



Kinetics of Al Evaporation from Liquid U–Al Alloys in Vacuum Induction Melting



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A research on kinetics of Al evaporation from liquid U–Al alloys was made in a vacuum induction melting (VIM) furnace at 1673–1843 K. The evaporation rate of Al was found to be first order with respect to Al content in the melt. The overall mass transfer coefficient of Al was determined and it was found that the evaporation rate of Al increased with increasing temperatures. The apparent activation energy of Al evaporation at 1673–1843 K was 171.5 kJ mol⁻¹. The value of mass transfer coefficient of Al in the liquid phase was estimated to be 3.77×10^{-6} , 7.41×10^{-6} , and 9.40×10^{-6} m s⁻¹ at 1673, 1753, and 1843 K, respectively. Meanwhile, rate determining steps were discussed and it was concluded that the evaporation rate of Al is mainly controlled by liquid phase mass transfer.

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1. Introduction

Fuel elements based on alloys of uranium (U) with aluminum (Al) are widely used in transport reactors^[1–3]. The recycling of U from U–Al becomes more and more important from the view point of saving energy, materials and protection of environment. Therefore, if Al as an impurity component in the U–Al system can be removed by evaporation, the process can be considered beneficial in terms of the melt product purity. In our previous work^[4], the thermodynamic properties of Al evaporation from U–Al system was studied by carrier gas method^[5]. Some researchers studied the thermodynamic properties of U–Al system^[6–9]. Meier et al.^[6] investigated the recycling of uranium from U–Al alloys by the chlorination route and initially evaluated the optimal reaction conditions by thermodynamic calculations. Slater et al.^[7] researched the electrochemical separation of Al from U. Some researches^[10–14] have been reported on the evaporation behavior of Al from Ti-based alloys and Ni-based alloys during vacuum melting. The research of Zhang et al. showed^[14] that the removal efficiency of Al is more than 90% during the Al evaporation from nickel. Siwiec^[13] studied kinetics of Al evaporation from a liquid Ti-

based alloy during its smelting in a vacuum induction furnace. To our knowledge, however, no work has been reported on the kinetics of Al evaporation behavior from U–Al alloys during vacuum melting. In this case, the kinetics of Al evaporation research such as reaction order, rate determining steps and temperature effects are significant for metallurgical processes of refining U from U–Al system.

The purpose of this work was to investigate evaporation rate of Al from liquid U–Al alloys in a vacuum induction melting furnace at 1673–1843 K. Meanwhile, rate determining steps and effect of melt temperature on the overall mass transfer coefficient of Al were discussed.

2. Experimental

In the present study, the metals used as starting materials for preparation of the targeted alloys were 99.999% pure Al and 99.9% pure U. U–Al alloys with the Al concentration about 1100 mass ppm were prepared by mixing the respective pure metals in a desired proportion and melted in a vacuum induction melting furnace.

The experiments were carried out by using a vacuum induction melting furnace (50 kW) containing about 12 kg of U–Al alloys under 2×10^{-3} to 8×10^{-3} Pa. A dense CaO crucible ($130^{\text{OD}} \times 106^{\text{ID}} \times 200^{\text{H}}$ mm), which was heat-treated at 773 K for 2 h before experiment, was used for melting the U–Al alloys. The U–Al alloy was placed in the CaO crucible and then heated to the

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desired temperatures. Temperature was measured by immersing a W·Re5/W·Re26 thermocouple contained in a protective alumina sheath into the melt. Power to the furnace was manually adjusted on the basis of temperature measurement to maintain the desired melt temperature and it is assumed that the temperature was controlled within ± 10 K. For the investigation of Al concentration change in U–Al melts, melting was performed at three different temperatures, i.e. 1673, 1753 and 1843 K. At each temperature, the melt was kept for well-defined periods of time, which were interrupted for casting small amount of melt into small molds. After homogenization at constant input power, the first sample was taken by casting small amount of melt into a small mold. The molds were coated with Y_2O_3 and preheated to 773 K for 2 h prior to casting. This moment was taken as starting time for the vacuum treatment ($t = 0$). Casting was performed at eight accumulated hold time: 10, 20, 30, 40, 50, 60, 70, 80 min. It is worth mentioning that chamber pressure and melt temperature did not change essentially during the sampling procedure. After the last sample was taken, the melt was cast into a mold.

Al concentrations were quantitatively measured using inductively couple plasma atomic emission spectrometer (ICP-AES).

3. Results

3.1. Reaction order of Al evaporation rate

Fig. 1 shows the typical changes in concentrations of Al during experiment at 1673, 1653 and 1843 K. In the present study, assuming first order rate law with respect to Al content in the melt, rate of Al evaporation is expressed by Eq. (1), and integration of Eq. (1) gives Eq. (2).^{15]}

$$-\frac{V}{S} \frac{dc(t)}{dt} = k_i c(t) \quad (1)$$

$$-\ln \frac{c(t)}{c(0)} = \frac{S}{V} k_i t \quad (2)$$

$$V = m/\rho \quad (3)$$

where $c(0)$ is the initial Al concentration in the melt (ppm); $c(t)$ is the Al concentration in the melt after a reaction time t (ppm); k_i is the apparent first order rate constant ($m s^{-1}$); S is the surface area of melt, which can be calculated by the radius of crucible and is assumed to be constant (cm^2); m is the mass of the melt, which can be accounted for by measuring mass of the samples (kg); ρ is the density of liquid uranium and was assumed to be $17.27 \times 10^3 \text{ kg m}^{-3}$, which is the density of liquid U at melting point

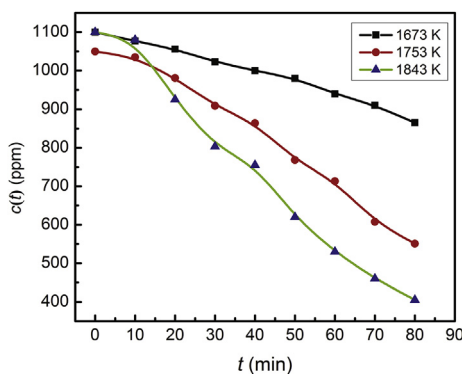


Fig. 1. Variation of Al concentration in liquid U–Al alloys with time.

(1408 K)^{16]}; V is the volume of the melt (cm^3), which was calculated by Eq. (3).

The results shown in Fig. 1 are plotted again according to Eq. (2) in Fig. 2, indicating that the rate of Al evaporation basically accords with first order law. This confirms that the evaporation of Al is represented by a reaction of first order under the present experimental conditions. The straight lines drawn by the least-squares method can be expressed as:

$$1673 \text{ K} : -\ln \frac{c(t)}{c(0)} = (175.3 \pm 4.8) \times 10^{-4} \times \frac{S}{V} \cdot t \quad (4)$$

(Adj. R – Square = 0.99)

$$1753 \text{ K} : -\ln \frac{c(t)}{c(0)} = (398.4 \pm 11.5) \times 10^{-4} \times \frac{S}{V} \cdot t \quad (5)$$

(Adj. R – Square = 0.99)

$$1843 \text{ K} : -\ln \frac{c(t)}{c(0)} = (547.7 \pm 17.7) \times 10^{-4} \times \frac{S}{V} \cdot t \quad (6)$$

(Adj. R – Square = 0.99)

Therefore, the evaporation rate of Al at different temperatures was expressed by the first order evaporation rate constant k_i ($i = 1, 2, 3$ for 1673, 1753 and 1843 K, respectively) ($m s^{-1}$), which was determined from the slope of the function $-\ln[c(t)/c(0)]$ against $(S/V(t)) \cdot t$. The rate constant k_i can be considered as an overall mass transfer coefficient, whereby greater values of k_i correspond to higher evaporation rates of Al in the melt. The values of the first order apparent evaporation rate constant of Al in uranium-based melts at 1673, 1753, and 1843 K was 2.92×10^{-6} , 6.64×10^{-6} , and $9.13 \times 10^{-6} \text{ m s}^{-1}$, respectively. The experimental results, along with the experimental conditions in each experimental run, are listed in Table 1.

3.2. Effect of melt temperature

The evaporation rate was strongly affected by the temperature of the melt. It was found that the evaporation rate of Al increased with increasing temperature. These findings were illustrated in Fig. 3, which shows a plot of $\ln k_i$ vs the reciprocal melts temperature $10^4/T$ (K^{-1}). The straight lines, which were drawn through regression analysis of experimental data, confirm that the dependence of evaporation rate on melt temperature is given by Eq. (7), which represents an Arrhenius type equation.

$$\ln k_i = -\frac{E_{Al}}{RT} + C_1 \quad (7)$$

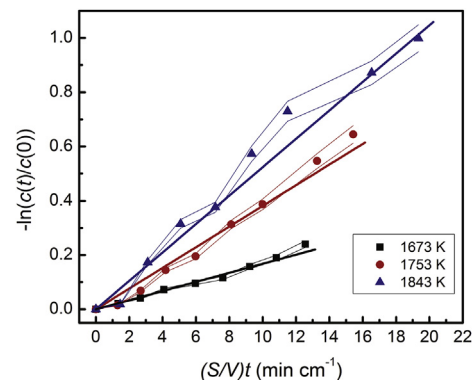


Fig. 2. Plot of $-\ln(c(t)/c(0))$ vs $(S/V) \cdot t$.

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