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[1]D. E. Nagle *et al.*, in *High Energy Physics with Polarized Beams and Targets*, edited by G. H. Thomas, AIP Conference Proceedings No. 51 (American Institute of Physics, New York, 1978), p. 224.

[2]R. Balzer *et al.*, Phys. Rev. Lett. 44, 699 (1980).

[3]V. Brown, E. M. Henley, and F. R. Krejs, Phys. Rev. C 9, 935 (1975).

[4]M. Simonius, Phys. Lett. 41B, 415 (1972), and Nucl. Phys. A220, 269 (1974).

[5]B. Desplanques, J. F. Donoghue, and B. R. Holstein, Ann. Phys. (N.Y.) 124, 449 (1980).

[6]J. D. Bowman *et al.*, Phys. Rev. Lett. 34, 1184 (1975).

[7]H. L. Anderson *et al.*, in *High Energy Physics with Polarized Beams and Targets*, edited by M. L. Marshak, AIP Conference Proceedings No. 35 (American Institute of Physics, New York, 1976), p. 273; R. Talaga, Ph.D. dissertation, University of Chicago, 1976 (unpublished).

[8]See Ref. 1, p. 231; D. Alde, Ph.D. dissertation, University of Illinois, 1978 (unpublished).

[9]E. M. Henley and F. R. Krejs, Phys. Rev. D 1, 605 (1975).

[10]M. Simonius, R. Henneck, C. H. Jacquemart, J. Lang, W. Haeberli, and C. H. Weddigen, to be published.

[11]H. Frauenfelder, R. E. Mischke, and D. E. Nagle, unpublished.

Search for Superheavy Elements in the $^{238}U + ^{238}U$ Reaction

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A search was made for spontaneously fissioning superheavy elements in damped collisions of two uranium nuclei. Different techniques were applied covering the elements 108 to 118 and $\simeq 126$, and a half-life range from 1 ms to more than 1 yr. No evidence for superheavy elements was found at upper cross-section limits of 10^{-32}, 10^{-33}, and 10^{-35} cm^2 for half-lives from 1 to 100 ms, 100 ms to 1 d, and 1 d to 1 yr, respectively.

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In most attempts to produce superheavy elements around atomic number $Z = 114$ and neutron number $N = 184$, complete fusion reactions have been tried. Although a large variety of target-projectile combinations have been examined, no positive results have been reported thus far.[1] This may indicate[2] that in the region of neutron-deficient superheavy nuclei accessible to fusion, fission barriers are lower than calculated in most theoretical studies, so that production cross sections are smaller and half-lives shorter than expected.

An alternative pathway to the superheavy elements has been opened by the first studies of the interactions between two uranium nuclei.[3,4] The resulting element distribution clearly indicates[4] the production of elements around $Z = 70$ in damped collisions with cross sections of about 10^{-28} cm^2 at 7.5 MeV/u bombarding energy. If element 70 is formed in a binary collision from one of the $Z = 92$ colliding partners, element 114 is the complementary fragment. According to potential-energy considerations[5] that describe the neutron-to-proton ratios of fragments in damped collisions, $N = 182$ should be the most probable neutron number associated with $Z = 114$ fragments, but $N = 184$ fragments should also be formed with sizable cross section, about 10^{-29} cm^2, as a result of the dispersion[6] in neutron numbers.

Most of the superheavy fragments formed in damped collisions will decay by fission, but a small fraction originating with very low excitation energy may survive. The distribution of excitation energies of $Z = 112$ to 114 fragments has been deduced from Q-value measurements.[3] By folding this distribution with calculated[7] survival probabilities against fission, a production cross section of the order of 10^{-33} cm^2 for element 114 has been estimated.[3] A value of about 10^{-34} cm^2 results from a similar analysis of excitation-energy distributions calculated[8] with the diffusion model for damped collisions. Such

cross sections exceed by one to two orders of magnitude the detection limits attainable with sensitive techniques using high-intensity uranium beams available at the UNILAC accelerator.

Therefore, we have performed searches for superheavy elements by bombarding thick uranium metal targets with uranium beams and using (i) radiochemical off-line techniques, and (ii) a rotating-wheel on-line experiment. In this way, we have attempted to utilize the extreme sensitivity of radiochemical techniques for the detection of long-lived spontaneously fissioning nuclides and to cover, on the other hand, the half-life region between 1 ms and 1 yr with a sensitive technique that does not depend on the chemical behavior of superheavy elements.

In the radiochemical experiments, 300-mg/cm^2-thick targets[9] of uranium metal on water-cooled copper backings were bombarded with 8.6-MeV/u ^{238}U beams typically for five days in order to accumulate up to 4×10^{16} particles. All reaction products were stopped in the uranium layer. To collect volatile species which may evaporate during bombardment, a nickel or palladium cylinder was placed upstream from the target followed by a trap immersed in liquid nitrogen. Radon and polonium activities found in the trap and on the cylinder, respectively, show that evaporation losses of typical homologs of superheavy elements are below 10%.

After irradiation the targets were processed with two alternative chemical separation procedures.[10] In the gas-phase chemistry, the predicted volatility[11] of elements 112 through 117 in their elemental state is utilized. The solution chemistry is based on the expected formation of strong bromide complexes of elements 108 through 116.[12] In both procedures provisions were made to condense noble gases (element 118) and other superheavy elements that may be volatile at room temperature.[13] In one experiment, the procedure was extended in order to search for superactinide elements ($Z \geq 121$). Thus we can state that a broad spectrum of elements was covered in our experiments, including the alternative region[14] of nuclear stability near $Z = 126$.

In most cases the isolated fractions were deposited on thin foils and counted between two surface-barrier detectors for spontaneous fission events and α-particle spectra. The efficiency for recording both fission fragments in coincidence was about 60%. Two of these counter pairs were positioned inside a neutron multiplicity counter so that neutrons emitted in coincidence with fission fragments could be recorded. This counter equipped with 30 ^{3}He tubes has an efficiency of 32% for detection of a single neutron.

Table I summarizes the experimental conditions and, in the last four columns, the spontaneous fission events registered[15] in the principal samples. In addition, the collector foils for volatile species and several side fractions in the chemical procedures were also assayed. It is difficult to identify the source of the observed events at such low counting rates: Background measurements in four fission-fragment detector pairs over approximately two years gave one coincidence. The two events observed in an early experiment (No. 2) show quite high fragment energies as might be due to heavy actinides but no evidence for such nuclides was obtained by α-particle spectroscopy. The occurrence of high-energy fragments could not be reproduced at 5 to 6 times higher beam integrals (Nos. 3 and 4) while the chemical yields of homologs of volatile superheavy elements were comparable. Samples prepared by solution chemistry sometimes gave events with low energies because of an energy loss in visible deposits. Here a contamination by about 50 μg of ^{238}U would explain the observed count rates. Such a contamination cannot be excluded because problems were encountered with traces of colloidal uranium compounds. The number of neutrons recorded[15] in coincidence with fission fragments is compatible with known spontaneous fission activities.

Although we do not attribute any significance to the observed events we have estimated the upper cross-section limits for the production of spontaneously fissioning superheavy elements on the basis of two observed events per 4×10^{16} particles. If Poisson statistics is applied then these two events are still compatible with six events at 95% confidence level. We assume 60% counting efficiency, 75% yield in the chemical procedure as was typically measured for homologs of superheavy elements, a counting time of 200 d, and a constant excitation function between 8.6 MeV/u and the interaction barrier at 6.1 MeV/u corresponding to an effective number of 3.2×10^{19} atoms of ^{238}U per cm^2. The results, denoted "chem," are shown in Fig. 1 as a function of the assumed half-life for superheavy nuclei. If an excitation function with a maximum at 6.8 MeV/u and a decrease at higher energies is assumed as observed for the production of actinide isotopes,[4] these limits increase by a factor of 4.

In our second approach, rotating 11-mg/cm^2

TABLE I. Search for superheavy elements in the $^{238}U+^{238}U$ reaction.

Experiment[a]	Beam integral (10^{16} particles)	Chemistry	Net counting time (d)	Observed spontaneous-fission events[b] Singles $E_{1(2)}$	Singles E_1+E_2	Coincidences $E_{1(2)}+n$	Coincidences E_1+E_2+n
1	0.5	Solution	51[c]	•••	0	•••	•••
2	0.7	Gas phase	320[c]	•••	2	•••	•••
3	3.6	Gas phase	80	0	0	1	0
4	4.0	Gas phase	303[d]	0	•••	0	•••
		Solution	303	0	0	2	0
5	2.9	Solution	100[c]	•••	0	•••	•••
6	2.8	Solution	239	2	0	2	0
		Superactinides	121[d]	1	•••	1	•••
5+6	5.7	Gases	99[e]	2	•••	•••	•••
4	4.0	Cf-fraction[f]	148[d]	97	•••	267	•••

[a]Thick uranium metal targets; ^{238}U beam energy 8.6 MeV/u.

[b]$E_{1(2)}$ singles event with $E \geq 30$ MeV (3.4%); E_1+E_2 fragment-fragment coincidence (18%); $E_{1(2)}+n$ fragment-neutron coincidence (9%); E_1+E_2+n fragment-fragment-neutron coincidence (40%). Values in parentheses are absolute detection efficiencies measured with a thin ^{248}Cm source.

[c]Assayed for fission fragment coincidences only.

[d]Sample on thick backing exposed to only one fission fragment detector.

[e]Condensed on a copper cryostat; single fission fragment measurement.

[f]Measured to control the counter; main constituent is 60.5-d ^{254}Cf formed with a cross section of 6×10^{-34} cm^2, Ref. 4.

uranium metal targets were bombarded with 8.3-MeV/u ^{238}U beams. In three runs $(0.4-1.8)\times10^{16}$ particles were accumulated within about one day. The reaction products recoiling from the targets in an emission cone of ±55° (lab) were stopped in a stack of five 8-μm aluminum catcher foils. These foils were continuously rotated out of the beam in order to expose the collected activities to stationary plastic fission-track detectors positioned at both sides of each rotating foil. Three different velocities were chosen to cover various half-life ranges down to 1 ms. After bombardment the catcher foils were exposed to new track detectors. With this system one recorded fission event per 10^{16} beam particles corresponds to a cross section of 9×10^{-36} cm^2 for short-lived products decaying during the bombardment time.

Four spontaneous fission activities were observed: Two short-lived components with 1.1 ± 0.1 and 13 ± 5 ms half-lives, a product decaying largely during irradiation (100 ms $\leq T_{1/2} \leq$ 1 d), and a long-lived component ($T_{1/2} \gg 1$ d). These activities can be assigned to the fission isomers 1.1 ms ^{244f}Am and 14 ms ^{242f}Am, and to 2.6 h ^{256}Fm and 60.5 d ^{254}Cf formed with mean cross sections of 9×10^{-33}, 1.3×10^{-32}, 1.0×10^{-33}, and 4×10^{-33} cm^2, respectively. The latter two values agree with cross sections measured with radiochemical tech-

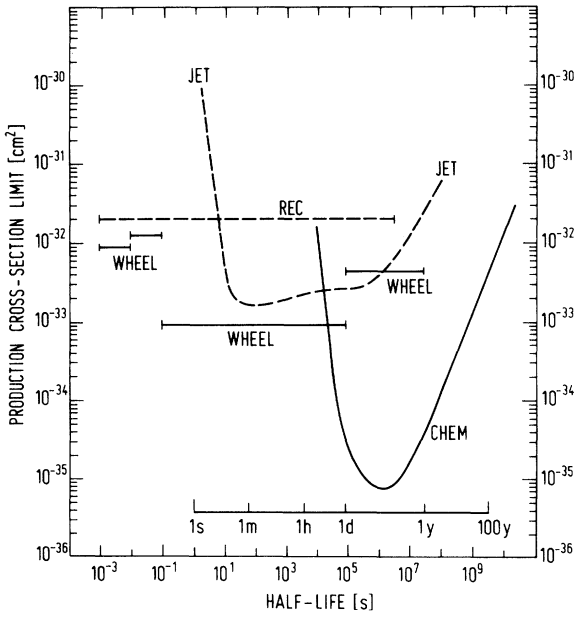

FIG. 1. Upper limits for the production cross section of spontaneously fissioning superheavy elements in the $^{238}U+^{238}U$ reaction. The limits denoted "chem" and "wheel" are from this work, those denoted "rec" from Ref. 3 and "jet" from Ref. 17. Note that the data have been obtained under somewhat different bombarding conditions.

niques[4] giving, thus, support for these assignments. For the two short-lived activities the observed cross sections appear to be somewhat lower than expectation values estimated by multiplying cross sections[4] for the ^{244}Am and ^{242}Am ground states with the ^{242}Am isomer-to-ground-state ratios found in other nuclear reactions.[16]

Hence, there is no evidence for the production of any spontaneous fission activity not attributable to actinides. We take the cross sections found for these four activities as an estimate for upper limits of the production of superheavy elements and show them denoted by "wheel" in Fig. 1. Also given are limits obtained by implanting recoil atoms in a surface-barrier detector ("rec")[3] and by transportation of recoil atoms to the detector arrangement with the gas-jet technique ("jet").[17] In contrast to the present experiment, spontaneously fissioning actinides have not been observed in these previous studies.

Our results, although negative, have increased the sensitivity limits for detection of superheavy elements in ^{238}U + ^{238}U collisions. Improvements are still conceivable, e.g., by an extension of radiochemical techniques to shorter half-lives. It is certainly important to attempt to increase the production rates, but bombardments at higher energies are not attractive since no gain in the production rates for fissionable heavy actinides has been observed.[4]

In summary, it is difficult to speculate on the possibility of producing superheavy elements in the laboratory because large half-life regions are still unexplored and because conceptional problems associated with both the complete fusion approach[2] and with the damped collision approach need further experimental and theoretical clarification.

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[1]See, e.g., G. Herrmann, Nature (London) 280, 543 (1979), and references therein.

[2]G. T. Seaborg, W. Loveland, and D. J. Morrissey, Science 203, 711 (1979).

[3]K. D. Hildenbrand *et al.*, Phys. Rev. Lett. 39, 1065 (1977); H. Freiesleben *et al.*, Z. Phys. A292, 171 (1979).

[4]M. Schädel *et al.*, Phys. Rev. Lett. 41, 469 (1978); M. Schädel, Gesellschaft für Schwerionenforschung Report No. 79-8, 1979 (unpublished).

[5]J. V. Kratz *et al.*, Nucl. Phys. A332, 477 (1979).

[6]J. Poitou *et al.*, Phys. Lett. 88B, 69 (1979).

[7]L. G. Moretto, Nucl. Phys. A180, 337 (1972).

[8]C. Riedel and W. Nörenberg, Z. Phys. A290, 385 (1979).

[9]H. Folger and U. Richter, Nucl. Instrum. Methods 167, 85 (1979).

[10]G. Herrmann, in *Proceedings of the International Symposium on Superheavy Elements, Lubbock, Texas, March 1978* (Pergamon, New York, 1978), p. 24.

[11]B. Eichler, Kernenergie 19, 307 (1976).

[12]J. V. Kratz, J. O. Liljenzin, and G. T. Seaborg, Inorg. Nucl. Chem. Lett. 10, 951 (1974).

[13]K. S. Pitzer, J. Chem. Phys. 63, 1032 (1975).

[14]D. Kolb, Z. Phys. A280, 143 (1977).

[15]The following coincidence events were observed: Experiment No. 2: 11 d, 88+124 MeV; 114 d, 101+53 MeV. No. 3: 24 d, 91 MeV, 1 *n*. No. 4: 145 d, >63 MeV, 1 *n*; 218 d, >23 MeV, 1 *n*. No. 6a: 19 d, >78 MeV, 0 *n*; 93 d, >23 MeV, 1 *n*; 292 d, >64 MeV, 0 *n*; 296 d, >34 MeV, 2 *n*. No. 6b: 110 d, >33 MeV, 1 *n*; 130 d, >112 MeV, 0 *n*, Nos. 5+6 (gases): 2 d, 111 MeV; 2 d, 88 MeV. Given above are the time of the event after the end of bombardment; the fragment energy corrected for minimum energy loss in the substrate (> indicates that correction cannot be estimated because of nonuniform sample thickness); and the number of neutrons in coincidence. The mean number of detected neutrons would be 0.65, 1.0, or 3.2, respectively, for an average of 2.0 (^{238}U), 3.15 (^{248}Cm), or 10 neutrons emitted per fission. The background coincidence event mentioned in the text gave 45+37 MeV.

[16]V. L. Kuznetsov *et al.*, Nucl. Phys. A324, 29 (1979).

[17]H. Jungclas *et al.*, Phys. Lett. 79B, 58 (1978); D. Hirdes *et al.*, Gesellschaft für Schwerionenforschung Annual Report No. GSI 79-11, 1978 (unpublished), p. 71; D. C. Aumann *et al.*, Phys. Lett. 82B, 361 (1979).