INL/EXP-20-59178-Revision-0

Radiological Degradation of Tetraoctyl Diglycolamide in n-Alkyl Solvents: Influence of Solvent Ionization Potential

Samuel Branch Tyndall, Gregory P Horne, Brittany D Hodges, Christopher A Zarzana

August 2020

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Prepared for the
U.S. Department of Energy
Office of Nuclear Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

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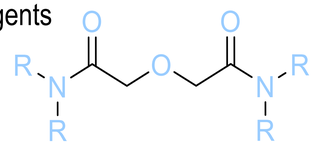
Abstract

Reprocessing of used nuclear fuel reduces the requirements for long-term storage repositories. Cost-efficient processes depend on knowledge of radiation effects, but gaps occur in understanding how radiation results in the degradation of fuel separation molecules. This project investigates the influence of different n-alkyl solvents on the radiolytic fragmentation behavior of N,N,N',N'-tetraoctyl-3-oxapentane-1,5-diamide (TODGA), a promising ligand for extraction of the lanthanides and minor actinides from dissolved nuclear fuel. Findings from this work suggest a linkage between the ionization potential of the solvent and the degradation products observed in the resulting mass spectrum.

Background and Motivation

- Separating and repurposing actinides and lanthanides from nuclear fuel minimizes storage repository use
- Diglycolamides (DGA's) are effective separating agents
- Radiolytic degradation of DGA's observed and characterized in n-dodecane
- Other n-alkyl solvents have not been used
- Different products seen in n-hexane
- **Hypothesis: Relationship between the ionization potential of the solvent, and the degradation products of DGA's seen after radiolytic degradation**



Methods

- Focus on one DGA called TODGA (R=octyl)
- Irradiate at different doses and in different solvents
- Separate column chromatography (Fig. 1)
- Ionize via electrospray ionization and send to the mass spectrometer (Fig. 2)

Figure 1

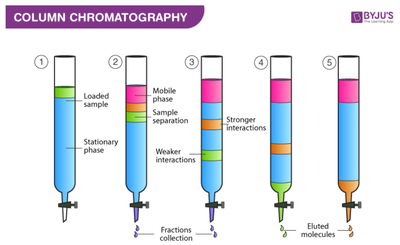
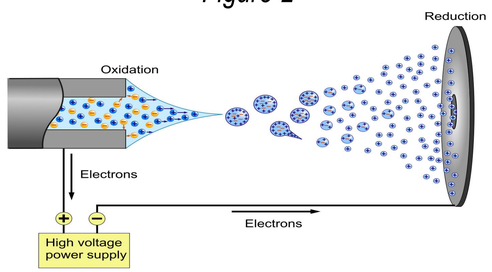


Figure 2



Materials and Instruments

- TODGA, n-alkyl solvents
- ^{60}Co Gamma Cell source (Fig. 3)
- Waters (Milford, MA) ACQUITY H-Class PLUS UPLC (Fig. 4)
- Bruker micrOTOF-II electrospray ionization quadrupole time-of-flight mass spectrometer (Fig. 5)

Figure 3

Figure 4

Figure 5

Results and Analysis

Figure 6: Previous work of TODGA Irradiated in Dodecane Mass Spectra

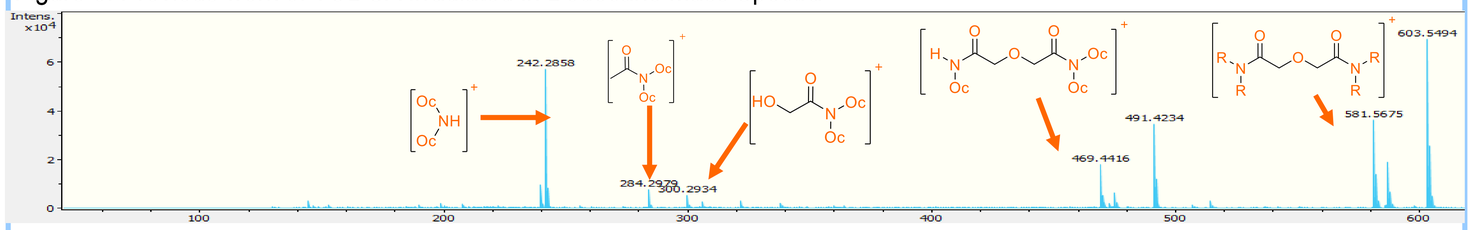
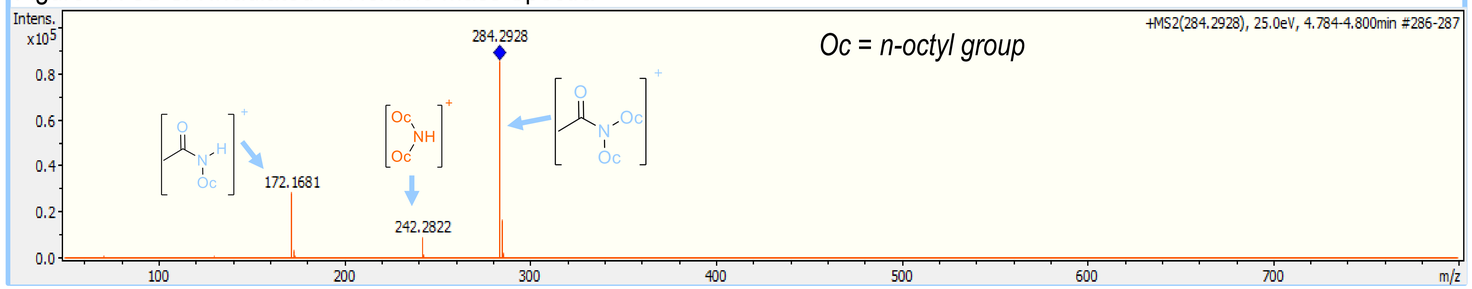


Figure 7:TODGA Irradiated in Hexane Mass Spectra



Oc = n-octyl group

Conclusions and Continued Work

- Fewer products produced in n-hexane than n-dodecane literature work
- Look to test TODGA in n-heptane, n-octane, n-decane, and n-dodecane
- Test different radiation doses and look for relationship with solvent ionization

Acknowledgments

This work was supported in part by the U.S. Department of Energy, Office of Science, Office of Workforce Development for Teachers and Scientists (WDTS) under the Science Undergraduate Laboratory Internships Program (SULI).

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Abstract

Reprocessing of used nuclear fuel reduces the requirements for long-term storage repositories. Cost-efficient processes depend on knowledge of radiation effects, but gaps occur in understanding how radiation results in the degradation of fuel separation molecules. This project investigates the influence of different n-alkyl solvents on the radiolytic fragmentation behavior of N,N,N',N'-tetraoctyl-3-oxapentane-1,5-diamide (TODGA), a promising ligand for extraction of the lanthanides and minor actinides from dissolved nuclear fuel. Placement of the vials in a 60Co Gamma Cell source occurred before analysis. Serial dilutions were made to achieve a 1µM concentration, and 3µL were injected into a Waters ACQUITY H-Class Plus UPLC coupled to a Bruker micrOTOF-II electrospray ionization quadrupole time-of-flight mass spectrometer for separation of impurities and analysis of the TODGA degradation products. Findings from this work suggest a linkage between the ionization potential of the solvent and the degradation products observed in the resulting mass spectrum.