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CYCLOTRON INVESTIGATION HEIDELBERG



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COMBINED INTELLIGENCE OBJECTIVES
SUB-COMMITTEE

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Report on
CYCLOTRON INVESTIGATION
HEIDELBERG, GERMANY

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U. S.

on behalf of the
U.S. Technical Industrial Intelligence Committee

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Subject: Cyclotron investigation at Heidelberg

TO : CIOS Secretariat

Investigators: Mr. C. W. Hansell, TIIC
Lt. Col. J. J. Slattery
Maj. J. M. Sanabria

The above investigators were accompanied by Dr. Max Knoll, a German scientist of the Telefunken Co., who had been separated from his laboratories in and around Liebenstein and who had attached himself to the headquarters of the Ferdinand Braun Institute at Brannenburg. He was released by the local military government at Rosenheim for the trip to Bad Liebenstein and vicinity. It was he who supplied names of scientists and institutions in which the investigators had a special interest.

It had been learned at Munich from Mr. Harry Marvin of the General Electric Co., another investigator, that the Bad Liebenstein area had been investigated by him but some of the targets had been overlooked. It was therefore agreed with Col. Jackson and Major Johnson-Ferguson at Munich that it would be proper to proceed to the Telefunken Laboratories in the Bad Liebenstein area with Dr. Knoll as guide and interpreter.

Interview with Prof. Walter Bothe, Kaiser Wilhelm Institute for Physics and Medicine, Heidelberg.

Since the route taken by the investigators passed through Heidelberg, advantage was taken of the opportunity to call on Dr. Walter Bothe, Im Backerfeld 6, Heidelberg, an internationally well-known nuclear physicist, who has been operating a cyclotron at Heidelberg. The cyclotron installation was shown to us by Dr. Bothe. He stated a Dr. Hogness of T Force had

asked for his reports but none were available as they had been previously destroyed.

According to Dr. Bothe, his activity with the cyclotrons during the war had been confined mostly to the preparation of tracer materials for use in biological research directed by Dr. Richard Kuhn of Wilckenstrasse 23, Heidelberg. No attempt was made to contact Dr. Kuhn because he was away from Heidelberg on that day and had already been questioned by a large group of investigators. No new particles or other outstanding discoveries were made with the cyclotron during the war according to Dr. Bothe.

Dr. Bothe said that, during the war, continuation of his work was permitted at first because the authorities had hope that the scientists might quickly learn to release and use atomic energy, even though the scientists secretly held no such hope. Later the authorities concluded that pure research was necessary and should be continued for its own sake.

As the Allied Armies swept toward Heidelberg, Dr. Bothe received orders from the German authorities to destroy all his scientific records and was forced to comply. The German Military personnel intended to destroy the laboratory but were dissuaded only with great difficulty and personal danger to Dr. Bothe. His cyclotron installation came thru the combat period substantially intact and in working order. It is still apparently complete except for a 150,000 volt X-ray type rectifier which had been removed by a Major (Br) Signals. This rectifier was used to supply the potential to draw out the accelerated proton beam to the target material.

Some of the bombarded tracer materials supplied by Dr. Bothe to Dr. Kuhn which were mentioned, included radioactive phosphorus, copper and bromine. Bombarded bromine, in particular had been used as tracer in a bromine compound intended for uses similar to penicillin. It was stated that the Germans had not developed penicillin production but that Dr. Kuhn had produced small amounts for research purposes.



The cyclotron appeared to have no outstanding characteristics to distinguish it from those most commonly used in the United States. The steel magnet had been supplied by Krupp and the water cooled magnet coils by Siemens. There were ceramic insulator seals for the drive circuit, placed close to the D's. The drive circuit was of two 4-inch diameter copper tubes, provided with a short circuiting slider for tuning and tapped by an open wire transmission line for introducing the drive power.

The magnetic field current was supplied by a generator on flexible mountings and required 53 KW.

Two Apiezon oil diffusion pumps and a mechanical pump in series provided the vacuum. There were Pirani and ionization vacuum gages, and as usual, there had been great difficulty with vacuum leaks, particularly because the vacuum chamber was made of cast material. There were no clever tricks for finding leaks. The usual method seemed to be that of putting the system under pressure and looking for leaks with soapy water.

The only flash-over protection provided seemed to be circuit breakers in the power circuits. Constant current circuits in the radio frequency circuits or power circuits were not used according to Dr. Bothe.

The radio frequency drive equipment was designed for 70 KW output power in the range of 15 to 30 meters wave length. It was provided with wave switching and frequency adjustment. It had been built by Ader and Schwartz, a small firm at Munich. It employed a conventional push-pull tuned grid tuned anode circuit arrangement and was designed for Telefunken water cooled tubes. It was supplied with direct current power from a continuously pumped tank type mercury rectifier, 6 phase, 12,000 volts, 180 KW, made by Brown Boveri at Mannheim.

The cyclotron was said to be capable of producing particles with 9,000,000 volts energy. A water barrier of approximately four feet thick proved inadequate and it was necessary to construct a control panel at another point some 75 feet away.

Professor Bothe had visited cyclotron installations in the United States. He seemed to be familiar with Professors Lawrence, Dunning and others

as well as with their equipment. He was familiar with the magnetic induction accelerator and said that he had started constructing one but this work was interrupted by the war.

An inconsequential but interesting item of information is that, although Dr. Bothe has been working for years on the frontiers of science he has never owned nor driven an automobile but gets about on foot or by bicycle.

3 enclosures:

Künstliche Radioaktivität durch thermische Neutronen bei den seltenen Erden.

Report on the work done during the war by the Institute for Physics of the Kaiser Wilhelm Institute for Medical Research. (Bericht über die Arbeit des Instituts während des Krieges)

Die Mindestgrösse der U-Maschine (Auszug)

INSTITUT FÜR PHYSIK

Heidelberg

June 26, 1945

Report on the work done during the war by the Institute for Physics of the Kaiser Wilhelm Institute for Medical Research

This abstract includes all investigations undertaken by our Institute since September 1939. In several cases, however, the numerical data can only be given approximately, because the Institute is closed for all its personnel including the Director. The same holds for the branch laboratory erected for reasons of air protection of Tauberbischofsheim, where also reprints and records of unpublished work and the private library of the Director are kept. This report is not intended for publication.*

I. The Heidelberg cyclotron

A full size cyclotron has been constructed since 1938. The donators were:

Stifterverband der Deutschen Forschungsgemeinschaft,
Helmholtz-Gesellschaft zur Förderung der physik-
alischen Forschung,
Kaiser Wilhelm-Gesellschaft zur Förderung der
Wissenschaften,
Max Planck-Stiftung,
Geheimrat C. Bosch (I.G. Farbenindustrie),
Badisches Kultusministerium,
Reichsforschungsrat.

About 90% of the costs came from private sources. No prescriptions nor restrictions of any kind regarding the working program of the cyclotron were connected with these grants. The cyclotron has not been used for any work concerning war problems.

* By military order the "secret reports" had to be destroyed. To avoid destruction, preparations had been made to bury these reports, but this proved not to be practicable, because there were always some unreliable persons on the premises.

The magnet has been constructed by Krupp and Siemens, the high frequency part by Rohde and Schwarz (München), the rectifier, the machine for exciting the magnet and the general installment by Brown, Boveri and Cie. The acceleration chamber has been designed by Dr. Gentner and built in our work shop.

Data: Diameter of the pole pieces 101 cms
weight of the magnet 80 tons
high frequency power 70 KW
maximum deuteron energy 12-13 MeV

The design of the whole plant followed closely that given by Prof. E. O. Lawrence of the University of California, to whom we are indebted for much valuable advice (the Director and his first assistant Dr. Gentner were guests of Prof. Lawrence at Berkeley in 1939). Further practical experience could be gathered from the Paris cyclotron, partly in co-operation with Prof. Joliot after his return to Paris in 1940. The Paris cyclotron had been left behind in an unfinished state by Prof. Joliot. After completing it, it was used by Prof. Joliot and his staff, and by a small group of German physicists, for purely scientific research. It was settled that no war work should be done by neither side with the Paris cyclotron.

The Heidelberg cyclotron was in action from Jan. to Sept. 1944; deuteron beams of 9 MeV were produced. Afterwards some changes had to be made, because the form of the magnetic field proved not to be quite satisfactory. Just before the occupation the cyclotron had begun to run again.

A new type of ion source after the principle of oscillating electrons is under construction (Dr. A. Papkow).

II. Work connected with medical, biological and chemical problems

With regard to the protection of personnel engaged in high power radiation plants, measurements on the absorption and spatial distribution of gamma rays and

neutrons were made, mainly in the cyclotron laboratory (Dr. Gentner, Dr. v. Droste, Dr. Jensen). The results combined with theoretical considerations (Bothe) showed that a protecting wall of 1 m of water, or 1,60 m of concrete is required to suppress sufficiently the direct radiations of a 8 MV cyclotron at a distance of 8 m. But if the wall is not closed at all sides, great care has to be taken of the stray radiation scattered round the edges of the wall.

Considerable quantities of artificially radioactive material have been produced and distributed among medical, biological and chemical colleagues for tracer work, therapeutic use etc., as for instance:

Cu 64 (12 h), was used by Dr. med. G. Schubert for clearing up the copper metabolism in rabbits and dogs. G. Schubert, H. Vogt, W. Maurer, W. Riezler, Naturwiss. 31,589, 1943.

P 32 (15 d), was used by Dr. med. Röder in the Kaiser Wilhelm Institute for Psychiatry, München, for investigating the penetration of phosphorus into the brain. It was stated, that the penetration is very small, with the exception of certain small regions, where considerable quantities of phosphorus can be accumulated (published in: Die Naturwiss.).

Na 22 (3 a). Dr. Schäfer of the I.G. Farbenindustrie showed conclusively that the potassium necessary for the growth of plants cannot be replaced by radioactive sodium. Therefore biologically the natural radioactivity of potassium plays no essential part. At the same time the sodium metabolism in plants was investigated (unpublished).

Ag 111 and Br 82 were made for Prof. Wagner (physico-chemical institute of the T.H. Darmstadt) for experiments on the photographic process. It was concluded that under normal conditions in the AgBr grain only the silver ions are wandering, whereas the bromine ions are fixed (unpublished).

Unfinished work: Production of larger quantities of radioactive Fe (47 d) and P (15 d) for treatment of anaemia and leucaemia has been prepared since a long time. Dr. Stodtmeister (Medizinische Klinik

der Universität Heidelberg) planned some research work on the different forms of anaemia, using the radioactive iron produced by our cyclotron. Experiments on neutron therapy were also on the working program of the cyclotron laboratory. - It was also planned to introduce radioactive Br^{82} into the new therapeutic substance 3065 discovered by Prof. Kuhn (K.W.I. Heidelberg, Institute for Chemistry) in order to clear up the working mechanism of this substance. - H^3 (30 a) and C^{14} (several 1000 a) have been produced till now in small quantities only; the intended biological and chemical work with these substances could not yet be begun.

III. Nuclear spectroscopy

The foundations of the spectroscopy of atomic nuclei have been laid by Bothe and Fränzl, Bothe and Becker (discovery of discrete particle groups and gamma rays produced by artificial disintegration; 1928, 1930) and Bothe and v. Baeyer (application of the coincidence method to nuclear problems; 1934). During the war this work was continued especially by Dr. Maier-Leibnitz in cooperation with Dr. Gentner and Dr. Zah-wei Ho. The technique of counting rays, and coincidences between rays, was highly refined. Exact measurements of the gamma sensibility curve of G.M. counters made of different materials have been made, the Norling curve having been extended down to energies of 50 keV. G.M. counters with sensibility either independent of $h\nu$ or proportional to $h\nu$ have been constructed. A cloud chamber with slow expansion was constructed especially for this type of work. A new type of apparatus for counting coincidences between an ionisation chamber and a G.M. counter has been developed, but was destroyed by foreign civil workers at Tauberbischofsheim, so that this work will be suspended indefinitely.

Details of disintegration processes with numerous radioactive isotopes were investigated. The following principal results have been obtained.

Na^{24} : There is a single beta ray spectrum ending at 5,4 MeV. More than one gamma ray per disintegration is emitted, the average gamma energy being ca. 4 MeV. The probable level system is discussed (ZS.f.Phys. 22, 233, 1944).

Na²²: has a single beta ray spectrum; each beta ray is coupled with one gamma ray of 1,3 MeV (published in ZS.f.Phys. 122, 233, 1944).

O¹⁹: The beta ray spectrum ends at 4 MeV. Gamma rays of ca. 1 MeV are emitted in approximately equal number with beta rays. The disintegration energy is 5 MeV. The new mass value of O¹⁹ fits well into the masses of neighbouring nuclei and with the Sargent diagram, whereas the mass value assumed before was certainly too low (published in ZS.f.Phys. 1944).

Mg²⁷: has a single beta ray spectrum ending at 1,7 MeV. Each beta ray is coupled with one gamma ray of 0,88 MeV. The disintegration energy therefore is 2,5 MeV (published in ZS.f.Phys. 1944).

Al²⁸: The limit of the beta ray spectrum is at 3,0 MeV. The gamma rays consist mainly of a strong group of ca. 1,7 MeV and a weak (ca. 6%) group of 2,5 MeV. The disintegration energy is probably $3,0 + 1,7 = 4,7$ MeV (published in ZS.f.Phys. 1944).

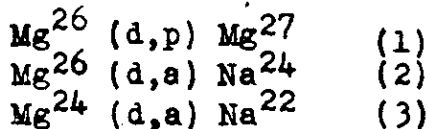
Cl³⁴: The beta ray spectrum is complex as proved by coincidence experiments. The limiting energy is 5 MeV. A complex gamma radiation was observed with an average γ of 2,3 MeV, and a maximum γ of ca. 3 MeV. The disintegration energy is 5 MeV. This leads to a mass value of Cl³⁴ fitting well into the neighbouring masses, whereas the value formerly assumed was too low (ZS.f.Phys., under press).

Mn⁵⁴: disintegration by capture only. Probably every capture process is accompanied by emission of a gamma ray of 0,6 MeV (unpublished).

Co, Zn, Ga: Unfortunately the results obtained with these elements are not available at the time being.

IV. Special nuclear reactions

A special investigation was made of the nuclear reactions



(Bothe, ZS.f.Phys., under press). The yields of the

3 radioactive products were measured as functions of the deuteron energy up to 7 MeV. From ca. 5 MeV up the yield of reaction (1) decreases; obviously reaction (1) is suppressed by reaction (2) with higher deuteron energies. The yields of reactions (2) and (3) increase in the whole region of deuteron energies, in a very similar way in both cases.

With a specially designed point counter no K radiation could be observed with Na^{22} . The number of K captures is certainly less than 1/10 the number of positrons emitted. This does not agree with theoretical predictions by Lamb.

Rare earth elements. A thorough study of radioactive isotope produced in these elements by slow neutrons has been made (Bothe, ZS.f.Phys., under press; an abstract appeared in Göttinger Nachrichten). A number of new radioactive isotopes have been observed. Some of the products emit K radiation, the origin of which was cleared up. Approximate values for the cross sections of slow neutron absorption in rare earth atoms were measured. For details cf. the enclosed reprint.

(n,2n) reactions. P. Jensen (ZS.f.Phys. 122,387, 1944) has measured the relative cross sections of several reactions of this type with different neutron sources. In general these cross sections go parallel with those determined for the corresponding (y,n)-reactions by Bothe and Gentner, but there are some significant differences. The results are in general agreement with the theory of Weiskopf and Ewing.

In this connection beryllium is of special interest, because here the threshold for the (n,2n)-reaction is very low. Fünfer and Bothe (ZS.f.Phys. 122,769,1944) observed that with Ra-Be-neutrons the neutron producing (n,2n) reaction even overbalances the neutron absorbing (n,a) reaction, whereas with Ra-F neutrons both processes are approximately equally effective in producing and absorbing neutrons.

Existence of He^5 ? P. Jensen examined the question of He^5 production by the reaction $\text{H}^2(a,p)\text{He}^5$. No indication of this reaction could be found. This result is uncompatible with previous findings of Joliot and Zlotowsky (published in ZS.f.Phys. 15,45,1940).

V. Passage of neutrons through matter

The "diffusion length" (average distance a thermal neutron travels before being absorbed) has been measured in carbon, beryllium and uranium after a method devised by Heisenberg. If the diffusion length (L) and the free path for scattering (λ_s) are known, the free path for absorption (λ_a) and therefore the absorption cross section can be calculated approximately by the Fermi formula $L = \sqrt{\frac{3}{2} \lambda_a \lambda_s}$, provided $\lambda_a \gg \lambda_s$. If, however, λ_a and λ_s are of the same order of magnitude, a more complicated formula has to be applied:

$$\frac{L}{2\lambda_s} \log \frac{L+\lambda_s}{L-\lambda_s} = 1 ; \frac{1}{L} = \frac{1}{\lambda_a} + \frac{1}{\lambda_s}$$

This exact formula has been derived by Bothe (ZS.f. Phys. 118,401,1941; 119,493,1942). The absorption cross sections determined in this way are

$(6,4 \pm 1) \cdot 10^{-27} \text{ cm}^2$ for pure carbon (Bothe and Jensen, ZS.f.Phys. 122,749,1944) $< 16 \cdot 10^{-27} \text{ cm}^2$ for Be (Fünfer and Bothe, ZS.f.Phys.122,769,1944) $6,2 \cdot 10^{-24} \text{ cm}^2$ for U (Bothe and Flammersfeld, unpublished).

The absorption cross section of Al was determined from the induced radioactivity as $0,4 \cdot 10^{-24} \text{ cm}^2$ (R. Fleischmann, unpublished). In the same way the absorption cross section of the rare earth elements were measured by Bothe (cf.IV).

The slowing down of fast neutrons has been treated theoretically by Bothe (ZS.f.Phys. 122,648,1944). Exact and approximate formulae for the spatial distribution of neutrons slowed down to a certain fraction of their original energy have been given. Experimentally the slowing down process has been investigated by P. Jensen (ZS.f.Phys. 122,756,1944) in H_2O , D_2O and carbon, after a method devised by Bothe; by this method the necessity of using large quantities of material is avoided. If B is the "Bremslänge" (average distance from the source which a fast neutron has gained when slowed down to thermal energy) the results obtained by Jensen are: $B = 45 \text{ cm}$ for D_2O (this value is probably too high); $B = 100 \text{ g/cm}^2$ for carbon. Fermi's value $B = 17 \text{ cm}$ for H_2O is assumed to be correct.

VI. Uranium fission

Like most of the other laboratories for nuclear

physics, our Institute too was engaged in experiments aiming at clearing up the details of the discovery of Hahn and Strassmann.

Nuclear cross sections of U. As mentioned in sect. V, the diffusion length of thermal neutrons in U_3O_8 was measured by a method devised by Heisenberg. A ball of U_3O_8 , immersed in H_2O could be tightly covered with cadmium sheet. The distribution of thermal neutrons in the U_3O_8 , if a neutron source was fixed in the center, was measured along a radius, with and without the Cd cover. From the difference of these two distribution curves the diffusion length was calculated. This combined with the total (absorption and scattering) cross section of U_3O_8 as measured by Whitaker gives the absorption cross section of the average U-atom, i.e. 2.10^{-24} cm^2 (Bothe and Flammersfeld, unpublished). This result was checked by P. Jensen (unpublished) by directly comparing the absorption of uranium with that of boron in solutions.

It is interesting that the same diffusion experiment repeated with crystallin U metal instead of oxyde gives a diffusion length greater than expected, probably as a result of neutron interferences (Bothe and Jensen, unpublished)

For the thermal fission cross section of the average U-atom v. Droste has found a value of $3,6.10^{-24}$ only (unpublished). The absorption cross section of pure U^{238} , as measured by production of the U^{239} activity, is very small ($0,1 - 0,2.10^{-24}$). Therefore a residual absorption cross section of ca. $2,6.10^{-24}$ remains to be explained. One possibility consists in assuming that the rare isotope U^{234} has an extremely high absorption cross section. Experiments to investigate this question were in preparation; a sample of U with a shifted isotope ratio, which had been promised us by Prof. Harteck, was to be used.

Some experiments on resonance absorption were made by P. Jensen (unpublished) comparing mixed solutions containing equal numbers of H-, N- and O-atoms, differing only by the U-contents. The number of 1 eV neutrons (Rh resonance) which have passed the resonance region of U was determined. It was concluded that the percentage of neutrons absorbed in the resonance region of U does not follow the $\sqrt{\text{concentration}}$ law expected in the case of a single resonance line, but that with higher concentrations the resonance absorption increases

in a linear way. This result is in perfect agreement with later experiments of Sauerwein at Berlin, showing that besides the resonance line at 7 eV there are broad regions of resonance absorption at somewhat higher energies. Still another type of experiment was carried through by Bothe, Flammersfeld and Jensen (unpublished): a hollow Al sphere was immersed in water, and the distribution of 1 eV neutrons in the water was measured, while a neutron source was fixed in the center. If then the sphere was filled with U or U₃O₈ the density of 1 eV neutrons near the surface of the sphere decreased in a way not compatible with the \sqrt{r} -law, but well accounted for by the existence of higher energy resonance bands.

Energy of fission neutrons. The upper limit of the energy spectrum of U-fission neutrons has been determined by several investigators with contradicting results. Therefore further experiments were made by Bothe and Gentner (ZS.f.Phys. 119, 568, 1942) using a photographic method. The fission neutrons acted on a layer of NaN₃, producing alpha particles by the N¹⁴(n,α) reaction. The range distribution of these alphas in a photographic plate was measured. It was concluded that the spectrum of the neutrons extends up to 11 MeV. This confirms the earlier results obtained by Joliot and by v. Droese, whereas Zinn and Szilard gave only 3,5 MeV as the upper limit.

Energy spectrum of fission products. An investigation carried through by Flammersfeld, Jensen and Gentner (ZS.f.Phys. 120, 450, 1943) aimed at determining not only the energy distribution of single fission products, but at the same time the ratio of kinetic energies of the two nuclei produced in one fission process. This was done by a coincidence method. The results are compared with the Bohr-Wheeler theory. A certain structure in the double distribution is indicated. For the various details of the publication.

Number of fission neutrons. Two types of experiments were carried out in order to determine the average number X of neutrons produced per thermal neutron absorbed (Bothe and Flammersfeld, unpublished). Firstly the spatial distribution of thermal neutrons round a Ra Be source was measured both in pure water and in a homogenous pulp of U₃O₈+H₂O. Secondly, a hollow sphere containing a Ra+Be source was immersed in water,

and the distribution of thermal neutrons was measured, both with the sphere empty and filled with U-powder. In both experiments the increase, produced by the U, of the total number of thermal neutrons is found by integrating. The results have to be corrected for the direct action of the fast neutrons emitted by the source and by fission, in order to obtain the increase produced by thermal fission only. By combining these two kinds of experiments, the influence of resonance absorption can be eliminated; the result is $X = 1,15$. The number of fission neutrons per thermal fission process therefore is 2,0. This result agrees with that obtained by the Heisenberg group.

Separation of U isotopes. After the discovery of Nier, that U^{235} is responsible for fission of natural U by thermal neutrons, it was tried to increase the U^{235} contents of U by the Clusius-Dickel method of thermal diffusion (Fleischmann, unpublished). These experiments were unsuccessful; obviously the thermal diffusion constant of UF_6 vapour is practically vanishing. But in another respect the Clusius-Dickel method proved to be very useful: by adding an inert gas to an aggressive gas (as UF_6) and keeping both separated in a Clusius-Dickel tube, one can vary and measure the gas pressure without any contact between the aggressive gas and metal parts of the apparatus taking place (Fleischmann, Naturwiss. 29,485, 1941). In this way the vapour pressure curve and the internal friction of UF_6 have been determined by Fleischmann; unfortunately the results are not available for the time being, Prof. Fleischmann having moved to Strassburg some time ago.

Chain reaction in U. Before the war already the idea of devising an apparatus in which a self maintaining reaction chain of U fission processes takes place has occupied theoretical and experimental physicist in all the world. Our Institute was interested in such a device as a powerful neutron source (it may be remembered that the director has played a part in the discovery of the neutron). Real large scale work, however, could not be done in our Institute for lack of material. Practically all the material available, as U metal and heavy water, were needed for the large scale experiments carried out at Berlin and later on at Hechingen-Haigerloch. At several occasions, however, the German scientists interested in the "U-Machine" gathered in Berlin in order to exchange views. It can be affirmed that the exclusive aim of all these common efforts was the construction of a device for steady and controlled production of neutrons.

and heat, whereas the question of an explosive bomb was not discussed. On the contrary it was eagerly searched for means to avoid with certainty processes of an explosive character.

In the last time experiments with a U-D₂O arrangement of 1,500 liters were made in the KWI for physics at Berlin and were meant to be continued at Hechingen-Haigerloch. The details of the last mentioned experiments were fixed between Prof. Heisenberg and Prof. Bothe, and 1-3 members of our Institute were delegated to Prof. Heisenberg to participate in the experiments. The experimental method for measuring neutron densities was one devised in our Institute.

In our Institute only a few small scale experiments were made with a 80 liter arrangement of U metal plates in D₂O (Bothe and Fünfer, unpublished). These experiments aimed at determining the optimal thickness of U plates and D₂O layers, giving the maximum rate of neutron production. The result was 1 cm of U metal (molten) + 18 cm of D₂O. In these experiments a new quick method for determining the total number of thermal neutrons in a large cylindrical volume was used. The method consists in arranging calibrated glass tubes filled with a Dy-solution parallel to the cylinder axis; after a given time the activity of each solution is measured in a double walled G.M. glass counter. By multiplying by the distance of the tube from the axis and integrating the relative total number of thermal neutrons present is found. The disturbance caused by the neutron absorbing power of the tubes has to be corrected for (cf. Bothe, ZS.f.Phys. 12,437, 1943). This method was used in the large scale experiments also, as mentioned above.

Some theoretical considerations about the function of the "U machine" have been made. The chief result is that there is a definite lower limit for the size of the machine, if the neutron production shall maintain itself, without supplying neutrons from outside. The formula and its derivation are contained in the accompanying abstract of the unpublished paper.

VII. Radiochemistry

For many problems in biology, chemistry and physics it is very important to have preparations of radioactive isotopes in highest concentrations. In certain cases the Szilard-Chalmers method has proved very useful.

Dr. Starke is engaged in developing a more general method based on adding an isomorphic compound, purifying and separating by sublimation. The method has already been successful in a number of cases, but the experiments have to be continued.

VIII. Electron physics

Single scattering of electrons with energies up to 2,5 MeV in Al and Ni has been investigated, using a magnetic lense as a "monochromator" and a point counter as a detector (Bothe and Ratzel, ZS.f.Phys. 115,497,1940). The results are supposed to be much more reliable than previous results obtained chiefly by the cloud chamber method. For scattering angles of $12 - 25^\circ$ the results are in agreement with the Mott formula at low energies. For higher energies the scattering cross section becomes larger than is predicted by the theory, f.i. twice for scattering of 2,4 MeV electrons in Al.

A betatron for production of beta- and gamma-rays of about 10 MeV is nearly finished. The construction follows essentially that of Kerst. Some parts have been transferred to our branch laboratory at Tauberbischofsheim, where the whole plant was intended to be installed for reasons of air protection. The betatron was planned chiefly for medical use.

Heidelberg, June 26th 1945

Professor Dr. W. Bothe,
Director.

Künstliche Radioaktivität durch thermische Neutronen bei den seltenen Erden

Von

W. Bothe

**Aus den Nachrichten der Akademie der Wissenschaften in Göttingen
Mathematisch-Physikalische Klasse 1944.**

Künstliche Radioaktivität durch thermische Neutronen bei den seltenen Erden

Von

W. Bothe

Vorgelegt in der Sitzung am 20. Oktober 1944 von R. POHL

Im Gebiet der seltenen Erden waren bisher die kernphysikalischen Kenntnisse lückenhafter und unsicherer als im übrigen periodischen System¹⁾. Der Grund war weniger die „Seltenheit“ dieser Elemente als die Schwierigkeit, sie genügend rein zu erhalten. Meist stehen nur mehr oder weniger hoch angereicherte Gemische seltener Erden zur Verfügung, deshalb ist es nötig, alle seltenen Erden im Zusammenhang zu untersuchen. Einen ersten Überblick über die radioaktiven Isotope, die durch Bestrahlung der seltenen Erden mit thermischen Neutronen, also durch (n, γ) -Prozesse entstehen, haben vor einer Reihe von Jahren HEVESY und LEVI²⁾ gegeben. In der vorliegenden Untersuchung wurde mit sehr viel stärkeren Neutronenquellen gearbeitet (Zyklotron), und die Messungen wurden mit tunlichster Genauigkeit über möglichst lange Zeiten ausgedehnt, um vor allem die längerlebigen Produkte zu erfassen und eine saubere Komponentenzerlegung der Abfallkurven zu ermöglichen. Weiter wurden auch die Absorptionskurven der Strahlungen stets mitgemessen, meist mehrmals in verschiedenen Stadien des Abfalls, so daß sie ebenfalls in ihre Komponenten aufgelöst werden konnten. Erst durch Hinzunahme der Absorptionskurven wird es möglich, eine beobachtete Aktivität mit Sicherheit zu identifizieren und den Einfluß von Verunreinigungen abzutrennen.

Die Ergebnisse sind in der Tabelle zusammengestellt. Zum Teil wurden ältere Ergebnisse bestätigt bzw. Fehlergebnisse richtiggestellt, doch wurde auch eine Reihe neuer Aktivitäten aufgefunden. Die eindeutige Zuordnung zu bestimmten Massenzahlen (Spalte 1)

1) Vgl. J. MATTAUCH u. S. FLÜGGE, Kernphysikal. Tabellen, Berlin 1942.

2) G. HEVESY u. H. LEVI, Danske Videnskab. Selskab. 14, Nr. 5, 1936.

ist in einigen Fällen noch nicht möglich. Die von anderer Seite manchmal vorgenommenen Zuordnungen auf Grund vermeintlicher ($n, 2n$)-Prozesse mit schnellen Neutronen sind nicht stichhaltig. Die Energiegrenzen der β -Spektren (Spalte 4) und die γ -Energien (Spalte 5) sind ziemlich rohe Schätzungen aus den Reichweiten bzw. Absorptionskoeffizienten. Die mehrfach auftretenden K-Strahlungen (Spalte 6) wurden, wo es die Intensität erlaubte, durch Absorptionsmessungen in selektiven Filtern identifiziert. In der 7. Spalte sind die wirksamen Kernquerschnitte für die Aktivierung aufgeführt, die auf folgende Weise gewonnen wurden. Aus den Aktivitäten pro Mol, die man durch eine bestimmte kurzzeitige Neutronendosis erhält, ergeben sich nach Korrektur für alle Absorptionseinflüsse, Berücksichtigung der Zählrohrempfindlichkeit und Multiplikation mit der jeweiligen Halbwertszeit Relativwerte für diesen Aktivierungsquerschnitt. Die Umrechnung auf Absolutwerte geschieht durch einen Normierungsfaktor, der daraus erhalten wird, daß für die Reinelemente (Ho, Tm) der *Aktivierungsquerschnitt* mit dem anderweitig gemessenen *Absorptionsquerschnitt* für thermische Neutronen (Spalte 8) übereinstimmen muß. Die Zahlen der Spalte 7 können nicht sehr genau sein; in einigen Fällen können nur untere Grenzen angegeben werden, weil weichere β -Strahlkomponenten vermutlich der Messung entgingen. Die Absorptionsquerschnitte in Spalte 8 sind den Messungen von RIEZLER und von REDDEMANN u. BOMKE entnommen¹⁾. Überall wo sie wesentlich größer als die Aktivierungsquerschnitte sind, läßt sich dies aus der Entstehung inaktiver Isotope erklären.

Bei den einzelnen Elementen ist folgendes bemerkenswert. Die drei neuen *Cer*-Aktivitäten werden trotz des kleinen Aktivierungsquerschnittes fast ausschließlich durch thermische Neutronen erregt, wie durch Cadmium-Filterung festgestellt wurde. Es entsteht der Verdacht, daß es noch ein seltenes stabiles Ce-Isotop gibt, das bisher der Beobachtung entging, etwa Ce^{144} . Das *Samarium* (47 h) geht zunächst in ein *metastabiles* Eu über, dessen γ -Strahlung vollständige innere Umwandlung erleidet. Beide aktive *Europium*-Isotope zeigen dualen β - und K-Zerfall. Die vorher unbekannte Halbwertszeit des langlebigen Eu-Isotops wurde hier auf indirektem Wege auf 7 Jahre geschätzt. Dieser Wert ergibt sich aus den gemessenen Intensitäten und aus der Forderung,

1) Vgl. den Bericht von K. DIEBNER u. a., Phys. ZS, 43, 440, 1942.

daß die Summe aller Aktivierungsquerschnitte des Eu mit seinem Absorptionsquerschnitt übereinstimmen muß. Für *Gadolinium* konnten trotz verhältnismäßig reiner Präparate keine klaren Ergebnisse erhalten werden, weil die Aktivität sehr schwach ist. Auch die *Erbium*-Aktivitäten sind schwach und schwer genau zu untersuchen. Die 6—7 h-Aktivität scheint aus zwei Komponenten verschiedener β -Energie zu bestehen. *Ytterbium*¹⁶⁹ ist ein neuer K-Strahler, der aus dem seltenen Isotop Yb¹⁶⁸ mit dem sehr großen Isotopenquerschnitt von etwa $20\,000 \cdot 10^{-24} \text{ cm}^2$ entsteht. Aus *Cassiopeium* entsteht ein Isomer des natürlich radioaktiven Cp¹⁷⁶, wie schon MATTAUCH vermutet hatte¹⁾; dieses zerfällt dual. Bei Yb und Cp wurden die Ergebnisse noch besonders durch Koinzidenzversuche von H. MAIER-LEIBNITZ²⁾ sichergestellt.

Die Methode der kombinierten Abfalls- und Absorptionsanalyse, wie sie hier zur Auffindung und Untersuchung der Aktivitäten benutzt wurde, kann nun auch umgekehrt zur Stoffanalyse auf kernphysikalischem Wege mit Vorteil angewandt werden. Im Laufe dieser Untersuchung konnten z. B. gewisse Verunreinigungen von der Größenordnung 0,01% leicht quantitativ bestimmt werden. Eine solche zerstörungsfreie Analysenmethode dürfte gerade im Gebiet der seltenen Erden wertvoll sein. Diese Fragen sollen an anderer Stelle ausführlicher behandelt werden.

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1) J. MATTAUCH, ZS. f. Phys. 117, 246, 1941; A. FLAMMERSFELD u. J. MATTAUCH, Naturwiss. 31, 66, 1943.

2) Wird demnächst veröffentlicht.

Aktives Isotop	Halbwertszeit	Zerfall	β -Strahlen eMV	γ -Strahlen eMV	α -Strahlen	Aktivier.-Querschn. 10^{-24} cm ²	Absorpt.-Querschn. 10^{-24} cm ²
1	2	3	4	5	6	7	8 ^{*)}
Y ⁹⁰	61 ± 1 h	β_-	2,1—2,6 ¹⁾	—	—	1	~ 4
La ¹⁴⁰	89,5 ± 0,5 h	β_-	1,3	0,9 ²⁾	—	> 7	20
Ce ^{141, 142, ?}	33 ± 2 h	β_-	1,8	?	—	0,12	} > 0,6
	12,5 ± 1 d	β_-	0,9	?	—	0,11	
	30 ± 2 d	β_-	0,4	+	—	> 0,4	
Pr ¹⁴²	19,2 ± 0,3 h	β_-	2	schwach?		13	< 12,5
Nd ^{147, 148, 151} od. 61 ^{147, 148, 151} (od. *Sm ¹⁵¹)	130 ± 3 m	β_-	1,6	?	—	0,16	} 0,6
	47,5 ± 1,5 h	β_-	1,1	?	—	0,21	
	11,1 ± 0,2 d	β_-	0,8	?	—	0,24	
Sm ^{151, 152, 155} (od. *Eu ¹⁵⁵)	~ 20 m	β_-	—	?	?	—	} 6500
	47 ± 1 h	β_-	0,75	—	Eu (inn. Umw.)	42	
Eu ^{153, 154}	} 9,3 ± 0,2 h	β_-	1,8	+	—	470	} 3300
		α	—	?	Sm	280	
		β_-	1,0	?	—	260	
} 7 ± 4 a	β_-	—	—	+	Sm	2300	} 3300
	α	—	—	+	Sm	2300	
Tb ¹⁶⁰	73,5 ± 1 d	β_-	0,7	+	?	> 22	—
Dy ¹⁶⁵	140 ± 1,5 m	β_-	1,4—1,9 ³⁾	—	—	600	820
Ho ¹⁶⁶	27,3 ± 0,5 h	β_-	1,8	—	—	49	49
Er ^{168, 171} (od. Tm ¹⁷¹)	~ 6 m	β_-	—	?	?	—	} > 1,4
	6—7 h	β_-	1,6	+	—	> 1	
	20 ± 2 h	β_-	0,6	+	—	> 0,4	
Tm ¹⁷⁰	127 ± 5 d	β_-	1	—	—	95	92
Yb ¹⁶⁹ Yb ^{175, 177}	33 ± 1,5 d	α	—	0,2; 0,4	Tm	11	} 34
	2,4 ± 0,2 h	β_-	1,3	?	?	1	
	99 ± 3 h	β_-	0,18; 0,5	0,35	Cp (inn. Umw.)	22	
*Cp ¹⁷⁶	} 163 ± 4 h	β_-	0,5	—	—	66	} 105
α		—	0,2	Yb	13		
β_-		1,15 ⁴⁾	—	—	26		
Cp ¹⁷⁷	220 ± 10 m	β_-	—	—	—	—	132

1) Vgl. J. MATTAUCH u. S. FLÜGGE, Kernphysik. Tabellen, Berlin 1942.

2) Nach W. MAURER (im Druck).

3) A. FLANNERSFELD und J. MATTAUCH, Naturwiss. 31, 66, 1943.

4) Nach d. Bericht v. K. DIEMER u. a., Phys. ZS. 43, 440, 1942.

Die Mindestgröße der U-Maschine (Auszug).

Von W. Bothe.

Eine kugelförmige Maschine, die sich selbst unterhalten soll, muß mindestens den Radius R haben, bei dem die stationäre Dichte der thermischen Neutronen am Rande Null und in der Mitte regular ist. Wir stellen die Differentialgleichung für φ auf, wobei wir der Einfachheit halber das System als eine homogene Mischung von U und Bremsmittel betrachten. D sei der Diffusionskoeffizient der thermischen Neutronen, k die Zahl der schnellen Spaltungsneutronen, die durchschnittlich für jedes im System absorbierte thermische Neutron entstehen, w die Wahrscheinlichkeit, daß ein Neutron im U-Resonanzgebiet wird, bevor es thermisch wird. Weiter sei s der Abstand zwischen dem Ort, wo ein Spaltungsneutron entsteht, und dem Ort, wo es thermisch wirkt; $s^2 = \xi + \eta^2 + \dots$. Schließlich sei $p(s)ds$ die Verteilungsfunktion der s ; $\int_0^\infty p(s)ds = 1$. Die Diffusionsgleichung lautet dann

$$D \Delta \varphi - v \varphi + \int_0^\infty v \varphi(x+\xi, y+\eta, z+\zeta) X(1-w) \frac{p(s)}{4\pi s^2} d\xi d\eta d\zeta = 0$$

Entwickelt man unter dem Integral bis zu Gliedern zweiter Ordnung in ξ, η, ζ , so fallen bei der Integration alle Glieder fort, die linear in einer der Größen ξ, η, ζ sind. Daher wird in dieser Näherung

$$D \Delta \varphi - v \varphi + v X(1-w) \left(\varphi + \frac{1}{2} \Delta \varphi \int_0^\infty \xi^2 p(s) ds \right) = 0$$

oder mit

$$\int_0^\infty \xi^2 p(s) ds = \frac{1}{3} \int_0^\infty s^2 p(s) ds = \frac{1}{3} \bar{s}^2$$

$$\left\{ D + \frac{1}{6} v X(1-w) \bar{s}^2 \right\} \Delta \varphi + \{ X(1-w) - 1 \} v \varphi = 0$$

Die Größe

$$D' = D + \frac{1}{6} v X(1-w) \bar{s}^2$$

kann als der "effektive Diffusionskoeffizient" bezeichnet werden. Die Lösung der Differentialgleichung, die bei $r = 0$ regular ist und bei $r = R$ verschwindet, lautet:

$$\varphi = \frac{1}{2} \sin \pi \frac{r}{R}$$

Dies eingesetzt gibt für R den Wert

$$R = \pi \sqrt{\frac{D'}{\{X(1-w) - 1\} v}}$$

Hierfür kann man auch schreiben, wenn $l = \sqrt{D/v}$ die Fermische Diffusionslänge der thermischen Neutronen bedeutet:

$$R = \pi l \sqrt{\frac{D' + \frac{1}{6} v X(1-w) \bar{s}^2}{X(1-w) - 1}}$$

Dies ist die Mindestgröße der freistehenden Maschine. Ist die Maschine von einem Mantel umgeben, der Neutronen zurückstreut, so kann sie erheblich kleiner sein, da γ am Rande nicht zu verschwinden braucht.