Identification of the Atomic Number of Nobelium by an X-Ray Technique

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A daughter x-ray identification scheme, in the original spirit of Moseley, has been applied to the identification of element 102, nobelium. The identification scheme is based on the coincident observation of K-series x rays from the daughter element, fermium in this case, with the α particle from the decay of the parent element, nobelium. We believe the technique provides an unequivocal identification of atomic number for the transfermium elements.

Identification of the atomic number of an element by its characteristic K-series x rays was first applied by Moseley[1] in establishing parts of the periodic table, and this method has been more recently applied to confirming the identification of the artificially produced elements, technetium ($Z=43$) and promethium ($Z=61$).[2] In all these cases, however, a large enough sample of the material was available so that x-ray emission could be induced by either electron bombardment or fluorescent excitation. Unfortunately, the direct application of this x-ray method to the identification of the transfermium elements is beyond present experimental capabilities as these elements are produced using heavy-ion accelerators and only very small samples (consisting of a few atoms) can be accumulated. For this reason, atomic number assignments for the transfermium elements have been based largely upon evidence from transmutation studies (such as genetic-linkage experiments[3]), excitation-function analyses, studies of nuclear systematics, and studies of chemical properties.[4] However, the x-ray method of Moseley can be extended, with a simple modification, to the study of many of the transfermium elements, even though many of these elements must be examined one atom at a time. This extended x-ray method is much more general for the identification of atomic number than those just mentioned because it is based on the measurement of a property which is closely related to the atomic number, the x-ray energies. And, since adequately accurate theoretical predictions already exist for the x-ray energies[5] of the transfermium elements, unambiguous atomic-number assignments can be made for these elements.

Our technique relies on the coincident observation of characteristic K-series x rays from the daughter element and α particles from the α decay of the parent element. This will be possible if the α decay of the parent proceeds to excited nuclear states in the daughter nucleus which subsequently de-excite by the internal conversion process. The characteristic K-series x rays of the daughter element then result from atomic rearrangements following K-shell internal conversion. Since the observed α particle has $Z=2$, the identification of K-series x rays from the daughter element (with atomic number $Z-2$) provides definitive proof of the existence of the parent element with atomic number Z.

Although experimental K-series x-ray energies or electron binding energies are not available for the elements in question ($Z \geq 100$), calculated values for $96 \leq Z \leq 120$ have been reported by Carlson, Nestor, Malik, and Tucker.[5] For the elements $Z=96$ through 99, our measurements[6] agree with their calculations to within 40 eV. Since, for Z near 100, a change in Z by one unit corresponds to a change in energy of about 3000 eV for a given K-series x ray, the accuracy of the calculated values is more than sufficient for the assignment of a unique atomic number. A comparison of calculated and experimental $K\alpha_2$ and $K\alpha_1$ x-ray energies for the elements $Z=96$-106 is given in Table I. In addition, K-series x rays can be distinguished from γ rays and other x rays on the basis of the relative line intensities within the corresponding K-series x-ray group ($K\alpha_2$, $K\alpha_1$, $K\beta_3$, $K\beta_1$, etc.).[6]

We produced the 200-sec ^{255}No isotope in the reaction ^{249}Cf(^{12}C, $\alpha 2n$) using a beam of $^{12}C^{+4}$ ions accelerated in the Oak Ridge isochronous cyclotron to an energy of 77.4 MeV. After a loss of 5 MeV in the 2.5-mg/cm^2 Be foil supporting the 350-μg/cm^2 ^{249}Cf target, the incident ^{12}C beam had the proper energy for maximizing the reaction yield. The recoiling reaction products were thermalized in a helium-filled, gas-jet collection assembly, pumped through a small orifice, and deposited on a 23-mg/cm^2 Ni disk. After a 6-min bombardment cycle, the disk was pneumatically transferred a distance of about 30 m to a counting

Table I. Calculated and experimental $K\alpha$ x-ray energies for $Z \geq 96$.

Element	$K_{\alpha 2}$ Calculated[a]	$K_{\alpha 2}$ Experimental	$K_{\alpha 1}$ Calculated[a]	$K_{\alpha 1}$ Experimental
Curium 96	104.607	104.589 ± 0.005	109.287	109.273 ± 0.005
Berkelium 97	107.214	107.165 ± 0.006	112.146	112.112 ± 0.006
Californium 98	109.864	109.818 ± 0.005	115.060	115.031 ± 0.005
Einsteinium 99	112.581	112.501 ± 0.010	118.057	118.018 ± 0.010
Fermium 100	115.320	(115.280 ± 0.090)	121.090	(121.070 ± 0.090)
Mendelevium 101	118.137		124.217	
Nobelium 102	121.020		127.424	
Lawrencium 103	123.939		130.684	
104	126.916		134.019	
105	129.951		137.431	
106	133.033		140.905	

[a]Ref. 5.

assembly. During the accumulation of a total integrated current of $^{12}C^{+6}$ ions of about 60 μA h, approximately 180 bombardment-counting cycles were completed, and about 7000 α particles from the decay of ^{255}No were detected.

The collection disk was positioned in the counting assembly between a surface-barrier detector for α-particle detection and a low-energy photon spectrometer for x-ray and γ-ray detection. The α detector, a 200-mm^2 × 300-μm Si(Au) surface-barrier detector, subtended a solid angle of 23.4% of 4π sr. The space between the detector and the disk was filled with He, and the measured energy resolution, in the He gas, was about 35 keV for the 5.806-MeV α group from the decay of ^{244}Cm. The photon spectrometer, a 16-mm^2 × 5.3-mm planar Ge(Li) detector, was equipped with a 11.7-mg/cm^2 Be window. Under our experimental conditions, for 122-keV photons, the measured photon-energy resolution (full width at half-maximum) was 509 eV, and the absolute efficiency was 4.65%.

The time interval between the α-particle event and the x-ray event was measured with a time-to-amplitude converter. An α pulse started the converter, and a photon pulse, occurring in the time range 0-100 μsec after the α-particle pulse, stopped the converter. Signals suitable for timing purposes were derived using amplitude- and rise-time-compensated timing discriminators. The three linear pulses constituting a decay event, α-particle energy, photon energy, and time correlation, were temporarily held in linear stretchers; then they were digitized sequentially by a single 8192-channel analog-to-digital converter and transferred to a computer using a fast, on-line, data-acquisition system.[7] All α-particle pulses were recorded, whether accompanied by a coincident photon pulse or not. The digitized events were scanned and processed after the experiment to yield information on the decay of ^{255}No.

The energy spectrum for the single α particles is shown in Fig. 1, and the α groups attributed to the decay of ^{255}No have been indicated. This spectrum is essentially identical to the spectrum reported recently by Eskola, Eskola, Nurmia, and Ghiorso[8] for the same reaction. In Fig. 2 is shown a portion of the photon energy spectrum for the photons observed in coincidence with all of the ^{255}No α groups. The observed lines have been identified as corresponding to Fm K-series x rays for the following reasons: The energies of these lines are in excellent agreement with values calculated for Fm K-series x rays (see Table I), and the relative line intensities are in good agreement with those expected for Fm K-series x rays. Since these x rays are uniquely

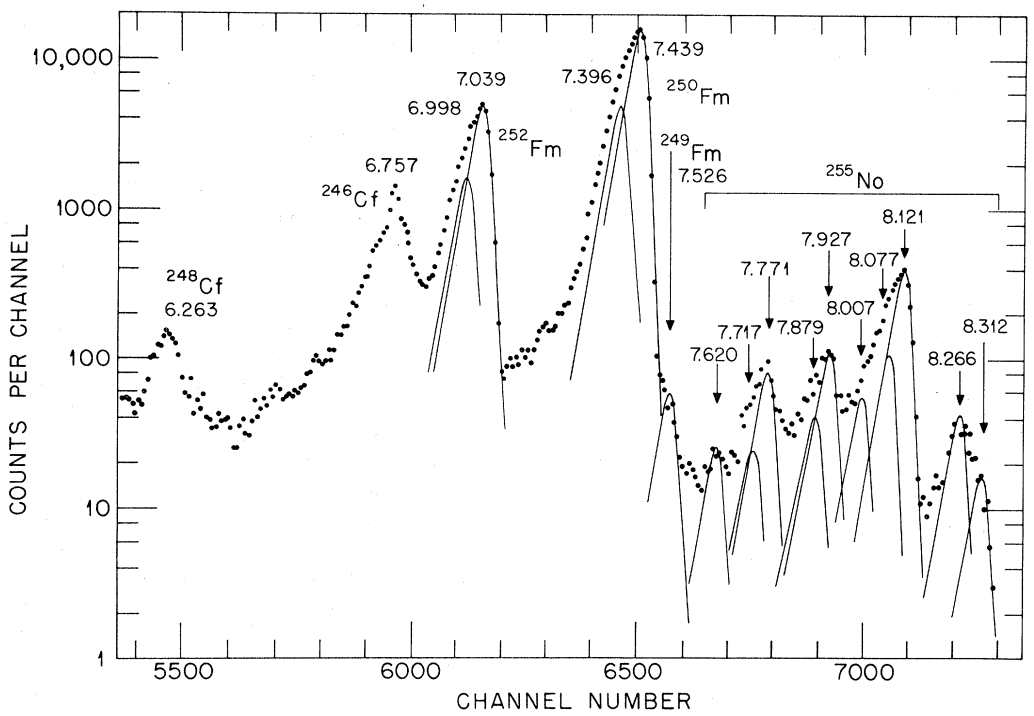

FIG. 1. α-particle energy spectrum for activities produced in the bombardment of ^{240}Cf with 72.3-MeV $^{12}C^{+4}$ ions.

associated with fermium ($Z = 100$) and since the x rays were in coincidence with α particles ($Z = 2$), these results provide an unequivocal identification of nobelium ($Z = 102$).

In addition to providing an identification of nobelium, our data provide detailed information concerning the level scheme for ^{251}Fm. In particular, approximately 43% of the observed Fm K-series x rays result from the de-exciting by K-shell internal conversion of a state at an excitation of 191 keV in ^{251}Fm. A lifetime of 15.2 ± 2.3 μsec was determined for this state by analyzing the time correlation between the Fm x rays and the ^{255}No α groups with energies of 8.121, 8.077,

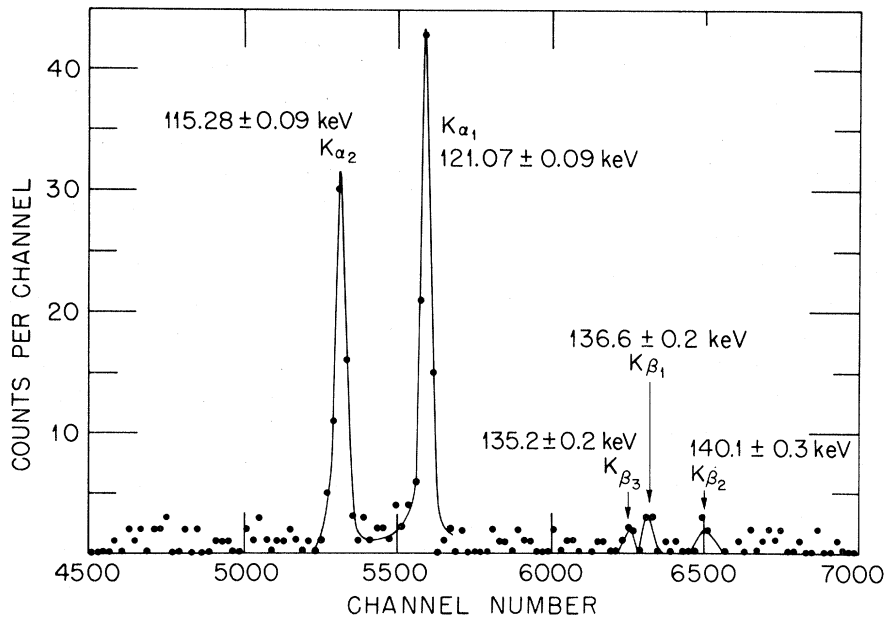

FIG. 2. A portion of the photon energy spectrum observed in coincidence with α particles in the decay of ^{255}No. The coincident Fm K-series x rays constitute a conclusive identification of nobelium.

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.