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Application of ab-initio molecular electronic structure calculations of radiolytic and hydrolytic stabilities of prospective extractants

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Many proposed sustainable nuclear fuel cycles include coextraction of minor actinides together with lanthanides from the high level liquid waste. The extraction of minor actinides is of particular importance due to their responsibility for the long-term radiotoxicity of the waste. For the trivalent actinides and lanthanides ions extraction, the diglycolamide (DGA) family of organic extractants is being used, among others. Since it is well known that the degradation of the extractants in the highly acidic and active environment leads to undesirable effects, it is necessary to demonstrate their hydrolytic and radiolytic stability.
For theoretical estimation of the general as well as the local chemical stability of the particular extractant's structure, the ab-initio calculations of some DGA extractants and their degradation products were performed. For the geometry optimization and electronic structure calculations, the Gaussian and DMol3 codes were used on the DFT level of theory (BLYP, B3LYP functionals / 6-31G**, DNP basis sets). The chemical stability was assessed according to various theoretical general and local stability indicators –electrostatic potential, HOMO-LUMO gap, spatial localization of HOMO, atomic charges, and bond orders. The results of local and general stability of the investigated extractants and their degradation products is in good agreement with the published experimental stability studies. The results for different computational codes and methods (exchange-correlation functionals, basis function types) were compared and found to be in good mutual agreement. Thus, such theoretical chemical stability predictions can provide a valuable support to experimental scientists in development of novel, more stable organic extractants and extraction methods.

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