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Evaluation of properties of soil-based filling material for radiation shielding and water sealing

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ABSTRACT

The 2011 off the Pacific coast of Tohoku earthquake affected Japan, causing severe damage to the Fukushima Daiichi Nuclear Power Station (1F). To decommission 1F reliably, it is necessary to protect workers from radiation exposure and to prevent leakage of contaminated water into the damaged nuclear buildings. The use of a filling type soil material, super heavy bentonite slurry (SHBS), which consists of sodium-bentonite slurry with barite powder, has been examined for water sealing with radiation shielding capabilities. In this study, radiation shielding capabilities test and falling head permeability test were done. This study proved that SHBS has superb performance as a filling-type water stopping material intended for radiation shielding during NPS decommissioning because it has greater gamma ray shielding capability and almost identical neutron beam shielding capability to that of tap water.

Keywords: hydraulic conductivity; radiation; slurry

1 INTRODUCTION

The 2011 off the Pacific coast of Tohoku earthquake caused a severe accident at Fukushima Daiichi Nuclear Power Station (1F) in Japan. Since the accident, it has been a severe difficulty that radiation-contaminated cooling water has subsequently leaked out from some cracks in the damaged nuclear building. According to the Mid-term and Long-Term Roadmap, preventing the diffusion of radioactive substances has required construction of a liquid confinement method under severe radiation conditions in three buildings (Nos. 1–3) that have fuel debris.

Many requirements are placed on materials used to facilitate the decommissioning of 1F promptly and reliably. For using materials in the damaged building, the especially necessary properties are low permeability for trapping contaminated water and radiation shielding capabilities to restrain intense radiation. Historically, in oil well drilling, a principal function performed by the drilling fluid is to form a low-permeability filter cake that seals cracks and other openings. The muddy water features of high density and high water contents are adequate for the materials to shield both gamma rays and neutron beams. Therefore, drilling fluids of some kinds can contribute to providing a steady condition in the damaged nuclear building with water-stopping materials that have radiation shielding capabilities.

Super heavy bentonite slurry (SHBS), the filling type soil material used for this study, has been developed based on drilling fluid (Yoshikawa et al., 2017). Figure 1 presents an image of the damaged nuclear reactor in 1F and the filling material. Table 1 shows some effects of SHBS on filling in the damaged nuclear reactor.

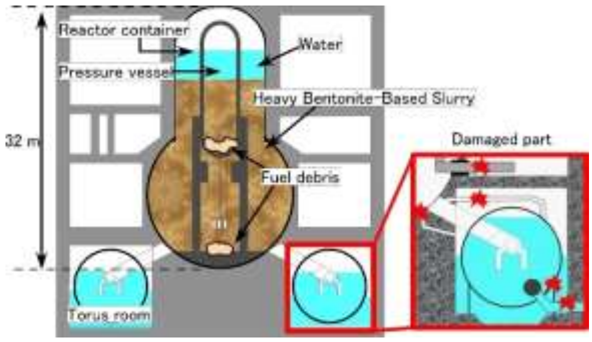

Fig. 1. An image of nuclear reactor buildings and using Super Heavy Bentonite Slurry.

Table 1. Some effects of using Super Heavy Bentonite Slurry.

Characteristics	Use and advantages
Fluidity	Filling construction
	Medium for removal of debris
Viscosity	Prevention of water leakage
High density	Gamma ray shielding
High water content	Neutron beam shielding
	Prevention of scattering of dust

2 FILLING TYPE SOIL MATERIALS ORIGINATED IN DRILLING FLUID

The slurry used for this study is a material obtained by adding barite as a weighting material, with sodium pyrophosphate as a dispersant for maintaining viscosity and stability to sodium-type bentonite suspension. Table 2 presents the SHBS compositions. For comparison, this study used samples with target specific gravity of 1.1, 1.8, and 2.5. In radiation shielding experiments, samples of three types were also added with sodium pentaborate so that the boron concentration in the slurry was 8000 ppm in each formulation in Table 2.

Table 2. Mixing ratio of Super Heavy Bentonite Slurry.

Specific gravity	Tap water (g)	sodium pyrophosphate (g)	Na-bentonite (g)	Barite (g)
2.5	100	0.2	7	400
1.8	100	0.2	10	140
1.1	100	0.2	12	10

3 MEASUREMENT OF SLURRY RADIATION SHIELDING CAPABILITIES

Few experimental works have quantified the radiation shielding capabilities of materials consisting of some materials. In this study, the gamma ray and neutron beam reduction rate of SHBS were quantified.

3.1 Experimental procedure

For this experiment, four acrylic containers filled with materials were prepared and arranged as presented in Fig. 2, whereby the shielding widths of 10 cm–40 cm were measured. Each ray source was placed to be in contact with the center of the radiation transmission surface of the acrylic container. The transmission dose was measured using each detector. Table 3 shows the working source. In this study, the radiation dose reduction rate, R_{rad}, was used as an index to evaluate shielding capabilities of these radiations of SHBS.

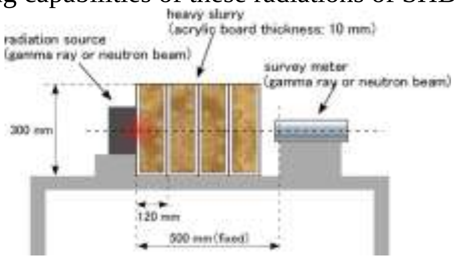

Fig. 2. Experimental setup of radiation shielding capabilities test.

Table 3. Radiation sources used in the experiment.

	Source	Activity (MBq)	Dose (μSv/h)	Energy (MeV)
Gamma rays	^{137}Cs	3.68	1.13	0.662
Neutron beams	^{252}Cf	1.067	5.18	1.406

3.2 Radiation shielding capabilities of SHBS at 10 cm width

To examine the radiation shielding capabilities of SHBS based on the phenomenon mechanism, the results were compared at the minimum width when the difference in reduction rates appeared most significantly in this experiment. Figs. 3–6 show the reduction rates of radiation for 10 cm width.

The gamma rays interact with electrons in contact when transmitting the substance to give some or all the energy to the electrons. The abundance of electrons in the shield, which is important for gamma ray shielding, is sorted out as wet density (Yoshikawa et al., 2017). Figure 3 presents the relation between the gamma ray reduction rate and the wet density for the 10 cm shield width. As depicted in Fig. 3, the gamma ray shielding capabilities possessed by SHBS show a higher reduction rate because the specific gravity is larger: about twice as large as that of tap water.

The moving neutron particles attenuate energy by causing elastic scattering with the nuclei of other substances, but their behaviors differ depending on the movement speeds of the particles. Neutron beams that move at 1/3 to 1/20 of the speed of light in vacuum immediately after being released from the nucleus by nuclear fission are called "fast neutron beams," which have extremely low probability of being trapped by other atoms. Neutron beams in which fast neutrons collide with atomic nuclei in a substance repeatedly and reach a state of thermal equilibrium with surrounding atoms and molecules are called "thermal neutron beams." They have a high probability of neutron capture by substances. Along with the two types above, this study investigated shielding by SHBS for "total neutron beams" including all of these neutron beams.

Because the energy attenuation of neutron beams accords with the law of conservation of momentum, water containing a large amount of hydrogen atoms, which is substantially equivalent in mass to neutron particles, is generally used as the moderating material. Therefore, this study used the volume moisture content as an indicator of water molecule abundance. As portrayed in Fig. 4, SHBS having a large solid fraction ratio showed the same total neutron shielding capabilities as that of tap water in all samples despite the inferior number of contained hydrogen atoms. Fig. 5 and Fig. 6 depict that the fast neutron and the thermal neutron radiation reduction rate increased in direct proportion to the volumetric water content. Moreover, as depicted in Fig. 6, boron-containing materials showed an approximately 70% higher thermal neutron reduction rate than others with the same volume moisture content. This rate is attributable to the fact that the motion velocity of boron-captured neutron particles had decreased to the thermal neutron level.

As described above, SHBS with a high wet density and a volumetric water content of about 50% or more has effective characteristics based on the shielding mechanism. Therefore, high gamma ray and neutron beam shielding capabilities will be exhibited even when the shield thickness can not be secured sufficiently, so that a superior shielding effect can be expected by filling the nuclear reactor building.

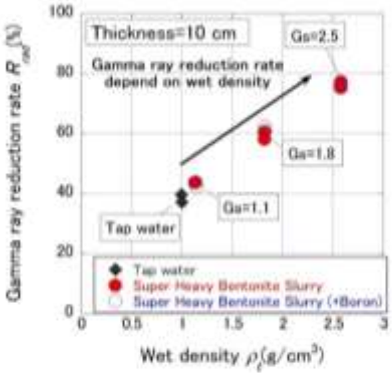

Fig. 3. Relationship between gamma ray reduction rate and wet density of shielding materials.

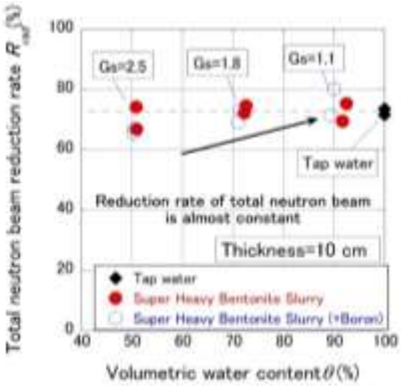

Fig. 4. Relationship between total neutron beam reduction rate and volumetric water content of shielding materials.

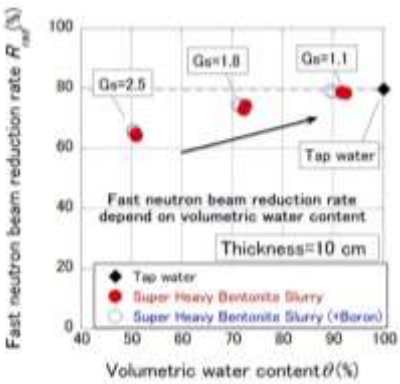

Fig. 5. Relationship between fast neutron beam reduction rate and volumetric water content of shielding materials.

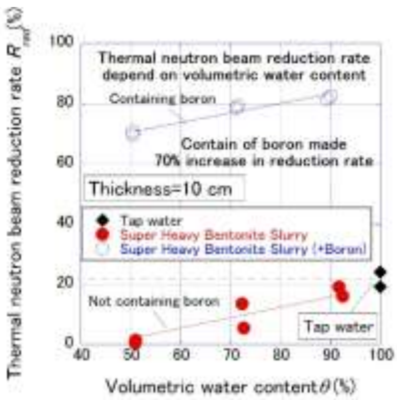

Fig. 6. Relationship between thermal neutron beam reduction rate and volumetric water content of shielding materials.

3.3 Changing the radiation shielding rate with material width

Because the probability that each particle collides with an electron or a hydrogen atom depends on the number of electrons or hydrogen atoms on the movement path of radiation, the shield thickness is an important factor. Figures 7–10 portray the relation between the rate of reduction of radiation and the shield thickness. Furthermore, as presented in Fig. 10, even in the presence or absence of boron, a decrease in the difference because of the increase thickness was confirmed. To shield thermal neutron beams, it is effective to capture neutron particles. However, even when capture does not occur, the direction of motion changes because of collision with nuclei. For that reason, the number of particles reaching the detector decreases as the thickness of the shield increases. Therefore, by providing a sufficient distance on the straight line of the dotted line source, the possibility was shown that the arrival number decreases without showing the neutron capture by boron. Based on the rationale presented above, using materials having high wet density and volumetric water content, such as SHBS, as a shield and securing its sufficient thickness can reduce the risk of radiation exposure to workers and equipment accompanying the decommissioning of reactors, even in intense radiation environments.

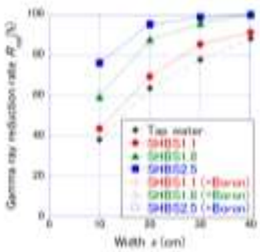

Fig. 7. Increasing of gamma ray reduction rate with material's width.

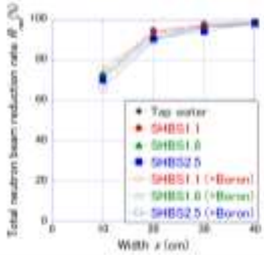

Fig. 8. Increasing of total neutron beam reduction rate with material's width.

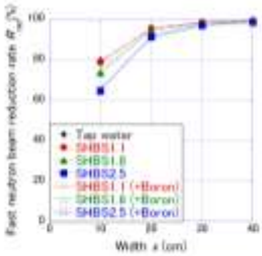

Fig. 9. Increasing of fast neutron beam reduction rate with material's width.

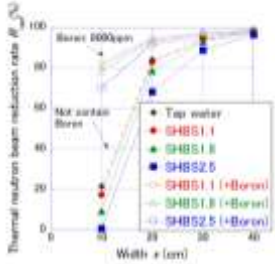

Fig. 10. Increasing of thermal neutron beam reduction rate with material's width.

4 HYDRAULIC CONDUCTIVITY OF THE SLURRY

Following Japanese Industrial Standards A 1218: 2009 "Permeability test of soil", a falling head permeability test using a rigid mold was executed in this study. The slurry volumes changed easily by seepage power (Imai, 1979). Therefore, a transparent acrylic mold was used to measure specimen length L changes with water passage. The initial specimen length, L_0, was 10 cm. After the quantities of inflow water and outflow water were measured, the hydraulic conductivity was calculated using equations (1) and (2) and the apparatus portrayed in Fig. 1.

$$k_T = 2.303\frac{(a_{in} \times a_{out})L}{(a_{in}+a_{out})A(t_2-t_1)}\log_{10}\frac{h_1}{h_2}\times\frac{1}{100} \quad (1)$$

$$k_{15} = k_T \cdot \frac{\eta_T}{\eta_{15}} \quad (2)$$

Therein, k_T (m/s) denotes hydraulic conductivity at T °C, L (cm) signifies the height of the specimen, a_{in} (cm^2) denotes the cross sectional area of the burette on the inflow side, a_{out} (cm^2) expresses the cross sectional area of the burette on the outflow side, A (cm^2) stands for the cross sectional area of the specimen, t_2-t_1 (s) is the measurement time, h_1 (cm) is the water level difference at time t_1, h_2 (cm) represents the water level difference at time t_2, k_{15} stands for hydraulic conductivity at 15 °C, and η_T/η_{15} is the correction coefficient for calculating the permeability coefficient at 15 °C.

Fig. 12 shows the decrease in hydraulic conductivity dependent on the elapsed time. The hydraulic conductivity was defined as the value caused by slurry sedimentation in this study. Sedimentation rates of slurries, $(L_0$-$L)/L_0\times100$, depend on the total quantity of water flow (Fig. 13). The hydraulic conductivity of SHBS dropped gradually into the order of 10^{-10} m/s with elapsed time. Reducing the pore size with development of filter cake is conceivable as a main factor decreasing the hydraulic conductivity over time.

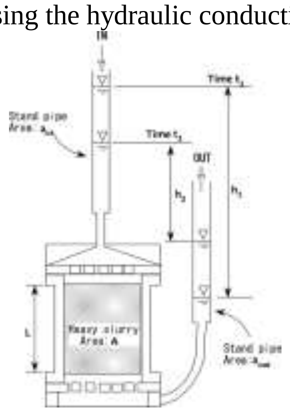

Fig. 11. Apparatus of falling head permeability test.

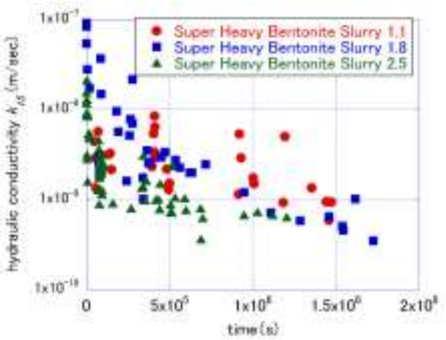

Fig. 12. Hydraulic conductivity changing with elapsed time.

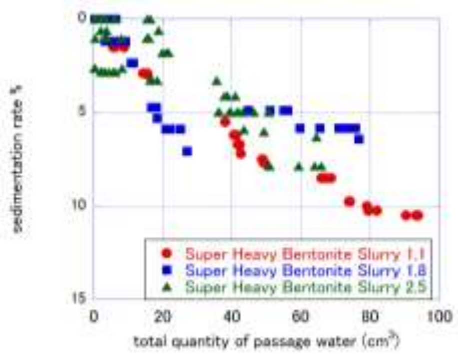

Fig. 13. Sedimentation rates of Super Heavy Bentonite Slurry depend on total quantity of water flow.

5 CONCLUSION

For eventual application to 1F decommissioning, this paper presents a summary of the radiation shielding capability and hydraulic conductivity of SHBS. Results demonstrated that the gamma ray reduction rate is dependent on the wet density if thickness occurs under the same conditions. Similarly, the neutron beam reduction rate depends on the volumetric water contents of materials. This study proved that SHBS has superb properties as a filling material intended for radiation shielding during 1F decommissioning because it has greater gamma ray shielding capability and almost equal neutron beam shielding capability to that of tap water. Also, SHBS has low hydraulic conductivity of 10^{-10} m/s. After these results are incorporated into plans for construction work, they are expected to be useful for recovery from the Fukushima accident.

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