Application of Extracellular Polymeric Substances Extracted from Wastewater Sludge for Reactive Dye Removal

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Abstract
This study aimed to investigate the adsorption of three commercial reactive dyes using extracellular polymeric substances (EPS) extracted from the waste sludge of a beer wastewater treatment plant in Hanoi, Vietnam. EPS was extracted from sludge by the HCHO-NaOH method and was characterized by measuring kaolin flocculation activity, dry weight, chemical composition, and functional groups. Adsorption of dyes on EPS was conducted by Jartest at different pH values, contact times, and EPS dosages. The EPS was composed of 25% sludge by weight. The FTIR analysis showed the presence of amine and carboxyl groups in the EPS structure. The removal efficiencies of reactive dyes were high at pH values below 6, a contact time of 30 to 60 min, and EPS dosage of 200 – 250 mg/L. At optimum condition, removal efficiencies of 85%, 99%, and 99% were obtained for Reactive Yellow 176 (RY 176), Reactive Blue 21 (RB 21), and Reactive Red 241 (RR 241), respectively. The adsorption process could be described by both Langmuir and Freundlich models. The maximum dye adsorption capacities for RY 176, RB 21, and RR 241 were 0.50 g/g, 0.72 g/g, and 0.95 g/g, respectively. It is concluded that EPS in wastewater sludge could be utilized as an effective adsorbent for dye removal, thereby enhancing the value of sludge in wastewater treatment.

Highlights
• EPS of wastewater sludge was extracted by HCHO-NaOH and used for dye removal.
• EPS extracted by HCHO-NaOH was composed of 25% of sludge weight.
• Removal of RY 176, RB 21, and RR 241 were 85%, 99%, and 99%.
• The high adsorption capacity of dye was obtained at a pH of less than 6.
Keywords Extracellular polymeric substances · Recovery · Wastewater sludge · Reactive dye adsorbent

1 Introduction

Textile dyeing industry generates a large volume of wastewater, which is difficult to manage. Dye is the primary pollutant of textile wastewater with complex structure, high molecular weight, and low biodegradability (Banerjee et al. 2015; Hua et al. 2021; Ringwal et al. 2021). As low biodegradable substances, dyes could be accumulated in the food chain; moreover, dyes can also have several harmful effects such as being carcinogenic and hindering the penetration of light in the water environment which negatively affect aquatic systems (Haque et al. 2021; Li et al. 2017; Saba et al. 2021).

Technologies such as coagulation, biological treatment, adsorption on activated carbon, and advanced oxidation processes (AOPs) have been used to treat textile wastewater (Akansha et al. 2019; Bulca et al. 2021). Coagulation techniques are cost-effective but are limited to highly soluble dyes such as azo and reactive dyes (Tianzhi et al. 2021). Biological methods require long hydraulic retention time and specific bacterial strains to degrade dyes; therefore, their industrial-scale applicability could be limited (Ogunlaja et al. 2020). AOPs (such as Fenton, TiO2 catalyst) can remove dyes completely, but they are costly and difficult to operate (Archina et al. 2016; Akansha et al. 2019).

Various studies have been conducted on low-cost adsorbents for dye removal, such as pear seeds, poplar sawdust, rice bran, walnut shell powder, mango leaf powder (Igwegbe et al. 2020; Tezcan and Ates 2019; Hong and Wang 2017; Uddin and Nasar 2020; Uddin et al. 2017). Recently, microbial EPS have been considered as environmentally friendly and low-cost adsorbents (Haque et al. 2021). The applications of EPS in dye removal have been investigated by several studies (He et al. 2021; Harshitha et al. 2021; Haque et al. 2021). It has been shown that EPS synthesized by bacterial strains have maximum dye adsorption capacity (145 – 178.6 mg/g), and this capacity could be comparable to that of commercial activated carbon (He et al. 2021; Harshitha et al. 2021).

EPS plays an essential role in protecting microbial cells from harsh environmental conditions and floc forming. It has been reported that the average particle sizes of flocs formed by crude EPS were bigger than those formed by chemical flocculants (Zhang et al. 2021). EPS mainly comprises polysaccharides, proteins, and other macromolecules such as DNA, lipids, and humic substances (Decho and Gutierrez 2017). Furthermore, it has been found that the biopolymer content in waste sludge varies from 10% to 30% depending on the extraction method used (Hoang et al. 2016). Although EPS content in wastewater sludge is substantially high, little information has been reported on the ability of using EPS extracted from wastewater sludge for commercial dye removal. Most of the prior studies investigated the ability of EPS extracted from bacterial strains like Bacillus stratosphericus, Bacillus cereus, Bacillus subtilis, Pseudomonas sp., Klebsiella sp. in dye degradation (Akansha et al. 2019; Hua et al. 2021; Siddharth et al. 2021).

In order to reuse the valuable component of wastewater sludge, this study aimed to extract EPS directly from wastewater sludge and assess the ability of the extracted EPS in the treatment of commercial reactive dyes. Factors affecting dye removal (pH, reaction time, and biopolymer dosage) and adsorption isotherm were also investigated. The use of EPS extracted from wastewater sludge for dye removal would be significant in terms of
environmental remediation, because it could contribute to the reduction of solid waste from wastewater treatment plants.

2 Materials and Methods

2.1 Materials

**Sewage Sludge** The sludge was taken from the aerobic tank of the wastewater treatment plant of the Hanoi beer factory (Me Linh district, Hanoi, Vietnam). The sludge was concentrated to 20 g MLSS/L and stored at -4°C for further experiments.

**Dyes** The dyes used in the study were reactive dyes, namely Reactive Yellow 176 (RY 176), Reactive Blue 21 (RB 21), and Reactive Red 241 (RR 241). The dyes were obtained from the Dong Xuan textile and garment factory in Hanoi, Vietnam. The structures of dyes are presented in Fig. 1. The dyes were prepared at concentrations of 80 mg/L. The maximum absorption wavelengths of RY 176, RB 21, and RR 241 are 418 nm, 626 nm, and 541 nm, respectively.

2.2 Methods

2.2.1 Separation of EPS

**Chemical Method** The HCHO-NaOH method was used to separate EPS from sludge. Prior to EPS extraction, the sludge was washed twice with distilled water and concentrated to a concentration of 20 g/L. 0.42ml of formaldehyde (HCHO) was added to 70 ml of sludge and incubated at 4°C for 1 hour. Then, 3 ml NaOH 10M was added to the sludge mixture and kept at 4°C for 3h. The reaction mixture was then centrifuged at 4°C for 15 min, 4000 x g to extract EPS from biomass (D’Abzac et al. 2010).

**Centrifuge Method** The process of separating EPS by centrifuge method is similar to the process of separating EPS by HCHO-NaOH but omitting the additional step of HCHO and NaOH.

2.2.2 Factors Affecting Dye Removal Efficiency by EPS

The effects of pH, reaction time, and EPS dosage on dye removal efficiency were performed separately on Jartest equipment (SJ-10, Yhana). The effect of pH was examined in the pH range from 2 to 10 by adding NaOH (0.1 M) or H₂SO₄ (0.1 M). The reaction time was in the range of 5 - 360 min to determine adsorption equilibrium. The effect of EPS dosage was examined with EPS volumes from 0.5 to 10 mL.

The test procedure of Jartest is described as follows: 500 mL of the dye solution (80 mg/L) was stirred at 120 rpm. 50 mg/L of Al³⁺ and crude EPS were added, and pH was adjusted. Then, the stirring speed was reduced to 60 rpm for dye treatment. Samples were settled for 30 min, the supernatant was filtered through a filter paper of 0.45 μm,
and the remaining dye concentration was measured. The dye removal efficiency was calculated according to the following Eq. (1):

\[
H = \frac{C_0 - C_1}{C_0} \times 100
\] (1)

where \( H \) is the efficiency (%); \( C_0 \) is the dye concentration of the sample without addition of EPS (mg/L); and \( C_1 \) is the dye concentration of the sample with addition of EPS (mg/L).
2.2.3 Adsorption Isotherms

The adsorption equilibrium of dyes by EPS can be described by Langmuir and Freundlich models. The linear form of the Langmuir isothermal equation is presented below:

\[
\frac{1}{q} = \frac{1}{q_e} + \frac{1}{q_e K_L} C_e
\]  

(2)

The linear form of Freundlich model is presented as follows:

\[
\log(q) = \log(K_f) + \frac{1}{n} \log(C_e)
\]  

(3)

In particular, \( q \) and \( q_e \) are the adsorption capacity and maximum adsorption capacity of dye on EPS, respectively (mg/g EPS); \( C_e \) is the residual dye concentration in the liquid phase (mg/L); \( K_L \) is the Langmuir constant (L/mg) related to the free energy of adsorption; \( K_f ((L/mg)^{1/n}) \) and \( n \) are the Freundlich constants.

From linear Eqs. (2) and (3), \( q_e \) and the coefficients \( K_L, K_f, n \) can be determined.

2.2.4 Analytical Methods

Purification of EPS from Sludge After the HCHO-NaOH extraction, the sludge mixture was centrifuged at 4000×g at 4 °C (Rotina 420R, Hettich Zentrifugen) for 15 min and the EPS containing the liquid phase was collected (the solid phase was removed). The EPS containing the liquid phase was purified by precipitation in cold ethanol with a volume ratio of ethanol and sample as 2:1 and stored at -20 °C overnight. The sample after precipitation was centrifuged at 4000×g at 4 °C for 15 min in order to obtain purified EPS. The EPS after purification was used to analyze dry weight, protein, polysaccharide, nucleic acid, and chemical functional group composition.

Determination of EPS Concentration The EPS concentrations were determined by weight method after drying the samples to constant weights at 105 °C.

Determination of Protein, Polysaccharide, and Nucleic Acid Concentrations The EPS collected after purification (by precipitation in cold ethanol) was dissolved in distilled water to the original volume. This solution was used to analyze proteins, polysaccharides, and nucleic acids. Protein was examined by the Lowry method with a standard solution of BSA (Bovine Serum Albumin) (Lowry et al. 1951). Carbohydrate was determined using the phenol - sulfuric acid method with glucose as the standard solution (DuBois et al. 1956). Nucleic acids were analyzed using the diphenylamine method (Dell’Anno et al. 1998).

Determination of Functional Groups in EPS EPS functional groups were determined using Infrared Spectroscopy (IR) spectrum. 0.1 - 0.2 mg of the dried purified EPS was mixed and milled with 100 mg of KBr. Then, the sample was pressed with a force of 5 tons to create pellets. The sample was placed on the infrared spectrum window to determine the compositions of functional groups.
Measure Kaolin Flocculation Activity (FA) The procedure for measuring kaolin flocculation activity of EPS was adopted from Bezawada et al. (2013). FA was measured at EPS dosage of 10 mg/L for both crude and purified EPS.

Measurement of Turbidity The supernatant after Jartest was used to measure the turbidity of the solution. The Hana turbidity meter (Model: HI 93703C) with a standard solution of Fomazin was used for the measurement.

Determination of Dye Concentrations Concentrations of RY 176, RB 21, and RR 241 dyes were measured by photometric method (UVD-3200, Labomed Inc.) at wavelengths 418 nm, 626nm, and 541 nm, respectively.

3 Result and Discussion

3.1 Properties of EPS Extracted by HCHO-NaOH Method

3.1.1 Chemical Composition of Extracted EPS

Concentrations and compositions of EPS extracted by the HCHO-NaOH method (compared with the centrifuge method) are shown in Table 1. EPS is an essential component contributing to forming activated sludge flocs in wastewater treatment systems (Pan et al. 2010). Concentration of the extracted EPS depends on the employed extraction methods. According to the results obtained in Table 1, concentration of EPS extracted by the HCHO-NaOH method was higher than that by the centrifuge method by a factor of 215.4 (5816 compared to 27 mg/L).

Physical methods such as centrifugation and heat have been applied to determine EPS concentrations in biomass in various studies (Laspidou and Rittmann 2002). However, only EPS extracted by those methods are mostly soluble forms or weakly bound to microbial cells (Laspidou and Rittmann 2002). In the aeration tank, microorganisms are significantly impacted by the turbulence of the air; consequently, EPS, which is weakly bound to microbial cells is likely to be released in the water, and only EPS strongly bound remain. Therefore, the concentration of EPS extracted from wastewater sludge by centrifugation was not high. In contrast, both loosely bound EPS and capsular EPS can be effectively extracted by chemical methods due to the reaction of chemical agents with the bonding between EPS and microbial cells. The concentration of EPS extracted by the HCHO-NaOH method was high, probably be due to the fact that chemicals as NaOH are not only able to separate high molecular weight polymer molecules outside the cell but also can break down the cell wall.

| Extraction Method | EPS (mg. L⁻¹) | Protein (mg.L⁻¹) | Polysaccharide (mg.L⁻¹) | Nucleic acid (μg.L⁻¹) | FA of EPS (%) |
|-------------------|---------------|------------------|------------------------|----------------------|---------------|
| Centrifugation    | 27±1.2        | 8.9±0.3          | 11.4±0.4               | 9.3±0.08             | 37±1.6        |
| HCHO-NaOH         | 5816±256      | 1762±63.7        | 439.8±15.9             | 420.3±8.2            | 96±7.2        |

Table 1 Concentrations and composition of EPS

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and separate large molecular organic compounds within microbial cells such as DNA and intracellular polysaccharides.

Nucleic acid is an indicator for cell lysis during extraction (Yang et al. 2019). The high concentration of nucleic acid obtained in EPS extracted by HCHO-NaOH indicates that a large amount of EPS is from high molecular polymer in the interior of the microbial cells. High nucleic acid content in EPS confirms again that the HCHO-NaOH method not only extracts EPS but also breaks the bacterial cell and releases high molecular substances to the liquid phase.

3.1.2 Determination of Functional Groups of EPS

Functional groups in EPS extracted from the biosludge were analyzed by the IR method. IR spectrums are shown in Fig. 2. The IR spectrum of the EPS obtained showed that the peaks occurred between 400 and 4000 cm\(^{-1}\). A peak near 3500 cm\(^{-1}\) was related to hydroxyl groups of polysaccharides (Liu et al. 2019). The presence of NH\(_2\) group was confirmed at the peak of 1643 - 1648 cm\(^{-1}\), whereas the presence of carboxyl (C=O)
and O-C (carboxylic acid group and their derivatives) was demonstrated at the peak of 1395 cm$^{-1}$ (Cavallero et al. 2020; He et al. 2021). Functional groups such as NH$_2$, C=O and O-C in EPS have been considered to be capable of binding and adsorbing organic substances and play an important role in the flocculation ability of EPS. It has been reported that the C=O and NH$_2$ groups of EPS are involved in the adsorption of dyes (He et al. 2021).

### 3.1.3 Flocculation Activity of EPS

Flocculation with kaolin solution is one parameter to measure EPS activity as it is a polymeric substance. Table 1 showed FA of crude and purified EPS extracted by centrifugation and HCHO-NaOH method. The results showed that the FA of EPS extracted by HCHO-NaOH was much higher than centrifugation. The FA of purified and crude EPS extracted by the HCHO-NaOH method was 96 and 90%, respectively, whereas the FA of purified and crude EPS extracted by centrifugation was only 37% and 73%, respectively. The high flocculation activity of EPS indicated that using HCHO and NaOH in extraction process did not affect much on EPS quality.

As shown in Table 2, kaolin flocculation activity of EPS extracted from wastewater sludge (96%) was comparable to that of EPS synthesized in synthetic medium (87.9%) (Yu et al. 2020b); nitrogen-free glucose medium with nitrile (90%) (Yu et al. 2020a); high saline medium (97.69%) (Hua et al. 2021); and crude glycerol medium (93.71%) (Yadav et al. 2021). These results confirm that HCHO-NaOH is a potential method to separate a high amount of EPS from sludge (25% by weight), which is capable of treating wastewater. The ability of EPS in dye removal is presented in the following sections.

### 3.2 Effect of Factors on Dye Treatment Performance of EPS

The capacity of crude EPS extracted by the HCHO-NaOH method in treating reactive dyes (RY 176, RB 21, and RR 241) was studied. The three reactive dyes used in the study are highly soluble and difficult to be removed by conventional coagulants such as PAC and alum.

| BFs producing bacterium | FA (%) | EPS (mg/L) | Growth medium | References |
|------------------------|--------|------------|---------------|------------|
| Bacillus sp.            | 97.69  | 5.2        | High saline medium | Hua et al. 2021 |
| Lipomyces starkeyi U9   | 87.9   | 9.2        | Synthetic     | Yu et al. 2020b |
| K. oxytoca GS-4-08      | 90     | 4          | Nitrogen-free glucose medium with nitrile | Yu et al. 2020a |
| Bacillus sp.8           | 93.71  | 50         | Crude glycerol | Yadav et al. 2021 |
| EPS of wastewater sludge extracted by HCHO-NaOH | 96 | 10 | Wastewater sludge of beer factory | This study |


3.2.1 Effects of pH

pH is the main parameter controlling the adsorption process due to its impact on the binding surface of the adsorbent and the ionization of dye molecules (Bhattacharyya and Sharma 2004). Effect of pH on dye treatment efficiencies (RY 176, RB 21, and RR 241) by EPS extracted by the HCHO-NaOH method are shown in Fig. 3. The results showed that adsorption efficiencies were highly dependent on pH. At neutral to alkaline pH (7, 8, 10), dye removal efficiencies were low for RY 176 and RR 241 (in the range of 2.7 - 7.2% and 0.4 - 2.9%, respectively). In contrast, at pH = 7 and 8, high removal efficiencies were recorded for RB 21 (65.7 and 39.8%). The highest removal efficiencies of all dyes were achieved at pH = 2 (87.9% for RY 176, 99.1% for RB 21, and 70.8% for RR 241).

pH is one of the environmental determinants of the dissociation of functional groups in the structure of dyes and ions chemistry of the active centers on the adsorbent surface. Therefore, the optimal pH depends on both the nature of the adsorbent and the chemical structure of the dye. High removal of reactive dye in acidic condition was reported in various studies (Igwegbe et al. 2020; Tezcan and Ates 2019; Hong and Wang 2017). On the other hand, the optimum pH for methylene blue removal was neutral (He et al. 2021; Uddin and Nasar 2020; Uddin et al. 2017; Tan et al. 2008).

EPS were separated from aerobic sludge with main components polysaccharides and protein with carboxyl, hydroxyl, and amino functional groups. The functional group of EPS plays a significant role in the adsorption process. At pH of approximately 7, amino functional groups are chemically positive (protonate), and carboxyl functional groups are negative (deprotonate). At lower pH, both these functional groups are positively charged (Benjamin and Lawler 2013). On the other hand, all three dyes contain negatively charged sulfonate groups (Fig. 1). Therefore, in an acidic environment, the surface of the positive adsorbent enhances the adsorption of negatively charged dyes. This may be the reason dye adsorption efficiency increases rapidly in acidic condition. This result also shows that the amino functional group does not play an essential role in the adsorption of RY 176 and RR 241 because, at pH> 7, the treatment effect is almost negligible.

Fig. 3 Effects of pH on dye treatment efficiencies by EPS extracted by HCHO-NaOH method
3.2.2 Effects of Contact Time

The effect of contact time on dye removal efficiencies is shown in Fig. 4. The results showed that in the first 5 min, removal efficiencies of all dyes rapidly increased, followed by gradual increases up to the end of the experiment. The removal efficiency of RB 21 dye reached 85%, whereas 65% and 36% were the removal efficiencies of RY 176 and RR 241 dyes, respectively. The rapid increase in the first 5 min could be due to the high concentration gradient of dyes between liquid and solid phases, leading to a high adsorption rate. After 60 min, insignificant increases in efficiency could be due to the remaining low concentrations of the dyes. The dyes were likely to be adsorbed by the extracted EPS rather than degraded, as synthetic dyes are generally difficult to degrade completely by microorganisms in a short time (less than 60 min) (He et al. 2021; Ogunlaja et al. 2020).

3.2.3 Effects of EPS Concentration

Correlations between EPS concentrations (in the range of 29 – 582 mg/L) and removal efficiencies of RY 176, RB 21, and RR 241 are shown in Fig. 5. Adsorption of dyes increased sharply when EPS concentration increased from 0 to 100 mg/L. Adsorption achieved highest at EPS concentration of 200 – 300 mg/L. Further addition of EPS led to only 1 – 2% increase in adsorption efficiency. The optimal dosage of EPS for RY 176, RB 21, and RR 241 were 200, 290, 350 mg/L, respectively. The removal efficiency of RY 176, RB21, and RR 241 were 86%, 99%, and 90%. The increase in EPS concentration means more adsorption sites, resulting in fast enhancement of dye adsorption. Further increase in EPS concentration showed a slight change in removal efficiency due to the fixed amount of adsorbate molecules.

The optimum dosage obtained in this study (200 – 350 mg/L) was lower than in other studies. As summarized in Table 3, the optimal dosage varied from 400 – 600 mg/L for EPS (Harshitha et al. 2021; He et al. 2021) and around 1000 – 3000 mg/L for other low-cost adsorbents (Tan et al. 2008; Uddin et al. 2017; Uddin and Nasar 2020; Hong and Wang 2017; Tezcan and Ates 2019; Igwegbe et al. 2020).

Fig. 4 Effects of contact time on dye treatment efficiencies by EPS extracted by the HCHO-NaOH method
Energy-dispersive X-ray spectrum (EDS) analysis of EPS before and after adsorption showed an increase of sulfur from 0.3% to 1.09 – 1.39% (Table 3). As shown in Fig. 1, all three dyes contain the sulfonate group in their chemical structure. The increase of sulfur after adsorption revealed the adsorption of dyes on the surface of EPS.

### 3.3 Adsorption Isotherms of EPS

The Langmuir and Freundlich adsorption models were used to describe the adsorption of dyes on EPS. Fig. 6 and Table 4 demonstrate the curve of the Langmuir and the Freundlich isotherm. The correlation coefficient ($R^2$) was 0.935 to 0.996 for the Langmuir model and 0.920 to 0.992 for the Freundlich model. The dye adsorption process on EPS is consistent with the Langmuir and Freundlich models. This indicates that the dyes were monolayer adsorbed on EPS in the condition of heterogeneous material surface.

The comparison of dye adsorption capacity of extracted EPS with other adsorbents is summarized in Table 5. The maximum adsorption capacity ($q_e$) of EPS for RB 21, RY 176, and RR 241 were 720, 520, and 952 mg/g, which were higher than the adsorption capacity of EPS of *Klebsiella oxytoca* (145 mg/g) (He et al. 2021) and *Lysinibacillus*...
sp. SS1 (178.6 mg/g) (Harshitha et al. 2021). The obtained adsorption capacity outperforms most reported activated carbon from low-cost material such as coconut husk (435 mg/g) (Tan et al. 2008), rice bran (151.3 mg/g) (Hong and Wang 2017), Dacryodes edulis Seeds (106.5 – 115.98 mg/g) (Igwegbe et al. 2020), mango leaf powder (156 mg/g) (Uddin et al. 2017) and other materials (Table 5).

It is well known that EPS compose protein and polysaccharides and has several functional groups for the adsorption of dyes and other organic compounds. However,

**Table 4** Parameters of Langmuir and Freundlich equation for dye adsorption

| Model      | Parameters       | RY 176 | RB 21  | RR 241 |
|------------|------------------|--------|--------|--------|
| Langmuir   | $q_e$ (mg/g EPS) | 502    | 720    | 952    |
|            | $K_L$ (L/mg)     | 0.04   | 1.07   | 0.01   |
|            | $R^2$            | 0.9624 | 0.9353 | 0.9960 |
| Freundlich | $K_f$ (L/mg)$^{1/n}$ | 3.91   | 12.17  | 1.58   |
|            | $n$              | 1.05   | 2.09   | 0.67   |
|            | $R^2$            | 0.9524 | 0.9203 | 0.9924 |
Table 5  The adsorption capacity of extracted EPS in comparison with other studies

| Adsorbent                          | Dyes            | \( q \) (mg/g) | pH   | Adsorbent dosage (mg/L) | Dye concentration (mg/L) | References                        |
|-----------------------------------|-----------------|----------------|------|-------------------------|--------------------------|-----------------------------------|
| *Klebsiella oxytoca*              | Methylene Blue  | 145            | 6-8  | 600                     | 200                      | He et al. 2021                     |
| *Lysinibacillus sp. SS1*          | Malachite Green | 178.6          | 6    | 400                     | 100                      | Harshitha et al. 2021             |
| *Dactyloides edulis* Seeds activated carbon | CR              | 106.5          | 2    | 1000 - 2000             | 100                      | Igwegbe et al. 2020               |
|                                   | VY4             | 115.98         |      |                         |                          |                                   |
| Pyrolysis poplar sawdust          | Disperse Orange 30 | 0.09          | 2    | 3000                    | 50                       | Tezcan and Ates 2019              |
| Modified rice bran                | Reactive blue 4 | 151.3          | 2    | -                       | 500                      | Hong and Wang 2017               |
| Walnut shell powder               | Methylene Blue  | 36.6           | 8    | 2000                    | 50 - 200                 | Uddin and Nasar 2020             |
| Mango leaf powder                 | Methylene Blue  | 156            | 7-10 | 3000                    | 350                      | Uddin et al. 2017                |
| Coconut husk activated carbon     | Methylene blue  | 435            | 7    | 1000                    | 50 - 500                 | Tan et al. 2008                  |
| EPS extracted from wastewater sludge | RB 21           | 720            | 4    | 290                     | 80                       | This study                        |
|                                   | RY 176          | 502            | 4    | 200                     | 80                       |                                   |
|                                   | RR 241          | 952            | 4    | 350                     | 80                       |                                   |
in biomass, the EPS is tightly attached with microbial cells, and most of the functional groups are occupied by linking with the cell membrane, therefore, they exhibit low adsorption capacity. By adding NaOH and HCHO, EPS can be detached from the cell, free the functional groups and thereafter exhibit high adsorption capacity. Besides adsorption, EPS could contribute to the coagulation process, enhancing dye removal. The high adsorption of RR 241 on EPS (952 mg/g) might be due to the contribution of the coagulation process.

The obtained results of this study showed that EPS could be extracted simply by adding HCHO and NaOH to the sludge solution. Extracted EPS was effective in crude form, which contains EPS, extracting chemical and bacterial cells. Other adsorbents such as EPS synthesized by specific strain, activated carbon required a more complex process to produce. Synthesized EPS required strict conditions and long fermentation time (Harshtitha et al. 2021; He et al. 2021); activated carbon required pyrolysis at high temperature or chemically modified material surface (Hong and Wang 2017; Tezcan and Ates 2019; Igwegbe et al. 2020).

The significant finding of this study was that EPS containing wastewater sludge could be used as an effective adsorbent for commercial reactive dyes. Simple extraction of EPS from wastewater sludge and high dye removal ability enhance its feasibility in the field application.

4 Conclusions

The adsorption of reactive dyes on EPS of wastewater sludge was successfully investigated. EPS extracted from wastewater sludge using the HCHO-NaOH method could be utilized as an effective adsorbent. Complete removal (85% – 99%) was achieved in acidic condition with EPS dosage from 200 – 350 mg/L. Langmuir and Freundlich isothermal equations could describe the adsorption of the reactive dyes Reactive Yellow 176 (RY 176), Reactive Blue 21 (RB 21), and Reactive Red 241 (RR 241) on EPS. The maximum dye adsorption capacities for RB 21, RY 176, and RR 241 were 0.72 g/g, 0.5 g/g, and 0.95 g/g, respectively. The results indicate that EPS of wastewater sludge could be reused as an alternative adsorbent for reactive dye removal.

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Availability of Data and Material The data in this study are available from the corresponding author on reasonable request.

Code Availability Not applicable

Authors’ Contributions All authors contributed to the conception and design. DTN and HVN: conceptualization. DTN and HXD: formal analysis and investigation. DTN, TTTN, and PMN: writing the original draft. HVN, QVT, and RDT: reviewing and editing. All authors read and approved the final manuscript.

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Declarations

Conflicts of Interest/Competing Interests  The authors declare that they have no competing interests

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