Search for transuranium elements (methods, results, and prospects)

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The article presents arguments in favor of existence of relatively stable transuranium elements with atomic numbers Z > 110.¹ A review is given of experimental attempts to detect such elements in accelerators, among materials of earthly, lunar, and meteoric origin, and in the most detail, in cosmic rays. The properties of the detectors used in these searches are discussed. A summary is presented of data on the composition of nuclei of cosmic origin in the region Z > 30, and recommendations are given further investigations.

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1. INTRODUCTION

In the last 3-5 years many physicists have gradually changed their ideas about the possible existence of relatively stable transuranium elements with charges Z > 100. One of the experimental bases and supports of the new point of view on this problem has been the existence, which became clear in 1970, of a slowing down in the rate of drop in the lifetimes of these elements with increasing Z on reaching charges of 104-105 (see Fig. 1). $[1^{-5}]$ A second and mainly theoretical argument lay in calculations of the lifetimes of transuranium elements with respect to their two dominant decay processes (spontaneous fission and α decay), made on the basis of the shell model by several groups of physicists in the period 1966-1970.^[6-11] The main result of these studies is that for a neutron number close to N = 184 and proton numbers near Z = 114 and Z = 126 there should exist an island of sharply increased nuclear stability (Figs. 1 and 2).

In spite of the high degree of uncertainty of the theoretical estimates (up to 8 orders of magnitude), they help to illustrate the tremendous effect of closed nucleon shells on nuclear stability and also to designate the main stategy of the theoretical searches. The fact is that lifetimes >10⁷ and in part down to 10⁴ years are favorable for use of cosmic-ray (including astronomi-cal), nuclear-physics, and geochemical methods, while lifetimes of 10^3-10^{-18} sec belong to the sphere of activity of heavy-ion accelerators.

The support of the shell model by theoretical physicists has a qualitative experimental basis in the region, among others, of very heavy nuclei close to uranium, such as the isotopes of Pb (Z = 82) and of element 102. In fact, from analysis of Fig. 3 we can be convinced that a departure from the relatively stable closed subshell with N = 152 by only two neutrons in either direction changes the lifetime of the isotopes of this nucleus with respect to spontaneous fission by several orders of magnitude. Even more important is the effect of magic numbers of doubly closed (proton and neutron) shells on a whole series of important properties of stable nuclei (including their production cross section) in the well investigated pre-uranium region. The principal magic numbers in the periodic table, 2, 8, 20, 50, 82, 126, determine to a significant degree the nonmonotonic behavior of such important nuclear characteristics as spin, quadrupole moment, sphericity, mass defect, and finally, the relative abundance. An experimental argument of no small importance but nevertheless inadequately convincing in favor of the existence of quasistable transuranium nuclei was obtained from a series of studies by P. H. Fowler and co-workers, which began in 1966^[13] with the emulsion method and later with dielectric detectors carried to the edge of the atmosphere by balloons. The next data, no less sensational but still less convincing, were published by Lal and co-workers^[14] in 1971; these data were obtained from space by means of natural dielectric detectors such as samples of meteoric and lunar material.

The main goal of the present article is to analyze the experimental aspects of the problem of searching for relatively stable transuranium elements with charge $Z \ge 110$, particularly as it refers to nuclei with lifetimes $>10^6$ years and has direct application to the program of study of the nuclear composition of the primary cosmic radiation.

There are good reasons to suppose that the main sources of cosmic rays are in many cases unique astrophysical objects in which the physical conditions (temperature, magnetic fields, density of nuclei, and neutron fluxes) create the possibility of synthesis of superheavy nuclei not suffering from a deficit of neutrons. Therefore, if we take into account the possibilities of direct observation of cosmic-ray sources by x-ray and

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 γ -ray astronomy, we can approach in principle a situation in which cosmic-ray and accelerator experiments (with use of the heaviest ions) will provide mutually supplementary information on the states of nuclear matter far from the current limits of the periodic table.

2. ATTEMPTS TO DETECT NUCLEI WITH $Z \ge 116$ IN ACCELERATORS

At the International Conference on Heavy-Ion Physics at Dubna in February, 1971, a number of papers were given on the first experimental attempts to take the big jump, along the staircase of ever increasing charge of the transuranium elements, calculated to reach the islands of increased stability.¹⁾

A. G. Demin et al.^[15] in 1971 carried out bombardment of ²³⁸U and ²⁴³Am targets by a beam of ⁶⁶Zn⁺¹⁰ ions with energy 390 MeV and intensity up to 5×10^9 particles per second in the U-300 accelerator at the Nuclear Reactions Laboratory, Joint Institute for Nuclear Research²⁾ (Fig. 4). For the condition that the lifetime of the fusion product exceed 10^{-8} sec, the upper limit of the cross section for production of the nucleus 122^{304} (for the case of a uranium target) was estimated



FIG. 1. Experimental data (O) and theoretical expectations (dashed curves) for the charge dependence of the lifetime of the transuranium elements. The shaded rectanges characterize the degree of uncertainty of the theoretical estimates.



FIG. 2. Theoretical dependence $[^{12}]$ on atomic number Z of the halflives with respect to spontaneous fission, α decay (α), and β decay (β). The dashed curves show the behavior of the periods for the set of all three processes. At the bottom of the figure the Z values achievable in principle by means of the fusion reactions $^{238}\text{U} + ^{131}\text{Xe}$ and $^{238}\text{U} + ^{238}\text{U}$ are indicated approximately.

FIG. 3. Lifetime with respect to spontaneous fission for element Z = 102 as a function of the number of neutrons N.



FIG. 4. Diagram of experiment on synthesis of superheavy nuclei in the Dubna accelerator. [15] 1-ion-beam collimators, 2-target, 3 and 4-fission-fragment detectors, 5-recoil-nucleus collector.

as 10^{-30} cm². G. N. Flerov^[17] noted, however, that there would be significantly greater chance of success in future experiments with the reactions U + Xe or even U + U, for only these reactions can come close to production of nuclei with the doubly closed shells Z = 114 and N = 184.

For this reason there is particular interest both in the progress in acceleration of still heavier ions in the accelerator at the Nuclear Reactions Laboratory, JINR; and in the plans also reported at the 1971 conference^[18] for startup of a new accelerator, the so-called Super Hilac at the Lawrence Radiation Laboratory in the USA, with an energy of 2.5-8.5 MeV/nucleon.

An interesting new direction in achieving collisions of heavy nuclei for the purpose of fusion is the attempt to use an ordinary proton accelerator, first undertaken by A. Marinov and others at the Rutherford Laboratory with materials bombarded at the CERN accelerator.^[19] The study consisted of detailed radiochemical analysis of a tungsten target which was placed for an extended period in an intense beam of 24-BeV protons with the purpose of looking in it for superheavy analogs of mercury with charge Z = 112. The hope was that, even if very rarely, an energetic proton could knock out of the tungsten nucleus some very heavy recoil nucleus with energy >1 BeV sufficient to overcome the Coulomb barrier of another W nucleus, with subsequent fusion of these nuclei. The positive indications of spontaneous fission of atoms with the properties of mercury obtained in this study (the effect amounted to 4.0 ± 0.3 per day) stimulated the planning of special experiments in this direction.

However, control experiments carried out by G. N. Flerov's group^[20] at the Serpukhov accelerator on ejection of rather heavy and energetic recoil nuclei (suitable for accomplishing a subsequent fusion reaction) with an appreciable cross section (greater than 10^{-30} cm²) led to a negative result, and this result alone already places in serious doubt the promise of this direction as a whole.

Still more convincing are attempts to make a direct check of Marinov's experiments, based on the results of parallel and independent work of three groups of physicists at the proton accelerators in the USA and at CERN.

In the work of R. Esterlund et al.^[21], uranium nuclei were bombarded by 11.5-BeV protons to study the possibility of producing already known transuranium elements in a two-stage reaction, for example, of the type

²³⁸U (p, ²⁰O) (²⁰O, 2n) ²⁵⁶Fm,

with ejection of ²⁰O nuclei in the first step and fusion

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of ²⁰O nuclei with ²³⁸U in the second step. Careful searches for ²⁵⁴ Fm and ²⁴⁶ Fm by detection of protons, γ rays, α particles, and fission fragments led to estimates of the upper limit of the cross section for this reaction $\sim 2 \times 10^{-32}$ cm² and $\sim 4 \times 10^{-34}$ cm², respectively.

In the work of J. P. Unik et al.^[22], samples of tungsten (of weight 5 and 25 g) and later also uranium (of weight 1 kg and 765 g) were bombarded by 12- and 28-BeV protons with a total bombardment dose of $\sim 10^{17}$ protons (Marinov used a target of 33 g of W and a dose of 7×10^{17} protons). In the experiments with uranium, ten transuranium nuclides up to ²⁴⁸Cf were detected by α spectrometry (the latter in a quantity of $\sim 10^{-36}$ cm²), but for the isotopes of Es, Fm, and Md, upper-limit cross sections at the level $\sim 10^{-36}$ cm² were already obtained.

The samples of W and U were subjected also to radiochemical analysis with detection of spontaneous fission fragments of possible products in the interval Z = 107-116. In all cases only upper-limit cross sections at the level $10^{-39}-10^{-40}$ cm² were found, even under conditions where, according to Marinov's data, an effect 5.0 ± 0.4 (in 115 days of measurement) should be observed.

A basic result was obtained by L. Westgaard et al.^[23] by bombardment of a uranium rod of thickness ~190 g/cm² by a flux of ~4 \times 10¹⁷ protons with energy 24 BeV. By chemical separation of the products with subsequent detection of α particles and fission fragments, it was established that there is a sharp drop in the production cross sections of the transuranium elements, beginning at 3×10^{-29} cm² for ²³⁶ Pu and extend-ing to 4×10^{-37} cm² for ²⁴⁸Cf (fragments of the type ¹⁴C are necessary for obtaining the latter). A rough estimate of the quantity and energy spectrum of ⁵⁶Ca fragments shows that their fusion with U nuclei could lead to formation of the isotope $^{200}112$ with a resulting cross section no greater than 2×10^{-40} cm². From comparison with the experimental data for ²⁴⁸Cf, for which the same method of estimation gives a clearly exaggerated value $\sigma \sim 3 \times 10^{-35}$ cm², it is evident that the estimates of the expected production cross sections for element 112 are too optimistic (not to mention that for the actual conditions of observation of the decay products an optimum lifetime $T_{1/2} \simeq 1$ month is assumed).

3. ATTEMPTS TO DETECT LONG-LIVED TRANSURANIUM ELEMENTS UNDER NATURAL CONDITIONS (Earth, Moon, Meteorites)

No particular doubt is raised by the fact that neither in the Sun or in any other bodies of the solar system are there at the present time conditions under which synthesis of transuranium elements could occur. The use of uranium itself and its decay products for dating the Earth (and in the near future other planets) is based on the assumption of a relict origin of uranium.

We note in passing that in the Earth equilibrium generally exists between the short-lived elements in the interval Z = 84-89 and the much longer-lived parents of the radioactive series—uranium (Z = 92), thorium (Z = 90), and actinium (Z = 89). The relative longevity of the latter (of the order 10^9-10^{10} years) is at once one of the manifestations of shell effects in islands of relative stability of very heavy nuclei. This means that searches for any relict elements with Z > 92 and lifetimes less than 10^8 years is actually doomed to failure even with the most sensitive detection methods. However, this does not exclude the possibility and necessity of searching for shorter-lived transuranium elements which could be carried by cosmic rays either to the Earth and then gradually (in millions of years) settle in the strata of the atmosphere and oceans and be collected in the corresponding deposits, or to the surface layers of the Moon and meteorites, which do not have the protection of an atmosphere.

We warn the reader beforehand that, in spite of a wide and ever increasing circle of investigations, there has not been a single completely reliable positive result obtained in this field. However, the main experimental indications deserve attention already, since they permit in principle a comparison (although a rough one) with the results of direct analysis of the chemical composition of cosmic rays.

The main groups of experimental facts (for more detail consult the earlier review of G. N. Flerov et al.^[24]) are as follows:

1) Indications of the existence either of unknown α -active elements with anomalously long-range α particles, or with chemical properties similar to osmium with the very modest energy $E_{\alpha} \approx 4.6$ MeV.^{[25-27]3)}

In this connection we note that short-lived artificially produced transuranium elements emit α particles with energies up to 9–9.5 MeV (the longest-range α particles from ThC^{'212} have an energy ~8.8 MeV^[28] and those from the long-lived U²³⁸ 4.2 MeV).

2) Indications [29, 30] of the existence of unknown spontaneously fissile impurities in samples of various origins in which elements such as Pb, W, Tl, Bi, and Hg have been in contact for a long time (tens and hundreds of years) with glass-a natural detector of spontaneous fission fragments; here estimates of the fission lifetime of the order of $10^{20} - 10^{21}$ years have been obtained (with concentrations of 10^{-12} - 10^{-13} g/g); control irradiations by slow neutrons from a reactor have made it possible to prove that the responsibility for the observed effect can in no way be placed on uranium impurities; subsequently, however, ^[31] these results have been reconsidered by the authors themselves, who do not exclude the possibility of a background effect due to fission of Pb nuclei by fast cosmicray nucleons.

3) Similar indications^[32] of unknown spontaneously fissile elements (other than uranium) have been obtained from observations on iron-manganese concretions taken from the ocean floor and with an age $\sim 2 \times 10^6$ years, which are substantially enriched in elements such as Pb, Tl, Hg, W, and others and which contain natural fission-fragment detectors in the form of pieces of feldspar; the density of fission-fragment tracks in various cases amounted to 7 to 120 tracks/mm².

4) An indication^[33] of the existence of elements which are analogs of Pt, Au, Hg, Tl, Pb, and Bi with anomalously long-range spontaneous-fission fragments, obtained by Von Gunten et al. by means of an original apparatus—a so-called spinner (a variation of a bubble chamber in which the super-saturation of the liquid necessary for formation of gas bubbles in particle

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tracks is produced by rapid rotation of a vessel containing a solution of the heavy element compounds being studied).

5) An indication obtained by a group of Indian physicists^[14] of the existance of elements with anomalously long-range spontaneous fission fragments in specially etched samples (pyroxene and feldspar) of meteoric and lunar origin: while Fe nuclei should produce in these samples tracks of length 10-13 microns and uranium fission fragments 16.5-18.5 microns, an appreciable fraction of tracks 20-25 microns long is actually also observed. However, as has been shown by detailed studies carried out by R. L. Fleischer et al. in the $\text{USA}^{[\,34\,]}$ on the optics of the television camera of Surveyor III (which was on the Moon about 2.5 years and was brought back to Earth by the crew of Apollo 12), in studies of this type it is important to take into account the background from high-energy fission of lead impurities by cosmic rays (in this case it amounted to \sim 10 tracks per cm²).

4. METHODS OF STUDYING THE HEAVY NUCLEAR COMPONENT OF COSMIC RAYS

a) <u>Nuclear emulsions</u>. As has been noted above, P. H. Fowler et al.^[35] obtained in emulsions the first important indication that in the flux of almost undistorted primary cosmic radiation near the edge of the atmosphere it is possible to observe, although extremely rarely, nuclei of the transuranium elements. It should be kept in mind that, since a detailed microscopic picture of a phenomenon is obtained, even single observations with emulsions can in principle be highly convincing and can exclude the possibility of imitating background processes which are so dangerous in the case, for example, of studying very rare events by electronic methods.

To obtain such a fundamental and simple characteristic of a particle as the electric charge in the case of fast and rather heavy nuclei (Z > 20), there exists a now well known method based on microphotometry of the profile of the track, i.e., the distribution of the fraction of light absorbed in the emulsion as a function of the distance from the particle trajectory and consequently from the axis of its track. The accuracy of such measurements, as illustrated in part in the fragments of a typical iron-nucleus track (Fig. 5), is due to the following factors:

1) The length of the measurable portion;

2) the accuracy in determination of the nuclear velocity;

3) the possibility of making calibration measurements under the most comparable conditions possible on nuclei with known charge;

4) the accuracy and reliability of determining the charge dependence of one of the three possible photometric characteristics of the track: a) the absorption of light a given distance from the track axis, b) an arbitrary average width of the track, and c) the integrated amplitude of absorption of light passing through a slit bounding the track image;

5) the influence of edge effects (at the surface of the emulsion layer);

6) the general background from unrelated particles and its fluctuations.



FIG. 5. Track of stopped iron nucleus in different portions of its range, recorded in type NIKFI R-2 emulsion of thickness 400 μ . The distances from the center of each portion to the point of complete stopping and the velocity of the nucleus are respectively: $R_d = 0$; $R_b = 150 \mu$ ($\beta_b = 0.20$); $R_c = 500 \mu$ ($\beta_c = 0.28$); $R_a = 500 \mu$ ($\beta_a = 0.50$).

We will discuss the first four factors in more detail.

1. The transverse structure (profile) of the track has a sharply inhomogeneous nature along the track; this is due to the fact that the track is formed by the ionizing action on sensitive AgBr crystals not of the primary nucleus itself but only of the δ electrons ejected from the atoms surrounding its trajectory. To estimate the role of fluctuations of this process in the photographic characteristics of the track, we will point out that, for example, the track widths of an iron nucleus averaged over segments 15 microns long far from the end of the range (Figs. 5a and c) are subject to a statistical spread of about 10% and consequently, for an accuracy of 2% in width measurement, measurements on a total length of the order of 400 microns are required.

2. In balloon flights of emulsions to altitudes of the order of 35-40 km and with special measures to recognize particles which entered the emulsion in the rising and falling portions,⁴⁾ the experiment should reveal a rather sharp lower limit to the velocity of the nuclei detected, as the result of the threshold action of the Earth's magnetic field. For example, in Fowler's experiments^[35] for vertically incident nuclei $\beta_{min} = 0.92$ (geomagnetic latitude ~42°) and consequently the maxi-

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mum possible spread in the estimates of the measured charge should be, it would appear, $(\Delta Z)_{max} = \pm 4\%$.⁵⁾ However, under actual experimental conditions the situation turned out to be appreciably poorer: As shown by Fig. 8, which is taken from ref. 35, for nuclei of the iron group the track density D at a distance 10 microns from the axis exceeds the minimum value D_{min} (which corresponds to $\beta = 0.95$) by no less than 20% (i.e., $\Delta Z/Z \ge 10\%$) in 1% of the events, while with the same energy spectrum for nuclei with Z = 96 such a large relative increase in density can occur in ~20% of the events. Thus, as the value of the nuclear charge Z increases, the accuracy and reliability of measurement of the charge Z due to the spread in velocities of the nuclei being studied deteriorate substantially.

When emulsions are exposed in outer space, as was done, for example, in the experiments of N. S. Ivanova et al.^[36], where the magnetic threshold does not have a fixed value, it is possible to determine β_{\min} from the condition of absence of a noticeable change in the photometric characteristics of a track in a sufficient length. In particular, for a length of 10 cm in emulsion it is possible to determine that the minimum energy is $\approx 1 \text{ BeV/nucleon}$, and hence $\beta_{\min} = 0.86$ and $(\Delta Z)_{\max} = \pm 6\%$.

Finally, a third method, suitable only for measurement of nuclei which stop completely inside the apparatus, consists of establishing the relation between the range R and the velocity β , which in turn depends, although weakly, on the desired charge value Z. This dependence gives rise to the method proposed by Zhdanov et al.^[37] for measuring Z by studying the longitudinal structure of the track, in particular from measurement of the maximum track width d_{max} and the corresponding residual range R_{max} of the nucleus.

Figure 6 gives the results of a study of the longitudinal structure of the track of a nucleus of the Fe group (from cosmic ray experiments), presented in the form of two variants of the dependence of the track width d on the residual range R. One of the variants (o) corresponds to the ordinary photometric measurement of the average width (i.e., smoothing the fluctuations of the width in the cell measured), and the other (x) to visual identification of the so-called core of the track formed by the significantly more numerous, and consequently less subject to fluctuations (in flux), short-range δ electrons. It turned out that a) measurements in both variants are satisfactorily approximated by a simple analytical function of the form

$$Y = \frac{AX}{1 + X^{\alpha}} , \qquad (1)$$

where $Y = d/d_{max}$, $X = R/R(d_{max})$, $\alpha = 1.5-1.7$.

The known law for approximation of the function d(R) permits a substantial increase in the accuracy of measurement of the maximum width d_{max} , without limitation to the portion directly adjacent to the maximum.

b) The dependence of the average width on the range agrees satisfactorily with the corresponding theoretical curve of R. $Katz^{[38]}$ and this permits the hope that a purely theoretical dependence $d_{max}(Z)$ can be a satisfactory approximation for the experimental estimation of the value of Z from measurements of d_{max} .

3. The only possibility up to the present time of

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FIG. 6. Experimental [³⁷] and theoretical [³⁸] dependence of track width on residual range R for iron group nuclei: x-measurement of track core, O-measurement of average width, solid curve-calculation of track widths determined from the condition $\overline{E} = E_0$, [³⁸] for Z = 22 and Z = 30 (E is the average rate of energy loss, \overline{E}_0 is the threshold for development of an AgBr grain).



FIG. 7. Summary of existing data on chemical composition of cosmic rays (for relativistic velocities) and of the material of various astrophysical objects. As the ordinate, in addition to arbitrarily normalized relative abundances, we have plotted a scale of absolute fluxes of cosmic rays with energy $E_{kin} > 1$ BeV/nucleon; the abscissa represents constant logarithmic intervals of Z values ($\Delta lgZ = 0.1$), in which the experimental data are combined. 1–cosmic rays, [³⁵] 2–cosmic rays, [³⁶] 3–cosmic rays, [³⁹] 4–cosmic rays, [⁴⁰] 5–solar-system matter, [⁴¹] 6–Earth's core, [⁴²] 7–sun's atmosphere. [⁴²]

experimentally calibrating emulsions for heavy nuclei of rather high energy has involved use of nuclei of the iron group. In particular, analysis of the existing data on the chemical composition of cosmic rays (Fig. 7) shows that in a background of a general and regular drop in abundance of elements with increasing atomic number, occurring roughly according to a Z^{-8} law, the iron group nuclei form a distinct peak with a particularly sharp drop on the high-Z side. This peak appears particularly well in Fig. 8, where the ordinate is the particle flux plotted on a linear, not logarithmic, scale and the abscissa, as a measure of Z, is the quantity D_{10}^{80} usually used by Fowler, i.e., the logarithm of the light absorption in the particle track at a distance of 10 microns from the axis with subtraction as a background of the absorption at a distance of 80 microns from the axis.

From Fig. 8 it is possible to estimate what statistics of calibration measurements are needed to provide a given calibration accuracy. Here it is necessary to bear in mind the observance of comparable conditions, of which the principal ones are, first, the closeness of the slope angles of the measured track to the emulsion plane, second, the constancy of the sensitivity, thickness, and development quality, and, finally, the absence of regression of the latent image and loss of sensitivity of the emulsion, which can occur during extended exposure of the emulsion.

Unfortunately, existing heavy-ion accelerators do not permit use for calibration of relativistic nuclei heavier than nitrogen, but in future years this possibil-

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FIG. 8. Results of photometric measurements of cosmic particles of the iron group according to Fowler. [³⁵]

ity must be kept in mind as a result of the rapid progress in the technology of heavy-ion acceleration.

4. Purely empirical direct calibration of the photometric characteristics of a track as a function of the nuclear charge over a wide range of Z is not possible at the present time even for a constant (but not too small) velocity β , since it cannot be done in accelerators.⁶⁾ Therefore it is necessary to use either purely theoretical methods or a combination of theory and empirical calibration of emulsions. In the first case, as was first done by Fowler et al., $^{[35]}$ the spatial distribution of the energy release is calculated as a function of the velocity and effective nuclear charge Zeff (i.e., the charge obtained with inclusion of the partial cladding of the nucleus by atomic electrons, which is important only for $\beta \leq 0.3$), and then, from the similarity of the distribution obtained to the spatial behavior of the density of real tracks for constant β , the conclusion is drawn that emulsion (within certain limits of the distance r) is a linear detector of the energy dissipated.

The main result of the calculations $[^{35}]$ is that the total energy release per unit volume at a distance r from the track axis is

$$\left(\frac{dE}{dx}\right)_{\text{tot}} = 2.4 \frac{Z_{\text{eff}}^2}{\beta^2 r^2} \times \left[1 + 0.012 \ln\left(\frac{\beta^2}{1 - \beta^2}\right)\right] eV/\mu^3,$$
(2)

where the second (unimportant) term in the brackets is due to so-called soft collisions involving excitation and subsequent de-excitation of the atoms of the medium, and the first (principal) term is due to the direct ejection (hard collisions) and subsequent ionization loss of δ electrons.

Detailed numerical calculations of the spatial dependence of the energy release E for emulsion have been made by R. Katz.^[38] These calculations,⁷⁾ in addition to the parameters Z and β , involved two parameters characterizing any radiation detector: these are the size of the sensitive element a_0 (the radius of an AgBr grain in the case of emulsion) and the critical (activation) energy dissipation in this element E_0 (in the case of normal emulsions of type Ilford G-5 and K-5, values of E_0 in the range 10-35 keV/ μ^3 were assumed). It is important that at distances $R_{max} \gg r \gg a_0$, the energy dissipated is given by $E \simeq r^{-2}$, i.e., is expressed by an extremely simple universal function (R_{max} is the maximum range of the δ electrons).

The calculation of Fowler et al.^[35] also showed that

for relativistic velocities of the nuclei a substantial part of the energy dissipation is determined by δ electrons with energy $W_{\delta} > 50$ keV; for small velocities the kinematic limitation of the δ -electron range becomes important (i.e., the condition $r \ll R_{max}$ is violated), and in this case it is necessary to add, on the right-hand side of Eq. (2), a cutoff factor of the type

$$f(r) = \exp(-\frac{10r^2}{R_{\max}^2}).$$

(3)

In just this velocity region it is necessary to take into account also another, though less important, factor, which has been studied in detail particularly by V. B. Semikoz^[43,44] This is the decrease in effective nuclear charge as the velocity decreases to values comparable with the velocities of the atomic electrons, that is, to $\beta = 0.3-0.4$ (this will be discussed in more detail below; see page 650).

Experimental verification^[38] (Fig. 9) of the results obtained in the range of distances $r = 5-50 \mu$ and charges Z = 26-100 (for relativistic nuclei) showed that the quadratic dependence of the energy dissipation on r predicted by Eq. (2) leads to a weaker dependence on r for the optical track densities (up to D = 3, where D is the natural logarithm of the light absorption).

As can be seen from comparison of the experimental data with the calculations of Katz for nuclei with charge $Z \sim 100$ in the range of distances $r = 10-40 \mu$, the effect of the deviation from linearity as the result of saturation of the density is already noticeable for small r and can lead to exaggeration of the true charge value by an amount $\Delta Z \sim 10\%$. One of the causes of non-linearity involves the threshold nature of the action of the sensitive element of the emulsion. As shown by Katz, as the result only of fluctuations of the energy release in the track volume, it is necessary to discuss in the general case only the probability of activation of a sensitive element of the detector—a microcrystal of AgBr. For a Poisson distribution of the fluctuations, this probability has the form

$$P = 1 - \exp\left(-\overline{E}/E_0\right),\tag{4}$$

where \overline{E} is the average energy dissipation per unit volume of the track and E_0 is the critical energy value necessary for formation of a development center in the AgBr grain.

Two very important properties of emulsion as a detector of multiply charged particles follow from Eq. (4). First, the track width d is determined by the distance from its axis at which the quantity $\overline{E} \ge E_0$ and reaches a definite, sufficiently large value; since $\overline{E} \sim Z^2/r^2$,

FIG. 9. Results of photometry of the profiles of tracks of relativistic nuclei according to Fowler, and comparison with the calculations of Katz. [³⁸]



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the width $d \sim Z$ (the method of measuring Z from the track width). Second, if $\overline{E} \ll E_0$ even on the axis of the track (i.e., for $r \sim a_0$), then everywhere $P \sim \overline{E} \sim Z^2$, the track becomes discontinuous, and the total number of developed grains per unit track length also is $\sim Z^2$ (the method of measuring Z by grain counting).

In addition, even for the condition $\overline{E} \ll E_0$, where the function P(E) becomes linear, the problem is substantially complicated by the inhomogeneity of the AgBr crystals in sensitivity (i.e., the finite spread in the value of E_0). Therefore an important means, but not the only means, of extending the region of linearity of the emulsion is reduction of the quantity \overline{E}/E_0 as the result of reduction of the size of the AgBr single crystals, which is extremely important also for reduction of the general background under conditions of extended exposure.

In addition to the calculations of Fowler and Katz, we can also use the detailed calculations of V. V. Varyukhin et al.^[36], who obtained data on the profiles of nuclear tracks over a wide range of velocities and charges (Figs. 10-12).

The general course of the discussions of the authors $[^{44]}$ is as follows. First the profile of the energy loss E(r) is calculated for iron nuclei of various velocities (Fig. 10). Then the density profiles B(r) of the tracks of the same nuclei are measured experimentally by photometry, which provides the possibility of unambiguously comparing the energy deposition E and blackening B for each specific type of emulsion and method of development. This comparison is equivalent to experimental determination of the activation threshold E_0 in the Katz model.

Finally, the energy depositions E(R) are calculated for a large set of charges and velocities, and these are converted to the blackening B(R). In this way the rangedependence has been obtained, in particular, for the blackening due to nuclei with Z = 26 and Z = 50 at $r = 3.3 \mu$ (Fig. 11); comparison of these functions permits in principle information to be obtained also on the range of linearity of a given emulsion.

If the track boundary is defined by some arbitrary condition (B = const), it would be possible to calculate by the same means the dependence of the track width on Z for β = const (Fig. 12). In this way, in particular, the expected photometric characteristics have been obtained of tracks of fast transuranium nuclei from the corresponding characteristics of slow Fe nuclei. From subsequent measurements of heavier stopped nuclei, a repeated procedure of this type permits in principle the



FIG. 10. Theoretical rate of energy deposition as a function of the distance from the trajectory of Fe nuclei of various velocities β . [³⁶]

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FIG. 12. Theoretical dependence of track width on the charge of a relativistic nucleus. $[^{36}]$

calibration of emulsions for slow transuranium nuclei.

Considerable attention has been devoted^[37] to experimental verification of the linearity of various emulsions by Zhdanov et al. who were unable to use charges higher than Z = 30; however, investigations have been made of stopping nuclei whose ionization is already close to that of relativistic nuclei of transuranium elements.

In Fig. 13 we have shown for the case of Ne nuclei (Z = 10) with energy 10 MeV/nucleon the longitudinal track structure (in region to the left of the maximum width) for four emulsions of different sensitivities. The sensitivity was in turn measured with an arbitrary limiting velocity β_{max} of singly charged particles (in particular, electrons), which are still visible in this emulsion as easily distinguishable tracks. From the point of view of the theory (particularly that of Katz^[38]) the track boundary is defined in such a way that the average energy dissipation, which is proportional to Z^2r^{-2} , corresponds on the average to the threshold sensitivity E_0 , which in turn is proportional to β_{\max}^{-2} . Hence it follows that the track width (other conditions being equal) should be proportional to β_{max} , and this is roughly in agreement with the data of Fig. 13.⁸

In spite of all the attractive features of using lowsensitivity emulsions as threshold detectors of very heavy nuclei which have a low sensitivity to the general cosmic-ray background, they also have negative aspects associated, first, with the reduced stability of the sensitivity and, second, with the lower relative accuracy in



FIG. 13. Dependence of track width of Ne nucleus as a function of residual range for emulsions of various sensitivity (in parentheses are shown the maximum energy of electron detectable by a given emulsion).

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measurement of the track width. On the other hand, from the point of view of Katz's theory, $[^{38]}$ for a detector threshold E_0 exceeding the average ionization loss even on the track axis, the track acquires an interrupted structure; in this case the experimenter uses grain counting, and the result of photometry in the ideal case turns out to be proportional to Z^2 , and not Z as in the case of a continuous track, which provides hope for an increase in the resolution of this method in charge.

In Figs. 14a and b we have shown how the longitudinal track structure of various nuclei is affected by dilution of the emulsion gelatin. It is evident that with increasing charge Z there is an increasing influence of nonlinear effects caused both by local depletion of the developer (so-called starved development) and the mutual displacement of developed AgBr grains from the central zone of the track as the result of their significant enlargement in the development process.

A very interesting and useful addition to ordinary photographic technique is the method developed recently by L. Pinsky et al. in the USA^[45] of detecting Cerenkov radiation of fast very heavy nuclei. The main elements of this detector (Fig. 15) are: a layer of transparent radiator of Cerenkov light (gelatin or a special gel in a polymer base), under which is placed (on an appropriate substrate) a thin layer (12–15 μ) of high-sensitivity Kodak emulsion which provides a visible blackening for a photon flux density of the order of 1–2 per square micron.

Obviously the effective radius of the spot obtained at the point where the trajectory of the nucleus intersects the emulsion depends both on the velocity β and the charge Z. A calibration experiment carried out in balloons at 30 km altitude with a set of other detectors with an area-time factor of ~1500 m²-h demonstrated the possibility of measuring nuclear velocity with an accuracy $\Delta\beta = \pm 0.02$ for thresholds Z_{min} = 60 and $\beta_{min} = 0.68$. It is proposed in later work to raise the charge threshold to Z_{min} = 50.

Attempts are being made^[46] to detect multiply charged particles also by detectors of a new type consisting of AgCl single crystals with an admixture of cadmium. On passage of a strongly ionizing particle, there occurs along the track a precipitation of the metallic silver phase which can be stabilized by simultaneous illumination with yellow light. After the entire exposure, tracks visible in a microscope can be obtained by irradiation with ultraviolet. In view of the



FIG. 14. Track widths of (a) Ne nuclei and (b) Zn nuclei in emulsions with various degrees of dilution with a normal size (0.27μ) of undeveloped AgBr grains. a) Dilution $\times 1$ (1), $\times 2$ (2), $\times 3$ (3), $\times 4$ (4); b) dilution $\times 1$ (4), $\times 2$ (3), $\times 4$ (1), $\times 8$ (2).

FIG. 15. Diagram of Cerenkov detector for heavy nuclei. [⁴⁵] 1–Substrates, 2–antihalation layer, 3–Kodak EK-2485 emulsion, 4–Cerenkov radiator.



limited dimensions and still inadequate reproducibility of the results, this type of detector is clearly inferior to nuclear emulsions, although it has the advantage of continuously erasing the background of weakly ionizing particles or particles not accompanied by preliminary irradiation.

b) <u>Solid track detectors (SSTD</u>). The principle of action of dielectric detectors of both the plastic and crystalline types is that the radiation effect of the passing particle on the element of the medium (in the form of a large organic molecule in the first case and a crystal in the second) leads either to breaking of chemical bonds or to displacement of atoms and as a consequence to an increase in the solubility of the detector material in suitable reagents (usually caustic alkalis) in the immediate vicinity of the particle trajectory. The effect turns out to be substantial (and moreover irreversible) only in the case where several (n) bonds in the molecule are broken simultaneously or the concentration of atomic defects is high.

The primary effect of passage of a strongly ionizing particle, in contrast to photographic emulsions, extends to very small distances (10-100 Å) from the trajectory, as is shown by direct observations by means of an electron microscope. One of the possible mechanisms of this effect in crystals is the so-called ion spike^[47] due to the instantaneous ejection of a large number of electrons from a narrow channel near the track axis.

The substantial difference in the mechanisms of particle track formation in emulsions and solid-state track detectors results, in particular, in the fact that measurement of the diameter of the channel left after etching cannot serve as a measure of the ionizing ability of the particle, particularly because the formation of the latent image cannot be reduced to the action of δ electrons.

One of the arguments in favor of the last statement is the detection^[48] of tracks of particles with energy substantially less than 0.5 MeV/nucleon and even as low as 0.2 keV/nucleon where there can be no possibility of δ electrons.

As stated by Katz,^[38] in contrast to emulsions, dielectric detectors can be considered as a set of targets requiring multiple hits, where the probability of striking each target is given by the expression

$$P = (1 - e^{-\tilde{E}/E_0})^n;$$
 (4a)

here $\overline{E}(Z, \beta, r)$ is the average energy deposition at a distance r from the particle trajectory; E_0 and n are parameters characteristic of the detector which determine the activation energy of the sensitive element and the number of elements forming one target. A value n > 1 has the result that, for the same fluctuations, particles with ionization $\overline{E} < E_0$ now give a substantially smaller contribution to the background than in the case of emulsion, even for comparable E_0 values.

There is, however, another point of view^[49] accord-

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ing to which, at least for plastic detectors, there is in general no activation threshold, and the possibility of observing tracks in the form of etched channels is determined by the increased velocity of penetration of the etchant through the chain of local defects left by the particle.

In addition, thermal motion, generally speaking, is less dangerous as the cause of regression of the latent image of the particle track with time for dielectric detectors. Furthermore, the stability of the properties of detectors of a given type (the degree of spread of the threshold values as a function of the manufacturing technology and the effect of the surrounding medium) depends directly on the thermal stability, i.e., the temperature dependence of the probability of irreversible annealing of the latent image of the track.

As has been shown, in particular, by Khan and Durrani^[50], the efficiency for detection of a given particle (in this case Cf^{252} fission fragments) drops sharply as the temperature approaches some critical value T_C ; this drop in efficiency occurs more rapidly than the expected probability of deactivation of the latent image of the track by thermal motion, which should be expressed by a factor of the form $exp(-E_0/kT)$ and should begin to play an appreciable role on reaching a temperature $T_C ~ E_0/k$. If we take as a characteristic annealing temperature the value of T_C at which the fragment-detection efficiency drops by a factor of 10, the following pattern is obtained for various detectors:

Detector	Plastics (pol	ycarbonates)	Crystals (glasses)		
Detector	Lexan	Macrofoil	Sodium glass	Tektite	
<i>T</i> _c , ℃	~ 175°	~ 165°	~ 300°	~ 475°	

For a given temperature and an isotropic angular distribution of detected particles, the efficiency f is determined by the ratio of the etching rates V_g for the detector material as a whole and V_t along the track axis, which determines the critical angle θ_c for entrance of a particle which will still leave a visible channel (since sin $\theta_c = V_g / V_t$ and $f = 1 - \sin \theta_c$).

The wide range of V_g/V_t values for different detectors is evident from the following table (we are considering detection of fission fragments):

Detector	v _g /v _t	Øc	Detector	v _g /v _t	θε
Sodium glass Obsidian Tektite Quartz	0.58 0.44 0.435 0,125	35,5° 26° 25°45′ 7°	Mica Macrofoil Lexan	0.08 0.052 0.044	4.5° 3° 2.5°

A quantitative measure of the radiation effect is usually the longitudinal etching rate Vt along the track axis (Fig. 16), which in practice lies in the range $0.1-100 \mu/h$. The velocity Vt is a unique but generally nonlinear function (of the form J^m , where $m \approx 2$) of the specific radiation effect^[40]

$$V_{t} = a \frac{Z_{\text{eff}}^{*}}{\beta^{2}} \left[\ln \left(\frac{\beta^{2}}{1-\beta^{2}} \right) + K - \beta^{2} - \delta \left(\beta \right) \right].$$
 (5)

While $\delta(\beta)$ is usually a small relativistic correction ($\delta = 0$ for $\beta < 0.8$) to the polarization effects in a dense medium, K is a very important parameter (it is usually in the range 10-60) which must be determined em-

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FIG. 16. Arrangement for measuring etching rates of dielectric detectors. V_t -along the track of the ionizing particle; V_g -detector material as a whole.

pirically for each combination of detector and etching agent.

With good reproducibility of the experimental conditions (in accelerators) it is possible to distinguish even the isotopes B^{10} and B^{11} or Ar^{36} and Ar^{40} by the rate of etching of plastic detectors. In cosmic-ray experiments it is possible to have a spread in Z values measured by the velocity V_t in different layers of the detector (but in the track of same heavy nucleus) $\Delta Z/Z \sim 1\%$.

In Figs. 17a we have shown the radiation effects J, taken from Price et al.^[40], as a function of energy for nuclei over a wide range of Z, and also the threshold values J_{\min} for a number of the most frequently used detectors (horizontal lines).

In order to have available in each specific detector dependences suitable for identification of particles, it is necessary to make careful calibration measurements in accelerators, taking into account a dependence $Z_{eff}(Z, \beta)$ of the type^[51]

$$Z_{\rm eff} = Z \left[1 - \exp\left(-\frac{125\beta}{Z^{1/3}}\right) \right]. \tag{6a}$$

However, as has been shown by V. B. Semikov,^[43] over a wide range of charges $(8 \le Z \le 100)$ better accuracy can be obtained with the equation

$$Z_{\rm eff} = Z \left[1 - \exp\left(-\frac{137\beta}{Z^{0.83}}\right) \right]. \tag{6b}$$

The calibration procedure usually consists either of measuring the longitudinal rate V_t (with inclusion of a correction for the rate of etching of a smooth surface V_g ; see Fig. 16), or in determination of the time necessary to obtain an etched channel of a given length (say, 10 microns).

Because of the lack of strict linearity in the kinetics of etching, these two calibration operations are in no way equivalent. Having measured the residual range R, converting by means of tables from ranges to velocities,



FIG. 17. a) Dependence of radiation effect J on velocity (lower horizontal scale) and energy (upper scale) of various nuclei; b) dependence of etching rate V on residual range R (calibration points and solid curves of the form of Eq. (5) are given for K-62).

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and using Eqs. (5) and (6), we can then either determine the constants K and m by a fit or, as was done by Price and Fleischer^[40], for example, for a Lexan detector (Fig. 17b), make direct calibration curves of the velocity Vt as a function of the range R for various Z.

It can be seen from Fig. 17a that in a given detector each nucleus for which the curve $J(\beta)$ lies above the threshold level J_{min} leaves a track of a definite length for a given method of etching. Therefore, in addition to accurate measurement of the etching rate V_t , identification of nuclei can be accomplished in principle by the simple measurement of the lengths of tracks.

The resolution of this type of measurement increases as the radiation effect approaches its threshold value (if the latter is sufficiently stable). Thus, from Fig. 17a it follows that the visible track lengths L of Cm nuclei (Z = 96) or U nuclei (Z = 92) in mica are distinguished by only ~20%, while in meteoritic samples (in particular, in olivine) the ranges of Zn nuclei (Z = 30) and Fe nuclei (Z = 26) should already differ by a factor of 2.5.

However, under the actual conditions of a cosmicray experiment it is necessary to take into account the effect of a number of other factors, in particular:

1) aging of the track with time;

2) the effect of heating;

3) the effect of the surrounding medium (gas composition and humidity) on the value of V_t ;

4) the effect of ultraviolet and ionizing radiation;

5) change of the etching rate V_t with the etching method and the concentration of the detector material in the solution.

We will consider in somewhat more detail the role of each of the factors enumerated.

1. Track aging, characterized by change of the velocity V_t or total length L as a function of the time elapsed between bombardment and etching, is a non-monotonic function of the time t. As in emulsions, the detector sensitivity first (for 1–2 months) rises and then begins to fall. A substantial difference, in particular, has been noted^[52] between the characteristics of old tracks and those obtained in accelerators of the same heavy ions (in particular, iron) in minerals of cosmic origin. It is unknown to what degree this difference is due to the effect of factors 2–4 listed above.

2. Several experimental studies^[53-55] have been made of the effect of additional thermal processing (before and after bombardment) of solid-state track detectors (both plastic and crystalline) on the etching rate V_t and the particle-track detection efficiency. In the case of plastic detectors (cellulose nitrate and acetate) it has been observed, in particular, that preliminary heating of the detector before bombardment to $T \sim 100^{\circ}$ C increases the stability and sensitivity for α -particle detection, while the same heating after bombardment reduces the sensitivity. These effects play an important role because the transition region from amorphous to polycrystalline structure of the organic polymer is reached.

For crystalline detectors, (in particular, mica) preliminary annealing is used to reduce the background, which is due, in particular, to spontaneous fission fragments from the U^{235} impurity.

3. Among the factors associated with the chemical composition of the surrounding medium, the greatest effect has been observed from ozone, ^[58] and the humidity of air also has a certain effect. It has been shown that ozone appreciably increases the sensitivity of plastic detectors, and therefore it can be used to accelerate the natural process of saturation of the sensitivity with time.

4. It was observed by Nicalae^[57] as long ago as 1968 that ultraviolet radiation had a favorable effect of shortening the time necessary for etching of particle tracks in the plastic detectors. It was found later^[50] that ultraviolet can significantly weaken the negative action of the annealing temperature (in the same plastics),

For crystalline detectors, which have a higher threshold, this effect (which could appear under the conditions of outer space) has not yet been observed.

On the other hand, study of the indirect effect of background on the detection characteristics is particularly urgent for high-threshold detectors which are suitable for use under the conditions of a high background of weakly ionizing particles. For example, Bastin-Scoffier carried out a parallel study of the effect of thermal annealing (at temperatures of $350-500^{\circ}$ C) and irradiation by a beam of O and He ions (with energy 42 MeV) on mica and silicate detectors (quartz, feldspar, pyroxene) containing in one case old tracks of cosmic origin and in the other case fresh tracks of Kr ions (E = 1 MeV/nucleon).

It was discovered that, first, radiation annealing of already existing tracks requires not only a large total dose but also some critical intensity of bombardment ($\sim (2-4) \times 10^{15}$ MeV per g/cm² per second) and does not reduce to a purely thermal effect and, second, old tracks left in detectors of lunar origin are much more stable than fresh tracks from bombardment in an accelerator.

5. The effective longitudinal etching rate V_t depends very strongly on the solution temperature, and therefore in the most careful work this temperature is maintained with an accuracy of ~0.05°. The dependence of V_t on the concentration of the etching products also requires a number of precautions: stirring of the solution, monitoring of the contents of the etching products in it, periodic replacement of the solution, and so forth.

Thus, solid-state track detectors have been studied to a smaller extent than nuclear emulsions (with respect to the mechanism of the effect) and are subject to a large number of additional factors which have not been well studied and which affect the quantitative characteristics. However, the large assortment of sensitivities, the relatively wider range of bombardment conditions, and in a number of cases the high degree of stability of the latent image of the track make this method an extremely dangerous competitor to nuclear emulsions, particularly for irradiations of great duration for the purpose of searching for very rare tracks and events.

c) Electronic detection methods. As a typical representative of electronic apparatus for study of rather heavy nuclei (up to Z = 26) we show in Fig. 18 a drawing of the apparatus of Webber and Ormes^[58] used in balloon flights in 1964–1965 at atmospheric depths of $2-7 \text{ g/cm}^2$.



FIG. 18. Diagram of the apparatus of Webber and Ormes. [⁵⁸] SCscintillation counters; lucite-Cerenkov radiator; PM-photomultipliers; RC-reflecting cone.

Each detected nucleus was represented as a point in a two-dimensional array S, (S + C), where S is the pulse from the upper (scintillation) counter and (S + C)is the pulse from the combined lower counter. The resolution in charge turned out to be quite adequate for reliable identification of all elements, at least up to Z = 14, for energies E > 0.8 BeV/nucleon, and is not greatly inferior to emulsion data.

The geometrical factor $\Gamma = 50 \text{ cm}^2\text{-sr}$ and the total area-time factor for 7 flights of ~500 m²-sr-sec turned out to be sufficient to determine the charge composition of cosmic rays in the interval Z = 3-26 and the energy spectrum up to 10-12 BeV.

The authors point out that for Z > 5 the unidentifiable background can be neglected, and corrections for nuclear interactions in the detector material (<3 g/cm²) increased with increasing Z, reaching 15% for the iron group.

In the case of heavier nuclei the corrections for nuclear interactions would increase still more rapidly.

The undoubted advantage of electronic apparatus is the possibility of telemetric transmission of the information. However, the problem of the role of background imitation events for very rare nuclei with Z > 40 requires special discussion.

5. RESULTS OF THE SUPERHEAVY NUCLEAR COMPONENT OF COSMIC RAYS

In this section we will consider results obtained mainly with track detectors (emulsions and dielectric detectors) and relating to the chemical composition of cosmic rays, beginning with Z = 30 and above.

The first experiments in this field were started in 1966 in Texas by the Fowler group; this work is being continued at the present time (see ref. 59); the supplementation of the emulsion method with dielectric detectors and the appreciable discrepancy in the results obtained by these independent methods has led to the appearance of a number of previously underestimated difficulties of a systematic nature.

Fowler's first apparatus had emulsion layers of total area 4.5 m² which were exposed in balloon flights in the stratosphere at altitudes with an atmospheric pressure of ~3 g/cm² for a period of ~11 h. A total of 11 nuclei with $Z \ge 40$ were observed (including 2 nuclei with $Z \sim 90$), although the total number of Fe nuclei which passed through the emulsion was estimated as ~2 × 10⁵. In a series of subsequent flights for which groups of specialists from the USA (Price, Walker, Fleischer, and others) prepared and processed also dielectric detectors of plastic materials (Lexan, cellulose triacetate, and others), the area-time factor increased to 1000 m²-h for an area of $\approx 25 \text{ m}^2$.

The combined results of these investigations, in Fowler's opinion, [50] are as follows:

1) There are no important differences between the general behavior as a function of Z for the chemical composition of cosmic rays and the abundance of the elements in the universe studied by astrophysical methods and meteoric observations (see also Fig. 7).

2) Noticeable peaks exist in the cosmic-ray chemical composition curve at Z = 52-54 and in the region $72 \le Z \le 78$ (Fig. 19).

3) Nuclei were observed with $Z \ge 90$.

The second of these conclusions is already a definite argument favoring the existence of processes for synthesis of superheavy elements as the result of rather fast neutron capture,⁹⁾ for only with a short time interval between two successive captures is it possible to jump over the corresponding radioactive elements. In addition, as was noted by Binus et al.,^[62] it is possible in principle even from a detailed analysis of the nuclear composition in the range of Z values 34-40 to make a choice between fast and slow synthesis, since the corresponding predictions for the ratios R of the fluxes $J(34 \le Z \le 36)$ and $J(37 \le Z \le 40)$ differ by roughly a factor of 5 (R = 7-8 for fast synthesis and R = 1.1-1.4 for slow synthesis).

Unfortunately the experimental data^[62] obtained by electronic methods (ionization chamber + Cerenkov counter) turned out to be too scanty (R = 11:2), al-though they favor the hypothesis of fast capture.

As a result of the limited area-time factor and also of instrumental effects the integrated electronic data for the entire region of nuclei clearly heavier than iron (Z > 30) turned out to be not only scanty but also contradictory. For example, the experiments of Volodichev et al.^[64] led to relative fluxes $I(Z > 30)/I_{Fe} \sim 3 \times 10^{-2}$, in clear contradiction to the results of L. V. Kurnosova et al.^[63], also obtained from artificial earth satellites, according to which $I(Z > 30)/I_{Fe} \sim 3 \times 10^{-4}$ (in agreement with the track-detector data).

Since for the transition to the region Z > 92 the area-time factor of the apparatus of Kurnosova et al. would have to be increased by at least 2-3 orders of



FIG. 19. Chemical composition of cosmic rays for Z > 60 according to the data of Fowler. [⁵⁹]

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magnitude, the electronic method is at the present a comparatively ineffective approach to the problem of the search for transuranium elements.

Returning again to the charge region Z = 40-92, we should mention the interesting though still preliminary results of P. S. Price and co-workers^[65], which were obtained with plastic detectors in an apparatus consisting of layers of Lexan, cellulose triacetate, emulsion, and x-ray films, detecting the Cerenkov radiation from heavy nuclei. After exposure of the equipment in balloons for a total area-time factor of ~8000 m²-h and scanning of 80% of the area, the authors observed 66 nuclei with Z > 30, including 8 nuclei in the interval $45 < Z \leq 70$, and 12 nuclei with Z > 70, among them 2 nuclei with Z > 92.

Special mention should be made of the experiments of N. S. Ivanova and co-workers, which were performed in space, on one occasion inside the Earth's magneto-sphere (in the satellites Cosmos 213 and Soyuz $5^{[66]}$) and on another occasion beyond the magnetosphere (in the automated interplanetary probes Zond 5 and Zond $7^{[36]}$).

In the studies in the Zond probes, which utilized lowsensitivity emulsions¹⁰⁾ of total thickness $20-40 \text{ g/cm}^2$, it turned out to be possible to estimate very roughly (on the basis of 4 events) the relative flux of nuclei $J(Z \ge 40)/J(Fe)$, which turned out to be close in order of magnitude (~10⁻⁴ instead of 2×10^{-4}) to the data of the same group for the region inside the magnetosphere.

In addition, under the same conditions (in Zond 5), absolute fluxes of iron nuclei were obtained in two energy intervals, namely:

 $(E \ge 0.15 \text{ BeV/nucleon}) = 0.47 \pm 0.06 \text{ m}^{-2} \text{ sr}^{-1} \text{ sec}^{-1}$,

 $(E \ge 1 \text{ BeV/nucleon}) = 0.23 \pm 0.04 \text{ m}^{-2} \text{ sr}^{-1} \text{ sec}^{-1}$.

This result can appropriately be compared with the conclusion of Fowler et al.^[59] that stopping nuclei (E < 1 BeV/nucleon) play an appreciable role in the flux of superheavy nuclei (Z > 40).

Price et al.^[67] made a comparison of the two independent methods of determining charge (plastic detectors Z_{pl} and emulsions Z_{em}) in the same interval 40 < Z < 92. As can be seen from Fig. 20, the discrepancy between the two methods is in the nature of a random spread amounting on the average, in the opinion of the authors, to ± 3 charge units. Still better agreement between the two methods, within $\Delta Z = \pm 1$, was obtained for emulsion and Lexan in a more recent study by O'Sullivan et al.^[68] in apparatus with a total thickness of 10 g/cm² (including an iron absorber).



FIG. 20. Comparison of results of two methods of measuring nuclear charge: emulsions and plastic detectors.

Adding together the world statistics at the end of 1970, Price et al.^[67] obtained a nuclear flux ratio $(Z > 83)/(70 < 83) \sim 0.3$, thereby confirming the earlier conclusions of Fowler et al.^[35], both that there is a peak in the Pt region due to the stabilizing influence of the shell of 126 neutrons, and that an important role is played by rapid neutron capture in cosmic-ray sources.

All of the data, although they are extremely scarce, on transuranium nuclei (Z > 92) deserve special attention. The revision by the Walker group (see ref. 68) of the 1969 emulsion data [69] was guite sensational. It turned out that, in spite of the high geomagnetic threshold (4.3 BeV), a nucleus considered to be relativistic stopped in the plastic layers under the emulsion, and a new determination of its charge led to the value Z = 76 \pm 4 (instead of the earlier value $Z \approx 110$). With a total exposure in this appratus of $\sim 3000 \text{ m}^2\text{-sr-h}$, no nuclei were observed with Z > 96. After adding the data obtained by Price, Fowler, and co-workers in another similar apparatus and taking into account the substantial absorption of the flux of transuranium nuclei in the atmospheric layer above the latter apparatus (7 g/cm^2) , O'Sullivan et al.^[68] obtain a final value of the absolute flux $J(Z > 96) = (2 \pm 1) \times 10^{-7} \text{ m}^{-2} \text{ sr}^{-1} \text{ sec}^{-1}$ and a relative flux $J(Z > 96) = (5 \pm 2.5) \times 10^{-7} J(Fe)$. The last value can appropriately be compared with the relative abundance of the group Z = 90-92 in the Universe, which Cameron^[70] gives as $J(90 \le Z \le 92)/J(Z = 26) = 6 \times 10^{-8}$.

For correct interpretation of the numbers presented above it is necessary to take into account not only the statistical error in estimation of the flux of nuclei with Z > 96 but also the probability that the true value of the charges of the nuclei observed does not exceed 96. Indicative in this regard are Tables II and III in the report of Fowler et al.^[35], who compare, on the one hand, the probability of imitation of tracks of relativistic nuclei with the measured Z values ($Z_1 = 104$, $Z_2 = 108$) by incompletely relativistic nuclei ($\beta < 1$) with different true values $Z < Z_1$, Z_2 , and, on the other hand, the possibility of not noticing the rise in the ionization of the particle due to its low velocity at the exit from the apparatus in comparison with the entrance (K = J_{ex}/J_{en}). In the first case a charge Z = 96 at energy $E_1 \simeq 1.1$ BeV/nucleon passes with a probability w = 14% and should lead to a rise in ionization K = 1.05; in the second case the corresponding numbers are E_2 \sim 0.8 BeV/nucleon, w = 6%, and K = 1.035. The experimentally measured average values of the relative ionization at the exit are $K'_{exp} = 1.00 \pm 0.003$ according to the emulsion data and $K'_{exp} = 0.97 \pm 0.03$ according to the solid-state-track-detector data (Lexan). It is evident that the probability of imitation of a far-transuranium nucleus by a Cm nucleus, at least in one event, is far from small. Moreover, the large angles between the particle tracks and the plane of the emulsion reduce the reliability of the calibration of its detecting properties.

In order to give a better representation of the general pattern of the results obtained in study of the superheavy (Z > 30) nuclear component of cosmic rays, we will give without any new comments a summary of all of the data discussed above, adding to them the information obtained by means of electronics (Table I).

In addition to experiments with artificially prepared and specially exposed detectors, unique information on

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the existence of transuranium elements in cosmic rays can be obtained from natural detectors—minerals of meteoritic, lunar, and in the future of planetary origin. As can be seen from Fig. 17a, such minerals can have a very high threshold, which assures a tolerable background from nuclei with small Z even with millions of years of exposure. Here, in addition to the total duration, this exposure may turn out to be advantageous also for the reason that the solar system periodically can enter the near zone of activity of supernovae.

A major deficiency of natural detectors is the limited possibility of calibration, as the result of both the very short (and therefore strongly fluctuating in length) tracks of the iron nuclei which serve as a calibration, and the interference from external effects, particularly heating by solar radiation (on the surface of the moon the maximum temperature is ~140°C).

At the present time the data obtained by this method and referring to charges Z > 80 are extremely scarce and not very convincing. In addition to the data already cited,^[14] refs. 71 and 40 discuss data obtained in a

TABLE I. Summary of results obtained in study of the nuclear composition of cosmic rays in the region Z > 30 (the flux of nuclei of the iron group is taken as unity, $I_{Fe} = 1$)

Authors	Method	Relative flux	Notes (conditions) of measurement)
L. V. Kurnosova et al., 1958,	Electr.	$I(Z > 30)^{\bullet})$ 3.6.10 ⁻⁴	Artificial Earth satellite
N. N. Volodichev et al., 1967,	*	$ \begin{array}{c} I(Z > 30) & I(Z > 48) \\ 3.8 \cdot 10^{-2} *) & 4 \cdot 10^{-3} *) \end{array} $	Artificial Earth satellite
P. H. Fowler, 1971 ref. 59	Emutsion	I(Z > 40) I(Z > 70) 2.10 ⁻⁴ 1.5.10 ⁻⁶	Balloons $(\lambda = 41^{\circ} N)$
N. S. Ivanova et al., 1970, ref. 66	Emulsion	a) $I(Z > 40) \sim 10^{4}$ b) $I(Z > 40) \sim 2 \cdot 10^{-4}$	a) Beyond the magnetosphere b) Inside the magnetosphere Balloons
P. H. Fowler, 1971, ref. 59 O'Sullivan et al., 1971, ref. 68	Emulsion + SSTD The same	$ \begin{array}{c} I(2>70) \ I(2>83) \\ 8\cdot10^{-6} \ 2\cdot10^{-6} \\ I(2>96) \\ \sim 5\cdot10^{-7} \end{array} $	$(\lambda = 43^{\circ} \text{ N})$ Balloons $(\lambda = 43^{\circ} \text{ N})$
*)I(Z = 21-	-28) = 1 (fe	$\int_{\mathbf{E}_{\mathbf{r}}} = 1$ the data must be divi	ded by ~1.2).

TABLE II. Results of conversion of chemical composition of cosmic rays to the edge of the atmosphere $[^{35}]$

Ζ	36-43	44-51	52-59	60-67	68-75	76-83	≥ 84
Measured in balloons Edge of the atmosphere Sources: a) $f(X) = \delta(X - X_0), X_0 =$ $= 1 \ g/cm^3$ b) the same, $X_0 = 3 \ g/cm^3$ (445 nuclei with $Z > 84$ in the sources); c) $f(X) = \exp(-X/\lambda_0), \lambda_0 =$ $= 4 \ g/cm^3$	21 23 20 148 15	19 23 16 139 25	13 17 10 81 35	4 8 18 3	5 8 12 24	3 5 12 23	3 6 25 6 39

TABLE III. Comparison of chemical composition of cosmic rays in sources (after conversion to the last variant of Table II) with predictions of the models of fast (r) and slow (s) neutron capture

Z	24-28 (Fe group)	36-43	44-51	52-59	60-67	6875	78-83	≥ 84
Spectrum in source $(\lambda_0 = 4 g/cm^2)$ Predictions:	1.1.10*	4	7	9	0	6	6	10
r process s process	2.4.10 2.6.10	48 200	10 9	16 17	2 1	1	6 12	3.3 0
<u>Note.</u> Normalization of all distributions in Z was performed in the region $Z \ge 52$.								

series of successive sections and etchings of samples
from the Patvar and Johnstown meteorites. The pres-
ence of tracks of length ~ 0.5 mm in the first case and
\sim 1 mm in the second case is, in the opinion of the
authors, an indication of the presence in cosmic rays
of nuclei with $Z > 82$ and, possibly, even $Z > 92$.

Maurette et al.^[71], who are cited by Price and Fleischer^[40], show a mosaic microphotograph of a track (of length 1 mm) of a fast strongly ionizing particle in a crystal of pigeonite taken from sample No. 12021 of lunar soil provided by the crew of Apollo 12. On the basis of results of artificial bombardment of minerals of similar properties under terrestrial conditions, the authors consider that the two cracks observed by them of this type are due to nuclei with $Z \ge 80$.

In order to draw from the experimental data presented above a conclusion as to the nature of the synthesis of elements in cosmic-ray sources, the data must first be corrected in two ways. First, it is necessary to take into account the change in chemical composition on passage of the cosmic rays through the comparatively thin $(\sim 3 \text{ g/cm}^2)$ layer of the atmosphere which separated the balloon equipment from outer space. As can be seen from Table II, this correction, according to Fowler's calculations^[35] amounts to only $\sim 10\%$ for the interval Z = 36-43, but is already $\sim 100\%$ for $Z \ge 84$. Second, it is necessary to take into account the effect of the interstellar medium in the path from the sources to the solar system. In this case a specific model is necessary for the distribution of sources in space, taking as a measure of the distance X the thickness of the material traversed (hydrogen), expressed in g/cm^2 . Two models are frequently used: a) a delta function $\delta(X - X_0)$; b) an exponentially falling distribution $f(X) = \exp(-X/X_o)$.

As also can be seen from Table II, for the first model the minimum permissible value $X_0 = 3 \text{ g/cm}^2$ (from the chemical composition in the region Z = 3-5) already gives an unacceptably large flux of nuclei with Z < 52 (for the observed number of nuclei with Z > 52), and only $X_0 = 1 \text{ g/cm}^2$ agrees with experiment. It must be concluded, therefore, that astrophysical objects approaching us in which uranium and transuranium elements are being "prepared" are separated by a layer of matter ~0.5 g/cm² thick, and the synthesis processes themselves occurred no more than 10⁶ years ago.

For the exponential model a value $\lambda_0 \leq 4 \text{ g/cm}^2$ (compatible with the data on chemical composition of the lightest elements) can still be reconciled with experiment (the expected fraction of elements in the source in the interval Z = 60-67 in this case almost fails to become negative in this region). If we assume that the exponential distribution of sources is more likely, we can compare, as was done by Fowler,^[35] the chemical composition of the superheavy nuclear component of cosmic rays, converted to the sources, with the corresponding predictions of Cameron and Clayton for fast (r-process) and slow (s-process) neutron capture. As can be seen from Table III, although the predictions are significantly closer to reality in the case of the r process, they are still in poor agreement with the observations in the interval Z = 36-43.

We will give a general summary of the results set forth in this section.

First, the data already accumulated on the nuclear composition of cosmic rays in the region Z = 36-92provide a basis (particularly after recalculation to the sources) for giving a clear preference, of the two possible mechanisms for synthesis of elements in the sources, to the process of fast neutron capture. Second, the data on existence of transuranium elements are extremely scarce and not very reliable. The existing experimental estimates of fluxes, which give more an upper limit than a true value, lie at the level ~10⁻⁶ of the flux of iron nuclei. The better estimates of charges have an error of ~3%, but these values do not include systematic errors, which act more in the direction of exaggerating the values than lowering the values of Z.

In connection with the last statement we recall that in the region Z > 92, three isotopes are known with lifetimes greater than 10⁶ years. These are $_{93}Np^{237}$ ($T_{1/2} = 2 \times 10^8$ years), $_{94}Pu^{244}$ ($T_{1/2} = 82 \times 10^6$ years), and $_{96}Cm^{247}$ (25×10^6 years). Therefore reliable experimental estimation of charge in the range Z = 93-96 is quite compatible with the set of existing data on the average age of cosmic rays; here the short range for fragmentation and fission of these nuclei permits us to speak of the existence of closer (both in space and in time) processes of synthesis of elements with subsequent acceleration of their nuclei to energies characteristic of cosmic rays. It is necessary to have in mind also the fact that incomplete ionization of atoms facilitates their escape from the shells of their respective stars, since for the same velocity the magnetic rigidity of the ion turns out to be a monotonically increasing function of the nuclear charge.

6. CONCLUSION. EXPERIMENTAL REQUIREMENTS AND PROSPECTS

When we consider our experience with experimental studies of the superheavy nuclear component of cosmic rays in recent years, we reach a number of conclusions important for planning of new experiments of the transuranium problem.

First, in selection of the measurement technique we must have in mind the following three main requirements:

1) sufficiently high area-time factor of the apparatus ($\gg 1 \text{ m}^2$ -month);

2) reliability of calibration of the detectors measuring the charge, over a sufficiently wide range (Z = 25-125);

3) the possibility of reliable identification and accurate measurement of the characteristics of extremely rare events under conditions of a high background of nuclei with lower charge and other imitating events. This last requirement greatly limits the use of electronic (nontrack) apparatus, particularly Cerenkov counters, in which imitating events may be produced by multiple production of particles.

Second, study of a wide range of charges and velocities of nuclei with a significant spread in the angles of arrival at the detector requires use of a large set of independent track detectors (both emulsion and dielectric) providing a charge resolution better than 2-3%.

Third, the combination of high area-time factor with the unavoidable limitations in weight and size of the apparatus used requires a careful approach to selection

TABLE IV. Integral distributions $N(> R_0)$ in range for nuclei with Z = 26 and Z = 100, converted from the energy spectrum of nuclei with Z \ge 20. [⁵⁸]

	R_0 , g/cm ²				
		t	3	10	35
Z = 26 (Fe)	$E(R_0), BeV/nucleon$ $N [> E(R_0)]$	0.1 0.97	0.2 0.90	0.4 0.77	0.85 0.35
Z = 100 (Fm)	$E(R_0), \text{BeV/nucleon} \\ N \mid > E(R_0) \}$	$\substack{0.2\\0.93}$	0.4 0.80	$0.85 \\ 0.36$	1.3. 0.26

of the optimum region of energy and range of the detected nuclei. Analysis of existing data on energy spectra of heavy nuclei (in particular, the data of Webber and Ormes^[58]) and conversion of the energies to ionization ranges (Table IV) show that the optimum range of detector thickness should be about $3-10 \text{ g/cm}^2$ (including the total thickness of the container).

Fourth, the requirements of high area-time factor and low energy thresholds (including both ionization loss and nuclear interaction in the material, as well as the geomagnetic cutoff of the detected particles) can be satisfied only in space flights of long duration with highlatitude orbits. The statistical reliability of the experimental results expected in this case amounts, in particular, to the order of 10 nuclei of transuranium elements for six-month exposure of detectors of total weight ~100 kg.

Fifth, the need of using detectors of large area (up to 10 m^2) requires increased attention to development and perfection of means of automation of the search, and preferably also a rough measurement of the track parameters associated with the charge and velocity of the nuclei detected.

Sixth, in addition to artificially prepared detectors, broad use must be made in solution of this problem of cosmic mineral detectors of meteoric and particularly of lunar origin. In choice of the optimum depths and in the corresponding calibration in accelerators, these detectors have basic advantages associated with their very high threshold for detection of charged particles and their very long (>10⁸ years) exposure times.

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¹⁾At one time it appeared that all of these attempts were meaningless as a result of the sharp drop in spontaneous fission lifetimes, according to a law $\exp(-Z^2/A)$.

- ⁵⁾This value is determined by the relativistic rise in ionization of the particles, which usually amounts to $\sim 14\%$.
- ⁶⁾In cosmic-ray experiments such calibration is possible over a limited range of Z as long as the experimenter can utilize observations of the fragmentation of relativistic nuclei for direct measurement of Z.
- ⁷⁾In a number of cases it turns out to be useful to find the spatial distribution of the characteristic quantity $(dE/dx)\beta^2/Z^2$, which depends only weakly on β .
- ⁸)Note that the threshold sensitivities of N-3 and Ya-2 emulsions have been determined very roughly and are not characterized by high stability.

²⁾Even before this, Thompson's group [¹⁶] attempted to use the reaction ${}^{40}Ar + {}^{248}Cm \rightarrow {}^{28x}114 + xn$, but the huge neutron deficit (~14) relative to the N = 184 shell left too few chances of success.

³⁾The corresponding element has been assigned a charge Z = 108.

⁴⁾This could be, for example, a device which rotates the emulsion block by 180°.

⁹⁾This process, which is called the r-process, must occur rapidly in com-

parison with β decay of the corresponding nuclei. [⁶⁰] Typical physical parameters which make this process possible are estimated [⁶¹] to be: temperature ~2 X 10⁹ degrees, neutron density 10²⁸ cm⁻³, average time of cycle ~10 sec. In the neutron-capture stage, nuclei can be obtained with $Z_{max} \approx 102-104$ ($A_{max} = 299-307$), and then as the result of a series of β decays they can be transformed to the region Z = 110-113 (A = 294-297).

- ¹⁰⁾The use of ordinary relativistically sensitive emulsions was excluded as the result of the extended exposure (~160 hours).
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