Volatile Carbonyl Compounds for New Refractory Beams at ISOLDE

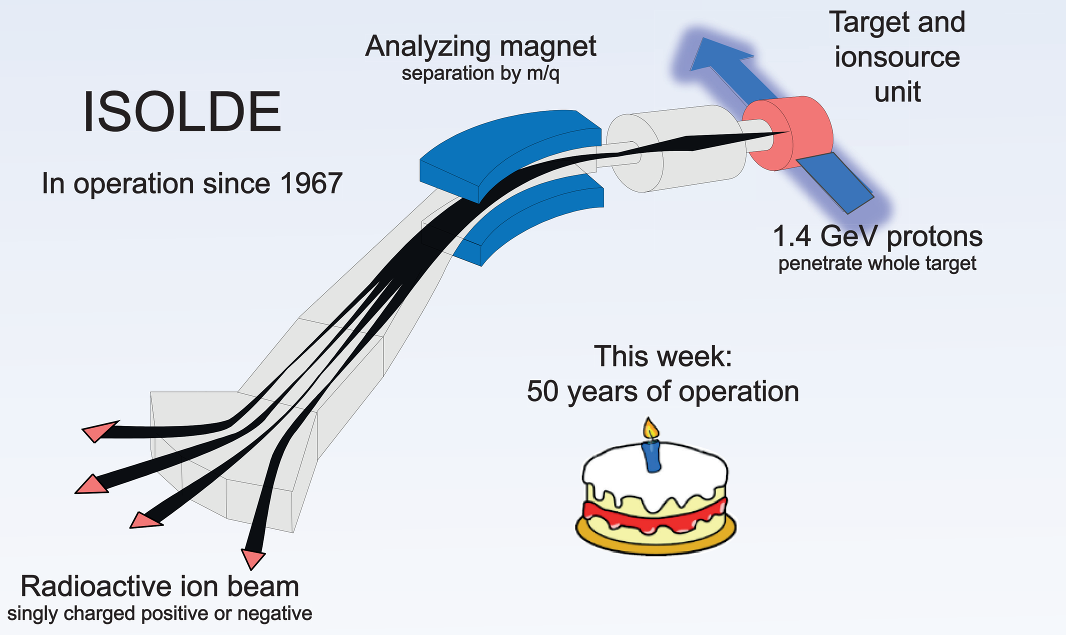
J. Ballof[1,2], C. Seiffert[2,3], Ch. E. Düllmann[1,4,5], J. P. Ramos[2], S. Rothe[2], T. Stora[2], A. Yakushev[4,5]

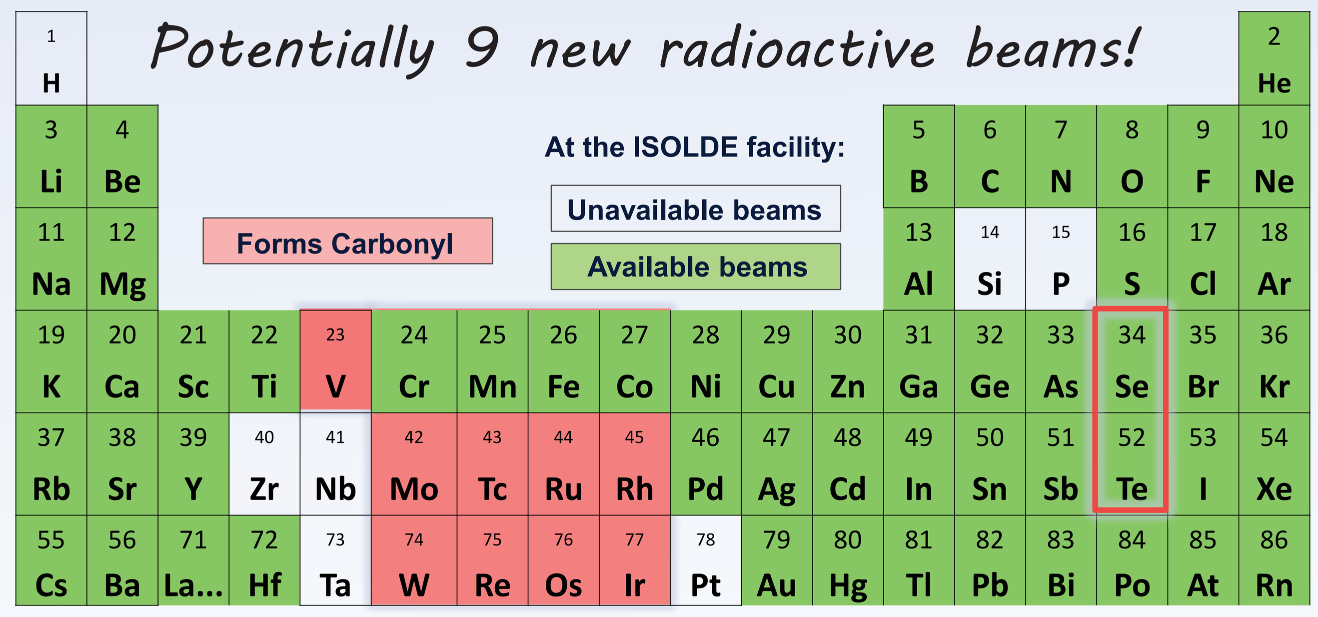
1) Johannes Gutenberg - Universität Mainz, Saarstr. 21, 55122 Mainz, Germany 2) CERN, Engineering Department, ISOLDE, 1211 Geneva 23, Switzerland 3) Technische Universität Darmstadt, Schlossgartenstr. 9, 64289 Darmstadt, Germany 4) GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstr. 1, 64291 Darmstadt, Germany 5) Helmholtz-Institut Mainz, Johannes Gutenberg-Universität, 55099 Mainz

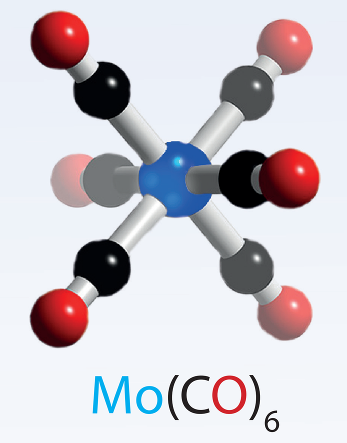
Despite the more than **50 year** old history of ISOL facilities, it is up to date still not possible to extract radioactive beams of many **refractory elements,** which is mostly due to their lack of volatility.

However, following recent developments which came up in the superheavy element research groups [1], the extraction of these elements has now come within reach.

We propose here the extraction of refractory metals as organometallic **carbonyl complexes.**

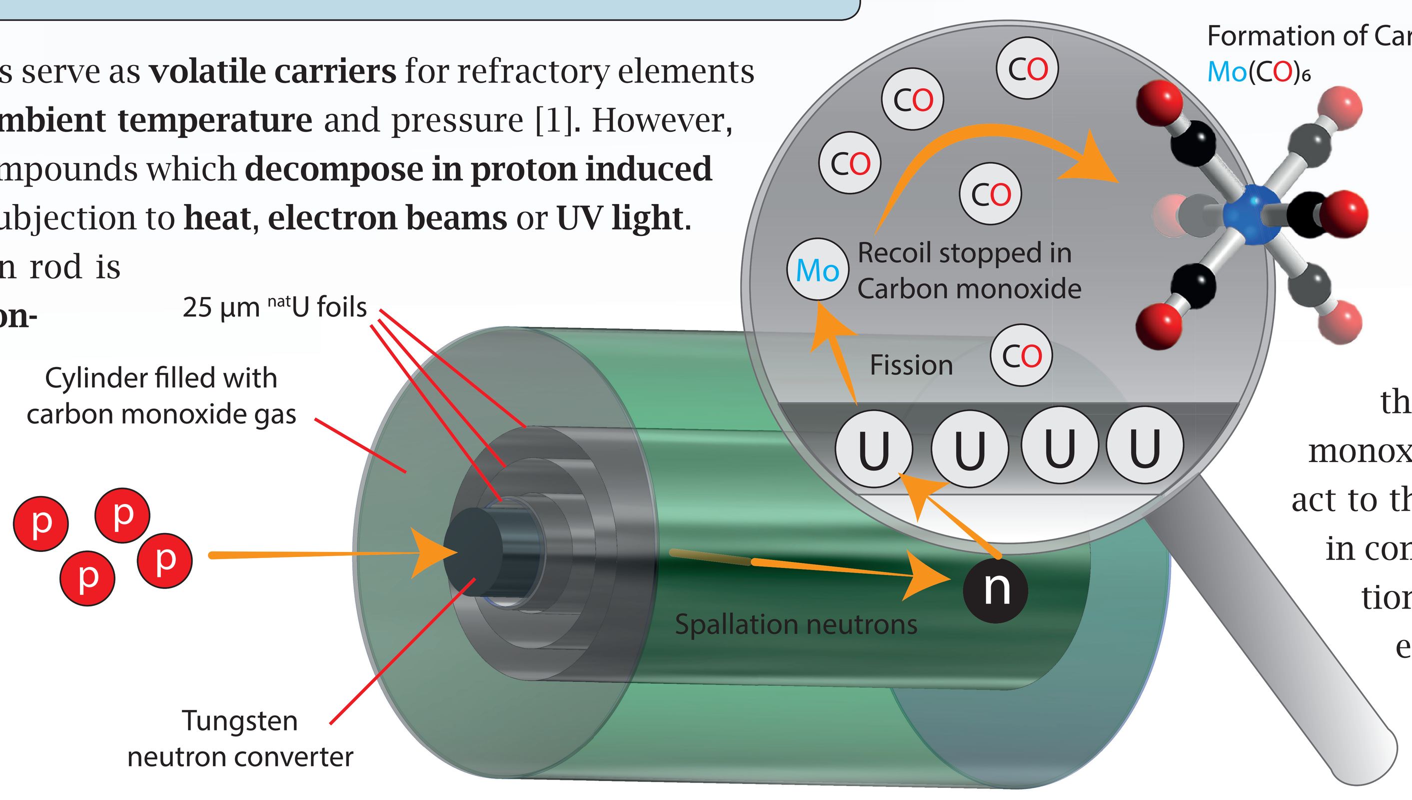






1 Production of radionuclides

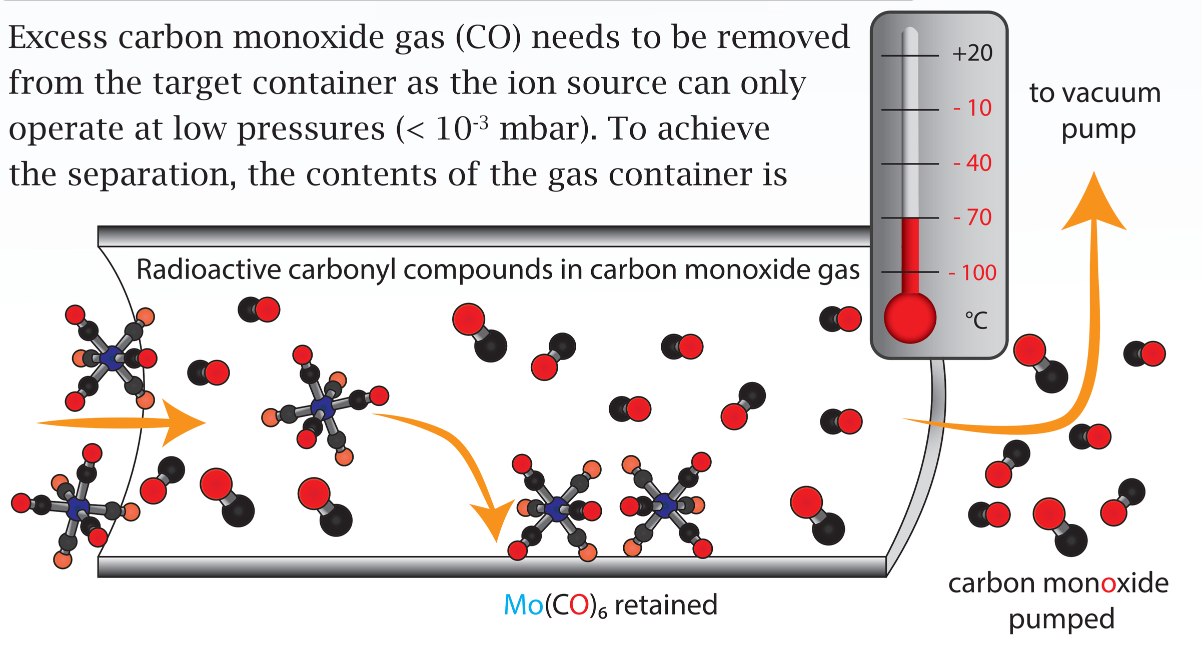
Carbonyl compounds serve as **volatile carriers** for refractory elements and form easily at **ambient temperature** and pressure [1]. However, these are delicate compounds which **decompose in proton induced plasmas** and upon subjection to **heat, electron beams** or **UV light**. Therefore a tungsten rod is used as **neutron converter** [2], which serves as **neutron spallation source** upon irradiation with protons. The **FLUKA** code was used to calculate the in-target production.



In the shown geometry, an *in-target* yield for ^{105}Mo of **10^8 ions / s** is predicted. Instead of diffusion through the solid materials, as in common ISOLDE target units, the **recoil effect** is exploited to extract the fission fragments into the carbon monoxide gas phase, where they readily react to the volatile carbonyl compound. **SRIM** in combination with a Monte-Carlo simulation was used to estimate the extraction efficiency to **11%** for leaving the 25 µm foil, thereof **51%** are stopped in the gas.

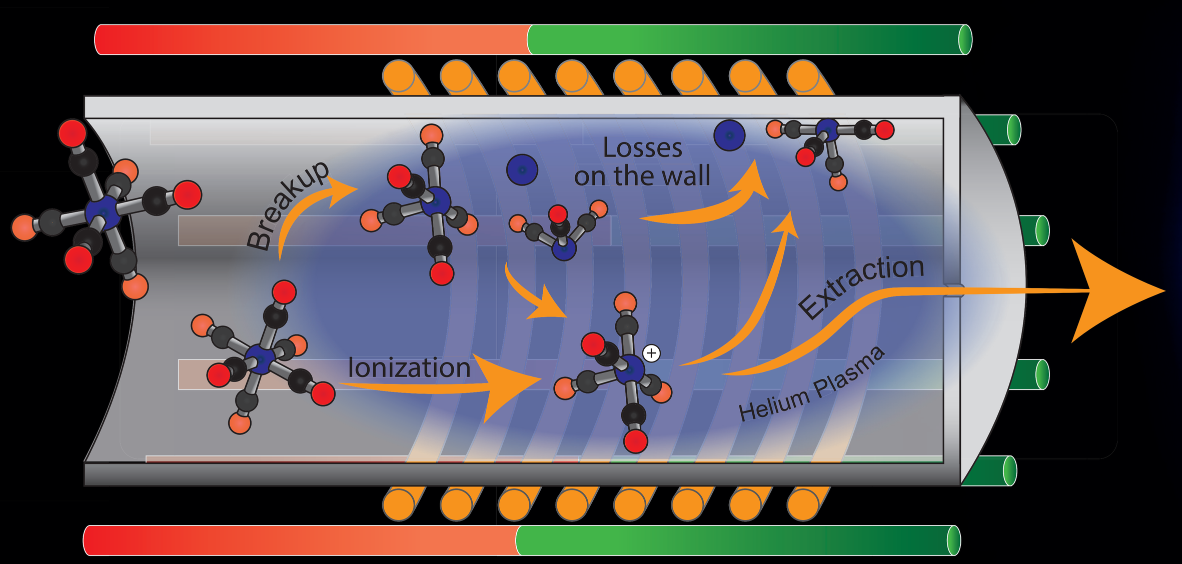
2 Excess-gas removal

Excess carbon monoxide gas (CO) needs to be removed from the target container as the ion source can only operate at low pressures (< 10^{-3} mbar). To achieve the separation, the contents of the gas container is pumped over a **cooling trap**. The gas flow and adsorption behavior has been investigated by Zvara [3] simulations. According to the simulations, temperatures between **-70 °C** and **-110 °C** are necessary to retain the compound.

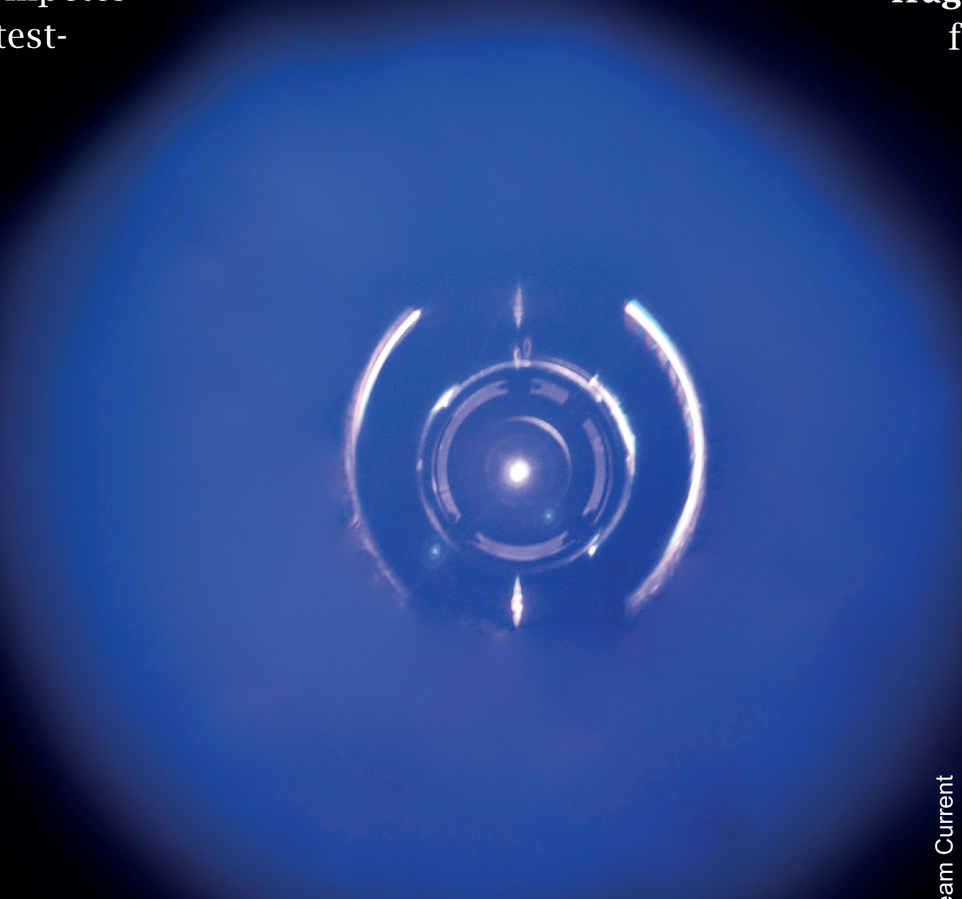


3 Ionization of the molecule

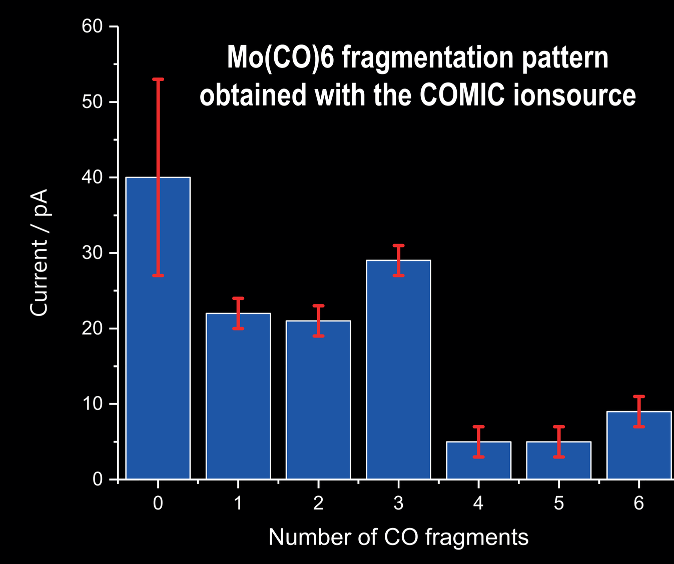
Ionization is one of the key steps in beam extraction. Due to the fragility of the compound, **break up** inside the ion source can occur. The remaining fragments are not volatile anymore, will stick to the walls and are lost. Therefore, the breakup competes with ionization processes. Within this work, several ion sources have been tested for compatibility with carbonyl beams. These are a customized FEBIAD-type **VADIS** [4] ion source, the microwave driven quartz **COMIC** [5] source and the radio frequency heated **HELICON** [6] source.



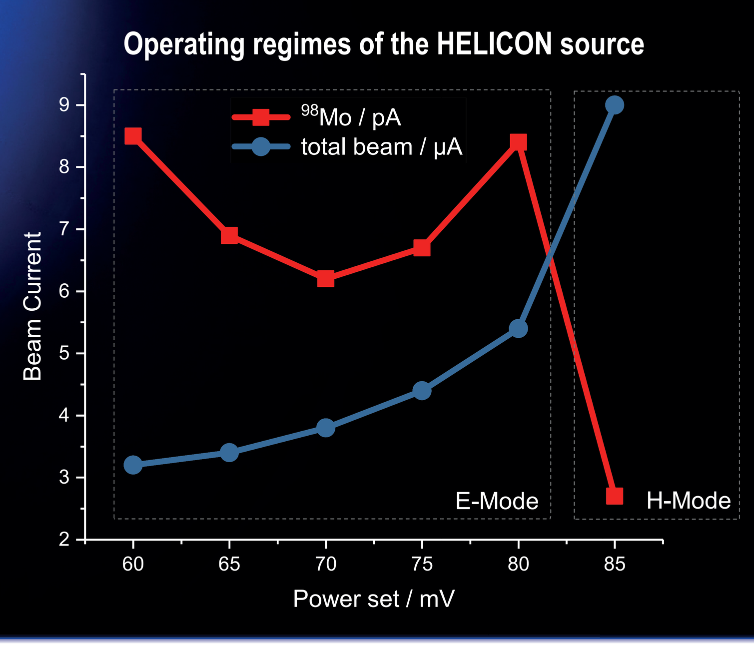
The *VADIS* ion source is operated at **high temperatures** (typically 2000 °C) but allows to use relatively **energetic electrons** for the ionization, which is advantageous to promote ionization rather than breakup. The *radio frequency driven sources* are operated **cold**, however, the **electron energies are low** in comparison to the electron impact source. With all investigated sources, it was possible to ionize the compound, but the so far achieved efficiencies need to be improved further for radioactive ion beam applications.



The **fragmentation pattern** could be measured with all ion sources, and a typical pattern for the COMIC source is shown below. As it can be seen from the graph, the **tricarbonyl fragment is predominant**, and the formation of tetra- and penta carbonyls is not favored.

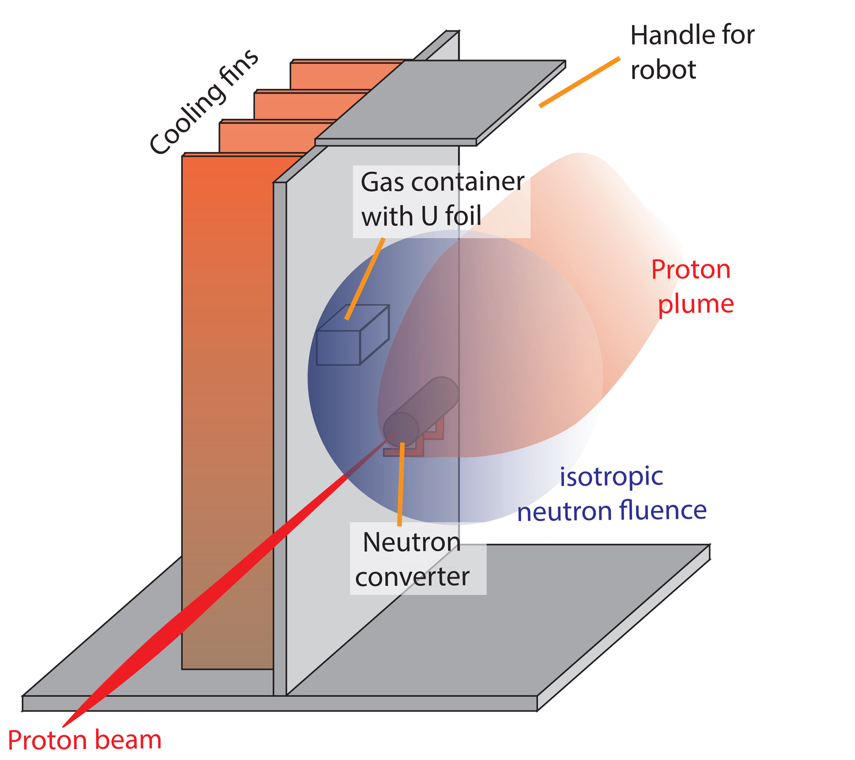


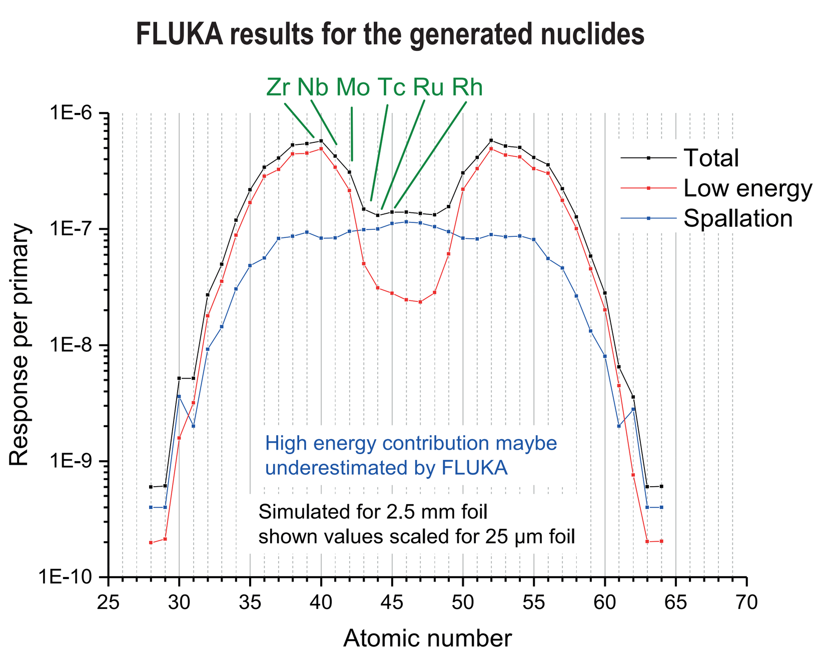
It turned out, that the quartz **COMIC** source exposes an unfavorable geometry for the extraction of fragile molecules, due to the positioning of the microwave antennas in the plasma volume. Therefore the **HELICON** source was tested. The source can be operated in an **capacitive** and an **inductive** mode. While the inductive mode offers relatively high efficiencies for noble gases (4% on Argon), the capacitive mode was found to be better suited for carbonyl compounds. As next step we plan to investigate the **MINIMONO ECR** source.



M MEDICIS irradiations

To verify the results of the FLUKA simulations, and to investigate carbonyl decomposition by the proton beam, we have prepared an irradiation experiment to be conducted at the new MEDICIS facility. A gas container filled with **carbon monoxide gas** and a **uranium foil**, will be placed on a irradiation unit equipped with passive **cooling fins** and a **neutron converter**.



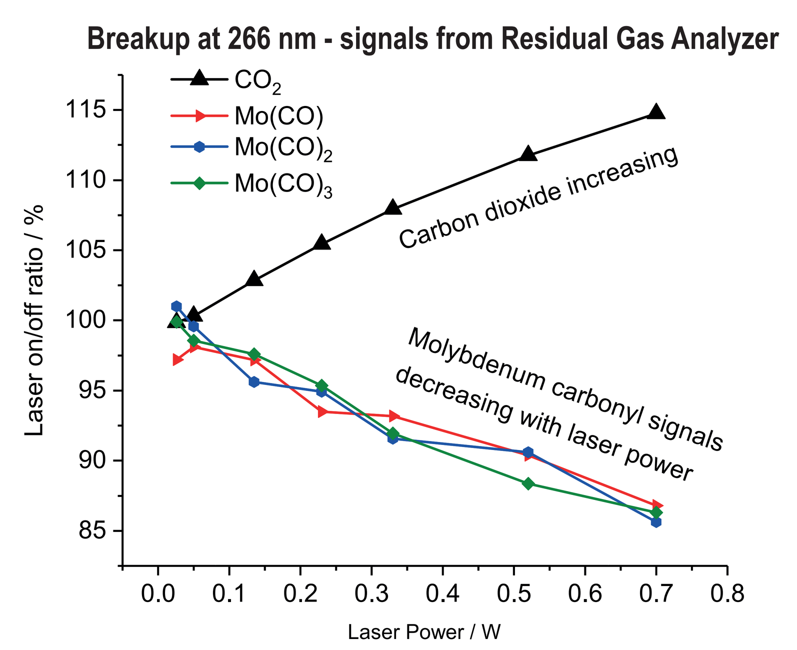


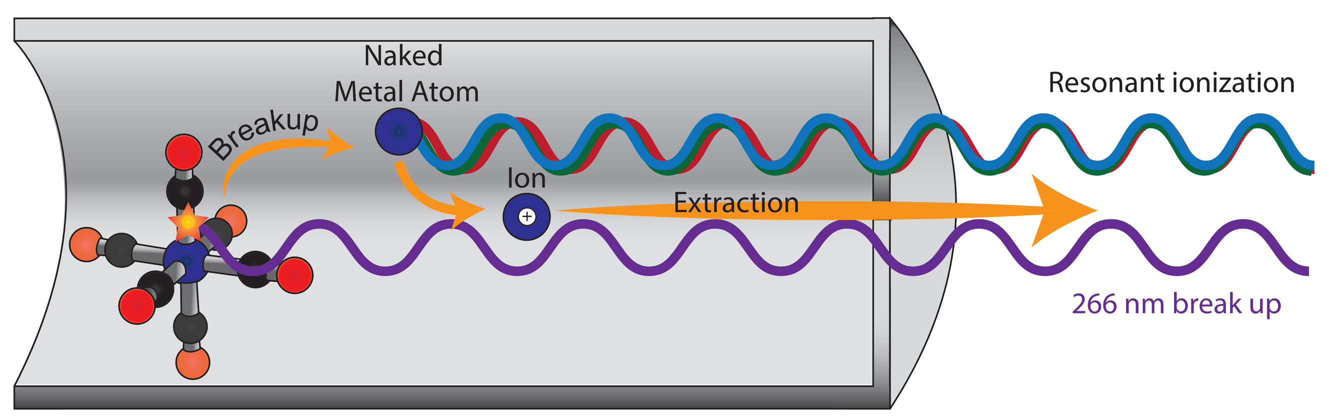
After irradiation, the unit is brought to the **hot cell**, where the gas volume is pumped over a **charcoal trap**. The trap will then contain **only volatile** compounds, like Mo(CO)$_6$, which are then subjected to **gamma spectroscopy**. For first tests we have chosen to place the gas container at a position which is receiving the lowest possible proton bombardment.

L Laser breakup and ionization

In addition to the sources discussed above, the possibility of ionizing the refractory metal by means of resonant laser ionization was also addressed. Prior to resonant laser ionization, the **compound needs to be broken up completely**. Due to the low first bond dissociation energy of only 1.7 eV [7], carbonyl complexes are well suited for laser induced breakup.

We investigated the laser breakup by constantly feeding molybdenum carbonyl into a vacuum chamber, which was equipped with a residual gas analyzer. By comparison of the „laser on" and „laser off" signals it was found out, that we **decompose 15%** of the injected metal carbonyls using a wavelength of 266 nm. Just recently, a **resonant ionization scheme for molybdenum has been successfully developed** by K. Chrysalidis, therefore the combination of laser breakup and ionization is ready to be tested.





References

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