



# Comparison of Gas Phase Fragmentation Behaviors of Nuclear Fuel Cycle Ligands in Lanthanide and Americium Metal Ligand Nitrate Clusters

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*Changing the World's Energy Future*

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## Impact

- Gas-phase investigation of the reactions of actinide and lanthanide complexes provide insights in understanding how metal coordination environments are influenced by electronic structure in the f-elements.

## Background

- Metal transportation across the boundary between the aqueous-organic phases is an essential step in nuclear fuel recycling process. Thus, understanding the behavior of the transport agents (ligands) guides designing of more efficient ligands with better separation efficiency towards higher recovery of materials from fission products. Gas-phase studies can isolate the metal-ligand interaction; however, limited gas-phase studies are currently available. This work studies the complexation of ligands to americium (Am) and lanthanides (Ln) and their fragmentations in the gas-phase. Comparisons are made between Am and Ln complexes.

## Result

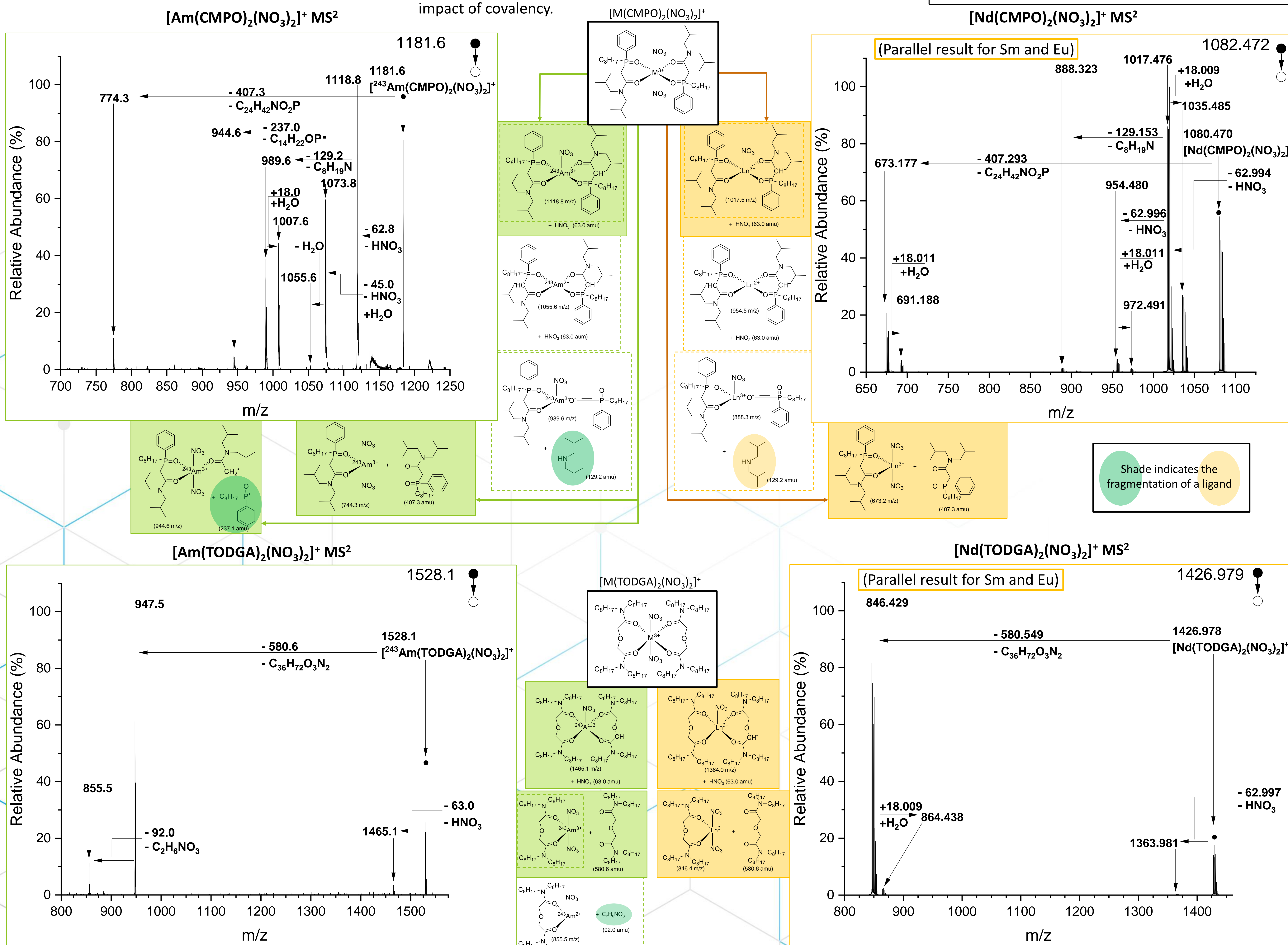
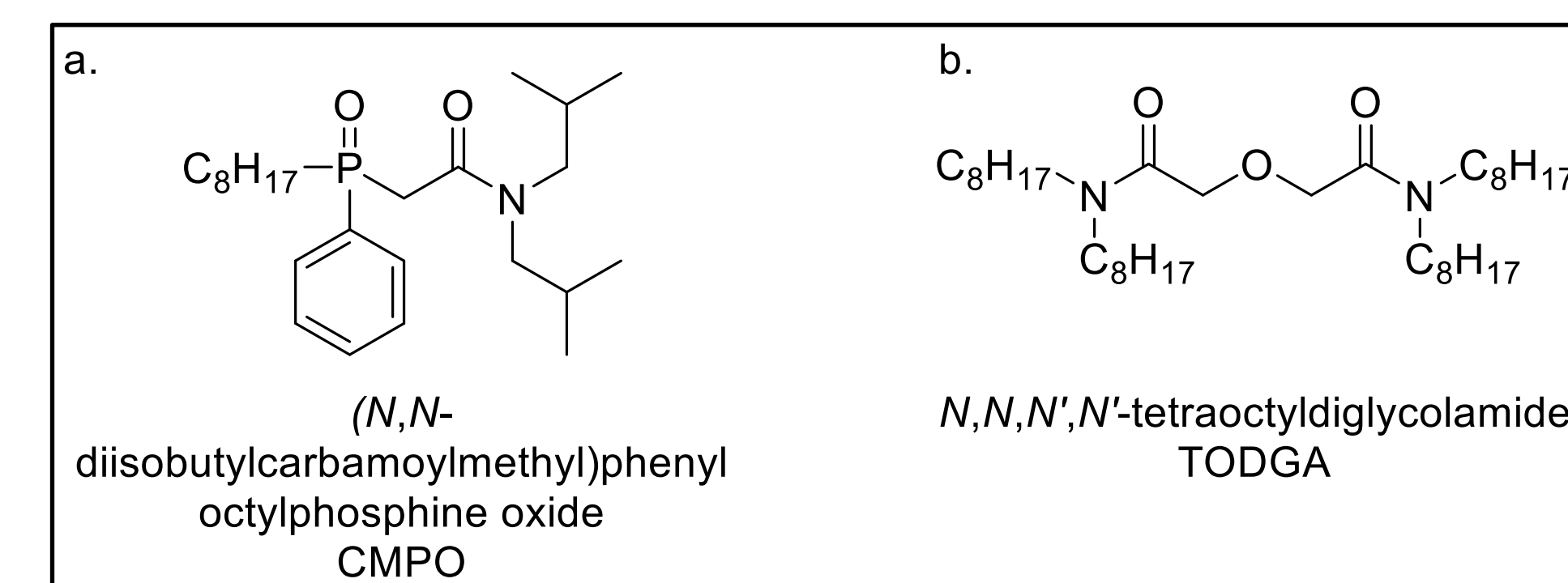
- Additional fragmentations were observed in metal-ligand complexes with CMPO than TODGA ligand complexes. Am complex exhibited additional fragmentations compared to the Ln complexes.

## Discussion

- Upon CID of Am-containing complexes, fragments of a ligand are lost in addition to intact ligands. It is hypothesized that an increase in the metal-ligand binding strength in the Am complexes compared to the Ln complexes weakens the intra-ligand bonds. This could be the result of increased actinide covalency. Calculations are ongoing to investigate the impact of covalency.

## Method

- Metal nitrates: Am (actinide), neodymium (Nd), samarium (Sm), and europium (Eu) (lanthanides; these lanthanides act as size and electron configuration analogues of Am)
- Ligands: (*N,N*-diisobutylcarbamoylmethyl)phenyloctylphosphine oxide (CMPO) and *N,N,N',N'*-tetraoctyldiglycolamide (TODGA)
- Metals complexed with two ligands and two nitrates ( $[M(NO_3)_2(Ligand)_2]^+$ ) are studied
- Bruker amazon Speed ETD quadrupole ion trap approved for use with transuranic elements. A Bruker micrOTOF-Q II quadrupole time-of-flight to collect high-resolution spectra of lanthanide-ligand complexes.



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