

## HOW CAN WE SHOW RADIOLYSIS AND RADIO-SHIELDING?

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Processing and reprocessing of nuclear materials often employs radiochemical separation technologies that are designed to allow separation and selective isolation of valuable radionuclides and precious metals, as well as removal of unwanted contaminants. One significant challenge to optimizing these high-level nuclear materials radiochemical separations methods is radiolysis of extraction materials. Breakdown of separations resins and materials is challenging to effective processing because degradation often leads to extraction processes that are unpredictable and costly. The extent and consequence of radiolysis is variable and dependent on a number of experimental factors; including the composition of the extractant, dose of radioactivity, hydrolysis, and the phases. Recently, Zalupski et al. (Idaho National Laboratory; INL) have demonstrated that extraction reagents used in the nuclear fuel cycle can be shielded from ionizing radiation by embedding them into the macrostructure of mesoporous carbon materials; specifically CMK-3. As a result, they saw no change in extraction behavior of an inorganic-organic CMK-3:HDEHP “hybrid” material for the extraction of <sup>241</sup>Am up to 1000 kGy. In collaboration with INL, we have attempted to build case based up quantitative and qualitative evidence of radiation shielding via direct comparison of hybrid systems to current extraction system; liquid-liquid systems and extraction chromatographic resins. A variety of spectroscopy techniques have been considered and performed. These include:

(1) Electron paramagnetic resonance spectroscopy to directly probe at the formation of radical species resulting from radiolysis and quantitate a difference between the formation or radiolysis products in hybrid systems versus conventional systems.

(2) Electron spray mass spectrometry to qualitatively observe radiolysis fragmentation products and, in conjunction with liquid chromatography, observe a decrease in the analyte concentration.

(3). Liquid and solid state NMR to qualitatively observe a peak shifts in NMR spectra (<sup>13</sup>C, <sup>31</sup>P, <sup>1</sup>H) to elucidate the formation of radiolysis products.

(4). Thermogravimetric analysis coupled with IR spectroscopy was used to qualitatively observe radiolysis through a change in the extractant weight loss profile and subsequent identification by IR.

While these investigations have improved our understanding of radiolysis, we have not been able to pin point a technique that allows for direct comparison among extraction materials.