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Short Note

On the decay properties of ^{269}Hs and indications for the new nuclide ^{270}Hs

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Abstract. In bombardments of ^{248}Cm with 143.7–146.8 MeV ^{26}Mg ions the nuclides ^{269}Hs and presumably ^{270}Hs were produced. After chemical isolation, Hs atoms were identified by observing genetically linked nuclear-decay chains. Three chains originating from ^{269}Hs confirmed the decay properties observed previously in the decay of ^{277}112. Two chains exhibited the characteristics expected for the new nuclide ^{270}Hs, which was predicted to be a deformed "doubly magic" nucleus. From the measured $E_\alpha = 9.16^{+0.07}_{-0.03}$ MeV an α-decay half-life of $3.6^{+0.8}_{-1.4}$ s was estimated.

PACS. 23.60.+e Alpha decay – 25.70.Gh Compound nucleus – 25.85.Ca Spontaneous fission – 27.90.+b $220 \leq A$

1 Introduction

Element 108 (hassium, Hs) was discovered in 1984 by Münzenberg *et al.* [1] using the heavy-ion fusion reaction ^{208}Pb(^{58}Fe, 1n)^{265}Hs. Until today, $^{264–267}$Hs and ^{269}Hs have positively been identified [1–5]. Furthermore, ^{263}Hs as a daughter of ^{267}110 [6] and one spontaneous-fission (SF) decay of ^{277}Hs as a decay product of ^{289}114 have been reported [7]. Two decays of ^{269}Hs have been observed in the decay chain of ^{277}112 [5,8,9]. In macroscopic-microscopic calculations increased stability was predicted at and near $N = 162$ and $Z = 108$ [10,11]. This new area of increased stability corresponds to gaps in the single-particle spectra which appear only at deformed shapes. By using a larger deformation space and a dynamic treatment of the fission barriers, Patyk *et al.* [10,11] analyzed the single-particle spectra of heavy nuclei and predicted ^{270}Hs to be a relatively strongly bound "doubly magic" deformed nucleus. They predicted rather long SF half-lives in this region. Therefore, ^{270}Hs is expected to decay predominantly by α-particle emission with $Q_\alpha = 9.44$ MeV [11]. In the most recent work, this value was slightly reduced to $Q_\alpha = 9.13$ MeV [12], resulting in a partial α-decay half-life of about 6 s, whereas the partial SF decay half-life was predicted to be 1.8 h [13]. Decay energies of $Q_\alpha = 8.7–9.9$ MeV have been derived from mass models [14–16], $Q_\alpha = 9.0$ MeV was calculated in the framework of the relativistic mean-field theory [17]. It is therefore of great interest to experimentally establish the decay properties of this nucleus. We report here on our experiments to chemically identify Hs, which resulted in the

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observation of ^{269}Hs and evidence for the production of the new nuclide ^{270}Hs.

2 Experimental

In our experiments ^{269}Hs and presumably ^{270}Hs were produced via the ^{248}Cm(^{26}Mg, 5n) and ^{248}Cm(^{26}Mg, 4n) reactions. The Universal Linear Accelerator (UNILAC) at the Gesellschaft für Schwerionenforschung, Darmstadt, provided very intense beams of up to 6.6 eμA 192.7 MeV $^{26}Mg^{5+}$. The beam entered the target chamber through a rotating 3-segment 3.68 mg/cm^2 Be vacuum window and the 2.82 mg/cm^2 Be target backing, before passing through the target material. The 3 banana-shaped ^{248}Cm targets (^{248}Cm 95.8 %, ^{246}Cm 4.2 %) with an average thickness of 239 μg/cm^2, 730 μg/cm^2, and 692 μg/cm^2, were prepared on Be foils by molecular plating. The ^{248}Cm target material was obtained by chemical separation from a ^{252}Cf neutron source [18]. The rotating vacuum window allowed for pressures of up to 1.3 atm in the target chamber. The target wheel rotated in the gaseous atmosphere of the target chamber with 2000 rpm. The rotation was synchronized with the beam pulses of the UNILAC in order to evenly distribute each 6 ms beam pulse over one of the three segments of the wheel. The 192.7 MeV beam energy of $^{26}Mg^{5+}$ from the accelerator resulted in projectile energies in the ^{248}Cm targets of 143.7–146.8 MeV.

In order to investigate the chemical properties of Hs, the gas chromatographic separation system IVO (*In situ* Volatilization and On-line detection) was developed and tested with short-lived α-particle emitting Os isotopes [19]. Element 108 is expected to belong to group 8 of the periodic table (like Os) and to form stable HsO_4 molecules, which should be volatile even at room temperature [20]. Nuclear-reaction products recoiling from the target are stopped in He containing 7.7% O_2. Os and presumably Hs are converted *in situ* to volatile tetroxides. The recoil chamber was continuously flushed at a flow rate of 1300 ml/min. At the exit of the recoil chamber the gas passed through a quartz column containing a quartz wool plug which was heated to 873 K. This quartz wool plug served as a filter for aerosol particles and provided a surface to complete the oxidation reaction of Os and Hs. Volatile species were then transported through a 10 m long Teflon (PFA) capillary (i.d. 2 mm) to an on-line detection system. From geometrical considerations, the transport time to the detection system was estimated to be between 1.5 and 3.0 s.

In order to efficiently detect the nuclear decay of isolated Hs nuclides, as well as to obtain chemical information about the volatility of the investigated compounds, a cryo thermochromatography detector was used [21]. The gas mixture containing volatile HsO_4 molecules is flowing through a narrow channel formed by a series of planar silicon diodes. Along this channel a longitudinal negative temperature gradient is established. The position at which the characteristic α-particle decay chain originating from the decay of a Hs nuclide is registered is indicating the deposition temperature of this molecule on the diode surface.

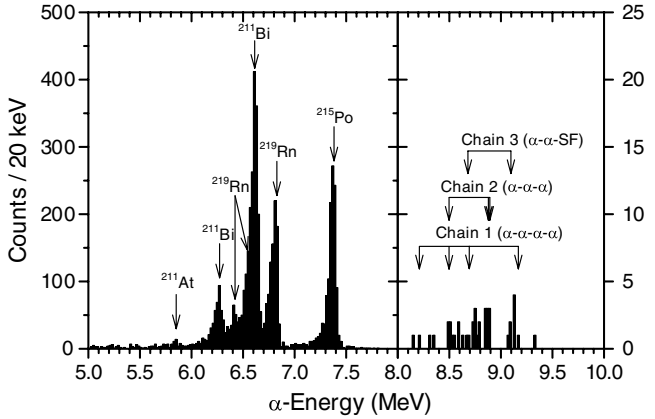

Fig. 1. α-particle spectrum of section 3 recorded during 64.2 h of beam (1.0×10^{18} ^{26}Mg ions).

Due to the close proximity of the silicon diodes facing each other, the probability to register a complete decay chain is rather high. A drawback of our method in comparison to kinematic separators is the lack of an initial implantation signal of the mother nucleus, which allows to measure the lifetime of the mother nuclide. In our apparatus the time at which a deposition of a HsO_4 molecule took place could not be detected, only the lifetimes of the daughter nuclei were measured. A cryo thermochromatography system named COLD (Cryo On-Line Detector) using an array of 2 × 36 PIN-diodes (Positive Implanted N-type silicon, 10 × 10 mm active area) was built. The COLD detector is an improved version of the Cryo Thermochromatographic Separator (CTS) [21] which was pioneered at Lawrence Berkeley National Laboratory. Two PIN-diodes mounted on ceramic supports (XRA-100, Detection Technology, Inc.) were glued together facing each other. Two T-shaped spacers made from silicon confined the gas flow to the active surface of the diodes. The gap in the channel was 1.5 mm wide. Always 3 neighboring PIN-diodes were electrically connected to form one detector unit. The COLD array consisted therefore of 12 sections each containing one pair of detector units, denoted with top (T) and bottom (B). The temperature ranged from 243 K down to 93 K. The detector units of the COLD array were calibrated with α-decaying ^{219}Rn (and its daughters ^{215}Po and ^{211}Bi) emanating from an ^{227}Ac source. The resolution was about 50–70 keV (FWHM) for detector units 1 through 8 (T and B) and about 80–110 keV (FWHM) for detector units 10 through 12 (T and B). The usually better resolution of PIN-diodes was degraded due to two effects. First, by electrically coupling 3 diodes together and, second, by the fact that about 30% of the registered α-particles were emitted by nuclides adsorbed on the opposite detector unit. Therefore, the energy of these α-particles was degraded by the He/O_2 atmosphere in the gap between the diodes. This energy loss was typically about 40 keV unless the α-particles were emitted under very shallow angles to the diode surface. The uncertainties of the α-decay energies in this work amount to typically $^{+70}_{-30}$ keV. A typical α-particle spectrum of section 3 is shown in fig. 1.

3 Results and discussion

In the course of the experiment, data was collected during 64.2 h and a beam integral of 1.0×10^{18} ^{26}Mg ions was accumulated. The count rate in all sections was very low. Only ^{219}Rn, ^{220}Rn, ^{211}At and their decay products were identified as background activities. While ^{211}At and its decay product ^{211}Po were deposited mainly in the first two sections, ^{219}Rn and ^{220}Rn and their decay products accumulated in the last three sections, where the temperature was low enough to condense Rn. Since detector unit 1B was not operating, section 1 was excluded from the data analysis. The average count rate per section was 0.6 h^{-1} in the relevant energy window $E_\alpha = 8.0$–9.5 MeV in sections 2 through 10. The data analysis revealed one four-member and 4 three-member decay chains each having a length ≤ 70 s. The four-member decay chain could unambiguously be attributed to the decay of ^{269}Hs. Also, the chain consisting of 3 α-particle decays must be attributed to ^{269}Hs. The remaining 3 three-member decay chains, each having a similarly low probability to be of random origin, must for chemical reasons be attributed to an isotope of Hs. From the calculated excitation functions of the reaction ^{26}Mg + ^{248}Cm [22], these are ^{269}Hs or ^{270}Hs at a beam energy of 143.7–146.8 MeV. Based on the poorly or unknown decay data of ^{269,270}Hs and their decay products, one of the remaining 3 decay chains was tentatively attributed to ^{269}Hs and two were tentatively assigned to the decay of the new nuclide ^{270}Hs (fig. 2).

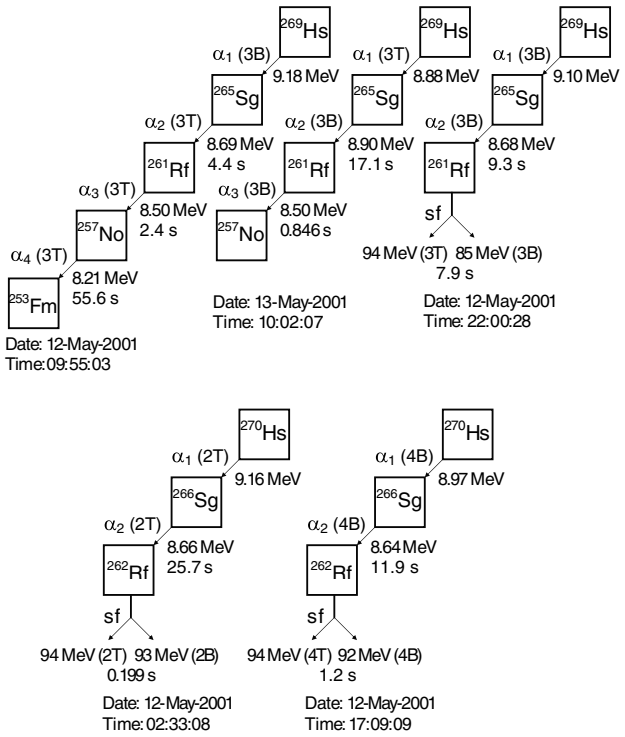

Fig. 2. Observed decay chains attributed to the decay of ^{269}Hs and ^{270}Hs produced in the reaction ^{26}Mg + ^{248}Cm.

The first decay chain of ^{269}Hs consisted of four α-particles occurring within 62.3 s in section 3. Based on the count rate in the energy window $E_\alpha = 8.0$–9.5 MeV in this section, the expected number of random four-member decay chains within 62.3 s not related to the decay of ^{269}Hs was calculated as $N_R \leq 2 \times 10^{-6}$. The first α-decay with $E_\alpha = 9.18$ MeV was followed by $E_\alpha = 8.69$ MeV ($\Delta t = 4.4$ s), in good agreement with the expected decay properties of ^{265}Sg. Decay energies $E_\alpha = 8.69$–8.94 MeV and a half-life of $7.4^{+3.3}_{-2.7}$ s have been measured for this nuclide [23,24]. The chain continued through ^{261}Rf ($E_\alpha = 8.50$ MeV, $\Delta t = 2.4$ s) and ^{257}No ($E_\alpha = 8.21$ MeV, $\Delta t = 55.6$ s). The last decay agrees well with literature data for ^{257}No [25]. However, ^{261}Rf is known to decay by the emission of 8.28 ± 0.02 MeV α-particles [26] with a half-life of 78^{+11}_{-6} s [27]. While in these earlier works the observation of 8.5 MeV α-particles could not be ruled out due to the presence of contaminants, the work of Lazarev *et al.* [28], where 69 ^{261}Rf-^{257}No correlations were registered, clearly showed no evidence for this α-decay energy. Nonetheless, the properties of our decay chain are in very good agreement with the ones observed for ^{269}Hs in the first decay chain of ^{277}112 produced in the reaction ^{208}Pb(^{70}Zn, 1n) [9]. There, an α-decay energy of 9.23 MeV was observed for ^{269}Hs. A striking feature was the observation of $E_\alpha = 8.52$ MeV for ^{261}Rf with a relatively short lifetime of 4.7 s [9], which now has also been observed in this work.

Two further decay chains attributed to ^{269}Hs were observed in the same section 3, *i.e.* at the same deposition temperature. In one decay chain the decay of ^{257}No was missed. This chain started with $E_\alpha = 8.88$ MeV, which is a relatively low decay energy for ^{269}Hs. Since it was not possible to determine on which side of the detector sandwich $^{269}HsO_4$ was deposited, it is conceivable that this α-particle lost a considerable part of its energy in the He/O_2 atmosphere. The decay chain proceeded through ^{265}Sg ($E_\alpha = 8.90$ MeV, $\Delta t = 17.1$ s) and through ^{261}Rf ($E_\alpha = 8.50$ MeV, $\Delta t = 846$ ms)($N_R \leq 7 \times 10^{-5}$). It appears as if in the decay of ^{269}Hs a different state of ^{261}Rf is populated. This is corroborated by the third observed decay chain, which is very similar to the first one, but is terminated by SF of ^{261}Rf ($\Delta t = 7.9$ s) ($N_R \leq 7 \times 10^{-6}$). The energies of the fission fragments have not been corrected for pulse height defects in the detectors and for neutron emission. The uncertainties associated with the fission fragment energies were estimated to be ± 6 MeV. The decay data of this chain is again in good agreement with a second decay chain observed recently for ^{277}112, which was also terminated by SF of ^{261}Rf with a lifetime of 14.5 s [8]. For the so far known 78 s ^{261}Rf, an upper limit for SF of $\leq 10\%$ was established [26]. Since $E_\alpha = 8.52$ MeV fits better into the systematics of ground-state α-decay energies, Hofmann *et al.* [9] tentatively assigned the α-decay and the SF event measured in their ^{277}112 decay chains to the ground state of ^{261}Rf. With the three additional events observed in this work, a half-life of $4.2^{+3.4}_{-1.3}$ s and a SF branch of about 40% is calculated for the decay of ^{261}Rf, whereas the previously known 78 s state should be denoted with ^{261m}Rf.

In our experiment, two decay chains exhibiting the expected decay characteristics of ^{270}Hs were observed. The so far unknown ^{270}Hs is expected to decay via

α-particle emission to the known nuclide ^{266}Sg (E_α = 8.63 ± 0.05 MeV) [23] which decays to ^{262}Rf ($T_{1/2}$ = 2.1 ± 0.2 s), a short-lived SF nuclide [29]. The first decay chain with E_α = 9.16 MeV, followed by E_α = 8.66 MeV (Δt = 25.7 s) and SF (Δt = 199 ms) occurred in section 2 ($N_R \leq 8 \times 10^{-6}$). Nearly identical energies were measured for both fission fragments. A similar decay chain starting with E_α = 8.97 MeV followed by E_α = 8.64 MeV (Δt = 11.9 s) and SF (Δt = 1.2 s) was observed in section 4 ($N_R \leq 5 \times 10^{-5}$). Again, nearly identical fragment energies were measured. In both decay chains the α-decay energy attributed to ^{266}Sg is in agreement with literature data [23]. The short correlation times of both SF events and the symmetric energy division among the fragments point to ^{262}Rf as terminating nuclide. The nuclide ^{262}Rf is known to fission symmetrically [29]. From the measured $E_\alpha = 9.16^{+0.07}_{-0.03}$ MeV attributed to ^{270}Hs, $Q_\alpha = 9.30^{+0.07}_{-0.03}$ MeV was calculated in good agreement with predictions [11,12]. Using the relationship between Q_α of ground state to ground state α-decays of even-even nuclides and the nuclear half-life by Buck *et al.* [30], resulted in $T^\alpha_{1/2}(^{270}\text{Hs}) = 3.6^{+0.8}_{-1.4}$ s. The formula of Buck *et al.* [30] reproduces the half-lives of known even-even nuclides within a factor of ∼ 2.

Since only about 77% of the inner surface of the COLD channel consisted of active detector surface, detection of a few incomplete decay sequences is expected. Two α-SF correlations were observed in sections 3 and 4 that still have a rather low random probability, but could not be assigned with certainty to either ^{269}Hs or ^{270}Hs. These were in section 3: E_α = 9.14 MeV followed by SF (108 MeV, only 1 fragment, Δt = 42.6 s) ($N_R \leq 4 \times 10^{-4}$) and in section 4: E_α = 8.72 MeV followed by SF (53 MeV, only 1 fragment, Δt = 3.1 s) ($N_R \leq 2 \times 10^{-4}$). Also, 4 uncorrelated SF decays with fragment energies ≥ 50 MeV were registered in sections 2, 3, and 4. All other sections 5 through 12 registered zero SF events. This may point to a SF branch in either ^{269}Hs or ^{270}Hs. Also, no three-member α-decay chains were observed in sections 5 through 10 with a length ≤ 300 s.

Assuming an overall efficiency of ≈ 40%, a production cross-section of 4 pb for α-decaying ^{270}Hs and of 6 pb for α-decaying ^{269}Hs was calculated at 143.7–146.8 MeV beam energy. The cross-sections are reported with an estimated accuracy of a factor of ∼ 3.

The isotopes ^{269}Hs and presumably ^{270}Hs were identified after chemical separation by observing genetically linked decay chains. The lower end of the decay chain of ^{277}112 was clearly reproduced and our work strongly supports the claim of Hofmann *et al.* [5] that in their work indeed the new element 112 had been produced. The efficiency and sensitivity of chemical methods competed favorably with physical separator systems and showed that chemical methods allow to work on the 1 pb cross-section level. The chemical implications of our experiment are discussed in a separate publication [31].

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