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RECOVERY AND HALF-LIFE DETERMINATION OF I^{129}

H. T. Russell

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RECOVERY AND HALF-LIFE DETERMINATION OF I^{129}

H. T. Russell

ABSTRACT

A simple, rapid, and efficient separation method for the recovery of I^{129} activity is described. Radioactive I^{129}, a product of U^{235} fission through the Sb^{129}-Te^{129} decay chain, was separated and purified for the radioisotope distribution program at Oak Ridge National Laboratory. It was found by mass spectrographic analysis that 51.28 ± 0.05% of the iodine in the product solution is present as I^{129} and that the remaining 48.72 ± 0.05% is stable I^{127}. The I^{129} activity was determined by beta–gamma coincidence counting to be 0.261 μc/ml. The total iodine concentration was ascertained by potentiometric analyses as 2.866 mg/ml. The half life, which is calculated from the above data, is 1.56×10^7 years.

INTRODUCTION

Radioiodine-129 is a product of U^{235} fission through the isobaric decay chain[1] as shown in Fig. 1. The iodine thus formed is trapped within the uranium metal. The iodine is released during metal dissolving and is distilled as elemental iodine from the nitric acid solution. The dissolver off-gases which carry the iodine activity are passed through a scrubber tower containing Berl saddles coated with silver nitrate, which traps the iodine as silver iodide. All short-lived iodine activity is allowed to decay before the long-lived I^{129} is recovered.

The first isolation of I^{129} and the characterization of its nuclear properties were done by Parker.[2] He estimated the energy of the beta particles to be 120 kev and that of the photons to be 30 kev. Later Borkowski[3] found a 39-kev gamma ray, in addition to the 30-kev x ray reported by Parker.

[1] W. H Sullivan, *Trilinear Chart of Nuclides*, GPO, Washington, January 1957.

[2] G. W. Parker *et al.*, ORNL-286, p 53 (Classified).

[3] C. J. Borkowski and A. R. Brosi, ORNL-607, p 52 (Classified).

RECOVERY OF IODINE ACTIVITY FROM BERL SADDLES

Iodine activity may be recovered by direct reduction of the silver to the metal on the Berl saddles. Berl saddles were submerged in a distillation flask in 1% sulfuric acid. The solution was brought to boiling, and powdered zinc was added to reduce the silver to the metal according to the following reactions:

$$2AgI + Zn \xrightarrow{H_2SO_4} 2H + ZnSO_4$$

$$2AgI + 2H \longrightarrow 2Ag + 2HI$$

The silver iodide may be removed from the Berl saddles before processing if desired. Berl saddles were covered with water and subjected to ultrasonic vibrations for approximately 5 sec, which quantitatively removed the silver iodide. One gram of silver iodide was obtained from 3 liters of Berl saddles.

RECOVERY OF IODINE FROM SILVER IODIDE

Iodine was recovered from silver iodide by reducing the silver to the metal with zinc in 1% sulfuric acid and freeing the iodine as HI in the

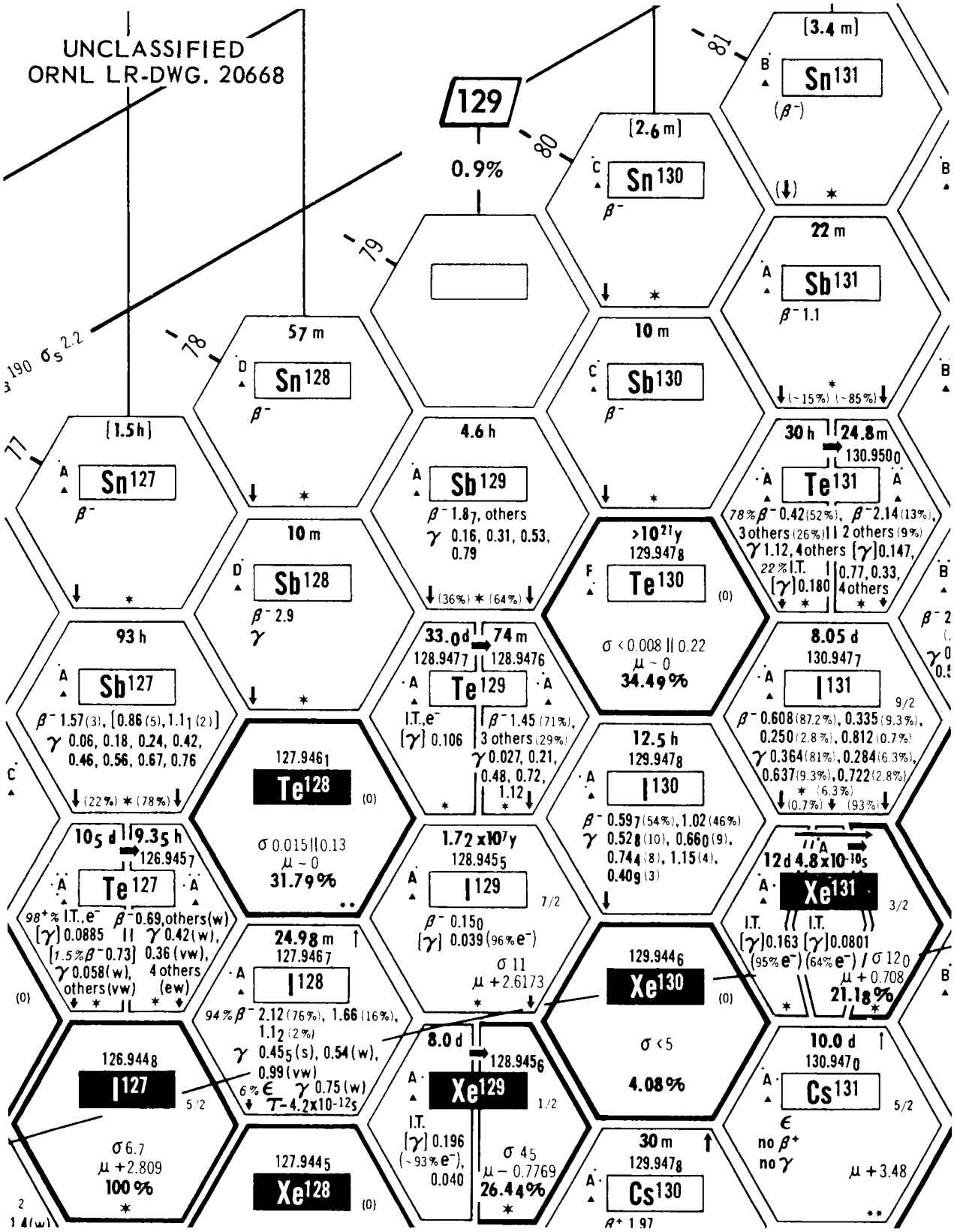

Fig. 1. Isobaric Decay Chain of I^{129}.

boiling solution. Metallic silver settles to the bottom along with a small excess of powdered zinc. Excess zinc metal is desirable to prevent dissolution of the silver in the subsequent distillation procedure. The apparatus used for the recovery and distillation of the iodine is shown in Fig. 2.

DISTILLATION OF IODINE

Following the reduction of silver, 16 *N* nitric acid is added dropwise to the surface of the boiling solution in the flask. Iodine evolves immediately and is trapped in weak sodium hydroxide solution along with a small amount of nitrogen oxide. The addition of excess nitric acid to the distillation flask should be avoided to prevent dissolution of the silver metal and reprecipitation of silver iodide.

Distillation is discontinued when iodine vapors are no longer visible in the distillation flask.

FINAL PURIFICATION

Further purification and concentration of the iodine activity were accomplished by a method developed by Rupp, Beauchamp, and Farmakes.[4] The preliminary separation method, described in the previous section, placed the iodine in a sodium hydroxide solution which contained traces of nitrate and nitrite ions. These impurities were removed by oxidizing the iodine to the nonvolatile iodate and distilling the nitric acid. The iodate was then reduced to iodine, which was distilled into sulfurous acid. The chemical steps involved are described below.

Oxidation of Iodine to Iodate

A 1 *N* potassium permanganate solution was added to the sodium hydroxide solution to oxidize iodine to the iodate. Then 18 *N* sulfuric acid was added to neutralize the caustic and to raise the sulfuric acid concentration in the solution to 6 *N*. When the purple color faded during the acidification, 1 *N* potassium permanganate solution was added to restore the permanganate color.

[4] A. F. Rupp, E. E. Beauchamp, and J. R. Farmakes, *Production of Fission Product Iodine 131*, ORNL-1047 (Dec. 18, 1951).

Distillation of Nitric Acid

Ten per cent of the solution was distilled and discarded to remove traces of nitric acid from the nonvolatile iodate.

Reduction and Distillation of Iodine

The sulfuric acid cancentration in the distillation flask was readjusted to 6 *N* by the addition of water.

Saturated phosphorous acid solution (H_3PO_3) was added dropwise to the boiling solution to reduce the excess potassium permanganate and to reduce the iodate to iodine. The purple color of the permanganate ion disappeared when the reduction was complete. Elemental iodine was then distilled and was trapped in a solution of weak sulfurous acid. The rate of distillation was accelerated by the addition of a few drops of 30% H_2O_2 to the distillation flask.

Sodium bicarbonate was added to neutralize the sulfurous acid and make the product solution slightly basic (pH $\sim$8).

DISCUSSION

No radiocontaminants were detected in a product solution containing 179 mg of I^{129}. The gamma-ray spectrum obtained by use of a single-channel, sodium iodide crystal gamma-scintillation spectrometer is shown in Fig. 3.

Mass spectrographic analysis[5] showed that 51.28 ± 0.05% of the iodine in the product solution was present as I^{129} and that 48.72 ± 0.05% was present as I^{127}. The total iodine concentration was determined, by potentiometric analysis,[6] to be 2.866 mg/ml.

The I^{129} activity was determined as 0.261 μc/ml by comparison with an I^{129} sample which had been standardized by beta-gamma coincidence counting.[7]

The half life can be calculated from the above data by the expression

$$T_{1/2} = \frac{N \ln 2}{|dn/dt|},$$

[5] W. D. Harman, Assay Laboratory Department, Y-12.

[6] J. H. Edgerton, Analytical Chemistry Division.

[7] W. S. Lyon, Analytical Chemistry Division.

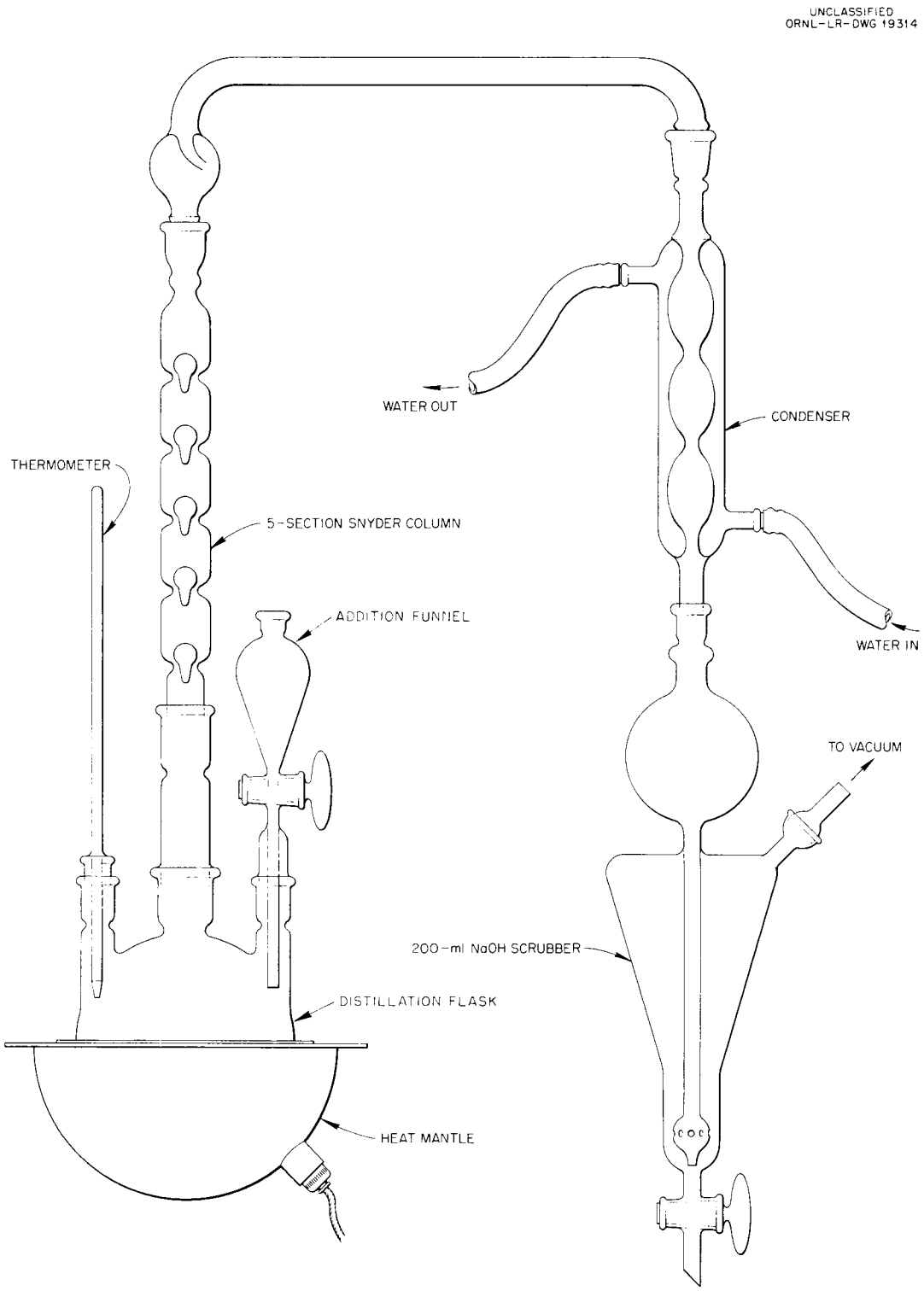

Fig. 2. Apparatus for Recovery and Distillation of Iodine.

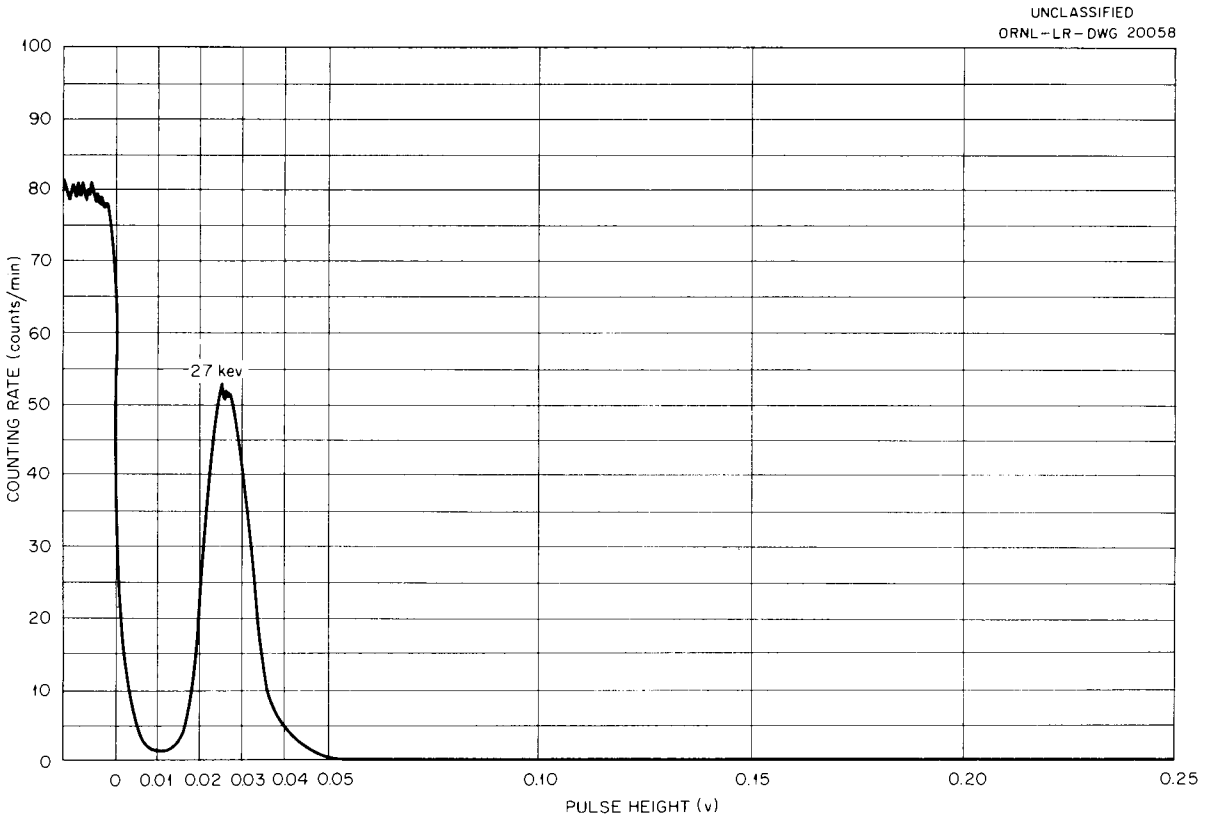

Fig. 3. Gamma-Ray Spectrum of I^{129} on NaI Spectrometer.

where

$T_{1/2}$ = the half life, in seconds,

N = the number of I^{129} atoms per milliliter,

$$= \frac{6.025 \times 10^{23}}{129} \times 1.470 \times 10^{-3} \text{ g of } I^{129}$$

per milliliter,

$|dn/dt|$ = the rate of decay, in atoms per second per milliliter, at time t.

The confidence limits were calculated[8] from the values listed below:

Amount of Activity (μc/ml)
0.2670
0.2617
0.2536
0.2500
0.2800
0.2536

Amount of I^{129} (%)
51.330
51.394
51.404
51.386
51.218
51.166
51.199
51.152

Total Iodine (mg/ml)
2.89
2.89
2.86
2.85
2.84

The value obtained for the half life by means of the expression $T_{1/2} = \frac{N \ln 2}{|dn/dt|}$, with the limit of error at the 95% confidence level, is $(1.56 \pm 0.06) \times 10^7$ years.

[8] These limits are approximations which were calculated by M. Kastenbaum, Mathematics Panel.