NAL PROPOSAL No. 119

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PRODUCTION OF HEAVY AND SUPER-HEAVY NUCLEI IN TARGETS IRRADIATED BY VERY HIGH ENERGY PROTONS

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Proposal to be submitted to the National Accelerator Laboratory, Batavia Illinois, USA

PRODUCTION OF HEAVY AND SUPER-HEAVY NUCLEI IN TARGETS IRRADIATED BY VERY HIGH ENERGY PROTONS

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The objective of the proposed experiment is to study the possible production of heavy and super-heavy nuclei by secondary reactions in targets irradiated by very high energy protons. The elastic or inelastic scattering of protons in the target could give high energy recoil nuclei. These nuclei could then react with other nuclei in the target to produce heavy or super-heavy nuclei. After irradiation chemical separations followed by the measurement of α-particle and fission fragment spectra would be made to try to isolate and identify the production from secondary reactions in the target.

The group has already carried out similar experiments using tungsten targets irradiated by about 10^{18} protons in the 24 GeV extracted proton beam from the CERN proton-synchrotron. These tungsten targets are those used for the production of secondary meson beams. Using sources prepared from these targets evidence has already been obtained for the possible existence of a super-heavy element with atomic number 112. (See attached preprint).

The group would like to obtain similar targets irradiated by the accelerator at NAL. The increased accelerator energy could give much higher energy recoil particles and hopefully a much larger number of nuclei produced by secondary reactions. It should be emphasised that we are not asking for a special irradiation for this experiment but any heavy element target which is used for the production of secondary beams from either the internal or external proton beam will be useful to us. (Alternatively a beam stopper which accepts a large fraction of the main beam intensity could be used).

Targets of any heavy element ($Z > 70$) of thickness greater than 50 gm/cm^2 and irradiated by more than 10^{16} protons would be suitable but if the group could have some limited choice of the target material to be used this could be very helpful. Almost any physical size of target could be used. It might also sometimes be desirable for the material to be supplied by this group so that specially purified material could be used or so that it could be subjected to prior chemical or spectroscopic analysis.

Rutherford Laboratory Preprint

RPP/NS/1

Evidence for the Possible Existence of a Superheavy Element with Atomic Number 112

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EVIDENCE FOR THE POSSIBLE EXISTENCE OF A SUPERHEAVY ELEMENT WITH ATOMIC NUMBER 112

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Abstract

Evidence is presented for the possible existence of a superheavy element produced by secondary reactions in a tungsten target bombarded by 24 GeV protons.

Introduction and Method

The possible existence of long lived superheavy nuclei due to the formation of a closed shell of protons around Z=114 and a closed shell of neutrons around N=184 has been the subject of many[1-6] theoretical papers. Predictions of the alpha, beta and spontaneous fission stability of the nuclei in this mass region have been made[7-11] and half-lives of up to 10^8 years, for some superheavy nuclei have been predicted.

It has been argued[12] that perhaps the best method to produce these new elements would be the bombardment of a heavy element target with very heavy

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ions, at energies above the Coulomb barrier which exists between the target nuclei and the projectile. Heavy incident ions are required to give the large neutron excess of the final nucleus. For some particular cases cross-sections of the order of about 10μb have been estimated[13] for the production of superheavy nuclei.

The present paper describes an attempt to produce superheavy elements by secondary reactions using presently available beams of very high energy protons. When such a proton is elastically scattering at a large angle from a heavy nucleus then a very high recoil energy may be achieved. For instance if a 24 GeV proton is elastically scattered from a W nucleus at angles between 45° and 180° to the incident beam, the recoil energy would be between 1.0 and 5.6 GeV respectively. A (p,n) or (p,p') reaction or alternatively reactions which involve a higher mass for the emitted particles such as (p,d) (p,α) etc, could also produce high energy recoil nuclei. These recoil energies can then exceed the Coulomb barrier of about 1.0 GeV which exists between two W nuclei. The superheavy elements could then perhaps be produced as a result of the interactions of the recoiling nucleus with another heavy nucleus in the target either due to an asymmetric fission reaction[13] or perhaps through an unusual type of transfer reaction.

Neither of the processes for the production of fast recoil nuclei mentioned above have been studied for the range of angles of interest in our experiment. Measurements[14,15] at 19.2 and 3 GeV for the elastic scattering of protons at small angles show a sharp decrease in the cross-sections with increasing angle and it is difficult to estimate from these measurements the cross-sections at much larger angles. On the other hand it may be argued that the cross-sections for the (p,n) and other inelastic reactions mentioned above will decrease much less rapidly with angle. If we assume a total

cross-section of about 1μb for the production of a "beam" of high energy heavy ions from 24 GeV protons (due to all the processes mentioned above and perhaps others) and we use the predicted cross-section of about 10μb for the induced fission reaction[13] we could expect a production yield of about 10^3 superheavy atoms in a typical target of 120 gm cm^{-2} of tungsten bombarded with about 10^{18} protons of 24 GeV energy. This may be a rather conservative estimate as the predictions of Karamyan and Oganesyan[13] probably underestimate the production cross-section.

In view of the fact that superheavy nuclei would decay to daughter nuclei of unknown properties, we cannot use the identification of the daughter nuclei to fix the Z and A of the parent decay. Also X-ray transitions cannot be used to identify the Z of the decaying nucleus because of the very small number of nuclei likely to be produced. It is therefore necessary to rely on the predictions[16] that elements, 110, 111, 112, 113 and 114 will be the chemical homologues of Pt, Au, Hg, Tl and Pb respectively, both for the identification of the atomic number of the element and for their separation from the 10^{23} atoms of other elements also present in the bombarded target.

Experiments have been carried out to separate actinides from the same targets as were used in the present work. The results so far indicate that significant alpha-activities have been observed from elements which closely follow the known chemistry of actinides. It has not so far been possible, however, to identify with absolute certainty, any specific alpha activity with known actinide isotopes. These results suggest that elements with $Z > 94$ may have been formed and it therefore seems possible that if superheavy elements ($Z > 110$) exist they could be formed in the same way.

In this paper we discuss measurements made on the Hg sources. Measurements on other sources are in progress. Both spontaneous fission and alpha activity have been observed in Hg sources prepared from the bombarded tungsten target. We believe that a possible explanation of these results is the presence of one or more isotopes of the element 112. Although the best evidence is that based on the observation of spontaneous fission we shall discuss the α-spectra first as some of these results are used in the interpretation of the fission data.

Chemical Separation

Two cylindrical tungsten targets, each 120 gm cm^{-2} thick and consisting of 33 gm of 99.95% tungsten were obtained after irradiation by the CERN proton-synchrotron. The first was bombarded by about 2×10^{18} protons of 24 GeV energy over a period of about a year and was 3-4 months old when it came into our possession. The second target was irradiated with about 7×10^{17} protons of the same energy over a period of about 4 months and we started the chemical separation a few days after the irradiation. We will refer to the first target as W1 and to the second as W2.

The targets were dissolved anodically at room temperature in an alkaline medium. Precautions were taken to avoid any loss of volatile products. The dissolution was carried out in the presence of 40 µg each of Os, Pt, Au, Hg, Tl and Pb as carriers. In addition, 1 mg quantities of Te, I, Ba, La, Zr, Ta and Sb were added as hold-back carriers for subsequent chemical operations.

The bulk of the radioactive products, including the elements Os to Pb were separated from the dissolved tungsten target by co-precipitation onto MnO_2. It was assumed that the eka-elements would follow their homologues

throughout these chemical separations.

The MnO_2 was dissolved in HCl/H_2O_2 (Au and Tl were extracted into ether at this point). The solution was adjusted to 1.5 M HCl and passed through a De-acidite FF anion exchange column. The elements Pt, Hg and Pb were retained on the column, and the actinides, among other elements, were not absorbed. The Hg was eluted with 0.01M HCl/thiourea. The complex was decomposed with HNO_3; the solution was adjusted to pH 2.5 with ammonia and the Hg extracted into CCl_4 as the dithizonate in the presence of EDTA as a masking agent. The Hg was back-extracted with dilute HCl and the dithizone extraction repeated as before. This separation procedure is specific for mercury.

Extrapolation of the periodic properties of the elements suggests that compounds of eka-mercury and the element itself may be more volatile than mercury. Sources were therefore prepared by vacuum evaporation of HNO_3 solutions at room temperature to minimise volatilisation of the eka-mercury. One of the main difficulties in preparing the sources was to reduce their weight in order to make them suitable for α-particle and fission fragment counting. This difficulty is severe with Hg as it is not possible to heat the sources in order to remove any organic material present. Both sources were about 2 mg cm^{-2} thick.

Alpha-Spectroscopy

Silicon surface-barrier detectors were used to detect the α-particles and extreme precautions were taken to avoid contamination of the detectors themselves. New detectors were used for the counting of all sources. Energy calibrations were made using an identical type of detector and it was shown that changing one detector for another of exactly similar type changed the

calibration by less than 0.2%. The calibration of the whole system of detector, amplifier etc, was observed to be stable to better than 0.5% over a period of several months. As confirmation that contamination of the apparatus was negligible no counts were observed over a period of 10 days with a blank platinum sheet in place of a source. The Hg sources were cooled to liquid nitrogen temperature to prevent the possible evaporation of the eka-mercury which was under vacuum for long periods.

Figure 1 shows α-particle spectra which were measured with the Hg sources. The source which was obtained from the second W target was counted twice, for 406 hours and for 236 hours and the results are shown in Figs 1a and 1b respectively. The average time difference between these two measurements was about 24 days. Fig 1c shows an α-particle spectrum obtained with the Hg source from the first W target. This spectrum was collected over a period of 280 hours. In all these spectra one can see a peak at $6.73 \begin{smallmatrix} + 0.150 \\ - 0.050 \end{smallmatrix}$ MeV. (The larger uncertainty towards the high energy is due to thickness of the sources). The total number of counts under the 6.73 MeV peak are 12, 7 and 12 in Figs 1a, 1b and 1c respectively. A comparison of Fig 1a and 1b shows that within the statistical accuracy this peak did not decay over a period of 24 days.

In the range of energies from 6.67 to 7.00 MeV there are more than 40 known α-groups which could in principle be candidates for producing the observed peak at 6.73 MeV which did not decay over a period of 24 days. However, most of the alpha groups are due to the decay of short lived isotopes which, since they do not have a parent of half-life greater than 10 days, could not be responsible for the observed peak. In the few other cases they can be excluded as candidates for producing this peak since if they were present, other pronounced alpha groups, which we did not see, should also be

observed. Perhaps the most problematic case is the 6.78 MeV group of ^{216}Po which exists in the decay chain of ^{228}Th (half-life of 1.91 years). However, in the decay of ^{228}Th there are other groups of α-particles at 5.68 and 6.29 MeV of equal intensity and at 5.43 and 8.78 MeV of comparable (70%) intensity to the 6.78 MeV group. In all the spectra shown in Fig 1, the peak at 6.73 MeV is very pronounced, it is the largest high energy group in Fig 1c and in Figs 1a and 1b only the groups at 5.14 MeV and 5.73 MeV which we believe are due to ^{210}Po and ^{236}Pu are larger than the 6.73 MeV peak. In Figs 1b and 1c we do not see a pronounced peak at 5.43 MeV and in Figs 1a and 1c we do not see a strong group at 5.68 MeV. (The few counts that exist in this region could also be due to the tail of the ^{236}Pu group). On statistical arguments, it therefore seems very unlikely that the observed 6.73 MeV peak is principally due to thorium contamination.

The peak at 5.14 MeV seen in Figs 1a and 1b with an upper edge at 5.30 MeV is probably due to the 5.31 MeV α-particle from the decay of ^{210}Po. The difference in the energy of the peak and the possible true alpha energy is a reflection of the thickness of the source. Possibly due to slight differences in the chemical separation techniques, this peak is not observed in the spectrum, Fig 1c from the W1 source. The peak at 5.73 MeV in Figs 1a and 1b may be due to the 5.75 MeV decay of ^{236}Pu. We do not at present have any clear explanation for the peak at 8.78 MeV; as we have shown above it is unlikely to be due to the decay chain of ^{228}Th. The peak at 3.1 MeV in Fig 1c is probably due to the decay of ^{148}Gd.

It might be argued that the peak at 6.73 MeV could be due to contamination by some unknown actinide which is also produced in the original target. The results of measurements on the actinides will be given later and here we

summarise only the important features which are relevant to the present discussion.

a Actinides appear to be produced in the tungstent target.

b The most pronounced group of α-particles observed in the spectra is at 3.18 MeV due to the decay of ^{148}Gd which is expected to be chemically separated with any actinides.

c A group at 6.75 MeV which appears in the spectra of most of the actinide sources has a total intensity of about 1/250 of the intensity of the 3.18 MeV group.

It is reasonable to assume that the decontamination factor for gadolinium would be similar to that for the transamericium actinides in the Hg chemical separation procedure. On this basis, and from the results of the actinide measurements mentioned above, one concludes that only 2 counts in the 6.73 MeV group observed in all the spectra from the Hg source may be due to the 6.75 MeV group of the actinides.

If, however, the 6.75 MeV group that appeared in the spectra from the actinide sources is due to decay of an unknown daughter of an actinide, the comparison with the intensity of the 3.18 MeV peak is no longer valid. The contamination in this case may be larger.

Finally we should comment that it seems unlikely, on the basis of half-life, that the α-group is due to any unknown decay of mercury even from an isomer state.

Observation of Spontaneous Fission

Since evidence for a superheavy element based solely on α-decay might not be considered convincing, a search was also made for spontaneous fission

events occurring in the Hg source using Polycarbonate films*, 1.2 mg cm^{-2} thick to detect the fission fragments[17]. Holes due to fission fragments were observed in several cases. It was confirmed that the spatial position of the holes corresponded with the area covered by the source. Measurements were started about 2 months after the separation of the mercury source from the W2 tungsten target. The results can be summarised as follows.

1 A foil which was kept for 7 days above the Hg source produced from the second W target (W2) had 8 holes in it.

2 Another foil held 4 days later for a period of 14 days above the same source had 34 holes in it.

3 Following attempts to reduce the thickness of the W2 source a third foil held above it for a period of 8 days had 28 holes.

4 A fourth foil held above the same source 13 days later for 8 days had 23 holes.

5 Four similar, but unirradiated foils processed in an identical way had no holes.

6 Eight foils kept for 12 days above Pt sheets identical to those used as backings for the Hg sources had on average less than one hole in each foil.

On the basis of these results$^{\phi}$ we would conclude that approximately 93 fission fragments from spontaneous fission in the Hg source from target W2 have been observed over a period of 37 days.

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* "Makrofol K G", supplied by Bayer Chemicals Ltd

$^{\phi}$ Preliminary measurements on a mercury source from a third tungsten target very recently received also give evidence for the observation of spontaneous fission.

Spontaneous fission is a rare mode of decay and is confined almost entirely to the elements above Uranium. However, we do not believe that the spontaneous fission events observed in the mercury source can be ascribed to contamination. Certainly they are not due to the well known spontaneous fission of ^{252}Cf (2.5 years) since we do not see the associated alpha group at 6.1 MeV. Estimates of the decontamination factors from various elements for the Hg sources made by comparing α-groups indicate that the fission events observed with the Hg source are unlikely to be due to any contamination. It is also unlikely to be due to the fission of a mercury isotope or of a metastable state since events of this type have not previously been reported in this mass region.

It is of some interest that so far we have no evidence for spontaneous fission occurring in the Pt, Au and Tl sources prepared at the same time as the Hg source. This indicates that the fission is specific to the Hg source and may be due to the decay of element 112 or one of its daughter products. For a crude estimate of the lifetime we assume that a maximum of 10^6 atoms of the isotope responsible were formed in the target. The rate of decay then implies an upper limit to the half-life of approximately 500 years. An estimated lower limit for the longest lived member in the decay chain producing the spontaneous fission is 1 month. No fission events were observed with the W1 source. This may be due either to the effects of source thickness or because it was about a year older than the W2 source when these measurements were made.

Clear evidence that the fission is due to a superheavy element would be given by a measurement of the fission energy spectrum. Unfortunately this is not at present possible owing to the thickness of the source.

Comparison with Predictions for Element 112

It seems reasonable to conclude that the α-group at $6.73 \begin{smallmatrix} + 0.15 \\ - 0.05 \end{smallmatrix}$ MeV and the fission fragments which we have observed may be due to decay of one or more isotopes of element 112. We already have shown that they seem unlikely to be due to the actinides or any other known activity. On the other hand the chemical properties of element 112 are predicted[16] to be similar to those of Hg and to be different from those of the elements from 104 to 111.

Our results give a disintegration energy of about 6.82 MeV assuming 290 as the mass of the recoil nucleus. This fits well with the predictions for β-stable isotopes of element 112 by Nilsson et al[8,10] and Muzychka[11] which lie in the range 6.5 to 7.0 MeV, and not so well with that by Lightman and Gerrace[9] of 7.5 MeV. The predicted half lives[8,10] lie in the range 10^3 to 10^4 years. Then if we assume a partial half-life of about 10^3 years for the isotope of element 112 which decays by α-particles of 6.73 MeV energy, the number of atoms of this isotope existing in our sources is estimated to be $10^5 - 10^6$.

Spontaneous fission half-lives in the range 10^6 to 10^{13} years were predicted by Nilsson et al[8,10] for β-stable isotopes of element 112. Our results would indicate a much shorter lifetime, however, they are still within the uncertainty of the predictions. It may also be that the observed fission events are due to a lighter isotope of element 112 where the spontaneous fission half-life is predicted to be shorter than the α-half-life.

To summarise we believe we may have observed the production of element 112 by secondary reactions in tungsten targets irradiated by 24 GeV protons. We have observed spontaneous fission (which is specific to the mercury source) and which is unlikely to be due to any contamination by actinides

or due to fission from some unknown isomeric state of mercury. We have also observed an α-decay at 6.73 MeV energy which, although with less certainty, is unlikely to be due to any contaminant and whose energy agrees with several sets of predictions for element 112.

Many people in different laboratories have helped us in various stages of the experiments and we are very grateful to them. In particular we would like to express our thanks to P H Standley and Ch Steinbach from CERN for providing us with the irradiated tungsten targets, to Mrs K M Glover and F J G Rogers from AERE, for continuing help at many stages of the experiment, to G B Stapleton for making the facilities of the radio chemistry laboratory available to us, and to J G Cuninghame, Drs A M Friedman, G Manning, G H Stafford and Professors K Bagnall, W D Allen and S G Cohen for very valuable discussions.

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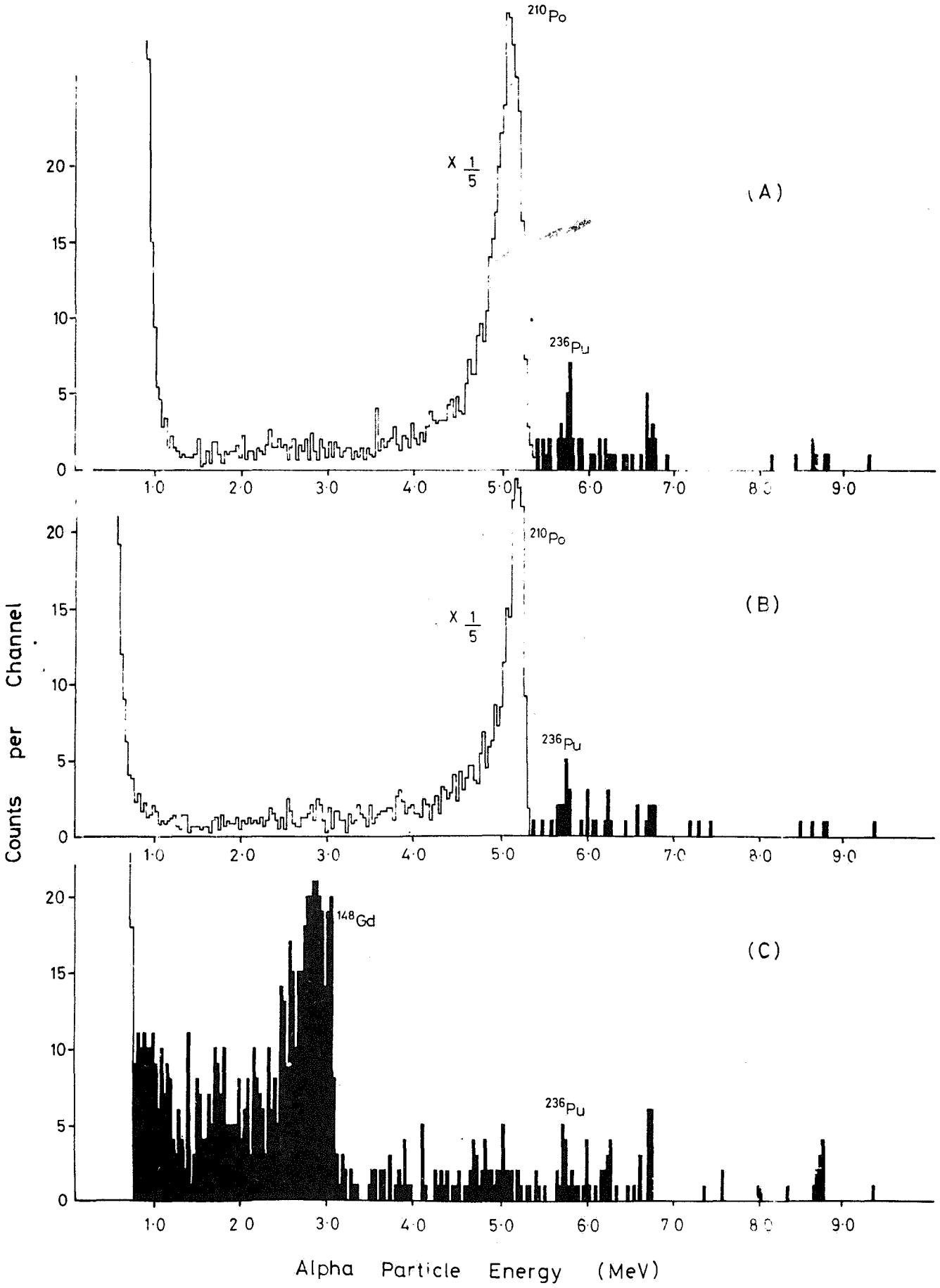
Caption to Figure

Alpha-particle spectra measured with the Hg sources. Figures (a) and (b) were obtained over periods of 406 and 236 hours respectively with the source obtained from the second tungsten target. Figure (c) was obtained over a period of 280 hours with the source from the first tungsten target.


210Po
X 1/5
(A)
236Pu
210Po
X 1/5
(B)
236Pu
148Gd
(C)
236Pu
Counts per Channel
Alpha Particle Energy (MeV)