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Neutron Absorber Qualification and Acceptance Testing from the Designer's Perspective

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Abstract

Starting in the mid 1990's, the USNRC began to require less than 100% credit for the ^{10}B present in fixed neutron absorbers spent fuel transport packages. The current practice in the US is to use only 75% of the specified ^{10}B in criticality safety calculations unless extensive acceptance testing demonstrates both the presence of the ^{10}B and uniformity of its distribution. In practice, the NRC has accepted no more than 90% credit for ^{10}B in recent years, while other national competent authorities continue to accept 100%.

More recently, with the introduction of new neutron absorber materials, particularly aluminum / boron carbide metal matrix composites, the NRC has also expressed expectations for qualification testing, based in large part on Transnuclear's successful application to use a new composite material in the TN-68 storage / transport cask.

The difficulty is that adding more boron than is really necessary to a metal has some negative effects on the material, reducing the ductility and the thermal conductivity, and increasing the cost. Excessive testing requirements can have the undesired effect of keeping superior materials out of spent fuel package designs, without a corresponding justification based on public safety.

In European countries and especially in France, 100% credit has been accepted up to now with materials controls specified in the Safety Analysis Report (SAR):

- Manufacturing process approved by qualification testing
- Materials manufacturing controlled under a Quality Assurance system.
- During fabrication, acceptance testing directly on products or on representative samples.
- Acceptance criteria taking into account a statistical uncertainty corresponding to 3σ.

The original and current bases for the reduced ^{10}B credit, the design requirements for neutron absorber materials, and the experience of Transnuclear and Cogema Logistics with neutron absorber testing are examined. Guidelines for qualification and acceptance testing and process controls, providing the basis for up to 100% ^{10}B credit, while satisfying all essential design requirements for transport package safety, are proposed.

1. Introduction

Baskets for spent fuel storage and transport containers usually include a neutron absorbing material between the fuel compartments as a means of assuring criticality safety. The most commonly used materials are boron–stainless steel alloy, boron-aluminum alloy, and boron carbide-aluminum composites. While other neutron absorbers continue to be investigated, the isotope ^{10}B has proven to be the most efficient for the configuration of a spent fuel basket and intact fuel. With the introduction of new borated materials into this arena in recent years, and with the reliance on improved neutron absorbers to provide criticality safety for increasing fuel enrichment, the testing used to qualify new materials (qualification testing) and the testing used to accept the material delivered for use (acceptance testing) have been the subject of increased attention by both regulators and designers.

2. Situation in the United States

In the 1980's and 90's in the US, there was not a great deal of attention to the means used for initially demonstrating the suitability of neutron absorber materials for long term use, or of acceptance testing of materials delivered for use. Designers took full credit for the specified mg ^{10}B/cm^2 (areal density). This changed in the mid-1990's in the US. Deterioration of Boraflex, a polymer-based neutron absorber, in wet storage led to awareness of the need to verify that materials could perform their design function in their design environment for their design lifetime [1]. At the same time the US Nuclear Regulatory Commission began to require that designers use less than the specified ^{10}B areal density in their criticality safety calculations, typically 75 to 90%.

The interest of both cask designers and the US NRC in these questions has accelerated with the introduction of a new class of materials for this use, boron carbide / aluminum metal matrix composites. The first US licensing of a transport packaging with this material was Transnuclear's TN-68 cask, which received a license for storage in 2000 and for transport in 2001 [2]. Many new transport packagings and license applications include this material, and several suppliers are manufacturing the material using various production techniques. Thus, there is a strong need for a standardized approach to qualify and accept these materials, an approach that provides the necessary and sufficient controls for public safety, while at the same time avoiding excessive requirements. This latter is not only for economic reasons, but because the response to such requirements is often to increase the boron content of the materials. This may have effects such as reduction of ductility and thermal conductivity that adversely affect design disciplines other than criticality safety. An effort by the ASTM International is underway to create such a standardized approach [3].

2.1 Reduced Credit for ^{10}B: Microscopic Uniformity

The USNRC guidance on this subject states "no more than 75% of the specified minimum neutron poison concentration of the packaging should generally be considered in the criticality evaluation" [4] and "a percentage of neutron absorber material greater than 75% may be considered in the analysis only if comprehensive tests, capable of verifying the presence *and the uniformity* of the neutron absorber, are implemented" [5, emphasis added].

Uniformity here was initially understood to be at the microscopic level. That is, uniformity is the relative freedom from the effects of heterogeneity, such as neutrons streaming between particles containing ^{10}B. This was based on experiments that found that heterogeneous neutron absorbers might be less effective for attenuation of collimated neutron beams than would be predicted by mathematical models such as those used in criticality calculations, which assume homogeneous ^{10}B distribution [5].

The most direct way to measure *effective* ^{10}B areal density is to use neutron transmission examination. By comparing the attenuation of thermal neutrons through the test material with the attenuation through a homogeneous or nearly homogeneous boron-containing material, one measures not the physical amount of ^{10}B, but rather one measures directly the ^{10}B that is effective in absorbing a collimated beam of thermal neutrons. If ^{10}B areal density is verified by measurement of the physical content of boron, e.g., by chemical and isotopic analysis, the chemical analysis can be benchmarked against neutron transmission testing, and a correction factor could be applied to the chemical analysis as necessary to account for any heterogeneity effects.

In recent Safety Evaluation Reports [6], the USNRC has further articulated the position that because of uncertainty in the criticality safety analysis methods, the designer must apply an additional 10% safety factor. According to this, if the designer can demonstrate that the material is 100% effective, that is, it behaves as if the ^{10}B dispersion were homogeneous, he should use 90% of the specified ^{10}B in criticality safety calculations; if the material is 83% effective, use 75% in the calculations, etc. This foundation for this position is not clear; uncertainty is considered by benchmarking the criticality calculations. Safety margin is provided by establishing a limit of 0.95 on the neutron multiplication factor, and (until now) by the assumption that the fuel is unirradiated.

2.2 Macroscopic Uniformity of ^{10}B

A second focus on boron uniformity is the macroscopic. That is, when we accept a neutron absorber by testing a small area on a coupon, to what extent is that coupon representative of the much larger area of sheet being delivered?

Transnuclear has accepted materials based on three methods of demonstrating macroscopic uniformity. One has been neutron radioscopy of coupons, which is a qualitative examination with acceptance based on uniformity of the radioscopic image. Another has been to perform acceptance testing on a coupon from the thinnest area of the master blank, after demonstrating that this is reliably the location with ^{10}B areal density less than or equal to that anywhere on pieces cut from that blank; that is, to use non-random sampling. The other approach has been the statistical analysis of areal density measurements from randomly sampled coupons contiguous to the delivered pieces. This analysis demonstrates that if the areal density of a large number of samples randomly located throughout the delivered material were measured, X percent of the results would be above the minimum specified areal density with Y percent confidence. The values of X and Y are usually both 95. This last has become the primary method used by Transnuclear in the US.

When this method was first implemented, entire lots were accepted or rejected based upon whether the lower tolerance limit at 95% confidence and 95% probability for the set of areal density measurements for the lot was equal to or greater than the specified minimum areal density. The method was improved by recognizing that areal density is the product of two independent characteristics, the ^{10}B volume density and the thickness.

In the current approach, test coupons are removed adjacent to the final pieces, and well-distributed throughout the lot. Test coupons that exhibit physical defects not acceptable in the finished product, or that would preclude an accurate measurement of the coupon's physical thickness, are not sampled for neutron transmission testing. The lot definition must create a set of material with consistent production history as well as with a sample size large enough provide a meaningful statistical analysis of results.

Neutron transmission measurements of ^{10}B areal density are reduced by three standard deviations, based on the number of neutrons counted, in order to conservatively account for statistical variations in measurement. The resulting value is called the "minimum ^{10}B areal density." The minimum ^{10}B areal density is converted to volume density, i.e., the minimum ^{10}B areal density is divided by the thickness at the location of the neutron transmission measurement. The lower tolerance limit of ^{10}B volume density is then determined, defined as the mean value of ^{10}B volume density for the sample, less K times the standard deviation, where K is the one-sided tolerance limit factor for a normal distribution with 95% probability and 95% confidence [7].

Finally, the minimum specified value of ^{10}B areal density is divided by the lower tolerance limit of ^{10}B volume density to arrive at the minimum thickness that guarantees the specified ^{10}B areal density. Any piece that is thinner than this minimum is treated as non-conforming.

2.3 Qualification Testing of New Materials

While acceptance testing is done to determine if material manufactured for use meets the acceptance criteria, qualification testing is performed, generally prior to first production, to determine if a material has the general characteristics required to perform its design functions in its design environment for its design lifetime.

The use of Boralyn® in the TN-68 introduced boron carbide/aluminum metal matrix composites into the storage and transport cask arena in the US. In order to demonstrate that it was not really another Boraflex, Transnuclear exposed the material to fast neutron irradiation, high temperature, corrosion testing, etc. Acceptance criteria such as "no formation of reaction products between the aluminum and boron carbide" that were not necessarily related to product performance were established and verified by transmission election microscopy (TEM). These tests became the basis for NRC expectations [8, 9].

The results confirmed what would be expected of metallic / ceramic systems. Metals barely begin to experience measurable changes in mechanical properties due to fast neutron fluences at about 10^{17} neutrons/cm^2 [10, 11], while the fluence in the basket of a spent fuel cask after 50 years at constant flux would be less than 10^{15}. Nor is there any reaction between aluminum and boron carbide below 350 °C [12], which is well above basket temperature under normal conditions of storage or transport. Furthermore, the temperatures of use in casks will not come near the temperatures at which the material was originally processed during ingot formation, extrusion, and hot rolling, so there is no reason to expect a temperature-induced physical change in the material that would be significant for the non-structural applications to which these materials are currently limited in the US. The conditions of dry storage and transport include immersion for only short duration in high purity deionized water or boric acid, so general corrosion, pitting corrosion, etc., do not have sufficient time to adversely affect the integrity of the material. For the purpose of hydrogen generation and compatibility with other basket materials, aluminum-based material may be regarded as identical to its aluminum alloy matrix, because the boron-containing particles are inert.

In essence, for metal-ceramic systems, the thermal, radiation, and corrosion environment of spent fuel dry storage and transport is not challenging. The focus of qualification testing should be to verify that the production process results in a material that has the necessary design properties (mechanical strength and ductility, thermal conductivity, etc.), and that has microscopic and macroscopic uniformity of ^{10}B distribution appropriate to the proposed means of acceptance testing for ^{10}B areal density.

3. Situation in Europe

In European countries, and especially in France, 100% credit for ^{10}B has been accepted for metals or metal matrix composites, whatever may be the fabrication process. This has not been based upon prior review of the material by the regulator, but rather upon obligations specified in the Safety Analysis Report and in other documents:

- Manufacturing process approved by qualification testing
- Acceptance testing directly on product or on representative coupons for production material
- Criteria taking into account a statistical error corresponding to 3σ for areal density measurement
- Manufacturing controlled by a Quality Assurance system

The main metals considered are:

- Borated stainless steel plates (ASTM-A887)
- Cast aluminum alloy (AS10B3 or other alloy)
- Extruded plates and profiles in aluminum alloy (6351) containing TiB_2.
- Metal matrix composites, formed by casting, powder metal processes, thermal spray, etc.

Final product form is achieved by casting, rolling, or extrusion.

Materials qualification testing and acceptance testing may include demonstration of mechanical properties and thermal conductivity, as required by the design, but the following discussion will focus on the demonstration of ^{10}B areal density.

3.1 Qualification of the Manufacturing Process

The principle of qualification for ^{10}B content is to demonstrate that the proposed measuring locations are representative of the pieces delivered, and to validate the acceptance testing measurements which guarantee a minimum ^{10}B areal density of the pieces delivered for use.

The average value of the ^{10}B areal density is determined on sample pieces representative of the lot. Average areal density must be greater than the specified value, which is generally larger than the value in the SAR. The sampling can vary with the form of the material.

3.2 Neutron Transmission Measurements

Two kinds of neutron sources are used to perform neutron transmission measurements of ^{10}B areal density:

- A portable neutron source whose neutron source strength decreases as an exponential function of time, but can be treated as constant over short periods, for example, one day.

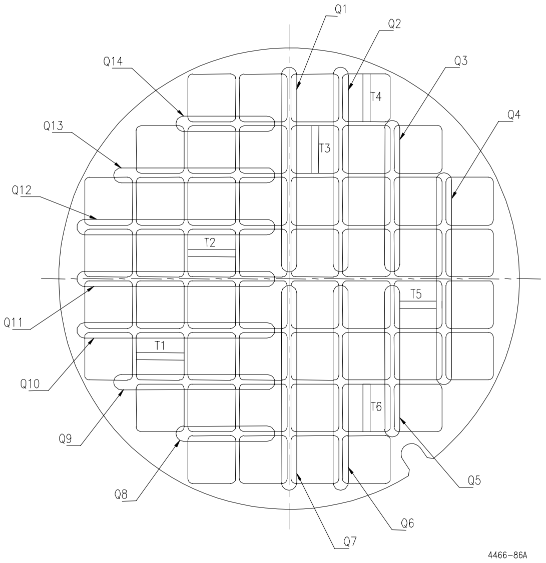
- A thermalized, collimated neutron beam from a research reactor: the neutron flux is greater and allows measuring higher content in ^{10}B. On the other hand, a reactor flux is not constant, and it is necessary to have neutron detector not only after attenuation by the test piece, but also for the incident neutron beam, in order to normalize the counting.

The measurement of ^{10}B areal density is made by comparison of the neutron count after the beam passes through the test piece to the count after the same number of neutrons passes through calibration standards of known uniform ^{10}B areal density. The test piece can be the component itself or a sample cut adjacent to the component. If the areal density is too high to be measured in a reasonable period of time, samples with reduced thickness can be prepared by machining. The standards should be material with a homogeneous distribution of boron, without other significant neutron absorbers, and with neutron scattering characteristics similar to the material being tested. Materials that have been used include titanium diboride or zirconium diboride paired with aluminum shims, boron carbide sheet, and composites with a very fine (≤ 25 micron) boron carbide in aluminum. There should be standards with both greater and less areal density than the test piece to form a calibration curve of transmitted neutron counts as a function of areal density. The number of counts must be great enough to obtain the desired degree of accuracy, that is, to reduce the standard deviation to the desired level; the time to achieve this number of counts increases with increasing ^{10}B areal density.

3.3 Materials Qualification Testing Examples

For cast pieces, the qualification consists of casting a complete piece with the same fabrication procedure as the production pieces, and cutting the qualification piece into coupons, as shown in Figure 1. The ^{10}B areal density of these coupons, taken in the more critical parts of the piece, is measured by neutron attenuation testing. Results are evaluated to determine if there are any differences attributable to location within the piece.

Figure 1: Sampling of Cast Part



Q1 to Q14 = coupons taken on qualification piece

T1 to T6 = coupons taken on fabrication pieces

For plates fabricated by rolling, the sample piece for qualification is a finished plate produced by the same manufacturing process to be used in production. A grid is drawn on the plate, and ^{10}B areal density measurements are performed in each area of the grid.

Qualification of extruded plates or profiles is made by measurements on samples representative of a complete heat. For example if a heat is cast into four logs, each log is cut into three billets, and ten profiles are extruded from each billet, then a sample will be taken from each log, then from each of the three billets from one log, from each of the ten profiles from one billet, and finally one profile will be cut into coupons for examination.

As an independent control on the neutron transmission measurements, a chemical analysis of the boron content, an isotopic analysis of the boron, and measurement of the density are made on a sample taken in the middle of the heat.

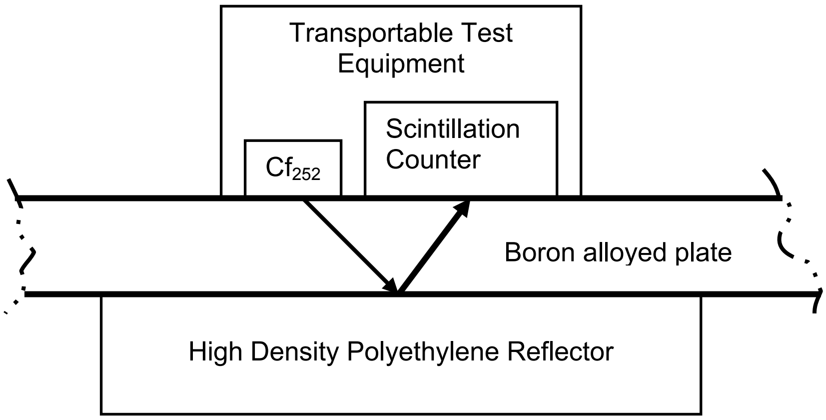
3.4 Examples of Testing Material During Manufacturing (Acceptance Testing)

Acceptance testing consists of a limited number of ^{10}B areal density measurements on the products or on representative coupons, which varies with the product form.

For cast pieces, coupons are taken from extensions on the casting, for it is generally not possible to have a direct measurement on the piece. These extensions are uniformly distributed in the piece, to be representative of boron content in all parts of the piece, as shown in Figure 1. To obtain a statistically significant number of results, and to verify ^{10}B areal density from top to bottom of the casting, more than one measurement may be taken on each coupon.

In the case of rolled pieces with low ^{10}B areal density, a direct measurement may be performed on the plate with a special apparatus including a neutron generator, detector, and reflector, as shown in Figure 2. This apparatus may be used only when the sensitivity of the measurements is demonstrated to be sufficient. It is placed on one or more points on each plate to be measured, and the neutrons are counted during a predetermined time; accuracy increases with increasing time. The ^{10}B areal density is determined from a diagram obtained by measurement of standards of verified ^{10}B areal density, and with same thickness as the plates being measured.

Figure 2: Portable Neutron Transmission Measurement Apparatus [13]



The measurement of extruded plates or profiles is made by ^{10}B measurement on samples taken at each end of a profile or plate, usually using a neutron beam from a research reactor.

To provide complementary measurements, one chemical analysis is made per plate (rolled products), or per heat (cast or extruded products). In case of boron enriched in isotope ^{10}B, an isotopic analysis is made in addition to the chemical analysis.

Confirmatory analysis is provided at the end of production, before assembly of the baskets. The sample with the lowest boron content, as determined from the control described in the previous paragraph, is sent to an independent laboratory for measurement of ^{10}B areal density by chemical and isotopic analysis.

In the case that any of these measurements yield a result less than the specified areal density, the entire lot is treated as non-conforming.

3.5 Criteria Taking into Account a Statistical Uncertainty of 3σ

The minimum areal density at any location on the piece *i* is given by:

$$T_i = X_0 (E_i / E_0)$$

where

- X_0 is the areal density measurement on the piece or on its associated coupon; to account for measuring uncertainty, the value (N + dN) is used to determine the areal density from the calibration curve, N being the neutron count and dN being 3 times its standard deviation,
- E_i is the minimum thickness on the piece, and
- E_0 is the thickness at the location of the neutron transmission measurement.

3.6 Manufacturing According to a QA System

The supplier of neutron absorber material must implement a quality system in compliance with ISO standard 9001 (2000). Particularly, non-conformities must be controlled by the supplier’s quality assurance system. Subcontractors, for example extruders, rolling mills, or testing agencies, must be audited by qualified personnel. The Supplier shall ensure that materials, documents, measuring equipment, inspection reports, etc., are completely traceable to the delivered product.

4. Proposed Qualification and Acceptance Testing Program with International Applicability

Incorporating the experience of both Transnuclear and Cogema Logistics, an appropriate neutron absorber material qualification program outline would

a) determine product density and compare to theoretical density to determine if the product is porous,

b) perform tensile testing for full density metal matrix composites (MMCs) and alloys (acceptance criteria as appropriate for the design, elongation as necessary to assure non-brittle material, etc.),

c) test for water absorption, hydrogen generation, and steam blister formation (delamination) for products with some porosity,

d) demonstrate that the manufacturing process produces a reasonably uniform distribution of boron by performing a statistically significant number of tests for ^{10}B areal density, at randomly sampled locations on the product, and showing that the standard deviation is less than some acceptable percentage of mean (probably 5 to 10% based on experience),

e) if the product may be susceptible to ^{10}B heterogeneity effects such as neutron streaming (very thin product, large boron-containing particles, low boron content), and if acceptance testing will be by chemical analysis, compare the results of chemical analysis to the results of neutron transmission analysis to determine if a correction factor for effective ^{10}B needs to be applied,

f) demonstrate the validity of acceptance testing techniques; for example, if acceptance testing will be non-random, demonstrate the validity of any assumptions, such as that the thinnest location will reliably yield the lowest areal density, and

g) verify other properties as required, e.g., thermal conductivity, suitability for anodizing, etc.

Process controls would be established by mutual agreement between designer and supplier so that the delivered product would be consistent with the qualification test material. Changes in the agreed-upon process controls that could affect mechanical properties would require re-qualification by mechanical testing, and changes that could affect the boron distribution would require repetition of the neutronic qualification testing. Quality assurance controls over the manufacturing process would provide documentation that manufacturing has conformed to the agreed-upon process controls.

Acceptance testing would remain the true determinant that the material delivered for use satisfies the design requirements. An appropriate acceptance testing program outline could include:

a) Testing by neutron transmission compared to homogeneous standards of known ^{10}B areal density or equivalent heterogeneous standards with a very fine, uniform dispersion of the borated phase. This provides a direct measurement of effective ^{10}B areal density; thickness and ^{10}B density should be treated as separate variables if practical.

b) Acceptance testing by chemical, spectrometric, and dimensional measurement. This provides a measure of the physical ^{10}B areal density, which may be more than the effective areal density if there are any neutron streaming or self-shielding effects due to heterogeneity of the neutron absorber. Therefore, acceptance testing by the chemical method should be benchmarked against neutron transmission measurements at the time of qualification testing. If the neutron transmission results are less than the chemical analysis results, the ratio should be applied as a correction factor to the chemical analysis for acceptance testing to account for the heterogeneity effects.

c) Statistical analysis, which may be applied to the results of ^{10}B density acceptance testing for each lot of material delivered for use. This analysis demonstrates that if the ^{10}B density of a large number of samples randomly located throughout the delivered material were measured, X percent of the results would be above the minimum specified areal density with Y percent confidence. The values of X and Y are usually both 95. Methods which achieve equivalent results, such as statistical process controls, rejection of all parts from a single master blank that fails the areal density measurement, neutron radioscopy, etc., may be acceptable.

d) Other acceptance criteria, such as mechanical properties, thermal conductivity, surface and dimensional inspection, etc, may be verified using standard industrial techniques.

5. Conclusion

The main elements of delivering quality neutron absorbers are qualification testing, process controls, quality assurance, and acceptance testing.

There is no need to perform extensive environmental testing to qualify metallic / ceramic systems; the dry spent fuel storage and transport environment is not challenging to such materials. Rather than examining characteristics that may affect performance, such as metallurgical structure, one should test performance directly. Mechanical testing is an appropriate means to demonstrate that such materials are sufficiently strong and ductile to perform their design functions in their design environment for their design lifetime.

An outline for qualification testing, process controls, and acceptance testing has been proposed. This outline addresses the primary concerns of both designer and regulator for providing up to 100% credit for the specified ^{10}B areal density: structural integrity, quantity of ^{10}B, and uniformity of ^{10}B distribution throughout the product at both the microscopic and macroscopic scales. Additional margin of safety (reduced ^{10}B credit) may be considered as appropriate depending on the design or the acceptance testing process, or as required by the national regulatory authority.

The proposed plan satisfies the goal of providing sufficient and necessary assurance that design characteristics are achieved. At the same time, it avoids introducing unnecessary testing requirements that could discourage the introduction of superior products or production techniques.

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