and 8.007 MeV. A more detailed account of the energy levels in ^{251}Fm populated by the α-particle decay of ^{255}No is the subject of a future publication.[9]

Based on our experience with the identification of nobelium, we feel that a conclusive atomic-number identification can be obtained with the observation of far fewer coincident events than were recorded in this experiment. We are currently applying this technique to the identification and study of α-active isotopes of transnobelium elements.

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Search for Superheavy Elements in Terrestrial Matter

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Negative results have been obtained in a search for spontaneously fissioning superheavy nuclei by neutron counting of a large variety of samples. An upper limit of about 10^{-11} g/g is estimated for the presence of such nuclei with a half-life of about 10^9 yr.

Extensive discussions on the physical[1] and chemical properties[2] of superheavy elements began after Myers and Swiatecki[3] pointed out that shell effects may lead to an island of relative stability beyond the present periodic table and after Meldner and Röper calculated that the next shell closure after 82 should probably occur at proton number 114. Detailed lifetime calculations were carried out for superheavy nuclei with even proton numbers by Nilsson[5,6] and Greiner[7] and their collaborators, and it was emphasized that additional stability is expected for odd-Z and/or odd-N nuclei.[7,8] These studies indicate for certain superheavy nuclei half-lives sufficiently long[5] for survival throughout geologic time, i.e., $T_{1/2} \gtrsim 10^8$ yr, and for occurrence in the cosmic radiation, $T_{1/2} \gtrsim 10^5$ yr, provided that such nuclei are produced in nucleosynthesis. The possibility of such production is still a matter of controversy.[1,9]

First attempts to detect superheavy elements in terrestrial matter were reported by Thompson *et al.*[10] and Flerov and Perelygin.[11] Taking into account the position of these elements in the periodic table,[2,12,13] Thompson *et al.* searched for element 110, eka-platinum, in platinum ores, but obtained negative results with a variety of detection methods. In contrast, Flerov *et al.* found positive evidence for element 114, eka-lead, in some lead-bearing samples by the observation of fission tracks in plastics[11] and glasses,[11,14] and of strongly ionizing events in proportional counters.[15] If these fission events were due to an element-114 isotope with a half-life of 1×10^9 yr, its concentration would amount to 5×10^{-13}-2×10^{-12} g per gram of lead.

Other searches in terrestrial material using these and other, more indirect, techniques produced negative results, as is reviewed in Ref. 12 and by Herrmann and Seyb,[16] and Flerov, Druin, and Pleve.[17] Superheavy elements have, however, been discussed as the explanation for some as yet unexplained evidence for either existing or extinct natural radioactivities.[12,16,17] Evidence for the occurrence of such elements in the cosmic radiation has also been reported.[18,19]

In the measurements to be summarized here, a more general approach was followed: First, we did not concentrate on a single element but considered the whole region from element 108, eka-osmium, to element 115, eka-bismuth. Second, we selected the samples not only according to their content in homologous elements but also according to general geochemical rules, and included by-products from the industrial treatment of pertinent material. This approach was chosen since the element of maximum stability cannot safely be predicted, and since the superheavy elements may not necessarily separate together with their homologues due to differences in chemical behavior.[2,19]

Our work is based on a search for spontaneous fission events. Since this decay mode is extremely rare among known natural radionuclides, it should be well suited for detecting superheavy nuclei. Even if the superheavy nuclide decays by alpha emission, the resulting decay chain should soon terminate in a short-lived, fissioning descendant present in radioactive equilibrium.[6,7] In view of the large variety and number of samples to be counted, we did not attempt to detect fission fragments but, rather, the neutrons accompanying the fission process.

This technique permits the counting of almost any kind of sample without special preparation. The drawback of neutron detection, its low efficiency, is counterbalanced by the large sample weight allowable (tens of kilograms) and by the large number of prompt neutrons (about ten) expected to be emitted in fission of superheavy nuclei.[20] Hence, with the neutron counter described in the next section, a concentration of 10^{-11} g/g of such nuclei should be detectable during a counting time of 2 days, which was typical for our work. For such short counting times, this detection limit compares to that achievable with other techniques. The sensitivity could be improved either by the detection of multiple neutron emission with counters operated below ground[21] or by using the "spinner" rotation counter[22]; but we felt that, for short counting times, the gain was not great enough to make these more elaborate techniques much superior to the simple one applied here.

We have used a neutron counter consisting of six ^{3}He counting tubes with an active length of 12 cm and a filling pressure of 4 atm. Two banks of three tubes were embedded in paraffin and the samples placed in between. Detection of γ rays was suppressed by pulse-height discrimination. After appropriate shielding against neutrons, a background of 60 counts/h was recorded at ground level. The detection efficiency for a single neutron was determined by counting at various positions a calibrated americium-lithium neutron source and averaging the results over the whole volume, and by counting spontaneous fission neutrons emitted from large amounts of ^{238}U. Calibrations involving the neutron source together with typical samples showed that self-absorption effects do not decrease the counting efficiency by more than a factor of 2.

If the 3σ error of the background is considered as the lowest detectable activity, a detection limit of 5×10^{-12} g/g is estimated for a superheavy nuclide of 1×10^9 yr half-life when counting a 30 kg sample for 48 h. This estimate is based on a counting efficiency of 1.5% for a single neutron, on a neutron multiplicity of ten neutrons per fission, and on a background of 60 counts/h. For longer-lived nuclides the limit increases proportionally to the half-life. In the case of metallic samples of the heaviest elements, the detection limit increases to 7×10^{-12} g/g because of the additional background described below.

In total, 102 samples were counted, falling into the following categories: (i) sulphidic ores and concentrates (36 samples), e.g., of lead (six), mercury (five), molybdenum (six), zinc (five), and copper (five); (ii) by-products from the industrial treatment of such ores (25), like flying ashes (nine) and slags (six); (iii) raw and pure metals (29), e.g., gold, platinum, and other noble metals (twelve), lead and adjacent metals (nine); (iv) miscellaneous samples (eleven), like manganese nodules (five) and wastes from the treatment of pollucites (two). Manganese nodules from the sea floor were included since they may collect superheavy nuclei accreting to the Earth from the cosmic radiation.[23] Most samples were available in tens of kilograms.

Counting rates well above background, namely 20 to 50 counts/h, were recorded with metallic samples of the elements platinum to bismuth.

Table I. Results of neutron counting of heavy metals.

Sample	Amount (kg)	Specific counting rate (counts/h mole) [a]
Osmium	0.63	0.3 ±0.8
Platinum	14.8	0.23 ±0.04
Gold	50.0	0.235±0.012
Mercury	13.0	0.22 ±0.05
Thallium	20.0	0.20 ±0.03
Lead	33.0	0.268±0.018
Bismuth	19.3	0.22 ±0.03
Weighted mean		0.240±0.017

[a]With statistical 2σ error.

However, the specific counting rates of these six elements were found to be identical within the statistical errors, as is shown in Table I for representative samples. Since it is very unlikely that all the samples contain exactly the same amounts of a spontaneously fissioning element, the neutrons have to be attributed to another source, most likely to high-energy spallation reactions induced in the samples by cosmic rays. This interpretation is supported by a comparison of the counting rates found for these and other elements with their total spallation cross sections estimated from cross-section systematics.[24] After normalization to the counting rate observed with silver, the relative cross sections fit the counting data over the whole region of elements studied, as is shown in Fig. 1. Uranium and thorium would contribute to the counting rates only in concentrations exceeding 0.01 and 1%, respectively, as can be estimated from data given in Fig. 1.

The maximum excess activity of the elements listed in Table I amounts to about 0.07 counts/h mole, except for osmium. Assuming the emission of two neutrons per fission, one obtains a lower limit of 2×10^{19} yr for the spontaneous fission half-life of these elements.

For the rest of the samples, the counting rates were within the errors identical with the background or, if a slight surplus was recorded, this effect could be attributed to spallation neutrons produced in main constituents of the samples. The data in Fig. 1 were used for the estimate of neutron production rates.

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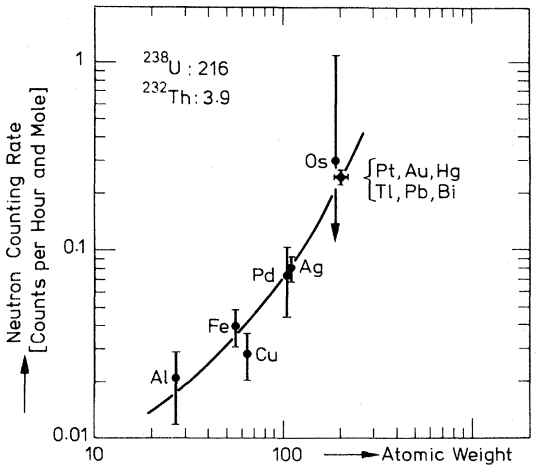

FIG. 1. Comparison of the neutron counting rates of metallic samples (points) with the total cross sections for high-energy spallation reactions (solid line) calculated from cross-section systematics (Ref. 24) and normalized to the counting rate of silver.

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N-Shell Conversion Electrons from the 2.33-keV Transition in ^{205}Pb

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We have measured the N_{II} and N_{III} conversion electrons from the ≈2.33-keV transition in ^{205}Pb with a $\pi\sqrt{2}$, iron-free, double-focusing β-ray spectrometer and have determined the energy of this transition to be 2.328 ± 0.007 keV. We obtain an N_{II}/N_{III} ratio of 0.70 ± 0.25 for this transition and a ratio $(N_{II}+N_{III})/L_I$, where the L_I is from the 26.22-keV $M2$ isomeric transition in ^{205}Pb, of $0.48^{+0.15}_{-0.05}$. These are in very good agreement with predicted conversion ratios and represent the first test of theoretical calculations for the N shell at such low energies.

The Michigan State University (MSU) $\pi\sqrt{2}$, iron-free, double-focusing β-ray spectrometer has been used to make measurements on the very low-energy portions of the electron spectra resulting from the EC decay of ^{205}Bi and ^{206}Bi. These were used to obtain the N-conversion spectrum for the 2.33-keV transition in ^{205}Pb. At ^{205}Pb the $p_{1/2}$ and $f_{5/2}$ neutron orbits have crossed and lie very close together, and indirect evidence[1-3] has established the 2.33-keV transition as being an $E2$ from the (predominantly $\nu p_{1/2}$) $\frac{1}{2}^-$ first excited state to the (predominantly $\nu f_{5/2}$) $\frac{5}{2}^-$ ground state. Measurements on conversion electrons in this energy range are extremely rare, and the present experiments represent the first direct observation of this transition. In fact, we believe that this is only the second time that an ≈2-keV transition has been measured,[4] and it is the first time such measurements have been made on an element with a high enough Z that the N subshell ratios could be used to test theoretical conversion coefficients directly.

The sources used for this study were prepared by bombarding mass-separated Pb isotopes (obtained from Oak Ridge National Laboratory) with protons from the MSU sector-focused cyclotron. The reactions ^{206}Pb$(p, 2n)^{205}$Bi ($t_{1/2}$ = 15.3 day) and ^{208}Pb$(p, 3n)^{206}$Bi ($t_{1/2}$ = 6.24 day), using 19- and 25-MeV protons, respectively, produced clean sources of the desired activities.

Standard precipitation and anion-exchange techniques were used to retrieve the active material from the targets. However, we found it necessary to take extreme precautions in order to minimize even trace amounts of impurities that would cause poor quality sources.[5] All glassware was replaced with quartz that had been specially washed and leached for several days in low-conductivity water (deionized, distilled, doubly deionized, distilled) whose purity was checked by measuring its conductivity. The acids used (HCl and HNO_3) were either "Ultrex" commercial grade[6] or were prepared by bubbling HCl and NO_2 gases through low-conductivity wa-