Uranium removal from the waste oil containing uranium by supercritical R-22

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1. Introduction

The waste oil used in the fuel processing plant sometimes contain uranium compounds because of the exposure of the oil to the uranium contained materials or environments. The waste oil containing uranium is collected and stored in the radiation controlled facility. The waste oil containing uranium is very difficult to be disposed in the radioactive waste disposal site under the current regulation law. The best and only method is to remove the uranium compounds from the waste oil containing uranium, and to dispose the cleaned oil by incineration. To satisfy the current regulation, the radioactivity of the waste oil should be from 0.0675 Bq/g to 0.0661 Bq/g in the case of mixed natural uranium and 5% enriched uranium, respectively. The radioactivity of the waste oil containing uranium stored in the radiation controlled facility ranged from 2 Bq/g (20 ppm) to 25 Bq/g (250 ppm). So, disposal of the waste oil containing uranium needs a process to remove uranium compounds from the waste oil containing uranium.

We had developed the decontamination process for radioactive waste oil containing cobalt by supercritical R-22 (chlorodifluoromethane) in our previous study[1]. The critical temperature of R-22 is 96.15℃ and the critical pressure of R-22 is 49.9 bar. The supercritical R-22 is excellent solvent to extract non radioactive pure oil components from radioactive waste oil.

In this study, we developed a removal process of uranium from the waste oil containing uranium using supercritical R-22.

2. Methods and Results

2.1 Chemicals

The R-22 was purchased from Dong A chemical Co. The chemical structure of R-22 was shown as Fig. 1. The nitric acid and other chemicals were used general grade reagents. The waste oil containing uranium which generated from nuclear fuel processing facilities was used.

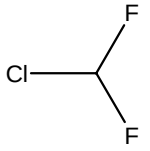

Fig. 1. Chemical structure of R-22

2.2 Equipments

The oil extraction equipment to extract pure oil components from the waste oil containing uranium manufactured shown as Fig. 2. It composed of a high pressure syringe pump, a preheating coil with 1/16 in * 20 m stainless steel tube, an extraction vessel with 120 ml stainless steel and a separator. The extraction vessel was wrapped with electric band heater and controlled by temperature controller. The experimental ranges of pressure and temperature were up to 150 bar and 150 ℃.

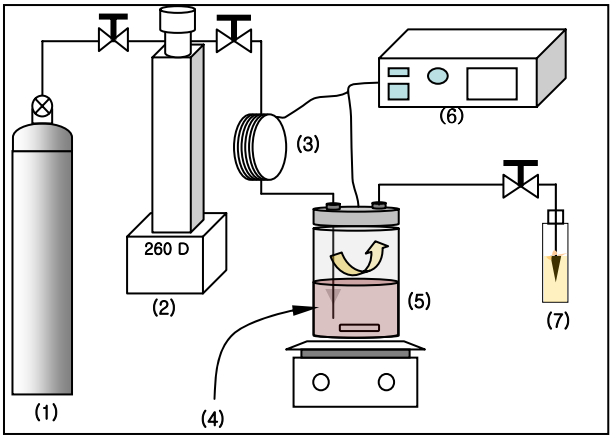

Fig. 2. The oil extraction equipment by supercritical R-22.
(1) R-22 tank (2) syringe pump (3) preheating coil
(4) extraction vessel (5) electric band heater
(6) temperature controller (7) separator

2.3 Experiments

At the first experiment, the removal efficiencies of uranium from the waste oil containing uranium by R-22 extraction and nitric acid treatment were determined. We removed uranium from the waste oil containing uranium by three steps. The first step is to extract pure oil from the waste oil containing uranium by supercritical R-22. The second step is to remove uranium from the 1st extracted oil by nitric acid. The third step is to extract pure oil from the 2nd treated oil by supercritical R-22. Firstly, the 55 g of the waste oil containing uranium was replaced in extraction vessel. The waste oil contained extraction vessel was heated by electric band heater to 100 ℃. The R-22 was introduced to the extraction vessel and pressurized to 75 bar by pressure syringe pump. After 20 min, the R-22 was introduced to the extraction vessel continuously with 4 ml/min of flow at 105 ℃, 75 bar and then

discharged with extracted oil. The extracted oil was accumulated in separator. The 1st extracted oil containing uranium by supercritical R-22 was analyzed by ICP-MS. Secondly, the 35g of the 1st extracted oil was placed in glass beaker with 35 g of 6M HNO_3. After 1 hour stirring with magnetic bar at room temperature, the treated oil by nitric acid was separated to other beaker. The 2nd treated oil containing uranium by nitric acid was analyzed by ICP-MS. Thirdly, the 2nd treated oil of 20 g was replaced in extraction vessel. The pure oil was extracted by supercritical R-22 same as the 1st extracted conditions. The 3rd extracted oil containing uranium by supercritical R-22 was analyzed by ICP-MS.

At the second experiment, the removal efficiencies of uranium from the 1st extracted oil according to nitric acid volume ratio to the 1st extracted oil and stirring time of nitric acid mixed the 1st extracted oil solution were determined. Firstly, the 15 g of the 1st extracted oil and 15 g of 3M HNO_3 were placed in glass beaker and then stirred with magnetic stirrer at room temperature during 1 hour. Secondly, the 15 g of the 1st extracted oil and 15 g of 3M HNO_3 were placed in glass beaker and then stirred with magnetic stirrer at room temperature during 2 hours. Thirdly, the 15 g of the 1st extracted oil and 30 g of 3M HNO_3 were placed in glass beaker and then stirred with magnetic stirrer at room temperature during 1 hour.
Three treated oils by nitric acid were separated to other beaker and three 2nd treated oils containing uranium were analyzed by ICP-MS, respectively.

2.4 Results and discussion

The removal efficiencies of uranium from the waste oil containing uranium by R-22 extraction and nitric acid treatment were summarized at Table 1. The removal efficiency of uranium from waste oil is highest in the 3rd extraction oil.

Table 1. Removal efficiency of uranium from waste oil by R-22 extraction and nitric acid treatment

	Concentration of U (ppm)	Removal efficiency of U (%)
waste oil containing U	61.9	-
1st extracted oil by R-22	4.72	92.4
2nd treated oil by HNO_3	1.57	97.5
3rd extracted oil by R-22	1.08	98.3

The removal efficiencies of uranium from the 1st extracted oil according to nitric acid volume ratio to the 1st extracted oil and stirring time of nitric acid mixed the 1st extracted oil solution were summarized at Table 2. The removal efficiencies of uranium from the 1st extracted oil are higher at higher volume ratio of 3M HNO_3 to the 1st extracted oil and long stirring time of nitric acid mixed the 1st extracted oil solution.

Table 2. Removal efficiency of uranium from the 1st extracted oil according to volume ratio and stirring time with 3M HNO_3.

	Concentration of U (ppm)	Removal efficiency of U (%)
1st extracted oil	14.6	-
volume ratio (1:1) stirring time (1 hr)	2.48	83.0
volume ratio (1:1) stirring time (2 hrs)	1.84	87.4
volume ratio (1:2) stirring time (1 hr)	1.52	89.6

3. Conclusions

Firstly, the uranium from the waste oil containing uranium was removed by extraction of supercritical R-22. But this extraction has limit to removal of uranium from the waste oil containing uranium. The waste oil containing uranium contained about 20 wt% of lube oil additives like dialkyldithiophosphates compound. The uranium compounds in waste oil seem to be combined with lube oil additives and then produce uranium chelating complexes. Because these uranium chelating complexes seem to be extracted by supercritical R-22, we assumed that some quantity of uranium was contained in the 1st extracted oil by supercritical R-22.

Secondly, the uranium from the 1st extracted oil containing uranium was removed by treatment of nitric acid. Because the uranium in the 1st extracted oil dissolved into aqueous nitric acid solution, the uranium concentration of the 2nd treated oil was reduced. In the case of uranium removal from the 1st extracted oil by nitric acid, uranium much removed at higher nitric acid concentration, higher nitric acid volume ratio to the 1st extracted oil and longer stirring time of nitric acid mixed the 1st extracted oil solution.

Thirdly, the uranium in the 2nd treated oil was removed by extraction of supercritical R-22 once more.

Through this study, we found that uranium removed from the waste oil containing uranium by R-22 extraction and nitric acid treatment but we have to study to remove uranium from the waste oil containing uranium in order to dispose the cleaned oil by incineration.

REFERENCES

[1] Jaeryong Yoo, Jinhyun Sung, Kwangheon Park, Hongdoo Kim, Hakwon Kim, Taeyoon Lim, Sang hak Yim, Weonseob Yoon, Purification of radioactive waste oil by a supercritical fluid, Transactions of the Korean Nuclear Society Spring Meeting, Korea, May 25-26, 2006