ON THE PRODUCTION OF HEAVY ELEMENTS BY COLD FUSION: The Elements 106 to 109

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1. INTRODUCTION—EARLY CHARGED-PARTICLE REACTIONS TO PRODUCE HEAVY ELEMENTS

After a short historical introduction (Section 1), this article presents new insights into the mechanism limiting the fusion of heavy nuclides (Section 135)

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2. Fusion is finally limited by the increasing Coulombic forces in the formation process of a compound system, as well as in its deexcitation. Moreover, nuclear structure effects in all stages of evaporation residue (EVR) formation are shown to be of importance. The wide field of fusion reaction studies and possible experimental techniques is projected onto the task of element synthesis, and only those aspects that are of relevance here are covered. The better understanding of EVR formation (Section 2) and the new experimental techniques (Section 3) that enabled the production of elements 107–109 (Section 4) are also discussed. In Section 5 ground-state properties and the nuclear structure of the heaviest isotopes, together with their production cross sections, are discussed. Finally, an outlook on how eventually to go beyond \( Z = 109 \) is given.

Isotopes of elements beyond Md were synthesized by fusion of heavy nuclei. This nuclear reaction was most successful in heavy-element production: The elements 102–109 were made during the last 25 years and about 40 isotopes of these elements were known in 1984. The synthesis of elements 102 to 106 started from the heaviest possible actinide targets fused with light projectiles ranging from \(^{14}\)B to \(^{26}\)Mg. This technique depends on the availability of heavy actinides in microgram quantities, and was widely used with the accelerators of LBL and ORNL in the US and of JINR in the USSR. The Berkeley and Dubna groups developed the pioneering techniques of using heavy-ion accelerators to fuse nuclei and detection systems to identify the new species. The many experiments done between 1955 and 1975 gave us the first five elements, 102–106, produced by fusion reactions and detected first not by chemical methods, as the elements from Pu to Md, but by careful analysis of the radioactive decay of the nuclear species. A gradual shift from chemical methods to physical methods occurred.

In the years 1955–1965, the years of apprenticeship, ingenious new methods were invented and perfected. Many isotopes were discovered, but retrospectively many of them had wrong mass assignments, wrong \( \alpha \) energies, decay times, and branchings, and very often the claims of discovery did not withstand rigorous inspection. For elements detected by physical methods, no clear criteria for naming elements exist. Element naming was taken by scientists as a right in itself. Claims have been maintained in spite of experiments proving the contrary; and controversy over the names of elements continues even now. From 1965 to 1975 results were consolidated. The number of new conflicting results became smaller and smaller. The method of fusing actinide target atoms with light projectiles had reached great perfection. Most of the data now accepted were obtained at Berkeley and Dubna during this time. The reactions investigated before 1969 are compiled in Reference (1). The more recent results are reviewed in References (2, 3).

The heaviest element made starting from actinide targets is 106 (4). The experiment performed at Berkeley in 1974 established a preliminary limit to the method. \(^{249}\)Cf bombarded by \(^{18}\)O yields \(^{261}\)106 via a 4n-reaction channel; 87 atoms of \(^{261}\)106 were identified via mother-daughter correlations. The \( \alpha \) lines of \(^{259}\)104 and \(^{255}\)102 known previously were correlated to \(^{261}\)106. The \( \alpha \) isotope decays via an \( \alpha \) decay, with a half-life of 0.9 + 0.2 s; and \( \alpha \) energies of 9.06 and 9.25 MeV were measured. The production cross section measured at 95-MeV bombarding energy is 0.3 nb. Dubna experiments (5) assign a fission branch to the isotope. Fission activities were later seen at Berkeley (6) as well, but were not assigned. The cross section obtained for the fission activity was 0.6 nb at Dubna and 9 nb at Berkeley. The fission branch of \(^{261}\)106 remains questionable, as the fission activity could well come from a target-like transfer product.

Different experiments designed to go beyond element 106 have been performed by bombarding \(^{249}\)Cf and \(^{249}\)Bk with \(^{20}\)Ne beams (7, 8). Atoms of elements 107 and 108 could not be detected in the large background of transfer products, which could not be suppressed sufficiently. We return to the actinide target reactions at the end of this article, in Section 5.5, and discuss future possibilities of the technique.

The possibility of using other nuclear reactions has been investigated thoroughly, but the results are discouraging. Successive neutron captures and subsequent \( \beta \) decays lead to \(^{251}\)Fm, but the short half-life of \(^{251}\)Fm (0.4 ms) prevents further buildup. Transfer of nucleons from lighter nuclei to the heaviest targets, such as \(^{248}\)Cm, \(^{249}\)Bk, \(^{250}\)Cf, and \(^{254}\)Es, produced a number of fission activities, which until now could not be assigned (6, 9, 10), but which might reflect isotopes of elements 102 and 103. The use of deep inelastic reactions for heavy-element production was disappointing, because the transfer of many nucleons is difficult to achieve without heating the highly fissileable heavy isotopes, and thus reducing their survivability to cross sections too small for current detection techniques (11, 12).

2. LIMITATIONS OF EVAPORATION RESIDUE FORMATION IN FUSION REACTIONS

We do not consider the disappearance of evaporation residue (EVR) formation at high bombarding energies here; instead we restrict ourselves to limitations at energies near the Coulomb barrier, which are relevant for heavy-element production. Following the assumption of a two-step formation process for EVRs—first step of amalgamating two nuclei to a compound system, and a second step of deexcitation of the compound system—we discuss limitations in the entrance channel (the compound nucleus formation) and then restrictions in the exit channel (the compound nucleus
deexcitation). An adequate presentation of the many facets of the physics of EVR formation goes beyond the scope of this article; only those aspects important for the synthesis of heavy elements are reviewed.

Because the formation cross sections for the heavy elements are smaller than 10 nb, that is $10^{-7}$ of the total reaction cross section, it is impossible to perform directly these investigations within reasonable beam times at EVRs of the elements in question. Therefore, the studies on entrance channel limitations were performed mostly with compound systems in the element range $Z = 80$--90. Studies on the deexcitation stop at the compound systems of element 104. Moreover, at a level below $10^{-3}$ of the total cross section, it becomes more and more difficult to make numerical predictions. We must combine experimental results with general theoretical concepts, such as "subbarrier fusion" (13, 14) and "extra-push energy" (15, 16) in order to gain some guidance for the reactions of interest. Subbarrier fusion may increase the fusion cross sections of heavy systems, before increasing Coulomb forces finally limit the formation of a compound system. It is shown below that nuclear structure effects are of importance for EVR formation up to the heaviest systems. Subbarrier fusion and the dynamical limitation of compound system formation are covered in Section 2.1, whereas the deexcitation of the compound system in its relevance to heavy element synthesis is discussed in Section 2.2.

2.1 The Entrance Channel Limitations

The phenomenon of increased subbarrier fusion of heavy systems was discovered experimentally (17, 18), whereas the limitation of compound system formation had been foreseen by theory early on (19). It seems appropriate to start the discussion with an experiment demonstrating both effects. Figure 1 shows the xn cross sections for the reaction $^{92}$Zr($^{90}$Zr, xn)$^{182}$Zr (20) together with the fusion barrier $B_B$ calculated using a standard potential—the Bass potential (21), which allows one to reproduce fusion barriers for all the light systems. The measurement shows that there is fusion far below the barrier, and at the same time still a smooth increase over a large energy range beyond the barrier.

In order to discuss the experiments we introduce a fusion probability $p(E)$ for central collisions (22). This quantity together with a survival probability $w(E + Q, l)$ allows us to separate numerically the phenomena in the entrance and exit channels. The procedure is especially adequate for highly fissionable compound systems, for which only in a small $l$ window near the zero angular momentum will EVRs surviving fission be found. The cross section for EVR formation at the kinetic energy $E$ in the c.m.s. system can be formulated by using the quantities $p(E, l)$ and $w(E + Q, l)$:

$$
\sigma(E) = \pi \lambda^2 \sum (2l + 1)p(E, l)w(E + Q, l).
$$

Here $E + Q$ is the excitation energy $E^*$ in the compound nucleus, $Q$ is the $Q$ value of the reaction, and $\lambda$ is the de Broglie wavelength corresponding to the entrance channel. If the compound nucleus hypothesis is assumed to be valid $w(E^*, l)$ does not depend on the entrance channel. We can now define an angular-momentum-weighted average of the fusion probability $\langle p(E, l) \rangle$:

$$
\langle p(E, l) \rangle = \pi \lambda^2 \sum (2l + 1)p(E, l)w(E + Q, l)/\pi \lambda^2 \sum (2l + 1)w(E + Q, l).
$$

This fusion probability $\langle p(E, l) \rangle$ can be determined from the measured EVR cross section $\sigma(E)$, provided $w(E^*, l)$ is known. The function $w(E^*, l)$ can either be determined from the EVR cross section of a corresponding asymmetric reaction (22), which allows us to determine $w(E^*, l)$ in an energy region undisturbed by entrance channel effects, or it can be determined from an evaporation cascade calculation (23), which has been tested to
reproduce EVR cross sections adequately (20, 24). The \( l \) dependence of \( \psi(E^*, l) \) is determined by the \( l \) dependence of the fission barrier, which is assumed to follow the rotating liquid drop model (25). This \( l \) dependence restricts the \( l \) values contributing to EVR formation to a range much smaller than the broad distribution of angular momenta in the primary fusion process. Because the weighting function \( \psi(E^*, l)(2l+1) \) used to obtain \( \langle p(E, l) \rangle \) is a narrow function peaking at all energies at small \( l \) values \((\sim 15)\), \( \langle p(E, l) \rangle \) becomes about \( p(E, l = 0) \) and the term "fusion probability for central collisions" \( p(E) \) is justified.

Figure 2 shows fusion probabilities for three systems: \(^{40}\text{Ar} + ^{180}\text{Hf} \) (26) showing subbarrier fusion, \(^{90}\text{Zr} + ^{90}\text{Zr} \) (27) approximating best a one-dimensional tunneling model (28), and \(^{90}\text{Zr} + ^{124}\text{Sn} \) showing a strong dynamical hindrance (22). In Figure 3 we give a scheme for the general dependence of \( p(E) \). It is characterized by three energies, the adiabatic barrier \( B_a \), the Bass potential barrier \( B_b \), and the dynamical barrier \( B \). The logarithmic slope \((\hbar \omega)^{-1} \) at small energies, and the two parameters, \( \sigma \) the barrier fluctuation parameter and \( c \) a truncation parameter, are also of importance. The latter can be replaced by the values of \( p(E) \) at the energies \( B_a \) and \( B_b \). The term \( B_a - B_b \) characterizes subbarrier fusion and \( B_b - B \) the energy shift due to the dissipation losses in the entrance channel, the extra-push energy. This six-parameter presentation describes both the subbarrier fusion effects and the entrance channel limitations. It replaces the two parameters \( B_a \) and \( \hbar \omega \) of a one-dimensional tunneling model (28).

The fusion probability \( p(E) \) may be presented as a sum over the tunneling through a sequence of barriers that are distributed like a Gaussian around a nominal fusion barrier \( B \) defined by \( p(B) = 0.5 \) (29):

\[
p(E) = (2\pi\sigma^2)^{-1/2} \int_{-\infty}^{\infty} \left[ \exp \left( -(E - B)^2 / 2\sigma^2 \right) \right] T(E - E') \, dE',
\]

with \( \sigma \) the width of the barrier distribution and \( T \) the transmission coefficient through a one-dimensional barrier calculated using a WKB approximation. As there is always a lowest barrier, \( B_a \)—the adiabatic barrier—that finally makes \( p(E) \) decrease with a slope given by \((\hbar \omega)^{-1} \), the lower integration limit may be truncated and replaced by a truncation parameter \( c \). For subbarrier fusion, Equation 3 relates the parameters \( \sigma \) and \( c \) to the adiabatic barrier \( B_a \) and the fusion probability \( p(B_a) \), which follow directly from the measured energy dependence of the fusion probability \( p(E) \).

The subbarrier fusion phenomena have been analyzed thoroughly. Static deformation of a collision partner increases fusion below the barrier, as the fusion barriers for the different orientations of the collision partners in
fission process must be small enough to allow the formation of a fused monosystem. Figure 4 demonstrates that nature has set a limit to combining two nuclei with atomic number \( Z_p \) and \( Z_T \) to form a nucleus with atomic number \( Z_p + Z_T \). Systems that were fused successfully are separated from combinations leading to no EVRs. The line of separation is characterized by a well-defined ratio of Coulomb to nuclear forces acting between the two amalgamating collision partners. Beyond that line open points indicate the different failures to produce superheavy elements. For \( Z_p + Z_T > 120 \) the very first step in the formation of an EVR, the overcoming of the Coulomb barrier, already becomes impossible for any combination of \( Z_p \) and \( Z_T \). In the range \( 120 > Z_p + Z_T > 80 \) fusion is hindered, not every combination of \( Z_p \) and \( Z_T \) is successful, e.g. the isotope \(^{244}\text{Pm}\) was made in the reaction \(^{206}\text{Pb}(^{40}\text{Ar},2n)^{244}\text{Pm}\), but could not be made out of the nearly symmetric partners \(^{110}\text{Pd}\) and \(^{136}\text{Xe}\) (34).

The ratio of disruptive Coulomb forces and attractive surface tension forces governs the amalgamation of two nuclei into one. For a monosystem this ratio is given by the fissility parameter \( x \). For a two-touching sphere configuration, Bass (35) defined a corresponding parameter making use of the proximity force. Taking into account that the proton and neutron ratio between the two partners is equilibrated very quickly \((10^{-22} \text{ s})\), and that the nuclear system at the Coulomb barrier for those systems of importance here

**Figure 3** Six parameters defining schematically the fusion probability. At low energies the slope \((\hbar \omega)^{-1}\) describes the Hill-Wheeler barrier penetration of the lowest possible adiabatic barrier \( B_p \). Subbarrier fusion is parametrized by \( P(B_p) \) and the subbarrier shift \((B_p - B_s)\), the dynamical hindrance by \( P(B_p) \) and the dynamical shift (extra-push) by \((B - B_s)\).

**Figure 4** Elements \( Z_p \) and \( Z_T \) fused to element \( Z_p + Z_T \). Each combination is a point in the triangle. Successful fusion reactions (in GSI experiments over the years detected by EVRs) are indicated by full points, failures by open points (36). Between \( x \) and \( x \) is indicated the production goes down below the 100-pb level. Between \( Z = 90 \) and \( Z = 120 \), more and more asymmetric combinations are necessary to produce surviving EVRs (34). Beyond \( Z > 120 \) no fusion is possible.
is more compact than the two-touching sphere configuration, a modified parameter \( x \), describing the ratio of Coulomb and nuclear forces has been defined and applied to organize the vast amount of data (34, 36):

\[
x_{\text{mean}}^{\text{UCD}} = 2x(κ² + κ + κ⁻¹ + κ⁻²)⁻¹/² \quad \text{with} \quad x = (Z²/A)/(Z²/A)_{\text{crit}};
\]

\[
(Z²/A)_{\text{crit}} = 50.88[1−1.78(N−Z/N+Z)²] \quad \text{and} \quad κ = (A_1/A_2)¹²³.
\]

Passing the Coulomb barrier, an appreciable amount of the radial energy on the long way from a two-touching nuclei configuration to a compound system is transferred into intrinsic excitation energy. The dissipated energy is reflected as a virtual increase of the Coulomb barrier; an extra-push energy is needed to fuse. The extra-push model introduced by Swiatecki (15) predicts a parabolic dependence of the extra-push energy on the scaling parameter \( x_{\text{mean}}^{\text{UCD}} \):

\[
B - B_0 = a²E_0(x_{\text{mean}}^{\text{UCD}} - x_{\text{thr}})² \quad \text{with}
\]

\[
E_0 = 7.6 × 10⁻⁴(Z²/A)²_{\text{crit}}A¹¹⁳(A¹¹³ + A¹⁹³)/(A_1 + A_2)³,
\]

where \( x_{\text{thr}} \) is a threshold value describing the onset of entrance channel limitation and \( a \) is a slope parameter. The numerical values for the parameters are calculated or fitted to the data (37). The calculation (38) gives \( x_{\text{thr}} = 0.72 \) and \( a = 22 \). The onset of entrance channel limitations is predicted by all friction models (37–40), but it is still open which of the models describes the data best. Accurate measurements of the quantity \( B - B_0 \) should finally settle which mechanism governs the dissipative losses in the fusion of heavy nuclei.

In Figure 5 are compiled all extra-push values obtained from fits to \( p(E) \) for different systems leading to EVR formation (20, 22, 24). The quantity \( B - B_0 \) with the above parameters follows roughly a quadratic dependence as predicted by the one-body dissipation model. It should be mentioned that the analysis of symmetric binary fragmentation data yields smaller extra-push energies and the limitation sets in at higher x values (41, 42). Binary fragmentation includes the cross section of quasi-fission; that is, a contribution leading to a symmetric fragmentation but not necessarily to an equilibrated compound system. For quasi-fission, which needs smaller overlap of the collision partners, we would indeed expect smaller losses by dissipative processes. The data of Figure 5 indicate the importance of nuclear structure effects in addition to the macroscopic dissipation losses. The smallest extra-push energies are observed for spherical, closed shell, collision partners. The \(^{48}\text{Ca} + ^{208}\text{Pb} \) collision system is found to fuse without any hindrance (24); \(^{90}\text{Zr} \) in the bombardments by \(^{124}\text{Sn} \) and \(^{90}\text{Zr} \) beams shows the smallest extra-push energy of all Zr isotopes (20, 22). Systems with several neutrons outside the \( N = 50 \) shell in Zr isotopes and two protons outside of the \( Z = 20 \) shell in \( N = 28 \) isotopes show considerably larger extra-push energies. The nuclear structure effects may contribute to the extra-push energies up to 10 MeV. These nuclear structure contributions to the barrier shifts have to be corrected for in order to obtain the dissipative losses in the macroscopic models. The macroscopic friction models are a first approximation that will describe the gross features, but to make any predictions for an actual collision system nuclear structure effects have to be considered.

Scaling of the extra-push energies may be possible for the macroscopic contribution, but certainly not for the microscopic, nuclear structure part. Whether the large values of the barrier fluctuation parameter observed for entrance-channel-limited systems, \( σ ≈ 10 \text{ MeV} \) (22, 24), reflect subbarrier fusion of microscopic origin or are a macroscopic property of the system is an open question. The analysis shows the barrier fluctuation parameter \( σ \) to increase with increasing extra-push energy \( (B - B_0) \). The shift of the dynamical barrier becomes a quantity that fluctuates with an increasing width around its mean value \( B \). The fluctuations getting comparable to the absolute value of the shift make the concept of a well-defined barrier doubtful. For the systems in question the potential energy surface shows no well-defined saddle point, but a large plateau region where small dynamical fluctuations will finally decide into which direction the system develops, to a binary or to a monosystem. The terms \( B - B_0 \) and \( σ \) seem to be coupled parameters, defining together the fusion probability at the Bass barrier, \( p(B_0) \), a quantity characterizing the hindrance in the entrance channel at the energy of the Bass barrier. The value of \( p(B_0) \) may be derived directly from the cross-section value at the barrier energy \( B_0 \).

Figure 6 shows \( p(B_0) \) as a function of \( x_{\text{mean}}^{\text{UCD}} \) for all systems investigated. In this presentation the cross-section values known for heavy-element produc-
tion beyond $Z = 104$ have been included (Section 5.4). Again, as in Figure 5, nuclear structure effects are evident. They modify $p(B_a)$ by factors larger than 10. A linear fit to all the data points gives for $p(B_a)$ an exponential dependence from $x_{\text{UCD}} - x_{\text{thr}}$ with a slope parameter $d$ [ln $p(E)/dx = 71$, equivalent to a factor of two for a change of 0.01 in $x_{\text{mean}}$.

$$p(B_a) = 0.5 \exp[-71(x_{\text{UCD}} - x_{\text{thr}})].$$

The entrance channel effects in the fusion of heavy nuclei extracted from the cross sections experimentally via $p(E)$, reduced to the energies $B_n$, $B_n - B_{\text{thr}}$, and the value of the slope parameter $\alpha$, the values of $p(E)$ at energies $B_n$ and $B_{\text{thr}}$ must be understood on the one hand from the macroscopic parameters describing tunneling, the fusion barrier, and the dissipation of energy of colliding, charged, liquid drops, and on the other hand from nuclear structure effects such as deformation, vibrational modes, and $Q$ values of competing transfer channels. Both theory and experiment are still far from being able to make reliable predictions to support the search for favorable new collision systems, systems that would eventually produce elements beyond the atomic numbers reached so far. The understanding of entrance channel effects is the most challenging task for future investigations, both for theoreticians and for experimentalists. New theoretical microscopic approaches, besides the macroscopic concepts, will have to be applied to the heavy-element synthesis problem, for example the “dissipative diabatic dynamics” of Nörenberg and co-workers (43–45).

2.2 Exit Channel Limitations

Newly born compound systems are hot. The nucleons in the compound system are excited. Excitation energies of 40–50 MeV at the barrier are found for many reactions. As long as the fission barriers are high, this excitation energy is dissipated by a cascade of evaporated particles. However, for small fission barriers the system may be destroyed by fission competing with particle evaporation. The probability of fission during the cooling-down phase of the fused compound system must be small enough to guarantee production rates of at least a few atoms per week. The losses during deexcitation of a fused compound system depend on the stability of nuclei against fission in states far above the ground state. The temperature dependence of the fission barrier is of great importance, as the production cross section of heavy elements is governed by its dependence on intrinsic excitation energy. To minimize the fission losses in the deexcitation phase, the highly fissionable nuclei should be produced with the smallest excitation energy possible, as each step in the competition between neutron emission and fission contributes to the losses. For a given compound system, a high survival probability is obtained if the number of steps in the evaporation cascade is kept small and if for each step the fission barrier has its largest possible value. Systems with the smallest possible excitation energies and angular momenta will survive.

The macroscopic fission barrier $B_{\text{fiss}}$, the barrier protecting a charged liquid drop against spontaneous fission, and the extra-push heating $B - B_{\text{nuc}}$ are closely related, as both depend on the value of the fissility parameter $x$ (defined in Equation 4). Decreasing fission barriers of liquid drops go together with increasing extra-push heating. Fortunately, the contributions of nuclear structure effects to the fission barrier and to the fusion probability are decoupled. We may try to profit from nuclear structure effects twice: in the entrance channel in order to fuse systems that would not fuse without nuclear structure effects, and in the exit channel in order to produce shell-stabilized heavy nuclei that would not survive deexcitation without shell stabilization.

2.2.1 Cold Fusion The minimum excitation energy $E_{\text{exc}}$ of a compound system is the sum of the energy $B$ to pass the Coulomb barrier and the negative reaction $Q$ value. In Figure 7 the minimum excitation for an element $Z_p + Z_T$ made out of a projectile $Z_p$ and a target nucleus $Z_T$ is shown. Those isotopes leading to the smallest excitation energy for a given combination have been chosen. “Extra-push” energy according to the parameters of the compound system of Figure 5 is added. The size of the squares runs from 15 to
neutrons in the early fusion studies at Berkeley and Dubna leading to elements \( Z = 101 - 106 \) (1-3) via two-neutron reactions found at Dubna in 1974 (49), and one-neutron deexcitation found in 1980 at Darmstadt (50). Deexcitation by pure \( \gamma \) emission was found recently in the reactions \( ^{90}\text{Zr} + ^{90}\text{Zr} \rightarrow ^{180}\text{Hg} \) at Darmstadt (46) and \( ^{90}\text{Ti} + ^{208}\text{Pb} \rightarrow ^{258}104 \) at Dubna (51). Excitation functions for the reaction \( ^{90}\text{Zr} + ^{90}\text{Zr} \) have been measured. They show below the barrier at an excitation energy of 17 MeV the one-proton, one-neutron, and the \( \gamma \) channels (see also Figure 1). The excitation function of the \( \gamma \) channel peaks at 20 MeV and has a width of only 6 MeV. The reaction combines all three mechanisms and leads to a cold fusion reaction. Heavy, symmetric, and shell-stabilized collision partners are fused below the barrier with no hindrance by extra-push heating. The cross section for "radiative fusion" is high, about 50 \( \mu \)b, as fission competition is still small. The fused system \( ^{180}\text{Hg} \) is situated at the triple point of the \( (N,Z) \) plane, where proton and neutron separation energies and the fission barrier are about equal. Around \( ^{180}\text{Hg} \) we find the nuclei the most resistive to intrinsic excitation. If these are produced cold, they reveal the long-sought "radiative fusion," the ultimate in cold fusion with surprisingly large cross sections. The large cross sections are reproduced by evaporation calculations assuming an E1 strength increased by a factor of five (20, 52, 53).

2.2.2 \textbf{Temperature Dependence of Shell Stabilization} The fission barrier protects the compound systems against immediate disintegration. The fission barrier is made up of two contributions, a smoothly varying macroscopic liquid drop part and a shell correction, mainly of the ground-state mass. Near element 106 the macroscopic fission barriers fall below 1 MeV. As is shown in Section 5.1 the stability beyond is governed by the shell correction of the ground-state masses alone. The fluctuations of the level density around a continuously increasing Fermi-gas level density determine the shell corrections, which strongly depend on the excitation energy and the deformation of the nuclear system. The study of the temperature dependence of the shell contributions to the fission barrier makes it possible to predict which excitation energy a shell-stabilized compound system may take in order to survive the deexcitation process.

Shell-stabilized, highly fissionable nuclei are found among the actinides. Using these nuclei produced by different nuclear reactions, one can study the contribution of shell effects to EVR formation at excitation energies varying from 10 to 60 MeV. We differentiate between shell-stabilized deformed and spherical nuclei. We choose the energy dependence of \( \Gamma_\pi/\Gamma_\ell \) for heavy actinide isotopes, as presented in (54, 55), to discuss deformed nuclei. For shell-stabilized spherical nuclei we refer to highly fissionable \( N = 126 \) nuclei, such as \( ^{216}\text{Th} \) (26, 56, 57).
A comparison of EVR cross sections for pairs of reactions leading to two neighboring compound systems gives $\Gamma_n/\Gamma_f$ as a function of excitation energy $(54, 55)$. All steps of the evaporation cascade except the first step are assumed to be equal for an $xn$ reaction and $(x+1)n$ reaction leading to the same EVR:

$$\frac{\Gamma_n}{\Gamma_f}(E^*_{n+1}) = \frac{\sigma_{n+1}/\sigma_{\exp}}{P_x(E^*)/P_{x+1}(E^*_{n+1}) \times \sigma_{CN}(E^*_n)/\sigma_{CN}(E^*_{n+1})}.$$

$P_x$ is the probability of emitting $x$ neutrons at an excitation energy $E^*_x$, $\sigma_{CN}$ is the cross section to produce the compound system at an excitation energy $E^*_n$, $P_x$ and $\sigma_{CN}$ are calculated following standard methods $(58, 59)$. The experimental values for $\Gamma_n/\Gamma_f$ obtained for three nuclei are presented in Figure 8. The values given refer to an angular momentum of zero. The $\Gamma_n/\Gamma_f$ values depend only weakly on the energy $E^*$ in the range investigated. The few points in the range $10–20$ MeV are even larger than at higher energies. The results of two calculations are given for comparison. The full line shows the expectation for $\Gamma_n/\Gamma_f$ with shell effects entering into level densities and the fission barriers. An exponential damping of the shell effects with $E_D = 18$ MeV, which follows from microscopic models, is used in the calculation $(60, 61)$. The damping of shell effects leads to the shallow minimum of $\Gamma_n/\Gamma_f(E^*)$, which becomes deeper for systems dominantly shell stabilized. The hatched line is calculated without any shell effects, and obviously the observed increase of $\Gamma_n/\Gamma_f$ in the low energy range cannot be explained. The measured cross sections must include shell effects for all the deformed nuclei analyzed. There would be no heavy elements made by fusion, unless $\Gamma_n/\Gamma_f$ were kept large at low excitation energies by shell effects.

![Figure 8](image)

Figure 8. $\Gamma_n/\Gamma_f(E^*)$ for the three deformed compound systems $^{244}$Cm, $^{248}$Cm, and $^{258}$Cf. Experimental values are taken from pairs of excitation functions $(54, 55)$. The lines indicate two calculations, full line with shell effects, dotted line without shell effects $(58, 60)$.

A very different behavior was found in experiments near $^{216}$Th. Here nuclei are spherical, highly fissionable, and again their barriers are partly shell stabilized. They may be produced within a range of excitation energies between 20 and 50 MeV. These nuclei are the best approximation to the shell-stabilized spherical superheavy nuclei near $N = 184, Z = 114$. Figure 9 shows measurements of the survival probability for thorium isotopes produced by $4n$ reactions $(57)$. Two calculations of survival probabilities are shown. The measured cross sections are smaller by about two orders of magnitude compared to an evaporation cascade calculation that included the ground-state shell effects in the fission barriers and a damping of shell effects with $E_D = 18$ MeV $(60, 61)$. A calculation with a reduced value of $E_D = 6$ MeV is given for comparison. It reproduces the trend of the experimental data fairly well.

Figure 10 shows a comparative study of EVR formation cross sections in the excitation energy range of $40–50$ MeV. Measured cross sections for nuclei with $B_f < B_n$ are compared to calculations $(36, 62)$. The deformed nuclei in the range $N = 140–155$ have shell corrections of their fission barriers similar to those of the spherical nuclei around $N = 126$ (Figure 10, bottom). The inclusion of shell effects with a damping energy of $18$ MeV reproduces well all data for deformed nuclei (Figure 10, top). However, for spherical nuclei the measured values are three orders of magnitude smaller than calculated. Neglecting all shell effects (Figure 10, middle) reproduces the data for spherical nuclei, whereas for deformed nuclei the calculated cross sections would be off by many orders of magnitude, in agreement with the analysis of Figure 8.

We know there is additional ground-state stability of spherical nuclei due to shell effects, which must be observable somewhere in the production.

![Figure 9](image)

Figure 9. The survival probability $w(E^*)$ for spherical Th EVRs produced by $4n$ reactions as a function of the neutron number of the EVR $(57)$. Circles are from $(^{40}$Ar+$\alpha\overline{H})$, squares are from $(^{48}$Ca+$\beta\gamma)$ reactions. The lines give two calculations using different damping energies for shell effects $(60)$; full line $E_D = 18$ MeV, dashed line $E_D = 6$ MeV.
cross sections at low energies. The analysis of the data shown in Figure 9 is compatible with shell effects becoming important at energies below 15 MeV ($E_0 = 6$ MeV). At higher energies the spherical compound systems behave as if no shell stabilization exists, whereas in deformed compound systems the shell stabilization is of importance up to 40–50 MeV ($E_0 = 18$ MeV). Introducing the well-known concept of collective enhancement of level densities into the calculation (63), one can explain the small experimental cross sections (64). A deformed nucleus like $^{252}$Fm is not affected by the collective enhancement of level densities, whereas $^{210}$Th shows a temperature-induced deformation. This reduces the influence of the spherical shell effect drastically, because even at moderate excitation energies the nucleus prefers the deformed shapes that are not shell stabilized. Collective enhancement of level densities may help to explain why spherical nuclei like $^{218}$U$_{126}$ or $^{298}$I$_{144}$ were not made until now.

The following conclusions for heavy-element production may be drawn from the studies on the limitations of EVR formation.

1. The entrance channel limitation by dissipative losses, as introduced by Swiatecki (15), hinders EVR formation at values of $\Delta E_{\text{ex}}/E > 0.70$. Further experiments indicate that there is no well-defined extra-push energy, but large fluctuations of the dissipative losses go together with increasing extra-push energies. The fusion probability at the Bass barrier $p(B_q)$ is proposed to be a more adequate ordering parameter than the extra-push energy for highly hindered systems.

2. Below and above the Bass barrier, nuclear structure effects modify the dissipative losses in EVR formation. Whether or not the same trends as found for subbarrier fusion are of importance when extra-push limitations are acting is an open question. Nuclear structure effects in EVR formation for the region of heavy elements could not until now be predicted.

3. Shell-stabilized deformed EVRs may be produced from compound systems with high excitation energies, as $\Gamma_n/\Gamma_f$ is only weakly dependent on the excitation energy.

4. Shell-stabilized spherical EVRs do not profit from shell stabilization in EVR formation unless their excitation energy is smaller than 15 MeV, a condition eventually to be fulfilled in the reaction $^{110}$Pd($^{110}$Pd, 2n)$^{210}$U, but never for reactions leading into the island of spherical superheavy nuclei.

5. To minimize losses in the evaporation cascade, the excitation energy should be kept as small as possible. The most symmetric combination of shell-stabilized collision partners, not yet limited by extra-push heating, should be chosen in order to fuse as coldly as possible. To produce elements 106 to 109, targets like $^{208}$Pb and $^{209}$Bi are bombarded by beams of $^{54}$Cr and $^{58}$Fe. This method of element production is called cold fusion in contrast to the hotter actinide-based reactions.

3. MODERN EQUIPMENT FOR ELEMENT SYNTHESIS

3.1 General Requirements Concerning the Techniques

To fuse two nuclei at the Coulomb barrier and thereby produce the heaviest elements, specific energies between 4.5 and 5.5 MeV/u must be available,
corresponding to energies of 80–350 MeV for projectiles between oxygen and nickel. Experimental equipment has to be designed in order to detect isotopes heavier than $Z = 104$ and $A = 250$, the decay modes of which are mainly $\alpha$ decay and fission. Electron capture (EC) branches are expected to be small, and detection of EC is desirable but not of great importance. Half-lives will be smaller than a few seconds. A lower limit on decay times cannot be foreseen. All isotopes are assumed to be made by fusion of heavy nuclei, that is, by a reaction with excitation functions as narrow as 10 MeV (10$^{18}$ atoms/cm$^2$). Production cross sections will be below $\sim 10$ nb, the cross sections found to produce isotopes of element 104. The cross-section limit should be pushed to as small a value as possible. The beam intensities should be as high as target technology allows. In thick Pb and Bi targets, about 3 kW are released by beam intensities of $5 \times 10^{13}$ ions/s. Pb and Bi targets of a few hundred $\mu$g/cm$^2$ at intensities of $4 \times 10^{12}$ ions/s will be heated by about 6 W. Thick targets have been cooled by forced liquid metal cooling at Dubna (65). Thin targets are cooled as rotating targets by black-body radiation cooling (66, 67) or by helium flow cooling at low pressures (68). The beam intensities given are upper limits reached with the most advanced ion sources at the Dubna U-400 or at the Darmstadt UNILAC.

In order to detect primary isotopes of the heaviest elements the separation method should be faster than seconds. In case of the detection of long-lived members of $\alpha$-decay chains, the separation time can be adjusted to the half-lives, which may be as long as days. Chemistry, on-line mass separators, and cluster-loaded He-jet transport systems have separation times of the order of seconds and will be useful only if isotopes in this time range are to be detected. Fast separation systems in the millisecond range are wheels (49, 65), systems in the microsecond range are recoil separators (69, 70).

The luminosity $L/$cm$^2$s = $N$/cm$^2$s, with $N$ the number of target atoms per cm$^2$ and $I$ the beam current in ions per second, is a quantity related to the reaction rate $A$ by $A \sim L\sigma$, where $\sigma$ is the production cross section. A luminosity of $10^{13}$/cm$^2$s is equivalent to about 1/pb$^{-1}$ d, that is a production rate of one atom per day at $\sigma = 1$ pb. Another important quantity of a separation method is its selectivity against primary beam particles, target recoil atoms, and transfer products. Rotating wheels and He jets do not discriminate radioactive transfer products at all. Recoil separators reduce transfer products by 2–3 orders of magnitude, whereas ISOL systems allow complete suppression of particles with deviating masses. Table 1 compares different methods either used or to be used for heavy-element production. The methods are presented in the following.

<table>
<thead>
<tr>
<th>Method</th>
<th>Rotating wheels</th>
<th>He-jet</th>
<th>Velocity selector</th>
<th>ISOL system</th>
<th>Gas-filled separator</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reference</td>
<td>(65)</td>
<td>(68)</td>
<td>(69)</td>
<td>(70)</td>
<td></td>
</tr>
<tr>
<td>Application</td>
<td>104–109</td>
<td>104–106</td>
<td>104–109 not used</td>
<td>102</td>
<td></td>
</tr>
<tr>
<td>Decay mode detected</td>
<td>fission</td>
<td>$\alpha$ chain or fission</td>
<td>EVR $\alpha$ chain fission</td>
<td>EVR $\alpha$ chain fission</td>
<td></td>
</tr>
<tr>
<td>Separation time (s)</td>
<td>$10^{-3}$</td>
<td>$10^{-1}$</td>
<td>$10^{-6}$</td>
<td>1</td>
<td>$10^{-6}$</td>
</tr>
<tr>
<td>Luminosity (pb$^{-1}$ d$^{-1}$)</td>
<td>5</td>
<td>0.4</td>
<td>0.2</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>Total efficiency</td>
<td>0.6</td>
<td>0.4</td>
<td>0.2</td>
<td>0.05</td>
<td>0.6</td>
</tr>
<tr>
<td>Event number detected at 1 pb in 10 days</td>
<td>30 (only fission)</td>
<td>2</td>
<td>0.4</td>
<td>0.05</td>
<td>1</td>
</tr>
<tr>
<td>Reduction of transfer products</td>
<td>none</td>
<td>none</td>
<td>$10^3$</td>
<td>excellent $10^3$</td>
<td></td>
</tr>
<tr>
<td>Single-event conclusive</td>
<td>no</td>
<td>no (fission)</td>
<td>yes ($\alpha$ chain)</td>
<td>yes</td>
<td>yes</td>
</tr>
</tbody>
</table>

### 3.2 Techniques

#### 3.2.1 ISOL SYSTEMS

ISOL systems until recently were never used in heavy-element synthesis experiments. They are included in Table 1 because they have some excellent features that in special cases may compensate for their slow separation time and low efficiency (only a few percent).

#### 3.2.2 HE-JET TECHNIQUES

As an example of an advanced He-jet facility stands the equipment used by Ghiorso and co-workers (4) for detection of $^{263}$Tb. The $^{18}$O beam of $3 \times 10^{17}$/s hits the He-cooled 0.8 mg/cm$^2$ thick $^{249}$Cf target mounted on a thin substrate. The recoils stopped in helium are
transported by a swift flow of helium seeded by a NaCl aerosol through a
5-m long capillary tube. The jet deposits its activity on a wheel, which is
moved after a preset collection time in front of surface barrier detectors.
These register either $\alpha$ decays or fission events coming from the parent or
daughter decays. A decaying parent atom sitting on the wheel either emits
its $\alpha$ particle toward the detector or is deposited itself as a recoil on the
detector surface. The $\alpha$ decay of the daughters is registered by the detector
with a high efficiency. A shuttle system allows one to move the detector in
front of a stationary second detector. The decay of a daughter atom
transferred as a recoil to the first detector can now be registered either by
this detector or by the second one. Three generations of $\alpha$ decays are linked
together by the wheel, the shuttling first detector system, and the fixed
second detector array. The system has an 80% transport efficiency, a
transport time of a second, but no discrimination against the copious
transfer products. Nevertheless, a cross section of 0.3 nb has been detected
for formation of the EVR $^{263}$106.

Because no correlation to the formation process of the atoms is possible,
fission events cannot be correlated. Fission half-lives can be measured by
varying the wheel transport time. With decreasing half-lives the restrictions
set by the long transportation times become more and more the decisive
handicap, e.g. the known isotopes of elements 107 to 109 could not have
been detected by any of the advanced He-jet systems installed at the various
laboratories.

3.2.3 WHEEL TECHNIQUES The wheel techniques were pioneered by the
Dubna group (49) and later applied by several other groups (6, 34). Their
advanced systems to study elements 104–109 are described here (see Figure
11) (65).

The target is a rotating cylinder made of copper, the circumference of
which is plated by a 3-mg/cm$^2$ layer of the target substance. The beam hits
the wheel tangentially; thus the target is equivalent to a thick target. EVRs
are stopped after having passed a layer of a few mg/cm$^2$ in beam direction.
Because fission products are emitted isotropically, there is a high proba-
bility of detecting at least one of them outside the target layer. Except for a
section of about 45° at the irradiation position, the wheel is viewed by mica
detectors, which allow for fission product detection. A 60% efficiency for
detection of a spontaneous fission event is achieved. The target wheel is
cooled and a beam power of 3 kW is safely handled over long irradiation
times. The heat generated in the copper wheel is transported via a thin
liquid metal layer to the standing water-cooled parts of the system. Beam
currents of $5 \times 10^{17}$/s have been used in irradiations with $^{55}$Mn ions. The
speed of revolution may be varied between 1 and $10^6$ cycles per min.

Shortest half-lives reported are a few milliseconds. Registration of fission
over periods of 100 h in specially selected micas is possible without any
disturbing background from spontaneously fissioning natural uranium
contaminants. Discrimination of fission products and scattered projectile
tracks is possible. The fact that the method can only detect fission is a most
serious disadvantage. This is compensated by the excellent luminosity,
which is larger by a factor of ten than that of other methods.

The thin layer of target material may be radiochemically treated after the
irradiation (72). Long-lived known daughter products of $\alpha$-decay chains
ending in elements Cm to Es have been separated and detected by their $\alpha$
energies. The detection of the heavy actinides is assumed to be related to
heavy-element formation. In the case of targets in the lead region these
isotopes are not produced as transfer products, nor in other decay channels
except for neutron evaporation. The latter is found to contribute predomin-
antly to EVR formation (51). This radiochemical technique is restricted to
the special case of reactions using targets in the lead region, and it is difficult
to transfer the method to actinide targets.

3.2.4 VELOCITY SEPARATORS The conservation of mass and momentum
in a fusion reaction demands that the fused product of mass $A = A_1 + A_2$
be emitted in beam direction with a well-defined velocity $v$, given by $v = A_1 v_1$/

![Figure 11](https://example.com/figure11.png)

*Figure 11* Wheel system developed by the Dubna group to synthesize isotopes by "cold fusion" (49, 71).*
$A_1 + A_2, A_1$ and $A_2$ are the masses of the projectile and the target; $v_1$ is the velocity of the projectile hitting the target nucleus at rest. Particles of velocity $v$ emitted in beam direction must be fusion products. A velocity filter selecting these particles is a kinematic mass separator. Since the projectiles and fusion products are both flying in the beam direction with different velocities, the separation problem is characterized by the suppression factor of primary projectiles in the velocity window of the fusion products. The ratio of projectiles to reaction products at the entrance to the filter is larger than $10^8$. Suppression factors of at least this order of magnitude have to be demanded. Radial and axial acceptance angles of $3^\circ$ guarantee a transport through the velocity filter of 20% for EVRs produced near the Coulomb barrier.

Different versions of velocity filters are discussed and reviewed in (73) in connection with the problem of isotope separation. Several filters are operating but only one system, the Separator for Heavy Ion Reaction Products (SHIP), is used to search for new elements. It has been operating since 1976 at the UNILAC (69, 74). A collaboration between II. Physikalisches Institut, Giessen, and GSI, Darmstadt—relying on common experiences gained during the construction and operation of the fission product separator LOHENSIN, Grenoble (75)—completed the SHIP in time to receive the first beams available from the UNILAC (76). Figure 12 shows a sketch of the filter.

The combination of two electric and four magnetic dipole fields, together with two quadrupole triplets, accepts particles radially and axially to $3^\circ$ and focuses all ionic charges within an ionic charge window of 20% (69, 77). The velocity dispersion necessary to separate projectiles and EVRs is maximum in the medium plane of the system. Here the beams are separated. The velocity dispersion is compensated in the second half of the filter, which ion-optically is antisymmetric to the first half. Suppression of the primary beam depends on the velocity difference between projectiles and EVRs. Ar, Kr, and Xe beams producing $A = 180$ EVRs have been suppressed by factors of $10^{12}, 10^{10}$, and $10^8$, respectively. The number of background particles from the primary beam is small enough to use detector systems directly at the focus position without any further stages of beam purification. A time-of-flight (TOF) system behind SHIP allows a redundant velocity measurement. Implantation of the unslowed EVRs with their full energy (30-300 MeV) into surface barrier detectors allows an energy measurement. Together with the TOF measurement, a rough value of the mass can be obtained ($A/\Delta A \approx 10$). These detector systems act as additional stages of beam purification with possible suppression factors up to $10^6$. In subbarrier fusion studies, cross sections were followed over six orders of magnitude and fusion cross sections in the microbarn region could still be measured (30).

**Figure 12** The velocity filter SHIP (69).

SHIP separates the reaction products spatially, and allows the detection of nuclei produced with sub-microbarn cross sections by exploiting their radioactive decay properties. Thus, it becomes not only a tool of reaction studies as magnetic spectrometers, but also a powerful on-line isotope separator (78). The subsequent $\alpha$ and spontaneous fission decays of the nuclei implanted are correlated among each other and to the signals obtained at the time of implantation into the surface barrier detector (79). Half-lives are determined from a few events by the maximum likelihood method. As an example, we give in Figure 13 the correlation time distribution of the $\alpha$ decay of $^{242}$Fm ($E_\alpha = 8.55$ MeV, $T_{1/2} = 0.18$ s) (80). As the recoil energy from $\alpha$ decay is small, an implanted nucleus does not

**Figure 13** The maximum likelihood method applied to the recoil $\alpha$ correlation observed in the reaction $^{208}$Pb($^{86}$Ar,3n)$^{242}$Fm in order to determine the half-life of $^{242}$Fm (79, 80).
change position. Position-sensitive detectors allow another large reduction of accidental correlations (81). The detection system used at SHIP is shown in Figure 14. An array of seven position-sensitive surface barrier detectors cover an area of 16 cm\(^2\). The cooled detectors allow for an energy resolution of 25 keV FWHM and position resolution of 0.2 mm FWHM, thus dividing each detector into 100 detector cells.

![Diagram showing the detector system behind SHIP allowing TOF energy measurements of the recoils and the registration of position, time, and decay energy for correlated members of a decay chain (24, 81). In the upper left corner the TOF-energy measuring system to determine the mass of the implanted recoil is shown. The Si detector in the right corner shows an implanted EVR and its decay by three a particles in three subsequent generations. The lower part shows a decay chain with the quantities defining the event.](image)

The detection of single decay chains was made possible by correlated event analysis. The error probability for a chain of correlated events is given as (82)

\[ P_{err} = N_0 \prod_{i=1}^{k} \lambda_i \Delta t_{0i} \]

with \( N_0 \) the number of implanted leading event nuclei during the measuring time, \( \lambda_i \) the counting rate in the energy window for the generation \( i \) in the decay chain, and \( \Delta t_{0i} \) the actually observed time difference between implantation and the \( i \)th decay. \( P_{err} \) is the probability that all \( k \) decay signals with their observed energy and time characteristics follow randomly and in any order the same implanted leading event nucleus. For chains with \( k \) generations the number of position cells of the position-sensitive detector \( F \) enters as \( F^{-k} \) into \( P_{err} \), e.g., for a three-member chain the SHIP detector reduces \( P_{err} \) by a factor of \( 10^{-6} \).

3.2.5 GAS-FILLED SEPARATORS Gas-filled separators have never been used for detection of elements beyond 102. But their high potentialities certainly make them a promising instrument in the future. They have been operated as fission product separators (83, 84), as well as fusion EVR separators (85). Facilities in operation now are the Juclsh On-line Separator for Fission products, JOSEP (86), and the Small Angle Separating System, SASSY (70). SASSY is a prototype instrument for separation of EVRs and all numbers given in the following refer to this instrument. It has a low atomic number resolution, but a high transport efficiency.

The deflection in a magnetic field depends on the average charge of the ion during its passage through the magnetic field:

\[ Bp/Tm = 0.027(v/u_0)A/\bar{q} \]

with \( v_0 \) the Bohr velocity \( ac \). The ratio \( v/\bar{q} \) only weakly depends on the velocity, and \( Bp/\bar{q} \) becomes a function depending mainly on the atomic number \( Z \). A large velocity window and the full ionic charge distribution can be accepted, which gives rise to the high efficiency of the method. The deflection in a gas-filled field, the gas providing the averaging over all possible ionic charges, depends on the mass and atomic number. The resolution is governed by two properties of the ion, the dispersion between neighboring atomic numbers and the fluctuations of the average ionic charges. A gas-filled separator has to be calibrated either with well-defined beams of heavy ions or with reaction products, which are known and can be detected by their radioactivity. For two velocity ranges, \( v/v_0 = 2.2 \) and \( v/v_0 = 4 \), the \( Bp/\bar{q} \) values for different ions in helium have been determined (Figure 15). The dependence is described by a \( Z^{-1/3} \) dependence, super-
imposed on an oscillating function determined by the electronic shell structure of the ions. These oscillations are most pronounced at small velocity. They govern the resolving power. At atomic numbers for which $Bp/A$ decreases with $Z$, $Z = 70 - 90$, the difference in $Bp$ for neighboring elements is small and separation is difficult, whereas for increasing $Bp/A$, $Z = 90 - 100$, the dispersion between elements is large and separation can be achieved.

Figure 16 shows the principal set-up of SASSY. The system is more compact (4 m) than SHIP and this allows still shorter separation times. The suppression of full energy projectiles in the $^{208}\text{Pb}(^{48}\text{Ca}, 2n)^{234}\text{U}$ reaction was found to be $10^{-15}$. The suppression of target recoils and target-like $\alpha$ emitters is $10^{-3}$. The transmission through the system was measured to be about 40%. A position-sensitive detector of $5 \times 2 \text{ cm}^2$ has registered 15% of all $\alpha$-decaying $^{234}\text{U}$ nuclei produced in the target. Use of large area detectors can achieve total efficiencies of more than 50%. Once instrument designers overcome the barrier of mixing dirty atomic physics with clean ion optics, gas-filled separators may become serious competitors to velocity filters as cheap, fast, and efficient separating devices.

4. PRODUCTION OF ELEMENTS 106–109 BY COLD FUSION REACTIONS

Only one experiment has successfully synthesized isotopes of elements beyond $Z = 105$ starting from actinide targets: the experiment that discovered $^{262}\text{Fm}$ (4). It was described in Sections 1 and 3.2.2. Until recently, all further attempts to synthesize other isotopes of $Z = 106$ or to reach higher elements failed. EVRs from fusion reactions, the cross sections of which are small compared to multinucleon transfer cross sections, could not be detected unambiguously in the large background of similar activities from transfer reactions. Moreover, all searches to synthesize elements around the predicted doubly magic nucleus $^{298}\text{Fl}$ failed (87). These search experiments are not the primary subject of this article, in spite of their interesting fall-out on different reaction aspects. The reactions leading beyond $Z = 106$ all make use of “cold fusion,” a method introduced by Oganessian and co-workers (49): Targets around Pb are bombarded by beams of Cr, Mn, and Fe with energies near the Coulomb barrier and isotopes in the chains $N-Z = 46-49$ are produced by In and 2n channels.

In order to establish the genetics of the isotopes in question, the daughter isotopes of elements $Z < 106$ had to be known. The isotopes of elements $Z < 102$ are well known and their spectroscopic properties are compiled in tables (88). Most of what we know about the isotopes of elements 104 and 105 comes from cold fusion studies and is presented in Table 2.
### Table 2 Compilation of data on isotopes of elements 104–109 obtained from cold fusion reactions

#### Element 104

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$ (s)</th>
<th>$E_0$ (MeV)</th>
<th>$b_0$</th>
<th>Reaction</th>
<th>$E_2^\gamma$ (MeV)</th>
<th>$E_1$ (MeV)</th>
<th>Dose $(10^{17})$</th>
<th>$\sigma$ (nb)</th>
<th>Comment</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>258</td>
<td>—</td>
<td>901(0.18)</td>
<td>898(0.29)</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},\gamma$</td>
<td>24.0</td>
<td>272</td>
<td>8.0</td>
<td>0.6</td>
<td>detection of $^{246}\text{Cf}$</td>
<td>(51)</td>
</tr>
<tr>
<td></td>
<td>990(0.06)</td>
<td>8.90(0.06)</td>
<td>8.70(0.06)</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},\nu$</td>
<td>24.0</td>
<td>233.5</td>
<td>—</td>
<td>6.1 ± 1.8</td>
<td>decay chain (a)</td>
<td>(50, 91)</td>
</tr>
<tr>
<td></td>
<td>8.60(0.12)</td>
<td>8.60(0.12)</td>
<td>—</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},\nu$</td>
<td>24.0</td>
<td>272</td>
<td>8.0</td>
<td>5.0</td>
<td>detection of $^{233}\text{Fm}$</td>
<td>(51)</td>
</tr>
<tr>
<td>256</td>
<td>5 ms</td>
<td>—</td>
<td>240.0</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},2n$</td>
<td>24.0</td>
<td>260</td>
<td>0.1</td>
<td>6</td>
<td>detection of 232$^{104}$ (a)</td>
<td>(90)</td>
</tr>
<tr>
<td></td>
<td>7.4 ± 0.2 ms</td>
<td>8.81</td>
<td>0.02</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},\nu$</td>
<td>24.1</td>
<td>239</td>
<td>—</td>
<td>6.0 ± 0.2</td>
<td>decay chain (a)</td>
<td>(50, 91)</td>
</tr>
<tr>
<td></td>
<td>6.7 ± 0.2 ms</td>
<td>6.70(0.09)</td>
<td>0.01</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},\nu$</td>
<td>28.0</td>
<td>271</td>
<td>8.0</td>
<td>6.0</td>
<td>detection of 232$^{104}$ (a)</td>
<td>(51)</td>
</tr>
<tr>
<td></td>
<td>1.2 ± 0.2 ms</td>
<td>1.20(0.09)</td>
<td>—</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},2n$</td>
<td>24.0</td>
<td>251</td>
<td>—</td>
<td>4.8 ± 0.2</td>
<td>decay chain (a)</td>
<td>(50, 91)</td>
</tr>
<tr>
<td></td>
<td>1.7 ± 0.2 ms</td>
<td>1.70(0.09)</td>
<td>—</td>
<td>$^{208}\text{Pb}^{(90}\text{Tl},2n$</td>
<td>28.0</td>
<td>271</td>
<td>8.0</td>
<td>4.0 ± 0.3</td>
<td>decay chain (a)</td>
<td>(50, 91)</td>
</tr>
</tbody>
</table>

#### Element 105

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$ (s)</th>
<th>$E_0$ (MeV)</th>
<th>$b_0$</th>
<th>Reaction</th>
<th>$E_2^\gamma$ (MeV)</th>
<th>$E_1$ (MeV)</th>
<th>Dose $(10^{17})$</th>
<th>$\sigma$ (nb)</th>
<th>Comment</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>258</td>
<td>9.30(0.08)</td>
<td>9.17(0.15)</td>
<td>—</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},\nu$</td>
<td>24.1</td>
<td>234</td>
<td>—</td>
<td>2.9 ± 0.3</td>
<td>EC-branch 33% to 232$^{104}$</td>
<td>(24, 50)</td>
</tr>
<tr>
<td></td>
<td>9.05(0.05)</td>
<td>9.05(0.05)</td>
<td>—</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},\nu$</td>
<td>24.1</td>
<td>265</td>
<td>2.6</td>
<td>2.7</td>
<td>Detection of 232$^{104}$ and $^{244}\text{Cr}$</td>
<td>(51)</td>
</tr>
<tr>
<td>[257]</td>
<td>5.0 ± 1.0 ms</td>
<td>0.8</td>
<td>1.58</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},\nu$</td>
<td>24.1</td>
<td>270</td>
<td>0.95</td>
<td>0.83</td>
<td>$\sigma$ refers to sf only, mass and element number revised in (51)</td>
<td>(92)</td>
</tr>
<tr>
<td>257</td>
<td>1.4 ± 0.4 ms</td>
<td>9.16(0.3)</td>
<td>9.07(0.3)</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},2n$</td>
<td>24.1</td>
<td>244</td>
<td>—</td>
<td>2.1 ± 0.8</td>
<td>decay chain (a, sf)</td>
<td>(24, 50)</td>
</tr>
<tr>
<td></td>
<td>8.97(0.4)</td>
<td>—</td>
<td>8.97(0.4)</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},2n$</td>
<td>24.0</td>
<td>310</td>
<td>2.3</td>
<td>0.03</td>
<td>sf from 257$^{105}$ after a decay of 261$^{107}$</td>
<td>(65)</td>
</tr>
<tr>
<td>[256]</td>
<td>1.2 ± 0.4 ms</td>
<td>—</td>
<td>207$^{99}\text{Mn},\nu$</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},2n$</td>
<td>24.0</td>
<td>310</td>
<td>23</td>
<td>0.3</td>
<td>$\sigma$ refers to sf only, mass and element number revised in (65)</td>
<td>(92)</td>
</tr>
<tr>
<td>258</td>
<td>2.6 ± 0.4 ms</td>
<td>—</td>
<td>—</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},2n$</td>
<td>24.1</td>
<td>310</td>
<td>55</td>
<td>~0.02</td>
<td>sf from 258$^{105}$ after a decay of 261$^{107}$</td>
<td>(65)</td>
</tr>
<tr>
<td>258</td>
<td>2.6 ± 0.4 ms</td>
<td>—</td>
<td>206$^{99}\text{Mn},\nu$</td>
<td>$^{209}\text{Bi}^{(90}\text{Tl},2n$</td>
<td>24.1</td>
<td>310</td>
<td>55</td>
<td>~0.02</td>
<td>sf from 258$^{105}$ after a decay of 261$^{107}$</td>
<td>(65)</td>
</tr>
<tr>
<td>[255]</td>
<td>1.2 ± 0.4 ms</td>
<td>0.9</td>
<td>209$^{99}\text{Bi}^{(90}\text{Tl},2n$</td>
<td>27.9</td>
<td>270</td>
<td>4</td>
<td>—</td>
<td>EC-branch 10% to 234$^{104}$</td>
<td>(65)</td>
<td></td>
</tr>
<tr>
<td>255</td>
<td>&gt; 0.5</td>
<td>209$^{99}\text{Bi}^{(90}\text{Tl},2n$</td>
<td>27.9</td>
<td>270</td>
<td>4</td>
<td>—</td>
<td>detection of 255$^{105}$ (a)</td>
<td>(65)</td>
<td></td>
<td></td>
</tr>
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Table 2 (continued)

Element 106

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$ (ms)</th>
<th>$E_i$ (MeV)</th>
<th>$b_s$</th>
<th>Reaction</th>
<th>$E_i^o$ (MeV)</th>
<th>$E_i$ (MeV)</th>
<th>Dose ($10^{17}$)</th>
<th>$\sigma$ (nb)</th>
<th>Comment</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>263</td>
<td>$0.9 \pm 0.2$</td>
<td>9.25</td>
<td>1.0</td>
<td>$^{269}$Cr($^{16}$O, 4n)</td>
<td>40.2</td>
<td>95</td>
<td>13.4</td>
<td>0.3</td>
<td>decay chain (a)</td>
<td>(4)</td>
</tr>
<tr>
<td>261</td>
<td>$0.26^{+0.16}_{-0.19}$</td>
<td>9.56(0.60)</td>
<td>&gt;0.9</td>
<td>$^{208}$Pb($^{14}$C, n)</td>
<td>22.8</td>
<td>264</td>
<td>1.6</td>
<td>0.5</td>
<td>decay chain (a)</td>
<td>(97)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>9.51(0.27)</td>
<td></td>
<td></td>
<td>22.8</td>
<td>300</td>
<td>0.3</td>
<td>0.3</td>
<td>detection of $^{233}$Es (a)</td>
<td>(98)</td>
</tr>
<tr>
<td>260</td>
<td>2.5 ± 1.5</td>
<td>—</td>
<td>&gt;0.8</td>
<td>$^{207}$Pb($^{14}$C, n)</td>
<td>22.4</td>
<td>290</td>
<td>—</td>
<td>0.3</td>
<td>detection of $^{235}104$ (af)</td>
<td>(94)</td>
</tr>
<tr>
<td></td>
<td>3.6 ± 0.9</td>
<td>9.76</td>
<td>0.5</td>
<td>$^{208}$Pb($^{14}$C, 2n)</td>
<td>22.8</td>
<td>290</td>
<td>2.3</td>
<td>0.4</td>
<td>decay chain (a, sf)</td>
<td>(95)</td>
</tr>
<tr>
<td>[259]</td>
<td>4–10</td>
<td>—</td>
<td>&gt;0.2</td>
<td>$^{208}$Pb($^{14}$C, 2n)</td>
<td>22.8</td>
<td>300</td>
<td>16</td>
<td>0.4</td>
<td>detection of $^{235}104$ (af)</td>
<td>(98)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$^{207}$Pb($^{14}$C, [2n])</td>
<td>22.4</td>
<td>280</td>
<td>0.3</td>
<td>1.0</td>
<td>mass and element</td>
<td>(93)</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$^{208}$Pb($^{14}$C, [3n])</td>
<td>22.8</td>
<td>280</td>
<td>0.2</td>
<td>1.0</td>
<td>number revised in (94)</td>
<td>(94)</td>
</tr>
<tr>
<td>259</td>
<td>—</td>
<td>—</td>
<td>~1.0</td>
<td>$^{206}$Pb($^{14}$C, n)</td>
<td>23.1</td>
<td>290</td>
<td>—</td>
<td>0.4</td>
<td>detection of $^{235}104$ (af)</td>
<td>(94)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$^{207}$Pb($^{14}$C, 2n)</td>
<td>22.4</td>
<td>290</td>
<td>—</td>
<td>0.4</td>
<td>detection of $^{235}104$ (af)</td>
<td>(94)</td>
</tr>
<tr>
<td>0.48 ± 0.51</td>
<td>9.63</td>
<td>&gt;0.8</td>
<td>$^{207}$Pb($^{14}$C, 2n)</td>
<td>22.4</td>
<td>266</td>
<td>1.4</td>
<td>0.3</td>
<td>decay chain (a)</td>
<td>(95)</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td>$^{208}$Pb($^{14}$C, [2n])</td>
<td>22.8</td>
<td>300</td>
<td>5</td>
<td>0.02</td>
<td>detection of $^{235}104$ (af)</td>
<td>(98)</td>
</tr>
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</table>

Element 107

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$ (ms)</th>
<th>$E_i$ (MeV)</th>
<th>$b_s$</th>
<th>Reaction</th>
<th>$E_i^o$ (MeV)</th>
<th>$E_i$ (MeV)</th>
<th>Dose ($10^{17}$)</th>
<th>$\sigma$ (nb)</th>
<th>Comment</th>
<th>Ref.</th>
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<tr>
<td>262</td>
<td>4.72 ± 0.3</td>
<td>10.38</td>
<td>1.0</td>
<td>$^{209}$Bi($^{14}$C, n)</td>
<td>22.3</td>
<td>262</td>
<td>1.2</td>
<td>0.2</td>
<td>decay chain (a)</td>
<td>(96)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$^{209}$Bi($^{14}$C, 2n)</td>
<td>22.3</td>
<td>290</td>
<td>5</td>
<td>0.2</td>
<td>detection of $^{235}104$ (af) and $^{244}$Cr (a)</td>
<td>(51, 65)</td>
</tr>
<tr>
<td>[261]</td>
<td>1–2</td>
<td>—</td>
<td>[0.8]</td>
<td>$^{209}$Bi($^{14}$C, [2n])</td>
<td>22.3</td>
<td>290</td>
<td>4.6</td>
<td>0.1</td>
<td>analysis modified later (65)</td>
<td>(92)</td>
</tr>
<tr>
<td>261</td>
<td></td>
<td>—</td>
<td>0.2</td>
<td>$^{209}$Bi($^{14}$C, [2n])</td>
<td>24.0</td>
<td>310</td>
<td>2.3</td>
<td>0.05</td>
<td>detection of $^{235}105$ (af)</td>
<td>(65)</td>
</tr>
<tr>
<td>260</td>
<td>—</td>
<td>—</td>
<td>1.0</td>
<td>$^{206}$Pb($^{14}$C, mn)</td>
<td>24.1</td>
<td>310</td>
<td>55</td>
<td>~0.02</td>
<td>detection of $^{236}104$ (af)</td>
<td>(65)</td>
</tr>
</tbody>
</table>

Element 108

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$ (ms)</th>
<th>$E_i$ (MeV)</th>
<th>$b_s$</th>
<th>Reaction</th>
<th>$E_i^o$ (MeV)</th>
<th>$E_i$ (MeV)</th>
<th>Dose ($10^{17}$)</th>
<th>$\sigma$ (nb)</th>
<th>Comment</th>
<th>Ref.</th>
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<tbody>
<tr>
<td>265</td>
<td>1.8±0.3</td>
<td>10.36</td>
<td>1.0</td>
<td>$^{208}$Pb($^{14}$Fe, n)</td>
<td>20.4</td>
<td>292</td>
<td>22</td>
<td>0.015±0.013</td>
<td>decay chain (a)</td>
<td>(97)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$^{208}$Pb($^{14}$Fe, 2n)</td>
<td>20.4</td>
<td>320</td>
<td>30</td>
<td>0.004</td>
<td>detection of $^{233}$Es (a)</td>
<td>(98)</td>
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<tr>
<td>264</td>
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<td>—</td>
<td>1.0</td>
<td>$^{207}$Pb($^{14}$Fe, 2n)</td>
<td>20.2</td>
<td>320</td>
<td>22</td>
<td>0.005</td>
<td>detection of $^{235}104$ (af)</td>
<td>(98)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$^{208}$Pb($^{14}$Fe, 2n)</td>
<td>20.4</td>
<td>320</td>
<td>32</td>
<td>0.002</td>
<td>detection of $^{235}104$ (af)</td>
<td>(98)</td>
</tr>
<tr>
<td>264</td>
<td>—</td>
<td>—</td>
<td>1.0</td>
<td>$^{209}$Bi($^{14}$Mn, n)</td>
<td>25.8</td>
<td>300</td>
<td>130</td>
<td>0.002</td>
<td>detection of $^{235}104$ (af)</td>
<td>(65, 98)</td>
</tr>
</tbody>
</table>

Element 109

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$T_{1/2}$ (ms)</th>
<th>$E_i$ (MeV)</th>
<th>$b_s$</th>
<th>Reaction</th>
<th>$E_i^o$ (MeV)</th>
<th>$E_i$ (MeV)</th>
<th>Dose ($10^{17}$)</th>
<th>$\sigma$ (nb)</th>
<th>Comment</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>266</td>
<td>3.57±0.4</td>
<td>11.10</td>
<td>1.0</td>
<td>$^{209}$Bi($^{14}$Fe, n)</td>
<td>19.8</td>
<td>299</td>
<td>2.8</td>
<td>0.015±0.013</td>
<td>decay chain (a, sf)</td>
<td>(99, 100)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$^{209}$Bi($^{14}$Fe, 2n)</td>
<td>19.8</td>
<td>320</td>
<td>36</td>
<td>0.003</td>
<td>detection of $^{235}104$ (af) and $^{244}$Cr (a)</td>
<td>(51)</td>
</tr>
</tbody>
</table>
258,257,104 were known from earlier studies (89). The isotopes 254,253,103 are \( \alpha \) emitters and were found in the reaction \( ^{209}\text{Bi}(^{50}\text{Ti},xn)^{259-210}\text{Bi} \) as correlated daughter decays of the \( Z = 105 \) isotopes (50). The half-life of 254,103 is 13.5 \( \pm \) 3 s, it emits \( \alpha \) particles of 8.46 MeV (64%), and 8.41 MeV (36%). 253,103 has a half-life of 1.3 \( \pm \) 0.3 s and emits \( \alpha \) particles of 8.80 MeV (56%) and 8.72 MeV (44%). In Table 2 we give the main references on the isotopes of elements \( Z = 104 \) and \( Z = 105 \).

At Dubna the spontaneous fission activities of the isotopes 256-253,104 were investigated (51, 90), whereas at Darmstadt \( \alpha \) decay and spontaneous fission activities of 256,255,104 were established (24, 50, 91). Spontaneous fission activities were assigned to 257,255,105 at Dubna (65, 92), \( \alpha \) decay of 258,257,105 was found at Darmstadt (24, 50). The production of the isotopes 256,255,105 was established from the occurrence of spontaneous fission (65). The isotopes 258,257,105 are assumed to have an electron capture branch leading to 258,257,104, which gives rise to fission activities with the half-lives of 258,257,105 (50, 65). Delayed fission found after \( K \alpha \) rays from the EC decay of 258,105 corroborates this interpretation directly (24). As Table 2 shows, there are no conflicting results from the different experiments, neither for the spectroscopic nor for the reaction data for element \( Z = 104 \) and \( Z = 105 \) isotopes. The inclusion of 1n reaction channels first found in the \( ^{208}\text{Bi}(^{50}\text{Ti},1n)^{257}104 \) reaction at Darmstadt (50) resolved all earlier discrepancies.

4.1 The Isotopes of Element 106

Three isotopes, 261-259,106, were produced by \( ^{54}\text{Cr} \) bombardments of 208-206Pb targets (Table 2). Experiments were performed in 1974 at Dubna (93). A 4-10-ms fission activity observed in 208,207Pb bombardments was then assigned to 259,106, an assignment that could not be confirmed later (94, 95). All irradiations performed since then are compiled in Table 2. Demin and co-workers (94), analyzing the spontaneous fission activities found in \( ^{54}\text{Cr} \) irradiations of 208-206Pb, concluded that the activities seen come from the isotopes 256,255,104 and not from element 106, the isotopes of which are assumed to be \( \alpha \) emitters. A value for the 260,106 half-life was given for an activity build-up analysis, and lower limits for the spontaneous fission half-lives of 261,259,106 were inferred. The results presented have been corroborated by experiments at Darmstadt (95, 97).

In an irradiation of 208Pb with \( ^{54}\text{Cr} \) at 4.85 and 4.92 MeV/u, events correlated to the implanted nucleus with an \( \alpha \) energy of 9.56 MeV were found. In the second generation of decays these events were correlated to the isotope 257,104, known to have an \( \alpha \) spectrum of several lines, the energies of which were found in the decay chains. It was established (97) that the isotope 261,106 is an \( \alpha \) emitter, \( E_{\alpha} = 9.56 + 0.03 \) MeV (60%), with a half-life of 0.26 \( \pm \) 0.11 s, in agreement with the conclusions in (94). Its formation cross section in the reaction \( ^{208}\text{Pb}(^{54}\text{Cr},n)^{261}106 \) is about 0.5 nb.

The isotope 260,106 has been produced at 4.92 MeV/u in the reaction \( ^{208}\text{Pb}(^{54}\text{Cr},2n)^{260}106 \) (95). It decays by the emission of an \( \alpha \) particle of 9.76 MeV into 256,104, which then disintegrates by fission (see Figure 21, Section 5.1). Besides the \( \alpha \) branch, a 50% spontaneous fission branch of 260,106 has been detected. The half-life of 260,106 is 3.6 \( \pm \) 0.2 ms, giving a spontaneous fission half-life of about 7 ms. This value is about equal to the half-life of the daughter isotope reached by \( \alpha \) decay. The spontaneous fission half-life going from 256,104 to 260,106 does not decrease, a finding again in agreement with the analysis of (94).

In the \( ^{207}\text{Pb}(^{54}\text{Cr},2n)^{259}106 \) reaction, 259,106 was found to have a half-life of 0.48 \( \pm \) 0.13 s, and to decay by \( \alpha \) emission with a main \( \alpha \) energy of 9.63 \( \pm \) 0.03 MeV. The spontaneous fission activity assigned to this isotope by the Dubna group in 1974 stems from 260,106 and from 256,104, but not from 259,106. In the reactions used in 1974, isotopes of element 106 were produced, but the isotope upon which the claim of element discovery was built was clearly assigned incorrectly.

4.2 The Isotopes of Element 107

Initial experiments designed to synthesize element 107 via the reactions \( ^{209}\text{Bi}(^{54}\text{Cr},xn)^{209}107 \) and \( ^{208}\text{Pb}(^{55}\text{Mn},xn)^{208}107 \) have been reported by the Dubna group (92). Two spontaneous fission activities detected were assigned to 261,107 (\( b_{\alpha} = 0.8 \)) and its daughter 257,105 (\( b_{\alpha} \approx 0.8 \)). Correlated event chains found in the reaction \( ^{209}\text{Bi}(^{54}\text{Cr},n)^{257}105 \) at Darmstadt (50, 96) showed that the spontaneous fission activity assigned in 1976 at Dubna to 257,105 is mainly due to 258,104, which fissions after EC decay of 258,105 (33%). In the reaction the isotope 262,107 unambiguously was established by seven correlated chains, one of which is given in Figure 17 (96). Both the \( \alpha \) decay and the fission branch in the 262,107 chain were confirmed by recent Dubna experiments (51, 65) showing the EC-delayed fission activity of 258,104 as well as the \( \alpha \) decay of 246,107, a late member of the chain (Figure 18). 262,107 is the one isotope of element 107 established in both laboratories independently and unambiguously.

The 1-2-ms activity assigned to 261,107 (92) was not confirmed directly, but it was argued that the spontaneous fission yield detected in 207Pb(55Mn, xn) was compatible with an 80% fission branch of 261,107 necessary to explain the fission yield found for 257,105 (51). A millisecond activity for 261,107 would point to a sharp disappearance of shell stabilization for this isotope or to a sudden loss of the additional stabilization of odd isotopes against spontaneous fission, which has been confirmed for all isotopes of elements \( Z = 104 \) to \( Z = 106 \) investigated.
From the systematics a fission half-life of about 0.1 s should be expected and 261.107 should be an α emitter in the millisecond range. A direct observation of 261.107 by correlated event analysis should be feasible and is highly desirable, as the claim of discovery of element 107 by the Dubna group is built upon the existence of a strong millisecond-fission branch of 261.107. This branch was not found; instead, α decay was observed in a recent experiment at Darmstadt (G. Münzenberg, private communication).

A third isotope, 260.107, was detected at Dubna in the reaction 206Pb(255Mn, n) by the EC-delayed fission activity of 256.104 (65). The isotope should be an α emitter as is 262.107. The production cross section was found to be a factor of 10 smaller than for 262.107. A confirmation by decay chain analysis is still lacking.

4.3 The Isotopes of Element 108

The synthesis of the even elements Z = 106 and Z = 108 was believed to be more difficult than the finding of the odd ones, Z = 107 and Z = 109, because their stability against spontaneous fission was expected to be reduced compared to the lifetimes of odd-element isotopes. Very small spontaneous fission half-lives were expected to range from 10^{-4} to 10^{-2} s. The finding of α decay for 264.109 (99) and the analysis of spontaneous

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**Figure 17** Sequence of correlated α decays following the implantation of 262.107 into a position-sensitive Si detector (96).

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**Figure 18** The α-particle spectrum for the Cf fractions obtained by bombarding a 206Bi target with 58Fe, 54Cr, and 50Ti ions (51). The arrows indicate energies and probabilities of the α transitions of nuclei present in the sample. The 3.18-MeV and 3.97-MeV peaks, as well as the complex spectrum having a maximum at 5.81 MeV, are from the tracer activities 146Cd, 149Yb, and 248Cf. The 6.75-MeV α particles are from 244Cf, a late member of the N−Z = 48 decay chain starting at 264.109, 262.107, and 258.105, respectively.
fission rates obtained in $^{54}$Cr bombardments of the isotopes $^{206-208}$Pb (94) led to the conclusion that the isotopes $^{259-261}$Pb might be $\alpha$ emitters and encouraged a search for isotopes of elements 106 and 108.

In the reaction $^{208}$Pb($^{54}$Fe, n) at a bombarding energy of 5.02 MeV/u, three correlated events were found; these were assigned to $^{265}$Pb (Figure 19) (97). The decay chains connect $^{264}$Pb with known isotopes, among which are found $^{257}$Tl (1.4 s) and $^{253}$Es (20 d), the latter being used by the Dubna group to prove formation of isotopes in the chain (98). The chains are correlated to $^{261}$Pb. The second-generation $\alpha$ energy seen in one event and the correlation times measured agree with the previous finding on this isotope. All chains are correlated in the third generation to the isotope $^{257}$Tl, which once more proves that the mother nucleus for these chains is $^{265}$Pb. $^{261}$Pb decays by $\alpha$ emission, $E_\alpha = 10.36 \pm 0.03$ MeV, with a half-life of 1.8$^{+2.7}_{-0.5}$ ms. In the three events no fission was observed in any generation. Up to element 108 in the sequence $N - Z = 49$ no fission branches have been found. The production cross section obtained is $19.2^{+15}_{-11}$ pb. The cross section reported by the Dubna group for the same reaction is considerably smaller, which points to either a favorable statistical fluctuation for the events at Darmstadt or to an overestimate of the detection probability in the Dubna measurements (98).

An 8-ms and a 6-ms fission activity were found in the reactions $^{208}$Pb($^{58}$Fe, x$n$) and $^{207}$Pb($^{56}$Fe, x$n$), respectively (98). This result was attributed to strong $\alpha$ branches of the isotopes $^{264}$Pb and $^{261}$Pb, which finally lead to $^{258}$Tl, the fission of which was observed. The even-even isotope $^{264}$Pb is concluded to be an $\alpha$ emitter with a negligible fission branch, because the cross section for $^{207}$Pb($^{58}$Fe, n) measured from the fission yield of $^{256}$Tl was as large as the cross section for the corresponding reaction leading to $^{265}$Pb using $^{208}$Pb targets. A direct observation of $^{264}$Pb $\alpha$ decay should be of high priority in order to obtain a mass excess value for an element $Z = 108$ isotope.

Another isotope of element $Z = 108$ with mass number 263 is assumed to exist (65, 98). In the reaction $^{209}$Bi($^{54}$Mn, n), a 1.1-s spontaneous fission activity was seen. This activity was assigned to $^{255}$I, the granddaughter of the $\alpha$-emitting isotope $^{265}$Pb. Finding the same pattern of fission yields for the $^{58}$Fe irradiations of Pb targets as for $^{54}$Cr irradiations makes an interpretation as proposed (98) and confirmed for the $Z = 106$ isotopes (94) very plausible also for the $Z = 108$ isotopes. A final confirmation of the Dubna analysis by decay chain analysis, as was done for the $Z = 106$ isotopes, is necessary, but even now the existing evidence makes a highly increased stability against spontaneous fission very probable not only for $^{265}$Pb but also for the isotopes $^{264,263}$Pb.

### 4.4 $^{266}$Pb—The Isotope with the Highest Mass and Atomic Number

In the synthesis of $^{266}$Pb, the experimental set-up used at Darmstadt was shown to be capable of detecting single event chains with a high degree of significance (96). This encouraged the attempt to synthesize element 109 using the fusion of $^{58}$Fe with $^{209}$Bi as a consequent next step. The isotopes reached with Bi targets and beams of $^{50}$Ti, $^{54}$Cr, and $^{58}$Fe via 1n channels are odd-odd isotopes. Their highly increased stability against spontaneous fission, about a factor $10^3$ in spontaneous fission half-life, makes them best candidates for $\alpha$-decaying isotopes. The $\alpha$ decays connect, via the $A = (4n + 2)$-decay chain, a sequence of odd-odd isotopes. As in previous studies the new isotopes $^{254}$Lr, $^{251}$I, and $^{262}$Pb had been found and their $\alpha$ energies measured; it was possible to identify a new member in the chain from one decay chain observed. From a simple logarithmic extrapolation of the production cross sections of the elements 104, 105, and 107, the cross section for element 109 was expected to be about one order of magnitude lower than that for element 107 (Figure 26, top).

In the course of 250 hours of irradiating $^{209}$Bi with $^{58}$Fe beams with specific energies of 4.95, 5.05, and 5.15 MeV/u, totalling a dose of $7 \times 10^{17}$ particles, one correlated decay chain (Figure 20) produced at the highest projectile energy could be observed (99, 100). The particles selected are all those leaving SHIP that have been mass-identified by TOF and energy measurements and that are followed at the same detector position ($\pm 0.4$ mm) by an $\alpha$ decay within 40 ms in the beam pulse, or within 14 s between beam pulses. Of the 529 events registered, there is one event with a mass value $264 \pm 13$ and followed by an $11.10 \pm 0.04$-MeV $\alpha$ particle that is
certainly an EVR. All other events can be attributed to $\alpha$ decays from transfer products in the mass range $A = 210$–216.

The event identified as EVR is followed by a further $\alpha$ decay within 22.3 ms. This $\alpha$ particle escaped from the detector; a rest energy of only 1.14 MeV was deposited in the detector. A $\gamma$ ray of 382.9 keV was found in coincidence. The correlation time found is compatible with the half-life determined for $^{262}$Tl. The next step observed in the decay chain is a spontaneous fission decay. An energy of 232 ± 10 MeV was registered with a time delay of 12.9 s. This is the only fission event found throughout the whole 250-hour irradiation. Via the 30% EC branch of $^{258}$Tl, the even-even isotope $^{258}$Tl is reached and it undergoes fission within ms. The two combined decays explain the virtually long correlation time of the fission event. Using Equation 8 and analyzing the background in the different decay steps, we find a probability of the event being random of $2 \times 10^{-18}$.

All pieces observed in the decay chain agree with an assignment of $^{266}$Tl as the mother isotope. All other decay channels were discussed thoroughly (100), and it was concluded from all possible deexcitation channels of the compound nucleus $^{267}$Tl that the most probable assignment of the observed decays was the isotope with mass 266 of the new element 109. The isotope $^{266}$Tl has a half-life between 2 and 20 ms; it decays by $\alpha$ emission with an energy of 11.10 ± 0.04 MeV. The $\alpha$ energy is within the predictions of different mass formulae (101, 102). The energy given might not correspond to the ground-state $Q$ value, as found for many other odd-odd isotopes. The actual $Q$ value may be higher by up to 0.5 MeV. The half-life is too long for the energy given, which suggests a hindrance that again is within the values known for other odd-odd isotopes. The isotope is produced via the $\gamma$ channel with an excitation energy in the range 20–26 MeV. The production cross section amounts to 3–50 pb.

The analysis of the one-event assignment by Münzenberg and coworkers (100) to prove the existence of element 109 is strongly supported by a Dubna experiment using the same reaction (51). At a dose 14 times larger than that at Darmstadt, wheel techniques combined with chemical separation (see Section 3.2.3) gave one fission event and seven $\alpha$ decays of $^{246}$Cf (Figure 18). The production cross section obtained in the experiment is five times smaller than at Darmstadt, a discrepancy observed for the synthesis of $^{265}$Tl as well. An increase of the Darmstadt detection probability by a factor of five to a value of 100% is impossible. A corresponding decrease of the detection probability for the Dubna experiments to a value of about 12% would be at variance with the value of 60% given in (65). The production of the isotopes $^{266}$Tl and $^{268}$Tl by two independent groups and methods makes the existence of elements 108 and 109 as assured as the existence of all the previous man-made elements.

4.5 The Naming of Elements

Not hiding a certain self-interest, we want to help settle existing controversies on the naming of elements (103, 104). As a rule we propose the following: Element synthesis becomes production of a given isotope, and a name should be accepted only if the experiment claiming the discovery is reproducible. An isotope is defined by its mass and atomic number, its fingerprints are its decay modes and its half-life. Decay modes of the heavy isotopes in question are electron capture and spontaneous fission, which sometimes are difficult to assign to a specific isotope, and $\alpha$ decay, which (with its decay energies measured with an accuracy of some parts in a thousand) gives a very reliable mode of assignment. The time correlation of subsequent decays is a further method of definite isotope identification. The proposed rule should be applied retrospectively for all elements discovered by isotope identification, that is elements 102–109. A joint commission of physicists and nuclear chemists urgently is needed to solve the longstanding controversies.

5. GENERAL TRENDS IN HEAVIEST ISOTOPES SYSTEMATICS

The measured values of $\alpha$ energies, $\alpha$-decay half-lives, and fission half-lives can be used to establish trends of these quantities up to element 109 and to extrapolate the trends to higher proton numbers. Comparison with theoretical predictions of mass excesses, shell corrections, and fission barriers becomes possible. Besides ground-state properties, production cross sections may be analyzed in order to understand the limitations of EVR formation.
5.1 Mass Excesses, Shell Correction Energies, and Fission Barriers

For the two even elements, $Z = 104$ and $Z = 106$, $\alpha$ energies of the even-even isotopes with $N - Z = 48$ have been determined. Figure 21 displays decay chains showing the $\alpha$ decay of $^{256}104$ (24, 91) and $^{260}106$ (95, 106). Only one chain was found for the $^{256}104$ $\alpha$ decay with $E_\alpha = 8.81 \pm 0.02$ MeV giving an $\alpha$ branch of smaller than $3\%$; the existence of this $\alpha$ branch was detected via $\alpha$ decay of $^{246}$Cm (27 d) at Dubna as well (98). The $\alpha$ energy of 9.76 MeV for $^{260}106$ is based on the detection of 11 events. The two $\alpha$ energies assuming ground-state to ground-state decay allow us to determine from the experimentally known mass excess of $^{253}$No (105) the mass excesses of $^{256}104$ and $^{260}106$. The mass excesses of $^{256}104$ and $^{260}106$ are $94.2 \pm 0.1$ and $106.6 \pm 0.1$ MeV, respectively (95, 106). These values are compared in Table 3 to different mass excess predictions (102, 107–110). Best agreement is obtained with the prediction of Liran & Zeldes (110); all other approaches fail to reproduce the experimental values by about 1 MeV, the nuclei being more bound by this amount than predicted.

Except for $^{254}108$, all isotopes of the $N - Z = 48$ sequence are known up to $^{260}109$. Taking for the odd elements the highest $\alpha$ energies observed, mass excesses were determined as for the even elements. For odd elements in case of transitions to excited states, the mass excesses obtained would be too large. Table 4 gives the new $\alpha$ energies, mass excesses, and shell correction energies, $\Delta E_{\text{shell}} = M_{\text{exp}} - M_{\text{macro}}$, for the $N - Z = 48$ isotopes beyond $Z = 102$. The macroscopic mass excesses are taken from the tables published by Møller & Nix (102). Figure 22 presents the shell correction energies for all $N - Z = 48$ isotopes between $Z = 91$ and 109 as given in Table 4 or taken from the latest Wapstra tables (105). There is a small systematic odd-even difference of about 0.2 MeV between odd and even elements all over the range of elements concerned. The new masses do not deviate more than the previously compiled ones. The shell correction increases up to $^{250}105$ and then stays about constant up to $^{266}109$ at a level of $-5.2$ MeV. The calculated shell corrections (102, 111) shown in Figure 22 (top) are systematically larger than the experimental ones, all nuclei are more stabilized than predicted. For the experimental shell correction of $^{276}114$, following the trend of the calculated shell corrections, a value of about $-5.5$ MeV is extrapolated. The shell correction going from $Z = 109$ to $Z = 114$ along $N - Z = 48$ stays constant or even becomes slightly more stabilizing. Figure 23 gives all known experimental shell corrections for

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**Table 3** Mass excess values for $^{256}104$ and $^{260}106$

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Ref. (107)</th>
<th>(108)</th>
<th>(109)</th>
<th>(110)</th>
<th>(102)</th>
<th>(106)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{260}106$</td>
<td>108.27</td>
<td>107.29</td>
<td>107.7</td>
<td>106.94</td>
<td>108.13</td>
<td>106.62±0.06</td>
</tr>
<tr>
<td>$^{256}104$</td>
<td>95.90</td>
<td>94.84</td>
<td>95.6</td>
<td>94.37</td>
<td>95.77</td>
<td>94.23±0.05</td>
</tr>
</tbody>
</table>

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**Figure 22**: For isotopes of the sequence $N - Z = 48$ from $Z = 92$–109 shell correction energies (top), $\alpha$ energies (middle), and fission barriers (bottom) are given. Dotted lines represent the calculated shell correction energies (102, 111), and the macroscopic $\alpha$ energies (102) and fission barriers (112) are given.
Table 4 The \( \alpha \) energies, mass excesses, shell corrections, and fission barriers for \( N - Z = 48 \) isotopes between \( Z = 102 \) and \( Z = 109 \)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>( ^{254}102 )</th>
<th>( ^{256}103 )</th>
<th>( ^{256}104 )</th>
<th>( ^{258}105 )</th>
<th>( ^{260}106 )</th>
<th>( ^{262}107 )</th>
<th>( ^{264}108 )</th>
<th>( ^{266}109 )</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>( E^{\alpha} ) (MeV)</td>
<td>8.42</td>
<td>8.46</td>
<td>8.81</td>
<td>9.30</td>
<td>9.76</td>
<td>10.38</td>
<td>11.10</td>
<td></td>
<td>( 95, 105 )</td>
</tr>
<tr>
<td>( M^{\text{ex}} ) (MeV)</td>
<td>82.87</td>
<td>89.66</td>
<td>94.23</td>
<td>101.52</td>
<td>106.62</td>
<td>114.48</td>
<td>128.17</td>
<td></td>
<td>( 102 )</td>
</tr>
<tr>
<td>( M^{\text{sh}} ) (MeV)</td>
<td>86.67</td>
<td>94.11</td>
<td>98.84</td>
<td>106.58</td>
<td>111.63</td>
<td>119.65</td>
<td>133.45</td>
<td></td>
<td>( 95 )</td>
</tr>
<tr>
<td>( \Delta E_{\text{sh}} ) (MeV)</td>
<td>-3.8</td>
<td>-4.4</td>
<td>-4.6</td>
<td>-5.1</td>
<td>-5.0</td>
<td>-5.2</td>
<td>-5.3</td>
<td></td>
<td>( 102, 111 )</td>
</tr>
<tr>
<td>( \Delta E_{\text{sh}} ) (MeV)</td>
<td>-2.4</td>
<td>-3.0</td>
<td>-3.1</td>
<td>-3.5</td>
<td>-3.5</td>
<td>-4.1</td>
<td>-4.8</td>
<td></td>
<td>( 102, 111 )</td>
</tr>
<tr>
<td>( B_{\text{f}} ) (MeV)</td>
<td>1.1</td>
<td>0.9</td>
<td>0.7</td>
<td>0.6</td>
<td>0.5</td>
<td>0.4</td>
<td>0.2</td>
<td></td>
<td>( 112 )</td>
</tr>
<tr>
<td>( B_{\text{f}}^{\text{exp}} ) (MeV)</td>
<td>4.9</td>
<td>5.3</td>
<td>5.3</td>
<td>5.7</td>
<td>5.5</td>
<td>5.6</td>
<td>5.3</td>
<td></td>
<td>( 95 )</td>
</tr>
<tr>
<td>( B_{\text{f}}^{\text{exp}} ) (MeV)</td>
<td>5.46</td>
<td>5.88</td>
<td>5.42</td>
<td>5.80</td>
<td>5.29</td>
<td>5.61</td>
<td>5.04</td>
<td></td>
<td>( 111 )</td>
</tr>
</tbody>
</table>

Figure 27 Experimental shell correction energy for nuclei with \( Z > 80 \). \( N > 152 \).

The fission barrier \( B_{f} \) is the sum of the macroscopic liquid drop surface and the shell contributions of the ground-state and saddle point masses. The experimental fission barriers are given in Table 4, Figure 27 (bottom), and are given for the \( N - Z = 48 \) isotopes in the following energy levels. The ground-state and saddle point energies are from (112). The macroscopic contribution to the fission barriers is difficult to measure and calculate, but it is known to be a small contribution. The macroscopic energy of the ground-state mass, \( -\Delta M^{\text{sh}} \), and the experimental fission barriers are shown as a function of the neutron number in Figure 27. The shell corrections are added to the macroscopic barriers giving the final fission barrier giving the main contribution.
The fission barrier protecting the isotopes of the new elements is high because of the strong ground-state stabilization of these nuclides. The stabilization shows an increasing trend in the entire range of the heaviest elements, as is demonstrated in Figure 23. Figure 22 (middle) compares the measured $\alpha$ energies with the $\alpha$ energies expected for nuclei having no shell corrections. For the calculation, the macroscopic masses of (102, 111) are used. The difference in $\alpha$ energies is always smaller than 0.4 MeV, which demonstrates again that the surface as a whole is shell stabilized independently of the individual proton and neutron numbers. The shell stabilization drastically changes the fission barriers, but only negligibly changes the $\alpha$ energies. The $\alpha$ energies reflect small local deviations of the mass surface, but they do not reveal the slowly changing trends toward higher shell correction energies. Only mass excess measurements disclose the increasing shell stabilization of the entire range of nuclides.

5.2 Half-lives for $\alpha$ Decay and Spontaneous Fission

The measured partial $\alpha$ lifetimes of the even-even $N - Z = 48$ isotopes were compared to lifetimes corresponding to the macroscopic $\alpha$ energies, Figure 24 (106). The $\alpha$ half-lives were calculated according to a prescription given in (113). All reduced $\alpha$ widths were set equal to one. Although a variation of 1 MeV in the $E_\alpha$ energies changes the lifetimes by three orders of magnitude, only a slight deviation (less than an order of magnitude from the macroscopic expectation) is observed experimentally. Once more this fact demonstrates that the shell corrections vary smoothly and the $\alpha$ lifetimes are affected only little by nuclear structure effects in this mass region. For a given element, $\alpha$ lifetimes are getting shorter and shorter with decreasing neutron number, a trend well described by macroscopic models and observed for many elements beyond $Z = 52$. As isotopes approach the proton drip line, their half-lives drop to the millisecond range. The short $\alpha$ half-lives observed are governed by this macroscopic trend, and nuclear structure is of minor importance only.

The contrary holds for the spontaneous fission lifetimes. To separate nuclear structure effects from macroscopic fission properties, we follow a slightly modified procedure (24), as originally introduced by Swiatecki (114). The fission half-life is given by

$$ T_{\alpha} \text{ (in seconds)} = 3 \times 10^{-21}/P, $$

with $P$ a barrier transmission factor, and the numerical factor the barrier knock-on time. Assuming a Hill-Wheeler-type transmission through the fission barrier with curvature $h_{\alpha} \beta$, we obtain

$$ P = [1 + \exp (2 \pi B_{\alpha} / h_{\alpha} \beta)]^{-1} \approx \exp (-2 \pi B_{\alpha} / h_{\alpha} \beta). $$

With $B_{\alpha} = B_{\alpha}^{\text{macro}} - \Delta E_{\text{shell}}$ and the shell corrections at the saddle point neglected, we obtain from Equations 10 and 11 an expression separating the nuclear structure effects from the macroscopic contribution:

$$ \log (T_{\alpha}) = 2.73 B_{\alpha}^{\text{macro}} / h_{\alpha} \beta - 2.73 \Delta E_{\text{shell}} / h_{\alpha} \beta - 20.5. $$

The experimental shell correction energies are obtained from mass excess values as described in Section 5.1, whereas the macroscopic fission barriers are calculated according to the semiempirical description given in (112). For isotopes with small shell correction energies and large values of $B_{\alpha}^{\text{macro}}$, the barrier curvature parameter $h_{\alpha} \beta$ may be fitted to the spontaneous fission half-lives. With this value of $h_{\alpha} \beta$ kept constant the macroscopic expectation may be calculated as a function of the fissility parameter $x$ (Equation 4). This macroscopic spontaneous fission half-life is presented, together with the experimental half-lives, in Figure 25 (top). For $^{260}$Fm the macroscopic fission half-life is about $10^{-12}$ s, which compares with 7 ms found experimentally. A stabilization by nuclear structure effects of 15 orders of magnitude is observed. Taking Equation 12 as a presentation with one adjustable parameter, values for $h_{\alpha} \beta$ as a function of $x$ may be obtained. Figure 25 (bottom) shows $h_{\alpha} \beta$ for all even-even spontaneous fission emitters. There is a smooth trend of $h_{\alpha} \beta$ for all isotopes between $U$ and $Cf$. For $^{254}$Fm and $Z = 102$ some isotopes still follow the trend, whereas, the isotopes of elements 104 and 106 show values of $h_{\alpha} \beta$ that are twice as large as the values for uranium.

Assuming no drastic changes in the inertia parameters, the increased values of $h_{\alpha} \beta$ point to a narrowing of the fission barrier by a factor of two. In Figure 25 (bottom) those isotopes for which a disappearance of the second fission barrier is predicted are indicated by points (115). Their positions support the hypothesis (48, 116, 117) that the change of half-life systematics
observed for $Z = 104$ is caused by such a disappearance. Moreover, these isotopes are shell stabilized. Their macroscopic barriers are smaller than 1 MeV. The values of $\hbar \omega_{1}$ were fitted to a linear dependence on the fissility parameter, giving for the $x$ dependence of the spontaneous fission half-life

$$\log (T_{1/2}) = (-3.55x + 6.62)B_{x} - 20.5.$$  

The smaller width of the barrier leads to a weaker dependence of the spontaneous fission half-life on the barrier height than that observed for lighter elements. A proportionality factor of 3.55 replaces the factor 6.88 found in (118). The fission through the narrow single-humped barrier most probably gives a symmetric mass distribution of fission fragments. Until now only the mass distributions of two of these nuclei ($^{260}104$ and $^{258}102$) have been measured and found to be symmetric (119). The total kinetic energy released in the fission of $^{260},^{258},^{256}104$ has now been measured (24, 119) and found to follow the systematics established for lighter elements (120).

The odd isotopes $^{255}104$ and $^{257}105$ have partial fission half-lives of 2.7 and 8.2 s, respectively. Comparison to $^{256}104$ allows us to determine the hindrance factors of spontaneous fission for odd protons and neutrons. Hindrance factors of 480 and 610 are obtained (24). Odd-odd isotopes may be hindered by the product of the individual hindrance factors, that is by a factor of $2 \times 10^{3}$. The $\alpha$ half-lives of the $Z = 107$ and $Z = 109$ isotopes and of the odd isotopes of $Z = 106$ are all shorter than the fission half-lives if we assume these hindrance factors and the fission half-lives of 7.2 ms ($^{260}106$) (95) and > 5 ms ($^{264}108$) (98) for the neighboring even-even isotopes as a reference. The unconfirmed spontaneous fission half-life of 1-2 ms obtained for $^{261}107$ by the Dubna group (92) was until recently the only exception.

Neither especially large hindrance factors nor nuclear structure effects in $\alpha$ half-lives are responsible for the absence of fission. The occurrence of $\alpha$ decay for the isotopes of elements 106 to 109 is mainly a consequence of a strong ground-state shell stabilization giving rise to increased fission half-lives.

5.3 Recent Theoretical Predictions of Shell Corrections and Half-lives

An island of macroscopically unstable, but shell-stabilized, spherical nuclei around $^{298}114,184$ was predicted as early as 1966 by Myers & Swiatecki (121). Many experiments sought to find these superheavy nuclei in nature or to produce them by nuclear reactions but failed (87). Predictions of shell corrections for nuclei between the heaviest isotopes known and the "superheavy" island are rare. Two calculations using the best macroscopic-microscopic models were published recently (111, 122). The shell corrections of the calculation of (111) are shown as an insert in Figure 23. Besides the strong shell effects at $^{298}114,184$ ($-7.08$ MeV) already known, another island at $^{272}109,163$ ($-6.93$ MeV) and $^{270}108,162$ ($-7.97$ MeV) has been predicted by (111) and (122), respectively. Nuclei not with $N = 184$ but with $N = 178$, 177 are found to have the largest shell corrections, e.g. $-8.97$ MeV for $^{298}114$ and $-8.38$ MeV for $^{298}116$. Following the successful path along $N - Z = 48$ leading to $^{260}109$ and further up to $^{276}114$, nearly constant shell corrections between $-4.6$ and $-5.0$ MeV are predicted. A continuous increase of the shell correction by another $-4$ MeV is expected.
going into the center of the island. For the isotopes of $Z = 114$ at $N = 163$, a change from deformed nuclei to spherical nuclei is predicted (111). The $N - Z = 42$, 49 paths lead to $Z = 114$ via a chain of deformed nuclei, whereas for $N - Z > 50$ spherical nuclei would be reached.

The new island around $^{272}_{109}$ consists of nuclei that are deformed ($\varepsilon_2 = 0.21$, $\varepsilon_4 = 0.09$) (111). The most interesting feature of these nuclei are the large $\varepsilon_4$ deformations (111, 122). Positive values of $\varepsilon_4$ correspond to a sausage-like shape of the nucleus. These sausage-like nuclei were seen before around $^{180}_{88}$Ta. But a microscopic explanation of why positive $\varepsilon_4$ deformation helps to stabilize nuclei was not given until recently. Whether the sausage shape at $N = 164 = 2 \times 82$ has to do with an underlying molecular cluster configuration of two $N = 82$ clusters is open to speculation.

The $N - Z = 48, 49$ isotopes of elements 112–114 are predicted to have $\alpha$ half-lives of about 0.2 $\mu$s and fission barriers of about 5 MeV (111). Experimentally for the elements $Z = 100–106$ the $\alpha$ half-lives are found to be larger than postulated; thus $\alpha$ half-lives may be around 1 $\mu$s for elements 112–114. Experimental shell corrections are stronger by about 1 MeV than calculated; thus fission barriers may be at least 5.5 MeV for the isotopes in question. Fission half-lives of about 20 ns can be extrapolated from Equation 13, and for all isotopes negligible fission branch can be expected. The $\alpha$ chains starting at elements 112–114 will end by fission in $^{257}_{105}$ or $^{256,258}_{104}$, or will continue to still lighter elements. Isotopes $^{271}_{114}$ and $^{273}_{112}$ decay by a chain that never shows fission and finally ends in $^{209}_{82}$Bi. Even for the light isotopes of elements 112–114 near $N < 163$, the ground-state stability may still be sufficient to allow detection with present techniques. The stability proceeding to heavier isotopes of element 114 will increase and the expectation of a maximum stability near $N = 184$ remains essentially unchanged. The new aspect that superheavy elements may not decay by fission, but could be detected by $\alpha$ chains and thereby directly give their atomic and mass number, is surprising indeed.

5.4 Production Cross Sections

Isotopes of elements $Z = 100–109$ were produced by cold fusion reactions between beams of $Z = 18–26$ and targets of $Z = 81–83$. Trends in production cross sections are discussed in the following. The first experiments producing heavy elements by cold fusion were performed at Dubna in 1974 (49). $^{40}_{18}$Ar projectiles bombarded Pb targets and spontaneous fission events were observed, which could be assigned to Fm isotopes. First cross sections and excitation functions were determined (49, 67). The experiments were later repeated at Berkeley (68) and Darmstadt (80). The Darmstadt experiment corroborated the earlier Dubna finding of 2n-

reaction channels and ended a controversy between Dubna and Berkeley on the existence of this cold fusion channel (123).

A compilation of the cross sections found in Ar bombardments of Pb targets for all the different reaction channels was published recently (34). The 2n and 3n channels have about equal cross sections, 4n channels are seen with a much smaller cross section. The existing data are reproduced for all reaction channels by modern evaporation codes (23) and do not indicate any dynamical hindrance in the entrance channel. However, subbarrier fusion has to be considered by a barrier fluctuation of 3% in order to reproduce the 2n cross sections, and the damping of shell effects at high temperature has to be taken into account by a damping energy $E_D = 18$ MeV in agreement with microscopic predictions (24, 34).

There are no cross sections known for the production of Md and element 103 by cold fusion. The production of isotopes of element 102 by fusion of Ca and Pb isotopes was studied extensively (68, 124). The high (several $\mu$b) production cross section for $^{254}_{102}$ in the reaction $^{40}_{20}$Ca($^{208}_{82}$Pb, 2n) found in experiments at Dubna (124) and Berkeley (68) could not be reproduced by the recoil spectrometer SHIP (24). The discrepancy at SHIP of nearly a factor of 10 in the cross section may be due to a reduced efficiency of the recoil spectrometer; indeed, in the case of a 100-ns isomer in $^{254}_{102}$, the efficiency might be considerably decreased by a conversion-induced Auger cascade (24). For element 102 isotopes, the 2n-reaction channel in the $^{48}_{21}$Ca reactions has become the dominant channel and all other channels are weaker by at least an order of magnitude. Again an evaporation calculation reproduces the data without assuming any hindrance in the entrance channel (24). More data on the weak channels would definitely help to determine whether or not a small hindrance might already be present. To describe the subbarrier fusion part of the excitation function, a small barrier fluctuation of 1.3% as expected for stiff nuclei is needed. The doubly-closed-shell nuclei $^{48}_{20}$Ca and $^{208}_{82}$Pb fuse without any hindrance at a well-defined barrier, as already found for the symmetric fusion of the two closed-shell $^{90}_{40}$Zr nuclei (Figure 5; Section 2.1).

For elements 104 and 105 some excitation functions were measured using reactions with $^{50}_{20}$Ti beams (50, 91). The 1n- and 2n-reaction channels have about equal yields. The small cross sections (only a few nb) for the 2n channel and the observation of 1n channels are reproduced by the evaporation calculations only, if a dynamical hindrance at the barrier of about a factor of 25–30 is assumed. Such a hindrance is equivalent to a barrier shift of $21 \pm 3$ MeV and an increase of the barrier fluctuation by a factor of two to about 6%. This extra-push value is in agreement with a $\frac{x_{\text{c}}}{\text{mean}}$ scaling (Figure 5) and the large fluctuation was observed for equivalent symmetric systems as well (22).
It was shown that the same parameters in the evaporation calculation reproduce well the excitation function in the $^{249}$Cr($^{12}$C, 4n)$^{253}$104 reaction without assuming any dynamical hindrance, which again corroborates the extra-push scaling that predicts for this reaction no entrance channel limitation. The sudden onset of a dynamical hindrance for the $^{250}$Ti reaction and its absence in $^{46}$Ca reactions points to a nuclear structure effect besides the macroscopic dissipation mechanism underlying the extra-push concept. The addition of two protons to $^{48}$Ca or of two neutrons to $^{50}$Zr in both cases leads to a considerable shift of the fission barrier and an increase in the barrier fluctuation parameter. The $\Gamma_n/\Gamma_f$ values at the low excitation energies present in cold fusion are of the order of a few percent in agreement with the analysis presented in Figure 8, and the high fission barriers as deduced from the analysis of ground-state shell corrections (Section 5.1). A damping of shell effects with temperature as expected from microscopic theory ($E_D = 18$ MeV) is compatible with the data.

Figure 26 (top) gives the cross sections as measured for reactions leading to isotopes of elements 102 to 109 via compound systems with $N - Z = 50$ and 49 for even and odd elements, respectively. All values are found in Table 2, except $\sigma_{2n} = 190$ nb and $\sigma_{3n} = 7.3$ nb obtained from the reaction $^{206}$Pb($^{46}$Ca, xn) (24). The slope of the cross sections for 3n and 2n reactions is larger than for 1n reactions. On the average the 1n cross section decreases by a factor of 3.5 for each element. For elements 104 and 105, the cross sections for 1n reactions become comparable to 2n cross sections and finally prevail for elements 107 to 109. Comparison of cross sections for pairs of nuclei like $^{207}$Pb/$^{208}$Pb or $^{49}$Ti/$^{50}$Ti shows that more neutrons may increase the yields, e.g. the 2n production cross sections of $^{256,255}$104 in reactions of $^{50}$, $^{49}$Ti with $^{208}$Pb differ by a factor of four, whereas no difference is found for reactions of the pair $^{208}$Pb/$^{209}$Pb with $^{50}$Ti leading to the same isotopes (Table 2). The difference of one neutron in the lead isotopes for pairs of 2n and 1n reactions leading to a given isotope changes the cross section in favor of $^{209}$Pb by factors of 9, 1.3, and 2.5 for $^{256,254}$104, $^{260}$106, and $^{264}$108, respectively. Q-value arguments favor neutron-rich collision partners. On the average, the available scarce data show an additional neutron to increase the cross section by a factor of two.

The excitation energy for all systems showing In cross sections between elements 104 and 109 decreases from 24 to 20 MeV, the fission barriers stay nearly constant (Figure 22, bottom) and still the cross section decreases by nearly three orders of magnitude. If we assume for $B_v$ constant approximately constant $\Gamma_n/\Gamma_f$ values for all reactions, this decrease can be attributed to an increase of the entrance channel limitation. Figure 27 shows the ln cross sections as a function of the scaling parameter $x_{\text{mean}}$. The

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**Figure 26** Upper part: Experimental cross sections for different xn channels observed in cold fusion reactions leading to compound systems with $N - Z = 49$ and 50 (Table 2). Lower part: The ln cross sections for cold fusion reactions compared to 4n cross sections for $^{249}$Cr- and $^{248}$Bk-based reactions (2, 4). The lines are fits to the data.

**Figure 27** The ln cross sections observed for cold fusion reactions as a function of the $x_{\text{mean}}$ scaling parameter.
odd-even effect clearly seen in Figure 26 (top) apparently disappears. As all EVRs are found near the Bass barrier, the fusion probability at the barrier \( p(B) \) is obtained from the cross section values, if the \( p(B) \) values are normalized to the \( p(B) \) value obtained for the \( ^{208}\text{Pb}(^{50}\text{Ti}, xn) \) reaction and if we assume the decrease in cross section is caused by a decrease of the fusion probability alone. The values of \( p(B) \) thus obtained (see Figure 6) are lower values because \( \Gamma_\nu/\Gamma_F \) is assumed to stay constant between \( Z = 104 \) and 109. The value of \( p(B) \) for production of \( ^{266} \text{109} \) is \( 5.5 \times 10^{-5} \). An extra-push energy of 85.4 MeV is calculated from Equation 5, with the parameters of Figure 5.

The dynamical barrier is shifted to energies where EVRs cannot survive. Their actual detection at the Bass barrier shows that fluctuations of the order of the shift exist. This corroborates the finding in Section 2.1 that the concept of a well-defined barrier breaks down for systems with large dynamical hindrance. Not only the barrier concept, but also the entrance-exit channel concept, may be questioned for small fusion probabilities and washed-out dynamical barriers. Single high energy neutrons could be emitted in an early stage of the fusion process and cool the system below the fission barrier faster than fission may occur (125). Such a mechanism may be of special importance for small 1n cross sections.

5.5 On the Production of Still Heavier Elements

In the experiments performed to produce highly fissionable isotopes, a three-fold limitation of the production process has been observed.

1. The limitation by fission losses in the evaporation cascade—a thermal limitation in the exit channel (Figure 26, bottom).
2. The “extra-push” limitation—a dynamical limitation in the entrance channel (Figures 5 and 6).
3. The nuclear structure–dependent limitation of the shell stabilization with excitation energy—a structural limitation (Figures 9 and 10).

The production of still heavier elements between \( Z = 110 \) and 114 is, as shown in the analysis of Sections 5.1 and 5.2, not a question of the ground-state stability of these elements, but a question of navigating between the three-fold limitation of the production process.

The first limitation has to be overcome if the production of heavy elements using actinide targets is to be continued. Figure 26 (bottom) compares the largest 4n cross sections using \( ^{249}\text{Bk} \) and \( ^{249}\text{Cf} \) targets to produce isotopes of elements 100–106 with 1n cross sections for cold fusion reactions. For actinide-based reactions, the two other limitations do not apply as long as the reaction aims at deformed isotopes for elements 110–114 (\( N < 165 \)). As the analysis of mass excesses and fission half-lives points to nearly constant fission barriers, the average \( \Gamma_\nu/\Gamma_F \) values in the evaporation cascade may, in contrast to the trend observed up to element 106, stay fairly large and the cross sections may decrease more slowly than observed for \( Z < 106 \). This speculation makes reactions such as \( ^{254}\text{Es}(^{23}\text{Na}, 4n)^{273}110 \), \( ^{249}\text{Cf}(^{26}\text{Mg}, 4n)^{271}110 \), or \( ^{235}\text{U}(^{48}\text{Ar}, 4n)^{271}110 \) possible candidates for proceeding beyond 109. The production cross sections will certainly be small. Estimates are uncertain and range between 100 and 0.1 pb. Only the detection of decay chains will allow us to identify the EVRs, since transfer reactions with actinide targets produce fission activities with much larger cross sections than do Pb- and Bi-based cold fusion reactions. The thermal limitation that since 1974 has prevented the actinide-based production of elements heavier than \( Z = 106 \) may be overcome by using recoil spectrometers and improved detection methods. Compared to the Berkeley 106 experiment (4), the sensitivity of the techniques used to detect \( ^{266}109 \) was improved by at least two orders of magnitude (100).

The entrance channel limitation reduces the cross section for production of element 110 by cold fusion to the level of a few pb (Figure 27). If the \( ^{56}\text{Fe} \) beams are replaced by \( ^{64}\text{Ni} \) beams, the cross sections may decrease only marginally because the decrease may be partly compensated by the higher number of neutrons in \( ^{64}\text{Ni} \), which will lead into a more stabilized \( N - Z = 52 \) compound system. The best cold fusion reaction for discovering element 110 will be \( ^{208}\text{Pb}(^{64}\text{Ni}, 1n)^{271}110 \) at an energy near the unshifted fusion barrier.

During the years, many attempts to detect superheavy elements have been undertaken (87). One of the more recent failures to produce isotopes of element 116 in the reaction \( ^{248}\text{Ca}(^{48}\text{Ca}, xn)^{296} - xn116 \) gave a lower limit of 100 pb for a time range of 14 orders of magnitude, starting from the microsecond range and ending at half-lives of several years (126). The excitation energy at the barrier in the above reaction is at least 30 MeV. The main reaction channels to be expected are 2n and 3n reactions leading to the isotopes of element 116 with 178 and 177 neutrons. According to the most recent calculations (111), these are the neutron numbers with the highest shell corrections (\( \sim -8.4 \) MeV). The time range covered in the experiment, following Equation 13, is equivalent to a change of 4 MeV in the shell correction, which is much larger than the uncertainty of the calculation. The spherical nuclei around \( N = 178 \) very probably are stable enough to be detected in the time range covered. They would have been seen if they were produced. The three-fold limitation in the production process explains at least qualitatively why we failed to produce them.

With \( x_{\text{mass}} = 0.805 \) we have have to expect about the same dynamical hindrance as for the production of \( ^{262}107 \), about \( 10^{-5} \) (Figure 6). With 2n
channels at the barrier \( E^* = 30 \text{ MeV} \), a thermal hindrance of the order of \( \Gamma_0/\Gamma_1 \approx 0.03 \) compared to a 1n channel in a cold fusion reaction is to be expected. Moreover, the production of the spherical nuclei is structurally limited. Taking as a reference the cross section for the production of \(^{216}\text{Th} \) (Figure 9), which is reduced by a factor of 50, a structural limitation of at least the same order of magnitude may be expected. Compared to the production of \(^{260}\text{107} \) \( (\sigma \approx 0.2 \text{ nb}) \), the additional hindrance of at least a factor of 10 explains why at a limit of 100 pb no atoms of element 116 were found in the above experiments. With cross sections of at most 0.1 pb it is difficult to imagine how the structural limitation blocking the access to the spherical superheavy nuclei could ever be overcome.

The outcome of other experiments attempting to go beyond \( Z = 109 \) is open. Only experiments can decide which of the limitations, the dynamical or cold fusion reactions or the thermal of actinide-based reactions, may be more easily overcome. The three-limitation will stop our search for man-made elements either at \( Z = 109 \) or at a few atomic numbers more. In any case, the game is limited and we should accept the study of the limitations to be a message in itself. The idea of superheavy elements—element shell stabilized, not existing in a world of macroscopic nuclear drops—has already been realized. The isotope \(^{260}\text{109} \) is already completely shell stabilized. For \(^{260}\text{106} \), the spontaneous fission half-life is increased by 15 orders, and without nuclear structure effects the isotope would never have been detected. Entering the island of \( \alpha \)-stabilized \( \alpha \) emitters at \( Z = 106 \) we produced elements that correspond to what superheavy elements are to be. They are not spherical nuclei, but deformed sausage-like entities; they live not billions of years, but milliseconds. We are reluctant to accept the idea that our dream of superheavy elements has already been fulfilled.

The detection of one atom of \(^{260}\text{109} \)—the discovery of an element via one atom—may be reward enough, and from a more distant point of view it is difficult to imagine a more beautiful end of the element game. The birth, life, and death cycle of the one atom of element 109, which was born out of two, lived for a short time as the heaviest atom ever made, and disintegrated again into two, reminds us of a Greek saying: "\( \epsilon v \to \nu \alpha v— \)The One that may stand for All."

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PRODUCTION AND DECAY OF THE $b$ QUARK

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