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PROCEDURE FOR THE RADIOCHEMICAL ANALYSIS OF ASTATINE IN BIOLOGICAL MATERIAL

Warren M. Garrison, Jeanne D. Gile, Roy D. Maxwell

and Joseph G. Hamilton

May 1950

Crocker Laboratory

Berkeley, California

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Warren M. Garrison, Jeanne D. Gile, Roy D. Maxwell* and Joseph G. Hamilton

May 1950

Introduction

Astatine, the artificially produced radioactive halogen of atomic number 85, accumulates[1,2] in the thyroid gland in a manner similar to radio-iodine. Recent[3] observations on the radiobiological properties of 7.5-hr. At^{211} show that the alpha particles emitted in the decay of this isotope produce severe radiation damage to thyroid tissue without apparent involvement of the adjacent parathyroids, indicating that astatine may have unique therapeutic application in hyperthyroidism. With rats, a dose of 10 μc of At^{211} is sufficient to produce a marked degree of injury to the thyroid follicles. As part of a detailed study of the metabolism and radiobiology of astatine now in progress at this laboratory, analytical procedures have been developed for the determination of astatine in biological material. These methods are reported in the present paper.

Experimental

Preparation of Astatine[1,4,6]. Bombardment of bismuth with alpha particles in the energy range 20-29 Mev produces, by (α,2n) reaction, 7.5-hr. At^{211}, free from other astatine isotopes, notably the 8.3-hr At^{210}. Sixty percent of the At^{211} nuclei decays by K-electron capture, with the emission of an 80-kev X-ray to Po^{211}, an alpha emitter with a 10^{-3} sec. half-life, and 40 percent decays by alpha particle emission to form Bi^{207}. Above 30 Mev, 8.3-hr At^{210} is produced by (α,3n) reaction. This isotope decays by K-electron capture to 140-day Po^{210} and was, therefore, an

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1 Corson, D. R., MacKenzie, K. R. and Segre, E.: Phys. Rev. 57, 459 (1940)

2 Hamilton, J. G. and Soley, M. H.: Proc. Nat. Acad. Sci. 26, 483 (1940)

3 Hamilton, J. G., Asling, C. W., Garrison, W. M., Scott, K. G. and Axelrod-Heller: Proc. Soc. Exper. Biol. and Med. 73, 51 (1950)

undesirable radioactive contaminant in the astatine preparations described here.

The bismuth was supported on a water-cooled aluminum target plate 10 mils thick. A small amount of C.P. bismuth was melted on the aluminum disc forming an aluminum-bismuth alloy which did not require flux. A bismuth layer of the desired thickness was obtained by fusing the required amount of bismuth to the alloyed surface. The plate was mounted in a bell-jar type target assembly and bombarded with 30 Mev alpha particles obtained by degrading the 40 Mev beam of the 60-inch cyclotron at Crocker Laboratory. The surface of the bismuth was cooled with a stream of helium. Beam currents up to 15 ua were used without noticeable fusion of the bismuth surface. After bombardment, the bismuth section was cut from the target foil and astatine was isolated by heating the bismuth to 425° C. in a stream of nitrogen carrier gas at a pressure of 10^{-2} - 10^{-3} mm. The astatine was collected at liquid-air temperatures on a cold-finger which had been covered with a thin layer of ice. After heating the bismuth for approximately 20 minutes, a solution of astatine in a minimum volume of water was obtained simply by removing the cold finger from the vacuum line and warming to room temperature.

Analytical Procedures: Since a 10 uc dose of At^{211} produces noticeable thyroid damage in the rat[3], the metabolism in normal animals had to be studied at the 1-5 uc dose level. Therefore, to obtain distribution data of the desired accuracy, an analytical procedure of maximum sensitivity was required. Although At^{211} emits 80 kev X-rays as well as alpha-particles, the low counting efficiency of the At^{211} X-rays made it necessary[5] to use alpha-counting methods so that the required sensitivity could be

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[4] Kelley, E. L. and Segre, E.: Phys. Rev. 75, 999 (1949)

[5] The experimentally determined alpha-particle/X-ray counting ratio was found to be approximately 850.

attained. In developing procedures for the quantitative separation of At^{211} from gram amounts of tissue so that alpha particles could be counted, several co-precipitation procedures [1,6,7] were tried. The most satisfactory of these involves the wet oxidation of the biological material with perchloric acid followed by a co-precipitation of astatine on elemental tellurium carrier from a 3 normal perchloric acid solution of the oxidation products.

Determination of Astatine by Co-precipitation with Tellurium: A sample of astatine-containing tissue (less than 10 gram wet weight) is digested in a minimum volume of 9 normal perchloric acid containing 30 percent by volume of 16 normal nitric acid. After the organic material has been oxidized[8], the clear solution is evaporated to 10-15 ml of concentrated perchloric acid. The solution is cooled and diluted to 3 normal. 5 mg. of tellurium as tellurous acid are added together with 1 ml of 12 normal hydrochloric acid[9]. A stream of sulfur dioxide is passed through the solution precipitating metallic tellurium which carries astatine quantitatively. The tellurium is separated by centrifugation, washed three times with distilled water and transferred to a porcelain counting dish. After drying at 70°C, the dish is counted for alpha activity. Typical results for a series of different tissues are shown in Table I.

The principal objection to the tellurium co-precipitation method is the

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[6] Johnson, G. L., Leininger, R. F. and Segre, E.: J. Chem. Phys. 17, 1 (1949)

[7] Corson, D. R., MacKenzie, K. R., and Segre, E.: Phys. Rev. 58, 672 (1940)

[8] Astatine is not volatile from concentrated oxidizing acids. (ref. 6)

[9] Tellurous acid is only slowly reduced in cold 3 normal perchloric acid in the absence of dilute hydrochloric acid.

time required in centrifuging, washing and transferring the co-precipitant. To obtain a more rapid analytical procedure, other chemical properties of astatine were studied, particularly the deposition of astatine on metallic foils which may be measured for alpha activity directly. Although astatine is chemically similar to iodine, the lighter halogen homologue, it also shows definite metallic properties. The similarity between adjacent elements in the last rows of the periodic table is quite marked and astatine shows a close resemblance to polonium in many of its properties[1,6,7]. Since polonium may be quantitatively removed from dilute acid by deposition on silver [10], a similar study was made with astatine. It was found from these studies that astatine in biological material may be quantitatively recovered by deposition on silver foil from a 3 normal perchloric acid solution after organic material was oxidized by the perchloric-nitric acid digestion.

Determination of Astatine by Deposition on Silver Foil: A sample of astatine containing tissue is digested in a mixture of perchloric-nitric acid as described in the tellurium procedure. After digestion is complete, the solution is evaporated until the perchloric acid fumes (approx. 10 ml), diluted to 3 normal with the addition of distilled water and transferred to a 50 ml beaker. A 2 mil thick circular silver foil of a size convenient for alpha counting[11] is placed in the bottom of the beaker and the solution is stirred for 30 minutes. The foil is washed in water and acetone, dried in air and counted for alpha activity. Typical analytical results are shown in Table I and II.

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10 Erbacher and Phillip: Zeit. f. Phys. 51, 309 (1928)

11 Although the astatine deposits principally on the top surface of the silver foil, for quantitative results both sides of the foil were counted for alpha activity.

Summary: Procedures for the radiochemical analysis of At^{211} in biological material have been described. Organic substances can be destroyed by perchloric acid-nitric acid digestion without loss of astatine by volatilization. Astatine may be isolated for alpha counting from a 3 normal perchloric acid solution of the oxidation product by (1) co-precipitation with metallic tellurium or (2) deposition on silver foil. Both procedures may be used for quantitive analysis. The silver foil method, however, is simpler, requires less time and is more easily adapted to routine determinations where large numbers of tissues are to be analysed.

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TABLE I

COMPARISON OF PROCEDURES FOR THE RADIO-CHEMICAL ANALYSIS OF ASTATINE IN BIOLOGICAL MATERIALS.

	Percent of Recovery			
Tissue (10 gm wet weight)	To Method		Ag Method	
Fat	96.0	(74.5)	96.0	94.0
Bone	93.5	105.0	103.0	93.0
Skin	98.5	92.5	100.0	98.5
Muscle	94.0	108.0	99.0	93.9

TABLE II

RATE OF ASTATINE DEPOSITION ON SILVER IN 3 N PERCHLORIC ACID

Time	Percent Deposited
0 - 10 minutes	73.5
10 - 20 "	20.0
20 - 30 "	8.0
30 - 60 "	2.0
Total	103.5