

ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ

Дубна.



E15 - 4651

G.N.Flerov

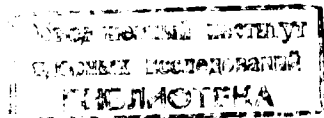
NEWEST ELEMENTS
OF THE MENDELEEV PERIODIC TABLE.
PERSPECTIVES AND CHEMICAL PROBLEMS
OF THE SEARCH FOR SUPER-HEAVY
ELEMENTS

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Report presented at the Paris Conference dedicated
to the centenary of the Mendeleev Periodic
Table



7988/2 p2,

It is now 70 years since D.I. Mendeleev wrote in "Foundations of Chemistry", when recognizing the significance and extreme importance of reasons for restricting a number of elements: "Being certain that the investigation of uranium starting with its natural sources will lead to still more new discoveries I safely advise to those who look for a subject of new research to pay a particular attention to uranium compounds".^{/1/}

It should be remembered that at a moment when the Periodic Table - one of the most fundamental laws of natural science was discovered, Marie Curie-Skłodowska was not yet one year old. Many years passed up to the discovery of natural and then artificial radioactivity. I believe that at that time the idea of element transmutation could even impede to some degree revealing the regularities generalized by D.I. Mendeleev in his greatest law. I should like to tell now about the situation in studying all the variety of atomic nuclei and to show the remarkable role the Periodic Law has been called to play in providing the progress in this region.

Nuclear properties are essentially effected by their charge. Here nuclear physics agrees with chemistry. An isotopic number -

atomic weight is also of a great value to nuclear physics (in contrast to chemistry). Nuclear properties change considerably with the atomic weight being changed from isotope to isotope. So the latter are usually systematized according to Z and A .

Fig. 1 shows the relationship of the already investigated part in nuclear studies to that to be investigated.

It looks like a map of isotopes, where Z - the proton number and N - the neutron number are plotted along the axes. Black squares present stable isotopes, their total number being about 300.

After the discovery of artificial radioactivity when the possibility appeared to create new unstable isotopes, the use was made of various types of accelerators and accelerated particles and more than 1500 isotopes were synthesized (dashed region in Fig. 1). Thus, the present amount of the known isotopes equals 2000, approx.

Analysis leads to a conclusion that the amount of isotopes of various elements with half-lives of thousandth fractions of a second and longer seems to be equal to about 5000 (region restricted by solid lines in Fig. 1).

A question naturally arises whether it is reasonable to continue studying isotopes already synthesized with a view of defining our knowledge of their properties or to synthesize new ones. Our opinion is that the information obtained in the investigation of an isotope being far off stability region is much greater as compared to that derived from studying isotopes near that region. It is a general methodological approach to study properties of a matter in extreme states which is used both by physicists and by chemists. Isotopes far off the β -stability region are

limited (1) when the proton number is small and the neutron number is relatively great and nuclear forces play the major part; (2) when there is excess of protons and Coulomb forces of repulsion are fairly important down to the possibility of radioactive decay of nuclei with the proton emission.

In this connection a heightened interest in study of nuclei of transuranium elements, where Coulomb forces are so great that they overcome nuclear tension forces, is quite understandable. Potential barrier that keeps a nucleus intact in equilibrium nearly disappears and the latter splits up into fragments. At the same time specifically nuclear effects associated with nuclear structure might express themselves very intensely. It is in this region of elements where a new kind of nuclear isomerism - isomerism of a shape has been discovered^{/2/}.

The maximum technical and scientific facilities at the disposal of scientists were always employed for the synthesis and study of new elements. By 1955 in the USA elements 93 to 101 have been created. To this powerful reactors, ground and underground nuclear explosions and accelerators were used.

Elements 99-100 were synthesized as a result of a thermo-nuclear explosion "Mike" conducted in 1952^{/3/}. An American theorist estimated the yield of the far elements produced via the reaction of instantaneous multineutron capture and predicted that elements 99 and 100 should be searched for in a cloud formed in an explosion. It took about a fortnight of strenuous work to catch isotopes of elements 99 and 100 out of this cloud and to estimate their properties.

Element 101-mendelevium was synthesized in the cyclotron by the bombardment of einsteinium (element 99) with alpha-particles.

It should be emphasized that after having synthesized an element in a nuclear reaction an investigator faces the extremely complex problem of extracting minor amounts of atoms of a new element from the target material where, as a rule, a mixture of tens of elements of Periodic Table was formed. Mass of the target material to be eliminated is hundreds of million times greater as compared to the amount of a new element. In addition, difficulties connected with the similarity of chemical properties of adjacent transuranium elements have to be overcome. The extremely complicated problem of chemical identification of new elements was solved successfully thanks to the analogous properties of actinide family and lanthanides. Thus the name mendelevium for element 101 in honor of Mendeleev was a natural recognition of the fact that without these fundamental indications of regularities of chemical properties of elements it would be more difficult for investigators to carry out successfully the titanic and at the same time minute work which has to be done in synthesizing and studying transuranium elements.

Further on the American scientists hoped to produce elements 104, 105, 106 and, possibly, even 108 by increasing density of neutron flux and duration of its influence. With this in mind a series of underground explosions has been carried out. Neutron flux in the centre of such an explosion amounts to 10^{25} neutrons passing through a centimeter squared. Directly after the explosion a borehole was started to take samples of fused rocks from the epicentre. These samples were rapidly taken to "hot" laboratories where an attempt was made to extract new elements chemically. It was a troublesome surprise for investigators that the only result of a special extremely powerful explosion "Cyclamen" was

an isotope of element 100 with mass number 257 - the heavier isotope of known element^{14/}. Any further elements have not by now been synthesized in nuclear explosions. This is likely to be caused by a small probability of capture of a greater amount of neutrons by nuclei of initial material or it is due to short life-time of heavy isotopes.

As far back as 1954 on Academician I.V. Kurchatov's initiative at the Atomic Energy Institute in Moscow the preparations to the synthesis of new elements started. Heavy ions accelerated in the cyclotron (carbon, nitrogen, neon, argon) were supposed to be taken for the synthesis instead of neutron fluxes of big density. Fusion of these nuclei with heavy nuclei of targets (uranium, plutonium) will result in production of elements 100 to 104 and heavier.

To this end, a heavy ion cyclotron has been designed and in 1961 put into operation in Dubna at the International Research Centre of socialist countries (Fig. 2). The magnet weight equals 2000 tonnes, a high frequency power - 1000 kw; power of magnet feeding is about 1000 kw, vacuum volume - 100 m³ (pressure 5.10⁻⁶ mm of Hg). From 1961 up to date this accelerator has been remained the most powerful machine around the world among the accelerators of heavy ions. Its beam intensity is tens and even hundreds times greater as compared to that available in the USA. By means of this machine we have managed to accelerate such particles as neon-22 being the major particle for bombarding targets of heavy elements.

While preparing the synthesis of elements 103 and 104 it was necessary to study in detail regularities of formation and properties of elements 102 known by that time. For this purpose 6

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its isotopes were obtained^{/5/}. Earlier American and Sweden analogous works on the synthesis of element 102 turned out to be erratic^{/6,7/}. Further on, when synthesizing element 104 we saw that its yield as well as its life-time compared to element 102 decline steeply. Even in optimum irradiation regime the yield observed accounted for 1 atom for 5 hours of operation^{/8/}.

The isotope of element 104 synthesized lives 0.3 sec. Despite so short life-time, I. Zvara and a chemists' group managed to extract element 104 out of a complicated mixture of radioactive products of nuclear reactions due to the gaseous method developed by this group^{/9/}. The fundamentals of this method are the follows. Atoms of element 104 via reaction in the gaseous phase are chlorinated, then with a view of separating from other atoms they are transported by gas stream through the adsorbent system and finally enter a chamber with mica detectors which register their decay. All the process has to be carried out for tenths of a second. Chemical investigations revealed that a new isotope belongs to an element whose volatile chloride is similar to that of hafnium and differs strongly from chlorides of kurium and californium, that is from chlorides of actinide elements. Thus, element 104 proved to be a chemical analogue of hafnium.

After that experiments on the synthesis of element 105 started. In order to provide a more single-minded search, it was decided to study first of all properties of element 103 isotopes and then, after the data on isotope properties of elements 102, 103 and 104 having been accumulated, to perform extrapolations with more assurance. We succeeded in creating two isotopes of element 103 with mass numbers 256 and 257^{/10/}. And in this case their real properties proved to be inconsistent with those ascribed to them by our colleagues in America^{/11/}.

At the present time at Dubna preliminary data on element 105 have been obtained by bombarding Am target with Ne ions. Further investigations are being carried out aimed at their improvement. Its life-time equals hundredth fractions of a second, and probability of its formation in the reaction is so that we managed to detect one atom per one day.

It is the main difficulty of experiments on the synthesis of new chemical elements that shows up most vividly here still lower effective cross sections of element production and still shorter half-lives with increasing atomic number. It is clear that element 106 and heavier cannot be created by similar method. Further advancement in the region of transuranium elements by any known method seemed to be impossible.

However, recently new data appeared which allows to consider perspectives in the synthesis of the far transuranium elements more optimistically.

The point is that in spite of the fact that a decline in stability with increasing charge and mass of a nucleus is a general trend in nuclide properties, the latter do not change monotonically. The situation reminds, in a definite sense, of the Mendeleev Periodic Table.

The periodicity in chemical properties of elements reflects the extent to which electron shells of atoms are populated. Nuclei possess shells too (closed and unclosed shells), however, in this case they are composed of two kinds of particles - of protons and neutrons. One more complication is presented by the fact that sizes of nucleon "orbits" in a nucleus are commensurable with sizes of particles themselves and they move in a force field created by themselves. Since the question of nature

of nuclear forces has been so far left open, calculations of nuclear shells are not accurate enough. However, all the known experimental data on nuclear properties indicate that these properties: a nuclear shape, its magnetic and mechanical momenta, spectra of excited states and the most important property - stability against various kinds of decay reveal strong changes near the certain proton and neutron numbers. These numbers are called "magic" and correspond to closed shells in nucleus. Nuclei containing a closed outer shell are characterized by particular stability, longer half-life as compared to adjacent isotopes or elements.

This enables one to hope that islands of relatively stable nuclei involving magic nucleon numbers may appear against a general background of decreasing stability as the far transuranium region is approached. Close to uranium magic numbers 82 (by protons) and 126 (by neutrons) are known. For further extrapolations experimental data on the possibly most remote transuranium elements is needed.

The analysis of properties of six isotopes of element 102 synthesized and studied so far provided evidence that the most stable isotope has mass number 254 which corresponds to neutron number $N = 152$ (Fig. 3). In this case 152 neutrons form a so-called subshell that also imparts enhanced stability to a nucleus. The existence of a subshell brings about the increase in yield of isotope 102^{251} and causes its longer half-life as compared to lighter and heavier neighbours.

Later on, theorists of several countries (USA, Sweden, Poland, England and USSR) have analysed the possibilities of existence in superheavy nuclei of real shells and their effect

on the nuclear stability. They concluded that if element 105 has as short half-life as 0.01 sec, approx., then from the point of view of the liquid drop model, elements 110 to 114, where Coulomb forces destroying a nucleus are very intense, should live only 10^{-20} sec. In the case of isotopes $^{294}_{110}$ or $^{298}_{114}$ however, with the neutron number 184 these should live considerably longer (value of $N = 184$ according to calculations, has to correspond to a closed neutron shell). Fig. 4 presents results of rather complicated calculations performed by theorists with computers for various isotopes of element 114. As is seen, isotope with mass 298 corresponding to $N = 184$, has to possess a half-life of 10^{20} years against spontaneous fission. This isotope is stable against β -decay and should have a fairly long life-time against α -decay, at least of the order of years. One can see the perspectives proceeding from these conclusions.

The group headed by Prof. Thompson in the USA made an attempt to synthesize element 114 during a year by irradiating a target of $^{248}_{96}\text{Cm}$ with $^{40}_{18}\text{Ar}$ ions. At first the results seemed to be positive, however the check experiments indicated that the back-ground of other reaction products had been registered^{/12/}. The experiments stopped.

It should be borne in mind that these experiments could produce nuclei of element 114 with neutron number 170, i.e. by 14 units less than in the closed shell. Using element 102 as an example, it was shown experimentally that life-time of isotopes and probability of their production via reaction decline sharply when moving off the shell. From this point of view, the result of Prof. Thompson is not surprising. Therefore, one had to take different way trying to approach the closed shell as near as

possible. For this purpose the heaviest target available has to be irradiated with the heaviest neutron-excessive nuclei. The possibility exists to apply plutonium-244 combined with calcium-48. Though deliberately short-lived atoms should be obtained, it is possible, however, that their life-time will be longer than one millionth fraction of a second. It will be sufficient for detecting new element formation and for measuring its half-life that, in its turn, will provide new evidences for further theoretical extrapolations.

Much the same situation is expected in the region of $Z = 126$, where the existence of relatively stable nuclei should be assumed.

It is possible that the above experimental difficulties will be overcome if instead of fusion reactions use will be made of fission reactions. It is to be explained here, why, say, in bombarding uranium by neon a very small amount of atoms of element 102 is produced. The point is that compound nuclei formed in fusion are excited state and in the process of deexcitation the fission occurs with the probability by a factor of tens of millions greater than the probability of processes resulting in element 102 production. Thus, the fission hinders synthesis and the idea concerning its application to solve this problem seems a bit unexpected.

The fission process yields products of a wide variety of charges and masses. If uranium is irradiated by neon, fission fragments are obtained in the region of $Z = 30-70$. In the case of $U + U$ reaction among fission fragments there would be found transuranium elements including the remote ones enriched with neutrons sufficiently. We do not yet possess beam of uranium

accelerated ions, but even by bombardment of uranium with argon polonium as a fission fragment could be obtained. The perspectives of this line are the further increase in sizes of accelerators widening the scope of operation, acceleration of xenon and next - of uranium, synthesis of the heaviest isotopes and study of their properties.

In this connection the following programme of the synthesis of transuranium elements may be proposed for the near future: (1) synthesis of light isotopes of elements 114 and 126 by all available means; (2) production of heavy isotopes by using accelerated ions heavier than xenon. Suddenly these calm forecasts were corrected. In summer 1968 an English physicist Prof. Powell at the general session of the Academy of Sciences of the USSR reported the experiments with nuclear emulsions which 25 years ago had resulted in the discovery of π -mesons in cosmic rays and provided a lot of other important information. At the end of his report he showed photographs obtained by his scholar Prof. Fowler who recently had launched balloons carrying photographic emulsions to a height up to 40 km to study element composition of cosmic radiation.

In cosmic rays there were detected a large amount of iron nuclei ($Z = 26$) and considerably smaller amount of nuclei in the region of $Z = 60-90$. The heavier is a particle, the thicker is its track in the photoemulsion. In developing large areas of photoplates Fowler found single very thick tracks which were ascribed to uranium.

In the second series of experiments a very long track of a particle was found that has ionizing effect much stronger than nuclei with $Z = 90$ (Fig. 5). Initially, Prof. Powell attributed charge 110 to this particle. Somewhat later, however, having taken into

account all possible experimental errors he reduced this value up to $106^{13/}$. Next experiments of Fowler carried out in collaboration with three American groups confirmed the existence of super-heavy component in cosmic rays. 15 tracks with $Z > 80$; 3 tracks with $Z > 100$; 1 track with $Z \approx 108$ were observed^{14/}.

Determination of particle charge by its track in a photoplate is a very complicated problem and it is difficult to discriminate between values of 90, 100 and 110. Processes occurring in photoemulsion in slowing-down of such heavy particles have not yet been studied well enough. For instance, the sensitivity of photoemulsion is known to depend on temperature. Whereas plates themselves were cold, the emulsion was heated along the path of a particle and its effect could be estimated only approximately.

At the moment as big amount of heavy particles as possible has to be registered and a function of particle yield on a charge has to be plotted. Elements with Z from 84 (polonium) up to 89 (actinium) should not be met in cosmic rays since they have no sufficiently long-lived isotopes. Therefore, the growth of the number of events registered allows to point where the region of

$Z = 90$ starts and to determine a charge of heavier particles with more confidence.

If the data reported by Prof. Fowler are true, it provides for very essential conclusions. One nucleus with $Z = 92$ falls onto meter squared of the Earth surface per 24 hours, i.e. a bit less than the amount of uranium nuclei. This means that the life-time of these nuclei is not less than tens of million years since the specific content of a component in cosmic rays is proportional to its life-time. So, it is sufficient to observe a few nuclei with good confidence for a conclusion to be made that nuclei with life-time of the order of tens and even hundreds of million

years exist in the region of $Z = 110-114, 126$. Close to these a group of tens of isotopes has to exist with a bit shorter life-time. If it is so, a new region involving both sufficiently long-lived and short-lived elements will be added to the latest element 105 of the Mendeleev Periodic Table.

It is obvious, thus, that theoretical extrapolations and experimental evidences do not limit practically the range of values of half-lives for elements near 114 and 126. These may be millions of years and millionth fractions of a second. So the investigator who hopes to prove the existence of these elements has to deal with a rather various experimental technique. Fig. 6 shows schematic view of the region of known elements from thorium up to kurchatovium and also presents conditionally hypothetic region of elements near $Z = 114$. Along horizontal axes proton number (Z) and neutron number (N) are plotted. For every isotope a height of column shows a half-life (vertical axis). Three planes cutting the time axis correspond to various values of half-life: 10^{-9} years (0,03 sec), 10^3 years and 10^8 years.

As far as the information on short-lived nuclei is concerned, it can be obtained, apparently, only by synthesizing them artificially, the only possibility to reach the appeared stability region being nuclear reactions induced by heavy ions. An attempt to create element 114 with the accelerator, experimental possibilities and perspectives in this direction were described above.

Much longer life-times are accessible in investigations of cosmic rays, which age has an upper limit of the order of $10^6 - 10^8$ years. Along with direct studies of the cosmic ray composition, one can try to detect superheavy transuranium elements on the Earth surface where they can fall constantly from the cosmic

space. As it was mentioned above, one atom with $Z > 92$ falls onto 1 m^2 of the Earth surface per 24 hours, i.e. about 70 mkg per year to all the Earth surface. It is a terribly small amount to be detected, however, data which can be gained therewith is so important that we have to make an attempt.

First of all chemical properties of elements 110-114 should be predicted. Then these elements have to be extracted from the deposits where they could be kept as a result of geochemical processes. Chemical studies of kurchatovium indicated that starting from $Z = 104$ an outer electron shell is populated that is kurchatovium turned out to be a chemical analogue of hafnium and not an actinide. Hence, elements 110-114 may be supposed to be a chemical analogues of Pt - Pb .

Cosmic new-comers are slowed-down at a big height in upper layers of stratosphere , mainly close to magnetic poles. Further on, these are spread over by atmospheric fluxes and fall onto the Earth surface with precipitation. Thus, one had to try extracting eka-lead and, possibly, other elements from water of lakes and oceans and to study their spontaneous fission and α -decay. Eka-lead and other new elements (eka-bismuth etc) fall from sea water by coprecipitating with hydroxides into sea deposits, where the content of these elements is likely to be higher as compared to natural lead. From this point of view, processing of iron-manganese nodules and other materials is of interest. It is also interesting to study meteorites and the moon surface which is likely to become accessible to man in the near future.

Finally, if elements we are searching for possess lifetimes comparable to the Earth age, then it is naturally to try to find their long-lived isotopes in minerals and to analyse possibilities of their extraction with a view of studying their physical and

chemical properties, determining mass and modes of radioactive decay.

If eka-lead (i.e. element 114) really coprecipitates with lead via geochemical processes, then it should be searched for in lead and lead minerals.

Experiments on the search for spontaneous fission in lead were performed at Dubna. 2 m² of thinnest mylar film have been contacted with a lead foil for a long time. As it was mentioned, eka-lead has to be an α -active and spontaneously fissioning element. If lead contains nuclei undergoing spontaneous fission, the film will detect fission fragments. The lead itself did not fission practically: its half-life should be equal to 10^{40} years, approx. (to illustrate this value: in 1 km³ of lead two fission events occur for one thousand years). "Sandwich" of mylar film and lead foil was put underground at a depth of 40 m to be protected from cosmic background. After 100 days 5 tracks of fission fragments were detected in the film. Calculations showed that if these fragments belong to lead its half-life will be not 10^{40} years, as should be expected, but 10^{21} years. The appeared reduction of lead half-life may be caused by an extremely small impurity of eka-lead which, however, decays many times faster than the common lead.

Long duration and laborious character of experiments with lead foils forced us to turn to samples where nuclei contained in lead have been registered for a long time. These samples are presented by glass contacted to metal lead for many years and glasses containing in their material lead compounds as well as lead crystalline minerals. Fission fragments leave in glass, some crystals and in mylar latent tracks which could be seen in the ordinary microscope after the proper etching.

In processing 10 cm² of glass surface of stained-glass panel of the XV century contacted to lead and 80 cm² of glass surface of Leiden cup with lead facing made at the end of the XIX century no one track of fragment was found. This result is in accord with the appeared low limit of half-life of spontaneous fission of lead $T_{1/2} = 2 \times 10^{22}$ years for the glass panel of the XIV century and $T_{1/2} = 10^{22}$ years for the Leiden cup. (For comparison: effect of thorium nuclei fission by cosmic rays at sea-level leads to the appeared low limit of spontaneous fission half-life of 1.5×10^{19} years ^(15/)). The absence of fission fragments in glass being in contact to lead seems to prove that the effect of nuclear fission induced by cosmic particles is very small.

One more series of experiments was performed with glass samples containing lead. To detect fission fragment tracks in the glass volume a consequent etching was employed. In each act of etching glass layer up to 20 μk was removed. Special check experiments revealed that fission fragment tracks differ strongly from accidental defects of glass.

In the experiment on lead manufactured in 1958 (lead concentration 40%) in the glass volume of 0,7 cm³ there were detected 27 fission tracks. The effect observed corresponds to the appeared spontaneous fission half-life of lead contained in glass:

$$T_{1/2} = (2 \pm 0.7) \times 10^{20} \text{ years.}$$

In the experiment with a cut-glass vase made at the end of the XVIII century (lead concentration 7%) in the volume of 0,27 cm³ there were detected 31 tracks of fragments which correspond to the appeared spontaneous fission half-life $T_{1/2} = (3 \pm 1) \cdot 10^{20}$ years.

It should be noted that the contribution of fission of thorium contained in these glasses by cosmic particles is negligibly small. Its concentration of thorium at the level of 1% by its weight which is dangerous. Check experiments showed that the content of thorium in these glasses does not exceed 10^{-5} g/g.

Background due to spontaneous fission of uranium contained in these glass samples (less than 10^{-7} g/g) can not exceed 3% of the effect being observed.

Thus, in the experiments with lead glass and lead contacted with mylar the effect of nuclear fission was detected which cannot be attributed neither to spontaneous fission of thorium or uranium contained in these samples nor to their fission induced by cosmic particles.

The hypothesis of existence of a heavy chemical element—an analogue of lead or of an element accompanying it with a half-life against spontaneous fission of over 10^8 years contained in samples in minor amounts, 10^{-12} - 10^{-13} g/g, may serve as a possible explanation of the effect being observed.

Difference in effects observed on lead glass samples and those contacted with metal lead may be caused by differences in chemical technology when preparing metal lead and its compounds introduced into glass composition.

More definite answer may be obtained by using possible difference in chemical properties of lead and eka-lead in order to develop chemical technique of its separation. Enrichment of 100 times is enough to check reliably the preliminary data of the investigation. Chemical properties of eka-lead are likely to be different from those of lead. For instance, eka-lead does not seem to have a stable tetravalent state. This had to be somewhat amphi-

teric. Volatility of its compounds and chloride dissolubility should be higher than those of lead. Moreover, a bigger ion radius allows to apply ion-exchange chromatography for its extraction. Further on, in addition to processing and treatment of new samples of lead glass the search for spontaneous fission effect is advisable in lead minerals which have been protected from cosmic irradiation by thickness of rocks and which have geological age of the order of tens of million years.

Side by side with this line at Dubna there will be carried out works on the synthesis of elements 114 and 126 with the heavy ion accelerators using $^{48}_{20}\text{Ca}$ and $^{68}_{30}\text{Zn}$ ions. If these experiments are successful, we could confirm yet preliminary conclusions concerning the existence of relatively stable super-heavy elements. This, in its turn, will make one to take a new look at many problems of nuclear structure and nuclear forces which act between nucleons.

To conclude, it can be said paraphrasing Mendeleev's words, that the investigation of lead and adjacent elements starting from its natural sources will lead to many discoveries and I can recommend with confidence to pay a special attention to studying lead compounds to all those who looks for a subject of new investigation.

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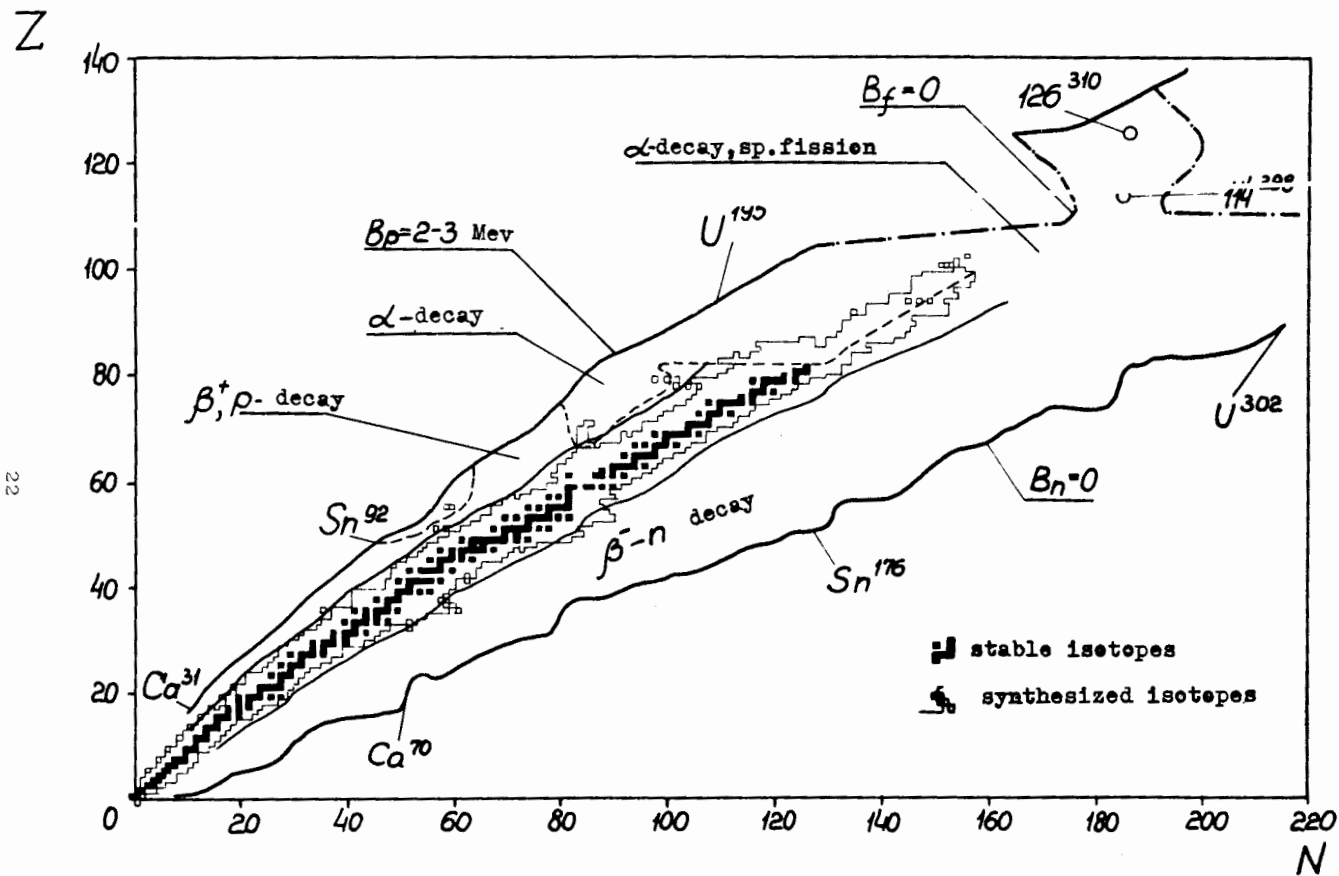


Fig. 1.

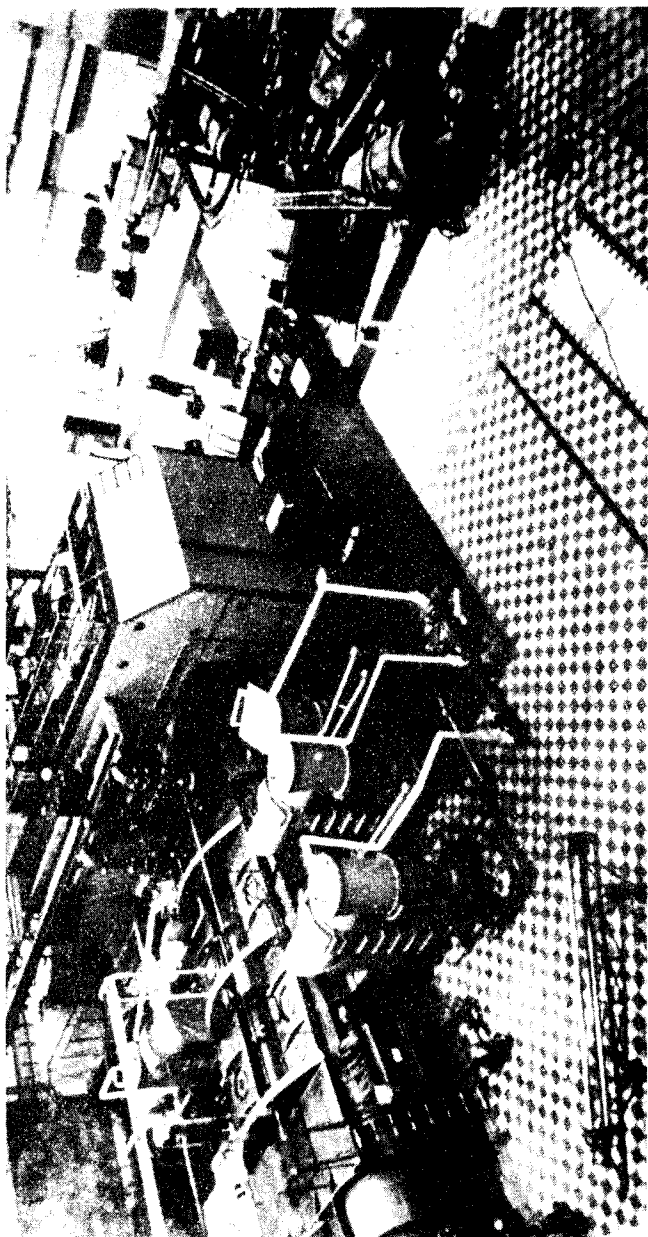


Fig. 2.

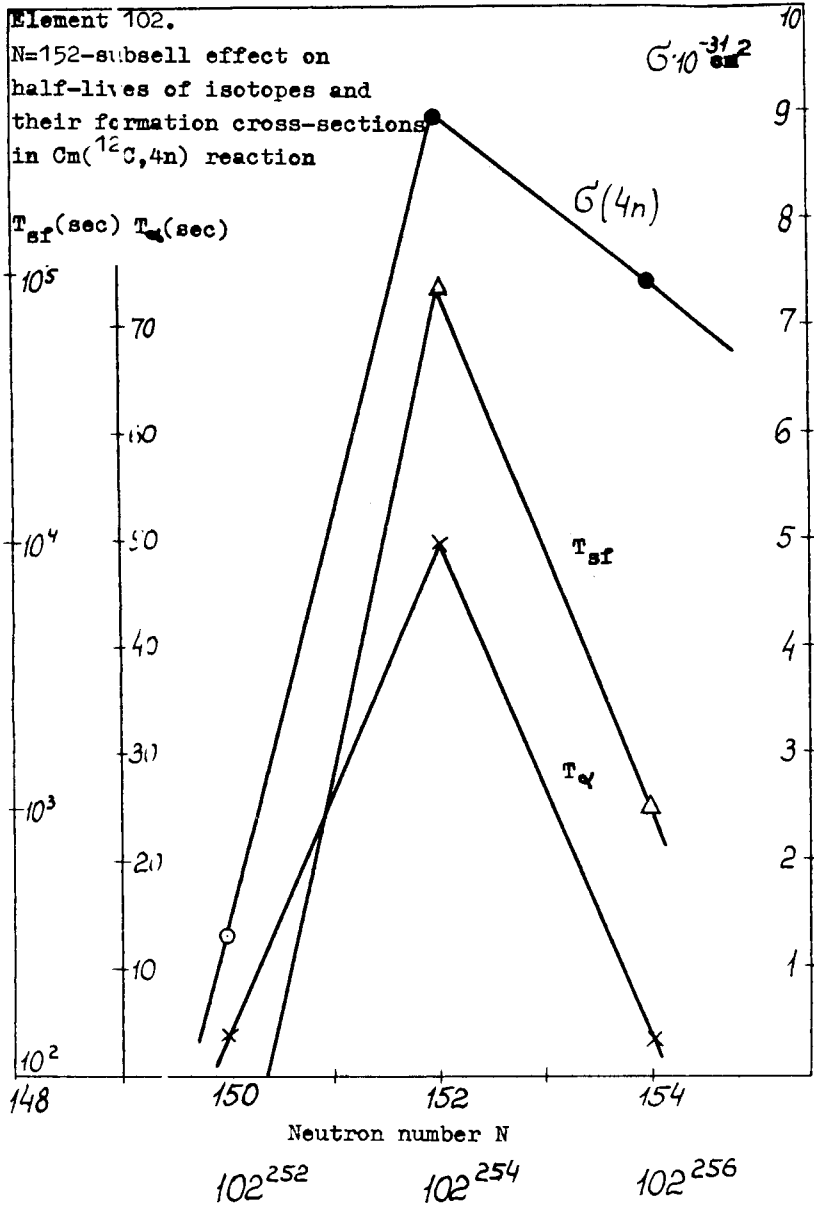


Fig. 3.

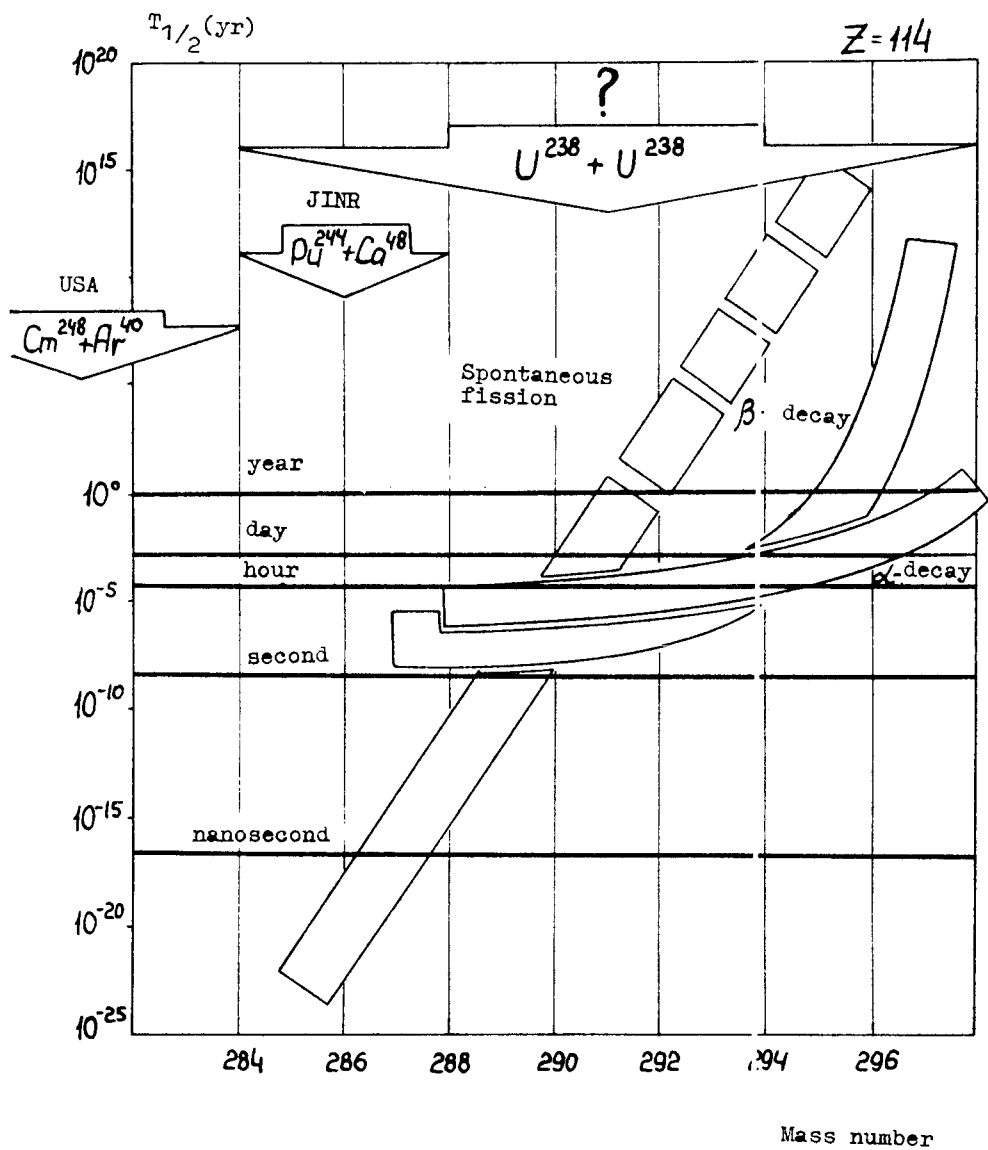


Fig. 4.

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Fig. 5.

Fig. 6.

