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Leaching of a Tailings from Oxy-sulfur Copper Ore with Sulfuric Acid

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Abstract. In this paper, mineralogy and leaching comprehensive utilization of tailings from an oxy-sulfide copper ore were studied. Tailings grade was 0.526%, of which copper was primary free copper oxide. Malachite was not completely dissolved although it flake aggregate size was much larger than other minerals. When the tailings were ground again to a fineness of -0.074 mm 93.5%, the copper grade in the tailings could only be reduced to 0.489% after one roughing and two scavenging. It was difficult to effectively recover copper with conventional flotation methods. Under the pulp concentration of 33%, the pulp temperature of 45 ℃ and 55 ℃, sulfuric acid dosage of 160 kg/t, leaching time of 0.5 h, immersion liquid contained copper 2.37 g/L, the leaching rate of 91.34% was obtained with acid leaching, which the copper tailings got effectively recycled.

1. Preface
Copper plays an important role in China's national economy and daily life and is an indispensable resource in production and life [1]. The tailings of an oxy-sulfur copper ore contained 0.526% of copper, which is much higher than the general natural grade of copper ore. Great waste of national important resources will be caused if the comprehensive utilization of copper ore is not carried out [2]. Zhai et al. [3] studied an oxide copper of abroad, the results showed that under the pretreatment conditions of the grinding fineness -74μm of 60%, pulp density of 35%, K1 concentration of 9%, temperature of 85 ℃, time of 1 h, acid leaching conditions of sulfuric acid of 25%, sulfuric acid concentration of 13.8%, temperature of 50 ℃, leaching time of 3 h, the copper leaching rate is 87.44%. Zhou et al.[4] studied an oxidized copper cobalt concentrate contained 5.75% copper and 0.34% cobalt, 78.96% oxidation rate with sulfuric acid as leaching agent. At the temperature of 50 ℃, with sulfuric acid and ore in the ratio of 0.3:1 and liquid solid ratio at 4:1, leaching time of 6 h, the copper and cobalt leaching rates of leach residue reaching 94.34% and 97.57%, respectively, and the leach solution contained 12.38 g/L copper, 0.73 g/L cobalt and trace of impurities of iron, manganese, magnesium, etc. Dai [5], Qiu [6], Wang [7], Xiong [8], Lv [9], et al. studied the leaching of different types of oxidized copper ore and obtained good indexes respectively. The object of this paper is to study the mineralogy and leaching comprehensive utilization of the tailings of an oxy-sulfide copper ore.
2. Ore properties

2.1 Phase analysis
Copper phase analysis results of tailings were shown in table 1.

| Phase            | Copper sulfide | Free copper oxide | Bonded copper oxide | Total copper |
|------------------|----------------|-------------------|---------------------|--------------|
| Contents (%)     | 0.0330         | 0.4835            | 0.0027              | 0.5192       |
| Distribution (%) | 6.36           | 93.12             | 0.52                | 100.00       |

It can be seen from table 1 that the primary copper in tailings was free copper oxide.

2.2 Tailings mineral composition
The tailings were prepared into sanding sheets which were then identified. The non-metallic minerals in sanding sheets were mostly fine granular with particle size between from 0.01 to 0.07 mm, a few of 0.07 to 0.1 mm, and some of greater 0.1 mm. Non-metallic minerals accounted for about 93%-94% and metallic minerals accounted for about 5%-7%. Malachite presented irregular flake or granular aggregate, which was mixed with limonite and other minerals to form round, elliptical and irregular pellets, and the particle size was mostly between 0.1-0.45 mm, with a small amount less than 0.1 mm. Goethite mostly presented in the form of small grains and their aggregates, the particle size was mostly between 0.01-0.05 mm, and the maximum was 0.15 mm, which mostly existed independently and partly wrapped in nonmetallic minerals. Chalcopyrite existed independently which were small and granular, with a particle size of about 0.01-0.02 mm. Chalcocite encased in nonmetallic minerals which were small granular, with a particle size of about 0.01-0.02 mm. The grain size of malachite flaky aggregate was much larger than that of other minerals in the thin sections, but the clay-like mineral with star point distribution could be seen, and malachite was not completely dissociation.

3. Experimental results and discussions

3.1 Exploration test of regrinding and re-separation for tailings
The regrinding and re-separation test of tailings was carried out according to the process and conditions in figure 1, and the test results were shown in table 2.
Figure 1. Flowchart of regrinding and re-separation for tailings.

Table 2. Results of regrinding and re-separation for tailings.

| Items       | Partly yield (%) | Grade (%) | Partly recovery (%) |
|-------------|------------------|-----------|--------------------|
| Concentrate | 0.69             | 2.82      | 3.70               |
| Middlings   | 1.38             | 1.99      | 5.22               |
| Tailings    | 97.93            | 0.489     | 91.08              |
| Total       | 100.00           | 0.526     | 100.00             |

It can be seen from table 2 that when the tailings are ground again to a fineness of -0.074 mm 93.5%, the copper content in the tailings can only be reduced to 0.489% through one roughing and two scavenging. The only way to reduce copper content in tailings was chemical methods.

3.2 Tailings acid leaching experiment

3.2.1 Sulfuric acid dosage of leaching test at room temperature
The leaching test of sulfuric acid dosage at room temperature was carried out as the procedure shown in figure 2. The leaching time was 1 hour and the test results were shown in table 3.
Figure 2. Flowchart of leaching test of sulfuric acid dosage at room temperature.

Table 3. Results of leaching test of sulfuric acid dosage at room temperature.

| Sulfuric acid dosage (kg/t) | Items            | Weight or volume (g or L) | Copper grade or g/L (%) | Recovery (%) |
|---------------------------|------------------|---------------------------|-------------------------|--------------|
| 40                        | Leaching solution| 1.0                       | 0.402                   | 15.49        |
|                           | Leaching residue | 470.5                     | 0.466                   | 84.51        |
|                           | Feed             | 500                       | 0.519                   | 100.00       |
| 60                        | Leaching solution| 1.0                       | 0.489                   | 18.84        |
|                           | Leaching residue | 461                       | 0.457                   | 81.16        |
|                           | Feed             | 500                       | 0.519                   | 100.00       |
| 80                        | Leaching solution| 1.0                       | 0.852                   | 32.82        |
|                           | Leaching residue | 462.5                     | 0.377                   | 67.18        |
|                           | Feed             | 500                       | 0.519                   | 100.00       |
| 100                       | Leaching solution| 1.0                       | 1.38                    | 53.11        |
|                           | Leaching residue | 453                       | 0.269                   | 46.89        |
|                           | Feed             | 500                       | 0.519                   | 100.00       |

It can be seen from table 3 that when the amount of sulfuric acid is 40 kg/t, the leaching rate of copper is extremely low. The leaching rate of copper increase gradually with the increase of the amount of sulfuric acid. When the amount of sulfuric acid increase to 100 kg/t, the leaching rate of copper is only 53.11%, indicated that the amount of sulfuric acid would be increased.
3.2.2 Leaching experiment of sulfuric acid in warming

In the pulp temperature of 45 °C and 55 °C, leaching time was 1 hour, the procedure as shown in figure 2 and the test results were shown in table 4.

Table 4. Results of leaching experiment of sulfuric acid dosage in warming.

| Sulfuric acid dosage (kg/t) | Items           | Weight or volume (g or L) | Copper grade (% or g/L) | Recovery (%) |
|----------------------------|-----------------|---------------------------|-------------------------|--------------|
| 80                         | Leaching solution | 1.0                       | 0.863                   | 33.27        |
|                            | Leaching residue | 452                       | 0.383                   | 66.73        |
|                            | Feed            | 500                       | 0.519                   | 100.00       |
| 100                        | Leaching solution | 1.0                       | 1.90                    | 73.23        |
|                            | Leaching residue | 448                       | 0.155                   | 26.77        |
|                            | Feed            | 500                       | 0.519                   | 100.00       |
| 130                        | Leaching solution | 1.0                       | 1.96                    | 75.51        |
|                            | Leaching residue | 438.5                     | 0.145                   | 24.49        |
|                            | Feed            | 500                       | 0.519                   | 100.00       |
| 160                        | Leaching solution | 1.0                       | 2.38                    | 91.71        |
|                            | Leaching residue | 418.5                     | 0.0514                  | 8.29         |
|                            | Feed            | 500                       | 0.519                   | 100.00       |

Table 4 illustrates that the effect of leaching in warming is much better than that of room temperature. The rate of leaching in warming is 20.12% higher than that of normal temperature leaching when the amount of sulphuric acid also is 100 kg/t. When the pulp temperature is 45 °C-55 °C, the leaching rate of copper is 91.71% with 160 kg/t sulfuric acid dosage. Therefore, sulphuric acid dosage (160 kg/t) was selected for subsequent leaching test.

3.2.3 Leaching time test in warming

Test process were the same as shown in figure 2, pulp temperature was 45 °C-55 °C, dosage of sulfuric acid was 160 kg/t, changed the leaching time, the results listed in table 5.

It can be seen from table 5 that with the increase of leaching time, copper leaching rate shows an upward trend, but the amplitude is not large. When the leaching time increase from 0.5 hours to 2.0 hours, the difference between the highest and lowest leaching rates is 1.52 percentage points, the difference between the highest and the lowest copper content is 0.04 g/L in the leaching solution, and the difference between the highest and the lowest copper content is 0.0094 percentage points in the leaching residue. It could be said that the leaching time had little influence on the leaching effect in the time from 0.5 hours to 2.0 hours, and the leaching time was determined to 0.5 hours.
Table 5. Results of leaching time test in warming.

| Sulfuric acid dosage (kg/t) | Items              | Weight or volume (g or L) | Copper grade (% or g/L) | Recovery (%) |
|---------------------------|--------------------|---------------------------|-------------------------|--------------|
|                           | Leaching solution  | 1.0                       | 2.37                    | 91.34        |
|                           | Leaching residue   | 418.7                     | 0.0537                  | 8.66         |
| Feed                      |                    | 500                       | 0.519                   | 100.00       |
|                           | Leaching solution  | 1.0                       | 2.38                    | 91.71        |
|                           | Leaching residue   | 418.5                     | 0.0514                  | 8.29         |
| Feed                      |                    | 500                       | 0.519                   | 100.00       |
|                           | Leaching solution  | 1.0                       | 2.39                    | 92.10        |
|                           | Leaching residue   | 418.2                     | 0.0490                  | 7.90         |
| Feed                      |                    | 500                       | 0.519                   | 100.00       |
|                           | Leaching solution  | 1.0                       | 2.41                    | 92.86        |
|                           | Leaching residue   | 418.0                     | 0.0443                  | 7.14         |
| Feed                      |                    | 500                       | 0.519                   | 100.00       |

4. Conclusions

(1) The copper in flotation tailings is primary free copper oxide existed as malachite. Malachite schistose aggregates can be seen with a larger particle size than other minerals, and clay minerals can be seen with star point distribution in sanding sheets.

(2) When the tailings were ground again to a fineness of 93.5% -0.074 mm, the copper grade in the tailings could only be reduced to 0.489% through one roughing and two scavenging. The flotation test results illustrate conventional flotation methods recover copper ineffectively.

(3) The leaching rate of 91.34% was obtained with acid under the 33% pulp density, pulp temperature 45 °C-55 °C, 160 kg/t sulfuric acid dosage, leaching time 0.5 h. The copper in tailings could be recycled effectively by sulfuric acid leaching.

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