

Potential Element Retention by Weathered Pulverised Fuel Ash : II. Column Leaching Experiments

Sanghoon Lee*

ABSTRACT: Column leaching tests were conducted using fresh and weathered pulverised fuel ash of some 17 and 40 years old from two major British power plants, with deionised water and simulated synthetic industrial leachate. The former was to see the leaching behaviour of weathered ash and the latter was to see if the formation of secondary products from water and PFA interaction and ameliorating effect in removing metals from industrial leachates. Fresh PFA liberates elevated concentrations of surface-enriched inorganics, including Ca, Na, K, B, C_{Total} , Li, Mo, Se and SO_4^{2-} . This might indicate their association with the surface of PFA particles. In the column leaching tests using weathered ash and deionised water, elements are not readily leached but are released more slowly, showing relatively constant concentrations. For the case of weathered ash, some readily soluble surface-enriched elements appears to have been liberated in their early stage of leaching and the liberation of glass associated elements are thought to be more important function in controlling the element concentration. The result from column leaching tests exceed for a number of elements when compared with various Water Standards and suggests the leachate from PFA disposal mound needs dilution to achieve target concentrations. PFA shows element retention effect for many elements, including B, Fe, Zn, Hg, Ni, Li and Mo, in the order of fresh Drax ash > weathered Drax ash > Weathered Meaford ash in retaining capacity. Geochemical modelling using a computer program WATEQ4F reveals some solubility controlling secondary solid products. These include $CaSO_4 \cdot 2H_2O$ for Ca, $Al(OH)_3$ for Al and $Fe(OH)_3$ for Fe.

INTRODUCTION

Pulverised Fuel Ash (PFA) is unstable in the weathering environment and as a result of weathering reactions secondary minerals form and elements are also lost into solution. This paper has been concerned with the long-term behaviour of PFA in the weathering environment and arising from this the possibility that weathered PFA could have an ameliorating effect on metal-rich, landfill leachates. To investigate this possibility batch-leaching and column leaching experiments were devised. The batch leaching experiments were described in a previous paper (Lee, this issue) and this report deals with the column leaching. The experiments are inter-related and therefore the conclusions from the first experiment are summarised below.

BATCH LEACHING EXPERIMENTS-SUMMARY

In the batch leaching experiments with fresh PFA from Drax power-station Ca, Na, K, S, B, C_{Total} , Cu, Li, Ni, Hg, Mo and Cl^- were readily leached and there is evidence of a surface association. Mg, Al, Ba, Si, V, As

and Se are less readily leached and these elements may be associated with the glass in the PFA. The samples of weathered PFA from Drax and Meaford demonstrate that the soluble elements associated with unstable phases such as the glass continue to pass into solution. In the experiments with a simulated leachate decreases in solution concentrations were noted for Fe, Ca, C_{Total} , Cu, Ni, Zn and Hg. The loss from solution and gain by the PFA was confirmed by analyses of the PFA, with the exception of Cd and Hg which were not included within the suite of elements analysed by XRF. The order of metal retention was determined as Meaford > weathered Drax > fresh Drax which is in agreement with the weathering sequence from most to least weathered. Confirmation of these results and conclusions will be sought in the column leaching tests.

EXPERIMENTS

Samples and Simulated Leachate

The same samples of PFA were used in the column leaching as in the batch leaching, namely fresh PFA from Drax, weathered PFA from the Barlow disposal mound [NGR SE 655277] and weathered PFA from the decommissioned power station at Meaford [NGR SS

*Institute of Natural Sciences, Yonsei University 134, Seoul 120-749, Korea

Table 1. Summary of test conditions for column leaching test.

Test Code	Material: (samples and solutions used)	D×H (cm)	Dry weight (gr.)	Moisture content(%)	Flow rate (mm/24 hour)	Bulk density (dry basis)
DFS	Drax fresh sample with synthetic solution	5×40	1,000	2.17	25	1.25
DFD	Drax fresh sample with deionised water	5×40	940	2.17	25	1.17
DWS	Drax weathered sample with synthetic leachate	5×40	1,000	4.99	25	1.21
DWDD	Drax fresh sample with synthetic leachate	5×40	970	4.99	25	1.17
MWSD	Drax fresh sample with synthetic leachate	5×40	1,000	11.36	25	1.13
MWDD	Drax weathered sample with deionised water	5×40	990	11.36	25	1.12

Table 2. Composition of synthetic leachate analysis by ICP(Unit : ppm unless indicated). Those elements whose concentrations are shown in brackets originate as impurities in the standard chemicals.

pH	Ca	Na	K	Mg	Fe	Al	B	Ba	Cd
4.55	615	1032	735	87	123	(0.12)	(0.23)	0.4	121
Cr	Cu	Li	Mn	Ni	Pb	Si	Sr	Ti	V
80	118	19	(0.18)	109	(0.22)	(0.81)	(0.16)	(0.05)	(0.11)
Zn	As	Hg	Se	Mo	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻		
108	(0.27)	13.8	(0.33)	(0.68)	1973	21	419		

896373].

The composition of the simulated leachate is shown on Table 1. In addition to the elements added other elements are present as trace impurities.

Design of Columns and Experimental Procedures

Six columns were constructed of acylyic plastic tube to the design shown on Fig. 1 which is modified from Brown *et al* (1976). The PFA samples in their natural moisture condition were loaded into the columns in stages and lightly packed to achieve uniform densities. The dimensions of the column and the bulk densities are given in Table 2. 1000 mls of solution, either deionised water or synthetic leachate, were passed through each column at a flow rate of 25 mm/day. The solution passing through the column was sampled every 50 ml. This sample was divided into two aliquots, one untreated for the anion determinations by ion chromatography (Dionex 2000i) and the other acidified with 1% HNO₃ for the determination of elements by ICP-AES. The results are given in Table 3 for the deionised water and Table 4 for the synthetic leachate. Difficulties were experienced with the flow rate with leachate for the Mea-

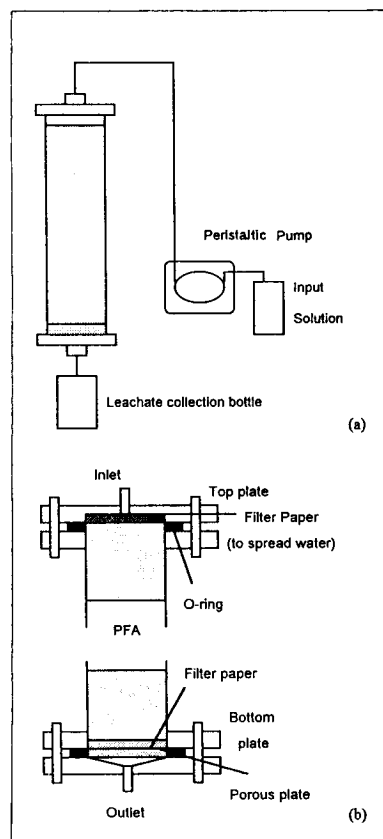


Fig. 1. Experimental setting for column leaching test in downflow configuration : (a) Whole system (b) Details for inlet and outlet parts.

ford weathered sample and only the first 150 ml of the solution passing through the column were analysed.

In the batch leaching 50 gms of sample were treated with successive aliquots of 500 ml of water or leachate. The solution volume is large compared with the sample mass. In the column leaching these proportions differ with 1000 gms of sample through which flows 1000 ml of solution.

The infiltration of solution through the PFA is more closely related to the natural system than is the batch leaching, but on the other hand the experiment takes longer to perform.

Column Leaching of Fresh Drax PFA with Deionised Water

The results (dfd1-1~1-13, Table 3) show for a number of elements high initial concentrations falling significantly as more water passes through the column. The

Table 3. Analytical results for leachates of column leaching test treated by deionised water (unit : mg/l unless indicated).

	pH	Eh(mv)	C(μ S/cm)	Ca	Mg	Na	K	Fe	Al	B	Ba	Cd	Co	Cr	Cu
dfd1-1	9.35	269.3	22300	353.4	36.7	5760.5	2113.5	0.17	0.99	72.21	0.08	0.05	0.05	23.95	0.14
1-2	9.39	262.4	3430	350.7	22.5	1571.4	804.6	0.17	0.27	45.03	0.06	0.01	0.02	5.16	0.03
1-3	9.49	267.0	1630	430.9	20.9	471.0	433.0	0.31	0.24	33.07	0.06	0.01	0.02	1.66	0.01
1-4	9.46	235.7	1030	499.8	21.7	179.4	282.9	0.17	0.26	29.23	0.05	0.01	0.00	0.86	0.01
1-5	9.29	248.5	899	499.9	22.3	86.6	194.1	0.19	0.30	26.91	0.05	0.01	0.00	0.51	0.01
1-6	9.42	257.2	821	548.5	23.7	53.3	156.4	0.16	0.30	27.16	0.05	0.01	0.01	0.34	0.00
1-7	9.44	263.0	793	565.2	22.4	35.8	117.3	0.41	0.32	25.14	0.04	0.23	nd	0.50	0.43
1-8	9.20	270.4	759	587.1	22.0	27.2	102.2	0.33	0.37	23.33	0.04	0.01	0.05	0.12	0.09
1-9	9.14	249.8	744	601.1	21.6	24.5	89.8	0.20	0.29	22.28	0.03	0.00	nd	0.16	0.01
1-10	9.12	223.2	747	595.7	20.0	21.2	81.8	0.16	0.32	20.70	0.03	nd	nd	0.13	0.02
1-11	9.13	226.3	736	590.4	18.3	17.8	73.8	0.13	0.34	19.13	0.03	nd	nd	0.10	0.03
1-12	9.01	-	714	582.7	17.7	17.5	65.8	0.12	0.26	17.53	0.04	nd	nd	0.12	0.03
1-13	9.11	-	716	588.6	15.5	15.6	59.5	0.06	0.29	16.00	0.03	nd	0.02	0.08	0.03
dwd3-1	7.49	278.7	9560	1626.2	232.5	141.8	157.1	0.21	0.21	2.10	0.33	0.02	0.03	0.20	0.04
3-2	8.10	276.4	2960	516.8	65.3	57.9	79.0	0.05	0.05	1.50	0.15	0.00	0.01	0.10	0.01
3-3	8.11	285.0	330	112.9	14.1	25.1	42.6	0.12	0.06	1.31	0.14	0.00	0.02	0.06	0.01
3-4	8.21	287.7	281	93.5	11.8	17.6	39.0	0.10	0.04	1.21	0.15	0.00	0.01	0.04	0.00
3-5	8.25	255.4	171	64.3	8.2	11.3	30.5	0.15	0.06	0.98	0.09	0.00	0.01	0.05	0.00
3-6	8.32	251.0	145	53.7	6.9	9.0	26.7	0.16	0.08	0.95	0.92	0.00	0.00	0.05	0.00
3-7	8.35	242.8	144	60.8	7.7	11.3	29.3	0.07	0.05	1.51	0.11	nd	0.02	0.05	0.01
3-8	8.32	249.6	128	54.3	6.8	9.0	27.5	0.03	nd	1.13	0.11	nd	nd	nd	0.01
3-9	8.34	231.7	123	53.3	5.5	6.4	22.4	0.00	nd	0.78	0.10	nd	nd	0.07	nd
3-10	8.13	241.2	104	47.9	6.0	8.4	24.9	0.04	nd	0.85	0.12	nd	nd	nd	0.01
3-11	8.31	-	109	47.9	6.0	6.6	23.1	0.00	nd	0.82	0.15	nd	nd	0.04	0.03
3-12	8.23	-	103	44.6	5.7	5.6	23.1	0.01	nd	0.68	0.14	nd	nd	0.00	0.03
mwd6-1	8.54	274.7	499	225.3	28.8	32.3	41.7	0.12	0.07	1.89	0.13	0.02	0.02	0.11	0.03
6-2	8.26	264.4	373	182.2	25.2	19.9	35.6	0.15	0.07	1.11	0.13	0.01	0.01	0.10	0.02
6-3	8.34	255.8	366	190.7	24.6	21.5	38.2	0.00	nd	0.89	0.13	0.08	0.04	0.11	0.00
6-4	8.28	239.3	279	146.2	19.1	17.3	32.0	0.03	0.03	0.83	0.11	nd	nd	0.04	0.04
6-5	8.34	235.0	210	98.8	13.0	13.7	28.4	0.04	nd	0.78	0.09	nd	0.00	0.07	0.01
6-6	8.43	230.5	177	106.5	14.1	13.8	27.1	0.04	0.05	0.72	0.12	nd	nd	0.03	0.02
6-7	8.34	-	154	80.6	10.7	11.7	25.8	0.03	0.05	0.78	0.11	nd	0.02	0.04	0.01
6-8	8.18	253.7	145	66.9	9.1	10.4	22.2	0.06	0.08	0.72	0.12	nd	0.01	0.02	0.00
6-9	8.26	-	140	62.4	8.3	9.7	23.1	0.01	0.03	0.78	0.12	nd	0.03	0.01	0.01
6-10	8.28	-	129	59.0	7.8	8.7	20.4	0.02	nd	0.78	0.13	nd	nd	nd	0.03
6-11	8.31	-	113	55.2	7.5	7.8	22.2	0.07	nd	0.71	0.12	nd	nd	0.01	0.03
6-12	8.28	-	100	48.5	6.3	7.4	16.0	0.04	0.00	0.56	0.11	nd	nd	nd	0.03
6-13	8.24	-	99	49.7	6.5	6.4	17.8	nd	nd	0.55	0.12	nd	nd	0.01	0.01

nd = not determined

electrical conductivity (Table 3) demonstrates the aggregate decrease in concentrations. The solution remains at a pH in excess of 9.0. Na and K show this pattern of high initial concentrations falling significantly with flow through the system. The initial concentrations for these two elements are 100 times the values recorded in the batch leaching and this reflects the relative change in proportion of sample mass to solution volume. Ca and S (as SO_4^{2-}), on the other hand, are present in concentrations closer to those noted in the batch leaching experiments and do not decrease significantly as more water passes through the system. The S values may

show a marginal decrease but the Ca concentrations actually increase. Saturation with respect to $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ is suspected.

Trace elements showing several orders of magnitude increase from batch to column leaching are B, Cr_{Total} , Li and Mo. These elements are readily leached and the anionic species for B, Cr_{Total} and Mo are soluble at the pH values encountered.

Other elements are recorded at concentration levels comparable with the initial batch leaching and concentrations either decrease, but proportionally less than elements such as Na and K, or remain approximately

Table 3. Continued.

	Li	Mn	Ni	Pb	Mo	Sr	V	Zn	As	Hg	Se	Si	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
dfd1-1	42.97	0.02	0.05	0.20	183.21	1.79	1.53	0.06	0.82	0.08	2.21	6.83	1122.0	93.9	1609.3
1-2	17.29	0.01	0.02	0.05	28.38	1.41	0.76	0.01	0.23	0.03	0.41	4.50	127.6	39.0	1506.2
1-3	8.42	0.01	0.02	0.07	6.62	1.37	0.49	0.04	0.08	0.02	0.12	3.37	21.4	15.9	1544.6
1-4	5.18	0.01	0.02	0.04	2.54	1.39	0.41	0.01	0.04	0.01	0.03	2.64	27.7	33.3	1078.0
1-5	3.78	0.00	0.02	0.03	1.71	1.38	0.39	0.01	0.08	0.02	0.07	2.53	10.6	29.8	1772.2
1-6	3.07	0.00	0.00	0.02	1.38	1.47	0.39	0.01	0.08	0.01	0.07	2.55	17.1	24.6	1573.2
1-7	2.45	0.00	0.01	0.07	0.65	1.41	0.26	0.24	0.01	0.07	nd	2.34	15.4	16.7	1446.8
1-8	2.14	0.00	0.07	0.25	0.64	1.45	0.38	0.03	0.00	nd	nd	2.40	10.5	19.1	1495.5
1-9	1.98	0.00	nd	0.17	0.80	1.48	0.29	0.01	0.00	nd	nd	2.49	5.9	6.2	1504.2
1-10	1.77	0.00	0.00	0.10	0.69	1.48	0.33	nd	0.07	nd	nd	2.40	10.3	7.7	1427.4
1-11	1.57	0.00	0.00	0.02	0.57	1.48	0.38	nd	0.15	nd	nd	2.31	6.8	4.3	1460.2
1-12	1.46	0.00	0.01	0.29	1.04	1.48	0.43	0.02	0.35	nd	0.30	2.40	2.5	3.2	1413.5
1-13	1.28	0.00	nd	0.09	0.97	1.50	0.39	nd	0.09	nd	0.06	2.39	2.2	1.2	1423.9
dwd3-1	0.24	0.02	0.04	0.14	0.64	6.94	0.28	0.37	0.29	0.01	0.55	8.87	1661.7	1224.7	3213.9
3-2	0.15	0.00	0.00	0.02	0.62	2.02	0.23	0.17	0.24	0.01	0.22	8.66	295.4	1056.0	676.5
3-3	0.14	0.00	0.03	0.02	0.38	0.50	0.19	0.10	0.23	0.00	0.13	7.41	54.3	167.2	138.4
3-4	0.10	0.00	nd	0.05	0.24	0.44	0.20	0.09	0.25	0.01	0.10	6.70	48.0	149.0	174.4
3-5	0.07	0.00	0.01	0.01	0.26	0.30	0.19	0.07	0.27	0.01	0.05	5.69	3.8	-	63.7
3-6	0.06	0.00	0.02	0.01	0.21	0.26	0.20	0.08	0.28	0.00	0.11	5.94	11.1	58.0	101.2
3-7	0.07	0.00	0.00	0.10	0.30	0.26	0.22	0.09	0.28	nd	0.03	5.84	3.5	8.5	75.5
3-8	0.05	nd	0.01	nd	0.27	0.24	0.07	0.07	0.18	nd	nd	5.44	4.8	6.6	61.5
3-9	nd	nd	nd	0.01	nd	0.26	0.12	0.05	0.17	nd	nd	5.37	6.5	8.3	57.3
3-10	0.05	nd	nd	0.06	0.03	0.23	0.13	0.03	0.16	nd	nd	4.98	2.2	6.4	42.5
3-11	0.05	nd	0.00	0.08	0.30	0.23	0.10	0.08	0.19	nd	0.07	5.13	2.4	4.4	35.7
3-12	0.06	nd	nd	0.17	0.23	0.21	0.13	0.04	0.13	nd	nd	5.05	1.3	4.2	27.4
mwd6-1	0.05	0.01	0.03	0.04	0.23	1.65	0.11	0.29	0.10	0.02	2.22	9.34	32.2	148.3	677.7
6-2	0.03	0.00	0.03	0.03	0.21	1.39	0.10	0.15	0.06	0.01	2.45	8.18	29.0	181.3	417.7
6-3	0.06	0.00	0.07	0.02	0.27	1.36	0.13	0.19	0.13	nd	2.08	6.41	120.5	18.2	354.1
6-4	0.04	0.00	nd	0.13	0.03	1.03	0.06	0.09	0.21	nd	1.70	5.77	10.3	47.0	267.0
6-5	0.02	0.00	nd	0.05	0.13	0.71	0.06	0.05	0.09	nd	0.21	5.55	4.5	16.9	162.6
6-6	0.03	0.00	nd	0.19	0.03	0.76	0.05	0.08	0.19	nd	nd	5.32	7.5	11.3	150.3
6-7	0.03	0.00	nd	0.21	0.09	0.60	0.07	0.06	0.07	nd	nd	5.25	1.7	4.5	34.8
6-8	0.01	0.00	nd	0.25	0.03	0.50	0.04	0.07	0.18	nd	nd	4.86	0.9	6.2	73.4
6-9	0.03	0.00	0.00	nd	nd	0.49	0.08	0.06	0.06	nd	nd	4.95	3.6	2.4	61.8
6-10	0.02	0.00	nd	0.15	0.00	0.46	0.01	0.05	0.15	nd	0.45	4.97	2.2	2.6	56.5
6-11	0.01	0.00	nd	0.24	0.07	0.44	0.01	0.03	nd	nd	nd	4.70	3.8	2.0	50.2
6-12	0.04	nd	nd	0.32	0.13	0.38	0.00	0.04	0.16	nd	nd	4.02	4.0	3.7	44.0
6-13	0.01	nd	nd	0.01	0.07	0.39	0.06	0.01	0.08	nd	nd	4.08	8.0	1.9	39.2

constant. In the latter category are Fe, Al, Ba and Sr and equilibrium reactions are indicated.

Column Leaching of Weathered PFA, Drax and Meaford, with Deionised Water

The analyses (dwd 3-1~3-12 and mwd 6-1~6-13, Table 3) show in general lower initial concentrations of the readily soluble elements and of Na and K in particular, which is to be expected as these are weathered samples. Na and K concentrations decrease as more water passes through the column, but with the K eventually achieving a near-constant concentration.

Compared with the fresh PFA B, Cr_{total}, Li, and Mo

are all present in much lower concentrations. This is essentially due to the removal of the readily accessible elements from the PFA during natural weathering. The PFA samples were not dried before the columns were packed and the initial concentrations recorded should be approaching those of the natural porewaters. Concentrations fall during the leaching experiment as the porewaters are progressively diluted. How effective dilution is may seem in the NO₃⁻ concentrations in the weathered PFA from Drax (Sample dwd analyses 3-1~3-12 Table 3). The high NO₃⁻ concentrations compared with the fresh PFA from Drax are probably derived externally from fertilizers (Lee and Spears, 1995). The leachate concentrations for NO₃⁻ decrease by a factor

Table 4. Analytical results of leachate of column leaching test treated by synthetic leachate (Unit : mg/l unless indicate).

	pH	Eh(mv)	C(μ S/cm)	Ca	Na	K	Mg	Fe	Al	B	Ba	Cd	Co	Cr	Cu	Li
Syn. Lea.	4.55	9.10	3550	615.9	1032.2	734.6	87.0	122.96	0.12	0.23	0.04	121.18	0.16	79.78	117.6	19.44
dfs2-1	9.90	267.8	8740	830.4	6171.1	2650.7	76.6	0.203	0.92	49.46	0.12	0.04	0.05	2.98	0.58	54.05
2-2	9.70	250.8	5380	2362.0	2063.7	1351.1	476.9	0.206	0.29	28.54	0.30	0.04	0.06	2.98	0.01	27.31
2-3	9.49	230.1	5270	2567.6	1725.1	977.5	682.3	0.218	0.23	36.06	0.32	0.02	0.07	4.98	0.00	20.68
2-4	9.33	243.3	5110	2623.3	1644.2	867.1	767.0	0.235	0.24	56.27	0.30	0.02	0.08	14.65	0.01	18.41
2-5	9.37	245.9	5190	2647.9	1616.5	816.9	678.6	0.260	0.24	61.69	0.30	0.02	0.07	21.01	0.01	17.69
2-6	9.49	259.8	5160	3356.8	2046.9	1051.1	650.7	0.505	0.00	63.21	0.30	0.00	0.05	21.85	nd	24.62
2-7	9.53	224.1	5190	3307.6	1996.7	1038.6	585.1	0.131	0.34	51.15	0.29	0.00	0.00	20.45	nd	24.85
2-8	9.35	252.4	5130	3295.2	2034.8	1045.8	598.4	0.094	0.21	43.16	0.25	0.00	0.00	20.14	nd	25.09
2-9	9.32	236.3	5230	3274.3	2031.7	1029.7	593.7	0.116	0.16	40.74	0.26	0.00	0.00	19.33	nd	25.23
2-10	9.29	219.1	5340	3333.0	2102.9	1069.9	643.2	0.123	0.29	46.55	0.27	0.00	0.00	18.56	nd	26.46
2-11	9.36	220.5	5190	3234.2	2014.3	1027.9	616.2	0.094	0.29	51.51	0.26	0.00	0.10	16.18	nd	24.91
2-12	9.19		5240	3197.1	2022.7	1025.2	626.8	0.058	0.08	59.51	0.24	0.00	0.00	14.74	nd	24.74
2-13	9.06		5230	3113.2	1964.8	1016.2	648.0	0.058	0.10	66.32	0.24	0.00	0.00	13.98	nd	24.73
dws4-1	7.22	281.8	8020	1451.6	121.3	143.3	211.2	0.176	0.17	2.91	0.35	0.03	0.04	0.19	0.13	0.27
4-2	7.55	282.8	12120	2754.1	783.9	236.6	336.0	0.127	0.17	2.46	0.56	0.02	0.05	0.61	0.05	0.77
4-3	7.93	288.3	15700	3300.7	1563.4	606.4	216.0	0.096	0.09	2.94	0.99	0.01	0.04	10.31	0.05	13.31
4-4	7.32	274.7	14730	2980.2	1535.7	736.3	181.2	0.108	0.12	3.89	1.03	0.01	0.04	25.28	0.02	15.21
4-5	7.37	273.4	15200	3157.3	1539.6	710.6	200.4	0.184	0.17	3.70	1.03	0.02	0.05	24.30	0.03	14.74
4-6	6.68	267.7	5850	3088.6	1643.9	758.1	177.6	0.192	0.19	3.54	1.13	0.03	0.06	21.55	0.03	17.43
4-7	6.40	250.4	5030	3079.4	1727.3	796.6	178.8	0.184	0.17	3.75	1.17	0.02	0.08	25.76	0.03	18.49
4-8	6.49	248.2	4990	2827.3	1611.9	797.2	184.8	0.232	0.18	3.60	1.07	0.08	0.11	23.29	0.03	17.47
4-9	6.26	260.5	4960	2818.1	1705.7	820.3	195.6	0.246	0.21	3.45	1.17	18.78	0.16	22.07	0.03	18.49
4-10	6.03	261.8	5530	3194.6	1999.2	1001.7	226.7	0.409	0.13	3.75	1.23	74.58	0.12	21.24	0.00	23.27
4-11	5.89	263.1	4920	2994.2	1914.6	968.8	213.6	0.181	0.34	3.21	1.21	105.96	0.16	17.80	0.07	21.78
4-12	5.82	256.4	4800	2982.5	1908.7	968.8	212.3	0.276	0.26	3.25	1.16	117.79	0.18	16.67	0.06	22.10
4-13	5.83	239.8	4960	3010.0	1945.9	986.7	207.7	0.218	0.45	2.92	1.16	127.78	0.18	16.85	0.09	22.27
mws5-1	7.12	262.0	3320	1930.3	919.6	358.8	202.8	0.230	0.15	1.48	0.84	0.02	0.04	5.88	0.01	7.19
5-2	7.08	246.9	5080	2866.1	1557.3	765.2	194.4	0.224	0.19	2.20	1.05	0.01	0.07	17.61	0.02	16.38
5-3	7.27	224.9	4990	4015.2	2325.9	1186.9	282.1	0.183	nd	3.92	1.03	2.74	nd	20.97	0.08	26.70

of approximately 300. This is greatly in excess of the decreases noted for B, Cr_{total}, Li and Mo, which all decrease by a factor of less than 10. The explanation for this is continued reaction of the PFA releasing these and other elements into solution. Although close to detection limits in a number of cases, for example Cd, Co, Cu and Hg, a range of trace elements are detected in the leachates. Concentrations are low and are mainly in the parts per billion range. Nevertheless, they are detected in solution and the rate of concentration decrease with flow through the system indicates that there is dissolution of the ash during the course of the experiment. For example, As falls from 0.29 mg/l to 0.13 mg/l (dwd 3-1~12, Table 3), from the initial to the final leachate.

Solution concentrations are reflected in the electrical conductivity values (Table 3). All the values for the leachates from the weathered PFA samples are lower than the equivalent sample from the fresh PFA and with few exceptions the differences are significantly large. Related to this the pH values for the weathered PFA samples average 8.26 whereas the values for the fresh PFA average 9.27. The solubilities of the anionic species

of B, Cr_{total} and Mo will be effected but this does not account for the difference between weathered and unweathered samples, which arises because the readily leached elements in the fresh PFA are largely removed during natural weathering.

Column Leaching of Fresh Drax PFA with Simulated Leachate

The initial leachate has similar concentrations of Ca, Na, K and Mg (Table 4) to that of the deionised water with the fresh PFA. Na and K concentrations fall as more solution passes through the column until the concentrations of the simulated leachate are achieved. Ca and Mg concentrations, on the other hand, increase to values in excess of those in the simulated leachate as expected from the behaviour with water, but values are also much higher than in the water leaching. There is greater dissolution in the synthetic leachate, but this would not appear to be simply related to pH, because the pH of the emergent solutions are in the 9.9~9.1 range. Greater dissolution is also noted for a number of trace elements

Table 4. continued.

	Mn	Ni	Pb	Si	Sr	V	Zn	As	Hg	Se	Mo	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
Syn. Lea.	0.18	109.40	0.22	0.81	0.16	0.11	107.60	0.27	13.85	0.33	0.68	1973.49	20.67	419.25
dfs2-1	0.01	0.05	0.14	10.11	4.16	2.30	0.04	0.71	0.11	1.98	156.94	1882.9	86.1	44057.7
2-2	0.02	0.06	0.17	5.57	9.90	0.63	0.04	0.42	0.09	0.56	19.85	2167.8	77.5	864.7
2-3	0.02	0.07	0.15	4.67	9.82	0.54	0.04	0.48	0.08	0.50	8.74	2408.1	44.6	994.9
2-4	0.02	0.06	0.18	4.54	8.39	0.56	0.03	0.50	0.07	0.43	4.37	2298.3	24.6	518.8
2-5	0.02	0.06	0.19	4.19	7.23	0.53	0.03	0.46	0.09	0.35	2.60	2672.4	26.5	4617.8
2-6	0.01	0.00	0.26	6.24	7.07	0.47	0.07	0.02	0.00	0.11	1.69	2325.1	62.7	1410.8
2-7	0.02	0.07	0.29	5.98	6.56	0.43	0.02	0.11	0.00	0.11	1.01	2847.6	31.5	1306.7
2-8	0.02	0.00	0.39	5.91	6.44	0.42	0.01	0.31	0.00	0.49	0.97	2389.4	37.0	1282.4
2-9	0.01	0.02	0.06	5.83	6.19	0.54	0.01	0.41	0.00	0.00	1.47	2311.4	44.0	1259.2
2-10	0.02	0.05	0.38	5.97	6.32	0.48	0.01	0.39	0.00	0.52	1.54	2391.5	18.0	1358.2
2-11	0.01	0.00	0.32	5.82	6.11	0.39	0.01	0.52	0.05	0.00	1.31	3349.4	11.1	1340.3
2-12	0.01	0.00	0.11	5.39	6.12	0.38	0.01	0.28	0.00	0.15	1.44	1765.7	7.1	1302.6
2-13	0.02	0.00	0.23	5.58	6.01	0.39	0.00	0.21	0.00	0.00	1.14	3342.5	18.1	1323.1
dws4-1	0.02	0.05	0.14	12.95	6.32	0.45	1.22	0.51	0.03	0.71	0.36	1054.7	1084.0	863.7
4-2	0.02	0.05	0.18	15.05	11.43	0.57	0.40	0.72	0.05	0.68	0.40	1880.9	1796.1	794.7
4-3	0.02	0.05	0.14	13.17	14.70	0.56	0.18	0.89	0.12	0.66	0.50	2209.4	249.4	635.1
4-4	0.04	0.05	0.16	15.07	13.30	0.56	0.17	0.81	0.08	0.49	0.48	2003.6	279.2	572.3
4-5	0.07	0.13	0.16	17.65	12.40	0.62	0.19	0.84	0.08	0.55	0.32	2326.0	173.1	782.7
4-6	0.10	12.38	0.18	18.94	10.19	0.62	0.21	0.87	0.07	0.43	0.42	4324.5	65.9	555.0
4-7	0.16	52.05	0.18	20.63	9.15	0.65	0.31	0.94	0.09	0.49	0.57	2135.5	23.7	478.0
4-8	0.45	90.97	0.18	21.01	8.64	0.61	0.46	0.92	0.08	0.43	0.73	2411.5	30.0	500.4
4-9	4.64	130.21	0.21	26.71	8.55	0.62	0.56	1.03	0.10	0.47	0.91	2702.4	28.7	645.5
4-10	9.09	136.10	0.49	34.36	7.88	0.54	10.19	0.69	0.00	0.00	0.12	2459.8	22.7	588.6
4-11	11.20	130.08	0.48	34.62	6.81	0.58	45.46	0.83	0.00	0.04	0.27	3339.5	7.9	536.5
4-12	11.79	127.43	0.19	34.01	6.36	0.32	82.12	0.51	0.02	0.30	0.34	3255.2	15.5	550.7
4-13	11.65	124.69	0.23	34.58	6.25	0.47	107.55	0.53	0.00	0.09	0.57	2004.3	4.1	517.9
mws5-1	0.02	0.08	0.09	8.72	16.24	0.24	0.24	0.34	0.04	1.67	0.29	1288.7	56.1	455.9
5-2	0.03	0.07	0.17	8.72	23.96	0.30	0.08	0.47	0.07	0.46	0.29	2802.9	29.5	426.8
5-3	0.14	0.93	nd	15.35	29.91	0.50	2.80	0.02	nd	nd	nd	2678.8	41.6	404.8

in the synthetic leachate, such as B and Ba.

The concentrations of Fe in the synthetic leachate is 100 mg/l (Table 1), but there is very little Fe present in the solutions to emerge from the column. Also present in the synthetic leachate and removed in the column are Cd, Cu, Ni, Zn and Hg. The concentrations of total Cr_{total} is reduced from synthetic leachate to final solution. Although there is some retention of Cr_{total} this is only partial, which is consistent with the solubility of the anionic species.

Elements present in significant concentrations in the synthetic leachate are removed from solution in passing through the fresh PFA. The pH of the leachate is 4.55 whereas the emergent solution is alkaline and this is an important factor in metal retention.

Column Leaching of Weathered PFA with Simulated Leachate

The analyses (Table 4) show a similar pattern with

flow through the column for Ca, Mg, Na and K in the weathered PFA as in the fresh PFA. The initial solution to emerge from this column is influenced by porewater concentrations within the column. Na and K concentrations vary with time and volume passed towards the leachate composition, whereas Ca and Mg are enhanced by reaction between leachate and PFA. Elements present as trace constituents in the leachate, such as B, are recorded in the emergent solution in much lower concentrations compared with the fresh PFA, demonstrating the effect of weathering, but in higher concentrations than in the water leaching demonstrating the greater dissolution achieved with the synthetic leachate.

The Fe concentrations in the emergent solution (Table 4) are uniformly low and demonstrate retention in the column. Cd, Cu, Ni, Zn and Hg are also retained but to differing degrees. Concentrations for all these elements are initially very low as the synthetic leachate passes through the column. The capacity of the column to retain all of the Cd, Ni and Zn is however exceeded

Table 5. Comparisons of PFA leachate with standards for drinking water and landfill leachate quality.

	Drinking Water quality		PFA leachate			Landfill Leachate Quality	
	CEC*	WHO*	Fresh Drax	Weathered Meaford	EC Landfill Directive	Barlow Consent (NRA)	
	GL* (mg/l)	MAC* (mg/l)	GV* (mg/l)	(mg/l)	(mg/l)		
Calcium (Ca ²⁺)	100	-	-	353	225		
Magnesium (Mg ²⁺)	30	50	-	37	29		
Sodium (Na ⁺)**	20	150	200	5760	32		
Potassium (K ⁺)	10	12	-	2113	42		
Chloride (Cl ⁻)	25	-	250	1120	32	1200-6000	
Sulphate (SO ₄ ²⁻)	25	250	400	1610	670	200-1000	
Aluminium (Al ³⁺)	0.05	0.5	0.2	0.99	0.07		
Nitrate (NO ₃ ⁻)**	25	50	45	94	150		
(as N)	5.65	11.3	10				
Nitrite (NO ₂ ⁻)**	-	0.1	-				
Ammonium (NH ₄ ⁺)	0.05	0.5	0.2				
Iron (Fe ³⁺)	0.05(Fe _T)	0.2(Fe _T)	0.3(Fe _T)	0.17	0.12		1.0
(Fe ₂ ⁺)	-	-	-				
Manganese (Mn ²⁺)	0.02	0.05	0.1	0.02	0.01		
Copper (Cu ²⁺)	3000	-	1000	140	30	2000-10000	100
Zinc (Zn ²⁺)	5000	-	5000	60	290	2000-10000	1000
Phosphate (HPO ₄ ²⁻)	400	5000	-				
Fluoride (F ⁻)**	8~12°C	1500	1500				
	25~30°C	700	-		1		
Barium (Ba ²⁺)	100	-	-	80	130		
Silver (Ag ⁺)	-	10	-				
Arsenic (As)**	-	50	50	820	100	200-1000	10
Cadmium (Cd ²⁺)	-	5	5	50	20	100-500	5
Chromium (Cr)**	-	50	50	24000	110	100-500	100
Mercury (Hg)**	-	1	1	80	20	20-100	
Nickel (Ni)	-	50	-	50	30	400-2000	500
Lead (Pb ²⁺)**	-	50	50	200	40	400-2000	500
Antimony (Sb)	-	10	-	420	30		
Selenium (Se)**	-	10	10	2200	2200		
Boron (B)	1000	-	-	72000	1890		5000
Temperature (°C)	12	25	-				
pH	6.5-8.5	-	6.5-8.5	9.3	8.5	4-13	6-9
Conductivity (μS/cm at 20°C)	400	-	-	22000	500		

*CEC : Council of the European Communities Directive 80/778

*GL : Guide level

*MAC : Maximum admissible concentration

*WHO : World Health Organisation. Guidelines for drinking water quality. 1984.

*GV : Guide value

**Inorganic constituents of health significance (WHO, 1984). [additionally : asbestos, vanadium, beryllium, nickel, silver, - no guide value set.]

and these elements achieve a "break-through" with concentrations increasing towards the synthetic leachate values. There is partial retention of Cr_{Total} in the column. Although Cr_{Total} appears in the emergent solution at a relatively early stage concentrations do not increase towards the leachate value as is the case with Cd, Ni and Zn.

Elements are lost from the synthetic leachate when passed through columns of both the fresh and weathered PFA. A "break-through" was achieved for Cd, Ni and

Zn in the weathered PFA but not in the fresh PFA. The capacity of the fresh PFA to extract elements from solution would therefore appear to be greater than that of the weathered PFA. A pH control on precipitation could be very important with solutions emerging from the fresh PFA in the 9.9~9.1 range whereas in the weathered PFA from Drax the pH range is 7.9~5.8. The fresh PFA with the readily soluble ions has a greater capacity to neutralise the acidic synthetic leachate than does the weathered PFA. The secondary minerals in the

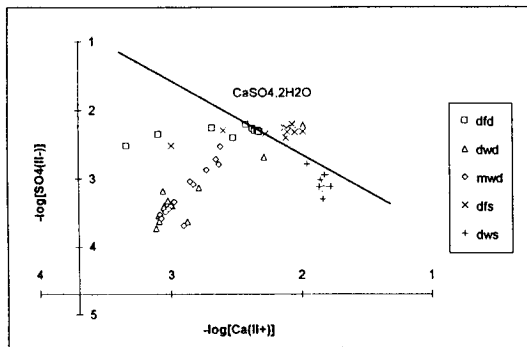


Fig. 2. Plot of calculated activities of Ca and SO_4^{2-} in leachates along with measured activity of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ (solid line).

weathered PFA could be important, but their influence would appear to be overshadowed by changes in the pH of the system. In the column leaching test the PFA sample to solution column is large hence the influence of pH. In the batch leaching the solution volume is large compared with the sample mass and there is little modification to the pH of the synthetic leachate (The pH values recorded were fresh Drax pH=4.80, weathered Drax=4.75 and weathered Meaford=5.00). Under these conditions of near constant pH the weathered PFA does demonstrate a greater potential for element extraction.

Comparison of Column Leaching Results with Water Quality Standards

The column leaching tests demonstrate the release of elements into solution and therefore the potential contribution that PFA in landfill sites might make to the water quality of surface and underground supplies. Recommended water quality concentrations for drinking water are given in Table 5. Also shown for comparison are column leachate analyses with water for fresh PFA from Drax (sample dfd 1-1, Table 3) and weathered PFA from Meaford (sample mwd 6-1, Table 3). In both cases the analyses selected are of the initial leachate to emerge from the column. These have the maximum concentrations and therefore give an indication of worst case scenarios. In the leachate produced from the fresh PFA most elements exceed recommended values and in some cases, such as Cr_{total} , Se and B, the margin is considerable. Major dilution is therefore required if all recommended values are to be achieved. In landfill sites there will be dilution from surface run-off for leachate draining into streams and rivers. Leachates passing into aquifers would also be diluted, but possibly not to the same extent. This would however, be a function of the

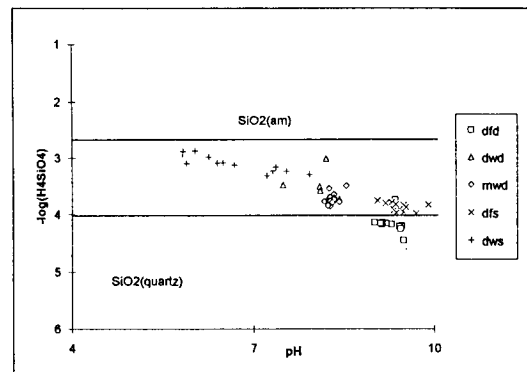


Fig. 3. Plot of calculated activity of H_4SiO_4 vs. pH with measured activity of amorphous and crystalline quartz (solid lines).

particular aquifer.

The leachate emerging from the column of fresh PFA after a period of time provides evidence of how PFA will behave as it is progressively weathered. The volume of flow can be related directly to rainfall and infiltration values, enabling the weathering behaviour over several years to be predicted. This does assume that the reactions in the field are duplicated in the columns, which need not follow because of the differing time scales. More realistic is to use weathered PFA and again the initial leachate gives an indication of possible water quality. Most elements in the leachate are present at concentrations which exceed maximum permissible concentrations but by a relatively small factor, with the exception of Se. Excessive dilution would not therefore be required to achieve drinking water standards.

Finally, on Table 5, are given a quality directive for landfill leachate and the NRA consent for the Barlow disposal mound at Drax power station. Elements in the leachate from the fresh Drax PFA which exceed the directive concentrations are SO_4^{2-} and Cr_{total} , falling within the range are As and Hg and below limits are Cu, Zn, Cd, Ni, and Pb. Elements in the leachate from the weathered PFA do not exceed the directive limits but SO_4^{2-} , Cr_{total} and Hg are within range and the other elements fall below. The Barlow consent values are lower than the landfill directive and comparable with drinking water standards. As, Cr_{total} , and possibly Cd in the leachate from weathered ash are higher than the consent values and leachate from the fresh ash would require significant dilution to achieve consent values.

Equilibrium Relationships in Leachates

In the previous discussion it was noted that Ca and

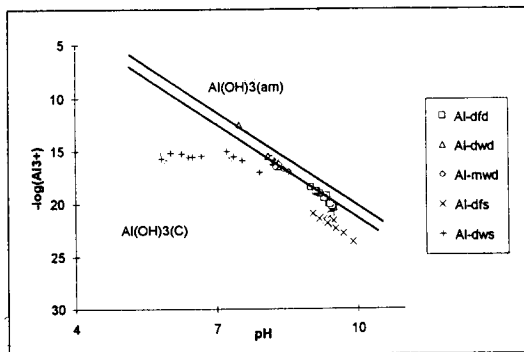


Fig. 4. Plot of calculated activity of Al^{3+} against pH with measured activities of Al^{3+} (solid lines) which are in equilibrium with amorphous and crystalline $\text{Al}(\text{OH})_3$.

SO_4^{2-} concentrations suggested an equilibrium with respect to gypsum. This and other possibilities have been investigated by equilibrium calculations, using a computer programme, WATEQ4F (Ball *et al.*, 1989).

The activities of Ca and SO_4^{2-} are plotted on Fig. 2 for all the leachates. Saturation with respect to gypsum is indicated by the line. Ca has been predicted or observed to occur in many different minerals in fresh and weathered ashes (Warren and Dudas, 1985; Fruchter *et al.*, 1988). Leachate samples generated using water do achieve saturation with respect to gypsum for the fresh PFA, but some samples are undersaturated. For the weathered PFA leachate samples are undersaturated, although there would appear to be a trend towards saturation. The leachates generated using the simulated landfill leachate are close to saturation, with the fresh PFA producing mainly oversaturated solutions. There is enhanced Ca dissolution compared with the water leachate.

The analysis of the anions in the leachate did not include HCO_3^- because precautions were not taken to exclude atmospheric CO_2 from the columns and the leachate. As a first approximation the HCO_3^- has been calculated as the difference between cation and anion totals. Using this derived value the leachates were found to be oversaturated with respect to calcite.

The concentrations of silica in equilibrium with quartz and amorphous silica are shown on Fig. 3. The PFA contains quartz and leachate concentrations would be anticipated to be at least saturated with respect to quartz. Although this is the case for most leachates there is a trend with pH values from which it may be concluded that the silica activities are not controlled by quartz or amorphous silica equilibrium either in dissolution or precipitation reactions. The trend with pH could be due

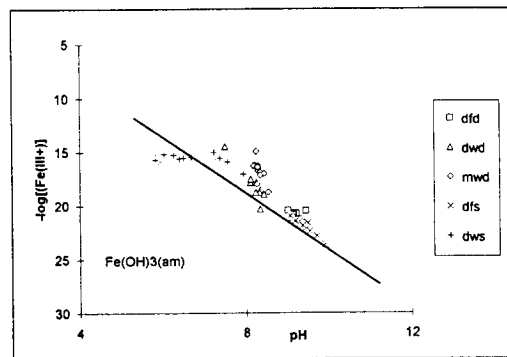


Fig. 5. Plot of calculated activities of Fe against pH, along with Measured activities of Fe in equilibrium with amorphous $\text{Fe}(\text{OH})_3$.

to kinetic factors. Both quartz and amorphous silica do take a finite time to achieve equilibrium in solution, which may exceed the time available in the column experiments.

Activity values of aluminium are plotted against pH on Fig. 4. The leachate produced using distilled water all lie on or very close to the $\text{Al}(\text{OH})_3$ solubility line. The simulated leachate samples fall below the solubility line but the trend is similar with most values showing a pH trend. It may be that the solution is saturated with respect to $\text{Al}(\text{OH})_3$ but the equilibrium calculations are not adequately modelling the more complex, organic acid rich synthetic leachate. This result agrees with the results of previous workers, such as Roy and Griffin (1984) and Fruchter *et al.* (1988).

On Fig. 5 is shown the activity of iron (Fe^{3+}) plotted against pH. There is again a trend with pH and all the samples plot close to $\text{Fe}(\text{OH})_3$ solubility line, but with some oversaturation. This discrepancy is thought to be due to the presence of some colloidal $\text{Fe}(\text{OH})_3$, which are not retained by $0.45 \mu\text{m}$ filter (Stumn and Morgan, 1981). In the water leachates iron originates entirely from the dissolution of the PFA, as do Si and Al considered above. Unlike these two elements, however, iron is a major component of the synthetic leachate and is retained to an important degree in the PFA column. Fig. 5 demonstrates that precipitation as $\text{Fe}(\text{OH})_3$ is the probable reaction and that pH is indeed important in metal retention within the column. $\text{Fe}(\text{OH})_3$ has been reported as a solubility controlling solid phase in PFA-water reaction (Ainsworth and Rai, 1987; Fruchter *et al.*, 1988).

CONCLUSIONS

1. When solutions containing heavy metals are passed

through columns of PFA metals are removed from solution, this confirming the findings of the batch leaching tests.

2. In column leaching the PFA is able to exert an important influence of the pH of the emergent leachate. The pH is an important control on the element retention in the column based on equilibrium calculations.

3. The order of element retention by the PFA is fresh Drax PFA > weathered Drax PFA. In batch leaching however, the reverse order was observed with the most weathered PFA (Meaford) having the greatest capacity for element retention. It is postulated that this is due to reaction with the secondary phases produced during weathering. In batch leaching the ratio of solution volume to PFA mass is large and hence the pH remains relatively constant during the course of the experiment.

4. Differential dissolution is observed as solutions pass through the columns and the distribution of elements in the PFA may be inferred. The weathered PFA has reduced concentrations of elements associated with fine particles and surfaces but nevertheless the PFA is not inert and elements are leached into solution.

5. The initial leachate concentrations generated with deionised water give an indication of porewater concentrations. The leachate from fresh PFA is considerably in excess of drinking water guide-line concentrations, as is to be expected, but the weathered PFA also generates a leachate which would require some dilution to achieve permissible concentrations for a number of elements.

6. Batch and column leaching provide valuable information on the potential behaviour of PFA in the natural environment. Column leaching more accurately reflects the natural system but does not eliminate the need for

field based observations, particularly as there may be kinetic constraints of some of the reactions in the columns. A comparison of the leaching work with the parallel study of porewaters in PFA disposal mounds should be of particular relevance.

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풍화 석탄연소 고�형폐기물(Pulverised Fuel Ash)의 중금속 제거가능성 :

II. 주사용출실험

이 상 훈

요 약 : 석탄연소 폐기물 (Pulverised Fuel Ash: PFA) 을 17년, 40년 매립 풍화받은 것 그리고 현재 발생하는 신선한 시료등으로 구분 영국내 2개 화력발전소로부터 각각 채취하여 주사용출실험 (Column leaching test)을 실시하였다. 용매는 증류수와 인공합성 침출수의 2가지를 이용하였다, 전자의 경우 풍화 시료의 용출행태를 주로 알아보고자 하였으며 후자의 경우 PFA시료와 용매간의 반응중에 발생하는 2차물질형성이 폐수처리중 중금속제거에 활용가능성이 있는가를 알아보고자 함이었다. 신선한 시료로부터 매우 높은 함량의 원소방출이 관찰된다. 이는 Ca, Na, K, B, Cr, Li, Mo, Se, 그리고 SO_4^{2-} 등을 포함한다. 이는 원소들의 분포위치에 관한 정보를 제공하며 이들원소의 입자 표면 수반을 시사한다. 풍화 받은 시료로부터 채취된 용액시료에서는 신선한 시료와는 달리 원소들의 급격한 초기방출은 관찰되지 않으며 비교적 일정함량을 보이며 매우 완만한 농도감소 감소현상을 보인다. 이는 17년과 40년이라는 매립기간중 표면의 용융성원소들의 방출을 의미하며 입자를 주로 구성하는 유리질물질에 수반되는 부분이 이들 시료에서의 원소방출에 중요한 역할을 하는 것으로 생각된다. 증류수를 이용한 실험결과를 여러가지 침출수배출기중, 음용수기준치등과 비교시 PFA는 이들 기준에 초과하는 농도를 나타내는바 침출수배출시에는 회석등의 처리가 필요할 것으로 생각되며 매립시 침출수가 지하수로 유입되지 않도록 하여야할 것으로 보인다. 인공 침출수가 PFA기등을 통과하며 많은 중금속 - B, Fe, Zn, Hg, Ni, Li Mo - 등이 침출수로부터 제거되며 이 제거정도는 신선한 Drax > 풍화 Meaford 시료의 순이다. 물분석시료를 지구화학 모델링 프로그램인 WATEQ4F를 이용하여 PFA 침출수내의 농도조정물질의 존재여부를 알아본 결과 Ca의 경우 $CaSO_4 \cdot 2H_2O$, Al의 경우 $Al(OH)_3$, Fe는 $Fe(OH)_3$ 등이 PFA침출수내에 각 해당원소의 농도를 제한하는 2차물질로 판단되었다.