Feasibility study of supercritical fluid extraction for the management of liquid and solid radioactive waste from uranium-plutonium fuel fabrication

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Supercritical fluid extraction (SFE) is a fairly common process used primarily in the food, pharmaceutical and petrochemical industries. SFE use in the nuclear industry is limited due to high operating pressure (i.e. 73 atm for CO_2 and 4,9 atm for CF_3CFH_2). This contradicts the principle of operation of technological devices under depression, which is used in radiochemical industries to maintain radiation safety.

However, SFE possesses such features as no trace of solvent in the final product, higher productivity of the technology compared to traditional liquid extraction, and the ability to adjust the solvent capacity of a supercritical solvent in a relatively wide range by changing of pressure and temperature. All this make the SFE process attractive for use in the spent nuclear fuel reprocessing and, above all, in the management of radioactive waste.

The purpose of this work is to identify the possibility of using SFE to reduce secondary waste and to conduct preliminary experiments.

In this work the review of the current state of SFE application in the field of nuclear energy is carried out. Moreover, the basic block diagram of radioactive waste purification and decontamination of metallic radioactive waste from the microquantities of uranium and plutonium is developed.

On the samples simulating different radioactive waste from nuclear fuel fabrication the study on selection and experimental confirmation of decontamination recipe efficiency were performed. The extraction efficiency of tributyl phosphate (TBP) has been shown to be insufficient for effective extraction of uranium, thorium and plutonium. Yet trioctylphosphine oxide (TOPO) extraction efficiency ensures efficient extraction of uranium, thorium and plutonium. However, TOPO under normal conditions is a solid substance and therefore the preparation of TOPO solutions in supercritical (SC)-CO_2 or freons is difficult.

The extractant recycling operation, which in SF technology is usually carried out by means of pressure drop and evaporation of the main substance, CO_2 or a freon, remains quite complicated. The use of a solid extractant, in this case, complicates its collection, as solids are usually crystallized from the supercritical fluid on the surface of the whole volume of the container, including the vertical walls and the lid of the device. Based on information on actinides extraction by phosphorus-containing extractant solutions in SC-CO_2 and freones [1, 2] it was suggested that trialkylphosphine oxide (TPO) solutions should be used.

Experimental studies have confirmed the effectiveness of extraction of uranium and plutonium from individual solutions of the analytical laboratory and from a mixed solution using 10% TPO solution in chloroform and freon. Thus, the use of TPO solution in SC freon allows to solve the task.

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2. Supercritical CO_2 extraction of molybdenum-ligand complexes from sulfuric solutions, L. Hung, A. Hertz, D. Hartmann, etc., *The Journal of Supercritical Fluids*, Volume 111 (May 2016), Pp. 97-103.