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

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Project Title:

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

June 10, 2002

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*Graduate Student(s) will be included on KRI-KIRSI team

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Dr. Gary Cerefice (Harry Reid Center)

Graduate Student (To be Determined, UNLV Chemistry)
* a second graduate student will be added Year 2

AAA Project Collaborator: Dr. James Laidler
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Chemical Technology Division, Argonne National Laboratory

AAA Research Area: Separations

Abstract:

The recovery of iodine released during the processing of used nuclear fuel poses a significant challenge to the transmutation of radioactive iodine. This proposal will develop and examine the use of Fullerene Containing Carbon (FCC) compounds as potential sorbents for iodine release from the reprocessing of nuclear fuel. This work will also include the development of bench-scale testing capabilities at UNLV to allow the testing of the FCC material in a simulated process off-gas environment. This experimental capability will also be used to test other potential sorbition materials and processes, such as natural organic matter (NOM) and other promising alternatives. This work will also examine the development of a process to convert the sorbed iodine into a ceramic material with the potential for use as either a transmutation target or as a waste form in a partitioning and sequestration strategy.

Work Proposed For Academic Year 2002-2003:

Bench-scale experimental apparatuses and methodologies to simulate iodine entrainment in the vapor phase released from the head end of the PUREX process (the 4M nitric acid dissolution of spent nuclear fuel) will be developed, along with procedures to test the sequestration of iodine from the vapor mixture. Long-term performance/suitability of FCC and NOM will be tested for sequestration of iodine released by nuclear fuel reprocessing. FCC-bearing materials will be prepared and evaluated under laboratory conditions by KRI-KIRSI. Simulated process evaluations will be done on the FCC-bearing materials, NOM, and other matrices suggested by the collaborators at UNLV. Conversion of the sequestered iodine to a ceramic-like material will be examined by the KRI-KIRSI team. Recovery of the iodine from the sequestering matrices will also be examined (by both teams).

Requested Funds: **FY02:** \$178,468 **FY03:** \$178,248

Background and Rationale

Iodine-129 (^{129}I) is an important fission product formed by both commercial nuclear power generation and nuclear weapons testing. Iodine-129 is released by the reprocessing of nuclear fuel. Reprocessing of spent nuclear fuel is generally accomplished using the PUREX or UREX processes (Choppin and Morgenstern, 2000). This procedure involves decladding of fuel rods by strong alkali and subsequent dissolution of UO_2 fuel pellets with concentrated nitric acid. The concentrated acid solution is extracted with an actinide complexing solvent or solvent mixture and the spent acid is further processed to immobilize or recover other fissile products.

When spent fuel rods are dissolved in concentrated nitric acid in preparation for actinide recovery, iodine is released as HI and I_2 . 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. Szente et al. (1999) has discussed the use of cyclodextrines for trapping iodine from nuclear waste. Hoskines and Karanfil (2002) have discussed the use of silver-impregnated activated carbon for removal and sequestration of iodide from aqueous solutions. Balsley et al. (1996) have discussed iodide retention by cinnabar and chalcocite. Ideally the iodine released during nuclear fuel reprocessing should be isolated in a form that can be transmuted to a stable product using nuclear transmutation technology such as those proposed in the Advanced Accelerator Applications program. The objective of the research proposed here is to immobilize iodine in a form that can easily be converted to a suitable target for neutron-induced transmutation. We believe that iodine released during fuel reprocessing can be immobilized in a Fullerene Containing Carbon (FCC) compound or an inexpensive Natural Organic Matter (NOM) matrix prepared from natural peat. Further processing of the trapped iodine using simple desorption or combustion processes should be able to produce iodine in a form suitable for transmutation.

Iodine can be highly mobile in the environment and ^{129}I has a half-life of 1.6×10^7 years. Radioactive iodine can potentially be transformed into short-lived or stable isotopes by neutron irradiation. In order to accomplish this transformation a suitable stable form of iodine must be prepared. We propose to bind iodine into FCC or NOM in order to immobilize this element and prepare a suitable target for transmutation.

Radioactive iodine has the same environmental and geochemical properties as stable iodine, and in its predominant oxidation states of -1 and 5 (I^- and IO_3^-), it is a highly soluble and mobile element that is likely to become rapidly dispersed in the environment. Iodine is also a biologically reactive element. For example, numerous volatile, organic iodine species are formed by marine algae and by bacteria. These biological reactions are responsible for transferring iodine to the atmosphere and this process is an important leg of the geo-chemical iodine cycle. Volatile species, that are formed biologically, include iodomethane and iodoethane (Laturnus et al., 2000; Amachi et al., 2000; Huo et al., 2000).

Iodine is also important in human nutrition and is incorporated into several thyroid hormones. Iodine is thus required for proper function of the thyroid gland and iodine deficiency can lead to severe metabolic disorders. Radioactive iodine isotopes can be concentrated in the human thyroid (VanMiddlesworth et al., 2000), thus, uncontrolled release would constitute a direct threat to human populations. For this reason recovery of iodine from fission materials is important for implementation of Advanced Accelerator Applications.

Recently workers at the Khlopin Radium Institute have demonstrated the sorption of iodine by a Fullerene Containing Carbon compound. Furthermore, these workers have demonstrated that this carbon material can be combined with ceramics for successful immobilization and storage. We propose to investigate the sorption properties of this matrix under conditions that simulate the dissolution and reprocessing of nuclear fuel.

Fullerenes are widely regarded as a unique phase of elemental carbon. The fullerene units are generally comprised of balls, tubes and ellipsoid shapes (Eletsii and Smirnov, 1995). The closed geometrical shapes of these small particles render their "aromatic" structures considerably more reactive than the basal planes

of graphitic carbon (Geckeler and Samal, 1999). Researchers at Khlopin indicated that the fullerene content is critical in sorption since extraction of fullerenes leads to a drastic decrease in sorption capacity. The interaction iodine with the fullerenes may therefore lead to the formation of extractable iodo-fullerene associations. One important aspect of this research will be to investigate the nature of these associations. Berdinsky et al. (1999) has studied the nature of iodine intercalated fullerene films. These materials were manufactured by gas-phase deposition, whereby the reacting substance (I_2 and C_{60}) were combined at high temperature and then deposited onto a cool substrate. Their studies indicate that the iodine-iodine bond is not broken during this process and that iodine exists in the form of I_2 while associated with the fullerene. The solubility and chemical reactivity of this iodine-intercalated material were not reported in this study. These observations are consistent with the study of Limonov et al. (1998) that investigated the spectra of different halogen-fullerene compounds. Limonov et al. found that Br_2 and Cl_2 form covalent bonds with the fullerene while the association of I_2 is primarily through van der Waals forces.

Clearly under some conditions iodine (for example as ICl) can add to double bonds. Furthermore, the relief of ring strain in the fullerene structure by formation of iodo-organic compounds may promote chemisorption of iodine. In addition, the FCC material is likely to contain numerous other compounds with unknown reactivity toward I_2 . For these reasons we will explore the possibility of the formation of iodine compounds on the FCC material.

The materials prepared by the Khlopin Radiation Institute consist of a fine powder with many particles less than 50 nm in diameter (B. Burakov, personal communication). This powder can be compressed into pellets that should be able to “sorb” iodine from the vapor phase. With the small particle size the permeability of a packed sampling tube may be very low and render sampling difficult. Another possibility would be to coat the FCC powder onto the walls of an annular denuder. Annular denuders are constructed with several concentric tubes mounted within an outer tube. Coating of the FCC material may require addition of a binding agent. Gas-flow is forced through the annular spaces between the tubes of the coated denuder. Denuders are available commercially for air and stack-gas sampling. We will attempt to adapt a commercially available model to this application. In this way high flow rates can be maintained while effective surface area is exposed to the gas stream. A third possibility is to utilize the FCC material in a fluidized bed reactor. This possibility will be explored if packed sampling tubes and denuders prove unworkable.

Organic chemical reactions of iodine with NOM have been investigated for decades. One reason for interest in these reactions is that volatile halogen species are formed as a byproduct of water disinfection procedures that utilize chlorine. Chlorine disinfection has been used for water treatment for over one hundred years. Numerous halogenated byproducts have been identified in chlorinated water. Iodine containing disinfection byproducts include triiodomethane (CHI_3) as well as several mixed halo-iodo-methanes (i.e. $CHCl_2I$, $CHBrClI$). Consumers can detect volatile iodinated compounds, in drinking water, at low concentrations by their unpleasant medicinal taste. Generally, volatile halogenated disinfection byproducts are considered potential human carcinogens and control of this class of disinfection byproducts is of vital concern for drinking water production. In many water treatment plants, chloramination has been substituted for chlorination. Chloramination generally produces lower amounts of chlorinated disinfection byproducts than chlorination, but has a greater tendency to produce iodinated byproducts (Bichel and Von Gunten, 1999).

Many researchers have demonstrated the incorporation of iodine into naturally occurring high molecular weight organic matter (natural organic matter such as humin, humic acids and fulvic acids). Oktay et al. (2001) investigated the speciation of iodine (^{129}I and ^{127}I) in the Mississippi river. Their work indicated that 70-85% of the iodine was associated with colloidal organic matter. Heumann et al. (2000) and Rädlinger and Heumann (2000) demonstrated that iodine becomes associated with higher molecular weight organic matter as a result of microbiological influences. Warner et al. (2000) studied the rapid interaction kinetics of aqueous I_2 with phenols and humic substances. Bichel and Von Gunten (2000) investigated the reaction of iodide with various model phenols and α -methylcarbonyl compounds during oxidative treatment of iodide containing waters with ozone, chlorine and chloramine. Oxidative reagents react rapidly with dissolved iodide converting it into hypiodous (HIO) acid and hypiodite (IO^-), depending on the pH.

Hypoiodous acid reacts rapidly with organic matter resulting in formation of iodine carbon bonds. Iodination reactions are most favorable with chloramines because chloramines selectively oxidize iodide to hypoiodite. Other oxidants, including hypochlorite, can convert iodine to iodate (IO_3^-) (Vel Leitner et al., 1998).

Model compound studies indicate that phenolic and α -methyl carbonyl groups in natural organic matter (NOM) should be especially reactive toward active iodine (I_2 and IOH). The reaction mechanism is with phenols and is reasonably well understood. The reaction is promoted by both H^+ and OH^- ions. At low pH the IOH_2^+ is the iodinating species. At high pH the ionization of the phenolic hydroxy group enhances the reaction of IOH with the phenolate. This reaction is promoted by electron-releasing functional groups on the reacting phenol. Reaction of iodine with α -methylcarbonyl involves the initial conversion of the α -methylcarbonyl to the enol, which then reacts with IOH . Enolization of the α -methylcarbonyl is promoted by both H^+ and OH^- .

The binding of iodine by high molecular weight natural organic matter (NOM) is also promoted by microbial activity or by chloramination. Iodination of α -methyl carbonyl groups results in the formation of triiodomethyl ketones that undergo hydrolysis to form iodoform and a carboxylic acid (haloform reaction). The hydrolysis step is however, considerably slower than the initial binding of active iodine and iodoform yields are generally only a small percentage of theoretical. The iodination of phenols (electrophilic aromatic substitution) is a rapid process, and many stable iodinated products have been isolated.

Natural organic matter (such as sphagnum moss, peat or brown coal) is an inexpensive and a renewable resource. NOM contains a myriad of reactive functional groups such as phenols and α -methyl carbonyl groups and that react with iodine or hypoiodite. The phenolic nature of natural organic matter is well established. For example degradation studies using CuO oxidation and tetramethylammonium hydroxide pyrolysis produce numerous aromatic products that can be related to lignin as a precursor (Hedges et al., 2000). Lignin is an abundant biopolymer that comprises approximately 30% of higher plants. The dissolution of spent nuclear fuels in concentrated nitric acid will produce iodine as I_2 and HI in an off-gas. We propose to allow iodine released from fission products (in dissolution off-gas) to react with NOM, isolating it from the off-gas and from the fuel rod dissolution process. The off-gas can be condensed and mixed with an aqueous suspension of NOM and a buffering agent (e.g. CaCO_3) that will increase the pH of the reaction medium to insure that most of the iodine will be in the form of the hypoiodite anion and iodide. The high pH of this medium will prevent the loss of volatile elemental iodine. Addition of chloramines to the off-gas should render all of the iodine reactive with the NOM. If CaCO_3 is used as a buffering reagent, the high calcium concentration in the medium resulting from dissolution of CaCO_3 , should minimize the dissolution of humic and fulvic acids from the NOM (Oste et al., 2002) and keep most of the bound iodine in an insoluble form. Trujillo et al. (1991) have immobilized peat biomass in polysulfone resin. This immobilized biomass was used to sequester heavy metals. A similar stabilization of the peat may work for iodine sequestration and prevent the release of soluble iodine species.

After reaction, the iodinated NOM substrate could be further processed in order to concentrate the iodine. For example, alkaline oxidation of the sphagnum moss should leave behind iodine as an iodate salt in the residual ash. Organic substrates such as sphagnum could perhaps be directly subjected to processing using advanced accelerator techniques for transmutation of radioactive iodine. If small amounts of volatile iodine species are generated during reaction with NOM, they could be trapped on activated charcoal or a Tenax™ and charcoal combination. Volatile iodides can be thermally released from activated charcoal-Tenax™ trap and oxidized to CO_2 and iodate salts, although charcoal has been utilized for neutron activation analysis of iodine in natural samples (Quintana and Thyssen, 2000).

Research Objectives and Goals

- Develop bench-scale experimental set-up and procedures for simulating PUREX head-end vapor phase.
- Develop experimental procedures for evaluating I sequestering methods using bench-scale procedures.

- Develop FCC-bearing material as potential I sequestration matrix.
- Determine binding of iodine to FCC and NOM.
- Examine alternate I 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). See Figure 1 for example.
- Develop methodology and host matrix for converting sequestered I to solid matrix for evaluation as transmutation target and/or disposal matrix.
- Examine recovery of iodine from I sequestration matrices.

Technical Impact

The proposed work is intended to enable researchers to optimize (develop a simple, economical and safe method) the sequestration and subsequent immobilization of iodine from the off-gas stream of the PUREX (or equivalent solvent extraction) process. This work will examine a number of approaches, with a preliminary focus on FCC or NOM matrices. The stability of the association of iodine with FCC and NOM products will be studied. The product distributions for the various matrices under various reaction conditions will be examined in order to maximize the binding of iodine to the insoluble fraction. The recovery of the iodine from the sequestration matrices will also be examined, along with the conversion of the iodine to matrices more suitable for geological storage and/or use as transmutation targets.

Research Approach/Scientific Investigation Plan

We are proposing a detailed characterization of the reactions of active iodine with FCC and NOM for the purpose of developing technology for trapping radioactive iodine released from processed nuclear fuel. The FCC compounds will be developed and produced by the KRI-KIRSI team, and will be studied at both locations. The KRI-KIRSI team will study impacts of process parameters on sorption of I, and will examine the material properties (such as how I attaches to the FCC compounds, etc.). The UNLV team will examine the FCC material, along with NOM and other potential sequestering agents, under simulated process conditions. The KRI-KIRSI team will also examine the conversion of the iodine-loaded FCC compound to a stabilized matrix (similar to a ceramic), for potential use as a disposal form, transportation phase, or potential target material. Both teams will examine recovery of the iodine from the sequestering matrices.

Initial experiments for FCC characterization will be performed by constructing a I₂ vapor generator. I₂ will be passed through the FCC substrate and then the nature of the sequestration will be studied. The proposed experimental apparatus is shown in Figure 1. Iodine crystals will be held and a constant temperature so as to generate a known concentration of iodine vapor. A stream of nitrogen gas will be passed through the I₂ chamber. This gas stream will be diluted with I₂ free nitrogen. The iodine vapor stream of known concentration will be directed through FCC test materials. The concentration of iodine in the gas stream from the generator will be measured by allowing a known volume of gas to flow through a NaOH-filled impinger where it can be measured using the DPD (N,N-diethyl-p-phenylenediamine) method. This experimental setup will allow the addition of other reagents such as water or nitric acid fumes, so that the influence of these compounds on iodine sorption can be assessed. A second NaOH-filled impinger placed after the sorbent trap can be used to measure breakthrough of iodine.

In addition we will construct a device for simulating nuclear fuel dissolution to simulate iodine sequestration under more realistic conditions. The general format of the experimental apparatus we envision is shown in Figure 2. Material with a known concentration of iodine will be placed into a round bottom flask that will be used as a reaction chamber. Nitric acid (i.e. 4M) will be added with a pressure-equalizing addition funnel. The mixture will be heated to a known temperature either with a heating mantle or a circulating temperature bath. The condenser will be used to control the loss of water from the system and to prevent flooding of the sampler. A carrier gas will be used if necessary, to strip iodine from the reaction chamber. Both of these devices will allow us to assess the breakthrough of iodine with various sorbents, and the effect of water and various oxides of nitrogen on sorption.

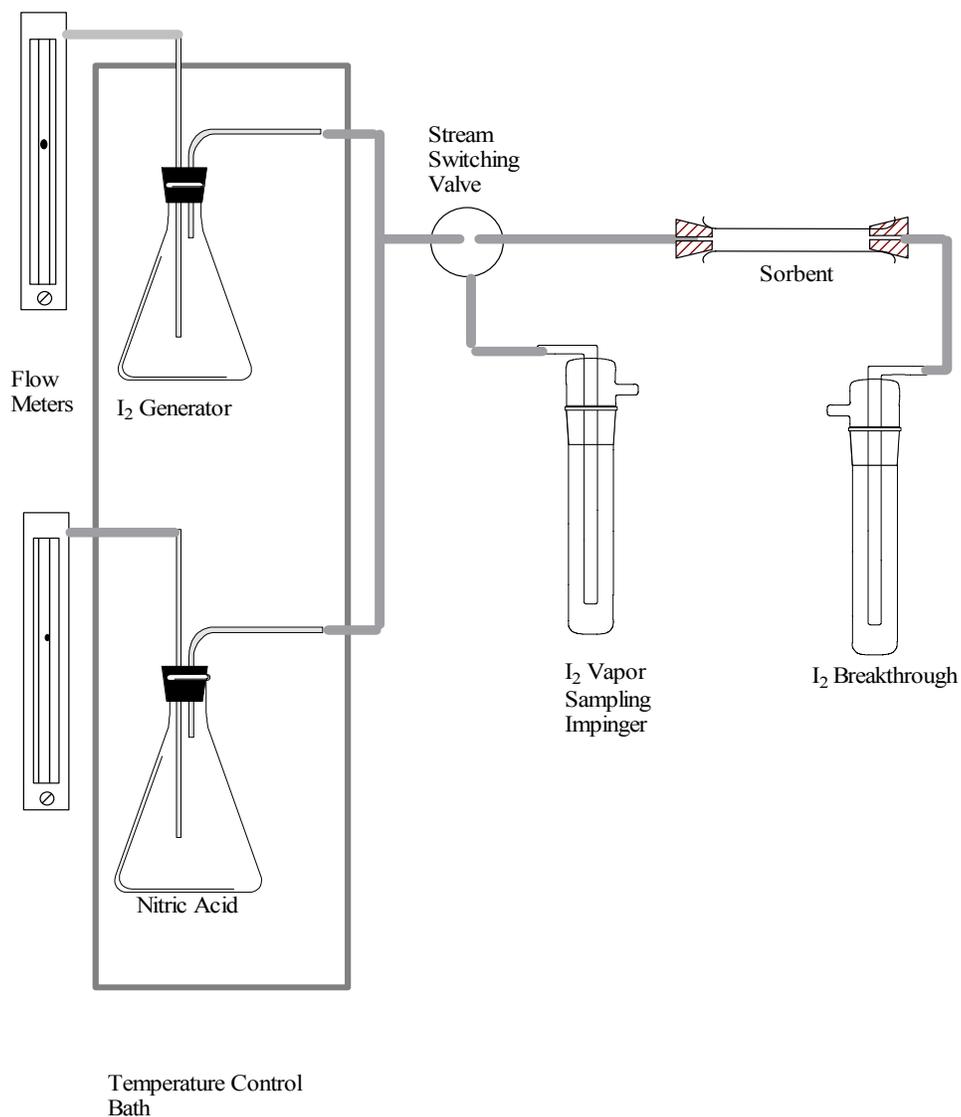


Figure 1: Iodine Vapor Generator

We propose to examine the reversibility of the sorption of iodine on FCC by performing a variety of experiments. We propose to assess the stability of the FCC to leaching with simulated groundwater and with solutions containing various reagents that can change the oxidation state of the iodine. For example, reaction of sorbed I_2 with reducing agents such as hydroxylamine or sulfite may form iodide which would be considerably more mobile in the environment and very susceptible to leaching from an FCC-ceramic composite. On the other hand, if the objective of the program is to concentrate the iodine for transmutation a simple leaching method for recovery from the FCC will be prerequisite.

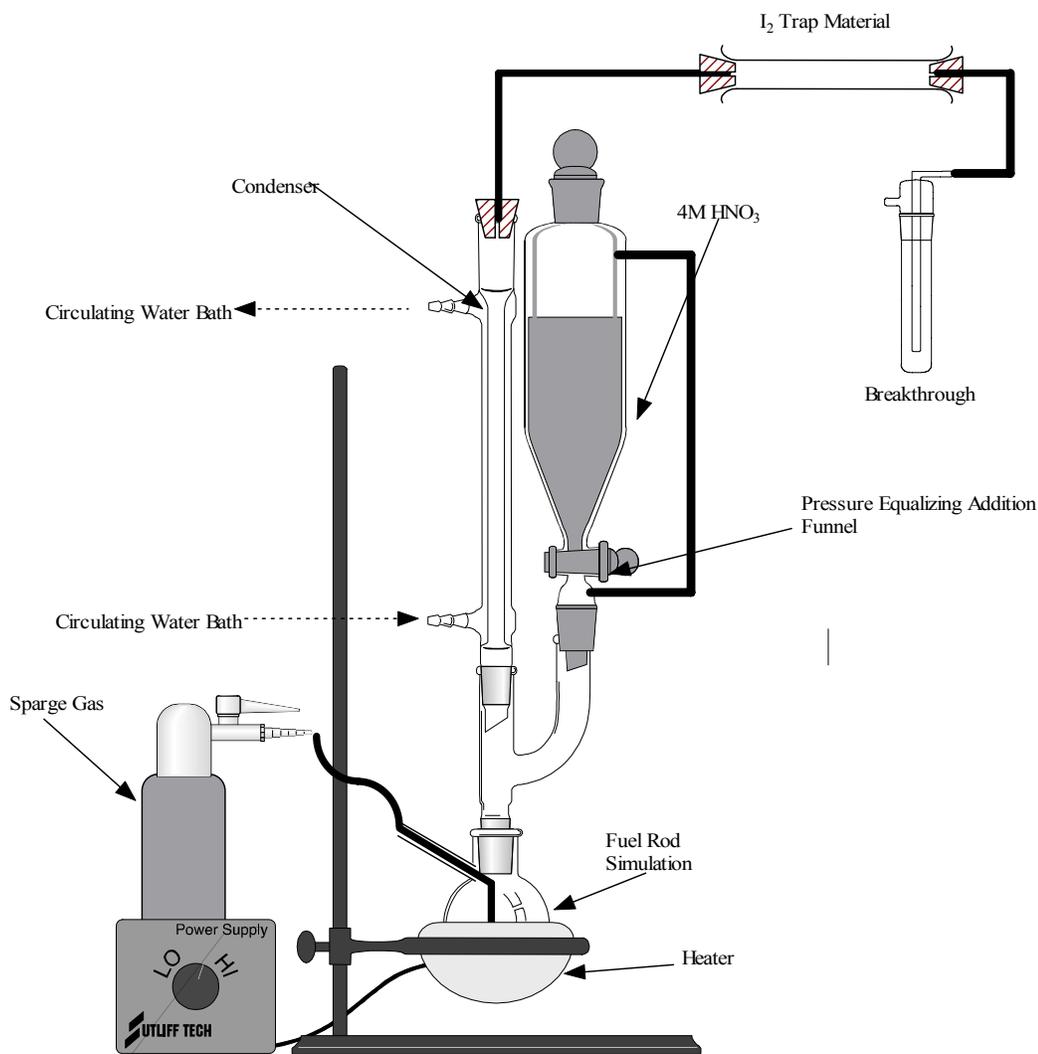


Figure 2: Simulation of Nuclear Fuel Dissolution

We propose to measure the thermal stability of the iodine-FCC association. This will be done by pyrolysis mass spectrometry of FCC material exposed to the iodine vapor in simulated fuel rod processing experiments. Pyrolysis will be performed using a temperature programmable pyrolyzer that can be interfaced to either a GC/MS for characterization of organic iodide compounds or to an ICP-AES or ICP-MS for measurement of iodine. This method will enable us to measure the decomposition temperature of the iodine-fullerene association and to characterize the products of decomposition. This information will be useful for assessing geological stability of FCC-iodine associations and for devising a method for recovery of iodine from FCC for transmutation. We anticipate doing additional studies to assess thermal stability of the iodine-FCC using Thermogravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC). Using these methods we may be able to associate iodine loss with thermally induced physical and chemical transformations of the FCC sorbent.

For initial studies with NOM we propose the following approach. The off-gas from nuclear fuel dissolution would be condensed using a cooled trap. This acidic condensate would be mixed CaCO_3 and NOM. A suitable oxidizing agent will be added to convert all of the iodine to HIO . Other buffering agents will be examined in order to determine the role of pH in the conversion process. The solid and aqueous fractions

will be centrifuged and separated for further characterization. Iodine bound in insoluble organic matter will be measured after alkaline oxidation. The approach utilized will be similar to the methods summarized in Copper et al. (1998), Olesky-Frenzedl et al. (2000), or Gu et al. (1997).

The potential production of volatile iodine species will be addressed as follows. The reaction system will be sparged with an inert gas and volatile species will be recovered on Tenax-activated charcoal. A commercial dynamic headspace concentrator will be used for this purpose. Volatile iodides will be thermally released from charcoal and analyzed by GC/MS.

Adsorbable organic iodide species will be measured after solid phase extraction (SPE) of aqueous phase with C₁₈ silica. The compounds retained by C₁₈ will be analyzed by GC/MS. Iodine associated with humic and fulvic acids will be recovered by resin extraction of the acidified aqueous solution. After alkaline destruction of the organic matter, this associated iodine will be measured using ion chromatography or by ICP-MS.

The speciation of iodine remaining in solution will be measured as follows. Under these reaction conditions remaining soluble inorganic iodine should consist only of HIO, IO⁻, IO₃⁻ and I⁻. Iodine/iodide and iodate can be analyzed by the method of Mishra et al. (2000). This method involves selective oxidation of iodide with 2-iodosobenzoate to produce active iodine that is subsequently reacted with N,N-dimethylaniline. The product p-iodo-N,N-dimethylaniline can be quantified by GC/MS. Iodate (IO₃⁻) will be determined by difference or by ion chromatography. Specific ion electrodes can be used to determine iodide in order to corroborate other analytical results. Total iodine in solution will also be measured using ICP-MS. If available we would also propose to use ICP-AES for sample screening and as a corroborative method of analysis. This approach is summarized in the Figure 3. We will also attempt to utilize the NOM for vapor phase sequestration of iodine using the experimental approaches outlined in Figures 1 or 2.

The execution of the project will involve using FCC prepared by the Khlopin Radiation Institute and NOM for preparation of vapor phase iodine trans. The chemical stability and form of the bound iodine will be inferred by various chemical and thermal experiments. In the case of NOM, the effect of pH, temperature on the reaction kinetics and the project distribution will be studied in depth. Once reaction conditions have been established refinements in the procedure such as pre-extraction of the NOM to remove soluble components and encapsulation of the NOM will be explored. In the final phases of the program conversion of the bound iodine (FCC or NOM) to a suitable transmutation target will be studied. This will likely involve an oxidation procedure as described above.

Proposed Experimental Fractionation Scheme

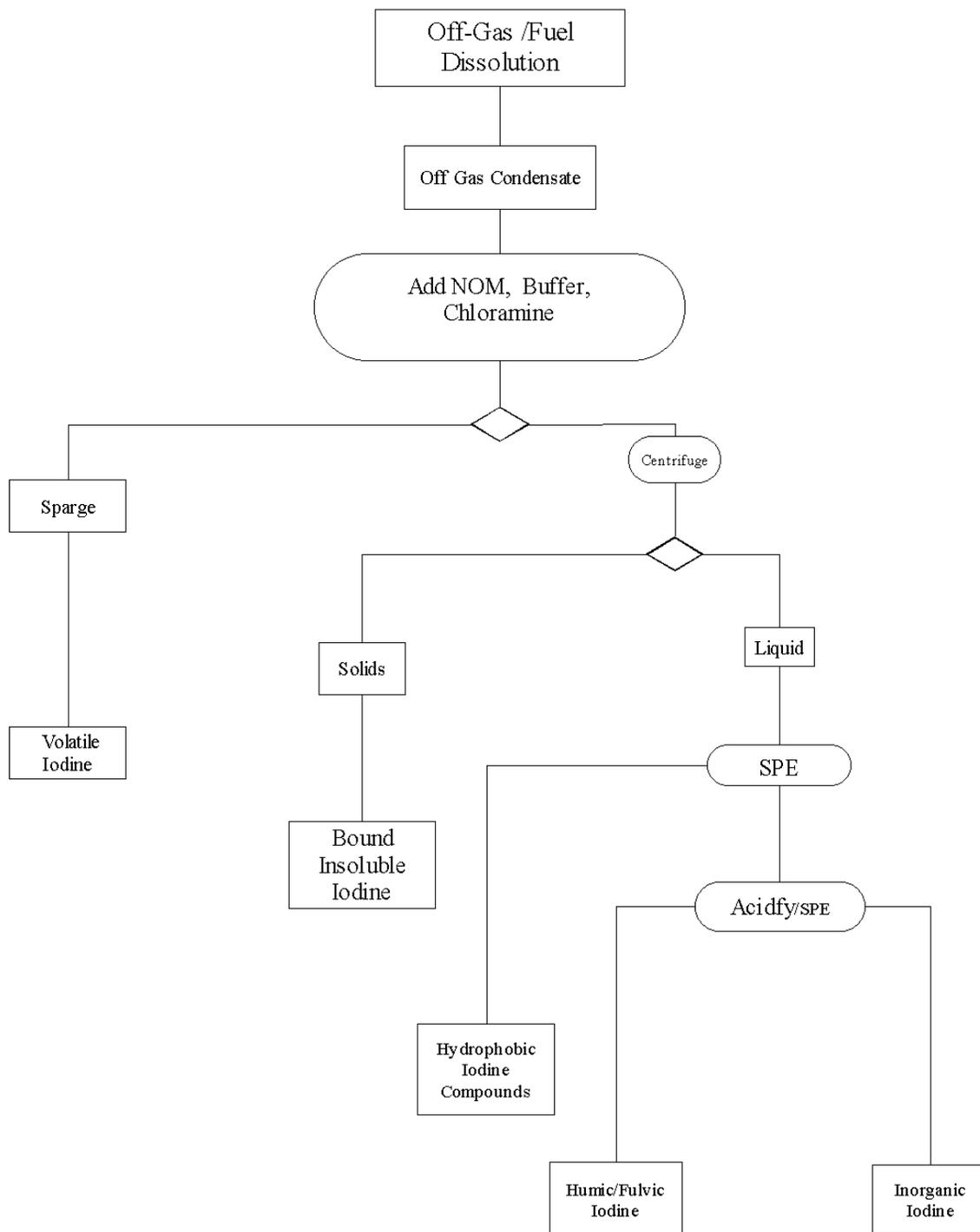


Figure 3: Proposed Fractionation

Capabilities at UNLV and KRI-KIRSI:

The majority of the equipment required to develop the sequestration matrices and characterize the reactions of iodine and these matrices is currently available at the UNLV and KRI-KIRSI facilities. These capabilities include gas chromatography, gas chromatography-mass spectrometry, ion-chromatography, ICP-MS (at UNLV); and XRD, SEM, EMPA, and experimental set-ups for MCC-1 protocol leach tests (at KRI).

A pyrolysis interface for use with the GC/MS will be acquired to support this work (this interface can also be used with the ICP-AES and ICP-MS systems). The bench-scale experimental set-up to simulate iodine release in the PUREX off-gas stream will be constructed using commercially available laboratory glassware, gas pressure regulators, flow regulators, and heaters. We are proposing to obtain two circulating temperature baths, and several heating mantles with a temperature controller for these experiments. In addition, various disposable supplies such as solid phase extraction cartridges and filters will be needed for the project.

If available at UNLV, an ICP-AES system capable of direct measurement of iodine (i.e. the Spectro CIROS CCD ICP-OES model FCE 12) will greatly increase the measurement capability of the UNLV team, allowing direct measurement of the off-gas vapor (with proper connections), and will provide another reliable technique for iodine measurements in the pyrolyzed sequestration matrices, and will be useful in examining solutions obtained from both leach test and iodine recovery experiments. If the ICP-AES system is not available, the available techniques will be used by the UNLV team.

Projected Timeline with Milestones and Deliverables:

This research program has been divided into several major tasks, which will be executed over a two- year period with the aid of a graduate assistant. Results will be communicated through written quarterly reports. The projected schedules for the UNLV and KRI components are broken down for this program and shown below.

Deliverables for Year 1:

- **Collaboration with DOE:** Regular communication with DOE collaborator to assess progress, discuss problems, and allow for refocusing if necessary to address shifts in direction by the National Program.
- **Quarterly Progress Reports:** Brief reports indicating progress will be provided every quarter in support of the DOE AAA quarterly meetings.
- **Annual Report:** Written reports detailing experiments performed, data collected and results to date.

Deliverables for Year 2:

- **Collaboration with DOE:** Regular communication with DOE collaborator to assess progress, discuss problems, and allow for refocusing if necessary to address shifts in direction by the National Program.
- **Quarterly Progress Reports:** Brief reports indicating progress will be provided every quarter in support of the DOE AAA quarterly meetings.
- **Final Report:** Written reports detailing experiments performed, data collected and results to date.

UNLV Work Timeline

Timeline Year 1

Task	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr
Literature Search	▨▨▨▨▨▨▨▨▨▨											
Preparation of Test FCC and NOM		▨▨▨▨▨▨▨▨▨▨										
Analytical Methods Testing			▨▨▨▨▨▨▨▨▨▨									
Set up Experimental Apparatus				▨▨▨▨▨▨▨▨▨▨								
Iodine Binding Experiments (FCC)					▨▨▨▨▨▨▨▨▨▨							
Iodine Binding Experiments (NOM)						▨▨▨▨▨▨▨▨▨▨						
Quarterly Reports			■			■			■			■

Timeline Year 2

Task	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr
Iodine Binding Experiments (FCC)	▨▨▨▨▨▨▨▨▨▨											
Iodine Binding Experiments (NOM)	▨▨▨▨▨▨▨▨▨▨											
Recovery of Bound Iodine		▨▨▨▨▨▨▨▨▨▨										
Conversion to Transmutation Target							▨▨▨▨▨▨▨▨▨▨					
Quarterly Reports			■			■			■			■
Write Final Report										▨▨▨▨▨▨▨▨▨▨		

KRI-KIRSI Timeline

ID	Task Name	2002					2003										2004											
		Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct
1	Laboratory Prep. for FCC fabrication																											
2	Synthesis of FCC																											
3	Examination of FCC loading for I																											
4	Synthesis of Immobilization forms (I-doped FCC & ceramic-like matrix)																											
5	Optimization of Immobilization forms (I-doped FCC & ceramic-like matrix)																											
6	Examination of Immobilization forms																											

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Associate Professor

Chemistry Department, University of Nevada, Las Vegas

EDUCATION

Post Doctoral Studies, Institute of Geophysics and Planetary Physics, University of California, Los Angeles, 1982-1984.

Ph.D. Marine Chemistry and Geochemistry, Scripps Institution of Oceanography, University of California, San Diego, 1982

B.A. (Summa Cum Laude) Chemistry, Revelle College, University of California, San Diego, 1978

PROFESSIONAL EXPERIENCE

- 1995- University of Nevada, Las Vegas
Associate Professor of Chemistry
- 1991 - 1995 University of Nevada, Las Vegas
Assistant Professor of Chemistry
- 1989 - 1991 C-E Environmental, Inc.
Senior Research Scientist
- 1988 - 1989 Radian Corporation, El Segundo, CA
Staff Scientist, Task Leader
- 1986 - 1988 Global Geochemistry Corporation, Canoga Park, CA Environmental Chemist and Project Manager
- 1985 - 1986 CT Agricultural Experiment Station, New Haven, CT Assistant Scientist
- 1983 - 1985 Institute of Geophysics and Planetary Physics, University of California Los Angeles, Los Angeles.
Post Graduate Research Geochemist

PROFESSIONAL AFFILIATIONS

American Chemical Society
-Environmental Chemistry Division

PUBLICATIONS

Jing, Linhong , Steinberg, S.; Johnston, B. J. (2001) Monocyclic Aromatic Hydrocarbon and Aldehyde concentrations at an Urban Site in Las Vegas, Nevada. *J. Air and Waste Management. Assoc.* 51, 1359-1366.

Kawamura, K. Steinberg, S., Kaplan, I.R. (2001) Wet deposition of low molecular weight mono- and dicarboxylic acids, aldehydes and inorganic species in Los Angeles, *Atmospheric Environment* 35,3917-3926.

Lantis I. Osemwengie, Spencer Steinberg (2001) In situ-Solid Phase Extraction and Laboratory Analysis of Trace Synthetic Musks in Municipal Effluent using GC/MS Full-scan mode. *J. Chromatography* 932(1-2), 107-118.

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- Johnson, E. A.; Rudin, M.J.; Steinberg, S. M.; Johnson, W. H. (2000) Sorption of selenite on Various Cement Formulations. *Waste Management* 7, 509-516.
- Kawamura, K.; Steinberg, S.; Kaplan, I. (2000) Homologous Series of C1-C10 Monocarboxylic Acids and C1-C6 Carbonyls in Southern California Air and Motor Exhaust.. *Atmospheric Environment* 34, 4175-4191
- George, S.; Steinberg, S. M. ; Hodge Vernon (1999) The Concentration, Apparent Molecular Weight and Chemical Reactivity of Silica from Groundwater in Southern Nevada, *Chemosphere* 38, 2143-2152.
- Steinberg, Spencer M.; Swallow, Charles Charles E.; Ma, W.K. (1999) Vapor Phase Sorption Of Benzene by Cationic Surfactant Modified Soil. *Chemosphere* 40, 57-63.
- Steinberg, Spencer M.; Schmeltzer, J. S.; Kreamer, D. K. (1996) Sorption of Benzene and Trichloroethylene (TCE) on a Desert Soil: Effects of Moisture and Organic Matter. *Chemosphere* 33 (5), 961-980.
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- Steinberg, S.M.; Poziomek, E.J.; Engleman, W.H. (1994) A Review of Applications of Luminescence to Monitoring of Chemical Contaminants in the Environment. *Chemosphere* 28, 1819-1857.
- Steinberg, S.; Fairely, J. P.; Kreamer, D. K. (1994) Slow Vapor-Phase Desorption of Toluene from Several Ion-Exchanged Montmorillonites. *J. Soil Contam.* 3, 249-264.
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- M.I. Venkatesan, E. Ruth, S. Steinberg and I.R. Kaplan (1987) Organic Geochemistry of Sediments from the continental shelf off Southern New England, USA-Part II. Lipids *Mar. Chem.* 21, 267-299.
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- Steinberg, S.M. and J.L. Bada. "The racemization of free and peptide bound serine and aspartic acid as a function of pH: Implications for in vivo racemization." *Bioorganic Chem.* 12, 349, 1984.
- Bada, J.L. and S.M. Steinberg. "Applications of amino acid racemization dating of fossil bones and teeth to problems in paleoanthropology". In: "Absolute Dating and Isotopic Analysis in Prehistory: Methods and Limits". Centre National de la Recherche Scientifique, Paris, 1984.
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- Steinberg, S.M. and J.L. Bada. "The diagenetic production of Alpha-keto acids in heated and fossil Chione shells." *Geochim. Cosmochim. Acta* 47, 1481, 1983.
- Steinberg, S.M. and J.L. Bada. "Peptide decomposition in the neutral pH region via the formation of diketopiperazines." *J. Org. Chem.* 48, 2295, 1983.
- Steinberg, S.M. and J.L. Bada. "The determination of Alpha-keto acids and oxalic acid in seawater by reversed phase liquid chromatographic separation of fluorescent quinoxilinol derivatives." *Mar Chem.* 11, 299, 1982.

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Steinberg, S.M. and J.L. Bada. "Diketopiperazine formation during investigation of amino acid racemization of dipeptides." *Science* 232, 544-545, 1981.

CURRICULUM VITAE

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Education, University: Degrees: Ph.D. in Chemistry, 1958, University of Michigan
M.S.Chem., 1954, University of Michigan
A.B. in Chemistry, 1952, Dartmouth College (cum laude, distinction in major)

Military Service: U.S. Army: April 1946-December 1947 (Infantry and Adjutant General's Dept.);
September 1950-September 1951 (Chemical Corps)

Professional Society Memberships: American Chemical Society, Royal Society of Chemistry, AAAS, AZ
NV Academy of Sciences, AAUP

Awards and Honors: **Engineer's Partner in Progress**, National Society of Professional
Engineers, Southern Nevada Chapter. **Exceptional Contributions to Engineering Education**, American
Society of Civil Engineers, Southern Nevada Chapter. **Gubernatorial Proclamation** for contributions to
higher education. **1999 Distinguished Service Award**, College of Sciences, UNLV

Employment Record: **1981 to 1998**, Professor of Chemistry, University of Nevada, Las Vegas (UNLV);
Emeritus, 7/1/1998. I taught introductory chemistry, general chemistry, organic chemistry, and polymer
chemistry and do research in organic chemistry, polymer chemistry, and analytical chemistry.
1988 - 1989: Dean, College of Science and Mathematics (UNLV). I had administrative and fiscal
responsibility for the Departments of Biological Sciences, Chemistry, Geoscience, Mathematical Sciences,
and Physics. There were approximately 60 faculty members distributed among the five departments.
1981 - 1988: Dean, College of Science, Mathematics and Engineering (UNLV). During this period I was
responsible for the science departments listed above plus the Engineering departments. Engineering was
developed from a five person, general engineering department, to a school with the Departments of Civil
and Mechanical Engineering and Electrical Engineering and Computer Science. Civil Engineering and
Mechanical Engineering earned ABET accreditation in 1987 and Electrical Engineering in 1988 about five
months after Engineering was spun off as a separate college. A \$12 million facility was planned and
constructed during this period. I was heavily involved in the planning and execution of this program, the
building project, and recruitment of faculty. The Architecture was affiliated with the College and was
started during the period. On the Science side, M.S. degree programs were developed in Chemistry and
Geology and a Biological Chemistry option for Chemistry majors was instituted.
1979-81: Dean, College of Arts, Sciences & Letters, The University of Michigan-Dearborn (UMD). This
undergraduate College encompassed the arts, humanities, mathematics, natural sciences, and social and
behavioral sciences. At the time, this was the third largest college in the three-campus University of
Michigan system at something over 3,000 full time equivalent students, and approximately 115 full time
faculty. A professional staff maintained the student records and managed a cooperative education program.

This was a time of severe financial distress in Michigan (17% unemployment) but budgetary cutbacks were achieved in a way that kept the College in the black and didn't wreck the programs.

1963 - 1981: Asst. Prof. of Chemistry, 1963, Assoc. Prof. 1966, Prof., 1969, (UMD) I joined UMD as the second chemist in two-man chemistry program and 525 students in the whole institution. I taught a heavy teaching load in analytical chemistry, organic chemistry, general chemistry, history of chemistry, and as a member of a team that taught a course entitled Matter, Energy, and Life which is still going strong today. We developed, with NSF support at the start, a Science Learning Center that is still going strong. The campus grew from 525 to 6,300 students while I was there and I was heavily involved more than my share of administrative tasks including department chair, division chair, and recruitment of a substantial number of faculty and administrators. I did research and published a number of papers.

1973-75: Chairman, Dept. of Natural Sciences, (UMD)

1967-69: Chairman, Division of Literature, Science, and the Arts (UMD)

1966: Acting Chairman of the Division of Literature, Sciences, and Arts (UMD)

1957-1963: Research Chemist, Shell Oil Co., Houston, TX and New York, NY I performed research in the general area of petroleum chemistry including, acid catalyzed oligomerization of light olefins, alkylation, isomerization of hexane, partial denitrification, desulfurization, and demetalizing of distillation residues. The final year was spent doing the work of 2 or 3 operators in the oil refinery during a labor dispute.

1954, summer: Research Chemist, Procter & Gamble Co. Ross, OH. I performed work attempting to learn something about the structure of lignin.

1952, summer: Research Engineer, Dow Chemical Co., Freeport, TX. I performed various analyses in a semi-plant that made alpha-methylstyrene oxide. During the last two weeks I assisted in a research project aimed at making synthetic glycerine from allyl chloride.

Courses taught at UM-D: Twenty-two different preparations

Courses taught at UNLV: Twenty-one different preparations including one in Physical Sciences and four in the Master of Arts in Liberal Studies program

Committee Service (UMD): Twenty-four different committees

Committees, UNLV and University and Community College System of Nevada System (UCCSN):
Thirty-six different committees

Graduate Student Committees: Daniel Fischer (Chair), Robert Curiale, Grazyna Orzechowska, Stephen Ward, Bhaskar Tadepalli, Russell Lidberg, Parviz Eivazi (Co-Chair), Zhen Wu, Ephraim Morris, Yu Zhang (Chair), Jill Jacoby (MALS), Diane Eggers (Physics), Mary Whelan, MALS

Current Research Interests:

Functional Group Polymers: I investigate the preparation, characterization, and reactions of polystyrene resins having as functional groups: sulfonyl chloride, sulfonamide, sulfonylhydrazine, thiol and sulfonic acid. The object is to carry out reactions in which the insoluble functional group polymer is one of the reactants and the other reactants are gases or are dissolved in liquids. I prepare polymers that coat surfaces and have chemically reactive functional groups that demonstrate uses in analytical chemistry. Some of the polymers incorporate fluorescent groups that are useful in detecting very small amounts of oxidizing agents such as chlorine or bromine. A current effort is directed toward detailed, quantitative determination of several different functional groups incorporated in the same polymer.

Mechanisms of organic reactions; sulfonium salts.

Publications since 1989:

Brian J. Johnson, David W. Emerson, Lining Song, Jennifer Floyd, Bhaskar Taddepali "Determination of Active Chlorine in Air by Bonded Phase Sorbent. Collection and Spectrophotometric Analysis" *The Analyst* **1999**, *124*, 1853-1857.

David W. Emerson, Richard L. Titus, Marlon D. Jones "A Ring-Opening Reaction of 6-Oxo-Substitued Spiropyrrolidinediones: Synthesis of 4-Substituted-1,5-dihydro-3-hydroxy-2-oxo-1,5-diphenyl-(2*H*)-pyrroles." *J. Heterocyclic Chem.* **1998**,*35*, 611-617.

Brian J. Johnson, David W. Emerson, Lining Song, Jennifer Floyd, Bhaskar Taddepali *Prepr. Pap. ACS Natl. Meet., Am. Chem. Soc. Div. Environ. Chem.* **1997**, *37*(2), 301-303.

David W. Emerson, Cheryl Grigorian, John W. Hess, Yu Zhang "Probing the Structures of Redox Polymers." *React.Funct.Polym.* **1997**, *33*, 91-101.

Stephen J. Butala; Kaveh Zarrabi; David W. Emerson "Sampling and Analysis for Lead in Water and Soil Samples on a University Campus." *J. Chem. Educ.* **1995**, *72*, 441-444.

David W. Emerson, "Microdetermination of Bromine, Chlorine and Chlorine Dioxide in Water in any Combination." *Microchem. J.* **1994**, *50*, 116-124.

David W. Emerson; Richard L. Titus; Spencer M. Steinberg; Marlon D. Jones "Kinetics and Mechanism of a Double Thiocyanate-Isothiocyanate Isomerization: A Round Robin Rearrangement." *J. Org. Chem.* **1994**, *59*, 5109-5110.

Boyd L. Earl, David W. Emerson, Brian J. Johnson, Richard L. Titus "A Course for teaching Practical Computer Skills to Chemistry Majors." *J. Chem. Educ.* **1994**, *71*, 1065-1068.

David W. Emerson, "Chlorine Dioxide Generated by Reaction of Sodium Chlorite with N-Halosulfonamide or N-Alkyl-N-halosulfonamide Groups on Styrene-Divinylbenzene Copolymers." *Ind. Eng. Chem. Res.* **1993**, *32*, 1228-1234.

David W. Emerson, Richard L. Titus, and Rowena M. González, "Evidence for Ketene Intermediates in the Decarbonylation of 2,4-Dioxo Acids and Esters and 2-Oxobutanedioic acid Esters." *J. Org. Chem.* **1991**, *56*, 5301-5307.

David W. Emerson, "Slow release of Active Chlorine and Active Bromine from Styrene-Divinylbenzene Copolymers Bearing N,N-Dichlorosulfonamide, N-Chloro-N-alkylsulfonamide and N-Bromo-N-alkylsulfonamide Functional Groups. Polymer Supported Reagents. 6." *Ind. Eng. Chem. Res.* **1991**, *30*, 2426.

David W. Emerson, Richard L. Titus and Rowena M. González, "Evidence for Ketene Intermediates in the Reactions of 2-Oxobutanedioic Acid Diesters with Alcohols and Water." *J. Org. Chem.* **1990**, *55*, 3572-3576.

Richard L. Titus, David W. Emerson, and Rowena M. González, "Synthesis of the (*E*) and (*Z*) Isomers of 1,2-diaryl-3-methyl-4,5-dioxo-3-pyrrolidincarboxylic Acid Esters. Structural Assignment by NMR and Mass Spectroscopy", *J. Heterocyclic Chem.* **1990**, *27*, 1857-1860.

David W. Emerson, "Polymer-Bound Active Chlorine: Disinfection of Water in a Flow System. Polymer Supported Reagents. 5." *Ind. Eng. Chem. Res.* **1990**, *29*, 448-450.

Presentations at Meetings since 1993

Emerson, D. W. "Classes Whose Member have Widely Different Levels of Preparation can be Taught Effectively" Forty Fifth Annual Meeting, Arizona-Nevada Academy of Sciences, Science Education Section, Las Vegas, NV , April 14, 2001

Emerson, D. W.; Titus, R. L.; Jones, M. D. "A Ring-Opening Reaction of 6-Oxo-Substituted Spiropyrrolidinediones" 214th American Chemical Society National Meeting, Las Vegas, NV Sept. 8, **1997**, Abstract # 135, Division of Organic Chemistry.

"Polymers for Protection and Detection" College of Sciences Colloquium, November 1, **1996**

"Probing the Structures of Redox Polymers" invited lecture, 7th Conference on Polymers and Organic Chemistry (POC'96), Wrocław, Poland, June 23-28, **1996**.

"Development of Polystyrene-Divinylbenzene Polymers with Sulfonamido Functional Groups" Wrocław Section of the Polish Chemical Society, May 31, **1994**.

BORIS E. BURAKOV

CURRENT POSITION:

Head of Mineralogical Group, Laboratory of Applied Mineralogy and Radiogeochemistry, V.G.Khlopin Radium Institute, Russian Ministry of Atomic Energy (MINATOM).

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PROFESSIONAL PROFILE:

- Experienced in material study of natural and artificial radioactive minerals, crystalline ceramic, glass etc.;
- Experienced in handling of highly radioactive materials including spent fuel and weapons Pu;
- Experienced in work under conditions of severe nuclear accident in Chernobyl.

EDUCATION:

MS Degree, Geology-Geochemistry-Mineralogy, Leningrad State University, former Soviet Union, graduation: June 1986; *Ph.D, Mineralogy*, Institute of Chemical Technology, Moscow, graduation: December 1991.

EXPERIENCE:

1985-86	Laboratory worker in V.G. Khlopin Radium Institute;
1986-91	Engineer-researcher in V.G. Khlopin Radium Institute, mineralogical and geochronological study of uranium ores;
1991-present time	Head of Mineralogical Group, V.G. Khlopin Radium Institute;
1990-92	Participation in the investigation of Chernobyl accident, material study of destroyed spent fuel, Chernobyl "lava" and hot particles;
1996	An expert of International Atomic Energy Agency (IAEA), personal No T636373, Nature of Services "Environmental Impact for the Chernobyl NPP Unit 4, Study of Interaction of Water and FCM";
1990-present time	Experimental research on the synthesis and examination of crystalline ceramic for immobilization of HLW including TRU and weapons Pu.

SELECTED PUBLICATIONS:

- (1) Burakov, B.E, Anderson, E.B. (1998) Development of Crystalline Ceramic for Immobilization of TRU Wastes in V.G.Khlopin Radium Institute. *Proceedings of the 2nd NUCEF International Symposium NUCEF'98, 16-17/11/98, Hitachinaka, Ibaraki, Japan, JAERI-Conf.99-004 (Part I), 295-306.*
- (2) Burakov, B.E, etc. (1999) Ceramic Forms for Immobilizing Pu Using Zr, Y, Al Metal Additives. *Environmental Issues and Waste Management Technologies IV*, 349-356.
- (3) Burakov, B.E, Anderson, E.B, Zamoryanskaya, M.V, Petrova, M.A. (2000) Synthesis and Study of ²³⁹Pu-Doped Gadolinium-Aluminum Garnet. *Material Research Society Symposium Proceedings Scientific Basis for Nuclear Waste Management XXIII, Vol.608, 419-422.*
- (4) Burakov, B.E, Hanchar, J.M, Zamoryanskaya, M.V, Garbuzov, V.M, Zirlin, V.A. (2002) Synthesis and Investigation of Pu-Doped Single Crystal Zircon, (Zr,Pu)SiO₄, *Radiochimica Acta*, 90, 95-97.