Spent fuel canister for geological repository: Inner material requirements and candidates evaluation

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ABSTRACT

One of the key aspects in designing Spanish spent nuclear fuel canister for geological repository is selecting the inner material to be placed between the steel walls and the fuel assemblies. This material has to primarily avoid the possibility of a criticality event once the canister gets breached by corrosion and flooded by groundwater. A detailed set of requirements for a material to fulfil this role in that environment have been devised and presented in this paper. With these requirements in view, eight potentially interesting candidates were evaluated: cast iron or steel, borosilicate glass, spinel, depleted uranium, dehydrated zeolites, haematite, phosphates, and olivine. Among these, the first four materials or their families are found promising for this application.

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1. Introduction

The present Spanish radioactive waste management policy [1] considers an open cycle for nuclear fuel and, accordingly, a geological repository – either in granitic rock or in clay formation – for the spent fuel. The preliminary design for the repository projects corrosion-controlled carbon steel canisters, with walls 100–120 mm thick that can simultaneously withstand mechanical loads up to 41 MPa (derived from bentonite swelling and hydrostatic pressure on hypothetically glacial period conditions with a 3000 m thick ice layer) and the foreseen effect of corrosion during the established confinement time [2–6], which is a minimum of a thousand years. Predicted durability, derived from corrosion studies carried out [7,8], extends over 10000 years. At later stages, the role of hindering radionuclide migration is mainly accomplished by the *bentonite barrier*, made up of compacted bentonite blocks placed between the canister and the host rock (see Fig. 1).

The canister is essentially an outer carbon steel shell into which 4 PWR (pressurized water reactor) or 12 BWR (boiling water reactor) spent fuel assemblies are inserted (see Fig. 2) (according to present spent fuel generation projections [9], over 80% of the canisters will be devoted to PWR fuel disposal). This configuration, planned for a maximum thermal load of 1200 W [2,6], leaves, according to our calculations, significant void space inside the canister. Although the exact dimensions of fuel assemblies may vary

slightly between power plants, when the representative assemblies' dimensions shown in Fig. 2 are used, the void space is 1.343 m³ for PWR canister, and 1.296 m³ for BWR canister. In other words, in a PWR canister, the void space is 81.1% of the total inner volume and 78.3% in a BWR canister. This void space, under repository conditions, would be filled with groundwater once corrosion penetrates the steel canister walls. As water is a good neutron moderator, and given a high water-to-fissile material ratio inside the canister, it is possible that self-sustained nuclear chain reaction could start, although the canister contains *spent fuel;* which, therefore, may be more appropriately designated *irradiated fuel.* Although there is no risk of explosion of any kind, the increase in heat generation within the repository associated to a criticality event may negatively affect the performance of the bentonite barrier (see Fig. 1). This can increase radionuclide migration rates at a disposal stage in which the canisters are no longer useful for confinement due to sustained corrosion. Additionally, the build up of fission products could be significant, as most of the original ones would have already decayed by this time, and spent fuel corrosion rates could also increase as a result of high temperatures [10–16]. In any case, a criticality event certainly impairs the performance of the repository, and so it should be avoided in any foreseeable scenario.

To achieve this, the canister will have to be properly designed with appropriate inner material. The main objective of this paper, therefore, is to provide a short list of material candidates that are considered suitable. The list could help identify the most promising alternatives for final selection or point out specific further research needs. Although in the final design different combinations of

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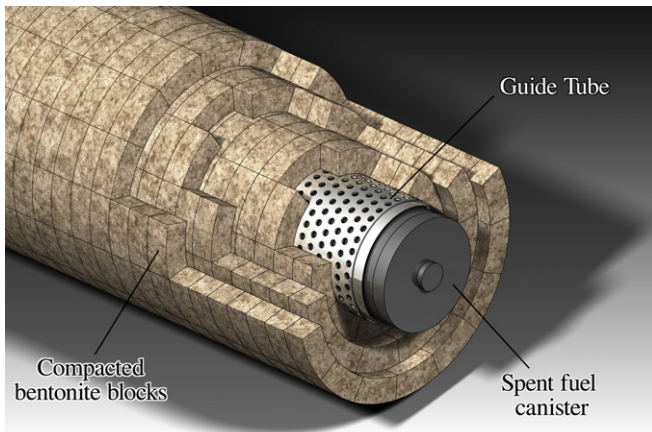E-mail address: zzrpuig@gmail.com (F. Puig).

Fig. 1. Computer recreation of spent fuel canister emplacement into a repository gallery. Bentonite barrier and guide tube are also shown.

materials can be considered for inner free volume fill, the initial objective is to select individual ones that could perform alone. If considered in particulate form, they might be accompanied by an inner steel frame to hold the fuel assemblies in place. The detailed study and evaluation of individual candidates presented here may also help to suggest potentially interesting combinations.

2. Material requirements

Prior to analysing different materials for suitability, it has been considered essential to develop a comprehensive list of evaluation criteria that cover all possible material properties or features related to fulfilling the demands of repository design. These criteria would help to quantify and evaluate the suitability of each candidate more objectively.

The evaluation criteria have been divided into five groups. The first group is focused on criticality avoidance, the second one on general desirable properties, the third on general undesirable properties, the fourth one on performance improvement features, and the last one on other parameters that, though not directly related to repository performance, could be certainly important in decision

making. In contrast to the last two groups, the other ones impose conditions (though not all strict simultaneous requirements), that must be either fulfilled or clearly made up by other favourable attributes. As a whole, the criteria should guarantee that the material can avoid critically along all repository life, while not harming any other confinement role, and, at the same time, assure its technical and economical feasibility.

2.1. Criticality

Usually, for preventing criticality, the required limit of effective neutron multiplication factor (k_{eff}) is 0.95. From the studies carried out for canisters of similar type and geometry, homogeneously filled, and containing 12 BWR fresh fuel assemblies enriched to 3.6% [17], it is inferred that at least 60% of the free volume will have to be filled to prevent k_{eff} reaching 0.95 limit if flooded. Existing data on criticality analyses for fresh fuel can serve as a conservative worst-case scenario in regard to two main uncertainty factors about canister fissile content. The first one is the need for disposal of PWR fuel with somewhat higher initial fuel enrichment than the 3.6% of BWR assemblies considered in Ref. [17], additionally to conceivable future fuel enrichment increases. The second one corresponds to the possible differences in fuel burnup between assemblies. Although such differences are, in principle, not expected to be huge, licensing procedure enforces the adoption of a conservative approach in dealing with fresh fuel if expensive accurate individual tracing, monitoring, and subsequent fuel composition calculations are to be avoided. These complex considerations for burnup credit are beyond the scope of the criteria presented in this paper, therefore the aforementioned fresh fuel canister criticality results [17] and those of Refs. [18–20] are used as reference. Further precision in criticality calculations is uncalled for, considering that the canister design is not fully established yet.

1. The first requirement for candidate materials, as implicitly stated, is to effectively fill a large portion of the canister inner free volume, that is, over about 60% of it. The effectiveness depends on different aspects, such as low porosity and material intrinsic geometry, that is, whether the material is in particulate form or in the form of manufactured blocks of certain shapes, or as single or multiple parts.
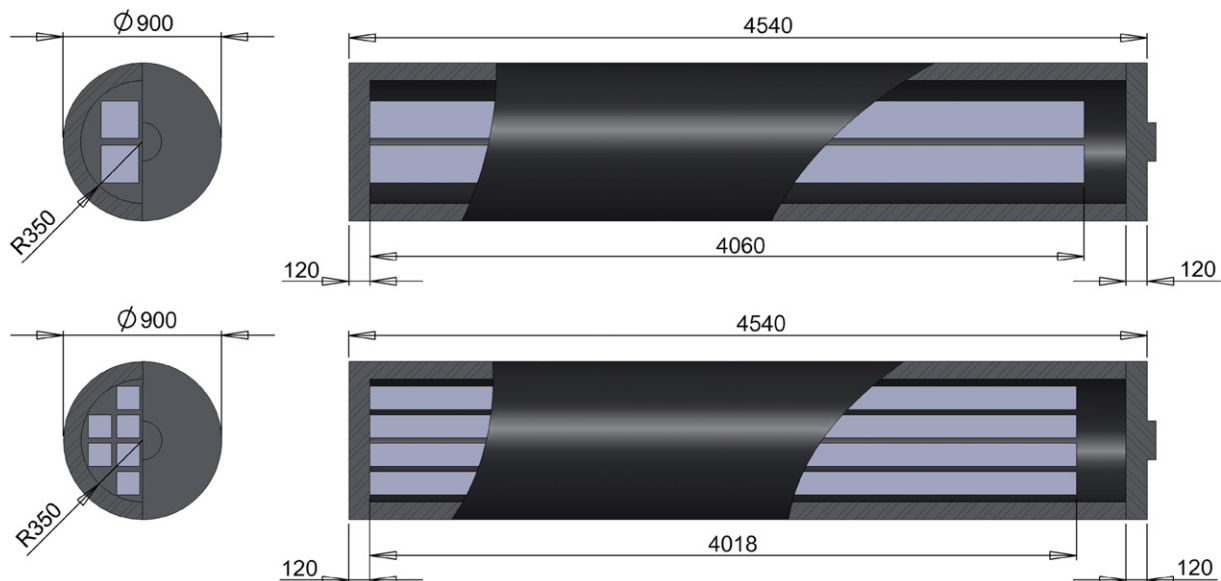

Fig. 2. Schematic broken-out section views of spent fuel canister and their dimensions (in mm). Above: canister holding 4 PWR 17 × 17 Westinghouse fuel assemblies. Below: canister holding 12 BWR 10 × 10 GB14 fuel assemblies.

- When dealing with particulate materials, it is relevant to consider if they can compact under their own weight after the canister has been filled. After the canister is sealed, the delayed compaction of the material should not reach 10% or more of its original volume. This is because, according to Agrenius [21], a portion 40 cm high of the fresh fuel assemblies extending above the fill material level is all that is needed to attain criticality in case the canister is flooded. Once the water manages to penetrate the canister, its effect, as a lubricant between particles, on facilitating compaction has also to be considered. In contrast to the conditions reported [21], the Spanish canister disposition is intended to be horizontal within the repository. Therefore, even if the material has compacted previously to some extent, the filling is likely to adopt a different, more favourable geometry when the canister is laid down. However, at this stage it would be difficult to assess accurately. The fact that the current canister design includes a gap of up to 25 cm between the top end of the fuel assemblies and the lid (see Fig. 2) could also significantly lessen the severity of this particular restriction. Nonetheless, Mennerdahl [22] pointed out that steel canister walls are a better neutron reflector than water. Therefore, a partially flooded horizontally disposed canister, with an empty gap on its inner upper section while the fuel is completely submerged, could involve a slightly higher neutron multiplication factor (k_{eff}) than a completely flooded canister.

2. For criticality avoidance, it should be useful if the material has significant neutron absorption capability.

3. It is also important to watch out for the presence of hydrogen, or any other light chemical element that could act as an efficient neutron moderator, the way water does.

4. Equally important is to ensure that previously specified criticality prevention demands are fulfilled throughout the repository life. This long-term general requirement of the material can be divided into three basic facets: radiation resistance, thermal stability and chemical stability.
   - For radiation resistance, it must be ensured that the essential material properties are not significantly affected, either by the total dose or by the dose rates foreseen for the inner material in the repository conditions.
   - Thermal stability requires, in the first place, appropriate thermal conductivity to avoid temperature values inside the canister that could affect any of the materials, particularly the fuel assemblies for which the rough maximum allowable cladding temperature is set at 350 °C [15,16] and about 400 °C for the fuel itself (due to accelerated corrosion rates above these levels). Obviously, the inner material itself should endure whatever temperature prevails in the canister as a result of its own thermal conductivity.
   - For chemical stability, the material should not be affected significantly by any corrosive substance in groundwater or by any other introduced during repository construction (namely corrosion resistance). Also, the solubility and lixiviation rates of the material should be low enough so that the inner material cannot be washed out and replaced by water prior to the fissile material itself.

2.2. General requirements to fulfil

Other general demands required to ensure the fulfilment of criticality criteria for the desired time extent are listed below:

1. The material should be in thermodynamic equilibrium with the whole system conditions and the materials in the repository, thus ensuring chemical compatibility.

2. The material should have good homogeneity between different batches to render performance assessment valid for all the canisters.

3. If the material is in particulate form, it should possess good rheological properties to ensure proper canister filling. For small particles, electrical surface charge effects, increasing friction, and the tendency to adhere to surfaces are to be considered.

4. The material has to be in such a form that it can be introduced or mounted inside the canister without the risk of damaging the canister or the fuel assemblies.

5. Fabrication, encapsulation, and any other process that may be needed should be allowed without any significant interference by the inner material. Welding the canister lid is one such important process susceptible to be affected by the inner material characteristics and derived conditions, and hence should be paid special attention to.

6. Disassembling the canister should be possible in case the quality control results after sealing are not satisfactory.

7. The canister should allow retrievability, if needed, for at least a few decades after its disposal.

2.3. General requirements to avoid

Under this head are brought together four issues which must be avoided for their negative implications either on repository performance or on the technical feasibility of using certain materials for this purpose:

1. Limited availability of the material.

2. Any material property with the potential to increase the corrosion rate of the canister, the cladding or the fuel itself.

3. Any potential capability to increase the radionuclide transport through the bentonite barrier or to chemically alter the barrier's basic properties, such as swelling, hydraulic conductivity, and diffusion resistance.

4. The propensity to retain significant amounts of air, especially on the surface of the materials in the form of small particles, that could lead to the formation of nitric acid by radiolysis, contributing to possible stress corrosion cracking (SCC).

2.4. Performance improvement properties

These properties could be divided into those oriented to improve the mechanical resistance of the canister and thus increase the time needed for corrosion processes to effectively breach through the canister's walls, and those aimed to improve radionuclide confinement.

1. For mechanical resistance improvement, it is desirable to use materials with moderate to high mechanical strength and/or those that could behave in an incompressible manner, without suffering significant particle deformation, fracture or crushing within the range of conceivable loads, so that they contribute to canister structural integrity, if inner material is properly packed.

2. For improving radionuclide confinement, sorption capability, especially of some key long-lived radionuclides, like ^{99}Tc, [129]I, [135]Cs, [241]Am and [243]Am, [237]Np, [226]Ra, [229]Th ([36]Cl, [94]Nb or [126]Sn could be included) and various uranium and plutonium isotopes such as [233]U, [234]U, [239]Pu, [240]Pu and [242]Pu, would be certainly useful. The same can be said for any other means to further reduce hydraulic conductivity through the canister. It should be noted that this would involve an additional benefit or safety margin, but still sorption is not included in repository

performance assessments, because it would require reliable quantitative data and full understanding of behaviour under repository conditions.

2.5. Other interesting properties

Under this head can be found some material traits, which, though not directly affecting performance, may become important in choosing between candidates that satisfactorily fulfil the requirements.

1. A well-documented long-term durability, in addition to laboratory testing results, either from natural geological formations' data, archaeological excavations, or some other relevant, particularly solid evidence.
2. Low material density to reduce the additional weight of the filling, thus allowing easier manipulation of the loaded canister and cutting down the expenses of the machinery needed.
3. Overall low cost of material, including expense for raw material acquisition and costs involved in processing and fabrication procedures that may have to be followed to attain the desired final form.
4. Material with good intrinsic radiation shielding properties to simultaneously reduce the total radiation dose received by the bentonite barrier (further ensuring its proper behaviour) and minimise the shielding or any remote manipulation techniques needed when handling the canister at any stage of disposal.
5. A material that inherently allows a relative simplicity along the whole process, from its obtention, treatment, and manufacturing to its transport, assembly, placing into the canister, and any other manipulation, including the necessary facilities and equipment for each step.

Besides the foregoing traits, there is another phenomenon related to material properties and repository performance that has been considered, but not used as an evaluation criterion, because there seems to be no consensus among scientific community on its implications for disposal. Such phenomenon is the production of hydrogen within the repository due to corrosion of certain materials, particularly steel. On the one hand, the presence of significant amounts of hydrogen is expected to maintain reducing conditions on the near field, thus preventing or slowing fuel corrosion [23–26]. On the other hand, high hydrogen concentrations are likely to cause steel embrittlement [27–29] and furthermore, if enough hydrogen pressure builds up, it could affect the performance of the bentonite barrier [29–32], as bubbles may leak through the clay, possibly allowing some amount of radionuclides to leak with them or opening preferential migration paths. Therefore, the possibility of inducing or preventing hydrogen generation should not be ignored, but it can be taken into account only when more convincing evidence about this topic is available.

Either way, this canister design is expected to generate significant amounts of hydrogen due to corrosion of its carbon steel shell. Whichever inner material is placed within the canister, it may not be of any use to prevent corrosion, because it remains isolated until the canister itself is breached by corrosion. Consequently, this issue should be basically considered from the point of view of avoiding additional hydrogen generation by the inner material or deliberately enhancing this phenomenon by appropriate material selection.

3. Candidate materials review

Initial selection of candidates for evaluation has been based on different criteria. These include previous knowledge and experience of the research team, similar works found in literature [33–38] and their conclusions, current designs for spent fuel canisters [15,16,39–51], the main basic requirements devised and already described for Spanish spent fuel canisters, and some key desirable properties.

The materials or material families studied for this purpose are cast iron or steel, borosilicate glass, spinel, depleted uranium, dehydrated zeolites, haematite, phosphates, and olivine.

3.1. Cast iron or steel

Cast iron has some interesting characteristics for its use as inner canister material for different reasons. In the first place, nodular cast iron has been selected, in preference to cast steel, and tested in similar designs by SKB [47,52–54] and Posiva [55,56] (Swedish and Finnish radioactive waste management companies, respectively). Cast iron (particularly SS 0717-02 [47,52] and its Finnish equivalent GRP-400 [55–57]) was preferred because it has better castability qualities, and it allows building a solid frame as an independent component; the frame allows an easy assembly of the canister, which does not introduce new problems in spent fuel insertion or later sealing. This solution, however, does not achieve very high occupation rates. Our calculations for Spanish canister, based on equivalent SKB cast iron insert design (with 230 mm wide channels in which to fit the fuel assemblies), give a remaining free volume occupation ratio of 54.74% for PWR canisters. Tighter insert channels and filling of their non-occupied length (Fig. 2) may be feasible if needed, to attain values of up to about 61% and 64%, respectively, although this might hamper the assemblies' insertion into them. A direct comparison with Swedish canister design for BWR is not possible, as the 160 mm insert channels of the latter would not fit into the somewhat slenderer Spanish canister. If *fuel channels* (BWR assemblies' metal casings for guiding reactor coolant flow) were previously removed from BWR assemblies, they could probably be inserted into 140 mm wide insert channels, which may still be practicable. In that case, occupation ratio would reach about 49%, and possibly 53.5% if non-occupied length of channels was completely filled. It has also to be noted that a solid frame does not fill all canister inner volume homogeneously, as it does not address itself to the task of filling free space between fuel pins (see Fig. 3). And it is precisely this volume between fuel pins, if flooded, which more efficiently contributes to the increase of k_{eff} and, consequently, to criticality risk. In addition, if cooling channels (or tubes) are to be added to the cast (as considered in Ref. [19]), to facilitate a more uniform solidification to reduce the risk of defects, the occupation ratios would decrease to about 44.5% and 48% for BWR and PWR, respectively, although the locations of these tubes would be in positions with lesser influence on criticality control.

If this option was not considered appropriate, steel shot could still be a candidate, as it is expected to fill all the volume with higher homogeneity if its size is wisely chosen (probably around 0.5 mm in diameter). Further, owing to its regular shape and high deformation resistance, no post-loading compaction should be expected. Despite the intrinsic geometrical limitations of small spheres in volume occupation, an appropriate combination of two different shot sizes can increase the theoretical volume-to-gap ratio to a rather high value. Experimentally, values of around 70% of occupation have been found feasible owing to their good rheological properties [48,58], and are likely to be enough for adequate safety margins. Furthermore, the possibility of combining a cast iron frame with steel shot fill on its fuel channels – a more complex process, though – can also be conceived. But, the use of steel shot has some problems. It is difficult to mix spheres of different sizes homogeneously and in the correct proportion. Also, it needs to be assessed if the spheres remain mixed adequately and permanently. It should be noted that these considerations for steel
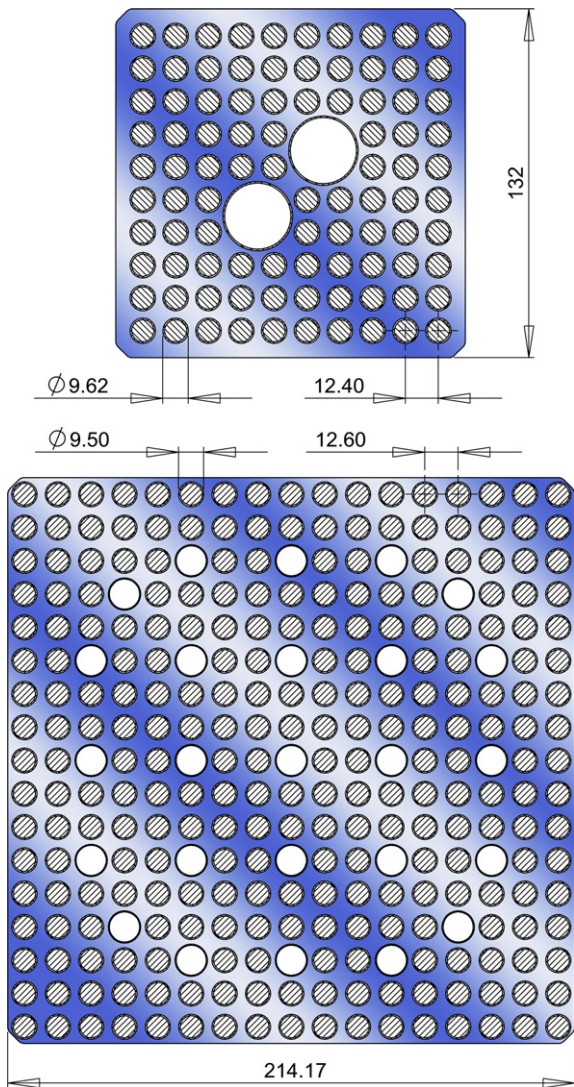
Fig. 3. Schematic cross section views of representative fuel assemblies with most relevant dimensions (in mm) displayed to illustrate gaps between fuel pins. Spacer grids, placed at regular height intervals to maintain assembly geometry, are not shown in the figure. Above: BWR 10 × 10 GE14 fuel assembly. Below: FWR 17 × 17 Westinghouse fuel assembly.

ite, goethite, or haematite) could also play an important role in maintaining reducing conditions and stimulating radionuclides sorption, thus hindering their mobility [60–64].

3.2. Borosilicate glass

Borosilicate glass (BSG) with a density of about 2250 kg/m^3 is more than three times lighter than steel. Among all the glass types, BSG has been selected as a candidate mainly because of its relatively good mechanical properties, its high neutron absorption capability (boron absorption cross section is approximately 750 barns for thermal neutrons), and its widespread use for high-level radioactive waste conditioning, which offers extensive valuable experience in this field. In addition, for similar reasons, it has been previously chosen as filling on the preliminary reference concept for the Spanish repository, and so it is essential to properly evaluate its suitability.

As with any other small spherical particulate material, the volume occupation ratio that can be achieved with borosilicate glass beds of the right size could be rather high, as shown previously for CANDU assemblies [48] and for PWR assemblies using steel shot [58]. This, coupled with an intrinsic high neutron cross section, offers good guarantees for criticality avoidance. Its reported very good leaching resistance and high thermal and radiation stability [37,65] are also favourable aspects, but its low thermal conductivity (around 1.2 W/m K) may pose a problem. Therefore, detailed canister thermal models that also include the selected inner canister gas fill properties should be developed to assess if enough heat evacuation rates are met. Rough calculations indicate that a global thermal conductivity of about 0.1 W/m K for the inner volume would probably be enough for the fuel cladding to remain below the mentioned 350 °C limit [15,16], although significant margin for uncertainties is recommended. Helium possesses a conductivity of 0.176 W/m K at 400 K and atmospheric pressure, and so this requirement could be expected to be fulfilled, if chosen. If another gas is finally selected, owing to other criteria, and the final conductivity in combination with BSG is found to be insufficient, other solutions, like the addition of thermal shunts made of aluminium or copper, could be conceived. Considering that these thermal shunts would be needed only during the first decades or centuries while the spent fuel cools down to lower residual power levels, the desired material requirements would probably be easy to meet, but it should be ensured that they will not interfere with any other process in the repository environment. All these difficulties are foreseen to be surmountable, but in the event of other suitable candidates being available, they probably are not worth the trouble because they may substantially increase the complexity of the whole disposal process.

It has been shown, concerning possible performance improvements, that the alteration layer formed on the surface of high level waste BSG matrix has important retention capabilities [66,67], thus hindering the release of certain radionuclides from the glass. Nevertheless, this capability is observed under conditions which significantly differ from those of borosilicate glass beds inside the canister, and as such, the extent of glass corrosion and the absorption capabilities of its alteration layer for external radionuclides present on groundwater will have to be evaluated to conclude that a significant positive effect would exist in this regard.

Glass beds, as any other particulate material considered for this application, is already foreseen to involve somewhat higher process complexity (compared to that of cast iron frame), as high packing efficiency needs to be ensured during the filling process, which would probably require induced vibration [48]. The combination of canister, spent fuel assemblies and filling material should be tested to find optimal vibration frequency, amplitude and duration for proper compaction, while simultaneously ensuring that

shot are rather generic and are applicable to most other candidate materials in particulate form that will be later reviewed.

Cast iron or steel has good radiation resistance [59] and good thermal conductivity, with values ranging from about 30 to 38 W/m K for the selected metals, which is considered more than enough for this purpose even in the form of steel shot. Regarding neutron absorption capability, iron has relatively low cross section for thermal neutrons. The same applies to most of the other chemical elements present in steel alloys, and they are only present in small amounts. Therefore, it acts more as a reflector than as an absorber and, as such, no positive evaluation can be made in this regard. Even so, boron-steel alloys (with high neutron absorption capability) could be used, but there are uncertainties concerning the possible selective lixiviation and washing out of boron upon steel corrosion, and so they do not appear to be interesting options.

Another prominent feature of iron is its capability to maintain reducing conditions in the near field which, in addition to preventing significant fuel corrosion rates, stimulate actinide precipitation and reduce mobility of most redox potential sensible radionuclides [40,60]. Furthermore, its corrosion products (like magnetite, sider-

this vibration is not harmful to the fuel assemblies' integrity, whose mechanical resistance could have been weakened by the long residence period inside the reactor.

3.3. Spinel

From the spinel family, spinel ($MgAl_2O_4$) has been chosen as a candidate for inner material because some recent developments by Nucon Systems Inc. have shown some potentially interesting applications for this material. The feasibility of obtaining large sintered pieces of this ceramic material [68], and welding them seamlessly using microwaves [69–71], opened the possibility of considering the spinel solid frame (similarly to the cast iron one) as an additional option to the more conventional particulate form. Spinel has also been considered previously in canister fabrication [36,72] although basically as a sprayed coating over metallic or metallic ceramic-lined containers.

The combination of its high mechanical strength (with Young's Modulus of 190 GPa, about the same as that of steel), outstanding corrosion resistance (only a few millimetres every million years [73,74]), good radiation resistance [71], high thermal stability (fusion at 2135 °C) [72], exceptional thermal conductivity for a ceramic material (around 15 W/m K) and the recently devised technique for seamless welding [69–71] has enabled it to be considered a material suitable to build the full spent fuel canister. Some uncertainties because of its fragility may prevent its use as a container, but these are certainly much less important for its intended use inside the canister. Even so, some potential technical problems – possible shrinkage, and internal stresses or other difficulties in producing solid blocks of the required size – regarding the fabrication of components may have to be resolved. And, this must be done on a rather large manufacturing scale prior to the material being considered a serious candidate in this solid form.

Regardless of these difficulties, spinel may still be used as small spheres because of its still interesting properties in this form. However, it probably cannot be used as fine powder (similar to the one used for sintering), as electrostatic repulsion may interfere with proper packing efficiency of such small particles. Additionally, as any other very small particulate, they may retain significant amounts of air and humidity on their surfaces, which could be difficult to eliminate, and that may contribute to undesirable SCC by nitric acid. Although the amount of nitric acid that could reasonably be formed inside the canister seems to be small and probably harmless [39], its formation should be avoided if possible.

3.4. Depleted uranium

Depleted uranium (DU) becomes an interesting material in view of its intrinsic physical properties and other technical, strategic, and administrative issues that could constitute additional advantages.

The United States Nuclear Regulatory Commission has recently (on 18th January, 2005) decided to consider DU a form of low-level waste [75], which may involve its final disposition as such, with significant cost. However, some uncertainty still remains regarding its classification in other countries and how the classification may change in future. Nonetheless, DU is still a huge energy source that could be used in fast breeder reactors in the future, like irradiated fuel itself, which could be employed for energy production after reprocessing. These considerations imply that even if today there is a need to dispose of these energy resources owing to lack of appropriate technology to exploit them and/or social pressure to find a final disposal solution, it is likely that these resources would need to be recovered in the future. If so, it may be an interesting option to store them in a way that could ease future retrieval and reduce recovery costs, without compromising safety or security, that is, storing them together, using DU as spent fuel canister filling material. This would also reduce disposal costs compared to the cost of two different facilities, which regardless of any possible future use, is probably the main issue today, except safety.

As regards its properties and performance under repository conditions, DU exhibits some interesting traits that make it appear as a candidate by itself. The foremost among them is its neutron absorption capability. If spent fuel, with its residual uranium enrichment and plutonium content, is placed in combination with a significant amount of DU, the mean concentration of fissile materials would be significantly reduced.

The fissile content calculations, worked out after taking into consideration the canisters projected dimensions (see Fig. 2), amount of spent fuel to be held, and remaining free space for eventual DU fill, show similar results for PWR and BWR assemblies, despite differences in the selected values for initial enrichment and burnup (according to usual real values), and different volume occupation ratios of the canister. Their conclusion is that the average fissile content in the resulting mix can easily be reduced to about 0.72% (the natural uranium enrichment in ^{235}U) or lower. As the mix also contains a number of fission products that act as neutron absorbers, despite not being homogeneously mixed, the resultant whole is expected to behave similarly as natural uranium does with regard to criticality, being unable to reach criticality under any possible configuration and in combination with any amount of groundwater, which is obviously light water. In fact, criticality on natural environments is not possible below 1 wt% ^{235}U in ^{238}U [76–78].

DU may also conduce to maintain reducing conditions in the near field [79], which decisively contributes to the stability of the spent fuel, and further prevents its lixiviation by means of saturating uranium content of groundwater that may get inside the canister; groundwater that in granitic environment should be already saturated.

DU could most probably be used inside the canister as a particulate fill in the form of small spheres of DUO_2. This ceramic material has essentially the same chemical properties as those of spent fuel, which is mostly UO_2 with only a small fraction of fission products, actinides, and their descendants. This serves to hinder the separation, by any chemical means at any stage of the disposal, of the irradiated fuel (and particularly its fissile content in the form of uranium) from the inner material that prevents its possible criticality, thus ensuring permanent safety. Plutonium, also present in the spent fuel UO_2 matrix, is incorporated into the UO_2 crystal structure, preventing its possible release until the matrix itself is destroyed. Additionally, plutonium is less soluble than uranium under reducing repository conditions [19,80] and so it is unlikely that it would get separated, specially because ^{239}Pu (the main fissile plutonium isotope) decays to ^{235}U with a half-life of 24110 years, which somewhat limits the extent of hypothetical ^{239}Pu lixiviation prior to conversion to ^{235}U. The other plutonium fissile isotope likely to be present in significant amounts in spent fuel is ^{241}Pu, which decays with a half-life of only 14.4 years to ^{241}Am, an important neutron absorber (around 700 barns for thermal neutrons and [n,γ] reaction [81]). Uranium is therefore the most important chemical element to be considered when dealing with long-term criticality issues on the repository.

With regard to its thermal conductivity, values ranging from about 6.9 to 8.1 W/m K are found for solid UO_2 at 500 K and 400 K, respectively [82], which are within the expected range inside the canister. UO_2 particulate conductivity in He atmosphere tests averages about 1 W/m K [58,83], for the foreseen packaging efficiency and temperatures. The expected temperature, according to 3D finite element computer calculations carried out on Ref. [6], would be under 150 °C for this conductivity value.

Other forms of DU, besides DUO_2, can also be considered for the spent fuel canister. These are mainly uranium silicates [84–86], DU_3O_8, pure metallic form and cermet [87–89], a kind of composite material that encloses DUO_2 powder on a steel matrix. These forms are considered to present no clear advantages over ceramic DUO_2 beds for the inner canister; instead, they add some performance uncertainties, and hence would not be considered for inner material selection analysis unless some of their features happen to be relevant in the future.

3.5. Dehydrated zeolites

Zeolites are considered a low density, high availability material with very high stability. They are also very resistant to corrosion and can withstand high temperatures without their properties being affected. Additionally, many studies showed their high absorption efficiency and retention capabilities toward some key radioactive elements, such as caesium [90–96], iodine [35,97], thorium [98–104], uranium [104–108], radium [105,109] or plutonium [96,110], and other elements like technetium [96], americium [96,111] or strontium [91–95,112].

Natural zeolites contain significant amounts of water by adsorption and composition; the water should be removed before its utilization inside the canister to prevent internal corrosion. This can be accomplished by heating the material up to 350 °C, when most of its water gets liberated, thus minimizing the possibility of corrosion at the canister loading stage.

Unfortunately, the highly porous nature of the material (reaching specific surface values up to $1000 \, m^2 \, g^{-1}$), which could be an advantage for radionuclide sorption, is a serious problem for effective volume occupation. Even if it is possible to achieve good compaction of the zeolites' particulate material, its intrinsic porosity, ranging from 40% to 70%, would result in a rather low ratio for total effective volume occupation. Considering that most or all of this inner and intergranular porosity would be filled with water, once the canister has been breached, the filling requirement for criticality avoidance will probably be impossible to achieve. Furthermore, the very low thermal conductivity of zeolites, about 0.20–0.30 W/m K [113], may compromise appropriate heat evacuation by the spent fuel, despite its high thermal resistance, as these values are near to the roughly estimated minimum for the whole inner material volume, and its particulate form could negatively affect this parameter. Additionally, air retention by zeolites on canister loading is expected to be very difficult to avoid, involving a possible risk of SCC by nitric acid.

Finally, some experimental results by Wang et al. [114,115] show that the zeolites are highly susceptible to irradiation-induced amorphization, resulting in drastic reduction of their sorption capabilities. The dose required for their complete amorphization by beta irradiation is found to be 10^8 Gy, which is quite low for its intended use as HLW immobilization matrix studied in those works. In its application as canister fill, irradiation dose rates are clearly lower, and highly dependent on considered position owing to self-shielding, but accumulated doses after long storage periods may also induce some serious amorphization.

3.6. Haematite

Haematite (α-Fe_2O_3) is very abundant and present in different kinds of mineral deposits. It possesses high sorption capabilities for heavy elements, like uranium [116–121], plutonium [122,123], neptunium [123,124], americium [125] or strontium [126,127]. Additionally, under reducing conditions, it can fix hydrogen while transforming to magnetite (Fe_3O_4).

This material could be processed to obtain near spherical particles, which are expected to fill the canister with high enough efficiency despite their sub-spherical shapes and slight porosity, up to a small percentage, depending on the degree of weathering. Nonetheless, rheological properties and resultant canister volume occupation should be experimentally tested to ensure the appropriate fulfilment of requirements.

Neither thermal stability nor conductivity would pose any problem, with thermal conductivity values ranging typically from 11.3 to 14.7 W/m K, which are foreseen to be high enough to fulfil, even when used in particulate form.

Per contra, uncertainty about radiation resistance, and general long-term behaviour of haematite under the reducing conditions of repository environment, and in the presence of bentonite clay and steel, could be of some concern; despite its well proven durability under other natural conditions. Very limited information has been found in this regard, but the possibility of radiation-induced dissolution of haematite has been pointed out [128,129], although in conditions that may not fit with those of the repository. On the other hand, the behaviour of magnetite from possible alteration of haematite would be relevant. In a granitic repository environment, magnetite may already be present as an accessory mineral, and hence the groundwater would already have been saturated with it; therefore, no further dissolution of magnetite would be expected [33]. In addition, the capacity of magnetite to buffer redox conditions could prove useful. In any case, the extent of long-term alteration or dissolution, the properties it could affect, and the influence of total dose and dose rates are key aspects that should be studied and well understood before considering haematite as a real candidate.

3.7. Phosphates

Among naturally occurring phosphates, those of the apatite group, typically $Ca_5(PO_4)_3(OH,F,Cl)$, are the most abundant and chosen here as reference. They are considered for this application mainly for their known ability to incorporate lanthanides and actinides, like uranium [130–138], thorium, and americium [136,139], into their lattice. Some experiments showed important sorption capability for strontium [137–140], iodine [35], plutonium [136,141] and neptunium [142], and even for caesium [130,140] in some cases, which could contribute to a better confinement of radionuclides in the repository. Apatite presents high thermal stability and also very high dissolution resistance, except at extreme pH values.

Hydrogen, though present in the composition of the phosphates of the apatite group, is present only in rather small amounts, and hence should not be a serious concern. On the contrary, its highly porous nature (between 48% and 58% according to de la Cruz et al. [35]) poses a much greater difficulty in meeting criticality avoidance requirements, in addition to its low thermal conductivity, which ranges from 0.19 to 0.23 W/m K [35] or from 1.25 to 1.39 W/m K [143] depending on the samples.

Another possible trouble with phosphates appears to be related to their behaviour under irradiation. Phosphates are usually associated in nature with some radioactive elements, but which would be its behaviour under the much higher ionizing radiation doses expected inside the canister is still uncertain, as little information is available on apatite behaviour under heavy radiation fields.

3.8. Olivine

Olivine $(Mg,Fe)_2SiO_4$, particularly that from Lovasjärvi intrusion (Finland) with fayalite (Fe_2SiO_4) fraction between 0.39 and 0.58, for a total FeO content of 28%, has been previously proposed by Hellmuth et al. [144,145] as canister filling material. Its main attractiveness lies in its sorption capacity, and Fe(II) content, which allows reducing conditions to be maintained in the near field.

Existing results show a high sorption capacity for U(VI) and other reducing conditions sensitive radionuclides [146–148]. Additionally, its redox buffering capability has been studied as a function of pH in the environmental conditions expected in a geological repository [149].

Olivine is expected to be available in particulate form (sand-like) with adequate rheological properties, capable of obtaining high enough packing efficiency [48] owing to their wide range of particle size, despite their sub-spherical shapes. As olivine porosity is low, fulfilment of volume occupation criteria is not considered a major problem; nonetheless, olivine's achievable packing efficiency and its amenability to form and its capacity to remain as a homogeneous mixture of different particle sizes during all the operations and disposal should be tested. Additionally, if olivine is to be presented in this form, it should be checked if it can meet the requirements in the light of the potential problems mentioned earlier with small particulate materials.

Serpentine, which appears on alteration of olivine by water in the presence of silicon, and thus will be present in a granitic repository environment [33], is a mineral which possesses similar capabilities as those of olivine with regard to sorption and reducing conditions fostering. Also, both olivine and serpentine swell on contact with water. Although dissolution of olivine or serpentine is not expected under the chemical conditions foreseen in the repository, further studies may be required to understand the possible influence of irradiation on its behaviour.

Thermal stability of olivine is not a problem; on the other hand, its rather low thermal conductivity, around 4.5 W/m K [150] at the temperature range inside the canister, could be of some concern, though not critical. The low cost and rather low density of olivine could be its other distinct advantages. Finally, as noted by Oversby [33], olivine usually contains impurities of other materials found in the rocks where olivine is mined. So, the amount of these impurities and their possible effect on canister or repository performance should be evaluated.

4. Discussion

There are several topics related to the materials evaluation that require further consideration, and are addressed in this section.

As a general issue, some of these materials do corrode under repository conditions, which may seem to make them unsuitable according to initially stated requirements. However, the corrosion criterion also involves other aspects and not all of these materials fail to fulfil it after all. For cast iron and steel the actual corrosion rates are too slow to endanger any short-term demands, like retrievability, heat transfer capabilities during the first few decades or centuries, or even additional positive contributions like increasing canister mechanical strength for up to thousands of years. This is particularly true as the inner material will not be exposed to significant corrosion agents until the canister gets breached and water flows inside. The long-term requirements of these materials are different and focused more on their stability, enabling them to hold their place around the spent fuel and not allowing the formation of gaps susceptible of being flooded with water. In this regard, iron corrosion products, like magnetite, siderite or goethite, swelling during the corrosion, are highly stable under the long-term repository conditions and so they are foreseen to perform well. BSG is neither completely resistant to lixiviation, although dissolution rate is anticipated to be rather slow under repository conditions. If criticality was dependent on boron concentration alone, the lixiviation and eventual washing out of boron content could become a serious problem. However, the packing efficiency achieved with borosilicate glass beds should be high enough to avoid criticality, and the resulting products from glass alteration

are likely to be stable enough to avoid large amounts of water around the spent fuel. DUO_2 may not be completely corrosion resistant, but shows very high stability under reducing conditions that prevail in underground formations which are over 500 m deep. Additionally, groundwater on granitic rock formations should already be saturated with uranium, further limiting its possible lixiviation. In any case, DUO_2 would behave the same way in this regard as irradiated fuel itself, ensuring its presence and assigned function around the fissile material as long as the latter remains in place.

Similarly, while cast iron or steel does not fulfil thermodynamic equilibrium criterion under repository conditions, their corrosion products do, being also true for haematite. It also applies to olivine and the serpentine that would appear in granitic repository environments, which is expected to fulfil its long-term role as a stable material.

The issue of borosilicate stability, due to possible devitrification, deserves a detailed discussion, being also highly linked to radiation resistance, thermal stability, and corrosion resistance requirements. Glasses are a well-known family of materials that evidently are not in thermodynamic equilibrium, as are crystals. Nonetheless, they remain stable under certain conditions, but it is difficult to assess if these conditions are maintained in the repository. This question has been extensively studied owing to the widespread use of BSG and other glasses of similar composition for high level waste immobilization. Despite these studies, it cannot be said that BSG long-term behaviour under foreseen repository conditions is fully understood and, therefore, that can be predicted with reasonable certainty. Many works can be found in literature that point out the main uncertainties and problems of this issue. To begin with, it has been previously shown [65,151,152] that radiation damage and high temperatures can devitrify glass, and thereby negatively affect its chemical stability, besides inducing glass fracturing and thus globally increasing corrosion rates. Experiments carried out by Ewing et al. [38] indicate a variety of problems affecting glass corrosion due to irradiation, although at doses probably significantly higher than those expected for inner canister material. It has also been noted [153,154] that glass corrosion and long-term behaviour is highly dependent on its composition, for example aluminium content [155]. Composition is, therefore, an important factor [153], and so every glass considered for this application should be carefully analysed. The effect of other materials present in the engineering barriers also needs to be evaluated, as other studies [154,156,157] pointed out its relevance. Some experiments were carried out on the influence of bentonite [158], magnetite [157,159,160] and the general solution composition [161] on glass corrosion and their possible deleterious effects. The glass alteration layer, which can present strong retention capabilities for actinides [66], also seems to be highly dependent on composition [162]. Other works [37,65], instead, conclude that the results obtained make BSG an excellent candidate with good long-term durability. To clarify this issue further, more detailed studies on BSG corrosion are needed, under temperature and radiation conditions representative of those in repository environment, and in the presence of engineering barrier materials and its corrosion products.

Present experience on canisters with cast iron frame (or insert) of similar design indicates that criticality avoidance requirements are not fulfilled with this geometry (at least for PWR assemblies) if burnup credit is not taken into account, whereas substantial safety margins are achieved if it is [18–20,163,164]. Oversby [165] also concluded that no credible scenarios for criticality could be found on the similar Swedish granite repository concept. If greater safety margins are desired, or burnup credit calculations and precise evaluation for each fuel assembly are to be avoided, then steel shot or other particulate materials would be the alternative as canister fill.

Simplicity, as most of the general requirements stated, is a concept difficult to evaluate accurately at this stage of design. Cast iron, as an already tested solution, is considered to involve relative simplicity, allowing separate fabrication of components and fairly easy assembly and sealing on-site, while all other candidates are considered mainly as particulate fills, involving packing (probably needing the use of vibrations) and other procedures required when using particulates. These may include storage, drying of the material if required, minimizing the amount of air they could carry, and even possible decontamination if canister quality tests are not passed and reopening is needed to extract the irradiated fuel. Most of these operations will probably have to be carried out on emplacement, increasing the on-site infrastructure needed. In spite of these drawbacks, no serious difficulty had been identified so far for any of these materials in this regard. Further evaluation of these parameters should be carried out once detailed specifications are available.

5. Conclusions

Based on Spanish spent fuel canister's preliminary design, assigned roles and all relevant information gathered, a complete and detailed set of requirements, desirable properties and other interesting features had been devised for its inner material or fill, with the main purpose of preventing a hypothetical canister criticality event once placed inside the repository. Based on these criteria, many potentially interesting materials as canister fill have been evaluated and the following conclusions drawn.

The first conclusion drawn from materials review is to discard dehydrated zeolites and phosphates from the list of candidates mainly because of their inability to safely avoid criticality, and other disadvantages arising from their very low thermal conductivity. Haematite and olivine constitute the second group of materials that have no intrinsic property which prevents them from being used as canister fill, but still present some problems or gaps in knowledge that need to be resolved before their use. The most important issues with olivine are related to uncertainties on its long-term behaviour under radiation fields, its likely impurities and their effects, and perhaps its low thermal conductivity. The main uncertainties regarding haematite are also related to long-term performance and radiation effects. Additionally, the most important feature of haematite for being considered as a candidate is its ability to suppress hydrogen production, but this seems to be of virtually no use when placed as inner material inside the canister. If any of these materials are considered interesting later, owing to some other aspects, it may be worthwhile to carry out further research and testing for this purpose, although it will be difficult to completely eliminate the drawback caused by the lack of experience with them in this field.

The remaining four material candidates (cast iron or steel, borosilicate glass, spinel, and DU) adequately fulfil the requirements, as far as this analysis shows, and may be used successfully. Nonetheless, some difficulties, uncertainties, and controversies still remain and require more detailed testing. With cast iron, the main problems are related to criticality margins without considering the burnup credit of the fuel, and possible undesirable effects due to the generation of significant amounts of hydrogen as a result of its corrosion. For BSG, the main problems arise from its low thermal conductivity, which could lead to excessive fuel temperature if not compensated by other means, such as thermal shunts. Additionally, borosilicate's long-term corrosion is not very well established [38,166,167] under high temperature and radiation conditions with regard to possible devitrification, which can even lead to preferential lixiviation of boron after the glass sustains significant corrosion [168]. This possibility could mean a serious difficulty if

criticality avoidance relies on boron concentration; however, if sufficient packing efficiency can be achieved and maintained, boron content would become less relevant, acting only as an additional safety measure. It should also be noted that many of the experiments conducted to study these phenomena were oriented to performance assessment of glass matrices for HLW immobilization, involving higher temperatures than those expected inside the canister, higher doses from radiation originated within the glass matrix, and considering much shorter relevant time spans than those related to spent fuel disposal. Nevertheless, this fact points to another interesting possible candidate material specifically for plutonium immobilization, that is, the gadolinium zirconate ($Gd_2Zr_2O_7$) [169–172]. Gadolinium zirconate offers much better radiation resistance and long-term stability than BSG [166,169,171,173–175], and the binding of gadolinium (the most effective neutron absorber known, with a thermal cross section of about 49 000 barns) to the crystal structure prevents its selective lixiviation. The only identified disadvantages could be thermal conductivity values similar to those of borosilicate, although somewhat higher (about 2 W/m K at expected canister temperatures) [176], and possibly higher costs.

Spinel, as a highly resistant ceramic material, has also a very good record. Its main problems may arise from fabrication process difficulties, lack of adequate experience using it, and possibly somewhat higher production costs as compared to those of some other alternatives, but none of them seem insurmountable. Depleted uranium dioxide, on its part, also exhibits a very good forecast performance, but for possible difficulties foreseen in legal or political issues about DU management, cost of the conversion from UF_6 to UO_2, possible processing problems (further testing would be desirable), and final canister weight.

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