

S. Y. Lin, Phys. Rev. Lett. **33**, 1487 (1974).

⁸*Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington, D. C.).

⁹T. Sikkeland, Phys. Rev. **135**, B669 (1964).

¹⁰R. Bass, Nucl. Phys. **A231**, 45 (1974).

¹¹D. L. Hendrie *et al.*, Phys. Lett. **26B**, 127 (1968);
N. K. Glendenning *et al.*, Phys. Lett. **26B**, 131 (1968);

W. Brückner *et al.*, Nucl. Phys. **A231**, 159 (1974);
A. H. Shaw and J. S. Greenberg, Phys. Rev. C **10**, 263 (1974).

¹²J. Randrup and J. Vaagen, to be published; R. Bass, private communication.

¹³R. G. Stokstad, Z. E. Switkowski, R. A. Dayras, and R. M. Wieland Phys. Rev. Lett. **37**, 888 (1976);
M. Arnould and W. M. Howard, Nucl. Phys. **A272**, 295 (1976).

Isotope Distributions in the Reaction of ^{238}U with ^{238}U

M. Schädel, J. V. Kratz, H. Ahrens, W. Brüchle, G. Franz,
H. Gäggeler, I. Warnecke, and G. Wirth
Gesellschaft für Schwerionenforschung, D-61 Darmstadt, Germany

and

G. Herrmann,^(a) N. Trautmann, and M. Weis
Institut für Kernchemie, Universität Mainz, D-65 Mainz, Germany
(Received 22 May 1978)

Radiochemically determined cross sections $\sigma(Z, A)$ were used to construct charge and mass distributions for the reaction of 1785-MeV ^{238}U ions with thick ^{238}U targets. Fission of the colliding nuclei is found to dominate. For the surviving uraniumlike fragments an enhancement of yields compared to the Kr + U and Xe + U reactions is observed. The formation of heavy actinides is shown to be associated with the low-energy tails of the broad excitation energy distributions in damped collisions.

The mechanism of strongly damped collisions between very heavy nuclei is of great current interest.¹ The ^{238}U beam available at the Unilac accelerator is presently being used to investigate the $^{238}\text{U} + ^{238}\text{U}$ reaction with several complementary techniques. First results of $\Delta E, E$ counter telescope measurements were published by Hildenbrand *et al.*² The aim of the present work was (i) to extend earlier radiochemical studies³⁻⁶ of mass and charge distributions in reactions of ^{238}U with ^{40}Ar , ^{56}Fe , ^{84}Kr , and ^{136}Xe ions to the U + U system, and (ii) to learn about the prospects of synthesizing superheavy elements in the latter reaction. Therefore, particular emphasis was put on the investigation of the survival probability of heavy actinide isotopes after their formation in damped collisions.

The experiments were performed with ^{238}U beams of 7.5 MeV/amu and up to 2.5×10^{11} particles/s incident on a thick, water-cooled uranium metal target (300 mg/cm²). All reaction products are stopped in the target. After bombardment the target was dissolved and the reaction products were chemically separated into 25 fractions which were assayed for x-ray, γ -ray,

α -particle, and spontaneous-fission activities over a period of several months. From these data integral cross sections $\sigma(Z, A)$ for individual isotopes with Z ranging from 26 to 100 and half-lives from 23 min to 7.4×10^3 yr were obtained. The cross sections represent mean values between the incident energy and the interaction barrier (6.1 MeV/amu) and were calculated⁷ using an effective number of 1.5×10^{19} target atoms per square centimeter. The $\sigma(Z, A)$ values were used to define a surface of independent formation cross sections in a Z - A plane [see Fig. 1(b)]; the process to generate the surface was discussed in Ref. 3. Corrections for products from reactions of ^{238}U ions with ^{16}O target impurities were made, if necessary, as described elsewhere.⁷

We interpret the charge distribution, Fig. 1(a), as being due to the superposition of four components: (i) products around uranium from quasi-elastic transfer (890 mb), (ii) nuclides with Z from ~ 73 to 100 arising from an originally symmetric distribution in the binary deep-inelastic transfer (290 mb), (iii) a nearly symmetric fission-product distribution originating from the sequential fission of highly excited binary inelas-

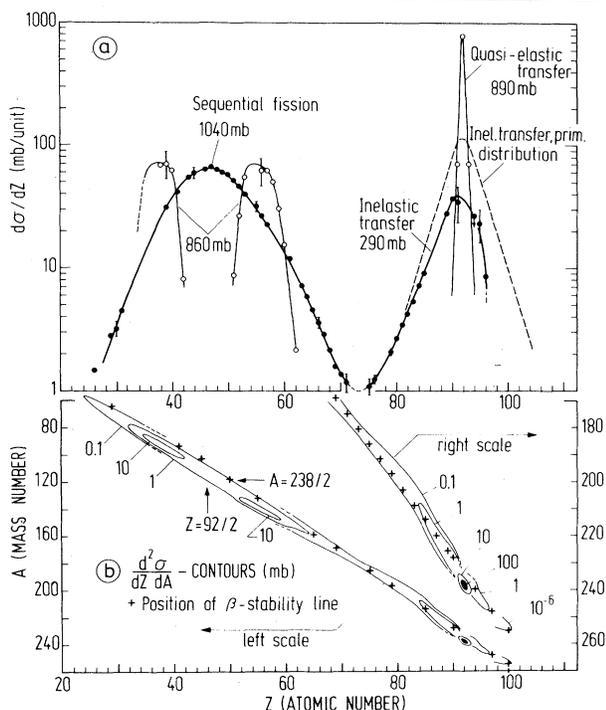


FIG. 1. The $^{238}\text{U} + ^{238}\text{U}$ reaction at ≤ 7.5 MeV/amu energy: (a) charge distribution for quasielastic transfer and sequential fission at low excitation energies (open circles), and for damped collisions with the associated sequential fission process (full circles), and the reconstructed primary fragment distribution (dashed curve). (b) Independent cross section isopleths in a Z - A plane.

tic fragments (1040 mb), and (iv) a double-humped distribution of neutron-rich fission products from sequential fission of transfer products being formed at low excitation energies (860 mb). In spite of the expected continuous dependence of yield on energy loss, a decomposition of the fission-product distribution in components (iii) and (iv) is strongly suggested by the measured isotope distributions for $33 \leq Z \leq 43$ and $50 \leq Z \leq 61$ which can be fitted by two Gaussians of different locations and widths.⁷ In the evaluation of cross sections for different reaction mechanisms one must take into account that two, three, or four fragments arise from a single collision. The sum of components (i) through (iv) represents the total reaction cross section, 1060 mb, which defines an effective beam energy³ of 6.9 MeV/amu in the thick target. The partial cross section for the damped collision process results as 405 mb from the sum of cross sections for components (ii) and (iii). This corresponds to a fraction of 0.4 of the reaction cross section. A

ratio of 0.5 has been deduced² by reconstructing the primary charge distribution of uraniumlike fragments at the higher beam energy of 7.42 MeV/amu.

In order to deduce the primary charge distribution for the damped collision process we use its integral cross section of 405 mb, and assume the distribution to be symmetric around $Z = 92$ and to have the usual shape as found, e.g., in the Xe + Au reaction.⁸ The width of the distribution is adjusted to the measured element yields near $Z = 80$ where fission probabilities are negligible.² The resulting primary distribution is shown as the dashed curve in Fig. 1(a). The maximum of the surviving products lies close to $Z = 91$ rather than at $Z = 80$ or 85 as observed in the Kr + U and Xe + U reactions,^{5,6} respectively. Apparently, the fission competition which truncates the original fragment distribution is less important in the U + U system. This is also evident in the counter experiments² which show that given elements in the U environment are produced at smaller average excitation energies in U + U collisions than in the other reactions.

The formation cross sections for actinide isotopes in the $^{238}\text{U} + ^{238}\text{U}$ reaction ($E \leq 1.24B$ where B is the interaction barrier) are shown in Fig. 2 and are compared with thick-target cross sections⁷ for the $^{136}\text{Xe} + ^{238}\text{U}$ reaction at $E \leq 1.40B$ which agree well with data⁹ at $E \leq 1.56B$. Such a comparison is meaningful because neither the Xe + U thick-target actinide cross sections^{7,8} nor the U + U thick-target actinide cross sections⁷ depend significantly on incident energy above $1.2B$. The enhancement of cross sections in the U + U reaction is obvious. In order to learn about mechanisms responsible for the formation of surviving actinides we reconstruct the measured product populations for the complementary elements $_{86}\text{Rn}$ - $_{98}\text{Cf}$, $_{85}\text{At}$ - $_{99}\text{Es}$, and $_{84}\text{Po}$ - $_{100}\text{Fm}$ with the following assumptions: (i) The isotope distributions for the light complements Ra, At, and Po are not significantly influenced by fission occurring in the deexcitation of these fragments. (ii) The most probable mass number of primary fragments $A_F'(Z)$ can be deduced following Volkov's¹⁰ generalized Q_{gg} systematics including neutron-pairing corrections δ_n . Comparison of $A_F'(Z)$ with the measured mean product mass number $A_F'(Z)$ gives the average number of neutrons $\bar{\nu}$ evaporated from the primary light fragments and their mean excitation energy as calculated with an evaporation code. The mean total excitation energy $\bar{E}_{\text{tot}}(Z)$ for the light and heavy fragments

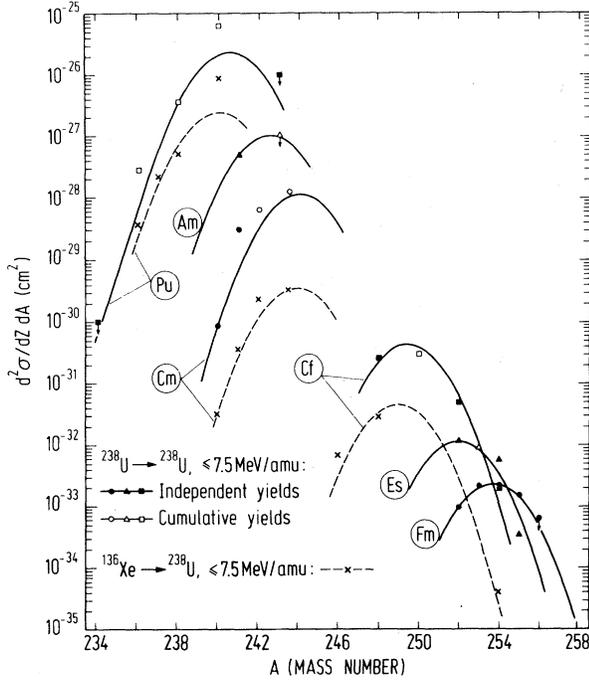


FIG. 2. Cross sections for the formation of heavy actinide isotopes in the U + U reaction at ≤ 7.5 MeV/amu. For comparison, similar data (Ref. 7) for the $^{136}\text{Xe} + ^{238}\text{U}$ reaction at ≤ 7.5 MeV/amu (dashed curves) are given. The curves are drawn to guide the eye.

is then estimated by assuming a partition of the excitation energy between the two fragments in proportion to their initial masses. (iii) The measured variance σ_F^2 of the isotope distribution for fixed Z results from the superposition of three dispersions: $\sigma_P^2 \approx \sigma_F^2 + \sigma_E^2 + \sigma_\nu^2$. Here, σ_F represents the width of the primary fragment isotope distribution around $A_F'(Z)$, σ_E accounts for the broad range of excitation energies associated with the formation of a given primary fragment $A_F(Z)$ which leads to a broad range of final products $A_P(Z)$ in the neutron evaporation process, and σ_ν reflects the fluctuations in the number of evaporated neutrons from a given fragment $A_F(Z)$ at fixed excitation energy.

The measured isotope distribution for polonium corresponds to $\sigma_P^2 = 6.9$ amu². The variance σ_ν^2 is not larger than 0.5 in the present experiments, as follows from evaporation calculations with the code ALICE. For σ_E^2 a value of ~ 4.5 amu² is estimated from the Q -value dispersions² of about 100 MeV FWHM (full width at half-maximum). We note here that the dispersion in the excitation energy is the dominant contribution in the U + U reaction. If we neglect the dependence of σ_F and σ_ν

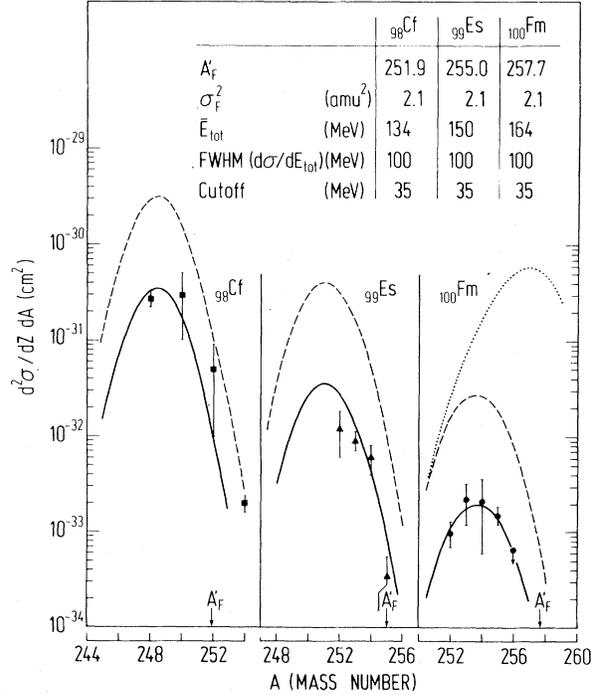


FIG. 3. Comparison of measured (symbols) and calculated isotope populations (curves) for the heaviest actinide elements observed in the U + U reaction. The calculations are outlined in the text.

on excitation energy, the variance of the primary fragment isotope distributions in the Po–Fm region can be estimated as $\sigma_F^2 \approx 2.1$ amu² (FWHM ≈ 3.4 amu).

We can now describe the population $Y(Z, A_P)$ of a given actinide isotope by starting from such a narrow primary fragment distribution $Y_i(Z, A_F)$ around the centroid mass number A_F' as derived from values of $Q_{gg} - \delta_n$ by simulating the evaporation of $x_i = A_F - A_P$ neutrons:

$$Y(Z, A_P) = \sum_{i=1}^k \left(\frac{\Gamma_n}{\Gamma_n + \Gamma_f} \right)^{x_i} Y_i(Z, A_F) \times \int \frac{d\sigma}{dE} P(x_i) dE. \quad (1)$$

Here, $\Gamma_n / (\Gamma_n + \Gamma_f)$ corrects for fission competition in the evaporation chain using an empirical approach,¹¹ $P(x_i)$ (extracted from ALICE code calculations) gives the probability for the evaporation of x_i neutrons, and $d\sigma/dE$ (assumed to be Gaussian) accounts for the dispersion in excitation energy and is adjusted so as to reproduce the isotope distribution of the complementary light element. While the most abundant Rn, At,

and Po isotopes are predominantly formed in collisions where energies close to the mean value are deposited in the system, the calculations show that the *surviving* transcurium isotopes originate exclusively from the low-energy tails of the excitation-energy distributions. In Fig. 3 the measured Cf, Es, and Fm isotope cross sections are compared with calculated populations using Eq. (1) and parameters derived from the experimental light-product isotope distributions as indicated in the figure. The dotted curve represents the Fm case where the total excitation energy is shared between the fragments in proportion to their masses. Since isotopes close to A_F' are not observed experimentally we conclude first that the Gaussian energy distributions do not extend to zero which indicates that a minimum energy dissipation is required for given charge transfers. We take care of this by introducing an energy cutoff. This yields the dashed curves in Fig. 3 which reproduce the position and width of the experimental distributions but still fail to reproduce the absolute cross sections by one order of magnitude. We discuss two possible reasons: (i) The Sikkeland formula¹¹ may systematically overestimate Γ_n/Γ_f by a factor of 2; for $\bar{\nu} \approx 4$ (Fm) this would account for the discrepancy. (ii) Agreement between experiment and our estimates is obtained (see solid curves in Fig. 3) if, starting with $Z_h \geq 98$ ($Z_l \leq 86$), it is assumed that the total excitation energy in the low-energy collisions is more and more concentrated in the heavy fragment. This may be correlated with the simultaneous formation of a nearly magic light fragment ($Z \approx 82$) which does not take up much excitation energy provided that the Q value is small enough for shell effects to be relevant. One may speculate that this effect might reverse for the formation of fragment pairs near $Z = 114, 70$ in the low-energy tails of the Q -value distributions, a feature highly desirable for the production of superheavy elements. With respect to the formation of primary fragments of such elements, it is interesting to note that an extrapolation of the original fragment distribution [dashed curve in Fig. 1(a)] indicates cross sections of 0.1 mb for $Z = 114, 70$. A more detailed analysis of the actinide cross sections including the influence of the angular momentum transfer on the survival probability

is in progress.⁷

In summary, the observed isotope distributions originating from damped collisions of U + U reveal large dispersions in the amount of energy dissipation at a given charge transfer. The production of highly fissionable heavy fragments is shown to depend entirely on the low-energy tails of these broad Q -value distributions and possibly on nuclear structure effects influencing the sharing of the total excitation energy between complementary fragments.

The authors are indebted to the staff of the Unilac for providing high-intensity uranium beams. Discussions with H. J. Specht and W. Nörenberg are gratefully acknowledged. The Mainz group has been supported by a research grant of the Bundesministerium für Forschung und Technologie. This work was submitted by one of us (M.S.) in partial fulfillment of a doctoral degree at the University of Mainz, Mainz, Germany.

^(a)Also at Gesellschaft für Schwerionenforschung, Darmstadt, Germany.

¹W. U. Schröder and J. R. Huizenga, *Annu. Rev. Nucl. Sci.* **27**, 465 (1977).

²K. D. Hildenbrand, H. Freiesleben, F. Pühlhofer, W. F. W. Schneider, R. Bock, D. v. Harrach, and H. J. Specht, *Phys. Rev. Lett.* **39**, 1065 (1977).

³J. V. Kratz, J. O. Liljenzin, A. E. Norris, and G. T. Seaborg, *Phys. Rev. C* **13**, 2347 (1976).

⁴U. Reus, A. M. Habbestad-Wätzig, R. A. Esterlund, P. Patzelt, and I. S. Grant, *Phys. Rev. Lett.* **39**, 171 (1977).

⁵J. V. Kratz, A. E. Norris, and G. T. Seaborg, *Phys. Rev. Lett.* **33**, 502 (1974).

⁶R. J. Otto, M. M. Fowler, D. Lee, and G. T. Seaborg, *Phys. Rev. Lett.* **36**, 135 (1976).

⁷J. V. Kratz, W. Bröchle, H. Gäggeler, G. Herrmann, M. Schädel, N. Trautmann, I. Warnecke, M. Weis, and G. Wirth, to be published.

⁸J. V. Kratz, H. Ahrens, W. Bögl, W. Bröchle, G. Franz, M. Schädel, I. Warnecke, G. Wirth, G. Klein, and M. Weis, *Phys. Rev. Lett.* **39**, 984 (1977).

⁹K. L. Wolf, J. P. Unik, E. P. Horwitz, C. A. Bloomquist, and W. Delphin, *Bull. Am. Phys. Soc.* **22**, 67 (1977), and private communication.

¹⁰V. V. Volkov, in *Proceedings of the International Conference on Reactions between Complex Nuclei, Nashville, June, 1974* (North-Holland, Amsterdam, 1974), Vol. II, p. 363.

¹¹T. Sikkeland, A. Ghiorso, and M. J. Nurmi, *Phys. Rev.* **172**, 1232 (1968).