

Development of an integrated sieve-crucible assembly for the sequential operation of liquid salt separation and distillation

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Abstract

The capacity of a salt distiller should be sufficiently large to produce the required throughput for the uranium electro-refining process. In this study, an assembly composed of a liquid separation sieve and a distillation crucible was developed for the sequential operation of liquid salt separation and vacuum distillation in the same tower. The feasibility of the sequential salt separation was examined by the rotation test of the assembly and sequential operation of a liquid salt separation – vacuum distillation. The adhered salt in the uranium deposits was removed successfully. The salt content in the deposits was below 0.1 wt% after the sequential operation of the liquid salt separation – salt distillation. The results of this study show that efficient salt separation work can be realised by the sequential operation of liquid salt separation – vacuum distillation in one distillation tower because the operation procedures are simplified and no extra operation of cooling and re-heating is necessary for the shift of the liquid salt separation step to the vacuum distillation step.

Introduction

The cathode deposit in the pyroprocess is uranium dendrites coated with 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 process. Distillation process was employed for the cathode processing due to the advantages of minimal generation of secondary waste, compact unit process, simple and low-cost equipment [8]. The basis for vacuum distillation separation is the difference in vapour pressures between a salt and uranium. A solid cathode deposit is heated in a heating region and salt vaporises, while non-volatile uranium remains behind.

Some studies have reported on the vacuum evaporation of the salt or cadmium in the pyroprocess [2] [4] [6] [7]. The efficiency of the vacuum distillation depends on evaporation rate, transportation of vapour, condensation and solidification. In addition, vacuum level, material of crucible, heating and cooling cycles play a major role in deciding optimum distillation process parameters.

The salt separation process is a crucial step since the process could be a bottle neck of the pyroprocess. It is very important to produce the required throughput for the salt separation system due to the high-uranium content of spent nuclear fuel and high salt fraction of uranium dendrites. The evaporation rate of the LiCl-KCl eutectic salt in a vacuum distiller is not high enough to produce the required generation capacity for the uranium dendrites in the electro-refiner. An increase in the distiller throughput could be made possible by the wide evaporation area or high distillation temperature. But the realisation of the wide evaporation area or high distillation temperature is limited by various factors such as material or structure of a distiller.

Kwon *et al.* has proposed a combined process to produce the required throughput for the salt removal process by the separation of the liquid salt prior to the distillation of the LiCl-KCl eutectic salt from the uranium deposits [5].

In this study, an assembly composed of a liquid separation sieve and a distillation crucible was developed for the sequential operation of the liquid salt separation – vacuum distillation in the same tower. The feasibility of the assembly was examined by the rotation test and sequential operation of a liquid salt separation – vacuum distillation.

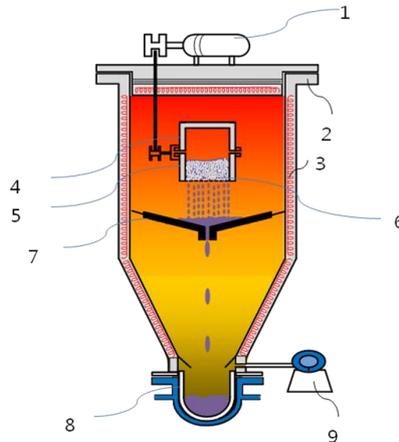
Experimental

Figure 1 shows a distillation tower for the sequential operation of the liquid salt separation – vacuum distillation. The distiller was composed of a distillation tower with an evaporator, a condenser, a control unit, and an off-gas treatment system. The assembly composed of a liquid separation sieve and a distillation crucible was hung under the top flange of the tower. The sieve was made of stainless steel mesh with an opening of 150 μm .

The feasibility of the assembly was examined by the rotation test and sequential operation of liquid salt separation and vacuum distillation. The salt separation experiments were carried out with uranium deposits, where the content of salt in the deposits was about 40 wt%. About 80 g of uranium deposits was put in the crucible and heated at 600°C for 30 minutes for liquid salt separation and the assembly was rotated. The temperature was increased to 800°C for the distillation of salt. The weight of uranium deposits was measured before and after distillation. The deposit samples were

dissolved into the nitric acid and the salt content was measured by using ICP spectroscopy.

Figure 1: Experimental set-up for the salt distillation

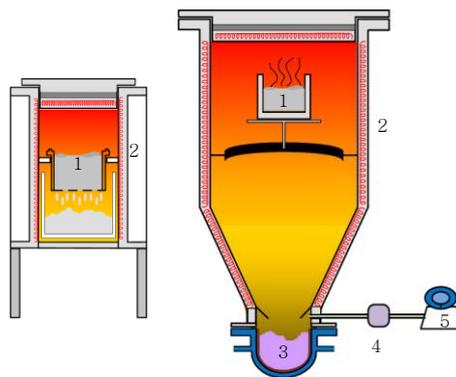


Note: 1: motor, 2: distillation tower, 3: heater, 4: crucible, 5: sieve, 6: uranium deposit, 7: heat shield, 8: recovered salt vessel, 9: vacuum pump.

Results and discussion

In the electro-refiner of the pyroprocess, the uranium deposits have more than 20 wt% of the electrolyte eutectic salt [6]. The capacity of a salt distiller should be sufficiently large to produce the required throughput for the uranium electro-refining process. To achieve a high throughput performance in the salt separation process, Kwon *et al.* proposed a method to combine liquid salt separation and vacuum distillation as shown in Figure 2 [5]. The salt separation system is composed of a liquid salt separation column and a vacuum distiller. In the combined process, the adhered salt is separated by heating on the sieve at a relatively low temperature compared to the operation temperature of the vacuum distiller, and then the residual salt is evaporated in the salt distiller at an elevated temperature.

Figure 2: Schematic of the salt separation system composed of (a) liquid salt separation column and (b) vacuum distiller



Note: 1: uranium deposit, 2: heater, 3: recovered salt, 4: filter, 5: vacuum pump.

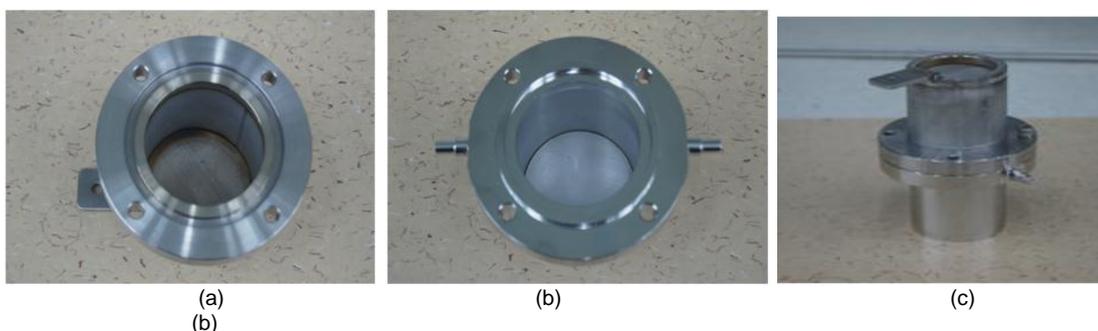
Uranium deposits in the sieve should be transferred to a ceramic coated crucible for the shift of the liquid salt separation step to the distillation step, since the iron-based alloy is not generally used as a crucible material to avoid the eutectic alloy formation between iron and uranium at high temperatures. For the transfer of uranium deposits, it

is necessary to cool the distillation tower. After the transfer of the uranium deposits, the distillation tower is to be reheated for the distillation of the salt.

In this study, an assembly composed of a liquid separation sieve and a distillation crucible was developed for the sequential operation of liquid salt separation – vacuum distillation in the same distillation tower without cooling and reheating of the chamber. The transfer of uranium deposits can be simplified by the rotation of the assembly.

The integrated sieve-crucible assembly was composed of a liquid separation sieve and a distillation crucible as shown in Figure 3. First, the uranium deposits are placed on the sieve and salt is separated to some extent as a liquid from deposits at lower than the distillation temperature. And then, the assembly is rotated (crucible downward) and the deposits fall down to the crucible. The salt in the deposits of the crucible is distilled at higher than 800°C.

Figure 3: Photographs of a sieve, a crucible and an assembly for the salt separation from the uranium deposits a) sieve for liquid salt separation, b) crucible for salt distillation and c) sieve – crucible assembly

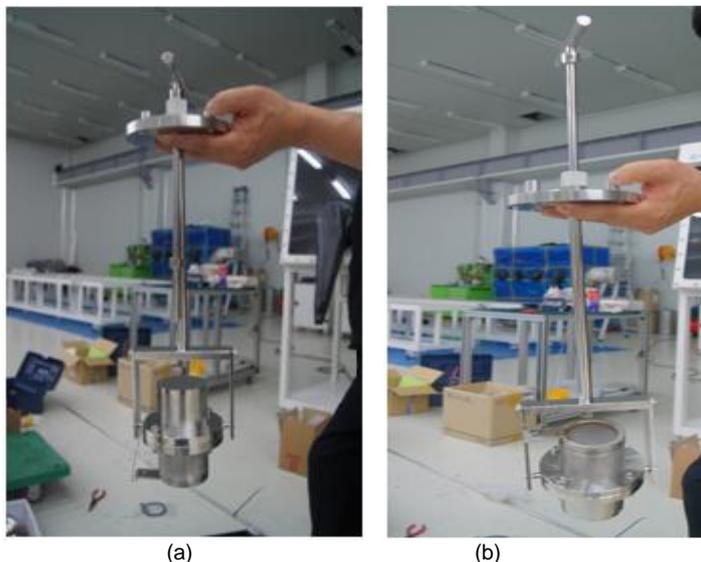


The feasibility of the integrated sieve-crucible assembly was examined through the rotation test of the assembly by the sequential operation of liquid salt separation – vacuum distillation in the distillation tower. Figure 4 shows a rotation test with a blank integrated sieve-crucible assembly. Initially, the pulling bar is placed at the low position. The assembly was rotated when the pulling bar was pulled up and the bar was in the upper position. It was found that the assembly rotated successfully with the aid of a pulling bar and a wire.

Figure 5 shows an integrated sieve-crucible assembly placed under the top-flange of the vacuum distillation tower. As shown in Figure 5, the assembly started to rotate when the pulling bar was lifted upward. The deposits in the sieve fell down into the crucible after the rotation of the assembly.

The vacuum level is important during the entire operation period of the vacuum distillation tower [1]. For the sequential operation of the integrated column, it is necessary to rotate the sieve- crucible assembly. The external movement should be transferred into the internal assembly through a column wall. Therefore, the preservation of the vacuum seal is very important during the rotation of the assembly to avoid a degradation of the vacuum level by the leak.

**Figure 4: Rotation test with a blank integrated sieve-crucible assembly
(a) sieve downward, (b) crucible downward**

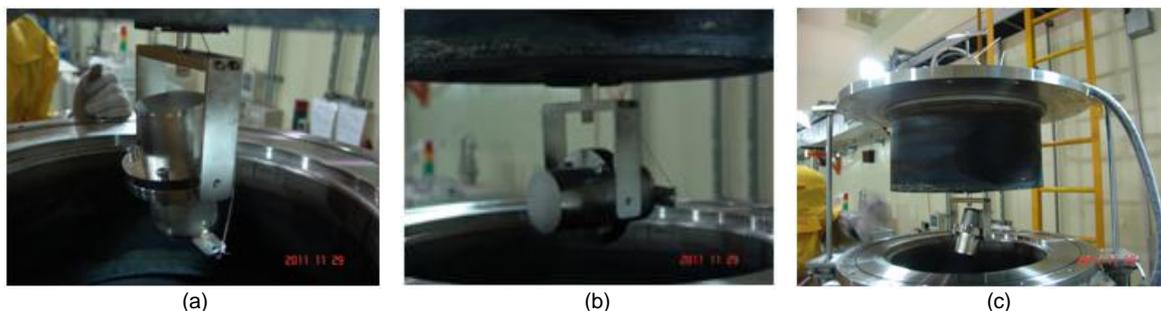


A method to preserve vacuum level was proposed and applied to the distiller in this study. For the rotation of the assembly in the distillation tower, the pulling bar placed outside the tower was connected to the assembly with a wire through the flange wall of the tower. The seal was assured by two O-rings during the movement of the connecting line through the wall of the flange. The gap between the guide bar and the hole of the top flange was sealed using a double O-ring. The O-rings were cooled by water to avoid degradation by heat.

The integrated assembly was tested in the distillation tower as shown in Figure 5. The assembly turned over successfully and the uranium deposits fell down from the sieve to the crucible. The leak of air was monitored using a vacuum pressure sensor after isolation of the vacuum pump by closing a valve. No signal was found about an air leakage during the test.

The experiment on the sequential operation of the liquid salt separation and salt distillation was carried out using an integrated sieve-crucible assembly. The uranium deposits are placed into the sieve side of the assembly and the adhered salt is separated by heating on the sieve. Then, the uranium deposits move to the crucible by the rotation of the sieve-crucible assembly and the residual salt is evaporated at an elevated temperature. The remaining salt in the uranium deposits was further separated by evaporation in the distillation tower for two hr at 850°C. The adhered salt in the uranium deposits was removed successfully. The salt content in the deposits was below 0.1 wt% after the sequential operation of the liquid salt separation and salt distillation. This residual salt after the salt separation process can be removed completely during the melting of the uranium metal in the following ingot preparation process.

Figure 5: Rotation of the integrated sieve-crucible assembly in the top flange of the vacuum distillation tower (a) sieve downward, (b) rotation of the assembly, and (c) crucible downward



From the above results, it could be concluded that efficient salt separation work was realised in an integrated sieve-crucible assembly by the sequential operation of liquid salt separation and vacuum distillation because the operation procedure is simplified and there is no need for further cooling and heating operations. It should be noted that the sequential operation of liquid salt separation and vacuum distillation using an integrated sieve-crucible assembly is one of the most effective ways to achieve a high throughput performance in the salt separation process.

Conclusion

In this study, it was proposed to produce the required throughput for the salt removal process by the separation of the liquid salt prior to the distillation of the LiCl-KCl eutectic salt from the uranium deposits of the solid cathode in an electro-refiner. The feasibility of liquid salt separation was examined by salt separation experiments on a stainless steel sieve. The adhered salt in the uranium deposits was separated as a liquid by heating the deposits at a low temperature. Therefore, the amount of salt to be distilled can be reduced by liquid salt separation prior to the salt distillation. It was found that the liquid salt can be separated from the uranium dendrites above 500°C. After the liquid salt separation had been removed by the vacuum distillation, the residual salt remained in the uranium deposits. It was concluded that the sequential operation of a liquid salt separation and a vacuum distillation using an integrated sieve-crucible assembly is one of the most effective ways to achieve a high throughput performance in the salt separation process.

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