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Submission date: 07-Aug-2024 10:46AM (UTC+0700)
Submission ID: 2428445472
File name: an_and_conventional_coagulants_for_coal_wastewater_treatment.pdf (1.04M)
Word count: 3056
Character count: 15030

IOP Conference Series: Materials Science and Engineering

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To cite this article: A Maria *et al* 2020 *IOP Conf. Ser.: Mater. Sci. Eng.* **980** 012077

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IOP Conf. Series: Materials Science and Engineering **980** (2020) 012077 doi:10.1088/1757-899X/980/1/012077

Comparing the effectiveness of chitosan and conventional coagulants for coal wastewater treatment

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Abstract. In this research, the potential of chitosan to be used as a coagulant to treat coal wastewater was investigated, in comparison to a conventional coagulant, i.e: $Al_2(SO_4)_3$ or aluminum sulfate, and Poly Aluminium Chloride (PAC). The parameters being studied were turbidity, pH, TDS, and TSS. The result of this research showed that chitosan worked as a more efficient coagulant to treat coal wastewater compared to alum and PAC, in terms of the needed dose of application. The optimum dose of chitosan was 20 mg/L that gave a 100% decrease in turbidity and TSS. On the other hand, a dose of 120 mg/L of alum was needed to have an optimum result, where the turbidity and TSS were decreased up to 100%. When PAC was used as a coagulant, the optimum dose was 120 ppm that reduced turbidity and TSS to 99.50% and 99.58%, respectively. Coagulation by chitosan, alum, and PAC were all influenced by pH, where the optimum pH for all three coagulants was within a range of neutral pH.

1. Introduction

One of the characteristics of coal wastewater released from coal processing activities is its high turbidity due to a high level of TSS. Before being released to the environment, coal industries often treat this wastewater by adding coagulants into it to reduce the turbidity. Aluminum-based coagulants, which include aluminum salts and polyaluminium chloride (PAC) are some of the most extensively-used coagulants. However, the use of aluminum-based coagulants comes with some drawbacks. Aluminum salts often yield sludge with a high level of metal concentration coming from the coagulant residual [1]. There is also a possibility of an increase in the concentration of dissolved aluminum ions in the treated water. It has been suggested that a high level of aluminum might be one of the causes of Alzheimer's disease [2]. Due to the potential negative impact on the use of conventional aluminum-based coagulants, an alternative coagulant is needed to have a more environmentally friendly treatment process on coal wastewater. In this paper, the potential of chitosan as a natural coagulant is presented. Comparison of the dose needed to have an optimum result in decreasing turbidity for between chitosan and conventional coagulants, i.e. PAC and $Al_2(SO_4)_3$ (aluminum sulfate) was investigated. The effect of pH on the effectiveness of each coagulant in decreasing turbidity and TSS level was also studied.

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2. Materials and methods

2.1. Materials

Coal wastewater samples were collected from the Kelanis site, one of the coal processing locations in South Barito, Central Kalimantan. Chemicals used were food-grade chitosan (100 mesh, MW = 50,000-80,000 Da); NaOH, HCl, CH_3COOH, $Al_2(SO_4)_3$, and PAC powder. All chemicals were reagent-grade from commercial sources (*Merck*) and used as received. To prepare a 2% (w/v) chitosan solution, 20 grams of chitosan was dissolved in 1 L CH_3COOH 2%. The solution was then labeled as (C-Chit). The other coagulants, i.e $Al_2(SO_4)_3$ and PAC were also prepared as 2% (w/v) solutions by dissolving them in distilled water. Each of the conventional coagulant solution was labeled as C-Al and C-PAC, respectively.

2.2. Experimental setup

To study the coagulation process, experiments were conducted by using a standard jar-test apparatus with six 1L-beakers. To determine the optimum dose, each beaker was filled with coal wastewater sample, and a specified amount of chitosan coagulant solution was added into it. The amount of chitosan coagulant solution added varied in each beaker. The mixture in each beaker was mixed rapidly with a 100-rpm speed for one minute, followed by a slow mixing at a 45-rpm speed for 20 minutes. The mixture was then let to settle for a 30-minute of sedimentation time. The supernatants were then withdrawn from the liquid phase for the analysis. The parameters being analyzed were turbidity, TSS level, and pH. All of the analyses were conducted in compliance with Indonesian National Standards (SNI, *Standar Nasional Indonesia*) for analysis of water and wastewater quality. The standard jar-test apparatus was also used to investigate the effect of pH on the coagulation efficiency. Coal wastewater samples were added into the beakers, and then the pH in each of the beaker was adjusted by adding 1.0 N NaOH and or 1.0 N HCl to have a set of samples with various pH, ranged from 4 to 9. Into each beaker, a specified amount of chitosan coagulant was added. The amount added was by the dose giving the most optimum result in decreasing turbidity, as obtained in the previous step. The samples were then rapidly mixed with a 100-rpm speed for one minute, followed by a flocculation process where the mixing speed was set at 45 rpm for 20 minutes, and a sedimentation time of 30 minutes. Supernatants were then withdrawn from the liquid phase. The pH, turbidity and TSS level from the supernatants were then analyzed. The same procedures to determine the optimum dose along with investigation on the effect of pH were repeated by using Al2(SO4)3 and PAC as coagulants.

3. Results and discussion

3.1. Optimum dose

In coagulation, the dose is one of the most crucial factors affecting the effectiveness of the coagulation process. While a dose that is too low would not give an optimum result, using an excessive amount of coagulant can lead to deflocculation [3]. The optimum dose can be different from one type of coagulant to another, and in many cases, the optimum dose also depends on the initial turbidity of the water being treated. Figure 1 shows the comparison between the decrease in turbidity of the coal wastewater samples when chitosan, PAC, and $Al_2(SO_4)_3$ were used as coagulants within the same dose range.

The result showed that while a 20 mg/L dose of chitosan coagulant was enough to have a 100% decrease in turbidity, samples treated by PAC and $Al_2(SO_4)_3$ practically did not show a decrease in turbidity nor TSS level. Pictures presented in figures 2-4 showed the clear difference of turbidity reduction when each of the coagulant being studied was used within the same dose range. Based on this result, the dose range of both PAC and $Al_2(SO_4)_3$was then increased until optimum results were obtained. The dose range that gave optimum results for each coagulant being investigated, in reducing turbidity and TSS levels is presented in figures 5 and 6.

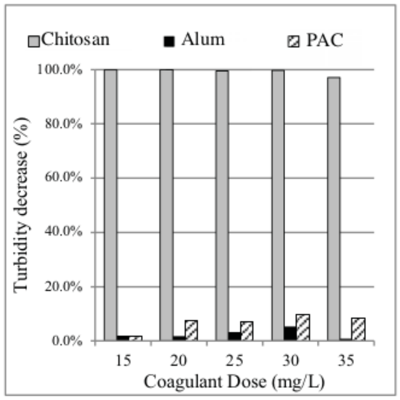

Figure 1. Turbidity decrease of coal wastewater treated by each type of coagulants.

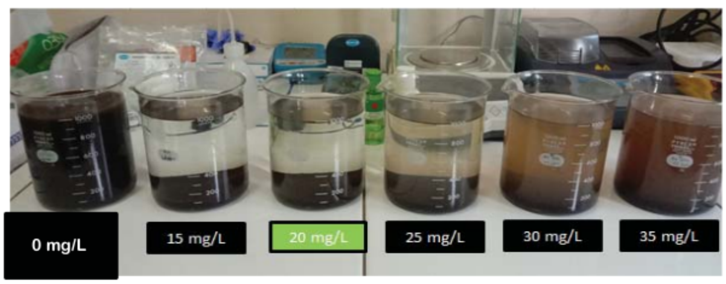

Figure 2. Coal wastewater samples after treated with chitosan as coagulant with a dose range of 0-35 mg/L.

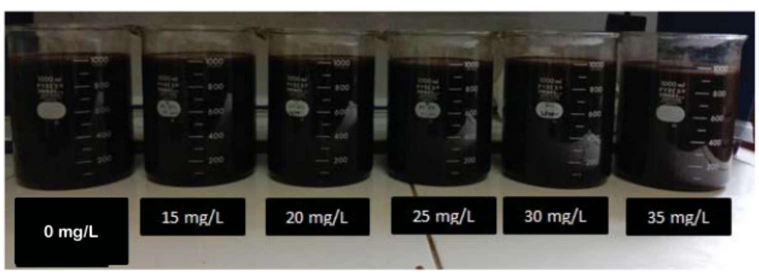

Figure 3. Coal wastewater samples after treated with $Al_2(SO_4)_3$ as coagulant with a dose range of 0-35 mg/L.

The results clearly showed that chitosan needs a much lower dose to give satisfying results, compared to both $Al_2(SO_4)_3$ and PAC. While chitosan needed a dose of 20 mg/L to have an optimum result, the optimum dose for $Al_2(SO_4)_3$ and PAC were 120 mg/L and 105 mg/L, respectively. The difference in the dose needed is likely due to the mechanisms of coagulation taking place. As coagulant-flocculant, chitosan can work through two mechanisms, i.e: charge neutralization and bridging mechanism [4, 5]. Chitosan can destabilize the colloidal particles in the suspension as chitosan is a biopolymer with polycationic properties. This is due to the presence of its amine groups that become protonated in an acidic medium. The long-chain structure of chitosan enabling it to form a bridge between the destabilized colloidal particles forms flocs that would then precipitate. In samples with a high initial TSS level, this mechanism of coagulation-flocculation seemed to work more effectively than the charge neutralization mechanism as the main mechanism of coagulation-flocculation by $Al_2(SO_4)_3$. In treatment using $Al_2(SO_4)_3$, hydrolyzed species were formed once $Al_2(SO_4)_3$ comes in contact with water that will be adsorbed by the colloidal particles [6]. However, a high dose of $Al_2(SO_4)_3$ will be needed to form flocs that are big enough to precipitate through a sweep-floc mechanism [7]. As a polymer, PAC can also work as coagulant-flocculant through the bridging mechanism. However, this mechanism can only take place in a high dose of PAC. If PAC is used in a low dose, the flocculation-coagulation can only occur through the charge-neutralization mechanism. The same pattern when chitosan was evaluated for its performance compared to conventional coagulants was observed by Ahmad *et al* [8]. They reported that chitosan needed a much lower dose (as low as 0.5 g/L) to decrease turbidity in palm oil mill effluent (POME) compared to $Al_2(SO_4)_3$ and PAC, which needed a dose of 8.0 and 6.0 g/l, respectively.

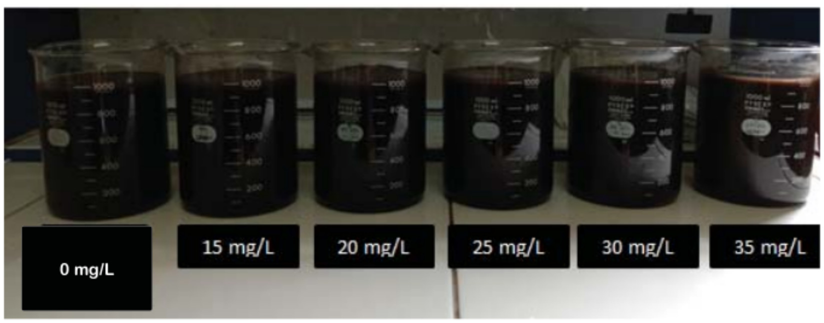

Figure 4. Coal wastewater samples after treated with PAC as coagulant with a dose range of 0-35 mg/L

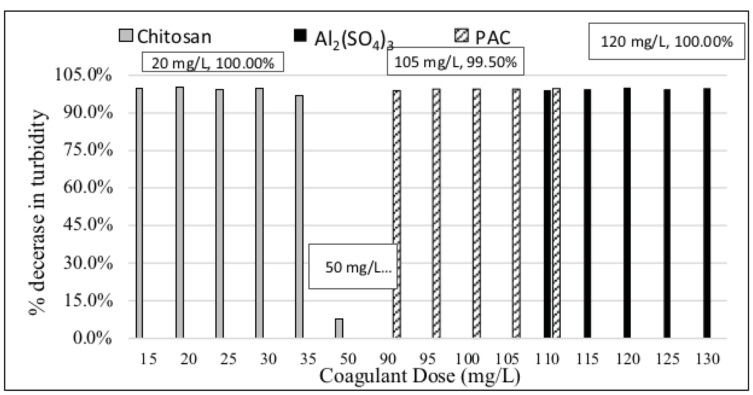

Figure 5. Optimum dose range for each type of coagulants in decreasing turbidity.

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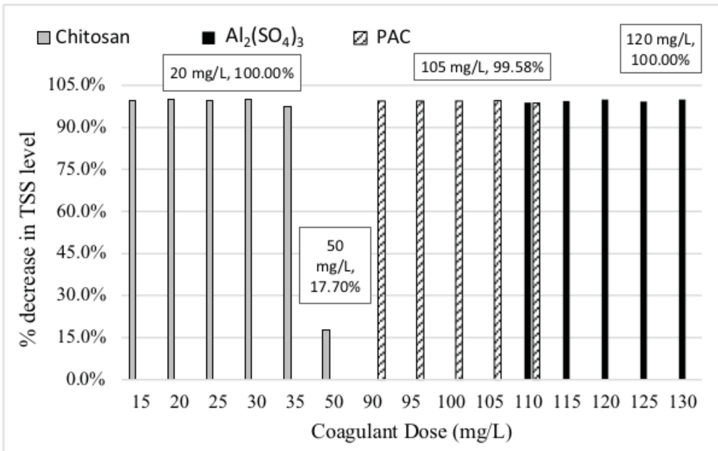

Figure 6. The optimum dose range for each type of coagulants in decreasing TSS level.

It is interesting to be noted that the performance of chitosan appeared to be very sensitive to overdosing. When the dose was increased to 50 mg/L, the decrease in turbidity and TSS value reduced significantly, indicating the occurrence of deflocculation. This is probably because of the nature of chitosan for being polycationic lead to an excess of positive charges in the system when the concentration of chitosan is above its optimum dose. As highlighted by Guibal *et al* [9], this excess of positive charge might cause the particles to be re-stabilized and breaking the formed flocs.

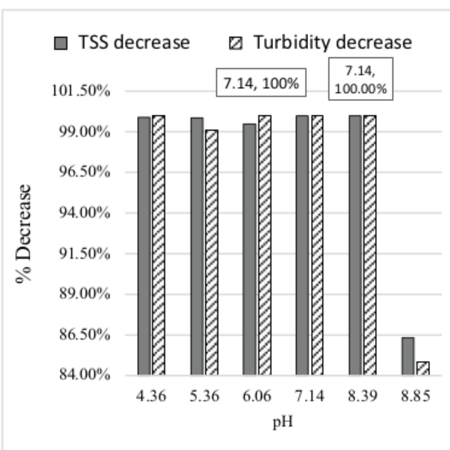

Figure 7. TSS level and turbidity decrease in treatment using chitosan at different pH.

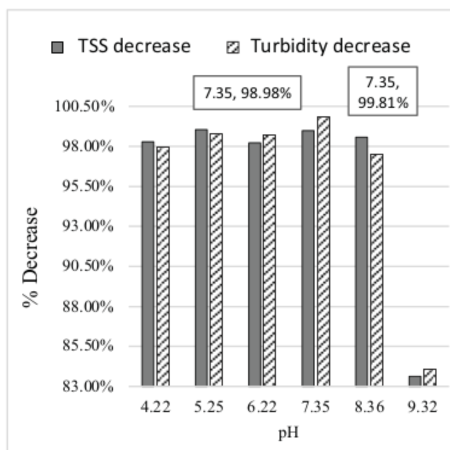

Figure 8. TSS level and turbidity decrease in treatment using $Al_2(SO_4)_3$ at different pH.

3.2. Optimum pH in coagulation

Another important factor in a coagulation process that has been well documented is pH [10]. In coagulation, pH will determine the presence of the protonated form of the coagulants. The figures below present how pH affected the performance of each coagulant in treating coal wastewater. Figures 7, 8, and 9 each show the decrease in turbidity and TSS levels when coal wastewater was treated using chitosan, $Al_2(SO_4)_3$, and PAC, respectively.

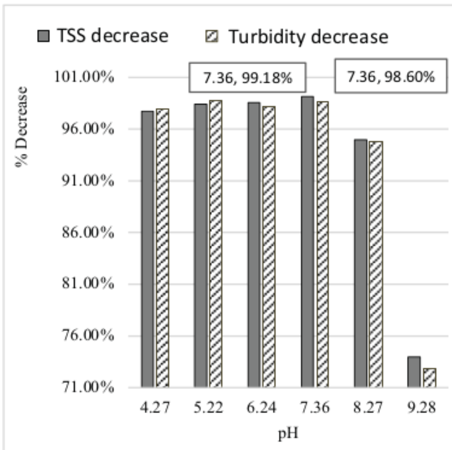

Figure 9. TSS level and turbidity decrease in treatment using PAC at different pH

From all of those figures, it can be observed that all of the coagulants work in a pH range where the system was acidic to a slightly alkaline condition. Once the pH of the system went to a more alkaline condition, the performance of all coagulants dropped significantly, as shown by a steep decline in the decrease in turbidity and TSS level. For all the coagulants, an optimum result was obtained when the pH is within a neutral range. This result is in accordance with some other studies that also showed a preferable condition for optimum coagulation was within a neutral pH range [9,11,12]. Particularly for chitosan, an acidic condition will ensure that the amine groups are protonated, enabling efficient charge neutralization in the coagulation process [4,13,14].

4. Conclusion

The result of this study has demonstrated that chitosan is a strong potential to be used as a natural coagulant as an alternative to conventional coagulant in treating coal wastewater. Chitosan worked more efficiently as it needed a much lower dose than $Al_2(SO_4)_3$ and PAC to give the result in decreasing turbidity and TSS levels in the coal wastewater samples. However, more research is still needed to explore the feasibility of using chitosan on a much larger scale than the laboratory scale that has been conducted in this study.

Acknowledgments

We would like to thank PT Adaro Indonesia who had generously allowed us to conduct this research in their facility.

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