Development of New Type Concrete for Spent Fuel Storage Cask

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Abstract

Heat resistant concrete has been developed to make it possible to design a new type cask that has been designed on the same concept of metal cask technologies for use in high temperature conditions. The allowable temperature of conventional concrete is limited to less than 100 degrees Celsius because most of its moisture is free water and therefore hydrogen, which is effective for neutron shielding, can be easily lost. Our newly developed concrete uses chemically bonded water and as a result can be used under high temperatures.

1. Introduction

In Japan, 12,940tU of spent fuels had been used from 1966, when the first commercial nuclear power plant started operating, to 1997 and more than 6,400tU of them are stored. In recent years, spent fuels produced in a year mount to about 900tU and will mount up to about 1,400tU in 2010 [1]. In Japan, spent fuels are planned to re-process and the re-processing plant is now under construction at Rokkasho in Aomori prefecture. The capacity of re-processing plant is 800tU in a year, therefore spent fuels are necessary to be stored safely before they go under re-processing operation.

Japanese electric companies and other key agencies are preparing the interim storage facilities of spent fuel in order to start the operation by 2010. At present, some metal casks are planned to be used for interim storage because they are more reliable and has been actually used for transport and storage, but in the economical point of view, some conventional concrete casks have more advantage than metal casks and the technological standards for concrete cask are now being prepared.

In the United States, concrete casks with conventional concrete have been actually used and it contains most of its moisture as free water that evaporates at over 100 degrees Celsius. As shown in Figure 1, the conventional concrete cask has air inlets and outlets, and the natural convection of air between concrete cask and canister prevents the concrete from being subjected to high temperature. Then, it prevents the free water that is effective for neutron shielding, from evaporating and the strength of concrete from degrading. However these air inlets and outlets cause the radiation streaming for lack of shielding material and the salt-contained particles by seawater induced into concrete cask cause the corrosion of canister. In Japan, the latter problem is especially serious because interim storage facilities are planned to be located near the sea. For resolving the problem, two-phase stainless steel, which has excellent corrosion resistance but is expensive, is one of the most actual candidate materials for canister [2].

In this paper, the newly developed heat resistant concrete containing enough water for neutron shielding even in high temperature of over 150 degrees Celsius is reported. As shown in Figure 2, using this heat resistant concrete, air inlets and outlets indispensable for conventional concrete cask are not needed and the new concrete cask can be designed with the same concept of technology as that of metal casks actually used.

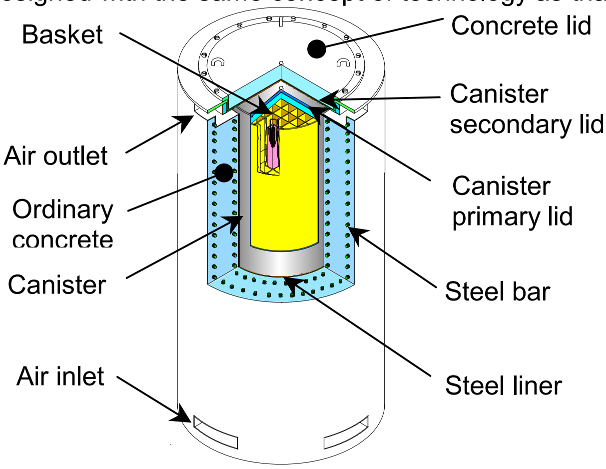


Fig.1 Conventional concrete cask

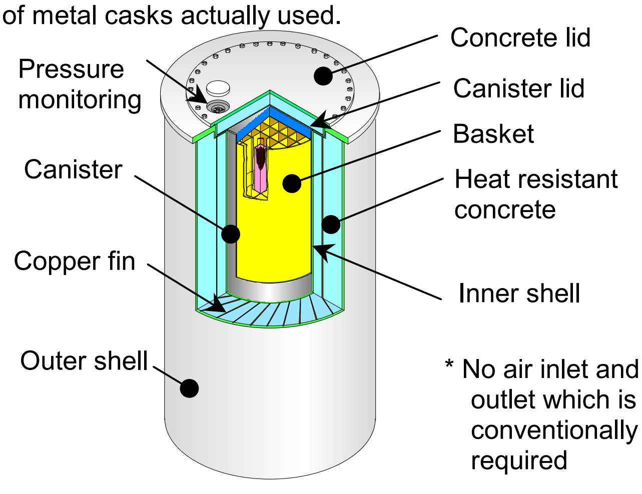


Fig.2 New type concrete cask

2. Development of heat resistant concrete

2.1 Purpose of development

The target of development is to make the heat resistant concrete material with keeping the shielding ability in the high temperature of over 100 degrees Celsius in order to remove the air inlets and outlets indispensable for the conventional concrete cask. The target temperature of heat resistance is more than 150 degrees Celsius taking account of the actual usage conditions.

2.2 Evaluation of material characteristics

2.2.1 Material composition

The most important characteristics of this material is being able to keep enough water (hydrogen) inside it under the high temperature condition of over 100 degrees Celsius to be effective for the neutron shielding. In this condition, conventional concrete material can not keep hydrogen which is in the form of the free water, but calcium hydroxide enables the heat resistant concrete to contain the enough hydrogen for neutron shielding since it has chemically bonded water. On the other hand, the concrete cask is also required to have gamma ray shielding ability. This heat resistant concrete has the excellent shielding ability not only for neutron but also gamma ray because it has the almost same density or even more as the conventional concrete with adding metal materials such as iron powder and steel fibers. Although this material does not contain coarse and fine aggregates, it is referred to as heat resistant “concrete” in this paper.

2.2.2 Basic material characteristics

Basic characteristics of this material to be important for practical use are measured at room temperature and 150 degrees Celsius and the results are shown in Table 1. Heat conductivity is measured by “Testing method for thermal conductivity of insulating fire bricks”(JIS R 2616), coefficient of linear expansion by “Measuring method of linear thermal expansion for building materials”(JIS A 1325), compressive strength by “Method of test for compressive strength of concrete”(JIS A 1108), specific heat by measuring liquid temperature including heated material and apparent specific gravity are calculated using weight and outer dimensions. The chemically bonded water content is calculated by measuring weight difference of calcium chloride before and after collecting water from sample material heated at 1000 degrees Celsius [7]. For comparison, the characteristics of ordinary concrete are also listed. From the results, the density of this material is almost same as ordinary concrete and the water content is two or three times as much as that of it. The difference of moisture content at room temperature and 150 degrees Celsius is about 5~6%, therefore this material has about 5 - 6% of free water evaporating at over 100 degrees Celsius. The compressive strength after heated is equivalent to that of high strength concrete.

Table 1 Properties of heat resistant concrete and ordinary concrete

	Heat resistant concrete		Ordinary concrete
	at 150 degrees-C	at room temp.	at room temp.
Density (g/cm^3)	2.2	2.3	2.25 - 2.3 [3]
Moisture content (mass%)	11	17	4 - 7 [4]
Heat conductivity (W/(mK))	1.4	2.0	2.6 - 2.8 [5]
Coefficient of linear expansion (1/K)	-	1.1×10^{-5}	1.0×10^{-5} [3]
Specific heat(kJ/(kgK))	-	0.9	1.0 - 1.3 [4]
Compressive strength (MPa)	90	60	18 - 40 [6]

2.2.3 Durability for long term heating

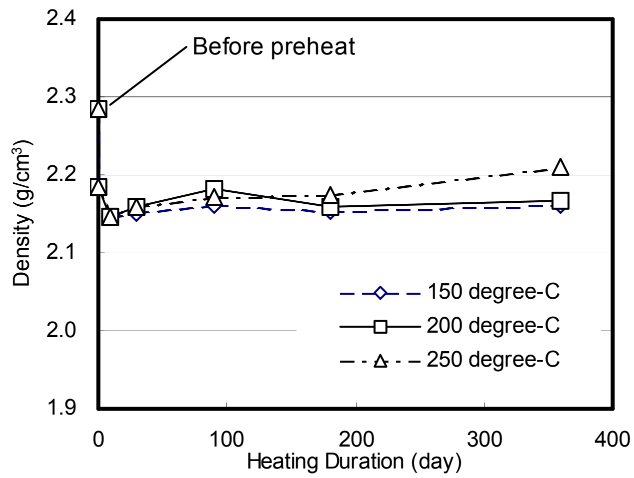
To apply this heat resistant concrete to practical use as shielding material, it is indispensable to verify the characteristics such as decreasing tendency of density and hydrogen (moisture content), etc. during its use. For the neutron shielding material such as resin and rubber practically used for metal cask, the extrapolating method is ordinary used in order to anticipate decreasing rate for more than 40 years by the data of accelerated heating durability test for approximate one year. For this material, the heating durability test for one year was performed and the variation of material characteristics during heated time was confirmed. The test contents and conditions are listed in Table 2. The heating temperature conditions are 150, 200 and 250 degrees Celsius. The samples were pre-heated at 105 degrees Celsius for 48 hours before the long term heating durability test in order to remove the free water. The test was performed with 3 samples for each test condition and the results were the average of them as shown in Figure 3. From the results, the density of each condition was kept with almost constant level after approximate 4% decrease by pre-heated because free water included in the sample was released. The chemically bonded water content was decreased slightly and gradually during heated period but after one year heated, it was verified that water content was converged into approximate 9%. The compressive strength of pre-heated sample was increased 50% compared with that of non-heated sample and then the constant value of approximate 100 MPa was kept during heated period. These tendencies of the results were the almost same regardless of the heating temperatures, then it is confirmed that the materials included in the heat resistant concrete are not influenced by the temperature range from 150 to 250 degrees Celsius.

The shrinkage rate, thermal conductivity, specific heat and coefficient of liner expansion of samples were also measured. The shrinkage rate and thermal conductivity were almost converged into the fixed value after one year heated. The specific heat and coefficient of linear expansion were almost constant during heated period.

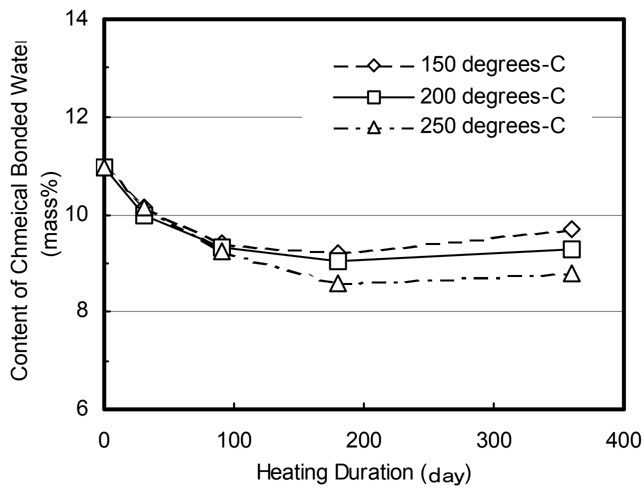
The differential thermal analysis was performed to check whether the chemically bonded water was from calcium hydroxide or not. Only the matrix materials, after removing iron powder and steel fiber using magnet, were analysed since the quantity of sample that could be analysed was very small (about 40 mg). From this test, the chemically bonded water in calcium hydroxide was verified to be kept in this material after the long term heated test with 150 - 250 degrees Celsius. Therefore the neutron shielding ability is not degraded on the temperature condition from 150 to 250 degrees Celsius.

Table 2 Test contents and conditions of long term heating durability

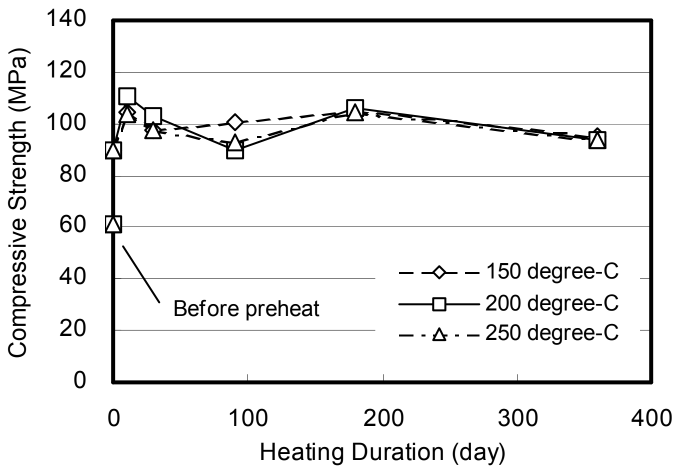
	Test contents and conditions
Measured characteristics	(a) Density (b) Chemically bonded water (c) Compressive strength (d) Shrinkage rate (e) Thermal conductivity (f) Specific heat (g) Coefficient of linear expansion
Conditions	(1) Temperature (degrees-C) : 150 / 200 / 250 (2) Heating duration (days) : 10 / 30 / 90 / 180 / 360 (3) Number of samples for each test conditoin : 3



(a) Density



(b) Chemically bonded water



(c) Compressive strength

Fig.3 Test results of long term heating durability

2.2.4 Additional characteristics

The cut sample of the heat resistant concrete was shown in Photo 1. This material does not include aggregates and steel rod and can be said to be a homogeneous material. In addition, this material contains steel fiber and is excellent for the crack resistance by thermal stress.

Photo1 Cut sample of heat resistant concrete

2.3 Evaluation of shielding ability

The shielding ability of concrete cask using heat resistant concrete was calculated with one dimension discrete ordinary shielding calculation code of the ANISN [10], and compared with the one using conventional concrete. Spent fuels were assumed to be in the condition listed in Table 3 and the concrete cask was modelled as shown in Figure 4. The radiation intensities of gamma ray and neutron were calculated using the ORIGEN2 code [9]. This calculating model was set to be 100 micro Sv/h at 1m from the central surface of the cask in case of using conventional concrete. The atomic densities of concrete materials used for the evaluation were listed in Table 4. In this evaluation, the cross section library was the DLC23/CASK library [11] and the conversion coefficients were calculated from the ICRP Publ.74 [12]. The calculated results were shown in Table 5. From this result, this new material is verified to be excellent for shielding performance especially neutron shielding, and approximate 20% of the total dose rate, especially approximate 50% of neutron dose rate could be reduced compared with conventional material in this calculation.

Table 3 Specification and condition of shielding calculation

	Condition	Remarks
(1) Fuel specification		
- Fuel type	BWR STEP III	
- Initial enrichment (%)	3.5	
- Average burnup (MWD/MTU)	45,000	
- Cooling time (year)	10	
(2) Calculation condition		note* : Result from long term heating tests.
- Number of fuel assemblies	52	
- Density of ordinary concrete (g/cm^3)	2.15 [8]	
- Density of heat resistant concrete (g/cm^3)	2.15 *	

Table 4 Atomic density of concrete material

Element	Heat resistant concrete (atoms/barn cm)	Ordinary concrete [8] (atoms/barn cm)
H	1×10^{-2}	5.34×10^{-3}
C	-	4.11×10^{-2}
O	2×10^{-2}	-
Mg	-	6.13×10^{-5}
Al	-	2.14×10^{-4}
Si	7×10^{-4}	1.78×10^{-2}
Ca	1×10^{-2}	2.22×10^{-3}
Fe	8×10^{-3}	6.35×10^{-4}

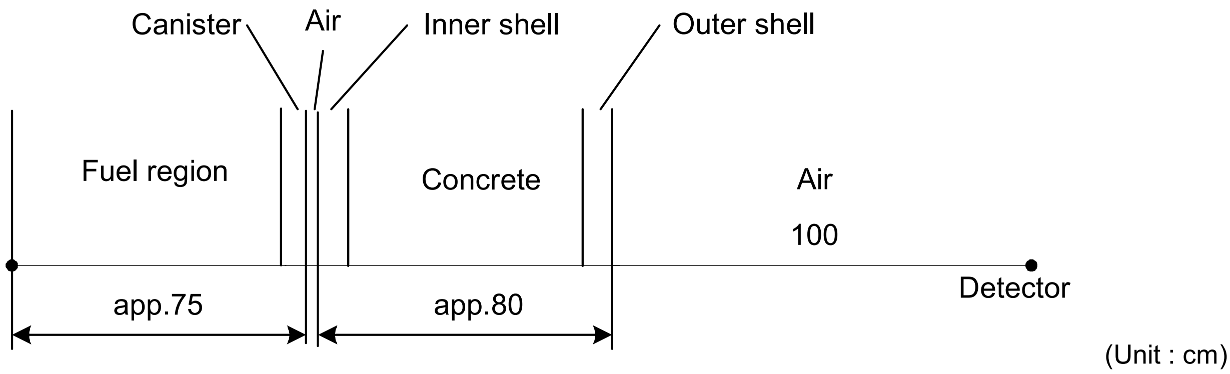


Fig.4 Shielding calculation model

Table 5 Result of shielding calculation

	Dose equivalent rate (micro Sv/h)		
	Gamma	Neutron	Total
Heat resistant concrete	60	20	80
Ordinary concrete	60	40	100

2.4 Evaluation of workability

Generally speaking, as concrete material is supposed to be cast into the case, it is very important to verify the workability of fresh material. In this development program, the casting test using the 1/3 scale model was per formed in order to check the casting workability. About 10 batches of heat resistant concrete material were manufactured, and samples taken from each mixing batch had about 70 cm slump flow then it was verified that this material had excellent flow property. This heat resistant concrete material could be self-filled and no compacting was needed. The photograph of outer view of the cast test model is shown in Photo 2 and the cross section is also shown in Photo 3. This new material was verified to be able to cast into the corner region of copper fin and inner or outer shell from this test result.

In order to check the homogeneity of cast material, the densities taken in random from the cast test material were measured and the result was shown in Table 6. The heterogeneity of cast material is caused by segregation of raw materials and by air included at the time of casting. From the result, the relative standards deviation of the density of cast material was approximate 2.5% and the cast material was almost homogeneous and stable without separation of included raw materials.

Photo 2 Outer view of 1/3 scale model

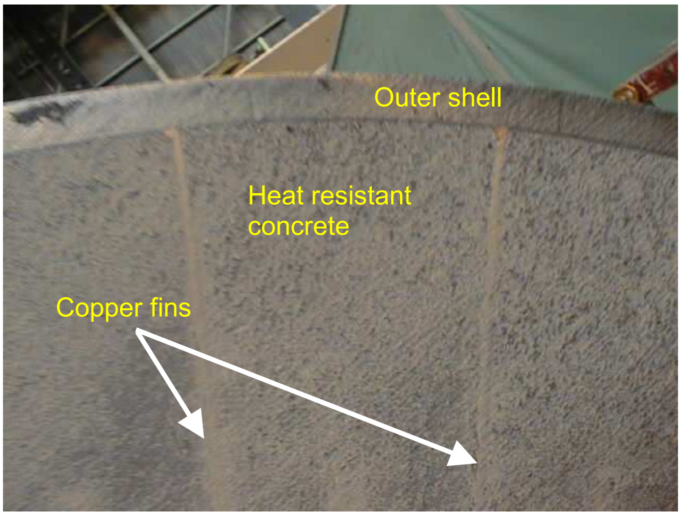


Photo 3 Cross section of 1/3 scale model

Table 6 Dispersion of concrete density

	Measured data
Sampling number	23
Average density (g/cm^3)*	2.4
Relative standard deviation (%)	2.5

Note* : Density data are measured at room temperature without heated.

3. Apply to new concrete cask

A new type concrete cask is shown in Figure 2 and a conventional one in Figure 1. Air inlets and outlets are indispensable to conventional concrete because the temperature of concrete has to be kept lower than 100 degrees Celsius to keep free water with it. But the heat resistant concrete material can contain necessary moisture content (hydrogen) as chemically bonded water effective for neutron shielding even at more than 100 degrees Celsius, therefore the same containment design as metal cask can be possible. The characteristics of this new type concrete cask are followed as below.

3.1 Measures against radiation streaming

It is necessary for conventional concrete cask to take any measure against radiation streaming because air inlets and outlets are lack of the shielding material; for example, the ducts of air inlets and outlets are to be bent. On the other hand, heat removal efficiency should be also needed with taking account for pressure drop of airflow through bent ducts. However this new concrete cask is excellent for shielding performance because it can have the containment structure as same as metal cask since the heat resistant ability of this new concrete is much enough to design this structure.

3.2 Containment monitoring

It is unable to set pressure sensor for containment monitoring on the canister of conventional concrete cask and also unable to set it on the main body of concrete cask since it is opened to outside. However this concrete cask is able to have the containment structure, which makes it possible to monitor air pressure between canister and main body of concrete cask. Therefore excellent containment performance of this concrete cask is ensured.

3.3 Corrosion resistance of canister

In Japan, since interim storage facilities are planned to be located near the sea, the canister installed in conventional concrete cask had possibility to be corroded and stress corrosion cracked by seawater particles including chlorine ion induced from air inlet. For resolving the problem, two-phase stainless steel, which has excellent corrosion resistance and is very expensive, is necessary to be used for canister. However this concrete cask is able to have the containment structure and ordinary austenitic stainless steel can be possible to be used for designing and manufacturing canister, therefore manufacturing cost of canister could be reduced.

4. Conclusion

The heat resistant concrete containing enough water (hydrogen) for neutron shielding even in high temperature of over 100 degrees Celsius was newly developed. Using this heat resistant concrete cask, air inlets and outlets indispensable for conventional concrete cask were not needed and the new concrete cask could be designed and manufactured with the same concept of technology as that of metal cask actually used.

In the next step, the heat resistant concrete material will be improved to have more excellent heat conductivity and higher density and the development of not only storage cask but also transport cask using this material will be proceeded.

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