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Radiation Stability of CyMe₄-BTBP and CyMe₄-BTPhen in their Solutions in Cyclohexanone-based Diluents

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CyMe₄–BTBP and CyMe₄–BTPhen are two prospective extractants for the European SANEX and/or GANEX processes for the recovery of minor actinides from a genuine spent nuclear fuel solution.[1] In such applications, radiation stability of the extractants and the influence of solvent irradiation on its extraction properties is one of the key factors determining feasibility of the extractant use in a process. [2]

In this study, radiation stability of CyMe₄–BTBP or CyMe₄–BTPhen was examined in systems with series of cyclohexanone-based diluents. In addition to following the degradation of the extractant, major degradation products were identified and the impact of irradiation of CyMe₄-BTPhen-based solvent on its extraction properties was followed.

Cyclohexanone and a series of different alkylated cyclohexanone derivatives were used as diluents. Two sources of ionizing radiation were used – in one part of the study CyMe₄–BTBP solutions were irradiated by accelerated electrons up to the absorbed dose 100 kGy; in the second part of the study both, the CyMe₄–BTBP or CyMe₄–BTPhen solutions were irradiated by gamma radiation up to the absorbed dose 400 kGy. Identifications and characterizations of the degradation products were performed by HPLC and MS analyses. Residual concentrations of both ligands were determined. The effect of the presence of HNO3 during the irradiation was studied for 2 selected solvents. Moreover, extraction properties of the irradiated CyMe₄–BTPhen solvents were compared with the extraction properties of non-irradiated solvents to estimate the influence of the degradation products in the organic phase.

In case of CyMe₄–BTBP in absence of the acid, a visible increase of stability was observed for systems containing 2-Me-cyclohexanone or 2,6-di-Me-cyclohexanone diluents. However, preliminary results indicate that this protective action is probably lost in presence of the acid. From this point, the system containing 3-Me-cyclohexanone seems better due to uniform stability in presence/absence of the acid. Composition of main degradation products corresponds to reactions with particular solvent molecules and oxygen or water. From these results, it could be concluded that the main issue of stability of the extraction system is apparently connected with reactivity of the carbonyl function (or other highly polarized bonds) present in the solvent, which leads either to nucleophilic reactions of BTBP with the solvent or radical reactions proceeding apparently also at the same sites of the solvent molecule.

The results of the study of extraction properties of the irradiated CyMe₄–BTPhen solvents revealed that the decrease in D(Am) is slower in 3-methylcyclohexanone at lover doses, but at the dose of 400 kGy 2,6-dimethylcyclohexanone seems to prevent the extractant degradation better.

References:

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Primary author: Dr GRÜNER, Bohumír (Institute of Inorganic Chemistry, AS CR, Řež)

Co-authors: EKBERG, Christian (Chalmers University of Technology, Gothenburg); LÖFSTRÖM ENGDAHL, Elin (Chalmers University of Technology, Gothenburg); JOHN, Jan (CTU in Prague, FNSPE, Department of Nuclear Chemistry, Prague); SOČUVKOVÁ, Jana (CTU in Prague, FNSPE, Department of Nuclear Chemistry, Prague); ŠVEHLA, Jaroslav (Institute of Inorganic Chemistry, AS CR, Řež); DISTLER, Petr (CTU in Prague, FNSPE, Department of Nuclear Chemistry, Prague); HÁJKOVÁ, Zuzana (Institute of Inorganic Chemistry, AS CR, Řež)

Presenter: SOČUVKOVÁ, Jana (CTU in Prague, FNSPE, Department of Nuclear Chemistry, Prague)

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