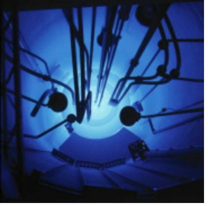


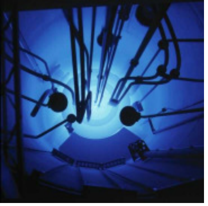
Metal-Carbonyl-Complexes: New perspectives in SHE chemistry and nuclear spectroscopy

Julia Even

Helmholtz-Institut Mainz, Mainz, Germany;
Johannes Gutenberg-Universität Mainz, Mainz, Germany

For a CO collaboration:

HIM – Uni Mainz – GSI – PSI – Uni Bern – LBNL – UC Berkeley - JAEA



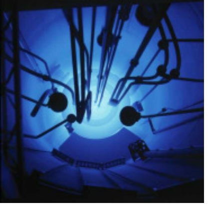
The CO-collaboration

Johannes Gutenberg-Universität Mainz, Mainz, Germany
Helmholtz-Institut Mainz, Mainz, Germany
GSI Helmholtzzentrum für Schwerionenfoschung, Darmstadt, Germany
J. Even, A. Yakushev, Ch.E. Düllmann, J. Dvorak, W. Hartmann, D.Hild, E. Jäger, J. Khuyagbaatar, B. Kindler, J.V. Kratz, J Krier, B. Lommel, L. Niewisch, I. Pysmentska, B. Schausten, N . Wiehl

PSI Paul Scherer Institut, Villigen, Switzerland
University Bern, Switzerland
R. Eichler, A. Türler, D. Wittwer

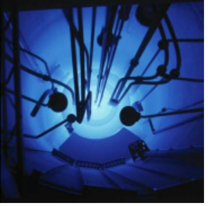
LBNL Lawrence Berkeley National Laboratory, Berkeley, USA
UC Berkeley, Berkeley, USA
O. Gothe, H. Nitsche

JAEA Japanese Atomic Energy Agency, Tokai, Japan
M. Schädel



Outline

- Motivation
- Metal carbonyl complexes
- Experiments @ TRIGA Mainz research reactor
- Experiments @ TASCA recoil separator at GSI
- Outlook and perspectives
- Summary



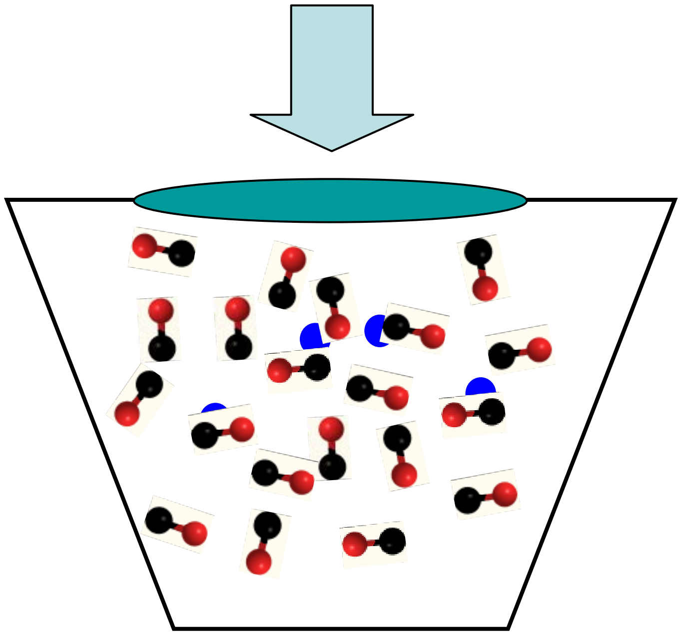
Gas-phase chemistry of the SHE

- Very limited number of compounds
  - Rf: $RfCl_4$, $RfBr_4$, $RfOCl_2$
  - Db: ($DbCl_5$), $DbBr_5$, $DbOCl_3$
  - Sg: SgO_2Cl_2, $SgO_2(OH)_2$
  - Bh: BhO_3Cl
  - Hs: HsO_4, $Na_2[HsO_4(OH)_2]$

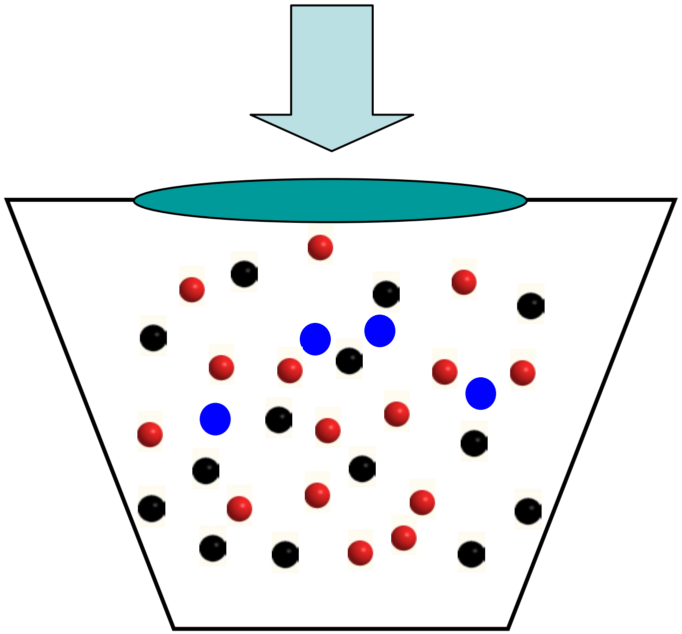
  - Cn and E114 – in their elemental states

New compound classes, in which relativistic effects might be better visible, are of interest!

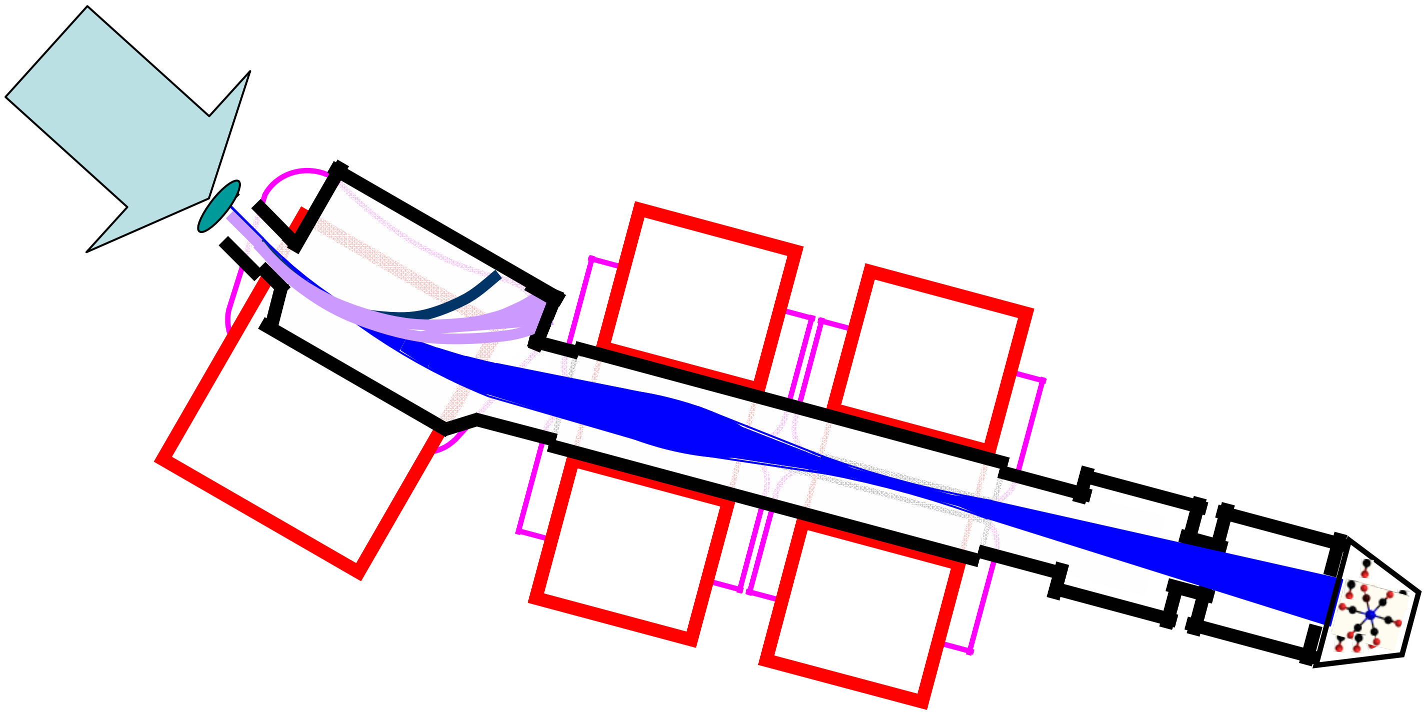
Limits of gas phase chemistry



Limits of gas phase chemistry



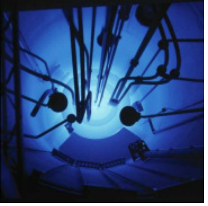
TransActinide Separator and Chemistry apparatus



TASCA @ GSI

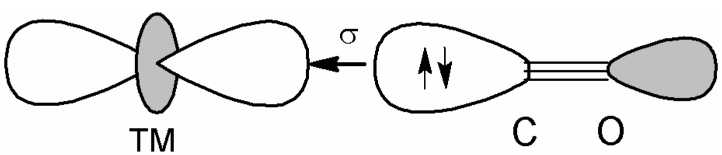
TransActinide Separator and Chemistry Apparatus

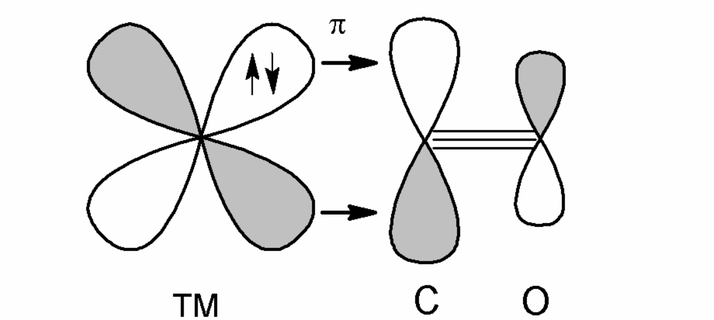
J. Even et al., NIMA 638 (2011) 157
A. Semchenkov et al., NIMB 266 (2008) 4153
Ch.E. Düllmann et al., NIMA 551 (2005) 528
M. Schädel, Eur. Phys. J. D 45 (2007) 67



Binary Metal Carbonyl Complexes

5	6	7	8	9	10
$V(CO)_6$	$Cr(CO)_6$	$Mn_2(CO)_{10}$	$Fe(CO)_5$	$Co_2(CO)_8$	$Ni(CO)_4$
	$Mo(CO)_6$	$Tc_2(CO)_{10}$	$Ru(CO)_5$	$Rh_2(CO)_8$	
	$W(CO)_6$	$Re_2(CO)_{10}$	$Os(CO)_5$	$Ir_4(CO)_{12}$	





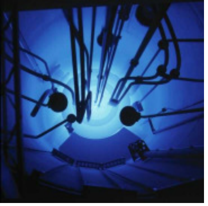
Highly symmetric complexes
with zero valent central metal atoms

J. Am. Chem. Soc. **1999**, *121*, 10830−10831

Prediction of the Bond Lengths, Vibrational Frequencies, and Bond Dissociation Energy of Octahedral Seaborgium Hexacarbonyl, $Sg(CO)_6$

Clinton S. Nash* Bruce E. Bursten*

Relativistic effects might be visible



Classical syntheses of carbonyl complexes

- General: M + CO @ high pressure (~300 bar)
- Hot Chemistry: "Baumgärtner´s method"
  $^{235}U(n,f)^{x}Mo + Cr(CO)_6 \rightarrow {}^{x}Mo(CO)_6$

Not suitable for SHE-chemistry!!!!

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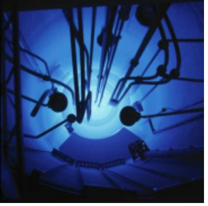

Studied d-elements

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57+* La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89+" Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 ---	114 ---	115 ---	116 ---	117 ---	118 ---

*	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
"	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

^{235}U fission ^{249}Cf fission Nuclear fusion at TASCA

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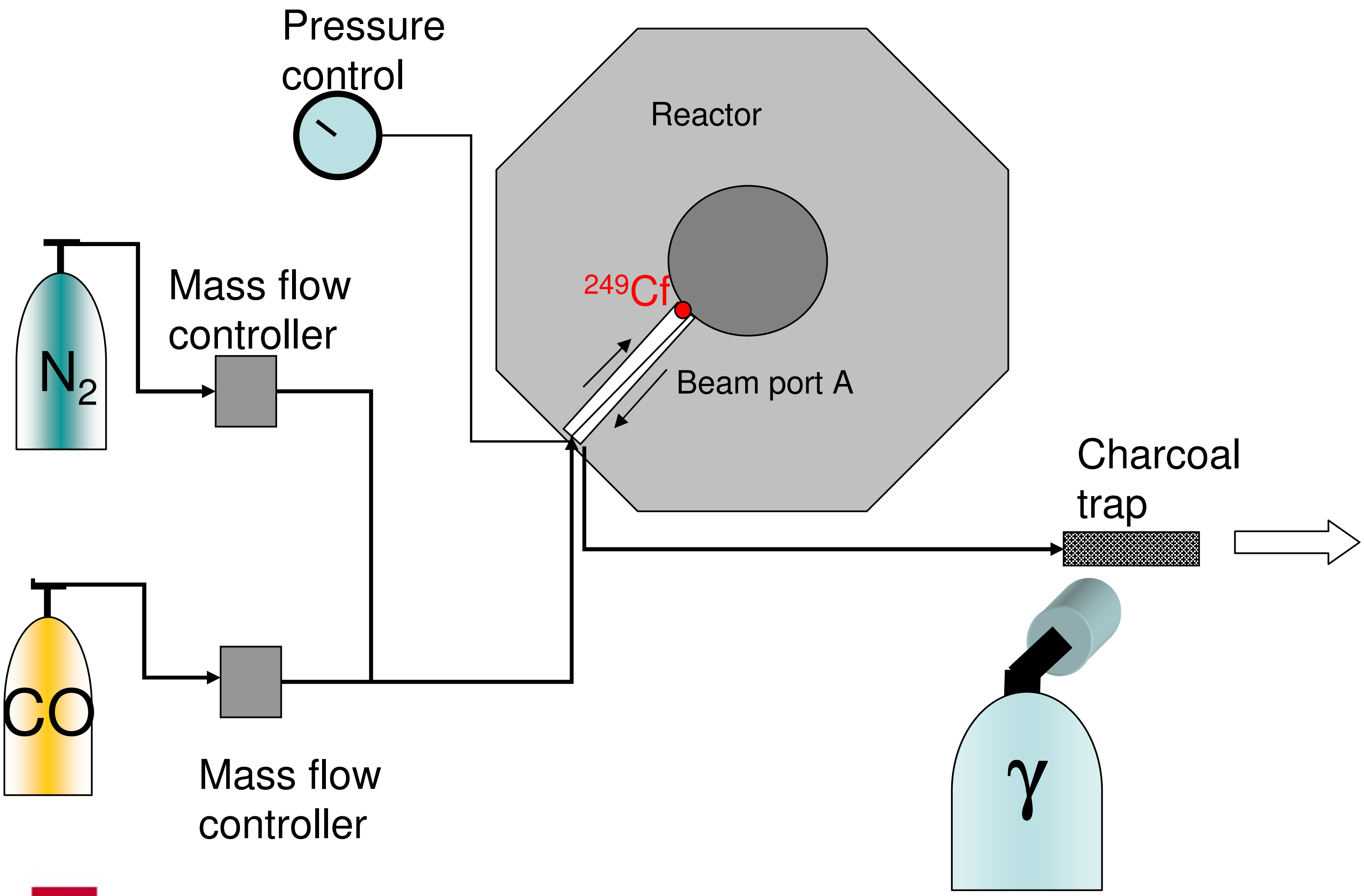
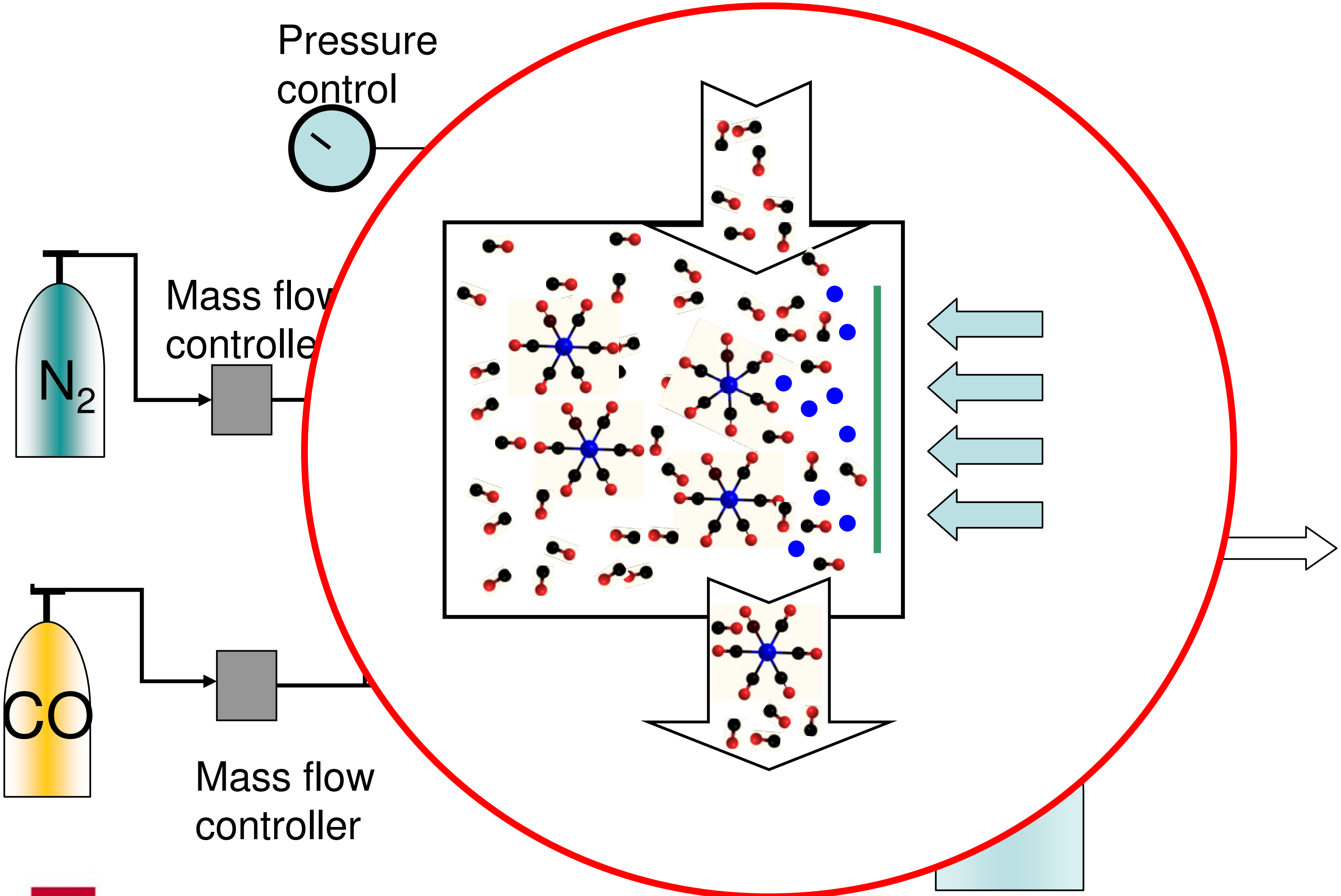
Experiment @ the TRIGA Mainz reactor



Experiment @ the TRIGA Mainz reactor



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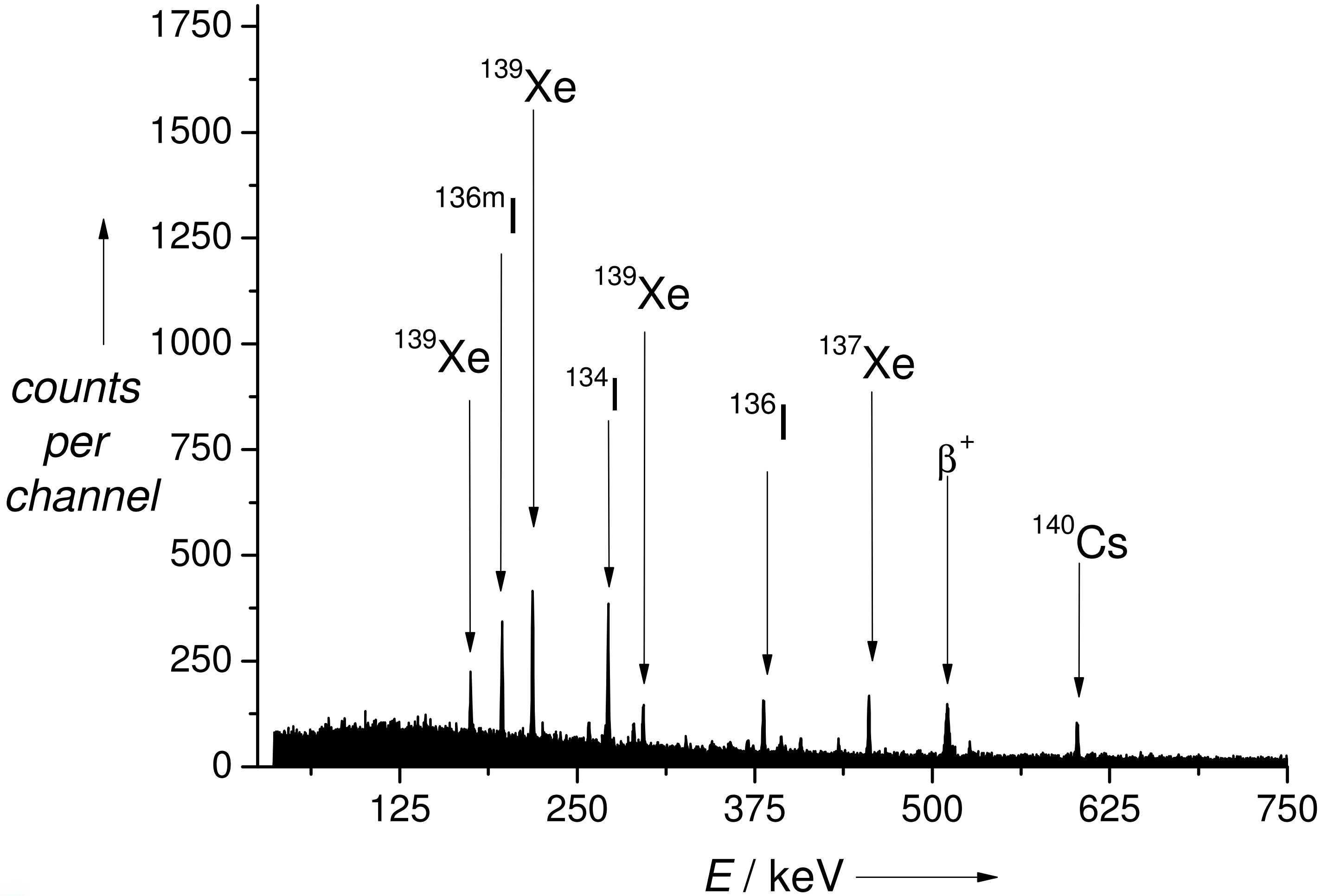
Transport with pure N_2



I
Xe

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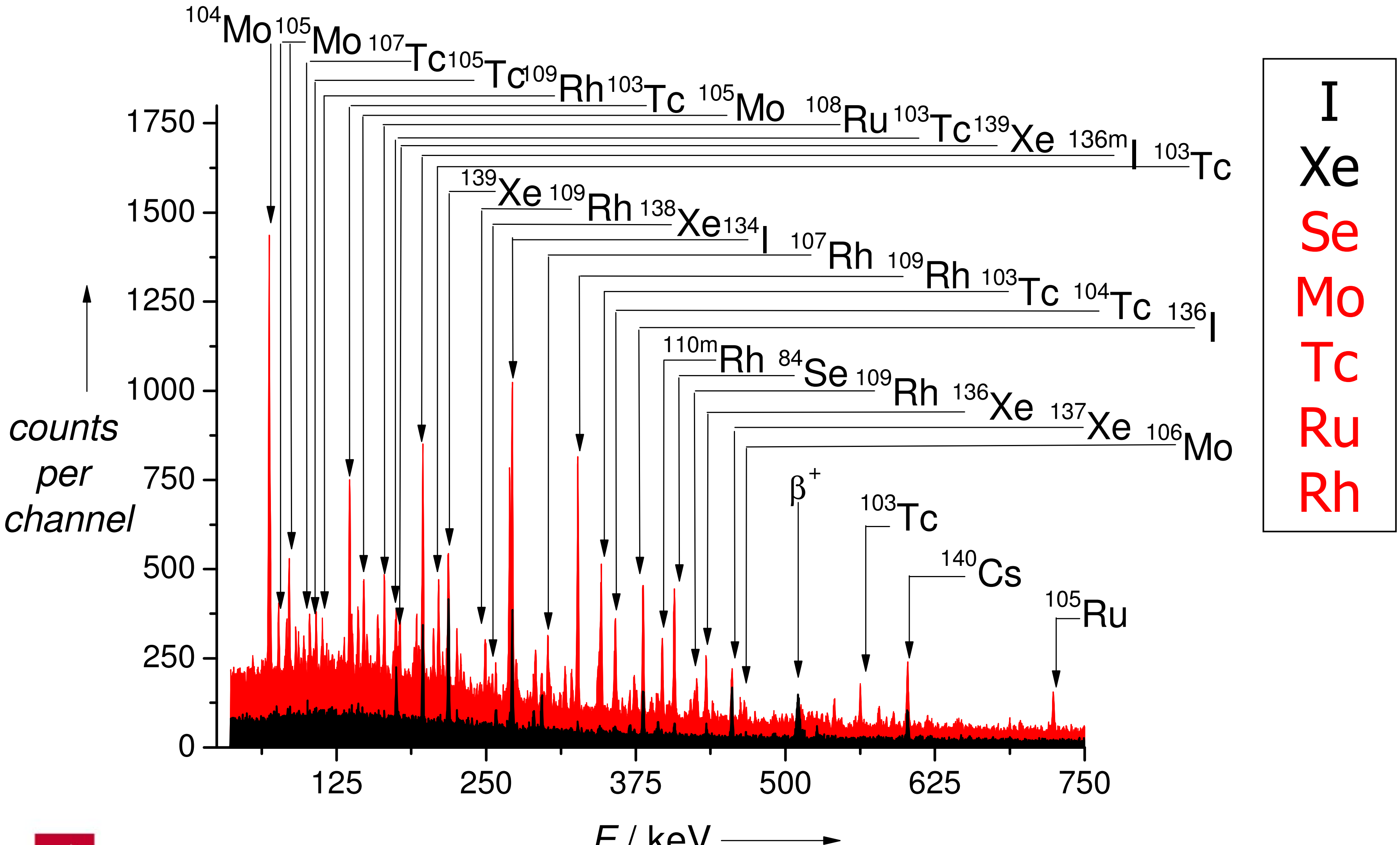
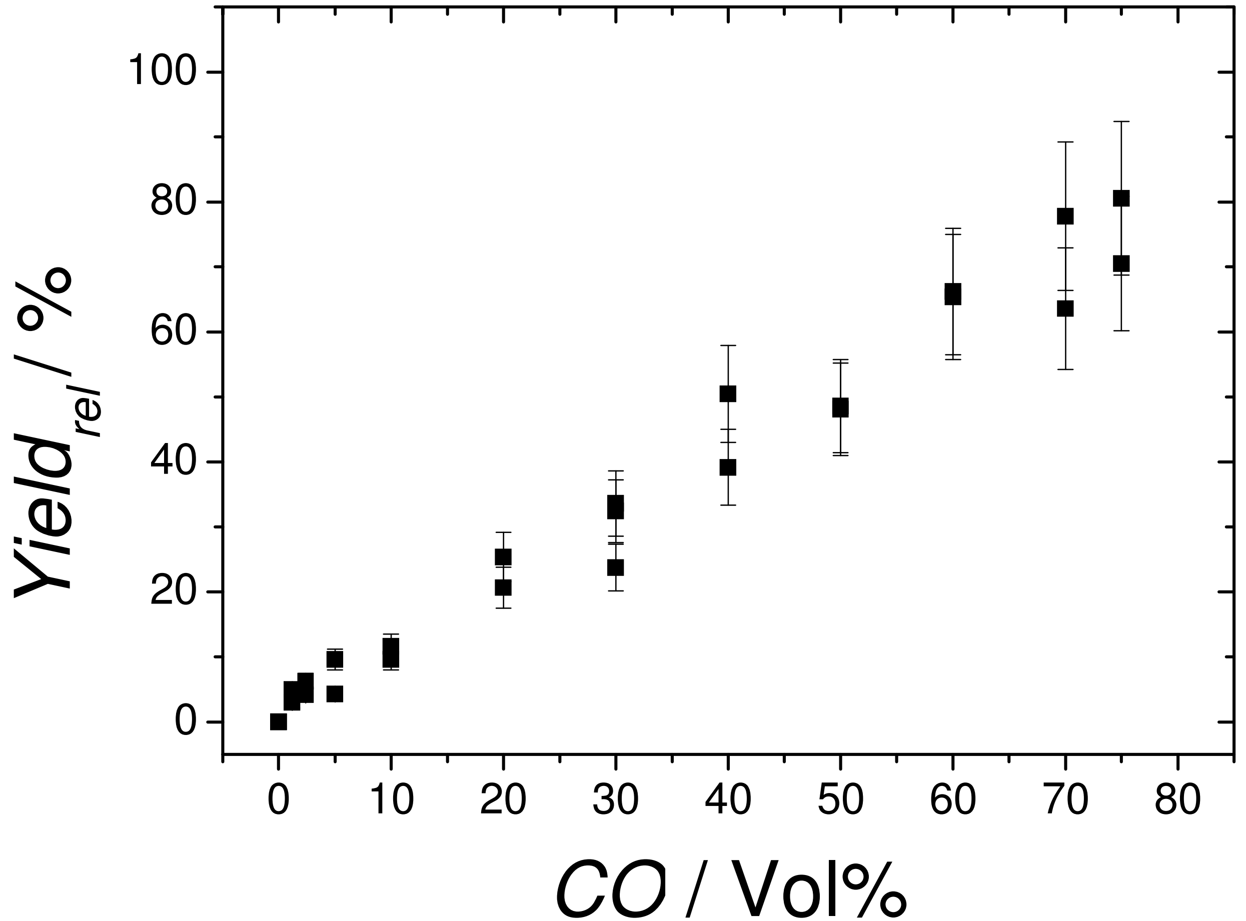
Transport with N_2 / CO mixtures



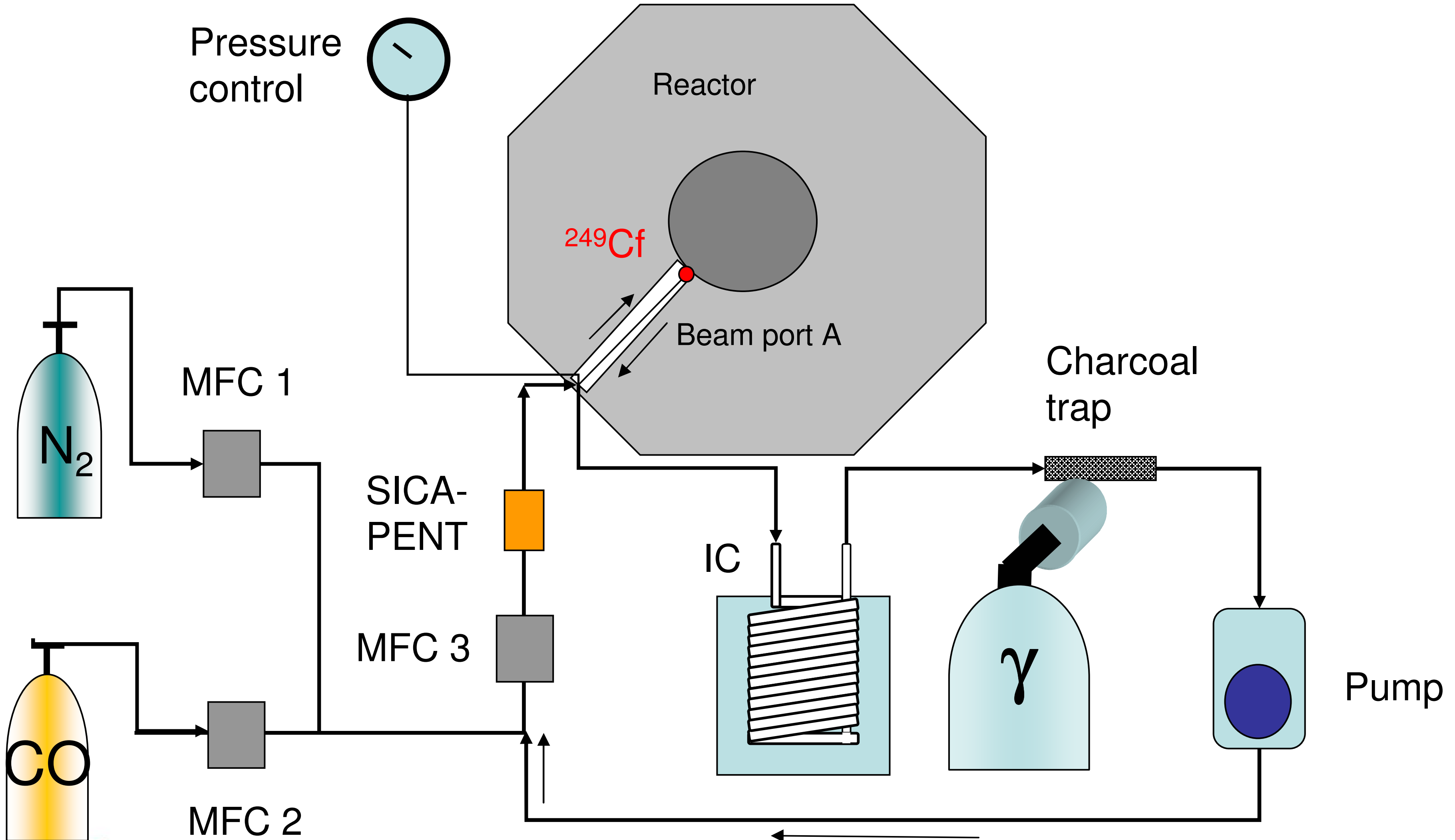
Transport compared to a N_2/KCl cluster jet



^{249}Cf(n,sf)^{104}Mo

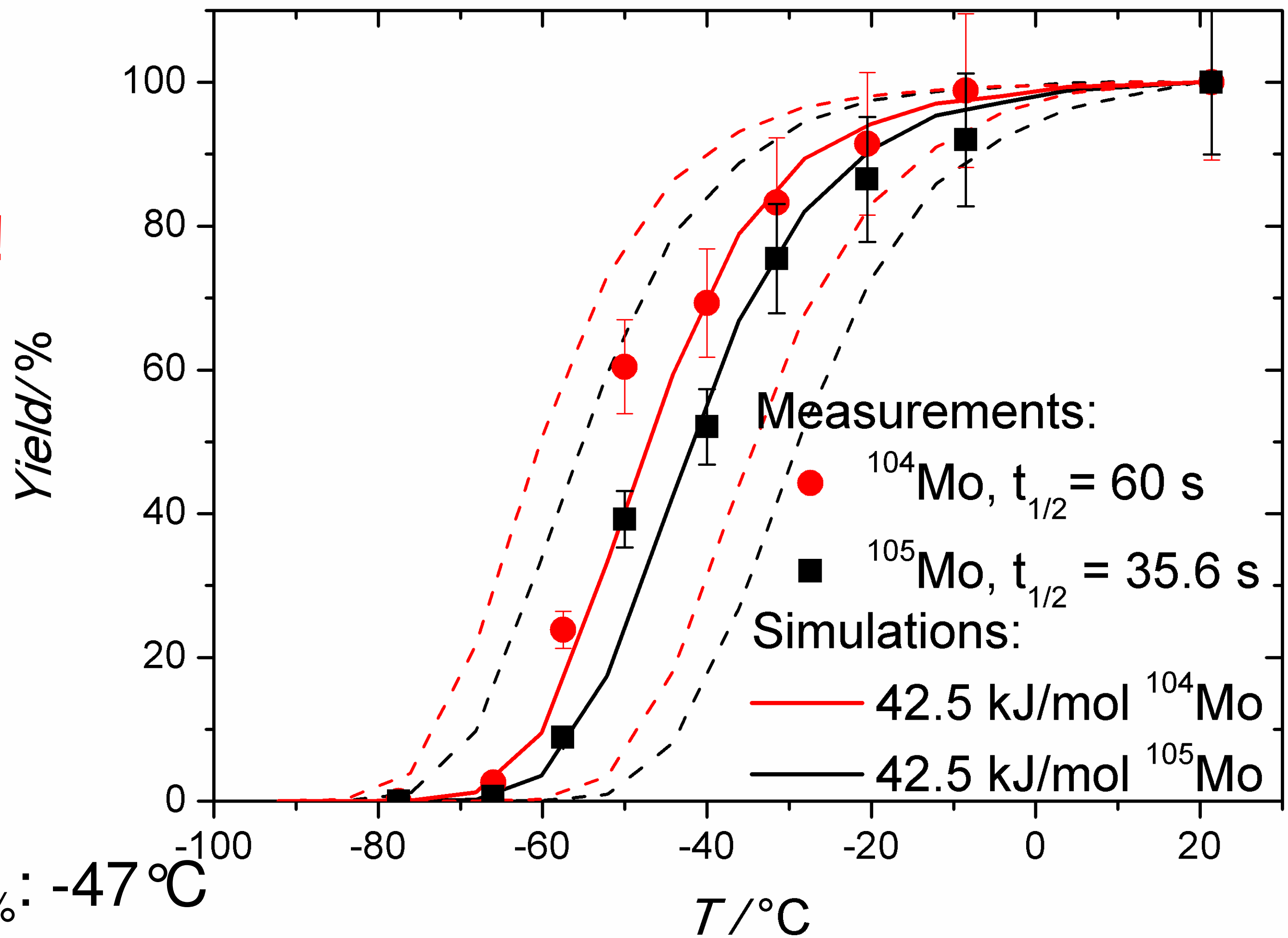
Total gas flow
500 ml/min

Isothermal chromatography - IC



IC of $Mo(CO)_6$ on SiO_2

Physisorption!!!

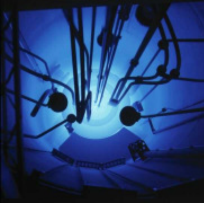


$^{104}Mo(CO)_6$ $T_{50\%}$: -47 °C

$^{105}Mo(CO)_6$ $T_{50\%}$: -42 °C

$-\Delta H_{ads}$ = 42.5 ± 2.5 kJ/mol

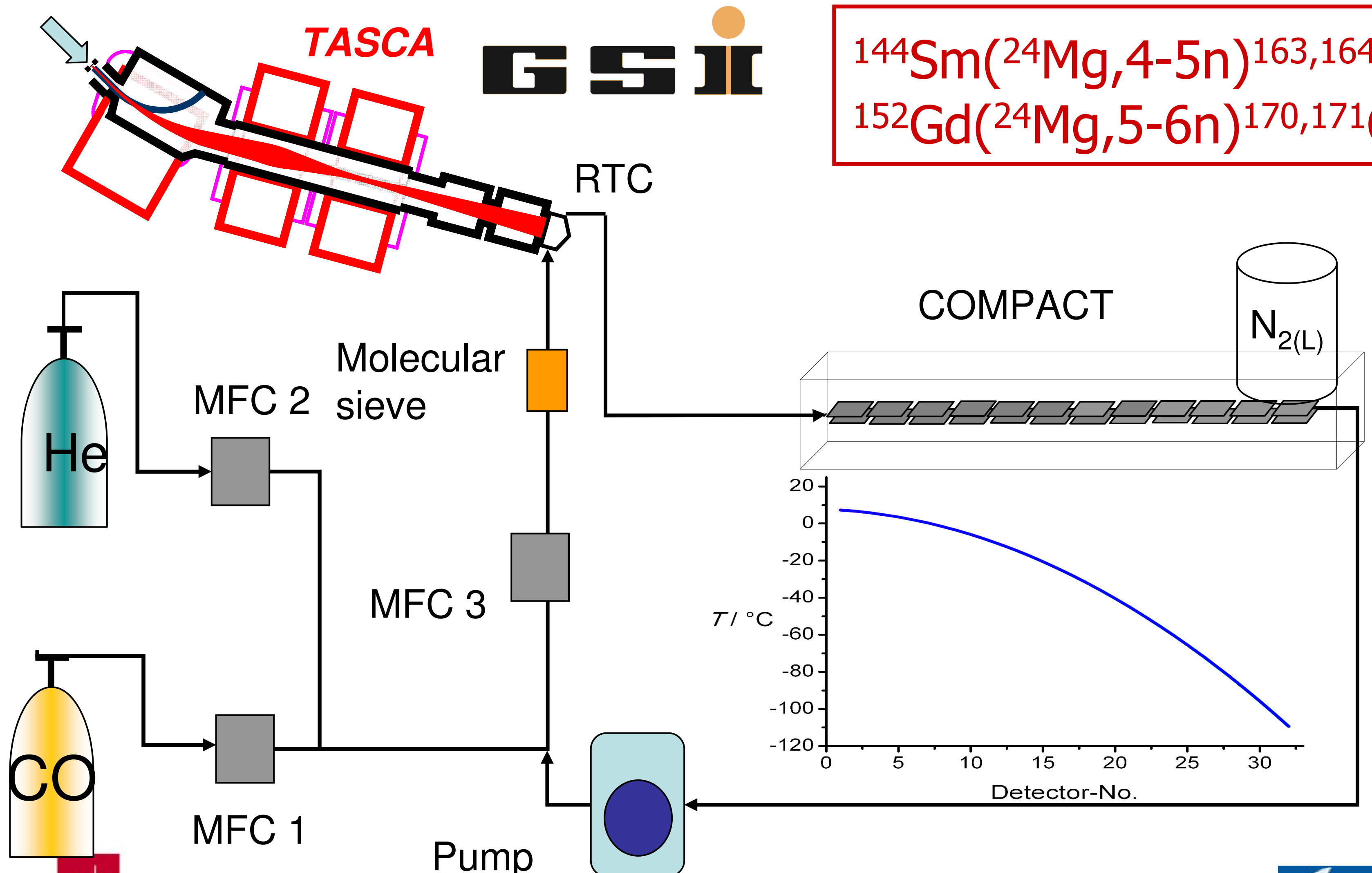
J. Even et al., Angew. Chem. Int. Ed. (submitted)



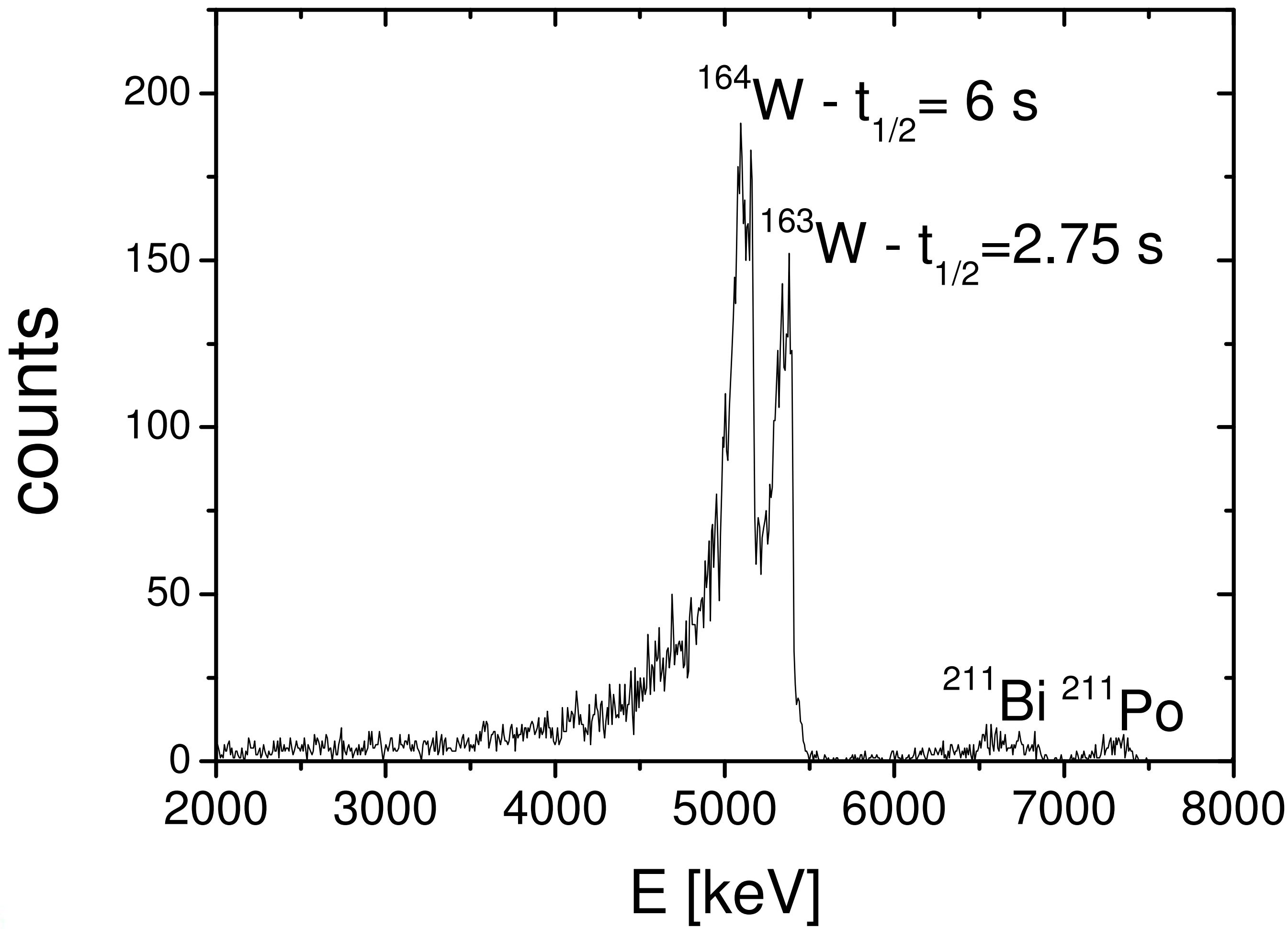
COMPACT @ TASCA

$^{144}Sm(^{24}Mg,4\text{-}5n)^{163,164}W$

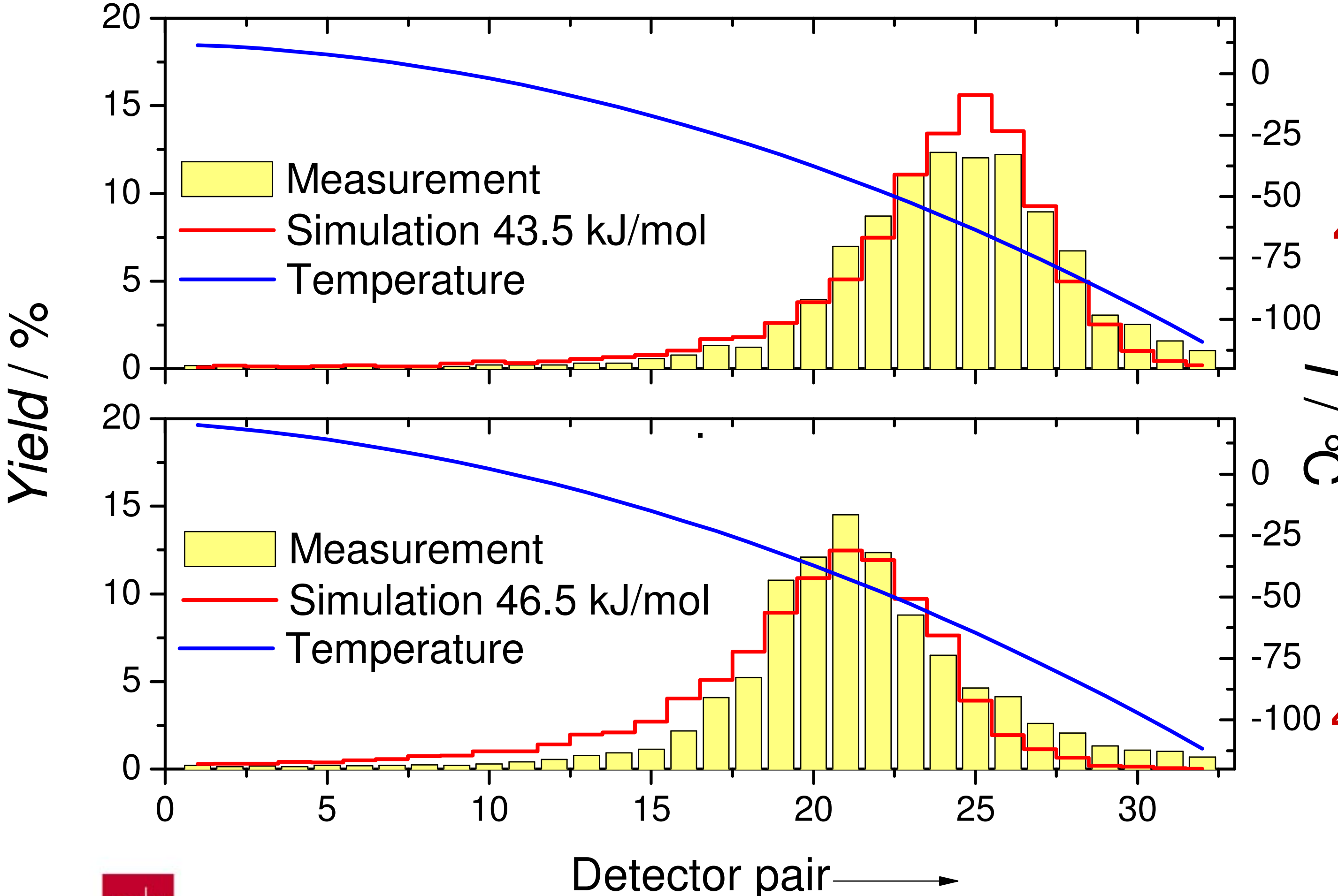
$^{152}Gd(^{24}Mg,5\text{-}6n)^{170,171}Os$



α-spectrum of $^{164,163}W$ measured with COMPACT



Thermochromatography



$Os(CO)_5$

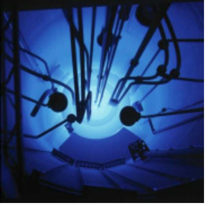
-ΔHads: $43.5^{+2.5}_{-3.5}$ kJ/mol

$W(CO)_6$

-ΔHads: 46.5±2.5 kJ/mol

Physisorption

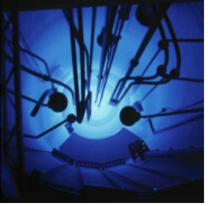
J. Even et al., Angew. Chem. Int. Ed. (submitted)



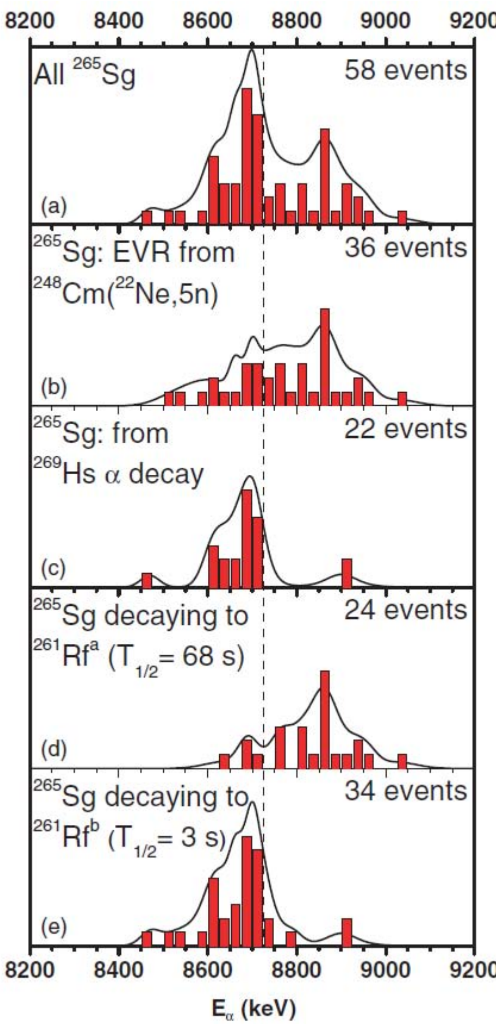
TAN-chemistry gains a new compound class

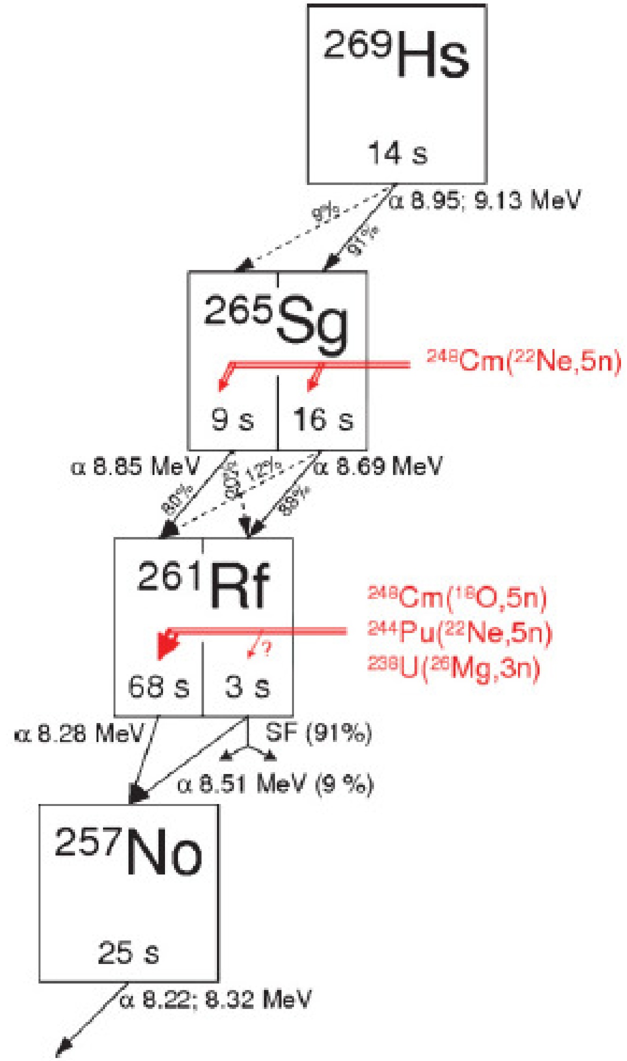
- $Sg(CO)_6$ and $Hs(CO)_5$ are now within reach

⇒ New compound classes of TAN are accessible, e.g., organometallic ones!



Isomeric states of ^{265}Sg



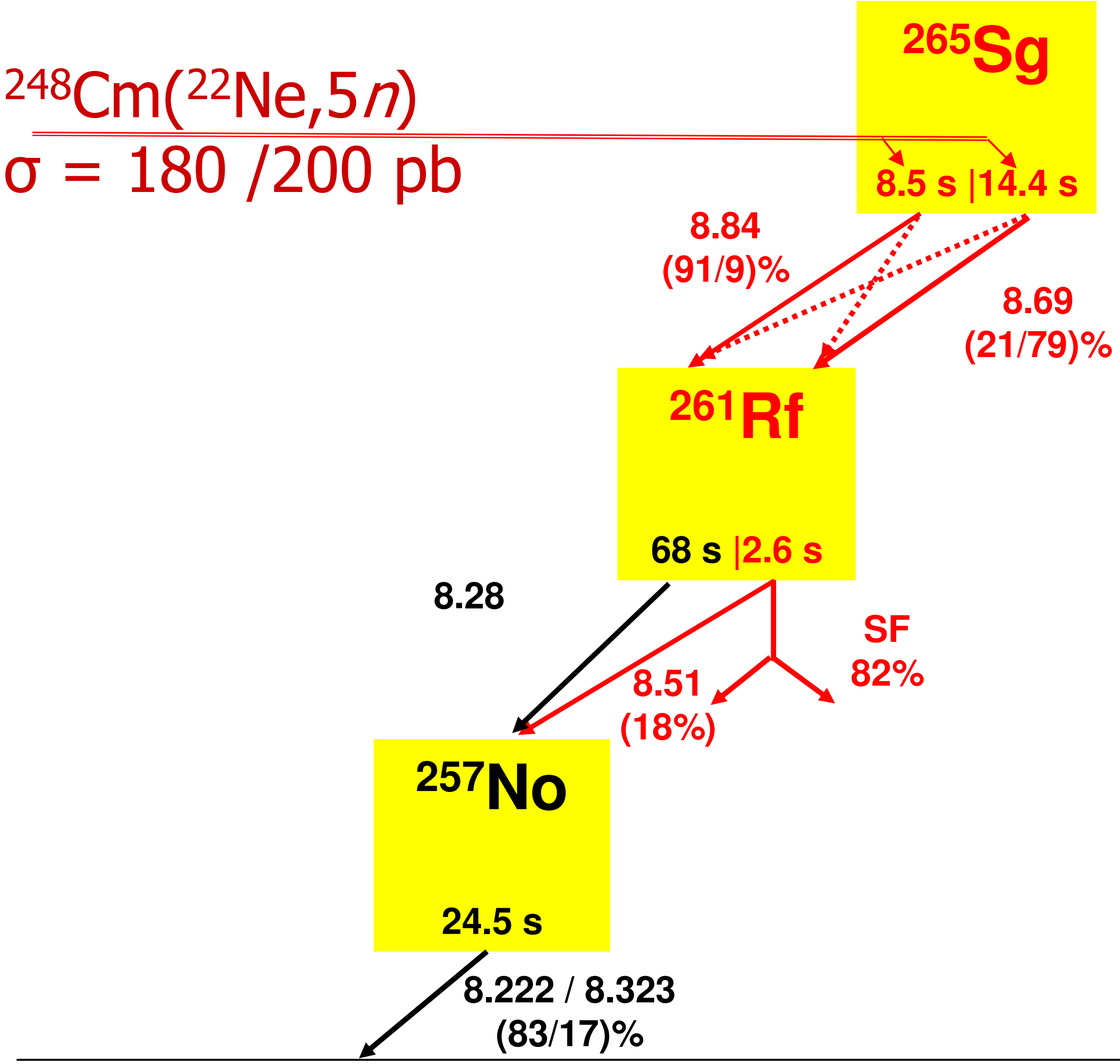


Dissagreement with theory:
Different calculations predict different ground states: ($7/2^+$; $9/2^+$; $11/2^+$)

But
None of these theoretical works predicts an occurrence of a long-lived isomeric state!

Ch.E.Düllmann, A. Türler, Phys.Rev.C **77**, 064320 (2008).

^{248}Cm(^{22}Ne,5*n*)^{265}Sg @ GARIS



H. Haba et al., TAN11, Sochi 2011

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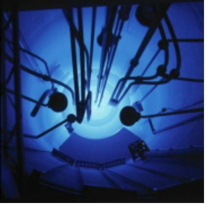
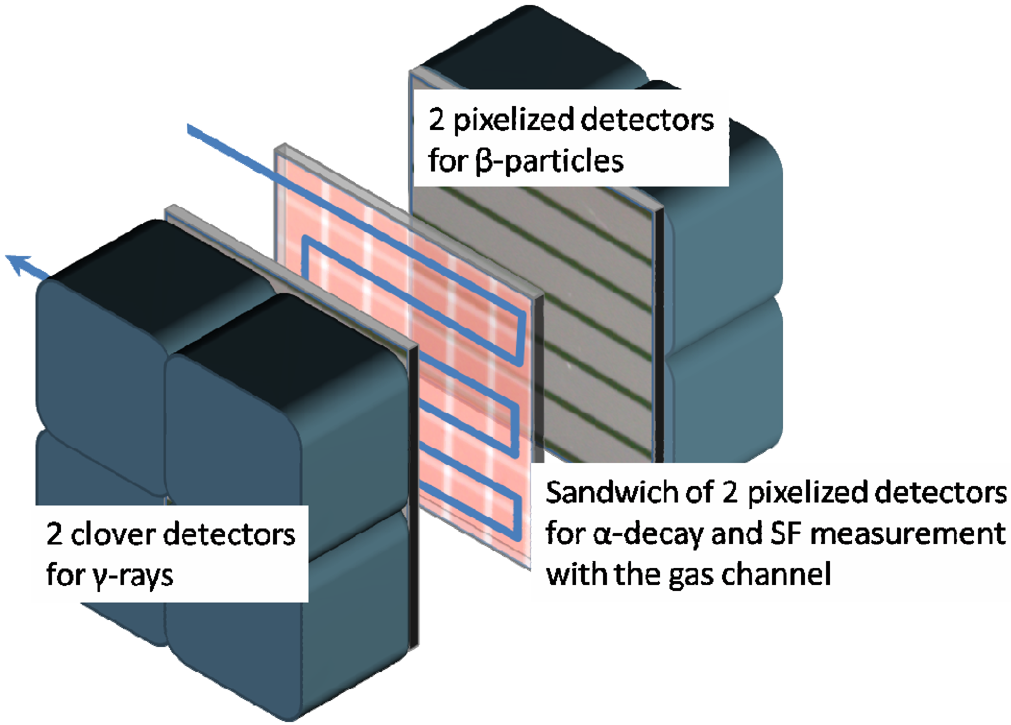


ALpha-BEta-GAmma spectroscopy with chemically separated samples



☺ Nuclear spectroscopy
+
chemistry

~ 100% efficiency
for α and SF detection
(under development)

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A. Yakushev

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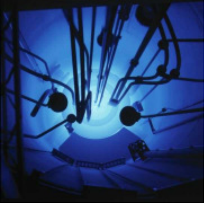
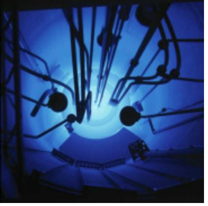


Summary

- Carbonyl complexes of group 4-9 elements
- Fast, efficient, in-situ chemistry (yield > 50%)
- Physisorption on SiO_2 surface (-30℃ to -70℃)

- Chemical and physical applications:
  - Access to a new **TAN-compound class**
  - TAN-carbonyl-chemistry opens the door for a new method in **nuclear spectroscopy** (Rf, Db, Sg, Bh, Hs,(Mt))



Acknowledgments

- Staff of the TRIGA reactor, the mechanical and electronics workshops at the Institute for Nuclear Chemistry, Uni. Mainz
- UNILAC operators, Target lab @ GSI

- Funding: BMBF and HIM

Thank you for your attention!

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