Electrochemistry in the superheavy element research

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Electrochemistry

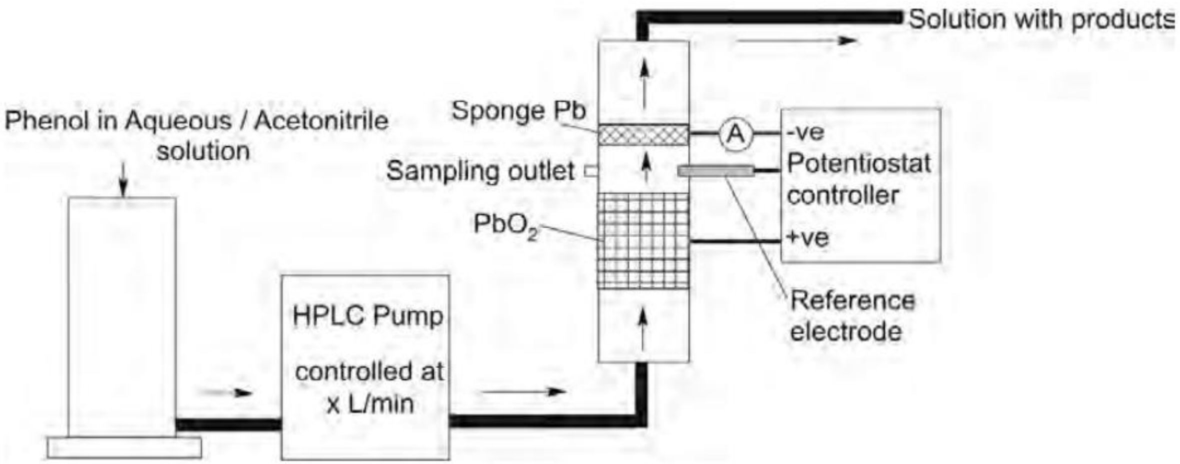

Fig. 1: Scheme of the flow-through electrochemical cell (Ferg, 2012)

- 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*

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Electrochemistry and SHE

Why?

- SHE radiotrace – low conc., $T_{1/2}$ = ~ms–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 → No, Md
- electrodeposition, α-spectroscopy (2007 -)

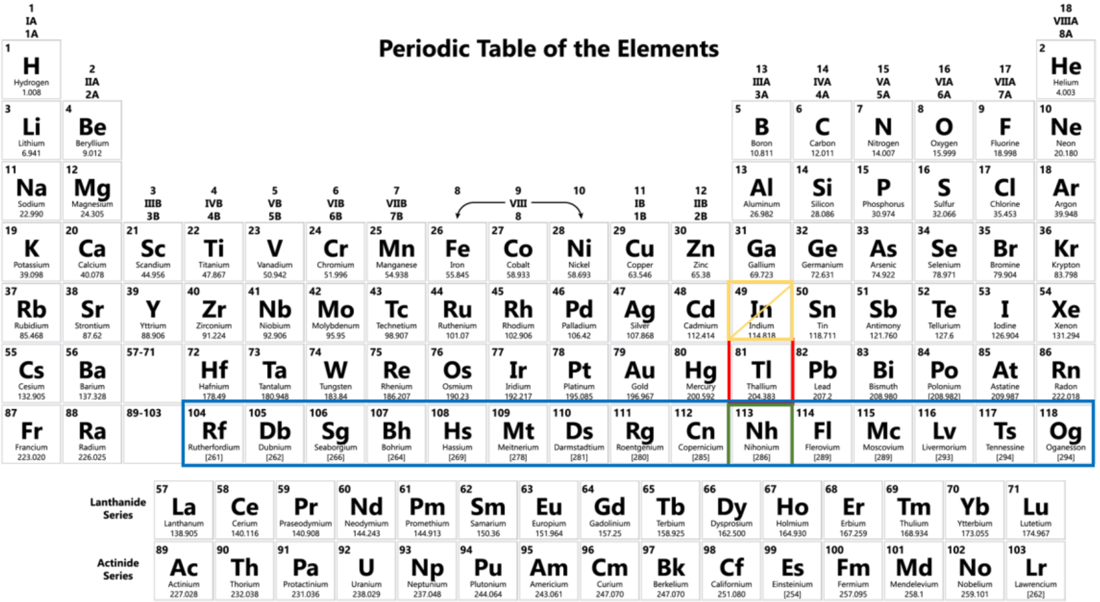

Fig. 2: Periodic table with SHE highlighted

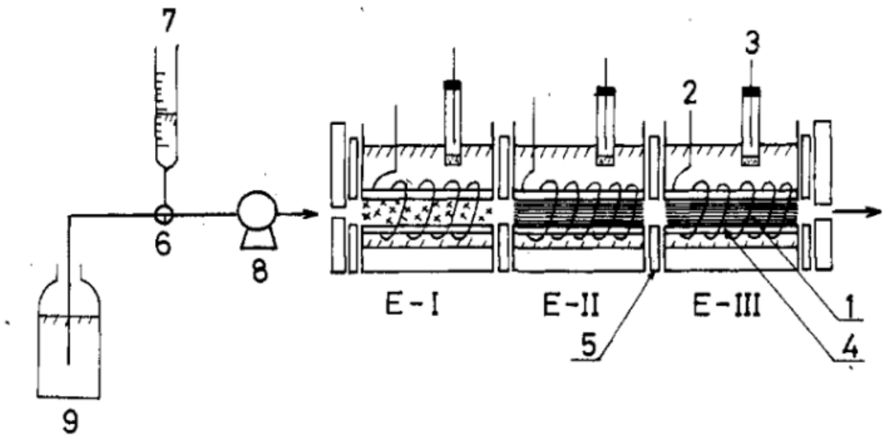

Fig. 3: Elchem cell for uranium analysis (Hideyo, 1986)

Source: Hideyo Takeishi et. al, Analytical Chemistry 1986 58 (2), 458-462, DOI: 10.1021/ac00293a043

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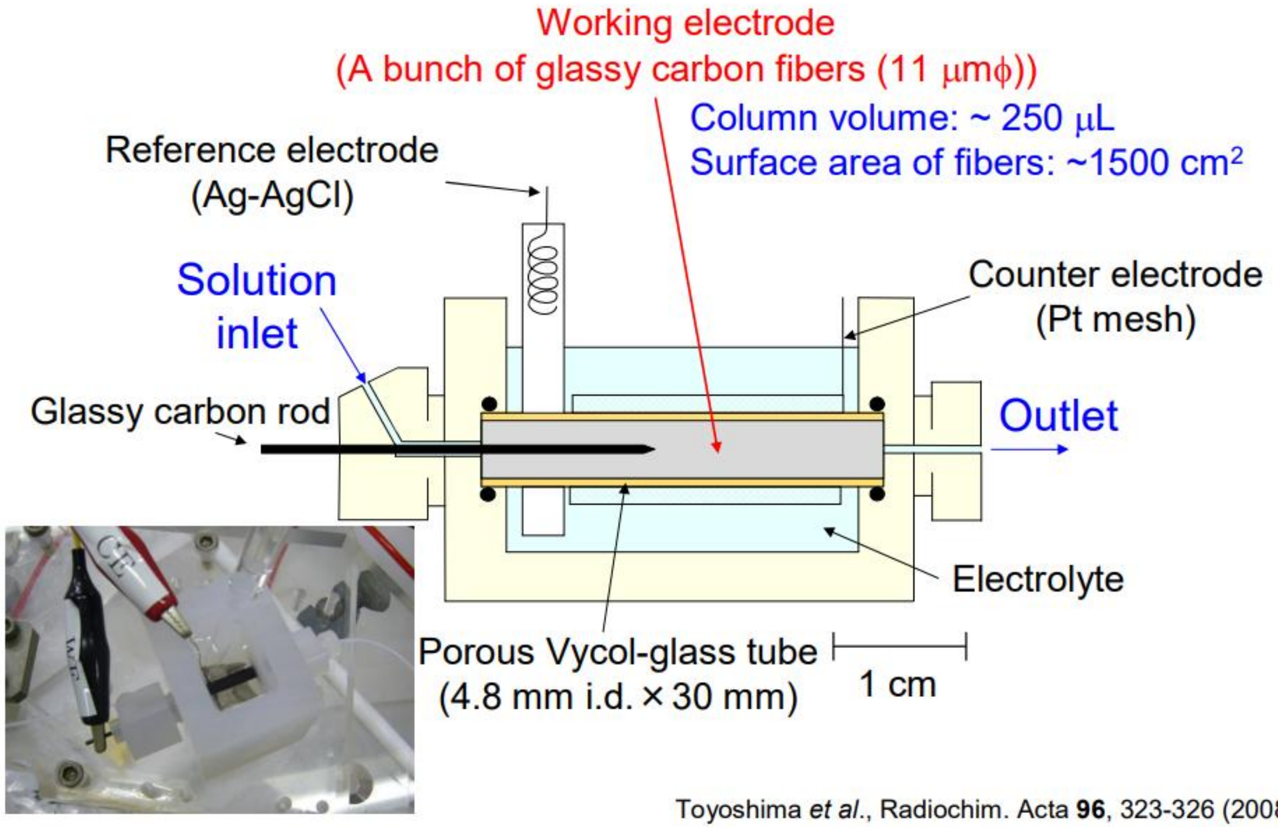

Toyoshima *et al.*, Radiochim. Acta **96**, 323-326 (2008).

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

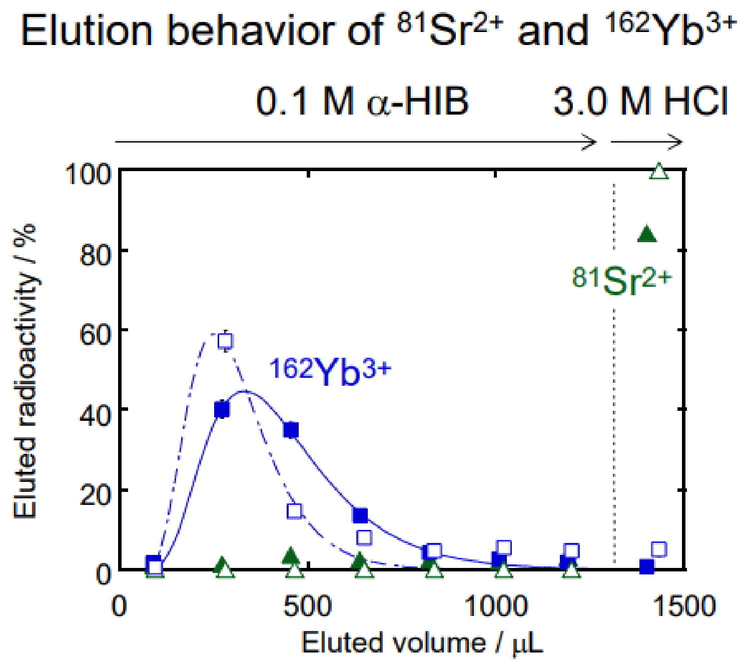

Fig. 5: Results of chromatography by (Toyoshima, 2009)

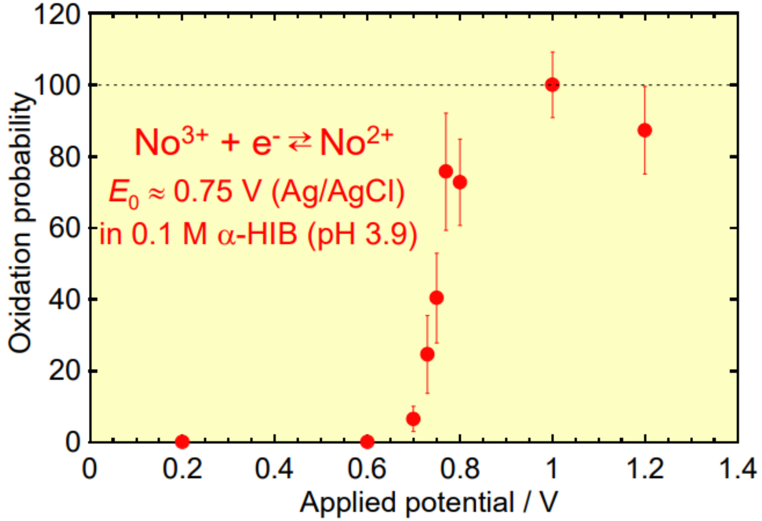

Fig. 6: Oxidation probability by (Toyoshima, 2009)

Source of the figures: Toyoshima et al.,. Radiochim. Acta. 2008-6-1, 96, Nr. 6, p. 323–326. doi: 10.1524/ract.2008.1498; Toyoshima et al., J. Am. Chem. Soc. 131, 9180-9181 (2009).

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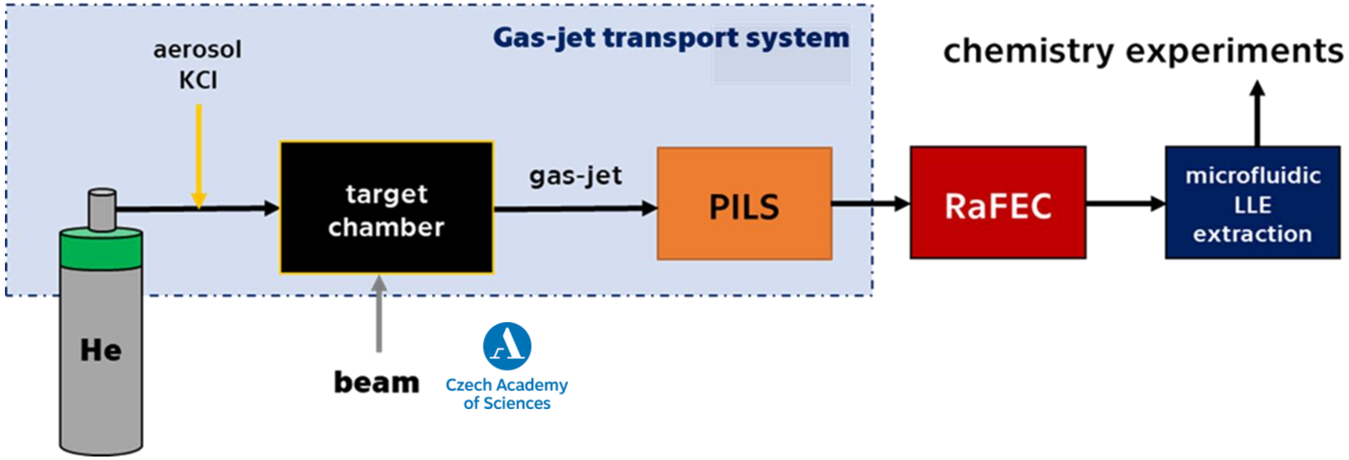

Fig. 7: Scheme of the whole FNSPE experimental apparatus

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**

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RaFEC

Rapid Flow Electrochemical Cell

Generation 0	Generation 1

RaFEC v0.0 — 10/2021 — CORR.CELL.S → **RaFEC v0.2** — 05/2022 — DBC cell → **RaFEC v0.3** — 03/2023 — teflon cell → **RaFEC v1.0** — 03/2024 — GCE → **RaFEC v1.1** — 04/2024 — GCE → **RaFEC v1.1b** — 04/2024 — GCE → **RaFEC v1.5** — 04/2025 — GCE

RaFEC v0.1 — listopad 2021 — corrosion cell

RaFEC v0.4 — 04/2023 — porcelain cell

RaFEC v1.4 — 11/2024 — GCE

Fig. 8a

Fig. 8b

Fig. 8c

Fig. 8d

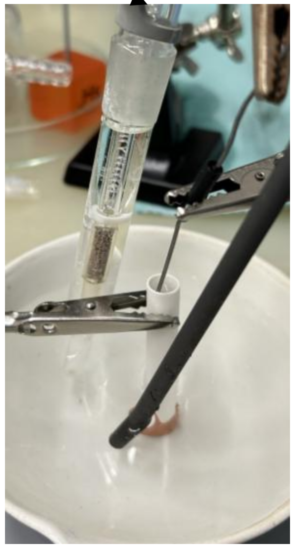

Fig. 8e

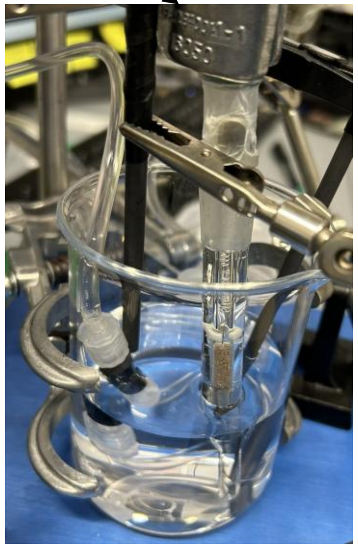

Fig. 8f

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^{197}Au (^{3}He, xn) Y

192Tl 9.6 min ε = 100.00%	193Tl 21.6 min ε = 100.00%	194Tl 33.0 min ε = 100.00% α < 1.0E-7%	195Tl 1.16 h ε = 100.00%	196Tl 1.84 h ε = 100.00%	197Tl 2.84 h ε = 100.00%

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

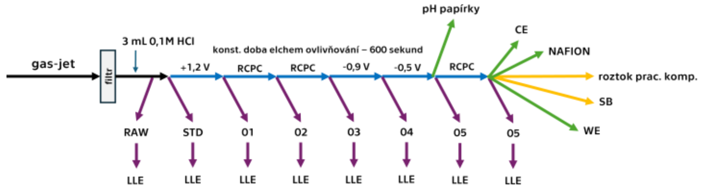
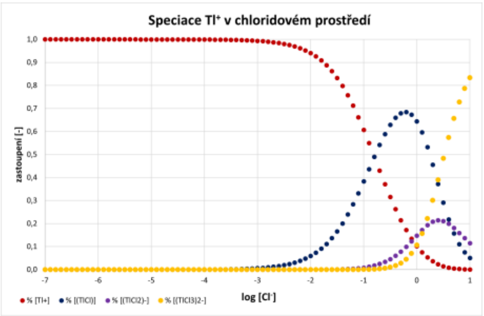


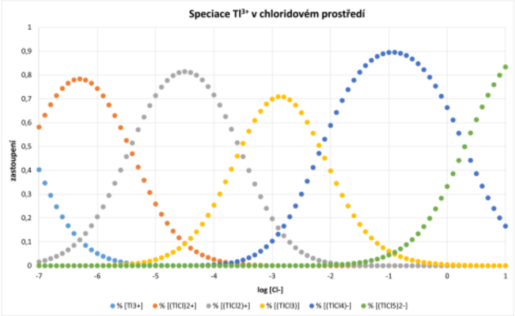
Fig. 10: sample experiment scheme

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

Fig. 11a+b: Tl speciation



(a) Pro Tl^{+I}



(b) Pro Tl^{+III}

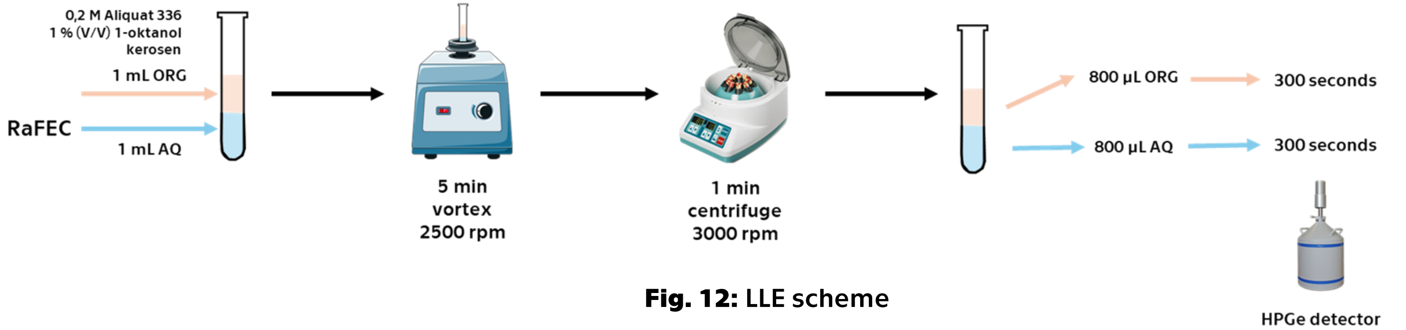


Fig. 12: LLE scheme

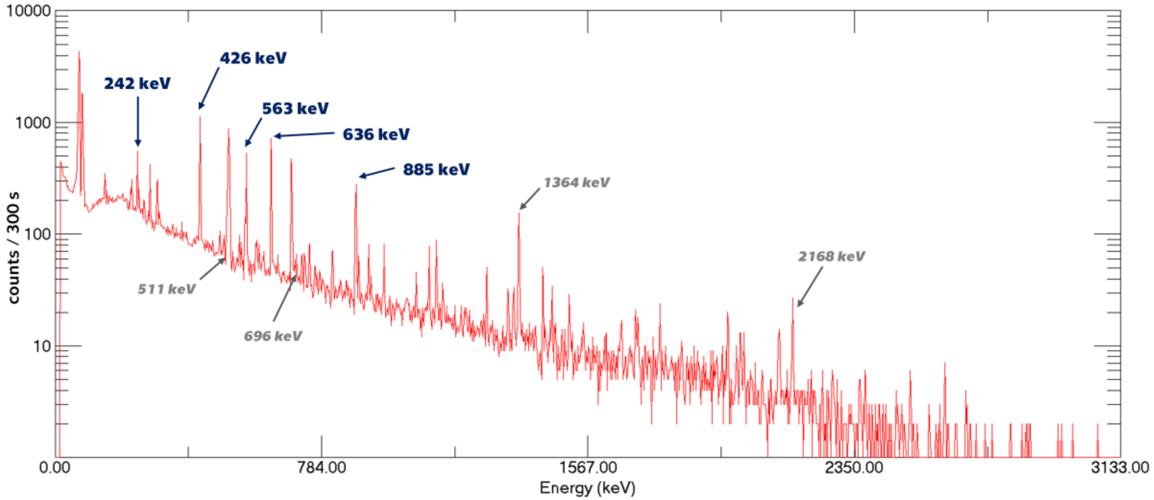


Fig. 13: sample spectrum with marked ROI

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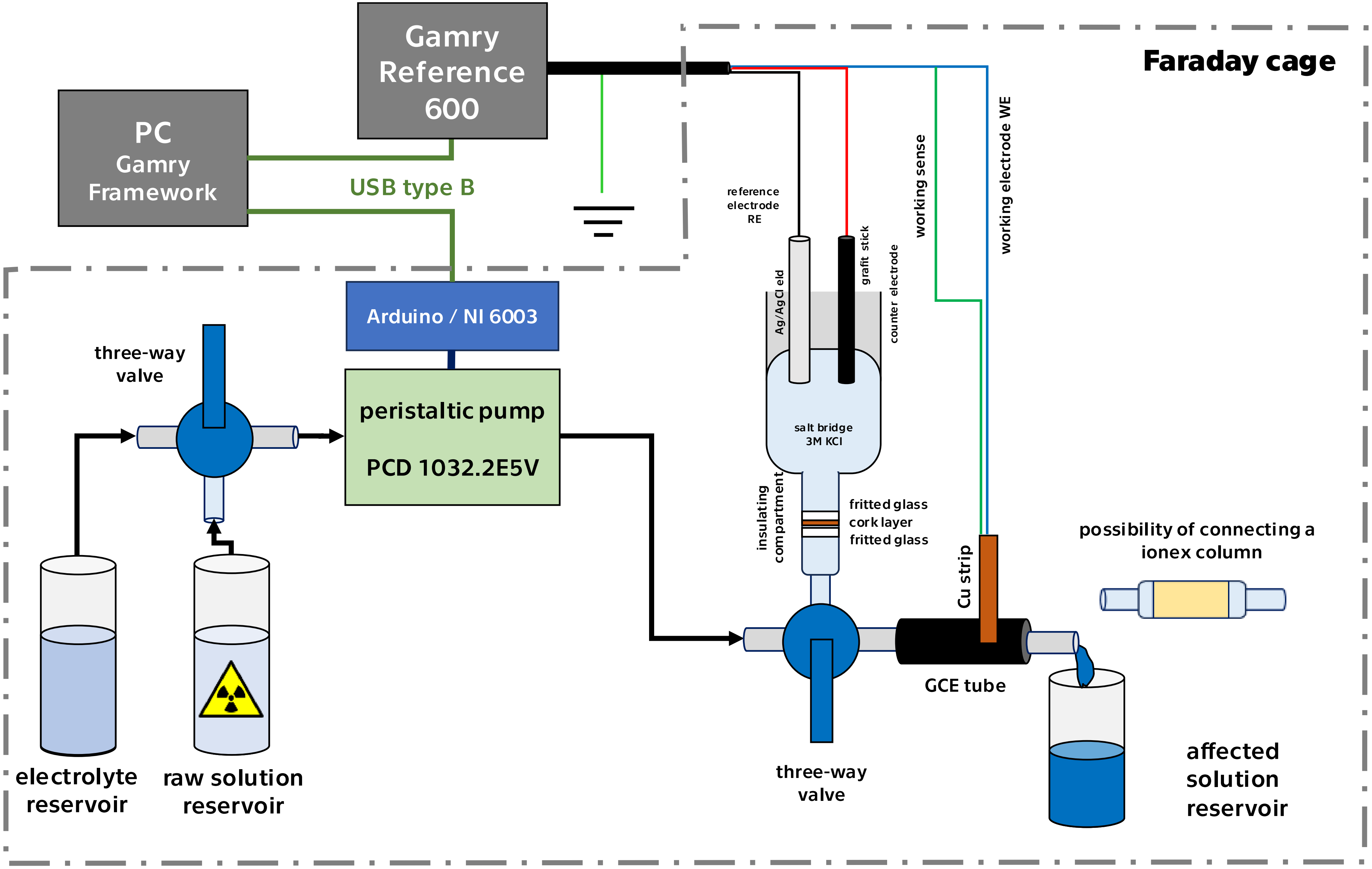

Fig. 14: Scheme of RaFEC v1.5 apparatus

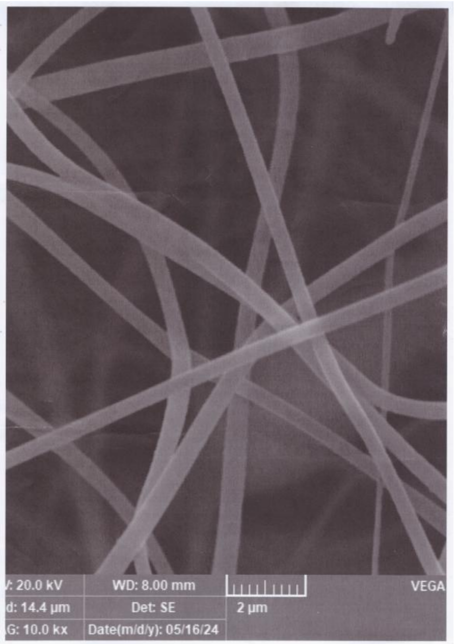

Fig. 16b: VYCOR glass (UCT Prague)

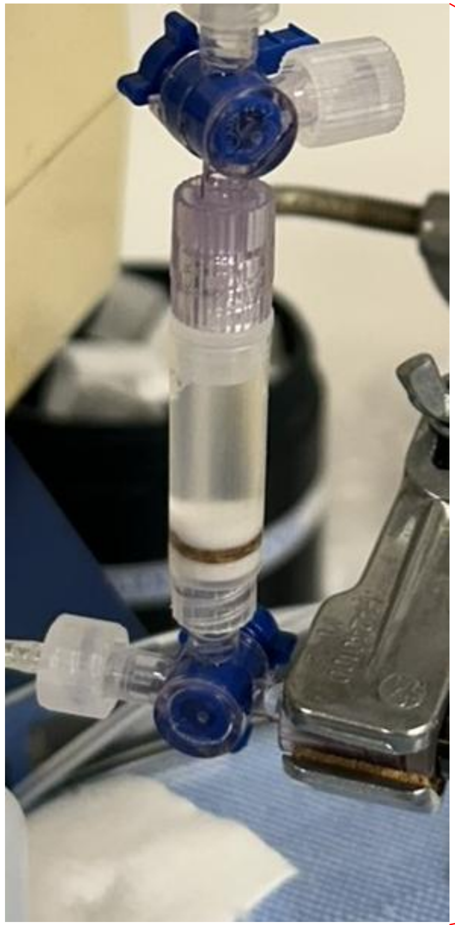

Fig. 16a: Detail of the apparatus – compartment of isolation

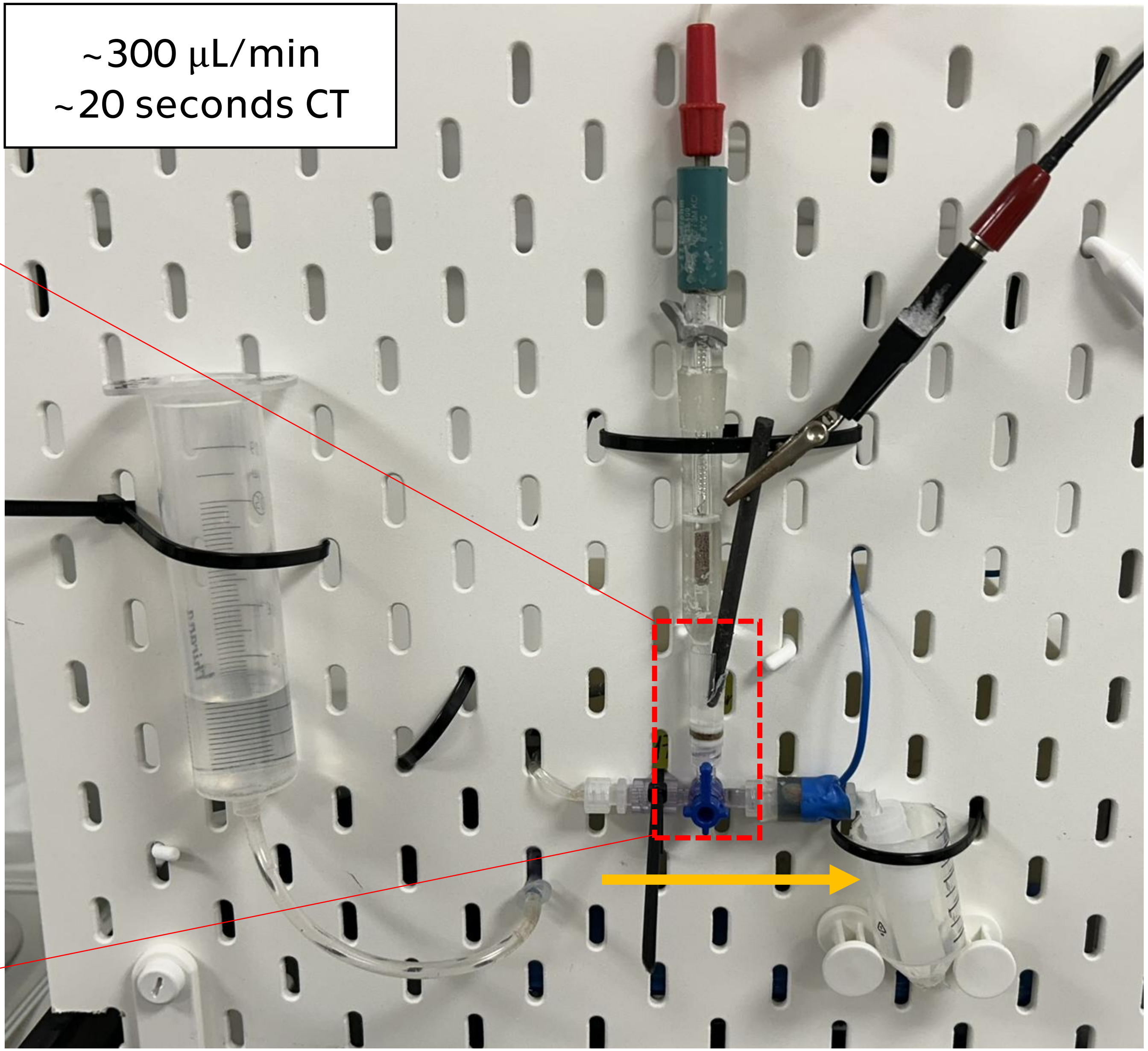

Fig. 15: Picture of RaFEC v1.2 apparatus (04/2025)

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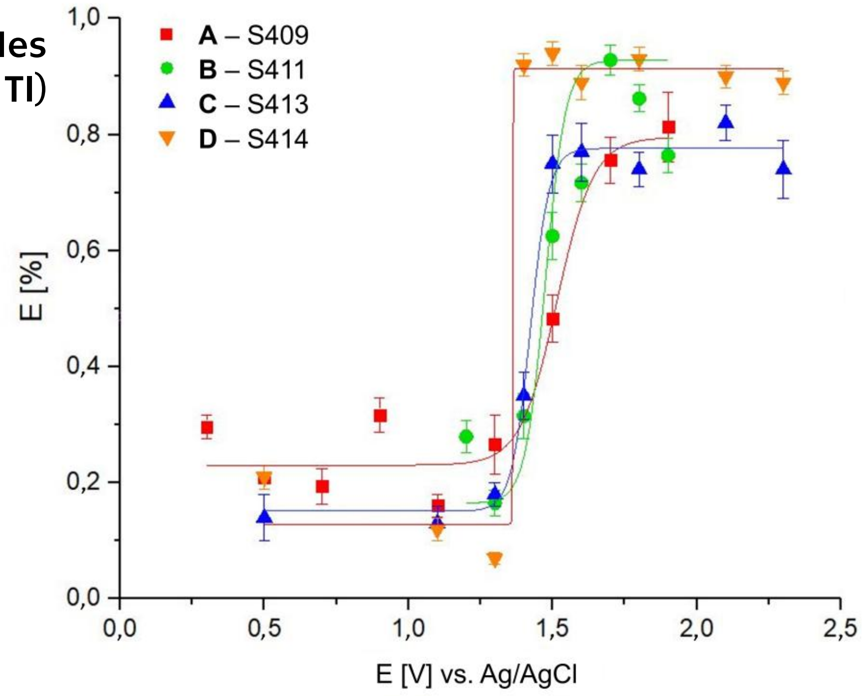
Results

- long way to optimization
- expected behavior in flow arrangement for cyclotron Tl
- problems with ^{201}Tl ($T_{1/2}$ = 3.04 d)

- significant sorption onto surfaces
- low activity for sufficient accuracy

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



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

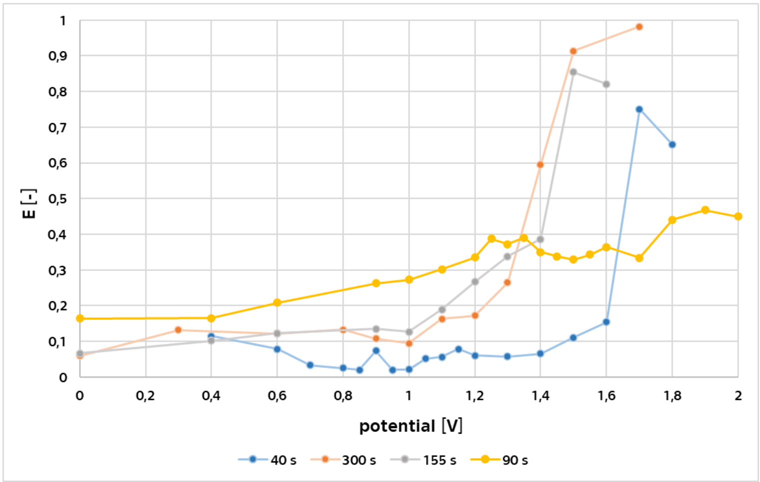


Fig. 17b: E for individual samples and potentials (Tl-201)

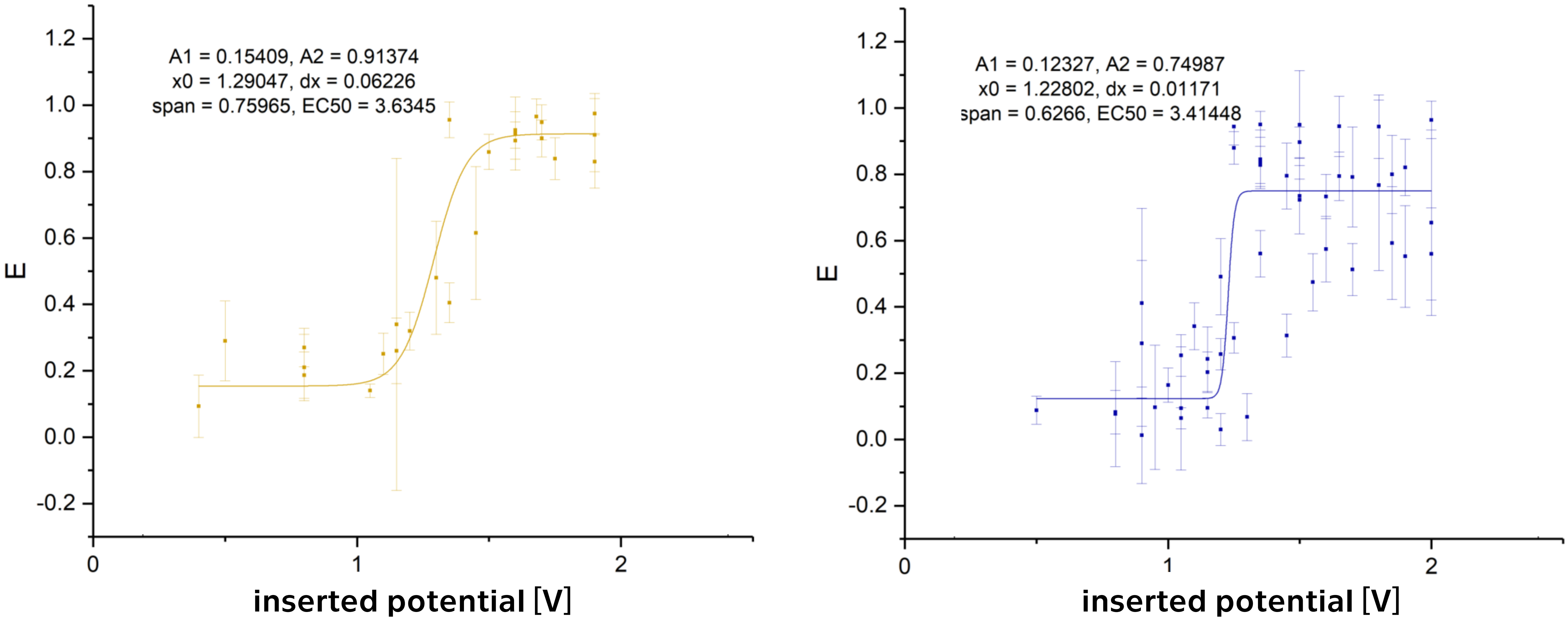


Fig. 18a+b: E for individual samples and potentials anion-exchange resin Dowex™ 1X2 100-200 mesh

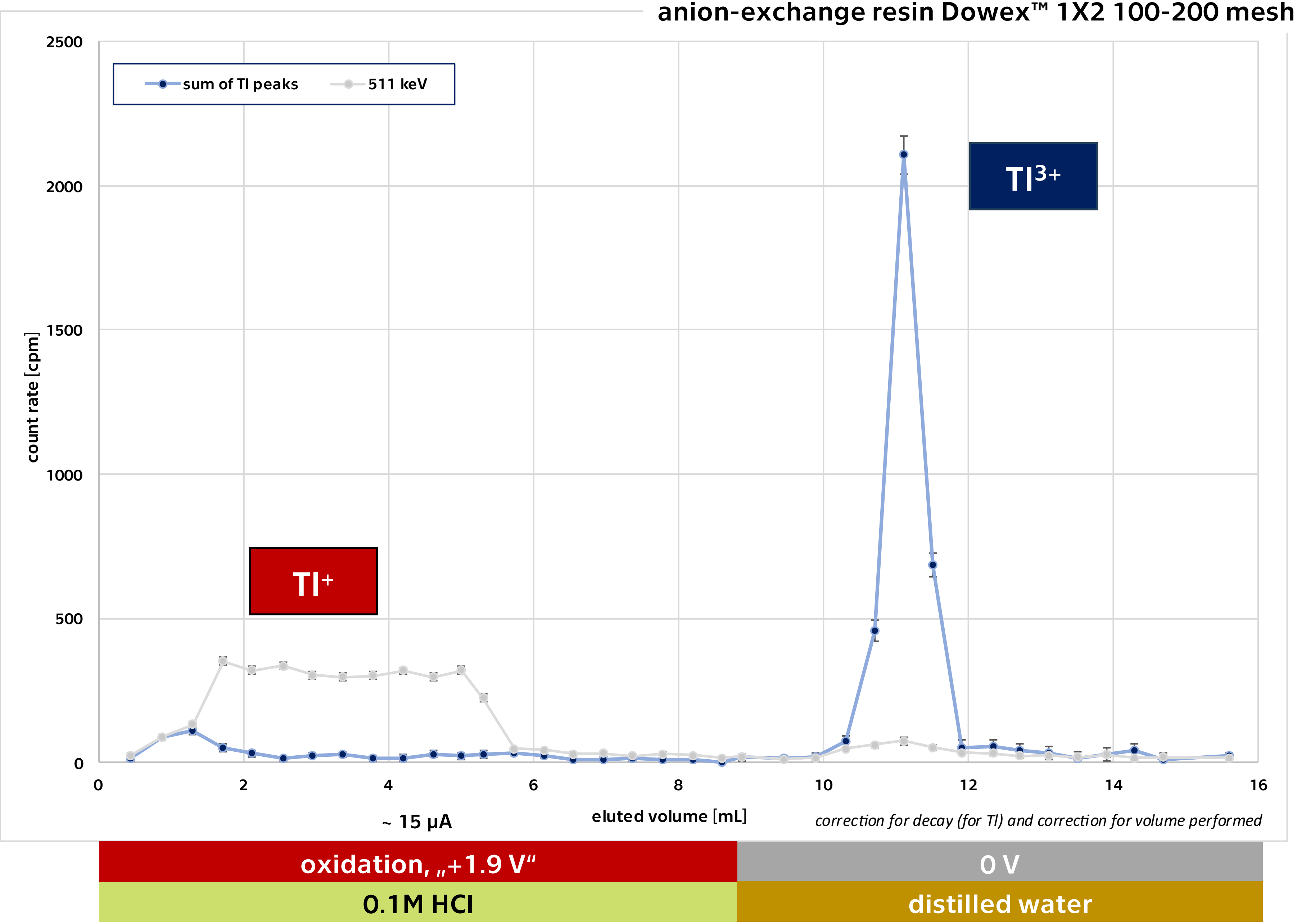


Fig. 19: Results of chromatography with anex

Conclusion

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

Further research

- optimization
- miniaturization
- isolation compartment
- sorption

- other elements
    - Ce, V-Db, Hg-Cn, Au-Rg
- automatization

Periodic Table 1–172

Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	Orbitals
1	1 H																	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											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

Period																Orbitals
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

Period																			Orbitals
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

[2] Phys. Chem. Chem. Phys., 2011, 13, 161-168

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