Electronic Supporting Information for:

Rock 'n' use of CO_2: Carbon footprint of carbon capture and utilization by mineralization

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S1. Functional unit for CCU by mineralization

There are several options for defining the functional unit of a LCA study on CCU by mineralization. Here, we discuss and compare 4 most common ones:

1. The *product of a mineralization pathway* is a suitable functional unit to compare diverse pathways that produce one identical product. This functional unit is, however, unsuitable to directly compare the LCA results to CCS or CCU by mineralization pathways producing not identical products.
2. The *main product of CO_2 source* (e.g., electricity) is a good choice for the functional unit to compare a variety of CCS by mineralization or CCU by mineralization pathways or other measures for climate change mitigation (e.g., geological storage, renewable energy) installed at one specific CO_2 source. If the main product of the CO_2 source is chosen as functional unit, the results of a LCA study depend strongly on the chosen CO_2 point source and are therefore difficult to interpret and adopt for other CO_2 point sources.
3. The *treated CO_2* can be used as a functional unit to compare technologies that capture and store CO_2. In this case, CCU by mineralization is regarded as a technology for off-gas treatment and can be compared to no action or alternative technologies. The LCA study thus starts with the raw off-gas and thus needs to explicitly consider all CO_{2e} emissions due to leakage or low efficiency of the

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technologies in CO_2 treatment and removal. As CCU by mineralization pathways are still in developing phase, required data for treated CO_2 (e.g., purity) is not always available.

4. The *stored CO_2* is a practical functional unit to compare CCS by mineralization or CCU by mineralization technologies. The technologies can produce diverse products and be installed at various CO_2 sources, but their main function in the LCA is now regarded as storage of CO_2.

S2. Life cycle stages for CCU by mineralization

Here, we describe the life cycle stages inside the CCU by mineralization system boundary (section 2.1) in more detail and discuss the required data for calculation of life cycle inventory for CCU by mineralization in general. Note that all the life cycle stages require data for mass balances and for the source and amount of energy (electricity, fossil fuel or thermal energy).

Feedstock supply includes all processes required to provide the feedstock to the mineralization plant (e.g., mining, transport). The feedstock of mineralization can be industrial by-products (e.g., steel slag) or natural minerals (e.g., serpentine, olivine). In contrast to natural minerals, no mining is required for industrial by-products. For the feedstock supply, transportation has to be considered including distance and transportation method (truck, train, or ship).

CO_2 supply stage covers the necessary processes to supply the CO_2 to the mineralization plant. In general, the CO_2 required for mineralization is obtained from a diluted gas stream, e.g., power plant off-gas, cement plant off-gas. In the CO_2 supply stage, CO_2 is separated (if required), compressed to the required pressure, and transported (if required). The effort of CO_2 supply has to be considered in the system boundary of the CCU system (cf. section 2.1). The life cycle inventory of CO_2 supply also depends on the CO_2 capture technology[1], the required CO_2 concentration and pressure for the CCU by mineralization pathway. Note that different pathways for CCU by mineralization require different CO_2 concentrations (pure CO_2 or 10-30% CO_2, off-gas)[2–4]. Hence, in some cases, CO_2 capture is not even required. Furthermore, most CO_2 capture technologies use special chemicals (e.g., solvents, adsorbents). The amount and environmental impacts of the required chemicals should also be included in the life cycle inventory of CO_2 supply stage.

Pretreatment consists of the required processes to prepare or activate the feedstock for the carbonation stage. Pretreatment of the feedstock is usually recommended to improve the slow reaction kinetics of carbonation. The pretreatment stage of the feedstock can be divided into 3 major categories: mechanical, thermal, and chemical treatment. The mechanical treatment produces more surface[5], the thermal treatment uses heat to change the morphology[6] and produces pores, while the chemical treatment dissolves impurities[7] and produces new pores for a higher rate of dissolution and reaction. The life cycle inventory of the pretreatment stage thus also requires the amount and environmental impacts of the required chemicals and whether they can be recycled.

The *Carbonation* stage of a mineralization pathway is where CO_2 reacts with magnesium oxide or calcium oxide. Carbonation technologies can be divided into two concepts: direct and indirect. Simply put, the direct mineralization concept directly react CO_2 with the feedstock in one step, whereas the indirect mineralization concept breaks down the process into multiple steps.[8]

The most common direct mineralization pathway is aqueous carbonation, in which CO_2 reacts with the feedstock in aqueous solution. The process includes three main chemical processes: dissolution of CO_2 in water, dissolution of alkaline earth metal from the feedstock in water, and the subsequent carbonation reaction. All processes for aqueous carbonation occur in one reactor.

In most indirect mineralization pathways, alkaline earth metals are leached out from the feedstock with help of a solvent in the first step of the carbonation process. In a subsequent step, the alkaline earth metals react with CO_2.

The life cycle inventory of the carbonation stage also depends on the environmental impacts of the required chemical and/or additives, the type of reactor, the reaction temperature, water losses, solvent recovery, the reaction yield, and the solid/liquid ratio of the carbonation stage.

Post-processing summarizes the required processes to prepare the product for the market (e.g., separation, classification, dewatering, drying, and transportation). The life cycle inventory of post-processing depends highly on the specific application of the product.

Use is the life cycle stage where we calculate the environmental impact avoided due to substitution of a conventional product (substitution credit). In general, for mineralization products, several applications have been proposed (Table S1), which differ widely in market size and price: from market sizes as large as 35 Gt/year for concrete admixture to small markets of 1.4 Mt/year for pigments; and prices varying from approximately 7 to 12000 €/ton.

Table S1: Potential applications (use stage) for end products of mineralization with approximate market size and price

End Product	**Application of mineralization product**	**Market size**	**Price [€/ton]**
Concrete or asphalt	Fine aggregates as admixture [9–11]	10-35 Gt/year[12]	7[13]
Iron and steel	Iron ore[14 15]	2.4 Gt/year[16]	57[16]
Blended cement	Pozzolan or self-cementing (SCM)[17,18,18–20,20]	1 Gt/year[21]	81[13]
Stabilized wastes	Reclassification of hazardous wastes[22]	1 Gt/year[23]	100[16]
Glass	Silicon dioxide (SiO_2) as feedstock[16]	58 Mt/year[24]	50[13]
Paper or tire	PCC* or PMC** as admixture or filler[25]	13 Mt/year[16]	350[16]
Nickel	Nickel feedstock[26]	2.3 Mt /year[13]	12000[13]
Pigment	Iron oxide and hydroxide as pigment[27]	1.4 Mt/year[16]	142[16]

* Precipitated calcium carbonate
**precipitated magnesium carbonate

S3. Sensitivity analysis on electricity grid mix

The pathways for CCU by mineralization are energy intensive. Thus, the carbon footprint of the electricity grid mix can have a high impact on the total carbon footprint of CCU by mineralization pathways. To analyze this effect, we computed the carbon footprints of the 7 CCU by mineralization pathways for the state-of-the-art scenario as function of the carbon footprint of the electricity grid mix (Figure S1). The electricity grid mix of Europe in 2014 (cf. Section S5) is assumed in both the ideal-mineralization and the state-of-the-art scenario.

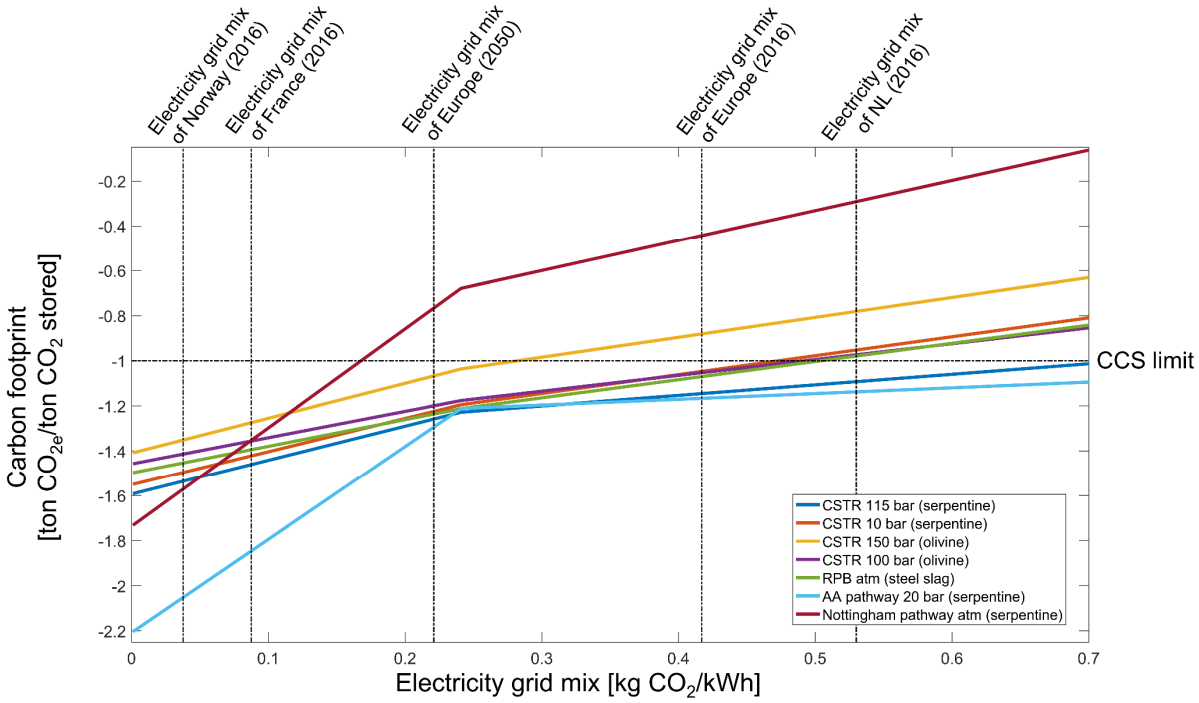

Figure S1: Carbon footprint for 7 CCU by mineralization pathways as function of carbon footprint of the electricity grid mix for the state-of-the-art scenario. CCS limit is the maximum potential of CCS technologies. RPB - rotary packed bed, AA – Abo Academy.

As expected, decreasing the carbon footprint of electricity grid mix decreases the total carbon footprint of all CCU by mineralization pathways. At the point where the carbon footprint of electricity is lower than carbon footprint of natural gas combustion (cf. Section S5), thermal energy is supplied by electric heating; due to this reason, the slopes of the graphs change at 241 gr CO_{2e}/kWh. Thus, from 700 to 241 grCO_{2e}/ kWh, the slope of the graphs depends only on the electricity demand of the CCU pathway; for electricity grid mixes lower than 241 grCO_{2e}/ kWh, the slope of the graphs depends on both the electricity and thermal energy demands of the CCU pathway.

With the electricity grid mix of France, all 7 CCU pathways avoid more than 1 ton CO_{2e} by mineralization and utilization of 1 ton CO_2. Due to reagent recovery processes, indirect pathways (Abo Academy and Nottingham pathways) require a higher amount of thermal energy than direct pathways; therefore, the dependence on the electricity carbon footprint becomes stronger once also heating is provided by electricity for the Abo Academy and Nottingham pathways.

In summary, changing the electricity grid mix from Europe-mix to France can change the carbon footprint of CCU by mineralization pathways dramatically. E.g., the carbon footprint of the Nottingham pathway decreases from -0.44 ton CO_{2e}/ ton CO_2 stored for the electricity grid of Europe-mix to -1.35 ton CO_{2e}/ ton CO_2 stored for the electricity grid of France. Therefore, proper accounting for the employed electricity is essential.

S4. Sensitivity analysis on feedstock transport distance

Large amounts of bulk feedstock need to be transported to the mineralization plants (2.5-5 ton feedstock/ton CO_2 stored). Thus, long feedstock transport distances can be critical for the carbon footprint of CCU by mineralization pathways. To analyze the impact, we compute the carbon footprints of the 7 CCU pathways for the state-of-the-art scenario as function of feedstock transport distance (Figure S2). The feedstock transport distance of 260 km has been assumed both in the ideal-mineralization and the state-of-the-art scenarios in the main text.

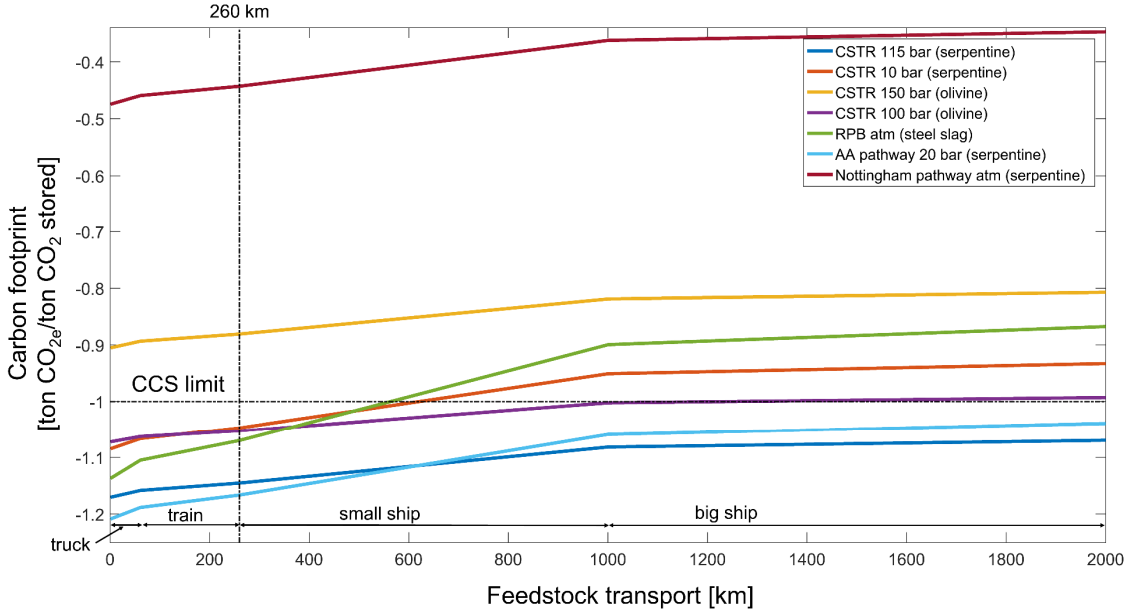

Figure S2: Carbon footprint for the 7 CCU by mineralization pathways as function of feedstock transport distance for the state-of-the-art scenario, 260 km is the assumption of both the ideal-mineralization and the state-of-the-art scenarios. CCS limit is the maximum potential of CCS technologies. RPB - rotary packed bed, AA – Abo Academy.

Increasing the feedstock transport distance increases the carbon footprint of all CCU by mineralization pathways. The slopes of the graphs change due to different transport methods: The first 60 km of transport is assumed by trucks that have high emissions (79 $grCO_{2e}$/ (km.ton)[28], steep slope).[29] The second transport method is by train until 260 km (25 $grCO_{2e}$/ (km.ton)[28], moderate slope).[29] After that, small ships are the common transport method until 1000 km, with 33 $grCO_{2e}$/ (km.ton)[30] (moderate slope).[29] For distances larger than 1000 km, big ships are needed with 4.7 $grCO_{2e}$/(km.ton)[30] emission (gradual slope).[31] Feedstock transport distance has a higher effect on pathways that require high amounts of solid handling. High solid handling is required for low reaction yields (e.g., Abo Academy

and Nottingham pathways) or low feedstock purity (e.g., rotary packed bed pathway). Overall, the impact of feedstock transport distance on the carbon footprints of the CCU by mineralization pathways is moderate. Due to the fact that the methods for long distance transport emit much less CO_{2e} than short distance transport methods, the effect of the feedstock transport distance is negligible after the first 1000 km.

S5. Considered LCA data sets for CCU by mineralization pathways

Table S2: Considered LCA data sets for CCU by mineralization pathways [28,32]

Product	Name of data set	Year	Database
Electricity	Electricity grid mix [EU-28]	2016	GaBi ts
Electricity	Electricity grid mix (2050) [EU-28]	2016	GaBi ts
Electricity	Electricity grid mix [NL]	2016	GaBi ts
Electricity	Electricity grid mix [FR]	2016	GaBi ts
Electricity	Electricity grid mix [NO]	2016	GaBi ts
Heat	Thermal energy from natural gas [EU-28]	2016	GaBi ts
Transport truck	Transport, small truck (up to 14 t total cap., 9.3t payload) [EU-28]	2016	GaBi ts
Transport train	Rail transport, average train, gross tonne weight 1000t / 726t payload capacity [EU-28]	2016	GaBi ts
NaCl	Sodium chloride (rock salt) [EU-28]	2016	GaBi ts
$NaHCO_3$	Sodium bicarbonate	2017	GaBi ts
Water	Process water [EU-28]	2016	GaBi ts
NH_3	Ammonia mix (NH3) [EU-28]	2016	GaBi ts
$MgSO_4$	Magnesium sulfate production [RoW]	2015	ecoinvent 3.3
$(NH_4)_2SO_4$	Ammonium sulfate production [RoW]	2015	ecoinvent 3.3
Diesel	Diesel mix at refinery [EU-28]	2013	GaBi ts
Ammonium nitrate	Ammonium nitrate production	2015	ecoinvent 3.3
Factory construction	Magnesium factory construction [RER]	-	ecoinvent 3.5

S6. Carbon footprint of background processes for CCU by mineralization

Table S3: Carbon footprint of background processes for CCU by mineralization pathways [28,32]

Product	Name of data set	Year	Database	Carbon footprint
Electricity	Electricity grid mix [EU-28]	2016	SP 39, GaBi databases 2019 ts	417 [gr CO_{2e}/kWh]
Heat	Thermal energy from natural gas [EU-28]	2016	SP 39, GaBi databases 2019 ts	241 [gr CO_{2e}/kWh]
Transport truck	Transport, small truck (up to 14 t total cap., 9.3t payload) [EU-28]	2016	SP 39, GaBi databases 2019 ts	79.1 [gr CO_{2e}/(km.t)]
Transport train	Rail transport, average train, gross tonne weight 1000t / 726t payload capacity [EU-28]	2016	SP 39, GaBi databases 2019 ts	25.8 [gr CO_{2e}/(km.t)]
source	GaBi Software and Database for Life Cycle Engneering			

S7. Laboratory data for the considered CCU by mineralization pathways

Table S4: Laboratory data for the considered CCU by mineralization pathways (RPB - rotary packed bed, AA – Abo Academy.)[4,33–38]

Mineralization Pathway	Heat pretreatment	Particle size [µm]	Pure CO_2	Carbonation reaction temperature [°C]	Overall reaction yield
CSTR 115 bar (serpentine)	Yes	37	Yes	155	92%
CSTR 10 bar (serpentine)	Yes	37	No	40	61%
CSTR 150 bar (olivine)	No	<10	Yes	185	81%
CSTR 100 bar (olivine)	No	<10	Yes	190	100%
RPB atm (steelslag)	No	125	No	25	48%
AA pathways (serpentine)	Yes	75	Yes	510	55%
Nottingham pathway (serpentine)	No	75	No	80	87%

S8. Life cycle inventory data of CCU by mineralization pathways for state-of-the-art scenario

Table S5: LCI data of CSTR 115 bar using serpentine for state-of-the-art scenario

Life cycle stage	Electricity [kWh/ton stored CO_2]	Thermal energy [kWh/ton stored CO_2]	Carbon footprint due to material use and transport [kg CO_{2e}/ton stored CO_2]	Total carbon footprint of the stage [kg CO_{2e}/ton stored CO_2]
Feedstock supply	13	-	25	30
Pre-treatment	215	292**	-7***	153
CO_2 in off-gas	-	-	-1000	-1000
CO_2 supply	98	650**	5	202
Carbonation	96	0**	85	125
Post-processing	53	-	-	22
Use	-	-	-710*	-710
Factory construction	-	-	34	34

* Credit due to ordinary Portland cement substitution

** Heat integration has been applied

*** Credit due to iron ore substitution

Table S6: LCI data of CSTR 10 bar using serpentine for state-of-the-art scenario

Life cycle stage	Electricity [kWh/ton stored CO_2]	Thermal energy [kWh/ton stored CO_2]	Carbon footprint due to material use and transport [kg CO_{2e}/ton stored CO_2]	Total carbon footprint of the stage [kg CO_{2e}/ton stored CO_2]
Feedstock supply	20	-	38	46
Pre-treatment	324	441**	-11***	230
CO_2 in off-gas	-	-	-1000	-1000
CO_2 supply	384	0	0	160
Carbonation	37	69	129	160
Post-processing	73	0	0	30
Use	0	0	-710*	-710
Factory construction	-	-	34	34

* Credit due to ordinary Portland cement substitution

** Heat integration has been applied

*** Credit due to iron ore substitution

Table S7: LCI data of CSTR 150 bar using olivine for state-of-the-art scenario

Life cycle stage	Electricity [kWh/ton stored CO_2]	Thermal energy [kWh/ton stored CO_2]	Carbon footprint due to material use and transport [kg CO_{2e}/ton stored CO_2]	Total carbon footprint of the stage [kg CO_{2e}/ton stored CO_2]
Feedstock supply	13	-	25	30
Pre-treatment	584	-	-	243
CO_2 in off-gas	-	-	-1000	-1000
CO_2 supply	103	530**	5	176
Carbonation	118	0**	93	142
Post-processing	61	-	-	25
Use	-	-	-531*	-531
Factory construction	-	-	34	34

* Credit due to ordinary Portland cement substitution

** Heat integration has been applied

Table S8: LCI data of CSTR 100 bar using olivine for state-of-the-art scenario

Life cycle stage	Electricity [kWh/ton stored CO_2]	Thermal energy [kWh/ton stored CO_2]	Carbon footprint due to material use and transport [kg CO_{2e}/ton stored CO_2]	Total carbon footprint of the stage [kg CO_{2e}/ton stored CO_2]
Feedstock supply	11	-	20	24
Pre-treatment	473	-	-	197
CO_2 in off-gas	-	-	-1000	-1000
CO_2 supply	93	358**	5	130
Carbonation	70	0**	43	72
Post-processing	53	-	-	22
Use	-	-	-531*	-531
Factory construction	-	-	34	34

* Credit due to ordinary Portland cement substitution

** Heat integration has been applied

*** Credit due to iron ore substitution

Table S9: LCI data of rotary packed bed pathway (RPB atm) using steel slag for state-of-the-art scenario

Life cycle stage	Electricity [kWh/ton stored CO_2]	Thermal energy [kWh/ton stored CO_2]	Carbon footprint due to material use and transport [kg CO_{2e}/ton stored CO_2]	Total carbon footprint of the stage [kg CO_{2e}/ton stored CO_2]
Feedstock supply	-	-	67	67
Pre-treatment	307	-	-	128
CO_2 in off-gas	-	-	-1000	-1000
CO_2 supply	0.3	-	-	0.1
Carbonation	330	-	38	175
Post-processing	137	-	-	57
Use	-	-	-531*	-531
Factory construction	-	-	34	34

* Credit due to ordinary Portland cement substitution

Table S10: LCI data of Abo Academy (AA) pathway using serpentine for state-of-the-art scenario

Life cycle stage	Electricity [kWh/ton stored CO_2]	Thermal energy [kWh/ton stored CO_2]	Carbon footprint due to material use and transport [kg CO_{2e}/ton stored CO_2]	Total carbon footprint of the stage [kg CO_{2e}/ton stored CO_2]
Feedstock supply	22	-	42	51
Pre-treatment	62	493**	-12***	133
CO_2 in off-gas	-	-	-1000	-1000
CO_2 supply	76	833**	5	237
Carbonation	17	2403**	56	642
Post-processing	82	-	-	34
Use	-	-	-1297*	-1297
Factory construction	-	-	34	34

* Credit due to ordinary Portland cement substitution

** Heat integration has been applied

*** Credit due to iron ore substitution

Table S11: LCI data of Nottingham pathway using serpentine for state-of-the-art scenario

Life cycle stage	Electricity [kWh/ton stored CO_2]	Thermal energy [kWh/ton stored CO_2]	Carbon footprint due to material use and transport [kg CO_{2e}/ton stored CO_2]	Total carbon footprint of the stage [kg CO_{2e}/ton stored CO_2]
Feedstock supply	17	-	32	39
Pre-treatment	46	0	-9***	10
CO_2 in off-gas	-	-	-1000	-1000
CO_2 supply	-	-	158	158
Carbonation	985****	2947**	54	1175
Post-processing	64	-	-	27
Use	-	-	-886*	-886
Factory construction	-	-	34	34

* Credit due to ordinary Portland cement substitution

** Heat integration has been applied

*** Credit due to iron ore substitution

**** Including the electricity demand of a compressor with polytropic efficiency of 86% for vapor recompression of the steam from the regeneration process to 1.44 bar

S9. Mass balance of the main components of CCU by mineralization pathways for state-of-the-art scenario

Table S12: Mass balance of the main components CCU by mineralization pathway for state-of-the-art scenario, all the numbers are in ton per ton CO_2 stored

Mineralization Pathway	Component	Feedstock supply	Pre-treatment	CO_2 supply	Carbonation	Post-processing	Use
CSTR 115 bar (serpentine)	Serpentine	2.3	2.3	-	0.18	0.18	-
	Magnetite	0.25	-0.25	-	-	-	-
	CO_2	-	-	1	-	-	-
	SiO_2	-	-	-	0.91	0.91	-0.91
	$MgCO_3$	-	-	-	1.92	1.92	-
	Water	-	-	-	0.28	-0.28	-
CSTR 10 bar (serpentine)	Serpentine	3.44	3.44	-	1.33	1.33	-
	Magnetite	0.38	-0.38	-	-	-	-
	CO_2	-	-	1	-	-	-
	SiO_2	-		-	0.91	0.91	-0.91
	$MgCO_3$	-		-	1.92	1.92	-
	Water	-		-	0.28	-0.28	-
CSTR 150 bar (olivine)	Olivine	1.97	1.97	-	0.37	0.37	-
	Impurity	0.5	0.5	-	0.5	0.5	-
	CO_2	-	-	1	-	-	-
	SiO_2	-	-	-	0.68	0.68	-0.68
	$MgCO_3$	-	-	-	1.92	1.92	-
	Water	-	-	-	-	-	-
CSTR 100 bar (olivine)	Olivine	1.6	1.6	-	0	0	-
	Impurity	0.4	0.4	-	0.4	0.4	-
	CO_2	-	-	1	-	-	-
	SiO_2	-	-	-	0.68	0.68	-0.68
	$MgCO_3$	-	-	-	1.92	1.92	-
	Water	-	-	-	-	-	-
RPB atm (steelslag)	Steel slag	4.05	4.05	-	2.1	2.1	
	Impurity	2.7	2.7	-	2.7	2.7	-
	CO_2	-	-	1	-	-	-
	SiO_2	-	-	-	0.68	0.68	-0.68
	$CaCO_3$	-	-	-	2.27	2.27	-
	Water	-	-	-	-	-	-
AA pathways (serpentine)	Serpentine	3.85	3.85	-	-	-	-
	Magnetite	0.43	-0.43	-	-	-	-
	CO_2	-	-	1	-	-	-
	SiO_2	-	-	-	1.67	1.67	-1.67
	$MgCO_3$	-	-	-	1.92	1.92	-
	Rest*	-	-	-	1.26	1.26	-
Nottingham pathway (serpentine)	Serpentine	2.88	2.88	-	-	-	-
	Magnetite	0.32	-0.32	-	-	-	-
	CO_2	-	-	1	-	-	-
	SiO_2	-	-	-	1.14	1.14	-1.14
	$MgCO_3$**	-	-	-	2.25	2.25	-
	Rest*	-	-	-	0.49	0.49	-

* The amount of feedstock in intermediate products

** In form of hydromagnesite

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