The atmospheric carbon capture performance of legacy iron and steel waste

SUPPORTING INFORMATION

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Pages: 21

Figures: 8

Tables: 6

Mineral Name	Formula	Mineral Group	CN*	Notes
Silicates				
Dicalcium silicate ¹⁻⁴⁴	Ca ₂ SiO ₄	Olivine	C_2S	Includes β - (larnite) and γ - (belite) phases
Tricalcium silicate ^{2, 8, 12, 16, 19-25, 27, 30, 33-35, 37-40, 42, 45}	Ca ₃ SiO ₅		C_3S	Includes alite and haturite
Akermanite ^{2, 13, 18, 21, 36, 43, 46, 47}	$Ca_2MgSi_2O_7$	Melilite	C_2MS_2	
Merwinite ^{1, 14, 17, 18, 21, 32, 33, 35, 37, 40, 42, 43, 47, 48}	$Ca_3Mg(SiO_4)_2$		C_3MS_2	
Gehlenite ^{2, 11, 13, 14, 18, 21, 26, 28, 31, 33-35, 37, 39, 40, 43, 44, 46-50}	$Ca_2Al_2SiO_7$	Melilite		
Quartz ^{1, 13, 15, 21, 34, 39, 48, 50-52}	SiO ₂	Quartz		
Christobalite ^{51, 52}	SiO ₂	Quartz		
Fayalite ^{3, 9, 31, 38, 52}	Fe_2SiO_4	Olivine		Includes divalent substitution
Kushiroite ⁴⁹	CaAl ₂ SiO ₆	Pyroxene	MS	
Enstatite ^{2, 3, 12, 21, 38, 42, 48, 53}	MgSiO ₃	Pyroxene		
Wollastonite ^{3, 9, 17, 25, 42, 54}	CaSiO ₃	Pyroxenoid	CS	
Ferrosilicate ³	FeSiO ₃	-		
Ferrosilite ^{9, 52}	$Fe_2Si_2O_6$	Pyroxene		Includes divalent cation substitution
Calcium magnesium aluminium iron silicate14	Ca ₂ MgOAlFeSiO ₅	-		
Clinoenstatite ⁹	Mg ₂ Si ₂ O ₆			
Diopside ^{21, 30, 42, 49, 50}	MgCaSi ₂ O ₆	Pyroxene	CMS_2	
Forsterite ³⁸	Mg ₂ SiO ₄	Olivine	-	
Monticellite ^{17, 21, 40, 42, 43}	CaMgSiO ₄		CMS	
Cuspidine ¹⁴	$Ca_4Si_2O_7F_2$	Sorosilicate		
Mullite ⁵²	$Al_6Si_2O_{13}$			
Plagioclase ⁵²	CaAl ₂ Si ₂ O ₈ -NaAlSi ₃ O ₈			
Bredigite ^{20, 21, 28-30}	$Ca_{12}Al_{14}O_{33}Ca_{14}Mg_2(SiO_4)_8$			
Alkali feldspar ⁵²	KAlSi ₃ O ₈	Feldspar		
Anorthite ^{31, 37, 43}	CaAl ₂ Si ₂ O ₈	Plagioclase		
Iscorite ³¹	Fe ₇ SiO ₁₀	e		
Dicalcium iron magnesium silicate ³³	$Ca_2Fe_{1,2}Mg_{0,4}Si_{0,4}O_5$			
Zircon ⁵²	ZrSiO ₄	Zircon		
Andradite ⁵²	$Ca_3(FeTi)_2Si_3O_{12}$	Garnet		
Rankinite ⁴²	Ca ₃ Si ₂ O ₇		C_3S_2	
Oxides	5 2 1		, 2	
Periclase ^{2, 3, 8, 13-15, 17, 19, 21, 27, 30, 32, 34-39, 42}	MgO			
Lime1, 3, 5, 8, 9, 15-17, 19-21, 24, 27, 31, 32, 38, 42, 55, 56	CaO			
Wüstite ² , 3, 5, 6, 8-10, 12, 14, 15, 19-21, 24-26, 29, 31, 34, 36, 38, 39, 42, 44, 45, 53, 55	FeO			
Corundum ^{2, 4, 9, 12, 18, 29, 31, 36}	α -Al ₂ O ₃			
Hematite ^{1, 3, 8, 17, 23, 24, 31, 33, 52, 56}	Fe ₂ O ₃	Iron oxides		
Magnetite1-4, 12-15, 18, 20, 21, 26, 28, 29, 34, 35, 38-40, 42, 44, 53	Fe ₃ O ₄	Iron oxides		Includes maghemite
Spine ^{[27, 30, 35, 48, 50, 52}	MgAl ₂ O ₄			Includes multivalent substitution
Magnesioferrite ^{20, 28, 37}	MgFe ₂ O ₄		MF	includes individual substitution
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Table S1. Common minerals in iron and steel slag samples as identified in academic literature

Calcium ferrite ^{1, 3, 5, 7-9, 14-20, 23, 24, 27, 32-38, 41, 42, 45, 55}	Ca ₂ Fe ₂ O ₅ (srebrodolskite) or CaFe ₂ O ₄		CF	Includes multivalent cation substitution
Hercynite ¹⁴	FeAl ₂ O ₄			
Brownmillerite ^{6, 12, 21, 39, 40, 56}	Ca ₂ FeAlO ₅		C_4AF	
Manganese oxides ^{14, 28, 31}	MnO ₂ or Mn ₃ O ₄			
RO phases (mixed oxides) ^{4, 7, 10, 12-14, 19, 21, 24, 27, 32, 35-38, 42, 52, 53, 55}	Various solid solutions			
Calcium aluminium oxides ^{2, 13, 14, 20-22, 34, 35, 38, 39, 42, 44}	CaAl ₂ O ₃ or Ca ₃ Al ₂ O ₆ or Ca ₁₂ Al ₁₄ O ₃₃ (mayenite)			
Hydroxides				
Portlandite1, 2, 4, 5, 7, 9, 10, 12, 14, 15, 17, 21-24, 27, 30, 37, 53, 55, 57	Ca(OH) ₂		СН	
Brucite ^{2, 4, 9}	Mg(OH) ₂		MH	
Goethite ⁵²	FeOOH			
Carbonates				
Calcite ^{1, 2, 4, 5, 7, 11, 12, 15, 17, 20, 23-25, 27, 37, 38, 51-53, 58}	CaCO ₃			Includes Mn cation substitution
Aragonite ^{5, 53}	CaCO ₃			
Dolomite ^{17, 49, 51}	$CaMg(CO_3)_2$			
Magnesite ¹⁷	MgCO ₃			
Ankerite ⁵³	Ca(FeMgMn)(CO ₃) ₂			
Kutnohorite ⁵³	$Ca(MnMgFe)(CO_3)_2$			
Tilleyite ⁴⁰	$Ca_2Si_2O_7(CO_3)_2$			
Sulphides and sulphates				
Oldhamite ^{31, 47, 49, 52}	(CaMg)S			Includes divalent cation substitution
Gypsum/Anhydrite ^{11, 30}	CaSO ₄			
Iron sulphide ^{42, 52}	FeS			
Magnesium sulphate ¹⁷	MgSO ₄ ·5H ₂ O			
Ettringite ¹¹	$Ca_6Al_2OH_{12}(SO_4)_3$ ·26 H_2O			
Others				
Metallic iron ^{3, 5, 21, 38, 47, 49, 52, 55}	Fe	Metals		
Fluorite ^{13, 21}	CaF ₂	Halite		
Graphite ⁵²	С			
Whitlockite ³³	$Ca_3(PO_4)_2$	Phosphates		
Calcium dihydrogen phosphate ³³	$Ca(H_2PO_4)_2 \cdot H_2O$	Phosphates		

* - Cement Notation





Figure S1. Photographs of the drilling works on site at Consett



Figure S2. Photographs of the material recovered from Consett

6 S1. Details of sample analysis

7 S1.1 Preparation

All slag samples were oven dried at 105 °C. Particle size distribution (PSD) analysis on bulk 8 9 samples was performed using sieves in accordance with BS ISO 11277:2009. ⁵¹ To prepare for 10 X-ray diffraction (XRD), X-ray fluorescence (XRF), acid digestion elemental inductively 11 coupled plasma-optical emission spectroscopy (ICP-OES), total carbon (TC) and total organic 12 carbon (TOC) analysis, samples were pulverised using a Siebtechnik vibrating-puck mill with 13 a tungsten carbide grinding bowl. until a homogenous powder was obtained. Equipment was 14 washed using distilled water and acetone between samples. All powdered samples were stored 15 in plastic, sealable bags until analysed. To prepare for microscopy, selected samples were 16 mounted in resin blocks, before being ground and polished with a sol-gel alumina (Al_2O_3) 17 suspension to a 0.25 µm finish. Before scanning electron microscopy analysis, the samples 18 were coated with 15 - 20 nm carbon (C) by thermal evaporation.

19 **S1.2 Sample analysis**

20 X-ray diffraction was performed by placing powders into aluminium (Al) holders, then 21 analysing with a copper (Cu) Ka radiation source operating at 35 kV and 40 mA. Samples were 22 scanned from 5 to 55 °2 θ , at a step size of 0.02 °2 θ , with a counting time of 1 s per step. At 23 Cardiff University, a Philips PW1710 Automate Powder Diffractomer was used, and diffraction patterns were analysed using PW1876 PC-Identify software, version 1.0b, and also 24 25 compared with JCPDS cards of standard materials. At the University of Leeds, a Bruker D8 26 diffractometer was used, and diffraction patterns were analysed using the EVA® software 27 using the ICDD PDF2 database. Semi-quantitative elemental analysis by XRF was carried out 28 on an Olympus X-5000 instrument on pressed, powdered samples in the laboratory at Cardiff 29 University. Accuracy of the XRF data was determined to be on average $\pm 6.7\%$ by the analysis 30 of silicate rich reference materials (Certified Reference Material: Stream Sediment STSD-1 31 and STSD-4 from Canmet Mining and Mineral Sciences, Ottawa, Canada) (Table S2). Total 32 carbon (TC) and sulphur measurements were performed on a Leco SC-144DR S/C (sulphur/carbon) analyser at 1350 °C in a pure (>99.9 %) oxygen (O₂) atmosphere. Total 33 organic carbon (TOC) measurements were performed using a Shimadzu TOC-L total organic 34 35 carbon analyser using phosphoric acid digestion. For both TC and TOC analysis, Leco 36 calibration samples were run prior to analysis to assess instrument accuracy. Twelve samples were digested and analysed by ICP-OES via a 4 acid digest method (EPA, 1996), ⁵² (for sample 37 38 details, see Table S3). Briefly, 0.1 g of sample was placed in a PTFE lined microwave digest 39 cell and 2 mL of analytical grade 45.71 % hydrofluoric acid (HF) was added. This was allowed 40 to stand for 12 hrs, then 6 mL of aqua regia solution (1:1 ratio of analytical grade 32 % 41 hydrochloric acid (HCl) and 70 % nitric acid (HNO₃)) was added, and then microwave digested 42 using an Anton Paar Multiwave 3000, at 200 °C (1400 W) for 30 minutes (after a 10 minute 43 up ramp time period). After a 15 minute cooling period, the resultant solution was neutralised 44 by adding 12 mL of analytical grade 4 % boric acid (H₃BO₃), and then microwave digested at 45 150 °C (900 W) for 20 minutes (after a 5 minute ramp-up time period). After a further 15 46 minute cooling period, the sample was analysed by inductively coupled plasma-optical 47 emission spectroscopy (ICP-OES) using a Perkin Elmer Optima 2100 DV instrument. Calibration standards (20, 40, 60, 80 and 100 mg/L) were prepared in 1% nitric acid from 1000 48 49 mg/L ICP standards. Experimental blanks and elemental standards at 50 mg/L were analysed 50 to check for accuracy and instrumental drift. Accuracy of the ICP data was determined to be 51 on average ± 3.6 % by the analysis of silicate rich reference materials (Certified Reference 52 Material: Stream Sediment STSD-1 from Canmet Mining and Mineral Sciences, Ottawa, 53 Canada) (Table S2). Selected samples from each borehole were chosen for scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDS) analysis (for sample details, 54

55 see Table S3). At Cardiff University, SEM analyses were performed using a FEI-XL30 FEG-ESEM under high vacuum conditions. A beam energy of 15 keV was used with a 50 µm 56 diameter final aperture with a nominal beam current of ~1 nA and a 10 mm working distance. 57 EDS spectra were acquired using a 10 mm² Oxford Instruments X-Act silicon (Si) drift detector 58 59 with Inca software with a 20 s livetime and a deadtime of less than 10 %. All totals were 60 normalised to 100 %. At the University of Leeds, SEM analyses were performed using a Tescan VEGA3 XM equipped with an X-max 150 SDD EDS and Aztec 3.3 software. A beam energy 61 62 of 15 keV was used, with a 15 mm working distance, mapping at a resolution of 2 µm. All 63 totals were normalised to 100 %.

64

Table S2. Comparison of recorded XRF elemental wt% concentrations (mg/kg) against
 known standards (Certified Reference Material: Stream Sediment STSD-1 and STSD-4 from
 Canmet Mining and Mineral Sciences, Ottawa, Canada)

			STSD-1 (mg/kg)		
Element	Si	K	Ca	Mn	Fe
Standard	22.493	0.996	1.905	0.387	4.456
1.	21.140	1.101	2.125	0.419	4.701
2.	20.240	1.077	2.082	0.409	4.715
3.	20.870	1.097	2.039	0.411	4.775
Mean	20.750±0.462	1.092 ± 0.013	2.082 ± 0.043	0.413±0.01	4.730±0.039
% Difference	-7.7	9.6	9.2	6.8	4.1
			STSD-4 (mg/kg)		
Element	Si	K	Ca	Mn	Fe
Standard	27.532	1.328	2.859	0.155	3.987
1.	25.67	1.392	3.078	0.166	4.312
2.	25.60	1.397	3.061	0.168	4.361
3.	25.67	1.385	3.064	0.168	4.382
Mean	25.648±0.0038	1.391±0.006	3.068±0.0094	0.167±0.001	4.352±0.036
% Difference	-3.8	4.8	7.3	8.0	9.1

68

69

Borehole	ICP-OES	SEM-EDX
	8.25	2.25
DII 1	20.25	10.25
ВП І	22.25	16.0
	23.75	
	4.25	12.5
рц 1	10.25	18.5
ЫП 2	14.25	
	16.25	
	2.25	4.25
DЦ 2	6.25	16.25
ВЦ Э	12.25	
	18.25	





Figure S3. Slag diffractogram plots from BH 1, BH 2 and BH 3 at various depths





S11



77

78 **Figure S5.** Total carbon concentrations of slag material (via furnace analysis) plotted against

79

- depth
- Key: Circle BH 1, Square BH 2, Triangle BH

								С	oncentratio	n (mg/kg)						
			Na	Ca	Mg	Si	К	Fe	Al	Mn	S	Ba	Sr	Ti	Zn	Р
BH 1	8.25	Slag	2659	283517	18799	213704	1887	5626	57750	4894	8276	974	1142	1486	65	104
	20.25	Slag	4513	287328	7095	176875	3416	9138	45388	3241	13138	5251	1170	1404	88	35
	22.25	Slag	5692	327550	13177	143574	3565	103980	56112	8990	11641	1646	1568	1670	93	32
	23.75	Clay	11724	18351	1850	339033	9218	25350	45003	501	4461	372	184	3562	170	347
BH 2	4.25	Slag	5652	373198	13952	174423	2792	4801	64068	6122	6598	1537	1489	1797	52	11
	10.25	Slag	3347	283069	14959	168147	2393	5179	48282	4063	9679	1111	1605	1656	43	17
	14.25	Slag	3713	332455	22902	141481	3285	44822	66405	9087	11065	1962	1790	1571	43	45
	16.25	Slag	3255	326317	25890	138728	1799	2667	52216	3890	17548	976	1409	1286	27	43
BH 3	2.25	Fayalite	14115	25997	1797	219154	1755	254148	11061	104433	906	4113	145	5285	51	0
	6.25	Slag	2625	291799	20001	179643	1992	5888	61990	6419	11579	2953	1708	1822	26	53
	12.25	Slag	5394	386777	19170	201957	2978	5890	59392	6727	13756	3230	1808	1808	31	39
	18.25	Slag	3459	350583	18939	159025	2718	14145	67563	5141	18652	942	1344	1542	21	46
	STSD-1	Standard	13353	25729	13267	198672	9962	45464	47632	3872	721	630	170	4600	178	1746
		Measured	13684	25030	13939	208823	9932	44534	46145	3759	691	659	161	4567	188	1758
		Accuracy	2.5	-2.7	5.1	-5.0	-6.4	-2.0	-3.1	-2.9	-4.1	4.6	-5.3	-0.7	5.6	0.7

Table S4: Analysis of borehole samples by HF digestion ICP-OES with known standard material (Certified Reference Material: Stream

Sediment STSD-1 from Canmet Mining and Mineral Sciences, Ottawa, Canada)



Figure S6: BSE images of typical slag material, from BH 1 at 4.5 m

Table S5: Semi-quantitative phase composition in slag sample from BH 1 (4.5 m depth) by

SEM-EDS spot analysis	Numbers refer to areas	mapped in Figure S5	D
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	1.	2.	3.	4.	5.	
Element	'Melilite'	'Fe- metal (oxidised)'	'Melilite'	'Melilite'	'Larnite'	
	Atom %±1σ	Atom %±1σ	Atom %±1σ	Atom %±1σ	Atom %±1σ	
Mg	2.8	n.d.*	2.6	2.6	0.4	
Al	5.3	n.d.*	7.7	7.7	n.d.*	
Si	8.8	n.d.*	9.3	9.0	9.7	
S	0.7	n.d.*	n.d.*	n.d.*	n.d.*	
Ca	25.9	1.6	23.9	27.2	35.1	
Fe	n.d.*	48.4	n.d.*	n.d.*	n.d.*	
0	56.5	50.0	56.6	56.4	54.8	

*not detected n, number of EDS spectra

Note: mineral names are inferred and are not definitive.



Figure S7. SEM-EDS elemental maps for major elements present in the slag sample shown in Figure 3

	1.	2.	3.	4.	5.	6.	7.	8.
Element	'Fe-metal'	'CaS'	'Larnite'	'Melilite'	'Melilite'	'Amphorous' Ca-Si-H'	'Blocky Ettringite'	'Needle Thaumasite'
	n = 1	n = 5	n = 6	n = 6	n = 5	n = 5	n = 4	n = 5
	Atom %±1σ	Atom % ±1σ	Atom % ±1σ	Atom % ±1σ	Atom % ±1σ	Atom % ±1σ	Atom % ±1σ	Atom % ±1σ
0	n.d.*	17 ±3	58 ± 0.3	59 ± 0.2	60 ± 0.4	70 ±2	74 ±1	72 ±2
Na	n.d.*	0.7 ± 0.70	n.d.*	n.d.*	n.d.*	n.d.*	n.d.*	n.d.*
Mg	n.d.*	n.d.*	1.0 ± 0.02	1.4 ± 0.05	3.4 ± 0.5	0.6 ± 0.8	n.d.*	n.d.*
Al	n.d.*	n.d.*	0.2 ± 0.4	14 ± 0.4	8.4 ± 0.5	n.d.*	4.5 ± 0.5	0.6 ± 0.1
Si	n.d.*	n.d.*	14 ± 0.1	9.7 ±0.1	9.7 ±0.1	7.5 ± 1.8	0.3 ± 0.2	4.7 ± 1.2
S	n.d.*	42 ± 1	n.d.*	n.d.*	n.d.*	0.2 ± 0.1	7.4 ± 0.4	8.0 ± 1.7
Ca	1.3	41 ±2	27 ± 0.5	17±0.3	16 ± 0.1	20 ± 1.4	14 ± 1	14 ± 1
Mn	n.d.*	0.4 ± 0.03	n.d.*	n.d.*	0.1 ± 0.1	n.d.*	n.d.*	n.d.*
Fe	98.7	n.d.*	n.d.*	n.d.*	n.d.*	n.d.*	n.d.*	n.d.*
Ca/Si	n.a.	n.a.	1.91 ±0.02	1.73 ±0.02	1.29 ±0.02	2.8 ±0.8		3.2 ±0.6
Ca/Al	n.a.						3.0 ± 0.2	-
Ca/S	n.a.						1.9 ±0.1	1.9 ±0.5

Figure 3C) (1 - 5) and Figure 3D) (6 - 8)

Table S6. Semi-quantitative phase composition in slag sample from BH 1 (16m depth) by SEM-EDS spot analysis. Number refer to spots in

*not detected n, number of EDS spectra

Note: mineral names are inferred and are not definitive.



Figure S8: Averaged particle size distribution curves for Consett slag material

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