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Separation of paraffin from the paraffin waste forms using a supercritical fluid

Jina Jang,a Seongwoo Yang,a Minsu Ju,b Kwangheon Park,a
a Green Nuclear Research Laboratory, Kyung Hee University, Suwon, 449-701, kpark@khu.ac.kr
b 25-1, Jang-Dong, Yuseong-Gu, Daejeon, 305-343, KHNP Nuclear Engineering & Technology Institute

1. Introduction

Nuclear energy currently becomes revitalized as a main energy source due to the rapid rise of oil prices and the growing demand for clean energy sources that do not produce carbon dioxide. However, nuclear energy indispensably produces radioactive nuclear wastes which should be very carefully kept in separation from the boundary of human environments for a long time.

Radioactive nuclear wastes should be maintained in a good stability against leaking out for a long period. For this purpose, nuclear power plants generally use a method of the solidification of radioactive wastes for disposal of intermediate and low level radioactive waste. Two forms of solidification are used- cement and paraffin. These waste forms must have good structural durability to allow for permanent disposal under even unfavorable conditions. In contrast to the cement waste form, the paraffin waste form has a high leaching rate[1]. A relatively low stability of the paraffin waste form makes it difficult to use for long-term storage. Therefore in order to solve this leaching problem, it may be recommended to separate the paraffin from the radioactive sludge in paraffin waste form above all. Then, the radioactive sludge can be processed into a more stable form of disposable wastes such as cement waste forms.

Paraffin is a group of alkane hydrocarbons with the general formula C_nH_{2n+2}, where n is greater than about 20. It is insoluble in water, but soluble in ether, benzene, and certain esters. Paraffin is unaffected by most common chemical reagents, but burns readily. Paraffin has the typical specific gravity of 0.9~1 and melts at 40~70℃[2].

We studied the extraction of paraffin using a supercritical fluid, because supercritical fluid is highly effective in selective extraction of a certain substance. This supercritical fluid has good penetration ability like gases with a good capability of containing high concentration of target substances like liquids. There are many applications exploiting the advantages of these properties [3,4]. We applied supercritical fluid in separation paraffin from radioactive sludge in the paraffin waste forms.

2. Experiment and results

2.1 Apparatus

We measured the solubility of paraffin in supercritical fluid. The experimental apparatus was comprised of a liquid syringe pump and an extraction cell with a thermo-controller. The ranges of pressure and temperature went up to 200bar and 200℃, respectively (Fig. 1). Operating temperature was controlled by the thermo-controller. The solubility of the paraffin in a fluid was measured by the optical observation in the variable volume cell by changing the volume and the pressure under the inserted fluid. By injecting the amount of the fluid with volume expansion, we were able to continuously find the cloud points.

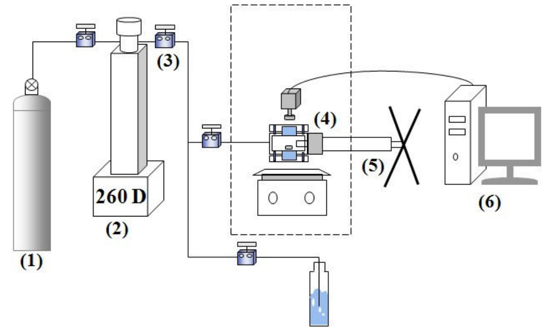

Fig.1 Solubility measurement apparatus (1) Cylinder (2) Syringe pump (3) Inlet valve (4) Variable cell (5) Rotator (6) Monitor

2.2 Solubility of paraffin in liquid/supercritical fluid

The solvent ratio (mass of solvent needed for extraction of unit mass of target material) of paraffin in CO_2 was measured. Liquid CO_2 has higher solvent ratio (Fig.2). The phase change was easily noticed. As the pressure increased, the solvent ratio of the paraffin decreased in both the liquid and supercritical CO_2. But the solvent ration turned out to be more than a thousand. This substantial amount of solvent required is a weak point for the use CO_2 as an extracting fluid.

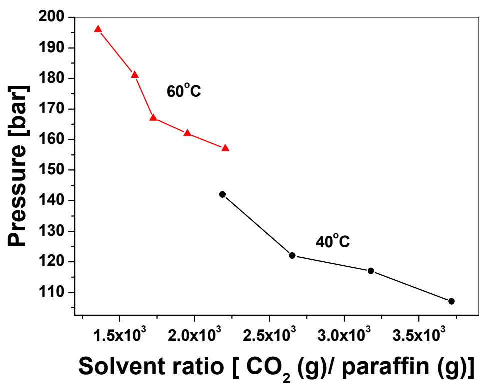

Fig.2 Paraffin solubility in CO_2

In order to get lower solvent ratio, an experiment was carried out using a R22. R22 turned out to be a good fluid to extract paraffin. The solvent ratio depends on the physical state of paraffin. The paraffin solubility changed little relative to pressure when it melted at 60℃, while higher pressure was needed in the solid state. The paraffin in R22 relatively had a much lower solvent ratio than that in the CO_2. The solid paraffin at 40℃ dissolved more slowly than the liquid state (Fig.3). Based on these solubility results, it was better to extract paraffin in liquid state at the higher temperature (greater than 60℃). Higher density of the fluid is the better to dissolve the paraffin at a given fluid amount.

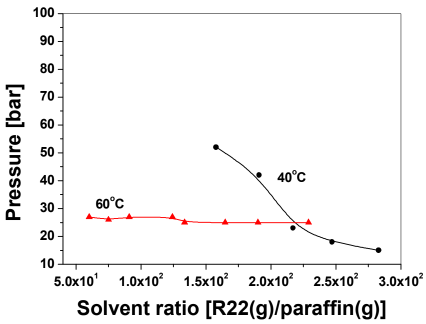

Fig.3 Paraffin solubility in R22

2.3 Extraction of paraffin

Based on the solubility result, we extracted the paraffin using R22. This extraction experiment was comprised of both static extraction and dynamic extraction. The volume of the extraction cell was 125ml and the density of R22 at 60℃ was 1.06g/ml, so the amount of R22 was 132.5g. Five grams of the paraffin were put into the extraction cell, which was then heated up to 60℃. The temperature was maintained. When 150ml of R22 was extracted, 0.6g of paraffin remained. So, more than 90% had been extracted. As a more dynamic extraction process was needed, to fully remove the paraffin, we proceeded to carry out an extraction experiment using a 330ml of R22. This completed eliminated the paraffin.

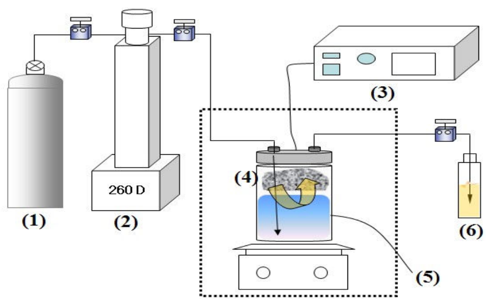

Fig.4 Extraction apparatus (1) Cylinder
(2) Syringe pump (3) Thermal controller
(4) Extraction cell (5) Specimen (6) Collector

3. Conclusion

To separate paraffin from the radioactive sludge in paraffin solid forms, we tested the feasibility of extraction of paraffin using supercritical fluid. R22 turned out to be a workable fluid. The solubilities of paraffin in both CO_2 and R22 were measured. The experiment confirmed the successful separation of paraffin be possible if we use R22 as a working supercritical solvent.

Acknowledgment

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REFERENCES

[1] J.M. Lee, A study on the characteristics of solidified waste form produced at Korea nuclear power plant and the durability of disposal facility for the safety assessment of low and intermediate level radwaste disposal, p. 75, 2002.
[2] J.M. Lee, A study on the characteristics of solidified waste form produced at Korea nuclear power plant and the durability of disposal facility for the safety assessment of low and intermediate level radwaste disposal, p. 57, 2002.
[3] L. T. Taylor, supercritical fluid cleaning – Fundamentals, technology and applications, Noyes Publications, New Jersey, 1998.
[4] M. D. Luque de Castro, M. Valcarcel, and M. T. Tena, Analytical supercritical fluid extraction, Springer-Verlag, Berlin Heidelberg, 1994.