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OF AMERICIUM-241 AND AMERICIUM-243

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THE SPECIFIC ACTIVITIES AND HALF LIVES
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All the values of the half life of americium-243 published heretofore have been based on determining, by means of an alpha-ray spectrograph or alpha-pulse analyzer, the relative numbers of Am^{241} and Am^{243} alpha particles originating from a sample of americium in which the mass ratio of the two isotopes is known. The determinations have yielded two nonconcordant values for the half life: 7600 years^{1,2} and 8600 years.^{2,3} The smallest estimated uncertainty in these determinations is ± 370 years, or about 5%. A determination of the half life by a specific-activity measurement should reduce the estimated error and thus lead to a more reliable value of the half life. As a result of the americium crystal structure work done in this laboratory⁴ we had available one piece of metallic americium-241 and two pieces of metallic americium-243 suitable for a specific-activity determination for each isotope.

An attempt was made to determine the specific activities of Am^{241} and Am^{243} using the compound AmF_3 . However, it was found that the extremely erratic results always gave a shorter half life. This could arise from the formation of AmOF during the drying of the tri-fluoride; indeed, subsequent work has indicated that americium tri-fluoride prepared under similar conditions to those used for the specific-activity determinations does incorporate a variable amount of water in the AmF_3 , and this could result in hydrolysis during

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drying. This fact and the difficulty of obtaining consistent impurity corrections have rendered the fluoride data so unreliable that they are not included here.

The specific activity of Am^{243} was measured by weighing and dissolving Am^{243} metal. A check on the technique was made, using Am^{241} metal. The metals were prepared by reduction of AmF_3 with barium in a tantalum crucible system heated in high vacuum.⁴ All of each sample of metal was dissolved, so that no spectrographic analysis on the actual metal used was possible. However, spectrographic analysis on metal similarly produced from the same starting materials showed 0.1% to 0.2% metallic impurities. An impurity content in the metals of 0.15% was assumed in calculating all the results. Mass-spectrographic analysis indicated $< 0.015\%$ Am^{243} in the americium-241 and 0.03% Am^{241} in the americium-243. The metals were scraped free of slag resulting from the reduction and weighed in quartz cones on a quartz fiber torsion microbalance calibrated to $\pm 0.1\%$. The weighed piece of metal was placed in a weighed 5-ml volumetric flask, dissolved by addition of acid, and diluted to a suitable volume by adding a solution of $6 \text{ M H}_2\text{SO}_4 - 2 \text{ M HNO}_3$. This solution was chosen because there was no detectable change in weight of an aliquot over the length of time necessary to weigh the aliquot. These gravimetric aliquots of the americium solutions were dried, flamed, and counted in a low-geometry counter⁵ of measured geometry known to 0.01%. In the first Am^{243} metal sample the amount of material was so small that after weighing the metal was dissolved directly on the counting plate in a drop of sulfuric acid containing about 1 λ of HCl. More than 10^6 counts were registered for each plate. An alpha-pulse analysis of each solution was made and no alpha particles other than Am^{241} alphas were found in that solution. The Am^{243} solution was found to contain 99.35% Am^{243} alpha particles, 0.50% Am^{241} alpha particles, and 0.15% Cm^{244} alpha particles.

Table I

Sample	Specific activity (alpha dis/m/ μ g)	Half life (years)
Am ²⁴¹	$(7.194 \pm 0.029) \times 10^6$	457.7 ± 1.8
Am ²⁴³ (1st sample)	4.113×10^5	7939
Am ²⁴³ (2nd sample)	4.100×10^5	7964
Am ²⁴³ average	$(4.107 \pm 0.025) \times 10^5$	7951 ± 48

The specific-activity determinations are summarized in Table I. This value of 457.7 years for the half life for Am²⁴¹ is to be compared with the value of 458.1 ± 0.5 years obtained by Hall and Markin⁶ from extremely careful determinations of the specific activity of Am₂(SO₄)₃ and AmCl₃. It should also be noted that the agreement between the metal and salt values indicates that no residue resulted from the dissolution of americium metal, unlike the situation with thorium, uranium, or plutonium. This is in accord with the lanthanum like properties of americium metal.⁴ The uncertainty in the half life of Am²⁴¹ is estimated to be 0.4% (balance calibration 0.1%, impurity content 0.15%, counting 0.1%, aliquoting 0.07%), yielding a half life of 457.7 ± 1.8 years; the uncertainty in the half life of Am²⁴³ is estimated to be 0.6% (balance calibration 0.1%, impurity content 0.15%, counting 0.1%, aliquoting 0.17%, pulse analysis 0.1%), yielding a half life of 7951 ± 48 years.

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REFERENCES

1. J. P. Butler, M. Lounsbury, and J. S. Merritt, *Can. J. Phys.* 35, 147 (1951).
2. John P. Hummell, University of California Radiation Laboratory Report, UCRL-3456, (1956). The value of Diamond et al.³ is also recalculated here using a revised value for the Am^{241} half life.
3. H. Diamond, P. R. Fields, J. Mech, M. G. Inghram, and D. C. Hess, *Phys. Rev.* 92, 1490 (1953).
4. P. Graf, B. B. Cunningham, C. Dauben, J. C. Wallmann, D. H. Templeton, and H. Ruben, *J. Am. Chem. Soc.* 78, 2340 (1956).
5. H. P. Robinson states that a description of this counter is to be published by the National Research Council, Subcommittee for Standards and Measurement of Radioactivity, in the proceedings of a conference held October 9 - 11, 1957 at Easton, Md.
6. G. R. Hall and T. L. Markin, *J. Inorg. Nucl. Chem.* 4, 137 (1957).