ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

E7 - 13040

G.N.Flerov

SOME NEW APPROACHES TO THE SYNTHESIS OF HEAVY AND SUPERHEAVY ELEMENTS

Submitted to Symposium on Nuclei Far from Stability (September 1979, Nashville, USA)

1980

We are living through the period when increasingly strong efforts are made to obtain qualitatively new results on the synthesis of heavy and superheavy elements. This fact forces us to critically examine what has been done in this field in the past decades. The history of synthesis of new elements in heavy ion reactions dates from the late 1950's. Elements of atomic numbers 102, 103, and 104 were synthesized [1] during the decade 1959-69. Since, elements of atomic numbers 105, 106, and 107 have been added to this list. Work on the synthesis of element 106 can be considered to be completed. At present two isotopes of this element with mass numbers 259 and 263 (refs. [2-4]) are known, and only chemical experiments remain to be carried out. There are certain indications of observation of a short-lived ($T_{1/2} \simeq$ $\simeq$1-2 ms) isotope of element 107 with mass number 261 (ref. [5]).

If we compare the efforts made to synthesize new elements with the rate of progress (see fig. 1), we have to express a considerable amount of entirely justifiable pes-

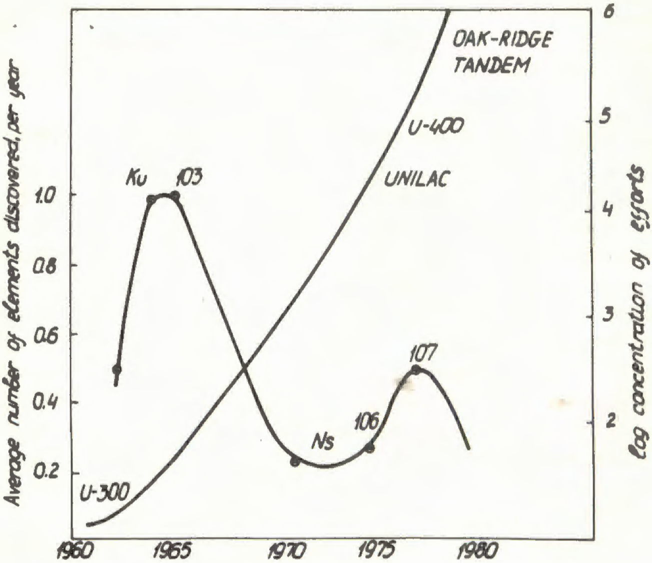


Fig. 1. An illustration of the rate of progress in discovery of new elements and the efforts concentrated (efforts made = number of accelerators x beam intensities x number of experimental setups).

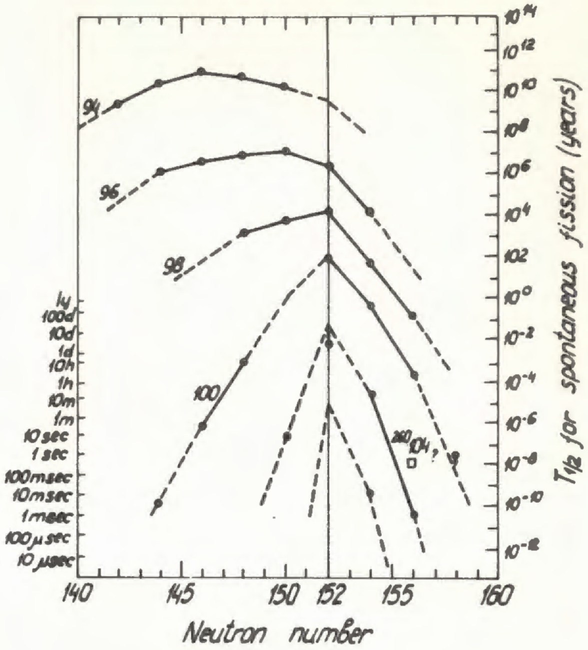


Fig. 2. Systematics of spontaneous-fission half-lives ($T^{SF}_{1/2}$), proposed by Ghiorso[6].

simism with regard to the situation that has occurred at the present time. However, despite this pessimism before considering new approaches to the problem of synthesis, it is worth estimating what else can be obtained by using traditional methods.

In the evaluation of the present situation, the fact attracts our attention that for many years further advance to the region of heavy nuclei has been objectively retarded under the influence of the known Ghiorso systematics[6] of spontaneous-fission half-lives, which were repeatedly referred to in review papers by Seaborg. If we believe these systematics shown in fig. 2, it is senseless to try to produce isotopes of elements of $Z > 102$, which are very neutron-rich compared with the "magic" number 152, since these isotopes should have vanishingly small lifetimes. Fortunately, as has been shown by the experiments carried out at Dubna, these predictions did not prove justifiable. In fact, as one goes from $Z = 102$ to $Z = 104$, the regularities of spontaneous-fission half-lives change drastically. In the region investigated, as the number of neutrons increases, a monotonic increase in $T^{SF}_{1/2}$ has been observed for the

$Z \geq 104$ isotopes. An increase in N by one unity corresponds to a $T^{SF}_{1/2}$ increase of, on the average, several times. This dependence has been convincingly traced in the case of the isotopes of kurchatovium - the element of atomic number 104 (see fig. 2). The recent experiments carried out by the Berkeley group[7] have yielded for ^{260}Ku a spontaneous-fission half-life value, which is still several times different from the value measured at Dubna. However, this is in principle not very important as both values are more than 10^4 times as large as that implied by Ghiorso's systematics.

The establishment of the new systematics of s.f. half-lives has helped one overcome a certain psychological barrier which prevented the performance of many reliable experiments. As a result, a number of heavy isotopes have been synthesized and their properties investigated (see fig. 3).

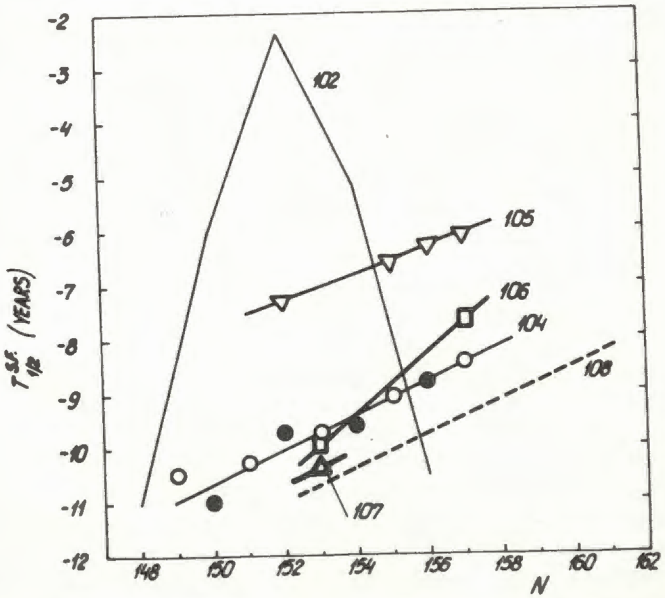


Fig. 3. Dependence of the $T^{SF}_{1/2}$ values on neutron number for the $Z \geq 102$ isotopes. The spontaneous-fission half-lives of the even-odd isotopes of kurchatovium (Z=104) are shown reduced by the average value of the hindrance factor (10^3). The extrapolation for the even-odd isotopes of element 108 is shown by a dashed line.

There are all grounds to hope that other neutron-rich isotopes will also be synthesized including the heavy isotopes of element 107 having lifetimes long enough for chemical experiments to be carried out. For this purpose the reactions $^{249}Bk + ^{22}Ne \rightarrow ^{266}107_{159} + 5n$, $^{249}Cf + ^{22}Ne \rightarrow ^{267}108_{159} + 4n$, $^{249}Cf + ^{26}Mg \rightarrow ^{271}110_{161} + 4n$, and others can be used. The group of Druin at Dubna has begun experiments to synthesize a heavy long-lived isotope of element 107, which very tentatively is expected to have a half-life of 1-2 seconds. Heavy ion beams from the U-400 cyclotron will make it possible to produce several tens of atoms of this isotope per day.

While in the field of synthesis of transfermium elements new ways of further advance have been outlined mainly because of the establishment of the new systematics of half-lives, the traditional methods used in the synthesis of superheavy elements have led to amazing results. Of course, the possibilities of these methods have not been exhausted yet. In this connection, a study of the reaction $^{248}Cm + U$, prepared jointly by the scientists of Darmstadt, Berkeley, Livermore and Oak Ridge would be of interest. This experiment, which is difficult from a technical point of view, seems to offer a novel possibility of synthesizing superheavy elements in the complicated process of the interaction of two extremely heavy nuclei. The performance of such experiments which we were dreaming of many years ago [8] causes the feeling of deep satisfaction. In all likelihood, these experiments will exhaust all the possibilities of such an approach to the synthesis of SHE. The proposal discussed by Seaborg et al. [9] to use the reaction $^{248}Cm + ^{244}Pu$ can hardly lead to radical changes in the existing situation. Moreover, its practicability is unlikely. Apparently, a more realistic way in this direction is to repeat the previous experiments at a substantially higher sensitivity to be achieved by increasing the beam intensity [10].

As to the other approach based on the use of complete-fusion reactions for the synthesis of SHE, a number of experiments still remain to be carried out. In the recent years, several experiments in which SHE were sought among the products of the complete-fusion reactions between the ^{48}Ca ions and ^{231}Pa, ^{232}Th, ^{233}U, ^{242}Pu, ^{243}Am, $^{246,248}Cm$ [11], and ^{248}Cm targets [12] have produced negative results. The experiments, in which the expected SHE were chemically extracted for about 1 hour and spontaneous fission events were detected were done especially thoroughly. The cross section sensitivity achieved was equal to 3×10^{-35} cm^2. The

new cyclotron U-400 put into operation at Dubna is capable of producing the ^{48}Ca ion beam with an intensity of 10^{14} part/sec, which will make it possible to increase the sensitivity by several hundred times.

The use of devices especially designed for the rapid separation of complete-fusion reaction products from the ion beam (see, e.g., [13]) will permit the search for SHE isotopes having very short lifetimes to 1 μsec.

However, despite the fact that some experiments remain to be performed, it is evident already now that one can hope for a daily production of several SHE atoms at best. This situation forces us to look for new approaches to the problem of reaching the region of stability.

One of such new approaches seems to emerge from the results of the experiments being carried out at Dubna. I shall remind you of the features of complete-fusion reactions, which play an important role in the synthesis of heavy and superheavy nuclides. The fissility factor of the compound nucleus seems to be the most substantial feature of these reactions. In order that the stabilizing effect of the closed nuclear shells might manifest itself, compound nuclei are usually produced with the lowest possible excitation energy. This can be achieved by choosing a projectile energy only slightly exceeding the Coulomb barrier. However, if the formation of a spherical compound nucleus requires an additional amount of energy, the probability of fusion near the Coulomb barrier may turn out to be considerably smaller than that expected, and this will lead to a many orders of magnitude decreases of (HI, xn) reaction cross sections compared with the available estimates.

Similar qualitative considerations were used to explain the negative results of the so far performed experiments aimed at the synthesis of SHE. However, Oganessian, Gierlik and Penionzhkevich have noticed that the close relationship between the interaction barrier and the minimum accessible excitation energy of the heavy fissionable nuclei formed in complete-fusion reactions may not occur in the case of such a mechanism of the interaction of complex nuclei, which involves the emission of energetic light particles.

The emission of such "pre-equilibrium" particles (neutrons, protons, and α-particles) was investigated by many authors. However, to clarify the possible role of this process in the synthesis of heavy fissionable nuclei, special experiments are needed, some of which have been carried out at Dubna. In the experiments involving the bombardment of a gold target with the ^{22}Ne ions at a laboratory energy of 175 MeV, we investigated the energy spectra of the α - particles emitted in coincidence with correlated fragment pairs at different angles with respect to the primary beam. Semiconductor Si-Au detectors were placed so that the fission fragments from the nuclei produced during the emission of α-particles by the compound nuclei could enter these detectors. The spectra of the α -particles emitted at non-zero angles were measured using an annular Si-Au detector. In zero-angle measurements, the experimental setup shown in fig. 4 was used including also a wide-range magnetic spectrometer. Figs. 5 and 6, respectively, show the angular distribution of the α -particles detected in coincidence with fission fragments, and the α -particle energy spectrum measured at zero angle to the beam. The angular distribution exhibits a sharp maximum at zero angle, which clearly indicates the pre-equilibrium mechanism of α -particle emission. As one can see from fig. 6, the energy spectrum is characterized by an exponential fall extending up to an energy of about 120 MeV, at which it sharply ceases. The particles having an energy of about 120 MeV make up a 10^{-5} th fraction of the total yield. Bearing in mind that the total cross section of this process is about 1 barn, we can estimate the cross section for the reaction involving the emission of energetic α -particles to be equal to about 10^{-29} cm^2.More estimates show that the residue formed after the emission of an α -particle should have considerable angular momentum, and the formation of the states of such nuclei on the yrast-line is very likely.

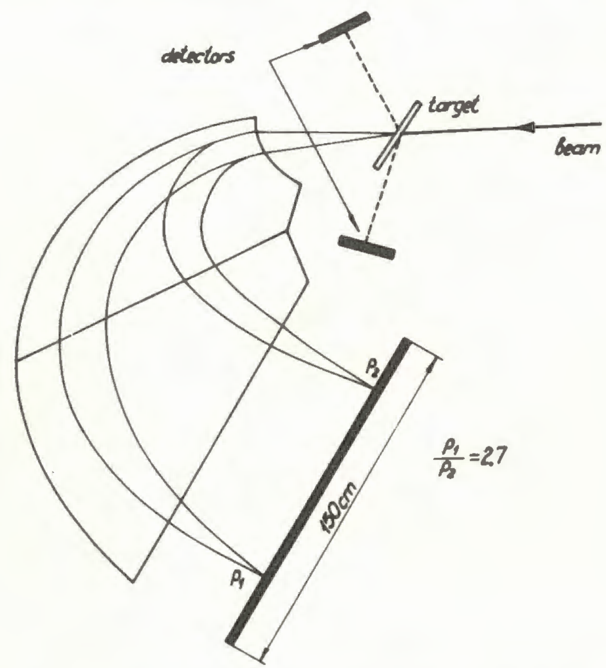


Fig. 4. Experimental setup used to study the spectra of α -particles emitted at zero angle.

In our view, a number of experiments should be carried out to clarify the details of the mechanism of the reactions mentioned. In particular, it is of interest to see how the α -particle spectrum and the cross section depend on the energy and mass of the bombarding ion. It is not

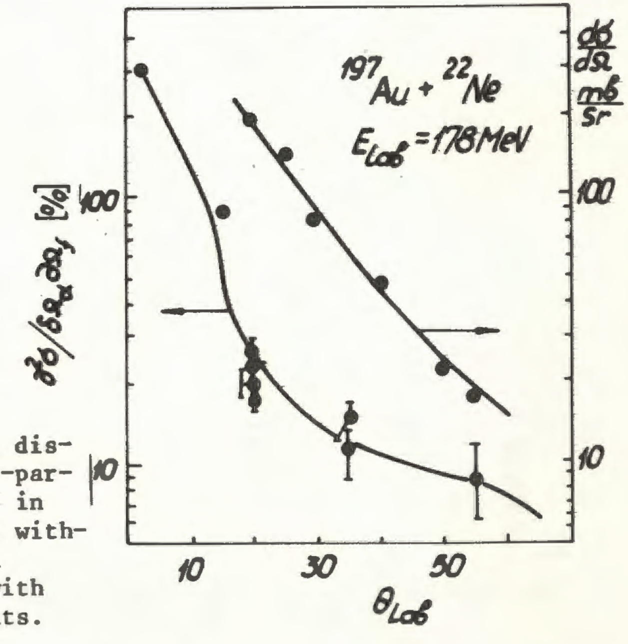


Fig. 5. Angular distribution of α -particles measured in correlation and without correlation (upper curve) with fission fragments.

excluded that the formation of rotating "cold" nuclei occurs at a projectile energy substantially exceeding the Coulomb barrier. The de-excitation probability seems to be determined for these nuclei by the competition between fission and γ-ray emission. The studies of the fission of such nuclei can yield experimental information on the fission barriers of states with large angular momentum. It is also very interesting to determine the multiplicity of γ-rays, their average and total energy, and to search for yrast traps and fission from isomeric states with large angular momentum. It is evident that the possibility of using the described mechanism of reactions to synthesize transfermium nuclides and SHE will be determined by the angular momentum dependence of the fission barrier of heavy nuclei. Much should be clarified by direct experiments such as those aimed at the determination of the probability of the reactions $^{238}U(^{18}O, \alpha)\,^{252}Cf$ and $^{238}U\,(^{22}Ne, \alpha)\,^{256}Fm$.

If the cross sections of these reactions turn out to be rather large, the (HI, α) and (HI, α1n) reactions can probably

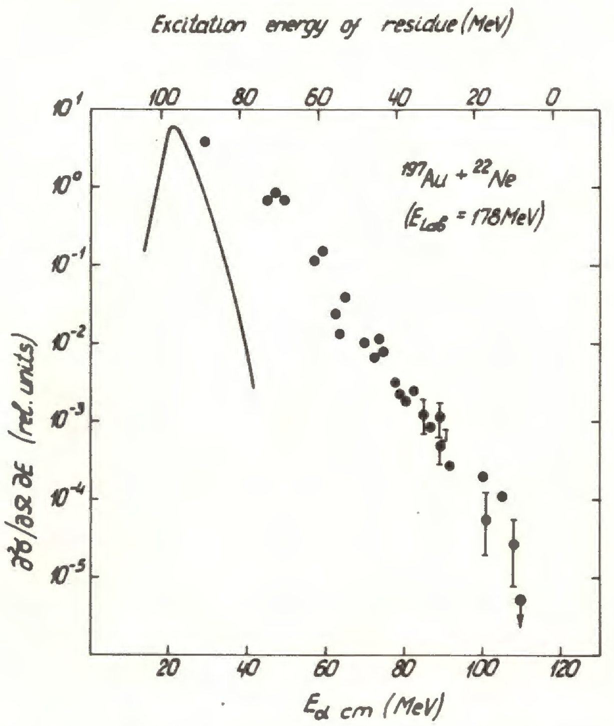


Fig. 6. Energy spectrum of α-particles emitted in the reaction $^{197}Au + ^{22}Ne$ (E_{lab} = 178 MeV) at an angle of 0-2° to the beam axis. The solid curve shows the calculated evaporation spectrum.

be used to synthesize the isotopes that are not producible by conventional complete-fusion reactions with sequential neutron emission. For instance, heavy isotopes can be produced in the reactions $^{249}Bk(^{22}Ne, \alpha)\,^{267}105$, $^{249}Cf\,(^{22}Ne, \alpha)^{267}106$, $^{249}Cf(^{26}Mg, \alpha)\,^{271}108$, and others. It is not excluded that such reactions may prove to be advantageous in terms of the SHE synthesis because of their attractive features such as, first, the possibility of producing superheavy nuclei in slightly excited states and, second, the possibility of producing isotopes lying closer to the β-stability line compared with usual complete-fusion reactions.

In the nearest future we are planning to carry out experiments to synthesize various nuclei by using nuclear reactions accompanied by the emission of pre-equilibrium particles. If we succeed in producing sufficiently high yields of the known isotopes of transuranium elements, it will be reasonable to use these reactions in the future experiments aimed at the synthesis of heavy and superheavy nuclei.

As to superheavy atomic nuclei, in our view, in considering the methods of their artificial production it is appropriate to bear in mind the possibility of detecting one or two naturally occurring long-lived nuclides from the hypothetic region of stability. Searches for such nuclides have been conducted by several groups since the late 1960's.

The history of these searches knows the periods of ups and downs in the activity of investigations, the latest sharp rise being associated with the report /14/ of an American group on the simultaneous observation of three superheavy elements in Madagascan monazites. It is known that the hopes have not materialized and this has caused a certain pessimism, which is also due to an excessive confidence in some theoretical calculations predicting short lifetimes for superheavy elements. However, taking into account that the prediction of spontaneous-fission half-lives of nuclei involves great difficulties and cannot be made with an accuracy better than 10 orders of magnitude, we do not share this pessimism. Moreover, the importance of this problem impels us to make considerable efforts in searches for superheavy elements in nature.

One of the directions in which searches are being carried out at Dubna /15/ is the study of the mass spectrum of the heaviest atomic nuclei in galactic cosmic ray nuclei. For this purpose, we use the property of silicate minerals incorporated in the stony-iron meteorites (pallasites) to record and preserve the tracks of galactic nuclei with $Z > 20$

for several tens and hundreds of million years. Mere estimates show that 1 cm^3 of such minerals located at a depth of 5 cm from the preatmospheric surface of the meteoritic body is expected to preserve for a period of 10^7 - 10^8 years several hundreds of tracks due to nuclei belonging to the uranium-thorium group. The investigations were carried out using olivines from Marjalahti, Eagle Station, and Lipovsky Khutor meteorites of the radiation ages of 175, 45 and 220 million years, respectively. These olivines were calibrated by xenon to titanium heavy ions, and the thermal stability of the tracks of atomic nuclei was investigated. By now, a total of 570 mm^3 of olivines have been investigated. Some data on the abundance of the $Z > 50$ nuclei in galactic cosmic rays have been obtained and the lower limit of the abundance of superheavy nuclei $(Z \geq 110)$ has been determined to be at a level of 1.6×10^{-9} relative to the iron group nuclei. The sensitivity achieved in our investigations was more than 10 times that obtained in all the experiments carried out since 1967, with nuclear emulsions and plastics (see ref. /16/).

Another trend pursued at Dubna during the last decade (see ref. /17/) is the search for long-lived spontaneously fissioning isotopes of superheavy elements in nature. To detect the rare events of spontaneous fission in different samples, we use neutron detectors installed in a salt mine 1100 m.w.e. underground in order to eliminate the cosmic ray background. This circumstance and a number of other measures taken have made it possible to achieve a sensitivity level at which, for samples weighing 10 kg, one could with confidence detect the spontaneous fission of a total of 2×10^{10} atomic nuclei with a half-life of 10^9 years. This corresponds to a concentration of 10^{-15} g/g. The indicated sensitivity is a factor of more than 100 larger than that obtained by other groups including, in particular, the Oak Ridge one /18/.

In 1972-75, by using neutron detectors we carried out measurements with the samples of carbonaceous chondrites which are generally known to belong to the Solar System's formations that are least differentiated in elemental abundance and are not depleted of heavy volatile metals (Hg , Tl , Pb, and Bi), whose homologues the hypothetical superheavy elements are supposed to be. As a result of these experiments, the multiple emission of neutrons has been revealed in the samples of Efremovka, Allende, and Saratov meteorites /19/. The observed counting rate was, on the average, one event per 5 days for samples weighing 10 kg, i.e., 10 to 30 times the rate for the spontaneous fission of the uranium contained in these samples. The possibility of explaining the observed phenomenon as being due to other sources of background was entirely eliminated. Therefore, we assumed that the meteoritic samples investigated contained a long-lived spontaneously fissioning nuclide, the concentration of which was 5×10^{-15} to 5×10^{-14} g/g or 10^{10} to 10^{11} atoms per kg of the meteorite.

In the subsequent experiments, in an attempt to find new samples more suitable for the extraction and concentration of the new nuclide, we investigated hot spring waters from the Cheleken Peninsula (the South-Eastern coast of the Caspian Sea), which are rich in heavy volatile metals, which seem to be separated together with volatile components from the comparatively little differentiated substance of the Earth's mantle. The spontaneous fission activity of the new nuclide was detected by measuring the multiple neutron emission in the samples of vinyl-pyridine anion-exchange resin, through which the Cheleken hot spring water was passed /20/. As a result of the elution of the mineral fraction from 170 kg of the resin, we obtained a 6 kg sample of precipitated hydroxide, which yielded the counting rate of spontaneous fission events of 5 counts per day. The thorough measurements of the α-activity of resin and hydroxide samples have shown that the Cm and Cf contamination, which, in principle, could penetrate the Cheleken water as a result of atmosphere fallout (nuclear tests) or during the treatment of the target material in the laboratory, are at least 100 times smaller than those required to account for the observed counting rate. Therefore we have drawn the conclusion that the Cheleken hot spring water also contains a new long-lived spontaneously fissioning nuclide. In our view, this nuclide lies in the region of superheavy elements.

Our current experiments are aimed at the extraction and concentration of the new nuclide from hot spring waters to carry out the unambiguous identification of the nuclide. In a number of extractions from the Cheleken Peninsula brines and other hot spring waters we have processed several hundreds of cubic meters of water by using the methods of ion exchange, cementation, extraction to the organic solvent, precipitation of hydroxides, and others. The results, being reproducible and mutually consistent, confirm the previous conclusions. During this work we succeeded in obtaining samples with a counting rate, which was a factor of 10^3 as large as that of the meteorite.

To determine the mass number of the new nuclide we are planning to use mass separators with very efficient ion sources. After the mass separation of the samples investigated the mass lines can be established in the region of $A > 270$ by detecting either spontaneous fission or the fission induced by intense γ-ray or neutron beams.

The unambiguous identification of the new nuclide can be done by measuring the energy of the X-rays induced by powerful synchrotron radiation. The equipment using the synchrotron radiation from a 20-pole Wiggler magnet, incorporated in the orbit of the VEPP-3 storage ring, permits such a measurement for 10^8 to 10^9 atoms of SHE at a concentration of 10^{-10} g/g in the sample.

Therefore we think it quite realistic that some time it will be possible to prepare targets from the heaviest accessible atomic nuclei-SHE nuclei. Already the present content of $\sim 10^{12}$ atoms in the samples available seems to be adequate for carrying out experiments on the synthesis of new atomic nuclei from the island of stability. We are currently involved in investigating one of such possibilities by irradiation with intense thermal and resonance neutron fluxes of the samples that contain the spontaneous fission activity. The integrated fluxes of 10^{22} thermal and 10^{21} resonance neutrons per cm^2 are accessible by high-flux nuclear reactors. As a result of the capture of one neutron, up to 1% of the atomic nuclei of the initial SHE isotope can transform to the nuclei of an isotope with mass by one unit larger. The spontaneous-fission half-life of the new isotope should be considerably shorter than that of the original nuclide, and this will facilitate its observation. Repeated irradiations in a reactor will permit production of increasingly heavier isotopes after the capture of 2, 3, 4, etc., neutrons.

This method of the synthesis of SHE isotopes, together with other ways of producing new nuclides from the island of stability, are illustrated in fig. 7. For instance, the long irradiations of targets with the fluxes of γ-rays having an intensity of 10^{15} /cm^2 sec that are accessible with modern facilities, will make it possible to produce in the reactions (γ, n) and (γ, p) the new SHE isotopes with the neutron and proton numbers one or two units less than that of the initial nucleus. Similarly, other isotopes can

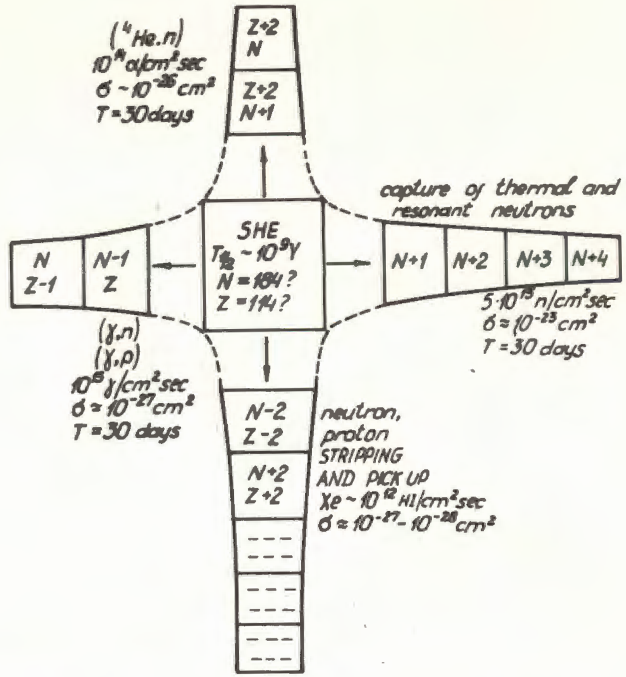
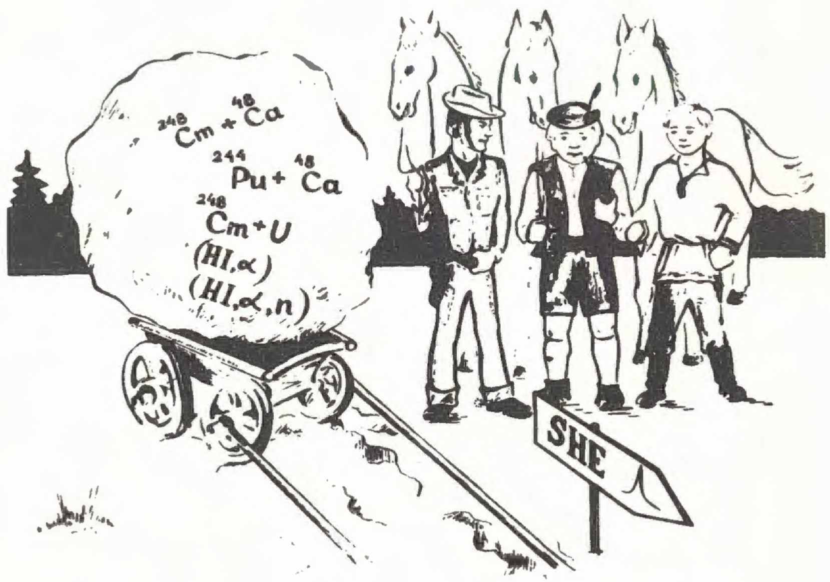


Fig. 7. A "stroll" along and across the "island of stability". An illustration of the possible ways of synthesiting new atomic nuclei by using a target made from a long-lived naturally occurring SHE. The preferable nuclear reactions, together with their cross sections, and the maximum accessible fluxes of particles and γ-rays are indicated.

be synthesized by reactions of the type (T,p), (^{3}He,p),(^{3}He,n), (^{4}He,n) and others induced by light projectiles, as well as by the pick-up and stripping reactions induced by heavy ions.

Finally, I would like to say that as any progress towards the new and unknown, the synthesis of new chemical elements is an exciting field of the knowledge of Nature by Man. This knowledge, however, is gained through tremendous intellectual efforts and large material expenses. I would like to express the hope that a reward for these endeavours will be not only new knowledge, but also the pleasure of a collaboration between the physicists and chemists of various countries (see fig. 8).



REFERENCES

1. Flerov G.N., Zvara I. JINR, D7-6013, Dubna, 1971.
2. Oganessian Yu.Ts. et al. Pisma ZhETPh, 1974, 20, p.580.
3. Ghiorso A. et al. Phys.Rev.Lett., 1974, 33, p.1490.
4. Druin V.A. et al. JINR, P7-12056, Dubna, 1978.
5. Oganessian Yu.Ts. et al. Nucl.Phys., 1976, A273, p.505.
6. Ghiorso A. Proc. R.A.Welch Found.Conf. on Chemical Research XIII: The Transuranium Elements, the Mendeleev Centennial, 1969, Houston, Texas, p.107.
7. Nitschke J.M. et al. Annual Report of Nuclear Science Division. LBL-8151, 1977-1978, p.84.
8. Flerov G.N., Druin V.A. Atomic Energy Reviews, 1970, 8, p.255.
9. Seaborg G.T., Loveland W., Morrissey D.J. Science, 1979, 203, p.711.

10. Riedel C., Nörenberg W. Z.Phys., 1979, 290, p.385.
11. Oganessian Yu.Ts. et al. Nucl.Phys., 1978, A294, p.213; Ter-Akopian G.M. et al. Yad.Fiz., 4979, 29, p.608.
12. Hulet E.K. et al. Phys.Rev.Lett., 1977, 39, p.385; Otto R.J. et al. J.Inorg.Nucl.Chem., 1978, 40, p.589.
13. Münzenberg G. et al. Nucl.Instr. and Meth., 1979, 161, p.65.
14. Gentry R.V. et al. Phys.Rev.Lett., 1976, 37, p.11.
15. Lhagvasuren D. et al. JINR, P7-11861, Dubna, 1978.
16. Shirk E.K., Price P.B. Ap.J., 1978, 220, p.719.
17. Flerov G.N., Ter-Akopian G.M. ATOMKI Közlemenyek, 1979, 21, p.93.
18. Stoughton R.W. et al. Nature Phys.Sci., 1973, 246, p.26.
19. Flerov G.N. et al. Yad.Fiz., 1977, 26, p.449.
20. Flerov G.N. et al. Z.Phys., 1979, A292, p.43.

Received by Publishing Department
on December 27 1979.