

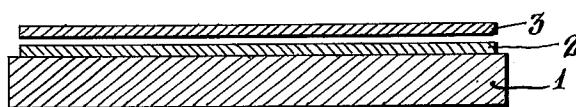
Oct. 10, 1950

G. JORIS

2,525,035

RADIO-ACTIVE DEVICE AND METHOD OF MAKING THE SAME

Filed Dec. 15, 1944



INVENTOR.
George Joris
BY *Moore & Flum*

ATTORNEYS

UNITED STATES PATENT OFFICE

2,525,035

RADIOACTIVE DEVICE AND METHOD OF
MAKING THE SAME

George Joris, Princeton, N. J., assignor to Canadian Radium & Uranium Corporation, New York, N. Y., a corporation of New York

Application December 15, 1944, Serial No. 568,382

3 Claims. (Cl. 117—33.1)

1

My invention relates to a new and improved carrier of radio-active material, and to a new and improved method of making the same.

One of the objects of my invention is to provide a carrier with a layer which comprises radioactive material, at one or more faces of said carrier. Said carrier is preferably made of metal or an alloy, or of any other suitable material.

The radio-active material is preferably, but not necessarily, a salt of radium which is insoluble in water.

As an example of my invention, I dissolve in water, a water-soluble salt of radium and also a water-soluble salt of an auxiliary metal. This water-soluble salt of the auxiliary metal is preferably a salt of a metal other than radium. I can use one or more auxiliary metals. I then precipitate, from said aqueous solution, a water-insoluble salt of radium and a water-insoluble salt or salts of said auxiliary metal or metals. I can thus provide a water-insoluble co-precipitate of the water-insoluble radium salt and of a water-insoluble salt or salts of the auxiliary metal or metals, which is very fine and uniform, so that the respective salts are very finely and uniformly dispersed in molecular or substantially molecular relation.

That is, the water-insoluble salt of the auxiliary metal may be silver carbonate and the water-insoluble radium salt may be radium carbonate. The silver carbonate has ultimate crystals in said co-precipitate.

The silver ion in some of said silver carbonate crystals is replaced by the radium ion. This produces a molecular distribution of the silver and the radium in said co-precipitate. The silver ion is thus replaced by the radium ion, in only some of said ultimate crystals, because I prefer to provide an excess of silver carbonate in said co-precipitate.

I then apply this co-precipitate to the surface of the carrier, so that said co-precipitate adheres to the respective surface of said carrier. I then decompose the salts or compounds of the auxiliary metal or metals in said co-precipitate, thus producing the respective metal or metals, without decomposing the radium salt.

I can thus provide the carrier with a surface layer which can be very thin, and in which the water-insoluble radium salt is very finely and uniformly held and distributed in the auxiliary metal or metals of said surface layer. The alpha rays and/or other rays or emissions of the radium salt can penetrate this very thin surface layer, thus providing a permanent radiation of alpha rays and/or the other rays. After the surface layer has been finished, it can be subjected to

2

gentle rolling pressure, in order to spread said surface layer more uniformly, and to produce a surface layer of uniform thickness.

Numerous additional important objects and advantages of my invention will be stated in the annexed description and diagrammatic drawing which illustrates a preferred embodiment thereof. The drawing is not to scale.

I will first illustrate the invention in an illustrative example in which the carrier 1 which is shown in the annexed drawing is made of metallic silver, the surface layer 2 at the respective surface of the carrier 1 is a mixture of metallic silver and radium carbonate, and the final protective surface layer 3 is made of gold. In the drawing, these elements are shown separated from each other, for convenience, but in practice these elements 1, 2 and 3 are in abutting relation. As an illustration, it is assumed that each square centimeter of the surface of the silver carrier 1 has a mass of radium carbonate, RaCO_3 , which corresponds to 220 micrograms of radium per se. Since the molecular weight of radium carbonate is 285.97, and the atomic weight of radium is 225.97, this corresponds to a distribution of a little less than 286.00 micrograms of radium carbonate per square centimeter of the surface layer 2, which is firmly fixed to carrier 1.

I dissolve in a minimum amount of water, a corresponding mass of radium nitrate, $\text{Ra}(\text{NO}_3)_2$, so that the mass of the radium per se, in said radium nitrate, is 220 micrograms, if it is desired to cover only one square centimeter of the surface of the carrier 1. It is assumed, for convenience, that there is no loss of material. In practice, such loss is very slight. I also dissolve in the water of said solution, a suitable amount of silver nitrate, AgNO_3 . The weight of the silver per se, in said mass of silver nitrate, can be from five to twenty times as much as the weight of the radium per se. This solution is uniform throughout.

I use a minimum amount of water in making this uniform solution, so as to avoid or minimize any loss of radium salt in the subsequent precipitation.

I then dissolve in the water of said uniform solution, at room temperature of about 20° C.—25° C., enough sodium carbonate or other suitable water-soluble carbonate, in order to co-precipitate all the radium and all the silver, in the form of their respective carbonates. Instead of sodium carbonate, I can use potassium carbonate, ammonium carbonate, etc.

Both silver carbonate and radium carbonate are substantially insoluble in water. I then separate the coprecipitate from the mother liquid by

decantation or filtration or in any other suitable manner.

In this co-precipitate, the radium carbonate and the silver carbonate are in molecular relation as previously explained.

This co-precipitate is then dried in any suitable manner. This co-precipitate can be dried in the open air at 20° C.-25° C., or by gentle heat or by placing said co-precipitate in a suitable desiccating chamber or the like. When so dried, said co-precipitate still consists of radium carbonate and silver carbonate in said molecular relation.

The respective surface of the silver carrier 1 is then carefully cleaned and dried. The co-precipitate is then applied to the respective surface of the silver carrier 1, so that said co-precipitate adheres to said surface of the carrier 1. For this purpose, the co-precipitate can be temporarily moistened with pure and substantially 100% ethyl alcohol or with water-free acetone or a water-free volatile essential oil or the like, just enough to make a paste which adheres to the respective surface of the silver carrier 1. This paste can be applied to said respective surface with or without pressure. Water can be used for making the paste, but I prefer a non-aqueous medium.

The carrier 1 and the surface layer of adhering co-precipitate are then heated to a suitable temperature at which the radium carbonate remains undecomposed, and at which the silver carbonate is decomposed to metallic silver and the ethyl alcohol, acetone, etc. are evaporated. By heating to said suitable temperature, the silver carbonate is first decomposed to form silver oxide, and the silver oxide is then decomposed to form metallic silver. Since silver oxide decomposes when heated to more than about 300° C. to produce metallic silver, and since radium carbonate is stable up to about 1400° C., I can easily form the metallic silver in situ, by heating to a temperature at which metallic silver is thus formed. This metallic silver adheres firmly to the material of the silver carrier 1. This metallic silver binds the radium carbonate to the silver carrier 1. The silver carbonate of the co-precipitate can be thus heated and decomposed in the open air, or in a reducing atmosphere of hydrogen or the like. I can use any suitable method for producing the metallic silver from any water-insoluble compound thereof. As previously stated, this finished surface layer of silver and radium carbonate can be subjected to gentle rolling pressure, to produce a surface of uniform thickness.

The thickness of the finished layer 2 is a maximum of one to two microns and it may be much thinner, as 0.1 micron or less, depending on the proportion of silver and the area which is covered. After the finished adhering layer 2 has been formed, it is preferably protected by a thin enclosing surface layer of gold 3, which is formed in the usual manner. The alpha rays and other rays from the radium of the radium carbonate easily penetrate this protective layer 3. The radon gas which results from the disintegration of the radium is wholly retained by the gold protective layer 3, until constant radioactivity is produced. That is, the radon gas is prevented from leaking through the protective gold layer 3, so that said radon gas disintegrates to emanate the alpha rays which pass through the layer 3.

After a certain period, a constant ratio is pro-

duced between the formation of the radon gas, and its disintegration, so that the emanation of the alpha rays becomes fixed.

The gold layer 3 is very thin, to permit the passage of the alpha rays and other rays.

The invention is not limited to the specific illustrative example, although I prefer to form the auxiliary metal of the layer in situ, by decomposing a compound of the auxiliary metal in situ.

Thus, the co-precipitate can be applied to a carrier made of a metal or alloy other than silver, or any suitable material. The metal of the layer is preferably the same as the metal of the carrier, or the metal of the layer can readily alloy with the metal of the carrier. Any suitable adhesive may be used for fixing the co-precipitate to the carrier.

The adhesive is preferably evaporated or decomposed, during the heating which decomposes the compound of the auxiliary metal.

Instead of using silver as the auxiliary material, I can use other metals.

Thus, cupric carbonate, which is insoluble in water, is produced by adding sodium carbonate to a solution of cupric nitrate. Hence, I can dissolve a water-soluble radium salt in water in which cupric nitrate is dissolved, and co-precipitate cupric carbonate and radium carbonate, by means of sodium carbonate. By suitable heating in the mother liquid, the cupric carbonate is gradually converted into black oxide of copper, namely, cupric oxide. This co-precipitate of cupric carbonate and radium carbonate, or the co-precipitate of cupric oxide and radium carbonate, can be applied adherently to a carrier of silver, copper, etc. If desired, the copper salt can be reduced to metallic copper, by heating the carrier and its co-precipitated surface layer, in a reducing atmosphere of anhydrous hydrogen. The copper oxide can also be readily reduced to metallic copper in an atmosphere of formic acid vapor, while leaving the radium carbonate undecomposed.

Numerous other suitable water-insoluble salts or compounds of various metals can thus be co-precipitated in unison with radium carbonate. The invention is not limited to any specific radium salt. It also includes compounds of other radio-active elements such as thorium, meso-thorium, actinium, polonium.

The above method is preferred in working with small quantities of a radium salt. However, the claims for the final article of manufacture are not limited to any particular method of making the same.

I can also use a water-soluble salt of radium or other radio-active element. For example, I can make an aqueous solution of radium nitrate and silver nitrate, and evaporate the water, thus producing a very fine and uniform mixture of the radium nitrate and silver nitrate. This mixture is dried and applied adherently to a silver carrier, and the silver nitrate is then decomposed by heating in hydrogen to metallic silver, without decomposing the radium nitrate. This surface layer is optionally wholly protected by a thin surface layer of gold or the like, which is applied very quickly, so as to minimize any loss of radium nitrate, in the gold-plating or gold-immersion bath. The gold layer can be applied in known manner, without plating.

Numerous changes and omissions and additions can be made to the illustrative disclosure herein,

without departing from the scope of this invention.

The carrier 1 may be of any thickness. However, I prefer that the carrier 1 should be sufficiently thick, so that the rays of the radio-active element do not pass through said carrier, and such rays pass only through the layer 2. The carrier 1 may be bendable or rigid. The proportion of the auxiliary metal or metals to the salt or other compound of the radio-active element may be varied as desired. I prefer to use an excess of silver, copper, etc., in the surface layer, in order to provide a metallic matrix in which the radio-active material is firmly held. In the completed article, the particles of silver, copper, etc., in the layer 2, are firmly united to each other to form a coherent metallic matrix, which holds the particles of the compound of the radio-active element. The invention includes the layer 2 per se, independently of a carrier.

The co-precipitate may consist of salts of the same acid. Thus, radium carbonate and silver carbonate are salts of carbonic acid. However, the co-precipitate, as previously noted, may be a mixture of copper oxide and radium carbonate before the co-precipitate is treated to decompose the copper oxide to metallic copper. The temperature at which the silver carbonate or copper oxide or the like is decomposed, is sufficiently high to sinter the particles of silver, copper, etc. to each other, in addition to sintering said metallic particles to the carrier 1.

Of course, the layer 2 may have a plurality of compounds of the same radio-active element or of different radio-active elements.

It is to be understood that the term "heat-resistant carrier" is used in the appended claims to denote a carrier which is adapted to carry, bonded to its surface, a coherent layer of the above described type formed by particles of a first metal and the water-insoluble salt of a radio-active metal, said salt being intermixed with the particles of said first metal, said carrier being substantially not affected by heating to decomposition temperatures of water-insoluble salts of said first metal, used in the process of this invention, and consisting preferably of a metal or an alloy.

Furthermore, it is to be understood that the terms "auxiliary metal" and/or "auxiliary carrier metal" are used in the appended claims to denote a metal or metals other than radioactive metals, said metal or metals forming water-soluble salts which can be precipitated from aqueous solutions together with salts of radio-active metals to form water-insoluble precipitates which can be decomposed to the metallic state at elevated temperatures below the decomposition temperature of the radioactive precipitates and below the melting point of the auxiliary carrier metal and below the melting point of the heat resistant carrier used in the process of my invention.

I claim:

1. A method of making a silver carrier which has an external adherent layer, said layer being a mixture of metallic silver and radium carbonate, the mass of the silver being greater than the mass of the radium in said radium carbonate, which consists in co-precipitating silver carbonate and radium carbonate from an aqueous solution of silver nitrate and radium nitrate by means of a water-soluble carbonate, the silver in said silver nitrate and the radium in said radium carbonate being in the desired final proportion in said layer, separating said co-precipitate from the mother liquid, drying said separated co-precipitate, mois-

tening said dry co-precipitate with a fluid to form a paste, said fluid being volatile at a temperature at which silver carbonate is decomposed to metallic silver, applying said paste adherently to a respective surface of said silver carrier, heating said carrier and said paste to said temperature in order to produce metallic silver in situ from said silver carbonate and to bond such produced silver to said silver carrier, said temperature being below the temperature at which radium carbonate is decomposed.

2. A method of providing a heat-resistant metallic carrier with a layer comprising particles of an auxiliary carrier metal selected from the group consisting of silver and copper, said particles being intermixed with a water-insoluble salt of a radio-active metal, which consists in dissolving a water-soluble salt of said radio-active metal and a water-soluble salt of said auxiliary carrier metal, in water, adding a precipitant to the aqueous solution thus formed to produce a co-precipitate of a water-insoluble salt of said radio-active metal and a water-insoluble salt of said auxiliary carrier metal, the decomposition temperature to metal of said water-insoluble salt of the auxiliary carrier metal being lower than the melting temperature of the auxiliary carrier metal and the melting temperature of the heat-resistant carrier and lower than the decomposition temperature of the water-insoluble salt of the radio-active metal, applying a surface layer of said co-precipitate to said heat resistant carrier, heating said carrier and the surface layer applied thereto to convert the water-insoluble salt of the auxiliary carrier metal into this metal and bond the resultant layer to the surface of the heat resistant carrier.

3. A coherent residue of a co-precipitate of a water-insoluble salt of a radio-active metal and a water-insoluble salt of an auxiliary carrier metal selected from the group consisting of silver and copper, said water-insoluble salt of said auxiliary carrier metal being decomposable by heat to produce the auxiliary carrier metal at a temperature which is below the melting point of said auxiliary carrier metal and below the decomposition temperature of said water-insoluble salt of said radio-active metal, said residue comprising particles of said water-insoluble salt of said radio-active metal intermixed with particles of said auxiliary carrier metal, said particles of said auxiliary carrier being the residue of the heat-decomposed water-insoluble salt of said auxiliary carrier metal, said intermixed particles of said water-insoluble salt of said radio-active metal and of said auxiliary carrier metal being heat-bonded to each other.

GEORGE JORIS.

REFERENCES CITED

The following references are of record in the file of this patent:

UNITED STATES PATENTS

Number	Name	Date
1,059,531	Ebler	Aug. 22, 1913
1,459,400	Hocker	June 19, 1923
1,505,209	Leach	Aug. 19, 1924
70 1,645,599	Jones	Oct. 18, 1927
1,655,184	Hahn	Mar. 3, 1928
1,670,483	Rentschler	May 22, 1928
1,956,948	Fattinger	May 1, 1934
2,326,631	Fischer	Aug. 10, 1943
75 2,333,710	Deimel	Nov. 9, 1943