Apparatus for muonium kinetics experiments on fluids at high pressure and temperature

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Generation IV International Forum (GENIV)

Canada, as a participant of GENIV is exploring the feasibility of a supercritical water nuclear reactor (SCWR) in which the cooling cycle operates at pressures around 250 bar and at temperatures up to 625°C.

There is very little information on the reaction kinetics of the aqueous species present in such an environment. This information is needed to predict corrosion rates and other water chemistry.

In this respect, muonium – as a light radioactive isotope of the hydrogen atom – turns out to be a very convenient probe for *in situ* kinetic measurements.

Muonium to study reactions in "hostile environment" (high pressure/temperature water)

- Muonium [μ+, e-] light isotope of hydrogen
- Muons injected into most materials. Optical windows are unnecessary.
- High sensitivity due to ~100% spin polarized muons, ie, no need for radio-frequency or other radiation to induce resonance.
- Spectroscopic information is conveyed by the direction of emission of the high energy decay positrons which can penetrate through the heavy reactor walls.
- The detectors are simple devices (plastic scintillators) outside the controlled sample environment.
- The short lifetime of the muon, μSR is inherently suited to the study of transients.

Chemical Kinetics in Supercritical Water

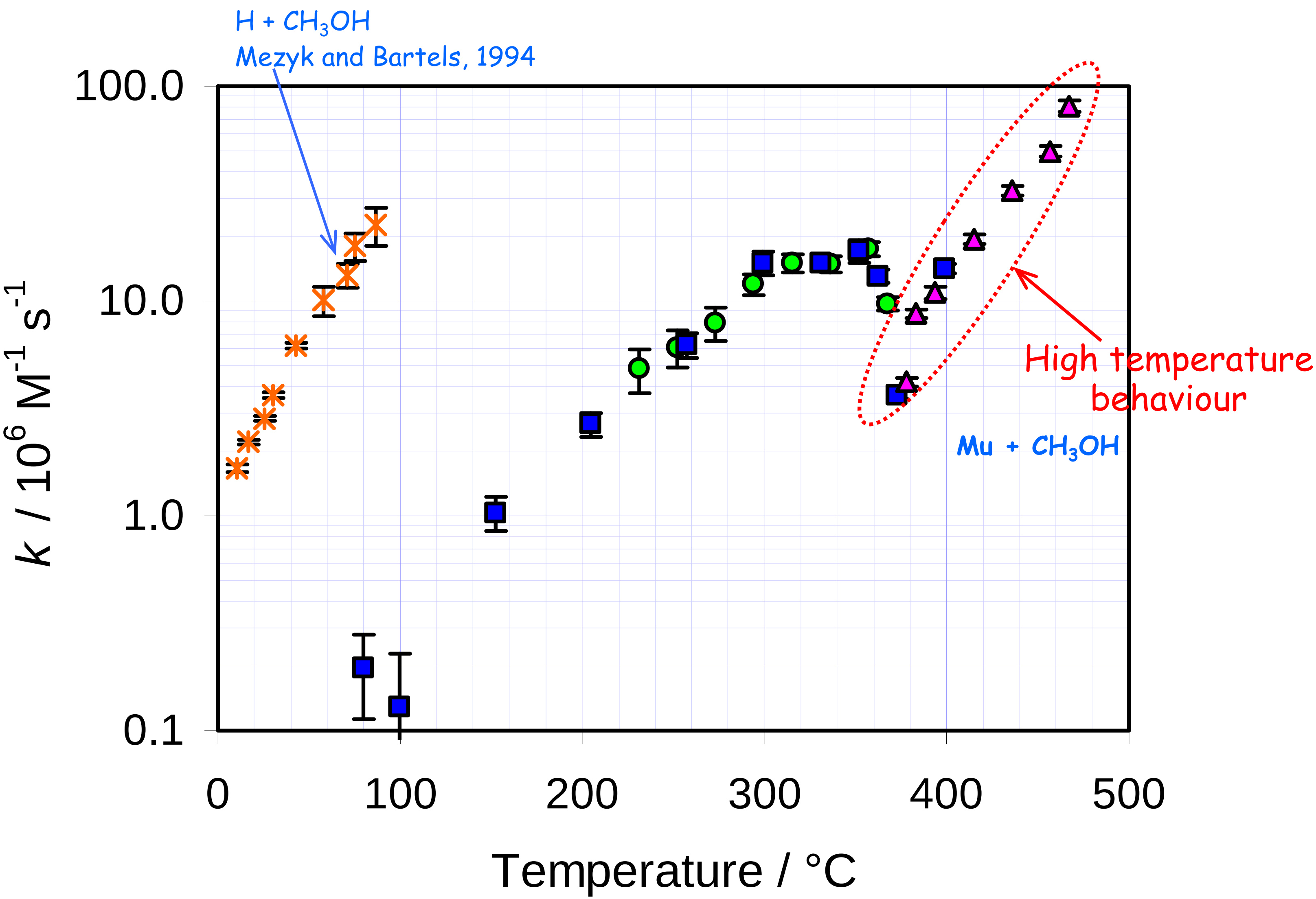
Drastic changes in the physical properties of water close to and above the critical point

(T_c = 374°C, P_c = 221 bar).

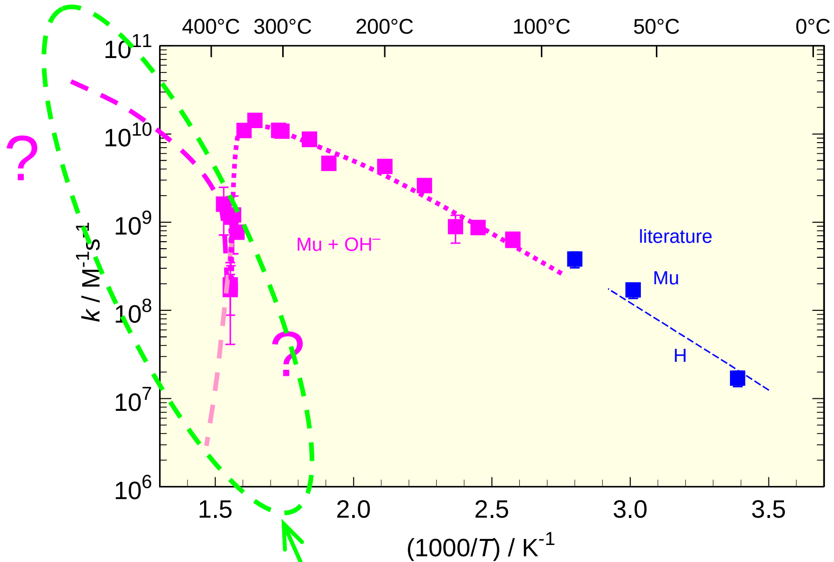
This leads to unusual kinetic behaviour (non-Arrhenius):

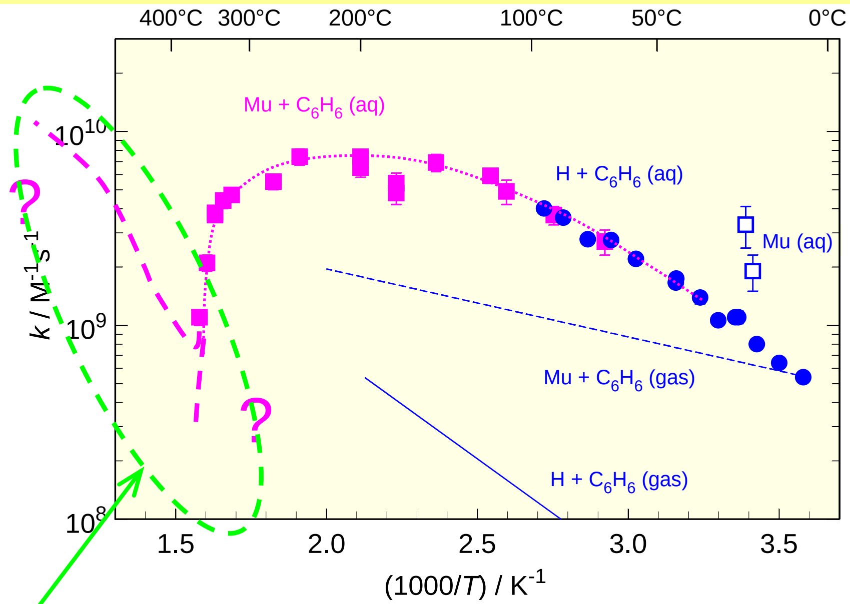
- Rate constants go through a maximum around critical conditions,
- Rate constants drop above critical conditions down to a minimum,
- Indications of recovery of the rate constants above this minimum

Example 1: H abstraction by Mu (or H)



Other examples





Mu (H) + OH^- → MuOH + e^-_{aq}

Mu (H) addition to benzene

Unknown behaviour

Practical Requirements for the Reactor Cell

- Strength of the container: in the present case, must be able to withstand without deformation the stress generated by internal pressure of at least 250 bar at temperature up to 650°C
- Entrance window: must be as thin as possible to facilitate muon penetration with a minimum of scattering.
- Cell material: ideally should be chemically inert.

Cell Design

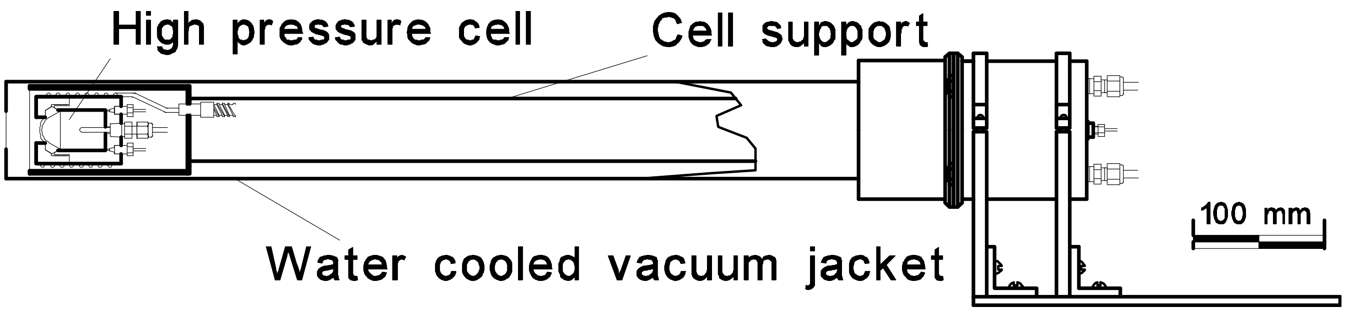
Version 1: for max pressure 450 bar, T < 450 °C

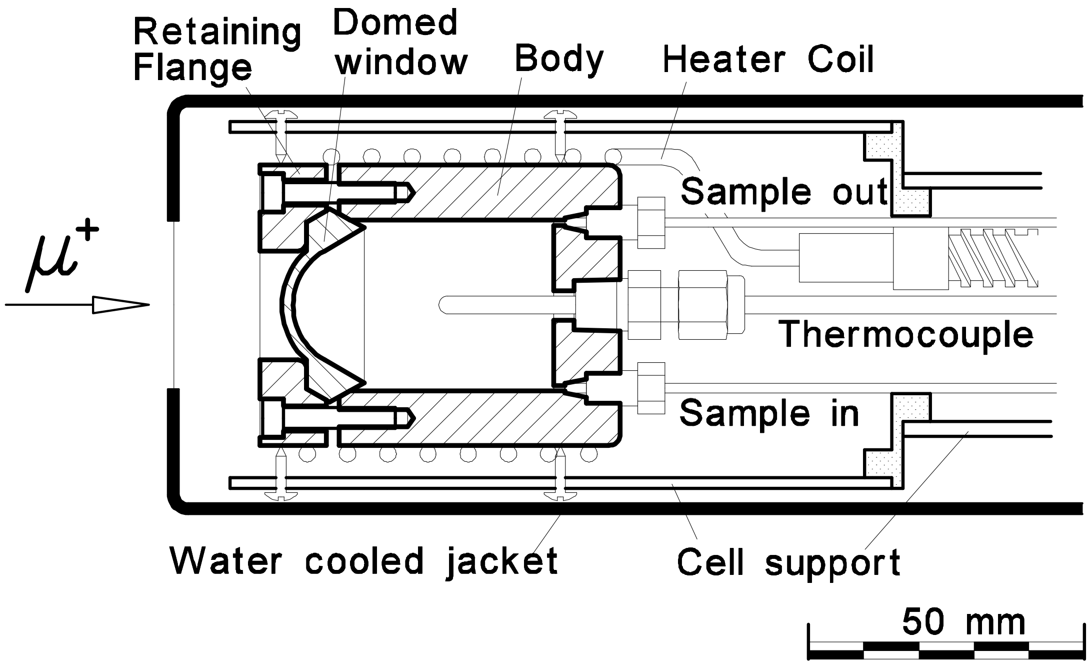
- Material: Ti 6Al4V
- Good for sample with density > 0.5 g cm^{-3}

Version 2: for p ~300 bar, T up to ~650 °C

- Alloy 625 can stand these temperature, but a bigger cell is need to accept low density sample (< 0.1 g cm^{-3})

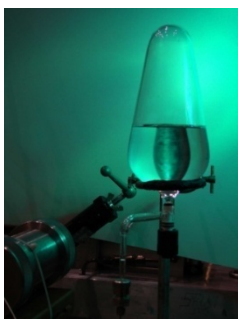
High pressure and temperature cell, version 1

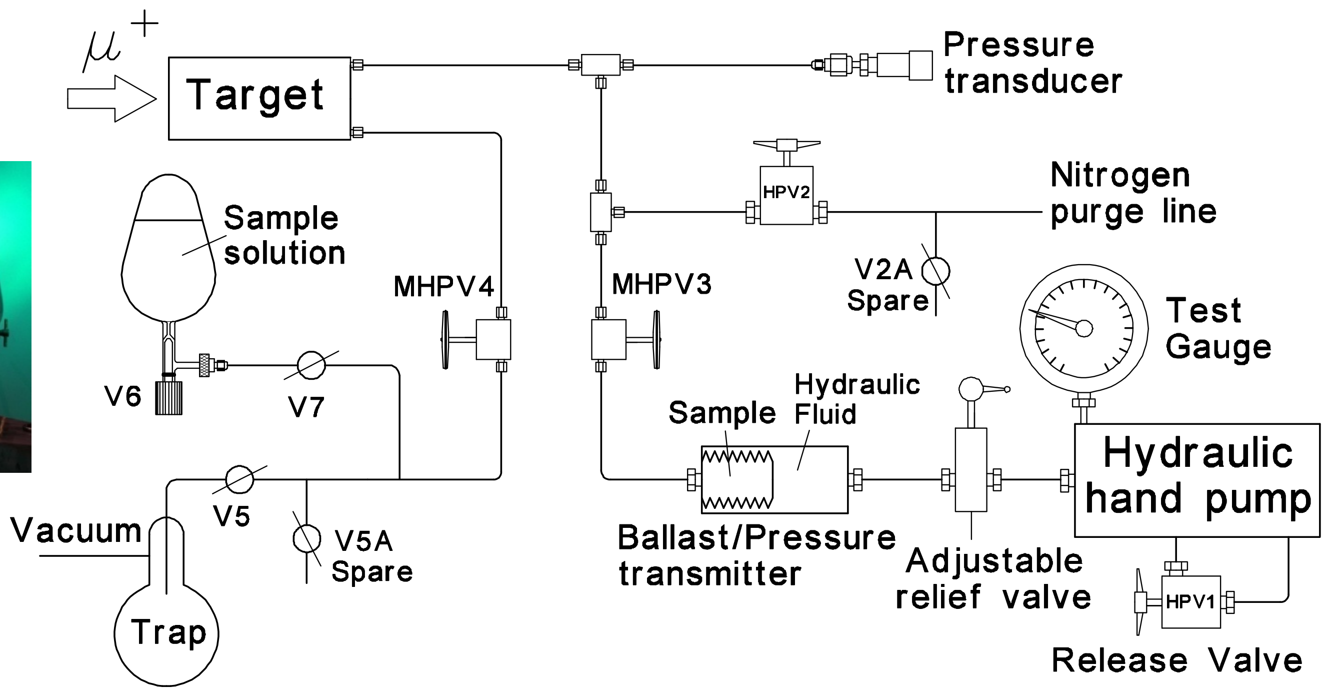




Block diagram of high pressure set-up

or

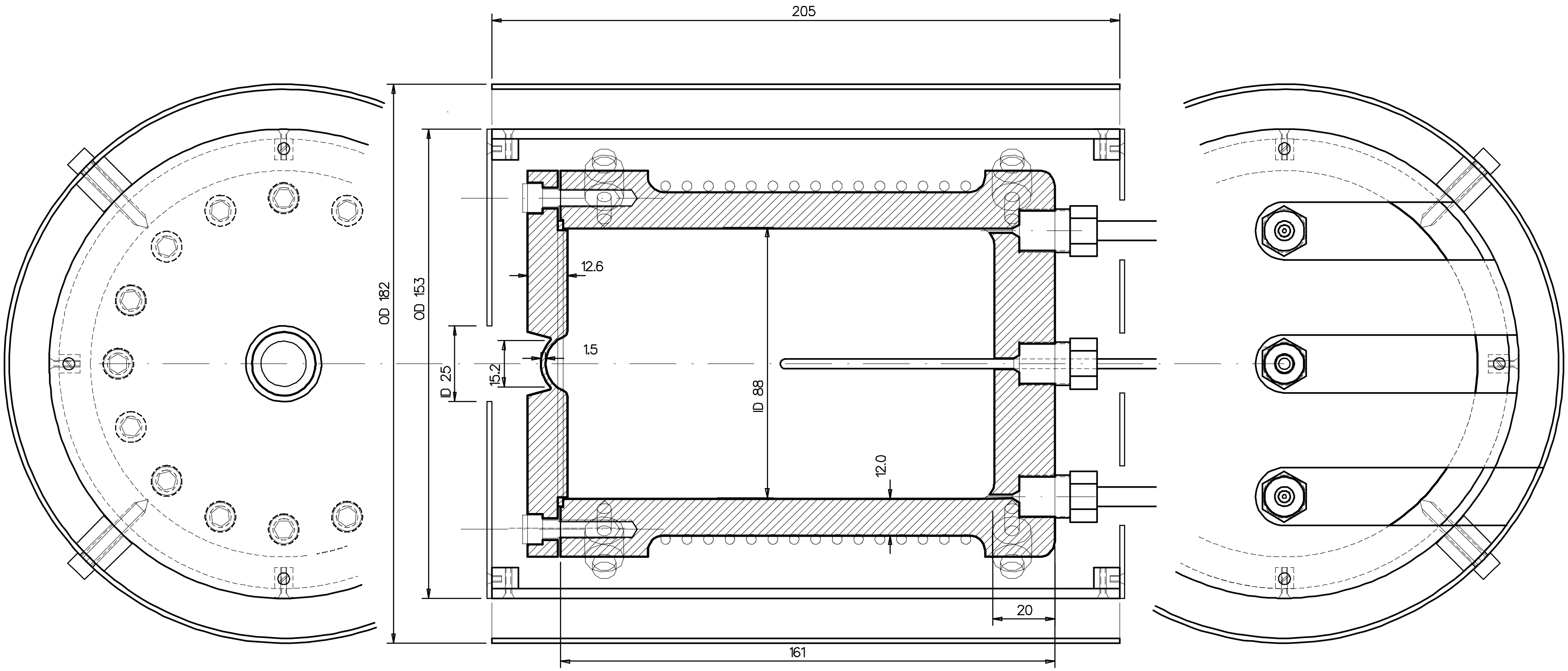




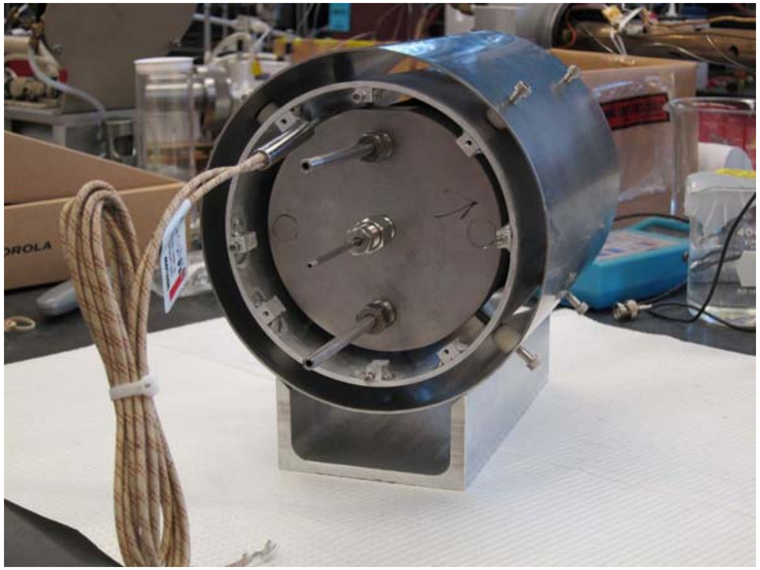
Version 1 Apparatus for Hydrothermal Studies: Conclusions

- In operation for several years
- Very robust
- Slow turn around
- Difficult to clean
- Limited to samples with d > 0.5 g cm^{-3}

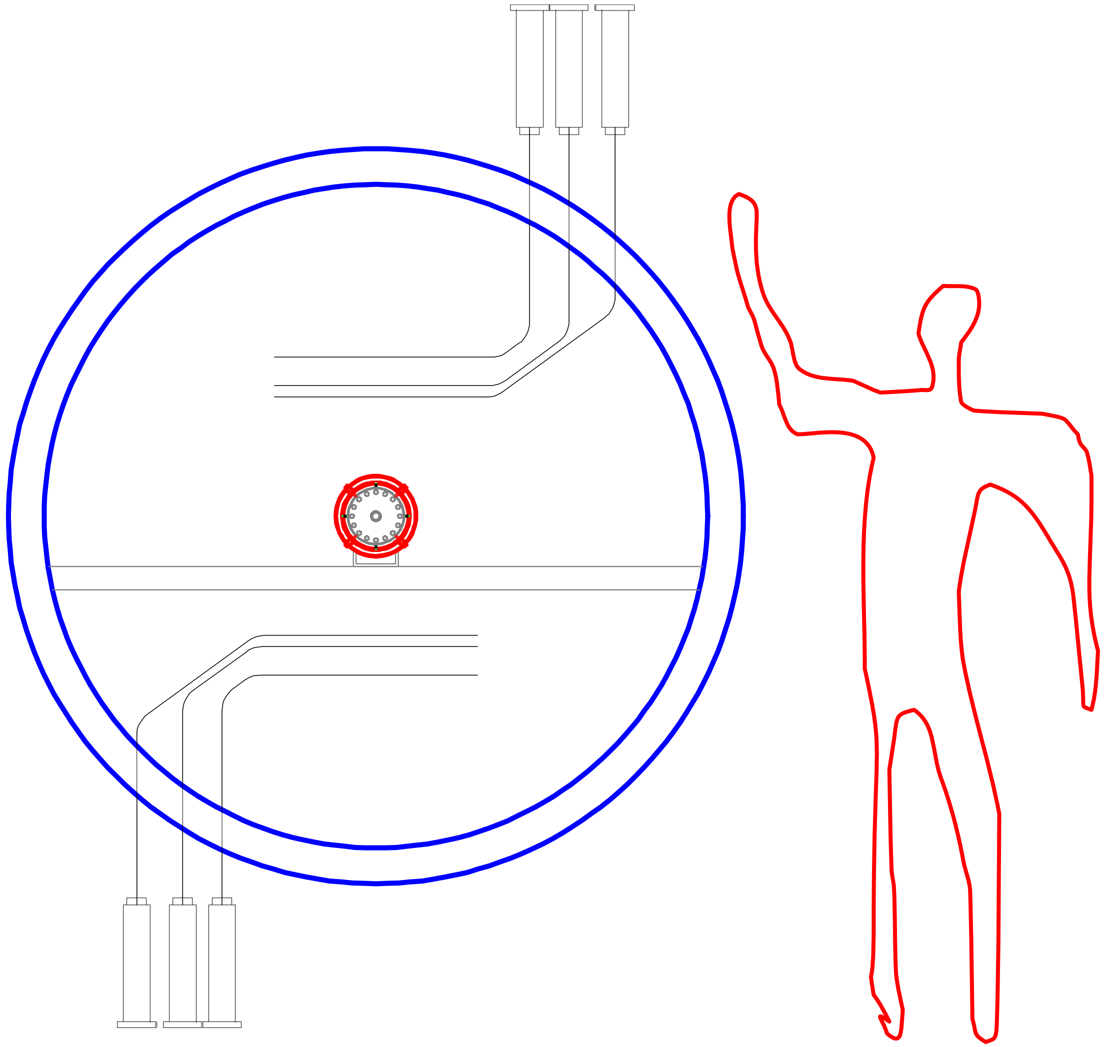
Version 2: p ~ 250 bar, T up to ~650 °C)



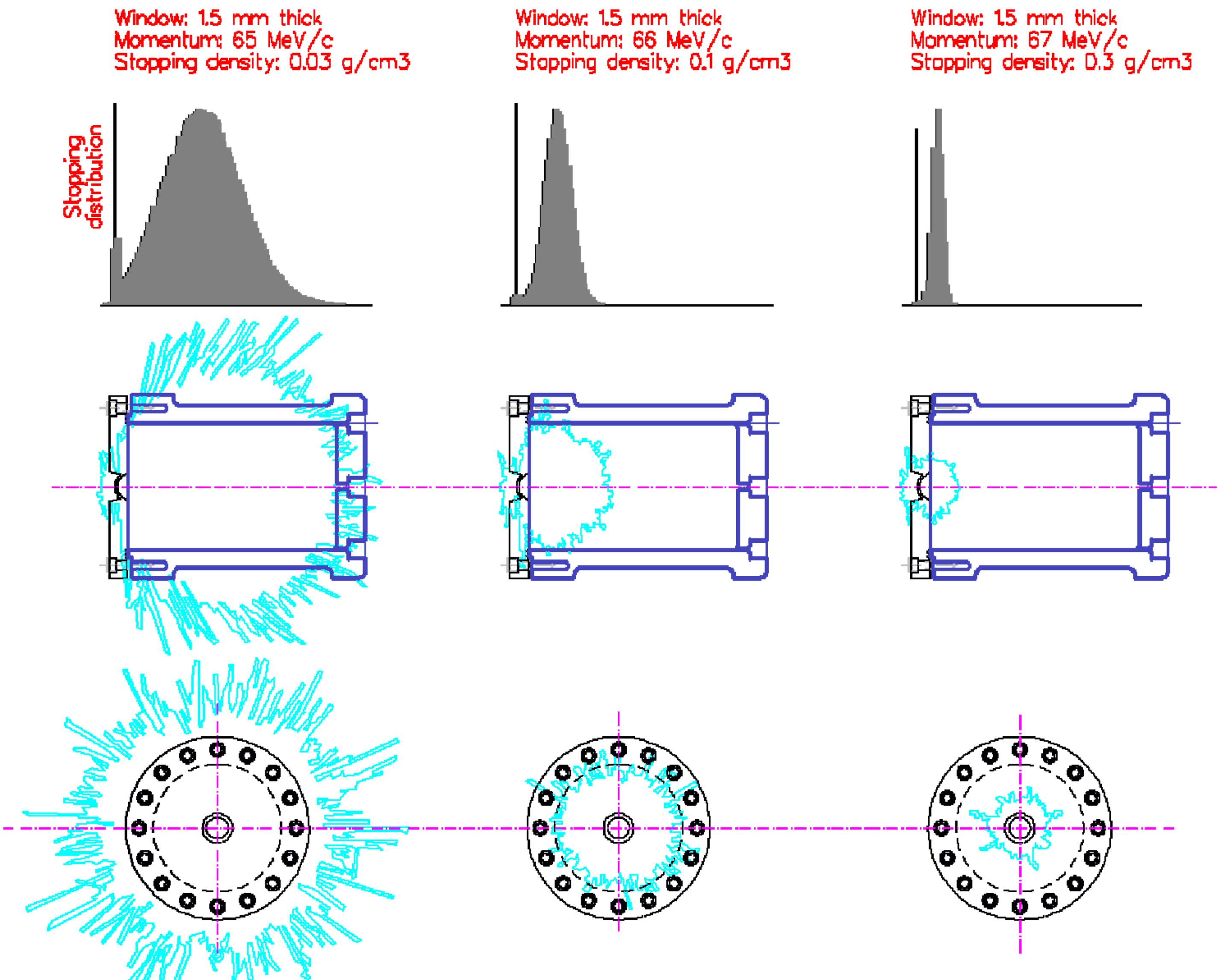
Constructed cell, Version 2



Cell in position in "Gas Cart"



Stopping distribution (SRIM calculations)



Version 2 Apparatus for Muonium Kinetics: Conclusions

- Needs to be fully tested
- Heavy
- Temperature Homogeneity ?
- Phase separation ?
- Good for samples with d < 0.1 g cm^{-3}
- Large Helmoltz magnet ("GasCart")

Acknowledgements

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