Removal of an anionic azo dye direct black 19 from water using white mustard seed (Semen sinapis) protein as a natural coagulant

Jingxi Tie, Zhihong Zheng, Guoting Li, NanNan Geng, Ganfang Chang, Lu Yu, Yifan Ji, Ling Shi, Yangyang Chang and Leilei Hu

ABSTRACT

In this study, standard jar tests were conducted using white mustard seed protein (WMSP) as a natural coagulant to remove direct black 19 (DB-19) from its aqueous solution. Comparative coagulation tests were performed using commercial polyaluminum chloride (PAC). The results showed that DB-19 removal by WMSP increased with increasing settling time and reached the maximum removal at 180 min. The DB-19 removal descended from 98.4 to 46.2% as pH increased from 4 to 10. The most effective temperature for DB-19 removal was 25°C. The removal of DB-19 was weakened by the presence of Na2S2O4. Overall, WMSP was more efficient than PAC for DB-19 removal in all experiments except at pH 4 and 5. The mechanism of the removal of DB-19 by WMSP could be attributed to adsorption and charge neutralization processes.

Jingxi Tie (corresponding author)
Guoting Li
NanNan Geng
Ganfang Chang
Lu Yu
Yifan Ji
Ling Shi
Yangyang Chang
Leilei Hu

School of Municipal and Environmental Engineering, North China University of Water Resources and Electric Power, Zhengzhou, 450046, China
E-mail: zzh_ncwu@163.com

Jingxi Tie
Henan Engineering Research Center of Water Pollution and Soil Damage Remediation, Zhengzhou, 450011, China
and
Henan Key Laboratory of Water Environment Simulation and Treatment, Zhengzhou, 450002, China

Zhihong Zheng
Henan Vocational College of Water Conservancy and Environment, Zhengzhou, 450008, China
and
Henan Engineering Laboratory of Lake & Reservoir Ecological Protection and Reclamation, Zhengzhou, 450008, China

INTRODUCTION

Globally, the textile industry is one of the largest fresh water consumers which consequently generates large amounts of wastewater. According to the data of the China statistical yearbook on the environment, the textile industry discharged 1.84 × 10^9 tons of wastewater in 2015, ranking as the third largest pollutant source among the 41 types of industries under investigation. The dyeing wastewater contains a large variety of contaminants including salts, dyes, heavy metals, surfactants, soaps, enzymes, oxidizing, reducing agents, and so on (Verma et al. 2012b; Bilińska et al. 2017; Costa et al. 2018). Among the pollutants, the residual dyes are the most peremptory sources of contamination.
since they are aesthetically undesirable, and can damage the aquatic ecosystem significantly by blocking light penetration and preventing oxygen transfer (Shi et al. 2007; Verma et al. 2012b; Lafi et al. 2018). Furthermore, most of the dyes used in the textile industry are synthetic and belong to azo derivatives, which make them and their metabolite products mutagenic and carcinogenic (Weisburger 2002; Bashiri et al. 2018). As a result, it is of great importance to remove the dyes from wastewater.

So far, many techniques have been developed to treat dyeing wastewater. For example, biological processes, coagulation/flocculation, adsorption and advanced oxidation processes including ozonation and photocatalytic reaction, etc. (Tie et al. 2016; Bilińska et al. 2017; Wang et al. 2017, 2018; Mahmoodi et al. 2018; Yan et al. 2018). Among the techniques, coagulation/flocculation has many advantages such as cost effectiveness, convenience in operation, low energy consumption, and no generation of harmful and toxic intermediates (Shi et al. 2007; Verma et al. 2012a).

Currently, the commercial coagulants used for water treatment consist of two major groups, namely, inorganic coagulants and organic polymers. Though both of them are playing a very important role in water treatment, many drawbacks arise in water treatment, such as inefficiency at low temperature, changes of pH, corrosion of instruments, and generation of a huge amount of non-biodegradable sludge and so on. Furthermore, their residuals in water can cause adverse impacts on living beings. For example, studies revealed that aluminum is neurotoxic and can cause pathogenesis of Alzheimer’s disease (Flaten 2001; Polizzi et al. 2002). Also, the residual monomers of polyacrylamide (PAM) present in treated water have been proven to be neurotoxic and carcinogenic (Šciban et al. 2009). Hence, the development of eco-friendly coagulant has been a major topic of research in water treatment.

Plant-based coagulants, including mustard seed, Jatropha curcas seed, guar gum, copra, Cactus latifaria, Prosopis juliflora seed, common bean, chitosan, orange waste and so on, have been used to remove a large variety of pollutants including turbidity, algae, dyes, humic acid, bacteria and metals etc. from water due to their advantages such as abundant source, high efficiency, low sludge production, biodegradability, and nontoxicity (Pritcharda et al. 2010; Antov et al. 2012; Abidin et al. 2013; Banerjee et al. 2013; Fatombi et al. 2013; Bodlund et al. 2014; Tie et al. 2015; Chethana et al. 2016; Ferhat et al. 2016; Tie et al. 2017; Kebaili et al. 2018).

For instance, Ndabigengesere et al. (1995) reported that the active agents in aqueous Moringa extracts were cationic proteins with a molecular weight of 15 kDa and an isoelectric point between 10 and 11. The optimal dosage of shelled Moringa oleifera seed was almost the same as alum, whereas purified proteins were more effective than alum for turbid water treatment. Furthermore, the innocuous coagulant did not affect the pH and conductivity of treated water with four or five times less volume of chemical sludge created compared to alum.

Recently, Bouladjoul et al. (2018) reported that Moringa oleifera seed powder was used to enhance the primary treatment of paper mill effluent. The results indicated that the turbidity and COD abatements were 96.02 and 97.28% using Moringa oleifera seed powder as coagulant, whereas the respective removals of turbidity and COD by alum were 97.1 and 92.67%, indicating that Moringa oleifera is a very efficient natural coagulant for the treatment of paper mill effluent.

Betatache et al. (2014) reported that the optimum dosage of prickly pear cactus was 0.4 g/kg for sewage sludge conditioning, which was much more efficient than three polyelectrolytes, namely, 0.8 g/kg for Chimfloc C4346, 80 g/kg for FeCl3, and 60 g/kg for Al2(SO4)3.

In recent research, mustard seed proteins with the molecular weight of approximately 6.5 and 9 kDa have been proven to be more efficient to remove turbidity from pond water than Moringa oleifera seed proteins (Bodlund et al. 2014). Although mustard seed has more advantages in wider distribution, abundance, and availability at low price compared with Moringa oleifera seed proteins, the only application of the mustard protein for water treatment obtained from the published literature was to remove the turbidity, which limits its versatility in water treatment. Hence, our study is aimed at investigating the potential of mustard seed protein as a natural coagulant for the removal of dye from its aqueous solution.

In this study, direct black 19 (DB-19, Figure 1) was selected as a target pollutant to test the coagulation ability of white mustard seed protein (WMSP) as it is a widely
used anionic dye, especially in some Asian countries (Shi et al. 2007). Meanwhile, both this dye and its reduction product have been proven to be mutagenic (Joachim et al. 1985). A commercial coagulant, polyaluminum chloride (PAC), that is most widely used for water treatment in China was selected to carry out the same experiment as a comparison to evaluate the coagulating performance of WMSP. The removal effects with respect to pH, settling time, temperature, coagulant dosage and influence of inorganic salts presence was investigated.

**MATERIALS AND METHODS**

**Preparation of coagulants and dye-containing wastewater**

White mustard seed (Figure 2), a traditional Chinese medicine used widely to resolve phlegm and dispel colds (vocabulary of traditional Chinese medical science), was purchased from a local pharmacy in Zhengzhou, China. The WMSP was extracted in the same way as described in our previous study (Tie et al. 2015). The only difference from the previous method was that the dialysis tube with a molecular weight cut-off of 3,500 Da was used in this study. The WMSP content was measured at 596 nm (UVmini-1240, Shimadzu, Japan) using bovine serum albumin as the standard (Zhou & Chen 2003). The PAC solution at the same concentration was synthesized by adding PAC into deionized water.

The DB-19 solution used in this study was prepared using the same method described in our previous study (Tie et al. 2015). The pH was adjusted to different values in the range of 4 to 9 using 0.1 M HCl and NaOH solution.

**Jar tests**

The jar tests were carried out using standard jar test equipment with six paddle gang stirers (Model ZR4-6, Shenzhen, China). The effect of reaction time and pH on the coagulation removal of DB-19 was conducted with an initial DB-19 concentration of 30 mg/L at 25 °C in the pH range of 4–10. The influence of coagulant dosage was studied at initial DB-19 concentrations of 40, 100, and 300 mg/L at pH 8 and 25 °C, respectively. The effects of temperature and inorganic salt were conducted at an initial DB-19 concentration of 30 mg/L and pH 8.

For all the tests the equipment was programmed to mix the beakers with 200 mL of the dyeing solutions in the following procedure: rapidly mixed (120 rpm) for 1 min and followed by slow mixing (45 rpm) for 20 min (Sánchez-Martín et al. 2010). The two coagulants were dosed into the beakers at the beginning of rapid mixing. Mixed matrices were collected from 2 cm below the water surface using a syringe, and the residual DB-19 concentration was determined using a UV-6000 spectrophotometer (Shanghai Metash instrument Co., Ltd, China) at the wavelength of
655 nm. The removal rate of DB-19 was calculated by the following equation:

\[ R = \frac{(C_0 - C_e)}{C_0} \times 100\% \] (1)

where \( C_0 \) and \( C_e \) are the initial and final concentrations of DB-19 in solution (mg L\(^{-1}\)), respectively.

**Characterization**

Fourier transform infrared spectroscopy (FTIR, Thermo Scientific NicoletiS10, USA) and X-ray photoelectron spectroscopy (XPS, Thermo Scientific ESCALAB 250Xi, USA) were used to characterize WMSP, DB-19, and the reaction products between WMSP and DB-19.

**RESULTS AND DISCUSSION**

**Effect of settling time and pH on DB-19 removal**

The commercial PAC, which has been used for the removal of dyes (Shi et al. 2007; Huang et al. 2014), was used as a comparison to evaluate the coagulation ability of WMSP. Figure 3 shows the effect of settling time on DB-19 removal for both WMSP and PAC coagulants in the pH range of 4–10. Generally, the residual concentration of DB-19 decreased with increasing settling time for both of the two coagulants. The DB-19 concentrations decreased quickly within the first 30 min since the large flocs formed by both of the two coagulants during the reaction stage of slow mixing sank quickly. However, the slow sedimentation of the residual fine flocs led to a decelerating decrease of DB-19 in the following stage, and the complete sedimentation of the flocs led to invariant residual DB-19 concentrations at the settling time of 180 min.

The DB-19 removals at different pH using two coagulants are shown in Figure 4 for the two coagulants. It can be seen that the DB-19 removals decreased from 98.4 to 46.2% and 99.0 to 41.1% as the pH increased from 4 to 10 for WMSP and PAC, respectively. This indicated that a lower pH was favorable for DB-19 removal for both coagulants. Although the performance of WMSP was comparable to PAC, except at pH 4 and 5, WMSP was still very efficient for DB-19 removal.

**Effect of coagulant dosage on DB-19 removal**

Coagulant dosage is a very important parameter to evaluate the efficiency of the coagulation process (Boulaadjoul et al.
Hence, the effect of coagulant dosage on DB-19 removal was tested at DB-19 concentrations of 40, 100 and 300 mg/L because it was important that different concentrations were selected to test the significance of their impact. As shown in Figure 5, the removal efficiency of DB-19 increased significantly from 58.7 to 83.5% when the dosage of WMSP was increased from 9.0 to 18.0 mg/L for the initial DB-19 concentration of 40 mg/L. The corresponding removals for the initial DB-19 concentrations of 100 and 300 mg/L increased from 61.9 to 90.9% and from 53.3 to 89.2% as the dosage of WMSP was increased from 9.0 to 36.0 mg/L and 33.8 to 63.0 mg/L, respectively, indicating that an increasing dosage of WMSP could improve the removal of DB-19 in the tests. In the case of the same tests using PAC as the coagulant, the removal efficiency was lower at each dosage. Therefore, WMSP was shown to be more efficient for DB-19 removal than PAC in the experiment.

**Effect of temperature on DB-19 removal**

Figure 6 shows the effect of temperature on DB-19 removal in the range of 10–45 °C. It can be seen that the removal rate varied at 31.7–43.7% for PAC and 70.0–83.2% for WMSP, respectively. At each temperature set for the experiment, WMSP performed better than PAC for DB-19 removal. The optimal temperature for DB-19 removal by both of the two coagulants was 25 °C, and the corresponding removal rates were 43.7 and 83.2% for PAC and WMSP, respectively.
The test results indicated that WMSP was more efficient than PAC at all selected temperatures.

**Effect of the presence of inorganic salt on DB-19 removal**

Sodium sulfate is widely adopted to improve dyeing performance and fixation property by enhancing the dye molecules transfer from the solution to cotton fibers when direct dyes are used (Teixeira *et al.* 2012). Hence, the effect of Na$_2$SO$_4$ on DB-19 removal was investigated. Figure 7 shows that, overall, Na$_2$SO$_4$ weakened the DB-19 removal for both coagulants, whereas WMSP still outperformed PAC in the presence of Na$_2$SO$_4$.

**Mechanism of DB-19 removal by WMSP**

The effect of pH on the flocculation is critical to explore the coagulation mechanism (Teixeira *et al.* 2012). Figure 8 shows the effect of pH on WMSP content. The lowest WMSP content occurred at pH 9.0, indicating the isoelectric point of WMSP was around pH 9.0 due to the fact that protein as an amphoteric compound has the least solubility at the isoelectric point (Zhao *et al.* 2011). Hence, WMSP was positively charged by adsorbing H$^+$ on its amino-groups in the pH range of 9 set for the experiment.

Figure 9(a) shows the spectrum of WMSP, DB-19 and their flocs formed by WMSP and DB-19. The band at 2,960 cm$^{-1}$ was ascribed to the CH$_3$ asymmetrical stretching (Fournier *et al.* 2008). The bands at 1,654.6, 1,540.8 and 1,241.9 cm$^{-1}$ were assigned to the C=O stretching (Amide I), the CN stretching (Amide II) and the NH bending (Amide III), respectively (Kong & Yu 2007). Figure 9(b) shows the spectrum of DB-19. The bands at 1,616.9 and 1,483 cm$^{-1}$ were ascribed to the C=C stretching of the benzenoid ring (Wang *et al.* 2007; Ge *et al.* 2012). The band at 1,045.2 cm$^{-1}$ in the dye was ascribed to SO$_3^-$ stretching (Abd-alredha *et al.* 2012). The band at

---

**Figure 6** | Effect of temperature on DB-19 removal (C$_0$ = 30 mg/L, pH = 8, settling time = 120 min).

**Figure 7** | Effect of inorganic salt on DB-19 removal by WMSP and PAC on DB-19 removal (C$_0$ = 30 mg/L, pH = 8, settling time = 2 h).

**Figure 8** | WMSP content variety as a function of pH.
838.9 cm\(^{-1}\) belonged to the C\(_2\)H\(_4\) bending (Khan et al. 2013). Figure 9(c) shows the spectrum formed by DB-19/WMSP flocs. The bands of WMSP at 1,654.6 and 1,540.8 cm\(^{-1}\) shifted to 1,631.5 and 1,562.0 cm\(^{-1}\), respectively. The bands of DB-19 at 838.9 cm\(^{-1}\) shifted to 833.1 cm\(^{-1}\), which resulted from the electrostatic attraction between the negatively charged sulfo groups of DB-19 and the protonated amino groups of the WMSP.

To further reveal the removal of DB-19 by WMSP, XPS was employed to monitor the elemental composition and chemical state of DB-19, WMSP and their flocs formed by WMSP and DB-19, respectively. As shown in Figure 10(a), DB-19, WMSP and their flocs formed by WMSP and DB-19 consisted of the same elements of C, O, N and S. Moreover, there was an Na 1 s peak around 1,068 eV in DB-19, which was assigned to Na\(^+\). Through the linear least-squares fit program by using Gauss–Lorentzian peak shapes (Yamashita & Hayes 2006), it was found that there were two chemical states of S 2p in DB-19 (2p\(_{3/2}\) 169.3 and 167.8 eV), which was attributed to -S(=O)\(_2\)-O-Na and -S(=O)\(_2\)-O-H (Figure 10(b)). Typically, there were also two chemical states of S 2p in WMSP, namely 2p\(_{3/2}\) of 167.6 and 163.5 eV, assigned to -S-S- and -SH, respectively (Figure 10(c)). After DB-19 reacted with WMSP, the chemical state of S 2p of -S(=O)\(_2\)-O-Na disappeared and a new chemical state S2p\(_{3/2}\) (168.7 eV) appeared. This was due to the formation of DB-19/WMSP flocs. Moreover, the binding energy of S2p\(_{3/2}\) of -S(=O)\(_2\)-O-H in DB-19 decreased to 167.3 eV, which was caused by the strong hydrogen bonding between -S(=O)\(_2\)-O-H of DB-19 and hydrogen bonding donors (e.g. -NH\(_2\), -COOH) (Figure 10(d)). It should be noted that -S-S- as the internal chemical information of the WMSP/DB-19 flocs could not be detected due to the limitation of XPS. This result is in accordance with the research of Iwahashi et al. (2009) and Du et al. (2015), who have undertook in-depth studies of the difference between the surface and interior of materials. According to the analysis of isoelectric point of WMSP, FT-IR and XPS results, it is concluded that adsorption and charge neutralization was the main mechanism for the coagulation reaction between WMSP and DB-19. Ndabigengesere et al. (1995) reported that adsorption and charge neutralization was the mechanism for the removal of turbidity using Moringa oleifera seed protein with isoelectric points between pH 10 and 11 as the coagulant. The reaction between WMSP and DB-19 can be described by the following equations:

\[
D - \text{S-O-Na} \rightleftharpoons D - \text{O}^- + \text{Na}^+ \tag{2}
\]

\[
W - \text{C-NH}_2 + \text{H}^+ \rightleftharpoons W - \text{C-NH}_3^+ \quad \text{(pH < 9)} \tag{3}
\]

\[
\text{D-S-O}^+ + \text{W-C-NH}_3^+ \rightarrow \text{D-S-O} - \text{W-C-NH}_3 \tag{4}
\]

where D and W denote DB-19 and WMSP, respectively.

The decrease of residual DB-19 content with descending pH shown in Figure 3 can be explained by the mechanism mentioned above. The adsorption and charge neutralization between WMSP and DB-19 was weakened, resulting in higher residual DB-19 content since WMSP molecular was negatively charged at pH 10 higher than its isoelectric point of pH 9. However, the reaction was reinforced by the increase of positive charge of WMSP with decreasing...
pH below its isoelectric point to create lower DB-19 residual.

CONCLUSIONS

This study describes an attempt of removal of the anionic dye DB-19 from water using proteins extracted from white mustard seed as a natural coagulant. The results indicate that the coagulation efficiency of WMSP was better than PAC at the same dosage for all other experiments except at pH 4 and 5. The main mechanism of the removal of DB-19 by WMSP could be adsorption and charge neutralization.

To our knowledge, it is the first report on removal of an anionic dye using WMSP as a natural coagulant, and the results showed that WMSP had excellent coagulation ability to remove DB-19.

ACKNOWLEDGEMENTS

This work was supported by the science and technology major project of Henan Province [grant numbers 161100310700].
REFERENCES

Abidin, Z. Z., Shamsudin, N. S. M., Madehia, N. & Sobri, S. 2012 Synthesis and characterization of azo dye para red and new derivatives. J. Chem. 9, 465–470.

Abidin, Z. Z., Shamsudin, N. S. M., Madehia, N. & Sobri, S. 2013 Optimization of a method to extract the active coagulant agent from Jatropha curcas seeds for use in turbidity removal. Ind. Crop Prod. 41, 519–523.

Antov, M. G., Šćiban, M. B. & Prodanović, J. M. 2012 Evaluation of the efficiency of natural coagulant obtained by ultrafiltration of common bean seed extract in water turbidity removal. Ecol. Eng. 49, 48–52.

Banerjee, C., Ghosh, S., Sen, G., Mishra, S., Shukla, P. & Bandopadhyay, R. 2013 Study of algal biomass harvesting using cationic guar gum from the natural plant source as flocculant. Carbohydr. Polym. 92, 675–681.

Bashiri, B., Fallah, N., Bonakdarpour, B. & Elyasi, S. L. 2018 The development of aerobic granules from slaughterhouse wastewater in treating real dyeing wastewater by Sequencing Batch Reactor (SBR). J. Environ. Chem. Eng. 6, 5536–5543.

Betache, H., Aouabed, A., Drouiche, N. & Lounici, H. 2014 Conditioning of sewage sludge by prickly pear cactus (Opuntia ficusIndica) juice. Ecol. Eng. 70, 465–469.

Bilińska, L., Murek, M. & Ledakowicz, S. 2017 Textile wastewater treatment by AOPs for brine reuse. Process Safe Environ. Prot. 109, 420–428.

Bodlund, I., Pavankumar, A. R., Chelliah, R., Kasi, S., Sankaran, K. & Rajarao, G. K. 2014 Coagulant proteins identified in mustard seed: a potential water treatment agent. Int. J. Environ. Sci. Technol. 11, 873–880.

Boulaadjoul, S., Zemmouri, H., Bendjama, Z. & Drouiche, N. 2018 A novel use of moringa oleifera seed powder in enhancing the primary treatment of paper mill effluent. Chemosphere 206, 142–149.

Chethana, M., Sorokhaibam, L. G., Bhandari, V. M., Raja, S. & Ranade, V. V. 2016 Green approach to dye wastewater treatment using biocoagulants. ACS Sustain. Chem. Eng. 4, 2495–2507.

Costa, A. F. S., Albuquerque, C. D. C., Salgueiro, A. A. & Sarubbo, L. A. 2018 Color removal from industrial dyeing and laundry effluent by microbial consortium and coagulant agents. Process Saf. Environ. Prot. 118, 203–210.

Du, C. B., Hu, X. L., Guan, P., Guo, L. X., Qian, L. W., Song, R. Y., Li, J. & Wang, C. L. 2015 Water-compatible surface-imprinted microspheres for high adsorption and selective recognition of pesticide drug from aqueous media. J. Mater. Chem. B 3, 3044–3053.

Fatombi, J. K., Lartiges, B., Aminou, T., Barres, O. & Caillet, C. 2013 A natural coagulant protein from copra (cocos nucifera): isolation, characterization, and potential for water purification. Sep. Purif. Technol. 116, 35–40.

Ferhat, M., Kadouche, S., Drouiche, N., Messaoudi, K., Messaoudi, B. & Lounici, H. 2016 Competitive adsorption of toxic metals on bentonite and use of chitosan as flocculent coagulant to speed up the settling of generated clay suspensions. Chemosphere 165, 87–93.

Flaten, T. P. 2001 Aluminium as a risk factor in Alzheimer's disease, with emphasis on drinking water. Brain Res. Bull. 55, 187–196.

Fournier, F., Gardner, E. M., Kidra, D. A., Donaldson, P. M., Guo, R. & Butcher, S. A. 2008 Protein identification and quantification by two-dimensional infrared spectroscopy: implications for an all-optical proteomic platform. Proc. Natl. Acad. Sci. USA 105, 15352–15357.

Ge, L., Han, C. C. & Liu, J. 2012 In situ synthesis and enhanced visible light photocatalytic activities of novel PANI–g-C3N4 composite photocatalysts. J. Mater. Chem. 48, 24959–25006.

Huang, X., Bo, X. W., Zhao, Y. X., Gao, B. Y., Wang, Y., Sun, S. L., Yue, Q. Y. & Li, Q. 2014 Effects of compound bioflocculant on coagulation performance and floc properties for dye removal. Biosens. Bioelectron. 65, 116–121.

Iwasashi, T., Nishi, T., Yamane, H., Miyamae, T., Kanai, K. & Seki, K. 2009 Surface structural study on ionic liquids using metastable atom electron spectroscopy. J. Phys. Chem. 113, 19237–19243.

Joachim, F., Burrell, A. & Andersen, J. 1985 Mutagenicity of azo dyes in the Salmonella/microsome assay using in vitro and in vivo activation. Mutat. Res. 156, 131–138.

Kebaili, M., Djellali, S., Radjai, M., Drouiche, N. & Lounici, H. 2018 Valorization of orange industry residues to form a natural coagulant and adsorbent. J. Ind. Eng. Chem. 64, 292–299.

Khan, M. S. U., Akhter, Z., Iqbal, N. & Siddiqui, M. 2013 Synthesis, characterization and morphological studies of some novel iloxane-based block copolymeric materials containing organometallic as well as organic polyesters. J. Organom. Chem. 745–746, 312–328.

Kong, J. L. & Yu, S. N. 2007 Fourier transform infrared spectroscopic analysis of protein secondary structure. Acta. Bioch. Biop. Sin. 39, 549–559.

Lafi, R., Gzar, L., Lajimi, R. H. & Hafiane, A. 2018 Treatment of textile wastewater by a hybrid ultrafiltration/electrodialysis process. Chem. Eng. Process. Process Intensif. 132, 105–113.

Mahmoodi, N. M., Abdi, J., Oveisi, M., Asli, M. A. & Vossoughi, A. 2018 Metal-organic framework (MIL-100 (Fe): synthesis, characterization and morphological studies of some novel metal-organic framework (MIL-100 (Fe)): synthesis, detailed photocatalytic dye degradation ability in colored textile wastewater and recycling. Mater. Res. Bull. 100, 357–366.

Ndabigengesere, A., Narasiah, K. S. & Talbot, B. G. 1995 Active agents and mechanism of coagulation of turbid waters using morgina oleifera. Water Res. 29, 703–710.

Polizzi, S., Pira, E., Ferrara, M., Bugiani, M., Papaleo, A., Albera, R. & Palmi, S. 2002 Neurotoxic effects of aluminium among foundry workers and Alzheimer's disease. Neurotoxicology 23, 761–774.

Pritchard, M., Craven, T., Mkandawire, T., Edmondson, A. S. & O’Neill, J. G. 2010 A comparison between Moringa oleifera and chemical coagulants in the purification of drinking water.
White mustard seed (Semen sinapis) protein for direct black 19 removal

First received 5 March 2019; accepted in revised form 22 May 2019. Available online 24 July 2019

– an alternative sustainable solution for developing countries. Phys. Chem. Earth 35, 798–805.

Sánchez-Martín, J., Ghebremichael, K. & Beltrán-Heredia, J. 2010 Comparison of single-step and two-step purified coagulants from moringa oleifera seed for turbidity and DOC removal. Bioresour. Technol. 101, 6259–6261.

Šćiban, M., Klašnja, M., Antov, M. & Škrbic, B. 2009 Removal of water turbidity by natural coagulants obtained from chestnut and acorn. Bioresour. Technol. 100, 6639–6645.

Shi, B. Y., Li, G. H., Wang, D. S., Feng, C. H. & Tang, H. X. 2007 Removal of direct dyes by coagulation: the performance of preformed polymeric aluminum species. J. Hazard Mater. 145, 567–574.

Teixeira, C. M. L. L., Kirsten, F. V. & Teixeira, P. C. N. 2012 Evaluation of moringa oleifera seed flour as a floculating agent for potential biodiesel producer microalgae. J. Appl. Phycol. 24, 557–563.

Tie, J. X., Jiang, M. B., Li, H. C., Zhang, S. & Zhang, X. W. 2015 A comparison between moringa oleifera seed presscake extract and polyaluminum chloride in the removal of direct black 19 from synthetic wastewater. Ind. Crop. Prod. 74, 530–534.

Tie, J. X., Chen, D. G., Zhao, M. Q., Wang, X. L., Zhou, S. G. & Peng, L. G. 2016 Adsorption of reactive green 19 from water using polyaniline/bentonite. J. Water Reuse Desal. 6, 515–523.

Tie, J. X., Zheng, Z. H., Zheng, X. M. & Hao, Y. R. 2017 Comparison of the performance of Moringa oleifera seed protein and polyaluminum chloride in removal of humic acid from water. Desalination. Water Treat. 90, 214–219.

Verma, A. K., Bhunia, P. & Dash, R. R. 2012a Supremacy of magnesium chloride for decolourisation of textile wastewater: a comparative study on the use of different coagulants. Int. J. Environ. Sci. Dev. 3, 119–123.

Verma, A. K., Dash, R. R. & Bhunia, P. 2012b A review on chemical coagulation/flocculation technologies for removal of colour from textile wastewaters. J. Environ. Manage. 93, 154–168.

Wang, X. F., Chang, S. L., Yang, J. K., Tan, J. C., Jia, H. H. & Yin, H. W. 2007 Detection of TNT in acetone using raman spectroscopic signature. In: Proceedings of SPIE, the International Society for Optical Engineering. Society of Photo-Optical Instrumentation Engineers, Beijing, China, pp. 662219.1–662219.8.

Wang, W. Y., Yue, Q. Y., Li, R. H., Song, W., Gao, B. Y. & Shen, X. 2017 Investigating coagulation behavior of chitosan with different Al species dual-coagulants in dye wastewater treatment. J. Taiwan Inst. Chem. Eng. 78, 423–430.

Wang, X. H., Jiang, C. L., Hou, B. X., Wang, Y. Y. & Hao, C. J. B. 2018 Carbon composite lignin-based adsorbents for the adsorption of dyes. Chemosphere 206, 587–596.

Weisburger, J. H. 2002 Comments on the history and importance of aromatic and heterocyclic amines in public health. Mutat. Res. 506, 9–20.

Yamashita, T. & Hayes, P. 2006 Effect of curve fitting parameters on quantitative analysis of FeO and FeO1.94 using XPS. J. Electron. Spectros. Relat. Phenomena 152, 6–11.

Yan, B., Xu, H., Yang, S. N., Bi, S. T., Li, F., Shen, C. S., ChunyanMa, C. Y., Tian, Q., JiansheLiu, J. S., XinshanSong, X. S., Sand, W. G. & Liu, Y. B. 2018 Treatment of industrial dying wastewater with a pilot-scale strengthened circulation anaerobic reactor. Bioresour. Technol. 264, 154–162.

Zhao, X., Tong, X., Chen, X., Han, M. Y., Xu, X. L. & Zhou, G. H. 2017 Yield, thermal denaturation, and microstructure of proteins isolated from pale, soft, exudative chicken breast meat by using isoelectric solubilization/precipitation. Process Biochem. 58, 167–173.

Zhou, Q. Z. K. & Chen, X. D. 2001 Immobilization of β-galactosidase on graphite surface by glutaraldehyde. J. Food. Eng. 48, 69–74.