Turkish Journal Of Nuclear Sciences, 9 (1982) 64-71

SULFUR COATED COLEMANITE AS SHIELDING MATERIAL

G. GÜNDÜZ

Middle East Technical University,
Department of Chemical Engineering, Ankara

ABSTRACT

Sulfur coated colemanite particles were used as coarse aggregate in concrete. It was found that the boron content of concrete could be increased to a value about four times larger than those reached in the past. As the amount of sulfur coated colemanite increases in the concrete, the density and the compressive strength decrease, but the total water content increases. The fast neutron flux slowly decreases with the increased colemanite, while the thermal flux is lowered in great extent. The total radiation dose is not considerably increased even though the colemanite content is increased twice that of the ones used in the past.

ÖZET

Kükürt kaplı kolemanit tanecikleri betonda iri kum olarak kullanıldı. Böylece betona konulabilen borun geçmişte erişilen değerlerin dört katına çıktığı bulundu. Kükürtlü kolemanit miktarı arttıkça betonun yoğunluğu ve basma dayanımı düşmekte, toplam su miktarı ise artmaktadır. Artan kolemanit miktarıyla hızlı nötron akısı yavaşça azalmakta, yavaş nötron akısı ise önemli miktarda düşmektedir. Toplam ışın dozu ise geçmişte kullanılan kolemanitin iki katına çıkılsa bile fazla artmamaktadır.

INTRODUCTION

The concrete shield of nuclear reactors usually contain baryte ($BaSO_4$) to attenuate gamma radiation and some boron compounds to absorb thermal neutrons. Fast neutrons can be thermalized by introducing small atomic weight compounds. This is usually done by increasing the water content of concrete. Magnesium oxy chloride added to the mortar can increase the water content of the cement phase twice. But it can be added

only in small quantities, otherwise it causes cracks /1/. The water content of the aggregate phase can be increased by using proper minerals such as limonite and serpentine /1-3/. Another way of introducing hydrogenous compounds is the polymer impregnation of set concrete /4/. About 4% polymer can be impregnated into baryte concrete.

Colemanite and borocalcite are two boron minerals added to concrete to absorb thermal neutrons. But they have deleterious effects on the setting of cement. In order to decrease the solubility of these compounds they are added to the coarse aggregate with a size larger than 20 or 30 mesh. The total amount added usually can not exceed 4-5%. Instead of boron minerals, boron frits can also be added. They are usually made from the fusion of boric acid, limestone, silicate and aluminum oxide. Frits also can be added in small quantities, about 5-6%. But it is also possible to add small amounts of frit to the cement phase /4-5/. It is possible to increase the amount of frit in concrete by first coating the frit particles by a thin layer of polymer and then using in mortar. This way the boron content of concrete can be increased by about 4 times /4/. But since all frits are prepared at high temperatures, they do not contain any crystalline water. But it is better if one can increase the amount of borocalcite ($CaB_4O_7 \cdot 4H_2O$) or colemanite ($Ca_2B_6O_{11} \cdot 5H_2O$) in concrete because they both contain large amounts of crystalline water to thermalize fast neutrons. The dissolution of these compounds can be minimized by coating the aggregate particles by a film.

EXPERIMENTAL

A preliminary experiment was carried out to coat colemanite particles by a copolymer (styrene 70 %, acrylonitrite 30 %) used in coating colemanite-baryte frit in the past /4/. But it did not work in this case. Because colemanite-baryte frit is quite porous and can easily absorb the monomer. Colemanite particles do not absorb the liquid monomer, so they can not be effectively coated. Therefore in this research colemanite particles were coated with sulfur which melts at around 393 K. The amount of sulfur in coated particles was 18.3 %. The coated particles were sieved from a set of sieves. The particles were then blended in proper ratios to meet the standards. The sulfur coated colemanite (SCC) particles were then added to the coarse aggregate. 75 % of the total aggregate was coarse (between 3.5-22 meshes) and 25 % was fine (below

22 mesh). The coarse aggregate contained both baryte and different proportions of SCC while the fine aggregate contained only baryte.

The samples were molded in cylinders, 5 cm in diameter, and 10 cm in height. For the concrete sample containing no SCC, the baryte/cement ratio was taken 6/1, and for those containing SCC, the ratio changed depending on the density of the aggregate. The water/cement ratio was 0.6. The molded samples were cured in a humid room for 28 days.

In the graphs the boron content is expressed in terms of boron oxide (B_2O_3) since the analysis of cement or ordinary concrete is usually reported in terms of oxides.

RESULTS AND DISCUSSION

Physical Properties:

The maximum amount of SCC used in the coarse aggregate was 50 %. Even though it was possible to increase this quantity further, it causes a decrease in density as seen in Fig. 1 and also a decrease in the barium content of concrete.

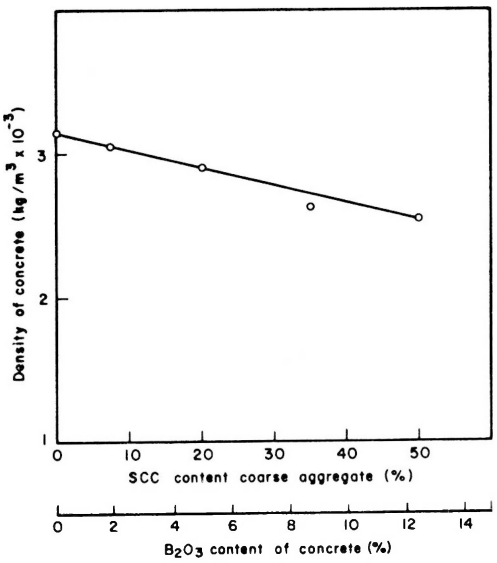


Fig. 1. Change of density.

Consequently the attenuation of gamma rays is weakened. A 50 % SCC in coarse aggregate denotes to a B_2O_3 content more than 12 % in concrete. This value is about four times more than those reached in the past. A similar value was obtained when polymer coated frit was used /4/. But it is possible to increase this value further by increasing the SCC content.

The water content in cement phase slightly decreases as seen from Fig. 2 even though cement/aggregate ratio slightly increases. This should be due to deleterious effects of colemanite on the setting of cement, such that sufficient water molecules can not bind to cement.

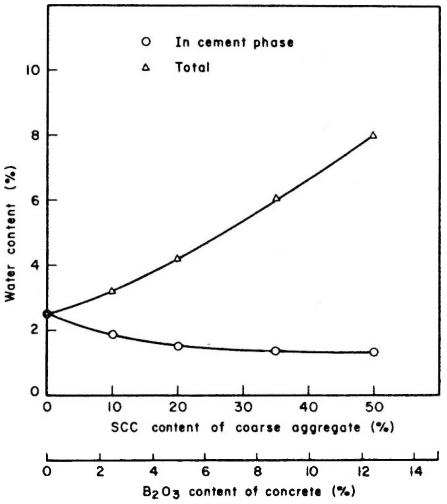


Fig. 2. Change of water content.

Consequently the strength of cement is badly influenced as seen from Fig. 3. Nevertheless the total water content of concrete increases with SCC as crystalline water is introduced with colemanite. This can be seen from the upper curve in Fig. 2.

Shielding Properties:

The shielding properties of concretes were determined through a computer. The neutron flux distributions fed in the program are typical of the flux distributions in a thermal reactor.

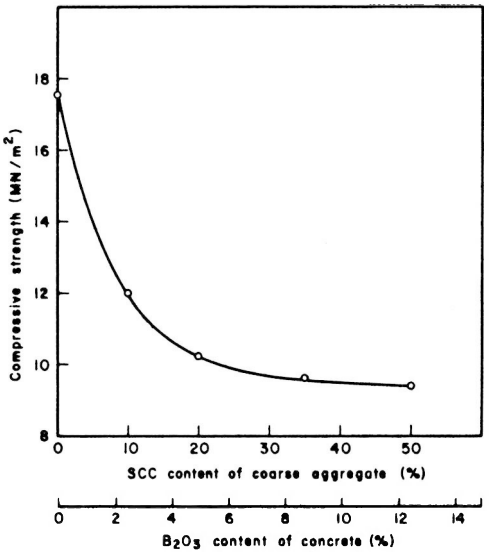


Fig. 3. Change of compressive strength.

The fast flux distributions are shown in Fig. 4. The fast neutron flux is lowest in 50 % SCC case, but the decrease is not very pronounced.

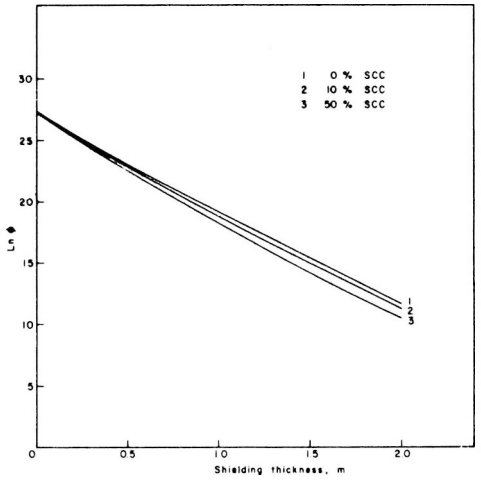


Fig. 4. Fast neutron flux (above 0.18 MeV).

Fig. 5 shows the thermal flux distributions. An increase in SCC content naturally decreases the thermal flux in great extent. This effect is more pronounced as one goes from 0 to 10 %.

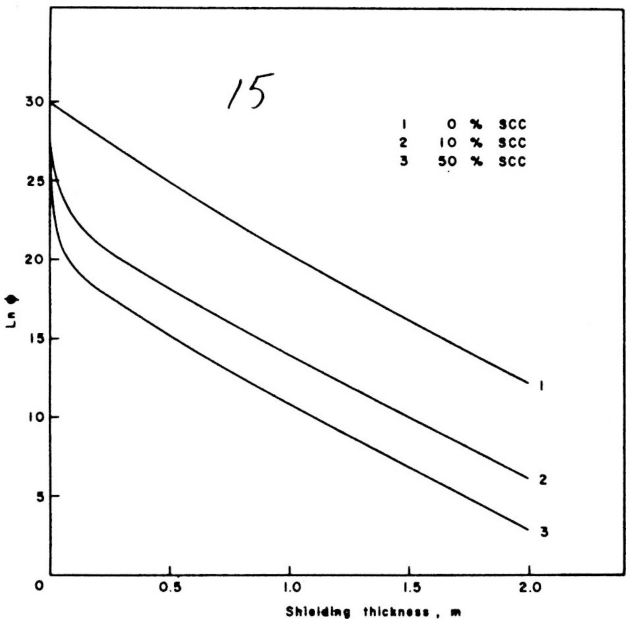


Fig. 5. Thermal neutron flux (below 0.41 eV).

The change of total radiation dose including both neutrons and gamma rays is shown in Fig. 6. The total dose has the lowest value in 10 % SCC and the largest value in 50 % SCC case. Large increases in SCC lowers the density, and reduces the amount of barium which is quite effective in attenuating the gamma radiation due to its relatively larger atomic number. The difference between 10 and 20 % SCC cases is very small. 10 % SCC content in coarse aggregate denotes to 5.12% colemanite in the concrete. This is the amount used in the past. By using 20 % SCC in coarse aggregate, the total amount of colemanite in concrete is increased twice compared to the ones used in the past. Since there is only a very slight difference in the total radiation dose one should prefer 20 % SCC case because fast and thermal neutrons are captured in larger extents. 50 % SCC concrete should then be used where neutron shielding is of primary importance.

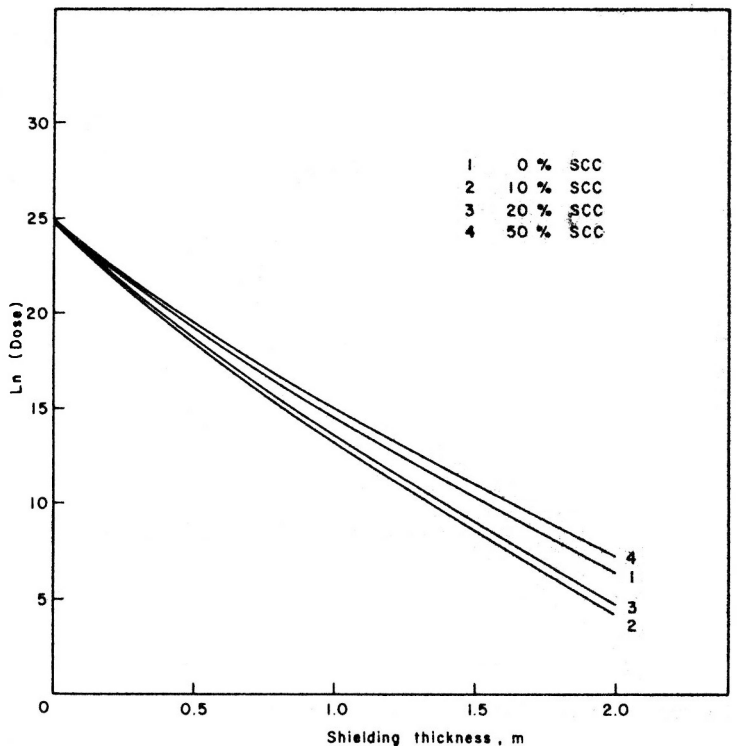


Fig. 6. Total radiation dose (mrem/hr).

ACKNOWLEDGEMENT

The author appreciates the help of Miss E. Boyacıoğlu in the preparation of aggregate.

REFERENCES

/1/ "Concrete For Radiation Shielding", American Concrete Institute, 1962.

/2/ Davis, H.S., Concrete for Nuclear Reactors (Proc. Symp. Berlin, 1970) 3, American Concrete Institute (1972) 1195.
/3/ Höniz, A., Vesely, F., Samalikova, M., Stoces, B., and Havranek, M., ibid., p. 1225.
/4/ Gündüz, G., Thesis, METU, Ankara (1980).
/5/ Çakaloz, T., Gündüz, G., Sunar, S., 7. Scientific Congress. Turkish Scientific and Technical Research Council, (1980) Kuşadası, İzmir (to be published) (in Turkish).