

A method for calculation of ion distribution in reaction system forming hydroxide

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Abstract: A formula was proposed to calculate the distribution of metal ions quantitatively in chemical reaction system forming hydroxide where precipitation and complex are formed together. The effects of some factors on formation of precipitation and complex were investigated, and the corresponding precipitation rates of zinc, iron (III), aluminum, copper and magnesium were calculated. As a result, it shows that the proposed formula is reliable. By the proposed formula, the existence state of metal ions in hydroxides reaction system with any metal ions can be well described and the effects of some factors on the distribution of metal ions were determined.

Key words: hydroxide; metal ions distribution; precipitation rate; complex forming rate

1 Introduction

In non ferrous metallurgy, hydroxide precipitate method is commonly used to recover valuable metals and purify the leaching solution usually [1–7]. For example, magnesium hydroxide and calcium hydroxide are used to form hydroxides precipitates to recover valuable metals such as nickel, cobalt, cadmium, copper, zinc and chrome from leaching solution or wastewater [1–5]. In zinc hydrometallurgy, zinc calcine is leached by sulfuric acid solution first, and then it is put into the leaching solution to neutralize in order to separate impure metals by hydroxides precipitation method [6,7]. In addition, hydroxide precipitation method is used in the manufacture of nano-sized metal hydroxides, e.g. nano-sized copper hydroxide and magnesium hydroxide [8–10], and complex metal hydroxides materials, e.g. rare earth (Pr, Nd, Sm, Eu, Gd, etc.) complex metal hydroxide, and Ni(II)–Cr(III) double hydroxides [11–13]. Except for the alkali metals and another few metals, most metal hydroxide solution contain precipitates

($M_i(\text{OH})_{n_i}$) and complex ions ($M_i(\text{OH})_{g_{ij}}^{(g_{ij}-n_i)^-}$) [14–16]. Therefore, the quantitative description and reasonable explanation for the distribution of metal ions in this complex metal hydroxide solution system have an important significance for scientific research and industrial production.

Research on chemical reactions in hydroxide solution system has been reported [1,5,14,15], but it is restricted to precipitation reaction system ($M_i(\text{OH})_{n_i}$) or coordination reaction system ($M_i(\text{OH})_{g_{ij}}^{(g_{ij}-n_i)^-}$) in the single component. Therefore, few works were reported on the multi-component chemical reaction system containing both precipitate ($M_i(\text{OH})_{n_i}$) and complex ($M_i(\text{OH})_{g_{ij}}^{(g_{ij}-n_i)^-}$).

Based on the quantitative study on the chemical reaction in the above solution system, a reaction model and a formula were proposed, which are suitable for the multi-component chemical reaction system containing both precipitate ($M_i(\text{OH})_{n_i}$) and complex ($M_i(\text{OH})_{g_{ij}}^{(g_{ij}-n_i)^-}$). According to the calculation results, the distribution of metal ions is analyzed in the multi-

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component solution containing zinc, iron (III), aluminum, copper and magnesium and pH changes with the increase of the total alkali concentration. Furthermore, the calculated results and experimental results are compared.

2 Multi-component hydroxide reaction system mode

When certain amount of alkali is added to multi-component solution containing several metal ions, the metal ions ($M_i^{n_i+}$) and hydroxide ion (OH^-) will react and form precipitate ($M_i(OH)_{n_i}$) and complex ($M_i(OH)_{g_{ij}}^{(g_{ij}-n_i)-}$).

Reaction system model is shown in Fig. 1, where $M_i^{n_i+}$ represents any metal ions in solution ($i=1, 2, \dots, p$), Y_k represents the complexing agent, while the solid line represents main reaction and the dotted line represents side reaction.

3 Theoretical derivation of formula

The precipitation rate (G) of metal cations is defined as the ratio of the concentration of metal cation used to form hydroxide precipitation ($M_i(OH)_{n_i}$) and the total concentration of the metal cation, expressed as

$$G_i = \frac{c_{M_i(OH)_{n_i}}(M_i^{n_i+})}{c_t(M_i^{n_i+})} \quad (i=1, 2, \dots, p) \quad (1)$$

where $c_t(M_i^{n_i+})$ represents the total concentration of any metal cations in solution, $c_{M_i(OH)_{n_i}}(M_i^{n_i+})$ represents the concentration of metal cation ($M_i^{n_i+}$) used to form hydroxide precipitation ($M_i(OH)_{n_i}$). The concentration of metal cation ($M_i^{n_i+}$) that reacts with hydroxide ion (OH^-) to form hydroxide precipitation ($M_i(OH)_{n_i}$) is given by formula (2).

$$c_{M_i(OH)_{n_i}}(M_i^{n_i+}) = c_t(M_i^{n_i+}) - c(M_i^{n_i+}) - \sum_{g_{ij}=1}^{g_{ij}} c_{M_i(OH)_{g_{ij}}}(M_i^{n_i+}) \quad (2)$$

where $c(M_i^{n_i+})$ represents the concentration of residual free metal cation remaining in solution after metal cation

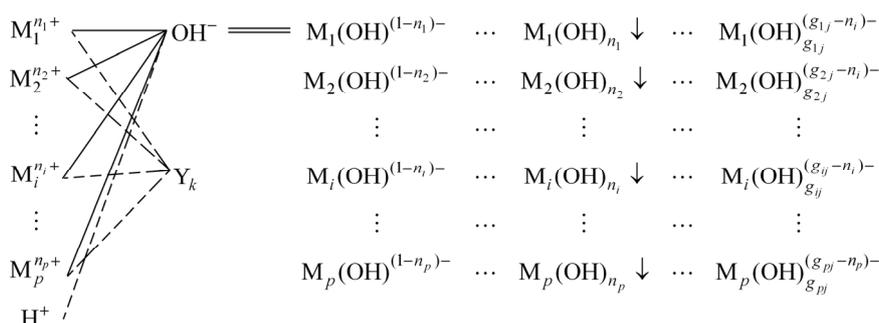


Fig. 1 Model of metal ion distribution when alkali is added to multi-component solution

($M_i^{n_i+}$) reacting with hydroxide ion (OH^-) to form precipitation and complex; g_{ij} represents the maximum coordination number of complex formed by metal cation ($M_i^{n_i+}$) and hydroxide ion (OH^-).

The concentration ($c_{M_i(OH)_{g_{ij}}}(M_i^{n_i+})$) of complex ($M_i(OH)_{g_{ij}}$) formed by metal cation ($M_i^{n_i+}$) and hydroxide ion (OH^-) is calculated by

$$c_{M_i(OH)_{g_{ij}}}(M_i^{n_i+}) = c(M_i^{n_i+})c(OH^-)\beta''_{M_i(OH)_{g_{ij}}} \quad (i=1, 2, \dots, p; g_{ij}=1, 2, \dots, g_{ij}) \quad (3)$$

where $c(OH^-)$ represents the concentration of residual hydroxide ion (OH^-) remaining in solution after metal cation ($M_i^{n_i+}$) reacting with hydroxide ion (OH^-) to form precipitation and complex; $\beta''_{M_i(OH)_{g_{ij}}}$ represents the conditional accumulative stability constants of complex ($M_i(OH)_{g_{ij}}$).

Deriving formula (1) to formula (3) gets formula (4):

$$G_i = 1 - \frac{c(M_i^{n_i+})(1 + \sum_{g_{ij}=1}^{g_{ij}} c(OH^-)^{g_{ij}} \beta''_{M_i(OH)_{g_{ij}}})}{c_t(M_i^{n_i+})} \quad (i=1, 2, \dots, p) \quad (4)$$

The concentration of residual metal cation remaining in solution after the reaction with hydroxide ion (OH^-) is expressed by conditional solubility product constant formula [1,4,16,17] as follows:

$$c(M_i^{n_i+}) = \frac{K''_{SP_{M_i(OH)_{n_i}}}}{(c(OH^-))^{n_i}} \quad (i=1, 2, \dots, p) \quad (5)$$

The sedimentation rate of any metal cations is obtained by substituting formula (5) into formula (1):

$$G_i = 1 - \frac{K''_{SP_{M_i(OH)_{n_i}}}(1 + \sum_{g_{ij}=1}^{g_{ij}} c(OH^-)^{g_{ij}} \beta''_{M_i(OH)_{g_{ij}}})}{c_t(M_i^{n_i+})c(OH^-)^{n_i}} \quad (i=1, 2, \dots, p) \quad (6)$$

The formation rate of complex ($M_i(OH)_{g_{ij}}$) is expressed by formula (7) as follows:

$$F_{i,j} = \frac{c_{M_i(OH)_{g_{ij}}}(M_i^{n_i+})}{c_t(M_i^{n_i+})} \quad (i=1, 2, \dots, p; g_{ij}=1, 2, \dots, g_{ij}) \quad (7)$$

Formulas (3) and (4) are deformed first and then substituted into formula (7), then,

$$F_{i,j} = \frac{(1-G_i)(c(OH^-))^{g_{ij}} \beta_{M_i(OH)_{g_{ij}}}''}{1 + \sum_{g_{ij}=1}^{g_{ij}} (c(OH^-))^{g_{ij}} \beta_{M_i(OH)_{g_{ij}}}''} \quad (i=1, 2, \dots, p; g_{ij}=1, 2, \dots, g_{ij}) \quad (8)$$

According to formulas (6) and (8), in order to get the sedimentation rate (G_i) of precipitate ($M_i(OH)_{n_i}$) and the formation rate of complex ($M_i(OH)_{g_{ij}}$), the concentration $c(OH^-)$ of residual hydroxide ion (OH^-) remaining in solution after metal cation ($M_i^{n_i+}$) reacting with hydroxide ion (OH^-) to form precipitation and complex must be obtained.

The total concentration of hydroxide ion (OH^-) in the reaction system was defined as $c_t(OH^-)$, then,

$$c_t(OH^-) = c(OH^-) + \sum_{i=1}^p c_{M_i(OH)_{n_i}}(OH^-) + \sum_{i=1}^p \sum_{g_{ij}=1}^{g_{ij}} c_{M_i(OH)_{g_{ij}}}(OH^-) \quad (9)$$

where $c_{M_i(OH)_{n_i}}(OH^-)$ represents the concentration of hydroxide ion (OH^-) that reacts with metal cation ($M_i^{n_i+}$) to form precipitate; $c_{M_i(OH)_{g_{ij}}}(OH^-)$ represents the concentration of hydroxide ion (OH^-) that reacts with metal cation ($M_i^{n_i+}$) to form complex.

According to formulas (1) and (7), $c_{M_i(OH)_{n_i}}(OH^-)$ and $c_{M_i(OH)_{g_{ij}}}(OH^-)$ can be expressed as

$$c_{M_i(OH)_{n_i}}(OH^-) = n_i c_{M_i(OH)_{n_i}}(M_i^{n_i+}) = n_i(G_i c_t)(M_i^{n_i+}) \quad (i=1, 2, \dots, p) \quad (10)$$

$$c_{M_i(OH)_{g_{ij}}}(OH^-) = g_{ij} c_{M_i(OH)_{g_{ij}}}(M_i^{n_i+}) = g_{ij}(F_{i,j} c_t)(M_i^{n_i+}) \quad (i=1, 2, \dots, p; g_{ij}=1, 2, \dots, g_{ij}) \quad (11)$$

Formulas (6), (8), (10) and (11) are substituted into formula (9), then,

$$c_t(OH^-) - c(OH^-) - \sum_{i=1}^p n_i \left[c_{M_i} - K_{sp_{M_i(OH)_{n_i}}}'' (1 + \sum_{g_{ij}=1}^{g_{ij}} (c(OH^-))^{g_{ij}} \beta_{M_i(OH)_{g_{ij}}}'') / (c(OH^-))^{n_i} \right] = 0$$

$$\sum_{i=1}^p \sum_{g_{ij}=1}^{g_{ij}} g_{ij} [K_{sp_{M_i(OH)_{n_i}}}'' (c(OH^-))^{g_{ij}-n_i} \beta_{M_i(OH)_{g_{ij}}}''] = 0 \quad (12)$$

4 Result and discussion

Solution components and their concentrations used in the experiment are listed in Table 1. The water for the experiment was deionized water.

Table 1 Components and concentrations of mixed solution

$c(Zn^{2+})/$ (mmol·L ⁻¹)	$c(Fe^{3+})/$ (mmol·L ⁻¹)	$c(Al^{3+})/$ (mmol·L ⁻¹)	$c(Cu^{2+})/$ (mmol·L ⁻¹)	$c(Mg^{2+})/$ (mmol·L ⁻¹)	pH
291	96.9	28.9	9.34	19.7	1.16

The precipitation rates of zinc, iron, aluminum, copper and magnesium were determined as follows. 0.5 L solution of metal ions was prepared firstly and its component is listed in Table 1. Then 1.06 mol/L sodium hydroxide solution was added and stirred for 30 min. After that the mixture was placed and held for 2 h. When the precipitation process was finished, the concentrations of metallic cations remaining in the solution were determined by flame atomic absorption spectrophotometry (AAS). The instruments used in AAS are as follows: WFX1F2B2 atomic absorption spectrophotometer (Beijing Second Optical Instrument Factory), XWT-S desk type recorder (Shanghai Automation Instrumentation Third Factory), magnesium, iron, zinc, aluminum, copper element hollow cathode lamp (Shanghai Optoelectronic Components Factory).

When alkali was added to the experimental solution whose components are shown in Table 1. The effects of alkaline quantities on the distribution of metal cation in the solution and the pH change are shown in Fig. 2. The data about the solubility product of metal hydroxides precipitation and cumulative stability constant of metal hydroxide complex are from literatures [1,5,14–16]. From Fig. 2(a), the zinc ion (Zn^{2+}) transformed into precipitate ($Zn(OH)_2$) gradually with the increase of $c_t(OH^-)$. When $c_t(OH^-)$ was 0.974 mol/L, the sedimentation rate could surpass 99%. When $c_t(OH^-)$ was higher than 1.03 mol/L, precipitate transformed into complex ion ($Zn(OH)_4^{2-}$). When $c_t(OH^-)$ was 1.76 mol/L, the sedimentation rate reduced to below 1.13%, the formation rate of complex ion ($Zn(OH)_4^{2-}$) could surpass 98.6%, the formation rate of complex ion ($Zn(OH)_3^-$) was as low as 0.21%, and complex ion ($Zn(OH)^+$) almost did not exist.

Figure 2(b) shows that Fe^{3+} existed in the form of metal cation Fe^{3+} (about 81%) and complex ion $Fe(OH)^{2+}$ (about 19%) in the solution first. After alkali

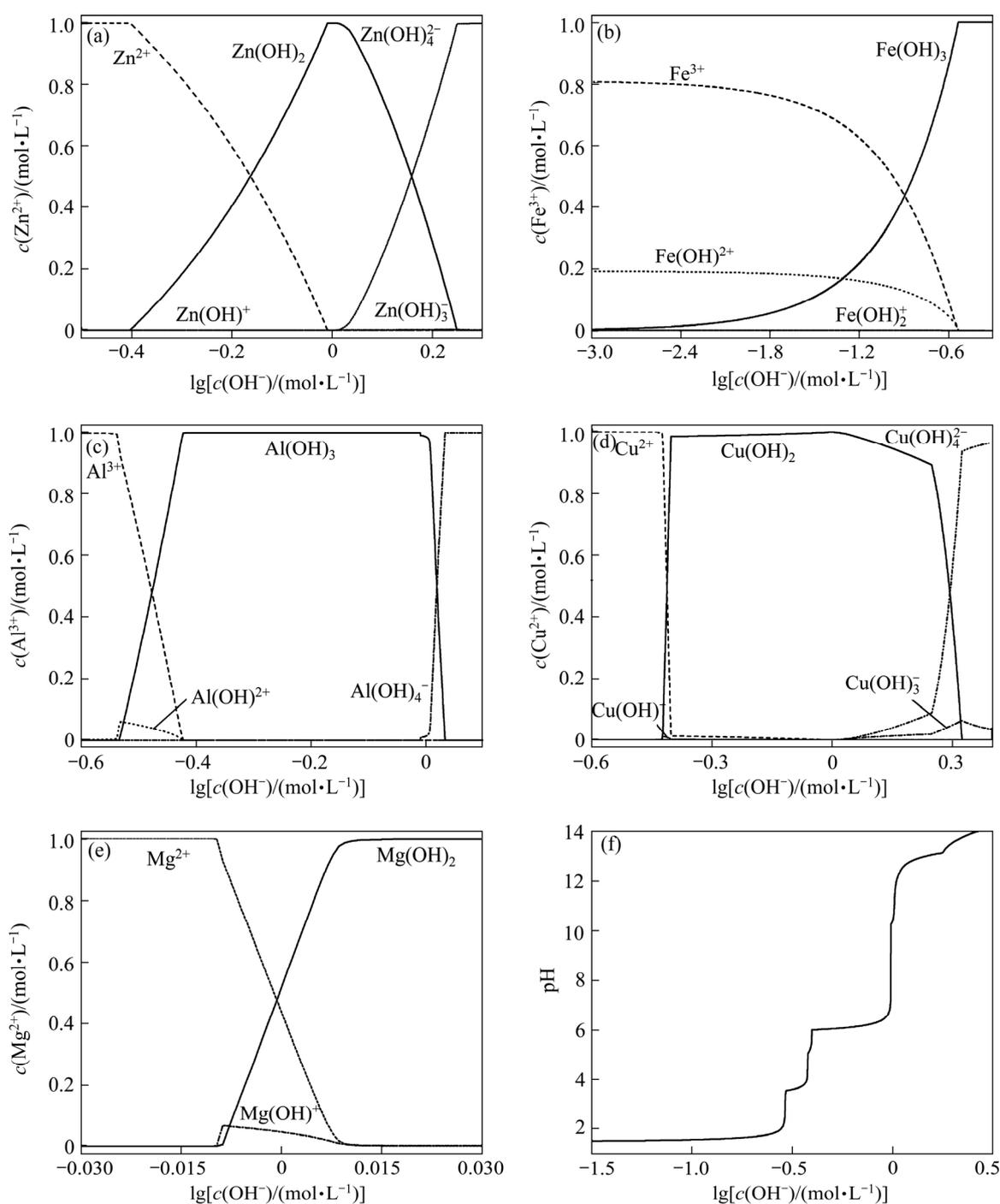


Fig. 2 Influence of total alkali concentration on distribution of Zn^{2+} (a), Fe^{3+} (b), Al^{3+} (c), Cu^{2+} (d), Mg^{2+} (e) and pH (f)

was added to solution, Fe^{3+} formed precipitate $Fe(OH)_3$ gradually, while complex ion ($Fe(OH)_2^+$) almost did not exist. When the concentration of alkali reached above 0.292 mol/L, the sedimentation rate could surpass 99.99%.

From Fig. 2(c), aluminium ion (Al^{3+}) transformed into complex ion ($Al(OH)_2^+$) and precipitated ($Al(OH)_3$) gradually with the increase of $c_t(OH^-)$. But the formation rate of complex ion ($Al(OH)_2^+$) was not large, even when $c_t(OH^-)$ was 0.294 mol/L, it reached 6.20%. When

$c_t(OH^-)$ increased continuously, the formation rate of complex ion ($Al(OH)_2^+$) reduced and sedimentation rate increased. When $c_t(OH^-)$ was 0.378 mol/L, the sedimentation rate could surpass 99.99%. When $c_t(OH^-)$ increased further, precipitate began to dissolve and form complex ion ($Al(OH)_4^-$). When the alkali concentration reached 1.08 mol/L, precipitate was completely dissolved in the form of complex ion ($Al(OH)_4^-$).

Figure 2(d) shows that copper ion (Cu^{2+}) transformed into complex ion ($Cu(OH)^+$) and precipitate

(Cu(OH)₂) gradually with the increase of $c_t(\text{OH}^-)$. But the formation rate of complex ion (Cu(OH)⁺) was not large, even when $c_t(\text{OH}^-)$ was 0.376 mol/L, it reached 1.47%. When $c_t(\text{OH}^-)$ increased continuously, the formation rate of complex ion (Cu(OH)⁺) reduced and sedimentation rate increased. When $c_t(\text{OH}^-)$ was 0.570 mol/L, the sedimentation rate could surpass 99.99%. When $c_t(\text{OH}^-)$ increased further, precipitate began to dissolve and form complex ion (Cu(OH)₃⁻) and complex ion (Cu(OH)₄²⁻). But the formation rate of complex ion (Cu(OH)₃⁻) was not large, even when $c_t(\text{OH}^-)$ was 2.11 mol/L, it reached 6.38%. With the increase of $c_t(\text{OH}^-)$, the formation rate of complex ion (Cu(OH)₄²⁻) increased, when $c_t(\text{OH}^-)$ was 2.00 mol/L, it could reach 55.60%.

From Fig. 2(e), magnesium ion (Mg²⁺) transformed into complex ion (Mg(OH)⁺) gradually with the increase of $c_t(\text{OH}^-)$. When $c_t(\text{OH}^-)$ was 0.980 mol/L, the formation rate of complex ion (Mg(OH)⁺) reached 6.56%. When $c_t(\text{OH}^-)$ increased continuously, the formation rate of complex ion (Mg(OH)⁺) reduced and sedimentation rate increased. When $c_t(\text{OH}^-)$ was 1.02 mol/L, the sedimentation rate of Mg(OH)₂ could surpass 99%.

Figure 2(f) shows that when alkali was added to solution, pH of the solution varied complicatedly. With the increase of $c_t(\text{OH}^-)$, pH increased slowly. When $c_t(\text{OH}^-)$ was 0.282 mol/L, pH reached 2.07, which was 0.91 higher than the initial pH 1.16. Then $c_t(\text{OH}^-)$ increased, pH increased rapidly. When $c_t(\text{OH}^-)$ was 0.294 mol/L, pH reached 3.56. When $c_t(\text{OH}^-)$ was 0.370 mol/L, pH increased slowly (pH=3.92). When $c_t(\text{OH}^-)$ increased continuously, pH increased rapidly. As $c_t(\text{OH}^-)$ was 0.370 mol/L, pH reached 6.02. As $c_t(\text{OH}^-)$ increased, pH increased gradually. When $c_t(\text{OH}^-)$ was 0.974 mol/L,

pH reached 7.09. When $c_t(\text{OH}^-)$ increased continuously, pH increased rapidly. As $c_t(\text{OH}^-)$ was 1.04 mol/L, pH reached 11.90. At this point, when $c_t(\text{OH}^-)$ was 9.84 mol/L (pH=10.3), a inflection point occurred. When $c_t(\text{OH}^-)$ increased, pH increased faster. As $c_t(\text{OH}^-)$ was 2 mol/L, pH reached 13.56.

The inflection points in some curves in Fig. 2 were related to the formation of precipitate and complex ion by the reaction of metal cation and hydroxide ion. Therefore, with the increase of $c_t(\text{OH}^-)$, if the concentration of any component of the solution changed, the existence form (sedimentation rate and formation rate of complex ion) of other components and the value of pH changed.

The reason that the distribution of metal cation in chemical reaction system containing both hydroxide precipitate and complex ion is that the ability (solubility product of hydroxide precipitate and cumulative stability constant of hydroxide complex ion) of forming precipitate and complex ion by the reaction of metal cation and hydroxide ion and their concentration is different. That is to say, the formation amount of precipitate and complex ion changes with the change of species and concentration of metal cation.

When alkali is added to solution, the sedimentation rate of Zn²⁺, Fe³⁺, Al³⁺, Cu²⁺, Mg²⁺ and pH change are calculated. The comparison of calculated values and experimental values is listed in Table 2. The comparative result proves the reliability of the proposed formula.

Based on the above formula, the maximum $c_t(\text{OH}^-)$ corresponding to the sedimentation of metal cation and pH of the solution can be obtained when alkali is added to multi-component solution containing some metal cations.

Table 2 Comparison of calculation result and experimental values of precipitation rates for Zn²⁺, Fe³⁺, Al³⁺, Cu²⁺, Mg²⁺ and pH

$c_t(\text{OH}^-)/(\text{mol}\cdot\text{L}^{-1})$	Method	G/%					pH
		Zn ²⁺	Fe ³⁺	Al ³⁺	Cu ²⁺	Mg ²⁺	
0.280	Experiment	–	96.15	–	–	–	1.97
	Calculation	0	96.30	0	0	0	2.03
0.362	Experiment	–	99.59	78.46	–	–	3.47
	Calculation	0	99.99	81.58	0	0	3.81
0.394	Experiment	–	99.94	96.38	89.24	–	5.14
	Calculation	0	100	99.99	88.67	0	5.55
0.630	Experiment	38.17	99.96	99.91	98.25	–	5.97
	Calculation	40.22	100	100	99.14	0	6.13
0.954	Experiment	94.32	99.99	99.99	99.94	–	6.73
	Calculation	95.87	100	100	99.92	0	6.71
1.022	Experiment	98.86	99.99	93.71	99.98	94.28	11.6
	Calculation	99.98	100	90.45	99.97	98.64	311.31

Temperature 18.9–20.3 °C

5 Conclusions

1) The distribution of metal cation in multi-component hydroxide reaction system is well described and the influence of several factors (the amount of alkali, cation concentration) on sedimentation rate and formation rate of complex ion is investigated quantitatively.

2) The distribution of metal cation and pH change are complicated when alkali is added to multi-component solution containing Zn^{2+} , Fe^{3+} , Al^{3+} , Cu^{2+} and Mg^{2+} .

3) The calculated values based on proposed formulas are in accordance with the experimental values significantly.

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氢氧化物反应体系中离子分布的计算方法

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摘要:提出一种计算公式,定量地计算同时形成氢氧化物沉淀和氢氧配合物的化学反应体系中金属离子的分布,考察影响氢氧化物沉淀和氢氧配合物形成的各种因素;基于提出的公式,计算锌、铁、铝、铜和镁的沉淀率,理论计算与实验结果一致。根据提出的计算公式,可以很好地描述在任意金属离子同时存在的氢氧化物反应体系中金属离子的存在形态,考察影响金属离子分布的因素。

关键词:氢氧化物;金属离子分布;沉淀率;配合物形成率

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