PROPERTIES OF SUPER-HEAVY NUCLEI AND SEARCH FOR ELEMENT 120*

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Dedicated to the late Professor Adam Sobiczewski

Experimental data related to the discovery of elements 107 to 112 measured at the velocity filter SHIP at GSI, Darmstadt, confirmed the predictions of a region of relatively high nuclear stability of deformed super-heavy nuclei. This region is located at proton and neutron numbers 108 and 162, respectively. The isotopes were successfully produced using cold fusion reactions. Experimental exploration of the predicted island of spherical super-heavy nuclei was only possible using hot fusion reactions. At the GSI SHIP, one of these reactions was studied using a ^{248}Cm target. In reactions with a ^{48}Ca beam, the previously known data on isotopes of element 116 were confirmed. Results from an attempt to search for element 120 using a ^{54}Cr beam are presented. In a complementary study, relative masses, model-dependent shell-correction energies, and related heights of fission barriers were deduced from measured Q_α values. The results are compared with predictions of macroscopic–microscopic models. The consequences for calculations of cross sections are discussed.

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1. Introduction

Definitely, a meaningful search for and production of super-heavy nuclei (SHN) was triggered by theoretical predictions. These started with the introduction of the nuclear shell model. An important improvement was obtained by Sobiczewski, Gareev and Kalinkin who predicted proton number $Z = 114$ and neutron number $N = 184$ as the next shell closures beyond

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^{208}Pb. This result was obtained using a Woods–Saxon potential-well in calculations within the framework of the macroscopic–microscopic (MM) model in 1966 [1]. In the following years and up to now, the models have been continuously improved. A review on theoretical works has been presented in [2].

On the other hand, erroneous theoretical developments were corrected on the basis of new experimental findings. Examples are the predictions of a so-called extra-push energy for fusion and the underestimation of quasi-fission in early cross-section calculations.

The nuclei presently known in the region at the upper end of the chart of nuclei are plotted in Fig. 1. These heavy and super-heavy nuclei were produced in so-called cold fusion reactions using targets of lead and bismuth

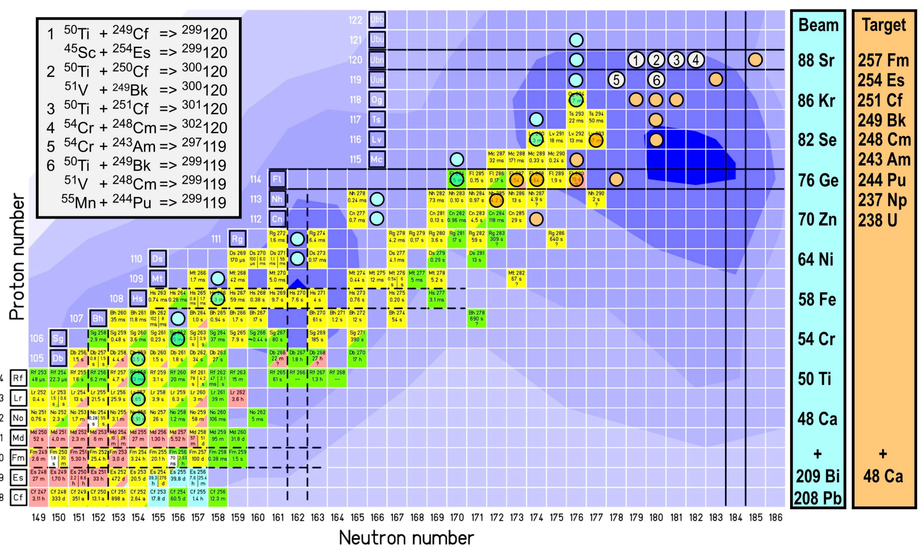

Fig. 1. (Colour on-line) Upper end of the chart of nuclei showing the presently known isotopes. Circles mark the produced and possible compound nuclei (CN) reachable in cold (blue) and hot (orange) fusion reactions. Beams are given for cold fusion on the left of the two columns to the right and targets for production of the most neutron-rich CN using hot fusion on the right of the two columns. CN of possible hot fusion reactions for production of isotopes of elements 119 and 120 (shown in the inset, top left) are marked with numbers 1–6. For completeness, CN produced in reactions with a ^{48}Ca beam and targets of ^{254}Es and ^{257}Fm are also shown, although sufficient material necessary for a production experiment can presently not be produced. See discussion in [3] for a tentative assignment of an electron capture (EC) branch of ^{287}Fl and ^{290}Fl and tentative re-assignment of the decay chains measured in [4–6]. The background structure shows the calculated shell-correction energies from -7 MeV (dark blue/black) in steps of $+1$ MeV obtained by Sobiczewski *et al.* using the macroscopic–microscopic model [2].

and in hot fusion reactions using actinide targets. Excitation energies of the produced compound nuclei (CN), shown as circles in Fig. 1, are 10–20 MeV in the case of cold fusion and 35–55 MeV for hot fusion. Therefore, the highest cross sections are measured for evaporation of 1 and 2–5 neutrons in the case of cold and hot fusion, respectively. The experimental results and descriptions of the experimental set-ups are presented in detail in review articles [3, 7–10].

The reason for the stability of heavy and super-heavy nuclei are shell effects. In MM models [2, 11], these are added as shell correction energies (SCE) to the macroscopic binding energy determined from the liquid drop model. The background colour in Fig. 1 shows this SCE for a wide range of heavy nuclei. Clearly, two regions of minimal SCE are recognized, one for deformed nuclei located at proton number $Z = 108$ and neutron number $N = 162$, the other for spherical nuclei near the double shell closure at $Z = 114$ and $N = 184$. In both cases, the experimental data related to the stability of these nuclei, like half-lives and decay modes, are well in agreement with the theoretical predictions developed and presented in various articles by Sobiczewski *et al.* [2].

2. Search for isotopes of element 120 at SHIP

For the production of isotopes of element 120, the reaction $^{54}\mathrm{Cr} + {}^{248}\mathrm{Cm} \rightarrow {}^{302}120^{*}$ was proposed in November 2010 [12]. The number of 37 scientists from laboratories of various countries gathered at GSI for contributing to the experiment with experience, equipment and material. Most substantial was the contribution of enriched $^{248}\mathrm{Cm}$ target material by Lawrence Livermore National Laboratory (LLNL) and the digital pulse processing by Oak Ridge National Laboratory (ORNL). The accelerator UNILAC and the separator SHIP and its position-sensitive detector system including the sensitive and well-tested analog signal processing were suggested for delivering the $^{54}\mathrm{Cr}$ beam and for separation and identification of the fusion reaction products, respectively.

Safe operation under experimental conditions was tested in a preparatory experiment in July 2010 [13]. In the reaction $^{48}\mathrm{Ca} + {}^{248}\mathrm{Cm} \rightarrow {}^{296}\mathrm{Lv}^{*}$, previously measured data on the decay of $^{293}\mathrm{Lv}$ and $^{292}\mathrm{Lv}$ [9] were confirmed.

A beam time of 140 days was proposed for the search for an isotope of element 120. It was estimated that within this time, a cross-section limit of 100 fb could be reached. In the first part of the element-120 experiment lasting for 38 days from April 24 to June 1 in 2011, a beam dose of 7.0×10^{18} was reached, which corresponds to a one event cross-section limit of 0.58 pb.

One chain of correlated signals was measured for which a probability of 4×10^{-8} was calculated that this chain occurred by chance. A possible assignment of the signals to a decay chain starting at $^{299}120$ is shown on the left-hand side in Fig. 2. Not shown in the figure are the signals from the implantations of nuclei into the detector, which serve as the start for the measurement of the lifetime of the potential parent nucleus of the chain.

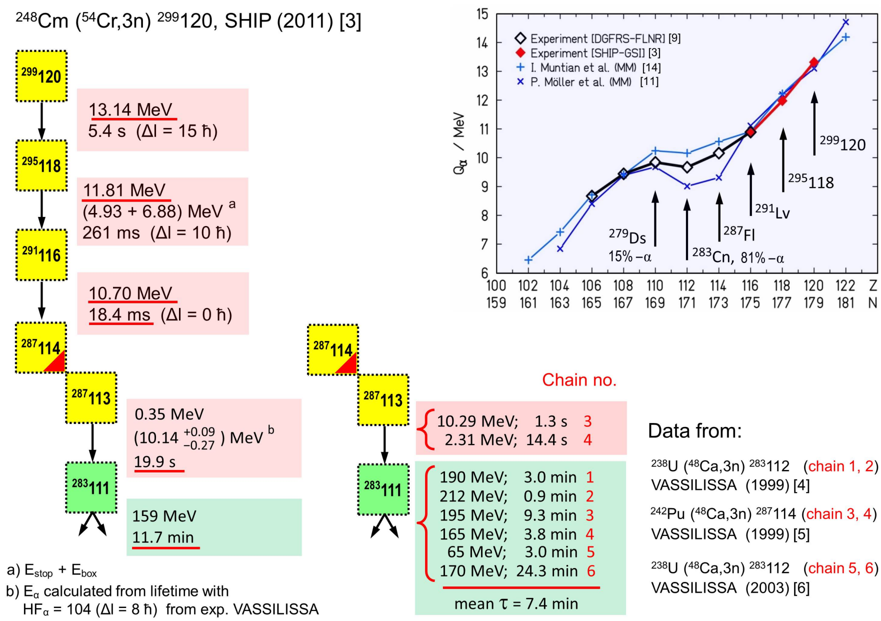

Fig. 2. Properties of the event chain measured at SHIP on May 18, 2011 and its possible tentative assignment (left) [3]. Properties of events measured in [4–6] and tentative re-assignment of the produced isotopes (bottom right). Comparison of measured [9] and calculated [11, 14] Q_α values of the decay chain passing through ^{291}Lv (top right). See [3] for details of the experiment at SHIP and arguments for the suggested assignments and possible re-assignments, respectively.

Arguments for the tentative assignment and a possible re-assignment of previously measured date are discussed in detail in [3]. Although some of the measured data are well in agreement with the given assignments, there exist also uncertainties which prevent definite conclusions. Under these circumstances, a continuation of the experiment would be the best and only solution. However, beam time was not available for this experiment at the UNILAC. Therefore, the known experimental data of even element SHN were reviewed in order to derive information from measured Q_α values on the trend of the binding energies of the nuclei within well-established decay chains. This information was used to extract 'experimental' but model-dependent SCE and related heights of fission barriers which are used in models for calculations of fusion–evaporation cross sections.

3. Review of known data and comparison with predictions

Investigated decay chains of nuclei of even elements are plotted in Fig. 3 in columns 1, 2, 4, 6, and 7. For details of the priority of discovery and of the assignment of the measured data, see review articles [3, 7, 9]. Not considered in the evaluation are the chains in columns 3, 5, and 8. In these cases, additional assumptions as EC or isomeric states are needed in order to place them into the systematics of known data.

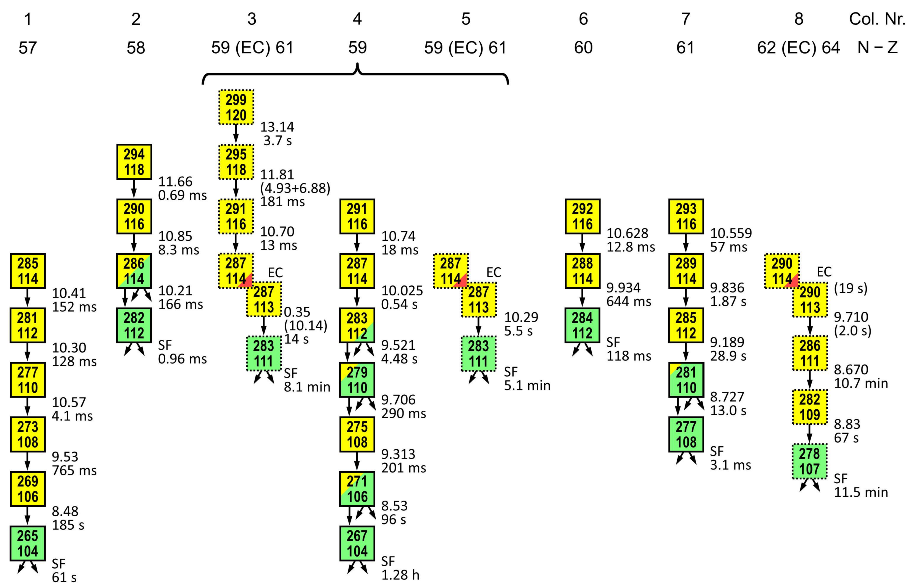

Fig. 3. Known and established α-decay chains of even element SHN are shown in columns 1, 2, 4, 6, and 7. The chains are ordered according to their difference $N - Z$ between neutron and proton number. The data are taken from review articles [3, 7, 9]. For each isotope, the lifetime and for each α emitter, the α energy is given in MeV. For a tentative assignment of the chain shown in column 3 and a tentative re-assignment of the decay chains shown in columns 5 and 8 considering a possible EC branch, see discussion in [3].

3.1. *Relative masses from measured Q_α values*

In a first step, the measured Q_α values were used to determine relative mass values. For none of the nuclei shown in Fig. 3, an experimental mass value is known. Therefore, the estimates of the AME 2012 mass evaluation [15] were used for normalizing the masses of the nuclei at the end of the chains, filled symbols in Fig. 4.

In Fig. 4, the difference of the so-determined relative masses and the predictions of the MM model of Sobiczewski *et al.* (MM-S, upper part) [2] and Möller *et al.* (MM-M, lower part) [11] are plotted. Although both models are based on the same principle — the MM model — significant differences are observed in the region of SHN extending up to values of 4.5 MeV.

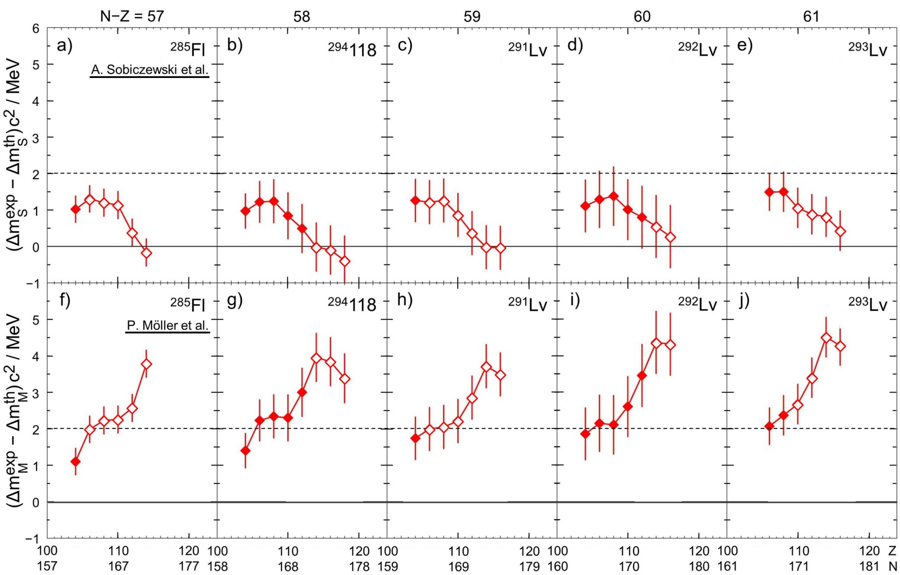

Fig. 4. Differences of experimental and theoretical masses of five neighbouring decay chains of even-element SHN. The data are compared with theoretical masses of the MM-S model in (a)–(e) [2] and with the MM-M model in (f)–(j) [11]. Filled symbols mark differences obtained with extrapolated masses given in the AME 2012 evaluation [15]. These data were used for normalizing the masses of the nuclei at the end of the α-decay chains. Open symbols mark the masses determined from the measured Q_α values [3, 7, 9] relative to the masses at the end of the chains. The horizontal dashed lines at 2 MeV are drawn to guide the eye. The figure was taken from [16].

3.2. Shell-correction energies and fission barriers

The relative masses were used determining trends of SCE and fission barriers. This is achieved subtracting the liquid-drop mass used in the specific model from the experimental mass. The so-determined model-dependent ‘experimental’ SCE are plotted in Fig. 5.

Common in both models is the minimum of SCE at $Z = 114$–116. However, the minimum is more pronounced in MM-M. There, it is in the range from -8.0 to -8.5 MeV, whereas in MM-S, it is at about -6.0 MeV.

The trend of the experimental SCE values normalized to the theoretical masses indicates a slightly stronger decrease from isotopes of element 110 into direction of 114–116 in MM-S, whereas significantly less shell strength is obtained in the comparison with MM-M. For the more neutron-rich chains of N–Z = 60 and 61, the difference amounts to about 2.0 MeV.

Fission barriers are dominantly determined by SCE in the region of SHN. Therefore, it is reasonable to compare the experimental SCE also with the calculated heights of the fission barriers. In fact, −SCE values were often used in calculations of cross sections as approximation to the heights of fission barriers, as long as these were not available. In Fig. 5, the negative values of the heights of fission barriers determined in MM-S and MM-M are also plotted. Only small differences are observed in the case of MM-S, whereas the fission barriers are about 1 MeV higher than the corresponding SCE values in the case of MM-M. Obviously, a pronounced positive saddle-point SCE is responsible for the increase of the heights of the fission barriers.

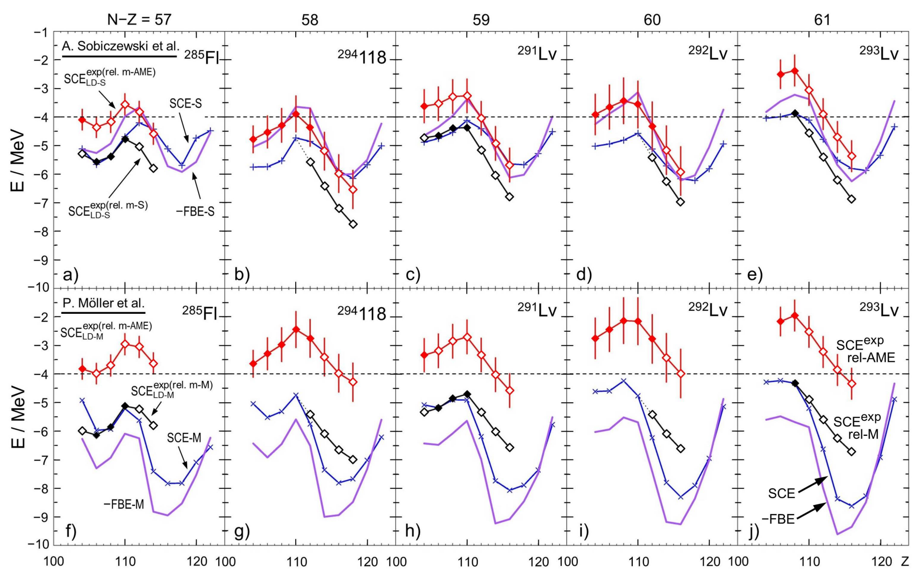

Fig. 5. Experimental model-dependent shell-correction energies of nuclei of five neighbouring decay chains are compared with theoretical values of the MM-S model [2] in (a)–(e) and of the MM-M model [11] in (f)–(j). Experimental data with error bars are based on the AME mass estimates [15], see also Fig. 4, from which the theoretical liquid-drop masses used in MM-S and MM-M, respectively, were subtracted. Experimental data without error bars were normalized to the theoretical masses of the nuclei at the end of the decay chains. Curves without symbols show the negative values of the heights of the fission barriers, −FBE-S [2] and −FBE-M [17], obtained in models MM-S and MM-M. The horizontal dashed lines at −4 MeV are drawn as reference lines to guide the eye. The figure was taken from [16].

3.3. Cross sections

The height of the fission barrier is primarily responsible for the survival of the CN and is, thus, a significant factor contributing to the cross section of the fusion–evaporation reaction. In the case of hot fusion reactions for production of SHN, a rough dependence of the cross section from the height of the fission barrier is given by the rule of thumb that a 1 MeV increase of the fission barrier (of the nucleus in the ground state) increases the cross section by a factor of ten and *vice versa* [18]. Considering the uncertainties of fission barriers suggested by the data shown in Fig. 5, we have to conclude that calculated cross sections will differ by many orders of magnitude when different predictions of fission barriers are used.

In many of these calculations, the values −SCE-M were used as approximation to the height of the fission barrier. This has two reasons. Firstly, the MM-M model is a global model which reproduces well the masses of the known nuclei and predicted well the masses of nuclei which were discovered after its publication, thus giving confidence to the predictive power of this model. Secondly, the binding energies together with their macroscopic and microscopic contributions are well-tabulated for a wide region of nuclei, in particular also for SHN [11], which enables a convenient use of these data in various calculations and comparisons. On the other hand, the parameters of the MM-S model [2] are adjusted to measured data of nuclei located in the limited region of nuclei beyond lead. Consequently, it is expected that the MM-S model will reproduce well the experimental binding energies of SHN, whereas the global MM-M model reveals good results also for lighter nuclei. However, as can be seen in the corresponding graphs presented in [19], this model exhibits larger differences for nuclei in the vicinity of closed shells.

In cross-section calculations for production of isotopes of elements 114 and 116, which used the high fission barriers of MM-M, the experimental data were well-reproduced due to using a correspondingly low fusion probability. If, in reality, as indicated by SCE deduced from experimental Q_α values, the heights of the fission barriers are lower, similar as predicted by MM-S, then the lower survival probability of the CN has to be compensated by an increase of the fusion probability. A rough quantitative estimate for the change of the calculated cross sections for production of isotopes of element 114 and comparison with isotopes of element 120 reveals that the cross sections for production of $^{299}120$ and $^{298}120$ in the reaction $^{54}\mathrm{Cr} + {}^{248}\mathrm{Cm} \to {}^{302}120^*$ could be higher by about a factor of ten than the values of 14 and 28 fb predicted in [20] for the $3n$ and $4n$ channel, respectively. This surprising and, at a first glance, contradictory result that lower fission barriers reveal higher cross sections, is due to the locally pronounced fission barriers for isotopes of elements 114 and 116 of the MM-M model used in calculations [20, 21], which are, as elaborated before, less pronounced in reality demanding for a higher fusion probability.

3.4. Conclusion

The reaction ^{54}Cr+^{248}Cm was experimentally investigated in an attempt to produce isotopes of the new element 120. Correlated α-decay-like signals and an also correlated SF event were observed. The probability is low that this chain of signals was produced by chance. Part of the data are in agreement with previously measured data assigned to decay chains starting at ^{291}Lv and ^{287}Fl, respectively, and with predictions for the α energies of ^{295}Og and ^{299}120. However, uncertainties exist, which hamper a definite assignment.

Experimental data on isotopes of even-element SHN measured in hot fusion reactions since 1998 were reviewed in order to prepare a comprehensive basis for comparison of experimental data among each other and with results of model calculations. Good agreement was observed between experimental data and the results of the MM models of Sobiczewski *et al.* [2] and Möller *et al.* [11] concerning properties of α decay and SF. In particular, the much higher probability for α decay compared to SF of nuclei in the island of SHN is verified.

However, from a detailed analysis of measured Q_α values aiming to extract shell-correction energies and related heights of fission barriers, lower heights for Fl and Lv isotopes were deduced than predicted in [11, 17] where particularly high fission barriers were calculated for isotopes of these two elements. Consequences for cross-section calculations were discussed.

From the very beginning of the GSI research on super-heavy nuclei, professor Adam Sobiczewski was our supporter and friend. Since the mid-1980s, he regularly visited GSI for one or two months usually accompanied by one or two students. In the emerging publications, GSI became his second affiliation. His theoretical predictions were most decisive for the formation of a physical and a chemical research program using the new heavy-ion accelerator UNILAC. Already the very early results on isotopes of the new elements from 107 to 109 produced in cold fusion reactions confirmed his prediction of a region of deformed super-heavy nuclei. Decay properties of isotopes of the elements from 110 to 112 observed at GSI and of one isotope of element 113 observed at RIKEN revealed and confirmed experimentally the centre and the upper end of this remarkable area of nuclear stability. The ongoing experimental research on super-heavy nuclei, since 1998 dominated by experiments performed at FLNR in Dubna using hot fusion reactions, eventually revealed the theoretically predicted island of spherical super-heavy nuclei. A comparison of experimental and theoretical data, the latter obtained in the framework of the macroscopic–microscopic model, demonstrated almost perfect agreement with the results obtained by Adam and his research team.

The valuable, humorous, and always enjoyable discussions with Adam will be remembered forever. For Adam's colleagues and friends, it was always a great pleasure to listen to his presentations and to learn and profit from the discussions with him.

The current article reproduces some text from an article published in *J. Phys. G: Nucl. Part. Phys.* **42**, 114001 (2015). That article provides a wider comparison of experimental work on cold and hot fusion reactions based on targets of ^{208}Pb, ^{209}Bi and isotopes of actinides, respectively.

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