Studies on the Ammonia Pressure Leaching of Metallic Nickel and Copper Powders

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In the current investigation, the difference in dissolution behaviour between metallic nickel and copper powders under the condition of ammonia pressure leaching has been studied. The leaching rates were determined at the NH_3 concentration of 1 or 1.5 mol/L and over the $(\text{NH}_4)_2\text{SO}_4$ concentration range from 0 to 1.5 mol/L, the temperature range from 15° to 200°C, the agitation range from 400 to 1200 rev/min and oxygen partial pressure range from 0.5 to 10 atmospheres. The solubility of nickel ammine sulphate in the $\text{H}_2\text{O}-\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$ system was also measured in the present work. The experimental results obtained under these conditions are summarized as follows. The leaching of the nickel powder is shown to be a reaction of the zero-th order, indicating that the leaching time curve is a linear relation. But the leaching of the copper powder is an autocatalytic reaction, showing an extended S-shaped curve with a certain incubation period. The average leaching rate of the nickel powder is directly proportional to the oxygen partial pressure, and that of the copper powder to the square root of the oxygen partial pressure. The leaching amount of the nickel powder in solutions containing both ammonia and ammonium sulphate in the presence of oxygen is very small compared with that of the copper powder. This fact is independent of the difference in solubility between the $\text{Ni}(\text{NH}_3)_6\text{SO}_4$ and $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ in the $\text{H}_2\text{O}-\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$ systems.

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I. Introduction

It has been known since 1858 that metallic copper dissolves as an ammine complex ion in aqueous ammonia in the presence of oxygen. The dissolution reaction was applied to the recovery of metallic copper from ores⁽¹⁾⁽²⁾. A similar application was made to metallic nickel and cobalt⁽³⁾. The effect of oxygen partial pressure on the dissolution rate of copper in aqueous ammonia has been investigated by Halpern⁽⁴⁾, as shown in Fig.1. Namely, at a low oxygen pressure, the rate is directly proportional to the oxygen pressure and is independent of the ammonia concentration. As the oxygen pressure is increased, the rate becomes independent of oxygen pressure and levels off at a value

Fig. 1 Dissolution rate of copper in aqueous ammonia at 26°C (the data obtained from Halpern⁽⁴⁾).

which depends on the ammonia concentration.

At the present time, the two mechanisms, the autocatalytic theory and the oxide-film theory, have been proposed with regard to the dissolution reaction of copper in aqueous ammonia in the presence of oxygen. According to the autocatalytic theory, the reaction process consists of the following (1)~(3) steps.

Thus, the equilibrium between $\text{Cu}(\text{NH}_3)_2^+$ and $\text{Cu}(\text{NH}_3)_4^{2+}$ ions acts as a catalysis. This mechanism has been supported by Yamasaki⁽⁵⁾ and Lu et al.⁽⁶⁾ On the other hand, according to the oxide-film theory, the following steps (4)~(6) have been reported as the dissolution process of copper in aqueous ammonia in the presence of oxygen.

where $\text{Cu} \cdots \text{O}$ indicates the adsorption of dissolved oxygen at the copper surface and $\text{Cu}^{2+} \begin{matrix} \text{NH}_3 \\ \text{O}^{2-} \end{matrix}$ the acti-

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(4) J. Halpern : J. Electrochem. Soc., 100 (1953), 421.

(5) E. Yamasaki : Sci. Rep. Tohoku Imp. Univ. Japan, Ser. 1, 9 (1920), 169.

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vated complex of copper. Eq. (5) is the rate-determining step supported by Halpern et al.^{(7)~(9)} Besides, a new interpretation of the mechanism has also been proposed by Habashi⁽¹⁰⁾.

It is generally known that nickel as well as copper dissolves in aqueous ammonia in the presence of oxygen and forms ammine nickel complex ions. In order to clarify the dissolution kinetics of nickel in aqueous ammonia in the presence of oxygen, the present authors conducted experiments with nickel plates used as samples. However, it was found that the nickel plates became passive as the oxygen partial pressure was increased, and also that the nickel powder could dissolve in an ammonia solution containing dissolved oxygen even under the experimental condition in which the nickel plates became passive. Therefore, as reported previously⁽¹¹⁾, the effects of the ammonia concentration, the oxygen partial pressure and other factors on the dissolution rate of the nickel powder in a solution containing both ammonia and ammonium sulphate in the presence of oxygen were measured under the condition of ammonia pressure leaching. However, since the accurate surface area of the nickel powder is unknown, the experimental results obtained cannot be compared with the data in Fig. 1 which are obtained by the use of a copper plate of the known surface area. For this reason, some experiments on the ammonia pressure leaching of the copper powder were carried out to determine the difference in dissolution behaviour between metallic nickel and copper powders under the condition of ammonia pressure leaching. The solubility of nickel ammine sulphate in the $\text{NH}_3\text{--}(\text{NH}_4)_2\text{SO}_4\text{--H}_2\text{O}$ system was also measured in the present work and compared with those reported on copper ammine sulphate. Based on the results obtained, the authors discussed whether the dissolution reaction of the copper powder in the ammonia solution containing ammonium sulphate in the presence of oxygen proceeds autocatalytically or not.

II. Experimental Procedure

The electrolytic nickel powder (100~325 mesh) and copper powder (−150 mesh) were used as samples. The leaching experiment was carried out by an autoclave which was constructed of stainless steel and had a volume capacity of 1 liter. The autoclave could endure the pressure up to 200 atm. and was equipped with a magnetic stirrer. The stirring rate could easily be adjusted between 400 and 1400 rev/min. A solution volume of 500 milliliter and a sample weighing 10 grams were used in most of the measurements. The solution temperature in the autoclave was measured by means

of a mercury thermometer and regulated to within $\pm 1^\circ\text{C}$. The oxygen pressure was controlled in consideration of the partial pressures of water and ammonia.

The experiment was carried out in the following way. After the sample and the leaching solution were put into the autoclave and sealed, the dissolved oxygen in the solution was replaced by nitrogen gas. The solution temperature was then heated to a desired temperature by an electric furnace and a constant oxygen gas was led into the autoclave. The course of the reaction was determined from the concentration of metal ions in the solution taken at the measured time intervals from the autoclave. Nickel and copper ions were analyzed by dimethylglyoxime and ammine methods using the Hiram's spectrophotometer.

III. Experimental Results

Since the dissolution rate of the copper powder in the ammonia solution containing oxygen was much faster than that of the nickel powder, the measurements could not be carried out under the same experimental condition. For this reason, the dissolution behaviour of the nickel powder under the condition of ammonia pressure leaching cannot be quantitatively compared with that of the copper powder, but the essential difference in dissolution behaviour between the nickel powder and the copper powder may be distinguished from the experimental results obtained.

1. Leaching time curves of nickel and copper powders in aqueous ammonia under oxygen atmosphere

Some typical leaching curves of nickel and copper powders in aqueous ammonia are shown in Fig. 2. These dissolution reactions can be given by eqs. (7) and (8), respectively.

According to the results obtained, it was found that the amount of the copper powder dissolved was much larger than that of the nickel powder. For example, the amount of the copper powder dissolved in 1.5M NH_3 solution at room temperature in 60 minutes was found to be about 10%, while that of the nickel powder in 9M NH_3 solution at 100°C only 3% in 60 minutes. Furthermore, the following distinct characteristic was observed on the leaching time curve as shown in Fig. 2. That is, at temperatures below 92°C , the dissolution of the nickel powder rapidly increases at the early stages of the dissolution reaction and then levels off to a definite value after 20 minutes. On the contrary, at temperatures above 92°C , the dissolution of the nickel powder rapidly increases in the initial period of the dissolution reaction; a maximal dissolution is attained in about 2 minutes or less and then rapidly falls on the final dissolution rate. These phenomena may be due to the fact that ammine nickel complex ions formed in aqueous ammonia are decomposed into nickel hydroxide.

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Fig. 2 Leaching curves of nickel powder and copper powder in aqueous ammonia.

Leaching condition :	Ni	Cu
Oxygen partial pressure	2 atm	1 atm
Temperature	9~128°C	16~84°C
NH ₃ concentration	9 mol/L	1.5 mol/L
Stirring velocity	700 rev/min	500 rev/min
Sample weight	10 grams	10 grams

As shown in Fig. 2, the amount of ammine nickel complex ions remaining in the leaching solution without decomposition decreases with increasing leaching temperature. On the other hand, the dissolution behaviour of the copper powder in the same solution differs from that of the nickel powder. That is, ammine cupric complex ions produced by the dissolution of copper powder decompose even at room temperature into copper hydroxide after a definite leaching time. The higher leaching temperature, the earlier becomes the decomposition. For example, it begins after 25 minutes at room temperature, after 10 minutes at 50°C and after 5 minutes at 70°C.

These results indicate that the equilibria between ammine complex ions and hydroxides in aqueous ammonia, which are given by eqs. (9) and (10), shift to the right-hand direction with rising temperature.

In the case of the nickel powder, the reaction (9) proceeds to the right-hand direction at temperatures above 92°C, precipitating nickel hydroxides. In the case of copper powder, however, the reaction (10) proceeds to the right-hand direction even at room temperature, precipitating copper hydroxides. Therefore, the effect of temperature on the apparent leaching degrees of the nickel and copper powders after 60 minutes is shown in Fig. 3. From these results, the leaching measurements of the nickel and copper powders were carried out at 100°C and room temperature, respectively.

2. Leaching curves of nickel and copper powders in solutions containing both ammonia and ammonium sulphate

The leaching degrees of the nickel and copper powders in solutions containing ammonia only are very

small as described in the preceding section. However, it is expected that the leaching degree increases greatly, if OH⁻ ions produced with the dissolution reactions (7) and (8) are neutralized by the addition of NH₄⁺ ions in accordance with the following eq. (11) and (12)⁽¹¹⁾:

In the present experiment, ammonium ions are added as ammonium sulphate. The leaching curves of the nickel and copper powders in solutions containing both ammonia and ammonium sulphate are shown in Fig. 4, indicating the increase in the leaching degree with increasing ammonium sulphate as was expected. In solutions containing ammonium sulphate insufficiently, there occurs a spontaneous formation of yellow-greenish precipitation of NiSO₄·8Ni(OH)₂ or greenish precipitation of CuSO₄·3Cu(OH)₂, when the solution pH exceeds a definite value with the advance of leaching. This is the reason why the leaching curve becomes flat as if the dissolution ceases, although the leaching reaction of the sample continuously proceed by the ammonia isolated with the decomposition of ammine complex ions. No precipitation was observed in the leaching solution containing a sufficient amount of ammonium sulphate. However, the initial rate of dissolution reaction is retarded with increasing ammonium sulphate because of the decrease of oxygen solubility in a solution of ammonium salt. It was further observed that the following two facts are the essential differences in dissolution behaviour between nickel powder and copper powder under the condition of ammonia pressure leaching.

(1) Although the leaching condition of the copper powder is less severe than that of the nickel powder, the copper powder dissolves far greater than the nickel

Fig. 3 Effect of temperature on the apparent leaching percentages of nickel powder and copper powder dissolved for 60 minutes in aqueous ammonia, respectively.

Leaching condition :	Ni	Cu
Oxygen partial pressure	2 atm	1 atm
Temperature	9°~144°C	16°~84°C
NH ₃ concentration	9 mol/L	1.5 mol/L
Stirring velocity	700 rev/min	500 rev/min
Sample weight	10 grams	10 grams

Fig. 4 Leaching curves of nickel powder and copper powder in NH_3 -(NH_4)₂ SO_4 aqueous solution.

Leaching condition :	Ni	Cu
Oxygen partial pressure	2 atm	1 atm
Temperature	100°C	17°C
NH ₃ concentration	3 mol/L	1 mol/L
Sample weight	10 grams	10 grams

powder, i. e., in a solution containing 1M (NH_4)₂ SO_4 , the dissolution of the copper powder is found to be about 90% in 30 minutes, while that of the nickel powder about 60% in 120 minutes.

(2) In solutions containing ammonium sulphate sufficiently, the leaching curve of the nickel powder indicates a linear increase, and that of the copper powder an extended S-shaped one with a certain incubation period.

3. Effect of temperature on the leaching degrees

The temperature dependence of leaching degrees of the nickel and copper powders in the NH_3 -(NH_4)₂ SO_4 aqueous solution is shown in Fig. 5. The amount of leaching of the nickel powder indicates a linear increase

with time at each temperature. The slope of this straight line increases with increasing temperature. Thus the leaching reaction of the nickel powder is found to be one of the zero-th order, independent of the concentrations of ammonia and ammonium sulphate. It may also be expected that the reduction in the size of nickel particles with the advance of the leaching reaction, i.e. a decrease in the effective surface area of the nickel powder, is compensated by an increase in the surface roughness of the nickel powder which gradually increases with the progress of the reaction⁽¹²⁾. Therefore, the actual surface area of the nickel powder in the leaching solution remains almost constant during the leaching reaction. On the other hand, the leaching

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curve of the copper powder shows a complex shape and consists of the following three stages; the initial stage with some incubation period, the second stage during which the leaching rate relatively remains constant, and the final stage during which the rate falls off with the leaching time. That is, the leaching time curve of the copper powder shows an extended S-shaped curve with an incubation period and the degree of the change in each stage of the leaching curve becomes larger with increasing temperature. At temperatures higher than 150°C, the decomposition of ammine cupric complex ions described in the preceding section are observed.

The apparent activation energies for the initial leaching rate of the nickel powder and the leaching rate in the second stage of the leaching curve of the copper powder, calculated from the Arrhenius plots, are nearly the same values, 1916 cal/mol and 1902 cal/mol, respectively.

4. Effect of stirring velocity on the leaching degrees

The effect of stirring velocity on the leaching degrees of nickel and copper powders under the condition of ammonia pressure leaching is measured by varying the

Fig. 5 Effect of temperature on the leaching percentages of nickel powder and copper powder in $\text{NH}_3\text{-(NH}_4)_2\text{SO}_4$ aqueous solution.

Leaching condition :

Oxygen partial pressure
 NH_3 concentration
 $(\text{NH}_4)_2\text{SO}_4$ concentration
 Stirring velocity
 Sample weight

Ni
 2 atm
 3 mol/L
 0.75 mol/L
 700 rev/min
 10 grams

Cu
 1 atm
 1 mol/L
 0.5 mol/L
 500 rev/min
 10 grams

Fig. 6 Effect of stirring velocity on the leaching percentages of nickel powder and copper powder in $\text{NH}_3\text{-(NH}_4)_2\text{SO}_4$ aqueous solution

Leaching condition :

Oxygen partial pressure
 Temperature
 NH_3 concentration
 $(\text{NH}_4)_2\text{SO}_4$ concentration
 Sample weight

Ni
 2 atm
 100°C
 1.5 mol/L
 0.75 mol/L
 10 grams

Cu
 1 atm
 15°C
 1 mol/L
 0.5 mol/L
 10 grams

revolution number of a stirring apparatus attached to the autoclave. The experimental results obtained are shown in Fig. 6. Irrespective of the stirring velocity, it is evident that the leaching time curves of the nickel powder are given by a linear relation and those of the copper powder by an extended S-shaped curve with a certain incubation period. However, as shown in Fig. 6, the incubation period in the leaching of the copper powder is gradually shortened with increasing stirring velocity. The quantitative relationship between the leaching rate and the stirring velocity can not be determined since the powdered sample is in the state of a turbulent and circular motion together with the leaching solution in the autoclave.

5. Effect of the oxygen partial pressure on the leaching rates

The measurements were made at several oxygen partial pressures ranging from 0.5 to 10 atmospheres and the results obtained are shown in Fig. 7. It is understood from Fig. 7 that both the leaching degrees of nickel and copper powders increase with increasing oxygen partial pressure and the incubation period observed on the leaching curve of the copper powder is shortened as the oxygen partial pressure is raised. Further, a difference in dissolution behaviour between the nickel powder and the copper powder was also observed in the relation between the leaching rates and the oxygen partial pressure, as shown in Fig. 8. The average

Fig. 7 Effect of oxygen partial pressure on the leaching percentages of nickel powder and copper powder in $\text{NH}_3\text{-(NH}_4)_2\text{SO}_4$ aqueous solution.

Leaching condition :		Ni	Cu
Temperature		100°C	17°C
NH_3 concentration		1.5 mol/L	1.0 mol/L
$(\text{NH}_4)_2\text{SO}_4$ concentration		0.75 mol/L	0.5 mol/L
Stirring velocity		700 rev/min	500 rev/min
Sample weight		10 grams	10 grams

Fig. 8 Effect of oxygen partial pressure on the leaching rates of nickel powder and copper powder in $\text{NH}_3\text{-(NH}_4)_2\text{SO}_4$ aqueous solution.

Leaching condition :		Ni	Cu
Temperature		100°C	17°C
NH_3 concentration		1.5 mol/L	1.0 mol/L
$(\text{NH}_4)_2\text{SO}_4$ concentration		0.75 mol/L	0.5 mol/L
Stirring velocity		700 rev/min	500 rev/min
Sample weight		10 grams	10 grams

leaching rate of the nickel powder increased in direct proportion to the oxygen partial pressure,

$$\text{or } -\frac{d[\text{Ni}]}{dt} = k_{\text{Ni}} \cdot P_{\text{O}_2},$$

but the leaching rate at the second stage of the leaching curve of the copper powder to the square root of the oxygen partial pressure,

$$\text{or } -\frac{d[\text{Ni}]}{dt} = k_{\text{Cu}} \cdot P_{\text{O}_2}^{1/2},$$

where $[\text{Ni}]$ and $[\text{Cu}]$ are the amounts of the nickel and copper powders dissolved in time t (min) (g/L), k_{Ni} and k_{Cu} rate constants and P_{O_2} the oxygen partial pressure (atm), respectively. These phenomena have also appeared in the case of the pressure leaching of sulfide ores. For example, the leaching rates of FeS_2 (pyrite)⁽¹³⁾ and ZnS ⁽¹⁴⁾ in H_2SO_4 solution, and of PbS ⁽¹⁵⁾ in NaOH solution are directly proportional to the oxygen partial pressure. On the contrary, the leaching rates of FeS ⁽¹³⁾, FeS_2 (marcasite)⁽¹³⁾, Cu_2S ⁽¹⁶⁾ and CuFeS_2 ⁽¹⁷⁾ in H_2SO_4 solution are proportional to the square root of the oxygen partial pressure. This suggests that the oxygen participates in the rate controlling step of the reaction either as a oxygen molecularly or as an atom dissociated. However, the reason why the difference in leaching behaviour reveals in the nickel and copper powders remains unknown.

6. The solubilities of nickel and copper ammine sulphates

The difference in the solubility between nickel ammine sulphate and copper ammine sulphate in the $\text{H}_2\text{O}-\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$ system may be considered to be one of the reasons why the amount of the copper powder dissolved in a solution containing both NH_3 and NH_4^+ ions is larger than that of the nickel powder. Therefore, the solubility of nickel ammine sulphate in the $\text{H}_2\text{O}-\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$ system was measured in the temperature range from 10° to 30°C. Nickel ammine sulphate, $\text{Ni}(\text{NH}_3)_6\text{SO}_4$, was prepared from the anhydrous nickel sulphate absorbed with ammonia gas at 30°C for 120 hours. The synthetic nickel ammine sulphate was a very unstable compound and easily decomposed into nickel sulphate in an air atmosphere. According to the result of measurement, when the $\text{Ni}(\text{NH}_3)_6\text{SO}_4$ powder was exposed in an air atmosphere for 5 minutes, the greater part of it was rapidly decomposed into $\text{Ni}(\text{NH}_3)_5\text{SO}_4 \cdot \text{H}_2\text{O}$. Then, the $\text{Ni}(\text{NH}_3)_6\text{SO}_4$ powder was completely decomposed into NiSO_4 powder after 196 hours in an air atmosphere. For these reasons, anhydrous nickel ammine sulphate powder synthesized was directly removed to the measuring cell from the apparatus of sample preparation.

The solubility of nickel ammine sulphate in the $\text{H}_2\text{O}-\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$ system measured in the present

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Fig. 9 Solubilities of $\text{Ni}(\text{NH}_3)_6\text{SO}_4$ (solid line) and $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ (interrupted line) in aqueous ammonia.

Fig. 10 Solubilities of $\text{Ni}(\text{NH}_3)_6\text{SO}_4$ (solid line) and $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ (interrupted line) in $(\text{NH}_4)_2\text{SO}_4$ aqueous solution.

Fig. 11 Solubilities of $\text{Ni}(\text{NH}_3)_6\text{SO}_4$ and $\text{Cu}(\text{NH}_3)_4\text{SO}_4 \cdot \text{H}_2\text{O}$ in $\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$ aqueous solution.

work is shown in Fig. 9, Fig. 10 and Fig. 11, respectively. In these figures, the solid lines represent the solubility of nickel ammine sulphate and the interrupted lines show the solubility of copper ammine sulphate which was determined by Okabe and Ito⁽¹⁸⁾. The solubilities of nickel and copper ammine sulphates in ammonia and ammonium sulphate solution in the presence of oxygen increase with rising temperature and decrease with increasing concentrations of ammonia and ammonium sulphate. Then the solubility of nickel ammine sulphate in the $\text{H}_2\text{O}-\text{NH}_3-(\text{NH}_4)_2\text{SO}_4$ system is about twice as large as that of copper ammine sulphate. From these results, it may be found that the difference between the leaching amount of nickel and copper powders is independent of the difference in the solubilities of nickel and copper ammine sulphates.

IV. Discussion

The leaching reaction of nickel powder in a solution containing both ammonia and ammonium sulphate may be considered to be of the zero-th order, because the leaching curve of the nickel powder is directly proportional to the leaching time. On the contrary, the leaching reaction of the copper powder in a solution containing both ammonia and ammonium sulphates may appear to be an autocatalytic one, because the leaching curve of the copper powder shows an extended S-shaped curve with an incubation period. The leaching reaction process of copper powder under the condition of the ammonia pressure leaching may be considered as follows. At the early stages of the leaching reaction, there is no complex ion in the leaching solution, but copper dissolved may easily form an ammine cupric complex ion with the progress of leaching reaction because both ammonia and oxygen are present sufficiently in the leaching solution. This reaction may be represented by eq. (12).

Then, the ammine cupric complex ion produced by eq. (12) reacts with metallic copper powder and is reduced to an ammine cuprous complex ion, as given by eq. (13).

Since both ammonia and ammonium ions are amply contained in the leaching solution, the ammine cuprous complex ion formed is again oxidized to the ammine cupric complex ion, as given by eq. (14).

Thus, the copper powder may dissolve in a solution containing both ammonia and ammonium sulphates in the presence of oxygen in accordance with eqs. (12) and (13). The reaction product, the ammine cupric complex ion plays a catalytic role in the leaching reaction and accelerates the leaching reaction of metallic copper powder in ammonia and ammonium sulphate solutions. From the facts described above, the leaching reaction of

metallic copper powder in a solution containing both ammonia and ammonium sulphates in the presence of oxygen can be given by eq. (15).

The explanation of the autocatalytic reaction of copper powder obtained in the present work, which is represented by eqs. (12), (13) and (14), may differ from that of the autocatalytic theory described in the introduction. That is, the concentration of the product caused by the leaching reaction of the copper powder is relative to the leaching rate equation under the condition of ammonia pressure leaching, since the reaction product, the ammine cupric complex ion plays the role of a catalysis and its amount increases with the leaching time.

If it is assumed that fine copper powder uniformly distributes in the leaching solution and its leaching reaction is considered to be a quasi-homogeneous one, the leaching rate equation of metallic copper powder in a solution containing both ammonia and ammonium sulphate can be given as follows;

$$\frac{dx}{dt} = k(a-x)(c+x)$$

where a is the initial concentration of copper powder, k a rate constant, x the amount of the copper powder dissolved in time t (min) and c the initial concentration of an ammine cupric complex ion. By integrating this equation, we can rewrite as

$$\frac{1}{a+c} \ln \frac{(c+x)}{(a-x)} = kt.$$

The relation between x and t is graphically shown in Fig. 12. Since c is equal to zero in the case of the leaching reaction of copper powder under the condition of ammonia pressure leaching, the leaching time curve of the copper powder must be consistent with the curve of $c=0$ in Fig. 12. This fact is in good agreement with the experimental results obtained in the present work. Consequently, the reasons why the leaching reaction of the copper powder in a solution both ammonia and ammonium sulphate shows an extended S-shaped curve with a certain incubation period and the leaching amount of the copper powder is considerably larger than that of the nickel powder, are demonstrated from the explanation described above.

Fig. 12 Reaction with the incubation period.

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V. Conclusion

The leaching behaviours of metallic nickel powder and copper powder under the condition of ammonia pressure leaching have been examined in the present work. From the experimental results obtained, the essential difference in leaching behaviour between metallic nickel and copper powders are phenomenally summarized as follows.

The leaching amount of the nickel powder in a solution containing both ammonia and ammonium sulphates in the presence of oxygen is very small compared with that of the copper powder. This fact is independent of the difference in solubility between nickel ammine sulphate and copper ammine sulphate in the H_2O –

NH_3 – $(\text{NH}_4)_2\text{SO}_4$ system. The leaching of the nickel powder is shown to be a zero-th order reaction, indicating that the leaching time curve is a linear relation, but that the leaching of the copper powder is an autocatalytic reaction, showing an extended S-shaped curve with a certain incubation period. The average leaching rate of the nickel powder is shown to be directly proportional to the oxygen partial pressure, but that of the copper powder to the square root of the oxygen partial pressure.

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