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Evaluation of dihexyloctanamide as extractant under plutonium rich feed conditions

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Tributyl phosphate (TBP) has been the work horse of Nuclear Fuel Reprocessing Technologists for more than five decades. However, based on five decades of experience on spent fuel reprocessing, certain problems have also been identified with the use of TBP as extractant. Prominent amongst them are: (a) high aqueous solubility, (b) poor radiation stability and interference of degradation products during stripping of Pu/U, (c) poor decontamination factor (DF) values of Pu/U with respect to fission products, (d) low limiting organic concentration (LOC) of Pu(IV), and (e) a large volume of secondary (phosphate) waste. In addition, as a consequence of the radiolytic degradation of TBP, the extraction and stripping behavior of U/Pu, and hydrodynamic properties such as viscosity, density, and phase disengagement time, are adversely affected. These problems are of particular concern to the separation scientists and technologists during the reprocessing of short-cooled thermal reactor fuels as well as of fast reactor fuels. In this context, completely incinerable N,N-dialkyl amides have been evaluated extensively as alternative extractants to TBP. Studies carried out at Radiochemistry Division, BARC, India, on the development of new extractants for the reprocessing of spent fuel suggested that a straight chain N,N-dihexyloctanamide (DHOA) was promising alternative to TBP for the reprocessing of irradiated uranium based fuels.
This paper deals with the evaluation of DHOA vis a vis TBP as an extractant for plutonium rich feed solutions encountered in fast reactor spent fuel reprocessing. Solvent extraction studies were carried out to evaluate the two extractants DHOA and TBP using pure 20 g/L Pu as well as simulated Pu rich feed solution containing 20 g/L Pu, 7 g/L U, fission products (FPs) and structural materials (SMs) at 4 M HNO₃. D_{Pu} values were higher for 1.1 M DHOA (26.4±1.2) for pure Pu as well as for simulated feed solution as compared to those for 1.1 M TBP (16.6±2.2) under identical conditions. Under simulated feed conditions, two successive stages were sufficient for quantitative extraction (>99.9 %) of Pu(IV) employing 1.1 M DHOA as extractant and maintaining organic-to-aqueous phase ratio (O/A) as 1. On the other hand, three stages were required in the case of 1.1 M TBP as extractant. Plutonium stripping experiments using 0.5 M HNO₃ as strippant (without any reductant) showed that only six stripping stages were sufficient for quantitative stripping of Pu from loaded DHOA phase. On the other hand, >10 stages were required for Pu stripping from loaded TBP phase and it became further difficult with the aging of the organic phase. By contrast, no problem related to Pu retention was observed for aged DHOA solution. These studies also suggested that DHOA is a promising extractant for coprocessing of U/Pu from spent fuels. In addition, DHOA was found distinctly better than TBP with respect to FPs and SMs decontamination.

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