Preparation of Carrier-Free Yttrium-90 by Paper Chromatography

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Although carrier-free yttrium-90 has been separated from strontium-90 by various methods[1], the authors have found that carrier-free yttrium-90 can be separated more simply and rapidly by paper chromatography than by other methods and the preliminary report of this investigation has been already published in this Bulletin[1]. The separation of yttrium-90 and strontium-90 by paper chromatography was also studied by H. Kamada and et al.[2], but they did not aim at the preparation of carrier-free yttrium-90. In this paper the detailed results of the study on the preparation of carrier-free yttrium-90 by this method and some types of behavior of yttrium on the paper are described.

Experimental

A mixture of strontium-90 and yttrium-90 was employed as a form of carrier-free nitrate in 0.3 N nitric acid. Yttrium oxide prepared by F. H. Spedding was converted into nitrate and was used as a carrier. The other reagents were all of extra pure grade.

A sample solution was placed at a point 3 cm. from one edge of the strip of the filter paper (Toyo No. 3, 2×40 cm.), allowed to dry in the air and then developed chromatographically by the ascending method. After the development, the paper strip was dried and cut off at an interval of 1 cm. Each piece of the paper strip was placed at the center of a sample pan and its radioactivity was measured with an end-window counter (Kobe Kogyo Corp.). All of the original positions of samples were in the middle of the pieces No. 0 and No. 1 of the histograms shown in the figures of the present paper.

Solvent and Developing Time

In the early work of this investigation a mixture of ethyl alcohol, butyl alcohol, and 10% aqueous solution of ammonium thiocyanate was used as a solvent, but subsequently it has been disclosed that butyl alcohol is not a substantial agent for the present purpose. Therefore a mixture of ethyl alcohol and 10% solution of ammonium thiocyanate was mainly employed as a solvent.

The dependence of the distribution of yttrium-90 and strontium-90 upon the ratio of ethyl alcohol and 10% solution of ammonium thiocyanate is illustrated in Fig. 1. A larger amount of thiocyanate solution gives a higher Rf value of strontium-90, while it also causes the upward diffusion of yttrium-90 and, on the other hand, a smaller amount of thiocyanate solution can hold yttrium-90 near the original position, but the Rf value of strontium-90 is lower. Therefore the solvent of the optimum composition is a mixture of 5 parts of ethyl alcohol and 2 or 3 parts of 10% solution of ammonium thiocyanate. When the ratio of ethyl alcohol and aqueous solution is fixed at 5 to 3, the concentration variation of thiocyanate in aqueous solution from 6 to 18% does not exert any remarkable influences on the distribution of yttrium-90 and strontium-90.

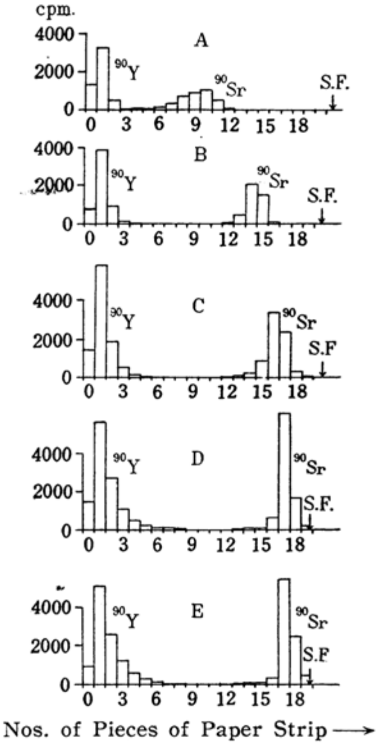

Fig. 1. Chromatograms of ^{90}Y and ^{90}Sr.
A. EtOH−10% NH_4SCN aq. (5:1).
B. 〃 〃 (5:2).
C. 〃 〃 (5:3).
D. 〃 〃 (5:4).
E. 〃 〃 (5:5).

Ammonium thiocyanate can be replaced by potassium thiocyanate. When the ammonium salt of chloride or nitrate is used instead of thiocyanate, yttrium-90 can be similarly separated from

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[1] cf. T. Kiba, S. Ohashi and T. Minabe, This Bulletin, **28**, 444 (1955).

[2] H. Kamada, et al., presented at the Meeting of The Japan Society for Analytical Chemistry in Tokyo, May 9, 1953.

strontium-90, but the Rf values of strontium-90 are relatively low. Ammonium sulfate does not provide a sharp separation of these nuclides. These results are shown in Fig. 2.

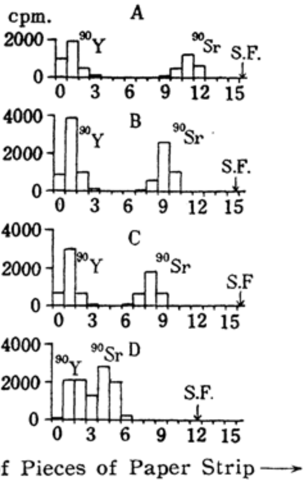

Fig. 2. Chromatograms of ^{90}Y and ^{90}Sr.
A. EtOH $-$ 2 N (15%) NH_4SCNaq. (2 : 1).
B. 〃 $-$ 2 N NH_4NO_3aq. (2 : 1).
C. 〃 $-$ 2 N NH_4Cl aq. (2 : 1).
D. 〃 $-$ 1 N $(NH_4)_2SO_4$ aq. (1 : 1).

As it was supposed that strontium may have an affinity to combine with thiocyanate, some examinations about it were carried out by spectrophotometric and polarographic methods. However it was found at least that strontium does not form a stable complex with thiocyanate.

The minimum developing time required to separate completely yttrium-90 from strontium-90 is thirty minutes as shown in Fig. 3.

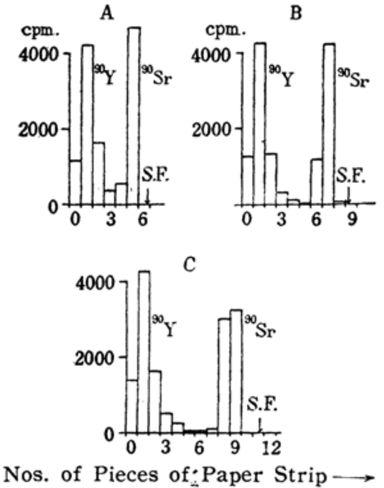

Fig. 3. Chromatograms of ^{90}Y and ^{90}Sr.
Solvent: EtOH $-$ 10% NH_4SCN aq. (5 : 3).
A. Developing time 15 min.
B. 〃 30 min.
C. 〃 45 min.

Leaching of the Separated Yttrium-90

Although several methods were tried to leach the separated yttrium-90 from the paper strip, the following method was the best.

As shown in Fig. 4 the paper strip holding the separated yttrium-90 was placed in a small glass tube with a flat bottom and 2 ml. of 3 N hydrochloric acid was added in it. After digesting the contents in the tube for a few minutes the solution was sucked up by means of a small filter stick. Repeating this procedure the results as shown in Table I were obtained.

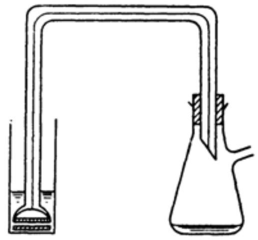

Fig. 4. Apparatus for leaching of ^{90}Y.

TABLE I

LEACHING OF THE SEPARATED YTTRIUM-90

3 N HCl, ml.	Yield, %
2	96.3
4	98.8
6	98.4
8	99.2
10	99.7

By the use of 10 ml. of 3 N hydrochloric acid the separated yttrium-90 can be almost completely released from the paper.

Purity of the Separated Yttrium-90

The half-life of the separated yttrium-90 was estimated as sixty-two to sixty-four hours as fairly coinciding with the value of sixty-four hours in the recent literature[3].

However it was found that the separated yttrium-90 had a very small amount of radioactive impurity, and by the redevelopment of the separated yttrium-90 it was also confirmed that the impurity was due to strontium-90. The amount of strontium-90 as the impurity was estimated by measuring the remaining radioactivity after all of the initially existing yttrium-90 had practically disappeared. As shown in Table II the impurity, strontium-90, in the separated yttrium-90 was less than 0.1% at the moment of the separation. Even in the case of the development of thirty minutes there was only 0.024% of strontium-90.

Relation between the Distribution of Yttrium on the Paper and the Amount of Yttrium-Carrier

As described above, the carrier-free yttrium-90 is located near the original position on the paper,

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[3] M. L. Salutsky and H. W. Kirby, *Anal. Chem.*, **27**, 567 (1955).

TABLE II

THE IMPURITY OF THE SEPARATED YTTRIUM-90

Solvent Ratio	EtOH	5	—	5	5	5	5
	BuOH	—	5	—	—	—	—
	10% NH_4CNS aq.	2	2	3	4	3	3
Developing Time, hr.		3	3	3	3	1	0.5
Solvent Front, cm.		19.7	17.8	19.8	18.7	11.2	8.3
Initial Activity, cpm.		3922	5211	5872	5599	4405	4582
Activity after t Days, cpm.		4.4	6.7	5.1	0.3	3.2	2.2
t Days		62	62	104	104	103	103
^{90}Sr in Initial Activity, %		0.056	0.064	0.044	0.003	0.036	0.024

when it is developed with a solvent composed of 5 parts of ethyl alcohol and 2 or 3 parts of 10% solution of ammonium thiocyanate. However, if yttrium-carrier is added to carrier-free yttrium-90, its distribution on the paper is varied as indicated in Fig. 5. Namely, when the amount of yttrium-carrier exceeds about 6×10^{-4} μg., yttrium goes gradually upward from the original position as the amount of its carrier increases. The similar behavior of lanthanum was observed by Y. Kitano[4]. On the other hand the Rf value of strontium-90 obtained by the development with the above mentioned solvent is kept constant, regardless of the presence of strontium-carrier.

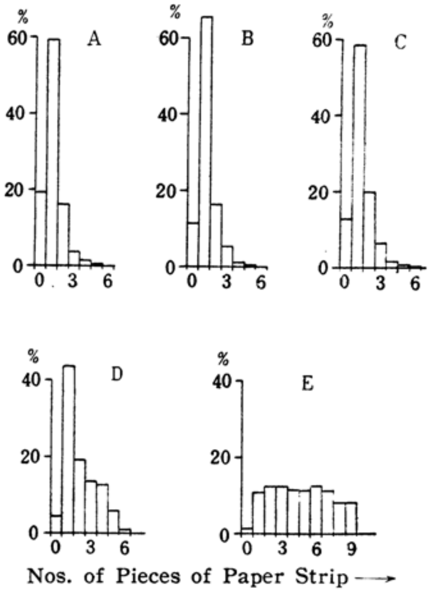

Fig. 5. Chromatograms of Y-carrier.
Solvent: EtOH−10% NH_4SCN aq (5:2).
Solvent front: 17.5−18.5 cm.
A. Y ca. 5×10^{-5} μg.
B. Y ca. 6×10^{-4} μg.
C. Y ca. 7×10^{-3} μg.
D. Y ca. 7×10^{-2} μg.
E. Y ca. 8×10^{-1} μg.

If the amount of yttrium-carrier is fixed on 0.8 μg., its distribution on the paper is varied according to the developing time as illustrated in (A) to (E) of Fig. 6. In the case of 2.5 μg. of yttrium-carrier its distribution at ninety minutes is given in (F) of Fig. 6.

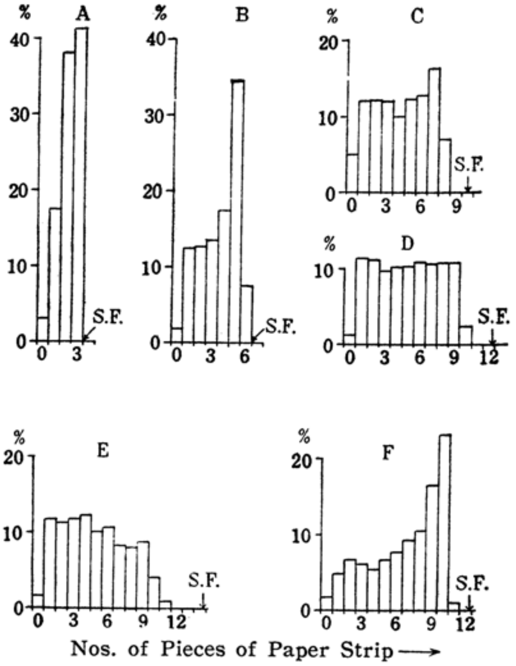

Fig. 6. Chromatograms of Y-carrier.
Solvent: EtOH−10% NH_4SCN aq. (5:2).
A. Y ca. 0.8 μg, developing time 15 min.
B. 〃 〃 30 min.
C. 〃 〃 60 min.
D. 〃 〃 90 min.
E. 〃 〃 120 min.
F. Y. ca. 2.5 μg. 〃 90 min.

These results may be explained by the assumption that yttrium is adsorbed on the paper and the adsorbable amount of the yttrium has a definite limit. The fact that the carrier-free yttrium-90 can be almost completely held near the original position may be due to its extremely small amount. If an amount of yttrium-carrier on a definite area of the paper exceeds the adsorbable amount, its excess part is accumulated at the top of its distribution pattern as shown in (B) or (F) of Fig. 6.

If a mixture of 5 parts of ethyl alcohol, 3 parts of 10% solution of ammonium thiocyanate, and

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[4] Y. Kitano, presented at the Symposium of The Japan Society for Analytical Chemistry in Osaka, May 3, 1955.

2 parts of 1 N hydrochloric acid is used as a solvent, yttrium gives a normal chromatographic spot with the Rf value of 0.79 regardless of the amount of yttrium-carrier, because the adsorption of yttrium on the paper is forbidden by the presence of hydrochloric acid.

Summary

(1) Carrier-free yttrium-90 can be separated rapidly and simply from strontium-90 by paper chromatography. The most suitable solvent is a mixture of ethyl alcohol and 10% solution of ammonium thiocyanate (5:3 or 5:2). The Rf value of carrier-free yttrium-90 is 0 and that of strontium is 0.7 to 0.8.

(2) Carrier-free yttrium-90 having a radiochemical purity more than 99.9% can be prepared within forty minutes.

(3) If yttrium-carrier is present, when it is developed with the above mentioned solvent, it is distributed upward from the original position on the paper. Its distribution pattern depends upon the amount of yttrium-carrier and the developing time and it may be caused by the adsorption of yttrium on the paper.

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