Confirmation of the Decay of $^{283}112$ and First Indication for Hg-like Behavior of Element 112

R. Eichler[a][b][*], N.V. Aksenov[c], A.V. Belozerov[c], G.A. Bozhikov[c], V.I. Chepigin[c], R. Dressler,[a] S.N. Dmitriev[c], H.W. Gäggeler,[ab] V.A. Gorshkov[c], F. Haenssler[b], M.G. Itkis[c], V.Ya. Lebedev[c], A. Laube[a], O.N. Malyshev[c], Yu.Ts. Oganessian[c], O.V. Petruschkin[c], D. Piguet[a], P. Rasmussen[a], S.V. Shishkin[c], A.V. Shutov[c], A.I. Svirikhin[c], E.E. Tereshatov[c], G.K. Vostokin[c], M. Wegrzecki[d], and A.V. Yeremin[c]

[a]Paul Scherrer Institute, CH-5232 Villigen, Switzerland

[b]Department for Chemistry and Biochemistry, The University of Bern, CH-3012 Bern, Switzerland

[c]Flerov Laboratory for Nuclear Reactions, JINR, 141980 Dubna, Russia

[d]Insitute of Electron Technology, 02-668 Warsaw, Poland

Two gas phase adsorption chemistry experiments aimed at the chemical characterization of element 112 using its isotope $^{283}112$ have been performed at the Flerov Laboratory for Nuclear Reactions (FLNR) Dubna, Russia. The applied Insitu-Volatilization and On-line Detection (IVO) technique is a thermochromatographic system combining the determination of the deposition temperature of volatile elements on a surface along a temperature gradient with an efficient detection of the deposited species by event-by-event alpha and SF-fragment spectroscopy. Two possibilities to produce the isotope $^{283}112$ were used: 1.) the direct production reaction $^{238}U(^{48}Ca,3n)^{283}112$; 2.) the reaction $^{242}Pu(^{48}Ca,3n)$, where the primary product $^{287}114$, decays via alpha emission to $^{283}112$ with a half-life of 0.5 s. The chemistry experiments were aimed at a chemical identification of $^{283}112$ and an independent confirmation of its decay properties. In the direct reaction no decays related to $^{283}112$ were observed. However, two decay chains unambiguously attributed to the decay of $^{283}112$ were observed using the second production path. Previously reported observation of $^{283}112$ and ^{279}Ds and their decay properties were confirmed. From its thermochromatorgaphic deposition first thermochemical data were deduced for element 112, unveiling it as a typical group 12 element.

1. Introduction

Since 1999 34 new isotopes of superheavy elements (SHE) 112-118 have been produced at FLNR in ^{48}Ca induced nuclear fusion reactions [1–5]. Their long half-lives, e.g. $^{283}112$ (4 s) and $^{289}114$ (2 s) indicate the close vicinity to the island of stability of SHE (114

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protons, 182 neutrons) as it was proposed already in 1966/67 [6,7]. However, the unambiguous assignment of the atomic number Z of these new isotopes is difficult since the observed α-decay chains always end in a previously unknown region of nuclides, which all decay by the unspecific spontaneous fission (SF). As it has been shown earlier [8–11] chemical techniques are available, which can be used to chemically identify genetically linked decay products of SHE. This approach allows for the identification of the atomic number of one member of the chains by chemical means, thus securing the elemental assignments of the entire decay chain. The observed half-lives of the produced isotopes of elements 112 and 114 are long enough [4] to chemically investigate these elements with recently available gas phase chemical techniques (see for review [11]). Their chemical identification is very exciting from the point of view of the periodic table. The observed new isotopes belong to the transactinide series of elements. The heaviest among them are elements of the groups 12-18, having electrons in their outer s and p electron valence shells. These electrons are expected to be severely influenced by relativistic effects induced by the high nuclear charge of the atoms [12,13]. The shape of their orbitals changes and also the energetic degeneration of the p, d, and f orbitals is abrogated. Element 112 has a predicted electronic ground state configuration $Rn:5f^{14}6d^{10}7s^2$. The relativistic contraction leads to stronger binding of the spherical 7s orbital. However, the resulting screening of the nuclear charge may lead to an expansion of the 6d orbitals and their looser binding. Therefore, a chemical behavior between noble-gas like and metallic was predicted for element 112 [14–22]. It was already suggested in 1976 that an exceptionally good possibility to reveal metallic properties of SHE will be the investigation of their interaction with metal surfaces [22]. The idea was that the interaction potential between SHE and transition metals promotes an intermetallic bond formation. If not, this element would simply show a physisorption interaction comparable to a noble gas, such as radon. Predicted thermodynamic properties regarding sublimation and adsorption of element 112 on a gold surface and the corresponding data for Zn, Cd, Hg, and Rn are summarized in table 1. The primary observation of a 5 min $^{283}112$ decaying by SF [1,2] in the nuclear fusion reaction $^{238}U(^{48}Ca,3n)$ initiated two types of chemistry experiments aimed at the investigation of element 112. 1.) The observation of 8 SF events in an ionization chamber coupled to a detector array with gold covered detectors held at room temperature was interpreted as an observation of a noble-gas like $^{283}112$ [23,24]. 2.) The IVO technique developed together with the Cryo On-Line Detector (COLD) proved to be successful for the chemical identification of Hassium (element 108) in the chemical state of the very volatile HsO_4 [8]. This technique required some modifications to be used for the investigation of chemical properties of element 112 [25,26]. The observation of high energetic events accumulating in the deposition region of radon in thermochromatography experiments performed with a 2-π detector array Cryo-On-Line Detector (COLD) at the Gesellschaft fr Schwerionenforschung, Darmstadt, Germany (GSI) [27] seemed to support the observation from [24]. However, these events could not be unambiguously assigned to SF decay [26]. Moreover, the observation of new decay properties for $^{283}112$ at the Dubna Gas filled Recoil Separator (DGFRS) as being 9.5 MeV alpha emitter with a half-life of 4 s [4] questioned the results. A second thermochromatography experiment with an improved version of the COLD detector (almost 4π-detection geometry) was performed at GSI. The experiment was sensitive to the reported new decay properties of $^{283}112$. However,

the sensitivity required to observe [283]112 was not reached [26]. Here we report on two further chemistry experiments joining the capabilities of the IVO thermochromatography method using the 4π-COLD detector with the availability of high [48]Ca beam intensities and long beam times at FLNR. The first experiment aimed at the direct production of [283]112 using the [48]Ca on [238]U fusion reaction. During the second experiment the isotope [283]112 was produced indirectly over the alpha decay of [287]114, which was formed in the reaction [242]Pu([48]Ca,3n) [4].

Table 1
Thermodynamic data for elements Hg, Rn, and element 112: Sublimation enthalpy ΔH_{subl} and adsorption enthalpy ΔH_{ads} on gold.

Element	Reference	$-\Delta H_{ads}(Au)$, kJ/mol	ΔH_{subl}, kJ/mol	Method
112	[22]	22		Empirical
112	[19]	84		Semi-empirical
112	[20]		39	Empirical
112	[29]	24		Empirical
112	[16]	$\sim$ 80		Calculation
112	[17]	$\sim$ 65		Calculation
112	[24]	< 60		Experiment
Rn	[29]	29±3	22	Experiment
Hg	[25]	98±3	64 [32]	Experiment
Zn	[32]		130	Experiment
Cd	[32]		111	Experiment

2. Experimental

2.1. Experiment with the [238]U targets

A grid supported stationary 1.4 mg/cm^2 [238]U target was irradiated for about 4 weeks with a total beam dose of 4.5*10^{18} particles [48]Ca at a center of target (COT) energy of 237 MeV. [nat]Nd (15 μg/cm^2) was added to the target in order to produce α-decaying [185]Hg, which has a half-life of 49 s. The collimator grid supporting the 4μm Ti vacuum window had a beam transmission of 55%. The uranium target was mounted on a copper cooling grid facing the incident beam. The recoiling products had to pass the 1.5 μm Ti target backing and subsequently this grid. An average loss of 20% due to the target grid was determined. The volatile products were transported by a 900 ml/min carrier gas mixture of He/Ar 70%/30% through an tubular oven with a quartz tube filled with a quartz wool plug and with Ta metal held at 850 °C. This arrangement ensured the removal of the aerosol particles produced by beam induced sputtering on the beam dump and the removal of traces of oxygen and water from the carrier gas. The volatile atomic and molecular

species were transported through an 8 m long PFA capillary to the thermochromatography detector COLD. Several α-decaying Rn isotopes ($^{219-221}$Rn) are produced in the ^{48}Ca induced nuclear fusion reaction with ^{238}U at high cross sections. Thus, Rn and Hg have been studied simultaneously with element 112 throughout the entire experiment. The COLD detector provided an on-line thermochromatogram of the adsorption of mercury (^{185}Hg) and radon (^{219}Rn) on a gold surface, covering a temperature gradient of -5 °C/cm from -24 °C down to -184 °C (see Figure 1A). The transport time was measured for ^{185}Hg as 3.6 s. The decay losses during the transport account for a transport efficiency of about 54% for a volatile species having a half-life of 4 s. The deposition efficiency was 100% and 88% for ^{185}Hg and ^{219}Rn, respectively. The detection efficiency for the ^{283}112(α)-^{279}Ds(SF) correlation was 75%. The effective target thickness was assumed to be about 1.0 mg/cm^2 with respect to the shape of the excitation function of the reaction ^{238}U(^{48}Ca,3n)^{283}112 as reported in [4]. However, the target material losses due to beam sputtering were considerable. These losses could be estimated using the absolute yield drop of ^{185}Hg during the irradiation. This drop was accounted for additionally as a virtual effective target thickness loss of 44% (see table 2). The target was changed once in a week. Together with the losses in the target cooling grid the overall efficiencies of the experiment of about 10% and about 9% were estimated for a Hg-like and for a Rn-like ^{283}112, respectively.

2.2. Experiments with the ^{242}Pu targets

A ^{242}Pu target (1.4 mg/cm^2) without grid support was irradiated with 3*10^{18} ^{48}Ca particles at a centre of target energy of 237 MeV using an otherwise same target assembly as in the experiments with the ^{238}U target. *nat*Nd (15 μg/cm^2) was added to the target material. Due to beam sputtering the target material losses were about 51%per week. Therefore, the target was changed about once in a week. In the reaction ^{242}Pu(^{48}Ca,3n)^{287}114 the isotope ^{283}112 is only indirectly formed via the α-decay of ^{287}114(T$_{1/2}$=0.5 s, E$_\alpha$=10.08 MeV). ^{287}114 is too short-lived to be transported to the COLD detector in this experiment. Therefore, it decays either in the recoil chamber or in the transport line. Perhaps, element 114 is adsorbed to a surface. Therefore, a recoil implantation of its alpha decay product ^{283}112 has to be assumed, lowering the transport yield for this isotope additionally by up to 50%. The experimental setup was kept the same as in the experiment with the ^{238}U target. The overall efficiency of the experiment was about 12% and 8% for a Hg-like and for a Rn-like ^{283}112, respectively. The experiment was started (Part A) establishing a temperature gradient of -5 °C/cm between -24 °C down to -184 °C (see Figure 1A) in the COLD. Later on (Part B) a temperature gradient of about -6.5 °C/cm between +35 °C and -180 °C (see Figure 1B) was installed.

3. Results and Discussion

3.1. Experiments with the ^{238}U targets

A typically measured distribution of ^{185}Hg and ^{219}Rn along the detector at these conditions is shown in the thermochromatogram (see e.g. Figure 1A, grey and white bars, respectively). From the distribution of atoms on a chromatographic surface along a temperature gradient it is possible to quantify the enthalpy of the adsorption interaction (-ΔH$_{ads}$) of the elements on the surface using the microscopic Monte-Carlo based model

of gas adsorption chromatography [30]. The Hg-isotopes were completely deposited on the gold-surface in the first 8 detectors down to -60 $^\circ$ C. This spontaneous, diffusion controlled deposition pattern of Hg on Au allows the determination of a lower limit of ΔH_{ads}^{Hg}(Au)> 47 kJ/mol. This value agrees well with previous observations [25] (see table 1). ^{219}Rn was deposited on the last 5 detectors peaking on detector 28 at a temperature of -170 $^\circ$ C. For radon the deposition distribution is reproduced by the Monte-Carlo model with an adsorption enthalpy of $-\Delta H_{ads}^{Rn}$(Au) $= 20\pm1$ kJ/mol, which is lower than data given in literature [29] (see table 1). The dew point of water in the carrier gas was measured as about -100 $^\circ$ C. This corresponds to water content of less than 0.1 ppm. We can therefore assume that the Au surface is covered by a thin ice layer at the very low temperatures (see Figure 1A). The determined adsorption enthalpy of radon on ice is therefore in good agreement with literature data [31]. During four weeks of irradiation no decays related to ^{283}112 were detected and no SF decays at all.

Table 2
Cross sections (σ) reported so far for ^{283}112 investigated in chemistry experiments directly produced in the reaction of ^{48}Ca with ^{238}U and from this work.

Ref.	Beam dose x10^{18}	Target thickness (effective) x10^{18}	Chemistry efficiency %	Energy (COT) MeV	Decay properties of ^{238}112 ($T_{1/2}$)		N^a
					α-SF (4 s)	SF (5 min)	
					σ, pb		
[24]	2.8	2.5	50	231	$> 7^b$	$2_{-1.2}^{+2}$ ^{c}	8
[26]	2.8	2.0	61.6	238	$< 2.3^c$	$< 0.8^c$	0
	1.4	2.0	44.4	241		$< 1.7^c$	
[this work]	4.3	1.4 ^{d}	17.6	237	$< 2.5^c$	$< 1.3^c$	0

^{a} Number of observed events attributed to the decay of ^{283}112

^{b} Some of the observed SF-decays in [24] were hypothetically attributed to the less efficient transport of the α-decaying ^{283}112(4 s) followed by SF of ^{279}Ds(0.2 s) [4];

^{c} Upper limit sensitivities at 95.45% c.i.;

^{d} accounting for a target material loss during the irradiation, which was quantified using ^{185}Hg yield, an effective target thickness of 1.4*10^{18} was calculated from the initially assumed effective target thickness of 2.5*10^{18} atoms.

In Table 2 the production cross sections were evaluated in dependence on the expected decay properties for ^{283}112 for all chemistry experiments. Summarizing the results from the two gas phase chemical experiments, which were sensitive to detect a 4 s α-decay of ^{283}112 followed by 0.2 s SF-decay of ^{279}Ds from [26] and this work it is possible to give an upper limit cross section of about 1.3 pb (95 c.i.) for the nuclear reaction ^{238}U(^{48}Ca,3n)^{283}112..
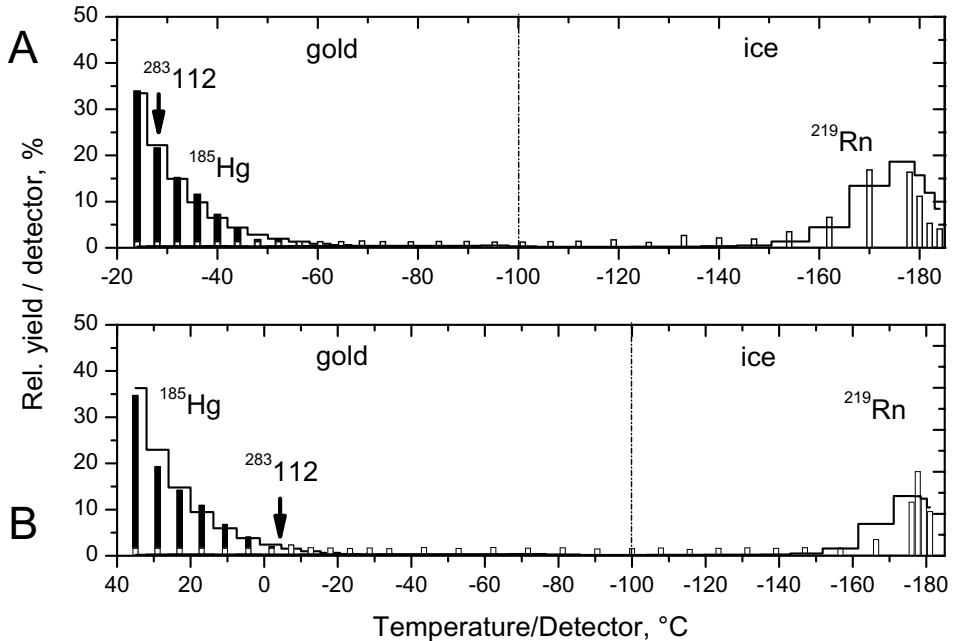
Figure 1. Thermochromatograms of ^{185}Hg (grey bars), ^{219}Rn (white bars) and ^{283}112 (black arrows) on gold and ice surfaces of the COLD detector. Each bar for ^{185}Hg and ^{219}Rn represents the yield measured in one detector sandwich of COLD. The border between gold and ice is indicated (dashed-dotted line). The stepped lines represent the Monte Carlo analysis of the chromatographic behaviour using the microscopic model of adsorption chromatography [30].

3.2. Experiments with the ^{242}Pu targets

The temperature gradient in Part A of the experiment was established from -24 to -184 ° C aiming at an almost quantitative deposition as well as for a Hg-like as for a Rn-like species. Indeed, the entire ^{185}Hg and 88% of ^{219}Rn which reached the COLD detector were deposited (see Figure 1A). The ^{185}Hg and ^{219}Rn showed the same deposition pattern as measured in the experiment with the ^{238}U target yielding the same thermochemical data for the adsorption of both species on gold (Hg) and ice (Rn). In this part of the experiment the first decay chain unambiguously attributed to the decay of ^{283}112 was detected. It was a 9.37 MeV alpha particle followed after 0.592 s by a high energy spontaneous fission. Both fragments having energies of 108 MeV and 123 MeV were detected in the same detector sandwich as the preceding alpha. This was the first SF decay detected after overall 5 weeks of beam time. In the first 15 detectors there were only five alpha particles measured between 9.15 and 10.5 MeV during the 4 h run. Therefore, we conclude that the measured decay chain has a very low random probability of (¡4x10^{-4}). Its deposition temperature of -28 ° C directly in the deposition region of Hg (detector 2) (see Figure

1A) allows to clearly distinguish it from the radon deposition. Only a 0.5%/detector in flight decay of ^{219}Rn(T$_{1/2}$=3.96 s) is expected, which makes the observation of an in flight decay of ^{283}112 followed by deposition and decay of ^{279}110 very improbable. To possibly distinguish the element 112 adsorption behavior on gold also from the mercury adsorption properties, a new temperature gradient starting from +35 ˚C down to -180 ˚C (see Figure 1B) was established in Part B of the Experiment. The deposition behavior of ^{185}Hg and ^{219}Rn at these conditions is shown in Figure 1B. From the spontaneous diffusion controlled deposition pattern of ^{185}Hg a lower limit adsorption enthalpy of -ΔH_{ads}^{Hg}(Au)>67 kJ/mol was deduced, which is consistent with the experimental value determined for ΔH_{ads} on gold [25](see table 1). As expected, the ^{219}Rn deposition maximum moved towards the end of the detector. Hence, a substantial fraction of about 35% ^{219}Rn did not deposit in the COLD. The corresponding adsorption enthalpy of Rn on ice (-ΔH_{ads}^{Rn}(ice)=20±1 kJ/mol) is in excellent agreement with the previous observations. At these experimental conditions we observed a second correlated decay chain with the clear signature of the ^{283}112. A 9.47 MeV alpha decay was followed within 0.536 s by a high energy spontaneous fission decay in the same detector sandwich. The fission fragments had energies of 127 MeV and 105 MeV. This decay chain was measured in detector 7 at a temperature of -5 ˚C. Detector 7 was reached by only 5% of the ^{185}Hg i.e. 2% of the Hg was deposited on the detector and 3 % reached the detectors 8-14 (see Figure 1B). A statistical analysis of the observation of both atoms of $^{(}$283)112 using the Monte-Carlo technique [30] allowed for a direct quantification of the adsorption interaction of element 112 with a gold surface. An adsorption enthalpy of -ΔH_{ads}^{112}(Au)=52$^{+46}_{-7}$kJ/mol (68% c.i.) was deduced, which represents the first ever thermochemical value determined for element 112 [28]. This value is indicative for a metallic interaction of element 112 with gold surfaces.

4. Conclusion

The upper production cross section limit of ^{283}112 in the reaction ^{238}U(^{48}Ca,3n) from all IVO chemistry experiments sensitive to the 4s α-SF decay scheme is 1.2 pb (95% c.i.), which lower than the cross section of 2.5$^{+1.8}_{-1.1}$ pb reported in [4]. On the same statistical confidence level there is no evidence for a 5-min SF decaying ^{283}112 as it was reported in [3,2,24] to a cross section limit of 0.39 pb. The observation of the isotopes ^{283}112 and ^{279}Ds in this gas phase chemistry experiment confirm the reports on their production in the nuclear fusion reaction of ^{48}Ca and ^{242}Pu and the reported decay properties [4]. The results of this experiment confirm also the discovery of ^{287}114 and ^{291}116 reported in [4], since the chemical identification of ^{283}112 and the confirmation of its decay properties are determining the atomic numbers attributed to the members of the observed alpha decay chains passing ^{283}112 and ^{279}Ds. The significantly higher interaction of element 112 with a gold surface compared to radon, observed from the thermochromatographic deposition of ^{283}112 in the COLD detector, reveals a clear indication for a metallic bond formation of element 112 with the gold surface, suggesting it to be a typical group 12 element [28]. The observed transport of atomic element 112 through the chemical setup is indicative for its high volatility. Further, more sensitive experiments are envisaged with ^{285}112 produced in the ^{48}Ca on ^{244}Pu reaction [4].

5. Acknowledgements

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