

Radiolytic stability of diglycolamides and N-donor extractants studied in nuclear waste management applications in Europe

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Abstract

The radiolytic stability is a very important prerequisite of organic molecules studied for nuclear waste management applications in Europe. Within several collaborative European projects in the past (e.g. ACSEPT, SACSESS) and the current GENIORS project, the stability of diglycolamides and N-donor extractants is studied. Diglycolamides are used in hydrophilic and lipophilic form for the complexation of trivalent lanthanide and actinide ions, while N-donor ligands are used for the selective complexation of trivalent actinides over lanthanides. Several process candidates for the separation of actinides from used nuclear fuel were developed and demonstrated on the laboratory scale, some of them even using genuine fuel solution. The radiolysis of both ligand families was investigated in solution, generally showing a good stability towards ionizing radiation. A remarkable difference in the two groups of ligands was found in their degradation compounds. While diglycolamides mostly form lower molecular weight degradation compounds through radical induced bond break, the N-donor ligands mainly form addition products with radicals formed through diluent radiolysis. Using solvent extraction methods with irradiated solutions, mass spectrometric analyses, pulse radiolysis experiments, and computational methods, we were able to gain fundamental insight into the reaction mechanisms.

Our main achievements in the field of ligand radiolysis relevant for nuclear waste management applications in Europe will be presented and discussed. A comparison of the radiolysis of the different ligands will be given and the role of radical formation from the diluent will be discussed in view of mechanistic understanding.