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Utilization of Cowpea Seeds (*Vigna Unguiculata L.*) Compare to Poly Aluminium Chloride (PAC) in the Coagulation-Flocculation Process of Peat Water

Dewi Fitria[1]*, Yuli Ashari Harianja[1], Edward[1]
Environmental Engineering Study Program, Faculty of Engineering, University of Riau, Indonesia

***Corresponding author:** dewifitria@lecturer.unri.ac.id

Peat water has special characteristics, namely brownish color, high organic matter and high ferric content. It requires a treatment before it can be used for daily needs. The water treatment used in this research was coagulation-flocculation with Cowpea seeds and poly aluminium chloride (PAC) as coagulants. The coagulation method aims to destabilize particles so that they can be combined with other particles to form larger aggregates and able to settle down. The purpose of this study was to determine the efficiency of using Cowpea seeds for removing organic matter and color in peat water, to evaluate the sludge volume index (SVI) and sludge mass and then to compare the results of using cowpea seeds with PAC as coagulants. In this study, Cowpea seeds can remove 91% of organic substance, 94.85% of color, has 38.29mL/g of SVI value and 98.26% of sludge mass. Meanwhile, PAC can remove 98.67% of organic, 98.26% of color, has 129.55mL/g of SVI value and 118.14% of sludge mass. The results of this study shows that Cowpea seeds have almost the same ability as PAC to be a coagulant but the dose is high. Cowpea seeds dose is more than 14 times of PAC dose.

Keywords: peat water, Cowpea seeds biocoagulant, PAC, organic matter, color, sludge volume index (SVI), sludge mass.

Introduction

The term "peat water" refers to water found in peatland areas. Indonesia has peatland areas of more than 36 million ha, which is 36% of the total tropical peat (UNOPS, 2022). These peatland areas spread across several islands including Sumatra. Riau is the region with the largest peatland on Sumatra Island, about 56.1% of the total peatland area in Sumatra. Based on the 2019 Peat Restoration Agency report, peatland in Riau Province is estimated to be 3,918,746 ha with a depth of > 2 m. As peat soil contains more than 90% of water, it makes peat water very potential to be a source of clean water for daily needs in peaty rural areas of Riau Province. If peat water could be treated effectively, it will provide a solution for communities in peat areas who still have difficulty obtaining clean water sources.

Peat water is surface water from peat soil that has special characteristics, with a brownish color, high organic content, high ferric (Fe) content and low hardness (Qadafi et al., 2023). Processing peat water into clean water can be done in several ways such as a combination of neutralization, aeration, coagulation-flocculation, sedimentation and filtration processes (Elma et al., 2021). Coagulation-flocculation are two interrelated processes to form larger sizes of flocs. This process uses coagulants and sometimes flocculants in a certain process. The coagulation method aims to destabilize particles so that they can combine with other particles to form larger aggregates and be able to settle down (Reynolds and Richards, 1996). The coagulation-flocculation process usually uses synthetic material as coagulants such as aluminium sulfate salts and poly aluminium chloride (PAC) (Zhou et al., 2021). Despite its good ability on coagulation process, synthetic coagulants such as alum compounds can trigger Alzheimer's disease and PAC has neurotoxic properties (Zhang et al., 2018). Furthermore, the basicity of PAC has been found to be closely related to the residual Al content in treated water (Zhang et al., 2018). Due to the negative impacts caused by the use of synthetic coagulants, research is needed to find alternative coagulants that are safe and environmentally friendly.

Natural coagulants are coagulants that come from natural materials. Those have advantages such as relatively cheap prices, available in large quantities, easily decomposed by microorganisms (biodegradable), non-toxic, less sludge volume, and stronger and more stable flocs formed (Amran et al., 2021). Based on the active ingredients of the coagulant, natural coagulants can be divided into polyphenols, polysaccharides, and proteins. Protein from legumes is one of the commonly used sources of natural coagulants, because in addition to being effective, legumes are easy to obtain, and require relatively simple treatment, including drying, size reduction, extraction, and purification (Kristianto, et al., 2018). Legumes that can be used as natural coagulants must have high protein content (Kristianto et al., 2018), one type of legume that has a high protein content is Cowpea seeds (*Vigna unguiculata L.*). Cowpea seeds contain 23–32% protein (Abebe and Alemayehu, 2022)

In this study, the effect of using cowpeas as a biocoagulant in reducing the organic content and color of peat water has been studied and the SVI and sludge mass produced have also been observed. As a new potential biocoagulant, the comparison of cowpeas performance as a biocoagulant with the performance of well known qualified coagulants is needed. This comparison will give further information about the ability of Soybean to remove contaminants in the coagulation process. The results of this cowpea biocoagulant were compared with PAC results to compare its effectiveness. PAC was used to represent metal coagulants commonly used in drinking water treatment.

Research Methods

Peat water sample

The water sample used in this study was taken from Rimbo Panjang Village, Tambang District, Kampar Regency, Riau Province-Indonesia. After that, the water sample was preserved in the refrigerator along the coagulation experiment. The initial characteristic of peat water is shown in *Table 1*.

Table 1. *Initial characteristic of peat water*

Parameter	Initial Value
pH	4.3
Organic Substance	281 mg/L
Color	389 PtCo

Cowpea seeds preparation

Vigna Unguiculata seeds were dried in the sun for 7 days (DeLong, 2006), then grounded using a blender to produce powder and sieved using a 200 mesh filter.

Analysis of Cowpea seeds characteristics

The characterization of Cowpeas seeds was analyzed using the Fourier transform infrared (FTIR) to determine the compounds or functional groups contained in the Cowpea seeds.

Cowpea seeds optimum pH test procedure

The optimum pH test was carried out to obtain the optimum pH which was used in the main research with a range of 2–8. This range was chosen based on pH in the previous investigation (Amran et al., 2021; Istiqomah et al., 2023; Yimer and Dame, 2021). The first step was to measure the pH of river water and then add NaOH solution to the water sample according to the predetermined pH range. After reaching the respective pH, Cowpea seeds biocoagulant was added following the jar test process. The jar test procedure is explained below. After the jar test, the organic substance and color were tested. The optimum pH determination was based on the highest removal of water parameters which was by pH 3.

Coagulant dosage determination procedure

The range of coagulant dosage was determined by using trial and error method. Initially, coagulant was added to peat water with a large dose variation. After obtaining the coagulant range that could remove the largest amount of organic matter, it was continued with a smaller range dose variation. This was continued until a small range was obtained that could remove the largest amount of organic matter.

Coagulation procedure

500 ml of the sample was put into each beaker glass, then 0.1 M NaOH or 0.1 M H_2SO_4 was added to reach the pH 3. Cowpea seeds biocoagulant or PAC were added into each beaker, then stirred at a speed of 100 rpm for 1 minute and continued at a speed of 40 rpm for 20 minutes. The sample was left for 15 minutes until the flocs settled. After that, samples were taken by decanting process to measure organic substance and color. Meanwhile remaining sludge was used to measure sludge volume index (SVI) and sludge mass. All of the experiments were undertaken by two repetitions.

Organic substance measurement

100 mL of the sample was pipetted into a 300 mL Erlenmeyer flask and 3 boiling stones were added. A few drops of $KMnO_4$ 0.01 N have been added to the sample until a pink color appears. 5 ml of 8 N organic free sulfuric acid was added then heated on an electric heater at a temperature of 105 ± 2. Once there was a smell of H_2S, continue boiling for several minutes. 10 mL of 0.01 N $KMnO_4$ standard solution was pipetted and heated until boiling for 10 minutes. 10 mL of 0.01 N oxalic acid standard solutions was pipetted and titrated with 0.01 N potassium permanganate until pink. The volume of $KMnO_4$ used was noted, if more than 7 mL of 0.01 N potassium permanganate standard solution was used, repeated the test by diluting the test sample.

Color measurement

Color was measured by using a Hach Spectrophotometer model DR/2010 using a wavelength of 455 nm according to colorimetric methods. Absorbance produced by spectrophotometer was used to determine the color of coagulated water (Pt-Co).

Sludge volume index (SVI) analysis

Determination of the sludge volume index was carried out after the flocculation coagulation process and the organic substances and color have been analyzed. The sludge was put into a 1000 mL Imhoff funnel and then allowed to settle for 30 minutes by gravity to form sediment. The SVI value of the sludge obtained was calculated using the following *Equation 1*:

$$SVI_{30}\ (\text{mL/gram}) = \frac{SV\left(\frac{mL}{L}\right) X\ 1000 \frac{mg}{g}}{MLSS\ \left(\frac{mg}{L}\right)} \quad (1)$$

where SV – volume of sludge in the Imhoff funnel (mL/L); MLSS – mixed liquor suspended solid (mg/L).

MLSS is the total amount of suspended solids in the form of organic and mineral materials including microorganisms. The way to determine MLSS is by filtering the mixed sludge with filter paper, then the filter paper is dried at a temperature of 105°C for 1 hour and the weight of the solids is weighed.

Sludge mass

To determine sludge mass was by filtering the sludge resulting from the flocculation process using Whatman filter paper no. 42 which has been weighed. Then the filter paper was dried at room temperature ± 30 minutes and continued drying using an oven at 103–105°C for 1 hour then weighed again (APHA, 2017). Calculating the sludge mass of a sample can use *Equation 2* below.

$$Sludge\ Mass\ (\%) = \frac{SMo - SMt}{SMo}\ \mathrm{X}\ 100\ \% \qquad (2)$$

where SMo – weight of wet sludge (g); SMt – weight of dry sludge (g).

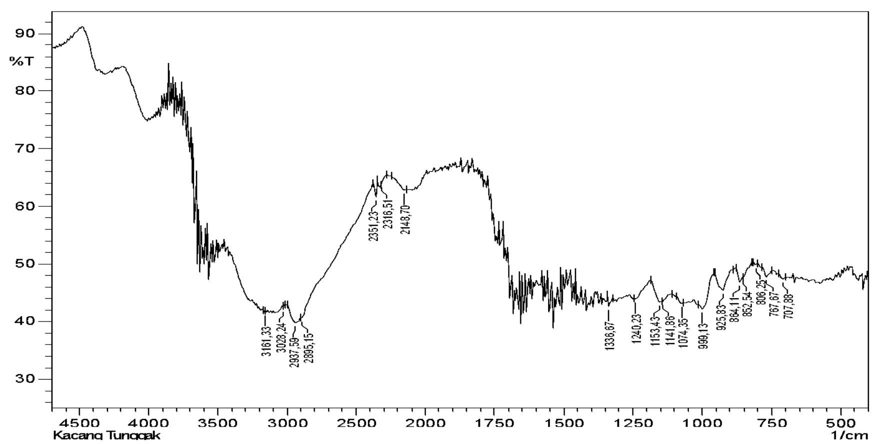
The weight of wet sludge is the sludge resulting from settling before drying, while dry sludge is sludge that has been dried at room temperature ± 30 minutes and continued drying using an oven at a temperature of 103–105°C for 30 minutes then weighed again.

Results and Discussion

FTIR analysis of Cowpea characteristics

Cowpea seeds characteristic test has been done to determine the active components in Cowpea seeds that can act as coagulant. Cowpea seeds characterization test was carried out using the FTIR to determine the functional groups of compounds in Cowpea seeds. The analysis was carried out by comparing the peak spectrum obtained with the wave number range for each functional group. The results of the infrared spectrum analysis with FTIR can be seen in *Fig. 1*.

Fig. 1. *Cowpea seeds FTIR test results*



From the FTIR test results above, it can be seen that there are absorption bands that indicate vibrations from functional groups in Cowpea seeds. *Table 2*. shows data of functional groups in Cowpea seeds.

Overall, the results of the infrared spectrum indicate the presence of polymers containing alkane, alkene, phenol alcohol, carboxylic acid and amine groups. According to Amran et al. (2021); Halder (2021); Yimer and Dame (2021), alkane, alkene and amine groups are active components and can act as coagulants because they can bind colloidal particles and form flocs that able to settle down. Furthermore, Carbonaro et al. (2008) said that the compound that plays a role in the coagulation flocculation process is a protein whose molecules consist of the elements C, H, N, O, and S.

Organic substance removal of Cowpea seeds and PAC

The dose of coagulant is one of the factors that affect the coagulation flocculation process. The addition of coagulant must be suitable with the needs in forming flocs, because the concentration of coagulant greatly affects the collision of particles (Reynolds and Richards, 1996). Flocs would be difficult to form if the addition of coagulant concentration is lacking, likewise the addition of too much coagulant can cause turbidity to occur again and the flocs are not formed properly. The effect of Cowpea seeds addition to remove organic content in peat water is shown in *Fig. 2* and the effect of PAC to remove organic content in peat water is shown in *Fig. 3*.

Table 2. *Cowpea Seeds FTIR spectrum results*

No	Functional Groups	Compounds	Wavelength (cm^{-1}) Cowpea seeds(1)	Frequency Range (cm $^{-1}$)(2)
1.	Alkane	C-H	2937. 59 2895. 15 1336. 67	2850–2960, 1350–1470
2.	Alkene	C-H	3028. 24 864. 11 852. 54 806. 25 767. 67 707. 88	3020–3080, 675–870
3.	Alcohol, phenol (H bond)	O-H	3161. 33 3028. 24 2937. 59 2895. 15 2351. 23 2316. 51 2148. 70	2000–3600
4.	Carboxylic Acid	O-H	3028. 24 3161. 33	3000–3600
5.	Amina	C-N	1240. 23 1336. 67	1180–1360

Fig. 2. *Cowpea seeds vs organic removal*

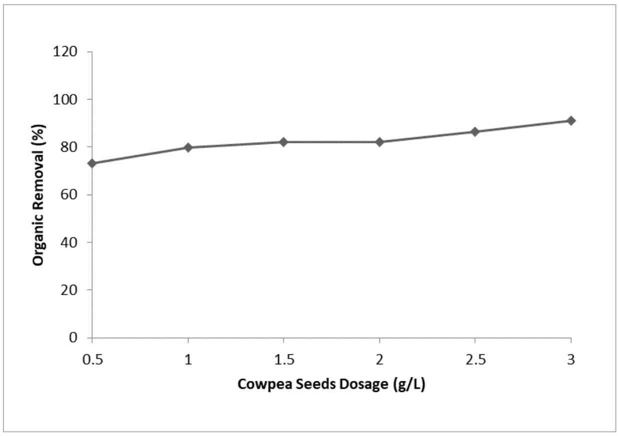
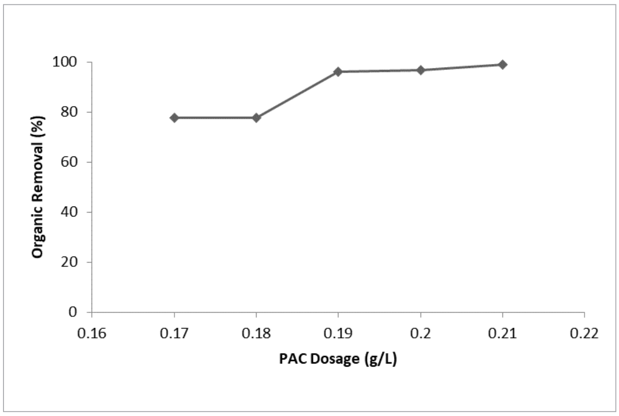


Fig. 3. *PAC vs organic removal*



From *Fig. 2*, the highest efficiency of organic matter removal by using Cowpea seeds biocoagulant was 91%. Furthermore, from *Fig. 3*, the highest efficiency of organic matter removal by PAC coagulant was 98.79%. Those Figures show that the higher the dose of cowpea and PAC added, the higher organic removal achieved. The higher Cowpea seeds and PAC dose will produce the more positive charge in water, so the more flocs formed. This happens because the more cationic produced by the coagulant, the more colloidal particles in peat water are neutralized and form flocs so that organic matter decreases (Putra et al., 2021).

Cowpea seeds as a coagulant can remove organic matter because of its high protein content. Protein from legumes i.e Cowpea seeds, is one of the commonly used coagulants (Kristianto et al., 2018). At acidic pH, the dissolved protein of Cowpea seed powder undergoes protonation, which causes the amino group (NH_2) to combine with the solution's H^+ to form $–NH^{3+}$. The $–NH^{3+}$ group supports the bond between the Cowpea seeds protein and the negatively charged peat water particles.

When Cowpea seeds powder is put into peat water and the coagulation-flocculation process is carried out, the protein will ionize to release positive and negative ions. Dissolved protein from Cowpea seeds contains $–NH^{3+}$ groups can bind negatively charged wastewater particles so that these particles are destabilized. As a result, it will form larger particle sizes that are able to precipitate. While the attraction between the protein charge and the peat water colloidal particles occurs, at the same time, a charge neutralization process also occurs. One of the working principles of the coagulant is the neutralization of the particle charge so that it facilitates the formation of flocs that can settle (Hussain and Haydar, 2020).

PAC as a coagulant can remove organic matter because of the same mechanism with Cowpea seeds which is ionization. Once PAC is added to water it will be ionized to release Al^{3+} ions which will stick to the peat water colloidal particles, neutralize the charge, and can reduce the repulsive force between particles and some of them form aluminum hydroxide ($Al(OH)_3$). As a result, organic matter will be gathered with PAC hydrolysis ions to form floc or absorbed by precipitate ($Al(OH)_3$). And, this will settle together due to gravity, so that the amount of organic matter in the water decreases (Yang et al., 2011).

From this result, it is known that Cowpea seeds are potential to be an alternative biocoagulant to remove organic content of peat water. Even though comparing the efficiency of organic matter removal by Cowpea seeds coagulant and PAC, the PAC coagulant is still slightly better at removing organic matter. This is due to PAC being more soluble than Cowpea seeds, PAC forms floc faster because of its polyelectrolyte group. PAC active aluminate group works effectively in binding colloids which are reinforced with polymer chain bonds from the polyelectrolyte group so that the floc clumps are denser (Zhang et al., 2008).

Color removal of cowpea and PAC

The peat water's reddish brown color comes from the high content of organic matter (humus material) that dissolved in the form of humic acid (Qadafi et al., 2023). The presence of this organic content causes the reddish brown color of peat water to be intense. In this experiment, Cowpea seeds and PAC have been used as coagulant to reduce peat water color, the result is shown in *Fig. 4* dan *Fig. 5*.

Fig. 4 shows that the highest efficiency of color removal using cowpea biocoagulant was at 94.85%. And, the highest color removal efficiency using PAC coagulant was 98.71% (*Fig. 5*). Peat water color reduction by using Cowpea seeds biocoagulant is in association with the removal of organic content. Color reduction can occur because the protein content in Legume seeds after ionization will be able to adsorb and form bonds with contaminants in water (Hussain and Haydar, 2020). Those bonds are stable. As a common process in coagulation with biocoagulant, it is predicted that Cowpea seeds coagulant hydrolysis product can bind with dissolved metal ions in peat water such as Fe and Mn synergistically so that the mass of the floc is greater and settleable. As a result, peat water color after coagulation flocculation will be reduced.

Fig. 4. *Cowpea seeds vs color reduction*

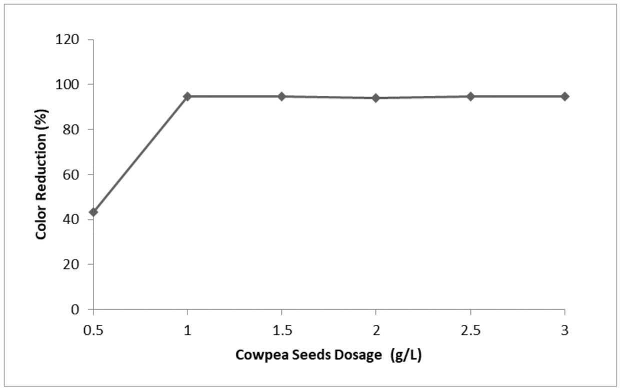
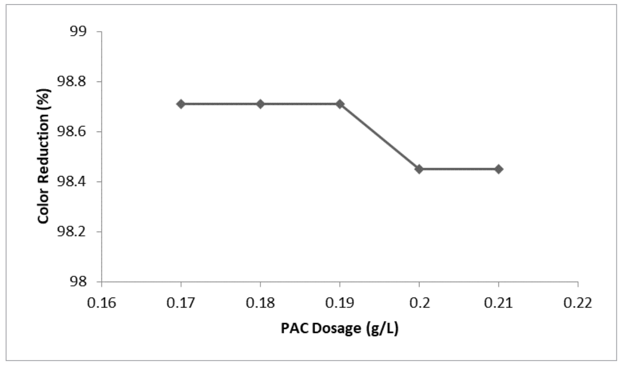


Fig. 5. *PAC vs color reduction*



Charge neutralization and adsorption are found to predominate due to the high efficiency at a low dosage (Otalora et al., 2023). This behavior was attributed to the fact that at acidic pH values, the ionization reaction of the hydroxyl and carboxyl groups (active sites of galacturonic acid) of the Cowpea seeds, as well as the hydrolysis of the glycosidic bonds of the polysaccharide chains, can cause an adsorption process and low bridging between the cowpea molecules and the colloidal particles present in the water sample. It is evidenced by the reduction of peat water color.

Fig. 5 shows that PAC is able to reduce the color of peat water. Once it is added to water, hydrolysis products will be produced. Some hydrolysis products are resulting in neutralization of the charge of color particles in peat water. And then, some of them are able to absorb humic and fulvic acids that cause color in peat water (Elma et al., 2021). Peat water particles will be adsorbed onto the surface of the solid coagulant hydrolysis product. It causes changes in the characteristics of the particle surface which will bind pollutants and then destabilizes stable color-causing particles. It will produce an attractive force and then form flocs that then can settle. Comparing the performance of Cowpea seeds biocoagulant with PAC, Cowpea seeds have almost the same ability as PAC to remove peat water's color. Furthermore, the comparison of Cowpea seeds and PAC performance to reduce color of peat water is presented in *Fig. 6*.

Fig. 6. *Peat water before and after coagulation with Cowpea and PAC*

(a) Peat water (b) Cowpea result (c) PAC result

Sludge volume index of Cowpea biocoagulant and PAC

SVI is the volume of sediment in mL units of 1 gram of sludge solids after 30 minutes of sedimentation in a 1000 mL cylinder. The purpose of measuring SVI is to determine the ability of the sludge to settle and to see the performance of the coagulant so that the resulting sludge does not cause new problems for the environment. SVI is measured by settling the volume of sludge using a 1000 mL Imhoff funnel for 30 minutes. A good SVI value ranges within 50–150 mL/g which can indicate the ability to settle large flocs (Oladoja and Aliu, 2009; Chetana et al., 2016). The SVI values of Cowpea seeds coagulant and PAC can be seen in *Fig. 7* and *Fig. 8*.

Fig. 7 shows that the best SVI value of cowpea coagulant was 38.29 mL/g, while from *Fig. 8*, the best SVI value of PAC coagulant was 129.55 mL/g. From *Fig. 7* and *Fig. 8*, the SVI value produced by cowpea coagulant is much lower compared to SVI value produced by PAC coagulant. If the SVI value obtained is low, it means that the sample condition is dominated by small flocs, conversely, if the SVI value is high, it means that the sample condition is dominated by large flocs (Taheriyoun et al., 2020). The small floc size causes the floc to be unable to unite so that it is difficult to form aggregates and is difficult to settle. Thus, increasing turbidity in the water will occur. Meanwhile, the large floc size

Fig. 7. *Cowpea seeds vs SVI*

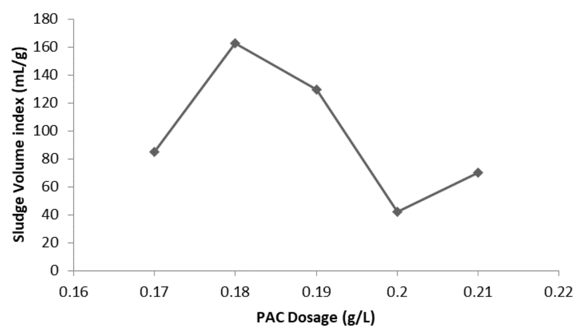
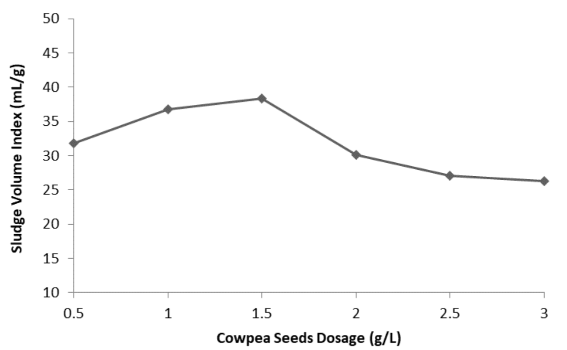


Fig. 8. *PAC vs SVI*



makes it easier for the floc to unite and form big aggregates which then can settle.

The SVI value of PAC coagulant is higher than Cowpea seeds biocoagulant because both have different chemical properties. PAC is an aluminum compound with chemical properties that allow polymerization and the formation of dense flocs, so that it can produce more sludge (Hussain and Haydar, 2020). Cowpea seeds can also form flocs but the flocs formed are not as dense as those formed by PAC coagulants. It seems that protein flocs of Cowpea seeds biocoagulant form a less dense hydrolysis product compared to PAC synthetic coagulant.

Sludge mass of Cowpea and PAC coagulants

Determination of the sludge mass value was obtained from the filtration process sludge. The sludge that was previously used to measure SVI, was dried using an oven at a temperature of 105°C. This sludge mass value indicates the aggregate settling ability. The sludge mass value can be used to determine the amount of sludge formed during the coagulation-flocculation process and also implies coagulant performance. *Fig. 9* and *Fig. 10* show the sludge mass value of Cowpea seeds biocoagulant and PAC coagulant.

Fig. 9. *Cowpea seeds vs sludge mass*

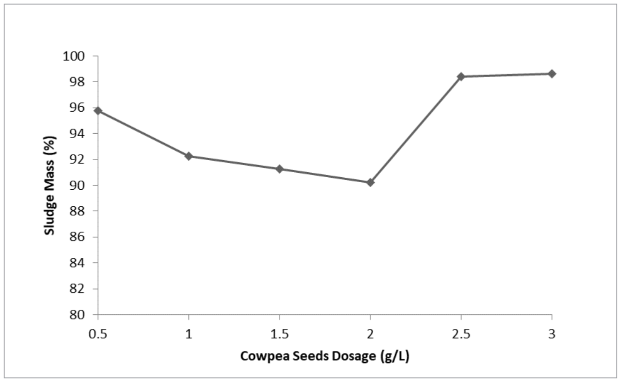


Fig. 10. *PAC vs sludge mass*

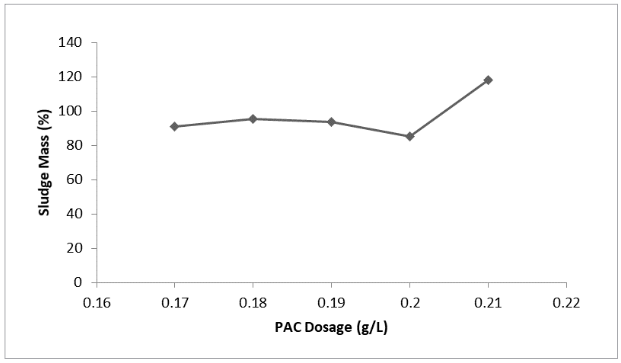


Fig. 9 shows that the highest value of Cowpea seeds sludge mass was at 98.6%. And from *Fig. 10*, the highest sludge mass value of PAC coagulant was 118.14%. From *Fig. 9* and *Fig. 10* above, it is known that the sludge mass value of Cowpea seeds biocoagulant is lower than the sludge mass of PAC coagulant. Sludge mass is correlated with floc size. The formation of larger flocs can allow better sedimentation in water. Larger floc sizes can produce higher sludge mass because the flocs are heavier and easier to settle, while small floc sizes produce lower sludge mass values because the flocs are lighter and difficult to settle (Chetana, et al., 2016).

The sludge mass value of PAC coagulant is higher than Cowpea seeds biocoagulant because of the differences in the chemical properties of the two coagulants. PAC is a polyelectrolyte compound that usually consists of aluminum hydroxide polymers that are more soluble in water compared to protein in Cowpea seeds. These chemical properties of PAC allow it to react faster with suspended particles in water and form larger flocs (Elma et al., 2013).

In theory, the SVI and sludge mass values are inversely proportional. A high SVI value will provide a low sludge mass value calculation, and vice versa if the sludge mass is low then the SVI value will be high (Clifton, 1998). From the results of this study, the SVI and sludge mass values are directly proportional where the SVI and sludge mass values are both large. The sludge mass value of Cowpea seeds coagulant is lower than PAC because PAC is more effective in coagulating small particles in water so that it can form larger flocs. A more effective coagulant is the one with a high sludge mass because it is able to make the floc settle faster thus more mud is deposited.

Conclusion

Based on the research results regarding the use of cowpea (*Vigna unguiculata L.*) and PAC as coagulants in the coagulation-flocculation process of peat water, it is known that PAC is still slightly better than Cowpea seeds as a coagulant. From organic removal, color removal, SVI and sludge mass of both coagulants, PAC is still slightly superior compared to Cowpea seeds. But

Cowpea seeds are still potential to be an alternative coagulant. The problem is related to the large dose of Cowpea seeds, which is more than 14 times the PAC dose. This requires further study if Cowpea seeds are to be implemented as a biocoagulant in peat water treatment.

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