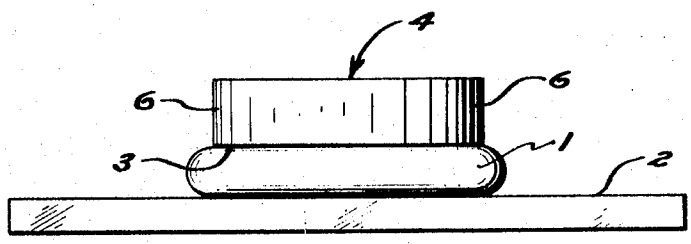
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METHODS OF MAKING SOURCES OF RADIOACTIVE ENERGY

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METHODS OF MAKING SOURCES OF RADIOACTIVE ENERGY

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ABSTRACT OF THE DISCLOSURE

In the manufacture of sources of radioactive energy, distribution of radioactive material within the source is controlled by the steps of (1) introducing a solution of non-radioactive solute into the matrix, and/or coating lateral exterior surfaces of the matrix with non-radioactive material, (2) contacting less than the entire matrix external surface with a solution containing a quantity of radioactive material, substantially all of which is capable of being introduced into the matrix prior to sealing the pores of the matrix by the application of heat.

BACKGROUND OF THE INVENTION

Field of the invention

This invention relates to improved methods of making sources of radioactive energy. More particularly, the invention relates to improvements in methods of introducing radioactive materials into the porous matrices to form sources of radioactive energy.

The term "source" is used to connote a usable combination of radioactive material within a matrix rather than the radioactive material per se. The sources are used for irradiation of various materials for a variety of well-known industrial and therapeutic purposes.

Description of the prior art

Such sources have been prepared by contacting matrices having a plurality of interconnecting pores or channels with a solution of radioactive material which is forced or drawn into the matrix by pressure and/or the capillary action of the pores. The solvent for the radioactive material is evaporated from the matrix which is then heated to collapse or shrink the pores thereby entraping the radioactive material. Such methods are described, for example, in U.S. patent application Ser. No. 392,312, filed Aug. 26, 1964, and now U.S. Patent No. 3,364,148, copending herewith and having a common assignee.

It has, hitherto, been the practice to totally immerse the matrix in the solution in order to obtain a uniform distribution of radioactive material. Merely contacting a portion of the matrix with solution has generally resulted in irregular distribution patterns. Uniform distribution of the radioactive material is required to provide uniform intensity of radiation across the source radiation face, e.g. the face directed at the target to be irradiated. In applications requiring controlled dosage to all areas of the target, as, for example, in therapeutic irradiation of animal tissues, uniform radiation intensity is critical.

Although uniform distribution can be obtained by immersion techniques, prolonged periods of immersion, presumably to allow establishment of equilibrium conditions, are often required. This, coupled with the relatively large volume of solution required for immersion is often uneconomical, and requires stringent measures to protect manufacturing personnel from over-exposure to radiation.

It should be noted that the immersion technique does not completely ensure uniform distribution since the formation of air pockets may exclude solution from portions of the matrix.

Incorporation of precise quantities of radioactive material into a matrix by immersion techniques is time-consuming and tedious. It is necessary to immerse the matrix in solution, remove the matrix and determine the quantity of material introduced and repeat this procedure until the desired concentration is obtained.

Furthermore, the quantity of radioactive material which can be introduced by the immersion technique is limited by the solubility of the radioactive material.

SUMMARY

It is an object of this invention to provide improved methods of making sources of radioactive energy. More specifically it is an object of this invention to provide material is obtained, increased quantities of radioactive methods whereby controlled distribution of radioactive material can readily be incorporated into the matrix, and exposure of manufacturing personnel to radiation is reduced.

Basically, these objectives are accomplished by applying a non-radioactive material to matrix surfaces and limiting the area of matrix external surface contacted by the solution. The methods, as well as other objects and advantages of the invention, will be more clearly understood from the drawings and the following description.

DESCRIPTION OF THE DRAWING

The figure schematically illustrates an arrangement of apparatus and materials utilized in preparing a source of radioactive energy according to an embodiment of this invention.

DESCRIPTION OF THE INVENTION

This invention can be practiced with porous matrices formed of polymers, metals, glasses or similar materials, which can be melted or deformed to seal the pores by application of heat. For example, the matrix can be of the type comprising a mass of sintered particles having a porous structure resulting from interstitial spaces between the particles or of the type produced by volatilizing, decomposing or leaching out particular constituents of various materials. It is desirable that the matrix material be strong, insoluble, and resistant to degradation induced by radiation. Porous matrices prepared by leaching insoluble constituents from various glasses are very satisfactory. Such matrices having silica contents in excess of 92% by weight are highly resistant to chemical attack and thermal shock and are, therefore, especially preferred. Methods of making such porous glass are described, for example, in U.S. Patent No. 2,196,744 and Corning Glass No. 7930 is representative of commercially available porous glass of this type. Such matrices are capable of chemically and/or physically adsorbing radioactive materials on both external and internal (pore wall) surfaces and of retaining such materials within the internal pore volume. Unless otherwise indicated, the term "matrix" surfaces" is used herein to include both internal and external surfaces.

It has been discovered that predetermined quantities of radioactive material can be conveniently introduced into porous matrices by effecting contact between a volume of a solution of radioactive material and an external surface area of the matrix which is less than the entire external surface area. This can be done, for example, in the manner illustrated in the figure. A volume of solution 1, containing a predetermined quantity of radioactive solute is placed on a substantially non-porous surface 2. An external surface 3 of a porous matrix 4 is placed in contact with the solution. The matrix must, of course, have a capacity sufficient to accommodate all of the predetermined quantity of radioactive material. If the volume of solution placed on the non-porous surface has a convex

upper surface due to surface tension, as for example, in the case of a "drop" of solution, the area of contact between solution and matrix can be readily controlled by adjusting contact pressure to flatten and spread the upper liquid surface to the desired extent. Contact between matrix and solution is maintained until substantially all of the solution is introduced into the matrix by the pressure of the solution against the matrix and/or the capillary action of the matrix pores.

While introducing the solution, it is desirable to expose the matrix to heat, low humidity, or other conditions effective to promote evaporation of the solvent for the radioactive material from the matrix. Promoting solvent evaporation from the matrix increases the rate of solution introduction and permits the use of solution volumes in excess of the matrix pore volume. When this technique is utilized, the amount of radioactive material which can be introduced in one matrix-solution contact step is not limited by the solubility of the radioactive material since the continuous evaporation of solvent prevents establishment of equilibrium in the matrix.

The method of this invention, as described thus far, permits the introduction of exact, predetermined amounts of radioactive material into the matrix, reduces the quantity of solution of radioactive material to which manufacturing personnel are exposed, and minimizes the duration of such exposure. However uniform or controlled distribution of radioactive material within the matrix is not readily obtained merely by use of the steps described above. For example, it has been found that these steps, alone, often result in sources having unduly high concentrations of radioactive material at the perimeter of the radiation face or irregular concentration patterns within the matrix. The reasons for such variations in distribution are not completely understood but may be due, at least in part, to variations in surface adsorptivity characteristics throughout the matrix and to progressive dilution of the solution via adsorptive removal of radioactive material from the solution as it pentrates into the matrix.

It has been found that applying a non-radioactive material to matrix surfaces prior to introduction of the solution of radioactive material provides uniform distribution of radioactive material.

In one embodiment of this invention lateral surfaces 6 of the matrix are coated with a non-radioactive material. The non-radioactive coating can be an absorbed layer of a non-radioactive isotype of the radioactive material or other material capable of being absorbed on the lateral surface. Alternatively, or additionally the lateral surfaces can be coated with lacquers, paints, greases, waxes, dyes, polymers or other well-known coating materials.

In a second embodiment of the invention, a volume of a solution of non-radioactive solute is introduced into the porous matrix. Preferably this is done according to the previously described procedure for introduction of radioactive material to ensure that the walls of the pores into which the radioactive solution is subsequently introduced have been previously contacted with the non-radioactive solute. The non-radioactive solute is chosen for its ability to be deposited or adsorbed on matrix surfaces thereby inhibiting adsorption of radioactive material thereon. In addition, the non-radioactive material should not enter into adverse chemical reactions with the matrix or radioactive material. Such factors as cost, toxicity, and the like may also be considered in selecting an appropriate material. Generally, a non-radioactive isotope of the radioactive material or a material selected from the same periodic group as the radioactive material proves most satisfactory.

If desired, the solvent for the non-radioactive material can be substantially completely evaporated from the matrix prior to contacting the matrix with the solution of radioactive material so that the latter solution will diffuse into the matrix more rapidly.

Preferably, the solvents used for both radioactive and non-radioactive solutes will be volatile in order to facilitate rapid solvent removal from the matrix during processing or prior to sealing. For example, water or low boiling organic solvents can be utilized.

In a third embodiment, the above described matrix pretreatments are combined. The order in which the pretreatments are applied is not critical. That is, the lateral surface coating can be applied either before or after introduction of the solution of non-radioactive material into the matrix.

Each of the embodiments described has been found to provide sources having substantially uniform radiation intensity across the radiation face. The mechanisms providing the desired distribution of radioactive material are not fully understood. However, the results may be due, at least in part, to inhibition or modification of matrix surface affinity for radioactive material or to modification of flow paths of radioactive material into the matrix due to lateral pore blockage effected by application of non-radioactive coatings to the lateral edges.

The invention is further illustrated by the following examples.

Example 1

A disk of Corning 7930 glass (a porous glass comprising more than 92% silica by weight) measuring about 1.0 cm. in diameter and about 0.1 cm. thick was pretreated by coating the lateral edges with an aniline dye marking ink.

A 20 micro-liter drop of an aqueous solution of radioactive strontium chloride (82 mg./ml. concentration) was placed on the surface of a smooth tetrafluoroethylene polymer sheet.

The treated disk was placed on this drop with sufficient pressure to spread the drop into contact with substantially the entire bottom face of the disk. While in contact with the drop, the disk was exposed to radiation from a conventional infrared heat lamp to promote evaporation of solvent from the porous matrix. Substantially all of the solution of radioactive material was introduced into the porous disk within 15 minutes.

The disk was heated to about 1100° C. to shrink the pores thereby "sealing in" the radioactive material.

A radiograph of the source indicated substantially uniform radiation intensity across the radiation face.

Example 2

A 20 micro-liter drop of an aqueous solution of non-radio active strontium chloride (82 mg./ml. concentration) was placed on the surface of a smooth tetrafluoroethylene polymer sheet. A disk of Corning 7930 glass having the same dimensions as the disk used in Example 1 was placed on the drop with sufficient pressure to spread the drop into contact with the entire disk face. Heat was applied, as in Example 1, to promote evaporation of solvent from the matrix.

A 20 micro-liter drop of an aqueous solution of radioactive strontium chloride (82 mg./ml. concentration) was then introduced into the disk in the same manner.

The matrix was sealed by heating to about 1100° C. and the distribution of radioactive material determined by a radiograph. The radiation intensity across the radiation face was uniform.

Example 3

A source was prepared as in Example 2 except that the lateral edges of the disk were coated with marking ink after the introduction of the solution of non-radioactive strontium chloride and prior to the introduction of radioactive material.

The radiation intensity across the radiation face, as measured by radiographs, was uniform.

Although this invention has been described by reference to specific embodiments, numerous variations therein can

be made. For example, the matrix can be contacted with additional volumes of radioactive material to provide higher concentrations in the source and techniques of applying coatings to and introducing solutions of non-radioactive materials and radioactive materials into the matrix can be modified to meet manufacturing requirements. Other modifications within the scope of the claims will be apparent to those skilled in the art.

I claim:

1. In a method of making a source of radioactive energy by introducing a solution of radioactive material into a heat-sealable porous matrix, and heating the matrix to seal at least the peripheral pores thereof, the improvement comprising applying a non-radioactive material capable of being adsorbed or deposited on surfaces of the matrix to said surfaces prior to introducing the solution of radioactive material into said matrix and introducing said solution by

(a) effecting contact between an external surface area of said porous matrix less than the entire external surface area of said matrix and a volume of said solution containing a predetermined quantity of radioactive material, said matrix having a capacity for all of said radioactive material, and

(b) maintaining contact between said matrix and said solution until substantially all of said solution is introduced into said matrix.

2. The method of claim **1** further comprising exposing said matrix to means for increasing the rate of vaporization of the solvent in said solution from said matrix while said selected surface of said matrix is in contact with said solution.

3. The method of claim **1** wherein applying said non-radioactive material to surfaces of the matrix comprises coating lateral external surfaces of the matrix with a non-radioactive material.

4. The method of claim **1** wherein applying said non-radioactive material to surfaces of the matrix comprises introducing a solution of non-radioactive solute into the matrix.

5. The method of claim **4** wherein said solution of non-radioactive solute is introduced into said matrix by

(a) effecting contact between an external surface area of said matrix less than the entire external surface area of said matrix and a volume of said solution of non-radioactive solute, and

(b) maintaining contact between the solution and the matrix surface until substantially all of the solution is introduced into the matrix.

6. The method of claim **5** additionally comprising the step of coating lateral external surfaces of said matrix with said non-radioactive material.

7. The method of claim **6** further comprising exposing said matrix to means for increasing the rate of vaporization of the solvent in each of said solutions from the matrix while the matrix is in contact with each of said solutions.

8. The method of claim **7** wherein said matrix comprises at least 92% silica by weight.

9. The method of claim **8** wherein the non-radioactive solute is a non-radioactive isotope of the radioactive material.

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