Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supplementary section A. Data for the proof of concept

Table A.1. Specific Activity change in graphite samples during leaching studies in LiCl-KCl eutectic at 723 K (0 mA current)

No	Sample ID*	Current, mA	m_{before}, g	m_{after}, g	m_{salt} g	Isotope	A_{before} Bq/g	A_{after}** Bq/g	A_{salt} Bq/g	Activity Balance*** %
1	O_1	0	0.2172	0.2172	176.026	^{60}Co	12145.87	12139.80	0.004	99.979
						^{133}Ba	228.80	228.50	-	99.889
						^{137}Cs	82.70	82.30	-	99.920
						^{154}Eu	73.20	72.90	-	99.966
2	S_1	0	0.4574	0.4574	176.037	^{60}Co	1489.66	1489.06	0.001	99.995
						^{133}Ba	204.00	203.80	-	99.993
						^{137}Cs	38.50	38.40	-	99.981
						^{154}Eu	55.90	55.60	0.001	99.970
3	W_1	0	0.4294	0.4294	176.012	^{60}Co	34870.12	34859.66	0.024	99.998
						^{133}Ba	423.70	423.40	0.001	99.997
						^{137}Cs	41.40	41.20	-	99.974
						^{154}Eu	146.30	145.60	0.002	99.973

* - O – Oldbury Magnox reactor, S – Sizewell Magnox reactor, W – Wylfa Magnox reactor

** - the 5 hours of leaching was tested.

*** - Activity balance was calculated using the following equation:

$$Activity\ balance = \frac{A_{after} \times m_{after} + A_{salt} \times m_{salt}}{A_{before} \times m_{before}} \times 100$$

Table A.2. Specific Activity change in graphite samples during the application of positive current in LiCl-KCl eutectic at 723 K (40 mA)

No	Sample ID*	Current, mA	m_{before}, g	m_{after}, g	m_{salt} g	Isotope	A_{before} Bq/g	A_{after} Bq/g	A_{salt} Bq/g	Activity Balance** %
1	O_1	40	0.2172	0.2172	176.066	^{60}Co	12139.80	12136.16	0.002	99.984
						^{133}Ba	228.50	228.40	-	99.994
						^{137}Cs	82.30	82.20	-	99.912
						^{154}Eu	72.90	72.40	0.001	99.919
2	S_1	40	0.4574	0.4574	176.034	^{60}Co	1489.06	1488.47	0.001	99.980
						^{133}Ba	203.80	203.70	-	99.997
						^{137}Cs	38.40	38.30	-	99.951
						^{154}Eu	55.60	55.30	0.001	99.973
3	W_1	40	0.4294	0.4294	176.047	^{60}Co	34859.66	34849.20	0.024	99.998
						^{133}Ba	423.40	423.00	0.001	99.993
						^{137}Cs	41.20	41.20	-	99.999
						^{154}Eu	145.60	144.70	0.001	99.643

* - O – Oldbury Magnox reactor, S – Sizewell Magnox reactor, W – Wylfa Magnox reactor

** - Activity balance was calculated using the following equation:

$$Activity\ balance = \frac{A_{after} \times m_{after} + A_{salt} \times m_{salt}}{A_{before} \times m_{before}} \times 100$$

Table A.3. Activity level in graphite samples after electrochemical treatment (40 mA current, up to two cycles) in LiCl-KCl eutectic at 723 K

Nº	Sample ID	Current, mA	m_{before}, g	m_1 g	m_{1salt} g	m_2 g	m_{2salt} g	Isotope	A_{before} Bq/g	A_1 Bq/g	A_{1salt} Bq/g	$\frac{A\,after+salt}{A\,before}$ %	A_2 Bq/g	A_{2salt} Bq/g	Activity Balance** %
1	O_1	40	0.217	0.215	176.040	0.213	176.025	^{60}Co	12136.16	8669.65	3.990	97.45	8095.44	0.578	97.96
								^{133}Ba	228.40	187.78	0.049	98.87	179.48	0.010	99.01
								^{137}Cs	82.20	76.45	0.006	97.87	74.65	0.002	98.39
								^{154}Eu	72.40	54.94	0.019	96.12	50.12	0.005	97.68
2	O_2	40	0.387	0.387	176.018	0.386	176.037	^{60}Co	14014.56	9119.00	9.648	96.38	8521.00	1.138	98.88
								^{133}Ba	213.69	180.66	0.067	98.76	171.41	0.018	99.11
								^{137}Cs	78.32	73.41	0.010	99.35	71.65	0.002	98.75
								^{154}Eu	77.42	61.21	0.034	98.89	55.11	0.012	98.41
3	O_3	40	0.232	0.230	176.040	0.229	176.016	^{60}Co	15114.12	8639.09	7.950	96.58	7955.35	0.710	97.97
								^{133}Ba	219.65	176.24	0.051	97.01	170.68	0.006	98.91
								^{137}Cs	80.23	74.88	0.006	98.55	73.05	0.002	98.70
								^{154}Eu	80.06	62.06	0.023	98.51	55.86	0.007	97.65
4	S_1	40	0.457	0.455	176.035	0.452	176.008	^{60}Co	1488.47	1224.44	0.591	97.18	1201.78	0.040	98.76
								^{133}Ba	203.70	170.95	0.077	98.10	166.01	0.006	97.89
								^{137}Cs	38.30	36.22	0.005	99.36	35.63	0.001	98.37

								^{154}Eu	55.30	51.60	0.006	97.12	48.82	0.005	97.95
5	S_2	40	0.459	0.458	176.019	0.448	176.030	^{60}Co	3639.96	2911.55	1.735	98.09	2823.33	0.274	98.47
								^{133}Ba	156.43	134.60	0.049	97.90	132.75	0.006	98.24
								^{137}Cs	26.92	24.96	0.004	98.82	25.12	0.000	99.06
								^{154}Eu	51.46	46.35	0.012	98.79	44.86	0.004	98.32
6	S_3	40	0.237	0.236	176.020	0.236	176.013	^{60}Co	1299.39	1079.15	0.259	97.51	950.44	0.141	97.84
								^{133}Ba	224.63	202.41	0.025	98.12	190.73	0.011	98.46
								^{137}Cs	63.13	60.00	0.004	98.99	58.42	0.001	98.31
								^{154}Eu	67.52	59.47	0.009	97.66	56.09	0.004	99.45
7	W_1	40	0.429	0.428	176.036	0.426	176.031	^{60}Co	34849.20	19100.22	35.769	96.80	15769.33	6.728	96.66
								^{133}Ba	423.00	297.56	0.281	97.41	276.88	0.035	97.43
								^{137}Cs	41.20	34.97	0.014	98.78	33.90	0.002	98.49
								^{154}Eu	144.70	120.19	0.056	98.64	107.59	0.026	97.91
8	W_2	40	0.600	0.596	176.023	0.595	176.024	^{60}Co	29726.33	21719.08	25.262	97.51	19568.23	5.888	97.95
								^{133}Ba	373.31	254.35	0.362	96.15	217.05	0.098	96.62
								^{137}Cs	67.69	56.83	0.035	98.68	53.28	0.010	98.62
								^{154}Eu	149.05	117.78	0.092	96.69	111.12	0.016	98.28
9	W_3	40	0.458	0.457	176.017	0.454	176.027	^{60}Co	29726.33	17791.39	29.030	97.25	15607.13	4.805	97.55

^{133}Ba	26.94	17.82	0.022	97.49	16.21	0.004	98.13
^{137}Cs	70.65	59.46	0.028	98.94	56.97	0.006	99.02
^{154}Eu	170.32	131.99	0.084	96.30	118.53	0.033	98.81

* - O – Oldbury Magnox reactor, S – Sizewell Magnox reactor, W – Wylfa Magnox reactor

** - Activity balance was calculated using the following equation:

$$Activity\ balance = \frac{A_{after} \times m_{after} + A_{salt} \times m_{salt}}{A_{before} \times m_{before}} \times 100$$

Supplementary section B. Data for the influence of applied current

Table B.1. Activity level in graphite samples after electrochemical treatment (up to 80 mA current, two cycles) in LiCl-KCl eutectic at 723 K

Nº	Sample ID	Current, mA	Cycle Nº	m_{before}, g	m_{after} g	m_{salt} g	Isotope	A_{before} Bq/g	A_{after} Bq/g	A_{salt} Bq/g	Activity Balance*** %
1	O_3	40	2	0.232	0.229	352.056**	^{60}Co	15114.12	7955.35	4.330	95.43
							^{133}Ba	219.65	170.68	0.028	96.15
							^{137}Cs	80.23	73.05	0.004	97.34
							^{154}Eu	80.06	58.78	0.015	96.71
2	O_4	20	2	0.487	0.486	176.016	^{60}Co	8070.00	7083.47	0.710	97.97
							^{133}Ba	205.19	202.54	0.006	98.91
							^{137}Cs	79.02	77.14	0.002	98.70
							^{154}Eu	69.32	61.29	0.007	97.65
3	O_5	60	2	0.380	0.377	176.020	^{60}Co	10575.68	4216.78	2.290	97.85
							^{133}Ba	207.09	166.42	0.004	99.26
							^{137}Cs	85.26	63.65	0.003	98.57
							^{154}Eu	71.36	51.39	0.016	96.51
4	O_6	80	2	0.384	0.381	176.016	^{60}Co	9573.28	2431.91	13.053	96.73
							^{133}Ba	142.58	49.52	0.080	97.73
							^{137}Cs	82.98	60.99	0.042	96.85
							^{154}Eu	81.35	50.01	0.041	98.11
5	S_4	20	2	0.616	0.615	176.008	^{60}Co	2952.99	2652.32	15.051	97.27
							^{133}Ba	93.44	91.40	0.192	96.23
							^{137}Cs	27.96	27.59	0.043	96.88
							^{154}Eu	79.48	72.49	0.062	95.79
6	S_5	40	2	0.600	0.599	176.030	^{60}Co	2839.30	2017.70	0.843	97.83
							^{133}Ba	146.80	118.55	0.004	98.85
							^{137}Cs	54.69	48.98	0.000	98.96
							^{154}Eu	731.56	585.70	0.018	97.51
7	S_6	80	2	0.315	0.314	176.022	^{60}Co	2862.45	853.05	2.495	96.73
							^{133}Ba	193.56	77.48	0.084	97.44
							^{137}Cs	24.68	17.93	0.017	98.64

							^{154}Eu	81.43	53.83	0.436	97.40
8	W_4	60	2	0.5872	0.5855	176.011	^{60}Co	28049.32	12266.30	3.398	96.04
							^{133}Ba	217.77	180.90	0.194	95.98
							^{137}Cs	39.13	30.34	0.011	98.36
							^{154}Eu	174.60	116.95	0.047	97.90

* - O – Oldbury Magnox reactor, S – Sizewell Magnox reactor, W – Wylfa Magnox reactor

** - total mass of salt is given after cycle 1 and cycle 2, for details on separate runs see table A.3.

*** - Activity balance was calculated using the following equation:

$$Activity\ balance = \frac{A_{after} \times m_{after} + A_{salt} \times m_{salt}}{A_{before} \times m_{before}} \times 100$$

Supplementary section C. Data for the influence of the cycle number

Table C.1. Activity level in graphite samples after electrochemical treatment (60 mA current, up to ten cycles) in LiCl-KCl eutectic at 723 K

No	Sample ID	Current, mA	Cycle No	m_{before}, g	m_{after} g	m_{salt} g	Isotope	A_{before} Bq/g	A_{after} Bq/g	A_{salt} Bq/g	Activity Balance** %
1	W_4	60	2	0.587	0.5855	176.032	^{60}Co	28049.32	12266.30	49.560	96.57
							^{133}Ba	217.77	180.90	0.107	97.58
							^{137}Cs	39.13	30.34	0.025	96.25
							^{154}Eu	174.60	116.95	0.172	96.39
2	W_5	60	1	0.449	0.446	176.014	^{60}Co	31297.56	13317.47	43.197	96.37
							^{133}Ba	363.16	316.78	0.100	97.49
							^{137}Cs	566.37	455.36	0.265	98.21
							^{154}Eu	136.66	109.87	0.061	97.36
3	W_6	60	4	0.498	0.497	176.022	^{60}Co	30954.34	11810.84	50.380	95.61
							^{133}Ba	228.76	177.74	0.127	97.12
							^{137}Cs	64.92	48.09	0.042	96.72
							^{154}Eu	123.55	83.12	0.106	97.56
4	W_7	60	6	0.265	0.260	176.034	^{60}Co	35014.78	12396.33	32.341	96.09
							^{133}Ba	263.51	77.67	0.267	96.16
							^{137}Cs	39.04	28.34	0.016	97.59
							^{154}Eu	156.32	87.86	0.096	96.07
5	W_8	60	8	0.217	0.213	176.010	^{60}Co	26072.99	9581.60	19.181	95.74
							^{133}Ba	453.22	83.20	0.441	96.91
							^{137}Cs	379.62	275.04	0.126	97.93
							^{154}Eu	161.03	50.73	0.130	96.44
6	W_9	60	10	0.491	0.472	176.008	^{60}Co	26978.51	6466.24	54.820	95.88
							^{133}Ba	440.95	72.42	0.990	96.25
							^{137}Cs	71.97	52.08	0.057	97.91
							^{154}Eu	163.58	29.01	0.361	96.09
7	W_10	60	8	0.459	0.448	176.020	^{60}Co	33648.72	11069.53	56.740	96.77
							^{133}Ba	369.85	62.01	0.767	95.88

No	Sample ID	Current, mA	Cycle No	m_{before}, g	m_{after} g	m_{salt} g	Isotope	A_{before} Bq/g	A_{after} Bq/g	A_{salt} Bq/g	Activity Balance** %
							^{137}Cs	51.33	38.04	0.036	98.91
							^{154}Eu	144.24	37.10	0.268	96.34
8	S_7	60	1	0.235	0.235	176.031	^{60}Co	957.43	389.57	0.715	96.64
							^{133}Ba	86.31	73.18	0.016	98.90
							^{137}Cs	39.47	32.40	0.008	97.33
							^{154}Eu	82.64	69.49	0.016	98.59
9	S_8	60	2	0.488	0.486	176.027	^{60}Co	1684.59	499.90	3.120	96.36
							^{133}Ba	113.89	95.73	0.045	97.91
							^{137}Cs	59.64	46.03	0.035	98.16
							^{154}Eu	597.32	431.92	0.439	98.53
10	O_7	60	4	0.469	0.464	176.019	^{60}Co	10845.90	4634.50	15.813	96.99
							^{133}Ba	147.63	114.20	0.087	98.73
							^{137}Cs	76.95	56.21	0.054	98.49
							^{154}Eu	81.35	54.19	0.068	97.45
11	O_8	60	6	0.457	0.452	176.013	^{60}Co	8784.59	3378.87	13.436	96.95
							^{133}Ba	213.53	71.03	0.352	96.47
							^{137}Cs	86.54	62.20	0.062	98.63
							^{154}Eu	67.23	41.59	0.064	98.06
14	O_9	60	10	0.4908	0.4716	176.027	^{60}Co	8573.28	2433.44	16.476	96.20
							^{133}Ba	163.54	35.63	0.347	97.04
							^{137}Cs	75.69	56.52	0.056	98.43
							^{154}Eu	75.63	19.40	0.152	96.52

* - O – Oldbury Magnox reactor, S – Sizewell Magnox reactor, W – Wylfa Magnox reactor

*** - Activity balance was calculated using the following equation:

$$Activity\ balance = \frac{A_{after} \times m_{after} + A_{salt} \times m_{salt}}{A_{before} \times m_{before}} \times 100$$

Supplementary section D. Data for potential-time profiles of the cyclic galvanostatic electrolysis

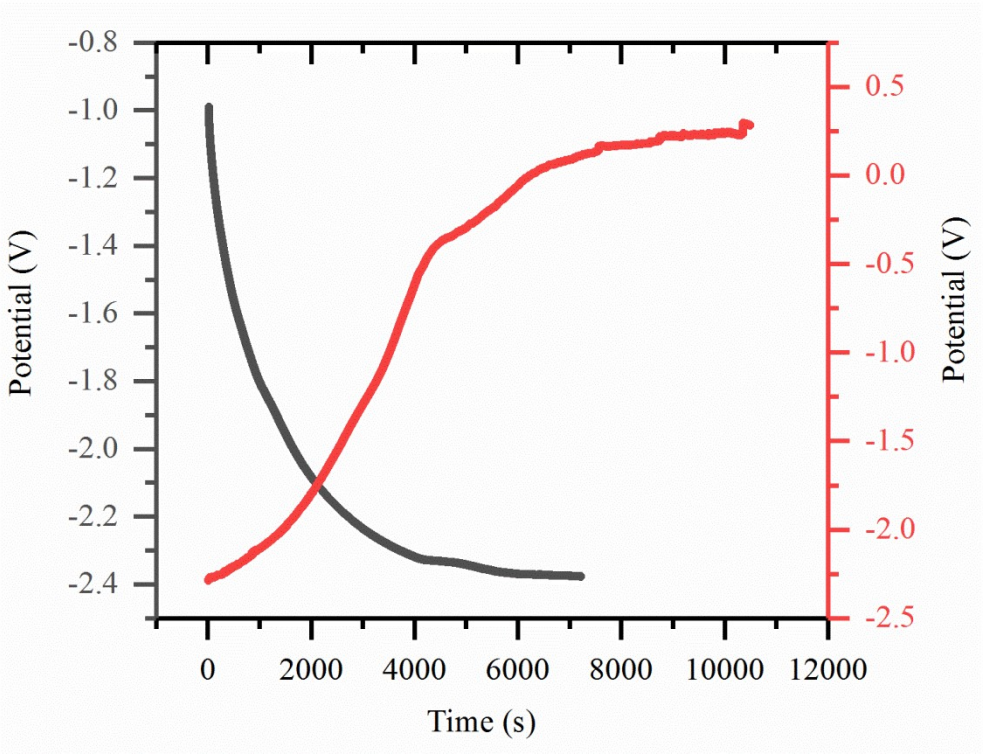

Figure D.1. Galvanostatic transients recorded under 40 mA negative (black) and 40mA positive (red) currents applied to irradiated graphite sample from Oldbury reactor placed in LiCl-KCl eutectic at 723 K

Supplementary section E. Data for cyclic voltammetry of the molten salts recorded before and after electrolysis

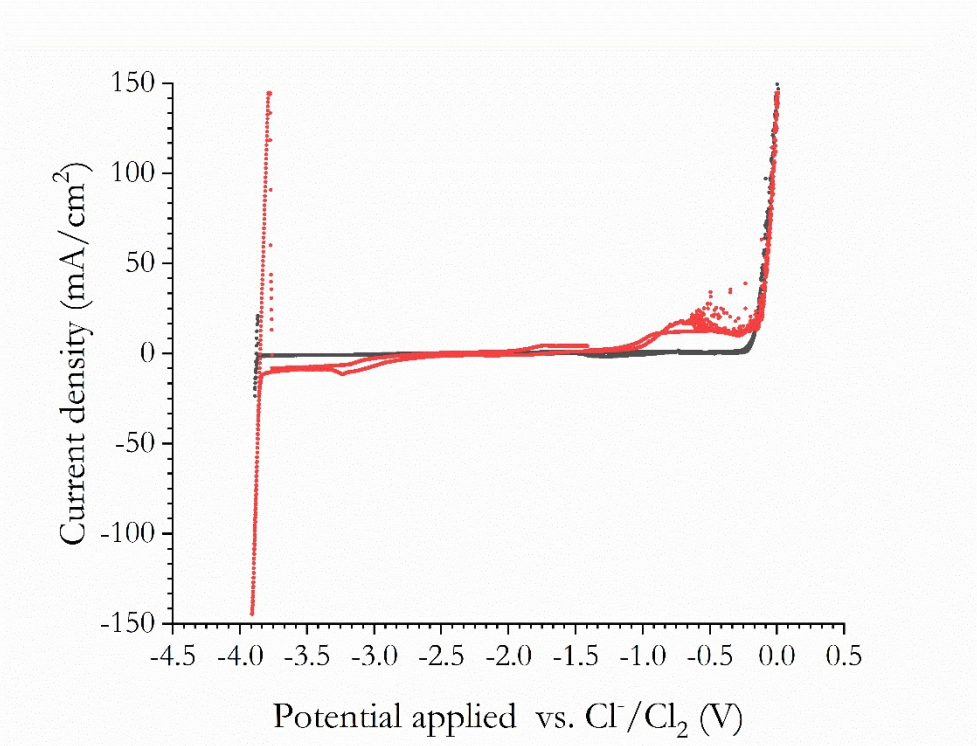

Figure E.1. Cyclic voltammograms of LiCl-KCl eutectic at 723 K recorded at scan rate of 100 mV/s before electrolysis (black) and after electrolysis (red) of irradiated graphite sample from Wylfa reactor at 40 mA. Working electrode: tungsten wire (S = 0.69 cm^2); reference electrode: 1 wt% Ag/AgCl; counter electrode: molybdenum wire (Ø 0.5 mm)