Det Kgl. Danske Videnskabernes Selskab.
Mathematisk-fysiske Meddelelser. **VII**, 11.

# ON THE MISSING ELEMENT 87

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KØBENHAVN
HOVEDKOMMISSIONÆR: ANDR. FRED. HØST & SØN, KGL. HOF-BOGHANDEL
BIANCO LUNOS BOGTRYKKERI
1926

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Between hydrogen and uranium three elements are still missing, namely those of atomic numbers 61, 85 and 87. Whilst the first element[1] belongs to the rare earth group, the two later ones lie in a part of the periodic system which contains only radioactive elements. Mercury is the heaviest element composed only of inactive isotopes; of the next three elements thallium, lead and bismuth, both radioactive and inactive isotopes are known, while from polonium upwards all the known isotopes show radioactive properties. When searching for the missing elements 85 and 87 we have thus first of all to consider the known series of radioactive disintegration and to investigate if the elements can be found as branch products. Let us first consider the element 87, which can either be the product of an $\alpha$-disintegration and have then its origin in the element 89, or the product of a $\beta$-disintegration in which case the parent element would have the atomic number 86. We have consequently to investigate if any of the known isotopes of 89 emit $\alpha$-rays, or any of the element 86 $\beta$-rays. Only two isotopes of 89 are known,

[1] Several fractions prepared by crystallisation of samarium and of neodymium preparations sent by Baron AUER VON WELSBACH, Dr. PH. HÖRNESS and Prof. L. ROLLA were investigated in this laboratory by the X-ray method. The very careful investigation carried out by Dr. NISHINA failed to reveal the presence of the element 61 in these fractions.

namely mesothorium and actinium and three of the element 86, namely radon, thoron and acton.

## The search for $\alpha$-particles emitted by MsTh2.

MsTh 2 emits during its disintegration, which has a half-value period T = 5,89 hours[1], hard $\beta$- and $\gamma$-rays. We purified by chemical methods this radio-element of all its disintegration products and tested whether any scintillations produced by $\alpha$-rays possibly emitted by the MsTh 2 could be observed. We have then to compare the number of the observed scintillations with those calculated on the assumption that the scintillations observed are solely due to the emission of $\alpha$-particles by the RaTh produced and its successive products. In the first few hours it is sufficient to take into account the $\alpha$-particles emitted by RaTh alone, after the lapse of three hours for example the number of $\alpha$-particles emitted by ThX will only be 1 % of those emitted by RaTh. But it has to be noticed that ThX comes practically at once into radioactive equilibrium with ThEm (T = 54,5 sec.) and ThA (T = 0,14 sec.) and therefore both these elements will increase the number of scintillations due to RaTh by 1 %. The number of ThX atoms (X) present after time t can be calculated from the formula

$$X = A\lambda_1\lambda_2\left[\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda)(\lambda_3-\gamma_1)}+\frac{e^{-\lambda_2 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_2)}+\frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right]$$

where A is the number of MsTh2 atoms originally present, $\lambda_1$, $\lambda_2$ and $\lambda_3$ are the radioactive constants of MsTh 2, RaTh and ThX ($3{,}2\cdot10^{-5}$, $1{,}16\cdot10^{-8}$ and $2{,}2\cdot10^{-6}$ sec.$^{-1}$). The number of scintillations caused by $\alpha$-particles expelled by ThX is proportional to X.

[1] W P. Widdowson and A. S. Russell, Phil. Mag. **49**, 137, 1925.

So long as we consider only the first few hours after the preparation of the pure MsTh 2, the curve indicating the rise of the number of scintillations will be identical

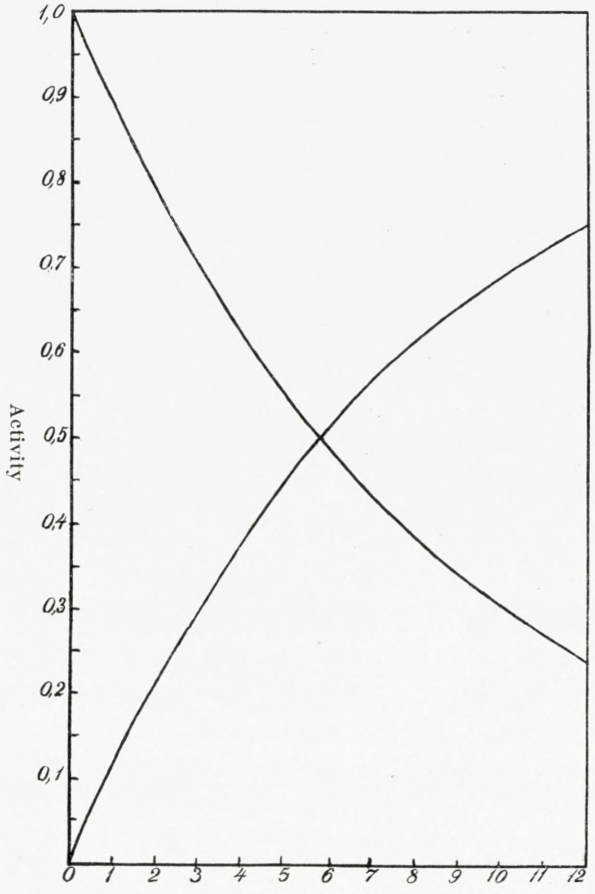


Fig. 1.

with the rise curve of MsTh 2, shown by Fig. 1. This clearly follows from the fact that the decay of every MsTh 2 atom gives rise to the formation of a RaTh atom and that the rise and decay curve of an element are reciprocal. The number of scintillations calculated on the assumption that they are solely due to the emission of $\alpha$-particles from the disintegration products of MsTh 2 and the number

of scintillations observed are seen in table 1, the later being the average of 10 sets of counting.

| Time in hours | Scintillations observed | Scintillations calculated |
|---|---|---|
| 1,23 | 13 | 13 (assumed) |
| 2,16 | 22 | 20 |
| 3,05 | 32 | 35 |
| 4,32 | 45 | 40 |

From the good agreement between the observed and calculated values we can conclude that our MsTh 2 preparation emitted practically no $\alpha$-particles immediately after its purification. To be able to make a more definite statement let us compare the number of scintillations observed after 1 hour with the number we should have observed if MsTh 2 could disintegrate with emission of $\alpha$-particles. The emission of $\alpha$-particles will in the later case be proportional to $0{,}89\,\lambda_1$, and in the former to $0{,}11\,\lambda_2$, the ratio of these values being equal to $2 \cdot 10^4$. Should therefore only $^1/_{20000}$ of the MsTh 2 atoms present disintegrate with expulsion of $\alpha$-particles, we should have observed twice as many scintillations as we did. Allowing for an error of experiment of 10 %, we can conclude that, if any MsTh 2 atoms disintegrate with expulsion of $\alpha$-particles, their number must be less than $^1/_{200000}$ of the disintegrating atoms.

We shall now describe shortly the preparation of the MsTh 2. A preparation of mesothorium, the $\gamma$-activity of which corresponded to that of about 1 mg. of radium, was dissolved in very dilute nitric acid and some 30 mg. of nitrate of thorium and the same amount of nitrate of barium was added. Then, by adding carbonate-free ammonia to this solution, all the radioelements present, except MsTh 1 and ThX, were precipitated. After the lapse of 6 hours, first 30 mg.

$La(NO_3)_3$ were added to the solution containing the MsTh 1 and then carbonate-free ammonia. The precipitate of MsTh 2 obtained was redissolved twice in acid and precipitated with ammonia to ensure a perfect absence of MsTh 1. The MsTh 2 was then dissolved in dilute nitric acid containing some thorium purified from disintegration products and the latter precipitated with $H_2O_2$. Then in the presence of some barium salt the ThX present was removed as sulphate and, after adding some $Pb(NO_3)_2$ and $Bi(NO_3)_3$, by leading a stream of $H_2S$ through the solution we removed also the ThB and ThC present. Every operation was repeated three times and carried out as quickly as possible. The solution ultimately obtained containing MsTh 2 only was evaporated very quickly and the scintillations produced by the preparation were counted in the usual way.

In the experiment described we did not search for the element 87 directly but tried to ascertain if the disintegration of some MsTh 2 atoms leads to the formation of atoms of the element 87. This method has the advantage that if at least $^1/_{200000}$ of the MsTh 2 atoms gives rise to an 87 atom, we can prove the existence of the latter, independently of the period of decay of the element 87. Another line of attack is to try to find the presence of a radioactive body in a solution of a mesothorium preparation from which all known radioelements have been removed by chemical treatment. Quite recently O. HAHN[1] has carried out such experiments. A strong mesothorium preparation, corresponding to 45 mg. radium, was used, and the solution contained a caesium salt, to prevent the removal of the element looked for by adsorption and similar processes. HAHN concludes that if the half value

[1] O. HAHN, Die Naturwissenschaften **14**, 158, 1926.

of the product sought for is not shorter than 2 hours and the branching ratio of MsTh 2 $\left(\frac{\alpha}{\beta}\right)$ is not more than $10^{-4}$, the element 87 would not have escaped detection, while assuming a half value of 10 years he would have detected the product even if the branching had been less than $10^{-7}$. Besides from MsTh 2, the element 87 could also originate from actinium by an $\alpha$-disintegration. Lacking a suitable actinium preparation the author could not determine the scintillation rise curve of actinium purified of all disintegration products.

## Search for the element 87 as a disintegration product of radon.

If some of the decaying radon atoms emitted $\beta$ instead of $\alpha$-particles, atoms of the element 87 would be produced. Since the decay of radon gives rise to a series of products emitting also $\beta$-rays there is very great difficulty in proving the possible presence of a very slight $\beta$-radiation due to the transformation of radon into 87. We have therefore abandoned the course followed in the case of MsTh 2 and tried to find directly a $\beta$-disintegration product of radon. A tube containing about 100 mg. radon has been pulverised after 3 days and treated with boiling diluted nitric acid, containing 50 mg each of $Cs(NO_3)$, $Pb(NO_3)_2$ and $Bi(NO_3)_3$. By boiling the solution the last traces of radon were removed and the solution was then treated with $H_2S$. To the filtered solution more lead and bismuth salt was added and the treatment with $H_2S$ repeated twice more. The activity of the evaporated solution was $^1/_{10}{}^6$ that of the active deposit removed and the remaining activity decayed with a period of 27 minutes, very nearly that of

RaB. If only $^1/_{10}{}^6$ part of the radon had disintegrated into a body having a half value not less than a few minutes, and if this were not an element emitting $\beta$-rays with a life of more than a few years[1], we would have had no difficulties in ascertaining its presence. No similar experiments were carried out with thoron and acton. Should the disintegration of these lead to a $\beta$-product Tn $\xrightarrow{\beta}$ 87 $\xrightarrow{\beta}$ . . . , the life of the element would be (comp. footnote 1) so short, that there would be very little hope of ascertaining its presence.

## Some remarks on the element 85.

The element 87 disintegrating by emission of $\alpha$-rays would lead at once to the formation of 85. The other way by which this element could be produced would be a $\beta$-emission by the nucleus of a polonium istotope. Trials to prove the existance in the case of polonium failed, in the case of the short-living polonium isotopes such an investigation encounters very great difficulties on account of the presence of several $\beta$-radiating bodies.

## Remarks on the existence of a stable isotope of 87.

Of none of the elements preceeding radon are inactive isotopes known, for such an isotope exhaustive search was made in the case of radon (84) by F. W. Aston using the method of positive rays. It is therefore improbable that a stable isotope of an element of higher atomic number than radon can be found especially if the atomic number is

[1] As the second $\beta$-product emitted by successive disintegration has a shorter life than the first one which has in the above hypothetical case T = 3,8 days, the later possibility must he considered as a very improbable one.

odd as in the case of 87. Exhaustive trials to find this element in caesium minerals by chemical methods failed. D. Druce and F. H. Loring[1] recently claimed to have discovered the element 87 by means of X-rays in manganese sulphate. We have to wait for further evidence as the presence of a few very faint X-lines coinciding more or less with the calculated values cannot be considered as conclusive for the presence of an element.

The above argument, though with less certainty, applies also to the element 85, which preceeds emanation. The limit where the elements with only stable isotopes known cease and the series of elements with both kinds of isotopes begins is well known and is seen from the table below, but the exact limit between the latter and the elements with only radioactive isotopes is still unknown[2]. We cannot say if polonium is found in stable form or not, and it would be of great importance to search for stable polonium isotopes in tellurium, bismuth or antimony minerals.

Table 1.

| | | |
|---|---|---|
| Au, Hg. only stable isotopes. | Tl, Pb, Bi. stable and radioactive isotopes. | Po, 87,85,Em. Ra... ?? Only radioactive isotopes. |

## Summary.

1. As only radioactive isotopes of the elements succeeding radon (86) are known, it is not probable that inactive isotopes of the missing element 87 can be discovered. This element having an odd atomic number we can only expect to find it as a comparatively short living body.

[1] F. H. Loring, Nature, **117**, 153, 1926.

[2] O. Hahn (l. c.) considers the existence of a stable isotope of radium and polonium to be possible.

2. Trials were made to discover by the scintillation method an $\alpha$-radiation emitted by MsTh 2. The emission of an $\alpha$-particle by this element 89 would lead to the formation of the element 87. It was found that, if such a disintegration occurs at all, fewer than $^1/_{200000}$ atoms of MsTh 2 can disintegrate in this way.

3. Trials to find 87 as a $\beta$ product of radon, by removing chemically all the known members of the active deposit of 100 millicurie failed.

4. It is known that the series of elements having both radioactive and inactive isotopes begins after mercury. Of thallium, lead and bismuth both kinds of isotopes are known. The limit between those with both kinds of isotopes and these with only radioactive isotopes known is uncertain. It is suggested that it may be possible to find an inactive isotope of polonium and the possible formation of the element 85 is discussed.

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