

VERIFICATION TESTS OF NEUTRON SHIELDING MATERIALS AND SHIELDING ASSESSMENT

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ABSTRACT

A combined test (neutron irradiation and heating) and a thermal test were performed for neutron shielding materials MREX[®] applied to MSF cask, which has been developed in Mitsubishi Heavy Industries, LTD (hereinafter called MHI), to confirm the extent of damage of neutron irradiation and fire. Simultaneously, shielding calculation was conducted considering the damage to verify shielding performance after the thermal test. These verification results proved that MREX[®] has sufficient shielding performance and its durability.

1. INTRODUCTION

Neutron shielding materials for spent fuel transport and storage cask are required to have characteristics to withstand heat decay and radiation released from spent fuel under normal conditions, and sufficient fire-resistance to minimize damage of cask under accident conditions. Therefore, various verification tests on neutron shielding materials for spent fuel transport and storage cask have been performed [1][2].

MHI have developed transport and storage cask for higher burn-up and shorter cooling time fuels (MSF cask fleet). MREX[®] developed by MHI is used as neutron shielding materials of this cask. Various verification tests for the materials have been conducted in the development of MSF cask. In these tests, damages of MREX[®] were confirmed, and shielding performance for the damaged one was verified. This paper describes various verification tests and characteristics of MREX[®].

2. SPECIFICATION OF MREX[®]

MREX[®] mainly consists of epoxy resin, which is mixture of bisphenol A type resin and alicyclic polyamine as hardener. To the epoxy resin, aluminum hydroxide to improve flame resistance and boron carbide to absorb secondary gamma ray are added. Table 1 shows specification of MREX[®], where main characteristics are high hydrogen content and superior shielding performance.

In addition, epoxy resin-based neutron shielding materials are difficult to mass-produce due to reaction heat caused by polymerization reaction. However, MREX[®] up to 600kg per batch can be produced by developing a mixing apparatus with improved cooling capability. As a result, the development allows shortening of manufacturing process of MREX[®] (See Figure 1).

Table 1. Specification of MREX[®]

| | |
|---|----------------------------------|
| Thermal Expansion Coefficient | 0.9×10^{-4} (30~150 °C) |
| Available Temperature | -40~150 °C |
| Density (g/cm ³) | 1.66 |
| Density of Boron Carbide (g/cm ³) | 0.020 |
| Density of Hydrogen (g/cm ³) | 0.10 |



Figure 1. Production of MREX[®]

3. NEUTRON IRRADIATION TEST

To confirm damage on MREX[®] due to neutron irradiation, a neutron irradiation test was conducted in the test reactor YAYOI, Nuclear Engineering Research Laboratory, University of Tokyo. Neutron fluence for this test is approximately 10^{15} n/cm², which is equal to an assumed fluence for 60 years in neutron shielding of MSF cask. Some samples of neutron shielding materials after irradiation were analyzed, while others are heated as a test combining irradiation and heat.

3.1 GAS ANALYSES

Qualitative analyses of sampling gas in exposure containers after neutron irradiation were conducted using a gas chromatograph mass spectrometer (GC-MS). In addition, components of the generated gas were analyzed.

The analysis results are shown in Table 2, which proved that components of organic gas were below the detection limits. Moisture contents were almost equal to water saturation, and a small amount of hydrogen was detected as a component of inorganic gas. The above results confirmed that deteriorative reaction by neutron irradiation such as decomposition of epoxy resin and filler, generation of gas, is rarely caused.

Table 2. Analysis Results of Gas Components after Irradiation

| Hydrogen (vol%) | Moisture content (mg/L) | Low-boiling organic element | Formic acid (mg/L) | Acetic acid (mg/L) | Propionic acid (mg/L) | Glycolic acid (mg/L) |
|-----------------|-------------------------|-----------------------------|--------------------|--------------------|-----------------------|----------------------|
| 1.76 | 26.8 | Air+Ar | <0.1 | <0.1 | <0.1 | <0.1 |

3.2 WEIGHT MEASUREMENT, DENSITY MEASUREMENT

There was no change in weight and density of the samples in the measurement before and after irradiation. These measurements showed that there was no decomposition or geometry variation of MREX[®] by neutron irradiation, like the analysis results of gas components.

3.3 COMBINED TEST (NEUTRON IRRADIATION AND HEATING TEST)

Neutron shielding is irradiated at higher temperature up to 150 °C in an actual cask. In this test, a heating test was carried out after neutron irradiation at room temperature because samples were impossible to be exposed to irradiation and heat at the same time in the test reactor. The heating test conditions are shown below.

- 1) Shape of samples : Cylinder (12mm in diameter, 65mm in height)
- 2) Heating temperature : 170 °C, 180 °C, 190 °C
- 3) Heating time : 1000h, 2000h, 5000h (170 °C)
100h, 200h, 500h (180 °C)
20h, 40h, 100h (190 °C)
- 4) Applied equipment : Heater, sealed tubes for heating
- 5) Heating atmosphere : Completely sealed system (Filling of N₂ in the early stages)

Each one of cylindrical sample was inserted into a stainless steel tube and sealed with plugs so that each sample could be heated in closed atmosphere (See Figure 2). After each heating period, stainless steel tube was pulled out from the heater and cooled down to room temperature. And each sample was weighted.

As described in section 3.2, the test results showed that weight of the samples with only irradiation was rarely reduced while weight of samples with only heating and with heating after irradiation was obviously reduced. There was little difference between only heating and heating after irradiation, which means that aging degradation of MREX[®] is greatly affected by heating (See Figure 3). Therefore, multiplier effect of irradiation and heating would be extremely low.

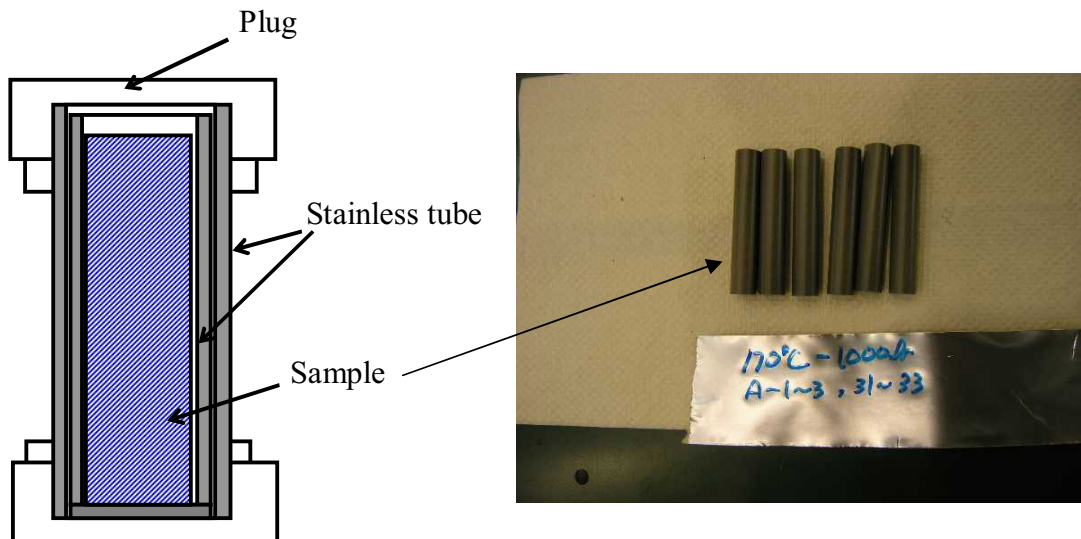


Figure 2. Sealed Tube for Heating Test and Samples after the Test

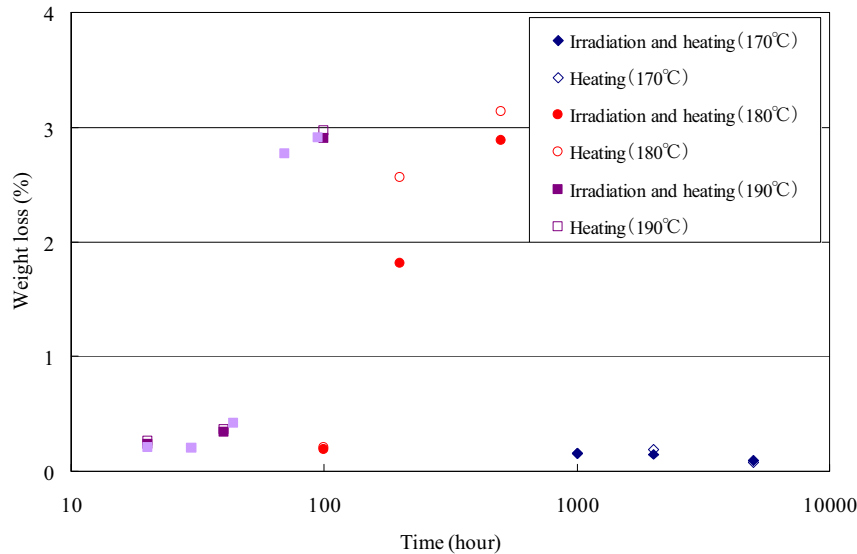


Figure 3. Combined Test Results and Influence of Neutron Irradiation

4. VERIFICATION OF FIRE RESISTANCE

4.1 THERMAL TEST

A thermal test was conducted under conditions based on TS-R-1 requirements (at 800 °C for 30 minutes) to identify extent of fire damage for MREX[®] under accident conditions (fire conditions)[3]. The test was conducted in the presence of BAM/T1.

a. Test model

Dimensions and state of the test model are shown in Figure 4. Body neutron shielding of MSF cask is a rectangular solid surrounded by thermal conductor, outer shell, and body shell. Therefore, the test model simulated the cross-section of the rectangular solid, which is 16cm high, 17cm wide, and 50cm long. In addition, a test model simulating penetration of a bar of 15cm in diameter into neutron shielding was prepared considering damage caused by 1m penetration test. Furthermore, several thermocouples were installed to observe internal temperature of the model.

b. Test procedure

- (1) Temperature of the test model was set to 130±10 °C before the thermal test.
- (2) Temperature in the furnace was set to 850 °C in advance and then maintained at the temperature for 1 hour or more.
- (3) The lid of the furnace was closed quickly after the test model was put into the furnace.
- (4) The test was started when the temperature in the furnace reached 800 °C again, and the test model was exposed to that environment for 30 minutes.
- (5) After 30 minutes, the test model was removed from the furnace and then cooled in the atmosphere.

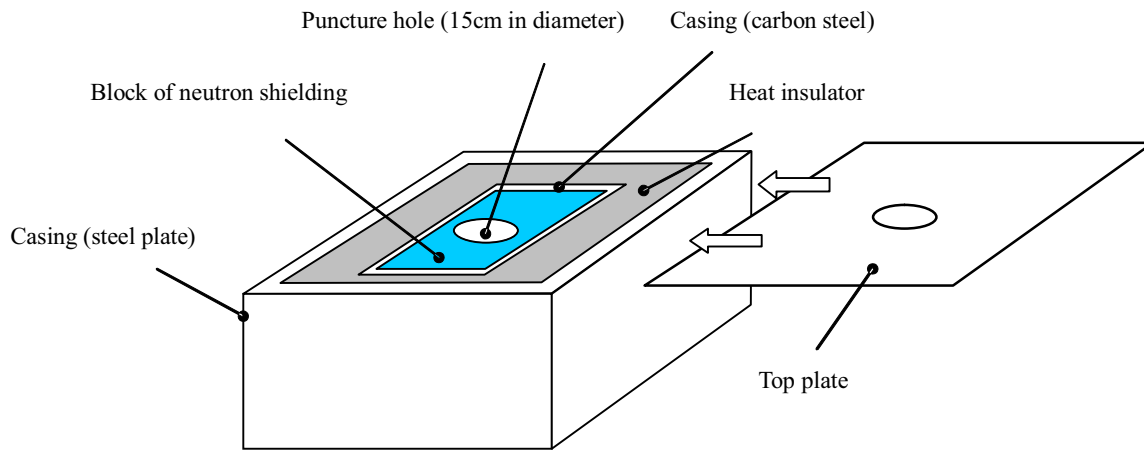


Figure 4. Test Model Simulating Neutron Shielding

4.2 TEST RESULTS

Weight test, cutting surface observation and chemical analysis for the test model were conducted after the test. The weight test results showed reduction of approx. 20%, and the cutting surface observation showed damage of approx. 10cm near the puncture hole, which was expanded to 25cm in diameter (See Figure 5). The chemical analysis results showed reduction of hydrogen content in neutron shielding by 10% compared with that before the test (See Figure 6).

These results proved that the total hydrogen content remained 70% compared with that in the early stages.

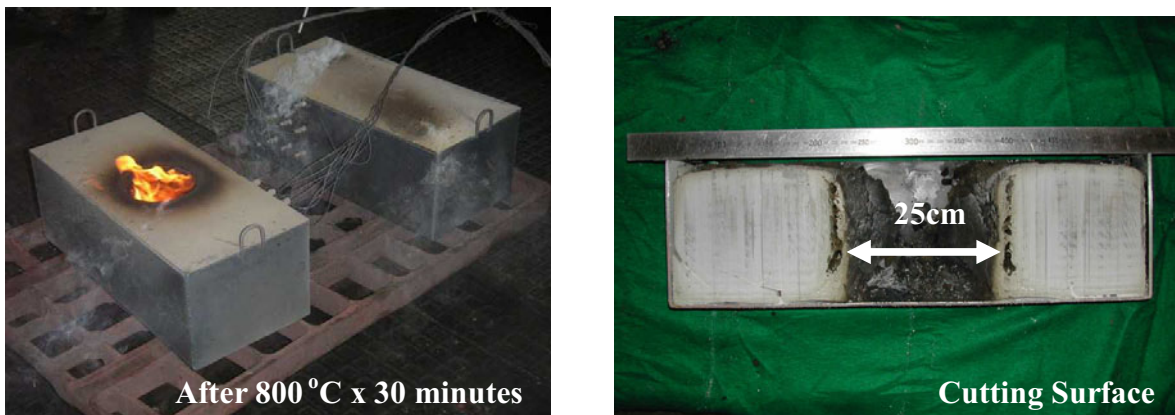


Figure 5. Test Model after Thermal Test

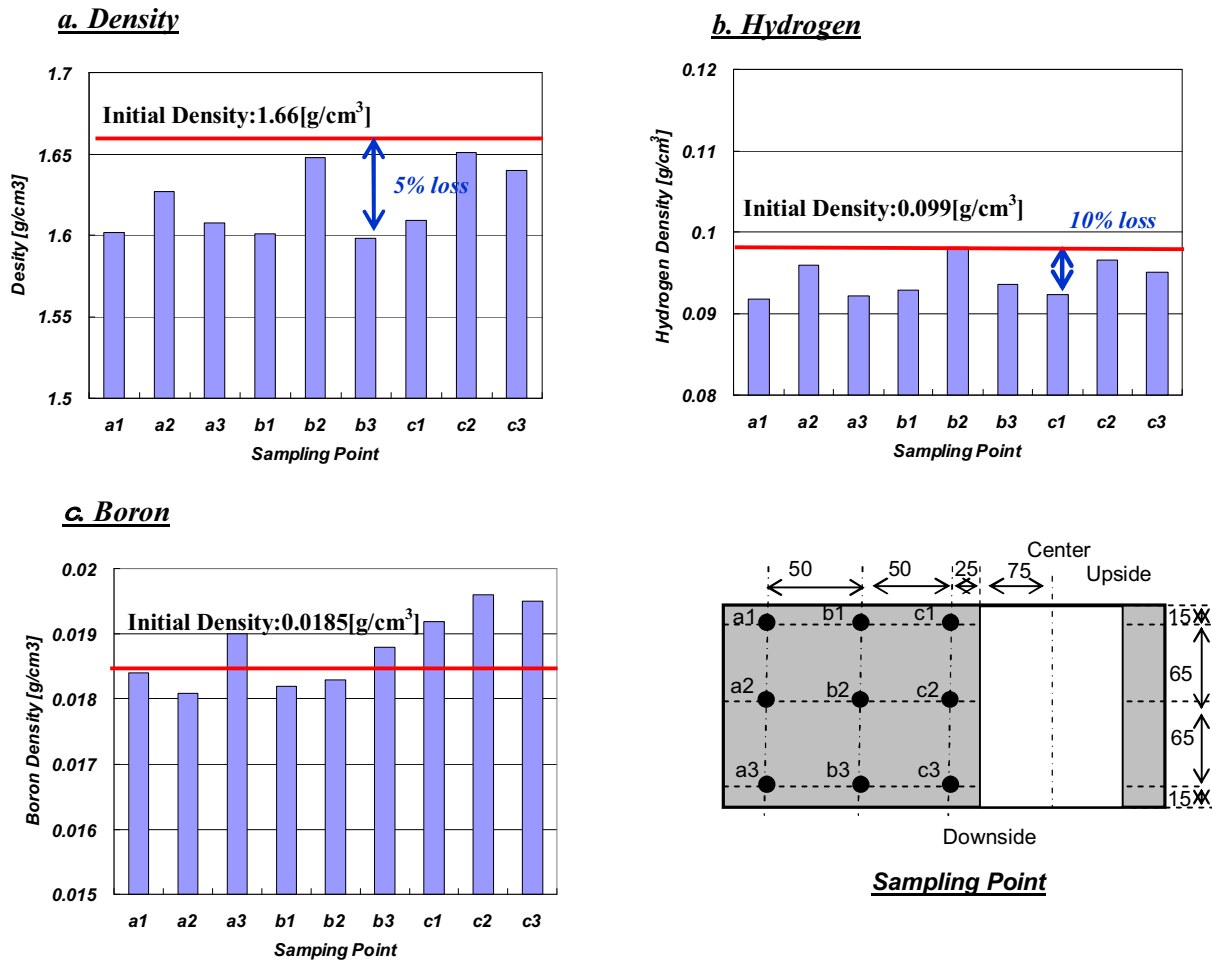


Figure 6. Chemical Analysis Results in Test Model

4.3 SHIELDING ASSESSMENT UNDER ACCIDENT CONDITIONS

Shielding calculation was conducted based on the verification test results described in section 4.2 to confirm shielding performance of damaged MREX[®] in the thermal test. Calculation conditions are as follows:

1) Modeling

- A two-dimensional model was used.
- Approx. 25cm thick cask body and approx. 18cm thick neutron shielding were applied to this model.
- Damage of neutron shielding was specified as weight loss of 50%.
- Two kinds of puncture hole diameter were modeled. One is 25cm, considering expansion caused by fire, and the other is 15 cm, not considering the expansion.

2) Source terms

- 69 BWR fuels
- Neutron source intensities were calculated by ORIGEN-2.

The calculation result showed in Figure 7, which shows dose rates of neutron in calculation cases of puncture holes 15cm and 25cm in diameter.

This result proved that the maximum dose rate at 1m from the cask surface near the puncture hole of 25cm was twice as much as that in the case of 15cm hole, which satisfies the criterion of 10mSv/hr.

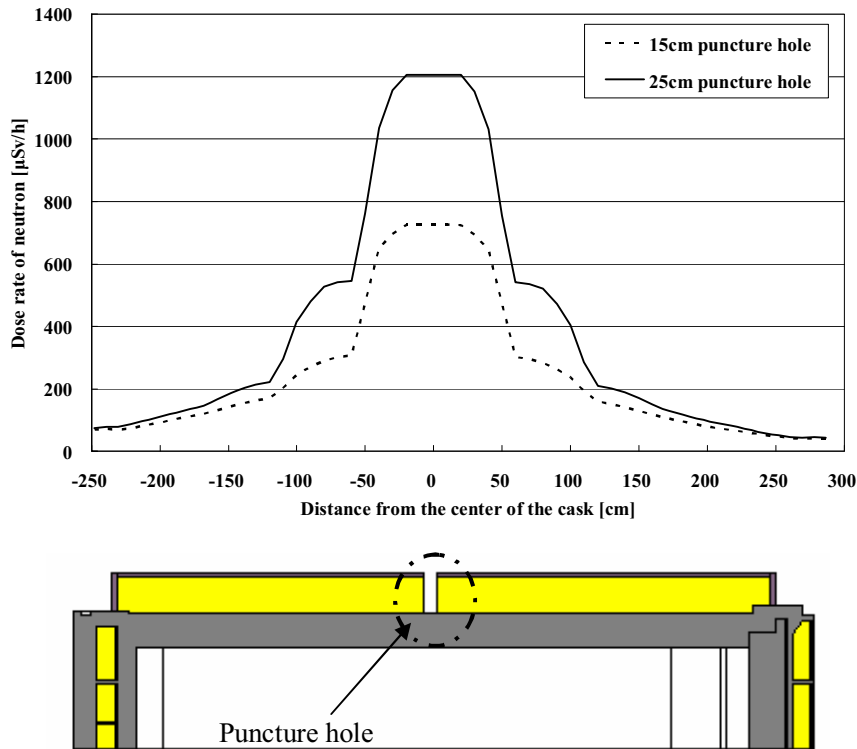


Figure 7. Shielding Calculation Results under Accident Conditions

5. CONCLUSIONS

- The neutron irradiation test proved that there was no degradation of MREX[®] by neutron irradiation such as decomposition and geometry variation. Moreover, aging degradation is unaffected by neutron irradiation.
- In the thermal test, little damage of MREX[®] were confirmed, which showed that the material has superior flame resistance.
- Supposing that a puncture hole is made in neutron shielding by penetration test before the thermal test, the extent of damage due to expansion of the puncture hole after the thermal test was confirmed. Shielding calculation showed that the maximum dose rate near the puncture hole after expansion of the hole, which increased twice compared with that before the expansion of the hole, completely satisfies the criterion.

ACKNOWLEDGMENTS

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REFERENCES

- [1] Momma.Y,et “Evaluation Test of the Thermal Stability of Resin as Neutron Shielding Material for Transport and Storage,” 12th PATRAM, Paris, France, May 10-15, 1998 .
- [2] Boonstra.R.H, “Thermal Testing of Solid Neutron Shielding Materials,” 10th PATRAM, Yokohama, Japan, September 13-18, 1992.
- [3] Regulations for the Safe Transport of Radioactive Materials, IAEA Safety Requirements No.TS-R-1, 2005 Edition.