

# Optimizing the synthesis of SHE metal carbonyl complexes and preparing nuclear structure studies of $^{265}\text{Sg}$ : preparatory experiments at Tandem Accelerator, JAEA Tokai, and at TRIGA Mainz

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## Abstract

In a series of complementary experiments at the TRIGA Mainz and the JAEA tandem accelerator, the in-situ synthesis of volatile transition metal carbonyl complexes with short-lived radioisotopes has been investigated to develop a new approach, which promises to allow more efficient carbonyl-complex formation than the previously used combination of gas phase chromatography with a physical recoil separator. Using a setup with decoupled recoil ion thermalization and chemical complex formation in two separate volumes, the feasibility of the method was demonstrated using fusion-evaporation-produced radionuclides at JAEA Tokai and short-lived fission products generated at the research reactor TRIGA Mainz. According to the currently available preliminary data, superior efficiencies were obtained, validating this novel approach. In parallel, construction of the ALpha-BEta-GAMMA decay spectroscopy detection setup ALBEGA continued.

## 1. Research Objectives

In case of superheavy elements, the investigation of carbonyl complexes is of broad interest, as relativistic effects, which scale with  $Z^2$ , are predicted to strongly influence the chemical behavior

of these elements [1]. The bonding in carbonyl complexes emerges from electron pair donation from the CO molecules surrounding the central atom into its empty d orbitals and subsequent  $\pi$ -back bonding due to overlapping of the filled d orbitals with antibonding  $\pi^*$ -orbitals of CO [2]. Comparative studies of the transactinides with their lighter homologs help to understand the influence of relativity on the electron shell and test the predictive power of the periodic table. Currently, the method of choice is the isolation of the respective ions behind a recoil separator, followed by the chemical carbonyl complex formation [3], which allowed synthesis of  $\text{Sg}(\text{CO})_6$  [4]. However, the evaporation residues of asymmetric fusion reactions, needed for the productions of suitably long-lived isotopes of the transactinides Sg, Bh, and Hs have a wide angular and energy distribution, thus the transmission efficiency through a forward-angle recoil separator is relatively low. In case of RIKEN GARIS, which was used in the Sg work, this partial efficiency is in the order of 13% for Sg [5]. Thus, the overall efficiency of the synthesis of carbonyl complexes in combination with physical preseparation is rather low. For future experiments a more efficient concept is highly desirable due to decreasing production rates for elements heavier than Sg.

## 2. Research Contents

### 2.1. A novel synthesis approach for volatile carbonyl complexes of transition metal radioisotopes

The possibilities for the chemical investigation of these systems without a physical preseparation stage is currently explored within a so called two-chamber approach, whereby the non-volatile fusion-evaporation products will be flushed out to a second, directly connected reaction chamber. Scientific goal is to avoid the high losses of about 80-90% currently inherent to the technique using a physical preseparator. First experiments performed at the JAEA tandem accelerator at Tokai, Japan, suggested that the successful synthesis of Os and W carbonyl complexes is feasible if the thermalization of the evaporation residues is spatially decoupled from the chemical

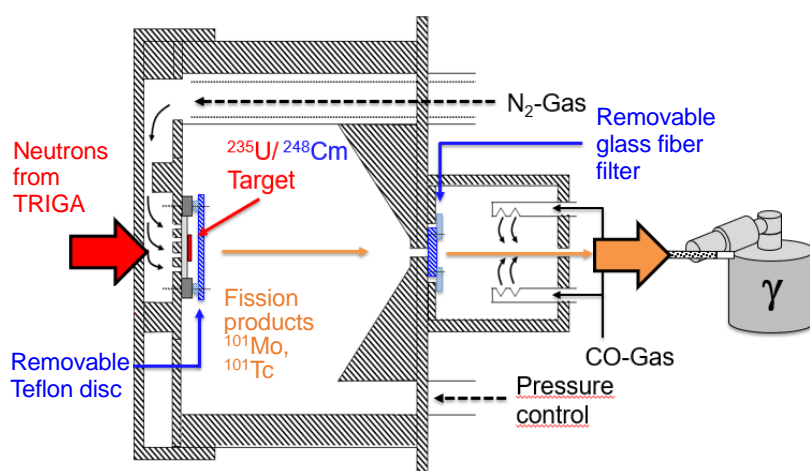


Fig 1. Schematic illustration of the experimental setup used for carbonyl chemistry studies at TRIGA Mainz (with a  $^{235}\text{U}$  target) and off-line (with a  $^{248}\text{Cm}$  fission fragment source).

synthesis. This allows forming the carbonyl complexes in the absence of beam. Further experiments were conducted using fission products obtained at TRIGA Mainz. Figure 1 illustrates the schematic structure of the two-chamber approach.

The fission products, either produced by neutron-induced fission (red, with a  $^{235}\text{U}$  target) or

by spontaneous fission (blue, with a  $^{248}\text{Cm}$  source), were thermalized in a gas-filled chamber behind the target in the thermalization chamber. To obtain absolute rates of fission fragments entering the thermalization chamber, a Teflon disk acting as a catcher for fission fragments emerging from the target could be mounted in close proximity covering the  $^{248}\text{Cm}$  source (see Fig. 1). In case of the on-line studies at TRIGA, previously obtained absolute rates were used [6]. The thermalization chamber was continuously flushed with inert gas in both cases. Directly at the exit of this thermalization chamber a reaction chamber was added, into which the fission products were flushed with the inert gas. In case of the flush-out efficiency studies, performed with the  $^{248}\text{Cm}$ -source, a removable charcoal filter was installed between the exit of the thermalization chamber and the entrance of the chemical reaction chamber. This filter was used to collect the non-volatile fission products that were flushed out of the thermalization chamber into the reaction chamber and was, after being removed, counted via  $\gamma$  spectrometry. For the chemical experiments, using the  $^{235}\text{U}$  target, carbon monoxide gas was injected into the second chamber, bypassing the thermalization chamber. The inert gas and the carbon monoxide were pre-cleaned by passing gas purification cartridges and hot titanium getters. Fission fragments volatile in their elemental form or forming volatile complexes with CO were transported with the gas stream through a Teflon capillary to a chemistry laboratory. Here the products were trapped on a charcoal filter and isotopically identified by examining the characteristic  $\gamma$  rays. All samples (Teflon disk; charcoal filter at the entrance of the chemical reaction chamber, and charcoal filter collecting volatile species after transport through the capillary) were counted in the same geometry to ensure identical efficiency.

## 2.2. On the way to the ALpha-BEta-Gamma multidetector system ALBEGA.

A next generation setup for measurements of ALpha-BEta-GAMMA decays (ALBEGA) after chemical isolation is currently under development. Building up on a first prototype [7], an advanced version, overcoming some limitations encountered during commissioning of the first version, is being built. It will allow nuclear spectroscopy studies of chemically isolated samples, including volatile carbonyl complexes. Therefore, this activity nicely complements the chemical studies described in Sect. 2.1.

## 3. Research results

### 3.1. Carbonyl compound synthesis

After first indications about the success of the approach were obtained at JAEA Tokai, more detailed and quantitative studies were carried out at TRIGA Mainz. Preliminary results from TRIGA for the total yield (comprising flush-out yield from thermalization chamber to reaction chamber and chemical yield for complex formation and transport to the external charcoal filter) as a function of gas composition (starting from a concentration ratio of  $\text{N}_2 : \text{CO}$  of 2:1 over

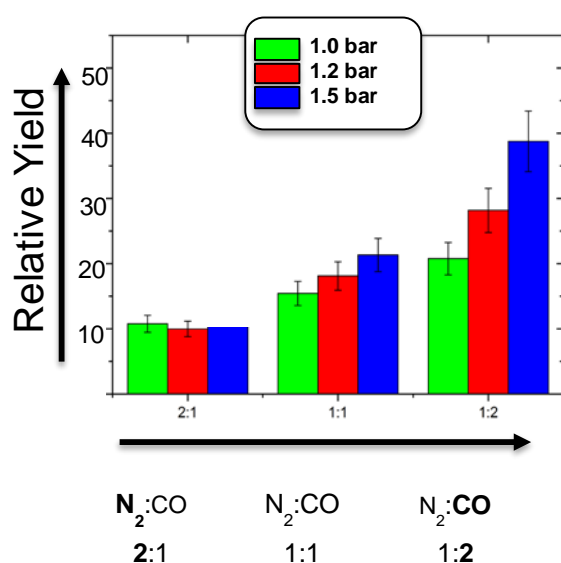


Fig 2. Transport efficiency for non-volatile reaction products like Tc and Mo, collected at the entrance of the reaction chamber.

shorter mean free path between collisions with CO molecules and slower diffusion to the wall, where non-volatile species are lost. Further studies aimed at investigating the influence of further parameters are currently under final evaluation.

### 3.2. ALBEGA development

Advanced prototypes of the two ALBEGA core detectors, whose inner surfaces are covered with a thin Al and SiO<sub>2</sub> layer, respectively, have been tested. They were developed to provide i) improved energy resolution and sensitivity to low energy signals produced by electrons, ii) higher photon detection efficiency, and iii) better mechanical stability against pressure differences. The tests performed with several radioactive sources including <sup>241</sup>Am ( $\alpha$  particles) and <sup>133</sup>Ba (conversion electrons) confirmed performance according to specifications, including the uniformity and desired very thin thickness of the dead layers, as well as the sensitivity to the low energy signals. In a next step, the detectors will be mounted in a sandwich configuration to produce the final version of the ALBEGA core detector.

## 4. Conclusion

After initial promising indications from the JAEA Tokai experiments, the main scientific goal of the TRIGA studies was to distinguish and optimize the transport efficiency for volatile carbonyl complexes of non-volatile radioisotopes between the thermalization- and the reaction chamber

decreasing N<sub>2</sub> content to a limiting ratio of 1:2) at constant total flow rate, and at different pressures, is shown in Figure 2. The efficiency for the transfer from the thermalization chamber into the reaction was found to reach values up to around 45%, which is substantially higher than the transmission of the GARIS separator. The positive influence of a high CO concentration on the yield is likely explicable by an increase in statistical collision probability with CO, which is necessary for the formation of a metal-CO bond and eventually a volatile carbonyl complex. This observation agrees with previous experiments performed by Even et al. [3, 8] behind a physical preseparator. The yield increases with increasing pressure inside the chamber and can be explained by

as well as to increase overall efficiency for the in-situ synthesis of carbonyl complexes. The transport efficiency was investigated at the TRIGA Mainz. Experiments with an  $^{235}\text{U}$ -target and offline measurements, performed with a  $^{248}\text{Cm}$  spontaneous fission source, revealed transport efficiencies up to 40% and thus, the new setup looks to be a promising alternative to the current method of choice.

## 5. Acknowledged

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