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RADIOCHEMICAL DETERMINATION OF SODIUM-24 AND SULFUR-35 IN SEAWATER

by D. Love D. Sam

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ADMINISTRATIVE INFORMATION

The work reported is part of a project sponsored by Bureau of Ships, under Project Index No. S-F011 05 11. The project is described in this Laboratory's USNRDL Technical Program for Fiscal Years 1960 and 1961, Revision #1, 1 July 1959, where it is designated as Program A-1, Problem 9.

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ABSTRACT

Rapid radiochemical procedures were developed for determining Na²⁴ and S35 in seawater containing fission product radionuclides. Na²⁴ is separated from other radionuclides by scavenging with lanthanum hydroxide; two sodium chloride precipitations with hydrogen chloride gas follow. The disintegration rate is determined by measuring the area of the 1.368 Mev gamma photopeak. The working time required for a single analysis is 1/2 hr; the precision, ± 1 %; the chemical yield, about 70 %; and the total effective decontamination factor from fission products, $\gg 10^{5}$.

 s^{35} in seawater is separated from other radionuclides through precipitating barium sulfate. This is followed by reducing the sulfate to hydrogen sulfide with hydrogen iodide and by subsequently oxidizing sulfide to sulfate in an alkaline peroxide solution. The resulting sulfate ion is precipitated as barium sulfate for chemical yield determination and counting. The working time required for a single analysis is less than 1 hr; the precision, ± 5 %; the chemical yield, 70-80 %; and the decontamination factor from fission products, > 10⁷.

Although the samples tested are salt solutions contaminated with fission products, as in the case of nuclear explosions in the ocean end in salt domes, these methods are applicable to many other types of camples containing fission products and induced activities.

SUI MARY

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The reliclogical composition of seawater and fallout originating from underwater nuclear devices is mostly a mixture of fission product radionuclides and of radionuclides induced in seawater and weapon debris. Of the radionuclides induced in seawater, Na²⁴ is the most abundant from 1 hour to 1 week after detonation and S35 is the most abundant for times greater than 1 week. The radiochemical procedures for analyzing contaminated seawater samples for Na²⁴ and S35 were time-consuming and difficult to perform. The problem was to improve these radiochemical procedures.

Findings

Rapid new radiochemical procedures for Na²⁴ and S³⁵ in fission product mixtures and in a seawater medium were developed. The working time required for a single analysis is greatly reduced; about 1/2 hour for Na²⁴ and 1 hour for S³⁵. The procedures are applicable to saltcontaining samples such as would result from nuclear detonations in the ocean and in salt domes, and to other types of contaminated materials.

INTRODUCTION

Nuclear detonation in seawater produces a mixture of radionuclides some of which are formed through neutron capture by those elements found in the water. The composition of the mixture of these induced radionuclides depends upon the time elapsed after the event.

Dyrssen and Nyman¹ calculated the relative activities of the slowneutron-induced radioelements in seawater. They showed that after the first hour, Na²⁴ constitutes nearly all the induced activity throughout the first week, after which almost all the activity is due to S³⁵. Sodium-24 is formed from a (n,r) reaction on stable Na²³, and S³⁵ is produced by the (n,p) reaction on stable Cl³⁵ and the (n,γ) reaction on stable S³⁴. Approximately 200 times more S³⁵ is formed by the (n,p)reaction on S³⁴.

Since Na²⁴ and S³⁵ are the two most abundant radionuclides induced in scawater following a nuclear detonation, it was desirable to develop simpler and more rapid radiochemical procedures for their analyses than those existing.

SODIUM-24

The Na²⁴ decay scheme consists essentially of a beta particle of maximum energy of 1.39 Mev, followed by two gammas in cascade at 1.368 and 2.750 Mev.² The half-period of Na²⁴ is 15.06 hr.³

R. J. Prestwood⁴ reported a radiochemical separation procedure of Na²⁴ from a mixed fission product solution. After an initial iron scavenge on the sample, sodium is precipitated as the sodium magnesium uranyl acetate. The sodium is finally converted to the chloride for counting. The chemical yield is about 50 to 60 %. While excellent decontantination is obtained, this procedure is tedious and times consuming.

Since Na²⁴ has a relatively short half-period, a more rapid method • of separation, adaptable to the analysis of numerous samples, is necessary. A procedure employing gamma-ray spectrometry in conjunction with • limited chemical separations was devised which provides a considerable saving in time and effort.

SULFUR-35

Sulfur-35 is a pure beta-emitter having a half-period of 87.16 days5 and a maximum beta energy of 0.1674 Mev.6

All of the sulfur in seawater is assumed to be present as the sulfate ion and is usually determined by precipitation as barium sulfate. The procedure and errors involved are adequately discussed in standard works.7

Two procedures are known for radiochemically separating S³⁵ from mixed fission products. Metcalf³ described a procedure in which bromine is used to convert sulfide to sulfate, which is then precipitated as benzidine sulfate. Decontamination is effected by repeated extractions of the benzidine sulfate in basic solution with isopropyl ether. This is followed by reprecipitation of benzidine sulfate with benzidine hydrochloride in acid solution. A ferric hydroxide scavenge is made and a final benzidine sulfate precipitation performed for chemical yield determination and counting.

Handley and Reynolds⁹ improved Metcalf's procedure by performing three ferric hydroxide scavenges, four benzidine sulfate precipitations, and three isopropyl ether extractions. A decontamination of greater than 107 and a chemical yield of 60 % were obtained from a mixed fission product sample. Approximately 3 hr are required to perform 4 determinations.

These procedures are rather slow and unsatisfactory for analyzing numerous 335 samples. A more rapid radiochomical procedure was therefore developed.

SEAWATER SAMPLES

In the development of the two radioshemical procedures, seawater was used which was obtained 40 miles off the coast of California. Since seawater contains large concentrations of goldup ion and sulfate ion. relatively constant at 8.650 and 2.372 g/kg of seawater, these carriers were not added.

No suspended matter (inorganic nor organic particles) was visible in the seawater. The concentration of particulate matter¹⁰ in the coastal waters off Massachusetts (depth 70 meters) is 0.4 to 2 mg/1, while that in the ocean water 100 miles beyond the continental shelf is 0.2 mg/1. Also, dissolved organic substances in seawater from the North Atlantic have been estimated to be about 5 to 6 mg/1. Na²⁴ and S35 are not concentrated by marine organisms.¹¹ Thus the presence of sodium and sulfur agglomerated in naturally occurring particulate or organic matter was considered negligible. Particulate matter formed from nuclear weapon debris also would contain negligible emounts of Na²⁴ and S35.

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AGALYSIS OF Ma²⁴

The radiochemical procedure developed is summarized as follows: Na24 is experted from other radionuclides by scavenging with lanthanum hydroxide; two sodium chloride precipitations with hydrogen chloride gas follow. The disintegration rate of Na24 is determined by measuring the area constituting the 1.368-May gamma photopeak, which is obtained by scintillation spectrometry with a NaI(T1) detector and a 256-channel pulse-height analyzer. The gamma-ray spectrometer is calibrated with Na24 samples of known specific activity.

PROCEDURE

Analysis

In detail the procedure is as follows: To a hot solution of 25 m of seawater and 1 ml of lanthanum carrier (10 mg La/ml) in a 50-ml contrifuge tube, add armonium hydroxide until lanthanum hydroxide precipitates. Centrifuge the colution and filter it through a No. 42 Whatman filter paper into a clean centrifuge tube. Cool the solution in an ice bath. Pass anhydrous hydrogen chloride into the solution by means of the anhydrous hydrogen chloride saturation apparatus (description below). Saturate the solution with the gas under pressure (5 psig) for 3 min after sodium chloride precipitates out. Centrifuge the solution for 30 sec and discard the supernatant solution. Wash the sodium chloride precipitate with 10 ml of hydrochloric acid wash solution (concentrated hydrochloric acid saturated with hydrogen chloride gas) and discard the wash. Dissolve the sodium chloride in 10 ml of distilled water. Again precipitate sodium chloride with anhydrous hydrogen chloride. Wash the precipitate once with 5 ml of the hydrochioric acid wash solution, then filter through a medium-pore sintered glass filter. Wash it twice with 5-ml aliquots of ethyl alcohol caturated with hydrogen chloride gas, and twice with 5-ml aliguots of diethyl ether saturated with hydrogen chioride gas. Spread out the precipitate at the bottom of a preve igned .

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weighing bottle (standard taper joint No. 29/12), for counting in a previously calibrated gamma-ray spectrometer. After the counting, dry the sodium chloride at 105° C in an oven, and cool in a desiccator. Weigh to determine its chemical yield. If necessary determine the initial amount of sodium in each sample by flame photometry. However, in almost all cases, the amount of sodium in the seawater is a known constant.

Spectrometer Calibration

Place a known aliquot of a Na²⁴ solution on a gold-plated VXNS film used for 4-pi beta counting. Dry it under an infra-red lamp and count in a 4-pi gas-flow beta proportional counter to determine the Na²⁴ absolute disintegration rate. Fate and Jaffel² give a detailed description of the 4-pi beta counting method. Then calibrate the gammaray spectrometer with this Na²⁴ solution. To obtain a precise measurement of Na²⁴ activity, evaluate the data obtained from the 1.368-Mev total absorption peak by the method of Covell.¹³ In this method a given fraction of the area of the total absorption peak is determined by analyzing the digital data defining that area of the peak.

256-Channel Gamma Anelyzer

The gamma-ray spectrometer used is a USNRDL modified version of the RCL 256-channel pulse-height analyzer, Mark 20, Model 2603 (Manufactured by Radiation Counter Laboratory Inc., Nucleonic Park, Skokie, Illinois). The detector is a 3 in. x 3 in. NaI(T1) crystal coupled to a 3 in. Dumont, Model 6363 photomultiplier tube, output from which is coupled to a linear amplifier through a cathode-follower preamplifier. The data print out in digital form.

Saturation Apparatus

A 50 ml-centrifuge tube, having a flared top, is fitted with a twohole rubber stopper. A clamp holds the rubber stopper tightly in the centrifuge tube. One hole holds a gas dispersion tube which is connected to a tank of anhydrous hydrogen chloride. This tube dips below the surface of the solution to be saturated. The other hole holds a glass tube fitted with a glass stopcock. This is connected to a funnel with Tygon tubing. The funnel is inverted and dips 1 mm into water to trap escaping hydrogen chloride gas.

RESULTS AND DISCUSSION

The present procedure for Na^{24} is based on a limited radiochemical separation followed by an assay of a high energy Na^{24} gamma-ray photopeak. This technique gives a very large effective decontamination from other radionuclides. The precision and accuracy of the procedure are determined mainly by the assay of the Na^{24} photopeak. The stability of the gamma-ray spectrumeter, the variation of photopeak counts of Na^{24} with specific activity, and the effect of other gamma-emitting radio-nuclides were determined and are described below.

The precision with which Na²⁴ is measured depends much on the stability of the gamma-ray spectrometer. The spectrometer is regularly subjected to quality control procedures in order to maintain a high degree of stability.¹³ Experience with the 256-channel analyzer has shown no measurable change in the photopeak counting efficiencies of Co60 for over a year. Also, repeated measurements with the same sample have shown that the variation in counting rate is that predicted by counting statistics.

The absolute disintegration rate of Na²⁴ is known through calibration of the spectrometer. The efficiency factor, N'/dpm, vs. the weight of sodium chloride is presented in Fig. 1. N' is the amount of activity from a Na²⁴ source counted for 100 seconds which is part of the arbitrarily chosen portion of the total absorption peak extending 10 channels on either side of the maximum activity channel.

Several radionuclides having garma energies above 1 Mev could interfere with the measurement of the 1.363-Mev gamma ray of Na24. In order to establish these interferences, an estimate is made of the relative activities of the pertinent fission products and induced radionuclides occurring in seawater at 15 hr after irradiation. Table 1 lists the relative activities of the possible interfering induced activities. Column 3 of this table was taken from an article by Senftle and Champion.14 Here As, is defined as the specific initial activity in 1 g of the target material for 1 sec of irradiation with a flux of 1012 neutrons/cm² sec. Column 4 takes into consideration the relative concentration of the target material in seawater, the relative abundance of the gamma rays having energies above 1 Nev, the decay of each nuclide for a period of 15 hours after fission, and the neutron cross-sections for competing reactions. The number of neutrons emitted per fission from y235 undergoing neutron fission is approximately 2.5.15 One neutron is required for the propagation of one fiscion reaction. Of the 1.5 neutrons left, two nuclides (H4 and C132)1 absorb approximately 99 2. Thus only a fraction, approximately 6 x 10-3, of all the neutrons



TABLE 1

Induced Radionuclide	^T 1/2	As ^a As	Gamma Emission Rate at 15 hr After Irradiation ^b (photons/sec)
Na24	15.06 h	1.8 x 10 ⁵	5.4 x 10 ²
C138	37.29 m	7 x 105	3 x 10-4
K42	12.44 h	1.5×10^4	0.8
Sc46	85 d	1.5×10^4	2 x 10-7
Mn56	2.576 h	1.0 x 107	5 x 10 ⁻⁴
Fe59	45.1 d	4	2 x 10-3
C060	5.27 x	8.5×10^2	1 x 10-9
Cu64	12.8 h	4 x 105	5×10^{-4}
N165	2.564 h	2.1×10^4	9 x 10-10
2n65	250 d	53	5 x 10-2
As75	26.1 h	2.4 x 105	6 x 10-4
Br.82	35.87 h	6.9 x 104	0.6
Agllom	270 d	2×10^2	8 x 10-9

Relative Activity of Most Abundant Radionuclides Induced by Thermal Neutron Irradiation of Seawater

a. From Reference 14.

b. Normalized to the activity induced by neutrons from 1.32 x 1012 fission events occurring in seawater.

released per fission is available for capture by the other components in seawater. Of this, 4.4×10^{-3} is taken by sodium and 2.6×10^{-4} by sulfur. The values in Column 4 of Table 1 were normalized to 1.32×10^{12} fission events to allow direct comparison with calculated fission product activities, given in Table 2. Table 2 lists the fission products having gamma energies above 1 Mev and having relatively high fission yields. Column 3 shows the gamma emission rate of each fission product nuclide at 15 hr post-fission, and takes into consideration the respective mass chain yields.¹⁶ Each value corresponds to the activity from 1.32×10^{12} fission events. This number of fissions corresponds to the fission products obtained by the irradiation, for 1 sec, of 1 g of enriched U²³⁵ of 93.2 % isotopic abundance, at a thermal neutron flux of 10^{12} neutrons/cm² sec. Column 3 of Table 2 is directly comparable to Column 4 of Table 1.

Fission Product Nuclide	T _{1/2} (hr)	Gamma Emission Rate at 15 hr After Irradiation (photons/sec)
Sr91 Y92 Agl12 Tel31m I132 I133 I135 La140 La141	9.7 3.6 3.2 30 2.4 22.4 6.7 40 3.7	2×10^{4} 4.9×10^{4} 1.1×10^{2} 1.6×10^{3} 7.3×10^{2} 9.2×10^{3} 2×10^{5} 3.7×10^{3} 1.2×10^{4}

Relative Activity of Most Abundant High Gamma Energy Fission Products in a Solution Containing 1.32 x 1012 Fissions

TABLE 2

Decontamination tests were made with various radionuclides in the absence of Na²⁴. The decontamination factors of the more abundant radionuclides, given in Table 3, are sufficient for satisfactory removal of the interfering radionuclides. Decontaminations of Y^{92} and Ag^{112} were not tested, because it has been observed that yttrium radio-isotopes are effectively removed by a lanthanum hydroxide scavenge, and that silver radioisotopes are separated by formation of the silver chloride complex during the precipitation of sodium chloride in the presence of a high hydrochloric acid concentration.

Excellent recovery by this procedure was obtained from a solution containing 4.06 x 10^4 dpm of Na²⁴, fission products of 2.1 x 10^{10} fissions, and 25 ml of seawater. The results for four aliquots are presented in Table 4. Chemical yields of 70 % were obtained. Gamma-ray spectra of a source containing 4.06 x 10^4 dpm of Na²⁴, of fission products (35 hr old) of 2.1 x 10^{10} fissions, and of a Na²⁴ source separated from the above solution, are compared in Figs. 2 and 3. The final socium chloride precipitate, after separation of the fission products, showed no visible contamination of the Na²⁴ gamma-ray photopeak.

TABLE 3	
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Decontamination Factors of Some Interfering Isotopic Radionuclides

Radionuclides	Decontamination Factor
Sr ⁸⁵ Bal33 Lal40 Te fission-product isotopes in equilibrium with res- pective I daughters	1×10^{4} 1×10^{2} 6×10^{3} 6×10^{2}
1131 Fission Products (2 months old)	·1 x 10 ⁴ 3 x 10 ⁵

TABLE 4

Recovery of Na²⁴

Sample	Na ²⁴ Determined
No.	(dpm)
1 2 3 4	4.15 x 10^{4} 4.02 x 10^{4} 4.12 x 10^{4} 4.10 x 10^{4} Average 4.10 x $10^{4} \pm 1.2 \pm$



Fig. 2 Combined Gamma Spectrum of a Solution Containing Na²⁴ plus Fission Products (35 hr old)



Fig. 3 Gamma Spectrum of Na²⁴ separated from a Solution containing Fission Products (35 hr old)

ANALYSIS OF S35

Briefly the procedure developed for analysis of S³⁵ is as follows:

The S³⁵ is separated from seawater by precipitation of barium sulfate, which is reduced to hydrogen sulfide with hydrogen iodide. The sulfide is subsequently exidized to sulfate in an alkaline peroxide solution. The resulting sulfate ion is precipitated as barium sulfate for determination of chemical yield and for counting.

PROCEDURE

Analysis

To a 10-ml aliquot of seawater add 0.5 ml of conc. animonium hydroxide and 5 ml of hydrogen peroxide. Boil. Add distilled water to make up a total volume of 500 ml. Add slowly 1 ml of conc. HCl and 5 ml of 1 M barium chloride solution to precipitate barium sulfate. Filter precipitate through a filter tower containing a filter paper circle, and wash with hot distilled water. If it is necessary to know the amount of sulfate in the seawater sample, at this point filter the barium sulfate quantitatively, wash with water, alcohol, and acetone, dry in an oven, and weigh.

Fig. 4 shows the hydriodic acid reduction apparatus.

Put the barium sulfate precipitate, filter paper, and 50 ml of concentrated hydriodic acid into a 200 ml 2-necked flask. Reflux this solution until all the barium sulfate dissolves. During this time, apply suction through a 60 ml separatory funnel (H). The hydrogen sulfide gas is absorbed in 75 ml of 1 M sodium hydroxide in a 150 ml flask (D) after passing through a gas dispersion tube (C). After the BaSO₄ reduction is complete, apply suction through a 150 ml flask (E). Expel the H₂S gas by adding 20 ml of concentrated hydrochloric acid to the



Fig. 4 HI Reduction Apparatus

A. HI reduction flask.

D. Flask containing 1 N NaOH for absorbing H29.

E. Flask containing NH₄OH and H_2O_2 for absorbing reformed H_2S_2 .

Sodium hydroxide solution in Flask D. Collect, with suction, hydrogen sulfide gas through a gas dispersion tube (F) into Flask E which contains a solution of 25 ml of concentrated ammonium hydroxide and 60 ml of 30 % hydrogen peroxide. An ice bath, G, is necessary to cool the reaction vessel and prevent forming. Rubber stoppers and tygon tubing are used.

When the oxidation and collection of hydrogen sulfide are completed, transfer the solution from Flask E to a 1-liter flask containing approximately 200 ml of distilled water. Boil to drive off excess ammonia and hydrogen peroxide. (Care is required, as this solution boils vigorously). Add about 0.5 ml of concentrated hydrochloric acid and 5 ml of 1 M barium chloride to precipitate the barium sulfate. After digestion, filter, with the aid of a filter tower, the barium sulfate onto a preweighed No. 42 Whatman filter paper circle. Wash the precipitate twice with hot distilled water, ethyl alcohol, and acetone. Then dry in an oven at 105° C, cool in a desiccator, and weigh. Mount the precipitate on a brass planchet assembly and cover with a thin film of rubber hydrochloride. A rubber 0-ring fitted around the brass planchet keeps the precipitate in place. Count the barium sulfate source in a low background beta counter.

Calibration

To facilitate determining the absolute disintegrations per minute of S^{35} , the counter must be calibrated for geometry, self-absorption, and self-scattering. An efficiency factor, the ratio of counts per minute (cpm) to disintegrations per minute (dpm) as a function of the source weight of barium sulfate, can be constructed as follows: Put an equal aliquot of S^{35} of known disintegration rate into each of six 250-ml erlenmyer flasks. (The S35 used was calibrated at the National Bureau of Standards and was reported to have been determined with a precision of ± 3 % by gas counting as sulfur dioxide.) The flasks contain various known amounts of a standardized sulfuric acid carrier. After mixing thoroughly, precipitate barium sulfate, weigh, mount on brass planchet assemblies, and count. Plot the ratio of the net count rate to the absolute disintegration rate precipitate (Fig. 5).



Fig. 5 Efficiency Factor vs. Weight of BaSC

The purpose of the initial treatment of the sample with hydrogen peroxide is to oxidize any hydrogen sulfide to sulfate ion. This serves to ensure complete exchange between \$35 and its sulfate carrier.

Bush17 reported that with concentrated sulfuric acid and excess concentrated hydrogen iodide the reaction product is mainly bydrogen sulfide according to the equation

$$2H^+ + SO_4^- + 8HI \longrightarrow H_2S + 4I_2 + 4H_2O_2$$

However, it is possible to obtain sulfur dioxide if the HI is slightly diluted.

The results of measurements of recovery of known emounts of 335 introduced into seawater are presented in Table 5. The usual corrections were applied for chemical yield and counting efficiency corresponding to the weight of barium sulfate precipitate recovered. Each

TABLE 5

Sample No.	dym of S35 Introduced	dpm of s35 · · · Determined
1 2 3	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$	$\begin{array}{r} 8.3 \times 10^2 + 5.1 \ \% \\ 7.9 \times 10^3 + 5.4 \ \% \\ 1.47 \times 10^4 + 3.1 \ \% \end{array}$

Recovery of Sulfur-35 From Seawater

value represents the average of triplicate determinations, and the precision of such determinations was about + 5 %. Errors are due mainly to difficulties in obtaining burium sulfate precipitate mounts which were sufficiently reproducible for precise counting of the weak beta-rays of S³⁵. This problem can be partially eliminated by adopting the procedure for assaying S³⁵ through gas counting¹⁸ or liquid scintillation counting¹⁹ techniques. The procedure gives decontamination factors of 107 from mixed fission products (Table 6), themical yields of 70 to 80 %, and a precision of about ± 5 %. This procedure is also adaptable to the simultaneous determination of a large number of samples.

TABLE 6

and the second second Radionuclides Decontamination Factor Ba140-La140 >107 Cel44 Pr144 >107 Cs137 >107 131 >107 1699-Tc99m >106 Pm147 >107 Ru106_Rh106 >109 sr90_y90 Ta¹⁸² >100 >100 2r95-1695 >106 Mixed Fission Products >10? · (18 days old)

Decontamination Factors of Various Radionuclides

REFERENCES

- 1. D. Dyrssen, P.O. Nyman. Acta Radiological 43, 421-427 (1955).
- 2. J. W. Knowles. Can. J. Phys. 37, 203 (1959).
- 3. J H. Sreb. Phys. Rev. 81, 469 (1951).
- 4. R. J. Prestwood. <u>Collected Radiochemical Procedures</u>. LA-1721 2nd edition. J. Kleinberg, editor, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, Aug. 1958.
- 5. H. H. Seliger, H. B. Mann, L. M. Cavallo. J. Research Nat'l. Bur. Standards 60, 447 (1958).
- 6. R. D. Conner, I. L. Fairweather. Proc. Phys. Soc. (London) 70A, 769 (1957).
- 7. H. H. Willard, N. H. Furman. <u>Elementary Quantitative Analysis</u>. pp. 363 fr. D. Van Nostrand, New York, (1940).
- 8. R. P. Metcalf. <u>Radiochemical Studies</u>, The Fission Products, Book I. C. D. Coryell and N. Sugarman, Eds. National Nuclear Energy Series, paper #47, p. 478, McGraw-Hill Book Co., 1951.
- 9. T. H. Handley, S. A. Reynolds. Oak Ridge National Laboratory, CF-56-7-118 (July 23, 1956).
- 10. H. W. Harvey. The Chemistry and Fertility of Seawater. Cambridge University Press, p. 9, 1960.
- 11. The Effects of Atomic Radiation on Oceanography and Fisheries. Publication No. 55, National Academy of Science, National Research Council, Washington, D. C. (1957).
- 12. B. D. Pate, L. Yaffe. Can. J. Chem. 33, 610 (1955).

- 13. C. F. Covell. Anal. Chem. 31, 1705 (1959). Also, Determination of 7 Ray Abundance Directly From the Total Absorption Peak. U. S. Naval Radiological Defense Laboratory Technical Report, USNRDL-TR-288, Dec. 1958.
- 14. F. E. Senfile, W. R. Champion. "Tables for Simplifying Colculations of Activities Produced by Thermal Neutrons," Supplemento al Nuovo Cimento 12, 549-571 (1954).
- 15. R. A. Charpie, J. V. Dunworth Editors, International Series of Monographs on Muclear Energy: Physics of Muclear Fission, Pergamon Press, p. 75 (1958).
- 16. S. Katcoff. Nucleonics 18, 201 (1958).
- 17. F. Bush. J. Phys. Chem. 33, 613 (1929).
- 18. W. F. Merritt, R. C. Havitings. Anal. Chem. 32, 308 (1960).

19. H. Jeffey, F. O. Olubajo, W. B. Jewell. Anal. Chem. 32, 306 (1960).

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