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The search for superheavy elements

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Keywords:

Superheavy elements, predicted properties, search for superheavy elements in nature, attempted syntheses by heavy-ion reactions.

Summary:

Extensive experimental efforts have been made during the past 12 years to detect superheavy elements in nature and to produce them in heavy-ion collisions. The search in nature remained inconclusive. Attempts to synthesize superheavy elements in the laboratory produced negative results. The concurrent study of heavy-ion reaction mechanisms has suggested new synthetic routes and modifications of old ones for future experiments.

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1. Introduction

For many years nuclear scientists believed that the periodic table of elements had been extended nearly to its limit, defined as the point where the Coulomb repulsion between the protons in the nucleus becomes so large that the cohesive nuclear forces cannot hold the nucleus together and the nucleus undergoes rapid spontaneous fission. This was based both on the liquid drop theory and on the observation of shorter and shorter spontaneous fission half-lives as the atomic number of the nucleus increased. However, in the period from 1966 to 1972 a number of detailed theoretical studies [1-5] showed that in the region of proton number $Z=114$ and neutron number $N=184$ the ground states of nuclei were again stabilized against spontaneous fission decay. The renewed stabilization was due to predicted shell closures at $Z=114$, $N=184$ and estimates of total half-lives involving all major modes of decay resulted in values for the most stable nuclei on the order of the age of the solar system. This was the reason for many nuclear chemists and physicists during the past 12 years to devote a considerable fraction of their scientific efforts and resources to attempts to detect these "superheavy elements" (SHE's), i.e. elements with atomic numbers $Z \geq 110$, either in natural samples or among the nuclear reaction products of heavy-ion collisions.

Today, the early optimistic view that a massive extension of the periodic table of the elements was possible through the production and identification of SHE's, has to be seen with scepticism. The present article will comment on current views of these expectations, summarize the results of attempts to synthesize SHE's at accelerators and briefly comment on recent searches for SHE's in nature. Reports on earlier work can be found in a number of review articles (e.g. Refs. 6 and 7) and conference proceedings [8,9].

2. Predicted properties of superheavy elements

2.1. Nuclear properties

In order to predict the stability of a superheavy nucleus one must rely on calculations of single-particle levels in nuclear potentials that have been extrapolated from the region of known nuclei. Most theoretical studies, e.g. those by Nilsson [10] and Nix [11], agree that the next major shell closures beyond $Z=82$ and $N=126$ occur at $Z=114$ and $N=184$, thus providing maximum stability against spontaneous fission for the doubly magic nucleus $^{298}_{114}\text{X}_{184}$. Besides spontaneous fission other decay modes, notably α -decay, beta decay, and electron capture, have to be considered for the calculation of the total half-life. In Fig. 1 two sets of predicted overall half-lives of superheavy nuclei are presented. In Fig. 1a, calculations by Fiset and Nix [12] are indicated. The maximum half-life of 10^9 y is predicted for $^{294}_{110}\text{X}_{184}$. The shift of the longest half-life from $Z=114$ to $Z=110$ is due to the competition of α -decay with spontaneous fission which decreases the half-lives for the higher atomic numbers. For the synthesis of SHE's by fusion reactions the shape of the shore of the "island of stability" in the west is extremely important because all possible landing places for this type of reaction are on the neutron-deficient side of the island. More recent calculations by Sobiczewski [13] reproduce this shape of the island but the half-lives are several orders of magnitude shorter with a maximum value of 10^3 y for $^{296}_{112}\text{X}_{184}$. A quite different shape of the island as predicted by Randrup et al [14] and Åberg et al. [15] is shown in Fig. 1b. Note the precipitous decrease in life-times (which is a consequence of largely reduced fission barrier heights) as the neutron number decreases from $N=184$ at a constant proton number. This trend of life-times and fission barrier heights indicates how important it may be to synthesize superheavy nuclei with the lowest excitation energy and the largest neutron number possible. With respect to the systematic uncertainty inherent in such calculations Bemis and Nix [16] have asserted that the accuracy of these half-life predictions is no better than $\sim 10^{\pm 10}$ for spontaneous fission decay and $\sim 10^{\pm 3}$ for α -decay. Effects like excitation

energy and angular momentum associated with the production of SHE's in nuclear reactions have also been considered. The fission barriers are predicted to vanish for 50 MeV of excitation energy and ~ 30 units of angular momentum [17,18].

Once formed, a SHE will probably give a unique signal in its decay which may provide the basis for specific detection techniques for such nuclei. Theoretical estimates indicate an average total kinetic energy for the fission fragments from a superheavy nucleus of 200-230 MeV and the evaporation of about ten neutrons from the fission fragments [19]. (For $Z=92$ these figures are 170 MeV and two neutrons, for comparison). It has also been speculated that the number of neutrons evaporated in the spontaneous fission of superheavy nuclei may only be of the order of 5 to 6, while the kinetic energy may be as high as about 270 MeV [20]. α -decay energies are considered to be a less unique fingerprint because these are expected [12] to fall in line with those of known α -particle emitters. Thus, the observation of spontaneous fission of high total fission-fragment energy and an unusually high neutron multiplicity should constitute the most distinct signal for the presence of SHE's.

2.2. Chemical properties

Predictions of chemical properties of SHE's are based on quantum mechanical relativistic Hartree-Fock or Hartree-Fock-Slater calculations of their ground state electronic structure [21], which defines their location in the periodic table of the elements, see Fig. 2. These calculations which can be carried out with fairly good accuracy indicate that the 6d shell is completed with element 112. The six 7p electrons are predicted to be split into two groups, two $7p_{1/2}$ electrons and four $7p_{3/2}$ electrons, with the splitting due to relativistic effects between their energies such that the filled $7p_{1/2}^2$ orbital (element 114, eka-lead) will act as a closed shell and additional $7p_{3/2}$ electrons will act like electrons outside a closed shell. Thus, element 115, eka-bismuth, is expected to have its valence electrons in the $7p_{1/2}^2 7p_{3/2}$

configuration with a stable +1 oxidation state in contrast to the stable +3 oxidation state of its homologue bismuth. Similarly, Pitzer [22] has pointed out that, due to strong relativistic effects, elements 112 (eka-mercury) and 114 (eka lead) may be very noble gases or liquids. The $8s^2$ orbital is filled with element 120. The group of super-actinide elements in which the inner 6f and 5g electron shells are filled starts with element 121.

The prediction of detailed chemical properties such as redox potentials in aqueous solutions requires, in addition to the calculation of electron configurations and ionization potentials for gaseous atoms [21], the prediction of properties such as the heat of hydration with its delicate dependence on the ionic radius. The prediction of such properties involves the judicious use of Mendeleev-like extrapolations of a smooth trend in the variation of the property among the members of a particular group in the periodic table (Fig. 2). This is very interesting field of chemical research has been reviewed by Fricke [21] and Keller and Seaborg [23].

Considerations on the chemical behaviour of SHE's are of importance for the selection of terrestrial and extraterrestrial samples and for the development of chemical methods for the isolation of SHE's from such natural samples or from irradiated targets [23]. Separations that have been applied in attempts to chemically identify SHE's are based on the high volatility and ease of reduction [24,21-23], on the predicted extreme stability of anionic bromide complexes [25] and the resultant ion exchange behaviour [26], and on the predicted strong affinity to sulfur [27,28] or sulfur compounds [29].

3. Search for superheavy elements in nature

One condition for the occurrence of SHE's in terrestrial samples is a half-life comparable to the age of the earth, about 10^9 y, the other one is the formation of these heavy elements in nucleogenesis. Heavy elements such as uranium are produced by the so-called r-process in supernovae by a sequence of neutron captures followed by β^- -decay. Whether or not the r-process can lead through a region of neutron-rich nuclei having low fission barriers to the region of SHE's is a question which has been answered controversially. In the most recent studies the production of SHE's in the r-process is considered unlikely [30-32], the results depend, however, on a delicate balance between extrapolated fission barriers and neutron binding energies.

Despite these uncertainties hundreds of natural samples were examined for superheavy elements. The most sensitive method used in these searches was the detection of spontaneous fission events [33]. For untreated samples detection limits of 10^{-12} to 10^{-15} grams of SHE's per gram sample were achieved if the spontaneous fission half-life was assumed to be 10^9 years. These limits have been improved in some cases by chemical enrichment and mass separation. Most of these efforts gave no conclusive evidence for the presence of SHE's but there have been a number of observations that do not definitely rule out the existence of SHE's. One of these spontaneous fission activities was detected in the hot spring waters on the Cheleken peninsula (USSR) by Flerov et al. [34]. This water is enriched with heavy volatile elements and is assumed to originate from a large depth. About 2000 m^3 of this water were passed through a barrel containing 850 kg of anion exchange resin. Some 170 kg of the resin were eluted with water and different acids, and hydroxides were precipitated in the effluent. The resulting samples were measured by high-efficiency neutron multiplicity counters. The spontaneous fission count rate was 0.5 per day per 9 kg of saturated resin, a rate which is much higher than background. The hydroxide precipitate contained an activity of 5 counts per day and had thus enriched about 50 % of the original activity of the resin. The neutron multiplicity distribution

obtained with the saturated resin and with the hydroxides is shown in Fig. 3 together with known multiplicity distributions occurring in the spontaneous fission decay of ^{238}U , ^{246}Cm , and ^{252}Cf , as well as distributions for some hypothetical emitters. The experimental neutron multiplicity distribution is compatible with that of ^{252}Cf , whereas ^{246}Cm and ^{238}U can be ruled out. In order to check for a possible contamination by fallout from nuclear weapons tests or by actinides present in the laboratory the authors isolated an Am through Cf fraction from a representative portion of the hydroxides. From the measured ratio of α -particle-to-spontaneous fission activity in this Am-Cf fraction the authors concluded that the activity cannot be due to a contamination by actinides with the exception of pure ^{252}Cf . The latter possibility was considered unlikely and it was suggested that the spontaneously fissioning nuclide detected in the Cheleken waters may be a SHE. Attempts to further concentrate this activity by chemical procedures which are more specific for the expected chemical behaviour of SHE's failed because of increased losses of the activity at each stage of processing [35]. No evidence for spontaneous fission decay has been found in solid deposits from hot springs at the bottom of the Red Sea which are assumed to be of a similar geological origin as the Cheleken samples [36,37], even though their chemical composition is different.

The Dubna group [38-40] also examined several meteorites for spontaneous fission activity, among them the meteorite Allende, a carbonaceous chondrite, in which Anders et al. [41-44] had previously found evidence for anomalous isotopic ratios for xenon isotopes which were interpreted as the result of spontaneous fission decay of a now-extinct, volatile SHE. The latter hypothesis was met with criticism, see e.g. Ref. 45. The meteorites examined with neutron multiplicity counters at Dubna showed, on the average, one event per five days in 10 kg of sample, which is 10-30 times higher than the counting rate expected from the uranium content of the meteorites and from other sources of background. Chemical separations based on the volatility of SHE's in the elementary or in an oxidized state were also applied by Zvara et al. [46] and it was suggested that the meteoritic samples contained a long-lived spontan-

eously fissioning nuclide of a SHE. However, in attempts to further concentrate the activity chemically the yield dropped down to a level close to the detection sensitivity [47].

Another possible indication for the natural occurrence of SHE's arose from studies of the heavy component of galactic cosmic rays by Dubna scientists [48]. Meteorites are exposed to the flux of cosmic rays during an average of 1×10^7 years; as a result of this exposure they accumulate an enormous amount of information on the charge spectrum of cosmic rays. By etching suitable minerals such as olivines located not too far from the surface of the meteorite one can obtain visible tracks the length of which is directly related to the atomic number of the nuclei producing the tracks. About 1 cm^3 of olivine crystals were microscopically scanned and about 2500 tracks produced by heavy nuclei with $Z > 70$ were registered. The distribution of track lengths after annealing shows a peak at $180\text{--}230 \text{ }\mu\text{m}$ which belongs to the uranium-thorium group. In addition, two tracks of 320 and $370 \text{ }\mu\text{m}$ length were found indicating nuclei with $Z \sim 110$. The annealing procedure reduces considerably the tremendous background associated with the abundant iron group in the galactic cosmic rays. Calibrations of track lengths were achieved by heavy-ion bombardments using Ti through Xe projectiles. Most recently, more olivine crystals were scanned and two more tracks of about $300 \text{ }\mu\text{m}$ length were found [38]. An important test of the method will be carried out in the near future when high-energy ^{238}U beams will be available from the Berkeley Bevalac accelerator.

4. Attempts to synthesize superheavy elements in heavy-ion collisions

During the period following the initial optimistic predictions concerning the production cross sections [49] for SHE's efforts began at the heavy-ion accelerators at Berkeley, Orsay and Dubna to "jump the gap" between the region of known nuclei and the predicted magic island by fusing two heavy nuclei together. Somewhat later, at Darmstadt, through the avail-

ability of projectiles as heavy as ^{238}U new prospects for the synthesis of SHE's showed up with the observation of a massive transfer of nucleons from one heavy nucleus to another at relatively low excitation energies. Both approaches are schematically illustrated in Fig. 4 which also gives some characteristic times (in units of 10^{-21}s). In a complete fusion reaction (see the upper branch) projectile and target amalgamate to a nearly spherical compound nucleus. The compound nucleus, even if formed with the minimum kinetic energy possible (i.e. at the barrier) carries several tens of MeV of excitation energy because, in general, more kinetic energy is required to overcome the Coulomb barrier than energy is consumed in the subsequent (endothermic) fusion process. The excitation energy can be carried away by the subsequent evaporation of light particles, mostly neutrons, which is followed by the emission of γ -rays before the ground state of the final evaporation residues is reached. For heavy nuclei the number of evaporation residues is a minute fraction of the number of compound nuclei that are primarily produced because at each step of the de-excitation chain fission into two fragments of comparable size competes heavily with particle evaporation. Thus the probability of producing a detectable superheavy nucleus is the product of two factors: i) the probability to fuse the reacting heavy ions to form an equilibrated superheavy compound nucleus, and ii) the cumulative probability of the excited superheavy compound nucleus to survive its de-excitation process.

The lower branch in Fig. 4 illustrates the alternative process termed deep-inelastic collision, in which the interacting nuclei stick together for a short time forming an excited, rotating double-nuclear system which then separates again into two heavy nuclei. During the life-time of the double-nuclear system kinetic energy of the relative motion is partially or completely transformed through friction into internal excitation energy, orbital angular momentum is dissipated into intrinsic spin of the fragments, and a substantial number of protons and neutrons can be transferred between the interacting nuclei. Ultimately, the separated fragments de-excite by particle emission and the competing fission as discussed above. In contrast to complete fusion where the

fused product is characterized by discrete mass and charge numbers, by a discrete excitation energy and a narrow bin of angular momenta, deep-inelastic collisions produce broad distributions of product mass and charge numbers, and broad distributions of excitation energies and angular momenta. In the following sections we shall summarize the results of attempts to synthesize SHE's using these alternative approaches and comment on the reasons why these attempts have failed. This is followed by a brief discussion of new insights into heavy-ion reaction mechanisms which might serve as guidance for future research.

4.1. Complete fusion reactions

Since complete fusion was so successful in the synthesis of the heaviest known elements it was natural to use this type of reaction in the first attempt to synthesize SHE's by bombarding $^{248}_{96}\text{Cm}$ with $^{40}_{18}\text{Ar}$ -ions to form $^{288}_{114}\text{X}$ compound nuclei [31,52]. The dilemma of this reaction and of many similar other ones lies in the fact that the possible compound nuclei have neutron numbers far below $N=184$ where fission barriers are predicted to be vanishingly small [4] or zero so that the cumulative survival probability is zero. Table 1 lists the fusion reactions tried so far together with the upper limit for the production cross section and the ranges of half-lives that were covered by the experiments. As suggested by Seaborg et al. [53] the reactions are ordered in three classes. Class 1 comprises those reactions where compound nuclei in the close vicinity of element 114 were produced. Unfortunately, this is only possible in projectile-target combinations with much less than the optimum number of neutrons resulting in very low survival probabilities precluding production and observation of SHE's. The magic neutron-number $N=184$ can be reached if compound nuclei with proton numbers as high as $Z=122$ are produced. These so-called "overshoot"-reactions (class 2 in Table 1) are expected to produce compound nuclei for which very good overall survival rates are predicted [53]. After de-excitation, decays by α - and beta-decay toward the center of the island of stability are predicted [12,14,15]. This is illustrated in Fig. 5 for the $^{136}\text{Xe} + ^{170}\text{Er}$ _{54 68}

reaction used by Münzenberg et al. [61] in an attempt to produce evaporation residues of the magic compound nucleus $^{122}_{306}\text{X}$.

This reaction was selected because the energetics is such that the compound nucleus is formed with very little excitation energy, so that only one neutron was expected to be evaporated in its de-excitation. The experiment was performed with the velocity separator SHIP at the UNILAC accelerator at Darmstadt. SHIP separated the fast projectiles ($10^{11}/\text{s}$) and transfer products by electrical and magnetic fields from eventual fusion products which would have been ejected from the target with much lower velocity. The slow fusion products would have been focussed onto a position-sensitive surface barrier detector which would i) register the implantation of a heavy nucleus and measure its velocity, ii) register the position of the implanted product inside the detector, and iii) register any α -particles or fission fragments that are emitted from this position inside the detector as a result of subsequent decay of the implanted evaporation residue. These decay events would be further characterized by the measurement of the associated energies and the correlation time elapsed between the implantation and the decay event. A significant signal for the production and subsequent decay of a SHE would then consist of an implantation signal indicating the proper product velocity followed by a correlated sequence of rapid α -decays ending by a spontaneous fission decay. Through combination of the velocity separation from the primary beam particles and transfer products with the requirement of such a unique sequence of signals a signal-to-noise ratio of $10^{18}:1$ was achieved. No single event was observed, defining an upper limit for the production cross section of about 1 nanobarn. The same technique was successfully applied in the recent discovery of element 107 in the $^{249}_{83}\text{Bi} + ^{54}_{209}\text{Cr}$ reaction by Münzenberg et al. [62].

Despite extensive searches using class 2 reactions over a wide range of bombarding energies, projectile-target combinations, and product half-lives, there were no successful SHE syntheses and rather low upper limits were set on SHE production. There is now, see subsection

4.4.2., solid evidence for the fact that the probability for fusion at the coulomb barrier has vanished for the class 2 reactions in Table 1. Thus, no SHE's appear to be formed for dynamical reasons by these overshoot reactions even though the survivability of the respective compound nuclei would have been rather promising.

Among the reactions listed in Table 1 the class 3 reaction of $^{48}_{20}\text{Ca}$ projectiles with a $^{248}_{96}\text{Cm}$ target has been studied most extensively both at Berkeley and at Dubna. This reaction represents the intriguing case in which both the fusion probability and the survivability have been considered favourable. Projectile and target nucleus were brought together $[55, 28, 66-68]$ with an energy exceeding the barrier by about 20 MeV. The extra energy was thought to be necessary to induce complete fusion to produce a compound nucleus with some 40 MeV of excitation energy which is expected to de-excite by the evaporation of 4 neutrons. The resulting evaporation residue $^{292}_{116}\text{X}$, according to Fiset and Nix [12], decays in the following major decay chain

with the final nucleus $^{288}_{112}\text{X}$ having a calculated spontaneous fission half-life of about 1 h. In the more recent studies by Randrup, Åberg and collaborators [14, 15] the life-times and decay modes are much less favourable, i.e. $^{292}_{116}\text{X}$ decays with a half-life of $1.6 \times 10^{-15}\text{ s}$ by spontaneous fission, thus precluding its observation in any of the experiments performed.

The $^{48}_{20}\text{Ca} + ^{248}_{96}\text{Cm}$ experiments were quite difficult and expensive because not only the target material (a highly radioactive isotope) but also the projectile $^{48}_{20}\text{Ca}$ with only 0.19% natural abundance are available (only in the USA and the USSR) in very limited amounts. The negative results produced in the course of many carefully planned and executed experiments both at Berkeley and at Dubna are summarized in Fig. 6 where upper limits for the production cross sections are plotted as a function of the half-lives covered by a given experimental technique.

The Dubna group [55] used two different chemical separation schemes followed by spontaneous fission (SF) and α -particle counting (α), denoted by D in Fig. 6. At Berkeley (BL in Fig. 6) a variety of techniques was applied: a) chemical separations [66,28] with two alternative procedures followed by α -particle and spontaneous fission counting (CHEM); b) examination [67] of condensed noble gases and extremely volatile species for α -particle decay and fission (GAS); c) direct fission fragment counting of stopped recoils [28] (FOLLS); d) gas-jet transport to a rotating wheel counting system [68] (WV), and e) spontaneous fission decay in flight [68], labeled DIF in Fig. 6.

Seaborg et al. [53] have analyzed reasons why SHE's were not seen in the $^{48}\text{Ca}+^{248}\text{Cm}$ reaction by calculating the relative neutron decay widths Γ_n/Γ_f at each step of the 4n-evaporation chain using the formalism outlined by Vandenbosch et al. [69] and two different sets [12,14] of fission barrier heights. The estimates of the latter quantities are either typical for most theoretical calculations done in the period from 1966 to 1972 [12], or represent a more pessimistic recent approach [14]. The resulting fraction of the compound nuclei surviving fission are then 96% or $<4 \times 10^{-9}\%$, respectively [53]. With an estimated fusion cross section of $\geq 10^{-27} \text{ cm}^2$ and the experimental upper limit on the production cross section of SHE's one can estimate an "experimental" upper limit for the survival rate in this reaction as $5 \times 10^{-35}/10^{27} = 5 \times 10^{-8}$. Clearly, this figure is dramatically smaller than the 96% resulting from the barriers of Fiset and Nix [12]. According to Seaborg et al. [53] the "experimental" survival rate requires the assumption of fission barrier heights that are 4 to 5 MeV lower than those originally predicted which are, thus, more in line with the barrier heights of Randrup et al. [14]. This conclusion is not unique, because reduced barrier heights are associated with largely reduced half-lives (recall the $1.6 \times 10^{-15} \text{ s}$ for ^{116}Xe predicted by Åberg et al. [15]) where the experimental sensitivity was much less than optimum or where no experimental technique was available at all, see Fig. 6. Also, the calculations [53] were performed with barrier heights corresponding to the full shell strength of a spherical SHE thus ignoring the reduction in barrier

heights by excitation energy and the extremely delicate dependence of the stability of a SHE on deformation. We shall come back to a discussion of these problems in section 4.4.3.

4.2. Deep inelastic transfer reactions

The failure of the complete fusion approach to produce SHE's may be associated in some direct or indirect way with the failure of even the best projectile-target combination to produce compound nuclei in the center of the island of stability where fission barriers are the highest and the losses of yield to prompt fission are the least. Therefore, it was natural to look for alternative approaches allowing production of more neutron-rich superheavy nuclei.

In 1977 studies of the interactions of two colliding uranium nuclei at the Darmstadt UNILAC accelerator [70,71,72] showed that the distributions in mass- and charge numbers of excited primary transfer products from deep-inelastic collisions extend into the region of SHE's. The evidence is based on the observation of the complementary light products. If the doubly magic $^{298}\text{X}_{114}^{184}$ would be formed through transfer of nucleons from one ^{238}U nucleus to another one, the complementary light product would be an ytterbium isotope:

Fig. 7 shows the element distribution in the reaction of 7.5 MeV/u ^{238}U -ions with a thick ^{238}U target obtained in radiochemical studies by Schädel et al. [72]. The distribution is interpreted as a superimposition of four components: (i) products in the close vicinity of ^{238}U from quasi-elastic few-nucleon-transfer reactions (890 mb), (ii) products with atomic numbers from Z=73 through 100 arising from an originally symmetric distribution around Z=92 produced by deep-inelastic transfer processes (290 mb), (iii) a nearly symmetric fission product distribution originating from highly excited deep-inelastic fragments (1040 mb), and (iv) a double-humped distribution

of neutron-rich fission products from the sequential fission of quasi-elastic transfer products (860 mb). Because the total cross section for deep-inelastic collisions has been measured (the sum of components (ii) and (iii)) the original pre-fission distribution can be reconstructed [72] as indicated by the triangles in Fig. 7. The resulting distribution is well reproduced by a semi-phenomenological theory by Riedel and Nörenberg in which the nucleon transfer is treated as a diffusion process [73]. The latter calculation predicts that the complementary elements Z=70 and Z=114 are formed with a cross section of 10^{-28} cm^2 . If one applies the rules [74] that describe the distribution of neutrons and protons between both complementary fragments on the basis of the phase space available above the potential energy as a function of N and Z, one predicts a relatively broad distribution in neutron numbers for Z=114 close to N=184. Thus, deep-inelastic transfer reactions lead to a much closer access to the center of the island of stability than any fusion reaction, as is demonstrated by Fig. 8. The important question to be asked is whether there is a chance for these heavy fragments around Z=114 to survive their de-excitation.

Counter experiments [70,71] revealed that more nucleons are transferred per MeV of excitation energy in the $^{238}\text{U} + ^{238}\text{U}$ reaction than in other deep-inelastic collisions involving heavy targets and lighter projectiles, indicating the production of "colder" products in the $^{238}\text{U} + ^{238}\text{U}$ system. In radiochemical studies the production cross sections of surviving transuranium nuclides were followed over eight orders of magnitude up to fermium [72], and it was found that the production rates for the heaviest actinides in the $^{238}\text{U} + ^{238}\text{U}$ reaction exceeded considerably those in the $^{136}\text{Xe} + ^{238}\text{U}$, $^{84}\text{Kr} + ^{238}\text{U}$, or $^{40}\text{Ar} + ^{238}\text{U}$ reactions. This result was not necessarily expected: In a deep-inelastic collision the amount of excitation energy showing up in the final fragments depends i) on the amount of kinetic energy that is dissipated into internal excitation energy during the collision, and ii) on the difference in the binding energies of the projectile and target and the emerging fragments in the exit channel. The latter is called the ground state Q-value, Q_{gg} , and

resumes negative values for endothermic transfer reactions. It appears now, that formation of ^{98}Cf in the bombardment of ^{238}U with the lighter projectiles ^{40}Ar , ^{84}Kr or ^{136}Xe is associated with strongly negative values of Q_{gg} of -43 MeV, -40 MeV, and -33 MeV, respectively. In the $^{238}\text{U} + ^{238}\text{U}$ reaction the same product is formed with $Q_{\text{gg}} = -2 \text{ MeV}$. Since a negative value of Q_{gg} reduces the excitation energy showing up in the final fragments and thus increases their survivability, one might expect, contrary to the experimental observation, increased actinide yields with the lighter projectiles if the primary yield before fission at the same amount of dissipated kinetic energy is the same in all reactions. In turn, the experimental observation of increased actinide yields in $^{238}\text{U} + ^{238}\text{U}$ collisions is evidence for a much higher primary yield at a given dissipated energy than in the other reactions.

Additional studies and analyses [75,76], see also subsect 4.4.4., demonstrated that the survival rates of these actinides are compatible with neutron evaporation-fission competition of fully equilibrated compound nuclei, i.e. can be calculated in the same way as for compound nuclei produced in fusion reactions. If we assume that this is also true for formation of Z=114 in deep-inelastic collisions we can fold the predicted production cross section for Z=114 with less than 30 MeV of excitation energy [73] with an energy-dependent survival rate [17] and obtain a cross section of about 10^{-35} cm^2 for surviving residues, a value which corresponds, with the accessible beam intensities, to production rates of a few atoms per week of beam-time, a rate just exceeding the detection limit of the most sensitive methods available at present.

In view of these not completely hopeless estimates direct searches for surviving SHE's in the $^{238}\text{U} + ^{238}\text{U}$ reaction were performed by a Darmstadt-Mainz collaboration. In these experiments thick ^{238}U metal targets were bombarded with 8.6 MeV/u ^{238}U beams. The thickness of the targets assured that the whole (unknown) excitation function for SHE production was physically integrated inside the target. Integral particle numbers of up to 5×10^{16} were accumulated within typical beam times of five days

[77]. After bombardment the targets were processed with two chemical separation procedures [78] based on different predictions of the chemical behaviour of SHE's. In the gas phase chemistry the predicted volatility and ease of reduction of elements 112 through 117 [24] was utilized. The solution chemistry was based on the predicted ion exchange behaviour of bromide complexes [26] and on the predicted affinity to sulfur [29] for elements 108 through 116. In both procedures provisions were made to condense noble gases and other species that are volatile at room temperature. In one experiment the solution chemistry was extended in order to search also for members of the superactinide series. Experimental details can be found in Ref. 78. Superheavy element fractions were assayed for α -particle and spontaneous fission events between two large-area surface barrier detectors with an efficiency of 60 % for recording both fission fragments in coincidence. More recently these counter pairs were positioned inside a neutron multiplicity counter so that neutrons could be registered in coincidence with fission fragments. Upper limits for the production cross sections of SHE's vs. the assumed half-life are summarized in Fig. 9. The limits obtained with chemical techniques [77] are labeled CHEM. The variation of these limits with half-life is the result of decay during the chemical separations for short-lived species, and incomplete saturation and decay for lifetimes comparable to or longer than the total counting time (usually about 200 d). The limits denoted WHEEL are the result of fission track detection with a rotating wheel system [77] where the sensitivity for spontaneously fissioning SHE's is limited by a background produced by the spontaneously fissioning actinide nuclei ^{244}Fm , ^{242}Fm , ^{256}Fm , ^{254}Cf , and ^{252}Cf in the unseparated reaction product mixture collected in the rotating catcher foils. Also included in Fig. 9 are upper limits from gas-jet experiments labeled JET [59,79] and from the implantation of recoil atoms [70] in a surface barrier detector (REC).

All experiments have produced negative results. As in the case of the $^{48}\text{Ca}+^{248}\text{Cm}$ reaction one notes a lack of sensitive measurements for very short-lived nuclei which would have to involve an efficient separation from actinide nuclei. Because the predicted cross sections for surviving

SHE's in the $^{238}\text{U}+^{238}\text{U}$ reaction were so close to the detection limits it was natural to think about ways to increase the production rates. Since no gain in the production rates for heavy actinides were observed in bombardments at higher projectile energies [75] it was not considered reasonable to use higher energies. More promising was the use of a heavier target material. Calculations with the diffusion model predict an increase in the production cross section for Z=114 at 30 MeV of excitation energy by two orders of magnitude if a thick ^{248}Cm target is used instead of a ^{238}U target [73]. After extensive development efforts at Berkeley, Darmstadt, Livermore and Mainz the first bombardments of thick ^{248}Cm targets with ^{238}U beams were performed at the UNILAC accelerator in October 1979. ^{248}Cm targets of 3.5 to 7 mg/cm² areal density were prepared by evaporation of ^{248}Cm metal onto Mo-foils of 4.5 mg/cm² thickness. These targets contain a spontaneous fission activity of about 10⁹ events per day, whereas a rate of a few fission events per year occurring in one of the SHE fractions would be considered a significant signal for SHE production. This shows that extreme care had to be taken in order to prevent an uncontrolled destruction of the target and subsequent contamination of the beam line, the accelerator and the laboratory. Details on the target cooling, temperature monitoring, beam wobbling, on the use of an interlock system, fast-acting valves, and glove boxes are given in Ref. 75. The target thicknesses were sufficient to degrade the beam energy from 7.4 MeV/u to or below the interaction barrier. In the first experiment, in which a beam integral of 3x10¹⁵ particles was accumulated, the heavy reaction products were stopped in a water-cooled stack of copper catchers. After the end of bombardment these catchers were heated to 1020 °C in the presence of hydrogen in order to separate volatile elements. The catchers were then dissolved in order to perform also the solution chemistry. The resulting chemical fractions mounted on thin foils were counted between opposed surface barrier detectors inside a neutron multiplicity counter. Elements that are volatile at room temperature like Rn were condensed on a cryostat faced by a single surface barrier detector. Preliminary upper limits resulting from these experiments were of the order of 2-3x10⁻³⁴ cm² for half-lives between several hours and about

100 d [78]. Simultaneously, various actinide fractions up to nobelium were separated in order to measure actinide production cross section, see section 4.4.4. A second experiment to detect gaseous SHE's ended after 4 hours of beam time (2×10^{14} particles) when the ^{248}Cm target developed a hole. Here, Krypton gas was used to stop the reaction products and to sweep gaseous products through a filter system (removal of non-volatile actinides) to a copper cryostat cooled to -150°C where they were continuously counted for fission events and high-energy α -particles. No such events were observed resulting in an upper limit of 10^{-33} cm^2 for volatile SHE's with half-lives between two minutes and several hours [80]. Both experiments were hampered by the unexpected early failure of the ^{248}Cm metal targets in the ^{238}U beam thus reducing the beam integrals to far below the integrals reached in the ^{238}U , ^{238}U experiments. Inspections of the ^{248}Cm targets, of irradiated targets of its homologue gadolinium, extensive bombardments of similar targets with pulsed electron beams, low-energy heavy-ion beams and additional 7.5 MeV/u ^{238}U bombardments led to the conclusion that there was a problem in transferring heat from the target through the Cm/Mo interface to the gas-cooled Mo substrate during heavy-ion bombardment causing rapid temperature cycling and tensile stresses that ultimately led to a mechanical failure of the targets along the rolling lines of the Mo substrate. An acceptable, partial solution to these heat transfer problems was achieved by modifications of the target apparatus which allows now to cool the target on both sides by 1.5 l/s nitrogen gas. In Juli 1981, with this improvement, about 2×10^{16} ^{238}U particles could be accumulated on target. The reaction products were again stopped in a stack of copper catchers and chemically processed as before. Counting of the isolated fractions is still being continued. Upper limits [81] for the production cross sections of SHE's after 180 d of counting are presented in Fig. 10. Even though there is the prediction of a hundred-fold increase in yield of SHE's in the ^{238}U , ^{248}Cm reaction relative to the ^{238}U , ^{238}U reaction [73], no detectable amounts of SHE's have been found in the bombardments of ^{248}Cm with ^{238}U ions in the half-life region of a few hours to some years with a sensitivity close to the edge of current technological limits.

4.3. The "fusion-after-instantaneous-fission" approach

A third series of experiments is based on the theoretical suggestion of instantaneous fission of a heavy nucleus during the close contact with another heavy nucleus followed by fusion of one of the fission fragments with the nucleus causing the fast fission process [82]. This suggestion has been shown [83] to produce negative results in the reaction of ^{208}Pb with ^{136}Xe where an upper cross section limit of $1.2 \times 10^{-30} \text{ cm}^2$ was obtained for half-lives between 10^{-12} and 10^4 s , and in the reaction of ^{208}Pb with ^{238}U using a rotating wheel system (Wheel), the gas-jet method (Gas-Jet) and off-line chemistry (Chemistry) [84], see Fig. 11. The continuation of experiments along this line appears to be the least promising because i) there is no evidence for production of known nuclei through this mechanism [85], ii) "fast fission" does not seem to occur at near barrier energies [86], and iii) fusion of an uranium fission product with a ^{208}Pb nucleus is dynamically hindered at near-barrier energies, see section 4.4.2.

4.4. New results on heavy-ion reaction mechanisms

Many of the above mentioned investigations, while failing to synthesize SHE's, have provided as a by-product valuable data on heavy-ion reaction mechanisms. Also, there is a growing body of relevant data emerging from concurrent studies of reaction mechanisms concerned with the limitations of complete fusion by the barrier and by entrance channel effects, with the production of cold heavy nuclei in transfer reactions, and with the survivability of excited heavy nuclei. These new results seem to provide highly significant guidance for future attempts to synthesize SHE.

4.4.1. Cold fusion through barrier penetration

Fusion above the classical barrier is reasonably well described by conventional one-dimensional fusion models using conservative nucleus-nucleus potentials [87]. However, from recent experiments [88-91] it follows that fusion well below the barrier (involving the quantum-mechanical penetration of the fusion barrier) cannot be described with such one-dimensional models of colliding rigid spheres involving WKB theory or the Hill Wheeler expression for the transmission through a potential barrier. As a general rule one observes unexpectedly large cross sections below the barrier. More specifically, there is evidence that the cross section enhancement is dependent on the nature of the amalgamating nuclei. In a comparative study [88,89] it was shown, first, that the cross section enhancement was larger for deformed target nuclei (^{154}Sm) than for spherical nuclei, and, second, that the global effect was much stronger if a heavier projectile (^{40}Ar) was used instead of a light one (^{16}O). As shown in a particularly clear way by Reisdorf et al. [91] in the fusion of ^{40}Ar with $^{112,116,122}\text{Sn}$ there is a striking gain in cross sections relative to the fusion cross sections calculated for rigid spheres even for spherical target nuclei, see Fig. 12. For statically deformed and "soft" nuclei the gain is larger. For even heavier projectiles than ^{40}Ar this trend is further accentuated [92]. Similar trends are likely to hold for superheavy target-projectile combinations. Thus, Fig. 12 tells us that lowering of the centre-of-mass bombarding energy below the barrier by 10 MeV reduces the fusion cross section by a factor of ~ 13 or less, whereas conventional fusion models predict a decrease by about 5 orders of magnitude. Since lowering of the entrance channel kinetic energy decreases the compound nucleus excitation energy by the same amount of MeV and since the survivability of a heavy, highly fissile compound nucleus depends so dramatically on its excitation energy the net result of choosing a bombarding energy ~ 10 MeV below the one-dimensional barrier will be a gain in the cross section for surviving superheavy evaporation residues by several orders of magnitude. Quantitative estimates of these gain factors are difficult to obtain because the survivability of a SHE (section 4.4.3.) depends also on the rate of

weakening of the shell effects near $Z=114$ and $N=184$ by excitation energy and compound nucleus deformation, both factors being poorly known at present.

The cross section enhancements for fusion below the barrier is far from being theoretically understood. Qualitatively, as the size of the projectile becomes comparable to that of the target, dimensions other than the separation of the centers of two rigid nuclei are expected to become important. Among the additional degrees of freedom that have to be considered we mention deformations, both static or momentary as a result of the zero-point motion of low-lying surface vibrations and the rapid formation of a neck between target and projectile which make the barrier penetration problem multidimensional.

4.4.2. Dynamical limitations of fusion near the barrier

The above statement on the time scale for the necking degree of freedom comes from low-energy fission studies [93] where fully paired even-even fragmentations into the ground states of the fission fragments have not been detected. The finding of pair breaking even for the coldest fragmentations allows to estimate a time for the necking-in process of at most 3×10^{-22} s. This time is short compared to the equilibration time for the mass asymmetry degree of freedom which is known from deep-inelastic heavy-ion collisions to be about 4×10^{-21} s. The time for formation of a neck in a fusion reaction, i.e. the transition time from a binary system to an elongated saddle point shape (the barrier crossing time) is, therefore, believed to be also much smaller than the time required for the disappearance of the mass asymmetry of the entrance channel. Therefore, it makes sense to study the dynamics of fusion at frozen mass asymmetry as a function of the separation of the colliding nuclei and as a function of the neck-size as proposed by Swiatecki [94]. Then, in analogy to fission where the fissility of the system is governed by the ratio between repulsive Coulomb forces and attractive nuclear forces, the fusability of two touching nuclei will be governed by an

effective value of Z^2/A defined as the ratio of forces in the entrance channel as

$$(Z^2/A)_{\text{eff}} = \frac{4 Z_1 Z_2}{A_1^{1/3} \cdot A_2^{1/3} (A_1^{1/3} + A_2^{1/3})},$$

if centrifugal forces are ignored. The latter is justified for the formation of very heavy fusion-evaporation-residues where only the lowest partial waves contribute. $(Z^2/A)_{\text{eff}}$ is a measure for the depth of the potential pocket which is required for fusion. Using a proximity potential and a Coulomb potential one can calculate that the pocket disappears for $(Z^2/A)_{\text{eff}} \sim 46$. Unfortunately, however, there is a geometrical limitation of fission in the entrance channel which is given by the elongation of the "conditional" saddle point shape with respect to the elongation associated with the touching point of projectile and target, as schematically illustrated in Fig. 13. In the upper part of the Figure the touching point lies within the separation R appropriate for the conditional saddle point, thus allowing the system to fall into the pocket and to fuse. For a system with $(Z^2/A)_{\text{eff}}$ being larger than a threshold value $(Z^2/A)_{\text{thr}}$ the system is not trapped unless an extra amount of kinetic energy is provided which brings the system inside the conditional saddle point. This "extra push", if supplied, results in additional excitation of the compound nucleus which, in turn, reduces its survivability. The extra push for systems with $(Z^2/A)_{\text{eff}} > (Z^2/A)_{\text{thr}}$ increases quadratically as

$$\Delta E_{\text{push}} = a^2 \eta_0 \left[(Z^2/A)_{\text{eff}} - (Z^2/A)_{\text{thr}} \right]^2$$

where the parameter a is the rate at which the extra push increases and η_0 is a dimensional constant. An important task for the experimenters was now to find out the value of $(Z^2/A)_{\text{thr}}$ and to check whether this value is independent of the fissility of the compound system. Sann et al. [95] and Swiatecki [94] asserted on the basis of measurements of the cross sections for symmetric fission in a large number of target-projectile combinations [95] that the threshold is located at $(Z^2/A)_{\text{thr}} \sim 33 \pm 1$

and the slope is ~ 12 . One may argue that the observation of symmetric fragmentation does not prove that a compound nucleus was existing. Therefore, measurements of compound nucleus evaporation residue cross sections are a more stringent test of the location of the dynamical threshold for fusion. Fig. 14 shows the experimentally determined extra push ΔE_{push} relative to the one-dimensional barrier for touching spheres as a function of $(Z^2/A)_{\text{eff}}$ [96]. Several important conclusions can be drawn from the data shown in Fig. 14: i) The concept of a conditional saddle point allowing scaling of the fusability of different target projectile combinations with the entrance channel parameter $(Z^2/A)_{\text{eff}}$ has been confirmed for systems with very different fissility (Z^2/A_0) . This confirms that the barrier crossing time (the time for formation of a neck) is short compared to the equilibration time for the mass asymmetry. ii) Up to $(Z^2/A)_{\text{eff}} \sim 38$ fusion has been observed unhindered with compound nucleus excitation energies as low as 15-20 MeV. iii) Below $(Z^2/A)_{\text{eff}} \sim 38$ fusion two nuclei which are very gently brought together with zero relative velocity and negligible angular momentum seems to be possible without the release of intrinsic energy due to friction, whereas one-body dissipation [94] already predicts "extra push" energies of up to 20 MeV. For $(Z^2/A)_{\text{eff}} < 38$ at the lowest possible bombarding energies the neck formation or barrier crossing is a friction-less transition. iv) Beyond $(Z^2/A)_{\text{eff}} \sim 38$ there seems to be a "thud wall" preventing heavy target-projectile combinations from being fused at low excitation energies.

The messages concerning the production of SHE's in fusion reactions that we get from these conclusions are as follows: Among the reactions listed in Table 1 that were hoped to form compound nuclei with possible survival all class 2 reactions are characterized by values of $(Z^2/A)_{\text{eff}} > 38$ so that compound nucleus formation did not take place at the near-barrier energies used in the experiments. Inasmuch as the class 1 reactions did not seriously test the existence of SHE's because of too unstable, neutron-deficient landing places, none of the many class 2 reactions can be seen as a serious test of the existence of the predicted island of stability because the synthetic route that was chosen does not exist.

The only access to the SHE island in fusion reactions is possible by combining heavy targets with $Z_{\text{88-98}}$ with medium size projectiles with $Z < 27$ ("class 3" reactions). When considering the required availability of weighable amounts of both projectile and target material with $(Z^2/A)_{\text{eff}} < 38$ we find only a few combinations using ^{48}Ca and ^{50}Ti beams with ^{244}Pu , ^{248}Cm and ^{252}Cf targets that fulfill the requirement of unhindered fusion into compound nuclei with possible survival.

But why did the extensive attempts with the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction fail? Both in Berkeley [28,66-68] and at Dubna [55] on the basis of some indirect hints for entrance channel limitations in heavy-ion fusion reactions [53] it was decided that some tens of MeV of extra push had to be provided to fuse ^{48}Ca with ^{248}Cm so that the compound system was formed with excitation energies between 34 and 54 MeV, which probably reduced their overall survival rates to hopelessly small values [53]. Today, in view of the results of the recent high-precision and high-sensitivity fusion studies mentioned above, we conclude that this decision was wrong. The reasons for this conclusion are summarized in Table 2 which compares some of the reactions investigated with the three most promising combinations reaching the island of stability. The excitation energies at the fusion barrier, E_{B}^{X} , are calculated from the experimentally determined barrier heights which are, for $(Z^2/A) < 38$, in agreement with calculated barriers using the proximity theory and reducing the resulting barrier heights by 4% [94]. The same recipe was used to predict fusion barriers for the three most promising superheavy target-projectile combinations and to estimate the respective compound nucleus excitation energies at the barrier, E_{B}^{X} . The column $E_{\text{min}}^{\text{X}}$ lists the lowest excitation energies at which evaporation residues were observed or are expected due to barrier penetration. Thus, it should be possible to form the superheavy compound systems at about 15 MeV excitation energy. Their entrance channel parameters are very similar to those, e.g., of the $^{94}\text{Zr} + ^{124}\text{Sn}$ reaction. However, there exists an important difference. ^{218}Th has a liquid drop barrier of 4-5 MeV at rather large deformations while the superheavy nucleus ^{296}Ows has its stability entirely to shell stabilization with a fission barrier at much smaller deformations. The important

question to be asked is how shell-stabilized narrow fission barriers of spherical nuclei are preserved at higher excitation energy.

4.4.3. A comment on the survivability of shell-stabilized compound nuclei

Sahm et al. [100-102] have studied the deexcitation by neutron emission of $^{214-220}\text{Th}$ produced by fusion of ^{40}Ar with $^{178-180}\text{Hf}$ and of $^{90-96}\text{Zr}$ with ^{124}Sn . The deduced relative neutron decay widths $< \Gamma_n / \Gamma_f >$, i.e. the average values of Γ_n / Γ_f over the neutron cascade, or the cross sections, show no increase at the magic neutron number $N=126$ for 4n or 3n channels. For 2n and 1n channels being observed at much lower excitation energies there is a small increase. As already pointed out by Schmidt et al. [102] these results are consistent with a reduction of the spherical ground state shell effect to 1/e of its full strength by an excitation energy of less than 10 MeV. On the other hand many actinide and transactinide isotopes with $Z < 106$ have been produced [103] in 4n reactions at 35 to 45 MeV of excitation energy. These nuclei are deformed in their ground state and their shell effects seem to be much more stable against intrinsic excitation than those of spherical nuclei. This is dramatically illustrated by Fig. 15 showing the ratio of measured to calculated $2n - 5n$ cross sections for a large number of deformed and spherical compound nuclei [101]. The upper part, where the calculations included the ground state shell effect, shows that for nuclei with deformed ground states the measured cross sections are fairly well reproduced by the calculations. In contrast, for nuclei with spherical or almost spherical ground states, the measured cross sections are much lower than predicted and are reproduced much better by calculations (middle part of Fig. 15) where the shell effects are completely neglected. This is to say that the stabilizing effect of spherical shells on the survival probability of compound nuclei is, if existing at all, extremely weak. One can appreciate the consequences of this result if one realizes that our test cases, i.e. the spherical compound nuclei near $N=126$ have solid (liquid drop) barriers of several MeV even in the complete absence of stabilizing shell effects. Never-

theless, their cross sections in fusion reactions are reduced by up to four orders of magnitude by the damping-out of their shell effects. Superheavy elements near $N=184$, on the other hand, have no stability at all from the liquid drop. Thus, their survival rates will depend entirely on the residual shell strength at the minimum excitation energy possible and at the unavoidable deformation introduced by the entrance channel of the fusion reaction. It is clear that future attempts to synthesize SHE's in the fusion reactions listed in Table 2 will have to be performed at energies 10 to 15 MeV below the calculated fusion barrier energies. The expected excitation energies are then similar to those in the $^{54}\text{Cr}+^{209}\text{Bi}$ reaction so that there is some hope for the formation of a superheavy evaporation residue by the evaporation of one neutron.

4.4.4. New results on deep-inelastic transfer reactions

After the original optimistic extrapolations [70-73] of production rates of SHE's in deep-inelastic collisions the question has been raised [86,75] whether the heavy fragments produced in these collisions are formed in statistical equilibrium and, thus, decay by a neutron evaporation cascade typical for an equilibrated compound nucleus, or, whether they are deformed (under the influence of the large angular momenta involved) to or beyond the fission saddle point. The latter situation would result in a fast non-equilibrium fission process which might be detectable experimentally.

Two very different types of experiments have addressed themselves to this important question. By using 1 m^2 large position-sensitive counters Glässel et al. [86] performed kinematically complete investigations of deep-inelastic collisions followed by sequential fission of the heavy reaction product in $^{238}\text{U}+^{238}\text{U}$ and $^{238}\text{U}+^{248}\text{Cm}$ collisions. These experiments provided no evidence for a fast sequential break-up of the heavy fragments. Even for the largest mass transfers leading to heavy fragments with $Z>110$, the data are still compatible with a two-step reaction

mechanism where fission of the heavy fragment follows the deep-inelastic collisions with minimum scission-to-scission times of $>10^{-20}$ sec. However, the observed fission fragment angular distributions were found [86] to be incompatible with the usual statistical distribution of states at the liquid drop saddle point. Also, in the region of the heaviest elements, Glässel et al. observed a considerable broadening of the fission fragment mass distribution. A possibly related broadening in the mass distribution has also been observed in fusion-fission reactions of lighter elements when the fission barriers reach zero because of very high angular momenta. Irrespective of the apparent two-step nature of the observed fission phenomena, Glässel et al. concluded [86] that non-equilibrated systems may be involved. It is important to note here, that their results refer to values of total kinetic energy loss (TKEL) of >150 MeV in the first reaction step. Similar investigations of the fission phenomena of $Z>110$ fragments at much lower values of TKEL are highly desirable.

The second type of experiments related to the basic question of a statistical equilibrium of the heavy fragments in deep-inelastic collisions was performed by Schädel et al. [72,76,106] by using the extreme sensitivity of radiochemical techniques. In the $^{238}\text{U}+^{238}\text{U}$ reaction the steep decrease in the element yields beyond uranium, see Fig. 7, was followed over eight orders of magnitude up to fermium ($Z=100$) [72]. Analyses of these actinide cross sections indicated that the surviving transuranium isotopes were produced in the low-energy tails of the broad excitation energy distributions at average excitation energies on the order of 35 MeV. In order to reproduce the residual Cf through Fm-yields relative neutron decay widths Γ_n/Γ_f a factor of two smaller than the empirical values by Sikkeland [107] have to be used [76]. This reduction is in agreement with semi-quantitative estimates of the influence of the angular momenta on Γ_n/Γ_f , however, alternatively, a loss of yield due to non-equilibrium fission could not absolutely be excluded. Here, the formation cross sections for transuranium isotopes in the $^{238}\text{U}+^{248}\text{Cm}$ reaction have helped to exclude the latter possibility. These data are shown in Fig. 16, where they are

compared to the cross sections observed in the $^{238}\text{U}+^{238}\text{U}$ reaction. The cross sections for Fm, Es, and Cf in the $^{238}\text{U}+^{248}\text{Cm}$ reaction are three to four orders of magnitude higher than in the $^{238}\text{U}+^{238}\text{U}$ reaction. These increases are qualitatively expected because fewer nucleons have to be transferred to the target nucleus ^{248}Cm to make Cf, Es, Fm or Md isotopes than need to be transferred to ^{238}U in order to produce the same product nuclei. Because the number of transferred nucleons and the intrinsic excitation of the product are correlated it is expected that Cf-, Es- and Fm-fragments produced in $^{238}\text{U}+^{238}\text{U}$ collisions have on the average much higher excitation energies than the same fragments originating from $^{238}\text{U}+^{248}\text{Cm}$ collisions. The resulting larger loss of yield to sequential fission explains the reduced yields of surviving actinides in the $^{238}\text{U}+^{238}\text{U}$ reaction relative to the $^{238}\text{U}+^{248}\text{Cm}$ reaction, see Fig. 16. A quantitative analysis of the observed yield ratios [106] revealed that on the average 3 to 4 neutrons were evaporated in the de-excitation chains leading to surviving heavy actinides in the $^{238}\text{U}+^{248}\text{Cm}$ reaction implying average excitation energies of about 30 MeV. This is entirely consistent with the earlier findings for the $^{238}\text{U}+^{238}\text{U}$ reaction [72] and largely excludes losses of yield to fast non-equilibrium fission processes. Based on these results and on the great similarities that exist in the reaction mechanisms of $^{238}\text{U}+^{238}\text{U}$ and $^{238}\text{U}+^{248}\text{Cm}$ collisions Schädle et al. [106] have asserted that a cross section of 10^{-32} cm^2 at excitation energies of 25-30 MeV is expected for $Z=114$ in the $^{238}\text{U}+^{248}\text{Cm}$ reaction. This is a factor of 30 higher than the estimate for the $^{238}\text{U}+^{238}\text{U}$ reaction.

If excited SHE's can be formed with <15 MeV of excitation energy and with millibarn cross sections in fusion reactions such as $^{48}\text{Ca}+^{248}\text{Cm}$ the deep-inelastic transfer approach appears to be clearly inferior. However, we should keep in mind that with deep-inelastic $^{238}\text{U}+^{248}\text{Cm}$ collisions we are capable of reaching neutron-rich areas in the island of stability inaccessible to fusion reactions, see again Fig. 8. If these neutron-rich species can be formed with little deformation, the chances of surviving their formation may be much better than those of any super-heavy compound nucleus produced in a fusion reaction. Thus, at present,

continued efforts along both alternative synthetic routes are necessary as well as continued studies of both reaction mechanisms which are of great interest in themselves.

5. Outlook

After more than a decade of extensive experimental efforts to search for SHE's in nature and to synthesize SHE's in heavy-ion reactions the anticipated massive extension of the periodic table of the elements has not been achieved. Still, the original reasons for this attempt are valid and motivate for further effort. There is the opportunity to test the predictive power of modern theories describing the stability of atomic nuclei. And there is still hope to test the predictions of chemical properties of the SHE's, predictions based on relativistic rules whose consequence are just becoming apparent in the atomic properties of the heaviest known elements. Experimental tests of these physical and chemical predictions would greatly expand our understanding of the periodic table of the elements.

The failure to observe SHE's has been interpreted [53] as an indication for considerably lower fission barriers of these elements than those predicted in the early "optimistic" calculations. This would preclude their observation in nature and diminish our expectations for the possible production of SHE's in the laboratory. However, as we have discussed in this review, recent insight into heavy-ion reaction mechanisms behooves us to believe that perhaps none of the heavy-ion experiments performed to date has provided a real test for the existence of the predicted island of stability. This is connected i) with a general lack of ultra-sensitive detection techniques for very short-lived species, ii) with the threshold for fusion reactions at $(Z^2/A)>38$ which precludes formation of superheavy compound nuclei in all "class 2" reactions and which leaves only very few target-projectile combinations for meaningful future experiments, and iii) with the extreme sensitivity of the ground state shell effect of spherical nuclei towards intrinsic

excitation which may have reduced the overall production rates of any residual SHE formed either in complete fusion reactions or in deep-inelastic transfer reactions to undetectable values.

For the near future we anticipate

- 1) Further studies on the de-excitation of spherical nuclei.
- 2) Further studies on energy- and system dependences of heavy actinide production in deep-inelastic heavy-ion collisions.
- 3) Developments of new methods for the detection of very short-lived SHE's and their application in future $^{238}\text{U}+^{248}\text{Cm}$ bombardments.
- 4) A repetition of the $^{48}\text{Ca}+^{248}\text{Cm}$ fusion experiments at substantially lower bombarding energies (4.5-4.9 MeV/nucleon) as well as attempts with the $^{48}\text{Ca}+^{244}\text{Pu}$ and $^{50}\text{Ti}+^{248}\text{Cm}$ reactions at comparably low energies.

None of these experiments will be simple because we need tens of milligrams of exotic target materials such as highly enriched ^{244}Pu and ^{248}Cm , large quantities of isotopically separated ^{48}Ca and ^{50}Ti , for which the world-wide supply is very limited, powerful heavy-ion accelerators producing intense beams of these exotic projectiles, highly sophisticated experimental techniques, and a considerable amount of man power. In none of the competing laboratories all these prerequisites are simultaneously fulfilled. This clearly calls for international collaboration.

If the new experiments fail we should keep in mind that in a fusion reaction there is a long way from the deformed configuration at the barrier showing no shell stabilization to the spherical ground state which is shell stabilized. Also deep-inelastic collisions are likely to produce SHE's in deformed shapes. How to bridge this gap is a question for which an answer is not at hand.

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Fig. 1. Predicted half-lives of SHE's indicated as isopleths of constant overall half-life as a function of proton and neutron number. (a) according to Fiset and Nix [12] and (b) according to Randrup et al. [14,15].

Fig. 2. Periodic table of the elements extended into the region of SHE's on the basis of calculations of their ground state electronic structure.

Fig. 3. Neutron multiplicity distribution of the fission events in hot-spring water from the Cheleken peninsula detected in experiments with saturated resin and hydroxides (black dots). The multiplicity distribution for various actinides and two hypothetical spontaneous fission activities are also shown. From Flerov et al. [34].

Fig. 4. Schematic representation of interactions between heavy nuclei leading to fusion into an equilibrated compound nucleus (upper branch) and to a deep-inelastic-collision (lower branch) with characteristic times in units of 10^{-21} s. After Brix [50].

Fig. 5. Predicted decay chain for the evaporation residues of the $^{136}\text{Xe}+^{170}\text{Er}$ reaction tried by Münzenberg et al. [61]. The experiment was optimized for the detection of a rapid sequence of high-energy α -particles followed by a spontaneous fission event, see text.

Fig. 6. Upper limits for the production cross section for superheavy elements produced in the $^{48}\text{Ca}+^{248}\text{Cm}$ reaction. vs. half-life. Labeling of the curves is explained in the text. The data are from experiments of the Dubna group [55] and of the Berkeley-Livermore collaboration [28,66-68].

Fig. 7. Charge distribution of reaction products from quasi-elastic transfer and sequential fission at low excitation energies

(open circles), and for deep-inelastic collisions with the associated mass-symmetric sequential fission product distribution (full circles) in the $^{238}\text{U}+^{238}\text{U}$ reaction at ≤ 7.5 MeV/u [72]. Triangles represent the reconstructed primary distribution of deep-inelastic transfer processes prior to sequential fission. The dashed curve is the result of a diffusion model calculation [73].

Fig. 8. Landing places for the fusion of ^{48}Ca with ^{248}Cm (dots) and for deep-inelastic transfer processes between two ^{238}U nuclei (diamonds) in the contour maps of Fig. 1a, b.

Fig. 9. Upper limits for the production cross section of superheavy elements in collisions of two uranium nuclei as a function of the assumed half-life. For labeling and References, see text.

Fig. 10. Preliminary upper limits for the production cross section of superheavy elements in the $^{238}\text{U}+^{248}\text{Cm}$ reaction [81].

Fig. 11. Upper cross section limits for spontaneously fissioning SHE's in the reaction $^{238}\text{U}+^{208}\text{Pb}$ vs. half-life from Ref. 84.

Fig. 12. Experimental fusion cross sections for $^{40}\text{Ar}+^{122}\text{Sn}$ from Reisdorf et al. [91]. The solid curve is a transmission calculation (WKB) using a proximity potential and assuming rigid sphericity for both target and projectile. The conventional fusion barrier is indicated by the arrow.

Fig. 13. Schematic representation of one-dimensional two-center potentials, $V(R)$, showing a reaction barrier and a pocket behind it. These quantities are defined in the one-dimensional space of two spheres separated by a variable distance R . Also shown are potential curves appropriate to two deformable bodies with fixed masses connected by a neck. Such curves represent cuts in a multi-dimensional deformation space, but there is in general a well-defined

saddle point, named the "conditional" saddle to emphasize the condition of fixed mass asymmetry at variable neck size and variable separation R . The top figure illustrates the cases where the conditional saddle is more elongated than two spheres in contact. In the lower figure the conditional saddle point is more compact than two spheres in contact. Here the existence of a pocket in the one-dimensional potential is not sufficient for the system to get trapped. An extra inward push of kinetic energy is required to bring the system inside the conditional saddle [94].

Fig. 14. Extra push of kinetic energy required to fuse given target projectile combinations [96]. The "fusability" of a given system is characterized by the entrance channel model parameter $(Z^2/A)_{\text{eff}}$ for angular momentum $l=0$. Systems that have been shown to fuse and to produce detectable amounts of evaporation residues are $^{40}\text{Ar}+^{208}\text{Pb}$ [97], $^{50}\text{Ti}+^{208}\text{Pb}$ and $^{50}\text{Ti}+^{209}\text{Bi}$ [98], $^{86}\text{Kr}+^{123}\text{Sb}$ and $^{94}\text{Zr}+^{124}\text{Sn}$ [99], and $^{76}\text{Ge}+^{170}\text{Er}$ [96]. No evaporation residues could be detected for $^{86}\text{Kr}+^{160}\text{Gd}$ and $^{136}\text{Xe}+^{110}\text{Pd}$ [96] indicating the existence of a "thud wall" for fusion beyond $(Z^2/A)_{\text{eff}} \simeq 38$. The solid line and the dashed lines are the result of a fit of Swiatecki's model to symmetric fission cross sections [94,95] using angular momentum dependent values of $(Z^2/A)_{\text{eff}}$ with $(Z^2/A)_{\text{eff}}^{\text{thr}} (l=0)=33\pm 1$ and a ω_{12} . The latter model [94] assumes that barrier crossing is a highly viscous process involving one-body friction.

Fig. 15. Ratio of measured and calculated xn cross section for $2 \leq x \leq 5$ as a function of the residual neutron number from Ref. 101. Upper part: σ_{calc} includes ground state shell effects. Middle part: σ_{calc} calculated by neglecting ground state shell effects. Lower part: Ground state shell effects for spherical nuclei near $N=126$ and for deformed nuclei near $N=152$.

Fig. 16. Cross section for the formation of transcurium isotopes in the $^{238}\text{U}+^{248}\text{Cm}$ reaction at 7.4 MeV/nucleon incident energy, from Ref. 106. Also plotted for comparison are data for the $^{238}\text{U}+^{238}\text{U}$ reaction from Ref. 72.

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Table 1: Reported attempts to synthesize superheavy elements by heavy-ion fusion reactions

Reaction studied	Compound nucleus	Upper-limit cross section for SHE production (cm ²)	Range of half-lives covered	Reference
Class 1: Compound nuclei with vanishing survivability				
$^{48}_{20}\text{Ca} + ^{232}_{90}\text{Th}$	$^{280}_{110}\text{170}$	3×10^{-35}	$> 3 \times 10^{-3}$ s	54
$^{48}_{20}\text{Ca} + ^{231}_{91}\text{Pa}$	$^{279}_{111}\text{168}$	4×10^{-35}	$> 3 \times 10^{-3}$ s	54
$^{48}_{20}\text{Ca} + ^{233}_{92}\text{U}$	$^{281}_{112}\text{169}$	7×10^{-35}	> 2 h	54
$^{40}_{18}\text{Ar} + ^{248}_{96}\text{Cm}$	$^{288}_{114}\text{174}$	2×10^{-32}	10^{-3} s - 1 d	51, 52
$^{48}_{20}\text{Ca} + ^{242}_{94}\text{Pu}$	$^{290}_{114}\text{176}$	1×10^{-35}	2 h - 1 y	55
$^{48}_{20}\text{Ca} + ^{243}_{95}\text{Am}$	$^{291}_{115}\text{176}$	2×10^{-35}	2 h - 1 y	55
Class 2: Dynamical hindrance for fusion				
$^{84}_{36}\text{Kr} + ^{208}_{82}\text{Pb}$	$^{292}_{118}\text{174}$	1×10^{-30}	$> 6 \times 10^{-7}$ s	56
$^{86}_{36}\text{Kr} + ^{208}_{82}\text{Pb}$	$^{294}_{118}\text{176}$	6×10^{-35} 1.5×10^{-30}	3×10^{-3} s - 100 d 2×10^{-9} s - 5 h	57 58
$^{59}_{27}\text{Co} + ^{238}_{92}\text{U}$	$^{297}_{119}\text{178}$	4×10^{-33}	1 s - 30 h	59
$^{60}_{28}\text{Ni} + ^{238}_{92}\text{U}$	$^{298}_{120}\text{178}$	2×10^{-33}	1 s - 30 h	59
$^{65}_{29}\text{Cu} + ^{238}_{92}\text{U}$	$^{303}_{121}\text{182}$	8×10^{-33}	1 s - 30 h	59
$^{65}_{29}\text{Cu} + ^{238}_{92}\text{U}$	$^{303}_{121}\text{182}$	2×10^{-33}	2 h - 1 y	60
$^{65}_{29}\text{Cu} + ^{238}_{92}\text{U}$	$^{303}_{121}\text{182}$	4×10^{-34}	2×10^{-6} s - 10 h	61
$^{76}_{32}\text{Ge} + ^{232}_{90}\text{Th}$	$^{308}_{122}\text{186}$	1×10^{-34}	5×10^{-3} s - 1 y	63
$^{136}_{54}\text{Xe} + ^{170}_{68}\text{Er}$	$^{306}_{122}\text{184}$	1.5×10^{-33}	2×10^{-6} s - 10 h	61
$^{76}_{32}\text{Ge} + ^{238}_{92}\text{U}$	$^{314}_{124}\text{190}$	1×10^{-34}	5×10^{-3} s - 1 y	63
$^{68}_{30}\text{Zn} + ^{243}_{95}\text{Am}$	$^{311}_{125}\text{186}$	5×10^{-32}	10^{-9} s - 5 d	64

Table 1 cont'd:

Reaction studied	Compound nucleus	Upper-limit cross section for SHE production (cm ²)	Range of half-lives covered	Reference
$^{84}_{36}\text{Kr} + ^{232}_{90}\text{Th}$	$^{316}_{126}\text{190}$	5×10^{-30}	$> 6 \times 10^{-7}$ s	56
$^{84}_{36}\text{Kr} + ^{238}_{92}\text{U}$	$^{322}_{128}\text{194}$	8×10^{-29}	$> 6 \times 10^{-7}$ s	58
$^{84}_{36}\text{Kr} + ^{238}_{92}\text{U}$	$^{322}_{128}\text{194}$	9×10^{-36}	2 h - 100 d	65
Class 3: Compound nuclei with possible survival				
$^{48}_{20}\text{Ca} + ^{246}_{96}\text{Cm}$	$^{294}_{116}\text{178}$	2×10^{-35}	2 h - 1 y	55
$^{48}_{20}\text{Ca} + ^{248}_{96}\text{Cm}$	$^{296}_{116}\text{180}$	see Fig. 5	55, 28, 66-68	

Table 2: Entrance and exit channel parameters for several heavy-ion fusion reactions with $(Z^2/A)_{\text{eff}} < 38$. The height of the fission barrier at zero excitation energy is given as $E_f = B_f - \delta U$, if an additional (small) shell effect at the barrier is ignored.

Reaction	Compound nucleus	Fission barrier B_f^a δU^b	Fusability $(Z^2/A)_{\text{eff}}$	Fissility (Z_o^2/A_o)	Fusion barrier B (MeV)	Excitation energy E_B^x	Energy (MeV) E_{min}^x	lowest xn channel observed	Reference
$^{40}_{18}\text{Ar} + ^{206}_{82}\text{Pb}$	$^{246}_{100}\text{Fm}$	1.5 -2.3	31.3	40.7	162 ± 3	33	19	2 n	97
$^{50}_{22}\text{Ti} + ^{208}_{82}\text{Pb}$	$^{258}_{104}$	0.8 -2.9	34.4	41.9	191 ± 3	22	18	1 n	98
$^{50}_{22}\text{Ti} + ^{209}_{83}\text{Bi}$	$^{259}_{105}$	0.6 -3.0	34.7	42.6	190 ± 3	15	12	1 n	98
$^{54}_{24}\text{Cr} + ^{209}_{83}\text{Bi}$	$^{263}_{107}$	0.4 -2.9	36.6	43.5	~ 209	20	15	1 n	62, 98
$^{86}_{36}\text{Kr} + ^{123}_{51}\text{Sb}$	$^{209}_{87}\text{Fr}$	7.2 -4.6	35.6	36.2	209 ± 3	40	28	3 n	99
$^{94}_{40}\text{Zr} + ^{124}_{50}\text{Sn}$	$^{218}_{90}\text{Th}$	5.3 -4.3	37.0	37.2	219 ± 3	31	20	1 n	99
$^{100}_{42}\text{Mo} + ^{124}_{50}\text{Sn}$	$^{224}_{92}\text{U}$	4.3 -0.4	37.7	37.8	229 ± 3	29	18	1 n	100
$^{48}_{20}\text{Ca} + ^{248}_{96}\text{Cm}$	$^{296}_{116}$	0 -4.9	33.9	45.5	196^c	25^d	<15		
$^{50}_{22}\text{Ti} + ^{244}_{94}\text{Pu}$	$^{294}_{116}$	0 -3.6	36.2	45.8	212^c	28^d	<18		
$^{50}_{22}\text{Ti} + ^{248}_{96}\text{Cm}$	$^{298}_{118}$	0 -5.7	36.6	46.7	216^c	28^d	<18		

- a) empirical liquid drop fission barriers, from Ref. 104
b) ground state shell correction, from Ref. 105
c) calculated using the proximity theory and reducing the barrier height by 4%
d) relative to the droplet model ground state in the absence of shell effects, Ref. 105

[illegible]

Fig. 5

Fig. 6

Fig. 7

Fig. 11

Fig. 10

Fig. 9

Fig. 8

Fig. 12

Fig. 13

Fig. 14

Fig. 16

Fig. 15