Professor A. L. Hughes and the cyclotron group of Washington University, St. Louis, Missouri.

Note added in proof: This experiment has been recently repeated. Cacodylic acid (Kahlbaum) in the form of an aqueous solution has been irradiated by slow neutrons, the Washington University cyclotron again being the neutron source. H_2S was passed into the solution, precipitating the activated As ions as a sulfide. $AsCl_3$ and HCl had been previously added.

The results were essentially the same, γ_A and γ_B again being present in about the same relative intensities. A better value of γ_B appears, however, to be 2.00 ± 0.04 Mev. Although the source was stronger and more highly concentrated, statistical uncertainty was still rather large in the neighborhood of 3 Mev, thus preventing any conclusive statement with regard to presence or absence of γ_C. Had

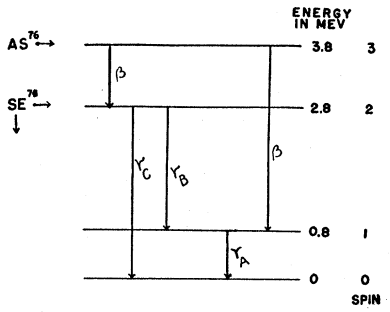

FIG. 3. Level scheme for Se^{76}.

γ_C been present with an intensity less than 20 percent of that of γ_B, it would have very probably escaped detection.

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The Search for Element No. 87

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An attempt to secure the element 87 $L\alpha_{1,2}$ x-ray lines in fluorescence spectra is made with negative results: no trace of the line sought for was found. The fluorescent material was $CsHSO_4$ derived from lepidolite, in which free thallium has been found. The writer suggests that the free thallium is derived from unstable element 87 atoms which had been present in the lepidolite molecule.

INTRODUCTION

THE status of the search for element 87 was fully discussed by the present writer in a previous paper.[1] Since then, there has been little further work done on the subject. Hulubei[2] has published x-ray spectrograms which reveal little or nothing. Moreover, the present writer's objections[1] to Hulubei's wave-length measurements still hold: the wave-lengths are far from the correct value; neither is the frequency separation of the "87" $L\alpha_{1,2}$ lines correct as given by Hulubei. As Hulubei admits,[3] they are "mésures très difficiles" and "raies très faibles."

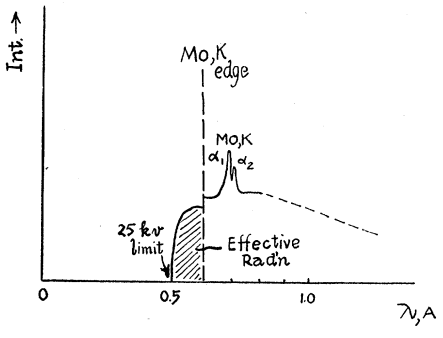


FIG. 1. Schematic spectral diagram showing effective portion of continuous spectrum emitted by molybdenum primary anticathode which might cause fluorescence of Mo portion of fluorescent target to effect registration of Mo K lines on plate.

One might possibly assume, then, that there is no evidence for the existence of element 87. There is, however, ample evidence for the existence of the unstable atom. J. A. Cranston,[4] in 1913, found that three atoms in 10^5 of $MsTh_2(89)$ emitted alpha-particles to form element 87 (see Fig. 4). He used the Geiger-Nuttall method. G. Guében,[5,6] of Liège, using the Geiger-Nuttall

* Now at the University of Southern California.

[1] F. R. Hirsh, Jr., Phys. Rev. **51**, 584 (1937).

[2] H. Hulubei, Comptes rendus **205**, 854 (1937).

[3] H. Hulubei, Comptes rendus **209**, 675 (1939).

[4] J. A. Cranston, Phil. Mag. **25**, 712 (1913).

[5] G. Guében, Ann. Soc. Sci. de Bruxelles **52**, 66 (1932).

[6] G. Guében, Ann. Soc. Sci. de Bruxelles **53**, 115 (1933).

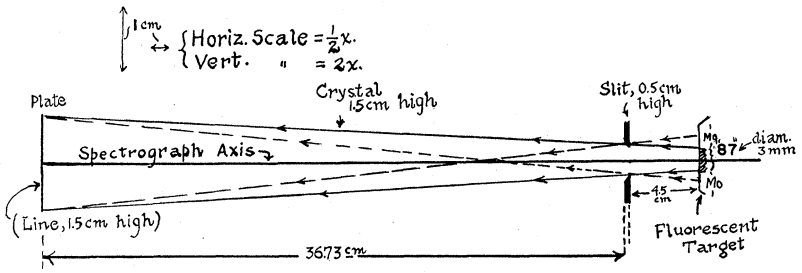


FIG. 2. Vertical section through spectrograph (note exaggerated scales) drawn from measured dimensions of spectrograph, and proving that both Mo K lines and element 87 $L\alpha_{1,2}$, produced by fluorescence, should register on photographic plate, since alignment is correct.

method, showed the existence of alpha-particles, estimated that one atom in 10^6 to 10^{10} atoms of $MsTh_2$ disintegrated to give alpha-particles and form an atom of element 87, instead of proceeding in the usual manner, with negative electron emission to form RaTh(90). He definitely showed that the alpha-particle activity had a half-life the same as that for the beta- and gamma-activity for $MsTh_2$. Thus the discovery of the unstable atom of element 87 is due to Cranston and Guében, with ample supporting evidence.

Paneth[7] has very recently reported the possible formation of element 87 by alpha-particle emission from Ac(89), forming radioactive AcK(87) of atomic weight 223. Meyer, Hess, and Paneth (see reference 7) found the alpha-activity, which was also found by Mlle. Perey. They estimate that one atom in one hundred of Ac(89) disintegrates by alpha-emission.

The present writer, however, at the instigation of Dr. T. G. Kennard of Claremont Colleges, California, who had an element 87 concentrate to be tested, decided to make a final search for the stable atom.

EXPERIMENTAL

In order to conserve the concentrate for further investigation, it was decided to attempt to excite the L x-radiations of element 87 by fluorescence. The writer has previously described the fluorescence x-ray tube[8] which was used. The Mo primary x-ray anticathode was operated at a potential of 25 kv, d.c. Since the Mo K radiation is excited at 20 kv, there should be a considerable beam of Mo K radiation leaving the primary x-ray anticathode and incident on the fluorescent target. The wave-length of the Mo $K\alpha_{1,2}$ lines is 0.707A; the L_{III} absorption edge of element 87, secured by interpolation from a Moseley graph, is located at 0.812A. Thus the Mo K radiation lies on the high absorption side of the L_{III} edge of element 87, and 87, $L\alpha$ should be strongly excited ($L\alpha$ comes from the transition $L_{III}M_V$).

The fluorescent target was made of Mo, with a three-mm hole in the center, into which the concentrate for element 87 was gently melted. The Mo-continuous radiation at 25 kv should excite the Mo K radiation, from the fluorescence target, as reference lines. The reference lines did not register on the photographic plate: this is not unreasonable, however, as very little of the continuous radiation is able to excite Mo K radiation (see Fig. 1). The shaded portion of the continuous spectrum alone can excite Mo K radiation by fluorescence. The short wave-length cut-off of the continuous spectrum is at 25 kv. Remembering the rule of 12.3 kv/A, we find the cut-off lies at $12.3/25 \approx 0.5$A. Mo K radiation is excited at 20 kv : $12.3/20 \approx 0.6$A. The Mo K radiation is at 0.7A approximately. Thus it will be seen that the Mo K reference lines should not be intense if they were registered. Lack of conditioning time for the x-ray tube made impossible the use of a higher potential for the primary anticathode. The axial alignment of the fluorescent target was excellent: this was done by rotating the glass insulating sleeve while the wax seal was hot. Figure 2, carefully constructed from the spectrograph and tube dimensions, shows that the photographic plate sees both top and bottom parts of the Mo part of the fluorescent

[7] F. Paneth, Nature **149**, 565 (1942).
[8] F. R. Hirsh, Jr. and F. K. Richtmyer, Phys. Rev. **44**, 955 (1933).

target, hence the Mo K reference lines should register if the fluorescence were intense enough. X-radiation was scattered by the fluorescence target, however, as evidenced by the presence of a central beam on the photographic plate (see Fig. 3). In this case:

$$h\nu = h\nu' + E\nu' + mc^2/2\,[(1-\beta^2)^{-\frac{1}{2}}-1] + MV^2/2,$$

where $h\nu$ is the original photon, $h\nu'$ the scattered photon, $E\nu'$ the recoil electron ejection energy, $mc^2/2\,[(1-\beta^2)^{-\frac{1}{2}}-1]$ the recoil electron kinetic energy, and $MV^2/2$ the energy of motion imparted to the atom.

Previously, under approximately the same operating conditions, an intense fluorescence plate had been secured in 10–20 hours. The plate used for testing the presence or absence of element 87 in the concentrate was exposed for 36 hours; the primary and fluorescence targets were cleaned hourly to minimize the absorption of tungsten deposits from the filament, which was run at an emission current of 15 ma, the primary anticathode was operated at 25-kv positive potential. No trace of element 87 $L\alpha_{1,2}$ lines was present on the photographic plate.

We conclude that element 87 was not present in the $CsHSO_4$ concentrate from lepidolite. Although this simple experiment shows that element 87, in the stable form, was not present in the lepidolite from which the $CsHSO_4$ was made, it does not prove it had not been present. In fact,

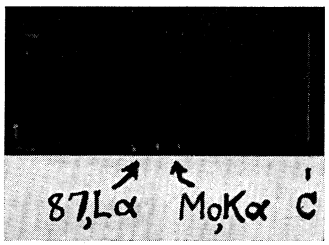


FIG. 3. Spectrogram showing registration of central (scattered) beam of x-radiation, and absence of element 87 lines. A quartz crystal was used: the Bragg angles for Mo, $K\alpha$ ($\lambda=0.707$A) and 87, $L\alpha$ ($\lambda=1.045$A) are 5° and 7°, respectively (approximately). (The plate faces in the direction from which the scattered x-rays came.)

Kennard and Rambo[9] have reported the presence of free Tl in this lepidolite molecule. The writer wishes to suggest that it is evidence that unstable element 87 had been present there. $MsTh_2(89)$ is derived by negative electron emission from

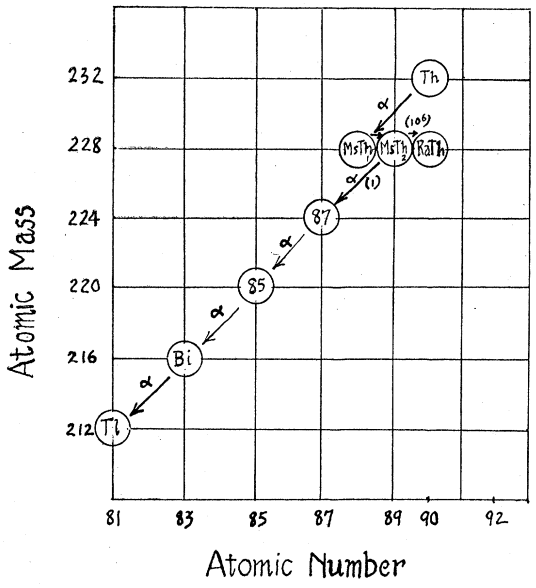


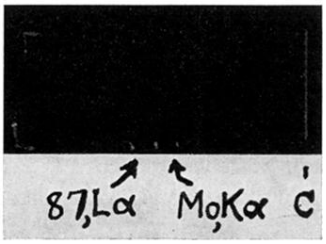
FIG. 4. Disintegration chart showing possible origin of Tl found in the lepidolite molecule. Atomic mass plotted against atomic number for the $MsTh_1-MsTh_2$ chain of disintegrations.

$MsTh_1(88)$, which in turn is derived by an alpha-emission from Th(90), (see Fig. 4). $MsTh_2(89)$ goes down by an alpha-emission to element 87; another alpha-emission produces element 85; two more alpha-emissions would produce Tl(81) of mass 212, and would account very nicely for the presence of Tl in the lepidolite molecule.

ACKNOWLEDGMENT

The writer especially wishes to thank Professors R. C. Gibbs and L. G. Parratt, at whose invitation this search was performed in the Cornell University Department of Physics, during the spring of 1941.

[9] T. G. Kennard and A. I. Rambo, Am. Mineral. **18**, 454 (1933).



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