Water Treatment Sludge as Coagulant and Adsorbent: A Recent Review

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Abstract The use of metal coagulants in water treatment generates huge amounts of sludge. Effective management to mitigate the challenges associated with the increasing amount of WTS remains a significant concern. This paper provides an overview of the beneficial reuses of water treatment sludge (WTS) as coagulant and adsorbent. Recent advancements in coagulant and adsorbent synthesizing techniques are discussed. Recovered coagulant (RC) and adsorbent have been successfully removed several pollutants in both water and wastewater, reducing the need for new coagulant production and disposal of sludge. Coagulant and adsorbent from sludge have potential to be utilized in both water and wastewater treatment processes and, hence promoting sustainability. Sulfuric acid is commonly used to recover contaminated coagulant. Calcination, activation, or impregnation have been studied in adsorbent production. Recovered coagulant/adsorbent could be as effective as or even better than fresh coagulants/adsorbent.

Keywords Water treatment sludge · Acidification · Heavy metal · Impregnation · Circular economy

1 Introduction

The coagulation–flocculation process plays an extremely vital role in water and wastewater treatment due to its low capital cost and easy operation [5, 21]. The use of these chemicals can result in the generation of sludge as a byproduct, which contains contaminants such as organic matter, heavy metals, and pathogens. It is crucial to properly manage this sludge to prevent environmental issues and health risks to public community. However, disposal of water treatment sludge (WTS) in the environment has been restricted for several reasons, including reduction in access

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to landfill site, expensive disposal prices, high transportation costs, and regulatory constraints. These issues have led to the exploration of potential reused of the WTS in various application.

Several studies have investigated the effectiveness of WTS as a coagulant and adsorbent for various contaminants and under different treatment conditions. These studies have shown promising results, indicating that sludge can be an effective coagulant and adsorbent for a wide range of contaminants [11–12] and this paper aimed to expand the knowledge through latest publications.

2 WTS as Coagulant

Acid recovery method widely used for coagulant recovery from WTS because of its high efficiency with superior quality [6]. Among different chemicals used, sulfuric acid showed better results from both practical and cost viewpoints [19]. This can be observed in Table 1 that most study using sulfuric acid for their recovery method. There were also other types of acid used such as nitric acid [3–4] and hydrochloric acid [10].

Water treatment sludge recovery using acidification methods reported to efficiently remove pollutants in wastewater. Mora-León et al. [10] compared the effectiveness of recovered polyaluminum chloride (PAC) coagulant using sulfuric and hydrochloric acid with that of commercial PAC and ferric coagulant. The authors found that the RC showed better turbidity removal of up to 96%, and at the optimal dose of RC, 89% total suspended solid (TSS), 62% total chemical oxygen demand (tCOD), 90% total phosphorus (tP), and 97% soluble phosphorus (sP) were removed from the wastewater. However, Chakraborty et al. [2] found that fresh coagulant outperformed recovered ferric-based coagulant, with RC showing a decrease in TSS, COD, and total nitrogen (tN) removal efficiency and a corresponding 10% increase in these pollutant's concentration in the treated wastewater compared to fresh coagulant; might be due to lower ionic strength of RC.

In comparison, there are studies that directly use WTS to treat wastewater without any recovery method. Kang et al. [7] studied the use of aluminum-based WTS as a substitute for conventional chemicals in animal farm wastewater treatment and found that the removal of TSS, PO_4^{3-}, and total organic carbon (TOC) was 87.8%, 96.9%, and 62.1%, respectively. Khedher et al. [8] studied the use of WTS as a coagulant aid to improve the dissolve organic matter (DOC) in natural surface water. The researchers found that the addition of WTS at a concentration of 3 g/L can reduce the optimum dose of fresh alum sulfate required up to 50% to achieve similar removal efficiency of DOC (70%). They also found that the addition of WTS reduced the sludge produced by approximately 50% compared to when it was not.

Several studies have also utilized sludge from water treatment plants to treat raw water, employing an acidification method to recover the coagulant from the sludge before use. Hamzah et al. [4] investigated the percentage recovery of aluminum-based coagulant by nitric acid and its performance in removing turbidity from raw water.

Table 1 Recovered coagulant performance various pollutant removal

Origin of sludge	Synthesizing method	Coagulant synthesized	Type of water treated	Coagulation condition	Removal performance	References
–	Acidification sulfuric acid	Alum–based coagulant	Raw water	Dosage 25 ppm pH 2.5	Turbidity 93.28%	Ruziqna et al. [14]
Ontario, Canada	Acidification sulfuric acid	Ferric–based coagulant	CEPT effluent	Dosage 40 mg/L pH 1.5	TSS 78% COD 63% tP 42% sP 29% tN 16%	Chakraborty et al. [2]
Sungai petani, Kedah	Acidification nitric acid	Aluminum sulfate	River water	Dosage 2 mg/L pH 7	Turbidity 99.47%	Hamzah et al. [4]
Dublin, Ireland	–	Aluminum–based coagulant	Animal farm wastewater	Dosage 1588 mg/L pH 7	TSS 97.8% (PO4)-3 96.9% TOC 62.1%	Kang et al. [7]
–	Acidification sulfuric and hystochloric acid	Polyaluminum chloride	Domestic wastewater	Dosage 40 mg/L	Turbidity 96% TSS 89% TCOD 62% tP 90% sP 97%	Mora-León et al. [10]
Nagpur, India	Acidification nitric acid	Polyaluminum chloride	Raw water	Dosage 1 ml/L	Turbidity 74%	Dahasahastra et al. [3]
South Australia	–	Aluminum sulfate	River water	Dosage 3 g/L pH 6	DOC 70%	Khedher et al. [8]

The study found that at the optimal dosage, the RC can remove the raw water turbidity up to 99.47%, which is better or comparable to the fresh coagulant. Dahasahastra et al. [3] performed the same recovery method and showed that RC has potential for use as a substitute for commercial alum in water treatment. The finding was that 1 mL/L of RC has a similar removal of turbidity efficiency (74%) to 0.6 mL/L of 1% (w/v) commercial alum solution. The same result was found by Ruziqna et al. [14], where slightly higher doses of RC were required to achieve the quality of pure coagulant in removing turbidity in raw water. They found that 25 ppm of recovered coagulant achieved similar reduction of turbidity of pure alum at 93.26%.

3 WTS as Adsorbent

Although activated carbon is a popular adsorbent for water and wastewater treatment, it can be expensive due to operation and regeneration costs, as reported by Azreen and Zahrim [1]. Thus, many studies have explored the use of adsorbents synthesized from waste materials like WTS as an effective and affordable solution. Among the various synthesis methods available, physicochemical methods have emerged as the most commonly used approach due to their effectiveness and ease of synthesis; needing only heat treatment. Methods such as calcination, activation, or impregnation have been studied to modify the properties of the WTS and create adsorbents with high adsorption capacity. Studies by Shahin et al. [16] and Truong and Kim [18] have demonstrated the high recovery efficiency and good quality of adsorbents synthesized using calcination and pyrolysis. Other synthesizing method also include chemical and physicochemical activation such as impregnation of $AlCl_3$ + Starch and calcination + H_3PO_4/KOH, respectively [9, 20]. These adsorbents have been found to effectively remove heavy metals, dyes, and organic pollutants from wastewater.

WTS has shown great potential as an effective and low-cost adsorbent for various pollutants. Table 2 summarizes some recent studies on the use of WTS as an adsorbent for contaminants in various solutions. The type of pollutant influences the adsorption capacity and removal performance of the adsorbent. Shahin et al. [16] used calcined powder adsorbent synthesized through physical aerobic calcination to remove copper. They obtained a high adsorption capacity of 35 mg/g and a removal performance of 90%. Separate studies found that phosphate achieve removal performance of 86–99% [18, 20], and endocrine disruptors achieving almost 100% [19].

It is worth noting that the adsorption capacity and removal performance of WTS as an adsorbent are generally comparable to or even higher than those of other commercial adsorbents. Zeng et al. [22] compared the adsorption capacity of granular adsorbent synthesized through physical methods with chitosan solution to that of commercial activated carbon and raw sludge for arsenic removal. They found that their adsorbent is able to remove As (V) at 14.95 mg/g adsorption capacity and solves the concern on the application in fixed beds system and the recovery and reuse of adsorbents resulting in lower operation cost. Similarly, Siswoyo et al. [17] compared the adsorption capacity of alum sludge to that of commercial activated carbon for heavy metal removal. They found that the adsorption capacity of alum sludge was comparable to that of commercial activated carbon.

The use of WTS as adsorbents for heavy metal removal has gained increasing attention due to their cost-effectiveness, eco-friendliness, and mainly high removal rate. Studies have shown that the effectiveness of WTS as an adsorbent for heavy metal at optimized condition where a study conducted by Siswoyo et al. [17] showed that WTS can achieve a removal rate of up to 95% for Cd. Furthermore, the removal performance of WTS for copper (Cu) and arsenic (As) achieved 90% and 85%, respectively [17, 22].

WTS as adsorbent has the potential to be utilized in water and wastewater treatment process in removal of pollutants. Further studies on hybridization of adsorbent should also be carried out. For instance, a study by Safie and Zahrim [15], where they studied on the combination of adsorbents to produce higher adsorption capacity which includes zeolites, chitosan, and biochar, and in line with this study, WTS-based adsorbent can be hybridized to achieve higher adsorption capacity.

Table 2 WTS as adsorbent for pollutant removal

Origin of sludge	Synthesizing method	Absorbent synthesized	Adsorption condition	Adsorption capacity (mg/g)	Removal performance (%)	References
PDAM Tirta Binangun, Yogyakarta, Indonesia	Physicochemical; sludge encapsulation	Alum raw, sludge powder Powder activated sludge (H_3PO_4), PAS alginate, PAS agar	Dose: 0.4 g/L pH: 7 CT (RSP, PAS): 30 min CT (PAS-AG/AR): 6 h	RSP: 40.26 PAS: 24.95 PAS–AG: 29.86 PAS–AR: 19.81	RSP: 91 PAS: 95 PAS–AG: 90 PAS–AR: 90	Siswoyo et al. [17]
Nanjing, China	Chemical ($AlCl_3$ + Starch)	PACS granular adsorbent	pH: 4	1.78	P: 86.06	Wu et al. [20]
Zagazig, Egypt	Physical (Aerobic calcination, 500 °C)	Calcined powder adsorbent	pH: 6.6 T: 80 °C	35.0	Cu^{2+}: 90	Shahin et al. [16]
Songbei, Harbin, China	Physical (mixture with chitosan solution)	Granular adsorbent	Dose: 1.8 g/L pH: 6.5	14.95	Ar(V): 85	Zeng et al. [22]
Brazil	Physicochemical (550 °C); (H_3PO_4 and KOH)	PAC modified sludge (PMS) Phosphoric acid sludge (PAS) Potassium hydroxide sludge (PHS)	Dose: 0.5 g/L pH: 5.5	Estradiol (E2): PMS: 8.748, PAS: 16.42, PHS: 17.903 Ethinylestradiol (EE2): PMS: 14.557, PAS: 4.233, PHS: 0.438	Estradiol (E2): PMS: 99.75, PAS: 99.96, PHS: 99.99 Ethinylestradiol (EE2): PMS: 99.99, PAS: 99.80, PHS: 99.75	Martins et al. [9]
Chuncheon City, Korea	Physical (Pyrolysis, 700 °C)	Pyrolyzed alum sludge	pH: 4–6	34.53	Phosphate: 99.0	Truong and Kim [18]
Ho Chi Minh City, Vietnam	Chemical (Precipitation)	DWS500@ZrO_2 ($Fe_2(SO_4)_3.nH_2O$	pH: 2	30.99	NO_3^-: 98.97	Phan Quang et al. [13]

4 Conclusion

The use of RC and sludge adsorbent shows the potential to provide a cost-effective and sustainable solution for water treatment. The acid sulfuric recovery process is widely used due to its high recovery efficiency and good quality. Most studies show that recovered coagulant is as effective as or even better than commercial coagulants, but at a higher dose. The recovered coagulants have been applied in removing pollutants in water treatment, especially in treating wastewater, where it efficiently removes contaminants such as turbidity, TSS, tP, and sP up to 90%. Physical methods have emerged as the most commonly used approach to synthesize adsorbents with high adsorption capacity. Most studies also show that the removal pollutant in wastewater using WTS adsorbent up to more than 90%. Further research should explore the economic potential of these materials in reducing the total operational cost of water and wastewater treatment processes. Multiple-objective optimization on the basis of ratio analysis could be employed to discretely measure multiple response characteristics of various coagulant and adsorbent as a function of assessment value.

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