Overview and Perspectives of SHE Research at GSI SHIP

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Abstract Theoretical studies present a quite detailed view of the stability of nuclei in the region of the heaviest elements. Three regions of increased stability exist. Two for deformed nuclei at proton and neutron numbers $Z = 100$ and $N = 152$ and at $Z = 108$ and $N = 162$. The third region is located at $Z = 114$, 120 or 126 and $N = 184$ for spherical nuclei due to closed shells or subshells. Experimentally, the existence of these regions of increased stability is established by synthesis of nuclei in cold fusion reactions using lead or bismuth targets and in hot fusion reactions based on actinide targets. Present experiments are trying to consolidate existing data and to explore the extension of the island of spherical nuclei into the direction of still heavier elements. The present status of experiments at the GSI SHIP is given as well as an outlook on investigations planned for the near future.

1 Introduction and Status of Experiments

For the synthesis of heavy and superheavy nuclei (SHN) fusion-evaporation reactions are used. Two approaches have been successfully employed. Firstly, reactions of a medium mass ion beam impinging on targets of stable lead and bismuth isotopes (cold fusion). These reactions have been successfully applied for producing elements up to $Z = 112$ at the GSI SHIP [1] and to confirm the results of these experiments at RIKEN [2] and LBNL [3]. Recently, a number of neutron deficient odd element isotopes were produced in a combination with ^{208}Pb targets and odd element projectiles at LBNL [4, 5]. Using a ^{209}Bi target the isotope ^{278}113 was synthesized at RIKEN [6].

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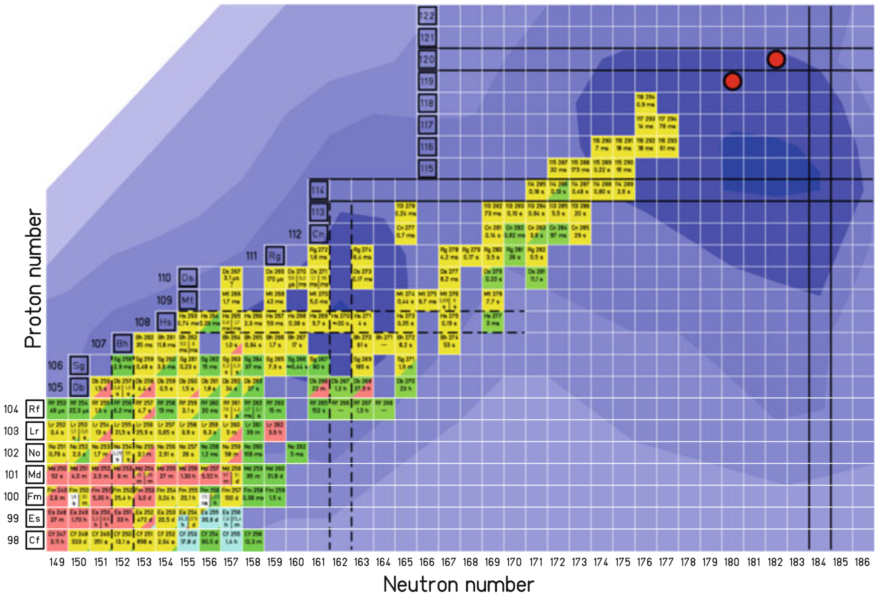

Fig. 1 *Upper end* of the chart of nuclei showing the presently (2012) known isotopes. For each known nucleus the element name, mass number, and half-life are given. The magic numbers for the protons at element 114 and 120 and for the neutrons at $N = 184$ are emphasized. The *bold dashed lines* mark proton number 108 and neutron numbers 152 and 162. Nuclei with that number of protons or neutrons have increased stability. However, they are deformed contrary to the spherical superheavy nuclei. In the region of the crossing between *bold* and *dashed lines* at $Z = 114$ and $N = 162$ it is uncertain, whether nuclei there are deformed or spherical. The *red dots* mark compound nuclei which can be formed in the reactions ^{50}Ti +^{249}Bk or ^{51}V +^{248}Cm (^{299}119*) and ^{54}Cr +^{248}Cm (^{302}120*). The background structure shows the calculated shell correction energy according to the macroscopic-microscopic model [16, 17]

Heavier isotopes of the element copernicium and new elements up to Z = 118 were produced in reactions with beams of ^{48}Ca and radioactive actinide targets (hot fusion) at FLNR [7, 8]. The results of four of these reactions, ^{48}Ca+^{242}Pu [9–11], ^{48}Ca+^{238}U [12], ^{48}Ca+^{244}Pu [13], and ^{48}Ca+^{248}Cm [14] were confirmed in independent experiments. A new isotope of element 114, ^{285}114 and its decay daughters, was synthesized by evaporation of five neutrons in the reaction ^{48}Ca+^{242}Pu at LBNL [15]. Figure 1 summarizes the data as they are presently known.

Element 112, 'copernicium', is presently the last element in the Periodic Table, which has received a name. Agreement between element-112 data of the GSI-SHIP work and the confirmation experiments was stated in a IUPAC Technical Report in 2009 [18]. Similarly, IUPAC has assigned priority of the discovery of elements 114 and 116 to the Dubna-Livermore group [19]. The names proposed by this group are 'flerovium' and 'livermorium', respectively [20].

Besides the insight that nuclei with such a high number of protons and resulting extremely high repulsive Coulomb forces are existing, two more important observations emerged. Firstly, the expectation that half-lives of the new isotopes should lengthen with increasing neutron number as one approaches the island of stability seems to be fulfilled. Secondly, the measured cross-sections for the relevant nu-clear fusion processes reach values of up to 10 pb, which is surprisingly high. Fur-thermore, the cross-sections seem to be correlated with the variation of shell-correction energies as predicted by macroscopic-microscopic calculations [16, 17, 21].

2 Continuation of SHN Experiments Using ^{248}Cm Targets

A comparison of various theoretical studies reveals that the location of the next closed proton shell beyond $Z = 82$ is uncertain. The question is still open whether $Z = 120$ is a closed proton shell or if strong shell closures exist at $Z = 114$ or 126. In addition, the possibility has to be considered that the island of superheavy nuclei is relatively flat and extents between sub-shells at 114, 120 and 126. Concerning the closed neutron shell, most theories agree with $N = 184$ as a strong shell. Experimental data—longer half-lives and decreasing negative shell-correction energies with increasing neutron number—as known so far, are in agreement with this finding, too.

As an important part of our work on the synthesis and properties of SHN we proposed to study also hot fusion reactions based on actinide targets, in addition to our cold-fusion program. Together with several technical improvements this proposal was made in a medium range plan already at the end 1998 [22]. However, at the beginning of 1999 our report was rejected and the proposed program was no longer pursued.

Now, times have changed, and in 2009 we suggested to start a program for studying superheavy nuclei using reactions based on ^{248}Cm targets. This target material has special properties which makes it favorable for the synthesis of heavy nuclei. It is one of the heaviest ($Z = 96$) and most neutron rich available targets. Increased shell effects at its neutron number $N = 152$ result in a relatively long half-life of 3.4×10^5 years and, thus, low specific activity. In combination with strongly bound projectile nuclei like ^{48}Ca or the neutron rich isotopes of the heavier elements up to nickel, relatively low excitation energies of the compound nuclei result, which are approximately 30–40 MeV at the fusion barrier. This advantageous property increases the probability for neutron emission instead of fission and thus results in relatively high fusion-evaporation cross-sections.

In the following, we list a number of general arguments which have to be considered selecting the best reaction with respect to cross-sections for production of new elements beyond 118, in particular the new elements 119 and 120.

Production cross-sections are strongly determined by fission barriers which again are built by shell effects in the region of SHN. The rising up of cross-sections to several picobarns for elements 114 and 116 is due to increasing shell effects when $N = 184$ is approached. This behaviour suggests using the most neutron rich projectile and target nuclei available for synthesis of the elements 119 and 120.

Shell effects and thus fission barriers are considerably reduced with increasing excitation energy of the compound nucleus. Therefore, selection of a reaction re-sulting in a minimum of excitation energy is mandatory.

Cross-sections are further strongly influenced by Coulomb re-separation in the entrance channel of the reaction due to quasi-elastic and quasi-fission processes. In order to reduce this most unwanted effect, reaction partners have to be used resulting in lowest repulsive Coulomb forces. This can be achieved using reaction partners of high asymmetry in the number of protons.

Other phenomena which also influence the cross-sections, but are difficult to predict quantitatively and in detail, originate from isotopic effects. The number of neutrons determines the nuclear radius, relatively more for the smaller projectiles, which influences the compactness of the system at the contact configuration. In the case of deformed nuclei of the actinides, nuclear orientation is another property, which strongly determines cross-section and beam energy. However, it is not possible to align the target nuclei in order to obtain an orientation which results in highest fusion probability.

Finally, the reaction must be technically possible, i.e. projectiles and targets have to be available. Heaviest isotopes which could be used as targets are ^{254}Es ($T_{1/2}$ = 276 d) and ^{257}Fm (100 d). However, the production of these isotopes is complex and only amounts of micrograms and nanograms, respectively, can be produced at high costs.

The next lighter isotopes available in principle are ^{252}Cf (2.6 y), ^{249}Cf (351 y), and ^{249}Bk (320 d). The isotope ^{252}Cf can be handled only with special radiation protection because of the high neutron flux being emitted from this fissioning material. The isotope ^{249}Bk has a relatively short half-life. It must be produced on demand and it is not available regularly. Due to the relatively short half-life, also the isotope ^{249}Cf has a high specific activity. In addition, the compound nucleus ^{299}120, which can be made in reactions with a ^{50}Ti beam, has three neutrons less then the compound nucleus ^{302}120, which can be produced with a ^{248}Cm target and a ^{54}Cr beam, see Fig. 1. Considering all pros and cons we conclude that the reaction ^{54}Cr $+^{248}$Cm $\rightarrow^{302}$120* is presently the most promising one being technically feasible to search for element 120.

Two reactions are available for the synthesis of element 119, ^{50}Ti $+^{249}$Bk and ^{51}V $+^{248}$Cm, both forming the compound nucleus ^{299}119, see Fig. 1. In this special case one can expect that the cross-section is independent whether the odd proton is located in the target or in the projectile. Already existing data on the synthesis of odd element isotopes using ^{208}Pb or ^{209}Bi targets and the corresponding odd or even element projectiles revealed similar cross-sections when the same compound nucleus is formed. A comparison was performed in [23] using experimental data measured at SHIP and at the LBNL gas-filled separator BGS.

Finally, the target ^{249}Bk opens an interesting aspect from the reaction point of view, apart from the difficulty with the production of the material and its short half-life. Using a ^{51}V beam the compound nucleus ^{300}120 can be formed. In this case the odd proton is expected to be transferred to the target in an early stage of the reaction, so that the cross-section could be similar as in the case of the reaction

^{50}Ti $+^{250}$Cf, also resulting in the compound nucleus ^{300}120. The homologous cold fusion reactions were not yet studied. The pairs of target nuclei ^{205}Tl, ^{206}Pb or ^{203}Tl, ^{204}Pb and beams of ^{51}V and ^{50}Ti, respectively, could be used supplementary to the reactions ^{208}Pb $+^{52}$Cr and ^{209}Bi $+^{51}$V.

Cross-sections calculated by various authors for the synthesis of elements 119 and 120 using ^{50}Ti or ^{54}Cr beams cover a range from 0.1 to 800 fb [24–28]. The reason for this large uncertainty is the extremely sensitive dependence of cross-sections from fusion barriers and resulting excitation energies at the barrier, from Coulomb re-separation and from fission barriers as outlined before. The model by Siwek-Wilczynska et al. [24] assumes a lower fusion barrier which results in an increase of the 3n cross-section. In the model by Nasirov et al. [25, 26] the quasi-fission processes result in strong reduction of cross-sections with increasing symmetry, whereas this effect changes the cross-section only within a factor of ten in the model by Zagrebaev and Greiner [27]. Finally, in the paper by Adamian et al. [28] various mass formula and various damping parameters of the fission barrier at increasing excitation energy were compared. Two of the results predict cross-sections differing by two and three orders of magnitude.

Experimental limits were obtained for reactions with ^{58}Fe and ^{64}Ni beams and targets of ^{244}Pu and ^{238}U at FLNR [29] and at SHIP [30], respectively. Although these limits are still high, they allow to reject unusually high fission barriers at element 120. Using the rule of thumb that a 1 MeV change of the fission barrier changes the cross-section by one order of magnitude at least [24], we obtain experimental limits for the fission barrier of element 120 isotopes of <8.9 and <8.3 MeV, respectively. As a starting point in this estimate we used the calculation of Zagrebaev and Greiner [27], who determined their cross-sections with a fission barrier of 7 MeV. At a fission barrier of 8.3 MeV their cross-section estimates would be a factor of 20 higher. This simple consideration and the very different predictions show that a sufficiently accurate estimate for the beam time necessary to produce the elements 119 and 120 cannot be made on the basis of the presently existing data and calculations.

Half-life and decay mode are nuclear properties which could hamper the identification, although isotopes of element 120 could be produced with high enough cross-sections. Theoretical calculations show that the heaviest elements decay by α emission. This result is proved by experimental data on elements up to ^{294}118, which has a measured Q_α value of 11.81 MeV and decays with a half-life of 890 μs [7]. In the region of interest, β decay and spontaneous fission are predicted to have significantly longer half-lives. This result is in agreement with the measured α-decay chains which end by spontaneous fission only at copernicium or below.

Whereas fission barriers and deduced fission half-lives are difficult to calculate, the access to Q_α values as difference of masses of neighbouring nuclei and deduced partial α half-lives is easier. In the following we compare experimental Q_α values of an established decay chain with few but representative theoretical predictions. In Fig. 2, calculated Q_α values are shown over a wide range from element 104 to 122 for the chain passing ^{292}116. Showing this figure, we are also aiming to obtain a sense for the uncertainties related to predictions on the stability of isotopes of the so far

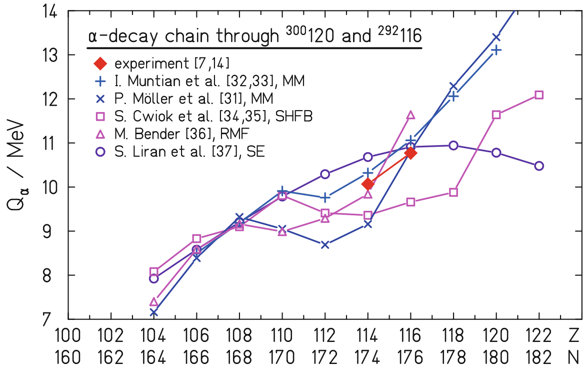

Fig. 2 Comparison of measured and calculated Q_α values of the α-decay chain passing the isotope $^{292}116$. Nuclei of this decay chain belong to the most neutron rich nuclei which can be produced in the laboratory. They are of special interest with respect to a future synthesis of so far unknown elements beyond $Z = 118$

unknown elements 119 and 120, their synthesis is presently the aim at the research centres JINR, RIKEN, and GSI.

Two of the theoretical data shown are based on the macroscopic-microscopic (MM) model [31–33], one on the self-consistent mean field model using the Skyrme-Hartree-Fock-Bogoliubov (SHFB) method [34, 35], one on the relativistic mean-field (RMF) model [36], and one on a semiempirical (SE) shell-model mass equation having Z = 126 and N = 184 as spherical proton and neutron shells after the double magic ^{208}Pb [37].

Obviously, the considered range of Q_α values can be subdivided in three parts concerning the variations of the predictions. One for elements below darmstad-tium, one for elements between darmstadtium and Z = 116, and a third one for elements up to 122.

The three regions are also related to different physical properties of the nuclei. Firstly, the region of well deformed nuclei below darmstadtium and $N < 170$; in this region the shape of the nuclei is determined by stronger binding energy at large deformation due to the compression of single particle levels below the ener-gy gaps at $Z = 108$ and $N = 162$ at quadrupole deformation parameters $\beta_2 = 0.25$. The second region up to element 116 for neutron numbers of the measured α-decay chain considered here is a transitional region of decreasing deformation into the direction of the third region extending up to element 122 and beyond, which is governed by shell effects of spherical closed shells or subshells.

Although the experimental Q_α values are scarce we notice that the gradient of the experimental data between elements 114 and 116 is less than in the results of the MM model [31] and the RMF model [36]. From this experimental observation we conclude that at neutron numbers 174 to 176 the proton shell strength at $Z = 114$ is less pronounced than predicted in [31, 36].

Concerning heavier elements beyond $Z = 118$, the experimental data is just at the limit which could settle the question if proton shells exist at $Z = 120$ or 126. Increasing Q_α values as predicted by the MM models would rule out shell closures at 120 and 126. As a consequence, the lifetimes of elements beyond 120 would fall below 1 μs which is the limit of present detection methods. The elements 119 and 120 would be the last ones which could be detected in the near future. At $Z = 120$ the 1 μs limit is reached at $Q_\alpha = 13.3$ MeV and at $Z = 126$ at 14.0 MeV.

A subshell closure at $Z = 120$ would result in relatively long α half-lives of element 120. At a Q_α value of about 11.7 MeV calculated for $^{300}120$ in [35], see Fig. 2, we obtain a half-life of 2.2 ms. In addition, also the α half-life of element 122 would be longer relative to the predictions of the MM models. The stronger trend to lower Q_α values of the semiempirical model would result in α half-lives of 350 ms and 43 s at $Q_\alpha = 10.8$ and 10.7 MeV [37] for isotopes of element 120 and 126 with mass numbers 300 and 310, respectively.

In the region of SHEs, fission barriers are mainly determined by ground-state shell effects. Because Q_α values are determined by the difference of binding energies between parent and daughter nucleus, the gradient of a Q_α systematics reflects the trend of increasing or decreasing fission barriers. The rapidly increasing Q_α values of the MM models for elements above 114 is related to increasing negative ground-state shell-correction energies and thus decreasing fission barriers. The opposite trend is valid for the semiempirical model.

The experimental Q_α values reveal differences to the theoretical data of up to 1 MeV, see Fig. 2. Similar differences must be expected for the ground-state shell-correction energies and the fission barriers. Fission barriers are an essential part in the calculations of cross-sections. As already reminded, a rough estimate shows that a 1 MeV increase of the fission barrier increases the cross-section by one to two orders of magnitude [24]. Uncertainties of this order of magnitude, which were revealed by the comparison of experimental and theoretical Q_α values, have to be considered in the discussions on the preparation of experiments aiming at searching for new elements. In other words, sufficiently long beam times have to be provided in order to perform experiments with the perspectives of being successful.

In conclusion we realize that half-lives of the isotopes of interest are predicted to be in the range from 1 to 30 μs, but could be significantly longer if the proton shell is at $Z = 126$ or 120 and not at 114. In any case special technical preparations are needed for detection of isotopes of element 120 in order to be prepared for short half-lives [43].

At SHIP, a minimum lifetime of 2 μs is needed so that the residues can pass through the separator, otherwise they will decay inside the separator. In this case, also the daughter nucleus after α decay will be lost with high probability due to the recoil momentum from the emitted α particle, an effect which reduces the transmission by a factor of ten.

The decay chains expected in the case of three and four neutron evaporation will populate isotopes of element 116, $^{290}116$ and $^{291}116$, which were measured previously at FLNR and which were confirmed indirectly by identification of the daughter nuclei $^{286}114$ and $^{287}114$ at LBNL [11]. In this case a well founded identification

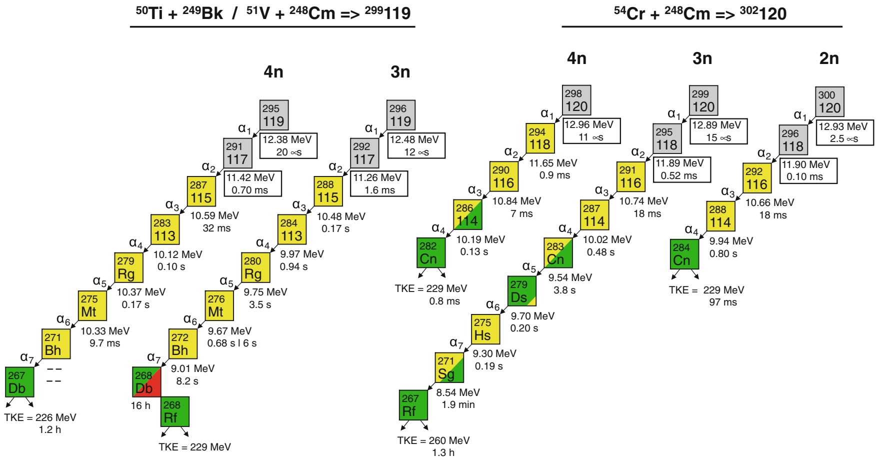

Fig. 3 Expected decay chains of element 119 populated in the reactions ^{50}Ti +^{249}Bk and ^{51}V +^{248}Cm, both forming the compound nucleus ^{299}119* and of element 120 formed in the reaction ^{54}Cr +^{248}Cm $\rightarrow^{302}$ 120*. Predicted decay data of so far unknown isotopes are given in frames. Values for E_α of ^{295,296}119, ^{291,292}117, ^{300}120, and ^{296}118 were taken from [38]. The half-lives of these isotopes were calculated using the WKB method [39]. Energies and half-lives of ^{298,299}120 and ^{295}118 were taken from [40]. Concerning the uncertainty of the calculated E_α values see also Fig. 2. Experimental data of the known isotopes were taken from publications of the Dubna-Livermore group, namely for the decay chain of ^{287}115 from [41], ^{288}115 from [42], ^{294}118, ^{291}116, and ^{292}116 from [7]. TKE values of the spontaneously fissioning isotopes terminating the chains were taken from Fig. 29 in [7]. Note the significant difference of the lengths of the decay chains for even-even and odd or odd-odd nuclei

of element 120 by genetic correlation to known nuclei is given. The expected decay chains are shown in Fig. 3. In the case of an also possible two neutron channel, we would observe as a granddaughter the isotope ^{292}116 and its daughter decays, which we observed in a confirmation experiment at SHIP using the reaction ^{48}Ca +^{248}Cm $\rightarrow^{292}$ 116 + 4n [14]. Similarly, the decay chains of ^{295}119 and ^{296}119 produced in 4n and 3n evaporation channels, respectively, will populate known decay chains after the first two α decays, see Fig. 3, left side.

A new search for element 120 using the reaction ^{54}Cr +^{248}Cm $\rightarrow^{302}$120* has been started at SHIP in spring 2011. Main aim of this first part of 33 days was to study the performance of the targets during irradiation with a chromium beam and to condition a second wheel for further irradiation in the future. Therefore, the beam current was limited to about 400 particle nA. Nevertheless, a cross-section limit of 560 fb was reached, which is, however, still far from calculated cross-sections being mainly in the range from 30 to 100 fb. An estimate based on higher beam intensities results in an additional measuring time of about three months in order to reach such low cross-section limits. From the performance of the accelerator, the targets, the separator, the detectors, and the data acquisition system in the first part of the experiment we conclude that our experiment is well prepared for this important next step in the study of superheavy nuclei.

3 Conclusion and Outlook

The experimental work of the last three decades has shown that cross-sections for the synthesis of the heaviest elements do not decrease continuously as it was measured up to the production of element 113 using cold fusion reactions. Recent data on the synthesis of elements 112 to 118 in Dubna using hot fusion show that this trend is broken when the region of spherical SHN is reached. Some of the results originally obtained in Dubna were confirmed in independent experiments and with different methods, including the use of chemical specific properties of the elements. We conclude that the region of the predicted spherical SHN has finally been reached and the exploration of the 'island' has started and can be performed even on a relatively high cross-section level.

An opportunity for the continuation of experiments in the region of SHN at low cross-sections afford, among others, further accelerator developments. High current beams and radioactive beams are options for the future. A wide range of half-lives encourages the application of a wide variety of experimental methods in the investigation of SHN, from the safe identification of short lived isotopes by recoil-separation techniques to atomic physics experiments on trapped ions, and to the investigation of chemical properties of SHN using long-lived isotopes.

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