Looping-accelerated CO_2 Mineralization for Cost-competitive Cementitious Materials and Hydrogen

Supplementary Information

Kyle Shank[1], Hefei Xu[1], Yunming Xu[2], Amirmohammad Arjomand Kermani[1], Jiangzhou Qin[1], Shang Zhai[1,3]

1 – Department of Mechanical and Aerospace Engineering, The Ohio State University, 201 West 19th Ave, Columbus, OH 43210, USA

2 – Department of Chemical and Biomolecular Engineering, The Ohio State University, 151 W. Woodruff Avenue, Columbus, OH 43210, USA

3 – School of Earth Sciences, The Ohio State University, 125 South Oval Mall, Columbus, OH 43210, USA

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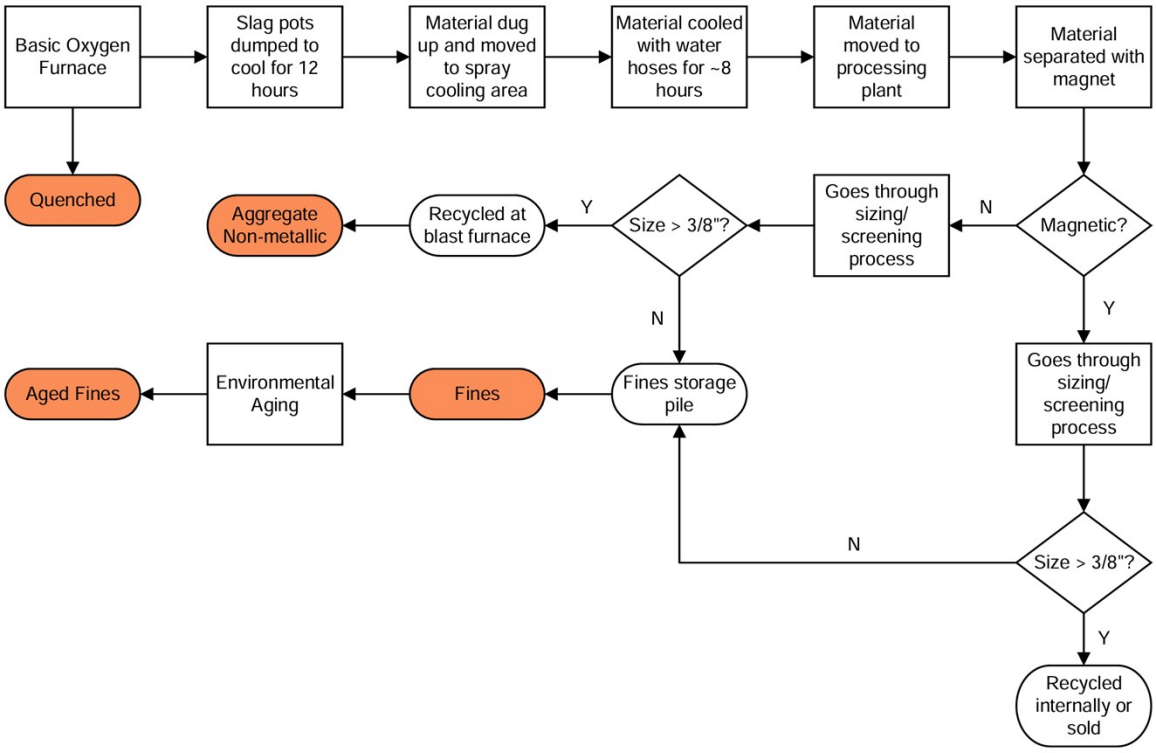

Fig. S1 *Cleveland-Cliffs – Cleveland Works BOF slag processing flow chart.*

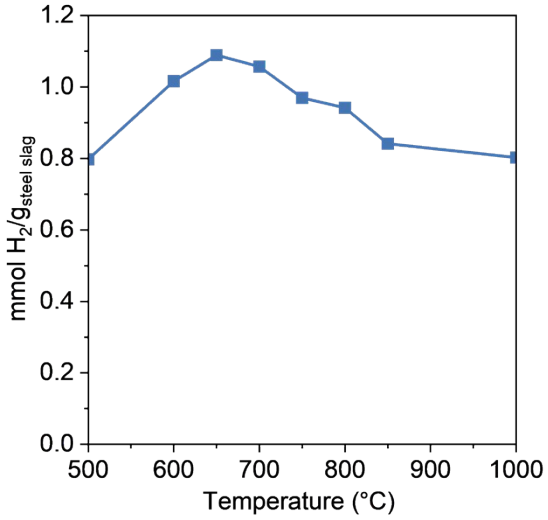

Fig. S2 *Dependence of hydrogen production on temperature for aggregate non-metallic slag.*

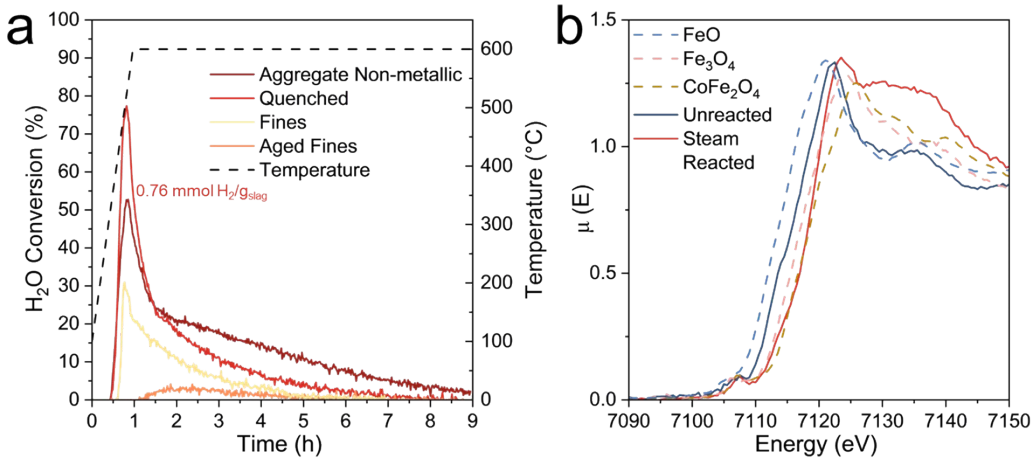

Fig. S3 *(a) H_2 production from water (H_2O) with BOF steel slags. Experiments conducted with 6 g steel slag at 600°C and 2 sccm H_2O mixed with 98 sccm Argon (Ar). Aggregate non-metallic slag is typically recycled for reuse in the blast furnace. Quenched slag was scooped directly out of the BOF. Slag fines are unrecyclable waste from the BOF. Aged fines are also the unrecyclable waste but aged outside for an unknown period. (f) X-ray absorption near edge structure (XANES) for Iron (Fe) K-edge showing the Fe oxidation in quenched slag after reacting with H_2O to produce H_2.*

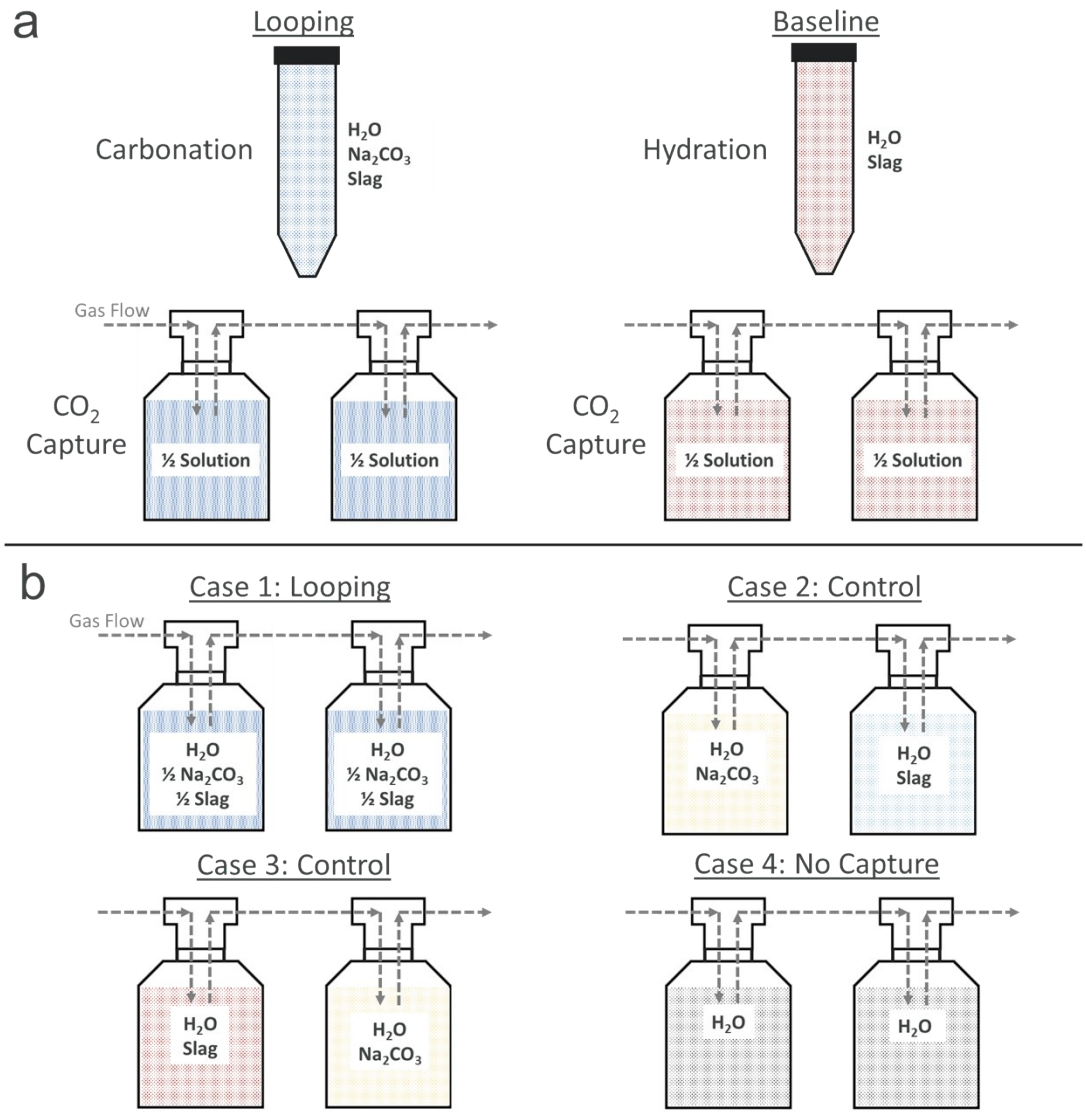

Fig. S4 *(a) Na_2CO_3 Looping Performance "looping" and "baseline" experimental scenarios. (b) Four experimental scenarios for mechanism testing: looping, controls, and blank run.*

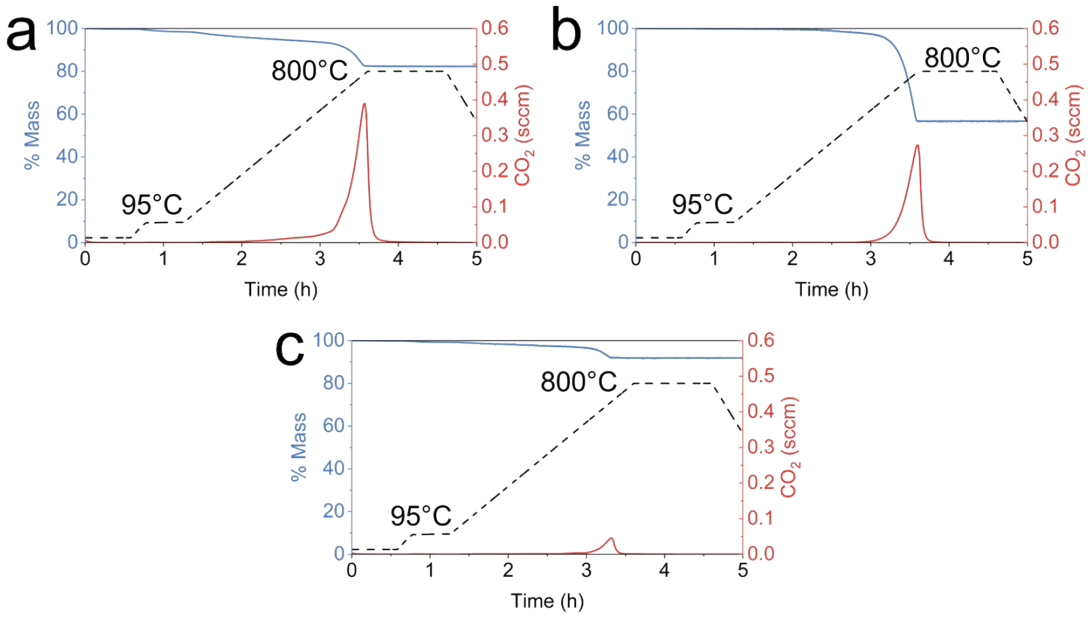

Fig. S5 *(a) Thermogravimetric analysis (TGA) and CO_2 evolution of carbonate-rich product from the 4th cycle of sodium carbonate looping capturing 4% CO_2 (Fig. 2a). (b) TGA and CO_2 evolution of precipitated $CaCO_3$ from the 4th cycle of the baseline experiment capturing 4% CO_2 (Fig. 2a). (c) TGA and CO_2 evolution of carbonate-rich product from the 4th cycle of air CO_2 sodium carbonate looping (Fig. 2e).*

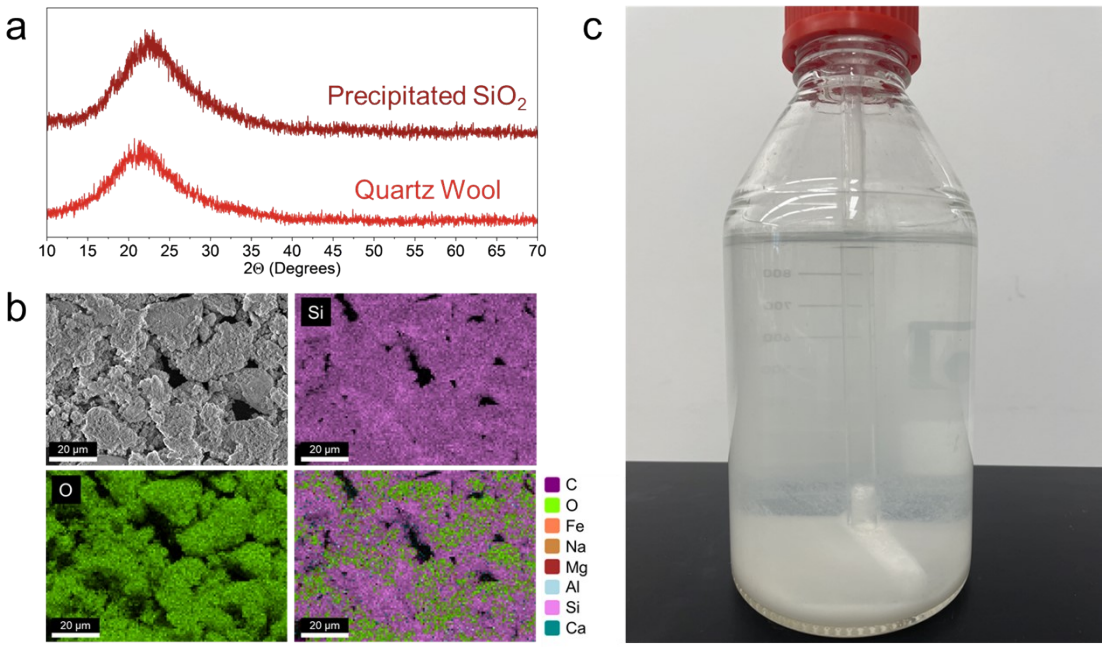

Fig. S6 *(a) XRD of amorphous precipitated SiO_2 compared to quartz wool as a reference. (b) SEM and EDS of precipitated SiO_2 showing main composition is silicon and oxygen. Sample preparation was done with 19 g of quenched BOF slag carbonated in an aqueous solution of 47 g of Na_2CO_3 in 900 mL DI H_2O for 12 hours at 60°C and 80 rpm. The mixture was then filtered, and the solution was subject to 48 hours of 90 sccm pure CO_2 to precipitate silicic acid which was washed, filtered and dried to produce SiO_2. (c) White silicic acid gel precipitating out of solution after 48 hours of pure CO_2 flow.*

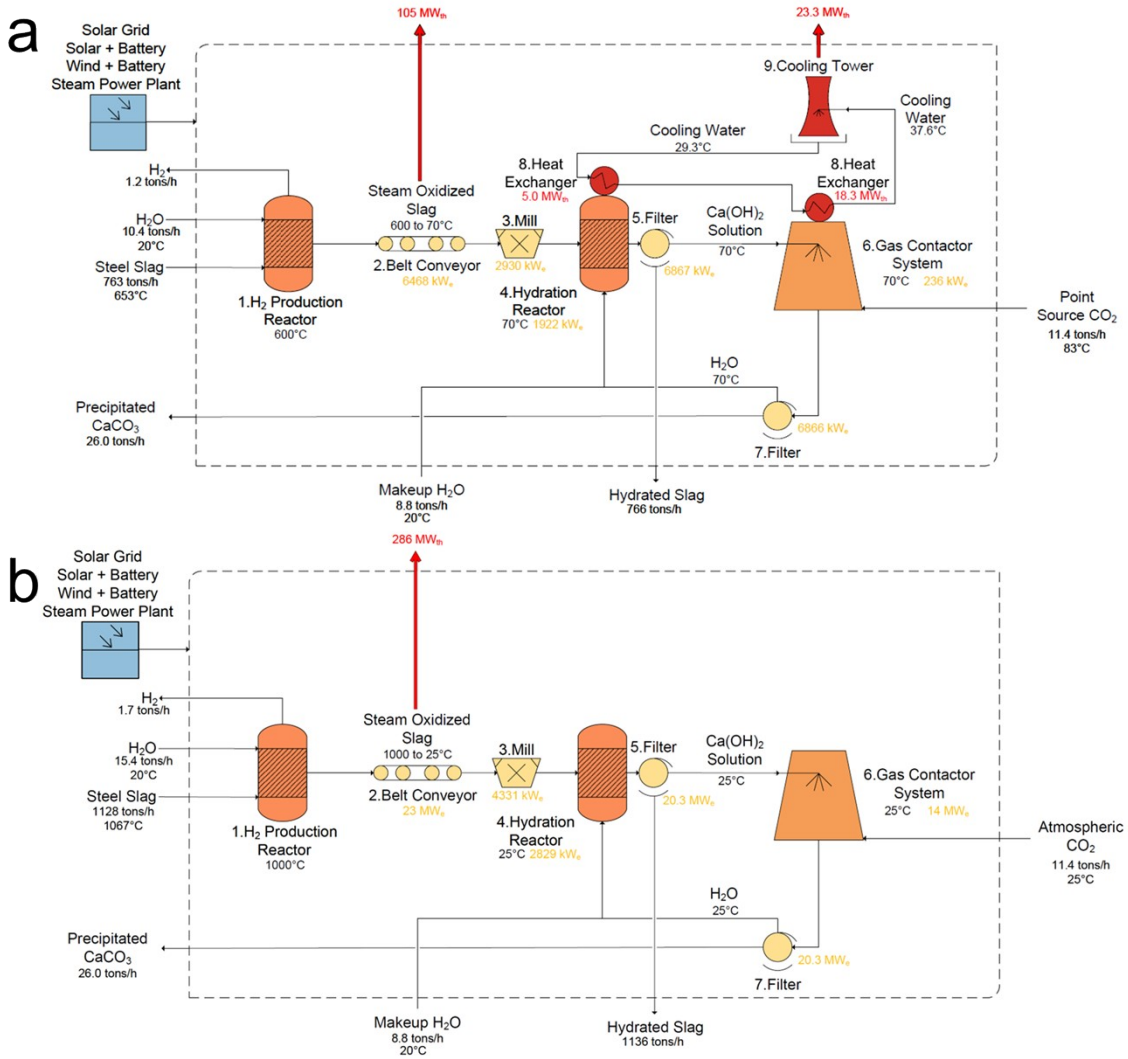

Fig. S7 *Proposed plant design for hydrogen production and CO_2 capture and mineralization with BOF steel slag through baseline process with a) Point-source CO_2 and b) air CO_2.*

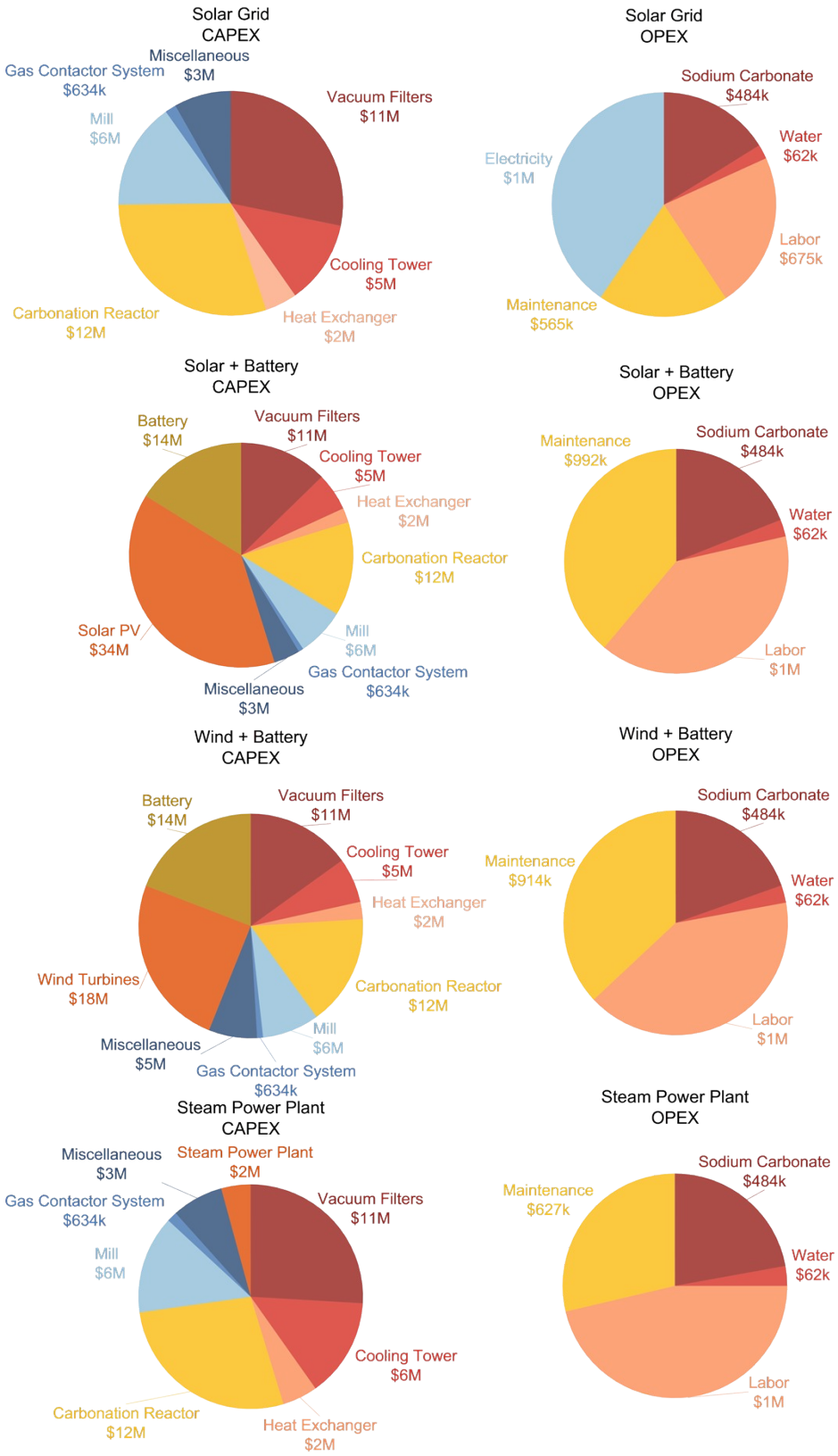

Fig. S8 *Point-source Na_2CO_3 looping CAPEX and OPEX for different electricity sources.*

Fig. S9 *Air CO_2 Na_2CO_3 looping CAPEX and OPEX for different electricity sources.*

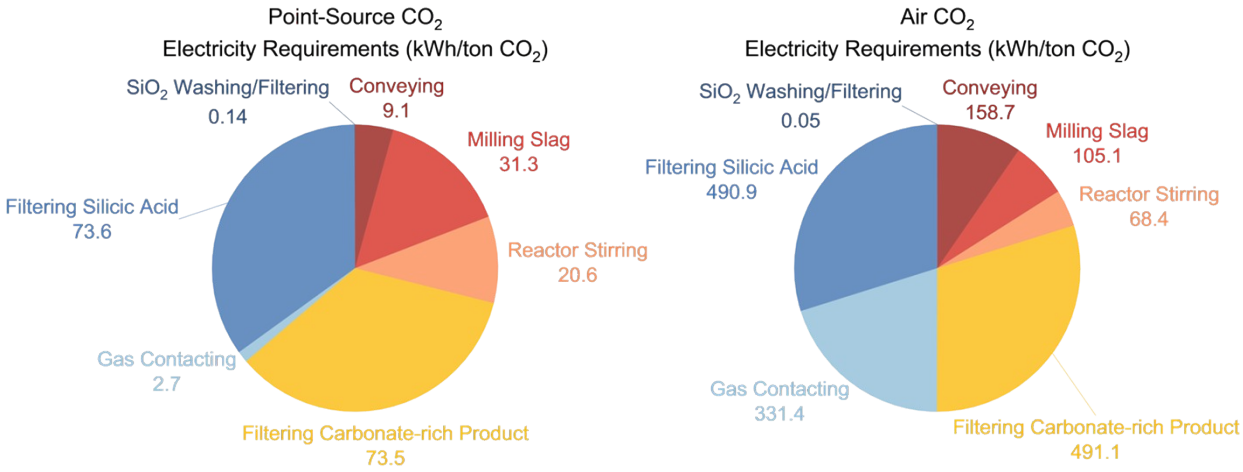

Fig. S10 *Point-source and air CO_2 Na_2CO_3 looping electrical energy requirements.*

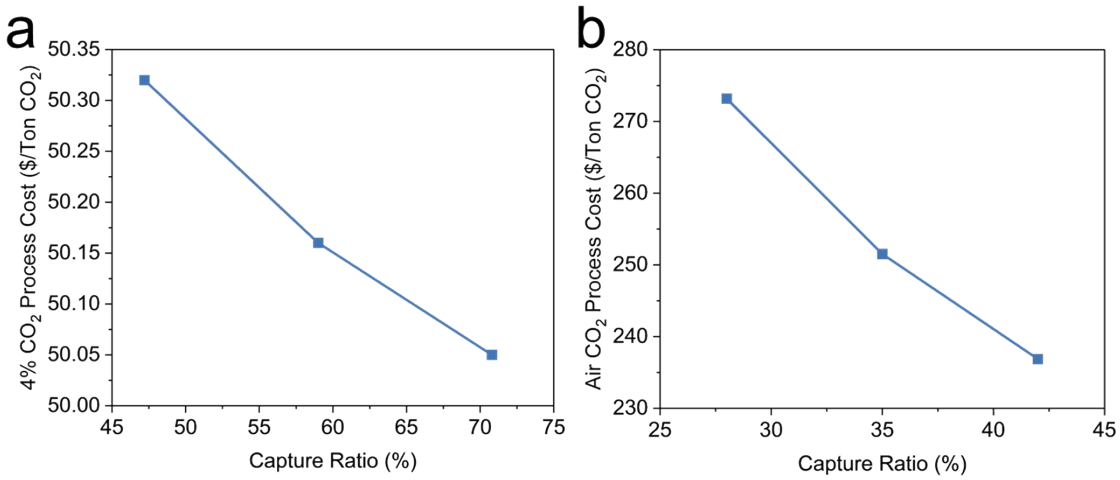

Fig. S11 *Sensitivity of process cost to the CO_2 capture ratio for (a) point-source and (b) air looping. Costs were evaluated by varying the CO_2 capture ratio by ±20% relative to our experimentally observed values for each case, assuming a solar-grid electricity scenario in the scale-up analysis.*

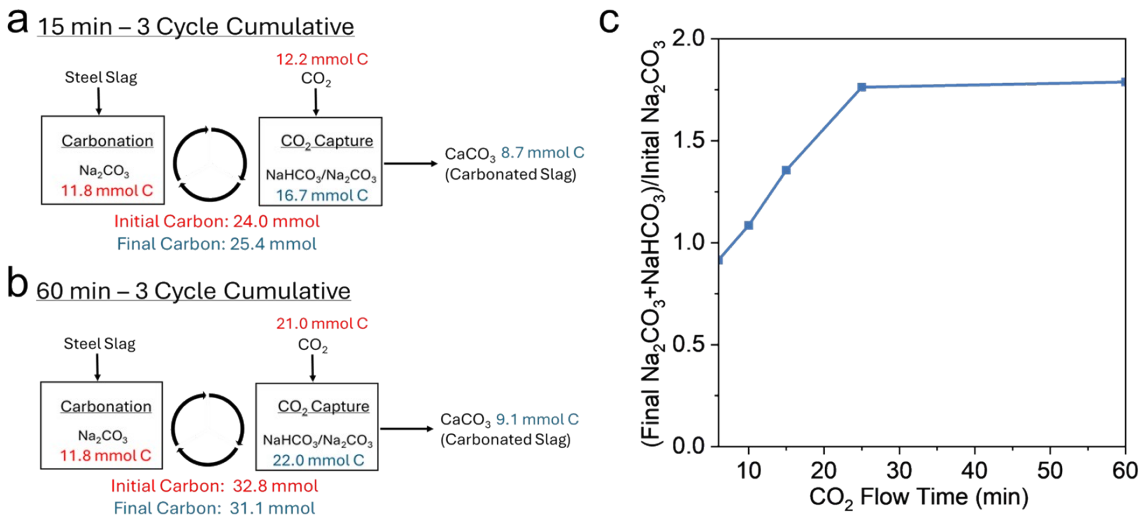

Fig. S12 *Carbon balance over 3 sodium carbonate looping cycles of (a) 15-minute and (b) 60-minute point-source CO_2 capture with quenched BOF slag at 45°C. 0.5 g of steel slag per cycle with 90 sccm 20% CO_2/Ar. (c) Ratio of final Na_2CO_3 and $NaHCO_3$ in solution to initial Na_2CO_3 when CO_2 capture step duration is varied. To quantify the Na_2CO_3 and $NaHCO_3$ in the solution, the pH was raised above 12.5 using sodium hydroxide (NaOH) to convert any bicarbonate ($NaHCO_3$) into carbonate (Na_2CO_3). Calcium chloride ($CaCl_2$) was then added to the solution, causing calcium carbonate ($CaCO_3$) to precipitate. The solution was filtered, and the precipitate was dried at 100°C. The amount of $CaCO_3$ was quantified using Thermogravimetric Analysis (TGA) and Gas Chromatography (GC) by heating to 800°C to decompose $CaCO_3$ into CaO and CO_2, which enabled the determination of the bicarbonate/carbonate in the solution.*

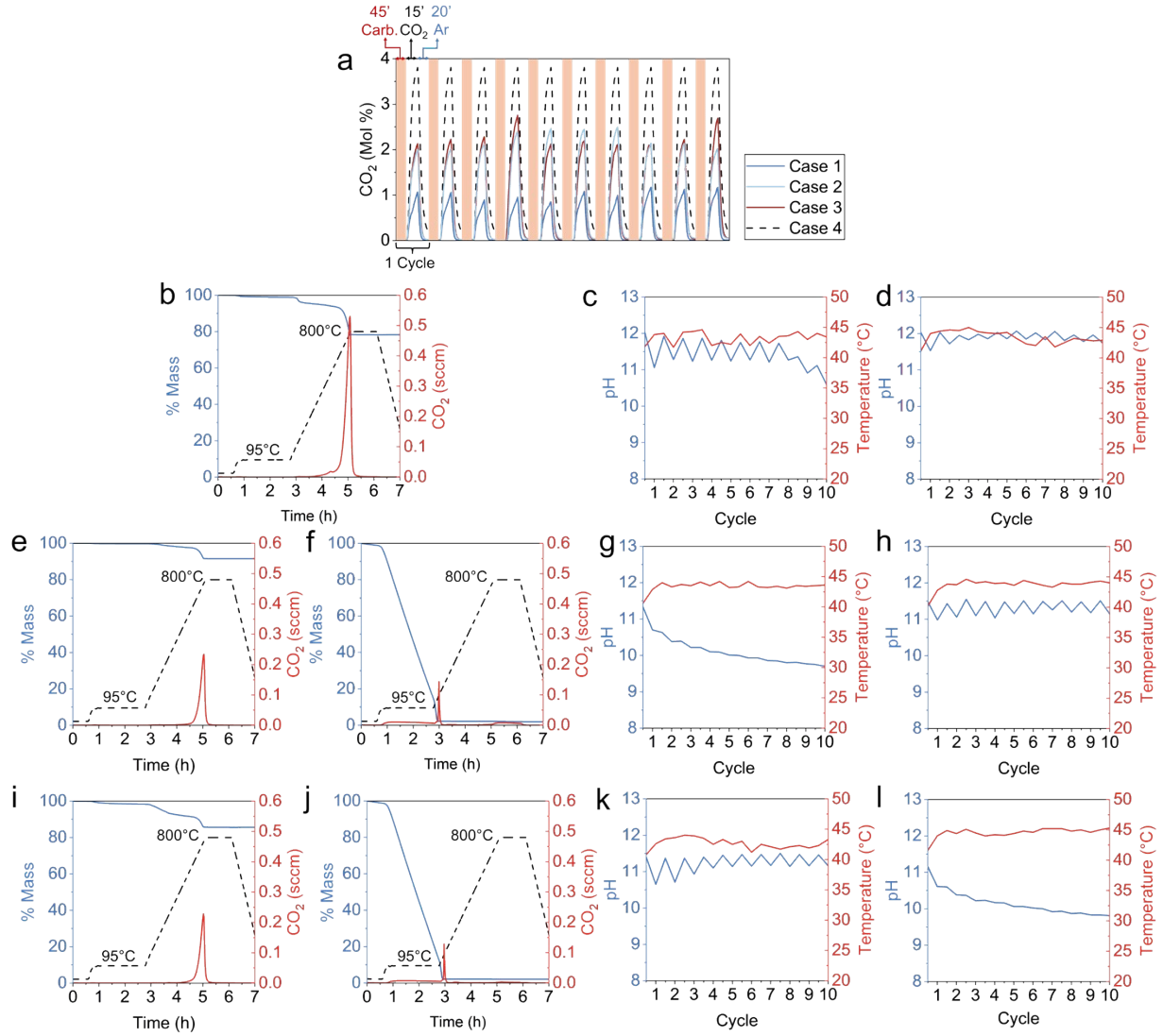

Fig. S13 *(a) 10 cycles of Case 1-4 point-source CO_2 capture with quenched BOF slag at 45°C. 0.5 g of steel slag per cycle with 90 sccm 4% CO_2/Ar. Cases are illustrated by Fig. S4b. (b) Thermogravimetric analysis (TGA) and CO_2 evolution of carbonate-rich product from Case 1 sodium carbonate looping capturing 4% CO_2. (c) pH and temperature in the first bottle of Case 1. Each half cycle is after carbonation, and each full cycle is after CO_2 capture. (d) pH and temperature in the second bottle of Case 1. (e) TGA and CO_2 evolution of carbonate-rich product from Case 2 control. (f) TGA and CO_2 evolution of Na_2CO_3/$NaHCO_3$ solution from first bottle in Case 2. pH and temperature in the first (g) and second (h) bottle of Case 2. (i) TGA and CO_2 evolution of carbonate-rich product from Case 3 control. (j) TGA and CO_2 evolution of Na_2CO_3/$NaHCO_3$ solution from second bottle in Case 3. pH and temperature in the first (k) and second (l) bottle of Case 3.*

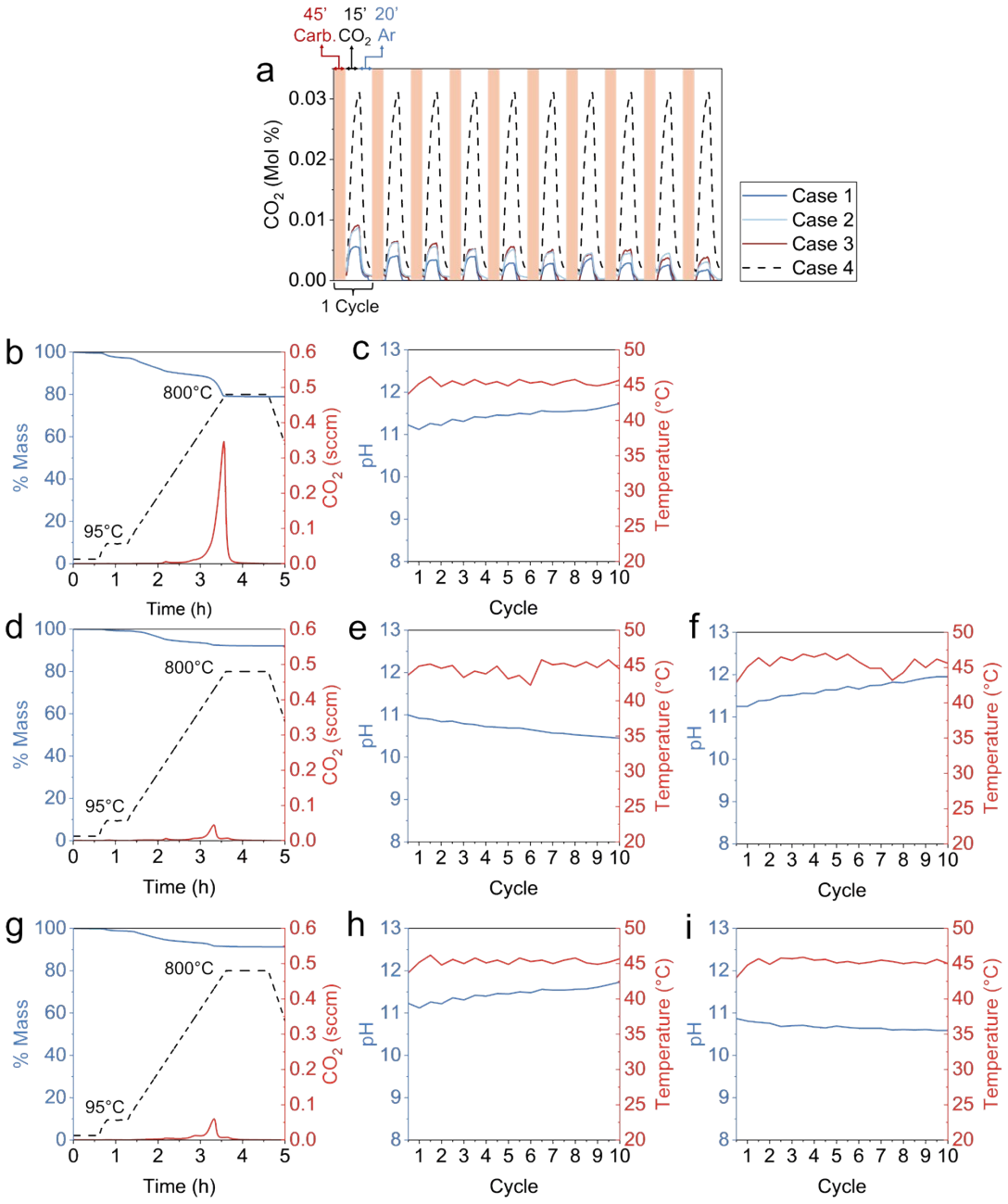

Fig. S14 *(a) 10 cycles of Case 1-4 using air with quenched BOF slag at 45°C. 0.05 g of steel slag per cycle with 90 sccm of 343 ppm CO_2/Air. (b) Thermogravimetric analysis (TGA) and CO_2 evolution of carbonate-rich product from Case 1 sodium carbonate looping capturing 343 ppm CO_2. (c) pH and temperature in the first bottle of Case 1. Each half cycle is after carbonation, and each full cycle is after CO_2 capture. (d) TGA and CO_2 evolution of carbonate-rich product from Case 2 control. pH and temperature in the first (e) and second (f) bottle of Case 2. (g) TGA and CO_2 evolution of carbonate-rich product from Case 3 control. pH and temperature in the first (h) and second (i) bottle of Case 3.*

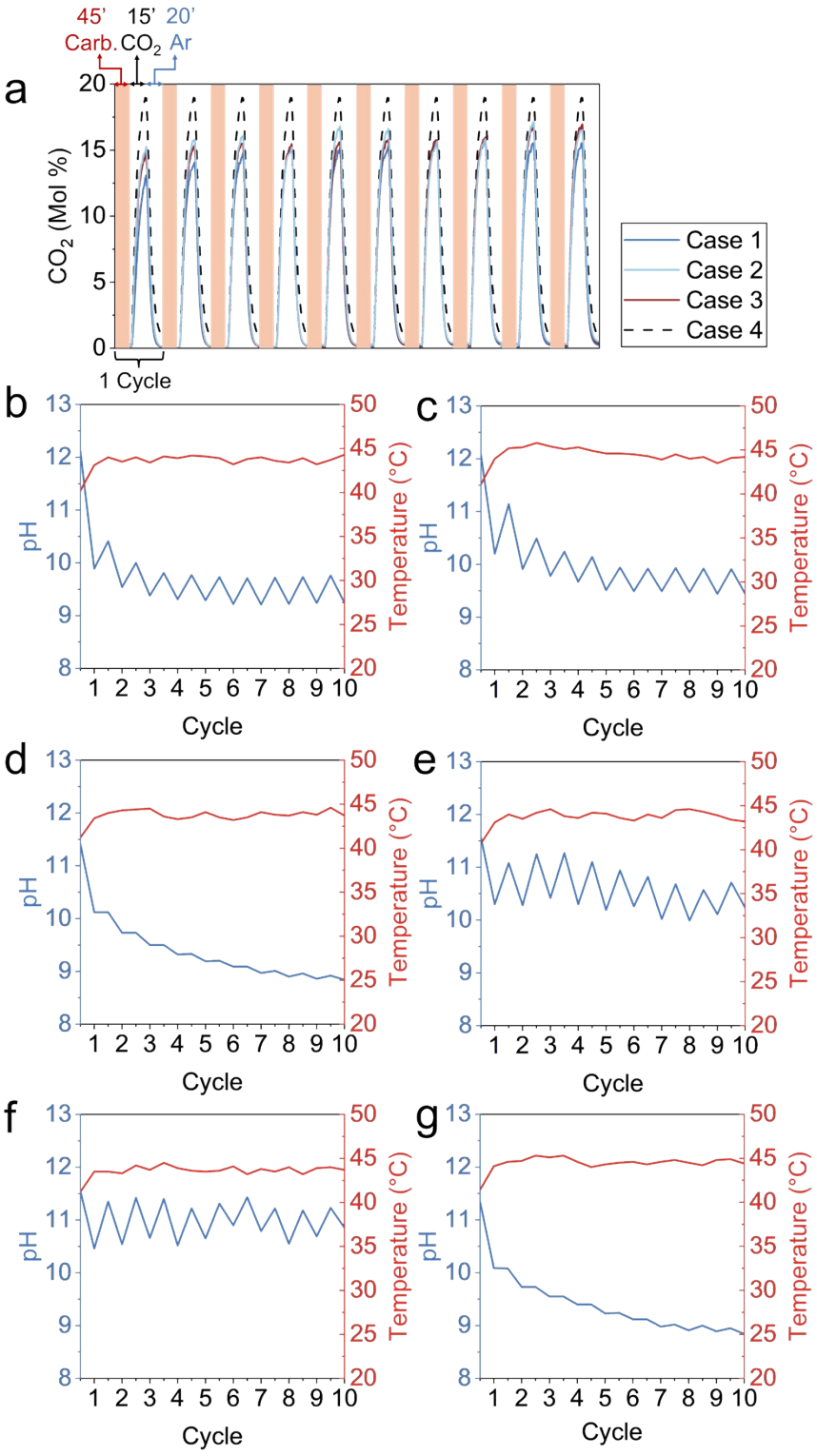

Fig. S15 *(a) 10 cycles of Case 1-4 point-source CO_2 capture with quenched BOF slag at 45°C. 0.5 g of steel slag per cycle with 90 sccm 20% CO_2/Ar. pH and temperature in the first (b) and second (c) bottle of Case 1. pH and temperature in the first (d) and second (e) bottle of Case 2. pH and temperature in the first (f) and second (g) bottle of Case 3.*

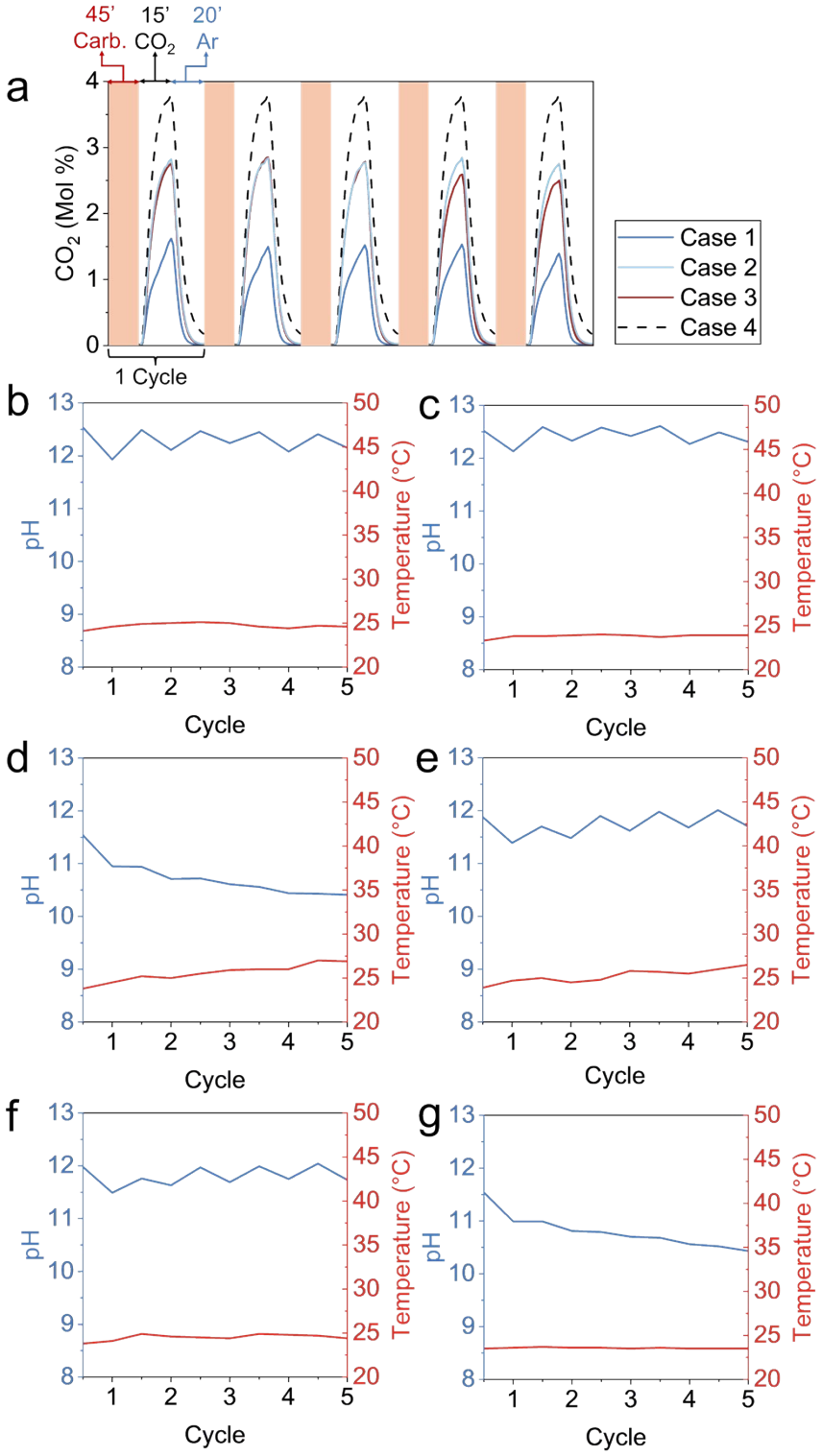

Fig. S16 *(a) 5 cycles of Case 1-4 point-source CO_2 capture with quenched BOF slag at 24°C. 0.5 g of steel slag per cycle with 90 sccm 4% CO_2/Ar. pH and temperature in the first (b) and second (c) bottle of Case 1. pH and temperature in the first (d) and second (e) bottle of Case 2. pH and temperature in the first (f) and second (g) bottle of Case 3.*

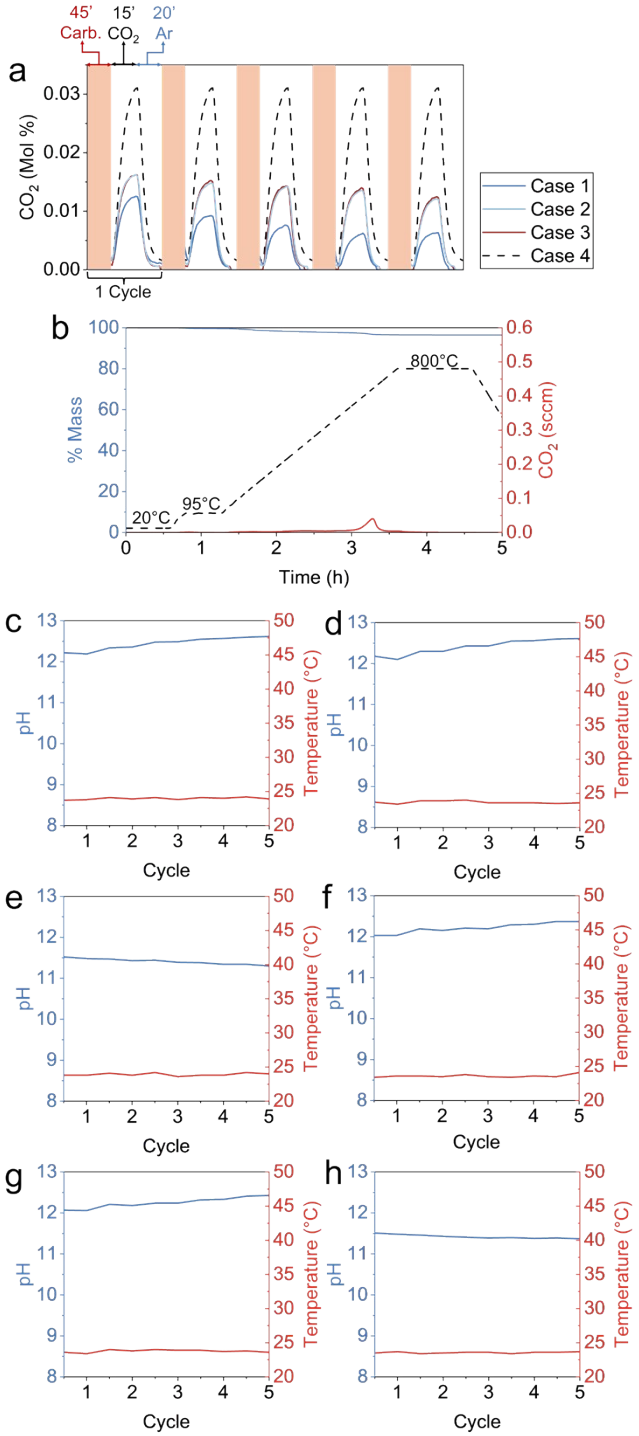

Fig. S17 *(a) 5 cycles of Case 1-4 using air with quenched BOF slag at 24°C. 0.05 g of steel slag per cycle with 90 sccm of 343 ppm CO_2/Air. (b) Thermogravimetric analysis (TGA) and CO_2 evolution of carbonate-rich product from Case 3. pH and temperature in the first (c) and second (d) bottle of Case 1. pH and temperature in the first (e) and second (f) bottle of Case 2. pH and temperature in the first (g) and second (h) bottle of Case 3.*

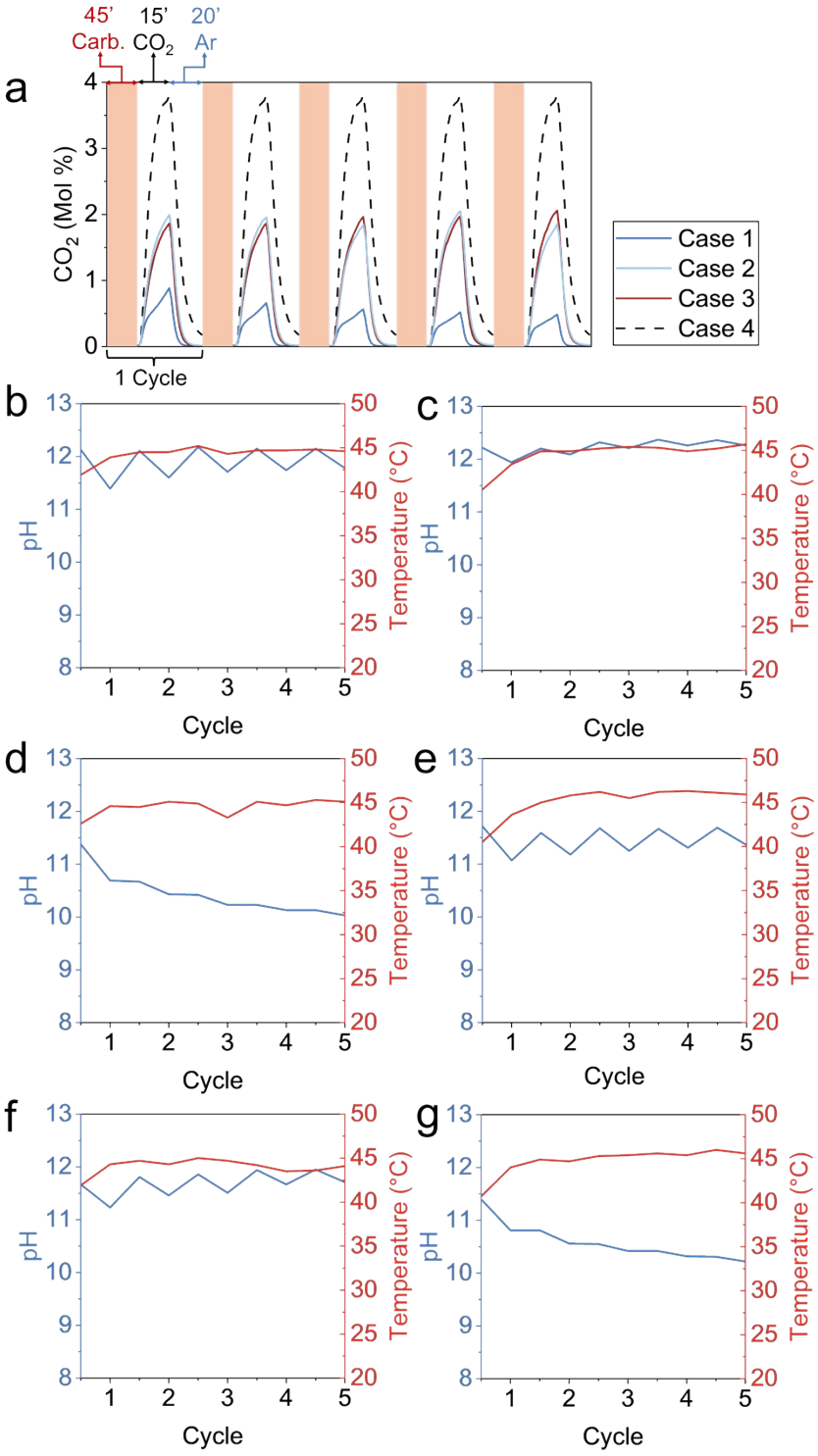

Fig. S18 *(a) 5 cycles of Case 1-4 point-source CO_2 capture with quenched BOF slag at 45°C with no H_2 production step beforehand. 0.5 g of steel slag per cycle with 90 sccm 4% CO_2/Ar. pH and temperature in the first (b) and second (c) bottle of Case 1. pH and temperature in the first (d) and second (e) bottle of Case 2. pH and temperature in the first (f) and second (g) bottle of Case 3.*

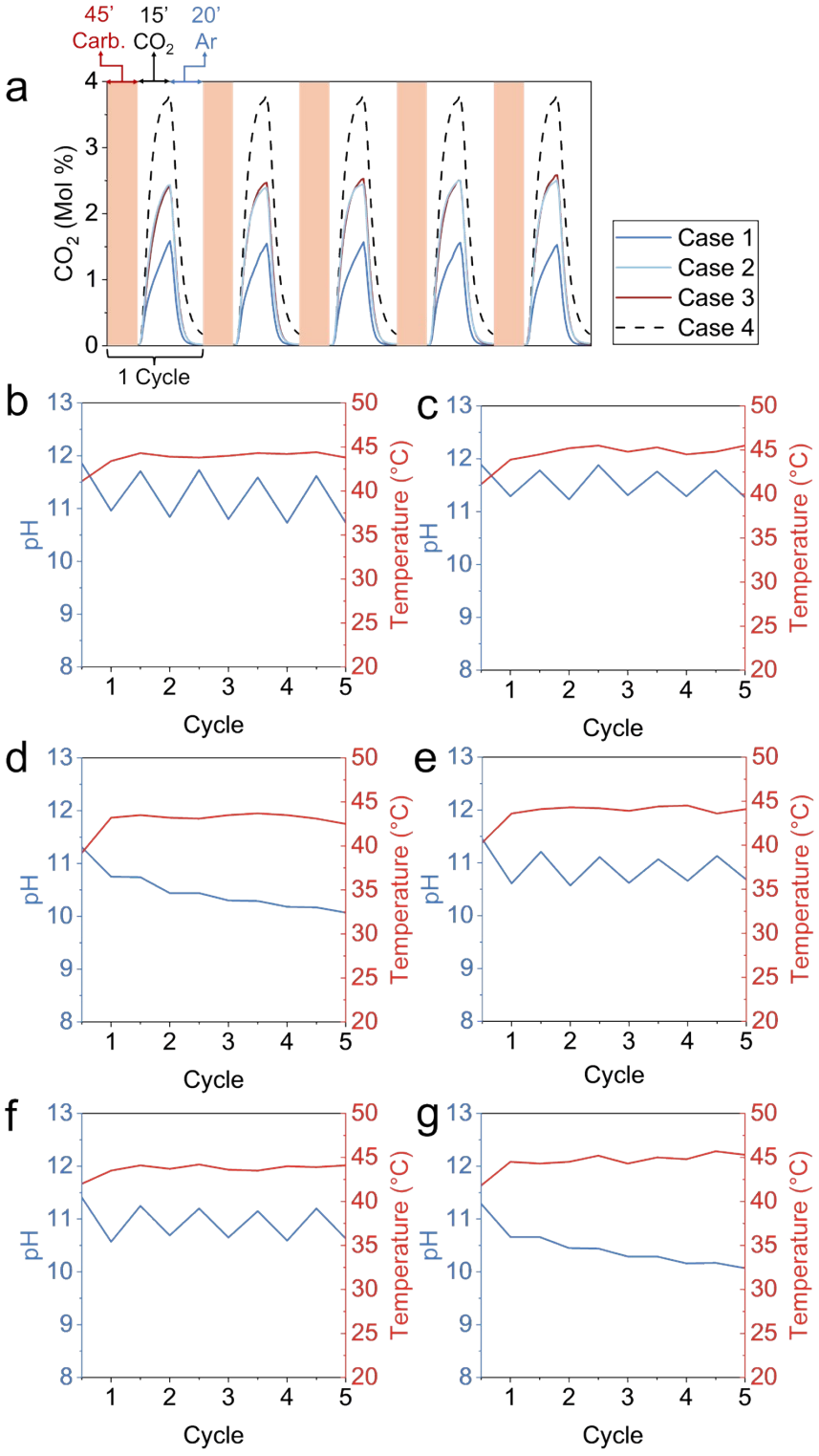

Fig. S19 *(a) 5 cycles of Case 1-4 point-source CO_2 capture with slag fines at 45°C. 0.5 g of steel slag per cycle with 90 sccm 4% CO_2/Ar. pH and temperature in the first (b) and second (c) bottle of Case 1. pH and temperature in the first (d) and second (e) bottle of Case 2. pH and temperature in the first (f) and second (g) bottle of Case 3.*

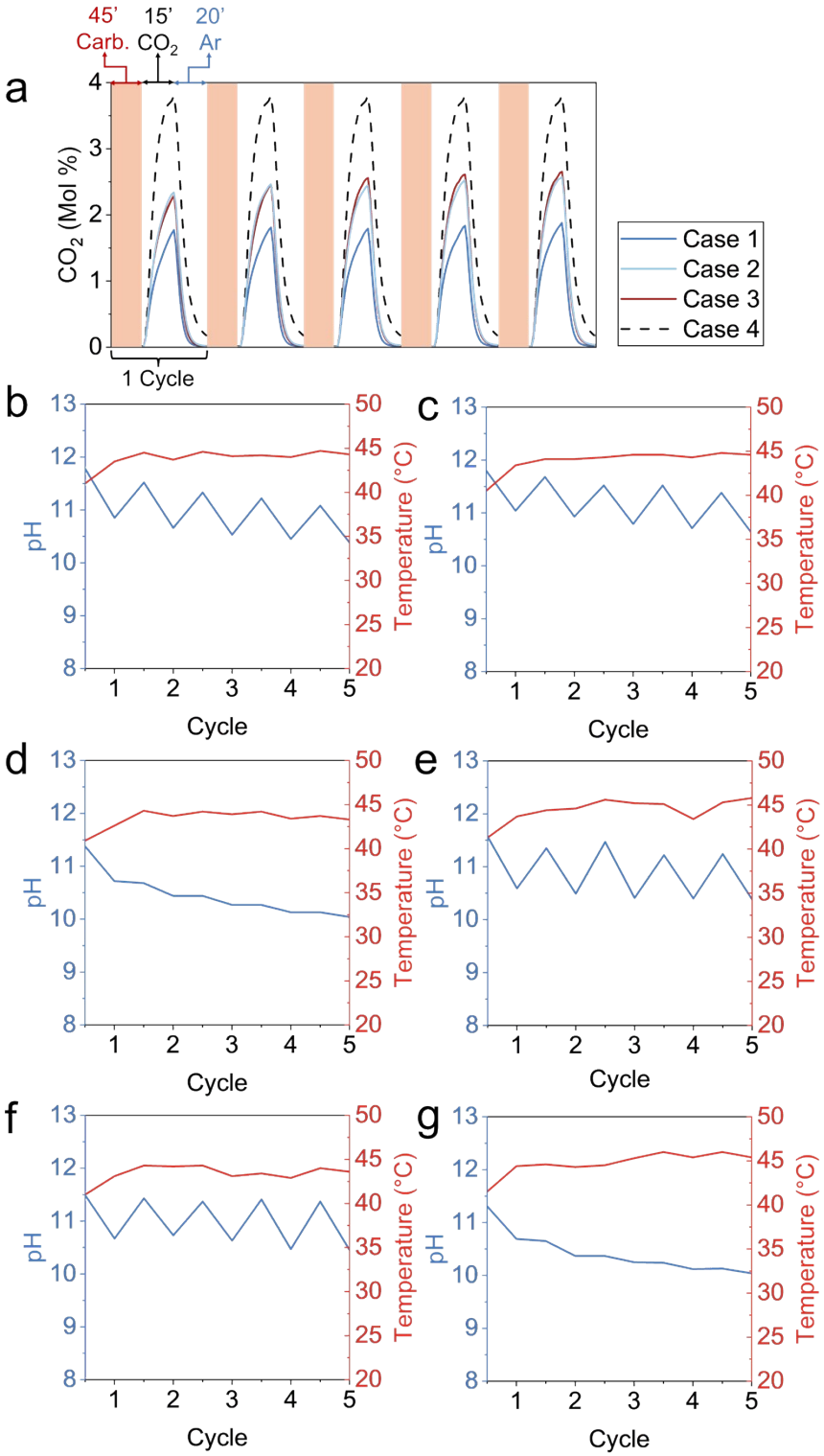

Fig. S20 *(a) 5 cycles of Case 1-4 point-source CO_2 capture with aged fines at 45°C. 0.5 g of steel slag per cycle with 90 sccm 4% CO_2/Ar. pH and temperature in the first (b) and second (c) bottle of Case 1. pH and temperature in the first (d) and second (e) bottle of Case 2. pH and temperature in the first (f) and second (g) bottle of Case 3.*

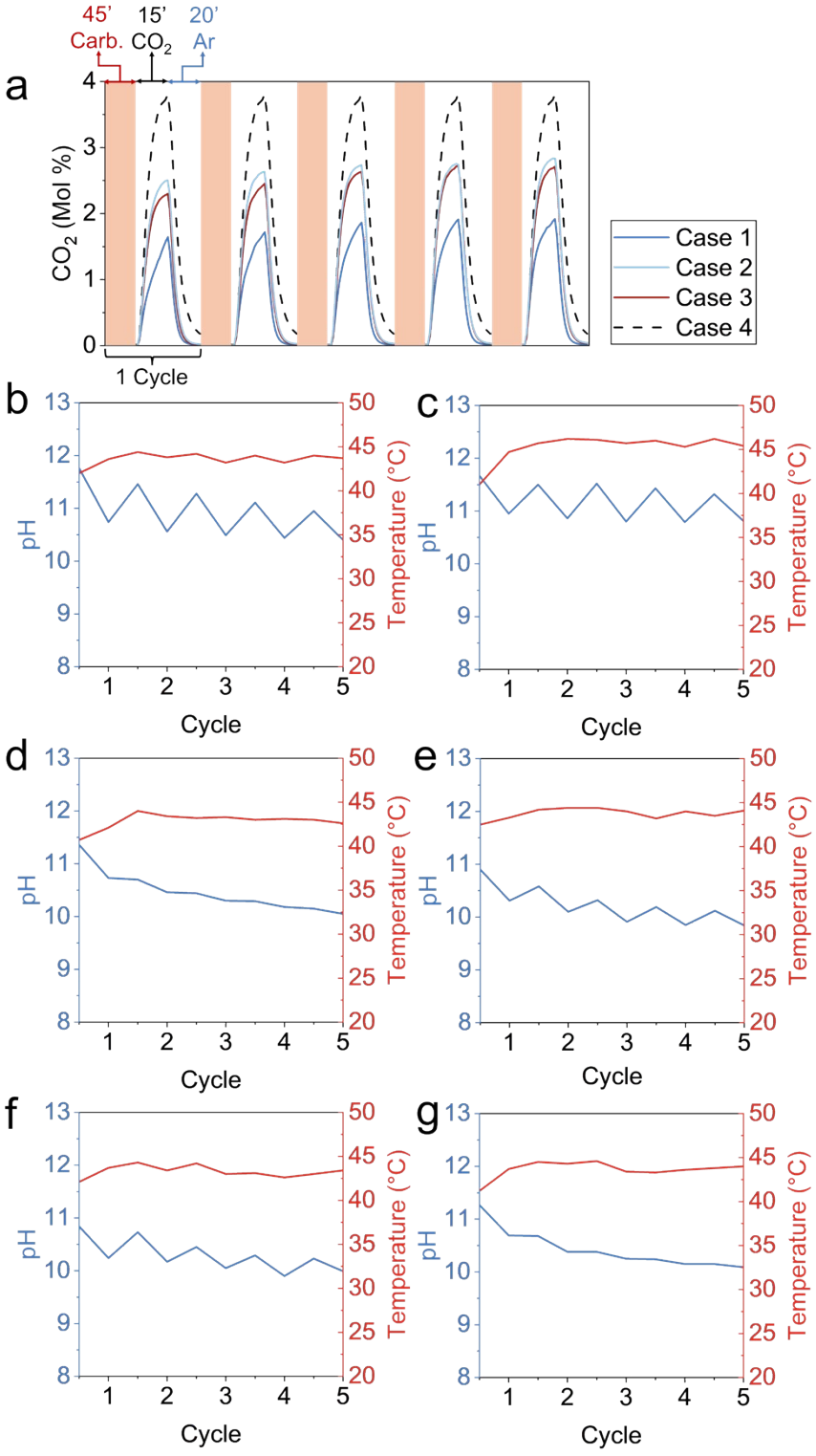

Fig. S21 *(a) 5 cycles of Case 1-4 point-source CO_2 capture with aggregate non-metallic BOF slag at 45°C. 0.5 g of steel slag per cycle with 90 sccm 4% CO_2/Ar. pH and temperature in the first (b) and second (c) bottle of Case 1. pH and temperature in the first (d) and second (e) bottle of Case 2. pH and temperature in the first (f) and second (g) bottle of Case 3.*

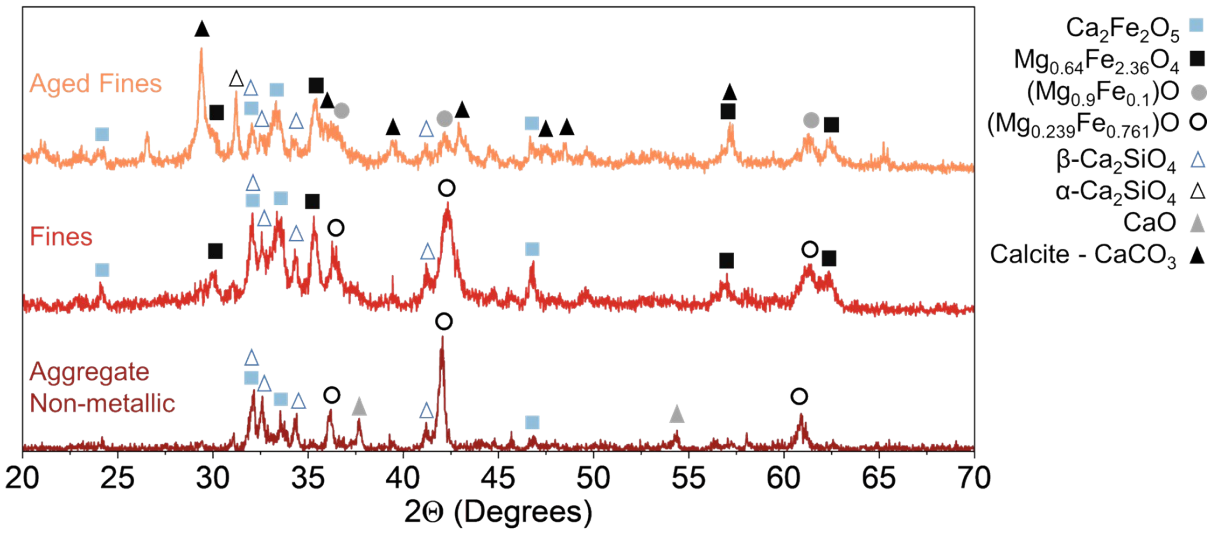

Fig. S22 *XRD of aggregate non-metallic, fines, and aged fines BOF steel slags with phase identification.*

Table S1 *ICP-OES analysis of quenched steel slag.*

Element	Ca	Si	Fe	Al	Mg
Mass %	41.00	27.30	15.23	2.76	10

Table S2 *ICP-OES analysis of amorphous SiO_2.*

Element	Ca	Fe	Al	Mg	Na
Mass %	0.59	0	2.7	0	0

Table S3 *Quenched BOF steel slag phases determined by XRD refinement.*

Phase	Content (wt%)
CaO	12.45
$Ca_2Fe_2O_5$	8.64
Ca_2SiO_4	17.04
Ca_3SiO_5	21.96
$Mg_{0.8}Fe_{0.2}O$	39.91

Table S4 *Calculated actively reacted phases per gram of quenched steel slag (mol/g-slag)*

Phase	Point-Source Looping	Point-Source Baseline	Air CO_2 Looping	Air CO_2 Baseline
Ca_3SiO_5	0.000379	0.00007866	0.0001127	0.0000532
Ca_2SiO_4	0.00039	0.00008091	0.0001159	0.0000547
CaO	0.000874	0.00018156	0.0002601	0.0001228

Table S5 *Process mass balance (tons/h) for annually 100,000 tons of CO_2 captured and mineralized. Inlet steel slag components labeled with "*". Outlet carbonate-rich product components labeled with "#". Outlet hydrated slag components labeled with "+".*

Na_2CO_3 Looping			Baseline		
Component	Point-Source	Air	Component	Point-Source	Air
Inlet Flow			Inlet Flow		
Steel Slag	92.97	312.51	Steel Slag	762.90	1127.76
FeO *	15.19	51.06	*FeO* *	124.65	184.27
Ca_3SiO_5 *	8.04	8.04	Ca_3SiO_5 *	13.70	13.70
Ca_2SiO_4 *	6.24	6.24	Ca_2SiO_4 *	10.63	10.63
CaO *	4.56	4.56	*CaO* *	7.77	7.77
Inactive slag *	58.94	242.61	*Inactive slag* *	606.15	911.39
H_2O(g) (Step 1)	1.27	4.27	H_2O(g) (Step 1)	10.42	15.40
CO_2	11.42	11.42	CO_2	11.42	11.42
H_2O(l) (SiO_2 Wash In)	6.44	6.44	Makeup H_2O	8.77	8.77
Intermediate Flow			Intermediate Flow		
Na_2CO_3	232.42	312.51	$Ca(OH)_2$	19.22	19.22
H_2O(l) (Step 2/3)	4648.47	31251.18	H_2O(l) (Step 2/3)	38144.82	112776.00
Na_2SiO_3	8.72	8.72	Product Flow		
NaOH	15.03	15.03	H_2	1.17	1.72
H_2SiO_3	5.58	5.58	Hydrated Slag	766.38	1135.67
H_2O(g) (Step 4)	1.29	1.29	*CSH* +	26.32	26.32
Product Flow			Fe_3O_4 +	133.91	197.95
H_2	0.14	0.48	*Inactive slag* +	606.15	911.39
Carbonate-rich product	101.22	323.43	$CaCO_3$	25.96	25.96
Fe_3O_4 #	16.32	54.85	Total Mass In/Out	793.50	1163.35
$CaCO_3$ #	25.96	25.96			
Inactive slag #	58.94	242.61			
SiO_2	4.29	4.29			
H_2O(l) (SiO_2 Wash Out)	6.44	6.44			
Total Mass In/Out	112.09	334.63			

Table S6 *Real Steam Rankine cycle state values.*

State	T (°C)	P (bar)	h (kJ/kg)
1-Saturated Liquid	45.82	0.1	191.83
2-Compressed Liquid	46.27	20	195.48
3-Superheated Vapor	312	20	3050.5
4-Wet Steam	45.8	0.1	2245

Table S7 *Lifespan for process equipment.*

Equipment	Lifespan (years)	Equipment	Lifespan (years)
Gas Catalytic Reactor	25 [1]	Pressure Vessel/Open Tank	30 [2]
Belt Conveyor	20 [3]	Shell and Tube Heat Exchanger	25 [4]
Mills	25 [5]	Cooling Tower	25 [4]
Basin/Propeller	30	Solar PV	30 [6]
Filters	20 [7]	Wind Turbine	30 [8]
Pump	15 [9]	Battery	15 [10]
Gas Contactor System	30 [11]	Steam Turbine	50 [12]
Tray Dryer	25 [13]	Condenser	30 [14]

Table S8 *Cost components by category.*

Cost Category	**Components**
Labor	- 2 laborers for overall process - 1 laborer for internal electricity generation (if applicable)
Material Handling & Processing	- Belt conveyor - Mill - Filters/Vacuum - SiO_2 Washer
Reactors	- H_2 production reactor - Carbonation reactor/Propeller - Gas or Air Contactor System - Dryer
Thermal Management	- Heat exchanger - Cooling tower - Increased size for steam power plant condenser (if applicable)
Land & Materials	- Land for processing facility - Land for electricity generation (if applicable) - Na_2CO_3 (capital and replenishment twice annually) - Water (capital and operating costs)
Maintenance	- Process equipment maintenance - Electricity generation equipment maintenance (if applicable)
Electricity & Storage	- Grid electricity (operating expense) - Equipment for on-site electricity generation (if applicable) - Battery storage system (if applicable)

Table S9 *Point-source Na_2CO_3 looping installed cost and related factors for process equipment.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
1	Gas Catalytic Reactor	31	m^3	0.52	3.6 (316 s/s)	4	0.54	$176,983	$707,934	$1,788,464
2	Belt Conveyor	1533	m	0.2	1.3 (316 s/s)	2.5	0.9	$68,727	$171,819	$614,930
3	Mills, Grinding Circuit	94	Mg/h	0.65	1	1	1	$2,406,205	$2,406,205	$6,078,833
4a	Basin	2349	m^3	0.74	1	4	1	$1,110,576	$4,442,305	$11,222,664
4b	Propeller	235	kW	0.52	1	2.5	1	$96,147	$240,368	$607,245
5a	Rotary Disk Filter	435	m^2	0.55	1	2.5	1	$552,734	$1,381,835	$4,945,515
6	Gas Contactor System	61	m^2	-	-	-	-	-	$316,773	$633,546
7a	Rotary Disk Filter	433	m^2	0.55	1	2.5	1	$550,761	$1,376,903	$4,927,865
5/7 b	Rotary Vane Vacuum Pump	1304	dm^3/s	0.38	1	4	1	$34,589	$138,356	$1,281,614
8	Tray Dryer	80	m^2	0.56	1	2.5	1	$37,593	$93,982	$237,428
9	Open Tank, Jacketed with Agitation	4.3	m^3	0.53	1	2.5	1	$32,871	$82,177	$207,604
10	Rotary Drum Filter, Vacuum	0.001	m^2	0.45	1	2.5	1	$82	$204	$730
11	Shell and Tube Heat Exchanger	395	m^2	0.71	2.4 (s/s)	3.5	0.64	$206,576	$723,016	$1,826,567
12	Cooling Tower	620	L/s	0.64	1	3.5	1	$539,612	$1,888,644	$4,771,310
	Solar PV	2407	kW	-	-	-	-	-	$16,941,933	$33,883,866
	Wind Turbine	2407	kW	-	-	-	-	-	$9,158,163	$18,316,326
	Battery	38504	kWh	-	-	-	-	-	$3,542,404	$14,169,617

Table S10 *Point-source Na_2CO_3 looping installed cost and related factors for process equipment within and changed by steam power cycle.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
2	Belt Conveyor	1194	m	0.2	1.3 (316 s/s)	2.5	0.9	$60,659	$151,647	$542,737
11	Shell and Tube Heat Exchanger	507	m^2	0.71	2.4 (s/s)	3.5	0.64	$246,979	$864,428	$2,183,817
12	Cooling Tower	798	L/s	0.64	1	3.5	1	$693,986	$2,428,949	$6,136,293
	Single Valve, Single Stage Steam Turbine	2407	kW	0.51	1	2.5	1	$183,033	$457,583	$674,332
	Pump Centrifugal	11	kW	0.29	1.93 (316 s/s)	4	0.7	$7,504	$30,016	$139,023
	Direct Contact Barometric Condenser	178	L/s	0.6	1	2.5	1	$4,006	$10,016	$25,304
	Vertical Pressure Vessel	14.4	m^3	0.52	2 (316 s/s)	4	0.69	$96,876	$387,504	$978,956

Table S11 *Point-source baseline installed cost and related factors for process equipment.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
1	Gas Catalytic Reactor	127	m^3	0.52	3.6 (316 s/s)	4	0.54	$368,759	$1,475,034	$3,726,403
2	Belt Conveyor	11687	m	0.2	1.3 (316 s/s)	2.5	0.9	$289,126	$722,814	$2,586,913
3	Mills, Grinding Circuit	772	Mg/h	0.65	1	1	1	$16,771,726	$16,771,726	$42,370,677
4a	Basin	19200	m^3	0.74	1	4	1	$5,257,149	$21,028,596	$53,124,874
4b	Propeller	1922	kW	0.52	1	2.5	1	$786,844	$1,967,111	$4,969,544
5a	Rotary Disk Filter	3560	m^2	0.55	1	2.5	1	$4,528,203	$11,320,508	$40,515,502
6	Gas Contactor System	475	m^2	-	-	-	-	-	$2,459,157	$4,918,314
7a	Rotary Disk Filter	3,533	m^2	0.55	1	2.5	1	$4,100,513	$10,251,283	$36,688,801
5/7 b	Rotary Vane Vacuum Pump	10,679	dm^3/s	0.38	1	4	1	$243,020	$972,080	$9,004,534
8	Shell and Tube Heat Exchanger	426	m^2	0.71	2.4 (s/s)	3.5	0.64	$218,187	$763,654	$1,929,232
9	Cooling Tower	670	L/s	0.64	1	3.5	1	$582,815	$2,039,853	$5,153,313
	Solar PV	25289	kW	-	-	-	-	-	$178,037,601	$356,075,201
	Wind Turbine	25289	kW	-	-	-	-	-	$96,240,338	$192,480,676
	Battery	161852	kWh	-	-	-	-	-	$37,226,044	$148,904,175

Table S12 *Point-source baseline installed cost and related factors for process equipment within and changed by steam power cycle.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
2	Belt Conveyor	9894	m	0.2	1.3 (316 s/s)	2.5	0.9	$266,019	$665,049	$2,380,174
8	Shell and Tube Heat Exchanger	1612	m^2	0.71	2.4 (s/s)	3.5	0.64	$561,219	$1,964,267	$4,962,358
9	Cooling Tower	2535	L/s	0.64	1	3.5	1	$1,577,676	$5,521,866	$13,949,978
	Single Valve, Single Stage Steam Turbine	25289	kW	0.51	1	2.5	1	$1,726,514	$4,316,285	$6,360,841
	Pump Centrifugal	115	kW	0.29	1.93 (316 s/s)	4	0.7	$46,572	$186,289	$862,814
	Direct Contact Barometric Condenser	1866	L/s	0.6	1	2.5	1	$282,754	$706,884	$1,785,813
	Vertical Pressure Vessel	69.4	m^3	0.52	2 (316 s/s)	4	0.69	$219,694	$878,774	$2,220,061

Table S13 *Air CO_2 Na_2CO_3 looping installed cost and related factors for process equipment.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
1	Gas Catalytic Reactor	26	m^3	0.52	3.6 (316 s/s)	4	0.54	$161,680	$646,718	$1,633,814
2	Belt Conveyor	7988	m	0.2	1.3 (316 s/s)	2.5	0.9	$199,962	$499,905	$1,789,134
3	Mills, Grinding Circuit	316	Mg/h	0.65	1	1	1	$6,870,346	$6,870,346	$17,356,663
4a	Basin	7810	m^3	0.74	1	4	1	$2,701,955	$10,807,821	$27,303,968
4b	Propeller	781	kW	0.52	1	2.5	1	$319,693	$799,233	$2,019,115
5a	Rotary Disk Filter	2883	m^2	0.55	1	2.5	1	$3,667,031	$9,167,577	$32,810,276
6	Gas Contactor System	7611	m^2	-	-	-	-	-	$39,420,400	$78,840,800
7a	Rotary Disk Filter	2883	m^2	0.55	1	2.5	1	$3,667,031	$9,167,577	$32,810,276
5/7 b	Rotary Vane Vacuum Pump	8651	dm^3/s	0.38	1	4	1	$196,867	$787,467	$7,294,432
8	Tray Dryer	40	m^2	0.56	1	2.5	1	$25,499	$63,748	$161,048
9	Open Tank, Jacketed with Agitation	2.1	m^3	0.53	1	2.5	1	$22,765	$56,912	$143,777
10	Rotary Drum Filter, Vacuum	0.079	m^2	0.45	1	2.5	1	$8,153	$20,384	$72,952
	Solar PV	18786	kW	-	-	-	-	-	$132,255,320	$264,510,640
	Wind Turbine	18786	kW	-	-	-	-	-	$71,492,183	$142,984,366
	Battery	300580	kWh	-	-	-	-	-	$27,653,385	$110,613,540

Table S14 *Air CO_2 Na_2CO_3 looping installed cost and related factors for process equipment within and changed by steam power cycle.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
2	Belt Conveyor	7127	m	0.2	1.3 (316 s/s)	2.5	0.9	$188,873	$472,182	$1,689,915
	Shell and Tube Heat Exchanger	2699	m^2	0.71	2.4 (s/s)	3.5	0.64	$882,609	$3,089,131	$7,804,122
	Cooling Tower	1385	L/s	0.64	1	3.5	1	$1,071,719	$3,751,016	$9,476,251
	Single Valve, Single Stage Steam Turbine	18786	kW	0.51	1	2.5	1	$1,282,542	$3,206,354	$4,725,154
	Pump Centrifugal	86	kW	0.29	1.93 (316 s/s)	4	0.7	$34,596	$138,385	$640,942
	Direct Contact Barometric Condenser	1386	L/s	0.6	1	2.5	1	$210,044	$525,109	$1,326,592
	Vertical Pressure Vessel	35.5	m^3	0.52	2 (316 s/s)	4	0.69	$154,946	$619,784	$1,565,771

Table S15 *Air CO_2 baseline installed cost and related factors for process equipment.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
1	Gas Catalytic Reactor	94	m^3	0.52	3.6 (316 s/s)	4	0.54	$315,121	$1,260,483	$3,184,379
2	Belt Conveyor	28006	m	0.2	1.3 (316 s/s)	2.5	0.9	$483,967	$1,209,917	$4,330,228
3	Mills, Grinding Circuit	1141	Mg/h	0.65	1	1	1	$24,792,987	$24,792,987	$62,634,914
4a	Basin	28288	m^3	0.74	1	4	1	$7,003,258	$28,013,034	$70,769,769
4b	Propeller	2829	kW	0.52	1	2.5	1	$1,157,929	$2,894,823	$7,313,237
5a	Rotary Disk Filter	10483	m^2	0.55	1	2.5	1	$13,335,282	$33,338,205	$119,315,682
6	Gas Contactor System	28040	m^2	-	-	-	-	-	$145,233,053	$290,466,105
7a	Rotary Disk Filter	10443	m^2	0.55	1	2.5	1	$13,284,366	$33,210,915	$118,860,118
5/7 b	Rotary Vane Vacuum Pump	31449	dm^3/s	0.38	1	4	1	$715,679	$2,862,717	$26,517,802
	Solar PV	84614	kW	-	-	-	-	-	$595,682,532	$1,191,365,063
	Wind Turbine	84614	kW	-	-	-	-	-	$322,003,262	$644,006,525
	Battery	1353824	kWh	-	-	-	-	-	$124,551,802	$498,207,208

Table S16 *Air CO_2 baseline installed cost and related factors for process equipment within and changed by steam power cycle.*

#	Cost Item	Size	Units	n	F_m	F_i	$f_{i,alloy}$	FOB Cost	Installed Cost	Total Cost – 50 years
2	Belt Conveyor	25047	m	0.2	1.3 (316 s/s)	2.5	0.9	$457,685	$1,144,213	$4,095,079
	Shell and Tube Heat Exchanger	9501	m^2	0.71	2.4 (s/s)	3.5	0.64	$3,106,692	$10,873,422	$27,469,699
	Cooling Tower	4876	L/s	0.64	1	3.5	1	$2,398,060	$8,393,208	$21,203,895
	Single Valve, Single Stage Steam Turbine	66126	kW	0.51	1	2.5	1	$4,514,414	$11,286,034	$16,632,050
	Pump Centrifugal	301	kW	0.29	1.93 (316 s/s)	4	0.7	$121,775	$487,101	$2,256,048
	Direct Contact Barometric Condenser	4880	L/s	0.6	1	2.5	1	$739,332	$1,848,330	$4,669,466
	Vertical Pressure Vessel	55.9	m^3	0.52	2 (316 s/s)	4	0.69	$196,263	$785,052	$1,983,289

Table S17 *Levelized cost of electricity (LCOE) for all scenarios (Units: $/MWh). The LCOE for internal electricity sources varies across process scenarios because each scenario assumes the same labor input, which does not scale proportionally with equipment sizing. * For the baseline air CO_2 scenario, additional electricity supplied by the solar grid is needed to satisfy the overall energy requirements.*

Electricity Source	Point-Source Looping	Point-Source Baseline	Air CO_2 Looping	Air CO_2 Baseline
Solar Grid	58	58	58	58
Solar PV + Battery	153	138	139	137
Wind + Battery	116	101	102	100
Steam Turbine	26	8	6	19*

Hydrogen Production

H_2 was produced by reacting steam with BOF slags from Cleveland-Cliffs – Cleveland Works. H_2 production experiments were conducted using 6 grams of BOF slag at 600°C, with a mixture of 2 sccm H_2O vapor and 98 sccm Ar. To determine the optimal temperature for H_2 production, experiments were conducted from 500 to 1000°C, revealing the highest production at 650°C (Fig. S2) demonstrating a similar trend to Li et al.[15] H_2 production is shown through water conversion in Fig. S3a, with oxidation of iron in slag (Fig. S3b). The aggregate non-metallic slag produced the most H_2, yielding 1.01 mmol-H_2 g^{-1}, followed by quenched slag with 0.76 mmol-H_2 g^{-1}. Slag fines and aged fines produced less H_2, with 0.34 and 0.07 mmol-H_2 g^{-1}, respectively.

The varying amounts of H_2 produced from different BOF slags highlight the important role of slag handling. Quenched slag, removed from the BOF and cooled as approximately 0.15 m chunks, and aggregate non-metallic slag, separated into chunks larger than 3/8", have small surface areas, limiting oxidation of reduced iron during cooling and storage and thus giving high H_2 production.

Technoeconomic Analysis Methods

For the technoeconomic analysis, four major cases were considered. Point-source Na_2CO_3 looping, point-source baseline, air CO_2 Na_2CO_3 looping, and air CO_2 baseline. Due to the complexity of the system, the process design is first outlined in the following sections for point-source looping, followed by additional sections highlighting the adjustments made for the other cases. The assumptions from the first case hold true unless stated otherwise.

TEA – Point-Source Looping

Process Chemistry and Thermodynamics

In the blast furnace-BOF steel making process, iron ore (Fe^{3+}) is reduced to iron in the blast furnace and then transferred to the BOF where O_2 is impinged at high pressure to further remove carbon. During the BOF process, fluxes are added to the steel to remove additional impurities. The major flux, CaO, reacts with SiO_2 to form various types of calcium silicates, mainly Ca_2SiO_4 and Ca_3SiO_5, which float to the top of the melt in the BOF furnace where they can be separated from the steel. Some iron (Fe^{2+}) from the steel is separated along with the calcium silicates due to incomplete reduction from the blast furnace or oxidation during the BOF process. After the steel slag is separated, the process studied in this paper begins.

The process begins with H_2 production by reacting steam with the steel slag at 600°C (Step 1). As outlined in Eqn S1, water vapor reacts with part of Fe^{2+} to form H_2 and Fe^{3+}. The exothermic reaction can be directly carried out by flowing water over high temperature slag which uses remaining heat from the BOF furnace to form high temperature steam. The oxidized iron remains an unreactive component through the remaining process, making the first step optional. As a simplified reaction equation neglecting other cations mixed with Fe ions in slag, we can write

$$3FeO(s) + H_2O(g) \rightarrow Fe_3O_4(s) + H_2(g);\ \Delta H(600°C) = -55.72\frac{kJ}{mol} \quad \text{(S1)}$$

The steel slag can then be cooled and milled into a powder which is fed into the cyclic carbonation and CO_2 capture steps. The carbonation step (Step 2) considers an aqueous solution of Na_2CO_3 reacting with Ca_3SiO_5, Ca_2SiO_4, and CaO at mild temperatures such as room temperature and 70°C, as shown in Eqns S2-4.

$$3Na_2CO_3(aq) + Ca_3SiO_5(s) + 2H_2O(l) \rightarrow Na_2SiO_3(aq) + 3CaCO_3(s) + 4NaOH(aq);\ \Delta H(70°C) = -189.15\frac{kJ}{mol} \quad \text{(S2)}$$

$$2Na_2CO_3(aq) + Ca_2SiO_4(s) + H_2O(l) \rightarrow Na_2SiO_3(aq) + 2CaCO_3(s) + 2NaOH(aq);\ \Delta H(70°C) = -139.78\frac{kJ}{mol} \quad \text{(S3)}$$

$$Na_2CO_3(aq) + CaO(s) + H_2O(l) \rightarrow CaCO_3(s) + 2NaOH(aq);\ \Delta H(70°C) = -5.25\frac{kJ}{mol} \quad \text{(S4)}$$

The $CaCO_3$ and other components in the carbonate-rich product can be filtered and removed from the solution. The remaining basic solution can then react with CO_2 in a flue gas or in air. Both the Na_2SiO_3 and NaOH can capture CO_2 at low temperatures to regenerate Na_2CO_3 (Eqns S5 and S6). It is important to note that the reaction enthalpy of CO_2 with NaOH is quite high which will result in low grade waste heat that will need to be managed. In addition, a portion of the water consumed in Eqns S2-4 will be regenerated in Eqn S6.

$$2CO_2(g) + 2Na_2SiO_3(aq) + 2H_2O(l) \rightarrow 2H_2SiO_3(s) + 2Na_2CO_3(aq);\ \Delta H(70°C) = -159.47\frac{kJ}{mol} \quad \text{(S5)}$$

$$4CO_2(g) + 8NaOH(aq) \rightarrow 4H_2O(l) + 4Na_2CO_3(aq);\ \Delta H(70°C) = -431.17\frac{kJ}{mol} \quad \text{(S6)}$$

The final step is to filter the H_2SiO_3 from the solution and dry it to form amorphous SiO_2 (Eqn S7), so that the remaining water consumed in Eqns S2-4 is regenerated. The amorphous SiO_2 can then be washed with water and sold. From this point, additional steel slag can be added to the Na_2CO_3 solution, and the cycle can continue.

$$2H_2SiO_3(s) \rightarrow 2SiO_2(s) + 2H_2O(g);\ \Delta H(100°C) = 72.87\ kJ/mol \quad \text{(S7)}$$

The reaction enthalpies were determined using the View Data function in FactSage 8.3, which provided enthalpy values for individual phases.[16,17] Solution enthalpy values not found in FactSage - Na_2CO_3, Na_2SiO_3, NaOH, and $Ca(OH)_2$ – are referenced from NIST[18]. Both data sources use the same standard reference state of 298 K and 1 bar for enthalpy values, ensuring consistency when combining FactSage and NIST thermochemical data.

Mass and Energy balance

To quantify the molar content of active phases in steel slag during the reactions, a combination of XRD refinement and TGA was employed. First, Rietveld refinement of XRD data was performed to determine the fraction of crystalline phases present in the slag (Table S3). Ca_3SiO_5, Ca_2SiO_4, and CaO are considered active components in the carbonation reaction. $Mg_{0.8}Fe_{0.2}O$ is reactive in H_2 production and $Ca_2Fe_2O_5$ is unreactive. Since amorphous phases cannot be directly quantified through XRD, their contribution was indirectly accounted for using TGA. The total amount of CO_2

mineralized during carbonation was determined by measuring the mass loss associated with $CaCO_3$ decomposition in TGA – 2.79 mmol CO_2 per gram of slag. The CO_2 mineralized was determined for the carbonation of quenched slag at 45°C with 4% CO_2 (Figs. 2a and S5a). By correlating our experimental CO_2 uptake with relative fractions of calcium-containing phases via Eqns S2-6, the absolute fractions of reacted Ca_3SiO_5, Ca_2SiO_4, and CaO were estimated. These fractions were then normalized to 1 g to obtain the molar quantities of each reacted phase in 1 g slag (Table S4).

The mass balance was initiated with the calculated amounts of Ca_3SiO_5, Ca_2SiO_4, and CaO. The Na_2CO_3 and water amounts were directly scaled from experiments with mass ratios of 2.5:1 Na_2CO_3 to slag and 50:1 water to slag. From there, the amount of FeO in the steel slag was assumed based on experimental H_2 production of 16.99 std. mL H_2/g slag. Then 100,000 tons of CO_2 captured annually was input into the system. A wash ratio (mass of water to mass of solid) of 1.5 was used to calculate the amount of water required to wash the amorphous SiO_2. The mass change of water after washing the SiO_2 was assumed to be negligible. Using these inputs the mass balance was completed and reduced to mass per hour (Table S5). The mass in and out of the system are highlighted in Fig. 3a along with the process diagram.

Heat balance in the reactors was calculated by considering the heat loss from natural convection from the reactor and heat generation or consumption due to exo/endothermic reaction. The cycle time was assumed to be 0.5 h based on the experimental procedure. Using the mass flow rate of materials, the reactors were sized based on volume, and the reactors were assumed to be a cube to estimate surface area interfacing with the environment. The density of steel slag and carbonated steel slag (carbonate-rich product), were assumed to be 3000 kg/m^3 and 3350 kg/m^3 respectively.[19,20] The density of silicic acid was assumed to be close to that of silica gel 600 kg/m^3 .[21] The density of the solution phase was calculated from Hitchcock and McIlhenny[22], and the effect of Na_2CO_3 and NaOH on density of solution were considered. Once the surface area was calculated, the heat loss to environment could be estimated using an assumed overall heat transfer coefficient of 0.003 kW/(m^{2}K) which was related to an insulated reactor into still air. [23] The heat generated or consumed during the reaction was calculated using the reaction enthalpy and mols of feedstock in the reactor. The active cooling of a reactor via cooling water or active heating of a reactor was then calculated by considering both the heat loss via reactor wall and the reaction heats.

Heat transfer for materials between reactors was calculated using literature enthalpy data or specific heat data. The specific heat of steel slag was assumed to be 935 J/kgK.[24] Steel slag exiting the BOF at 1650°C is allowed to naturally cool to 653°C. At 653°C, the heat available is enough to generate steam from the flow of water into Step 1. The steam reacted slag leaving Step 1 must then be cooled down to 70°C which is required for Step 2 and 3. The temperature for Step 2 and 3 was increased from 45°C in our experiments to 70°C to prevent a small temperature difference in the heat exchanger to dissipate the reaction heat, which would otherwise require a larger surface area and increase costs. At 70°C, performance is expected to be similar, or better than, the experimental results at 45°C. The heat from the steel slag between Steps 1 and 2 can then be used to power a steam power cycle and dry the silicic acid in Step 4. Milling and filtering are assumed to happen at 70°C. After the second filtering process, the silicic acid must be heated from 70°C to 100°C and then the water vapor that forms must be cooled to 70°C for return to Step 2. The specific

heat of silicic acid was calculated from FactSage as 1050 J/kgK.[16,17] Additionally, the flue gas (point-source CO_2) was assumed to enter the system at 83°C coming from a natural gas combined cycle power plant.[25] The flue gas releases heat to the Step 3 reaction and was assumed to have a specific heat of 1050 J/kgK.[26]

The heat transfer components are highlighted in Fig. 3a. The H_2 production reactor does not involve active heat dissipation or supply because the heat loss to the environment was about equal to exothermic heat generation by altering the reaction time and reactor volume for H_2 production. The waste heat from the steel slag on the conveyor is used to dry the silicic acid while releasing the remaining heat to the atmosphere. The heat exchanger on the carbonation reactor handles the exothermic reaction heat and heat release from the condensation of water vapor from the dryer. The analysis is for steady state operation and thus initial heating of water and Na_2CO_3 feedstock are ignored. The heat exchanger on the CO_2 capture spray tower also handles exothermic reaction heat as well as sensible heat from the flue gas.

Electric energy input for the process was calculated for the conveyor belt (between Step 1 and milling), milling, filtering, stirring, pumping solution (between Step 2 and 3), and fan-driven gas flow which were determined to be the major energy consumption processes. Energy input for transporting water vapor from Step 4 to 2 was determined to be negligible. The power requirement of the conveyor belt was 0.72 kW/Mg/h per km of horizontal distance.[23] Milling steel slag was assumed to consume 3.84 kWh/ton [27] and filtering consumes 0.18 kWh/m^3-water.[28] Agitation is required for Step 2 and washing SiO_2 and the energy consumption was assumed to be 0.1 kW/m^3.[23] The energy requirement for the pumps and fans are calculated based on assumptions from Carbon Engineering[29] and are detailed in the *Discussion of Specific Equipment* section. The power requirements for the individual components are shown in Fig. 3a.

Four energy scenarios for providing electricity were considered for each case: 1) Energy purchased from an external solar grid, 2) Internal solar photovoltaic (PV) system with integrated 16-hour battery storage, 3) Internal wind turbines with integrated 16-hour battery storage, and 4) Internal steam power plant powered by waste heat from the steel slag. Energy purchased from an external solar grid was assumed to be \$0.0576/kWh.[30] The internal solar PV system was designed to be in Cincinnati, Ohio (4.5 sun hours) with an inefficiency factor of 1.2. The solar panel cost was assumed as \$1.1/W.[31] To account for intermittency of solar energy, a grid scale battery system was sized for 16 hours of electricity with a cost of \$92/kWh.[32] The internal wind turbines were assumed to have an average installed cost of \$1370/kWh with a capacity factor of 36%.[8] Similarly to solar PV, a 16-hour battery system was included to accommodate the intermittency of wind energy. The steam power plant was designed as a real steam Rankine cycle with isentropic pump and turbine efficiencies of 0.8 and 0.9, respectively.[33,34] The state values are shown in Table S6. The steam power plant is powered by direct contact heat exchange from the steel slag to water as the working fluid directly after leaving the H_2 production reactor.

Capital Cost

Capital cost of the system was estimated using data from engineering design textbooks.[23,35] To estimate the capital cost, the purchase cost, or free on-board cost (FOB cost), was calculated and additional factors were applied. The general equation for capital cost of equipment is shown as Eqn (S8).

$$Cost = \left[(FOB\ cost)_{ref}\left(\frac{size}{size_{ref}}\right)^{n}\frac{CEPCI}{1000}(F_m f_{i,alloy})\right][F_i]\left[\frac{(1+F_o)}{F_u}\right] \quad \text{(S8)}$$

Where the FOB cost of reference equipment is multiplied by a ratio of sizes which corrects for economy of scale by the factor of n. If the size of the designed equipment was outside the range specified for n, then linear scaling was assumed to be conservative. The CEPCI is the Chemical Engineering Plant Cost Index where the CEPCI index is 1000 while the analysis considers the plant being manufactured in 2030 when the CEPCI index will be an assumed 725. F_m is the materials factor to account for purchasing equipment that is designed with materials other than carbon steel. The installation factor, F_i, considers installation costs such as electrical, piping, insulation, painting, concrete, and labor. F_i is independent of construction material for equipment and needs to be lowered by the factor $f_{i,alloy}$. F_o is the offsite investment factor which accounts for improvements to the site infrastructure and is assumed to be 0.4 for a new site.[35] F_u is the plant utilization factor and is assumed to be 0.95 as the plant can operate continuously. In the following sections, the equipment cost without F_i, F_o and F_u is considered the FOB cost while the installed cost includes F_i and not F_o and F_u. The lifespan of the plant was assumed to be 50 years, and the lifespan of equipment was estimated (Table S7) to help determine their replacement schedule. The offsite factor, F_o, is not included in the price of the replacement equipment because additional improvements to the infrastructure are not needed. The FOB cost, installed costs, and total cost over 50 years for the equipment are detailed in Table S9 for electricity cases 1-3.[23,35] When a steam power plant is considered, the cost components for the power cycle are added and the size of the heat exchanger, cooling tower, and conveyor belt are updated (Table S10).

Additional capital costs include Na_2CO_3, water, and land. The initial feedstock of Na_2CO_3 is based on a market price of $1,042/ton.[36] Water is required for Step 2 and 3 reactions as well as the cooling tower. The initial water volume required to fill the cooling tower system consists of the water needed to fill the cooling tower basin and connected piping. A reasonable approximation is to assume that this volume is approximately twice the system's circulating flow rate. Water cost is set as $1.58/ton.[37] The cost of land was estimated as 1% of the installed cost of equipment.[23] Additional land cost was included for the solar PV and wind turbine systems requiring 8.3 acres/MW and 85.25 acres/MW respectively.[38,39] The cost of land was assumed to be $3,343/acre.[40]

Discussion of Specific Equipment

Carbonation Reactor: Due to the high water-to-slag ratio, a large volume of solution is present in the carbonation reactor at any given time. To reduce costs, a concrete basin was sized for the carbonation reaction instead of a metal reactor. It is assumed that if the basin is open to air, any CO_2 uptake from the atmosphere would form silicic acid during carbonation which would be negligible compared to the 4% CO_2 during the capture step. This assumption does not affect the overall process and may even provide additional CO_2 capture from the air. For mixing, top-entry propellers were sized for the open basin, assuming an agitation power requirement of 0.1 kW/m^3.

Gas Contactor System: The design of the gas contactor system follows Carbon Engineering's air-contactor concept.[29] It includes a slab contactor equipped with packing for contacting the flue gas with the absorption solution, along with fans for gas handling and pumps for solution circulation. The capital cost is based on the required contactor frontal area, A, determined from Eqn (S9). The

reacted CO_2 mass flow rate, $\dot{m}_{CO2}$, was assumed to correspond to 100,000 tons of CO_2 captured per year. The gas velocity, V, and density, ρ, were taken as 1.6 m/s and 0.92 kg/m^3, respectively, representing the properties of the flue gas.[41] The CO_2 mass fraction, w_{CO2}, was calculated from a 4% CO_2 concentration in air (corresponding to 6% by mass), and the capture fraction, X_{CO2}, was set to the experimental value of 59%.

$$A = \frac{\dot{m}_{CO_2}}{V\rho w_{CO_2} X_{CO_2}} \tag{S9}$$

The total capital cost of the contactor was determined using Eqn (S10), where C_A is the capital cost per unit frontal area, valued at \$3700/m^2, C_{pack} is the cost of the packing and fluid distributor, taken as \$250/m^3, and D is the packing depth of 8.6 m.

$$C_{capital} = \left(C_A + C_{pack}D\right) * A * F_u \tag{S10}$$

The cost estimates reported in Carbon Engineering's study were originally based on 2008–2009 Alberta prices. To update these costs, conversion from Canadian dollars to U.S. dollars and adjustment to 2030 values were required. An exchange rate of 1.07 Canadian dollars per U.S. dollar was applied for 2008, and a CEPCI value of 575.4 was used for that year. It was also noted that Alberta capital costs during this period were approximately 30–50% higher than those on the U.S. Gulf Coast. To remain conservative in this analysis, the capital costs were therefore reduced by 40% to better represent projected U.S. conditions.

The fan pressure drop, ΔP, and energy requirement, E, were calculated using Eqn (S11) and (S12). The fan efficiency, η_{fan}, was assumed to be 71%, but reduced by 15% (to 56%) to account for additional pump energy.

$$\Delta P = D7.4V^{1.4} \tag{S11}$$

$$E = \Delta PV/\eta_{fan} \tag{S12}$$

Mill: Since the installation cost was already incorporated into the FOB cost, an additional installation factor was omitted.[23]

Filters: The filtration rate was assumed to be 3 L/s/m^2.[23] The selected rotary disk filters did not include a vacuum pump, so a rotary vane vacuum pump was sized separately for a single filter. Since the two filters are of similar size, the calculated pump cost was then multiplied by two to account for both units.

Dryer: The drying rate was assumed to be 0.27 g_{water}/s/m^2 and the capacity was 35 kg/m^2.[23]

Heat exchanger and cooling tower: The overall heat transfer coefficient for the heat exchanger was chosen as 1500 W/m^2/°C for water to water.[35] For cooling reactors, the hot side (reactor) was designed to not change temperature. The minimum achievable temperature by the water was calculated by adding the wet bulb temperature, 23.8°C, to the approach temperature, 5.5°C, which results in 29.3°C.[23] The cooling capacity of the tower was chosen to be 8.3°C which leads to 29.3°C cold fluid out and 37.6°C cold fluid in.[23]

Belt Conveyor: The conveyor was designed to cool slag to the carbonation temperature after H_2 production. The radiative heat from the slag is used to dry the silicic acid. The conveyor belt was designed to have a width of 72 in (1.83 m) and a velocity of 3 m/s.[42,43] The slag was assumed to be a flat plate on the conveyor with a constant rate of cooling. Radiation and convection cooling were considered in the design with ambient conditions considered as air at 25°C. The radiation component was calculated using a steel slag emissivity of 0.81.[44] The convection component was calculated by using the Nusselt correlation for turbulent flow over a flat plate with constant heat flux conditions (Eqn S13).[45] Heat transfer was calculated using the average temperature formula provided in Eqn S14.[46] To determine the Reynolds number and convective heat transfer coefficients, an initial conveyor length of 100 m was assumed and later iteratively updated to match the calculated length. The total conveyor length was determined based on two sequential cooling stages: (1) from the outlet temperature of the H_2 production step to the slag outlet temperature from the steam power plant, and (2) from that steam power plant outlet temperature to the carbonation temperature. For energy scenarios 1–3 (no steam power cycle included), the total conveyor length includes both cooling stages. For the steam power plant scenario, only the second cooling stage was considered.

$$Nu_x = 0.0308 Re_x^{4/5} Pr^{1/3} \tag{S13}$$

$$T_{avg} = \frac{T_{in}^5 - T_{out}^5}{5(T_{in} - T_{out})} \tag{S14}$$

Steam Power Cycle: The steam power cycle was designed with a single-valve, single-stage steam turbine, a centrifugal pump, a direct-contact barometric condenser, and a direct-contact heat exchanger for steam generation. The turbine operates at 20 bar with 100°C superheat, which adds a 0.97 factor to the FOB cost. Steam generation assumes direct contact between the hot steel slag and the working fluid, requiring a vertical pressure vessel as a direct-contact heat exchanger. To size this vessel, the volume of steel slag and steam within the system at any given time was determined. The convective heat transfer coefficient was estimated using the Wakao and Kaguei Nusselt correlation for heat transfer in a packed bed (Eqn S15).[47] The Nusselt number was calculated based on an assumed heat transfer time of 10 minutes, a slag particle diameter of 0.05 m, and a reactor diameter of 1 m. Once the convection coefficient was determined, the actual heat transfer time was calculated. The total slag surface area available for heat transfer was assumed to be the calculated conveyor belt surface area. The final calculated heat transfer time was found to be within the same order of magnitude as the initial assumption. An additional cost factor of 1.15 was included for the pressure vessel to handle 2 MPa.

$$Nu_x = 2 + 1.1 Pr^{1/3} Re^{0.6} \tag{S15}$$

After the steel slag leaves the direct-contact heat exchanger, it is at 245°C before entering the belt conveyor. The heat exchanger and cooling tower were increased in size to handle the heat release from the condenser in the steam power cycle, if included.

Annualized Capital Cost

The capital cost of the plant was calculated using the annualized capital cost formula shown in Eqn S16, where the total capital cost is multiplied by the capital recovery factor which depends on

interest rate, i, and lifespan of the plant, n. The interest rate was assumed to be 5% [48] and the lifespan of the plant was estimated at 50 years.

$$Annualized\ Capital\ Cost = Total\ Capital\ Cost * \left(\frac{i(1+i)^n}{(1+i)^n - 1}\right) \quad \text{(S16)}$$

Operating Costs

The operating costs include maintenance, labor, Na_2CO_3 replenishment, and water inlet for H_2 production and washing SiO_2. Maintenance of equipment was assumed to be 4% of the total installed cost of the equipment annually. Maintenance for solar PV and wind turbines were assumed to be \$22/kW/year and \$39/kW/year, respectively.[30,49] Maintenance for grid scale batteries was assumed to be 2.5% of the capital cost.[50] An operating and maintenance cost equal to 5% of the total capital cost was included for the gas contactor system.[29] For the case of using external solar grid electricity, the plant is operated by 2 laborers per shift. An additional laborer is added per shift for the other cases where electricity is generated on site. The laborers are paid an average wage of \$38.50/hour.[51] Na_2CO_3 is estimated to be replenished 2 times a year for the market price of \$1,042/ton. Water cost for H_2 production and washing SiO_2 is considered \$1.58/ton. Evaporation is considered negligible, and the water is assumed to not need replenishment for Step 2 and 3 or the cooling tower.

Levelized Cost of Electricity

The levelized cost of electricity (LCOE) was calculated by dividing the sum of the annualized capital cost and operating cost by the total annual electricity generation (Eqn S17).

$$LCOE = \frac{Annualized\ Capital\ Cost + Annual\ Operating\ Cost}{Annual\ Electricity\ Generation\ (MWh_e)} \quad \text{(S17)}$$

Revenues

Revenues from H_2, carbonate-rich product, precipitated silica, and the 45Q tax credit were accounted for. The H_2 selling price was assumed to be \$2/kg.[52] The selling price of the point-source Na_2CO_3 looping carbonate-rich product was assumed to be \$26/ton.[53] Precipitated silica has a selling price ranging from \$600 to \$1200 per ton, depending on purity.[54] The price was set at \$600 to be conservative. Finally, the 45Q tax credit provides \$85/ton of CO_2 captured for projects that convert point-source CO_2 into useful products.[55]

Breakeven Time

The breakeven time was calculated by dividing the total capital expenses by yearly profit. The profit is the difference between yearly revenue and yearly operating expenses (Eqn (S18)).

$$Breakeven\ Time\ (years) = \frac{CAPEX}{Yearly\ Revenue - Yearly\ OPEX} \quad \text{(S18)}$$

The breakeven time was calculated for the scenario where electricity is provided from an external solar grid and is shown:

$$CAPEX = Equipment + Land + Water + Na_2CO_3 = \$39{,}144{,}316 + \$142{,}708 + \$14{,}437 + \$242{,}124 = \$39{,}543{,}585$$

$$Revenue = H_2 + Carbonated\ Slag + Silica + 45Q\ Tax\ Credit = \left(\frac{\$2}{kg_{H_2}}\right)\left(1{,}244{,}677\frac{kg_{H_2}}{year}\right) + \left(\frac{\$26}{ton_{carbona}}\right.$$
$$+ \left(\frac{\$600}{ton_{silica}}\right)\left(37{,}599\frac{ton_{silica}}{year}\right) + \left(\frac{\$85}{ton_{CO_2}}\right)\left(100{,}000\frac{ton_{CO_2}}{year}\right) = \$56{,}602{,}824$$

$$OPEX = Maintenance + Labor + Water + Na_2CO_3 + Electricity = \$564{,}980 + \$674{,}520 + \$62{,}28$$
$$= \$3{,}000{,}202$$

$$Breakeven\ Time = \frac{\$39{,}543{,}585}{\frac{\$56{,}602{,}573}{year} - \frac{\$3{,}000{,}202}{year}} = 0.8\ years$$

<u>*TEA – Point-Source Baseline*</u>

Process Chemistry and Thermodynamics

The H_2 production step in the baseline case follows the same reaction as the looping scenario. After H_2 production, the steel slag is cooled and milled into a powder which is fed into cyclic hydration and CO_2 capture steps. The hydration step (Step 2) considers water reacting with Ca_3SiO_5, Ca_2SiO_4, and CaO as shown in Eqns S19-21. The hydration of Ca_3SiO_5, Ca_2SiO_4, forms $3CaO{\bullet}2SiO_2{\bullet}8H_2O$ referred to as calcium silicate hydrate (CSH). The heat of hydration and reaction equations for Ca_3SiO_5 and Ca_2SiO_4 are referenced from Mindess et al.[56]

$$2Ca_3SiO_5(s) + 11H_2O(l) \rightarrow 3CaO \cdot 2SiO_2 \cdot 8H_2O(s) + 3Ca(OH)_2(aq);\ \Delta H = -0.52\frac{kJ}{g - Ca_3SiO_5} \quad \text{(S19)}$$

$$2Ca_2SiO_4(s) + 9H_2O(l) \rightarrow 3CaO \cdot 2SiO_2 \cdot 8H_2O(s) + Ca(OH)_2(aq);\ \Delta H = -0.26\frac{kJ}{g - Ca_2SiO_4} \quad \text{(S20)}$$

$$CaO(s) + H_2O(l) \rightarrow Ca(OH)_2(aq);\ \Delta H(70°C) = -87.2\frac{kJ}{mol} \quad \text{(S21)}$$

The CSH and other components in the hydrated steel slag can be filtered and removed from the solution. The remaining $Ca(OH)_2$ solution can then react with CO_2 in a flue gas or in air at low temperatures to precipitate $CaCO_3$. Only a portion of the water consumed in Eqns S19-21 will be regenerated in Eqn S22 which leads to an overall deficit of water in the cycle which requires replenishment.

$$5CO_2(g) + 5Ca(OH)_2(aq) \rightarrow 5CaCO_3(s) + 5H_2O(l);\ \Delta H(70°C) = -453.1\frac{kJ}{mol} \quad \text{(S22)}$$

The $CaCO_3$ can then be filtered from the water. From this point, additional steel slag and makeup water can be added, and the cycle can continue.

Mass and Energy balance

The molar content of actively reacted phases was quantified in the same way as the point-source looping but using the hydration reaction equations and our experimentally measured 0.34 mmol CO_2 mineralized per gram of slag. The CO_2 mineralized was determined for the baseline carbonation of quenched slag at 45°C with 4% CO_2 (Fig. 2a and S5b). The molar quantities of each phase in the slag are shown in Table S4.

The major differences between the looping and baseline scenario in mass balance include: (1) the baseline produces hydrated slag instead of carbonated slag (carbonate-rich product) in Step 2, (2) the baseline produces precipitated $CaCO_3$ during the CO_2 capture step instead of silicic acid, (3) the process consumes water in the baseline which requires makeup. For direct comparison to the looping scenario the baseline considers the outlet of hydrated slag plus precipitated $CaCO_3$ as the supplementary cementitious material product (carbonate-rich product). The mass balance for the point-source baseline is shown in Table S5 and Fig. S7a.

Active heating or cooling needed for the reactors was once again calculated by considering the heat loss from natural convection and heat generation or consumption due to exo/endothermic reactions. To calculate the reactor size, the density of hydrated steel slag was input as 2604 kg/m^3.[57] It was assumed that $Ca(OH)_2$ concentration has no effect on solution density, as a saturated solution would have a density close to that of water.[58] The density of $CaCO_3$ was assumed to be 2700 kg/m^3.[59]

The heat transfer between the reactors was handled the same as looping scenario; however, there is no H_2SiO_3 production and therefore no drying step is necessary. The heat from the steel slag on the conveyor belt is completely released to the atmosphere or partially used for driving steam power cycle. After the steam power plant, the steel slag leaves at 146°C. In addition, the makeup water is assumed to enter the hydration reactor at 25°C which requires heating to the reaction temperature of 70°C. The heating need is less than the exothermic heat release for Step 2. The heat balance is shown in Fig. S7a.

Capital Cost

The process cost was estimated in the same manner as the looping scenario. For the point-source baseline, a CO_2 conversion of 7% was determined from the CO_2 capture step of baseline quenched slag at 45°C with 4% CO_2 in our experiments (Fig. 2a). The FOB cost, installed costs, and total cost over 50 years for the equipment are detailed in Table S11 for electricity cases 1-3. The updated steam power plant components are listed in Table S12. No Na_2CO_3 is required for the process so there is no capital cost of Na_2CO_3 included.

Operating Costs and Revenue

The operating costs include maintenance, labor, and water inlet for H_2 production and makeup.

Revenues from H_2, carbonate-rich product, and the 45Q tax credit were accounted for. The baseline considers the outlet of hydrated slag plus precipitated $CaCO_3$ as the carbonate-rich product. The selling price of the point-source baseline carbonate-rich product was assumed to be $3.32/ton, which was scaled to have equal yearly revenue for carbonate-rich product as the point-source looping case, considering the mixed effects of larger product quantity and lower product carbonation extent of baseline product than looping.

TEA – Air CO_2 Looping

The process chemistry and thermodynamics for the air CO_2 looping case is the same as the point-source looping. The molar content of actively reacted phases was quantified in the same way as the point-source looping, but with 0.83 mmol CO_2 mineralized per gram of slag. The CO_2 mineralized was determined for the carbonation of quenched slag at 24°C with air CO_2 by our experiment (Fig. 2c and S5c). The molar quantities of each phase in the slag are shown in Table S4.

The mass balance for the air CO_2 looping is shown in Table S5 and Fig. 4a.

The mass ratios of 1:1 Na_2CO_3 to slag and 100:1 water to slag were directly adopted from our experiments. Additionally, the carbonation time was 15 minutes to be consistent with experiments. The CO_2 capture step is assumed to take 1.5 hours, with one batch of solution cycling in and out of the air contactor system every 15 minutes. The air was assumed to enter the system at 25°C. Due to the large mass of air entering the system, the exothermic heat release from the reaction of $NaOH/Na_2SiO_3$ solution with CO_2 can be removed with a negligible air temperature rise. Additionally, during the carbonation step, the increase in temperature of the solution from exothermic reaction heat is negligible due to the large water to slag ratio. This allows the heat exchanger and cooling tower for the carbonation and air contactor system to be removed. The heat balance is shown in Fig. 4a.

Due to the large amount of electricity needed for the secondary processes (conveying, milling, filtering, stirring, pumping, and fan-driven gas flow), the steel slag input temperature needed to be increased for an internal steam power plant to supply electricity. Therefore, steel slag enters the system at 1067°C which supplies enough power to heat water to 1000°C where H_2 production occurs. After the steam power plant, the steel slag leaves at 176°C which still supplies enough heat to dry the H_2SiO_3.

The process cost was estimated in the same manner as the previous cases. For the air CO_2 looping a CO_2 conversion of 35% was determined from the CO_2 capture step of quenched slag at 24°C in our experiment (Fig. 2c). For the air contactor design, an air density of 1.24 kg/m^3 was assumed, along with a CO_2 mass fraction of 0.06% corresponding to an atmospheric concentration of 400 ppm. The FOB cost, installed costs, and total cost over 50 years for the equipment are detailed in Table S13 for electricity supply methods 1-3. The updated steam power plant components are listed in Table S14.

Revenues from H_2, carbonate-rich product, precipitated silica, and the 45Q tax credit were accounted for. The selling price of the air CO_2 looping carbonate-rich product was assumed to be \$26/ton.[53] The 45Q tax credit provides \$180/ton of CO_2 captured for projects that convert air CO_2 into useful products.[55]

The breakeven time was calculated using Eqn S18 for the scenario where electricity is provided from an external solar grid and is shown:

$$CAPEX = Equipment + Land + Water + Na_2CO_3 = \$202{,}246{,}954 + \$782{,}764 + \$53{,}290 + \$325{,}554 = \$203{,}408{,}562$$

$$Revenue = H_2 + Carbonated\ Slag + Silica + 45Q\ Tax\ Credit = \left(\frac{\$2}{kg_{H_2}}\right)\left(4{,}183{,}915\frac{kg_{H_2}}{year}\right) + \left(\frac{\$26}{ton_{carbona}}\right.$$

$$+ \left(\frac{\$600}{ton_{silica}}\right)\left(37{,}599\frac{ton_{silica}}{year}\right) + \left(\frac{\$180}{ton_{CO_2}}\right)\left(100{,}000\frac{ton_{CO_2}}{year}\right) = \$122{,}590{,}794$$

$$OPEX = Maintenance + Labor + Water + Na_2CO_3 + Electricity = \$3{,}898{,}964 + \$674{,}520 + \$81{,}5$$
$$= \$14{,}784{,}426$$

$$Breakeven\ Time = \frac{\$203{,}408{,}562}{\frac{\$122{,}590{,}794}{year} - \frac{\$14{,}784{,}426}{year}} = 1.9\ years$$

TEA – Air CO_2 Baseline

The process chemistry and thermodynamics for the air CO_2 baseline case is the same as the point-source baseline. The molar content of active phases was quantified in the same way as the point-source baseline, but with 0.23 mmol CO_2 mineralized per gram of slag. The CO_2 mineralized was determined for the baseline carbonation of quenched slag at 24°C in our experiment (Fig. 2c). The molar quantities of each reacted phase in the slag are shown in Table S4.

The mass balance for the air CO_2 baseline is shown in Table S5 and Fig. S7b.

Similarly to the air CO_2 looping case, the CO_2 capture step takes place at 25°C due to the large amount of air entering the system and cooling the solution. The heat balance is shown in Fig. S7b.

Steel slag was assumed to enter the system at the same temperature as the air CO_2 looping scenario. Therefore, steel slag enters the system at 1067°C which supplies enough power to heat water to 1000°C where H_2 production occurs. Due to the high electricity demand, waste heat from the slag was insufficient to meet the process requirements. An additional 18.5 MW of grid electricity was supplied, resulting in the steel slag exiting the power cycle at 196°C.

The process cost was estimated in the same manner as the previous cases. For the air CO_2 baseline a CO_2 conversion of 9% was determined from the CO_2 capture step of baseline quenched slag at 24°C in our experiment (Fig. 2c). The FOB cost, installed costs, and total cost over 50 years for the equipment are detailed in Table S15 for electricity cases 1-3. The updated steam power plant components are listed in Table S16.

Revenues from H_2, carbonate-rich product, and the 45Q tax credit were accounted for. The selling price of the air CO_2 baseline carbonate-rich product was assumed to be \$7.24/ton, which was scaled to have equal yearly revenue for carbonate-rich product as the air CO_2 looping case, considering the mixed effects of larger product quantity and lower product carbonation extent of baseline product than looping. The 45Q tax credit provides \$180/ton of CO_2 captured for projects that convert air CO_2 into useful products.

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