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ORIGINAL PAPER

Magnetically assisted coagulation using iron oxide nanoparticles-Leucaena leucocephala seeds' extract to treat synthetic Congo red wastewater

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Abstract

This study focused on utilization of Leucaena leucocephala seeds' extract and iron oxide nanoparticles (IONPs) under magnetic field to provide effective coagulation process. Leucaena seeds' extract was obtained using 3 M NaCl solution as solvent at feed-to-solvent mass ratio of 1:20. IONPs were synthesized using co-precipitation method. The obtained IONPs were analyzed using scanning electron microscopy, X-ray diffraction and transmission electron microscopy. Based on analysis results, the obtained IONPs were the agglomerate of Fe_3O_4 phase with average particle diameter of 14.6 nm. The extract and IONPs were used as natural coagulant at various pHs (3–12), coagulant dosages (4–40 mL extract/L), and IONPs concentrations (0.5–3 mg/mL extract) under external magnetic field. It was obtained that pH 3 was the best pH because positively charged Leucaena protein at pH 3 acts to neutralize negatively charged Congo red molecules. The highest removal was obtained at coagulant dosage of 20 mL/L, and further increase in dosage was decreasing the removal due to colloid re-stabilization effect. The variation of IONPs concentration did not give any effect on the %removal and sludge volume. However, further investigation using pseudo-second-order kinetic model showed that the increase in IONPs concentration gave increase to the kinetic constant, signifying faster settling kinetics. The best condition was obtained at pH 3, coagulant dosage 20 mL/L, and IONPs concentration of 3 mg/mL extract that gave 90% Congo red removal, 30 mL/L sludge volume, and 30 min settling time.

Keywords Congo red · Iron oxide nanoparticles · Leucaena leucocephala · Magnetically assisted coagulation · Natural coagulant

Introduction

Nowadays, dye-containing wastewater becomes a serious environmental issue as it could cause problems to environment and human health. Various wastewater treatment techniques have been explored, namely adsorption using biomass-based adsorbents (Albadarin et al. 2017; Daneshvar et al. 2017; Naushad et al. 2016b; Sharma et al. 2015, 2018), nanocomposite materials (Alqadami et al. 2016, 2017, 2018; Tatarchuk et al. 2019), and other materials (Mironyuk et al. 2019; Naushad et al. 2016a); photodegradation (Naushad et al. 2019; Sharma et al. 2018); membrane separation (Dasgupta et al. 2015); and advanced oxidation processes (AOPs) (Hudaya et al. 2017). Although the previously mentioned techniques have advantages to treat dye-containing wastewater, coagulation and flocculation are still commonly used method to treat water and wastewater (Meric et al. 2005).

Generally, coagulants could be categorized as chemical and natural coagulants. Furthermore, chemical coagulants could be classified as hydrolyzing metallic salts, pre-hydrolyzing metallic salts, and synthetic cationic polymers (Freitas et al. 2018). Although chemical coagulants are widely used, there are some downsides of their utilization. Metallic salts are known to produce high-volume residual sludge with possible environmental impact, potential neurotoxic diseases (dementia, Alzheimer's) on human health, and changes of treated water's pH (Choy et al. 2015; Yin 2010). Synthetic cationic polymers that were meant to replace metallic salts also have their drawback. These come from its toxic derivatives that come from unreacted monomers, unreacted

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chemicals from the production of synthetic polymers, and reaction of synthetic polymers and water (Oladoja 2015).

In recent years, many researchers have explored utilization of natural coagulant from plant and non-plant resources. Several plant-based coagulants such as *Moringa oleifera* (Baptista et al. 2017), *Moringa stenopetala* (Dalvand et al. 2016a), *Jatropha curcas* (Abidin et al. 2017), *Plantago major* (Ramavandi 2014), *Carica papaya* (Kristianto et al. 2018a), and orange (Kebaili et al. 2018) have been explored. Some non-plant-based coagulants are also used, namely fish scales (Musa et al. 2015), chitosan (Meraz et al. 2016), and algal alginate (Vijayaraghavan and Shanthakumar 2016). These natural resources could provide alternative solution to treat water and wastewater with similar effectiveness to chemical coagulants.

Leucaena leucocephala, also known as leucaena, white popinac, or *lamtoro gung* (in Indonesian), is a tropical tree that easily found in Indonesia. Utilization of leucaena is relatively limited, mainly as food source and forage (Evitayani et al. 2004). Leucaena seeds is known to have high protein content (approximately 50%w dry basis) (Sethi and Kulkarni 1994), and most of the proteins are globulin fraction, which is soluble in saline water (Sethi and Kulkarni 1993). In our previous works, we have utilized leucaena as natural coagulant to treat turbidity and color in synthetic wastewater (Kristianto et al. 2018b, 2019). Leucaena seeds' extract gave comparable performance to commercial alum with 50% lower sludge volume. However, long settling time is still needed (> 1 h) to separate flocs and treated water. This long residence time results in bigger size of settling tanks, leading to higher capital cost. Some efforts are needed to make this process more effective in shorter time.

Iron oxide nanoparticles (IONPs) have gained a lot of interest in these recent years. Various applications have been explored, such as heavy metal and organic substance adsorption from water, photocatalyst, and support in wastewater treatment using immobilization techniques (Lofrano et al. 2016; Xu et al. 2012). There are some advantages and unique characteristic of IONPs such as relatively cheap price, low toxicity, long shelf life, and biocompatible (Laurent et al. 2008). Its biocompatibility property is making possible interaction of IONPs with various functional groups in organic substances, such as carboxylic acid, phosphonic acid, dopamine, trimethoxy silane, cysteine, and amine (Dias et al. 2011).

Using this property, IONPs could be utilized together with protein as natural coagulant. Previously, several researchers have explored the utilization of Fe_3O_4, α and γ-Fe_2O_3 for this application. Okoli et al. (2012) have used purified moringa (*Moringa oleifera*) extract to functionalize mixture of Fe_3O_4 and γ-Fe_2O_3 nanoparticles. The IONPs–moringa combination in this study gave 80% turbidity removal, which was higher when compared to only moringa extract (70%) and IONPs (40%) were used independently, with faster removal (12 min), compared to gravity settling (240 min) (Okoli et al. 2012). Santos et al. (2016) observed the effect of oleic acid on the coating of γ-Fe_2O_3 IONPs. It was found that the presence of oleic acid coating decreased the coagulation performance, and furthermore, increased the organic demand in the treated water. The best result was obtained in the presence of IONPs under magnetic fields, with 90% of removal 10 mg/L IONPs concentration and 400 mg/L extract at 30 min of settling (Santos et al. 2016). Santos et al. (2018a) utilized α-Fe_2O_3 nanoparticles and moringa extract to treat textile wastewater. From this study, it was confirmed that iron oxide nanoparticles-natural coagulant combination could give comparable coagulation performance with natural coagulant only at shorter settling time (Santos et al. 2018a). Similar result was also obtained by Mateus et al. (2018b) who used Fe_3O_4 nanoparticles with moringa extract to treat tartrazine-containing water (Mateus et al. 2018b).

In this study, we combined IONPs and extract of leucaena seeds to treat synthetic Congo red wastewater. To the best of authors' knowledge, the combination of IONPs and leucaena seed extract in magnetically assisted coagulation process has never been used before. IONPs were synthesized using basic co-precipitation method. This method is known to have some advantages, such as simple preparation method, fast reaction, high yield, and ease in scale-up (Wu et al. 2008, 2015). The obtained IONPs would be used in coagulation process with crude extract of leucaena. The effect of pH, coagulant dosage, and IONPs concentration on the removal of Congo red was observed. This research was carried out at Laboratory of Water and Waste Treatment Technology, Department of Chemical Engineering, Parahyangan Catholic University, from January to August 2019.

Materials and methods

Synthesis of IONPs and characterization

The IONPs were synthesized using co-precipitation method at room temperature and atmosphere, based on previous researchers (Khalil 2015; Rashad et al. 2012; Mateus et al. 2018a). To summarize, $FeSO_4$ and $Fe(NO_3)_3 \cdot 9H_2O$ with molar ratio of 1:2 were dissolved in 10 mL distilled water. 1.2 g NaOH was dissolved in 2 mL distilled water and then added dropwise to the iron solution. The mixture was agitated for 15 min. The obtained black precipitate was separated using magnet, then repeatedly washed using demineralized water and ethanol, and oven-dried at 50 °C for 4 h. The IONPs were characterized using scanning electron microscope–energy-dispersive X-ray spectroscopy (SEM–EDS) Hitachi SU3500, transmission electron microscope (TEM)

Hitachi HT7700, and X-ray diffraction Bruker D8 Advance. The crystallite size (d) was estimated from XRD spectra using Scherrer Eq. (1), where K is Scherrer constant (0.9), λ is the wavelength, β is the full width at half maximum peak (FWHM), and θ is the Bragg angle.

$$d = \frac{K \cdot \lambda}{\beta \cdot \cos\theta} \tag{1}$$

Leucaena seeds' extract preparation

The leucaena seeds' crude extract was prepared following our previous experiment (Kristianto et al. 2019). To summarize, 5 g leucaena seeds' kernel powder was mixed with 100 mL 3 M NaCl solution and mixed for 30 min. The extract was separated from the solids by means of centrifugation (5000 rpm, 10 min) and filtration, and used as natural coagulant without further treatment. The extract was freshly made before every experiment to prevent degradation of active coagulating agent due to storage.

Jar test experiment

Congo red was used as a model substance in this coagulation study. Congo red solution with the initial concentration of 50 ppm was mixed with the IONPs–extract coagulant. The mixture pH was adjusted prior to coagulation using 0.1 HCl or NaOH. The coagulant preparation was done by mixing various amounts of IONPs into 20 mL leucaena seeds' extract. The mixture was sonicated for 5 min in a sonicator bath and agitated for 1 h. The mixture of Congo red solution and coagulant was rapidly mixed at 200 rpm for 3 min, followed by slow mixing at 60 rpm for 30 min. The settling was done with the presence of external magnetic field, which was provided by array of neodymium magnets for 1 h. The sample was taken every 5 min until 1 h to observe the effect of magnetic field on the settling process. The %removal was calculated using Eq. 2, where C_i and C_f are the initial and the final Congo red concentrations (ppm), measured using spectrophotometer visible (General Scientific UV 1800 PC) at maximum wavelength of 570 nm. The sludge volume (mL sludge/L) was measured using Imhoff cone after 1 h settling without magnet, which was calculated using Eq. 3. Variation of variables that were studied is presented in Table 1.

$$\%\text{removal} = \frac{C_\mathrm{i} - C_\mathrm{f}}{C_\mathrm{i}} \times 100\% \tag{2}$$

$$\text{sludge volume} = \frac{V\ \text{sludge (mL)}}{V\ \text{waste water(L)}} \tag{3}$$

The evaluation of the removal kinetics was done by using pseudo-first- and pseudo-second-order adsorption kinetics, calculated using Eqs. 4 and 5, respectively. The k_1 (1/min) and k_2 (g min/mg) are the pseudo-first- and pseudo-second-order kinetic rate constants, and q_t and q_e are the amount of Congo red adsorbed (mg/g) as a function of time and at equilibrium, respectively. The q value was calculated using Eq. 6, where C_i (mg/L) is the initial concentration, C_t (mg/L) is the concentration as a function of time, V (L) is the volume of wastewater, and m is the coagulant dosage (g).

$$\ln\left(q_\mathrm{e} - q_t\right) = \ln\left(q_\mathrm{e}\right) - k_1 t \tag{4}$$

$$\frac{t}{q_t} = \frac{t}{q_\mathrm{e}} + \frac{1}{k_2 q_\mathrm{e}^2} \tag{5}$$

$$q = \frac{\left(C_\mathrm{i} - C_t\right) \times V}{m} \tag{6}$$

Results and discussions

Characterization of IONPs

The morphology of IONPs was observed using SEM and is presented in Fig. 1a. It could be observed that the IONPs obtained were in the form of spherical iron nanoparticles agglomerate. The EDS spectra (Fig. 1b) showed high purity of IONPs with 72.67%w Fe and 27.33%w O, omitting the C peak that came from the carbon tape used in the SEM–EDS

Table 1 Variation of pH, coagulant dosage, and IONPs concentration in this study

Study	Variables		
	pH	Coagulant dosage (mL extract/L waste water)	IONPs concentration (mg IONPs/mL extract)
pH effect	3, 4, 6, 8, 10, 12	20	2
Coagulant dosage effect	Best pH	4, 8, 12, 16, 20, 24, 28, 32, 36, 40	2
IONPs concentration effect	Best pH	Best dosage	0.5, 1, 1.5, 2, 2.5, 3

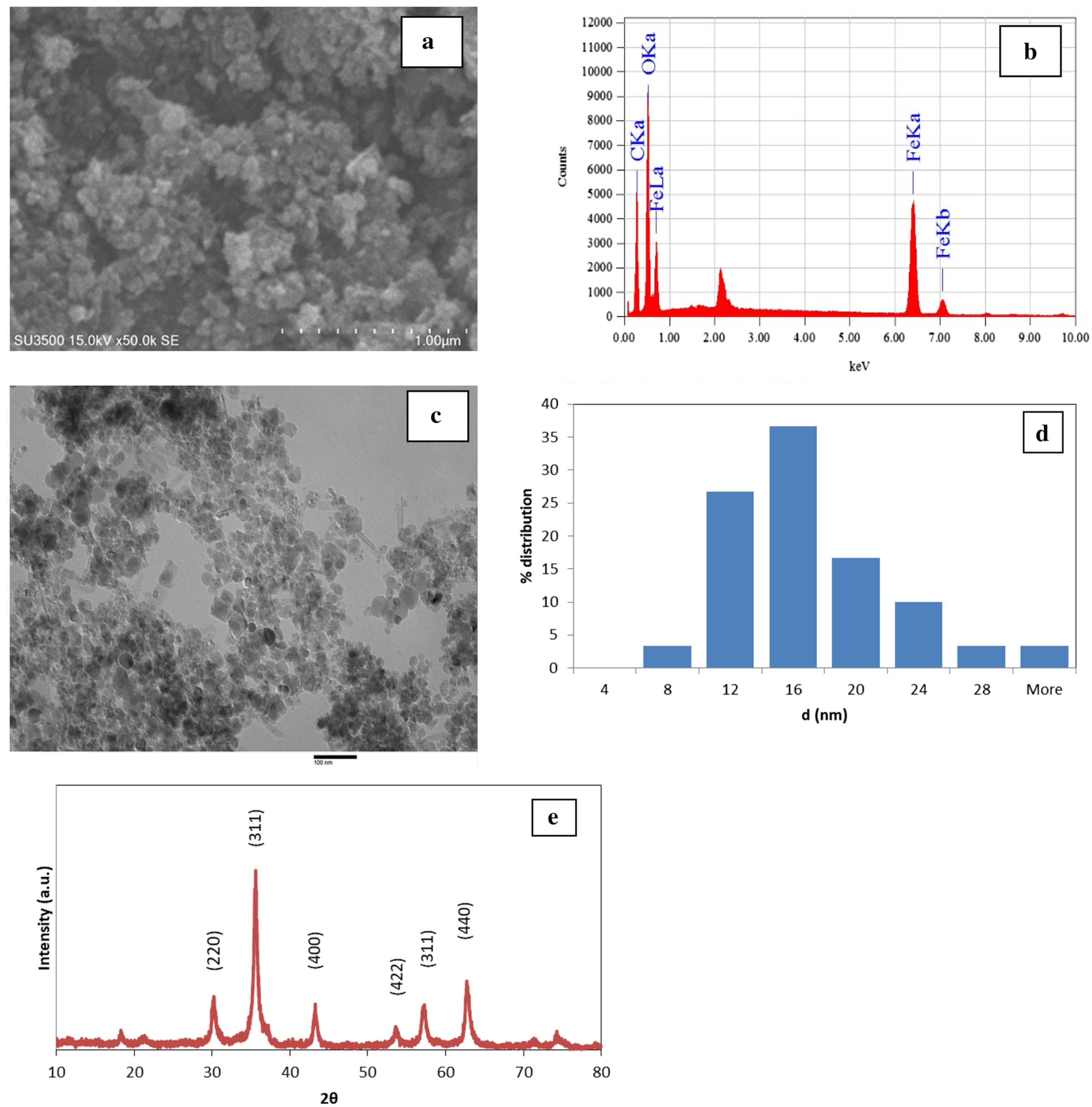

Fig. 1 Morphology of IONPs at magnification of 50,000× (**a**), EDS analysis (**b**), TEM image (**c**) and its diameter distribution (**d**), and XRD spectra of IONPs (**e**)

analysis. This showed a typical Fe_3O_4 composition that has been reported by previous researchers (Mateus et al. 2018b; Hariani et al. 2013). Furthermore, from the TEM image (Fig. 1b), it could be observed that the IONPs had diameter size 9–30 nm following normal distribution (Fig. 1c), with average of 14.6 nm. XRD spectrum of IONPs is presented in Fig. 1d. It could be observed that the IONPs sample exhibited 2θ peaks and its Miller index as follows: 30.19° (220), 36° (311), 43.23° (400), 53.61° (422), 57.19° (511), and 62.77° (440) (Iida et al. 2007; Loh et al. 2008), which are commonly found in magnetite (Fe_3O_4) nanoparticles. In addition, the particle diameter was calculated using Scherrer equation from the XRD spectra indicated with an approximate diameter of 12.6 nm. This result is in good agreement with average diameter measured from TEM image.

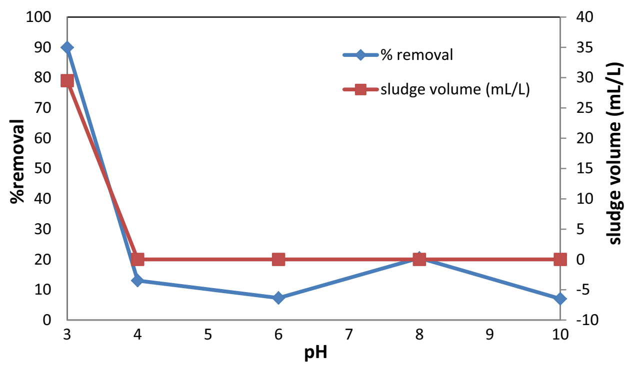

Fig. 2 Profile of %removal and sludge volume at variation of pH

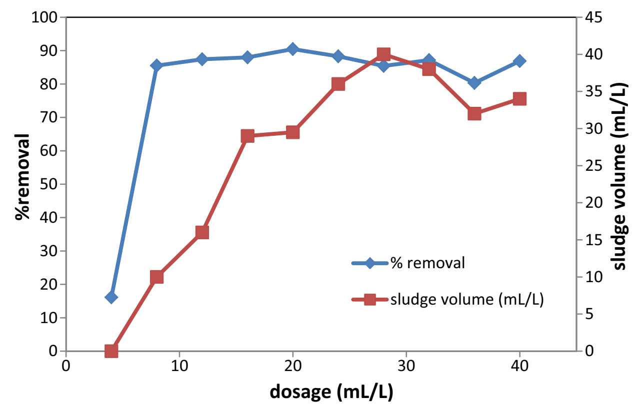

Fig. 3 Profile of %removal and sludge volume at variation of coagulant dosage

Effect of pH on coagulation

The effect of pH on the %removal and sludge volume is presented in Fig. 2. It could be observed that coagulation at pH 3 exhibited the highest %removal, while further increase in pH decreased the %removal. Similar trend was also observed for generated sludge volume. The same trend was obtained when only leucaena seeds' extract was used as coagulant (Kristianto et al. 2019). From this observation, it was clear that the coagulation process was pH-sensitive, and the presence of IONPs did not alter the best pH for coagulation. Thus, it could be concluded that the protein in leucaena seeds' extract was the active coagulating agent. It is known that protein molecule is amphoteric that has positive charge at pH lower than its pI and negative charge at pH higher than its pI value. It is also known that Congo red is an acidic dye with isoelectric point around 3, thus resulting in negatively charged molecule at pH 3.0 and above (Tie et al. 2015). Based on these facts, it could be predicted that adsorption–charge neutralization was the possible mechanism in this study.

Effect of coagulant dosage

The effect of coagulant dosage on the %removal and sludge volume is presented in Fig. 3. It could be observed that at low dosage, low %removal was obtained, as there were not enough active coagulating agents to neutralize the Congo red. With the increase in coagulant dosage, the %removal was also increased; the highest %removal was obtained at dosage of 20 mL/L. Further increase in coagulant dosage resulted in the decrease in %removal. This phenomenon could happen due to the presence of excessive active coagulating agent that resulted in colloid re-stabilization. The active coagulating agent could be adsorbed on previously neutral flocs, thus leading to charge reversal and resulting in electrostatic repulsion between flocs (Choy et al. 2015). The sludge volume trend was also increased with the increase in %removal. The volume was still increasing until 30 mL/L coagulant dosage, regardless of the slight decrease in %removal. As stated before, at excessive coagulant dosage condition, the flocs became re-stabilized, thus hindering the formation of agglomerates. This condition could result in smaller floc size; thus, the sludge became more porous and increasing the sludge volume (Kristianto et al. 2019). Further addition of coagulant decreases the sludge volume, as the % removal was also decreased.

Effect of IONPs concentration

The effect of IONPs concentration on the coagulation performance is presented in Fig. 4. It could be observed that variation of IONPs concentration from 0.5 to 3.0 mg/mL extract did not give any effect to the %removal and sludge volume generated, compared to coagulation without the presence of IONPs (0 mg/mL extract). This was possible because the active coagulating agent was the protein in leucaena extract, while the IONPs did not contribute to the coagulation process. This statement was confirmed by doing similar experiment using only IONPs (Fig. 5), where IONPs did not give any contribution to the removal of Congo red. Further observation of the sludge using light microscope (inset in Fig. 4) confirmed that the sludge that was obtained from IONPs–leucaena extract and leucaena extract only had similar structure and floc size.

The kinetics of Congo red's removal (Fig. 5) was further investigated by using pseudo-first- and pseudo-second-order adsorption kinetics. It is generally known that one of the commonest natural coagulant mechanisms is adsorption followed by charge neutralization, prior to floc formation and settling. Furthermore, according to Beltrán-Heredia et al. (2012) the adsorption stage is the rate determining step; thus, the whole process could be modeled using adsorption equations. The parameters of adsorption kinetics are

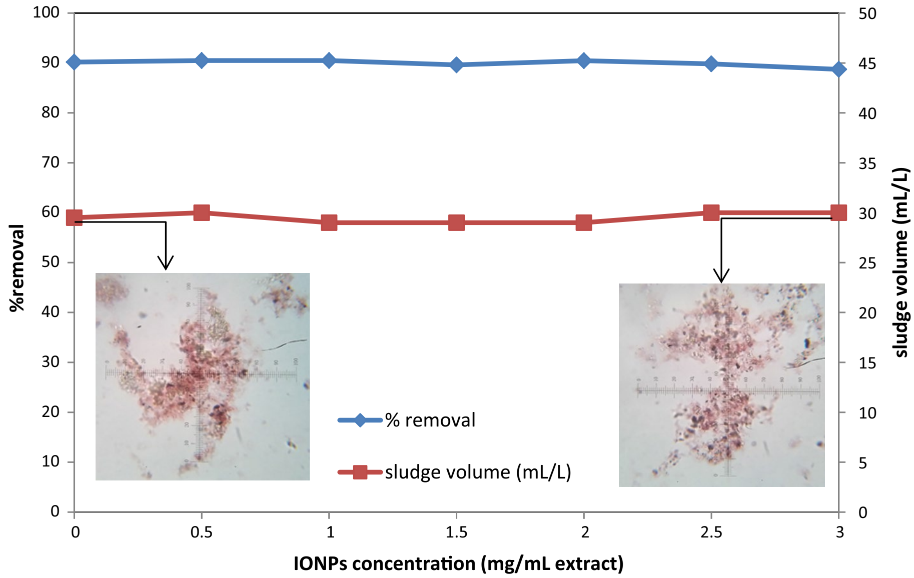

Fig. 4 Profile of %removal and sludge volume at variation of IONPs concentration and its visual floc observation at 0 and 3.0 IONPs concentration mg/mL extract (magnification 400×)

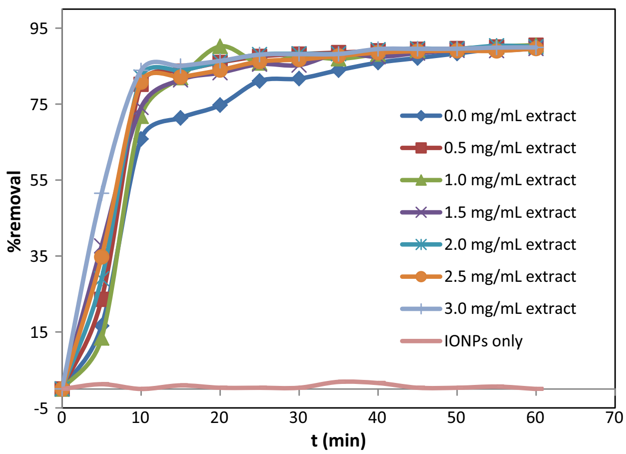

Fig. 5 Profile of %removal kinetics at variation of IONPs concentration

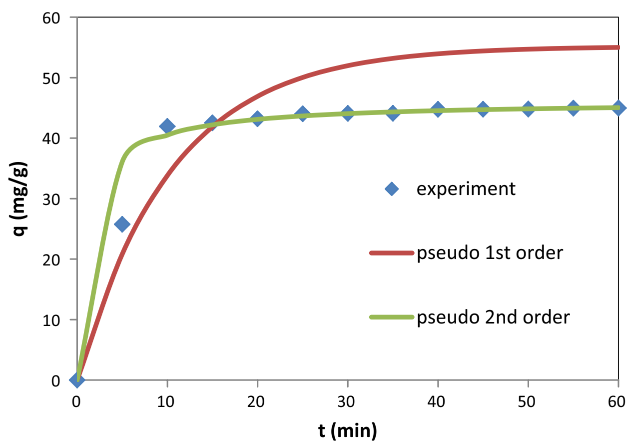

Fig. 6 Fitting for experimental and pseudo-first- and pseudo-second-order model data (pH 3, coagulant dosage 20 mL/L, and IONPs concentration 3.0 mg/mL extract)

Table 2 Parameters of adsorption kinetics

IONPs concentration (mg/mL extract)	Pseudo-first order			Pseudo-second order		
	q_e (mg/g)	k_1 (1/min)	R^2	q_e (mg/g)	k_2 (g min/mg)	R^2
0	55.1823	0.0948	0.8785	49.019	0.001923	0.9996
0.5	15.70001	0.078302	0.8917	46.2963	0.003293	1
1.0	28.41188	0.087744	0.9261	46.72897	0.009785	0.9991
1.5	20.66332	0.083369	0.9345	46.51163	0.009208	0.9998
2.0	17.55497	0.088666	0.8697	46.08295	0.015696	0.9999
2.5	14.96925	0.074157	0.9035	46.08295	0.012624	0.9999
3.0	16.56533	0.133804	0.89	45.45455	0.024948	0.9998

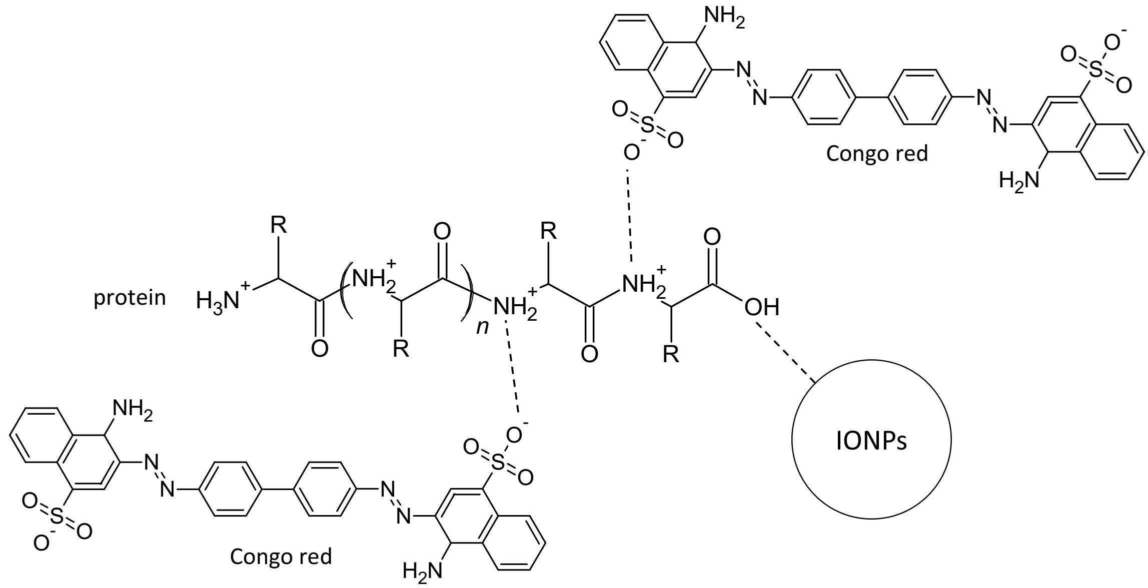

Fig. 7 Illustration for coagulation mechanism

presented in Table 2. It could be observed that pseudo-second-order kinetics was suitable kinetic model for this study, shown by high R^2 value. The suitability of this model, compared to pseudo-first-order kinetics, is also shown in Fig. 6. Similar results were also obtained by previous researchers (Mateus et al. 2018b; Dalvand et al. 2016b). Suitability to the pseudo-second-order kinetics implies chemisorption occurred between the active coagulating agent and colloid. This was possible through the presence of positive sites in the protein and negative functional groups on the Congo red molecules, interacting via hydrogen or dipole–dipole bonding (Jadhav and Mahajan 2014). The illustration of this mechanism is presented in Fig. 7. The interaction of charge neutralization between proteins could be seen as the positive active coagulating agent with negatively charged Congo red molecules. Furthermore, there are interactions between protein and the IONPs that could make faster settling kinetic possible.

Based on the kinetics of %removal, presented in Fig. 5, and the kinetic parameter in Table 2, it could be observed that the increase in IONPs concentration could increase the formation of flocs, thus increasing %removal within shorter time. With the increase in IONPs concentration, the faster the floc formation, the higher the %removal was observed at 5 min. After 10 min, the increase became less significant (less than 10%) and constant after 30 min of settling, where the coagulation with only leucaena seeds' extract needed 60 min to reach same %removal. Similar result was also

Table 3 Comparison of the previous study using magnetically assisted natural coagulant—IONPs coagulation

Natural coagulant	Wastewater	pH	Coagulant dosage	IONPs	Result	References
Leucaena leucocephala (extraction NaCl 3 M, 5 g/100 mL)	Congo red (50 ppm)	3	20 mL/L	Fe_3O_4 3 mg/mL extract	90% color removal 30 min	This study
Moringa oleifera (extraction NaCl 1 M, 1 g/100 mL)	Tartrazine (50 ppm)	3	66 mL/L	Fe_3O_4 1 mg/mL extract	70.16% color removal 10 min	Mateus et al. (2018b)
Moringa oleifera (extraction NaCl 1 M, 1 g/100 mL)	Pirapo River water (80 NTU)	n/a	40 mL/L	γ-Fe_2O_3 1 mg/mL	94% turbidity removal 30 min	Santos et al. (2018b)
Moringa oleifera (extraction NaCl 1 M, 0.02 g/100 mL)	Pirapo River water (143 NTU)	7.12	66 mL/L	Fe_3O_4 1 mg/mL extract	96.8% turbidity removal 30 min	Mateus et al. (2018a)
Moringa oleifera (extraction NaCl 1 M, 0.04 g/100 mL)	Textile wastewater (707.3 mg Pt–Co/L)	3.27	66 mL/L	α-Fe_2O_3 1 mg/mL	92.37% apparent color removal 10 min	Santos et al. (2018a)

shown by the kinetic constants, which increased along with addition of IONPs.

Comparison of our experimental result to that of the previous study is presented in Table 3. It could be seen that the result obtained in this study is comparable in terms of removal and settling time. Further study of coagulation condition optimization, IONPs reusability, and protein purification using IONPs could be explored.

Conclusion

In this study, combination of leucaena seeds' extract and IONPs has been utilized to treat Congo red synthetic wastewater. The IONPs were synthesized using co-precipitation of Fe^{2+} and Fe^{3+} at basic pH solution and atmospheric condition, resulting in the agglomeration of spherical Fe_3O_4 nanoparticles of with average diameter of 14.6 nm. The best pH of coagulation was found to be at pH 3, regardless of the presence of IONPs; thus, it could be concluded that charge neutralization was the coagulant mechanism, and IONPs did not affect the coagulant mechanism. The highest removal was found at leucaena extract dosage of 20 mL/L, and further increase in dosage decreases the removal due to floc re-stabilization. At variation of IONPs concentration, it could be observed that the removal and sludge volume were similar to coagulation without IONPs. Further investigation of settling kinetics showed that the increase in IONPs concentration gave shorter settling time. In addition, the settling kinetics was well fitted to pseudo-second-order kinetics. The best coagulation condition was found to be at pH 3, coagulant dosage 20 mL/L, and IONPs concentration 3.0 mg/mL extract that gave 90% Congo red removal and 30 mL/L sludge volume with half settling time of the one without IONPs (60 min).

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Compliance with ethical standards

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

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