

## Research Article

# Effect of Deposit on the Evaporation Rate of Adhered Salt in Uranium Dendrite

Sang Woon Kwon <sup>1</sup>, Si Woo Park,<sup>2</sup> and Sung Jai Lee<sup>1</sup>

<sup>1</sup>Korea Atomic Energy Research Institute, 989-111 Daedukdaero, Yuseong-gu, Daejeon 34057, Republic of Korea

<sup>2</sup>Wonjin Co., 17 Nonggong-gil, Boseong-gun, Jeollanamdo 59443, Republic of Korea

Correspondence should be addressed to Sang Woon Kwon; [swkwon@kaeri.re.kr](mailto:swkwon@kaeri.re.kr)

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Electrorefining is a key step in pyroprocessing. The solid cathode processing is necessary to separate the salt from the cathode of the electrorefiner since the uranium deposit in a solid cathode contains electrolyte salt. Moreover, it is very important to increase the throughput of the salt separation system due to the high uranium content of the spent nuclear fuel and high salt fraction of uranium dendrites. Therefore, in this study, the effect of deposit on the evaporation of the adhered salt in a uranium deposit was investigated by using the samples of salt in the uranium deposit and salt in the deposit of the surrogate material for the effective separation of the salt. It was found that the salt evaporation rate is dependent on the deposit type and bulk density in the crucible. Additionally, the evaporation rate was found to be lower when the deposit structure is complex; the rate also decreases as the bulk density of the deposit is increased owing to the retardation of the salt vapour transport process. It was concluded that the mass transfer of the salt vapour is an important parameter for the achievement of a high throughput performance in the salt distillation process.

## 1. Introduction

Pyroprocessing is a promising way for the recovery of actinide elements from the used nuclear fuel [1–4]. Electrorefining, which is a key process in pyroprocessing, generally comprises two recovery steps: (1) deposition of uranium onto a solid cathode and (2) the recovery of actinide elements by a liquid cathode. Because the uranium deposit in a solid cathode contains electrolyte salt, solid cathode processing is necessary to separate the adhered salt from the cathode.

The cathode deposit in pyroprocess is uranium dendrites coated by eutectic salt loaded with fission product chlorides. The purpose of the solid cathode processor is to remove entrained salt from the uranium electrodeposited and to consolidate the dendritic deposits. The cathode process has two separate steps of a salt separation and a consolidation of uranium deposits at the Korea Atomic Energy Research Institute.

A physical separation process, such as distillation separation, is more attractive than a chemical or dissolution process because physical processes generate much less secondary waste. Distillation processes are often preferred for cathode processing because of the following advantages: minimal generation of residual waste, a compact unit process, and simple and low-cost equipment [1–5]. As an example, in vacuum distillation separation, which is possible because of the difference in vapor pressures between salt and uranium, a solid cathode deposit is heated in a pre-determined heating region, thereby vaporizing the salt and leaving behind the nonvolatile uranium.

Several researchers have reported on the vacuum evaporation of salt or cadmium in pyroprocessing [6–12]. The efficiency of the vacuum distillation is dependent on the evaporation rate, transportation of vapor, and variables related to condensation and solidification [13, 14]. In addition, the vacuum level, crucible material, and heating and

cooling cycles play a major role in the optimization of the distillation process parameters.

Because of the high uranium content of the spent nuclear fuel and high salt fraction in uranium dendrites, it is critical to increase the throughput of the salt separation system. The evaporation rate of the LiCl-KCl eutectic salt in a vacuum distiller is not sufficiently high to satisfy the generation capacity requirements for the uranium dendrites in an electrorefiner. Thus, the evaporation rate of the salt in the uranium deposit is an important parameter that must be considered in the design and operation of the salt distiller. Although increased distiller throughput can be made feasible by employing a wide evaporation area or high distillation temperature, the realization of a wide evaporation area or high distillation temperature is complicated by various factors such as the material or structure of the distiller [15].

The distillation crucible can accommodate more uranium deposit if the deposit in the crucible is compressed. The compression increases the bulk density of the deposit. However, it is probable that the transport of the salt vapour is hindered due to the narrower path in the deposit heap by the compressing. The reduced transport of the salt vapour results in the decrease of the evaporation rate [13, 14]. Thus, the relationship of the bulk density and salt evaporation rate is important for the throughput of the salt distiller. The type of the deposit is also important due to the transport rate of the salt vapour between the deposit particles. The deposit particles with complex shape leads to the reduction of the salt evaporation rate due to the complicated path of the vapour in the heap of the deposit.

In this study, the effect of uranium deposit on the evaporation of the adhered salt was investigated to develop a salt separation system with a high throughput performance for the pyroprocess.

## 2. Materials and Methods

**2.1. Sample Preparation and Experimental Setup.** Figure 1 shows the experimental setup. The setup includes a distillation tower with an evaporator, a condenser, a control unit, and an off-gas treatment system. A load cell installed above the top flange of the distillation chamber was used to measure changes in the sample weight during the salt evaporation experiment.

Surrogate deposits were used for the investigation of type effect because the uranium deposits with various morphologies were difficult to prepare in the electrorefiner. The samples were prepared with LiCl-KCl eutectic salt powder and/or surrogate deposits to examine the effects of the deposit type on the evaporation rate under the condition of a 59/41 mole fraction ratio of lithium to potassium (lithium chloride-potassium chloride, Alfa aesar, chlorine 61.6%). Four types of samples were prepared for the investigation into the deposit type effect as summarized in Table 1.

- (i) Salt sample: 101.7 g of LiCl-KCl eutectic salt without deposit
- (ii) Steel ball sample: 100.1 g of salt with 100.6 g of steel balls

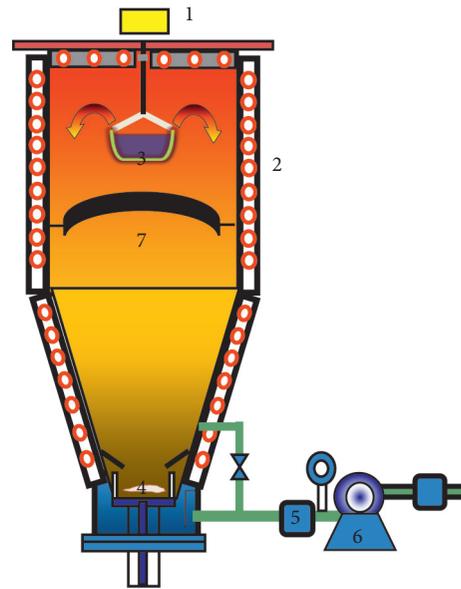


FIGURE 1: Experimental setup for the salt separation experiments (1: load cell, 2: heater, 3: uranium deposits, 4: recovered salt, 5: filter, 6: vacuum pump, and 7: heat shield).

- (iii) Steel chip sample (steel chip I): 150.4 g of salt with 50.1 g of steel chips
- (iv) Steel chip sample (steel chip II): 100.4 g of salt with 100.8 g of steel chips

The structure of the steel balls is much simpler than the genuine uranium deposits. The steel balls have a diameter of approximately 10 mm, as shown in Figure 2(a). The structure and length of the steel chips are more complicated than the uranium deposits. The steel chips have a length of 5–15 mm and width of 1–2 mm, as shown in Figure 2(b). The salt powder and surrogate deposits were well mixed to ensure homogeneity.

Four kinds of uranium deposit samples were prepared by using genuine uranium dendrites in order to investigate the significance of the deposit bulk density on the evaporation rate. The density of the samples was varied from 0.63 to 1.45 by pressing the samples with a spatula (150~200 g uranium deposits). The density was calculated as the deposit weight divided by the volume in the crucible. The uranium dendrites were produced by the uranium electrorefiner in the pyroprocessing facility at the Korea Atomic Energy Research Institute (Figure 2(c)).

**2.2. Salt Separation Experiments.** The crucible of the sample was made of stainless steel with a diameter and height of 58 mm and 80 mm, respectively, and was hung under the load cell rod through the top flange of the distillation tower. A rotary pump was used to evacuate the distillation chamber before heating. The samples were heated to 825°C at a rate of 5°C/min before allowed to settle for two hours. The pressure was maintained below  $2 \times 10^{-2}$  Torr during the salt evaporation.

TABLE 1: Types of samples used for the investigation of the type effect.

Sample name	Amount of eutectic salt (g)	Deposit (surrogate)	
		Type of deposit	Amount of deposit (g)
Salt	101.7	—	—
Steel ball	100.1	Ball	100.6
Steel chip I	150.4	Chip	50.1
Steel chip II	100.4	Chip	100.8



(a)



(b)



(c)

FIGURE 2: Photographs of the (a) steel balls, (b) steel chips, and (c) uranium deposit.

### 3. Results and Discussion

The effect of deposits on the evaporation behavior of the salt was investigated for several types of salt-deposit samples: a salt (LiCl-KCl eutectic) without deposit, a salt in steel balls, a salt in steel chips, and a salt in a uranium deposit. Figure 3 shows the profiles of the temperature and weight for a steel ball sample during the experiment. The apparent salt evaporation rates were calculated based on the slope of the weight reduction curve. The evaporation rate of the salt was not perfectly constant throughout the entire experimental period, even though the vacuum pressure and temperature were maintained at a constant level. Considering this, the averaged value was regarded as an apparent evaporation rate in the experimental range.

Figure 4 shows the apparent evaporation rates of the salt in various types of deposits. The apparent evaporation rate of the salt without the deposit was  $8.3 \text{ g/h-cm}^2$ . The apparent evaporation rate was slightly reduced to  $8.15 \text{ g/h-cm}^2$  in the steel ball sample. This very small reduction was likely related to the type of the deposits. The structure of the steel balls is much simpler than the uranium deposit, and the transportation of the salt vapour was hindered slightly by the steel ball deposits. During the distillation of the salt, most of the balls separated from the salt settled to the bottom of the crucible. Therefore, the evaporation surface area of the salt was nearly same as in the sample without deposits, as illustrated in Figure 5(b).

Two types of salt-steel chip samples were implemented in the experiments: steel chip I and steel chip II. The structure and length of the steel chips are more complicated than the

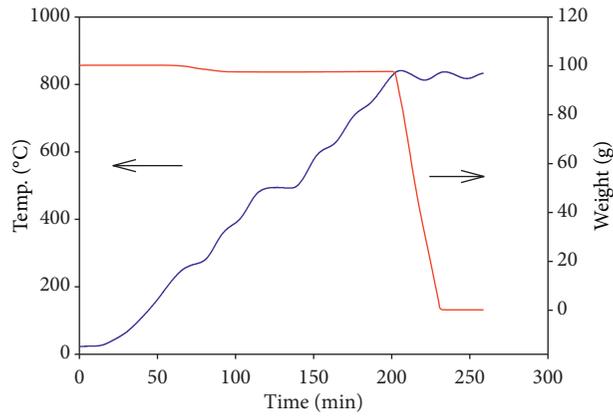


FIGURE 3: Profiles of sample weight and temperature as a function of time (steel ball sample).

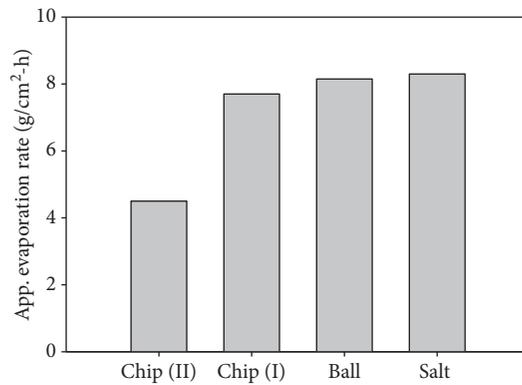


FIGURE 4: Comparison of apparent evaporation rates of LiCl-KCl eutectic salt in various surrogate deposits.

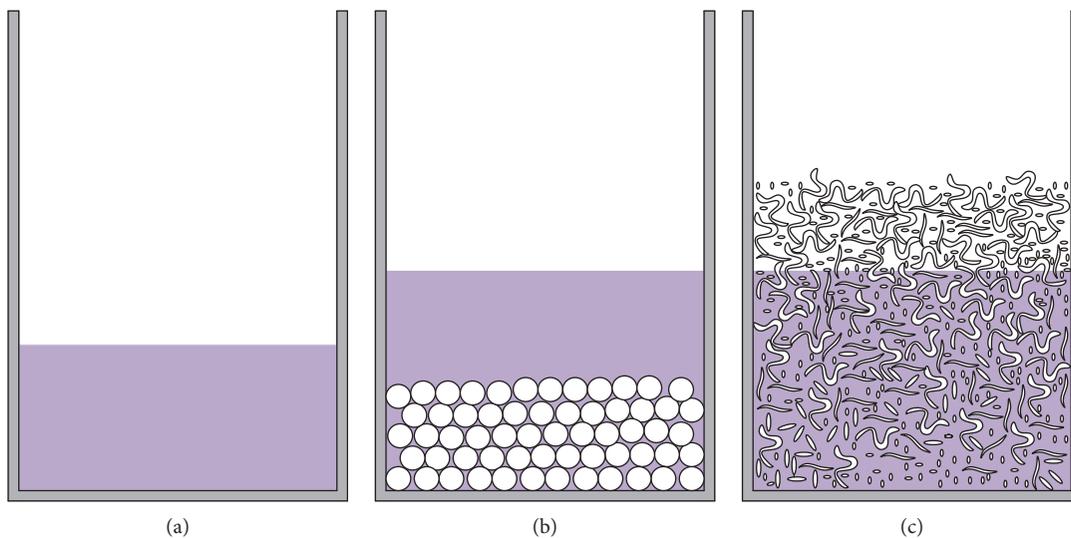


FIGURE 5: Diagrams of salt and deposits in the crucible during evaporation experiments for the samples comprising (a) salt, (b) salt and steel balls, and (c) salt and steel chips.

uranium deposits. The apparent evaporation rate for steel chip II, in particular, was considerably lower than that for the salt-steel ball sample or salt sample without deposits, as

shown in Figure 4. The apparent evaporation rate of the sample with a relatively small amount of steel chips (steel chip I) was 7.7 g/h-cm<sup>2</sup>, whereas the apparent evaporation

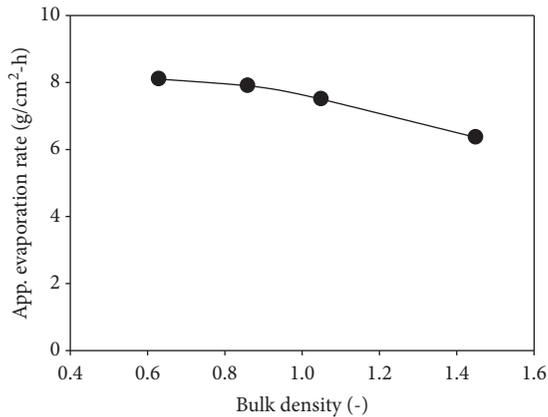


FIGURE 6: Relationship between the uranium deposit bulk density and apparent evaporation rates of LiCl-KCl eutectic salt.

rate was markedly reduced to  $4.5 \text{ g/h}\cdot\text{cm}^2$  as a result of incorporating a larger volume of steel chips (steel chip II).

The salt-steel chip samples likely have lower evaporation rates because of the reduced mass transfer rate, which is a consequence of the complex structure of the steel chips. The evaporation rate of the adhered salt was also found to be related to the transportation of salt vapour, as the transfer of salt vapour was hindered by the complex structure of the deposit, as illustrated in Figure 5(c). As can be seen, as the evaporation surface is always below the top surface of the steel chip layer, the pressure near the salt surface was increased as a result of the retardation of the expulsion process of the salt vapour to the outside of the crucible. It was probable that the evaporation rate decreased because of the low transfer rate of salt vapour. From the abovementioned results, it could be concluded that the evaporation rate is affected significantly by the type of the deposit.

The effect of bulk density on the apparent evaporation rate was also investigated by using the uranium deposit samples produced by electrorefining. The density of the uranium deposit in the crucible was varied in the range of 0.63~1.45. The results demonstrated that the evaporation rate increased as the bulk density of uranium deposits was reduced, as shown in Figure 6. Additionally, the reduced evaporation rates at higher bulk densities are presumed to be due to the narrow path of the salt vapour, which reduces the mass transfer rate. However, an increase in the bulk density results in an increase in the throughput of the salt distiller; this is because the loading capacity of the deposit correspondingly increased as the bulk density in the distillation crucible was increased. These results demonstrate that it is necessary to optimize the bulk density of uranium deposits in the distillation crucible.

From the abovementioned results, it was found that the salt evaporation rate is dependent on the deposits in the crucible because the evaporation rate of the adhered salt influences the expulsion of the salt vapour. Thus, it could be concluded that the mass transfer of the evaporated salt is an important parameter needed to enhance the throughput of the distilled salt adhered to cathode deposits. Therefore, the distiller geometry must be designed to optimize mass

transfer such that high throughput performance can be achieved in the salt separation process.

#### 4. Conclusions

In this study, the effect of uranium deposit on the evaporation of the adhered salt was investigated to develop a salt separation system with a high throughput performance for the pyroprocess. The evaporation rate was found to be dependent on the type of the deposit. Additionally, a more complex deposit structure was found to correspond to a lower evaporation rate, which can be further reduced by increasing the bulk density of the deposit. It is presumed that the evaporation rate of the adhered salt is also related to the transportation of salt vapour, as the expulsion of salt vapour was found to be hindered by a complex deposit structure or a narrow path for the vapour transfer. The reduced evaporation rate was a consequence of a reduced mass transfer rate. Therefore, it could be concluded that the distiller geometry must be designed to optimize mass transfer such that high throughput performance can be achieved in the salt separation process.

#### Data Availability

The data used to support the findings of this study are included within the article.

#### Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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