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

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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. Cerfice, 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.

NOM (such as spaghnum 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) in St. Petersburg, Russia, have proposed that the iodine-loaded FCC compound, 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.

RESEARCH ACCOMPLISHMENTS

Observations related to the oxidation of iodide to iodine (I₂) or hypoiodic acid (HIO) by MnO₂ were continued. The formation of triiodide presumable involves the adsorption of iodide onto the MnO₂ surface (perhaps displacing a surface hydroxyl group). The
iodide should be subsequently oxidized and released back into solution as \( \text{IOH} \) or \( \text{I}_2 \), which rapidly forms \( \text{I}_3^- \). The kinetic data has been modeled as a first order process. First order rate constants have been obtained for the formation of iodine in the presence of \( \text{MnO}_2 \). The increase in iodide oxidation rates with \( \text{MnO}_2 \) concentration is evident in the data. The reaction rate increases with iodide concentration although the dependence is not first order (an order of 1.4 appear to fit the data). The oxidation rate also increases with temperature and has a apparent activation energy of 16.2 kJ/mol.

The total yield of iodine form these materials was compared with the resulting Mn (II) concentration to estimate the oxygen to manganese ratio (\( \text{MnO}_x \)) for the starting material. This ratio \( (x) \) is a function of the average oxidation state of the material and is given by:

\[
x = 1 + \frac{I^-}{Mn^{+2}}
\]

The rate of iodide oxidation can be seen to vary significantly for the various preparations.

It is clear that some manganese oxide can oxidize iodide to iodine under mild pH and temperature conditions. By comparison, laboratory grade \( \text{MnO}_2 \) reacted sluggishly under these conditions. Because of the wide distribution of this mineral in nature, it is believed that the oxidation of iodide by manganese oxide may result in the formation of organic iodine bonds in sedimentary and soil organic matter.

**TASK 15 PROFILE**

Start Date: August 2002  
Completion Date: October 2007

**Theses Generated:**


**Journal Articles:**


S.M. Steinberg, B. Buck, J. Morton, and J. Dorman, “The speciation of iodine in the salt impacted Black Butte Soil Series along the Virgin River, Nevada, USA” Accepted for publication with minor revision.

**Conference Proceedings:**


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<th>Mineral</th>
<th>O/Mn Ratio</th>
<th>K(sec(^{-1}))</th>
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<tr>
<td>Cryptomelane Type II</td>
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<tr>
<td>Sodium Manganese Oxide Hydrate</td>
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<td>Black Birnessite + Cryptomelane</td>
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<tr>
<td>Cryptomelane Type II</td>
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<tr>
<td>Manganese Oxide Hydrate</td>
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<td>1.27 E-02</td>
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</table>

Rate constants for iodide oxidation (at pH 4 and solid concentration of 1 mg/mL) are tabulated along with the measured oxygen to manganese ratio of the manganese oxide. The mineral was identified by powdered X-ray analysis.

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