Neutron Absorbers: Qualification and Acceptance Tests

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Abstract

Materials containing the element boron, as well as other materials with a high cross-section for absorbing thermal neutrons, are used widely in the nuclear industry. These materials are most commonly produced in the shapes of plates and rods. One common use of these absorber materials is to ensure subcriticality during normal and off-normal/accident service conditions inside storage casks and transportation packages of spent nuclear fuel. Frequently, little or no structural credit is given to these materials. The boron isotope, ^{10}B, with its high cross section for thermal neutrons, is one component that can be used to prevents criticality. Commercial products are tailored to meet a particular material specification of minimum areal density of the ^{10}B isotope. When an absorber material is introduced into the marketplace, acceptance tests and qualification tests must be performed to ensure compliance with regulatory requirements. The materials must be demonstrated to contain prescribed levels of the ^{10}B isotope, that are homogeneously distributed throughout the product. In addition, the products must be sufficiently durable to withstand the conditions of service. For each product and manufacturing process, qualification tests are conducted at least once. These tests are used to demonstrate that the material will be of adequate quality and durability for the intended service. In addition to qualification tests, acceptance tests are used to ensure that material properties for plates and other shapes are in compliance with the licensing requirements of the application. Usually this is a demonstration for a production run that the distribution and level of ^{10}B is adequate for all plates that will be used in service. Statistical information on selected parameters can be used to ensure that the product will meet the service requirements.

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Introduction

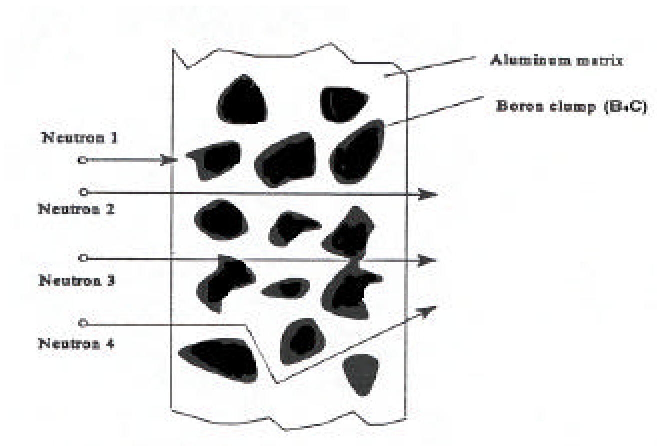
Various boron-containing materials are used widely in the nuclear industry as neutron absorbers. Since these materials are components of containers used to store and transport radioactive elements with long half lives, they are required to have adequate resistance to radiation and corrosion, and adequate physical and chemical properties. Further, these materials should experience negligible reduction in effectiveness under normal/off-normal and accident conditions of storage or transport. Neutron absorbers can consist of alloys of boron compounds with aluminum or steel in the form of sheets, plates, rods, liners, and pellets. Likewise, neutron absorbers are commercially produced as composites of fine particles in a matrix. One example is composite material with a matrix of an aluminum alloy containing a dispersion of particles of boron carbide (B_4C). The composite can be clad on both sides with an aluminum alloy to further enhance environmental durability. Another example would be that of stainless steel with boron as a precipitate. Materials containing boron as a natural precipitate are to benefit from the laws of nature, which promote a uniform chemical potential of species (such as boron) in the solid state. Man-made composite materials (such as those produced as neutron absorbers) require blending and processing technologies to promote homogeneity. This paper discusses some of the tests that are used to qualify a material and to assess homogeneity and durability in the approval of neutron absorber materials for use in licensed Dry Cask Storage Systems (DCSS) and transport containers in the United States.

Background

Neutron absorber materials are used to ensure subcriticality during normal/off-normal and accident conditions in containers for storing or transporting fissile materials. Typically, they are expected to absorb neutrons only during the very unlikely accident scenarios involving unborated water entering the primary containment barrier of a spent fuel containment system, e.g. a DCSS or transport container.

Fine particles that are uniformly distributed are required for good absorptivity. The distribution of ^{10}B in a commercial neutron absorber material may not always be ideal, and streaming or channeling of neutrons, as depicted in two dimensions in Figure 1[1], may occur due to non-evenly distributed ^{10}B. Results of thermal neutron attenuation/absorption tests have shown that three factors influence channeling effects in absorber materials. These factors are enrichment, particle size and areal density. Neutron streaming has been observed[2] in Boral? , which is a commercial material containing boron. The streaming is attributed to particle size effects of the B_4C, which is dispersed in an aluminum matrix in Boral? [3]. Particle sizes for Boral? are considered coarse because 95 percent of the particles range between 80 and 180 ?m. For heterogeneous samples of Boral? , the measured neutron absorption was found to be about 18 percent less than that for a homogeneous material containing the same amount of ^{10}B. This finding led to a recommendation, in several NRC NUREGs, to limit the credit of absorber material effectiveness to only 75 percent.[4,5]

Based upon recommendations in the applicable standard review plans, NUREGs 1609, 1617, and 1537, it has been the NRC's practice to either (a) limit the credit for absorber materials to only 75 percent of the minimum amount of neutron poison shown to be present, or (b) consider giving credit up to 90 percent if comprehensive measures are implemented to establish the presence, uniformity, and neutronic effectiveness of an absorber material. The 75-percent limit has served primarily to bound the observed effects of neutron channeling in heterogeneous absorber materials such as Boral? . When this is done, the material is regarded as being 83 percent effective. The 90-percent limit, as applied to absorber materials that are regarded to be 100 percent effective (i.e. finely dispersed and homogeneous) such as boron carbide pellets, has been used to offset specific uncertainties in the validation of the criticality computational methods.



Discussion

Current NRC practice is to use qualification and acceptance tests to ensure the acceptability of neutron absorber materials for DCSS and transport containers Qualification tests are conducted on material produced by a given manufacturing process and material specification to demonstrate the quality, effectiveness and durability of the material over its licensed service life. Once qualified, only acceptance tests need to be conducted on plates to be used in service. This is done to ensure that the material produced in a given production run meet the absorptivity requirements. A material may need to be re-qualified if significant changes in production processes or material specifications are made.

Neutron absorber plates for use in DCSS and transport containers are fabricated to provide a specified areal density of ^{10}B, which is the mass per unit volume of the isotope in the material multiplied by the thickness of the material in which that isotope is contained. Acceptance tests and qualification tests are used to ensure that the finished absorber material contains no less than a prescribed areal density of the absorber isotope.
The neutron absorber material must be demonstrated to be adequately durable for the service conditions of the application. This is usually done using qualification test data for the material. In addition, acceptance tests are performed on samples from each production run, so as to ensure that the absorptivity values for the plates or other shapes produced are in compliance with the specifications and requirements of the application. The uniformity of the distribution of ^{10}B may be addressed in both the qualification and the acceptance tests.

Qualification Testing

Qualification tests are conducted at least once, for a given set of materials and manufacturing processes, to demonstrate acceptability and durability of the resulting product as a neutron absorber over the licensed service life. Table 1, Qualification Tests for Neutron Absorbers, describes the qualification tests that are one acceptable means as preliminary requirements for qualifying the candidate materials for containment systems used in the storage or transport of commercial spent nuclear fuel.

Table 1. Qualification Tests for Neutron Absorbers

Materials Environmental Factors	**Testing/Analysis**
Short-Term Exposure during Loading and Unloading Operations	The absorber material shall be evaluated for environmental interactions, such as thermal conditions and the generation of hydrogen in the environment of the spent fuel pool. Applicable periods of time should exceed those expected for loading, unloading and transfer operations. For these evaluations, the absorber materials shall be evaluated for dissimilar metal interactions, as may be appropriate to the application, and the environment may be borated or deionized water, as appropriate.
Neutron Absorption	The absorptivity shall be measured on plates taken from lots of (production) materials or materials intended for commercial use. The number and choice of location shall be determined so as to have adequate statistical assurance that the absorptivity is above the minimum required. The goal is to obtain assurances that the production methods yield products that are sufficiently homogeneous within and between lots.
Thermal	The absorber material shall be analyzed or examined for alterations and damage that could occur from exposure to the entire range of temperatures for the basket under normal conditions and hypothetical accident conditions over the licensed life of the system, under helium (or other gas as appropriate) and with appropriate imposed stresses. The test sample(s) shall be metallurgically examined for the following changes: 1. Redistribution of the boron. 2. Dimensional changes, due to material instability. 3. Cracking, spalling, or debonding of the matrix from the boron or boron-containing particles. 4. Weight reduction, due to outgassing. 5. Embrittlement. 6. Chemical changes, e.g. oxidation or hydriding.
Radiation	The absorber material shall be examined for damage which could occur from exposure to the maximum fast and epithermal neutron integrated flux over the licensed lifetime of the system, under helium or other environment, and under appropriate levels of imposed stress. The test sample(s) shall be metallurgically examined for the following changes: 1. Redistribution of the boron. 2. Dimensional changes (material instability). 3. Cracking, spalling, or debonding of the matrix from the boron or boron-containing particles. 4. Weight reduction (outgassing).

Table 1. Qualification Tests for Neutron Absorbers	
	5. Embrittlement. 6. Chemical changes, e.g. oxidation or hydriding. 7. Effects due to radiolysis. Note. The long-term storage environment is dominated by fast neutrons; the material is exposed to thermal neutrons only during loading under water. Therefore, “damage evaluations” are required only for damage from exposure to fast neutrons. For an aluminum matrix containing B_4C, gamma exposure need not be evaluated because its effects on aluminum and B_4C are negligible when compared with the effects of fast neutrons, which are the primary source of radiation damage to metal and ceramics in this application.
Neutron Radiography	Macroscopic uniformity of ^{10}B can be qualitatively verified by radiography or radioscopy of the coupons[6]. A common acceptance criterion is uniform luminance across the coupon. Variations in luminance indicates variation in ^{10}B coverage. This inspection usually covers the entire coupon. In addition, for plates not fabricated from a wrought alloy, e.g. for plates fabricated from composite materials, randomly selected plate shall be scanned in its entirety to demonstrate uniform luminance. The significance of any apparent non-uniformity should be analyzed quantitatively. Radiography may be useful in arguments for 1) qualifying a material and its production methods, and 2) making an argument for a specific level of credit.

Acceptance Testing

Acceptance tests are used to ensure that material properties for plates and other shapes produced in a given production run are in compliance with the materials requirements of the application. Acceptance tests are used to ensure that the process is operating in a satisfactory manner. Statistical tests may be conducted, as appropriate, to augment findings relating to isotopic content, impurity content, or uniformity of the ^{10}B. Table 2, Acceptance Tests and Criteria for Neutron Absorbers, describes the types of acceptance tests commonly conducted on absorber materials.

Table 2 Acceptance Tests and Criteria for Neutron Absorbers

Test Procedure	**Property being Measured**	**Test Description and Criteria**
Chemical Analysis	Boron Content	An assay performed to determine whether the material has the specified boron content[7]. Analytical samples shall be randomly taken from coupons removed from the plates. When these measurements are used in lieu of neutron transmission the area of each sample should be comparable with the area of the “spot” size of the neutron transmission test.
Mass Spectrometric	Isotopic content	Determination of the isotopic content of the boron (^{10}B and ^{11}B)[7] is made to establish whether the content is in compliance with the purchaser’s specifications.
	Impurity	Impurity content is determined[7] to ensure that the maximum

Table 2 Acceptance Tests and Criteria for Neutron Absorbers		
Spectrochemical	content	concentration limit, for selected impurity elements, is not exceeded. For example, iron is particularly important in aluminum alloys, as it tends to migrate to and "ripen" in grain boundaries.
Neutron Transmission	^{10}B content	Effective ^{10}B content is measured locally by neutron transmission tests of coupons, to determine absorption values of the coupons in relation to values of appropriate standards, e.g. ZrB_2. This is done for various locations to establish within-plate and plate-to-plate variability as needed with the number of locations to be measured being dependent on the variability and the required accuracy. Assuming that all other requirements have been met, the material may be approved for greater than 75 percent credit (this is 83.3 percent credit for the material--see Discussion) only after sufficient data have been compiled on production or other materials, to ensure that the minimum required absorber level will always be present despite normal inhomogeneities. Statistical data on transmissivity may be coupled with luminescence test data to obtain reasonable assurance of uniformity of the neutron absorber material. The area of the "spot" sizes normally used in neutron transmission tests is about one square centimeter. When chemical analysis is used as a surrogate for these measurements, an area of comparable size shall be used.
Visual Examination	Defects	The finished plates shall be examined to verify that they are free of cracks, porosity, blisters, or foreign inclusions. Removal of such defects, where possible, shall be permitted if the removal does not result in a dimensional non-conformance.

Coupons taken for Qualification and Acceptance Testing

Test coupons are randomly selected for use in qualification and acceptance testing of the candidate materials. Coupons should be taken so as to be representative of the neutron absorber. To the extent practical, test locations on coupons will be randomized to minimize errors due to location, or position within the coupon. Some suggested locations should include the ends, corners, centers, and irregular locations. These locations represent the most likely areas that contain variances in thickness. Adequate numbers of samples should be taken from every other component (plate, rod, etc.) produced in a lot to obtain a good representation. A lot is defined as all plates from a single billet. Overall, the coupons should be taken as a random samples of the material.

Sampling of 100 percent of the coupons taken from each new billet is normal practice. Sampling 100 percent of all plate materials may be scaled back if warranted. For example, in the acceptance testing for neutron absorber plate, a reduced level of sampling (to 50 percent of all coupons) was allowed after acceptance of all coupons in the first 25 percent of the lot. A measured value less than the required minimum areal density during reduced inspection results in a rejection. Plates materials contiguous with the location of the rejected coupon are themselves rejected. If a coupon rejection occurs during normal or reduced inspection, then 100 percent inspection of coupons is again mandated.

Conclusion

Acceptance and qualification tests are used to establish the presence, uniformity, effectiveness, and durability of a neutron absorber, material (usually ^{10}B) so as to ensure that throughout the service period the material will continue to meet the applicable safety requirements. The tests and guidelines (or their equivalents) discussed in this paper are required for the approval of neutron absorber materials. Modified or additional tests may be warranted based on level of poison credit requested or type of absorber to be evaluated (e.g. gadolinium).

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