International Conference on Ion Sources (ICIS 2021)

TRIUMF, Vancouver, September 2021

Dynamics and Reactivity of Thermalized Ions in a Radiofrequency Quadrupole Gas-Reaction Cell Used for the Production of Radioactive Molecular Ions

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2) A.E. Lalonde AMS Laboratory, University of Ottawa, Canada
3) TRIUMF, Vancouver, Canada

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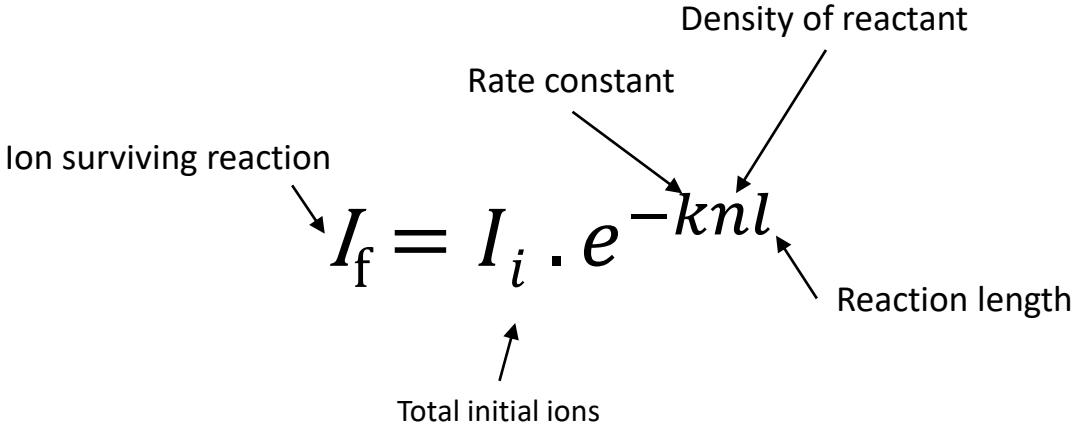
Agenda

- Ion-Molecule Chemistry in Radio Frequency Quadrupoles
- Isobar Separator for Anions – Univ. of Ottawa AMS Laboratory
- Energy Profile in Gas (Thermal Relaxation)
- Formation/Transmission of Radioactive Molecules (TRIUMF, Dr. C. Charles)

Common Ion-Molecule Chemistry at Low Energy



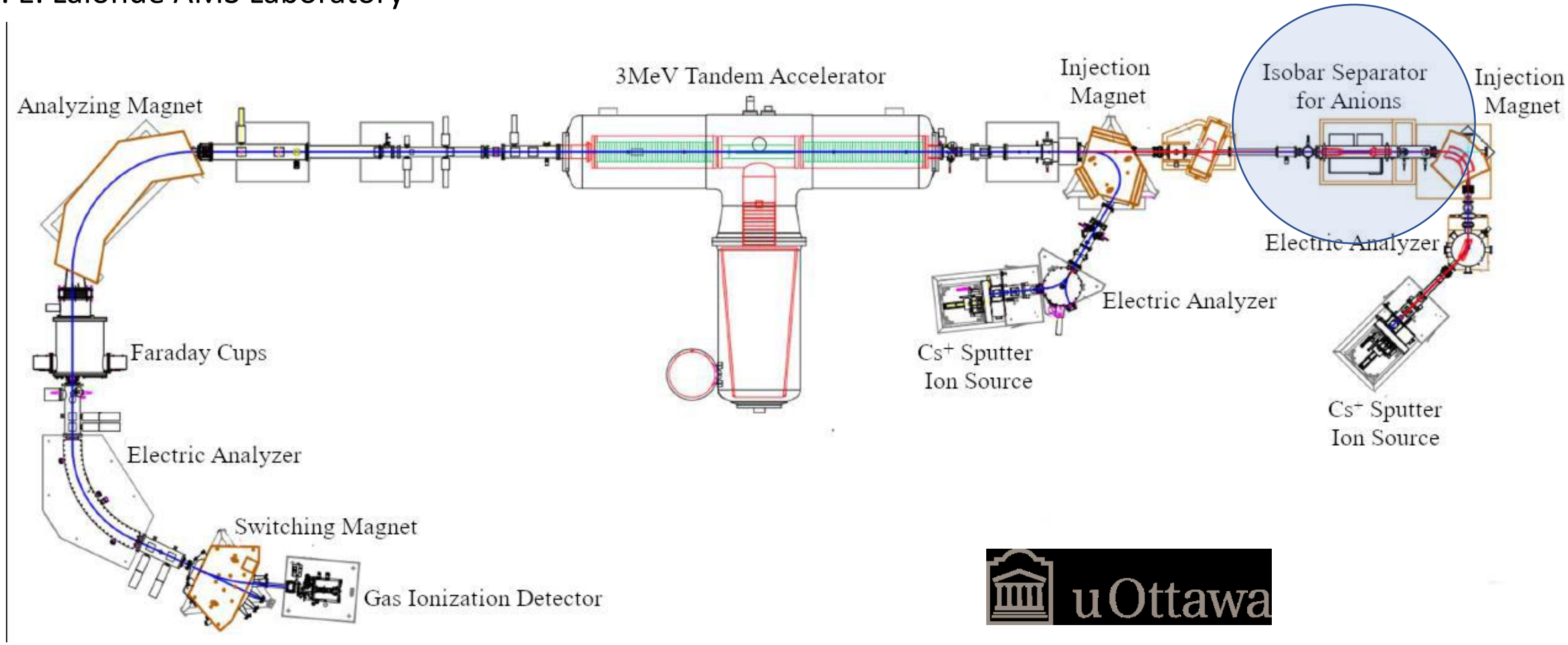
$$k = 2\pi q \left[\frac{\alpha}{\mu}\right]^{1/2}$$

$A^+ + B$	⟶	$B^+ + A$	IE(A)>IE(B)	Charge Transfer
$AC^- + B$	⟶	$A^- + C + B$		Ion Dissociation
$A^- + B$	⟶	$B^- + A$	EA(A)<EA(B)	Electron Transfer
$AH^+ + B$	⟶	$BH^+ + A$	PA(A)<PA(B)	Proton Transfer
$A^+ + B$	⟶	$[A.B]^+$		Adduct formation
$A^+ + BC$	⟶	$AB^+ + C$		Cluster Reaction
$AC^+ + BDE$	⟶	Products		Allowed chemistry

Isobar Separation in Accelerator Mass Spectrometry

Isobar Separator for Anions – Accelerator Mass Spectrometry

A. E. Lalonde AMS Laboratory



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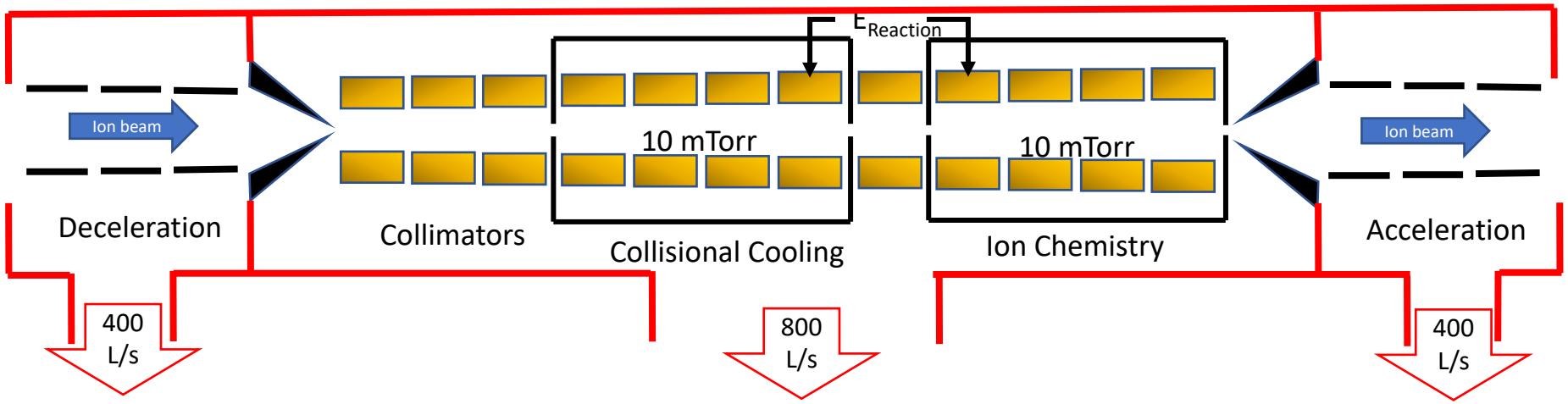
Ion Chemistry in Radio Frequency Confinement Fields



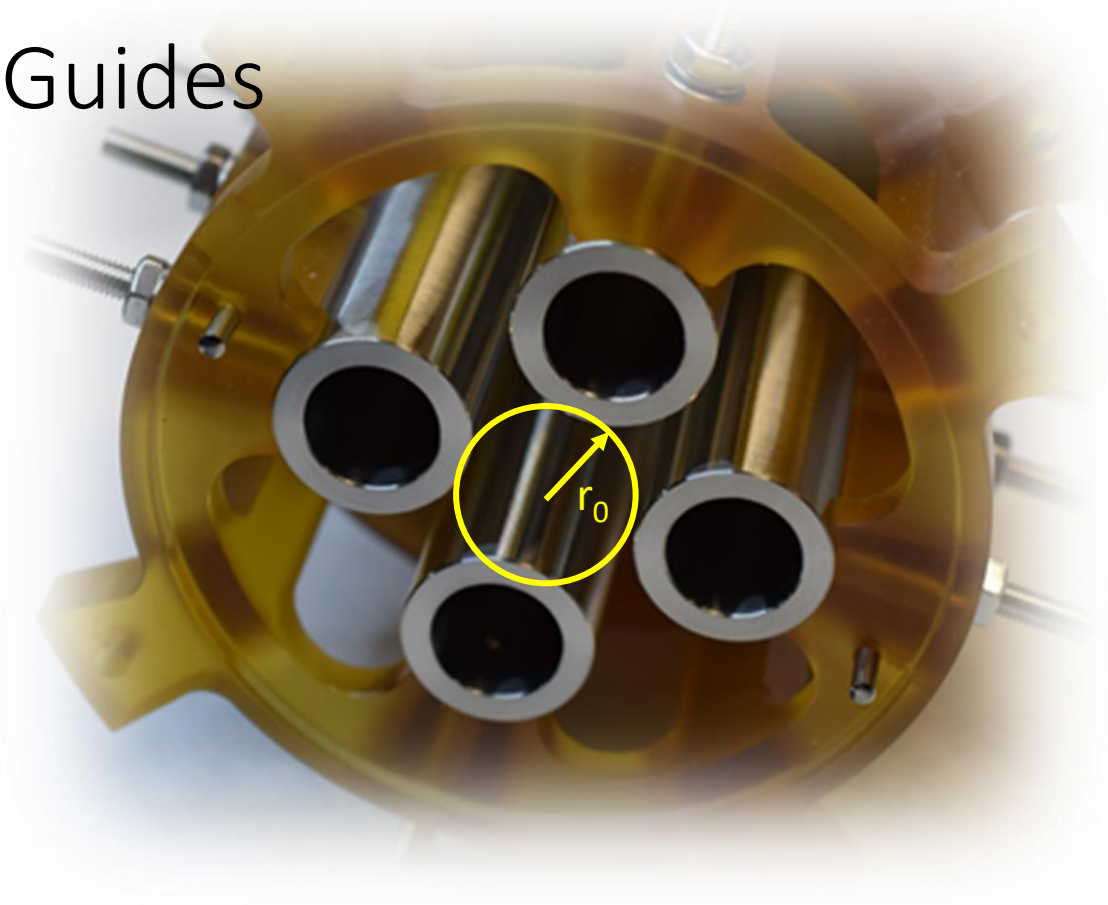
- Reduction of translational ion beam energy by DC lenses
- Minimizes diffusion losses by quadrupolar RF fields, efficiently confine low energy ion beam
- Maximizes acceptance of the quadrupolar fields -> better transmission
- Collisional cooling/focusing and deep RF potential well depth
- Achieve ion chemistry at known and controlled energy near thermal, $E_{lab} < 1eV$
- Controlling reaction length/time using axial fields/segmented quadrupole ion guide

Acceptance of Quadrupolar Ion Guides

Mathieu's stability parameters

$$q = 2\frac{e\,V_{rf}}{m\omega^2 r_0^2}$$

$$a = 4\frac{e\,V_{dc}}{m\omega^2 r_0^2} = 0$$

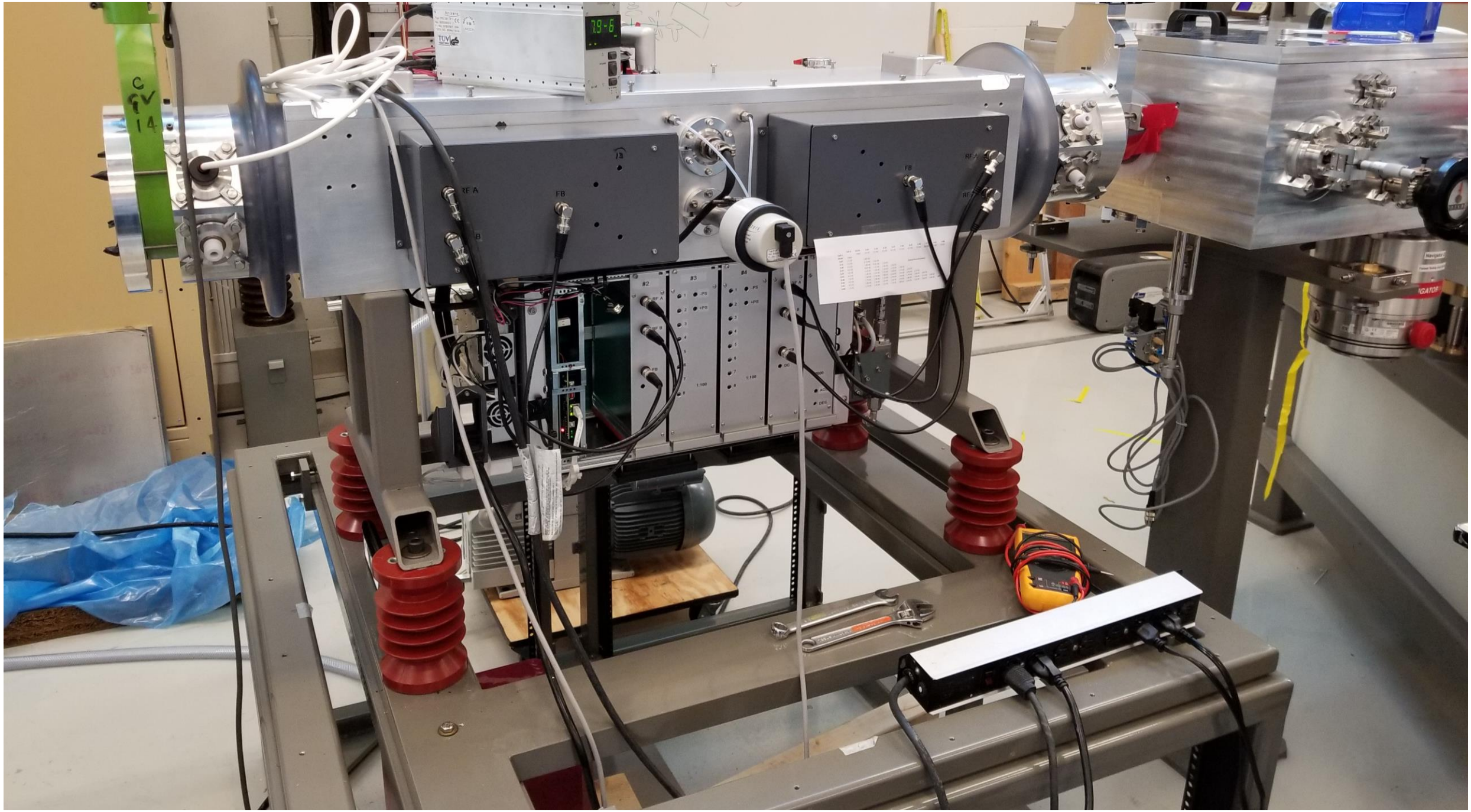


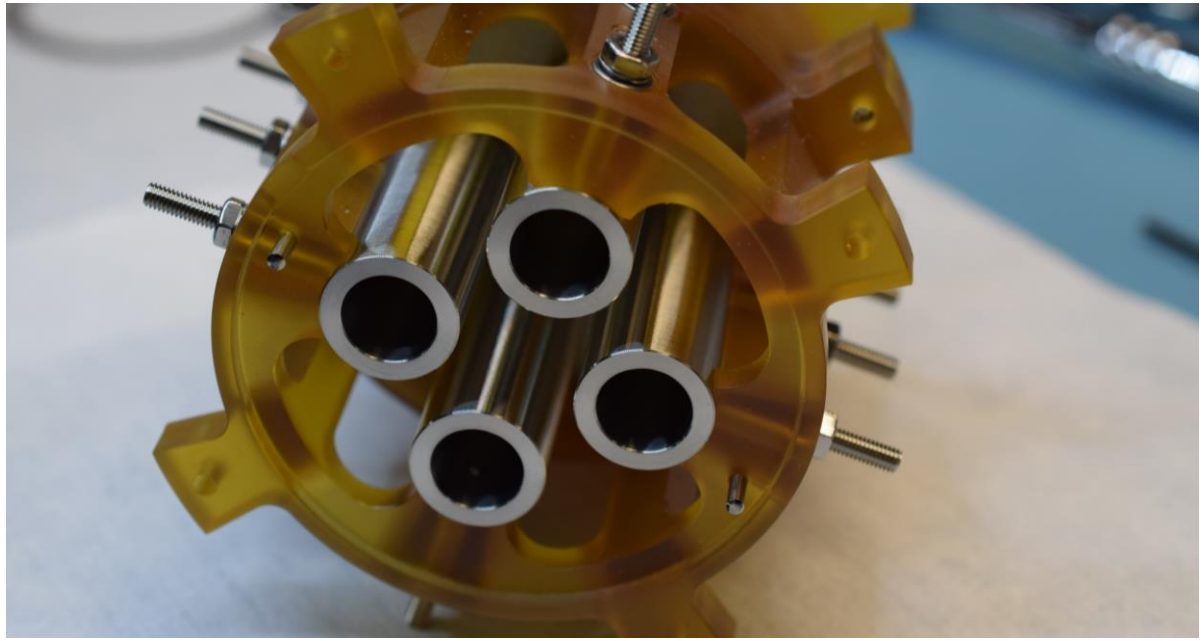
where:

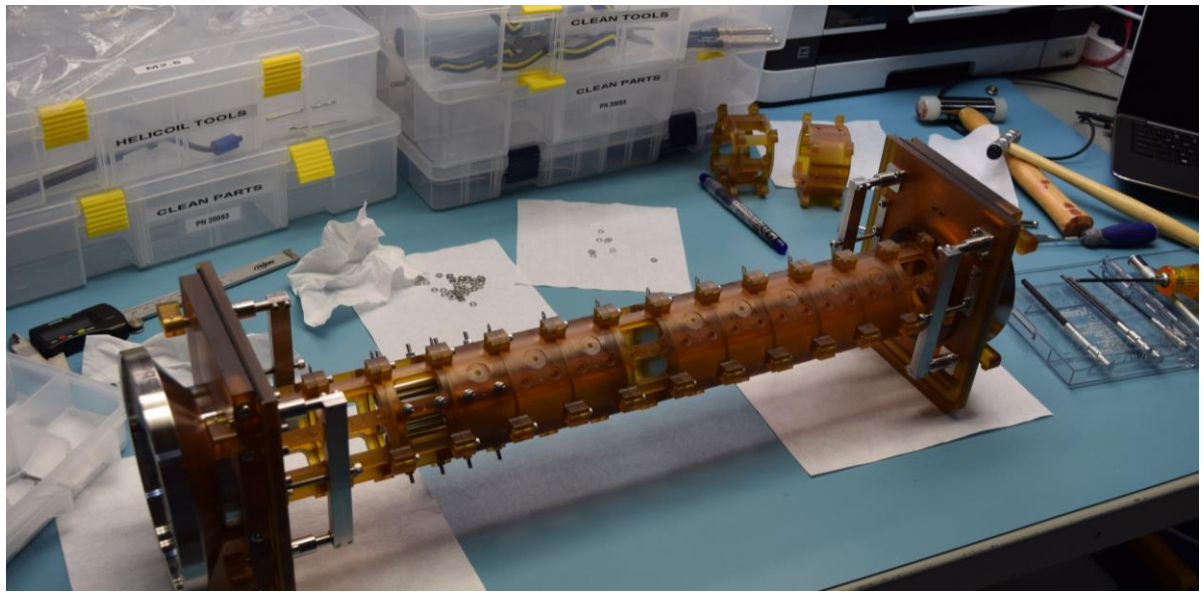
- e is the elementary charge
- V_{rf} is the peak-to-peak RF voltage
- V_{dc} is the DC voltage between the pole pairs
- m is the ion mass
- ω is the angular RF frequency, and
- r_0 is the radius of a circle tangential to the inner surface of the rods.

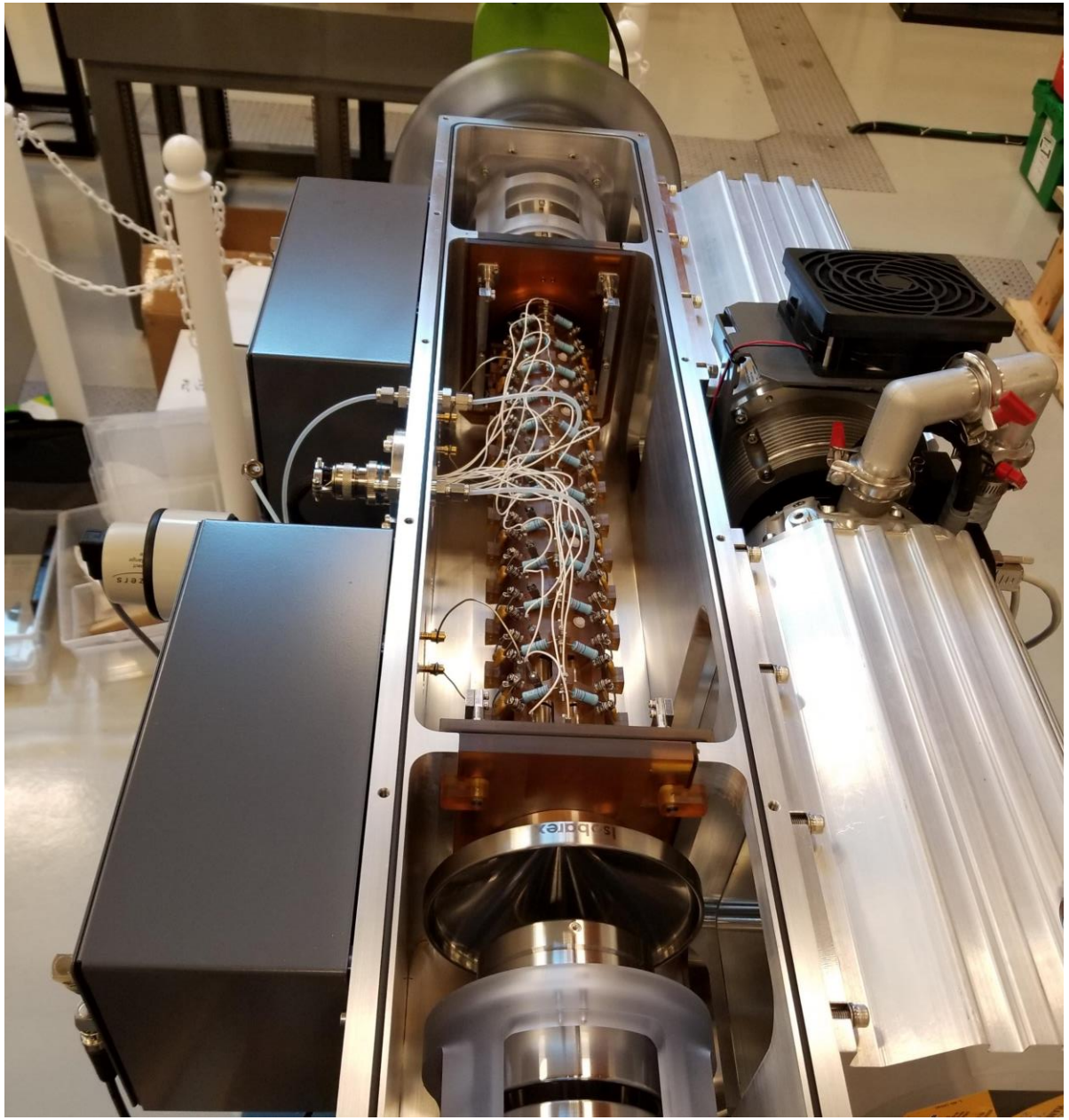
$$\mathrm{Acc} \approx r_0^{\,4}\,\omega^2$$

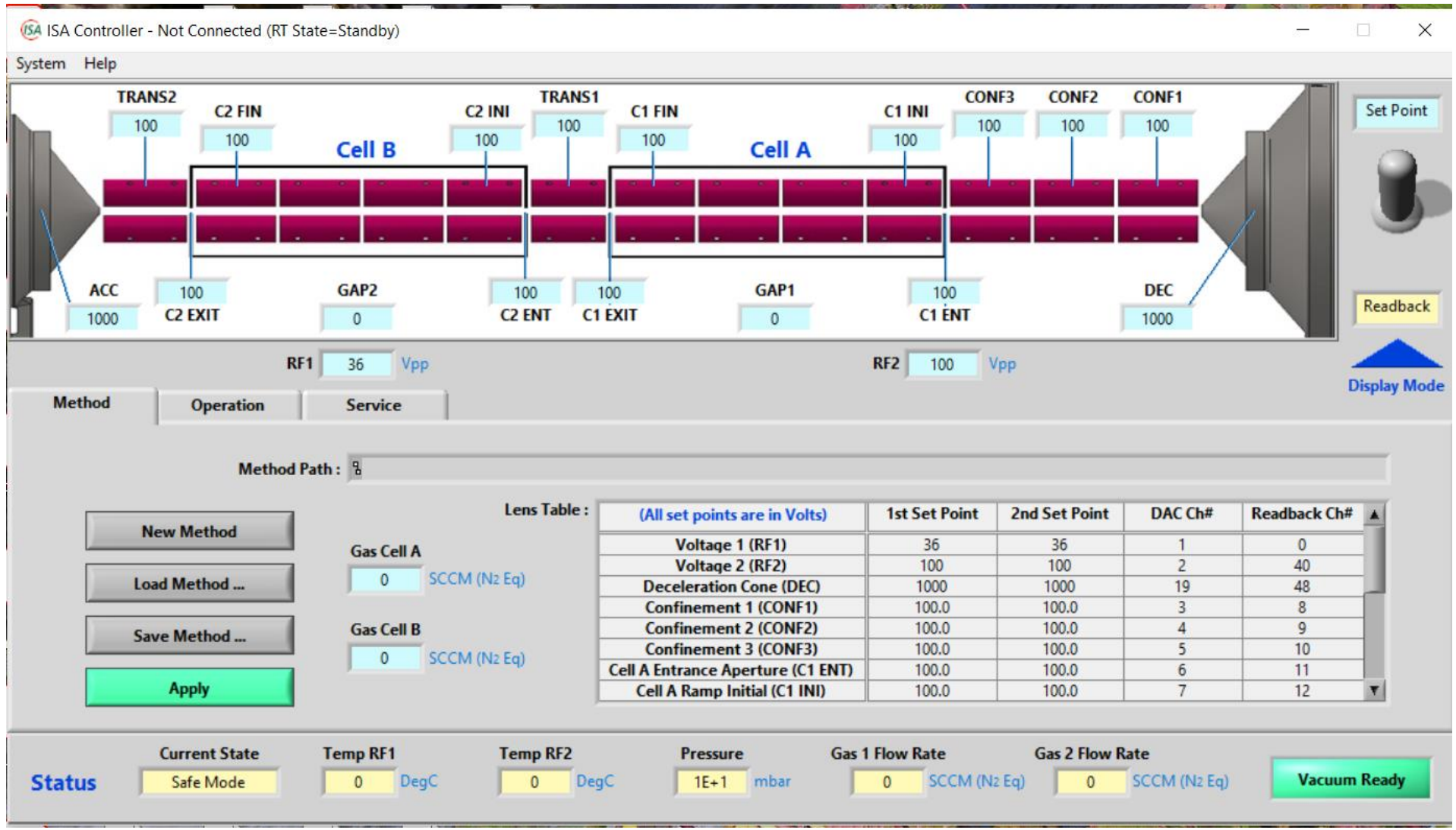
ISA hardware designed to maximize acceptance of quadrupolar fields


RF A
FB
#2
#3
#4



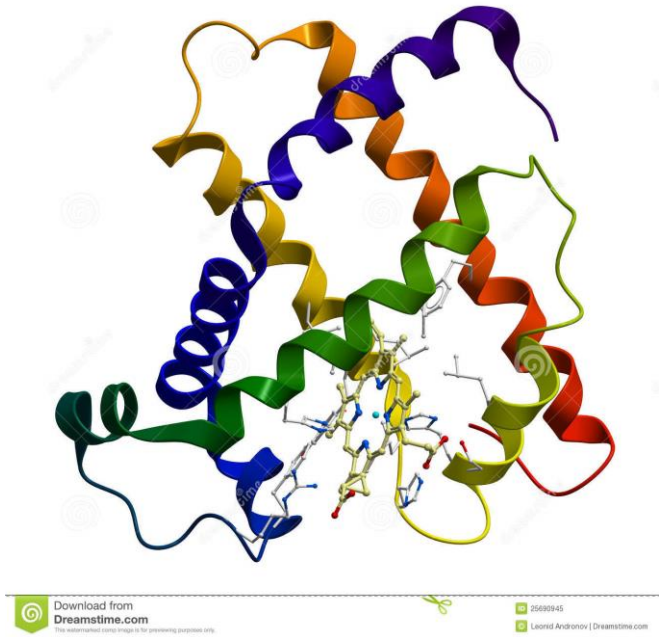

CLEAN TOOLS
CLEAN PARTS
HELICOIL TOOLS
CLEAN PARTS




ISA Controller - Not Connected (RT State=Standby)
System
Help
TRANS2
C2 FIN
Cell B
C2 INI
TRANS1
C1 FIN
Cell A
C1 INI
CONF3
CONF2
CONF1
100
ACC
1000
C2 EXIT
GAP2
0
C2 ENT
C1 EXIT
GAP1
C1 ENT
DEC
Set Point
Readback
Display Mode
RF1
36
Vpp
RF2
100
Vpp
Method
Operation
Service
Method Path :
New Method
Load Method ...
Save Method ...
Apply
Lens Table :
Gas Cell A
0
SCCM (N2 Eq)
Gas Cell B
0
SCCM (N2 Eq)
(All set points are in Volts)
1st Set Point
2nd Set Point
DAC Ch#
Readback Ch#
Voltage 1 (RF1) 36 36 1 0
Voltage 2 (RF2) 100 100 2 40
Deceleration Cone (DEC) 1000 1000 19 48
Confinement 1 (CONF1) 100.0 100.0 3 8
Confinement 2 (CONF2) 100.0 100.0 4 9
Confinement 3 (CONF3) 100.0 100.0 5 10
Cell A Entrance Aperture (C1 ENT) 100.0 100.0 6 11
Cell A Ramp Initial (C1 INI) 100.0 100.0 7 12
Status
Current State
Safe Mode
Temp RF1
0
DegC
Temp RF2
0
DegC
Pressure
1E+1
mbar
Gas 1 Flow Rate
0
SCCM (N2 Eq)
Gas 2 Flow Rate
0
SCCM (N2 Eq)
Vacuum Ready

Collisional Focusing Effects in Radio Frequency Quadrupoles

Douglas & French, J. Am. Soc. Mass Spectrom. 1992, 3, 4, 398–408



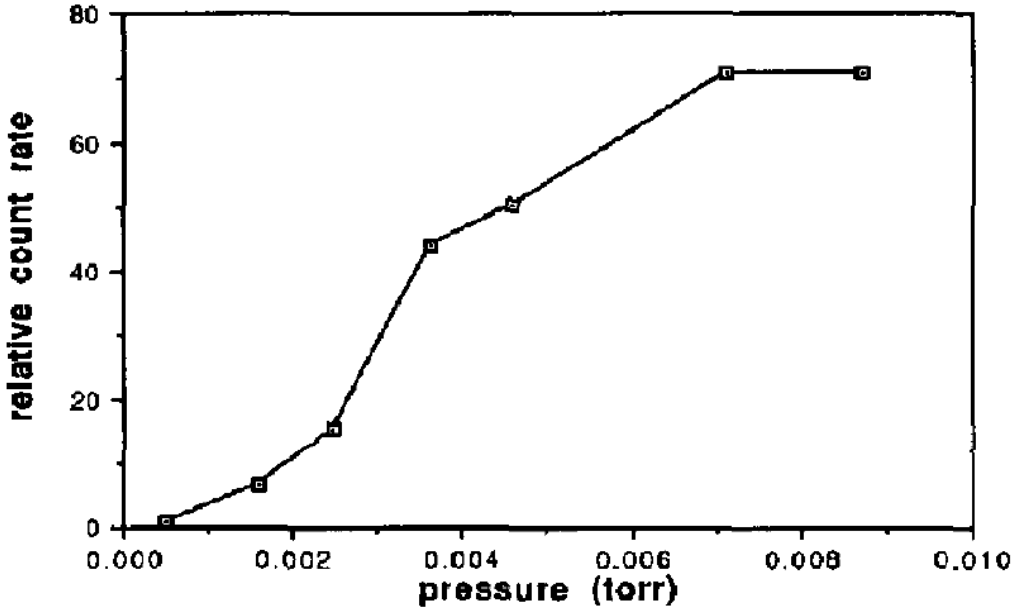


Figure 5. The variation of relative count rate for ions of m/z 1304 (+13) from myoglobin with pressure in the rf quadrupole. The results are normalized to 1.0 at a pressure of 5.0×10^{-4} torr.

Myoglobin/N_2 mass ratio = 46.6

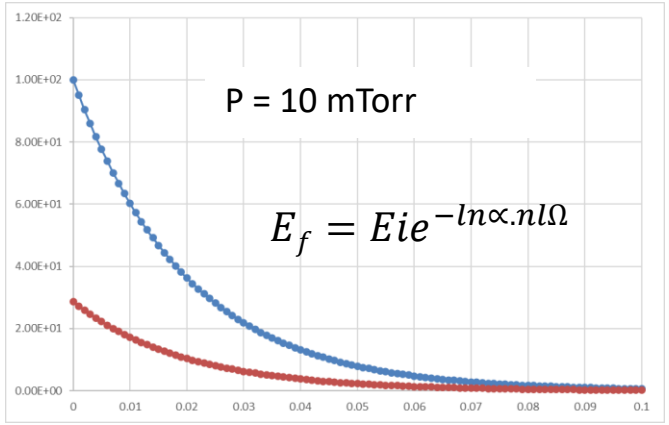
Single Cell ISA

Collisional cooling proceed before ion chemistry

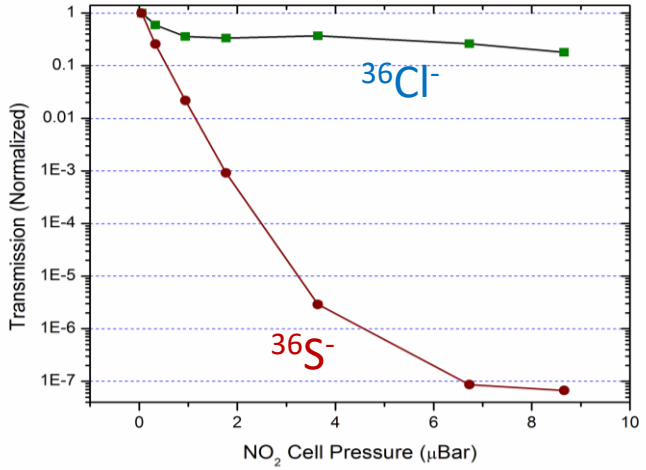
$$V_d = \frac{3}{16}\frac{V_{acc}e}{dn}\left(\frac{2\pi}{\mu kT}\right)^{1/2}\frac{1}{\Omega_T}$$

E_{lab}

E_{CM}

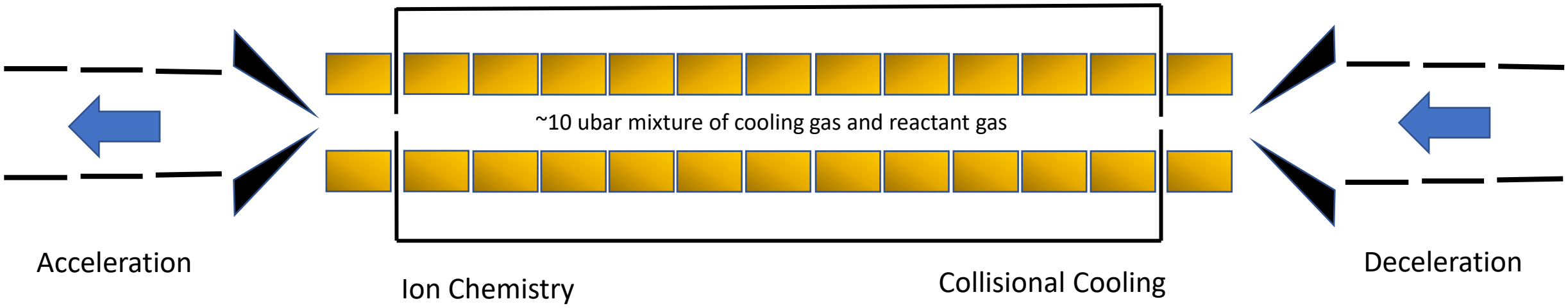


$$I_f = I_i . e^{-knl}$$



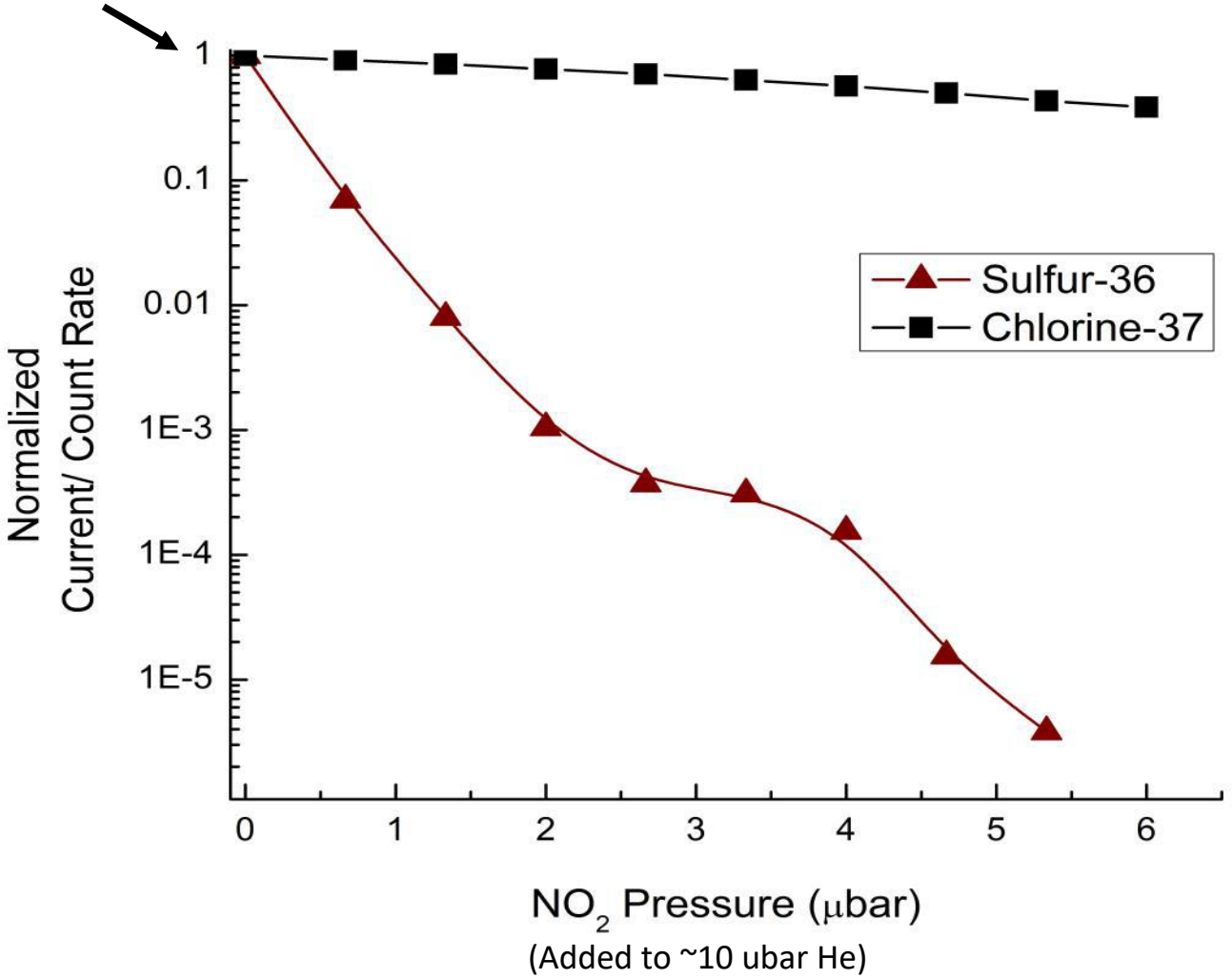
$$V_d = \frac{3}{16}\frac{V_{acc}e}{dn}\left(\frac{2\pi}{\mu kT}\right)^{1/2}\frac{1}{\Omega_T}$$

$$\Omega_T = \frac{2\pi q}{V_d}\sqrt{\frac{\alpha}{\mu}}$$



Selective Suppression of S^-

40-50% transmission for $37Cl^-$ established first in ~10 ubar He
(Mass ratio 37Cl/4He = 9.25)



$Cl^- + NO_2 \rightarrow Cl + NO_2^-$

endothermic by 1.34 eV
No reaction

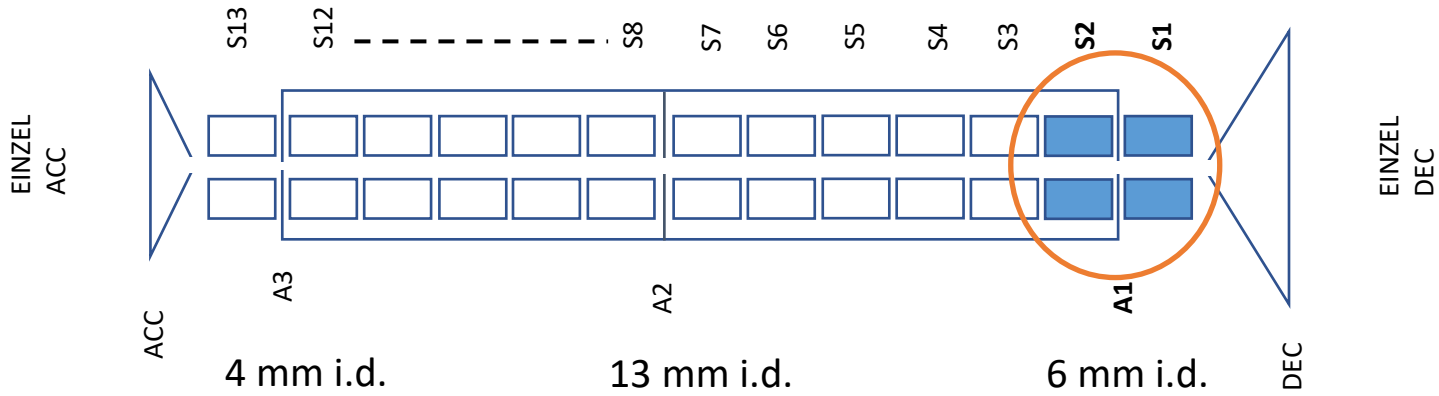
$S^- + NO_2 \rightarrow S + NO_2^-$

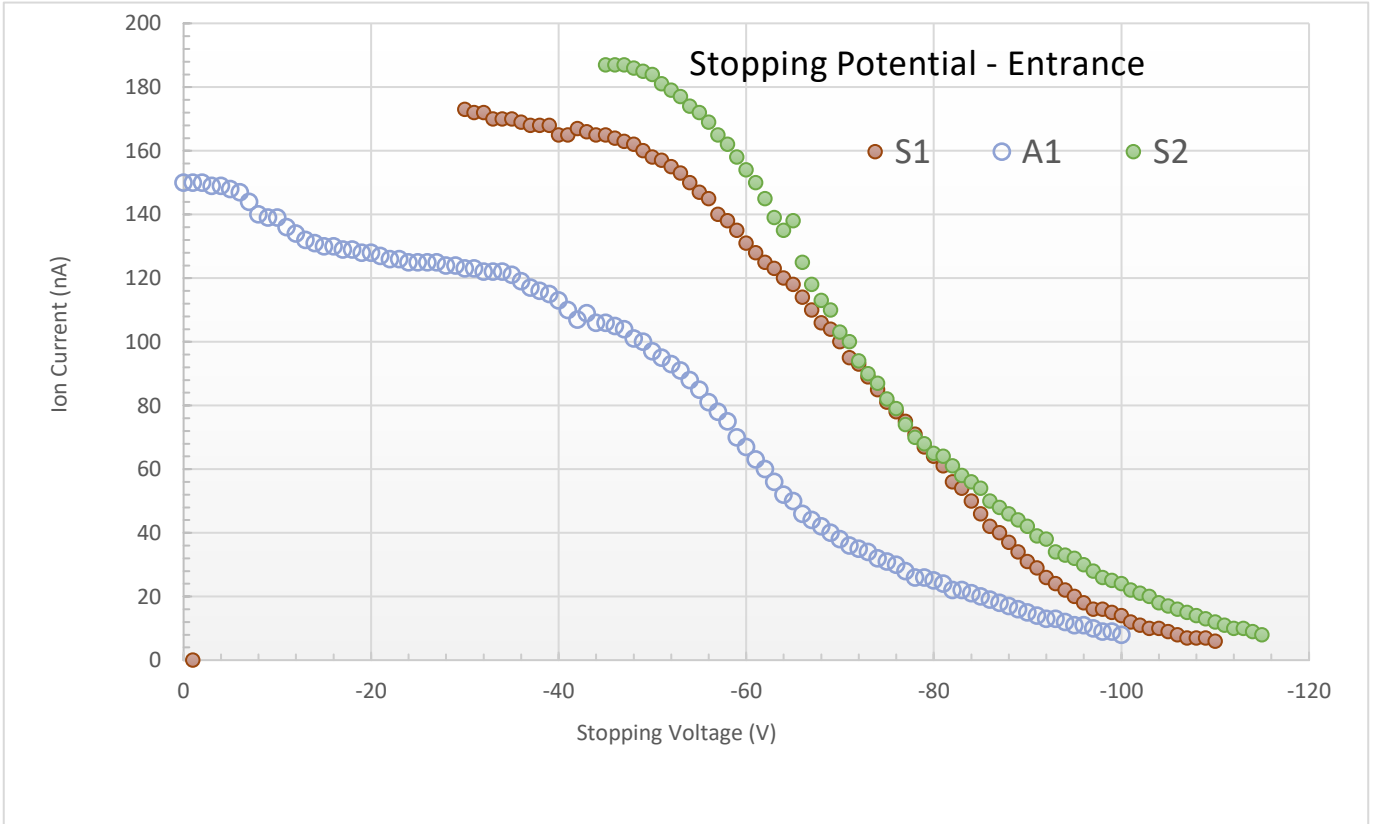
exothermic by 0.2 eV
Fast reaction

$^{37}Cl^-$

Source Target: 7 kV
Beam: 20 kV
Source Einzel: 13.8 kV (readback)
Magnet Slits: 4 mm (symmetrical)
ΔVDeck: +62 V (-20.053kV)

Helium: 9 SCCM (12 µbar in cell)
Mean Free Path: 8.6 mm





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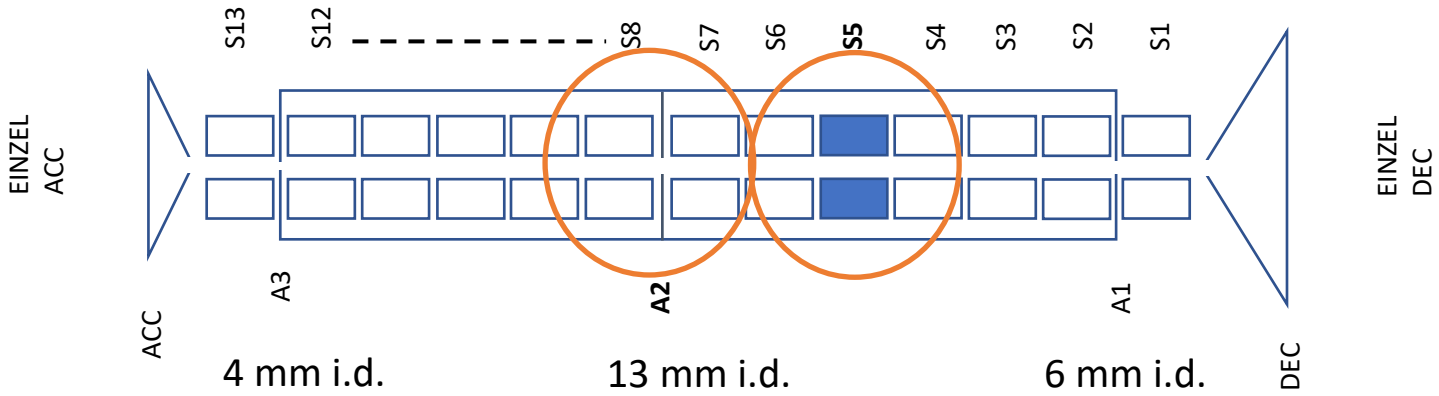

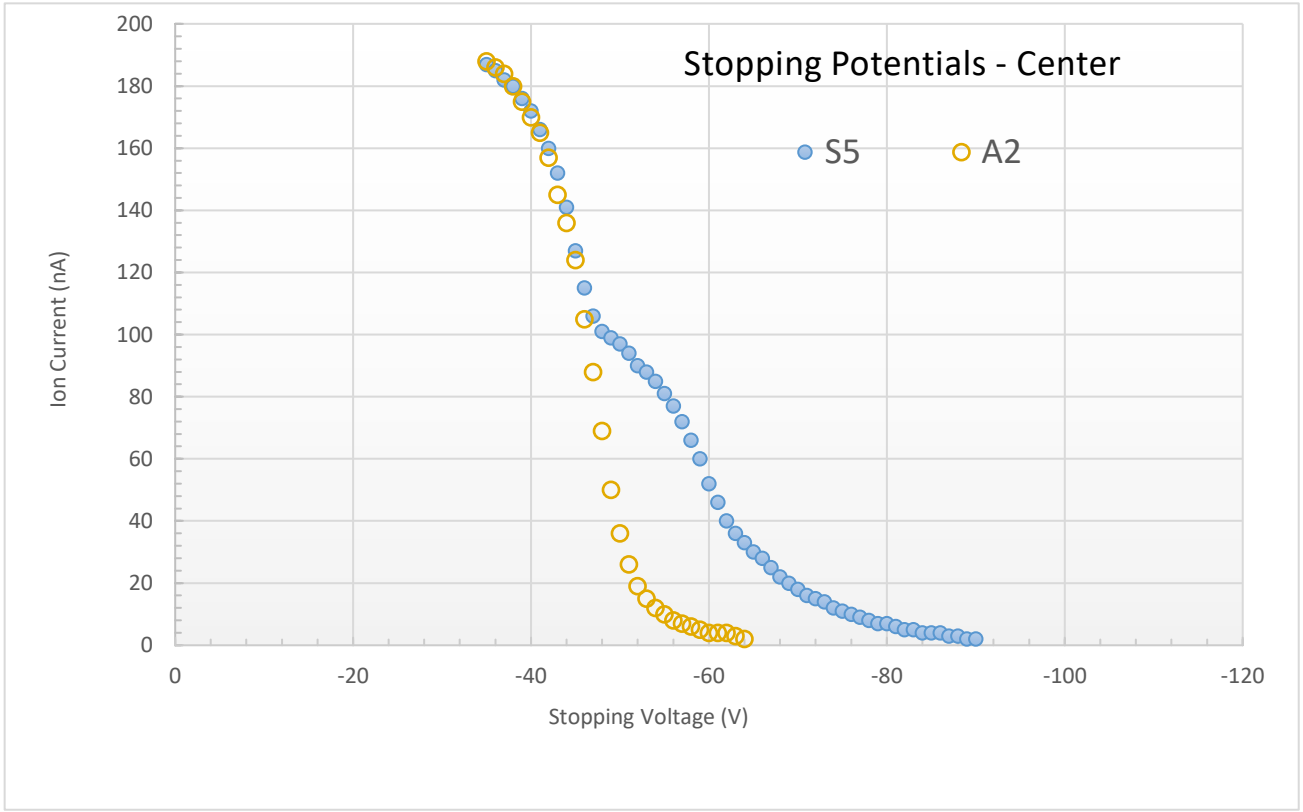

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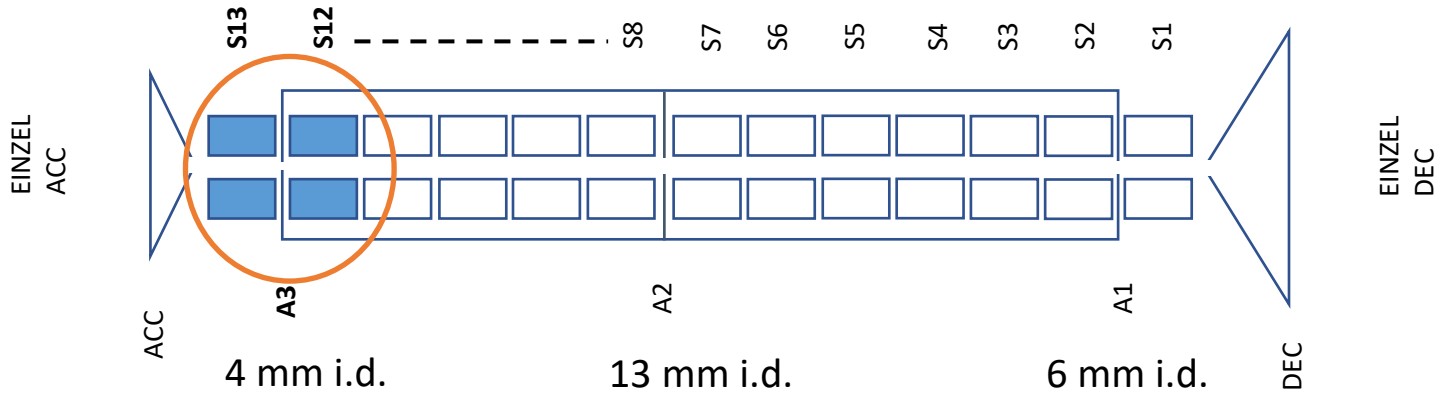

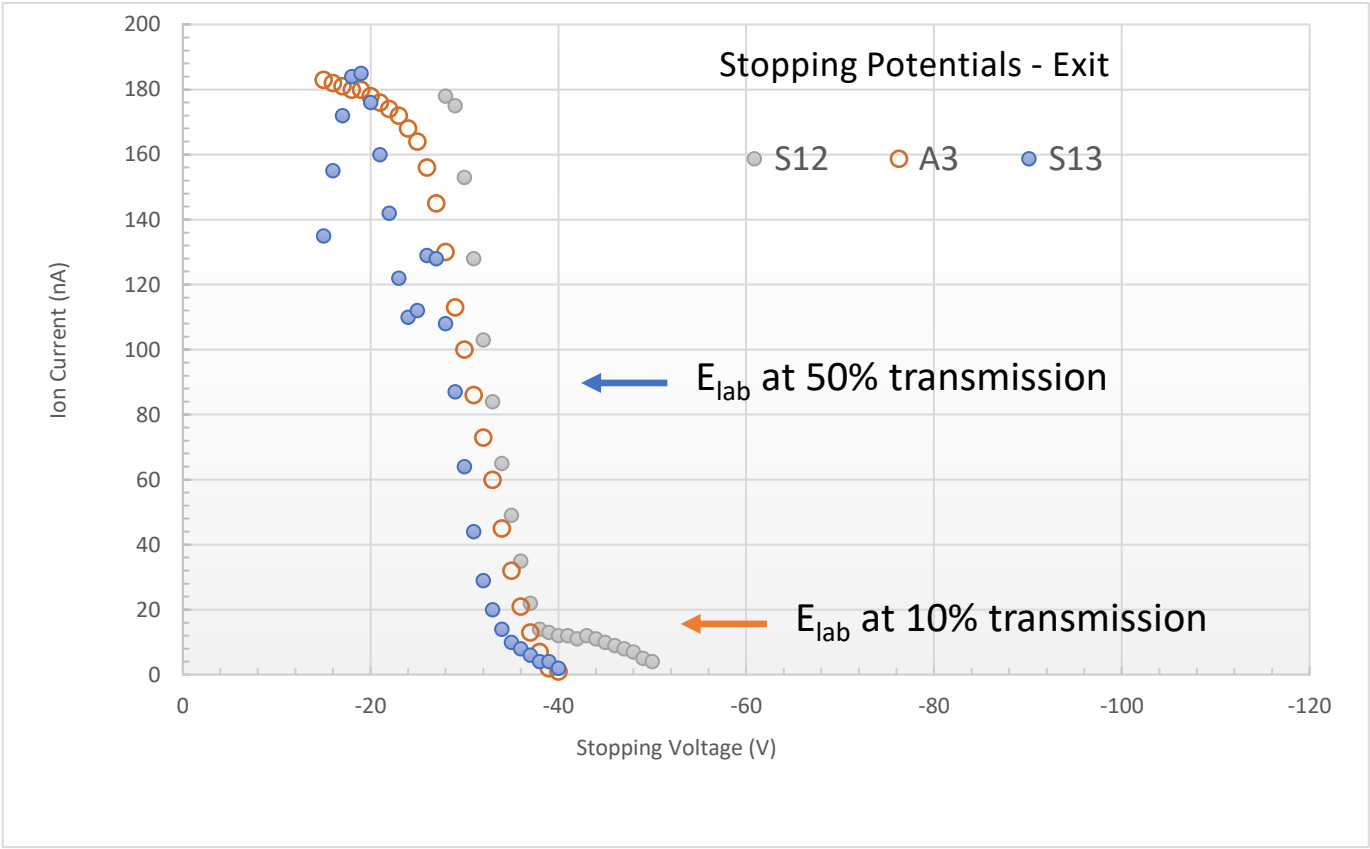

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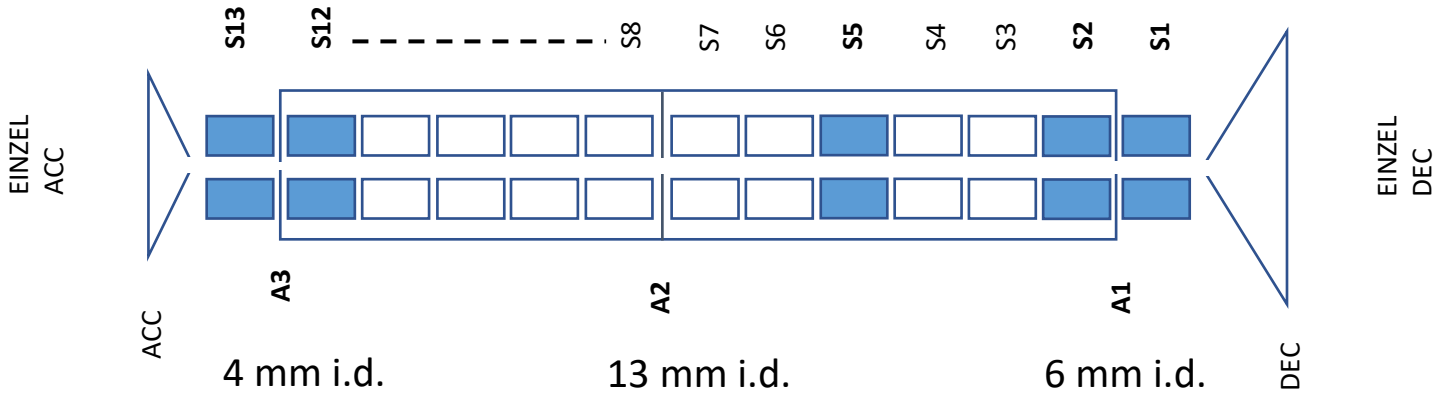
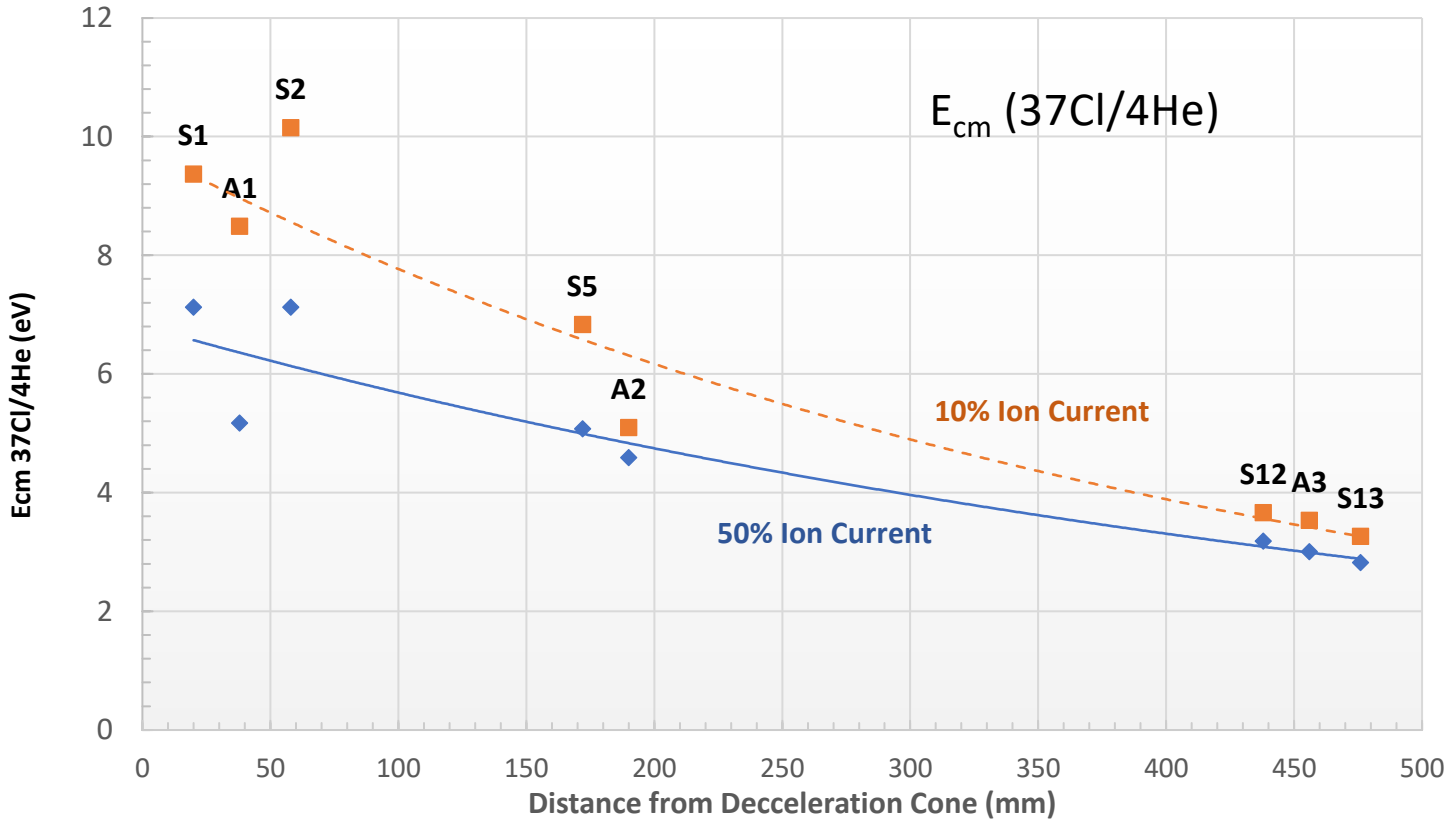
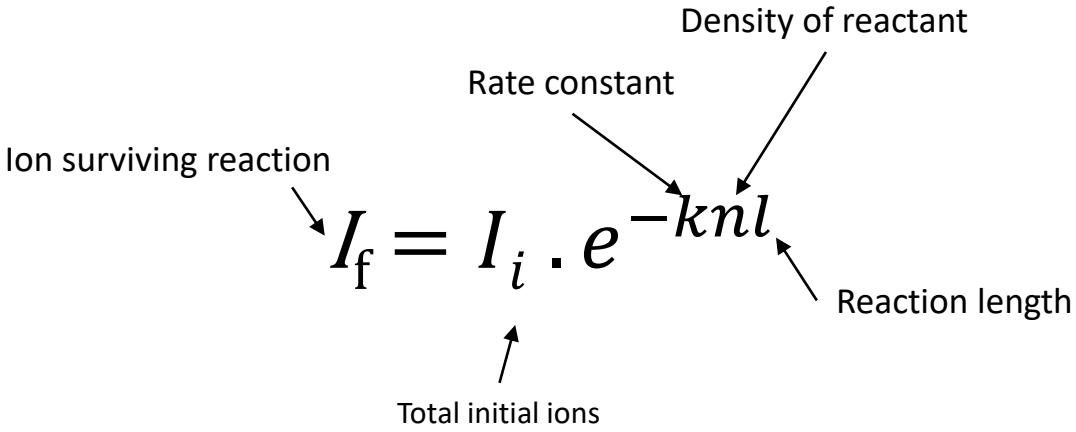
 # Collisions: 53

$E_{cm} = E_{lab} m_n/(m_n+m_i)$





Common Ion-Molecule Chemistry



$A^+ + B$	$B^+ + A$	IE(A)>IE(B)	Charge Transfer
$AC^- + B$	$A^- + C + B$		Ion Dissociation
$A^- + B$	$B^- + A$	EA(A)<EA(B)	Electron Transfer
$AH^+ + B$	$BH^+ + A$	PA(A)<PA(B)	Proton Transfer
$A^+ + B$	$[A.B]^+$		Adduct formation
$A^+ + BC$	$AB^+ + C$		Cluster Reaction
$AC^+ + BDE$	Products		Allowed chemistry

On-line Synthesis of Radioactive Molecules

Examples of Ion-Molecule Chemistry at Thermal Equilibrium

Adduct Formation Reactions

$Ga^+ + C_6H_6 \xrightarrow[1.2x\ 10^{-9}]{100\%} Ga.C_6H_6^+$

$Cu^+ + C_6F_6 \xrightarrow[1.0x\ 10^{-9}]{100\%} Cu.C_6F_6^+$

$Pt^+ + SF_6 \xrightarrow[2.3x\ 10^{-11}]{100\%} Pt.SF6^+$

Cluster Formation Reactions

$W^+ + CH_3F \xrightarrow[1.2x\ 10^{-10}]{95\%} W.CHF^+$

$\xrightarrow[1.2x\ 10^{-10}]{5\%} W.CH_3F^+$

Allowed Chemistry

$V^+ + CH_3F \xrightarrow[1.7x\ 10^{-10}]{100\%} VF^+$

$Hf^+ + CO_2 \xrightarrow[1.0x\ 10^{-9}]{100\%} HfO^+$

$Hf^+ + CS_2 \xrightarrow[1.2x\ 10^{-9}]{100\%} HfS^+$

Reported by the Center of Mass Spectrometry at York University (Toronto, Canada) in a SIFT apparatus
https://www.yorku.ca/dkbohme/research/selection_table.html

Formation of Radioactive Molecules in Ion Reaction Cell – Perspectives

- Cooling must be improved/ E_{cm} must be lowered (< 1 eV)
- Nearly 100% chemical conversion and transfer possible at E_{cm}= 0.1 eV
- Segmented RFQ a very versatile tool for beam diagnostic and reaction control
- Positive ion chemistry offers broader possibilities/ is easier to accomplish
- ISA for AMS a good test system

Ion Reaction Cell on RIB – TRIUMF/ Collaboration with Dr. C. Charles

Acknowledgements

- A. E. Lalonde AMS Laboratory – Dr. X. Zhao, N. St-Jean
- Isobarex Team – M. Yang, D. Valyaev, V. Titov, M. Omidvar

Thank You!

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