United States Patent Office

2,986,533

Patented May 30, 1961

2,986,533

COMPACT HIGH DENSITY RADIATION SCREENING MATERIAL CONTAINING TUNGSTEN

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No Drawing. Original application Nov. 12, 1958, Ser. No. 773,174. Divided and this application June 19, 1959, Ser. No. 821,359

7 Claims. (Cl. 252—478)

The present invention relates in general to a new and useful composition of matter having the characteristics of a high density and of absorbing or screening radiations of X-rays and gamma rays without producing excessive secondary gamma ray radiation upon impingement of such rays upon it. Co-pending application Serial No. 773,174, filed November 12, 1958, of which this application is a division relates in general to an improved method of making this new composition of matter. Said application describes both the new composition and the improved method and claims the latter.

The shielding materials of the prior art, although efficient as shields, are in many instances of little practical or commercial use because they do not have the structural strength to be self-supporting. Therefore, when it is desired to shield an area from radiation of the type described it is necessary to construct a composite element a portion of which has the shielding element and another portion of which has the structural supporting element. It is obvious that a material which has the ability to serve as both the shielding element and the structural supporting element offers many practical and commercial advantages.

It is an object of the present invention to provide a useful composition of matter which has a combination of these advantages, radiation shielding and structural strength.

In the past lead used alone and certain alloys of tungsten with nickel and copper have been used as radiation shields with varying degrees of success. Lead alone is easily deformable, is mechanically weak and requires special handling. Tungsten and its known alloys are expensive and are very difficult to work with because of their hardness which makes them difficult to machine and otherwise fabricate to desired sizes and shapes.

I have discovered that the combination of lead and tungsten combined in the special manner hereinafter described produces a new and commercially useful product which has the qualities desired, radiation shielding and structural strength.

Attempts have been made in the past to combine lead and tungsten by means such as melting the lead and mixing powdered tungsten therein. This resulted in segregation of the two metals upon solidification of the lead without an alloy or even alloy-like structure being formed. No useful composition of matter resulted.

The present application discloses and claims a new and useful composition of matter including a combination of tungsten and lead in a compact body of high density capable of absorbing or screening radiations and describes preferred methods of manufacture of said composition of matter as brought out and exemplified in the disclosure hereinafter set forth, the scope of the invention being indicated in the appended claims.

While a preferred embodiment of the invention is described herein, it is contemplated that considerable variation may be made in the method of procedure and the

combination of elements, without departing from the spirit of the invention.

I prepare my novel and useful composition of matter by using the materials and following the procedures hereinafter set forth. First, granulated tungsten powder particles are prepared in the usual manner to have a bulk density in the order of 60–140 grams per cubic inch. Then these prepared particles are treated by placing them in an aqueous solution containing decomposable salts of lead, tin, copper and nickel. The solution is then evaporated and upon evaporation particles of all of the metal salts present are deposited as a coating on the tungsten particles.

These coated particles after being sieved through a screen of suitable mesh, say 200, are placed in a suitable container and heated to decompose and reduce the metal salts. In order to accomplish the desired decomposition and reduction of the salts the heating should be done in a reducing atmosphere. The reducing atmosphere may be provided in any well known and conventional way such as by heating in a furnace chamber that is completely evacuated or contains an atmosphere of hydrogen or of a suitable inert gas. Another alternative is to heat the particles in a graphite boat. The formation of this metallic coating is beneficial to the promotion of a honeycomb type of structure which, as is later explained, is an essential step in the formation of the compact body made of my novel composition of matter.

The next step is to compress the tungsten particles now covered with a metal coating of alloy-like nature in a suitable mold under controlled conditions of pressure and temperature to produce a pressed metallic body having a honeycomb structure. The amount of pressure applied and the temperature used are regulated to produce a pressed body having a predetermined desired density. The next step, which is accomplished while the body is still in the mold is to added lead in the form of shot or in any other suitable and convenient form and to heat the mixture in a suitable furnace or by means of high frequency coils placed over the mold to a temperature above the melting point of lead to cause the lead to become molten and run into the voids in the pressed body whereby said voids therein are filled and the final product is formed. This can then be ejected from the mold. The heating should be done in a protective atmosphere to prevent oxidation. This protective atmosphere can be produced by placing the mold and lead in a chamber that is completely evacuated or contains hydrogen or a suitable inert gas.

An alternate method is to place the coated tungsten particles, after the metal salts forming the coating thereon have been decomposed and reduced, between a series of heated rollers and to pass them therethrough and thus to compress the powder to a predetermined desired density. When this density is reached sheets of lead are placed on top and bottom of the resulting pressed tungsten body and between that body and a series of rollers. Further compression between and heating by the rollers will force the lead through the interstices in the surface of the pressed tungsten body and into the voids therein. The compression and heating by the rollers is continued at a temperature just below the melting point of lead until all voids are substantially filled with lead and a homogenized compact of full desired density is obtained.

In some instances it has been found advantageous to reinforce the coating on the tungsten particles. A preferred way of doing this is to immerse the coated particles before they have been heated and pressed into a pressed tungsten body in an aqueous solution of lead nitrate. The solution is then evaporated whereby lead nitrate is added to the coating on the tungsten particles and this is then decomposed and reduced in the manner described above whereby metallic lead becomes an integral part of the alloy-like metal coating on the particles. These particles are then sieved through a screen of the size desired and are pressed into a body having a honeycomb structure as has been previously described. Lead is then added by either of the methods described above to fill the voids therein. It has been found under some conditions that this method results in a higher content of lead in the final compact and, therefore, in a compact of higher density.

The final density of bodies made as above described is determined by the density of the pressed tungsten body before any of the lead is added and by the percentage of voids still remaining in the body after the addition of the lead. The latter is controlled by the amount of lead added, the temperature at which it is added and the time allowed for the lead to enter the voids. Densities in excess of 99% of the theoretical maximum density have been attained.

The composition of matter formed by the above described novel treatment of tungsten particles with lead comprises a compact body having strong bonds between the particles of coated tungsten and the lead filling the voids of the original honeycomb structure. Due to the production of an alloy-like coating on the tungsten particles the resultant pressed tungsten body develops a strong catalytic-like action in making the composite body one of high strength and high density by promoting the substantially complete filling of the voids. The densities of bodies produced in this manner range from 12.0–18.5 grams per cubic centimeter. The compacts so produced are structurally strong, readily machineable and are not deformable even at elevated temperatures. Specimens have been subjected to standard tensile tests and have shown tensile strengths of from 12,000 pounds per square inch to 25,000 pounds per square inch, the difference depending upon the pressure used to form the pressed tungsten body before the lead was added. Standard compression tests showed speciments to have a value of from 20,000–30,000 pounds per square inch. From these values it is obvious that the composition of matter invented by me which forms the compact has tensile and compression strengths well within the ranges making them suitable as self-supporting structural elements.

The following are specific examples of compact bodies of the novel composition of matter produced by me and are given by way of illustration, but do not in any way limit the scope of this invention.

Example I

One kilogram of pure tungsten powder having a bulk density of substantially 60 grams per cubic inch was first treated with an aqueous solution containing stannic chloride, nickel nitrate and cupric nitrate. The amounts of the metal salts present in the batch of solution into which the particles were immersed were computed to have a combined metal content of 5 grams or 0.5% of the weight of the tungsten particles immersed in the solution. This comprised 1.25 grams of tin, 2.5 grams of nickel and 1.25 of copper, thus the ratio of the metal contents of the specific salts used was 50% by weight of nickel and 25% by weight each of copper and tin. After immersion and thorough mixing in the solution heat was applied to evaporate the solution. Constant stirring during heating was necessary to assure uniform distribution of the metal salts on the surface of the tungsten particles to form a coating. The coated particles then were sieved through a 200 mesh screen and the screened particles were then placed in a nickel boat and heated in a reducing atmosphere whereby the particles of metal salts on the surface of the tungsten particles were decomposed with the result that the coating was converted to an alloy-like coating of tin, nickel and copper.

500 grams of these coated particles were then placed in a mold and again heated and compressed to a density of 10.4 grams per cubic centimeter (170 grams per cubic inch). When this density was reached there was added in the mold 260 grams of lead in the form of shot around the compressed particles now forming a pressed body. This amount was slightly in excess of the theoretical amount required to fill the voids in the pressed body. The mixture in the mold was then heated in a furnace chamber containing an atmosphere of hydrogen to a temperature of 850° C. for a period of approximately 15 minutes during which period all the lead had become molten and had seeped down through the honeycomb structure of the compacted body of coated tungsten particles. The mold was allowed to cool and the compact ejected therefrom. Excess lead was removed and the density of the piece was determined by water displacement. For the density to which the tungsten body was pressed before addition of the lead in this example, the theoretical density would be 15.6 grams per cubic centimeter if all the voids were filled. The actual density of the compact made as described in this example, measured by water displacement, was 15.4 grams per cubic centimeter.

Example II

The composition of matter produced by this example was made in substantially the same manner as that described in Example I except that before the coated particles of tungsten were pressed to form a pressed tungsten body the particles were given an additional coating of lead by immersing them in an aqueous solution of lead nitrate in which the lead present weighed 30 grams or 3.0% by weight of the tungsten in the coated particles immersed therein. The particles were well mixed in the solution which was heated during constant stirring until the solution was evaporated and the particles dried. These particles were then screened through a 200 mesh screen and the lead nitrate now included in the coating thereon was reduced to lead by heating the coated particles in a nickel boat in a reducing atmosphere whereby the lead nitrate was decomposed and reduced. The result was that metallic lead was added to the alloy-like coating on the tungsten particles. These particles were then treated by the same methods described in Example I to form a compressed tungsten body and then a compact of high density containing lead in the former voids. The compact produced by the method described in this example was homogeneous and had a density of 15.4 grams per cubic centimeter very close to the full theoretical density of 15.6 grams per cubic centimeter for the density of 10.4 grams per cubic centimeter (170 grams per cubic inch) of the pressed tungsten body before the lead was introduced.

Example III

In this example 500 grams of tungsten powder having a bulk density of substantially 95 grams per cubic inch were coated with the same metals and in the same manner as set forth in Example I. The entire batch of coated particles was then placed in a mold and again heated and compressed to a density of 11.6 grams per cubic centimeter (190 grams per cubic inch). When this density was reached 200 grams of lead were added as set forth in Example I, this amount being slightly in excess of the theoretical amount required to fill the voids in the pressed body. The actual density of the compact made as described in this example, measured by water displacement, was 16.0 grams per cubic centimeter whereas the theoretical density, if all the voids were filled, would have been 16.2 grams per cubic centimeter.

Example IV

In this example 500 grams of tungsten powder having a bulk density of substantially 120 grams per cubic inch were coated with the same metals and in the same manner as set forth in Example I. The entire batch of coated particles was then placed in a mold and again heated and compressed to a density of 12.2 grams per cubic centimeter (200 grams per cubic inch). When this density was reached 220 grams of lead were added as set forth in Example I, this amount being slightly in excess of the theoretical amount required to fill the voids in the pressed body. The actual density of the compact made as described in this example, measured by water displacement, was 16.1 grams per cubic centimeter whereas the theoretical density, if all the voids were filled, would have been 16.3 grams per cubic centimeter.

Example V

One kilogram of pure tungsten powder having a bulk density of substantially 140 grams per cubic inch was first treated with an aqueous solution containing nickel chloride, cupric chloride and stannic chloride. The amounts of the metal salts present in the batch of solution into which the particles were immersed were computed to have a combined metal content of 30 grams or 3.0% of the weight of the tungsten particles immersed in the solution. This comprised 7.5 grams of tin, 15 grams of nickel and 7.5 grams of copper, thus the ratio of the metal contents of the specific salts used was 50% by weight of nickel and 25% by weight each of copper and tin. With the exception of metal salts and the total weight of metal present in the metal salt solution, the same steps were followed, pressures used, temperatures used and weight of lead added as were followed and used in Example I. The theoretical density of a compact for the density to which the tungsten body was pressed before addition of the lead in this example would be 15.4 grams per cubic centimeter if all the voids were filled. The actual density of the compact made as described in this example, measured by water displacement, was 15.3 grams per cubic centimeter.

Example VI

The composition of matter produced by this example was made in substantially the same manner as that described in Example V except that as described in Example II, before the coated particles of tungsten were pressed to form a pressed tungsten body the particles were given an additional coating of lead by immersing them in an aqueous solution of lead nitrate in which the lead present weighed 30 grams or 3.0% by weight of the tungsten in the coated particles immersed therein. The procedures, pressures, temperatures and weight of lead added from this point on were those described in Example I as varied by Example II. The compact produced by the method described in this example was homogeneous. The theoretical density of a compact for the density to which the tungsten body was pressed before addition of the lead in this example would be 15.2 grams per cubic centimeter if all the voids were filled. The actual density of the compact made as described in this example, measured by water displacement, was 15.2 grams per cubic centimeter.

What I claim is:

1. A radiation screening material of high density, high tensile strength and high compression strength consisting of compacted tungsten particles coated with at least one of the metals in the group consisting of tin, nickel and copper and bonded together by lead which fills the voids between said particles to the extent that the actual denisty of said material is substantially from 98,7% to 100.0% of the theoretical density thereof if all the voids were completely filled.

2. A radiation screening material in accordance with claim 1 in which said metallic coating is between substantially .5% and substantially 3.0% by weight of said tungsten particles.

3. A radiation screening material in accordance with claim 1 in which said metallic coating is substantially .5% by weight of said tungsten particles.

4. A radiation screening material in accordance with claim 1 in which said metallic coating is substantially 3.0% by weight of said tungsten particles.

5. A radiation screening material of high density, high tensile strength and high compression strength consisting of compacted tungsten particles coated with at least one of the metals in the group consisting of tin, nickel and copper in which said metallic coating is between substantially .5% and substantially 3.0% by weight of said tungsten particles and said metallic coating has added thereto a lead coating comprising substantially 3.0% by weight of said tungsten particles and in which said particles are bonded together by additional lead which substantially fills the voids between said particles.

6. A radiation screening material of high density, high tensile strength and high compression strength consisting of compacted tungsten particles coated with at least one of the metals in the group consisting of tin, nickel and copper in which said metallic coating is substantially .5% by weight of said tungsten particles and said metallic coating has added thereto a lead coating comprising substantially 3.0% by weight of said tungsten particles and in which said particles are bonded together by additional lead which substantially fills the voids between said particles.

7. A radiation screening material of high density, high tensile strength and high compression strength consisting of compacted tungsten particles coated with at least one of the metals in the group consisting of tin, nickel and copper in which said metallic coating is substantially 3.0% by weight of said tungsten particles and said metallic coating has added thereto a lead coating comprising substantially 3.0% by weight of said tungsten particles and in which said particles are bonded together by additional lead which substantially fills the voids between said particles.

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