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Treatment of laundry wastewater using extracellular polymeric substances (EPS)

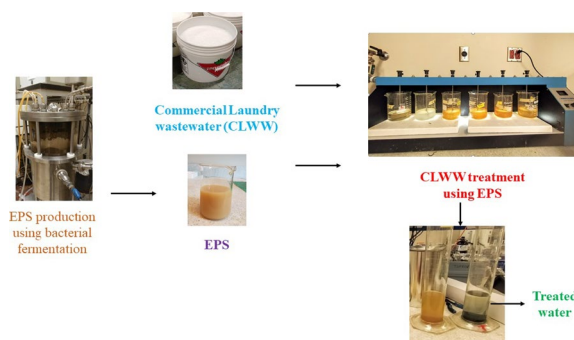
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Abstract

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 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)

Graphical Abstract



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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 m³ 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; Elazouzi 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, 2021); biological processes (Andersen et al. 2002; Ashfaq et al. 2017; Bagheri & Mirbagheri 2018; Deowan et al. 2015; Emaminejad et al. 2019; Hamed 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; Hamed 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 with an explicit application for LWW treatment (Bokhary et al. 2018; Kumar et al. 2021).

Anaerobic treatment and membrane bioreactors are potential methods for the 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 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; Hamed 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. 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) (Lapidou & 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 90 s. 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 LWV (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 LWV treatment. Three chemical coagulants (Alum, FeSO_4 , and CaCl_2) were selected and used for the treatment of commercial laundry wastewater (CLWV).

Materials and methodology

Origin and characterization of commercial laundry wastewater (CLWV)

Raw laundry wastewater (LWV) 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 LWV effluent was stored in 5 buckets of 20 L each, over a period of 24 h for further analysis. The samples were stored in a cold room at a temperature of -4°C .

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 4°C 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 -20°C overnight for phase separation. The precipitates (solid phase) of S-EPS were collected by centrifuging at 6000 g at 4°C for 20 min. The

centrifuged pallet of S-EPS was dried at 60°C to a constant weight.

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

$$\text{EPS (g/L)} = (\text{W}_2 - \text{W}_1)/\text{V} \quad (1)$$

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 60°C 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 4°C for 20 min.

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

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

The collected S-EPS and B-EPS in liquid form were used for LWV treatment. Figure 1 shows the mechanism of CLWV 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.

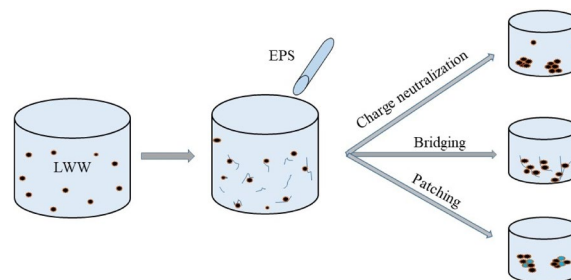


Fig. 1 Mechanism of LWV treatment process

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 H₂SO₄) 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 s. 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):

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

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, 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 s 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 s 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 LW/W. To enhance the performance of EPS, a chemical coagulant was used in combination with EPS. During the LW/W 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 LW/W. 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 LW/W 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 1 M H₂SO₄ or base 2 M 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 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 and 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.

Results and discussion

Characteristics of CLWW

The LW/W samples collected from the discharge basin (raw LW/W) were characterized for turbidity, pH, conductivity, SS, TS, COD, NPEOs, TN, TP & metals. The characterization results are depicted in Table 2. It was

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

found that LW/W 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.

Treatment of CLWW using chemical coagulants

Identification of optimized pH for chemical coagulants

The chemical coagulants (alum, FeSO₄, and CaCl₂) 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 (Additional file 1: Figure S1). 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 were 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 FeSO₄ was used as a coagulant the zeta potential value varied from pH 3 to

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

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 were found at pH 3, 5, 7, 9, and 11 respectively. The maximum FA of 29% was observed at pH 5. Figures 2a, 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

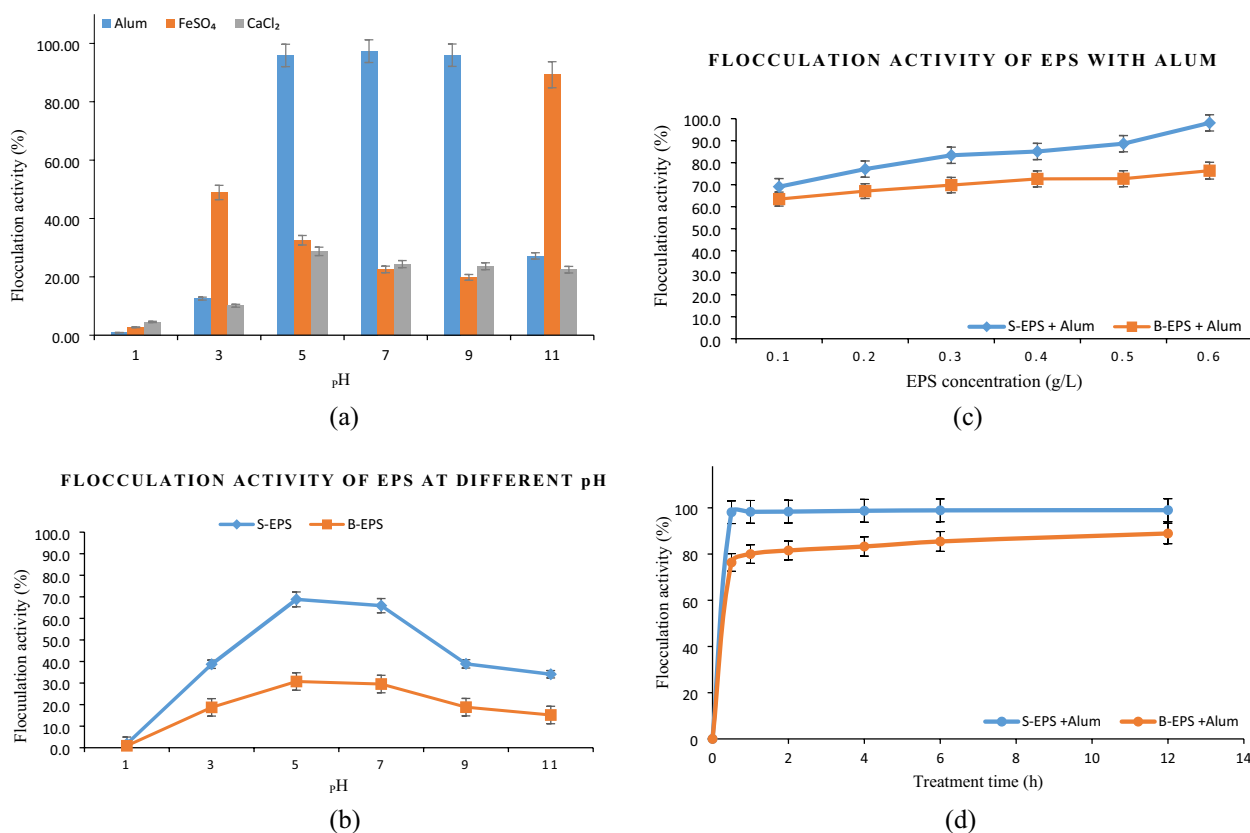


Fig. 2 Flocculation activity of: **a.** Chemical coagulants at different. **b** S-EPS and B-EPS at different pH. **c** EPS in the combination of alum at different EPS concentrations at pH 7. **d.** S-EPS and B-EPS at different treatment time

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.

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 LW/W). 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 FeSO₄ 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 FeSO₄ dosage. When the concentration of CaCl₂ 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 FeSO₄ provided higher turbidity, SS, and TS removal as compared to CaCl₂. 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 FeSO₄. However, for CaCl₂, the maximum removal efficiency was achieved at

Table 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
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

1500 mg/L dose. The removal of turbidity, SS, and TS for CaCl₂ (1500 mg/L) was found to be 36.18%, 29.17%, and 36.09% respectively.

COD removal of 85.10, 42.60 and 47.80% COD was achieved using alum (2000 mg/L), CaCl₂ (1500 mg/L), and FeSO₄ (2000 mg/L), respectively. It was seen that when the dosage of alum and FeSO₄ 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 FeSO₄, respectively. However, CaCl₂ gave a slightly different performance. When the dosage of CaCl₂ was increased from 100 to 1500 mg/L, COD removal was enhanced. But when the dosage of CaCl₂ increased further from 1500 to 2000 mg/L, COD removal decreased. The maximum COD removal (42.60%) was obtained at a CaCl₂ concentration of 1500 mg/L. The reason behind the lower COD removal in the case of CaCl₂ 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 were 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 were 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 2a, 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 and 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 Fig. 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.

Table 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 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-floculant, 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%.

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

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

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%).

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, 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 Fig. 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 12 h of treatment. It was observed that treatment time (after 0.5 h) did not have a significant impact on the turbidity removal rate for S-EPS (Fig. 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 12 h 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 (Fig. 3d, 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. 3d, 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 (Fig. 3d–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).

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 LWV. However, EPS produced by different bacterial

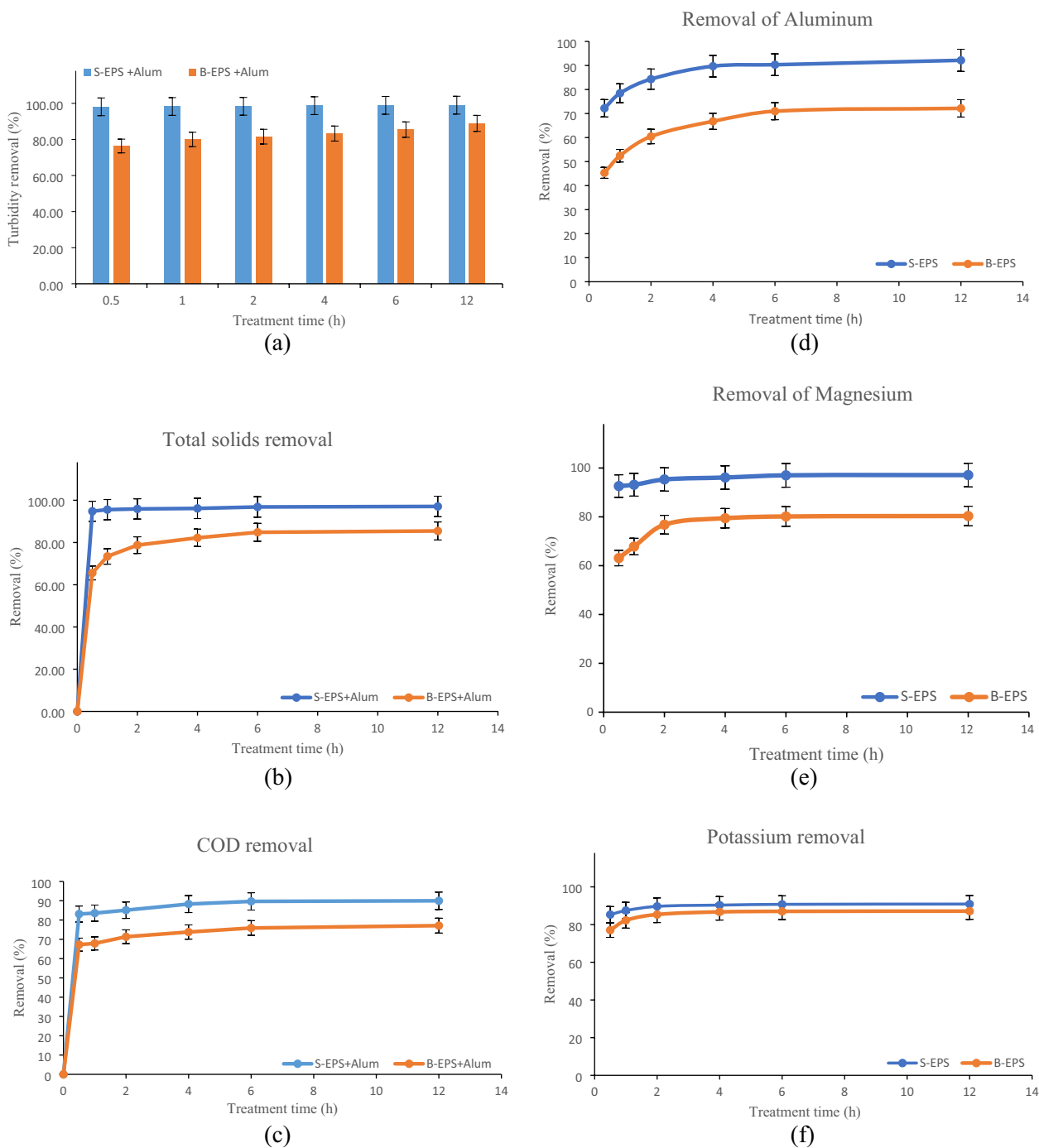


Fig. 3 Removal of pollutants using B-EPS and S-EPS: **a** Turbidity removal. **b** TS removal. **c** COD removal. **d** Aluminum removal. **e** Magnesium removal. **f** Potassium removal

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 (0.6 g/L) when combined with alum (0.3 g/L) at pH 7 gave the best results for LWW treatment after 30 min. 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 4 h.

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 (0.5 g/L) and FeSO₄ (2 g/L) at pH 8 for a treatment time of 8 h.

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 (0.015 g/L) and FeSO₄ (2 g/L) were used together at pH 8 for 2 h. 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.

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, FeSO₄ and CaCl₂), 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 4 h, it resulted in more than 88.00% COD removal) from CLWW.

Supplementary Information

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Additional file 1: Figure S1. Effect of pH on Zeta Potential.

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Author contributions

SK, AKM, PD, and RDT conceived and planned the idea of the study. SK experimented and collected the data. All authors carried out the analysis and contributed to the interpretation of the results. SK wrote the manuscript in consultation with AKM, PD, and RDT. 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 Environmental System Research.

Competing interests

The authors declare that they have no competing interests to declare.

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