DEVELOPMENT OF RUBBER-TYPE NEUTRON SHIELDS FOR TRANSPORT/STORAGE PACKAGINGS

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Abstract

DEVELOPMENT OF RUBBER-TYPE NEUTRON SHIELDS FOR TRANSPORT/STORAGE PACKAGINGS.

Two kinds of rubber-type neutron shields for transport/storage packagings have been developed. In Type I, silicone rubber is the base material. Titanium hydride, boron carbide and other additives are included. This material offers good liquidity at processing, which makes it possible to apply this rubber shield to shapes with complicated geometry. The density is 1.8 g/cm^3. With Type II, ethylene-propylene rubber is the base material. Boron carbide, antimony oxide and other additives are included. This is a solid type shield, and machining is needed before fitting to the proper location. The density is 1.0 g/cm^3. Several measurements were performed for both types to evaluate the shielding property, heat resistance, fire resistance, strength, weather resistance, thermal conductivity and thermal expansion. The neutron shielding ability of Type II is found to be better than that of water, while that of Type I is slightly less than that of Type II. A shielding calculation using a Monte Carlo code was performed to evaluate the measurement results. The experimental results and analytical results are in close agreement. The other main properties of the shields are as follows: The allowable temperatures of Types I and II are 170°C and 140°C, respectively; in the event of fire, both types are self-extinguishable in air.

1. INTRODUCTION

The development of neutron shielding materials is becoming more and more important for the design of transport/storage packaging. Burnup of fuels is increasing owing to the trouble free operation of nuclear reactors and the employment of high performance fuels. Consequently, the quantity of neutrons generated in spent fuels is greatly increasing, so that packaging must have a neutron shield which is highly effective from the point of view of both design and materials.

Many conventional spent fuel transport packages use water or a mixture of ethylene glycol and water as their neutron shield. These liquids are easily contained in cavities and are readily available at low cost. However, these shields require a great deal of space in the cavities to compensate for the increase in the volume of the liquid resulting from the decay heat of the spent fuel. Since neutrons pass through this space without attenuation, a complicated arrangement must be installed in the package to prevent neutron streaming. Another disadvantage of a liquid shield is that, because these shields are usually located near the outer surface of the packaging, there is the danger that all of the liquid might escape under accident conditions. As a result, precautions to ensure that a sufficient part of the material survives to provide suitable shielding after an accident are a serious consideration. These precautions usually increase the thickness and the weight of the packaging wall and reduce the spent fuel payload.

To overcome the disadvantages of a liquid shield, rubber-type neutron shields have been developed. Since rubber-type neutron shields are not liquid they do not require any container. Therefore, additional space is not necessary in the shield and there is no liquid to escape from the shield under accident conditions.

2. REQUIREMENTS FOR NEUTRON SHIELDS FOR PACKAGINGS

Generally, the density of a neutron shielding material is lower than that of steel, which is often used for the main body of packagings. Therefore, it is desirable to position the neutron shields outside the main body of the packaging rather than inside, since the overall weight of the packaging will be less.

The neutron shields which would be located near the outer surface of the package must fulfil the following requirements:

(a) *Shielding property:* The shield should have high neutron shielding efficiency. The shielding property in this case must cover the dose rate, not only of the neutrons, but also of the secondary gamma rays which are emitted as the result of neutron absorption.

(b) *Heat resistance.* The packaging shield is heated by the decay power of the spent fuel and the temperature becomes very high. The shield should be resistant to heavy heat damage which could reduce the efficiency of the shielding, even if the packaging is exposed to high temperatures for a long storage period.

(c) *Fire resistance.* The shield should retard fire even under fire test conditions, which by regulation include exposure of the packaging to an atmosphere of 800°C for 30 minutes. If the shield begins to burn during the test, the fire should extinguish itself immediately after the heat source is removed. This means the shield should be self-extinguishing.

(d) *Strength.* The packaging must survive several drop tests, during which the shield should not sustain any heavy damage which would reduce the efficiency of the shielding, such as cracks or drop out.

(e) *Weather resistance.* The packaging will be used for transport or storage for a long period, during which the shield should not suffer any heavy damage which would lead to deterioration of the shielding property under adverse weather conditions.

3. SPECIFICATIONS OF RUBBER-TYPE SHIELDS

Two kinds of rubber-type neutron shields have been developed for packagings. The specifications of the shields are as follows.

3.1. Type I shields

Silicon rubber is the base material. Titanium hydride, boron carbide and some other additives are included. Silicon rubber offers good liquidity at processing and is excellent in terms of heat and fire resistance. However, the silicone rubber has less neutron shielding ability than water because of the smaller atomic number density of hydrogen. The atomic number density values of hydrogen in silicone rubber and hydrogen in water are 5.0×10^{22} and 6.7×10^{22} atoms/cm^3, respectively.

Titanium hydride is the best neutron shield, as its hydrogen atomic number density is 9.0×10^{22} atoms/cm^3. There are many restrictions on the use of titanium hydride directly for the neutron shielding of a packaging because this material is a kind of powder metal. The Type I shields are made by mixing titanium hydride into silicone rubber. The material keeps the merits of silicone rubber and has a higher hydrogen content than the original.

If the low energy neutrons that are moderated enough by hydrogen in the shield are absorbed by hydrogen, secondary gamma rays of high energy are emitted by the (n, γ) reaction of hydrogen. The gamma dose rate may be several times as great as the neutron dose rate in some situations. In order to control the gamma rays emitted from Type I, the proper quantity of boron carbide must be included. Boron carbide contains boron-10 which can absorb numbers of the low energy neutrons and never emits secondary gamma rays.

The density of Type I is 1.8 g/cm^3. The atomic number densities of hydrogen and boron-10, the elements which are the most important for neutron shielding, are 5.5×10^{22} and 4.1×10^{20} atoms/cm^3, respectively.

Type I offers good liquidity at processing, which makes it possible to apply this shield to shapes with complicated geometries. Furthermore, Type I is excellent in terms of heat and fire resistance, which means that this shield can be set up on the outer surface of the packaging directly with no cover.

3.2. Type II shields

Ethylene propylene rubber (EPR) is the base material. Boron carbide, antimony oxide and other additives are included.

The atomic number density of hydrogen in EPR is 7.4×10^{22} atoms/cm^3, which is higher than that of water. This means EPR is a better neutron shield than water. With respect to heat and fire resistance, EPR is a good material in the rubber category but is less effective than silicone rubber. Type II shields include some quantity of antimony oxide as the flame retardant to improve the heat and fire resistance. For the same reason as with Type I shields, boron carbide is included in Type II.

The density of Type II is 1.0 g/cm^3. The atomic number densities of hydrogen and boron-10 are 6.7×10^{22} and 3.2×10^{20} atoms/cm^3, respectively.

In terms of the total dose rate of neutrons and secondary gamma rays, Type II is a better shield than water because it has the same hydrogen content as water and includes more boron-10.

The fire resistance of Type II is not sufficiently high, though it is improved by the addition of antimony oxide. It is desirable for Type II to be installed inside the outer wall of the packaging or to have an appropriate cover to protect against direct fire. The liquidity of Type II at processing is also less. Therefore, Type II needs machining before fitting to the proper location.

4. PROPERTIES OF RUBBER-TYPE SHIELDS

4.1. Shielding property

4.1.1. Experiment

Two types of experiments were performed. Firstly, the variation of the neutron dose rate as a function of the thickness of each shield was measured with a californium-252 neutron source. Secondly, the ratios of the secondary gamma dose rate to the neutron dose rate and the neutron spectrum after the penetration of each shield were measured, using the YAYOI fast neutron source reactor at the University of Tokyo.

Four test pieces were prepared for the Type I and Type II shields, each 50 cm in width, 50 cm in height and 3 cm in thickness. Three kinds of measurements were performed for each type; with a single piece, two pieces together and four pieces together.

4.1.1.1. Measurement of neutron dose rate

The neutron source was 1 mCi[1] of californium-252 and the detector was a Studsvik 2202D rem counter. The distance between the source and the detector was about 1 m and the test pieces were located between them near the detector.

[1] 1 Ci = 37 GBq.

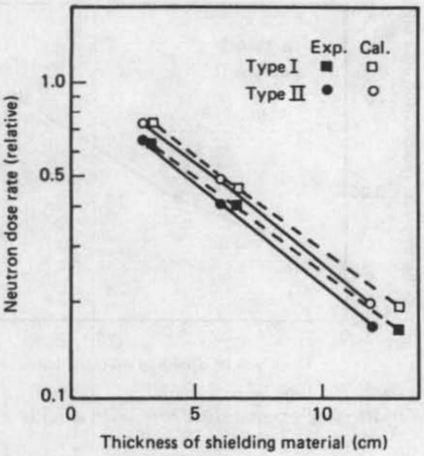

FIG. 1. Comparison of measured and calculated neutron dose rates (^{252}Cf point source).

The results of measurements are shown in Fig. 1. It can be seen that the Type II is a better neutron shield than Type I.

The removal cross-section, which is an important shielding property, is usually obtained by means of the following equation:

$$D_i/D_0 = \exp(-\Sigma_R X)$$

where D_i is the neutron dose rate with shielding
D_0 is the neutron dose rate without shielding
Σ_R is the removal cross-section (cm^{-1})
X is the shielding thickness (cm).

The removal cross-sections were slightly varied in accordance with the number of specimens in the test. In the measurements made on four pieces together, the removal cross-sections of Type I and Type II were 0.14 and 0.15 cm^{-1}, respectively.

In addition, measurements with a single piece, using three different test pieces of the same kind of material, were performed three times. There were no significant differences between the three measured values. This confirmed that the qualities of the three test pieces were consistent.

4.1.1.2. Measurement of secondary gamma dose rate and neutron spectra

The neutrons from the YAYOI neutron source reactor are collimated to a beam source of 5 cm in diameter. The detectors for the neutron dose rate, gamma dose rate and neutron spectrum were a rem counter, an ion chamber and

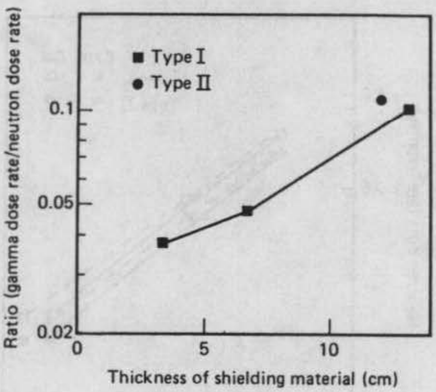

FIG. 2. Variation of gamma dose rate with shielding thickness.

an ^{3}He proportional counter, respectively. The experimental configuration was the same as in the measurement using californium-252.

The ratios of the secondary gamma dose rate to the neutron dose rate are shown in Fig. 2. The thicker the shield, the higher this ratio becomes. The secondary gamma dose rates were about 10% of the neutron dose rates in the measurements with four pieces together, which showed the highest figure in these experiments. This figure is acceptable for the neutron shield of the packaging. The boron-10 included in the shield worked effectively.

If a package requires an extremely thick shield, it is recommended that the amount of boron carbide in the shield be increased to offset the increased gamma rays that result from the increased thickness.

The neutron spectra after the penetration of each shield and the neutron source spectrum are shown in Fig. 3. The source spectrum has a 1/E form. The neutron spectra after penetration of the shields are nearly flat. There were no special peaks in the spectra after penetration of the shields. This means that both shields are effective over the whole neutron energy range.

4.1.2. Calculation

The calculations were performed by using the MORSE-CG three dimensional Monte Carlo calculation code with a surface crossing estimator, which could take into account the detector size.

The cross-section library used in MORSE-CG was DLC-23/CASK [2]. The number of neutrons generated in one calculation was 30 000 histories. The fractional standard deviations of each calculation were less than 0.05.

The calculation values are shown in Fig. 1 together with the measured ones. These values were normalized to the dose rate with no shield. The variation in

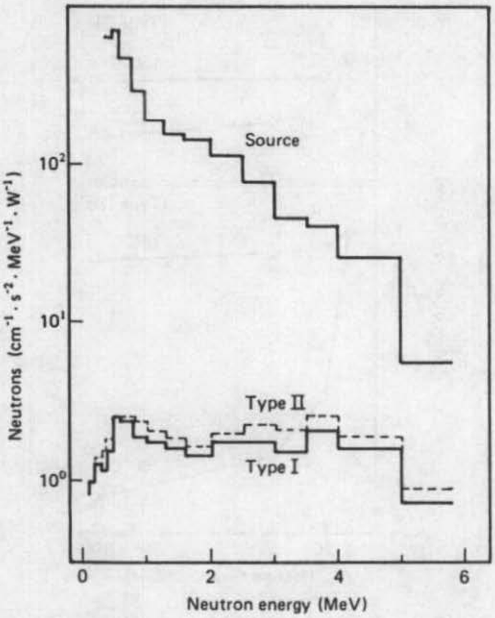

FIG. 3. Neutron spectra measured by ^{3}He counter.

attenuation of the calculated values agrees well with that of the measured ones, but the absolute value of the calculated results seems to be slightly higher.

The removal cross-sections were calculated as 0.124 and 0.135 for the specimens with four pieces together (Type I and Type II, respectively). These values agree closely with the experimental ones.

Calculations were also performed for water. The calculated neutron dose rate with water was almost the same as with Type II, i.e. the two shields had an equivalent neutron shielding property.

4.2. Heat resistance

Thermal decomposition of a neutron shield would affect its shielding property and is closely related to its mass. Therefore, for each shield the mass reductions over time at various temperatures were measured. The results are shown in Fig. 4. The mass reduction occurred in direct proportion to temperature and time, with a rapid reduction at the beginning.

The heat resistance of the two types of shield was evaluated using the results of these measurements. The allowable temperature of a packaging neutron shield was defined as follows – the temperature at which the mass would be

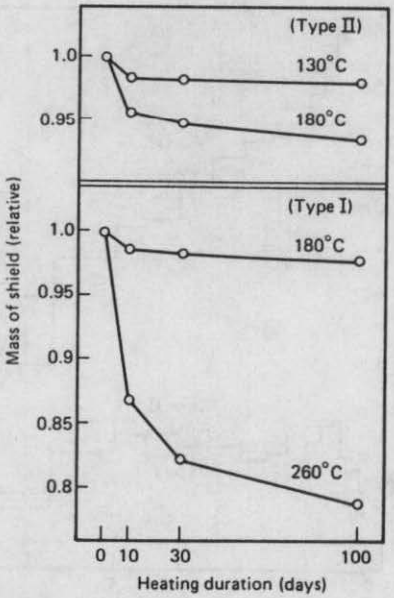

FIG. 4. Mass reduction under high temperature conditions.

reduced by 10% over the entire service period, assuming, for example, a neutron shield for a transport packaging under the following circumstances:

Service period	:	20 years
Transportation	:	1.5 times per year
Period of transportation	:	100 days

Since the mass reduction was measured for only 100 days at the longest, the periods over which mass would be reduced by 10% at each temperature were obtained by extrapolation and interpolation of the measured results. The following values were obtained:

Type I:	180°C	1250 days
	260°C	8 days
Type II:	130°C	6400 days
	180°C	300 days.

The correlation between the temperature (T) and time (t) for rubber can be expressed by the following equation [3]:

$$\ln(t) = A + E/RT$$

where A is a constant
E is the activation energy
R is the gas constant.

From the above equation and figures, the following allowable temperatures were obtained:

Type I ~ 170°C
Type II ~ 140°C.

These temperatures are high enough for the material to be used in packaging neutron shields. If the service temperatures of Type I and Type II shields are assumed to be 150°C and 130°C, the service periods would be approximately 70 and 30 years, respectively.

4.3. Fire resistance

The oxygen indices of Type I and Type II shields were measured to evaluate their fire resistance. The oxygen index represents the volume percentage of the minimum oxygen content needed to maintain combustion of material under a certain test method. The oxygen content of air is 21%. Therefore, materials of which the oxygen index is larger than 21% would be classified as self-extinguishable in air. The larger the oxygen index, the more self-extinguishable the material. The oxygen indices of Type I and Type II were 35.5 and 21.5, respectively. This means both are self-extinguishable in air.

4.4. Other properties

4.4.1. Strength

A rubber-type shield would not crack or drop out under drop tests because of its elasticity. Further, if necessary, it is possible to protect and reinforce the shield by proper casing or covering.

4.4.2. Weather resistance

The base materials of Type I and Type II shields, silicone rubber and EPR, have excellent intrinsic weather resistance and this would remain intact even with additives present.

4.4.3. Thermal conductivity

The thermal conductivities of Type I and Type II shields are 1.84×10^{-3} and 4.28×10^{-4} cal/(cm·s·°C), respectively.

4.4.4. Thermal expansion coefficient

The thermal expansion coefficients of Type I and Type II shields are 1.80×10^{-4} and 2.22×10^{-4} (deg C)$^{-1}$, respectively.

5. CONCLUSION

The shielding and other important properties mentioned above show that the Type I and Type II shields are very effective for a transport/storage packaging.

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