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METHOD OF MAKING A NEUTRON-ABSORBING CALCIUM BORIDE BRICK

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FIG. 1

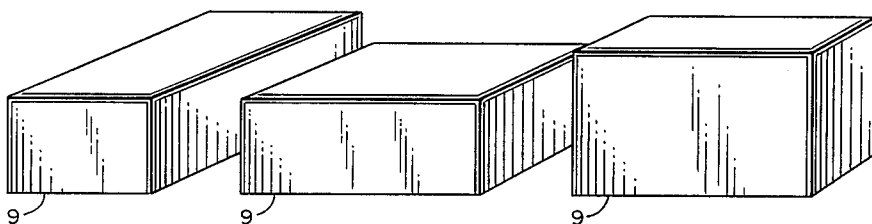
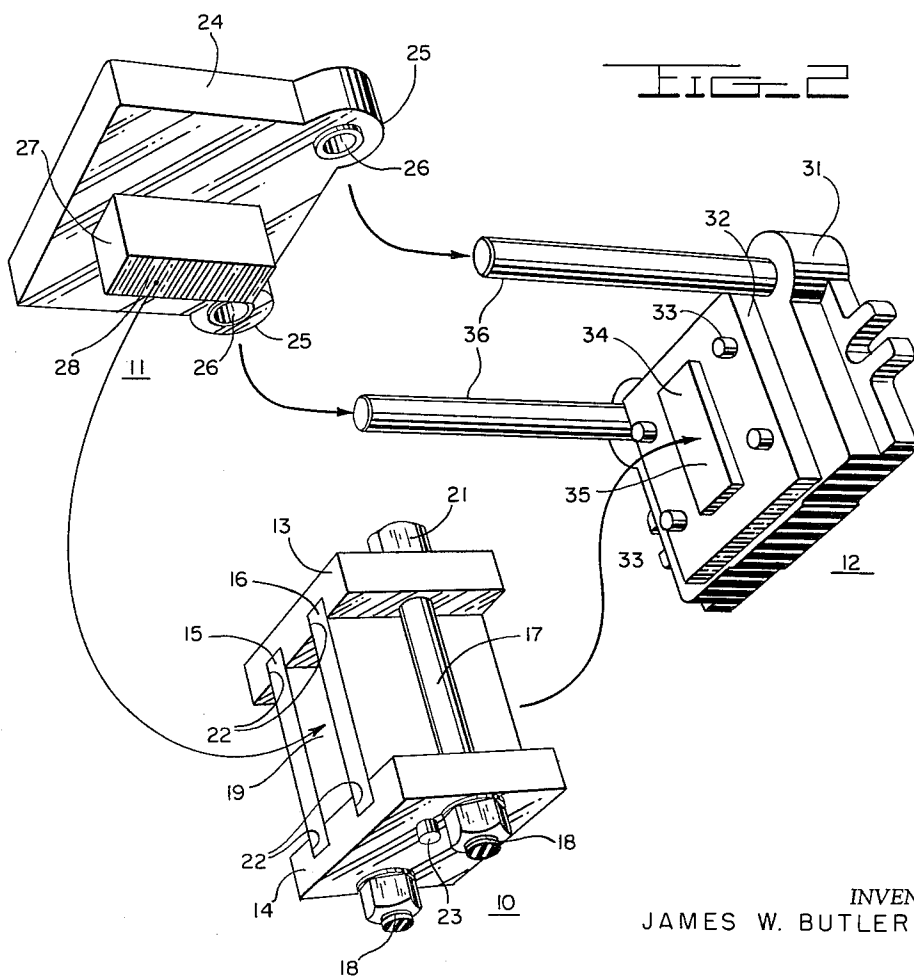


FIG. 2



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METHOD OF MAKING A NEUTRON-ABSORBING CALCIUM BORIDE BRICK

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(Granted under Title 35, U.S. Code (1952), sec. 266)

The invention described herein may be manufactured and used by or for the Government of the United States of America for governmental purposes without the payment of any royalties thereon or therefor.

The present invention relates to a shielding medium for providing protection against emanations of radioactivity, especially neutrons, and more particularly to a method of making a shielding medium of universal application, primarily against neutrons.

It is well known that people and equipment must be protected from harmful effects of certain radiation and the protection thereof is very important. In the past, radiation shielding against emanations of low energy was satisfactorily effected by providing a barrier of a sufficient quantity (in thickness) of any relatively dense material to absorb the radiation. With the advent of the nuclear reactor, the cyclotron, and other devices and processes wherein radiation, including neutrons, in enormous quantity and of great energy may be produced, the problem of shielding has become a much more complicated matter. These devices require more practical considerations and make desirable special radiation absorption materials to reduce the weight, volume, and cost of the materials. Protection against radiation emanating from such devices have been made in the form of walls, ceilings, and floors in, or on, which a sheet of lead or other radiation absorption metals have been used. Heretofore, attempts have been made to reduce the weight, area coverage, and cost of suitable shielding structures. As such, special concrete compositions of enhanced shielding efficiency have been devised which include metallic elements such as iron or lead having a high-atomic-weight. Such shielding materials have their drawbacks since it is known that in absorbing neutrons, especially fast neutrons, "secondary" radiation such as secondary gamma radiation is generated.

This requires additional shielding to absorb the "secondary" gamma radiation thus generated.

A somewhat suitable thermal neutron shield has been developed and patented as Patent Number 2,727,996 by T. Rockwell and V. L. McKinney. The patented shield is made with a continuous matrix of a malleable metal with a solid refractory material substantially constituted of boron discontinuously and uniformly dispersed therein with a sheathing of a malleable metal, such as aluminum, bonded thereto. Suitable refractory material being boron carbide (B_4C), boron oxide (B_2O_3), sodium tetraborate, boron phosphide and boron sulfide. Suitable shield structures made of the above material are hard to fabricate and are extremely costly both in material and manufacturing cost. The cost of (B_4C) is so great that large scale shielding use would be prohibitive.

The present invention overcomes the drawbacks of prior art neutron shielding structures and provides a method for making a simple, relatively inexpensive neutron shield in the form of bricks made of calcium boride (CaB_6) which can be used continuously without any noticeable deterioration.

It is therefore an object of the present invention to provide a method of making a relatively inexpensive neutron shielding element.

Another object is to make a simple, inexpensive neu-

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tron shield element which has a fairly high density and is long-lived.

Still another object is to make a neutron shielding element which does not deteriorate and can be formed into different shield structures and can be used at high temperatures if necessary.

Other and more specific objects of this invention will become apparent upon a careful consideration of the following detailed description when taken together with the accompanying drawings, in which;

FIG. 1 illustrates a plurality of bricks illustrating the different dimensions of a neutron shield structure made by the method of this invention; and

FIG. 2 illustrates a mold by which neutron shield bricks are formed.

Neutron shield bricks made in accordance with the method of the present invention are of calcium boride (CaB_6) made into a thick, dry mud-like substance by mixing water with the CaB_6 , placing the substance into a mold to form bricks by compression and then baking the formed bricks for a period of time.

More specifically, inexpensive neutron shield bricks 9 made according to the method of the present invention are made by a specially built mold shown by illustration in FIG. 2. The mold is made of the utmost simplicity which requires operation of only two nuts to assemble and disassemble the mold during the process of forming bricks. The mold includes a central section 10, a bottom section 11, and a top section 12.

The center section comprises similar end plates 13 and 14 separated by similar side plates 15 and 16, which are held together to form a rectangular space 19 therebetween by two bolts 17 and nuts 18 in which the bolts pass through the end plates 13 and 14 parallel to the outer surfaces of side plates 15 and 16. The bolts are provided with heads 21 on the outside of end plate 13 and the nuts 18 on the outer side of end plate 14 whereby the nuts can be tightened against plate 14 to hold the center section together. As shown, end plates 13 and 14 have slots 22 therein within which the ends of plates 15 and 16 are fitted to hold the plates 15 and 16 in their correct position. In order to correctly align side plates 15 and 16 with end plates 13 and 14, side plate 15 is secured to end plate 13 by bolts passing through plate 13 and screwed into the end of side plate 15 and side plate 16 is secured to end plate 14 by bolts 23 passing through end plate 14 and screwed into the end of plate 16. Another manner would be to secure pins in the end plates which would match with holes in the ends of side plates 15 and 16.

The bottom section 11 comprises a bottom plate 24 which has two rounded corners 25 with two spaced guide holes 26 therein. The center of the bottom plate is made with a brick shaped portion or plunger 27 that fits into the area 19 of the center section bounded by the end and side plates. The plunger 27 has a close fit with the surfaces of the center section and is provided with a suitable clearance to allow excess water to escape during molding of the bricks.

The upper section 12 is provided with an upper plate 31 to which a top plate 32 similar to the bottom plate 24 is secured by suitable means such as bolts 33. The plate 32 has a brick shaped portion or plunger 34 made in the center thereof and which fits from the top into the open area 19 of the center section bounded by the side and end plates. The upper section 12 is also provided with spaced guide rods 36 which match with the guide holes 26 in the bottom section. The guide bars insure that the top and bottom sections remain in proper alignment during compression of the bricks. In order to

remove more easily the bricks from the mold, the end and side plates as well as the sides 28 and 35 of the brick shaped plungers which are facing outwardly from the bottom and upper plates have smooth surfaces facing the open area 19 of the center section.

In carrying out the teaching of the method of the present invention, calcium boride (CaB_6) containing a minimum of about 50% boron by weight is preferred. For example, a satisfactory composition has been determined to contain the following elements by weight: boron 52%, calcium 31%, carbon 8%, iron 3%, and others 3%. The neutron absorbing bricks are made from about 100 parts by weight of CaB_6 in the dry powder form mixed with about 5 to 10 parts by weight of water. The water is sufficient to make a thick, fairly dry mud. The amount of water is not critical. If excessive amounts of water are used, the excess will simply seep out of the mold, the main disadvantage being the inconvenience caused by the leakage of the dirty water. If too little water is used, the "green" brick has too little cohesive strength and might break or crumble upon transferral to the oven. Furthermore, a brick made with no water at all has less cohesive strength, even after the baking operation, than do those which have a few parts of water per 100 parts of CaB_6 . A "green" brick is one which has been molded but not cured as by heating. The center section of the mold is assembled and the bottom portion is placed with the center brick shaped plunger 27 inserted from the bottom into the rectangular space 19 formed by the end and side plates of the center section. The CaB_6 -water mixture is weighed to the proper amount of about 2 lbs. to form a suitable brick 2 in. x 3 in. x 5 in. The weighed mixture is placed into the rectangular space of the center section of the mold and then the upper section is placed over the center section with the guide rods 36 inserted into the guide holes 25 of the bottom section. Thus the top portion is guided into place with the brick shaped plunger 34 fitting into the upper portion of the center section over the CaB_6 -water mixture which has been placed into the center section. The mold is then placed in a suitable device for applying pressure to the bottom and top sections such that the pressure is applied directly over the brick shaped plungers. It has been determined that suitable bricks are formed by application of from about one ton per square inch to about two tons per square inch pressure to the upper plate wherein the higher pressures make a denser brick. After compressing the CaB_6 -water mixture in the mold to form a "green" brick, the "green" brick is removed from the mold by removing the top section from the mold and then removing the bolts 17 such that each of the end plates along with their attached side plates of the center section can be removed. The "green" brick is then transferred from the bottom section to a ceramic baking tray by a gentle horizontal push on the brick. A number of "green" bricks are formed and placed onto the tray and then baked in a suitable baking oven.

The temperature required to bake the "green" bricks is rather critical to prevent decomposition of the compound and to prevent the bricks from crumbling by handling. The "green" bricks are preferably placed into a cold oven and the temperature brought up slowly to the desired temperature to prevent too-rapid evaporation and expansion of the residual water left in the green bricks. Too-rapid heating causes fissures and sometimes complete breaking of the green bricks. The best quality bricks are formed by bringing the heat up slowly during a warmup time of about two hours to a baking temperature of from about 600 degrees centigrade to about 900 degrees centigrade with about 750 degrees preferred and maintaining the baking temperature constant for a period of from about two hours to about six hours. After baking for the preferred time, the heat source is cut off and the bricks are allowed to cool in the oven

at the slowest heat off cooling rate, preferably overnight or for at least several hours. The bricks are now cured and ready to be removed from the oven for use as desired in a neutron shield structure.

In order to insure accuracy in making bricks of uniform size, the initial weighed charge is made slightly lighter than normal and then compressed. The upper plunger is then removed and the compressed CaB_6 -water mixture is visually inspected. If the brick is undersized, additional mixture is added to the mold and again compressed to form a brick of the correct size. The brick is then removed and cured as described above.

The process taking place during the making of the brick is not fully understood; however it is believed that a slight amount of boron oxide B_2O_3 is formed from a reaction between very small amounts of decomposed CaB_6 and the water mixed with the CaB_6 . CaB_6 does not melt because its decomposition point is lower than the melting point. Since B_2O_3 has a melting point of about 577 deg. C., and the required temperatures are greater than 600 deg. C., it is believed that the minimum critical temperature is dictated by the melting point of the slight amount of B_2O_3 formed, and that it is important that this small amount of B_2O_3 be melted. The B_2O_3 is then believed to act as a binder to hold the brick together. It is further believed that above 900 deg. C. so much of the CaB_6 decomposes that the brick is deformed and will not maintain its shape.

It has been determined that the bricks made in accordance to the method of this invention are very stable and are not affected by normal indoor temperatures and humidities, and can be used for indoor structures without any further treatment. However, for outside wall structures it is necessary to paint the brick structure with a waterproof non-aqueous masonry paint or other waterproof non-aqueous coating.

The bricks of a size 2 x 3 x 5 inches weigh about two lbs. each and have an average density of about 100 lbs./ft.³ or 1.6 gm./cm.³. Such bricks can be used as a structural material for reactor rooms, accelerator rooms, experiment rooms, etc. and have withstood a test of 12,000 lbs. of force along the 2-inch dimension after considerable use. Temporary shielding walls may be erected without the use of any bonding cement, but for permanent walls, a non-aqueous cement, such as thermal setting plastics, may be used.

Obviously many modifications and variations of the present invention are possible such as, for example, the size and shape of the bricks in the light of the above teachings. It is therefore to be understood that within the scope of the appended claims the invention may be practiced otherwise than as specifically described.

What is claimed is:

1. A method of making neutron-absorbing bricks which comprises mixing water with calcium boride powder to form a thick, dry mud-like substance, placing a portion of said mud-like substance into a mold, compressing said mud-like substance in said mold at a pressure of about one ton/in.² to about two tons/in.² to form a "green" brick, removing said "green" brick from said mold by disassembling said mold, placing said "green" brick into an unheated oven, applying heat slowly to said oven during a warmup time of about two hours to raise the oven to a baking temperature of about 600° C. and baking said brick at a temperature from about 600° C. to about 900° C. for about two to six hours, discontinuing the application of heat and allowing said temperature to return slowly to room temperature, said brick remaining in said oven until room temperature is reached.

2. A method of making neutron-asborbing bricks which comprises mixing water with calcium boride powder to form a thick, dry mud-like substance, placing a portion of said mud-like substance into a mold, compressing said mud-like substance in said mold at a pressure of about one

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ton/in.² to about two tons/in.² to form a "green" brick, removing said "green" brick from said mold by disassembling said mold, placing said "green" brick into an unheated oven, uniformly applying heat slowly to said oven during a warmup time for a period of about two hours to raise the temperature of said oven up to about 600° C. and baking said brick at a temperature from about 600° C. to about 900° C. for from two to six hours, discontinuing the application of heat and allowing said temperature to return slowly to room temperature and removing said brick from said oven.

3. A method of making neutron-absorbing bricks which comprises mixing 5-10 parts water by weight with 100 parts by weight of calcium boride to form a thick dry mud-like substance, placing a portion of said mud-like substance into a mold, compressing said mud-like substance in said mold at a pressure of about one ton/in.² to about two tons/in.² to form a "green" brick, removing said "green" brick from said mold by disassembling said mold, placing said "green" brick into an unheated oven, applying heat slowly to said oven during a warmup time of about two hours to raise the oven to a baking temperature of about 600 deg. C. and baking said brick at a temperature from about 600 deg. C. to about 900 deg. C. for about two to six hours, discontinuing the application of heat and allowing said temperature to return slowly to room temperature, said brick remaining in said oven until room temperature is reached.

4. A method of making neutron-absorbing bricks in accordance to the method claimed in claim 3 wherein the amount of boron in said calcium boride is at least 40% by weight.

5. A method of making neutron-absorbing bricks which comprises mixing 5-10 parts water by weight with 100

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parts by weight of calcium boride to form a thick dry mud-like substance, placing a portion of said mud-like substance into a mold, compressing said mud-like substance in said mold at a pressure of about one ton/in.² said "green" brick from said mold by disassembling said mold, placing said "green" brick into an unheated oven, to about two tons/in.² to form a "green" brick, removing applying heat slowly to said oven during a warmup time of about two hours to raise the oven to a baking temperature of about 600 deg. C. and baking said brick at a temperature of about 750 deg. C. for about two to six hours, discontinuing the application of heat and allowing said temperature to return slowly to room temperature, said brick remaining in said oven until room temperature is reached.

References Cited in the file of this patent

UNITED STATES PATENTS

1,858,413	Noack et al. -----	May 17, 1932
1,913,373	Golyer -----	June 13, 1933
2,647,061	Lowe -----	July 28, 1953
2,728,128	Sheer et al. -----	Dec. 27, 1955

FOREIGN PATENTS

209,936	Australia -----	Feb. 2, 1956
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OTHER REFERENCES

- Rockwell, Reactor Shielding Design Manual, 1956, p. 190.
 Nuclear Science Abstracts (NSA), vol. 11, No. 20, Supplement, Oct. 31, 1957, p. 1304, abstract No. 11,745.
 NP-8086, NRL Quarterly on Nuclear Science and Technology, Oct. 1, 1959, pp. 13-16.