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Granite recrystallization: The key to the nuclear waste problem?

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ABSTRACT

We report the outcome of high-temperature, high-pressure experiments showing that granite can be partially melted and completely recrystallized on a time scale of years as opposed to millennia as widely believed. This could prove the key to secure, very deep borehole disposal in the continental crust for small to moderate volumes of particularly problematic radionuclides. Removal of these problematic isotopes from spent nuclear fuel and other forms of high-level waste could open the way to safe and acceptable disposal of the remaining bulk of high-level waste with large volumes of intermediate-level waste in geologically shallow, conventional repositories.

Keywords: radioactive waste disposal, continental crust, melting, crystallization, borehole.

INTRODUCTION

One of the seemingly more intractable challenges to contemporary science is how to deal with radioactive waste, especially spent nuclear fuel and other forms of high-level waste. The solution toward which most countries with inventories of high-level waste and long-lived intermediate-level waste are tending is the mined and engineered repository at depths of a few hundred meters. In these repositories the waste is encapsulated and sealed into an extensive tunnel system, usually in excavations in the walls or floors. Individual designs vary (Miller et al., 2000), but the generic pattern is the Swedish KBS-3 concept. As evidenced by the British Government's refusal to allow Nirex to develop a similar repository in the UK, the mined and engineered multibarrier concept is not without its problems. However, most of these (and certainly the more serious) are related to two aspects of the waste.

First, the presence of heat-generating radionuclides (HGRs) places engineering constraints on the design of the repositories and raises questions about the performance of the construction materials. The waste has to be packaged in relatively small units dispersed throughout a large volume to prevent excessive temperature rises in the enclosing rock. Even so, the fact that significantly elevated temperatures (above ~150 °C) can be generated in and around the packages casts doubts on the abilities of cements, grouts, and seals to function. Because some of the high-heat-generating radionuclides (HHGRs), such as ^{134}Cs, ^{137}Cs, ^{90}Sr, and ^{90}Y, have relatively short half-lives, the scale of the heating problem can be reduced by allowing the waste to cool for several decades prior to disposal. However, this is not a complete solution, because many of the HGRs are long-lived or very long lived radionuclides (VLLRs).

Second, the presence of VLLRs requires that the wastes be isolated from the biosphere for 10^5–10^6 yr. Because of the difficulties of predicting climatic, hydrologic, and geologic conditions over such long times, it is almost impossible to make the necessary performance assessments and safety cases for such repositories.

ALTERNATIVE APPROACH

If the HGRs, HHGRs, and the problematic VLLRs (e.g., Am, Np, Cm, and possibly Pu), which in total are volumetrically minor, could all be removed from the waste, disposal of the rest—including the U (unless it is to be reprocessed)—becomes much less of a problem. The remainder of the high-level waste can be put into mined repositories without the need to spread small packages, each with its own multibarrier system, throughout large volumes of rock. This could enable construction of more massive and secure barriers around large single masses of waste while making the repository smaller, more economical, and less environmentally disruptive. This approach would also eliminate heating problems and remove concerns over the performance of materials because temperatures throughout the repository would be little different from ambient. The most important benefit, however, is that it would reduce the period over which performance assessments and safety cases need to be made to a few thousand years (i.e., a <10 k.y. repository). This is well within the predictive capabilities of geological and engineering sciences.

Traditionally the nuclear industry has considered that the components of high-level waste, especially spent nuclear fuel, should be stored and/or disposed of together. However, considerable interest has arisen recently, especially in France and the United States, in partitioning and transmutation of spent nuclear fuel as a possible way of dealing with the troublesome VLLRs. The science of transmutation is complex, uncertain, expensive, and long term (DEFRA, 2001), but the apparent willingness of the industry to contemplate partition of the waste offers the prospect of attractive alternative solutions such as the suggested <10 k.y. repository. In this option, the bulk of the high-level waste, including U (unless it is to be reprocessed), but minus the problematic radionuclides, is disposed (or at least located) with intermediate-level waste in a shallow (<1 km) mined repository. The inclusion of UO_2 is unlikely to be a problem because, notwithstanding its radioactive longevity, it is extremely immobile, as evidenced by the stability of uraninite and pitchblende deposits in Earth's crust over millions of years (Miller et al., 2000).

VERY DEEP DISPOSAL

To enable use of the <10 k.y. repository option, a safe and satisfactory way of disposing of the relatively small amounts of HHGRs, HGRs, and VLLRs removed from the high-level waste destined for the repository is required.

Several schemes have been proposed for the disposal of moderate volumes of radioactive waste in deep or very deep boreholes (e.g., Juhlin and Sandstedt, 1989; Watts, 1997; Gibb, 1999, 2000). Potentially the safest and most robust of these is an adaptation of the scheme (Gibb, 2000) in which high-level waste in special containers is placed in the lower part of a 4–5-km-deep borehole in granitic continental crust. Radioactive decay gradually heats up the waste packages to a peak temperature sufficient to generate a substantial zone of partial melting in the granite surrounding the containers. As the heat output of the waste decreases, the melt slowly cools and recrystallizes to seal the packages into a sarcophagus of solid granite surrounded by zones of thermal metamorphism in which any pre-existing fractures are sealed by annealing and low-temperature hydration mineralization. Widths of the zones of melting and metamorphism are maximized by preventing sinking of the packages (Gibb, 2000), in contrast to deep self burial schemes (e.g., Logan, 1974). The scheme depends on two crucial premises. First, sufficient melting of the granite will occur at low enough temperatures for the containers to survive, and second, the partial melt can be completely recrystallized to a fine- to medium-grained holocrystalline rock—both on time scales appropriate to the thermal decay of the waste.

The rate at which the rock around a high-level-waste package heats up depends on the heat output of the waste and conductive transfer. For the case modeled by Gibb (2000), it would take between 60 and 70 days for the granite adjacent to the container to reach 850 °C. Once the rock has reached its maximum temperature, the rate at which it cools is a function of the decay of the waste. For typical spent pressurized water reactor fuel cooled for 5 yr after removal from the reactor, it would take ~2.5 yr for the temperature of the granite to fall from 850 to 600 °C, i.e., a cooling rate of ~0.0114 °C/h (see Fig. 4 of Gibb, 2000). Heating of the rock is thus controlled by the thermal loading of the packages, and the subsequent cooling rate depends on the type and age of the waste.

The strength of this scheme is that, by going to such depths at an appropriate site, the waste is placed in an environment where any fluids present in fractures in the enclosing rock are physically and chemically isolated from near-surface groundwaters and have been so for many millions of years—a situation that is not likely to change in the next 10^5 yr. Hence, as has been argued elsewhere (Gibb, 2000), in the unlikely event of complete failure of the near-field barriers, the geologic barrier provides an ultimate safeguard. This scheme could readily be adapted for combinations of HHGRs, HGRs, and VLLRs. Unfortunately, despite the more than adequate confining pressure at depths of >3 km, the association of radioactive waste with high temperatures and hydrous fluids is likely to conjure up imagined scenarios in the lay mind that could make political acceptability of the scheme difficult to achieve.

An alternative that avoids this unfortunate association is a low-temperature, very deep disposal scheme being researched by one of us (Gibb). In this scheme the packages, which do not contain high enough concentrations of HGRs to significantly heat the adjacent rock, are placed in the lower reaches of a borehole. The borehole is then sealed at intervals above the waste packages, but still within the zone of deep, isolated rock fluids. As in the high-temperature scheme, sealing is done by melting and recrystallizing the host granite and can be achieved either by special sealing packages of HHGR-bearing waste or by controlled electrical heating.

Although safety is the paramount criterion for any disposal option, it is inevitable that technical feasibility and economics will be significant. Great advances have been made in deep-drilling technology in recent years, and large-diameter (to 1 m) holes to depths of >4 km are now considered commercially feasible (Harrison, 2000). The principal costs of any deep borehole disposal scheme will be those arising from the drilling and casing of the holes. One of us (Gibb, 2000) conservatively estimated the cost of a 4 km, 0.5-m-diameter hole as 6.25 million U.S. dollars, and Harrison (2000) more accurately projected the cost of a 4 km, 0.8-m-diameter hole as 4.62 million U.S. dollars. Significant savings are likely to arise if multiple, slightly splayed, holes are sunk from a single surface location. We are aware of no realistic final-cost projections available for any of the mined and engineered repository concepts under consideration, but it is clear that the costs of the exploratory investigations and repository construction alone would be two or three orders of magnitude greater than the cost of boreholes. It seems probable therefore that the cost of the necessary boreholes would be more than recovered from the savings arising from substitution of the smaller <10 k.y. repository enabled by very deep borehole disposal of the problematic radionuclides.

GRANITE MELTING AND RECRYSTALLIZATION

However the waste is sealed into the borehole, it is essential that the granite can be partially melted and completely recrystallized in a matter of years and, in two of the cases, on the time scale of the heating and cooling of the waste (Gibb, 2000). From what is known about the kinetics of melting in granitic systems (e.g., Piwinski, 1967; Scaillet et al., 1995), it is unlikely that much of the partial melting generated in the disposal scenarios would be equilibrium melting. Crucially, in the context of the schemes, little or nothing is known about how rapidly granitic magma can be cooled and still give rise to a holocrystalline rock. Similarly, there are no directly determined data for the minimum times needed for the crystallization of medium- and coarse-grained granites. Cooling rates of natural granites have been deduced from the times taken for the intrusions in which they occur to cool from their emplacement temperatures, but such times (derived by various techniques) have indicated cooling rates mostly in the range 10–500 °C/m.y. (Attrill and Gibb, 2003b). These have encouraged a widely held belief that granites can only form by extremely slow crystallization over thousands, if not millions, of years. There are, however, good grounds for believing that silicic magmas can be completely crystallized at cooling rates orders of magnitude faster.

To test this, and hence the feasibility of the borehole disposal schemes, partial melting and linear cooling (recrystallization) experiments were undertaken on a typical upper crustal granite under the conditions likely to occur in the schemes. The rock used was an S-type granite of Caledonian age from northern England. The experiments were carried out mainly on powdered rock at a pressure of 0.15 GPa, corresponding to the pressure at depths of 4–5 km in continental crust. Total H_2O contents of the charges varied from 0.58 wt% (no added H_2O) to 10.6 wt%, so many of the data are for the H_2O-undersaturated conditions appropriate to the disposal schemes (Gibb, 2000).

The partial melting experiments were initially intended to simulate the continuous heating of the high-temperature disposal scheme, but several factors encouraged the use of prograde isothermal runs in which the sample was heated as quickly as practicable to the target temperature and held for the required time before quenching. On the time scale of the experiments (200–2650 h), this leads to a slight underestimation of the amount of melt generated for any temperature compared with true simulation of disposal conditions (Attrill and Gibb, 2003a). The partial melting relationships for the granite were determined for H_2O vapor-saturated and H_2O vapor-undersaturated conditions between 650 and 850 °C and are portrayed as a phase assemblage diagram in Figure 1. (Full accounts of the methods and products of the experiments were given in Attrill and Gibb [2003a], and only the key results are summarized here.)

Significant in the context of the disposal schemes is that melting begins just below 700 °C irrespective of H_2O content. However, for undersaturated melting there is a positive correlation between the amount of melting (determined by modal analyses of quenched samples) and H_2O content (Fig. 2). Once vapor saturation is reached the amount of melting becomes independent of H_2O content. From experiments undertaken to ascertain how close the partial melting is to equilibrium and other evidence (Attrill and Gibb, 2003a), we consider that Figure 1 is close to being an equilibrium melting diagram in its higher-temperature and higher-H_2O-content regions. However, at lower temperatures there is little doubt that the melts produced are a long way from equilibrium. Nevertheless, it is prograde melting of crystalline rock similar to that represented by our experiments and not equilibrium melting that would occur during heating of a granite host by packages of high-level waste.

Some of the partial melting experiments carried out on crushed granite were repeated on solid rock. For example, powdered granite with a total H_2O content of 2.58 wt% held at 800 °C for 570 h closely approached equilibrium, and generated 60 vol% melt. Repeating this experiment with a 20-mm-long, 7-mm-diameter core of rock produced the same phase assemblage, but with only 40 vol% melt. Clearly this experiment did not attain equilibrium. Melting was initiated at the outer surface of the core, producing an envelope of liquid. As melting progressed it permeated throughout the core to give an interlocking net-

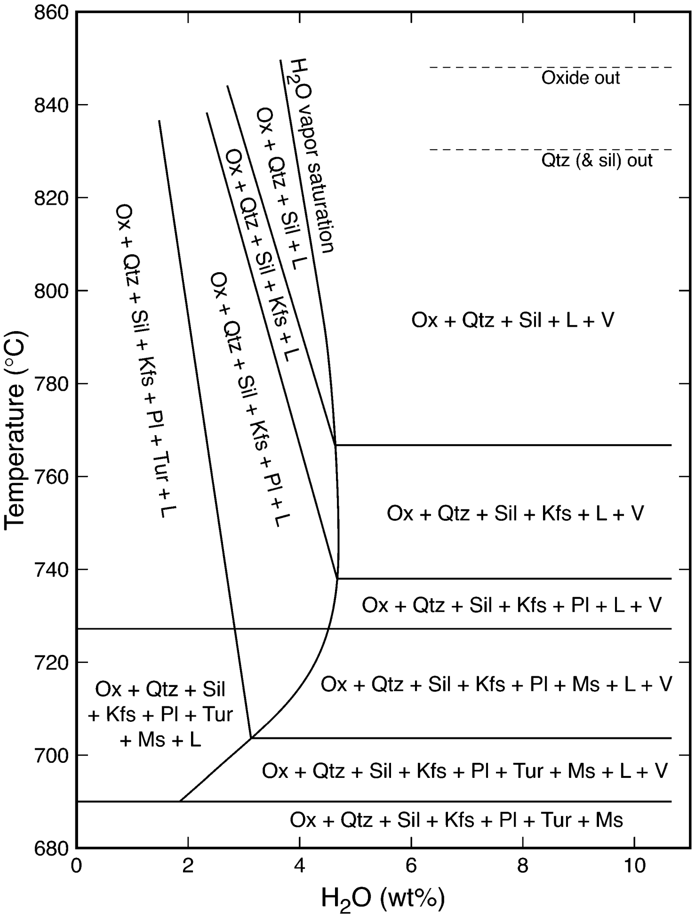

Figure 1. Phase assemblage diagram for granite melting experiments at pressure, *P* = 0.15 GPa. Abbreviations: Kfs—alkali feldspar; L—liquid; Ms—muscovite; Ox—iron-titanium oxide; Pl—plagioclase; Qtz—quartz; Sil—sillimanite; Tur—tourmaline; V—vapor. Sillimanite is not primary phase, but comes from breakdown of chlorite (Attrill and Gibb, 2003a).

work of liquid (Fig. 3). Bearing in mind the likely kinetic effects of the difference between the powdered granite and the solid core, it is remarkable that as much as 40% melting occurred at 800 °C in only 570 h. From the standpoint of the disposal schemes this result is most encouraging, and even more so is the fact that melting permeates rapidly through the solid rock.

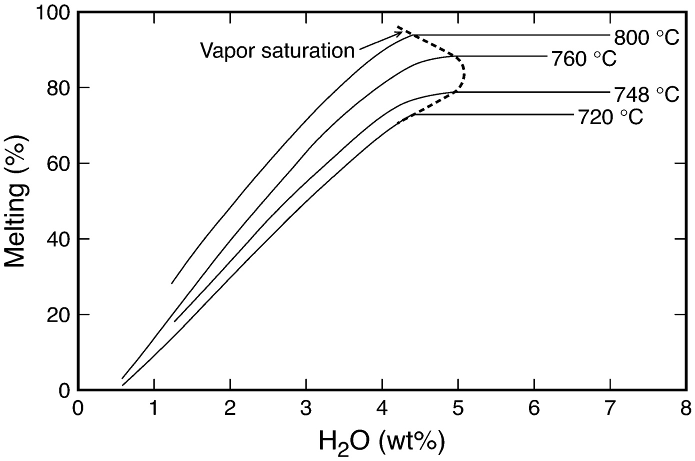

Figure 2. Variation in amount of partial melting of granite with water content at different temperatures.

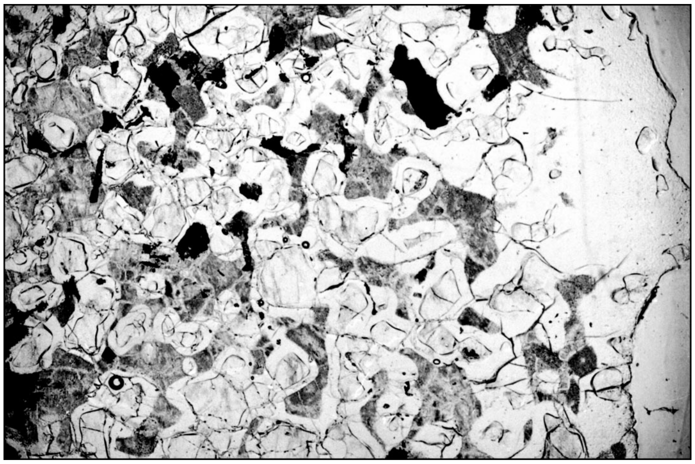

Figure 3. Thin section cut from core of granite partially melted at 800 °C (pressure, *P* = 0.15 GPa; 570 h; 2.5 wt% H_2O). Note continuous network of glass around relict crystals. Envelope of glass developed on surface of core is conspicuous at right side of photomicrograph (plane-polarized light; width of field is 6 mm).

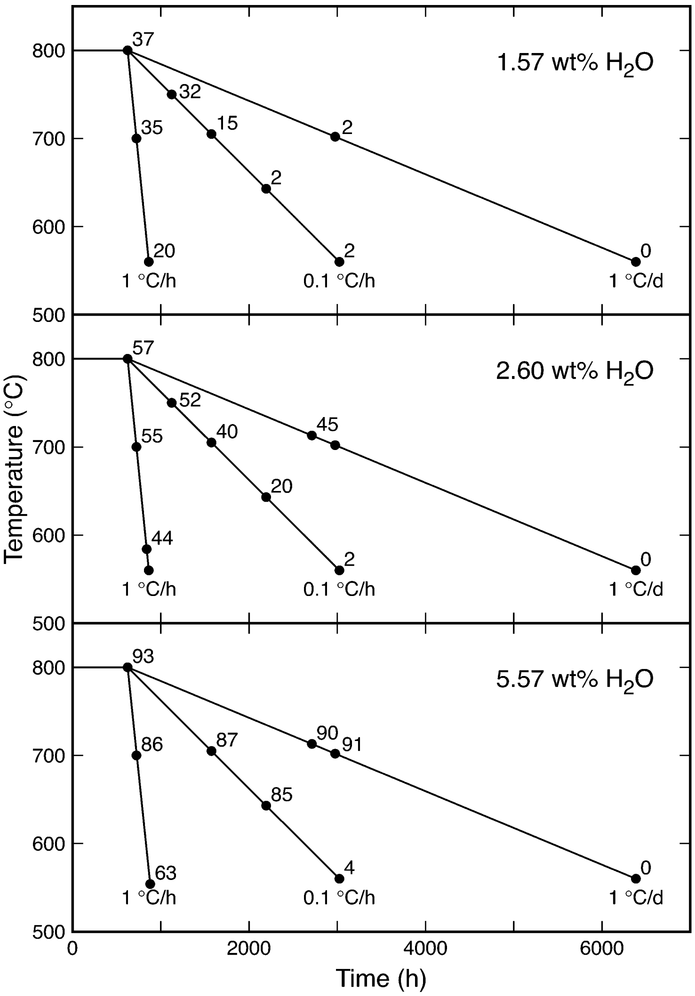

Figure 4. Summary of recrystallization experiments on granite for different water contents and cooling rates. Solid circles represent quenching temperatures and adjacent numbers are volume percent of melt remaining.

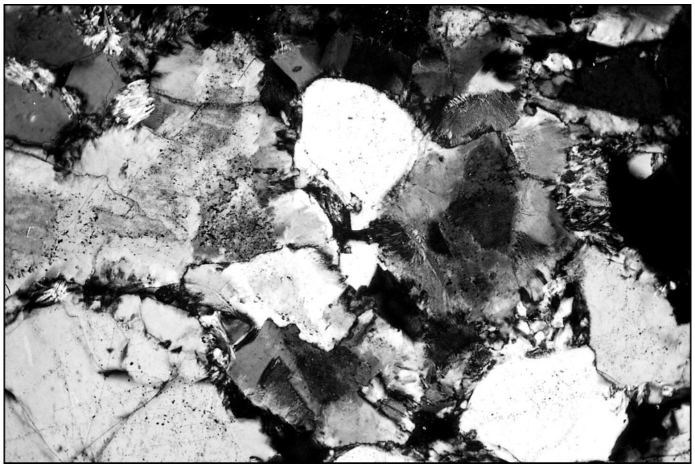

Figure 5. Thin section cut from core of granite (see Fig. 3) that was completely recrystallized by cooling from 800 to 560 °C at 0.1 °C/h (pressure, $P = 0.15$ GPa; 2.54 wt% H_2O; cross-polarized light; width of field is 1.2 mm).

The recrystallization experiments used three linear cooling rates (1 °C/h, 0.1 °C/h, and 1 °C/day) and three total H_2O contents (1.57, 2.6, and 5.57 wt%). To avoid the well-known nucleation problems in granitic systems (Naney and Swanson, 1980), it is essential that seed crystals be present in the melt. Usually this is achieved experimentally by incorporating large pieces of the seed phase in the starting material (Scaillet et al., 1995; Simakin and Chevychelov, 1995). In our experiments this was unnecessary; before cooling, the starting materials were run at 800 °C for 624 h such that only a partial melt was generated. Hence, the seeds were already present as relict crystals, exactly as they would be in the disposal schemes. The recrystallization experiments are illustrated in Figure 4, where the horizontal lines from time zero represent the 800 °C melting prior to cooling at the rate indicated. Samples were quenched from the points shown by the solid circles so that the progress of recrystallization could be traced, and the volume of melt remaining in each case is given in Figure 4. For further details of the recrystallization experiments and products see Attrill and Gibb (2003b).

The temperature at which any phase begins to recrystallize in the cooling experiments is lower than its melting temperature. The difference varies with cooling rate and is directly proportional to the H_2O content. Water effectively suppresses crystallization. An important consequence is that, for the cooling rates investigated, recrystallization continues to temperatures well below 700 °C (i.e., below the solidus). For example, for cooling at 0.1 °C/h, almost complete recrystallization does not occur until 640 °C with 1.5 wt% H_2O, until 560 °C with 2.5 wt% H_2O, and until even lower temperatures with higher water contents.

CONCLUSION

To be a suitable host rock for low-temperature, very deep disposal it is necessary that a reasonable amount of partial melting is possible at temperatures below ~1000 °C and that the melts can be recrystallized at cooling rates consistent with the method of sealing employed. For electrical heating, these could be controlled over several decades. For high-temperature, very deep disposal the constraints are tighter. A substantial amount of melting (perhaps >50%) must be achieved below ~850 °C, and complete recrystallization must occur at cooling rates faster than that of the waste package. As already indicated (Gibb, 2000), realistic cooling rates for the disposal of appropriate high-level waste are likely to be less, and often considerably less, than 0.03 °C/h. The experiments demonstrate that S-type crustal granite can be melted to yield suitable amounts of liquid at temperatures below 850 °C and the liquids can be completely recrystallized when cooled to temperatures of ~550 °C at rates slower than 0.1 °C/h. Thus, such granites, which are abundant throughout the continental crust, would be appropriate hosts for the very deep disposal of HGRs, HHGRs, and/or VLLRs.

Particularly significant in the context of such schemes are the cooling experiments carried out on cores of solid granite that not only confirmed that the partial melts can be recrystallized (Fig. 5), but demonstrated that the silicate liquids will flow into any fractures in the rock before sealing them completely on recrystallization.

Very deep borehole disposal, whether high or low temperature, therefore looks to be a viable means of dealing with the problematic radionuclides in high-level waste (provided their separation does not prove technically or economically prohibitive) and could contribute to resolving the wider nuclear waste problem as outlined herein.

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