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High-density support matrices: - key to the deep borehole disposal of spent nuclear fuel.

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Abstract

Deep (4-5 km) boreholes are emerging as a safe, secure, environmentally sound and potentially cost-effective option for disposal of high-level radioactive wastes, including plutonium. One reason this option has not been widely accepted for spent fuel is because stacking the containers in a borehole could create load stresses threatening their integrity with potential for releasing highly mobile radionuclides like ¹²⁹I before the borehole is filled and sealed. This problem can be overcome by using novel high-density support matrices deployed as fine metal shot along with the containers. Temperature distributions in and around the disposal are modelled to show how decay heat from the fuel can melt the shot within weeks of disposal to give a dense liquid in which the containers are almost weightless. Finally, within a few decades, this liquid will cool and solidify, entombing the waste containers in a base metal sarcophagus sealed into the host rock.

PACS Code 28.41 – Radioactive wastes – waste disposal.

1. Introduction

The world's inventory of spent nuclear fuel is already considerable, with over 55,000 tons in the USA alone, and the current management strategies in most of the nuclear nations are that spent fuel should end up in some form of geological disposal. The wider issues surrounding disposal of spent fuel are discussed briefly in section 5.4 but, in the context of this paper, the two most significant facts are that none of the options proposed to date for the disposal of spent fuel are without their technical difficulties and over 50 years since nuclear power generation began no country yet has an operational disposal facility.

Disposal of high-level radioactive wastes, including spent nuclear fuel and fissile materials like Pu, in deep boreholes could offer many advantages over more conventional mined and engineered repositories [1–4]. In particular, the greater depths (3-5 km as against 300-800 m) and less dynamic hydrogeological conditions increase confidence in the geological barrier against return to the biosphere of any radionuclides. Deep borehole disposal relies more on the geological barrier and less on engineered barriers, the performances of which are less certain on the timescale necessary for the isolation of HLW ($10^5 - 10^6$ years). In addition to greater safety, other potential benefits of deep borehole disposal include higher security (against terrorist or accidental intervention), wider availability of geologically suitable sites, less environmental disruption [2,4] and potentially better cost-effectiveness and public acceptability.

In the U.S.A. a MIT study on the Future of Nuclear Power [3] recommended that deep boreholes for spent fuel “merited a significant R & D program”. In the UK the Committee on Radioactive Waste Management (CoRWM), in recommending geological disposal for all high-level wastes [5], stated that decision making about the exact form of such disposal “should leave open the possibility that other long term management options [*than mined repositories*] (for example, borehole disposal) could emerge as practical alternatives”.

2. Deep borehole disposal of spent fuel

2.1. Packaging

Among the different versions of deep borehole disposal currently under investigation [2,4,6,7] is one designed for spent nuclear fuel from light water reactors (LWR) that we refer to [4] as “low-temperature very deep disposal – variant 2” (LTVDD-2). In LTVDD-2, after removal from the reactor and several years of cooling, the spent fuel rods (pins) are removed from the assembly and packed into a cylindrical stainless steel, copper or other metal container. The length of the container should be such as to avoid the need to cut down the fuel pins. After the container is heated slowly in an inert atmosphere, such as nitrogen or argon (mainly to prevent oxidation of the zircaloy cladding), a suitable material, such as molten glass or lead, is poured in to fill the voids between the pins. As little space as possible is left above the contents consistent with the need to secure the lid. The temperature to which the container and its contents must be heated to prevent quenching and cracking of the infill varies with the material, e.g., for Pb it is $\sim 330^{\circ}\text{C}$. After slow cooling to solidify and anneal the infill, the containers are sealed by welding on the lid and any surface contamination is removed before storing or transporting to the disposal site.

At first sight, close packing (70-80% by volume) of the irradiated fuel rods might suggest an increased risk of criticality but, in fact, the opposite is true. Even without the glass or Pb infill, and if the spaces between the rods somehow became filled with water, the reduction in the moderator:fuel ratio to well below the optimum value for fuel assemblies would eliminate the possibility of criticality. This is the same as the principle behind the consolidated storage of spent LWR fuel rods in some reactor cooling ponds, where the pins are removed from their assemblies and packed into long metal boxes for storage under water. In the case of MOX fuel, criticality is equally unlikely provided the burn-up of the fuel is sufficient and, in any event, should it be

perceived as an issue (for UO₂ or MOX fuel), the risk could be further reduced by the use of neutron poisons.

A problem which could arise, at least in theory, stems from the fact that microcracks occasionally develop in the zircaloy cladding of the fuel rods during reactor operation and minute amounts of water might gain access. If any such water is still present when the hot glass or Pb fill is poured into the container it might cause miniature steam explosions that could threaten the integrity of the cladding and enable the escape of volatile fission products, such as Xe, Kr, Cl, I and Cs, especially if these had become concentrated in the gap between the cladding and the fuel pellets during reactor operation. However, sufficiently slow pre-heating of the container and fuel rods would evaporate any such water and eliminate the risk, however small.

There is also the possibility that, in the case of Pb infill, some reaction could occur between the Pb and the zircaloy cladding that might threaten the integrity of the latter. Phase relations in the system Pb-Zr are not fully understood but it is known that Pb is very slightly soluble in Zr at 330°C suggesting some diffusion would occur at the interface during the infilling of the spaces between the fuel rods. However, diffusion is a very slow process and, given the short time at elevated temperatures during infilling, any reaction between the Pb and the zircaloy would be confined to the surface of the cladding. We are investigating this further but our best estimate at present is that for there to be any risk of the Pb penetrating and destroying the cladding on the timescale involved the temperatures would need to be several hundred °C higher.

2.2. Borehole deployment.

At the disposal site the containers are deployed, singly or in small batches, over the bottom 1 km or so of a 4-5 km deep, large diameter (up to 0.8 m), fully-cased borehole sunk into granitic continental crust (Fig. 1). When deployment of the waste packages is complete the borehole is sealed at intervals above the deployment zone.

This could be accomplished satisfactorily by a number of methods and using different materials but perhaps the best would be 'rock welding' through the partial melting [6-8] and recrystallization [6,7,9] of the wall rock and crushed granite backfill. This could be done with special containers filled with high heat generating HLW [2,6] but could equally easily be achieved by down-hole electrical heating. For additional security the uppermost casing could be removed and the top of the hole obliterated to conceal its location once sealing was completed.

Depending on the type of spent fuel, filler, pin packing density and the material and thickness of the container, the waste package will have a specific gravity (SG) in the range 8 to 11. Even under a hydrostatic head of 3-5 km of borehole fluid, the number of containers that could be stacked on top of each other before the physical integrity of those at the bottom is jeopardized is limited (40-90 depending on type). The ability of the containers to deform without rupture decreases with the clearance between the container and the casing (Fig. 1). Unfortunately, a requirement of 'engineering out' the slight risk of waste packages jamming and having to be recovered during deployment is a substantial clearance between the container and casing. For a 0.8 m diameter hole with 0.7 m inside diameter steel casing the clearance should exceed 0.03 m.

Disposing of complete fuel assemblies (rather than individual pins) with a low density filler such as glass might reduce the density of the packages enough to avoid the problem, but it would be inefficient use of borehole space and would greatly increase costs. A better solution might be to deploy containers singly and grout the gaps with cement that is allowed to set before the next emplacement. Alternatively, the containers could be deployed in small batches separated by bridging plugs to spread the overlying load onto the borehole walls. However, the former would slow down and reduce the simplicity of borehole disposal while the latter would present considerable engineering difficulties. Both could subsequently fail as a result of the heat from the waste and do

little to prevent access of aqueous fluids from the host rock to the containers. Below we show how this loading problem could be eliminated by the introduction of a novel type of high-density support matrix (HDSM) with many additional benefits to the safety and effectiveness of the geological disposal.

3. High-density support matrices

We propose emplacing a controlled volume of dense, Pb-based, low-melting point alloy in the form of fine shot after the deployment of each container or small batch of containers. The shot would sink rapidly through the aqueous drilling/emplacement fluid to surround the container(s), filling the gaps between the container(s) and the casing and, through perforations in the casing (required for weight reduction [10]), the spaces between the casing and rock. The solid shot would itself function as an effective interim support matrix. However, shortly after emplacement, heat from the radioactive decay of the waste would begin to melt the metal allowing it to flow into any remaining voids. It would displace upwards the aqueous fluid from between the shot grains. [Note that this fluid will not vaporize even at temperatures well in excess of those generated by the waste due to the pressure (~ 150 MPa) at such depths [6,7].] Over tens of years the molten HDSM will cool and solidify to encase the waste packages, effectively ‘soldering’ the containers in to the borehole.

The alloy is chosen to have an appropriate melting point and liquid SG for the disposal. The SG should be such that the filled containers will only just sink, reducing both the effective weight of the stack and the load on the bottom containers so they can withstand deformation until long after the HDSM has solidified around them. The melting point is less critical as long as the temperatures generated by the waste are sufficient to melt the shot (section 4). A range of suitable alloys exists in the system Pb-Sn-Bi but Zn, Sb, etc. could replace Sn or Bi or be added to make quaternary alloys. SG decreases with Sn content while increasing the amount of Bi or Sn (towards the

eutectic compositions) will lower the liquidus temperature (Fig. 2) [11]. The one atmosphere ternary eutectic in the system Pb-Sn-Zn [11] occurs at 177°C and $\text{Pb}_{23}\text{Sn}_{72}\text{Zn}_5$. As far as we know, the phase relations in these alloy systems have not been determined for 150 MPa but, since this pressure only raises the melting points of pure Pb, Sn and Zn by 10°C, 4°C and 6°C respectively [12,13], increases in melting temperatures for binary and ternary alloys should be similarly small. If the SG of the waste packages was greater than 11.3, Pb could be used alone as the HDSM, with additional benefits (section 5), although raising the melting temperature to over 327°C. Binary alloys of Pb and Sn offer a range of specific gravities between 7.27 and 11.3 and most begin to melt at 183° at one atmosphere (Fig. 2a). Alloys of Pb and Bi have higher SGs but begin to melt as low as 125.5°C (Fig. 2b).

The selected alloy could be made as homogeneous shot and emplaced through the drill-hole deployment string releasing it a short distance (5-20 m) above the last container emplaced in each batch. Alternatively, the components of the alloy in shot form could be mixed in the required proportions before emplacement. The decay heat of the waste then creates the alloy *in situ* by melting. The latter has obvious benefits of economics and flexibility but could be limited by the capacity of the heat output from the waste being too low to melt pure end members.

4. Thermal modelling

To demonstrate how the Pb-based HDSM would work we have modelled two realistic cases of LTVDD-2 for spent LWR fuel using a computer code [GRANITE] created for determining temperature distribution with time in and around deep borehole disposals of heat-generating nuclear wastes [4].

Case A is a stack of 10 stainless steel containers with 73% (by volume) of 30 year old pressurized water reactor UO_2 fuel pins (45 GWd/t burn-up) and a lead infill. The containers, which are 3.75 m long with an outside diameter of 0.63 m and wall

thickness of 0.05 m, are deployed at two day intervals with enough shot emplaced immediately after each container to cover it to a depth of at least 1.5 m. Such packages would have a SG of 10.1 requiring a HDSM with a SG of ~ 9.7 . This is provided by a $\text{Pb}_{70}\text{Sn}_{30}$ (wt. %) alloy with 1-atmosphere solidus and liquidus temperatures of 183°C and 256°C respectively (Fig. 2a). The corresponding temperatures for 150 MPa should be only a few degrees higher. The modelling results are shown in Fig. 3a in which each curve represents the evolution of temperature for one point within the HDSM (see figure caption). The HDSM would first begin to melt against the container wall at the mid point of the stack 17 days after initial deployment while at the bottom of the stack it would take 29 days for melting to begin. Against the borehole wall melting would begin less than half a day later than against the container. The small time differences between the HDSM attaining its solidus temperature at its inner and outer surfaces are a consequence of the high thermal conductivity of the alloy. At the top of the stack it would take 120 days for the HDSM to start melting. All of the HDSM would be at least partly molten four months after deployment. The maximum temperature attained in the HDSM would be 356°C (against the container at the mid point of the stack) after 5.5 years. The HDSM would begin re-solidifying against the borehole wall at the top of the stack after 49.5 years and solidify completely (against the container at the mid point of the stack) after ~ 125 years.

Case B is a stack of 10 copper containers with 73% of 30 year old spent fuel (45 GWd/t burn-up) from an AP-1000 light water reactor of the type likely to be used for any nuclear 'new build' in the UK or USA. The containers are longer (4.3 m) and thinner walled (0.035 m) than in case A and deployed at one week intervals. The packages would have a SG of 10.6 making the ideal SG of the HDSM around 10.3, which could be provided by a $\text{Pb}_{40}\text{Bi}_{60}$ or a $\text{Pb}_{83}\text{Sn}_{17}$ alloy. The 1 atmosphere solidus and liquidus temperatures of the former are 125.5°C and 160°C respectively and for the latter 205°C and 285°C (Fig. 2). The latter is preferred on the grounds of cost and Pb

content but the former would be more suitable if complete melting is required or the ambient temperature at 4 km depth is lower than the 100°C used in the modelling. The results of temperature/time modelling are given in Fig. 3b. In this case the Pb-Sn HDSM would begin to melt against the container at the bottom of the stack 36 days after the first container was emplaced. At the mid-point of the stack the HDSM would begin to melt after 47 days and peak at 364°C in just under 6 years. At the top of the stack the HDSM against the borehole wall would not begin to melt until after 143 days and would solidify after 47 years. The final solidification of the HDSM (against the containers at the mid point of the stack) would occur after ~103 years.

Using spent fuel with a lower heat output (e.g., older than ~ 40 years) might result in failure of the HDSM at the very top of the stack to achieve partial melting. This need not be seen as an obstacle since the heat from the bottom of the next container (or stack) would melt it when emplacement resumes. However, should this be perceived as problematic it would be a simple matter to use one or more packages containing spent fuel with a higher heat generating capacity at the top of the stack to ensure melting of the HDSM occurs all around the stack. We have modelled such ‘management’ of the temperature gradients along the length of the stack elsewhere [4]. Alternatively, using a HDSM with a lower solidus temperature could achieve the same result.

5. Discussion

The above examples demonstrate that Pb-based HDSM could overcome the problem of load-stressing the containers provided sufficient heat is available from the decay of the waste. The temperatures attained in and around a borehole disposal (and the times taken to reach them) can be tailored to the requirements of the disposal by adjustment of parameters such as the type and age of the spent fuel; the ratio of fuel pins to filler in the containers; and the composition, dimensions, number, spacing and emplacement rate of containers in the stack.

5.1. Thermal expansion

In this, as with all borehole disposal concepts involving heating, it is important to consider the effects of differential expansion of the various materials involved, especially in the context of the HDSM. Full quantitative analysis of these effects requires consideration of all the materials concerned in each specific case combined with the temperature variations throughout the system as predicted by the thermal modelling and is beyond the scope of this paper. However, it is useful to consider a ‘worst scenario’ general case based loosely on case A modelled above. At the mid point of the stack the temperature of the stainless steel container rises to a maximum of 356°C (Fig. 4). With a coefficient of linear expansion (α) of around $17.3 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ the volume expansion of the container from an assumed pre-disposal temperature of 25°C would be 1.72%.

5.1.1. Inside the container

The maximum temperature attained by the contents of the container at the mid point of the stack varies from just over 360°C on the borehole axis to about 356°C against the container. Expansion of the contents needs to be considered in two stages: up to the point at which the Pb infill begins to melt and then up to the maximum temperatures attained.

When the Pb infill begins to melt (at 327°C) on the axis of the borehole the temperature against the inner surface of the container is 324°C (Fig. 4). By this stage the steel container will have expanded by 1.55%. Taking an average of 325°C for the contents, the fuel pins with $\alpha = \sim 8.0 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ (UO_2), will expand by 0.72% while the Pb infill ($\alpha = 28.9 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$) will expand by 2.60%. With a fuel pin to infill volume ratio of 73:27 the contents of the container will therefore expand by 1.23%, which is less than the 1.55% expansion of the container. Consequently no significant stresses would arise within the container up to the point at which the Pb infill begins to melt. Substantially lower spent fuel to infill ratios could lead to the contents expanding more

than the container resulting in internal stresses in the container. Small amounts of stress within the container could be accommodated by the steel without any threat to its integrity but, for cases where calculations indicate that expansion of the contents would be too great, a Pb-Sn or Pb-Bi alloy infill with a lower coefficient of expansion could be used to avoid this.

When Pb melts it undergoes an expansion of $\sim 2.9\%$ and the liquid continues to expand. Consequently, by the time the contents of the container have reached their maximum temperature of $\sim 357^\circ\text{C}$ (Fig. 4) the Pb will have undergone a total expansion of 6.1% giving an overall expansion of the contents of 2.2% , which is greater than the 1.72% expansion of the container. However, once the Pb is liquid it can readily expand into the space at the top of the container left on filling (to enable welding of the lid) and increased by contraction of the original liquid Pb infill, so eliminating any significant stresses inside the container.

5.1.2. Outside the container

At the mid point of the stack the borehole casing (carbon steel with $\alpha = 11.7 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$) reaches a maximum of about 355°C and will expand by 1.16% . Prior to the onset of melting in the $\text{Pb}_{70}\text{Sn}_{30}$ HDSM at 183°C the container will expand by 0.82% and the casing by 0.56% resulting in a decrease in the space between the two. Over the same interval the HDSM ($\alpha = 25 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$) will expand by 1.18% but being in the form of shot the expansion would be accommodated by 'flow' up the annulus into the space above the top of the stack. Once melting begins the molten alloy will sink down the annulus displacing the aqueous inter-grain fluid resulting in a large effective volume decrease in the HDSM with any resulting (fluid-filled) space in the annulus being filled by more alloy from the reserve in the space above the top container.

It is also instructive to consider what will happen in the annulus during cooling once the HDSM has solidified. While the volume changes between the solidus of the HDSM (183°C) and ambient temperature (100°C) will be quite small, the container will

contract more than the casing and the HDSM more than both. At first sight this would suggest small gaps arising between the containers and HDSM and between the HDSM and casing. However, the Pb-based HDSM will remain malleable and the vertical pressures should be more than enough to ensure slow compression of the HDSM fills any such gaps.

5.2. Other benefits of HDSM

Pb-based HDSM offer additional benefits for the safe disposal of spent nuclear fuel. Because of the high SG and relatively low viscosity of the molten metal it will flow efficiently into all voids around the waste packages and between the casing and wall rock. It will also flow into any fissures in the wall rock rendering the entire system impenetrable to aqueous fluids during the deployment stage and indefinitely beyond, so effectively eliminating any potential radiolysis problems. By contrast, cementitious grouts, which could fulfil a partial support role, would have to be pumped down-hole under pressure, are unlikely to fill the voids so effectively and could be prone to cracking after setting due to the heat from the waste and/or the load of subsequent container deployments. They could not give the sealing efficiency of the HDSM. Also, because HDSM are at least partly molten for a period exceeding the operational (filling) period of the borehole, they would allow gradual physical adjustment (settling) of the containers with minimal risk. Further, the malleable nature of Pb and its alloys means that not only could such adjustments continue indefinitely but minor tectonic deformations (earthquakes) in the enclosing rock could probably be accommodated by the HDSM without risk to the containers long after the borehole is sealed, thus further enhancing safety.

Another benefit of the HDSM is that it could enable the use of thinner walled containers since they have to withstand only the stresses of transport and deployment and not the weight of an overlying stack of unsupported packages, thus saving on material and simplifying manufacture and welding. The decrease could be to either the

outer or inner diameters of the containers: the former enabling a reduction of borehole diameter and the latter an increase in the amount of spent nuclear fuel disposed of. Both would increase cost effectiveness.

Yet a further advantage of using Pb-based HDSM is that there is much radioactively contaminated Pb in the nuclear industry requiring safe disposal, although the most highly contaminated Pb might best be used for container infill rather than the HDSM. In spent nuclear fuel disposals where sufficient heat is available, the SG of the package could be raised above 11.3 to allow use of 100% Pb HDSM, thus simplifying manufacture and maximizing the disposal of Pb. The higher SG could be deliberately achieved by using U metal (from spent metal fuels or depleted U) as a replacement for some of the oxide fuel pins or as granules mixed with the Pb infill.

5.3. Environmental aspects

The use of large amounts of toxic heavy metals such as Pb in the proposed disposal scheme would appear, at first sight, to raise environmental concerns. Although at temperatures below $\sim 400^{\circ}\text{C}$ there is no reaction between Pb, Sn, Bi etc. and the silicate minerals of a rock like granite, there is a theoretical possibility of some Pb getting into the deep crustal fluids in and around the borehole. While Pb is insoluble in hot water, these fluids are likely to be dense brines in which Pb could form soluble halide complexes and be transported in small quantities through the rock. However, quite apart from the fact that the density stratification of these brines would prevent any uprise to the biosphere [1,2,4], the Pb complexes are unlikely to travel far before cooling, encountering a different geochemical environment and precipitating as Pb sulphides in much the same way as natural hydrothermal ore deposits of Pb are formed. The presence of high local concentrations of Pb in rocks, even in deposits close to the surface, causes no real environmental problems.

5.4. Wider considerations

It was pointed out in section 4 that using spent fuel more than ~40 years old might generate insufficient decay heat to adequately melt the HDSM so requiring ‘fortification’ with some younger fuel. While a substantial proportion of the world’s inventory of spent UO_2 fuel is currently suitable for disposal using the proposed scheme, there are some non-scientific issues that could affect this and could have a bearing on the potential application of the scheme.

Technically, the deep borehole disposal scheme proposed above could be implemented within 10 years but delays over political decisions, site selection, legal debates, planning permission etc. make it unlikely that any disposal facility would be available in less than 20 years. Consequently, the proportion of the inventory that could be disposed of without the addition of younger fuel will decrease. Also, some countries have a policy that spent fuel must undergo a statutory cooling period (40 to 100 years) before going to long-term storage or disposal. Unless such policies are revoked (quite possible if a suitable route for earlier disposal is identified) they could effectively foreclose on the proposed scheme as all the fuel will be incapable of providing sufficient heat by the time it is released. Nevertheless, the prospects for the disposal scheme are good if current and ‘new build’ reactors operate on the ‘once through’ fuel cycle (the current policy in the USA, the UK, Sweden, Finland, etc.) and there is an ongoing supply of young spent fuel for disposal.

On the other hand, there is a view that spent fuel should not be regarded as waste and disposed of. Given the likely increase in demand for uranium and an inevitable rise in price, the pressures for reprocessing of existing stocks of spent fuel could become ever greater and future reactor operations, e.g. Generation IV, may well be designed with reprocessing in mind. In such circumstances direct disposal of spent UO_2 and MOX fuel could be confined to nuclear operators with small programmes that do not justify the costs and risks of transportation and reprocessing. It is not inconceivable, however, that further into the future, reactor technology could be based on inert matrix

fuels that are again designed for ‘once through’ cycles with closure by direct disposal, for which deep boreholes could re-emerge as the ideal route (although the need for HDSM might be reduced).

6. Conclusion

Notwithstanding any misgivings about the long term wisdom of disposing of spent nuclear fuel, there is currently a large inventory awaiting geological disposal and the proposed scheme could provide the necessary route for a substantial proportion of this. High density support matrices, deployed as described above, could overcome the container loading problem, offer several additional benefits, and provide the key to the disposal of spent nuclear fuel in deep boreholes with all the advantages of increased safety, security, cost, etc. that this option offers.

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FIGURE CAPTIONS

- Fig. 1. Schematic of the bottom container of the lowermost stack in a deep borehole disposal for spent nuclear fuel in granitic host rock. (HDSM = Pb-based high-density support matrix).
- Fig. 2. One atmosphere phase diagrams for binary alloys; (a) the system Pb – Sn; (b) the system Pb – Bi; (Both after [11]).
- Fig. 3. Temperature/time curves for two cases of deep borehole disposal of spent nuclear fuel. Ambient temperature at a depth of 4 km in the continental crust is taken as 100°C. (a) Case A is a stack of 10 stainless steel containers of 30 year old spent PWR fuel (see text). Each curve represents a point within the HDSM adjacent to the container surface as follows: 1 = mid-point of the stack; 2 = bottom of the stack; 3 = top of the stack. The broken horizontal line is the one atmosphere solidus temperature of the HDSM. (b) Case B is a stack of 10 copper containers of 30 year old spent AP-1000 reactor fuel (curves as in (a)).
- Fig. 4. Temperature variation along the horizontal radius of case A for the mid-point of the stack when the temperature on the borehole axis reaches 327°C (after 361 days - dashed line) and when the temperature of the container surface attains its maximum value (after 5.6 years - solid line).

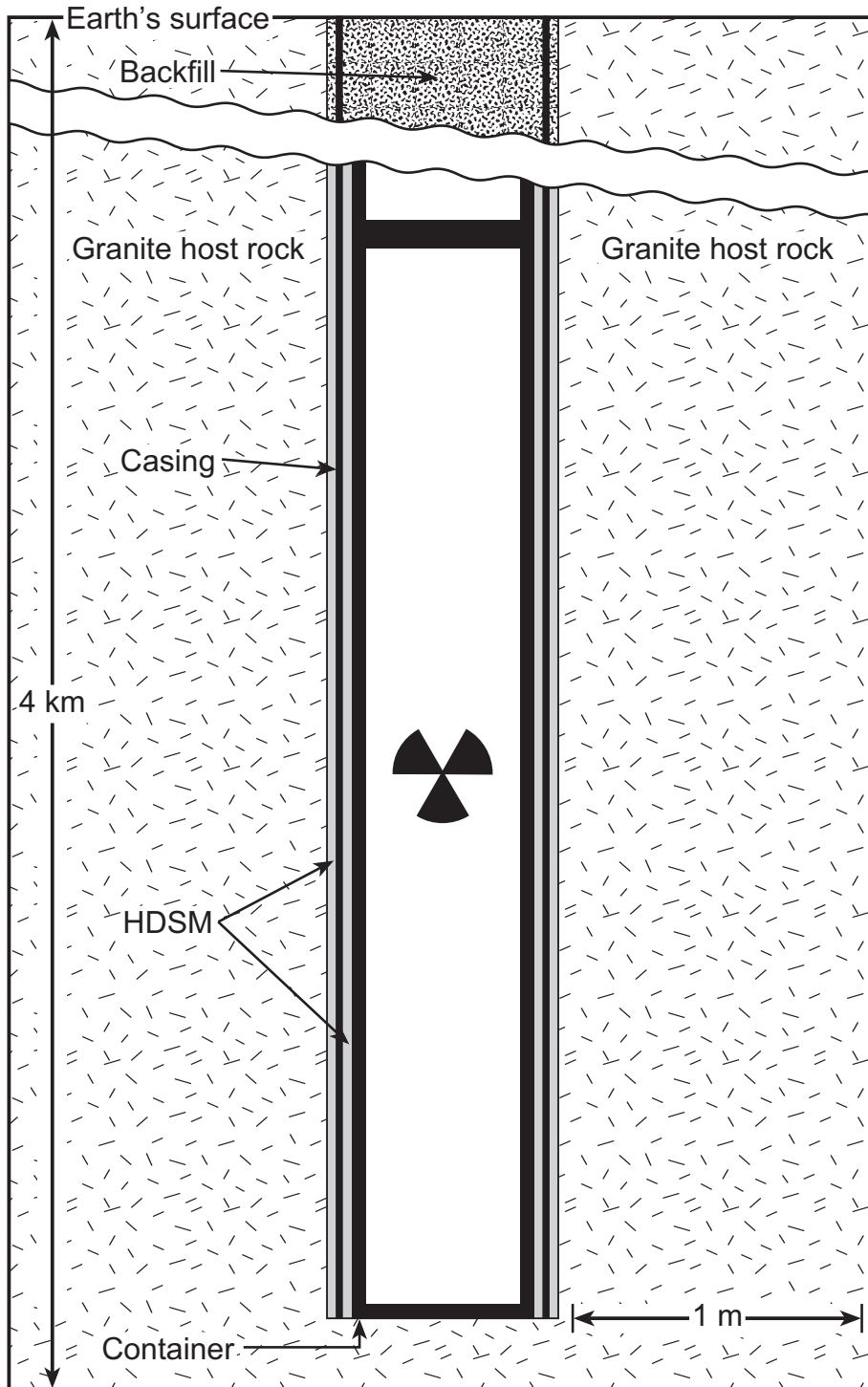


Figure 1

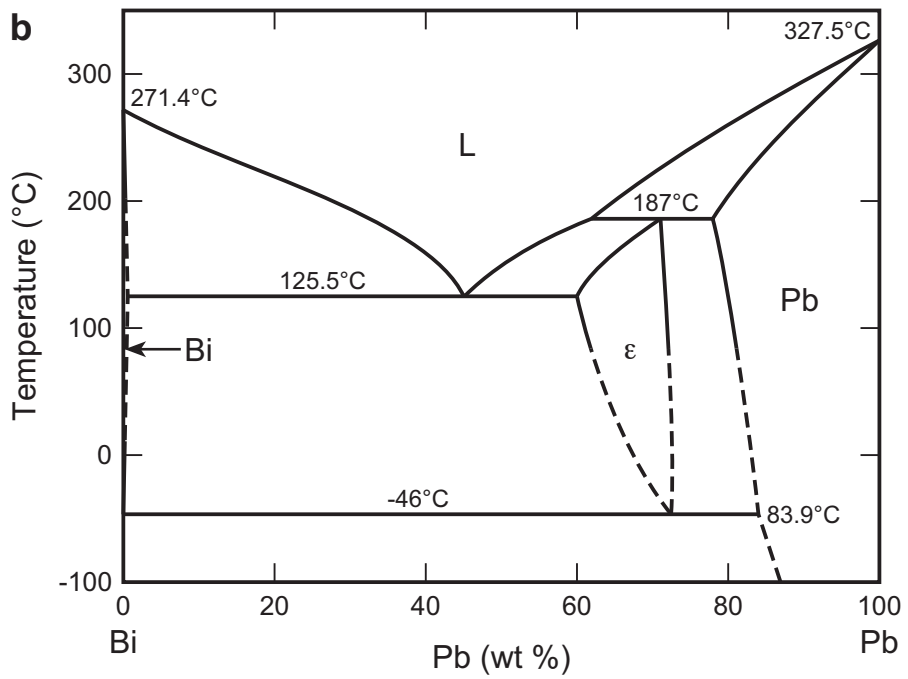
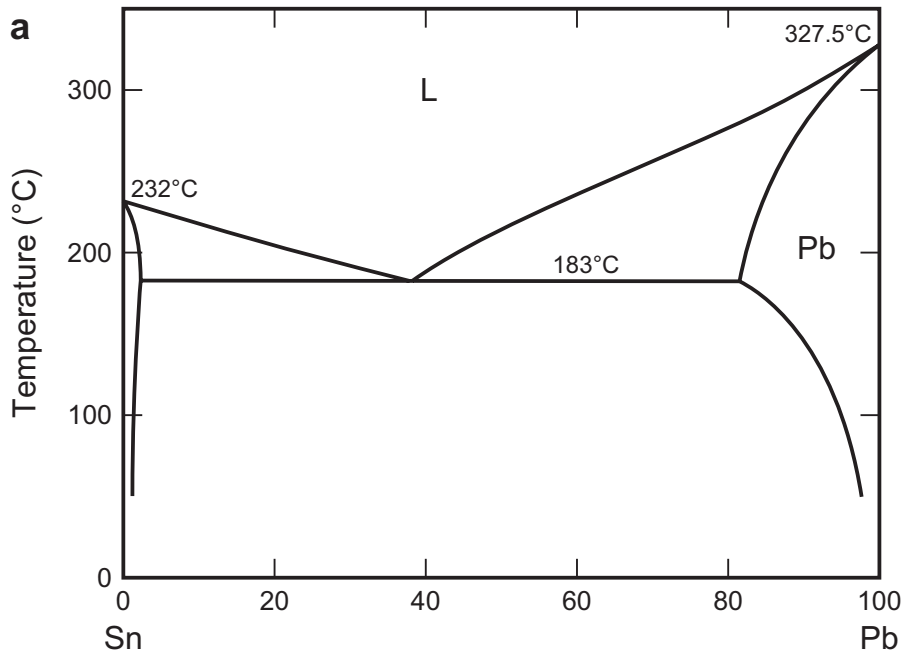


Figure 2

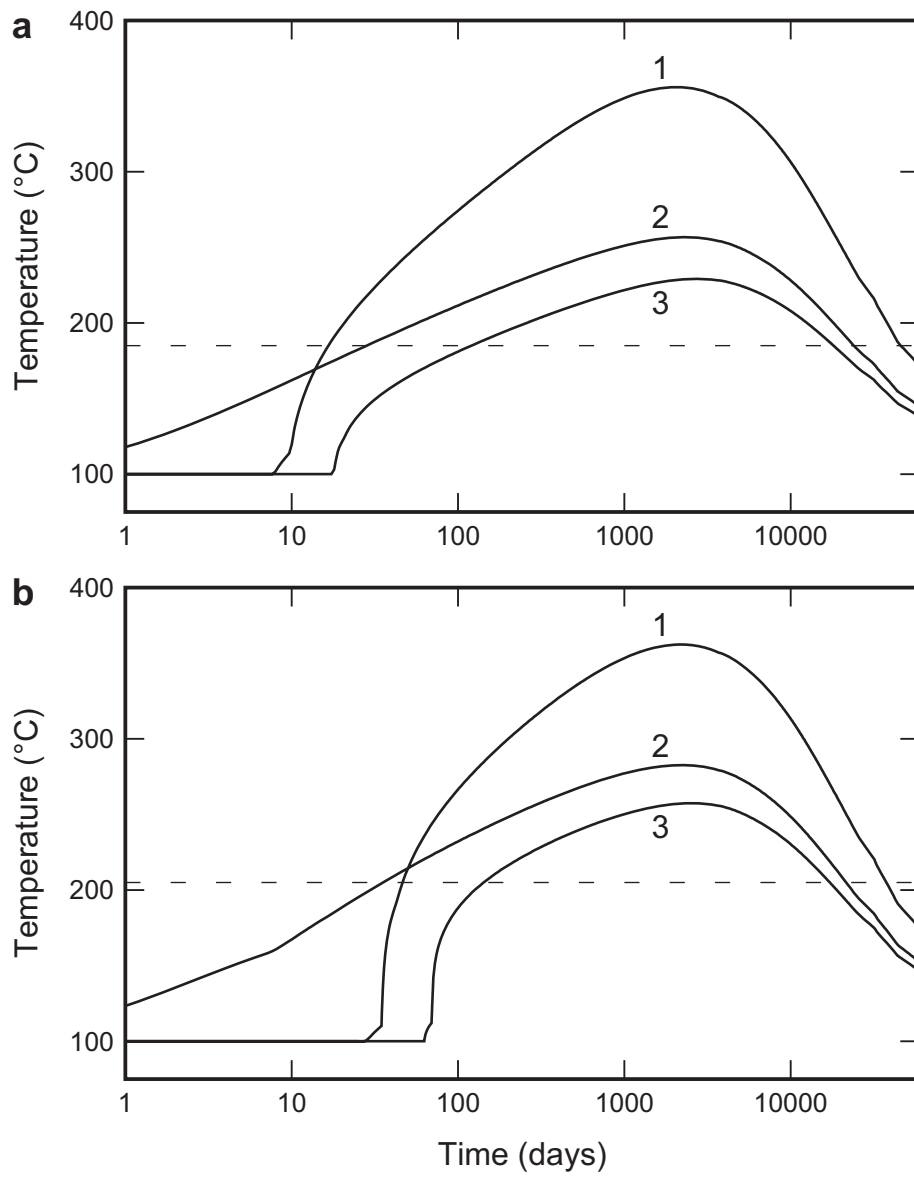


Figure 3

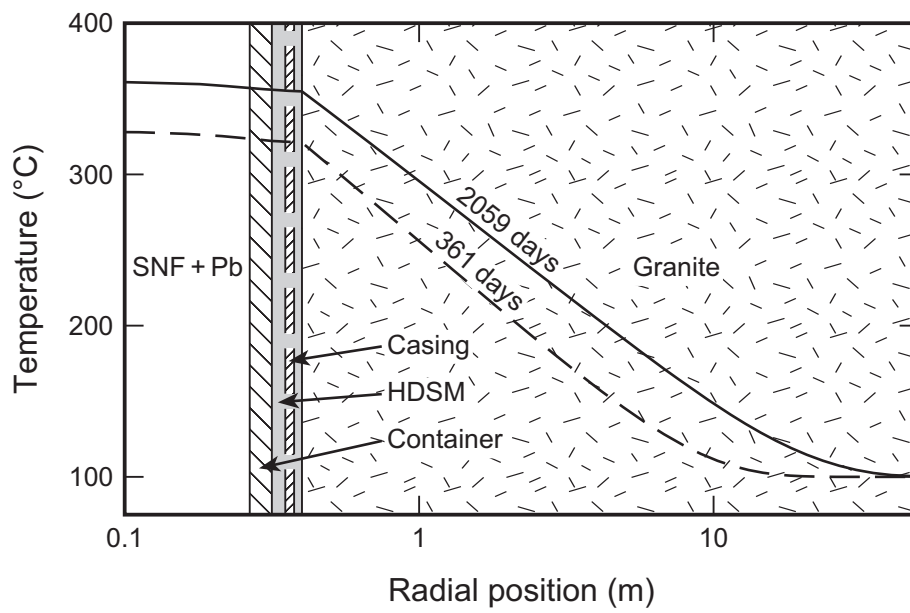


Figure 4