

Nuclear Materials Characterization in the Materials and Fuels Complex Analytical Hot Cells - 9275

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ABSTRACT

As energy prices skyrocket and interest in alternative, clean energy sources builds, interest in nuclear energy has increased. This increased interest in nuclear energy has been termed the “Nuclear Renaissance”. The performance of nuclear fuels, fuels and reactor materials and waste products are becoming a more important issue as the potential for designing new nuclear reactors is more immediate. The Idaho National Laboratory (INL) Materials and Fuels Complex (MFC) Analytical Laboratory Hot Cells (ALHC) are rising to the challenge of characterizing new reactor materials, byproducts and performance.

The ALHC is a facility located near Idaho Falls, Idaho at the INL Site. It was built in 1958 as part of the former Argonne National Laboratory West Complex to support the operation of the second Experimental Breeder Reactor (EBR-II). It is part of a larger analytical laboratory structure that includes wet chemistry, instrumentation and radiochemistry laboratories. The purpose of the ALHC is to perform analytical chemistry work on highly radioactive materials. The primary work in the ALHC has traditionally been dissolution of nuclear materials so that less radioactive subsamples (aliquots) could be transferred to other sections of the laboratory for analysis.

Over the last 50 years though, the capabilities within the ALHC have also become independent of other laboratory sections in a number of ways. While dissolution, digestion and subdividing samples are still a vitally important role, the ALHC has stand alone capabilities in the area of immersion density, gamma scanning and combustion gas analysis. Recent use of the ALHC for immersion density shows that extremely fine and delicate operations can be performed with the master-slave manipulators by qualified operators. Twenty milligram samples were tested for immersion density to determine the expansion of uranium dioxide after irradiation in a nuclear reactor. The data collected confirmed modeling analysis with very tight precision. The gamma scanning equipment in the ALHC has taken on a new role also as a micro-gamma scanning system and has been put into service; allowing the linear and radial counting of a spent fuel segment to determine reaction characteristics within a small section of nuclear fuel. The nitrogen, oxygen and carbon analysis allows the identification of these impurities in spent nuclear fuel and also most oxides, nitrides, carbides, C-14 and tritium.

INTRODUCTION

Increasing energy costs over the last few years have caused a great deal of interest to be generated in alternative methods of conserving and generating energy. The upsurge in new, more fuel-efficient cars, better lighting concepts and other conservation methods is evidence of this. Also evidenced is the proliferation of windmills in the west and interest in many other alternative energy sources such as solar, biofuels and coal. Among the alternatives is the generation of electricity using nuclear energy. Ten new reactor applications have been submitted in the past two years. Nuclear energy is also attractive as because of its lower emissions at a time when carbon dioxide damage to the environment is debated with great contention.

One aspect of the Nuclear Renaissance is the further development of nuclear energy sources for higher efficiency, lower cost, improved safety and proliferation resistance. The world’s reactor designers now have several “third generation” reactors that require less time to build and reduce capital costs. The Westinghouse AP1000 is expected to reduce the construction cycle to 36 months, provide additional

inherent safety measures, have an operating life of 60 years and produce electricity for the consumer at \$0.035 per kilowatt hour. Other designs make up to 40% more electricity per reactor than previously expected and can operate on less desirable fuels than enriched uranium.

The Department of Energy (DOE) has a strong role in providing leadership for the nuclear renaissance. Besides teaming with industry to accelerate building of existing technology reactors, the DOE is actively developing the next generation of nuclear reactors and working on closing the fuel cycle. DOE leads the Generation IV International Forum (Gen IV) working on six advanced nuclear reactor designs that employ significantly different fuels. As part of the Gen IV forum, DOE has an Advanced Fuel Cycle Initiative (AFCI) that seeks to develop and demonstrate proliferation resistant technologies that will allow the recycling and treatment of commercial spent fuel. Part of the AFCI is partitioning of the spent fuel components and fission (or burning) of the actinides to make a waste that is much more resistant to proliferation. A favored option for burning the actinides is the use of a fast neutron reactor.

Reactor and Fuel Reprocessing Facilities at the INL

The Idaho National Laboratory (INL) has been a leader in reactor development for over 50 years. There have been 50 different reactors built and operated at the INL. In addition to the conventional boiling and pressurized water reactors the INL has operated two experimental breeder reactors. Fuel reprocessing has also been featured at the INL, primarily at the Idaho Nuclear Technology Engineering Center (previously known as the Idaho Chemical Processing Plant), where about a dozen different spent fuels (mainly oxide fuels) were reprocessed to recover the highly enriched uranium in PUREX (plutonium, uranium extraction) and REDOX (reduction, oxidation process to extract uranium) processes. A separate effort at the Experimental Breeder Reactor – II Facility (formerly the Argonne National Laboratory – West) developed and demonstrated the electrochemical treatment of metallic, fast reactor spent fuel. A major effort at the EBR-II was the development of the Integrated Fast Reactor (IFR) to close the fuel cycle for breeder reactors. The electro-refining process continues as a method of treating sodium bonded spent fuel.^{1, 2}

Several hot cells (remote facilities shielded from highly radioactive materials) at the INL's Materials and Fuels Complex (MFC) supported the EBR-II, IFR and currently support the electro-refining treatment processes and fuel experiments. The electro-refining portion of the IFR takes place in the Fuel Conditioning Facility (FCF), which is attached to the EBR-II. Sodium-bonded fuel from fast reactors around the country is processed at the FCF where uranium and plutonium can be recovered from the metallic fuel. Treating the fuel to react with the metallic sodium bonding allows the waste products to be disposed of without undue hazards. Experiments and fuel examination take place in the Hot Fuel Examination Facility (HFEF). Chemical and physical analyses of the electro-refining feed and products take place at the Analytical Laboratory's Hot Cells (ALHC). Support of other projects, such as characterization of highly radioactive waste or source materials from other facilities, may also be accomplished in the ALHC.

ALHC REMOTE CHEMICAL AND PHYSICAL ANALYSIS

The ALHC is part of a large analytical chemistry laboratory facility at the MFC. This 1,050 m² laboratory complex is the sole remaining moderate or high level radiological laboratories at the INL. It consists of the A-Wing, which houses the six connected hot cells, a series of glove boxes and mechanical operations, the B-Wing, which houses general laboratory, high alpha contamination glove boxes and radiological hoods and the Sodium Wing, which houses general laboratories and glove boxes for casting metallic nuclear fuel. A basement floor houses most of the ventilation blowers, filters and storage areas. Each hot cell is about 2 meters on a side and uses 2 Central Research Laboratories Model L heavy duty master/slave manipulators. Samples are primarily received from the FCF (and from HFEF through FCF)

via a pneumatic sample transfer system. A cart system moves samples and materials between the six cells.³

Dissolution Capabilities

Routine electrorefining samples are derived from two sources, driver and blanket sodium bonded fuel rods. These two fuels are chopped into segments of sizes of 1 inch or less. These pieces are then run through the electrorefining process. The material collected is melted and cast into ingots. The types of samples received from this process are shown on Table I and detailed as follows⁴:

- Fuel Segment – Fuel rod from a driver or blanket assembly that is chopped into pieces of 1 inch or less
- Casting Pins – Material from ingot drawn up through a glass tube and cut into ¼ to ½ inch pieces
- Drill Fines – Material drilled from an ingot
- Cladding Hulls – Fuel segment material left over from the electrorefining process
- ER Salt – Electrorefiner Salt material collected using a dip cup or drawn up through a filtered tube
- ER Cadmium - Electrorefiner Cadmium material collected using a dip cup or drawn up through a filtered tube
- Casting Pins/ Drill Fines Silica Determination – Same material as above, but uses a cold dissolution process.

The following shows the sample type and the acid matrix used for dissolution.

Table I, Samples Routinely Dissolved for Analysis

Sample	Fuel Type	Acid Matrix
Casting Pins/ Drill Fines	Driver	Nitric/ HF
Casting Pins/ Drill Fines	Blanket	Nitric/ HCl
Fuel Segment	Driver	Nitric/ HF
Fuel Segment	Blanket	Nitric/ HCl
Cladding Hull	Driver	Nitric/ HF
Cladding Hull	Blanket	Nitric/ HCl
ER Salt	n/a	Nitric
ER Cadmium	n/a	Nitric
Casting Pins/ Drill Fines, Silica	Driver/ Blanket	Nitric/ HF

New fuel characterization utilizes the ALHC dissolving capabilities. The different types of new fuels that are provided for dissolution vary from spent fuel to developmental fuel. While working on the developmental fuels there is un-irradiated and irradiated material. While the hot cells concentrate on the irradiated material, the remaining Analytical Lab provides support by doing dissolution research on the

un-irradiated material as well. The different fuels the ALHC have worked on include Belgium Reactor 3 (BR3), Mixed Oxide (MOX), Japanese Atomic Energy Agency (JAEA) and Commercial (M5).

Immersion Density

One example of the physical analyses performed in the ALHC is the immersion density of highly radioactive samples. This procedure measures the density of a material based on the Archimedes' Principle. The Archimedes' Principle used the displacement of water to determine the mass per unit volume, thus determining the density for objects of complex geometry (a solid gold kings crown for example versus an alloyed crown). By immersing a sample of the material in water a separate density may also be determined that differs from "bulk density"; small voids will be filled with fluid during the measurement. This measurement is especially useful for determining the growth or swelling changes that may go on during irradiation of materials. Extremely small voids and cracks may not be filled by the water and this measurement does not give an absolute density or porosity. During the procedure a sample is immersed in a water bath and weighed. The temperature of the water is then measured by a thermocouple and the density of displaced water is calculated for that temperature.

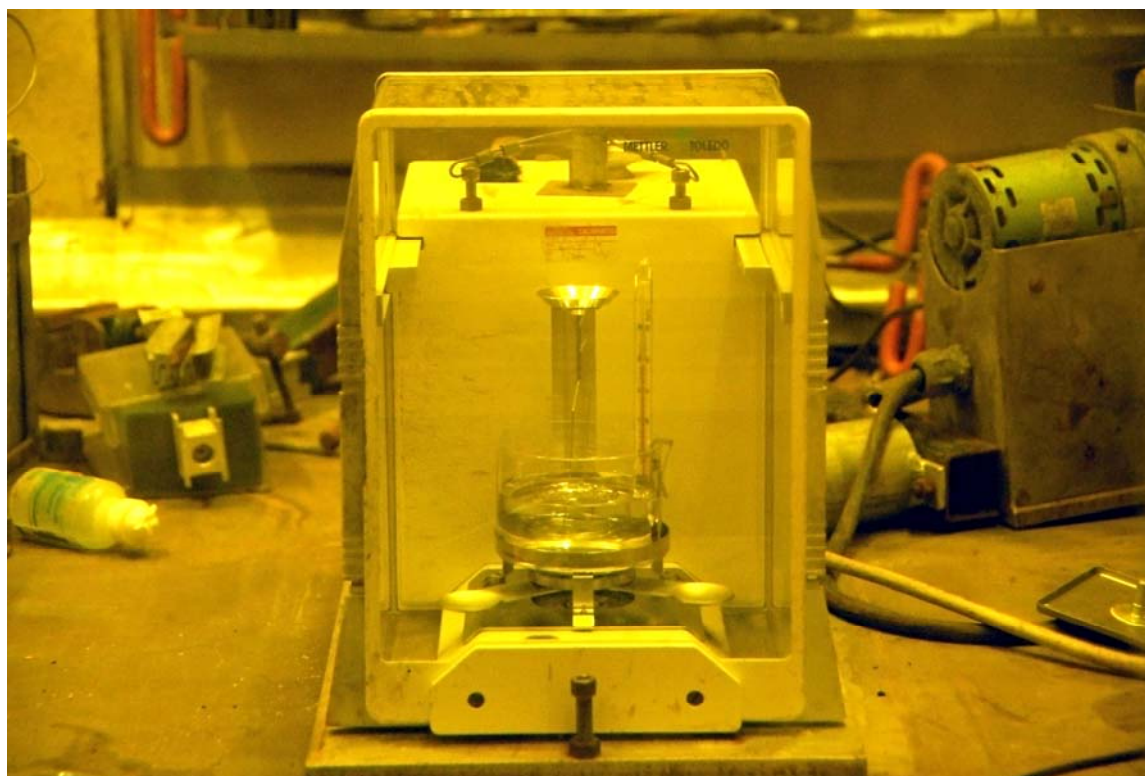


Figure 1, Remote Immersion Density System.

For the purpose of performing immersion density a standard analytical balance and an additional piece of equipment that substitutes for the balance pan are used. A Mettler AT-400 balance, shown in Figure 1, was used. This balance carries a precision of about 0.3 mg (as configured in the hot cell) and is modified with remote electronics and fixtures to aid in remote manipulations. The immersion density determination kit (Figure 1, mounted in the balance) is available from Mettler as an add-feature (PN 210485) for the AX/AT balance. The kit consists of a bracket that suspends the holder, holder/basket that is immersed in the water, a glass beaker and a thermometer. The kit was not difficult to assemble, but required some dexterity to mount on the balance.

The use of immersion density equipment requires very fine use of the hot cell manipulators because of the delicate nature of the equipment and samples. The spent fuel samples being examined were pieces of spent fuel pellet, averaging about 2 mm in size (about 20 mg) as shown in figure 2. The requested precision for the measurement was 1%. Bench testing performed with such small samples indicated that that level of precision would require multiple small samples pieces being weighed together five times (removed from pan and reentered each time). Immersion control standards were purchased from the Quantachrome Company and used to validate the method both on the bench top and in cell. Agreement between the density determined by the spent fuel modeling code and the analysis was less than two standard deviations (<1%) in each case and was approximately correct in terms of bias (slightly higher) for all samples.

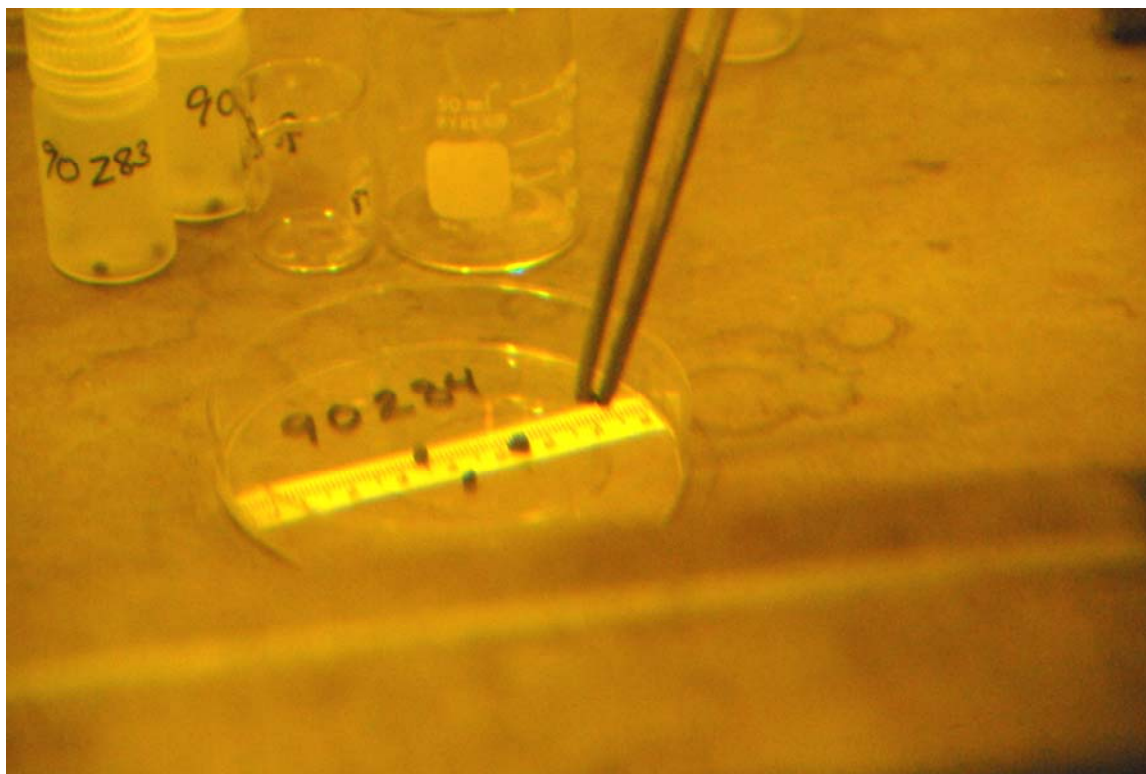


Figure 2, Small Fuel Pieces Manipulated in the ALHC for Immersion Density

Gamma Detector System

ALHC Cell #4 has a four-inch penetration through the rear wall to allow for the insertion of a germanium gamma-ray detector. The detector is isolated from the hot cell environment using only relatively thin aluminum foil that is functionally transparent to gamma rays. Connected to a modern DSP-based acquisition system, this system provides a valuable tool in the radioanalytical arsenal of the analytical lab at the Materials and Fuels Complex of the INL.

This gamma-ray detector system is currently calibrated for three basic geometries, a 30 ml liquid, a point source, and a 2 gallon can. A simple sample-transport track provides the ability to vary the sample-to-detector distance. Additionally, one of three lead collimators, approximately seven inches in length, can be placed between the sample and the detector. Four collimators are currently available: one-eighth inch,

one-quarter inch, one-half inch and 1 inch. Working in concert, the various pieces of this system provide a flexibility that allows for a wide range of samples and activity levels.

The hot-cell gamma detector system is generally used for three types of samples. It is used to assay dissolved, irradiated nuclear fuel, to assay hot-cell waste products, and to assay irradiated fuel segments for PIE purposes. Dissolved nuclear fuel is gamma-scanned in the hot cell as a matter of convenience as much as anything. Conceptually, it would be possible to dilute the solution sufficiently to remove it from the hot cell and count it on a standard gamma-ray detector. It is somewhat easier, however, to perform a minimal dilution/geometry correction in the hot cells and count them. This also helps to minimize waste and to cut down on the amount of sample material that must be returned to the hot cells for disposal. Hot cell waste is also counted using the same gamma ray detection system. Predominantly, waste is counted in 2 gallon cans that are crushed before removal from the hot cells. However, various other waste forms are counted using different geometries. The waste assay results are combined with process knowledge to allow for disposal of waste of all types.

An exciting innovation in the Analytical Lab uses the hot cell gamma ray detector and tungsten collimators with very small apertures to perform areal scans of irradiated materials for post-irradiation examination (PIE). Using electrical discharge machining, a four-inch tungsten collimator was built with a 0.5 mm aperture. Using a precision linear stage, samples can be moved across the collimator in very small steps (< 0.5 mm). Using this "microgamma" scanning technique, the analytical lab staff is able to radially scan fuel pin segments to measure the mobility of volatile species, to scan fuel compacts for position-specific burnup measurements prior to disassembly, as well as any measurement with small areal requirements.

The hot cell gamma-ray detector system at the analytical lab provides a strong capability for measuring various samples at an extreme range of activity levels. Performing these measurements in the hot cell minimizes waste, saves time, and, in keeping with ALARA principles, keeps dose to the staff at a minimum.

Combustion Gas Analysis

The Analytical Laboratory Hot Cells have a unique ability for analysis of nitrogen, oxygen and carbon content of radiological samples. A LECO TC-436 and IR-412 have been reengineered for radiological Hot Cell usage (Figure 3). The remote furnace nitrogen/oxygen analyzer is a bench-top LECO TC-436 with an EF-400 electrode furnace and the remote furnace carbon analyzer is a bench-top LECO IR-412 carbon determinator with HF-400 induction furnace modified for remote furnace operation in the hot cell. The hot cell remote furnace nitrogen/oxygen and carbon analyzers have a remote (hot cell operating area) and local (hot cell service area) operating station, which is a moveable instrument cart that consist of a video monitor, keyboard control, and printer. A HEPA-filtered vacuum cleaner in hot cell No. 3 is used to for cleaning the remote electrode furnace of the N/O analyzer and for trapping particulate contamination ejected during the pinch valve operation, cleaning of combustion tube and lance assembly, and for cleaning of remote induction furnace of the Carbon analyzer. Inlet and exhaust for both instruments are mitigated through engineering controls in conjunction with in line HEPA filters and Hot Cell ventilation. The accuracy and precision of the hot cell remote furnace nitrogen/oxygen and carbon analyzers meet manufacturer's bench-top performance specifications.



Figure 3, Remote LECO equipment for Nitrogen/Oxygen and Carbon Analysis

The remote furnace nitrogen/oxygen analyzer is used to determine total nitrogen and oxygen content and nitride/oxide concentrations of:

- Irradiated or contaminated reactor hardware (stainless steel or others) samples
- Unirradiated and conditioned metallic spent fuel
- Plutonium metal
- Li/Pu metal oxides

The remote furnace carbon analyzer is used to determine the carbon content of:

- Irradiated or contaminated reactor hardware (stainless steel) samples or other metals and alloys carbon-14 (C-14), and tritium (H-3)⁵
- Unirradiated conditioned spent metallic fuel
- Beryllium metal C-14 and H-3
- Unirradiated plutonium, neptunium, and uranium metals.

Irradiated fuel samples are not analyzed by the remote furnace nitrogen/oxygen analyzer and unconditioned irradiated fuel samples are not analyzed by the remote furnace carbon analyzer due to radiological hazards associated with released fission gases in the system piping outside the hot cells.

CONCLUSION

The ALHC is a versatile facility for the remote characterization of highly radioactive nuclear and radiological materials. It is integrated into a facility system that allows dilutions of dissolved materials to be processed at much lower hazard to personnel in other parts of the laboratory. The versatility of the hot cells allows operators to do remote manipulations that require great dexterity such as combustion gas, immersion density and micro-gamma scan analyses. The results of these analyses show that even under remote conditions very tight precision can be obtained.

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