

SEARCH FOR SUPERHEAVY ELEMENTS IN DAMPED COLLISIONS OF ^{238}U WITH ^{238}U

Günter Herrmann

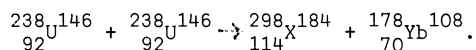
Institut für Kernchemie, Universität Mainz, D-6500 Mainz, and
Gesellschaft für Schwerionenforschung, D-6100 Darmstadt, FRG

Abstract - A search was made for spontaneously fissioning superheavy elements in damped collisions of two uranium nuclei. Different radio-chemical techniques and a rotating-wheel system were applied covering the elements 108 to 118 and ≈ 126 , and a half-life range from 1 ms to more than 1 y. No evidence for superheavy elements was found at upper cross-section limits of 10^{-32} , 10^{-33} , and 10^{-35} cm² for half-lives from 1 to 100 ms, 100 ms to 1 d, and 1 d to 1 y, respectively. First attempts to produce superheavy elements in the $^{238}\text{U} + ^{248}\text{Cm}$ reaction are briefly described.

INTRODUCTION

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 (Ref.1). This may indicate (Ref.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 first studies of the interactions between two uranium nuclei (Ref.3 & 4) which became possible after uranium beams of sufficient energy to overcome the Coulomb repulsion had been produced at the UNILAC heavy ion accelerator at Darmstadt. This approach is based on a novel type of nuclear reactions, the damped collision (Ref.5) in which both interacting nuclei stick together in a dinuclear system existing for a short time, about 10^{-22} to 10^{-20} s, dissipate kinetic energy and angular momentum, exchange nucleons, and separate again. Damped collisions lead to broad element and mass distributions in the reaction products peaking at or close to the atomic and mass number of the initial reaction partners and also to broad distributions in excitation energy and angular momentum. In damped collisions between two uranium nuclei the element and mass distributions may be broad enough to reach even the doubly-magic centre of the stability island:



Evidence that this takes place comes from radiochemical (Ref.4) and counter experiments (Ref.3 & 6) on the $^{238}\text{U} + ^{238}\text{U}$ reaction showing that fragments complementary to superheavy elements, i.e. with $Z \approx 70$, are formed in damped collisions. This does not necessarily demonstrate that superheavy elements in their ground state are also produced in detectable quantities. As will be discussed below, by far most of the superheavy fragments originate from such collisions with much too high excitation energy and angular momentum and, possibly, in much too elongated shapes to circumvent disintegration by fission. Only if the dispersions in excitation energy and angular momentum are sufficiently broad, the few superheavy fragments may have a chance to survive. Hence, only if there is sufficient overlap between the tails of the dispersions in atomic and mass number, excitation energy, angular momentum, and shape, there is a chance to produce detectable amounts of superheavy elements.

The present talk is a status report on a search for superheavy elements in the $^{238}\text{U} + ^{238}\text{U}$ reaction (Note a). Before the experimental techniques are outlined and the results obtained so far are described, some aspects of the reaction mechanism related to the survival probability of the heaviest fragments in $^{238}\text{U} + ^{238}\text{U}$ collisions are discussed. First attempts to

Note a. Survey of work by a GSI-Mainz collaboration: H. Gäggeler, N. Trautmann, J. V. Kratz, M. Schädel, W. Brühle, G. Herrmann, P. Peuser, G. Tittel, G. Wirth, H. Ahrens, H. Folger, G. Franz, K. Sümmerer and M. Zendel.

produce superheavy elements with the $^{238}\text{U} + ^{248}\text{Cm}$ carried out by an international collaboration are briefly described in the final section.

THE REACTION OF ^{238}U WITH ^{238}U

The distribution of nuclear mass and charge between the fragments in the reaction of ^{238}U with ^{238}U was studied in detail (Ref.4) after the bombardment of thick uranium metal targets with ^{238}U beams of 7.5 MeV/u incident energy. After bombardment the target was dissolved and the reaction products were chemically separated into 25 fractions which were assayed for x-ray, γ -ray, α -particle and spontaneous-fission activities. From these data energy-integral cross sections for individual isotopes with Z ranging from 26 to 100 were obtained. These were used to define a surface of independent formation cross sections in a Z-A plane. The data are condensed in Fig.1 in an element distribution obtained by integrating the cross sections for the isotopes (with mass numbers A) of each element Z. This distribution is

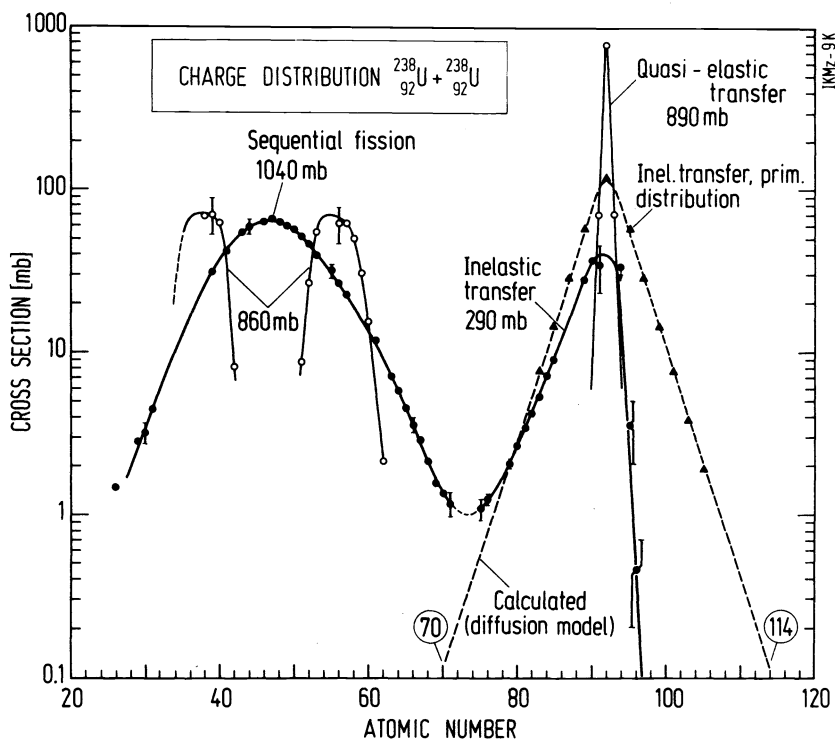


Fig. 1. Element distribution in the $^{238}\text{U} + ^{238}\text{U}$ reaction at 7.5 MeV/u incident energy: total production cross section of the isotopes of each element in thick-target bombardments as a function of the atomic number (Ref.4). The two main mechanisms contributing are quasi-elastic transfer reactions and sequential fission at low excitation energies (open circles), and damped inelastic collisions with the associated sequential fission at higher excitations (full circles). Also displayed are the reconstructed primary, pre-fission distribution (full triangles) and its extrapolation to Z=70, 114 (dashed curve) with the diffusion model for damped collisions (Ref.10).

interpreted (Ref.4) in terms of a superposition of four components: (i) a narrow symmetric distribution around uranium from quasi-elastic transfer of a few nucleons between the interacting nuclei (total cross section 890 mb), (ii) a broader distribution from $Z \approx 73$ to 100 arising from an originally symmetric distribution in the damped collision process sharply truncated beyond uranium by sequential fission (290 mb), (iii) a nearly symmetric, broad distribution originating from the sequential fission of highly excited primary reaction products (1040 mb), and (iv) a double-humped distribution of neutron-rich nuclei from sequential fission of primary fragments being formed with low excitation energy (860 mb). Taking into account that two, three or four fragments arise from a single collision, the sum of components (i) through (iv) represents the total reaction cross section, 1060 mb, and the

partial cross section for the damped collision process results as 405 mb from the sum of cross sections for components (ii) and (iii) corresponding to a fraction of 0.4 of the reaction cross section.

In order to deduce the primary, pre-fission element distribution for the damped collision process we use its integral cross section of 405 mb, and assume the distribution to be symmetric around $Z=92$ and to have the usual shape as found, e.g., in the $\text{Xe} + \text{Au}$ reaction (Ref.7). The width of the distribution is adjusted to the measured element yields near $Z=80$ where fission probabilities are negligible (Ref.6). The resulting primary distribution is indicated by triangles in Fig.1. The maximum of the surviving products lies close to $Z=91$ rather than at $Z=85$ or 80 as observed in the $\text{Xe} + \text{U}$ and $\text{Kr} + \text{U}$ reactions (Ref.8 & 9), respectively. Apparently, the sequential fission process is less important in the $\text{U} + \text{U}$ system. This is also evident in the counter experiments (Ref.6) which show that given elements in the uranium environment are produced at smaller average excitation energies in $\text{U} + \text{U}$ collisions than in the other reactions. Also shown in Fig. 1 (dashed curve) is an extrapolation to $Z=70$, 114 based on a semiphenomenological theory in which the exchange of nucleons between interacting nuclei in damped collisions is treated as a diffusion process (Ref.10).

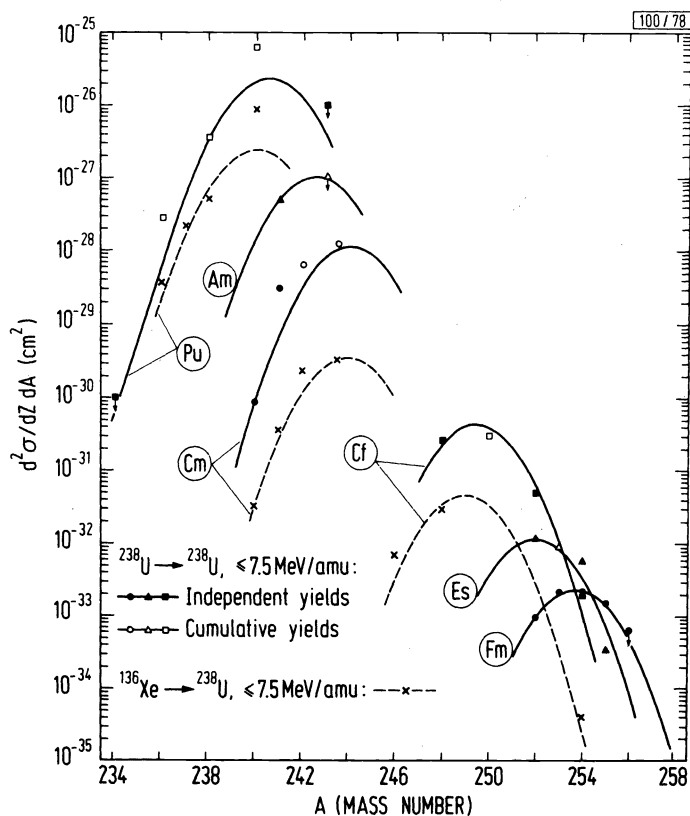


Fig. 2. Cross sections for the formation of heavy actinide isotopes in the $^{238}\text{U} + ^{238}\text{U}$ reaction at 7.5 MeV/u incident energy in thick-target bombardments (solid curves). For comparison, similar data for the $^{136}\text{Xe} + ^{238}\text{U}$ reaction at 7.5 MeV/u incident energy are given (dashed curves). From (Ref.4).

This extrapolation to element 70 of the primary distribution for damped collisions results in a formation cross section of 10^{-28} cm². Primary fragments of element 114 should have the same cross section. The most probable neutron number associated with $Z=114$ fragments can be deduced to be close to $N=182$ from the potential energy available for different distributions of the neutrons in a $Z=114$, $Z=70$ split as follows from the Q_{gg} -systematics with pairing corrections which was found to explain the experimental N/Z -ratios in the $\text{Xe} + \text{Au}$ reaction (Ref.7) and in many other reactions (Ref.11). Taking into account the dispersion in the neutron distribution (Ref.12), a cross section in the order of 10^{-29} cm² is estimated for the doubly-magic configuration $Z=114$, $N=184$ at 7.5 MeV/u incident energy.

But what fraction of these heavy fragments around $Z=114$ may have a chance to deexcite without undergoing fission? Is it not more likely that fragments from damped collisions are formed in very elongated shapes beyond the saddle point shape for fission so that they quickly undergo fission rather than achieve the spherical shape in which stabilization by proton and neutron shell closures becomes most effective?

One may speculate about this question with an analysis of the steep decrease in the element yields beyond uranium, see Fig. 1, followed with radiochemical techniques over eight orders of magnitude up to fermium, $Z=100$ (Ref.4). The experimental cross sections are shown in Fig. 2 where they are compared to thick target cross sections in the $^{136}\text{Xe} + ^{238}\text{U}$ reaction. The enhancement of cross sections in the $^{238}\text{U} + ^{238}\text{U}$ reaction is obvious. It reflects again, at least qualitatively, that nucleon transfer proceeds "colder" (Ref.3 & 6) in the $^{238}\text{U} + ^{238}\text{U}$ reaction than in other reactions. In order to learn whether the experimentally determined survival probabilities for the heaviest actinides are compatible with the usual statistical deexcitation of excited heavy nuclei, we analyzed (Ref.4) the cross sections for the complementary element pairs radon-californium ($Z=86,98$), astatine-einsteinium ($85,99$),

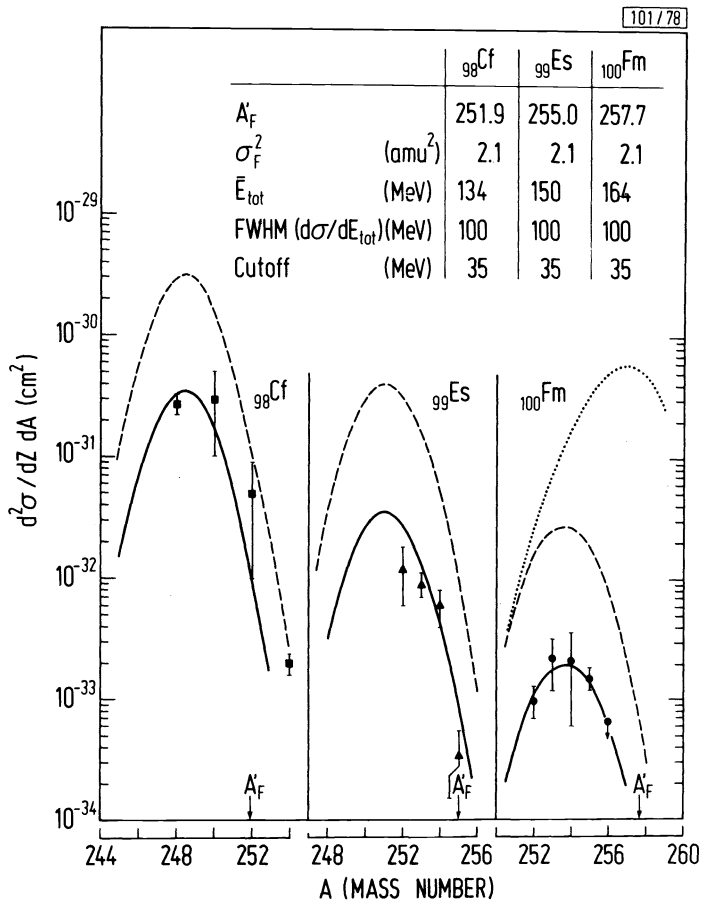


Fig. 3. Comparison of measured (symbols) and calculated isotope populations (curves) for the heaviest actinide elements in the $^{238}\text{U} + ^{238}\text{U}$ reaction. The calculations are outlined in (Ref.4). A'_F is the most probable mass number and σ_F^2 is the variance of the primary mass distribution for a given element, \bar{E}_{tot} is the mean total excitation energy of the light and heavy fragments and $d\sigma/dE_{\text{tot}}$ accounts for its dispersion, and "cutoff" gives the minimum energy which has to be dissipated in collisions leading to a given actinide element.

and polonium-fermium ($84,100$). With some straightforward assumptions and with data from counter experiments (Ref.3 & 6), the primary distributions for the lighter complements could be reconstructed as well as their excitation energy distributions, and could be transformed

into the respective distributions of the heavier complements. Starting from there, the expected isotope cross sections of the heavy complements were calculated taking into account (Ref.13) the competition between neutron evaporation and fission. The calculations show that the surviving transcurium isotopes originate exclusively from the low-energy tails of the excitation energy distributions. In Fig. 3 the measured cross sections for californium, einsteinium and fermium isotopes are compared with calculated distributions using parameters as indicated in the Figure. The dotted curve for fermium represents the case where the total excitation energy is shared between the fragments in proportion to their masses. Since the experimental isotope distribution is centered at a lower mass number we concluded that a minimum energy dissipation is required for given charge transfers and took care of this by introducing an energy cutoff. This yields the dashed curves in Fig. 3 which reproduce the position and width of the experimental distributions but still fail to reproduce the absolute cross sections by one order of magnitude. Agreement between experiment and our estimate is obtained (see solid curves in Fig. 3) if, starting with $Z_1 \geq 98$ ($Z_1 \leq 86$), it is assumed that the total excitation energy in the low-energy collisions is more and more concentrated in the heavy fragment. This may be related to the simultaneous formation of a nearly magic light fragment ($Z=82$). One may speculate that this effect might reverse for the formation of fragment pairs near $Z=114$, 70 in the low-energy tails of the excitation energy distributions, a feature highly desirable for the production of superheavy elements. Similar conclusions were drawn in (Ref.6).

Of course, the indications for shell effects are very indirect. A more pessimistic view would relate the low experimental californium, einsteinium and fermium yields to a stronger fission competition than expected according to the empirical systematics (Ref.13). An increase by a factor up to three in the fission probability would remove the discrepancy. This may be caused by angular momentum effects which were not taken into account, or because a large fraction ($\approx 90\%$) of the californium, einsteinium and fermium fragments is formed with deformations at or beyond the critical saddle-point shape for prompt fission.

Some hint for the latter process has been obtained in investigations of three-body exit channels in $^{238}\text{U} + ^{238}\text{U}$ and $^{238}\text{U} + ^{248}\text{Cm}$ collisions using large-area position-sensitive counters (Ref.14). Even for mass reactions leading to fragments with $Z > 110$, the data are still compatible with a two-step reaction mechanism where the heavy fragment from the deep inelastic collision exists at least for times of 10^{-20} sec until it fissions. However, the observed fission fragment angular distributions and broad mass distributions are quite different from what is normally observed in fission of heavy nuclei at such excitation energies, and may be explained by fission from very elongated shapes. It is important to note that these results refer to heavy fragments with much higher excitation energy than allowed for survival. Whether reactions at correspondingly lower excitations proceed also via unusual configurations remains an open question.

In order to learn how the yields of very heavy nuclei depend on the energy of the uranium projectile, radiochemical cross-section measurements with thick uranium targets were also performed at 6.49, 6.84, and 8.65 MeV/u incident energy. Within the experimental uncertainties, the centroids and widths of the isotope distributions seem to be independent of the incident energy. These data were used to derive excitation functions for transcurium elements in the $^{238}\text{U} + ^{238}\text{U}$ reaction from the differences of the thick-target production rates at subsequent energy steps, i.e. for the energy intervals between 8.65 and 7.50, 7.50 and 6.84, 6.84 and 6.49, and 6.49 and 6.10 MeV/u, the latter energy corresponding to the interaction barrier. Figure 4 shows the result for californium, integrated over all isotopes. These excitation functions agree in that there is a steep increase in the cross sections up to about 6.6 MeV/u and then a decrease at the higher incident energies whereas the cross sections for the lighter complementary elements as radon increase with increasing energy. These observations again support the conclusion that, at each projectile energy, only the low-energy tails of the energy dispersions are of importance for the survival of very heavy fragments. At higher incident energies, the enhanced mass transfer which is caused by the stronger penetration of the colliding nuclei starts to be overcompensated for incident energies beyond ≈ 6.6 MeV/u by the higher excitation energies involved which drastically reduce the survival probability of the product nuclei. This is illustrated by the insert in Fig. 4 giving the cross-section ratio for the complementary element pair californium-radon as a measure for the average survival probability of californium isotopes. This ratio is found to decrease exponentially with increasing projectile energy.

The fraction of element 114 nuclei that may survive after $^{238}\text{U} + ^{238}\text{U}$ collisions can be estimated by folding the excitation energy distributions for element 114 nuclei with the energy-dependent survival probabilities which have been calculated (Ref.15). This approach implies the assumptions that element 114 nuclei originate from damped collisions with nearly spherical shapes and that fission and neutron evaporation compete statistically in the deexcitation of the excited primary fragments. Again, as for heavy actinides, only the low-energy tails of the energy distributions are found to be relevant for survival. If excitation energies derived from the counter experiment (Ref.3 & 6) are used in this analysis a total cross section in the order of 10^{-33} cm² results for element 114 nuclei (Ref.6)

whereas a value of 10^{-34} cm² is obtained with excitation energy distributions calculated (Ref.10) with the diffusion model for damped collisions. Such cross sections exceed by one to two orders of magnitude the detection limits attainable with high-intensity uranium beams

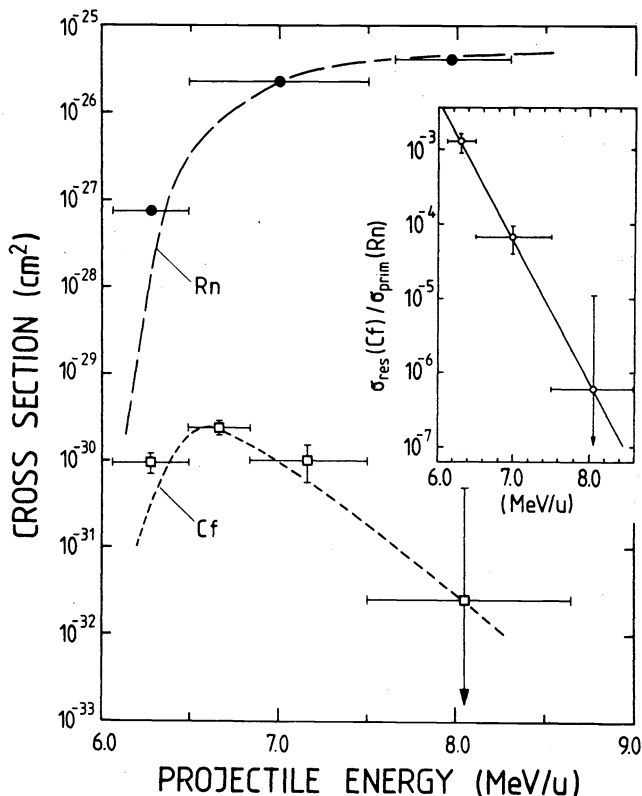


Fig. 4. Integral cross sections for all isotopes of the complementary fragment elements californium (Z=98) and radon (Z=86) in the $^{238}\text{U} + ^{238}\text{U}$ reaction plotted versus projectile energy. The radon cross sections are corrected for a small loss by sequential fission. The cross section ratio (insert) is a measure for the average survival probability against prompt fission of californium fragments with increasing excitation energy.

available at the UNILAC accelerator if techniques are applied which should allow the detection of superheavy elements even at a few-atom scale. However, although the observed actinide cross sections in the $^{238}\text{U} + ^{238}\text{U}$ reaction provide some evidence for shell effects in the sharing of excitation energies between fragments which would increase the cross section for element 114, these estimates might rather be considered as upper limits because of not yet fully explored effects working in the opposite direction: the role of extreme deformations and of high angular momenta in the reaction and - most important - the uncertainty in the theoretically calculated fission-barrier heights. Notwithstanding, the $^{238}\text{U} + ^{238}\text{U}$ reaction appears to be very attractive for attempts to produce superheavy elements.

SEARCH FOR SUPERHEAVY ELEMENTS IN THE $^{238}\text{U} + ^{238}\text{U}$ REACTION

In view of such cross section estimates for the production of surviving superheavy elements direct searches were performed during the past three years by bombarding thick uranium metal targets with uranium beams 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 y with a sensitive technique that does not depend on the chemical behaviour of superheavy elements. By the application of thick targets we have assured that the whole range of projectile energies from

the incident energy down to the interaction barrier is available thus covering the optimum energy range as long as it falls within this region.

Radiochemical methods

In the radiochemical experiments, 300 mg/cm^2 thick targets (Ref.16) of uranium metal on water-cooled copper backings were bombarded with $8.6 \text{ MeV/u } ^{238}\text{U}$ 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 the 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 volatile elements are below 10%.

After irradiation the targets were processed with two chemical separation procedures based on entirely different principles with regard to the chemical behaviour of superheavy elements. In the gas-phase chemistry, the predicted volatility (Ref.17) of elements 112 through 117 in their elemental state is utilized. The other procedure, the solution chemistry which should cover elements 108 through 116, is based on the expected formation of strong anionic bromide complexes as is utilized in a cation exchange procedure (Ref.18), and on the expected strong affinity to sulfur used in the solvent extraction with diethyldithiophosphate $(\text{EtO})_2\text{PSSH}$ (Ref.19). In both procedures provisions were made to condense noble gases (element 118) and other superheavy elements that may be volatile at room temperature (Ref.20) on a copper cryostat. In one experiment, the procedure was extended in order to search for super-actinide elements ($Z \geq 121$). Thus we can state that a broad spectrum of elements was covered in our experiments, including the alternative region (Ref.21) of nuclear stability near $Z=126$.

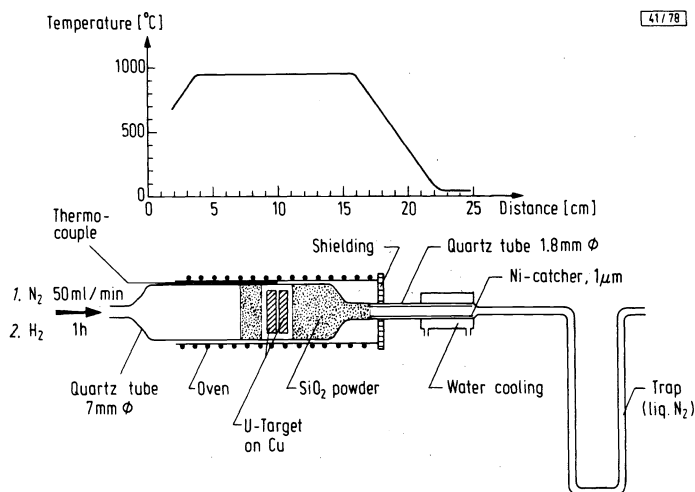


Fig. 5. Set-up for the gas-phase chemistry to volatilize and collect super-heavy elements in the elementary states (bottom) and temperature distribution along the quartz tube (top).

Figure 5 illustrates the gas-phase chemistry. The irradiated uranium target was placed in a quartz tube and heated in a nitrogen stream up to $950 \text{ }^\circ\text{C}$ in order to convert the thick metallic target into fine-dispersed uranium nitrides. From this matrix volatile species were evaporated in hydrogen gas at $1000 \text{ }^\circ\text{C}$ for several hours to ensure quantitative recovery of typical homologues of superheavy elements. By isothermal gas chromatography at $900 \text{ }^\circ\text{C}$ separation from actinides and uranium was achieved. The transported volatile elements were deposited on thin nickel or silver foils kept in a steep temperature gradient. The foils were finally mounted for counting with surface barrier detectors. Gaseous species passing the collector foils were collected in a charcoal trap immersed in liquid nitrogen.

Figure 6 gives an outline of the solution chemistry. During dissolution of the bombarded uranium target in strong hydrobromic acid, escaping gases which may contain element 118 (and possibly 112 and 114) were collected in charcoal which was later counted in a neutron-multiplicity counter. The solution in dilute hydrobromic acid was passed through a cation exchange column which was eluted with dilute hydrobromic acid. Superheavy elements should be contained in the column effluent and in this first eluate. These fractions were combined

and extracted with $(\text{EtO})_2\text{PSSH}$, and the organic as well as the aqueous phases were evaporated onto thin foils for counting with surface barrier detectors. The organic phase should preferentially contain elements 111 to 116, the aqueous phase elements 108 to 110. To obtain the production cross sections of nuclides in the vicinity of uranium, the cation exchange column was further eluted with hydrochloric acid and oxalic acid. The fraction containing plutonium, the actinides and lanthanides was purified with ion exchange columns and finally separated by high-pressure liquid-liquid extraction chromatography (Ref.22) into several fractions of transplutonium elements.

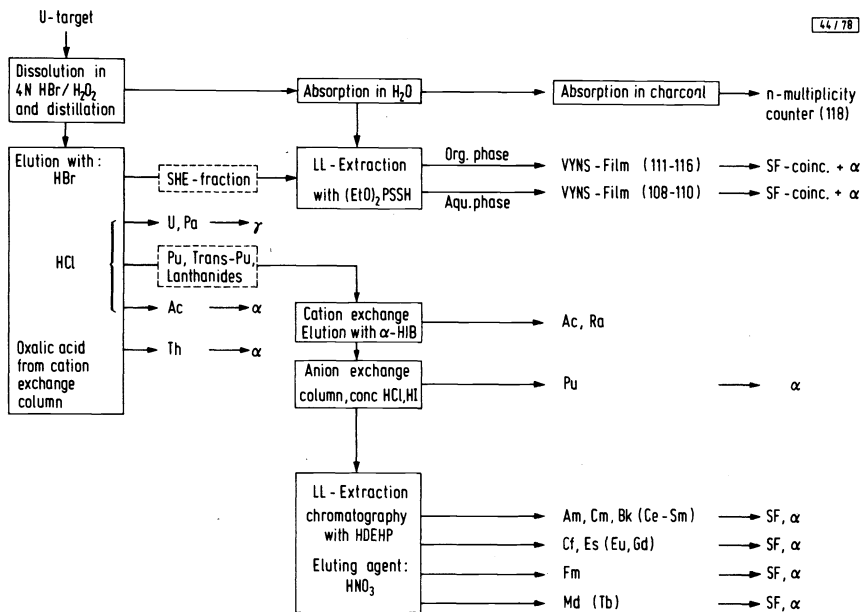


Fig. 6. Schematic flow-diagram of the solution chemistry applied to separate superheavy and actinide elements from thick uranium metal targets.

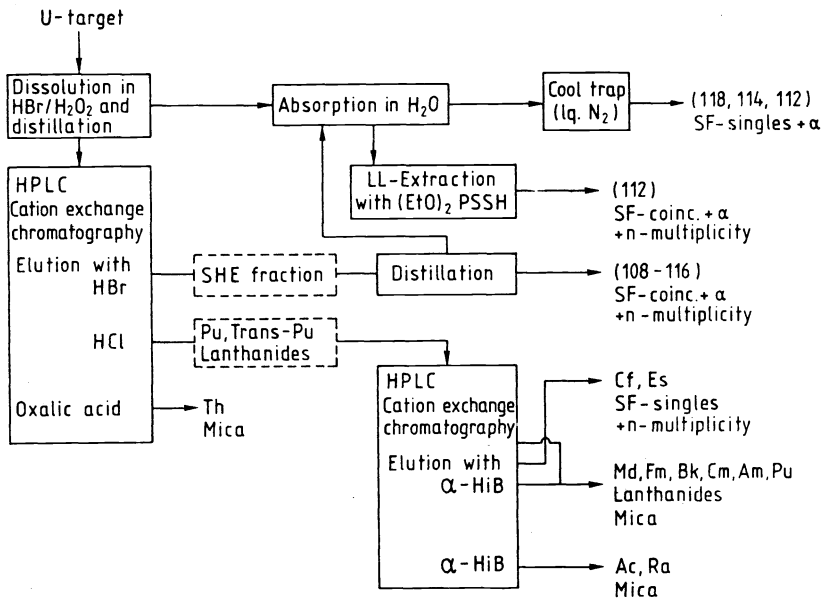


Fig. 7. Schematic flow-diagram of the solution chemistry modified to condense volatile species on a copper cryostat and to prepare fractions that should contain any member of the superactinide elements series.

After bombardments 3 and 4 (see below) the target was first processed with the gas-phase chemistry; then the target material was dissolved and further processed with the solution chemistry. In bombardments 5 and 6, where only the solution chemistry was used, the chemistry scheme was modified as shown in Fig. 7 in order to condense volatile species escaping during dissolution of the target on a copper cryostat faced by a surface barrier detector, and to prepare additional counting samples of chemical fractions that are expected to contain any member of the superactinide series. These latter samples were inspected with mica fission-track detectors and, later, with surface barrier detectors.

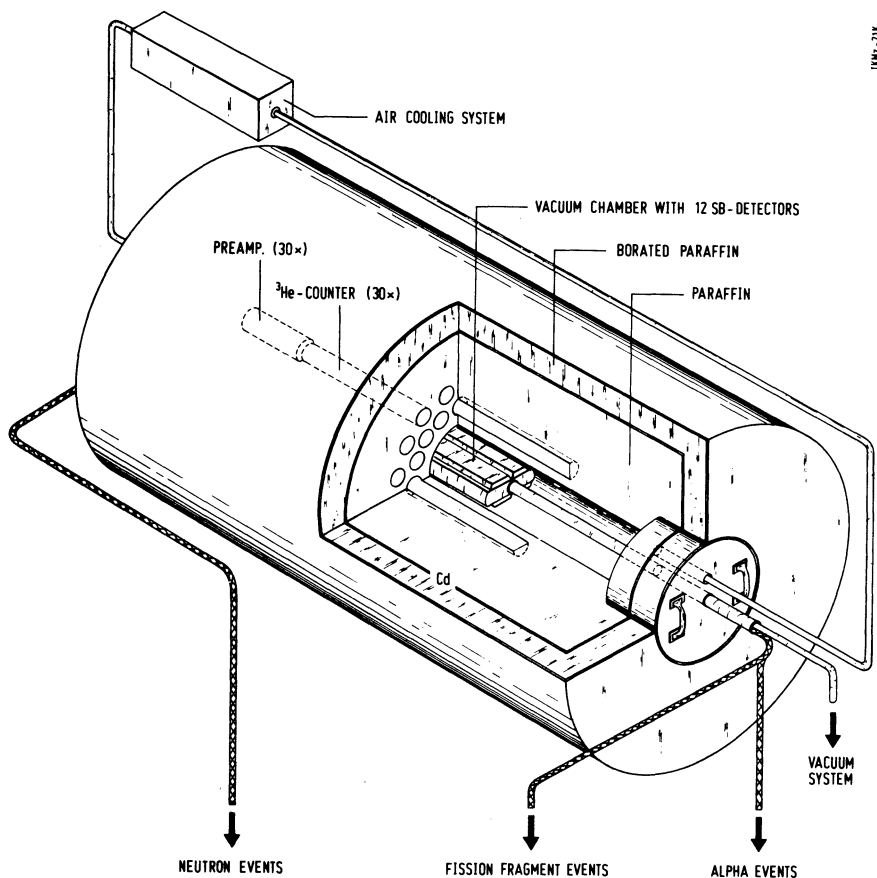


Fig. 8. Schematic view of the fission-fragment neutron-multiplicity counter. The sample deposited on a thin backing is placed between one of the six large-area surface barrier (SB) detector pairs which record fission-fragment and α -particle energies. Whenever a fission fragment triggers the counter neutrons emitted from fission fragments are counted after slowing down in paraffin in a system of 30 ^3He counters surrounding the sample region. This version of the counter was used in the $^{238}\text{U} + ^{248}\text{Cm}$ experiment, a preceding version with two detector pairs in the $^{238}\text{U} + ^{238}\text{U}$ experiments.

In most cases the isolated superheavy element fractions were deposited on thin foils and counted between two large-area surface barrier detectors for spontaneous fission events and α -particles. The efficiency for recording both fission fragments in coincidence was about 60%. For experiments 3 through 6 two of these counter pairs were positioned inside a neutron multiplicity counter, Fig. 8, so that neutrons emitted in coincidence with fission fragments could be recorded. This counter equipped with 30 ^3He tubes has an efficiency of 32% for detection of a single neutron.

Rotating wheel system

The rotating wheel system used in our second approach is shown in Fig. 9. A rotating 11 mg/cm^2 thick uranium metal target was bombarded in a vacuum chamber and the reaction

products recoiling from the target under an emission cone of $\pm 55^\circ$ (laboratory system) were stopped in a stack of five 8μ thick aluminium catcher foils. During bombardment 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 of the aluminium catcher foils were chosen to cover various half-life ranges down to 1 ms. After bombardment the catcher foils were exposed to fresh track detectors for the detection of long-lived spontaneous fission activities. For the foils exposed on-line, the number of tracks recorded at increasing angles from the target position were plotted versus the decay time obtained from the rotational frequency.

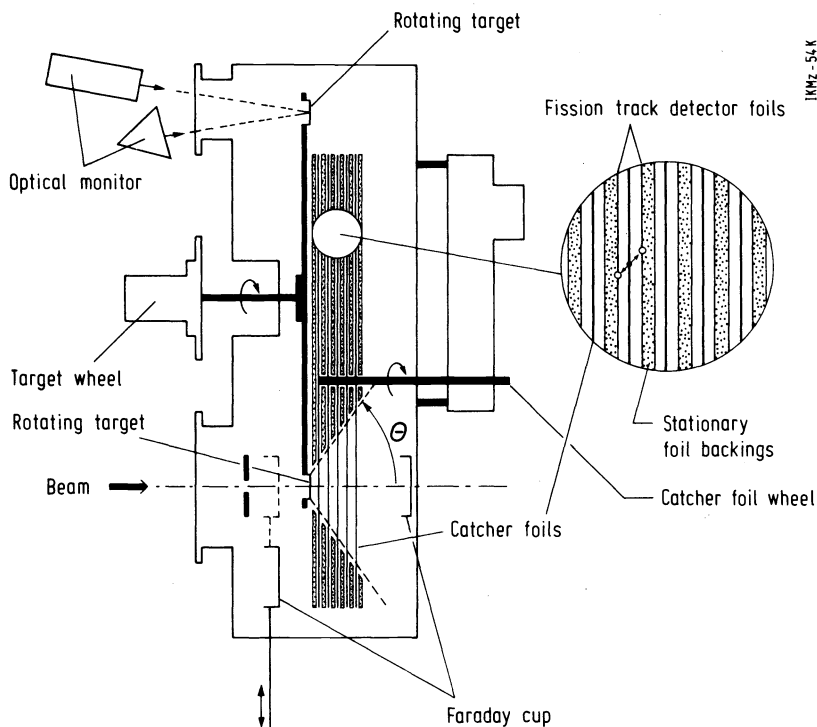


Fig. 9. Schematic view of the rotating wheel system for on-line detection of short-lived spontaneous fission activities in damped collisions. The target rotates in a vacuum chamber. Reaction products recoiling from the target under an emission angle θ are stopped in a stack of aluminium catcher foils which continuously rotate in order to expose the collected activities to stationary plastic fission-track detectors positioned at both sides of each rotating foil.

Results

Six bombardments were performed in the series of radiochemical experiments. Experimental conditions and the spontaneous fission events registered in the principal samples are summarized in Table 1. Further information about these events is given in Table 2. In addition to the samples listed in Tables 1 and 2, the collector foils and traps for volatile species placed close to the targets during bombardment 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 (45+37 MeV). The two events observed in an early experiment (No.2) show quite high fragment energies (Table 2) as might be due to heavy actinides but there was no evidence for such nuclides in the simultaneously taken α -particle spectra. The occurrence of such high-energy fragments could not be reproduced at five to six times higher beam integrals (Nos. 3+4) while the chemical yields of homologues of volatile superheavy elements were comparable. Samples prepared by solution chemistry gave sometimes events with low energies due to an energy loss in visible deposits. Here a contamination by about $50 \mu\text{g}$ of ^{238}U would explain the observed count rates. Such a contamination cannot absolutely be excluded because problems were encountered with traces of colloidal uranium compounds which irregularly pass the ion exchange column into the superheavy element fraction. The two events recorded in the condensed noble gas fraction (Nos. 5+6) may correspond to the usual electronic background of

TABLE 1. Search for superheavy elements in the $^{238}\text{U} + ^{238}\text{U}$ reaction

| Experiment ^{a)} | Beam integral in 10^{16} particles | Chemistry | Net counting time in d | Observed spontaneous-fission events ^{b)} | | | |
|--------------------------|--------------------------------------|---------------------------|------------------------|---|-----------|---------------------------|-------------|
| | | | | Singles $E_{1(2)}$ | E_1+E_2 | Coincidences $E_{1(2)+n}$ | 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 |
| 4A | 4.0 | Gas-phase | 303 ^{d)} | 0 | - | 0 | - |
| B | | Solution | 303 | 0 | 0 | 2 | 0 |
| 5 | 2.9 | Solution | 100 ^{c)} | - | 0 | - | - |
| 6A | 2.8 | Solution | 239 | 2 | 0 | 2 | 0 |
| B | | Super-actinides | 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 system; main constituent is 60.5-d ^{254}Cf formed with a cross section of 6×10^{-34} cm^2 (Ref.23).

TABLE 2. Spontaneous fission events observed in the $^{238}\text{U} + ^{238}\text{U}$ reaction

| Experiment | Time after irradiation in d | Fragment energy in MeV ^{a)} | | Number of neutrons ^{b)} |
|------------|-----------------------------|--------------------------------------|----------|----------------------------------|
| | | E_1 | E_2 | |
| 2 | 11 | 88 | 94 (124) | - |
| | 114 | 101 | 23 (53) | - |
| 3 | 24 | 91 | 0 | 1 |
| 4B | 145 | >63 | 0 | 1 |
| | 218 | 0 | >23 | 1 |
| 6A | 19 | >78 | 0 | 0 |
| | 93 | 0 | >23 | 1 |
| | 292 | >64 | 0 | 0 |
| 6B | 296 | >34 | 0 | 2 |
| | 110 | >33 | - | 1 |
| | 130 | >112 | - | 0 |

a) Measured fragment energy in the upper (E_1) and lower (E_2) detector with energy (in parentheses) corrected for minimum energy loss in the sample backing; > indicates that correction cannot be estimated because of unknown sample thickness.

b) Neutron number counted in coincidence with fission fragments. The average number recorded 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.

a single surface barrier detector. The number of neutrons counted in coincidence with fission fragments (Table 2) 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 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 homologues 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. 10 as a function of the assumed half-life for superheavy nuclei. The variation in cross sections is a result of decay during the chemical separations (usually a few hours) for short half-lives, and incomplete saturation and decay for half-lives comparable to or longer than the total counting time. If an excitation function with a maximum at 6.6 MeV/u and a decrease at higher energies is assumed as observed for the production of actinide isotopes (Fig. 4), the "chem" limits increase by a factor of four.

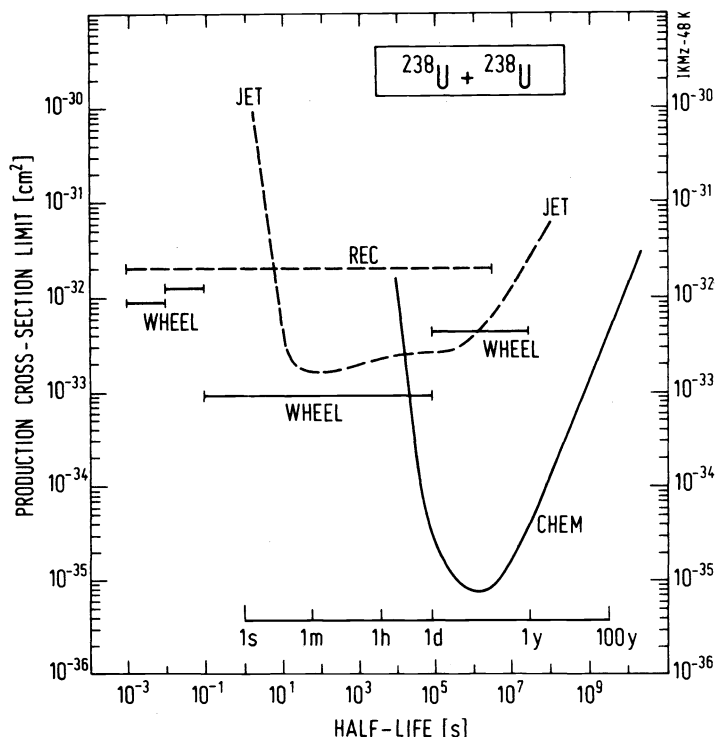


Fig. 10. Upper limits for the production cross section of spontaneously fissioning superheavy elements in the $^{238}\text{U} + ^{238}\text{U}$ reaction. The limits denoted "chem" and "wheel" are from work summarized in this contribution, those denoted "rec" are from (Ref.3 & 6) and "jet" from (Ref.25,26,27). Note that the data have been obtained under somewhat different bombarding conditions with regard to projectile energy and target thickness.

Also included in Fig. 10 are the results, denoted "wheel", of three runs with the wheel system in which $(0.4-1.8) \times 10^{16}$ ^{238}U particles of 8.3 MeV/u energy were accumulated on the targets within about one day. As is illustrated by the decay curves shown in Fig. 11, four spontaneous fission activities were observed: Two short-lived components with 1.1 ± 0.1 ms and 13 ± 5 ms half-life, a product decaying largely during irradiation ($100 \text{ ms} \leq T_{1/2} \leq 1 \text{ d}$), and a long-lived component ($T_{1/2} \gg 1 \text{ d}$). These activities can be assigned to the fission isomers $1.1\text{-ms } ^{244\text{m}}\text{Fm}$ and $14\text{-ms } ^{242\text{m}}\text{Fm}$, and to $2.6\text{-h } ^{256}\text{Fm}$ and $60.5\text{-d } ^{254}\text{Cf}$ formed with mean cross sections of $9 \times 10^{-33} \text{ cm}^2$, $1.3 \times 10^{-32} \text{ cm}^2$, $1.0 \times 10^{-33} \text{ cm}^2$, and $4 \times 10^{-33} \text{ cm}^2$, respectively. The latter two values agree with cross sections measured with radiochemical techniques (Ref.23) 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 (Ref.23) for the ^{244}Am and ^{242}Am ground states with the ^{242}Am isomer-to-ground-state ratios found in other nuclear reactions (Ref.24). Hence, there is no evidence for the production of any spontaneous fission activity not attributable to known actinide nuclides. We take the cross-sections measured for these four activities as an estimate for upper limits of the production of superheavy elements as they are shown in Fig. 10. Also given in Fig. 10 are limits obtained (Ref.3 & 6) by implanting recoil atoms in a surface barrier detector ("rec") and by transportation of recoil atoms to the detector arrangement with the gas-jet technique ("jet") as reported in (Ref.25,26,27). In contrast to these previous studies, the sensitivity limit set by the background due to presence of spontaneously fissioning actinides in unseparated reaction products has been reached in the present experiment.

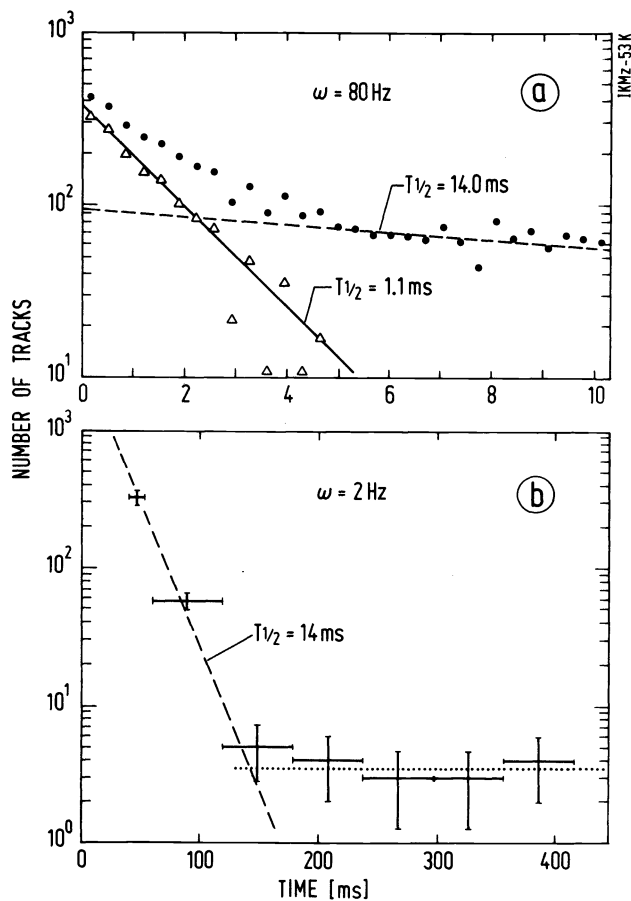


Fig. 11. Decay-curves of spontaneous fission activities observed with the rotating wheel system in the $^{238}\text{U} + ^{238}\text{U}$ reaction at 8.3 MeV/u incident energy. Parts a) and b) refer to two different time regions as determined by the rotational frequency.

Conclusions

Our experimental efforts to search for surviving superheavy nuclei in the uranium-on-uranium reaction, although negative thus far, have established new limits for the production cross section. Further improvements are still conceivable, e.g. by an extension of radiochemical experiments to shorter-lived species. It is certainly important to attempt to increase production rates, but bombardments at higher energies appear not to be attractive since no gain in the production rates for fissionable heavy actinides has been observed. More promising is the use of heavier targets: in the $^{238}\text{U} + ^{248}\text{Cm}$ reaction, the production cross section for surviving superheavy elements should increase by two orders of magnitude compared to the $^{238}\text{U} + ^{238}\text{U}$ reaction, as follows from calculations with the diffusion model (Ref.10).

FIRST ATTEMPTS WITH THE $^{238}\text{U} + ^{248}\text{Cm}$ REACTION

In October 1979 a Berkeley-Darmstadt-Livermore-Mainz-Oak Ridge collaboration (Note b) joined for the first bombardments of thick ^{248}Cm targets with ^{238}U beams at the UNILAC. The targets were prepared by evaporation of ^{248}Cm metal on 4.5 mg/cm^2 thick molybdenum foils. Target thicknesses between 3.5 and $7 \text{ mg } ^{248}\text{Cm/cm}^2$ were used. These targets contain a spontaneous fission activity of about 2×10^9 events/day. The target set-up is shown in Fig. 12. Accelerator and beam line were protected by a molybdenum entrance window. Window and target were cooled by a carefully purified, fast nitrogen gas flow while the target was additionally cooled by a helium gas jet. The target temperature was monitored on-line with an infrared

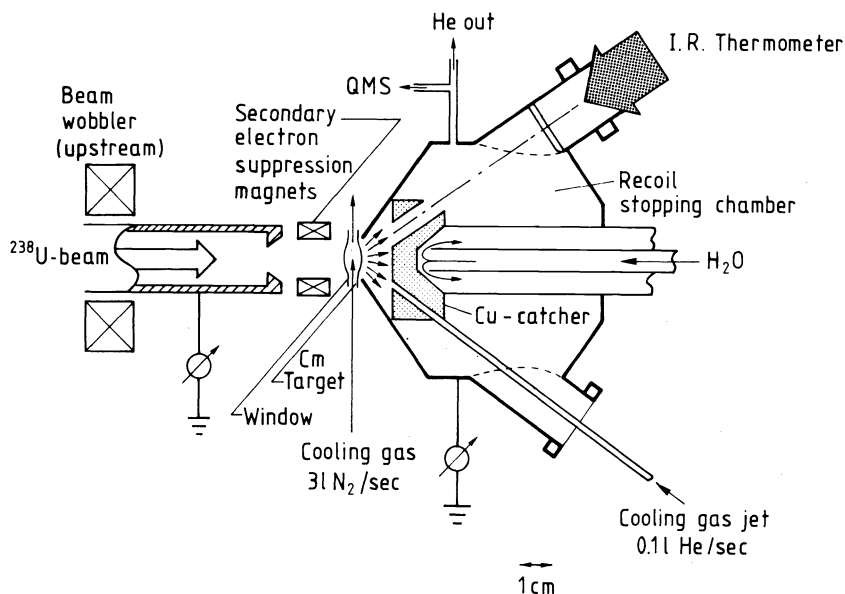


Fig. 12. Target apparatus for the $^{238}\text{U} + ^{248}\text{Cm}$ experiments (schematically).

thermometer. The outgoing helium gas was continuously assayed for nitrogen by a quadrupole mass spectrometer. A sudden increase of the signal for mass 28 indicated that the target plus backing had developed a hole so that nitrogen entered the recoil chamber. The ^{238}U beam was spread over the whole target area by a beam wobbler. In case of a break of the target and window a fast acting valve would prevent contamination of the accelerator. A failure of any of these safety measures produced an interlock signal that shut off the beam in less than 1 ms. Also included in the interlock conditions was the occurrence of beam spikes exceeding a preset value of the peak current.

A water-cooled recoil-atom catcher was inserted into the recoil chamber (see Fig.12) to stop any reaction products escaping under laboratory angles of $\pm 55^\circ$. The beam energy was 10.0 MeV/u in front of the entrance window and 7.4 MeV/u incident to the target where it was further degraded below the interaction barrier. After bombardment the copper catcher was removed and processed chemically, first with the gas-phase chemistry and then with the usual solution chemistry. Somewhat faster chemical separations of the transcurium elements were also performed after several short bombardments (1-3 hours). Production cross sections for actinide isotopes are presented and discussed in a separate contribution to this conference (Ref.28).

After an irradiation with a beam integral of 3×10^{15} particles, several fractions were isolated. Counting is still being continued. Preliminary upper limits for the production

Note b: Participants are J. M. Nitschke (Lawrence Berkeley Laboratory), W. Bröchle, H. Gäggeler, J. V. Kratz, M. Schädel, K. Sümmerer, G. Wirth (GSI Darmstadt), E. K. Hulet, R. W. Lougheed (Lawrence Livermore Laboratory), N. Trautmann, G. Herrmann, P. Peuser, R. Stakemann, G. Tittel (University of Mainz), R. L. Hahn and R. L. Ferguson (Oak Ridge National Laboratory).

cross section of superheavy elements are displayed in Fig. 13. The data were derived with similar assumptions to that for the $^{238}\text{U} + ^{238}\text{U}$ reaction except for the effective number of target atoms which is now $1.5 \times 10^{19} \text{ } ^{248}\text{Cm}$ atoms/cm². The shift to 10^{-34} cm^2 in the $^{238}\text{U} + ^{248}\text{Cm}$ reaction compared to the 10^{-35} cm^2 in the $^{238}\text{U} + ^{238}\text{U}$ reaction (Fig.10) is mainly due to the lower beam integral achieved because of the unexpected early failure of the curium metal targets in the uranium beam. Even at target temperatures well below those that had been safe in electron gun bombardments the targets developed pin holes and cracks after approximately one day of beam time. The reason is not yet understood. Clearly, before any further attempts with the $^{238}\text{U} + ^{248}\text{Cm}$ reaction will be undertaken, the target technology has to be improved substantially.

OUTLOOK

In principle, the production of superheavy elements should be possible in damped collisions of heavy nuclei as follows from the identification of complementary fragments with quite large production cross sections. The success of such an approach hinges on several questions related to the state in which the heavy fragment finds itself after the separation of the

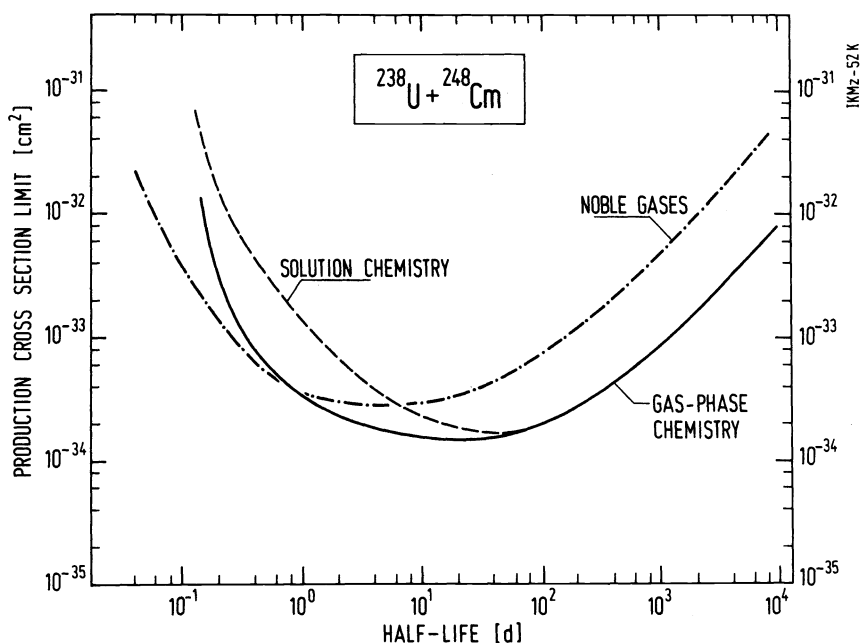


Fig. 13. Preliminary upper limits for the production cross section of spontaneously fissioning superheavy elements in the $^{238}\text{U} + ^{248}\text{Cm}$ reaction at 7.4 MeV/u incident energy.

dinuclear collision complex. Both the observation of a life-time exceeding 10^{-20} sec for $Z > 110$ fragments (Ref.14) and of surviving heavy actinides (Ref.4) are encouraging. However, at present, neither one of these experiments can exclude that the reaction proceeds through elongated shapes located already outside the fission saddle point and, hence, quickly leading to fission. Besides this difficulty there are the uncertainties of the theoretical fission-barrier heights and the question of the persistence of ground-state shell effects at high angular momenta. With the rapid growth of knowledge on the interactions between very heavy nuclei, it seems to become more and more evident that the production rates for superheavy elements in damped collisions of very heavy nuclei might be close to, if not below, the detection limit. Hence, a convincing proof for the formation of superheavy elements in such reactions will very likely be an extremely difficult undertaking. Considering, however, the fundamental problem behind this search - where is the upper end of the periodic table of the elements located and what forces terminate it - extensive efforts to shed light into this question seem to be justified.

Acknowledgement - The work summarized in this paper would not have been possible without the continuous and successful efforts of the UNILAC crew to provide high-intensity uranium beams, and without the cooperation of H. Gäggeler-Koch, D. Gembalies, B. Haefner, R. Heimann, H. J. Keim, U. Knitz, E. Schimpf, W. Schorstein, U. Tharun and W. Weber.

REFERENCES

1. See, e.g., G. Herrmann, Nature **280**, 543-549 (1979), and references therein.
2. G. T. Seaborg, W. Loveland and D. J. Morrissey, Science **203**, 711-717 (1979).
3. K. D. Hildenbrand, H. Freiesleben, F. Pühlhofer, W.F.W. Schneider, R. Bock, D. v. Harrach and H. J. Specht, Phys. Rev. Lett. **39**, 1065-1068 (1977).
4. M. Schädel, J. V. Kratz, H. Ahrens, W. Bröchle, G. Franz, H. Gäggeler, I. Warnecke, G. Wirth, G. Herrmann, N. Trautmann and M. Weis, Phys. Rev. Lett. **41**, 469-472 (1978).
5. See, e.g., W. U. Schröder and J. R. Huizenga, Ann. Rev. Nucl. Sci. **27**, 465-547 (1977), and references therein.
6. H. Freiesleben, K. D. Hildenbrand, F. Pühlhofer, W. F. W. Schneider, R. Bock, D. v. Harrach and H. J. Specht, Z. Physik A292, 171-189 (1979).
7. J. V. Kratz, W. Bröchle, G. Franz, M. Schädel, I. Warnecke, G. Wirth and M. Weis, Nucl. Phys. **A332**, 477-500 (1979).
8. J. V. Kratz, A. E. Norris and G. T. Seaborg, Phys. Rev. Lett. **33**, 502-505 (1974).
9. R. J. Otto, M.M. Fowler, D. Lee and G. T. Seaborg, Phys. Rev. Lett. **36**, 135-138 (1976).
10. C. Riedel and W. Nörenberg, Z. Physik A290, 385-391 (1979).
11. V. Volkov, Phys. Rep. **44**, 93-157 (1978), and references therein.
12. J. Poitou, R. Lucas, J. V. Kratz, W. Bröchle, H. Gäggeler, M. Schädel and G. Wirth, Phys. Lett. **88B**, 69-72 (1979).
13. T. Sikkeland, A. Ghiorso and M. J. Nurmi, Phys. Rev. **172**, 1232-1238 (1968).
14. P. Glässel, D. v. Harrach, Y. Civelekoglu, R. Männer, H. J. Specht, J. B. Wilhelmy, H. Freiesleben and K. D. Hildenbrand, Phys. Rev. Lett. **43**, 1483-1486 (1979).
15. L. G. Moretto, Nucl. Phys. **A180**, 337-362 (1972).
16. H. Folger and U. Richter, Nucl. Instr. Meth. **167**, 85-89 (1979).
17. B. Eichler, Kernenergie **19**, 307-311 (1976).
18. J. V. Kratz, J. O. Liljenzin and G. T. Seaborg, Inorg. Nucl. Chem. Lett. **10**, 951-957 (1974).
19. W. Bröchle, G. Tittel, N. Trautmann and M. Zendel, in Transplutonium Elements (eds. W. Müller and R. Lindner), North-Holland, Amsterdam, p. 29-36 (1976).
20. K. S. Pitzer, J. Chem. Phys. **63**, 1032-1033 (1975).
21. D. Kolb, Z. Physik A280, 143-147 (1977).
22. M. Schädel, W. Bröchle, B. Haefner, J. V. Kratz, W. Schorstein, N. Trautmann and G. Herrmann, Radiochim. Acta **25**, 111-117 (1978).
23. M. Schädel, Gesellschaft für Schwerionenforschung Darmstadt Report GSI 79-8 (1979).
24. V. L. Kuznetsov, L. E. Lazareva, V. G. Nedorezov, N. V. Nikitina and N. M. Parovic, Nucl. Phys. **A324**, 29-38 (1979).
25. H. Jungclas, D. Hirdes, R. Brandt, P. Lemmertz, E. Georg and H. Wollnik, Phys. Lett. **79B**, 58-60 (1978).
26. D. Hirdes, H. Jungclas, R. Brandt, P. Lemmertz, R. Faß and H. Wollnik, Gesellschaft für Schwerionenforschung Darmstadt Annual Report 1978 (GSI 79-11) p. 71.
27. D. C. Aumann, H. Faeschini, L. Friedmann and D. Weismann, Phys. Lett. **82B**, 361-364 (1979).
28. E. K. Hulet, R. W. Loughheed, J. M. Nitschke, R. L. Hahn, R. L. Ferguson, W. Bröchle, H. Gäggeler, J. V. Kratz, M. Schädel, G. Wirth, G. Herrmann, G. Tittel and N. Trautmann, these proceedings.