



Pressure leaching of manganotantalite by sulfuric acid using ammonium fluoride as an assistant reagent

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ABSTRACT

Manganotantalite is difficult to decompose by sulfuric acid alone. Even at sulfuric-acid mass concentrations of 60%, only 8.5% tantalum and 9.9% niobium leached. To obtain a high leaching efficiency of tantalum and niobium, ammonium fluoride is added as an assistant reagent. Over 93% tantalum and 96% niobium were leached by mixing $-74\ \mu\text{m}$ manganotantalite with 50% sulfuric acid and ammonium fluoride, with heating for 2 h at 200 °C in a closed vessel. The ammonium fluoride/manganotantalite mass ratio was 0.8:1. A comparison of the novel technology that is proposed in this article with traditional hydrofluoric-acid technology shows that the mass concentration decreases significantly from 60 to 70% to 5.3%. Excess fluoride ions in solution can be recycled as the assistant reagent. Therefore, hardly any fluoride ions will be discharged as environmental pollutants. The local-density approximation was used for an adsorption-energy calculation of the fluoride ion on the MnNb_2O_6 (100) surface. The calculation results indicate that fluoride-ion adsorption on the MnNb_2O_6 (100) surface is a chemisorption process. The partial density-of-states analysis indicates that fluoride ions interact mainly with surface Nb atoms through a complicated orbital hybridization. The charge transfer from Nb to F confirms the ionic characteristics of the Nb–F bonds.

1. Introduction

Tantalum and niobium are distributed sparingly in the earth's crust with Clark values of 0.0002 wt% and 0.0020 wt%, respectively, and so they are classified as rare metals. Because of their good corrosion resistance, high melting point and good thermal conductivity, tantalum and niobium are used widely in metallurgy, high-energy physics, aerospace, chemical anticorrosion and electronic industries (Wang et al., 2013; Yang et al., 2013). Moreover, some alkali-metal niobates have been used in laser technology.

Tantalum and niobium can be extracted from tantalum–niobium ores (Deblonde et al., 2016a; Deblonde et al., 2016b; El-Hussaini and Mahdy, 2002; Nete et al., 2014). China possesses the second-most-abundant global tantalum–niobium resources. Tantalite and niobite and columbite–tantalite, microlite and pyrochlore are the main ores that are used for tantalum and niobium extraction (Yuan et al., 2015). Tantalite, niobite and columbite–tantalite ($(\text{Fe},\text{Mn})(\text{Ta},\text{Nb})_2\text{O}_6$) are decomposed mainly by 60–70% hydrofluoric acid at 90–100 °C to produce tantalum and niobium-fluoride complexes, and sometimes by using mixed solvents of hydrofluoric acid and concentrated sulfuric acid to decrease the solution boiling point and to reduce the dosage of hydrofluoric acid

(Guo and Wang, 2009). It is difficult to decompose these ores using sulfuric acid alone. The ore can also be decomposed by alkali fusion, such as by using sodium hydroxide and potassium hydroxide. A certain amount of corresponding carbonate is added to lower the melting point and smelt viscosity. The main disadvantage of this method is the high alkali consumption, which is 6 to 8 times the reaction requirement. Microlite and pyrochlore ($(\text{Ca},\text{Na})_2(\text{Nb},\text{Ta})_2(\text{O},\text{OH},\text{F})_7$) are decomposed easily by sulfuric acid to produce $\text{Nb}_2\text{O}_4\text{SO}_4$ in dilute sulfuric-acid solution and $\text{Nb}_2\text{O}_3(\text{SO}_4)_2$ in more concentrated sulfuric-acid solution (Guo and Wang, 2009; Yang et al., 2014).

Manganotantalite ($\text{Mn}(\text{Ta}_{0.5}\text{Nb}_{0.5})_2\text{O}_6$) is similar to tantalite and can be decomposed by hydrofluoric acid, but because of its high volatility, approximately 10% hydrofluoric acid will be lost during the decomposition process and cause serious environmental pollution. Gaseous fluoride (HF) is highly phytotoxic, and harms plants in the surrounding environment. People are prone to dental disease and plants are affected to necrosis, yellowing and death.

In present work, sulfuric acid (60%) was used to decompose manganotantalite at 200 °C and 1.5 MPa for 2 h, and only 8.5% and 9.9% tantalum and niobium were leached, respectively. The X-ray pattern of the sample in Fig. 1 shows that the main phases are manganotantalite,

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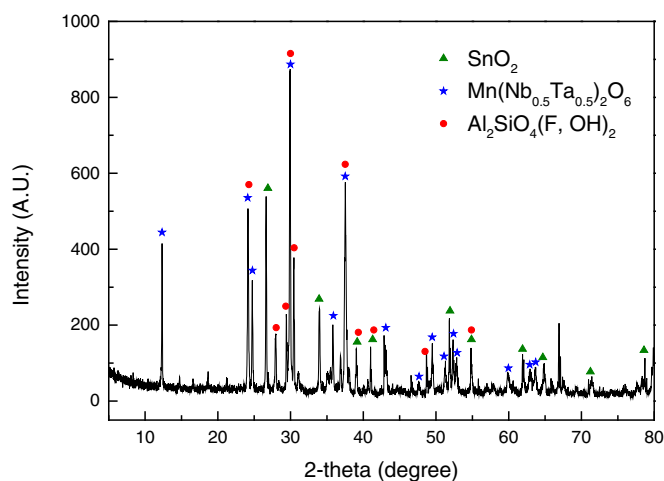


Fig. 1. XRD pattern of the sample.

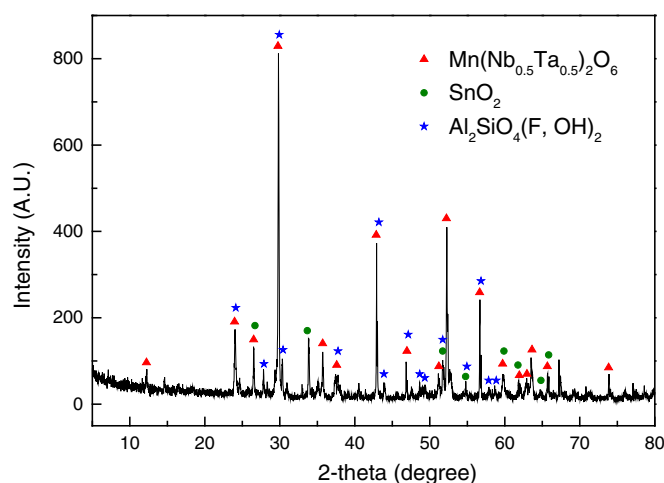


Fig. 2. XRD pattern of leaching residue.

topaz and cassiterite. The XRD pattern of the leaching residue is shown in Fig. 2. Figs. 1 and 2 show that most manganotantalite was not leached. To promote manganotantalite decomposition and to reduce the volatilization of hydrofluoric acid, ammonium fluoride is introduced as an assistant reagent, instead of hydrofluoric acid. The leaching process was carried out in a pressure-sealed system, and thus, the sulfuric-acid and ammonium-fluoride dosages can be decreased. To date, the pressure leaching of manganotantalite by sulfuric acid using ammonium fluoride as an assistant reagent has not been reported in detail.

Detailed information on the effect of leaching parameters, such as sulfuric-acid concentration, ammonium-fluoride dosage, pressure, temperature, liquid-to-solid ratio and time, does not exist, so research was conducted to study such effects on the leaching of manganotantalite in sulfuric acid. Density-functional theory (DFT) calculations were carried out to investigate the mechanism of interaction between fluoride ions and the MnNb_2O_6 surface. The mechanism of interaction was proposed based on adsorption energy, Milliken population, molecular orbital and electronic structure analysis.

2. Experimental and theoretical calculations

2.1. Experimental

2.1.1. Materials and reagents

Manganotantalite was obtained from Jiangxi Yichun Mine, China. The ore was ground to $-74\ \mu\text{m}$ and analyzed by inductively coupled

Table 1
Elemental analysis of the manganotantalite.

Constituent	Ta ₂ O ₅	Nb ₂ O ₅	Al ₂ O ₃	FeO	MnO ₂	SnO ₂	SiO ₂
Content, wt%	18.7	13.9	16.3	17.6	7.0	14.2	10.9

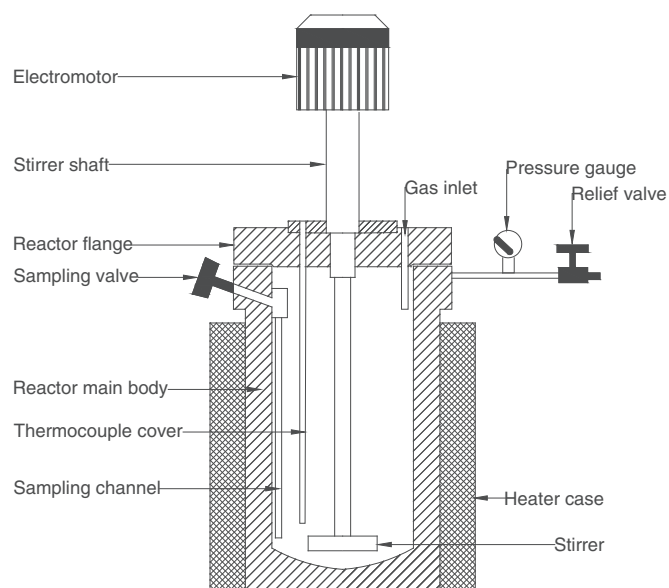


Fig. 3. Schematic representation of pressure reactor.

plasma-atomic emission spectroscopy (ICP-AES) and X-ray diffractometry. The chemical analysis in Table 1 indicates that the contents of tantalum pentoxide and niobium pentoxide are 18.7% and 13.9%, respectively, and the impurity elements include mainly aluminum, silicon, iron, manganese and tin (Kuang, 2011).

Sulfuric acid, hydrochloric acid and ammonium fluoride (all analytical-reagent grade) were from Ganzhou Angesi Science and Technology Co., Ltd., Jiangxi, China. Distilled water was used in the experiments.

2.1.2. Procedure

All experiments were carried out in a Hastelloy® pressure vessel that was equipped with a mechanical stirrer. A schematic of the reactor is presented in Fig. 3. An exact amount of manganotantalite was introduced into the reactor, followed by addition of sulfuric-acid and ammonium-fluoride solution. After sealing, the temperature was increased under air pressure. After each experiment, the solid and liquid were separated by using a water-circulation vacuum filter. The residue was washed with dilute hydrochloric acid three times, dried and weighed. The tantalum and niobium contents in the residue were analyzed by ICP-AES. Prior to analysis, the residue was roasted and smelted with an appropriate amount of sodium carbonate and boric acid at 1000 °C for 20 min, and then dissolved using hydrochloric-acid solution in a 1:1 (m/v) ratio. The solutions were diluted to a suitable concentration for analysis.

The tantalum or niobium leaching efficiency was calculated by mass balance, as follows:

$$\text{Leaching efficiency} = \left(1 - \frac{m}{m_0}\right) \times 100 \quad (1)$$

where m is the mass of tantalum or niobium in the residue and m_0 is the mass of tantalum or niobium in the manganotantalite.

2.1.3. Analysis

To examine the phase composition of the manganotantalite and

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