Granite Recrystallization – The Key to an Alternative Strategy for HLW Disposal?

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ABSTRACT

An alternative strategy is proposed for the disposal of spent nuclear fuel (SNF) and other forms of high-level waste (HLW) whereby the integrity of a mined and engineered repository for the bulk of the waste need be preserved for only a few thousand years. This is achieved by separating the particularly problematic components, notably heat generating radionuclides (HGRs) and very long lived radionuclides (VLLRs) from the waste prior to disposal. Such a solution requires a satisfactory means of disposing of the relatively minor amounts of HGRs and VLLRs removed from the waste. This could be by high-temperature very deep disposal (HTVDD) in boreholes in the continental crust [1,2]. However, the viability of HTVDD, and hence the key to the entire strategy, depends on whether sufficient melting of granite host rock can occur at suitable temperatures and whether the melt can be completely recrystallized. The high-temperature, high-pressure experiments reported here demonstrate that granite can be partially melted and completely recrystallized on a time scale of years, as opposed to millennia as widely believed. Furthermore, both can be achieved at temperatures and on a time scale appropriate to the disposal of packages of heat generating HLW. It is therefore concluded that the proposed strategy, which offers, environmental, safety and economic benefits, could be a viable option for a substantial proportion of HLWs.

INTRODUCTION

Most countries with inventories of HLW are looking to eventual disposal in so-called "deep", mined and engineered repositories utilising the multi-barrier approach. While designs differ in detail, they almost all involve the wastes being encapsulated in metal and cement before being sealed into a system of tunnels, caverns and silos at depths between 300 and 1000 metres, i.e. in geological terms they are quite shallow. The technical problems associated with such repositories are well known and can have serious consequences, e.g. the failure to progress an ILW repository in the UK in the late 1990's. Most of the difficulties arise either from the siting of the repository within the zone of active near-surface groundwater circulation or from the inherent properties of the HLW itself (and frequently both).

The HLW based problems are essentially of two kinds. Firstly, the waste contains substantial amounts of heat generating radionuclides (HGRs) which puts constraints on the design and materials of the repository and, more importantly, necessitates that the HLW be packaged in relatively small units. Further, each of these units requires its own multi-barrier system and they have to be dispersed throughout a large volume of host rock to avoid potentially damaging temperature rises. Secondly, the waste contains sufficient quantities of very long lived radionuclides (VLLRs) to necessitate that it be isolated from the biosphere for up to a million years. Since geological, climatic and other conditions can not be predicted so far into the future,

it is, to say the least, extremely challenging to make the performance assessments and safety cases frequently needed for political acceptability and widespread public confidence.

The heating problem can be ameliorated by allowing HLWs, such as spent reactor fuel, to 'cool' for a period (usually a few decades) prior to disposal but this is only a partial solution as many of the HGRs are also long or very long lived.

AN ALTERNATIVE STRATEGY

The serious HGRs and problematic VLLRs, such as Np, Cm, Am, Tc etc., are a volumetrically small part of HLW/SNF. If these and possibly Pu (should it be designated as waste) could be removed from the waste and dealt with separately, disposal of the remaining bulk of the HLW in a mined and engineered repository would be greatly simplified in every respect.

Separation of the components of HLWs, other than the recovery of U and Pu through reprocessing, has tended to be dismissed in the past, not least because of the inevitable generation of further secondary wastes. However, recent speculation about partitioning and transmutation as a way of dealing with the troublesome VLLRs, especially the minor actinides, has awakened interest in both the possibility and technology of separation. Irrespective of the arguments for and against transmutation, the readiness of the nuclear industry to seriously contemplate partitioning of HLW/SNF opens up the prospect of attractive alternative strategies for HLW management. Perhaps foremost among these is the "under 10,000 year repository" which is used to dispose of the bulk of the waste, including U (unless reprocessing).

The "under 10,000 year repository"

The absence of the more problematic VLLRs from HLW would reduce the period for which the waste needs to be isolated from the biosphere to a few thousand years. This radically changes the entire scenario since making performance assessments and safety cases for an "under 10,000 year repository" is well within the predictive capabilities of contemporary geological and engineering sciences.

The bulk of the HLW minus its HGRs (and VLLRs) can be disposed of in the repository without the need to disseminate small amounts, each with its own near-field barrier system, throughout a large volume of rock. Hence, more massive and secure barriers could be constructed around quite substantial masses of HLW while significantly reducing the overall size and cost of the repository and the ensuing environmental disruption.

In the absence of reprocessing, much of the HLW/SNF is likely to be uranium oxide. The inclusion of this (despite its radioactive longevity) with the bulk of the waste in the repository is unlikely to constitute significantly more of a problem than do the natural uraninite and pitchblende deposits that have remained stable in the upper regions of the Earth's crust over millions of years [3].

To enable the option of the "under 10,000 year repository" it is necessary to find a safe and acceptable means of disposing of the relatively small quantities of HGRs and VLLRs removed from the HLW before consigning it to the repository. The remainder of this paper attempts to show that not only does such a means exist but that it is perfectly feasible in terms of the underlying science.

High temperature very deep disposal (HTVDD)

Disposal of radioactive waste in deep or very deep boreholes in the continental crust offers many advantages [4] and several schemes have been proposed for the secure disposal of small to moderate volumes of HLW [e.g., 1,2,5,6,7]. Possibly the most robust and potentially safest of these is a proposed high temperature, encapsulated, very deep disposal [2] that could be well suited to the HGRs and VLLRs separated from HLW/SNF.

In this scheme special cylindrical containers filled with heat generating HLW are deployed in the lower reaches of a 4 – 5 km deep, large-diameter (0.5 – 0.8 m) borehole sunk into granitic continental crust. Energy from the radioactive decay of the waste heats the packages to temperatures in excess of 850°C, which are sufficient to generate a substantial zone of partial melting in the adjacent granite. As the heat output gradually declines this melt cools and recrystallizes to seal the waste packages into a sarcophagus of solid crystalline granite surrounded by zones of thermally metamorphosed rock. In the latter any fractures, whether pre-existing or created during heating, are annealed by recrystallization or sealed by hydration mineralization.

Locating long lived heat sources in host rocks that almost certainly contain saline fluids will inevitably affect the surrounding environment. Effects are likely to include elevated temperature distribution and heat flow, enhanced solution/precipitation, fracture fluid flow and convection. Such effects have been discussed and, in some cases, modelled [2] and predicted to be relatively insignificant in the context of the viability and safety of the disposal scheme.

This HTVDD differs from earlier 'deep rock melting' schemes in that the waste itself does not become incorporated into the re-solidified host rock and no unconstrained sinking of the packages [7] occurs. The major benefit of such deep disposal is that it can locate the waste in a host rock in which any hydrous fluids present in fracture systems have been physically and chemically isolated from near-surface groundwaters for millions of years – a situation that is not likely to change in the next 10^5 years. Consequently, even if the near-field barriers of waste form, container, granite sarcophagus and cocoon of metamorphosed rock were all to be breached, the geological barrier constitutes an enormous safeguard.

For this scheme to work fully two things are crucial. First, sufficient melting of the granite must occur at temperatures low enough to preserve the integrity of the containers and, second, the melt must be able to crystallize completely to a holocrystalline rock. Both have to happen on a time scale appropriate to the thermal decay of the HLW.

EXPERIMENTAL RESULTS

From what is known about melting kinetics in granitic systems it is unlikely that much of the partial melting generated in the disposal scenario would be equilibrium melting. Even more crucially, almost nothing is known about how quickly granitic melts can be cooled and still give rise to a wholly crystalline rock. Solidification rates for natural granites have been deduced from the times taken for the intrusions in which they occur to cool from their emplacement temperatures but these times (derived by a number of methods) mostly indicate cooling rates between 10°C and 500°C per million years. This has led to the view that granites can only form by extremely slow crystallization over thousands, if not millions, of years despite there being

good geological evidence that acid magmas can be completely crystallized at cooling rates orders of magnitude faster.

To test this, and hence the feasibility of the disposal scheme, a series of melting and recrystallization experiments [8, 9] were carried out on an S-type granite (E93/7) of Caledonian age from the north of England under the conditions predicted to arise in the disposal scenario.

Partial melting of granite

The melting relations of the granite were determined for H_2O-saturated and undersaturated conditions at a pressure of 0.15 GPa, corresponding to the likely pressure at depths between 4 and 5 km in continental crust. Starting materials were normally powdered rock and total H_2O contents varied from 0.58 wt.% (= the natural H_2O^+ content of the granite) to 10.6%. Hence, many of the data are for the undersaturated conditions appropriate to the disposal scheme [2]. For a number of practical reasons, isothermal experiments in which the sample was heated as quickly as possible to the target temperature were used instead of the continuous heating over months that would occur in an actual disposal. On the time scale of the experiments (200 – 2650 hours) this will result in a slight underestimation of the amount of melt generated at any temperature compared with the simulation of disposal heating but it is not significant. The results of these experiments are shown as a phase assemblage diagram in figure 1.

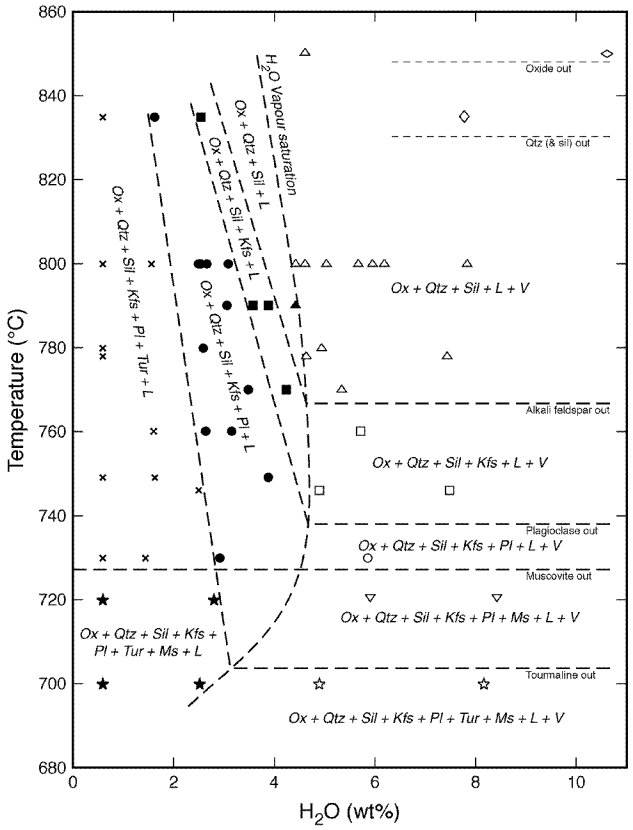

Figure 1. Phase assemblage diagram for granite E93/7 at 0.15 GPa (see [8] for details).

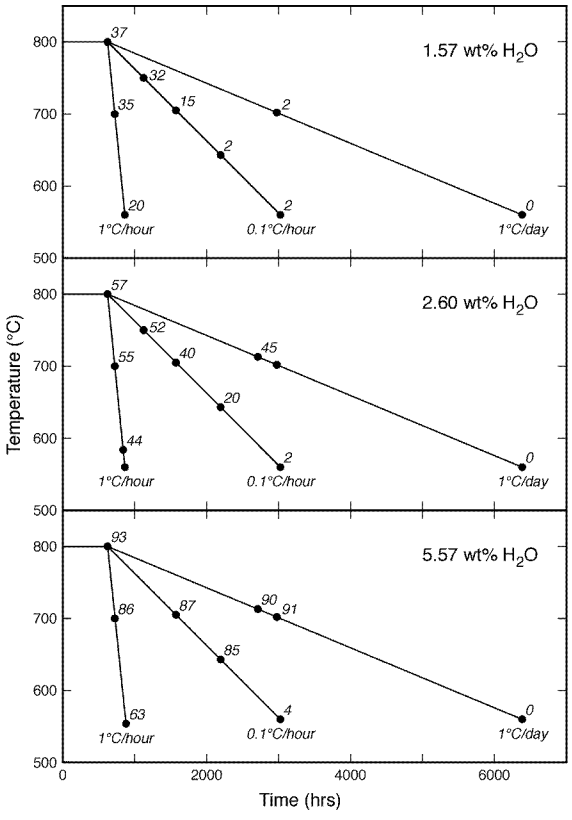

Figure 2. Recrystallization experiments on granite E93/7 for different H_2O contents & cooling rates (see text).

Melting begins just below 700ºC irrespective of H_2O content. For undersaturated melting above the solidus there is a positive correlation between the amount of melt generated and H_2O content but once vapour saturation is attained the amount of melt becomes independent of H_2O content. Figure 1 is close to being an equilibrium melting diagram in its higher temperature and H_2O content regions but the melts produced at lower temperatures are a long way from equilibrium. However, it must be born in mind in the context of HTVDD that it is not equilibrium melting that would occur during heating of granite by packages of HLW but prograde partial melting similar to that generated in the experiments.

Some of the experiments were duplicated using solid rock instead of powder as starting material. E.g., the 800ºC, 2.58% H_2O experiment in figure 1 closely approached equilibrium after 570 hours and generated 60% of melt (by volume). Repeating this experiment with a 20 mm x 7 mm diameter core of granite produced the same phase assemblage but with only 40% of melt and clearly did not attain equilibrium. The melting was initiated around the outer surface of the core and, as it progressed, it permeated right through the rock to produce an interlocking network of liquid. Considering the difference between sub-90 micron powdered granite and solid rock from the standpoint of reaction kinetics, it is remarkable that as much as 40% of melting took place at 800ºC in only 570 hours and it is extremely encouraging for the HTVDD concept. Even more significant are that melting migrated rapidly through the solid rock and that well over 50% of melting is likely to be achieved in under 1000 hours.

Granite recrystallization

In the 0.15 GPa recrystallization experiments (illustrated schematically in figure 2) samples of powdered granite with total H_2O contents of 1.57%, 2.6% or 5.57% were held at 800ºC for 624 hours before cooling at one of the three linear rates indicated. In order to follow the progress of recrystallization, experiments were quenched from the points shown in figure 2 and the volumes of melt remaining (italic numbers in figure 2) were determined by modal analysis.

To overcome the well-known problems of nucleation from feldspar bearing [10] and granitic [11] liquids, seed crystals have to be present in the melt. Experimentally, this is normally achieved by incorporating large pieces of the seed phase in the starting material [12]. However, this was not necessary in the present case since only a partial melt is generated at 800ºC after 624 hours for all the H_2O contents concerned, i.e., seeds were already present in the form of relic crystals. This is exactly as it would be in the HTVDD scheme.

For full details of these experiments the reader is referred to the papers by Attrill and Gibb [8, 9]. One of the more significant results is that the temperature at which any of the main minerals (feldspar, quartz and biotite) begins to crystallize during linear cooling is always lower than its melting temperature (figure 1). The difference is a function of the cooling rate and is directly proportional to the H_2O content, which effectively suppresses crystallization. An important consequence of this is that complete recrystallization does not occur until well below 700ºC, i.e. below the solidus, for the cooling rates studied. For example, during cooling at 0.1ºC per hour the last trace of melt does not disappear until 640ºC with 1.5% H_2O, until 560ºC with 2.5% H_2O, and until even lower temperatures with higher H_2O contents. Nevertheless, complete recrystallization of the granitic partial melts was achieved in the experiments for cooling rates of 0.1ºC per hour and slower.

DISCUSSION

To be suitable for HTVDD a host rock must undergo enough melting (possibly > 50%) at a temperature below ~850ºC under the conditions appropriate to the disposal. Further, recrystallization must be achievable at cooling rates faster than those which would prevail in the zone of partial melting around a disposal as a result of the thermal decay of the waste.

The experiments demonstrate that a typical S-type crustal granite can be melted to yield suitable amounts of liquid below 850ºC and that these liquids can be completely recrystallized when cooled to temperatures around 550ºC at rates of 0.1ºC per hour or slower.

For HTVDD, heating is a function of the thermal loading of the container and subsequent cooling is controlled by the type and age of the HLW, giving considerable scope for control. Gibb [2] modelled HTVDD for a case using spent pressurised water reactor fuel 'cooled' for 5 years after removal from the reactor. It was calculated that it would take around 65 days after disposal for the granite adjacent to the container to reach a maximum temperature of 850ºC, after which it would cool to 600ºC in about 2.5 years. This is equivalent to a cooling rate of ~0.011ºC per hour.

It is therefore concluded that S-type and similar high-Si granitic rocks, which are abundant at suitable depths over large areas of the continental crust, could be appropriate host rocks for HTVDD of heat generating HLWs. This could provide a route for the disposal of HGRs and VLLRs separated from large quantities of HLW/SNF thus opening the way for disposal of the remaining bulk of these wastes in mined and engineered "under 10,000 year repositories". The latter should greatly facilitate a solution to the "nuclear waste problem", technically, economically and politically.

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