1 Supplementary information

Table 1 contains the raw data from solution for each sample.

Table 1: Element concentrations (ppb) measured at each time point during the leach test.

Sample	Time point (hr)	Al (ppb)	Si (ppb)	K (ppb)	Sr (ppb)	Cs (ppb)
Sr_1	2	9428.4	5965.3	211166.7	15.8	-
	7	4297.1	16336.7	216000.0	9.7	-
	24	9292.6	31083.3	223233.3	11.9	-
	48	21889.6	23670.0	100923.3	13.6	-
	72	8273.9	22763.3	70713.3	23.4	-
	96	32246.6	22260.0	67176.7	21.6	-
	120	28503.1	21430.0	51790.0	22.3	-
	144	23597.1	21020.0	49223.3	15.4	-
	168	22557.9	20460.0	45560.0	10.6	-
	192	21640.8	22020.0	44446.7	11.1	-
	216	20577.4	18676.7	38053.3	11.9	-
	240	19480.1	19470.0	38610.0	11.4	-
	264	18835.4	20843.3	32300.0	10.3	-
	336	33243.0	24646.7	66423.3	17.1	-
	504	46074.7	24423.3	94830.0	19.3	-
	672	36889.8	25073.3	68250.0	15.8	-
	840	35522.8	26550.0	58770.0	16.3	-
Sr_3	2	6984.7	12715.7	317566.7	16.5	-
	7	4272.3	27593.3	338633.3	22.3	-
	24	8908.8	42063.3	332933.3	34.2	-
	48	8291.5	29323.3	144200.0	45.7	-
	72	7151.6	25823.3	92366.7	54.4	-
	96	28954.8	23416.7	79370.0	47.5	-
	120	25868.0	21810.0	59233.3	41.7	-
	144	22471.8	21636.7	54830.0	43.7	-

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Table 1 – *Continued from previous page*

Sample	Time (hr)	Al (ppb)	Si (ppb)	K (ppb)	Sr (ppb)	Cs (ppb)
	168	22324.0	21193.3	52306.7	47.3	-
	192	21234.2	22516.7	46166.7	41.7	-
	216	24834.4	20956.7	46190.0	31.6	-
	240	18439.1	20830.0	41400.0	41.8	-
	264	17310.2	21563.3	30133.3	35.1	-
	336	30513.6	25856.7	63113.3	66.1	-
	504	39460.0	25426.7	84710.0	66.7	-
	672	33043.7	26766.7	61743.3	52.4	-
	840	32255.8	27810.0	53826.7	57.6	-
	2	10743.3	7130.3	304733.3	-	18199.3
	7	5135.4	11344.0	148603.3	-	12058.1
	24	11255.7	25233.3	178166.7	-	9301.0
	48	42231.1	22876.7	96006.7	-	6049.7
	72	22508.3	22296.7	77326.7	-	4509.7
	96	41114.8	22876.7	72860.0	-	4330.7
	120	35936.1	22083.3	61656.7	-	3692.8
	144	32334.2	22306.7	61260.0	-	3407.4
Cs_1	168	29742.7	22136.7	59573.3	-	3017.6
	192	29714.8	24446.7	56413.3	-	2816.8
	216	23870.6	20143.3	43263.3	-	1606.6
	240	25061.6	23806.7	50020.0	-	2494.5
	264	23549.2	23496.7	37070.0	-	2291.0
	336	38003.5	24873.3	67533.3	-	3612.8
	504	43044.2	25170.0	90446.7	-	4439.3
	672	38719.3	25520.0	67003.3	-	3960.8
	840	36871.3	25276.7	56813.3	-	3452.5
	2	5978.1	5280.3	277466.7	-	52142.2
	7	4878.2	14.9	896.8	-	48647.2

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Table 1 – *Continued from previous page*

Sample	Time (hr)	Al (ppb)	Si (ppb)	K (ppb)	Sr (ppb)	Cs (ppb)
	24	11213.6	29060.0	201966.7	-	32505.1
	48	28192.3	22423.3	90230.0	-	18001.0
	72	22794.7	22446.7	85193.3	-	13764.4
	96	39813.7	22216.7	69330.0	-	12861.5
	120	34574.2	21773.3	56183.3	-	11044.3
	144	30524.8	21963.3	55780.0	-	9925.4
	168	26513.4	21160.0	51796.7	-	8547.7
	192	25365.1	23233.3	45536.7	-	7580.1
	216	25356.0	20073.3	42416.7	-	7278.3
	240	22662.2	22596.7	44653.3	-	6977.6
	264	21026.3	21946.7	29353.3	-	6444.7
	336	36840.6	24783.3	64856.7	-	10881.3
	504	43391.4	23680.0	87533.3	-	13956.7
	672	38896.1	25173.3	63786.7	-	12467.3
	840	37733.0	23870.0	54903.3	-	11009.8

Figure 1 shows the CFL calculated for aluminium, silicon, and potassium.

Table 2 contains the pH data measured at each time point.

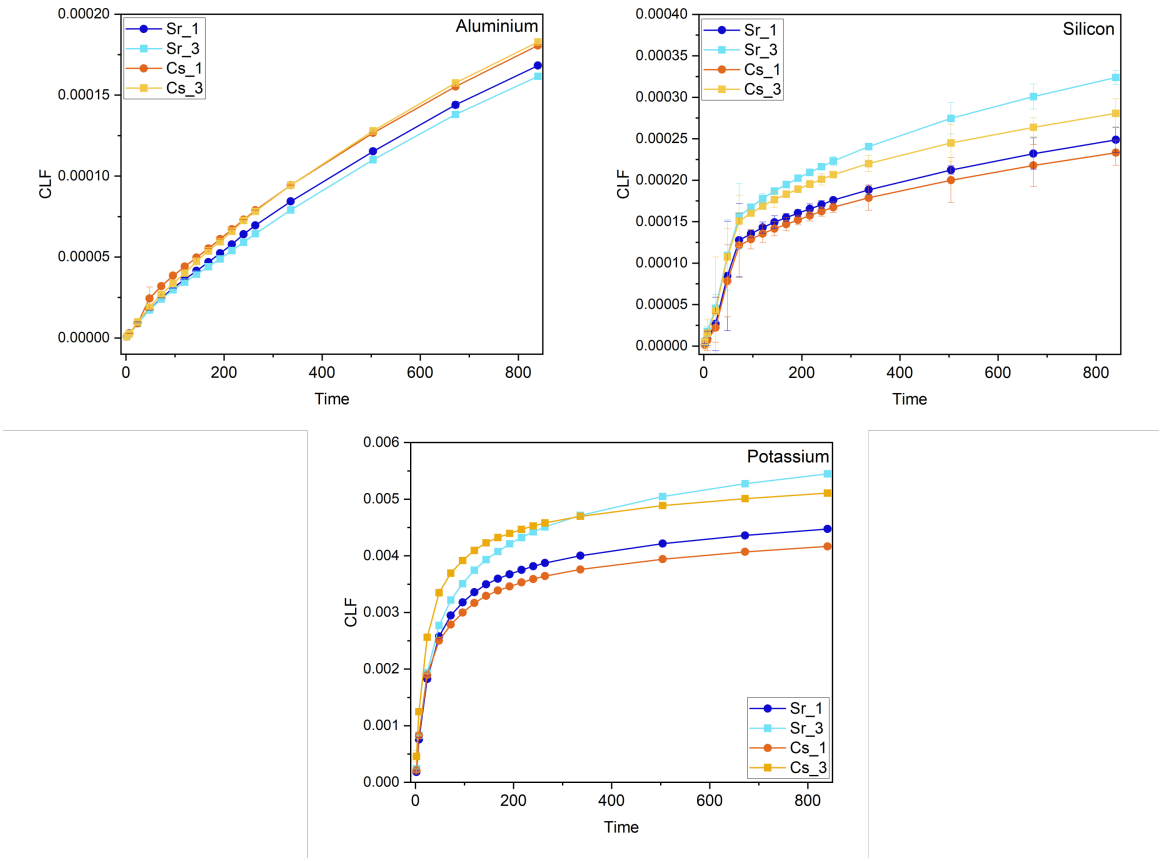

Figure 1: Calculated CFL for Al, Si, and K.

Table 2: pH measured at each time point during the leach test.

Time point (hr)	**Control**	**Cs_1**	**Sr_1**	**Cs_3**	**Sr_3**	**Blank**
2	9.80	9.99	10.07	10.72	10.63	7.02
7	10.47	10.69	10.71	11.10	11.17	6.99
24	10.95	11.16	11.32	11.28	11.18	6.71
48	10.84	10.87	10.91	10.91	11.02	6.26
72	10.58	10.73	10.71	10.76	10.78	6.34
96	10.48	10.62	10.57	10.59	10.69	6.37
120	10.41	10.57	10.49	10.47	10.53	6.57
144	10.33	10.40	10.40	10.40	10.51	6.00
168	10.17	10.22	10.30	10.28	10.43	6.46
192	10.01	10.18	10.14	10.35	10.35	6.20
216	9.93	10.09	10.15	10.13	10.26	6.47
240	10.08	10.11	10.02	10.03	10.26	7.50
264	9.77	10.02	10.01	10.03	10.39	6.65
336	10.35	10.32	10.44	10.30	10.61	6.79
504	10.31	10.41	10.47	10.41	10.73	7.39
672	10.17	10.20	10.27	10.19	10.39	7.82
840	9.84	10.01	10.16	9.98	10.32	7.20

Tables 3 - 5 show the PHREEQC modelling input parameters and saturation indices of selected mineral systems of Cs and Sr.

Table 3: Input parameters for PHREEQC modelling.

Parameter	**Value**	**Units**
Temperature	20	Celsius
pH	10.0	pH unit
Redox state	±4	pe
Solution volume and density	Volume = 1 kg Density = 1	-
Concentration Al	0.9	ppm
Concentration Si	12.5	ppm
Concentration K	1.1	ppm
Concentration Sr*	0.006	ppm
Concentration C(4)	1**	ppm
Concentration Cs*	30	ppm

*Sr removed for Cs system, Cs removed for Sr system. **Estimated (system is open to air).

Table 4: Saturation index of selected mineral Cs system.

Mineral	**Formula)**	**Saturation index**
Boehmite	AlO(OH)	0.97
Gibbsite	$Al(OH)_3$	0.89
Illite	$K_{0.85}Al_{2.8}Si_{3.15}O_{10}(OH)_2$	0.54
Kaolinite	$Al_2(Si_2O_5)(OH)_4$	0.30
Caesium carbonate	$CsCO_3$	-21.37

Table 5: Saturation index of selected mineral Sr system.

Mineral	**Formula)**	**Saturation index**
Boehmite	AlO(OH)	0.97
Gibbsite	$Al(OH)_3$	0.89
Illite	$K_{0.85}Al_{2.8}Si_{3.15}O_{10}(OH)_2$	0.54
Kaolinite	$Al_2(Si_2O_5)(OH)_4$	0.30
Strontianite	$SrCO_3$	-2.21
Strontium hydroxide	$Sr(OH)_2$	-15.15

Figure 2 shows the unchanged XRD diffractograms before and after leaching.

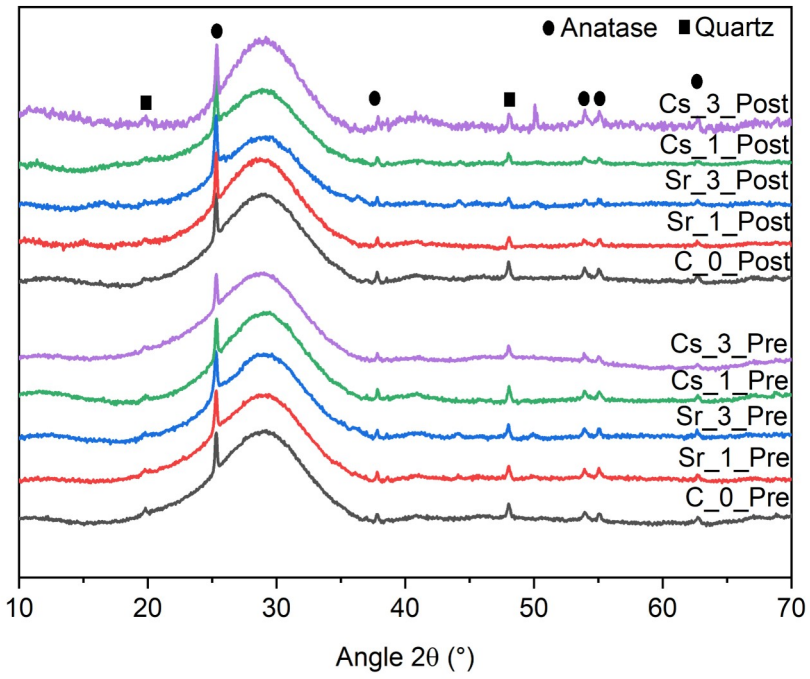

Figure 2: XRD data for geopolymer gels with varying incorporation of Sr and Cs, both pre- and post-leach. Sample codes are formatted as (1,3) % RN where RN is the radionuclide Sr or Cs and 1 or 3 refers to the mass percentage of its addition in hydroxide form.

Figures 3 - 5 show EDX scans of the bulk sample, to determine the elemental composition of the bulk sample.

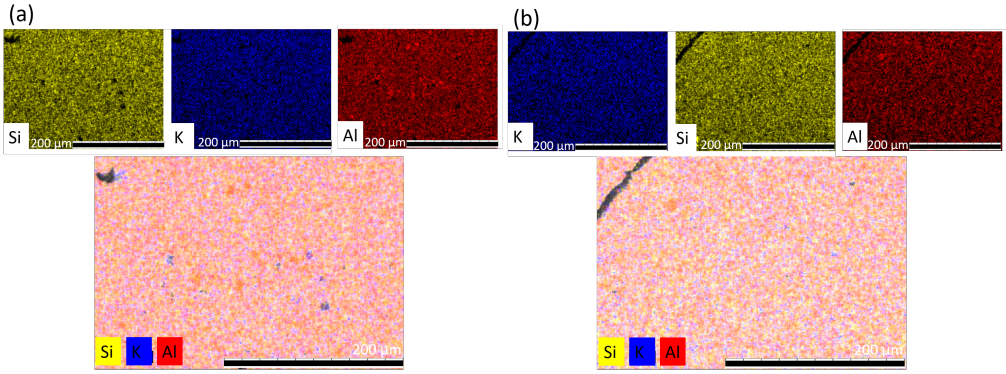

Figure 3: EDX scans for (a) pre-leach control and (b) post-leach control samples. Scans taken for 10 minutes for Si, Al, Cs, and K.

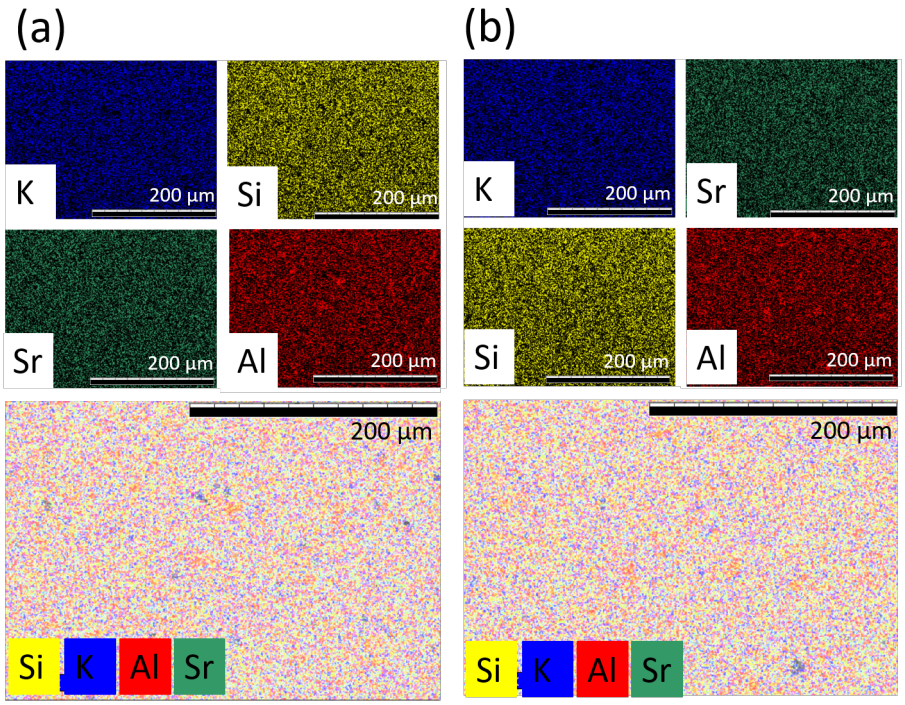

Figure 4: EDX scans for (a) Sr_3_Pre and (b) Sr_3_Post samples. Scans taken for 10 minutes for Si, Al, Cs, and K.

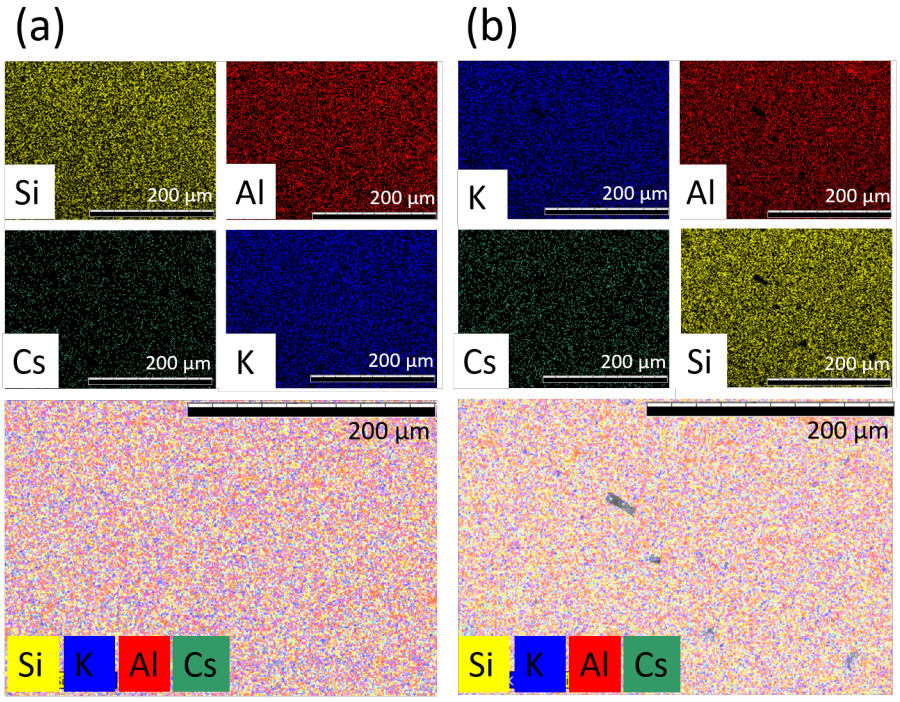

Figure 5: EDX scans for (a) Cs_3_Pre and (b) Cs_3_Post samples. Scans taken for 10 minutes for Si, Al, Cs, and K.

Figure 6 shows low-resolution SEM images of the sample edge, before and after leaching, to determine if there is the formation of an alteration layer.

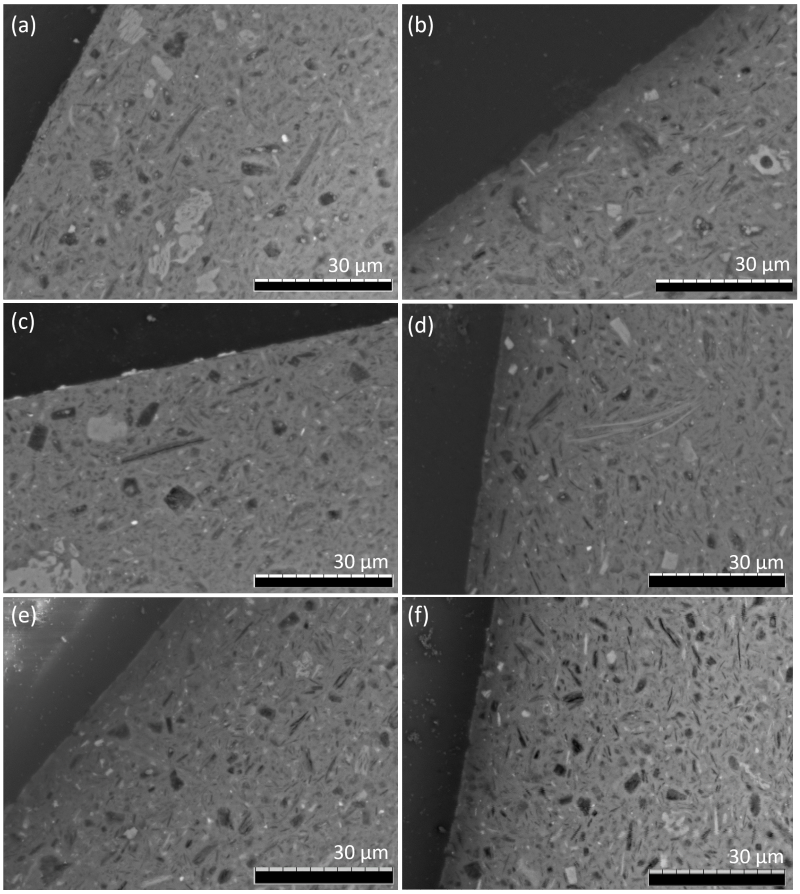

Figure 6: Low-resolution SEM images of the sample edge for (a) C_0_Pre, (b) C_0_Post, (c) Sr_3_Pre, (d) Sr_3_Post, (e) Cs_3_Pre, and (f) Cs_3_Post.

Figure 7 shows the FTIR spectra before and after leaching. A red box is included to indicate where carbonation peaks occur.

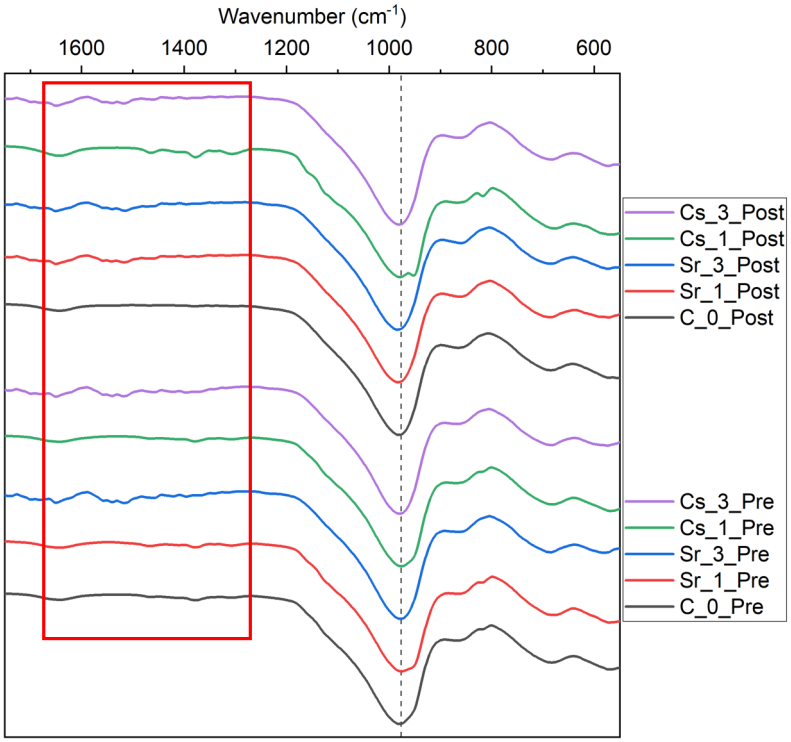

Figure 7: FTIR spectra of geopolymer gels with varying incorporation of Sr and Cs both pre- and post-leach. Red box indicates area where carbonation peaks occur.