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Task 15

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.

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 are 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.

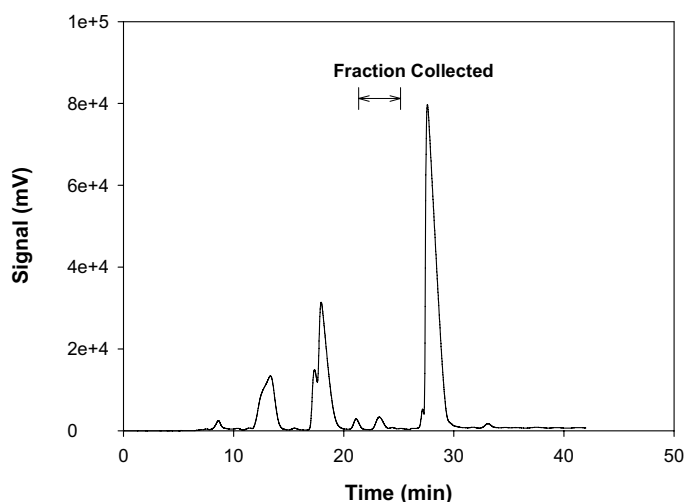
The following are the specific research objectives and goals:

- Develop bench-scale experimental set-up and procedures for simulating 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 are developed and prepared by the KRI Research Industrial Enterprise (KIRSI). The KRI-KIRSI team research the impacts of process parameters on sorption of iodine, and examine the material properties, such as how iodine attaches to the FCC compounds. The KRI-KIRSI team also examines 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 of the reduction of iodate by NOM were continued. Incorporation of iodine into various forms of NOM and various model compounds was demonstrated. Several methods for trapping iodine to further demonstrate that iodine or HOI was an intermediate in this process were utilized. Iodine trapping with N,N-dimethylaniline and selective oxidation of leuco crystal violet was also utilized. Kinetic measurements of this process were taken.



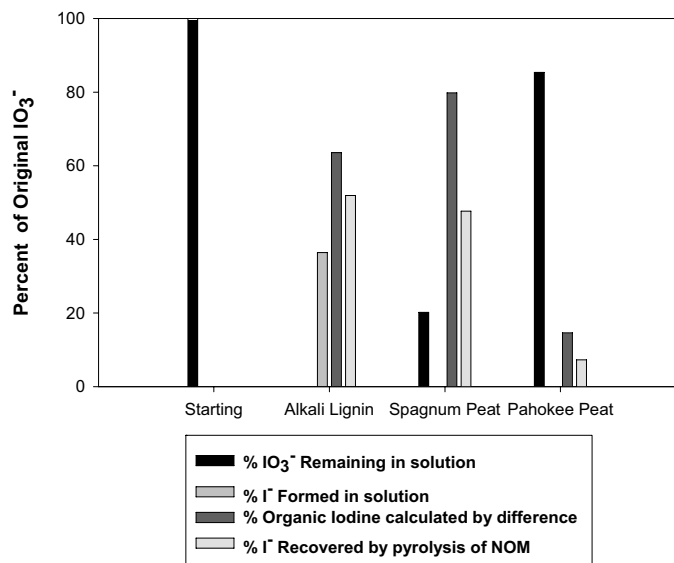
An example chromatogram from pyrolysis of 0.05 g of iodinated alkali lignin at 400 °C.

Utilizing preparative gas chromatography, it was demonstrated that methyl iodide that was released by pyrolysis (of .05 to 0.5 grams of OM) could be purified and recovered.

Several methods for the conversion of methyl iodide to sodium iodide were explored. The Wüster reaction was attempted and was not successful. Methyl iodide reacted rapidly with several nucleophiles including thiourea, mercapto ethanol. Methyl iodide could be easily reacted with GT-73 resin (mercaptan containing resin) in the sodium form. The result of this reaction was the release of sodium iodide that could be rinsed free of the resin.

The methyl pyridinium resin was prepared. Further experiments demonstrated that iodide could be recovered from aqueous solution and converted to methyl iodide by pyrolysis. Competitive effects from other aqueous anions were observed.

A number of potential reducing organic compounds as candidates for iodate reduction were examined. Lignin phenols and carbohydrates did not seem to be viable candidates. Hydroquinone however, rapidly reduces iodate to iodide. Furthermore, iodine (or HOI) was demonstrated to be an intermediate in this process and can be trapped as an organoiodine compound. For example a solution of hydroquinone and iodate can iodinate p-hydroxybenzoic acid or vanillic acid. It can be routinely observed that diphenolic



Distribution of iodine after iodate-NOM experiments at room temperature and pH 2.

ACADEMIC YEAR HIGHLIGHTS

- ◆ Steinberg S.M., Kimble G.M., Schmett G.T., Emerson D.W., *Abiotic reaction of iodine and iodate with sphagnum peat*. Preprints of Extended Abstracts presented at the ACS National Meeting, American Chemical Society, Division of Environmental Chemistry, 45(1), 476-481, 2005.
- ◆ Steinberg S.M., Kimble G.M., Schmett G.T., Emerson D.W., *Abiotic Reaction of Iodate and Iodine with Natural Organic Matter*. University of Nevada, Las Vegas: Las Vegas, NV. Presented at ACS 40th Western Regional Meeting, Anaheim, CA, January 22-25, 2006.
- ◆ Steinberg S.M., Kimble G.M., Schmett G.T., Emerson D.W., Turner M.F.*, Rudin M., *Immobilization of Fission Iodine by Reaction with Insoluble Natural Organic Matter*. Presented at MARC VII, Honolulu, HI, April 3-7, 2006.
- ◆ Steinberg S.M., Kimble G.M., Schmett G.T., Emerson D.W., *Abiotic Reaction of Iodate with Sphagnum Peat and Other Natural Organic Matter*. Presented at MARC VII, Honolulu, HI, April 3-7, 2006.

moieties are produced during pyrolysis of lignin and other forms of NOM. Others have evoked this functionality to explain the redox properties of humic and fulvic acids.

It was demonstrated that iodide can be trapped in NOM in the presence of manganese dioxide (g-MnO₂). The oxidation of iodide to iodine (or I₃⁻) can be observed at moderate pH (4-9). Also, it was demonstrated that at pH 7 iodide is incorporated into NOM in the presence of MnO₂. Observations indicate that the reaction occurs when microcrystalline MnO₂ (prepared by us) is utilized. Reagent grade MnO₂ does not appear to promote this reaction.

FUTURE WORK

It has been proposed that the reaction of NOM with iodide in the presence of various forms of MnO₂ be continued, and the effect temperature and pH on this reaction be investigated.

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