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PROSPECTS OF SYNTHESIS OF NEW ISOTOPES AND ELEMENTS*

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NOWADAYS it is difficult to imagine that there are people anywhere in the world who do not know the significance of the word "radioactivity" or what radium is. Marie Sklodowska-Curie together with Pierre Curie gave humanity both the element and the concept.

The extensive and systematic investigation of natural and subsequently also of artificial radioactivity started with that great discovery. Indeed, this turned out to be an invaluable contribution to our concepts of the structure of matter and to our knowledge about the atomic nucleus.

Nuclear physics investigates the structure of both stable and radioactive nuclei. The larger the number of the various isotopes which have been investigated, the clearer a picture of the structure of nuclear matter is revealed. It is therefore natural to analyze how far we have progressed in this direction, how many isotopes have already been synthesized and investigated, and how many must still be obtained and investigated by experimenters in the future. An answer to this question can be obtained by considering Fig. 1. This figure shows the isotopes which have already been synthesized and the possible isotopes with various Z (Z is the number of protons in the nucleus) and N = A – Z (N is the number of neutrons in the nucleus and A is the mass number) which can apparently be studied in the future. Stable isotopes are marked by little black squares. About 1500 additional isotopes with around a hundred of them in the transuranium region were obtained by various methods in nuclear reactions after the Joliot-Curies discovered artificial radioactivity in 1934.

The scale of the diagram is so small that nuclear fission which determined the technological progress of the twentieth century left only small excursions on this isotope chart. The outer contour indicated by a continuous line marks the boundaries of the region of stability obtained on the basis of theoretical calculations. It is readily seen that the region of synthesized isotopes is very small compared with the region which remains to be filled. The estimated number of possible isotopes in this region is four to five thousand. This is considerably more than the number obtained so far.

Two questions arise: 1) what can one expect from an investigation of these isotopes and, consequently, should they be obtained? 2) can they be obtained?

In the opinion of the body of physicists and chemists working in Dubna, one should undoubtedly attempt to obtain these isotopes and investigate them. Synthesizing such isotopes as, say, calcium-70 or calcium-31, we have at our disposal nuclei with very unusual ratios of numbers of protons and neutrons (20p and 11n or

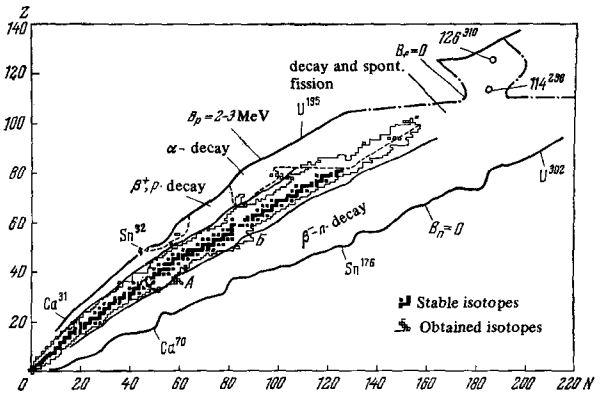

FIG. 1.

20p and 50n). In this case we can study nuclear matter in extremely unusual states. Such a methodological approach is very characteristic of physics in general. Thus, for example, investigators study the behavior of matter in strong magnetic fields, at very high pressures, under conditions of high electric fields, etc. From this point of view the study of nuclear matter when the ratio of Coulomb and nuclear forces is unusual will allow one to obtain very useful information about its properties. In this context one can understand the special interest in the study of far transuranium elements where both the Coulomb and nuclear forces are very great.

How can new isotopes be obtained? Attempts were made to solve this problem by many methods. In particular, underground nuclear explosions were used in the U.S.A. to obtain very heavy isotopes of transuranium elements. The essence of this method consists in the fact that in neutron fluxes of limitingly large density nuclei capture several neutrons one after another without having time to undergo beta decay. This makes it possible to obtain from the initial uranium-238 very heavy uranium isotopes which transform after several beta decays into further removed elements.

The first experiments were not specially intended for obtained heavy isotopes. The 100th element with a mass of 255 was discovered in 1952 as a result of an explosion (not underground but above ground) without a very large neutron flux. Starting from very general considerations, one could assume that if one were to obtain much larger fluxes and if one were to carry out a specially rapid chemical analysis, then this should lead to success in an attempt to advance to higher atomic numbers. Optimistic estimates indicated that one could thus obtain elements with a large value of Z. It was expected that one could then synthesize the 110th, 112th element, etc.

There are few instances in which so much effort was lost to reach a goal—the obtaining of large neutron fluxes and the chemical extraction of the trans-

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uranium fraction, but scientists were inspired by the lofty task of penetrating into the region of long-lived isotopes of remote elements and we believe that everything possible was in general done. However, only the isotope of the 100th element with the mass 257 was successfully obtained by 1966, i.e., in the course of fourteen years this method resulted in no progress in Z and in all in two units of A. It does not appear to us that this indicates that the method is completely hopeless; there exist apparently some additional possibilities. It is seen, however, that there is a physical reason which has so far not made it possible to proceed by this method into the region of new elements.

A second method of synthesis of new nuclei whose development began in 1954 in the U.S.A. and in the U.S.S.R., apparently on the same conceptual basis, is an attempt to utilize heavy accelerated particles: carbon, oxygen, neon, and argon. In principle two procedures are possible in this case. First, pieces of nuclear matter, say five to seven neutrons together, may break off from the compound nucleus; thus instead of sequential capture the target nucleus can capture at once a whole complex of neutrons.

Reactions of this type were indeed investigated. Transfer of 5–8 neutrons was observed in Dubna. The cross section for such a process is small, although in this case we do not venture far from the region of stability. In order to penetrate into the region of instability (see Fig. 1), stable nuclei must be enriched with 40–50 neutrons. The cross section for such a process is so small that this method is of little promise even if heavier particles are used.

The second procedure consists in the fact that instead of a transfer process of neutron complexes one makes use of fusion reactions. Let us, for example, consider what one should obtain on bombarding lead-208 with neon-20. In this case the compound nucleus (uranium-228) will have an excitation of about 60 MeV. As a result it emits six neutrons and the light uranium-222 isotope is synthesized. This method yields neutron-deficient isotopes.

In particular, this method was used to synthesize proton emitting neutron-deficient isotopes. V. A. Karnaukhov and co-workers in Dubna, Bell in Canada, and subsequently also the American investigators R. Macfarlane, A. Paskanser, et al. succeeded in going a little outside the region of stability and nevertheless to observe about two dozen proton emitters.[1-3] On proceeding further into the region of neutron-deficient isotopes one should observe the two-proton radioactivity predicted by V. I. Gol'danskiĭ. This requires the emission of 15–20 neutrons. However, here a very strict law known to all who deal with synthesis of nuclei far from the stability region begins to operate. We would like to get away from this region as far as possible. At the same time, we utilize for this purpose nuclear processes which although they occur in 10^{-14} sec are in essence adiabatic. Therefore after the emission of 6–8 neutrons the probability of emission of neutrons by the nucleus decreases whereas the probability of fission of the nucleus of the emission of protons increases appreciably. In the case of neutron-deficient isotopes of medium masses the nucleus attempts to turn back and actually returns to the stability region emitting protons.

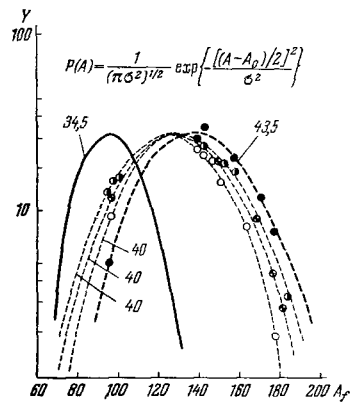

FIG. 2.

In the process of synthesis of transuranium elements there is a large probability of fission, as a result of which one obtains instead of a nucleus of the 100th element, for example, two nuclei with Z = 50. This indeed leads to the circumstance that in solving the problem of the synthesis of transuranium elements one must deal with catastrophically small cross sections. The latest experiments carried out in Dubna showed that whereas the cross section of the 102nd element of which six isotopes have been synthesized amounts to 10^{-9} barn, the cross section for the production of the 104th element is only 10^{-10} barn, and for the 105th element it is even smaller. All that can be done to increase the yield is to increase the intensity of the accelerated particles, or, instead of exposures of months, to carry out the experiment in the course of many years. Both of these methods are linear, whereas the decrease in the cross section is exponential. Therefore, in our opinion, the 105th element whose properties are at present being refined at Dubna will apparently be the last element that can be approached by this method.

A natural question is: is there a way out from the resulting situation? Analyzing the possibilities, we have reached the conclusion that nuclear fission is the main factor which makes the synthesis of new isotopes and elements difficult. Let us attempt to utilize a method sometimes used by experimenters—to make the interfering factor work to our advantage. We attempt to accomplish the synthesis of new elements by means of nuclear fission, increasing gradually the charge of the bombarding nucleus. The fission fragments will then also gradually increase their charge, and finally in the limiting case, say in the bombardment of uranium with uranium, one can obtain isotopes of remote transuranium elements, and not only transuranium ones, but at the same time also all the elements shown in Fig. 1. In addition this method is particularly promising for obtaining nuclei in isomer states of various types.

The idea appears sensible; however, one must carry out experiments which would indicate what is actually obtained, and how many isotopes of various elements are produced in the fission process. To this end Yu. Ts. Organesyan in conjunction with a group of chemists carried out during the past year in Dubna

systematic investigations of fission products of heavy nuclei under the action of multiply-charged ions. At first we turned to the work of 1956–1958 where comparatively light particles (carbon and nitrogen) were used to bombard uranium. A distribution curve with a maximum at Z = 50 was obtained in these investigations.

There arose the question of what would happen to the distribution curve when the charge of the incident particle is gradually increased. Undoubtedly, the maximum of the curve will be shifted, namely: a replacement of a particle with Z = 6 by a particle with Z = 22 (titanium instead of carbon) will lead to a compound nucleus with Z = 114, and fission into halves will yield Z = 57, i.e., a displacement of eight units will already have taken place. In addition, as V. M. Strutinskiĭ and co-workers showed, the spread of the distribution should increase.[4,5]

Figure 2 shows the distribution of fragments with respect to A for the case of uranium fission by a light particle with Z = 6 and by heavier particles with Z = 10 and Z = 18. The dashed curves show the dependence of the width of the distribution on the excitation energy. The experiment was carried out for several energies of the bombarding particles. A shift of the distribution maximum, an appreciable broadening of the curve, and a rather weak dependence on the energy of the incident particles are observed.

Experiments show that even when uranium is fissioned by neon this method of obtaining isotopes is appreciably more efficient than the usual method when fission is achieved by means of thermal neutrons. Inspite of the fact that in fission by thermal neutrons two peaks are obtained in the mass distribution of the fission fragments, the single peak of fragments in the fission of uranium by accelerated neon ions is so broad that for large A the isotope yield turns out to be larger by a factor of thousands and millions than in fission by neutrons (Fig. 3).

I recalled the fact that both transfer and fusion reactions are adiabatic processes. Therefore, synthesizing transuranium elements in nuclear fusion reactions with heavy ions, we obtain neutron-deficient isotopes lacking several—five, six and at times even more—particles. There is never any excess and the spread is very small. If, on the other hand, use is made of the fission process, then owing to the fact that

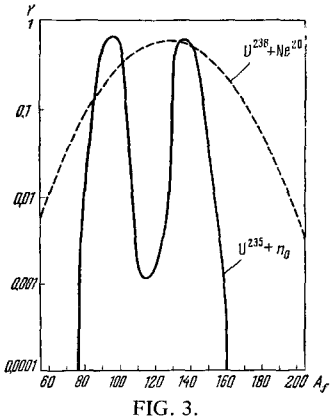

FIG. 3.

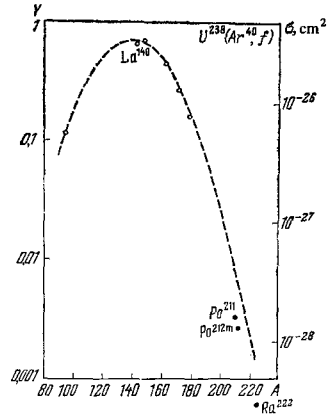

FIG. 4.

in this process there is a very complex distribution over excitation energies, the spread in A for given Z is also large. Of greatest interest perhaps is a recently obtained curve (Fig. 4). Bombarding uranium (Z = 92) with argon (Z = 18) we should have obtained a compound nucleus with Z = 110 and with fission into halves—nuclei with Z = 55. However, because of the spread in Z polonium and astatine (Z = 84, 86) were observed. This means that in this experiment we almost came close to the transuranium elements. Based on methods whose foundations were already laid many years ago by Marie Sklodowska-Curie, when polonium was first extracted from large quantities of ore, it has now been possible to study the A distribution for polonium on a large number of isotopes obtained in a uranium target bombarded with neon and argon ions.

The following figure (Fig. 5) depicts the cupola-like distribution of fission products for this case drawn in three dimensions in accordance with data from our experiments. Here the distribution in Z encompasses about 50 units and several dozens of isotopes are obtained for each given Z. Therefore, about 1000 isotopes are obtained even in an experiment in which we use comparatively light particles. There are no doubts that by accelerating heavier particles one can obtain many thousands of isotopes including isotopes of remote transuranium elements.

It has already been mentioned that it is essential to accelerate such particles as krypton, xenon, tungsten, and uranium. Special accelerator projects are at present being developed for this purpose all over the world; A. Ghiorso spoke of one of these in his lecture ("Synthesis of Transuranium Elements"). We at the JINR have chosen a way which differs somewhat from that followed by the U.S. physicists. At the present moment the ion source which operates in Dubna exceeds in its intensity of multiply-charged ions (with 7–8 charges) by a factor of 25 those obtained in the U.S. At the same time, not all the possibilities have so far been utilized in the sources. Our ion source was developed almost twenty years ago by L. A. Artsimovich and his co-workers for separating isotopes, and by changing its parameters but a little it was possible to obtain ions with 8–12 charges instead of singly-charged ions. We have full confidence that by making full use of the technology employed in plasma investigations we shall be able to obtain ions with 14–16

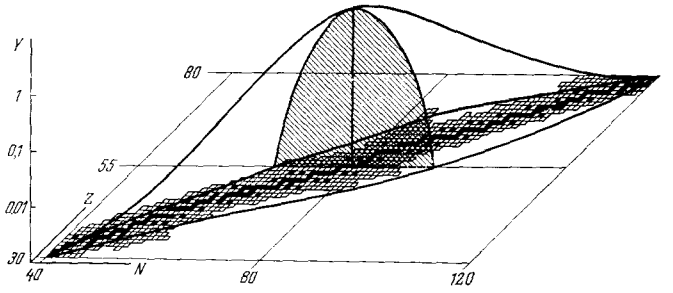

FIG. 5.

charges. Tungsten ions are being accelerated in the multiply-charged ion cyclotron of the JINR; true, so far only up to energies of about 100 MeV. Utilizing multiply charged tungsten (+10) and increasing the diameter of the device (but not as much as indicated in the lecture of A. Ghiorso), it will be possible to obtain tungsten and uranium ions with intensities of tens of microamperes and with the energies required for carrying out nuclear synthesis.

Investigations of ion sources reduce in essence to attempts to "drive" the largest power into a gas discharge. This is also done by those who are concerned with ion sources and controlled thermonuclear processes. It seems to us that here a new possibility has been discovered relatively recently. As a result of the development of laser technology it has been possible to obtain light beams of very high intensity. It suffices to say that a laser with an average power of 1 watt is equivalent to a pulsed source with a power of many hundreds of thousands of kilowatts. Naturally there appeared therefore thoughts of utilizing these gigantic powers for obtaining highly charged ions. The power of a laser is large, but the energy per quantum amounts in all to a few electron volts, and by means of these quanta one must detach from the atom electrons with binding energies of 100 and sometimes 1000 eV. Therefore, our attitude to the use of lasers was even as recently as a year ago not very serious; this was so up to the appearance of the first publications in which it was reported that ions with 8–10 and even 15 charges of such elements as calcium and iron are obtained as a result of irradiating metal plates with ultrapowerful lasers.

Apparently at such high powers there occurs something referred to by physicists as collective interaction with simultaneous absorption of a large number of light quanta, or some other processes take place which lead to electrical fields sufficient for the deep ionization of atoms. Therefore, along with further refinement of plasma sources, it is essential to develop laser sources.

If one considers the collective efforts in developing methods of accelerating the heaviest ions, then one will have in the first place to think about possible work directed towards the refinement of ion sources. The transuranium problem can serve as an example of such collective efforts in the solution of the most complex problems. Without entering into the fine details of the discussion of the chemistry of the 102nd element, I note that apparently the results of the American scientists substantially supplement our results based on gas chemistry. Of course, all this is far from what we should like to know about the 102nd element. The word " chemistry" includes both physico-chemical, electrochemical and many other properties, and it is essentially inexhaustible. The hope that we shall have a volume describing the properties of the 102nd element similar to the contents of Gmelin's handbook for other elements is, I believe, unfounded. But we can, say, determine the valence, which is also nominal. (It is well known that the valence depends on external conditions.) From this point of view it appears to me that gas chemistry—one of the basic methods of investigation used in our laboratory (the properties of both the 102nd and 104th element were studied by the method of gas chemistry)—represents already in itself a large step forward.[6,7]

There arises the question of the legitimacy of defining the physico-chemical properties of individual atoms. It should be said that when we began experiments on the chemistry of the 104th element we started from the premise that the chlorine compounds of the 104th element and the chlorides of hafnium will have analogous properties. Data on the temperature dependence of the vapor pressure for hafnium compounds were found in the textbooks. But how can one apply this concept to a single atom? A difficult situation resulted. It turned out, however, that an appropriate physical analogy exists for a single atom. If a single atom collides many times in an ion exchange column with a surface or settles on the resin and again escapes from it, then the number of adsorption and desorption acts is very large, and one introduces then instead of the vapor pressure the concept of the mean time during which this atom stays on the surface. This concept is valid and we have no doubt that the behavior of a single atom will coincide with the way in which a large complex of atoms would behave if there were no interaction between them. But the interaction need only be taken into account in weighable quantities; therefore, if we consider chemistry valid for hundreds of thousands of atoms, then it is just as valid for a single atom.

It appears to me that in this discussion, which will, I hope, still continue for a little while, this contrasting of chemistry and physics is very artificial. The fact is that if we deal with elements with live, say, millionth parts of a second then neither gas nor liquid chemistry will yield anything. At the same time, the nuclei will during those times capture K electrons and a study of the x rays will essentially yield as reliable data about Z as are, for example, obtained as a result of a chromatography study of the element.

Special consideration must also be given to the problem of the determination of the mass number of the new isotopes. In recent syntheses of short-lived nuclei, use has been made of so-called physical identification. According to these methods the mass numbers of the reaction products were identified, for example, from the excitation function of the given reaction, i.e., from the yield of the investigated product as a function of the energy of the bombarding particles. However, these methods of indirect identification do not always yield unambiguous results. A direct answer to the question regarding the mass number of an isotope can

be obtained by the use of a mass spectrometer or separator. However, this cannot be the usual variant of this instrument, but a rapidly operating one which permits one to work with very short-lived nuclei. As you well know, such instruments are at present constructed in a number of laboratories. Their main feature is that the products of nuclear reactions are rapidly separated from the target and rapidly transported to the ion source of a continuously operating mass separator. The detection of the decay of the separated isotopes is accomplished in the separator itself. Such a rapidly operating separator on line with a heavy-ion cyclotron is being constructed in Dubna.

It should be said that thus far the success of an experiment on the synthesis of isotopes is in many instances determined by the beam intensity, the isotope purity of the target, and the absence in it of impurities such as lead. Unfortunately, the first experiments on the synthesis of elements 102 and 103 did not satisfy these conditions, neither in the USA, nor in Sweden, nor in the Soviet Union. There were at that time no single-isotope targets and the obtained data could therefore be ascribed to this or that isotope. In particular, the isotope which was synthesized in the Swedish work "took on" in the course of a decade five values of the mass number from 251 to 255, and many years of work were required in Dubna[8] to synthesize and investigate the six isotopes of the 102nd element, in order to leave no room for ever new hypotheses of persistent authors.

The second circumstance also turned out to be important. During the initial stage of the experiments on heavy ions it was not clear that lead will be a very dangerous impurity. The yield of analogous alpha emitters for lead is larger by a factor of a million than the yield of the far transuranium elements obtained in irradiating plutonium, curium, or californium. Therefore the purification of targets of lead is absolutely essential. Since there is much lead in physical and chemical laboratories, this task proved to be very difficult. The laboratories are saturated with lead: soldering is done with lead solder, cables have lead jackets, shielding is made of lead, etc.

We have apparently succeeded in proving that the Swedish work did not deal with the 102nd element but with an irradiation product of lead, i.e., an isotope of the 88th element or a product of a stripping or fission reaction with $Z = 90$.

The work of synthesizing the 105th element also depends to a large extent on the purity of the target—only a limitingly small content of lead is admissible.

Another approach is to produce such conditions that even a single decay event is so reliable that it does not admit contradicting interpretations and that no background appears. In our work with the 105th element we are following both paths: on the one hand, we purify our elements of lead, and, on the other hand, we try to obtain a maximum of information per alpha decay, information that would absolutely exclude the possibility of the appearance of any interfering factors (the method of amplitude-time correlations).

The young participants in the work with the 105th element believed at one time that the work is complete. However, those complications which occurred with the 102nd element as well as the difficulties in the work concerning the 103rd element (our results conflict with what was published by American investigators in 1961), lead us to conclude that the investigation should be continued, inspite of the fact that the information about the 105th element which is at our disposal is more reliable than the data initially obtained for the 102nd and 103rd element.

I do not know whether I have succeeded in indicating the state of the problem at present and in the future. In the past there have apparently been very many hasty conclusions. This required an additional decade of work and it appears to me that we must now think more about the present and about the future. I have therefore not analyzed in detail the work of the Fifties and of the early Sixties.

If we are to recall the past, then only the remote past, namely the heroic efforts of Marie Sklodowska-Curie and Pierre Curie in their discoveries of the new elements. It should be mentioned that at that time they discovered new elements in the complete absence of data on their chemical properties, without application of any (from our point of view) complicated methods, but with a very exacting attitude towards their own results and a profound analysis of the obtained data. Therefore, in the work of the Curies there are essentially no instances similar to any extent to the situation that occurred with the discovery of the 102nd and 103 element, and which we hope shall not be repeated in the future.

In conclusion, allow me to express my hope that the problem of nuclear synthesis and of the investigation of numerous new elements and isotopes will be successfully solved by the joint efforts of scientists of the various countries.

[1] V. A. Karnaukhov, G. M. Ter-Akop'yan, and V. G. Subbotin, JINR Preprint R-1072, Dubna, 1962.

[2] A. M. Paskanser, R. M. Pherson, K. A. Esterlund, and R. L. Reeder, Report 10362 (1962).

[3] R. Barton, R. McPherson, R. E. Bell, W. R. Frisken, W. T. Link, and R. B. Moore, Canad. J. Phys. 41, 2007 (1963).

[4] Yu. A. Muzychka, Yu. Ts. Oganes'yan, B. I. Pustyl'nik, and G. N. Flerov, Yad. Fiz. 6, 306 (1967) [Sov. J. Nucl. Phys. 6, 222 (1968)].

[5] V. M. Strutinskiĭ, Yad. Fiz. 1, 821 (1965) [Sov. J. Nucl. Phys. 1, 588 (1965)].

[6] I. Zvara, T. S. Zvarova, T. Tsaletka, Yu. T. Chuburkov, and M. R. Shalaevskiĭ, JINR Preprint R-2548, Dubna, 1966.

[7] I. Zvara, T. S. Zvarova, R. Tsaletka, Yu. T. Chuburkov, and M. R. Shalaevskiĭ, Radiokhimiya (Radiochemistry) 9, 2 (1967).

[8] G. N. Flerov, JINR Preprint D7-3444, Dubna, 1967.

Translated by Z. Barnea