

Methods of Production of
Phosphorus-32, Sulfur-35 and Iodine-131

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Summary

This paper presents a review of methods of production of phosphorus-32, sulfur-35 and iodine-131 which have been developed in many countries. In addition to this review the method of production of each radioisotope adopted in Japan and our present status of the production are described.

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リン-32, イオウ-35 およびヨウ素-131の製造法

要 旨

この報告は欧米諸国で開発されたP-32, S-35およびI-131の製造法をレビューしたもので、これら放射性同位体の製造に際して種々の方法のなかから適当な方法を選択するための参考となるように、それぞれの方法の概略を述べ、長所・短所を明らかにした。また、日本原子力研究所におけるこれら放射性同位体製造の現状についても併記した。

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INTRODUCTION

The radionuclides P-32, S-35 and I-131 are now produced in curie amount in several countries. According to the kind of the target materials and the processing method after the irradiation, many different methods have been developed for the production of these radionuclides.

Which method should be adopted for the practical production of these radionuclides, it may be decided depending on the amount of the radionuclide to be produced or the condition of the irradiation in a given nuclear reactor.

Intending to serve for the choice of the production method, the authors have reviewed the production methods of P-32, S-35 and I-131 along the history of the development and clarified their advantages and disadvantages.

1. METHODS OF PRODUCING P-32

To produce P-32 in reactor, one of two possible methods is to utilize the $^{31}\text{P}(n,\gamma)^{32}\text{P}$ reaction by irradiating red phosphorus with thermal neutrons, and the other is to utilize the $^{32}\text{S}(n,p)^{32}\text{P}$ reaction on sulfur with fast neutrons.

The former (n, γ) method is simpler but the P-32 produced is of low specific activity, and a neutron flux of 1×10^{13} n/cm²/sec will only yield about 1,000 mc/g at saturation. Products of higher specific activity are often demanded. The latter (n,p) method has the limitation of requiring a fast neutron flux which can only be obtained by placing the target very close to the fuel elements, but it will produce carrier-free P-32 satisfying the requirement for high specific activity.

There is strong demand for high specific activity P-32, and production is undertaken with large targets of the order of 100 g or even kilograms. Elementary sulfur is used as target material. The different methods of separating P-32 from sulfur may be roughly classified into the three types described hereafter.

1.1 Liquid-Liquid Extraction

This method was first published in 1946 in the U.S. as a process for preparing curie amounts of P-32.^{P-1),P-2)} The irradiated sulfur is heated in concentrated nitric acid. The boiling point of concentrated nitric acid (120°C) is higher than the melting point of sulfur (113°C), so that the sulfur melts in boiling nitric acid and is mixed by the boiling bubbles. The P-32 is extracted into the nitric acid as ortho-phosphoric acid. Part of the sulfur target is oxidized into sulfuric acid and extracted into the nitric acid; then the P-32 is coprecipitated with iron hydroxide by adding iron to separate it from the sulfuric acid; then the iron is separated by extraction with isopropyl ether. Coprecipitation of P-32 with iron hydroxide is described in detail in a separate report.^{P-3)}

The above method was improved in 1947 by replacing concentrated nitric acid by 0.2 M nitric acid and pressurizing to melt sulfur at the same temperature as before.^{P-4)} The use of dilute nitric acid reduced the oxidation of sulfur, but iron from the stainless steel autoclave required for pressurization dissolved into the extract, and the product had to be purified. This method of extraction under pressure was practiced for several years in U.S., U.K. and France.^{P-5)~P-11)} In the U.K. an improvement was introduced by using an inner vessel made of glass in the autoclave to prevent attack of nitric acid on the stainless steel, and by rocking the autoclave. These improvements facilitated better mixture of the two phases and increased yield from 40 to 70% as compared to the original method using concentrated nitric acid.

They also replaced the iron hydroxide by lanthanum hydroxide for the coprecipitation process. P-5) ~ P-8)

Other processes devised in the liquid-liquid extraction category include fusion extraction by heating irradiated sulfur with a mixture of acetic acid and acetic anhydride, and extraction with acid or alkali from the dissolved sulfur in an organic solvent such as carbon disulfide or bromoform.

The acetic acid method uses the addition of acetic anhydride as a means of raising the boiling point. After the extraction the mixture is evaporated and dried to crystallize the small amount of extracted sulfur; P-32 is then leached into an inorganic acid. P-12) The product is finally purified by coprecipitation with iron hydroxide. P-3)

Other methods using organic solvent include a process of extracting P-32 with nitric acid or ammonia from carbon disulfide solution of the irradiated sulfur. P-13) In this method, the extract is first evaporated and dried, then ammonium molybdate is added to 0.5 N hydrochloric acid solution; P-32 is extracted into butyl acetate, and back extracted into water; and finally purified by anion exchange resin. An alternative method of purifying the extract from carbon disulfide solution with anion and cation exchange resins has also been discussed.

Organic extraction other than with carbon disulfide, such as with trichloroethylene, chloroform, benzene and aniline, has also been reported. P-14) The report compares these solvents for extraction ratio and ease of solvent handling, and concludes that trichloroethylene should be adopted.

1.2 Liquid-Solid Extraction

This method was developed in Norway in 1955. P-15) ~ P-19) A fine powder of sulfur is irradiated and the P-32 extracted with boiling water. Sulfur transforms from rhombic system to monoclinic at 95.5°C, and P-32 is released from the crystal during this transformation. The extraction is accelerated by adding a small amount of 2-octanol as wetting agent. The 2-octanol is easily eliminated by distillation after extraction, but tends to dissolve a small amount of sulfur which must be removed from the extract by purification.

To save this trouble, instead of using wetting agent, a method of extracting P-32 by violently stirring the aqueous suspension of sulfur with high frequency agitator has also been devised. P-20) It is reported that 15 minutes of agitation produced a yield of 75%.

1.3 Distillation

A distillation method developed in U.K. in 1954^{P-21)} is to place the irradiated sulfur in a quartz vessel and to distill the sulfur at 500°C in nitrogen flow. The involatile P-32 remains in the vessel, and after thus eliminating the sulfur the residue is extracted with dilute hydrochloric acid containing a small amount of hydrogen peroxide. The product does not require purification, and more than 98% of the P-32 is recovered in the form of orthophosphoric acid. Yield is between 80 and 100%. This method is now adopted in many countries, and several reports^{P-20)~P-22)} have discussed details of operation procedures.

The distillation method requires a very pure target since involatile impurities found in the sulfur remains in the distilling vessel. Purification of the target is done by the following method.^{P-20)}

Sulfur powder is boiled in 2 N hydrochloric acid for three hours, during which the acid is changed every hour; it is then washed with water. This eliminates heavy metal impurities but leaves organics which must be further removed. This is done by melting in a flask, maintaining at 150°C for six hours during which a small amount of nitric acid is added and agitated every 15 minutes, and finally washed with water after cooling. This purification is not complete for hydrocarbons, and carbon remains in the sulfur after distillation. The hydrocarbons are removed by extraction with carbon tetrachloride for two days. The sulfur thus purified is reported to contain less than 0.2 ppm of heavy metals and only 1 ppm of hydrocarbons.

Another paper^{P-22)} reports purification with distillation repeated four times as in the case of irradiated sulfur distillation.

Distillation has a number of advantages over other methods, apart from a yield of 80-100%, which is higher than other processes: the procedure is simple and does not require complicated purification as in the case of extraction. Time required for the process is one day as compared to four days for liquid-liquid extraction. It also permits the increase of specific radioactivity by repeating the process of sulfur distillation and irradiation. The Oak Ridge National Laboratory has succeeded in increasing specific activity by 1 to 2 x 10³ by this repetition.

Liquid-solid extraction is simple as procedure but the use of fine powder of sulfur as target material largely increases the target volume. Furthermore, the method cannot be adopted when reactor temperature exceeds sulfur melting point. The distillation method is more convenient also in these respects.

In our Institute too, we have been using the distillation method for producing P-32. One hundred grams of sulfur is

irradiated for 15 hours in the JRR-1 core, which produces 80 mc of P-32. The fast neutron flux is about 1×10^{12} n/cm²/sec. Using this size of target, we expect to cover the whole domestic demand of P-32 when irradiation in JRR-3 is commenced in the near future.

2. METHODS OF PRODUCING S-35

Sulfur-35 is usually produced by neutron irradiation on potassium chloride, utilizing the $^{35}\text{Cl}(n,p)^{35}\text{S}$ reaction. The $^{34}\text{S}(n,\gamma)^{35}\text{S}$ reaction can also be utilized, but it produces a product of low specific activity and is not practical.

Neutron irradiation on potassium chloride produces at the same time K-42 with half-life of 12.47 hr, Cl-38 of 37.5 min, Cl-36 of 3.2×10^5 yr besides P-32 of 14.30 days. Of these, K-42 and Cl-38 have relatively short half-lives and are eliminated by decay if the product is cooled for about a week after irradiation. The P-32 is produced by fast neutrons through the $^{35}\text{Cl}(n,\alpha)^{32}\text{P}$ reaction whereas the $^{35}\text{Cl}(n,p)^{35}\text{S}$ reaction is by thermal neutrons, so that the proportion between P-32 and S-35 differs according to the position of the target in the reactor. The fraction of P-32 can be largely reduced by placing the target away from core center to eliminate or facilitate the subsequent process of removing the P-32. S-1)

The simplest way of separating the S-35 from potassium chloride is by cation exchange resin: An aqueous solution of the irradiated potassium chloride is passed through a hydrogen form cation exchange resin to remove potassium. The effluent is evaporated to dryness to remove chlorine and Cl-36. S-2) ~ S-6) The S-35 is leached by dilute hydrochloric acid. It is obtained directly in the form of sulfuric acid without any oxidation of the product.

The S-35 produced by this procedure is adequately pure except for minute quantities of P-32, which must be separated when present in non-negligible amount. This is done by adsorption of the P-32 in a column of aluminum shavings before passing the potassium chloride solution through the ion exchange resin, S-3) by coprecipitation with iron hydroxide, S-4) or by evaporating the effluent from the ion exchange resin, leaching the S-35 and P-32 and adsorbing the P-32 in iron form cation exchange resin. S-6)

A method suited for treating large amounts of potassium chloride is to cool a saturated aqueous solution of irradiated potassium chloride with dry ice and to precipitate and eliminate most of the potassium chloride by passing hydrogen chloride gas. S-2), S-7)

In the cation exchange resin method, a strong acid type resin containing a sulfonic group is generally used, and the resin decomposes to form a product which tends to lower the specific activity of the S-35. It also necessitates the introduction of a separation process for P-32. To avoid these complications a simple method of producing highly specific active S-35 is reported

which utilizes anion exchange resin. S-8) A carrier of phosphate is added to a 0.1 M aqueous solution of irradiated potassium chloride and is passed through a chloride form anion exchange resin column. After washing the resin with water, P-32 and S-35 are successively eluted with 0.1 N hydrochloric acid. The disadvantage of the anion exchange method is that it is highly influenced by potassium chloride concentration, and does not permit treatment of highly concentrated solution. The method is thus unsuitable for processing large amounts of targets. A method has been developed in which ethanol is added to a hot saturated aqueous solution of irradiated potassium chloride to precipitate about 90% of the potassium chloride, and the S-35 is separated by anion exchange from the filtrate. S-9)

Some reports describe a simple process of producing S-35 with alumina column. S-10) ~S-12) Irradiated potassium chloride is dissolved in 0.5 N hydrochloric acid to give a 2 N solution, and passed through a column containing 1 to 2 g of alumina thoroughly washed with 0.5 N hydrochloric acid; Cl-36 is then eluted with 0.5 N hydrochloric acid, and S-35 with 0.2 N ammonia. P-32 remains in the column. The eluted S-35 is purified by cation exchange.

Another method is coprecipitation of S-35 from an aqueous solution of irradiated potassium chloride. S-13) Cl-36 is first separated by adding chromate and barium to let S-35 and P-32 coprecipitate with the resulting barium chromate. The filtrate is evaporated and the residue treated with sulfuric acid; then the hydrogen chloride evolved is absorbed in potassium hydroxide, and the resulting potassium chloride is used for the next irradiation. The process is repeated to concentrate the Cl-36. The barium chromate precipitate containing S-35 and P-32 is dissolved in hydrochloric acid; then the chromic acid is reduced with ethanol, and separated in the form of hydroxide. The precipitate contains S-35. After repeating the precipitation three times it is dissolved, and chromium separated by cation exchange.

We in the Institute have adopted the cation exchange method, and we use iron form cation exchange resin for separating P-32. A 30 g target of potassium chloride is irradiated in a flux of 2×10^{11} n/cm²/sec in JRR-1 for 15 hours, which produces about 4 mc of S-35. When we start using JRR-2 and JRR-3, we expect to obtain about 1 c with an irradiation of 10 days in a flux of 10^{13} n/cm²/sec.

3. METHODS OF PRODUCING I-131

The various methods of producing I-131 can be classified, according to the nuclear reaction utilized, form of target material and method of treatment, into the following categories.

Nuclear Reaction	Target	Treatment
$^{235}\text{U}(n, f)^{131}\text{I}$	Natural uranium	Distillation
$^{130}\text{Te}(n, r)^{131}\text{Te}$	Elementary tellurium	Distillation
$\beta^- ^{131}\text{I}$	Tellurium dioxide	Distillation
	"	Sublimation
	Telluric acid	Distillation
		Adsorption
		Column chromatography

Using tellurium as target material the saturation activity of I-131 produced per gram of tellurium irradiated in a neutron flux of 1×10^{12} n/cm²/sec is about 10 mc, while starting from U-235 the same irradiation will produce about 8 mc per gram of natural uranium. Neutron economy in the reactor would favor production by fission when curie amounts of I-131 are required. The latter method also has the advantage of yielding shortlived fission products such as Ba-140 at the same time. On the other hand, this process is accompanied by undesirable fission products which, with their high radioactivity, set back the advantages of the process by complicating the handling of the product. Another shortcoming is that the I-131 is distilled from a nitric acid solution of irradiated uranium so that the product tends to be contaminated with undesirable products such as nitric and nitrous acids as well as long-lived fission products like Sr-90 and Cs-137.

When in 1946 the Oak Ridge National Laboratory started to produce I-131 for distribution, they irradiated elementary tellurium and separated the I-131 by distillation. I-1) It was already anticipated at that time that the tellurium target method would eventually become inadequate to cope with the rapidly increasing demand for I-131. Supply of the product increased from 300 mc per month in 1946 to 30 c in 1950, and in 1948 ORNL changed to production by fission. About 1,000 c were produced by the latter method by December 1951. I-2)

3.1 Tellurium - Distillation

This was the method first adopted at Oak Ridge.^{I-3)} They irradiated elementary tellurium; then dissolved it in a mixture of chromium (VI) oxide and concentrated sulfuric acid, thus oxidizing tellurium into tellurate and I-131 into iodate. The I-131 was then reduced into elementary iodine by adding an excessive amount of oxalic acid; and finally distilled. This method involves the use of large amounts of reagents for dissolution and reduction, which tended to introduce small amounts of the reagents into the distillate during distillation, so that the distilled I-131 had to be redistilled for purification. This was done by acidifying the distillate with sulfuric acid; adding a small amount of potassium permanganate to decompose the oxalic acid; then adding oxalic acid to reduce the excess permanganate and iodate; and finally distilling I-131. The distillate was collected in a basic solution containing a reducing agent, and the I-131 fixed in the form of I⁻. Reports have described the equipment and procedure for producing I-131 in curie amounts by this method.^{I-4)~I-7)} Another report has discussed the oxidation, reduction, distillation and other processes related to I-131 production.^{I-8)}

The process of dissolving irradiated elementary tellurium in a mixture of chromic and concentrated sulfuric acids is violent, and the operation is not easy. To avoid this difficulty a method has been devised of using a target of telluric acid, which is soluble in water, and inorganic acids.

3.2 Telluric Acid - Distillation

In 1958, R. Constant published his method of dissolving irradiated telluric acid in sulfuric acid and distilling the I-131.^{I-9)} The adoption of telluric acid as target material permitted a product of high purity by a simplified procedure with a yield of 90%.

The chemical form of the I-131 as produced by the dissolution of irradiated telluric acid in sulfuric acid affects the efficiency of distillation. This has been discussed in relation to the mechanism of the distillation of I-131 from telluric acid solution.^{I-10)} At Saclay, elementary iodine is isolated from the telluric acid solution by the addition of a trace of potassium iodide and iodate to accelerate the process.^{I-11)}

3.3 Telluric Acid - Adsorption^{I-12)}

Irradiated telluric acid is dissolved in 1 N sulfuric acid in a platinum dish; agitated to absorb the I-131 on the dish; then returned to solution by electrolysis. A report discusses the treatment of large targets by this method.^{I-13)}

3.4 Telluric Acid - Column Chromatography I-14), I-15)

Irradiated telluric acid is dissolved in a small amount of water; and passed through a column filled with glass paper; alcohol is then passed through the column, which leaves the telluric acid in the form of fine precipitate and elutes I-131 alone.

Telluric acid undergoes dehydration and becomes insoluble above 90°C during irradiation in reactor, so that it cannot be used as target material in such cases. I-9) For this reason tellurium dioxide has come to be used as target material for its stability under heat and ease in handling as compared to elementary tellurium, as described below.

3.5 Tellurium Dioxide - Distillation I-16)

Irradiated tellurium dioxide is dissolved in 10% sodium hydroxide solution; acidified with sulfuric acid; then iron (III) sulfate added, and I-131 distilled under reduced pressure.

3.6 Tellurium Dioxide - Sublimation I-17), I-18)

Developed in Norway in 1954, this method is to irradiate tellurium dioxide powder; which is transferred to a quartz boat; heated in electric furnace to a temperature of between 680 and 700°C with pressure reduced to 2×10^{-2} mm Hg to sublime the I-131 as elementary iodine; which is then collected in a sodium hydrogen sulfite solution. It is reported that 90% of the I-131 was recovered by treating from 50 to 150 g of tellurium dioxide for 2 to 4 hours. This method has been improved, and a report describes equipment for performing the sublimation in air flow. I-19)

The sublimation technique is in operation in many countries, and reports have been published on production methods, I-20), I-21) and mechanism of I-131 release. I-22) Advantages of the method are simplicity of operation, high purity of the product, and possibility of increasing specific activity by repeated use of the tellurium dioxide target.

3.7 Uranium - Distillation I-2), I-23), I-24)

About 3.5 kg of irradiated uranium is dissolved in six liters of nitric acid; I-131 is then distilled by steam - air distillation; the distillate is treated with hydrogen peroxide to oxidize the nitrous acid; and finally the I-131 is redistilled. During redistillation the steam temperature is kept slightly above 95°C in order to distill only the I-131 and leave the nitric acid which has a low vapor pressure when dilute. The distillate is collected in a 10% sodium hydroxide solution and concentrated; then the reducible substances are decomposed by the addition of potassium permanganate. The excess permanganate and iodate are reduced with phosphorous acid in sulfuric acid solution; the I-131 is then

distilled; and finally collected in sodium hydroxide - sodium sulfite solution.

I-131 is mainly used in medicine for research and diagnosis, and high chemical and radiochemical purity is demanded. Methods of assay include filter paper electrophoresis for chemical form, I-25) and paper chromatography, evaporation and extraction for radioactive impurities. I-26)

We in the Institute are now comparing the telluric acid and tellurium dioxide methods of production. We have already completed studies with JRR-1, which is a relatively small reactor, and we are now proceeding with similar studies with JRR-2 in order to make a final choice.

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A review to cover production methods of the other radioisotopes is intended to be realized in near future.

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