Development of Highly Effective Neutron Shields and Neutron Absorbing Materials

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ABSTRACT

A wide range of materials, including polymers and hydrogen-occluded alloys that might be usable as the neutron shielding material were examined. And a wide range of materials, including aluminum alloys that might be usable as the neutron-absorbing material were examined. After screening, the candidate material was determined on the basis of evaluation regarding its adaptabilities as a high-performance neutron-shielding and neutron-absorbing material. This candidate material was manufactured for trial, after which material properties tests, neutron-shielding tests and neutron-absorbing tests were carried out on it. The specifications of this material were thus determined.

This research has resulted in materials of good performance; a neutron-shielding material based on ethylene propylene rubber and titanium hydride, and a neutron-absorbing material based on aluminum and titanium hydride.

INTRODUCTION

Japanese electric utilities are making efforts to enhance the burnup of fuels in order to make efficient use of light water reactors, causing the fuels to be more highly enriched and the radiation source intensity (neutron source intensity in particular) of used fuels to be increased further.

Accordingly, in the course of research aimed at designing and developing en irradiated fuels storage casks capable of efficiently containing such fuels with high burnup, a high-performance neutron-shielding material for use as the main body of the packaging and a neutron-absorbing material to be used for the basket in the shell were developed.

NEUTRON SHIELD

· Requirement Approach

Neutron shields which would be located near the outer surface of the packaging must fulfill the following requirements:

a) Shielding property: The shield should have high shielding efficiency not only for neutrons, but also for the secondary gamma rays which are emitted as a result of neutron absorption.
b) Heat resistance: The shield should be resistant to heat damage caused by the decay power of spent fuel.

c) Fire resistance: The shield should be self-extinguishing when fired at 800 ℃ for 30 minutes.

d) Strength: The packaging must survive several drop tests, during which the shield should maintain shielding efficiency. For a liquid shield such as water, there is the danger that all or some of the liquid might escape during the tests.

With the above requirements taken into account, 5 types of materials were selected. Table 1 shows the specification of each material. As examples of conventional materials, resin and water were also listed. Among these, resin underwent the same tests to which the selected materials were subjected, for reference.

· Applicability tests

Neutron-shielding materials were subjected to the applicability test for the purpose of studying their applicability as materials for shipping casks and storage casks. In the applicability test, consisting of a mechanical test, a heat resistance test, a fire test, and a thermophysical property test, the testing conditions were determined in detail in accordance with the characteristics of the materials to be tested.

In particular, with titanium hydride, the compactness to be achieved was determined after conducting a compactability test, since titanium hydride is in the form of a powder in its normal state. Figure 1 shows the results of the compactability test. A pressure ranging from 8 to 10 tonne/cm^2 is required to achieve a compacting density of 90% of the theoretical density, even though this is affected by both the particle size and the boron content of the powder.

Figure 2 shows the results of a long-term heat-resistance test conducted as part of the applicability test. Titanium hydride is superior to other materials in terms of its long-term resistance, followed by silicone rubber. Polypropylene and ethylene propylene rubber, both of which are superior to the reference materials, can be used at relatively low temperatures.

· Capability Tests

Two types of test were performed. One was designed to obtain the fundamental shielding property of each material when combined with a steel plate that simulates the main body of the packaging. The steel plate thickness is 20cm. Table 2 summarizes the type of shielding experiment.

Fig. 3 shows the case combined with a steel plate.When the steel plate is not provided, the neutron dose rate after passing through the 15-cm-thick EP-B shield is about 70% that of the resin shield, while the dose rate after passing through the 15-cm-thick TiH_2 shield is about 20% that of the resin shield. A TiH_2 shield about 10cm thick can provide the same dose rate as a 15-cm-thick resin shield. When a steel plate is combined, the neutron dose rate after passing through the 15-cm-thick EPT-B shield decreases by about 10% when neutrons pass though the 15-cm-thick TiH_2 shield. Because the ratio of thermal neutrons increases when a steel plate is combined, attenuation in the shield generally increases. Fig. 4 shows how gamma rays attenuate in the shield when a steel plate is used. Each specimen exhibits a larger gamma dose than that of the resin sield, in particular in the 0.5cm range of the shield. After passing through the 15-cm shield, the gamma dose increases by about double that of the resin shield. Neutron-shielding requires the neutron scattering and moderation process. With this process, gamma rays are generated. Needless to say, the greater the attenuation, the more gamma rays are generated.

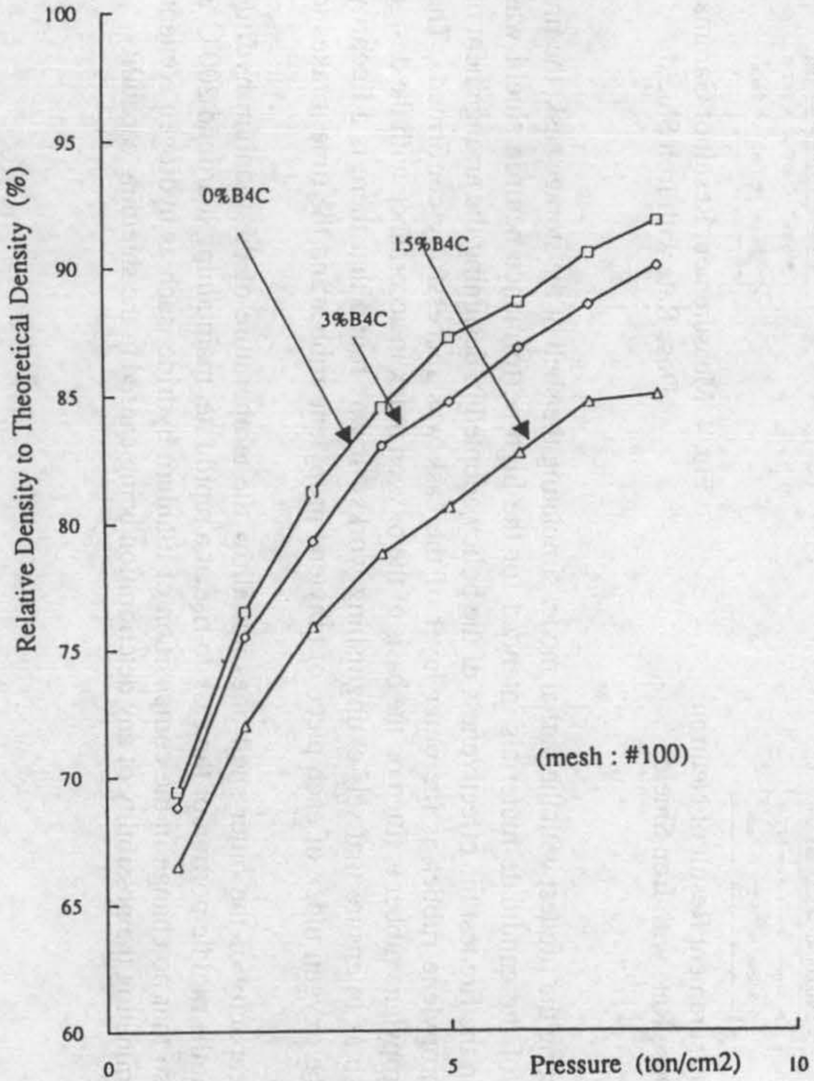

Figure 1. Forming Pressure of Titanium Hydride Powder

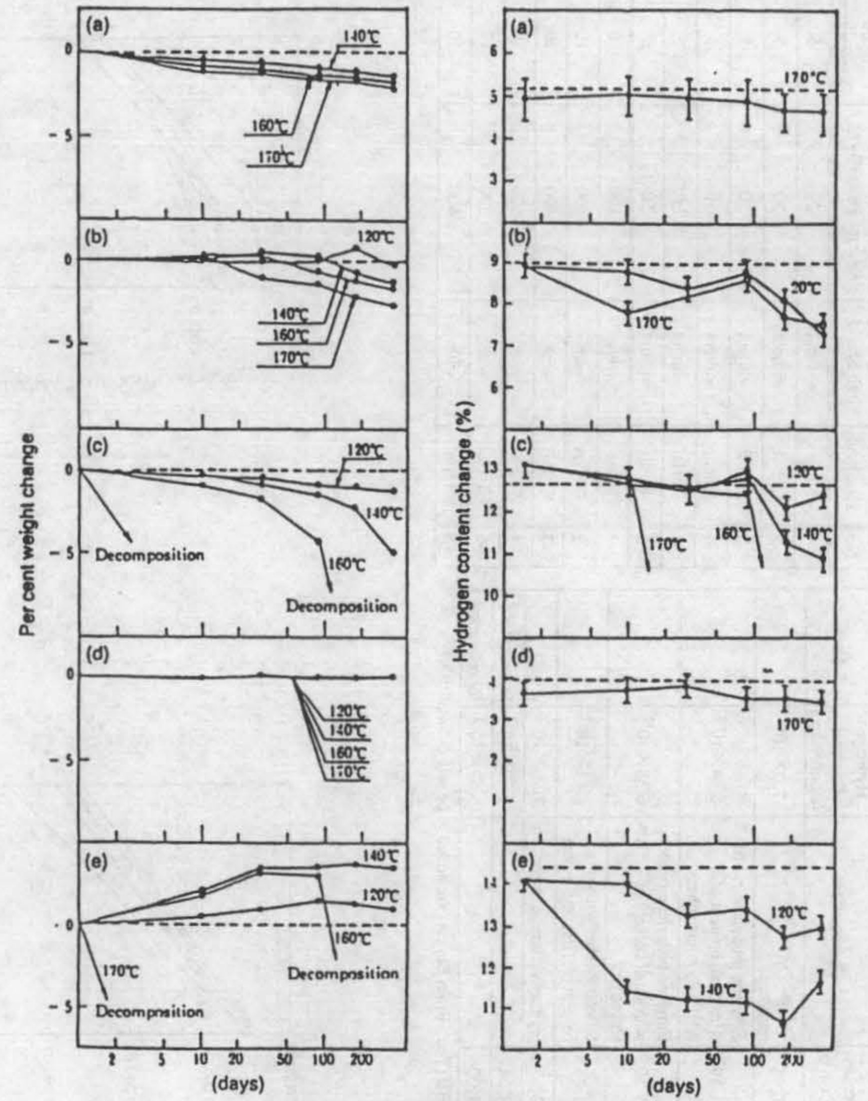

Figure 2 Thermal Degradation data

(a) Silicone rubber (b) Ethylene-propylene rubber (c) Polypropylene

(d) Titanium hydride (e) Polyethylene

Table 1 Specification of Shield Materials

Specimen	Density (g/cm^3)	Composition	Hydrogen Contents (atoms/b cm)	Note
PP	0.95	Polypropylene with fire retardant	7.0×10^{-2}	
EPT-A	1.19	Ethylene propylene rubber with small content of $Al(OH)_3$ * and others	6.44×10^{-2}	
EPT-B	1.50	Ethylene propylene rubber with small content of TBE * and others	6.38×10^{-2}	
Silicon	1.75	Silicon Rubber with small content of TiH_2 and others	5.41×10^{-2}	
TiH_2	3.33	Hydrogen storage material	7.76×10^{-2}	
Resin	1.58		4.70×10^{-2}	for reference
Water	1.00		6.69×10^{-2}	for reference

* : $Al(OH)_3$ and TBE (Tetra Brom Ethane) are added to be self-extinguishing

Table 2 Case of Experiment

No	Specimen	Iron Shield	Reactor Power(Wh)	Beam Diameter(cm)
1-1	None	without	20	5
-2	EPT-A	without	50	5
-3	EPT-B	without	50	5
-4	Silicon	without	50	5
-5	TiH_2	without	150	5
-6	Resin	without	50	5
2-1	None	with	100	10
-2	EPT-A	with	400	10
-3	EPT-B	with	400	10
-4	Silicon	with	500	10
-5	TiH_2	with	300	10
-6	Resin	with	400	10

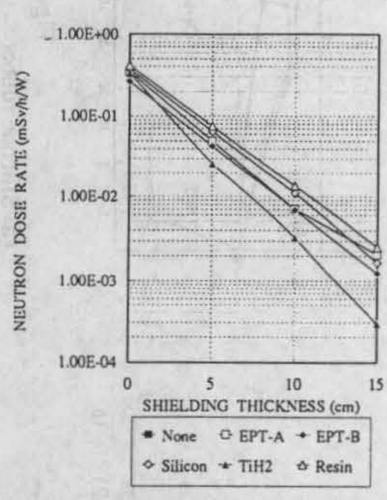

Fig. 3 Measurement Result of Neutron Dose Rate with Iron Shield

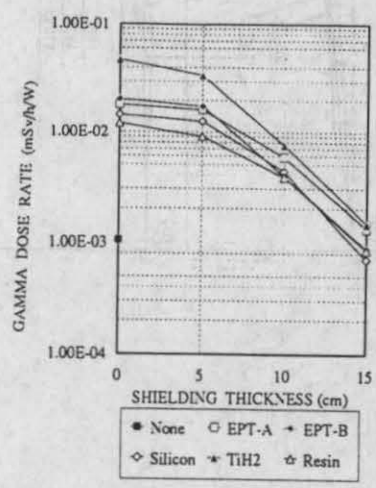

Fig. 4 Measurement Result of Gamma Dose Rate with Iron Shield

· Fire test

By conducting the fire test with the partial model simulating the shell of the storage cask, the fire resistance of the candidate materials intended for the high-performance neutron shield was verified. In the fire test the effectiveness of the policy adopted in designing the arrangement of ethylene propylene rubber as the outer layer of the cask was successfully confirmed. The ethylene propylene rubber is fitted on the basis of the oxygen index in accordance with the design principle so as to ensure that self-extinguishing works. It was found that there is a linearity between the oxygen index of each piece of ethylene propylene rubber and the time it takes to extinguish.

The heat capacity of the outer shell does not allow the temperature of the titanium hydride disposed at the middle portion of the cask to rise at a rapid rate, maintaining it around 200℃ at the most, so that no change in the composition of titanium hydride, such as hydrogen content, occurs, eliminating the possibility of any deterioration being caused in the shielding capability.

· Conclusion

Table 3 gives an overall evaluation of the high-performance neutron shielding materials on the basis of the results of the applicability tests which have been carried out so far, their fundamental capability and operational suitability as neutron-shielding material, and the value added as newly developed material. As shown in the table, as high-performance neutron-shielding material, ethylene propylene rubber and titanium hydride (high density) have such excellent applicability that it is practicable from the viewpoint of designing to employ the former as an outer layer of the cask and the latter at such portions as the trunnion where shielding tends to be insufficient locally.

NEUTRON-ABSORBING MATERIALS

· Requirements

The increment in both the initial enrichment and calorific value in the spent fuel, initiated by the recent movement to achieve high burnup of fuel for the light water reactor, has given rise to the following requirements regarding neutron-absorbing materials:

a. Absorbing property; It is desirable that neutron-absorbing material cannot only absorb thermal neutron effectively but also lower fast neutron by effectively thermalizing it.
b. Heat transfer; To dissipate the heat generated in spent fuels at a noticeable rate, neutron-absorbing materials have to possess reasonably high heat conductivity.
c. Mechanical properties; shipping casks and storage packages should maintain their mechanical integrity even if subjected to mechanical shocks.

In accordance with these requirements, the three kinds of alloys having aluminum as their base material and the four kinds of compacts of titaniun hydride expected to improve both moderation and absorption abilities were tentatively selected.

· Applicability test

The candidate neutron-absorbing materials tentatively chosen on the basis of the requirements stated in the previous section were submitted to the applicability test with the aim of studying the applicability of the selected materials to the shipping and storage package. The applicability test was composed of four items, mechanical, corrosion, thermophysical test and workabiity test, and the testing condition for each candidate material was definitely created according to their peculiarities. The results from the applicability test showed that alaminum alloy with addition of rare earth elements is inferior in corrosion resistance, reducing the possibility of employing it in actual packages.

· Capability test

The capability test has demonstrated that with the three cases, test 5 (structural material; Al, moderator; TiH -15 % B_4C), test 6 (structural material; SUS, moderator; TiH -15 % B_4C), and test 7 (structural material; B_4C 15%-Al, moderator; water), a low thermal neutron flux was detected on the downstream side of the moderator. Each of these tests, as a whole system, contained much more boron than the other systems did. With test 7, in particular, a low thermal neutron flux was measured on the upstream side of the absorbing material, followed by test 5 and test 6 in this order, with which the strength of the thermal neutron flux decreased(Fig.5). This can be attributed to the properties of the structural materials employed.

Table 3 (1/2) : Summary of development of high-performance neutron absorbing materials

Item		Polypropylene	Ethylene propylene rubber		
			Type A Fire-resistant materials in halogen family	Type B Fire-resistant materials in hydroxide family	Type C Fire-resistant materials
Characteristics		Superior to polyethiene in heat-resistance. Materials having the same neutron shielding capability were provided with fire resistance	Produced by polymerization of the ethylene group and propylene group, with additional fire-resistant material in the halogen family having excellent heat-resistance with hign hydrogen content	Produced by adding fire-resistant material in halogen to the material mentioned in the column on the right	Produed by adding a mixture of fire-resistant material in the halogen family and in the hydroxide family to the material mentioned in the column on the right
Fundamental capabilities	Neutron shielding capability	Excellent, because of high density of hydrogen atoms ○	Excellent, because of high density of hydrogen atoms ○	Excellent, owing to high density of hydrogen atoms, best in EPT ○	Excellent, owing to high density of hydrogen atoms, best in EPT ○
	Weight reduction	Excellent, owing to low mass density and superior shielding capability ○	Excellent, owing to low mass density and superior shielding capability ○	Excellent, owing to low mass density and superior shielding capability ○	Excellent, owing to low mass density and superior shielding capability ○
	Long-term heat resistance	Good; the result of the long-term heat test proved that it could be used at about 120℃ △	Excellent; the result of the long-term heat test proved that it could be used at about 140℃ ○	Excellent; the result of the long-term heat test proved that it could be used at about 140℃ ○	Excellent; the result of the long-term heat test proved that it could be used at 160℃ ○
Applicability	Workability	Excellent, No problem because of extensive use in general industries ○	Excellent, No problem because of extensive use in general industries ○	Excellent, No problem because of extensive use in general industries ○	Excellent, No problem because of extensive use in general industries ○
	Cost	Excellent, nearly as costly in both manufactruring and processing as in general industries ○	Excellent, nearly as costly in both manufactruring and processing as in general industries ○	Excellent, nearly as costly in both manufactruring and processing as in general industries ○	Excellent, nearly as costly in both manufactruring and processing as in general industries ○
Material characteristics		Excellent, but cannot be used as heat conduction member for the same reason that regular plastics cannot be used ○	Excellent, but cannot be used as heat conduction member for the same reason that regular plastics cannot be employed ○	Excellent, but cannot be used as heat conduction member for the same reason that regular plastics cannot be used ○	Excellent, but cannot be used as heat conduction member for the same reason that regular plastics cannot be used ○
Other applicabilities (Possibilities of using in other apparatus)		Excellent, usable in environment where heat resistance greater than that of polyethylene is required ○	Excellent, usable in environment where heat resistance greater than that of polyethylene is required ○	Excellent, usable in environment where heat resistance greater than that of polyethylene is required ○	Excellent, usable in environment where heat resistance greater than that of polyethylene is required ○
Summary		Good, althhough inferior in heat resistance, usable in actual machine with ease △	Excellent, can be used in actual machine with ease, and well balanced both in neutron shielding capability and heat resistance ○	Excellent; the statement in the next column on the left is true for the material, inferior in mechanical property and superior in neutron shielding capability compared with material mentioned in the next column on the left ○	Excellent; the statement in the second column on the left is true for this material. Well balanced in mechanical properties and neutron shielding capability ○

Table 3 (2/2) : Summary of development of high-performance neutron absorbing materials

Item		Titanium hydride		Reference materials	
		Low density(about 70%)	High density(about 90%)	TN resin	Polyethyle
Characteristics		Produced by solidifying by using press material having the highest hydrogen content among materials in metal hydride	Produced through using press by solidyfying material having the highest hydrogen content among aetal hydride, attaining greater density than that of the material mentioned in the column on the left	Used in France in actual shipping casks made of unsaturated poiyester	Common as a neutron shielding material in nuclear faci??ties and empioyed in boty Germany and the US in actual storage casks
Fundamental capabilities	Neutron shielding capability	Excellent, have about the same hydrogem atom density as water ○	Excellent, highest density of hydrogen atoms among the candidate materials ○	Good, but inferior to the other, materials in density of hydrogem atoms △	Excellent, due to high density of hydrogen stoms ○
	Weight reduction	Excellent from vlempoint of density and shielding capability ○	Excellent, due to superior neutron shielding capability ○	Good in terms of density and shielding capability △	Excellent, due to iow mass density and high shielding capability ○
	Long-term heat resistance	Excellent; the result of the long-term heat test proved that it could be used at 160℃ ○	Excellent; the result of the long-term heat test proved that it could be used at 160℃ ○	Good, technical data provided by the manufacturer proved that it can be used at about 120℃ △	Fair, long-term heat test proved that it can be used at temperatures a little higher than 100℃ ×
Applicability	Workability	Excellent, Can be solidified by using low pressure press ○	Good, High pressure press is required, can be cut with case △	Excellent, Compiicated can be done with injection method ○	Excellent, No problem bacause of extensive use in general industries ○
	Cost	Good, reiatively castly in purchase of raw material and in fabrication △	Fair, Extremely castly in fabrication ×	Excellent, expensive because of specificity ○	Nearly as costly in both manufacturing and processing as in general industries industries ○
Material characteristics		Fair, fairly brittle ×	Excellent, strong enough to maintain the shape, heat conduction takes place to some extent ○	Excellent, but cannot be used as heat conduction member for the same reason that regular plastics cannot be used ○	Excellent, but cannot be used as heat conduction member for the same reason that regular plastics cannot be used ○
Other applicabilities (Possibilities of using in other apparatus)		Excellent, usabie in environment where great heat resistance is required ○	Excellent, usabie in environment where neutron flux is so intense that high heat resistance is required ○	Existent material △	Existent material △
Summary		Good, application in actual machine is hard to achieve because of instability of the material △	Excellent, but use is limited to local area because of the poor workability and expensiveness, although it is superior in neutron shielding capability and heat resistance ○	Existent material △	Existent material △

Table 4 Combination of materials in neutron absorption test

Test No.	Material	
	Neutron absorbing material	Structural material
1	TiH_2	Al (B 3%)
2	TiH_2	SUS(B 1%)
3	TiH_2 (B4C 3%)	Al
4	TiH_2 (B4C 3%)	SUS
5	TiH_2 (B4C 15%)	Al
6	TiH_2 (B4C 15%)	SUS
7	H_2O	Al (B_4C 15%)
8	H_2O	SUS(B 1%)

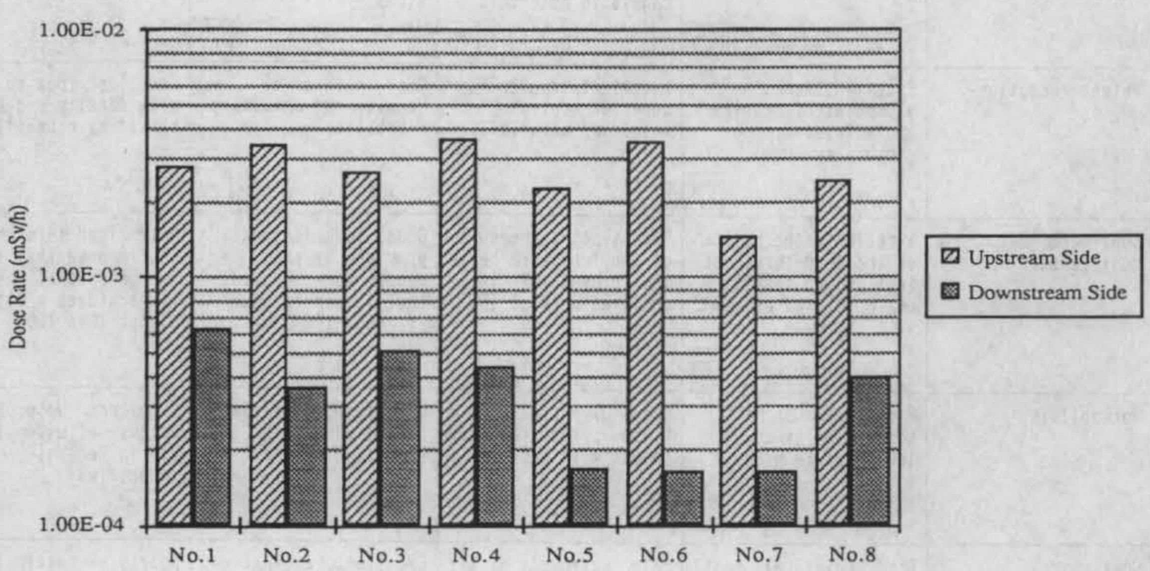

Fig.5 Result of Neutron Absorption Test

· Conclusions

Table 5 gives the results of the overall evaluation of the high-performance neutron-shielding materials conducted on the basis of the result gained from the applicability tests which have been conducted so far, the fundamental properties and usability as neutron-absorbing material, and value added as newly developed material. As shown in the table, as high-performance neutron shielding material, both titanium hydride and powder aluminum alloy are superior in usability, so that it is practical from the viewpoint of designing to utilize the former as neutron-moderating material having neutron-absorbing ability, and the latter as both structural members and heat-conducting members, as it is excellent in neutron-absorbing capability.

ACKNOWLEDGMENTS

The authers wish to express their thanks to Mr. H.Yokoyama and other staff members of Central Research Institute of Electric Power Industry for their assistance and helpful discussions.

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Table 5 (1/2) : Summary of development of high-performance neutron absorbing materials

Item		Neutron absorbing materials (structural material)			Neutron moderator
		Cast aluminum alloy	Powder aluminum alloy		Titanium hydride (B_4C : 0-15%)
		(rare earth element)	With addition of B_4 C	With addition of BN	
Characteristics		Aluninum is used base material with addition of a conbination of rate earth metal having large thermal neutron absorption cross section	With aluminum used as base metal, addition of boron (B_4 C) is increased by empoloying powder metallurgy method	Aluminum is used as base metal, and addition of boron (BN) is increased through use of powder metallurgy method	To effectively moderate and absorb neutron. TIH2, which has a high density of hydrogen atatoms, is solidified
Fundamental capabilities	Neutron shielding capability	Good, but not swo effective aince epithermal neutron is prominent lm package system △	Excellent, due to high boron concentration ○	Excellent, due to high boron concentration, but inferior in neutron absorbing capability to material deait with in the next column on the left ○	Excellent, depending on how this material is combined with structural material ○
	Weight reduction	Good, but low neutron absorption ability and low mechanical strength prevent improvement of the loading factor from improving △	Excellent, absorption capability and great mechanical strength contribute to improvement of the loading factor ○	Excellent, absorption capability and great mechanical strength contribute to improvement of the loading factor ○	Excellent, appropriate combination with structural material allows the loading factor to be improved ○
	Long-term heat resistance	No probiem, in cace of using for storage cask ○	No probiem, in cace of using for storage cask ○	No probiem in cace of using for storage cask ○	Inferior in maintaining shape under water, and thus requiring reinforce-ment such as stainless steel ciadding Γ
Applicability	Workability	Good, Vacuum casting is necessary. Excellent in workability, especially, in shaping △	Good, Ingots are manufactured with high-temperature static-pressure water press. Extruding, rolling, and cutting are applicable △	Good, Ingots are manufactured with high-temperature static-pressure water press. Extruding, rolling, and cutting are applicable ×	Goo, manufactured with high-pressure press and cam be cut △
	Cost	Good, expensiveness of rare earth elements contributes to imcreasing purchase cost of the material △	Good, high in production cost △	Good, high in production cost △	Good, high in production cost △
Characteristics of material		Good, but inferior in both mechanical properties and dispersion of neutron absorbing elements △	Excellent, possesses properties which fulfill requirements of material for the basket, except for defect of being reiatively hard and brittle ○	Excellent, possesses properties which fulfill requirements of material for the basket, except for defect of being reiatively hard and brittle ○	Good, strong enough to maintain its shape, and has reasonabie tharmal conductivity △
Other capabilitiews (such as applicabilities to different apparatus)		Good, can be used in a system where effective moderation is meeded △	Excellent, applicablic to areas other than packages that require a high thermal neutron absorbing avility ○	Excellent, applicable to areas other than packages taht require a high thermal neutron absorbing ability ○	Excellent, usable in areas that requira both heat resistance and high neutronmoderating capability ○
Summary		Good, inferior to powder aluminum alloy in fundamental capabilities, applicablilities, and characteristics △	Excellent, improvement in workability and reduction of cost are decisive factors for future use ○	Good, but inferlor to the material dealt with in the column on the left in both fundamental capabilities and workability △	Excellent, improvement in operational suitability is decisive factor for future use, but inferlor to powder aluminum alloy in fundamental capabilities ○

Table 5 (2/2) : Summary of development of high-performance neutron absorbing materials

Item		Reference material: Aluminum alloy with addition of B (low temperature)	Reference material: Stainless steel alloy with addition of B (low temperature)	Remarks
Characteristics		This existent material, which is employed in shipping casks, acts as the reference material to compare with the material (structural material) mentioned in the column on the left. (boron concentration : about 1%)	This wxistent material, which is employed in shipping casks, acts as the reference material to compare with the material (structural material) mentioned in the column on the left. (boron concentration : about 3%)	
Fundamental capabilities	Neutron absorption capability	Good, lower in boron concentration compared with powder aluminum alloy △	Good, lower in boron concentration compared with powder aluminum alloy △	
	Improvement in loading number	Inferior in neutron absorbing ability to the material mentioned in the next column on the left △	Inferior in neutron absorbing ability to the material mentioned in the next column on the left △	
	Long-term corrosion resistance	No probiem, in cace of using for storage cask ○	Excellent, no probiem in using this material in actual storage casks ○	
Applicability	Workability	Excellent, existent material, no probiems exist ○	Excellent, existent material, no probiems exist ○	
	Cost	Excellent, existent material, available at relatively low price ○	Excellent, existent material, available at relatively low price ○	
Characteristics of material		Good, but while having appropriate mechanical properties as structural material, Inferior in thermal conductivity △	Excellent, possesses satisfactory mechanical properties and thermal conductivity ○	
Other capabilities (such as applicabilities to different apparatus)		Existent material △	Existent material △	
Summary		Existent material △	Existent material △	While powder aluminum alloy still has probiems to be solved before being empioyed in practice. It seems reasonable to study the possibility of lowering the production cost of aluminum alloy added with