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Immobilization of Fission Iodine by Reaction with a Fullerene Containing Carbon Compound and Insoluble Natural Matrix

S.M. Steinberg, G.S. Cerefice, and D.W. Emerson

BACKGROUND

The recovery of iodine released during the processing of used nuclear fuel poses a significant challenge to the transmutation of nuclear waste. Iodine-129, a long-lived fission product formed by both commercial nuclear power generation and nuclear weapons production, is released when reprocessing nuclear fuel. Since iodine can be concentrated in the human thyroid, any uncontrolled release of iodine may result in an increased rate of thyroid cancer in the exposed population. For this reason, recovery of iodine is important for implementing any nuclear transmutation strategy.

The first step in any transmutation strategy is the processing of the used nuclear fuel. This step involves separating the used fuel into its constituent elemental components, allowing the recovery of the uranium, transuranic actinides, long-lived fission products, and other components, depending on the strategy and processes involved.

When used fuel rods are dissolved in concentrated nitric acid in preparation for actinide recovery, iodine is released from the fuel. A significant fraction of the iodine is lost to the vapor phase during this process, where it may potentially become a fugitive emission and be released from the plant. To avoid this, specialized filtration systems are used to try to trap and sequester the released iodine (and other fission product gases).

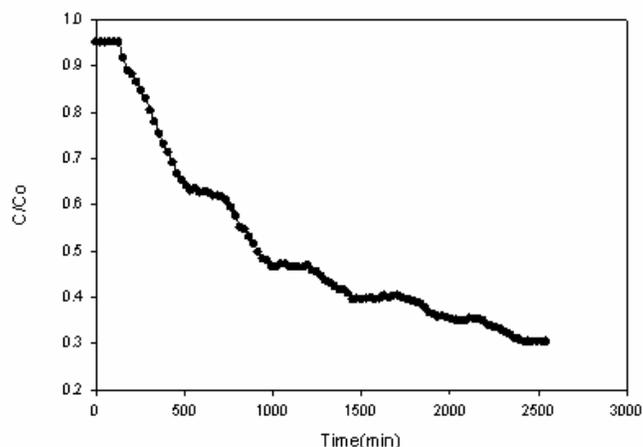
The primary goal of this research is to capture and immobilize the iodine released from these processes in a form that can easily be converted to a suitable target for neutron-induced transmutation. The investigators believe that iodine released during fuel reprocessing can be immobilized in a Fullerene Containing Carbon (FCC) compound or a Natural Organic Matter (NOM) matrix.

Natural organic matter (such as sphagnum moss, peat or brown coal) is an inexpensive and a renewable resource. Further processing of the trapped iodine using simple desorption or combustion processes should be able to produce iodine in a form suitable for transmutation. Furthermore, collaborators at the Khlopin Radium Institute (KRI) have proposed that the iodine-loaded FCC material, when combined with ceramics, is stable enough for use as a long-term storage form, and may be usable as a transmuter target matrix.

RESEARCH OBJECTIVES AND METHODS

The stability of the association of iodine with FCC and NOM products were studied. Product distributions for the various matrices under various reaction conditions were examined in order to maximize the binding of iodine. The recovery of the iodine from the sequestration matrices was also examined, along with the conversion of the iodine to matrices more suitable for geological storage and/or use as transmutation targets.

Iodide Sequestration at pH 4
Continuous Flow method



Sequestration of iodide ($10^{-4}M$) in the presence of sphagnum peat and MnO_2 . The solution was circulated through a column at 5 mL/min.

The following are the specific research objectives and goals:

- Develop bench-scale experimental set-up and procedures for simulating plutonium extraction process (PUREX) head-end vapor phase.
- Develop experimental procedures for evaluating iodine sequestering methods using bench-scale procedures.
- Develop FCC bearing material as potential iodine sequestration matrix.
- Determine binding of iodine to FCC and NOM.
- Examine alternate iodine sequestration matrices using techniques developed for FCC and NOM studies.
- Examine the effect of reaction conditions on binding.
- Elucidate the nature of the reaction products (volatile, hydrophobic, soluble, insoluble).
- Develop methodology and host matrix for converting sequestered iodine to solid matrix for evaluation as transmutation target and/or disposal matrix.
- Examine recovery of iodine from sequestration matrices.

The FCC compounds were developed and prepared by the KRI Research Industrial Enterprise (KIRSI). The KRI-KIRSI team researched the impacts of process parameters on sorption of iodine, and examined the material properties, such as how iodine attaches to the FCC compounds. The KRI-KIRSI team also examined the conversion of the iodine loaded FCC compound to a stabilized matrix (similar to ceramic) for potential use as a disposal form, acceptable transportation material, or potential target material.

RESEARCH ACCOMPLISHMENTS

Observations related to the oxidation of iodide to iodine (I₂) or hypoiodic acid (HIO) by MnO₂ were continued. During this study, a number of synthetic (nano-particle) manganese (III, IV) oxide preparations have been shown to be capable of oxidizing iodide to iodine (or IOH) at pH as high as 5. Various synthetic preparations of MnO₂ were examined. These various nano-particle manganese oxides have been characterized by transmission electron microscopy, scanning electron microscopy, and atomic force microscopy. In addition, surface areas, average manganese oxidation states and the kinetics of iodide oxidation by these materials have been measured. The rate of iodide oxidation by various manganese oxide preparations varies significantly. The oxidation rate appears to have some relationship to the average oxidation state of the manganese.

The incorporation of iodine into NOM was demonstrated in batch mixtures of MnO₂ and alkali lignin or sphagnum peat. The sequestration of iodine by NOM is illustrated to the left.

In another facet of this project, iodine distributions in salt impacted soils from the Virgin River, Nevada were examined. This study indicates that organic iodine was the most abundant form of iodine in the soil samples and that the content of organic iodine was correlated to total organic matter and to the lignin content (as measured by chemolysis) of the samples. These observations are consistent with the notion of the formation of organic iodine resulting from the cycling of iodine between iodide and iodate, with organic iodine resulting from the iodine or hypoiodic acid (I₂ and HIO) intermediate reacting with phenolic moieties in sedimentary and soil organic mater.

ACADEMIC YEAR HIGHLIGHTS

- ◆ S.M. Steinberg and G. Kimble, "Immobilization of Fission Iodine By Reaction with Insoluble Natural Organic Matter," *Journal of Radioanalytical and Nuclear Chemistry*, (in press).
- ◆ S.M. Steinberg, G.M. Kimble, G.T. Schmett, and D.W. Emerson, "Abiotic Reaction of Iodate with Spahgnum Peat and Other Natural Organic Matter," *Journal of Radioanalytical and Nuclear Chemistry*, (in press).
- ◆ S.M. Steinberg, B. Buck, J. Morton, and J. Dorman, "Speciation of iodine in the salt-impacted Black Butte soil series along the Virgin River, NV," accepted, 234th American Chemical Society National Meeting, Boston, MA, August 19-23, 2007.
- ◆ S.M. Steinberg and N.R. Birkner, "Formation of organic iodine in soil and sediment by reaction of manganese oxide with iodide," accepted, 234th American Chemical Society National Meeting, Boston, MA, August 19-23, 2007.
- ◆ M.A. Cheney, N.R. Birkner, L. Ma, T. Hartmann, P.K. Bhowmik, V.F. Hodge, and S.M. Steinberg, "Synthesis and characterization of inorganic double helices of cryptomelane nanomaterials," *Colloids and Surfaces A: Physicochemical Engineering Aspects* 289, pp 185–192, 2006.

FUTURE WORK

The oxidation of iodide with MnO₂ will continue to be investigated using remaining funds under a no cost extension. MnO₂ preparations will continue to be characterized. Presently, surface acidity of the various materials is being examined.

Combined Data							
	Depth	Org Iodide	Lignin	%C	% Clay	I	IO ₃
Avg depth	1.000						
Org Iodide	0.458	1.000					
Lignin/mg	0.519	0.623	1.000				
%C	0.407	0.394	0.608	1.000			
% Clay	0.186	0.388	0.130	0.535	1.000		
I	0.339	0.303	0.030	0.023	0.084	1.000	
IO ₃	0.260	0.223	-0.065	0.175	0.334	0.684	1.000

Correlation analysis for Black Butte soils

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