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# Recent Progress in Pulse Radiolysis Detection Methods and their Application to Ionic Liquid-Based Systems for Closing the Nuclear Fuel Cycle

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Recent developments in instrumentation have opened up new vistas for radiation research using pulse radiolysis techniques. Picosecond pulse radiolysis has become a routine technique in several countries, and new techniques are under development to further expose physical events occurring at very short times following radiolysis. Optical fiber-based "single-shot"methods providing picosecond-timescale transient spectroscopy detection for sub-milliliter liquid and solid samples now exist. New technologies such as mid-IR quantum cascade lasers enable detailed mechanistic studies of radiation-induced reactions using the high resolution and structural specificity of vibrational spectroscopy to identify intermediates. Broadband multispectral detection has increased the power and throughput of pulse radiolysis detection methods in the infrared and UV-Vis-NIR. In addition to the current progress described above, a perspective will be offered identifying important areas for future instrumentation development in support of cutting-edge radiation science.

Ionic liquids (ILs) attract the interest of researchers and industry due to their remarkable properties, and many applications in the fields of energy and technology, including as a potential medium for the treatment of spent nuclear fuel for the sustainable use of nuclear energy. For several years we have studied aspects of ionic liquid radiation chemistry from primary species reactivity through long-term product accumulation, to elucidate their degradation pathways. We aim to develop innovative, effective and durable IL-based separations systems, so we must also describe how radiolysis interferes with the separation process. During this project we found several classes of ions that are resistant to ionizing radiation and we will present some examples. The recent development of mid-infrared transient absorption detection has been particularly useful. For example, we observed the immediate formation of acetaldehyde and vinyl alcohol during pulse radiolysis of choline NTf2 (see figure). This work and use of the LEAF Facility of the BNL Accelerator Center for Energy Research, were supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, Geosciences and Biosciences, under contract DE-SC0012704 (BNL).

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