Study on the Dispersion Mechanism of Citric Acid on Chlorite in Hematite Reverse Flotation System

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Abstract: The adhesion behavior between particles in the pulp will seriously affect the index of concentrate. In this paper, the effect of citric acid on chlorite in hematite reverse flotation system was studied. The flotation test and settlement test of artificial mixed ore showed that a lower dosage of citric acid could significantly improve the removal rate of chlorite. The mineral contact angle measurement and zeta potential analysis, combined with the extended DLVO theory showed that chlorite is easily adsorbed on the surface of hematite, results in a lower removal rate, and the phenomenon was analyzed by a laser particle size analyzer and optical microscope. The results showed that citric acid can reduce the apparent grain size of artificial mixed ore and improve the dispersion degree of pulp by changing the wettability and surface potential of the minerals, thus increased the removal rate of chlorite.

Keywords: hematite; chlorite; citric acid; dispersion mechanism; reverse flotation

1. Introduction

With the increasing exploitation of mineral resources, the content of silicate minerals such as chlorite in hematite ore gradually increases and the size of hematite particles become finer. In order to achieve the full liberation of different hematite particles, the ore needs to be finely ground, which leads to serious slime of the chlorite. In addition, the adsorption behavior of fine particles is prone to occur due to their high surface reactivity and charge [1]. Fine-grained chlorite easily adheres on hematite surfaces in subsequent flotation operations, reducing the Fe grade in flotation concentrate. Therefore, it is important for flotation separation of different minerals to effectively eliminate the adsorption among mineral particles.

Previous studies have showed that dispersants can significantly improve the flotation performance of fine-grained ore [2]. Biopolymers, sodium hexametaphosphate and lignosulfonate have been used for the flotation of copper ores associated with kaolinite, bauxite and clay minerals, respectively [3–6]. Although chemical dispersants are widely used to separate the nonferrous metal minerals by flotation, they are rarely used for ferrous metal flotation.

Citric acid, as a dispersant, has been widely used in the flotation separation of silicate minerals, sulfide ores and some calcium carbonate minerals [7–11]. In the field of hematite flotation, the dispersing effect of citric acid on siderite has been investigated by some scholars [12,13], but its effect on chlorite in flotation of hematite has been rarely reported. In this paper, the effect of citric acid on separating chlorite from hematite ore was studied by artificial mixed ore flotation. Based on the extended DLVO theory, the settlement tests, zeta-potential measurements and particle size analyses were conducted to further reveal the dispersion mechanism of citric acid on chlorite in improving the quality of iron concentrate. Furthermore, the agglomeration and dispersion between chlorite and
hematite before and after adding citric acid were confirmed by optical microscope. Finally, the results provide a reference for effectively separating fine-grained minerals by flotation.

2. Materials and Methods

2.1. Materials and Reagents

2.1.1. Materials

The hematite and chlorite were taken from Anshan in Liaoning Province, China. Quartz was selected from Linqu in Shandong Province, China. High grade raw ores were crushed, ground and then purified by gravity methods. The purity of hematite and quartz was above 98%, and the purity of chlorite was more than 90%, which were proved by the X-ray Diffraction (XRD) method and multi-element chemical analysis. The above three kinds of pure minerals were sieved into four fractions for the implement of the following experiments. In this study, hematite was used in four size fractions of $-74 + 45 \, \mu m$, $-45 + 25 \, \mu m$, $-25 + 10 \, \mu m$, and $-10 \, \mu m$, and the quartz was selected at $-74 + 45 \, \mu m$, and the chlorite is selected at $-10 \, \mu m$.

2.1.2. Reagents

The sodium oleate (collector) and methyl isobutyl carbinol (MIBC, frother) that used in the experiments was chemically pure, and the purity of citric acid (dispersant), calcium chloride (activator), sodium hydroxide and hydrochloric acid (pH adjusters) were analytically pure, and the starch (depressant) was an industrial product. All reagents were prepared with deionized water, and the pH of deionized water was ca. 6.5.

2.2. Analyzing Methods

2.2.1. Artificial Mixed Ore Flotation Experiment

The artificial mixed mineral flotation test was carried out in a mechanical agitation flotation machine at a constant rate of 1100 r/min. The mineral suspension was prepared by adding 2.0 g of the minerals (which hematite: quartz: chlorite mass ratio of 5:3:2) to 25 mL deionized water, stirred for 2 min. The pH of the mineral suspension was adjusted to the desired value by adding NaOH or HCl and conditioning for 2 min; then, the appropriate amount of citric acid, starch, calcium chloride, sodium oleate and MIBC were added and conditioned for 2 min. Flotation was performed for 3 min. Both the floated and unfloated products were dried, weighed and analyzed. After that, the recovery rate of each minerals was calculated, respectively.

2.2.2. Settlement Test of Artificial Mixed Ore

The dispersion effect of citric acid on mineral particles in the pulp was investigated by using settlement tests. The specific steps were conducted as follows: 30.0 g artificial mixed ore was obtained, in which the mass ratio of hematite ($M_1$), quartz ($M_2$) and chlorite ($M_3$) was 5:3:2, add it to a 1000 mL volume separatory funnel and fill it with deionized water, with a speed of 600 r/min electric stirrer for 5 min to ensure that the pulp was dispersed evenly. Then, according to the flotation agent system, the pH regulator, dispersant, inhibitor, activator and collector were added. After standing for 1 min, open the separatory funnel and drain 500 mL. The quality of hematite ($m_1$), quartz ($m_2$) and chlorite ($m_3$) can be obtained by weighting and testing the content of elements. The dispersion degree of mineral particles in the pulp is related to the settlement rate ($\beta$), and the smaller the settlement rate, the better the dispersion of the corresponding mineral particles. The settlement rate of hematite ($\beta_1$), quartz ($\beta_2$) and chlorite ($\beta_3$) is expressed as follows:

$$\beta_{1,2,3} = \frac{m_{1,2,3}}{M_{1,2,3}} \times 100\%$$

(1)
2.2.3. Contact Angle and Zeta Potential Measurements

Contact angle measurement was carried out with a JC2000A contact angle analyzer (Powereach Instruments, Shanghai, China). Take 2.0 g fine granule sample into a beaker, followed by adding flotation agent and stirred for 2 min, and then dried in a vacuum oven at 40 °C. In addition, obtain a smooth sheet by a tablet press machine. During the measurement, a stable and suitable water about 2–3 mm in diameter was produced on the surface of the sample with a microsyringe. Each sample was repeated five times, and the average value was reported.

Zeta potentials were measured by a Malvern Instruments Nano-ZS90 zeta potential analyzer (Malvern Instruments, Malvern, UK) at room temperature. In accordance with the flotation reagent dosage, a pulp suspension containing 0.01 wt % -5 μm hematite (quartz/chlorite) was conditioned in a beaker for 10 min at a given pH. The zeta potentials at each condition were measured at least five times and the average value was calculated and reported.

Residues of chemicals in glassware can have a significant effect on the reproducibility of the contact angle and zeta potential, and therefore must ensure their cleanliness. At the beginning of each test, all the glassware and flotation columns were thoroughly rinsed with an ultrasonic cleaner, then rinsed with acetone, and finally rinsed with deionized water for at least 3 times.

2.2.4. Analysis of the Particle Size of Artificial Mixed Ore

Particle size was measured by a Malvern Mastersizer 3000 laser particle size analyzer (Malvern Instruments, Malvern, UK). Hematite and chlorite were mixed at a mass ratio of 1:1, then transferred it into a beaker and stirred it to ensure it mixed evenly. Particle size analysis was carried out by extracting 1 mL from the middle of the suspension. In this paper, the Sauter mean diameter d32 (it is defined as the diameter of a sphere that has the same volume/surface area ratio as a particle of interest) was used to indicate that, the larger the particle size, the worse the particles’ dispersibility.

2.2.5. Observation of Particle Aggregation and Dispersion

Aggregation and dispersion behavior of hematite and chlorite in suspension were observed by an optical microscope (BDS200, OPTEC, Chongqing, China). Hematite and chlorite were mixed in a beaker with deionized water (Water Treatment Equipment Instruments, Sichuan, China), and the reagent was added according to the flotation reagent system. The droplets were extracted from the pulp and dropped to the surface of a glass plate for observation. This method can be more intuitive and accurate to obtain the agglomeration of the mineral particles in the pulp.

3. Results and Discussion

3.1. Effect of Citric Acid Dosage on Flotation of Artificial Mixed Ore

In order to study the effect of citric acid on chlorite in hematite reverse flotation system, the dosage of citric acid with different hematite granularity size were tested. The pH of the pulp was adjusted to 11.5 [14,15], and the results were shown in Figure 1.
Figure 1. Effect of citric acid dosage on hematite reverse flotation (the product indexes in concentrate including Fe grade, the hematite recovery, the removal rate of quartz and chlorite were shown in (A–D), respectively; sodium oleate dosage: 160 mg/L; starch dosage: 60 mg/L; calcium chloride dosage: 100 mg/L; MIBC (methyl isobutyl carbinol) dosage: 20 mg/L; pH: 11.5).

As can be observed from Figure 1A, the content of citric acid has a great influence on the index of flotation concentrate. With the increase of citric acid dosage, the grade of flotation concentrate increased first and then decreased, indicating that, when citric acid is in a lower dosage (less than 30 mg/L), it will be helpful for flotation separation of minerals in the pulp. It can also be found that coarse-grained hematite flotation concentrate index was significantly higher than fine-grained one, indicating that the greater the size of hematite, the better the separation effect. From Figure 1B, we know that citric acid has a great influence on the flotation recovery of fine-grained hematite. With the increase of citric acid dosage, the recovery rate of hematite was increased significantly and then gradually stabilizes. Figure 1C showed that low concentration of citric acid has little effect on the removal rate of quartz, but with the increase of citric acid dosage, the removal rate of quartz decreases significantly, which indicates that a high concentration of citric acid can inhibit the floatability of quartz. According to Figure 1D, we found that the removal rate of chlorite increased significantly with the increase of citric acid dosage, but the high dosage of citric acid can also inhibit the floatability of chlorite.

In order to study the mechanism of citric acid on removal rate of chlorite, the contact angle of hematite and chlorite was tested before and after citric acid added. From the measurement results in Table 1, we found that chlorite is a hydrophobic mineral, and citric acid has small impact on the contact angle of hematite, chlorite and quartz, but the artificial mixed ore flotation test showed that the removal rate of chlorite was increased from about 30% to about 75% by adding citric acid. Thus, the reason that citric acid improved the removal rate of chlorite is not by changing the surface hydrophobicity of chlorite.

Table 1. Mineral contact angle measurement results (pH: 11.5; sodium oleate dosage: 160 mg/L; starch dosage 60 mg/L; calcium chloride dosage: 100 mg/L; citric acid dosage: 30 mg/L).

<table>
<thead>
<tr>
<th>Measuring Medium</th>
<th>Without Citric Acid</th>
<th>Citric Acid Added</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Hematite</td>
<td>Chlorite</td>
</tr>
<tr>
<td>Water</td>
<td>36.3°</td>
<td>114.6°</td>
</tr>
<tr>
<td>Glycerol</td>
<td>61.8°</td>
<td>136.2°</td>
</tr>
</tbody>
</table>

3.2. Citric Acid Dosage on the Settlement Effect of Artificial Mixed Ore

Citric acid is a kind of strong acidic ternary organic carboxylic acid, due to its molecular structure contains carboxyl, it has good affinity with oxidized minerals, and is easily adsorbed on the surface of oxidized minerals. The adhesion between particles is eliminated by changing the surface properties of the minerals [16]. The flotation test of artificial mixed ore showed that low concentration of citric
Acid could significantly improve the removal rate of chlorite in hematite reverse flotation. In order to analyze its mechanism, the settlement behavior was quantitatively evaluated by artificial mixed ore.

![Figure 2](image_url)

**Figure 2.** Effect of citric acid dosage on settlement rate (the settlement rate of minerals in hematite flotation system with different hematite size were shown in (A–D), respectively; sodium oleate dosage: 160 mg/L; starch dosage: 60 mg/L; calcium chloride dosage: 100 mg/L; pH: 11.5).

According to the results showed in Figure 2, citric acid dosage has a great impact on the settlement rate of chlorite, and the influence on the settlement rate of fine-grained hematite (−10 μm and −25 + 10 μm) is also significantly, but has little impact on the coarse grained hematite and quartz. Taking Figure 2A as an example, with the citric acid dosage increased to 50 mg/L, the hematite settlement rate increased from 87.21% to 92.87%, while the chlorite settlement rate decreased from 88.83% to 76.67%. The mechanism may be that, without citric acid, a large amount of chlorite adsorbed on the surface of hematite, causing it to sink with the hematite to the bottom of the pulp. While citric acid added, the dispersion degree in the pulp was increased, and chlorite was desorbed from the hematite surface, thus the chlorite settlement rate decreased significantly.

### 3.3. The Extended DLVO Theoretical Calculation

The classical DLVO theory, which is used to study the stability of charged colloidal particles, holds that there is the electrostatic and van der Waals interaction between charged particles. However, the extended DLVO theory, which not only concerns the two kinds of action energy above, but also involves the interaction of polar interfaces [17–20]. The extended DLVO theory can be used to explain the cohesion and dispersion behavior of mineral particles under the action of various flotation agents. Hence, in this paper, the extended DLVO theory was used to calculate the interaction energy between hematite and chlorite particles, and the total interaction energy \( V_{T}^{ED} \) was calculated as follows:

\[
V_{T}^{ED} = V_W + V_E + V_H
\]
where $V_W$ is Van der Waals interaction; $V_E$ is Electrical interaction; and $V_H$ is Hydrophobic (or hydration) interaction.

(1) Van der Waals interaction, $V_W$

For a granule system with large particle size differences ($-74 + 45 \mu m$ grain grade hematite and $-10 \mu m$ grain grade chlorite):

$$V_W = -\frac{A_{122} R_1}{6H}$$

(3)

For granule system with similar particle size (the remaining grains of hematite and $-10 \mu m$ grain grade chlorite):

$$V_W = -\frac{A_{122} R_1 R_2}{6H R_1 + R_2}$$

(4)

where $R_1$ and $R_2$ are the radius of the two particles, respectively; $H$ is the interaction distance between two particles. $A_{122}$ is the effective Hamaker constant of the substances 1 and 2 in medium 3, and it can be calculated as follows:

$$A_{122} = (\sqrt{A_{11}} - \sqrt{A_{33}})(\sqrt{A_{12}} - \sqrt{A_{33}})$$

(5)

where $A_{11}$, $A_{22}$, and $A_{33}$ are the Hamaker constants of substances 1, 2 and medium 3 in vacuum, respectively.

(2) Electrostatic interaction, $V_E$

$$V_{ER} = \frac{\pi \varepsilon_0 \varepsilon_r R_1 R_2}{R_1 + R_2} \left( \frac{\phi_1^2 + \phi_2^2}{\phi_1^2 + \phi_2^2} \right) \left[ \frac{2\phi_1 \phi_2}{\phi_1^2 + \phi_2^2} p + q \right]$$

(6)

where $p, q$ can be obtained by the following formula,

$$p = \ln \left[ \frac{1 + \exp(-kH)}{1 + \exp(-kH)} \right]$$

(7)

$$q = \ln \left[ 1 - \exp(-2kH) \right]$$

(8)

where $\varepsilon_0$ is the absolute permittivity in the vacuum and $\varepsilon_0 = 8.854 \times 10^{-12} \text{ C}^2 \text{ J}^{-1} \text{ m}^{-1}$; $\varepsilon_r$ is the absolute dielectric constant of the dispersion medium (for water $\varepsilon_r = 78.5 \text{ C}^2 \text{ J}^{-1} \text{ m}^{-1}$); $\phi_1$ and $\phi_2$ are the zeta potentials of two minerals, respectively; $k$ is the Debye constant and $k^{-1} = 0.104 \text{ nm}$.

(3) Hydrophobic (or hydration) interaction, $V_H$

For granule system with large particle size differences:

$$V_H = 2\pi R_1 h_0 V_H^0 \exp \left( \frac{H_0 - H}{h_0} \right)$$

(9)

For granule system with similar particle size:

$$V_H = 2\pi \frac{R_1 R_2}{R_1 + R_2} h_0 V_H^0 \exp \left( \frac{H_0 - H}{h_0} \right)$$

(10)

where $h_0$ is the attenuation length, usually 1-10 nm; $H_0$ is the equilibrium contact distance between the two surfaces; $V_H^0$ is the interaction energy constant of the polar interface, which can be obtained by the following formula:

$$V_H^0 = 2 \left\{ \sqrt{\gamma_1^0} (\sqrt{\gamma_1} + \sqrt{\gamma_2} - \sqrt{\gamma_3}) + \sqrt{\gamma_2^0} (\sqrt{\gamma_1} + \sqrt{\gamma_2} - \sqrt{\gamma_3}) - \sqrt{\gamma_1^0} \gamma_2 - \sqrt{\gamma_1} \gamma_2^0 \right\}$$

(11)
where $\gamma_1$, $\gamma_2$, $\gamma_3$ are the electron acceptor components of particles 1, 2 and 3; and $\gamma_1^-$, $\gamma_2^-$, $\gamma_3^-$ are the electron donor components of particles 1, 2 and 3, respectively.

The surface of the minerals studied in this paper is regarded as a single polarity surface, so $\gamma_S^+ \approx 0$, while $\gamma_{SS}^+$, $\gamma_{SS}^-$ of particles 1 and 2 can be obtained by measuring the contact angle of solids in two polar liquids [21],

$$ (1 + \cos \theta) \gamma_L = 2(\sqrt{\gamma_S^+ \gamma_L^+} + \sqrt{\gamma_S^- \gamma_L^-}) $$

(12)

where $\gamma_L$, $\gamma_L^+$, $\gamma_L^-$ were the surface energy of liquid, the surface energy dispersion component, and the electron acceptor component. The variable $\gamma_{SS}^+$ has the following relationship with the Hamaker constant $A$:

$$ A = 2.05 \times 10^{-21} \gamma_{SS}^+ $$

(13)

The surface tension parameters of water and glycerol are shown in Table 2, and the results of the zeta potential are shown in Figure 3.

<table>
<thead>
<tr>
<th>Liquid</th>
<th>$\gamma_L$ (mJ/m$^2$)</th>
<th>$\gamma_L^+$ (mJ/m$^2$)</th>
<th>$\gamma_L^-$ (mJ/m$^2$)</th>
<th>$\gamma_L^-$ (mJ/m$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>72.8</td>
<td>25.5</td>
<td>21.8</td>
<td>25.5</td>
</tr>
<tr>
<td>Glycerol</td>
<td>64</td>
<td>3.92</td>
<td>34</td>
<td>57.4</td>
</tr>
</tbody>
</table>

Figure 3. Effect of citric acid on the zeta potential of hematite and chlorite surface (citric acid dosage: 30 mg/L; sodium oleate dosage: 160 mg/L; starch dosage: 60 mg/L; calcium chloride dosage: 100 mg/L; pH: 11.5).

Based on Equations (2)–(13), and substituting the data measured in Tables 1 and 2 and Figure 3, total interaction energies ($V_{TOT}$) between hematite and chlorite with adding citric acid or not can be calculated, and showed in Figures 4 and 5. The results show that the total interaction energies of hematite and chlorite particles is negative when the citric acid is not added, confirming that the force between those two is gravitational, fine-grained chlorite adhered to the surface of hematite, resulting in a low removal rate of chlorite. When citric acid is added to the pulp, the total interaction energies between hematite and chlorite can be changed to a positive value, indicating that the force between them is repulsive, and the particle dispersion in the pulp is enhanced, then the chlorite removal rate.
increased. The results further explain the effect of citric acid on the dispersion of the pulp during the flotation process.

**Figure 4.** Potential energy curve of extended DLVO in hematite and chlorite in the absence of citric acid.

**Figure 5.** The curve of extended DLVO potential between hematite and chlorite after citric acid is added.

It can also be found from Figures 4 and 5 that, whether the citric acid is added or not, the interaction energies of coarse-grained hematite and chlorite is greater than that of fine-grained hematite and chlorite. However, the artificial mixed mineral flotation tests showed that the coarser the particle size of hematite, the greater the removal rate of chlorite. The results indicated that a small amount of chlorite adsorbs on the surface of coarse-grained hematite due to its small specific surface area.

### 3.4. Particle Size Analysis of Artificial Mixed

The apparent particle size in the pulp can indicate the agglomeration/dispersion state between the minerals indirectly. In order to verify the dispersion effect of citric acid between hematite and chlorite, the particle size of hematite (−10 μm grain grade as an example) and chlorite, and the particle size of artificial mixed ore with adding citric acid or not was analyzed, respectively.
Considering the results showed in Figure 6, with the absence of citric acid, the artificial mixed ore due to its adhesion behavior between hematite and chlorite particles, the Sauter mean diameter is up to 26.2 μm, and about 63.76% of the particle in the pulp was greater than 14.5 μm. After the addition of citric acid, the Sauter mean diameter of the artificial mixed ore decreased to 10.7 μm, and only 34.68% of the particles was greater than 14.5 μm. The results showed that the addition of citric acid can significantly reduce the adhesion behavior between hematite and chlorite, and increase the dispersibility of the pulp, thus significantly reduce the apparent size of artificial mixed ore.

3.5. Optical Microscopy Image of Particles in the Pulp

In order to observe the dispersion effect of citric acid on hematite (−74 + 45 μm grain grade so that the image more clearly) and chlorite more intuitively, the micrograph of the state between these two particles in the pulp was obtained [22] and showed in Figure 7. As can be seen from Figure 7A that, without citric acid added, a large amount of chlorite is adsorbed to the surface of hematite, resulting in a lower removal rate of chlorite in the artificial mixed ore flotation test. After citric acid is added, hematite and chlorite are fragmented. At this point, the hydrophobic chlorite adsorbed on the surface of the bubble and separated from the pulp, thus improved significantly the removal rate of chlorite.

The purpose of this study is to reveal the mechanism that citric acid can significantly improve the removal rate of chlorite. Through the contact angle measurement, zeta potential tests and optical microscopy analysis, the mechanism of citric acid was discussed. However, these findings are just a preliminary understanding of the pure mineral system. Subsequent studies should be conducted to test the effects of citric acid on the actual ore flotation, and much more work should be carried out.
Figure 7. Optical microscopy image of particles in the pulp with citric acid added (B) or not (A) (sodium oleate dosage: 160 mg/L; starch dosage: 60 mg/L; calcium chloride dosage: 100 mg/L; pH: 11.5; citric acid dosage: 30 mg/L).

4. Conclusions

In this investigation, we conducted batch flotation tests with artificial mixed ore of hematite, quartz and chlorite by a mechanical agitation flotation machine. The results showed that citric acid can increase significantly the removal rate of chlorite in hematite reverse flotation. Based on the extended DLVO theory, the dispersion mechanism of citric acid was studied. The results obtained are summarized as follows.

(1) In the reverse flotation process of hematite, chlorite is easily adsorbed on the surface of hematite, resulting in a lower chlorite removal rate and a poor concentrate index.

(2) Extended DLVO theory studies indicate that citric acid by changing the contact angle and zeta potential of minerals, thus the interactions between hematite and chlorite is from attractive forces to repulsive forces, so that the particles are scattered between each other, and ultimately significantly improve the removal rate of chlorite in the flotation.

(3) Artificial mixed ore settlement test and laser particle size analysis results showed that, with the addition of citric acid, the settlement rate of chlorite was decreased significantly, and the apparent particle size of the pulp was decreased obviously, the results further confirmed the dispersion effect of citric acid in the hematite flotation system.
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Conflicts of Interest: The authors declare no conflict of interest.

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