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IMMOBILIZATION OF FISSION IODINE BY REACTION WITH FULLERENE CONTAINING CARBON COMPOUNDS OR INSOLUBLE NATURAL ORGANIC MATTER

by

Gregory Tye Schmett

Bachelor of Science in Chemistry
University of Nevada, Las Vegas
2002

A thesis submitted in partial fulfillment of the requirements for the

Master of Science Degree in Chemistry
Department of Chemistry
College of Sciences

Graduate College
University of Nevada, Las Vegas
August 2005
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The Thesis prepared by

Gregory T. Schmett

Entitled

Immobilization of Fission Iodine by Reaction with Fullerene Containing
Carbon Compounds or Insoluble Natural Organic Matter

is approved in partial fulfillment of the requirements for the degree of

Master of Science

Examination Committee Chair

Dean of the Graduate College
ABSTRACT

Immobilization of Fission Iodine by Reaction with Fullerene Containing Carbon Compounds or Insoluble Natural Organic Matter

by

Gregory Tye Schmett

Dr. Spencer Steinberg, Examination Committee Chair
Professor of Chemistry
University of Nevada, Las Vegas

Since the start of the nuclear age, weapons testing and commercial nuclear power plants have been producing radioactive iodine-129 as a fission by-product. Iodine-131 has a half-life of 8 days and is generally not deemed a serious storage problem. Iodine-129 has a half-life of sixteen million years and decays by Beta emission.

The Department of Energy (DOE) has determined there are three major contributors to public radiation dose as a consequence of radionuclides releases. The three isotopes of major concern for the DOE are iodine-129, technetium-99 and neptunium-237 ($^{129}$I, $^{99}$Tc, $^{237}$Np). DOE has a research and development program called the “National Department of Energy Advanced Fuel Cycle Initiative” to recover and transmute iodine-129 to a stable isotope of xenon ($^{130}$Xe) in which UNLV program is the “Transmutation Reprocessing Program” (TRP).
This research explores the use of hitherto unexplored materials to trap iodine from nuclear fuel reprocessing, by either using Natural Organic Matter (NOM) or Fullerene Carbon Compounds (FCCs) for immobilization or conversion to a suitable target for nuclear transmutation.
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And last but not least I want to thank my father Roy Schmett for all of his support and help during my college “career”, thanks Dad.
CHAPTER 1

INTRODUCTION

Background

Iodine is a bluish-black, lustrous solid that mainly occurs in nature as stable iodine-127 (Latimer & Hildebrand, 1953). Small amounts of radioactive iodine-129 are formed naturally in the upper atmosphere by interaction of high-energy particles with xenon by a process known as spallation (R. Michel et al., 2002). Iodine is present in nature in diverse materials including soils, rocks, seaweed, sponges and mammals.

Since the start of the nuclear age, weapons testing and commercial nuclear power plants have been producing radioactive iodine-129 as a fission by-product. When an atom of uranium-235 fissions, it generally splits asymmetrically into two large fission fragments with mass numbers ranging from 90 to 140, and two or three neutrons. Iodine-129 and iodine-131 are two such by-products. The fission yield of iodine-129 is about 1% while the yield of iodine-131 is about 3% (Human Health Fact Sheet, 2001). For every one hundred nuclide decay events, one atom of iodine-129 is produced along with three atoms of iodine-131 from Uranium-238. The iodine-131 has a half-life of 8 days and is generally not deemed a serious storage problem. Iodine-129 has a half-life of sixteen million years and decays by Beta emission with a specific activity of (0.00018 Ci/g)(Human Health Fact Sheet, 2001).
The Department of Energy (DOE) has determined that there are three major contributors to public radiation dose as a consequence of radionuclides releases. The three isotopes of major concern for the DOE are iodine-129, technetium-99 and neptunium-237 ($^{129}$I, $^{99}$Tc, $^{237}$Np) (Bresee et al., 2001). DOE has a research and development program called the “National Department of Energy Advanced Fuel Cycle Initiative” to recover and transmute iodine-129 to a stable isotope of xenon ($^{130}$Xe) in which UNLV program is the “Transmutation Reprocessing Program” (TRP).

During the reprocessing of used nuclear fuel volatile iodine species are released as HI and I$_2$. This occurs particularly during the decladding and dissolution of the fuel rods in nitric acid. This iodine should be trapped to prevent the release of radioactivity to the environment. This research explores the use of hitherto unexplored materials to trap iodine from nuclear fuel reprocessing, by either using Natural Organic Matter (NOM) or Fullerene Carbon Compounds (FCC). These materials may be stable for geological storage or be further processed to prepare targets for transmutation. Radioactive iodine-129 has the same environmental and geochemical properties as stable iodine-127, with its predominant oxidation states of -1 or +5 (I$^-$ and IO$_3^-$).

Iodine may interact with FCCs by chemical or physical adsorption. It has been demonstrated that iodine exists in molecular form in fullerene crystals (Berdinsky et al., 1999). Chemisorptions of iodine onto fullerene structures may relieve ring strain by the formation of carbon-iodine bonds. A suitable transmutation target could then be achieved by desorption from FCCs or combusted to release a form of iodine ready for transmutation.
There is a large body of environmental and geological evidence that iodine can become associated with NOM (Warner et al., 2000). Several studies shown that I-129 is strongly retained in organic-rich surface soils (Sheppard & Thibault, 1992). This research explores the use of NOM, in particular sphagnum peat, to sequester iodine from nitric acid vapor (simulating the off-gas stream from the head-end dissolver). The sequestered iodine could then be recovered from the NOM for immobilization or converted to a target suitable for nuclear transmutation. It is potentially practical to convert (through transmutation reactions) long-lived radionuclides to short-lived or even stable isotopes (Bresee et al., 2001). As part of this work, the nature of the sphagnum-iodine association was explored as well as method that can be used to recover sequestered iodine from sphagnum.
CHAPTER 2

LITERATURE REVIEW

Background and Rationale

Iodine-129, and other stable and unstable isotopes of iodine are released during the reprocessing of nuclear fuel using either the PUREX or UREX processes (Choppin & Morgenstern, 2000). Releases of iodine from the PUREX or UREX processes are in the forms of several chemically reactive species (Taghipour & Evans, 2000). The UREX and PUREX processes are solvent extraction procedures in which the oxide fuel is decladed and dissolved in concentrated nitric acid. Uranium (UREX) or Plutonium/Uranium (PUREX) are extracted into an organic phase of tributylphosphate, which extracts actinide cations in the oxidation states IV and VI. Iodine is released as HI and I\(_2\) during the dissolution of the UO\(_2\) fuel pellets. A significant fraction of the iodine is lost to the vapor phase where it may potentially become a fugitive emission; or it can be trapped using specialized filtration systems that may incorporate activated carbon. The use of cyclodextrines for trapping iodine from nuclear waste steams have been explored (Szente et al., 1999). Hoskines & Karanfel, 2002, have discussed the use of silver-impregnated activated carbon for sequestration and removal of iodide from aqueous solutions. Cinnabar and chalcocite have been examined for iodide retention in fuel rod dissolution (Balsley et al., 1996). Iodine-129 is released during the nuclear fuel reprocessing as a fugitive vapor emission, ideally this emission should be trapped and
isolated from the environment. Transmutation can convert the long half-life of iodine-129 to a stable isotope of xenon-130 ($^{130}\text{Xe}$) through neutron capture. In order to complete this transformation a suitable stable target form of iodine-129 must be prepared.

\[ ^{129}_{53} \text{I} (1.5 \times 10^7 \text{ yr}) \gamma ^{130}_{53} \text{Xe} (10 \text{ yr}) \beta ^{130}_{54} \text{Xe} \]

Figure 1: Transmutation Reaction

We propose to bind fugitive iodine vapor onto FCCs or an inexpensive form of NOM, such as peat moss or sphagnum. Once the iodine has been trapped it could then possibly be concentrated and converted to NaI, which would be the transmutation target.

**Fullerene Carbon Compound**

Carbon clusters are molecules containing 60, 70, 76, 84, etc. carbon atoms arranged on the surface of a sphere or of a spheroid (Eletskii et al., 1995). $C_{60}$ was named “Buckminsterfullerene” after the American architect Buckminster Fuller who designed and built the geodesic dome; and also known as Bucky Balls. $C_{70}$ is known as “Falmerene” (Geckeler & Samal, 1999). The physical and chemical properties of FCCs have spawned the development of a new field of chemistry. Many reactions that have been studied with fullerenes include; alkoxylation, arylation, halogenation, hydroxylation, and osmylation (Aldersey, 1995). $C_{60}$ is comprised of 32 rings, in which 12 are pentagons and 20 are hexagons; the number of hexagons in a fullerene type
structure can be predicted by using Euler's theorem, \( m = (n-20) \ 2^{-1} \). Each fullerene family member \( (C_n) \) consists of 12 pentagons and \( m \) number of hexagons, \( m = (n-20) \ 2^{-1} \) (Geckeler & Samal, 1999). Scientists from the Khlopin Radium institute, located in St. Petersburg Russia have demonstrated the sorption of iodine onto FCC material; which then can be combined with ceramics for long-term immobilization and storage. FCCs are considered to contain a unique phase of elemental carbon, which can be found as balls, tubes (nano-tubes) and as ellipsoids shapes (Eletskii & Smirnov, 1995).

Having closed geometrical shapes these small particles are considered strained aromatic structures, which have considerable more reactivity than the basal planes of graphitic carbon (Geckeler & Samal, 1999). This interaction of FCC with iodine may lead to the formation of extractable iodo-FCC associations. An investigation into the nature of iodine conjunctions with intercalated FCC films has also been conducted (Berdinsky et
The FCC materials were produced by gas-phase deposition where the reactive substances (I$_2$ and C$_{60}$) were mixed at high temperatures and deposited on a chilled substrate. This study indicated that the iodine-iodine bond is not broken, but exists in the I$_2$ form while associated with the FCC materials. The solubility and chemical reactivity of this FCC iodine mixture was not reported in the study. The spectra of several halogen-FCC compositions have been investigated and the conclusion is that Cl$_2$ and Br$_2$ form covalent bonds with FCCs, while the association of I$_2$ bonds is primarily by van der Waals forces (Limonov et al., 1998). Iodo-organic formation with FCCs could relieve ring strain and may promote chemisorption of the iodine. The FCCs used in the experiments were prepared by the Khlopin Radiation Institute for UNLV. They consisted of a very fine powder with a particle diameter of less than 50 nm. The powder has been compressed into pellets that were able to absorb iodine from the vapor phase (Burakov, 2003).

Natural Organic Matter

Natural organic matter (NOM) is comprised of partially decomposed plant materials or plant remains (biomass) known as humus (McDonnell et al., 2001). The structure of humic acids and fulvic acids are not well established, however some general characteristics have been described. Humic acids have average molecular weights greater than 5000 amu, are dark in color, soluble under alkaline conditions, have high water retention capacity, have high capacity of cation exchange sites and are relatively stable structures (Properties of Humic Substances, 1997). Fulvic acids have molecular weights less than 5000 amu, are pale yellow in color, are soluble in both acid and
alkaline solutions, and have low water retention capacity, low cationic exchange and high chelating ability (Warner et al., 2000). Reactions with NOM and iodine have been investigated for decades. Iodine has been used in drinking water treatment for over a hundred years. However do to its high cost, limited availability, physical and chemical characteristics along with a medicinal taste it gives to the water, iodine has not generated sufficient interest to displace chlorine. The use of iodine has been limited to emergency water treatment when chlorinating water is not practical. The first known use of iodine in water treatment was during World War I, where iodine was used as an alternative to chlorinated water disinfection for the United States Military (White, 1992). For iodine, the disinfection by-products identified include, triiodomethane (CHI₃) along with other mixed halo-iodomethanes (CHCl₂I, CHBrClI). Iodinated compounds in drinking water can be detected by consumers at concentrations as low as (0.2 mg/L); due to their distinct medicinal odors and taste (Bruchet et al., 1989; Chang, 1958). Volatile halogenated by-products of this character are of vital concern to drinking water production facilities because they are considered potential human carcinogens and control of these disinfection by-products are critical and federally mandated.

Incorporation of iodine into naturally occurring high molecular weight organic matter such as humic acids and fulvic acids has been demonstrated by researchers for over 4 decades. In the sediment of the Mississippi river, studies of iodine-127 and iodine-129 distributions indicated that 70% to 85% of the iodine found in the river sediment is associated with colloidal organic matter (Oktay et al., 2001). It has been established that microbiological processes influences iodine uptake into the higher
molecular weight organic matter (Heumann et al., 2000). Recent studies have demonstrated the rapid interactions of aqueous \( I_2 \) with humic substances and phenols (White, 1992). Various studies demonstrate that phenols and acetyl substituted compounds react with iodide during oxidative treatment of waters with ozone, chlorine and chloramines (Bichsel & Von Gunten, 1999; Bichsel & Von Gunten, 2000).

Depending on the pH, oxidative reagents react rapidly with dissolved iodide thus converting it to hypoiodous acid (HIO) along with hypoiodite (IO\(^{-}\)). Organic matter reacts rapidly with hypoiodous acid resulting in the formation of iodine carbon bonds. Chloramines favor iodination reactions because chloramines selectively oxidize iodide to hypoiodite. Hypochlorite can also convert iodine to iodate (IO\(_3\)) (Vel Leitner et al., 1998).

Phenolic and acetyl model compound studies have indicated that NOM is reactive toward both active iodine and IOH. The reaction with phenols is well understood, and is promoted by both the OH\(^{-}\) and H\(^{+}\) at a low pH where the IOH\(_2\)\(^{+}\) is the iodinating species. Ionization of phenolic hydroxyl groups at a high pH enhances the reaction of IOH, a very weak acid, with phenolate. Electron releasing functional groups on the reacting phenol also promotes ring substitution. For reactions at acetyl groups the enol is produced by the initial conversion of the acetyl to the enol tautomeric form, which then reacts with iodine or IOH. Conversion of a ketone to an enol takes place by way of two solvent-mediated proton transfers. A proton is transferred to oxygen in the first step, and then removed from a CH\(_3\) in the second step. Enolization of acetyl, is produced when a base reacts with any acetyl group in the fulvic or humic acid.
Figure 3: Enol Reacting with Iodine, acid promoted

Figure 4: Enolate Reacting with Iodine, base promoted
iodination of an acetyl group results in a formation of triiodomethyl ketones, which then undergo hydrolysis to form a carboxylic acid and iodoform. This process is known as the haloform reaction. The rate-limiting step in the reaction is the hydrolysis.
moss, peat moss or brown coal is known as NOM and is a very inexpensive, renewable and abundant natural resource. NOM has several reactive functional groups including phenols, acetyl groups and other ketones that can react with hypoiodite and iodine.
NOM has a phenolic nature, and from degradation studies using CuO, oxidation and tetramethylammonium hydroxide pyrolysis produces several aromatic products, that are related to lignin (Hedges et al., 2000). Lignin is a phenolic biopolymer that comprises approximately 30% of all woody plants.
The dissolution of the spent nuclear fuel rods, which are dissolved in concentrated nitric acid produce (in the off-gas stream) I$_2$ and HI. This work showed off-gas iodine released from fission products reacts with NOM that has been mixed with an inorganic base such as CaOH$_2$. The base promotes disproportionation of I$_2$ to I$^-$ and IO$_3^-$. The base increases the pH of the NOM medium to insure that most of the iodine will be in the form of the hypoiodite anion, iodide or iodate. The base will raise the pH of the NOM trapping material and thus prevent the loss of any volatile elemental iodine; but the pH may be lowered because of the nitric acid. If Ca(OH)$_2$ is used as a buffering reagent the high calcium concentration in the medium should minimize the dissolution of humic and fulvic acids from the NOM thus keeping most of the bound iodine in an insoluble form (Oste et al., 2002). Previous studies have used a polysulfone resin to immobilize peat biomass, which was then used to sequester heavy metals (Trujillo et al., 1991). A similar method maybe applied to the NOM for iodine sequestration to prevent
the release of any soluble iodine species. Also, iodinated NOM could be further processed to concentrate iodine by alkaline oxidation, which then could leave iodine as an iodate salt in the residual ash. Perhaps the sphagnum could be directly subjected to advanced accelerator techniques for transmutation of the iodine-129.

Research Approach and Scientific Investigation Plan

Stable iodine-127 has the same environmental and geochemical properties as radioactive iodine-129, with its predominant oxidation states of -1 and -5 (I⁻ and IO₃⁻); therefore iodine-127 was used for all experimentation. A detailed characterization of the interactions of iodine with FCC and NOM for the purpose of developing new technology for trapping radioactive iodine released from nuclear fuel rods was conducted. The initial experiment consisted of building an I₂ vapor generator. Iodine vapor was passed through FCC substrates and in separate experiments NOM using this generator. The nature of the sequestered iodine was then studied. A nitrogen gas stream passes through the iodine crystals that were held at a constant temperature, then directed to an impinger filled with Na₂S₂O₅ reagent for trapping I₂. The concentration of iodine-127 in the gas stream was measured by allowing a known volume of gas in a measured amount of time to flow to a sodium meta-bisulfite (Na₂S₂O₅) filled impinger where iodide could then be measured using an iodide selective electrode. The iodide concentration was then used to determine the iodine concentration. Then iodine vapor stream was then diverted through FCC materials and in later experiments through NOM. With a known amount of iodine passing through the system in a measured amount of time, break-through can be ascertained by employing a second impinger that
was placed after the sorbent trap materials. This experimental design allows for the addition of other reagents such as water or nitric acid fumes to enter the system where their effect on iodine sorption can be assessed.

In addition to the \( \text{I}_2 \) generator a second apparatus for simulating nuclear fuel dissolution was constructed to simulate iodine sequestration under more realistic conditions. By placing a known amount of iodine into a three neck round bottom flask containing concentrated nitric acid, a nitrogen stream can sweep the iodine out where it will pass through the trap material to an awaiting impinger for measurement using an iodide selective electrode.
CHAPTER 3

METHODOLOGY

Experimental Design: Iodine Generator 1

An iodine generator was designed for break-through studies. Constant nitrogen (N₂) flow was achieved with a regulator and needle valve. Tygon™ tubing was connected to the regulator and connected to a Pyrex™ glass “Y” where the nitrogen flow was split. From the Pyrex™ “Y” each nitrogen stream was connected to a flow meter, both having the same range of (0 to 100 mL/min). The inclusion of two flow meters was to insure each nitrogen stream had the same flow rate. From the exit port on the flow meters the nitrogen streams were directed to two separate Wheaton™ bell jars (high volume sparging vessels). Each jar was water jacketed so circulating baths could be used to control the temperature in each jar. Each bell jar had a total volume of 1 liter. Each bell jar had a large mouth screw on cap with a Teflon™ seal. The seal was made out of silicone 3mm thick, with a 1mm thick Teflon™ liner. Both bell jars had entrance ports and exit ports to the reaction chamber with ¼ inch O.D. tubes adaptors. The adaptors had a ¼ inch diameter hole in the center so a glass tube could be inserted and sealed with an O-ring. One bell jar was used for elemental iodine chips while the other held nitric acid or nano-pure water depending on the experiment. From the exit port on the flow meters Tygon™ tubing was connected to the bell jar entrance port. A Pyrex™ glass tube with an outside diameter of ¼ inch was pushed into the Tygon™ tubing and
then secured with a hose clamp. The opposite end of the glass tube was connected to the bell jar entrance cap using an O-ring seal, which was then screwed into place. From the exit ports on each bell jar another glass tube as described above was connected to Teflon™ tubing. The tubing was changed from Tygon™ to Teflon™ to insure inert conditions existing in each bell jar. The Teflon™ tubing was connected to a Pyrex™ glass “Y” where the iodine and nitric acid streams combined and mixed. This single Teflon™ tube went back to a three-way stopcock to direct the stream to either an impinger or to a column “trap” followed by an impinger. The trap consisted of a packed Pyrex™ glass column filled with NOM or FCC. A wet test meter was connected to the exit port of the second impinger to keep track of the total volume of gas that was passed through the system during each experiment. An additional flow meter was connected after the wet test meter to monitor the flow rate. The wet test meter and the final flow meter were attached to the exit port of the impinger, depending on which apparatus was running. Flow meters only work accurately if they are open to the atmosphere on the exhaust port. The flow meters mentioned earlier, allows the experimenter to maintain the same flow on each side of the system. Flow rates are adjusted by the needle valves on each flow meter to the same scale reading. By monitoring the flow meter at the end of the system, then adjusting the internal flow meters, a desired flow rate could be maintained. By running the system for a known amount of time, to the impinger (with no in line trap) a total amount of iodine in moles (average I\textsubscript{2} concentration) could be calculated. In addition the average iodine concentration in the vapor phase was calculated. By running the system for the same amount of time to the trap material placed before the impinger, the break-through on iodine was obtained by measuring the
milli-volt reading of the iodide concentration in the impinger with an iodide selective electrode.

(Schematic 1): Iodine Generator 1

Each impinger held (15 mL) of sodium meta-bisulfite (Na$_2$S$_2$O$_5$) at (0.154 mol/L).

Sodium meta-bisulfite reduces elemental iodine to iodide, which can then be measured with the iodide selective electrode. The iodide selective electrode was calibrated by generating a calibration curve using potassium iodide solutions ranging from (10$^{-1}$ to 10$^{-4}$ M). The calibrating solution also contained an ionic strength adjuster (Na$^+$NO$_3^-$) with a concentration of (5 mol/L); (Ratio: 1ml ISA for every 50mL of Na$_2$S$_2$O$_5$). A calibration curve was used only when an R$^2$ value of 0.995 or greater was achieved. The bell jars (which were water jacketed) were connected to re-circulating thermal baths that
maintained the temperature at 40° C. This temperature was selected from a pressure curve generated for iodine (Figure 5) using the Antoine equation from Lange’s handbook of chemistry (lange’s, 1979).

\[
\log p = A - \left(\frac{B}{T + C}\right)
\]

\[
P = 1.02 \text{ Torr}; \ A = 9.810; \ B = 2901.0; \ C = 256.00; \ T = 40^°C
\]

Pressure curve generated from Lange’s handbook of chemistry.

(Figure 5)

The system was held at a constant temperature of 40° C using heat tape that was wrapped around the Teflon™ tubing and all connections; in addition, aluminum foil was used to insulate the tubing. The temperature was regulated by a variable autotransformer, and was adjusted to maintain a temperature of 40° C. Temperature was monitored with a Tyrek (type K) thermocouple that was inserted underneath the aluminum foil. All glass connections were sealed with sealed silicone grease.
Experimental Design: Iodine Generator 2

The only change made to the iodine generator described above was the bell jars were replaced with Midget™ bubblers (schematic 2), two in series for each bell jar replaced.

(Schematic 2): Iodine Generator 2

The bubblers were not jacketed so additional heating tape was wrapped around the bubblers and a second variable autotransformer was employed to maintain the bubblers at a constant temperature of 40° C. One set of bubblers held elemental iodine and the other held the concentrated nitric acid or nano-pure water, (depending on the experiment).
Experimental Design: Fuel Rod Simulator 1

To simulate nuclear fuel rod dissolution a fuel rod simulator was designed. The starting materials consisted of a nitrogen tank with a regulator and needle valve. Tygon™ tubing was connected to the regulator and connected to a flow meter with a range of (0 to 100 mL/min). On the exit port of the flow meter Tygon™ tubing was secured with a hose clamp. A ¼ inch outside diameter Pyrex™ tube was inserted into the opposite end of the Tygon™ tubing and secured with a hose clamp. A thermometer adapter with a 24/40 ground glass fitting that had an O-ring and screw top cap was employed. The glass tube was inserted through the cap and then an O-ring was placed over the glass tube. The adapter was then screwed together and this assembly was placed onto the left port of the three neck round bottom flask (RBF). The center port on the three neck RBF held a condenser that has a length of 40 cm. The last opening on the three neck RBF housed 50 mL pressure equalized addition funnel for adding nitric acid or nano-pure water to the RBF depending on the experiment. All fittings were (24/40) ground glass. From the top of the condenser another thermometer adapter was connected that held a ¼ inch outside diameter glass Pyrex™ tube sealed with an O-ring. The glass tube was then connected to Teflon™ tubing and was secured with a hose clamp. The Teflon™ tubing was connected to a glass column trap that was installed in the Teflon™ line. The glass column trap would either hold NOM or FCC materials during trapping experiments. From the end of the trap, the Teflon™ tubing was then connected to an impinger that held (15 mL) of sodium meta-bisulfite, that was connected to a second impinger to catch any iodine that was not trapped in the first impinger. This solution will reduce any elemental iodine that passes through the trap to
iodide. At the exit port of the impinger a wet test meter was installed (the same one used on the iodine generator) to measure the total volume of gas generated and passed through the system. A final flow meter was installed after the wet test meter. The exit port on the flow meter was vented to the atmosphere. By adjusting the needle valve of the internal flow meter the desired flow rate was maintained.

(Schematic 3): Fuel Rod Simulator 1

Experiments were run by placing a known amount of elemental iodine (in a solution of water) into the RBF holding 25 mL of nitric acid then letting the system run for a measured amount of time. The total amount of iodine sequestered by the trap could then be calculated by measuring the concentration of iodide that had broken through the trap and become reduced in the impinger; then measuring the iodine remaining in the bottom of the RBF. By mass balance the known amount of iodine was calculated in the trap material. The RBF was held at a constant temperature of 40° C by use of a heating
mantle plugged into a variable autotransformer. The condenser was kept at 40°C by a re-circulating bath set to 40°C. The Teflon™ tubing exiting the top of the condenser was held at a constant temperature by using heat tape and aluminum foil insulation.

Experimental Design: Fuel Rod Simulator 2

The principal difference between fuel rod simulator 1 and 2 was the condenser, it was removed from the middle neck of the RBF and replaced with a thermometer adapter. A glass connecting tube was inserted into the thermometer adapter and sealed with an O-ring. The Teflon™ tubing was then shortened by three-fourths of its original length. This shortened system was held at 40°C by use of heat tape and aluminum foil insulation.

(Schematic 4): Fuel Rod Simulator 2
Experimental Design: Fuel Rod Simulator 3

The differences between fuel rod simulator 2 and 3 was the RBF. It was replaced with an Ace bell™ jar and head. The bottom of the jar has a volume of 1 liter and was jacketed for water circulation. The head of the bell jar has three necks with the standard (24/40) tapered glass.

![Schematic 5: Fuel Rod Simulator 3](image)

A flat O-ring Teflon™ seal was inserted between the head and the bottom of the bell jar, which was then sealed with a clamp to hold the glassware together (Schematic 5). The bottom of the reaction flask was jacketed so water can circulate around the reaction chamber. The head of the bell jar was heated to 40° C with heat tape and insulating with aluminum foil. This modification eliminated silicone grease that was found to trap some of the I₂ in previous systems.
Glassware

Prior to each run, all glassware was washed with Alconox™ dishwashing detergent and rinsed twice with tap water, then five times with nano-pure water. The glassware was then dried in an oven at 110°C for an hour prior to use.

Reagents

The chemicals and reagents used in various parts of this study include those listed below. From Aldrich Chemical Co: Iodine flakes [7553-56-2], Bucky Balls or tubes [11203-BB], Calcium Carbonate [471-34-1] and Sodium Nitrate [7631-99-4]. From Fisher Scientific: Sodium Meta-Bisulfite [7631-90-5], Calcium Hydroxide [1305-62-0]. From EM Science: Potassium Iodide [7681-11-0], Sodium Carbonate [497-19-8], Sodium Bicarbonate [144-55-8].

Instruments Used

The instruments employed during the experiments included: Elemental Analyzer (CHN/O/S), model # 440, Exeter Analytical INC; X-Ray Photoelectron Spectroscopy, model # SSX-100 with x-ray source from Aluminum K-alpha, Surface Science Instruments; Netzsch STA Jupiter 449C Simultaneous Thermal Analyzer (STA-DSC/TGA); LaMotte DHA 3000 Digital mV meter, model # DHA 3000, LaMotte Co.; Smart Colorimeter, Model # LMC202 REV E, LaMotte Co.; Iodide Selective Electrode, model # 27504-18 LL2, Cole-Parmer Instrument Co.; Precision Wet Test Meter, Precision Scientific Co.; Flow Check meter, model # 7080, Alltech Associates Inc.
Experiments: Iodine Generator 1

Before the start of each experiment a calibration curve was generated using an iodide selective electrode then plotted. An $R^2$ value of .995 or greater had to be achieved before each experiment to ensure proper calibration of the electrode (figure 6). An initial reading for the iodide selective electrode was recorded for each impinger holding the 15 m/L of sodium meta-bisulfite. The nitrogen gas flow was set to 20 mL/min and an iodide measurement was recorded every 10 minutes to determine the iodine vapor concentration for the blank side of the system. The nitrogen gas flow was set to 20 mL/min and an iodide measurement was recorded every 10 minutes to determine the iodine vapor concentration for the blank side of the system. Total iodine sorbed by the impinger was calculated by subtracting the final millivolt reading from the initial millivolt reading (figure 7).

Iodide calibration curve generated using $10^{-1}$ to $10^{-4}$ sodium meta-bisulfite.

(Figure 6)
Iodine generator 1, free flow of iodine through the system.

(Figure 7)

Two experiments were ran using iodine generator 1 (Figure 8a & 9a) before it was replaced by iodine generator 2. One inherent problem with the initial design was the large lag time in starting and stopping of the nitrogen flow, this was because of the large volumes of the bell jars. In addition, the silicone seal tended to ripple when the lid was attached thus allowing iodine vapors to leak past the Teflon™ sheet and absorb into the silicone backing. The two experiments conducted with this system were 30% Ca(OH)₂ mixed with 70% sphagnum and activated carbon. Nitric acid is used in all experiments unless otherwise specified. The experiments consisted of filling a glass column with 30% Ca(OH)₂ and 70% sphagnum then clamping the trap in line with the system. After the iodine concentration was measured for the blank side, the iodine/nitric acid stream was directed through the NOM. Figure 8a shows the break-through curve for three runs of sphagnum with 30% calcium hydroxide in minutes. Figure 8b is the average of three
runs in millimole (mmol) vs. bed volume (BV) of the substrate. BV is calculated by measuring the pore volume (PV) of the trap material by pushing water into the column.

Break-through of iodine with 30% Ca(OH)\textsubscript{2} and sphagnum. Flow rate 20 mL/min.

(Figure 8a)

Average break-through of iodine with 30% Ca(OH)\textsubscript{2} and sphagnum.

(Figure 8b)
with a 1mL syringe then measuring the volume of water required to saturate the trap material. Break-through times were converted to BV by multiplying the flow rate by Activated carbon with different weights. Flow rate 20 mL/min.

(Figure 9a)

Activated carbon with different weights converted to bed volumes.

(Figure 9b)
time, then dividing by PV. (BV = (Flow * Time)/ PV). Statistics were calculated by combining the results from several experimental runs and are indicated by the error bars in the results. Activated carbon was studied because current trapping of iodine from dissolved nuclear facilities fuel rods uses activated carbon. Two separate runs were made each with different weights of activated carbon. Figure 9a has both runs plotted together and figure 9b is BV vs. mmole. The experiment with 0.100 grams did not break-through after 2 ½ hours and would not done so for several more hours; so the experiment was stopped and a weight of 0.020 grams was selected and replaced in the trap.

Experiments: Iodine Generator 2

Break-through of iodine with trap filled with 30% Ca(OH)$_2$ and sphagnum.

Flow rate 20 mL/min.

(Figure 10a)
Average-break through of iodine with trap filled with 30% Ca(OH)$_2$ and sphagnum.

(Figure 10b)

Break-through of iodine with trap filled with 30% Ca(OH)$_2$ and sphagnum.

Flow rate 20 mL/min. No acid in the system.

(Figure 11a)
Average-break through of iodine with trap filled with 30% Ca(OH)$_2$ and sphagnum.

(Figure 11b)

Break-through of iodine with trap filled with Khlopin FCC. Flow rate for 20 mL/min.

(Figure 12a)
Average-break through of iodine with trap filled with Khlopin FCC.

(Figure 12b)

Break-through of iodine with trap filled with Aldrich FCC. Flow rate 20 mL/min.

(Figure 13a)
Average break-through of iodine with trap filled with Aldrich FCC.

(Figure 13b)

Experiments: Fuel Rod Simulator 1

All experiments conducted with the various fuel rod simulators required the generation of an iodine calibration curve using an iodide selective electrode, then recording the initial (mV) reading of sodium meta-bisulfite filled impinger. A LaMotte™ Smart colorimeter iodine test kit was used to measure iodine in a stock solution (ppm). A known volume of the iodine stock solution was placed into the RBF, the nitrogen flow was started and the nitric acid was added using the pressure equalized addition funnel. Usually 3 mg of elemental iodine was used for the experiments. The system was then run for a known amount of time. The impinger iodide concentration was measured every ten minutes with the iodide selective electrode until a break-through of iodine was observed. Once a break-through occurred the nitrogen flow was
stopped and the solution remaining in the RBF was re-measured using the LaMotte™ Smart colorimeter. From the amount of iodine held by the impinger and the quantity remaining in the RBF, the sequestered iodine trapped in the NOM or FCC was calculated by mass balance in experiments that do not have copper in the system.

Figures 14 and 15 illustrate the effect copper has when added to the system. No trap was used in either experiment to observe the sparging rate of iodine with copper. Copper (0.5 grams) was added to simulate fuel rod dissolution because it generates NO₂ and NO by reacting with HNO₃ in the off gas stream. The copper impacted the iodide selective electrode and the LaMotte™ Smart colorimeter. In figure 15 the experiment was repeated except the sodium meta-bisulfite was replaced with sodium hydroxide.

System run with nitric acid, iodine and copper. Flow rate 20 mL/min.

(Figure 14)
System run with sodium hydroxide in impinger. Flow rate 20 mL/min.

(Figure 15)

Experiments: Fuel Rod Simulator 2

Break-through of iodine with 30% Ca(OH)$_2$ and sphagnum. Flow rate 20 mL/min.

(Figure 16a)

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Average break-through of iodine with 30% Ca(OH)$_2$ and sphagnum.

(Figure 16b)

Break-through of iodine with Khlopin FCC plotted with blank. Flow rate 20 mL/min.

(Figure 17a)
Average-break through of iodine with Khlopin FCC.

(Figure 17b)

Experiments: Fuel Rod Simulator 3

Break-through of iodine with 30% Ca(OH)$_2$ and sphagnum plotted with blank.

(Figure 18a)
Average break-through of iodine with 30% Ca(OH)$_2$ and sphagnum.

(Figure 18b)

Break-through of iodine with Khlopin FCC plotted with blank. Flow rate 20 mL/min.

(Figure 19a)
Average-break through of iodine with Khlopin FCC.

(Figure 19b)

Break-through of iodine, 30% Ca(OH)$_2$ and sphagnum, with copper, plotted with blank.

Flow rate 20 mL/min.

(Figure 20a)
Average break-through of iodine, 30% Ca(OH)$_2$ and sphagnum with copper.

(Figure 20b)

Break-through of iodine with Khlopin FCC and 0.01 g of copper, plotted with blank.

Flow rate 20 mL/min.

(Figure 21a)
Average-break through of iodine with Khlopin FCC and 0.01 g of copper.

(Figure 21b)
CHAPTER 4

RESULTS

Surface Area Analysis

Surface area analysis was conducted on the Khlopin FCC that was preloaded with 189 mg/g of I₂ to determine how much surface area was occupied by iodine. As a comparison the Khlopin FCC that had no sorbed iodine in the matrix was also analyzed, along with activated carbon. Two separate instruments were used for comparison, one was the Micromeritics Gemini 2370 and the other was the Micromeritics ASAP 2010. Each machine quantified the BET surface area (m²/g), micropore volume (cm³/g), micropore area (m²/g) and the external surface area (m²/g). The largest difference seen in table 1 was in the Khlopin FCC that was preloaded with iodine. The difference between the surface areas of the two samples is believed to arise from differences in the two instruments. The Gemini 2370 dries and degasses the sample with nitrogen at 200°C and is open to the atmosphere during the drying process where water and other sorbents are thermally degassed and carried away by the nitrogen stream. The Micromeritics ASAP 2010 is a closed system that uses vacuum to out-gas the sample at a temperature of 200°C Celsius. This operational difference could account for differences the BET surface area, micropore volume, micropore area and the external surface area seen between the two machines. The Khlopin FCC that had no iodine loaded in the matrix gave similar surface area results for both instruments. The activated carbon
surface areas are also very close for the two instruments. It is noteworthy that the activated carbon has almost 4 times the measured BET surface area, micropore volume, micropore area and external surface as the Khlopin FCC.

**Surface Area Analysis**

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<th>Micromeritics ASAP 2010</th>
<th>Micromeritics Gemini 2370</th>
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<tr>
<td>Dried under vacuum at 200° Celsius</td>
<td>Dried under nitrogen at 200° Celsius</td>
</tr>
<tr>
<td>Khlopin (FCC) preloaded with 189 mg/g of Iodine in matrix.</td>
<td>Khlopin (FCC) preloaded with 189 mg/g of Iodine in matrix.</td>
</tr>
<tr>
<td>Date: 8/8/2003</td>
<td>Date: 8/10/2003</td>
</tr>
<tr>
<td>BET surface Area</td>
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<tr>
<td>±0.2223</td>
<td>±2.826</td>
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<tr>
<td>Micropore Volume</td>
<td>0.004882 cm³/g</td>
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<tr>
<td>Micropore Area</td>
<td>29.4762 m²/g</td>
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<tr>
<td>External Surface Area</td>
<td>Correlation Coefficient: 0.9999113</td>
</tr>
<tr>
<td>Correlation Coefficient:</td>
<td>0.999948</td>
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<tr>
<td>Ktilopin (FCC)</td>
<td>0.00 mg of Iodine in matrix.</td>
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<tr>
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**Table 1**

**Elemental Analysis**

Elemental analysis was conducted using an Exeter Elemental Analyzer 440; that measured carbon, nitrogen and hydrogen.
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<th>Run #2</th>
<th>Run #3</th>
<th>Ave</th>
<th>Std</th>
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<td>Sphagnum</td>
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<td>Run #2</td>
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<td>Wt: ug</td>
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<td>1321</td>
<td>984</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>% Carbon</td>
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<td>45.25</td>
<td>46.23</td>
<td>45.58</td>
<td>0.56</td>
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<td></td>
<td>% Hydrogen</td>
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<td>0.05</td>
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<td>0.98</td>
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<td>1.30</td>
<td>1.30</td>
<td>0.33</td>
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<td>% Oxygen</td>
<td>79.86</td>
<td>59.39</td>
<td>75.51</td>
<td>71.59</td>
<td>10.78</td>
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<tr>
<td>Khlopin (FCC)</td>
<td>8/11/2003</td>
<td>2474</td>
<td>1808</td>
<td>2776</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Run #1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Run #2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Run #3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Wt: ug</td>
<td>2474</td>
<td>1808</td>
<td>2776</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>% Carbon</td>
<td>88.30</td>
<td>85.11</td>
<td>83.75</td>
<td>85.72</td>
<td>2.34</td>
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<tr>
<td></td>
<td>% Hydrogen</td>
<td>0.08</td>
<td>0.03</td>
<td>0.09</td>
<td>0.07</td>
<td>0.03</td>
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<tr>
<td></td>
<td>% Nitrogen</td>
<td>1.01</td>
<td>1.26</td>
<td>2.24</td>
<td>1.50</td>
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<td>% Oxygen</td>
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<td>13.60</td>
<td>13.92</td>
<td>12.71</td>
<td>1.83</td>
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<td>Activated Carbon</td>
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<td>1839</td>
<td>Ave</td>
<td>Std.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Run #1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Run #2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Wt: ug</td>
<td>1520</td>
<td>1839</td>
<td>Ave</td>
<td>Std.</td>
<td></td>
</tr>
<tr>
<td></td>
<td>% Carbon</td>
<td>90.98</td>
<td>89.62</td>
<td>90.30</td>
<td>0.96</td>
<td></td>
</tr>
<tr>
<td></td>
<td>% Hydrogen</td>
<td>1.27</td>
<td>1.14</td>
<td>1.21</td>
<td>0.09</td>
<td></td>
</tr>
<tr>
<td></td>
<td>% Nitrogen</td>
<td>6.56</td>
<td>6.24</td>
<td>6.45</td>
<td>0.29</td>
<td></td>
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<tr>
<td></td>
<td>% Oxygen</td>
<td>1.10</td>
<td>3.00</td>
<td>2.05</td>
<td>1.34</td>
<td></td>
</tr>
</tbody>
</table>

Table 2
Five different samples were analyzed and ran three times each to establish an average for; sphagnum, dried peat moss, potting soil, Khlopin FCC and activated carbon. The oxygen value calculated was by difference assuming that C, N, H and O account for the entire sample. Sphagnum peat had the highest values for carbon, nitrogen and hydrogen. This organic mixture was consistent in texture and dark brown in color; no large particles were observed in the matrix. Based upon the results from the elemental analysis sphagnum was selected for the study because the matrix was the most uniform; along with the observation it had the highest carbon content of the NOM's tested. The dried peat moss had low carbon, nitrogen and hydrogen results but had high oxygen content. This organic material was light brown in color and had what appeared to be perlite mixed into the matrix. The third sample analyzed was typical potting soil, which could be obtained at any nursery; this soil also had lower carbon, nitrogen and hydrogen content, but had a large oxygen content. The potting soil contained large amounts of, sticks, rocks and what appeared to be perlite in the matrix. The forth sample analyzed was the Khlopin FCC, it had large carbon content with small values for nitrogen, hydrogen and oxygen. The activated carbon had similar values for carbon, hydrogen and nitrogen as the Khlopin FCC material but with a slightly higher carbon content.

Thermal Gravimetric Analysis

The thermal gravimetric analysis (TGA) was conducted using a Netzsch STA Jupiter 449 C under N$_2$ purge gas at a heating rate of 10°C/min. TGA traces any weight loss the materials undergo upon heating including decomposition temperatures. Iodine has a melting point of 113.7° C, a boiling point of 185° C and has a vapor pressure @
25° C at 0.31 mmHg. By looking at figure 22 the weight loss starts at approximately 150° C and then starts to level out around 350° C. The loss is around the boiling point of iodine and thus indicates physical adsorption of iodine on the FCCs. The total mass loss from the sample is 10% thus indicating that the Khlopin FCC’s can hold about 100mg/g of iodine in it matrix. The Khlopin FCC experimented on was sample #5 which held 95 mg/g of iodine according to the Khlopin Radium Institute.

Iodine lost from Khlopin FCC using TGA.

![TGA: Khlopin FCC loaded with Iodine.](image)

(Figure 22)

**Differential Scanning Calorimetry**

Differential Scanning Calorimetry (DSC) was conducted using the Netzsch STA Jupiter 449 C with the sample under N₂ purge gas at a heating rate of 10°C/min. DSC is sensitive to any phase changes or chemical reactions the sample may undergo during
thermal cycling. This analysis indicates that iodine is being lost from the sample. As the sample is heated the iodine started to sublime away indicating that iodine was being evaporated from the sample matrix, as indicated in the figure 23. The Khlopin FCC experimented on was sample #5 which held 95 mg/g of iodine according to the Khlopin Radium Institute.

Iodine lost from Khlopin FCC using DSC.

(Figure 23)

X-Ray Photoelectron Spectroscopy

X-ray photoelectron spectroscopy (XPS) is a surface technique that analyzes the chemical environment of individual atoms. The principle lies in measuring the binding energy (BN) of core electrons ejected from the atom under photo-ionization. X-ray photoelectron spectroscopy was conducted on the Khlopin material preloaded with iodine at 189 mg/g. The sample was probed for the carbon, oxygen and iodine
absorption edge. The iodine 3d edge was expected to be more intense because the matrix was loaded with 189 mg/g of iodine. The problem that occurred with the XPS was that the ultra high vacuum pumped most of the iodine away before the spectrum was obtained. This caused problems for the instrument itself because the iodine attacked the copper O-ring seals in the XPS machine. Since the iodine was so easily pumped out of the matrix this confirms the TGA analysis and indicates purely physical adsorption of iodine in the large surface area matrix with no measurable ionic or covalent character. This is consistent with the TGA measurement conducted on the same matrix. The carbon edge 1s peak is quite pronounced in the spectrum. The binding energy was calibrated by assigning the top of the C 1s signal at 284.7 eV for the Khlopin FCC matrix.

XPS Spectrum of Khlopin FCC loaded with 189 mg/g of Iodine.

(Figure 24)
High Resolution C 1s Peak of Khlopin FCC Matrix Loaded 189 mg/g of Iodine.

(Figure 25)

High Resolution for I 3d Khlopin FCC Matrix Loaded with 189 mg/g of Iodine.

(Figure 26)
The iodine 3d edge is just barely above the background noise of the instrument, which indicates that the majority of the iodine has been pumped away before the spectrum was analyzed, the peak fell at 626.9 eV.

Iodine Generator 1

The results obtained from the first iodine generator are of comparison of sphagnum with 30% Ca(OH)$_2$ and activated carbon, figure 27. The results are given in bed volumes that are calculated from the pore volume of the trap material, time and flow rate of the nitrogen gas. Sorption is however reversible for activated carbon, but not for the sphagnum.

Comparison of sphagnum and Ca(OH)$_2$ to activated carbon in break-through times.

Flow rate 20 mL/min.
Iodine Generator 1.

<table>
<thead>
<tr>
<th>N. A. = Nitric Acid</th>
<th>Mole of Iodine</th>
<th>Mole of Iodine</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ave. Trapped</td>
<td>STDEV</td>
</tr>
<tr>
<td>30% Ca(OH)₂ &amp; 70% Sphagnum &amp; N. A.</td>
<td>5.68E-07</td>
<td>1.34E-07</td>
</tr>
<tr>
<td>Activated Carbon &amp; N. A.</td>
<td>1.54E-05</td>
<td>4.12E-06</td>
</tr>
</tbody>
</table>

(Figure 27)

Even though the activated carbon can hold more iodine based upon its surface area this is by purely physical adsorption.

Iodine Generator 2

Comparison of sphagnum with activated carbon and Ca(OH)₂, to Khlopin (FCC) and Aldrich (FCC) in break-through times. Flow rate 20 mL/min.
<table>
<thead>
<tr>
<th>Iodine Generator 2.</th>
<th>Mole of Iodine</th>
<th>Mole of Iodine</th>
</tr>
</thead>
<tbody>
<tr>
<td>N. A. = Nitric Acid</td>
<td>Ave. Trapped</td>
<td>STDEV</td>
</tr>
<tr>
<td>Khlopin (FCC) &amp; N. A.</td>
<td>6.38E-07</td>
<td>2.29E-07</td>
</tr>
<tr>
<td>16.6% Act. Carbon/20.37% Ca(OH)2/Sphagnum &amp; N. A.</td>
<td>6.08E-07</td>
<td>1.71E-07</td>
</tr>
<tr>
<td>Aldrich (FCC) &amp; N. A.</td>
<td>5.40E-06</td>
<td>2.25E-07</td>
</tr>
</tbody>
</table>

(Figure 28)

Of the three sorbents shown in figure 28 the Khlopin FCC adsorbed the largest percent of iodine from the system because of its high surface area; while the Aldrich FCC showed lower adsorption of iodine. The sphagnum had quite a large adsorption that resulted from the activated carbon that was mixed into the sample. In the sphagnum sample it is believed the activated carbon initially trapped the iodine, the base then aided in binding of the iodine released from carbon into the NOM. The initial iodine adsorption for sphagnum with 30% Ca(OH)₂ (figure 27) is not nearly as high as when the sample has been amended with activated carbon. Sorption into the NOM matrix is limited by the reaction kinetics and the concentration of reactive functional groups in the NOM.
Comparison of sphagnum and Ca(OH)$_2$ to activated carbon in break-through volumes.

Flow rate 20 mL/min.

(Figure 29)

Sphagnum was mixed with several different bases to see which combination gave the highest adsorption of iodine (Figure 29). The results presented in figure 29 indicate that the strength of the base influences the amount of iodine that is adsorbed by the NOM.
The conclusion is that the base reacts with the iodine (perhaps leading to disproportion) providing the sphagnum functional groups time to react with the sequestered iodine.

\[ 6H_2O + I_2 \rightarrow 2IO_3^- + 12H^+ + 10e^- \]
\[ (2e^- + I_2 \rightarrow 2I^-) 5 \]
\[ (3I_2 + 3H_2O \rightarrow 5I^- + IO_3^- + 6H^+) \]

Once Ca(OH)_2 was determined to be the best base of those tested it was then mixed in at 10%, 30% and 60%(wt/wt) with the sphagnum. The 30% mixture absorbed the highest amount of incoming iodine. It is believed the base plays two roles when mixed in with the sphagnum. The added base deprotonates the phenolic moieties in the sphagnum making the functional groups more reactive toward iodine. Also the hydroxide promotes the disproportion of iodine, which immobilizes fugitive iodine. However disproportion may slow down the reaction of iodine with the sphagnum by trapping iodine in a less reactive form.

Fuel Rod Simulator 1

The experiment summarized in figure 30 was conducted for two reasons. First to determine how KI reacted NO gas generated by the reaction of HNO_3 with copper once it was added to the system; and second, to see how fast hydrogen iodide would transfer through the system without a trap in the line. With copper in the system the breakthrough of NO_2 caused a large change in the ISE response. The colorimeter testing of the remaining iodine in the bottom of the reaction flask, also gave erratic readings.
Comparison of how 0.5 grams of copper added effects the experiment.

Flow rate 20 mL/min.

(Figure 30)

Comparison of how 0.5 grams of copper added effects the experiment with trap material

Flow rate 20 mL/min
Comparison between Khlopin (FCC) and Sphagnum, with the addition of 0.5 grams copper in the system. Flow rate 20 mL/min. The addition of copper to the system was intended to simulate the dissolution of atomic fuel rods with nitric acid (Figure 31). Initially 0.5 grams of copper was added to the reaction chamber. The generation NO\textsubscript{2} from the copper interfered with the iodide selective electrode and also exhausted the bisulfite filled impinger. The colorimeter testing of the remaining iodine in the bottom of the reaction flask also had erratic concentration readings.
Fuel Rod simulator 2

Comparison of nano-pure water to nitric acid with no trap in system.

Flow rate 20 mL/min.

(Figure 32)

In figure 32 the experiment was conducted as a comparison between nano-pure water and nitric acid to see how the presence of nitric acid impacted sparging behavior in the system with no trap. In both experiments the break-through time for iodine to reach the impinger was the same about 10 minutes. The difference seen between the two experiments can most likely be attributed to the difference in iodine’s solubility in the two solutions. Nitric acid likely salts out the iodine from solution thus causing more rapid sparging.
Comparison of sphagnum with 30% Ca(OH)$_2$, to Khlopin (FCC) for break-through times. Flow rate 20 mL/min.

(Figure 33)

Figure 33 illustrates a large difference is seen in break-through behavior between the Khlopin FCC and NOM. Khlopin FCC material is able to sorb more iodine than the 30% Ca(OH)$_2$ and sphagnum mixture. However sorption on NOM is irreversible, whereas sorption onto the Khlopin FCC was easily reversible. The trap material for each experiment, was washed with 10 mL of KI (as I$_3^-$) to see how much iodine could be removed as I$_3^-$ from the matrix. It was then measured colorimetrically using the DPD method. The 30% Ca(OH)$_2$ and sphagnum trap had less than 10% of the total iodine
removed off of the trap indicating that the iodine had undergone chemical reaction and was now incorporated into the sphagnum matrix. The FCC trap material was not washed right after being removed from the system and was capped with paraffin™ film for a few minutes. After setting for several moments the paraffin™ film turned pink. This indicated that the iodine was escaping from the FCC matrix. The trap was washed with KI (as I\textsubscript{3}) and measured using the iodide selective electrode and indicated that 90% of the iodine had been washed off the column. Our results demonstrate that Ca(OH)\textsubscript{2} and sphagnum mixture is able to chemically react with the iodine where as the FCC (due to its large surface area) material only traps iodine by physical adsorption.

Fuel Rod Simulator 3

Comparison of different concentrations of base with sphagnum.

Flow rate 20 mL/min.

![Graph showing the comparison of different concentrations of base with sphagnum.](image)
Fuel Rod Simulator 3 used all Teflon™ seals. Figure 34 shows results for sphagnum mixed with Ca(OH)\(_2\) in various proportions to confirm that the 30% Ca(OH)\(_2\) and 70% sphagnum is still the most effective tested mixture for iodine sequestering.

Comparison of Khlopin (FFC) with and without 0.01 gram of copper added to the system. Flow rate 20 mL/min.
An experiment was conducted with 0.01 grams of copper to compare the effects on Khlopin FFCs with and without copper in the system. Figure 35 indicates that NO$_2$ generation still enhances iodine break-through with Khlopin FCC. The copper added to the system posed several challenges, with one being the generation of nitrogen dioxide when mixed with nitric acid, which increased the flow rate. The nitrogen flow rate of 20 mL per minute had to be continuously adjusted on the flow meters to keep the proper flow rate. The smaller amount of copper prevented the impinger from being overwhelmed by NO$_2$ and allowed the tracking of the break-through, but the calculation for how much iodide was trapped in the impinger gave results higher for iodine than what was placed in the system initially. The colorimeter method of measuring the initial iodine in the RBF gave erratic results; no values obtained.
Comparison of Sphagnum with and without 0.01 gram of copper added to the system.

Flow rate 20 mL/min.

(Figure 36)

In figure 36, one experiment consisted of 30% Ca(OH)2 and 70% sphagnum with 0.01 grams of copper added, then for comparison the same experiment was ran with no copper in the system. The added copper increased the flow rate when mixed with nitric acid. The nitrogen flow rate of 20 mL per minute had to be continuously adjusted on the flow meters to maintain the proper flow rate. The smaller amount of copper prevented the impinger from being overloaded by NO₂ and allowed the tracking of the breakthrough, but the calculation for how much iodide was trapped in the impinger gave
erroneous results. Testing of the RBF gave erratic results using the colorimeter method; no values were obtained for the experiment with copper added.

Discussion

These experiments demonstrated that iodine can be sequestered by several different types of materials. The ability to provide detailed mass balance for the experiments may have been better if an ICP/MS was available. The methods used to determine the quantity of iodine each had advantages but also had limitations during the different experiments. The iodide selective electrode worked well when nitric acid or water was used in the system but failed when copper was added to the system. If more than 0.01 grams of copper was added to the system the impinger was overloaded and failed, but when 0.01 grams of copper was added the iodide selective electrode did not reverse directions and graphs could be plotted but gave erroneous results to calculate the amount of iodine that broke-through the trap.

The colorimetric method used to measure the iodine remaining in the R.B.F. is quite accurate when nitric acid and water is used in the system but gave erratic measurements when copper is introduced to the system, perhaps because CuI is forming.

\[
2\text{Cu} + \text{I}_2 \rightarrow 2\text{CuI}
\]

The use of 70% sphagnum mixed with 30% Ca(OH)\textsubscript{2} was the best combination of NOM and base concentration tested for sequestering iodine. This mixture allowed the base to deprotonate the phenolic structures making the sphagnum more reactive with
iodine, in addition the high pH promotes the disproportionation of iodine. This immobilizes fugitive iodine. Only a small percentage of the trapped iodine could be washed off with KI solution (<10%). The addition of copper to the system changed the dynamics of the reaction because NO$_2$ and HNO$_3$ are capable of oxidizing iodide. When copper is added to the system the iodine passed through the trap material with little apparent interaction. Several possibilities exists which include: the conversion of iodine to volatile species of INO$_2$ or INO, or the destruction of the reactive functional groups in the NOM. The mechanism of how NOx decreased the break-through time for iodine is not clear at this time. The problem with the shorter break-through time could be diminished by increasing the size of the trap.

The Khlopin FCC material had a high capacity for iodine trapping because of its large surface area. The trapping of iodine using either Khlopin or Aldrich FCCs is reversible since the iodine is lost from the matrix as soon as it is opened to the atmosphere. Iodine loss from the FCC material when opened to the atmosphere indicates that iodine is purely physically sorption; this conclusion is supported by the thermal gravimetric measurements. Iodine is released from the Khlopin FCC matrix at approximately 140°C, which is just above the melting point for iodine. FCC material that has trapped iodine and is then rinsed with KI solution can remove more than 90% of the iodine. Copper added to the experiments impacted the break-through times as NO$_2$ and nitric acid are both capable of oxidizing iodide. When copper is added to the system the iodine passed through the trap material with a lower hold up time. One possibility is the conversion of iodine to volatile species of INO$_2$ or INO which are not
as readily sorbed. The presence of NOx decreases the break-through time for iodine in both FCC and NOM type traps.

To use FCCs to trap iodine is not economically feasible because FCC material is currently expensive to make at this time. Activated carbon can trap more iodine than FCC material do to its larger surface area; and is relatively in expensive. The combination of activated carbon and NOM could be a viable alternative to other chemically modified activated carbon compounds, such as silver impregnated activated carbon.

Recommendations for future work would include more measurements with activated carbon and sphagnum combinations. Devise away to separate the NOx out of the gas stream before reacting Iodine with NOM. Then all experiments should be scaled up.
APPENDIX I

IODINE GENERATOR 2

Iodine Generator 2: 30% NaHCO₃ & 70% Sphagnum.

(Index 1a)

Iodine Generator 2: 30% NaHCO₃ & 70% Sphagnum.

(Index 1b)
Iodine Generator 2: 30% Na₂CO₃ & 70% Sphagnum.

(Index 3a)

Iodine Generator 2: 30% Na₂CO₃ & 70% Sphagnum.

(Index 3b)
Iodine Generator 2: 10% Ca(OH)$_2$ & 90% Sphagnum.

(Index 4a)

Iodine Generator 2: 10% Ca(OH)$_2$ & 90% Sphagnum.

(Index 4b)
Iodine Generator 2: 60% Ca(OH)$_2$ & 40% Sphagnum.

(Index 5a)

Iodine Generator 2: 60% Ca(OH)$_2$ & 40% Sphagnum.

(Index 5b)
Iodine Generator 2: 20.37% Ca(OH)$_2$ & 63.03% Sphagnum & 16.6% Activated Carbon.

(Index 6a)

Iodine Generator 2: 20.37% Ca(OH)$_2$ & 63.03% Sphagnum 16.6% Activated Carbon.

(Index 6b)
APPENDIX II

FUEL ROD SIMULATOR 1


Fuel Rod Simulator 1: 30% Ca(OH)₂ & 70% Sphagnum. With Copper.

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Fuel Rod Simulator 1: 30% Ca(OH)$_2$ & 70% Sphagnum. With Copper.

(Index 2b)
APPENDIX III

FUEL ROD SIMULATOR 2


(Index 1a)


(Index 1b)

(Index 2a)


(Index 2b)
APPENDIX IV

FUEL ROD SIMULATOR 3

Fuel Rod Simulator 3: 10% Ca(OH)$_2$ & 90% Sphagnum, Plotted with Blank.

(Index 1a)

Fuel Rod Simulator 3: 10% Ca(OH)$_2$ & 90% Sphagnum.

(Index 1b)
Fuel Rod Simulator 3: 60% Ca(OH)$_2$ & 40% Sphagnum.
Plotted with Blank.

(Index 2a)

Fuel Rod Simulator 3: 60% Ca(OH)$_2$ & 40% Sphagnum.

(Index 2b)
REFERENCES


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