Development of Neutron Shielding Concrete Containing Colemanite and Peridotite

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The present paper describes development of Neutron shielding (NS) concrete and its application to the shield for a neutron spectrometer, SPICA at BL-09 beamline of J-PARC (Japan Proton Accelerator Research Complex). NS concrete is a new concrete using colemanite and peridotite rocks as an aggregate of concrete to build a compact and low-activation shields in the facilities of the high energy particle accelerators, instead of conventional radiation shields composed of normal concrete and some kinds of shielding materials such as a boric acid resin, polyethylene and/or iron. The shielding performance of the present NS concrete was evaluated with the Monte Carlo code PHITS and clarified through the transmission experiments by using $^{252}$Cf spontaneous fission source. Additive mechanical and chemical tests showed better properties than those of the normal concrete. The present concrete was applied to the SPICA shields of a 52 m long neutron beam line and a neutron scattering room. Simplification of the shields and much shield thickness saving was attained comparing with a conventional shielding design. The shields were constructed in September 2011. It was also clarified in calculation that the neutron-induced activities in the NS concrete applied to the shields for a BNCT (Boron Neutron Capture Therapy) facility are several order magnitude lower than those even in the limestone concrete.

KEYWORDS: Radiation Shield, Neutron, Concrete, Colemanite, Peridotite, J-PARC, SPICA, Dose Rate, BNCT Facility, Neutron-Induced Activity.

1. Introduction

Neutron plays an important role as a probe of advanced material science investigating physical properties on microscopic structures and their functions, mineral phase analysis of archaeological remains and medical application as a BNCT (boron neutron capture therapy). Typical facility for investigating physical properties is the MLF (Materials and Life Science Experimental Facility) of J-PARC (Japan Proton Accelerator Research Complex) [1]. In civil construction, many collapse accidents of huge infrastructure such as building, bridge, tunnel and roads have been reported in the world. Neutron is also applicable to detect a structural failure due to degradation of material in the infrastructure. Recently, many compact accelerators are developed for the purposes of physical property studies[2], BNCT[3] and nondestructive measurement to detect water/void inside slabs by backscattered neutrons[4]. In these facilities, radiation shields are built to protect people and equipment from radiation exposure.
Recently, compact shields are desired to keep a wide experimental area of neutron scattering facility even in MLF, of which 22 neutron-beam-lines have already occupied among 23 ones[5]. The compact proton accelerator facilities will be generally constructed in the local room area of the university and laboratory, and used by carrying it to bridge, tunnel and road. In order to keep the space of working area, compact shields are needed.

The present paper describes development of neutron shield (NS) concrete by using a colemanite and peridotite rocks as an aggregate of concrete to build a compact shield additively having a property of low material-activation, and its typical application to the SPICA spectrometer of J-PARC for special environment measurements on battery research are described together with analysis of induced activities of materials in the shield for a BNCT facility.

2. Neutron Shield (NS) Concrete

2.1 Development of the NS concrete

Concrete is usually used for neutron shielding, since it can be easily made any shape and has moderate neutron-shielding performance. Although it is inexpensive, massive thickness is required. Thus, development of the inexpensive concrete having better shielding performance and the same mechanical properties as normal concrete was a long-time dream. Boron has a property of high neutron capture and mixture of boron and hydrogen very effective to slim shields. They are contained in natural rock, and it is better to use natural rock as aggregate of concrete from the viewpoints of cost and demand for construction. However, most of past works to develop the concrete of colemanite that is a natural rock containing boron reported that a hardening delay occurred. The reason is that the colemanite slightly dissolved in water and did not have uniform mechanical strength with keeping a rigid shape. The authors found[6] through the neutron transport calculations that a few tens per cent of colemanite was enough to give a good shielding performance. Such small amount of colemanite used as a coarse aggregate with portland cement resulted in forming a hard mass of plate[7]. Serpentinite being hydrogen-rich with iron, which were effective on neutron slowing-down, was used for concrete in the past, and the typical example was the pedestal concrete shield of the fast breeder demonstration reactor Monju[8]. Since the serpentinite contains harmful asbestos, the authors selected similar hydrogen-rich peridotite instead of serpentinite in order to develop the NS concrete: it has been developed with colemanite, peridotite, and ordinary Portland cement by controlling the content of the colemanite up to around 15 wt% of the concrete composition aiming at improved shielding performance and production of the concrete. The density of neutron shield concrete is the same as that of normal concrete; 2.2 g/cm³.

2.2 Shielding property of NS concrete

Basic shielding property of the NS concrete and other shielding materials used for the shields of neutron spectrometers was calculated for a cylindrical geometry with a point source at the bottom with the particle transport code MCNP5[9] and the JENDL-3.2-base library[10]. Table I shows the atomic composition of the NS concrete. The calculated results are shown at various incident neutron energies as a function of shield thickness in Figure 1. Table II shows that the NS concrete mostly occupies the
second position concerning neutron shielding performance except for thermal energy that B$_4$C and boric acid resin are superior to the NS concrete. Iron shield is effective in high energy region due to large removal neutron cross section of inelastic scattering and (n, 2n) reactions etc. above 2 MeV but not beyond 40 cm because of window effect of moderated neutrons at cross section minima in the resonance energy region. Typical example is shown in case of 30 keV neutron source. There is a large s-wave resonance at 27.7 keV which is very effective to neutron shield but followed by a cross section minimum at 24 keV giving window effect inducing deep penetration in iron.

Since the NS concrete absorbs neutrons well, secondary gamma-rays are quite lower

| H   | B10        | B11 | O    | Na   |
|-----|------------|-----|------|------|
| 2.28E-02 | 2.80E-04  | 1.14E-03 | 4.18E-02 | 2.99E-05 |
| Mg  | Al         | Si  | P    | S+Cl |
| 9.04E-03 | 6.03E-04  | 6.92E-03 | 1.87E-05 | 5.87E-05 |
| K   | Ca         | Ti  | Mn   | Fe   |
| 2.81E-05 | 2.84E-03  | 1.99E-05 | 1.87E-05 | 9.18E-04 |

Table I. Atomic compositions of the NS concrete. (unit: atoms/barn*cm)

| Source Energy | 1st position | 2nd position | 3rd position | Others |
|---------------|--------------|--------------|--------------|--------|
| 20 MeV        | Iron         | NS concrete  | Polyethylene | Normal concrete (up to 50cm) |
| 2.45 MeV      | Polyethylene | NS concrete  | Iron (up to 50cm) | Normal concrete (up to 70cm) |
| 1 MeV         | Polyethylene | NS concrete  | Normal concrete | Iron |
| 100 keV       | Polyethylene | NS concrete  | Normal concrete | Iron |
| 30 keV        | Polyethylene | Iron (up to 10 cm) | NS concrete | Normal concrete |
|               |              | NS concrete  | Normal concrete | Iron (beyond 15 cm) |
| 1 keV         | Polyethylene | NS concrete  | Iron          | Normal concrete |
| Thermal       | B$_4$C       | Boric acid resin | NS concrete | Iron, Polyethylene |

Table II. Order of neutron shielding performance of shielding materials.

Fig. 1. Comparison of neutron transmission through various shielding materials.
in the NS concrete than in iron, polyethylene and normal concrete. Figure 2 shows a comparison of total and secondary gamma-ray dose rates between the normal and the NS concrete. In case of 10 keV neutron source, the secondary gamma-ray dose rate of the NS shield is about 1.5 order of magnitude lower than that of the normal concrete beyond 40 cm thickness and total dose rate beyond 20 cm thickness is mostly contributed by the secondary gamma-ray.

It is also noticed that the present shielding performance was clarified through the transmission experiments and its analyses for the NS concrete slabs up to 100 cm thickness performed at the laboratory of Hazama-Ando Corporation by using $^{252}$Cf neutron source: the shielding performance of the NS concrete was about 1.7 times with the one hundredth attenuation length of the normal concrete[7].

2.3 Mechanical properties concrete

The mechanical and chemical property tests for the standard test piece of 10 cm in diameter and 20 cm in length were also carried out [7]. The results showed that the compressive strength of the NS concrete up to 13 weeks after placing the concrete was equivalent to that of the normal concrete. The drying shrinkage test proved 8% - 10% smaller than the normal concrete.

Concrete is basically alkaline which protects reinforcement from corrosion. The concrete carbonation by atmospheric CO$_2$ reduces its alkalinity (i.e., neutralization of concrete) and often induces reinforcement corrosions. The carbonation test was made under the condition of different water/cement ratios which influences the mechanical strength of concrete. The test result measuring depth of neutralized layer from the concrete surface given in Table III showed that the neutralized layer depth of NS concrete was smaller than normal concrete: it means that carbonation speed of the NS concrete is lower than that of the normal concrete.

| Water/Cement Ratio | Depth of neutralized layer (mm)  |
|-------------------|----------------------------------|
|                   | 50%                              | 60%                              |
| Normal concrete   | 3.0                              | 4.5                              |
| NS concrete       | 2.0                              | 3.5                              |
concrete is slower than the normal concrete.

3. Application to SPICA shields

The present NS concrete has been used for 4 neutron spectrums such as NOVA[7], SPICA[11], TAIKAN[12] and Super HPRD of the MLF of J-PARC. The followings are a description on SPICA shield design and construction as an example of the NS concrete application.

3.1 Shield design

SPICA is a large neutron spectrometer. Its flight path from moderator to a sample position is 52 m, the second longest one in MLF. Central axis of the neutron beam line was located at 1773 mm height. SPICA has three detector banks that are the backward detector bank, the multipurpose detector bank and the forward (small angle) detector bank. The distance from the sample to detectors is about 2 m for all banks. Thus, the scattering room was taken to be the largest in MLF: approximately 9 m in length, 6 m in width and 3.5 m in height. Radiation shield design for SPICA has been performed by using the PHITS code [13] as following two steps: (1) designs aiming at optimized multilayer shields using conventional shielding materials of iron, normal concrete, and boric acid resin which were generally used for neutron-spectrometer shields and (2) advanced design of novel monolayer shields using the NS concrete aiming at the mostly same results as the first design. Figure 3 shows calculation models of the first designs with the multilayers. In the advanced design, most multilayers were changed into

![Calculation model of the first designs with the multilayers using conventional shielding materials and model description for the advanced design.](image)
monolayers of the NS concrete except for the normal concrete blocks located in the neutron beamline to shield streaming neutrons. Thus, the SPICA shields can be mentioned to be composed of only concrete.

Shield thickness was determined so as to satisfy the design criterion of radiation dose rates both outside of the biological shield and at the boundary of the radiation controlled area. Since the SPICA NBL is straight, polyethylene which was often applied to the NBL shields of the other neutron spectrometers of MLF was initially considered as the most inner shield to reduce influence of streaming neutrons. However, the calculation indicated that the lower concrete blocks partially carrying a saddle-shaped iron shield, which were set under the guide tube between guide-tube supporters, effectively reduced so much neutron streaming that polyethylene was substituted with normal concrete as shown in Figure 3(a). Figure 3(b) shows that shield thickness was reduced in stepwise as a distance from the neutron moderator. Shield designs for the neutron scattering room were made to reduce radiation dose rates outside the shields and the neutron background in the detector bank as shown in Figure 3(c). Main shield was determined to be multilayer composed of 500 mm or 600 mm thick concrete. An inner liner of 50 mm thick boric acid resin was used together with the shield blocks located along the NBL in order to reduce background neutrons in the room. Beam dump was composed of 100 mm thick boric acid resin, 100 mm thick iron and 300 mm thick concrete surrounding 300 mm x 300 mm beam hole.

Further advanced design was performed to change the multilayer shields into monolayer shields was made by using NS concrete and ascertaining that radiation dose rates become lower than those of multilayer shields. In the NBL shields, all shielding materials were substituted with the NS concrete except for the lower concrete blocks against neutron streaming from the neutron moderator to the neutron scattering room. Thus, volume size of the shields was unchanged. Main shields for the neutron scattering room were modified into thinner NS concrete shield by removing the boric acid resin lining and resulted that the room became extended. Backward shield of the beam dump was also reduced by 200 mm by using Eponite®[14] which was mixture of boron carbide with a phenol-based resin instead of boric acid resin. Special care was paid to the local spaces of disk choppers shown in Figure 3(d) in order to suppress neutron streaming by using a partial shield of polyethylene. Table IV shows shield saving or simplification due to design change from multi-layers of different shielding materials to

| Places                     | Conventional Design                                      | Optimized Design                                      | Improving       |
|----------------------------|----------------------------------------------------------|-------------------------------------------------------|-----------------|
| NBL Shields around Disk Choppers | Multi-layer shields of Normal Conc., Iron and Polyethylene | 760 – 910 mm NS Concrete                               | Simplification  |
| Main Shields for Spectrometer | 50 mm Boric Acid Resin Lining + 500 mm or 600 mm Normal Concrete | 500 mm or 600 mm NS Concrete                           | 50mm save       |
| Entrance to Spectrometer   | 50 mm Boric Acid Resin Lining + 600 mm Normal Concrete   | 400 mm NS Concrete                                    | 250mm save      |
| Around NBL in Main Shields | 100 mm Normal Concrete+200mm Iron+50 mm Boric Acid Resin | 350 mm NS Concrete                                    | Simplification  |
| Backward of Beam Dump      | 200 mm Boric acid resin + 400mm Iron+1100 mm Normal Concrete | 200 mm Eponite® +200 mm Iron +1100 mm NS Conc.        | 200mm save      |
| Hatched Door               | 50 mm Boric Acid Resin + 600 mm Normal Concrete          | 550 mm NS Concrete                                    | 100mm save      |
mono-layer shields of NS concrete.

Finally, radiation dose rates were calculated for the region including neighboring beamlines BL08 and BL10. Calculated radiation dose rates satisfy the design criterion and seem to have some kind of margins around the NBL shields. Neutron spectra in the detector bank also calculated to clarify that the present monolayer shield reduce background neutrons as much as boric acid resin. Figure 4 compares the calculated total dose rate distributions between the first design and the advanced one. Figure 4(a) shows some mountain of springing dose are observed around the step altering shield thickness in the first design and diminished in the advanced one. Figure 4(b) shows that the advanced design succeeded to decrease neutron streaming through gaps in the disk.

Fig. 4. Comparison of the dose rate distributions between the first design and the advanced one.

Fig. 5. Calculated radiation dose rate distributions around the SPICA shields[11].
copper room at the distance of 13m from the moderator. Figure 4(c) shows nearly the same result even though the advanced design adopted thinner shields of the NS concrete compared to the two-layers shields of borated resin and normal concrete in the first design.

Thus, the calculation with the model considering effect of radiation from the neighboring neutron beam lines gave sufficient shielding performance as shown in Figure 5. It was also found that the NS concrete exhibits the same spectrum as the 10 cm boric acid resin and 10 cm B$_4$C liner which are strong absorbers of thermal neutron [11]. It can be mentioned that the NS concrete realize a simple monolayer shield, compared to the conventional multi-layer shields.

3.2 Fabrication and Construction of the SPICA Shield

The iron shields were fabricated by Mitui Engineering & Ship Building Co. Ltd. and

![Fig. 6](image1.jpg)

(a) Size check  (b) Placing concrete  (c) Fabricated concrete block

**Fig. 6** Photos of fabrication of the shield blocks.

![Fig. 7](image2.jpg)

(a) Side view of the NBL shield  (b) Top view of the NBL shield

(c) Main shield and beam dump  (d) Hatch door

**Fig. 7** Photos of the SPICA NBL and main shields construction.
the shields of the NS concrete were by P.S. Concrete Co. Ltd. based on the contract with Hazama-Ando Corporation which did the overall construction. Figure 6 shows photos of fabrication of the concrete shields from size check of the iron reinforce to concrete blocks. Finally, the shields have been constructed at the 9th NBL of the MLF in 31 October in 2011 by Hazama-Ando Corporation as shown in Figure 7.

4. Neutron Induced Activities of Concrete

The shielding performance of the NS concrete occupy the second position in the allover energy regions as described in Section 2; it means that the neutron fluxes in the NS concrete are very low in comparison with that in the other shielding materials. On the other hand, since radioactivities of the concrete will become of problem to make a decommissioning of facility as well as preservation, neutron induced activities of the concrete at that time are expected to be very low. The activation of concrete is not induced by main elements of concrete composition which have low activation cross sections but done by impurity elements of which contents are not identified in general. Thus, the neutron activation experiments for different concretes in the same neutron field of JRR-4 to certify the contents of impurity elements of the concretes and an activation estimation for 3 kinds of concrete wall in the accelerator neutron source facility for BNCT was made as an example of application[15].

4.1 Comparison of neutron induced activities irradiated in the same neutron field

The authors performed the neutron activation experiments of concrete at the fourth research reactor of JAERI, JRR-4. JRR-4 is a swimming pool-type reactor with low-enriched uranium fuels, moderated and cooled with light water. The reactor core is established in “a core tank” in the reactor pool as shown in Figure 8. The maximum thermal power of JRR-4 is 3,500 kW. Maximum thermal neutron flux is $7 \times 10^{13}$ n/cm$^2$/s. The reactor power, operating time, and pattern are available to meet the requirements set by users. The reactor was used with five irradiation ports for the shielding mock-up experiment, Boron Neutron Capture Therapy, activation analysis, production of semiconductor silicon, and education and training for engineers and researchers in various fields.

In the present work, the samples used in the experiment were the neutron shield concrete, peridotite concrete and limestone concrete which is well known as a low activation concrete. The concrete sample was crushed until the particle diameter was 1

| Activated Nuclides | Activity (Bq/g) | NS Concrete | Peridotite Concrete | Limestone Concrete |
|--------------------|----------------|-------------|---------------------|-------------------|
| $^{51}$Cr          |                | 3077.5 ± 43.1 (1.0) | 6543 ± 140.5 (2.1) | 878.5 ± 14.5 (0.29) |
| $^{59}$Fe          |                | 399.5 ± 3.6 (1.0)  | 617 ± 5.5 (1.5)  | 268 ± 3.5 (0.67)  |
| $^{60}$Co          |                | 270.5 ± 2.4 (1.0)  | 444 ± 2 (1.6)    | 65.0 ± 1.2 (0.24) |
N.B. The value in a parenthesis is a ratio of activity to the NS concrete.

mm and 100 mg of the sample was enclosed in a polyethylene bag, which was then packed in a polyethylene capsule. The capsule was sent to the irradiation Pn port in a reactor core by pneumatic tube and was irradiated for 120 s; irradiation neutron flux was estimated $10^{15}$ n/cm$^2$. The dose of the irradiated sample was immediately measured by a GM survey meter without opening the capsule. Then, after cooling, the gamma spectrum of the sample was measured by a germanium semiconductor detector. A quantitative analysis was made to identify radioactive nuclide and its activity and to derive atomic compositions of the impurity elements. A part of the results is given at Table V. Activities of different concretes are in agreement within a factor of 7, i.e, the ratio of maximum to minimum of activity is about 7, while the activities of the limestone concrete is the smallest.

4.2 Calculation of activities in BNCT facility

The activation characteristics in concrete walls formed from neutron shield concrete, limestone concrete, and peridotite concrete were compared to each other in a BNCT facility of 5.3m L x 5.0m W x 7.3 m H irradiation room with 1.8 m thick concrete shields followed by a proton accelerator. Neutron fluxes were calculated with the PHIT code. To evaluate the depth distribution of the activation amount in the wall in the beam dump side, the volume of the wall was subdivided into slices parallel to the wall surface, each of 10-cm thickness. Although the accelerator neutron source has a suitable moderator that prepares epithermal neutrons from the neutrons that are generated from a Be or Li target, the 10 keV source was assumed in the calculation. Then, neutron induced activities were calculated by DCHAIN code[16] from reaction rate between the neutron and medium elements in individual slices of the walls.

The calculated result of neutron spectrum in the wall showed that slow neutrons below 0.6 eV of the NS concrete is lower by about two orders of magnitude than the others. The $^{60}$Co activities in the NS concrete decreases the most promptly with a depth from the surface as shown in Figure 9. The neutron induced activities for short half life nuclides between the limestone and the NS concrete are also compared in Table VI.

![Graph showing spatial distribution of $^{60}$Co activities in wall A](image)

Fig. 9 Spatial Distribution of $^{60}$Co Activities in wall A[15].
Table VI  Produced Quantities of Short-Half-Life Nuclides in Floor and Wall A. (unit: Bq/g)

| Position       | Depth  | $^{28}$Al | $^{24}$Na | $^{56}$Mn |
|----------------|--------|-----------|-----------|-----------|
| NS concrete    | Floor  | 0 to 10cm | 0.017 (1.0)| 0.002 (1.0)| 0.020 (1.0) |
|                | Wall A | 0 to 10cm | 0.009 (1.0)| 0.001 (1.0)| 0.010 (1.0) |
| Limestone conc.| Floor  | 0 to 10cm | 1.59 (93)  | 0.94 (410) | 0.38 (19)   |
|                | Wall A | 0 to 10cm | 1.52 (77)  | 0.89 (810) | 0.35 (35)   |

N.B. The value in a parenthesis is a ratio of the amount of activity in wall A to that of NS concrete.

From these figure and table, it can be mentioned that the NS concrete has several orders of magnitude lower activities than the other the concretes, even the limestone concrete, because neutron attenuation is very large in the NS concrete.

5. Conclusion

Neutron shielding (NS) concrete using a colemanite and peridotite rocks as an aggregate of concrete has been developed to build a compact and low-activation shields in the facilities of the high energy particle accelerators, instead of conventional radiation shields composed of normal concrete and some kinds of shielding materials such as a boric acid resin, polyethylene and/or iron. It has been found that the NS concrete has a good shielding performance in the whole neutron energy range from thermal to a couple of tens MeV. As a result, NS concrete has realized thinner monolayer shields for long NBL and large scattering room of a neutron powder diffractometer SPICA of J-PARC. The SPICA shields were fabricated well and have been constructed on 31th November 2011.

The shield simplification to the concrete also enables us to built-and-disassemble the shields quite easily and hence might reduce construction cost. Other merit of the neutron shield concrete is to reduce a neutron-induced activities, since it effectively diminish thermal neutrons. The activation experiment clarified the neutron induced activities of various concretes and quantities of impurity elements contained in concrete. The example calculations for BNCT facility showed larger reduction of several order of magnitude in neutron induced activities in the wall and floor of the NS concrete in comparison with limestone concrete, well known as a low activation concrete.

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