Available online at www.sciencedirect.com

SCIENCE DIRECT®

Hydrometallurgy 80 (2005) 170–178

hydrometallurgy

www.elsevier.com/locate/hydromet

A kinetic study of the leaching of a low-grade niobium–tantalum ore by concentrated KOH solution

Hong-ming Zhou [a,b,*], Shi-li Zheng [a], Yi Zhang [a], Dan-qing Yi [b]

[a]Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100080, China
[b]School of Materials Science and Engineering, Central South University, Changsha, Hunan 410083, China

Received 21 October 2003; received in revised form 1 June 2005; accepted 19 June 2005
Available online 3 August 2005

Abstract

A kinetic study of the leaching of a low-grade niobium–tantalum ore by concentrated KOH solution under atmospheric pressure has been investigated. The effects of reaction temperature, KOH concentration, agitation speed, particle size and alkali-to-ore mass ratio on the dissolution rate of niobium were examined. It was found that the dissolution rate of niobium is significantly influenced by the temperature, particle size and concentration of KOH solution. The experimental data were well interpreted with a shrinking core model under diffusion control through the product layer. By using the Arrhenius expression, the apparent activation energy for the dissolution of niobium was evaluated. Finally, on the basis of the shrinking core model, the following rate equation was established:

$$1-3(1-X_{\mathrm{b}})^{2/3}+2(1-X_{\mathrm{b}})=\frac{k_0}{R^2}\mathrm{e}^{-\frac{72200}{RT}}t.$$

Keywords: Clean process; Leaching; Kinetics; KOH; Niobium–tantalum ore; Diffusion control

1. Introduction

At present, most ores containing niobium and tantalum are decomposed by concentrated hydrofluoric acid. However, such an approach is only appropriate for high-grade niobium–tantalum ore, and special containers are required due to the remarkable corrosiveness of hydrofluoric acid (HF) (Miller, 1959; El-Hussaini, 2001; He et al., 1998a; Gupta and Suri, 1994a). Moreover, the decomposition process is accompanied by the loss of a considerable amount of HF (He et al., 1998b) because of the volatilisation, which is harmful to human beings and equipment. A large amount of fluorspar sludge is generated and needs to be disposed of (Bludssus and Eckert,

---

* Corresponding author. Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100080, China. Tel.: +86 01 82610244; fax: +86 01 62561822.

E-mail address: ipezhm@yahoo.com.cn (H. Zhou).

0304-386X/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.hydromet.2005.06.011

1993). In addition, although the resource of niobium–tantalum ore is rich in China, most of it is hard to decompose with hydrofluoric acid (Li et al., 1992). Therefore, it is imperative to develop new and cleaner production processes, so as to maximize resource utilization.

Recently, a new process for the leaching of a low-grade niobium–tantalum ore has been proposed by the Institute of Process Engineering, Chinese Academy of Sciences, China. Based on the principles of cleaner production, the new process can serve as an alternative solution for the traditional process. In the new process, low-grade niobium–tantalum ore is decomposed in fluid medium of concentrated KOH solution under atmospheric pressure, and the reaction and mass transfer are significantly enhanced. As a result, the decomposition rate of niobium–tantalum ore is greatly improved at a relatively low temperature.

Concentrated KOH solution leaching of niobium from low-grade niobium–tantalum ore remains so far a developing field. Much fundamental work needs to be carried out. Although some previous studies had been performed on the atmospheric alkaline leaching or pressure alkaline leaching process for niobium–tantalum ore (Oka and Miyamoto, 1949; Bhattacharya, 1952; Cardon, 1962; Dickson and Dukes, 1957), no previous work has ever been reported on the kinetics of concentrated KOH solution leaching. The present work mainly focuses on the kinetics of concentrated KOH solution leaching process for low-grade niobium–tantalum ore. The reaction and operation parameters examined include the agitation speed, the initial KOH concentration, the temperature, the particle size of the ore and the alkali-to-ore mass ratio.

2. Theoretical

The reactions of niobium–tantalum ore with KOH solution are as follows (Orekhov and Zelikman, 1963; Zelikman and Orekhov, 1965, 1972):

$$\begin{aligned}&3(\mathrm{Fe,Mn})\mathrm{O}\cdot(\mathrm{Ta,Nb})_2\mathrm{O}_5 + 8\mathrm{KOH} + (n-4)\mathrm{H_2O}\\&\quad = \mathrm{K}_8[(\mathrm{Ta,Nb})_6\mathrm{O}_{19}\cdot n\mathrm{H}{\pm}_2\mathrm{O}] + 3(\mathrm{Fe,Mn})\mathrm{O} \qquad (1)\end{aligned}$$

$$\begin{aligned}&\mathrm{K}_8[(\mathrm{Ta,Nb})_6\mathrm{O}_{19}\cdot n\mathrm{H_2O}] \rightleftharpoons 6\mathrm{K}(\mathrm{Ta,Nb})\mathrm{O}_3 + 2\mathrm{KOH}\\&\quad + (n-4)\mathrm{H_2O} \qquad (2)\end{aligned}$$

At the first stage, the soluble $K_8[(Ta,Nb)_6O_{19}.nH_2O]$ is obtained, and then converted into insoluble $K(Ta,Nb)O_3$ at the second stage. Depending on the reaction conditions niobium and tantalum will be present in the leaching solution as soluble $K_8[(Ta,Nb)_6O_{19}.nH_2O]$ or as insoluble $K(Ta,Nb)O_3$. For the new process (Zhou et al., 2003), the main object is the production of soluble $K_8[(Ta,Nb)_6O_{19}.nH_2O]$. The reaction rate of reaction (2) is significantly influenced by the leaching temperature and KOH concentration. It can be almost restrained when the leaching temperature is 150 °C, or in the range of 150–200 °C and the initial KOH concentration is higher than 82 wt.% (Orekhov and Zelikman, 1963). So the leaching kinetics of niobium from a low-grade niobium–tantalum ore by concentrated KOH solution is investigated under the conditions of the leaching temperature ranging from 150 °C to 200 °C in the present study.

3. Experimental

3.1. Materials

The niobium–tantalum ore used was supplied by the Ningxia Orient Tantalum Industry Co., Ltd, Ningxia, PR China. The main chemical composition of the ore, determined by ICP-AES, is given in Table 1.

The result of X-ray diffraction analysis (XRD, using Phillips PW223/30) of the ore is shown in Fig. 1. The XRD pattern indicates that the main constituent of the ore is niobite, the universal chemical formula of which is expressed as $(Fe,Mn)O\cdot(Ta,Nb)_2O_5$. The ore was dried, screened and grouped into different size fractions, 104–147 μm, 61–74 μm, 43–61 μm and 38–43 μm, in order to examine the possible effect of particle size. The content of Nb_2O_5 in different size fractions is presented in Table 2.

Table 1
Main chemical composition of niobium–tantalum ore

Component (%)							
Nb_2O_5	Ta_2O_5	SiO_2	TiO_2	WO_3	P	MnO	Fe_2O_3
28.72	28.25	1.61	5.73	0.83	0.077	7.53	11.85