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Jack D. Law
David A. Meikrantz
Dean R. Peterman
Catherine L. Riddle
Terry A. Todd

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Jack D. Law, David A. Meikrantz, Dean R. Peterman, Catherine L. Riddle, Terry A. Todd

Idaho National Laboratory: P.O.Box 1625, Idaho Falls, Idaho 83415 jack.law@inl.gov

INTRODUCTION

As part of the Advanced Fuel Cycle Initiative (AFCI), the reduction in volume and heat generation of spent nuclear fuel requiring geologic disposal is currently being addressed. The goal is to optimize utilization of the nation's first repository and potentially reduce or eliminate the need for additional repositories. This will be achieved through separating long-lived, highly toxic elements, reducing high-level waste volumes and the toxicity of spent nuclear fuel, and reducing the heat generation of spent nuclear fuel. The Idaho National Laboratory (INL) is working closely with a team of national laboratories and other organizations to support development of these separations processes.

Key to the reduction of short-term heat load in a geological repository is the separation of ^{137}Cs and ^{90}Sr. Removal of these highly radioactive fission products reduces the short-term (~100 yr) heat generation source of the spent nuclear fuel. Once separated, the Cs and Sr would be placed in storage until the activity has decayed to LLW levels, at which time it could potentially be disposed of as a non-transuranic (TRU) low-level waste (LLW).

DESCRIPTION

To support development of this separations process, technologies for the simultaneous separation of Cs and Sr from dissolved spent nuclear fuel are being developed. In previous work, cesium and strontium have been removed from acidic nuclear waste solutions and related alkaline wastes using separate solvent extraction processes designed specifically for these elements. In general, crown ethers have been selected for the removal of strontium, and calixarenes have been selected for separations of cesium. A novel process is being developed at the INL which combines these two types of extractants to create a solvent that will extract both cesium and strontium from acidic media, simultaneously. This technology, the Fission Product Extraction (FPEX) Process, utilizes 4,4',(5')-di-(t-butyldicyclohexano)-18-crown-6 (DtBuCH18C6) for the extraction of Sr, calix[4]arene-bis-(tert-octylbenzo-crown-6) (BOBCalixC6) for the extraction of Cs, 1-(2,2,3,3-tetrafluoropropoxy)-3-(4-sec-butylphenoxy)-2-propanol (Cs-7SB) as a modifier, TOA (trioctylamine) and an Isopar® L diluent[1]. Initial development of the FPEX process at the INL began in FY-03. Results from this initial testing were promising and further testing was performed in FY-05 and FY-06. Based on these test, a flowsheet was designed and tested using 24 stage of 3.3-cm centrifugal contactor.

RESULTS

Process goals for the separation of Cs and Sr from the spent LWR fuel require ≥99.9% removal. Additionally, the TRU content of the Cs/Sr product must be less than 100 nCi/g so that this waste stream will be classified as a non-TRU LLW once the ^{137}Cs and ^{90}Sr decay. This requires a decontamination factor of approximately 1E+05 to 1E+06, depending on the fuel composition and preceding separation processes, for the TRU from the Cs/Sr product.

Based upon the process goals and the results of laboratory testing, a flowsheet was designed for testing in 24 stages of 3.3-cm contactors. This flowsheet was successfully demonstrated with spent LWR fuel simulant. The flowsheet testing consisted of approximately 2 hours of startup, including the initiation of feed flows and filling of contactor stages, followed by 270 minutes of operation with feed simulant. The solvent was recycled during testing for a total of 2.6 solvent turnovers within the 24 stages of contactors. Hydraulic performance of the process was excellent and low distribution coefficients were obtained for Eu (Am surrogate). Removal efficiencies of >99.99 and >99.98% were obtained for Cs and Sr, respectively. These results exceeded the process goals of 99.9% separation.

[1] References herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the U.S. Government, any agency thereof, or any company affiliated with the Idaho National Laboratory.

CONCLUSIONS

An FPEX flowsheet has been successfully demonstrated with spent LWR fuel simulant. Hydraulic performance of the process was excellent. Removal efficiencies of >99.99 and >99.98% were obtained for Cs and Sr, respectively. These results exceeded the process goals of 99.9% separation. Additionally, distribution coefficients for Eu were very low indicating little of the TRU should be extracted with this flowsheet. Flowsheet testing with actual spent fuel is recommended in order to verify these results.