EVALUATION TEST ON THE THERMAL STABILITY OF RESIN AS NEUTRON SHIELDING MATERIAL FOR SPENT FUEL TRANSPORT CASK

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SUMMARY

Epoxy-resin based neutron shielding material, NS-4-FR, is used for spent fuel transport and/or storage cask. In this paper the outline of thermal aging test performed to evaluate the heating effect on this neutron shielding material, NS-4-FR, is introduced. The test is consisted of two kinds of thermal aging test, one is "Basic Test" and the other is "Block Heating Test". The former is cooperatively performed by ten Japanese Electrical Power Companies, and the latter is done by GESC and NOF Corporation.

Outline of Basic Test

In order to obtain the basic data for evaluation of heating effect on the neutron shielding efficiency of NS-4-FR, weight of isothermally heated sample was measured and composition of gaseous compounds released from sample were analyzed. Each NS-4-FR sample was sealed in a stainless steel tube with end plugs and heated isothermally at 150～190℃ in thermostats.

Based on the results of this "Basic Test", it was confirmed that the heating under 170℃ in the closed system gives little effect on the shielding efficiency of NS-4-FR from the view-point of cask design.

Outline of Block Heating Test

Temperature of neutron shielding material in the actual cask is not uniform. To aid to evaluate the effect of temperature gradient on the shielding efficiency, thermal aging test using test blocks was performed.

Cylindrical test block covered by thermal insulation was set upright on the electrically heated hot

plates, and the bottom of the test block was kept at 170℃ in the air. Along the axis of the blocks, temperature distribution during heating was measured, the density and chemical element distribution were measured after the heating.

Based on the measured density distribution in the block, it was confirmed that it is sufficiently conservative to evaluate the weight change of NS-4-FR in the actual cask by using isothermal heating data.

BASIC TEST

Test Conditions

"Basic Test" was performed with a view to obtaining the data for evaluation of heating effect on the properties of NS-4-FR, especially on its shielding efficiency. In this test the external appearances of samples were observed, their weights and composition of gaseous compounds released from samples were measured after isothermal heating over 150℃.

Thermal aging conditions were determined in conformity to the international guide for the thermal evaluation and classification of electrical insulation (IEC Pub.216) and German specification on the thermal evaluation of plastics (DIN53 446).

Thermal aging conditions such as heating temperature, heating time, sample size, sample number, etc., are shown in Table 1-1.

Sample Preparation

Samples were prepared by GESC through the same process by using dedicated mixing machine for the actual installation so that they have just the same properties as NS-4-FR in actual cask. GESC is the company concerning in installation of NS-4-FR into the actual casks.

Test Procedure

Each one of cylindrical sample was inserted into a stainless steal tube (sample tube) and sealed with end plugs so that each sample could be heated in closed atmosphere. Sample for gas analysis was sealed in sample tube by end plugs with valve. Before sealing air contained in sample tube was replaced with nitrogen-oxygen mixed gas with appointed composition. Samples were heated in several thermostats at appointed temperature for appointed period shown in Table 1-1. Each temperature of these thermostats was controlled within ±0.5℃.

After achieving of each heating period, sample tube was pulled out from thermostats and cooled down to room temperature. External appearances of samples were observed and their photographs were taken. Each sample was weighed by direct reading balance. A constituent of gas collected from sample tube was analyzed by gas chromatography-mass spectrograph(GC-MS) and its composition was determined by gas chromatography(GC).

Test Results

External Appearance of Heated Samples

All the samples were observed before and after heating. While they showed a slight change in color, no deformations nor cracks which affect neutron shielding efficiency was observed in heated samples.

Weight Change of Heated Samples

All the samples showed some weight loss after heating. Weight change at each heating temperature is shown in Figure 1-1. It is noticeable that weight loss has a tendency to saturate in the early stage of heating period, especially earlier than 1000hr. The portion of weight loss after 1000hr was extremely small. Each weight loss at all the temperatures except at 190℃ was relatively small, and those after 5000hr at 150℃, 160℃, 170℃ and 180℃ were 1.4wt%, 2.3wt%, 2.9wt% and 3.9wt%, respectively. The weight loss at 190℃ exceeded 4wt% only after 200hr, thermal aging at this temperature was completed at 200hr.

The averaged rate of weight loss during each heating period ($\triangle W/\triangle t$) can be estimated based on this result. Figure 1-2 shows this estimated average rate of weight loss during each heating period. From this figure, it can be confirmed that weight loss rate decreases rapidly with heating period. For instance, the averaged rate of weight loss at 170℃ between 1000～2000hr(1) becomes smaller than 1/60 of that between 0～100hr(1), and 1/10 of that between 100～200hr(1).

Composition of Gas released from Heated Sample

The result of analysis on the composition of gaseous compounds released from sample is shown in Figure 1-3. From this result it is confirmed that over 90wt% of gaseous compounds is consisted of water (vapor). Therefore it could be considered that the weight loss is mainly caused by release of water from NS-4-FR and that the sauce of water is that absorbed in NS-4-FR and/or crystal water in aluminum hydroxide added in NS-4-FR as flame retarder.

BLOCK HEATING TEST

Test Method

In order to simulate the temperature distribution of the shielding material in the actual cask under its working condition, cylindrical test blocks of NS-4-FR were continuously heated at its bottom side over a hot plate at 170℃ for 300hr and 2,000hr in the air. Test blocks were covered with thermal insulation, and the temperature distribution in test blocks was measured during heating. Figure 2-1 shows the shape and size of the test blocks. After the heating, the test blocks were cut off vertically along its central axis to observe the appearance of cross section and to measure the density and chemical composition in several points.

Test Result

Appearance of Heated Test Blocks

In both test blocks heated for 300hr and 2000hr, slight change of the color was observed in the range of about 50mm above the bottom. However, neither deformations nor cracks was observed in the samples. The range where the color changed did not expand further regardless of heating time, showing its dependence only on temperature.

Note(1) : Averaged rates of weight loss between 0～100hr, 100～200hr and 1000～2000hr are plotted at the points of 100hr, 200hr and 2000hr, respectively

Weight Loss Due to Heating

Figure 2-2 shows the relation between the temperature distribution and weight loss estimated from measured density change in the test block heated for 2000hr. While some weight loss was observed it was comparatively small. The weight loss of NS-4-FR in the cask heated under the condition of temperature gradient can be estimated by using this result. Assuming the temperature of NS-4-FR in the cask is between 100℃ and 170℃, the average weight loss of this sample in the region of this temperature range can be estimated to be less than 1%. This value is considerably small, compared with the weight loss in the "Basic Test" for 2000hr heating at 170℃.

CONCLUSIONS

The thermal aging test on epoxy-resin based neutron shielding material, NS-4-FR, was performed. The following conclusions are led, and it was confirmed that NS-4-FR sufficiently keeps its neutron shielding efficiency under heating up to 170℃ in closed system from the view point of cask design ;

- No deformation nor crack which may affect the shielding efficiency was observed in the heated sample,
- Large portion of weight change (weight loss) at 150～180℃ occurred in early stage of heating, earlier than 1000hr, weight loss after this was very small,
- Weight loss under 180℃ in closed system was small. Those at 180℃ / 5000hr and 170℃ / 5000hr were < 4wt% and < 3wt%, respectively,
- The main component of released gas from sample was water (vapor),
- Weight loss due to heating under the condition of temperature gradient was much smaller than that isothermally heated sample. Therefore it is conservative to evaluate a thermal aging effect on the neutron shielding efficiency of NS-4-FR in actual cask based on the result of isothermal heating test.

REFERENCES

DIN53 446 : "Deutsche Normen : Bestimmung von Temperatur-Zeit-Grenzen", (1962)

IEC Pub.216-1,2 : "Guide for the Determination of Thermal Endurance Properties of Electrical Insulating Materials : Part 1 and Part 2"

Table 1-1 Thermal Aging Condition in Basic Test

Item	Test Condition in this test and suggested in DIN and/or IEC		
	This Test	DIN or IEC Pub.216[1]	Remarks
Number of temp. condition	5 conditions	≧ 3 conditions	satisfied
Temp. interval	10℃	10～25℃ 20℃ (≧ 10℃)*	satisfied
Heating temp.	150,160,170,180 and 190℃	lowest test temp. ≧temp. which gives longer life than 5000hr * highest test temp. ≧temp. which gives longer life than 100hr *	satisfied
Heating condition	continuous heating	continuous(or cyclic*) heating	satisfied
Heating period	100,200,500,1000, 2000, 3600 and 5000hr	1, 2, 4, 8, 16 and 32weeks	satisfied
Sample size	6mm ϕ x 60mm	120mm x 15mm x 10mm (same size for measurement of Brinell hardness, etc.)	conservative from the view-point of weight change evaluation because of larger S/V ratio
Sample number	5 samples for weight meas. 1 sample for gas analysis	5 samples not specified	satisfied
Atmosphere	closed condition	to consider actual working condition*	satisfied

(note 1)

*mark : based on IEC Pub.216-1,2

no marks : based on DIN53 446

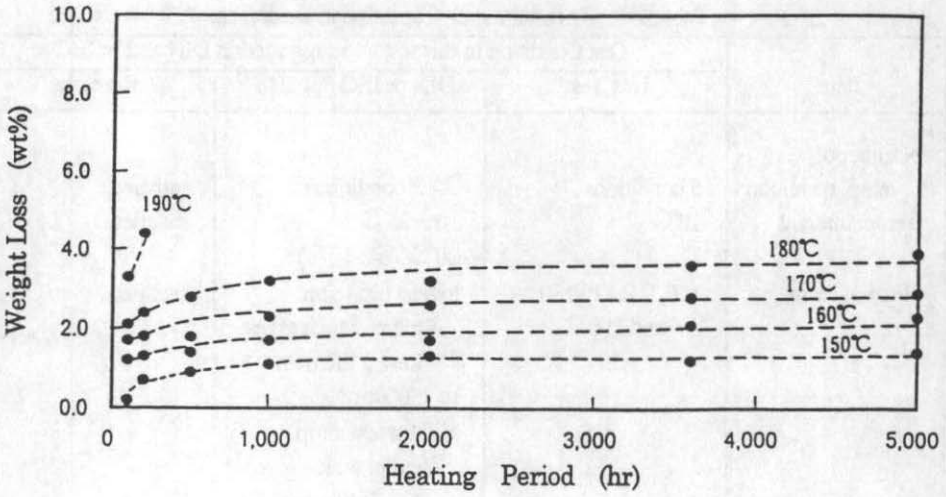


Figure 1-1 Weight Loss of NS-4-FR by Heating in Closed System

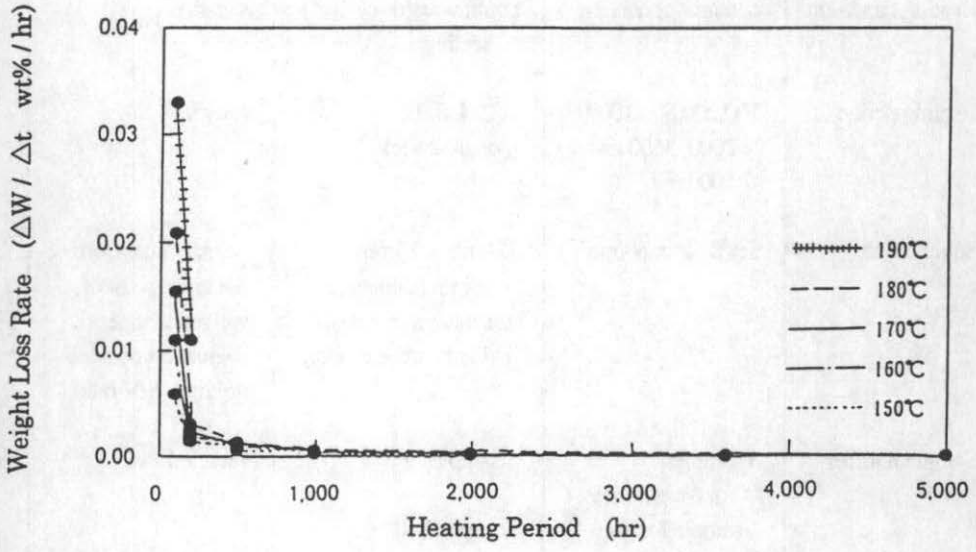


Figure 1-2 Change of Weight Loss Rate

Averaged weight loss rate between tn-1 and tn : $\triangle W / \triangle t = (\triangle Wn - \triangle Wn\text{-}1) / (tn - tn\text{-}1)$

here $\triangle W / \triangle t$ is plotted at the point of time tn

$\triangle W$: weight loss between tn-1 and tn

$\triangle t$: time interval between tn-1 and tn

$\triangle Wn$: weight loss at tn = $(W - Wn) / W$

$\triangle Wn\text{-}1$: weight loss at tn-1 = $(W - Wn\text{-}1) / W$

W : sample weight before heating

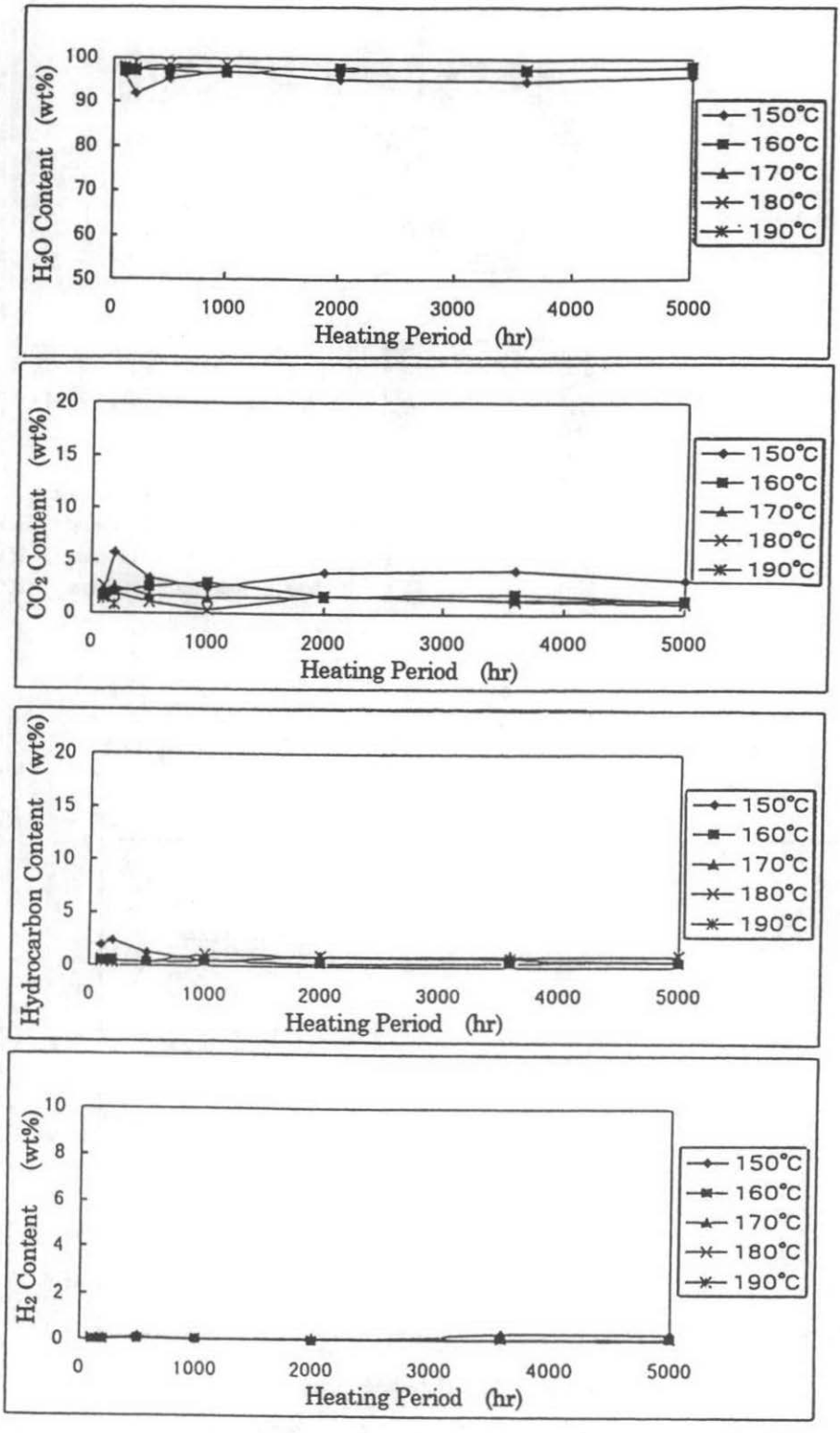
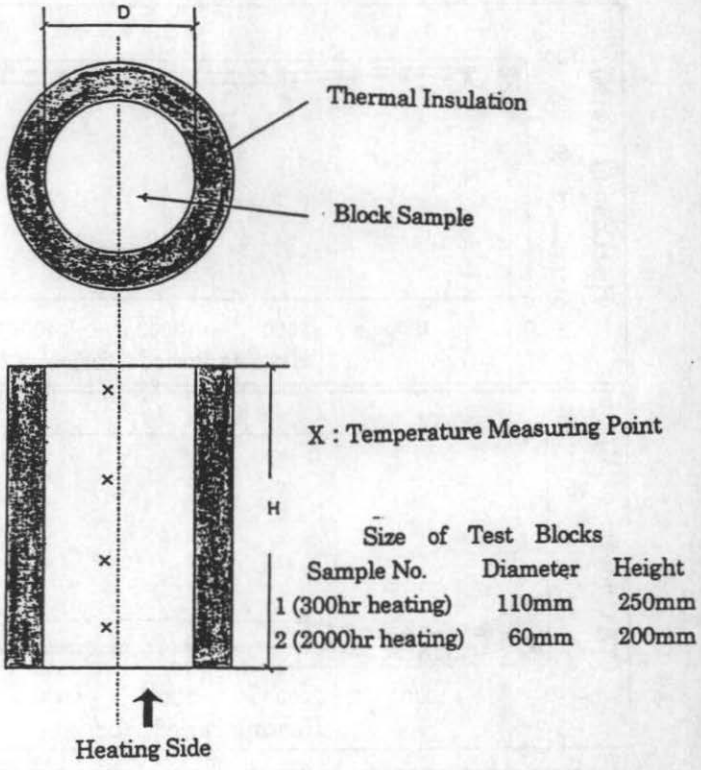


Figure 1-3 Content of Gaseous Compound released by Heating



Size of Test Blocks

Sample No.	Diameter	Height
1 (300hr heating)	110mm	250mm
2 (2000hr heating)	60mm	200mm

Figure 2-1 Shape and Size of Test Blocks for Block Heating Test

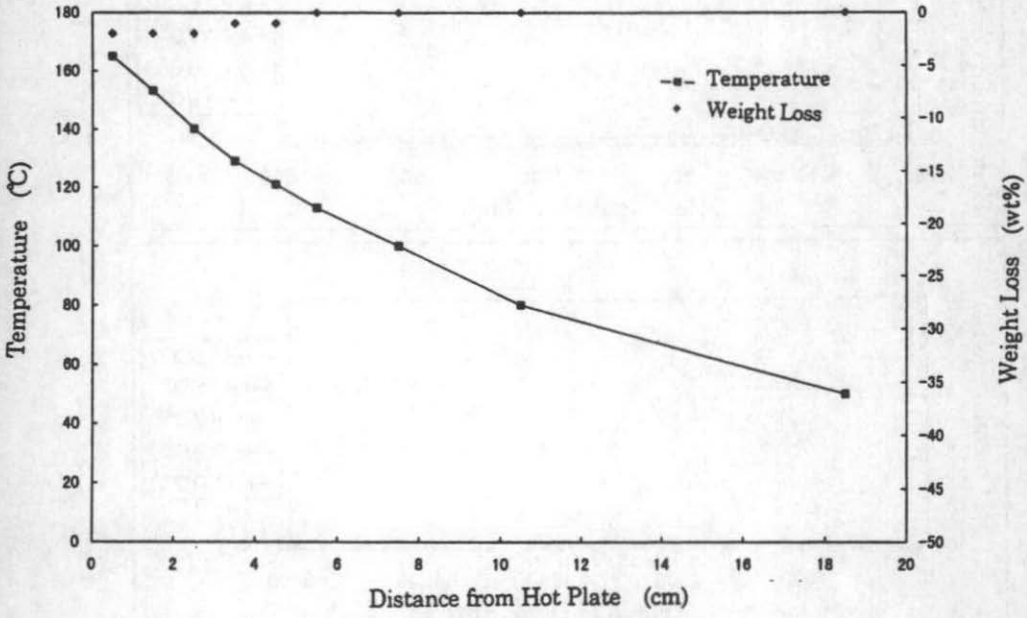


Figure 2-2 Temperature Distribution and Weight Loss of Block Heating Test Sample after 2000hr