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Purification of radioactive waste oil by a supercritical fluid

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1. Introduction

The radioactive waste oil from the nuclear industry is potentially hazardous due to its possibility to contaminate soil and underwater. Pollutants in waste oil are generally radioactive heavy metals or organo-metals. Radioactive waste oils are highly viscous fluids that are similar to used-motor oils. Several processes have been developed to regenerated used motor oil, such as acid-clay treatment, chemical addition, vacuum distillation, thermal cracking and hydrofinishing. However, these technologies are difficult to apply to separating radioactive nuclides from radioactive waste oils. In recent years, our laboratory developed a membrane method for the regeneration of used motor oils [1]. We applied supercritical CO_2 ($scCO_2$) as a viscosity-reducing additive to waste oils at a lower process temperature in order to improve membrane permeability and thus the energy saving [2, 3, 4]. However, the membrane cannot filter the contaminants in radioactive waste oil that are not particles, such as radioactive ions in impurity water in the oil. In this paper, we suggest a method extracting clean oil from the radioactive waste oil rather than filtering by a supercritical fluid. We selected R22, a refrigerant, as a solvent for extraction. R22 has a mild critical point - 96.1℃ and 49.9bar. Regeneration of waste oils by extracting clean oil using a supercritical fluid such as R22 is easy to handle and reduce secondary wastes. In this paper, we examine the feasibility of R22 in extracting clean oil from radioactive waste oils.

2. Methods and Results

2.1 Apparatus

The experimental apparatus contains a liquid syringe pump, an extraction cell with a preheating line and a collector. The design pressure and temperature range up to 200bar and 200℃ (Fig 1). The volume of the extraction cell is 150ml. The extraction cell is manufactured by HANWOUL ENGINEERING CO. The preheating line was used in order to ensure constant operating temperature. It was rolled tube 1/16 inch dia. The radioactive waste oil was obtained from Ulchin nuclear power plant.

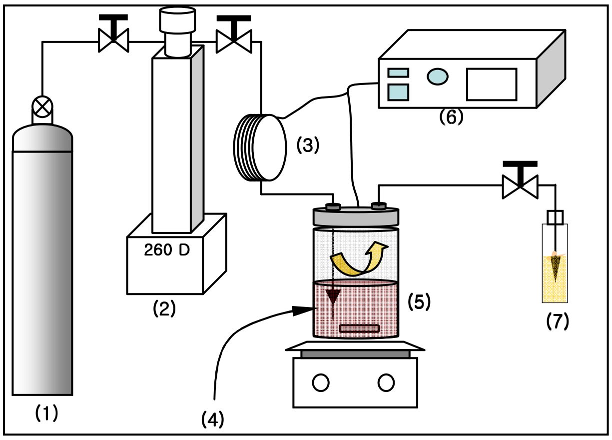

Figure 1.(1) R22 Tank (2) Syringe pump
(3) Preheating (4) Waste oil
(5) Extraction cell (6) Thermo-controller
(7) Collector

2.2 Experiments

Before the extraction test, first we measured the solubility of the oil in $scCO_2$ and in R22. Figure 2 shows the results. The pressure above the solubility line is needed for the formation of the single phase of the oil and the fluid. It was found that oil required higher pressure in $scCO_2$ than in R22 to reach the same solubility in the fluid. Lower solubility means larger amount of solvent is required for extraction, or higher 'solvent ratio' is needed. The solvent ratio refers to the required mass of solvent per unit mass of extracted oil.

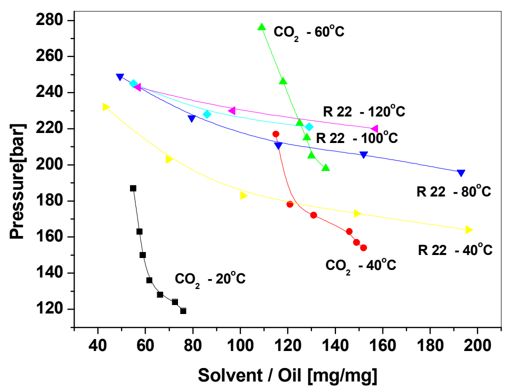

Figure 2.Comparison of solubility oil in $scCO_2$ and scR22

The density of the oil (0.84g/ml) is heavier than that of R22 (0.65-0.75g/ml). At first, we inserted the waste oil (86ml) into the extraction cell, and then supplied R22 up to the experimental pressure by a syringe pump. We set the temperature, 100℃-110℃ and the pressure, 60bar -70bar. Thermocouples were inserted into the extraction cell and the preheating line. The temperature of the extraction cell containing R22 and radioactive waste oil were controlled by PID temperature controller. After 10 minutes of stabilization, clean oil was extracted by R22 from the waste oil. The flow rate of R22 was under 10ml/min, to prevent overflowing out of mixture of the oil and R22.

2.3 Results and discussion

Since the density of the radioactive waste oil is higher than supercritical R22, the oil is at the bottom and the clean oil (hydrocarbon) is dissolved into and extracted by R22 that is at the top.

Gamma ray activity of the waste oil was measured by a HP Ge gamma detector before the experiment. Several gamma rays were detected including Co-60. The gamma activity of Co-60 was 88mBq/55ml in the radioactive waste oil. Gamma activity of the extracted oil was also measured. After extraction the activity of Co-60 was below the MDA (Minimum Detectable Activity), which means regeneration of radioactive waste oils is possible by this R22-extraction method. The extracted oil shows a different color (brighter than waste oil) as shown in Figure 3. The original color of radioactive waste oil is somewhat dark. The extracted oil has the lighter color, close to that of pure oil. Depending on the extracting condition, the color of extracted oil changes, which indicates the fractionation of oil is possible by this method.

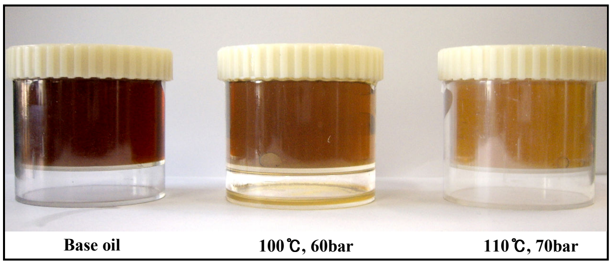

Figure 3.Comparison of base oil and extracted oil

3. Conclusion

The feasibility of the extraction of clean oil from the radioactive waste oil was tested. R22 turned out to be a good solvent for extraction. The extracted oil was clean of gamma radioactivity. The color of extracted oil changes depending on the extraction condition. Fractionation of oil seems possible by this method. We can also apply this extraction method for regeneration of conventional waste oils such as waste car oils.

Acknowledgment

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