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New results on elements 111 and 112

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This paper is dedicated to our former department leader P. Armbruster on the occasion of his 70th birthday

Abstract. Experiments on the synthesis and identification of the nuclei ^{272}111 and ^{277}112 were performed in order to confirm previous results. Three additional decay chains were measured in the reaction ^{64}Ni + ^{209}Bi → ^{273}111*. The study revealed considerably improved data on the decay chain originating from ^{272}111. One additional chain was measured in the reaction ^{70}Zn + ^{208}Pb → ^{278}112*. The decay properties of the chain starting at ^{277}112 are in excellent agreement with the second chain of the first experiment down to ^{265}Sg, where the new chain ends by a previously unknown spontaneous-fission branch. A re-analysis of all the data on elements 110, 111, and 112 measured at GSI since 1994 (a total of 34 decay chains was investigated) revealed that for 2 chains (the second chain of ^{269}110 measured in 1994 and the first chain of ^{277}112 measured in 1996) the results of the new analysis differed from the previous one. In all other cases the earlier data are exactly reproduced.

PACS. 23.60.+e Alpha decay – 25.70.-z Low and intermediate energy heavy-ion reactions – 25.85.Ca Spontaneous fission – 27.90.+b $A \geq 220$

1 Introduction

In order for the discovery of a new element to be recognized, criteria that must be satisfied were established by the 1992-report of IUPAC's Transfermium Working Group (TWG) [1]. These criteria served as a guide for the new report of IUPAC/IUPAP's Joint Working Party (JWP) [2] which treated the discovery of the elements 110, 111, and 112. These reports will probably also be the basis for the work of similar committees in the future.

One criterion of more general nature is the reproducibility of an experimental result, and the TWG suggested that no new element should be officially recognized until the data have been reproduced. However, a need for repetition could be waived in those cases where the data are of such nature that no reasonable doubt is possible and a repetition of the experiment would imply an unreasonable burden.

Concerning our results on elements 110, 111, and 112 obtained at the GSI UNILAC in Darmstadt, the JWP report concluded that the element 110 was convincingly identified in the 1994 SHIP experiment [3,4]. However, when the same criteria were applied to the results for elements 111 and 112 [5,6], which we identified by three and two decay chains, respectively, internal redundancy was found to be insufficient to warrant certitude of identification.

The cross-section for the synthesis of ^{272}111 was 3.5 pb and 1.0 pb for ^{277}112. The latter value allows for detection of one nucleus per week using the presently available techniques. The half-lives are within a range of a few hundred microseconds which is ideal for detection using recoil separators. Therefore, it seemed to us not to be an unreasonable burden to repeat the two experiments in order to certify the previously measured data.

2 Experimental procedure

We performed the measurements aiming at new data on the synthesis and the decay of ^{277}112 and ^{272}111 in the

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Table 1. Summary of previous and present irradiations performed to synthesize elements 111 and 112. $E_{\rm proj}$ is the beam energy in front of the target, E^* is the calculated excitation energy of the compound nucleus for reactions in the middle of the target thickness. The compilation of ref. [7] was used for projectile and target mass and the calculations of ref. [8] for the mass of the compound nucleus. The "observed events" are decay chains which were assigned to the one-neutron evaporation channel by genetic correlation to known daughter nuclei. Also given are the results from short irradiations of a ^{207}Pb target with ^{64}Ni [9] after the element 111 run and of a UF_4 target with ^{70}Zn after the element 112 run.

Date	Time (day)	Target isotope	Thickness (μg/cm^2)	Beam isotope	$E_{\rm proj}$ (MeV)	Compound nucleus	E^* (MeV)	Ion dose (10^{18})	Observed events	σ (pb)
01.12.–06.12.1994	5.0	^{209}Bi	450	^{64}Ni	316.1	^{273}111	9.4	1.0	0	< 2.9
06.12.–12.12.1994	5.8				318.1		11.0	1.1	1	$1.7^{+3.3}_{-1.4}$
12.12.–18.12.1994	5.9				320.0		12.5	1.1	2	$3.5^{+4.6}_{-2.3}$
16.10.–29.10.2000	13				320.0		12.5	2.2	3	$2.5^{+2.5}_{-1.4}$
29.10.–06.11.2000	7.3	^{207}Pb	435		317.0	^{271}110	14.0	1.3	8	13 ± 5
26.01.–18.02.1996	24	^{208}Pb	450	^{70}Zn	343.8	^{278}112	10.1	3.4	1[a)]	$0.5^{+1.1}_{-0.4}$
03.05.–22.05.2000	19		450		346.1		12.0	3.5	1	$0.5^{+1.1}_{-0.4}$
22.05.–29.05.2000	7		426		343.8		10.1	1.2	0	< 2.6
29.05.–01.06.2000	3	$^{238}UF_4$	305		370.3	^{308}122	18.6	0.6	0	< 7.2

[a)] See sect. 2.

year 2000. The irradiations took place from May 3–29 and October 16–29, respectively. The reactions were the same as in our first experiments, ^{70}Zn + ^{208}Pb $\rightarrow$ ^{278}112* and ^{64}Ni + ^{209}Bi $\rightarrow$ ^{273}111*. A summary of the experimental conditions is given in table 1. First results from the $Z = 112$ experiment were already published in ref. [10].

The nuclei were identified by position and time correlation analysis which allows to establish genetic relations of the nuclei within a decay chain. The data were measured by using position-sensitive Si detectors. Details of the experimental set-up and the analysis procedure are given elsewhere [4,10–12].

The targets were prepared by evaporation of lead or bismuth deposited in layers of 450 μg/cm^2 on carbon backing foils of 40 μg/cm^2. The target layers were then covered by evaporation of a 10 μg/cm^2 thick carbon layer to protect the targets from sputtering and to enhance radiative cooling. At a distance of 16 cm behind the target, a carbon foil of 60 μg/cm^2 thickness was mounted for equilibration of the ionic-charge state. Before the reaction products were implanted into the Si detector, they passed through three time-of-flight detectors each consisting of two carbon foils. The total thickness of these carbon foils amounted to 190 μg/cm^2.

In extension of our electronic set-up, we installed a circuit for the $Z = 111$ run which allowed to switch off the beam within 50 μs after an implanted residue was detected by coincidence of energy and time-of-flight signal. In a subsequent time window of 10 ms, a preset number of α-particles (in this experiment one) was required which then prolonged the beam-off period up to the expected measurable end of the decay chain. In our experiment 10 minutes were chosen, thus making provision for the detection of a possible α decay of ^{252}Md, $T_{1/2} = 2.3$ min. This improvement considerably reduced the background during the period of α decays and allowed for the safe detection of signals from long-lived decays. The circuit was prepared already in May for the $Z = 112$ experiment, but not yet used since the trigger conditions could not be set properly, mainly due to the use of degrader foils in front of the Si detector. No degrader foils were used in the $Z = 111$ experiment.

Behind the stop detector we mounted a second Si detector. This detector was of the same type as the stop detector, however, the detector strips were galvanically connected so that three energy sensitive segments were formed. In the analysis, the signals from the detector were used to identify and reject protons of about 11 MeV energy. The protons leave in the stop detector (thickness 300 μm) energy loss signals of about 1.9 MeV with a distribution of about 0.5 MeV width (FWHM). This is the same range in which also the signals from escaping α-particles are measured. Therefore, an efficient suppression of the protons reduces considerably the probability for chance correlations in those cases, when escaping α-particles are not stopped in the back detectors and the device for switching off the beam cannot be used.

We suppose that the protons are emitted from reactions with carbon nuclei of the target backing, cover foil and/or the charge equilibration foil. The protons have a high magnetic rigidity, and a part of them passes SHIP and is focused in the right-hand side of the detector (see fig. 3 in ref. [4]). At beam currents of 2.0×10^{12} /s, the counting rate of these protons is 0.4 /s.

Preceding the main irradiation, we tested the experimental set-up using reactions of higher cross-section. In the case of the element 111 we chose the reaction ^{64}Ni + ^{208}Pb, which has a cross-section for the 1n channel

of 15 pb at $E^* = 12.1$ MeV. During 3 days we measured 3 decay chains of ^{271}110, which were in agreement with the previous result [4]. In one case we even observed the α decay of ^{255}Md at the end of the chain. The nucleus ^{255}Md has an α branching of 8%. It is produced by electron capture of ^{255}No. Our measured α energy was 7.32 MeV and the lifetime 19 min, both values are in agreement with the literature data for the decay of that nucleus [13].

During the same irradiation period we used the ^{64}Ni beam also for an investigation of the even-even nucleus ^{270}110 using a ^{207}Pb target (see table 1). The results obtained were published in ref. [9]. From that data we received useful information for the comparison of the ^{277}112 decay chain with the results from theoretical studies (see subsect. 4.2).

In the case of the element 112 run (^{70}Zn beam) we used, for testing purposes, a ^{50}Ti beam and chemical compounds of ^{208}Pb as targets. Subsequent to the main experiment we used the ^{70}Zn beam also to irradiate a $^{238}UF_4$ target (see table 1). In that reaction the compound nucleus would be $^{308}_{186}$122. This nuclide is located near the predicted closed neutron shell $N = 184$, where shell effects could result in relatively high cross-sections. In that irradiation, however, we did not observe any decay chain which could be attributed to a superheavy element. The upper cross-section limit obtained was 7.2 pb.

In order to prove consistency of the results from the earlier analysis and the presently used one, we also reanalyzed all our data measured since 1994. In the course of that work we reviewed a total of 34 decay chains, four of ^{269}110, eight of ^{270}110, thirteen of ^{271}110, six of ^{272}111 and three of ^{277}112. In two cases (the second chain of ^{269}110 measured in 1994 and the first chain of ^{277}112 measured in 1996) we found inconsistency between the original raw data, stored in binary files on magnetic tape (which were used for re-analysis), and the event-by-event text files (which were used at the time as the basis for the assignment). For reasons not yet known to us the contents of these text files had been modified for the case of the two events so that event chains were spuriously created.

The data, which were assigned to the second event chain of ^{269}110 at the time, were taken from a text file. These data consisted of an implanted residue succeeded by three α decays within 375 ms [3]. However, the data which were re-analyzed from the binary raw data file, revealed only one α decay with $E_\alpha = 10.53$ MeV (this is the same energy as α_2 of the earlier analysis) subsequent to an implanted nucleus after 20 μs (the electronic dead time is 15 μs), but not after 201 μs as in the case of α_1 of the chain.

The first decay chain assigned to ^{277}112 consisted of five α decays subsequent to an implanted nucleus [6]. The new analysis of the binary raw data file revealed agreement with the 1996 event-by-event text file only in the case of the event α_1, an α decay with 11.65 MeV energy. It was preceded by an implanted nucleus at 11 s (but not at 400 μs as deduced in 1996). This sequence of two correlated events fits perfectly to the transfer product ^{212}Po.

An explanation for the disagreement between the original raw data (fully consistent with the present re-analysis) and the extracted text file data (showing the disagreement just discussed) on the basis of errors in the computer program was ruled out, because all neighboring single events and all other decay chains measured at the time were exactly reproduced by the new analysis. We cannot exclude, for example, human failure in the analysis of these two events. But to reiterate: all other events, a total of 32 decay chains, are exactly reproduced in the re-analysis.

3 Element 111

3.1 Results

The new results on the decay of ^{272}111 are in full agreement with the data measured previously. In our first experiment in 1994 we studied the reaction $^{64}\mathrm{Ni} + {}^{209}\mathrm{Bi} \rightarrow {}^{273}111^*$ at three different beam energies. The energy values and the results of the experiment are listed in table 1.

The cross-section maximum could not be definitely determined from the data due to the large statistical error bars and few number of data points. However, taking into account the trend set by the previously measured excitation functions for the synthesis of nuclei of lighter elements [4], we concluded that the maximum yield should be close to an excitation energy of 12 MeV. In order to improve the statistical accuracy at that energy, we chose the already measured data point at 320 MeV beam energy also in the confirmation experiment.

During an irradiation time of 13 days we collected a beam dose of 2.2×10^{18} ions. A total of 3 decay chains was measured from which, in agreement with the first result, a cross-section of 2.5 pb was deduced. The efficiency of SHIP was estimated to be 40%, the same value was used in our first experiment. The mean cross-section value from both experiments at 320 MeV beam energy (5 events at 3.3×10^{18} projectiles) is $(2.9^{+1.9}_{-1.3})$ pb.

The newly measured decay chains are plotted in fig. 1. The decay data of the three new chains are compared with those from the first experiment and with literature data on ^{260}Db and ^{256}Lr [13–15] in fig. 2. The results are discussed in the following.

3.2 Discussion

The trigger for the switching off the beam worked properly. All three chains were measured in full length during beam-off periods. The 10 minutes beam-off trigger was activated only in one case without detection of a decay chain.

The implantation energies are grouped around the mean value of 39.2 MeV. The individual energy values are given in fig. 1. The signals were calibrated using a fraction of projectiles which hit the detector due to low ionic-charge states. No corrections for nuclear stopping or recombination effects were applied.

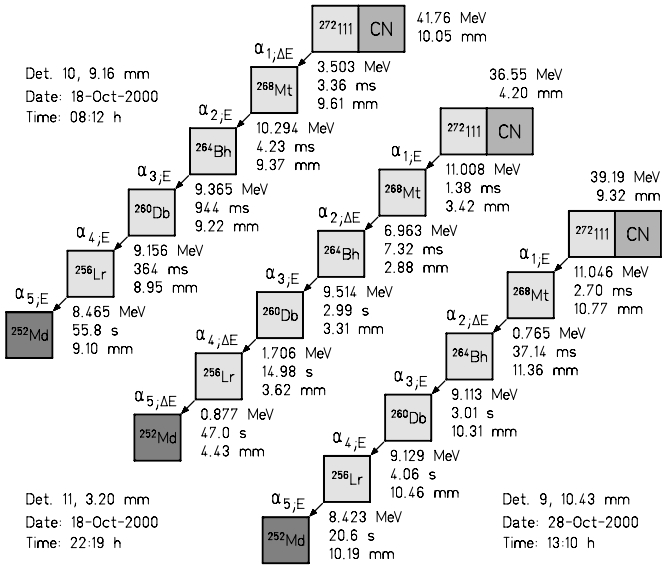

Fig. 1. Three decay chains and decay data measured during an experiment aiming at the confirmation of element 111.

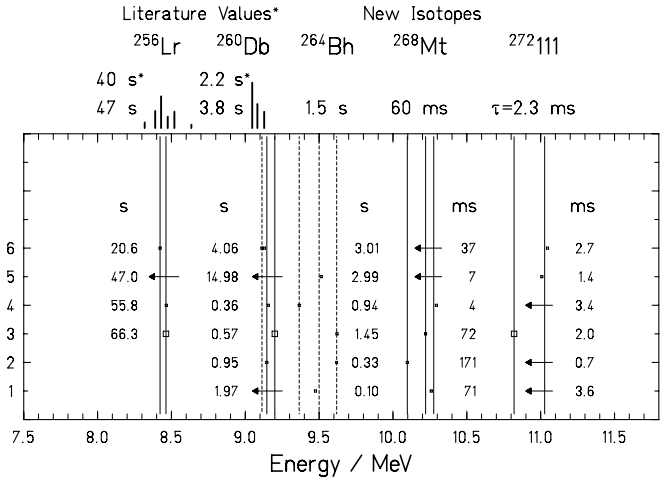

Fig. 2. Comparison of α-decay data from the six events which were assigned to the decay of ^{272}111. The event chains are chronologically ordered and numbered from 1 to 6. The size of the data points reflects the detector resolution, small dots stand for α's stopped in the main detector, larger squares for escape α's stopped in the back detectors and arrows for escape α's delivering only a ΔE signal from the main detector. Vertical lines are drawn at energies of single data points or at the mean energy values of decays which have the same energy within the detector resolution. Data points assigned to ^{264}Bh are marked by dashed lines for better distinction from the ^{260}Db data. Also given are the individual and mean values of lifetimes (not half-lives). Above the upper abscissa the α spectra and lifetimes deduced from the literature (marked by an asterisk) are plotted for the decays of ^{260}Db and ^{256}Lr.

We compare now the measured energies with those calculated for a fusion process, taking into account the energy loss in the target, in the various carbon foils and in a detector dead layer of 11.6 μg/cm^2 Si. For the two extreme cases of reactions at the front side and at the end of the target, we get kinetic energies of the evaporation residues of 54.5 and 58.7 MeV, respectively, when they enter the active detector material. The energy losses of projectiles and the heavy residues were calculated using the computer program SRIM-2000 [16]. For the residues the extrapolation $\mathrm{d}E/\mathrm{d}x(^{272}111) = \mathrm{d}E/\mathrm{d}x(^{238}\mathrm{U})$ at the same E/A-value as the residue was applied, because uranium is the heaviest element included in SRIM.

The difference of the mean values of implantation energies from measurement and calculation is 17.4 MeV (31%) which seems a reasonable fraction for the sum of various pulse height defects. An amount of 9.4% of signal loss due to vacancies and phonons is determined by SRIM for ^{238}U ions in Si. If that data is correct, then the remainder of 12.1 MeV (21.6%) must be due to recombination effects.

The width of the measured implantation energies from 36.5 to 41.8 MeV is in reasonable agreement with the limits set by the target thickness and additional statistical fluctuations.

The chains were measured from the detector strip numbers 10, 11 and 9 (in order of their appearance) which are located at 7.5, 12.5 and 2.5 mm right from the center of the 80 mm wide detector. The mean vertical positions were at 9.16, 3.20 and 10.43 mm, respectively, measured from the bottom of the 35 mm long detector strips. The position distribution close to the center of the detector evidences the correct setting of the SHIP electromagnetic-field values and proper estimate of the ionic-charge state of the residues.

All signals, which were assigned to a decay chain, had identical strip number and the vertical positions were distributed around the mean value according to the detectors position resolution of ± 150 μm for α-particles of about 10 MeV energy. The mean values were calculated from the more accurate data of the fully stopped α-particles, marked with letter E in fig. 1, except for the case of α_2 of chain two, which was also used because of the high-energy loss value of 6.96 MeV. In all cases, the vertical position was calculated from the signals of the bottom connector, which had the higher amplitude and hence higher accuracy. The redundant signals from the top connector resulted in complementary values. The positions from low-energy escape α's and high-energy implantations deviate up to 1 mm from the mean values, because of decreasing position resolution at decreasing energy and signal processing in a different branch of low amplification, respectively.

We conclude from the implantation signals and position measurement that the three measured sequences of signals arise from correlated events. They show all characteristic properties of decay chains of implanted fusion products.

The first three α decays of the chains, which were assigned to the new nuclei ^{272}111, ^{268}Mt and ^{264}Bh, are grouped at energies of 11.0, 10.2 and 9.4 MeV, respectively (see fig. 2). The α decays of ^{260}Db and ^{256}Lr are located at 9.05 and 8.45 MeV. The trend of decreasing energy by about 0.7 MeV per α decay along a decay chain was

also observed for the neighboring decay chains of ^{266}Mt, ^{269}110, ^{270}110 and ^{271}110. This is a result of the smoothly varying potential-energy landscape below neutron number 162. The trend is broken when $N = 162$ is crossed, as it is in the case of the ^{277}112 decay chain (see sect. 4).

Each of the nuclei in the decay chain of ^{272}111 decays by transitions of more than one energy. In particular, we measured two transitions from ^{272}111 itself, at least two from ^{268}Mt and four from ^{264}Bh. Three and six different energies are known from ^{260}Db and ^{256}Lr, respectively.

A commensurate condition for a convincing assignment of the decay chains is the agreement of the data for the decay of known daughter nuclei with literature values. However, this condition must not necessarily be fulfilled, which could occur, *e.g.*, in the case of incompletely known decay schemes of the daughter nuclei.

In the case of dubnium we measured three α's which were fully stopped in the detector. Their energies are 9.146, 9.156 and 9.129 MeV (from chain 2, 4 and 6) with a mean value of 9.144 MeV. The relative error bar for each of the single energies is determined by the detector resolution of 20 keV (FWHM) and amounts to $\sigma = 8.5$ keV. However, the absolute uncertainty is affected by the calibration procedure. In our case α decays were used from evaporation residues of reactions with ^{142}Ce and ^{150}Nd targets and the ^{64}Ni beam. We estimate an absolute uncertainty of $\sigma = 20$ keV.

Ghiorso *et al.* [14] assigned three α lines to the decay of ^{260}Db with energies and intensities of 9.06 (55%), 9.10 (25%) and 9.14 MeV (20%). The accuracy of the energy was estimated to be 0.02 MeV. From a second experiment on the decay of ^{260}Db, Bemis *et al.* [15] reported values of 9.041 (48%), 9.074 (25%) and 9.120 MeV (17%) for the α decay and a 9.6% spontaneous-fission branch. The α energies were accurate within $\pm$0.014 MeV for the first two lines and within $\pm$0.017 MeV for the last line, however, the values are systematically smaller by about 0.020 MeV than in the first experiment. Our energy of 9.144 MeV is in agreement within the given accuracy with the third high-energy line of the literature data.

A slightly different, 56 keV higher energy was measured from chain 3 in the 1994 experiment. However, the energy value of 9.200 MeV was determined from an escape event stopped in the back detector. For such events the energy resolution is only 40 keV (FWHM). Therefore, this transition is still in agreement with the other three decays measured at higher accuracy.

Two of the α-particles from the ^{260}Db decay escaped from the detector (chain 1 and 5). However, from the energy loss signal we could determine the lifetimes of 1.97 and 14.98 s, respectively. Together with the lifetimes of the other four decays we determine an arithmetic mean value of $(3.8^{+2.6}_{-1.1})$ s, from which a half-life of $(2.6^{+1.8}_{-0.8})$ s follows. The literature values for the half-life are (1.6 ± 0.3) s [14] and (1.52 ± 0.13) s [15]. The data are in agreement, although we notice that the relatively long lifetime measured in chain 5 shifts our mean value upwards.

A statistical analysis as worked out by Schmidt *et al.* [17] for an exponential distribution shows that the upper limit for a confidence level of 68% is at 5.8 times the true value in the case of one event, which means in our case a limit at 12.7 s for the lifetime using the most accurate half-life of 1.52 s ($\tau = 2.19$ s). On average, each 6th decay must be even beyond that limit. In addition, the lifetimes of the other members of the chain do not show any peculiarity which would justify the assignment to an isomeric state. Therefore, we consider the 14.98 s decay to be a statistical fluctuation in the decay of ^{260}Db.

Six α transitions are known from the decay of ^{256}Lr, $T_{1/2} = 28$ s [13] (see fig. 2). In our first experiment only one decay was measured. The other two could not be unambiguously correlated due to long lifetimes. An energy of 8.463 MeV was determined from the energy loss and residual energy of that escape event (chain 3). In the new experiment we measured almost the same energy (8.465 MeV) from chain 4, but now with better accuracy from a fully stopped α-particle. Another different α energy of 8.423 MeV was measured from chain 6. The same precision of the energy measurement holds as already discussed in the case of ^{260}Db. The two energies agree with the known transitions of 8.475 (13.3%) and 8.430 MeV (37%) [13], both energies given with an accuracy of $\pm$0.015 MeV.

One decay of ^{256}Lr was detected as an escape event with a lifetime of 47 s. The arithmetic mean of the lifetimes from the four measured decays is 47 s, the half-life is (33^{+33}_{-11}) s in agreement with the literature value.

We conclude that, in all of the six measured decay chains, the energies and lifetimes of the 4th and 5th α decay are in agreement with the literature data on the decay of ^{260}Db and ^{256}Lr. Especially, high precision and completeness was obtained in the case of chain 4 and 6 of the confirmation experiment. The chains end for being detectable after the α decay of ^{256}Lr due to the large electron capture branch of ^{252}Md ($b_{\mathrm{EC}} > 50\%$ [13]) and the long half-life of ^{252}Fm ($T_{1/2} = 25.4$ h).

As a consequence of the discussion presented before, we assign the α transitions preceding ^{260}Db to the previously unknown nuclei ^{264}Bh, ^{268}Mt and ^{272}111. The arithmetic mean values of the lifetimes determined from the six decay chains are 1.5 s, 60 ms and 2.3 ms, respectively, resulting in half-lives of $(1.0^{+0.7}_{-0.3})$ s, (42^{+29}_{-12}) ms and $(1.6^{+1.1}_{-0.5})$ ms. The distribution of the individual lifetimes is in agreement with statistical fluctuations (see discussion before), although a possible existence of isomeric levels cannot be excluded. A candidate for the decay of an isomer could be the 171 ms, $E_\alpha = 10.097$ MeV transition of ^{268}Mt in chain 2, which has a relatively long lifetime and also an energy different from the three other decays by 0.16 MeV.

Analogous is the situation in the nucleus ^{264}Bh. There, the α energies are spread across a range from 9.11 MeV (chain 6) to 9.62 MeV (chain 2 and 3). Similar wide energy and lifetime distributions were measured previously in the case of the neighboring odd-odd isotope ^{262}Bh [18]. In that case, however, there was clear evidence for an isomeric state deduced from the higher number of 29 measured events. The feeding of both the isomer ($T_{1/2} = 8.0$ ms) and the ground state (102 ms) by the α decay of ^{266}Mt

was established later [19]. Despite a wide-energy distribution of the ^{266}Mt α decays from 10.5 to 11.7 MeV based on 14 events, the lifetimes could be described by one common value corresponding to $T_{1/2} = 1.7$ ms. However, as discussed in ref. [19], the existence of isomers with similar half-life in ^{266}Mt could not be excluded. Guided by a theoretical study of Ćwiok *et al.* [20] and the correspondence of the experimental data, we conclude that an analog level structure exists in the case of ^{268}Mt and ^{264}Bh as in the two neutrons lighter isotopes ^{266}Mt and ^{262}Bh.

For the decay of ^{272}111 redundancy in the α energies was measured in two cases (chain 5 and 6). The mean energy value is 11.027 MeV. Compared with the previously measured energy of 10.820 MeV (chain 3), a 210 keV difference of the Q_α-values results. In this context the coincidence of the ^{272}111 α decay from chain 1 with a signal of (155.0 ± 0.8) keV energy in the Ge detector is especially interesting (see discussion in [5]). The energy of 155 keV is close to the predicted $K_{\alpha 1}$ X-ray energy of meitnerium ($E_{K\alpha 1} = 151.7$ keV) [21]. If the low-energy α transition populates a level at 207 keV in ^{268}Mt, then the transition energy from that level would be sufficient to eject one of the K-electrons which are bound by an energy of 177 keV [21]. Unfortunately, we failed to measure if the ^{272}111 α-particle in chain 1 belongs also to the low-energy group, because that α-particle escaped from the detector.

At the limit of belonging together are the three α decay energies of ^{268}Mt from chain 1, 3 and 4. They are grouped at a mean energy value of 10.258 MeV. An argument that the origin of the 10.221 MeV α decay (chain 3) is different from the other two is the fact that this transition was in coincidence with a 93 keV signal in the Ge detector. Therefore, we tentatively split the three transitions into two groups consisting of the transition measured from chain 3 at 10.221 MeV and the two transitions from chain 1 and 4 at a mean energy value of 10.276 MeV.

In the case of ^{264}Bh the energies from chain 2 and 3 and from chain 1 and 5 agree well within the detector resolution. The mean values are 9.619 and 9.494 MeV, respectively.

Our conclusion of the recent $Z = 111$ experiment is that our first results are confirmed and that the new data reveal considerably improved information on the decay pattern of the chains starting at ^{272}111.

4 Element 112

4.1 Results

The experiment for confirmation of the synthesis of element 112 was performed in May 2000 (see table 1). Using the same reaction as in the first experiment, $^{70}\mathrm{Zn} + {}^{208}\mathrm{Pb} \rightarrow {}^{278}112^*$, we measured one additional chain of ^{277}112. The decay properties are in agreement with the second chain of the first experiment down to ^{265}Sg. The new chain ends at ^{261}Rf by a previously unknown spontaneous-fission branch.

In our first experiment we chose a beam energy of 343.8 MeV, which resulted in a 10.1 MeV excitation energy of the compound nucleus. A beam dose of 3.4×10^{18} was collected during the 24 days experiment. The cross-section which resulted on the basis of one decay chain was 0.5 pb.

The beam energy had been chosen according to a measured trend of cross-section maxima into the direction of lower excitation energy with increasing element number [4]. In the new experiment we chose a slightly higher beam energy which resulted in 12.0 MeV excitation energy in order to determine the trend of the cross-section at varying beam energy. Arguments for increasing instead of decreasing the beam energy resulted from theoretical studies which predicted the cross-section maximum at $E^* = 12.6$ MeV [22] or even a sharp increase up to about 100 pb at $E^* = 14$ MeV [23].

During the 19 days experiment in May 2000 we collected a beam dose of 3.5×10^{18} ions. A cross-section of 0.5 pb was deduced from the only one decay chain observed, using the same efficiency of 45% as in the first experiment. The measured decay data are shown together with our assignment in fig. 3. A comparison between half-lives and Q_α-values from both chains is presented in fig. 4.

The decay chain starts with an implantation signal from the detector strip 16 at an energy of 24.1 MeV. This energy is lower than the implantation energies in the element-111 run, because we used degrader foils in front of the Si detector in order to stop a part of the low-energetic background of projectiles. Another reason was to avoid high-amplitude signals which cause tailing and make an exact α energy measurement more difficult in the case of lifetimes shorter than 500 μs. The degrader was composed of a 3 μm thick Mylar foil and an Al wedge-shape degrader of 1.1 μm thickness on the left side (there the background has higher energy, but is deflected more by the last SHIP dipole magnet due to higher-charge states, see fig. 1 in ref. [10]) and 0.5 μm on the right side (where strip 16 is located). The foils of the time-of-flight detectors were the same as described before, however, the thickness of the charge equilibration foil was only 30 μg/cm^2 carbon in this run.

We tried to reproduce the measured implantation energy using SRIM-2000 for the calculation of the energy loss of projectiles and fusion products. We obtained a kinetic energy of 41.8 MeV for the ^{277}112 ion when entering the active detector material. This value differs from the measured one by 17.7 MeV or 42%. Taking into account larger uncertainties of energy loss values due to the relatively thick degrader foils and a relatively increased contribution from nuclear stopping at lower kinetic energy as in the element 111 run (which was performed without degrader foils), a pulse-height defect of 42% seems reasonable.

The implantation signal was in coincidence with signals from all three time-of-flight detectors. Subsequent to implantation three α-particles were measured with full energy in the stop detector. The fourth α escaped. The energy loss signal of 0.2 MeV was relatively low due to the

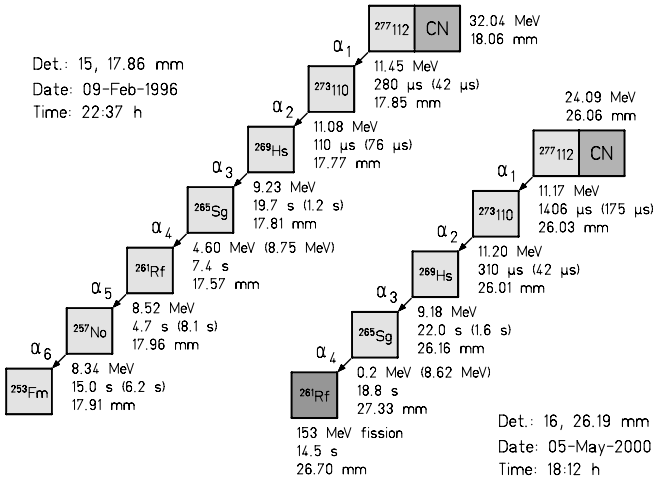

Fig. 3. Decay chains attributed to the decay of $^{277}112$ produced in reactions $^{70}Zn + {}^{208}Pb$. The newly measured chain is plotted on the lower right side of the figure. The previously measured chain from our 1996 experiment [6] is also plotted for better comparison. The lifetimes given in brackets were obtained from calculations using the WKB method (see text). In the case of escape α-particles the energies were calculated from the lifetimes. The results are given in brackets.

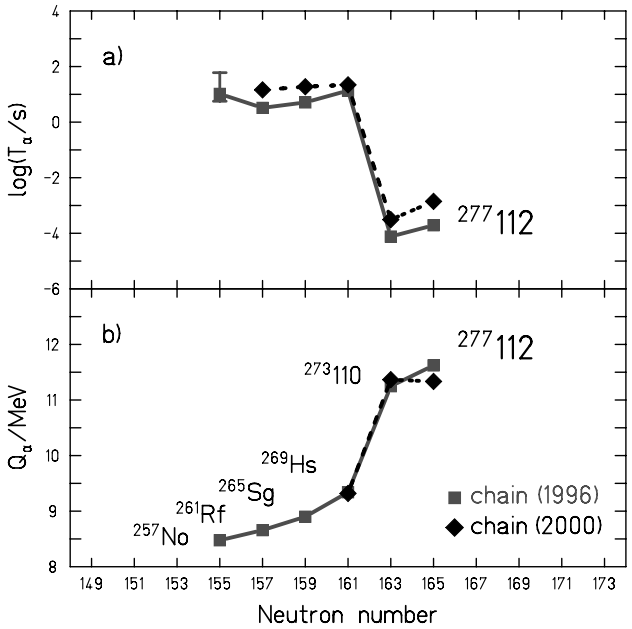

Fig. 4. Systematics of the decay chains of $^{277}112$: upper part, measured half-lives; lower part, Q_α-values. An example of the error bar of the half-lives is shown in one case. It reflects the statistical uncertainty in the case of one event and demonstrates the agreement between the lifetime measurements for each of the nuclei of the decay chains.

location of the parent nucleus close to the detector surface. All α events occurred during the 5.5 ms beam-on intervals, however, none of the time-of-flight detector signals was in coincidence, which indicates that the pulses from the Si detector were caused by radioactive decays. No signal from the γ detector was in coincidence neither with the implantation nor with the α-particles.

Finally, a high-energy event of 153 MeV was measured in the stop detector. This value is given without any correction for energy deficit. The event occurred 6.2 ms after the beginning of the 14.5 ms beam-off period and, therefore, must be assigned to a radioactive decay. No high-energy signal was measured from the back detectors, but there was a 2.3 MeV signal from back-detector segment 6 in coincidence within a time window of 1 μs. This segment is located closest to strip 16 of the stop detector. The low-energy signal could be due to the residual energy of a fission fragment, but electrons and a cascade of γ-rays emitted during the fission process are another possible explanation. From the Ge detector we received a coincident signal of (1857 ± 6) keV. The sum of information obtained from that high-energy event identifies it as a spontaneous-fission event. The relatively high energy measured in the stop detector indicates that most of the fission energy was deposited there.

All signals from the stop detector were tagged by a logic signal which identifies them to originate from detector strip number 16. This strip is located at the border of the detector, 35–40 mm on the right side from the center. The vertical positions are grouped around a mean value of 26.19 mm from the bottom of the strip within a position window set by the detector resolution. The mean value was calculated from both the bottom and top signals from all events except the escaped α from which only the top signal was above the discriminator level.

The measured time intervals between the detector signals are given in fig. 3. The first two α decays are on the order of one millisecond, the subsequent two and the fission event on the order of few tens of seconds. This is still short relative to the intervals arising from background events if the position information is included. Therefore, the origin of the correlated event chain by chance is unlikely. During the whole experiment no other similar chain was observed.

4.2 Discussion

The decay chain of $^{277}112$ is located in a region of the chart of nuclei, where the decay properties of the daughter nuclei were not studied in SHIP experiments before. The reason is that those nuclei can be directly produced only in reactions using actinide targets for which SHIP has a low transmission of only a few percent.

At the time of our first experiment (1996) no literature data was available for comparison with the decay properties of the daughter and granddaughter nuclei $^{273}110$ and ^{269}Hs. Today, the data on these nuclei is still scarce and incomplete. Only one work was published dealing with the synthesis of the daughter nucleus $^{273}110$ [24] and only recently a paper was submitted on the decay of the granddaughter ^{269}Hs [25]. Therefore, we begin the discussion with presenting the available information on the decay properties of nuclei at the lower end of the decay chain,

aiming to work out a consistent picture of the decay properties of nuclei relevant to the decay chain of $^{277}112$.

The measurable end of the decay chain is set by the long-living nuclide ^{253}Fm which decays dominantly by electron capture ($T_{1/2} = 3.0$ d, $b_{EC} = 88\%$) [13]. It is populated by the α decay of ^{257}No ($T_{1/2} = 25$ s, $b_\alpha \approx 100\%$).

Three α lines were measured from the decay of ^{257}No studied in the reaction $^{13}C + ^{248}Cm$ which produced ^{257}No directly in a 4n evaporation channel [26]. The energies of the transitions were 8.22, 8.27 and 8.32 MeV with relative intensities of 55, 26 and 19%, respectively.

The decay of ^{257}No was observed again subsequent to the α decay of ^{261}Rf. This nucleus was first identified by Ghiorso *et al.* [27] using the reaction $^{18}O + ^{248}Cm \rightarrow ^{266}Rf^*$. An α line at (8.28 ± 0.02) MeV was measured superimposed on the α lines of the daughter decay. A half-life of (65 ± 10) s was obtained for the mother activity. A new half-life of (78^{+11}_{-6}) s, based on a greater number of decays, was published in ref. [28].

Recently, the decay of ^{261}Rf and ^{262}Rf was reinvestigated by Lazarev *et al.* [29] using the reaction $^{22}Ne + ^{244}Pu \rightarrow ^{266}Rf^*$. Using a α-α-correlation method the previously obtained data on the decay of ^{261}Rf were confirmed. From 69 measured correlations no α-particle energy was greater than 8.4 MeV neither from the parent nor from the daughter decay.

For the neighboring spontaneously fissioning isotope ^{262}Rf an upper limit for α decay of 3% was measured. This value is in agreement with the more restrictive 0.8% limit measured by Lane *et al.* [30]. In the latter paper also the presently most accurate half-life value of (2.1 ± 0.2) s was presented, and a previously measured half-life of 47 ms [31] was considered as a candidate for a spontaneously fissioning K-isomer.

Both isotopes, ^{261}Rf and ^{262}Rf, were observed as α decay daughter products from the corresponding parent nuclei of seaborgium ($Z = 106$). The experiments were performed at the Dubna gas-filled separator [32] and at GSI using chemical separation by ARCA [33] and OLGA [34]. Alpha decays with energies in the range from 8.6 to 9.0 MeV were assigned to ^{265}Sg and ^{266}Sg. The two isotopes were distinguished by the decay properties of the daughter nuclei known at the time, α decay of ^{261}Rf and spontaneous fission of ^{262}Rf.

Four decay chains were measured by Lazarev *et al.* [24] in the reaction $^{34}S + ^{244}Pu \rightarrow ^{277}110^*$, which were assigned with different significance to the decay of $^{273}110$. In the chain which was given the highest weight the decays of ^{269}Hs and ^{261}Rf were missed, however, the measured data are not in contradiction (taking into account the resolution of 120 keV FWHM for that chain) to the data measured in our experiment down to the decay of ^{265}Sg, although the combined lifetimes measured in [24] for the decay of ^{269}Hs–^{265}Sg (158 s) and ^{261}Rf–^{257}No (384 s) were unusually long.

In a very recent paper Türler *et al.* [25] reported on three and two decay chains, which they had assigned to ^{269}Hs and the new isotope ^{270}Hs, respectively. The nuclei were produced in reactions $^{26}Mg + ^{248}Cm \rightarrow ^{274}Hs^*$ and transported in a He/O_2-carrier gas in form of volatile HsO_4 molecules into a cryo thermochromatography detector system which was built from two arrays of PIN diodes facing each other at a distance of 1.6 mm. The results on the decay of ^{269}Hs fully confirm the data which we obtained from the decay of $^{277}112$.

The following conclusions can be drawn from that work, from the previous studies discussed above and from our decay chains assigned to $^{277}112$.

1. *Decay of* ^{269}Hs: One component of the α decay has an energy of 9.18 MeV, determined from our new chain. The uncertainty of the α energy is ±0.02 MeV given by possible systematic deviations and a detector resolution of 20 keV FWHM. This transition likely populates an excited level in ^{265}Sg as indicated by the 50 keV higher energy measured in the 1996 chain. The energies measured in ref. [25] were 9.18, 8.88 and 9.10 MeV with an uncertainty of typically $^{+0.07}_{-0.03}$ MeV. The asymmetric error bar is caused by possible energy loss in the gas between the detector arrays. The energy loss could be even more than 40 keV in the case that α-particles are emitted under shallow angles to the detector surface. The half-life deduced from our work is (14^{+26}_{-6}) s (arithmetic mean of two events). This value is long enough to secure survival of collection and transport time needed in the chemical experiment [25]. There, ^{269}Hs was the parent nucleus and its half-life could not be measured. We notice that the data on the decay of ^{269}Hs from our work are in agreement with the results obtained by Türler *et al.*, which proved by chemical means, that the measured nucleus belongs to element 108.

2. *Decay of* ^{265}Sg: In our experiment the α-particles from the decay of ^{265}Sg escaped from the detector, however, their lifetime could be measured from the energy loss signals. From the two events we determine a half-life of (9^{+17}_{-4}) s. An estimate of the α-particle energies can be obtained using the WKB method and a potential barrier given by Igo [35]. From the individual lifetimes we determine α energies of 8.75 MeV and 8.62 MeV, respectively. Decay energies between 8.6 and 9.0 MeV were measured for this nuclide [32,34], and a half-life of $(7.4^{+3.3}_{-2.7})$ s was deduced [34]. In the recent work by Türler *et al.* [25] α energies of 8.69, 8.90 and 8.68 MeV were measured and individual lifetimes of 4.4, 17.1 and 9.3 s, respectively. We notice that also the data from the decay chains of $^{277}112$, which were assigned to ^{265}Sg, agree with the data from the other experiments.

3. *Decay of* ^{261}Rf: The literature data on the decay of this nucleus are $E_\alpha = 8.28$ MeV and $T_{1/2} = 78$ s ($\tau = 112$ s). In our experiments we measured for the decays of this chain member an α-particle with an energy of 8.52 MeV and a lifetime of 4.7 s (old chain), and a spontaneous-fission event with a lifetime of 14.5 s (new chain). The α energy of 8.52 MeV and spontaneous fission were not observed in experiments, in which ^{261}Rf was produced as evaporation residue. Also not the short lifetime of 9.6 s deduced from our chains. However, in the recent experiment by Türler *et al.* [25] these observations were confirmed. Two α decays with an energy of 8.50 MeV each

and one fission event were measured from the decay chains of ^{269}Hs with lifetimes of 2.4, 0.8 and 7.9 s, respectively.

In experiments which produced ^{261}Rf as daughter of the evaporation residue ^{265}Sg (a total of 21 decays are cited in the literature [32–34]), an 8.52 MeV α decay was measured only in one case [33]. This decay, however, occurred with a lifetime of 142 s and could not be definitely assigned to the decay chain of ^{265}Sg [36]. In few other cases it could be possible that due to the previously unknown fission branch of ^{261}Rf, α-fission correlations were erroneously assigned to the decay sequence ^{266}Sg–^{262}Rf.

Summarizing the data from our experiments on ^{277}112, that on ^{269}Hs from ref. [25] and the previously obtained literature data, we conclude that two levels exist in ^{261}Rf with half-lives of (78^{+11}_{-6}) s and $(4.2^{+3.4}_{-1.3})$ s, which decay by emission of 8.28 MeV α-particles in the first case and by emission of 8.52 MeV α's and by spontaneous fission with a branching of about 40% in the latter case. The α energy of 8.52 MeV fits better into the systematic of ground-state α energies [37]. Therefore, we tentatively assign the 4.2 s level to the ground state, whereas the 78 s level could be a higher-spin isomeric state, because it is preferably produced in heavy-ion fusion reactions.

4. *Decay of* ^{257}No: The α decay of this isotope was measured in the first experiment with $E_\alpha = 8.34$ MeV and $\tau = 15.0$ s. These data are in agreement with the literature data (see before). In the work by Türler *et al.* [25] α decay of ^{257}No was measured only in one of the two cases subsequent to α decay of ^{261}Rf. The non-observation of the ^{257}No α decay in 1 from a total of 3 cases, although its energy and lifetime should be easily detectable, is indication for an electron capture branch. Such a decay channel would populate the 5.5 h ^{257}Md which has an α/EC ratio of 15/85 [13]. The decay of ^{257}Md could probably not be observed in ref. [25]. In previous works [26,27,29] the ^{257}Md α lines at 7.0 MeV were covered by intense background lines.

A calculation of the electron capture half-life for ^{257}No is difficult, because of its low Q_β-value [38]. However, a logarithmic interpolation of the EC half-lives of ^{255}No and ^{259}No results in $T_{1/2,\mathrm{EC}} = 42$ min and thus $b_{\mathrm{EC}} \approx 1\%$ for the decay of ^{257}No. Considering the experimental result, we conclude that ^{257}No has an EC branch probably near the middle between 1 and 30%.

So far, we discussed in detail the data which were measured in our work in the reaction ^{70}Zn + ^{208}Pb $\rightarrow$ ^{278}112* and the results which were obtained in other experiments on lighter nuclei performed up to the study of ^{269}Hs. We showed, that our assignment of the data measured for the second chain in the 1996 experiment [6] and the results and interpretation deduced from the new decay chain measured in 2000 [10], were fully confirmed by independent investigations. The agreement of our decay chains with literature data up to the decay of ^{269}Hs proves the assignment of the two preceding α decays to the nuclei ^{273}110 and ^{277}112. Other possible evaporation residues as starting points of the decay chain were already discussed and excluded in ref. [6].

The first two α decays of the new chain have energies of 11.17 and 11.20 MeV, respectively, which are succeeded by an α of only 9.18 MeV, an energy step by about 2 MeV. Correspondingly, the lifetime increases by about five orders of magnitude between the second and third α decay. This decay pattern is in agreement with the one observed for the chain in our first experiment (see fig. 4). It was explained as the result of a local minimum of the shell correction energy at neutron number $N = 162$ which is crossed by the α decay of ^{273}110 [39,40].

Recently, Ćwiok *et al.* [41] performed a Hartree-Fock-Bogoliubov calculation with Skyrme-Sly4 interaction and a zero-range pairing in order to calculate the low-energy quasiparticle states of nuclei along the ^{277}112 decay chain. The calculations clearly provide evidence for high- or low-spin isomeric states near the ground state in both ^{277}112 and ^{273}110. Due to the large single-particle energy gap at $N = 162$, the low-spin levels $1/2^+$ and $3/2^+$ are shifted upwards to higher energy by about 1 MeV in ^{269}Hs. Therefore, the Q_α-values of the favorite transitions from the low-spin state in ^{273}110 is reduced by that amount of energy. The population of an excited level at about 1 MeV seems unusual if compared with α-decay properties in the region of lighter elements. There, however, the ground-state Q_α-values are of the order of about 6 MeV, whereas in the case of ^{273}110 they are almost twice as high.

Beyond $N = 162$ the predicted Q_α-values of the favored transitions between the low- and high-spin levels are almost equal. This result is in line with our experimental data, if we tentatively assign the measured α decays to transitions between the high-spin states. Search for low-spin isomeric states as predicted theoretically remains a task for future experiments.

Support for the quality of the model was obtained recently by a convincing description of a high-spin K-isomer which was observed experimentally in ^{270}110 at an excitation energy of 1.13 MeV with a half-life of 6.0 ms [9]. In that nucleus the same single-particle levels for the neutrons are responsible for the low-energy level scheme as in the case of the nuclei at the upper part of the decay chain from ^{277}112.

We also calculated the α-decay probabilities using the WKB method and a potential barrier given by Igo [35]. This model reproduces the α-decay probabilities of even-even nuclei in the region of heavy elements within a factor of two. The result for the lifetime of each individual transition within the ^{277}112 decay chains, assuming $\Delta l = 0$ transitions, is given in brackets in fig. 3. Some of the transitions are reproduced accurately, others reveal hindrance factors of about 10 on the average. In order to explain these hindrance factors solely by centrifugal barrier, Δl-values of about 4–6 are needed. However, another more likely reason to explain the hindrance factors is a strong configuration mixing as suggested by the theoretical description.

A cross-section of $(0.5^{+1.1}_{-0.4})$ pb was measured for the new data point at 12.0 MeV excitation energy. This value fits well into the systematic of cross-sections. A cross-

section increase with increasing beam energy as predicted by theoretical investigations [22,23] was not observed.

During our experiment we also decreased for a period of 7 days the energy of the ^{70}Zn beam to the previously used value of 343.8 MeV. No further event was measured at a beam dose of 1.2×10^{18}. A new mean value for the cross-section of $(0.4^{+0.9}_{-0.3})$ pb follows for the data point at 10.1 MeV excitation energy (1 event at 4.6×10^{18} projectiles).

5 Summary and outlook

Two experiments were carried out in 2000, which resulted in the detection of three additional decay chains of ^{272}111 and one more decay chain of ^{277}112. The reaction $^{64}\mathrm{Ni} + {}^{209}\mathrm{Bi}$ was used for the synthesis of the element 111 and $^{70}\mathrm{Zn} + {}^{208}\mathrm{Pb}$ for the element 112. These were the same reactions as used in our first experiments. The new data are in full agreement with the results obtained in 1994 from three decay chains measured of ^{272}111 and with the second chain of ^{277}112 measured in 1996. The quality of the data was improved and new, previously not known decay properties were measured. The decay data of daughter nuclei of the chains was compared in detail with literature data available until October 2001. The agreement of our data with the data on the daughter nuclei obtained independently of other experiments, results in an unambiguous assignment of the parent nuclei to ^{272}111 and ^{277}112, respectively.

Part of the data was also compared with the results from theoretical calculations. The decay properties are in agreement with structure calculations of nuclei near $N = 162$ and $Z = 108$–110. For these nuclei a local minimum of the shell correction energy was calculated, which results from a low single-particle level density at large quadrupole plus hexadecapole deformation.

We performed a re-analysis of our data measured since 1994 in order to confirm the previously obtained results and to prove consistency with the presently used computer programs. In the course of this work we reviewed 34 decay chains, four of ^{269}110, eight of ^{270}110, thirteen of ^{271}110, six of ^{272}111 and three of ^{277}112. In two cases (second chain of ^{269}110 measured in 1994 and first chain of ^{277}112 measured in 1996) we found inconsistency of the data, which led to the conclusion, that for reasons not yet known to us, part of the data used for establishing these two chains were spuriously created. In all other cases the previously obtained data are exactly reproduced.

The measured cross-sections were on a level of 1 pb. Beam times of several weeks were necessary for the detection of few events, although beam currents and detection sensitivity had been continuously improved in recent years. The results presented in this paper and recent work in Dubna on elements 114 and 116 [42] demonstrate that further investigation of superheavy nuclei is promising. However, systematic work on an extremely low cross-section level is mandatory. At the UNILAC, which was our working horse for now 25 years, a further increase of the sensitivity is not easily possible. The reasons are that the duty factor of 28% cannot be increased and that necessary long beam times cannot be arranged due to the multifold use of the accelerator also as injector of beams into the high-energy facility SIS. A possible solution might be the construction of a high-current accelerator dedicated to heavy element research delivering beams of 100% duty factor. With such a machine, improved target technology, a separator upgrade with respect to higher transmission and further background suppression, a detailed investigation of the properties of superheavy nuclei will become feasible.

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