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

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

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 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 that perhaps the best method to produce these new elements would be the bombardment of a heavy element target with very heavy ions, at energies above the Coulomb barrier which exists between the target nuclei and the projectile (see ref. 12, for instance). 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 10 μbarn have been estimated for the production of superheavy nuclei (S. A. Karamyan and Yu. Ts. Oganesyan, unpublished).

This article describes an attempt to produce superheavy elements by secondary reactions using beams of very high energy protons. A very high recoil energy may be achieved when such a proton is elastically scattering at a large angle from a heavy nucleus. 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) and so on, 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nucleus with another heavy nucleus in the target either due to an asymmetric fission reaction (Karamyan and Oganesyan, unpublished) or perhaps through an unusual type of transfer reaction.

Neither of the processes for the production of fast recoil nuclei mentioned here has been studied for the range of angles of interest in our experiment. Measurements[13,14] 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 will decrease much less rapidly with angle. If we assume a total cross-section of about 1 μbarn for the production of a "beam" of high energy heavy ions from 24 GeV protons (due to all the processes mentioned here and perhaps others) and we use the predicted cross-section of about 10 μbarn for the induced fission reaction (Karamyan and Oganesyan, unpublished) we could expect a production yield of about 10^3 superheavy atoms in a typical target of 120 g 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 probably underestimate the production cross-section.

Because superheavy nuclei will 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[15] 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 article we discuss measurements made on the Hg sources. Measurement 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 alpha spectra first as some of these results are used in the interpretation of the fission data.

Chemical Separation

Two cylindrical tungsten targets, each 120 g cm^{-2} thick and consisting of 33 g 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

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period of about a year and was three to four 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 four 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 coprecipitation 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 0.01 M HCl/thiourea. The complex was decomposed with HNO_3; the solution was adjusted to *p*H 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 minimize volatilization of the eka-mercury. One of the chief difficulties in preparing the sources was to reduce their weight in order to make them suitable for alpha particle and fission fragment counting. This difficulty is severe with Hg because 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 and so on 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 ten 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.

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

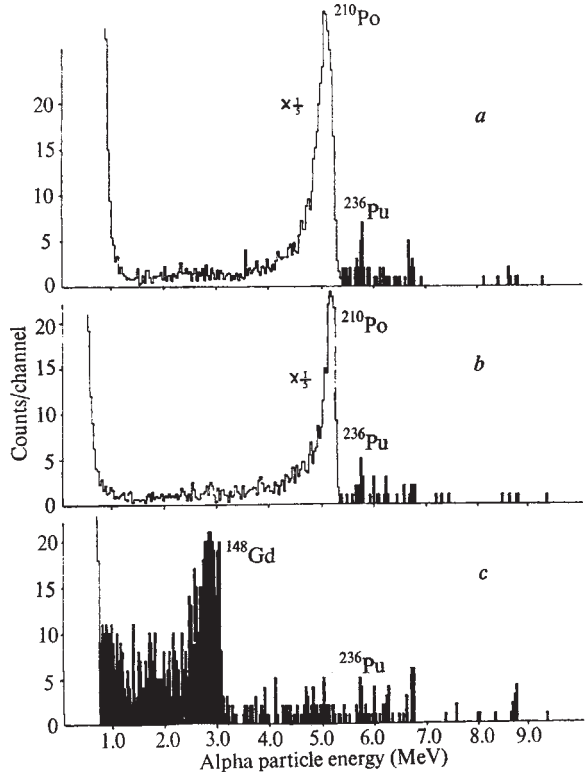

Fig. 1 Alpha particle spectra measured with the Hg sources. *a* and *b* were taken over periods of 406 and 236 h respectively with the source obtained from the second tungsten target. *c* was obtained over a period of 280 h with the source from the first tungsten target.

In the range of energies from 6.67 to 7.00 MeV there are more than forty known alpha 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. Most of the alpha groups, however, are due to the decay of short lived isotopes which, because 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 because, 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). In the decay of ^{228}Th, however, there are other groups of alpha 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. 1*c* and in Figs. 1*a* and *b* 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. 1*b* and *c* we do not see a pronounced peak at 5.43 MeV and in Figs. 1*a* and *c* 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. 1*a* and *b* with an upper edge at 5.30 MeV is probably due to the 5.31 MeV alpha particle from the decay of ^{210}Po. The difference in the energy of the peak and the possible true alpha energy is a reflexion 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. 1*c*, from the W1 source. The peak at 5.73 MeV in Figs. 1*a* and *b* 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, it is unlikely to be due

to the decay chain of ^{228}Th. The peak at 3.1 MeV in Fig. 1*c* 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 summarize only the important features which are relevant to the present discussion.

(*a*) Actinides seem to be produced in the tungsten target.

(*b*) The most pronounced group of alpha 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 of 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 here, one concludes that only two 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 alpha group is due to any unknown decay of mercury even from an isomer state.

Observation of Spontaneous Fission

As evidence for a superheavy element based solely on alpha decay might not be considered convincing, we also searched for spontaneous fission events occurring in the Hg source using polycarbonate films (*Makrofol KG*, supplied by Bayer Chemicals Ltd) 1.2 mg cm^{-2} thick to detect the fission fragments[16]. 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 two months after the separation of the mercury source from the W2 tungsten target. The results can be summarized as follows.

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

(2) Another foil held four days later for a period of fourteen days above the same source had thirty-four holes in it.

(3) Following attempts to reduce the thickness of the W2 source a third foil held above it for a period of eight days had twenty-eight holes.

(4) A fourth foil held above the same source thirteen days later for eight days had twenty-three holes.

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

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

On the basis of these results 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. (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. But 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) as 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 alpha groups indicate that the fission events observed with the Hg source are unlikely to be due to any contamination. They are also unlikely to be due to the fission of a mercury isotope or of a metastable state as 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 one 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 because of the thickness of the source.

Comparison with Predictions for Element 112

It seems reasonable to conclude that the alpha group at $6.73^{+0.15}_{-0.01}$ MeV and the fission fragment which we have observed may be due to decay of one or more isotopes of element 112. We have shown already 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[15] 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 for the mass of the recoil nucleus. This fits well with the predictions for beta-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 alpha 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 beta-stable isotopes of element 112. Our results would indicate a much shorter lifetime, but 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 alpha half-life.

To summarize, we believe that 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 alpha-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.

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Protein Polymorphism as a Phase of Molecular Evolution

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It is proposed that random genetic drift of neutral mutations in finite populations can account for observed protein polymorphisms.

SINCE one of us[1] put forward the theory that the chief cause of molecular evolution is the random fixation of selectively neutral mutants, some have supported the theory[2–5] and others have criticized it[6–8].

Rate of Evolution

Probably the strongest evidence for the theory is the remarkable uniformity for each protein molecule in the rate of mutant substitutions in the course of evolution. This is particularly evident in the evolutionary changes of haemoglobins[5], where, for example, the number of amino-acid substitutions is about the same in the line leading to man as in that leading to the carp from their common ancestor. Similar constancy is found on the whole for cytochrome C, although the rate is different from that of the haemoglobins. The observed rate of amino-acid substitution for the haemoglobins is very near to one Pauling (10^{-9}/amino-acid site/yr) over all vertebrate lines[5]. The rate for cytochrome C is roughly 0.3, while the average rate for several proteins is about 1.6 times this figure[2].

If we define the rate, k, of mutant substitution in evolution as the long term average of the number of mutants that are substituted in the population at a cistron per unit time (year, generation and so on), then under the neutral mutation–random drift theory, we have a simple formula

$$k = u \qquad (1)$$

where u is the mutation rate per gamete for neutral mutants per unit time at this locus. Note that this rate k is different from the rate at which an individual mutant increases its frequency within a population. The latter depends on effective population size.

The uniformity of the rate of mutant substitution per year for a given protein may be explained by assuming constancy of neutral mutation rate per year over diverse lines. Moreover, the difference of the evolutionary rates among different molecules can be explained by assuming that the different fraction of mutants is neutral depending on the functional requirement of the molecules.

On the other hand, it can be shown that if the mutant substitution is carried out principally by natural selection

$$k = 4N_e s_1 u \qquad (2)$$

where N_e is the effective population number of the species, s_1 is the selective advantage of the mutant and u is the rate at which the advantageous mutants are produced per gamete per unit time[9]. In this case we must assume that in the course of evolution three parameters N_e, s_1 and u are adjusted in such a way that their product remains constant per year over diverse lines. The mere assumption of constancy in the "internal environment" is, however, far from being satisfactory to explain such uniformity of evolutionary rate. In our example of carp–human divergence, we must assume that $N_e s_1 u$ is kept constant in two lines which have been separate for some 400 million years in spite of the fact that the evolutionary rates at the phenotypic level (likely to be governed by natural selection) are so different.

Polymorphism in Sub-populations

Kimura[1] also suggested that the widespread enzyme polymorphisms in *Drosophila*[10] and man[11] as detected by electrophoresis are selectively neutral and that the high level of heterozygosity at such loci can be explained by assuming that most mutations at these cistrons are neutral. This suggestion, however, has been much criticized[12–14]. One of the chief objections is that the same alleles are found in similar frequencies among different sub-populations of a species and that some kind of balancing selection must therefore be involved.

Robertson[15] suggested that if a large fraction of mutations at a locus is selectively neutral, we find either very many alleles segregating in large populations, or a small number of different set of alleles in different isolated small populations. He considered that because neither of these alternatives is found, most polymorphisms have at some time been actively maintained by selection.