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PROCESS FOR MAKING RADIOACTIVE IODINE

Geoffrey I. Gleason, Oak Ridge, Tenn., assignor to Abbott Laboratories, North Chicago, Ill., a corporation of Illinois

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This invention relates to a method of producing radioactive iodine^{131}, in particular, it relates to a method of producing said radioactive iodine by neutron bombardment of meta-telluric acid.

Radioactive iodine^{131}, hereinafter referred to as I^{131}, is an important tool in the hands of the clinician. Among its many uses may be mentioned its use as an index of basal metabolism and treatment of thyroid tumors. I^{131} is administered to the subject intravenously and is collected by the thyroid gland in accordance with the particular affinity of that organ for iodine. Activity of the thyroid gland is an index of basal metabolism. Such activity can be measured in terms of the amount of iodine which is attracted thereto. The tracer properties of radioactive iodine provide an evaluation of the degree of attraction to the thyroid gland by employing well-known methods of radioactive counting. This particular relationship of the thyroid and iodine also explains the mechanism whereby thyroid tumors can be treated by radioactive emissions after I^{131} has been deposited in the thyroid gland. Other adaptable practices of I^{131} are known or are actively being investigated.

The prior art provides two methods for producing I^{131}, both of which are characterized by serious disadvantages. One method provides production of I^{131} as a fission by-product following neutron bombardment of uranium in a reactor. This uranium by-product process results in a 2% to 3% fission yield of I^{131}, but the actual chemical yield is low due to processing and aging. The other well-known method employs metallic tellurium or tellurium oxide to produce I^{131}. The tellurium metal is a target material in a neutron flux reactor, and after a suitable period of irradiation, metallic tellurium, which is 35.4% tellurium^{130}, is converted to the radioactive isotope tellurium^{131}. This latter material is spontaneously converted to I^{131} following β-emission. While this method results in a higher recoverable yield than the fission by-product method, it is actually less desirable. This is attributed to the complex and difficult separation steps necessary to obtain I^{131} from the irradiated target material.

Ortho-telluric acid has recently been described as a target material for I^{131} production (R. Constant, J. Inorg. Nucl. Chem., 1958, vol. 7, pp. 133–139). This particular target provides fairly easy separation, but this particular acid has the disadvantages of low-temperature stability, small tellurium content and low specific density.

It is an object of this invention to provide a more efficient method for the production of I^{131}.

Another object of this invention is to provide a method whereby the production of I^{131} is characterized by higher yields.

A still further object of this invention is to provide a method of I^{131} production which is complemented by a simple separation procedure.

In the accomplishment of the foregoing objects and other objects which will be apparent hereinafter, it is now provided that meta-telluric acid comprises the target material in a high-flux neutron reactor. Meta-telluric acid, H_2TeO_4, has a density of 3.50 and a tellurium content of 66%. Meta-telluric acid is unstable above the elevated temperature of 160° C. By the term "unstable" is meant the loss of a mole of water from a mole of meta-telluric acid after the temperature of stability has been exceeded. This temperature of stability for meta-telluric

acid is high and comprises a decided advantage over ortho-telluric acid as will be more fully disclosed in a later portion.

The improved method substantially provides deposition of the target material in a capsule or equivalent container made of aluminum or other suitable materials. The term "target material" will be employed herein as a synonym for meta-telluric acid. The capsule is placed in a high-flux, low-temperature (less than 100° C.) reactor which may be air-cooled such as the Brookhaven reactor or water-cooled such as a heterogenous water-cooled medium testing reactor (MTR) type. A definite neutron flux is directed at the target material for a specified period of time. The neutron flux is expressed as the number of neutrons traversing a square centimeter of space per second (n./cm.^{2}/sec.). The method is successfully operated at varying neutron flux, but it is desirable to employ higher flux in order to obtain more I^{131} activity per gram of target material. The quantity concept relates to the radioactivity of I^{131} in units of curies (c.) or millicuries (mc.). The curie is defined as the quantity of any radioactive nuclide in which the number of disintegrations per second is 3.700×10^{10}.

The radioactive activity of I^{131} obtained by this method may be approximately calculated from the formula:

$$A=N\sigma f(i-e^{-\lambda t})$$

In the foregoing formula, A represents the activity of I^{131} in disintegrations/sec., N is the number of Te^{130} target atoms, σ is the capture probability in barns (10^{-24} cm.^{2}) for Te^{130} per each gram of elemental tellurium irradiated, t represents the irradiation time in days, f represents the neutron flux or n./cm.^{2}/sec. and λ is the decay constant of the I^{131} (days^{-1}). According to this formula, one gram of elemental tellurium in meta-telluric acid will provide the following maximum (or saturation) activities for each given flux:

Flux	Approximate Saturation Yield
10^{11} n./cm.^{2}/sec.	1.0 mc./gm.Te.
10^{12} n./cm.^{2}/sec.	10.0 mc./gm.Te.
10^{13} n./cm.^{2}/sec.	100.0 mc./gm.Te.
10^{14} n./cm.^{2}/sec.	1,000.0 mc./gm.Te or 1 curie.

After meta-telluric acid has been exposed to the prescribed irradiation conditions, I^{131} is easily separated therefrom by dissolving the target material in distilled water and adding thereto a strong mineral acid such as sulfuric, phosphoric and the like.

A preferred practice provides the addition of a carrier to the solution. A carrier such as potassium iodide is added in an amount of 50–100 micrograms. This amount of stable iodide is sufficient to act as a carrier for the radioactive I^{131} atoms formed in the process.

The solution is distilled and the formed iodine^{131} can be converted from the ionic state to the molecular state or I^- to I_2. It is also provided in alternative practice that the iodine ions are passed into a container with a solution of sodium sulfite, thereby obtaining I^{131} in the sodium iodide form. This separation procedure is obtained with facility because the target material is soluble in water and is a mild oxidizing agent.

In the practice of this process, it is provided that a high-flux, low-temperature reactor can be utilized, thus, providing a higher neutron flux without incurring difficulties from the development of excessively high temperatures. The presence of high temperatures beyond the workable limits of the process would result in decomposition of meta-telluric acid or undesirable vaporization pressures arising from liberated water. This is a limiting disadvantage for a capsule or a container of a given volume. As described hereinbefore, meta-telluric acid is

unstable at temperatures in excess of 160° C. Such excessive temperatures should result in liberation of water in meta-telluric acid in vapor form. The build-up of vapor pressure would result in rupturing the capsule containing the target material. This is an occurrence which obviously should be avoided and it is very difficult to avoid with ortho-tellurium acid which has a low-temperature stability of 90° C. It is, therefore, apparent that the high-temperature stability of meta-telluric acid comprises an advantage in the use of this target material. Cooling the reactor with water will allow use of a high-flux rate and control of temperature conditions at a level below the decomposition temperature. Such desirable high-flux rates can be attempted in a low-temperature, high-flux, air-cooled reactor without leading to temperatures which would decompose the target material.

The process is practiced to advantage over ortho-telluric acid because meta-telluric acid has a higher tellurium content which amounts to 66%. This allows a large amount of tellurium in a given capsule. The use of meta-telluric acid is also superior to ortho-telluric acid because meta-telluric acid has a higher density of 3.50. This provides a good amount of tellurium target in a relatively small target volume. Such features provide economical advantages for neutron bombardment in a reactor.

Embodiments of the process are presented in the following illustrations, but it is intended that such illustrations be considered only as a teaching rather than an exclusive practice.

Example I

An aluminum capsule measuring one inch in diameter and three inches in length is filled with 100 gms. of meta-telluric acid. The capsule is placed in a water-cooled reactor of the MTR type affording 10^{14} n./cm.^{2}/sec. The temperature of the target material is maintained below 160° C. The irradiation period is continued for 16 days. The target material is transferred from the aluminum capsule to a mixture comprising 1500 ml. of distilled water, 100 ml. of concentrated sulfuric acid and 100 micrograms of potassium iodide. The mixture is present in an enclosed flask to which is attached distilling apparatus. The solution is distilled under application of heat. The distillate is collected in a receiver containing 10 ml. of Na_2SO_3 solution. Distillation is continued until a volume of 20 ml. has been collected. This is a recoverable yield of 36–40 curies I^{131}.

The distillation steps are not characterized by any critical features. Conventional procedures well known in the art of distillations are operable. It is desirable to perform the distillation in an acid medium to obtain the best results; any strong mineral acid which is non-volatile and is not carried away by the steam distillate is useful. Various aqueous-acid mixtures may be employed, but the preferred mixture comprises about a 6% solution of acid in water.

Example II

In an aluminum capsule is placed 1.52 gms. of meta-telluric acid and the capsule with target is irradiated at 1.65×10^{13} n./cm.^{2}/sec. for 11.7 days in the north face of the Brookhaven reactor (air-cooled).

The target material is transferred to a distilling flask containing 15 ml. of 1 N H_2SO_4 plus 100 micrograms of potassium iodide carrier. The mixture is warmed until the target dissolves and is then distilled with a nitrogen purge. The distillate is collected to a volume of 3 ml. in a trap containing 3 ml. of Na_2SO_3 solution (2 mg./ml.)

The obtained yield is 90 mc. I^{131} at pile-out time. Examining the bottoms of the distilling apparatus shows that no emissions due to I^{131} occur. This indicates the recovery is quantitative.

I claim:

1. A method for making radioactive iodine^{131} which comprises irradiating meta-telluric acid with a neutron flux in a high flux, low temperature reactor, irradiating said meta-telluric acid at a reactor temperature not in excess of 160° C., and separating iodine^{131} from the irradiated meta-telluric acid.

2. A method for producing radioactive iodine^{131} which comprises irradiating meta-telluric acid with a neutron flux in a high-flux, low-temperature, air-cooled reactor, irradiating said meta-telluric acid at a reactor temperature not in excess of 160° C., dissolving the irradiated meta-telluric acid in water made acidic by a strong, non-volatile mineral acid and separating I^{131} by distillation.

3. A method for producing radioactive iodine^{131} which comprises irradiating meta-telluric acid with a neutron flux in a water-cooled reactor, irradiating said meta-telluric acid at a reactor temperature not in excess of 160° C., dissolving the irradiated meta-telluric acid in water made acidic by a strong, non-volatile mineral acid and separating iodine^{131} by distillation.

4. A method according to claim 3 where the irradiated meta-telluric acid is dissolved in an aqueous medium containing about 6% of a strong, non-volatile mineral acid.

5. A method for making radioactive iodine^{131} which comprises irradiating about 100 gms. of meta-telluric acid with a neutron flux of 10^{14} n./cm.^{2}/sec. in a water-cooled reactor for about 16 days at a reactor temperature not in excess of 160° C., dissolving the irradiated meta-telluric acid in about 1500 ml. of a water solution containing about 6% concentrated sulfuric acid and separating iodine^{131} by distillation.

6. A method for making radioactive iodine^{131} which comprises irradiating about 1.5 gms. of meta-telluric acid at a neutron flux of about 1.65×10^{13} n./cm.^{2}/sec. in a high-flux, low-temperature, air-cooled reactor for about 12 days at a reactor temperature not in excess of 160° C., dissolving the target material in about 15 ml. of 1 N H_2SO_4, adding thereto between 50 and 100 micrograms of a carrier and separating I^{131} from the carrier by distillation.

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