Search for naturally occurring superheavy element $Z = 110$, $A = 294$

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A search for the superheavy element $Z = 110$ and $A = 294$ has been made using the Tandem accelerator as a highly sensitive mass spectrometer. The observed background leads to an upper limit on the presence of $^{294}110$ in a sample of placer platinum of 1 in 10^{11}. This result implies that $^{294}110$ either has a lifetime less than about 2×10^8 yr, or was not produced in the amounts predicted by Schramm and Fowler, or did not follow its homolog platinum in the geophysical and chemical processing of the material.

[NUCLEAR STRUCTURE Search for superheavy element (110,294), upper limit found, use of Tandem accelerator as sensitive mass spectrometer.]

The superheavy element predicted to have been synthesized in nature with a half-life greater than 10^8 yr and hence still detectable is the element $Z = 110$ and $A = 294$ ($^{294}110$). Nilsson, Thompson, and Tsang[1] have calculated that this isotope may have a half-life for alpha emission of greater than 10^8 yr, a half-life for spontaneous fission of 10^{12} yr, and beta stability. Although Randrup *et al.*[2] agree that $^{294}110$ is the longest lived superheavy isotope, they predict its total decay half-life to be 10^5 yr, again with considerable uncertainty in absolute value. Larsson *et al.*[3] suggest that inclusion of quadrupole pairing and Coulomb shell corrections will raise the barriers and increase the life times. Schramm and Fowler[4] have estimated the yield of this isotope produced by multiple neutron capture in a supernova explosion (r process) to be 0.02 to 0.06 of the abundance of ^{195}Pt similarly produced. But Howard and Nix[5] find a cutoff of the r process at $Z = 96$, $N = 186$, similar to Bengtsson *et al.*[6] and Boleu.[7] Only Meldner, Nuckolls, and Wood[8] speculate on possible neutron capture paths to the superheavy island. So, if it is postulated that a nearby supernova (SN) triggered the formation of the solar system, then $^{294}110$ might have been synthesized to the extent of about 0.01 of platinum. If $^{294}110$ follows its homolog through the condensation of the earth and geologic history, then its abundance now would be reduced only by its decay. With these assumptions we have plotted in Fig. 1 the fractional number abundance $F = n(110)/n(\mathrm{Pt})$ to be expected now after 4.7×10^9 yr for various possible values of the decay half-life of $^{294}110$. Figure 1 also shows the presently expected alpha decay rate of $^{294}110$ per gram of platinum per second for various values of the half-life and the spontaneous fission rate for several possible fission half-lives of $^{294}110$. $^{290}110$, on the other hand, formed from the alpha decay of $^{294}110$ and two subsequent beta decays, would presumably promptly undergo fission after formation. Following Nilsson *et al.* (Ref. 1), $^{290}108$ would have a fission lifetime of 100 years and a beta decay lifetime of 10 years. The resulting $^{290}109$ would decay promptly to $^{290}110$ or would itself quickly undergo fission. Consquently, following chemical segregation of element 108 from 110 (such as during purification of an ore), the spontaneous fission rate for 110 would rise, over a ten-year period, from the fission curves for $^{294}110$ to the d_α curve in Fig. 1.

A number of searches for superheavy elements in platinum have already been carried out with negative results.[9-11] As Seaborg, Loveland, and Morrissey[12] point out, the recent theoretical estimates of low barriers, hence shorter lifetimes and lower r-process cutoff, are supported by the failure to detect superheavies in the ^{48}Ca+^{248}Cm reaction. Nevertheless, the feasibility of the use of a heavy ion accelerator as a highly sensitive mass spectrometer[13] prompted a further search for the most likely superheavy isotope on the possibility that its lifetime was greater than 2×10^8 yr but had escaped previous searches.

Our search was made possible by the ease of producing appreciably large platinum negative ion beams from our sputter source.[14] Using a nugget of placer platinum from the Goodnews Bay in Alaska, we have been able to produce steady beams of 2.6 μA of platinum negative ions. [In the geophysical condensation and subsequent geochemical concentration of the heavy elements in ultramafic rocks the association of possible eka platinum element 110 (homologous to platinum[15]) and platinum might be expected, as evidenced by the presence of group VIII metals rhodium and nickel.[16] Placer platinum nuggets have had no subsequent purification which might segregate possible element 110 from platinum.] The trans-

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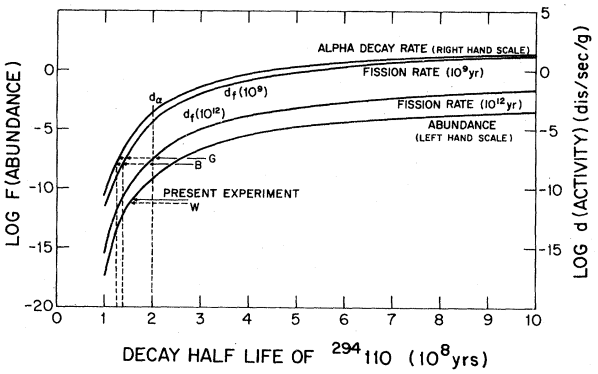

FIG. 1. The left hand vertical $\log_{10}$ scale gives the present fractional abundance compared to platinum of the isotope $Z=110$, $A=294$ expected from SN production initiating solar system formation. The lower curve shows this abundance as a function of assumed lifetime for ^{294}110. The upper curves show the activity in disintegrations per second per gram of platinum on the right hand vertical $\log_{10}$ scale again as a function of a assumed half-life of ^{294}110. The top curve indicates the alpha activity of ^{294}110. The middle curves show the fission activity of ^{294}110 for assumed half-lives for spontaneous fission of ^{294}110 of 10^9 or 10^{12} yr, respectively. See text for further details.

mission of our FN Tandem Accelerator for heavy ions as measured with gold and platinum ions is about $(\frac{1}{4})\times10^{-4}$ using gas stripping at 3.15 MV on the stripper canal and a good resolution injector magnet. After acceleration the positive ion beam was filtered through a crossed field velocity selector, with a resolution of $M/\Delta M=75$, an analyzing magnet ($M/\Delta M=375$), a time-of-flight detector ($M/\Delta M=62$), an argon ionization chamber, and a silicon surface barrier detector as indicated in Fig. 2.

The time-of-flight detector involved a pulse pickup unit consisting of a few μg/cm^2 carbon foil in the beam to produce secondary electrons which were accelerated by a 500 V grid onto a microchannel plate amplifier (Galileo Electro-Optics Corporation) working at 900 V with the output accelerated by 2 kV onto a silicon surface barrier detector as described by Andrews.[17] This pulse was delayed 540 nsec and used to stop the time-to-amplitude converter. The start pulse to the TAC was taken from another silicon surface barrier detector located in the beam path behind the argon ionization chamber. A 2.5μm Mylar window formed the front of this chamber which contained 45 Torr of argon over a path length of 6.4 cm. The flight time was measured over a total length of 144 cm, traversed by the platinum 6$^+$ ion in 337 nsec (calculated time of flight after correcting for the slowing down in argon). The signal from the argon ionization chamber, rather than the linear signal from the "stop" surface barrier detector, was used to measure the energy of the detected particle in order to avoid the serious problems with incomplete charge collection caused by recombination of charge carriers. The argon pressure was adjusted so that ions of large Z lost most of their kinetic energy in the gas. The time of flight for each ion reaching the solid state detector was computer plotted versus the argon pulse height as in Fig. 3, allowing discrimination for mass and energy. The setup was calibrated primarily by using the ^{196}Pt 6$^+$ ion from the breakup at the stripper of a ^{196}Pt ^{103}Rh negative ion which was injected simultaneously with mass 294 by the negative ion injector magnet ($M/\Delta M=60$). The peak in Fig. 3 is ascribed to this weak ion beam and the expected position of a ^{294}110 9$^+$ ion is shown as the neighboring circle. The observed

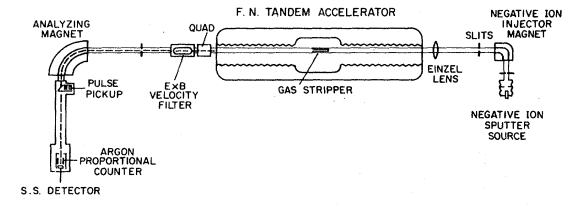

FIG. 2. General plan of FN Tandem accelerator as used for super sensitive mass spectroscopy with time-of-flight and argon chamber detector. (Sketch not to scale.)

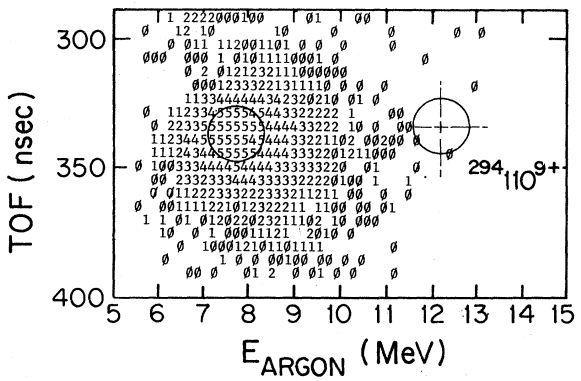

FIG. 3. Computer display showing plot of number of observed ions (binary logarithmic scale) as a function of (vertical scale) a fixed delay of 540 nsec minus the time-of-flight time in nsec over 144 cm flight path length and (horizontal scale) energy loss in 6.4 cm of 45 Torr argon (0.675 mg/cm^2). The main peak is ascribed to the ion ^{196}Pt 6$^+$ produced in the sputter source as ^{196}Pt ^{103}Rh$^-$ and injected as mass 294 by the negative ion magnet. The ^{196}Pt breaks off from the molecule during gas stripping at 3.15 MV, is accelerated as a 6$^+$ ion to 21.0 MeV and consequently has nearly the same magnetic rigidity as a particle of 31.5 MeV at mass 294 and charge 9$^+$. The circle shows the expected location of an isotope $Z=110$, $A=294$ injected as a negative ion, subsequently stripped and accelerated as a 9$^+$ ion.

background of 2 counts in 1260 sec. leads to an upper limit on $^{294}110$ in this sample of platinum of less than 1 in 10^{11} or a fractional number abundance of less than 1×10^{-11} (at the 95% confidence level). This result is shown in Fig. 1 as an arrow pointing to the abundance curve. That is, if $^{294}110$ had been produced in the trigger SN explosion to the extent suggested by Schramm and Fowler,[4] and had followed platinum in the geophysical and chemical processing of terrestrial material, then it must have a half-life for decay of less than 1.7×10^8 yr to be undetected at this level.

These results are consistent with and confirm the negative results of the other searches that have been made. Wesolowski *et al.*[9] have examined placer platinum by searching for neutron-induced fission of energy greater than 215 MeV and assuming the (n,f) cross section to be the same as for ^{235}U. As shown in Fig. 1 this corresponds to a decay half-life for $^{294}110$ of less than 1.5×10^8 yr with an uncertainty due to the assumed (n,f) cross section. Behringer *et al.*[10] used their "spinner" detector for spontaneous fission and found a rate of fissioning of about one in 2.5×10^4 hours per gram of platinum. This rate of 1.1×10^{-8} fissions per second per gram is shown by the arrow *B* in Fig. 1 pointing at the fission rate curves. It corresponds to a limiting decay lifetime of $^{294}110$ of less than 1.8 to 1.25×10^8. Geisler, Phillips, and Walker[11] have looked for spontaneous fission fragment tracks from a platinum sample and set a limit corresponding to 2 to 5×10^{-8} fissions per gram per second as shown by the arrow *G* in Fig. 1 and limiting the decay half-life of $^{294}110$ to less than about 1.4×10^8 yr depending on the fission fragment range.

All of these results agree in indicating either that $^{294}110$ was not produced to the extent predicted by Schramm and Fowler (as suggested by Wene and Johansson,[18] Viola,[19] and Howard and Nix[5], or that the half-life for decay is less than 2×10^8 yr or eka platinum has segregated differently than platinum. Our limits on the presence of $^{294}110$ could probably be lowered by using a platinum sample devoid of rhodium since PtRh supplies much of our background. However, as Fig. 1 shows, it will be difficult to lower the half-life limit very much. Even if more recent material were available (such as accumulated cosmic rays), any $^{294}110$ would probably lack the concentration available from platinum ore and be too dilute to allow detection at these levels of sensitivity.

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