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MCCLOY, John, RILEY, Brian, DIXON WILKINS, M., EVARTS, Jonathan, BUSSEY, John, VIENNA, John, BINGHAM, Paul <http://orcid.org/0000-0001-6017-0798>, GREGG, Dan, OJOVAN, Michael, SCHULLER, Sophie, URUGA, Kazuyoshi, PERRET, Damien, REGNIER, Elise, GIBOIRE, Isabelle, UM, Wooyong, XU, Kai, GOEL, Ashutosh and KRUGER, Albert

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This document is the Accepted Version [AM]

Citation:

MCCLOY, John, RILEY, Brian, DIXON WILKINS, M., EVARTS, Jonathan, BUSSEY, John, VIENNA, John, BINGHAM, Paul, GREGG, Dan, OJOVAN, Michael, SCHULLER, Sophie, URUGA, Kazuyoshi, PERRET, Damien, REGNIER, Elise, GIBOIRE, Isabelle, UM, Wooyong, XU, Kai, GOEL, Ashutosh and KRUGER, Albert (2024). International Perspectives on Glass Waste Form Development for Low-Level and Intermediate-Level Radioactive Waste. Materials Today, 80, 594-618. [Article]

International Perspectives on Glass Waste Form Development for Low-Level and Intermediate-Level Radioactive Waste

John S. McCloy* [1],[2], Brian J. Riley [2], M. C. Dixon Wilkins [1], Jonathan Evarts [1],[2], John Bussey [1], John Vienna [2], Paul A. Bingham [3], Dan Gregg [4], Michael Ojovan [5], Sophie Schuller [6], Kazuyoshi Uruga [7], Damien Perret [6], Elise Regnier [6], Isabelle Giboire [6], Wooyong Um [2],[8], Kai Xu [9], Ashutosh Goel [10], Albert Kruger [11]

[1] Washington State University, Pullman, WA, USA
[2] Pacific Northwest National Laboratory, Richland, WA, USA
[3] Sheffield Hallam University, Sheffield, UK
[4] Australian Nuclear Science and Technology Organisation, Kirrawee, Australia
[5] The University of Sheffield, Sheffield, UK
[6] CEA/DES/ISEC/DPME, Montpellier University, Marcoule, France
[7] Central Research Institute of Electric Power Industry, Yokosuka, Japan
[8] Pohang University of Science & Technology, Pohang, Korea
[9] Wuhan University of Technology, Wuhan, China
[10] Rutgers, the State University of New Jersey, Piscataway, NJ, USA
[11] United States Department of Energy, Richland, WA USA

Keywords: nuclear waste; glass; waste processing; international perspectives

*corresponding author, john.mccloy@wsu.edu

Abstract

The global energy transition to low-carbon energy sources will require a significant contribution of nuclear energy to achieve emission goals. Low-level radioactive wastes (LLW) and intermediate-level radioactive wastes (ILW) are created in various phases in the nuclear fuel cycle for power generation, as well as from nuclear accidents, legacy weapons production, contaminated site decommissioning, and other nuclear fuel cycle activities such as radiopharmaceutical production. In this review, we will summarize recent developments, state-of-the-art glass formulations, and thermal treatment process developments for vitrification of nuclear LLW and ILW from programs in Europe, Asia, Australia, and North America. Throughout, we will discuss the selection of glass over other possible waste forms and any special processing considerations due to the nature of the waste. The characteristics of the wastes, such as mixed technological waste, waste coming from dismantling of reprocessing facilities, site decommissioning, accident site decontamination, are important considerations. This is balanced with the suite of technologies available to vitrify these wastes, such as variations of incineration, in-can melting, and plasma treatment. Glass properties and microstructural aspects – such as inclusion of crystals or metallic phases – are compared to give an overview of the versatility of packaging matrices, such as homogeneous glasses, composites, and crystalline matrices. The volume and heterogeneity of the waste, as well as radionuclide, chemical and low silicate solubility components, factor into the selection of a given waste form, processing route, and technology. Case studies include examples from the United States, United Kingdom, Russia, France, Australia, Japan, Korea, and China.

1 Introduction

In recent years, it has become widely recognized that the global goals of decarbonization will not be met without a significant role for nuclear energy in electricity generation, of which the power supplied by ~450 commercial nuclear reactors provide ~30% of the world's total low carbon electricity [1]. That said, the public acceptance of nuclear energy is uneven between countries, often due to concerns about the disposal of radioactive waste and the public uncertainty of available policies and technology for the safe handling and disposition of such wastes [2]. It is the purpose of this review to summarize the current state-of-the-art relating to the disposal of low- and intermediate-level radioactive wastes (LILW) in various parts of the international community. Rather than summarizing all waste form types, we focus on glass and the vitrification processes used to produce them, while only briefly discussing alternative waste form technologies. Glass waste forms have long been considered as some of the safest and most chemically durable waste form classes, thus allowing for very long-term immobilization (thousands to millions of years) of radionuclides and preventing their entry into the biosphere [3]. This review will also avoid discussion of used nuclear fuel or reprocessed nuclear fuel leading to high-level radioactive waste (HLW), as other suitable recent reviews on this subject are available [4-13]. One of the motivations of the current review is to consolidate information from the international community from disparate sources, often including a large amount of 'grey literature' [14] available in specialist conference proceedings, and company and laboratory reports. The public message to be told is that there exists a robust, science-based, and long-standing international engineering experience for treating and immobilizing a large variety of radioactive wastes safely and effectively. Materials science and engineering is a key discipline threaded throughout radioactive waste management. A high-level of international collaboration exists in this field as evidenced by this paper and the cited references.

Previous reviews have focused on general considerations for radioactive waste management [15-18], specific strategies for individual countries [19-21], or overview of different waste form types [22, 23].

Additionally, there is some overlap between vitrification considerations for LILW and that of hazardous non-radioactive waste [24-27].

In the following sections, we focus our attention on technological considerations, which lead to (1) vitrified (glassy) waste forms for immobilization of LILW, (2) the treatment and conditioning processes required prior to thermal treatment of waste and (3) the safety systems needed to manage any gaseous emissions from the thermal processing. Other potential or currently used waste forms and strategies for LILW, including cements, bitumen, and direct disposal, have been reviewed elsewhere [23, 28]. The basic requirements for a long-term immobilization matrix for radioactive waste are its radiation stability and chemical durability against the environment when stored in an appropriate disposal site [28-30]. Additionally, for vitrification of wastes, processing constraints must be considered due to potential solid phase precipitation, volatility, melt viscosity, and electrical conductivity [25, 27, 31].

In the current review, we first summarize the considerations for classification of radioactive waste, by hazard. Next, we summarize the state of the field by looking at the considerations for designing vitrified waste forms for the specific waste and available technologies, including consideration of off-gas treatment. Here, we also highlight the important role of glass formulation along with the historic and current methods for performing this part of the design. The bulk of the review is dedicated to case studies of various countries working on technology development and implementation of vitrified waste forms for LILW. Finally, we summarize and conclude with recommendations for areas of targeted future research for the international community.

2 Radioactive Waste Classification

2.1 Radioactive waste classification and regulation

Waste form design, and the associated technology, is dependent upon the waste disposition strategy. Thus, it is necessary to develop concise waste classification prior to specifying material requirements for the safe long-term management of radioactive wastes. In most cases, it is appropriate to classify wastes according to the requirements of the necessary disposal safety strategy, with the knowledge that any one strategy will not be feasible for all wastes. For the context of this work, an understanding of how material is classified as ILW or LLW is important. Waste classifications depend on laws specific to various countries; however, most national nuclear waste classifications systems have at their core considerations for the radioactive characteristics of the wastes.

Different characteristics of a radioactive waste under consideration must be examined to determine its final classification. The most obvious criterion is the radioactivity of the waste, which includes a holistic assessment of specific activity (activity per unit mass), activity concentration (activity per unit volume), and total activity of the waste. The half-lives of the contained radioisotopes relative to the timescale of institutional control and management will also impact the waste classification, with particularly long-lived isotopes [*e.g.*, the fission products ^{129}I ($t_{1/2}$ = 1.57×10^{7} y), ^{99}Tc ($t_{1/2}$ = 2.13×10^{5} y), and ^{93}Zr ($t_{1/2}$ = 1.53×10^{6} y)] posing significantly different challenges to disposal than short-lived isotopes, which are more common in wastes from nuclear medicine, *e.g.*, ^{192}Ir ($t_{1/2}$ = 73.83 days), ^{60}Co ($t_{1/2}$ = 5.271 y). Some wastes containing high concentrations of 'medium-lived' isotopes [chiefly the high-yield fission products ^{90}Sr ($t_{1/2}$ = 29.1 y) and ^{137}Cs ($t_{1/2}$ = 30.17 y)] will generate significant radiogenic heat, possibly necessitating specific engineered solutions during management and disposal [32, 33].

2.2 International recommendations

The International Atomic Energy Agency (IAEA) recommends dividing radioactive wastes into six classifications (**Figure 1**) according to the type(s) of radionuclides present in the waste and their concentrations [34]. These classifications are general and written in the context of the geological disposal of wastes; further development of the classifications and any quantitative boundaries between them is left to the individual IAEA member states and may be state, agency, site, or even process specific.

1. Exempt waste (EW): material that contains sufficiently low concentrations of radionuclides that it does not need to be disposed of or managed as radioactive waste after clearance (i.e., release from regulatory control [28]).

2. Very short-lived waste (VSLW): material that will fall below regulatory standards for radioactive waste after a limited storage period of up to a few years.
3. Very low-level waste (VLLW): material that, although not meeting regulations for EW, does not require containment or isolation, and so can be disposed of in near-surface landfill facilities.
4. Low-level waste (LLW): material containing limited amounts of long-lived radionuclides that will remain above clearance levels for an extended period, up to a few hundred years. These are wastes that require engineered isolation and containment, though are suitable for disposal in near-surface facilities. It is noted that this class comprises a very wide range of wastes. In some countries, there is considerable complexity to this classification, as described further below.
5. Intermediate-level waste (ILW): material that contains significant amounts of long-lived radionuclides but does not generate significant radiogenic heat. The amount and type of radionuclides in ILW will not decay in reasonable timescales to levels suitable for near-surface storage and so will require disposal in deeper facilities than LLW.
6. High-level waste (HLW): material that contains sufficiently high concentrations of radionuclides as to be significantly heat generating on disposal timescales. Waste containing high concentrations of long-lived radionuclides where deep geological disposal is recommended.

2.3 Example implementations, relation to disposal depth

Many nations base their regulations on the IAEA waste classifications detailed above, including China [35], South Korea [36], Russia [37, 38], Australia [39], and the United Kingdom (UK) [33, 40]. Aside from the quantitative boundaries dividing the categories, other differences also exist. For example, the omission of ILW as a separate category in Japanese regulations, with HLW and LLW designated the only top-level classifications, and LLW subdivided according to isotopic contents and specific activity/activity concentration [41].

In many nations, wastes that are predominantly α-emitters are treated differently from those that are predominantly β- or γ-emitters or materials containing transuranic materials; for example, in Russia, solid materials are designated as LLW if their specific activity is 0.1 – 1 MBq/kg, 1 – 10 MBq/kg, and 0.01 – 0.1 MBq/kg for α emitters, β-/γ-emitters and transuranic-containing wastes respectively [37].

Each country can classify and regulate radioactive waste as they see fit, with or without guidance from IAEA. For example, in the US, the US Nuclear Regulatory Commission (NRC) defines low-level radioactive waste (LLRW) as a regulatory category through the 1954 Atomic Energy Act. Regulatory categories of commercial LLRW/LLW in the US are defined by the NRC in the US Code of Federal Regulations, part 61 "Licensing requirements for land disposal of radioactive waste" where waste classification is in 61.55 [32] defining different classes (A, B, C, greater than C) radioactive waste based on specific radionuclides and concentrations. As compared to commercially produced LLW, US Department of Energy LLW is governed by specific DOE orders [42]. The US Environmental Protection Agency (EPA) [43] further defines categories, without different regulatory statuses, in the EPA 2003 Advanced Notice of Proposed Rulemaking (ANPR), such as low-activity radioactive waste (LARW) and low-activity mixed waste (LAMW), the latter which contains small amounts of radionuclides plus chemically hazardous components.

In a departure from IAEAs classifications, the US defines HLW as "…the highly radioactive material resulting from the reprocessing of spent nuclear fuel…" [44] and other material designated as HLW by the US NRC, which must be disposed in a deep geological repository. All other wastes are designated as transuranic wastes, byproduct materials, or LLW. This source-based definition of HLW leads to significant differences in the classification of LLW. This definition has since been interpreted by the Department of Energy (DOE) to mean that some lower-activity reprocessing wastes can be designated as LLW, so long as they fall within the definition for Class C LLW, or it is demonstrated that they do not require disposal in a deep geological repository [45, 46]. Current regulatory controls in India also follow a similar source-based definition of HLW ("…the radioactive liquid containing most of the fission products and actinides present in spent fuel…" and other wastes with similar radiological characteristics [47]), with Low and Intermediate Level Waste (LILW) defined as wastes with activities and heat generation below HLW but above clearance levels.

Another case illustrating national departures from IAEA recommendations is the nuclear waste classification of Russian Federation [48], which generally follows IAEA recommendations and guidelines concerning final end point, apart from separation of nuclear waste into special waste (disposed of at its location) and disposable nuclear waste, which is then categorized into six classes according to waste type and final end point [49].

The IAEA acknowledges that different final disposal facilities are suitable for different radioactive wastes. While disposal in a deep facility is currently the option with the highest margin for long-term safety, it is untenable, for both financial and engineering reasons, for the high volumes associated with LILW. Most countries (and the IAEA) accept that final disposal in facilities not more than 100 m deep, with or without engineered barriers, is suitable for the majority of LLW.

Internationally, consistent features of LILW category wastes including the following: relatively lower specific activities/activity concentrations (usually on the order of ≤ 10 MBq/kg), no significant generation of radiogenic heat or no need for heat dissipation, high overall waste volumes, and compatibility with surface or near-surface disposal (< 100 m deep).

3 Technologies and considerations

This review focuses on LILW immobilized in glassy waste forms. Borosilicate glass was selected in the 1970s as the preferred matrix for immobilization of HLW in most countries [3, 42], due to the radiation stability, chemical stability, flexibility of the amorphous structure to accommodate varying compositions, and maturity of the industrial process. It is not necessarily obvious, however, why glass would be the preferred waste form for LILW, though cost and risk are the main factors involved in selecting a class of waste form for LILW.

Two examples follow. At the Fukushima Daiichi Nuclear Accident site in Japan, the overarching waste management concept is built around the need to treat large amounts of waste in an easy, reliable, fast, and affordable method [50]. In this schema, cementitious waste forms have priority for low to moderate concentrations of α-emitters, β-emitters, and γ-ray emitters, and long-lived nuclides. If cements do not meet requirements due to leachability, hydrogen generation, or other considerations, then alkali-activated materials (AAM, i.e., geopolymers) will be adopted. If AAM materials do not meet the needs, then glass will be considered, as it is fast and flexible and adoptable for many wastes but requires additional considerations for volatile components with off-gas processing. This strategy is depicted in **Figure 2**.

In another example, Hanford low-activity waste (LAW) has significantly higher volume than the HLW. While the initial plans focused on vitrification in Joule-heated ceramic melters, concerns about cost led the US Congress to request the Government Accountability Office conduct a study looking at alternatives to glass for Hanford LAW [51]. This study produced a series of reports assessing cost and risk of alternative technologies, including cement (grout), bulk vitrification in-container vitrification (ICV, see section 3.2), and fluidized bed steam reforming (FBSR) [51]. The grouting process includes use of ordinary Portland cement as well as often other supplemental cementitious materials of all types including blast furnace slag and fly ash [52]. The latter process involves pyrolyzing the liquid waste and converting it to crystalline phases, which can then be consolidated and bound in various ways. Other technologies for LAW and secondary wastes have been considered, such as geopolymers (DuraLith) [53], phosphate-bonded cements (Ceramicrete) [54], and large-batch vitrification [55]. In terms of these three options, glass is generally considered to be the most expensive but also the most chemically stable. However, at many nuclear sites in the US, liquid LLW is grouted, such as at the West Valley Demonstration Project (WVDP) [56, 57], the Savannah River Site (SRS) (*i.e.*, saltstone produced at the Salt Waste Processing Facility [58, 59]), or the grouted incinerator ash produced at Idaho National Engineering and Environmental Laboratory (INEEL) [60].

3.1 Forms of Waste

One key factor in selecting the waste form and the pertinent process, assuming vitrification in this case, is the form of the waste – i.e., liquid or solid, organic or inorganic, metal containing, mixed, etc. The primary waste streams for LILW can vary widely depending on the source, such as nuclear fuel reprocessing and fuel cycle, radiological operations, medical waste, nuclear site decommissioning, or accident and contamination. Thus, the primary waste could be soil, trees, buildings, needles, plastic gloves, liquids, gases, and everything in between. The form(s) and size(s) of the waste(s) can vary widely. In principle, processes are available to ultimately vitrify any of these materials, though vitrification may or may not be the overall best process for a given waste stream.

Generally, the processing of waste prior to the immobilization step, which for glasses is high-temperature thermal treatment, is described as ‘treatment’ and/or ‘conditioning’. In some literature, these are distinguished whereby ‘treatment’ implies a change for handling and economic transportation, while ‘conditioning’ also assumes a change to facilitate storage and disposal [61]. One of the critical factors for waste management is volume reduction. This is often done as a conditioning step, and can involve thermal processes such as incineration, pyrolization, plasma treatments, or non-thermal treatments such as compaction.

For example, liquid inorganic waste, which is mostly either acidic or alkaline aqueous heterogeneous mixture, is dehydrated by either calcination at elevated temperatures (400 to 1000°C) or dried at low-temperatures (90 to 120°C) and is stored as a slightly more stable form. Even these processes require management of the off-gas, as toxic and radioactive gases can be evolved and must be captured and treated – more so with calcination. Often multiple steps are integrated together in technologies where, for example, pyrolization, melting, and immobilization in glass are integrated into a single industrial platform. Mixed waste containing organics, such as organic ion exchange (IEX) resins, for instance, can be thermally gasified to powder in a fluidized bed (VTT process [62]) or burned in plasma and vitrified (“SHIVA” process [62]) **Table 1** summarizes some considerations, with select technologies involving vitrification discussed further in 3.2.

The formulation of glasses (Section 3.4) for final immobilization depends strongly on the compositions of the wastes and the criteria processing and disposal criteria. These can vary considerably depending on their origin, and thus may require different technologies for processing. **Figure 3** presents different types of LILW including: solid deposits, liquid fission products, ashes from incineration, sludges and slurries from co-precipitation processes, or solid sorbents such as zeolites or silicotitanates containing ion exchanged radionuclides.

3.2 High Temperature Thermal Treatment Options

Development of technologies for immobilization of radioactive waste in glassy waste forms normally proceeds from small batch studies through to full pilot-scale demonstrations (**Figure 4**). At the same time that nuclear waste vitrification was being developed, large-scale bulk melter technology was also being developed [63, 64]. Most melting technologies are based on resistive heating (Joule-heating) or induction heating, but a few projects have employed heating via thermal plasma or via high power lasers (e.g., [65-67]). **Table 1** and **Figure 5** show a set of comparisons amongst some of the candidate thermal processing technologies for LILW, and the following section summarizes these technologies.

The hot-wall induction melter (HWIM) was developed in France in 1962 in Marcoule and is still in use to vitrify HLW raffinate from the reprocessing of used nuclear fuel at La Hague, France and Sellafield, UK. Slightly later, the Joule-heated ceramic-line melter (JHCM) was developed in the US starting in 1973 and became the basis for HLW vitrification in the WVDP in New York, the Defense Waste Processing Facility (DWPF) at the SRS, the M-area melter at the SRS, and soon at the Hanford Waste Treatment and Immobilization Plant (WTP) for HLW and low-activity waste (LAW). Outside the US, the JHCM was further developed in Japan, Russia, India, and Germany [63]. Higher temperatures (compared to the JHCM) can be achieved using a cold-crucible induction melter (CCIM), now operational treating HLW in La

Hague, France, treating LILW at Ulchin, Korea, and Radon, Russia, and is under development in many countries for a variety of waste streams.

The details of delivering the waste and glass-forming chemicals into the melter can vary considerably depending on the melter technology and the selected process flow. For example, in the commercial French HWIM process, the liquid waste is calcined to solid in a separate apparatus [27, 28] before being fed to the melter along with glass frit. By contrast, in JHCM, liquid waste is added directly, and combined with powdered glass frit (US, DWPF), glass marbles (Japan, Rokkasho [68]), glass wool (Japan, Tokai), or as a slurry containing added glass-forming chemicals (US, WTP) [69]. In all these cases, the homogenized glass melt is poured out of the melter into a canister or container where the melt cools into a an amorphous or partially crystalline glassy solid.

In other waste management scenarios, it may be desirable to add large, irregularly-shaped contaminated parts directly without cutting and grinding. In this case there may be significant advantages to single-use melters, which are also the containers themselves [70, 71]. One type of these solutions is Joule-heated in-container vitrification (ICV), known commercially as GeoMelt®. This technology is being used by Veolia Nuclear Solutions. GeoMelt is a Joule-heated technology that uses a single-use, refractory-lined vessel where the waste vessel acts as the melter and disposal container. The single use melter with ceramic lining and graphite electrodes allows temperatures to exceed 1600°C, higher than the typical 1150°C for nuclear JHCMs. Melting generally proceeds as with slurry-fed JHCM, where a cold cap [72] is formed due to dehydrated and partially reacted feed, while the molten glass develops at the bottom of the melter. To avoid volatilization of radioactive components like cesium, some top-off frit (TOF) is used whereby only the glass-formers are added as the container is nearing full. Multiple engineering scale vitrification tests (~212-240 kg in 43×43×43 cm^3 container) were conducted, and waste loading was >80 wt.% for co-melting of Fukushima waste zeolite with carbonate slurry (CS) and iron co-precipitated slurry (IS). Single pass cesium retention in glass was >96 wt.% for most cases. Volatilized Cs was almost completely captured by a sintered metal filter and recycled [73]. This technology has been deployed in the

US to treat a variety of complex (containing combinations of metal, cement, organic, and dry powder LILW) and mixed radioactive/hazardous wastes [74]. It is also under testing for application to a broad range of complex LILW in Japan, U.S., France, Taiwan, and the UK.

A second type of in-container vitrification process is known generically as in-can melting (ICM). or in France DEM&MELT, as developed by a consortium of CEA, Orano, ANDRA, and ECM Technologies [75]. In this technology, the waste, solid or liquid, is placed directly in the canister that will become its final storage container. The canister is then heated externally by a refractory-lined resistance furnace, and when fully melted the container is cooled and the canister is removed and sealed. This simple and, to an extent, portable set-up is ideal for small-batch demolition and decommissioning projects. Temperatures are generally limited to 1150°C with Inconel 601 canisters to avoid creep and high temperature oxidation [70]. DEM&MELT is fully qualified by the CEA to treat α-emitting liquid waste and may be rated at a technology readiness level (TRL) of 7 [76]. The process has been designed to treat ILW and HLW and has been demonstrated for immobilization of secondary waste from the Fukushima Daiichi accident (see Section 4.1.1 and 4.2.2).

Another process variation which has been recently developed by CEA is the Process for Incineration and Vitrification In Can (PIVIC) [15, 77]. In PIVIC, mixed wastes including organics and metals are inserted into a vertical multi-part chamber. An oxygen plasma torch is used to incinerate the organic component at ~800°C. In the lower section, an induction melter heats the ceramic and metal fractions together. The cylindrical melter operates at low frequency (~50 Hz) [15] where the skin depth for stainless steel is large. Thus, the alternating current (AC) fields penetrate the melter can, its refractory lining, and the waste volume itself. The melt volume is effectively electromagnetically stirred, as it consists of a lower layer of metal phase and upper layer of oxide ‘slag’ containing the other components. Some studies have been performed so far to ascertain the partitioning to metal or oxide phase in the melt [78]. The great advantage of the PIVIC process is its ability to on-site process highly mixed types of waste from, for example, decontamination and decommissioning work, where separation of materials is not desirable.

The final waste form is an in-can layered structure of a lower metal fraction and upper ceramic fraction [15].

The final technique for consideration is hot isostatic pressing (HIP), which includes vitrification as an option. For HIP, the waste and specifically designed additives are mixed and calcined to produce a free-flowing powder that is dispensed into an engineered HIP canister, sometimes with additional metallic powder for redox potential control. The canister is then evacuated and sealed *via* welding and subjected to high temperatures (typically 1000 – 1300°C) and pressures (~30 – 100 MPa) where the material is consolidated into a dense monolithic form. The technology has several advantages over conventional high-temperature consolidation methods [79]. A range of waste form classes can be produced using the technology, including glass, ceramic, and advanced composite waste forms such as cermets and glass-ceramics. The waste form can therefore be tailored to the chemical, physical, and radiological properties of the specific waste stream with the target phase(s) that make up the waste form effectively immobilizing the entire range of waste species. Further, consolidation into the final waste form occurs within sealed HIP canisters, which effectively removes the potential for radionuclide volatilization and corrosive chemical emissions into the off-gas system during high-temperature consolidation. This reduces the chemical and radiological load on the off-gas system during thermal processing and minimizes secondary wastes.

3.3 Off-gas

The off-gas treatment systems at vitrification plants are designed to capture the contaminants in the off-gas stream(s), producing secondary wastes that can be processed further for safe storage and disposal. Characteristics of the off-gases of LILW vitrification plants depend on waste composition, temperature, and redox conditions in the melter with main constituents being air, water vapor, gases from decomposition reactions and, most importantly, volatilized feed materials including some radionuclides such as ^{129}I, ^{14}C, ^{99}Tc, ^{137}Cs, ^{134}Cs, and ^{106}Ru. Besides gases, aerosols significantly contribute to the emission source term. The off-gas treatment system typically comprises [80]:

- Off-gas cooling to remove condensable components and reduce the volumetric flow rate.

- Removal of the airborne particulates by wet scrubbing with low- and then high-efficiency removal.
- Removal of residual liquid aerosols generated during scrubbing by a mist eliminator to protect the final filters.
- Final high-efficiency filtration by high-efficiency particulate air (HEPA) filters to remove residual aerosols.
- In some cases, the chemical conversion of noxious gases (*e.g.*, halides, NO_x, SO_x) into benign compounds.

Usually, LILW vitrification plants are supplied with off-gas treatment systems comprising dust scrubbers and/or sleeve filters, condensers, NO_x absorbers, catalytic reactors for residual NO_x decomposition and, at the final stage, HEPA filters. **Figure 6** illustrates the range of technological apparatuses used on LILW vitrification, with an example from Russia, with overall effectiveness shown by data in **Table 2**.

3.4 Glass Formulation Issues and Strategies

Several factors, including the form of waste (solid, liquid, etc.), method of addition of glass forming chemicals (GFCs), thermal treatment technology, and off-gas treatment, must be considered when aiming to vitrifying the LILW. Specific chemistries are needed for forming glass, normally borosilicates (containing SiO_2 and B_2O_3, which is typically added as sand and boric acid or a pre-melted glass frit) or occasionally phosphates. Additional components must be added to allow thermal processing and to ensure good product performance. Some of these components are often part of the waste stream itself, such as sodium in some US LLWs or silica within soils, ashes, and zeolites. Some example compositions of LILW glasses are shown in **Table 3**. The specific example given below applies to the US LAW at the Hanford site [69], but the same principles are generally true. For cost reasons, it is ideal to maximize the so-called waste loading (normally, the mass fraction of the final glass that originated as a waste source) while maintaining desired processing envelope and final properties.

Glass formulation for LILW typically optimizes the waste loading or the waste stream flexibility while simultaneously satisfies processing and product quality related constraints [31, 71, 81-83]. The process constraints cover requirements for vitrification, which typically include melt viscosity (*i.e.*, $1 \leq \eta_{\mathrm{TM}} \leq 10$ Pa·s for process efficiency, mixing, and corrosion [84, 85]; electrical conductivity if using Joule- or Induction-heating of the melt (JHCM, CCIM, or ICV) (i.e., $10\ \mathrm{S \cdot m^{-1}} \leq \varepsilon_{\mathrm{TM}} \leq 70\ \mathrm{S \cdot m^{-1}}$) [85]; and melter refractory/electrode corrosion rates [86]. The product property constraints include: radionuclide inventory/dose, crystal content in the melt [85], solubility of troublesome components such as S, Cl, F, Cr, Tc, Ti, and/or Zr [25], chemical durability response to standardized tests [87]; phase change during slow-cooling that may impact performance [88]; and additional regulatory constraints [88]. Property-composition models can be used in most cases to formulate successful glass compositions to meet all the necessary requirements discussed above [31, 71, 81-83, 89]. When models cannot be used, this is attributed to one of the following reasons or combination thereof: (1) insufficient data to produce a model, (2) limited mechanistic understanding of the model, and/or (3) the property does not vary enough to approach the limit. These models are used to numerically optimize glass compositions for a specific waste stream, melter technology, and disposal approach. **Figure 7** gives an example of the process used.

4 LILW vitrified waste forms by region and country

In the following section, more specific narratives are given for ongoing research and practice in different nations, starting with Europe and North America, then moving to Asia and the Pacific.

4.1 Europe & North America

4.1.1 France

Low- and intermediate-level waste resulting from dismantling, rinsing of installations, or decontamination materials from effluents can be very diverse in nature and have complex physico-chemical compositions. Due to their moderate activity (see section 2), monolithic conditioning matrices (composite, crystallized or partially crystallized glasses) – which are dense, stable, and durable allowing for the long-term confinement of radionuclides, waste stabilization, and volume reduction – can be considered. In

France, a methodology is being implemented to produce vitrified waste packages compatible with long-term storage or temporary on-site storage pathways.

Tests are first conducted at the laboratory scale (i.e., masses ~ 10s – 100s of grams) to study the chemical reactivity of precursors (waste and additives), optimize the composition, the form of vitrification additives (glass frit, powders), and waste loading. They also allow the study of issues such as foaming, redox reactions, and volatilization, and help define the most suitable physico-chemical properties of additives (especially glass melt viscosity).

Subsequently, tests can be conducted in vitrification mock-ups allowing the production of up to 1 kg of vitrified material to study volatility and interactions between the melt and the crucible. Large-scale testing phases (*ca.* 300 kg) are carried out after characterization of materials obtained in the laboratory and optimization of various process parameters such as temperature, refining duration, feeding mode, and waste loading rate.

The CEA (Commissariat à l'énergie atomique et aux énergies alternatives) has a full-scale in-can vitrification demonstration tool (named DEM&MELT) to conduct these tests. The DEM&MELT process [75, 76, 90-96] is particularly suitable for vitrifying solid and liquid waste, including complex wastes such as powdery materials and particularly viscous or sticky sludges. Clogging, deposit formation, and dust entrainment are limited due to optimized waste feeding. The overall loss from volatility of radionuclides (especially cesium) is very low due to the efficiency of the gas treatment system and management of the thermal treatment cycle. Around twenty vitrification tests in an inactive environment have been conducted with the DEM&MELT process to confine waste types such as zeolites, silicotitanates, sands, ashes, and coprecipitation sludges contaminated particularly with Cs and Sr.

Table 4 provides an illustration of the main waste types studied (*Fukushima Effluent Treatment Waste*: zeolites, various mineral adsorbents, and also sludge and ash) and the results of the tests implemented. The processing temperatures used are relatively low (800–1100°C) to limit the volatility of

Cs and Sr (<0.01 % by mass), and sulfur in some cases, but still allow waste stabilization in the form of a stable monolith. The obtained vitrified material can be homogeneous at the microscopic scale or partially crystallized. Waste loading rates range 40 – 80 wt.%.

4.1.2 United Kingdom

The UK's LLW has historically been, and will continue to be, disposed of in near-surface facilities [33, 97, 98] consistent with IAEA guidelines [99]. UK ILW is derived from a range of sources, including spent fuel reprocessing, legacy wastes, and reactor operations and decommissioning, and consists of steels, graphite, concrete, cement, sand, sludges, ion exchange resins, flocculants, mixed wastes [33, 97], and ceramics and glass [99]. No single immobilization route has been designated. Cementation/grouting has traditionally been used to immobilize UK ILW [100]; however, a wider range of treatment routes has been under consideration for over 20 years [101-103]. UK wastes studied include [27, 103-109]:

- Pond sludges (SiO_2, Fe_2O_3, MgO, Al_2O_3, CaO, ZnO, UO_2, PuO_2, organics);
- Plutonium-Contaminated Materials (PCMs) (Steel, Cu, Pb, PVC, masonry, glass, PuO_2);
- Spent sand + clinoptilolite media [SiO_2, $M_{3\text{-}6}(Si_{30}Al_6)O_{72}\cdot 20H_2O$ (M = Ca, Na, K, Cs, Sr)];
- Spent IEX media (organic resins, radionuclides, process contaminants);
- Magnox sludges [$Mg(OH)_2$, Mg, U, Pu, Cs, Sr, I];
- Contaminated asbestos wastes [$Mg_3Si_2O_5(OH)_4$, concrete, masonry]; and
- Miscellaneous β-/γ-emitting solids (HEPA filters, maintenance/equipment scrap, laboratory wastes, thermocouples).

Regarding the radioactivities of the above UK ILW, based on data from multiple sources [97, 101, 105, 110], major elements and radionuclides of concern (particularly long-lived isotopes) include: ^{3}H, ^{14}C, ^{90}Sr, ^{99}Tc, ^{137}Cs, Pu, Am, and β-/γ-emitting daughter isotopes.

As discussed in a 2019 UK position paper [105], several thermal treatment/vitrification technologies have been considered and most tried at laboratory or even pilot scale with simulated UK ILW.

These include: (i) JHCM technology [111, 112]; (ii) Joule-heated ICV [106, 113, 114]; (iii) CCIM (not yet tried for UK ILW); (iv) Plasma melting [115, 116]; and (v) HIP [117-120]. Each of these technologies may be more or less suited to the treatment of the above ILW categories and it is unlikely that a single technology solution could apply optimally to all of them.

PCM wastes are typically packaged in PVC bags and stored in 200 L drums, with each drum having different metallic compositions, masonry, glass (soda-lime silicate and borosilicate), all contaminated with Pu [108]. They have received considerable attention in the UK [106, 108, 109, 115, 121-123]. Early work utilized pilot-scale plasma thermal treatment of simulated PCM waste, which resulted in glass/slag-like SiO_2-Al_2O_3-Fe_2O_3-CaO products [115]. Lab-scale vitrification trials have considered different waste blends with components representing PVC, metal, masonry, and mixed wastes, adding Ce as a Pu surrogate and ground granulated blast furnace slag or a soda-lime silicate cullet as glass-formers. The produced materials comprised glassy waste forms with metallic layers or inclusions, with Ce strongly partitioned to the amorphous component [108, 109]. In a series of trials using ICV technology [106], a simulated mix of PCM and Magnox sludge was vitrified with the addition of "local soils and fluxant" as glass-forming additives. The resulting product comprised heterogeneous vitreous waste forms, with magnesium-iron silicates dispersed throughout a glassy matrix, with Ce primarily partitioned into the glassy phase [121].

Simulant sand + clinoptilolite waste forms have been produced using JHCM [111], Joule heated ICV [110], and HIP [118, 119] technologies [121, 124]. Melting in the JHCMs with waste loadings of 75 to 80 wt.% and additions of B_2O_3, Li_2O, and Na_2O as glass-forming additives has been developed and tested at the maximum service temperature of 1150 °C [111]. Active trials undertaken using ICV did not document waste loading, but analyses showed retention of 76-77% of ^{137}Cs and ^{85}Sr, noting the unoptimized nature of the test [110]. Heath et al. [118, 119] and Gardner et al. [120] demonstrated the feasibility of treating these wastes, alone and in combination with Magnox sludge, using HIP, with up to 95 wt.% waste loading demonstrated.

Vitrification of spent IEX resins has received less attention in the UK, though a detailed survey of over 80 candidate glass compositions for immobilization of IEX resin or decommissioning sludge(s) has been reported, with down-selection based on a range of relevant criteria, including: waste loading, volatility of radionuclides and glass components, and chemical durability of the final waste form [107, 125]. Eight candidate compositions were taken forward into lab-scale trials, from which three compositions were selected as suitable. Pilot-scale trials were also performed utilizing one of these compositions.

The processing of Magnox sludge wastes has been studied using HIP [118, 120] and ICV [110, 121] and candidate glass formulations for Magnox sludge vitrification developed [126, 127]. These achieved 30-50 wt.% waste loading in a range of iron-alkali-alkaline earth borosilicate and SiO_2-B_2O_3-Al_2O_3-Na_2O-MgO glasses. Utton et al. [124] conducted detailed chemical durability testing of a laboratory simulant ILW vitrified in a borosilicate glass and two full-scale simulant vitrified products (a slag containing simulant PCM and Magnox sludge; and a glass containing clinoptilolite).

4.1.3 Russia

Nuclear waste classification and nuclear waste management are held under an overarching government strategy defined within the regulatory system of the Russian Federation [48, 128]. LILW has been generated in Russia from various sources including commercial nuclear power plants (NPPs), defense weapons production programs, research and development activities, nuclear medicine, industry, agriculture, and geology with a wide range of radionuclides being involved [129, 130]. Current LILW industrial vitrification methods in Russia are based on CCIM, with demonstration-scale testing of JHCM having been performed [129, 131]. For example, in 2001, the vitrification plant processed 254.3 m^3 of liquid LILW producing 2900 kg of glass [131].

The focus of LILW vitrification programs in Russia was on operational aqueous waste generated at NPPs. Corrosion of activated parts leads to accumulation of such radionuclides as ^{60}Co, ^{54}Mn, ^{59}Fe and shorter-lived radionuclides ^{51}Cr, ^{58}Co, ^{122}Sb while fission products such as ^{90}Sr and ^{137}Cs contaminate the coolant due to leakage from fuel elements. **Table 5** shows typical radionuclide compositions of some NPP operational LILW.

JHCM was selected for early tests with the first small-scale ceramic melter for LILW vitrification being tested at the end of the 1970s, with a pilot plant including full off-gas treatment in operation by 1987. Vitrification of both institutional and NPP radioactive wastes was successfully demonstrated at the plant at pilot level where ^{134,137}Cs retention ranged 2.5-7%. Drawbacks revealed during plant operation, including refractory and electrode corrosion, large size and weight, low specific productivity, resulted in the decision to replace it with a CCIM. During the 1980s, the CCIM-based process was developed, with three types of crucibles tested with consideration for different wastes and targeted waste forms.

CCIM vitrification tests were performed at lab-, pilot- and industrial-scale, producing various simulant waste form, including borosilicate, aluminosilicate, alumina-silica-phosphate glasses, and glass-crystalline composite materials, melted incinerator ashes and contaminated loamy soils, as well as various Synroc formulations (i.e., A, B, C) [79]. Based on this, a full-scale vitrification plant started its operation in 1999 [131], which has served as the basis for the currently recommended vitrification for operational LILW at NPP with WWER-type reactors, *i.e.*, water-water energetic reactor [132].

The vitrification plant consists of three CCIMs working in parallel (see Fig. 3 of Ref. [129]). LILW is first dewatered in a rotary film evaporator increasing the salt content in it up to 1000 $kg \cdot m^{-3}$. Then, it is mixed with glass-forming additives in the form of natural datolite, bentonite, and silica sand producing a slurry with about 20-25 wt.% water content. The slurry is then fed into the CCIM from which the glass melt is poured into 10 L containers, which are then annealed. Immiscible waste components such as chlorides, sulfates, molybdates, refractory oxides, and/or noble metals are dispersed within the melt using the mechanical stirrer, which operates periodically, increasing the capacity of the melter by 1.20 to 1.25-fold [129, 131]. **Table 6** gives the main parameters of CCIM, and glass formulations used. The liquid waste capacity is up to 200 L/h, producing up to 80 kg/h at 30-35 wt.% waste loading on an oxide basis for the three CCIM operating in parallel. Properties of the vitreous products are shown in **Table 7** . Extensive laboratory, long-term field- and computer-based tests of vitrified LILW carried out since 1987 demonstrate very good retention of contaminants by the glass [28, 133-139]. For example, the average leaching rate of radionuclides from the borosilicate glass with RBMK LILW gradually diminished during the test period from 9.4×10^{-7} $g\cdot cm^{-2}\cdot day^{-1}$ over the first year to 2.2×10^{-7} $g\cdot cm^{-2}\cdot day^{-1}$ after 16 years [135, 136]. Other areas of vitrification technologies examined in Russia include tests (TRL4 – TRL5) of self-sustaining vitrification [140] and utilization of glass-crystalline composite waste forms [8, 141].

4.1.4 United States

In the US, a large fraction of LLW is immobilized in cementitious waste forms ("grouted"), or encapsulated in polymer matrices, with notable exceptions. As with other countries [100, 121], occasionally previously grouted wastes found later to be unstable are reprocessed to the more stable glass waste form. The US has supported different LILW vitrification efforts including:

- Duratek mixed waste vitrification at SRS M-Area;
- ATG LLW Vitrification at Richland, WA;
- Fernald Environmental Management Project (FEMP) Defense LLW vitrification in Fernald, OH;
- MDA vitrification at Los Alamos, NM;
- Permafix mixed waste vitrification in Richland, WA;
- Defense LLW vitrification at Oak Ridge, TN;
- Mixed LLW vitrification in Andrews, TX;
- Defense LAW vitrification at Hanford, WA.

The US DOE has supported vitrification efforts at the Savannah River Site in Aiken, South Carolina including both LLMW and LLW; a summary of these efforts was provided by Jantzen et al. [142, 143]. Duratek, Inc. (currently Atkins-Realis) vitrified mixed LLW at the SRS M-Area. 750,000 gallons (~2.8 ML) of sludge was converted to 2.1 million lbs. (~952 metric tons) of glass using JHCM technology. ATG vitrified 2.6 million lbs. (~1179 metric tons) of LLW including dry active waste (trash), sludges, resins, and miscellaneous sources at the Safglas facility in Richland WA between 1997 and 2003. The FEMP began vitrification of radium and radon bearing silo residues using a high-temperature JHCM (1350°C). Four campaigns produced ~32 metric tons of glass before a melter failure in 1996 caused the program to end [144, 145].

GeoSafe (now Veolia Nuclear Services, VNS) performed immobilization of soils containing LILW and mixed LLW at a number of sites in the US and Australia via in situ vitrification (ISV) [146] (see **Figure 8**). A total of 24,500 tons of glass were produced at 23 active sites (e.g., Maralinga, Australia; Hanford, WA; Los Alamos, NM; Idaho Site, ID; and Oak Ridge, TN) from 1991 through 2000. Geomelt ISV vitrification was performed at Hanford, WA; Hashimoto and Ube, Japan; Permafix in Richland, WA; and Andrews, TX with mixed LILWs and commercial hazardous wastes. Note that for ISV the contaminated soils are immobilized in place (in situ) into a solid glassy block, often requiring some additives to the soil [146, 147], while for ICV a single-use container is used. Since outgassing of the molten material was often a problem, the ISV rigs were outfitted with an off-gas capture system. Testing and demonstrations of the ISV and ICV processes was performed or is underway by VNS at Richland, WA; Sellafield, UK; Fukushima, Japan; Hanford, WA; Idaho National Laboratory, ID; and in France.

The US DOE is currently supporting LAW and HLW vitrification efforts at the WTP on the Hanford Site in Richland, WA. The LAW melters at WTP are currently (as of this writing, 2024) undergoing commissioning. The source of this waste is from US Cold War weapons production efforts. Overviews of the origin of this waste, how the waste treatment process will be conducted, and property constraints for the glass waste forms are provided elsewhere [69]. Before vitrification, the Hanford LAW will be pre-treated to remove high-heat isotopes (i.e., ^{137}Cs, ^{90}Sr) through a process called Tank-Side Cesium Removal (TSCR), and the liquid streams will be concentrated through the Effluent Management Facility (EMF) whereby the stream will be partitioned into concentrated streams for recycle and dilute streams for treatment and disposal through evaporation. The Hanford LAW melters will be operated at 1150°C. Once operating, the Hanford LAW vitrification plant will be the largest of its kind in the world, with two 300-ton melters producing glass at a combined 30 tons of glass per day according to design supported by 1/3rd scale pilot-scale test data.

4.2 *Asia & Pacific*

4.2.1 Republic of Korea

Nuclear energy generation in the Republic of Korea (ROK, South Korea) is very prolific, with 26 operating NPPs accounting for about 30% of total national electricity generation [148]. LILW accounts for about 60% of the total waste volume generated during NPP operation [2]. These LILW are generally classified into dry active waste (DAW), borate waste, spent filter, and spent IEX, accounting for 56%, 28%, 2%, and 14% of the total volume of LILW, respectively [149]. These wastes will be dispositioned to the Wolsong disposal center located in the southeastern region of the Korean peninsula in Gyeongsangbuk-do province, which is ~27 km southeast of Gyeongju city. It has been built as an underground silo-type concrete-based facility, where the silos are located between 80–130 m below the surface, and any groundwater passing through the silos proceeds towards the sea in a southeasterly direction [150, 151]. The six silos at the Wolsong disposal center can hold about 100,000 drums. The facility is operated by the Korea Radioactive Waste Agency (KORAD) since 2015, and 17,498 drums (3,500 m^3) of LLW have been disposed of as of the end of March 2019 [151]. According to waste acceptance criteria (WAC) in South Korea, homogeneous radioactive wastes such as spent IEX, borate waste, and other liquid wastes should be solidified before disposal. Cement has been used for this purpose, but other waste forms have been also examined for use [152-158].

Glass (or vitrification) is also one of the waste forms widely used in the ROK for LILW immobilization [156, 158]. Since 1994, the Korea Hydro & Nuclear Power Co. Ltd. (KHNP) has been studying the vitrification of LILW generated from NPPs [159]. Sodium alumino-boro-silicate glass forms the primary compositional baseline for vitrification. Several types of glasses developed by KHNP are shown in **Table 8**. The commercial Ulchin Vitrification Facility (UVF) was completed in 2009 and has been vitrifying combustible LILW using a CCIM, with a regular throughput of 20 kg/h (max. 25 kg/h) [160].

Concentrations of hazardous off-gases such as CO, HCl, NO_x, and SO_x were determined to be well below the emission limits. The final glass waste form showed high durability and low leachability of contaminants of concern as assessed by the Material Characterization Center-1 (MCC-1), product consistency test (PCT), vapor hydration test (VHT), and ANS 16.1 methods [157, 159, 161]. The volume reduction factor of most glasses is > 33 compared to the initial bulk volume of the waste, and all other objectives meet the performance criteria, operational safety, and stability requirements of the facility [160]. Due to the limited space of the existing repository in the ROK, vitrification of LILW is highly recommended and can significantly contribute to radioactive waste management.

4.2.2 Japan

In Japan, LLW is generated by nuclear power plants, spent fuel reprocessing facilities, MOX fuel processing facilities, and Fukushima Daiichi Nuclear Power Station (FDNPS) decommissioning and fallout radiological waste [162]. Unique waste streams, for example, rubble, cut trees, and secondary wastes, such as effluent from the Advanced Liquid Processing System (ALPS) for contaminated water treatment from the FDNPS accident, present unique challenges. Japan and the international community have been working to immobilize the FDNPS wastes since shortly after the accident in 2011. Many organizations in Japan are actively performing R&D on waste disposition, including waste forms and geological repositories, *e.g.*, NUMO, JNFL, JAEC, NSC, NISA, JAEA.

Common routes for LLW disposition are incineration, compression, and cement waste forms, while vitrification technology has yet to be adopted industrially. Disposition of LLW waste in Japan began in 1992 at the Low-Level Radioactive Waste Disposal Center of Japan Nuclear Fuel Limited (JNFL). Due to the successful implementation of vitrification technology in other countries, such as the US, France, and Korea, Japan has begun the development of vitrification technology, including Fused Glass Solidification (FGS) in conjunction with CCIM technology and ICV technology.

Following the FDNPS accident, large quantities of seawater and freshwater were injected into the reactors to provide cooling. Within the cooling water, the primary radionuclides of concern are ^{137}Cs and

^{90}Sr [75, 163]. To decontaminate the cooling water, three purification systems were implemented following the accident (**Figure 9**): (1) Kurion-Areva/Veolia system, (2) Simplified water retrieval and recovery system (SARRY), and (3) Advanced Liquid Processing System (ALPS). Each purification system carried design improvements to replace or complement the previous system. About 70% of the secondary water treatment waste is from ALPS. Some details on the processing of this waste were discussed in section 4.1.1.

Despite vitrification of LLW being an active area of research for decades, Japan has yet to implement vitrification technology for LLW. In 2014, the Agency for Natural Resources and Energy of the Ministry of Economy, Trade and Industry initiated the "basic research programs for the next generation vitrification technology." One of the objectives of the primary research programs is to develop vitrification technology for LLW into a stable solidified waste form with substantially reduced volume. To meet this objective, the primary research programs have brought together industry-leading experts in vitrification technology including the IHI corporation, JNFL, the Japan Atomic Energy Agency (JAEA), and the Central Research Institute of Electric Power Industry (CRIEPI).

IHI Inc. (formerly Ishikawajima-Harima Heavy Industries Co., Ltd.) is developing "fused glass technology" to treat LLW in Japan. This technology is similar to conventional vitrification, but the fused glass solidification technology uses the silica already contained within the waste (i.e., in the soil) as a glass-former [162]. Fused glass solidification technology has the potential to reduce the volume of unique waste streams, such as those from the FDNPS radiological fallout, by reducing the amount of glass additive and increasing final waste loading.

For FDNPS waste streams, IHI's review of vitrification melting technologies concluded that CCIM is the most appropriate melting technology for fused glass solidification due to (1) the formation of a skull layer minimizes corrosion of the furnace wall, thereby extending melter furnace life and permitting the use of highly corrosive glass within the melter, (2) short start-up and shutdown times wastes can easily be changed from day to day, and (3) the opportunity of treating many types of wastes (*e.g.*, sludge, zeolites, spent resin, liquid wastes, ash and combustible wastes) [162, 164]. Due to Korea Hydro and Nuclear Power

(KHNP) experience with CCIM, IHI entered into an agreement with KHNP to study CCIM, with testing carried out from 2013 to 2015. IHI confirmed a waste loading of 20-65 wt.% for ALPS waste streams, including carbonate slurry, iron coprecipitation slurry, zeolite (spent resin), silicotitanate (ash), and ferrocyanide sludge, at temperatures < 1200°C. Zeolites were vitrified at 1050°C due to prevent Cs volatilization. Carbonate slurries were supplied at greater than 40 L/h. No crystals were observed in the glass confirming homogeneity. For zeolite and incinerated ash, volatilization rate of Cs_2O was kept to less than 8%. All vitrified waste streams, except for ferrocyanide sludge, meet US standards for high-temperature viscosity, electrical conductivity criteria and leaching rates.

Concurrently, the Japanese government is also supporting joint development of GeoMelt® and DEM&MELT In-Container Vitrification technologies. Finucane et al. [73] processed three melts and performed durability testing. Test melts utilized non-radioactive Cs and Sr simulants. Single-pass retention tests of Cs and Sr ranged 91.46-99.30 wt.% and 99.76-100 wt.%, respectively. Waste loadings ranged from 70-82 wt.% and volume reductions ranged from 74-79 vol.%. Durability testing was performed on all resulting glasses and compared to one US glass standard (EA), and two Japanese reference glasses (P0797, P0798), with measured leaching rates and normalized mass losses comparable to or lower than the references.

The DEM&MELT ICV process being developed at CEA (see section 3.2 and 4.1.1), while primarily designed for the treatment of ILW and HLW, has also shown process for treatment of secondary FDNPS wastes [76]. Three waste chemistries were studied: (1) all-waste (comprising zeolites, silicotitanate sand, precipitation sludge simulants); (2) Cs-rich waste; and (3) Sr-rich waste. Localized crystallization was observed in all melts, predominantly at the top and bottom-layer of the crucible due to the high concentrations of Ti, Zr, and Nb [76]. Waste loading was limited to 50 wt.% in the Sr-rich melt to limit the Mg from the ALPS carbonate sludge and minimize magnesium silicate crystallization. Simulated FDNPS ALPS slurries (a combination of the ferric and carbonate slurries) have also been vitrified with borosilicate glass additives. The successful full-scale test (*ca.* 280 kg) attained a 60 wt.% waste loading, after being heat treated to 1100°C for 17 min.

4.2.3 India

In India, LILW are characterized according to established criteria [165]. Liquid LILW is treated by processes such as chemical precipitation, ion exchange, evaporation, and reverse osmosis [165-167]. Solid LILW arises in two types: 1) 'primary wastes' comprising radiologically contaminated components and equipment (e.g., metallic hardware) and spent radiation sources; and 2) 'secondary wastes' resulting from site operation [166]. Solid LILW includes protective rubber and plastic wear, miscellaneous metallic components, cellulosic and fibrous materials, spent organic IEX resins and filter cartridges [166]. Combustible LILW is incinerated and compactable wastes are reduced in volume by mechanical compaction [166]. Non-α-emitting LILW solid and solidified wastes generated during reactor operation are disposed in near surface disposal facilities [168, 169].

ILW was typically immobilized with cement, bitumen, and composite polymers since 1985 [168, 169]. All the intermediate level liquid wastes (ILLWs) of nuclear fuel reprocessing origin are now treated using IEX resins to separate them into LLW and HLW, greatly reducing waste form volumes [169], hence ILLWs are no longer directly immobilized [168, 169]. Legacy alkaline ILLWs from reprocessing have been treated by sorbents and resins and the organic bitumen and polymer matrices previously proposed for direct immobilization were never implemented [168].

As of 2022 [170], liquid ILW generated during spent fuel reprocessing is alkaline in nature and is rich in ^{137}Cs, inactive salts and dissolved organics. It is treated using a Cs-selective resorcinol formaldehyde (RF) IEX resin. The Cs-rich eluate is concentrated and immobilized in a vitreous matrix as HLW. The LLW effluents are managed by various treatment methodologies involving industrially usable precipitants. ILW treatment plants operate at Trombay and Kalpakkam [170]. Liquid LLW is typically treated by co-precipitation and immobilized in a cement matrix [166, 169], whereas solid LLW are treated by compaction (polymers and rubber); combustion/incineration (cellulosic materials); and melt densification (PE), with untreatable wastes directly disposed [169]. A plasma-assisted incinerator/pyrolizer has been developed for treating Category I solid waste, as a conventional incinerator only catered to cellulosic wastes (volume reduction factor, VRF 30-50). The plasma based system caters to all rubber/plastic/cellulosic wastes (VRF 30) [169]. The system has been successfully applied to treat 500 kg of inactive mixed waste and 500 kg of radioactive cellulosic waste [169]. Recent research [171] compared decomposition and Cs, Sr retention of representative solid LILW (PE, PVC, rubber, cellulose, mixed wastes) using conventional thermal gravimetric analysis, lab furnace and plasma-based approaches, and determined that the engineering scale plasma pyrolysis based approach with mixed waste forms confirmed superior weight and volume reduction factors compared to the other methods with Cs and Sr "well confined" [171].

4.2.4 China

Vitrification of LILW in other Asian countries is still being considered an alternative to lower-cost alternatives. In China, for instance, LILW is typically immobilized in cement [172]. However, Chinese researchers have developed thermal plasma technology (TPT) since 2009 to decontaminate solid wastes for safety and volume reduction. In this technology, organic matter is incinerated (gasified) by a thermal plasma torch, and the resulting inorganic ash can be melted with glass frits or glass-forming chemicals at temperature below 1250℃ in a melter [173, 174]. The pyrolysis characteristics of typical LILW, as well as the glass formulation for the residual ashes from LILW, have been investigated, and hundreds kilograms of durable simulated LILW glasses have been produced from a research-scaled LILW TPT treatment furnace [175, 176]. The construction of a LILW TPT demonstration plant (treatment capacity: ~500 t/a) was completed in 2023 in Gansu Province, and a few cold tests have been performed.

The formulation of LILW glasses is challenging, due to the complexity and variability of the waste sources generated, causing extensive variations in the composition of residual ashes from LILW TPT gasification. As developed for immobilizing HLW, borosilicate glass is considered a candidate host glass to immobilize residual ashes of LILW. Different borosilicate glass formulae have been developed based on the compositions of the inorganic residues from LILW. For example, the filter media of high efficiency particulate air (HEPA) filters is composed of ~95 wt.% glass fiber and bears a relatively high viscosity at 1200°C, thus, Na_2O and CaO were added to ensure low enough viscosity at the melting temperature where volatility of radionuclides is minimal [177]. In addition, the filter media of HEPA filters, which is >50 wt.% SiO_2, was proposed to be a glass additive to immobilize the combustible waste ashes, resulting in a minimization of the glass additives.

4.2.5 Australia

The Australian Nuclear Science and Technology Organisation (ANSTO) is the home of Australia's most significant landmark and national infrastructure for research. Importantly, it operates Australia's only nuclear reactor, the Open Pool Australian Light Water Reactor (OPAL). This 20 MW thermal reactor is one of the world's most modern multipurpose research reactors and it supports nuclear medicine production, several neutron beam instruments, as well as industrial irradiations, including silicon ingots for the manufacture of high-performance semiconductors. For 50 years prior to OPAL, ANSTO operated Australia's first research reactor, HIFAR. Australia does not operate any nuclear power reactors.

ANSTO is a major producer of medical radioisotopes, including ^{99}Mo, which is used to generate ^{99}Tc for medical imaging. The majority of radioactive waste produced at ANSTO is directly associated with its production of nuclear medicine. As a result, ANSTO produces ILW and LLW which has been managed safely and securely on ANSTO's Lucas Heights site for 70 years.

In Australia, the safe use and management of radiation and nuclear technologies is regulated by ARPANSA (the Australian Radiation Protection and Nuclear Safety Agency). One important aspect of ARPANSA's role includes ensuring Australian entities such as ANSTO safely manage their radioactive waste holdings. Further, ARPANSA provides the definitions for the classification of radioactive waste in Australia [39], with alignment to IAEA's General Safety Guide No. GSG-1 [34]. The Australian Safeguards and Non-proliferation Office (ASNO) is Australia's nuclear regulatory authority for nuclear security and nuclear safeguards. ASNO is the state authority responsible for Australia's compliance with its international and domestic obligations to nuclear safeguards. In addition, the Australian Radioactive Waste Agency

(ARWA) is responsible for the delivery and operation of a permanent, purpose-built facility for Australia's nuclear wastes. ARWA is also responsible for managing this yet-to-be-established future storage and disposal site, as well as its associated waste acceptance criteria.

The spent fuel from ANSTO's research reactors is currently exported to either France or the UK for reprocessing, which allows recycling and reintroduction of the fissile isotopes back into their fuel cycle programs. The remaining waste is vitrified in those countries, and an equivalent radionuclide inventory of fission products is returned to Australia as either a UK or French-vitrified product. ANSTO continues to store two TN-81 storage casks (6.5 m long × 3 m diameter with 20-cm thick steel walls) on its Lucas Heights site until a National Radioactive Waste Management Facility is operational. One of these TN-81 casks contains 4 × 500 kg canisters of vitrified nuclear waste from the UK, the other 20 × 500 kg canisters of vitrified nuclear waste from France. More than 90% of the radioactive waste produced by ANSTO is LLW, and this includes paper, plastic, gloves, clothes, and filter systems. This waste is shredded and compressed into 200-L drums. ANSTO will use super-compaction to reduce the waste volume, with cement overpack providing a radiation shield.

The two primary liquid ILW streams produced from ^{99}Mo production have an activity of $\sim 10^{10}$-10^{11} Bq·L^{-1} (following ∼3 years decay time from processing), with the majority of the radioactivity from ^{137}Cs [79]. These two wastes differ due to the different irradiation targets and ^{99}Mo processing routes. The first liquid ILW is a uranyl nitrate solution (~120-200 g U·L^{-1}) in 0.5-1 mol·L^{-1} HNO_3 solution and contains fission products and process impurities. It was produced from the 1980s – 2005 following "acidic route" ^{99}Mo processing. The second liquid ILW is comprised of a NaOH and $NaAlO_2$ solution with fission products (with a Na concentration of 6-7 mol·L^{-1} and Al concentration of ~1.5 mol·L^{-1}) and is currently produced at ANSTO following an "alkaline route" ^{99}Mo processing.

ANSTO Synroc® is a technology platform that is being developed by ANSTO for the treatment of ANSTO's ILW streams. It is a highly flexible waste processing technology with unique characteristics, which make it suited to the treatment of problematic wastes generated from current and future fuel cycle activities [79, 178]. A key aspect of the technology is the employment of hot isostatic pressing (HIP), which uses temperature and pressure during waste form consolidation into the final monolithic form (see section 3.2 for more details on the HIP process technology). ANSTO are currently commissioning a first-of-a-kind Synroc Waste Treatment Facility (SWTF) which is designed to treat the primary ILW from the "alkaline route" ^{99}Mo processing. The waste form design is a sodium aluminoborosilicate glass formulation with ~25 wt.% (oxide basis) waste loading, although this varies somewhat with the Na and Al molarity in the waste feed. The fully automated process technology has been designed to transform a mixture of liquid waste and waste-forming additives into to a tailored granular powder that is subsequently consolidated through HIPing (high level process flow is provided in **Figure 10**). The SWTF will produce a sodium aluminoborosilicate glass *via* HIP in a 30 L HIP canister at 1150°C. The product is suitable for long-term storage or future repository disposal in Australia, displaying normalized mass losses for all elements in the ASTM C1285 (PCT) aqueous durability protocol [179] of $< 1.0\ \mathrm{g{\cdot}m^{-2}}$. This is in line with requirements for the Hanford Immobilized Low-Activity Waste Product Acceptance in the US (PCT-A, response ($NL_{[Na,B,Si]} \leq 2\ \mathrm{g{\cdot}m^{-2}}$) [180].

ANSTO is also progressing the application of Synroc Technology for the treatment of the primary ILW from "acidic route" ^{99}Mo processing. For this uranium-bearing waste, a pyrochlore-rich multiphase ceramic [181] or glass-ceramic [182] has been designed with 30 – 50 wt.% waste loading (oxide basis). The tailored glass-ceramic waste form demonstrates flexibility in the waste form design to receive the required waste variability; it also suitably passes aqueous durability performance requirements and has been demonstrated successfully at scales up to 1 kg. In the glass-ceramic design, the addition of glass facilitates the incorporation of fission products, while the inclusion of pyrochlore immobilizes the uranium with high waste loading. The up-scaled dense HIPed sample showed normalized elemental releases (NL_i) of < 2 g/L for all elements in the ASTM C1285 aqueous durability experiment [179].

There is a secondary waste stream generated during ^{99}Mo production at ANSTO which is also worthy of discussion. This waste is challenging to immobilize due to its high concentration of lithium (Li^+)

and sulfate ions (SO_4^{2-}), its acidic nature, and its radioactivity (again predominantly Cs). Optimized glass [183] and glass–ceramic [184] laboratory scale waste forms are currently under consideration for this waste, again with processing *via* ANSTO Synroc® technology. The waste form design aims to maximize sulfate incorporation while also achieving acceptable chemical durability. In this case, a glass waste form with high sulfate incorporation of 2.8 wt.% SO_3 (corresponding to a waste loading of 11 wt.% as Li_2SO_4) was achieved. However, tailored glass–ceramic waste forms were able to be produced at lower temperatures and with higher waste loadings (~16 wt.% on an oxide basis). The glass-ceramic waste form produced $BaSO_4$ crystals within a glass matrix as designed. Satisfactory chemical durability was indicated using the ASTM C1285 standard test method [179] and normalized mass losses below 2 $g \cdot m^{-2}$ for all elements were again observed.

5 Summary and Conclusions

Stabilization of wastes and immobilization of waste radionuclides to prevent contaminant and radionuclide transport into the biosphere is of paramount concern to maintaining the health and safety of the world. This review provided an overview of radioactive waste classification, focusing on low-level and intermediate-level radioactive waste (LILW). One option for the stabilized form for disposal of these types of wastes involves vitrification into glassy waste forms, which has been one of the leading candidate technologies for immobilizing radioactive waste over the past several decades. This review covered current and proposed LILW vitrification activities that have been conducted internationally (i.e., Europe, Asia, North America, and the Pacific). Various thermal treatment technologies have been considered and are mature and appropriate for certain wastes. Among these are Joule-heating melting, hot-wall induction melting, cold-crucible induction melting, and Joule-heated in-container vitrification. Pre-treatment technologies include plasma processes, incineration, and calcination. Since glass is an amorphous material which can incorporate any radionuclide at the atomic scale, it is very flexible for waste variability. Each application must be evaluated for suitability of the composition, additives needed, off-gas treatment, economics, and tolerance of crystalline phases. Future application of portable or rapidly transported thermal treatment such as the types of in-container vitrification promise to make decommissioning of legacy nuclear sites faster and more efficient. A robust international community of researchers working together has coalesced around radioactive waste management, and their collective action has resulted in, and will continue to result in, the deployment of robust and safe radioactive waste management technology across the globe.

Acknowledgements and funding

This paper has been a collaboration of the authors and their respective agencies and organizations. Pacific Northwest National Laboratory (PNNL) is operated by Battelle Memorial Institute for the DOE under contract DE-AC05-76RL01830. JSM, MCDW, and JB acknowledge funding from the United States Department of Energy (US DOE) Waste Treatment & Immobilization Plant (WTP) Federal Project Office, contract number 89304022CEM000015. The DEM'N'MELT project was supported by Andra as part of the "*Nucléaire de Demain*" programme of "Investissements d'avenir" under grant agreement No RTSCNADAA160017 and No RTSCNADAA17-0010. The Fukushima waste applicability study was funded by the Japanese Ministry of Economy, Trade and Industry as The Subsidy Program "Project of Decommissioning and Contaminated Water Management". CEA also acknowledges H2020 PREDIS: Pre-disposal management of radioactive waste project, European Union's Horizon 2020 research and innovation programme under grant agreement No 945098 (2020-2024). KX acknowledges funding from the National Key R&D Program of China under grant agreement No 2018YFB1900203.

Data availability

No new previously unpublished data was used for the research described in the article.

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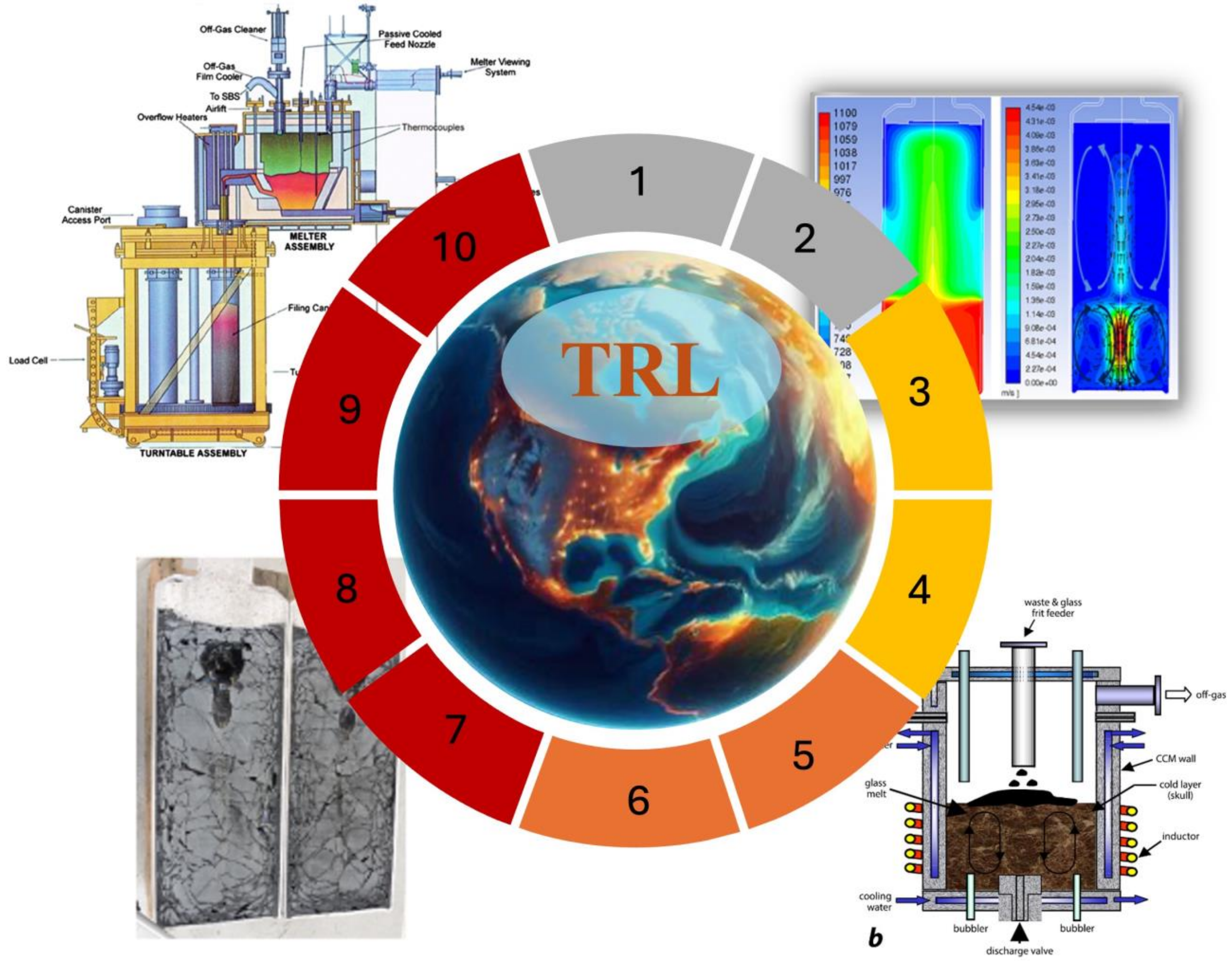
Figures



Graphical abstract

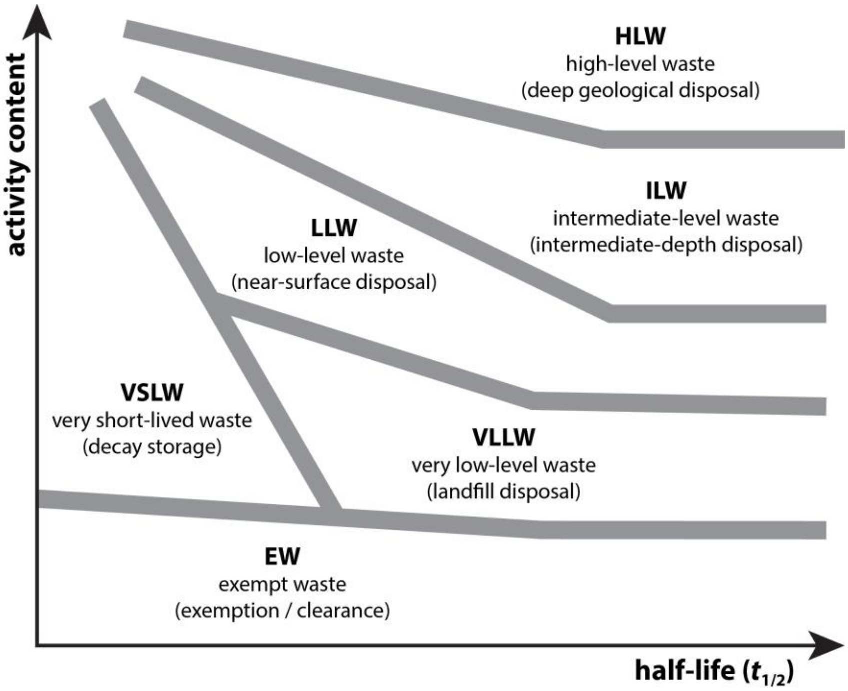


Fig. 1. IAEA classification guideline for radioactive waste, after [34].

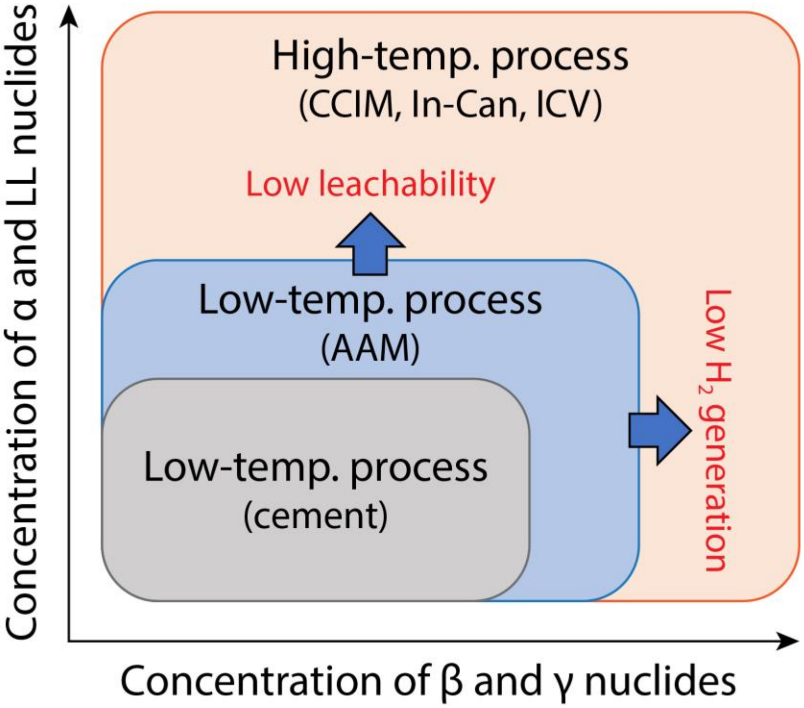


Fig. 2. Selection of waste forms for Fukushima Daiichi accident waste, after [50]

Fig. 3. Different types of LILW streams

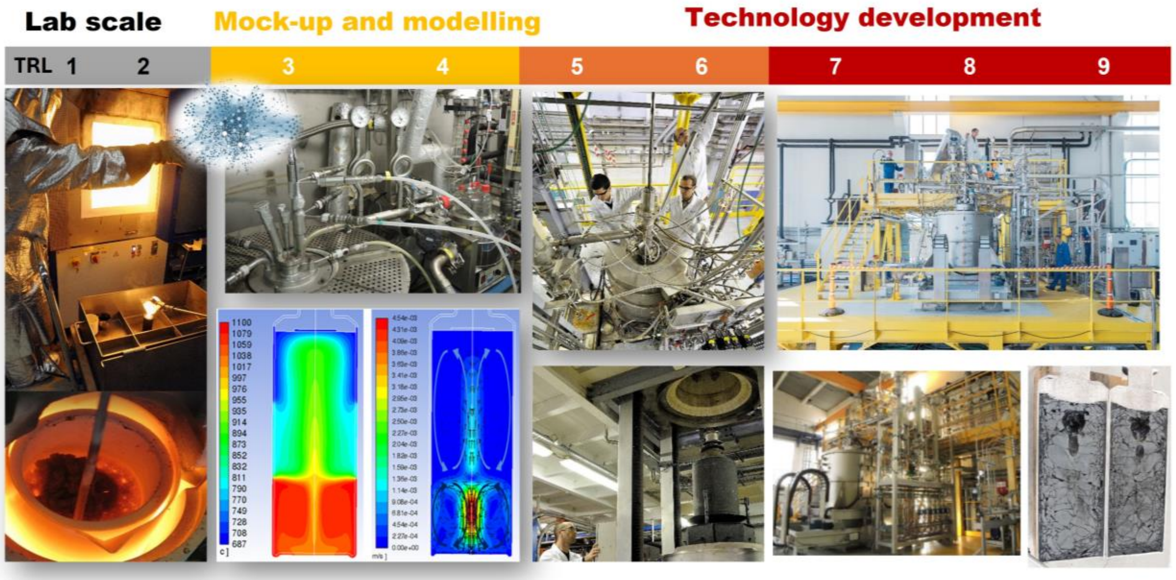


Fig. 4. Vitrified waste form development cycle, exemplified by CEA France.

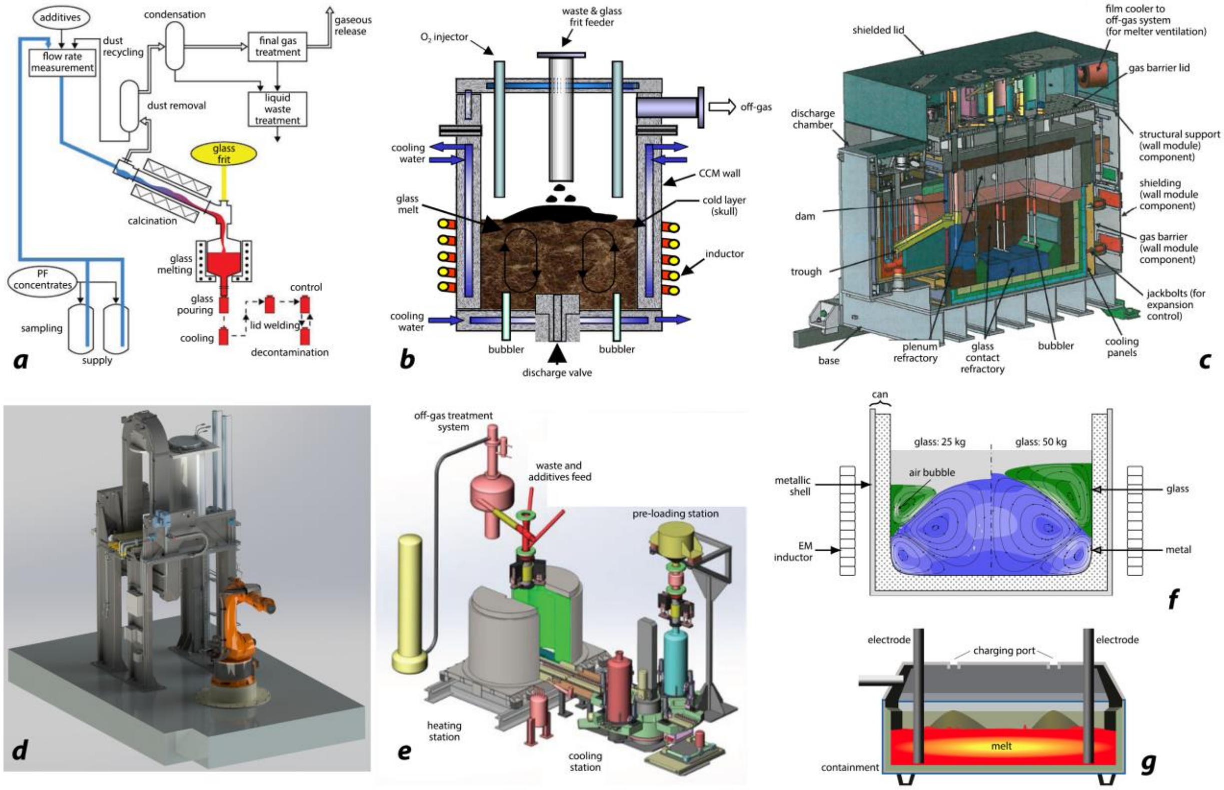


Fig. 5. Montage of thermal processing technologies for vitrification of LILW. (a) hot wall induction melter (HWIM), modified from [185]; (b) cold crucible induction melter (CCIM), modified from [186]; (c) Joule-heated ceramic melter (JHCM), for vitrifying Hanford LAW, courtesy of US Department of Energy; (d) hot isostatic press (HIP), courtesy of ANSTO; (e) in-can melting (ICM) using external heating, example DEM&MELT, France, modified from [75], CC BY 4.0; (f) PIVIC process showing metal and glass melt, modified from [187]; (g) in-container vitrification (ICV) using Joule heating, example Geomelt®.

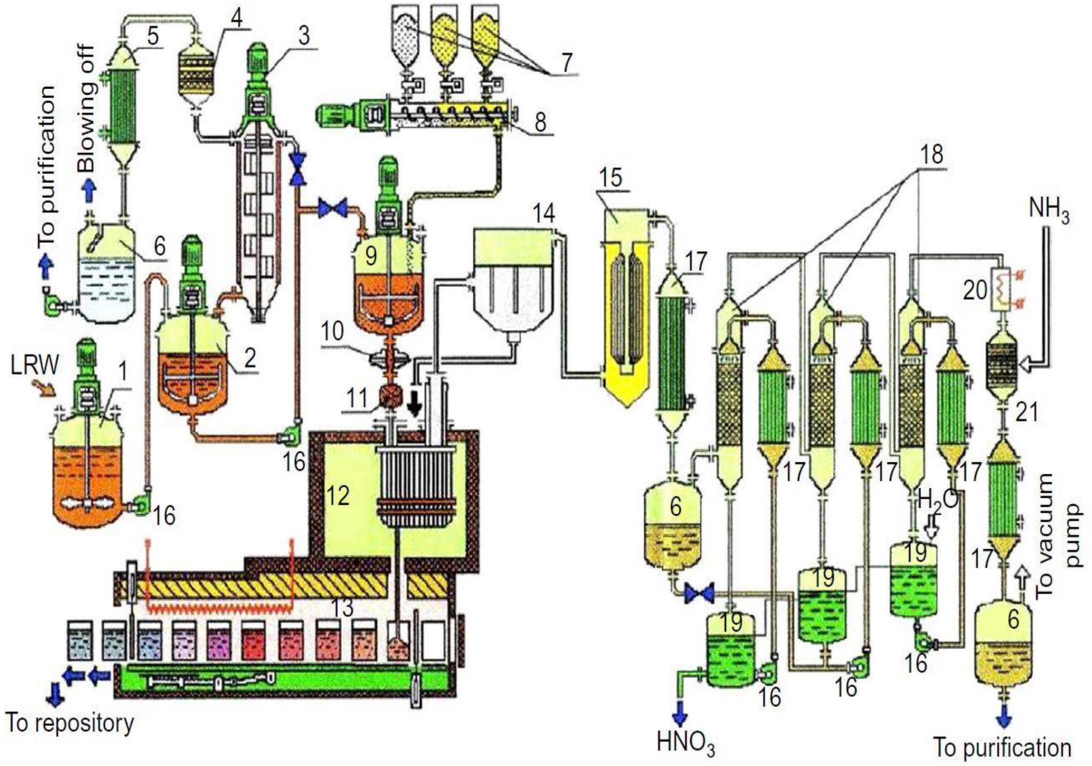


Fig. 6. Technological sheet flow diagram of “Radon” LILW vitrification plant. 1 – interim storage tank, 2 – concentrate collector, 3 – rotary film evaporator, 4 and 15 – HEPA filters, 5, 17 and 21 – heat exchangers, 6 and 19 – reservoirs, 7 –glass-forming additives hoppers, 8 – screw feeder, 9 – batch mixer, 10 – mechanical activator, 11 – peristaltic pump, 12 – cold crucible, 13 – annealing furnace, 14 – sleeve (coarse) filter, 16 – pumps, 18 – absorption columns, 20 – heater. Reproduced from [80] with permission of IAEA.

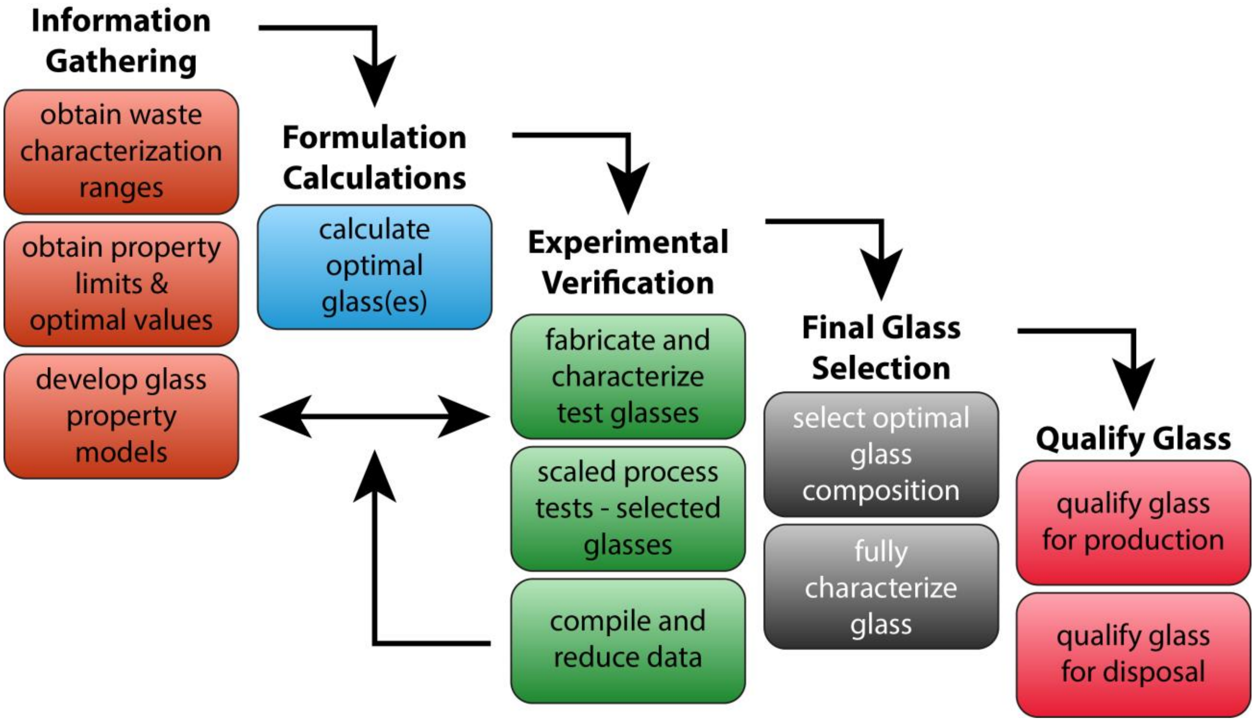


Fig. 7. Process of glass formulation for radioactive waste glasses.

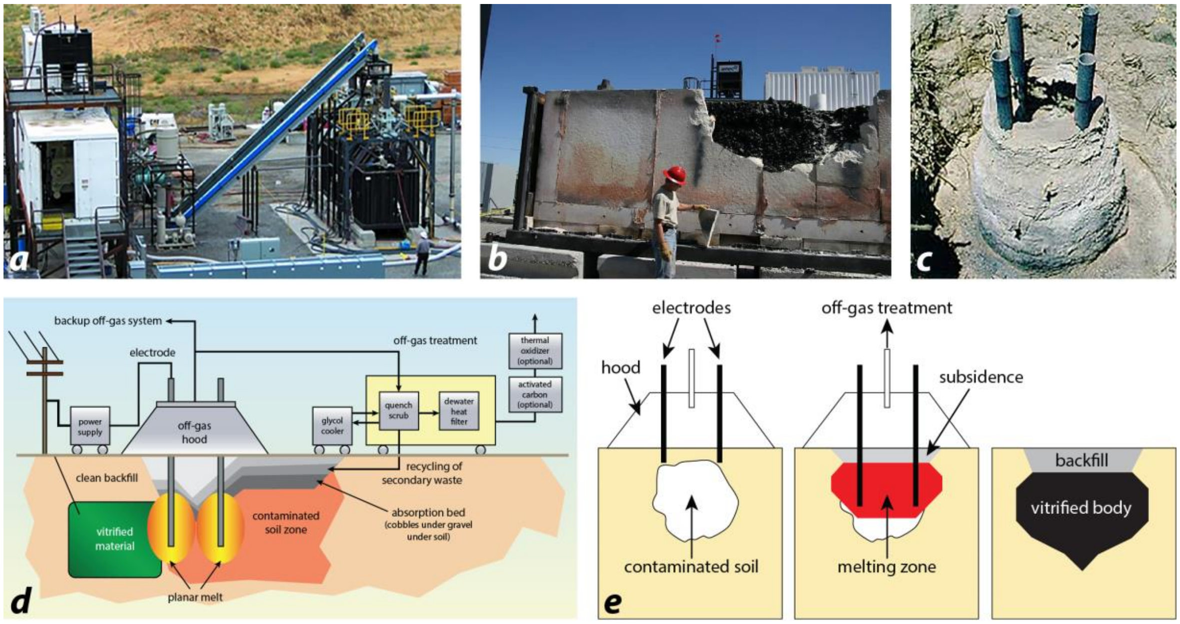
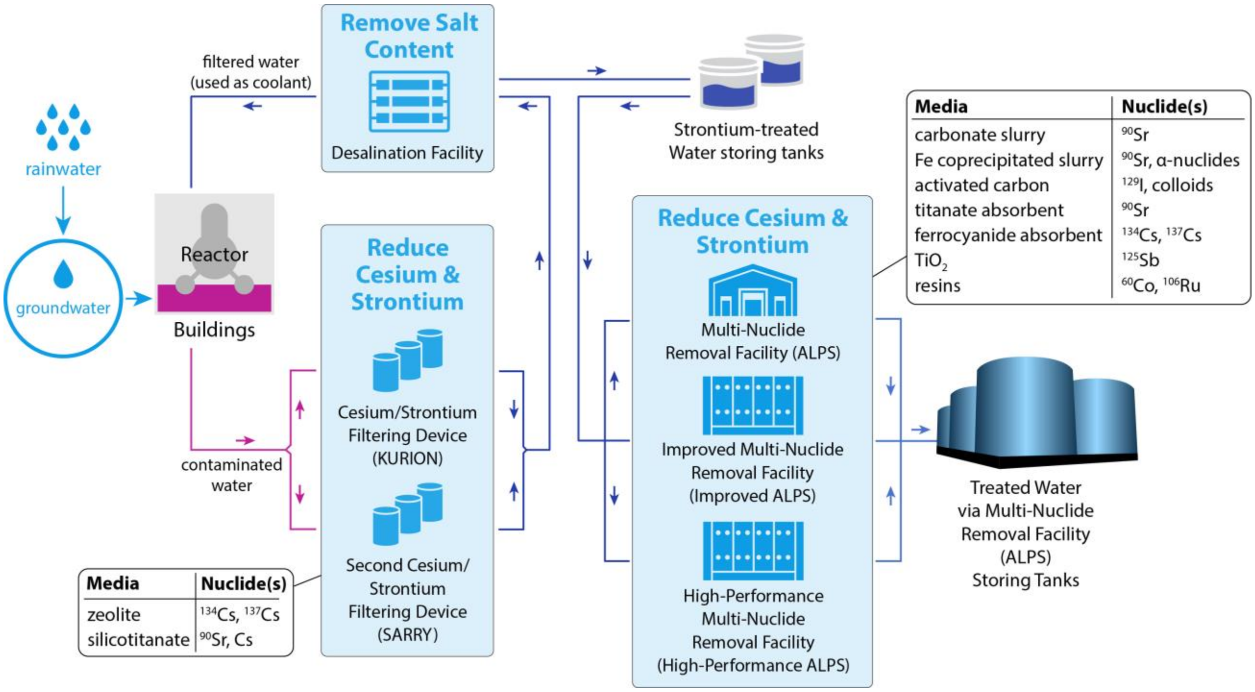


Fig. 8. Bulk vit and in-situ vit processes. a), b) bulk vitrification, see also [55, 188]; c) vitrified soil through ISV, see also [147]; d), e) ISV process, redrawn after [27, 189]. Photos courtesy of Brett Campbell, Veolia Nuclear Solutions.



Media	Nuclide(s)
carbonate slurry	^{90}Sr
Fe coprecipitated slurry	^{90}Sr, α-nuclides
activated carbon	^{129}I, colloids
titanate absorbent	^{90}Sr
ferrocyanide absorbent	^{134}Cs, ^{137}Cs
TiO_2	^{125}Sb
resins	^{60}Co, ^{106}Ru

Media	Nuclide(s)
zeolite	^{134}Cs, ^{137}Cs
silicotitanate	^{90}Sr, Cs

Fig. 9. Schematic for processing of Fukushima Daiichi wastes, radionuclide inventory, and secondary waste streams, redrawn after [190, 191].

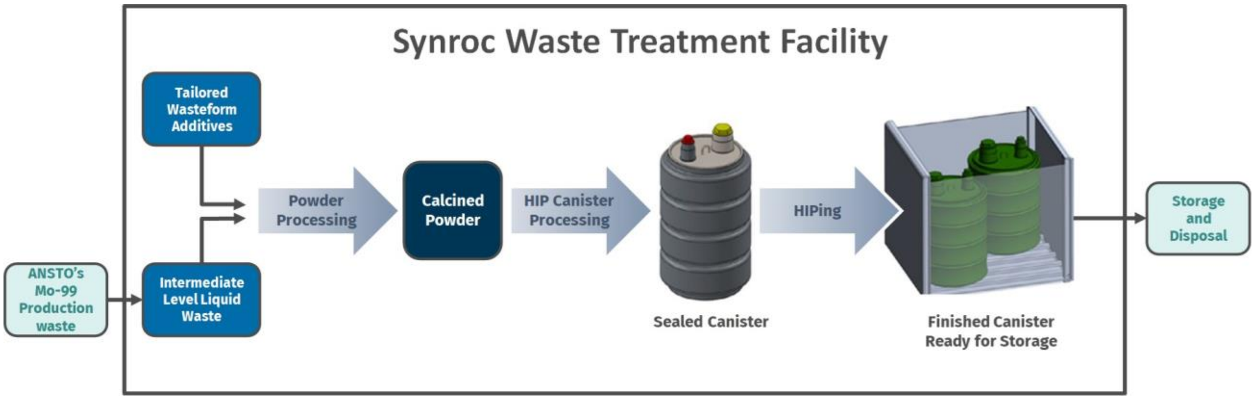


Fig. 10. Process flow for the Synroc Waste Treatment Plant. Courtesy of ANSTO.

Tables

Table 1. Comparison of thermal treatment processes (continued on multiple pages)

Treatment Technology	**Technology Description**	**Technology Status**	**Considerations**
Joule-heated ceramic melter (JHCM)	Waste may require pretreatment (e.g., Hanford LAW to remove ^{137}Cs and ^{90}Sr). In a continuous process, liquid waste is added directly, and combined with powdered glass frit, glass marbles, or as a slurry containing added glass-forming chemicals. The melters (e.g., Hanford LAW melters) are typically operated at 1150°C. The homogenized melted glass is poured out of the melter into a canister.	Developed in the US since 1973 Industrially operated for HLW on several sites, e.g., US, Germany, Belgium, Russia, and Japan Demonstration testing for LILW in Russia. Trialed in UK with simulated LILW e.g., Simulant sand + clinoptilolite	Off-gas treatment systems must be designed to capture contaminants, producing secondary wastes. Noble metal accumulation. Glass viscosity and electrical conductivity characteristics.
Hot-wall induction melter (HWIM)	Liquid waste is calcined to solid in a separate apparatus before being mixed with glass-forming powders and charged into the melter. The homogenized melted glass is poured out of the melter into a canister.	Developed in France since 1962 Industrially operated in La Hague and the UK to vitrify HLW.	Off-gas treatment systems must be designed to capture contaminants, producing secondary wastes.
Cold-crucible induction melter (CCIM)	The cold crucible is a compact cooled melter in which the radioactive waste and the glass are melted by direct high frequency induction heating. The melter containment is water cooled to prevent the outer layer of feed/frit from melting. Therefore, the melt is carried out within a ‘skull’ of unmelted feed/frit. The homogenized melted glass is poured out of the melter into a canister.	Commissioned also at La Hague for certain HLW. LILW industrial vitrification in Russia since 1999. Developed in the Republic of Korea since 1994 and commercially operated since 2009 for LILW. Some development work in Japan.	Off-gas treatment systems must be designed to capture contaminants, producing secondary wastes. The skull prevents adherence of materials to the wall and thus corrosion and decontamination is reduced. The skull also allows operation at higher temperature (e.g., 1300°C – 1400°C). Allows processing of wastes requiring more refractory glass compositions.

Hot isostatic-pressing (HIP)	The waste and additives are mixed and calcined to produce a free-flowing powder that is dispensed into an engineered HIP canister. In a batch process, the canister is evacuated and sealed and HIPed at high temperatures (typically 1000 – 1300°C) and pressures (~30 – 100 MPa) where the material is consolidated into a dense monolithic form.	Developed in Australia since 1995 and currently commissioning an industrial facility for LILW. Trialed in UK with simulated waste. e.g., Simulant sand + clinoptilolite e.g., Magnox sludge, and for the immobilization of Pu residues at Sellafield.	Waste pre-treatment required to render the feed materials suitable for consolidation *via* HIP (e.g., calcination). Can produce glass, glass-ceramic, ceramic and other advanced composite waste forms. Removes potential for radionuclide volatilization and corrosive chemical emissions into the off-gas system during high-temperature consolidation. Minimizes secondary wastes. Removes melter corrosion problems as processing unit is separated from the waste. This also allows processing of varied feeds (orphan/niche wastes).
In-situ vitrification (ISV)	Developed to remediate contaminated soils by stabilizing and immobilizing them in place (in situ) in a solid glassy block. In most cases, some additives are mixed with the soils before heating to ensure adequate melting. Graphite electrodes are placed in the ground and a graphite starter path was used to begin Joule-heated melting of the soil.	Developed by Battelle Pacific Northwest National Laboratory. Various crucible tests on actual soils as well as bench-top and full-scale tests were completed.	Requires an off-gas capture system due to outgassing of the molten material. In-situ operation. Some methods for mixing the flux with soil have been explored, as well as water removal and organic pyrolysis of soils.
Plasma Treatment	The use of plasma to transfer energy into melts. This has been studied quite extensively in the treatment of municipal waste. Provides the complete dissociation of organics but requires extensive off gas treatment facilities. Process vitrifies residue into final form.	LILW is currently treated in Switzerland at the ZWILAG facility, in Bulgaria at the KOZLODUY plasma plant , and in Russia by SIA RADON. Trialed in the UK with simulated wastes with pilot-scale plasma thermal treatment of simulated PCM waste which resulted in glass-like slag.	This same system allows processing of combustible materials and the melting of metallic parts, concrete and other solid matter. The burden on the off-gas can be considerable. Variable feedstock can challenge the operational stability on the plasma system.

Thermal treatment process based on thermal gasification (VTT)	The thermal gasification process developed by VTT is based on fluidized-bed (FB) gasification. Produces a fine dust, which is collected by high temperature filtration. In addition to filter dust, larger inorganic particles are removed from the process together with bed material. This mass stream consists primarily of bed material. In most cases filter dust and bottom ash are immobilized after waste treatment before final disposal.	Developed by VTT since the 1980s. Applied to the treatment of LILW containing organic matter (IEX or operational wastes, etc.).	Reduces the volume of organic radioactive waste (solid or liquid). The developed process is compact to allow co-location with waste production.
Joule-heated in-container vitrification (JHICV),	Known commercially as GeoMelt® or Perma-Fix Solutions. Liquid waste and glass-forming chemicals are mixed with soils and melted 1250–1500°C using graphite electrodes placed within the mixtures. A single-use, batch process where a refractory-lined vessel is used where the waste vessel acts as the melter and disposal container.	Developed by Veolia subsidiaries Kurion Japan, Veolia Nuclear Solutions, and Veolia Nuclear Solutions Federal Services, LLC. Trialed in the UK (NNL) with simulated waste, e.g., simulated mix of PCM, Magnox sludge, Simulant sand + clinoptilolite, Magnox sludge, and failing cemented products. Some engineering-scale testing for supplementary LAW treatment at Hanford	Maximum temperatures are generally limited to 1450°C with Inconel electrodes in a ceramic-lined vessel. Can directly treat large, irregular-shaped contaminated parts. Removes requirement to pour the glass. Similar off-gas challenges to joule heated systems.
In-can melting (ICM).	Known commercially as DEM&MELT. The waste (solid or liquid) is placed directly in the canister that will become its final storage container. The canister is then heated externally by a refractory-lined resistance furnace, and when fully melted the container is cooled, removed and sealed. Processing temperatures are relatively low (800–1100°C) to limit radionuclide (e.g., Cs and Sr) and chemical (e.g., S) volatility, but still allow waste stabilization in the form of a stable monolith. Suitable for wastes such as zeolites, silico-titanates, sands, ashes, and coprecipitation contaminated sludges.	Developed by a consortium of CEA, Orano, ANDRA, and ECM Technologies. CEA has a full-scale in-can vitrification demonstration tool (DEM&MELT). DEM&MELT has TRL = 7 and is fully qualified by the CEA to treat α-emitting liquid waste. Some development work has also been undertaken in Japan. Designed to treat ILW and HLW.	Simple and portable set-up. Ideal for small-batch demolition and decommissioning projects. Suitable for solid, liquid and complex wastes. Clogging, deposit formation, and dust entrainment are limited. Loss from volatility of radionuclides is very low though does need consideration. Allows crystallization in the product

Process for Incineration and Vitrification In-Can (PIVIC).	The waste (solid or liquid) including organics and metals are inserted into a vertical multi-part chamber. An oxygen plasma torch is used to incinerate the organic component at ~800°C. In the lower section, an induction melter heats the ceramic and metal fractions together. The In-Can Melter is a metallic crucible heated in a refractory furnace. The melt volume is electromagnetically stirred, as it consists of a lower layer of metal phase and upper layer of oxide 'slag' containing the other components. The final waste form is an in-can layered structure of a lower metal fraction and upper ceramic fraction.	Developed by the CEA.	Allows on-site processing. Can treat solid, liquid and highly mixed wastes, e.g., from decontamination and decommissioning work, where separation of materials is not desirable. Off-gas treatment systems must be designed to capture contaminants, producing secondary wastes. Final product can be glass, glass ceramic or simply a high-density waste product including a metal phase.
Hybrid System for Advanced Incineration Vitrification (Système Hybride d'Incinération Vitrification Avancé), SHIVA	An incineration-vitrification process using a cold wall direct glass induction melter and plasma burner. Specifically designed to operate in a hot cell for high or intermediate level waste. It allows, in a single reactor, waste incineration by plasma burner and ashes vitrification.	Developed by the CEA. A full-scale inactive pilot has been tested by the CEA since 1998 for various wastes. TRL of 5-6.	Suited for the treatment of organic (e.g., organic ion-exchange resins) and mineral waste with high alpha contamination and potentially high chloride or sulfur content. Can treat solid or liquid wastes but must not contain metals.

Table 2. Operational parameters of off gas treatment system of “Radon” LILW vitrification plant [80, 129].

Volumetric flow rate, m^3/h		Up to 100
Off gas temperature, ^{o}C	At the inlet	Up to 200
	At the outlet	Less than 50
Radioactive aerosols concentration, Bq/L	At the inlet	Up to 15
	At the outlet	Less than 0.015
Concentration of dust, mg/m^3	At the inlet	Up to 2000
	At the outlet	Less than 0.02
Concentration of NO_x, g/m^3	At the inlet	Up to 70
	At the outlet	Less than 0.01

Table 3. Some typical compositions of LILW glasses (wt.%)

Glass ID	ISG	LAW-A44	LRM	AG8W1	DG-2	SG	CSD-B	RBMK	WWER	N1	HM09-30MS	G11_D2	CORA	VP2	BV	ISV	M1	ALPS-40ox*
Refs.	[192]	[193]	[194]	[195, 196]	[195, 196]	[195]	[197]	[129, 131]	[129, 131]	[198]	[126]	[125]	[199]	[177]	[200]	[146]	[201]	[75]
	Durab. ref. (HLW)	Durab. ref. (LAW)	Durab. ref. (LAW	Durab. ref. (LLW)	Durab. ref. (LLW)	Comp. ref. (LLW)	Durab./ comp. ref. (LLW)	In use CCIM (Mixed waste)	In use CCIM (DAW)	In use plasma (SRW)	Exp. ILW (Magnox)	Exp. ILW (Resin)	Prop. CCIM (LLW)	Exp. LILW (Filters)	Prop. bulk-vit	Prop. in-situ vit	Exp. in contain vit	Exp. In-can vit (ALPS)
Country	Ref. HLW	US	US	Korea	Korea	Korea	France	Russia	Russia	Russia	UK	UK	Italy	China	US	US	Japan	Japan
SiO_2	56.0	44.6	54.9	43.1	41.2	37.5	50.3	48.1	46.8	30.6	33.6	50.0	42.0	49.0	46.3	64.7	44.7	35.5
B_2O_3	17.2	8.9	8.0	10.0	11.3	10.6	14.4	7.5	9.0		8.3	9.1	17.1	4.8	5.0	0.1	5.6	15.2
Al_2O_3	6.4	6.2	9.6	12.3	7.1	7.4	8.7	2.5	4.3	9.0	1.2	8.9	13.3	5.5	9.2	8.2	7.1	0.6
Li_2O			0.1	1.2	5.3	5.1	2.2				1.3	4.7					3.7	
Na_2O	12.2	20.0	20.3	17.6	10.1	4.5	12.6	17.2	24.8	3.6	3.1	15.5	15.7	10.1	20.0	15.4	13.0	12.4
K_2O		0.5	1.5	1.6	4.5	7.3		0.5	1.9	5.5	0.4	0.1		5.1	1.5	0.5		
MgO		2.0	0.1	2.1	4.6	2.2				5.9	10.8				1.3	0.4	13.0	12.6
CaO	5.0	2.0	0.6	4.8	9.8	18.1	3.1	15.5	6.2	13.8	4.0	0.2	1.5	20.1	2.7	5.5	8.0	16.5
SrO					0.1						0.2				0.03	0.01		0.8
BaO					0.1		0.4				21.0			3.1	0.1	0.01		
TiO_2		2.0	0.1	1.2	3.1					1.3					0.8	0.8	0.2	0.3
ZrO_2	3.3	3.0	0.9	0.9	1.1		2.0					0.2	2.0		7.0	0.1	2.2	
V_2O_5				1.4	0.1										0.01			
Cr_2O_3		0.02	0.2				0.1								0.1			
MoO_3		0.01	0.1				0.7											
MnO_2			0.1	0.1	0.2	0.3	0.2								0.1	0.01		
Re_2O_7		0.1																
Fe_2O_3		7.0	1.4	1.8	0.4	2.9	2.8	1.7	1.8	10.0	11.8	10.7	0.7		4.3	4.3	2.0	5.5
CoO				0.3	0.0		0.3						0.2					0.3
NiO			0.2		0.1		0.3	5.7	4.3				0.2					
ZnO		3.0			0.2	1.3								2.4				0.2
La_2O_3			0.02															
F		0.01	0.9												0.1			
Cl		0.7						0.7	0.6	1.1	0.6				0.2			0.2
SO_3		0.1	0.3				0.4	0.6	0.5	1.5	0.1	0.4	0.5		0.8			
P_2O_5		0.03	0.6	0.4	0.8		0.2			14.8			0.1		0.6			
Other			0.3	1.2	0.0	2.9	1.3			3.2	3.6	0.1	6.7				0.6	
TOTAL	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.1	100.0	100.0	100.0

* given as NaCl, **Given as Na_2SO_4; Durab. (durability); Ref. (reference); Comp. (composition); Prop. (proposed); Exp. (experimental); Vit. (vitrification)

Table 4: Illustration summary of non-radioactive Fukushima wastes vitrification tests processed in DEM&MELT pilot at CEA Marcoule

Type of waste	**Characteristic of waste, precursors/compositions/experimental conditions**	**Homogeneity/Volatility/Chemical durability**	**Ref.**
ALPS (Advanced Liquid Processing System-Multi Radionuclides Removal) **Ferric, magnesium oxides calcium carbonated slurries**  *Humidified mix of slurries*  Glass frit (SiO_2, B_2O_3 and Na_2O) *Fukushima Effluent Treatment Waste*	Elaboration temperature: 1100°C ≈ 280 kg of glass produced Feeding rate range tested is [5-10 ~ 15-28] kg/h *Dry composition of ALPS slurries, Wt.%* $CaCO_3$ 47.3 | Al_2O_3 0.9 $Mg(OH)_2$ 29.5 | $Co(OH)_2$ 0.5 Na_2CO_3 3.5 | $Ti(OH)_2$ 0.5 SiO_2 3.0 | $Zn(OH)_2$ 0.4 $SrCO_3$ 1.9 | $Ca(OH)_2$ 0.3 $FeO(OH)H_2O$ 12.0 | Cl 0.3	Waste loading: 40 wt.% oxides Homogeneous glass (with few crystalline phases) Radionuclide volatility: Sr <0.01 wt.% Chemical durability: Leaching test (MCC-1 type: 28 days, 90°C in pure water, S/V of 2 m^{-1})  Full-scale half-cut canisters	[75, 93, 94, 96]
Cs-loaded zeolites (Natural chabazite)  *Zeolites samples 0.7-2 mm*  Glass frit (SiO_2, B_2O_3 and Na_2O) *Fukushima Effluent Treatment Waste*	Elaboration temperature : 1100°C ≈ 260 kg of glass produced Feeding rate range tested is [6-20] kg/h *Composition of zeolite Wt.%, with Cs : 10 mg/g* SiO_2 51.5 Al_2O_3 17 CaO 5.5 K_2O 4.5 Fe_2O_3 3.0 MgO 1.3 Na_2CO_3 3.5 Others 13.7	Waste loading: 5 wt. % oxides Macroscopic homogeneous glass high retention of Cs Radionuclides volatility Cs: 0.08 wt.%  Full-scale Half-cut canister produced	[93, 94, 96]
Cs-loaded zeolites – mix zeolite, silicotitanate and sand  *Zeolites*  Silicotitanates *Fukushima Effluent Treatment Waste*	Elaboration temperature : 1100°C ≈ 270 kg of glass produced Feeding rate range tested is [6-20] kg/h *Silicotitanate : Na_2O-TiO_2-SiO_2 and Zeolites, Wt.%* SiO_2 51.5 Al_2O_3 17 CaO 5.5 K_2O 4.5 Fe_2O_3 3.0 MgO 1.3 Na_2CO_3 3.5 Others 13.7	Waste loading: 60 wt. % oxides Macroscopic homogeneous glass with high retention of Cs and Sr Radionuclides volatility: Cs: 0.04 wt.% Sr: 0.01 wt.%  Full-scale half-cut canister	[93, 94]

Table 5. Typical operational NPP LILW radionuclide composition [131].

Radionuclide, Bq/m^3	NPP with RBMK*			NPP with WWER**		
	Leningrad	Kursk	Chernobyl (Ukraine)	Kalinin	Kola	Novo-Voronezh
^{137}Cs	5×10^6	4.1×10^9	3.7×10^8	1.8×10^9	7.5×10^{10}	1.2×10^{10}
^{134}Cs	-	8.5×10^8	1.0×10^7	1.0×10^9	8.0×10^9	9.0×10^8
^{60}Co	2.8×10^7	5.3×10^7	n.m.	4.5×10^7	1.5×10^9	2.6×10^7
^{239}Pu	n.m.	1.5×10^5	n.m.	4.4×10^5	n.m.	n.m.

*RBMK – Reaktor Bolshoy Moshchnosti Kanalnyy; channel type uranium graphite reactor. **WWER – water-water energetic reactor, an analogue of western pressurized water reactors. N.m. – not determined.

Table 6. Operational data of CCIM LILW vitrification plant [129, 131].

Parameter / LILW source	**RBMK**	**WWER**
Vitreous waste form composition, wt.%	16.2Na_2O 0.5K_2O 15.5CaO 2.5 Al_2O_3 1.7Fe_2O_3 7.5B_2O_3 48.2SiO_2 1.1Na_2SO_4 1.2NaCl 5.6Misc.	24.0Na_2O 1.9K_2O 6.2CaO 4.3Al_2O_3 1.8Fe_2O_3 9.0B_2O_3 46.8SiO_2 0.8Na_2SO_4 0.9NaCl 4.3Misc.
Specific radioactivity, Bq/kg	7.0×10^6	2.7×10^6
Melting temperature, ˚C	1200 – 1250	1200 – 1250
Viscosity (Pa s) at 1200^{o}C	3.9	1.6
Specific resistivity (Ω m) at 1200 ^{o}C	0.028	0.027
Capacity by glass, kg/h	6.8-9.0	7.9-10.0
Specific capacity, kg/m^2 h	120-160	140-180
Specific power consumption, kW h/kg	5.0-6.0	4.5-6.0
Carry over of solid fraction, wt.%	1.0-1.2	1.0-1.2
Carry over of radionuclides (as ^{137}Cs),%	3.0-3.9	3-4

Table 7. LILW vitreous waste forms parameters [129, 131].

Parameter / Waste form, source		**Borosilicate glass, RBMK**	**Borosilicate glass, WWER**	**Glass composite material, LILW with sulphates**
Waste oxides loading, wt.%		30-35	35-45	30-35 and up to 15 vol.% of yellow phase
Density, g/cm^3		2.5-2.7	2.4-2.6	2.4-2.7
Compressive strength, MPa		80-100	70-85	50-80
Leach rate, g/cm^2×d after 28-th 62d doo leaching for given nuclide	^{137}Cs	$10^{-5} – 10^{-6}$	~10^{-5}	~10^{-5}
	^{90}Sr	$10^{-6} – 10^{-7}$	~10^{-6}	$10^{-6} – 10^{-7}$
	Cr, Mn, Fr, Co, Ni	~$10^{-7} – 10^{-8}$	~10^{-7}	~$10^{-7} – 10^{-8}$
	REE, Actinides	~10^{-8}	~10^{-8}	~10^{-8}
	Na	$10^{-5} – 10^{-6}$	~10^{-5}	$10^{-4} – 10^{-5}$
	B	$<10^{-8}$	$<10^{-8}$	$<10^{-8}$
	SO_4^{2-}	~10^{-6} at content less than 1%	~10^{-6} at content less than 1%	$10^{-4} – 10^{-5}$ at content less than 15%

Table 8. Glass types developed in the ROK and their properties

Glass ID	DG-2	SG	AG8W1	KEV-A PVC	KEV-A Borate	ISG-1
Waste streams	Dry active waste	Ion-exchange resins	Mixed LLW wastes	Polyvinyl chloride	Liquid borate	Reference
Melter type	CCIM	CCIM	CCIM	-	-	-
Current use	Yes - UVF	Yes - UVF	Yes - UVF	Proposed	Proposed	-
Reference	[195, 196]	[195, 196]	[195, 196]	[202]	[202]	[192]
Processing T_p (°C)	1150	1150	1150	1110	1110	1300
Waste loading (wt.%)	25	40	40	50	<15	-
Viscosity at T_p (poise)	10	4	67	20	30	6
Conductivity at T_p (S/cm)	0.46	0.40	0.31	-	-	-
PCT 7 d, total NL (g/m^2)	2	3	2	2	24	1
SiO_2 (wt.%)	41.2	37.5	43.1	35.1	47.6	56.4
Al_2O_3 (wt.%)	7.1	7.4	12.3	3.6	4.2	6.4
B_2O_3 (wt.%)	11.3	10.6	10.0	7.5	25.7	17.3
Total alkali oxide (wt.%)	19.8	16.9	24.6	11.6	19.5	13.0
Total alkali earth oxide (wt.%)	14.4	20.4	2.3	24.9	0.4	5.0
Transition metal oxide (wt.%)	4.9	6.0	8.4	17.1	2.6	2.1
Other (wt.%)	1.4	0.0	0.1	0.3	0.0	0.0