

## Evidence for a very noble element 112

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In recent years, chemical properties have been investigated for transactinide elements up to hassium (Hs, element 108). It could be shown, that hassium forms a very volatile tetroxide [1] and that this tetroxide adsorbs strongly on a sodium hydroxide surface [2]. Both investigations confirm for hassium a typical behavior of a group 8 element in the transition element series of the periodic table.

Due to relativistic effects in the electron shells, increasing deviations of the periodicity of chemical properties along the rows of the periodic table are expected to occur for the heaviest elements. In this context element 112 is exceptional. From the systematics of the periodic table element 112 is expected to be a member of group 12. Hence, it should behave like a volatile metal similar to mercury. However, due to its closed shell  $[Rn]5f^{14}6d^{10}7s^2$  electronic structure and the mentioned relativistic effects, the relativistic stabilization of the  $7s^2$  electron orbital could be so strong that element 112 might behave like a noble gas, similar to radon [3,4].

Indeed from two experiments performed at Dubna [5,6] it was concluded that element 112 does not behave like mercury. Products from the reaction  $^{48}\text{Ca} + ^{238}\text{U}$  that form a long-lived spontaneously-fissioning isotope of element 112 ( $^{283}112$ ,  $T_{1/2} = 3$  min) [7-9] were continuously transported with a carrier gas to a counting device consisting of gold covered PIPS detectors followed by a gas-ionization chamber. Hg-isotopes produced in fusion reactions of  $^{48}\text{Ca}$  with the  $^{\text{nat}}\text{Nd}$  admixture in the target, were quantitatively adsorbed on the noble metal covered detectors, while element 112 was detected in the adjacent gas ionization chamber. Hence, this experiment yielded evidence that element 112 does not adsorb on gold at room temperature, and seems therefore to behave like a very inert metal or even like a noble gas.

In the studies of hassium tetroxide performed by our collaboration the IVO (In-situ Volatilization and On-line counting) device [10] was used. This technique permits *in-situ* thermochromatographic measurements of deposition temperatures of very volatile species on inert surfaces. Based on the expected inertness of element 112 we decided to apply a modified IVO technique for its chemical investigation. Products of the heavy ion induced nuclear fusion reaction recoiling out of the target were thermalized in dried 1 l/min helium carrier gas. The inner surface of the recoil chamber was completely covered by a quartz insert to prevent adsorption of volatile metallic reaction products. Gaseous products were swept out of the recoil chamber through an open quartz column to an oven with a quartz wool filter heated up to 850°C. Aerosol particles produced accidentally by e.g. beam particle induced sputtering processes in the beam dump were stopped in this filter. Separation factors of about  $10^7$  have been determined for lanthanides (model elements for heavy actinides) in test experiments with ytterbium, produced in the reaction  $^{142}\text{Nd}(^{20}\text{Ne};6n)^{156}\text{Yb}$ . Volatile products that passed this filter were transported through a 10 m long PFA-capillary to a Ta/Ti-getter kept at 1000°C. The getter served as trap for trace

amounts of water and oxygen. It was experimentally verified that the transport of carrier free amounts of mercury and radon occurred with almost no loss through the PFA or quartz capillary tubes at room temperature and through the Ta/Ti-getter. Finally, the gaseous products were injected into the modified detector COLD (Cryo On-Line Detector) [1]. This device consists of an array of 32 silicon PIN-photodiodes ( $10 \times 9.8 \text{ mm}^2$  active area) mounted in a Teflon coated copper channel at a distance of 1.6 mm opposite to a gold covered surface, forming a rectangular gas chromatographic column. The gold surface was produced by vapor deposition of gold on a copper frame. A negative temperature gradient from  $+35^\circ\text{C}$  to  $-187^\circ\text{C}$  (see Fig. 1, black line) was established along this chromatographic channel using a thermostat heating and a liquid nitrogen cooling. The whole detector set-up was placed in a vacuum tight steel box kept at 900 mbar to isolate it thermally against ambient air.

Event-by-event spectroscopy in a  $2\pi$ -detection geometry provided the identification of nuclides that were deposited on the gold surface. Calibration of the PIN-photodiodes was performed by  $\alpha$ -decaying  $^{219}\text{Rn}$  emanating from a  $^{227}\text{Ac}$  source and its daughters  $^{215}\text{Po}$  and  $^{211}\text{Bi}$ . The determined energy resolution was 40-70 keV except for a few detectors that suffered from a reduced resolution of only 100-180 keV. This new set-up was tested on-line with short-lived Hg-isotopes produced in nuclear fusion reactions of  $^{20}\text{Ne}$  and ytterbium (enriched in  $^{168}\text{Yb}$ ) at the PHILIPS cyclotron at the Paul Scherrer Institute and with  $^{219}\text{Rn}$  emanating from a  $^{227}\text{Ac}$  source. The efficiency of detecting a fission fragment was 77 %. With  $^{186-190}\text{Hg}$  and  $^{219}\text{Rn}$  an overall process efficiency of about 60 % and a transport time of less than 25 s was determined [11].

In February/March 2003 three banana shaped segments of a  $^{238}\text{U}$  target (average thickness  $1.6 \text{ mg cm}^{-2}$ ), prepared on  $2.33 \text{ mg cm}^{-2}$  thick Be foils, were mounted in the rotating target assembly ARTESIA [12] and were bombarded by  $1.9 \times 10^{12} \text{ }^{48}\text{Ca}^{7+} \text{ s}^{-1}$  at a Gesellschaft für Schwerionenforschung mbH/UNILAC energy of 346 MeV. After passing the  $3.7 \text{ mg cm}^{-2}$  Be vacuum window and the target backing-foil the  $^{48}\text{Ca}$  projectile energy was degraded to 239 MeV. The energy inside the  $^{238}\text{U}$  target was then 222 – 239 MeV. During 16.8 days  $2.8 \times 10^{18}$   $^{48}\text{Ca}$  particles were accumulated on the target. One of the three banana shaped segments of the  $\text{U}_3\text{O}_8$  target contained  $21.7 \text{ } \mu\text{g cm}^{-2} \text{ natNd}$  in order to produce  $\alpha$ -decaying Hg-isotopes, mainly in the reaction  $^{142}\text{Nd}(^{48}\text{Ca},4-6n)^{184-186}\text{Hg}$ . Since Rn-isotopes were produced in nuclear transfer reactions [13] it was possible to compare the chemical properties of element 112 with both mercury and radon during the entire experiment. The volatile reaction products were transported to the COLD detector with 1 l/min helium. The Hg-isotopes were completely deposited on the gold surface opposite the first nine detectors. About 65 % of  $^{220}\text{Rn}$  were adsorbed in the low temperature part of the COLD detector (see Fig. 1).

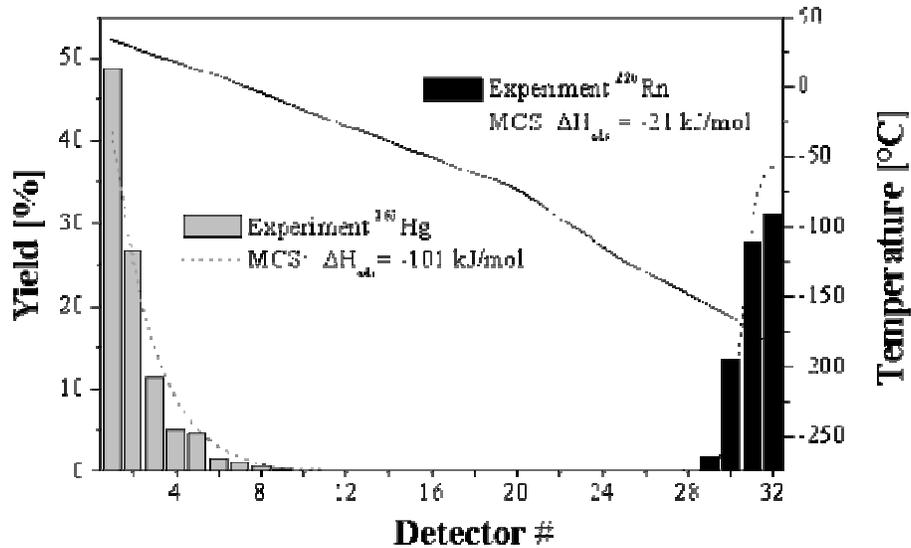


Figure 1. Thermochromatograms of  $^{185}\text{Hg}$  (grey bars, left-hand scale) and  $^{220}\text{Rn}$  (black bars, left-hand scale) on COLD at a helium carrier gas flow of 1 l/min. Also shown are Monte Carlo simulations of the observed depositions (grey and black dashed lines, left hand scale). The temperature gradient is indicated (black line, right-hand scale).

The spontaneous, diffusion controlled deposition of mercury on gold agrees well with literature values ( $\Delta H_{\text{ads}} = -101 \pm 2$  kJ/mol) [14]. For radon the deposition distribution is reproduced assuming an adsorption enthalpy of  $\Delta H_{\text{ads}} = -21 \pm 1$  kJ/mol. This value is somewhat lower compared to a literature value  $\Delta H_{\text{ads}} = -29 \pm 3$  kJ/mol for radon on gold as measured under molecular flow conditions [15]. We can therefore not exclude that at these very low temperatures the gold surface is covered by a thin ice layer. Indeed, in earlier thermochromatographic experiments of radon on ice surfaces an adsorption enthalpy of  $\Delta H_{\text{ads}} = -20 \pm 2$  kJ/mol was found [16]. Moreover, a careful analysis of the resolution of the  $\alpha$ -spectra yielded evidence for an ice formation at temperatures below about  $-90^\circ\text{C}$  (detector #21). This corresponds to a dew point of water at a partial pressure in the helium carrier gas of 0.2 ppm.

During the experiment 12 fission-like events (with energies above 40 MeV) were detected (Fig. 2). A background measurement performed after the run clearly indicated a non-zero background with scattered events along the entire array at a count-rate of approximately 0.0055 events/detector and day.

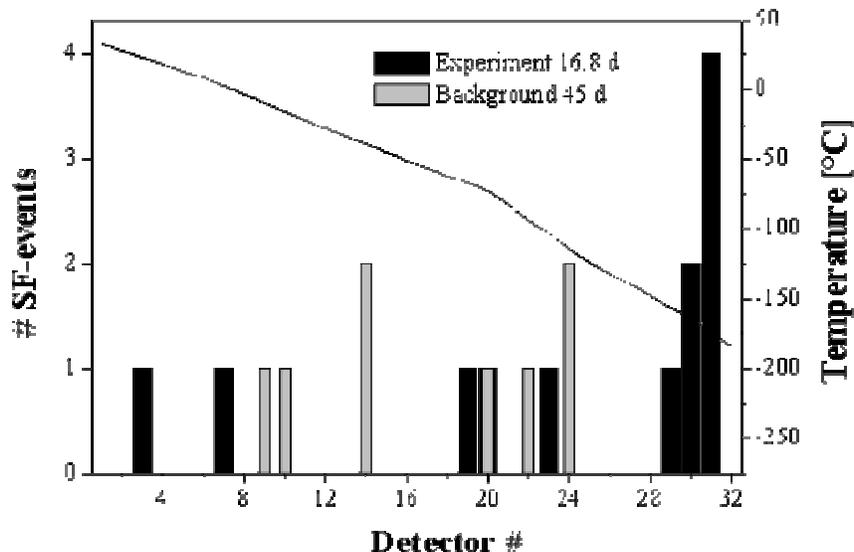


Figure 2. Observed events (with energies > 40 MeV) throughout the  $^{48}\text{Ca} + ^{238}\text{U}$  experiment (black bars, left-hand scale) and during the background measurement performed immediately after the run (grey bars, left-hand scale). The temperature gradient is indicated (black line, right-hand scale).

The assignment of the measured SF events to the decay of  $^{283}\text{112}$  is only convincing if no actinides are found along the detector array. Therefore, a very careful analysis of the  $\alpha$ -spectra was performed. First, the quartz wool filter kept at  $850^\circ\text{C}$  was leached with concentrated nitric acid. From the solution a sample was prepared to measure off-line  $\alpha$ - and SF-events. Pronounced peaks of  $^{226-228}\text{Th}$ ,  $^{224-226}\text{Ac}$ , and  $^{221-228}\text{Ra}$  were observed but no SF-decays detected during a counting time of 30 days (Fig. 3).

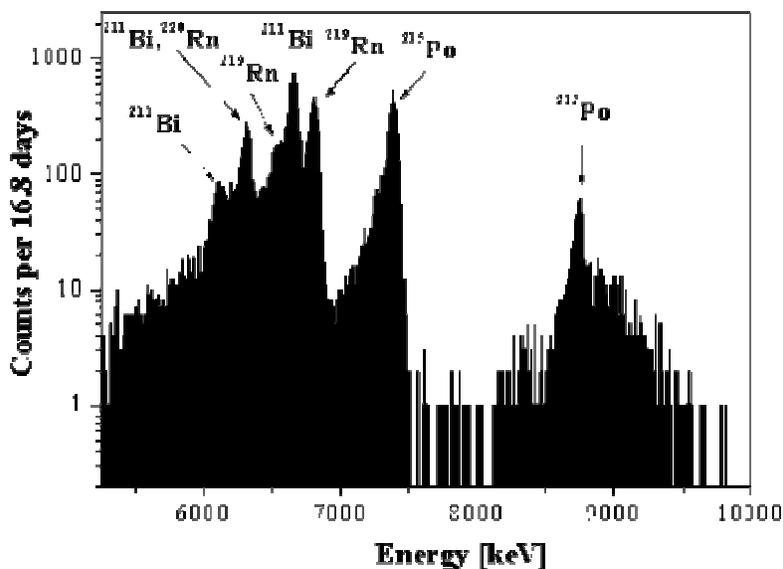


Figure 3.  $\alpha$ -spectra of detector # 21 in the energy-range between 5.3 MeV and 10 MeV.

In addition, a thorough analysis of the recorded 32  $\alpha$ -spectra did not indicate any  $\alpha$ -peak that might be assigned to a heavy actinide nuclide. Moreover, it is extremely unlikely, that any isotope of radon has a SF-branch. Hence, we assign most of the measured SF events to an isotope of element 112, presumably  $^{283}112$ .

In the detectors 29, 30 and 31 a total of 7 events were measured during the experiment at an expected background of 0.28 events. This deposition peak almost coincides with the deposition peak of radon. Already this preliminary data from the ongoing data analysis of the experiment allow us to conclude, that our experiment yielded strong evidence for a very noble element 112. The final results will be presented in our contribution.

## Acknowledgements

We would like to thank the crews of the Gesellschaft für Schwerionenforschung mbH UNILAC and of the Paul Scherrer Institut Philips Cyclotron for providing stable and intense beams of  $^{48}\text{Ca}$  and  $^{20}\text{Ne}$ . We are indebted to the staff of the Laboratory for Micro- and Nanotechnology at Paul Scherrer Institute for adapting the PIN-diodes for the COLD. These studies were supported in part by the Swiss National Science Foundation.

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