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Qualification of neutron absorber material families for dry storage systems

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ABSTRACT

The need for qualification of neutron absorber materials has become an urgent topic in the field of dry fuel intermediate storage systems in Switzerland due to requalification of existing cask designs for increased content requirements. A special issue is the material which is used to manufacture the baskets of casks for spent fuel, mainly borated aluminium. This material has to fulfil several requirements for example to ensure subcriticality, to sustain stability under mechanical loads and to dissipate heat. These requirements also depend on the design of the casks and have to persist over the long period of intermediate storage under continuous high thermal load.

This paper starts with the definition of the necessary requirements by the help of numerical analysis of accident and service conditions at storage sites with specific respect to regulatory environment. In addition the applied methods for the qualification of the long term material behaviour will be described from the perspective of the Swiss regulator. Special challenges of the current qualifications are the variations of boron content and geometrical dimensions within the same family of material.

In addition there are manufacturing limitations which can also have impact on the material behaviour and properties. These aspects require an adapted supervision of the manufacturing process. Qualification requirements have to be respected during the on line material production. Examples for necessary procedures during supervision of the mass produced material are provided as well as specific long term demonstrations.

INTRODUCTION AND CURRENT SITUATION IN SWITZERLAND

In the year 2008 the new ENSI guideline G05 [1] has come into force. This guideline regulates design, production and operation of interim stored casks including the documentation and tests during fabrication. Besides to several mechanical and thermal properties which must be fulfilled by the cask design consideration of aging effects is required. Fulfilment of all requirements of the ENSI guideline G05 must be maintained over the whole storage period. The applicant must find a way to demonstrate the development of the material behaviour during several decades under thermal loads.

Today Switzerland operates two dry interim storage facilities with some casks already stored. Table 1 shows the actually stored designs and quantities. In addition to these casks, two completely new designs were introduced for ENSI approval.

Figure 1: Interim storage site in Switzerland

Design/Typ	Quantity
TN24G/GB	4+4
TN24BH/BHL	6+8
TN97L	9
TN52L	1
TN81CH	6
HAW20/28CG	5
Castor I-c-Diorit	1
Total	44

Table 1: Quantity of stored casks in Switzerland

All these casks are of different designs including a variety of different materials. One major difference is the basket design (Figure 2) and material. The basket has to fulfil different safety functions like heat dissipation, structural strength, control of subcriticality and shielding. All these safety functions are influenced by design and material.

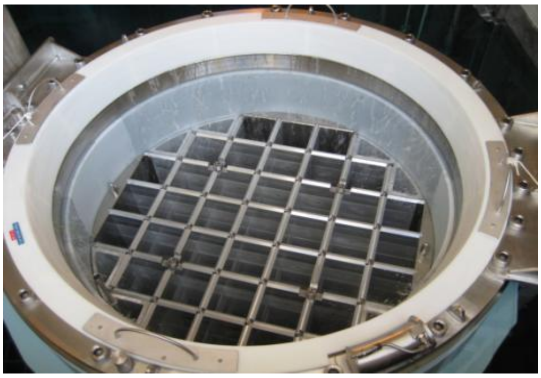

Figure 2: Basket inside a cask

A qualification process for the different basket materials, which are designed and fabricated specially for this use, has to be established. There is no standard code which describes the long term behaviour under high temperature for any of these new materials. The development of the qualification is also needed for the envisaged new licensing process for transport and interim storage casks in Switzerland. ENSI would like to implement the dual purpose cask (DPC) licensing process in a few years and one major part of such licenses is the control of aging effects. Therefore new basket materials must be extensively qualified. Currently ENSI is involved in four different material qualification procedures. Additionally to the qualification process the supervision and monitoring of the routine production has to be developed according to the ENSI-approved specifications. The material tests and monitoring during fabrication have to ensure that the routinely produced material fulfils the properties of the qualified material as specified in the respective Safety Analysis Reports (SAR).

ENSI has implemented a product based system for surveillance of nuclear components in general, which is as well applied to transport and storage casks for Switzerland. Prior to the manufacturing every new assembler will be audited and his quality management system will be approved. In addition to the audit, major steps and tests during fabrication will be monitored by a third party organisation. Normally ENSI designates SVTI to do all supervision action during fabrication of a cask. Therefore important tests like leak tightness test, pressure test and load test will be performed in presence of SVTI inspectors. Important fabrication steps like welding actions will also be checked. The supervision of each cask also includes the approval of selected manufacturing documents as e.g. quality plans, welding procedures, purchase specification of the cask designer and test procedures before fabrication, the review of all dysfunction treatment sheets and also the check of the complete manufacturing documentation in the end. The complete volume of supervision is defined by ENSI for each cask taking into account the ENSI G05 [1] guideline. Independent from SVTI supervision, the future cask owner also has to define his scope of supervision to ensure the quality of the cask supplier and the product.

After fabrication the requirements for operation of interim storage facilities are described in authority regulatory guide. The guideline ENSI-G04 [2] came into force 2010 and defines the requirements for interim storage facilities in Switzerland including responsibilities, requirements to the facilities, inspections and also the systematic safety evaluation. This safety evaluation is based on the national law and applies to the whole storage period of each waste type. For the cask systems in Switzerland two main issues must be respected.

(1) Transportability of each cask at each time
(2) Maintenance of the requirements for each cask defined by guideline ENSI-G05

QUALIFICATION PROCESS

According to ENSI-guidelines a qualification process includes several steps. This process is independent from the kind of material and the use. It is a general approach to the qualification.

(1) By definition of intended use of the cask the respective requirements are clear. For Type B packages the requirements of the service and accident conditions defined by ADR [3] must be fulfilled as well as for storage the requirements of the national guidelines.
(2) The applicant develops a cask design by use of standardized and/or special materials and their properties.
(3) The safety analysis reports both for transport and for storage provide evidence that the package design fulfils all safety functions and requirements.
(4) Identification of the entire safety relevant parameters and properties for each item and its material.
(5) These safety relevant parameters and properties must be ensured by the manufacturing documents and the suggested test methods from the applicant.
(6) During examination of the cask design the applicant must show that these parameters and properties can be maintained over the whole lifetime of the cask.

(7) The product based monitoring of the fabrication ensures that these parameters and properties can be maintained during fabrication process.
(8) To ensure these parameters and properties during whole lifetime actions must be implemented such as real time aging tests and updated periodic safety analysis.

In step (1) the national guidelines and the ADR defines the service and accident conditions and each dual purpose cask design has to fulfil these requirements. In Switzerland for example the transport requirements are such as 9 m drop test or fire come from the regulation of dangerous good onto street and rail (ADR [3]) and the storage requirements are such as aircraft crash with following kerosene fire and earthquake event come from the ENSI-G05 [1] guideline. In step (2) the applicant develops a cask design fulfilling the above mentioned requirements. One bases for the designs is the material properties. For some needs mainly for the neutron absorbing special materials must be developed by the cask designer. After defining all material properties the applicant makes all necessary analyses to show that all safety functions and requirements can be fulfilled by the cask design (step (3)). These analyses also must described the safety margins in respective to the material properties. If the cask designer can show that all requirements are covered, the material parameters are defined.

To identify all safety relevant parameters and properties a lot of different analyses for the safety targets must be checked by the assessor (step (4)). These analyses address different material parameters such as strength and strain for the mechanical properties, emission coefficient and heat conductivity for the thermal behaviour, density for the radiation protection and boron content to secure subcriticality. Sometimes the required material parameters from the storage SAR are different compared to the transport SAR. For dual purpose casks it is always the most restrictive parameters which have to be fulfilled. In addition to these material properties delivered from the safety reports, other facts are also important. It is necessary to define the maximum storage duration of the cask under constant maximal thermal conditions. Otherwise a temperature-time-history over the whole storage duration under consideration of the cooling of the fuel elements must be included in the safety analysis report for storage. Another point is how the cask is used and stored is described normally in both reports. Therefore often both safety reports must be compared. For example normally the aging effects are considered in the storage SAR but if the cask must be transported during storage it could be lead to problems if the mechanical strength criteria in the transport SAR are higher than in the storage SAR.

This theoretical description of design qualification and verification shall now be explained using the basket material, an issue currently receiving high regulatory attention.
In the beginning of the qualification process of the basket material for a specific cask the design and the safety relevant properties of the material must be identified. For example there are three different designs (shown in Figure 3-5) and each design addresses different properties to the special basket material. For design 1 (Figure 3) the complete basket is manufactured out of one material. Therefore all safety functions must be fulfilled by this special material. In design 3 (Figure 5), a sandwich structure, maintaining sub criticality is provided by a special material. The other safety functions like heat dissipation, structural strength and radiation protection are provided by a standardized structural material. Quite similar in design 2 (Figure 4), which has a skeleton out of standardized material, which provides mechanical strength special the absorber material can be less rigid than in design 1.

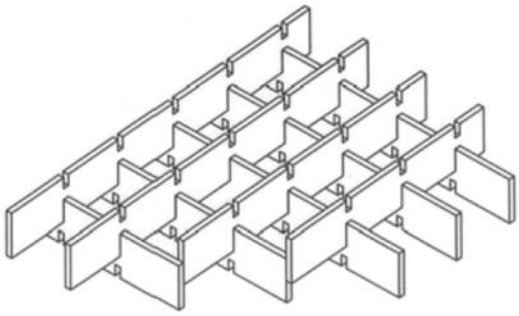

Figure 3: Basket made out of one material

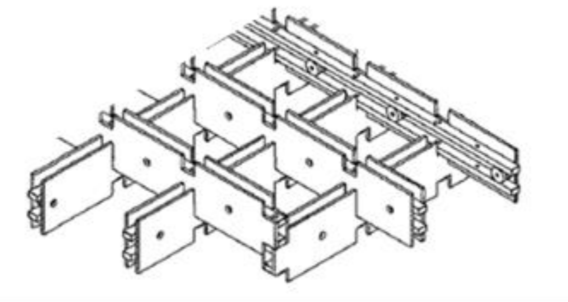

Figure 4: Basket with skeleton (AREVA TN Concept)

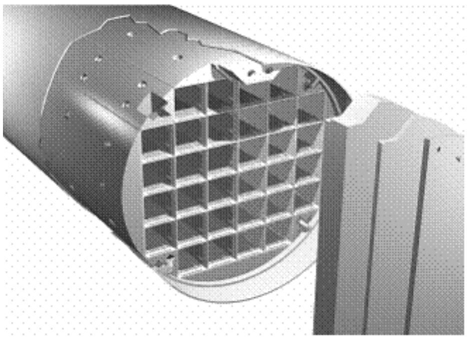

Figure 5: Basket with sandwich structure

In addition to these safety relevant parameters and properties which were defined by the correlation of design to the safety functions further aspects can be important too. For example if the cask is filled under wet conditions the applicant has to show the resistance against corrosion of the used basket material. Also if the cask is stored in horizontal position the creep effect coming from the weight of the fuel assemblies must be considered in the context of qualification. Additional to these points another basic requirement is the homogeneity of the material. If the material is for example a metal matrix composite (MMC) this means homogeneity of boron dissipation in the matrix as well as the homogeneity of the base material alloy relating to grain size, mechanical properties etc.
The last aspect to be considered in the qualification is the production process of the material and of the complete basket. If there are special steps in the material production process, like for example heat treatments or additives, they also have to be covered by the qualification. This applies equally to special processes during assembling of the basket welding processes or higher elongation to ensure local deformation.

RECOMMENDATIONS FOR QUALIFICATION

Based on experience from production control of more than 40 casks of various designs ENSI have recommendations in several topics.
The first topic is the physical parameters of the basket material. Normally the thermal properties such as heat conductivity, heat transfer coefficient and emission coefficient are not affected by the storage time and the temperature. If the material is specially coated and not coated using a standard procedure like anodization, the coating has to be specifically qualified.
The second topic is the neutron physical properties. They must be stable over the whole storage duration. To show the resistance against neutron and radiation inside the cask, the applicant can perform a calculation of the total accumulated dose over the lifetime. For

verification coupons irradiated with the same accumulated doses are to be tested. Isotopic tests must be performed to show the behaviour during and after storage duration.
For all mechanical properties like strength and elongation the applicant must show that all criteria are met over the whole storage duration under service temperature and accumulated neutron dose. Also the development of safety margins until the end of the storage period must be addressed in the safety analysis report. Of specific interest are materials showing degeneration of mechanical properties in real aging tests which are representing the conditions in a loaded cask. Here the applicant has to demonstrate sufficient safety margins for the chosen material on the basis of the temperature-time-history of the cask. Material should be preferred that does not show any degeneration. If the material shows a stable strength plateau (for example yield) after a long time under high temperature, this value should be taken for the justification in the safety report. If such a material has a higher strength at the beginning than at the final plateau, the material should be processed directly after fabrication to make use of the higher safety margin at the beginning of the storage period as this is the period with the highest heat load (freshly discharged fuel elements). It must however be ensured that the final plateau still provides enough safety margin. If the qualification demonstrates major deviations in the results of this stable plateau (strength or time) either the material cannot be accepted or the safety demonstrations must be modified. To take credit from the temperature-history-curve of each loaded cask can only be accepted in single exceptional cases. Such a procedure is not acceptable as a basis for the safety analysis report.
A major challenge is to simulate the behaviour of the mechanical properties during storage. It is not possible to make a real test over for example 40 years and 200°C beforehand. Therefore another approach is used: the Larson-Miller approximation [4]. The idea of this approximation is to perform the ageing test with higher temperature than the service temperature inside the cask but for a shorter time. This is described by the Larson-Miller-Equation factor P with a constant C, the time (t) logarithmic and the temperature (T) as multiplier:

$$P = T \cdot (C + \log t)$$

C is only depending on the material. For aluminium normally 20 is used for C.
Originally this approach was delivered for aluminium alloys. The validity for special boron containing composite materials like MMC still has to be verified. This also applies to the value of C.
If it is unknown whether the material can change its parameters and behaviour under higher temperature than the normal service temperature inside the cask a validation of this temperature depending behaviour must be done by the applicant.

RECOMMENDATIONS FOR FABRICATION

Material used for qualification tests has to be manufactured under the same conditions like the material used for the real manufacturing of the baskets. The qualification process itself has been approved by the authorities. Therefore the routine production must include all the quality-relevant manufacturing steps as for the material produced for the qualification. Especially the heat treatments, mixture procedures or extrusion temperature and speed must be the same for all lots of the material (mass produced and qualification).
The same is true for the same chemical properties of qualified material. This applies especially to the boron content (also B-10 content) and the base material composition. To show that the mass production material is comparable to the qualification, test coupons must be recovered from the fabrication of each material lot. Recovery of such samples has to

address specific geometric values of the profiles like thickness and special characteristic of the manufacturing process such as extrusion direction.
Due to the fact, that the heat treatment has a significant influence on the mechanical behaviour of the basket material the final heat treatment temperature during fabrication must be higher than the service temperature inside the stored cask.

FABRICATION MONITORING

Concerning the basket material, the volume of the supervision depends on the design of the basket and also on the used material. For example if the material is only used to ensure sub criticality the mechanical behaviour has minor importance in contrast to the thickness of the profiles. This topic is also linked to the beginning of the qualification program where the significant parameters are determined in accordance with the safety analysis report.
For the basket material and the basket manufacturing, the process parameters, the chemical composition of the base material and the boron content can be checked in advance on the basis of the process documentation. Thus their compliance with the qualification can be confirmed. During the manufacturing, chemical analysis has to be carried out to ensure the correct composition of the produced lots. Also the homogeneity of the mixed material shall be confirmed in an appropriate manner. It is recommended to do these steps at an early stage of the manufacturing. Before the final assembly of the basket starts, representative coupons shall be taken and tested for several requirements. One of them is the neutron attenuation behaviour, which is influenced by the B10 quantity. To ensure adequate neutron attenuation according to the qualification, radiation tests and isotopic analysis must be done.
If material sensitive for degradation with time is used it is necessary to find a way to simulate the ageing of the material for getting information on the long-term behaviour. This could be done with the before qualified Larson-Miller-Approximation and thus by using higher temperature than the loaded cask has under service temperature. With this approach it has to be ensured that there will be no increase of the mechanical strength by the simulated ageing heat treatment. After the ageing, tensile tests must be carried at those temperatures for which mechanical values are required in the safety analysis reports. The extent of the tests depends on the production process and the dispersion of test results within one lot. Generally in the first production run a higher number of tests should be done. If these tests show a small dispersion it is possible to reduce the quantity of tests for the next series step by step.

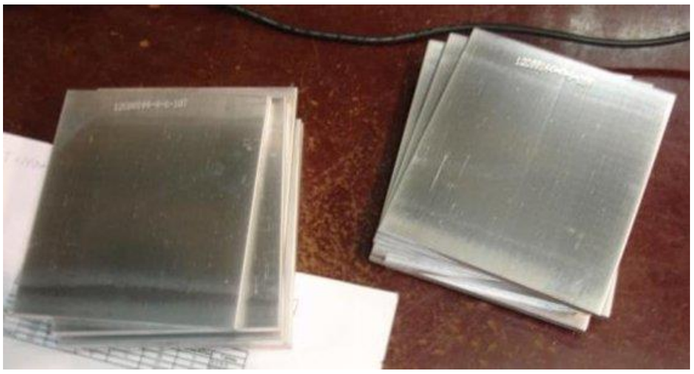

Figure 6: Marked test specimens, taken during fabrication

Besides the chemical and mechanical properties all other relevant characteristics shall be controlled as for example dimensions, flatness and straightness of the profiles or surface condition (Figure 7).

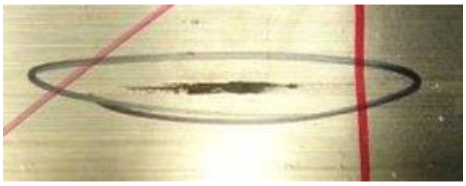

Figure 7: Surface scratch, detected during visual inspection

MONITORING DURING STORAGE

In respect to the guideline ENSI-G04 [2] all safety relevant properties of the basket material must be ensured over the storage period i.e. during “operation” of the cask. To have a better understanding of the different influences during storage and to have the knowledge about the material behaviour and the safety margins ENSI requests the cask owner to do real aging tests in parallel to the storage of each cask. Test specimens of the used basket material will be used to simulate the real aging inside the loaded cask. These specimens will be put into an oven with the same temperature of the loaded cask. This will start before the loaded cask will be stored. These specimens will be examined regularly and the material behaviour and properties will be checked for example in performing tensile tests. This is also a part of the systematic safety evaluation every 10 years enforced by the guideline ENSI-G04.

CONCLUSIONS

To fulfil the requirements defined by international standards and by the responsible safety authorities different basket types have been designed for specific applications. Depending on these designs the neutron absorber materials of the basket have to fulfil different safety functions. The materials used are specially developed for this field of nuclear industry. There are no general documents like ASME material specifications which specify the long term properties of such materials. The applicant has to provide additional evidence that the specified properties of such material can be ensured over the complete interim storage period.

For such qualifications interdependencies have been recognized and some suggestions derived. If material sensitive for degradation is used combination of aging effects and the development of service temperature inside the cask requires special attention. Another challenge is achieving the required material properties during routine production. Therefore specific methods to control alternative aging mechanisms are used. Such methods must be qualified to be applicable for the special materials.

Due to this complex topic it is recommended to reduce the application of different materials as possible. This also applies to variation in boron contents of the same base material. The cask owner should be more involved in the qualification process or at least he shall be capable to ensure in depth understanding of the safety relevant features of his own spent fuel casks.

To ensure the properties during the whole interim storage period different methods were used by the applicants in the qualification process. To validate the aging tests used for the qualification real aging tests shall be performed in real time on specimens from routine production.

A successful material qualification must in addition demonstrate the safety margins over the whole interim storage period thereby leading to an in-depth understanding of the time-dependant development of safety relevant material properties. This knowledge can be used for ensuring compliance with transport requirements during the final transport of the stored spent fuel to its final destination – the repository – after the interim storage.

ACKNOWLEDGEMENTS

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Figure 1: ZWILAG
Figure 2: AXPO Power AG
Figure 3: TNI
Figure 4: AREVA TN Concept
Figure 5: TN Inc.

REFERENCES

[1] ENSI
Richtlinie G05: Transport- und Lagerbehälter für die Zwischenlagerung
April 2008

[2] ENSI
Richtlinie G04: Auslegung und Betrieb von Lagern für radioaktive Abfälle und abgebrannte Brennelemente
September 2010

[3] ADR
Europäisches Übereinkommen vom 30. September 1957 über die internationale Beförderung gefährlicher Güter auf der Straße
Stand vom 1. Januar 2013

[4] J. Miller F.R. Larson
A Time-Temperature Relationship for Rupture and Creep Stresses, Transactions of the ASME, Vol. 74, ASME New York, July 1952, pp 771-785