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Hybrid processes for the treatment of commercial laundry wastewater using extracellular polymeric substances, electrocoagulation, and ultrafiltration

Procédés hybrides pour le traitement des eaux usées de buanderies commerciales en utilisant des substances polymériques extracellulaires, l'électrocoagulation et l'ultrafiltration.

Par

Sushil Kumar

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Examinateur externe:	Prof. Robert Hausler STEPPE-ETS, Montreal
Examinateur externe:	Prof. Elsayed Elbeshbishy Ryerson University, Toronto
Examinateur interne et président de jury:	Prof. Jean François Blais INRS-ÉTÉ
Directeur de recherche:	Prof. Patrick Drogui INRS-ÉTÉ

Dédicace

This thesis is dedicated to my family.

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RÉSUMÉ

La gestion et le traitement des eaux usées provenant de buanderies commerciales posent d'importants défis environnementaux et économiques. Cette thèse aborde cette problématique en proposant des solutions novatrices et respectueuses de l'environnement pour le traitement des eaux usées de buanderies commerciales (EUBC) au Québec, Canada. Trois approches distinctes sont explorées : i) l'utilisation de substances polymériques extracellulaires (SPE) comme bio-floculants, ii) l'application de l'électrocoagulation constituée de morceaux de métaux angulaires d'électrodes sacrificielles utilisées sous forme de lit fixe dans le réacteur électrolytique (procédé EC-ELF) et; iii) l'emploi combiné d'ultrafiltration (UF) et de SPE pour le traitement des des eaux usées de buanderies commerciales.

La première partie de l'étude consiste à produire les SPE à partir de glycérol brut et de boues de papeterie par une souche bactérienne (BS-04). Deux types de SPE, SPE visqueux (S-SPE) et SPE de bouillon (B-SPE), sont utilisés pour le traitement des EUBC. Une analyse comparative avec des coagulants chimiques conventionnels ($FeSO_4$, $CaCl_2$, Alun) montre des résultats prometteurs. Le S-SPE présente une activité de floculation (AF) plus élevée, éliminant 83,0 ± 1,0 % de la turbidité, 77 ± 2 % des solides en suspension (SS) et 76,0 ± 1,0 % de la demande chimique en oxygène (DCO) des EUBC. La combinaison de S-SPE et d'alun atteint des taux d'élimination remarquables de 98,09 ± 0,09 % de la turbidité, 95,42 ± 0,58% des SS et 83,08 ± 1,10 % de la DCO des EUBC.

La deuxième partie de l'étude a consisté à appliquer le procédé EC-ELF utilisant des électrodes en morceaux de métaux de fer angulaires installés dans le réacteur sous forme de lit fixe, suivi de l'ajout de polymère de synthèse ou de SPE. Un plan d'expérience constitué du plan factoriel (PF) suivi du plan central composite (PCC) a permis d'optimiser le procédé pour le traitement des EUBC. L'application des conditions optimales (intensité de courant de 3.0 A, un temps de traitement de 60 minutes et une dose de SPE améliorée de 5,8 mg/L à 8,0 mg/L) a permis une élimination efficace de la turbidité (98 ± 2 %), des solides totaux (TS) (95 ± 3 %), de la DCO (89 ± 5 %) et de nonylphénol ethoxylates (NPEO) (53 ± 2 %). Les paramètres opératoires optimisés reprèsentent un coût de traitement de 1,20 $CAN/m^3.

La dernière partie des travaux a consisté à introduire les SPE dans la boucle de recirculation de l'ultrafiltration de manière à atténuer l'encrassement de la membrane et à allonger ainsi sa durée de fonctionnement normal. Des études d'optimisation révèlent une augmentation du débit de perméat, de l'élimination de la turbidité et de l'élimination de la DCO. Dans des conditions optimisées (TMP : 18 psi, température : 60 ˚C, dose de SPE : 14,17 mg/L), l'UF couplée à des SPE atteint un débit de perméat de 0,81 L/min, une élimination de la turbidité de 99,0 ± 1,0 %, une élimination de la DCO de 80,0 ± 1,0 % et un temps de lavage de 31 minutes. Les estimations de coûts (0.93$ CAD/ m^3) indiquent des dépenses énergétiques (0.86 $/ m^3) comparables à celles du procédé EC-ELF, confirmant la faisabilité économique de cette approche.

Cette thèse offre des perspectives précieuses sur les approches novatrices et durables pour le traitement des EUBC et la promotion d'une gestion responsable des eaux usées dans les installations de blanchisserie commerciale.

Mots-clés: Eaux usées de blanchisserie commerciale (CLWW), Substances polymériques extracellulaires (SPE), L'électrocoagulation constituée d'électrodes sacrificielles utilisées sous forme de lit fixe dans le réacteur électrolytique (EC-ELF), Ultrafiltration (UF), Traitement durable des eaux usées.

ABSTRACT

The management and treatment of wastewater from commercial laundries pose significant environmental and economic challenges. This thesis addresses this issue by proposing novel, environmental-friendly solutions for treating commercial laundry wastewater (CLWW) in Quebec, Canada. Three distinct approaches are explored: (i) the utilization of extracellular polymeric substances (EPS) as bio-flocculants, (ii) packed bed electrocoagulation (PBEC) with scrap sacrificial iron electrodes, and (iii) combined use of ultrafiltration (UF) membrane and EPS for the treatment of CLWW.

The first part of the study consists of producing EPS from crude glycerol and paper mill sludge using a bacterial strain (BS-04). Two types of EPS, Slime EPS (S-EPS) and Broth EPS (B-EPS), are utilized for CLWW treatment.

A comparative analysis with conventional chemical coagulants ($FeSO_4$, $CaCl_2$, Alum) shows promising results. S-EPS demonstrates higher flocculation activity (FA), removing 83.0 ± 1.0 % of turbidity, 77.0 ± 1.5 % of suspended solids (SS), and 76.0 ± 1.0 % of chemical oxygen demand (COD) from CLWW. The combination of S-EPS and alum achieves remarkable removal rates of 98.09 ± 0.09 % turbidity, 95.42 ± 0.58 % SS, and 83.08 ± 1.1 % COD from CLWW.

The second part of the study consists of applying PBEC using scrap sacrificial iron electrodes followed by addition of synthetic polymer/EPS as flocculants. An experimental design consisting of factorial design (FD) followed by the central composite design made it possible to optimise the process for the treatment of CLWW. The application of optimal conditions (current intensity of 2.99A, treatment time of 58.8 minutes, and an enhanced EPS dose from 5.8 mg/L to 8.0 mg/L) allowed effective removal of turbidity (98.0 ± 2%), total solids (TS) (95.0 ± 3%), COD (89.0 ± 5%), and Nonylphenol Ethoxylates (NPEOs) (53 ± 2%). The optimized operating parameters result in a treatment cost of 1.20\$CAN/m^3.

The final part of the work involved introducing EPS into the recirculation loop of ultrafiltration to mitigate membrane fouling and thus extend its normal operating life. Optimization studies reveal an increase in permeate flow rate, turbidity removal, and COD removal. Under optimized conditions (TMP: 18 psi, temperature: 60°C, EPS dose: 14.17

mg/L), UF coupled with EPS achieves a permeate flow rate of 0.81 L/min, turbidity removal of 99.0 ± 1.5 %, COD removal of 80.0 ± 1.0 %, and a wash time of 31 minutes. Cost estimates (0.93$ CAN/m^3) indicate energy costs (0.86$CAN/$m^3$) comparable to those of the PBEC process, confirming the economic feasibility of this approach.

This thesis provides valuable insights into innovative and sustainable approaches for treating CLWW and promoting responsible wastewater management in commercial laundry facilities.

Keywords: Commercial laundry wastewater (CLWW), Extracellular polymeric substance (EPS), Packed bed electrocoagulation (PBEC), Ultrafiltration (UF), Sustainable wastewater treatment

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF EQUATIONS

LIST OF ACRONYMS AND ABBREVIATIONS

AO	Advanced oxidation
AOPs	Advanced oxidation processes
AOS	Alpha olefin sulfonate
B-EPS	Broth extracellular polymeric substances
BOD	Biochemical oxygen demand
CCD	Central composite design
CEAEQ	Center of expertise in environmental analysis of Quebec
CEPS	Capsular extracellular polymeric substances
CLWW	Commercial laundry wastewater
COD	Chemical oxygen demand
COV	Les composés organiques volatils
EC	Electrocoagulation
EC-ELF	L'électrocoagulation constituée d'électrodes sacrificielles utilisées sous forme de lit fixe dans le réacteur électrolytique
EGSB	Expanded granular sludge bed
EO	Electrochemical oxidation
EPS	Extracellular polymeric substances
FA	Flocculation activity
FOG	Fats, oils, and greases
HAP	Les hydrocarbures aromatiques polycycliques
HRT	Hydraulic retention time
IEP	Isoelectronic point
LAS	Linear alkylbenzene sulfonate
LCA	Life cycle assessment
LDH	Layered double hydroxide
LWW	Laundry wastewater
MBBR	Moving bed biological reactor
MBR	Membrane bio rector
MF	Microfiltration
MW	Molecular weight
NF	Nanofiltration
NPEO	Nonylphenol ethoxylate
NSERC	National sciences and engineering research council of Canada
PAC	Powdered activated carbon
PAHs	Polycyclic aromatic hydrocarbons
PBEC	Packed bed electrocoagulation
PES	Polyethersulfone
RBC	Les disques biologiques rotatifs/ Rotating biological contractor
RO	Reverse osmosis

RSM	Response surface methodology
SABP	Systèmes améliorés de biopolymères perméables
SBR	Sequential batch reactor
S-EPS	Slime extracellular polymeric substances
SPE	Substances polymériques extracellulaires
SS	Suspended solids
TMP	Transmembrane pressure
TN	Total nitrogen
TOC	Total organic carbon
TP	Total phosphorous
TS	Total solids
TSS	Total suspended solids
UF	Ultrafiltration
VOC	Volatile organic compounds

1. SYNTHÈSE

1.1 Introduction

À une époque marquée par la menace imminente de pénurie d'eau douce, la gestion responsable des ressources en eau s'impose comme une priorité mondiale. Cette thèse entreprend une exploration exhaustive visant à aborder un élément clé de ce défi : le traitement et la réutilisation des eaux usées issues des opérations de lavage de linge dans les buanderies commerciales (Zhou *et al.*, 2022). Cette recherche se concentre sur l'intégration de méthodologies avancées, telles que les Substances Polymériques Extracellulaires (SPE), l'électrocoagulation constituée d'électrodes sacrificielles utilisées sous forme de lit fixe dans le réacteur électrolytique (EC-ELF) avec des anodes sacrificielles, et les membranes d'ultrafiltration (UF) combinées avec les SPE. À travers une approche holistique, cette étude s'efforce non seulement de traiter efficacement les eaux usées de buanderies commerciales, mais aussi d'établir des voies pour leur réutilisation ultérieure, atténuant ainsi la pression sur les réservoirs limités d'eau douce.

Face aux demandes croissantes en ressources en eau engendrées par l'augmentation des populations et des activités industrielles, l'industrie textile, notamment les opérations de de buanderies commerciales, émerge comme un contributeur significatif à la consommation excessive d'eau et à la pollution de l'environnement. Le rejet des eaux usées de buanderies, chargées de détergents, de produits chimiques et d'autres contaminants, pose non seulement des risques environnementaux, mais aggrave également la pression sur les réserves d' eau douce (Kumar *et al.*, 2022b). Reconnaissant le besoin urgent de résoudre cette question, ces travaux de recherche visent à approfondir les connaissances en lien avec le traitement et la récupération des eaux usées provenant des opérations de buanderies. L'objectif global est de développer un modèle non seulement durable, mais également évolutif, atténuant l'impact environnemental tout en contribuant à la protection des ressources en eau.

La portée de cette recherche revêt une importance capitale dans le contexte de la pénurie d'eau mondiale. Elle entend offrir des approches innovantes pour le traitement et la réutilisation des eaux usées. En ciblant spécifiquement le traitement des eaux usées de buanderies, réputées pour leur consommation importante d'eau, cette étude entend apporter des contributions significatives au développement de stratégies applicables non seulement à cette industrie, mais également à d'autres secteurs industriels réputés pour leur consommation excessive d'eau (ex. industries agro-alimentaires, industries du textile, etc.),

L'objectif global de cette étude consiste à mettre au point un système hybride combinant l'électrolyse, l'ultrafiltration et l'ajout de substances polymériques extracellulaires (SPE) pour le traitement des eaux de buanderies commerciales. Ces travaux comportent trois objectifs spécifiques : **i**) Évaluer l'efficacité d'un procédé d'électrocoagulation constitué de morceaux de métaux angulaires (électrodes sacrificielles) utilisés sous forme de lit fixe dans le réacteur électrolytique (procédé EC-ELF) pour le traitement des eaux de buanderies commerciales; ii) Traiter les eaux de buanderies à l'aide de SPE seules et effectuer une comparaison avec le procédé EC-ELF (l'électrocoagulation constituée d'électrodes sacrificielles utilisées sous forme de lit fixe dans le réacteur électrolytique); iii) Clarifier les eaux de buanderies par ultrafiltration en présence de SPE de manière à réduire le colmatage membranaire.

1.2 Synthèse de la littérature

1.2.1. Eaux usées de buanderies

Les caractéristiques des eaux usées de buanderies (EUB) varient en fonction des habitudes de lavage, des types de machines et de facteurs régionaux. Différentes méthodes de traitement sont nécessaires pour diverses sources d'EUB, telles que les eaux usées de blanchisserie domestiques, commerciales et hospitalières. Les propriétés telles que la température, le pH, la turbidité, la conductivité et la composition chimique diffèrent selon les sources d'EUB. Des surfactants tels que LAS, NPEO et des micro-plastiques sont des contaminants émergents présents dans les EUB, posant des risques environnementaux et sanitaires (Oktiawan *et al.*, 2021). Les technologies de traitement visent à éliminer ces contaminants pour une réutilisation ou un rejet. Les normes internationales fixent des limites admissibles pour les EUB traitées, guidant leur réutilisation à des fins telles que l'irrigation, l'abreuvement du bétail, voire comme source d'eau potable après un traitement rigoureux (Corona *et al.*, 2021). De plus, l'impact des EUB sur la santé humaine et l'environnement, et ses effets sur la vie aquatique et les mutations génétiques potentielles, soulignent l'importance d'un traitement et d'une réglementation efficaces (Centurion *et al.*, 2018).

1.2.2. Traitement des eaux usées de blanchisserie

Dans l'ensemble, le système de traitement des EUB peut être divisé en trois types de traitement : : biologique, physico-chimique et combinaison de méthodes biologiques et physico-chimiques (Bering *et al.*, 2018).

1.2.3. Traitement physico-chimique

La coagulation chimique, méthode traditionnelle de traitement des eaux usées, utilise des coagulants inorganiques ou organiques comme le sulfate d'aluminium et les sels de fer. Ces coagulants produisent des sous-produits de métal hydroxyde dangereux, nécessitant une purification ultérieure par adsorption (Corona *et al.*, 2021). Des études récentes mettent en évidence l'efficacité des coagulants chimiques dans l'élimination des microfibres des eaux usées de buanderie (EUB). L'électrocoagulation (EC) utilise un courant électrique pour générer des coagulants in situ et a montré une solution prometteuse pour éliminer divers polluants des eaux usées, y compris les matières en suspension, DCO et les micro-plastiques (Samadikun *et al.*, 2021). Les défis tels que la passivation des électrodes et la consommation d'énergie doivent être abordés pour une application durable. La filtration membranaire offre une efficacité de séparation élevée pour le traitement des EUB, mais l'encrassement membranaire reste un défi qui nécessite des traitements ou des modifications supplémentaires. Les récents progrès explorent l'utilisation de membranes céramiques pour résoudre les problèmes de stabilité et de perméabilité (Shang *et al.*, 2015). L'adsorption est une technique rentable pour éliminer les polluants des EUB en utilisant divers adsorbants. Des études récentes démontrent l'efficacité de différents adsorbants dans l'élimination des surfactants, des métaux lourds et des radionucléides des EUB, mais l'optimisation des processus d'adsorption est encore nécessaire pour une efficacité maximale.

1.2.4. Traitement biologique

Les méthodes de traitement biologique des eaux usées, comprenant la décomposition aérobie et anaérobie, offrent des solutions efficaces pour éliminer les contaminants. Les processus aérobies, tels que les bioréacteurs aérobies et les filtres percolateurs, ont démontré des taux élevés d'élimination des polluants tels que la DBO_5, les surfactants et la DCO. La biodégradation anaérobie, réalisée à travers divers réacteurs comme les SBR et les AFBR, montre des possibilités de pouvoir éliminer des polluants tels que LAS et la matière organique, selon la composition des eaux usées (Ziemba *et al.*, 2018). Cependant, le traitement biologique peut entraîner la formation de mousse dans le bassin d'aération en raison des surfactants présents dans les eaux usées. Les agents anti mousses ou les ajustements des paramètres opérationnels tels que le temps de rétention des boues peuvent contribuer à contrôler la formation de mousse.

1.2.5. Procédés de traitement intégrés

Les processus intégrés de traitement des eaux usées combinent des méthodes physiques, chimiques et biologiques pour atteindre les normes réglementaires. La combinaison des procédés tels que l'électrocoagulation, l'adsorption, la filtration membranaire et la floculation améliore l'efficacité de l'élimination des polluants des eaux usées. Par exemple, des études ont montré que la combinaison de l'électrocoagulation bipolaire et de l'électro flottation peut réduire significativement la concentration de polluants tels que la DCO et les MBAS dans les eaux usées industrielles (GICHARU, 2012). De même, la combinaison de la coagulation avec l'adsorption ou la filtration membranaire a entraîné une élimination substantielle de contaminants tels que la DCO, les surfactants, la turbidité et l'azote total.

De plus, les approches intégrées impliquant un traitement biologique, telles que les bioréacteurs à membrane (MBR), offrent des avantages tels que des taux d'élimination élevés pour divers polluants et une facilité d'automatisation (Ude *et al.*, 2021).

1.2.6. Aspect économique

Lors de l'analyse économique, des facteurs tels que la consommation d'énergie, les dépenses en capital et les coûts d'exploitation et de maintenance affectent considérablement l'économie du traitement. La consommation d'énergie dans les stations de traitement varie en fonction de la technologie et des pratiques de lavage du linge. Les coûts d'exploitation et de maintenance incluent la main-d'œuvre et la consommation en électricité. La maintenance constitue une dépense non négligeable. Comprendre ces aspects économiques aide à sélectionner des méthodes rentables pour le traitement des eaux usées de blanchisserie (Ran *et al.*, 2022).

1.2.7. Réutilisation des eaux résiduaires de buanderies commerciales : rôle dans l'économie circulaire

La transition vers un modèle d'économie circulaire dans le traitement des eaux usées est motivée par la nécessité de récupérer les nutriments et l'eau recyclable des eaux usées, offrant des économies de coûts et des avantages environnementaux par rapport au modèle d'économie linéaire. Les EUB contiennent des ressources précieuses telles que l'eau traitée et les boues, qui peuvent être réutilisées à diverses fins, réduisant la dépendance aux ressources non renouvelables et à l'eau douce (Within, 2015). Cependant, les méthodes de traitement conventionnelles des EUB sont confrontées à des défis tels que des coûts élevés et des impacts environnementaux. Des méthodes alternatives telles que l'utilisation des bio-floculants et l'électrocoagulation sont explorées, ainsi que des stratégies pour améliorer l'efficacité de la filtration membranaire. Le développement d'adsorbants rentables et de stratégies de remédiation combinées est crucial pour un traitement durable des EUB (Makropoulos *et al.*, 2018). Des recherches intensives sont nécessaires pour développer des procédés de traitement des EUB économiquement viables et respectueux de l'environnement qui répondent aux normes de réutilisation de l'eau.

1.3 Problématique, hypothèses, objectifs et originalité

La nécessité de traiter durablement les eaux usées de buanderies est motivée par la rareté mondiale des ressources en eau douce. Pour permettre la réutilisation, l'eau traitée doit satisfaire à des critères spécifiques. Pour relever efficacement ce défi, il est nécessaire de procéder à l'élimination complète des constituants organiques, des particules colloïdales, des contaminants et des agents tensioactifs afin de se conformer aux réglementations sur la réutilisation de l'eau. Cette étude se concentre principalement sur la phase initiale de ce processus complexe.

L'investigation porte sur l'utilisation des SPE en association avec l'UF et l'EC. Cette approche intégrée vise à éliminer efficacement la matière organique, les particules colloïdales et diverses impuretés présentes dans les eaux usées. Notamment, la phase ultérieure ciblant l'élimination des agents tensioactifs fait partie d'une initiative de recherche indépendante menée par Mahdieh en 2024. Cette recherche ultérieure explore l'utilisation de techniques d'oxydation avancée (AO) et d'oxydation électrochimique (EO) spécifiquement pour l'élimination des agents tensioactifs.

En divisant le projet en phases distinctes, cette thèse de recherche établit un cadre complet pour relever le défi complexe du traitement des eaux usées de blanchisserie en vue d'une réutilisation durable. L'étude met l'accent sur l'utilisation des SPE, de l'UF et de l'EC pour traiter le contenu organique et les impuretés particulaires, tandis que la phase ultérieure vise à optimiser les méthodes d'élimination des agents tensioactifs par le biais de techniques d'oxydation avancée - une approche essentielle pour répondre aux normes de réutilisation de l'eau, comme le font Mahdieh et al., 2024.

1.4 Problématique

1.4.1. Problèmatique générale

Impact hydrologique

Le monde est confronté à une réalité frappante : la rareté des réserves d'eau douce est une préoccupation pressante. Seulement 2,5 % de l'eau de la Terre est de l'eau douce, et à peine 0,3 % est accessible dans les lacs, les rivières et les marais. Un impressionnant 4 milliards de personnes font déjà face à une grave pénurie d'eau pendant au moins un mois par an. D'ici 2025, près des deux tiers de la population mondiale pourraient vivre dans des conditions de stress hydrique. Étonnamment, 80 % de l'eau que nous utilisons se retrouve sous forme d'eaux usées, regorgeant d'un potentiel inexploité. Le recyclage et le traitement des eaux usées à des fins agricoles, industrielles et même potables peuvent soulager la pression sur les sources d'eau douce (Mauchauffee et al., 2012). Pour minimiser la demande en eau douce, il est nécessaire de réutiliser les eaux usées traitées dans les systèmes de blanchisserie.

Rejet des eaux usées de blanchisserie : Impacts sur les réseaux d'égouts et l'efficacité des stations de traitement

L'industrie de la blanchisserie génère une énorme quantité d'eaux usées, qui présentent des caractéristiques assez différentes de celles des eaux usées municipales normales. Les eaux usées générées par l'industrie de la blanchisserie sont très riches en SS, en DCO, en tensioactifs tels que le NPEO et en métaux lourds. Lorsque les eaux usées de blanchisserie sont rejetées dans les égouts municipaux sans traitement préalable, elles contribuent à augmenter la concentration de SS, de DCO, de métaux lourds et de tensioactifs dans les eaux usées municipales. Dans les eaux usées municipales, la contribution des détergents aux métaux lourds est de 31,9 % pour le Cd, de 0,24 % pour le Cu et de 0,30 % pour le Zn (Aonghusa & Gray, 2002). Cela a des implications importantes pour l'élimination des boues d'égout. La présence de NPEO dans le système de traitement des eaux usées peut affecter la croissance microbienne ou réduire l'efficacité du traitement (Weng et al., 2010).

Limitations de l'utilisation d'une méthode de traitement conventionnelle unique pour les initiatives de réutilisation de l'eau

La pénurie économique d'eau est sévère dans les pays en développement. Ainsi, l'utilisation de stratégies de traitement des eaux usées peu coûteuses peut aider à minimiser ce problème. Une quantité abondante d'eaux usées de blanchisserie est générée quotidiennement et diverses recherches sur le traitement des eaux usées ont été menées pour trouver des techniques appropriées, mais le traitement des eaux usées de blanchisserie est assez complexe car aucun processus unique n'est assez efficace et ne répond aux normes de réutilisation. En raison de la présence de particules en suspension et de tensioactifs (composés réfractaires : NPEOs), il est assez difficile de traiter les eaux usées de blanchisserie par le processus de traitement conventionnel. L'étude montre que le processus de traitement conventionnel comme le traitement primaire peut éliminer efficacement les particules en suspension mais qu'il est difficile d'éliminer les tensioactifs (seuls 13 % de NPEO peuvent être éliminés) (Xu et al., 2014). Dans le même temps, l'ultrafiltration peut éliminer efficacement les particules en suspension et les micro-organismes mais elle ne peut pas atteindre une efficacité élevée d'élimination de la DCO, des métaux et des détergents (Huisman et al., 1997).

Manque de recherche et de connaissances sur les méthodes de traitement intégrées pour le traitement des eaux usées de blanchisserie

L'état actuel de la recherche sur les méthodes de traitement intégrées pour les eaux usées de blanchisserie est insuffisant, ce qui indique une lacune critique dans notre compréhension. Malgré l'accent croissant mis sur la gestion durable des eaux usées, il existe une pénurie notable d'études axées sur des approches intégrées spécifiquement adaptées aux défis uniques posés par les eaux usées de blanchisserie. Cette lacune de connaissances limite notre capacité à optimiser les processus de traitement en termes d'efficacité et d'impact environnemental, notamment étant donné la nature diversifiée et dynamique des contaminants présents dans les eaux usées de blanchisserie. L'absence de recherche sur les méthodes de traitement intégrées entrave le développement de stratégies efficaces pour les usines de traitement des eaux usées de blanchisserie, laissant l'industrie sans directives fondées sur des preuves. Pour combler cette lacune, les futures recherches devraient donner la priorité à l'étude de la performance et de la faisabilité des méthodes de traitement intégrées pour les eaux usées de blanchisserie. Établir une base de connaissances solide grâce à une enquête scientifique rigoureuse est crucial pour faire progresser les pratiques durables en matière de traitement des eaux usées et garantir la préservation de la qualité de l'eau et de la santé environnementale.

1.4.2. Problèmatique spécifique

L'utilisation excessive de coagulant chimique et la génération de sous-produits toxiques

Dans le traitement des eaux usées de blanchisserie, une utilisation excessive de coagulants chimiques peut entraîner des coûts opérationnels accrus et un impact environnemental. Les effluents de blanchisserie contiennent divers contaminants organiques et inorganiques, nécessitant des processus de coagulation-floculation efficaces pour le traitement (Wang & Zhou, 2018). Cependant, le surdosage des coagulants peut non seulement être économiquement inefficace, mais peut également entraîner la présence de produits chimiques résiduels dans l'eau traitée, posant des préoccupations environnementales.

Remplacement fréquent des anodes et passivation des électrodes lors du processus EC

Pendant le processus d'EC, la dégradation des anodes et leur remplacement fréquent posent un défi. La présence de polluants dans les eaux usées de blanchisserie nécessite la génération in situ de coagulants, qui est produite par l'oxydation de l'anode, causant la dégradation de l'électrode. Cette dégradation entraîne le besoin de remplacer fréquemment les électrodes, ce qui conduit à des cycles de fonctionnement courts. Le principal défi du processus d'EC réside dans le contrôle de la passivation, c'est-à-dire la formation de la couche d'oxyde à la surface des électrodes.

Colmatage de membrane pendant le processus de UF

L'encrassement de la membrane en raison de la présence de particules colloïdales, d'huiles et de tensioactifs dans les effluents de blanchisserie peut entraîner une efficacité de filtration réduite et une augmentation de la consommation d'énergie. Cela nécessite également un lavage fréquent de la membrane.

1.5 Hypothèses

Approche écologique : Les SPE bactériens pour la coagulation et la floculation durable

Le processus de coagulation et de floculation a été considéré comme une méthode facile, rentable et efficace pour éliminer les matières particulaires, l'argile, les colloïdes, les acides humiques, fulviques, le phosphore et les métaux lourds des eaux usées. Malgré les avantages économiques, l'utilisation de coagulants chimiques pose de graves problèmes environnementaux et de santé ; par conséquent, il existe un intérêt croissant pour l'utilisation de biopolymères écologiques et durables afin de réduire les risques posés par les coagulants chimiques et de diminuer leurs doses (Guo et al., 2017). Parmi les biopolymères, les substances polymères extracellulaires (SPE) bactériennes ont été explorées en tant qu'agents de floculation pour la décantation des boues et la déshydratation, et pourraient être utilisées pour le traitement des eaux usées de blanchisserie et l'élimination des métaux lourds et des tensioactifs. L'ajout de SPE dans les eaux usées de blanchisserie peut être utile pour réduire l'encrassement des membranes lors de la filtration en bout de ligne (Lin et al., 2019). Cela aidera à former des flocs et à empêcher la pénétration des particules à l'intérieur des pores de la membrane. Les SPE à base de polymères pourraient favoriser l'hydrophilicité de la membrane (Nouha et al., 2016).

Performance améliorée des électrodes et durée de vie opérationnelle prolongée grâce à l'emballage d'électrodes en fer sacrificiel

Le potentiel du processus d'électrocoagulation pourrait être utilisé pour l'élimination des composés organiques ; en même temps, la neutralisation de la charge peut aider à éliminer les particules colloïdales et en suspension des eaux usées (Li et al., 2022a). Dans le processus d'EC, le mécanisme de déstabilisation des contaminants, de suspension des particules et de rupture des émulsions peut être résumé en trois étapes successives. (1)

Compression de la double couche diffuse autour des espèces chargées par les interactions des ions générés par l'oxydation de l'anode sacrificielle (Oktiawan et al., 2021). (2) Neutralisation de la charge des espèces ioniques présentes dans les eaux usées par des contre-ions produits par la dissolution électrochimique de l'anode sacrificielle. Ces contre-ions réduisent la répulsion électrostatique entre les particules au point que l'attraction de van der Waals prédomine, provoquant ainsi la coagulation. Un résultat de charge nette nulle est obtenu dans le processus (Verma, 2017). (3) Formation de flocs : les flocs formés à la suite de la coagulation créent une couverture de boues qui emprisonne et relie les particules colloïdales qui restent encore dans le milieu aqueux (Nidheesh et al., 2020).

En augmentant la surface disponible grâce à l'empilement d'électrodes sacrificielles en fer, la passivation peut être considérablement réduite, ce qui entraîne des intervalles plus longs entre les cycles de régénération des électrodes. Cette méthodologie permet de prolonger le fonctionnement de l'installation initiale des électrodes, facilitant ainsi le traitement de grands volumes d'eau dans des délais plus courts.

Utilisation des EPS pour améliorer les performances de l'ultrafiltration

La littérature publiée (Kabsch-Korbutowicz et al., 2006 ; Shon et al., 2004) suggère que l'ultrafiltration a un énorme potentiel pour éliminer les particules en suspension et offre une barrière pratiquement complète contre les particules plus grandes que la taille des pores, les bactéries et les virus beaucoup plus petits habituellement présents dans l'eau d'alimentation. Toutes les particules plus grandes que la taille des pores sont rejetées. Cette caractéristique rend les membranes d'UF idéales pour répondre aux exigences de qualité de filtration absolue largement indépendantes de la qualité de l'eau d'alimentation, à condition que l'intégrité initiale de la membrane reste intacte (Benis et al., 2021). L'UF pourrait permettre d'éliminer les composés colloïdaux/particulaires qui pourraient interférer avec le processus ultérieur, tel que l'adsorption ou la dégradation catalytique utilisée pour l'élimination du NPEO (Kornaros & Lyberatos, 2006). De même, le prétraitement des Eaux Usées de Blanchisserie (LWW) pourrait permettre de concentrer les polluants colloïdaux dans un petit volume, suivi du traitement de ces polluants à l'aide de la coagulation électrochimique. Une telle approche peut permettre d'augmenter le rendement faradique pendant le traitement électrochimique (Mollah et al., 2001).

Le flux tangentiel (circulation du fluide parallèlement à la surface de la membrane) peut être utilisé pour contrôler l'encrassement de la membrane (Watabe et al., 2016). L'incorporation de SPE dans les eaux usées de blanchisserie atténue l'encrassement de la membrane lors de la filtration en bout de ligne, facilite la formation de flocs et empêche la pénétration des particules dans les pores de la membrane. Les substances polymères extracellulaires à base de polymères renforcent en outre l'hydrophilicité de la membrane (Mauchauffee et al., 2012).

1.6 Objectifs

Objectif Global: Développement d'un processus hybride pour la décontamination et la réutilisation des eaux usées des blanchisseries commerciales en utilisant la technologie de traitement par membrane, électrolytique et SPE.

Objectif Spécifique

• Application de la méthode de traitement SPE pour les eaux usées brutes de blanchisserie et le flux concentré en utilisant différentes combinaisons de coagulants chimiques.

• Enquêter systématiquement sur efficacité du traitement des eaux usées de blanchisserie par l’électrocoagulation constituée d’électrodes sacrificielles utilisées sous forme de lit fixe dans le réacteur électrolytique (EC-ELF) et SPE, dans le but d'évaluer l'impact synergique de ces méthodes sur l'élimination des polluants, la stabilisation colloïdale et l'amélioration globale de la qualité de l'eau.

• Développer et optimiser la technologie de membrane UF pour le traitement des eaux usées de blanchisserie à l'échelle pilote, en mettant particulièrement l'accent sur la régulation des problèmes d'encrassement de la membrane. De plus, évaluer la faisabilité du traitement du flux concentré par EC-ELF.

1.7 Originalité

L'originalité de ce travail peut se résumer comme suit :

- Pionnier dans le traitement des eaux usées de blanchisserie par l'application de SPE dérivé de la fermentation du pétrole brut. Cette approche novatrice exploite le potentiel des SPE, un sous-produit de la fermentation microbienne, pour un traitement efficace des eaux usées, offrant ainsi une solution respectueuse de l'environnement et durable.
- Introduction d'un système EC-ELF pour le traitement des eaux usées de blanchisserie, mettant en vedette des électrodes sacrificielles en fer granulaire. Cette approche EC innovante vise l'élimination globale des contaminants, y compris les solides en suspension, les solides colloïdaux, la demande chimique en oxygène (DCO), les métaux et les tensioactifs. L'utilisation simultanée de SPE renforce davantage l'efficacité de ce processus de traitement.
- Avancement dans le traitement des eaux usées de blanchisserie grâce à l'application de la technologie de membrane UF. Cela implique non seulement la gestion efficace de l'encrassement de la membrane, mais aussi l'utilisation stratégique de SPE. Le contrôle de l'encrassement est réalisé grâce à la modulation des paramètres de fonctionnement et à l'optimisation du temps de lavage à contre-courant, améliorant ainsi les performances globales du système de membrane UF.

1.8 Méthodologie

Traitement des EUB à l'aide de SPE

Les eaux usées de blanchisserie brutes (EUB) ont été collectées auprès d'une laverie commerciale et soumises à une caractérisation rigoureuse, mesurant divers paramètres tels que le pH, la turbidité, la conductivité, les matières en suspension (MES), les matières totales en suspension (MTS), la demande chimique en oxygène (DCO), l'azote total (NT), le phosphore total (PT), les nonylphénols éthoxylés (NPEOs) et les métaux.

Des substances polymériques extracellulaires (SPE) ont été produites en utilisant la souche bactérienne BS-04 cultivée sur du glycérol brut et des boues secondaires. Le processus impliquait une fermentation, conduisant à la production de substances SPE en bouillon (B-SPE) et en boue (S-SPE). La concentration de SPE a été calculée à l'aide d'équations spécifiques basées sur le poids sec des substances produites.

Les S-SPE et B-SPE collectées ont été utilisées pour traiter les EUB. Le mécanisme de traitement des EUB à l'aide de SPE en tant que bio-floculant a été étudié, en se concentrant sur les processus de neutralisation de charge, de pontage et de correction, qui ont facilité l'agrégation des particules pour leur élimination.

Des expériences ont été menées pour optimiser le pH pour divers coagulants chimiques tels que le sulfate ferreux, l'alun et le chlorure de calcium. L'activité de floculation a été évaluée à différents niveaux de pH pour déterminer les conditions les plus efficaces pour le traitement. De plus, l'impact de la concentration en coagulant sur l'activité de floculation a été étudié pour établir les doses idéales pour un traitement efficace.

Des expériences supplémentaires ont exploré l'influence de la concentration en SPE (S-SPE et B-SPE) sur l'efficacité du traitement. Différentes concentrations ont été testées pour identifier la dose optimale pour une élimination maximale des polluants, en tenant compte de paramètres tels que la turbidité, les MES, les MTS, la DCO, l'azote, le phosphore, les métaux et les NPEOs.

L'étude a également examiné l'utilisation combinée de SPE et de coagulants chimiques. La synergie entre ces agents a amélioré l'activité de floculation grâce à des mécanismes de neutralisation de charge, de coagulation en balayage et de pontage des flocs. L'association SPE-coagulant a facilité une meilleure agrégation des particules, conduisant à un meilleur traitement des EUB.

Pour déterminer l'efficacité du traitement, l'effet du temps de traitement sur le processus a été évalué. Des expériences ont été menées sur différentes périodes pour identifier la durée optimale nécessaire pour atteindre une efficacité de traitement maximale.

Des méthodes analytiques certifiées fournies par le Centre d'expertise en analyse environnementale du Québec ont été utilisées pour évaluer les différents paramètres dans les eaux usées traitées, assurant une standardisation et une fiabilité dans le processus d'analyse.

EC-ELF utilisant des anodes sacrificielles

Le réacteur d'électrocoagulation constituée d'électrodes sacrificielles utilisées sous forme de lit fixe dans le réacteur électrolytique (EC-ELF) a été conçu comme un système complet visant à éliminer efficacement les contaminants de l'eau et des eaux usées. Il comprenait un réacteur en plexiglas de 4,5 L divisé en cinq compartiments séparés par des feuilles de plexiglas, permettant un écoulement en serpentin. Cette disposition facilitait un temps de contact plus long entre les coagulants chimiques générés in situ et les polluants dans les eaux usées. Chaque compartiment abritait des cathodes en acier inoxydable et des anodes en graphite, créant un champ électrique lorsque l'eau contaminée traversait le lit emballé, déclenchant des réactions électrochimiques pour produire des cations métalliques agissant comme des coagulants.

La disposition d'une garniture sacrificielle en fer entre la cathode et l'anode favorisait la coagulation et la floculation, améliorant le contact entre les électrodes et l'eau pour une déstabilisation et une agrégation efficace des particules. Cette configuration prévenait les courts-circuits en plaçant des déchets de fer sacrificiels séparés des cathodes en acier inoxydable par des feuilles de plexiglas perforées. Les électrodes étaient connectées en parallèle à un générateur CC, établissant un mode dynamique en lot où l'eau traitée était recirculée dans le réacteur. La recherche portait sur les EUB échantillonnées dans une installation à Montréal, Canada. Une caractérisation détaillée des eaux usées comprenait l'analyse du pH, de la turbidité, de la conductivité, de la DCO, des SST, des TS, des NPEO, des nitrates, des phosphates et des métaux. Cette analyse visait à comprendre la composition des eaux usées, essentielle pour la conception de méthodes de traitement efficaces. La comparaison avec les normes de rejet et de réutilisation a fourni des informations précieuses sur la qualité des eaux usées et leur déviation par rapport aux limites réglementaires.

La méthodologie de surface de réponse (MSR) a guidé la planification et l'optimisation statistique des expériences. Trois facteurs opérationnels clés - intensité du courant, temps de traitement et dose de polymère - ont été identifiés, et leur impact sur l'élimination de la turbidité, de la DCO et des SST a été évalué. Une matrice de conception factorielle composée de 8 expériences et une matrice de conception composite centrale (CCC) avec 12 essais supplémentaires ont été exécutées pour déterminer les paramètres optimaux.

Le remplacement des Substances Polymères Extracellulaires (SPE) par un polymère synthétique visait à améliorer l'efficacité du traitement EC-ELF et à rendre le processus plus durable et respectueux de l'environnement. L'étude a remplacé les polymères par des SPE dans des expériences optimisées, améliorant potentiellement l'élimination des contaminants des eaux usées.

Des méthodes analytiques prescrites par le Centre d'Expertise en Analyse Environnementale du Québec ont été utilisées pour les mesures de pH, de conductivité, de DCO, de SST, de TS, de NPEO et de métaux, assurant une standardisation et une fiabilité du processus d'analyse.

Les aspects économiques ont été pris en compte, en mettant principalement l'accent sur la consommation d'énergie et les coûts des polymères et des électrodes associés au processus d'électrocoagulation. La consommation d'énergie a été estimée au taux provincial de 0,06 \$CAN/KWh, tandis que le coût global par mètre cube de solution traitée a été évalué en dollars canadiens ($\$CAN/m^3$).

La recherche englobait la conception complète du réacteur, la caractérisation des eaux usées, l'optimisation statistique, l'incorporation des SPE, le respect des méthodes analytiques certifiées et les considérations économiques, visant à élaborer une stratégie de traitement efficace et économiquement viable pour les eaux usées de blanchisserie commerciale.

UF combiné avec SPE

Le traitement des eaux usées de blanchisserie à l'aide de membranes d'ultrafiltration (UF) en polyéthersulfone (PES) impliquait une méthodologie systématique. La membrane UF présentait des caractéristiques spécifiques : une surface de 0,5 m^2, une plage de coupure moléculaire de 100 à 150 kDa et une taille de pore de 0,02 microns. La conception de l'expérience suivait une approche factorielle et de conception composite centrale (CCC), en se concentrant sur trois variables codées :

• Pression transmembranaire (TMP) (X_1) : Variée entre 10 et 18 psi, codée comme U_1 avec un point médian de 14 psi et un changement (ΔU_1) de 4.

• Température (X_2) : Allant de 20 à 60°C, codée comme U_2 avec un point médian de 40°C et un changement de 20.

• Dose de Substances Polymères Extracellulaires (SPE) (X_3) : Allant de 5 à 15 mg/L, codée comme U_3 avec un point médian de 10 mg/L et un changement de 5.

Ces variables codées permettaient d'explorer systématiquement l'impact de la pression transmembranaire, de la température et de la concentration en SPE sur le processus d'UF pour le traitement des eaux usées de blanchisserie. Les conceptions factorielles et CCC fournissaient un cadre complet pour évaluer l'interaction de ces variables sur l'efficacité du système de membrane UF dans l'élimination des contaminants des eaux usées.

La configuration expérimentale visait donc à optimiser les paramètres du processus d'UF en évaluant l'influence de la TMP variable, de la température et de la dose de SPE sur la qualité du perméat et les débits de flux à travers la membrane UF. Cette approche systématique visait à déterminer les conditions les plus favorables pour un traitement efficace des eaux usées de blanchisserie en utilisant la configuration spécifique de la membrane UF.

1.9 Résultats

Traitement des EUB à l'aide de SPE

Les EUB ont été analysées pour divers paramètres, notamment la turbidité, le pH, la conductivité, les solides en suspension (SS), les solides totaux (TS), la demande chimique en oxygène (DCO), les nonylphénols éthoxylates (NPEO) et les métaux. Les résultats ont indiqué des niveaux élevés de turbidité, de SS, de TS, de DCO et de NPEO. La présence de ces composés dans les EUB est attribuée aux résidus/saletés fixés sur les vêtements et aux détergents utilisés pour le lavage du linge. Les EUB contenaient également du calcium, de magnésium, d'aluminium, de sodium et de potassium, ainsi que des traces de métaux toxiques comme le zinc, le mercure, le plomb, le nickel et le cuivre.

La comparaison avec les normes de rejet d'eau établies par Santé Canada et la Ville de Québec a montré que les concentrations d'EUB étaient près de cinq fois supérieures aux valeurs recommandées, soulignant la nécessité d'un traitement avant le rejet dans les plans d'eau.

Le processus de traitement impliquait des tests de coagulants chimiques (alun, $FeSO_4$, $CaCl_2$) pour déterminer leur efficacité dans le traitement des EUB. Les niveaux de pH optimaux pour chaque coagulant ont été identifiés : pH 7 pour l'alun, pH 11 pour le $FeSO_4$ et pH 5 pour le $CaCl_2$, mettant ainsi en évidence le rôle t du pH dans le processus de coagulation-floculation. Les coagulants chimiques déstabilisent les particules, provoquant la formation de flocs, ce qui entraîne l'élimination des polluants.

Les concentrations variables de coagulants ont influencé l'élimination de la turbidité, des SS, des TS et de la DCO. L'alun et le $FeSO_4$ ont permis d'enregistrer des taux d'enlèvement de contaminants plus élevés, comparativement à ceux enregistrés avec le $CaCl_2$. La combinaison d'alun et de SPE (S-SPE) a considérablement amélioré l'élimination des polluants, atteignant des taux d'abattement allant jusqu'à 98,09 ± 0.09 % d'enlèvement de la turbidité et 83,08 ± 1,1 % d'enlèvement de la DCO. L'activité de floculation est améliorée lors de la combinaison de coagulant métallique et de bio-floculant grâce à trois mécanismes principaux se déroulant simultanément : i) la neutralisation de charge ; ii) la coagulation par balayage ; et iii) le pontage des flocs (Bo *et al.*, 2012). La nature ionique du métal, ainsi que sa taille et sa densité de charge, régulent son interaction avec les SPE chargées négativement.

Les expériences avec le SPE (S-SPE et B-SPE) seul ont montré une efficacité d'élimination plus faible par rapport aux coagulants chimiques. Cependant, lorsqu'ils sont utilisés en combinaison avec l'alun, les SPE ont fait preuve d'une bonne efficacité, notamment le S-SPE atteignant 98 ± 1 % de turbidité et 83 ± 1 % de DCO à des conditions optimisées.

La combinaison SPE et $Al_2(SO_4)_3$ a permis d'enregistrer des taux d'abattement en métaux relativement élevés. Des taux d'abattement allant jusqu'à 87 % d'enlèvement d'Al et 96% d'enlèvement de Ca ont été enregistrés lors du traitement des EUB.

Dans l'ensemble, la combinaison d'alun avec le SPE, en particulier le S-SPE, a montré des résultats prometteurs dans le traitement des EUB, démontrant un potentiel significatif pour une élimination efficace de la DCO, de la turbidité et des métaux, soulignant sa viabilité pour le traitement des eaux usées.

Traitement des EUB par EC-ELF utilisant des électrodes sacrificielles en lit fixe

Le procédé EC-ELF a été testé pour le traitement des eaux réelles de buanderies commerciales (EUB). Lors de l'application du procédé EC-ELF utilisant des anodes sacrificielles en lit fixe, plusieurs paramètres tels que, l'intensité de courant, le temps de traitement et la concentration de polymère de synthèse ont été étudiés. En appliquant un plan factoriel 2^3, les meilleures efficacités d'enlèvement des polluants (93±2% d'élimination SST; 87± 2% d'élimination de DCO) ont été obtenues pour une intensité de courant de 3 A pendant une période de 60 min de temps de traitement et lorsqu'une concentration 6 mg/L de polymère est utilisée. L'intensité de courant et le temps de traitement étaient les paramètres principaux les plus importants affectant l'enlèvement de la DCO et SST. Les contributions de l'intensité de courant et du temps de traitement étaient de 64% et 25%, respectivement, alors que la contribution concentration de polymère était de 10%.

L'analyse a révélé des informations sur l'effet d'interaction de X_1X_2 entre l'intensité du courant (X_1) et le temps de traitement (X_2) sur l'efficacité de l'élimination des polluants. Alors que l'interaction X_1X_2 avait un impact négligeable (moins de 1 %) sur la turbidité

(Y_1) et l'élimination de la DCO (Y_3), elle a influencé de manière significative l'élimination des SST (Y_2), contribuant à 9 %.

Un plan de surface de réponse (Plan Central Composite, PCC) a par la suite été appliqué pour déterminer les conditions optimales d'enlèvement simultané de la DCO, SST et la turbidité. Les critères choisis pour l'optimisation sont les suivants : (i) Le temps de traitement et l'intensité du courant doivent être maintenus dans une plage spécifiée avec un niveau d'importance élevé (facteur de pondération de 3/5) pour faciliter l'élimination des polluants ; (ii) La concentration en polymère devrait être minimisée avec un niveau d'importance élevé (facteur de pondération de 4/5) afin de réduire l'utilisation de polymère synthétique et de promouvoir la durabilité dans le processus. En fonction de ces critères, la solution optimale proposée par le logiciel consiste à imposer une intensité de courant de 3.0 A, pendant 59 min du temps de traitement et en appliquant une concentration en polymère de 5.8 mg/L pour la séparation solide/liquide des EUB électro-coagulées. Des taux d'abattement allant jusqu'à 89 ± 5 % de la DCO, 95± 3 % de SST et 98 ± 2 % de la turbidité ont été enregistrés avec un coût d'opération de CAD \$0.93/ m^3 (incluant, énergie consommée, la consommation d'électrode et de polymère) dans ces conditions optimales.
- Dans ces conditions optimales, des composés tels que les phosphates (84% d'élimination), l'azote ammoniacal (77% d'élimination) et le sulfate (81%) peuvent être simultanément éliminés. Les modifications chimiques de surface améliorent les SPE chargées négativement en utilisant des cations multivalents positivement chargés comme ponts pour attirer les anions. Les cations positivement chargés et les groupes anioniques des SPE facilitent l'élimination des nitrates–nitrites par échange de cations et forces coulombiennes. L'application du procédé EC-ELF suivi de la floculation à l'aide d'un polymère de synthèse n'a pas eu d'effet sur le NPEO.

Lorsque le polymère de synthèse est substitué par les SPE (ajout de 8 mg/L), on note une amélioration significative des taux d'abattement des polluants cibles (incluant le NPEO). Des taux d'abattement de la turbidité (97 ± 1 %), des SST (95± 1 %), de la DCO (89 ± 2%) et des NPEO (53 ± 5 %) ont été enregistrés, montrant ainsi l'influence cruciale de la dose de SPE sur les performances épuratoires. Les groupes fonctionnels présents dans les SPE facilitent l'adsorption et la complexation des NPEO, tandis que les microorganismes associés aux SPE contribuent à la bioaccumulation. De plus, les SPE peuvent physiquement piéger les NPEO, agissant comme une barrière de filtration lors du traitement des eaux usées.

Ces résultats soulignent l'importance des paramètres opérationnels optimisés et de l'intégration de matériaux appropriés, tels que les biopolymères, dans l'amélioration de l'efficacité des systèmes de traitement électrochimique pour une élimination efficace des polluants dans les eaux usées de blanchisserie.

Traitement des EUB par l'emploi combiné d'UF et de SPE

Cette partie des travaux a consisté à évaluer l'efficacité d'un procédé hybride combinant UF et SPE pour le traitement des EUB. Les eaux initiales de buanderie avaient une concentration de DCO de 577 ± 30 mg/L et une concentration de NPEO de 568 ± 150 µg/L. Elles étaient également caractérisées par une turbidité initiale relativement élevée de 109 ± 5 NTU et un pH basique (pH 10,0 ± 0,5). Un plan de surface de réponse a été utilisé pour modéliser le procédé hybride pour l'enlèvement simultané de la DCO, de NPEO et de la turbidité. Les paramètres étudiés sont la pression transmembranaire (TMP) (10 et 18 psi), la température (20 à 60°C) et la concentration de SPE (5 à 15 mg/L).

La première étape a consisté à construire un plan factoriel afin de caractériser les effets associés aux différents paramètres principaux contrôlant l'efficacité du procédé UF/SPE pour l'enlèvement des contaminants. Ce plan a également permis d'étudier les effets des interactions des paramètres sur la réponse. Un ensemble de 2^3 expériences a été réalisé. Les valeurs de réponses enregistrées pour ce plan factoriel varient entre 62 et 78% pour l'enlèvement de la DCO, alors qu'elle varie entre 97 et 99% pour l'élimination de la turbidité. La meilleure réponse ayant été obtenue en imposant une pression trans membranaire de 18 psi, une température de 60°C et une concentration en SPE de 14.2 mg/L correspondant à un rendement d'enlèvement de la DCO de 80 ± 1% et un taux d'enlèvement de la turbidité de 99 ± 1%.

Le EC-ELF a démontré une efficacité remarquable dans l'élimination des contaminants des concentrés de UF, surpassant les méthodes EC conventionnelles et offrant des solutions prometteuses pour des pratiques durables de gestion de l'eau. L'estimation des coûts (0,86$ CAD/ m^3) a montré une efficacité énergétique comparable entre le EC-ELF et la UF dans le traitement des EUB, soulignant le potentiel des techniques innovantes comme le EC-ELF dans la résolution des défis de qualité de l'eau.

1.10 Recommandations et perspectives

Conclusion

Cette étude a permis de développer et d’évaluer les performances épuratoires d’un procédé hybride combinant électrolyse, ultrafiltration et ajout de substances polymériques extracellulaires (SPE) pour le traitement des eaux de buanderies commerciales.

De façon plus spécifique, l’étude menée sur le traitement des EBC en utilisant les SPE seuls ou en combinaison agents coagulants a fait ressortir les points suivants :

- Les SPE produits par la souche bactérienne BS-04 ont présenté une teneur notable en protéines de 1,45 g/L, ainsi que des propriétés spécifiques telles que la viscosité (31 mPas), le potentiel zêta (-38 mV), l'activité de floculation (82 %) et l'indice de déshydratation (40 %). Ces caractéristiques rendent les SPE efficaces pour le traitement des EUB, comme en témoigne leur capacité à améliorer les processus de floculation et de déshydratation. L'absence de teneur significative en glucides dans les SPE suggère un avantage potentiel en termes de stabilité et de longévité dans les applications de traitement (Hu et al., 2023).
- Les expériences avec le SPE (S-SPE et B-SPE) seul ont montré une efficacité d'élimination plus faible par rapport aux coagulants chimiques. Des taux d’enlèvement de 76,0 ± 1,0 % de DCO, de 83,0 ± 1,0 % d’enlèvement de la turbidité et de 77,0 ± 1,5 % d’enlèvement des solides totaux lorsque les SPE sont utilisées seuls.
- En revanche, lorsque les SPE sont utilisées en combinaison avec l'alun, des taux d’enlèvement des polluants ont été majorés. Des taux d’abattement allant jusqu'à 98,09 ± 0,09 % d’enlèvement de la turbidité et 83,08 ± 1,1 % d’enlèvement de la DCO sont enregistrés.

L’étude menée sur le traitement des EUB par EC-ELF utilisant des électrodes sacrificielles en lit fixe (en présence ou non de SPE) a fait ressortir les points suivants :

- Le processus EC-ELF optimisé statistiquement démontre une élimination efficace dans des conditions spécifiques, avec une turbidité éliminée à 98 ± 2,0 %, des TS à 95 ± 3,0 %, une DCO à 89 ± 5,0 %, et des NPEO à 53 ± 2,0 %.
- L'estimation économique révèle la consommation d'énergie comme principale dépense, totalisant 1,20 $CAN/m^3, avec les coûts énergétiques contribuant à eux seuls à 0,86 $CAN/m^3, tandis que les dépenses en électrodes et en polymères étaient comparativement minimes.
- Les expériences de conception factorielle soulignent l'influence cruciale de la température et de la concentration de SPE, avec la température comme facteur dominant dans l'élimination de la turbidité (contribuant à environ 89,7 % de la variation observée) et la concentration de SPE contribuant significativement à l'élimination de la DCO.

L'étude menée sur le traitement des EUB par l'emploi combiné d'UF et de SPE a fait ressortir les points saillants suivants :

- L'optimisation des conditions de traitement avec la méthodologie de conception composite centrale a produit des valeurs prédites alignées sur les résultats expérimentaux, démontrant la robustesse des modèles développés.
- Pour le traitement optimisé des EUB commerciales avec UF et SPE, les conditions comprenaient une TMP de 18 psi, une température de 60 °C et une dose de SPE de 14,2 mg/L, permettant un débit de perméat de 0,81 L/min, une élimination de la turbidité de 99,0 ± 1,0 %, une élimination de la DCO de 80,0 ± 1,0 % et un temps de lavage de 31 minutes.
- L'incorporation de SPE dans la UF a réduit l'encrassement des membranes, améliorant l'efficacité du processus et prolongeant la durée de vie des membranes.
- Le EC-ELF a produit des réductions notables de la turbidité, des SST, des ST, de la DCO et des NPEO, démontrant son efficacité dans l'élimination complète des contaminants.
- Les estimations des coûts pour le traitement des EUB avec UF et SPE se sont élevées à 0,865$CAD/m^3, avec une dépense énergétique de 0,01$CAN/m^3 et de 0,67$CAN/m^3 pour les SPE. En revanche, pour le EC-ELF, le coût a été évalué à

0,17$CAN par mètre cube, incluant 0,01$CAN par mètre cube pour l'énergie et 0,005$CAN par mètre cube pour le polymère, avec une efficacité énergétique comparable à la UF.

Cette étude fournit des informations sur les conditions de traitement optimisées et la rentabilité de différentes méthodes pour le traitement des EUB, mettant en avant le potentiel des SPE dans la UF et des techniques innovantes comme le EC-ELF pour traiter le concentré de UF afin de relever les défis environnementaux et promouvoir des pratiques durables de gestion de l'eau.

Perspectives

En capitalisant sur les résultats riches et les perspectives éclairantes de cette étude exhaustive, plusieurs perspectives prometteuses et des voies d'exploration scientifique supplémentaires se dessinent dans le domaine du traitement des eaux usées de buanderie (LWW).

En revanche, il serait très important d’effectuer des travaux supplémentaires dans le but d’optimiser l’efficacité de ce procédé hybride :

- Réaliser une étude dynamique du procédé EC-ELF permettant de comprendre l’écoulement du fluide à l’intérieur du réacteur électrolytique (étude dynamique des fluides computationnelle).
- Optimiser et étudier davantage le mécanisme d’enlèvement des NPEO par les SPE. Les futures études peuvent se concentrer sur l'amélioration de la capacité d'adsorption des SPE grâce à des techniques de modification de surface et sur l'exploration de consortiums microbiens capables de dégrader les NPEOs avec le soutien des SPE.
- Prévoir dans l’étude subséquente la combinaison des procédés d’oxydation électro-catalytique (OEC) et les SPE. La génération in situ d’espèces oxydantes (OH, H_2O_2, $H_2S_2O_8$, etc.) et l’utilisation d’électrodes catalytiques offrant la plus grande surface effective possible (électrodes sous forme de métal déployé) constituent les deux éléments clés pour oxyder efficacement les NPEO. En revanche, lorsque le procédé d’oxydation électrochimique est limité par le processus de transfert de masse des espèces électro-actives (ex. NPEO présent en élément de trace dans l’eau), il peut

être intéressant de concentrer au préalable les polluants par une séparation solide/liquide en utilisant par exemple les SPE (comme bio-floculant).

- Tester et mettre à l'échelle préindustrielle le procédé hybride: Mener des recherches supplémentaires pour optimiser le procédé combinant SPE, électrocoagulation et ultrafiltration pour une application à grande échelle.
- Évaluer le cycle de vie (LCA) du procédé hybride : réaliser une évaluation complète du cycle de vie de l'ensemble du procédé de traitement des eaux usées, incluant l'utilisation de SPE, l'électrocoagulation, l'ultrafiltration et toutes les étapes de traitement ultérieures. Évaluer les impacts environnementaux associés à chaque étape du processus, de l'extraction des ressources à la génération de déchets, pour identifier les domaines potentiels d'amélioration et orienter les efforts futurs en matière de durabilité.
- Analyser les coûts et la viabilité économique du procédé hybride : Effectuer une analyse détaillée des coûts englobant les dépenses liées à la mise en œuvre et à l'exploitation de l'approche de traitement multimodale. Évaluer les coûts initiaux, les dépenses opérationnelles, la maintenance, ainsi que les économies ou avantages potentiels découlant de l'eau traitée ou des ressources récupérées. Cette analyse aidera à déterminer la faisabilité économique et la compétitivité de l'approche.

2. LITERATURE REVIEW

2.1. Article Information

Article's title	Advancements in Laundry Wastewater Treatment for Reuse: A Review
Names of the authors and their professional affiliations	Sushil Kumar[1], Ali Khosravanipour Mostafazadeh[1,2], Lalit R Kumar[1], R. D. Tyagi[3,4], Patrick Drogui[1]*, Emmanuel Brien[5] [1.] *INRS Eau, Terre et Environnement, 490, rue de la Couronne, Québec G1K 9A9, Canada* [2.] *Institut de recherche et de développement en agroenvironnement, 2700 Rue Einstein, Québec, QC G1P 3W8, Canada* [3.] *Chief Scientific Officer, BOSK Bioproducts, 100-399 rue Jacquard, Québec G1N 4J6, Canada* [4.] *Distinguished Professor, School of Technology, Huzhou University, Huzhou, China* [5.] *VEOS Water, 265 Rue Armand-Majeau, Saint-Roch-de-l'Achigan, QC J0K 3H0, Canada*
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2.2. Abstract

Laundry related activities produce huge quantity of wastewater that is very rich in lint, dyes, surfactants, and detergents. The large amount of laundry wastewater (LWW) is generated by extensive human activities. LWW needs to be treated in order to tackle the challenging problem of water pollution and to accomplish water sustainability. To achieve this success, LWW can be reused for several purposes such as irrigation, construction activities, vehicle washing etc. However, there are several challenges in the reclamation of LWW like effective handling of the wastewater and meeting the regulatory criteria. Based on the literature review it seems that a single treatment process is not sufficient to treat LWW up to acceptable reuse standards. To that end, different treatment chains have been proposed: i) hybrid processes combining membrane filtration with adsorption; ii) advanced oxidation process using ultraviolet (UV) and hydrogen peroxide (H_2O_2); iii) coagulation-flocculation combined with sand filtration; iv) combination of ozonation process, adsorption, and ultrafiltration (UF). This review paper discusses the selection of suitable treatment technology depends on several factors: i) a well-designed equipment, ii) cost-effectiveness of treatment method and iii) desired characteristics of the treated water. The review paper also presents solutions for treatment and reclamation of LWW.

KEYWORDS: Laundry wastewater (LWW); Treatment technology; Reclamation; Reuse; Sustainability; Economic analysis; Circular economy; Integrated Processes; Membrane Filtration; Adsorption

2.3. Introduction

Water is among the most valuable natural resources available on the earth. Among the existing water on Earth, 97% is saline (unfit for consumption), and merely 3% is available for human utilization (Shirazi *et al.*, 2012). However, 68.7% of the freshwater is in the form of icecaps and glaciers, which makes it hard to use for drinking, household, or industrial purposes (Shirazi *et al.*, 2012). As a result, approximately 99% of the existing water on Earth cannot be used for daily purposes. Pollutants leading to the deterioration of communal health and natural surroundings are found in the wastewater that emerges from the laundries. This eventually results in water pollution that has been a matter of concern for all the countries around the globe in recent years. Also, there have been concerns related to the increased production of laundry wastewater (LWW), which impacts the environment, specifically in countries with emerging economies that are progressing rapidly. The laundry industry consumes a huge amount of water on a daily basis. To handle 1 kg of washing clothes, the laundry industry consumes 15 L water, producing 400 m3 of wastewater per day (Mauchauffee *et al.*, 2012). Such a substantial quantity of LWW demands operative procedures for wastewater remediation. Nevertheless, the lack of LWW treatment policies has been a perpetual setback.

Laundry additives, baking soda, soap, and detergent are utilized for different processes in the laundry (bleaching, white-work washing, starching and bluing) to make it free from dirt, oil, and grease. Therefore, the composition of LWW mainly contains compounds from soap, soda, and detergent. (Sheth *et al.*, 2017b) There is a large difference in the products used on commercial and residential scales, and hence there is a considerable variation in the composition of LWW at different scales. The sources of LWW generation are commercial laundries, household laundries, hotels, restaurants, and hospitals. The attributes of wastewater generated from all these laundries are almost the same except for hospitals laundries. These characteristics are dependent upon several factors like place of origin, the total number of people using the service, habits of the occupants, lifestyle, storage period, etc. LWW originated from commercial laundries have several pollutants like dye, phosphate-based detergents, surfactants like nonylphenol ethoxylates (NPEOs), sodium perborate, linear alkylbenzene sulfonate (LAS), and sodium percarbonate. The pollutants may contaminate the water system and results in eutrophication or harm aquatic life. In addition, hospital laundries consist of pharmaceutical products and chemicals. The aforesaid chemicals in the usable water can create a threat to the ecosystem. (Estahbanati *et al.*, 2021) A proper effective and eco-friendly management of LWW is necessary as utilization of surfactants over 0.5 mg L^{-1} poses a major health threat. (Rao, 2007)

The existing innovative technology arrangements conclude how the reuse of LWW is quite difficult but still achievable by applying integrated processes. (Dimoglo *et al.*, 2019; Panizza *et al.*, 2005) This review aims to summarize previous research published by experts round the globe, describing the characteristics of LWW and methods of LWW treatment. This would be extremely helpful in lieu of prior planning before any research is carried out on the subject. This work is also unique in its approach to resolve the drawbacks of single technology utilization for dealing with LWW. This review paper also emphasizes on combination of two or more treatment processes to give better-augmented results for the reusability of the LWW.

2.4. Laundry wastewater treatment

The habits and procedures followed during the washing of the clothes, along with the nature of the washing machine affect the composition of LWW. (Pakula & Stamminger, 2010) The orientation (horizontal vs. vertical axis) provided by the washing machine supplier defines the two categories of the machine available in the market. European countries have a large market for the horizontal ones, whereas the vertical ones are more popularised in American, Asian, and Australian cities. (Gooijer & Stamminger, 2016) Washing behavior includes the temperature, average load, and the total number of washing rounds. In a research on 38 countries, it was conveyed that the yearly water expenditure in Europe's laundry was approximately 10 m3 less than the other countries. (Pakula & Stamminger, 2010) The maximum water consumption was reported in Japan and Malaysia, as a consequence of which the highest amount of 60 to 80 m3 LWW/household/year was generated in these countries. The main reason credited by the author for this huge variation was given to the utilization of vertical axes in Asian countries that leads to more number of washing rounds. (Manouchehri & Kargari, 2017) Therefore, this evidence establishes the necessity for treatment and reusing of treated LWW for other purposes. The sources and reuse purposes of treated laundry wastewater are depicted in Figure 1.

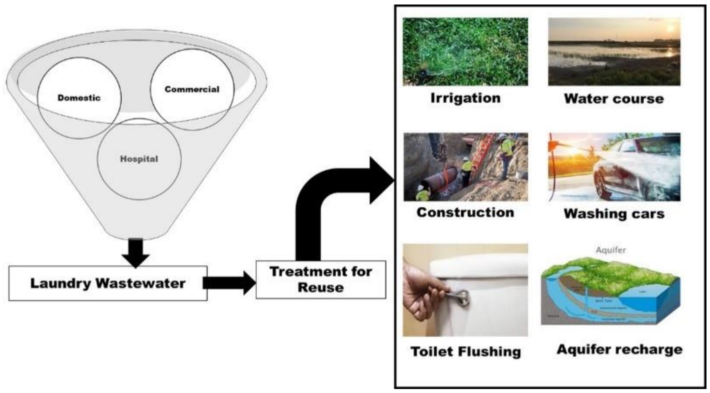

Figure 2.1:Depiction of source and LWW reclamation for reuse

As the initial characteristics of LWW differ widely between the individual apartments, it is not practical to install the same treatment unit for all of them. (Lu & Leung, 2003) However, several studies reported some applications for water recycling after small modifications in treatment approach for different types of LWW. (Ahmad & Hisham, 2008; Ciabattia *et al.*, 2009; Dimoglo *et al.*, 2019; Lu & Leung, 2003; Omolara, 2017) It was proposed in a study to collect separately LWW from all the individual apartments in a collection basin and then apply the treatment process. (Lu & Leung, 2003) The suggested processes can be implemented during the design stage of the building by providing separate plumbing facilities to each apartment for LWW collection. (Lu & Leung, 2003) The study suggests that LWW has high reuse potential for secondary purposes. (Lu & Leung, 2003) A study reported that, after simple treatment (for instance using chemical coagulation or ultrafiltration process), domestic LWW can be effectively used as toilet flushing water, irrigation water and also in the first wash cycle of the laundry. (Ciabattia *et al.*, 2009; Omolara, 2017)

Another study reported the reuse of treated commercial LWW in the wash cycles of the laundry. A treatment process combining (i) physico-chemical pre-treatment (coagulation, flocculation and Dissolved Air Flotation or DAF); (ii) sand filtration; (iii) ozonation; (iv) Granular Activated Carbon (GAC) filtration and (v) cross-flow ultrafiltration (UF) on flat membranes realised in polyvinyledene fluoride (PVDF). (Ciabattia *et al.*, 2009) The study indicates that, treated wastewater from GAC outlet fulfills the required permissible limit for discharge in Italy. Further treatment from UF membrane enhances the quality of treated wastewater, allowing it to reuse in washing process. (Ciabattia *et al.*, 2009) Other researchers reported the possibility to reuse of LWW generated from petroleum refineries in Pakistan. (Ahmad & Hisham, 2008) It was achieved by using an economical treatment system comprising of sedimentation and filtration units. (Ahmad & Hisham, 2008) The treated water was only used in the first rinsing cycle of the laundry. (Ahmad & Hisham, 2008) Another study reported the reuse of industrial LWW previously treated by electrocoagulation and electroflotation process. (Dimoglo *et al.*, 2019) The treated LWW were used in the washing process. At least, 90% removal of pollutants was achieved under the best operating conditions imposed: current density of 5.26mA/cm2, pH 5.5, and treatment time of 5 min. (Dimoglo *et al.*, 2019) Likewise, hospital LWW were subjected to treatment using coagulation-flocculation, anthracite filtration and advanced oxidation process (UV/H2O2) with a view to reuse treated effluent for washing process. During the treatment process low pressure UV-C lamp was used and treatment efficiency was investigated using factorial block design. (Zotesso *et al.*, 2017)

Properties of LWW

LWW generation source, along with the washing load, its procedure, and proficiency of the machines, sums up the measurable attributes of LWW. (Guilbaud *et al.*, 2010) LWW principally is categorized into industrial, domestic, and hospital laundry wastewater. (Manouchehri & Kargari, 2017) Amongst these, hospital LWW has been considered dangerous. (Lutterbeck *et al.*, 2020)

Physical parameters of wastewater are temperature, suspended solids (SS) concentration, color, turbidity, and electrical conductivity. Temperature is among the most noteworthy parameter as it governs the percentage of reaction and existence of microbes, which is necessary for aerobic wastewater treatment. Normally, temperature for LWW lies between 30-36˚C. (Eriksson *et al.*, 2002; Nicolaidis & Vyrides, 2014) Measurement of the turbidity and SS of LWW provide some data about the concentration of colloidal fragments that might clog pipes, used for transportation of wastewater. Turbidity for domestic laundries, commercial laundries, and hospital laundries wastewater has been described in the range of 14-400 NTU, 40-150 NTU, 87.9 NTU, respectively. (Mostafazadeh *et al.*, 2019; Omolara, 2017) Electrical conductivity for wastewater lies between 190-1400 µS/cm for domestic laundries, 640-3000 µS/cm for commercial laundries, and 808-2000 µS/cm for hospital's laundry wastewater. (Mostafazadeh *et al.*, 2019; Noutsopoulos *et al.*, 2018; Omolara, 2017) SS has been reported in range of 16-330 mg/L, and total solids (TS) vary between 113-2410 mg/L. (Eriksson *et al.*, 2002) The biological and chemical characteristics of LWW are very important from the treatment perspective. The main chemical characteristics of LWW are pH, biochemical oxygen demand (BOD), nanophenol ethoxylate (NPEO), chemical oxygen demand (COD), linear alkylbenzene sulphonate (LAS), Dialkyldimethylammonium chlorides (DDAC, DSDMAC), nitrates and nitrites, and phosphate. The biological parameters include the occurrence of microorganisms in wastewater. For the commercial laundry, the typical value of pH lies between 9-11, the

COD value is in the range of 376-910 mg/L, and BOD value is between 48-1200 mg/L. (Moura *et al.*, 2019; Sheth *et al.*, 2017a) Hospital laundry wastewater encompasses all the above parameters along with some pathogens and some pharmaceutical products. (Lutterbeck *et al.*, 2020) So, for the treatment of hospital laundry wastewater, a separate treatment process such as disinfection using catalytic photozonation is required, which can take care of the contaminants along with surfactants. (Kist *et al.*, 2008) The properties of laundry wastewater in accordance with the source of origin have been stated in Table 1. Table 2 outlines LWW characteristics in different countries.

Table 2.1 Characteristics of LWW in accordance with the source of origin

Parameter	Domestic Laundry (Omolara, 2017; Sheth *et al.*, 2017a)	Commercial Laundry (Omolara, 2017)	Hospital Laundry (Omolara, 2017)	Commercial Laundry (Mostafazadeh *et al.*, 2019)	Commercial laundry (Moura *et al.*, 2019)
Temperature (˚C)	30-34	u.p.	u.p.	u.p.	u.p.
pH	7.88-10.32	9-11	11.4-11-6	9.2±0.5	8.9±1.3
Conductivity (µS/cm)	190-1400	640-3000	808-2000	715±20	u.p.
Turbidity (NTU)	14-400	40-150	87.9	71±3	u.p.
TDS (mg/L)	640-1455	420	456-800	400±20	u.p.
TSS (mg/L)	445-1550	Apr-0	66-71	60±20	84±58
COD (mg/L)	376-910	u.p.	477-876	628±20	745±152
BOD (mg/L)	48-1200	218-9810	44-50	u.p.	u.p.

**u.p..: Unpublished*

Table 2.2 Characteristics of Laundry wastewater according to different countries

		India	China	Kenya	Israel	Canada
Physical	Turbidity (NTU)	390	858 ± 111	u.p.	u.p.	71 ±3
	Temperature (˚C)	30.7	40 ± 1.0	24	u.p.	u.p.
	TSS (mg/L)	u.p.	359 ± 82	u.p.	188	60 ±20
	TDS (mg/L)	u.p.	357 ± 52	u.p.	u.p.	400 ±20
Chemical	pH	7.3	12.5 ± 0.5	7.3-10.3	7.5	9.2 ±0.5
	Conductivity(mS/m)	9000	124	1526	2457	715 ±20
	TOC (mg/L)	u.p.	u.p.	u.p.	361	u.p.
	TN (mg/L)	u.p.	u.p.	u.p.	4.9	u.p.
	TP (mg/L)	4.2	22 ± 4	u.p.	169	u.p.
	Salinity (ppt)	u.p.	u.p.	0.6	u.p.	u.p.
	BOD (mg/L)	u.p.	u.p.	6250	462	u.p.
	COD (mg/L)	1339	1138 ± 58	u.p.	4200	628 ±20
	DO (mg/L)	u.p.	u.p.	3.7	u.p.	u.p.
	References	(Gunarathne & Rajakaruna)	(Kim *et al.*, 2014)	(Kotut *et al.*, 2011)	(Friedler *et al.*, 2013)	(Mostafazadeh *et al.*, 2019)

**u.p.: Unpublished*

The limits in accordance to British regulations for raw LWW on total Kjeldahl nitrogen (TKN), *E. coli*, chemical oxygen demand (COD), and biochemical oxygen demand (BOD) measurably surpassed at a margin of 92.0%, 89.0%, 73.8%, and over 100%, respectively as per the studies and experimentation of Ho et al. (Ho *et al.*, 2020). In the residential LWW, considerable COD, BOD, and anionic surfactants have been detected in high concentration. (Ho *et al.*, 2020) The augmentation of the biodegradability of LWW was established by the mean ratio of COD to BOD as 1:5, which was calculated by Makropoulos et al. (Makropoulos *et al.*, 2018). Residential, domestic laundries showed a presence of heavy metals too. A small concentration of lead and chromium was detected, but elevated concentration of zinc (0.20 ± 0.16 mg/L) and copper (43 ± 29 µg/L) was found probably because of plumbing materials and coatings of the water outlets. The plumbing materials and coatings from the pipe network leach out during LWW disposal. The exceeding levels of turbidity, total phosphorus (TP), conductivity, and total suspended solids (TSS) were detected in the LWW produced from industries when compared to domestic LWW sources, reported in Kim et al.(Kim *et al.*, 2014) and Shang et al. (Shang *et al.*, 2015). Some of these values were found to be more than that of the discharge limits

(Table 3). For example, TP was 26 mg/L, which was more than the required limits of 18 mg/L. All solids classes combined were discovered in high levels when residential LWW was mixed with university campus LWW. (Jacangelo *et al.*, 1991) When compared with another LWW sources, relatively less polluted wastewater was discovered in LWW collected at the university campus. (Chrispim & Nolasco, 2017) Alternatively, extensive COD (500-580 mg/L) along with high amounts of linear alkylbenzene sulfonate (LAS) (13.2-15.0 mg/L) were observed in LWW that was generated after washing of the bedding used in trains. (Leu *et al.*, 1998) Finally, slightly excessive total organic carbon (TOC), COD, and hardness like magnesium and calcium were observed by Guilbaud et al. (Guilbaud *et al.*, 2010); while experimenting and studying with LWW that was generated from a ship. (Guilbaud *et al.*, 2010) Treatment of LWW according to the discharge limits and its reuse is the need of the hour because of several applications of treated LWW. Therefore, effective LWW remediation technologies have been advocated for water-scarce regions to resolve the problem of water availability and management (Espósito *et al.*, 2009).

Regulatory standards for reuse of treated LWW

Documentation describing LWW reclamation and reuse benefits has been reported. (Ahmad & Hisham, 2008; Ahn & Song, 1999; Benis *et al.*, 2021; Bilad *et al.*, 2020; Ciabattia *et al.*, 2009; Dimoglo *et al.*, 2019; Hoinkis *et al.*, 2012; Kim *et al.*, 2005; Li *et al.*, 2009; Noutsopoulos *et al.*, 2018) Water reclamation and reuse contribute to solving water scarcity problems. (Ho *et al.*, 2020; Kumar *et al.*, 2021) The development of a new water source after reclaiming LWW has been reported, which helps in reducing the expenses of freshwater consumption. (Estahbanati *et al.*, 2021; Kumar *et al.*, 2021; Zaroual *et al.*, 2006) Nonetheless, purpose of reusing or reclaiming wastewater are restricted to their aims like drinking water for human or livestock, irrigation, industrial cooling water, or construction. The recommended specifications and regulations related to suitable water consumption are set all over the world by various local and international institutions, following which if the limit is obtained, the wastewater is consumable and can be utilized.

It is observed that regular LWW do not meet the acceptance limits of phosphate, nitrate, pH, SS, nitrite, iron, COD, ammoniacal nitrogen, and copper. Thus, the required, acceptable levels of these parameters need to be brought down by additional treatment. National Water Quality Standards (NWQS), as set by the governmental agencies all over Canada regulating irrigation and livestock drinking purpose, can only be achieved after treating LWW by efficacious methods. (Ahmad *et al.*, 2012) The LWW used for irrigation purpose has reduced stringent necessities compared to livestock drinking, which takes into account the health hazards on cattle and the customers. Employing treated LWW for agricultural purposes, can act as a substitute to commercial fertilizers because of presence of phosphorus and nitrogen in treated LWW.

LWW falls into the category of greywater, which can be reclaimed and reused for drinking purposes. In some countries like Australia, Singapore, and Namibia have developed and applied technologies to treat LWW for drinking purposes. (Kim *et al.*, 2015b) A standardized guideline for drinking water quality was created by the World Health Organization (WHO, 2011) on an international level. These guidelines were adapted and further utilized to set domestic drinking water consumption guidelines by the countries worldwide. The Public Utilities Board (PUB) in Singapore, based on WHO’s strategies, developed a product called NEWater that strictly followed these guidelines. Utilizing its innovative island-wide sewage protocol, this product is made fit for reuse through a chain of treatment methods using double membrane filtration, conventional wastewater procedure, and ultraviolet (UV) treatment technology. (Mauchauffee *et al.*, 2012) Since 2003, NEWater technology has been extensively consumed in domestic as well as in industrial purpose all over Singapore.

The data shown in Table 3 was included from several references from innumerable international specifications for watercourse discharge, drinking water, cattle drinking water, irrigation water, and construction water eminence. Table 3 gives all the LWW parameters and their permissible limits in various watercourses. Conversely, strict limits are followed for the extreme occurrence of COD and LAS. Consequently, merely by handling COD along with LAS can create LWW throwaway to watercourses.

Table 2.3 International permissible limits for LWW

Parameters	Average values for LWW	Permissible limits					
		Irrigation	Cattle drinking water	Water course	Drinking water	Reuse in laundry	Toilet flushing
pH	5.6± 0.9	6.5- 8.4	5.0-9.0	6.0-9.0	6.5-8.5	6-8	--
Turbidity (NTU)	--	--	--	--	--	<25	≤ 5
Total Alkalinity (mg/L)	25.9± 20.2	1.5-8.5	--	200	--	<100	--
BOD_5(mg/L)	--	--	--	--	--	--	≤ 20
LAS (mg/L)	163±247	--	--	15	--	--	
TSS (mg/L)	--	--	--	--	--	5-25	≤ 20
Sulphate (mg/L)	21.1 ±19.1	0-20.0	< 500	500	400	<10	--
Sulphide(mg/L)	0.2 ± 0.1		0.0-0.33	0.2	10.1	--	--
Nitrate (as NO_3^-) (mg/L)	8.4 ± 6.8	5.0-30.0	45-132	--	50	--	--
Nitrite (as NO_2^-) (mg/L)	2.1 ± 0.8	--	--	--	1.5	--	--
N-ammoniacal (mg/L)	7 ± 10.8	--	--	--	3	--	--
Phosphate (mg/L)	94.6 ±75.4	0.0-2.0	0.0-1.0	<10	--	5-10	--
Zinc (mg/L)	0.56 ± 0.8	2	24	1	5	--	--
Lead (mg/L)	0.06 ±0.05		0.1	0.1	0.01	--	--
E coli (CFU)	--	--	--	--	--	--	≤ 200
References	(Braga *et al.*, 2015)	(Energy *et al.*)	(Heinz *et al.*, 2011)	(Nelson *et al.*, 2008)	(Edition, 2011)	(Dimoglo *et al.*, 2019)	(Health Canada, 2010)

Surfactants, microfibers and microplastics: source of emerging contaminants

Surfactants like NPEOs, LAS, and DDAC are widely employed as detergents in the laundry industry. They are considered as emerging contaminants and are continuously discharged, creating an adverse potential effect on the environment. Surfactants are amphoteric in nature, which permits them to behave like acid or base therefore, they are employed as cleansers in several industries. Surfactants may exist in numerous forms like cationic, anionic, and non-ionic; mainly, they exhibit LAS. However, surfactants, when enter the water bodies, they can adversely impact aquatic life and also human health. The literature clearly demonstrates the adverse effect of surfactants on flora, fauna, and human health. The excessive concentration of LAS may cause respiratory problems in humans and may also cause skin irritation. (Ríos *et al.*, 2018a; Ríos *et al.*, 2018b) On the other hand, anionic surfactants exhibit toxic character attributable to their highly reactive nature with cellular units and combine with several macromolecules like DNA, peptides, and protein, causing damage of their function. (Ríos *et al.*, 2018a; Ríos *et al.*, 2018b) The measure of inhibitory activity of a substance on a specific biological or biochemical function is referred as the half-maximal inhibitory concentration (IC50). Könnecker and co-workers discovered that the IC50 standards for Daphia magna, fish, and algae are 9.6 mg/L, 8.9 mg/L, and 12.8 mg/L, respectively. (Könnecker *et al.*, 2011)

Thus, eliminating LAS from laundry wastewater is essential to decrease deleterious environmental effects. Although various categories of chemical and physical treatment methods exist, most of the techniques are economically not feasible and yields toxic by-products. Toxic by-products are more hazardous than preliminary compound. The practice of biological treatment processes can eradicate the poisonous compounds and change them into less poisonous by-products. (Annadurai *et al.*, 2008)

Microplastics (MPS) and nanoplastics (NPS) are other emerging contaminants that are present in LWW. Synthetic fibers released during the washing process are the major source of MP in LWW. (Galvão *et al.*, 2020) Studies indicate that LWW is one of the primary contributors of microfibers (MFS) in the aquatic system. (Shen *et al.*, 2022) Studies also indicate that these microplastics can stay in the environment for many years due to chemical stability. Due to its large surface area, endurance, and fluidity, MPS can readily adsorb metals, harmful bacteria, and organic chemical pollutants, which results in the increase of microplastics in the aquatic environment. (Antunes *et al.*, 2013; Fendall & Sewell, 2009; Li *et al.*, 2018) Because of the small size and light density MPS and NPS are easy to consume by aquatic animals. These MPS can cause an ecotoxicological effect on aquatic animals and adversely affect human health. (Galvão *et al.*, 2020; Kiendrebeogo *et al.*, 2022) The accumulation of MPS can cause intestinal blockage and abrasion in aquatic animals. (Slootmaekers *et al.*, 2019)

Currently, MPs have not been included in the Effluent Discharge Objectives (EDOs) of municipalities, but due to their serious environmental effects, it is necessary to develop feasible and cost-effective treatment technology. Although most of the MPS (>90%) can be eliminated in the wastewater treatment plants, but still some fraction of MPS bypasses the system and enters to the environment. So far, no specific treatment process has been implemented in the wastewater treatment plants (WWTPs) for the removal of MPs and NPs, with the exception of existing technologies, such as sedimentation, grit chamber, skimming, screening, membrane bioreactor, and tertiary filtration. Moreover, many researchers are working to develop new technology approaches for MPs and NPs degradation in wastewater. (Kiendrebeogo *et al.*, 2021; Kiendrebeogo *et al.*, 2022)

Effects on human health and the environment

The wastewater generated from the laundry activity is highly rich in SS, energy (heat), dye, surfactant, total solids, and COD. The surfactants like NPEOs are the chief ingredients of detergent, which is considered into consideration as an endocrine-disrupting compound. When water containing NPEOs is directly discharged within the open water bodies, it can adversely affect the ecosystem and the aquatic animals. It may lead to genetic mutation of the aquatic animals and can affect the reproduction capability of the fish. (Sandbacka *et al.*, 2000) NPEO may be toxic when it exceeds a certain aggregate in water bodies. (Talmage, 1994)

The surfactants that are cationic, anionic, and non-ionic in nature exhibit hydrophilic and lipophilic nature. The lipophilic tail is biologically active with surfactants. The anionic surfactants possess the ability to get attached to the bioactive macromolecules such as peptides, enzymes, DNA. As a result, surface charge on the bioactive macromolecules get altered along with improper folding of polypeptide chain. On the other hand, the cationic surfactants may attach to the bacterial membrane, and can alter the performance of the bacteria. The non-ionic surfactants are capable of combining with phospholipid membrane and proteins, resulting in the increased permeability of the membrane and vesicles. An increase in the permeability of the cell membrane can lead to cell disruption due to release of low molecular weight compounds from the cell. (Ivanković & Hrenović, 2010)

2.5. Laundry Wastewater Treatment Processes

Overall, the LWW treatment system may be divided into three principal procedures: biological, physicochemical, and the combination of biological and physicochemical methods (Fig. 2).

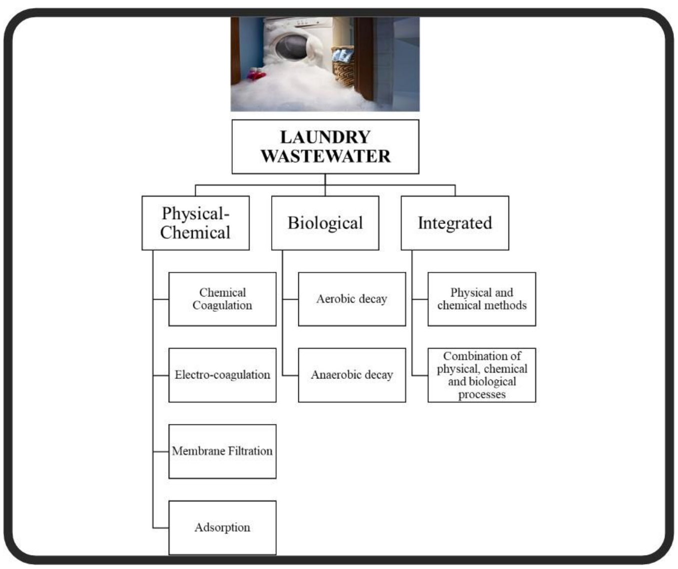

Figure 2.2: Laundry Wastewater Treatment Processes

2.6. Physical – Chemical

Chemical Coagulation

Since many decades, chemical coagulation has been utilized for wastewater treatment. The inorganic coagulants like aluminium sulphate or iron salts (ferric sulphate, ferrous sulphate or ferric chloride) were the first ones to be used for the treatment. However, $FeCl_3$ is corrosive and $FeSO_4$ works only in the pH range of 8.5-11. Therefore, preference was given to aluminium sulphate and ferric sulphate which can be utilised in a varying pH range of 5-9 and 4-11, respectively. (Mostafazadeh et al., 2019) Hazardous by-products like metal hydroxides are produced with the usage of above coagulants, which creates an additional problem of removing these metal ions from the treated water. Therefore, chemical coagulation process must be combined with adsorption. A high efficacy has been found with ligand based composite adsorbents that are selective for removal of metal ions from the LWW. (Awual, 2017) Natural and synthetic organic coagulants are supposed to give high efficacy with lower doses and hence, they tend to produce less volumes of sludge. They provide higher removal of turbidity and COD than the inorganic coagulants. (Dotto et al., 2019) However, best results were obtained when low molecular weight polymers were added as coagulants during LWW treatment. (Kim et al., 2014)

For chemical coagulation process, COD and turbidity removal rate is dependent on SS concentration in LWW. (Shang et al., 2015) High COD and turbidity removal rate is generally observed for high SS concentration. For high removal rates, working pH, coagulant concentration, sedimentation time and mixing speed must be experimentally optimized. Generally, jar tests are employed for this type of experiments. (Shang et al., 2015)

The effectiveness of chemical coagulants like ferric chloride and polyaluminum chloride (PACl) was studied for microfibers removal from LWW. (Li *et al.*, 2022b) It was found that 90-94% of microfibers were removed using ferric chloride in the absence of detergent, and 90% removal efficiency was obtained using PACI. (Li *et al.*, 2022b)

Electrocoagulation

The pollutants like ions, organic colloids, and inorganic colloids are removed from various wastewater effluents by using electrocoagulation process (EC). During the treatment process wastewater passes through the EC cell and voltage is applied. At the anode electrode, EC delivers the coagulant in situ by anodic dissolution in wastewater subjected to treatment. At the cathode surface, water is reduced into hydrogen gas and hydroxyl ion. The dissolved metal ions (coagulating agents: Al^{3+} or Fe^{2+}/Fe^{3+}) then encounter suspended particles to neutralize the negative surface charge. The coagulating agents can also react with hydroxyl ions and produce iron (or aluminum) hydroxides having a considerable sorption capacity. These metal hydroxides can simultaneously remove suspended solids, heavy metals, and other pollutants by co-precipitation or complexation. Figure 3 shows the treatment mechanism of EC process.

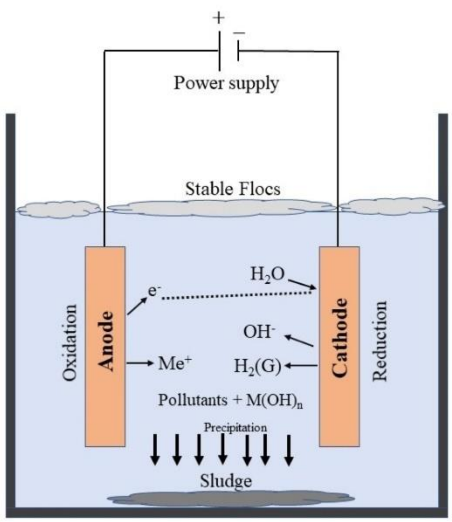

Figure 2.3: Treatment Mechanism of EC Processes

The active coagulant precursors are produced when sacrificial anodes are dissolved via applied potential. (Hoinkis & Panten, 2008) Table 4 summarizes some remarkable findings using electrocoagulation for handling LWW. Arrangement of dissimilar electrodes in EC has proved to be beneficial in terms of minimizing energy consumption, as discussed by various researchers. (Chen, 2004; Mollah et al., 2001; Tyagi, 2022)

A moving particle electrode granular activated carbon (GAC) was utilized under optimized specific operating conditions (current intensity 354.3 mA, pH 5.4, reaction time of 55 minutes, and GAC dosage of 47.1 g/L) for LWW treatment. Then, treated LWW was used for irrigation and construction activities. (Jung et al., 2015) In supplementary research by Janpoor et al. (Janpoor et al., 2011) , utilizing additional four aluminum electrodes, created an extensive range of managed laundry effluent evaluations and also testified optimum circumstances (pH= 6, 30 V, and inter-electrode distance of 1.5 cm) to accomplish best elimination efficacy: 93.2% COD, 90.1% color, 96.7% phosphorus, and 93.5% detergent. (Janpoor et al., 2011) The maximum turbidity removal rate was achieved after 90 min of treatment. The enmeshment in a precipitate and the neutralization of surface charges (consequently stimulating coagulation) are the two elimination procedures that eradicates these contaminants utilizing the classes of aluminum species generated. An impermeable film on the cathodes is formed as a consequence of the passivation of aluminum anodes which may interfere with the performance of electrocoagulation. (Holt et al., 2005) The energy consumption increases when a high voltage for long terms is applied for the breaking of the inhibition layer produced on the electrode surface. Also, the electrodes are exhausted after such long-run usages. The performance of this technology was improvised through the application of a low-frequency sonic field (40-200Hz) to overcome the above-mentioned problems SO^{4-} and Cl^{-} were coagulated after augmenting the iron hydroxide concentration. (Kovatcheva & Parlapanski, 1999) Some discrete combinations of electrodes in a relative investigation were also utilized underneath acidic circumstances (pH=3). (Dehghan et al., 2018) An efficacious 87.1% and 75.1% COD removal was achieved with iron electrode pair and iron-aluminum electrode pair, respectively, due to the dissipation of iron into divalent Fe(II) and trivalent Fe(III) ions and sturdily dissipation of aluminum into trivalent Al(III) ions. Lastly, the decay of harmful contaminants was simplified by the supplementation of oxidizing materials (like hydrogen peroxide) under specific operating conditions like pH=5, current density of 0.5 mA/cm, and 10 min of treatment time. In the study 81.6% of elimination efficacy was achieved for SDS removal using above process. (Yüksel et al., 2009)

Likewise, electrocoagulation process can effectively be used to remove MPs and NPs from LWW. More than 90% of MPs and NPs were removed from wastewater while using aluminum electrode operated with 10V applied voltage density during 6h of retention time at pH 7.2. (Shen et al., 2022) In a similar study, 98% removal of MPs was obtained using Fe-Al electrode with 2.16 A current intensity at pH 9 and for 60 min treatment time. (Akarsu & Deniz, 2021) Another study suggests that electrooxidation (EO) and electro-peroxidation (EO-H_2O_2) can effectively degrade 86.8% of NPs from wastewater. (Kiendrebeogo et al., 2022)

In another similar study, Kiendrebeogo et al. (Kiendrebeogo et al., 2022) showed that up to 89 ± 8% of MPs could be degraded by using electrooxidation process comprising of boron doped diamond (BDD) anode operated at a current density of 108.4 $mA \cdot cm^{-2}$ and in the presence of 0.03 M Na_2SO_4 used as supporting electrolyte. (Kiendrebeogo et al., 2022) The scanning electron microscopy (SEM) analysis of the remained MPs in water after the electrooxidation demonstrated all of them were unchanged, suggesting the complete mineralization of MPs at the anode surface. (Kiendrebeogo et al., 2022) Dynamic light scattering (DLS) also confirmed this suggestion by observation of the formation of no MPs with lower size than the initial MPs. It was also verified by Fourier transform infrared spectroscopy (FTIR) detecting no C=O bond by FTIR analysis on the MPs after electrooxidation. (Kiendrebeogo et al., 2022)

This process has innumerable benefits: easy, cost-effective, environment-friendly, flexible, competent, and broad-spectrum (being capable to eliminate concurrently various kinds of contaminants) (Chen, 2004). Nonetheless, a consistent substitute of the sacrificial anode is essential while the anode disperses in the solution, and cathode passivation may happen that diminishes the ability of electrocoagulation. To tackle the challenge of passivation and short operation time, the suitable electrode material can be investigated, which can provide better treatment efficiency and less passivation. Another strategy could be the addition of continuous coagulant dose while maintaining the cell potential low during extended EC operations. Using alternative current (AC) supply, polarity reversal, ultra-sonification, and increasing the flow velocity to increase the scouring could be another solution. Besides this, additional research needs to be performed to accomplish high performance in electrocoagulation through statistical modeling and computational fluid dynamics.

Table 2.4 LWW Treatment using Electrocoagulation

Material Used	Pollutant concentration	Operating Conditions	Removal %	References
GAC (Moving Particle Electrode)	COD = 334.13 mg/L	Current = 354.3 mA Reaction time = 55 min GAC concentration = 47.1 g/L pH = 5.4	COD = 98.56%	(Jung *et al.*, 2015)
Iron	COD = 398 mg/L Turbidity = 186 NTU	Initial pH = 3 Operating Voltage = 30 V Treatment time = 60 min	COD = 87.1%	(Dehghan *et al.*, 2018)
Iron	SDS dose = 60 mg/L	Current density = 0.5 mA/cm^2 Treatment time = 10 min Initial pH = 5	SDS = 81.6%	(Yüksel *et al.*, 2009)
Aluminium	COD = 4155 mg/L Surfactant= 463 mg/L Turbidity = 245 NTU TSS = 987 mg/L	Initial pH = 6-8 Operating Voltage = 30 V Distance between electrodes = 1.5 cm HRT = 90 min	COD = 93.2% TP = 96.7% Surfactant = 93.5% Color = 90.1% Turbidity = 95.9%	(Janpoor *et al.*, 2011)

Membrane filtration

The high separation efficiency has been a major reason for adopting membrane technology as an economical and eco-friendly process for treating wastewaters (Bilad *et al.*, 2020). The existing volumes of surfactants in LWW have been the center for the selection of an appropriate membrane. (Tripathi *et al.*, 2013) The critical micelle concentration (CMC) is attained when coalescence of surfactants happens inside the aqueous solution, and spherical micelles are generated.

Inside an aqueous solution, surfactants combine to form spherical micelles when CMC is attained in solution. The range of spherical micelles generated by CMC can be between 2-20nm, as a consequence, microfiltration and ultrafiltration membranes are chosen when the retrieval of surfactants is on or beyond CMC. Alternatively, nanofiltration membrane is acclaimed during circumstances where it cannot surpass the CMC value. (Tripathi *et al.*, 2013) The statistics on the efficiency of membrane filtration for LWW treatment are presented in Table 5.

Table 2.5 LWW treatment using membrane filtration

Membrane used	Operating conditions	Feed	Removal	References
UF membrane (Submerged: pore size = 6.2 nm)	Operating Temperature = 20˚C Initial pH = 7.5 Applied Pressure = 0.12 bar	TOC = 161 mg/L Turbidity = 140 NTU	TOC = 83.4% Turbidity = 99.9%	(Li *et al.*, 2009)
NF (AFC 80)	pH = 7 Temperature = 40˚C Pressure = 3.5 bar	COD = 1350 mg/L	COD = 93%	(Guilbaud *et al.*, 2010)
MF (pore size: 0.22 μm)	Pressure = 1 bar Flow rate = 44 L/h	TSS = 240 mg/L COD = 2538 mg/L Turbidity = 360 NTU BOD = 1190 mg/L	BOD = 93.9% COD = 90.8% TSS = 98.7% Turbidity = 98.7%	(Manouchehri & Kargari, 2017)
UF membrane (MWCO = 5 kDa)	pH = 10.9 Operating Temperature = 45˚C Applied Pressure = 2.5 bar	COD = 360 mg/L	COD = 90.0% TSS = 92.7%	(Ashfaq *et al.*, 2017)

**UF: Ultrafiltration, NF: Nanofiltration, MF: Microfiltration, MWCO: Molecular weight cut-off*

Studies have been reported for reusing treated LWW post-MF treatment utilized a MF membrane made of cellulose nitrate for LWW treatment and permeate (treated LWW) could be employed for sanitary toilet flushing or landscaping. (Omolara, 2017) But treated LWW could not be reused for washing laundries because it did not follow permissible limits of BOD content (10-30 mg/L). (Omolara, 2017) However, application of a separate MF membrane - mixed cellulose ester (MCE), generated an effluent of 233.5 mg/L COD with COD elimination of 90% in the permeate, that needed further dilution before its use in sanitary toilet flushing. (Manouchehri & Kargari, 2017) Ashfaq et al. (Ashfaq *et al.*, 2017) studied that more advanced treatment process is required in case of hospital LWW, besides MF. (Ashfaq *et al.*, 2017) As UF membrane was utilized with molecular weight cut-off (MWCO) of 5kDa, after which the authors found 90% COD elimination in the permeate under following operating conditions: transmembrane pressure = 2.5 bars, flow rate = 2.5 LPM.

The reutilization of LWW (initially treated by MF) in toilet flushing does come with a considerable number of challenges. Even though UF membranes procedures effectively eliminate SS, the permeate still possess high concentration of organic materials and according to EU guidelines, can not be used as bathwater. (Li *et al.*, 2009) Implementation of a rotating disk system (RDM, at 2000 rpm) attached to NF270 membrane to treat genuine detergent wastewater produced 350 L/m2 h permeate flux and 97% COD elimination. (Luo *et al.*, 2019) Apart from the various innumerable advantages of membrane technology, it has certain shortcomings as well. The major shortfall is membrane fouling, which occurs when the particles are accumulated on the surface and/or within the pores of the membrane. The membrane fouling demands repeated washing to re-establish the permeate flux and, utilizes more energy at high pressures. (Chrispim & Nolasco, 2017; Mat Nawi *et al.*, 2020) To alleviate the drawbacks of membrane filtration, solutions like hydrodynamic operation (control of turbulent flow), pre-treatment techniques, and membrane surface modifications can be employed to treat the LWW as per the limits.

In general, it has been determined that single-step treatment is inefficient for producing high-quality effluent which fulfills the allowed limits for wastewater reuse. (Abdel-Kader, 2013; Ashfaq *et al.*, 2017; Schouten *et al.*, 2007; Veli *et al.*, 2019) One-step membrane filtration is a feasible process for LWW treatment and there have been recent advancements in membrane manufacturing techniques. It has been reported that use of small pore size membranes (NF and UF membrane) in single-phase filtration, may generate high quality permeate for on-site recovery. (Guilbaud *et al.*, 2010) Hospital LWW treated through a single-step UF filtration (MWCO: 5 kDa) has been investigated to generate reusable water in China, USA, and Italy. (Ashfaq *et al.*, 2017) NF filtration system has been installed at marine ships to handle LWW to remove microbes and SS while resulting in low COD (48 mg/L) and TOC (7 mg /L) in treated LWW. (Guilbaud *et al.*, 2010) Eighty percent of treated LWW was recovered and reused for washing of the laundry. The authors reported 35% decline in permeate flux due to membrane fouling by fibres and pollutants in LWW. To resolve the above problem, the authors recommended use of NF RDM. Shear strength dynamic filtration was used to reduce gel layer formation and concentration polarization. This enhanced permeate flux from 26% to 57%, while COD removal increased by 4%. (Luo *et al.*, 2019)

UF membranes made of polysulfone could remove up to 80% sodium dodecycl sulphate (SDS) surfactant from LWW. (Majewska-Nowak *et al.*, 2005) However, the surfactant retention rates, and the permeability of membranes rapidly decrease with the increase of surfactant concentrations. (Majewska-Nowak *et al.*, 2005) In order to solve the problem of membrane permeability and stability during laundry wastewater treatment (for long-time operation), many researchers have suggested the use of ceramic membranes. Ceramic membranes have higher chemical and thermal stability, excellent structural integrity, and low fouling tendency and can be effectively used for LWW treatment containing relatively high surfactant concentrations. (Kim & Park, 2021; Šostar-Turk *et al.*, 2005; Suárez *et al.*, 2014) For instance, the permeability of the ceramic membrane was much higher (50 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$) than the polymeric organic membrane (4.2 and 29.2 $L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$). (Fernández *et al.*, 2005; Majewska-Nowak *et al.*, 2005) Another study shows that the ceramic UF membrane was more effective in removing suspended particles compared to removing organic matter removal. (Kim & Park, 2022)

Adsorption

Various kinds of adsorbents retain active functional groups, which deliver selective affinity through electrostatic interaction or synchronization mechanisms with respect to innumerable surfactants (BHAGAT, 2018). Schouten et al. (Schouten *et al.*, 2007) reported variation of adsorption efficacy due to the surface charge and the adsorbents' pore size. (Schouten *et al.*, 2007) The ionic interfaces of the cationic resin will help in promoting adhesion of an anionic surfactant. On the same hand, dispersion of surfactant molecules will be controlled by both macropores (450 nm) or mesopores (2–50 nm) together. They adsorb the surfactant molecules at the interior core of adsorbents. (Ríos *et al.*, 2018b) There have been substantial attempts for activation of different adsorbents at varying temperature and pH Zhang et al. (Zhang & Ding, 2015) studied the primary amino-cross-linked chitosan microspheres (ACCMs) to demonstrate strong affinity towards anionic surfactants that preserved 94.3% adsorption following 8 adsorption-desorption cycles under sub-optimal temperature and pH. (Zhang & Ding, 2015) Adsorbents for LWW treatment can be chosen based on type of surfactant – anionic or cationic. (Terechova *et al.*, 2014) Approximately 95% heavy metals were eliminated from treated LWW by using activated carbon generated from banana stem waste. (Gomes *et al.*, 2007)

Moreover, iron oxide powder (hematite and magnetite) is an efficient adsorbent for removal of cationic surfactants. Large amount (17 g/L to 51 g/L) of the adsorbent was crucial to attain 90% elimination of cationic surfactants in 10 minutes. (Braga *et al.*, 2015) In another study, zeolite-4A provided better performance than other adsorbents materials (vermiculite and zeolite) to reach adsorption equilibrium, which was found to be effective for 98.7% removal of radionuclide species ($Sr^{2+,}$ Cs^{+}, and Co^{2+}) from LWW of nuclear facilities. (Jia *et al.*, 2014) A record of notable studies on adsorption have been depicted in Table 6. To sum up, adsorption for LWW management is contemplated as an efficient and cost-effective technique, which can eliminate pollutants from wastewater. As cited, several parameters influence the efficacy and functioning of adsorbents like pH, preliminary pollutant concentration, stirring speed, ionic strength, contact time, adsorbent amount, coexisting ions, and temperature.

Table 2.6 LWW treatment by Adsorption

Adsorbent	Feed	Operating Conditions	Removal Efficiency	References
Allophane	LAS Dosage = 0.002 M	pH = 6 Treatment time = 5 h Allophane dose = 33.3 g/L	20%	(Nikaeen *et al.*, 2016)
Micrometric iron oxide powder (magnetite and hematite)	Surfactant dosage = 0.2–4.2 mg/L	Adsorbent concentration: (Magnetite = 17–51 g/L and Hematite = 1- 17 g/L) pH = 8 Operating Temperature = 25˚C Duration = 10min.	Anionic surfactant = 20% Cationic surfactant= 90% Non-ionic surfactant = 40%	(Bokhary *et al.*, 2018)
Primary amino crosslinked chitosan microspheres	Anionic surfactant = 10-2000 mg/L	pH = 3 Temperature = 25˚C	SDBS = 1220 mg/L SLS = 888 mg/L SDS = 825 mg/L	(Zhang & Ding, 2015)
Multi-walled carbon nanotubes	LAS dosage = 50 mg/L Na_2SO_4 dose = 0.1 mol/L	Operating pH = 5 Operating Temperature = 25˚C CMMWCNTs concentration = 0.01% (w/v) Stirring speed = 250 rpm	LAS = 350 mg/L	(FENG *et al.*, 2017)

2.7. Biological Treatment

Aerobic decay of contaminants has been reported via numerous techniques, such as batch biodegradation, aerobic bioreactor, or trickling filters. (Baumann *et al.*, 1981) The study of a two-stage aerobic moving bed bioreactor (MBBR) (Kaldnes K5 as bio-filter media) was carried out to effectively treat LWW. MBR was successfully operated for four months which resulted in removal of 95-98% BOD_5, 85-96% surfactant and finally 89- 94% of COD. (Bering *et al.*, 2018) The sequencing batch biofilm reactor (SBBR) setup removed approximately 86.5% of COD, > 85% of micropollutants, 98.4% ammonium, and 71.4% TN from LWW. (Tombola *et al.*, 2019)

Aerobic degradation may resolve drawbacks of other LWW treatment methods. The River Die-Away (RDA) biodegradation test on shampoo surfactants reported mercantile shampoo products comprised of surfactants, which are resilient to biodegradation compared with biodegradable shampoo. (DeFerrer *et al.*, 1991) Varied modifications in LAS elimination efficacy through an assortment of industrial detergents have revealed that additives like softeners, antibacterial additives, bleaching products, and fragrance in detergents can limit the microbial efficiency for biodegradation. (Khandare & Govindwar, 2015)

It has been reported that anaerobic biodegradation of LWW varies analytically on the composition of LWW. Many studies used SBR, anaerobic fluidized bed reactor (AFBR) for LWW treatment, expanded granular sludge bed (EGSB), and up-flow anaerobic sludge blanket (UASB). (Andersen *et al.*, 2002; Delforno *et al.*, 2015; Faria *et al.*, 2019) Denitrifying microbes in anaerobic batch reaction were able to eliminate 99.9% LAS with 97.5 mg/L ethanol and 88 mg/L nitrates. (Andrade *et al.*, 2017) LAS elimination (84%) was observed in an EGSB reactor after the supplementation of Fe^{3+} ions. (Centurion *et al.*, 2018; Delforno *et al.*, 2014) The role of Fe^{3+} as terminal electron acceptor can produce energy and provide microbial growth. In contrast, the microbial activities accountable for LAS elimination were found to be inhibiting after supplementing the effluents with a sulfide concentration of more than 20 mg/L. (Delforno *et al.*, 2015) LAS mixture and other substances present in detergent can produce harmful volatile fatty acids in the effluent.

(Carosia *et al.*, 2014) The presence of xenobiotics as non-ionic surfactants was beneficial for the decay of LAS through the promotion of microbial growth. (Okada *et al.*, 2014) In the synthetic LWW, LAS decay observed was 45% whereas in the actual LWW, 82% decay of LAS was reported in a UASB reactor. (Okada *et al.*, 2014) Similarly, a study reported that no influence on LAS decay was found in anaerobic bioreactor if there were impurities like metals, organic acids, and xenobiotic compounds in sanitary sewage. (Okada *et al.*, 2014) Effluent intensity is a major reason for impacting the anaerobic biodegradation efficacy of LWW. (Okada *et al.*, 2014) In a different study, when LAS intensity in LWW was enhanced to 5-36 mg/L in a pilot-scale EGSB reactor (LWW co-digestion with municipal sewage), elimination of COD and LAS was found to be 44-61% and 32-71%, respectively. Literature also supports the anaerobic degradation efficacy in AFBR through different media. (Moura *et al.*, 2019) The elimination of LAE, organic matters, and LAS was found to be 98%, 40.4%-85.1%, and 24.7% 56%, respectively in AFBR. (Braga *et al.*, 2015) In the meantime, 99% elimination of LAS and 97% COD removal was obtained after operating AFBR with clay, glass beads, activated carbon, and sand with 18 hours of HRT. (Oliveira *et al.*, 2017)

The use of wetland to manage wastewater has been observed as noteworthy accomplishment as testified in the collected works. (Pérez *et al.*, 2019) Elimination of 77% LAS in LWW was established in an ideal artificial wetland operating at 15 days of HRT and comprising *Schoenoplectus americanus* (an aquatic flowering plant). The treated LWW was targeted for re-utilization in gardening. *S. americanus* was dipped fully in a 40 cm water depth with gravel, tezontle, and agave fiber as supports in the prototype synthetic wetland. (Pérez *et al.*, 2019)

During biological treatment there is a formation of foam in the aeration basin owing to the presence of surfactants in LWW. The stable foams were generated due to the interaction of gas bubbles with hydrophobic particles and surfactants. (Petrovski *et al.*, 2011) The hydrophobic particles come together at the water-air interface and reinforce foam stability. (Petrovski *et al.*, 2011) If the presence of surfactant causes excessive forming, antifoam agents (such as Zeta Airspel 300®, dimethyl siloxane (DMS), silicone defoamer, etc.) can be used to minimize foam formation in LWW. (Khan *et al.*, 2001; Kulkarni *et al.*, 2017;

Mangundu, 2017) Some studies also suggest that foaming can be controlled by reducing the sludge retention time (SRT) (Hug, 2006; Rossetti *et al.*, 2005).

2.8. Integrated Processes

Integrated physical and chemical processes

As mentioned above, a single technology cannot attain the regulatory limits in an economical and eco-friendly manner. The physicochemical procedures combined by other treatment processes for the effective management of LWW are depicted in Table 7. Combining coagulation with other treatment technologies can enhance treatment efficiency of LWW. (Matilainen *et al.*, 2010) In a study, a combination of bipolar electrocoagulation and electro flotation procedure has been reported to treat industrial LWW in an electrochemical apparatus. (Ge *et al.*, 2004) The charged hydroxyl-cationic complexes such as $Al_6(OH)_{15}^{3+}$ and $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$ and $Al_{13}O_4(OH)_{24}^{7+}$, were generated on the anodic side as a consequence of the hydrolysis and polymerization of Al^{3+}. The higher coagulation rate was promoted by the hydroxyl-cationic complexes, while the promotion of floc floatation was done by the by-products gases (H_2 and O_2). (Ge *et al.*, 2004) The same experiment was replicated at a pilot-scale reactor of 1.5 m^3/h treatment capacity with actual LWW from Harbin, China. The pilot-scale reactor resulted in 84.80% removal of methylene blue active substances (MBAS) and 95.84% removal of COD. In another study, the utilization of ultrasound along with electrocoagulation-and-electroflotation cell has been reported for LWW treatment. (Wang *et al.*, 2009) The contaminant level in treated LWW was brought down by the combined technique. (Wang *et al.*, 2009) Electrocoagulation/electrofloatation process successively operated at the laboratory and large scale for LWW treatment was studied by Dimoglo et al. (Dimoglo *et al.*, 2019). Color, surfactant and turbidity could be respectively removed up to 90% under the best operating conditions (current density = 5.26 mA/cm^2, pH = 5.5, and treatment time of 5 min). (Dimoglo *et al.*, 2019)

Combination of coagulation and chemical adsorption has several advantages. The combination of adsorption with electro-coagulation has been reported for treatment of industrial LWW. (Veli *et al.*, 2019) In this study, two different adsorbents obtained from organic waste (left-over hazelnut shell (HS)) were used: polyaniline (PANI/HS) and

polypyrrole (PPy/HS). PANI/HS (0.70 USD/g) resulted in COD elimination of 75% while PPy/HS (3.21 USD/g) resulted in COD removal of 20% from LWW. (Veli *et al.*, 2019)
Noutsopoulos et al. (Noutsopoulos *et al.*, 2018) combined three processes for LWW management: i) coagulation ii) sequential sand filtration, iii) adsorption using activated carbon, iii). (Noutsopoulos *et al.*, 2018) The combined technique resulted in 85% COD removal. The treated LWW met the regulatory standards in Greece for urban utilization, irrigation of crop fields and toilet flushing.

Combination of coagulation with membrane filtration has been reported for LWW treatment. (Hashim *et al.*, 2017) Such combination has two major benefits: enhanced elimination of natural organic matter (NOM) compounds and decrease in membrane clogging. (Haan *et al.*, 2018) Earlier, studies focused on membrane filtration for LWW treatment where, UF and reverse osmosis (RO)were widely used. A sequential UF followed by RO arrangement resulted in 98.9% COD removal, 99.2% BOD_5 removal and 75% mineral oil removal from LWW. (Kim *et al.*, 2015b) The membrane fouling in UF/RO can be reduced to 50% by combination of a pre-treatment method. (Kamarudin *et al.*, 2003; Kim *et al.*, 2014) Membrane clogging can also be reduced through hydraulic washing amid filtration rounds.

Effective LWW treatment can be obtained by combining coagulation along with another removal processes like flotation, membrane filtration, adsorption, and flocculation. Such combinations result in a robust, efficient and faster treatment process. (Costa *et al.*, 2018) However, the chemical treatment process often generates non-recyclable waste that increases the overall costs after treatment. As a result, exploration for new alternatives is focussed on sustainable, biodegradable, and non-chemical-based materials (organic substances). However, intensive research needs to be carried out with these materials for elimination of pollutants from LWW.

Integrated treatment approach using combination of biological and other techniques

The most common example of physical--biological treatment is membrane bioreactor (MBR). (Iorhemen *et al.*, 2016) There are several advantages of MBR: i) it can treat elevated organic loading though biological treatment ii) it has advantage of membrane separation process iii) ease of automation and iv) requirement of smaller space. (Huang *et* *al.*, 2000) The domestic-legislated release criteria were linked with the efficient treatment of industrial LWW in Poland that showed promising results on the MBR efficacy. (Bering *et al.*, 2018) The study testified removal of various pollutants in the following range: 95%–98% BOD_5, 89%–94% COD, 55%–71% TP, 32%–84% TN, 98.8%–99.4% non-ionic surfactants and 94.5%–99.5% anionic surfactants. In a different study, the authors presented a pilot-scale finding of side-stream MBR, operating for 4-month period. (Andersen *et al.*, 2002) The treated LWW from MBR was used for laundry washing cycles. (Andersen *et al.*, 2002) The treated LWW from MBR do not require further treatment technology like RO.

An amalgamation of phytoremediation with physicochemical treatment process for LWW treatment was recommended. (Siswoyo *et al.*, 2019) Expending plants *Kiapu* (*Pistia stratiotes*) and water hyacinth (*Eichhornia crassipes*) were used to simplify phytoremediation, followed by adsorption to deal with LWW. It was reported that when adsorption functioning reached 50%, the phytoremediation process removed 54.3% phosphate, 77.5% COD, and 99.9% surfactants. Apart from adsorption, centrifugation also has been recommended to remove SS in LWW that may not be eliminated through the phytoremediation process. The authors reported that centrifugation of LWW, enhanced the removal of pollutants by microalgal phytoremediation *(Chlorella variabilis).* During the combined treatment, removal of COD, BOD_5, TP, and TN were observed to be 92%, 91%, 85% and 97%, respectively.

Addition of pre-treatment steps (e.g., flotation, membrane filtration, flocculation, adsorption) is the most common method to selectively eliminate pollutants harmful to the MBR unit.

Table 2.7 Integrated processes for LWW treatment

	Treatment Process	Feed	Operating Conditions	Removal Efficiency	References
Integrated Physio-chemical Processes	EC–EF	ILWW COD = 785-1090 mg/L MBAS concentration= 64.5-72.3 mg/L Turbidity = 471-583 NTU	Electrodes = Ti, Al Current intensity = 0.8-2 A Reactor volume = 2800 cm^3 HRT = 5-30 min S.A. = 50 cm^2	COD = 95.82-95.87% MBAS = 84.51-85.12% Turbidity = 97.89-98.25%	Ge *et al.*, 2004)
	Coagulation, sand adsorption, and membrane filtration	ILWW Surfactant concentration = 10.06 mg/L TN dose = 2.75 mg/L BOD = 195 mg/L NH3-N dose = 2.45 mg/L COD = 280 mg/L TP dose = 9.92 mg/L SS concentration = 35 mg/L	Coagulants = $Al_2(SO4)_3.18H_2O$ Adsorbent = Silicic Diameter = 3.2 cm Height = 1 m Velocity = 10 m/h Sand grain size = 0.5-2 mm Treatment time = 6 min Membrane filtration Type = UF and RO S.A. = 0.13 m^2 (UF) = 1.5 m^2 (RO) Pore size = 20-400 kDa (UF) Pressure = 4-5 bar (UF) = 20-30 bar (RO) Flow velocity = 15-20 m/s (UF)	TP = 98.59% SS = 75% Surfactant = 90.95% TN = 98.91% COD = 98.93% NH3-N = 98.78% BOD = 99.23%	(Šostar-Turk *et al.*, 2005)
	EC/EF	Simulated LWW COD = 226 mg/L	Electrode = Al S.A. = 72 cm^2 Ultrasound frequency = 43 kHz Voltage = 1-7 V Operating pH = 2.5-9.5	COD = 62%	(Wang *et al.*, 2009)
	Coagulation, membrane Filtration	ILWW COD = 1,138 mg/L	Coagulation Mixing time = 2-20 min Coagulant dose= 40-250% Membrane Type = Micro Filtration (dead-end, recirculated flow)	COD = 74-82%	(Kim *et al.*, 2014)

Coagulation, membrane Filtration	ILWW Turbidity = 110-735 NTU TP = 23-23.5 mg/L COD = 414-1196 mg/L	Coagulation Operating Temperature = 40 ℃ Coagulant dose= 0-750 μl/l Membrane Type = MF Flow rate = 3.36 L/h S.A. = 11.3 cm2	Turbidity = 100% TP = 20-40% COD = 55-65%	(Shang *et al.*, 2015)
Coagulation/ flocculation/ sedimentation, membrane filtration	ILWW Turbidity = 64 NTU TOC = 86.1 mg C/L TDS = 473 mg/L Color = 425 mg/L TS = 532 mg/L COD = 245 mg O_2/L Surfactant = 15.9 mg/L TN = 4.8 mg TN/L	Coagulation/ flocculation/ sedimentation pH range= 4.6-8.0 Cogulant dose = 60-180 mg/L Agitation speed = 20-120 rpm Membrane filtration Flow Rate= 30 L/h (MF), 48 L/h (UF) S.A. = 0.027 m^2	Turbidity = 99.1% TOC = 56.3% TDS = 55.6% Color = 98.4% TS = 100% COD = 68.6% Surfactant = 71.7% TN = 27.1%	(Nascimento *et al.*, 2019)
EC/EF	ILWW MBAS = 77 mg/L COD = 1,237 mg/L Turbidity = 626 NTU	Electrode material = aluminium S.A. = 350 cm^2 Reactor Volume = 1.5 dm^3 Electroflotation Reactor Volume = 3 dm^3 Electrodes = graphite, steel	MBAS = 30-40% COD = 70% Turbidity = 60%	(Dimoglo *et al.*, 2019)
Coagulation/ flocculation/ sedimentation-adsorption - membrane filtration	ILWW Turbidity = 75 NTU Color = 1,185 mg/L Surfactant = 29.5 mg/L COD = 305 mg/L	Coagulation/ flocculation/ Sedimentation Type = Tanfloc POP Concentration = 110-150 mg/L Agitation Speed = 20-120 rpm Adsorption Material = AC S.A. = 900 m2/g Weight = 0.1 g Membrane Type = MF Pore size = 0.4 μm F.A. = 0.027 m^2	Turbidity = 99.4% Color = 99.9% Surfactant = 92.9% COD = 80%	(Huang *et al.*, 2019)

			Membrane Permeability = 210.89 L/h m^2 bar Applied Pressure = 1.4 bar Flow rate = 30 L/h		
	Membrane Bioreactor	ILWW Conductivity = 1.8 mS/cm Fe = 210 μg/L Non-ionic surfactant = 1 mg/L	Reactor Volume = 3.5-4.0 m^3 Membrane type = UF S.A. = 0.95 m^2 Pore size = 500 kDa	Conductivity = -5.9% IFe = 96.1% Non-ionic surfactant= 99.3%	(Andersen *et al.*, 2002)
	Coarse screening, membrane Bioreactor	Commercial LWW Conductivity =1,000-3,000μS/cm COD = < 100 mg/L	Membrane type = MF Capacity = 0.72 L/min. S.A. = 360 m^2	Conductivity = 98% COD = 95%	(Buchheister *et al.*, 2006)
Integrated Physio-chemical and Biological Processes	Membrane Bioreactor, membrane filtration	ILWW COD = 1,050 mg/L Conductivity = 1,900 μS/cm TN = 40 mg/L TP = 5 mg/L	Membrane Bioreactor Flux = 14 L/m^2 h Applied Pressure = 0.11 bar Air flow rate = 42 m^3/h HRT = 20-30 h Membrane Type = RO Flux = 25 L/m^2 h (13 bar)	COD = 95.71% Conductivity = 98.68% TN = 98.75% TP = 84%	(Hoinkis *et al.*, 2012)
	Screening, Membrane Bioreactor	ILWW TP = 16.8 g/L Turbidity = 92 NTU TN = 5.5 g/L Conductivity = 2,249 μS/cm TS = 15 g/L COD = 317 mg/L	Pore size = 0.4 μm Air flow rate = 48 m^3/h Feed Flow rate = 0.5 m^3/m^2 d Applied Pressure = 1-2 kPa	TP = 57.14% Turbidity = 99.57% TN = 56.36% Conductivity = 0.40% TS = 98.67% COD = 74.76%	(Nicolaidis & Vyrides, 2014)
	Membrane Bioreactor	ILWW Non-ionic detergent = 14.6-63.9 mg/L BOD = 368-626 mg/L Anionic detergent = 22.6-50.8 mg/L COD = 750-1,150 mg/L	Reactor Volume = 20 L (3 units) Flow rate = 13-25 L/h S.A. = 0.93m^2 Pore size = 0.04 μm Applied Pressure = -0.05 bar MLSS concentration= 7-11 kg/m^3	Non-ionic detergent = 98.8-99.4% BOD = 95-98% Anionic detergent = 94.5-99.5% COD = 89-94% TP = 55-71% TN = 32-84%	(Bering *et al.*, 2018)

	TP = 4.2-7.47 mg/L TN = 8.75-20 mg/L			
Adsorption, phytoremediation	Household LWW COD = 365-975 mg/L Surfactant = 9-27 mg/L Phosphate = 1-2 mg/L	Adsorbent = Municipal sludge Concentration = 1-400 mg Treatment time = 0-120 min Phytoremediation Species = Kiapu (*Pistia stratiotes*), water hyacinth (*Eichhornia* crassipes)	COD = 77.5% Surfactant = 99.9% Phosphate = 54.3%	(Siswoyo *et al.*, 2019)

2.9. Economic Analysis of LWW treatment

Conventional approaches for treating laundry wastewater comprise of innumerable arrangements of chemical, physical, and biological methods. Concerning high operational expenses of above processes, it is necessary to explore cost-effective and efficient techniques. (Lalit R. Kumar, 2022) Low energy consumption, minimum chemical expenditure and reduced installation space should be the criteria for these cost-effective techniques. Following are the factors that need to be considered for economics of treatment processes:

Power consumption

Power utilization of current wastewater treatment plants varies between 20 and 45 kWh population equivalent per year. Smaller plant has lower power expenditure. Generally, cost of power consumption is 50% of capital cost of the treatment plants. (Kowalska, 2008) Authors reported that the energy consumption for NF is higher (45.5 kWh/m^3) as compared to EC (1.25kWh/m^3) and thermophilic aerobic membrane reactor (TAMR) (10 kWh/m^3). The automatic laundry washing services bring about a huge difference in power consumption cost of treatment plant. For example, power consumption cost of treatment plant is dependent on the variation in the number of annual laundry wash cycles, influent temperature set, and type of the washing machine. The common washing practices vary from laundry to laundry, which impact the influent quality and electricity consumption cost. Japanese families, for instance, require supplementary water for washing compared to other countries. European households use lower amount of water due to horizontal axis washing machine, leading to more contaminated LWW. This may perhaps be a paradigm additionally for further countries, like Japan and South Korea, where extreme concentrations of pre-treatments and post-treatments of laundry are recounted, which depart various of cleaning processes from the washer. This directs to the query of washing implementation that is accomplished by the washer and uncertainty it is adequate for user requirements.

Higher concentrations of washing treatments exterior to the washer indicate a depleted concentration of washing implementation endowed by the machines. These supplementary practices consume furthermore pertinent expanses of water and energy, which are not contemplated in the inspection. (Sandrolini & Franzoni, 2001) In several studies, it has been reported that energy consumption largely depends upon the type of treatment technology used. (Guilbaud *et al.*, 2012; Nicolaidis & Vyrides, 2014; Šostar-Turk *et al.*, 2005) Figure 3 presents the cost breakdown of different treatment processes used for the LWW treatment. The processes like NF and NF combined with RO has high-energy consumption cost contribution (9% and 30% respectively), while processes like adsorption using GAC and submerged MBR has lower energy consumption cost contribution (4% and 4.5% respectively). (Guilbaud *et al.*, 2012; Nicolaidis & Vyrides, 2014; Šostar-Turk *et al.*, 2005)

Capital cost of the treatment system

A study reported that capital cost of LWW treatment plant has increased in the previous 15 years due to increase in wastewater recycling rates. Studies shows that the capital cost is one of the main and largest cost associated with the LWW treatment process. Researchers estimated that the capital cost for adsorption, NF, NF combined with RO and submerged MBR are 21%, 47%, 27% and 95% respectively. (Guilbaud *et al.*, 2012; Nicolaidis & Vyrides, 2014; Šostar-Turk *et al.*, 2005) From the above data it can easily concluded that the capital cost of submerged MBR is high and it requires 95% of the total cost of the LWW treatment system. (Nicolaidis & Vyrides, 2014) In another study Jamil et.al. (Ahmad & Hisham, 2008) designed a LWW treatment system by using sand and gravel filter. The treatment unit was able to treat 1.8 m^3 of LWW on the daily basis. The capital & electricity cost for the system was estimated to be 1336 US $ and 6.75 US $/year respectively. (Ahmad & Hisham, 2008)

Operation and maintenance cost (O&M)

A study implied that data for operation and maintenance costs incorporate costs of substances (chemical procurement for operation and maintenance), workforce, and electricity for treatment, but it does not include water circulation costs. WHO reported O&M cost of $807 million annually in 2007. (Organization, 2003) The highest contributor of this expenditure was workforce expenses ($302 million), while material and energy charges contributed $198 million and $199 million, respectively. Additional supplementary expenses were estimated to be $108 million. (Organization, 2003) The O&M cost linked to the procurement and treatment will be determined by type of treatment technology, volume of LWW, and type of water handled. (Organization, 2003) In figure 4, the cost breakdown of LWW treatment process shows that the maintenance cost is one of the major cost contributors. The treatment process like submerged MBR and NF combined with RO requires lower operating cost (0.5% and 23% of total cost), while NF & adsorption requires higher operating (64% and 74.5% of total cost) for the operation. (Guilbaud *et al.*, 2012; Nicolaidis & Vyrides, 2014; Šostar-Turk *et al.*, 2005)

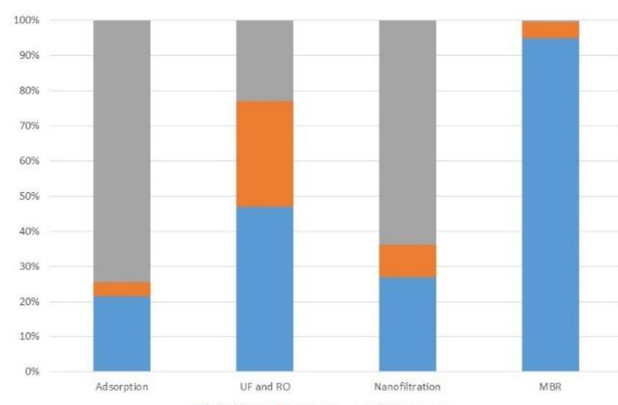

Figure 2.4: Cost breakdown of different LWW treatment processes

Table 8 presents the total cost associated with the different LWW treatment process. Highest treatment cost was reported for NF (5 €/m^3) while lowest treatment cost was reported for sand filtration (0.04 €/ m^3). (Guilbaud *et al.*, 2012; Nicolaidis & Vyrides, 2014; Šostar-Turk *et al.*, 2005)

Table 2.8 Cost of laundry wastewater treatment

Treatment method	Cost (€/ m^3)	Reference
Chemical Coagulation	0.42	(Ciabattia *et al.*, 2009)
Adsorption	1.15	(Collivignarelli *et al.*, 2019)
Sand filtration	0.04	(Ciabattia *et al.*, 2009)
UF	0.16	(Ciabattia *et al.*, 2009)
NF	5	(Collivignarelli *et al.*, 2019)
Membrane bio-reactor	0.35	(Nicolaidis & Vyrides, 2014)
Coagulation, Sand filtration, Ozonation, Adsorption and UF	0.81	(Collivignarelli *et al.*, 2019)
UF and RO	1.35	(Šostar-Turk *et al.*, 2005)
Sand filtration and Ozonation	1.44	(Collivignarelli *et al.*, 2019)
Coagulation and Adsorption	0.51	(Šostar-Turk *et al.*, 2005)

2.10. LWW Reuse: role in circular economy

Over the last years, the main objective of wastewater treatment plants (WWTPs) is treatment of wastewater and sewage sludge. However, there is an enhanced demand of the recovery of nutrients and recyclable water from wastewater. It allows to modify a WWTP into a sustainable technological. A study reported that linear economy model creates an annual economic burden of 7.2 trillion euros in Europe. (Within, 2015) The replacement of the linear economic model by a circular economy model has several advantages: enhanced resource productivity, reduction in waste and decrease in dependency on natural resources. As a result, circular economy model can generate a direct profit of up to 0.6 trillion euros/year and an indirect profit of up to 1.2 trillion euros by 2030. This will lead to 25% cost reduction when compared to the linear economy model. (Within, 2015)

LWW contains huge amounts of useful resources which can contribute to circular economy. (Sfez *et al.*, 2019) Amongst them, treated wastewater (reusable for several applications) and wastewater sludge (a valuable source of nutrients). Figure 5 shows reuse of treated LWW in context of circular economy. Hence, recovering resources from LWW can contribute to the concept of circular economy. (Molina-Moreno *et al.*, 2017; Verstraete & Vlaeminck, 2011) There is considerable nutrient content in wastewater sludge: 50-55% carbon, 10%-15% N and 1%- 3% P, 0.5%-1.5% S. (Tyagi & Lo, 2013) As a result, sludge can be used as a resource for many products like fertilizer, animal feed etc. This will also counter a serious environmental impact by reducing dependency on non-renewable resources. (Fijalkowski *et al.*, 2017),(Tyagi & Lo, 2013)

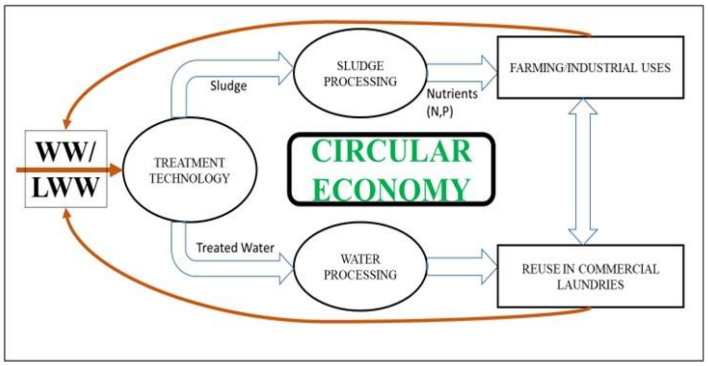

Figure 2.5: Circular Economy in Laundry wastewater treatment

Apart from recovering nutrients from treated wastewater streams, water reuse is also necessary due to high water requirement for industrial and agricultural activities, tacking climate change, urbanisation and population growth. (Goswami *et al.*, 2018) As a result, treated water can reduce the freshwater demand for industrial and agricultural uses. (Division *et al.*, 2004)

Under the "European Green Deal", European Union (EU) proposed an action plan towards zero pollution. The target to achieve zero pollution ambition has been set by the year 2050. (COMMISSION, 2021) The goal of the plan is to decrease pollution up to a level that will no longer be considered dangerous to the ecosystem and health. (COMMISSION, 2021) Under this action plan, they set a target to reduce significantly total waste generation and by 50% residual municipal waste generation by 2030. (COMMISSION, 2021) The zero-pollution hierarchy follows reversing the pyramid of action in which it has been proposed to prevent the pollution at the initial level, minimize and control it at the next level and, at the bottom, eliminate and remediate it. Under Urban Wastewater Treatment Directive, the EU plans to recover nutrients from the wastewater and reuse the treated wastewater for other purposes. (COMMISSION, 2021) The deal also addresses emerging contaminants like microplastics and micro-pollutants like pharmaceuticals. The plan also aims to improve water quality by reducing microplastic release (by 30%) in the environment. (COMMISSION, 2021)

2.11. Challenges and Future perspectives

Chemical coagulation is one of conventional methods for LWW treatment. However, high cost of chemicals, adverse environmental impact and handling large volume of sludge are challenges associated with chemical coagulation. Bio-flocculants can act as alternative to chemical flocculants. Extensive research needs to be done to produce cost-effective flocculants.

Electro-coagulation is a replacement for chemical coagulation and has several benefits: easy & cost-effective process, environment-friendly, flexible, competent, and broad-spectrum (being capable to eliminate concurrently various kinds of contaminants). However, cathode passivation and electrode degradation may happen that diminishes the efficiency of electrocoagulation. Besides, additional research works need to be performed to accomplish high performance in electrocoagulation through statistical modeling and computational fluid dynamics. To tackle the challenge of passivation and short operation time, the suitable electrode material can be investigated, which can provide better treatment efficiency and less passivation. Another strategy could be the addition of continuous coagulant dose while maintaining the cell potential low during extended EC operations. Using alternative current (AC) supply, polarity reversal, ultra-sonification, and increasing the flow velocity to increase the scouring could be another solution. Membrane filtration is commonly used for LWW treatment. The major shortfall in membrane filtration is membrane fouling, which occurs when the particles are accumulated on the surface and/or within the pores of the membrane, which demands repeated washing to re-establish the permeate flux and utilizes more energy at high pressures. Membrane filtration is unable to remove surfactants from LWW as per the desired limits. To resolve above problems, solutions like hydrodynamic operation (controlling turbulence flow), pre-treatment techniques, and membrane surface modifications can be employed to treat LWW as per desired limits. In the combined treatment systems, a pre-filtration technique (by means of sand filtration or a bag filter) followed by membrane filtration using ceramic membranes could be the basis of a process able to reduce clogging issues during LWW treatment.

Adsorption for LWW management is considered as an efficient and cost-effective technique. However, high cost of commercial adsorbents and high regeneration cost (due to high temperature, high volume of solvents) are major disadvantages of this technology. Low-cost adsorbents need to be produced from agricultural wastes and tested at laboratory scale. Effective regeneration of adsorbents can be one of the solutions to reduce the overall cost of the adsorption process and minimize the use of costly adsorbents.

Combined remediation strategies like biological, physicochemical, or integrated processes can be used to treat LWW treat as per the required limits. MBR is a combined method, capable of treating LWW with high removal of pollutants. However, it has high capital cost and energy expenditure. Also, due to membrane clogging in MBR, there is high operational cost due to repetitive washing and replacement of membranes. Additional pre-treatment steps (e.g., flotation, membrane filtration, adsorption, and/or flocculation) are the most common procedure to selectively eliminate pollutants harmful for the MBR unit. However, this will enhance the overall operating cost and affect environmental footprint of the treatment process. Also, developing and underdeveloped countries may not have required infrastructure or resources to install this type of technology. Intensive research is required to develop LWW treatment processes, which are cost-effective, ecological, and acceptable to the community and can meet the permissible limits for water reuse.

2.12. Conclusion

Due to rapid urbanization, large amount of untreated LWW effluents is discharged into sewage lines, which imparts load on wastewater treatment plants. Surfactants (like LAS), nitrate, COD, BOD, phosphate, SS, turbidity, and sulphates are the chief impurities in LWW. Besides, additional tiny aggregates of volatile acids, alkalinity, fabric, deodorizing agents, food, non-biodegradables, body fluids, a limited concentration of metals (Zn, Cd, Cu, Fe) and different xenobiotic organic compounds are present in LWW. Proper management of LWW can create a concept of circular economy, resulting in reclaimed water for many useful purposes, such as irrigation, construction, car washing, toilet flushing etc. Based on literature review it seems that a single treatment process is not sufficient to treat LWW up to acceptable reuse standards. To that end, different treatment chains have been proposed: i) hybrid processes combining membrane filtration with adsorption; ii) advanced oxidation process using ultraviolet (UV) and hydrogen peroxide (H_2O_2); iii) coagulation-flocculation combined with sand filtration; iv) combination of ozonation process, adsorption, and ultrafiltration (UF) techniques. By use of appropriate treatment technology, the freshwater supply dependency can be reduced, leading to the minimization of freshwater consumption cost.

Conflict of Interest: The authors declare that they have no conflict of interest.

Data availability statement: Data sharing not applicable – no new data generated.

3. PROJECT CONTEXT AND PREVIOUS STUDY

3.1. Basis of this study

The necessity to treat laundry wastewater sustainably is driven by the global scarcity of freshwater resources, as depicted in Figure 3.1. To enable reuse, treated water must satisfy specified criteria outlined in Table 3.1. Effectively addressing this challenge requires comprehensive removal of organic constituents, colloidal particles, contaminants, and surfactants to comply with water reuse regulations.

This study focuses on employing EPS for the treatment of commercial LWW using the following processes: (a) EPS alone, (b)PBEC with synthetic polymer/ EPS, and (c) UF with EPS. These integrated approaches aim to efficiently eliminate organic matter, colloidal particulates, and various impurities present in the CLWW. Notably, the subsequent phase targeting surfactant removal is part of an independent research initiative led by Mahdieh in 2024. This subsequent research explores the utilization of Advanced Oxidation (AO) and Electrochemical Oxidation (EO) techniques specifically for surfactant removal.

By dividing the project into distinct phases, this research thesis establishes a comprehensive framework for addressing the multifaceted challenge of treating commercial laundry wastewater for sustainable reuse. The study focuses the use of EPS, UF, and EC to address organic content and particulate impurities, while the subsequent phase aims to optimize methods for surfactant removal through advanced oxidation techniques—an essential approach for meeting water reuse standards, as undertaken by Mahdieh et al. in 2024, after project aims to optimize methods for surfactant removal through advanced oxidation techniques—an approach vital for meeting water reuse standards (in the washing cycle of laundry) as being done by Mahdieh, et al., 2024.

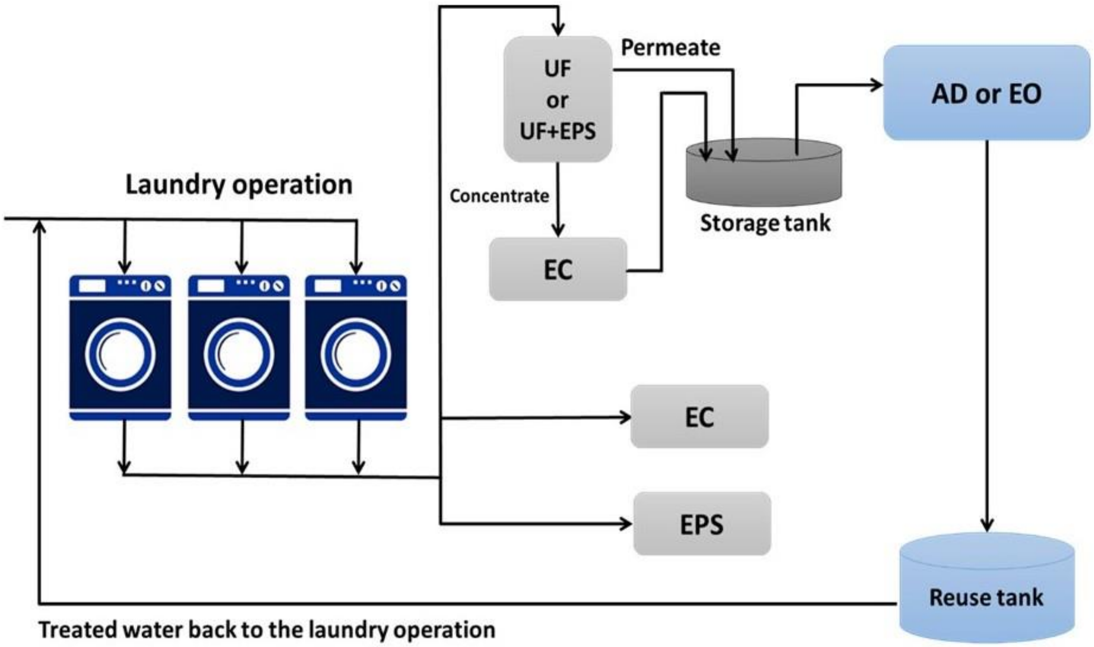

Figure 3.1 Project overview for the treatmet of LWW

Table 3.1 LWW rejection and reuse values

Parameters	Measured Values for LWW in this thesis	Rejection standard	Water reusing standard
Turbidity (NTU)	110 ± 5	n.r.	≤ 2
pH	10.4 ± 0.5	5.5-9.5	5-9
Conductivity (µS/cm)	494 ± 20	n.r.	≤1500
Suspended solids (mg/L)	60 ± 10	≤ 80	10-50
Total solids (mg/L)	622 ± 20	<100	n.r.
COD (mg/L)	579 ± 30	<150	<60
NPEOs (µg/L)	570 ± 150	<200	n.r.
Sulphate (mg/L)	22± 5	n.r.	n.r.
Sulphite (mg/L)	0.2 ± 0.05	n.r.	n.r.
Nitrate (mg/L)	4 ± 1	n.r.	n.r.
Nitrite (mg/L)	1.5 ± 0.5	n.r.	n.r.
Ammoniacal nitrogen (mg/L)	2.3 ± 1	n.r.	n.r.
Total nitrogen (mg/L)	9.7 ± 0.5	n.r.	n.r.
Phosphate (mg/L)	21 ± 2.0	n.r.	n.r.
Aluminium (mg/L)	0.096 ± 0.02	n.r.	n.r.
Calcium (mg/L)	18.6 ± 2.0	n.r.	n.r.
Magnesium (mg/L)	3.87 ± 0.5	n.r.	n.r.

Sodium (mg/L)	104 ± 10	10kg/day	n.r.
Potassium (mg/L)	3.76 ± 0.5	n.r.	n.r.
Sulphur (mg/L)	20.6 ± 2.0	n.r.	n.r.
Zinc (mg/L)	0.42 ± 0.15	n.r.	n.r.
Copper (mg/L)	0.03 ± 0.01	n.r.	0.01
Iron (mg/L)	0.23 ± 0.2	n.r.	0.1
Manganese (mg/L)	0.019 ± 0.005	n.r.	0.05
Lead (mg/L)	<0.003	n.r.	n.r.
References	This thesis	(Corona *et al.*, 2021; Li *et al.*, 2022a; Manouchehri & Kargari, 2017; Omolara, 2017)	(Benis *et al.*, 2021; BHAGAT, 2018; Faria *et al.*, 2019; Moura *et al.*, 2019; Nascimento *et al.*, 2019)

n.r.: Not reported

3.2. Problems

3.2.1. General Problems

Hydrological Impact

The world is grappling with a stark reality: the scarcity of freshwater bodies is a pressing concern. Only 2.5% of the Earth's water being freshwater, and a mere 0.3% accessible in lakes, rivers, and swamps. A striking 4 billion people already face severe water scarcity for at least one month a year. By 2025, nearly two-thirds of the global population could be living in water-stressed conditions. Astonishingly, 80% of the water we use ends up as wastewater, laden with untapped potential. Recycling and treating wastewater for agricultural, industrial, and even potable purposes can alleviate the strain on freshwater sources. For processing 1kg of cloths approximately 15 litres of fresh water is required (Mauchauffee *et al.*, 2012). In commercial laundries huge number of clothes are washed daily, which ultimately demand tremendous amount of fresh water. To minimise the freshwater demand, it is necessary to reuse treated wastewater in the laundry systems.

Laundry Wastewater Discharge: Impacts on Sewer Networks and Treatment Plant Efficiency

LWW has quite different characteristics than normal municipal wastewater. It is highly rich in SS, COD, surfactants like NPEO and heavy metals. When laundry wastewater disposed to the municipal sewer lines without prior treatment then it contributes to the increase in the concentration of SS, COD, heavy metals and surfactants in municipal wastewater. In municipal wastewater, the contribution from detergents of heavy metals are 31.9% for Cd, 0.24% Cu and 0.30% for Zn (Aonghusa & Gray, 2002). This has important implications for sewage sludge disposal. The presence of NPEO in the wastewater treatment system may affect the microbial growth or it can reduce the efficiency of the treatment (Weng *et al.*, 2010).

Limitations of using single conventional treatment method for water reuse initiatives

Water scarcity is severe in developing countries; thus, the use of inexpensive wastewater treatment strategies can help minimize this issue. An abundant amount of laundry wastewater (LWW) is generated daily and various wastewater treatment research have been performed to achieve suitable techniques but, the treatment of laundry wastewater is quite complex as any single process is not quite efficient and do not fulfill the reuse standard. Due to the presence of suspended particles and surfactants (refractory compounds), it is quite challenging to treat laundry wastewater by the conventional treatment process. At the same time, it is quite challenging to remove emerging, contaminate like NPEO from the wastewater by conventional treatment processes.

The study shows that the conventional treatment process like primary treatment can remove suspended particles effectively but difficult to remove surfactants (Only 13 % NPEO can be removed) (Xu *et al.*, 2014). Chemical coagulation is highly effective in removing suspended solids, organic matter, and contaminants, providing reliable treatment outcomes. Moreover, it requires minimal space, making it suitable for compact treatment systems. However, this process utilizes chemical coagulants leading to potential environmental impacts (Sun *et al.*, 2022). Electrocoagulation offers effective removal of various contaminants including heavy metals, organic pollutants and suspended solids. However, its disadvantages include electrode passivation, which reduces treatment efficiency and necessitates periodic cleaning or activation to maintain performance. Additionally, electrode replacement is frequently required (Samadikun *et al.*, 2021). At the same time ultrafiltration can effectively remove suspended particles and microorganism but it can not achieve high removal efficiency of COD, metals and detergents (Huisman *et al.*, 1997).

Lack of Research and Knowledge on Integrated Treatment Methods for LWW treatment

The current state of research on integrated treatment methods for LWW is lacking, indicating a critical gap in our understanding of this vital area. Despite the growing emphasis on sustainable wastewater management, there is a notable scarcity of studies focused on integrated approaches specifically tailored to the unique challenges posed by LWW.

This gap in knowledge limits our ability to optimize treatment processes for efficiency and environmental impact, particularly given the diverse and dynamic nature of contaminants present in LWW. The absence of research on integrated treatment methods hinders the development of effective strategies for LWW treatment plants, leaving the industry without evidence-based guidelines.

To address this shortfall, future research should prioritize investigating the performance and feasibility of integrated treatment methods for LWW. Establishing a solid knowledge base through rigorous scientific inquiry is crucial for advancing sustainable practices in wastewater treatment and ensuring the preservation of water quality and environmental health.

Sustainability challenges for LWW treatment

Sustainability challenges in current laundry wastewater treatment practices are increasingly evident, reflecting the need for a paradigm shift in the industry. Conventional treatment methods, such as activated sludge processes, often struggle to address the complex composition of laundry effluents, leading to incomplete removal of contaminants (Smith & McCullers, 2014). Furthermore, the environmental impact of energy-intensive treatment processes raises concerns about the overall sustainability of current practices. Some researchers emphasize the need for more energy-efficient alternatives to mitigate the carbon footprint associated with wastewater treatment plants (Padmanabhan *et al.*, 2017; Teixidó *et al.*, 2011; Valentino *et al.*, 2017). Additionally, the reliance on chemical-intensive treatments poses a threat to ecosystem health. The persistence of synthetic detergents and surfactants accentuates the ecological implications of current laundry wastewater treatment practices (Pasciucco *et al.*, 2022).

3.2.2. Specific Problems

Excessive chemical coagulant usage and toxic by-product generation

In LWW treatment, excessive chemical coagulant is excessive chemical coagulant usage can lead to increased operational costs and environmental impact. Laundry effluents contain various organic and inorganic contaminants, requiring effective coagulation-flocculation processes for treatment (Wang & Zhou, 2018). However, overdosing coagulants may not only be economically inefficient but can also result in residual chemicals in the treated water, posing environmental concerns.

Frequent replacement of anodes and passivation of electrodes during EC process

During the EC process, degradation of anodes and frequent replacement is a challenge. The presence of pollutants in the LWW require in-situ generation of coagulants which is generated by oxidation of the anode which causes the degradation of electrode. This degradation generates the need for replacing and changing the electrodes frequently which leads to short operation cycles. The major challenge of the EC process lies in controlling the cause of passivation, the oxide layer formation on the surface of the electrodes.

Membrane clogging during UF process.

Membrane fouling due to the presence of colloidal particles, oils, and surfactants in laundry effluents can lead to reduced filtration efficiency and increased energy consumption. It also requires frequent backwashing of the membrane.

3.3. Hypothesis

Eco-friendly Approach: Bacterial EPS for sustainable coagulation and flocculation

The coagulation and flocculation process has been considered as an easy, cost-effective, and efficient method for removing particulate matter, clay, colloids, humic acid, fulvic acid, phosphorus, and heavy metals from wastewater. Despite the economic benefits, the use of chemical coagulants poses serious environmental and health issues; therefore, there has been growing interest in the use of eco-friendly and sustainable biopolymers in order to reduce the risks posed by chemical coagulants and to decrease their doses (Guo *et al.*, 2017). Among biopolymers, bacterial extracellular polymeric substances (EPS) have been explored as a flocculating agent for sludge settling and dewatering and could be used for the treatment of laundries wastewater and the removal of heavy metals and surfactants.

Enhanced Electrode Performance and Prolonged Operational Lifespan through Sacrificial Iron Electrode Packing

Potential of electrocoagulation process could be utilized for organic compound removal; at the same time the charge neutralization can help to remove colloidal and suspended particles from the wastewater (Li *et al.*, 2022a). In the EC process, the destabilization mechanism of the contaminants, particle suspension, and breaking of emulsions can be summarized as three successive steps. (1) Compression of the diffuse double layer around the charged species by the interactions of ions generated by oxidation of the sacrificial anode (Oktiawan *et al.*, 2021). (2) Charge neutralization of the ionic species present in wastewater by counter ions produced by the electrochemical dissolution of the sacrificial anode. These counter ions reduce the electrostatic inter-particle repulsion to the extent that the van der Waals attraction predominates, thus causing coagulation. A zero net charge results in the process (Verma, 2017). (3) Flocs formation: the flocs formed as a result of coagulation creates a sludge blanket that entraps and bridges colloidal particles that are still remaining in the aqueous medium (Nidheesh *et al.*, 2020).

However, EC exhibits some drawbacks too that include elevated energy consumption, sludge generation, inefficiency for certain contaminants, and susceptibility to electrode passivation (Elazzouzi *et al.*, 2018; Nidheesh *et al.*, 2020; Yasri *et al.*, 2020). Therefore, there is a need of advanced processes such as packed bed electrocoagulation (PBEC) that may offer enhanced contaminant removal, reduced energy demands, diminished sludge formation, and prolonged electrode durability (Holt *et al.*, 2005; Yasri *et al.*, 2020). By increasing the surface area through the packing of sacrificial iron electrodes, passivation can be significantly reduced, leading to longer intervals between electrode regeneration cycles. This methodology may also enable prolonged operation of the initial electrode setup, facilitating the treatment of large volumes of water.

Use of EPS to enhance the performance of Ultrafiltration

Published literature (Kabsch-Korbutowicz *et al.*, 2006; Shon *et al.*, 2004) suggests that ultrafiltration has a huge potential in removing suspended particles and provides an essentially complete barrier against particles larger than the pore size, bacteria and the much smaller viruses usually found in the feed water. Any particles greater than the pore size are rejected. This characteristic makes UF membranes ideal for meeting absolute filtration quality requirements largely independent of the feed water quality provided the initial integrity of the membrane remains intact (Benis *et al.*, 2021). UF could allow removing colloids/particles compounds that could interfere with the subsequent process, such as adsorption or catalytic degradation used for NPEO removal (Kornaros & Lyberatos, 2006). Likewise, the pre-treatment of LWW could allow concentrating colloidal pollutants in a small volume, followed by the treatment of these pollutants using electrochemical coagulation. Such an approach can allow increasing faradaic yield during electrochemical treatment (Mollah *et al.*, 2001)**.**

Tangential flow/ cross flow (circulation of the fluid parallelly to the surface of the membrane) can be used to control the clogging of the membrane (Watabe *et al.*, 2016). The incorporation of EPS in LWW mitigates membrane clogging during cross flow filtration, facilitating floc formation, and preventing particle penetration into membrane pores. The polymer-based extracellular polymeric substances further enhance membrane hydrophilicity (Mauchauffee *et al.*, 2012).

The incorporation of EPS in LWW mitigates membrane clogging during cross flow filtration, facilitating floc formation, and preventing particle penetration into membrane pores. The polymer-based extracellular polymeric substances further enhance membrane hydrophilicity (Mauchauffee *et al.*, 2012).

3.4. Objectives

Global Objective: Development of hybrid process for the decontamination and reuse of wastewater from commercial laundries using EPS, electrolytic and membrane treatment technology.

Specific Objectives:

- Applying use of EPS for treatment of commercial LWW using different combination of chemical coagulants.
- To systematically investigate the treatment efficacy of laundry wastewater through PBEC and EPS, aiming to assess the synergistic impact of these methods on pollutant removal, colloidal stabilization, and overall water quality improvement.
- To enhance the performance of UF membrane technology using EPS for the treatment of LWW on a pilot scale, with a specific focus on regulating membrane clogging issues. Additionally, to assess the feasibility of treating the concentrated stream through PBEC.

3.5. Organization of the thesis

The thesis is organized into several chapters, each contributing to a comprehensive understanding of the research topic. The first chapter serves as an introduction, providing an overview of the research. It includes an introduction to the topic and a synthesis of existing literature related to laundry wastewater (LWW) treatment, covering various aspects such as treatment methods, economic considerations, and the role of LWW reuse in circular economy models. This chapter also addresses the research problem, hypotheses, objectives, methodology, results, recommendations, and conclusions. The second chapter comprises a literature review, offering an in-depth analysis of relevant literature pertaining to the research topic. It surveys existing studies, theories, methodologies, and findings related to LWW treatment and the use of specific treatment methods. The third chapter contextualizes the research within the broader project scope and discusses any previous studies or related work conducted in the field, providing necessary background information. Subsequent chapters focus on specific treatment methods, including the use of extracellular polymeric substances (EPS), electrocoagulation (EC) treatment using packed bed reactors with sacrificial anodes, and methods to enhance the treatment performance of ultrafiltration (UF) membranes using EPS. Each chapter presents detailed discussions on the principles, applications, effectiveness, and results of the respective treatment methods. The final chapter summarizes the key findings of the research, draws conclusions based on the results obtained, and provides insights into future research directions and potential areas for further investigation. The thesis concludes with a comprehensive list of references cited throughout the document, facilitating further reading and verification of information. This structured organization ensures a logical progression of the research and facilitates comprehension for readers.

3.6. Novelty of the research

The originality of this work can be summarized as:

- Pioneering the treatment of commercial LWW through the application of EPS derived from the fermentation of crude glycerol. This novel approach harnesses the potential of EPS, a by-product of microbial fermentation, for effective wastewater treatment, showcasing an environmentally friendly and sustainable solution.
- Introduction of PBEC system for the treatment of commercial LWW, featuring granular sacrificial iron electrodes packing. This innovative EC approach targets the comprehensive removal of contaminants, including suspended solids, colloidal solids, COD, metals, and surfactants. The concurrent use of EPS further enhances the efficiency of this treatment process and provides a sustainable alternative of chemical polymers.
- Advancement in the treatment of commercial LWW through the application of UF membrane technology along with EPS. This involves not only the effective management of membrane clogging but also the strategic utilization of EPS. The control of clogging is achieved through the modulation of operating parameters and employing EPS, enhancing the overall performance of the UF membrane system.

4. TREATMENT OF LAUNDRY WASTEWATER (LWW) USING EXTRACELLULAR POLYMERIC SUBSTANCES (EPS)

4.1. Article Introduction

Article's title	Treatment of laundry wastewater using extracellular polymeric substances (EPS)
Names of the authors and their professional affiliations	Sushil Kumar[1], Ali Khosravanipour Mostafazadeh[2], Patrick Drogui[1], R D Tyagi[3,4] 1. Institut National de la Recherche Scientifique (INRS), Centre-Eau Terre Environnement (ETE), Université du Québec, 490 Rue de la Couronne, Québec (QC), Canada G1K 9A9 2. Quebec Center for Studies of Chemical Processes (CÉPROCQ), 6220 Rue Sherbrooke Est Montréal, Québec, CANADA 3. BOSK-Bioproducts, 100-399 rue jacquard, Quebec (QC) G1N4J6 4. Distinguished Professor, School of Technology, Huzhou University, China.
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Status of the paper	Published, available online
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DOI	https://doi.org/10.1186/s40068-023-00289-5

4.2. Abstract

The problem of management and treatment of wastewater from commercial laundries is a matter of concern. The present study provides an effective and eco-friendly solution to the treatment of wastewater from commercial laundries in Quebec (Canada) by using the extracellular polymeric substance (EPS) as a bio-flocculant. EPS was produced from the valorization of crude glycerol and paper mill sludge by a bacterial strain (BS-04). Two different types of EPS: Slime EPS (S-EPS) and Broth EPS (B-EPS) were used for the treatment of commercial laundry wastewater (CLWW). This is the first study for the treatment of CLWW using bio-flocculant EPS. A comparison between the conventional treatment of laundry wastewater (LWW) by chemical coagulants ($FeSO_4$, $CaCl_2$, Alum) and enhanced treatment by bio-flocculant EPS has been drawn in the study. Moreover, LWW treatment by a combination of EPS and chemical coagulants was also investigated. It was observed that S-EPS (0.6 g/L) gave better flocculation activity (FA) than B-EPS. S-EPS alone can remove 83.20% of turbidity, 77.69% of suspended solids (SS), and 76.37% of chemical oxygen demand (COD). The best results were obtained by combining S-EPS (0.6 g/L) with alum (300 mg/L) at pH 7 for a treatment time of 30 min. This combination was able to remove 98% of turbidity, 95.42 % of SS, and 83.08% of COD from LWW. When treatment time has been increased to 4 h at pH 7, it resulted in more than 88% COD removal from CLWW.

Keywords:

Commercial laundry wastewater (CLWW); Extracellular polymeric substance (EPS); Slime EPS (S-EPS); Broth EPS (B-EPS); Linear alkyl benzene sulfonates (LAS); Nonylphenol Ethoxylates (NPE); Chemical oxygen demand (COD)

4.3. Introduction

There has been a persistent presence of contaminants in the water that leads to its pollution, because of which treating this polluted water has been a huge concern, all over the globe. Commercial laundries have been contributing to this pollution over the years as 1 kg of clothing requires about 15 L of water for washing, which in turn produces approximately 400 m^3 of wastewater on daily basis (Ciabattia *et al.*, 2009). Laundry wastewater (LWW) has been composed of organic contaminants that include surfactants, fats, and detergents along with inorganic contaminants such as sand and soil dust (Zhu *et al.*, 1998). Linear alkylbenzene sulfonates (LAS) and nonylphenol ethoxylates (NPEOs) are the most used surfactants in laundry detergent formulations, which often lead to environmental degradation causing health problems for animals and humans both. It has a serious negative impact on the aquatic biota as well (Jardak *et al.*, 2016). Many studies have found the presence of microplastics in LWW (Akarsu & Deniz, 2021; Conley *et al.*, 2019). It must be noted that such emerging contaminants are of great concern due to their endocrine and hormonal disrupting nature. These emerging contaminants can also lead to a genetic mutation in aquatic animals (Lissens *et al.*, 2003). Therefore, the urgent demand of the hour is to treat this polluted water.

Various physicochemical processes such as electrocoagulation (Chou *et al.*, 2009; Elazzouzi *et al.*, 2018; Gomes *et al.*, 2007; Han *et al.*, 2002; Holt *et al.*, 2005; Hu *et al.*, 2003; İrdemez *et al.*, 2006; Janpoor *et al.*, 2011; Kabdaşlı *et al.*, 2009; Kumar *et al.*, 2004; Larue *et al.*, 2003; Yüksel *et al.*, 2009; Zaroual *et al.*, 2006), membrane filtration (Ahn & Song, 1999; Bhattacharyya *et al.*, 1978; Bilad *et al.*, 2020; Carbonell-Alcaina *et al.*, 2016; Corbatón-Báguena *et al.*, 2015; Gitis *et al.*, 2006; Guilbaud *et al.*, 2010; Manouchehri & Kargari, 2017), adsorption (Ahmad *et al.*, 2019; Chen *et al.*, 2008; Corona *et al.*, 2021; Veli *et al.*, 2019; Veli *et al.*, 2021); biological processes (Andersen *et al.*, 2002; Ashfaq *et al.*, 2017; Bagheri & Mirbagheri, 2018; Deowan *et al.*, 2015; Emaminejad *et al.*, 2019; Hamedi *et al.*, 2019; Hoinkis *et al.*, 2012; Iorhemen *et al.*, 2016; Lohaus *et al.*, 2018; Madaeni *et al.*, 1995; Mahmoudi *et al.*, 2020; Paris & Schlapp, 2010) and combined treatment processes (Bokhary *et al.*, 2018; ElSherbiny *et al.*, 2019; Emaminejad *et al.*, 2019; Fan *et al.*, 2001; Hamedi *et al.*, 2019; Howe & Clark, 2006; Huang *et al.*, 2019; Jia *et al.*, 2017; Kamarudin *et al.*, 2003; Kamaruzzaman *et al.*, 2019; Kim *et al.*, 2014; Mostafazadeh *et al.*, 2019; Siswoyo *et al.*, 2019) have been applied for the treatment of LWW. Biological processes are inefficient in eliminating persistent organic pollutants. Also, the availability of excessive chemical oxygen demand (COD) makes biological processes incompatible with an explicit application for LWW treatment (Bokhary *et al.*, 2018).

Anaerobic treatment and membrane bioreactors are potential methods for the treatment of CLWW (Braga *et al.*, 2015; Delforno *et al.*, 2015; Faria *et al.*, 2019; Moura *et al.*, 2019). However, challenges associated with them consist of the expensive treatment process, the existence of excessive ammoniacal nitrogen in the effluents, and membrane clogging in membrane filtration makes them less effective for industrial-scale application (Corbatón-Báguena *et al.*, 2015; Hamedi *et al.*, 2019; Howe & Clark, 2002). The treatment of LWW by utilizing chemical flocculants has been found in the literature (Šostar-Turk *et al.*, 2005; Terechova *et al.*, 2014). The removal of particulate matter along with phosphorus and heavy metals has been effectively performed by the easy and economical coagulation (alum and ferrous sulfate) and flocculation processes. The chemical flocculants are not considered eco-friendly despite the advantages of being cost-effective. On the other hand, environment-friendly biopolymers like extracellular polymeric substance (EPS) can be used for LWW treatment as they are sustainable and environmentally friendly, and they minimize the harmful risks posed by the chemical flocculants such as their carcinogenic nature with low biodegradability.

Bio-flocculants originate from the natural secretions of bacteria, and cell lysis which is harmless, sustainable, biodegradable, and poses no secondary pollution risk. The predominant constituents of bio-flocculant (or extracellular polymeric substances or EPS) such as protein, glycoprotein, polysaccharide, and nucleic acid are generated by microbes during the growth phase. They have been investigated as a flocculating agent for dewatering and sludge settling (Huang *et al.*, 2019; Kaur *et al.*, 2022; Liu *et al.*, 2009). EPS is present both outside of cells and in the interior of microbial aggregates. The forms of EPS that occur outside of microbial cells can be divided into bound EPS (loosely bound polymers, sheaths, condensed gels, capsular polymers, and attached organic matters) and soluble EPS or loosely bound EPS (soluble macromolecules, colloids, and slimes) (Laspidou & Rittmann, 2002).

The use of EPS as a flocculating agent has been used by other researchers in the biological treatment of wastewater (Kaur *et al.*, 2019; Ndao *et al.*, 2022), but the application of EPS to treat LWW is not reported in the existing literature. In recent studies, EPS was effectively used to treat composting and landfill leachate (Kaur *et al.*, 2019; Ndao *et al.*, 2022). The study achieved a removal efficiency of 84% for COD, 96% for phosphorus, 97% for nitrates-nitrites, 75% for ammoniacal nitrogen, 29%-99% for metals, and 95% for colors (Kaur *et al.*, 2019). In another study, when EPS was used in combination with $FeSO_4$ for the treatment of landfill leachate. During the Jar test study, $FeSO_4$ was added to the glass beaker and stirred fast at 120 rpm for 90s. Thereafter at adjusted pH, the process effectively removed 82.00% of COD, 44.00% of total nitrogen, 50.00% of phosphorus and 64.30% of Ca, and 62.40% of Mg (Ndao *et al.*, 2022). However, there is a lack of studies using bio-flocculants. Resulting in negligible utilization of bio flocculants for the treatment of LWW (Mohan, 2014).

Therefore, the purpose of this study was to use bacterial EPS produced by the use of waste streams as substrate (crude glycerol in combination with industrial pulp and paper mill sludge) with chemical coagulants for LWW treatment. Three chemical coagulants (Alum, $FeSO_4$, and $CaCl_2$) were selected and used for the treatment of commercial laundry wastewater (CLWW).

4.4. Materials and Methodology

Origin and characterization of commercial laundry wastewater (CLWW):

Raw laundry wastewater (LWW) sample was collected from commercial laundry by VEOSwater (Terrebonne, Quebec) and the samples were supplied to the INRS laboratory. The effluents have been collected at the water outlet point of the commercial laundry room (Montreal, Quebec). The 100 L of LWW effluent was stored in 5 buckets of 20 L each, over a period of 24 hours for further analysis. The samples were stored in a cold room at a temperature of -4ºC.

The commercial laundry wastewater was characterized to measure pH, turbidity, conductivity, suspended solids (SS), total solids (TS), chemical oxygen demand (COD), total nitrogen (TN), total phosphorous (TP), nonylphenolethoxylate (NPEOs), and metals.

EPS Production using crude glycerol and industrial sludge.

EPS was produced using the bacterial strain BS-04 (isolated from wastewater sludge), which was grown on crude glycerol and secondary sludge (from pulp and paper mill wastewater treatment) as carbon substrates (Subramanian *et al.*, 2010). The process involved 96 h of fermentation at controlled pH, temperature, and dissolved oxygen. The fermented broth obtained after the fermentation process is called broth EPS (B-EPS). Centrifugation was done to the fermented broth at 4ºC for 20 min at 6000 g to obtain slime EPS (S-EPS) in the supernatant (Nouha *et al.*, 2016a).

To measure the dry weight of S-EPS the supernatant obtained was mixed with ethanol (95%v/v) in a 1:2 ratio and kept at -20ºC overnight for phase separation. The precipitates (solid phase) of S-EPS were collected by centrifuging at 6000 g at 4ºC for 20 min. The centrifuged pallet of S-EPS was dried at 60ºC to a constant weight.

The concentration of EPS was calculated using Eq. (1):

$$\text{EPS (g/L)} = (W_2 - W_1)/V \qquad \textbf{Equation 1}$$

Where W_1 = initial weight of the empty container (in g); W_2 = final dry weight of the container with dried sample (in g); and V = volume of the sample (in L).

The dry weight of capsular EPS (C-EPS) was determined by mixing the biomass pellets (obtained by centrifugation of fermented broth) with water in equal volume and then the mixture was heated at 60ºC in the water bath for 60 min to release the C-EPS in the liquid phase. Released C-EPS in the liquid phase was then collected by centrifugation at 6000 g at 4ºC for 20 min.

The total EPS concentration or broth EPS (B-EPS) concentration contains was calculated by using Eq. (2):

$$\text{B-EPS (g/L)} = \text{S-EPS (g/L)} + \text{C-EPS (g/L)} \qquad \textbf{Equation 2}$$

The collected S-EPS and B-ESP in liquid form were used for LWW treatment. Figure 1 shows the mechanism of CLWW treatment using EPS as a bio-flocculant. During the process of charge neutralization, the bio-flocculant particles get adsorbed on the surface of colloidal particles, resulting in reducing the repulsive force between the adjacent particles. This causes the particles to combine by increasing van der Waals interaction. During the bridging process, high molecular weight bio-flocculants are adsorbed on colloidal particles. Due to their large sizes, the bio-flocculants bridge particles and hence creates large flocs. During the patching mechanism, bio-flocculant particles come in contact with colloidal particles and neutralize a portion of the particle's surface. The opposite charge makes adsorbed bio-flocculants act as patching points for nearby particle.

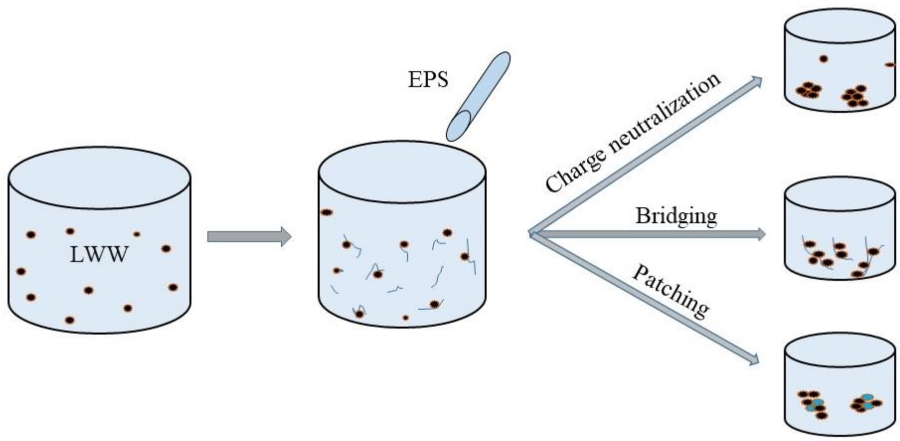


Figure 4.1 Mechanism of LWW treatment process

CLWW treatment

Identification of optimized pH for chemical coagulants

The jar test experiments were conducted for ferrous sulfate, alum, and calcium chloride to study flocculation activity at different pH (3, 5, 7, 9, and 11). During the experiment, 300 mL of LWW sample was taken into 1 L glass beaker and different pH were maintained by adding acid (1 M H_2SO_4) or base (2 M NaOH). A fixed concentration (300 mg/L) of each coagulant was added to different beakers, followed by rapid mixing (120 rpm) for 90 seconds. Then beakers were kept for agitation at 40 rpm for 30 min. Subsequently, the liquid mixture was shifted into a 1 L glass cylinder for 30 min settling, after that, the supernatant was taken and tested for FA.

Flocculation activity can be determined by using Eq. (3):

FA= (B – A) *100/B **Equation 3**

Where A = turbidity of the sample (with added coagulant) after settling for 30 min, and B = turbidity of the control (without coagulant) after settling for 30 min.

Effect of coagulant concentration

Jar test experiments were performed at suitable pH, and different coagulant doses (100 mg/L, 300 mg/L, 600 mg/L, 900 mg/L, 1200 mg/L, 1500 mg/L, and 2000 mg/L) were added to the glass beakers containing CLWW, respectively. Followed by 90 seconds of rapid mixing (120 rpm) and 30 min of slow agitation (40 rpm) in the jar test, the liquid mixture was transferred to a 1 L glass cylinder for settling. After 30 min of settling, the supernatant was collected and analyzed for flocculation activity.

Identification of optimum pH using EPS (S-EPS or B-EPS)

Experiments were performed for S-EPS and B-EPS. The pH of the LWW samples was adjusted to different values (3, 5, 7, 9, and 11). Rapid agitation (120 rpm) was done for 90 seconds to stabilize pH. Then fixed concentration (0.2 g/L of LWW) of S-EPS and B-EPS were added to different beakers containing LWW. Following the addition of EPS, slow agitation (40 rpm) was carried out for 30 min. After the agitation, the liquid mixture was transferred to a glass cylinder for settling to determine the flocculation activity.

Identification of optimum EPS concentration

In search of the optimal dose of EPS for maximum treatment efficiency, different concentrations (0.1 g/L, 0.2 g/L, 0.3 g/L, 0.4 g/L, 0.5 g/L, and 0.6 g/L) of S-EPS and B-EPS were used to perform the flocculation experiments. Each type of EPS was added to the glass beakers containing LWW and then jar tests were performed separately (as mentioned in the above sections) to determine the treatment efficiency. The concentration at which the visible flocs can be seen was considered for further optimization. After the jar test experiment followed by 30 min settling, the supernatant was analyzed (Turbidity, SS, TS, COD, nitrogen, phosphorous, metals, and NPEOs) to determine the pollutant removal efficiency.

Combination of EPS and Chemical Coagulant

Aluminum salt (alum) is as of now the most favored coagulant due to its efficiency and economics; however, traces of aluminium residuals in treated water are a strong agent for Alzheimer's disease (Campbell, 2002). Another environmental risk associated with the widespread use of aluminium salt as a primary coagulant is sludge disposal (Priya *et al.*, 2017). The use of natural coagulants such as bio-flocculants is a cost-effective and efficient remediation strategy for the treatment of extremely turbid water, but its composition tends to increase the organic content in water (Verma *et al.*, 2012). They also have insignificant flocculant activity in low turbid water because of limited repulsive force between dispersed colloidal particles. As a result, low flocculating properties, insignificant yields, and high production costs have limited the usage of only natural coagulants in water treatment plants (Huang *et al.*, 2014). Therefore, it was a scientific and economic approach to use metal coagulants along with the bio-flocculants.

Flocculation activity is improved while combining metal coagulant and bio-flocculant owing to three main mechanisms taking place simultaneously: i) the charge neutralization; ii) the sweep coagulation and iii) flocs bridging (Bo *et al.*, 2012). The ionic nature of metal and its size and charge density regulate its interaction with negatively charged EPS. The mechanism behind the removal of organic matter and other contaminants is that the introduction of positively charged coagulants destabilizes the stable negative charge of the target particles by compressing the double layer. This upset decreases the distance or repulsion between particles, in turn decreasing the zeta potential. The particles are then able to get close enough together due to Vander Waals forces. As a result, van der Waals interactions are predominant, allowing aggregation of suspended fine particles and subsequent flocculation.

The association of EPS with coagulants leads to a better particle aggregation due to high molecular weight and structure of EPS. EPS has hydroxyl sites, amine and carboxyl functional groups. In fact, a previous study has shown the presence of these functional sites in EPS structure produced from glycerol and activated sludge by using FTIR (Nouha *et al.*, 2016a). Also, EPS molecules can attach to neutralized particles in solutions and simultaneously cross-link other EPS molecules which are also attached to other neutralized colloids (More *et al.*, 2014). All these phenomena under combination of EPS and coagulant resulted in enhanced treatment of LWW. To enhance the performance of EPS, a chemical coagulant was used in combination with EPS. During the LWW treatment process, suitable pH (as determined in the section on optimization of pH for EPS) and chemical coagulant (as determined in the section effect of chemical coagulant) were used to carry out the experiments. Different concentrations (0.1 g/L, 0.2 g/L, 0.3 g/L, 0.4 g/L, 0.5 g/L, and 0.6 g/L) of each type of EPS were added to the glass beakers containing LWW. Rapid mixing was done for 90 s for each set followed by 30 min of slow mixing at 40 rpm. Treated water was kept for settling in a glass cylinder for 30 min. The collected supernatant was then analyzed to determine the removal efficiency of different parameters (Turbidity, SS, COD, nitrogen, phosphorous, metals, and NPEOs).

Effect of treatment time

After the determination of suitable pH and EPS concentration, experiments were performed to determine the treatment time to achieve the maximum treatment efficiency. During the treatment process, 300 mL of commercial LWW was added to each of the glass beakers. Then the suitable concentration of S-EPS (as determined in the section on optimization of EPS concentration) was added to each beaker. Then pH was adjusted to a suitable value (as determined in the section on optimization of pH) by using 1M H_2SO_4 or base 2M NaOH. After 90 s of rapid agitation, agitation speed was reduced from 120 to 40 rpm and kept for incubation. The jar test experiments were performed for different time periods (0.5, 1, 2, 4, 6, and 12h), and treated water was set aside to settle by gravity for 30 min. After settling, the supernatant sample was analyzed to determine the treatment efficiency for each treatment time.

Analytical Methods

The methods used for the analysis of samples are certified methods by CEAEQ (Center of Expertise in Environmental Analysis of Quebec) (l'Environnement & climatiques, 2014) under the responsibility of the Government of Quebec. The mission of CEAEQ is to standardize the methods and analytical tools used in environmental analysis. Table 1 depicts the analytical methods for different parameters.

Table 4.1 Analytical methods

Parameters	Analytical Method
Ph	pH meter (Fisher scientific)
Turbidity	Turbidity meter, (Fisher scientific)
COD	Spectrophotometer, CEAEQ, 2014 (Method of analysis -. 315 MA-COD 1.0)
SS	Gravimetric analysis, CEAEQ, 2017(MA. 100 – S.T. 1.1)
TS	Gravimetric analysis, CEAEQ, 2012 (MA. 104 – S.S. 2.0)
NPEOs	LC-MS at 350ºC, CEAEQ (MA 400 – NPEO 1.0)
Metals	ICP-AES

4.5. Results and Discussions

Characteristics of CLWW

The LWW samples collected from the discharge basin (raw LWW) were characterized for turbidity, pH, conductivity, SS, TS, COD, NPEOs, TN, TP & metals. The characterization results are depicted in Table 2. It was found that LWW contained high amount of turbidity, SS, TS, COD, and NPEOs. The presence of a high amount of turbidity and SS was a result of residual dirt/soil in the dirty clothes/fabrics. The analysis was also carried out to detect the amount of NPEO present in the laundry wastewater. A significant amount of NPEO (570-720 µg/L) was found in the sample. The CLWW also contained a significant amount of metals like calcium (Ca), magnesium (Mg), aluminum (Al), sodium (Na), and potassium (K). A small amount of heavy metals were also detected in the CLWW.

The characteristics of CLWW were compared with the standards for water discharge outlined by Health Canada and Ville de Quebec. It was found (Table 2) that the concentration of different parameters (turbidity, SS, TS, COD, NPEO, metals) of CLWW was almost 5 times higher than the recommended value. Hence it was necessary to treat CLWW before discharge to the water bodies.

Table 4.2 Characteristics of CLWW

Parameters	Value of the parameter in CLWW	Discharge standard (Ville de Quebec)	Reuse standard (Toilet flushing) (Health Canada)
Turbidity (NTU)	110 ± 5	n.a	<5
pH	10.4 ± 0.5	6.5 - 8.5	6-9
Conductivity (µS/cm)	494 ± 20	n.a	n.a
Suspended solids (mg/L)	60 ± 10	n.a	<20
Total solids (mg/L)	622 ± 20	<100	n.a
COD (mg/L)	579 ± 30	<150	<100
NPEOs (µg/L)	570 ± 150	<200	n.a
Sulphate (mg/L)	22± 5	n.a	n.a
Sulphite (mg/L)	0.2 ± 0.05	n.a	n.a
Nitrate (mg/L)	4 ± 1	n.a	n.a
Nitrite (mg/L)	1.5 ± 0.5	n.a	n.a
Ammoniacal nitrogen (mg/L)	2.3 ± 1	n.a	n.a
Total nitrogen (mg/L)	9.7 ± 0.5	n.a	n.a
Phosphate (mg/L)	21 ± 2.0	n.a	n.a
Aluminium (mg/L)	0.096 ± 0.02	n.a	n.a

Calcium (mg/L)	18.6 ± 2.0	n.a	n.a
Magnesium (mg/L)	3.87 ± 0.5	n.a	n.a
Sodium (mg/L)	104 ± 10	n.a	n.a
Potassium (mg/L)	3.76 ± 0.5	n.a	n.a
Sulphur (mg/L)	20.6 ± 2.0	n.a	n.a
Zinc (mg/L)	0.42 ± 0.15	n.a	n.a
Copper (mg/L)	0.03 ± 0.01	n.a	n.a
Iron (mg/L)	0.23 ± 0.2	n.a	n.a
Manganese (mg/L)	0.019 ± 0.005	n.a	n.a
Lead (mg/L)	<0.003	n.a	n.a
Nickel (mg/L)	0.004 ± 0.002	n.a	n.a
Cadmium (mg/L)	<0.0003	n.a	n.a
Chromium (mg/L)	0.0015 ± 0.001	n.a	n.a

n.a.: Not applicable

Treatment of CLWW using chemical coagulants

Identification of optimized pH for chemical coagulants

The chemical coagulants (alum, $FeSO_4$, and $CaCl_2$) were investigated for the treatment of CLWW. The results were compared based on zeta potential and FA. The treatment of CLWW by alum resulted in an initial increase in zeta potential (during pH 3-7) and then it gradually decreased with an increase in pH from 7 to 11 (Supplementary figure 1). The increase in zeta potential (pH 3-7) is probably due to the surface charge neutralization (Cosa *et al.*, 2013). Zeta potential values of −7.7 mV, −2.7 mV, 0.4 mV, −4.7 mV, and

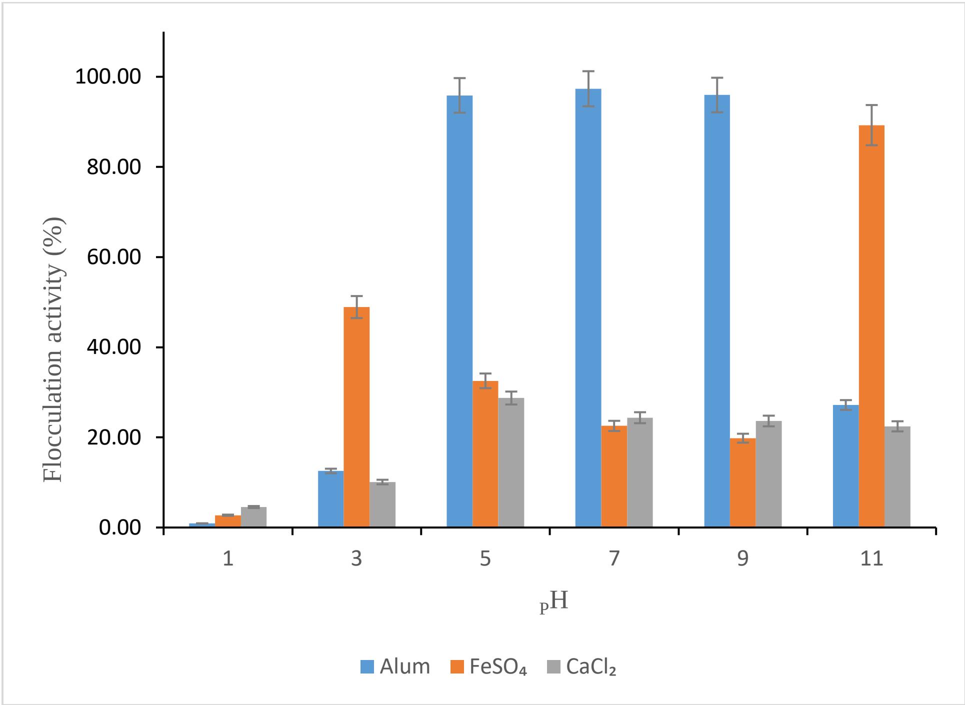
−11.9 mV was obtained at pH 3, 5, 7, 9, and 11 respectively. Similarly, the FA also increased with an increase in pH up to pH 7, and then it decreased with a further increase in pH. At pH 3, only small FA (12.55%) was observed. FA increased to 97.00% at pH 7 and then decreased to 27.00% at pH 11. However, when $FeSO_4$ was used as a coagulant the zeta potential value varied from pH 3 to 11. Zeta potential values of −7.7 mV, −8.3 mV, −9.4 mV, −9.7 mV, and −3.9 mV was found at pH 3, 5, 7, 9, and 11 respectively. The maximum FA of 89.00% was observed at pH 11.

However, when $CaCl_2$ was used as a chemical coagulant, it was observed that the value of zeta potential initially increased with an increase in pH (3 to 5) and then became almost constant (from pH 7 to 9). Zeta potential values of −10.7 mV, −10.3 mV, −10.7 mV, −11.7 mV, and −13.2 mV was found at pH 3, 5, 7, 9, and 11 respectively. The maximum FA of 29% was observed at pH 5. Figures 2(a) and (b) show the impact of pH on flocculation activity using chemical coagulants and EPS, respectively.

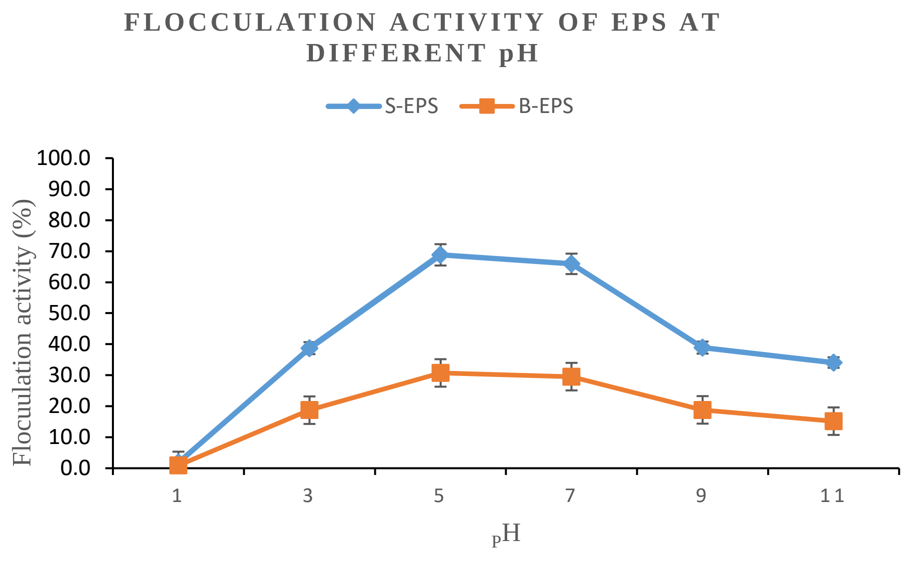
The results for FA for different coagulants indicate that pH has a significant role in flocs formation and removal of pollutants from CLWW. It has been identified that the suitable pH for Alum, $FeSO_4$, and $CaCl_2$ were 7, 11, and 5, respectively. From the results, it is evident that the coagulation-flocculation process is widely dependent on the pH. It imparts a huge effect on the ionization and solubility of chemical coagulants. However, the optimized pH will depend upon the type of chemical coagulant used. Considering the optimized pH for chemical coagulant alum (pH=7), it was decided to keep pH 7 as the optimized pH for future experiments.

The main factors behind the removal of pollutants by coagulation-flocculation are charge neutralization, double-layer compression, bridging, and particle entrapment. During the process, positively charged chemical coagulants neutralize the negatively charged contaminants present in the wastewater sample. It destabilizes the stable particles by compressing the double layer (Kaur *et al.*, 2019).

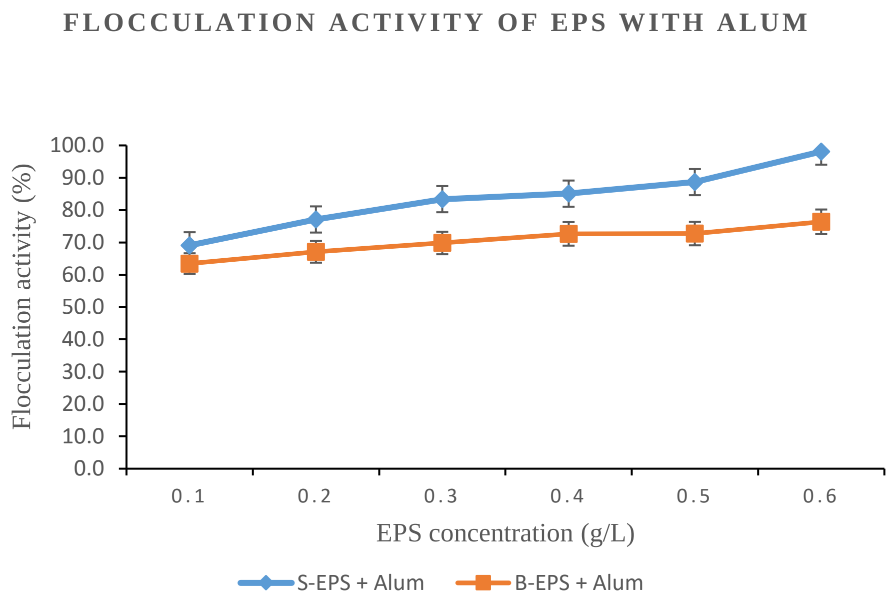
Due to a decrease in the distance between the double layer, the repulsive forces between the particles also decrease which results in a decrease in zeta potential. Once the particle comes closer, it starts bridging with the other adjacent particles and results in flocs formation. This process of flocs formation followed by settling helps in the removal of pollutants from the wastewater sample.



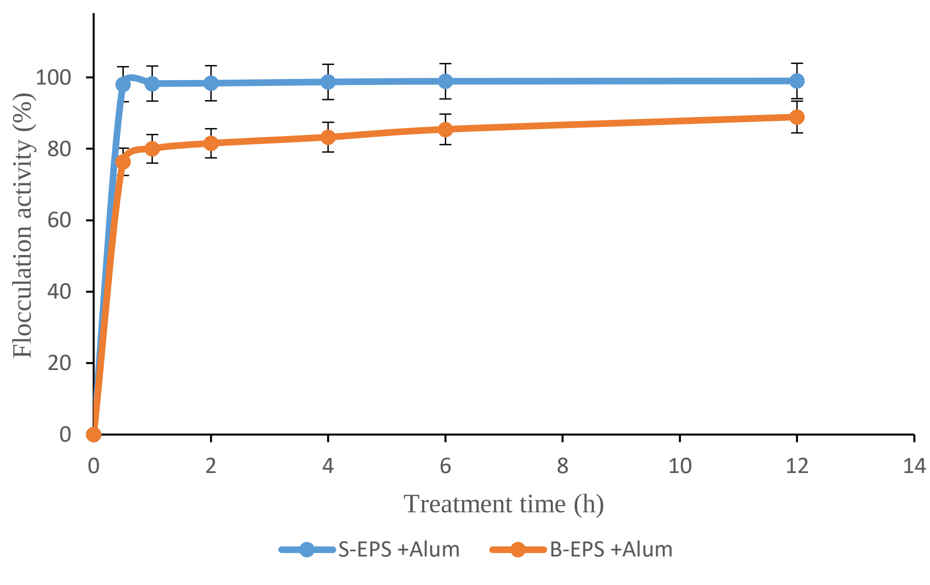
(a)



(b)



(c)



(d)

Figure 4.2: Flocculation activity of (a) Chemical coagulants at different pH, (b) S-EPS and B-EPS at different pH, (c) EPS in the combination of alum at different EPS concentrations at pH 7, (d) S-EPS and B-EPS at different treatment time

Effect of coagulant concentration

To identify the optimum concentration of each coagulant, jar test experiments were performed by using different coagulant doses (100, 300, 600, 900, 1200, 1500, and 2000 mg/L of LWW). When alum concentration was raised from 100 to 2000 mg/L at pH 7, the zeta potential value also increased from −12.8 to 1.8 mV, respectively. With the increase in alum concentration, FA also increased from 21.82% to 99.18%. However, when $FeSO_4$ concentration increased from 100 to 2000 mg/L at pH 11, the value of zeta potential increased from −14.7 to −5.8 mV, respectively. Similarly, FA also improved from 14.09% to 91.09% by increasing the $FeSO_4$ dosage. When the concentration of $CaCl_2$ was increased from 100 to 2000 mg/L, the zeta potential value increased from −13.9 mV to −8.3 mV, respectively. With the increase in zeta potential value, the flocculation activity also improved from 4.55% to 22.45%. It has been clear that the concentration of chemical coagulants used for the treatment depends upon the properties of laundry wastewater samples (Huang *et al.*, 2019; Mahvi *et al.*, 2015). A reported study indicates that the appropriate coagulant dosage can produce stable and consistent flocs due to adsorption and stable interaction between particles (Huang *et al.*, 2019; Mahvi *et al.*, 2015). However, a lower coagulant dose may result in the formation of small flocs and can interfere with the settling of the particles. This may also result in a decrease in flocculation activity. Similarly, a higher coagulant dose may result in unstable flocs due to particle repulsion and weak flocs formation.

Table 3 shows the removal of pollutants by using different chemical coagulants. It is indicated from Table 3 that alum and $FeSO_4$ provided higher turbidity, SS, and TS removal as compared to $CaCl_2$. Alum (2000 mg/L) alone was effective in removing 99.18% turbidity, 98.00% SS and 91.33% TS. Similarly, 91.09% turbidity, 78.67% SS and 68.09% TS were removed from CLWW at 2000 mg/L dose of $FeSO_4$. However, for $CaCl_{2,}$ the maximum removal efficiency was achieved at 1500 mg/L dose. The removal of turbidity, SS, and TS for $CaCl_2$ (1500 mg/L) was found to be 36.18%, 29.17%, and 36.09% respectively.

Table 4.3 Treatment of CLWW using different concentrations of chemical coagulants

Coagulant	Coagulant dose (mg/L)	Removal (%)			
		Turbidity	TSS	TS	COD
Alum	100	21.82	6.17	7.36	8.17
	300	33.64	21.33	31.99	22.98
	600	65.45	69.67	44.53	33.68
	900	94.36	76.33	65.92	44.35
	1200	98.09	84.67	79.39	63.32
	1500	98.55	94.67	85.14	80.98
	2000	99.18	98.00	91.33	85.10
$CaCl_2$	100	12.73	7.67	7.68	6.37
	300	20.00	11.00	12.04	11.57
	600	24.36	19.17	21.13	19.13
	900	26.36	24.83	27.09	26.95
	1200	34.55	27.83	29.97	36.92
	1500	36.18	29.17	36.09	42.60
	2000	25.36	20.67	32.72	41.29
	100	14.09	12.67	9.60	6.23
	300	23.36	22.67	20.39	18.47

$FeSO_4$	600	78.73	52.67	44.94	25.29
	900	83.36	68.83	53.95	31.37
	1200	89.64	76.17	60.51	36.75
	1500	89.91	77.17	65.85	40.19
	2000	91.09	78.67	68.09	47.80

COD removal of 85.10, 42.60and 47.80% COD was achieved using alum (2000 mg/L), $CaCl_2$ (1500 mg/L), and $FeSO_4$ (2000 mg/L), respectively. It was seen that when the dosage of alum and $FeSO_4$ increased from 100 mg/L to 2000 mg/L, corresponding COD removal was increased from 8.17% to 85.10% for alum and 6.23% to 47.80% for $FeSO_4$, respectively. However, $CaCl_2$ gave a slightly different performance. When the dosage of $CaCl_2$ was increased from 100 to 1500 mg/L, COD removal was enhanced. But when the dosage of $CaCl_2$ increased further from 1500 to 2000 mg/L, COD removal decreased. The maximum COD removal (42.60%) was obtained at a $CaCl_2$ concentration of 1500 mg/L. The reason behind the lower COD removal in the case of $CaCl_2$ at higher concentration (>1500 mg/L) was due to an excessive increase in positive charge, which increased the repulsive forces between the pollutant particles and resulted in a tendency of lower flocs formation. Since FA and pollutant removal rates were the highest for alum when compared to the other coagulants, a combination of EPS with coagulant was tested only with alum.

Treatment of CLWW using EPS (S-EPS and B-EPS)

Identification of optimum pH for CLWW treatment

To determine the best pH for CLWW treatment using EPS, experiments were conducted at various pH (3, 5, 7, 9, and 11). The EPS concentration was fixed at 0.2 g/L to conduct the jar test experiments. The results were compared based on flocculation activity and zeta potential. The treatment of CLWW using S-EPS resulted in an initial increase in zeta potential (from pH 3 to 5) and then it gradually decreased with an increase in pH from 5 to 11. Zeta potential values of −8.9, −1.4, −2.1, −8.7, and −10.9 mV was obtained at pH 3, 5, 7, 9, and 11, respectively. Similarly, the FA also increased while increasing the pH from 3 to 5. Then, FA remained almost constant till pH 7 and decreased with a further increase in pH. At pH 3, only small FA (38.73 %) was observed. FA increased to 68.8% at pH 7 and then decreased to 34.09% at pH 11. The isoelectronic point (IEP) seems to be reached at pH 5. When IEP is reached, the positive and negative charges of particles get neutralized, resulting in a higher probability of combining the dispersed and/or colloid particles. However, when B-EPS was used as a bio-flocculant the zeta potential value increases with an increase in pH up to 5, is almost constant from 5 to 7 pH, and then decreases with an increase in pH 11. Zeta potential values of −11.2 mV, −8.1 mV, −8.3 mV, −10.9 mV, and −13.1 mV was found at pH 3, 5, 7, 9, and 11 respectively. Similarly, the FA also increased with an increase in pH up to pH 5 and after that remains almost constant up to pH 7 and decreased with further increase in pH up to pH 11. At pH 3 it was observed that B-EPS shows only small FA (18.73%), at pH 5 it shows maximum FA (30.70%) and then decreased to 15.20% at pH 11. The results indicated that S-EPS showed higher FA (68.80%) when compared to B-EPS (30.70%) at pH 5. However, at pH 7 the S-EPS and B-EPS showed a huge difference in the flocculation activity (65.90% and 29.50% respectively). Figure 2(a), (b), (c), and (d) shows the flocculation activity of chemical coagulants at different pH, EPS (S-EPS and B-EPS) at different pH, EPS (S-EPS and B-EPS) in combination with alum at various EPS concentrations and EPS (S-EPS and B-EPS) in combination with alum at different treatment time.

The results for FA for EPS indicate that pH has a significant role in floc formation and removal of pollutants from CLWW. The bio-flocculant adsorbed on the surface of colloidal particles during the charge neutralisation process, lowering the repulsive force between the nearby particles. High molecular weight bio-flocculants are adsorbed on colloidal particles during the bridging process. The bio-flocculants bridge particles because of their huge diameters, resulting in massive flocs. Bio-flocculant particles interact with colloidal particles during the patching mechanism, neutralising a section of the surface. Adsorbed bio-flocculants function as patching points for surrounding particles due to the opposite charge. The suitable pH for S-EPS and B-EPS is pH 5. However, FA at pH 7 is almost similar to pH 5 (Fig 2b). Therefore, pH 7 can also be considered, because it will reduce the consumption of chemicals to adjust the pH. By adding H_2SO_4 and NaOH, the pH of the

CLWW sample can be adjusted to the desired optimized value. At these optimized pH values (pH 7), the visible flocs were formed efficiently, resulting in the removal of pollutants from the wastewater.

The variation in FA of S-EPS and B-EPS is possibly caused by the existence of various types of proteins and carbohydrates moiety. These compounds are complex molecules, which are structurally different and contain many functional groups. The structure and functional groups may change with the microorganism used for fermentation, the type of carbon substrate, and the time of fermentation (Higgins & Novak, 1997).

Also, another possible reason may be the fact that B-EPS is the combination of S-EPS and C-EPS. C-EPS can affect the FA of B-EPS because it contains a hydrophilic compound (hydroxyl groups) (Tian *et al.*, 2006). B-EPS also contains organic, inorganic, dead cells, etc. which may also affect the overall performance of B-EPS.

Effect of EPS concentration

Different concentrations of S-EPS and B-EPS were used at the optimum pH. Various concentrations (0.1, 0.2, 0.3, 0.4, 0.5, and 0.6 g/L) of S-EPS and B-EPS were used to remove the contaminants from CLWW. The zeta potential and FA were studied at different EPS concentrations. It was observed that when S-EPS concentration increased, the zeta potential value also increased. The values of zeta potential in mV were −7.7 mV, −3.4 mV, −1.4 mV, −0.6 mV, 0.4 mV, and 1.2 mV at corresponding S-EPS concentrations of 0.1 g/L, 0.2 g/L, 0.3 g/L, 0.4 g/L, 0.5 g/L, and 0.6 g/L, respectively. Similarly, the FA also increased with the increase in S-EPS concentration. The maximum FA of 83.27% was attained at an S-EPS dose of 0.6 g/L (Fig 2c). However, when B-EPS was used the zeta potential value was −16.3 mV, −8.1 mV, −6.5 mV, −4.3 mV, −3.7 mV, and, −3.5 mV at corresponding B-EPS concentration of 0.1 g/L, 0.2 g/L, 0.3 g/L, 0.4 g/L, 0.5 g/L, and 0.6 g/L. Similarly, the FA was raised with the rise in the B-EPS doses. The maximum FA (51.64%) was achieved when 0.6 g/L B-EPS was used. The results indicated that S-EPS displayed higher (83.27 %) FA when compared to B-EPS (51.64 %) at an EPS concentration of 0.6 g/L.

Combining EPS and chemical coagulant for the treatment of CLWW

Experiments were also conducted using EPS in the combination of alum (300 mg/L) at pH 7. When S-EPS is combined with alum for the treatment of CLWW it gives better FA as compared to S-EPS alone (Fig 2c). This indicates that EPS with or without alum plays a significant role in the removal of pollutants from CLWW samples. The FA increased with an increased in S-EPS concentration (Fig. 2c). The maximum FA of 98.09% was achieved by using S-EPS (0.6 g/L) and alum (300 mg/L. The flocculation activity for the combination of EPS and alum has been depicted in Figure 2c. Similarly, when B-EPS was used in combination with alum, it gave better FA as compared to B-EPS alone. The maximum FA of 76.36% was achieved using 0.6 g/L B-EPS and 300 mg/L of alum. The outcomes of the experiments were compared for the removal of pollutants (turbidity, SS, TS, COD, metals, etc.).

Removal of turbidity, SS, and TS

When EPS (S-EPS and B-EPS) were used alone for the treatment of CLWW, it resulted in slightly lower removal efficiency as compared to chemical coagulants (Table 4). The results indicated that the removal efficiency of turbidity, SS, and TS increased with the increase in EPS dosage. It was also observed that S-EPS gave better removal efficiency than B-EPS. S-EPS (0.6 g/L) alone can remove 83.27% turbidity, 77.69% SS and 78.66% TS. However, B-EPS (0.6 g/L) was able to remove only 51.64% turbidity, 48.38% SS and 46.15% TS from CLWW. Statistical analysis of the results obtained under optimum conditions by combining S-EPS with alum (analysis done in triplicate) indicates that the percentage of turbidity removal had a mean value of 98.09% with a standard deviation of 0.09, which means that it can be considered as constant with 0.09% accuracy. Also, the percentage TSS removal had a mean value of 95.42% with a standard deviation of 0.58 with accuracy 0.61%, and the percentage TS removal had a mean value of 94.80% with a standard deviation of 1.56 with accuracy 1.64%.

A higher removal of turbidity, SS, and TS was achieved by using S-EPS (0.6 g/L) in combination with alum (300 mg/L). S-EPS and alum together effectively removed 98.09% turbidity, 95.42% SS and 94.80% TS from CLWW. Similarly, B-EPS (0.6 g/L) in combination with alum (300 mg/L) removed 76.36% turbidity, 67.76% SS and 65.57% TS.

From the outcomes, it can be easily determined that the combination of S-EPS with alum gave higher removal efficiency for turbidity, SS, and TS when compared to S-EPS alone.

Removal of COD

When EPS (S-EPS and B-EPS) were used alone for the treatment of CLWW it gave lower COD removal (Table 4). During the treatment process, 76.37% COD removal was obtained at 0.6 g/L S-EPS concentration. However, 43.64% of COD removal was achieved using B-EPS (0.6 g/L) alone. An increase in EPS concentration increased the COD removal efficiency. The reason behind the lower COD removal at lower EPS dosage (<0.2 g/L) may be due to the less availability of bio-flocculant, which leads to the insufficient bridging of the particles and results in low settling of the suspended material. These results are in agreement with those reported on composting leachate treatment with EPS (Kaur *et al.*, 2019).

The combination of EPS (S-EPS and B-EPS) with alum enhanced the COD removal from CLWW. The combination of S-EPS (0.6 g/L) and alum (300 mg/L) facilitated COD removal (83.08%). Similarly, the combination of B-EPS (0.6 g/L) and alum (300 mg/L) was able to remove 67.20% of COD from the wastewater sample. The Statistical analysis shows that the percentage COD removal had a mean value of 83.08% with a standard deviation of 1.1 with accuracy 1.32%.

Table 4.4 Treatment of CLWW using EPS alone and in the combination with alum

Treatment	EPS concentration (g/L)	Removal (%) using S-EPS				Removal (%) using B-EPS			
		Turbidity	TSS	TS	COD	Turbidity	TSS	TS	COD
CLWW	0.1	34.45	29.65	30.49	34.47	10.55	9.46	10.79	11.22
	0.2	68.82	56.28	51.36	60.21	30.73	23.54	23.60	32.94
	0.3	76.00	68.08	60.01	68.82	38.64	32.85	33.98	36.35
	0.4	78.91	70.28	66.48	71.64	45.55	39.25	38.32	38.36
	0.5	81.73	76.21	73.41	74.73	50.55	45.19	42.96	41.55
	0.6	83.27	77.69	78.66	76.37	51.64	48.38	46.15	43.64
CLWW + Alum	0.1	69.09	36.13	45.00	44.76	63.45	42.43	37.26	27.84
	0.2	77.09	63.78	52.01	49.94	67.09	47.61	44.48	36.13
	0.3	83.36	72.46	65.91	63.06	69.82	52.63	47.16	40.27
	0.4	85.09	77.80	76.46	66.68	72.64	59.62	54.43	50.63
	0.5	88.64	85.02	83.19	71.17	72.73	66.66	62.93	56.84
	0.6	98.09	95.42	94.80	83.08	76.36	67.76	65.57	67.20

Removal of metals

CLWW contains a significant amount of metals like Al, Na, Mg, K, and Ca (Table 2). A small amount of heavy metals was also detected in the wastewater sample. When CLWW was treated alone with S-EPS, it displayed considerable removal of metals. At the initial S-EPS dose of 0.1 g/L, only a small concentration of Al (5.2%), Na (6.3%), Mg (5.7%), K (2.1%), and Ca (10.8%) was removed from the wastewater sample (Table 5). When S-EPS concentration was increased, it displayed considerable removal of metals from wastewater. When S-EPS concentration increased to 0.6 g/L, removal of Al, Na, Mg, K, and Ca was 72.2%, 94.3%, 92.6%, 85.3%, and 98.4% respectively. Table 5 indicates data for the metal removal at different EPS concentrations. For the treatment of CLWW using B-EPS, the elimination of metals enhanced with the rise in B-EPS concentration (Table 5). At the initial B-EPS dose of 0.1 g/L, the removal of Al, Na, Mg, K, and Ca was 3.12%, 4.41%, 3.76%, 1.85%, and 8.32% respectively. When the concentration of B-EPS was raised to 0.6 g/L the elimination of metals enhanced to 45.32% (Al), 68.01% (Na), 63.12% (Mg), 77.06% (K), and 78.77% (Ca).

When EPS was used in combination with alum, the metal removal was increased significantly with the increase in EPS concentration. For the combination of S-EPS and alum (0.1 g/L and 300 mg/L), only a small concentration (2-13%) of metals was removed. When the dose of S-EPS was raised to 0.6 mg/L the removal of Al, Na, Mg, K, and Ca was increased to 87.08 84.76, 97.3, 92.68, and 96.2%, respectively.

When B-EPS was used in combination with alum, it also shows a significant impact on metals removal. For the initial B-EPS dose of 0.1 g/L, the elimination of Al, Na, Mg, K, and Ca was 3.49%, 4.94%, 4.1%, 1.76%, and 9.4% respectively. When the concentration of B-EPS was increased to 0.6 g/L, the removal of Al, Na, Mg, K, and Ca increased to 52.52%, 75.93%, 69.62%, 73.31%, and 81.62% respectively. It was observed that the removal of Na (75.93%) and Ca (81.62%) was better than the removal of Al (52.52%) and Mg (69.62%).

Table 4.5 Metals removal from CLWW using EPS and chemical coagulants

Treatment	EPS concentration (g/L)	Removal (%) using S-EPS					Removal (%) using B-EPS				
		Al	Na	Mg	K	Ca	Al	Na	Mg	K	Ca
CLWW	0.1	5.20	6.30	5.70	2.10	10.80	3.12	4.41	3.76	1.85	8.32
	0.2	15.40	20.30	18.70	12.50	27.90	9.24	12.21	11.34	14.20	20.48
	0.3	32.60	38.10	36.90	27.40	46.70	18.56	25.67	20.35	27.11	37.96
	0.4	55.70	66.40	65.40	44.20	69.10	31.42	47.48	45.16	45.90	51.21
	0.5	67.80	84.20	79.70	78.60	85.70	41.68	55.94	56.60	62.17	64.99
	0.6	72.20	94.30	92.60	85.30	98.40	45.32	68.01	63.12	77.06	78.77
	0.1	6.34	5.80	6.38	2.33	13.23	3.49	4.94	4.10	1.76	9.40

CLWW + Alum	0.2	15.79	17.68	20.94	14.88	29.02	11.35	16.92	14.45	11.45	23.28
	0.3	37.77	39.05	41.33	31.41	48.57	22.91	27.87	28.55	23.91	45.63
	0.4	69.95	65.09	73.25	47.06	75.86	39.43	53.06	43.05	32.95	61.12
	0.5	84.72	79.46	89.26	84.25	89.13	46.56	68.01	56.34	61.71	72.57
	0.6	87.08	84.76	97.30	92.68	96.20	52.52	75.93	69.62	73.31	81.62

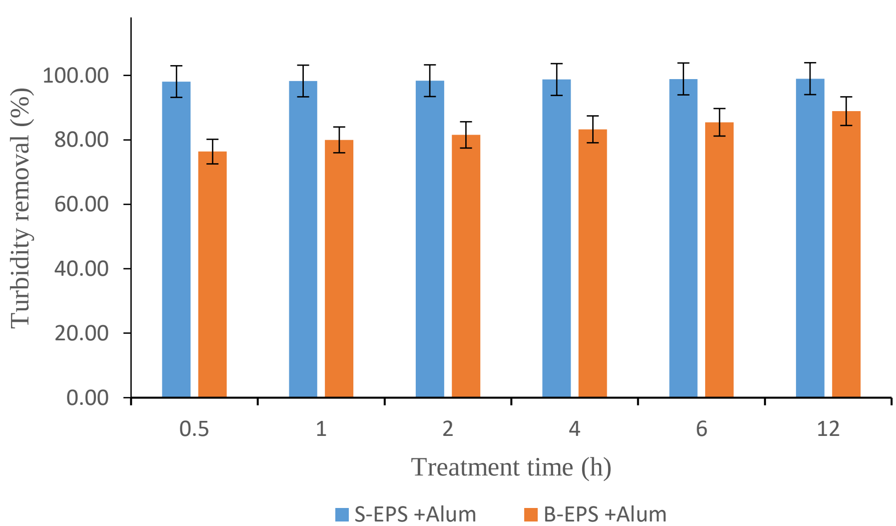
The removal of metals is because EPS possesses several adsorption sites for metals, which consist of proteins with aromatics and aliphatic sites and carbohydrates with hydrophobic sites (More *et al.*, 2014). The EPS contains a high amount of polysaccharides and proteins, which play a very important role in metal removal. Studies indicate that C=O (carbonyl groups), amides groups, and -OH (hydroxyl group) present in the proteins can effectively eliminate heavy metals by electrostatic interaction (Nouha *et al.*, 2016a; Nouha *et al.*, 2016b; Ruan *et al.*, 2013). In addition to that other functional groups like the C-O-C group (ether) with polysaccharides, C=O group with phenolic alcohol, -OH group with alcohol, and phosphorous and sulfur groups also contributes to complex reactions. Apart from this, the presence of a phosphorus group in a nucleic acid (particularly DNA) which is present in EPS also provides additional binding sites for metals. Additionally, nucleotides and uronic acids containing phosphorous groups available in EPS are having a negative charge and they can effectively attach with multivalent positive charged ions resulting in metal removal (Nouha *et al.*, 2018). It has been also reported that S-EPS contains a higher portion of proteins as compared to B-EPS (Pan *et al.*, 2010). Therefore, the amount of protein and its types play a significant role in the flocculation process.

Effect of treatment time for CLWW treatment:

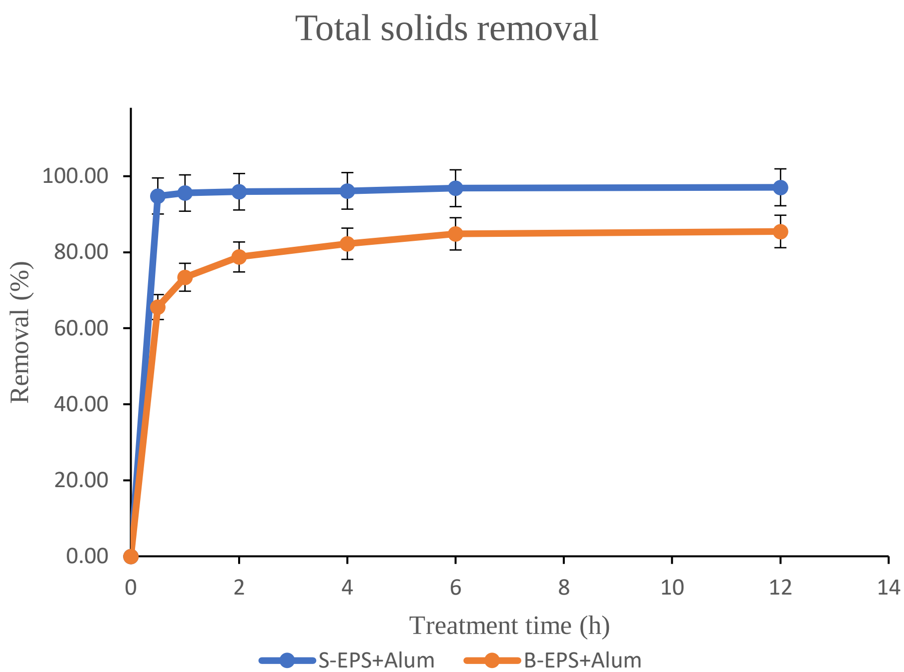
The impact of treatment time on flocculation activity is depicted in Figure 2d. It was observed that as the treatment time increased, it has a significant impact on the removal of contaminants like turbidity, SS, TS, COD, and metals.

When S-EPS (0.6 g/L) was used in combination with alum (300 mg/L), it was found that the value of turbidity decreased from 2.1 NTU to 1.1 NTU after 12h of treatment. It was observed that treatment time (after 0.5h) did not have a significant impact on the turbidity removal rate for S-EPS (Figure 3a). Similarly, the concentration of SS, TS, and COD also decreased with an increase in treatment time. The concentration of SS, TS, and COD after 12h of treatment was 1 mg/L, 18.18 mg/L, and 58.2 mg/L, respectively. It was also found that the removal of metals like Al and Mg increased significantly with an increase in treatment time (Figure 3(d) and 3(e)). The removal of Al and Mg increases from 72.2% and 92.6 % (30 min of treatment time) to 92.15% and 97.11%, respectively (Figure 3(d) and 3(e)). For metals like Na and Ca, only small removal (2-5%) was observed.

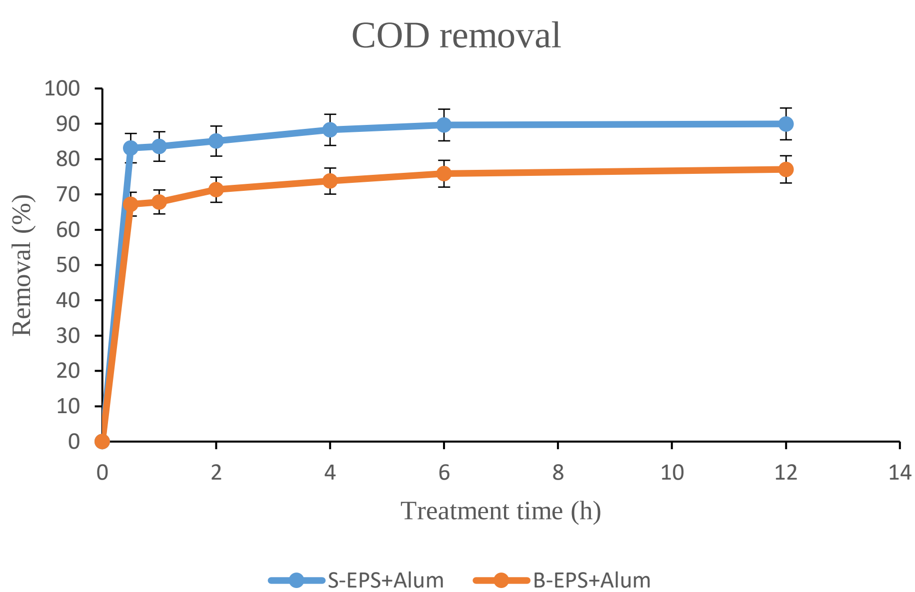
When B-EPS (0.6 g/L) was used in combination with alum (300 mg/L), it was found that with the increase in treatment time, better removal of turbidity, TS, TSS, COD, and metals was observed as compared to S-EPS and alum treatment. The value of turbidity decreases from 26 NTU to 12.2 NTU after 12 h of treatment. Similarly, the concentration of SS, TS, and COD also decreased from 19.34, 214.13, and 190 mg/L to 6.99, 90.43, and 132.9 mg/L respectively after 12 h of treatment time. It was also observed that a significant amount of metal removal was achieved with an increase in treatment time (Figure 3(d), 3(e), and 3(f)). The removal of Al, Na, Mg, K, and Ca was increased from 45.32, 68.01, 63.12, 77.06% (30 min of treatment time) and 78.77 to 72.15, 83.14, 80.34, 87.12, and 85.46%, respectively (12h of treatment time).



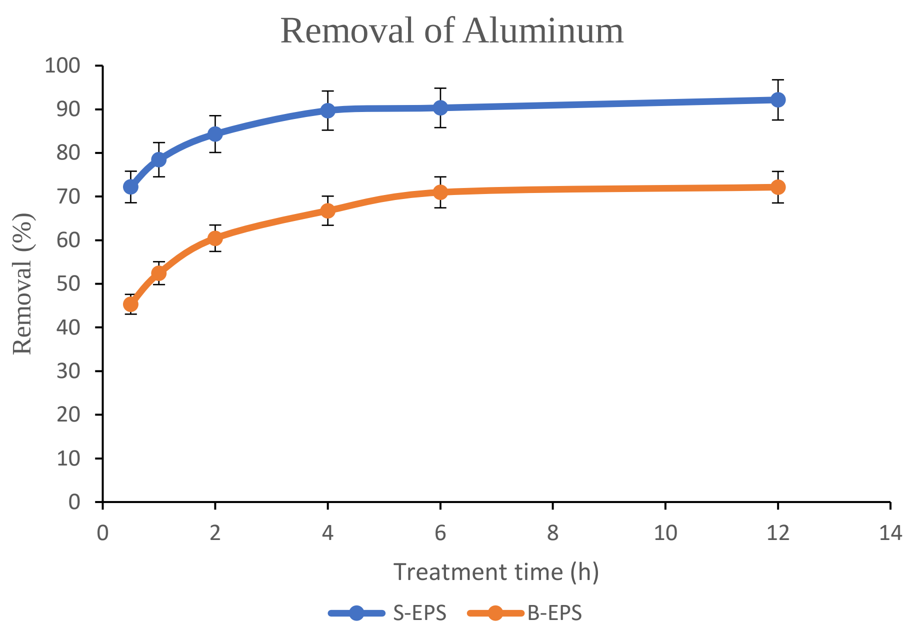
a)


Total solids removal
Removal (%)
100.00
80.00
60.00
40.00
20.00
0.00
0
2
4
6
8
10
12
14
Treatment time (h)
S-EPS+Alum
B-EPS+Alum

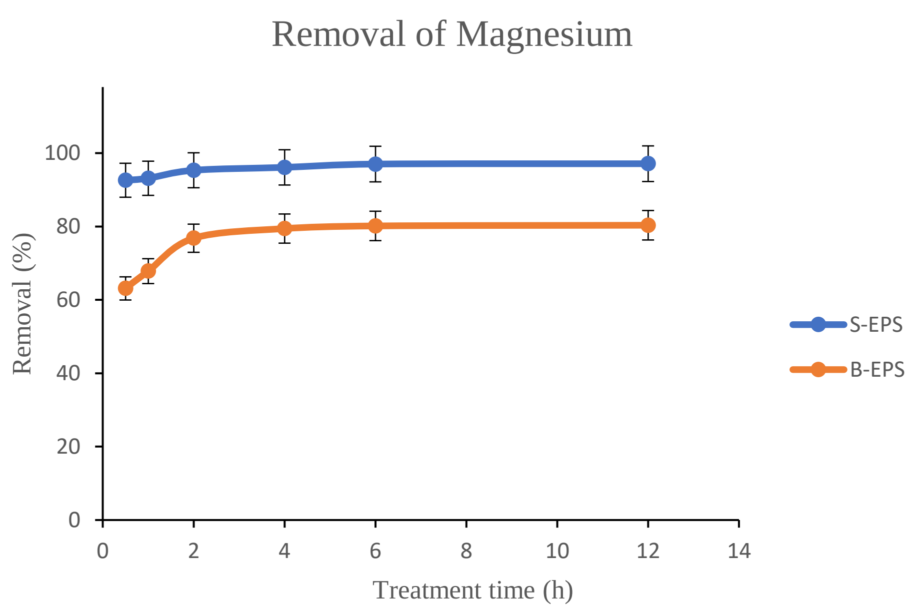
b)


COD removal
Removal (%)
100
90
80
70
60
50
40
30
20
10
0
0
2
4
6
8
10
12
14
Treatment time (h)
S-EPS+Alum
B-EPS+Alum

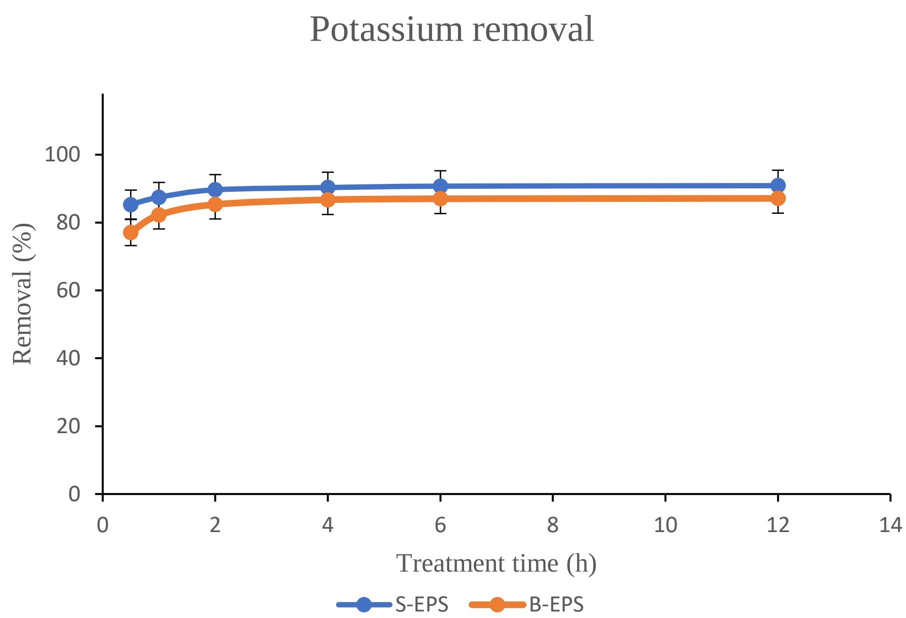
c)


Removal of Aluminum
100
90
80
70
60
50
40
30
20
10
0
Removal (%)
0
2
4
6
8
10
12
14
Treatment time (h)
S-EPS
B-EPS

d)


Removal of Magnesium
100
80
60
40
20
0
Removal (%)
0
2
4
6
8
10
12
14
Treatment time (h)
S-EPS
B-EPS

e)



f)

Figure 4.3: Removal of pollutants using B-EPS and S-EPS: (a) Turbidity removal, (b) TS removal, (c) COD removal, (d) Aluminum removal, (e) Magnesium removal, (f) Potassium removal

Comparison with previous studies

To the best of our knowledge this is the first time that a process using EPS has been used for the treatment of LWW. However, EPS produced by different bacterial strains has been used as a bio-flocculent to treat landfill leachates. We compare below the present study with other studies mentioned in the literature using EPS for the treatment of composting leachate and landfill leachate.

For this study, we used EPS produced by the bacterial strain BR04. The S-EPS (0.6g/L) when combined with alum (0.3 g/L) at pH 7 gave the best results for LWW treatment after 30 minutes. The removal efficiency for turbidity, SS, COD and TS were recorded as 98%, 95.42%, 83.08% and 65.57%, respectively. COD removal efficiency increased to 88% after the treatment time of 4h.

By comparison, Rajwinder et al., 2019 (Kaur *et al.*, 2019) used EPS (produced by bacterial strain BS04) in order to treat composting leachate. Up to 69% of COD, 92% of phosphorus, 65.7% of ammonia and 63.32% of colour could be removed simultaneously removed from landfill leachate using a combination of S-EPS (0.5g/L) and $FeSO_4$ (2g/L) at pH 8 for a treatment time of 8h.

Another study carried out by Adama et al., 2022 (Ndao *et al.*, 2022) used EPS (obtained from bacterial strain BS04) for the treatment landfill leachate. The best results from this study were obtained when S-EPS (0.015g/L) and $FeSO_4$ (2g/L) were used together at pH 8 for 2h. 83% COD, 50% Phosphorus, 44% nitrogen and 64-79% of metal removal were obtained.

The discrepancy of these results can be attributed to two main reasons: (i) the type of wastewater having different initial characteristics; (ii) the EPS is produced by different bacterial strains.

4.6. Conclusion

This work concludes that the biopolymer (EPS) produced from the bacterial fermentation process using BS-04 bacterial strain and waste streams (crude glycerol and secondary sludge from pulp and paper mill) can be used for the treatment of commercial laundry wastewater. Out of the three common chemical coagulants (Alum, $FeSO_4$, and $CaCl_2$), alum (2000 mg/L) achieved the highest flocculation activity (99.18 %) and pollutant removal rates (SS-98%, TS- 91.33%, COD-85.1 %) for CLWW treatment at pH 7 and treatment time of 30 min. It was observed that S-EPS (0.6 g/L) gave better FA than B-EPS. S-EPS alone can remove 83.20% of turbidity, 77.69% SS, and 76.37% COD. When S-EPS (0.6 g/L) was used in combination with alum (300 mg/L) at pH 7 and a treatment time of 30 min, the removal of turbidity, SS, and COD reached 98.00%, 95.42%, and 83.08%, respectively. When treatment time has been increased to 4h, it resulted in more than 88.00% COD removal) from CLWW.

4.7. Acknowledgement

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Author Contributions

Sushil Kumar, Ali Khosravanipour Mostafazadeh, Patrick Drogui, and R D Tyagi conceived and planned the idea of the study. Sushil Kumar experimented and collected the data. All authors carried out the analysis and contributed to the interpretation of the results. Sushil Kumar wrote the manuscript in consultation with Ali Khosravanipour Mostafazadeh, Patrick Drogui, and R D Tyagi. All authors read and approved the final manuscript.

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Availability of data and materials

All data generated or analyzed during this study are included in this published article.

Decalarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

All authors read and approved the final manuscript and mutually agreed that it should be submitted to Environmental System Research.

Competing interests

The authors declare that they have no conflicts of interest to declare.

5. EC TREATMENT OF LWW USING PACKED BED REACTORS WITH SACRIFICIAL ANODES

5.1 Article Introduction

Article's title	Application of Central Composite Design for Commercial Laundry Wastewater Treatment by Packed bed Electrocoagulation using sacrificial iron electrodes
Names of the authors and their professional affiliations	Sushil Kumar[1], Patrick Drogui[1], R D Tyagi[2,3] 1. Institut National de la Recherche Scientifique (INRS), Centre-Eau Terre Environnement (ETE), Université du Québec, 490 Rue de la Couronne, Québec (QC), Canada G1K 9A9 2. BOSK-Bioproducts, 100-399 rue jacquard, Quebec (QC) G1N4J6 3. Distinguished Professor, School of Technology, Huzhou University, China.
Title of the journal	Process Safety and Environmental Protection
Status of the paper	Submitted
Date of submission, acceptance, or publication	Not applicable
DOI	Not applicable

5.2 Preface

This paper, based on using PBEC for the treatment of LWW is related to the preceding paper on the use of EPS as a bio-flocculant for the LWW treatment in the context of addressing the common concern of wastewater management from commercial laundries.

Both studies share a common starting point, acknowledging the challenge of managing and treating wastewater from commercial laundries. The previous study proposes an eco-friendly solution using bio-flocculant EPS derived from crude glycerol and paper mill sludge. It evaluates the effectiveness of different types of EPS, including Slime EPS (S-EPS) and Broth EPS (B-EPS), in treating CLWW. Additionally, it compares the performance of bio-flocculation with EPS against conventional chemical coagulation methods.

In contrast, this study presents an alternative environmentally friendly solution to treat LWW by employing PBEC. The electrochemical reactor utilizes stainless steel and graphite electrodes, along with sacrificial iron electrodes, to achieve effective removal of turbidity, SS, TS, and COD from the wastewater.

The relation between the two studies lies in their common objective of providing sustainable and eco-friendly solutions for the treatment of wastewater from commercial laundries. While the first study focuses on bio-flocculation with EPS, the second study explores EC with a specific emphasis on the use of sacrificial iron electrodes. Both studies contribute valuable insights and methodologies to the broader goal of addressing the environmental impact of commercial laundry wastewater.

5.3 Abstract

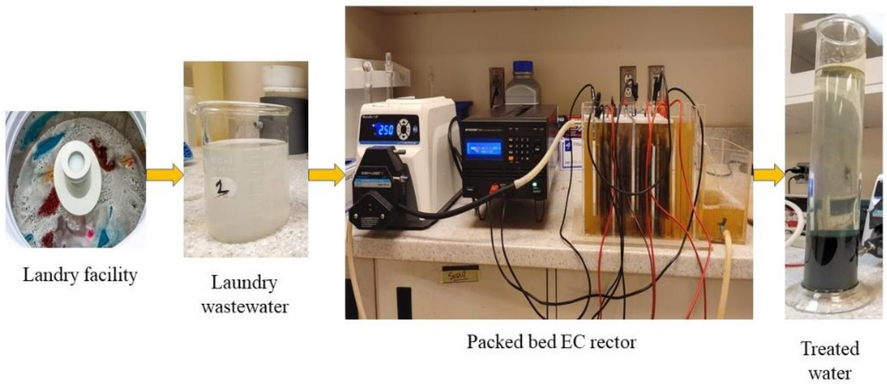

Figure 5.1: Graphical abstract

This research paper presents a novel method utilizing packed bed electrocoagulation (PBEC) packed with sacrificial iron electrodes, and synthetic polymer/extracellular polymeric substances (EPS) as flocculants, statistically optimized via factorial design (FD) and central composite design (CCD) for treating commercial laundry wastewater (LWW) in Quebec, Canada. The study employs stainless steel cathodes, graphite anodes, and scrap iron pieces as sacrificial electrodes, ensuring efficient treatment in dynamic batch mode operation with enhanced contact time facilitated by serpentine flow. The initial characteristics of LWW was COD 579 ± 30 mg/L, TSS of 60 ± 10 mg/L, TS of 622 ± 20 mg/L, turbidity of 110 ± 5 NTU, pH of 9 ± 0.5, NPEOs of 570 ±150 µg/L and conductivity of 494 ± 20 mS/cm. The results demonstrate effective removal of turbidity (98 ± 2%), TS (95 ± 3%), COD (89 ± 5%), and NPEOs (53 ± 2%) under optimized current intensity: 2.99A, treatment time: 58.8 minutes and enhanced EPS dose from 5.8 mg/L to 8.0 mg/L. The economic feasibility analysis reveals energy consumption as the primary expenditure, with a treatment cost of 1.20$CAN/m^3. This research introduces sustainable treatment for commercial LWW, meeting Quebec's reuse standards, implying reuse potential and responsible wastewater management.

Keywords: Commercial laundry wastewater, wastewater treatment, packed bed electrocoagulation, response surface methodology, environmentally friendly.

5.4 Introduction

Laundry facilities generate substantial amounts of wastewater containing a complex mixture of organic and inorganic contaminants, posing ecological and human health risks if left untreated (Kumar *et al.*, 2023). This issue has garnered significant attention in the context of escalating concerns surrounding water pollution and the imperative for sustainable wastewater management solutions. Particularly, the discharge of untreated or inadequately treated laundry wastewater (LWW) into aquatic systems necessitates effective treatment methods to mitigate environmental contamination (Akarsu & Deniz, 2021). The diverse composition of LWW, including detergents, suspended solids, organic compounds, oils, and fats, underscores the need for comprehensive analysis and treatment strategies tailored to its constituents (Samadikun *et al.*, 2021). Understanding the interactions and concentrations of these constituents guides the design of appropriate treatment technologies to ensure effective removal or reduction of contaminants before discharge, emphasizing the importance of responsible wastewater management (Li *et al.*, 2022a).

Electrocoagulation (EC) has emerged as a promising technology for LWW treatment, leveraging controlled electrochemical reactions to generate coagulant species that facilitate the agglomeration and subsequent precipitation of contaminants (Kumar *et al.*, 2022a). In the context of LWW treatment, EC offers several advantages, effectively removing suspended solids, oils, organic compounds, and even heavy metals and colorants (Corona *et al.*, 2021; Oktiawan *et al.*, 2021; Veli *et al.*, 2021). However, EC exhibits some drawbacks too that include elevated energy consumption, sludge generation, inefficiency for certain contaminants, and susceptibility to electrode fouling (Elazzouzi *et al.*, 2018; Nidheesh *et al.*, 2020; Yasri *et al.*, 2020). Therefore, there is a need of advanced processes such as packed bed electrocoagulation (PBEC) that may offer enhanced contaminant removal, reduced energy demands, diminished sludge formation, and prolonged electrode durability (Holt *et al.*, 2005; Yasri *et al.*, 2020). Synthetic polymers have conventionally been utilized as flocculants in EC systems (Lin *et al.*, 2019). Yet, their environmental drawbacks, rising operational costs, and the formation of stable compounds hindering the process advocate for exploring alternatives such as extra polymeric substances (EPS) (Drogui *et al.*, 2022; Janpoor *et al.*, 2011). EPS are superior to synthetic polymers in electrocoagulation due to their natural origin, higher biodegradability, stronger floc formation, and lower cost, providing both environmental and economic benefits (Kumar *et al.*, 2023; Liu *et al.*, 2022; Nouha *et al.*, 2016).

This research paper presents an innovative method based on PBEC utilizing sacrificial iron electrodes and incorporating EPS as an alternative to synthetic polymers. The novelty of this investigation lies in combining these innovative techniques with factorial deign (FD) and central composite design (CCD) to statistically optimize various process parameters. This study conducts a thorough analysis of the cost and energy requirements of the entire process to assess its economic feasibility. By increasing the surface area through the packing of sacrificial iron electrodes, passivation is significantly reduced, leading to longer intervals between electrode regeneration cycles. This methodology enables prolonged operation of the initial electrode setup, facilitating the treatment of large volumes of water in shorter timeframes. This research optimizes statistically the process parameters using FD and CCD in PBEC for commercial LWW treatment. The utilization of packed sacrificial iron electrodes and replacement of synthetic polymers with EPS as flocculants, the study suggests a more sustainable and efficient approach for commercial LWW treatment.

5.5 Materials and Methodology

EC packed bed reactor design:

The PBEC reactor (figure 2) consists of a systematic arrangement to optimize contaminant removal from commercial LWW. The 4.5 L Plexiglass reactor consists of 5 compartments, each compartment is separated by a plexiglass sheet and arranged in such a way that it can provide serpentine flow in the reactor. The serpentine flow provides better contact time with the in situ generated coagulants and pollutants present in LWW. The four compartments consist of stainless steel (SS) (surface area: 110 cm^2) as cathode and graphite (surface area: 110 cm^2) as anode. In each compartment, vertically oriented electrodes are strategically placed, inducing the formation of an electric field as the contaminated water passes through the packed bed. This electric field initiates electrochemical reactions, culminating in the production of metal cations that serve as coagulants.

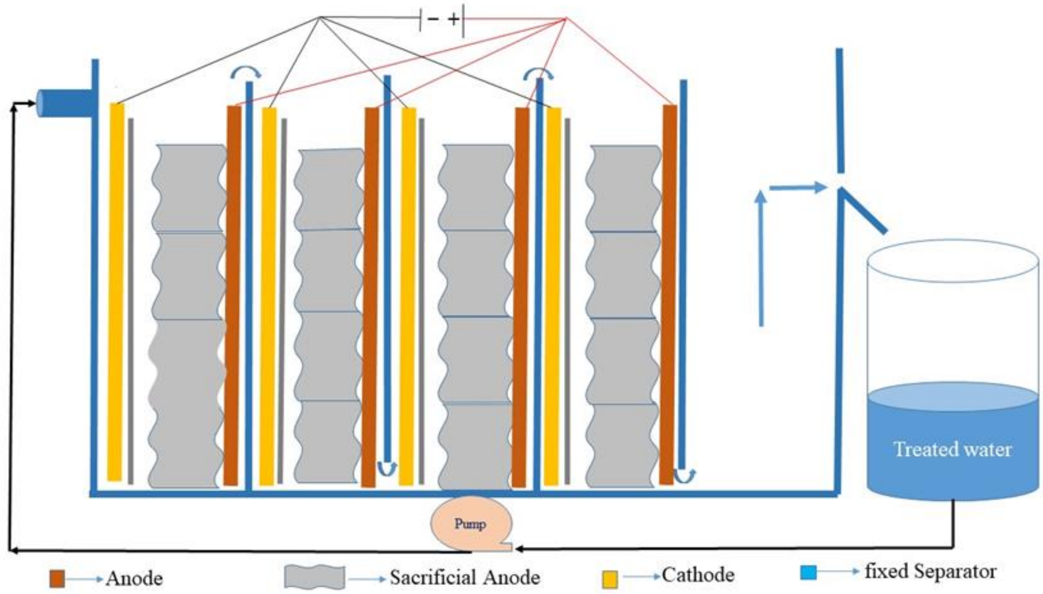

Figure 5.2: Packed bed electrocoagulation (PBEC) rector design

Sacrificial iron scrap packing has been done in between cathode and anode to promote coagulation and flocculation. To avoid any short-circuiting sacrificial iron scraps has been separated from SS cathode by a perforated plexiglass sheet. The electrodes are interconnected in parallel to a current source, employing a direct current (DC) generated through a DC generator. The packing material optimizes the interaction between the electrodes and water, augmenting effective particle destabilization and aggregation. These resulting flocs are trapped within the packed bed, while the treated water undergoes recirculation within the reactor, ensuring a dynamic batch mode. The reaction mechanism involved during this EC process has been shown in Equations 4-6 below.

At anode:

$Fe_{(s)} \longrightarrow Fe^{2+}{}_{(aq.)} + 2e^-$ ---------------------- **Equation 4**

In the solution:

$Fe^{2+}{}_{(aq.)} + 2OH^-{}_{(aq.)} \longrightarrow Fe(OH)^2{}_{(s)}$ -------------------------- **Equation 5**

At Cathode:

$2H_2O_{(l)} + 2e^- \longrightarrow H_{2(g)} + 2OH_{(aq.)}$ -------------------------- **Equation 6**

LWW Sampling and characterization:

The samples of LWW were collected from a commercial laundry facility situated in Montreal, Canada. The sample was collected from outlet point of commercial laundry in 200 L container and transported to the experiment location (INRS laboratory). The 200 L of LWW was divided into 10 buckets, each containing 20 L, and subsequently stored in a cold room at a temperature of -4℃ for further experimentation.

The collected samples were then analyzed to assess parameters such as pH, turbidity, Conductivity, COD, TSS, TS, NPEO, nitrate, phosphate and metals. These data aids in understanding the wastewater's characteristics, which is crucial for designing effective treatment methods.

Response Surface Methodology (RSM)

Experiment plan and statistical optimizations were done using response surface methodology (Design expert-10). Response Surface Methodology entails employing regression analysis to establish a link between a response variable and controlled variations in independent factors. It generates numerous experimental parameter permutations, enhancing laboratory test efficiency. Utilizing estimated parameters, it becomes possible to identify the pivotal variable exerting the most influence on the predicted outcome. Consequently, researchers can prioritize variables crucial to product acceptance (Rao *et al.*, 2019).

Three distinct operational parameters, namely current intensity (X_1), treatment time (X_2), and polymer concentration (X_3), were identified as critical for PBEC in this study. The response variables selected were turbidity, COD, and TSS removal.

To conduct the FD, a matrix of 8 experiments was constructed ($2^k = 8$, where k represents the number of factors). Each factor was set at two levels (-1 and +1), as outlined in Table 1. Additionally, the CCD matrix was created, comprising the FD matrix (8 experiments) with an additional 12 assays.

Table 5.1 Experimental variables and their range

Coded Variables	Factor	Range		$U_{1,0}$	ΔU_1
		Minimum value (-1)	Maximum value (+1)		
X_1	U_1: Intensity (A)	1	3	2	1
X_2	U_2: Time (min)	20	60	40	20
X_3	U_3: Polymer concentration (mg/L)	2	6	4	2

Combining Packed Bed Electrocoagulation with EPS

The study involves the utilization of EPS derived from bacteria *BS-04* to augment the treatment efficacy of PBEC in handling LWW (Kumar *et al.*, 2022a). The experimental design includes the replacement of conventional synthetic polymers with varying concentrations of EPS within the PBEC system. Characterized EPS, obtained through established cultivation and extraction methods (Kaur *et al.*, 2019), is integrated into the PBEC setup. Results are statistically analyzed to elucidate the impact of EPS substitution on PBEC performance. The findings aim to provide insights into the viability of EPS as an alternative to conventional polymers in enhancing PBEC's effectiveness in treating commercial LWW.

Analytical methods

The pH measurement was conducted utilizing a pH-meter (Fisher Acumet model 915) furnished with a double junction Cole-Palmer electrode featuring an Ag/AgCl reference cell. Likewise, conductivity was assessed using a conductivity meter (Oakton Model 510). During the study COD, TSS, TS and $NPEO_s$ were measured by using standard method prescribed by CEAEQ (Center of Expertise in Environmental Analysis of Quebec) (l'Environnement & climatiques, 2014). Metals were measured by ICP-AES method proposed by CEAEQ.

Passivation studies

To investigate passivation phenomena, we conducted experiments to measure the variation of potential (voltage) in two different electrochemical systems: PBEC and EC without packing. The PBEC setup involved electrodes surrounded by a packed bed of sacrificial iron, while the EC without packing had exposed electrodes without any surrounding material. The experiments were conducted under controlled conditions to monitor the changes in potential over time.

Economic estimate

The energy utilized was calculated at a rate of 0.06\$CAN/KWh, aligning with the rate established by the province of Quebec (Canada) (Kumar *et al.*, 2023). Additionally, expenses associated with the process encompassed the cost of EPS and electrodes. The cost of EPS production was calculated at a rate of 0.95\$CAN/L (Kumar *et al.*, 2020) and the cost of electrode material was calculated at a rate of 0.3\$CAN/Kg (Yasri *et al.*, 2020). The overall expenditure was assessed in terms of Canadian dollars spent per cubic meter of treated LWW (\$CAN/m^3).

5.6 Results and discussion

LWW Characteristics

The analysis of LWW reveals a comprehensive profile of its composition, as shown in Table 2 and its potential environmental implications.

Table 5.2 Initial characteristics of the LWW

Parameters	Average Values	Recommendation for effluent discharge in the sewer (Ville de Quebec)	Recommendation for reuse in the laundries
Turbidity (NTU)	110 ± 5	<10	<2
pH	10 ± 0.5	6.5-8.5	6.5-8.5
Conductivity (µS/cm)	494 ± 20	n.r.	<2000
Total Suspended solids (mg/L)	60 ± 10	<30	<5
Total solids (mg/L)	622 ± 20	<100	<500
COD (mg/L)	579 ± 30	<150	<100
NPEOs (µg/L)	570 ± 150	<200	<25
Sulphate (mg/L)	22± 5	n.r.	<250
Sulphite (mg/L)	0.2 ± 0.05	n.r.	n.r.
Nitrate (mg/L)	4 ± 1	<10	n.r.
Nitrite (mg/L)	1.5 ± 0.5	<1	<1
Ammoniacal nitrogen (mg/L)	2.3 ± 1	<10	n.r.
Total nitrogen (mg/L)	9.7 ± 0.5	<10	<10

Phosphate (mg/L)	21 ± 2.0	<1	n.r.
Aluminium (mg/L)	0.096 ± 0.02	n.r.	n.r.
Calcium (mg/L)	18.6 ± 2.0	n.r.	n.r.
Magnesium (mg/L)	3.87 ± 0.5	n.r.	n.r.
Sodium (mg/L)	104 ± 10	n.r.	n.r.
Potassium (mg/L)	3.76 ± .5	n.r.	<50
Sulphur (mg/L)	20.6 ± 2.0	n.r.	n.r.
Zinc (mg/L)	0.42 ± 0.15	<2	n.r.
Copper (mg/L)	0.03 ± 0.01	<1	n.r.
Iron (mg/L)	0.23 ± 0.2	<1	<0.3
Manganese (mg/L)	0.019 ± 0.005	<0.1	n.r.
Lead (mg/L)	<0.003	<0.1	<0.01
References	This study	(Cárdenas *et al.*, 2023; Zhang *et al.*, 2022)	(Exall *et al.*, 2006; Van Rossum, 2020)

A turbidity reading of 110 ± 5 NTU suggests a significant concentration of suspended solids. Such high levels can obstruct light penetration in water bodies, potentially affecting photosynthesis and aquatic ecosystems (Trigueros *et al.*, 2023). The alkaline nature of the wastewater, with a pH of 10.38 ± 0.5, signifies potential alterations in natural pH levels, posing a threat to aquatic life if discharged in the water bodies without treatment (Middelboe & Hansen, 2007). Elevated conductivity at 494 ± 20 µS/cm suggests the prevalence of dissolved salts and ions, which, at heightened levels, can negatively impact freshwater organisms (French *et al.*, 2010). COD measures the oxygen required for oxidizing organic and inorganic matter, indicating a COD of 579 ± 30 mg/L. This high COD implies a significant presence of organic compounds, that may potentially deplete oxygen levels in water bodies and endangering the aquatic life (Sharma & Gupta, 2014). The presence of various nutrients such as nitrate, nitrite, ammoniacal nitrogen, total nitrogen, and phosphate underscores the risk of eutrophication. Elevated nutrient levels can spur excessive plant growth, leading to oxygen depletion and harming aquatic organisms (Zhu *et al.*, 2010). Metal concentrations, including aluminum, zinc, copper, iron, and manganese, even in trace amounts, pose risks to aquatic life and can bioaccumulate in the food chain (Tekin-Özan, 2008). Furthermore, sulfate, sulfite, and sulfur levels in the wastewater can affect water taste and quality, potentially generating harmful by-products (Fernando *et al.*, 2018). Although lead concentration is minimal (<0.003 mg/L), any trace of lead in water is concerning due to its toxicity and adverse health effects (Lytle *et al.*, 2021). Therefore, effective LWW treatment can reduce the environmental impact, safeguarding water ecosystems and human health from potential hazards associated with LWW.

Effect of operating parameters

The influence of various factors, current intensity (X_1), treatment time (X_2), and polymer concentration (X_3), on commercial LWW treatment was examined through FD methodology. Each factor was set at two levels, denoted as minimum and maximum, and normalized to (-1) and (+1). The experimental region and corresponding coded values are outlined in Table 1. The factorial matrix generated consisted of 8 experiments, focusing on three responses: turbidity, TSS and COD removal. A summary of the experimental plan and corresponding results can be found in Table 3.

Table 5.3 Factorial design matrix and response

Assays	Experiment design			Plan of experiments			Results (removal %)		
	X_1	X_2	X_3	U_1 (A)	U_2 (min)	U_3 (mg/L)	Turbidity	TSS	COD
1	-1	-1	-1	1	20	2	36.6	36.8	24.2
2	1	-1	-1	3	20	2	62.5	56.8	51.2
3	-1	1	-1	1	60	2	44.1	38.2	40.7
4	1	1	-1	3	60	2	76.8	74.6	68.5
5	-1	-1	1	1	20	6	38.2	36.2	26.3
6	1	-1	1	3	20	6	68.5	62.4	50.4
7	-1	1	1	1	60	6	64.4	54.8	56.4
8	1	1	1	3	60	6	97.4	93.2	87.7

From the factorial design three mathematical models were obtained for three different responses.

$Y_1 = 61.06 + 15.24X_1 + 9.61X_2 + 6.06X_3 + 1.19X_1X_2$ **Equation 7**

$Y_2 = 56.63 + 15.13\,X_1 + 8.58X_2 + 3.57X_1X_2$ **Equation 8**

$Y_3 = 50.68 + 13.78\,X_1 + 12.65X_2 + X_1X_2$ **Equation 9**

Where Y_1 is experimental response for turbidity removal, Y_2 is experimental response for TSS removal and Y_3 is experimental response for COD removal. The Pareto graphic analysis (Figure 3) visually illustrates the relative magnitudes of the main effects and interactions, providing a comprehensive overview of their contributions to the removal efficiencies of Y_1, Y_2, and Y_3.

The analysis revealed insights into the interaction effect of X_1X_2 between current intensity (X_1) and treatment time (X_2) on pollutant removal efficiency. While the X_1X_2 interaction had a negligible impact (less than 1%) on turbidity (Y_1) and COD removal (Y_3), it significantly influenced TSS removal (Y_2), contributing to 9%. Despite its influence on TSS removal, the main effects of X_1 and X_2 remained crucial, as they had the highest contributions (64.02% and 25.46%, respectively) to pollutant removal. This highlights the importance of optimizing current intensity and treatment time for effective LWW treatment via PBEC.

To assess the significance of these models, Analysis of Variance (ANOVA) was conducted using the Design Expert software. The resulting correlation coefficients (R^2) for the proposed models were determined to be 0.95 for turbidity removal, 0.88 for TSS removal, and 0.90 for COD removal, respectively.

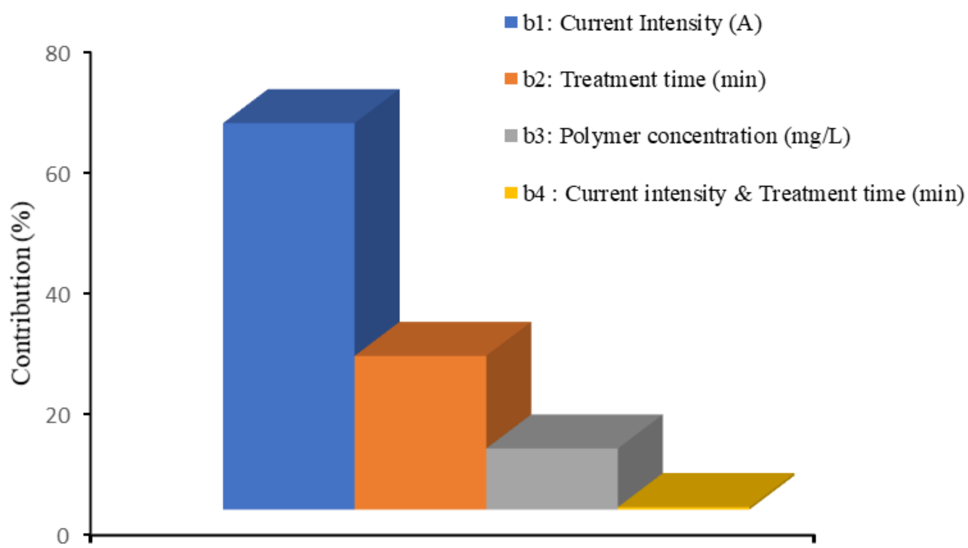

Figure 5.3: Graphical Pareto analysis of the effect of current intensity, treatment time and polymer concentration on pollutant removal

Optimization of commercial LWW treatment using CCD methodology:

As previously mentioned, three distinct operational parameters, namely current intensity (X_1), treatment time (X_2), and polymer concentration (X_3), were selected as pivotal factors in the treatment process of commercial LWW. To achieve optimization, a CCD methodology was employed. The CCD matrix consisted of 20 experiments, encompassing the FD matrix, which comprised of 8 assays.

Table 5.4: Central composite design matrix

Assays	Experiment design			Plan of experiments			Results (removal %)		
	X_1	X_2	X_3	U_1 (A)	U_2 (min)	U_3 (mg/L)	Turbidity	TSS	COD
1	-1.68	0	0	0.32	40	4	22.3	18.4	18.2
2	1.68	0	0	3.68	40	4	90.7	88.5	87.4
3	0	-1.68	0	2	6.4	4	10.2	7.8	8.2
4	0	1.68	0	2	73.6	4	72.6	66.7	68.4
5	0	0	-1.68	2	40	0.64	44.6	40.4	38.9
6	0	0	1.68	2	40	7.36	63.6	61.2	58.6
7	0	0	0	2	40	4	67	59.8	58.4
8	0	0	0	2	40	4	66.6	67.2	62.3
9	0	0	0	2	40	4	66.6	64.5	60.9
10	0	0	0	2	40	4	67.5	62.1	64.2
11	0	0	0	2	40	4	63.6	62.5	58.7
12	0	0	0	2	40	4	64.9	59.4	56.7

$$Y_1 = 65.67 + 17.35X_1 + 13.32X_2 + 5.89X_3 + 1.19X_1X_2 + 0.59X_1X_3 + 4.16X_2X_3 - 0.98 (X_1)^2 - 6.32 (X_2)^2 - 1.83(X_3)^2$$ **Equation 10**

$Y_2 = 62.25 + 17.49X_1 + 12.28X_2 + 5.51X_3 + 3.58X_1X_2 + 1.03X_1X_3 + 3.78X_2X_3 - 1.04 (X_1)^2 - 6.76 (X_2)^2 - 1.97(X_3)^2$ **Equation 11**

$Y_3 = 60.04 + 16.59X_1 + 14.82X_2 + 5.08X_3 + X_1X_2 + 0.07X_1X_3 + 4.2X_2X_3 - 1.59 (X_1)^2 - 6.71 (X_2)^2 - 3.02(X_3)^2$ **Equation 12**

Table 5.5*:* Lack of fit tests summary for different responses

Response	Source	Sum of squares	*df*	Mean square	*F* value	*p*-Value Prob > *F*	
	Linear	1513.33	11	137.58	62.92	0.0001	Suggested
	2FI	1360.67	8	170.08	77.78	< 0.0001	
Y_1	Quadratic	763.41	5	152.68	69.82	0.0001	
	Cubic	409.83	1	409.83	187.42	< 0.0001	Aliased
	Pure error	10.93	5	2.19			
	Linear	1633.95	11	148.54	17.23	0.0028	Suggested
	2FI	1409.29	8	176.16	20.43	0.0021	
Y_2	Quadratic	724.53	5	144.91	16.81	0.0038	
	Cubic	345	1	345	40.02	0.0015	Aliased
	Pure error	43.11	5	8.62			
	Linear	1217.85	11	110.71	14.33	0.0043	
	2FI	1068.68	8	133.59	17.29	0.003	
Y_3	Quadratic	331.52	5	66.3	8.58	0.017	Suggested
	Cubic	76.13	1	76.13	9.85	0.0257	Aliased
	Pure error	38.64	5	7.73			

Lack of Fit testing, which is central to the testing process, provides a detailed examination of the suitability of regression models in clarifying the complex relationships between response variables. The results of Lack of Fit testing were examined for the responses (Y_1, Y_2, Y_3):

Y_1* *(Turbidity Removal): p-values for the linear, 2FI, and quadratic terms are low (< 0.005), highlighting their statistical significance in describing the turbidity removal model. This indicates that the linear, interactions, and quadratic terms play a significant role of the observed changes in turbidity removal. The low p-value of quadratic model (0.0001) suggests that it has a very significant impact in explaining the changes in turbidity removal.

Y_2* *(TSS Removal): Similar to Y_1, the linear, 2FI, and quadratic terms exhibit low p-values (< 0.005), emphasizing their statistical significance in describing TSS removal in this which is important in linear interaction quadratic terms that will affect TSS extraction efficiency to determine the activity. Notably, the low p-value (0.0038) for the quadratic model indicates its significant effect in the TSS removal model.

Y_3* *(COD Removal): Like Y_1 and Y_2, the linear, 2FI, and quadratic terms for Y_3 exhibit low p-values (< 0.005), indicating statistical significance for COD removal description These terms are very helpful in predicting COD removal. However, the high p-value of the quadratic term (0.017) compared to the other models indicates that the effect of the quadratic term is weaker in explaining the variation in COD removal.

Furthermore, consistently low p-values due to non-normality (equal error) indicate that the residuals estimated by the models are negligible but high p-values for the cube term is in the case of Y_2 and Y_3 makes it possible to improve the model by reconsidering the role of cube effects in these responses.

The Lack of Fit test validates the significance of linear, 2FI, and quadratic terms in elucidating variations in turbidity, TSS, and COD removal. However, the significance of the cubic term varies among different responses and may be weakly influenced by specific instances. This comprehensive analysis facilitates the refinement and refinement of the model provides insights for more accurate predictions and a better understanding of the commercial LWW treatment process. Prediction models derived from PBEC process for the variables (current intensity, treatment time, and polymer concentration) and the responses (turbidity removal, TSS removal, and COD removal). This equation provides a systematic description of how variations in these parameters affect pollutant removal efficiency in commercial LWW treatment.

Lack of Fit tests summarize the adequacy of the model, which assesses whether the models are adequate to capture the relationship between variables and responses and determine the p-values associated with each term in the models what is important. In this case, the low p-values (<0.05) for the linear, 2FI (second order interaction), and quadratic terms in models Y_1, Y_2, and Y_3 indicate the importance of response control prediction that these terms significantly affect turbidity, TSS, and COD removal efficiency during the treatment process.

High probability values indicate that the terms in the models are not very likely to occur by chance. This indicated the robustness and reliability of the models in explaining variation in responses. In contrast, the marginal probability indicates the importance of the terms, suggesting that these factors primarily affect the effectiveness of treatment.

As such, the high significance of linear, 2FI, and quadratic terms in models indicates that current intensity, treatment time, and polymer concentration play an important role in the effective removal of impurities. Thus, during the treatment process, these variables must be carefully controlled, and the process adjustments must be made.

Additionally, the presence of alias cubes in some cases (Y_1 and Y_3) show interactions between multiple factors that were not explicitly captured by the model, which may require further examination or refinement of the model if their impact is understood together. Ultimately, these findings emphasize the importance of considering and controlling these variables to improve the efficiency. Figure 4 shows the comparison of predicted value and the actual value for turbidity, TSS and COD removal. It is clear from the figure that the predicated value and measured value lies on the same line, hence the results are satisfactory.

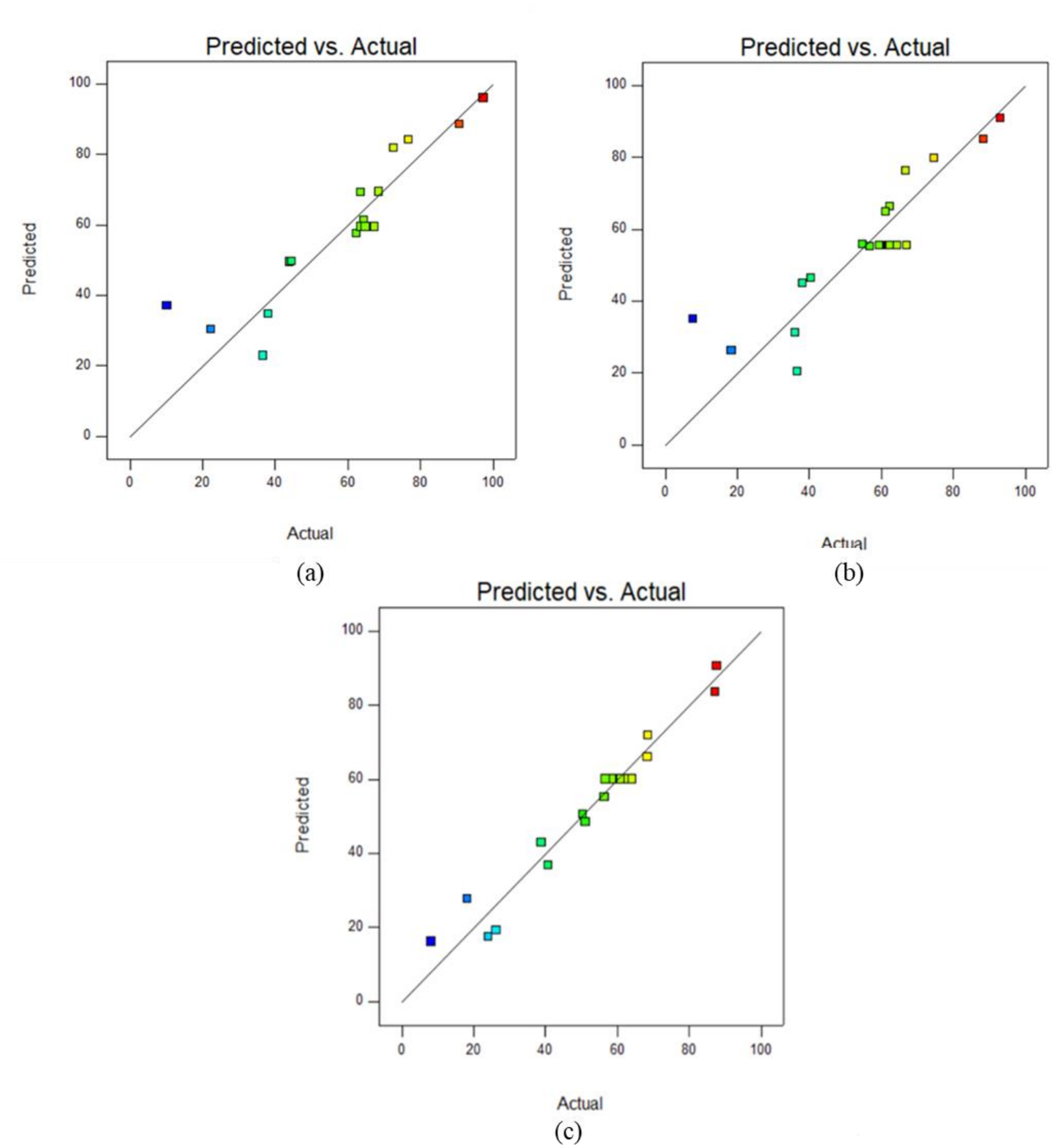

Figure 5.4: Actual value and predicted value of removal of (a) Turbidity, (b)TSS, and (c) COD removal

These results reveal the efficacy of regression models in the treatment, meeting discharge and reuse guidelines, as given in table 2.

Figure 5 shows that with the increase of current intensity and treatment time, the removal of turbidity, TSS and COD increases. Analysis of the 3D model portrayed in Figure 6 unveils a notable escalation in turbidity, TSS and COD removal efficiency as the current intensity varies between 2.25-3A and the treatment time spans from 45-60 minutes. This observation implies a direct correlation between increasing current intensity and treatment time and the improved efficacy of the process in eliminating suspended particles and colloidal matter from LWW, consequently enhancing the overall turbidity removal efficiency. Furthermore, the comparison between actual and predicted values, as depicted in Figure 4, demonstrates a satisfactory alignment, indicating the statistical significance of the quadratic model employed. The maximum turbidity, TSS and COD removal can be seen during 2.25-3A (current intensity) and 45-60 min (treatment time).

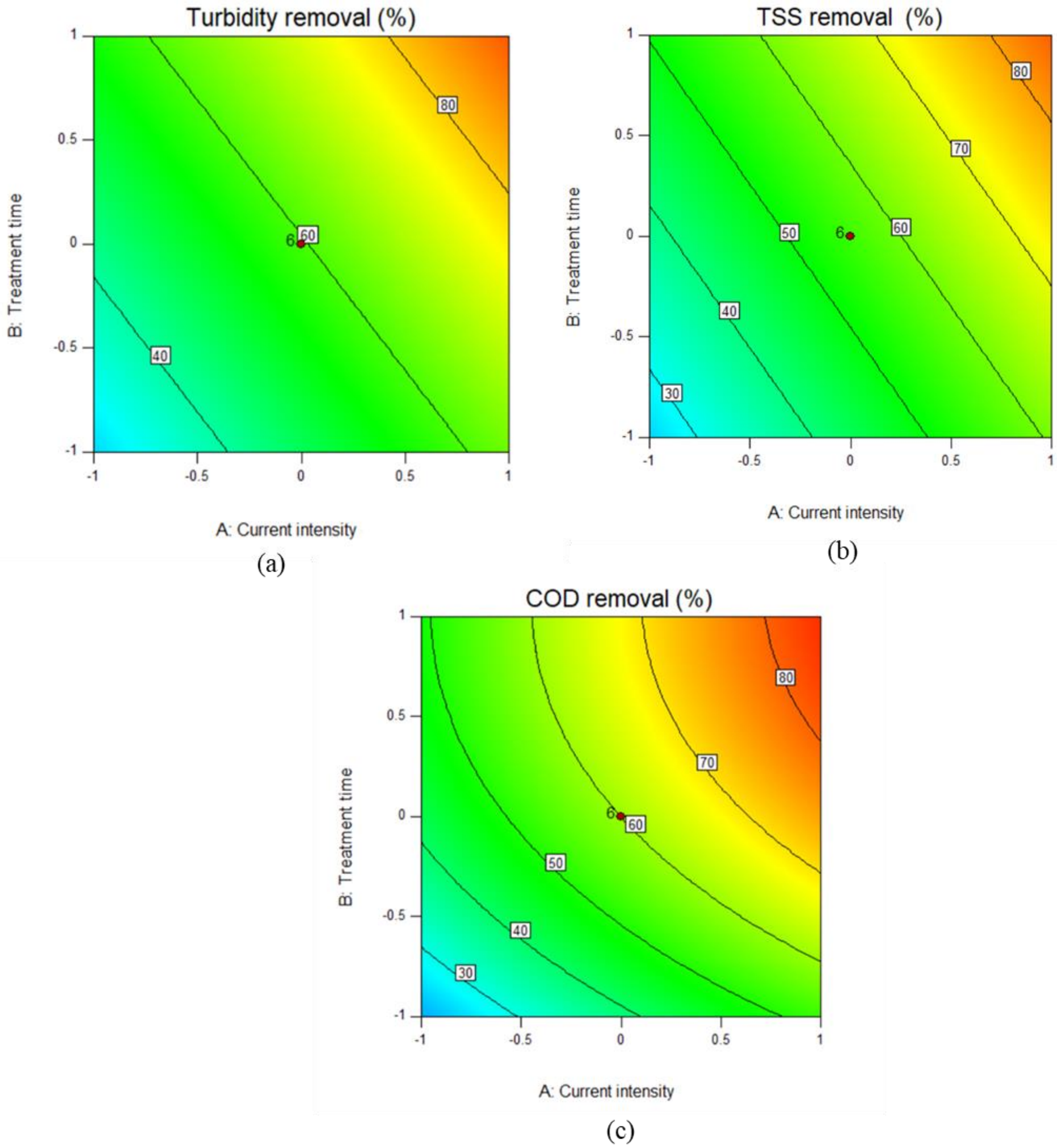

Figure 5.5: (a) Turbidity removal, (b) TSS removal, and (c) COD removal

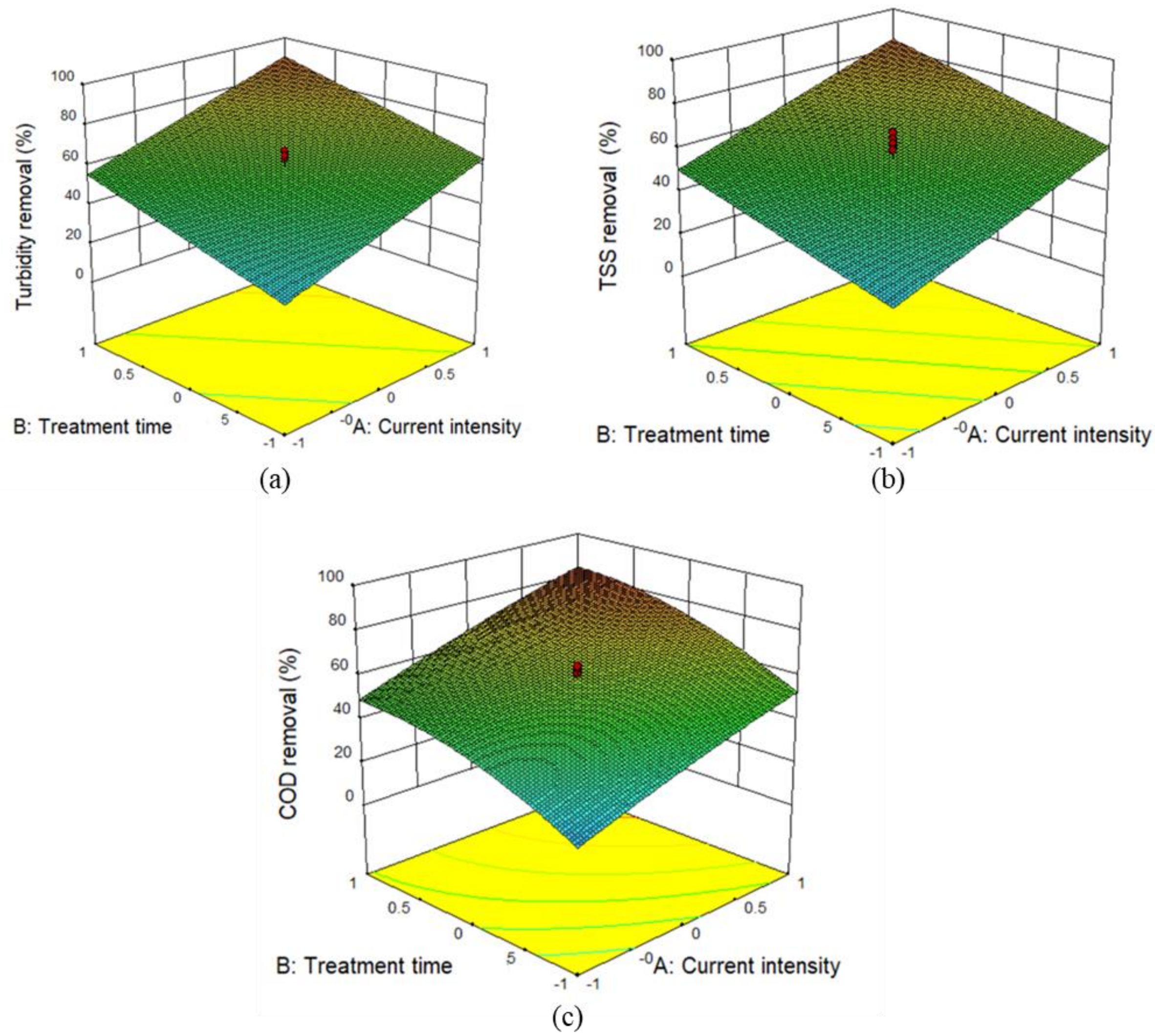

Figure 5.6: 3D profiles: (a) turbidity removal, (b) TSS removal, and (c) COD removal

Optimization using desired criteria:

To rigorously ascertain the optimal conditions for wastewater treatment concerning pollutant removal, it is imperative to consider the energy consumption throughout the process. The criteria chosen for optimizing the treatment of LWW are as follows: (i) Treatment time and current intensity must be maintained within a specified range with a high level of importance (weighting factor of 3/5) to facilitate pollutant removal; (ii) Polymer concentration should be minimized with a high level of importance (weighting factor of 4/5) to mitigate the utilization of synthetic polymer and promote sustainability within the process.

Desirability functions serve as objective functions, exhibiting values ranging from zero beyond the defined limits to one at the targeted goal. The optimization program aims to maximize this desirability function. The pursuit of the objective commences from a randomly chosen starting point, progressing along the steepest ascent to reach a peak. The presence of multiple peaks can arise due to curvature in the response surfaces and their amalgamation within the desirability function. Initiating the process from various points within the design space enhances the likelihood of identifying the most optimal local maximum.

By these experiments, we discovered that the ideal settings are a current intensity of 2.995 A, a treatment time of 58.82 minutes, and a polymer dose of 5.778 mg/L. Under these conditions, the elimination of turbidity, TSS and COD attained to 97.80%, 94.94% and 89.63% respectively with a desirability score of 1. These optimal values were subsequently experimentally validated, resulting in a 97.73% removal of turbidity, 94.75% removal of TSS and 89.26 % removal of COD.

A controlled approach to statistical optimization sets a specific goal for each factor and response variable, and with a variety of possible goals such as maximizing, minimizing, targeting, or resolving in a predetermined range. Each value is assigned a weight, indicating its precise preference function. These desire functions range from zero beyond the defined threshold to one within the target range and guide the optimization process to maximize this functionality.

Given the sharpness of the responses and their consistency, there may be multiple peaks. Starting from multiple points in the design space increases the chances of finding the best local maximum. In optimizing multiple reactions with current strength, treatment time, polymer dose parameters of turbidity, TSS removal and COD were set as 'max' in specific ranges. The objective of this method removal of turbidity, TSS and COD was to identify a viable and cost-effective solution.

Through the process optimization, we discovered conditions that improved the removal of turbidity, TSS, and COD during treatment. The experimentally validated results confirm that the numerical optimization method is robust and reliable in determining the optimal conditions to achieve pollutant removal from water high sewage sample levels. These results highlight the effectiveness of this efficiency method in designing and enhancing wastewater treatment efficiencies for effective pollutant removal.

Removal of compounds, metals and NPEOs:

Quantitative analysis of metals and NPEOs was carried out under at the optimal conditions (Current intensity: 2.995 A, treatment time: 58.82 min and polymer dose: 5.778 mg/L). During the process 84.34% of phosphate, 76.80% of ammonia and 81.32% of sulphate were removed from LWW. Additionally, it was noted that the removal of Al, K, Na, Ca and Mg was also noteworthy. In the study it was also found that PBEC reactor can also remove NPEOs at certain extent (less than 48% removal). It was also observed that the concentration of iron in the supernatant was increased due to dissolution of iron ions from the sacrificial iron packing.

Optimized conditions of current strength, treatment time, and polymer dosage significantly influenced the removal efficiency of various components and chemicals in the wastewater and observed phosphate removal, ammonia, and sulfate are removed with suitable yields of 84.34%, 76.80%, and 81.32%, respectively. Furthermore, the enhanced removal of metals such as Al, Mg, Na, K, and Ca indicated the effectiveness of the process in reducing the metal content. However, the reaction showed limitations in the removal of NPEOs, resulting in removal rates lower than 48%.

An interesting observation was that the dispersion of metal ions from the sacrificial metals increased the concentration of metal surface. This phenomenon indicates a negative effect of the treatment process, suggesting its importance for further research on the potential of metal dispersion and its potential impact on overall treatment efficacy. Overall, these data demonstrate the versatility of the treatment system, highlighting its effectiveness in removing specific pollutants such as phosphate, ammonia, sulfates and some metals but focusing on NPEO controlling removal limitations and unintended consequences of metal dispersion.

Combining PBEC with EPS:

Following the integration of PBEC with EPS (replacing synthetic polymer with biopolymer), there was a decreased in the efficiency of removing turbidity, TSS, and COD at optimized condition (Current intensity: 2.995 A, treatment time: 58.82 min and polymer dose: 5.778 mg/L. To achieve the desired target EPS doses has been increased to 8.0 mg/L. The integration of EPS in place of the synthetic polymer within the PBEC system brought about notable changes in the removal efficiency of various contaminants.

Following this adjustment, a distinct improvement in the removal efficiencies were observed. Turbidity removal reached 97.73%, demonstrating a substantial increase from the initial reduced efficiency. Similarly, TSS removal improved to 94.80%, showcasing a notable enhancement compared to the initial lower performance. The removal of COD also exhibited an increase to 89.63%, indicating an improvement from the reduced efficacy observed after the integration of EPS. Moreover, the removal efficiency of NPEOs notably increased to 53.6% after the adjustment in EPS dosage. This elevation in NPEOs removal suggests that the modification in the EPS dosage positively impacted the system's ability to mitigate these specific contaminants.

These results underscore the critical role of EPS dosage in influencing the efficiency of the PBEC system. The increased EPS dosage led to significant enhancements in the removal of turbidity, TSS, COD, and NPEOs, effectively addressing the decreased efficiency observed previously. This emphasizes the importance of optimizing operational parameters in electrochemical treatment systems to achieve desired treatment goals and improve contaminant removal efficiencies.

Passivation Studies

Our findings indicate notable differences in the potential variation between the PBEC and EC without packing configurations (Figure 5.7). In the EC without packing, we observed a rapid increase in potential over time compared to the PBEC setup. This observed behavior can be attributed to the formation of a passivation layer on the electrode surface in the EC without packing configuration. The passivation layer acts as a barrier, increasing the surface resistance and hindering the electron transfer process, leading to the observed increase in potential. In contrast, the presence of packing material in the PBEC setup likely mitigates passivation effects by providing a more stable environment and preventing direct contact between the electrode surface and potentially passivating species, resulting in a slower increase in potential over time. These findings highlight the importance of considering electrode configuration and passivation effects in electrochemical studies, as they can significantly influence the performance and efficiency of electrochemical processes.

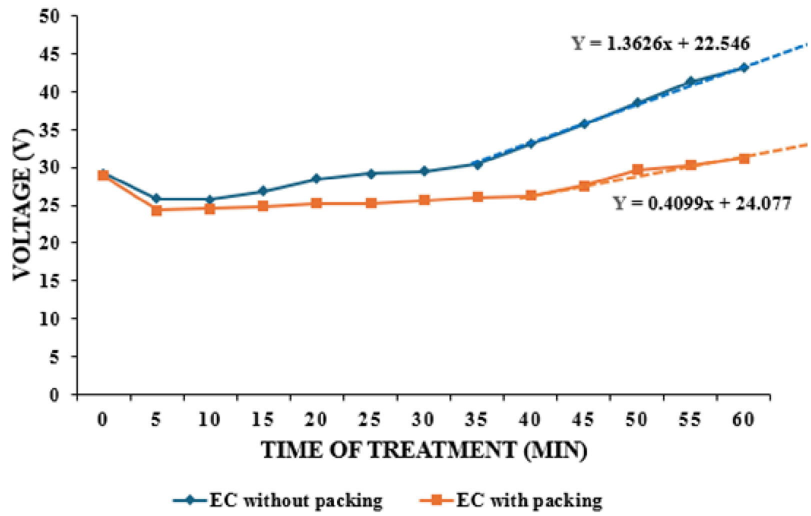

Figure 5.7: Passivation studies

Cost estimation

The economic evaluation of electrocoagulation for wastewater treatment under optimized conditions yielded insights into expenditure distribution. Energy consumption emerged as the primary cost, totaling the total cost of treatment 0.93\$CAN/m^3. Calculated at 0.06\$CAN/KWh, energy costs dominated at 0.86\$CAN/m^3. Electrode expenses, at 0.3\$CAN/Kg, contributed 0.65\$CAN/m^3, while polymer costs were 0.27\$CAN/m^3 at 0.95\$CAN/L.

The substantial proportion of the total cost attributed to energy consumption implies the importance of optimizing energy efficiency in the electrocoagulation process. Strategies such as utilizing renewable energy sources or implementing energy recovery systems could potentially mitigate this cost burden and enhance the economic viability of the process.

Furthermore, the relatively modest expenses associated with electrodes and polymer highlight the affordability of these consumables in the context of electrocoagulation based LWW treatment. However, ongoing efforts in research and development are required to explore cost-effective alternatives or improvements in electrode and polymer materials, aiming to further reduce operational costs.

5.7 Conclusion

The analysis of LWW composition reveals a complex profile with significant environmental implications. High levels of turbidity, alkalinity, conductivity, COD, metals, and nutrients pose risks to aquatic ecosystems and human health. Thorough treatment is imperative before discharge to mitigate these potential hazards.

Investigations into operating parameters using FD and CCD facilitated the development of predictive models for turbidity, TSS, and COD removal. The significance of linear, 2FI, and quadratic terms in these models underscores the crucial role of current intensity, treatment time, and polymer concentration in removal efficiencies. The Lack of Fit tests validated these terms, suggesting further exploration of cube effects for specific responses.

The optimization process, targeting maximum removal efficiencies for turbidity, TSS, and COD, involved exploring multiple starting points. Optimal conditions were identified at a current intensity of 2.995 A, treatment time of 58.82 min, and polymer dose of 5.778 mg/L. Experimental validation confirmed the high removal percentages achieved under these conditions.

At optimized conditions, the treatment process demonstrated significant removal of phosphate, ammonia, sulfate, and certain metals. However, limitations were observed in NPEOs removal, indicating the need for further investigation. The dispersion of metal ions from sacrificial metals highlighted a potential negative impact on treatment efficacy, warranting deeper research.

The integration of EPS in place of synthetic polymers initially reduced the efficiency of turbidity, TSS, and COD removal. Adjusting the EPS dosage to 8.0 mg/L, however, notably improved removal efficiencies across all parameters. This emphasizes the critical role of

EPS dosage in enhancing PBEC system efficiency and underscores the need for careful consideration of operational variables in treatment processes.

The economic estimate of this study under optimized conditions highlights the dominance of energy consumption as the primary expense, totaling 1.20$CAN/m^3, with energy costs alone contributing 0.86$CAN/m^3. While electrode and polymer expenses were comparatively minimal, optimizing energy efficiency remains paramount to mitigating overall costs. Strategies such as utilizing renewable energy sources or implementing energy recovery systems show promise in this regard. Additionally, the affordability of electrodes and polymers underscores the feasibility of electrocoagulation-based wastewater treatment, although ongoing research is needed to explore cost-effective alternatives.

In conclusion, this study provides comprehensive insights into LWW treatment, emphasizing the significance of optimizing operational parameters and integrating novel components like EPS. It highlights the need for targeted strategies to address specific contaminants and underscores the critical role of understanding and controlling operational variables in enhancing treatment efficiency and ensuring environmental safety. Our analysis emphasizes the importance of technological advancements and operational strategies in enhancing the economic viability of the process as a sustainable solution for LWW treatment. Further research and real-world implementations are crucial for refining cost estimations and improving overall economic feasibility.

5.8 Future Perspectives

Looking ahead, future research on LWW treatment should focus on refining and extending the prediction model developed in this study. Further investigation of the cubic effects, particularly in response to disproportionate importance, may contribute to a more comprehensive understanding of the complex relationship between performance dimensions and extraction efficiency. Such maintenance will be important for accurate modeling and for predicting treatment outcomes in different settings.

Furthermore, the study highlights the need for in-depth studies of metal ions from metal sacrifices and their potential adverse effects on treatment efficacy. Future research could investigate the mechanisms by which metals scatter, their consequences for overall treatment efficacy, and strategies to mitigate or optimize these effects thus allowing for electrochemical effects systems are more robust and reliable.

The addition of EPS to the treatment process gave promising results after dose adjustment. Future strategies should investigate different types and concentrations of biopolymers to further improve their performance. This research approach has the potential to promote a more sustainable and environmentally friendly alternative to synthetic wastewater treatment.

In addition, the study demonstrates the limitations of the removal of NPEOs, indicating the need for dedicated research to understand and address these challenges. Alternative research methodologies are adopted to treat or modify existing processes to increase the efficiency of NPEO removal.

In practical applications, the optimal conditions identified in this study for LWW treatment can be tested and applied in a larger scale study and real-world applications that will provide valuable insights into how the proposed treatment can be scalable, practical and economically viable methods.

In summary, future perspectives in this paper revolve around refining prediction models, exploring metal diffusion processes, investigating new biopolymer processes, solving NPEO removal challenges, and conducting experiments will be the best implementation of research to help improve the solution.

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Author Contributions

Sushil Kumar, Patrick Drogui, and R D Tyagi conceived and planned the idea of the study. Sushil Kumar experimented and collected the data. All authors carried out the analysis and contributed to the interpretation of the results. Sushil Kumar wrote the manuscript in consultation with Patrick Drogui, and R D Tyagi. All authors read and approved the final manuscript.

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Availability of data and materials

All data generated or analyzed during this study are included in this published article.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

All authors read and approved the final manuscript and mutually agreed that it should be submitted to Process Safety and Environmental Protection.

Competing interests

The authors declare that they have no conflicts of interest to declare.

6. ENHANCING THE TREATMENT PERFORMANCE OF UF MEMBRANE USING EPS FOR LWW TREATMENT

6.1. Article Introduction

Article's title	Enhancing the ultrafiltration membrane for laundry wastewater treatment using extracellular polymeric substances (EPS)
Names of the authors and their professional affiliations	Sushil Kumar[1], Patrick Drogui[1], R D Tyagi[2,3] 1. Institut National de la Recherche Scientifique (INRS), Centre-Eau Terre Environnement (ETE), Université du Québec, 490 Rue de la Couronne, Québec (QC), Canada G1K 9A9 2. BOSK-Bioproducts, 100-399 rue jacquard, Quebec (QC) G1N4J6 3. Distinguished Professor, School of Technology, Huzhou University, China.
Title of the journal	Journal of Environmental Management
Status of the paper	Submitted
Date of submission, acceptance, or publication	Not applicable
DOI	Not applicable

6.2. Preface

The preceding paper on electrocoagulation treatment using packed bed reactors with sacrificial anodes delves into an electrochemical approach for treating laundry wastewater. It implements a process involving sacrificial anodes to induce coagulation and precipitation of contaminants in the wastewater, aiding in their removal. This method focuses on utilizing electrochemical principles to facilitate the separation of pollutants from the wastewater by forming flocs.

In contrast, this paper focuses on enhancing the performance of UF membranes for treating LWW by incorporating EPS. It explores the potential of EPS to enhance the filtration and separation efficiency of UF membranes. The goal is to improve the removal of contaminants, such as suspended solids and organic matter, from laundry wastewater by modifying the membrane's characteristics through the inclusion of EPS.

Despite employing different methodologies, both studies target the challenge of improving the treatment efficacy of laundry wastewater. The electrocoagulation approach focuses on inducing coagulation and precipitation of contaminants for their removal, while the enhancement of UF membranes with EPS aims to augment the filtration capabilities to separate pollutants more effectively. Both methodologies contribute valuable insights into innovative techniques for addressing the complexities associated with managing and treating LWW, offering distinct yet complementary approaches to wastewater treatment.

6.3. Abstract

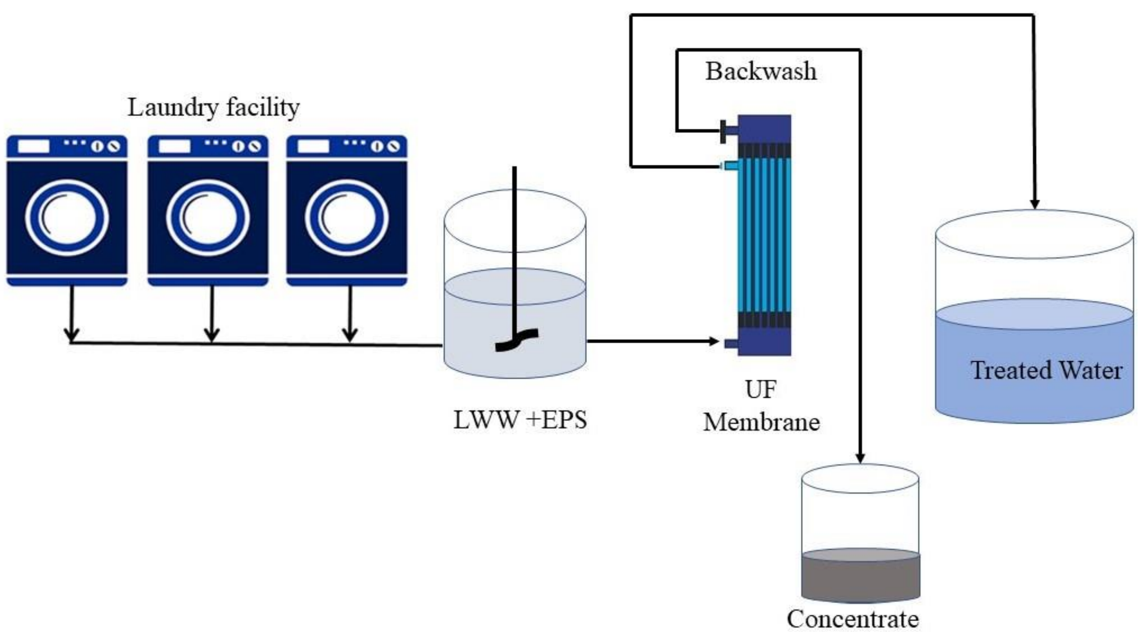

Figure 6.1: Graphical Abstract

This study presents the use of extracellular polymeric substances (EPS) derived from bacterial strain BS-04 for enhancing the treatment performance of ultrafiltration (UF) membrane for the commercial laundry wastewater (LWW). EPS characterization revealed a predominantly proteinaceous composition (1.45 ± 1.5 g/L) with negligible carbohydrate content, a zeta potential of -38.2 ± 0.5 mV and a viscosity of 30.99 ± 0.5 mPas. LWW exhibited elevated levels of turbidity (109 ± 5 NTU), COD (576.7 ± 30 mg/L), and Nonylphenol ethoxylates (NPEOs) (568 ± 150 μg/L). The EPS dose was the main influent parameter on the turbidity and COD removals. The optimal experimental parameters for Turbidity and COD removals have been investigated by using a Central Composite methodology. Optimized conditions were established while imposing a TMP of 18 psi, a temperature of 60˚C, and EPS dose of 14.17 mg/L. These conditions resulted in a permeate flow rate of 0.81 L/min, turbidity removal of 98.95%, COD removal of 80.53%, and a backwash time of 31 minutes. Adding EPS to UF helps prevent membrane fouling, making the process more efficient. Treating the concentrate from UF with packed bed electrocoagulation (PBEC), using specific conditions (current intensity: 2.99A, treatment time: 58.80 minutes, and EPS dose: 8 mg/L), resulted in significant removal of contaminants. These contaminants include turbidity (99.34% removal), total suspended solids (99.57% removal), total solids (99.70% removal), COD (87.60% removal), and NPEOs (53.95% removal). The costs for UF with EPS and PBEC were similar, suggesting that EPS-based strategies have potential for sustainable wastewater treatment and water management. This research offers valuable insights into optimizing and making innovative approaches cost-effective for treating LWW with UF.

6.4. Introduction

The Laundry wastewater (LWW) presents a formidable challenge due to its diverse and often hazardous composition, necessitating effective treatment to safeguard environmental and public health (Chavan *et al.*, 2024; Kumar *et al.*, 2023). Originating from laundry facilities, this wastewater contains a complex mixture of contaminants including detergents, surfactants, organic matter, suspended solids, and microorganisms (Akarsu & Deniz, 2021). If left untreated or inadequately treated, the discharge of LWW poses significant risks to water quality, aquatic ecosystems, and human well-being (Kim *et al.*, 2014). Thus, there is an urgent need to implement efficient treatment methods to mitigate its adverse environmental impact and maintain the integrity of water resources (Kumar *et al.*, 2022c).

Among the array of treatment technologies available, ultrafiltration (UF) emerges as a promising solution for addressing the challenges posed by untreated commercial LWW. UF, a membrane-based separation process, offers several advantages including high removal efficiency of suspended solids, pathogens, and dissolved contaminants (Hu *et al.*, 2023). UF has been harnessed to treat wastewater, effectively recovering water resources and reducing pollutant concentrations (Hembach *et al.*, 2019). However, UF encounters its own set of challenges, primarily fouling, which occurs when contaminants accumulate on the membrane surface or obstruct membrane pores. Fouling compromises UF efficiency, leading to decreased performance and increased operational costs (Vela *et al.*, 2009).

Extracellular polymeric substances (EPS) are intricate biopolymers synthesized by microorganisms, crucial for biofilm formation and microbial cohesion in diverse ecosystems (Kumar *et al.*, 2020). EPS serve multiple functions, such as promoting cellular aggregation, shielding microorganisms from environmental stresses, and acting as a scaffold for microbial attachment on surfaces (Nouha *et al.*, 2016). Composed of polysaccharides, proteins, nucleic acids, lipids, and humic substances, the composition of EPS varies based on microbial species, environmental factors, and microbial communities (Matilainen *et al.*, 2010). Their diverse composition influences functional properties, particularly in applications such as wastewater treatment. In biofilms, EPS play pivotal roles in cell attachment, structural support, environmental stress mitigation, nutrient retention, intercellular communication, and resistance to antimicrobial agents (Lin *et al.*, 2019). This multifunctionality has attracted significant attention in disciplines like environmental microbiology, biotechnology, and wastewater treatment. The versatile nature of EPS offers promising avenues for enhancing processes like ultrafiltration in treating complex wastewater streams (Peng *et al.*, 2022).

In the quest to develop an optimal treatment process, response surface methodology (RSM) emerges as a valuable tool. RSM allows for systematic exploration of process parameters using statistical optimization techniques such as Central Composite Design (CCD) and factorial design (FD). Through RSM, researchers can identify the optimal conditions for UF-EPS treatment, maximizing efficiency and meeting regulatory guidelines (Rao *et al.*, 2019). This approach enables the development of a scientifically sound and robust treatment process for laundry wastewater.

This paper presents a novel approach, integrating ultrafiltration UF with EPS, aiming to enhance the efficiency of commercial LWW treatment and meet stringent guideline limits set by the Quebec government for reuse in laundries. Utilizing CCD and FD as statistical optimization techniques, this study seeks to improve the efficacy of UF-EPS treatment and draw scientifically robust conclusions. The originality of this research lies in its thorough analysis of the integrated UF-EPS system, providing insights into innovative strategies for sustainable water management practices in the context of laundry wastewater treatment. Additionally, the findings presented here not only contribute to the advancement of wastewater treatment technologies but also pave the way for future research, innovations, and applications aimed at addressing the challenges of environmental conservation.

6.5. Materials and Methodology

EPS Production and Characterization

EPS production was initiated using a blend of crude glycerol and industrial sludge. The bacterial strain BS-04, isolated from wastewater sludge, was cultivated in this mixture of crude glycerol and secondary sludge sourced from pulp and paper mill wastewater treatment (Kaur *et al.*, 2019) . This complex process took 96 hours of fermentation, maintaining control over pH (6.8-7.0), temperature (28-30 ˚C), and dissolved oxygen (30-50%) levels. The culmination of this fermentation process yielded the fermented broth termed broth EPS (B-EPS). Subsequent centrifugation at 4ºC for 20 minutes at 6000 g enabled the extraction of slime EPS (S-EPS) from the supernatant (Nouha *et al.*, 2016).

Quantification of the dry weight of S-EPS involved a meticulous procedure. The supernatant was mixed with 95% v/v ethanol in a 1:2 ratio and refrigerated at -20ºC overnight to induce phase separation. The resulting precipitates (solid phase) of S-EPS were isolated by Sorvall centrifugation at 6000 g and 4ºC for 20 minutes, subsequently dried at 60ºC until a consistent weight was achieved.

Determining the concentration of EPS was calculated using Eq. (13):

$$\mathrm{S-EPS} = (\mathrm{W_2} - \mathrm{W_1}) * \frac{1000}{\mathrm{V}}$$ **Equation 13**

Where (W_1) represents the initial weight of the empty container in grams, (W_2) signifies the final dry weight of the container with the dried sample in grams, and (V) denotes the volume of the sample in liters.

Another variant, capsular EPS (C-EPS), was ascertained by mixing biomass pellets (obtained via centrifugation of fermented broth) with an equivalent volume of water. This mixture underwent heating at 60ºC in a water bath for 60 minutes to liberate C-EPS into the liquid phase. The freed C-EPS in the liquid phase was isolated through centrifugation at 6000 g and 4ºC for 20 minutes.

The cumulative EPS concentration, or broth EPS (B-EPS) concentration, was computed using Eq. (14):

$$\mathrm{Total\ EPS} = \mathrm{S-EPS} + \mathrm{C-EPS}$$ **Equation 14**

The liquid form of collected S-EPS was employed for treating LWW. S-EPS is favored in wastewater treatment because it has higher binding capacities for contaminants in wastewater, enhancing the removal of pollutants such as heavy metals, organic compounds, and nutrients. Its adhesive properties allow for the formation of biofilms on surfaces like filter media or membranes, improving the efficiency of biological treatment methods like activated sludge processes, biofiltration, and membrane bioreactors (Guo *et al.*, 2017). Whereas, C-EPS protects individual cells, while B-EPS, being soluble, doesn't contribute as effectively to biofilm formation or pollutant removal. (Hu *et al.*, 2023). The treatment mechanism utilized S-EPS as a bio-flocculant in this study. The process encompassed S-EPS forming a protective biofilm on PES ultrafiltration membrane. This biofilm acts as a barrier, preventing foulants like organic matter and bacteria from sticking to the membrane (Toto, 2018). The EPS matrix discourages direct contact between foulants and the membrane while promoting the growth of beneficial bacteria. This helps reduce fouling, improving the membrane's performance and lifespan in wastewater treatment (Peng *et al.*, 2022).

Viscosity (mPas) and zeta potentials (ζ) were measured using Viscometer (DV DV-II PRO (Brookfield Engineering Laboratories, Inc., Stoughton, MA, USA)) and Zetaphoremeter (Zetaphoremeter IV, Zetacompact Z8000, CAD Instrumentation, France), respectively (Nouha et al. 2017). The protein content of the extracted S-EPS was determined using bovine serum albumin (BSA) as a standard (Kruger 2002). The carbohydrate content of the extracted EPS was determined by LC-MS-MS (More et al. 2015). To determine flocculation activity, the turbidity of the sample supernatant was measured using Turbidometer (Micro 100 Turbidity meter, Hach Company). FA measured was based on the relative decrease in turbidity of suspension after settling (More et al. 2012a). FA was calculated according to the equation 15.

$$\mathrm{FA} = (1 - \frac{\mathrm{S}}{\mathrm{C}}) * 100$$ **Equation 15**

Where, C is the control turbidity (NTU), and S is the sample turbidity (NTU). Kaolin suspension without EPS served as control. For quantifying the effects of EPS on dewaterability, Capillary Suction Time (CST) of the settled kaolin sediments was measured. CST was determined by the CST instrument (Triton electronics, model 304 M CST, Dunmow, Essex), using a 10-mm diameter reservoir. Lower CST value as compared to the control CST indicated better dewaterability (More et al. 2012a). The sediments from jar test samples without EPS addition was the control. The increase in the dewaterability was calculated by equation 16.

$$\text{Dewaterability } (\%) = (1 - \frac{\mathrm{CST_{sample}}}{\mathrm{CST_{control}}}) * 100$$ **Equation 16**

Where, CST is in seconds.

LWW Collection and Characterization

Sampling and characterizing laundry wastewater (LWW) involved a systematic process initiated at a commercial laundry facility situated in Montreal, Canada. The LWW sample was specifically drawn from the outlet point of the commercial laundry, collected within a 200 L container, and subsequently transported to the experimental site, the INRS laboratory. To ensure its integrity, the 200 L LWW batch was partitioned into ten 20 L buckets and securely stored in a cold room maintained at -4°C for further experimentation and analysis.

Upon collection, the LWW samples underwent comprehensive analysis to assess various crucial parameters encompassing pH, turbidity, conductivity, COD (Chemical Oxygen Demand), TSS (Total Suspended Solids), TS (Total Solids), NPEO (Nonylphenol Ethoxylates), nitrates, phosphates, and a spectrum of metals. These parameters provided critical insights into the distinct characteristics of the wastewater. Such comprehensive characterization serves as a cornerstone for devising effective treatment methodologies tailored to the specific composition of the LWW. Parameters such as turbidity, pH, conductivity, suspended solids, COD, NPEOs, and the presence of various metals were compared against the reuse standards set up by Health Canada and Quebec Municipality (Collivignarelli *et al.*, 2019), showcasing the variance between the actual LWW composition and the prescribed thresholds for safe discharge or potential reuse. These standards are mentioned in Table 1. This detailed comparison laid the foundation for crafting targeted treatment approaches aimed at mitigating or meeting the designated standards for safe disposal or potential reutilization of the wastewater.

Table 6.1*:* Water rejection standard and Reusing standards

Parameters	Rejection standard	Water reusing standard
Turbidity (NTU)	n.r.	≤ 2
Ph	5.5-9.5	5-9
Conductivity (µS/cm)	n.r.	≤1500
Suspended solids (mg/L)	≤ 80	10-50
Total solids (mg/L)	<100	n.r.
COD (mg/L)	<150	<60
NPEOs (µg/L)	<200	n.r.
Sodium (mg/L)	10kg/day	n.r.
Zinc (mg/L)	n.r.	n.r.
Copper (mg/L)	n.r.	0.01
Iron (mg/L)	n.r.	0.1
Manganese (mg/L)	n.r.	0.05
References	(Corona *et al.*, 2021; Li *et al.*, 2022a; Manouchehri & Kargari, 2017; Omolara, 2017)	(Benis *et al.*, 2021; BHAGAT, 2018; Faria *et al.*, 2019; Moura *et al.*, 2019; Nascimento *et al.*, 2019)

*n.r. : Not reported

Ultrafiltration Membrane Setup

Setting up the ultrafiltration process involves meticulous preparation and handling of the membrane system, as shown in fig. 2. The Polyethersulfone (PES) UF membrane with a surface area of 0.5 m^2, molecular cut off value of 100-150 kDa and the pore size of 0.02 micron was carefully assembled into the filtration unit following manufacturer guidelines. Prior to experimentation, the membrane undergoes conditioning procedures such as pre-soaking, cleaning, and flushing to activate the membrane's surface and eliminate any residual impurities that might affect performance.

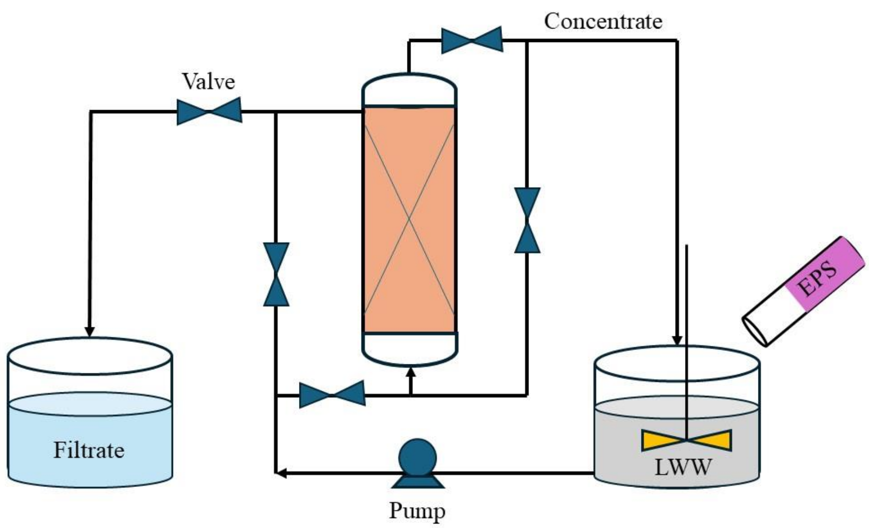

Figure 6.2: Crossflow Ultrafiltration combined with EPS

Design of experiments

The experimental design incorporates a Factorial and Central Composite Design to evaluate the impact of multiple factors on the ultrafiltration process, as shown in Table 2. The experimental design and statistical optimization were conducted through Response Surface Methodology (RSM) using Design Expert-10. RSM involves employing regression analysis to establish connections between response variables and controlled variations in independent factors. This methodology allows for the rapid generation of multiple experimental permutations, significantly improving laboratory testing efficiency. By leveraging estimated parameters, it becomes feasible to pinpoint the primary variable that holds the most sway over the anticipated outcome. As emphasized by Mu et al. in 2009, this approach enables researchers to prioritize the variables critical for product acceptance, refining focus and enhancing efficiency.

• Factorial Design: Factors and coded variables were TMP (Transmembrane Pressure), Temperature, EPS concentration are the primary factors to be tested, each having a coded variable range (-1 to +1) that corresponds to their minimum and maximum values. TMP ranges from 10 to 18 psi (coded as -1 to +1), Temperature ranges from 20 to 60 ºC (coded as -1 to +1), and EPS Concentration ranges from 5 to 15 mg/L (coded as -1 to +1).

• Central Composite Design (CCD): CCD supplements the factorial design by introducing center points to explore the quadratic response of the system within the factor ranges. This design enables the examination of curvature in the response surface and aids in optimizing the process parameters.

• Reponses: The responses were calculated in the form of Y_1: Permeate flow rate (L/min), Y_2: Turbidity removal (%), Y_3: COD removal (%) and Y_4: Back wash time (min).

Table 6.2*:* Experimental variables and their range

Coded Variables	Factor	Range		$U_{1,0}$	ΔU_1
		Minimum value (-1)	Maximum value (+1)		
X_1	U_1: TMP (psi)	10	18	14	4
X_2	U_2: Temperature (ºC)	20	60	40	20
X_3	U_3: EPS concentration (mg/L)	5	15	10	5

Studying Fouling behavior of the membrane

The fouling UF membranes poses a significant challenge, impacting performance, increasing operational costs, and raising environmental concerns (Hu *et al.*, 2023). Understanding the fouling behavior is crucial for optimizing processes, reducing costs, minimizing environmental impact, and driving innovation in membrane technology (Derlon *et al.*, 2016). Accordingly, quantification of fouling arising from residual pollutants is imperative, achieved through the assessment of specific resistance to filtration (SRF). This entails plotting a graph depicting filtration time/filtration volume against filtrate volume based on experimental data. The slope of this curve yields the magnitude of filtration resistance, it is derived from Darcy's equation.

$J = \frac{dV}{dt}.\frac{1}{A} = \frac{\Delta P}{\mu.(R_F+R_m)}$ (Darcy's equation) **Equation 17**

After integration and rearrangement, we can get equation (18),

$\frac{t}{V} = \left(\frac{\mu.R_F.a}{2PA^2}\right)V + \frac{\mu.R_m}{A.P}$ **Equation 18**

$\frac{t}{V} = f(V) = K_b.V + K_0$ **Equation 19**

K_b is the slope of straight line presented in equation (20):

$K_b = \frac{\mu.R_F.a}{2PA^2} \Rightarrow R_F(SRF) = \frac{2.K_b.P.A^2}{\mu.a}$ **Equation 20**

Where:

V= volume of filtrate, t = time of filtration, P= pressure across filter medium, A = surface area of filtration, a= A/V = surface areas per volume of filtrate, R_F = specific resistance to filtration and R_m = initial resistance of the membrane.

To study this phenomenon, the optimized experiments were done with and without EPS to see the correlation and effect of EPS on membrane fouling.

Concentrate Management using PBEC

The optimization of the PBEC reactor for concentrate obtained after ultrafiltration treatment involved structural modifications. A 5-liter glass reactor was specifically configured into six compartments using plexiglass dividers. Each segment housed stainless steel cathodes (110 cm²) and graphite anodes (110 cm²) strategically aligned to facilitate a serpentine flow pattern, maximizing contact time between the wastewater and electrodes. Integration of sacrificial iron scrap between the cathodes and anodes served to significantly enhance coagulation and flocculation processes. For effective electrochemical reactions, a parallel connection to a DC generator was established across the electrodes.

Electrocoagulation, powered by a DC current, was employed for contaminant removal. Sacrificial iron scrap electrodes induced coagulation and flocculation. PBEC parameters—current intensity, treatment time, EPS concentration—were optimized using RSM via factorial and central composite designs.

The setup and optimizations were exactly the same as those used in the study on electrocoagulation treatment of LWW using packed bed reactors with sacrificial anodes (Kumar *et al.*, 2023).

Analytical Method

Analytical techniques were employed for various measurements throughout the study. The pH levels were determined utilizing a pH-meter (Fisher Acumet model 915) equipped with a double junction Cole-Palmer electrode incorporating an Ag/AgCl reference cell. Similarly, conductivity measurements were conducted using a dedicated conductivity meter (Oakton Model 510). Key parameters such as Chemical Oxygen Demand (COD), Total Suspended Solids (TSS), Total Solids (TS), and Non-ionic Polyethylene Oxide Surfactants (NPEOs) were assessed using established methodologies outlined by CEAEQ (Center of Expertise in Environmental Analysis of Quebec) (l'Environnement & climatiques, 2014). Additionally, the quantification of metals was performed using the ICP-AES method as suggested by CEAEQ.

Cost Estimate

<u>LWW treatment using UF and EPS</u>: The energy utilized was calculated at a rate of 0.06$CAN/KWh, aligning with the rate established by the province of Quebec (Canada) (Kumar *et al.*, 2023). The cost of EPS production was calculated at a rate of 0.95$CAN/L (Kumar *et al.*, 2020).

<u>Concentrate Management using PBEC</u>: The cost of electrode material was calculated at a rate of 0.3$CAN/Kg (Yasri *et al.*, 2020), along with the cost of energy consumption and cost of EPS as mentioned above.

The overall expenditure was assessed in terms of Canadian dollars spent per cubic meter of treated solution ($CAN/m^3).

6.6. Results and discussion

EPS Characterization

The EPS generated by bacterial strain BS-04 through the utilization of crude glycerol underwent comprehensive characterization, analysis of its composition, viscosity, flocculation activity, and dewatering potential. At the termination of a 96-hour period, the protein and carbohydrate constituents within the final B-EPS were quantified using the Bradford assay and LC-MS-MS techniques, respectively. Notably, no significant amounts (< 20 mg/L) of carbohydrates such as glucose, fructose, lactose, sucrose, galactose, xylose, or trehalose were detected in the B-EPS. Conversely, the analysis revealed a presence of 1.45g/L of proteins. Additionally, the B-EPS exhibited specific properties, with a viscosity of 30.99mPas, a zeta potential of -38.2mV, a flocculation activity of 82%, and a dewaterability index of 40.21%.

LWW Characterization

The comprehensive measurements provide detailed information about the composition and quality of the collected commercial LWW under study, summarised in Table 3.

Table 6.3: Commercial laundry wastewater characterization and the standards for effluent discharge and reuse

Parameters	Values	Recommendation for effluent discharge in the sewer	Recommendation for reuse in the laundries
Turbidity (NTU)	110 ± 5	<10	<2
pH	10 ± 0.5	6.5-8.5	6.5-8.5
Conductivity (μS/cm)	494 ± 20	n.r.	<2000
Total Suspended solids (mg/L)	60 ± 10	<30	<5
Total solids (mg/L)	622 ± 20	<100	<500
COD (mg/L)	579 ± 30	<150	<100
NPEOs (μg/L)	570 ± 150	<200	<25
Sulphate (mg/L)	22± 5	n.r.	<250
Sulphite (mg/L)	0.2 ± 0.05	n.r.	n.r.
Nitrate (mg/L)	4 ± 1	<10	n.r.
Nitrite (mg/L)	1.5 ± 0.5	<1	<1
Ammoniacal nitrogen (mg/L)	2.3 ± 1	<10	n.r.
Total nitrogen (mg/L)	9.7 ± 0.5	<10	<10
Phosphate (mg/L)	21 ± 2.0	<1	n.r.
Aluminium (mg/L)	0.096 ± 0.02	n.r.	n.r.

Calcium (mg/L)	18.6 ± 2.0	n.r.	n.r.
Magnesium (mg/L)	3.87 ± 0.5	n.r.	n.r.
Sodium (mg/L)	104 ± 10	n.r.	n.r.
Potassium (mg/L)	3.76 ± 0.5	n.r.	<50
Sulphur (mg/L)	20.6 ± 2.0	n.r.	n.r.
Zinc (mg/L)	0.42 ± 0.15	<2	n.r.
Copper (mg/L)	0.03 ± 0.01	<1	n.r.
Iron (mg/L)	0.23 ± 0.2	<1	<0.3
Manganese (mg/L)	0.019 ± 0.005	<0.1	n.r.
Lead (mg/L)	<0.003	<0.1	<0.01
References	This study	(Cárdenas *et al.*, 2023; Zhang *et al.*, 2022) (Ville de Quebec)	(Exall *et al.*, 2006; Van Rossum, 2020)

Turbidity (109 ± 5 NTU): This indicates the degree of cloudiness caused by suspended particles in the water, which in turn suggests a substantial presence of suspended solids in the LWW, potentially originating from detergents, textile fibers, or other sources. High turbidity levels could impact light penetration in water bodies, influencing photosynthesis and affecting aquatic organisms by reducing available light (Trigueros *et al.*, 2023).

pH (10.34 ± 0.5): This indicates the high alkalinity of the LWW attributed to alkaline detergents used in laundry processes., which can alter the chemical equilibrium in aquatic environments, potentially affecting nutrient availability, biological processes, and aquatic life (Ran *et al.*, 2022).

Conductivity (496 ± 20 µS/cm): This elevated value implies the presence of dissolved ions and salts, that can affect the osmoregulation of aquatic organisms and alter the overall water quality, impacting the ecological balance of the receiving water bodies (French *et al.*, 2010).

Suspended Solids (60 ± 10 mg/L) and Total Solids (620 ± 20 mg/L): These measurements indicate that LWW may consist of organic and inorganic materials, detergents, textile fibers, and other particulate matter which can lead to sedimentation, reducing water clarity and potentially disrupting aquatic habitats (Kumar *et al.*, 2022a).

COD (576.7 ± 30 mg/L) and NPEOs (568 ± 150 µg/L): The high COD suggests a significant amount of biodegradable organic compounds, indicating the potential for oxygen depletion upon discharge into water bodies. NPEOs are surfactants commonly found in detergents and their presence indicates a potential source of non-biodegradable pollutants in the water (Sharma & Gupta, 2014).

Chemical Composition:

Sulfate (23 ± 5 mg/L) and Sulphite (0.3 ± 0.05 mg/L): Likely originating from detergent additives, contribute to the overall chemical composition of the wastewater and may affect water quality and taste (Bo *et al.*, 2012).

Nitrate (5 ± 1 mg/L) and Nitrite (1.78 ± 0.5 mg/L): Instigating from detergents or other sources and may pose risks related to eutrophication and potential health hazards (Zhou *et al.*, 2022).

Ammoniacal Nitrogen (2.5 ± 1 mg/L), Total Nitrogen (9.4 ± 0.5 mg/L), and Phosphate (23 ± 2.0 mg/L): Originating from detergents and organic matter, can contribute to eutrophication, impacting water quality and aquatic ecosystems (Kim *et al.*, 2015a).

Metallic Elements: The presence of various metallic elements such as aluminium (0.094 ± 0.02 mg/L), calcium (18.8 ± 2.0 mg/L), magnesium (3.82 ± 0.5 mg/L), sodium (108 ± 10 mg/L), potassium (3.73 ± .5 mg/L), sulphur (20.6 ± 2.0 mg/L), zinc (0.45 ± 0.15 mg/L), copper (0.07 ± 0.01 mg/L), iron (0.26 ± 0.2 mg/L), manganese (0.017 ± 0.005 mg/L), and trace amounts of lead (<0.003 mg/L) raises concerns about potential toxicity to aquatic organisms and environmental contamination.

These comprehensive measurements show a detailed picture of the composition and quality of the collected laundry wastewater from the commercial laundries of Quebec. The observed parameters highlight potential environmental risks associated with its discharge, emphasizing the importance of effective treatment processes before releasing

LWW into natural water bodies to mitigate its adverse impacts on aquatic ecosystems and human health.

Factorial Design and Significant Parameters

The study aimed to explore the impact of various factors, namely TMP (X_1), temperature (X_2), and EPS concentration (X_3), on the treatment of LWW with a Full Factorial Design (FD) approach. Each variable was set at two levels, denoted as (−1) and (+1), corresponding to minimum and maximum values, respectively. The experimental region and associated coded values can be found in Table 2.

The resulting factorial matrix consisted of experiments, wherein four responses, namely Permeate flow rate (L/min) (Y_1), Turbidity removal (%) (Y_2), COD removal (%) (Y_3)and Back wash time (min) (Y_4) were investigated. A summary of the findings is presented in Table 4.

Table 6.4*:* Factorial design matrix and response

Assays	Experiment design			Plan of experiments				Results (removal %)		
	X_1	X_2	X_3	U_1 (psi)	U_2 (C)	U_3 (mg/L)	Permeate Flow rate L/min	Turbidity	COD	Back wash time min
1	-1	-1	-1	10	20	5	0.49	97.1	62.8	18
2	1	-1	-1	18	20	5	0.77	97	65.3	16
3	-1	1	-1	10	60	5	0.56	98.6	77.4	22
4	1	1	-1	18	60	5	0.82	98.4	74.6	20
5	-1	-1	1	10	20	15	0.51	97.5	72.9	23
6	1	-1	1	18	20	15	0.76	97.3	70.4	24
7	-1	1	1	10	60	15	0.52	99.2	84.2	34
8	1	1	1	18	60	15	0.83	99.1	78.4	32

From the factorial design these mathematical models were obtained for the different responses:

$Y_1 = 0.66 + 0.14X_1 + 0.025X_2 + 0.001X_3 + 0.002\ X_1X_2$ **Equation 21**

$Y_2 = 98.02 - 0.075X_1 + 0.80\ X_2 + 0.25X_3 + 0.00\ X_1X_3 + 0.075X_2X_3$ **Equation 22**

$Y_3 = 73.25 – 1.08\ X_1 + 5.04\ X_2 + 3.23\ X_3$ **Equation 23**

$Y_4 = 23.63 - 0.63X_1 + 3.38\ X_2 + 4.63\ X_3 + 0.38\ X_1 X_3 + 1.38\ X_2X_3$ **Equation 24**

Where Y_1 is experimental response for permeate flow rate, Y_2 is experimental response for turbidity removal, Y_3 is experimental response for COD removal and Y_4 is the experimental response for Back wash time. To assess the validity of these models, an Analysis of Variance (ANOVA) was conducted using design expert software.

Effect of experimental parameters

The conducted study utilized a factorial design approach to investigate the impact of various experimental parameters, including TMP, temperature, and EPS concentration, on the treatment of liquid waste. The resulting mathematical models and Pareto graph analysis as shown in figure 3, provided valuable insights into the relative contributions of these parameters to the response.

The mathematical model for permeate flow rate (Y_1) revealed that TMP had the most significant effect, contributing approximately 96.89% to the variation observed in the response. This emphasizes the critical role of TMP in determining the rate at which permeate is produced during the treatment process. Furthermore, temperature and EPS concentration were found to have minor but discernible effects on permeate flow rate, with contribution percentages of 3.09% and 0.005%, respectively. These findings align with previous studies highlighting the influence of TMP on membrane filtration processes and emphasize the need for precise control and optimization of TMP to maximize permeate flow rates (Li *et al.*, 2020).

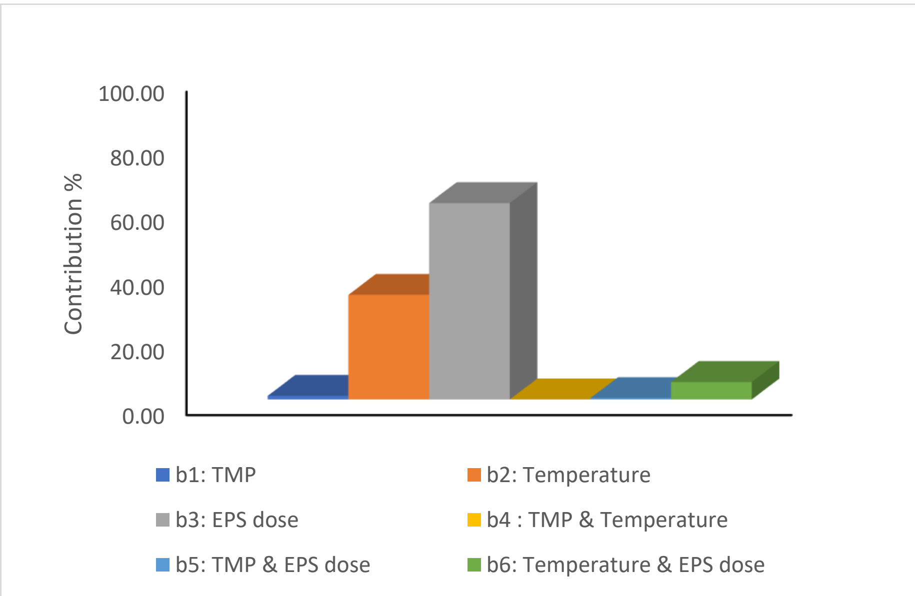

Figure 6.3: Effects of different factors during factorial design

Temperature emerged as the dominant factor influencing turbidity removal (Y_2), contributing approximately 89.67% to the observed variation in the response. This underscores the sensitivity of turbidity removal to temperature variations, with higher temperatures generally enhancing the efficiency of turbidity removal processes. Additionally, TMP and EPS concentration were found to play supplementary roles in determining turbidity removal, contributing 0.79% and 8.76%, respectively. The combined effect of temperature and EPS concentration further highlighted their importance, contributing an additional 0.79% to turbidity removal. These findings corroborate previous research highlighting the influence of temperature and EPS concentration on membrane filtration efficiency and draw attention to the need to consider these factors when designing treatment systems (Mukherjee & Ghosh, 2022).

EPS dose emerged as a significant factor influencing COD removal (Y_3), contributing approximately 28.20% to the observed variation in the response. This highlights the importance of EPS concentration in facilitating the removal of chemical oxygen demand from liquid waste streams. Furthermore, temperature was found to be a major determinant of COD removal, contributing 68.65% to the observed variation, while TMP played a minor role, contributing 3.15%. These findings underscore the complex interplay between temperature, EPS concentration, and membrane filtration efficiency in COD removal processes and emphasize the need for holistic optimization strategies to maximize treatment performance (Guo *et al.*, 2017).

The model for backwash time (Y_4) indicated that temperature and EPS concentration exerted substantial effects, contributing 32.36% and 60.72%, respectively, to the observed variation in the response. This shows the sensitivity of backwash time to temperature and EPS concentration variations, with higher temperatures and EPS concentrations generally necessitating longer backwash durations to maintain treatment efficiency. Additionally, interaction with TMP was 1.12% and the combined interactions between TMP and EPS concentration, as well as temperature and EPS concentration, were found to have minimal effects on backwash time, contributing 0.41% and 5.39%, respectively. These findings highlight the importance of considering temperature and EPS concentration when optimizing backwash protocols to ensure efficient membrane cleaning and prolonged system operation (Siddiqui *et al.*, 2020).

Optimization using CCD Design Methodology:

The Central Composite Design (CCD) was employed to depict the response of the three numerical factors within the entire experimental domain, as summarised in Table 5. This methodology offers several advantages, including: (i) comprehensive exploration of the experimental space; (ii) facilitation of coefficient determination for fitting a mathematical model; and (iii) effective interpolation of responses. Three quantitative factors denoted as U_1, U_2, and U_3 are hypothesized to influence the response variables. The response can me be presented by following model equation 25:

$$Y = b_0 + \sum_{i=1}^{k} b_i X_i + \sum_{i=1}^{k} b_{ii} X^2 + \sum_j \sum_{i=1}^{k} b_{ij} X_i X_j + e_i \qquad \textbf{Equation 25}$$

In this equation, Y represents the experimental response, b_0 signifies the mean of the experimental response, b_i denotes the estimated primary effect of factor j on response Y, b_{ii} represents the estimated secondary effect of factor j on response Y, and b_{ij} indicates the estimated interaction effect between factors i and j on response Y.

Table 6.5: Design of experiments using CCD methodology

	Coded Value			Experimental Value						
				U_1	U_2	U_3	Y_1	Y_2	Y_3	Y_4
Run	X_1	X_2	X_3	TMP (psi)	Temperature (°C)	EPS doses (mg/L)	Permeate flow rate (L/min)	Turbidity removal (%)	COD removal (%)	Back wash time(min)
1	0	0	0	14	40	10	0.65	98.8	80.8	24
2	-1	1	-1	10	60	5	0.56	98.6	77.4	22
3	0	0	0	14	40	10	0.65	98.8	80.8	24
4	0	0	-1.68	14	40	1.6	0.63	98.7	60.5	21
5	0	0	0	14	40	10	0.65	98.8	80.8	24
6	0	1.68	0	14	73.6	10	0.67	98.2	80.3	25
7	0	0	0	14	40	10	0.65	98.8	80.8	24
8	1	1	1	18	60	15	0.83	99.1	78.4	32
9	0	0	0	14	40	10	0.65	98.8	80.8	24
10	0	0	0	14	40	10	0.65	98.8	80.8	24
11	-1	-1	1	10	20	15	0.51	97.5	72.9	23
12	-1	1	1	10	60	15	0.52	99.2	84.2	34
13	1	-1	-1	18	20	5	0.77	97	65.3	16
14	1	1	-1	18	60	5	0.82	98.4	74.6	20
15	0	0	1.68	14	40	18.4	0.66	98.6	86.2	30
16	1	-1	1	18	20	15	0.76	97.3	70.4	24
17	-1	-1	-1	10	20	5	0.49	97.1	62.8	18
18	0	-1.68	0	14	6.4	10	0.64	96.8	79.3	24
19	-1.68	0	0	7.28	40	10	0.32	98.8	80.4	26
20	1.68	0	0	20.72	40	10	0.9	99	81.6	25

Based on the experimental results of CCD design, following equations were obtained for different response variables:

$Y_1 = 0.65 + 0.15X_1 + 0.018X_2 + 0.0002X_3 + 0X_1X_2$ **Equation 26**

$Y_2 = 98.81 - 0.019X_1 + 0.64X_2 + 0.13X_3 + 0X_1X_2 + 0X_1X_3 + 0.075X_2X_3 - 0.027X_1^2 - 0.52X_2^2 - 0.12X_3^2$ **Equation 27**

$Y_3 = 76.96 - 0.48X_1 + 3.29X_2 + 5.05X_3 - 1.07X_1X_2 - 1X_1X_3 + 0.57X_2X_3 - 0.91X_1^2 - 1.33X_2^2 - 3.61X_3^2$ **Equation 28**

$Y_4 = 24.20 - 0.49X_1 + 2.10X_2 + 3.82X_3 - 0.37X_1X_2 + 0.38X_1X_3 + 1.38X_2X_3 + 0.19X_1^2 - 0.16X_2^2 + 0.19X_3^2$ **Equation 29**

From the above equations (26)-(29) it is clear that temperature and EPS dose are very important factors for COD removal. However, in case of permeate flowrate TMP and temperature play very important role. From the model equation it can be also seen that the interaction X_1X_2 and X_2X_3 plays very significant role on COD removal and back wash time.

Examination of the 3D model depicted in Figure 4(a) reveals a discernible increase in turbidity removal efficiency as the temperature ranges between 40°C and 60°C. This observation suggests a direct correlation between rising temperature and the enhanced efficacy of the process in eliminating suspended particles and colloidal matter from LWW, consequently improving the overall turbidity removal efficiency. The comparison between actual and predicted values of turbidity removal, as illustrated in Figure 5(a), demonstrates a satisfactory agreement, indicating the statistical significance of the quadratic model employed. The calculated p-value of 0.0006 further implies the robustness of the model in accurately predicting turbidity removal efficiency.

Examination of the 3D model portrayed in Figure 4(b) discloses a significant trend: as the temperature spans from 40°C -60°C and transmembrane pressure (TMP) escalates within the 16-18 psi range, there is an augmentation in the efficiency of permeate flow rate. This observation posits a direct correlation between increased temperature and TMP levels and the enhancement in permeate flow rate. The comparison between actual and predicted values of turbidity removal, as delineated in Figure 5(b), reveals a satisfactory concurrence, underscoring the statistical significance of the quadratic model employed. The calculated p-value for permeate flow rate, which is less than 0.0001, underscores the robustness of the model and its ability to accurately predict permeate flow rate removal efficiency.

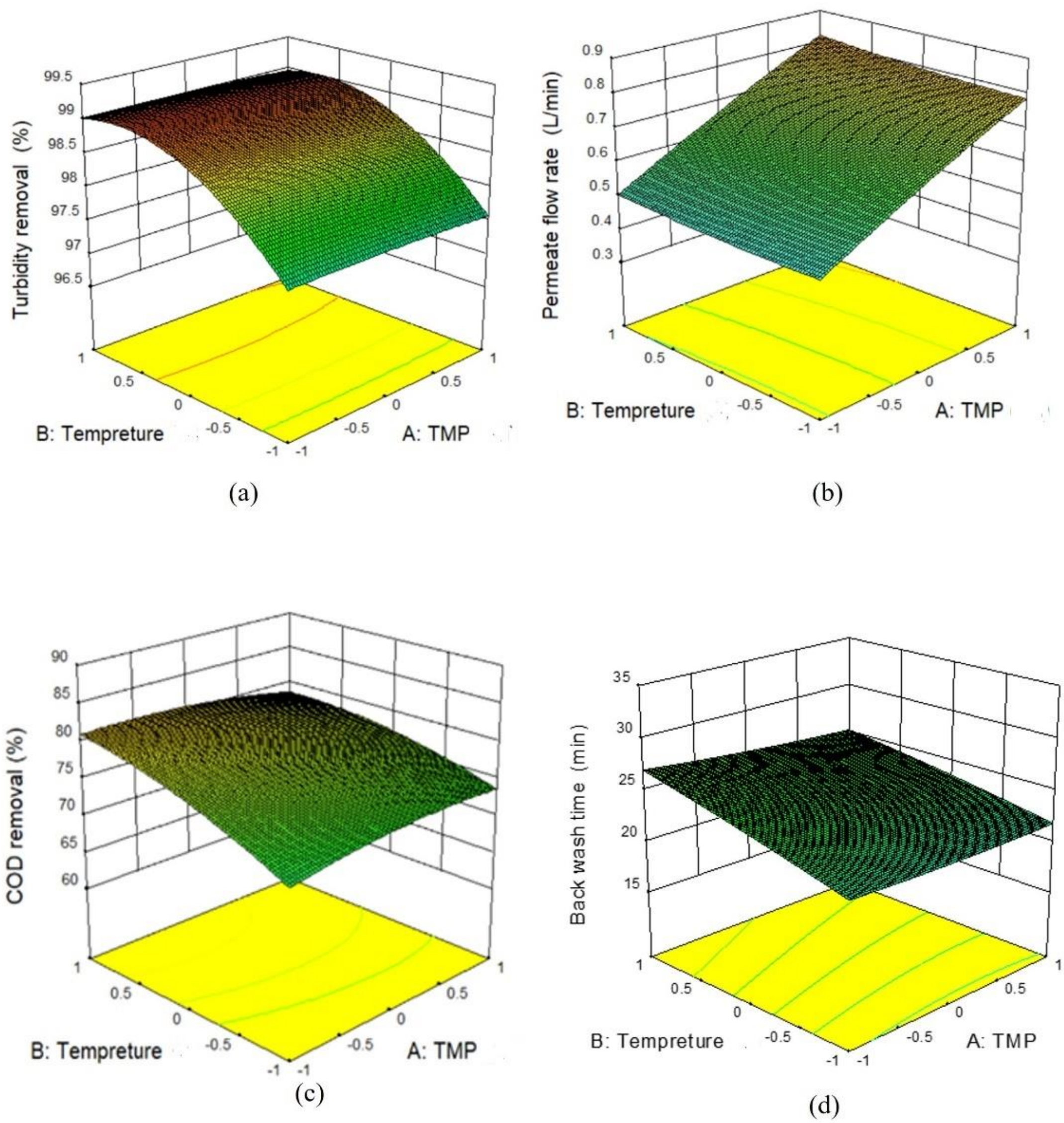

Figure 6.4: 3D profile of: (a) turbidity removal vs TMP and temperature, (b) Permeate flow rate vs TMP and temperature, (c) COD removal vs TMP and temperature, and (d) Backwash time vs TMP and temperature

The analysis of the 3D representation in Figure 4(c) reveals that as the temperature ranges between 40°C - 60°C, there is a notable escalation in the COD removal efficiency. This observation emphasises a direct correlation between increasing temperature and the augmented efficacy of the process in removing COD from LWW. Additionally, it is observed that TMP has a negligible impact on COD removal. When EPS dosage are increased, it has significant impact on COD removal. The maximum COD removal can be seen in the range of 10-15mg/L. Furthermore, the comparison between actual and predicted values of COD removal, as depicted in Figure 5(c), suggests a satisfactory alignment, thereby affirming the statistical significance of the quadratic model applied in this analysis. The calculated p-value of 0.036 further supports the statistical reliability of the model in accurately predicting COD removal efficiency.

Analysis of the 3D model in figure 4(d) unveils an influence of temperature (40-60°C) on the backwash efficiency, indicating an impact on the process ability to effectively clean the filtration membranes. Conversely, TMP is observed to have a negligible effect on backwash efficiency, suggesting its minimal contribution to the process. Moreover, it is evident that an increase in EPS dosage results in an extended backwash time, with the maximum duration occurring within the dosage range of 12-15 mg/L. This finding implies a direct relationship between EPS dosage and backwash duration, highlighting the importance of dosage optimization for efficient membrane cleaning. The comparison between actual and predicted values of backwash time, as presented in Figure 5(d), exhibits a satisfactory agreement, indicating the robustness and statistical significance of the quadratic model utilized for prediction. Furthermore, the low p-value of 0.0055 for backwash implies the statistical significance of the model in accurately predicting backwash time.

To meticulously ascertain the optimal parameters conducive to achieving heightened permeate flow rates, maximal turbidity and COD removal, as well as prolonged backwash duration, the following experimental conditions have been delineated: (a) TMP targeted at 18 psi, (b) Temperature maintained within a specified range, and (c) Dosages of EPS falling within predefined thresholds. Assigning high importance levels of 5/5 for maximum permeate flow rate and backwash time, and 4/5 for maximal turbidity and COD removal, these conditions were systematically assessed. Consequently, the optimal configuration emerged as TMP of 18 psi, temperature of 60˚C, and EPS dose of 14.17 mg/L, under this condition the predicted values of

0.82L/min for permeate flow rate, 98.98% for turbidity removal, and 80.83% for COD removal, with a backwash duration of 30 minutes. Upon experimentation, the observed values closely aligned with the predicted results, with a permeate flow rate of 0.81 L/min, turbidity removal of 98.95%, COD removal of 80.53%, and a backwash time of 31 minutes. The close alignment between predicted and experimental outcomes indicates satisfactory model performance.

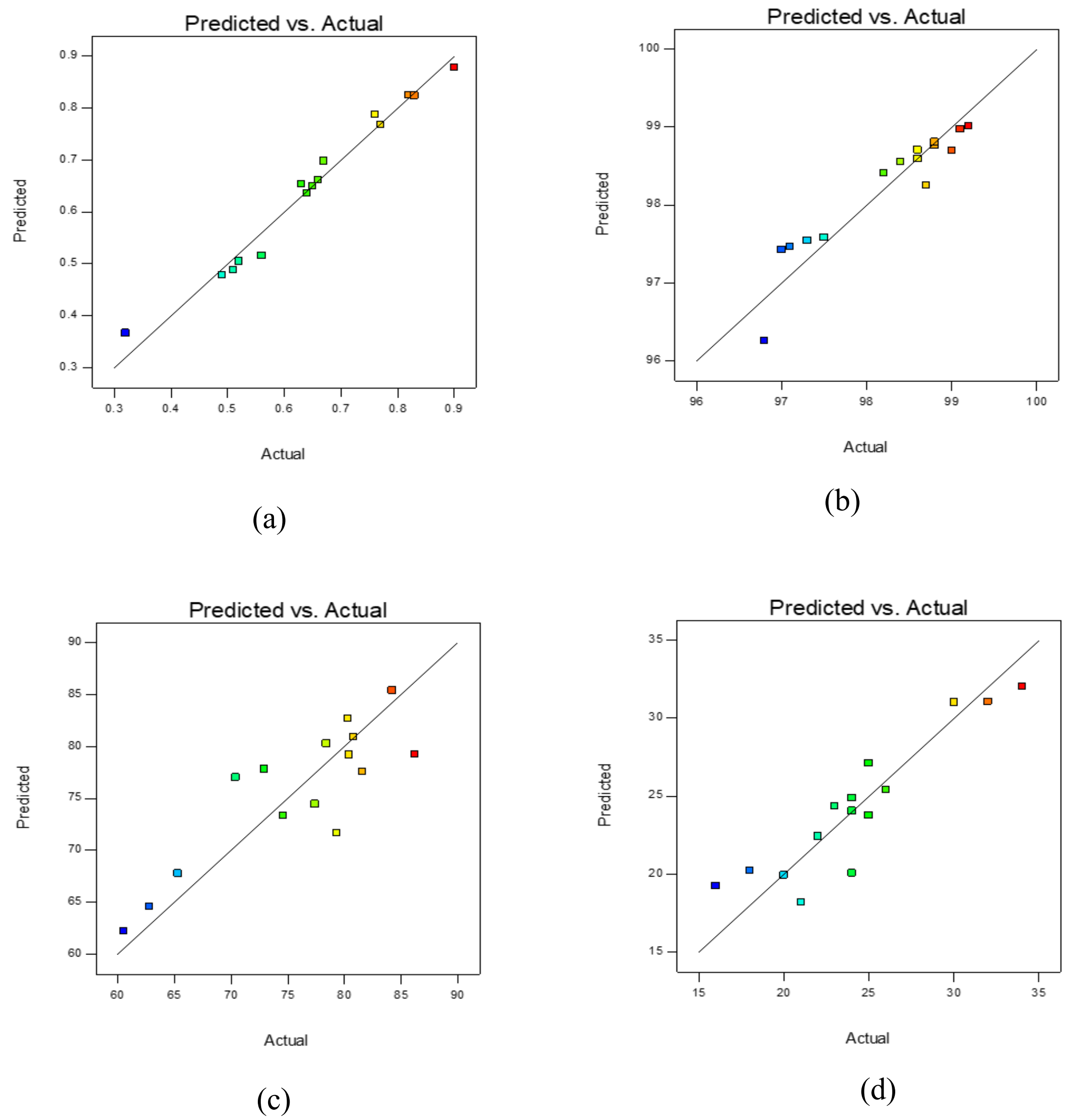

Figure 6.5: comparison of predicted vs actual for: (a) Permeate flowrate, (b) Turbidity removal, (c) COD removal and (d) Backwash time

Fouling behavior of UF membrane

The investigation into the fouling behavior of UF membrane with the incorporation of EPS presents intriguing findings with significant implications for process efficiency and membrane performance enhancement. The results indicate a noteworthy improvement in process efficiency when EPS is used in combination with UF membrane. This finding is a pivotal development, demonstrating the potential of EPS to mitigate fouling and enhance overall process performance. Such enhancements are crucial for optimizing UF processes, particularly in applications where membrane fouling poses a significant operational challenge.

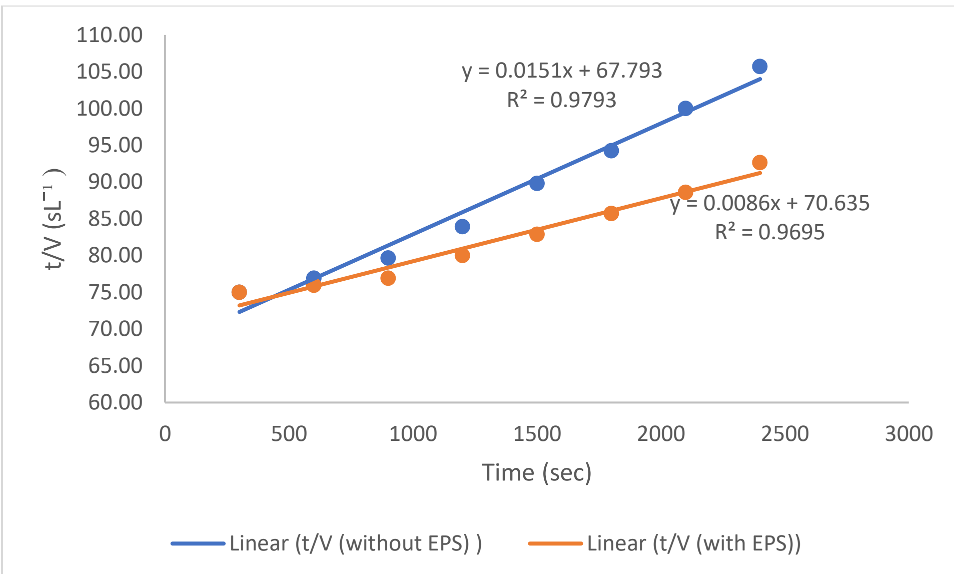

Figure 6.6: Graph of t/V vs time

The graphical representation of fouling behavior, depicted in the figure 6, provides valuable insights into the dynamic nature of membrane fouling with and without EPS. The distinct slopes of the lines corresponding to UF with and without EPS signify differences in fouling kinetics and membrane resistance buildup over time.

At the initial stages of the process, the fouling behavior appears comparable between UF with and without EPS, indicating similar trends in membrane fouling initiation. However, as the process progresses, the divergence in fouling behavior becomes evident. The steeper slope observed in the line representing UF without EPS suggests a more rapid increase in membrane resistance compared to UF with EPS. This observation implies that the incorporation of EPS mitigates fouling and slows down the rate of membrane fouling progression over time.

By forming a protective layer on the membrane surface, EPS interferes with foulant-membrane interactions, acting as a barrier that reduces foulant adhesion and prevents deposition onto the membrane (Luo *et al.*, 2022). Additionally, EPS exhibits inherent properties, including hydrophilicity, surface charge, and molecular conformation, which promote fouling resistance by facilitating foulant repulsion and enhancing ease of removal during cleaning operations (Nouha *et al.*, 2016). Furthermore, EPS can modify the surface properties of UF membranes, such as surface roughness, hydrophobicity/hydrophilicity, and surface charge, thereby altering the interaction forces between foulants and the membrane surface, ultimately reducing fouling propensity and enhancing overall fouling resistance (Kumar *et al.*, 2023).

The observed differences in fouling behavior highlight the potential of EPS as a promising additive for mitigating membrane fouling in UF processes. By slowing down the accumulation of fouling layers on the membrane surface, EPS can prolong membrane lifespan, improve process efficiency, and reduce the frequency of cleaning and maintenance operations. Moreover, the utilization of EPS may offer cost-effective solutions for enhancing UF performance, particularly in applications where fouling poses operational challenges.

Treatment of LWW concentrate using PBEC:

Table 6.6: Characteristic of concentrate from UF and effluent

	pH	Conductivity (µS/cm)	Turbidity (NTU)	TSS (mg/L)	TS (mg/L)	COD (mg/L)	NPEOs (µg/L)
LWW concentrate	10±0.5	610±20	301±5	303±10	2856±30	2089±50	760±50
After treatment with PBEC	9.5±0.5	580±20	2±0.5	≤ 1	5±0.7	130±20	350±50

The characteristic of concentrate generated from UF, as shown in table 6 indicates that it is loaded with high suspended solids, COD and NPEOs. Due to high concentration of suspended solids, concentrate is loaded with high turbidity. This emphasize that proper management of concentrate is needed. Treatment using PBEC combined with EPS indicates that, the process is self sufficient in removing Turbidity, TSS, TS and COD. The result also indicates that the packed bed electrocoagulation is not highly effective in removing NPEOs from the concentrate.

The comparison between Electrocoagulation (EC) treatment and PBEC treatment for the removal of contaminants from the UF concentrate reveals notable differences in efficacy. In previous studies utilizing EC, substantial reductions in turbidity, TSS, and COD were achieved, with removal efficiencies of approximately 80%, 70%, and 60%, respectively (Elazzouzi *et al.*, 2018; Oktiawan *et al.*, 2021; Prajapati *et al.*, 2016). However, the removal of NPEOs remained limited, with only a minimal reduction of around 20% (Oktiawan *et al.*, 2021; Samadikun *et al.*, 2021). In contrast, the application of PBEC in the present study resulted in remarkable improvements across all parameters. Turbidity, TSS, TS, and COD were reduced to extremely low levels, with removal efficiencies of 99.34%, 99.57%, 99.70% and 87.60% respectively. The removal of NPEOs is limited but approximately 53.95% with PBEC. This significant enhancement in contaminant removal can be attributed to the unique configuration of the PBEC reactor, which optimizes the electrocoagulation process. Additionally, the incorporation of sacrificial iron scrap and the utilization of EPS further augment coagulation and flocculation processes, leading to enhanced contaminant removal. The substantial reduction in NPEOs, a challenging class of contaminants to treat, highlights the effectiveness of PBEC in addressing a wide range of pollutants present in the concentrate. Overall, the findings highlight that PBEC is more efficient than the conventional EC methods in achieving comprehensive and efficient treatment of UF concentrate, offering promising solutions for sustainable water management practices.

Cost Estimate

For LWW treatment employing UF and EPS, the total treatment cost estimation (0.865\$CAN/m^3) indicates an energy expenditure of 0.010\$CAN per cubic meter and 0.670\$CAN per cubic meter for EPS for the UF process. On the other hand, for PBEC (UF concentrate management), the cost is evaluated at 0.170\$CAN per cubic meter, including 0.010\$CAN per cubic meter for energy and 0.005\$CAN per cubic meter for polymer in PBEC. Notably, the energy expenditure in PBEC aligns with that of UF in LWW treatment, indicating a comparable efficiency in energy utilization between the two processes.

6.7. Conclusion

In this study, the comprehensive characterization of bacterial EPS and commercial LWW led to significant insights into their properties and treatment potential. The findings have several implications supported by existing scientific literature and provide a deeper understanding of wastewater treatment processes.

The EPS generated by bacterial strain BS-04 exhibited a significant protein content of 1.45g/L, with specific properties such as a viscosity of 30.99mPas, a zeta potential of -38.2mV, a flocculation activity of 82%, and a dewaterability index of 40.21%. These properties make EPS an effective agent for wastewater treatment, as demonstrated by its ability to enhance flocculation and dewatering processes.

Analysis of LWW revealed elevated levels of suspended solids (60 ± 10 mg/L), alkalinity (pH 10.34 ± 0.5), conductivity (496 ± 20 µS/cm), and organic compounds (COD 576.7 ± 30 mg/L), suggesting the environmental risks associated with its discharge. The high turbidity (109 ± 5 NTU) and COD levels are consistent with findings from similar studies, indicating the prevalence of suspended particles and organic pollutants in laundry effluents.

Factorial design experiments elucidated the influence of various factors on LWW treatment, highlighting the critical role of temperature and EPS concentration. Temperature emerged as the dominant factor influencing turbidity removal (Y_2), contributing approximately 89.67% to the observed variation in the response. Furthermore, the significant contribution of EPS concentration to COD removal (Y_3) underscores the importance of EPS in facilitating organic pollutant removal through enhanced floc formation and sedimentation.

The optimization of treatment conditions using central composite design methodology resulted in predicted values closely aligned with experimental outcomes, indicating the robustness of the models developed. The ability to accurately predict treatment performance based on experimental parameters enhances the efficiency and reliability of wastewater treatment processes, contributing to the advancement of sustainable water management practices. For the optimized treatment of LWW using UF with EPS, the conditions included TMP of 18 psi, temperature of 60°C, and EPS dose of 14.17 mg/L. These conditions resulted in a permeate flow rate of 0.81 L/min, turbidity removal of 98.95%, COD removal of 80.53%, and a backwash time of 31 minutes.

The incorporation of EPS mitigated fouling in ultrafiltration membranes, offering promising solutions for improving process efficiency and prolonging membrane lifespan. By minimizing fouling, EPS enhances the operational stability and cost-effectiveness of membrane filtration systems, facilitating their widespread application in wastewater treatment.

Comparison between conventional EC and PBEC for UF concentrate treatment highlighted the superior performance of PBEC. PBEC achieved remarkable reductions in turbidity (2 ± 0.5 NTU), TSS (≤ 1 mg/L), TS (5 ± 0.7 mg/L), COD (130 ± 20 mg/L), and NPEOs (350 ± 50 µg/L), showcasing its efficacy in comprehensive contaminant removal. The incorporation of sacrificial iron scrap and EPS further enhanced coagulation and flocculation processes, emphasizing PBEC's potential for sustainable water management practices.

The cost estimation (0.865$CAD/m^3) for LWW treatment employing UF and EPS indicated an energy expenditure of 0.010$CAN per cubic meter and 0.670$CAN per cubic meter for EPS for the UF process. On the other hand, for PBEC, the cost was evaluated at 0.170$CAN per cubic meter, including 0.010$CAN per cubic meter for energy and 0.005$CAN per cubic meter for polymer in PBEC. Notably, the energy expenditure in PBEC aligned with that of UF in LWW treatment, indicating comparable efficiency in energy utilization between the two processes.

The study provided valuable insights into the optimized treatment conditions and cost-effectiveness of different methods for LWW treatment, emphasizing the potential of EPS in UF and innovative techniques like PBEC for treating the UF concentrate in addressing environmental challenges and promoting sustainable water management practices.

6.8. Future Perspectives

Future research could focus on further enhancing the removal efficiency of NPEOs from laundry wastewater concentrate using advanced oxidation processes (AOPs) or adsorption techniques.

Another important aspect for future consideration is the management of sludge generated during UF concentrate treatment using PBEC. Sludge management is crucial for ensuring environmental sustainability and compliance with regulations. Future research could explore innovative approaches for sludge dewatering, treatment, and resource recovery. Techniques such as sludge conditioning, mechanical dewatering (e.g., centrifugation, filtration), and advanced thermal or biological treatment processes for sludge stabilization could be investigated.

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Author Contributions

Sushil Kumar, Patrick Drogui, and R D Tyagi conceived and planned the idea of the study. Sushil Kumar experimented and collected the data. All authors carried out the analysis and contributed to the interpretation of the results. Sushil Kumar wrote the manuscript in consultation with Ali Khosravanipour Mostafazadeh, Patrick Drogui, and R D Tyagi. All authors read and approved the final manuscript.

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Availability of data and materials

All data generated or analyzed during this study are included in this published article.

Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

All authors read and approved the final manuscript and mutually agreed that it should be submitted to Journal of Environmental Management.

Competing interests

The authors declare that they have no conflicts of interest to declare.

7. CONCLUSION AND PERSPECTIVES

7.1. Conclusion

This study provides a comprehensive characterization of bacterial EPS and commercial LWW, yielding significant insights into their properties and treatment potential. The EPS produced by bacterial strain BS-04 exhibited a notable protein content of 1.45g/L, along with specific properties such as viscosity (30.99mPas), zeta potential (-38.2mV), flocculation activity (82%), and dewaterability index (40.21%). These characteristics render EPS an effective agent for CLWW treatment, as evidenced by its capacity to enhance flocculation and dewatering processes. Importantly, the absence of significant carbohydrate content in EPS suggests a potential advantage in terms of stability and longevity in treatment applications (Hu et al., 2023).

Analysis of commercial LWW revealed elevated levels of suspended solids (60 ± 10 mg/L), alkalinity (pH 10 ± 0.5), conductivity (496 ± 20 µS/cm), and organic compounds (COD 576 ± 30 mg/L), indicating environmental risks associated with its discharge. The high turbidity (109 ± 5 NTU) and COD levels are consistent with findings from similar studies, suggesting the prevalence of suspended particles and organic pollutants in laundry effluents (Chen et al., 2015).

During CLWW treatment using EPS it was observed that out of the three common chemical coagulants (Alum, $FeSO_4$, and $CaCl_2$), alum (2000 mg/L) achieved the highest flocculation activity (99.18 %) and pollutant removal rates (SS-98%, TS- 91.33%, COD-85.1 %) for CLWW treatment at pH 7 and treatment time of 30 min. It was observed that S-EPS (0.6 g/L) gave better FA than B-EPS. S-EPS alone can remove 83.0±1% of turbidity, 77±1.5% TSS, and 76±1% COD. When S-EPS (0.6 g/L) was used in combination with alum (300 mg/L) at pH 7 and a treatment time of 30 min, the removal of turbidity, SS, and COD reached 98.09±0.9%, 95.42%, and 83.08±1.1%, respectively. When treatment time has been increased to 4h, it resulted in more than 88.00% COD removal) from CLWW.

The results of statistically optimised PBEC process for treating commercial LWW demonstrate effective removal of turbidity (98 ± 2%), TS (95 ± 3%), COD (89 ± 5%), and NPEOs (53 ± 2%) under optimized conditions of current intensity: 2.99A, treatment time:

58.8 minutes and enhanced EPS dose from 5.8 mg/L to 8.0 mg/L. The economic estimate of this study under optimized conditions highlights the dominance of energy consumption as the primary expense, totaling 1.20$CAN/m^3, with energy costs alone contributing 0.86$CAN/m^3. While electrode and polymer expenses were comparatively minimal, optimizing energy efficiency remains paramount to mitigating overall costs.

Factorial design experiments elucidated the influence of various factors on commercial LWW treatment, highlighting the critical role of temperature and EPS concentration. Temperature emerged as the dominant factor influencing turbidity removal (Y_2), contributing approximately 89.67% to the observed variation in the response. The dominance of temperature in influencing turbidity removal aligns with existing research demonstrating the temperature dependence of coagulation-flocculation processes in wastewater treatment (Wang et al., 2023). Furthermore, the significant contribution of EPS concentration to COD removal (Y_3) underscores the importance of EPS in facilitating organic pollutant removal through enhanced floc formation and sedimentation (Luo et al., 2022).

Optimization of treatment conditions using central composite design methodology resulted in predicted values closely aligned with experimental outcomes, indicating the robustness of the models developed. The ability to accurately predict treatment performance based on experimental parameters enhances the efficiency and reliability of CLWW treatment processes, contributing to the advancement of sustainable water management practices (Liu et al., 2022). For the optimized treatment of commercial LWW using UF with EPS, the conditions included TMP of 18 psi, temperature of 60°C, and EPS dose of 14.17 mg/L. These conditions resulted in a permeate flow rate of 0.81 L/min, turbidity removal of 98.95%, COD removal of 80.53%, and a backwash time of 31 minutes.

Incorporation of EPS mitigated fouling in ultrafiltration membranes, offering promising solutions for improving process efficiency and prolonging membrane lifespan. By minimizing fouling, EPS enhances the operational stability and cost-effectiveness of membrane filtration systems, facilitating their widespread application in CLWW treatment (Estahbanati et al., 2021).

PBEC achieved remarkable reductions in turbidity (2 ± 0.5 NTU), TSS (≤ 1 mg/L), TS (5 ± 0.7 mg/L), COD (130 ± 20 mg/L), and NPEOs (350 ± 50 µg/L), showcasing its efficacy in comprehensive contaminant removal. The incorporation of sacrificial iron scrap and EPS further enhanced coagulation and flocculation processes, emphasizing PBEC's potential for sustainable concentrate management practices.

Cost estimates (0.865$CAD/m3) for commercial LWW treatment employing UF and EPS indicated an energy expenditure of 0.01$CAN per cubic meter and 0.67$CAN per cubic meter for EPS for the UF process. On the other hand, for PBEC, the cost was evaluated at 0.17$CAN per cubic meter, including 0.01$CAN per cubic meter for energy and 0.005$CAN per cubic meter for polymer in PBEC. Notably, the energy expenditure in PBEC aligned with that of UF in CLWW treatment, indicating comparable efficiency in energy utilization between the two processes.

This study provides valuable insights into the optimized treatment conditions and cost-effectiveness of different methods for CLWW treatment, emphasizing the potential of EPS in UF and innovative techniques like PBEC for treating the UF concentrate in addressing environmental challenges and promoting sustainable water management practices.

7.2. Perspectives

By capitalizing on the rich findings and insightful perspectives of this comprehensive study, several promising avenues and additional scientific explorations emerge in the field of laundry wastewater (LWW) treatment. However, it is crucial to conduct further work to optimize the efficiency of this hybrid process:

- Perform a dynamic study of the EC-ELF process to understand the fluid flow within the electrolytic reactor (CFD study).
- Further optimize and investigate the mechanism of NPEO removal by EPS. Future studies can focus on enhancing the adsorption capacity of EPS through surface modification techniques and exploring microbial consortia capable of degrading NPEOs with EPS support.
- Include in subsequent studies the combination of electro-catalytic oxidation processes and EPS. In situ generation of oxidizing species (OH, H_2O_2, $H_2S_2O_8$, etc.) and the use of catalytic electrodes offering the largest effective surface area possible

(deployed metal electrodes) are key elements for effectively oxidizing NPEOs. However, when the electrochemical oxidation process is limited by the mass transfer of electro-active species (e.g., NPEO present in trace amounts in water), it may be advantageous to pre-concentrate pollutants through solid/liquid separation, using EPS, for example, as a bio-flocculant.

- Test and pre-industrially scale up the hybrid process: Conduct further research to optimize the process combining EPS, electrocoagulation, and ultrafiltration for large-scale application.
- Evaluate the life cycle assessment (LCA) of the hybrid process: Conduct a comprehensive life cycle assessment of the entire wastewater treatment process, including the use of EPS, electrocoagulation, ultrafiltration, and all subsequent treatment stages. Assess the environmental impacts associated with each step of the process, from resource extraction to waste generation, to identify potential areas for improvement and guide future sustainability efforts.
- Analyze the costs and economic viability of the hybrid process: Conduct a detailed cost analysis encompassing expenses related to the implementation and operation of the multimodal treatment approach. Evaluate initial costs, operational expenses, maintenance, as well as potential savings or benefits arising from treated water or recovered resources. This analysis will help determine the economic feasibility and competitiveness of the approach.

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