



ČVUT
ČESKÉ VYSOKÉ
UČENÍ TECHNICKÉ
V PRAZE

Electrochemistry in the superheavy element research

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Electrochemistry

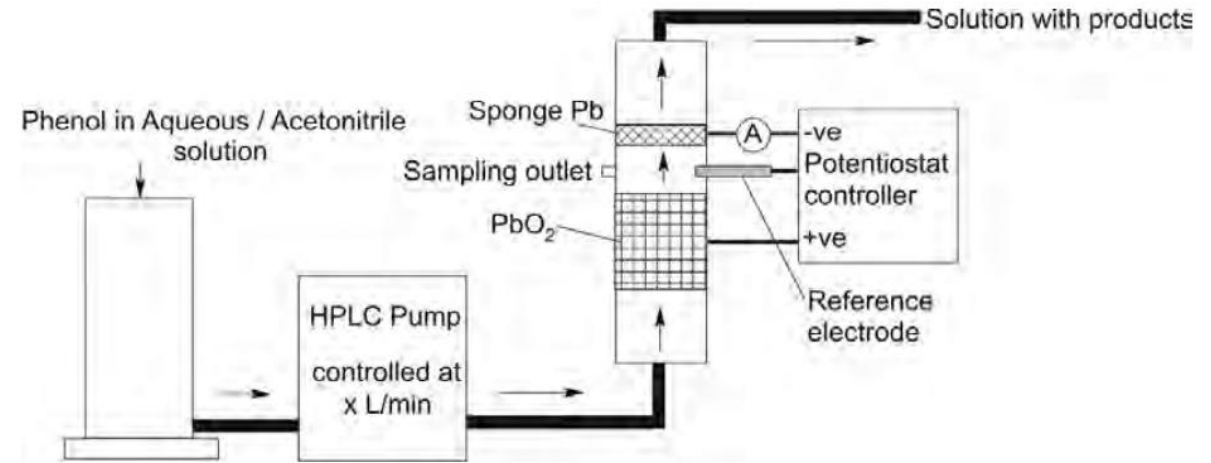


Fig. 1: Scheme of the flow-through electrochemical cell (Ferg, 2012)

- a wide range of different methods and procedures
- **adjusting ox. states in the flow apparatus**
 - mainly for the analytics of organics, not a separation
- *Czech electrochemistry tradition*



Electrochemistry and SHE

Why?

- SHE radiotracer – low conc., $T_{1/2} = \sim \text{ms} - \text{s}$
- oxidation state adjustment
 - without chemicals – **green chemistry**
 - chemical properties, characterization
 - further experiments, separation
- not very widespread in radiochemistry
- for transuraniums (**Toyoshima**, 2008)
 - Yb, Ce, Sr $\rightarrow$ No, Md
- electrodeposition, α -spectroscopy (2007 -)

Periodic Table of the Elements																		2																	
1 1A H Hydrogen 1.008		2 2A He Helium 4.003																13 3A B Boron 10.811		14 4A C Carbon 12.011		15 5A N Nitrogen 14.007		16 6A O Oxygen 15.999		17 7A F Fluorine 18.998		18 8A Ne Neon 20.180							
3 Li Lithium 6.941		4 Be Beryllium 9.012																5 Al Aluminum 26.982		6 Si Silicon 28.086		7 P Phosphorus 30.974		8 S Sulfur 32.066		9 Cl Chlorine 35.453		10 Ar Argon 39.948							
11 Na Sodium 22.990		12 Mg Magnesium 24.305		3 III B Boron 10.811		4 IV C Carbon 12.011		5 V N Nitrogen 14.007		6 VI O Oxygen 15.999		7 VII F Fluorine 18.998		8 VIII Ne Neon 20.180		9 IX Na Sodium 22.990		10 X Mg Magnesium 24.305		11 XI Al Aluminum 26.982		12 XII Si Silicon 28.086		13 XIII P Phosphorus 30.974		14 XIV S Sulfur 32.066		15 XV Cl Chlorine 35.453		16 XVI Ar Argon 39.948					
19 K Potassium 39.098		20 Ca Calcium 40.078		21 Sc Scandium 44.956		22 Ti Titanium 47.867		23 V Vanadium 50.942		24 Cr Chromium 51.996		25 Mn Manganese 54.938		26 Fe Iron 55.845		27 Co Cobalt 58.933		28 Ni Nickel 58.693		29 Cu Copper 63.546		30 Zn Zinc 65.38		31 Ga Gallium 69.723		32 Ge Germanium 72.631		33 As Arsenic 74.922		34 Se Selenium 78.971		35 Br Bromine 79.904		36 Kr Krypton 83.798	
37 Rb Rubidium 85.468		38 Sr Strontium 87.62		39 Y Yttrium 88.906		40 Zr Zirconium 91.224		41 Nb Niobium 92.906		42 Mo Molybdenum 95.94		43 Tc Technetium 98.906		44 Ru Ruthenium 101.07		45 Rh Rhodium 102.905		46 Pd Palladium 106.42		47 Ag Silver 107.868		48 Cd Cadmium 112.414		49 In Indium 114.818		50 Sn Tin 118.710		51 Sb Antimony 121.757		52 Te Tellurium 127.6		53 I Iodine 126.905		54 Xe Xenon 131.294	
55 Cs Cesium 132.905		56 Ba Barium 137.327		57-71 Lanthanide Series		72 Hf Hafnium 178.49		73 Ta Tantalum 180.948		74 W Tungsten 183.84		75 Re Rhenium 186.207		76 Os Osmium 190.23		77 Ir Iridium 192.222		78 Pt Platinum 195.084		79 Au Gold 196.967		80 Hg Mercury 200.592		81 Tl Thallium 204.383		82 Pb Lead 207.2		83 Bi Bismuth 208.980		84 Po Polonium [209]		85 At Astatine [210]		86 Rn Radon [222]	
87 Fr Francium [223]		88 Ra Radium [226]		89-103 Actinide Series		104 Rf Rutherfordium [261]		105 Db Dubnium [262]		106 Sg Seaborgium [266]		107 Bh Bohrium [264]		108 Hs Hassium [265]		109 Mt Meitnerium [268]		110 Ds Darmstadtium [271]		111 Rg Roentgenium [272]		112 Cn Copernicium [285]		113 Nh Nihonium [284]		114 Fl Flerovium [289]		115 Mc Moscovium [288]		116 Lv Livermorium [293]		117 Ts Tennessine [294]		118 Og Oganesson [294]	
				57 La Lanthanum 138.905		58 Ce Cerium 140.116		59 Pr Praseodymium 140.908		60 Nd Neodymium 144.242		61 Pm Promethium [145]		62 Sm Samarium 150.36		63 Eu Europium 151.964		64 Gd Gadolinium 157.25		65 Tb Terbium 158.925		66 Dy Dysprosium 162.500		67 Ho Holmium 164.930		68 Er Erbium 167.259		69 Tm Thulium 168.934		70 Yb Ytterbium 173.055		71 Lu Lutetium 174.967			
				89 Ac Actinium [227]		90 Th Thorium 232.038		91 Pa Protactinium 231.036		92 U Uranium 238.029		93 Np Neptunium 237.048		94 Pu Plutonium 244.064		95 Am Americium 243.061		96 Cm Curium 247.070		97 Bk Berkelium 247.070		98 Cf Californium 251.080		99 Es Einsteinium [252]		100 Fm Fermium 257.095		101 Md Mendelevium 258.1		102 No Nobelium 259.101		103 Lr Lawrencium [262]			

Fig. 2: Periodic table with SHE highlighted

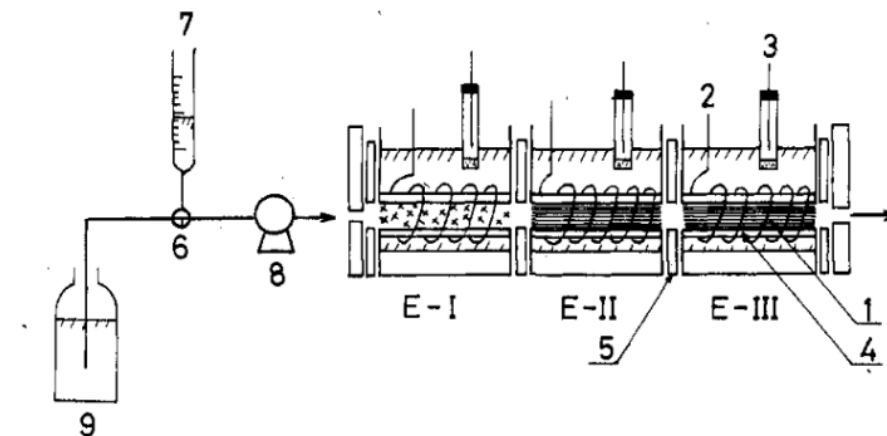
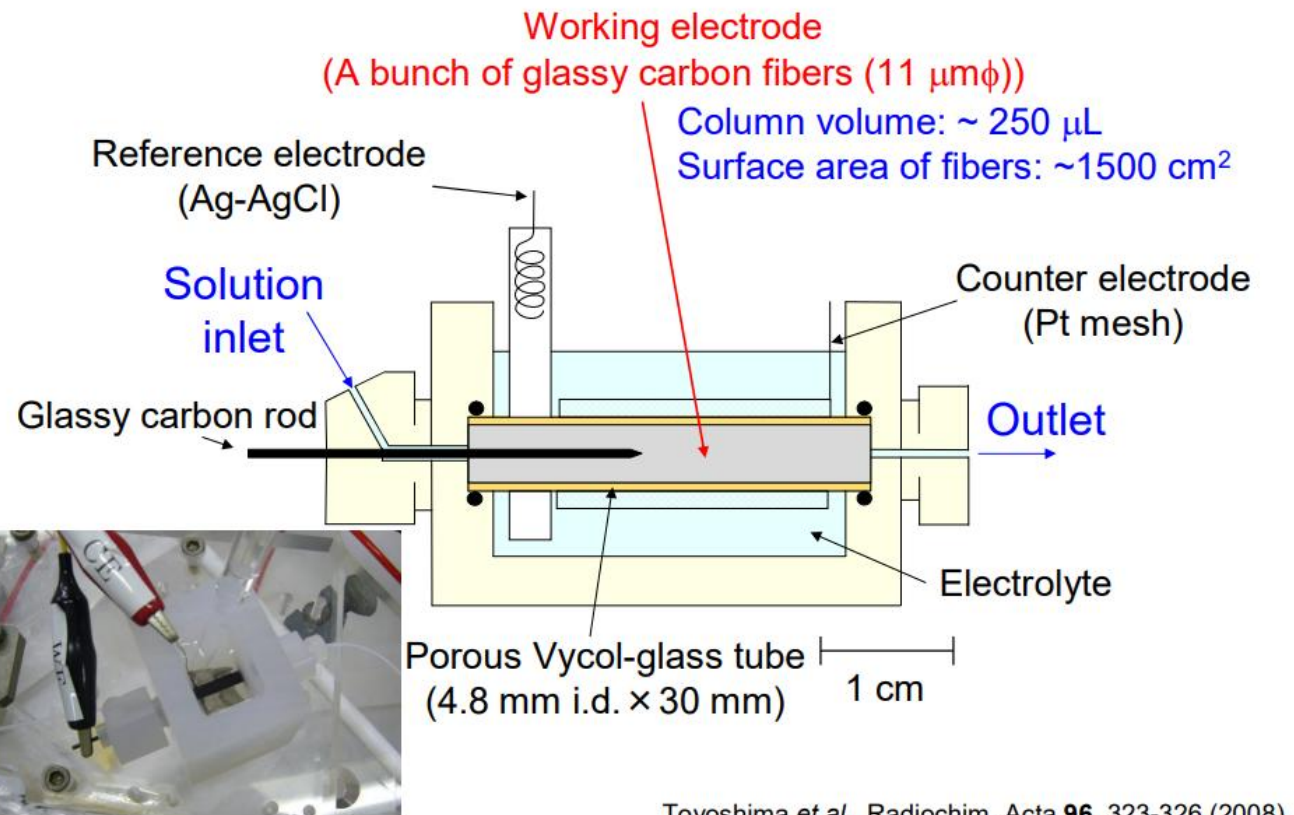


Fig. 3: Elchem cell for uranium analysis (Hideyo, 1986)



Toyoshima *et al.*, *Radiochim. Acta* **96**, 323-326 (2008).

Fig. 4: electrochemistry apparatus made by (Toyoshima, 2008)

Elution behavior of $^{81}\text{Sr}^{2+}$ and $^{162}\text{Yb}^{3+}$

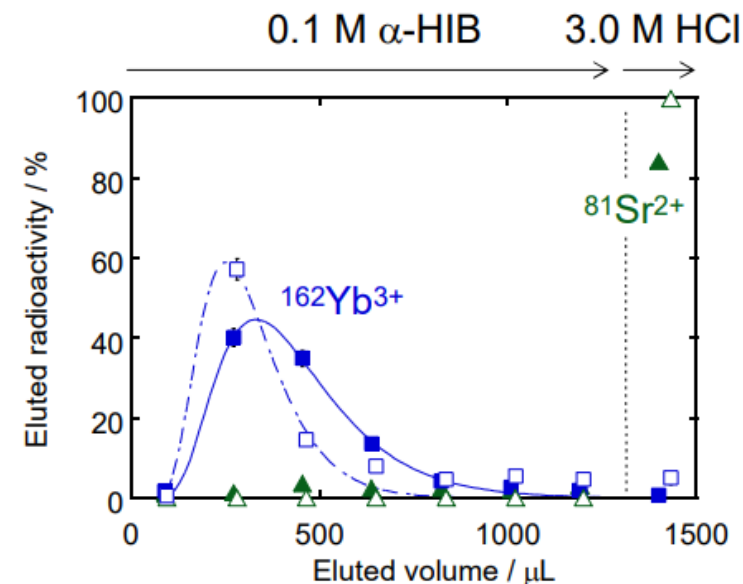


Fig. 5: Results of chromatography by (Toyoshima, 2009)

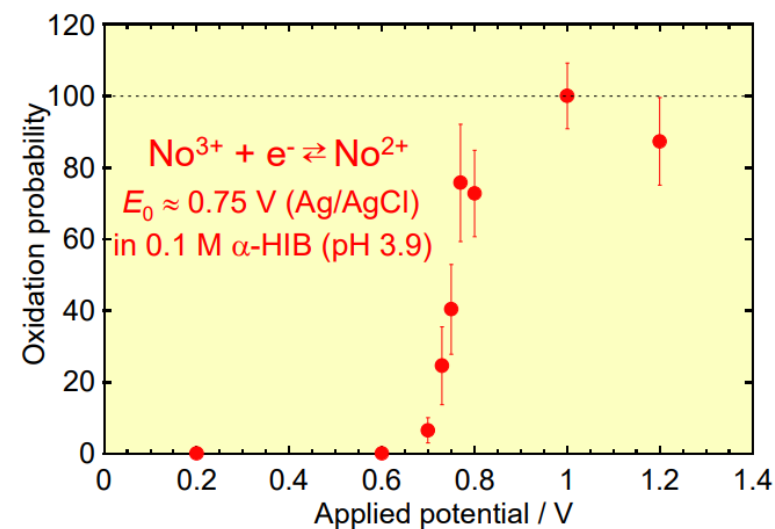


Fig. 6: Oxidation probability by (Toyoshima, 2009)



Basic assumptions

- **SHE oxidation state adjustment**
- **one-atom-at-a-time** mode, fast
- minimalization of sorption and electrodeposition
- compatibility with other parts of the CTU apparatus
- **flow arrangement**
- **three-electrode system**

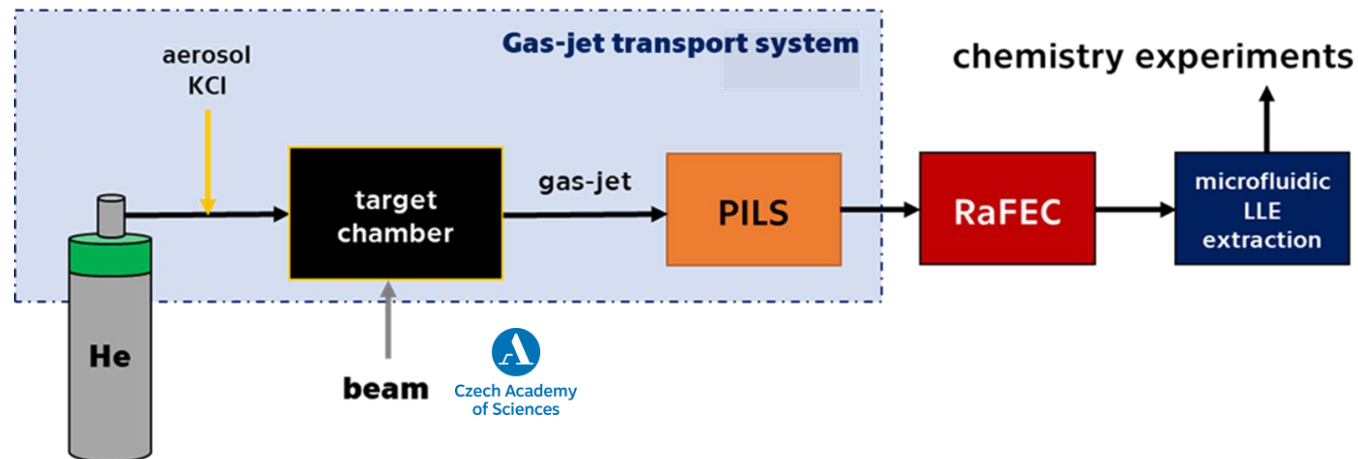


Fig. 7: Scheme of the whole FNSPE experimental apparatus



RaFEC

Rapid Flow Electrochemical Cell

Generation 0

Generation 1

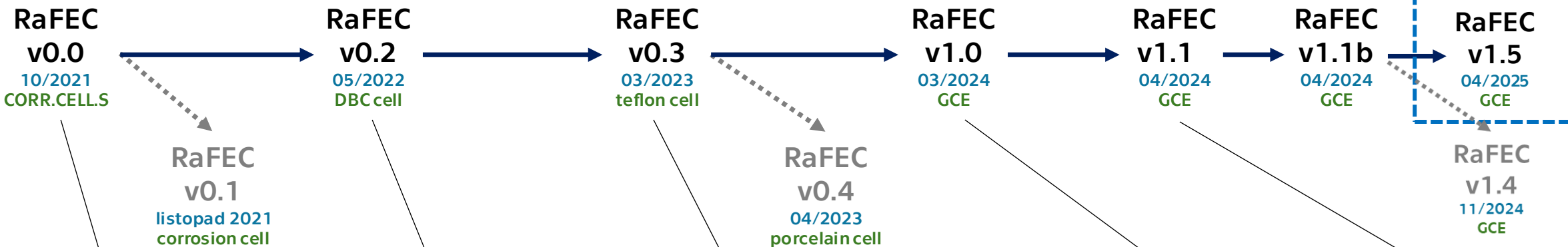


Fig. 8a



Fig. 8b



Fig. 8c

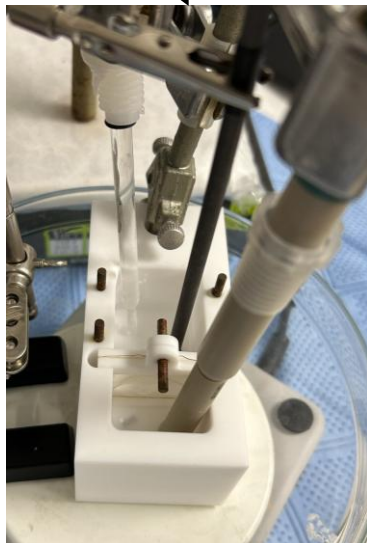


Fig. 8d

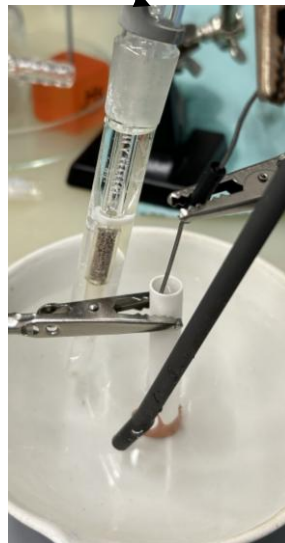


Fig. 8e



Fig. 8f





192Tl 9.6 min $\epsilon = 100.00\%$	193Tl 21.6 min $\epsilon = 100.00\%$	194Tl 33.0 min $\epsilon = 100.00\%$ $\alpha < 1.0\text{E-}7\%$	195Tl 1.16 h $\epsilon = 100.00\%$	196Tl 1.84 h $\epsilon = 100.00\%$	197Tl 2.84 h $\epsilon = 100.00\%$
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Fig. 9: possible Tl radioisotopes, source [1]

[1] NuDat 3.0: Search and plot nuclear structure and decay data interactively. Upton: National Nuclear Data Center (NNDC) at Brookhaven National Laboratory, 2021. web: <https://www.nndc.bnl.gov/nudat3/>.

^{201}Tl

electrochemistry cell

RCPC, ASV, chronoamperometry, chronocoulometry, CV

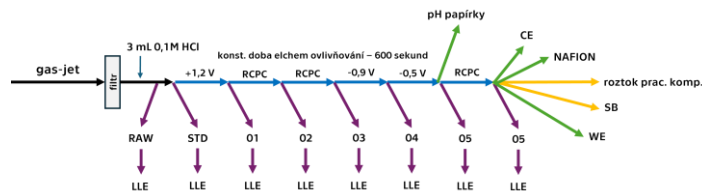
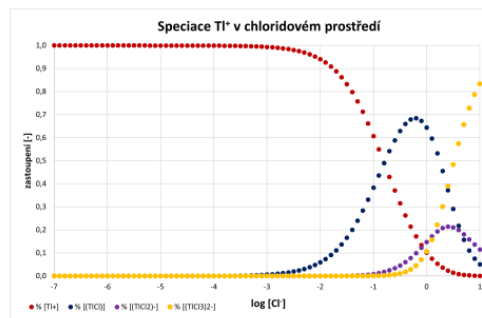
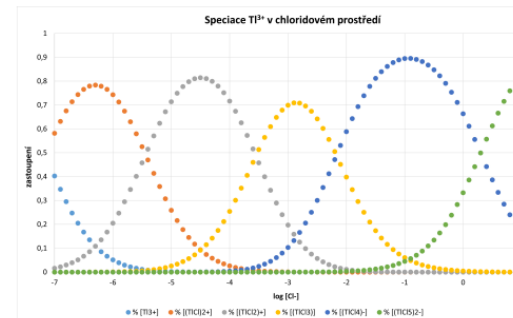


Fig. 11a+b: Tl speciation



(a) Pro Tl^{+I}



(b) Pro Tl^{+III}

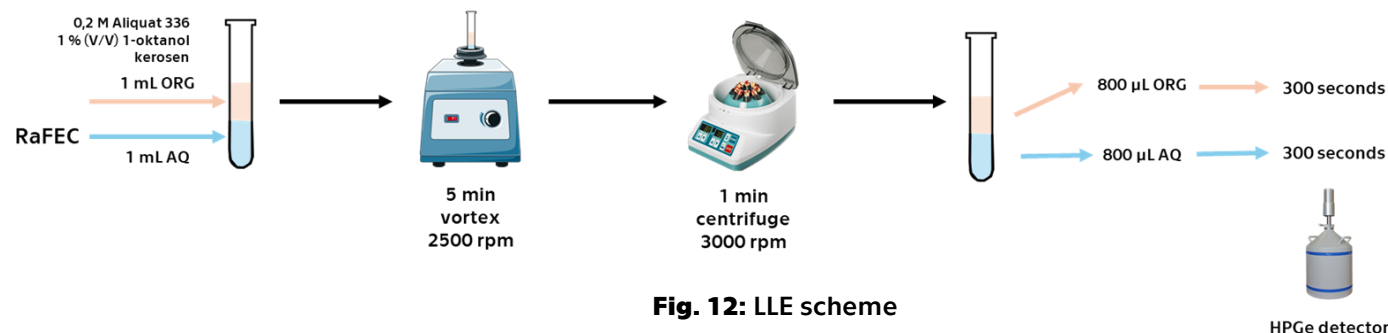
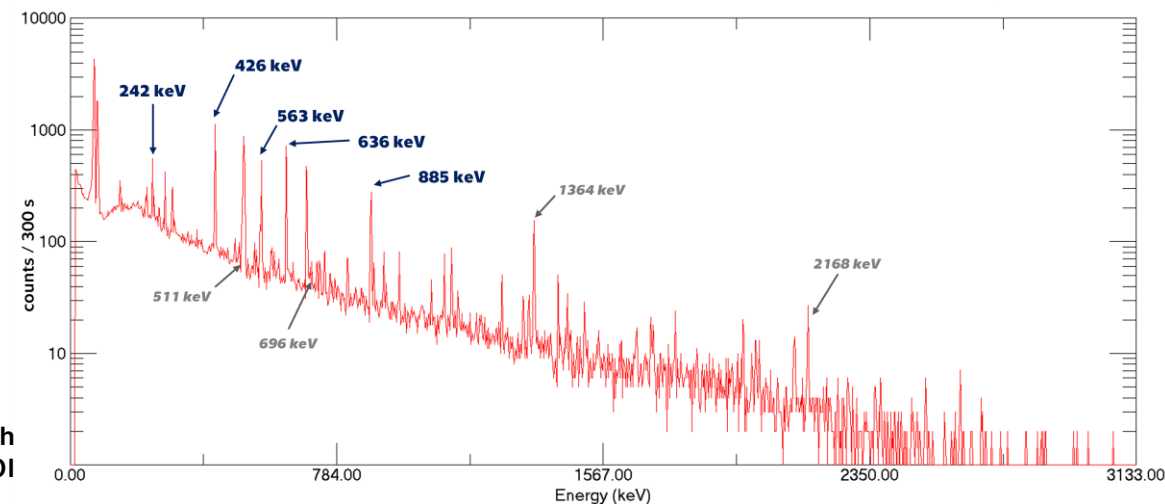


Fig. 12: LLE scheme

0,2M Aliquat 336
kerosene
1 m/m % 1-octanol

Fig. 10: sample experiment scheme

Fig. 13: sample spectrum with marked ROI



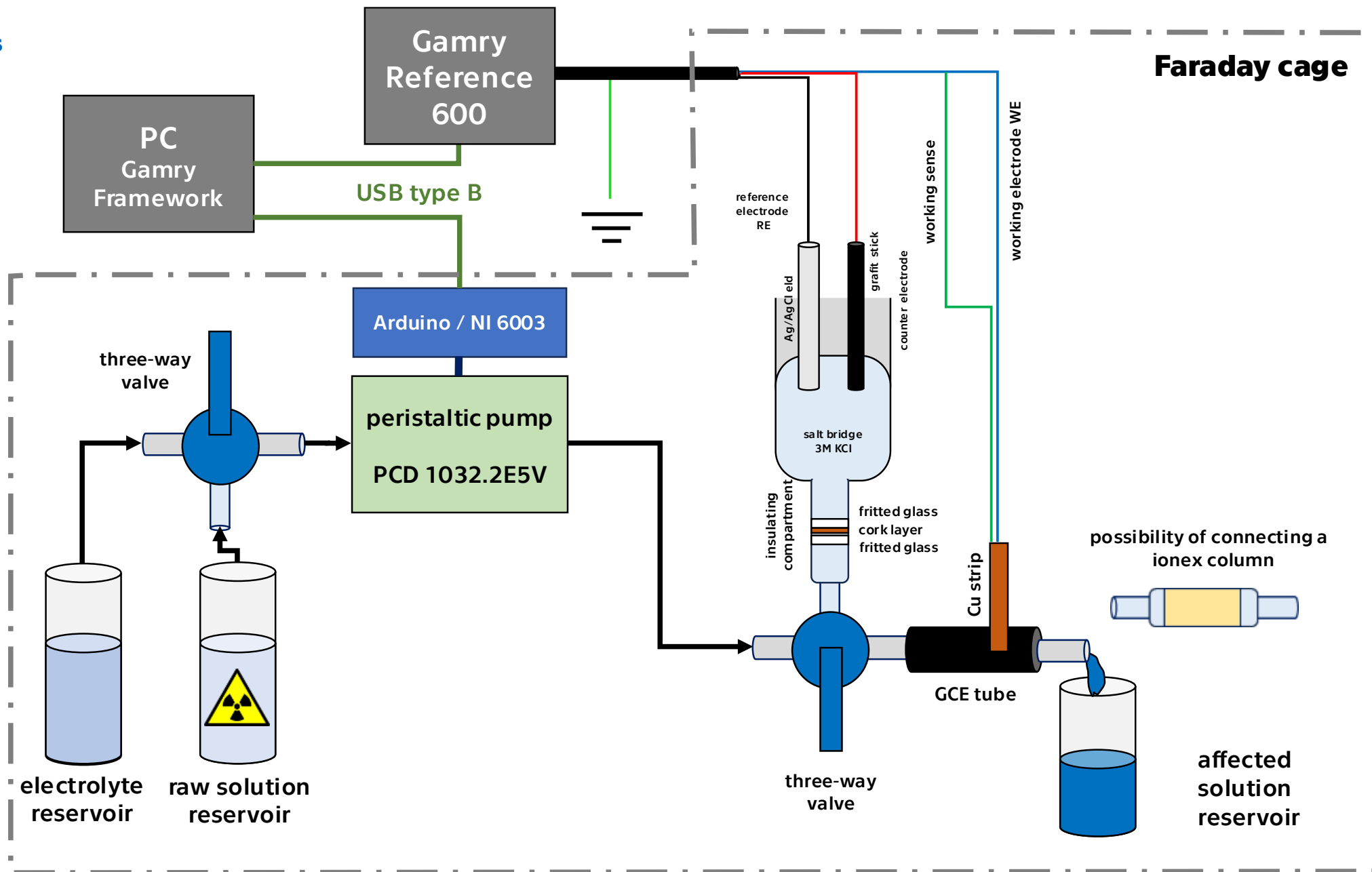


Fig. 14: Scheme of RaFEC v1.5 apparatus

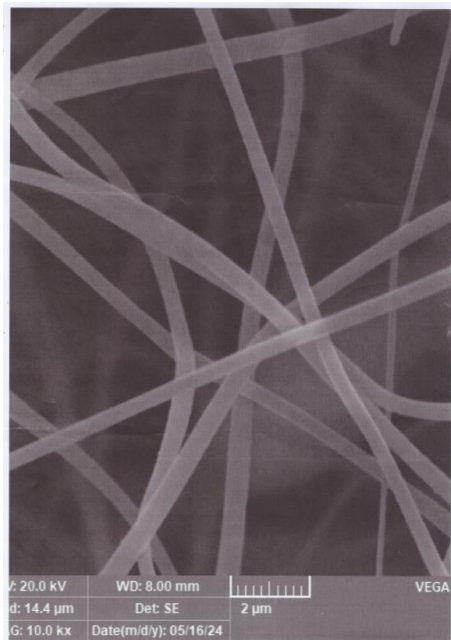


Fig. 16b: VYCOR glass
(UCT Prague)



Fig. 16a: Detail of the apparatus –
compartment of isolation

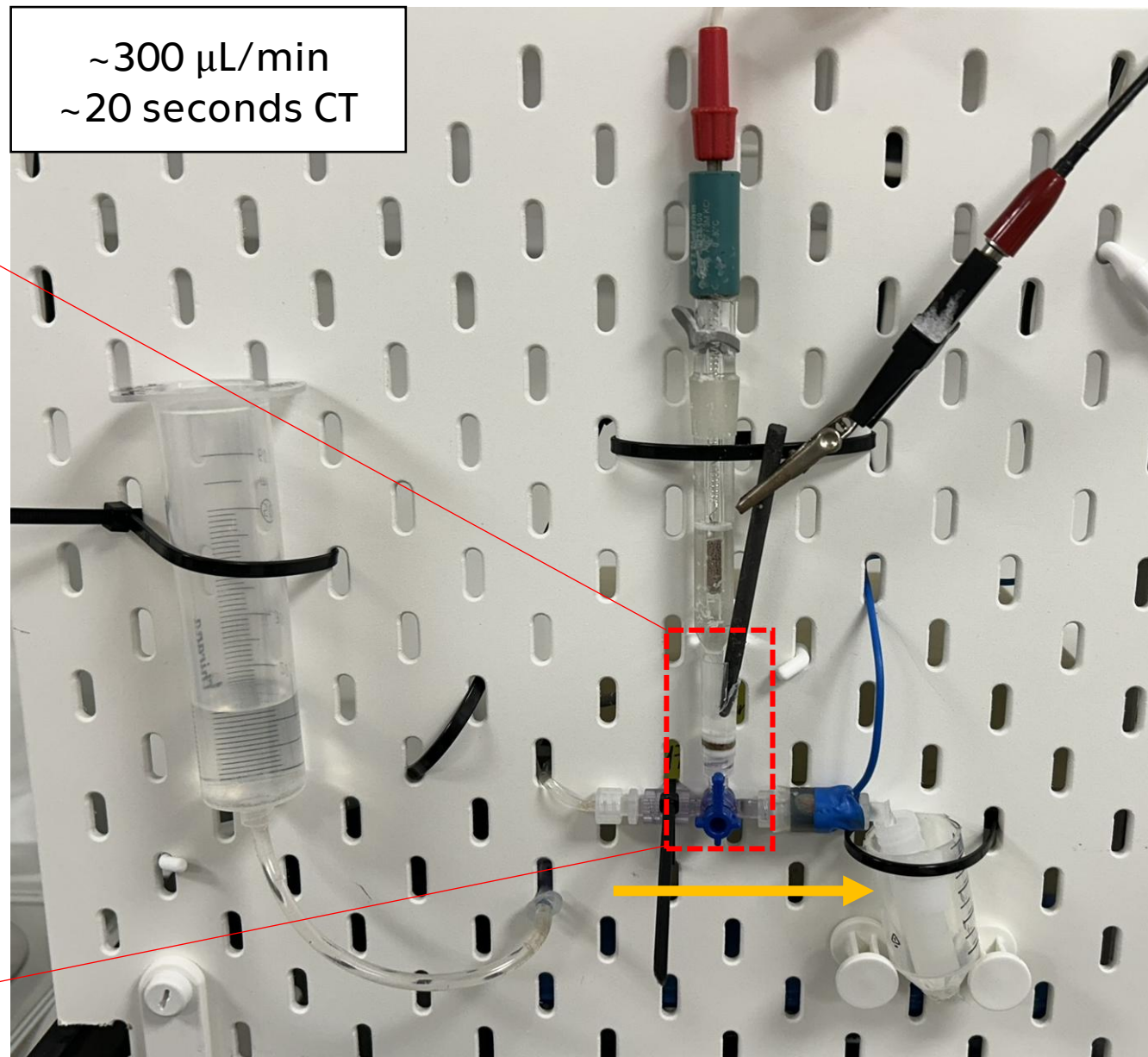


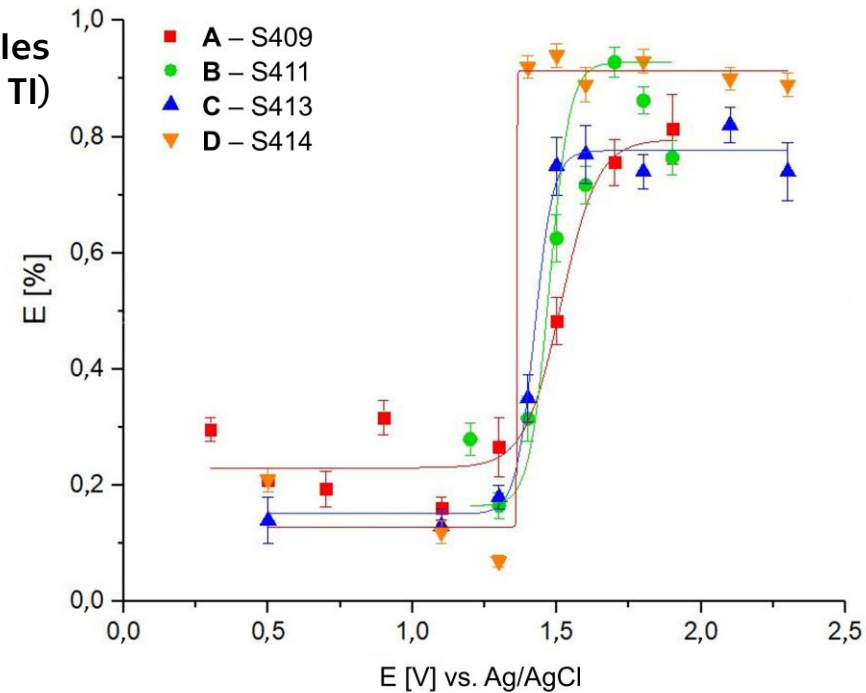
Fig. 15: Picture of RaFEC v1.2 apparatus (04/2025)



Results

- long way to optimization
- expected behavior in flow arrangement for cyclotron TI
- problems with ^{201}TI ($T_{1/2} = 3.04 \text{ d}$)
- significant sorption onto surfaces
- low activity for sufficient accuracy

Fig. 17a: E for individual samples and potentials (cyclotron TI)



$$y = \frac{A_1 - A_2}{1 + e^{(x-x_0)/dx}} + A_2$$

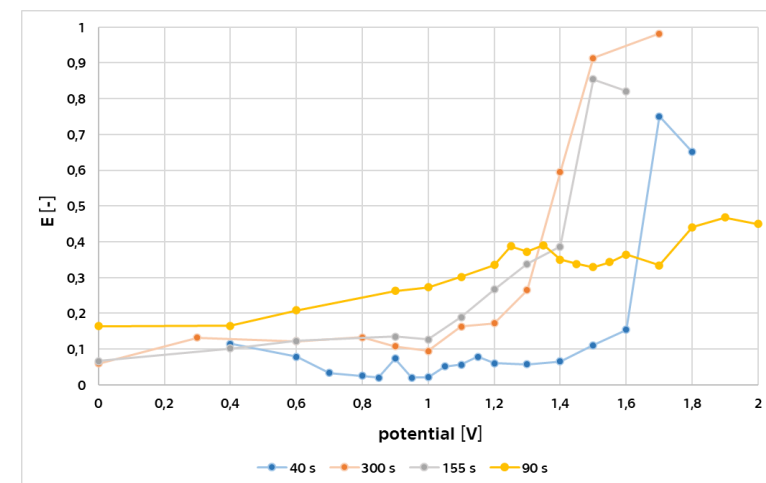


Fig. 17b: E for individual samples and potentials (TI-201)

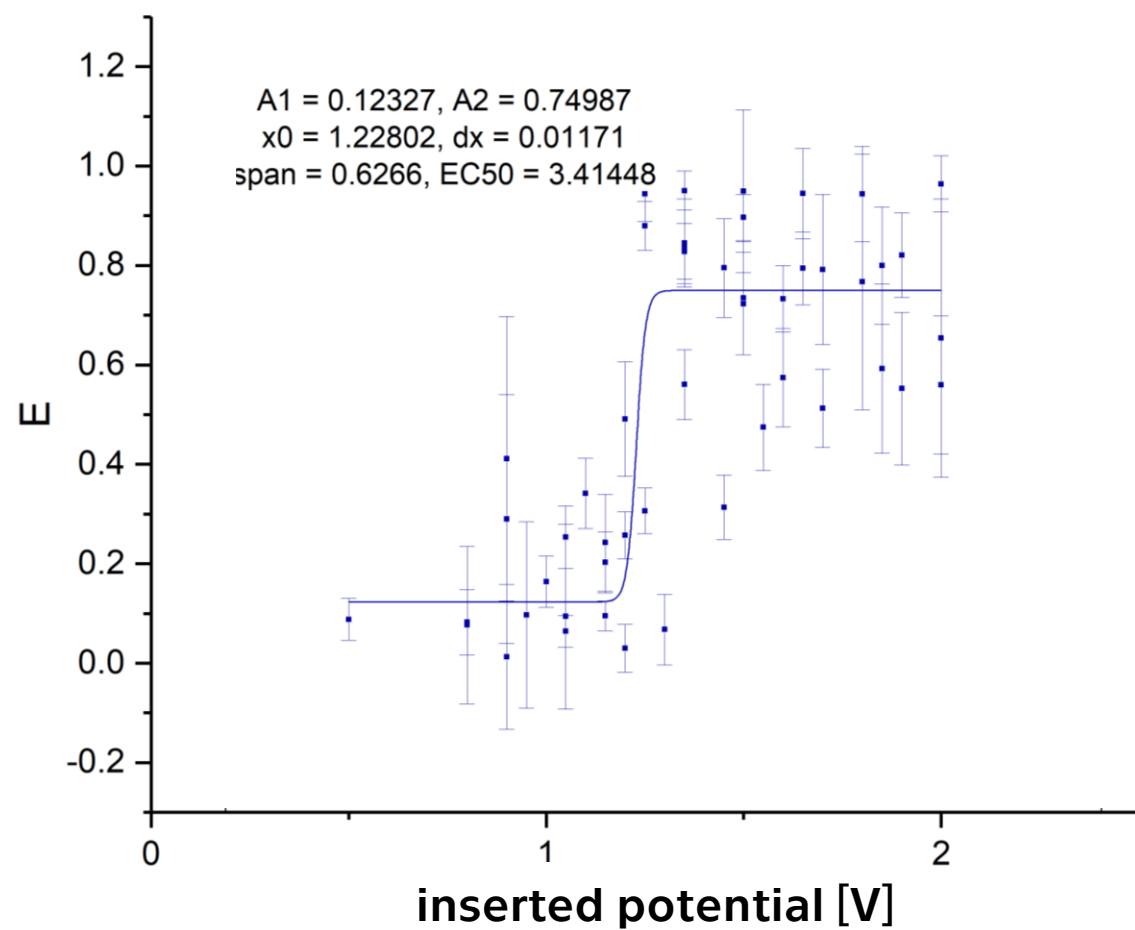
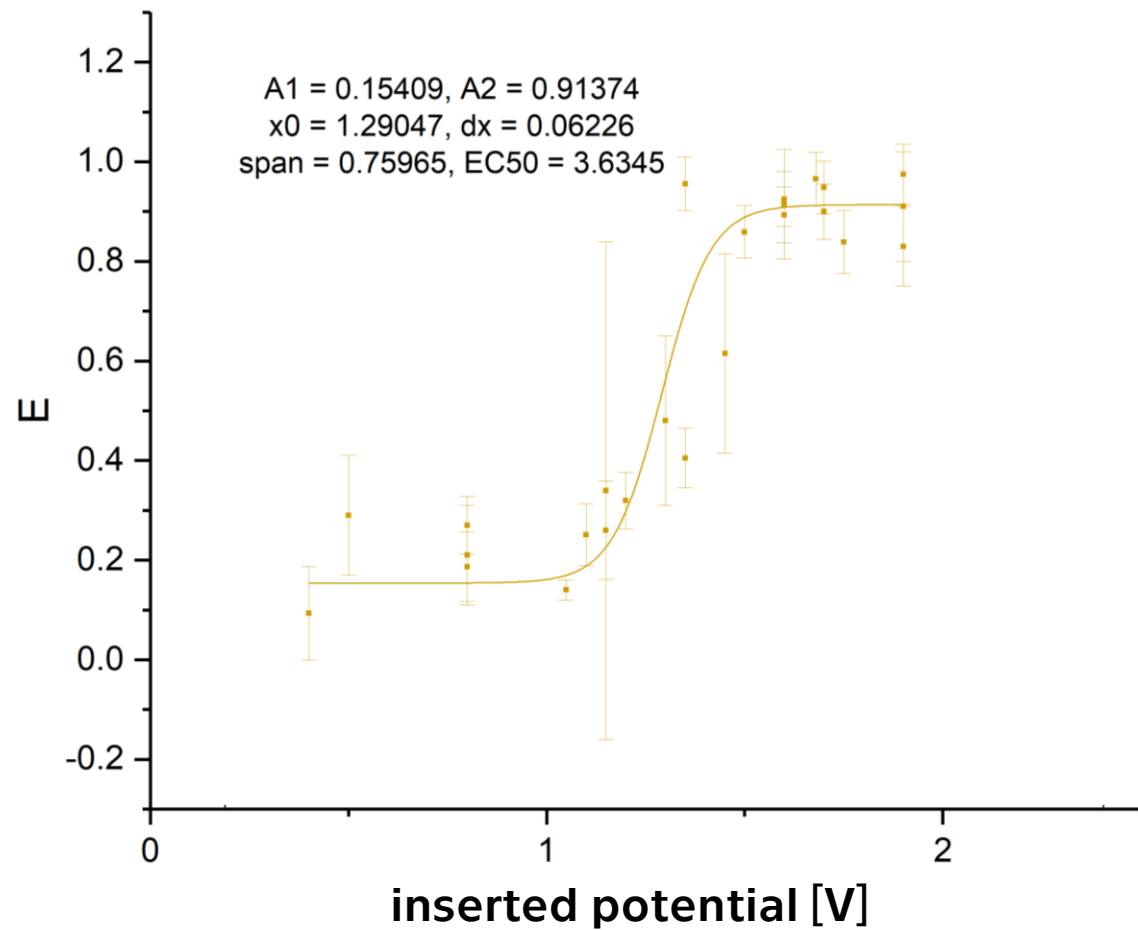


Fig. 18a+b: E for individual samples and potentials

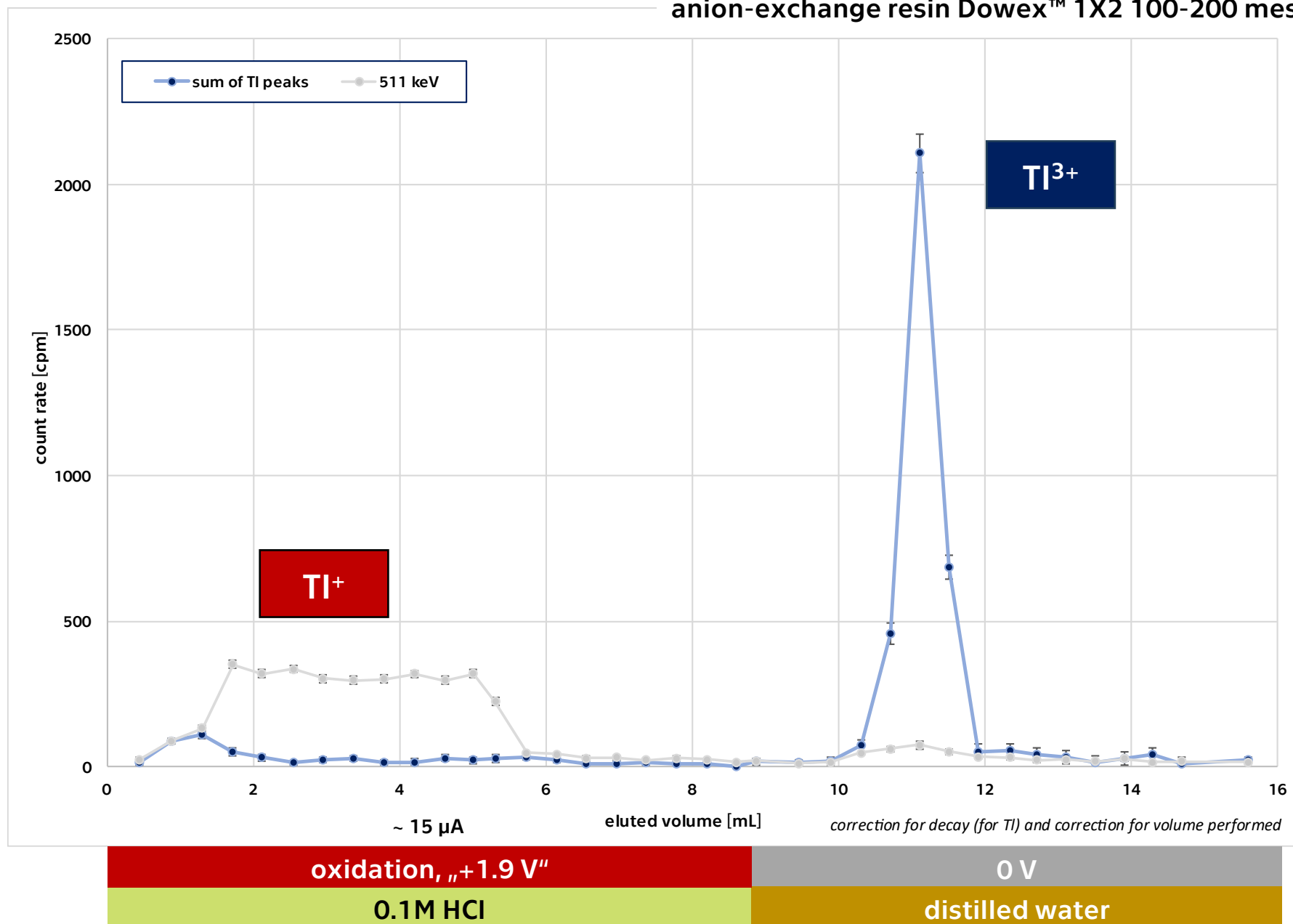


Fig. 19: Results of chromatography with anex



Conclusion

- **electrochemistry apparatus RaFEC v1.2**
- capable of adjusting the oxidation state of Tl (cyclotron) radiotracers
- ^{201}Tl : further research needed



Further research

- optimization
- miniaturization
- isolation compartment
- sorption
- other elements
 - Ce, V-Db, Hg-Cn, Au-Rg
- automatization

Period	1	Periodic Table 1-172																18	Orbitals	
1	1 H	2											13	14	15	16	17	2 He	1s	
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne	2s2p	
3	11 Na	12 Mg	3	4	5	6	7	8	9	10	11	12	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar	3s3p	
4	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	4s3d4p	
5	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	5s4d5p	
6	55 Cs	56 Ba	57-71	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	6s5d6p	
7	87 Fr	88 Ra	89-103	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og	7s6d7p	
8	119	120	121-	156	157	158	159	160	161	162	163	164	139	140	169	170	171	172	8s7d8p	
9	165	166												167	168					9s9p
6	57 La	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	4f				
7	89 Ac	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	5f				
8	141	142	143	144	145	146	147	148	149	150	151	152	153	154	155	6f				
8	121	122	123	124	125	126	127	128	129	130	131	132	133	134	135	136	137	138	5g	

Fig. 20: Periodic table (Pyykkö), source: [2], changed



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