

# Heavy Metals Removal by Hydroxide Precipitation and Coagulation-Flocculation Methods from Aqueous Solutions

Fong Moi Pang, Sheau Ping Teng, Tjoon Tow Teng,\* and A.K. Mohd Omar

*Environmental Technology Division, School of Industrial Technology,  
University Science of Malaysia, 11800 Minden, Penang, Malaysia*

The hydroxide precipitation and coagulation-flocculation methods were used to treat wastewater containing lead, zinc, copper, and iron. The concentrations of heavy metals in the synthetic wastewater range from 1 to 14 mg/L for lead, 5 to 90 mg/L for zinc, 3 to 90 mg/L for copper and 5 to 45 mg/L for iron. Individual Zn(II) and Cu(II) with concentrations below 90 mg/L and Fe(III) with concentrations below 45 mg/L were removed up to 99% by the precipitation method in the pH range of 8.7 to 9.6, 8.1 to 11.1, and 6.2 to 7.1, respectively. Though the highest percent removal of Pb(II) by hydroxide precipitation alone was approximately 98%, the final dissolved concentrations did not fulfill the Standard A discharge limit required by the regulations, thus further treatment by the coagulation-flocculation process was performed. Aluminum sulphate (alum), polyaluminum chloride (PACl) and magnesium chloride ( $MgCl_2$ ) have been used as coagulants together with Koaret PA 3230 as coagulant aid to determine the effectiveness of the coagulation method for the removal of individual heavy metals in the wastewater. The effects of parameters such as pH, type, and dosage of coagulant on the percentage of metal removal, and the amount of coagulant aid on the flocs settling time were investigated. The jar test showed that up to 99% removal of Pb(II) was attained by the addition of 1,200 mg/L of alum, 150 mg/L of PACl, and 2,000 mg/L of  $MgCl_2$  in a pH range of 6.5 to 7.8, 8.1 to 8.9, and 9.7 to 10.9, respectively.

**Key words:** precipitation; coagulation-flocculation, heavy metals

## Introduction

Heavy metal pollution is an existing and growing worldwide problem, especially in developing countries. Heavy metals such as Pb, Zn, Cu, and Fe are often discharged by metal plating, metal cleaning and fabrication, battery manufacturing, chemical manufacturing, and paint and pigment industries. The treatment and removal of heavy metals have drawn considerable attention because of their association with various health problems. Although metals such as Mn, Fe, Cu, and Zn are essential nutrients at low levels (but are toxic at higher concentrations), others such as Hg, Cd, and Pb could bring severe physiological or neurological consequences even in small amounts (Angino et al. 1977; Maessen et al. 1985; Laws 1993; Nebel and Wright 1996; Macchi et al. 1996; Mohammed and Najar 1997).

Owing to the harmful effects of heavy metals to human beings and to aquatic life, permissible limits have been set by environmental authorities for different kinds of waters. In Malaysia, industrial wastewater discharge is regulated by *Sewage and Industrial Effluent Regulations*, 1979 (EQA 1974 [2002]). Various waste constituents may have to be removed or reduced to meet standard discharge limits before being released to the receiving streams. Currently, most of the industries in Malaysia treat their wastewater in order to meet the Standard B discharge limits. The Standard B discharge limits for lead,

zinc, copper, and iron are 0.5 mg/L, 2.0 mg/L, 1.0 mg/L, and 5.0 mg/L, respectively. However, the Department of Environment of Malaysia may raise the industrial discharge limits to become more stringent, especially in the heavy metal concentrations, for protection of the environment from serious pollution in the future. Considerable investment and research have been focused on the improvement of many well-established technologies. Common technologies for the removal of heavy metals from aqueous solutions include chemical precipitation, coagulation-flocculation, ion exchange, and adsorption (Clifford et al. 1986; Grasso et al. 1990; Chung 1997; Mellah and Chegrouche 1997; Mohammed and Najar 1997; Eckenfelder 2000).

The heavy metal concentrations after a precipitation process alone do not necessarily accomplish the limit of pollutants required by the regulations, which is one of the most important criteria in choosing a treatment process (Netzer et al. 1974; Baltpurvins et al. 1995). Even though heavy metal concentrations in the final effluent may meet the discharge limit, the precipitates are too small and take a long time to settle. This is not desirable since a long settling time requires a larger settling tank. Normally, the processes that form the basis for the treatment of heavy metals begin with hydroxide precipitation, followed by coagulation-flocculation-settling, and subsequently the separation of the unsettled flocs from the wastewater stream by filtration.

Coagulation-flocculation is a commonly used physico-chemical method in the treatment of metal-bearing industrial wastewater because it removes

\* Corresponding author: tteng@usm.my

colloidal particles, some soluble compounds, and very fine solid suspensions initially present in the wastewater by destabilization and formation of flocs (Burke et al. 2000). Both aluminum and ferric salts, either in monomer or polymeric forms, have been reported as effective coagulants in treating heavy metals from wastewater (Hannah et al. 1977; Chu 1999; Fan et al. 2003; Kang et al. 2003). Magnesium chloride, as compared with aluminum or iron salts, is a less commonly used coagulant in the field of wastewater treatment. It has been used in a study to investigate the effectiveness of the coagulation process for the removal of colouring matters (Tan et al. 2000; Ooi et al. 2005; Wong et al. 2007). It has been shown that the presence of Mg enhanced the removal of some pollutants (Leentvaar and Rebhun 1982). The effectiveness of seawater liquid bittern as a source of Mg to remove heavy metals has been evaluated (Ayoub et al. 2001); it gave a removal of 92.2, 75.2, and 63.2% for Pb(II), Zn(II), and Cu(II), respectively, at a pH range of 11.0 to 11.5.

It is important to investigate and determine the effectiveness of hydroxide precipitation and coagulation-flocculation in removing heavy metals (lead, zinc, copper, and iron) without major modification of the current wastewater treatment facilities in order to meet the Standard A discharge limits of 0.1 mg/L for lead, 2.0 mg/L for zinc, 0.2 mg/L for copper, and 1.0 mg/L for iron. The effects of parameters such as pH, type, and dosage of coagulant on the percentage of metal removal and the amount of coagulant aid on the flocs settling time have been studied.

### Materials and Methods

The experiments conducted were part of the study of a Master's Degree which had been carried out in the Environmental Laboratory, School of Industrial Technology, University Science Malaysia. All experiments were conducted under the room temperature.

#### Chemicals

Lead(II) nitrate, zinc(II) sulfate heptahydrate, copper(II) chloride dihydrate (Merck KGaA, Germany) of purity >99%, and iron(III) nitrate nanohydrate (Fluka Chemie, Switzerland) of purity >97% were used to prepare the individual heavy metals solutions. Sodium hydroxide, NaOH of purity 99% supplied by Merck KGaA, Germany, was used as the base. Alum of industrial grade and PACl (30%) in powder of analytical grade were obtained from T.C. Chem. Technology (Kulim, Kedah, Malaysia), while magnesium chloride of analytical grade was supplied by Fluka Biochemika. Polyelectrolyte, Koaret PA3230 of commercial grade from Giulini Chemie was used as the coagulant aid throughout the experiment at a concentration of 0.1 g/L. All heavy metal solutions, coagulants, coagulant aids, and sodium hydroxide were prepared by using distilled water.

#### Analyses

The pH of the solution was measured by using a HACH Sension 3 pH meter (HACH, Colorado, U.S.A.), while the concentrations of the heavy metals were determined using a Perkin-Elmer A. Analyst 100 Atomic Absorption Spectrophotometer (Perkin Elmer, Singapore).

#### Jar Tests

The experimental work was divided into two parts. The first part consisted of experiments to determine and compare single metal removal by means of hydroxide precipitation. The percent of metal removal with respect to increasing initial concentrations was studied at various pH levels. Different amounts of NaOH were added to adjust the pH of 150-mL heavy metal solution. The solution was stirred for 1 minute by using the jar test apparatus, and allowed to settle for 30 minutes. The content was then filtered by using Whatman No. 4 filter paper. The pH and the metal concentrations of the supernatant were measured.

In the second part, a coagulation-flocculation process was carried out on Pb at a concentration of 7 mg/L since this is the highest concentration normally found in battery manufacturing wastewater. All experimental work was conducted by using a six-beaker jar test apparatus. Each beaker contained 150 mL of the heavy metal solution. The pH of the solution was adjusted by adding NaOH prior to the addition of coagulant. The samples were mixed at 60 to 65 rpm for 3 minutes. Coagulant aid was then added and the samples were allowed to mix further for 1 minute, followed by 30 minutes of settling. At the end of the settling period, the samples were filtered and analyzed for the concentration of the heavy metal ions. The conditions of the experiments are summarized in Table 1.

### Results and Discussion

#### The Effect of pH on Metal Hydroxide Precipitation

The results of hydroxide precipitation on the selected heavy metal ions are illustrated in Fig. 1 to 4, where the percent removals of Pb(II), Zn(II), Cu(II), and Fe(III), respectively, are plotted against solution pH. Pb(II), Zn(II), Cu(II) and Fe(III) were found to form precipitates at various pHs in the absence of coagulants. All metals precipitate as hydroxides by the addition of NaOH. For each metal concentration, a curve with a maximum point, which represents the highest percentage of metal removal (minimum solubility), was obtained. These maximum points fell in a pH range of 7.8 to 8.8, 8.7 to 9.6, 8.1 to 11.1, and 6.2 to 7.1 for Pb(II), Zn(II), Cu(II), and Fe(III), respectively. The results agree with those reported in the literature (Hautala et al. 1977; Eckenfelder 2000; Kim et al. 2002).

The percent removals of Pb(II) were 69.0, 93.3, 96.9, and 98.3% for the initial concentrations of 1.5, 3, 7, and

TABLE 1. Summary of the conditions of experiments

<i>(a) Hydroxide precipitation</i>		
Type of metal	Concentration range (mg/L)	pH range
Pb(II)	1–14	4–11
Zn(II)	5–90	3–11
Cu(II)	3–90	4–11
Fe(III)	5–45	2–11

<i>(b) Coagulation -flocculation process on Pb(II) of 7mg/L</i>			
Type of coagulant	Coagulant dosage (mg/L)	pH range	0.1 g/L coagulant aid (mL)
Alum	200–4,000	4–11	2
PACl	150–1,200	4–10	2
MgCl <sub>2</sub>	600–6,000	9–12	2

14 mg/L, respectively. The results show that the removal efficiency increased with the initial metal concentration. Though the highest percent removal of Pb(II) was approximately 98%, the final dissolved concentrations (i.e., 0.235 mg/L) do not fulfill the discharge limit required by the regulations, thus further treatment by the coagulation-flocculation process was performed.

Figure 2 shows a plot of percent removal of Zn(II) at different pH values. A similar plot is that of Pb(II) (Fig. 1), with a maximum point corresponding to the minimum solubility of Zn(II) and that falls in the pH range of 8.7 to 9.6 for each Zn(II) concentration; the results obtained are in agreement with those reported in the literature

(Baltpurvins et al. 1996; Saari et al. 1998). According to Baltpurvins et al. (1996), the minimum Zn(II) solubility point falls in the pH range of 9.5 to 10, whereas Saari et al. (1998) indicated that the respective pH range was slightly lower (8.5 to 9.5). The percent removal of 10 mg/L of Zn(II) found in this study was 99.7%, which is higher compared with the 97.3% reported by Saari et al. (1998). Both minimum solubility points fall in the same pH range. The percent removal of Zn(II) increased tremendously from pH 5 to 7. An average increment of 53.6% removal was observed in that pH range for 5, 8, and 10 mg/L of Zn(II). Between a pH of 7 to 9.6, the final concentrations of Zn(II) (at <0.406 mg/L) in the

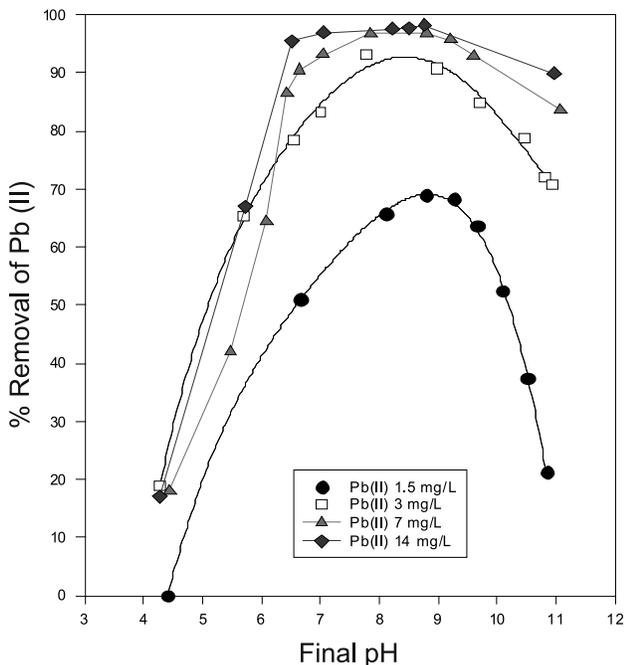


Fig.1. Percentage removal of Pb(II) at different pH in the absence of coagulant.

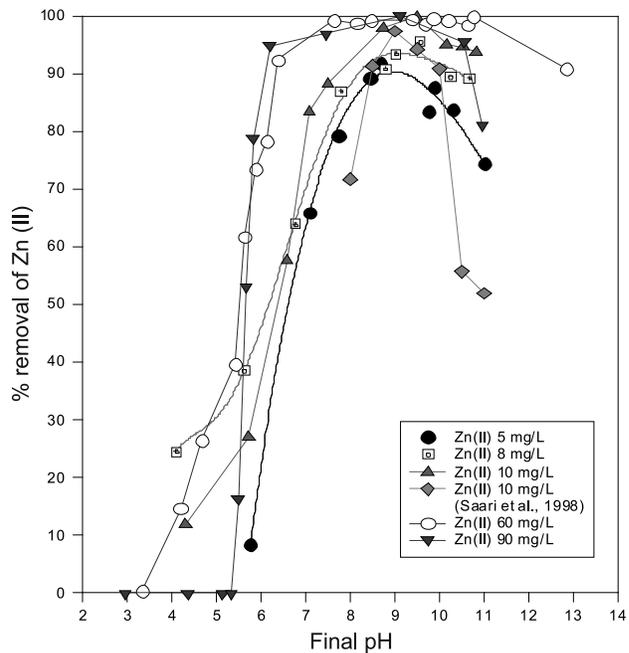
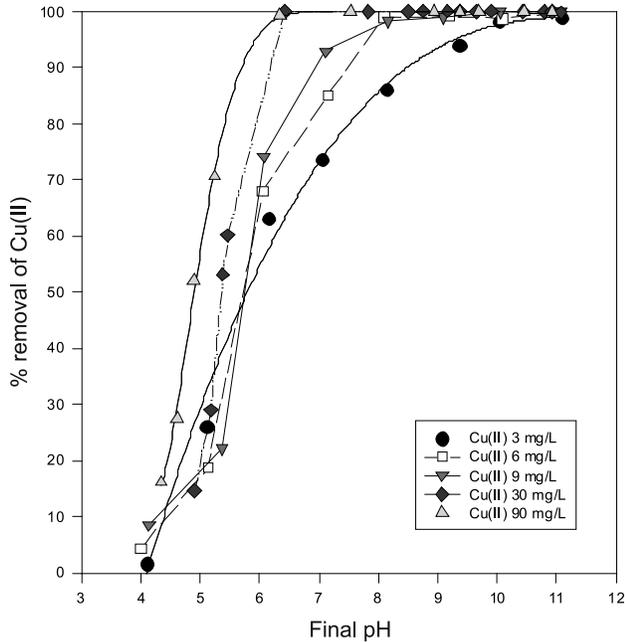


Fig.2. Percentage removal of Zn(II) at different pH in the absence of coagulant.

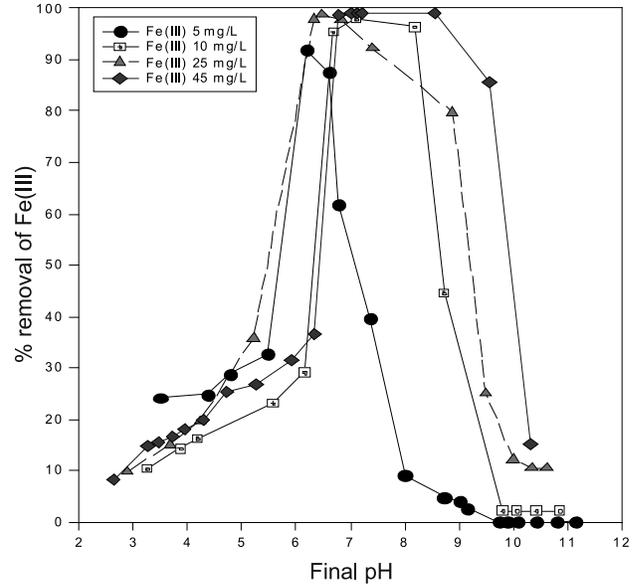


**Fig.3.** Percentage removal of Cu(II) at different pH in the absence of coagulant.

solutions fulfilled the standard discharge limit for all the three initial concentrations.

Generally, the percent removals of Cu(II) with initial concentrations of 3 to 90 mg/L by means of chemical precipitation are relatively high ( $\geq 99\%$ ) as shown in Fig. 3. The effective pH range for the best Cu(II) removal occurred from 8.1 to 11.1 for the initial concentrations of 3, 6, and 9 mg/L of Cu(II). This pH range agrees with the value of Baltpurvins et al. (1996). When the Cu(II) concentration increased to 30 and 90 mg/L, the effective pH range became broader (i.e., pH 6.3 to 11.1), and the final concentration of Cu(II) in the solutions was  $<0.2$  mg/L, thus meeting the standard A discharge limit.

Figure 4 shows the plot of the percent removal of Fe(III) as a function of the solution pH at initial concentrations of 5, 10, 25, and 45 mg/L of Fe(III). It shows that the optimal pH range for Fe(III) removal occurred at a lower pH as compared with the other three heavy metals. The optimal pH range became wider when the initial concentration of Fe(III) increased from 5 to 45 mg/L. For an initial concentration of 5 mg/L Fe(III), the optimal pH range was 6.2 to 6.6, while for the other concentrations, the optimal pH range was 6.7 to 8.6. The optimal pH ranges obtained are similar to those of Hautala et al. (1977). Iron is soluble in acidic solution, but as the pH rose, Fe(II) and Fe(III) were converted into their respective hydroxides. According to Hautala et al. (1977), there was sufficient dissolved oxygen to oxidize  $Fe^{2+}$  to  $Fe^{3+}$  in a dilute neutral solution of iron (50 ppm or less). It was then assumed that probably only the Fe(III) species existed in the prepared solutions. Neutralization of an acidic iron solution with NaOH resulted in the precipitation of  $Fe(OH)_3$ . The precipitate was slow to



**Fig.4.** Percentage removal of Fe(III) at different pH in the absence of coagulant.

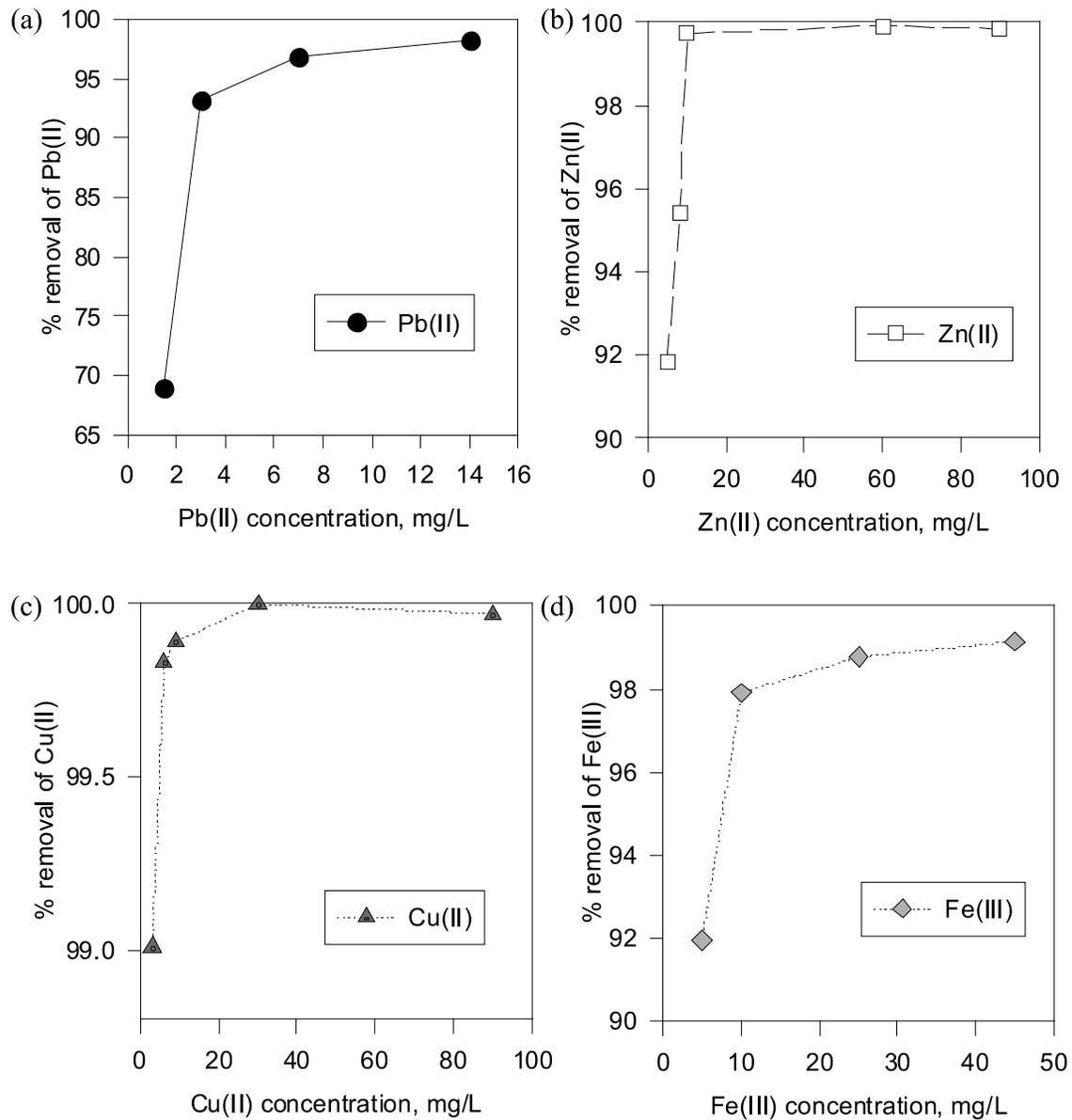
settle and easily redispersed by slight agitation. The percent removal of Fe(III) in the alkaline zone was very poor. Beyond a pH of 9, there was almost no Fe(III) removal from the solution of lowest concentration. In this region, percent removal of Fe(III) increased with the concentration of Fe(III). The percent removals of Fe(III) were 2.2, 25.0, and 85.5% for the initial concentrations of 10, 25, and 45 mg/L of Fe(III), respectively.

At pH corresponding to the minimum solubility, all metals form slightly soluble, gelatinous hydroxides in the presence of equivalent quantities of hydroxide ions, but the hydroxide precipitates may redissolve in water after the minimum solubility point by forming soluble anionic hydroxyl complexes because of amphoteric effects (Maruyama et al. 1975; Saari et al. 1998).

The percent removal of metal increased with initial metal concentrations, as shown in Fig. 5. This is in agreement with the findings of Daniels (1975) and Ayoub et al. (2001) who reported that higher influent metal concentrations result in greater metal removal. It was also found that each heavy metal experienced different removal efficiencies by hydroxide precipitation, which is probably due to its hydroxide solubility properties as well as affinity toward precipitating suspended solids.

#### The Effect of Coagulants on Pb(II) Removal

Chemical precipitation of the heavy metals of different initial concentrations showed that only Pb(II) needed further treatment after the hydroxide precipitation process in order to further reduce the final concentration of Pb(II) in the wastewater. The results of the alum, PACl, and  $MgCl_2$  coagulation experiments of various coagulant



**Fig.5.** Percentage removal of metal as a function of metal initial concentrations. (a) Pb(II); (b) Zn(II); (c) Cu(II) and (d) Fe(III).

dosages on synthetic wastewater containing 7 mg/L of Pb(II) are shown in Fig. 6, 7, and 8, respectively. Upon the addition of coagulant to the alkaline solutions, metal removal was enhanced significantly. Fig. 6 shows that treatment with alum dosages of 1,200, 2,000, and 4,000 mg/L worked well to give >99% removal in the pH range of 6.9 to 8.0. Final concentrations of Pb(II) in the solutions were 0.038, 0.028, and 0.020 mg/L, respectively, which are lower than the Standard A discharge limit.

Figure 7 shows the use of PACl with excellent percent removal for almost all the dosages studied. For the initial concentration of Pb(II) (7 mg/L), percent removal as

high as 98.6% needed to be achieved in order to meet the discharge limit. The corresponding pH range for the effective Pb(II) removal (>99%) was 8.1 to 8.9 when 150 mg/L of PACl was used. When the PACl dosage increased to 600, 800, and 1,200 mg/L respectively, the optimum pH range for the 99% removal became broader (i.e., 7.6 to 9.1) and the solutions contained only a small amount of Pb(II), which complied with the Standard A discharge limit.

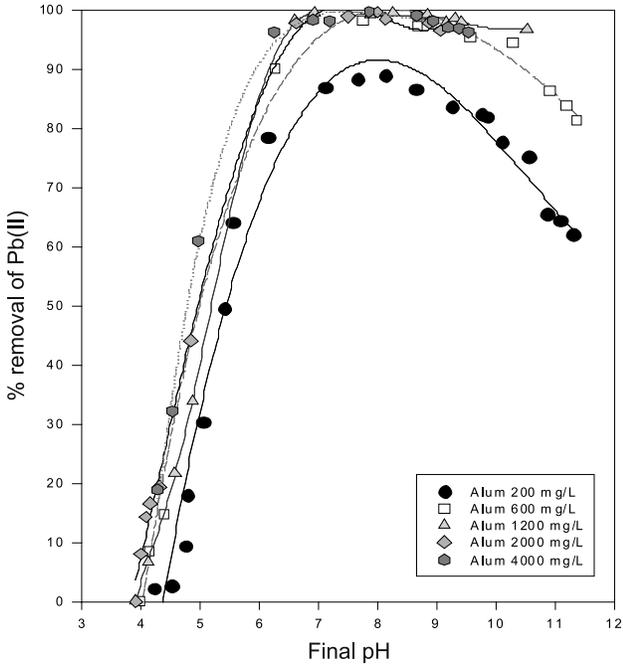
The results of the jar tests with 7 mg/L of Pb(II) treated with  $MgCl_2$  in a pH range of 9.0 to 12.5 are presented in Fig. 8. It shows that a higher coagulant

dosage is needed to remove 99% of Pb(II). MgCl<sub>2</sub> dosages of 600, 1,200, 2,000, 4,000, and 6,000 mg/L gave 92.6, 96.6, 99.4, 98.9, and 98.7% removal, respectively. It was observed that the pH for the maximum percent removal shifted slightly to the lower range when the MgCl<sub>2</sub> dosage increased. This is in agreement with the

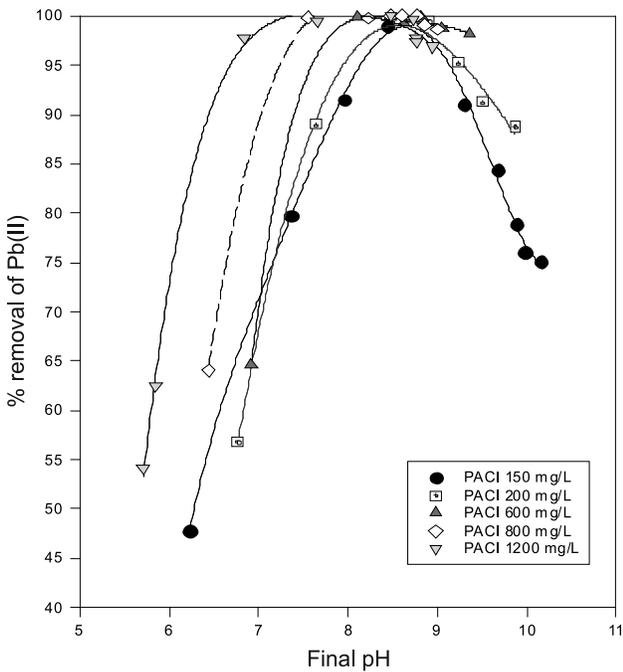
findings of Lefers et al. (1987), who showed that the pH value at which the precipitation of Mg(OH)<sub>2</sub> flocs starts depends on the Mg<sup>2+</sup> concentration in the wastewater. This pH value decreased with increasing Mg content in the wastewater. Therefore, the treatment of wastewater containing relatively high concentration of Mg can be carried out at lower pH values.

Figure 9 shows the percent of Pb(II) removal by various coagulants as a function of the coagulant dosage. The curves were drawn in different optimal pH ranges of 6.5 to 7.8, 8.1 to 8.9, and 9.7 to 10.9 for alum, PACl, and MgCl<sub>2</sub>, respectively. For alum and PACl, the results showed that increasing the concentration of the coagulant increased the efficiency of the Pb(II) removal. The percent removal of Pb(II) increased with the increase of the coagulant dosage up to a limit, after which the percent removal attained an almost constant value. An almost similar removal efficiency for different dosages of coagulants means that the concentration of the coagulants remaining in the solution is higher, which may contribute to toxicity (Stephenson and Duff 1996).

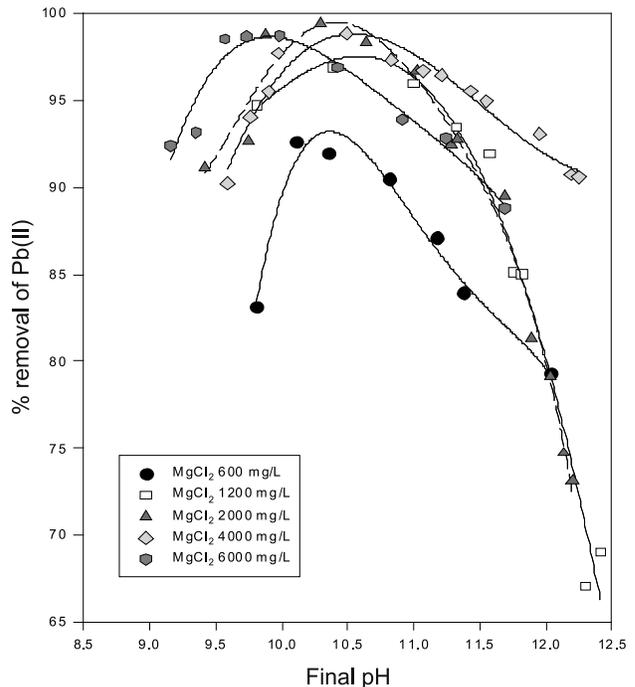
The percent removal of Pb(II) increased from 89.0 to 98.1% when the alum dosage increased from 200 to 600 mg/L. A removal of Pb(II) higher than 99% can be attained by the addition of 1,200 mg/L of alum. The percent removal of Pb(II) was found to be >99% as the PACl dosage was increased from 150 mg/L to higher dosages. A complete Pb(II) removal was achieved when 800 and 1,200 mg/L of PACl were used. In the case of treatment with MgCl<sub>2</sub>, precipitation with NaOH in a pH range of 9.8 to 10.5 in the absence of MgCl<sub>2</sub> was insufficient to achieve good removal of Pb(II). The



**Fig.6.** Treatment of 7 mg/L of Pb(II) with different alum dosages.



**Fig.7.** Treatment of 7 mg/L of Pb(II) with different PACl dosages.



**Fig.8.** Treatment of 7 mg/L of Pb(II) with different MgCl<sub>2</sub> dosages.

final concentration of Pb(II) in the solution was 0.562 mg/L (i.e., 91.9% removal).  $MgCl_2$  dosages of 600 and 1,200 mg/L exhibited lower percent removal of Pb(II) compared with alum and PACl at the same dosages. However, a  $MgCl_2$  dosage of 2,000 mg/L gave almost the same percent removal (i.e., 99.4%) as an alum dosage of 2,000 mg/L (i.e., 99.6%). Further increase of the  $MgCl_2$  to 4,000 and 6,000 mg/L slightly decreased the percent removal. This may be due to the restabilization of the colloidal particles by the excess  $MgCl_2$  hydrolysis species, and therefore the Pb(II) ions remained in solution (Chu 1999).

Among the three coagulants used, PACl, which gives almost 100% removal of Pb(II) at a dosage of 150 mg/L, exhibited better removal efficiency than alum and  $MgCl_2$ . Less PACl was needed than alum and  $MgCl_2$  to achieve the desired heavy metal removal. It has been reported that PACl has a few superior characteristics, including stronger metal removal ability than alum (Van Benschoten and Edzwald 1990; Sonu et al. 1997; Fan et al. 2003). In the presence of an aluminum-based coagulant, Al will remain as a "free ion" in acidic solutions. When the pH of the solution increases, the Al in PACl will form polymerized aluminum hydroxide, which provides a larger surface area over alum for Pb(II) to be adsorbed onto, and to trap the  $Pb(OH)_2$  particles (Stephenson and Duff 1996; Chu 1999).

### The Effect of the Coagulant Aid on Flocs Settling Time

Prior to the addition of the coagulant aid to the solutions, the tiny particles are dispersed in the solution and it takes a long time for the particles to settle down. The use of a coagulant aid greatly improves the flocs separation performance by promoting the growth of agglomerates of fine particles of such size and density that effective solid-liquid separation can be accomplished (Ødegaard 1987). The effect of polyelectrolyte on flocs settling time was studied on the 7 mg/L of Pb(II) aqueous solutions at the corresponding optimal coagulant dosage and pH value. The results displayed in Fig. 10 show that the settling time of the flocs treated with alum, PACl, and  $MgCl_2$  with 0.5 mL of Koaret PA 3230 were 60, 81, and 569 s, respectively. The shortest settling time of the flocs treated with alum and PACl was about 31 s when 2.0 mL of Koaret PA 3230 was added to the solutions. Varying the amount of coagulant aid does not have the significant effect for the treatment by alum and PACl as they could easily settle in a short time. This is in good agreement with the result of Amokrane et al. (1997), who reported that the coagulation-flocculation process using alum as a coagulant and an anionic polymer as the coagulant aid provided flocs of greater size and lower sedimentation time. For the solutions treated by  $MgCl_2$ , the settling time decreased when the amount of coagulant aid increased. This shows that in order to bring down the settling time to about 100 s when  $MgCl_2$  is used, a minimum amount of 5 mL of coagulant aid must be used.

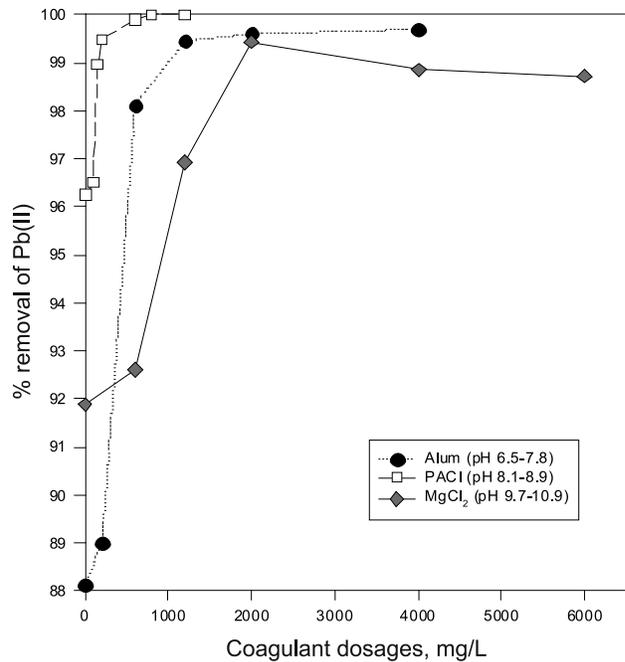


Fig.9. Effect of the use of different coagulants on the removal of 7 mg/L of Pb(II).

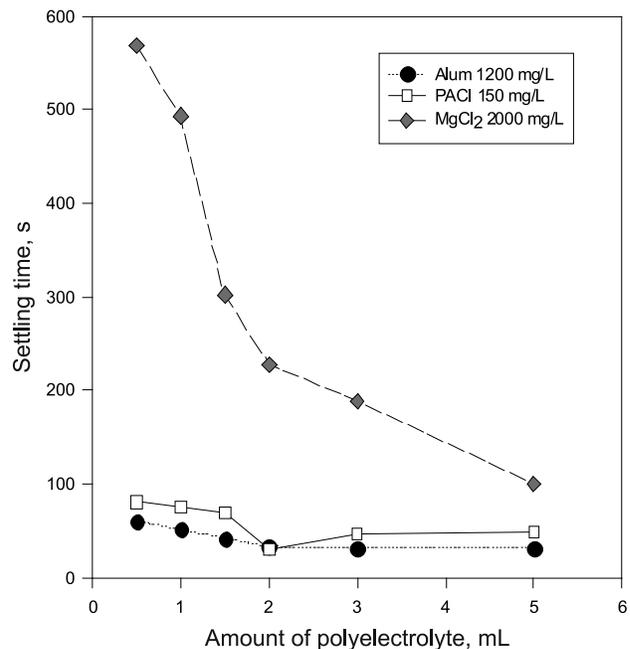


Fig.10. Flocs settling time as a function of amount of polyelectrolyte used.

### Conclusions

The removal efficiency of heavy metals by hydroxide precipitation increased with increased metal concentration. Optimal removal of metals by hydroxide precipitation was attained at a fixed pH, which varied with the type

of metal. Individual Zn(II) and Cu(II) of concentrations below 90 mg/L and Fe(III) of concentration 45 mg/L in aqueous solutions were removed well by the precipitation method in the pH ranges of 8.7 to 9.6, 8.1 to 11.1, and 6.2 to 7.1, respectively. Alum, PACl, and MgCl<sub>2</sub> at dosages 1,200, 150, and 2,000 mg/L could achieve up to 99% removal for Pb(II) with an initial concentration of 7 mg/L at a pH range of 6.5 to 7.8, 8.1 to 8.9, and 9.7 to 10.9, respectively.

### Acknowledgment

The authors gratefully acknowledge the financial support by the Ministry of Science, Technology and Environment Malaysia under “Skim Biasiswa Khas.”

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Received: 22 November 2007; accepted: 7 January 2009.