Effect of the Temperature and Pressure on a Cadmium Distillation

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1. Introduction

The long-lived actinides could be extracted and fabricated into a fuel for use in advanced reactors. In addition, the extraction of actinides from a spent fuel will significantly reduce the radioactivity of the spent fuel and the volume of the spent fuel to be buried in a repository. Pyroprocessing has been developed for the separation of the long-lived elements due to its advantages of compactness, nuclear proliferation resistance, and a reduction of the secondary waste generation [1].

Electrorefining is a key step in the pyroprocessing. Electrorefining process is generally composed of two recovery steps– deposit of uranium onto a solid cathode and the recovery of the remaining uranium and TRU elements simultaneously by a liquid cadmium cathode. After the recovery step of the actinides by the liquid cadmium cathode, cadmium is separated from the actinide products in the liquid cadmium cathode by distillation process [2].

Physical separation processes, such distillation separation, are more attractive than chemical or dissolution process because physical processes generate much less secondary process.

Besides the nuclear industry, cadmium distillation process has other applications in the purification of cadmium and a waste treatment of spent Ni-Cd battery [3].

In this study, cadmium distillation experiments were carried out to examine the effect of the temperature and pressure on the cadmium distillation for the development of an actinide recovery process from a liquid cadmium cathode in the laboratory scale cadmium distiller.
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2. Experimental

Fig. 1 shows a layout of the experimental set-up for the cadmium distillation. The experimental set-up is composed of an evaporator, a condenser, a control unit, an off gas treatment system. The evaporation area is surrounded by a carbon dome. The maximum heating temperature is 1,500℃. The temperature in the carbon dome was measured using a thermocouple that is connected to the center of the dome. A Kanthal super heating element is used for the heating the evaporation area.

The loading capacity is 1kg-Cd/batch. Cadmium ingot was cut into small pieces and used as received without any further treatment. The cadmium ingot has a purity of 99.99 wt. %. The condenser is connected to the rotary vacuum and diffusion pumps. The diffusion pump was used to remove the oxygen in the evaporator and the condenser chamber before an evaporation of cadmium. The absolute vacuum pressure can be reached below 1×10^{-3} torr before a heating. The vacuum pressure was measured by a MKS Baratron pressure sensor and a readout system. During the distillation, the pressure was kept constant by using a throttle valve [MKS] and a controller [MKS 600].

A filter was used to avoid any contact between the cadmium vapor and the pressure sensor. Weight loss of cadmium in the crucible was measured by a load cell. The distillation experiments were performed in a vacuum at various temperatures in the range of 400-700℃. The ceramic filter was placed before the vacuum pump to avoid an entrainment of cadmium vapor to the pump. The off gas from the distiller was treated in a wet scrubber before its release to the atmosphere.

The cadmium crucible with about 170g of cadmium is bottom loaded into the carbon dome by raising the bottom assembly until seated. The evaporator was evacuated and heated up to the distillation temperature. The temperature was held until the complete evaporation of the cadmium. Cadmium vapor was transported to the condenser through the 2" diameter pipe which is preheated by a jacket heater. Cadmium is recovered by allowing the vapor strike a cold receiving crucible, where it condenses.

The evaporation rates of cadmium were measured at various controlled vacuum pressures to obtain the basic data for the design of cadmium cathode processor.
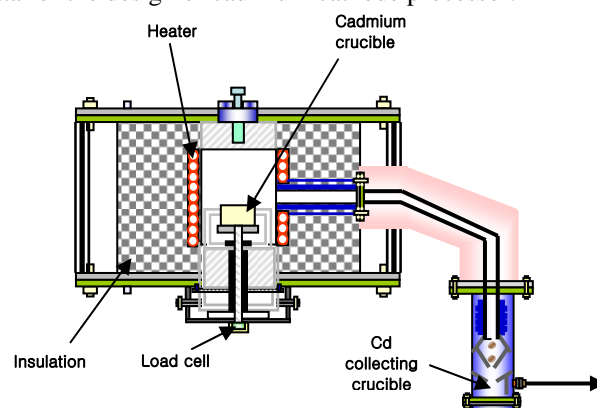

Fig. 1. Experimental set-up for the cadmium distillation experiments.

3. Results and Discussion

Cadmium distillation experiments were carried out to examine the effect of the temperature and pressure on the cadmium distillation for the development of an actinide recovery process from a liquid cadmium cathode.

The evaporation rate depends on various parameters such as the pressure, temperature, geometry of the evaporator and the crucible, and the amount of impurities. The exact evaporation rate per unit surface area is not easy to measure since the surface of the cadmium splat at a high temperature during an evaporation. In this study, the apparent evaporation rates were measured by the average weight loss during evaporation at a controlled temperature and pressure.

The apparent evaporation rates were measured at various temperatures and pressures as shown in Fig. 2. The distillation rate of cadmium was varied from 4.5 to 33 g/cm^2/h in the temperature range of 500- 650℃ and in the vacuum pressure range of 0.5 to 10 torr.
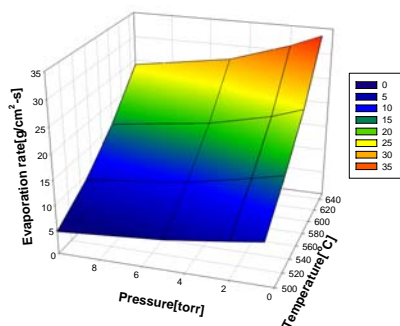

Fig. 2. The evaporation rate of cadmium at various temperatures and vacuum pressures.

The distillation rate of cadmium increased with an increasing temperature, whereas the distillation rate decreased with an increasing vacuum pressure. High evaporation rate may cause a difficulty in the following consolidation process by a scattering of the remaining actinide powders[4]. Therefore the optimum operating conditions should be found for a successful recovery of the actinides from the liquid cadmium cathode.

The measured evaporation rates were compared with the values calculated by a Hertz-Langmuir equation. The vaporization rate of the metal can be expressed by the Hertz–Langmuir relation (Eq. (1)) based on the kinetics of gases

$$M = (Pi - Pa)/(2\pi mRT)^{1/2} \qquad (1)$$

where M is the net vaporization rate, m is the molecular weight, R is the gas constant, T is temperature, Pi is the saturation vapor pressure of the gas i in the ambient space, and Pa is the partial pressure of the gas i in the ambient space.

Fig. 3 shows the amount of evaporated cadmium calculated by the Hertz-Langmuir relation and the experimental values. This theoretical value calculated by the Hertz-Langmuir relation is much higher than experimental value of the evaporated cadmium. This high value is probably explained by following; the Hertz-Langmuir equation assumes that the distillation rate is solely controlled by vaporization at the surface and ignores such factors as the rate of vapor transport, heat transfer at the surface. Moreover, parameters such as the heat transfers, retro-diffusion phenomena, impurities and mixing interactions are ignored by the Langmuir expression [5]. Thus the apparent rates are less than the theoretical rates. This deviation is compensated for by evaporation coefficient (*α*) obtained empirically. The evaporation coefficient is a function of temperature.
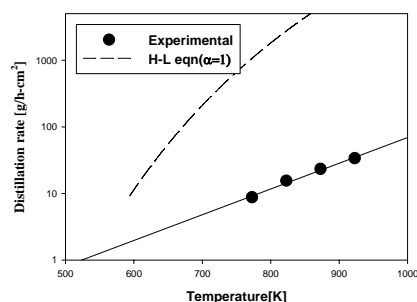

Fig.3. Comparison between the experimental and calculated distillation rates at 0.5torr.

4. Summary

Distillation experiments of cadmium were carried out to examine the effect of temperature and pressure on a cadmium distillation for the development of an actinide recovery process from a liquid cadmium cathode. The distillation rate of cadmium increased with an increasing temperature, whereas the distillation rate decreased with an increasing vacuum pressure. The optimum operating conditions should be found for the following ingot formation step. The theoretical value calculated by the Hertz-Langmuir relation was higher than experimental value of the evaporated cadmium.

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