Treatment of Commercial Laundry Wastewater Using Extracellular Polymeric Substances (EPS)

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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 extracellular polymeric substance (EPS) as bio-flocculant. EPS was produced from 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 treatment of commercial laundry wastewater (CLWW). This is the first study for 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 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% suspended solids (SS) and 76.37% chemical oxygen demand (COD). The best results were obtained by combining S-EPS (0.6 g/L) together with alum (300 mg/L) at pH 7 for 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.

Introduction

There has been a persistent presence of the 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, detergents along with the 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 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; Estahbanati et al., 2021; 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; Mollah et al., 2001; 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., 2012; 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., 2014; Kamarudin et al., 2003; 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 for explicit application for LWW treatment (Bokhary et al., 2018; Kumar et al., 2021).

Anaerobic treatment and membrane bioreactors are potential methods for treatment of CLWW (Braga et al., 2015; Delforno et al., 2020; Faria et al., 2019; Moura et al., 2019). However, challenges associated with them consist of 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 the chemical flocculants have been found in literature (Šostar-Turk et al., 2005; Terechova et al., 2014). The removal of particulate matter along with phosphorus and heavy metals have been effectively performed by the easy and economical coagulation (alum and ferrous sulphate) 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 minimise 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 are harmless, sustainable, biodegradable and pose 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 growth phase. They have been investigated as a flocculating agent for dewatering and sludge settling (Huang et al., 2019; Kaur et al., 2019; 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 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). Study achieved removal efficiency of 84% for COD, 96% for phosphorus, 97% for nitrates-nitrites, 75% for ammoniacal nitrogen, 29%-99% for metals and 95% for colours (Kaur et al., 2019). In another study, when EPS was used in combination with $FeSO_4$ for the treatment of landfill leachate. During the Jartest 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 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 the 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).

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 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 the cold room at temperature of -4ºC.

The commercial laundry wastewater was characterised 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 dry weight of S-EPS the supernatant obtained was mixed with ethanol (95%v/v) in 1:2 ratio and kept at -20ºC for 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 the Eq. (1):

$\text{EPS (g/L)} = (W_2 – W_1)/V$ (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-ESP) was determined by mixing the biomass pellets (obtained by centrifugation of fermented broth) with water in equal volume and then mixture was heated at 60ºC in water bath for 60 min to release the C-EPS in the liquid phase. Released C-EPS in 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 the Eq. (2):

B-EPS (g/L) = S-EPS (g/L) + C-EPS (g/L) (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 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 particles surface. Opposite charge makes adsorbed bio-flocculants to act as patching point for nearby particles.

CLWW treatment

Identification of optimized pH for chemical coagulants

The jar test experiments were conducted for ferrous sulphate, 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 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):

$$FA = (B - A)*100/B \quad (3)$$

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, 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 1 L glass cylinder for settling. After 30 min of settling, the supernatant was collected and analysed 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 were 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 concentration (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 were considered for further optimization. After the jar test experiment followed by 30 min settling, supernatant was analysed (Turbidity, SS, TS, COD, nitrogen, phosphorous, metals and NPEOs) to determine the pollutant removal efficiency.

Combination of EPS and Chemical Coagulant

To enhance the performance of EPS, chemical coagulant was used in combination with EPS. During the LWW treatment process, suitable pH (as determined in section of optimization of pH for EPS) and chemical coagulant (as determined in 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 was 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 analysed to determine the removal efficiency of different parameters (Turbidity, SS, COD, nitrogen, phosphorous, metals and NPEOs).

Effect of treatment time

After 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 suitable concentration of S-EPS (as determined in section of optimization of EPS concentration) was added to each beaker. Then pH was adjusted to a suitable value (as determined in section of 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 12 h) and treated water was set aside to settle by gravity for 30 min. After settling, the supernatant sample was analysed 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 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 350°C, CEAEQ (MA 400 – NPEO 1.0)
Metals	ICP-AES

Results And Discussion

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 high amount of turbidity and SS was as 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). Small amount of heavy metals was 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 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	10.38 ± 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)	579.3 ± 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 ± .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

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 initial increase in zeta potential (during pH 3–7) and then it gradually decreased with increase in pH from 7 to 11. The increase of 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 were obtained at pH 3, 5, 7, 9 and 11 respectively. Similarly, the flocculation activity (FA) also increased with increase in pH up to pH 7 and then it decreased with further increase in pH. At pH 3, only small flocculation activity (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 during pH 3 to 11. Zeta potential values of − 7.7 mV, − 8.3 mV, − 9.4 mV, − 9.7 mV and − 3.9 mV were found at pH 3, 5, 7, 9 and 11 respectively. The maximum flocculation activity 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 increase in pH (3 to 5) and then became almost constant (during pH 7 to 9). Zeta potential values of − 10.7 mV, − 10.3 mV, − 10.7 mV, − 11.7 mV and − 13.2 mV were found at pH 3, 5, 7, 9 and 11 respectively. The maximum flocculation activity of 29% was observed at pH 5. Figure 2(a) and (b) shows impact of pH on flocculation activity using chemical coagulants and EPS, respectively.

The results for FA for different coagulants clearly indicate that pH has 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 coagulation-flocculation process is widely dependent on the pH. It imparts 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 optimized pH for the future experiments.

The main factors behind 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 removal of pollutants from the wastewater sample.

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, zeta potential value also increased from − 12.8 to 1.8 mV, respectively. With increase in alum concentration, FA also increased from 21.82–99.18%. 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 14.09–91.09% by increasing $FeSO_4$ dosage. When 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 4.55–22.45%. 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, lower coagulant dose may result in formation of small flocs and can interfere the settling of the particles. This may also result in a decrease in flocculation activity. Similarly, 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 was removed from CLWW at 2000 mg/L dose of $FeSO_4$ 2000. 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) were found to be 36.18%, 29.17% and 36.09% respectively.

Table 3
Treatment of CLWW using different concentration 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
$FeSO_4$	100	14.09	12.67	9.60	6.23
	300	23.36	22.67	20.39	18.47
	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 8.17–85.10% for alum and 6.23–47.80% for $FeSO_4$, respectively. However, $CaCl_2$ gave slightly different performance. When dosage of $CaCl_2$ was increased from 100 to 1500 mg/L, COD removal enhanced. But when dosage of $CaCl_2$ increased further from 1500 to 2000 mg/L, COD removal decreased. The maximum COD removal (42.60%) was obtained at $CaCl_2$ concentration of 1500 mg/L. The reason behind the lower COD removal in 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, 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

In order 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 increase in pH from 5 to 11. Zeta potential values of − 8.9, − 1.4, − 2.1, − 8.7 and − 10.9 mV were obtained at pH 3, 5, 7, 9 and 11, respectively. Similarly, the FA also increased while increasing the pH 3 to 5. Then, FA remained almost constant till pH 7, and decreased with 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, positives and negatives charges of particles gets neutralized, resulting in higher in probability of combining the dispersed and/or colloids particles. However, when B-EPS was used as bio-flocculant the zeta potential value increases with increase in pH up to 5, and almost constant from 5 to 7 pH, and then decreases with increase in pH 11. Zeta potential values of − 11.2 mV, − 8.1 mV, − 8.3 mV, − 10.9 mV and − 13.1 mV were found at pH 3, 5, 7, 9 and 11 respectively. Similarly, the FA also increased with increase in pH upto 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 comparable flocculation activity (65.90% and 29.50% respectively). Figure 2(a), (b), (c) and (d) shows 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 clearly indicate that pH has significant role in floc formation and removal of pollutants from CLWW. 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 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 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 contains many functional groups. The structure and functional groups may change with the microorganism used for fermentation, type of carbon substrate and, 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 possibly affect the FA of B-EPS due to the fact that it contains hydrophilic compound (hydroxyl groups) (Tian et al., 2006). B-EPS also contains organic, inorganic, and dead cells, etc. which may also affect the overall performance of B-EPS.

Effect of EPS concentration

Different concentration 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 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, respectively. Similarly, the FA also increased with the increased in S-EPS concentration. The maximum FA of 83.27% was attained at S-EPS dose of 0.6 g/L (Fig. 2c). However, when B-EPS was used the zeta potential value were − 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 EPS concentration of 0.6 g/L.

Combining EPS and chemical coagulant for the treatment of CLWW

Experiments were also conducted using EPS in 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 significant role in removal of pollutants from CLWW samples. The FA increased with 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 combination of EPS and alum has been depicted in Fig. 2c. Similarly, when B-EPS was used in combination of 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 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.

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 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. 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 insufficient bridging of the particles and resulted in low settling of the suspend material. These results are in agreement with those reported on composting leachate treatment with EPS (Kaur et al., 2019).

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, combination of B-EPS (0.6 g/L) and alum (300 mg/L) were able to remove 67.20% of COD from wastewater sample.

Table 4
Treatment of CLWW using EPS alone and in combination of 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 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 initial S-EPS dose of 0.1 g/L, only 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 rise in B-EPS concentration (Table 5). At 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 metals removal was increased significantly with the increase in EPS concentration. For combination of S-EPS and alum (0.1 g/L and 300 mg/L), only small concentration (2–13%) of metals were 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 of alum, it also shows 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 removal of Na (75.93%) and Ca (81.62%) was better than removal of Al (52.52%) and Mg (69.62%).

Table 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
CLWW + Alum	0.1	6.34	5.80	6.38	2.33	13.23	3.49	4.94	4.10	1.76	9.40
	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

Removal of metals is due to the fact that EPS posses several adsorption sites for metals, which consists of proteins with aromatics and aliphatic sites and carbohydrates with hydrophobic sites (More et al., 2014). The EPS contains high amount of polysaccharides and proteins, which play very important role in metals removal. Studies indicates 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 C-O-C group (ether) with polysaccharides, C = O group with phenolic alcohol, -OH group with alcohol and phosphorous and sulphur groups also contributes in complex reactions. Apart from this, presence of phosphorus group in nucleic acid (particularly DNA) which is present in EPS also provides additional binding sites for metals. Additionally, nucleotides and uronic acids containing phosphorous group available in EPS are having negative charge and it can effectively attach with multivalent positive charged ions resulting in metals removal (Nouha et al., 2018). It has been also reported that S-EPS contains higher portion of proteins as compared to B-EPS (Pan et al., 2010). Therefor, the amount of protein and its types play significant role in the flocculation process.

Effect of treatment time for CLWW treatment:

The impact of treatment time on flocculation activity is depicted in Fig. 2d. It was observed that as the treatment time increased, it has 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 of alum (300 mg/L), it was found that 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 significant impact on turbidity removal rate for S-EPS (Fig. 3a). Similarly, the concentration of SS, TS and COD also decreased with 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 removal of metals like Al and Mg increased significantly with increase in treatment time (Fig. 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 (Fig. 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 of 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 significant amount of metal removal was achieved with increase in treatment time (Fig. 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 (12 h of treatment time).

Conclusion

This work concludes that the biopolymer (EPS) produced from 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 the 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 treatment time of 30 min, the removal of turbidity, SS and COD reached to 98.00%, 95.42% and 83.08%, respectively. When treatment time has been increased to 4 h, it resulted in more than 88.00% COD removal) from CLWW.

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 Environmental System Research.

Competing interests

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

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Author contributions

Sushil Kumar, Ali Khosravanipour Mostafazadeh, Patrick Drogui, R D Tyagi conceived and planned the idea of the study. Sushil Kumar performed the experiment 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 analysed during this study are included in this published article.

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Figures

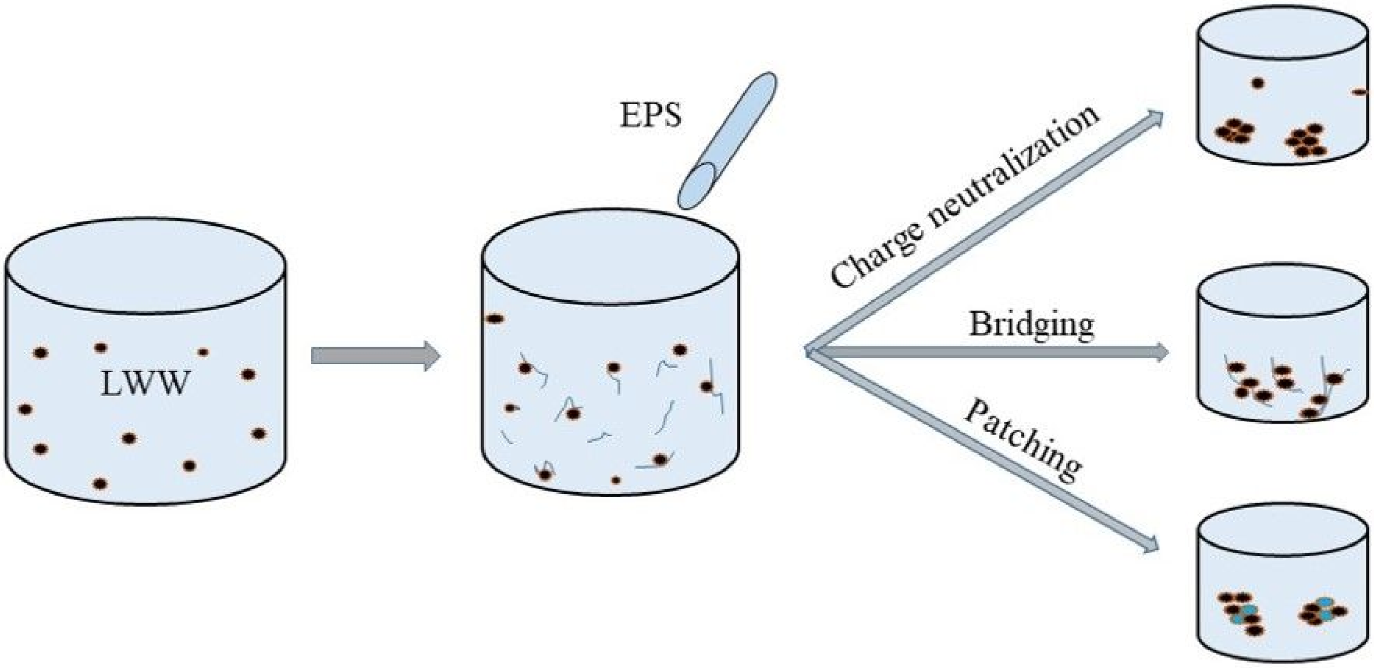

Figure 1

Mechanism of LWW treatment process

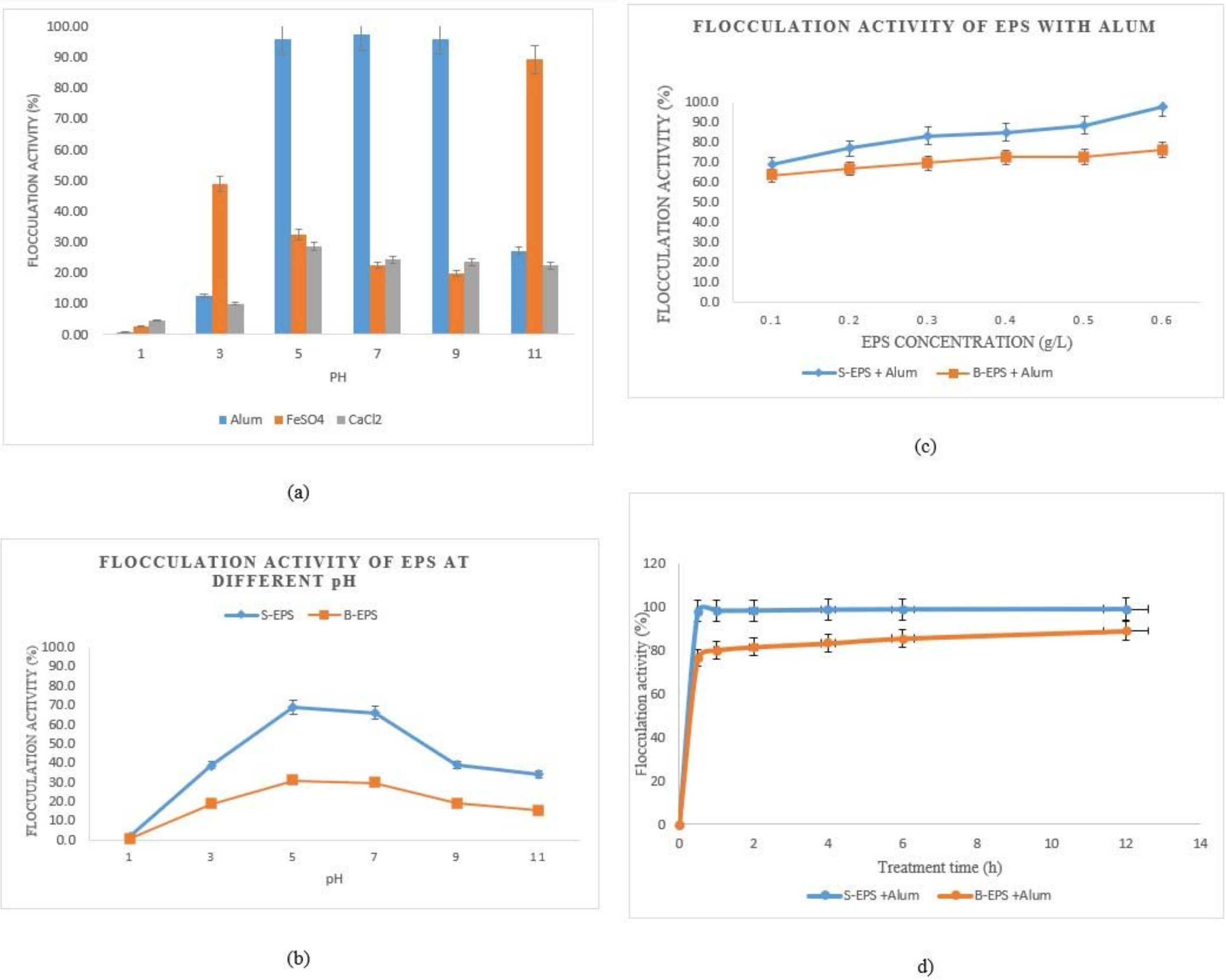

Figure 2

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

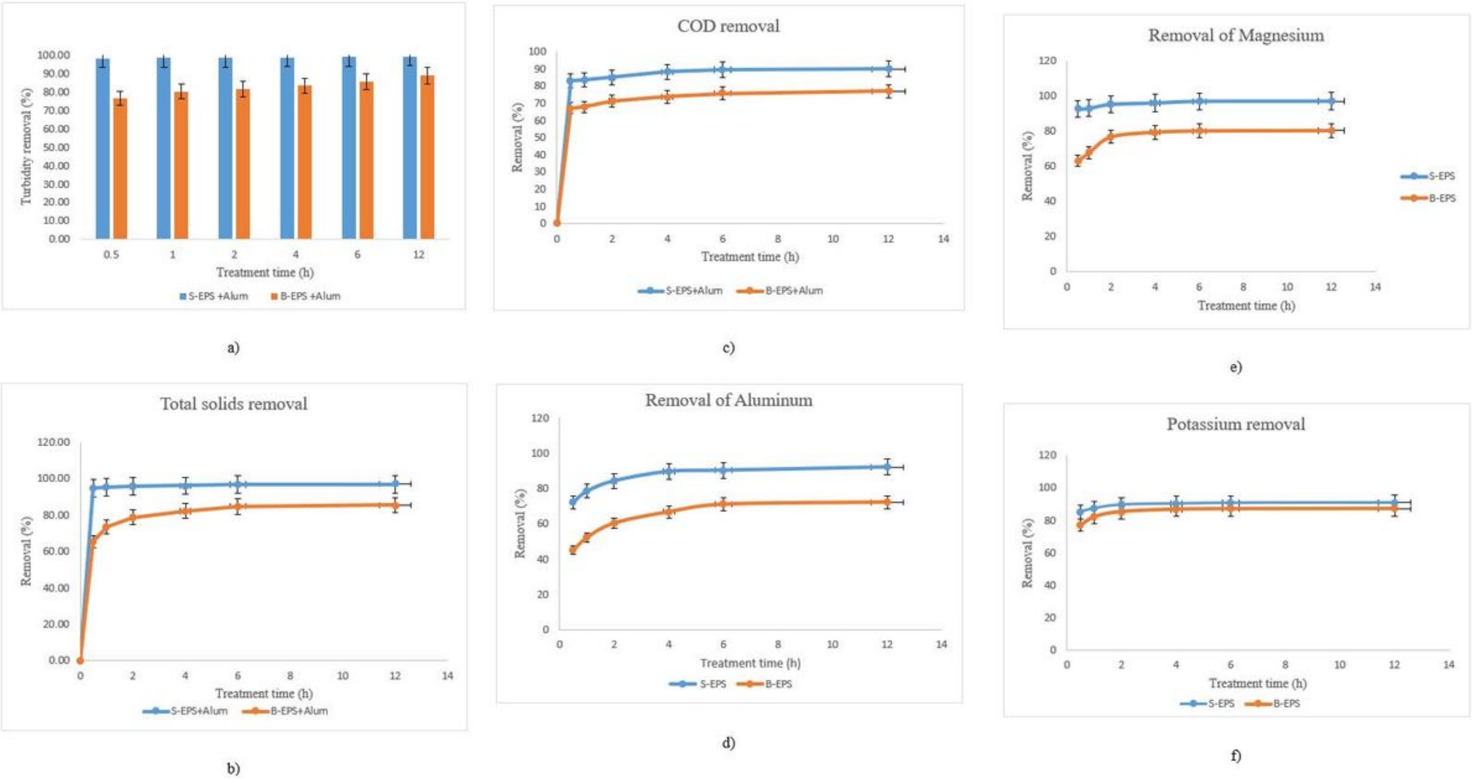

Figure 3

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

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