Systematic Approach to Determination of Maximum Achievable Capture Capacity via Leaching and Carbonation Processes for Alkaline Steelmaking Wastes in a Rotating Packed Bed

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Figure S1. Schematic diagram of the experimental set-up for carbonation of BOFS slurry in an RPB. (1) BOFS slurry storage tank, (2) pump, (3) slurry inlet, (4) RPB reactor, (5) rotor, (6) gas inlet, (7) gas outlet, (8) gas flow rate controller, (9) rotameter, and (10) CO_2 concentration analyzer.

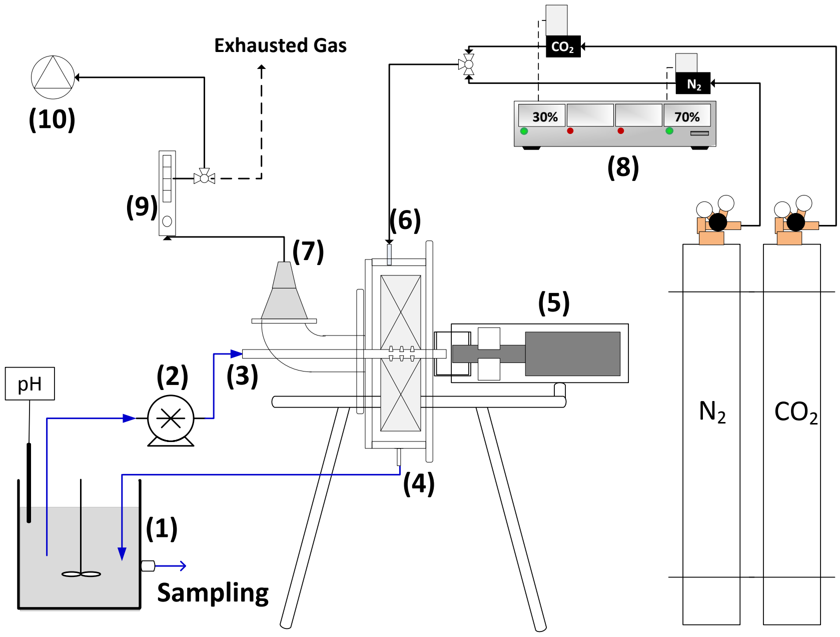


Figure S2. QXRD Procedure using Rietveld method via GSAS program.

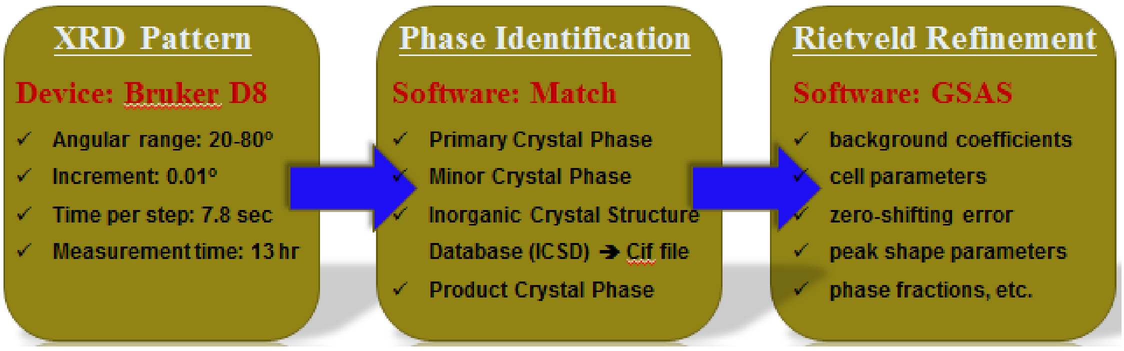
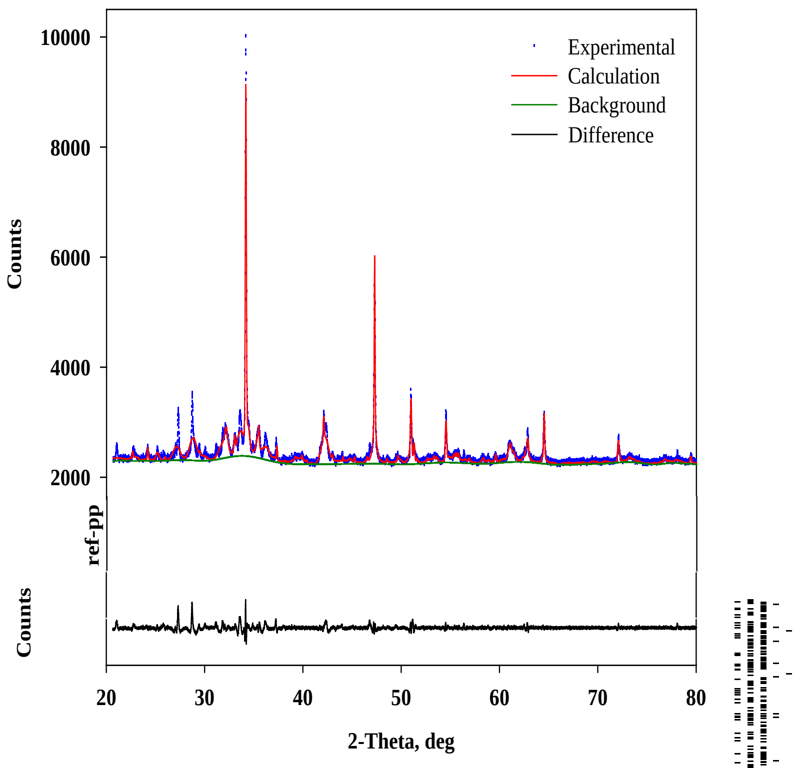


Figure S3. Experimental and calculated XRD diffractogram by Rietveld method in GSAS program for (a) fresh and (b) carbonated BOFS.

(a)



(b)

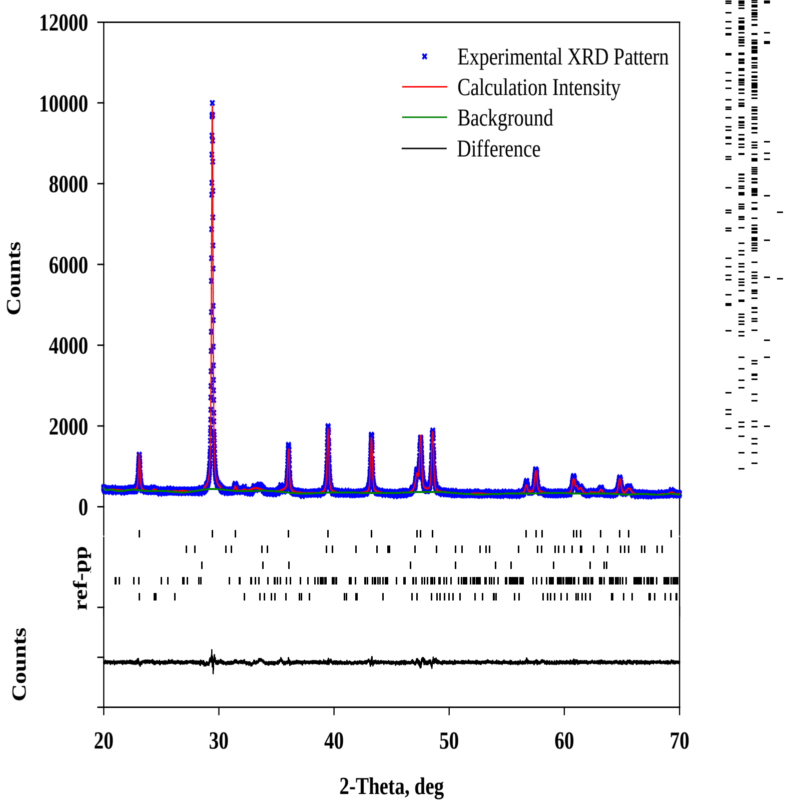


Figure S4. Rietveld refinement results for BOFS before and after carbonation in the case of CRW, with 95% confidence interval. Abbreviation: BRO: brownmillerite ($Ca_2Fe_{1.014}Al_{0.986}O_5$); DSH: α–dicalcium silicate hydrate (C_2-S-H); POR: portlandite ($Ca(OH)_2$); WUS: wustite (FeO); WOL: wollastonite (C_1S); CAL: calcite ($CaCO_3$).

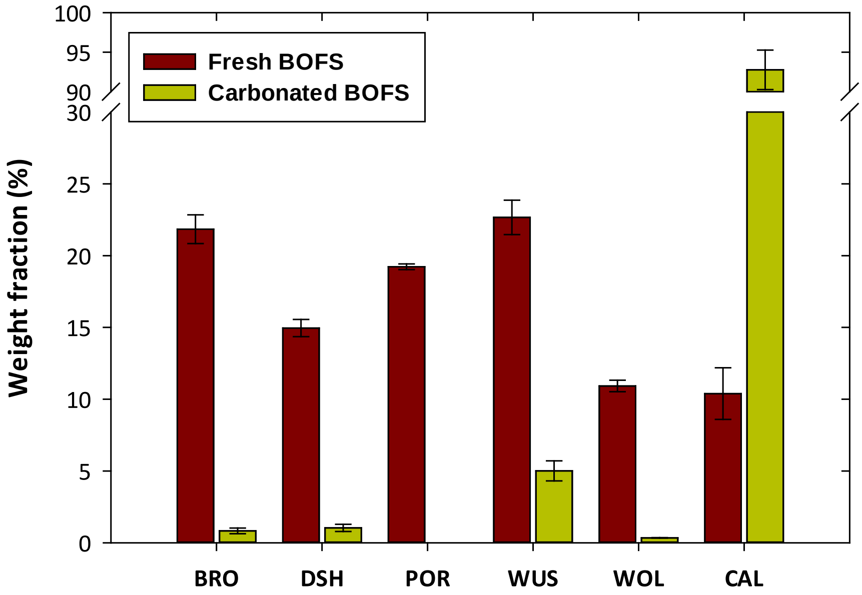


Figure S5. Summary of Mass balance results of CO_2 and calcium ions for carbonation reaction of BOFS via an RPB, as reported in our previous research work.[12,36]

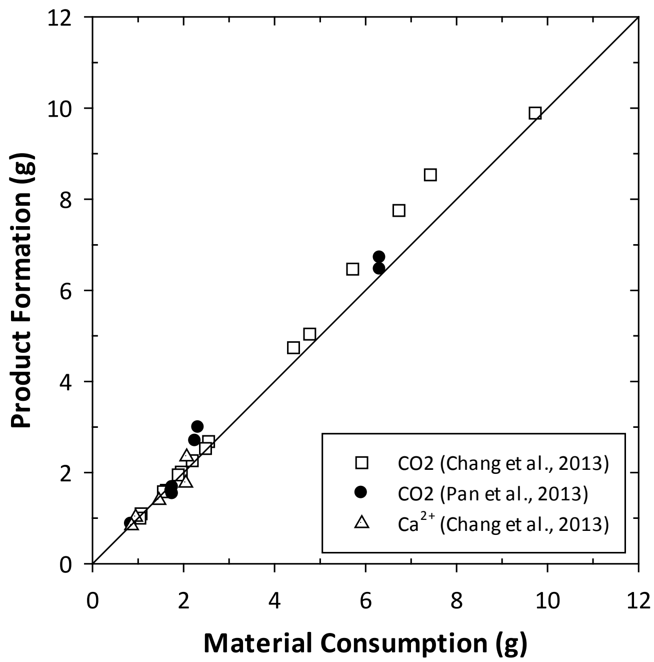
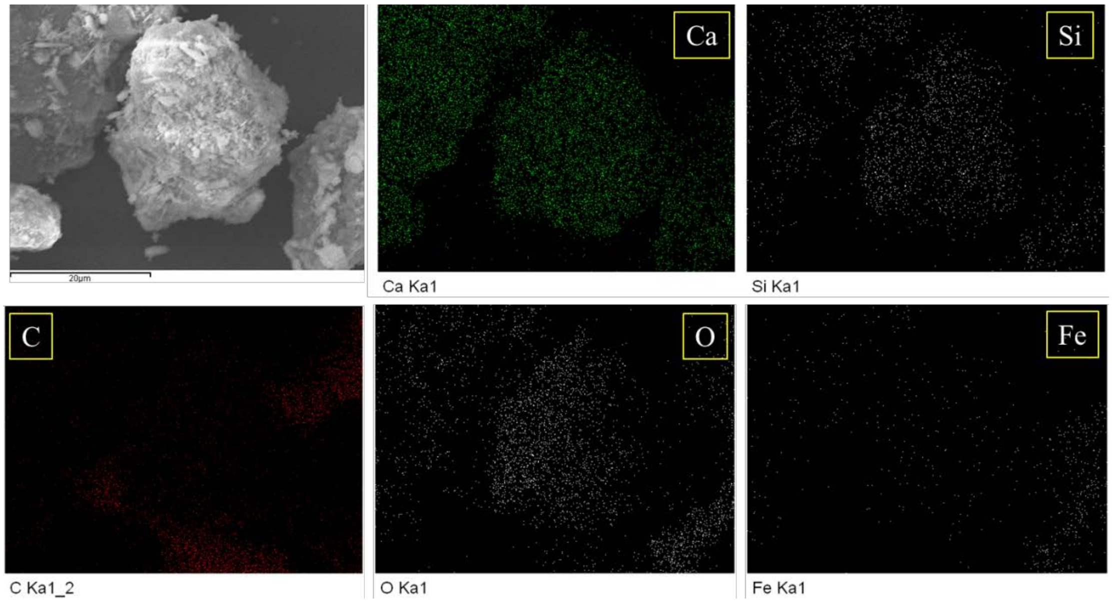


Figure S6. SEM image with elemental mapping of Ca, Fe, Si, C and O for (a) fresh and (b) carbonated BOFS.

(a)



(b)

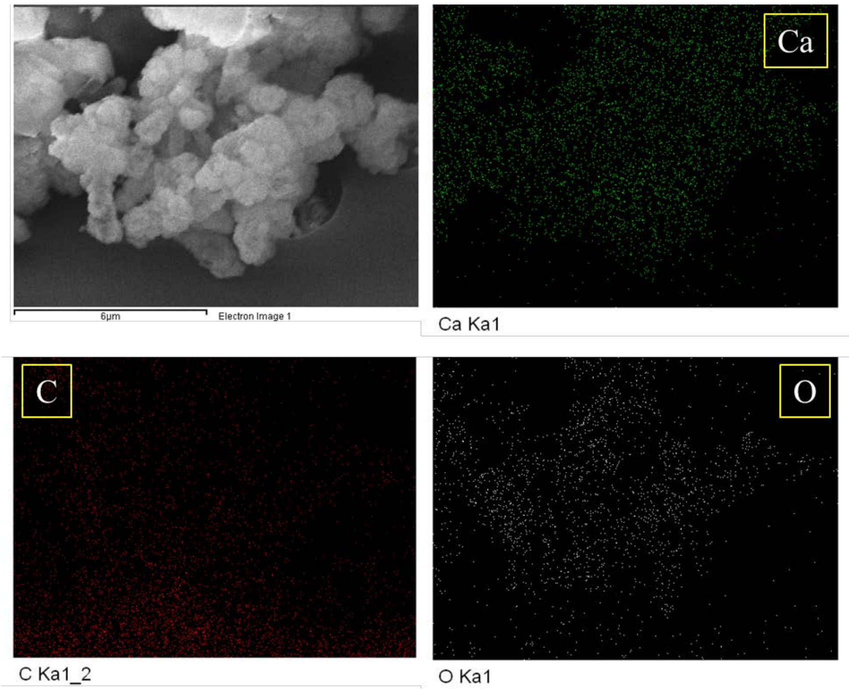


Figure S7. Systematic approach to determination of maximum achievable capture capacity (MACC) via leaching and carbonation processes for BOFS in an RPB.

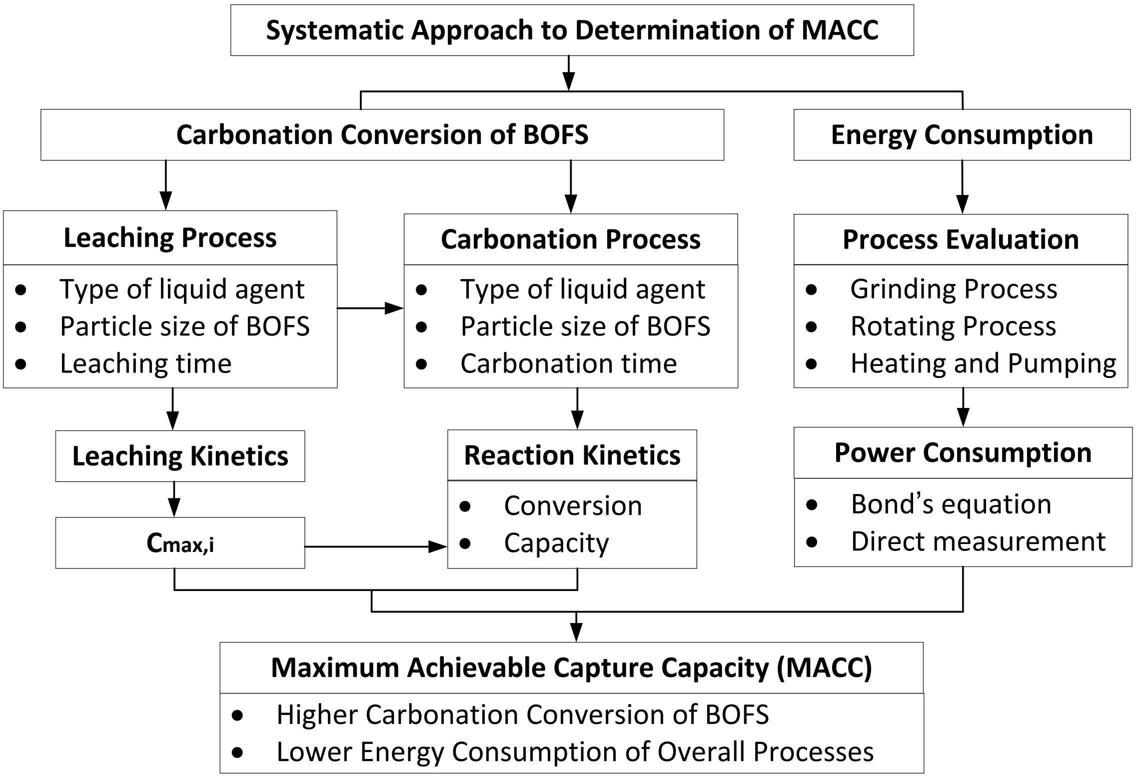


Table S1. Physicochemical properties of basic oxygen furnace slag (BOFS) used in this study.

Item		Unit	Categories				
			< 125 μm	125–350 μm	350–500 μm	500–840 μm	840–1190 μm
Physical Properties	Weight fraction [a]	%	27.9	23.3	6.23	10.0	6.5
	True density [b]	g cm^{-3}	3.04	3.40	3.54	3.63	3.66
	BET surface area [c]	m^2 g^{-1}	5.615 ± 0.015	2.248 ± 0.005	1.975 ± 0.003	1.745 ± 0.006	1.945 ± 0.009
	Langmuir surface area	m^2 g^{-1}	7.342 ± 0.205	2.960 ± 0.081	2.599 ± 0.678	2.301 ± 0.062	2.544 ± 0.071
Chemical properties [d]	Loss on ignition (LOI)	%	8.9	4.3	2.9	1.7	1.3
	SiO_2	%	8.6	9.1	9.2	9.3	9.6
	Al_2O_3	%	0.8	1.1	1.1	1.2	1.3
	Fe_2O_3	%	26.0	29.4	31.0	31.9	32.9
	CaO	%	48.2	47.2	46.5	45.3	43.6
	MgO	%	3.5	4.9	5.2	5.4	5.5
	SO_3	%	0.2	0.2	0.2	0.2	0.2
	TiO_2	%	0.4	0.5	0.5	0.5	0.6
	MnO	%	2.8	2.8	2.9	2.9	2.9

[a]: The rest 26.1% in weight fraction is for those particle size greater than 1190 μm; [b]: Density was analyzed with Micromeritrics Accupyc 1340 by Particulate Technology Laboratory (NTU); [c]: BET Surface Area was analyzed with Micromeritrics ASAP2010 by Particulate Technology Laboratory (NTU); [d]: Analyzed with XRF by CHC Resources Corporation;

Table S2. Physicochemical properties of cold-rolling wastewater (CRW) used in this study.

Item		Unit	Value
Water quality	pH	-	11.20~11.87
	Conductivity	μmho cm^{-1}	4860
	TDS	mg L^{-1}	2680
Cation concentration [a]	Na^{+}	mg L^{-1}	837
	K^{+}	mg L^{-1}	268
	Ca^{2+}	mg L^{-1}	147
	Mg^{2+}	mg L^{-1}	0.28
	Fe^{3+}	mg L^{-1}	1.36
	Cd^{2+}	mg L^{-1}	0.58
	Zn^{2+}	mg L^{-1}	75
	Al^{3+}	mg L^{-1}	2.1
	Cr^{3+}	mg L^{-1}	0.31
	Ni^{2+}	mg L^{-1}	1.53
Anion concentration	Cl^{-} [b]	mg L^{-1}	1400
	SO_4^{2-} [c]	mg L^{-1}	232
	NO_3^{-} [d]	mg L^{-1}	8.74

[a]: Analyzed with ICP-AES (JOBIN YVON, JY24); [b]: Measured by precipitation titrimetry (i.e., Mohr method); [c]: Measured by Nephelometer method; [d]: Analyzed with IC (DIONEX DX-100).

Table S3. Specification of XRD operation for Rietveld refinement in this investigation.

Diffractometer	Bruker D8 Advance
Goniometer	θ-2θ, radius 217.5 mm
Source	Cu Kα, (λ = 1.54 Å), line focus
Generator	40 kV, 40 mA
Sample	
Sample Surface diameter	20 mm
Preparation	Drying at 105 ^{o}C for 3 hr
Incident optics	
Monochromator	Vario1 Johansson focusing
Programmable divergence slit	0.5^{o} (fixed)
Incident anti-scatter slit	0.5^{o}
Receiving optics	
Programmable anti-scatter slit	1.5^{o} (fixed)
Soller slit	0.02×0.02 radians
Detector	Vantec
Scan information	
Angular range (2θ)	20-80^{o}
Increment (2θ/step)	0.01
Time per step (s)	7.8
Measurement time (min)	780

Table S4. Variations of $C_{max,i}$, k_i and n_i values for various metal ions leaching into CRW for 90 min.

Dp of BOFS	Model Parameter	Unit	Metal ions: Ca	Na	K	Zn	Pb	Al	Ni	Fe	Cr	Mg
< 125 µm	C_{max}	ppm	2690.2	1177.4	232.4	79.0	43.0	22.6	10.37	9.89	3.25	2.96
	k	ppm/s	0.0188	0.0158	0.0618	0.1181	0.0685	0.0921	0.1469	0.1231	0.1995	0.0417
	n	-	1.14	1.42	1.12	1.0	1.0	1.0	1.0	1.24	1.0	1.0
	r^2	-	0.999	0.997	0.996	0.999	0.984	0.990	0.999	0.999	0.999	0.977
125–350 µm	C_{max}	ppm	2115.6	1071.7	229.6	77.1	37.9	20.8	8.34	9.82	3.11	2.85
	k	ppm/s	0.0146	0.0194	0.0429	0.1282	0.1044	0.0880	1.0121	0.1322	0.1127	0.0502
	n	-	1.26	1.53	1.12	1.0	1.0	1.0	1.0	1.18	1.0	1.0
	r^2	-	0.996	0.986	0.997	0.999	0.998	0.992	0.997	0.999	0.998	0.999
350–500 µm	C_{max}	ppm	1310.4	1045.2	218.0	70.5	25.7	19.1	7.51	9.89	2.56	2.61
	k	ppm/s	0.0114	0.0192	0.0411	0.0947	0.0612	0.1149	0.2091	0.1240	0.0559	0.0600
	n	-	1.26	1.44	1.21	1.0	1.0	1.0	1.0	1.26	1.0	1.0
	r^2	-	0.973	0.955	0.995	0.998	0.981	0.996	0.999	0.998	0.997	0.999
500–840 µm	C_{max}	ppm	1256.3	867.1	208.0	67.0	21.7	16.6	7.31	9.76	2.14	1.99
	k	ppm/s	0.0116	0.0188	0.0379	0.1246	0.0835	0.0848	0.1419	0.1162	0.0680	0.0899
	n	-	1.28	1.48	1.18	1.0	1.0	1.0	1.0	1.24	1.0	1.0
	r^2	-	0.973	0.962	0.990	0.999	0.993	0.996	0.999	0.998	0.976	0.982
840–1190 µm	C_{max}	ppm	876.0	826.9	201.4	63.2	19.0	14.1	7.05	9.25	2.02	1.67
	k	ppm/s	0.0118	0.0184	0.0341	0.1420	0.0805	0.0833	0.1107	0.1091	0.0744	0.0826
	n	-	1.39	1.50	1.19	1.0	1.0	1.0	1.0	1.32	1.0	1.0
	r^2	-	0.983	0.950	0.998	0.999	0.987	0.990	0.998	0.999	0.986	0.986

Table S5. Comparisons of CO_2 capture capacity estimated by Rietveld refinement results (in this study) with those calculated by TG technique in the literature.[36] (operating condition: direct carbonation using BOFS/CRW system in RPB, 20 min).

Mineral phases	Abbre.	Molecular weight (g/mol)	Rietveld refinement		Degree of carbonation		Capture capacity (t CO_2/t BOFS)[b]	
			Weight fraction in fresh BOFS (%)	Weight fraction in Carbonated BOFS	In terms of Ca content (%)	Contribution fraction (%)[a]	Estimation in this study	TG technique (Chang et al., 2013)[36]
$Ca_2Fe_{1.04}Al_{0.986}O_5$	-	244.7	21.85 ± 1.24	0.83 ± 0.21	6.47	25.86	0.071	-
$Ca_2(HSiO_4)(OH)$	C2SH	190.1	14.95 ± 0.69	1.04 ± 0.25	5.71	22.82	0.063	-
$Ca(OH)_2$	-	74.0	19.23 ± 0.26	0.02 ± 0.00	9.43	37.69	0.104	-
FeO	-	71.9	22.67 ± 1.22	5.01 ± 0.73	0.0	0.0	0.0	-
$CaSiO_3$	C1S	116.1	10.92 ± 0.41	0.34 ± 0.02	3.41	13.63	0.037	-
$CaCO_3$	-	100.0	10.39 ± 1.81	92.78 ± 2.59	-	-	-	-
Total			100.00	100.00	25.02	100.00	0.275	0.277

[a]: Contribution fraction of each mineral phase for achieving the estimation value of CO_2 capture capacity (i.e., 0.275 t CO_2/ t BOFS) in this study.

[b]: Relative percent difference (RPD) between the two studies was calculated to be 0.63%.