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IODINE-132 GENERATOR AND SHIPPING CONTAINER

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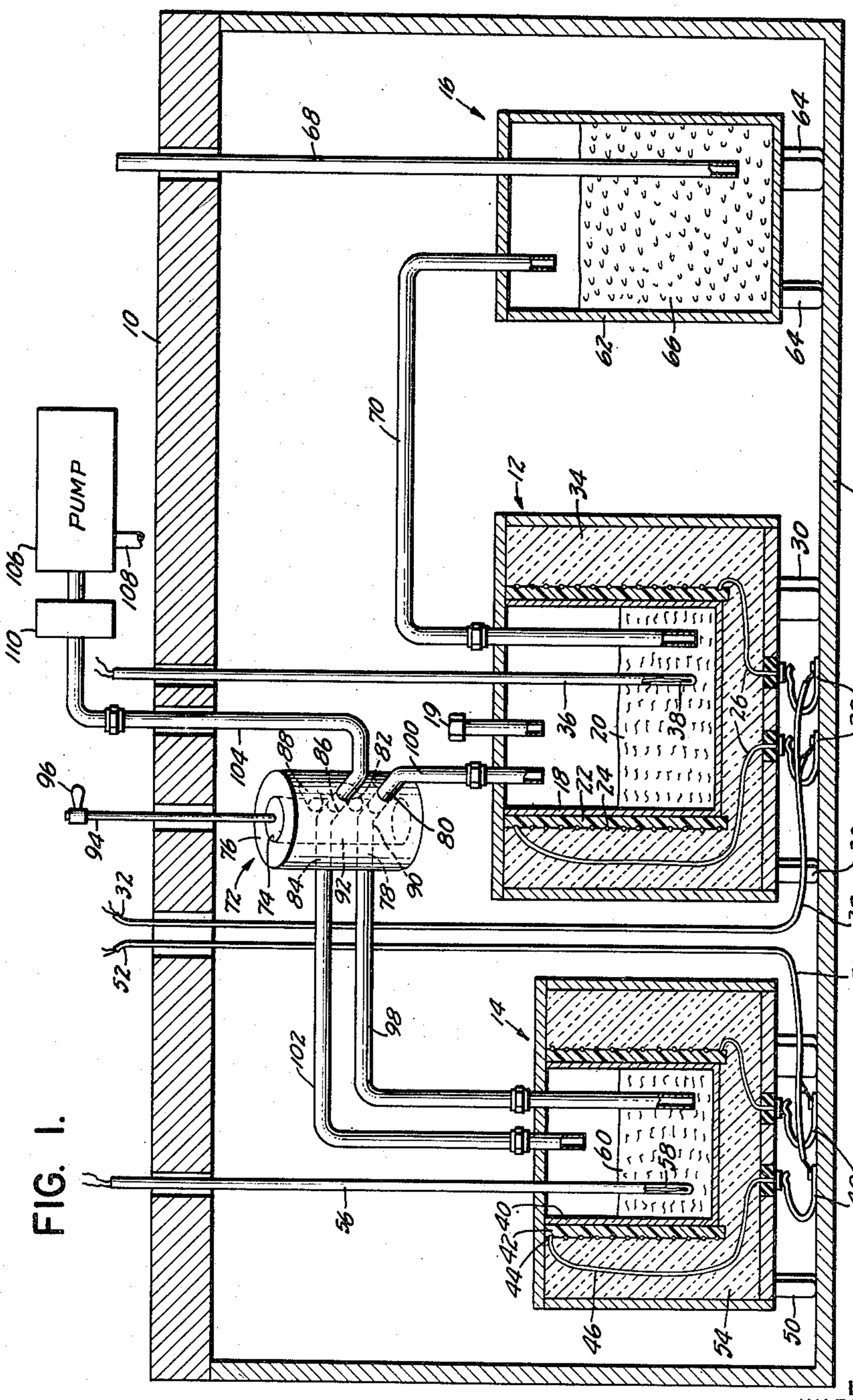

FIG. 1.

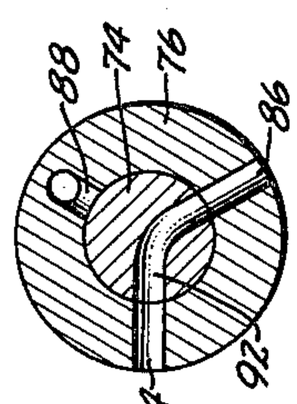

FIG. 2.

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IODINE-132 GENERATOR AND SHIPPING CONTAINER

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5 Claims. (Cl. 23—216)

This invention relates to the production of radioactive iodine. More particularly the invention relates to a novel source of iodine-132, a new method of recovering the radioactive iodine-132 in substantially pure form from this source, and apparatus particularly adapted to carry out the method of the invention.

In recent years extensive uses have been developed for radioactive iodine in the medical and biological fields, wherein it has been used as a tracer for investigative and diagnostic purposes and also, in particular cases as a therapeutic agent. Heretofore it has been customary to use iodine-131 for these purposes, both because of the ready availability of this isotope and because its half life of 8 days is in the range where the activity of the iodine remains at a useful level for a sufficient period of time to permit its shipment to relatively distant points of use. However for many clinical and therapeutic purposes a short-lived isotope can be used and in fact is more desirable than a long-lived isotope. For example, in the so-called Smith and Quimby method for determining blood circulation time, less than two minutes is required to make the determination. Also in certain known methods of locating brain tumors with di-iodo-fluorescein containing radioactive iodine the measurement of radiation must be made in the first two hours after the radioactive material has been injected, since the tumor starts to throw off the dye after that time.

In cases where the nature of the objective sought permits a short-lived isotope to be used, the use of such an isotope yields substantial advantages. Thus the isotope decays rapidly after it is no longer needed, and hence within a relatively short period of time a second test or treatment can be made. Also the amount of radiation received by the whole body is minimized, and so is that received by such organs as the kidneys through which the radioisotope must pass before being eliminated. A further advantage of the shorter-lived isotope resides in the fact that when such isotopes are used there is no problem of disposing of radioactive wastes eliminated by the body.

A study of the known radioactive isotopes indicates that iodine-132 with a half-life of 2.4 hours is well suited for many applications of the type outlined above. However, its half-life is so short that it cannot be packaged in the conventional manner and shipped to a considerable distance without losing an excessive proportion of its activity.

It is accordingly an object of the present invention to provide a novel source of iodine-132 and a novel method of packaging this source which permits effective use of iodine-132 from the source after the package has been shipped a considerable distance. It is another object of the invention to provide a container for the source as well as means for causing the iodine-132 to be delivered from the container or package in controllable amounts at the point of use. Other objects of the invention will be in part obvious and in part pointed out hereafter.

In general the objects of the present invention can be

achieved by dispersing tellurium-132, which is the radioactive parent of iodine-132, in a solid but readily liquefiable vehicle. The tellurium may be in either elemental form or in the form of a suitable tellurium compound. Tellurium-132, which has a half-life of 77 hours occurs in the fission products resulting from the irradiation of natural uranium in a nuclear reactor and can be readily separated from these fission products. Iodine-132 formed in the solid mixture of tellurium-132 and vehicle remains imprisoned therein until removed by the method described below. The solid mixture is the source of iodine-132 referred to above and is preferably packaged in a special container for shipment and storage, the container being so constructed as to permit the source to be "milked" at the point of use to obtain controllable quantities of iodine-132 therefrom. This "milking" can be carried out, for example, by heating the source to liquefy it and passing a stream of a suitable dry gas, such as air, through the liquid mixture to remove the iodine-132 therefrom.

The many objects and advantages of the present invention can best be understood and appreciated by reference to the accompanying drawing which illustrates container apparatus adapted to contain the iodine source of the present invention and capable of being used to carry out the method of the invention. In the drawing:

Figure 1 is a schematic vertical section through the container showing the iodine generator centrally located therein, flanked by an air drying chamber on the right and an absorbing chamber on the left, and also showing the piping interconnecting the chambers and a control valve for regulating flow through the chambers, and

Figure 2 is a horizontal section through the control valve of Figure 1 taken through the upper set of ports of the valve.

Referring to the drawing, the numeral **10** designates a container or casing within which are located an iodine generator **12**, an absorber **14**, and an air dryer **16**. The generator **12** includes an inner chamber **18** which contains a body **20** of the iodine source material referred to above. This material is preferably a solid solution of a suitable tellurium-132 compound, such as the dioxide, in a suitable readily fusible vehicle, such as, for example, a eutectic mixture of lithium and potassium chlorides. It can be conveniently charged into chamber **18** as an aqueous solution by removal of filling plug **19** at the top of the chamber, and converting to the desired solid form by driving off the water of solution by heat supplied by a heating element **24**.

The chamber **18** is surrounded by a sleeve **22** of insulating material that supports the electrical heating element **24**, the ends of which are connected by conductors **26** to the terminals **28**. As shown, generator **12** is supported and positioned by legs **30** which rest upon the bottom of casing **10**, and the terminals **28** are located in the space between the bottom of generator **12** and the bottom of casing **10**. Electrical energy to heat the element **24** can be supplied to terminals **28** from a suitable source (not shown) by the lead **32**. The space between chamber **18** and the outer casing of generator **12** is filled with insulation **34** to minimize heat loss from chamber **18**. It will be understood that the function of element **24** is to heat and thereby fuse the source material **20** when it is desired to withdraw radioactive iodine from the generator and that the element **24** is connected to a source of electrical energy only when it is desired to melt the material **20**. To measure the temperature of the source material a thermocouple well **36** containing a thermocouple **38** is provided, the lower end of which is embedded in the body **20** of source material and the upper end of which extends through the top of casing **10**.

The absorber **14** is similar to the generator **12** in construction in that it includes an inner chamber **40**, in-

sulating sleeve **42**, heating element **44**, conductors **46**, terminals **48**, legs **50**, lead **52**, insulation **54**, thermocouple well **56** and thermocouple **58**, all similar to the corresponding parts of generator **12**. However, in place of the source material **20** of chamber **18** the chamber **40** contains a body **60** of a suitable absorbing agent for iodine, such as for example, silver nitrate. As will be described more fully hereafter, the generator **12** should be purged before it is used as a source of iodine-132 and the function of absorber **14** is to absorb iodine delivered by generator **12** during this purging step.

The drier **16** comprises a casing **62** supported and positioned by legs **64** that rest on the bottom of causing **10**. The drier is charged with a suitable water-absorbing material **66** such as silica gel and is provided with a gas inlet tube **68**, the discharge end of which extends into the silica gel bed. The top of drier **16** is conected to generator **12** by a pipe **70**, the discharge end of which extends into the body **20** of source material.

In operation of the device a gas, for example atmospheric air, is drawn through pipe **68** into drier **16** wherein it is dried. The dry gas then flows through pipe **70** and bubbles through the molten mixture of tellurium-132 dioxide and alkali metal chlorides in such a way as to pick up iodine-132 contained in the mixture. Flow of iodine-containing gas from generator **12** is controlled by a valve **72** comprising a rotatable plug **74** and housing **76**. The housing **76** contains a series of three lower ports **78**, **80**, and **82** and a series of three upper ports **84**, **86**, and **88**. The plug **74** has a lower passage **90** and upper passage **92** adapted to interconnect adjacent ports as best indicated in Figure 2. Ports **82** and **88** are interconnected within housing **76**. The plug **74** is provided with an upwardly extending actuating rod **94** having at its upper end a handle **96** by means of which the valve plug can be turned.

As shown in Figure 1, port **78** of housing **76** is connected by a pipe **98** with a point within chamber **40** near the bottom of silver nitrate bed **60**; port **80** is connected to the top of chamber **18** by a pipe **100**; port **84** is connected to the top of chamber **40** by a pipe **102**; and port **86** is connected by a pipe **104** to an absorber **110** containing a suitable absorbing medium through which iodine-containing gas is pulled by a pump **106**, the iodine-free gas being discharged through pipe **108**.

It may be noted that the container **10** is adapted to perform the dual function of encasing the generator absorber and drier and also acting as a radiation shield to prevent escape of a harmful amount of radiation.

The manner in which the apparatus shown in the drawing can be used to carry out the method of the present invention should be largely apparent from the foregoing description. The chamber **18** is initially charged with a mixture of tellurium-132 or a tellurium-132 compound and a normally solid but readily liquefiable vehicle. The use of a solid mixture is advantageous since the radiation hazard that might be created by breakage of a package containing a radioactive liquid or gas can be largely avoided by shipment of the material in solid form. Also the solid vehicle serves to imprison the radioactive iodine-132 as it is formed by decay of the tellurium-132.

When it is desired to remove iodine-132 from the apparatus for use, the generator is first purged free from iodine-132. To accomplish this purging the heating elements **24** and **44** are energized to melt the contents of the generator and absorber, valve **72** is positioned to interconnect the generator and absorber, and a gas is drawn into the system through pipe **68** by pump **106**. The gas passes through the molten iodine source **20**, where it picks up iodine-132, and then through the molten body of silver nitrate wherein it is absorbed. Upon completion of the purging step the heating elements are de-energized, pump **106** shut off and the apparatus allowed to stand for a predetermined interval until the desired quantity of iodine-132 has accumulated. The iodine source is then remelted, valve **72** positioned to cause the iodine-bearing gas stream to by-pass the absorber, and gas is drawn through the molten source to remove iodine-132 therefrom for use.

From the known properties of the radioactive materials used and by means of known mathematical relationships it is possible to calculate quite precisely how long the apparatus should stand after completion of a purging step in order to build up in the generator a predetermined desired quantity of radio-iodine. For any given source the maximum build-up of iodine will occur about 12.4 hours after purging. If less than the maximum quantity of iodine is desired this lesser amount can be readily and accurately obtained by using a shorter time interval between purging and "milking" of the generator.

It is of course to be understood that the foregoing description is illustrative only. Thus the tellurium-132 may be incorporated in compounds other than the dioxide, such as one of the chlorides, bromides or iodides or tellurous acid. Also although the preferred vehicle material, as previously indicated, is an eutectic mixture of lithium and potassium chlorides, numerous other readily liquefiable vehicles e. g. zinc chloride, may be used and these may or may not be salt mixtures. When elemental tellurium is used it can be conveniently dispersed in ferric nitrate or elemental sulfur. Also tellurium in the form of telluric acid may be added to a hot gelatine solution to form a jelly. Any iodides formed from the generated iodine are oxidized to free iodine by an excess of the telluric acid. The jelly can be liquefied by heating as described above or by adding water thereto. The tellurium-132 used may or may not be associated with a non-radioactive vehicle such as natural tellurium.

It may be further noted that the silica gel in drier **16** can be replaced by other known drying agents, and the silver nitrate of absorber **14** can be replaced by other known iodine absorbing agents. Other modifications within the scope of the invention will be apparent to those skilled in the art.

We claim:

1. A method of producing a gaseous stream containing radioactive iodine which comprises forming a solid mixture comprising a liquefiable vehicle and tellurium-132, confining said mixture in a closed vessel to cause iodine-132 formed by decay of said Te^{132} to accumulate therein, liquefying said mixture in said vessel, and passing a dry gas inert to the vehicle containing the iodine through the molten mixture to remove iodine therefrom.

2. A method of producing a gaseous stream containing radioactive iodine which comprises forming a low melting point, solid mixture comprising tellurium-132 and a fusible vehicle, confining said mixture in a closed vessel to cause iodine-132 formed by decay of said tellurium-132 to accumulate therein, heating said mixture in said vessel to melt the same, and passing a dry gas inert to the vehicle containing the iodine through the molten mixture to remove iodine therefrom.

3. The method of producing a gaseous stream containing radioactive iodine which comprises forming a low melting point, solid mixture of a tellurium-132 compound and a fusible vehicle, confining said mixture in a closed vessel to cause iodine-132 formed by decay of said tellurium-132 to accumulate therein, heating said mixture in said vessel to melt it, passing a stream of dry air in sequence through the molten mixture and then through a bed of a substance capable of absorbing iodine from said stream for a period sufficient to purge said molten mixture of iodine, and thereafter continuously passing air through said molten mixture and conducting it to a point of use without passing it through said absorbent bed whereby said stream contains freshly formed iodine-132.

4. The method of producing a gaseous stream containing radioactive iodine which comprises forming a solid solution of tellurium-132 dioxide in a mixture of lithium and potassium chlorides, confining said solid solution in a closed vessel to cause iodine-132 formed by decay of said tellurium-132 to accumulate therein, heating said

solid solution in said vessel to melt it, and passing dry air through the molten solution to remove iodine-132 therefrom.

5. Apparatus for generating a radioactive vapor comprising in combination a casing; a generating chamber, a purging chamber and a gas drying chamber mounted in said casing, said generating chamber being interconnected with both said drying chamber and said purging chamber, both said generating chamber and said purging chamber having a container located therein for receiving a quantity of a solid material, each of said containers being provided with heating means for heating the solid contained therein; pump means connected to said chambers to produce a flow of gas through said drying chamber, generating chamber, and purging chamber in sequence to a discharge conduit; and valve means connected to said generator chamber, said purging chamber, and said discharge conduit and selectively movable to interconnect directly said generating chamber and discharge conduit to bypass said purging chamber.

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