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Final Report
for
Grant NGR-05-009-170
National Aeronautics and Space Administration
Washington, D. C. 20546

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Title: Fission Products of Superheavy Elements. An investigation of the naturally occurring fission products of elements heavier than Uranium.

Duration: One Year (April 1, 1971-72)

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(NASA-CR-138798) FISSION PRODUCTS OF SUPERHEAVY ELEMENTS. AN INVESTIGATION OF THE NATURALLY OCCURRING FISSION PRODUCTS OF ELEMENTS HEAVIER THAN (California Univ.) 7 p HC $4.00 CSCL 18G G3/24
N74-28151
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1. Introduction.

Much theoretical work has been carried out on the stability and decay properties of nuclei in the region of the doubly closed shell nucleus $^{298}114$. It was suggested that the nuclide $^{294}110$ might be the most likely candidate for a long-lived extinct superheavy nucleus. There are certain indications in favor of extinct superheavy elements in some meteorites, and this evidence comes from isotopic anomalies of meteoritic Xe and possibly Kr.

The fact that there is evidence for fission products in nature which cannot be attributed to presently naturally occurring elements clearly suggests that extinct elements may be their parents. The fission products from element X which are present in many of the very old chondrites indicate a quite different mass yield distribution not only in the mass region 130 to 140, but also around mass 86. If fission excesses on the Kr isotopes can be substantiated, then an asymmetric fission mode is required with relatively high yields at mass 86. These requirements, together with the absence of a correlation with U, seem to rule against Actinide elements as candidates for element X. It appears possible, therefore, that element X has to be found among the superheavy elements with $Z \geq 104$. It seems that a half-life of about 10^7 years would be required, although at present it is not possible to give an exact value. Such a half-life does appear to be compatible with theoretically predicted α-half-lives of about 10^8y for elements with $Z = 110$ to 114.

2. Progress Report.

We have proposed to study the fission mass yields in different structural elements and mineral separates and then to carry out a systematic search for element X. We have separated several structural elements (matrix, different chondrules, a dark inclusion, some Ca-rich inclusions) and also separated a heavy fraction (density >4.05) which then was magnetically separated into a magnetic and non-magnetic fraction. Each fraction was microscopically inspected, and the percent enrichment of metal and sulfides was estimated. These two fractions were considered essential, since some of the superheavy elements are expected to belong to either the siderophile or calcophile element groups. We have analysed many of these separates in an attempt to locate the fission components. We have not yet succeeded in locating the element X fission component exactly, but on the basis of our results, we can already exclude a number of possibilities. We have, however, located the ^{244}Pu fission component, and also we have secured some very important information on the genesis and chronology of this meteorite. We will briefly summarize the evidence. More detailed discussions will be given in a paper at the AGU spring meeting and in a paper which is in preparation.

2.1 The ^{244}Pu Fission Component.

A distinct fission excess with a large relative yield on ^{132}Xe was found in the Ca-rich inclusions. The decomposition of the isotopic yields for the heavy Xe isotopes into a trapped, a spallation and a fission component gives fission mass yields which are in very good agreement with the ^{244}Pu mass yields obtained by Alexander et al. (1971). The ^{136}Xe fission concentration is 1.1×10^{-11} cc STP/g which is more than one order of magnitude larger than the $^{238}U_{s.f.}$ fission component. The latter component, therefore, will only slightly affect the relative mass yields.

2.2 Element X Fission Component.

This fission component is found in the matrix and in the chondrule fractions. Surprisingly, it is not found in either one of the heavy mineral separates. Our current best values for the relative mass yields are derived from the data obtained in the heavy mineral separates as well as the 1000° and 1800°C temperature fractions of a bulk sample and a chondrule sample. The yields (^{130}Xe ≡ 0, ^{132}Xe: ^{134}Xe: ^{136}Xe = 0.20: 0.66: 1.00 indicate even smaller yields on ^{132}Xe and ^{134}Xe than previously estimated. Work on the matrix is in progress. The nature and origin of the element X fission component has to be further investigated. It is still not clear whether element X has been incorporated together with the other extinct radioisotopes and whether it can, in fact, be used for age dating. The present evidence can already be used to eliminate a number of possible precursors.

Effects from neutron irradiation in space will be used to study in detail any differences in the time of formation of individual separates, including chondrules and inclusions. Criteria of geochemical affinity of element X and of the heavy elements in the periodic table can, of course, only be applied if element X decayed in situ. It should also be pointed out here that this information will be of much help in interpreting lunar data: Lunar Xe appears to include fission components, and, on the other hand, meteorites appear to include solar-type Xe. In most of these investigations, Kr data will also be obtained. The fission yields in the Kr mass region can be expected to be much smaller. Nevertheless, they will be very important for an assessment of the fission yields and of the asymmetric or symmetric fission modes.

2.3 Radiogenic ^{129}Xe and Neutron Activation in Space.

The isotopic composition of Xe in one Ca-rich inclusion is unique: 99% of the total Xe is radiogenic ^{129}Xe, 0.5% is ^{244}Pu fission Xe and another 0.1% is neutron produced ^{128}Xe from neutron capture in I. The ^{129}Xe/^{132}Xe ratio in this inclusion is 350, the largest ratio ever measured. The question of the ^{129}I - ^{129}Xe age of this inclusion has to be answered. The $^{129}Xe_r$ concentration is ≈ 2 x 10^{-8}cc STP/g! Fortunately, we have obtained the answer from a neutron activation in space. The ^{128}Xe and ^{80}Kr anomalies from neutron capture in I and Br respectively show that the halogens in the Ca-rich inclusion are enriched by a factor of 12 relative to the bulk sample. This factor accounts for all the $^{129}Xe_r$ and the ^{129}I - ^{129}Xe age of the inclusion does not appear to be much different from the bulk rock age.

4. Facilities and Instruments.

All the work was performed at UCSD, and the equipment for this study was available in our laboratories. Major items include:

(a) A six inch, single focussing noble gas spectrometer with electron bombardment source, electromagnetic source and deflection magnets and with two detection systems. The instrument is operated at ultrahigh vacuum (~10^{-10} torr) and achieves a detection limit of 2 x 10^{-15}cc STP per isotope at 2mA emission current and a maximum resolution of 750.

(b) A 12 inch solid source spectrometer, thermal ionization, single focussing with both straight and multiplier detection systems. The latter is being used in either the integrating or the pulse-counting modes.

(c) A techtron Atomic Absorption Instrument with analog output and hollow cathode lamps of a large number of chemical elements. A precision of 2% was achieved on this instrument.

These instruments are complete systems with integrated electronic and vacuum supplies.

5. Personnel.

In addition to the Principal Investigator, one co-investigator (Dr. T. W. Osborn) and one graduate student have participated in this research.

We have taken advantage of a technical staff (part time) for maintenance, service and updating of all the instruments and electronic equipment.

6. Concluding Remarks.

We have located the ^{244}Pu fission component and succeeded in enriching the "element X" fission component. Any information on superheavy elements is of importance to the nuclear chemist and physicist. A possible identification of a naturally occurring superheavy element, therefore, would be an extremely welcome contribution to this knowledge. The occurrence of such an element in nature, together with an estimate of its half-life, would be of utmost value to the astrophysicist and cosmochemist with regard to nucleosynthesis and the time-scale and evolution of the solar system.

Relevant Papers and Publications

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