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Optimizing High Level Waste Disposal

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ABSTRACT

If society is ever to reap the potential benefits of nuclear energy, technologists must close the fuel-cycle completely. A closed cycle equates to a continued supply of fuel and safe reactors, but also reliable and comprehensive closure of waste issues. High level waste (HLW) disposal in borosilicate glass (BSG) is based on 1970s era evaluations. This host matrix is very adaptable to sequestering a wide variety of radionuclides found in raffinates from spent fuel reprocessing. However, it is now known that the current system is far from optimal for disposal of the diverse HLW streams, and proven alternatives are available to reduce costs by billions of dollars. The basis for HLW disposal should be reassessed to consider extensive waste form and process technology research and development efforts, which have been conducted by the United States Department of Energy (USDOE), international agencies and the private sector. Matching the waste form to the waste chemistry and using currently available technology could increase the waste content in waste forms to 50% or more and double processing rates. Optimization of the HLW disposal system would accelerate HLW disposition and increase repository capacity. This does _not_ necessarily require developing new waste forms, the emphasis should be on qualifying existing matrices to demonstrate protection equal to or better than the baseline glass performance. Also, this proposed effort does _not_ necessarily require developing new technology concepts. The emphasis is on demonstrating existing technology that is clearly better (reliability, productivity, cost) than current technology, and justifying its use in future facilities or retrofitted facilities. Higher waste processing and disposal efficiency can be realized by performing the engineering analyses and trade-studies necessary to select the most efficient methods for processing the full spectrum of wastes across the nuclear complex. This paper will describe technologies being evaluated at Idaho National Laboratory and the facilities we've designed to evaluate options and support optimization.

Keywords: HLW, high level waste, disposal, CCIM

INTRODUCTION

Einstein said, "*The definition of insanity is doing the same thing over and over again and expecting a different result.*"

In the United States the nuclear industry has an opportunity to do better, to finally realize the potential of nuclear energy as a clean, essentially unlimited source of energy. Coupled with hydrogen generation, both clean fixed sources of electricity and innovative clean fuels for transportation can be developed. Matched with water desalination, industry can continue to make arid land productive and continue to provide high quality water for a healthy growing population. However, technologists also have the potential to fall victim to habits and live Einstein's famous quote describing insanity.

Enjoying the benefits of hindsight, it could be said society was blinded by the enormous potential seen in a nuclear age, where energy would be almost free…

"It is not too much to expect that our children will enjoy in their homes electrical energy too cheap to meter..."

- Lewis Strauss, Chairman of the Atomic Energy Commission, 1954

"Heat will be so plentiful that it will even be used to melt snow as it falls....[T]he central atomic power plant will provide all the heat, light, and power required by the community and these utilities will be so cheap that their cost can hardly be reckoned."

-Robert M. Hutchins, president of the University of Chicago, site of the first nuclear chain reaction, 1946

Industry has learned much since then. Three Mile Island, Chernobyl, and tens of billions of dollars spent on cleaning up radioactive contamination around the world are material proof that the nuclear industry can do better. Idaho National Laboratory (INL) is now working on Generation IV (GenIV) reactor design and a more efficient fuel cycle. INL will collaborate with foreign and domestic organizations to design reactors that can only fail safely, with inherent limits on temperature and criticality making a core meltdown physically impossible. The focal point for the GenIV research in the USA is INL

Further, industry has much greater insight into the fuel-cycle than ever before. Though the USA is not currently reprocessing fuel, industry is evaluating new fuel-cycles. The fuel-cycle of the future may not only produce electrical power and heat while "burning" (fissioning) weapons-grade plutonium; it may also "burn" unwanted transuranic (TRU) elements, and breed new fuel to make a self-sustaining fuel supply. In addition to making it possible to recycle selected isotopes for beneficial applications or concentrated to be destroyed, new separations concepts can be designed with the knowledge of how the wastes are to be dealt with in the long term. It could make it possible to remove isotopes that cause the long-term heat burden on a repository, or to remove isotopes that are extremely challenging to sequester in a waste form. These isotopes can be removed for destruction in the reactor as targets, or isolated for innovative uses or tailored disposal. The Advanced Fuel Cycle Initiative is also centered at INL.

This brings us to the third and perhaps our greatest responsibility, closing the fuel-cycle by addressing the waste issues. Reactors designed to burn problematic TRU and separations processes that minimize high level waste (HLW) are important steps forward, but if industry ever expects popular support to build these new designs, the waste segment of the fuel-cycle must be closed. Waste treatment is the Achilles heel of nuclear science and engineering. A nuclear future in medicine, space exploration, and diagnostics as well as energy to light homes, and to provide clean fuel and water rests on our ability to responsibly close the cycle completely. Closure should include reliable technology designed to safely dispose of wastes in a system optimized for cost effectiveness. The public is concerned with spent fuel management, shipping, storage, and what becomes of the wastes. In years passed, there was significant concern over radiation in general, and horrific effects were the plots of many books and movies in the popular media. Today, industry has the benefit of 60 years of epidemiological data, can focus concerns on specific wastes issues; in some cases industry can focus on particular nuclides and their potential impacts on human health and the environment. Industry can readily identify these issues, because they are the same problems that have been researched for 30 years, namely waste form manufacture, immobilization efficiency and performance, and clean emissions.

WASTE ISSUES

Making durable waste forms that can accept a wide variety of elements at significant concentrations is challenging. Making these waste forms homogenous and making them last for 10,000, 100,000, or 1,000,000 years is even tougher. Proving that they will last has not yet been done to the satisfaction of the public. It has been over 20 years since the decision was made to convert HLW into glass in the USA[i,ii] but industry is still faced with problematic elements that are very difficult to immobilize. Glass was chosen because its vitreous nature makes it a flexible host matrix for cations of different sizes and charge, but anions like sulfur and phosphorous have very limited solubility in borosilicate glass (BSG), which results in low waste loading and more glass with some wastes. Iodine can be lost as a gas at the temperatures it takes to make glass. Chloride and pertechnetate are notoriously soluble in water, and challenge glass durability in a repository. All of these facts suggest that additional waste forms may be necessary to optimize HLW disposal by matching the waste form to the waste chemistry. In addition, industry must be able to manufacture waste forms that provide high-performance immobilization, without generating secondary waste emissions that complicate licensing of the waste processes. Since the 1960s, the public has insisted on and won representation in the decision making process. Thus, nuclear technologists must design acceptable waste forms with processes to manufacture them, and ensure that secondary wastes are dealt with safely if the world is to realize the benefits of sufficient energy, clean fuel and water to support a sustainable future. The R&D conducted over the last 30 years should be reevaluated to optimize HLW disposal.

WASTE FORM MANUFACTURE

United States Department of Energy HLW immobilization systems are based on joule-heated melters (JHM), which is also the standard technology for producing HLW glass in Russia and Japan.. This technology, based on melting glass by passing electrical current through a molten pool contained in a refractory brick basin, has many years of successful deployment in industry. This technology is best suited to process consistent glass chemistry, in large quantities, within the a relatively modest temperature range. The designs for processing HLW are much more expensive that industrial designs, using highly corrosion resistant refractories and high-nickel alloy electrodes, but the technology is essentially the same. Many HLW applications will push the operating envelope for the technology to the extreme for materials durability.

- The chemistry of HLW is inconsistent.
- The temperatures required to incorporate refractory elements into BSG (i.e. Al, Cr, and Zr) in significant quantities exceed the normal operating range of the materials available for these melters.
- Many HLW compositions are highly corrosive and contain significant volatile species.
- Maintenance in a highly radioactive environment is very expensive.
- Melter lifetime is very limited (4-7 years); even state-of-the-art facilities are ill equipped to dismantle and dispose of the large, highly contaminated spent units.

Efforts to optimize the DOE HLW disposal system must include investigation and implementation, as appropriate, of alternative process technologies to produce the waste forms more effectively, which will provide both mid-term and long-term benefits to the USDOE. This is because some of the additional waste forms that have been shown to be the most beneficial for specific HLW inventories cannot be produced effectively, if at all, in a JHM.

While the JHM is a proven technology, it also has significant limitations due to temperature and corrosion constraints, particularly in regard to processing the widely varied and challenging chemistry of the DOE HLW inventory.[iii,iv] Alternative melter technologies have repeatedly been shown to offer potential improvements in cost-effectiveness and system optimization than can be achieved through continued use of JHMs.[v,vi,vii]

For example, the operational life of a JHM is relatively short (i.e. 5 to 7 years) and this can be further reduced when processing waste that is particularly aggressive, or operating at higher temperatures to enhance waste loading. Replacement of a JHM is costly and significantly impacts the USDOE schedule commitment to the host state. Recently, the JHM at the heart of the Defense Waste Processing Plant (DWPF) at the Savannah River Site in South Carolina was replaced for approximately $115M in combined equipment and facility downtime costs. In addition, these costs do not include the final dismantling and disposal of the highly radioactive melter carcass. Conversely, a substitute melter concept such as the cold crucible induction melter (CCIM) is smaller, modular, and has the potential to provide much longer operational life.[viii,ix,x] As a matter of fact, CCIM units do not appear to have any particular limitations on service life, should last for the life of the facility, and their compact design makes them ideal candidates for retrofit. A CCIM could replace the JHM in the DWPF at their next replacement with little additional cost, thereby avoiding all future melter replacement. If the DWPF plans to replace their JHM only one additional time over the projected life of the plant, there would a return on investment of nearly 100%. This does not include the avoided costs of melter disposal, additional potential savings due to increased waste loading, and increased production rate made possible by the enhanced operational flexibility offered by CCIM technology.

One engineering analysis commissioned by the USDOE to evaluate options for the Hanford Waste Treatment Plant (WTP) showed that CCIM technology offered the potential for waste forms containing up to 47% more waste, and production rates more than double the baseline JHM technology.[xi] The CCIM has also been shown to provide a greater operating envelope with the ability to effectively process a broader range of waste forms in terms of corrosion and processing temperature. The baseline JHM is nominally limited to 1150^{o}C, whereas optimal temperature for vitrifying Hanford HLW has been estimated to be near 1350^{o}C[xi], a compromise between waste loading and volatilization of problematic species. The JHM design also limits glass formulation by constraints on liquidus temperature to prevent buildup of crystals on the bottom of the melter. Another study for Hanford by Pacific Northwest National Laboratory showed that this single constraint limits waste loading to less than 34 wt% for neutralized acid waste, whereas glass durability would allow waste loading as high as 70 wt%.[xii] Estimates range as high as $670 million for a 1 wt% increase in waste loading for processing Hanford HLW.[xiii] The potential for savings by applying currently available technology through retrofit of current facilities is staggering at about $2.4B. To put this in perspective, if even 10% of the implied savings was realized, it would be equivalent to full scholarships providing 20,000 college degrees. For an operating facility such as the DWPF, retrofit of the JHM with CCIM technology would be likely to allow for processing of challenging waste streams with higher waste loadings at a faster rate, while increasing the operational life of the melter. The technology offers the potential for higher waste loading and processing rate because it is particularly advantageous for operation at elevated temperatures (i.e. up to 2000^{o}C versus a maximum of 1150^{o}C for a JHM) and its design enhances convective mixing without reducing melter life. Due to its compact size and modular construction, the CCIM also offers simpler decommissioning. The CCIM has been cited in the referenced reports for potential benefits to the DOE in immobilization of HLW.

A variable frequency CCIM testbed has been built at INL for demonstration of the advantages of this technology. This system is fully instrumented and capable of both slurry and solid feeds. The offgas treatment system is also fully instrumented and designed for highly efficient treatment of the CCIM offgas. In addition, several test melts have been done in CCIM units at the Khlopin Radium Institute in St. Petersburg, Russia and the Radon facilities near Moscow, Russia in collaboration with those institutions and Savannah River National Laboratory.

Other technologies have also been evaluated. Hot iso-static pressing (HIPing) has been investigated in collaboration with the Australian Nuclear Science, and Technology Organization (ANSTO). Fluidized bed mineralization has been evaluated in collaboration with Studsvik (Sweden), Thermochem and Science Applications International Corporation (SAIC) (USA) at test facilities owned by INL. Steam reforming and HIPing also appear to be potentially advantageous technologies to produce cost-effective alternative waste forms, but they have not been investigated to determine their true feasibility for wide-spread implementation within the DOE. In the long-term, validation of these alternative technologies, development of quantitative operational data, and life-cycle cost-effectiveness analyses will be crucial to implementation of advanced fuel cycles in support of the next generation nuclear power plants.

Efforts to investigate alternative process technologies must be focused, based on the programmatic strategy, on only those approaches that provide clear, and significant benefit, while offering realistic opportunities for implementation into DOE facilities and systems. Significant expertise and capability has been developed in waste processing capabilities within the USDOE, international agencies (i.e. Commissariat a l'Energie Atomique (CEA) in France and ANSTO in Australia), and private/semi-private industry (e.g. AREVA/Cogema in France, Studsvik, and Thermochem). Collaborations with industry and international agencies should be strengthened and leveraged to realize the maximum benefit of the proposed approach. Feasibility studies for retrofit opportunities of existing facilities, as well as implementation in future facilities, require the support of private industry. The INL testbed was developed specifically for these efforts and is available for collaborative testing and evaluation.

WASTE FORM

Focus on a specific waste form like BSG limits the overall efficiency of the HLW disposal system. Borosilicate glasses are very durable and have the flexibility to immobilize many cations but are limited for some species as described above. Manufacturing BSG in joule-heated melters further limits the efficiency of the system due to constraints caused by the technology on glass properties such as melting and liquidus temperatures. By setting glass property constraints due to the processing technology, glass chemists are limited in their flexibility in designing durable glasses that can tolerate more waste.[xiv] Another factor that further complicates this situation is that treatment systems must not only address the wide variety of HLW streams at Hanford and Savannah River, but must also address sodium bearing waste and calcine waste at Idaho, which are dramatically different in chemical composition and physical form. Limiting the flexibility of the glass chemistry unnecessarily results in more glass, which translates directly into greater impact (cost) on a repository.[xv] Expanding the envelope of acceptable waste forms to include non-BSG, glass-ceramic, and ceramic matrices, in conjunction with implementing currently available technologies could significantly reduce the costs of waste immobilization.[xi]

The immobilization systems designed today can be expected to ultimately process wastes that will be generated in meeting the nation's future energy needs. Industry knows today that waste forms other than BSG are acceptable choices to immobilize key radioactive and hazardous components in existing and future waste streams. As large volume waste streams are addressed (e.g. Idaho calcines), it is imperative to match the waste form to the waste stream characteristics. As an example, Idaho calcines contain components that are difficult to incorporate in a BSG waste form. Collaboration amongst ANSTO, CEA and Cogema demonstrated that waste loadings exceeding 50% can be achieved using a cold crucible induction melter to generate a glass-ceramic waste form.[xvi] If calcine is mandated to be immobilized in a BSG, the resulting waste form is neither economical nor an optimally performing waste form. Requiring a single host matrix for a variety of wastes, some of which are nearly insoluble in the BSG matrix (e.g. phosphorous and sulfur) results in lower waste loading, that leads to greater waste volume, and, in turn, higher processing and operations costs at both the treatment and disposal facilities. These overall HLW disposal system inefficiencies cost more money and take more time. Higher waste processing and disposal efficiency can be realized by performing the engineering analyses and trade-studies necessary to select the most efficient methods for processing the full spectrum of wastes across the DOE complex.

Matrix chemistry may include multiple phase BSG, non-BSG (i.e. aluminosilicate glass, iron phosphate glass), glass-ceramics, and ceramics, as long as standards are met for durability and stability. Iron phosphate glasses are just one example of a proven waste form, that if included now based on current technical performance requirements, could realize near term benefits by the USDOE at the WTP currently under construction at Hanford. Experimental data shows the low-activity glass production could potentially be cut in half at WTP using iron phosphate glasses, without relaxing waste form durability requirements.[xvii,xviii]

Validation of the performance of these candidate waste matrices may require additional durability testing, as well as an understanding of the interaction of chemical species with the waste package and the geology that could result during long-term waste form degradation. These data will be necessary to feed into the modeling that the USDOE uses to predict behavior and transport of contaminants in the subsurface under the repository. Fortunately, the basis for most of this testing already exists. It is primarily *what* is measured that must change to evaluate other materials. For example, boron is measured to determine the durability of a BSG, whereas the appropriate analysis for an iron-phosphate glass may be iron. This flexibility to measure the appropriate constituents to evaluate alternative waste matrices must be built into the waste/package/disposal system performance requirements documents to allow consideration of these waste forms.

The work proposed here does not suggest a need to develop new waste forms or processing technologies, only to take advantage of the research and development already paid for in the nuclear community over the last 30 years.

SECONDARY WASTES AND OFFGAS TREATMENT

After waste form and manufacturing, the third leg of the stool is controlling gaseous and liquid emissions. Generating copious amounts of low-level waste can be a costly trade for enhanced processing technology. In the USA, this issue can be exacerbated by the regulations relative to "mixed-wastes" which contain both chemically hazardous and radioactive species. Some of these wastes, particularly those contaminated with transuranic elements may literally have no disposal options under current regulations.

To meet many of the research needs in this area, INL has equipped the CCIM testbed with a state-of-the-art modular offgas treatment system. The offgas system is designed to provide highly efficient thermal destruction of NOx and products of incomplete combustion, capture of acid gases, metals, radionuclides, and particles, and sorption of any residual mercury if necessary. The modular design allows the various unit operations to be mixed and matched to treat offgases from CCIM testing, a bench-scale steam reformer, and experimental offgas mixtures created for testing by using one of the thermal processes as an artificial source term with or without addition of containerized gases. The offgas system in the thermal testbed is also mirrored in a larger pilot-scale facility currently equipped with a fluidized-bed steam reformer. These offgas treatment systems are currently available to help resolve emission issues while minimizing secondary wastes like spent carbon. The system is also designed to minimize offgas component size by minimizing offgas flow through design innovations. One example, is the simple heated offgas connection to the melter plenum that eliminates components like the film-cooler commonly used to reduce plugging immediately downstream of a melter plenum.

CONCLUSIONS

The nuclear industry knows and understands the waste issues, and must accept the challenge of deploying new technology, or it is doomed to repeat the past. More efficient waste forms can be used now to reduce the volume of stabilized HLW by as much as 50%. The technology to produce these waste forms is available today, and requires little or no additional development. The CCIM is an excellent example of a technology that has over a decade of commercial nonradioactive operating time that could be immediately integrated into existing facilities. The CCIM can reduce or eliminate melter failure, potentially double processing rates and take advantage of more efficient waste forms, thus incorporating more waste in less volume. Offgas treatment and secondary waste reduction for all processes, and particularly high-temperature systems are areas of significant interest to the public. Currently available innovations can improve the designs of future facilities, reduce treatment system size and cost, and potentially improve operation of current facilities. The integrated thermal test bed at INL is available to demonstrate better technology, available now, to save the nuclear community billions of dollars in closing the waste side of the fuel cycle.

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