Impact of alkalinity sources on the life-cycle energy efficiency of mineral carbonation for CO_2 storage

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SUPPORTING INFORMATION

Methodology

LCA Model

Examples of direct and indirect consumption in two sectors (natural gas and steel) are shown in Table 1. In this example, it can be seen that consuming 1 MJ of natural gas directly in the reactor results in the consumption of more than 1 MJ of natural gas in total, due to upstream use (e.g., in pipeline compressors), as well as consumption of other primary fuels such as coal (e.g., indirectly consumed in electricity used during natural gas processing and distribution). For the purposes of this paper, this indirect consumption is considered as the embodied energy contained in a product or energy source (i.e., direct energy content of the energy product is not considered embodied energy).

SI Table 1. Example of direct and embodied energy (economy-wide) inputs per unit of energy (natural gas) and material consumed (steel)

	Natural Gas		Steel
	Direct (MJ / MJ natural gas)	**Embodied (MJ / MJ natural gas)**	**Embodied (MJ / tonne of steel)**
Coal	0.0	0.032	11254
Petroleum	0.0	0.007	1343
Natural Gas	1.0	0.037	5973
Bio/waste	0.0	0.001	226
Non-fossil electricity	0.0	0.008	2611

The life cycle energy consumption values are then converted to equivalent CO_2 emissions using CO_2 emissions factors, provided in Table 2.[1]

SI Table 2. CO_2 emission factors for primary and secondary fuels

	g CO_2 / MJ fuel consumed
Coal	89.1
Petroleum	70.6
Natural Gas	50.3
Bio/waste	0.0
Non-fossil electricity	0.0
Coke	96.8
Residual Fuel	74.0
Diesel	69.3
Gasoline	67.2
Average electricity	176.8

For extraction and transportation process stages, the embodied energy in capital investment is accounted for within the EIO-LCA results for the corresponding NAICS sectors; for other process stages the embodied energy in capital investment is specified below.

Process modeling

Physical preprocessing

SI Table 3. Bond work indices for alkalinity sources

	W_i	**Case used**
Blast furnace slag	12.00	SS

Cement clinker	13.49	CKD
Dolomite	11.31	Olivine
FA	13.39	FA
Serpentine	11.61	Serpentine

Chemical conversion (dissolution and precipitation reactions)

Heating

Table 4 provides the heat capacity and heat of reaction used for various reaction materials.

SI Table 4. Heat capacities, C_p, and heat of reactions, ΔH, for reaction materials

	C_P (MJ/t-K)	**ΔH (kJ/mol)**
CKD	0.84	-179
FA	0.84	-179
Olivine	0.805	-88
Serpentine	1.09	-35
SS	0.49	-179
Water	4.19	--

Table 5 provides the thermal conductivity, k, and emissivity, ε, for tank materials.[2]

SI Table 5. Thermal conductivity and emissivity for tank materials

	k (W/m-K)	**ε (unitless)**
Insulation (mineral fiber)	0.058	0.050
Stainless steel	16.25	0.195

Mixing and mass transfer

There are two different reaction factors that could limit the reaction speed. First, the mass transfer of CO_2 into the liquid phase could be limiting. Second, the rate of dissolved CO_2 removal through the carbonation reaction could be limiting. We assume here that the carbonation reaction rate is the limiting rate, and that this reaction is limited by the alkaline feedstock dissolution (rather than the precipitation of carbonate minerals, which is comparatively fast). Thus, we scale reactor size and throughput rates using the rate of reaction, not the rate of

CO_2 diffusion into the liquid phase. Two arguments are presented below to suggest that this is a reasonable assumption.

First, the type of mixing has been selected to so that the conditions are fully turbulent for the high and low pressure cases. For instance, the Reynolds number for the stirred reactor vessel for the Ol-155˚C case = 1,320,000 and for the FA-25˚C case = 7,090,000, calculated according to:

$$Re = ND^2 \frac{\rho}{\mu}$$

where N = the impeller speed (rps), D = impeller diameter (m), ρ = mixture density (kg/m^3), and μ = mixture viscosity (kg/m-s). Given the turbulent flow in the tanks, additional recirculation of CO_2 to ensure adequate mixing and mass transfer to the liquid phase (beyond the mixing already included in the design) was deemed unnecessary.

Second, the assumed rate of mass transfer of CO_2 from the gas phase to the liquid phase has also been considered, to ensure that our system design is reasonable in terms of mass transfer and that no additional recirculation is required to ensure adequate dissolution. We include calculations of the mass transfer of CO_2 in two cases: FA—25˚C (1 bar, 25˚C) and Ol-155˚C (100 bar, 150˚C). Table 6 table summarizes the parameters and data used for the calculations, which are explained in detail below.

SI Table 6. Parameters for mass transfer calculations

Parameter	unit	FA-25˚C	Ol-155˚C
T	˚K	298.15	428.15
P	bar	1.00	100.00
c*	M	0.08	0.73
μ_{H2O}	Pa-s	8.90E-04	1.79E-04
ρ_{H2O}	kg/m^3	997.05	917.69
R_{CO2}	m	1.28E-10	1.28E-10
D_{CO2}	cm^2/s	1.92E-05	1.37E-08
ν_{H2O}	m^2/s	8.93E-07	1.95E-07
d	mm	0.50	0.50
P/V	kg/m-s^3	115.00	1249.63
f_{CO2}	mol/s	262.99	262.99

The diffusion of CO_2, D_{CO2}, into water at 25°C, 1 bar is 1.92 x 10^{-5}.[3] From this value, the radius of CO_2, R_{CO2}, can be estimated according to:

$$R_{CO2} = \frac{k_B T}{6\pi\mu D} \qquad \text{(Eq. 1)}$$

where k_B is the Boltzmann constant equal to 1.38x10^{-16} g cm^2/s^2-K, T is the temperature, μ is the dynamic viscosity of the solvent. R_{CO2} = 1.28 x 10^{-10} m, and this value can be used to determine the D_{CO2} for the Ol-155°C (using Eq. 1).

For mass transfer enhanced by dispersion of bubbles into a stirred tank, the mass transfer coefficient of CO_2, k, into the fluid can be determined from D_{CO2}, according to:

$$\frac{k\,d}{D_{CO2}} = 0.13\left(\frac{d^4(P/V)}{\rho\upsilon^3}\right)^{1/4}\left(\frac{\upsilon}{D_{CO2}}\right)^{1/3} \qquad \text{(Eq. 2)}$$

where d is the bubble diameter, P/V is the mixing power per volume, ρ is the density of water, v is the kinematic viscosity of water.[4]

The flux of CO_2 through the gas-liquid interface can be described based on the mass transfer coefficient according to:

$$V\frac{dc}{dt} = AN = Ak(c^* - c) \qquad \text{(Eq. 3)}$$

where V is the volume of solution (m^3), A is the area of the interface (m^2), N is the mass-transfer flux (mole/m^2-s), k is the mass-transfer coefficient m/s, c is the initial concentration of CO_2 in the water assumed to equal 0 M, and c^* is the concentration of CO_2 in the water at equilibrium at given values of temperature and pressure, estimated using EQ3/6 in conjunction to the Redlich-Wong equation of state for a water-CO_2 mixture.[5, 6]

By dividing the input rate of CO_2, f_{CO2}, by the calculated N, the required interfacial area, A_{req}, can be determined. The interfacial area required can be normalized to the total tank cross-sectional area of the system, A_{req}/A_{cross}. Table 7 shows the results for the mass transfer calculations.

SI Table 7. Results for mass transfer calculations

Parameter	unit	FA-25°C	Ol-155°C
k	m/s	3.88E-05	5.04E-04
N	mol/m^2-s	3.00E-03	3.69E-01
A_{req}	m^2	87561.50	713.25
A_{req}/A_{cross}		28.5	0.7

To ensure that CO_2 recirculation is not necessary, the actual interfacial area of the system must at least be equal to the A_{req} based on the mass transfer calculations, to ensure that the rate of CO_2 uptake into the solution is fast enough. For both cases the required interfacial areas and ratios to tank cross-sectional areas are reasonable. While the design / selection of a CO_2 dispersion system was beyond the scope of this work, the required interfacial areas are low enough that it is reasonable to assume a dispersion system could be used to create enough area to ensure adequate interphase mass transfer. It is important to keep in mind that our system is scaled (i.e., the number of reactors / amount of total reactor volume) based on the amount of CO_2 the alkalinity source can react with at the selected operating conditions, and it is assumed that the CO_2 injection rate will be tailored such that a steady state is reached (i.e., just enough CO_2 is injected to saturate the fluid and maintain CO_2 pressure in the tank).

Post-reaction processing

The energy and separation data for the various separation processes are summarized in Table 6.

SI Table 6. Separation process parameters

Parameter	unit	Clarifier	Liquid	Centrifugal

			cyclone	**filter**
diameter or width	*m*	25	1	1.4
volumetric flow rate	m^3/s	0.20	0.075	0.076
product % solid	*wt. %*	30	50	88
% water separated	*vol. %*	75	0	0
water recyclable	*yes/no*	yes	no	no
power consumption	*kW*	2.8	15.0	462.7

Model uncertainty

In order to evaluate model uncertainty, process efficiency ranges are defined based on values from literature or reasonable ranges of assumption. Table 7 summarizes the efficiency assumptions used to estimate the uncertainty in results.

Process	**Efficiency basis**	**Inefficient**	**Average**	**Efficient**
Extraction	efficient and average case correspond to low intensity mining, inefficient case corresponds to high intensity mining	high intensity	low intensity	low intensity
Train transport	percentage of calculated results	120%	100%	85%
Truck transport	percentage of calculated results	105%	100%	90%
Grinding	percentage of energy input to electric motor converted to power	90%	95%	100%
Reaction	percentage of materials reacted	80%	90%	100%
Mixing	percentage of energy input to electric motor converted to power	90%	95%	100%
Heating	percentage of energy input to electric motor converted to power	90%	95%	100%
Water recycle	percentage of heat loss during separations	30%	20%	10%
Separation	percentage of energy input to electric motor converted to power	90%	95%	100%
Disposal	(same as mining)	high intensity	low intensity	low intensity

SI Table 7. Process efficiency assumptions

References

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