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## From Fermi Transuranic Elements to Nuclear Fission Physical and chemical aspects

In this paper, we will try to reconstruct the birth of Fermi hypothesis of transuranic elements, and the path starting from this idea and leading to the discovery of nuclear fission.

The hypothesis of transuranic elements is a consequence of the discovery of neutron induced artificial radio-activity, made by Enrico Fermi, working alone, in March 1934 [6], under the influence of his theory of beta decay [12].

By bombarding some elements (Platinum, Copper, Aluminum, Fluorine, ...) with a neutron source, he found that Aluminum and Fluorine became radio-active, with mean lives of twelve minutes and ten seconds, respectively.

Fermi interpretation for the case of Aluminum was the following  $(n, \alpha)$  activation reaction

$$(n, \alpha): Al_{13}^{27} + n_0^1 \rightarrow Na_{11}^{24} + He_2^4,$$

where a new radio-active isotope of Sodium is created, which beta decays in a stable isotope of Magnesium

$$Na_{11}^{24} \rightarrow Mg_{12}^{24} + e^{-} + v.$$

A similar reaction was proposed for Fluorine.

With this discovery, a new chapter of research is open, trying to see whether there are other substances to be activated, and whether the activation reaction is only  $(n, \alpha)$ , or there are other reactions.

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Starting from April, a large research group was formed, including the Chemist Oscar D'Agostino, and moreover Edoardo Amaldi, Emilio Segrè and Franco Rasetti [1].

A systematic exploration of the periodic table was made in the period April-July 1934.

63 elements were irradiated, and 38 were found to be activated!

As a result of chemical analysis, it was possible to find three reactions taking place in neutron bombardment, with emission of an alpha particle, a proton and a gamma ray, respectively

$$(n, \alpha): X_Z^A + n \to X_{Z-2}^{A-3} + \alpha,$$
  
$$(n, p): X_Z^A + n \to X_{Z-1}^A + p,$$
  
$$(n, \gamma): X_Z^A + n \to X_Z^{A+1} + \gamma.$$

Moreover, it was found that only  $(n, \gamma)$  reactions where possible for heavy irradiated elements, with  $Z \ge 30$  [7].

This general scheme of interpretation was very strong, well founded on the previous Fermi theory of  $\beta$  decay, with validity along all periodic table, Uranium included [8].

Obviously the study of Uranium was more complicated. It was necessary firstly to eliminate the products of natural radioactivity.

In May induced radio-activity was found, with many decay periods, one minute, thirteen minutes, plus others not well identified. After careful chemical analysis on the thirteen minute active element, Fermi and his coworkers could conclude that it was not an isotope of the elements from Uranium Z = 92 to Lead Z = 82.

The active element was found to precipitate together with Rhenium Sulphide. Since Rhenium was homologue to the hypothetical Z = 93 element, according to the periodic table of the times, the conclusion was that the thirteen minute element was the transuranic element Z = 93! [9]

Also a further transuranic element Z = 94 was introduced, in order to give the interpretation of the other long period. These new elements were called Ausonium,  $Ao_{93}$ , and Hesperium,  $Hs_{94}$  [10], [17].

After the discovery of the effects of slow neutrons, research on Uranium were refined. The final conclusion in Rome, in May 1935 [4], is summarized in the  $(n, \gamma)$  activation reaction

$$(n, \gamma): U_{92}^{238} + n \rightarrow U_{92}^{239} + \gamma,$$

followed by the chain of  $\beta$  decays

$$U_{92}^{239} - {}_{15''}^{\beta} \to Ao_{90}^{239} - {}_{13'}^{\beta} \to Hs_{94}^{239} - {}_{100'}^{\beta} \to X?.$$

It is a triumph for Fermi interpretation scheme!

After, the interests of Fermi group in Rome concentrate on the study of other problems, in particular on the slow neutrons [11].

The problem of transuranic elements, starting from the beginning of 1935, until the end of 1938, is at the center of interests of Otto Hahn and Lise Meitner in Berlin, and moreover of Irene Curie in Paris, in 1937-38.

Researches of Hahn and Meitner fully confirm Fermi interpretation. Ausonium and Hesperium are called EkaRhenium and EkaOsmium, in order to stress their chemical affinity with lower homologue. Many more periods were found, and many more transuranic elements were identified by the technique of the lower homologue [13].

In order to solve the puzzle, they used the newly discovered phenomenon of isomerism, according to which the same isotope can have different decay rates. So they could build three different series of transuranic elements, all coming from the  $(n, \gamma)$  activation of Uranium, according to Fermi scheme [18].

The elements were called EkaRhenium, EkaOsmium, EkaIridium, EkaPlatinum, EkaAurum?. The isomeric series were

$$U_{92}^{239} - {}_{10''}^{\beta} \rightarrow EkaRe_{93} - {}_{22'}^{\beta} \rightarrow EkaOs_{94} - {}_{59'}^{\beta} \rightarrow EkaIr_{95} - {}_{66br}^{\beta} \rightarrow EkaPt_{96} - {}_{25br}^{\beta} \rightarrow EkaAu_{97}?,$$
$$U_{92}^{239} - {}_{40''}^{\beta} \rightarrow EkaRe - {}_{16'}^{\beta} \rightarrow EkaOs_{94} - {}_{57br}^{\beta} \rightarrow EkaIr_{95},$$
$$U_{92}^{239} - {}_{23'}^{\beta} \rightarrow EkaRe_{93}?$$

At the end of July 1938, a last transuranic element is added to the second series, EkaPlumbum, coming from a 60 day decay of EkaIridium [14].

This is the very complex, but perfectly consistent, isomeric transuranic castle devised by Hahn and Meitner.

However, new measurements made by Irene Curie and coworkers, in Paris, made necessary a further complication of the picture.

In May 1937, Irene Curie and Savitch begin the study of Uranium, and introduce a new techique of measurement of induced radio-activity [2]. In this way, they found a new active body, with a decay rate of around 3.5 hours, called  $R_{3.5hr}$ . The discovery of this new active body will be a fundamental landmark, because it will cause the shift of attention from the unknown region of transuranic elements, to the well known region of periodic table below Uranium.

In fact, they found that the new body shared the properties of rare earth elements, and is carried down with Lanthanum in precipitation. Since Lanthanum is the lower homologue of Actinium Z = 89, it was natural to identify the new body

with Actinium. Before accepting this identification, they attempt a fractional precipitation of a mixture  $(La+R_{3,5br}+Ac)$ , in order to differentiate the substances.

This is the first time that the procedure of fractional precipitation is employed in the field of *artificial* radiochemistry! This technique will play a major role, in the hands of Hahn and Strassmann, for the discovery of fission!

With this method, they found that the new active body separates from Actinium. Therefore it can not be Actinium!

After these results, their conclusion is that  $R_{3.5b}$  must be a transuranic, of very difficult location indeed [3]. In the transuranic region one could not foresee properties of rare earth elements!

About the nature of this new and strange "transuranic", Otto Hahn and Fritz Strassmann, in November 1938, communicated their results [15].

They were able to confirm the new Curie-Savitch body, with an decay rate corrected to 4 hours, but they found also five more different periods, given approximately by 2.5 minutes, 40 minutes, 110 minutes, 60 hours, several days.

In their November 1938 paper, they give a general scheme for the six activities based on chemical evidence, of precipitations with Barium (homologue of Radium) and Lanthanum (homologue of Actinium).

The previously established transuranic series are left unchanged. Three new series are introduced based on isomerism of Radium and Actinium

$$\begin{aligned} & Ra_{88}^{231} - (2.5min) \to Ac_{89} - (40min) \to Tb_{90}?, \\ & Ra_{88}^{231} - (110min) \to Ac_{89} - (4bours) \to Tb_{90}?, \\ & Ra_{88}^{231} - (severaldays) \to Ac_{89} - (60bours) \to Tb_{90}?, \end{aligned}$$

This general picture, if satisfactory according to the chemical analysis done in Berlin, was quite in conflict with established physical facts. In fact it required that neutron irradiated Uranium should emit two successive alpha particles, passing over Thorium (Z = 90), in order to become Radium, i.e. the reaction was

$$(n, 2\alpha): U_{92}^{238} + n \rightarrow Ra_{88}^{231} + \alpha + \alpha.$$

This reaction was in conflict with Fermi general interpretation, and moreover was difficult to imagine from an energetic point of view. How could be that a neutron (even a slow neutron!), could kick off two alpha particles from the nucleus.

Moreover, G. Frh. v. Droste, a former coworker of Meitner in Berlin, did not find experimental evidence for the emission of alpha particles [5].

It is clear that the central point was the presence of Radium. However, now research had been shifted from the transuranics region to a region well known in the periodic table, where all sophisticated chemical techniques were available. Further research by Hahn and Strassmann, in December 1938 [16], led to a final proposed picture involving now four isomeric species for Radium and four for Actinium, according to the following chains of beta decays

Notice that the "Radium" species (in quotation marks!) are identified in the paper as "alkaline earth metals".

In fact, immediately they report on the astonishing fact that the well known techniques of fractional crystallization and fractional precipitation did not allow them to separate the "Radium isotopes" from Barium. Analogously for "Actinium" with respect to Lanthanum.

They say "As Chemists we really ought to revise the decay scheme given above and insert the symbols Ba, La, Ce in place of Ra, Ac, Th."

The part of the castle connected with the four isomeric "Radium" chains falls in pieces. But now, with the proof that lower species are formed, also all reasoning leading to transuranic elements must be revised.

They say "The "transuranic group" of elements are chemically related but not identical to their lower homologue, Rhenium, Osmium, Iridium and Platinum. Experiments have not been made yet to see if they might be chemically identical with the even lower homologue, Technetium, Ruthenium, Rhodium and Palladium. After all, one could not even consider this as a possibility earlier. The sum of the mass numbers of Barium + Technetium, 138 + 101, gives 239!"

Also the transuranic castle falls to ruin, and nuclear fission has been discovered!

In conclusion, Fermi hypothesis of transuranic elements played the important role of raising the problem of identifying the active products of Uranium neutron irradiation. The path to fission was very tortuous. Chemical and physical aspects were strongly intermixed.

Between 1934 and 1938, an enormous transuranic castle was build, where nuclear transmutations, starting from Uranium, extended in the unknown region of the periodic table up to Z = 96, and beyond. In this region, chemical methods could give only indirect evidence.

With the discovery of the new Curie-Savitch element  $R_{3.5b}$  there was a turning point. Transuranic elements are no more sufficient. It is necessary to build a new castle of isomeric species, extending in the known region of the periodic table.

This new castle is not very solid from a physical point of view (the two alpha problem). However, efficient work can be done on the chemical side.

In one month this new castle falls in ruin, carrying with itself also the transuranic construction.

Most of the active elements should be recognized as products of the fission.

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