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RESEARCH ARTICLE - SPECIAL ISSUE - CHALLENGES AND RECENT ADVANCEMENTS IN NUCLEAR ENERGY SYSTEMS

Vitrification as a Key Solution for Immobilisation Within Nuclear Waste Management

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Abstract
Vitreous materials in the form of both relatively homogeneous glasses and composite glass crystalline materials (GCM) incorporating disperse crystalline phases are currently the most reliable wasteforms effectively used on industrial scale for nuclear waste immobilisation. Glasses are stable solid-state materials with a topologically disordered atomic structure in the form of solid solutions, i.e. solutions frozen via vitrification to a solid state without forming regular crystalline phases. Nuclear waste vitrification is attractive because of technological and compositional flexibility enabling hazardous elements to be safely immobilised and providing a glassy material characterised by high corrosion resistance, mechanical and radiation durability, as well as effectively reducing the volume of the resulting wasteform.

Keywords Nuclear waste · Immobilisation · Vitrification · Glass · Durability · Waste disposal

1 Introduction

Nuclear energy is statistically the cleanest source of power being also at the top level by its greenhouse gas emissions along with wind and solar power which are not only sustainable in practice but also far better compared with coal, oil, gas, and biomass burning as well as hydropower [1]. Figure 1 modified after the open access publication [1] is illustrating this being based on detailed analysis with normalisation of death rate data per 1 TW × h of energy produced and for CO_2 emissions per 1 GW × h of energy produced during the lifecycle of power plant of concern [1].

Nuclear energy has a potentially exceedingly valuable role to play in securing power supplies and can do it safely. Nuclear power gives both long-term security of supply against economic and political threats and short-term load following capabilities with maximised efficiency [2]. As the world is struggling to meet climate change goals, the IAEA concludes that the global nuclear power generation will need to significantly expand beyond its historical markets involving new countries interested in safe sources of energy [3]. This implies that building the trust of the public into nuclear energy usefulness and safety and an important way to do this is to demonstrate safe decommissioning of existing nuclear facilities including nuclear power plants and safe management of all nuclear waste produced during activities associated with use of nuclear energy. Nuclear (radioactive) waste is that waste that contains, or is contaminated with, radionuclides at activity concentrations greater than clearance levels which are established by the regulatory organisations. Naturally, radioactive wastes with activity concentrations below that levels are considered as non-radioactive, these are termed exempt wastes. The highest hazard for humans and environment poses the high-level waste (HLW).

Nuclear waste management at all stages of its utilisation and decommissioning has become one of the most important activities of nuclear industry [4]. While the end point of nuclear waste management is always disposal [4, 5], predisposal activities are needed aiming to prepare the waste properly for subsequent storage, transportation, and final disposal. Immobilisation of nuclear waste using durable materials which provide a high degree of retention of radionuclides plays a key role in predisposal activities [6, 7].

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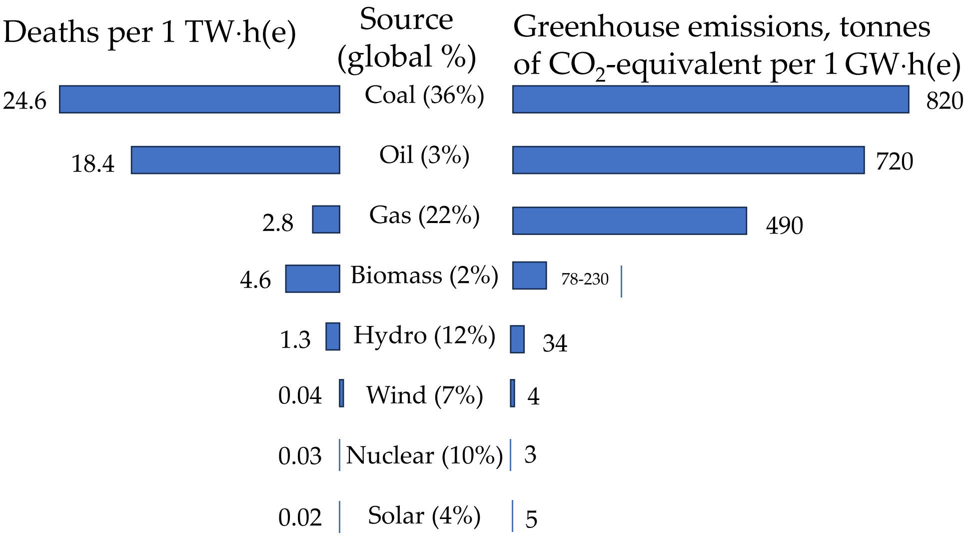


Fig. 1 Characterisation of energy sources by death rates caused by accidents and pollutions and greenhouse gas emissions. Modified after [1]

Selection of materials suitable for waste immobilisation is based on a multifactor analysis which assumes a graded approach with most durable materials such as silicate glasses and oxide ceramics aimed at most hazardous nuclear waste such as long-lived and highly radioactive waste—high-level waste (HLW) [8–16]. Nuclear waste glasses which immobilise the toxic radionuclides are produced via vitrification which envisages dissolution of toxic components into a liquid molten mass which being cooled below the so-called glass transition temperature T_g solidifies transforming into a durable solid material which retains the toxic radionuclides within its structure [17].

2 The Glass

Glasses are solid-state materials which have a topologically disordered internal atomic structure. There is a high similarity in the structural arrangements of glasses and liquids as both these distinctly different phases of matter are characterised by a topologically disordered structure despite of the fact that their properties are completely different, e.g. mechanically the vitreous phase is brittle whereas the liquid phase is plastic. This causes many discussions (Fig. 2a) and frequent attempts to rely on rheology rather than structure in defining the T_g below which the melt is transformed into a glass [19].

This unconformity is resolve utilising the mathematical apparatus of set theory. Indeed, utilisation of the set theory provides an unambiguous clarification of structural differences between glasses and melts based on their different Hausdorff–Besicovitch dimensionalities of glass ($D = 3$) and liquid ($D = 2.5$) bonding systems [19–21]. Upon heating, glasses continuously change most of their properties to those of a liquid-like state in contrast to crystals where such changes occur abruptly at a fixed temperature (the melting point). The solid-like behaviour of amorphous materials at low temperatures is separated from liquid-like behaviour at high temperatures by the glass transition temperature (T_g). Both glasses and melts (liquids) have an internal structure made of a network of interconnected structural blocks via interconnecting bonds however whether a material behaves as either liquid or solid depends on the connectivity between its elementary building blocks—molecules. Solids (glasses) are characterised by a high degree of connectivity, whereas structural blocks in liquids (melts) have a lower connectivity [22]. Figure 2b illustrates the obvious structural difference between glasses and liquids seen explicitly for bonding system—in the glass almost all bonds (apart from some point-like defects) are intact while the bonding system of a liquid is significantly disrupted. Kantor and Webman have proved in 1984 that the rigidity threshold of an elastic percolating network is identical to the percolation threshold [23] thus allowing to find the degree of disruption when a glass transforms to a liquid. Indeed, an increase of temperature causes the break of some of bonds between glass structural blocks (e.g. atomic or molecular species) so that broken bonds (termed configurons) start forming clusters; moreover, the higher the temperature the larger are the clusters made of broken bonds. Melting of an amorphous material occurs when the broken bonds form a percolation cluster, which penetrates the whole volume of the disordered network; moreover, formation of percolating clusters at glass transition temperature is evidenced both by experimental and computer modelling results [18–20].

The stability of glassy form of matter is of fundamental importance as disposal times span for HLW beyond million years [5]. Glasses always existed on the Earth, e.g. formed from volcanic melts that cooled rapidly being quite stable and durable to survive in a corrosive environment for many

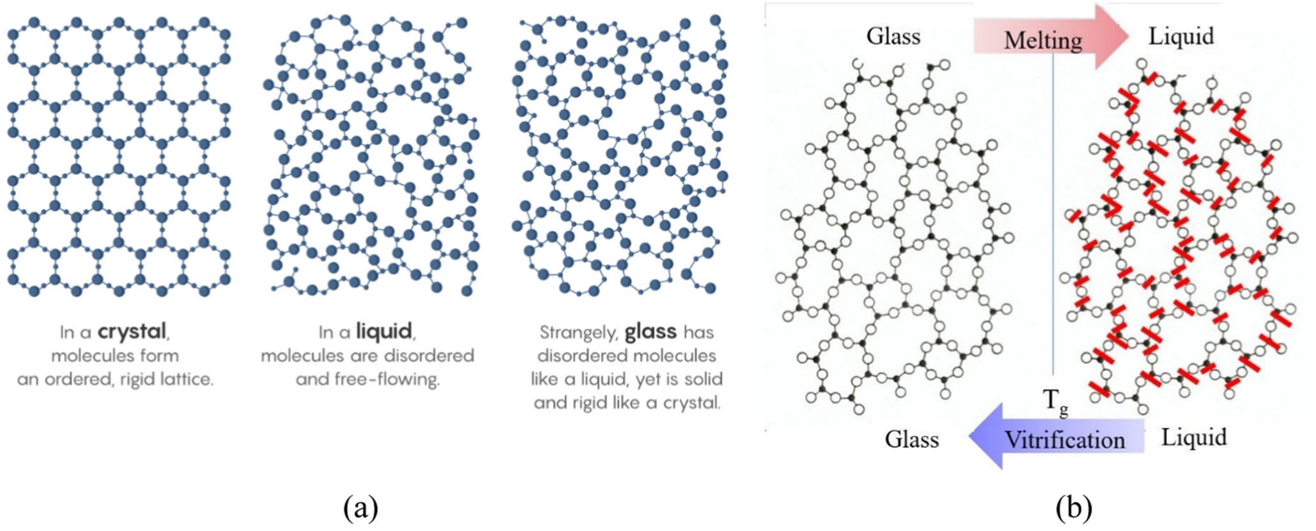


Fig. 2 **a** The structure of glass is different from that of a crystal however it looks similar to that of a liquid (see open access reference [18]). **b** Melting of glass (glass–liquid transition) occurs when bonds are broken (schematically shown by red dashes) while vitrification (glass formation) is due to creation of bonds between the atoms

millennia and millions of years. Natural glasses are found in nature almost unaltered such as obsidians (volcanic glasses), fulgarites (formed by lightning strikes), tektites found on land in Australasia and associated micro-tektites from the bottom of the Indian Ocean, moldavites from central Europe, and Libyan Desert glass from western Egypt which have been in the natural environment for many millions of years with extremely low alteration rates being less than a nm/year. The highly siliceous massive (thousand tonnes) vitreous deposits found in the volcanic caldera in Krasnokamensk, Russia, retain the radioactive uranium at concentrations exceeding 20 ppm at the age of about 140×10^6 years [24]. Although, compared to crystalline materials of the same composition, glasses are metastable materials, their relaxation to crystalline structures is kinetically impeded. Because of this phase changes cannot occur in practice, e.g. crystallisation would take in silica glass about 10^{98} years which significantly exceeds the lifetime of Universe being about 1.5×10^{10} years [25].

Except for alkali–aluminophosphate glass used in Russia, borosilicate glass has been universally selected as the vitreous wasteform to immobilise HLW as shown in Table 1.

However, some nuclear waste streams, such as legacy low- and intermediate-level radioactive waste (LILW) that was accumulated during completion of various nuclear development programmes, are preferably immobilised using bespoke selected silicate or phosphate systems that are better suited for given compositions of waste.

3 The Vitrification

The IAEA recommends vitrification for high-level waste conditioning as explicitly specified in the Table II-3 of Reference [4]. It should be also noted that there is a distinct correlation between the immobilisation technology used and the class of nuclear waste which reflects the graded approach on managing radioactive waste [8, 25]. The higher the nuclear waste hazard, the more durable materials are being used (Fig. 3).

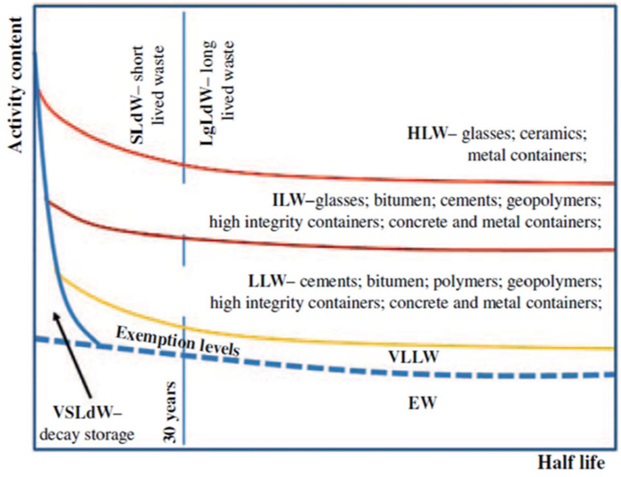


Fig. 3 Classification of radioactive waste following IAEA scheme [27] and preferred materials used for immobilisation. HLW—high-level waste, ILW—intermediate-level waste, LLW—low-level waste, VLLW—very low-level waste, EW—waste exempt of regulatory control. Reproduced from [25] with permission of Elsevier

Table 1 Typical composition of glasses for HLW immobilisation [26]

Country	Facility*	Composition of glasses by oxides**, wt%
Belgium	Pamela	$70.7P_2O_5\cdot7.1Al_2O_3\cdot22.2Fe_2O_3$ and $52.7SiO_2\cdot13.2B_2O_3\cdot2.7Al_2O_3\cdot4.6CaO\cdot2.2MgO\cdot5.9Na_2O\cdot18.7$ Misc
France	AVM	$46.6SiO_2\cdot14.2B_2O_3\cdot5.0Al_2O_3\cdot2.9Fe_2O_3\cdot4.1CaO\cdot10.0Na_2O\cdot17.2$ Misc
France	R7/T7	$54.9SiO_2\cdot16.9B_2O_3\cdot5.9Al_2O_3\cdot4.9CaO\cdot11.9Na_2O\cdot5.5$ Misc
Germany	Karlsruhe	$60.0SiO_2\cdot17.6B_2O_3\cdot3.1Al_2O_3\cdot5.3CaO\cdot7.1Na_2O\cdot6.9$ Misc
India	WIP	$30.0SiO_2\cdot20.0B_2O_3\cdot25.0PbO\cdot5.0Na_2O\cdot20.0$ Misc
India	AVS	$34.1SiO_2\cdot6.4B_2O_3\cdot6.2TiO_2\cdot0.2Na_2O\cdot9.3MnO\cdot43.8$ Misc
Japan	Tokai	$46.7SiO_2\cdot14.3B_2O_3\cdot5.0Al_2O_3\cdot3.0CaO\cdot9.6Na_2O\cdot21.4$ Misc
Russia	EP500	$53.3P_2O_5\cdot15.8Al_2O_3\cdot1.6Fe_2O_3\cdot23.5Na_2O\cdot5.8$ Misc
UK	WVP	$47.2SiO_2\cdot16.9B_2O_3\cdot4.8Al_2O_3\cdot5.3MgO\cdot8.4Na_2O\cdot17.4$ Misc
USA	DWPF	$49.8SiO_2\cdot8.0B_2O_3\cdot4.0Al_2O_3\cdot1.0CaO\cdot1.4MgO\cdot8.7Na_2O\cdot27.1$ Misc
USA	WVDP	$45.8SiO_2\cdot8.4B_2O_3\cdot6.1Al_2O_3\cdot11.4Fe_2O_3\cdot1.4MgO\cdot9.1Na_2O\cdot17.8$ Misc
USA	WTP	$50.0SiO_2\cdot20.0B_2O_3\cdot5.0Al_2O_3\cdot25.0Na_2O$

*DWPF, Defence Waste Processing Facility, Savannah River Site, USA; SRNL, Savannah River National Laboratory, USA; WVP, Waste Vitrification Plant, Sellafield, UK

**_Misc_Miscellaneous, including oxides of radioactive waste

In addition to its high durability, glass can incorporate a vast range of elements into its structure. Indeed, Michael Faraday described the glass as “a solution of different substances one in another”. Glasses are highly tolerant to compositional changes and characteristic properties of glasses change continuously with variation of composition. Because of this, vitrification is little sensitive to slight compositional variations typical for most waste streams. There is a clear trend currently of using glass crystalline (composite) materials (GCM) containing both crystalline and amorphous phases rather that homogenous glasses [6, 8]. Glasses are optimal immobilising materials to ensure safe long-term storage, transportation, and consequent disposal of nuclear waste due to their physical and chemical durability combined with their high capability to incorporate into their structure most elements. The main advantages of radioactive waste vitrification process are as follows [8, 25]:

1. High capability of glass to immobilise various elements.
2. Simple production technology adapted from glass production industry.
3. Relatively small volume of the resulting vitreous material.
4. High chemical durability of glasses in natural waters, and
5. High tolerance of glasses to radiation damage.

Vitrification involves melting of wastes with glass-forming additives so that the final vitreous product incorporates the waste contaminants in its macro- and microstructure [17]. The vitrification process generally comprises evaporating the water from liquid waste, decomposing the volatile anions (e.g. nitrates), fusing the waste with oxide glass additives until they form a homogeneous liquid, pouring the melt into canisters and cooling to form the solid glass containing the waste components. The thermal energy required for the conditioning of 1 L of typical commercial HLW containing 120 g of salts is roughly 1.2 kW × h. The major portion of this energy of about 67% is required for evaporation. The energy required for decomposition of nitrates is about 20% and for fusion is about 13% of the total. Two basic types of vitrification technologies which are being used industrially are either the one-stage process in which both waste calcination and melting occur in the melter (Fig. 4a) or the two-stage process when the waste is calcined prior to melting (Fig. 4b) [28].

In the one-stage process (Fig. 4a), the excess water is first evaporated from the waste using apparatuses such as thin film evaporators. The remaining salt concentrate is then mixed with the required additives aiming to obtain a durable final glass product. The glass-forming batch is thus produced in the form of a wet paste on mixing, which is then pumped into the melter where it is subject to further water evaporation, calcination with decomposition of salts and complete melting. The glass-forming melt is finally poured from the melter into metallic containers (canisters) where it forms a solid glass on its cooling down the glass transition temperature T_g. In the two-stage vitrification process schematically shown in Fig. 4b, the calcination of the waste concentrate after evaporation occurs in a separate device—calciner which operates

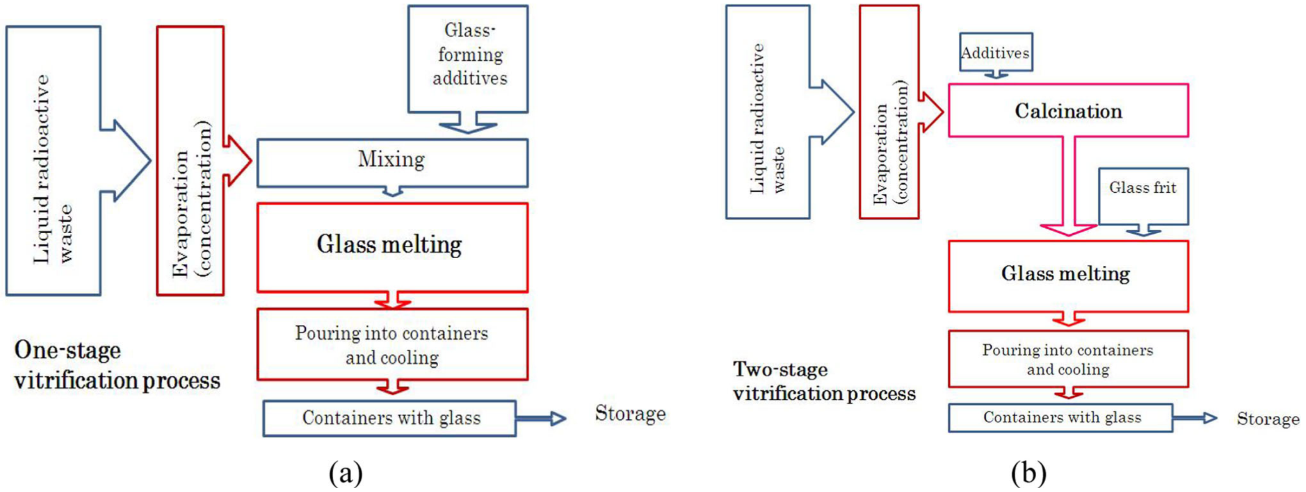


Fig. 4 Schematic of one-stage (**a**) and two-stage **b** technological processes used for nuclear waste vitrification

at much lower temperatures compared melters [25]. After that the calcine is fed into the melter together with required glass-forming additives typically in the form of glass frit. In the melter, similar to the one-stage vitrification process, a glass-forming melt is produced although the sizes of melters in the case of two-stage vitrification are considerably smaller as shown in Fig. 5 which shows the vitrification technology used to immobilise HLW in the UK.

The main types of melters include ceramic, metallic and cold crucible melters. Metallic melters have the advantages of high thermal conductivity, thermal shock resistance and small volume. Their disadvantages include a lower temperature of operation, microstructural changes at high temperature creating failure modes, welding flaws, and, in the case of induction heating, the potential for hot spots. Cold crucible melters provide frozen glass contact materials maintained by active cooling which separates the melt from crucible walls. Their advantage is very high-temperature process capability, relatively low volume and tolerance to corrosive melts. There is a distinct trend currently of using pot (in-can) method of vitrification for immobilisation of legacy and decommission radioactive waste which envisages mixing of waste with glass-forming additives and feeding the mixture at a constant flow rate directly into vessels–crucibles where water evaporation, calcination and vitrification occur [30]. Refractory canisters–containers are used to ensure containment of radioactive waste during processing (glass melting), storage, transportation and disposal. In the in-can vitrification process, the melting pot is disposable and serves as the primary canister for both metallic and vitreous wasteforms.

Hazardous waste radionuclides are finally immobilised in the vitreous materials either by direct incorporation into the glass molecular structure or by encapsulation when the waste components such as refractory crystals are surrounded by the glass matrix. Vitrification is one of the best solutions in providing safety of nuclear waste storage, transportation and final disposal: France, India, Japan, Russia, Slovakia, South Korea, the UK and USA have operating vitrification facilities used to immobilise both HLW and (LILW), with Germany having recently completed their HLW vitrification programme (Table 2) [8].

The hazard of waste $H(t)$ is characterised by the ingestion radiotoxicity which is defined as the volume of drinking water which potentially can be contaminated above the allowable level of radionuclides IL_i: $H(t) = \Sigma[C_i\ (0)\ \exp(-\ \lambda_i t)\ \Phi_i V/\mathrm{IL}_i]$, where C_i (0) is the initial concentration of i-th radionuclide, λ_i is the decay constant, t is time, V is the initial volume of waste and the dimensionless factors $\Phi_i \leq 1$ are equal to the released inventory fractions for each radionuclide from the given type of wasteform used. The toxicity of HLW resulting from reprocessing of nuclear fuel used during one year at a typical 1 GW nuclear power plant is immense with potentially contaminated water volumes exceeding 10^{12} m^3 (e.g. the volume of lake Michigan in the USA). Vitrification reduces the risk of contamination to levels close to almost zero [25]. Waste vitrification also provides a high waste volume reduction [25], because of that the overall cost of vitrified radioactive waste is usually lower compared with alternative options specifically with due account of storage, transportation and disposal expenses.

4 The Durability

Glass was selected as a nuclear waste immobilising matrix (wasteform) due to its both compositional flexibility and high resistance to corrosion in aqueous systems which ensures the retention of radionuclides over required time frames which in the case of actinides extend over many hundreds thousand

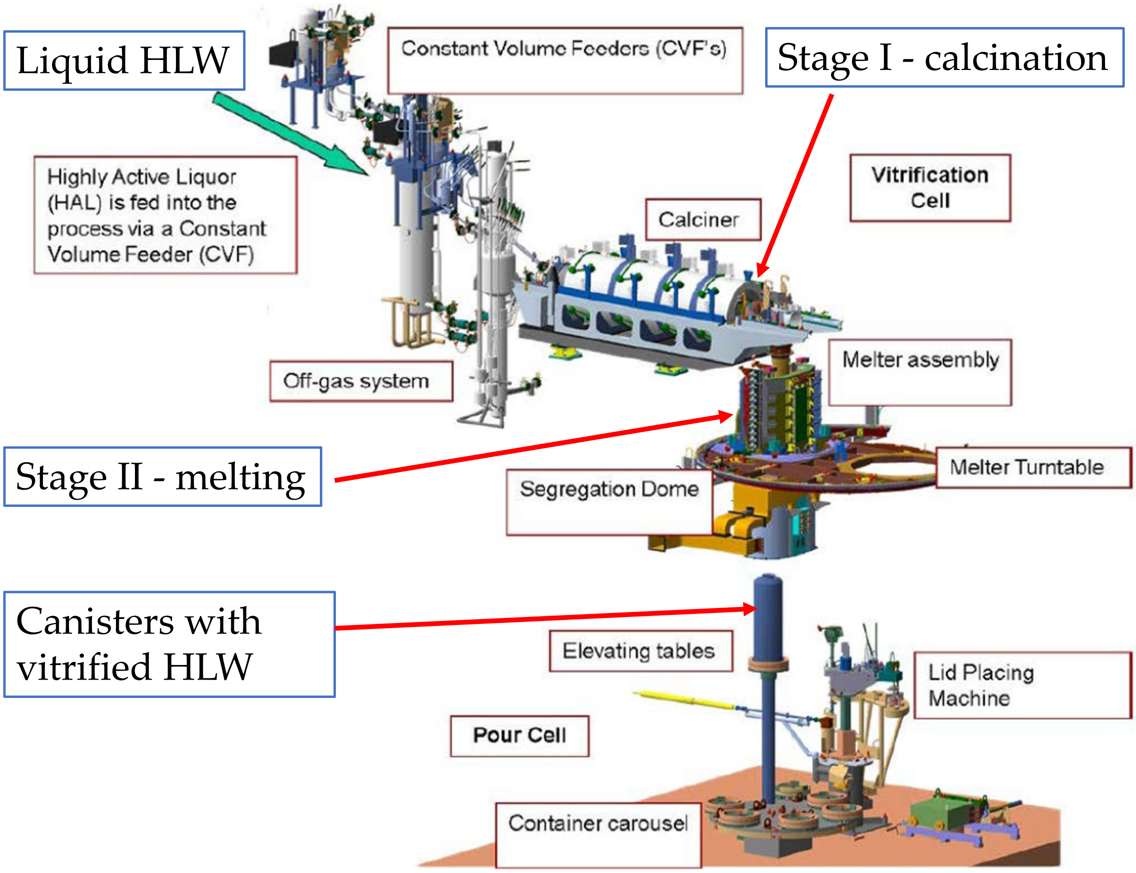


Fig. 5 Schematic of the two-stage vitrification process used in the UK at the waste vitrification plant (WVP). Adapted from [29], publicly available

Table 2 Successful industrial implementation of HLW vitrification in the world

Country	Vitrification facilities	Operating years reported	Immobilised activity, TBq	Total produced quantity, tonnes
France	R7/T7 (La Hague), AVM (Marcoule)	1978–2019	$29\cdot10^7$	8252
USA	DWPF (Savannah River), WVDP (West Valley), WTP (Hanford)	1996–2012	$2.7\cdot10^6$	7870
Russia	EP-500 (MCC, Mayak)	1987–2020	$28\cdot10^6$	7700
UK	WVP (Sellafield)	1990–2012	$33\cdot10^6$	2200
Belgium	Pamela (Mol)	1985–campaign completed	$0.5\cdot10^6$	500
Japan	TRP (Tokai)	1995–2007	$14.8\cdot10^3$	70
Germany	VEK (Karlsruhe)	2010–campaign completed	$0.8\cdot10^6$	55
India	WIP (Tarapur), AVS (Tarapur), WIP(Trombay)	2012	$9.62\cdot10^3$	28

and even millions of years [5]. Corrosion of glass is a complex process dependent on glass composition, environmental conditions, and time [12, 15, 16, 25] being investigated as a function of glass composition, temperature, pH, and solution chemistry [31] for both alkali–silicate [12–16, 32–36] and phosphate glasses [37, 38]. Table 1 reveals that most of glasses currently used to immobilise the nuclear waste are of alkali–silicate family. The corrosion of alkali–silicate glasses in water occurs via two main mechanisms, namely the diffusion-controlled ion exchange (IEX) and the hydrolysis (labelled as H) [25, 39]. The IEX mechanism involves the mutual diffusion and exchange of a cation (M^+) in glass with a proton or hydronium (H_3O^+) from water via the reaction $(\equiv\mathrm{Si{-}O{-}M})_{\mathrm{glass}} + H_2O \leftrightarrow (\equiv\mathrm{Si{-}O{-}H})_{\mathrm{glass}} + \mathrm{M{-}OH}$. The IEX mechanism almost completely controls the initial release of cations (radionuclides) and at relatively low ambient temperatures and not extremely high pH can dominate over the hydrolysis for many hundreds or even thousands of years [40]. Ion exchange reactions selectively extract cations from the glass being characterised by the normalised leaching rate NR_{xi} ($\mathrm{g/cm^2 \cdot day}$):

$$
\begin{aligned}
NRxi &= \rho(Di/\pi t)\,1/2 \\
&= \rho 10 - pH/2[\kappa D_{0H}/Ci(0)\pi t]\,1/2\exp\left(-Edi/2RT\right),
\end{aligned}
$$

where ρ ($\mathrm{g/cm^3}$) is the glass density, D_i ($\mathrm{cm^2/day}$) is the effective interdiffusion coefficient, E_{di} (J/mol) is the interdiffusion activation energy, R is the universal gas constant $R = 8.314$ J/mol, T (K) is the temperature, D_{0H} ($\mathrm{cm^2/day}$) is the pre-exponential coefficient in the diffusion coefficient of protons (hydronium ions) in glass, C_i (0) (mol/L) is the concentration of cations at the surface of the glass and κ is a constant that relates the concentration of protons in glass to the concentration of protons in water, i.e. to the pH of the solution. NR_{xi} therefore decreases exponentially with pH of contacting water solutions. The hydrolytic near-surface reactions of hydroxyl ions with the glass network destroys it which leads to a congruent dissolution of all glass components into the contacting water and finally leads to deposition of silica gel layers on the glass surface. The hydrolysis proceeds through the reaction: $(\equiv\mathrm{Si{-}O{-}Si}\equiv)_{\mathrm{glass}} + H_2O \leftrightarrow 2(\equiv\mathrm{Si{-}O{-}H}$ leading to a complete dissolution of the topologically disordered glass network with formation of ortho-silicic acid H_4SiO_4. The hydrolytic dissolution of alkali–silicate glasses is characterised by the normalised dissolution rate NR_H ($\mathrm{g/cm^2 \cdot day}$):

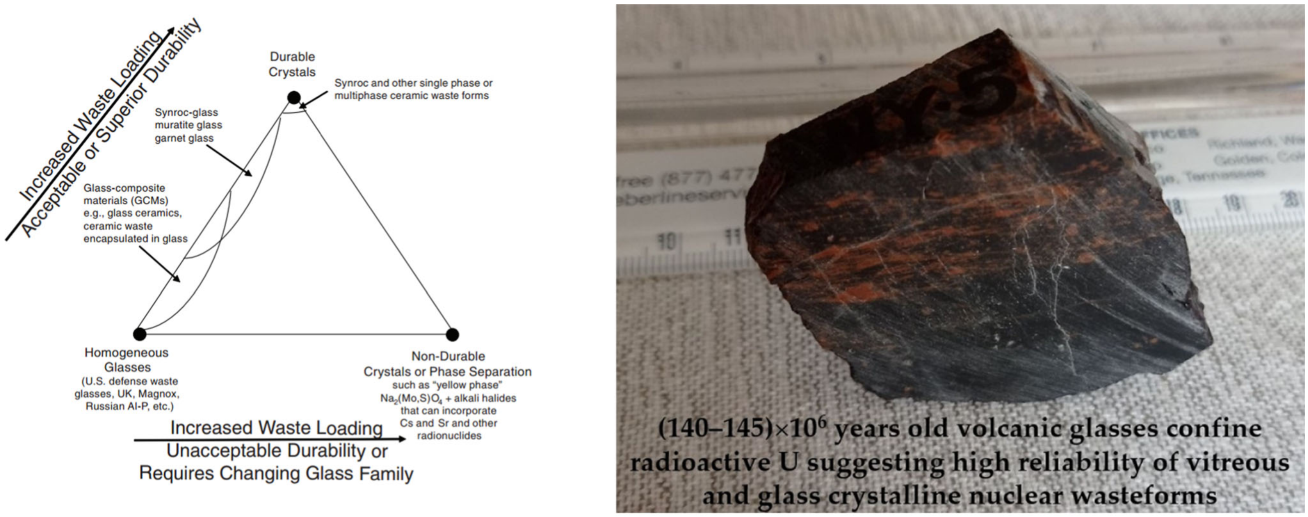
$$
NR_H = \rho r_c = \rho k a^{-\eta}\left[1-(Q/K)^{\sigma}\right]\exp(-E_{\mathrm{a}}/RT)
$$

where r_c is the linear rate of dissolution (cm/day), k is the characteristic rate constant, a is the activity of hydrogen cations, η is the pH-dependent exponent ($\eta \sim 0.5$), E_{a} is the activation energy of hydrolysis, Q is the product of the ionic activity of the rate control reaction, K is the equilibrium constant of this reaction and σ is the order of the reaction. Although the rate of hydrolytic dissolution of glass does not depend on time, it increases exponentially with the pH of the contacting water. Overall the corrosion rate NR_i normalised with respect to the i-th of component of glass is given by the sum of the contributions from the IEX and hydrolysis plus the contribution from the instantaneous dissolution of surface-bound radionuclides $\mathrm{NR}_i = \mathrm{NR}_{Xi} + \mathrm{NR}_H + N\Phi_{si}$, where $N\Phi_{si}$ accounts for instantaneous dissolution [25, 40]. The analysis shows that the typical rate of corrosion via the hydrolysis (r_{c}) of alkali–borosilicate glasses used to immobilise nuclear waste is about 10^{-7} m/y [41]. Generically, the dependence of the glass corrosion rate NR_i on the pH of contacting water is given by a U-shaped curve with the minimum corrosion rate located about pH 7, i.e. for near-neutral solutions [25]. Glasses in a deep geological environment are typically in contact with saturated solutions, therefore $\mathrm{Q} \to \mathrm{K}$ and thus $\mathrm{NR_H} \to 0$ which predetermines the IEX as the main mechanism of long-term corrosion [25, 34, 40, 42]. As the IEX is characterised by leaching rates NR_{xi} which decrease with inverse square root of time (proportionally to $t^{-1/2}$), the total inventory of radionuclides which could be potentially mobilised and migrate into the environment is limited and enables a reliable confinement of radionuclides until their natural decay [15, 25, 40]. Without contact of glass with water, the radionuclides including actinides will remain retained within the glass body structure.

The vitrification provides immobilisation of hazardous waste radionuclides either by direct incorporation into the glass network structure at molecular scale or by encapsulation when the waste components such as refractory crystals are surrounded by the glass matrix, i.e. the wasteform in this case is a GCM. The recent trend in the vitrification technology is on using GCMs which provide superior waste loading and durability compared to fully homogeneous vitreous materials [7, 8, 32, 43–45], which is illustrated by Fig. 6a. Figure 6b shows an example of a prehistorical GCM in the form of silicate glass containing crystalline particles and retaining primordial U and Th naturally occurring radionuclides over hundred million years[24].

5 Conclusions

Nuclear waste vitrification is a mature technology which provides a reliable immobilisation of waste toxic constituents and reduces the volume of waste to be disposed of. The glass produced has a high corrosion resistance, mechanical and radiation durability. Borosilicate and to a lesser extent phosphate glasses are the overwhelming worldwide choice for the immobilisation of high-level radioactive wastes (HLW) resulting from nuclear fuel reprocessing and low- and intermediate-level radioactive wastes (LILW) such as



(a) (b)

Fig. 6 The phase assemblage and durability wasteforms (**a**) [32]; the image of an ancient GCM—a volcanic glass confining the natural radioactive uranium and thorium for over 140–145 million years (**b**) [24]

the legacy waste and the waste resulting from operation of nuclear power plants. Continued advances in glassy wasteforms and nuclear waste vitrification technologies will be keys in enabling widespread deployment of nuclear energy.

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