Supplementary Text:

Absorber column size

Michaelis-Menten rate law can be applied to the CO_2 hydration reaction. By making a quasi steady state approximation for the concentration of enzyme-substrate complex, assuming that the enzyme is not in great excess over substrate, and assuming that substrate is never depleted (since new substrate is constantly added), the following equation can be derived:

$$v_{\mathrm{catalyzed}} = \frac{v_{\max}[S]}{(K_m + [S])} \qquad (S1)$$

where $v_{\max} = (k_{\mathrm{cat}})(\text{moles enzyme})$ is the maximum reaction velocity, $K_m = 8$ mM, $k_{cat} = 6\text{x}10^5$ second^{-1}, and [S] = solubility of CO_2 at 40°C = 22 mM.[31, 32] The desired $v_{catalyzed}$ = the CO_2 flux = 5.09 moles CO_2 second^{-1}. A rearrangement of equation (1) shows the moles of enzyme needed to catalyze the reaction in the absorber.

$$\text{Moles enzyme} = \frac{(v_{\mathrm{catalyzed}})(K_m + [S])}{[S](k_{\mathrm{cat}})} \qquad (S2)$$

Solving this expression for reactor conditions gives a value of $1.16\text{x}10^{-5}$ moles of enzyme. To determine the volume of the scaled up reactor for the commercial process, the concentration of enzyme in the reactor must be calculated. It was assumed that it was best to work at a high concentration of bCA2-yeast, to minimize the reactor volume and the associated capital equipment cost. A concentration of cells $1.1\text{x}10^8$ cells mL^{-1} was chosen because above this concentration the mixture becomes to thick to handle.

Assuming 100,000 enzymes per cell (which was confirmed by flow cytometry), the final concentration of enzyme will be around 27 nM.

The volume of the reactor then can be calculated:

$$\text{Volume}_{\mathrm{reactor}} = \frac{(\text{moles}_{\mathrm{enzyme}})}{[\text{enzyme}]} \qquad (S3)$$

which leads to a catalyzed reaction liquid volume of 420 L. This liquid volume, plus a liquid hold-up of 5% gives an active absorber volume of 8400 L, which becomes 11300 L including the top and bottom dead zones (see table 3).
To determine the impact that the enzyme has on the reactor volume it is also possible to calculate the volume necessary for the uncatalyzed reactor. To do this, the following equation is used:

$$v_{\mathrm{uncatalyzed}} = k_1[S] \qquad (S4)$$

Using the uncatalyzed reaction rate parameter value of 0.045 sec^{-1} for k_1,[33] and a value for [S] that is equal to the solubility of CO_2 at 40°C = 0.022 moles L^{-1},[31] gives an uncatalyzed reaction rate of 0.001 moles L^{-1} sec^{-1}. Dividing the CO_2 flux by the uncatalyzed reaction rate gives a required liquid volume of 5090 L, more than 10 times the volume required if using the carbonic anhydrase yeast. The associated equipment cost increase for this process modification is reported in supplementary table 4.

Mineralization reactor size

The calculations to determine the size of the mineralization reactor are based on the mineralization rate of $CaCO_3$ and the amount of CO_2 to be mineralized. The volume of the reactor can be calculated using equation 5.

$$\text{Volume}_{\mathrm{reactor}} = \frac{CO_2\ \text{capture rate}}{\text{rate}_{\mathrm{mineralization}}} \qquad (S5)$$

where CO_2 capture rate = (CO_2 flux)(CO_2 capture efficiency of CaO) = 4.33 moles second^{-1} and $\text{rate}_{\mathrm{mineralization}}$ is the baseline rate measured in the laboratory experiments for yeast with no mineralization peptide = $6.3\text{x}10^{-4}$ moles $CaCO_3$ L^{-1} second^{-1}. Substituting these values into equation 5 gives an estimated mineralization reaction volume of roughly 6900 L. If the mineralization rate could be increased by 10x, then the mineralization reactor could be eliminated and the reaction could occur in the bottom of the absorption reactor.

It is also worth considering what the mineralization rate would be in the absence of the mineralization template provided by the yeast cells. To do this, the uncatalyzed mineralization rate from the mineralization rate measurement experiments was used = $5.2\text{x}10^{-4}$ moles $CaCO_3$ L^{-1} second^{-1} (see figure 2). Substituting this value into equation 5 gives an estimated reaction volume of roughly 8300 L or 20% larger than the catalyzed reactor volume. The associated equipment cost increase for this process modification is reported in supplementary table 4.

Settling reactor size

The presence of yeast cells in the mineralization reactor results in much larger particles than when the CO_2 is mineralized without the yeast cells (see supplementary figure 3). Dozens of yeast cells interconnect with dozens of calcium carbonate crystals whereas the carbonate minerals formed in the absence of yeast cells stay as individual crystals. The average particle sizes for the two conditions were estimated to be 1 mm for the yeast-CaCO3 particles and 25 μm for the individual $CaCO_3$ particles.

The settling time was measured in the lab-scale mineralization reactor by turning off the stirring and monitoring the amount of time that it took for the materials to settle to the bottom of the reactor. The engineered yeast in supplementary figure 3 had a settling time of roughly 14 cm min^{-1}, whereas the no yeast sample took more than 4 hours to fully settle (settling time < 0.029 cm min^{-1}), which leads to a settling rate enhancement of almost five hundred fold. Even if the inactive materials in the fly ash (which are typically less than 100 μm in diameter) are able to increase the settling rate of the $CaCO_3$ in the settling reactor by a factor of 10 (to 0.3 cm min^{-1}), the difference is still almost fifty fold. This slower settling rate would have a significant impact on the residence time in the settling reactor, and, thus, the cost of this piece of equipment. The associated equipment cost increase for this process modification is reported in supplementary table 4.

Production of bCA2-yeast

In order to determine the amount of bCA2-yeast required for the process, the following equation is used:

$$\text{yeast flux} = (\text{Volumetric rate}_{\text{absorber}})\left(\frac{\text{yeast cells}}{\text{L}}\right)(\text{MW}_{\text{cells}}) \approx 7.5\ \frac{\text{kg}_{\text{DCW}}}{\text{hour}} \qquad \text{(S6)}$$

where the volumetric rate in the absorber is 6.98×10^4 L $hour^{-1}$, the concentration of cells in the absorber is 1.1×10^8 cells mL^{-1}, and the molecular weight of the cells is 1×10^{-12} g_{DCW} $cell^{-1}$. The bCA2-yeast are grown in fermenters with an assumed yield of 50% dry cell weight yeast per gram of sugar (glucose) and with an assumed growth rate of 0.05 kg DCW L^{-1} day^{-1}. The calculated yeast fermenter volume is thus 4.5 m^3.

There are several assumptions underlying the calculations in this section, which are worth a brief discussion. First, the yeast-display system implemented in this work uses plasmid (i.e., non-genomic) DNA to host the Aga2 and carbonic anhydrase genes, while the Aga1 gene is chromosomally integrated. The plasmid is retained in successive generations of cells with auxotrophic pressure. In practice, this means that minimal media must be used to retain the plasmid. Even with this selection pressure, however, only 40%-60% of the cells in a population retain the plasmid, presumably because the missing nutrient is secreted into the media at a sufficient concentration by the cells with the plasmid so as to support the cells without it. In the implementation proposed for the final process, however, the Aga2-bCA2 gene would be genomically integrated, thus avoiding the problems associated with the use of the plasmid DNA.[32]

Second, expression of the Aga1 and Aga2-CA2 genes is under the control of a galactose inducible expression cassette. Since transcription from this cassette cannot occur in the presence of glucose, glucose would need to be exhausted and then galactose must be used as the sole carbon source during the induction period. This would require more than 100 kg day^{-1} of galactose, which would be cost prohibitive.[33] In the implementation proposed for the final process, however, the Aga2-bCA2 gene would be placed under the control of a copper inducible expression promoter, like Cup1, which would allow for the use of glucose as the carbon source.[34] Using 100 μM copper sulphate to induce expression would require roughly 40 grams day^{-1} copper sulphate pentahydrate (Sigma Aldrich #209198), or roughly \$1 day^{-1} at a price of \$31 kg^{-1}.

High temperature stable CA2 process modification

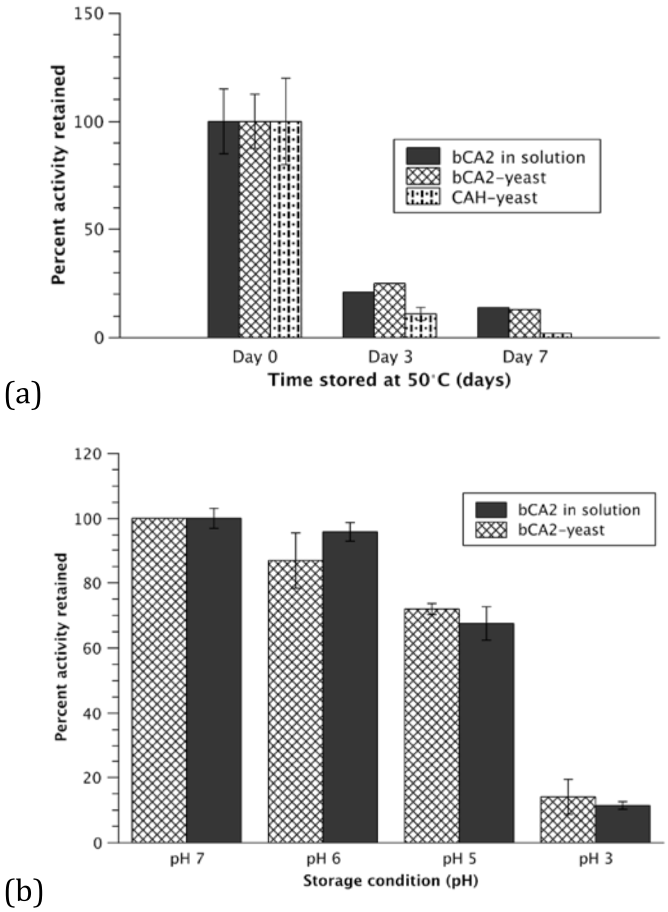
The CO_2 mineralization process was modified using a recombinantly produced CA2 that is stable at temperatures of up to 60°C for 2 hours and that can be purchased at a price of \$10 / kg. For this modification (see process 6 in table 5) we assumed an equivalent CO_2 hydration rate for the high-temperature CA2 as our bCA2-yeast has. The process was modified by removing the second heat exchanger, E2, and the well cooling water and by reducing the size of the first heat exchanger, E1. Without the yeast in the system, the mineralization rate is slower and the settling rate is slower which means that a larger mineralization reactor, S4, and settling reactor, M2, are required. The elimination of E2, the well cooling water, all the yeast growth equipment (S6, M6, S0, and P0) and the yeast growth raw materials does not offset the costs incurred with a larger S4 and M2 and the purchase price of recombinant CA2 (at \$10 / kg). Our model predicts that the bCA2-yeast process is ~2% less expensive per tonne of CO_2 mineralized (~$6 / tonne).

High temperature stable CA2 and wild-type yeast process modification

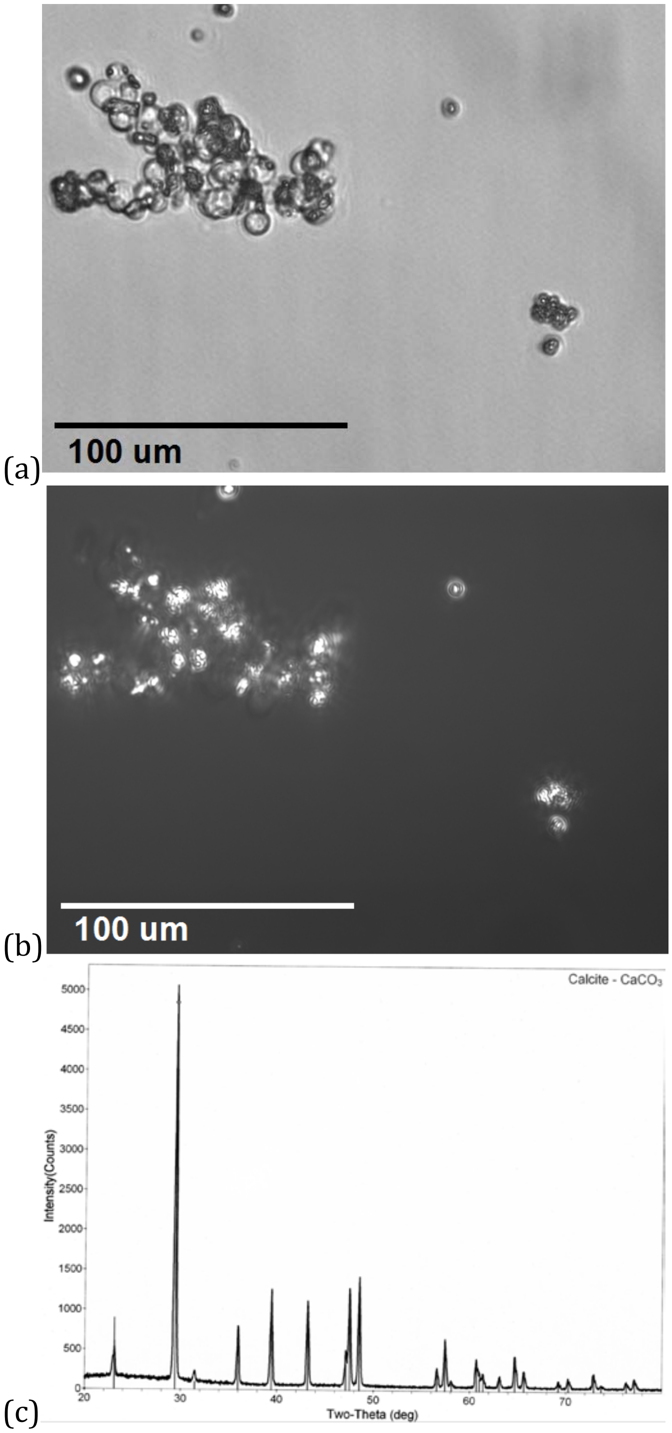
To further benchmark the bCA2-yeast and fly ash process, we added a case in which we purchased high temperature CA2 to enhance the CO_2 hydration rate and wild-type yeast to enhance the mineralization rate and the settling rate. This allowed us to eliminate E2, S6, M6, S0, P0, and the well cooling water, to use a smaller E1, and to not increase the sizes of S4 or M2. This effectively gave us a two-component (CA2 and yeast) system with high-temperature stability, and no need to grow the biological components ourselves. Even with these generous assumptions, our model predicted that our proposed bCA2-yeast process would be able to mineralize CO_2 at a lower cost than this two-component system (see table 5).

Supplementary Figures:

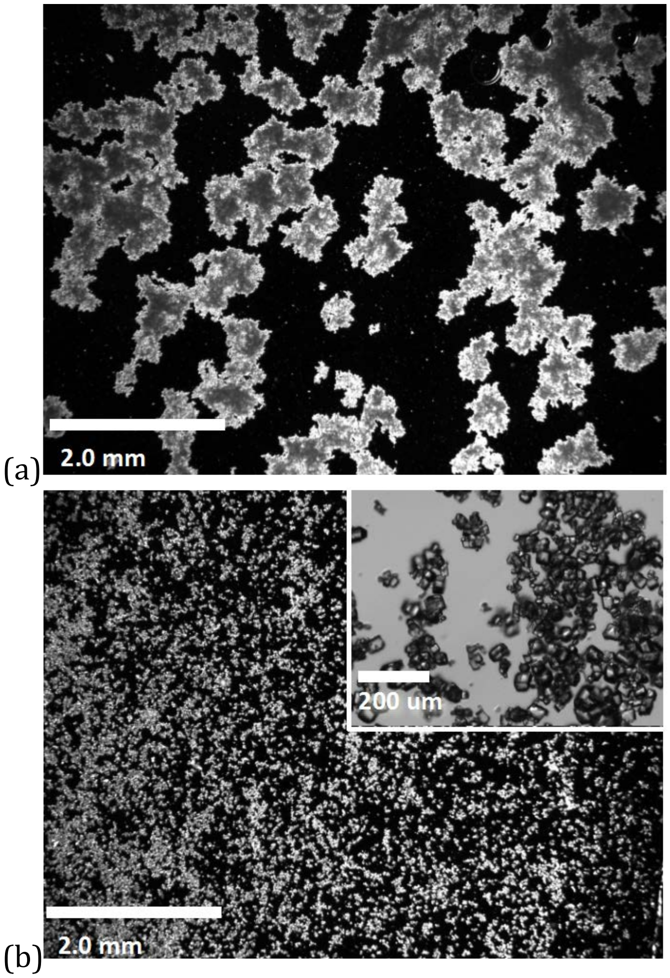
SUPPLEMENTARY FIGURES:



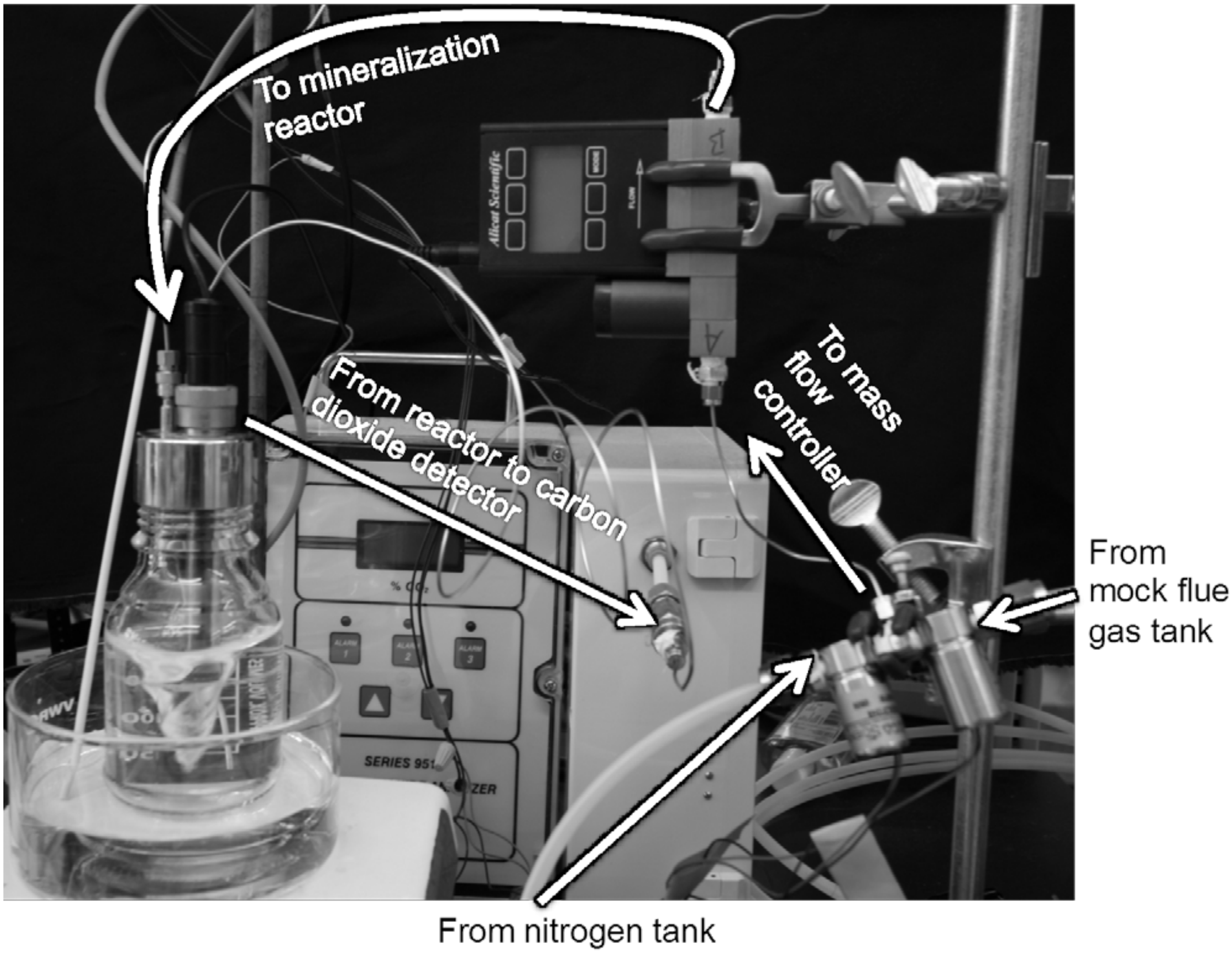
Supplementary Figure 1: Temperature and pH stability of yeast displayed carbonic anhydrase isoforms. (a) Day 0 samples are freshly prepared enzyme solutions. The samples were stored at 50°C for 3 days and 7 days and activity was measured for each sample using the assay described in the experimental section. The results for each sample were normalized to the starting activity levels so that the stability of each sample could be compared to that of the other samples. (b) pH stability of bCA2-yeast and bCA2 in solution. Samples were stored for 16 hours at 4°C from pH 7.0 to pH 3.0. Samples were then transferred to the CA2 assay buffer, activity was measured and compared to the starting activity.



Supplementary Figure 2: Cross-polarized microscopy and X-ray diffraction confirmation of $CaCO_3$ mineralization (a) Representative image of the N66-yeast from the mineralization reactor. The dark objects that are attached to the yeast cells are $CaCO_3$. (b) Cross-polarized microscopy image of the same sample from (a). In this image, only objects that can change the polarization of light (i.e., $CaCO_3$ crystals) are illuminated (c) XRD chromatogram confirms that calcite is mineralized in the presence of N66-yeast. The N66-yeast sample peaks are shown in black and the calcite reference peaks are shown in grey.



Supplementary Figure 3: Size of calcium carbonate particles mineralized in the presence and absence of engineered yeast cells (a) This image shows the typical particle sizes of the yeast-$CaCO_3$ particles when CO_2 is mineralized in the presence of GPA-yeast cells with a final concentration of $6x10^{11}$ peptides mL^{-1} ($2x10^7$ cells mL^{-1}). (b) This image shows the typical particle sizes of the $CaCO_3$ particles when CO_2 is mineralized without yeast cells. Both reactions took place in the CO_2 capture reactor described in the main text with a starting concentration of 0.1 M Tris-base and 0.1 M $CaCl_2$ and with a flow rate of 0.1 L $minute^{-1}$ of 15% CO_2.



Supplementary Figure 4: Photograph of bench-scale CO_2 capture reactor. Solenoid valves were used to switch between a nitrogen gas purge and a mock flue gas. An Alicat Scientific mass flow controller calibrated to the mock flue gas was used to control the flow of gas into the reactor. The gas was directed to the bottom of the stirred liquid in the reactor and the off gas was collected from the head space in the reactor and sent to an in-line CO_2 detector. A Ca^{2+} sensitive probe was placed in the reactor liquid to monitor the Ca^{2+} concentration in real time.

Sample	**Concentration (nM)**	**Mineralization rate (M / sec)**
No biomolecules	0	$1.07x10^{-5} \pm 4.1x10^{-7}$
bCA2	67	$1.41x10^{-5} \pm 5.1x10^{-7}$
bCA2-yeast	67*	$1.35x10^{-5}$

*Concentration reported in terms of active bCA2 molecules

Supplementary Table 1: Conversion of gaseous CO_2 to $CaCO_3$ in a lab-scale CO_2 capture reactor demonstrates that the bCA2-yeast and the soluble bCA2 enhance the conversion of CO_2 to $CaCO_3$ in the reactor. These data show the results of a process that is the combination of CO_2 dissolution, CO_2 hydration, and $CaCO_3$ mineralization in the lab-scale CO_2 capture reactor. These results are specific to this reactor and cannot be directly compared to results from other reactors.

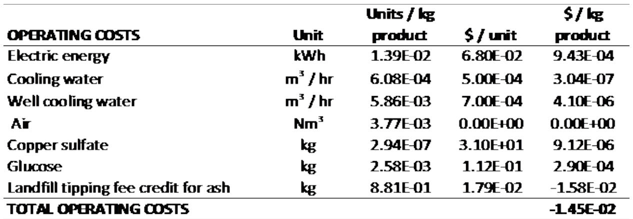
ISBL investment cost

Direct Cost	**Man power** (thousand $)	**Material** (thousand $)	**Total** (thousand $)
Equipment	19.6	3412.2	3431.8
Piping	201.3	395.0	596.3
Civil	119.6	106.1	225.7
Steel	26.0	99.5	125.5
Instrumentation	85.2	455.3	540.5
Electrical	107.0	354.5	461.5
Insulation	8.9	11.0	19.9
Paint	68.9	38.6	107.5
Total	**636.5**	**4872.0**	**5508.5**

Indirect Cost	**Total** (thousand $)
Engineering (15% of total direct cost)	826.3
Contingency (15% of total direct cost)	826.3
G & A Overheads (3.3% of total direct cost)	181.8
Contract Fee (3.5% of total direct cost)	192.8
Total	**2027.1**

Total Investment (ISBL)	**7535.6**

Supplementary Table 2: Inside battery limits (ISBL) costs for the CO_2 mineralization process using bCA2-yeast and fly ash. The itemized list for the total material cost is in table 4.

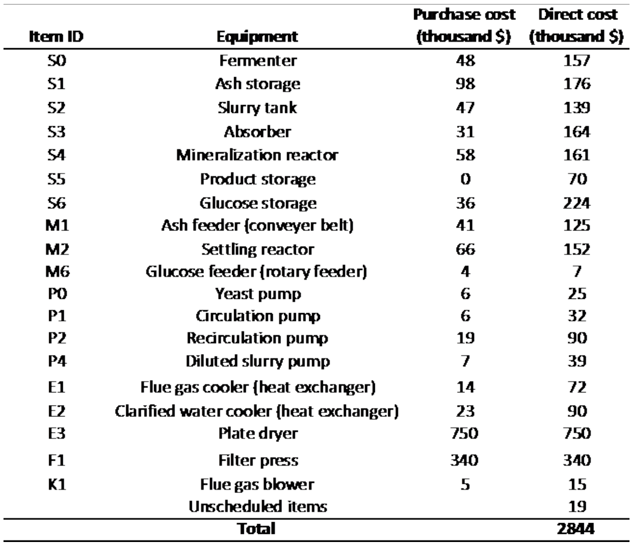


OPERATING COSTS	Unit	Units / kg product	$ / unit	$ / kg product
Electric energy	kWh	1.39E-02	6.80E-02	9.43E-04
Cooling water	m^3 / hr	6.08E-04	5.00E-04	3.04E-07
Well cooling water	m^3 / hr	5.86E-03	7.00E-04	4.10E-06
Air	Nm^3	3.77E-03	0.00E+00	0.00E+00
Copper sulfate	kg	2.94E-07	3.10E+01	9.12E-06
Glucose	kg	2.58E-03	1.12E-01	2.90E-04
Landfill tipping fee credit for ash	kg	8.81E-01	1.79E-02	-1.58E-02
TOTAL OPERATING COSTS				-1.45E-02

Supplementary Table 3: Operating costs for CO_2 mineralization process using fly ash and bCA2-yeast. The contribution of each of these utilities or raw materials to the price per kg of product generated is based on the amount of each that is required to make one kg of product and on the respective cost per unit.

Item ID	Equipment	Processes that use this equipment	Fold increase volumetrically without rate enhancment provided by biological components	Purchase cost (thousand $)	Direct cost (thousand $)
S3	Absorber	1, 2, 3, 6, 7		31	164
S3-L	Large absorber	4, 5	10x	82	290
S4	Mineralization reactor	1, 3, 5, 7		58	161
S4-L	Large mineralization reactor	4, 6	1.2x	65	169
M2	Settling reactor	1, 2, 3, 5, 7		86	276
M2-L	Large settling reactor	4, 6	50x	546	1103

Supplementary Table 4: Equipment costs for modifications to the CO_2 capture processes using fly ash (see table 3, table 5, and figure 3 for full equipment lists, process flow diagrams, and cost analyses for these processes). The bCA2-yeast enhances CO_2 hydration rate, $CaCO_3$ mineralization rate, and the settling rate of the product. Without bCA2-yeast, the reactors for these steps would be larger and the reactor costs would be higher. This table shows which processes would require the more expensive reactors.



Item ID	Equipment	Purchase cost (thousand $)	Direct cost (thousand $)
S0	Fermenter	48	157
S1	Ash storage	98	176
S2	Slurry tank	47	139
S3	Absorber	31	164
S4	Mineralization reactor	58	161
S5	Product storage	0	70
S6	Glucose storage	36	224
M1	Ash feeder (conveyer belt)	41	125
M2	Settling reactor	66	152
M6	Glucose feeder (rotary feeder)	4	7
P0	Yeast pump	6	25
P1	Circulation pump	6	32
P2	Recirculation pump	19	90
P4	Diluted slurry pump	7	39
E1	Flue gas cooler (heat exchanger)	14	72
E2	Clarified water cooler (heat exchanger)	23	90
E3	Plate dryer	750	750
F1	Filter press	340	340
K1	Flue gas blower	5	15
	Unscheduled items		19
	Total		**2844**

Supplementary Table 5: Process equipment for CO_2 mineralization process using paper mill ash and bCA2-yeast. Equipment costs were determined using Aspen Process Economic Analyzer V7.2.1 where applicable. For equipment not available in Aspen Process Economic Analyzer, estimated costs were provided from equipment vendors. Costs are in US dollars for a US location in 2011. The equipment in this list corresponds with the equipment in figure 3.

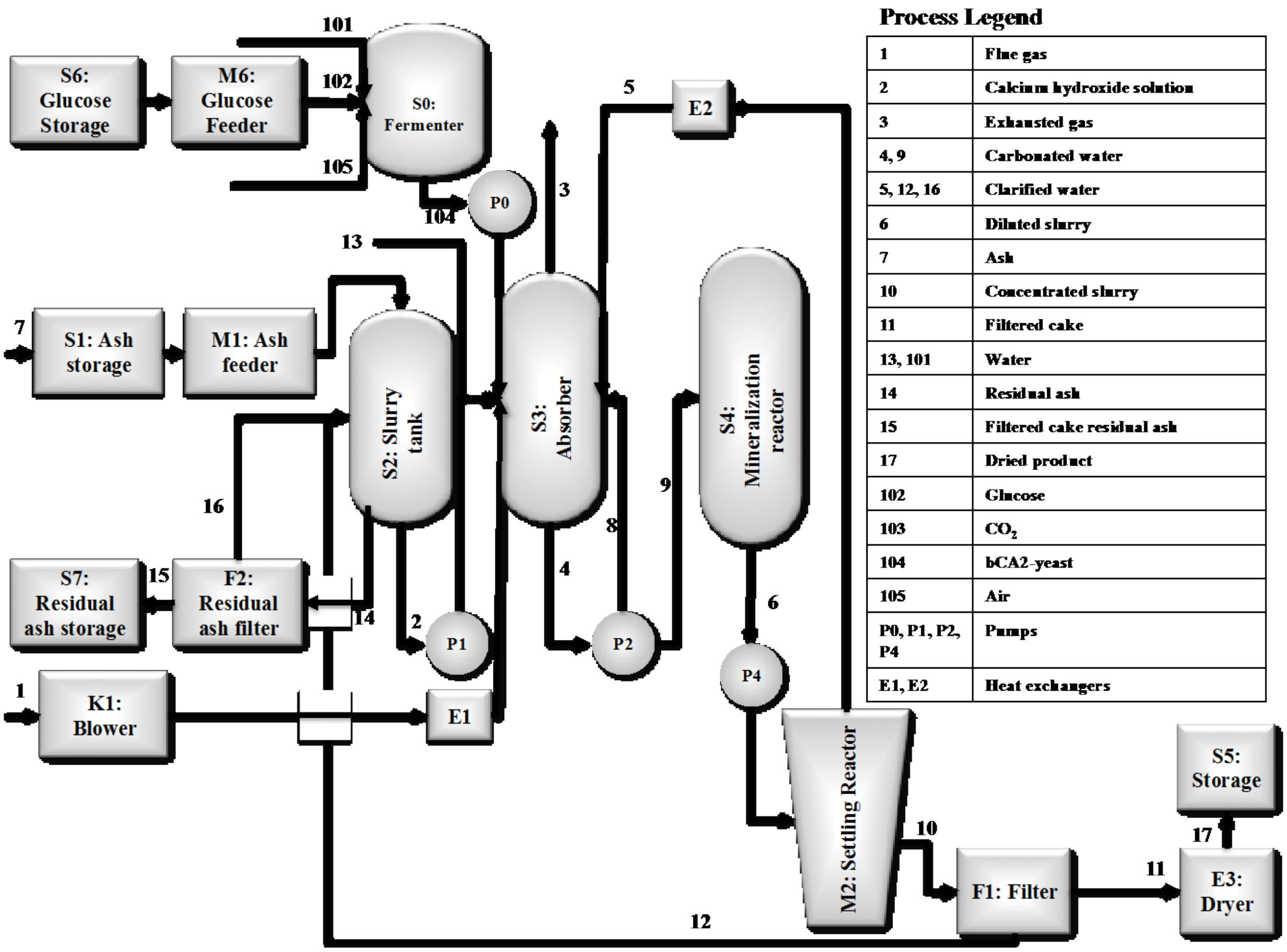
	1 Flue gas		2 Ash slurry		3 Exhausted gas		4 Carbonated water		5 Clarified water	
	Rate [tonne/h]	Comp %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2	2.8	70.2			2.8	85.0				
O_2	0.15	3.7			0.15	4.5				
CO_2	0.81	20.5			0.12	3.7				
H_2O	0.22	5.6	7.8	84.7	0.22	6.8	58.8	92.5	21.2	100.0
Mg^{2+}			0.028	0.31						
$CaSO_4$			0.056	0.61			0.19	0.30		
SiO_2			0.28	3.06			0.085	0.13		
Al_2O_3			0.14	1.53			0.96	1.50		
Fe_2O_3			0.028	0.31			0.44	0.69		
CaO			0.87	9.49						
$CaCO_3$							3.0	4.7		
$MgCO_3$							0.12	0.19		
Glucose										
bCA2-yeast							6.5E-03	1.0E-02		
tonne/hr	3.9	100.0	9.2	100.0	3.3	100.0	63.6	100.0	21.2	100.0

	6 Diluted slurry		7 Ash		8 Recycled water		9 Carbonated water		10 Concentrated slurry	
	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2										
O_2										
CO_2										
H_2O	29.4	92.5			29.4	92.5	29.4	92.5	8.2	77.5
Mg^{2+}			0.028	2.0						
$CaSO_4$	0.10	0.30	0.056	4.0	0.10	0.30	0.10	0.30	0.10	0.90
SiO_2	0.042	0.13	0.28	20.0	0.042	0.13	0.042	0.13	0.042	0.40
Al_2O_3	0.48	1.5	0.14	10.0	0.48	1.50	0.48	1.50	0.48	4.5
Fe_2O_3	0.22	0.7	0.028	2.0	0.22	0.69	0.22	0.69	0.22	2.1
CaO			0.87	62.0						
$CaCO_3$	1.5	4.7			1.5	4.7	1.5	4.7	1.5	14.0
$MgCO_3$	0.059	0.19			0.059	0.19	0.059	0.19	0.059	0.56
Glucose										
bCA2-yeast	3.2E-03	0.010			3.2E-03	0.010	3.2E-03	0.010	3.2E-03	0.031
tonne/hr	31.8	100.0	1.4	100.0	31.8	100.0	31.8	100.0	10.6	100.0

	11 Filtered cake		12 Clarified water		13 Water		14 Dried Product	
	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2								
O_2								
CO_2								
H_2O	0.42	15.0	7.8	100.0	0.42	100.0	0.012	0.50
Mg^{2+}								
$CaSO_4$	0.10	3.4					0.10	4.0
SiO_2	0.042	1.5					0.042	1.8
Al_2O_3	0.48	17.0					0.48	19.9
Fe_2O_3	0.22	7.9					0.22	9.2
CaO								
$CaCO_3$	1.49	53.0					1.5	62.0
$MgCO_3$	0.059	2.1					0.059	2.5
Glucose								
bCA2-yeast	3.2E-03	0.12					3.2E-03	0.13
tonne/hr	2.8	100.0	7.8	100.0	0.4	100.0	2.4	100.0

	101 Water		102 Glucose		103 CO_2		104 bCA2-yeast		105 Air	
	Rate [tonne/h]	Comp %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2									9.2	76.7
O_2	3.5E-03	11.3							2.8	23.3
CO_2					4.7E-03	100.0				
H_2O	2.7E-02	88.7					0.029	90.0		
Mg^{2+}										
$CaSO_4$										
SiO_2										
Al_2O_3										
Fe_2O_3										
CaO										
$CaCO_3$										
$MgCO_3$										
Glucose			6.47E-03	100.0						
bCA2-yeast							3.2E-03	10.0		
tonne/hr	3.06E-02	100.0	6.47E-03	100.0	4.75E-03	100.0	0.0	100.0	12.0	100.0

Supplementary Table 6: Mass balance for CO_2 mineralization process using paper mill ash and bCA2-yeast. The streams in this table correspond to those with matching labels in figure 3.



Process Legend

1	Flue gas
2	Calcium hydroxide solution
3	Exhausted gas
4, 9	Carbonated water
5, 12, 16	Clarified water
6	Diluted slurry
7	Ash
10	Concentrated slurry
11	Filtered cake
13, 101	Water
14	Residual ash
15	Filtered cake residual ash
17	Dried product
102	Glucose
103	CO_2
104	bCA2-yeast
105	Air
P0, P1, P2, P4	Pumps
E1, E2	Heat exchangers

Supplementary Figure 5: Process diagram for CO_2 mineralization process using CaO-leached fly ash and bCA2-yeast. Equipment sizes and costs are listed in supplementary table 7. Mass balances for the fly ash and bCA2-yeast process are listed in supplementary table 3. Mass balances are listed in supplementary table 8. The itemized equipment list is in supplementary table 7.

Item ID	Equipment	Purchase cost (thousand $)	Direct cost (thousand $)
S0	Fermenter	48	157
S1	Ash storage	188	275
S2	Slurry tank	210	309
S3	Absorber	96	294
S4	Mineralization reactor	208	397
S5	Product storage	0	53
S6	Glucose storage	36	224
S7	storage-offside	0	72
M1	Ash feeder (conveyer belt)	45	128
M2	Settling reactor	106	367
M6	Glucose feeder (rotary feeder)	4	7
P0	Yeast pump	6	25
P1	Circulation pump	28	141
P2	Recirculation pump	47	218
P4	Diluted slurry pump	28	141
E1	Flue gas cooler (heat exchanger)	14	72
E2	Clarified water cooler (heat exchanger)	16	73
E3	Plate dryer	500	500
F1	Filter press	960	960
K1	Flue gas blower	5	15
	Unscheduled items		41
	Total		**4468**

Supplementary Table 7: Process equipment for CO_2 mineralization process using CaO-leached fly ash and bCA2-yeast. Equipment costs were determined using Aspen Process Economic Analyzer V7.2.1 where applicable. For equipment not available in Aspen Process Economic Analyzer, estimated costs were provided from equipment vendors. Costs are in US dollars for a US location in 2011. The equipment in this list corresponds with the equipment in supplementary figure 5.

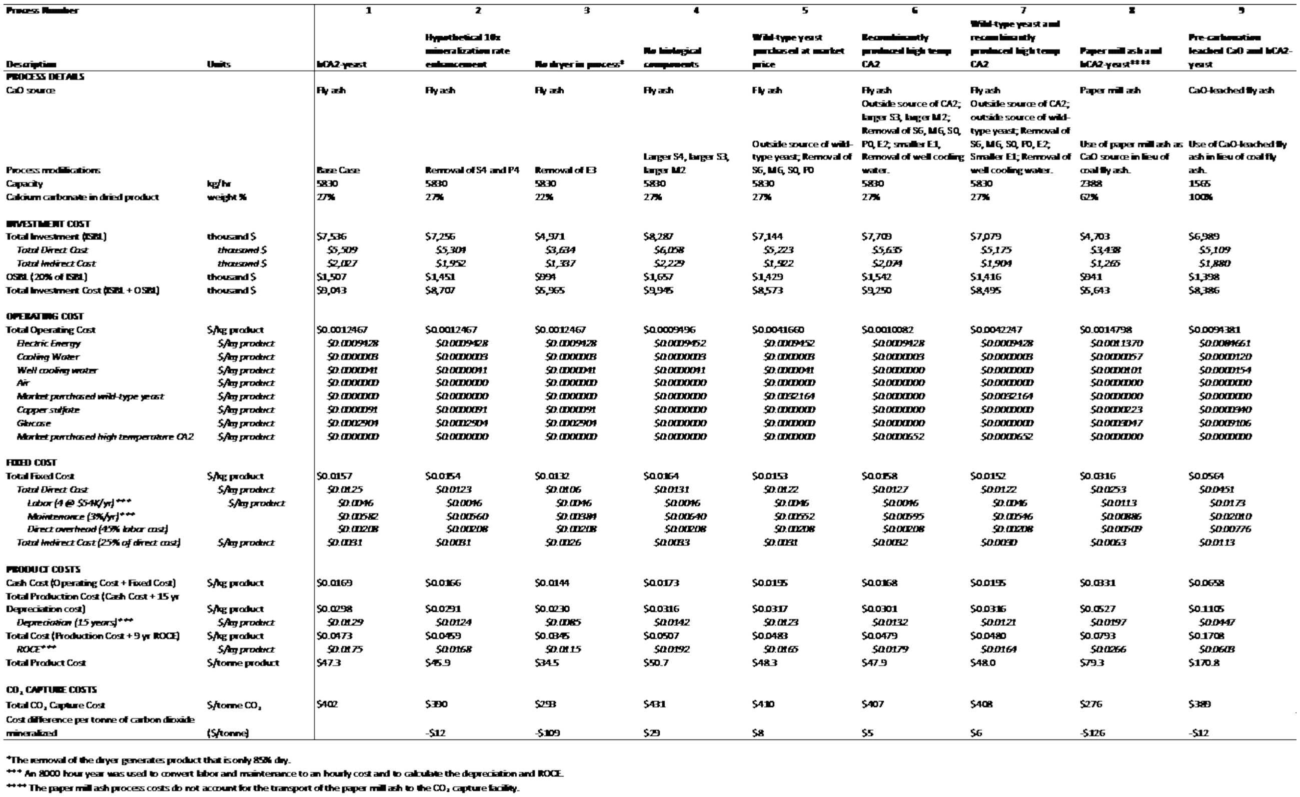
	1 Flue gas		2 Calcium hydroxide solution		3 Exhausted gas		4 Carbonated water		5 Clarified water	
	Rate [tonne/h]	Comp %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2	2.8	70.2			2.8	85.0				
O_2	0.1	3.7			0.15	4.5				
CO_2	0.8	20.5			0.12	3.7				
H_2O	0.2	5.6	576.5	99.8	0.22	6.8	576.5	99.7	565.0	100.0
Ca^{2+}			0.63	0.11						
OH^-			0.53	0.09						
SiO_2										
Al_2O_3										
Fe_2O_3										
CaO										
$CaCO_3$							1.6	0.27		
Glucose										
Yeast1							6.3E-03	1.1E-03		
tonne/hr	3.9	100.0	577.6	100.0	3.3	100.0	578.0	100.0	565.0	100.0

	6 Diluted slurry		7 Ash		9 Carbonated		10 Concentrated slurry		11 Filtered cake	
	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2										
O_2										
CO_2										
H_2O	576.5	99.7			576.5	99.7	11.5	88.0	0.28	14.9
Ca^{2+}										
OH^-										
SiO_2			2.6	50.0						
Al_2O_3			1.3	25.0						
Fe_2O_3			0.41	8.0						
CaO			0.87	17.0						
$CaCO_3$	1.6	0.27			1.6	0.3	1.6	11.9	1.6	84.7
Glucose										
Yeast1	6.3E-03	1.1E-03			6.3E-03	1.1E-03	6.3E-03	4.8E-02	6.3E-03	0.34
tonne/hr	578.0	100.0	5.1	100.0	578.0	100.0	13.1	100.0	1.8	100.0

	12 Clarified water		13 Water		14 Residual Ash		15 Filtered cake residual ash		16 Clarified water		17 Dried Product	
	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2												
O_2												
CO_2												
H_2O	11.2	100.0	0.01	0.50	9.9	70.0	0.75	15.0	9.2	100.0	7.9E-03	0.50
Ca^{2+}												
OH^-												
SiO_2					2.6	18.1	2.6	51.2				
Al_2O_3					1.3	9.0	1.3	25.6				
Fe_2O_3					0.41	2.9	0.41	8.2				
CaO												
$CaCO_3$			1.6	99.1							1.6	99.1
Glucose				0.00								
Yeast1			6.3E-03	0.40							6.3E-03	0.40
tonne/hr	11.2	100.0	1.6	100.0	14.2	100.0	5.0	100.0	9.2	100.0	1.6	100.0

	101 Water		102 Glucose		103 CO_2		104 bCA2-yeast		105 Air	
	Rate [tonne/h]	Comp %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt	Rate [tonne/h]	Comp. %wt
N_2	2.8	70.2			2.77	85.03				
O_2	0.1	3.7			0.15	4.48				
CO_2	0.8	20.5			0.12	3.72				
H_2O	0.2	5.6	576.45	99.80	0.22	6.77	576.45	99.73	576.45	99.73
Ca^{2+}			0.63	0.11						
OH^-			0.53	0.09						
SiO_2										
Al_2O_3										
Fe_2O_3										
CaO										
$CaCO_3$							1.56	0.27	1.56	0.27
Glucose										
Yeast1							6.3E-03	1.1E-03	6.3E-03	1.1E-03
tonne/hr	3.9	100.0	577.6	100.0	3.3	100.0	578.0	100.0	578.0	100.0

Supplementary Table 8: Mass balance for CO_2 mineralization process using CaO-leached fly ash and bCA2-yeast. The streams in this table correspond to those with matching labels in supplementary figure 5.



Process Number		**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**
Description	**Units**	**hCA2-yeast**	**Hypothetical 10x mineralization rate enhancement**	**No dryer in process***	**No biological components**	**Wild-type yeast purchased at market price**	**Recombinantly produced high temp CA2**	**Wild-type yeast and recombinantly produced high temp CA2**	**Paper mill ash and hCA2-yeast******	**Pre-carbonation leached CaO and hCA2-yeast**
PROCESS DETAILS										
CaO source		Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Fly ash	Paper mill ash	CaO-leached fly ash
Process modifications		Base Case	Removal of S4 and P4	Removal of E3	Larger S4, larger S3, larger M2	Outside source of wild-type yeast; Removal of S6, M6, S0, P0	Outside source of CA2; larger S3, larger M2; Removal of S6, M6, S0, P0, E2; smaller E1, Removal of well cooling water.	Outside source of CA2; outside source of wild-type yeast; Removal of S6, M6, S0, P0, E2; Smaller E1; Removal of well cooling water.	Use of paper mill ash as CaO source in lieu of coal fly ash.	Use of CaO-leached fly ash in lieu of coal fly ash.
Capacity	kg/hr	5830	5830	5830	5830	5830	5830	5830	2388	1565
Calcium carbonate in dried product	weight %	27%	27%	22%	27%	27%	27%	27%	62%	100%
INVESTMENT COST										
Total Investment (ISBL)	thousand $	$7,536	$7,256	$4,971	$8,287	$7,144	$7,709	$7,079	$4,703	$6,989
Total Direct Cost	*thousand $*	*$5,509*	*$5,304*	*$3,634*	*$6,058*	*$5,223*	*$5,635*	*$5,175*	*$3,438*	*$5,109*
Total Indirect Cost	*thousand $*	*$2,027*	*$1,952*	*$1,337*	*$2,229*	*$1,922*	*$2,074*	*$1,904*	*$1,265*	*$1,880*
OSBL (20% of ISBL)	thousand $	$1,507	$1,451	$994	$1,657	$1,429	$1,542	$1,416	$941	$1,398
Total Investment Cost (ISBL + OSBL)	thousand $	$9,043	$8,707	$5,965	$9,945	$8,573	$9,250	$8,495	$5,643	$8,386
OPERATING COST										
Total Operating Cost	$/kg product	$0.0012467	$0.0012467	$0.0012467	$0.0009496	$0.0041660	$0.0010082	$0.0042247	$0.0014798	$0.0094381
Electric Energy	*$/kg product*	*$0.0009428*	*$0.0009428*	*$0.0009428*	*$0.0009452*	*$0.0009452*	*$0.0009428*	*$0.0009428*	*$0.0011370*	*$0.0084661*
Cooling Water	*$/kg product*	*$0.0000003*	*$0.0000003*	*$0.0000003*	*$0.0000003*	*$0.0000003*	*$0.0000003*	*$0.0000003*	*$0.0000057*	*$0.0000120*
Well cooling water	*$/kg product*	*$0.0000041*	*$0.0000041*	*$0.0000041*	*$0.0000041*	*$0.0000041*	*$0.0000000*	*$0.0000000*	*$0.0000101*	*$0.0000154*
Air	*$/kg product*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*
Market purchased wild-type yeast	*$/kg product*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0032164*	*$0.0000000*	*$0.0032164*	*$0.0000000*	*$0.0000000*
Copper sulfate	*$/kg product*	*$0.0000091*	*$0.0000091*	*$0.0000091*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000223*	*$0.0000340*
Glucose	*$/kg product*	*$0.0002904*	*$0.0002904*	*$0.0002904*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0003047*	*$0.0009106*
Market purchased high temperature CA2	*$/kg product*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000000*	*$0.0000652*	*$0.0000652*	*$0.0000000*	*$0.0000000*
FIXED COST										
Total Fixed Cost	$/kg product	$0.0157	$0.0154	$0.0132	$0.0164	$0.0153	$0.0158	$0.0152	$0.0316	$0.0564
Total Direct Cost	*$/kg product*	*$0.0125*	*$0.0123*	*$0.0106*	*$0.0131*	*$0.0122*	*$0.0127*	*$0.0122*	*$0.0253*	*$0.0451*
*Labor (4 @ $54K/yr)****	*$/kg product*	*$0.0046*	*$0.0046*	*$0.0046*	*$0.0046*	*$0.0046*	*$0.0046*	*$0.0046*	*$0.0113*	*$0.0173*
*Maintenance (3%/yr)****		*$0.00582*	*$0.00560*	*$0.00384*	*$0.00640*	*$0.00552*	*$0.00595*	*$0.00546*	*$0.00886*	*$0.02010*
Direct overhead (45% labor cost)		*$0.00208*	*$0.00208*	*$0.00208*	*$0.00208*	*$0.00208*	*$0.00208*	*$0.00208*	*$0.00509*	*$0.00776*
Total Indirect Cost (25% of direct cost)	*$/kg product*	*$0.0031*	*$0.0031*	*$0.0026*	*$0.0033*	*$0.0031*	*$0.0032*	*$0.0030*	*$0.0063*	*$0.0113*
PRODUCT COSTS										
Cash Cost (Operating Cost + Fixed Cost)	$/kg product	$0.0169	$0.0166	$0.0144	$0.0173	$0.0195	$0.0168	$0.0195	$0.0331	$0.0658
Total Production Cost (Cash Cost + 15 yr Depreciation cost)	$/kg product	$0.0298	$0.0291	$0.0230	$0.0316	$0.0317	$0.0301	$0.0316	$0.0527	$0.1105
*Depreciation (15 years)****	*$/kg product*	*$0.0129*	*$0.0124*	*$0.0085*	*$0.0142*	*$0.0123*	*$0.0132*	*$0.0121*	*$0.0197*	*$0.0447*
Total Cost (Production Cost + 9 yr ROCE)	$/kg product	$0.0473	$0.0459	$0.0345	$0.0507	$0.0483	$0.0479	$0.0480	$0.0793	$0.1708
*ROCE****	*$/kg product*	*$0.0175*	*$0.0168*	*$0.0115*	*$0.0192*	*$0.0165*	*$0.0179*	*$0.0164*	*$0.0266*	*$0.0603*
Total Product Cost	$/tonne product	$47.3	$45.9	$34.5	$50.7	$48.3	$47.9	$48.0	$79.3	$170.8
CO_2 CAPTURE COSTS										
Total CO_2 Capture Cost	$/tonne CO_2	$402	$390	$293	$431	$410	$407	$408	$276	$389
Cost difference per tonne of carbon dioxide mineralized	($/tonne)		-$12	-$109	$29	$8	$5	$6	-$126	-$12

*The removal of the dryer generates product that is only 85% dry.

*** An 8000 hour year was used to convert labor and maintenance to an hourly cost and to calculate the depreciation and ROCE.

**** The paper mill ash process costs do not account for the transport of the paper mill ash to the CO_2 capture facility.

Supplementary Table 9: CO_2 capture cost calculations for the various process modifications discussed in this paper. The results in this table differ from those in Table 5 because the landfill tipping fee credit for non-disposal of fly ash is not included in this table. Thus, the CO_2 capture costs presented in this table represent the costs of the process if no purchaser for the mineralized CO_2 were found.

Capture of CO_2 using MEA absorption. Case 10 from Woods, et al. U.S. DoE, NETL, 2010.

Line	Value	Units	Description	Source	Calculation
W1	594,757	kg/h	CO_2 captured	Woods, et al. Case 10	
W2	549,960	kWe	net electicity production	Woods, et al. Case 10	
W3	61,200	kWe	CO_2 capture consumption	Woods, et al. Case 10	
W4	611,160	kWe	gross electricity production	Woods, et al. Case 10-A6	W2+W3
W5	0.10	kWh/kg	electrical energy /CO_2 captured	Woods, et al. Case 10	W3/W1
W6	0.97	kg/kWh	CO_2/gross electricity	Woods, et al. Case 10	W1/W4
W7	59	mills/kWh	Cost of electricity without CO_2 capture	Woods, et al. Case 9	
W8	140	mills/kWh	Levelized cost of electricity with CO_2 capture	Woods, et al. Case 10	

CO_2 mineralization using fly ash and bCA2-yeast

Line	Value	Units	Description	Source	Calculation
B1	686	kg/h	CO_2 captured	Barbero, et al. Fly ash and bCA2-yeast case.	
B2	668	kWe	gross electricity production		B1*W6
B3	0.0139	kWh/kg	electrical energy /CO_2 captured	Barbero, et al. Fly ash and bCA2-yeast case.	
B4	9.54	kWe	CO_2 capture consumption		B1*B3
B5	658	kWe	net electricity production		B2-B4
B6	59	mills/kWh	Cost of electricity without CO_2 capture	Woods, et al. Case 9	
B7	60	mills/kWh	including the reduction in electrical energy production		B6/B5*B2
B8	268	mills/kgCO_2	Cost of CO_2 capture	Barbero, et al. Fly ash and bCA2-yeast case.	
B9	279	mills/kWh	including the reduction in electrical energy production		B8*B1/B5
B10	340	mills/kWh	Levelized cost of electricity with CO_2 capture		B6*B2/B5+B8*B1/B5

CO_2 mineralization using fly ash and bCA2-yeast and sale of product to downstream user

Line	Value	Units	Description	Source	Calculation
B11	5,830	kg product / hr	Amount of product made by our process	Barbero, et al. Fly ash and bCA2-yeast case	
B12	8.7	kg product / kWh	Amount of product per kWh		B11/B2
B13	$10.00	$ / tonne product	Sale price of product	Benchmarked from reported fly ash and $CaCO_3$ prices	
B14	$0.01	$ / kg product	Sale price of product		B31 / 1000
B15	-$0.09	$ / kWh	Impact of sale of product on cost of electricty		B14*B12
B16	-87.33	mills / kWh	Impact of sale of product on cost of electricity		B15*1000
B17	252	mills/kWh	Levelized cost of electricity with CO_2 capture and sale of product		B10+B16

Supplementary Table 10: This table details the calculations used to arrive at a levelized cost of electricity for the CO_2 capture process described in this work. The cost of electricity is benchmarked against that reported for an MEA absorption process as describe in in Woods, et al., case 10. Sources for values in the table are listed in the source column and the calculations used to derive the other values are listed in the calculation column.

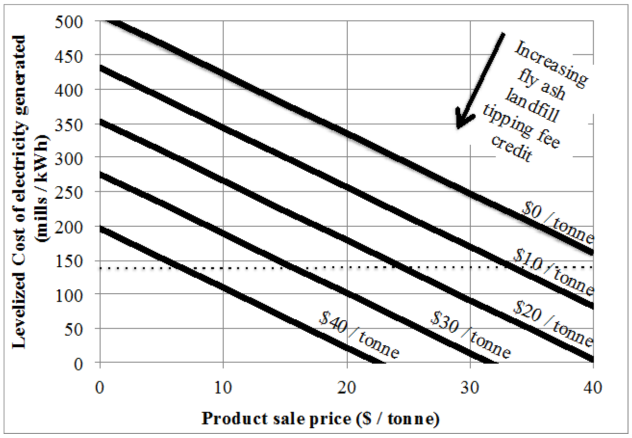
Calculating LCOE for CO_2 capture:

$$LCOE = \frac{(COE\ without\ CO2\ capture)(gross\ electricity\ production) + (cost\ of\ CO2\ capture)(CO2\ captured)}{Net\ electricity\ production} - \frac{(Product\ sale\ price)(product\ made\ per\ hour)}{gross\ electricity\ production}$$

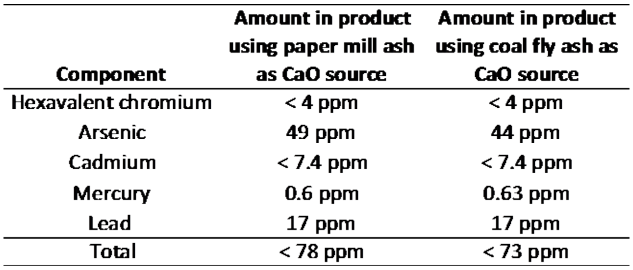
MEA absorption cost of electricity:
The MEA absorption process captures 595,000 kg of CO_2/hr. That process produces 549960 net kW with a consumption of 61200 kW for capture and compression (Woods, et al. 2010, Table 4-20). Thus, it captures 0.973 kg of CO_2/kWh gross with an electrical consumption of 0.103 kWh per kg CO_2.

CO_2 mineralization using bCA2-yeast and fly ash:
The bCA2-yeast and fly ash process captures 686 kg/hr of CO_2 (85% CO_2 capture efficiency on CO_2 in flue gas from supplementary table 3), which corresponds to 668 kWe gross. There is an electrical consumption of 0.0139 kWh per kg CO_2 which gives

658 kWe of net production. For this table 268 mills per kg CO_2 was used, which is the resulting cost of CO_2 capture for the fly ash and bCA2-yeast case that incorporates a fly ash landfill tipping fee credit of $17.89/tonne (from Woods, et al. 2010). Using a cost of electricity of of 59.4 mills/kWh (Woods, et al., case 9) there is a levelized cost of electricity of 340 mills/kWh with CO_2 capture. Selling the mineralized CO_2 product at $10/tonne (which is at the low end of the range of prices that were explored) would reduce the cost of to 252 mills/kWh. The impact of the value of the landfill tipping fee and the product sale prices on the levelized cost of electricity for the bCA2-yeast and fly ash process is reported in figure 4. The impact of the value of the landfill tipping fee and the product sale prices on the levelized cost of electricity for the process with fly ash and no biological components is reported in supplementary figure 6.



Supplementary Figure 6: This figure shows the impact that the fly ash landfill tipping fee credit and the sale price of the mineralized CO_2 product have on the levelized cost of electricity for a power plant with a CO_2 mineralization process utilizing fly ash and with no biological components. Each of the thick black lines shows the LCOE for a set fly ash landfill tipping fee. The x-axis is the product sale price and the y-axis is the LCOE of the process. The horizontal dashed line at 139 mills / kWh represents the reported LCOE for a pulverized coal subcritical power plant capturing 90% of the CO_2 using an MEA absorption process reported by Woods, et al. 2010. Note that the LCOE for the MEA process does not include CO_2 storage costs. Without bCA2-yeast, mineralization of CO_2 using fly ash is roughly 30 mills / kWh more expensive.



Component	Amount in product using paper mill ash as CaO source	Amount in product using coal fly ash as CaO source
Hexavalent chromium	< 4 ppm	< 4 ppm
Arsenic	49 ppm	44 ppm
Cadmium	< 7.4 ppm	< 7.4 ppm
Mercury	0.6 ppm	0.63 ppm
Lead	17 ppm	17 ppm
Total	< 78 ppm	< 73 ppm

Supplementary Table 11: Mineralized CO_2 from the lab-scale reactor using coal fly ash and using paper mill ash as the CaO sources was analyzed for lead, mercury, cadmium, hexavalent chromium, and arsenic. Both CaO sources produced material that met the Toxics in Packaging Materials requirement of less than 100 ppm of all five of these metals in total. Both CaO sources produced material that had ~ 50% SiO_2 by weight.