Cross section limits for the $^{248}\text{Cm}(^{25}\text{Mg}, 4n-5n)^{268,269}\text{Hs}$ reactions

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We report on an attempt to produce and detect ^{268}Hs and ^{269}Hs in the nuclear fusion reaction $^{25}\text{Mg} + ^{248}\text{Cm}$ using the gas phase chemistry apparatus COMPACT. No decay chains attributable to the decay of hassium isotopes were observed during the course of this experiment. From the nonobservation of ^{269}Hs we derive a cross section limit of 0.4 pb (63% confidence limit) for the reaction $^{248}\text{Cm}(^{25}\text{Mg}, 4n)^{269}\text{Hs}$ at a center-of-target beam energy of 140 MeV. The evaluated cross section limit for the $^{248}\text{Cm}(^{25}\text{Mg}, 5n)^{268}\text{Hs}$ reaction depends on the assumed half-life of unknown ^{268}Hs . Current systematics of the half-lives for even-even Hs isotopes suggests a value of 0.5 s, resulting in a cross section limit of 1.3 pb.

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Superheavy elements exist only because of nuclear shell effects, which stabilize them against spontaneous fission (SF). Nuclear stability in the region of elements with proton number $104 \leq Z \leq 108$ is primarily influenced by the closure of deformed shells at $Z = 108$ and $N = 162$. Macroscopic-microscopic as well as self-consistent mean-field calculations [1–4] predicted ^{270}Hs to be a deformed doubly magic nucleus. The nuclide ^{270}Hs was identified in one of our previous experiments and its observed properties confirm theoretical predictions of nuclear shell stabilization at $Z = 108$ and $N = 162$ [5]. To explore the strength of the shell stabilization in this entire region, it would be interesting to compare properties of ^{270}Hs to those of other even-even nuclei in its vicinity. However, most of the nearby even-even nuclides are presently unknown (see Fig. 1 in Ref. [5]), including both neighboring even Hs isotopes, ^{268}Hs and ^{272}Hs .

In this Brief Report we report on an experiment aimed at identifying the unknown nuclide ^{268}Hs . In the nuclear fusion reaction $^{25}\text{Mg} + ^{248}\text{Cm}$, the isotopes ^{268}Hs and ^{269}Hs were expected to be produced in the $5n$ and $4n$ evaporation channel. The HIVAP code [6] predicts maximum cross sections of 1.2 and 0.6 pb, respectively, for these two channels at beam energy of $E_{\text{lab}} = 140$ MeV. HIVAP predictions of cross sections of the $3n$, $4n$, and $5n$ channels of the reaction $^{26}\text{Mg} + ^{248}\text{Cm}$ proved to be remarkably close to experimental values [7] and we expected a similar agreement for the reaction $^{25}\text{Mg} + ^{248}\text{Cm}$.

Various predictions for the decay of ^{268}Hs through emission of an α particle or by spontaneous fission (SF) exist in the literature, but these predictions have large uncertainties and vary considerably as shown below. These uncertainties result from the strong dependence of the half-life on the α decay energy, Q_α , and shape of the fission barrier. Small differences in Q_α , in height of the fission barrier, or an alternative path over the multidimensional fission barrier often yield half-lives that differ by orders of magnitude. A partial α decay half-life can be derived using a Viola-Seaborg-type formula from Q_α values. Half-life estimates using the most recent set of parameters [8] are typically accurate to within a factor of two for even-even nuclei. Assuming a smooth trend of Q_α values in this region of the chart of nuclides, an empirical estimate of Q_α of ^{268}Hs can be obtained by interpolating the Q_α values of its known even-even neighbors, ^{266}Hs [9] and ^{270}Hs [7]. From the interpolated value of $Q_\alpha = 9.58$ MeV, a partial α decay half-life of $T_{1/2}(\alpha) = 0.5$ s is derived [8]. Macroscopic-microscopic calculations [10], which show very good agreement with experimental values for nuclei in this region, predict $Q_\alpha = 9.49$ MeV, which correspond to half-life of $T_{1/2}(\alpha) = 0.92$ s [8]. In Ref. [11] predictions on both, partial α and SF half-lives are made based on the density-dependent cluster model. Depending on the used set of parameters, $T_{1/2}(\alpha)$ was predicted as ~ 40 ms or 7.3 s, whereas the partial SF half-life $T_{1/2}(\text{SF})$ was predicted as 84 s. These calculations suggest α decay to be the preferred decay mode of ^{268}Hs . The predicted $T_{1/2}(\text{SF})$ is in good agreement with the value calculated in Ref. [4] ($T_{1/2}(\text{SF}) = 96$ s), although other calculations result in significantly shorter SF half-lives (e.g., $T_{1/2}(\text{SF}) = 0.4$ ms–1.5 s [12]). For all but the shortest predicted $T_{1/2}(\text{SF})$ we expect ^{268}Hs to decay predominantly by α decay with a half-life of about 0.5 s to the known nuclide ^{264}Sg , which decays by SF with $T_{1/2} = 37$ ms [13].

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It was shown [5,7,14] that a chemical separation of hassium as volatile HsO_4 from other nuclear reaction byproducts and the detection of the nuclear decay of Hs isotopes and their daughters in a thermochromatography detector is the most effective and efficient method for studying production and decay of hassium isotopes. The nuclear reaction $^{248}\text{Cm}(^{26}\text{Mg}, 3n-5n)^{269-271}\text{Hs}$ as well as the decay properties of the formed Hs isotopes and their daughters were investigated using this technique [7]. For the work reported here, we have improved our previously used Cryo Online Multidetector for Physics and Chemistry of Transactinides (COMPACT) to reduce the transport time to about one second. It is therefore suitable for searching ^{268}Hs with an expected half-life of ~ 0.5 s.

In the recent experiment, a beam of $^{25}\text{Mg}^{4+}$ ions was accelerated by the UNILAC at the GSI to 7.45 MeV/nucleon and impinged on a rotating ^{248}Cm target. The irradiation setup was identical to the one used in our previous experiments [5,7]. Before entering the target material, the beam passed through 20.2 μm thick Be vacuum windows, 6 mm of He/O₂ mixture (10% O₂) at atmospheric pressure, and 15.3 μm thick Be backings of the target material. The target consisted of three arc shaped segments with initial average thicknesses of 788 $\mu\text{g}/\text{cm}^2$, 743 $\mu\text{g}/\text{cm}^2$ and 244 $\mu\text{g}/\text{cm}^2$ of Cm (^{248}Cm : 95.8%, ^{246}Cm : 4.2%). One segment contained 2% in weight of ^{152}Gd (30% isotopic enrichment) for the simultaneous production of α -decaying Os isotopes. Os is a chemical homolog of Hs. The beam energy in the center of the target was $E_{\text{lab}} = 140$ MeV [15], corresponding to a compound nucleus excitation energy, E^* , of 49 MeV [16]. HIVAP [6] predicts the maximum cross section of 1.2 pb for ^{268}Hs produced in the $5n$ channel at about this energy. With typical beam intensities up to 1.5 particle/ μA , a beam integral of 3.61×10^{18} was accumulated in 170 h of irradiation. Products of the nuclear reactions recoiled from the target and were stopped and thermalized in a He/O₂ mixture in the recoil chamber (RC) heated to 350°C. Because the gas mixture contained 10% of O₂, recoiling products of nuclear reactions were partially oxidized in the RC. A cluster-less gas flow (1.4 l/min) swept the RC and transported volatile nuclear reaction products and their volatile compounds to a heated (600°C) quartz wool plug. This plug served as a filter for aerosol particles and provided a surface for completion of the oxidation reaction. All nonvolatile species were efficiently retained by this filter, while volatile species like HsO_4 , OsO_4 , Rn or At passed through the filter. The efficiency to extract synthesized Hs nuclei in the form of HsO_4 from the RC was estimated to be 80%, and the separation factor from non-volatile species was $> 10^6$ under current experimental conditions [17]. Species volatile at room temperature were transported further through a PTFE (Teflon) capillary kept at ambient temperature to the detection system COMPACT [18]. This is constructed as a thermochromatography column (TC) [19], formed by PIPS (Passivated Implanted Planar Silicon) diodes suitable for the detection of α particles and fission fragments. A sum spectrum obtained in the course of the experiment is shown in Fig. 1.

Because of the intense radiation field close to the target during irradiation, the detection setup was placed behind a thick concrete wall in a neighboring room in previous experiments

FIG. 1. Sum spectrum of all 64 detectors obtained during the course of the experiment. Due to the chemical separation, only peaks of Os, Rn, and At and their daughters are visible. The inset shows an expanded view of the area of interest ($8 < E_\alpha < 10$ MeV), where decays of Hs isotopes were expected.

[5,7]. Besides massive shielding of the detector and electronics from neutrons and γ -rays caused by irradiation, this has the advantage of direct access to the detection setup during the irradiation. As a consequence, the transport capillary leading through the thick shielding wall is rather long, of the order of 10 m. Considering a typical gas flow rate of 1.0 l/min and a capillary with a typical inner diameter (i.d.) of 2 mm, this results in transport times of 1.8 s or more. To study nuclides with $T_{1/2} \leq 1$ s, which is expected to be the case for ^{268}Hs , the transport time t_{trans} has to be minimized. Therefore the complete detection setup including electronics and the data acquisition system was placed inside the irradiation cave, close to the RC in a position heavily shielded by a 40-cm thick concrete wall, a 15-cm thick boron-paraffin layer, and a 7-cm thick Pb layer. The shielding proved to be satisfactory as no electronic noise was observed and the background of events in the detector did not increase significantly when compared to previous experiments, when COMPACT was located outside of the cave [5,7]. The detection setup had to be fully automated to allow its operation inside the cave, which was not accessible during irradiation. Moving COMPACT inside the cave allowed using a capillary with an i.d. of 1.5 mm and a length of only 4 m. From the total volume of the transport path including the RC of 24 cm³ and the gas flow rate of 1.4 l/min we estimated a mean transport time of about 1 s.

The transport time of the optimized setup was measured in a dedicated experiment with short-lived Os isotopes [18]. ^{171}Os and ^{172}Os with half-lives of 8.3 s and 19.2 s, respectively, were produced in bombardments of an enriched ^{152}Gd target (750 $\mu\text{g}/\text{cm}^2$ thick) with a chopped (1 s on/59 s off) ^{25}Mg beam. Using the same experimental setup as described above, Os was transported as OsO_4 to the COMPACT, where OsO_4 was adsorbed and the nuclear decay was measured. Each event was time-stamped and the time between the beam pulse and the decay could be evaluated. The measured time distribution of Os decays is presented in Fig. 2. A full account of this

FIG. 2. Time distribution of $^{171,172}\text{Os}$ decays, as measured in COMPACT during the transport time measurement experiments. Zero corresponds to the start of the beam pulse. A fit derived from the residence time distribution model [18] is shown as a thick line. The inset shows the first three seconds in detail.

experiment and the evaluation of the transport time along with the used model can be found in Ref. [18]. For the purpose of the present paper, it can be briefly summarized that the transport of the activity from the RC to the detector was modeled using the theory of residence time distribution [20,21]. In this model, the RC can be described as an ideally stirred reactor with a mean sweep time t_r . Transport through the capillary can be simplified to plug flow with a mean transport time t_c , because of the high diffusion coefficient. Hence, transport of the activity through both, the RC and the capillary, can be modelled as a combination of an ideally stirred reactor and plug flow. We have used this model to obtain an analytical fit of the time distribution of Os decays, as measured in our experiment. The best fit was obtained for values of $t_r = 0.39$ s and $t_c = 0.60$ s. The resulting mean transport time $t_{\text{trans}} = t_c + t_r \approx 1$ s agrees with the expected transport time, estimated from the known volume and the gas flow rate.

Because of the chemical separation [5], only α lines originating from $^{172-174}\text{Os}$ (transported as volatile OsO_4), ^{211}At , $^{219,220}\text{Rn}$, and their daughters were identified. Unfortunately, a small fraction of aerosol particles, created by beam induced sputtering of target material, reached the detection system, resulting in an evenly distributed SF background. Therefore SF-decaying Hs isotopes could not be identified. During 170 h of beam time 533 SF events with at least one fragment with $E_{\text{SF}} > 20$ MeV and 249 α particles with energies in the range of interest, $8 \leq E_\alpha \leq 10$ MeV, were detected on the whole detector array.

^{268}Hs , produced in the $5n$ evaporation channel, is expected to α decay to ^{264}Sg , a known SF isotope with $T_{1/2} = 37$ ms with an α branch $< 36\%$ [84% confidence limit (C.L.) [13]]. Therefore, we searched for short α -SF or α - α correlations. For a decay chain to be considered a true ^{268}Hs chain, the following conditions had to be met: an α decay within an energy range of $8 \leq E_\alpha \leq 10$ MeV followed by another α decay ($8 \leq E_\alpha \leq 10$ MeV) or SF (at least one fragment > 20 MeV) within 1 s in the same or a neighboring detector pair. Because of the present

α particle and SF background, pseudocorrelated sequences can be formed with nonzero probability. The number of ^{268}Hs like random correlations expected to occur during the course of experiment was 0.03 [17].

^{269}Hs , the $4n$ evaporation channel product, is known. Its decay scheme was recently revised [22,23]. ^{269}Hs decays by α decay ($T_{1/2} = 9.7$ s, $E_\alpha = 8.95$ – 9.23 MeV) to ^{265}Sg . Two states of ^{265}Sg , referred as ^{265a}Sg and ^{265b}Sg , are known. Both states decay by α decay ($^{265}\text{Sg}^a$: $E_\alpha = 8.85$ MeV, $T_{1/2} = 9$ s and $^{265}\text{Sg}^b$: $E_\alpha = 8.69$ MeV, $T_{1/2} = 16$ s) to ^{261}Rf , ^{269}Hs decays preferably (91%) to $^{265}\text{Sg}^b$, although decay to $^{265}\text{Sg}^a$ has also been observed (9%) [23]. In ^{261}Rf again two states are known, $^{261}\text{Rf}^a$ and $^{261}\text{Rf}^b$. The decay of $^{265}\text{Sg}^b$ feeds preferably (88%) $^{261}\text{Rf}^b$, which decays with the half-life of $T_{1/2} = 3$ s either via α decay ($E_\alpha = 8.51$ MeV, 9%), or undergoes SF (91%). $^{261}\text{Rf}^a$, populated mostly in $^{265}\text{Sg}^a$ decay (80%), is known to α decay ($E_\alpha = 8.28$ MeV, $T_{1/2} = 68$ s) to ^{257}No . Consequently, α - α -SF or α - α - α correlations were expected as typical patterns of ^{269}Hs decays. The correlation criteria, which were applied, were two subsequent α decays within 300 s within an energy range of $8 \leq E_\alpha \leq 10$ MeV followed by another α decay ($8 \leq E_\alpha \leq 10$ MeV) or SF (at least one fragment > 20 MeV) within 300 s in the same or a neighboring detector pair. Because of α particle and SF background, only a three- or four-member chain was considered as a valid correlation. The number of expected random correlations mimicking a three- or four-member ^{269}Hs decay chain was 0.1 [17].

During the course of the experiment, none of the above mentioned ^{268}Hs or ^{269}Hs like correlations were detected. The yield of short-lived α decaying Os isotopes was monitored during the course of the experiment to ensure that all experimental conditions were optimized for HsO_4 synthesis and detection. The overall efficiency to detect 9.7-s ^{269}Hs as a three- or four-member decay chain is 51% [17], resulting in a one event cross section limit of 0.4 pb (63.2% confidence

FIG. 3. One event cross section limit for ^{268}Hs synthesized in the $5n$ evaporation channel in the fusion reaction $^{25}\text{Mg} + ^{248}\text{Cm}$ at a beam energy of $E_{\text{lab}} = 140$ MeV. The cross section limit is shown as a function of the unknown ^{268}Hs half-life. There is more than a 63.2% probability that the true cross section is below the derived function.

limit [24]). The unknown nuclide ^{268}Hs is expected to decay by α particle emission, with a half-life of ~ 0.5 s. Because the predicted half-life of ^{268}Hs (~ 0.5 s) is comparable to the mean transport time (1 s), the overall efficiency to detect ^{268}Hs can be expressed only as a function of its half-life. Figure 3 presents the $^{248}\text{Cm}(^{25}\text{Mg}, 5n)^{268}\text{Hs}$ cross section limit calculation for a mean transport time of 1 s. The calculated detection efficiency for ^{268}Hs with an assumed $T_{1/2} = 0.5$ s was 15%. This corresponds to a one event cross section limit of 1.3 pb (63.2% C.L.).

To summarize, it was shown that transport times as short as one second can be achieved in chemistry experiments, which makes such technique suitable for studying isotopes of superheavy elements with short half-lives. Despite the experimental improvements, no decay chains from Hs nuclei

were observed in the $^{248}\text{Cm}(^{25}\text{Mg}, 4n-5n)^{268,269}\text{Hs}$ reaction. A one event cross section limit of 0.4 pb for the $4n$ evaporation channel was reached in 170 h of irradiation. This cross section limit is below the value predicted by HIVAP [6] (0.6 pb). From the nonobservation of ^{268}Hs it follows, that either α decay is not the preferred decay mode of ^{268}Hs , or the half-life of ^{268}Hs is shorter than 0.5 s, or the cross section of the $^{248}\text{Cm}(^{25}\text{Mg}, 5n)^{268}\text{Hs}$ reaction at 140 MeV is lower than 1.3 pb.

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