### Encapsulation of ILW Metals by Melting Technology - 15360

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## ABSTRACT

This paper outlines how encapsulation using melting technology can be performed and describe the positive effects in a handling and repository perspective. Based on a feasibility study of a conditioning method for reactor core components and other ILW metals with a high content of long-lived radionuclides, the paper gives an overview of the concept and how the method can be facilitated.

The paper covers identification and analysis of alternative techniques that supports the aim from a repository perspective, which is to strengthen the defense in depth for long-lived waste to be disposed. Embedding of radioactive metallic ILW components can isolate the activated component from corrosion and leakage of radionuclides into the repository barrier systems. By implementing a system that delays the mobility of nuclides the leakage to the biosphere can be delayed and by then decreased significantly. Conditioning by embedding has potential to strengthen a repository concept significantly. It may in certain cases also reduce the requirements on the repository barriers. In addition to the above, embedding also serves as an efficient radiation protection, which simplifies post treatment handling of the waste. The concept can be applied on a wide spectrum of metallic components.

Positive effects in a handling and repository perspective can be achieved if low level metal not subject to free release is used as embedding material, since conditioning of metallic ILW waste can be carried out at significant lower dose rates and without increasing the total amount to be disposed. The paper discuss why there are embedding alternatives for ILW metals that are considered to be both technically feasible and reasonable in cost both with regards to investment and operational cost. A cost-benefit analysis as well as a risk analysis is discussed in order to support the feasibility assessment of a facility for encapsulation of ILW metals by melting technology.

## **INTRODUCTION**

The radioactive waste from the Swedish nuclear industry is categorized into three major groups:

- Short-lived low and intermediate-level waste
- Long-lived low and intermediate-level waste (ILW)
- High-level waste (used fuel).

There is yet no final disposal solution for the long-lived low and intermediate-level waste. This situation is applicable not only for Sweden for also for other countries.

Swedish Nuclear Fuel and Waste Management Co. (SKB) started a project, Disposal of long-lived waste (SFL) concept study in the beginning of 2011 with assignment to develop alternative methods for final repository and conditioning of long-lived low and intermediate-level waste and to compare them. The goal was to by 2013 present a comparison between identified repository concepts. The study [1] should be of such a level that SKB can make decisions of which concepts that are to be further investigated in a safety analysis. As a part of this concept study, Studsvik was assigned to investigate whether melting of

long-lived low- and intermediate-level metallic waste is technically possible and if so, what is required to build and operate such a facility. Low-level metallic waste can be treated in the existing melting facility in Studsvik and other facilities on commercial basis. These facilities however are not equipped for melting of ILW. In the assignment from SKB, Studsvik found melting of metallic waste from the ILW category neither technically feasible nor cost effective [2].

During evaluation of melting, embedding of radioactive components was expected to be more technical feasible and advantageous from a long term safety perspective. Studsvik investigated if there are alternative techniques for conditioning of metallic ILW waste. The result of this investigation is presented in current paper.

# DESCRIPTION

Current study presents the concept of embedding radioactive metallic components in a corrosion resistant material. A qualitative comparison of different embedding materials is made. Three embedding techniques are presented: i) embedding by die casting, ii) embedding by use of Hot Isostatic Pressing (HIP) powder technology and iii) direct loading into thick-walled containers. Each embedding alternative is presented from a technique, safety and current application perspective. A short reasoning about cost for the three different techniques is also given.

### **Embedding concept**

Embedding of radioactive metallic components in a dense, non-brittle and corrosive resistant material protects the component from corrosion in final repository. Leakage of radionuclides from repository to biosphere can thereby be both delayed and reduced under accepted limits, in order to be clear that this technology is suitable from a regulatory point of view. Except for the corrosive protection, embedding also serves as an efficient radiation protection which simplifies post treatment of waste. An example of an embedded segmented component is visualized in Figure 1.

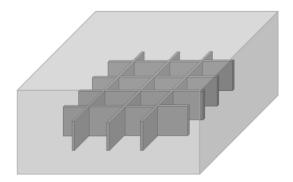


Fig. 1. Sketch of an embedded piece of a BWR core grid.

Above mentioned conditioning advantages are expected to be obtained for a broad spectrum of dose rates and component geometries. The conditioning concept would complement todays melting of metallic components with lower contamination levels and form an overall concept for conditioning of all radioactive metallic components in a nuclear plant, see Figure 2.

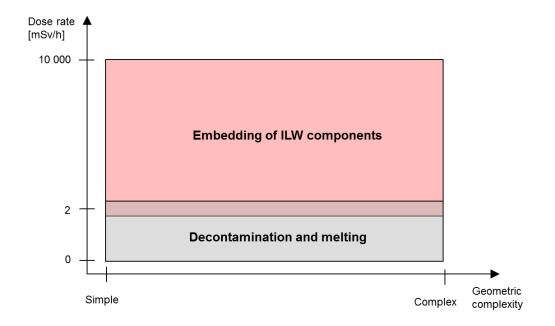


Fig. 2. Potential application window for embedding of metallic components in the ILW range.

### **Embedding material**

Embedding material should if possible be corrosive resistant in repository environment to prevent leakage of radionuclides to biosphere. A corrosive resistant material also gives the nuclides in the component enough time to decay to more harmless levels once corrosion of component starts. To outlive the geological movements in the bedrock and the temperature changes with time in final repository environment, the material also has to be non-brittle and dense to avoid cracks from tensions. Other practical parameters such as: dose absorption, life cycle perspective, weight and cost were also compared.

Corrosion rate for carbon and stainless steel has been investigated for prevailing environment in final repository. The main part of corrosion take place under anaerobic and chloride present environment resulting in a similar corrosion rate of 0.1  $\mu$ m/year for both carbon and stainless steels [3]. Regarding corrosion rates for Ni and Ti based alloys there are no data for corrosion in repository environment. Regarding corrosion of oxides, the corrosion is closely related to surrounding pH value. The approximated idem value for final disposal environment (concrete present) around the steel is about 12.5. As conventional glass is a rather acidic oxide the resistance for corrosion in prevailing alkaline environment was assumed poor compared to that of the other materials.

In this brief comparison, metals like carbon- and/or stainless steel were determined to be most advantageous both from a long-term safety perspective and from a practical waste handling perspective.

#### Long term safety - delayed corrosion due to anode corrosion of shield

In SKB Preliminary safety assessment report for deep repository for long-lived low and intermediatelevel waste, transport of radionuclides from final repository to biosphere was calculated to estimate

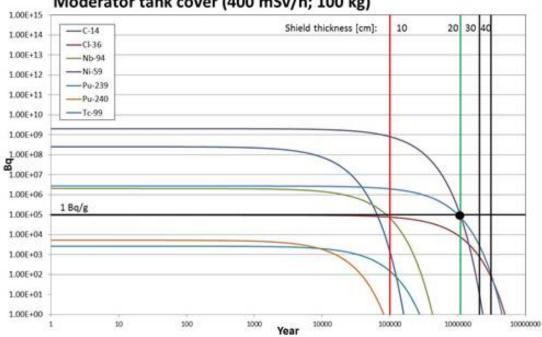
resulting dose to critical group peaking close to 1E-5 Sv/y [4]. Cl 36 and Mo 93 are the dominating nuclides at the time for release to biosphere (1E5 - 1E6 y). Calculation is based on direct corrosion.

Embedding of ILW have the possibility to delay corrosion and by then leakage of radioactivity from final repository to biosphere. Embedding material should if possible outlive the decay of dangerous nuclides in the component until significant activity reduction has been achieved.

Figure 3 shows the decay for long-lived nuclides for two different components with different geometry and contamination levels in the ILW category. Vertical lines illustrate approximated corrosion start for different shield thicknesses. Horizontal line shows when the specific nuclide activity concentration has reached 1 Bq/g. Embedding material was assumed to be carbon steel with a corrosion rate of 0.1  $\mu$ m/year As can be seen in Figure 3, it is theoretically possible to design a shield thickness so that all nuclides have decayed to harmless levels at time for corrosion start.

If leakage could be delayed a million years, the dose to critical group would principally be delayed the same number of year. Giving another million years for decay, also the dose from radionuclide at leakage would be significantly lower.

Hence, embedding of metallic components as conditioning concept has the possibility to ensure long-term safety.



# Moderator tank cover (400 mSv/h; 100 kg)

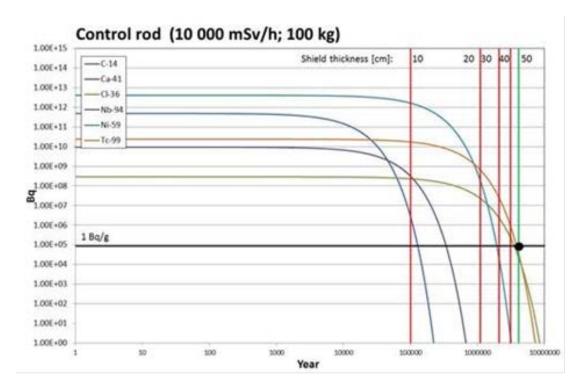


Fig. 3. Decay for component specific nuclides for two core components in the ILW range with different contamination and geometry; a) moderator tank cover and b) control rod. Vertical lines illustrate corrosion start for different steel shield thicknesses (for a corrosion rate of  $0.1 \,\mu$ m/year).

## Long term safety - delayed corrosion due to less are/volume ratio

Also, embedding of components will for some complex geometries, give a lower area exposure for corrosion, see example in Figure 4. A low area/volume ratio prolongs leakage of radionuclides through corrosion and is thereby favorable from a long-term safety point of view.

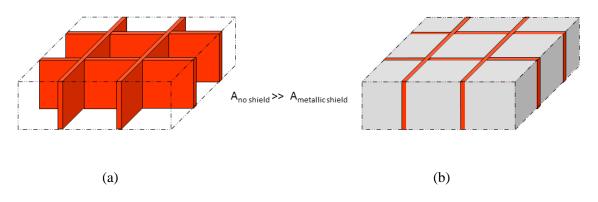


Fig. 4. Schematic sketch of corrosion area for a piece of: a) non-embedded and b) embedded part of a core grid.

## Long term safety - problem with possible leaking of package lid avoided

Above mentioned advantage with delayed corrosion through anode corrosion of a protecting shield surrounding the active components could also be obtained by having a thick steel wall of the repository waste container. If a safe sealing of lid without cracks can be guaranteed both concepts have potential to ensure long-term safety by delayed corrosion. Still, the embedding concept has the advantage of avoiding the negative effect on long-term safety from a leaking package lid.

To summarize, embedding of metallic components has potential to ensure long-term safety. Nuclides can be kept safe from corrosion and leakage until they have decayed to low levels. Also, for some components a lower area exposure for corrosion will further delay corrosion. Embedding of metallic radioactive waste compared to direct loading into thick-walled repository waste containers also has the advantage of avoiding the negative effect on long-term safety from a leaking package lid.

## **Disposal volume**

Adding more material for embedding also means adding volume. If low level waste just above clearance limits can be used as embedding material, a total positive effect on disposal volume can be expected. As neither the conditioning technique nor the vessel design for HLW is yet decided and as the shield thickness of component will vary a lot depending on contamination level, the effect of direct embedding on storage volume is left out of discussion.

## Waste handling – reduced surface dose rate

Except for the corrosive protection, embedding also contributes to a significant decreasing of dose rate, associated with decreasing of personnel doses. Significant reduction of dose rate will be obtained for the complete component register of metallic ILW waste. This will simplify further handling of components like packing and transportation of RWC container after conditioning.

### Waste handling - suitable waste forms

As the nuclide content for the different components varies a lot, future application of the embedding concept could customize the shield thickness to better suit the embedded components need for corrosion and radiation protection and thereby avoid over- or under dimensioning of waste packages. Embedding of components with a thick shield questions the need of an extra metallic package to load the components in. Instead, if the final outer geometry after embedding is standardized, the lump itself could directly fit into the final repository container.

### **Reuse of low level waste**

Current melting facility for contaminated metals in Studsvik (SMA) generates metal containing activity higher than limits for clearance. This material has to be disposed as radioactive waste. If this metal was used as embedding material, decommissioning of nuclear power plants could be carried out from a better life cycle perspective, see Figure 5.

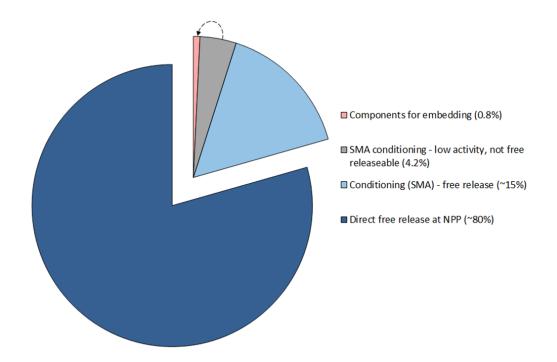


Fig. 5. Approximation of weight proportion of metallic waste with different contamination levels for a BWR reactor.

## **Embedding techniques**

Applied embedding techniques need to create a homogeneous and evenly thick shield around the active components to ensure long-term safety due to delayed corrosion and radionuclide leakage to biosphere. A uniform shield is also important from a waste handling perspective, avoiding hot spots.

Three techniques for embedding metallic ILW components in a protecting metallic shield were considered to have potential to meet the demands for a homogeneous and evenly thick shield; i) die casting for embedding, ii) HIP (Hot Isostatic Pressure) powder metallurgy and iii) direct loading into thick-walled containers. Future conditioning of metallic ILW waste will include transportation and need for temporary storage before and after conditioning. Regardless of used embedding technique, segmentation of radioactive components before embedding will have to be carried out. The list of needed equipment in the ILW metal treatment facility is quite extensive due to the complex and remote handling that is needed in the facility. No detailed specification of equipment is made. Focus is set to generally present the process flow for the three alternatives. In the following, each embedding alternative is presented from a technique, safety and current application perspective.

### Die casting for embedding - Technique

Die casting of low active melt in a mould filled with segmented metallic ILW parts is one technique to embed components in a homogeneous shield around the component. Die casting of the metallic shield would need some kind of positioning advice in the mould to ensure that components are centered in the ingot after solidification.

Before die casting the mould with segmented parts are recommended to go through a drying step to avoid vapor explosions from liquid in contact with melt. It is important that metal enclosure is homogeneous without voids and pores. Segmentation of component before die casting could be carried out to favor a compact filling of mould. Also casting and filling procedure can be designed to favor compact filling of mould.

Filling of liquid hot metal around solid radioactive components risks to melt the component and thereby cause a radioactive mixing zone in the metal shield. Unless melting temperature of the component is higher than the casted melt, component surface would melt and liquid radioactive melt could be transported due to natural convection within the liquid melt. Also, surface particles and oxides attached to the component surface risk to float within the melt. See Figure 6 for illustration.

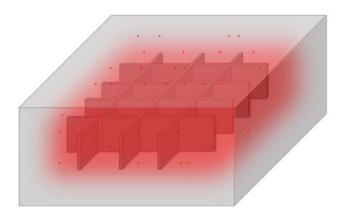


Fig. 6. Sketch of a piece of a BWR core grid enclosed in a metallic shield by die casting.

To further evaluate the spread of radioactivity within the metal enclosure more extensive calculations and experiments are needed, this is beyond the scope of current study.

#### Die casting for embedding - Safety

Handling of liquid melt around 1 500 °C is dangerous from a safety point of view. Vapor explosions of liquids coming into contact with melt or breakthrough of ladle shell are examples of mishaps frequently occurring in melting facilities. Die casting of melt around highly radioactive components also involves safety risks. For example, tipping of the mould would result in extensive clean-up efforts. Also, loading of radioactive component into the mould would have to be remote and in radiation shielded area. Risk analysis of die casting is left out of current study. Safety risk for die casting was roughly assessed to manageable in current study since treatment not includes handling of radioactive melt.

#### Die casting for embedding – Current application on radioactive waste

The authors found one similar application of above described technique in Germany [5]. Core components were segmented and loaded into a MONOLITH container made of cast iron with 150 mm thick walls. After loading, a lid was mounted onto the container. Thereafter components were dried by routing hot dry air through the container via the lid openings. A filling funnel was mounted after drying on one of the lid openings and low active melt (< 200 Bq/g) was used for filling. Containers were cooled

and transported to interim storage.

## Powder Metallurgy HIP for embedding - Technique

Hot Isostatic Pressing (HIP) is a process to densify powders or cast and sintered parts in a furnace at high pressure (100-200 MPa) and at temperatures from 900 to 1 250 °C. The gas pressure acts uniformly in all directions to provide isostatic properties and 100 % densification. The powder is filled into a dense form which is tightly closed after filling. The form is then positioned into the HIP and pressurized. Capsule is removed afterwards by machining.

Centering of contaminated parts of metallic waste in the ILW range using HIP technology could be obtained by filling a separate cylinder with waste and concrete for minimizing the void. The inner cylinder is later centered into the outer form. Powder is then filled around the inner and outer cylinder form and furnace is pressurized.

For application of HIP technique for conditioning of ILW waste, it is important that the shield thickness can be guaranteed to avoid corrosion and hot spots on the waste cylinder.

## Powder Metallurgy HIP for embedding - Safety

HIP technique has the advantage of not handling a liquid melt. If any failure arises, neither active components nor the embedding material would be in liquid form. This strongly lowers the safety risk and simplifies the clean-up efforts compared to die casting as previously discussed. Still, handling of component would have to be remote to secure that dose to workers is limited. HIPing means pressurizing a chamber to very high pressures, a possible failure causing an explosion could be one risk with HIP-processing. The authors, have in dialogue with HIP furnace suppliers and HIP users not found one such documented event of explosion. Explosion risk was therefore assessed to be low. Safety risk for HIPing was roughly assessed to manageable and lower compared to die casting since treatment not includes any handling of liquid melt.

## Powder Metallurgy HIP for embedding - Current application on radioactive waste

HIP technology has been one of the tested techniques in the development of fuel disposal. In 1978, ASEA performed an experiment of embedding an active fuel rod into a ceramic container of alumina using HIP technology. The experiment was carried out in Robertsfors [6].

The Institute of Materials Engineering (IME) in Australia, is the home of ANSTO's Synroc waste form technology. Synroc is a suite of technologies for immobilizing various forms of intermediate- and high-level radioactive wastes for disposal using the HIP technique. Synroc is basically a ceramic made from several natural minerals which together incorporate into their crystal structures nearly all of the elements present in high level radioactive waste [7]. HLW waste is mixed with Synroc powder and filled into a form which is pressurized in a HIP furnace.

## Direct loading into thick-walled containers - Technique

The final presented alternative for conditioning is also the most technically simple alternative. Metallic ILW waste is segmented and thereafter directly loaded into a thick-walled container. After loading, the

tank is filled with concrete to remove void in the container. Container lid is thereafter positioned and sealed by welding.

#### Direct loading into thick-walled containers - Safety

Conditioning by segmentation and direct loading into thick-walled containers do not include any risk from handling of liquid melt or explosion risk as for previous discussed techniques. One risk from direct loading into thick-walled containers could be dropping active components during loading, or dropping filled container after loading.

Risk analysis of direct loading into thick-walled containers is left out of current study. However, conditioning by segmentation and direct loading into thick-walled containers is expected to be the best alternative from a safety point of view.

#### Direct loading into thick-walled containers - Current application on radioactive waste

Loading into metallic tanks is the currently applied technique for existing metallic ILW waste. Inner metallic box with holes is filled under water. After being filled, it is remotely lifted into BFA tanks and the tanks are finally sealed with bolts. This technique is also named the zero alternative for conditioning of metallic ILW waste.

Above described technique of direct loading into thick-walled tanks, which are filled with concrete and sealed is not internationally applied for disposal of metallic ILW waste in similar environment.

## **Cost estimates**

A first coarse economical comparison of the two first described embedding technique: die casting and PM HIP technology was made. The direct loading alternative is expected to have the lowest investment and production cost and was not given further attention in this cost estimation. The cost for production of thick-walled containers needs better be assessed before it can be confirmed as the most economical alternative. Such estimation is however left out of this study. The cost was estimated for design and licensing the facility, the initial investment, the operation, and the decommissioning.

#### DISCUSSION

A comparison of the three presented embedding techniques is thereafter made. The three different conditioning alternatives are compared and discussed with respect to: i) long-term safety, ii) disposal volume, iii) waste handling, iv) total life cycle, v) technical complexity, vi) safety, vii) cost and viii) long term operation.

A facility for final conditioning ILW for SFL will be needed before disposal can take place, and no such facility exists in Sweden today, meaning that a zero alternative does not exist. The major choice is then to decide when to start and what to aim for. Based on experience from the waste treatment facilities at Studsvik and elsewhere, it can be concluded that it takes several years to establish new treatment methods, and to build enough competence, facilities, and supporting systems.

Three different embedding alternatives for conditioning of internal core components in the ILW range were discussed and compared in this study: i) die casting, ii) PM HIP and iii) direct loading into thick-walled containers. Comparison was made mainly with respect to their effect on: i) long-term safety, ii)

disposal volume, iii) waste handling, iv) total life cycle, v) technical complexity, vi) safety, vii) economical cost and viii) long term operation. Most technical feasible and cost effective alternatives were finally selected based on the comparison.

It is important to mention that current study only makes a first brief comparison of three different embedding techniques. More detailed facility specifications and cost estimates are needed. Further studies supporting assessment of safety barriers, final storage volume approximations and detailed cost estimations are also suggested.

### Long-term safety

All three embedding alternatives are considered to have potential to be accounted as safety barriers.

Thick-walled containers, is the best alternative to guarantee a uniform thickness of the shield and also non-active shield. As long as centering of active components using HIP technology can be controlled HIP is considered the second best alternative for a uniform thickness of the shield and also non-active shield. As previously mentioned, HIP technique has the advantage that the radioactive component itself will not melt and be spread into the shield as for die casting. Loose surface oxides and particles do no risk to float as for die casting. The isostatic pressure will most likely lock particles to the component surface.

Using HIP technology for embedding would most likely gives the best quality of the protecting shield itself. In general, equal or better material properties are obtained provided by the fine and homogeneous isotropic microstructure from HIPing.

Embedding by HIP or by die casting both has the advantage of avoiding the negative effect on long-term safety from a leaking package lid.

Also, embedding by die casting or by use of HIP technology will give a lower area/volume ratio for some components which favors delayed corrosion and leakage of radionuclides to biosphere. This advantage on long-term safety is not obtained for the direct loading alternative.

All three embedding alternatives were considered to have equal conditions for minimizing void in the waste packages.

## **Disposal volume**

The effect on final storage volume for embedding compared to packing into standardized tanks is left out in this study. A more close application of the protecting shield to the radioactive component is however considered to have a beneficial effect on disposal volume compared to the thick walls of the repository waste containers serving as corrosion protection. Also, if low level waste just above clearance limits can be used as embedding material, a total positive effect on disposal volume can be expected.

## Waste handling

All three embedding alternatives are expected to give similar low dose rates from conditioned waste packages.

## **Total life cycle**

Earlier mentioned advantage with reuse of low active metals as embedding material, can technically be obtained for all three presented embedding techniques. However, it is expected to be easiest applied for the die casting alternative. Liquid low level melt from current melting facility in Studsvik can be directly casted around the segmented metallic components.

Reuse of low level waste can also be obtained for both HIP technology as well as the direct loading alternative. However, reuse for these alternatives would be more complicated.

Low active metals could be used for powder production in HIP application. That would require a separate gas atomization facility handling radioactive melt for powder production. Low active metals could also be used as raw material for production of cylinder forms. The benefit from reuse of low active melt for HIP technology is not considered economically realistic due to extensive need of new equipment.

Also, low active melt could be used as raw material for production of repository waste containers. This would also need large equipment investments and is not considered economically realistic.

Die casting for embedding of metallic ILW components is considered the best alternative for reuse of LLW. Die casting is for this reason also considered the best from alternative from a total life-cycle perspective.

## **Technical complexity**

Die casting was considered to be the most technically complex alternative due to the high risk of handling liquid melt and thereby extra need of safety barriers in the case of a mishap. The technical solution avoiding radioactive melt from components to be spread into the embedding shield must also be tested out.

### Safety

Direct loading into thick-walled containers was assessed to be the safest alternative from an operational point of view. Once radioactive components are loaded into the package and the lid is mounted the dose rates are significantly reduced. There is no risk coming from handling of liquid melt as for die casting or explosion risk from pressurizing a HIP chamber.

HIPing was considered to be the second best alternative from a safety point of view.

#### Cost

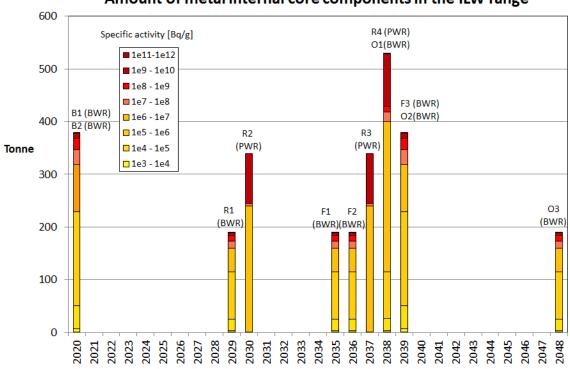
The cursory cost estimate indicated both highest investment and operation cost for the HIP alternative, followed be the die casting alternative. The direct loading alternative is expected to be most economically favorable. Direct loading into thick-walled containers is assessed to be the most favorable alternative from a cost perspective.

It is important to remember that the cost approximation made is a first rough estimation with great uncertainty. It should therefore not be used as decision making for selection of most economical alternative. Again, it only gives a first indication.

#### Long term operation

As can be seen in Figure 7 the waste stream of metallic ILW waste from Swedish NPPs will be of instantaneous character with longer gaps of ILW waste.

The availability of technically advanced conditioning techniques like die casting or HIP may give rise to an increase in demand from international customers to use facilities in Sweden, which may have the effect of leveling out the demand for metallic ILW conditioning.



# Amount of metal internal core components in the ILW range

Fig. 7. Waste streams of metallic internal core components in the ILW range from decommissioning of Swedish NPPs [2].

#### Technical feasible and cost effective alternatives

All three embedding alternatives are considered to have potential to be accounted as safety barriers. All three alternatives are also expected to be technically feasible.

Only two alternatives however are considered to be enough cost effective for a future application; i) die casting and ii) direct loading into thick-walled containers.

#### **Further studies**

Primary, die casting and direct loading alternatives are suggested to be more detailed described so that better cost estimates can be performed.

The technical design of the facility should preferably ensure a homogeneous shield around the component.

Possible effect from melting of active component is suggested to be investigated.

Sealing of package lid must also be further investigated before the direct loading alternative can be accounted as safety barrier.

Also, repository waste container design needs to be worked out.

## CONCLUSIONS

- There is yet no final disposal solution in Sweden and many other countries for the long-lived low- and intermediate-level waste and therefore no WAC.
- Embedding of radioactive metallic ILW components protects the component from corrosion and leakage of radionuclides from repository to biosphere can thereby be both delayed and decreased.
- Conditioning by embedding has potential to strengthen the long-term safety.
- Embedding also serves as an efficient radiation protection which simplifies post treatment of waste.
- Embedding of metallic ILW components is a broad conditioning concept suitable for a wide spectrum of dose rates and component geometries.
- If the low level melt is used as embedding material, conditioning of metallic ILW waste can be carried out from a better life-cycle perspective.
- Three different embedding alternatives for conditioning of internal core components in the ILW range were discussed and compared in this study; i) die casting, ii) PM HIP and iii) direct loading into thick-walled containers.
- All three embedding alternatives were considered to have potential to be accounted as safety barriers.
- Two alternatives were considered to be both technical feasible and cost effective for a future application; i) die casting and ii) direct loading into thick-walled containers.

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