Design of Precipitation System for the Removal of Total Suspended Solid, Turbidity and Mineral Content from Coal Processing Plant Wastewater

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Abstract

A precipitation system was designed for the removal of total suspended solid (TSS) from wastewater coming from a coal processing plant (CPP). The wastewater from the studied CPP had a pH in the range of 3.4-3.6 with TSS and turbidity of 1000 and 150 mg·L^{-1}, respectively. The selection and dose optimization of the cationic coagulants (Ferric chloride (FeCl$_3$) and Aluminium sulphate (Al$_2$[SO$_4$]$_3$)), inorganic polymer coagulant (Poly Aluminium Chloride (PAC)) and a commercial coagulant (brand name (N8100)) were carried out. The results showed that N8100 at 5 mg·L^{-1} dose and pH 6-8 gave the best performance with the final wastewater had TSS 10 mg·L^{-1}, Turbidity 3 mg·L^{-1}, Fe 0.11 mg·L^{-1}, Mn and Al each <0.01 mg·L^{-1} (undetectable). These results were below the limit set by the Indonesian wastewater standard for coal mining industry. The designed precipitation system used for the removal of total suspended solid consisted of equalization pond (960 m^2), inlet channel with baffles and coagulant-lime control for optimum mixing and three identical sedimentation ponds (each with the lower dredge volume of 105 m^3 and the whole pond volume of 675 m^3). These ponds retained the sediments for 3 months prior to cleaning. Coagulants N8100, FeCl$_3$, Al$_2$(SO$_4$)$_3$ and PAC had the operating costs 10.03 US$/day(lowest) 118.95, 22.06 and 21.77 US$/day, respectively. Besides the low operating cost N8100 had produced least sediments compared to FeCl$_3$, Al$_2$(SO$_4$)$_3$ and PAC.

**Key words:** Coagulant, jar test, total suspended solid (TSS), sedimentation, turbidity.

Introduction

Recently, coal mining in Indonesia has steadily increased to 178.8 million metric tons in a year. With this rate of production, coal mining in Indonesia will continue for more than a hundred year to come[1]. Besides making profits, mining activity always has negative impacts, especially on the environment of the surrounding of mining area. Moreover, inappropriate water management has reduced the amount of available resources[2]. Acid mine drainage (liquid-phase waste produced from sulfides minerals (commonly pyrite) when exposed to water and/or air) is the primary waste produced not only during coal mining but also in the extraction and processing phases. In the extraction phase can result in the acid mine drainage with ratio of a ton of acid mine drainage for a thousand tons of coal produced whereas during processing phase (such as washing, crushing, and water removal) acid mine drainage is also produced in large quantity[3]. A precipitation system is commonly utilized to meet the demands for coal mine wastewater disposal. The type of coagulant used, its dose and pH of the wastewater are some of the parameters that determine the effectiveness of the precipitation system[4,5,6]. Ferric chloride (FeCl$_3$), Aluminium sulphate (Al$_2$[SO$_4$]$_3$), Poly-aluminium chloride (PAC), and other poly-electrolyte have been used in wastewater treatment facilities to reduce TSS, turbidity and mineral content[7,8,9]. FeCl$_3$ and Al$_2$(SO$_4$)$_3$ undergo hydrolysis When dissolved in water, Ferric salt in water hydrolyzes to monomer species ([Fe(H$_2$O)$_6$]$^{3+}$, [Fe(H$_2$O)$_5$(OH)]$^{2+}$ (pK_1 2.2), [Fe(H$_2$O)4-(OH)$_2$]$^+$ (pK_2 3.5), Fe(OH)$_3$(s) (p$K3$ 6), and [Fe(H$_2$O)$_2$(OH)$_4$]$^-$ (pK_4 10)), and polymeric species ([Fe2(H2O)$_8$(OH)$_2$]$^{4+}$ and/or [Fe^{2-} (H$_2$O)$_7$(OH)$_3$]$^{3+}$)^{10}. The

overall reactions are normally simplified by the following reaction.

$$2FeCl_3 + 3Ca(HCO_3)_2 \rightarrow 2Fe(OH)_{3(s)} + 3CaCl_2 + 6CO_2 ...... 1$$
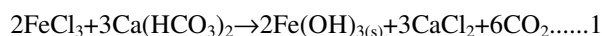
Likewise, aluminium salt in water hydrolyzes to monomer species ([Al(H$_2$O)$_6$]$^{3+}$, [Al(H$_2$O)$_5$(OH)]$^{2+}$ (p$K1$ 4.9), [Al(H$_2$O)$_4$(OH)$_2$]$^+$ (p$K2$ 5.6), Al(OH)$_3$(s) (pK_3 6.7) and [Al(OH)$_4$]$^-$ (pK_4 7.6)) and polymeric [Al$_2$(OH)$_2$]$^{4+}$, [Al$_3$(OH)$_4$]$^{5+}$ and/or the known "Al$_{13}$" [Al$_{13}$O$_4$(OH)$_{24}$]$^{7+}$species[11]. A precipitation charge neutralization (PCN) model was also proposed[12]. According to PCN model coagulation with aluminium salts involves three steps: (a) destabilization begins after addition of a dose of coagulant that exceeds the operational solubility of aluminium, (b) aluminium hydroxide species are then deposited onto colloidal surfaces, and (c) under typical conditions, the metal hydroxide is positively charged, while the original colloidal particles are negatively charged. The overall reactions are normally simplified by the following reaction,

$$2Al_2((SO_4)_3.14H_2O) + 3Ca(HCO_3)_2$$
$$\downarrow$$
$$2Al(OH)_{3(s)} + 3CaSO_4 + 6CO_2 + 14H_2O \ ...2$$
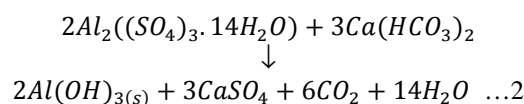
Apart from the inorganic coagulans, Polymermic coagulant such as PAC has been extensively used due to its buffering effect. Various base species have been formed during the production of PAC. These include the monomers Al^{3+}, [Al(OH)]$^{2+}$, [Al(OH)$_2$]$^+$, [Al(OH)$_3$]$_{(am)}$ and [Al(OH)$_4$]$^-$, as well as a dimmer [Al$_3$(OH)$_2$]$^{4+}$, a trimer [Al$_3$(OH)$_4$]$^{5+}$ and the tridecamer ([Al$_{13}$O$_4$(OH)$_{24}$]$^{7+}$, often denoted by Al$_{13}$)[13].

The wastewater from one of the CPP in Kalimantan, Indonesia was posing a big risk to the environment of the surrounding area because the precipitation system there was not designed properly. This CPP wastewater had a pH in the range of 3.4-3.6 with total TSS and turbidity of 1000 and 150 mg·L^{-1}, respectively. Below pH 4.5 we expect no metal hydroxide formation[14]. The existing facility was basically a sedimentation pond dug in the available land without concrete walling and any lining. Apart from this the coagulant dosing was inefficient and was manually carried out. Prior to disposal into the river, the water from the CPP had high turbidity and blackish colour. The conditions further exacerbates during monsoons season.

The objectives of this research was to; (1) select the type and optimize the concentration of a coagulant for the removal of TSS, turbidity and mineral content in the wastewater coming from the CPP Kalimantan, and (2) to design a precipitation system which can meet the local and Coal Mining Industry standards for wastewater[15, 16]. We believe our design will be a potential step forward in curtailing the wastewater problems associated with CPP.

Material and Methods

Materials: Wastewater samples were collected from the inlet of CPP, Kalimantan, Indonesia. Sodium hydroxide (NaOH), cationic coagulants (FeCl$_3$ and Al$_2$[SO$_4$]$_3$), inorganic polymer coagulants (PAC) and were purchased from Merck whereas a commercial coagulant N8100 was obtained from Nalco. All the chemicals were of analytical grade and were used as received.

Methods: To optimize the precipitation process a series of jar tests were conducted in 1 L flask containing 500 mL wastewater (original pH in the range of 3.4-3.6). The pH of the waste water samples was initially adjusted to 5, 6, 7 and 8. Then the coagulants (FeCl$_3$ Al$_2$(SO$_4$)$_3$, PAC and N8100) were added to the samples in 50, 100, 150, and 200 mg.L^{-1} and were stirred first at 100 rpm for three minutes (destabilization phase of colloid particles), followed by stirring at 20 rpm for 12 minutes (flocculation phase). Finally settling was allowed for 15 minutes to form flock by precipitation. Measurements of pH, turbidity, TSS, and mineral content were carried out with pH mater (Multi 340i WTW), turbidity meter (Hi 98703 HANNA), standard method (SNI M-03-1989-F), and Atomic Absorption Spectrometer (AA-6300 GC FID-9A Shimadzu), respectively.

Results and Discussion

Coagulant Tests: To remove TSS, turbidity and dissolved mineral content efficiently, and to comply with the pH in wastewater, the initial pH (adjusted with 1N Sodium hydroxide), type and dosage of coagulants are very important. The effect of dose of four coagulants (FeCl$_3$, Al$_2$(SO$_4$)$_3$, PAC, and N8100) on the pH, turbidity, TSS and mineral content (Fe, Mn and Al) was studied.

Effect of coagulant dose on Turbidity and Total Suspended Solid: Figure 1 and figure 2 show the removal of Turbidity and TSS as function of the four coagulants' doses. figure 1(a) and figure 2(a) show that at optimum dose (100 mg·L^{-1}) of FeCl$_3$ and at initial pH 8, TSS decreased from 1000 mg·L^{-1} to 150 mg·L^{-1}. At minimum solubility, metal hydroxide becomes saturated and readily precipitated. Visual observation also revealed that flock forming was larger at this dose and pH than those at other conditions. These complied with standard value of wastewater disposal from coal mining industry (i.e. <200 mg·L^{-1})[16]. At optimum dose (100 mg·L^{-1}) of FeCl$_3$ and at initial pH 8 turbidity was also decreased from 150 mg·L^{-1} to 5 mg·L^{-1}, The decrease turbidity might be attributed to the fact that as metal hydroxides precipitation increased, the sweep flock efficiency became higher. Therefore final turbidity value became lower. At iso-electric point (where zeta potential value of ferric hydroxide approaches zero) coagulation process run optimally because enmeshment mechanism of the colloid particles in precipitant took part i.e., colloid particles were captured in metal hydroxide flock structure. At low dose the flock did not appear since the coagulant was not adequate to compress the double layer of the colloid particles or to bind the colloid particles to form bridging. At coagulant dose lower than 100 mg·L^{-1}, low colloid particles enmeshment occurred, which had resulted in lower efficiency of sweep flock mechanism and in small size flocks formation. High dose (more than100 mg·L^{-1}) of the coagulant in the suspension caused charge stabilization of colloid particles due to the adsorption of counter ions (which in this case was Fe^{3+}). Excess adsorption of the counter ions caused the charge of colloid particles to become positive, which had resulted in the separation of the particles due to electrostatics repulsion and in low flock appearance.

In case of Al$_2$(SO$_4$)$_3$ (figure 1(b) and figure 2(b)) turbidity and TSS decreased, regardless of the initial pHs, to less than 20 NTU and 20 mg·L^{-1}, respectively. These were far less than the standard value of wastewater disposal from coal mining industry. The optimum dose was achieved at150 mg·L^{-1} where the turbidity and TSS dropped to lowest value. Other doses still had turbidity and TSS values under allowable value. However, further addition of Al$_2$(SO$_4$)$_3$ coagulant affected the pH of the solution as mentioned in the next section.

Figure 1(c) and figure 2(c) show that optimum dose for PAC was attained at 100-150 mg·L^{-1} and pH 8. At 150 mg·L^{-1} big flocks were formed and readily observed but the turbidity was higher than it was at the dose of 100 mg·L^{-1}. Increase in dose of PAC (from 100 to 200 mg·L^{-1}) raised the turbidity (again due to re-stabilization of the colloid particle. The turbidity value at initial pH 8 was better observed than at other initial pH. PAC contains Al$_{13}$([AlO$_4$Al$_{12}$(OH)$_{24}$(H$_2$O)$_{12}$]$^{7+}$) that has a significant role in flock formation[9, 18]. Al$_{13}$ created bigger and better flocks at base conditions[19]. However, PAC can do coagulation at low alkalinity. As PAC hydrolization takes place, the flock

formed incorporates the chloride ion into the flock structure so it is not available for producing acid.

Figure 1(d) and Figure 2(d)) show that N8100 dose at 5 mg·L^{-1} had already reduced the Turbidity and TSS to below 10 NTU and 20 mg·L^{-1}. Increase in dose of N8100 slightly affected the turbidity and TSS of the solution. N8100 coagulant works by neutralizing particle charge and by bridging mechanism, which was the main advantage of this coagulant. Experiments with N8100 used lower concentration compared to the previous experiments with FeCl$_3$, Al$_2$(SO$_4$)$_3$ and PAC. In this experiment, 15 mg·L^{-1} visually gave the most observable flock formed. This coagulant has quaternary ammonium chloride functional group with positive charge active sites in its polymer structure. The active sites have a function as colloid particle charge neutralize and act as an inter-particle bridge between colloid particles to form flock. The formation of

big flock structure helps to destabilize the suspension with sweep flock mechanism.

Effect of coagulant dose on pH: Figure 3 (a-d) depicts the effect of coagulants (FeCl$_3$, Al$_2$(SO$_4$)$_3$) PAC and N8100 dose on the pH in the wastewater samples. The pH of the samples decreased significantly with addition of the 150 mg.L^{-1} of FeCl$_3$ and Al$_2$(SO$_4$)$_3$ coagulants (figure 3a-b). The decrease in pH was due to the formation of Fe(OH)$_3$ and Al(OH)$_3$ metal hydroxides precipitates. Addition of 200 mg.L^{-1} of FeCl$_3$ and Al$_2$(SO$_4$)$_3$ further decreased the pH down to the range of 2-4 (figure 3 (a-b)), which was not favourable for precipitation. A pH reduction was also observed in PAC addition, although the reduction gradient was smaller (figure 3c) compared to FeCl$_3$, and Al$_2$(SO$_4$)$_3$. PAC contains monomer, dimmer and trimmer bases such as Al^{3+}, [Al(OH)]$^{2+}$, [Al(OH)$_2$]$^+$, [Al(OH)$_3$]$_{(am)}$, [Al(OH)$_4$]$^-$, [Al$_3$(OH)$_2$]$^{4+}$, [Al$_3$(OH)$_4$]$^{5+}$ and [Al$_{13}$O$_4$(OH)$_{24}$]$^{7+}$. These
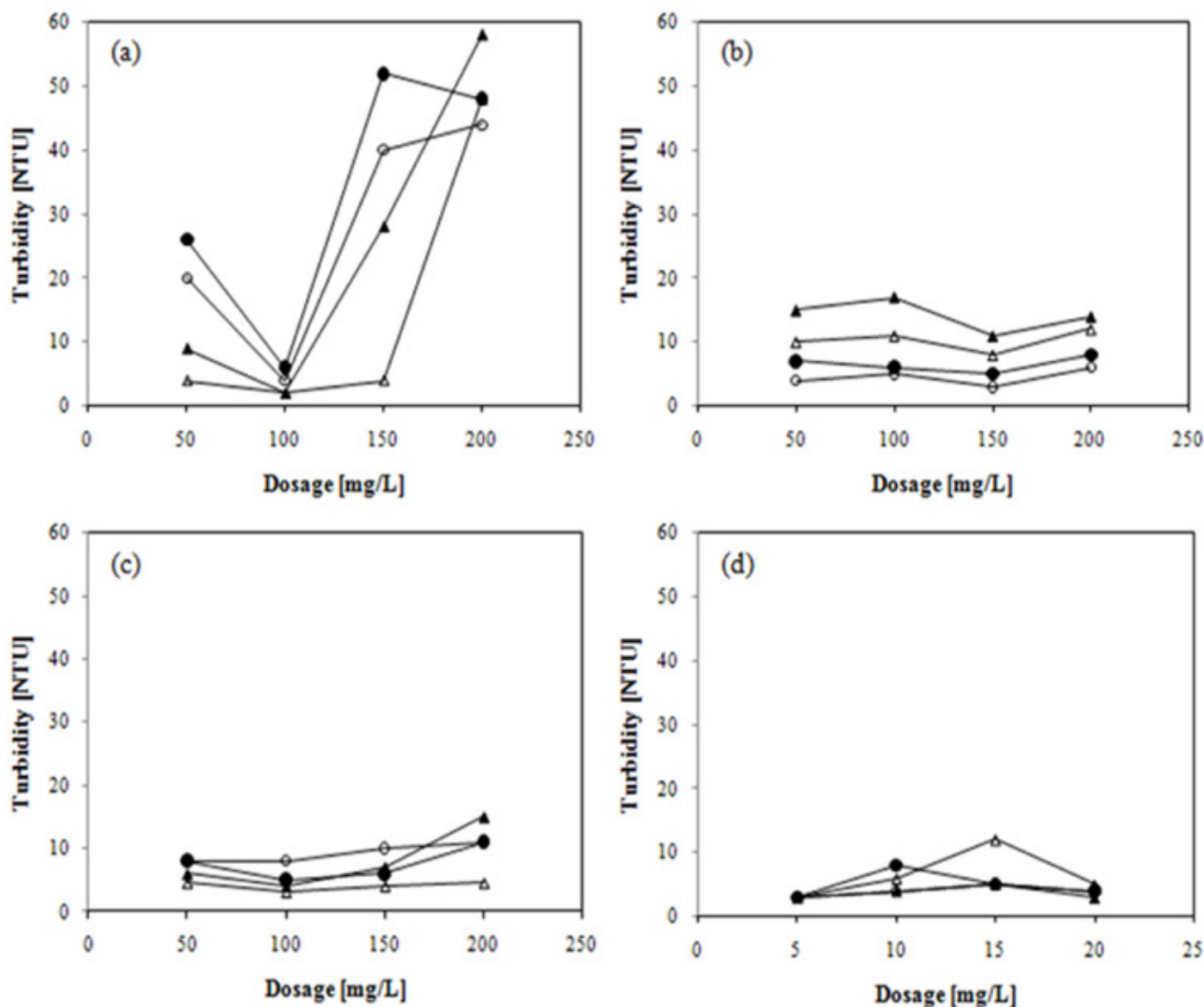
Figure-1
Turbidity after addition of A. FeCl$_3$; B. Al$_2$(SO$_4$)$_3$; C. PAC; and D. N8100; in pH 5 to 8
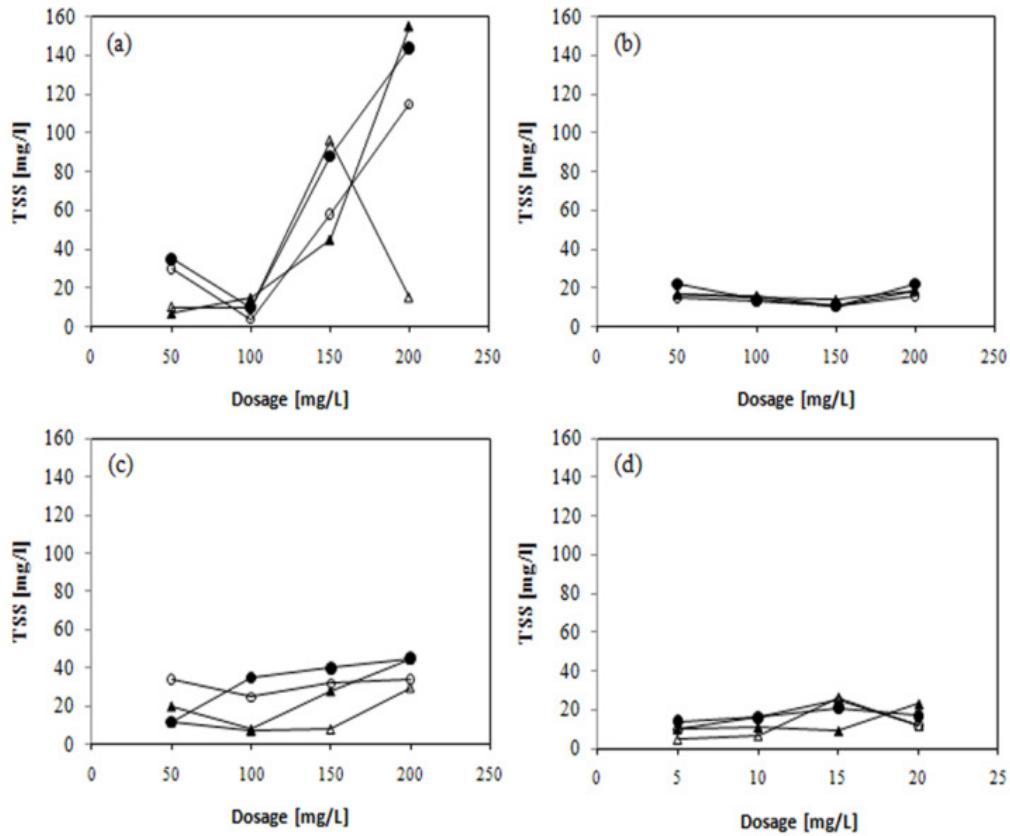
Figure-2
Total Suspended Solid (TSS) after addition of A. FeCl₃; B. Al₂(SO₄)₃; C. PAC; and D. N8100; in pH 5 to 8

Figure-3
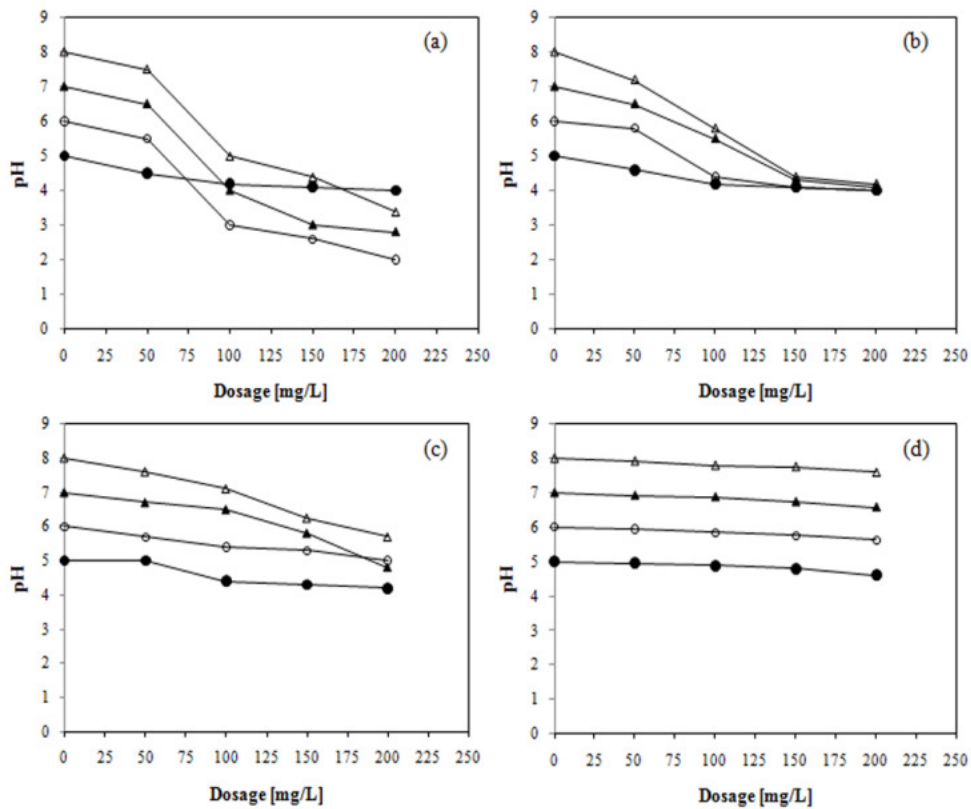**Conditions of pH after addition of (a) FeCl₃; (b) Al₂(SO₄)₃; (c) PAC; and (d) N8100; in pH 5 to 8**

bases resists pH reduction in wastewater, thus, the pH reduction was slightly smoother compared to the one in case of $Fe(OH)_3$ and $Al(OH)_3$. N8100 is a cationic organic polymeric material (containing epichlorohydrin dimethylamine) with between 50,000 to 150,000 $g\cdot mole^{-1}$ molecular weight [20]. Addition of N8100 at 200 $mg\cdot L^{-1}$ reduced the pH only by 0.4 pH scale (figure 3d), which might be due to the presence of large number of basic amine groups.

Dissolved Metal: Table 1 shows the maximum allowable pH and dissolved metal content of wastewater before and after precipitation with selected dose of coagulants and adjusted pH. The results showed that proper coagulant dose and adjustment of pH had reduced Fe, Mn and Al concentration to below allowable limit, regardless of coagulant used. However, lowest metal content ((Fe (0.11 $mg\cdot L^{-1}$), Mn (<0.01 $mg\cdot L^{-1}$), Al(<0.01 $mg\cdot L^{-1}$)) was observed by adding 5 $mg\cdot L^{-1}$ of N8100 at pH 7 to the waste water samples, which proved the effectiveness of this coagulant to reduce metal concentration in wastewater. The N8100 has long polymer chains and hence, this significant metal content reduction might be to the effective bridging mechanism[20]. A rather moderate effect of $FeCl_3$ and PAC at 100 $mg\cdot L^{-1}$ and pH 8 and, and $Al_2(SO_4)_3$ at150 $mg\cdot L^{-1}$ and pH 6. on the dissolved metals was observed. In case of $Al_2(SO_4)_3$, at lower dose (100 $mg\cdot L^{-1}$) and pH 7, dissolved Al increased in the wastewater due to increased solubility of aluminium hydroxide.

Table-1
Dissolved metal content and pH of the wastewater before and after the precipitation at selected coagulant dose and adjusted pH

	Dose $mg\cdot L^{-1}$	Fe	Mn [$mg\cdot L^{-1}$]	Al	pH
Maximum allowable	-	7[1]	4[1]	n.s [2]	6.0 - 9.0[1]
Before treatment	-	20.00	0.38	0.76	3.4 - 3.6
After precipitation with:					
$FeCl_3$	100	0.43	0.12	<0.01	8[3]
$Al_2(SO_4)_3$	150	0.77	0.31	<0.01	6[3]
PAC	100	0.39	0.19	<0.01	8[3]
N8100	5	0.11	<0.01	<0.01	7[3]

[1] Environmental Ministry Act 2003 and East-Kalimantan Governor Law 2002, [2] n.s. = not stated, [3] Measurement was done after pH adjustment and prior to coagulant addition

Design: The TSS removal plant was an open system. Measurements in the field and precipitation data collected were used to calculate the capacity of this new design. First, the precipitation data was collected and the rainfall intensity was calculated by Mononobe eqn.1[14].

$$I = \frac{X_T}{24}\left(\frac{24}{t}\right)^m \qquad ....3$$

The precipitation in 24 hour (X_T) was 105.14 mm, which was calculated using Gumber distribution method (not discussed here). From eqn.1, the rainfall intensity (I) was 36.45 $mm\cdot h^{-1}$, the runoff coefficient (C) for the mining area with no plantation was 0.9; and the rain water catchment area (A) was 5.174 Ha. Using these values the wastewater rate (Q) was calculated as 472 $m^3\cdot h^{-1}$ with eqn. 2;

$$Q = 0.278 \ CIA \ ....4$$

Lay out of the ponds is given in figure 4(a). The designed wastewater precipitation system consisted of: equalization pond (figure 4(b)), inlet channel (figure 4(c-d)), and sedimentation ponds (Figure 4(e)). Pond 1 is the main sedimentation, while Ponds 2 and 3 are the reserved sedimentation ponds. For optimum dose, lime and coagulant control were added into the system.

Inlet Channel: Figure 4(c-d) depict the dimension of inlet channel and its cross section area. The designed inlet channel is a small trapezium channel. This trapezium channel has a 60° slope with the height of wet channel (h) and channel base width (B) is 2h, The wet cross sectional area (A') is $2.575h^2$ and the hydraulic radius (R) is 0.906h. By using Gauckler–Manning formula for open channel (eqn 3) with conversion constant (k) equal to 1, gradient (S) 1%, roughness coefficient (n) 0.016, then the h could be calculated as 0.0051 m (or 5 cm).

$$Q = (k/n) \ R^{2/3} \ S^{1/2} \qquad .... 6$$

To obtain steady-state flock-size distribution inside the sedimentation pond, shear mixing of the coagulant and wastewater should take place before wastewater get into first sedimentation pond. Shear mixing of the solution (coagulant, lime and wastewater) inside the inlet channel could be achieved by putting baffles along the flow.

Sedimentation pond: The design of sedimentation pond is illustrated in figure 4 (e). The pond was designed to allow the settling of sediments formed during coagulation. The actual sedimentation time in lab was 20 minutes. However, for safety precautions sedimentation time (t) was set at 54 minute. The pond volume was calculated using the following equation:

$$V = \frac{t \ Q}{24}f ....5$$

If the wastewater rate (Q) is 472 $m^3\cdot h^{-1}$ t is 54 minute, f (cleaning time factor of sediments) is 6 days then the volume of sedimentation pond can be calculated as 105 m^3. This volume is the volume of lower part of the sediment pond (dredge part). The long slope is made to allow sedimentation fall onto the dredge part. The whole pond can retain sediment up to 1 month without any cleaning 675 m^3. This pond is surrounded by concrete lining to avoid coagulant absorption by the coal-soil layer wall. The second and third ponds are identical dimensionally with the first pond. These ponds are provided to allow further sedimentation and for avoiding flooding when more

wastewater flow come or when cleaning of ponds are delayed until 3 months.

Coagulant and Lime: The amount of coagulant needed is the product of optimum dose multiplied by wastewater debit. After addition of coagulant, the pH decreased to certain level and therefore lime is needed to bring the pH level to their optimum pH level for coagulation. Experiments were carried out to measure the lime needed to achieve optimum pH level in 1 litre flasks containing different coagulants. Multiplying the lime needed in 1 litre flask by the wastewater rate resulted in the need of lime (kg) per day. The results of both coagulant and lime need are listed in Table 2. This table also shows the cost of coagulant, lime and total cost for each type of coagulant used. It is evident that the cost of lime is negligible in the total cost compared to coagulant cost. Among the four coagulants, N8100 has the lowest operating cost for removing TSS. Experiments also showed that no lime was needed for N8100 since this coagulant can work in pH range 6-8. The total cost different between $Al_2(SO_4)_3$, PAC per day is not significant. Nevertheless, cost difference between $Al_2(SO_4)_3$, PAC and N8100 become much evident per year. Sediment formed: The table 3 below illustrates the sedimentation volume formed. The volume of sediment formed depends on wastewater rate, coagulant dose and the characteristic of the coagulant. Handling precipitation improperly will increase the cost of sludge management operations[21]. Experiments were done to measure the sediment formed per litre of wastewater for respective coagulant and pH conditions. Using eqn. $D = Q.d_i$, N8100 produce least sediments than the other coagulants.

Conclusion

All the coagulants used were capable to reduce the TSS from 1000 $^{mg·L^{-1}}$ to far below the allowable concentration, i.e. < 200 mg·L^{-1}. Each coagulant has its optimum conditions to treat this coal processing wastewater. Overall, N8100 at 5 $^{mg·L^{-1}}$ dose and pH 6-8 gave the best performance with the final wastewater has 10 mg·L^{-1}(TSS), 5 mg·L^{-1}(Turbidity) , 0.11 mg·L^{-1}(Fe), where as Mn and Al each <0.01 mg·L^{-1}, which was undetectable. Design of the precipitation system consists of equalization pond, inlet channel with control and baffles for optimum mixing, sedimentation pond (with volume of lower dredge = 105 m^3; and the volume of whole pond = 675 m^3); and two reserve ponds with the same design as main pond that altogether can retain the sediments until 3 months prior to cleaning. Coagulant N8100 had least sediment and the lowest operating cost, i.e. 10.03 US\$/day. FeCl$_3$, $Al_2(SO_4)_3$ and PAC had the total cost of 118.95, 22.06 and 21.77 US\$/day, respectively. The different of these coagulant costs become much evident if we expand the time to a year or more.

Acknowledgment

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Table-2
Coagulant and Lime Cost per day

Coagulant					pH[2]	Lime		Total
Type	**Dose mg·L^{-1}**	**Need[1] [kg]**	**Price/kg**	**Cost**		**Need[3] [kg]**	**Cost[4] [US$]**	**Cost [US$]**
			[US$]					
$FeCl_3$	100	47.2	2.5	118	8	9.5	0.95	119
$Al_2(SO_4)_3$	150	70.8	0.3	21.24	6	8.2	0.82	22.06
PAC	100	47.2	0.45	21.24	8	5.3	0.53	21.77
N8100	5	2.4	4.25	10.03	6-8	-	--	10.03

[1]Based on 472 m3/day wastewater inlet; [2]The optimum pH for coagulation;
[3]Determined by lab experiments; [4]The price of lime is 0.1 US$/kg

Table-3
Sedimentation formed in the pond

Sediment[1]			
Coagulant	L [ml.L^{-1}]	Daily [m^3]	Annually[2] [10^{3}x m^3]
$FeCl_3$	60	28.32	8,5
$Al_2(SO_4)_3$	75	35.4	10,6
PAC	70	33.04	9,9
N8100	50	23.6	7,1

[1]calculated based on lab experiment;
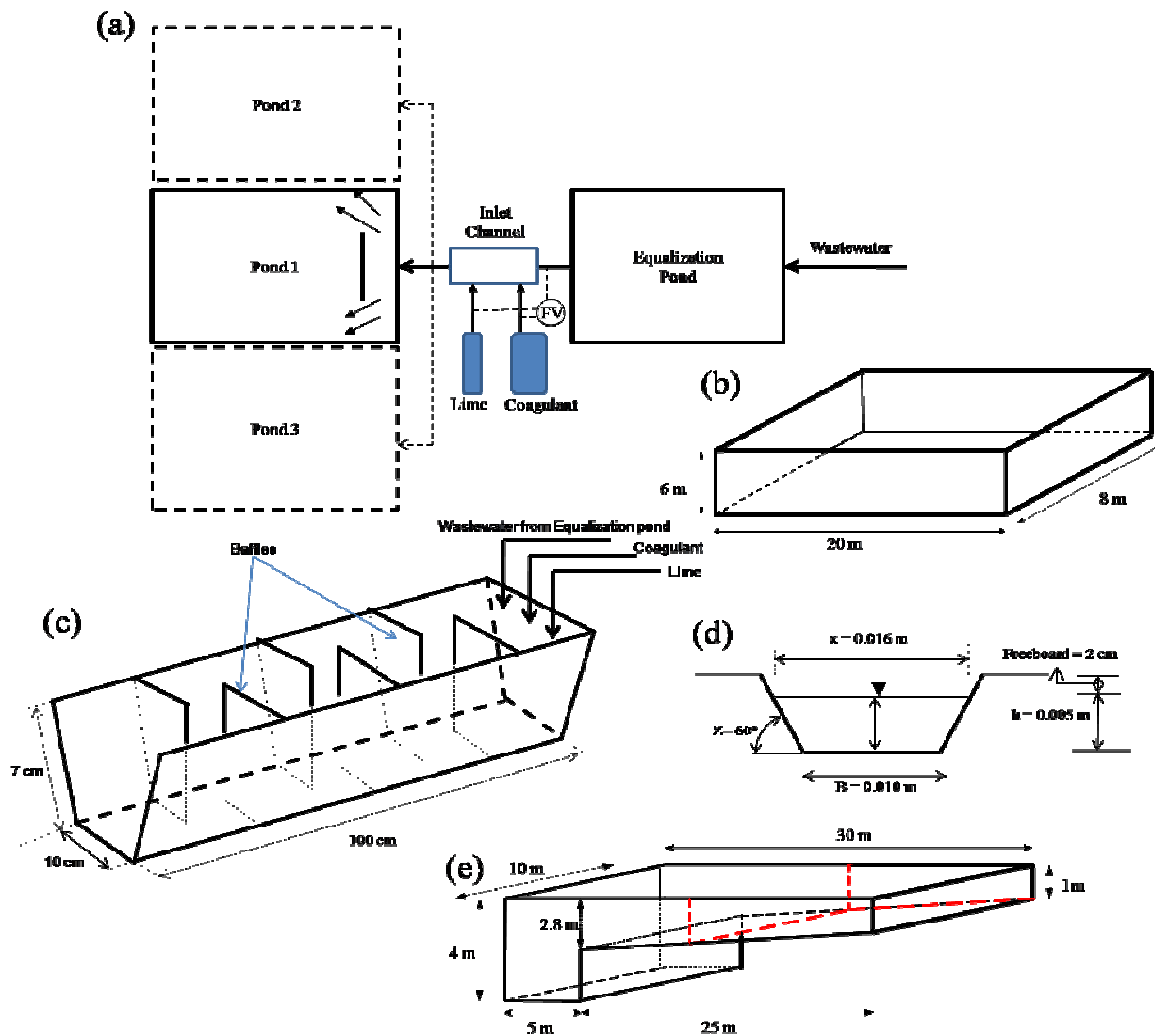[2]Based on 300 day per year

Figure-4
Design of wastewater precipitation system (a) birdview layout; (b) Equalization pond; (c) inlet channel; (d) cross sectional inlet channel; (e) pond 1 (sedimentation pond)