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Composite Coagulant (PACSPP) Ability to Remove Ammonia and Colour from Stabilised Leachate Under Ratio, pH and Dosage Influence

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Abstract: Along with chemical coagulants, coagulation-flocculation is one of the applications that works well for stable leachate treatment. Polyaluminium chloride (PAC) and sweet potato peels (SPP) were operated as a composite coagulant (PACSPP) to investigate the efficiency of removing colour and ammonia from Simpang Renggam landfill's leachate. From the analysis of the results, the optimum ratio of composite coagulant was PACSPP(b), with a 4:6 ratio. The optimum dose and pH for composite coagulant of PACSPP(b) was 5,000 mg/L at pH 6, with removal percentages of colour and ammonia being 92% and 66%, respectively. The removal percentage of colour and ammonia of composite coagulant is at par but with less chemical coagulant. These results show that a composite coagulant made up of PAC and natural coagulant of SPP combined the best qualities of both coagulants and improved the coagulation effectiveness.

Keywords: coagulation-flocculation, PAC, composite coagulant PACSPP

1. Introduction

In recent decades, fast global and commercial development has contributed to the growing development of municipal solid waste (MSW). Food waste management for 41% of the total weight of waste components in 2020, according to SWCorp Federal Territories, followed by diapers (23.7%), plastic (19.3%) and paper (7.8%) in urban areas such as Kuala Lumpur [1]. According to a study by Zulkifli et al. [2], more than one million tonnes of MSW are produced per day in Asia, with 1.8 million tonnes projected by 2025. Landfilling is the safest practice for disposing of solid waste. There are three ways of landfilling, i.e., sanitary, unsanitary landfills, and open dumping. Due to unplanned construction, conducting landfills will eventually develop poisonous gases and leachates that can avoid containment and make their way to groundwater and soil [3]. The leachate production will continue even when the landfill shuts down [4]. Leachate is black or brown and contains many organic, non-organic and heavy metals. Leachate is usually divided into three levels, i.e., leachate less than 5 years old (young), 5-10 years (medium) and more than 10 years (mature) [5]. Organic and inorganic pollutants, heavy metals and potentially dangerous chemicals harmful to living creatures and ecosystems, such as ammonia, may be found in leachate [6],[7]. High ammonia nitrogen levels may harm aquatic life in the long run [8],[9]. According to the categorisation and characterisation of leachate, landfill operators

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have commissioned various biological, physical, and chemical treatments. Physical and chemical treatments are preferred for stabilised leachate, and these techniques have been developed throughout the years, such as coagulation-flocculation [10],[11]. Coagulation-flocculation is more effective in handling intermediate and stable or matured leachate than young leachate [12].

Coagulation destabilises a colloidal suspension [13]. Flocculation is how destabilised particles are propagated to gather and agglomerate into larger flocs [14]. Charge neutralisation is achieved using coagulants [15]. Coagulants destabilise negatively charged colloids, allowing fine particles and colloids to agglomerate into large particles, eliminating turbidity, natural organic matter, and other soluble organic and inorganic contaminants in wastewater [15]. Coagulation-flocculation has long been a prevalent method, with polyaluminium chloride (PAC), alum and polyaluminium silicate chloride as the typical coagulants [16]. The formation of PAC precipitates significantly improved coagulation performance, effectively removing suspended matter, dissolved organic matter, turbidity, COD, colour and humic acid [13]. PAC is a typical polymeric inorganic salt that can coagulate more than alum at lower temperatures and produce less sludge. However, PAC has several drawbacks, including comparatively high procurement costs, negative impacts on human health and the atmosphere, and high sludge production [17]. Disadvantages of using PAC in leachate treatment are higher cost, health risks to humans and other living organisms (Alzheimer’s disease and lung cancer), and hazardous sludge [4],[18],[19],[20],[21],[22]. Natural coagulants are commonly studied and used as next-potential coagulants due to the detrimental disadvantages of chemical coagulants [4]. Natural materials are also readily available, cost-effective, and come from sustainable sources, rendering coagulants from natural materials an extra persuasive argument to create [23]. Natural coagulants, such as plant extracts, are plentiful, safe for human health, and toxic-free in general. This research uses sweet potato peels (SPP) as a natural coagulant. SPP is not widely used in leachate treatment. Sweet potato (*Ipomoea batatas* L.) is a tropical food plant belonging to the morning glory family (Convolvulaceae). Based on a previous study, orange sweet potato has the highest amylose content, which helps starch’s functional properties, such as clarity and swelling will deteriorate as amylose content increases [24].

Sweet potato is categorised as plant-based and similar to tapioca [25]. Tapioca starch contains amylose and is made up of long, essentially unbranched chains that tangle violently when released, making it a very efficient thickening that traps huge volumes of liquid inside its tangled chains [25]. The increasing need for efficient and sustainable materials in the wastewater treatment sector has prompted the creation of composite made up of various possible components, such as organic and natural polymers [26]. Premixing two coagulants form a composite coagulant into a single reagent under specific working conditions. However, based on the literature study, a composite coagulant consisting of PAC and SPP has never been used in the coagulation-flocculation process as a stabilised leachate treatment. Therefore, this study aims to determine the efficiency of PACSPP coagulation in removing ammonia and colour from leachate under the influence of different PACSPP ratios, doses and leachate pH.

2. Materials and Method

2.1 Landfill Leachate

The leachate sample used in this study was taken from the Simpang Renggam landfill located at Simpang Renggam, Kluang, Johor. The characterisation tests were done to classify the leachate during the preliminary study, and the sample was taken twice a month for three months, from October to December 2021. The sample was transported to the Wastewater Laboratory, Faculty of Civil Engineering and Built Environment (FKAAB), Universiti Tun Hussein Onn Malaysia (UTHM), and stored in a cool room of 4ºC. All samples were collected and stored according to Water and Wastewater APHA standard (2005). table 1 shows the characterisation data of raw leachate used for this study. 500 mL of leachate was used in the study for the coagulation process during jar tests [27]. Based on the studied leachate characteristics, the sample leachate used is old leachate. However, the results did not comply with the leachate standard Environmental Quality Act (EQA) Regulation 2009. Therefore, the coagulation-flocculation method is used as an efficient method for leachate treatment.

Table 1 - The characteristics of Simpang Renggam landfill

Parameter	Range	EQA Regulation 2009
Temperature	**22.83**	**<40**
pH	**8.1**	**6.0 – 9.0**
COD (mg/L)	**706.30**	**400**
Ammonia (mg/L)	**1,218.10**	**5**
SS (mg/L)	**82.92**	**50**
Turbidity (NTU)	**23.04**	**-**
Colour (ADMI)	**2,069.13**	**-**

No. of sample: Nine samples for three months

2.2 Coagulation Process

The coagulation process was performed in a series of jar tests using VELP-Scientific, Model JLT6 (Italy), according to ASTM D2035 standard procedure. This method used six paddle rotors, and 500 mL of leachate samples were put in 1-L beakers for the test [25]. It was rapidly mixed at 200 rpm for 4 minutes. Then, slowly mixed at 30 rpm for 15 minutes before settling for 30 minutes [25],[27]. After that, a plastic syringe was used to collect the supernatant three centimetres from the surface for analytical analysis.

2.2.1 Composite Coagulant Preparation (PACSPP)

The composite coagulant was prepared by injecting 200 mL of SPP amount of a range of PAC using a peristaltic pump and magnetic stirring [27]. The peristaltic pump adjusted the flow rate to 2 mL/min with a water bath temperature of 65 – 70ºC. There were six PAC:SPP ratios used in this study (i.e., 10:0, 2:8, 4:6, 6:4, 8:2 and 0:10). The selected dose range in the composite coagulant process was from 1,000 mg/L to 5,000 mg/L, and the pH range was from 3 to 10. The optimum conditions for composite coagulants are based on the highest removal of ammonia and colour. The analysis was done immediately to prevent any changes in the treated samples

2.2.2 Single Coagulant Preparation (PAC & SPP)

The single coagulant preparation started by weighing 10 g of PAC powder and diluting it into 100 mL of distilled water [25]. For the SPP coagulant, 10 g of SPP was weighed in 100 mL of distilled water for dilution. The dose range for PAC single coagulant was from 500 mg/L to 4,000 mg/L, while for SPP single coagulant was in the range of 50 mg/L to 500 mg/L [4],[31]. Both single coagulants at various pHs, 3 to 10, were applied. The optimum dose and pH obtained from a single coagulant were used for the optimisation dose, and pH for dual coagulant (PAC + SPP).

2.2.3 Dual Coagulant Preparation (PAC + SPP)

For dual coagulants, the same method was used for stock solution preparation from a single coagulant (10 g of PAC or SPP was diluted into 100 mL of distilled water). PAC solutions become the primary coagulant for dual coagulation that is added at rapid mixing, while SPP is an addition to support the PAC to ensure the efficiency of the treatment [25]. Firstly, the optimum dose from a single PAC is used as a fixed dose of PAC with various doses of SPP in the range of 50 to 500 mg/L. Then, fixed doses of PAC and SPP were tested with various pH ranges of 3 to 10. Thus, the removal percentage of single coagulant, dual coagulant, and composite coagulant were compared.

2.3 Analysis Method

The removal parameters were analysed according to Standard Methods in table 2. The samples were tested at room temperature, with the findings based on a triplicate analysis. All respective parameters were tested using HACH DR6000TM UV-VIS Spectrophotometer (USA).

Table 2 - The laboratory test parameters and list of Standard Methods used [28]

Table	Standard Method
Ammonia	Conductrimetric (APHA Method: 4500-N) & (HACH Method: 8038)
Colour	Spectrophotometric (APHA Method: 2120) & (HACH Method: 8025)

3. Results and Discussion

3.1 PAC Optimisation as a Single Coagulant

In preliminary research, PAC was analysed at various dosages to determine the ideal condition for this chemical coagulant to accomplish excellent removal of the colour and ammonia parameters. The PAC single coagulant dose ranged from 500 to 4,000 mg/L at pH 7. figure 1 shows the removal percentage of PAC single coagulant in ammonia and colour removal. Based on the graph, the highest removal percentage for colour and ammonia was at the dose of 3,000 mg/L PAC, with 89% and 40%, respectively. Thus, the PAC dose at 3,000 mg/L was selected as the optimum dose for a single PAC due to the higher removal percentage recorded. Next, the experiment on various pH values of the sample was carried out between 3 to 10. figure 2 shows the results of the experiment. Based on the graph, the removal percentage showed the highest removal at pH 6 for colour (92%) and ammonia (41%). Hence, the optimum pH of PAC single coagulant was selected at pH 6 due to the higher removal percentage of colour and ammonia.

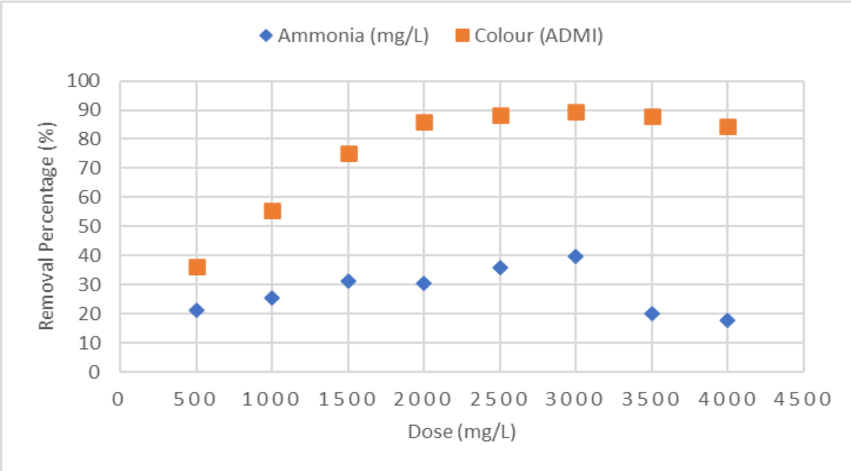

Fig. 1 - Optimum dose of PAC at pH 7

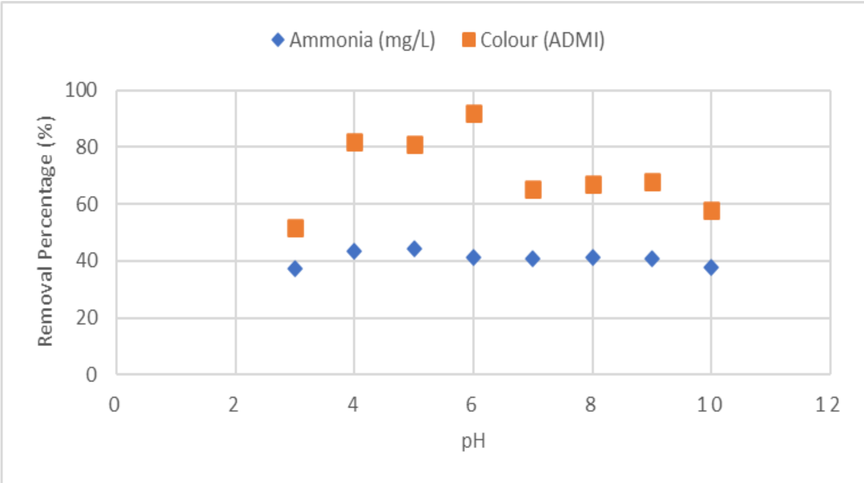

Fig. 2 - Optimum pH of PAC at 3,000 mg/L

A study by Mohd-Salleh et al. [27] for PAC as a single coagulant shows the optimum dosage at 3,750 mg/L and pH 6 of leachate (Table 4) with 98% (colour) and 28% (ammonia) removal percentages. When comparing the results from this study with the study by Mohd-Salleh et al. [27], the removal percentage of colour for this study was at par, but the removal percentage of ammonia shows increasing from 28% to 44%. An investigation by Aziz et al. [15] showed that PAC at a 5,000 mg/L dose at pH 6 could remove 90% of colour. In contrast, this study found that the optimum dose and leachate pH for PAC as a single coagulant were at 3,000 mg/L at pH 6 and could remove 90% of colour. Furthermore, when the coagulant dose was raised, the proportion of parameters removed increased until an optimal value was reached, and PAC achieved 90% colour removal but low ammonia degradation [28].

3.2 SPP Optimisation as a Single Coagulant

The results of the removal percentage of SPP as a single coagulant at pH 7 are shown in figure 3. The highest removal percentage of colour and ammonia was shown at 250 mg/L dose with 4% and 6%, respectively. Based on figure 4, the highest removal of ammonia is at pH 8 (5%), while the highest removal of colour is at pH 3 (8%) compared to pH 8 (4%). Thus, pH 8 was selected as the optimum pH for SPP as a single coagulant due to the highest removal percentage of ammonia and less reagent used in adjusting pH. It was selected because pH 3 is not practical for on-site application due to the high chemical volume necessary to alter pH.

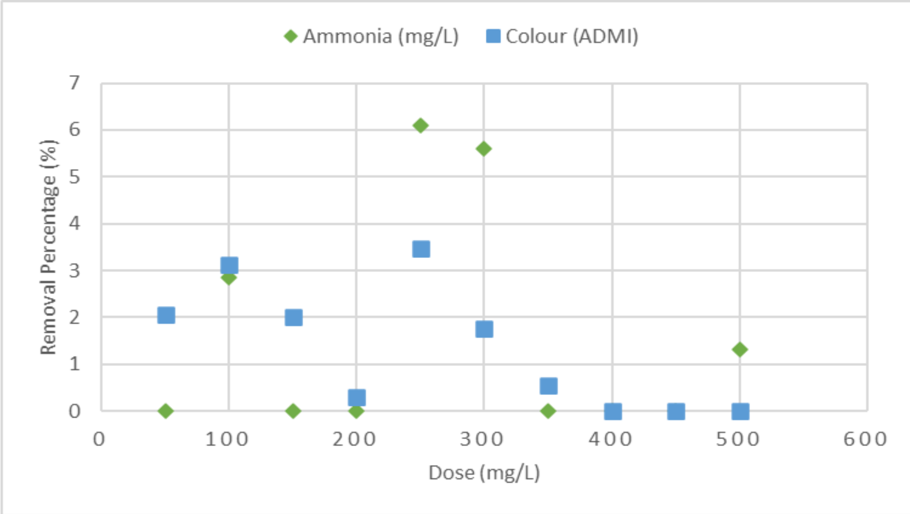

Fig. 3 - Optimum dose of SPP at pH 7

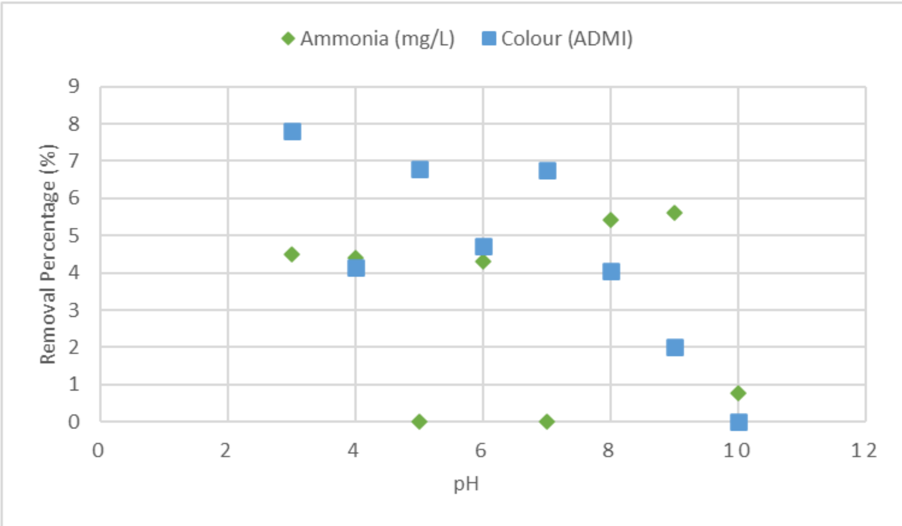

Fig. 4 - Optimum pH of SPP at 250 mg/L

Based on the previous study by Azizan [25], a single coagulant using tapioca starch was able to remove 30% of colour and 38% of ammonia at 2.5 g/L dose with pH 8 leachate (Table 4). Compared with this study, SPP efficiency (colour: 4% and ammonia: 5%) shows lower results than tapioca starch. Comparing the optimum pH of this study with the Azizan [25] shows that both studies have a similar optimisation pH at pH 8. Physical observation shows that SPP produces low sludge as a single coagulant. Based on Mohd-Zin, et al. [16], tapioca flour as a natural coagulant shows that sludge production was lower than chemical coagulants. This is because starch ions, unlike chemical coagulants, were not strongly positive enough to interact with negatively charged particles in leachate [16].

3.3 PAC and SPP Optimisation as a Dual Coagulant

The experiment was performed with a range of doses of SPP at 50 to 500 mg/L and fixed-dose PAC at 3,000 mg/L (optimum single coagulant dosage), as shown in figure 5. Based on the graph, at a 250 mg/L dose of SPP, the dual coagulant removed 95% of colour and 39% of ammonia. Then, the experiment continued with various pHs of the samples and a fixed dose of PAC at 3,000 mg/L and SPP at 250 mg/L. figure 6 shows the removal percentage for dual coagulant at a fixed dose of PAC at 3,000 mg/L and a fixed dose of SPP at 250 mg/L with various pH values. The highest removal percentage of colour occurred at pH 8 (93%). Meanwhile, the highest removal percentage of ammonia was 5 (35%). Therefore, the optimum pH for dual coagulant was selected at pH 8 since the difference between the removal percentage of ammonia at pH 8 and pH 5 is 3%. The optimum pH of 8 was selected due to the high removal percentage of colour, and it is nearer to raw leachate pH. Next, the experiment continued with the varied dose of PAC in the range of 500 mg/L to 4,000 mg/L at pH 8, and the SPP dose was 250 mg/L. Based on figure 7, the highest removal percentage of colour shows at 2,500 mg/L with 94%. Meanwhile, a 2,000 mg/L dose is able to remove 93% of colour. The difference in removal percentage at 2,000 and 2,500 mg/L is 2%. However, the highest removal percentage of ammonia was at a 3,000 mg/L dose with 41%. Here, the difference in the removal percentage of ammonia for doses 2,000 mg/L and 3,000 mg/L is 4%. Thus, 2,000 mg/L was selected as the optimum PAC dose of dual coagulant due to the lower dosage needed.

Based on a study by Mohd-Zin, et al. [29], a dual coagulant (alum 3 g/L + barley 0.8 g/L) consisting of alum as a chemical coagulant and barley as a natural coagulant has successfully removed 98% of colour from Simpang Renggam landfill leachate (Table 4). The study by Mohd-Zin, et al. [29] showed a higher removal percentage of colour compared to 3.0 g/L of alum coagulant, probably due to the addition of barley may minimise the amount of alum needed while also improving coagulation. Compared to this study using PAC as a chemical coagulant in the dual coagulant (PAC: 2,000 mg/L + SPP: 250 mg/L) with Mohd-Zin, et al. [29], PAC used low dosage alum. PAC is more efficient than alum because lesser dosages are used [30]. According to a study by Mohd-Salleh et al. [31], they used *Manihot esculenta* peel extract (MEP) as a natural coagulant and PAC as a chemical coagulant in the leachate treatment that undergoes coagulation-flocculation process at dose 250 mg/L and 3,500 mg/L through dual coagulant at pH 7. The removal percentage shows that dual coagulants studied by Mohd-Salleh et al. [31] could remove 96.6% of colour and 33.8% of ammonia. Compare the study by Mohd-Salleh et al. [31] with this study; dual coagulant can remove 93% of colour and 37% of ammonia. The removal percentage of colour shows a slight decrease from Mohd-Salleh et al. [31], with a difference of 4%, but the removal of ammonia shows an increment of up to 3%. This shows that SPP was more efficient in removing ammonia than MEP.

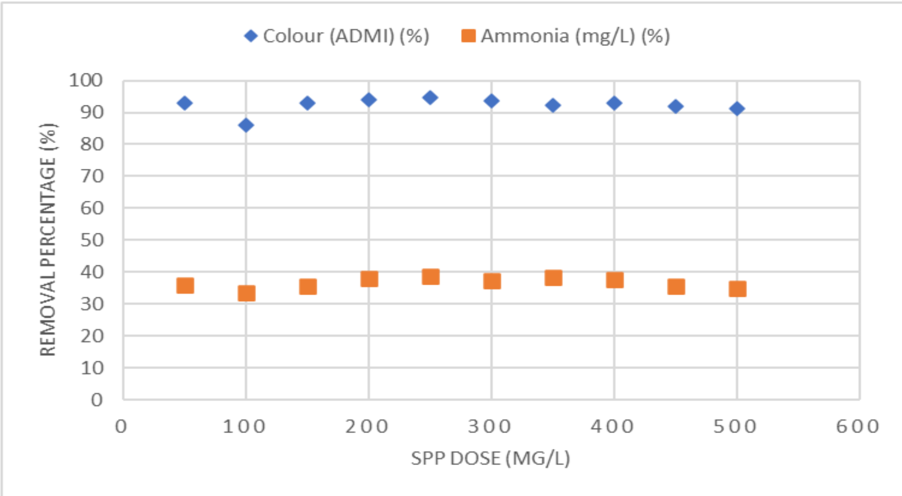

Fig. 5 - Optimum dose of SPP dual coagulant and fixed PAC dose (3,000 mg/L) at pH 7

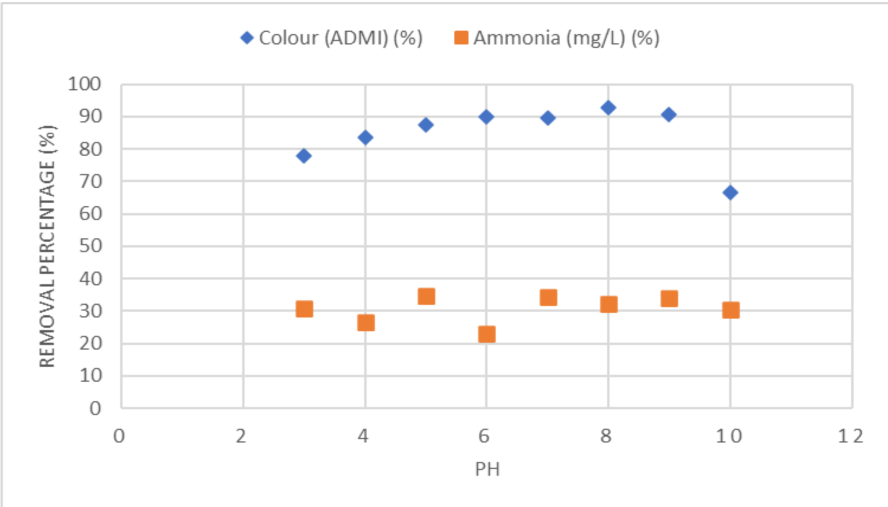

Fig. 6 - Optimum pH of dual coagulant with fixed PAC dose (3,000 mg/L) and SPP dose 250 mg/L

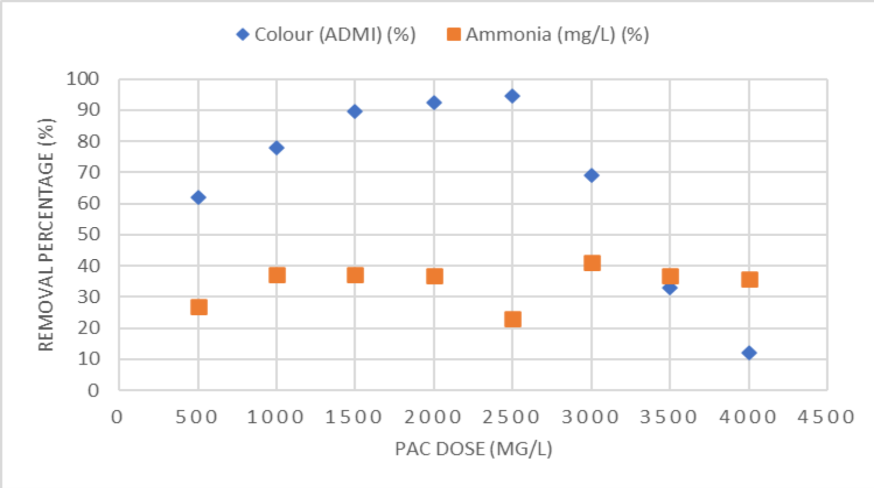

Fig. 7 - Optimum PAC dose of dual coagulant with fixed SPP dose (250 mg/L) at pH 8

3.4 Composite Coagulant (PACSPP) Performance Under the Influence of Different Ratios, pH and Doses

Composite coagulant has four ratios labelled as PACSPP(a), PACSPP(b), PACSPP(c) and PACSPP(d) with 2:8, 4:6, 6:4 and 8:2, respectively. table 3 shows the composite coagulant PACSPP with the optimum dose and pH. The removal percentage of colour and ammonia for PACSPP(a) shows 75% and 44% at 4,000 mg/L doses at pH 7. Composite coagulant PACSPP(b) removed 92% of colour and 66% of ammonia at 5,000 mg/L dose at pH 6. The data showed that PACSPP(b) was more effective in removing colour and ammonia. Compared with PACSPP(c), 3,000 mg/L dose at pH 7 removed 95% of colour and 49% ammonia. Composite coagulant PACSPP(c) is able to remove colour 3% from PACSPP(b), but PACSPP(c) is not able to remove ammonia efficiently as PACSPP(b) with a 17% difference. Based on their concentration of PAC between PACSPP(b) and PACSPP(c), PACSPP(b) with 0.04 g/mL concentration is lower than PACSPP(c) with 0.06 g/mL

Thus, PACSPP(b) seemed to be better than PACSPP(c) due to the lower dosage of PAC used, and this study's objective is to use less PAC dosage for leachate treatment. Next, PACSPP(b) was compared with PACSPP(d) at 4,000 mg/L dose and pH 7 with 93% of colour and 57% ammonia removed. PACSPP(d) shows a slight difference in removing colour compared to PACSPP(b), with 93% to 92%. In removing ammonia, PACSPP(b) shows more efficiency than PACSPP(d), with 66% than 57%. Moreover, the concentration of PAC in PACSPP(d) is higher, with a value of 0.08 g/mL, compared to PACSPP(b), which is 0.04 g/mL. The amount of Al contained in PACSPP(d) also shows higher concentration (985.6 mg/L) than PACSPP(b) (616 mg/L), PACSPP(a) (246.4 mg/L) and PACSPP(c) (554.4 mg/L). Thus, PACSPP(b) was selected as an optimum composite coagulant ratio due to the lower concentration of PAC and the higher ammonia removal percentage.

Table 3 - Composite coagulant PACSPP optimum dose and pH

Type of coagulant	Optimum dose mg/L (pH) PACSPP	Percentage removal (%)		Dose of Al (mg/L)
		Colour	Ammonia	
PACSPP(a)	4,000 (7)	75	44	246.4
PACSPP(b)	5,000 (6)	92	66	616
PACSPP(c)	3,000 (7)	95	49	554.4
PACSPP(d)	4,000 (7)	93	57	985.6

Table 4 - Removal percentage results from the previous study

Sample	Coagulant (dose)	Percentage removal (%)		Reference
		Colour	Ammonia	
Leachate	PAC (900 mg/L)	93.5	9.7	[7]
Leachate	PAC (5,000 mg/L)	97.4	-	[15]
	Tamarindus Indica seed (5,000 mg/L)	41.9	-	
	Dual- PAC (2,750 mg/L) + *Tamarindus Indica* seed (2,000 mg/L)	97.3	-	
Leachate	Tapioca Starch (2.5 g/L)	54.7	13.2	[32]
Leachate	Dual – Alum (3 g/L) + Barley (0.8 g/L)	98	-	[29]
Leachate	Dual – PAC (3,500 mg/L) + *Manihot esculenta* peel extract (250 mg/L)	96.6	33.8	[31]
Leachate	Dual – PAC (2,200 mg/L) + *Durio zibethinus* seed starch (400 mg/L)	96.1	-	[33]
Leachate	PAC (2.5 g/L)	89	19	[25]
	Tapioca starch (2.5 g/L)	30	38	
	Dual – PAC (1.5 g/L) + Tapioca starch (0.2 g/L)	92	21	
	Composite – PAC and Tapioca starch (1.5 g/L)	82	24	
Leachate	PAC (3,750 mg/L)	98	28	[27]
	Tapioca peels (1,000 mg/L)	80.2	18.2	
	Composite – PAC and tapioca peels (2,446.18 mg/L)	88.5	23.7	

Based on the study by Azizan [25], using tapioca starch (TS) as a natural coagulant combined with PAC was able to remove 24% and 82% of ammonia and colour with an optimum dose of 1.5 g/L at pH 5 (Table 4). Meanwhile, Mohd-Salleh et al. [27] used a composite coagulant made of tapioca peel, and PAC obtained 23.7% and 88.5% of ammonia and colour, respectively. The result from Azizan [25] and Mohd-Salleh et al. [27] were compared with the optimum composite coagulant PACSPP(b) of this study, and it showed that PACSPP(b) was able to remove colour and ammonia better than both of the previous studies. Furthermore, PACSPP(b) obtained the best ammonia removal compared to the previous studies listed in table 4. Compared to the pH condition, Azizan [25] shows pH 5 as the optimum pH, while for PACSPP(b), it happens at pH 6 as an optimum, which is more stable. The stable performance for most natural coagulants was in the range of 6 to 8 [34].

3.5 Composite Coagulant PACSPP(b) Efficiency Comparison with Single Coagulants (PAC & SPP) and Dual Coagulant (PAC + SPP)

In the case of a single coagulant, the optimum dose and pH of single coagulant PAC and SPP are 3,000 mg/L at pH 6 (colour: 92% and ammonia: 41%) and 250 mg/L at pH 8 (colour: 5% and ammonia: 4%), respectively. figure 8 compares the composite coagulant with single coagulant and dual coagulant. figure 8 shows that the single coagulant was more effective in removing ammonia than the dual coagulant due to the high dosage of PAC used in the single coagulant (3,000 mg/L and 462 mg/L of Al) compared to the dual coagulant (2,000 mg/L and 308 mg/L of Al). Meanwhile, dual coagulant (PAC + SPP) removed colour and ammonia at 93% and 37% with a 2,000 mg/L dose at pH 8. Different pH values of single coagulant and dual coagulant also affect the values of removal percentage. The reagent for adjusting the pH value can affect the removal percentage of the leachate sample and remove some impurities in the leachate [25]. Single coagulant PAC at 2,000 mg/L dose removed 86% and 30% of colour and ammonia, respectively. It shows that the removal percentage for dual coagulant (colour: 93% and ammonia: 37%) at the same dose of PAC had increased the values of removal percentage. As a result, compared to single coagulant PAC and SPP, adding SPP to a dual coagulant improved removals and reduced dose utilisation. The application of composite coagulant PACSPP(b) with 5,000 mg/L dose at pH 6 has a higher removal percentage (92% for colour and 66% for ammonia) compared to the removal percentage for dual coagulant. This study shows the composite coagulant PACSPP(b) works best in removing ammonia compared to the optimum dual coagulant at 2,000 mg/L dose for PAC and 250 mg/L for SPP (colour: 93%

and ammonia: 37%). Compared to dual coagulant at 4,000 mg/L dose (616 mg/L of Al; colour: 12% and ammonia: 36%) with the composite coagulant (492.8 mg/L of Al; colour: 88% and ammonia: 68%), it shows that composite coagulant has better efficiency in wastewater treatment.

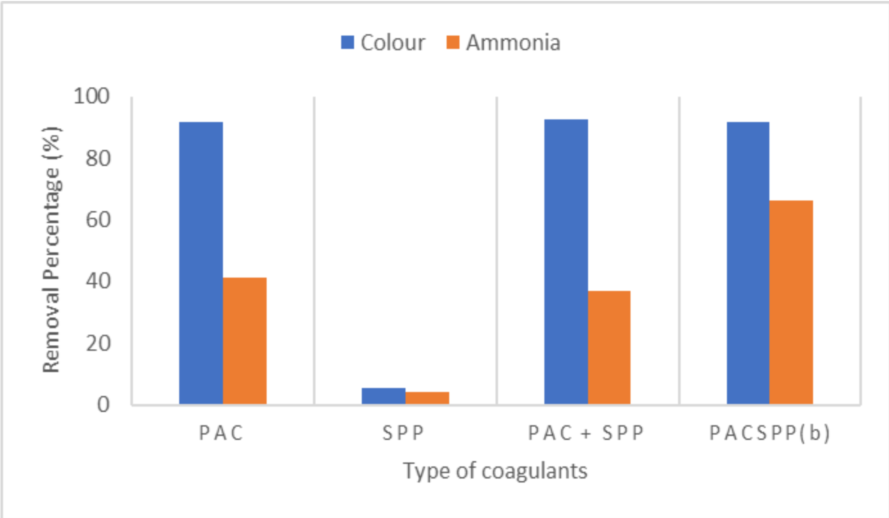

Fig. 8 - Comparison of the composite coagulant with single coagulant and dual coagulant

4.0 Conclusion

The optimum dose and leachate pH for the composite coagulant of PACSPP(b) was 5,000 mg/L at pH 6, with the removal percentages of colour and ammonia being 92% and 66%, respectively. The removal percentage of colour by PACSPP(b) is at par with the single coagulant (PAC) and dual coagulant (PAC+SPP). Meanwhile, ammonia removal by PACSPP(b) is better than the single coagulant (PAC) and dual coagulant (PAC+SPP). These results show that a composite coagulant made up of a chemical coagulant (PAC) and natural coagulant (SPP) combined the best qualities of both coagulants and improved the coagulation effectiveness. Some redocumentation can be made such as use a variety of chemical coagulants as well as natural coagulants, examine other removal parameters such as COD, turbidity and heavy metal and study the characteristics of coagulant and determine the sludge formation through XRD, SEM, zeta potential and IEP.

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