In-situ High Temperature Radiation-Induced Metal Cation Redox Chemistry

Jacy K. Conrad[1, S, C], Aliaksandra Lisouskaya[2], Logan Barr[3], Robert V. Fox[1], Stephen Mezyk[4], Craig Stuart[3] and David M. Bartels[2]

[1]*Center for Radiation Chemistry Research, Idaho National Laboratory, Idaho Falls, ID, U.S.A.*
[2]*Notre Dame Radiation Laboratory, University of Notre Dame, Notre Dame, IN, U.S.A.*
[3]*Reactor Chemistry and Corrosion, Canadian Nuclear Laboratories, Chalk River, Ontario, Canada*
[4]*Department of Chemistry and Biochemistry, California State University Long Beach, Long Beach, CA, U.S.A.*
jacy.conrad@inl.gov

Aqueous systems at elevated temperatures containing dissolved metal ions from the corrosion of stainless steels and other alloys are exposed to ionizing radiation fields throughout the nuclear fuel cycle. Ionizing radiation breaks the solvent down into highly reactive oxidizing and reducing species that drive changes in the oxidation states of metal ions, which determine their chemical properties, including solubility, complexation, and reactivity. Therefore, understanding the aqueous radiation chemical behavior of metal ions from key alloying materials, such as iron (Fe) and chromium (Cr), is critical to model and monitor the performance of several processes, including: the corrosion of nuclear reactor materials, the extent of extraction in used nuclear fuel reprocessing systems, and the partitioning of materials for vitrification in nuclear waste storage. Despite the importance and prevalence of these radiation-induced reactions, only two reactions between the primary radiolysis products of water and dissolved Fe or Cr ions were reported above 100 °C prior to this project. Here, we addressed this critical knowledge gap by studying the radiation chemistry of Fe and Cr ions in aqueous solution via a combination of kinetic measurements, using high temperature (up to 325 °C) time-resolved electron pulse radiolysis, and steady-state changes in metal ion redox distribution, using gamma irradiations over a wide range of temperatures (37–195 °C) facilitated by a custom-built vessel designed and benchmarked by this project. Time-resolved and steady-state data are essential for the development of multiscale computational models for the prediction of the behavior of these metal ion systems from ambient to hydrothermal conditions in radiation environments.