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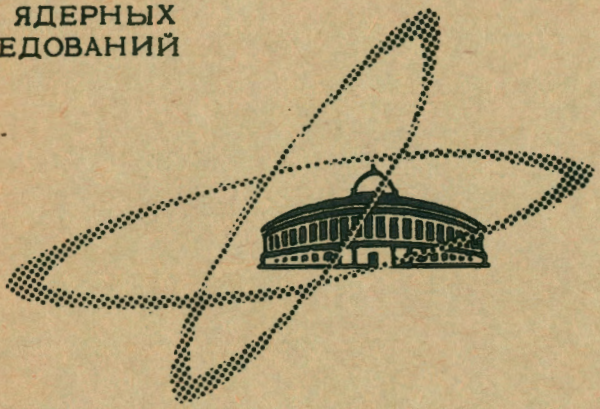

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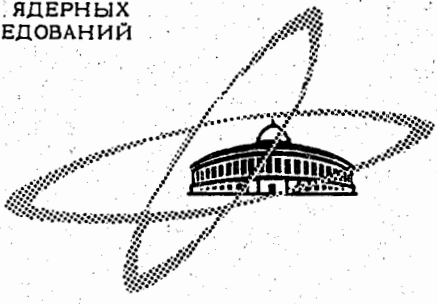

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SYNTHESIS AND STUDY OF NEW ISOTOPES AND ELEMENTS

Investigations using accelerated complex nuclei are being intensively developed today in middle energy nuclear physics. Thus, the ways inaccessible to other methods of synthesis are opened, which lead to the study of new isotopes and elements in a wide range of nuclear atomic numbers Z and mass numbers A.

These possibilities attract particular attention and in recent years a number of heavy ion accelerators have been designed in different countries all over the world: in the USA several accelerators of heavy ions have been devepoled and are being constructed now (omnitron, tandems and heavy ion cyclotrons), in France an accelerator is under way, with which aid it seems possible to obtain crypton with an energy required to overcome the Coulomb barrier of heavy ions, in West Germany a powerful linear accelerator is being constructed.

What are the main trends in the investigations with the beams of accelerated complex nuclei?

First of all, it is the synthesis of new elements and isotopes in a wide range of Z and A, then - the study of nuclear interactions of heavy ions, including those accelerated up to relativistic energies, and, finally, the utilization of heavy ions for a number of investigations in solid state physics and radiochemistry.

In the Soviet Union experiments on the new element synthesis were started in 1955 at the Kurchatov Atomic Energy Institute in Moscow on the cyclotron U-150 designed for the acceleration

of light particles. Beams of heavy ions were accelerated there. A number of so far unknown isotopes was studied and an isotope of a new element 102 was synthesized.

In these first experiments on the synthesis of far transuranium elements the intensity of the beams of accelerated ions was insufficient and, besides, a set of accelerated particles did not permit staging required experiments widely.

In this connection, it was decided to construct at the Joint Institute for Nuclear Research in Dubna a special accelerator of multicharged ions and in 1961 the cyclotron with a 310 cm diameter of pole dees and the highest parameters in the world (set of particles, their intensity etc.) went into service. Due to the proper choice of the heavy ion acceleration technique (cyclotron technique) and the high parameters of the ion source worked out at the Kurchatov Atomic Energy Institute, there were obtained fluxes of accelerated ions of neon and argon with the intensity up to 10^{14} ion/sec. With the beams of such particles experiments on the study of the mechanism of interaction of complex nuclei were conducted, special attention being paid to the problem of new element synthesis.

Both in the process of the synthesis of kurchatovium and during the production of 102 and 103 elements it was required to use neutron-enriched separated isotopes of oxygen ^{18}O, neon ^{22}Ne, nitrogen. Moreover, the targets irradiated with the beam of such accelerated particles had also to be neutron-excessive (^{242}Pu, ^{243}Am). The isotopic industry of the Soviet Union provided materials required for experiments on the synthesis of new elements and isotopes, which were performed with the heavy ion beams from the cyclotron of multicharged ions of the Laboratory of Nuclear Reactions of JINR.

Several tens of the far transuranium isotopes were obtained and their properties were studied. In 1964 element 104 was discovered and named kurchatovium. Though in the process of synthesis only extremely small quantities of element 104 are produced, in 1966 its chemical properties were investigated and it was shown that the far elements obey the regularities predicted by the Mendeleev periodic law.

The synthesis of kurchatovium and the study of its chemical properties took about six years. When the new element had been synthesized, it became evident that its chemical investigation would require an entirely new rapid method. Conventional chemical method did not fit: reactions in water proceed too slowly to investigate the chemical properties of kurchatovium during its half-life (0.3 sec). Great difficulties arose from the fact that one had to analyse individual atoms.

Element 103 became "closing" in the actinide family. It was known that actinide chlorides may be precipitated on the filter. Whereas element 104, according to the periodic law, should have properties similar to those of hafnium forming volatile compounds with chlorine, which do not settle on the filter. This analogy underlies the method of the chemical investigation of the new element.

The experimental procedure was the following. The recoiled nuclei were stopped by the nitrogen stream in the chamber behind the target of plutonium - 242. This stream picked up the nuclei of the 104th element recoiled from the target and accessory products and transported them into a volume where they were chlorinated by interaction with a stream of chlorinating agent-zirconium tetrachloride so that tetrachloride of 104 element and actinide chlorides were formed. Carried by the chlorinating agent, which at the same time played the part of a bearer, these compounds reached the filter, where the actinide chlorides were precipitated and element 104 tetrachloride passed through the filter. The temperature during chlorination was 350°C, what presented high requirements to the corrosion resistance of the apparatus. Due to the measures taken in this direction, the background was practically eliminated.

After the filter the reaction products passed into a chamber, where fission fragments were registrated by mica detectors. Since the filter shut the way for actinides, the tracks on the detectors may be only those of element 104. They undoubtedly indicated that kurchatovium is an analogue of hafnium.

Intensive beams of the accelerator U-300 and the application of various identification techniques permitted six isotopes of element 102 to be also synthesized. Probability of forming the nuclei of the far transplutonium elements in heavy ion reactions is rather small. Thus, from 10 milliard compound nuclei with the charge 104 resulting from the fusion of neon (Z=10) with plutonium (Z=94), under optimal conditions only one nucleus may be the subject of physical and chemical investigation, while the others break into fragments during 10^{-15} sec, due to the excitation energy introduced into them by neon ions.

It should be noted that the hopes of American scientists for the synthesis of far elements in the neutron fluxes of the thermonuclear explosion were not justified. Only one new isotope of fermium (Z=100) was so far obtained and there are some grounds to believe that in this way one can not go further than element 102.

The synthesis of new elements and the study of their properties is one of the basic problems of experimental nuclear physics. There are about 300 stable isotopes, and what about radioactive, ones, produced by all experimental methods available during 32 years since the discovery of the artificial radioactivity by the Joliot-Curies, they account for 1500 (see Fig.1).

The question arises naturally: what new radioactive nuclei might yet be obtained, how many of them altogether and what are their properties? The answer to this question was found in a number of theoretical papers based on the available experimental material and present conceptions of nuclear stability. Figure 1 presents the range of nuclei, for which the expected life time is not less that 10^{-10}sec. It is much more extensive and contains 3000-4000 isotopes.

Though it is very important that a result of the investigations of isotopes in this region nuclear physics will get a great deal of new objects for study, it is no less important that in this region are nuclei over-enriched in protons and neutrons, which are impossible to be synthesized by other methods. The study of such "saturated" nuclei like, for example, $^{31}_{20}Ca$ or $^{70}_{20}Ca$ is especially valuable, since it allows obtaining more extensive data than the study of isotopes with the ratio of the numbers of protons and neutrons close to "normal".

The studied properties of the far transuranium elements synthesized in reactions on heavy ions at the Laboratory of Nuclear Reactions permitted one to outline the directions of the synthesis of higher elements in new regions of stability with Z ranging form 114 to 126 (fig.2).

In this connection, let us examine fig.2, which shows the dependence of half-lives on the number of neutrons in a nucleus for even nuclei of element 102 relatively to spontaneous fission and alpha decay. The influence of the closed neutron subshell of 152 neutrons is strongly pronounced- the half-lives T_{sf} and T_{α} of the isotope $^{254}102$ exceed the corresponding periods of the adjacent even isotopes approximately by two orders of magnitude. It should be particularly emphasized that the cross section of the formation of the isotopes with a closed subshell is also higher than that of the formation of the adjacent isotopes. On this ground it should be expected that for nuclei with closed neutron and proton shells in the region with high Z the "shell" correction will compensate for the influence of Coulomb forces and double magic nuclei will be relatively stable against spontaneous fission and alpha decay. Cross sections of such nuclei formation will be considerably higher than those of the adjacent isotopes.

The calculations of possible closed neutron and proton shells and their influence on the stability of nuclei in the range of Z=114, 126 and N=184 were made in the Soviet Union by V.M.Strutinsky et al./1/ and W. Swiatecki et al. in the USA /2/. From their data the half-life relatively to the spontaneous fission of "supernuclei" with closed proton and neutron shells may amount to 10^{16} years (Z=114, A-Z=184). Theoretical estimations have shown, however, that the life time of such nuclei is limited by order modes of decay and does not exceed several years (see fig.3). Element 114 and adjacent ones lie in the beta-stability region. Thus the element

close to 114 might possess a significantly long life time as well. These estimations were somewhat unexpected and allowed to hope that the synthesis of superelements is possible.

The results reported this summer by Professor Powell turned out to be very surprising. He told that nuclei with the charge $Z \geq 110$ were discovered in cosmic rays by the group of Professor Fowler. Later on, English scientists came to the conclusion that the charge of these particles is equal to 106.

They used nuclear photographic emulsions exposed for hours at a high atmospheric depth over the Earth surface. The width of the etch pits on the tracks of particles in the emulsion is proportional to Z^2. Cosmic iron nuclei were observed reliably and with great statistics (Z=26). The charge of nuclei of higher elements is determined by extrapolating from this value to larger Z. However, the extrapolation from Z=26 to Z ≈110 is too far, so it may cause a great inaccuracy in determining Z. In our opinion, there is a possibility of more accurate determination of Z for cosmic heavy nuclei. The calibration of emulsions may be performed during the measurements, provided statistics being sufficiently large. This is for lack of long-lived isotoped in the range of atomic numbers from 83 to 90, which might reach the Earth undecayed. Therefore, if statistics is sufficiently large, a "gap" in the values of Z will be observed just in this region, and since its boundaries are distinctly determined by the values of Z=83, Z=90 the required calibration data will become available to experimentalists and this will make it possible to determine the atomic numbers of "superelements" coming to the Earth from space with a sufficiently great accuracy.

It should be noted that the time of flight of these supernuclei from their sources, say , from supernew stars , to the Earth is of the order of hundred million years. Therefore, if these nuclei reach the Earch undecayed, their life times should be at least of the same order, that is 10-100 million years. The synthesis of isotopes of elements 102, 103 and 104 and the study of their properties allowed one to estimate reliably life times of the total of isotopes in the re-

gion of ordinary numbers 101-108. According to our estimates, life times of the most long-lived isotopes in this region do not exceed several days[3] . Therefore, among the cosmic heavy nuclei with Z > 100 only nuclei of elements belonging to a new stability region with atomic numbers close to Z= 114 could exist.

Chemical investigation of kurchatovium made it clear that beginning from Z=104 an outer electron shell is already populated that is kurchatovium turned out to be a chemical homologue of hafnium. Proceeding from the latter one can undoubtedly predict chemical properties of higher elements in the wide region of values Z as well. In particular, chemical properties of element 114 can be predicted. No deviations from the normal population of 7p-shell could be expected in the 104-114 range of atomic numbers. In this case element 114 has to be a chemical homologue of lead. This enables one to hope for the 114 element discovery in lead minerals. Basing on the data obtained by Fowler et al.[4](according to the latter, about 1 mg of "eka-lead" falls down to the Earth per 24 hours) one can conclude that the possible concentration of "eka-lead" in the primary lead could not exceed 10^{-14} g/g at the assumption that the half-life of "eka-lead" is of the order of 100 million years. At these conditions one could observe the spontaneous fission effect with the seeming $T_{1/2} = 10^{22}-10^{23}$ years in the earth lead. So in order to detect spontaneous fission events in the natural lead one needs to develop a technique which allows using large quantities of matter and which is not sensitive to the fission fragment background due to cosmic rays etc. Later on attempts could be made to enrich the "eka-lead" fraction using differences between its chemical properties and those of the ordinary lead.

The artificial synthesis of superelement isotopes is the most promising one. The heavy ion method is the only possible way of superelement synthesis available so far. However, only short-lived isotopes of superelements could be produced by this method by means of existing technique of heavy ion acceleration, since the uranium nuclei accelerators are not constructed yet. From fig.3 which

shows the dependence of isotope half-life upon various kinds of decay as a function of a mass number one can see that the half-life of element 114 isotopes increases strongly with the increase of the neutron number in a nucleus. (The diagram is plotted on the basis of the data of one of Swiatecki's calculations). Therefore in the process of the element 114 synthesis in the reactions via a compound nucleus there is need to bombard the neutron enriched targets with accelerated complex nuclei which A/Z ratio is rather high. The most appropriate reaction is the following one:

$$^{244}_{94}Pu + {}^{48}_{20}Ca \rightarrow {}^{292-xn}_{\ \ 0}114 + x{}^{1}_{0}n \ .$$

Both calcium-48 and plutonium-244 could be accumulated only by means of high developed industry. In such a reaction element 114 could be already synthesized with a half-life sufficient for its identification.

The synthesis and investigation of new isotopes and elements in the heavy nuclei fusion reactions resulting in compound nuclei formation of the type $^{476}184(^{238}U + {}^{238}U)$ hold more promise. These compound nuclei undergo fission practically immediately. However, among their fission fragments nuclei of element 114 with a large value of the mass number can be found too, a relatively large yield of superelement nuclei with a large half-life being expected.

Thus , the main trends in the synthesis of new isotopes and elements could be pointed out:

1) Production of isotopes and elements in heavy ion reactions via a compound nucleus. Neutron-enriched separated isotopes are of particular importance in this method.
2) Synthesis of new isotopes and elements in the reactions of the type U+U. The development of these methods is connected with the special accelerator construction.
3) Search for and identification of new elements in cosmic rays and natural minerals.

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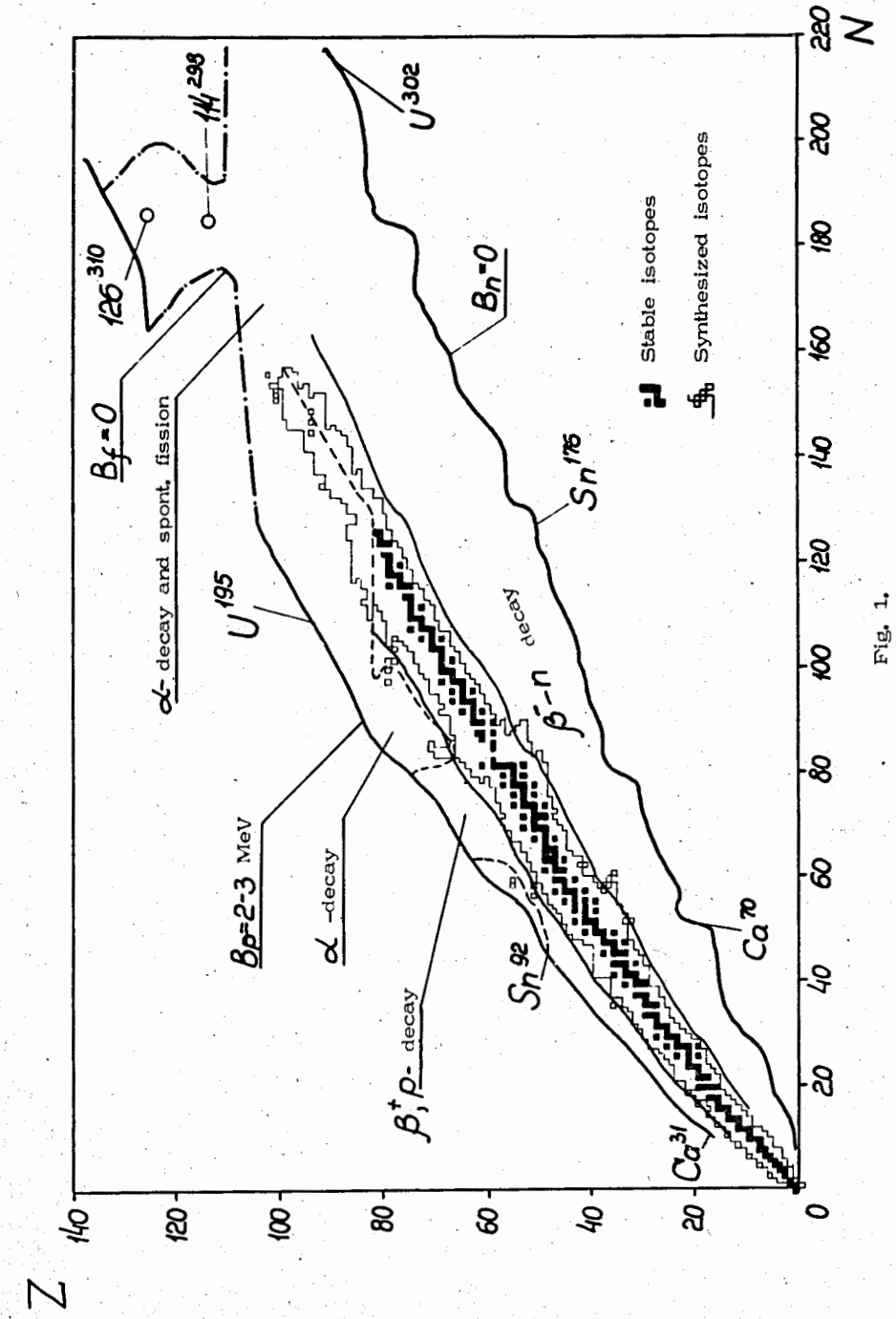
Z
N
Stable isotopes
Synthesized isotopes
α- decay and spont. fission
$Bf=0$
126^{310}
114^{298}
U^{302}
$Bn=0$
Sn^{176}
U^{195}
$Bp=2-3$ MeV
α -decay
β^--n decay
β^+,p- decay
Sn^{92}
Ca^{70}
Ca^{31}

Fig. 1.

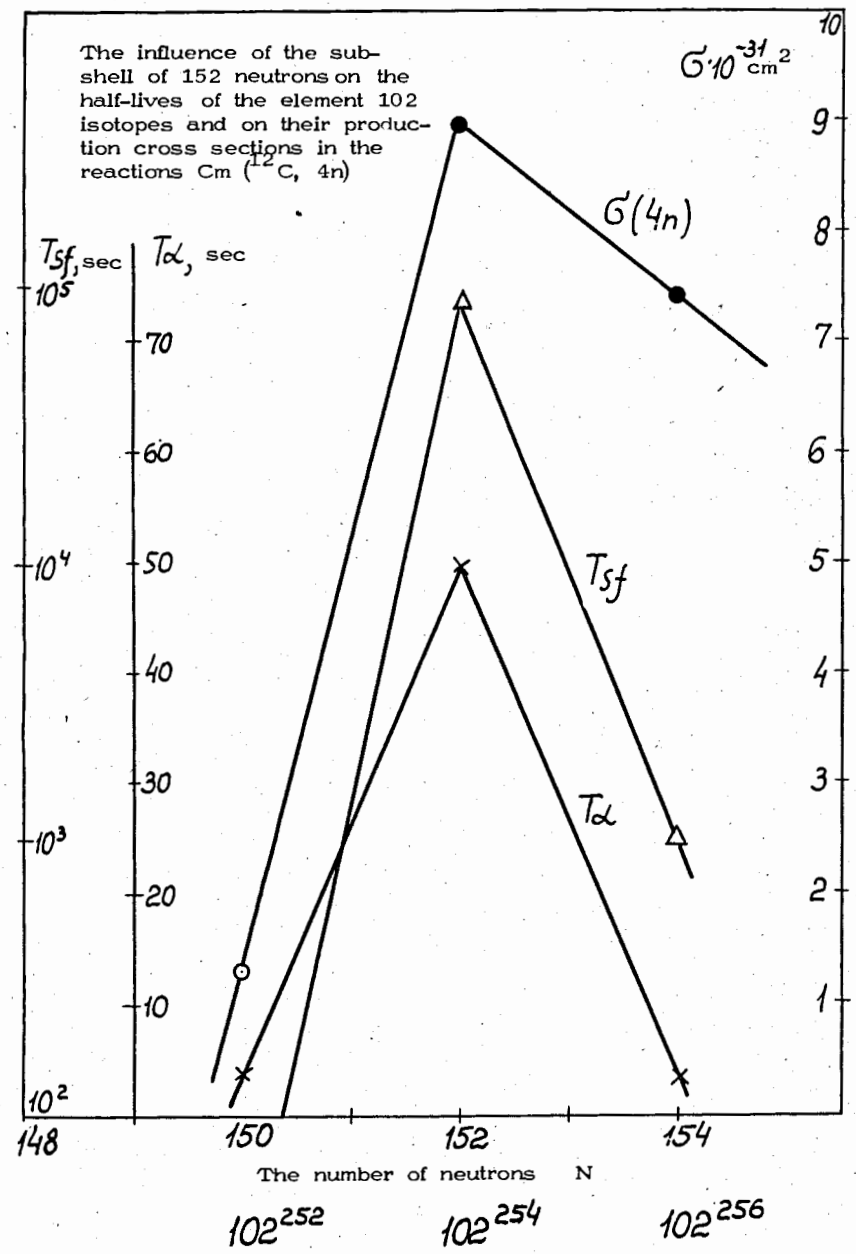
The influence of the sub-shell of 152 neutrons on the half-lives of the element 102 isotopes and on their production cross sections in the reactions Cm (^{12}C, 4n)
$\sigma \cdot 10^{-31}$ cm^2
T_{sf}, sec
T_α, sec
$\sigma(4n)$
T_{sf}
T_α
The number of neutrons N
102^{252}
102^{254}
102^{256}

Fig. 2.

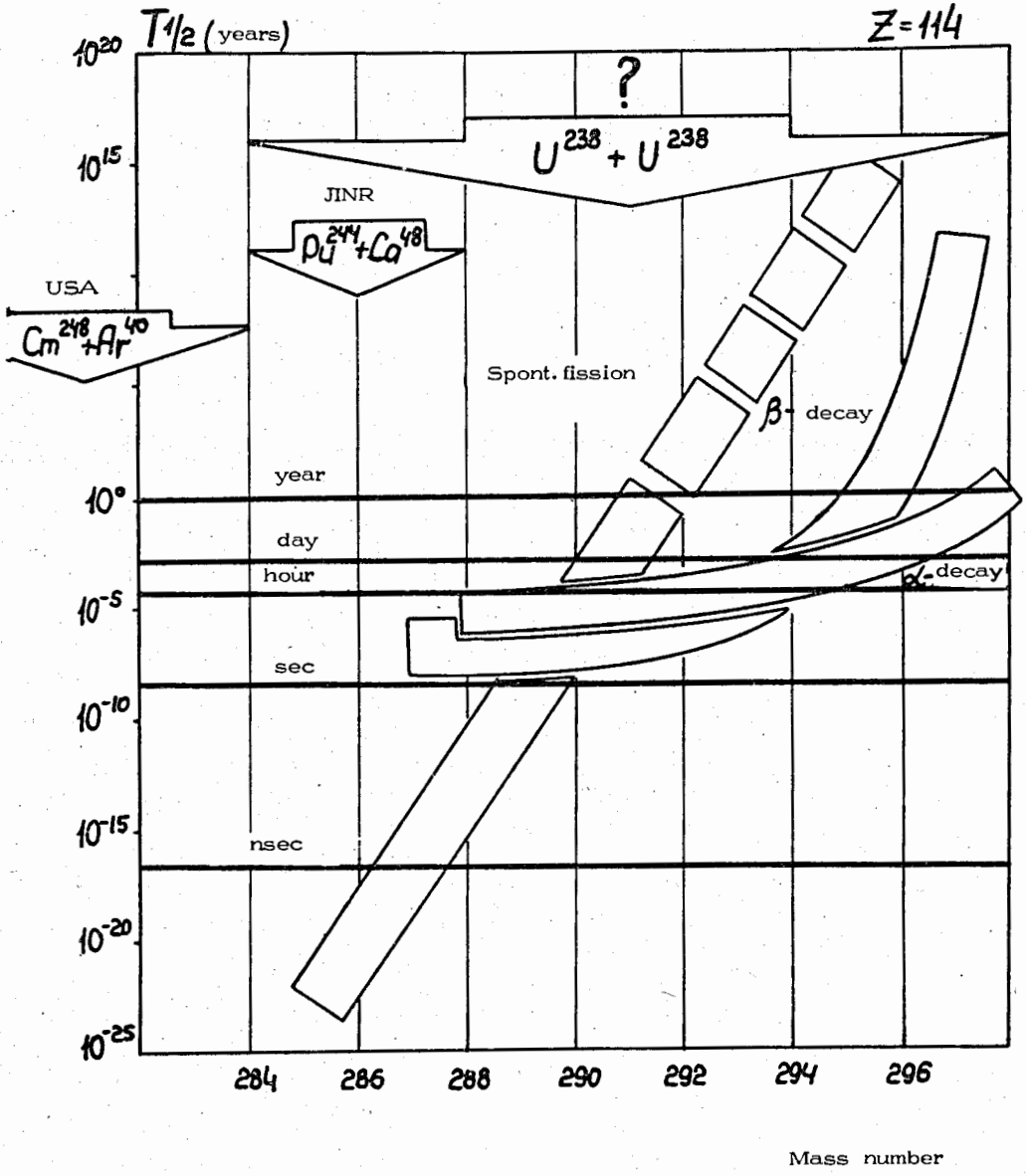

Fig. 3.

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