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# SPONTANEOUS EMISSION OF HEAVY-IONS FROM URANIUM\*

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The present paper reports experimental evidences that  $^{238}\text{U}$ , and perhaps other heavy nuclei, besides undergoing spontaneous fission, are also emitters of ions in the mass-range from 20 to 70. Estimates obtained by means of the WKB method indicate half-lives of  $10^{15}$  to  $10^{18}$  years for some of these processes, which agree with our findings. Our results are supported by a systematic observation of neon and argon with abnormal isotopic abundance in both radioactive minerals and helium-bearing natural gases.

The average value for the half-life of spontaneous fission of  $^{238}\text{U}$  obtained by different methods of measurement is  $8.8 \times 10^{15}$  years (see Table I). Such low fission activity makes precise measurements difficult, leading to large disagreement among different methods and individual measurements. Thus, using

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an integral recording method such as loaded nuclear emulsion, for statistical reasons and for greater accuracy, very long exposures are necessary in order to collect the desired number of tracks from fission events.

We were successful in conducting a long exposure experiment, using total exposures up to 12,500 mg-days of uranium per square centimetre of loaded nuclear emulsion. For this experiment a special development technique was worked out, which aimed at recording, in the bulk of the emulsion, very legible tracks from the spontaneous fission events, in spite of the presence of a background of about  $10^6$  alpha particles per fission event<sup>1-5</sup>. To hinder the fading of the latent image from fission tracks, the nuclear emulsion pellicles were stored at low temperatures during the entire exposure time (in some cases up to 8.5 years). The method used allows development of the very legible fission tracks, and at the same time does not record alpha particle tracks at all. Moreover, the development technique we used does not register tracks from ions with charge  $Z$  lower than 9. The long total exposures yielded a sufficiently high population of tracks (about  $10^3$  per square centimetre). For discrimination and precise counting of tracks from spontaneous fission, we made a detailed range-track histogram of all track-ranges recorded under our experimental conditions. This histogram showed a very surprising result: the appearance of a new peak, together with the characteristic peak of spontaneous fission tracks from  $^{238}\text{U}$  (see Fig. 1). The new peak was attributed to tracks from heavy-ions with  $Z$  equal to or greater than 10, since they are not related to the tracks originated by ordinary fission processes. The chance of instru-

mental error was ruled out. The possibility of short range tracks being caused by an uneven sensitivity in the nuclear emulsion leading to the recording of alpha particle tracks in local high sensitivity regions is unlikely, because such a high population of alpha particles would yield local clumps of a high number of tracks, and not dispersed isolated tracks in the bulk of the emulsion. Moreover, there was no observable development gradient in the emulsion thickness.

The first observation of these unexpected shorter tracks in the nuclear emulsion led us to search for the same short tracks on plates used in previous spontaneous fission measurements, carried out in our laboratory and submitted to a wide range of total exposures<sup>6</sup>. We observed systematically the same range-track distributions with the same clear peak related to the shorter length tracks.

The origin of these short range tracks was attributed by us to a spontaneous nuclear phenomenon, a case of large-nucleon-cluster emission, and considering that the nuclear emulsion plates were loaded with isotopes from the natural uranium, our hypothesis was that this heavy-ions emission arises from the isotope  $^{238}\text{U}$  and not from  $^{235}\text{U}$  or  $^{234}\text{U}$ , which are both less abundant. By microscopic inspection of the ionization and careful analysis of the range distributions of these short tracks, we concluded that we were dealing with spontaneous emission from  $^{238}\text{U}$ , of ions of intermediate masses in the region from neon to nickel. Energetically, this heavy-ion decay mode from  $^{238}\text{U}$  (and from other heavy nuclei) is possible, although in our experiment there was no means of identifying the charge and the mass of the

recorded ions. We calculated the range and the mean relative ionization of heavy-ions in emulsion from the Q-value, for some probable spontaneous decay modes. The measured ranges of the short length tracks agree quite well with the calculated energies and ranges of such ions (see Table II). The calculated mean ionization of the ions at such energies indicates that these heavy-ions are in the average less ionizing than an ordinary fission fragment, only by a factor of two, and therefore adequate to be recorded by means of our nuclear emulsion discrimination technique.

In the case of spontaneous fission fragments, the recorded long tracks are so legible that 90% efficiency is achieved in counting them, even with a relatively low population of tracks. Unfortunately, this high efficiency is not achieved in the case of the short range tracks. Microscopic scanning losses due to shorter range and lower ionization, together with very low track population, yielded a low counting efficiency (30%) for our shortest total exposure. However, applying the statistical method of double scanning<sup>7</sup> it was possible to estimate the true counting rate of the short range tracks. By this method, the half-life estimated for the new spontaneous phenomenon was  $(2 \pm 1) \times 10^{15}$  years, for all heavy-ion emission modes with an amount of activity large enough to contribute tracks during total exposures. The peak, which is centered at about  $24\mu\text{m}$ , corresponds to ordinary fission tracks (shown in Fig. 1) measured with good accuracy, and leads to a spontaneous fission half-life of  $(6.0 \pm 0.4) \times 10^{15}$  years. This result is comparable to reported values obtained from different experimental me

thods (see Table I).

Experimental evidence which strongly supports our heavy-ion decay hypothesis, is the presence of small amounts of neon and argon in the radioactive minerals and in the helium from natural gases. The neon and argon may have their origin in the spontaneous emission from either  $^{238}\text{U}$  or  $^{232}\text{Th}$ . In fact, an isotopic abundance considered highly abnormal in relation to the atmosphere has been observed for isotopic abundance of this neon and argon<sup>8-12</sup>. The explanation for the origin of this unusual isotopic abundance is still not very clear<sup>12,13</sup>, and it may result, as we stated above, from a spontaneous emission from either  $^{238}\text{U}$  or  $^{232}\text{Th}$ . The  $^{22}\text{Ne}$  isotope excess found in some radioactive minerals which contain uranium and thorium was attributed to the reaction of  $^{19}\text{F}$  with alpha particles to yield  $^{22}\text{Ne}$  as a final product. This mechanism is unlikely, since the concentrations of fluorine are not sufficient to produce the observed amount of  $^{22}\text{Ne}$ , as pointed out by Sharif-Zade et al.<sup>12</sup>. Assuming, however, that the excess of  $^{22}\text{Ne}$  observed in the neon gas contained in uranium minerals comes from the spontaneous emission of  $^{22}\text{Ne}$  from  $^{238}\text{U}$ , we were able to calculate the half-life of such a process from the data available in literature. The result from G-3 sample of Eastern USSR pitchblende measurements<sup>12</sup> was  $2.2 \times 10^{15}$  years. Considering the large errors expected in such a kind of measurement, this result is consistent with our observations. The same happens with the excess of  $^{38}\text{Ar}$  isotope from minerals which contain uranium<sup>9</sup>, whose results show half-lives of  $10^{17}$  to  $10^{19}$  years, which are compatible with our findings.

The spontaneous emission of heavy-ions from  $^{238}\text{U}$  de-

tected in the present experiment may contribute to some of the anomalous pleochroic halos (dwarf halos) recorded in black micas and observed by mineralogists long ago<sup>14,15</sup>, and whose agent was not yet properly explained<sup>16,17</sup>. Pleochroic halos are colored concentric rings formed by prolonged alpha particle emission from small radioactive inclusions in the host material, and each of the rings can be identified by the alpha-emitters of the uranium or thorium decay series. Recently, a variety of dwarf halos was also discovered with radii ranging from 1.5 $\mu$ m to 11 $\mu$ m and it was shown that the small bleached regions immediately surrounding the radioactive inclusions are regions of high radiation damage<sup>17</sup>. Furthermore, in emulsion, the radii of these dwarf halos would correspond to ranges between 4 $\mu$ m and 15 $\mu$ m. The emission from <sup>238</sup>U of heavy ions with similar ranges may also contribute to such halos.

Radiochemists, searching for true ternary fission of <sup>235</sup>U induced by thermal neutrons have reported a few cases of strong abnormal yield of light nuclides such as <sup>28</sup>Mg, <sup>35</sup>S, chlorine isotopes, argon isotopes, scandium isotopes, <sup>45</sup>Ca, <sup>56</sup>Co, <sup>59</sup>Fe, <sup>66</sup>Ni and <sup>67</sup>Cu<sup>18-21</sup> which may originate not from tripartition but from the heavy-ion decay modes of the excited nuclei. In addition, radiochemical studies of spontaneous fission of <sup>252</sup>Cf have also reported relative yields of some light nuclides far from the fission mass-yield curve such as <sup>28</sup>Mg, <sup>43</sup>K and <sup>66</sup>Ni<sup>22</sup>, whose origin may be attributed to a case of spontaneous heavy-ion emission.

Experimental energy cut-off in fission experiments at energies above the Q-value of heavy-ion emission, together

with a lower activity for such processes, may be responsible for non-observation of the heavy-nucleon-cluster emission phenomena. The mass and energy yielded by such processes are strongly asymmetrical, for the recoil nuclide and heavy-ion emitted does not yield an energy released above the usual cut-off of the fission experiments.

In this note we do not intend to discuss models which explain the mechanism of the phenomena observed, but only to give some results of rough calculations obtained by means of the classical WKB method for penetration of potential barriers for the half-life of this possible spontaneous process. Assuming the previous existence of heavy-nucleon-clusters, a simple systematic calculation of penetration through a potential barrier for these nucleon-agregates points out the possibility of a few heavy-ion emissions with mass number ranging from 20 up to 70, whose yield is of the same order of magnitude as the yield of spontaneous fission (see Fig. 2). In these estimations, the formalism of the alpha-decay process was used in the case of heavier cluster emission modes. The total decay-constant for all heavy-ion emission modes is given by

$$\lambda = \sum_i \lambda_i \quad (1)$$

where  $\lambda_i$  is the decay-constant for a given specific decay mode, which is obtained as follows:

$$\lambda_i = \lambda_{0i} e^{-G_i} \quad (2)$$



$$\lambda_{oi} \text{ [yr}^{-1}\text{]} = \frac{2.191929 \times 10^{29}}{R - R_{xi}} (0.85 Q_i / m_i)^{1/2} \quad (3)$$

$$G_i = 0.52494 P_i (Z_{xi} Z_{yi} m_i b_i)^{1/2} \quad (4)$$

$$b_i \text{ [fermi]} = 1.439898 Z_{xi} Z_{yi} / 0.85 Q_i \quad (5)$$

$$P_i = \arccos \left( \frac{R_i}{b_i} \right)^{1/2} - \left[ \frac{R_i}{b_i} - \left( \frac{R_i}{b_i} \right)^2 \right]^{1/2} \quad (6)$$

In these expressions  $Q_i$  (expressed in MeV) is the total energy released and  $m_i$  (expressed in amu) is the reduced mass of the system. Both these quantities were calculated as is indicated in Table II.  $Z_{xi}$  and  $Z_{yi}$  are the charges of the recoil and the emitted nuclides, respectively.  $R_i$  represents the sum of the nuclear radii of these two nuclides, i.e.,  $R_i = R_{xi} + R_{yi}$ . The nuclear radii were calculated according to  $R = r_0 A^{1/3}$ , where  $A$  is the mass number and  $r_0$  was taken to be a adjustable parameter (in Eq. (3)  $R$  is the nuclear radius of the parent nucleus ( $^{238}\text{U}$ ) and  $R_{xi}$  is that of the emitted ion). Such a simplified model gives calculated half-lives in agreement with our experimental results when 15% of the total energy released is assumed to be excitation energy of recoil and emitted nuclides, and the nuclear radius parameter is assumed to be 1.324 fermi. The decay-constant,  $\lambda$ , is strongly affected by the nuclear radius parameter and by the masses of both the recoil and the emitted nuclide. Obviously, the resulting half-lives as calculated by the above procedure, are subjects to errors of several orders of magnitude and therefore must be seen as indicative only of the pos-

sibility of some heavy-ion emission modes. Within the referred limits of large errors, caused mainly by uncertainties in the calculated mass of the recoil nucleus, the calculations indicate that about twenty of such processes are expected to occur with a half-life of less than  $10^{24}$  years, the most probable processes being the emission of  $^{50}\text{Ca}$ ,  $^{48}\text{Ca}$  and  $^{49}\text{Ca}$ . About ten of the possible processes occur with emission of heavy-ions with magic numbers of neutrons and protons. For instance, the heavy-ion  $^{48}\text{Ca}$  would be emitted with a half-life of about  $10^{16}$  years.

Fission induced in heavy nuclei by high energy particles shows also the production of individual fragments (single unpaired tracks recorded, for instance, by means of the mica-target-mica sandwich technique<sup>23</sup>). The origin of such single fragments has not been clearly explained. It may be attributed to a local high energy nuclear excitation (leading to the so-called fragmentation phenomenon), but could also, on the contrary, be the result of a very low energy excitation process, which happens in a non-negligible percentage of all high energy induced fission processes. This fact could be responsible for the emission of the unpaired fragments. To obtain experimental confirmation of such a process we exposed nuclear emulsions loaded with natural uranium to the low energy photon beams produced at the reactor "Saphir" of the EIR (Würenlingen, Switzerland). The irradiations were performed in collaboration with Dr. W. Wölfli<sup>24</sup> by means of an intense 8.86 MeV gamma-rays flux (about  $10^{11}$  photons per square centimetre) obtained by the (n, $\gamma$ ) reaction on nickel. The targets were positioned at right angles with respect to the incident beam. The irradiated plates were submitted to the same

development technique previously used in spontaneous fission studies. Since the photofission cross-section at the energy considered is about 30 mb and the time lapse between the loading of the emulsion and its development was only 70 days, the spontaneous events represent less than 1% of the total recorded tracks. We observed this time a clear fine structure in the range-track distribution (see Fig. 3) with a definite peak at about 15 $\mu$ m.

It is our conclusion that the experimental facts outlined above, such as the strongly abnormal isotopic abundance of neon and argon found in minerals containing uranium and thorium and in helium-bearing natural gases, and the peaks of the short range tracks observed in nuclear emulsion reported in the present note, seem to support evidence of a new type of radioactivity, i.e., the spontaneous emission of heavy-ions from  $^{238}\text{U}$ , and perhaps from other heavy nuclei as well. We anticipate seeing our results confirmed by other researchers.

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24. A detailed discussion on this subject will be presented in a future paper.

TABLE I - Some Experimental Results Concerning the Spontaneous Fission Half-life of  $^{238}\text{U}$

| Authors               | Year | Experimental Method | Half-life (units of $10^{15}\text{yr}$ ) | Ref. (*) |
|-----------------------|------|---------------------|--|----------|
| Libby                 | 1939 | Radioch.            | > 0.1                                    | 1        |
| Petrzhak and Flerov   | 1940 | Fission Chamber     | 100                                      | 2        |
| Perfilov              | 1947 | Nuclear Emulsion    | 13.0 ± 2                                 | 3        |
| Perfilov              | 1947 | Ioniz. Chamber      | 13.17                                    | 4        |
| Segrē                 | 1952 | Ioniz. Chamber      | 8.04 ± 0.30                              | 5        |
| Kuroda and Edwards    | 1954 | Radioch.            | 5.9 ± 0.6                                | 6        |
| Kuroda and Edwards    | 1954 | Radioch.            | 6.1 ± 1.5                                | 6        |
| Kuroda et al.         | 1956 | Radioch.            | 10.3 ± 1.0                               | 7        |
| Parker and Kuroda     | 1956 | Radioch.            | 8.4 ± 0.8                                | 8        |
| Kuroda and Edwards    | 1957 | Radioch.            | 5.9 ± 0.4                                | 9        |
| Parker and Kuroda     | 1958 | Radioch.            | 8.0 ± 0.5                                | 10       |
| Gerling et al.        | 1959 | Radioch.            | 5.8 ± 0.5                                | 11       |
| Kuz'minov et al.      | 1960 | Ioniz. Chamber      | 6.5 ± 0.3                                | 12       |
| de Barros et al.      | 1963 | Nuclear Emulsion    | 8.5 ± 0.5                                | 13       |
| Fleischer and Price   | 1964 | Mica                | 10.5                                     | 14       |
| Fleischer and Price   | 1964 | Radioch.            | 10.0                                     | 14       |
| Rao and Kuroda        | 1966 | Radioch.            | 8.88                                     | 15       |
| Roberts et al.        | 1967 | Mica                | 10.1                                     | 16       |
| Spadavecchia and Hahn | 1967 | Spinner             | 8.23 ± 0.10                              | 17       |
| Ishmori et al.        | 1967 | Radioch.            | 7.191 ± 0.158                            | 18       |
| Roberts et al.        | 1968 | Mica                | 9.85                                     | 19       |
| Galliker et al.       | 1970 | Spinner             | 8.19 ± 0.06                              | 20       |
| Leme et al.           | 1971 | Mica                | 9.5                                      | 21       |
| Condē and Holmberg    | 1971 | Ioniz. Chamber      | 11 ± 2                                   | 22       |

(\*)References for this Table are quoted in the Appendix.

TABLE II - Calculated Range and Mean Relative Ionization in Standard Emulsion for Some Possible Cases of Spontaneous Emission of Heavy-ions from  $^{238}\text{U}$ .

| Emitted Nuclide  | Q-Value <sup>a</sup> (MeV) | Kinetic Energy <sup>b</sup> (MeV) | Range <sup>c</sup> ( $\mu\text{m}$ ) | Relative Ionization <sup>d</sup> |
|------------------|----------------------------|-----------------------------------|--------------------------------------|----------------------------------|
| $^{10}\text{Be}$ | 7.4                        | 6.0                               | 8.0                                  | 4.8                              |
| $^{11}\text{B}$  | 11.1                       | 9.0                               | 9.0                                  | 5.6                              |
| $^{12}\text{C}$  | 20.7                       | 16.7                              | 13.6                                 | 5.2                              |
| $^{14}\text{C}$  | 23.7                       | 18.9                              | 15.4                                 | 5.3                              |
| $^{14}\text{N}$  | 19.3                       | 15.5                              | 10.4                                 | 7.7                              |
| $^{15}\text{N}$  | 26.0                       | 20.7                              | 14.0                                 | 6.6                              |
| $^{15}\text{O}$  | 20.0                       | 15.9                              | 9.1                                  | 9.5                              |
| $^{16}\text{O}$  | 31.7                       | 25.2                              | 14.5                                 | 7.5                              |
| $^{18}\text{F}$  | 28.4                       | 22.4                              | 11.2                                 | 10.3                             |
| $^{20}\text{F}$  | 36.0                       | 28.1                              | 14.1                                 | 9.6                              |
| $^{20}\text{Ne}$ | 41.5                       | 32.3                              | 14.3                                 | 10.3                             |
| $^{21}\text{Ne}$ | 43.0                       | 33.3                              | 14.8                                 | 10.4                             |
| $^{22}\text{Ne}$ | 49.5                       | 38.2                              | 16.9                                 | 9.9                              |
| $^{24}\text{Ne}$ | 54.1                       | 41.3                              | 18.3                                 | 9.9                              |
| $^{22}\text{Na}$ | 39.6                       | 30.6                              | 12.2                                 | 12.8                             |
| $^{26}\text{Na}$ | 56.3                       | 42.7                              | 16.9                                 | 11.7                             |
| $^{24}\text{Mg}$ | 50.8                       | 38.9                              | 14.0                                 | 13.3                             |
| $^{28}\text{Mg}$ | 67.3                       | 50.5                              | 18.1                                 | 12.6                             |
| $^{27}\text{Al}$ | 58.4                       | 44.0                              | 14.5                                 | 14.8                             |
| $^{30}\text{Al}$ | 68.8                       | 51.1                              | 16.8                                 | 14.5                             |
| $^{28}\text{Si}$ | 60.8                       | 45.6                              | 13.8                                 | 16.4                             |
| $^{31}\text{Si}$ | 74.7                       | 55.2                              | 16.6                                 | 15.7                             |

TABLE II (Continued)

|                  |       |      |      |      |
|------------------|-------|------|------|------|
| $^{30}\text{P}$  | 59.5  | 44.2 | 12.9 | 18.9 |
| $^{31}\text{P}$  | 69.1  | 51.1 | 14.6 | 17.9 |
| $^{32}\text{S}$  | 68.6  | 50.5 | 13.8 | 19.9 |
| $^{36}\text{S}$  | 93.4  | 67.4 | 17.8 | 18.3 |
| $^{38}\text{S}$  | 95.2  | 68.0 | 18.0 | 18.7 |
| $^{42}\text{Ar}$ | 103.6 | 72.5 | 17.6 | 22.1 |
| $^{47}\text{K}$  | 112.1 | 76.5 | 18.0 | 24.3 |
| $^{40}\text{Ca}$ | 85.2  | 60.3 | 13.8 | 26.7 |
| $^{48}\text{Ca}$ | 119.7 | 81.2 | 18.2 | 25.5 |
| $^{50}\text{Ca}$ | 120.4 | 80.8 | 18.3 | 26.0 |

- <sup>a</sup> The Q-values reported in this Table were calculated from nuclear masses of both the parent nucleus ( $^{238}\text{U}$ ) and emitted heavy-ion taken from the compilation of Wapstra and Gove - Nuclear Data Tables. (1971), 19, 265 - ; the masses of the recoil nuclei were calculated assuming the semi-empirical nuclidic mass equation of Wing and Fong - Phys. Rev., (1964), 136, B923 - .
- <sup>b</sup> Kinetic energies were calculated from the Q-value for each specific process assuming, in all cases, 15% of the total energy released as excitation energy of the product nuclides.
- <sup>c</sup> Ranges and relative ionizations in standard emulsion for heavy-ions listed above were calculated following the paper of Heckman et al. - Phys. Rev., (1960), 117, 544).
- <sup>d</sup> Relative Ionizations are to that of an alpha particle of 4.2MeV.



APPENDIX

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FIGURE CAPTIONS

Fig. 1 - Typical range distribution of tracks recorded in uranium loaded nuclear emulsion plates. This histogram was obtained by means of our nuclear emulsion discrimination technique. A large, clear peak appears centered at about  $10\mu\text{m}$ . The nearly gaussian distribution centered at about  $23\mu\text{m}$  undoubtedly corresponds to the spontaneous fission tracks from  $^{238}\text{U}$ . No correction for counting efficiency is taken into account in this histogram. The maximum error in obtaining individual track-range (which originates mainly from a shrinkage factor  $\approx 4$ ) is  $\pm 3\mu\text{m}$ .

Fig. 2 - Mass-yield distribution of spontaneous emission processes from  $^{238}\text{U}$ . The full circles are the results of calculations for some specific heavy-ion decay modes as indicated in the text, and correspond to the experimental total half-life of  $(2 \pm 1) \times 10^{15}$  years. The full line is the experimental mass-yield curve of light fragments of spontaneous fission obtained by Rao and Kuroda (Phys. Rev., 147, 884 (1966)), and corresponds to a half-life of  $(6.0 \pm 0.4) \times 10^{15}$  years. The total yield for these two processes is normalized to 100%. We also reported in this figure the upper limits of the yields of some light nuclides from thermal neutron-induced fission on  $^{235}\text{U}$  ( $\Delta$ ,  $\square$ ,  $\blacksquare$ ,  $\nabla$ ) and from spontaneous fission of  $^{252}\text{Cf}(0)$ . Experimental data are taken from:  $\Delta$ , Ref. 18;  $\square$ , Ref. 20;  $\blacksquare$ , Ref. 19;  $\nabla$ , Ref. 21;  $\circ$ , Ref. 22.

Fig. 3 - Track-range distribution of an uranium loaded nuclear emulsion exposed to a photon beam from nickel radioactive neutron capture spectrum (main line energy of 8.86MeV). To improve accuracy in individual track-range measurements, only tracks with dip equal to or less than  $2\mu\text{m}$  have been used in this histogram.

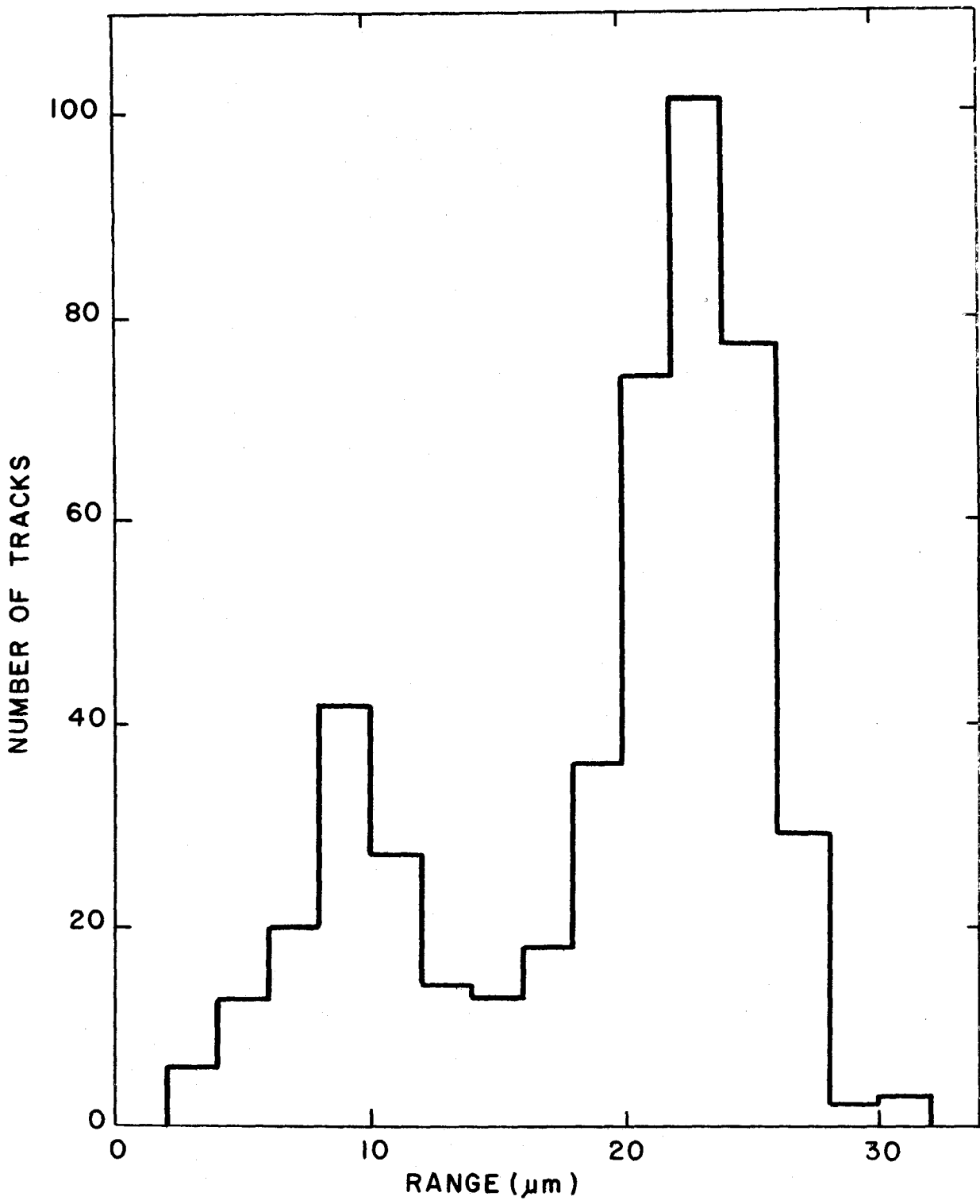


FIG. 1

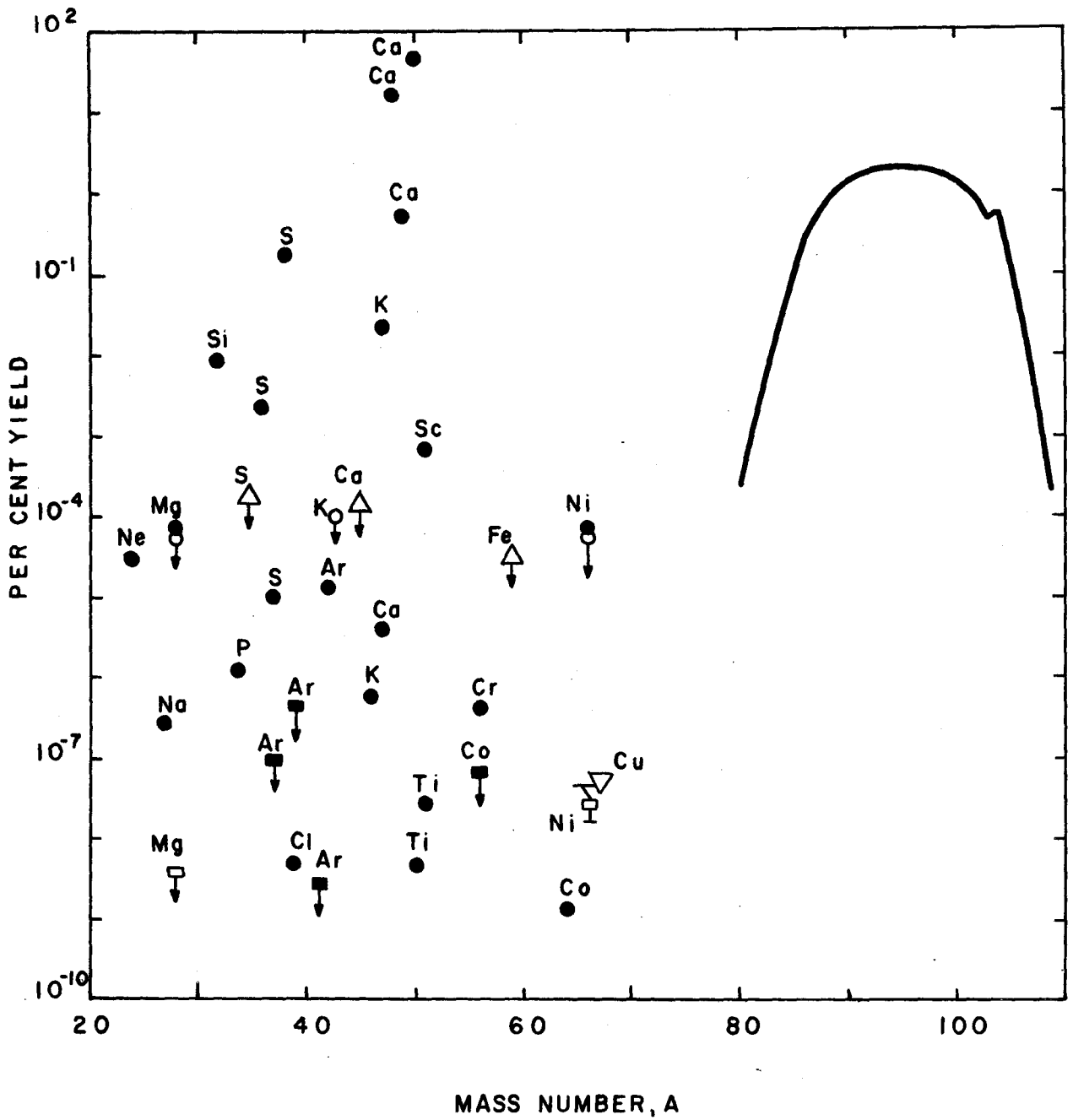


FIG. 2

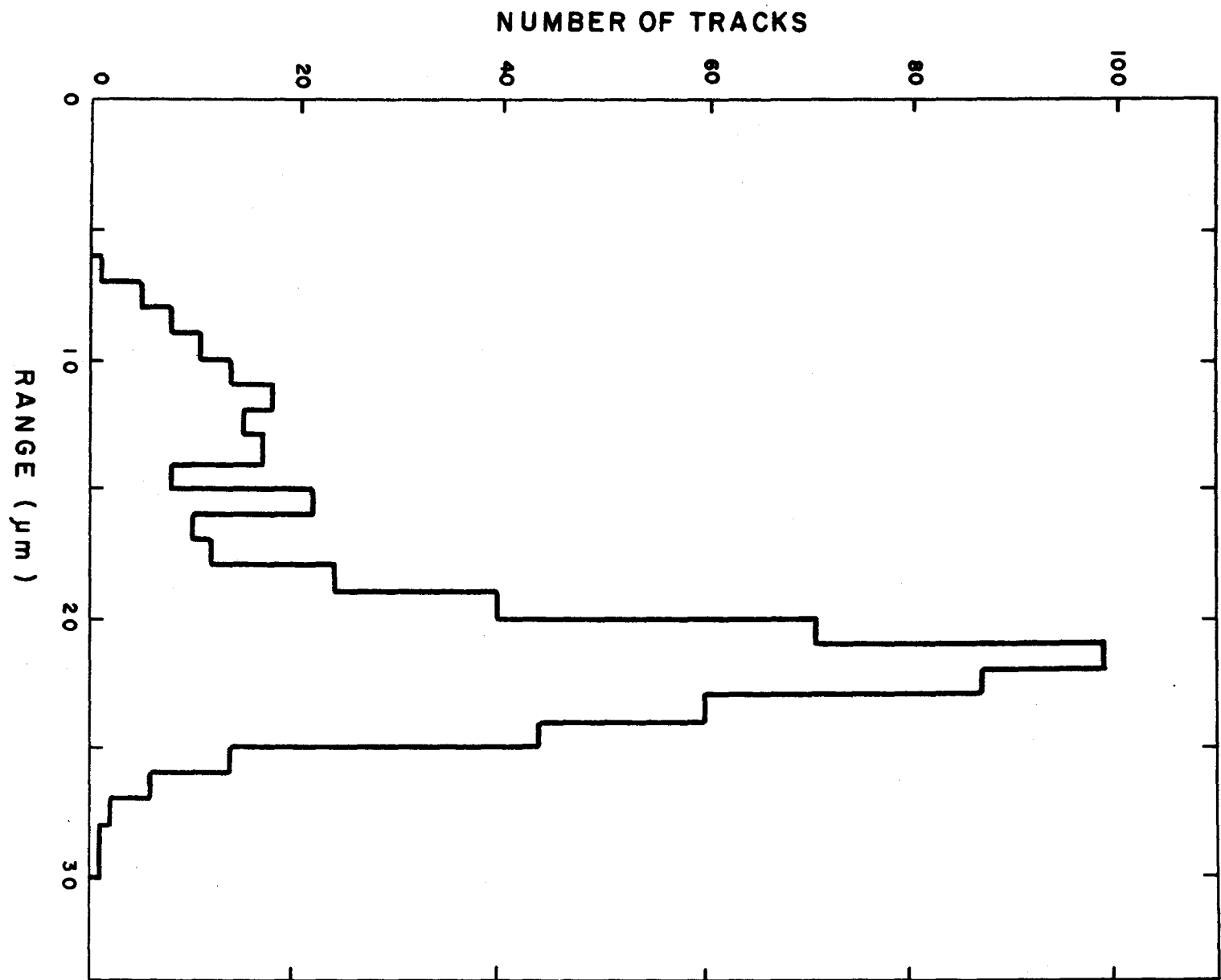


FIG. 3