R&D Programs and Policy within the CEA-AREVA Joint Vitrification Lab (LCV) - 13592

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ABSTRACT

Waste management is a key issue for the reprocessing industry; furthermore, vitrification is considered as the reference for nuclear waste management. In order to further improve and strengthen their historical cooperation in high temperature waste management, the CEA, R&D organization, and AREVA, Industrial Operator, decided, in September 2010, to create a Joint Vitrification Laboratory within the framework of a strategic partnership. The main objectives of the CEA-AREVA Joint Vitrification Laboratory (LCV) are (i) support AREVA's activities, notably in its La Hague plants and for new projects, (ii) strengthen the CEA's lead as a reference laboratory in the field of waste conditioning. The LCV is mandated to provide strong, innovative solutions through the performance of R&D on processes and materials for vitrification, fusion and incineration, for high, intermediate and low level waste. The activities carried out in the LCV include academic research on containment matrices (formulation, long-term behaviour), and the improvement of current technologies/development of new ones in lab-scale to full-scale pilot facilities, in non-radioactive and radioactive conditions, including modelling and experimental tools. This paper focuses on the programs and policy managed within the LCV, as well as the means employed by the CEA and AREVA to meet common short-,mid- and long-term challenges, from a scientific and industrial point of view. Among other things, we discuss the technical support provided for the La Hague vitrification facilities on hot melter and CCIM technologies, the start-up of new processes (decommissioning effluents, UMo FP) with CCIM, the preparation of future processes by means of an assessment of new technologies and containment matrices (improved glasses, ceramics, etc.), as well as incineration/vitrification for organic and metallic mixed waste or metallic fusion. The close relationship between the R&D teams and industrial operators enables the LCV to propose attractive waste management solutions, with appropriate schedules and optimized development costs, making allowance for R&D constraints, engineering requirements and the industrial environment.

INTRODUCTION

Vitrification is now recognised internationally as the reference industrial solution for managing high level waste, and is therefore key to the industrial and commercial sustainability of spent fuel recycling. As demonstrated for several decades, mastering this technology and adapting it permanently to new market challenges and changing safety requirements requires extensive scientific, technological and industrial expertise in all aspects of vitrification.
Industrial startup of the CCIM (Cold Crucible Induction Melter) technology was a particularly major step in that direction, and was only possible thanks to the close cooperation between the CEA and AREVA [1]. Leveraging this historical partnership to bring it to the next level, the

Joint Vitrification Laboratory (LCV) will not only support industrial spent fuel recycling, but will also aim to develop new high temperature waste processing technologies for the international market.

The industrial challenges for vitrification specialists are to i) enhance the performance of current processes ii) acquire technological leadership for future facilities iii) develop and qualify new glasses and containment matrices and iv) make R&D competencies available to operators around the world.
The R&D work being carried out by the LCV to meet these challenges has resulted in a strengthening of basic research into the materials used for waste packages and their long-term behavior, with the aim of proposing innovative solutions and adding to existing knowledge.

The LCV boasts wide range of experimental equipment, from laboratory-scale to full-scale, which can be used in both radioactive and non-radioactive conditions. Dimensional extrapolation is almost impossible at present due to the very high non-linearity and complex coupling of the physical and chemical phenomena involved, and as a result, extensive full-scale experiments are necessary at the process qualification stage.
However, the ever-larger and more complex installations required and the high cost of the simulants needed for testing is prompting researchers to opt increasingly for simulation. This gives them a better understanding of the tests to be carried out and helps to optimize the corresponding costs.
While high level waste vitrification accounts for a large share of the R&D carried out at LCV, solutions also have to be developed for packaging intermediate level long-lived waste generated by fuel cycle activities and nuclear facility dismantling. Examples of this are mixed organic, mineral and metallic waste, structural waste from nuclear fuel treatment and long-lived radionuclides (iodine, chlorine, etc.).
The combination of scientific, industrial and engineering skills produces the high-level expertise needed to enhance the performance of facilities currently in operation.

EXPERIMENTAL RESOURCES

A multi-scale formulation and characterization approach is required in the laboratory to develop a full waste treatment vitrification process; this also includes studies of radioactive laboratory glasses and the use of mock-ups and pilot industrial facilities to develop containment matrix production processes.

Laboratory resources

In the laboratory, around 35 researchers work on matrix formulation studies, basic vitreous material studies and industrial support. The experimental resources available make it possible to create samples weighing anything between one gram and one kilogram in temperatures of up to 1600°C. This is done not only in conventional furnaces but also in induction furnaces, thermal gradient furnaces, controlled atmosphere furnaces and by means of presses. The associated characterization lab has all the conventional resources needed to measure the physical properties of molten and solid glass (viscosity, thermal conductivity, electrical resistivity, laser particle size analyzers, pycnometers, density meters, IR spectrometers, DSC, TG-DTA, etc.) and perform structural characterization (SEM with heating stage, field effect SEM, variable temperature X-ray diffraction, etc.). A considerable amount of time is devoted to basic studies of the physical and structural properties of glasses and the physical and chemical mechanisms by which they are created. Further information is obtained through modeling, making the researchers able to answer operators' immediate questions or anticipate future developments in radioactive waste vitrification (new glasses, improved processes).

For high level waste, 25 LCV researchers operate two shielded systems, a liquid waste analysis laboratory and a solid waste characterization laboratory. Using this equipment, they can produce radioactive materials weighing a few hundred grams via the "pot" process (glass containing Pu, Cm, Tc, specific containment ceramics, etc.) and the continuous process (fission product and minor actinide glass) reflecting the 2-stage calcination/vitrification process used on an industrial scale in France. The glasses produced can be prepared (cut, bored, encapsulated, polished) for analysis in the two support laboratories where solids and liquids are analyzed. Techniques such as SEM, EPMA, Raman spectroscopy, gamma scanning and X-ray diffraction are used. The glasses studied can also undergo weathering experiments with water in static or dynamic conditions, after which chemical analysis of the leachates is carried out.

Approximately 30 researchers spend their time studying the long-term behavior of glasses or new containment matrices, particularly the way in which they are affected by water in repository conditions. In this discipline, the timescales involved rule out any form of experiment and robust predictions can only be made by demonstrating an understanding of the innermost mechanisms by which water weathers glass, and modeling them. Original water experiments to weather glass (in static, dynamic and repository conditions) are carried out to determine the durability of the matrices. Operational models are produced to predict the performance levels over time in a variety of conditions.

Mock-ups

Around 30 researchers specializing in high-temperature process engineering develop and operate specific tools such as mock-ups for studying calcination and melting processes and the recycling of entrained matter in the off-gas treatment unit. From what they learn, they can develop chemical models of process behavior and industrial-scale pilot installations representative of industrial facilities.

Experiments are carried out on the 1:200 calcination mock-up to determine the calcination reactions of the glass and study heat transfer over the entire tube length.

The dust recycler mock-up was designed to represent the flows and volumes involved on a scale of 1:50 in relation to the full-scale dust recycler. It can be connected to a furnace mock-up to generate a gas flow representative of a hot melter. The furnace mock-up can be fed with solids and liquids to study glass production reaction mechanisms.

The 1:10 DIVA calcination facility comprises a calciner and a melting pot. It is used to conduct parametric studies during the two stages of the calcination-vitrification process, mainly to support operation of the vitrification facilities in La Hague and Marcoule.

The DIVA In-Can facility is a vitrification unit into which the effluents to be vitrified are fed directly. It produces glass at a rate of 5 kg/h in a metal melting pot, with no possibility of producing a glass melt. It is used to vitrify small quantities of effluents.

Pilot facilities and prototypes

Around 30 mechanical, electrical and I&C engineers and technicians are responsible for round-the-clock operation of industrial-scale pilot facilities and prototypes, as well as for operational maintenance.

The Evolving Vitrification Prototype (PEV) comprises a calciner and a melter which can be either a cold crucible or a hot metal pot. It is used for R&D programs carried out to support operation of the vitrification facilities in La Hague or any other export programs based on a 2-stage calcination-vitrification process.

The CFA platform is a calciner-free facility for carrying out continuous experiments in cold crucibles of varying diameters, both conventional and manufactured by Megaver, with induction heating through the bottom of the crucible. Programs based on a vitrification process with liquid feed directly into the cold crucible, such as the DoE programs in the United States, and programs to develop an optimized vitrification process for future reprocessing plants will be run on this platform [2].

The platforms, pilot facilities and mock-ups are operated in conjunction with a Laser Induced Breakdown Spectroscopy (LIBS) chemical analyzer which analyzes solid and liquid samples in near real-time. Adjustments are currently being made to enable on-line analysis of processes to study transient phenomena which often generate additional entrainment, and identify safe interim solutions.

The vitrification facilities are supplemented by waste treatment facilities involving high-temperature processes such as combustible waste incineration and the melting of metal.

The Solid Waste Incineration Research Facility (IRIS) comprises a pyrolyzer for solid organic waste and a calciner for obtaining ash. The equipment has its own off-gas treatment unit comprising a post-combustion chamber, a dilution cooler, an electrostatic precipitator and a scrubbing column. This facility is used to carry out R&D into the incineration of combustible waste.

An in-can plasma incineration-vitrification mock-up is currently being installed and will share the off-gas treatment unit of the IRIS facility. It will comprise a melter for matter with a metallic phase and a vitreous phase which may be partly crystallized, and a plasma incineration unit capable of operating with gases with a high oxygen content. This mock-up will be used to obtain design data for a treatment process for technological waste containing a mixture of mineral, organic and metallic material.

Modeling tools

The researchers have 3D modeling tools to support their process development and design work.

Electromagnetic simulation is carried out in 3D with the Flux 3D® application to optimize existing facilities and devise, design and build new induction heating facilities. Complex geometries can be handled and important aspects, such as the amount of electromagnetic energy to be supplied at precise points, can be calculated. The Flux 3D® application is used intensively, for hot or cold crucible induction heating facilities or for new applications which are still at the development stage.

This electromagnetic code is coupled with the FLUENT® code for thermal-hydraulic studies in molten glass baths. The algorithms have been adapted to make allowance for the large variations in the physical and chemical properties of molten baths as a function of temperature. The two applications are coupled by exchanging files. The challenge for melter modeling in the future is to make allowance for the chemical reactions and changes in physical and chemical properties in the thickness of the cold layer when the glass is being produced. Coupling to the electromagnetic-thermal-hydraulic model of the melter to understand the thermal impact on both sides will be an important step in the overall modeling process.

The close working relationship between the R&D teams and industry also makes it possible to obtain operating data on industrial facilities which are fundamental sources of information when carrying out research programs. More data samples are available and once they have been interpreted by the R&D teams they can be compared with R&D figures, taking knowledge to a higher level.

MAIN R&D PROGRAMS

R&D teams supporting industry

The support the R&D teams provide for industrial operators makes it possible to (i) enhance the performance levels of existing processes (ii) assist operators when new processes are being commissioned and ramped up (iii) suggest ways in which the changing characteristics of waste to be treated should be taken into account.

The close relationship between the R&D teams and the industrial operator is a major advantage when seeking ways to enhance the performance levels of existing processes.
Furthermore, it gives the R&D teams the opportunity to encounter operating problems and constraints first-hand, including those which cannot be reproduced at the R&D stage. Consequently, the recommendations of the R&D teams can be adapted, leading to the best compromises in the interests of continuous improvement (end product quality, increased production rates, optimization of parameters to protect the industrial tool, equipment life extension, etc.).

The support the R&D teams provide for operators is also essential for the successful commissioning of new processes. Once a development plan has been completed, including formulation of the containment material, and the process developed and qualified on laboratory-scale to full-scale facilities in nominal conditions and in transient and degraded modes, we are in a position to provide a process book and all the data nuclear operators require to put together the document reference base needed to obtain Nuclear Safety Authority licenses and write the operating and maintenance procedures.
One of the stages in the qualification process involves carrying out a series of tests in conditions which represent as accurately as possible the first test campaign to be conducted in radioactive conditions in the industrial facility [3]. This enables the R&D teams to provide the support needed when the new process is commissioned. During this phase, the R&D teams and the operator work together as one to ensure that start-up is successful and that nominal operation is

reached as quickly as possible, with operating parameters being adjusted in near real-time. The R&D specialists also provide training for operators and maintenance workers.
The successful hot startup of the R7 vitrification facility at AREVA's La Hague plant, where rinsing effluents from the UP400 plant are treated and UMo high-level liquid waste is packaged, is the perfect example of the value added provided by LCV.

Lastly, the R&D teams help operators to adapt their operating conditions to the changes in the types of waste to be treated, giving them greater production flexibility. One example of this is the allowance made for the stream from the treatment of RTR UAl fuel in the UOx stream.

Resources other than those in the LCV are also available, for example the test platform of the CCIM in the Beaumont-Hague Research Hall (HRB) near the La Hague site and operated by AREVA's engineering company; this is used to qualify certain operating configurations, notably in remote operating and maintenance conditions.

Developments in the vitrification of HLW concentrated fission product solutions

The solutions resulting from the processing of light water reactor fuels are successfully vitrified in the La Hague facilities. Once a cold crucible has been started up successfully, it should be possible to vitrify them in this kind of furnace while seeking to increase production capacity and platinoid content. The maximum content in glasses for fission product, actinide and noble metal oxides is currently limited to 18.5 wt%. A 10-year study program has been devised with the aim of reducing glass storage volumes and determining the impact of an increased incorporation rate on the glass containment properties; glass matrices with higher aluminum and sodium contents have been developed. The possibility of using chemical dilution to reprocess UAl and USi fuels from research reactors is also being examined.

The impact on the glass structure and hence on the thermal stability and self-irradiation of these glasses in the long and medium term are the key points of the program.

The purpose of the self-irradiation studies is to determine whether the glass properties will be modified by the cumulative decay which occurs while they are in the repository.
To this end, several complementary lines of research are being carried out at the LCV. They consist mainly of specific laboratory-scale experiments to study the aging of nuclear glass in repository conditions over a period of around one year, as well as atomistic simulations to determine the atomic scale mechanisms behind the phenomena observed. All of the information obtained is used to create robust long-term behavior prediction models.

For the study of glasses doped with radioactive elements, radioactive elements with a short half-life are incorporated into the glass in order to obtain the highest cumulative decay doses possible in the time devoted to the laboratory studies. The advantage of this methodology is that it is the most representative since the entire glass volume is irradiated as it would be in real conditions and all decay phenomena are taken into account. For example, for the study of alpha decay, this technique simulates both the consequences of the alpha particle (helium nucleus) and the recoil nucleus.

However, the disadvantage is that radioactive samples are used, limiting the characterization techniques to those which are adapted to this kind of environment. Furthermore, given that the decay doses are integrated into the laboratory glasses more quickly that in actual repository conditions, the dose rate also has to be examined. This is done by creating materials doped to varying degrees so that irradiation effects at different dose rates can be investigated.

For the study of real radioactive glasses, radioactive laboratory glasses with a composition comparable to that of the real glass are produced; industrial radioactive glasses are also used. Even though accelerated aging is not possible with this technique, it has the advantage of representing actual situations and enabling important observations to be made on a human scale (10 to 30 years). In fact, almost half of the energy released by beta and gamma decay is released in the first 30 years.

External irradiation of the glasses is the third aspect studied using simulated non-radioactive glasses in which irradiation loading is simulated by means of external irradiation techniques (neutrons, heavy ions, electrons, γ). These fairly easy experiments shed light on the macroscopic and microscopic changes which occur in the material. By characterizing the glasses using sophisticated spectroscopic techniques, experiments can be carried out to determine how the atomic structure of the glass behaves under irradiation. Moreover, by varying the type of irradiation, different decay types can be simulated and the electronic and nuclear effects differentiated, which is necessary to determine the cause of the phenomena observed. The disruptive effects resulting from the high injected dose rates and the low irradiated volumes are the major disadvantages of this kind of experiment. By having a precise knowledge of these two factors and comparing them with the results of the other lines of research, the pertinent effects of experimental artefacts can be determined.

Atomistic modeling of glass self-irradiation

Considerable progress has been made in the last few years as regards computing capability and the phenomena occurring on an atomic scale during self-irradiation in glass can now be simulated numerically. Until now, dynamic molecular studies were most commonly used to study the ballistic (or nuclear) effects of the recoil nuclei emitted during alpha decay. This approach takes advantage of the fact that the recoil nucleus trajectory is fairly short, primary and secondary projectile energy decay is rapid and most of the energy decay mechanisms are elastic. Thus it is possible to simulate the trajectory of a recoil nucleus and all the dynamic phenomena it produces in its wake in modeled vitreous systems comprising a few tens to a few hundred thousand atoms. This gives an atomic-scale view of the ability of the glass matrix to withstand radiation.

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When combined, all these different lines of study make it possible to assess the impact of self-irradiation of the glass on its macroscopic behavior and therefore to judge its radioactive element containment capability in the long term; the atomic origin of the phenomena observed can also be determined, information which is essential for creating long-term behavior prediction models.

Treatment of mixed technological waste

The research program for the treatment of mixed metallic, organic and mineral waste has been designed to make allowance for the constraints resulting from different kinds of waste with varied compositions.
The process envisaged is based on an incineration-vitrification process involving a combination of proven and partially validated technologies. It consists in incinerating the organic fraction of the waste using plasma torches and incorporating the ashes obtained into the mineral waste in a containment glass. The metal fraction will be melted down. The confined waste package will comprise two fractions and further studies will need to be carried out to determine how they interact during the production phase.
The first thing to be done to finalize this process will be to carry out preliminary R&D in order to formulate the containment glass.
Preliminary tests on the combustion of mixed waste and the phosphate treatment of metals likely to form volatile chlorides will also be carried out on existing facilities to remove any uncertainty in these two areas. Likewise, mock-ups will be used to assess the preliminary glass compositions selected.
At the end of this phase, a number of process and containment glass options will be proposed.
The second stage of the program consists in testing these containment glasses on a laboratory scale (sensitivity to waste, glass/metal interaction) and validating the process and containment glass options.
The next stage will be to start designing and building a full-scale industrial prototype which can be adapted to the nuclear industry.
A methodology for studying the long-term behavior of the both vitreous and metallic containment materials in the package will have to be drawn up.
The third phase of the R&D program covers full-scale testing on the industrial prototype, process qualification, characterization of the containment materials produced and investigation into the long-term behavior of the package as a whole. At the end of this phase, a process book will be proposed.

CONCLUSION

The R&D policy rolled out by the CEA and AREVA and materialized by the creation of the LCV enables the two entities to work together on the short-, medium- and long-term challenges of high-temperature waste treatment and vitrification.
This policy is designed to i) reflect a shared vision of the long-term and the research needed (materials, processes, technologies, waste package durability) to meet the requirements of the nuclear industry ii) maintain and develop skills to ensure scientific excellence and the long-term future of a successful industry iii) ensure continuous improvement in terms of industrial performance and end-product quality.

The R&D programs in progress at the LCV are concrete proof of the desire to put this policy into action, with timescales which are compatible with the challenges facing the CEA and AREVA.

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