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LONG-RANGE FISSION FRAGMENTS FROM RADIOGENIC LEAD†

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ABSTRACT

Fission tracks with a range 19% longer than tracks produced by uranium fission fragments were observed on mylar foils after a three year underground (100 m w.e.) exposure to 3600 cm^2 of radiogenic lead. Ordinary lead which was simultaneously exposed to mylar foils under the same conditions yielded a background effect of only 3% of the effect from radiogenic lead.

The observed long-range tracks are best explained as resulting from the spontaneous fission of natural, superheavy elements in the suggested element 114 stability region which were coprecipitated with the radiogenic lead. Most likely they are created by a decay chain starting in the proposed Z= 124 to 126 region from a superheavy element which chemically follows uranium.

INTRODUCTION

The stimulation for the work described herein came from the observation of a spontaneous fission activity in several fractions isolated from technical HfO_2 by thermochromatographic separation of chlorides (1). These preliminary data as well as the extended results reported in (2) gave rise to a hypothesis which explained the decaying and growing activities as belonging to some natural decay chain of superheavy elements (analogous to the ^{238}U decay chain, with some branching into spontaneous fission). In the work described in (2) it was found that some of the fractions contained besides some regular (decaying) fission activity also an activity resulting in fission fragments capable of penetrating at least 26μm of mylar foil and producing unusually "fat" tracks. The latter were interpreted as resulting from fission fragments with $54 \leq Z \leq 72$ having energies $150 < E < 250$ MeV per fragment. The fat tracks were attributed to the fission of elements 126 and/or 154. The fission fragments from element 126 could have a total energy of $E_t = 293$ MeV (3), whereas fission of element 154 should have $350 < E_t < 450$ MeV. Therefore the light single fragments from fission of elements 126 and 154 should have $E_{126} \leq 180$ MeV and $E_{154} > 220$ MeV, respectively. On the other hand, the light fission fragments of elements with $110 \leq Z \leq 114$ should have only 130 to 150 MeV per fragment as compared to 90 to 110 MeV per fragment from fission of uranium.

At the onset of this work it was assumed that the postulated natural decay chain of superheavy elements would begin in the Z=126 region and decay, at least partially, by an alpha-chain into the Z=114 region. It was further assumed that elements with $110 \leq Z \leq 114$ should at least partially decay by spontaneous fission with a total energy of 215 to 235 MeV (as theoretically predicted (3)), producing fission fragments detectable on mylar foils. Another assumption was that some amount of element 124 should always be present along with natural uranium. Thus if element 124 is present in uranium ores then element 114 eka-lead should chemically follow radiogenic lead (hereafter called radiolead) as waste material during industrial isolation from uranium ores.

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EXPERIMENTAL PROCEDURES

Source Preparation and Exposure

Based on the hypothesis and assumptions set forth above three sources of 1200 cm^2 each of radiolead were prepared. The radiolead used was in form of $PbSO_4$, obtained from the separation of lead during the processing of uranium ores in the radiochemical laboratories of Arcueille, France. This lead was primarily composed of the isotopes ^{206}Pb, ^{207}Pb, and ^{210}Pb from the decay chains of ^{238}U and ^{235}U in pitchblende, with a small amount of natural Pb also present from the uranium ores. No traces of either the ^{226}Ra or the ^{228}Th decay chains were detected by means of alpha-spectroscopic measurements. The radiolead was spread onto three aluminum foils by means of acetone and dried in air. The source thickness was about 12 mg $PbSO_4/cm^2$. All three sources were placed underground (under 36m soil or ~100 m w.e., i.e. under the shielding equivalent of 100m of water). The sources were covered with 13μm thick double mylar foils. The first mylar foil was used to detect fission fragments from the radiolead, the second to detect background effects.

The exposure lasted a total of 1129 days. Thereafter the mylar foils were removed from the sources and washed with water to remove any residual radiolead. The aboveground exposure of the foils while in contact with radiolead for all operations was 8 hrs.

The fission background created in lead by cosmic rays, primarily muons, still present after a shielding thickness of 36m of soil was also measured. Six 3mm thick Pb-plates were exposed in parallel in similar fashion to double mylar foils in the same location as the radiolead for a period of 1038 days. Exposure of the mylar foils to the Pb-plates was only underground (i.e., no 8 hr above ground exposure as for the radiolead). The plates were infinitely thick for escaping fission fragments ($\gg$20 mg/cm^2). Just prior to the development of the mylar foils the second or background pages of the mylar foils for both radiolead and metallic Pb were exposed to metallic uranium sources for calibration. Exposure was to either one or two sources for periods ranging from 20 to 100 hrs. It defined the efficiency of each foil development in relation to the known fission rate and fission fragment distribution of uranium.

Fission Fragment Detection

After exposure to the uranium sources the mylar foils were developed in 6M NaOH to reveal the fission fragment tracks utilizing the "Mylar Foil Technique" described elsewhere (2). A second development with NH_3 showed the location of each track as a blue dot on ozalid paper. The mylar foil was then microscopically scanned above each blue dot at 200X magnification to identify real fission tracks and separate them from mechanical and related defects. The size of all real tracks was measured at 1000X magnification. Similar microscopic scanning (1000X magnification) of mylar foil page No. 2 allowed measurement of the size of the uranium fission fragment tracks in the same development. The dots found outside the areas covered by the uranium sources on page No. 2 were also scanned and no real tracks were found. None of the fission fragments from the radiolead sources had enough energy to penetrate at least 26μm of mylar.

Efficiency of Fission Fragment Detection and Correction for Background Effects

It can be assumed that the ratio of the number of tracks from fission of uranium in the radiolead source and from the uranium source is proportional to the uranium concentration in radiolead, C_U gU/g$PbSO_4$:

$$\frac{N_{T,\,U\text{-}PbSO_4}}{N_{T,\,U}} = \left(\frac{R_{U\text{-}PbSO_4}}{R_U}\right)\left(\frac{A_{PbSO_4}}{A_U}\right)\left(\frac{t_{PbSO_4}}{t_U}\right)C_U \quad , \tag{1}$$

where $N_{T,\,U\text{-}PbSO_4}$ = number of radiolead fission fragment tracks detected on one foil caused by uranium impurity,

$N_{T,\,U}$ = number of U-fission fragment tracks detected on one foil,

$R_{U\text{-}PbSO_4}$ = range of fission fragments in $PbSO_4$, mg/cm^2,

R_U = range of fission fragments in metallic U, mg/cm^2,

A_{PbSO_4} = surface area of $PbSO_4$, cm^2,

A_U = surface area of U, cm^2,

t_{PbSO_4} = time of exposure to $PbSO_4$, days,

t_U = time of exposure to U, days,

C_U = concentration of impurity (uranium) causing the fission effect, $gU/gPbSO_4$.

Equation (1) is also valid for the muon effect in lead, but all indices U have to be replaced by the index Pb (assuming that the radiolead source thickness is larger than the range of fission fragments in lead, R_{Pb}). The ranges for light fission fragments (110 MeV - ^{88}Sr) in U, Pb and $PbSO_4$ are obtained from (4) as 15.73, 14.17 and 10.88 mg/cm^2, respectively. For the muon fission effect in lead, $C_{Pb} = Pb/PbSO_4 = 0.683$ (concentration of Pb in $PbSO_4$ of radiolead).

Uranium Contamination in the Radiolead

The uranium contents in the radiolead was measured by means of alpha-spectroscopy of a large-area source of radiolead. The latter was dissolved in $HClO_4$, evaporated on a Pt-disc and accurately weighed. The foil was then heated to 600°C (to reduce the ^{210}Po- contamination) and weighed again. The alpha-activity was counted in a standard alpha-chamber attached to a pulse hight analyzer. The amount of uranium present in the radiolead was calculated from the integrated number of alpha-counts in the 4.18 MeV energy region. It was found to be $C_U = 3.85 \times 10^{-5}$ g U/g of radiolead.

RESULTS

A total of 87 tracks were detected on three mylar foils after an exposure of 1129 days to $3600 cm^2$ of radiolead. Similarly, a total of 134 tracks were detected from uranium fission fragments on mylar foil page No. 2 after a 98 hour (3.667 days) exposure to 180 cm^2 of metallic uranium. According to Eq. (1), $C_U = 3.85 \times 10^{-5}$ $gU/gPbSO_4$ should have produced the following detectable fission effect during the 1129 day exposure to 3600 cm^2:

$$N_{T,\,U\text{-}PbSO_4} = \left(\frac{R_{U\text{-}PbSO_4}}{R_U}\right)\left(\frac{A_{PbSO_4}}{A_U}\right)\left(\frac{t_{PbSO_4}}{t_U}\right) N_{T,\,U} \times C_U$$

$$= \left(\frac{10.88}{15.73}\right)\left(\frac{3600}{180}\right)\left(\frac{1129}{3.667}\right) 134 \times 3.85 \times 10^{-5} = 22 \text{ tracks.}$$

The six mylar foils which were exposed in parallel to $7200 cm^2$ of ordinary lead plates (to determine the muon fission effect) in the same location were developed similarly. A total of 10 fission tracks were detected for the 1038 day exposure period. The metallic uranium calibration sources yielded a total of 813 detectable tracks for an exposure period of 150.5 hours to an area of $360 cm^2$.

The metallic uranium sources produced detectable fission fragment tracks at a rate of 2.701/hr and 1.523/hr of exposure to $180 cm^2$ each of mylar foil page No. 2 for the cases of radiolead and ordinary lead, respectively. With these detection efficiencies the background effect due to muon-caused lead fission in the radiolead for the underground location (100 m w.e.) can be determined from Eq. (1) as:

$$N_{T,\,Pb\text{-}PbSO_4} = \left(\frac{R_{PbSO_4}}{R_{Pb}}\right)\left(\frac{A_{PbSO_4}}{A_{Pb}}\right)\left(\frac{t_{PbSO_4}}{t_{Pb}}\right)\left(\frac{1.523}{2.701}\right) N_{T,\,Pb} \times C_{Pb}$$

$$= \left(\frac{10.88}{14.17}\right)\left(\frac{3600}{7200}\right)\left(\frac{1129}{1038}\right)\left(\frac{1.523}{2.701}\right) \quad 10 \times 0.683 = 1.61 \text{ tracks.}$$

It should be noted that it was assumed that the 110 MeV-^{88}Sr ion as determined for lead from (4) also represents the fission fragments from muon fission of lead.

After subtraction of the uranium fission background (22 tracks) and the muon lead fission background (2 tracks) the net fission effect left is 63 tracks. This represents the detected fragments from an unknown, spontaneously-fissioning impurity present in the radiolead.

The microscopic examination of the tracks was carried out at 1000X magnification and one division on the graticule corresponded to 1.44μm. The tracks from radiolead and uranium (exposed to the same mylar foil) differed from each other in two ways. First, no tracks longer than 10.3 divisions were found among the uranium-caused tracks, whereas there were 14 tracks (16.1%) among those from the radiolead the length of which was between 10.5 and 14 divisions. Secondly, the radiolead produced a number of double tracks, i.e. two fission tracks in very close proximity. In some of these pairs the tracks were almost equal in width whereas in others one track was substantially wider than the other. In many of

these cases the two tracks covered an area smaller than 10×10 divisions (14×14μm). In contrast, only 3.7% of the tracks produced by uranium were double tracks. The 10 tracks detected in the lead background experiment somewhat resembled the effect from radiolead (2 were doubles and one was long). In addition to these 10 tracks the background lead also produced 13 fat tracks which resembled the fat tracks observed in (2). These 13 fat tracks might indicate the presence of elements from the proposed stability region near Z = 164. However, none of the fat tracks in this investigation penetrated two mylar foils (26μm). Among the radiolead and the uranium tracks there was only one such fat track in addition to the 87 and 134 regular tracks mentioned above. Table 1 summarizes the results of the track survey including the percentage contribution of each variety of track. Three of the 14 long tracks from radiolead were double tracks and they were also included in the 22 double tracks. Of particular significance appears to be the large fraction of double tracks from radiolead. The last line in the table, denoted as "Unknown Fission in Radiolead", is obtained by subtracting the 22 uranium fission background tracks and the 2 muon lead fission background tracks in the ratio of the track distribution for uranium and lead (B.G.).

TABLE 1 Survey of Tracks

Source	Surface Area cm^2	Total No. of Tracks	Single Tracks	Double Tracks	Long Tracks (>19.5μm)
Radiolead	3600	87	54(62.1%)	22(25.3%)	14(16.1%)
Uranium	180	134	128(95.5%)	5(3.7%)	1(0.8%)
Lead (B.G.)	7200	10	7(70%)	2(20%)	1(10%)
Unknown Fission in Radiolead		63	31(49.2%)	21(33.3%)	14(22.2%)

The distribution of track lengths (measured at 1000X magnification) from radiolead, uranium and lead background is shown in the histogram of Fig. 1. Each double track is treated as two separate tracks.

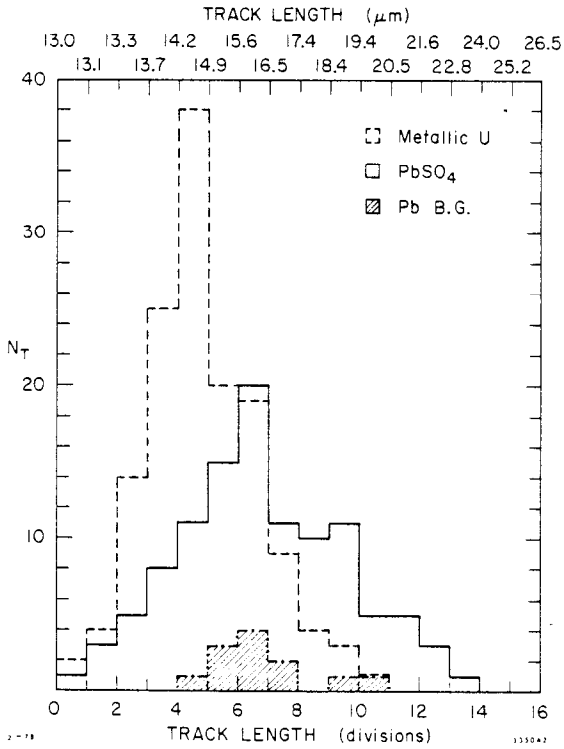

Fig. 1. Histogram of detected number of tracks N_T versus measured track length in μm for metallic uranium (139 tracks), radiolead (109 tracks) and lead background (12 tracks). (Double tracks were counted twice).

Long-Range Fission Fragments From Radiogenic Lead

The distribution of uranium fragment tracks shows a maximum at 5 divisions = 14.9μm length and shows a sharp decrease beyond 8 div. = 17.4μm. Only one track (10.3 div. = 19.7μm) is in the 11 division length range. In contrast, the maximum of the radiolead distribution falls at 7 div. = 16.5μm. Moreover, a secondary maximum is indicated at 10 div. = 19.4μm with a gradual decrease up to a length of 14 div. = 24.0μm. The 14 tracks located between 11 and 14 div. are clearly from fragments having a longer range than the single track from uranium at 10.3 divisions. The lead background track distribution resembles that of the radiolead but with poor statistics.

Some preliminary conclusions can be drawn from the data presented in Fig. 1: The radiolead contains some unknown spontaneously-fissioning element X which produces fission fragments the range of which relative to that of uranium fragments varies as 23.4μm (X): 19.7μm(U) = 1.19:1 (utilizing only the maximum measured length for both, since those are considered tracks from light fission fragments almost parallel to the mylar foil with the full range in the first mylar foil).

Also measured was the maximum diameter or width of all tracks represented in Fig. 1. The arithmetic average diameter for all radiolead tracks was found to be 3.81 div. = 5.49μm. Similarly, the average track diameter from uranium fragments was 3.34 div. = 4.81μm. The track diameters from both radiolead and uranium are statistically distributed primarily in the interval between the "average ± 1 division". The arithmetic average of the maximum track diameters for each track length interval is plotted in Fig. 2. All the long-range fragment tracks from radiolead (length = 9 to 14 div.) show an arithmetic average between 3 and 5 div., i.e. they belong to normal fission fragments with correspondingly high dE/dx along the track. The average diameter or width of the 10 tracks from lead was slightly larger (between 3 and 6 div., average 4.39) due to longer mylar foil development in NaOH. This is in agreement with the previously observed higher detection efficiency for tracks from uranium exposed to the lead background foils (2.703 tracks/hr as compared to 1.523 tracks/hr for the uranium exposed to the radiolead foils).

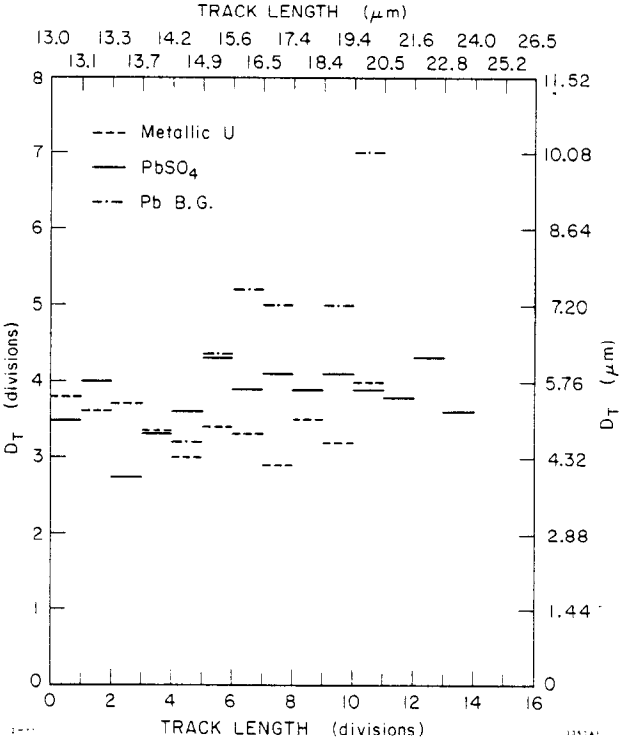

Fig. 2. Maximum diameter or width of tracks, D_T (arithmetic average for each length interval) for 139 tracks from metallic uranium, 109 tracks from radiolead and 12 tracks from lead background.

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ENERGY AND Z OF LONG-RANGE FISSION FRAGMENTS

General Considerations

The measured range R_{ion} (μm) and the maximum track diameter or width, D(μm) were used to estimate the kinetic energy and Z of the long-range fission fragments. The ionization loss dE/dx along the fission fragment track was determined from the following relationship between the leaching rate in NaOH, V_t, and the ionization effect in a plastic, I:

$$V_t = K_1 D = B\left[I(\beta, Z^*)\right]^{1.8} = B\left[K_2 \frac{dE}{dx}(\beta, Z^*)\right]^{1.8} \quad , \tag{2}$$

where K_1, K_2 and B are proportionality constants. Equation (2) was verified for lexan (5) utilizing different ions and by measuring the etching velocity V_t according to the growth of the etched cones along the length of the track in very thick foils. In the investigation reported herein etching progresses simultaneously from both sides of the relatively thin mylar foil. During approximately the first 20 minutes of etching the track becomes a small hole. Thereafter, further growth of the diameter D of such a hole can be assumed to be proportional to the etching velocity V_t as described in Eq. (2).

Any two tracks developed in the same development on the same mylar foil with maximum diameters or widths $D_{1,max}$ and $D_{2,max}$ can be related by their ionization losses $(dE/dx)_{1,max}$ and $(dE/dx)_{2,max}$ according to Eq. (2) in the ratio:

$$D_{1,max}/D_{2,max} = (dE/dx)^{1.8}_{1,max}/(dE/dx)^{1.8}_{2,max} \tag{3}$$

Here $(dE/dx)_{max}$ is the maximum ionization loss along the track producing the maximum diameter or width; it is solely used in all further considerations.

Calibration with Uranium Tracks

The most probable fission fragment having the exact range of light uranium fission fragments was determined first. The latter is obtained from Fig. 1 as 10.3 div. = 19.7μm of mylar (for ρ = 1.4 g/cm^3) or R_U = 2.758 mg mylar/cm^2. The energies for different ions from ^{238}U-fission were calculated using the semiempirical tables given in (4); they are presented in Table 2. For each ion $^{Z_i}A_i$ given in Table 2, the kinetic energy which gives the exact range R_U = 2.758 mg mylar/cm^2 was found (4). The complementary fission fragment was then calculated (assuming Z = 92, A = 238 for ^{238}U-fission) according to the following equation:

$$Z_{c(A_c)} = {}^{Z - Z_i}{}_{(A - A_i)} \quad . \tag{4}$$

The kinetic energy E_{A_c} of $^{Z_c}A_c$ in Table 2 was calculated according to the classical relationship:

$$E_{A_c} = E_{A_i}(A_i/A_c) \quad . \tag{5}$$

The total translational kinetic energy E_{tot} in Table 2 was then simply determined as:

$$E_t = E_{A_i} + E_{A_c} = (A/A_c)E_{A_i} \quad . \tag{6}$$

The value for ^{238}U is the same as for ^{235}U + n (6), namely E_t = 172 MeV (7). Using data given in (7), the average of ^{235}U + n and ^{239}Pu + n can be obtained as E_t = 175 MeV. If these values are compared to the data shown in Table 2, it is evident that for a range of 19.7μm in mylar, only the Rb-ions give the proper total kinetic energy, and that the most likely light uranium fission fragment having a range of 19.7μm is ^{95}Rb. This result is in good agreement with the peak of the light fission fragment mass distribution for ^{238}U which shows a maximum yield at A_i = 95.5 (8). The charges Z = 36 for A = 92 and Z = 37 for A = 95, etc. as given in Table 2 were chosen according to the hypothesis of equal charge displacement of fission fragments and data given in (7). This hypothesis states that the most probable Z_p of a fission fragment with a mass A is that Z which results in the same number of β-decays for both complementary fragments:

$$Z_p(A) = Z_{UCD}(A) + \Delta(A) \quad , \tag{7}$$

where $Z_p(A)$ is the most likely charge of fragment A, $Z_{UCD}(A)$ is the charge of this fragment with "Unchanged Charge Distribution" (UCD) as compared to the fissioning nucleus; and $\Delta(A)$ is a correction factor for Z_{UCD} (usually $-0.7 \leq \Delta(A) \leq -.4$) which was taken as the average of the data for thermal neutron fission of ^{235}U and ^{239}Pu as given in Fig. 11 of (7). The charge Z_p was chosen according to Eq. (7) for all isotopes in Table 2 with the exception of ^{91}Rb. The latter was used for comparison only and its total energy agrees within 2 MeV with that of ^{95}Rb as obtained from Eq. (6). All other isotopes in Table 2 are assumed before neutron emission which is probably correct during the time of flight of the fragments in mylar.

TABLE 2 Energy of Uranium Fission Fragments in Mylar for a Range of R_U=2.758 mg/cm^2

$Z_{i_{A_i}}$	E_{A_i}	$\left(\frac{dE}{dx}\right)_{A_i, max}$	$Z_{c_{A_c}}$	E_{A_c}	E_{total}
	MeV	MeV/(mg/cm^2)		MeV	MeV
$^{88}Se_{34}$	98.7	53.21	$^{150}Ce_{58}$	57.9	156.7
$^{89}Br_{35}$	101.1	54.61	$^{149}La_{57}$	60.4	161.5
$^{92}Kr_{36}$	102.7	56.00	$^{146}Ba_{56}$	64.7	167.4
$^{91}Rb_{37}$	106.4	57.40	$^{147}Cs_{55}$	65.9	172.3
$^{95}Rb_{37}$	104.2	57.40	$^{143}Cs_{55}$	69.2	173.4
$^{97}Sr_{38}$	106.2	58.81	$^{141}Xe_{54}$	73.1	179.3
$^{100}Y_{39}$	107.6	60.21	$^{138}I_{53}$	78.0	185.6

Energy and Z of Fission Fragments from Radiolead

The kinetic energy of different fission fragments from radiolead with a total range R_X = 23.4 μm = 3.276 mg/cm^2 in mylar was calculated analogous to that from uranium fragments above. The data is presented in Table 3. The kinetic energy E_{A_i} was found with the aid of semiempirical tables (4) for each fission fragment (having a charge Z_p chosen as in Table 2) and the total energy, E_t, was calculated. Table 3 gives E_t for five possible fissioning nuclei: ^{238}U, $^{274}110$, $^{278}114$, $^{294}110$, and $^{298}114$; it is denoted as $^{238}E_t$, $^{274}E_t$, $^{278}E_t$, $^{294}E_t$, and $^{298}E_t$, respectively. The choice of these nuclei was based on the assumption of closed neutron shells at either N=184 ($^{298}114$ and $^{294}110$) or N=164 ($^{278}114$ and $^{274}110$). This choice is meant to demonstrate the sensitivity of E_t to the mass and N of the fissioning nucleus.

Based on the ionization loss, $(dE/dx)_{max}$ i.e., the maximum diameter or width of the tracks, the data presented in Table 3 allows selection of the most probable Z for fragments having a range of 23.4 μm in mylar. Figure 2 demonstrates that tracks from long-range fission fragments (11 to 14 div.) have a regular diameter, close to the average of 3.81 divisions. By comparision the uranium tracks have an average maximum track diameter or width of 3.34 div., i.e., $D_{X,max} \simeq 1.14\ D_{U,max}$ (where D_X is the average maximum diameter of all tracks from radiolead). On the other hand, the average maximum diameter or width of the 14 long-range tracks only is 3.93 div., i.e. $D_{X,max} = (3.93/3.34) D_{U,max} = 1.18\ D_{U,max}$. If this ratio is valid then the use of Eq. (3) for the ^{95}Rb-fragment from fission of uranium yields:

$$D_{X,max}/D_{Rb,max} = 1.18 = (dE/dx)^{1.8}_{X,max}/(57.40)^{1.8} ,$$

or

$$(dE/dx)_{X,max} = 62.93 \text{ MeV/(mg/cm}^2\text{) of mylar.}$$

This $(dE/dx)_{max}$ corresponds to ^{103}Nb with E_t equal to 220.6, 222.2, 229.3, 231.3 and 254.5 for fission of $^{298}114$, $^{294}110$, $^{278}114$, $^{274}110$ and ^{238}U, respectively. If an error band of $\pm$ 0.5 divisions is added to the average located at 3.93 div. (this covers possible diameter inhomogeneity due to track development) then the upper limit for the maximum ionization loss is obtained from Eq. (3) as:

TABLE 3 Energy of Fission Fragments From Radiolead for a Range R_X = 3.276 mg/cm^2 in mylar

$Z_{i_{A_i}}$	E_{A_i}	$\frac{dE}{dx}_{A_i, max}$	$^{238}E_t$	$^{274}E_t$	$^{278}E_t$	$^{294}E_t$	$^{298}E_t$
	MeV	MeV/(mg/cm^2)	MeV	MeV	MeV	MeV	MeV
$^{92}Kr_{36}$	131.1	55.98	213.7	197.4	195.9	190.8	189.6
$^{97}Sr_{38}$	136.2	58.81	229.9	210.9	209.2	203.3	202.0
$^{102}Zr_{40}$	141.1	61.61	247.0	224.8	222.9	216.1	214.6
$^{103}Nb_{41}$	144.4	63.01	254.5	231.3	229.3	222.2	220.6
$^{108}Mo_{42}$	145.0	64.39	265.5	239.4	237.2	229.3	227.5
$^{110}Tc_{43}$	147.7	65.77	274.7	246.8	244.4	236.0	234.2
$^{113}Ru_{44}$	149.7	67.17	285.0	254.8	252.2	243.2	241.2
$^{118}Pd_{46}$	154.3	69.95	306.0	271.0	268.0	257.7	255.4
$^{125}Cd_{48}$	157.5	72.72	331.7	289.7	286.2	274.0	271.3
$^{130}Sn_{50}$	161.6	75.46	356.1	307.5	303.6	289.7	286.7

$$(dE/dx)_{u\text{-}max} = [(4.43/3.34)(57.40)^{1.8}]^{1/1.8} = 67.15 \text{ MeV/(mg/cm}^2) \quad .$$

As reasonable lower limit for the maximum ionization loss, $(dE/dx)_{\ell\text{-max}}$, the average diameter or width of all tracks from uranium and radiolead can be used:

$$(dE/dx)_{\ell\text{-max}} = [(3.81/3.34)(57.40)^{1.8}]^{1/1.8} = 61.60 \text{ MeV/(mg/cm}^2) \quad .$$

From this consideration the upper limit for Z in Table 3 is obtained as Z=44 (^{113}Ru fragment) with total kinetic energies of 241.2, 243.2, 252.2, 254.8, and 285.0 MeV for fission of $^{298}114$, $^{294}110$, $^{278}114$, $^{274}110$ and ^{238}U, respectively. As a lower limit the ^{102}Zr fragment is obtained with similar energies given in Table 3.

The fission of ^{238}U with $247 \leq E_t \leq 285$ MeV (within the lower and upper limits) is clearly excluded since its total fission energy is known, $^{238}E_t$ = 172 MeV (the same as for ^{235}U + n).

The most probable fission fragment in Table 3, ^{103}Nb, resulting from the fission of an isotope with a total energy of $^{294}E_t$ = 222.2 MeV is in good agreement with theoretical predictions of 216 MeV (3) and 216.5 MeV (9). The latter paper gives a semiempirical formula for the prediction of the total energy:

$$E_t = 0.119\ Z^2/A^{1/3} \quad . \qquad (8)$$

Equation (8) yields $^{274}E_t$ = 221.7 MeV or $^{278}E_t$ = 237.0 MeV for $^{274}110$ or $^{278}114$, respectively, whereas ^{103}Nb, the most probable fragment given in Table 3, resulted in 231.3 or 229.3 MeV, respectively. The lower limit as represented by ^{102}Zr in Table 3 shows surprisingly exact agreement in $^{294}E_t$ = 216.1 MeV with predictions of 216 and 216.5 MeV (3 and 9). Also $^{274}E_t$ = 224.8 MeV for ^{102}Zr in Table 3 is very close to the value of 221.7 MeV as calculated with Eq. (8).

The most likely fission fragment (^{103}Nb) and the fragment representing the lower limit (^{102}Zr) as given in Table 3 lend strong support to the following conclusion: the tracks observed from radiolead are due to spontaneous fission of either ^{294}110 or ^{274}110 which are the result of two alpha-decays of the corresponding isotopes of element 114, eka-lead. On the other hand, the predictions for $^{298}E_t$ are 235 MeV (3) and 231.5 MeV Eq. (8). Both values are in good agreement with $^{298}E_t$ = 234.2 as given for ^{110}Tc in Table 3. Similarly, $^{278}E_t$ = 237.2 MeV for fragment ^{108}Mo agrees exactly with the value of 237 MeV as predicted by Eq. (8) for fission of ^{278}114. The $(dE/dx)_{max}$ for the ^{108}Mo and ^{110}Tc fragments is still below the upper limit of 67.17 MeV/(mg/cm^2), as determined above. This leads to the further conclusion that the experimental data reported herein can also be well explained by fission of ^{298}114 or ^{278}114. However, it should be stressed that a clear distinction between fragments from ^{298}114 or from ^{294}110, ^{278}114, ^{274}110 is not possible within the precision of this experiment.

DOUBLE TRACKS

A number of double tracks were reported in Table 1 (22 from radiolead, 5 from uranium, 2 from lead). Some of these could be due to scattering of the fission fragment and resulting recoil of the ^{16}O nucleus in mylar. This was probably the case for four doubles from U and two from radiolead. These doubles are composed of one track of regular width and one that is significantly narrower. In 20 other cases from radiolead both tracks were rather similar in size and often touched each other, suggesting a common vertex. In some of these events a single track entered into the mylar foil and branched into two tracks of equal size. In other events the two tracks were separate and not in the same plane. In these two latter cases the effect could be explained as either fission in flight of a heavy fragment or fission after the heavy fragment had stopped (i.e., cascade fission or secondary fission of a heavy fragment during the exposure period).

In about 50% of the double events the two fragments were coplanar. This indicates either ternary fission as previously theoretically predicted (3) or a heavy recoil, as for example of ^{32}S from $PbSO_4$, with ^{32}S appearing as second fragment.

It is concluded that at least some of the events resulted from three fission fragments, emitted simultaneously or in cascade. This suggests the presence of a superheavy element in radiolead. Coincidence measurements are necessary to verify ternary fissions.

DISCUSSION OF RESULTS

The observed long-range tracks from radiolead are most probably the result of the spontaneous fission of a superheavy element from the predicted stability region near Z=114. The effect can be explained by the presence of element 114 having long half-life in uranium ores which coprecipitated with radiolead during chemical separation. If this is the case, it is difficult to understand why natural lead did not yield a significant number of long-range tracks under similar exposure conditions. Element 114 or eka-lead should geochemically follow lead and not uranium or vanadium (in pitchblende or carnotite ores). The presence of element 120 in radiolead (which also should precipitate well with sulfates) is excluded since its chemical homologue, Ra, was not contained. On the other hand, the observed effect can well be explained if the hypothesis is accepted that a long eka-uranium or eka-plutonium chain exists in nature, decaying predominantly by alpha-emission. In this case eka-uranium should follow uranium well. Element 126, the mother element of eka-uranium, could also behave chemically very similar to uranium. This is plausible if the stable valencies of samarium (3+ and the less stable 2+) are compared with its chemical homologue in the actinide series, namely plutonium (3+, 4+ and 6+). In a shift from Sm to Pu the oxidation potential for the metal oxide reaction $MeO_2^{2+} \rightarrow Me^{4+}$ decreases from large positive values (for the nonexistant SmO_2^{2+}) to + 1.052V for PuO_2^{2+}. The eka-Pu in the superactinide series should again have an even lower oxidation potential than Pu for the formation of (126) $O_2^{2+} \rightarrow 126^{4+}$; it might be close to the oxidation potential of + 0.334V for $UO_2^{2+} \rightarrow U^{4+}$. This means that element 126 could be most stable as a hexavalent (126) O_2^{2+}-ion and it could chemically and geochemically closely follow uranium. In this case element 114 can be expected to be found accompanying radiolead isolated from uranium ores as it appears to do in this investigation.

The present results support the existence of a long alpha-decay chain starting in the region of stability near 126-124 which was earlier postulated in general form (1, 2). The detection ratio as defined in Eq. (1) can be related to the fission half-lives of element X and uranium as follows:

$$T_{\frac{1}{2}(X\text{-}PbSO_4)} = \left(\frac{R_{X\text{-}PbSO_4}}{R_U}\right)\left(\frac{N_{T,U}}{N_{T,X\text{-}PbSO_4}}\right)\left(\frac{A_{PbSO_4}}{A_U}\right)\left(\frac{t_{PbSO_4}}{t_U}\right) T_{\frac{1}{2}(^{238}U)} \quad , \tag{9}$$

where $T_{\frac{1}{2}(X\text{-}PbSO_4)}$ = apparent half-life for fission of element X in radiolead = $T_{\frac{1}{2}(X)}/C_X$,

$T_{\frac{1}{2}(^{238}U)} = 6.65 \times 10^{15}$ years (2) ,

$T_{\frac{1}{2}(X)}$ = real half-life of element X ,

C_X = concentration of element X in radiolead .

Thus Eq. (9) allows calculation of the apparent half-life for spontaneous fission of element 114 in radiolead. If 63 (of 87 observed) fragments are assigned to element 114, the apparent half-life becomes

$$T_{\frac{1}{2}(X\text{-}PbSO_4)} = \left(\frac{10.88}{15.73}\right)\left(\frac{134}{63}\right)\left(\frac{3600}{180}\right)\left(\frac{1129}{3.667}\right) \times 6.65 \times 10^{15} = 6 \times 10^{19} \text{ years.}$$

This value corresponds to a concentration of element 114 of 1.7×10^{-11} g/gU in uranium ores (if $T_{\frac{1}{2}}(114)$ is assumed to be 10^9 years). It is possible that element 124 is of similar concentration in uranium ore (assuming $T_{\frac{1}{2}}(124) \geq 10^9$ years). If, however, the half-life of element 114 is shorter (of the order of years) than that of element 124, then the weight concentration of the former in radiolead might be much less than 1×10^{-11} g/g. Based on the data presented herein, the authors feel that chemical concentration and separation on a kilogram scale could lead to isolation of element 114 in quantities sufficiently large to allow accurate fission energy measurements, fission-neutron coincidence counting and mass spectrometric analysis. Based on the data presented herein the counting rate of element 114 in radiolead is 80 fissions/(kg-day). Whether or not element 114 shows a high neutron multiplicity, it should be directly observable from kilogram quantities of radiolead measured underground.

Samples of radiolead and ordinary lead were neutron irradiated (10). Some alpha-activity was observed in the mass-separated radiolead at mass position 292. No such activity was found in the ordinary lead and neither samples showed any unknown fission activity. In light of the results reported herein it cannot be ruled out that this alpha-activity originated from a more short-lived isotope of element 114 produced by neutron-irradiation of the practically stable element 114 in radiolead. If this were true, then the spontaneous fission activity observed in this paper represents perhaps only a small fission branch in the decay scheme of element 114. Thus the latter could be present in radiolead at concentrations substantially higher than 10^{-11}g/g. Further investigation of neutron-irradiated lead is certainly warranted.

A high neutron multiplicity was recently reported in samples from the Allende meteorite (11). It was assumed to have originated from a superheavy element in the region from Z=112 to 118. Based on an assumed $T_{\frac{1}{2}} = 10^9$ years the concentration was between 3×10^{-14}g/g and 3×10^{-15}g/g. The observed effect was extremely low and corresponded to 5 to 50 fissions/(kg-year), or 10^{-3} of the effect observed from radiolead in this paper. This low effect, if at all real, could be understood within the framework of the present work. The meteorite contained very little uranium, only 0.02 ppm (13). This agrees with known data of 0.01 - 0.02 ppm for average concentrations of U in meteorites as for example reported in Fig. 11 of (12). The meteorites are assumed to be primordial matter from which also the solar system and thus the earth were formed. In the geochemical concentration process uranium was transported to the earth crust and concentrated. This resulted in an average uranium concentration of 3 to 5 ppm in the crust, whereas the core was depleted in U to a level of <0.006 ppm (12). Further concentration takes place in the crust, up to 6×10^5ppm in pitchblende. Based on the data reported herein it can be said that probably any uranium contains some small amount of element 124 or 126 both of which chemically could resemble uranium. From this and the proposed alpha-decay chain it can be assumed that the concentration of element 114 in some ores is related to the concentration of uranium in the ore, for example at a ratio of $\simeq 10^{-11}$g of element 114/g of uranium (yielding 80 fissions/day from 1 kg of radiolead). Using this ratio for the Allende meteorite (0.02 ppm U) the spontaneous fission effect in 1kg due to element 114 (produced from the proposed element 124 decay chain following uranium) should be extremely low: 2×10^{-8} gU/g $\times$ 80 fissions/(kg-day) $\times$ 365 days = 5.8×10^{-4} fissions/(kg-year). The observed value was 5 to 50 fissions/(kg-year) indicating that the meteoritic material contained perhaps some amount of superheavy material unrelated to the element 124-126 decay chain and present in the original universal material before geochemical concentration. The absolute counting rates in the

experiment described in (11) are very low. Based on the considerations set forth above, it appears that the extensive efforts reported in (11, 13) to confirm the existence of natural super-heavy elements are devoted to less than optimal materials. Better and faster results can be obtained if geochemical and other concentration processes are taken advantage of.

CONCLUSIONS

Long-range fission fragments emitted from radiolead sources exposed on mylar foils in the underground were observed. Ordinary lead sources were measured in parallel under the same conditions. They produced a very small effect; only about 3% of the spontaneous fission effect from radiolead can be assigned to ordinary lead underground fission (caused by muons at the 100 m.w.e. level).

The fission fragments from radiolead have a range 19% longer than the fission fragments from uranium exposed on the same mylar foils. Analysis of the length and width or diameter of the tracks yielded the conclusion that the observed long-range fragments belong to fragments in the range between ^{102}Zr and ^{113}Ru. They originated from fission reactions with a total kinetic energy between 215 - 255 MeV. This energy range coincides with that predicted for elements in the proposed region of stability from Z=110 to 114.

A number of cases with two tracks were also observed indicating the possibility of ternary fission. It was concluded that either element 126 and/or 124 are or were present in uranium ore after its formation at concentrations of $\sim 10^{-11}$g/g and that they decay by an alpha-decay chain to the proposed region of stability near Z=114. The results strongly support the postulated hypothesis of the existence of such an alpha-decay chain in the region of 126-124 and they extend it to the region near Z=114. Another, less likely, explanation is that element 114 was present with some other elements (which later formed the uranium ores) at the time of the last nucleosynthesis.

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