

Indium recovery from zinc oxide flue dust by oxidative pressure leaching

LI Xuan-hai(黎铨海)¹, ZHANG Yan-juan(张燕娟)¹, QIN Quan-lun(覃全伦)¹,
YANG Jian(阳 健)¹, WEI Yan-song(韦岩松)²

1. School of Chemistry and Chemical Engineering, Guangxi University, Nanning 530004, China;

2. Department of Chemistry and Biology, Hechi College, Yizhou 546300, China

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Abstract: Indium was recovered from zinc oxide flue dust (ZOFD) with sulfuric acid by oxidative pressure leaching in an autoclave, and the effects of different technological conditions on indium leaching were studied. Potassium permanganate and hydrogen peroxide were used as oxidants. The atmospheric pressure leaching experiments were also carried out. The experimental results show that the leaching rate of indium can be effectively improved by oxidative pressure leaching. The optimum conditions of pressure leaching are determined as sulfuric 5.10 mol/L acid, leaching time 150 min, temperature 90 °C, and the H₂O₂ dosage of 0.5 mL/g or 2.5% KMnO₄. The leaching rate of indium is more than 90%, which is increased by 13% compared with that of atmospheric pressure leaching process without oxidant under the optimum conditions.

Key words: indium; zinc oxide flue dust; oxidative pressure leaching; potassium permanganate; hydrogen peroxide

1 Introduction

Indium has a wide range of applications due to its unique physical and chemical properties[1], which has been widely used in optoelectronic devices, transparent electrodes for liquid crystal display and solar cells[2–6]. Indium is most commonly found in zinc-bearing materials such as sphalerite or marmatite[7–8]. At present, many raw materials for recovering indium are the previously abandoned zinc smelter slag and leaching residue. These waste residues are calcined at high temperature, the flue dust was collected and then the zinc oxide flue dust (ZOFD) containing 0.1%–0.3% (mass fraction) indium was got[9–10]. Sphalerite and marmatite are sulphur-bearing minerals. When being calcined at high temperature, indium sulfide (In₂S₃) will be formed and enter into the crystal lattice of zinc ferrite (franklinite, ZnFe₂O₄) or other solid solutions. The stability of indium sulfide, zinc ferrite and solid-melt in sulphuric acid creates problems in indium recovery by hydrometallurgical processes.

Pressure leaching can increase the temperature to more than 100 °C, which cannot be achieved under atmospheric pressure. In addition, the oxidative effect of

oxidants can be enhanced under the pressure leaching. In recent investigations, oxidative pressure leaching was widely used in the leaching of sulphur-bearing nickel minerals[11–14], sulphur-bearing zinc minerals[15–16], sulphur-bearing copper minerals[17–18] and other sulphide concentrates or slag[19–21]. The oxidative pressure leaching has very significant effect on recovering metals from sulphur-bearing minerals or slag. Using hydrogen peroxide (H₂O₂) and potassium permanganate (KMnO₄) as oxidant in pressure leaching has been reported[19, 22–24]. In the presence of oxidant, the sulfides and other stable phases can be dissolved efficiently, then the valuable metals are recovered. In order to efficiently extract indium from ZOFD, potassium permanganate and hydrogen peroxide are used as oxidants, and the effect of oxidative pressure leaching on indium extraction in the autoclave is studied in this work.

2 Experimental

2.1 Materials

The ZOFD sample used was got from a zinc production plant in Liuzhou, Guangxi, China, which was obtained by roasting the smelting residues in the rotary

kiln at high temperature. The sample was characterized by X-ray diffractometry (XRD, Rigaku model D/max-2500) and the result is shown in Fig.1. The composition of the ZOFD is very complex, including wurtzite (ZnS), zincite (ZnO), quartz (SiO_2), galenite (PbS), aluminium oxide (Al_2O_3), franklinite (ZnFe_2O_4), lead silicate (Pb_2SiO_4), zinc antimony oxide ($\text{Zn}_{2.33}\text{Sb}_{0.67}\text{O}_4$), and so on. The chemical composition is presented in Table 1. Industrial sulphuric acid (89.5%) was used for leaching. Two oxidants, potassium permanganate (KMnO_4) and hydrogen peroxide (H_2O_2), were of analytical grade.

Table 1 Main chemical composition of zinc oxide flue dust (mass fraction, %)

Zn	Pb	SiO_2	Fe	Al_2O_3	S	In
19.88	11.82	9.16	8.31	5.05	3.24	0.121

2.2 Leaching procedure

The atmospheric pressure leaching experiments were carried out in a 500 mL three-neck flask immersed in thermostatically controlled water bath, equipped with a reflux condenser, mechanical stirrer and thermometer. The temperature could be controlled within $\pm 1^\circ\text{C}$. Deionized water and sulphuric acid were used to prepare the desired leaching solutions (400 mL) in the three-neck flask. When the temperature reached the pre-set value, 50.0 g of ZOFD and variable dosage of oxidant were added. After reaction for 150 min, the slurry was filtered and the residue was washed adequately with deionized water. The indium concentration in the filtrate was analyzed by polarography (Model JP-303, China)[25].

The pressure leaching tests were performed in a 1 L 1Cr18Ni9Ti stainless steel autoclave (GSH-1) with polytetrafluoroethylene lining, equipped with a PID temperature controller, a heating mantle, a variable speed stirrer, and a feeder. In a typical experiment, the slurry of mixing solid sample and distilled water was added into the autoclave, then the autoclave was sealed and heated.

When the pre-set temperature was approached, sulphuric acid and oxidant were fed into the autoclave by the air compressor device. The pressure in the autoclave was kept constant at 0.5 MPa (gage pressure). At the end of the experiment, the autoclave was rapidly cooled by cold water through the feeder. The slurry was filtered and the residue was washed adequately with deionized water. The filtrate was analyzed for indium concentration by polarography. In these atmospheric pressure and pressure leaching experiments, the reaction temperature, reaction time, initial concentration of sulfuric acid, solid/liquid ratio, and stirring speed were fixed at 90°C , 150 min, 5.10 mol/L, 1:8, and 575 r/min, respectively.

3 Results and discussion

The oxidative pressure leaching experiments were first carried out, then the atmospheric pressure leaching experiments were performed for comparison.

3.1 Effect of dosage of H_2O_2 on oxidative pressure leaching

The experiments were carried out with the H_2O_2 dosage of 0.25, 0.50, 0.75, and 1.0 mL/g. The results are shown in Fig.2. It can be seen that the leaching rate of indium increases obviously with the increase of the dosage of hydrogen peroxide when the dosage is less than 0.50 mL/g. Above 0.50 mL/g, the dosage of H_2O_2 has slight influence on the leaching rate. The leaching rate of indium is more than 90% with H_2O_2 dosage of 0.50 mL/g.

3.2 Effect of sulfuric acid concentration with H_2O_2 as oxidant

The effect of sulfuric acid concentration on indium leaching rate was studied in the range of 2.04–6.12 mol/L with the H_2O_2 dosage of 0.50 mL/g. As seen from Fig.3, indium leaching rate is increased from 63.7% to 91.3% with increasing H_2SO_4 concentration from 2.04 to

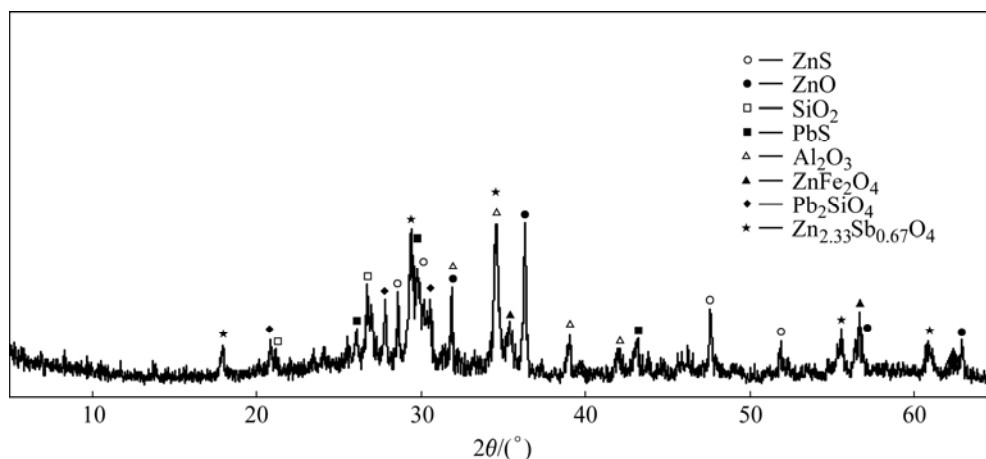


Fig.1 XRD pattern of zinc oxide flue dust

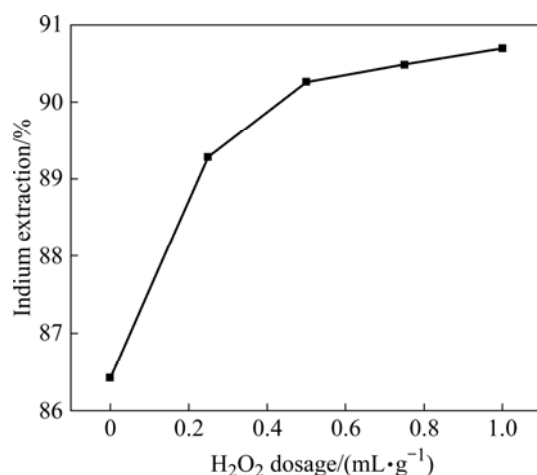


Fig.2 Effect of H₂O₂ dosage on indium extraction in oxidative pressure leaching

6.12 mol/L. It is also observed that the leaching rate is enhanced slowly when the H₂SO₄ concentration is higher than 5.10 mol/L. Increasing the H₂SO₄ concentration from 5.10 to 6.12 mol/L, the leaching rate of indium is slightly improved from 90.3% to 91.3%. Therefore, an initial sulfuric acid concentration of 5.10 mol/L is sufficient for the recovery of indium.

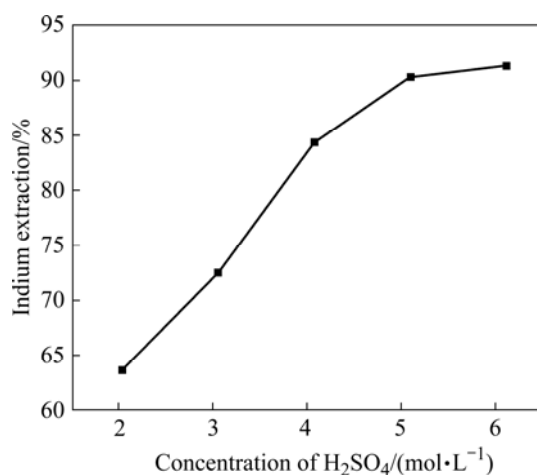


Fig.3 Effect of H₂SO₄ concentration on indium extraction in hydrogen peroxide oxidative pressure leaching

3.3 Effect of dosage of KMnO₄ on oxidative pressure leaching

Another oxidant, potassium permanganate, was used to study the effect of dosage of oxidant on the indium leaching rate. The experiments were performed with the KMnO₄ dosage of 0, 1.25%, 2.5%, 3.75%, 5%, and 6.25%. The results are plotted in Fig.4. Not surprisingly, the leaching rate of indium increases with increasing the dosage of potassium permanganate. It can also be observed that the leaching rate almost increases

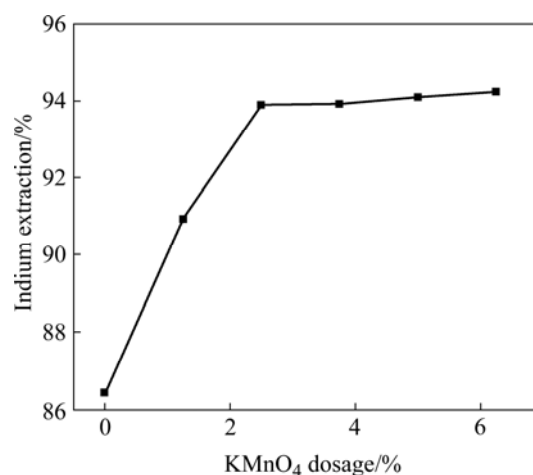


Fig.4 Effect of KMnO₄ dosage on indium extraction in oxidative pressure leaching

linearly increasing when the KMnO₄ dosage is less than 2.5%, while above 2.5%, the leaching rate is not increased obviously. The leaching rate of indium reaches 93.9% with the KMnO₄ dosage of 2.5%, only 94.3% of indium is extracted with 6.25% KMnO₄. Therefore, the KMnO₄ dosage of 2.5% is chosen for the recovery of indium.

3.4 Effect of H₂SO₄ concentration with KMnO₄ as oxidative

The effect of H₂SO₄ acid concentration on indium leaching rate was studied in the range of 2.04–6.12 mol/L with the KMnO₄ dosage of 2.5%. It can be observed from Fig.5 that the extraction of indium increases sharply with the H₂SO₄ concentration increasing from 2.04 to 3.06 mol/L. The increasing trend becomes slight while the H₂SO₄ concentration is above 3.06 mol/L, and the leaching efficiency of indium is about

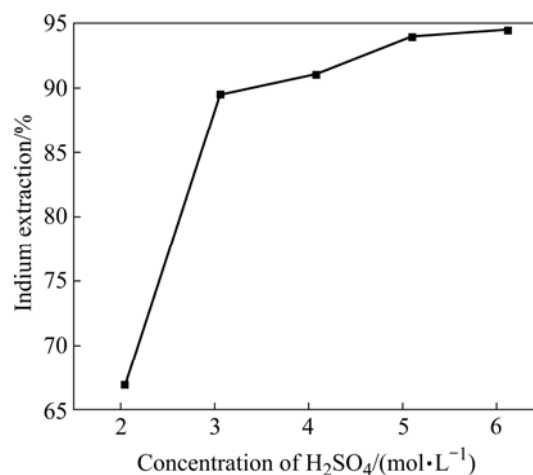


Fig.5 Effect of H₂SO₄ concentration on indium extraction in KMnO₄ oxidative pressure leaching

90% with acid concentration of 3.06 mol/L. Increasing the H_2SO_4 concentration from 3.06 to 6.12 mol/L slightly improves the recovery of indium from 89.5% to 94.4%. This shows that the extraction of indium is not strongly dependent on the acid concentration while the H_2SO_4 concentration is above 3.06 mol/L with potassium permanganate as oxidant.

3.5 Comparison of two leaching technologies

In order to study the different effects of atmospheric oxidative leaching and oxidative pressure leaching, the experiments were carried out in a 500 mL three-neck flask with potassium permanganate as oxidant. The conditions of atmospheric oxidative leaching were the same as those of the experiments of changing the dosage of potassium permanganate in oxidative pressure leaching. The results are shown in Fig.6. The results demonstrate that the pressure leaching has obvious influence on the extraction of indium. Without oxidant, the leaching rate of indium is 77.6% in atmospheric oxidative leaching, while 86.4% in pressure leaching, so the recovery of indium is increased by 8.8% just through enhancing the pressure. In these experiments, the leaching rate of indium is significantly increased with the increase of the dosage of potassium permanganate, and the recovery of indium in pressure leaching is about 8% higher than that in atmospheric pressure leaching. These results indicate that both the oxidant and pressurizing can strengthen the indium extraction from zinc oxide flue dust.

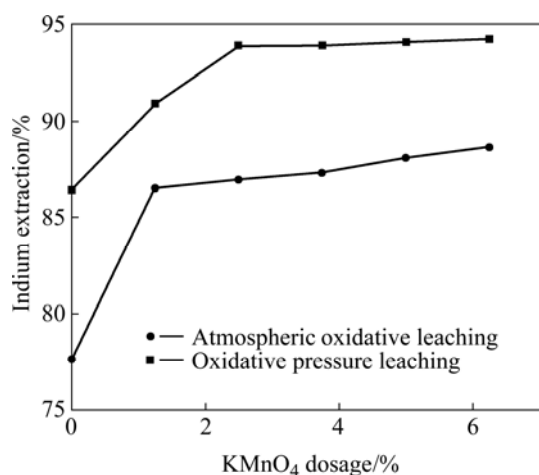


Fig.6 Effect of different leaching processes on indium extraction

These phenomena are related with the composition of the indium-containing minerals. In the pressure leaching, the concentration of oxygen in the solution is higher than that in the atmospheric leaching, so some of the stable indium-bearing phases can be destroyed and then dissolved easily into the solution. The addition of oxidant not only destroys the stable indium-bearing

phases, but also oxidizes the indium sulfide which is difficult to dissolve in the normal leaching process, thus increasing the recovery of indium.

4 Conclusions

1) The oxidative pressure leaching can effectively recover indium from zinc oxide flue dust, which is a new process of extracting indium from low indium-bearing minerals.

2) The leaching rate of indium increases with increasing the dosage of oxidant and the concentration of H_2SO_4 . The pressure leaching has obvious influence on the extraction of indium. Both adding the oxidant and enhancing the leaching pressure increase the indium extraction from ZOFD. The effect is more significant using potassium permanganate as oxidant than using hydrogen peroxide. The use of potassium permanganate in pressure leaching can obviously decrease the dependence on the H_2SO_4 concentration.

3) The optimum conditions of pressure leaching are 5.10 mol/L H_2SO_4 , the H_2O_2 dosage of 0.5 mL/g or 2.5% KMnO_4 , at 90 for 150 min. The leaching rate of indium is more than 90%.

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