



## Carbonation Workshop

# Carbon mineralization: principles, main process routes and product properties

**Giulia Costa**

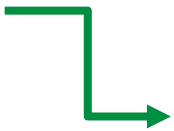


**TOR VERGATA**  
UNIVERSITÀ DEGLI STUDI DI ROMA

*Laboratory of Environmental Engineering  
Department of Civil Engineering and Computer Science Engineering  
University of Rome "Tor Vergata", Italy*

## Topics that we will address

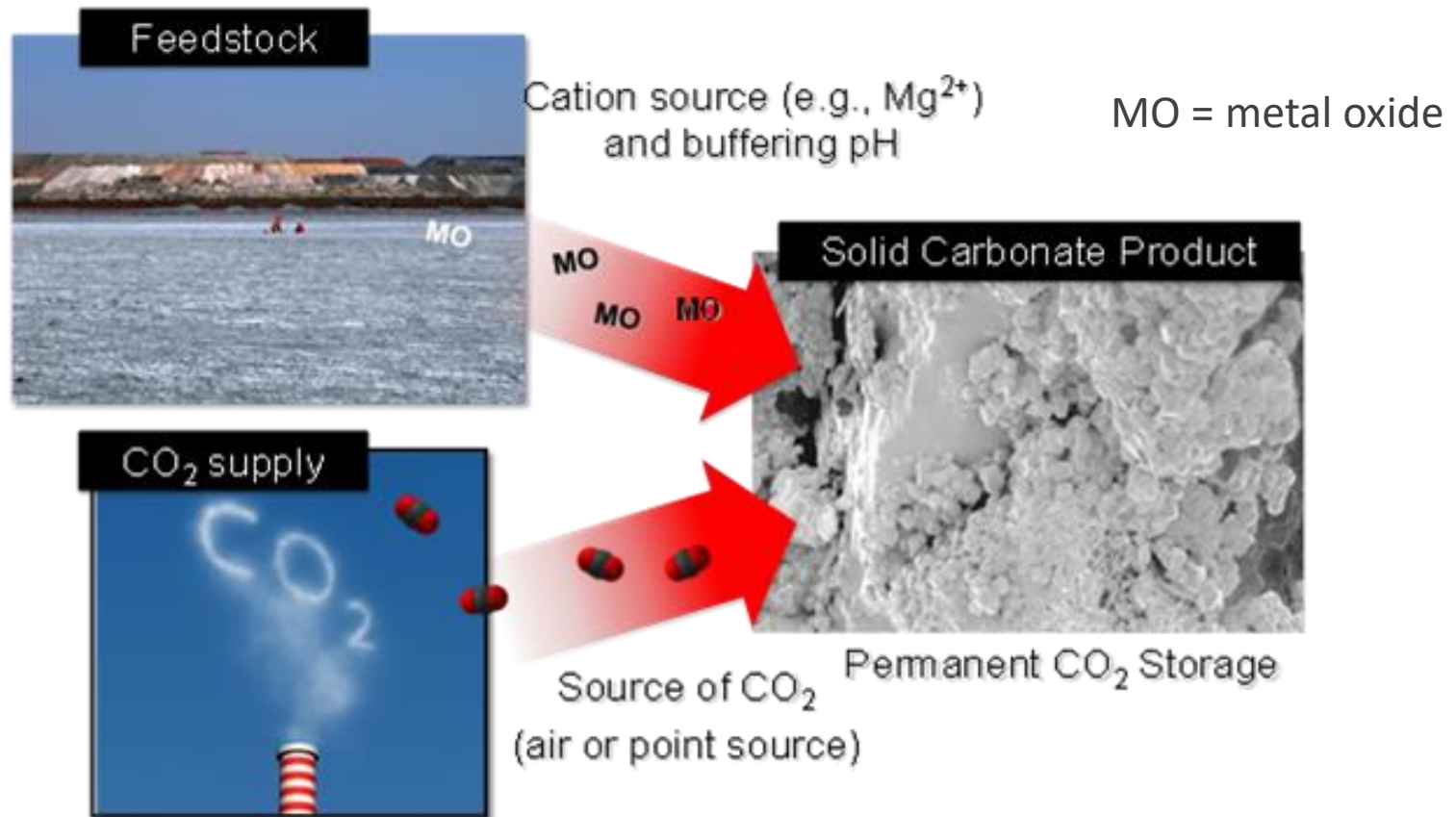
---

- Carbonation reaction and its applications
  - Feedstocks
  - Process routes and operating conditions
  - Products
  - Effects of the process in terms of product properties and CO<sub>2</sub> uptake
- 
- leaching behavior





*M: Calcium, Magnesium (Iron)*

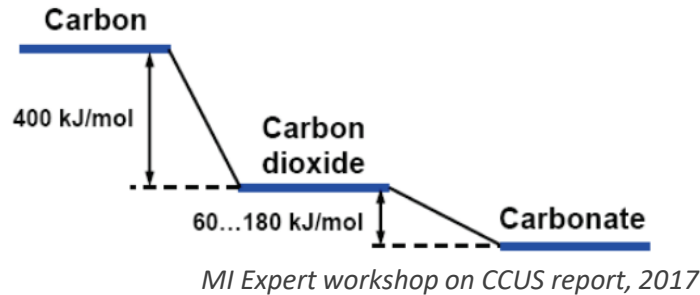


*MI Expert workshop on CCUS report, 2017*



# Carbon capture, storage and utilization: CO<sub>2</sub> conversion to solid carbonates

## Specific traits of carbonation as a CCUS strategy

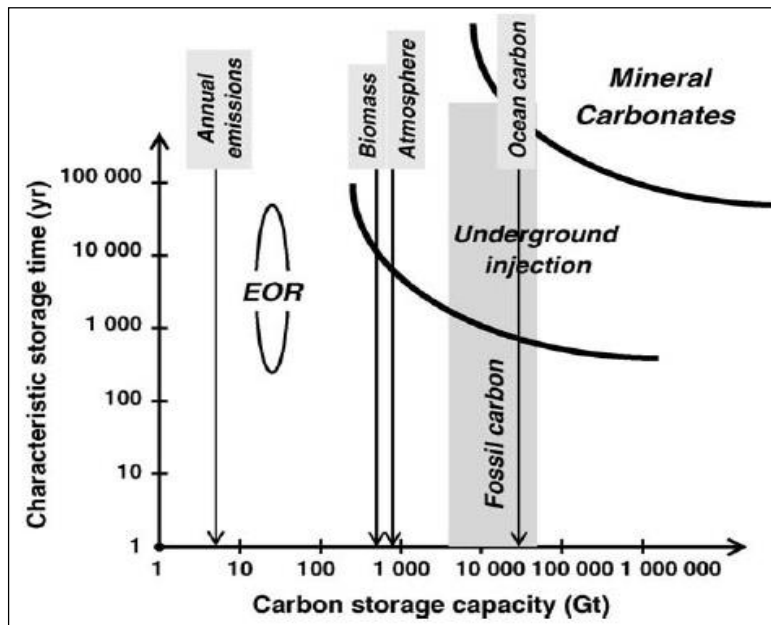


Compared to other CO<sub>2</sub> utilization strategies:

*Exothermic reaction*

Compared to other CO<sub>2</sub> storage strategies:

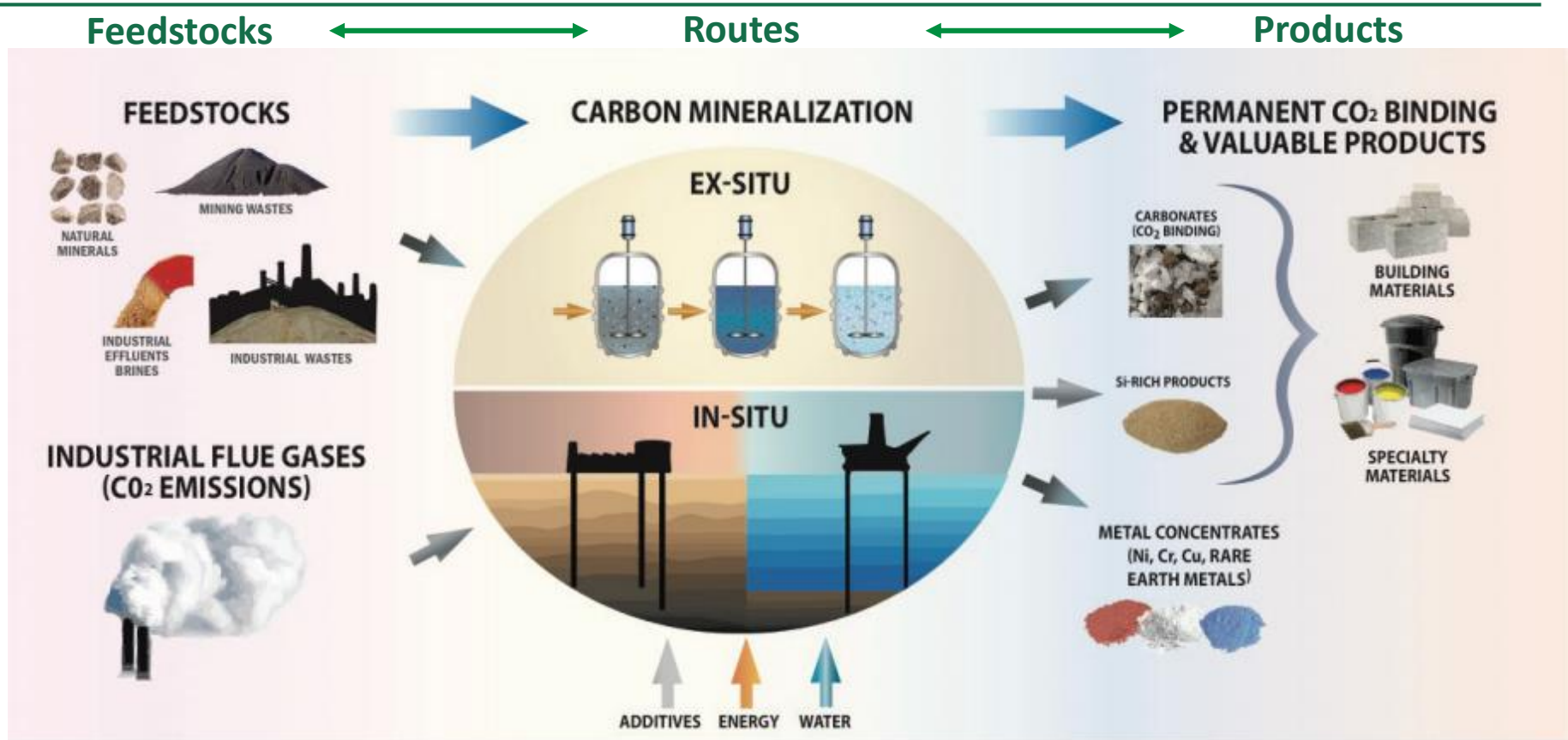
- *Permanent storage and no need for monitoring*
- *Large capacity (considering minerals)*
- *Possibility of employing waste materials or industrial by-products as feedstocks*
- *Possibility of obtaining a product (and co-products) to use for different applications*



*Zevenhoven et al, 2006*



# Applications of carbon mineralization



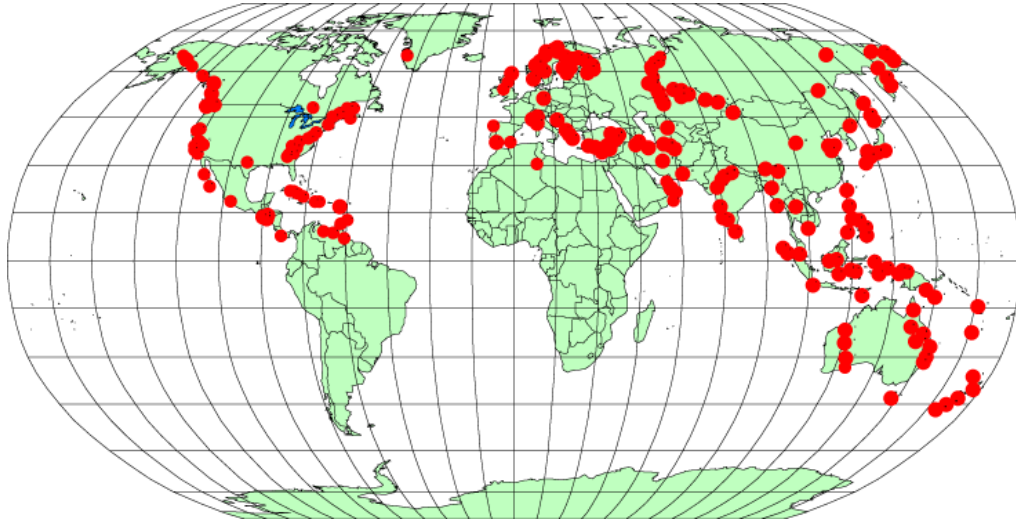
MI Expert workshop on CCUS report, 2017

- **Ex situ engineered processes (minerals & industrial residues)**
- In-situ passive weathering (mine tailings)
- Ex situ treatment or curing step (cements/concrete)



## Feedstocks for carbon mineralization

- *Ultramaphic rocks : ophiolite belts - Mg silicates*



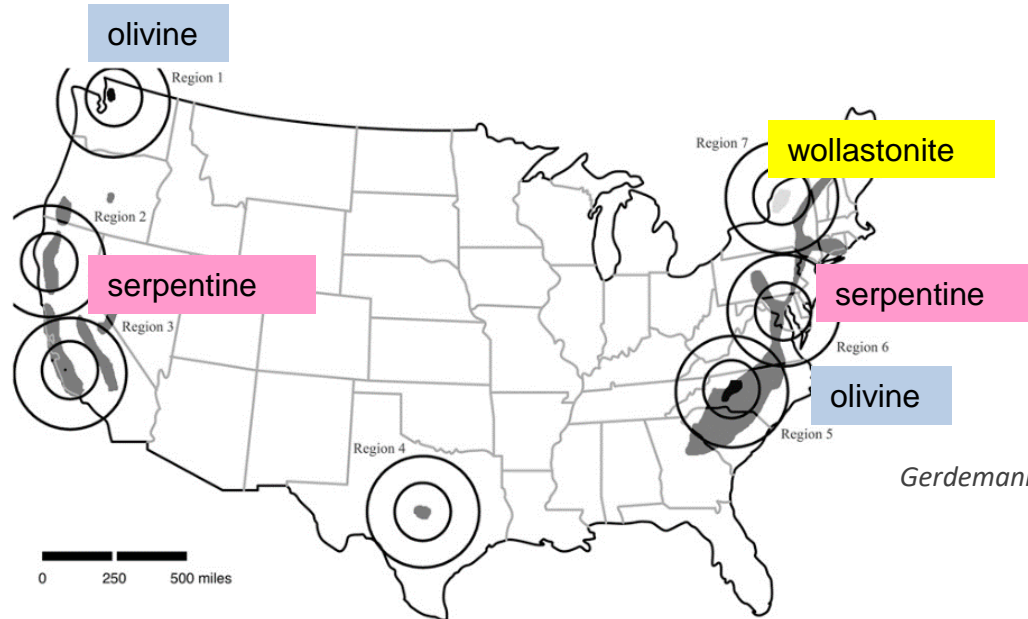
Stoichiometric ratio  
between tons of rock  
needed to store 1 ton of CO<sub>2</sub>

Rock Type		MgO, wt%	CaO, wt%	$R_C$	$R_{CO_2}$
Peridotites	Dunite	49.5	0.3	6.8	1.8
	Harzburgite	45.4	0.7	7.3	2.0
	Lherzolite	28.1	7.3	10.1	2.7
Serpentinite		~40	~0	~8.4	~2.3
Gabbro		~10	~13	~17	~4.7
Basalt	Continental tholeiite	6.2	9.4	26	7.1



# Feedstocks for carbon mineralization

- Silicate minerals*

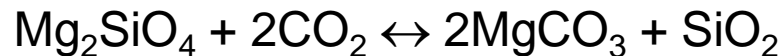


region	ore mineral and grade
1	olivine, 100%
2–4	serpentine (lizardite), 100%
5	olivine, 70%
6	serpentine (antigorite), 100%
7	wollastonite, 50%

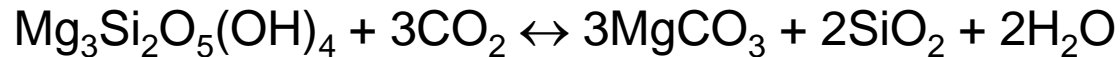
*Gerdemann et al, 2007 Albany Research Center*

Reserves estimated to exceed 2 Gton just in Norway, production 8 Mtons/y

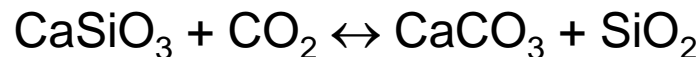
Olivine:



Serpentine:



Wollastonite:



Reserves estimated to exceed 9700 Gton just in the US

World reserves >100 Mton (China, India, US, Finland), production 700 000 tons/y





## Feedstocks for carbon mineralization

---

- *Alkaline industrial residues*

- **Steel industry**

- ✓ Slag from converter units
- ✓ Slag from steel refining treatments
- ✓ Slag from stainless steel production

- **Energy sector**

- ✓ Pulverized fly ash
- ✓ Lignite fly ash
- ✓ Slag from thermal treatment processes
- ✓ Air pollution control residues
- ✓ Fly ash

- **Cement industry**

- ✓ Cement kiln dust
- ✓ Waste cement



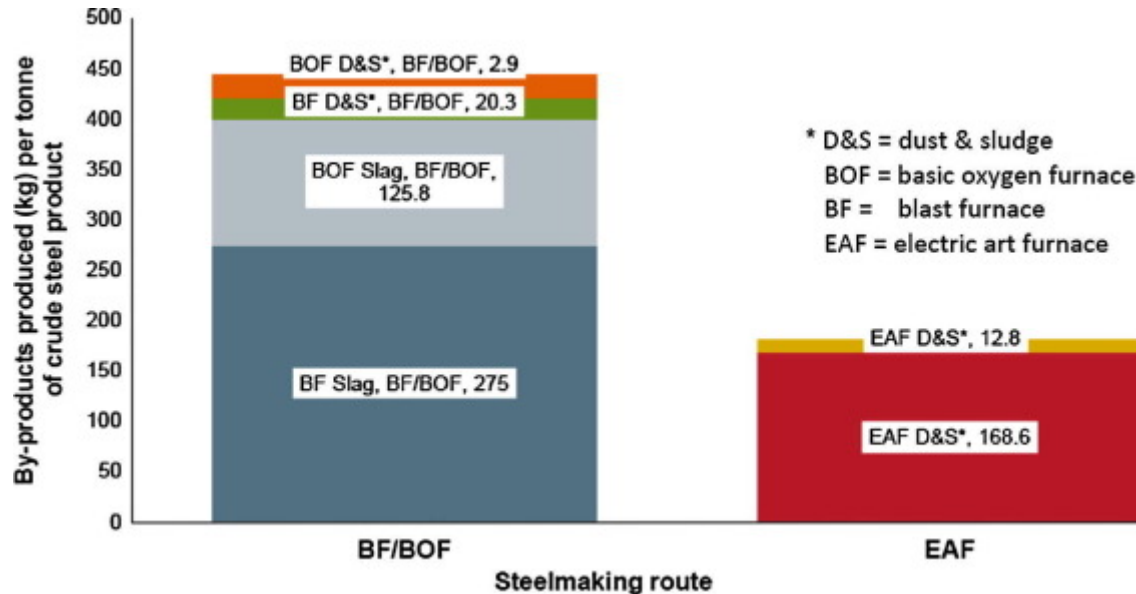
Total maximum sequestration potential  $\approx 100\text{s Mt CO}_2/\text{y}$





## Feedstocks for carbon mineralization

- Steel slag production data (2014)

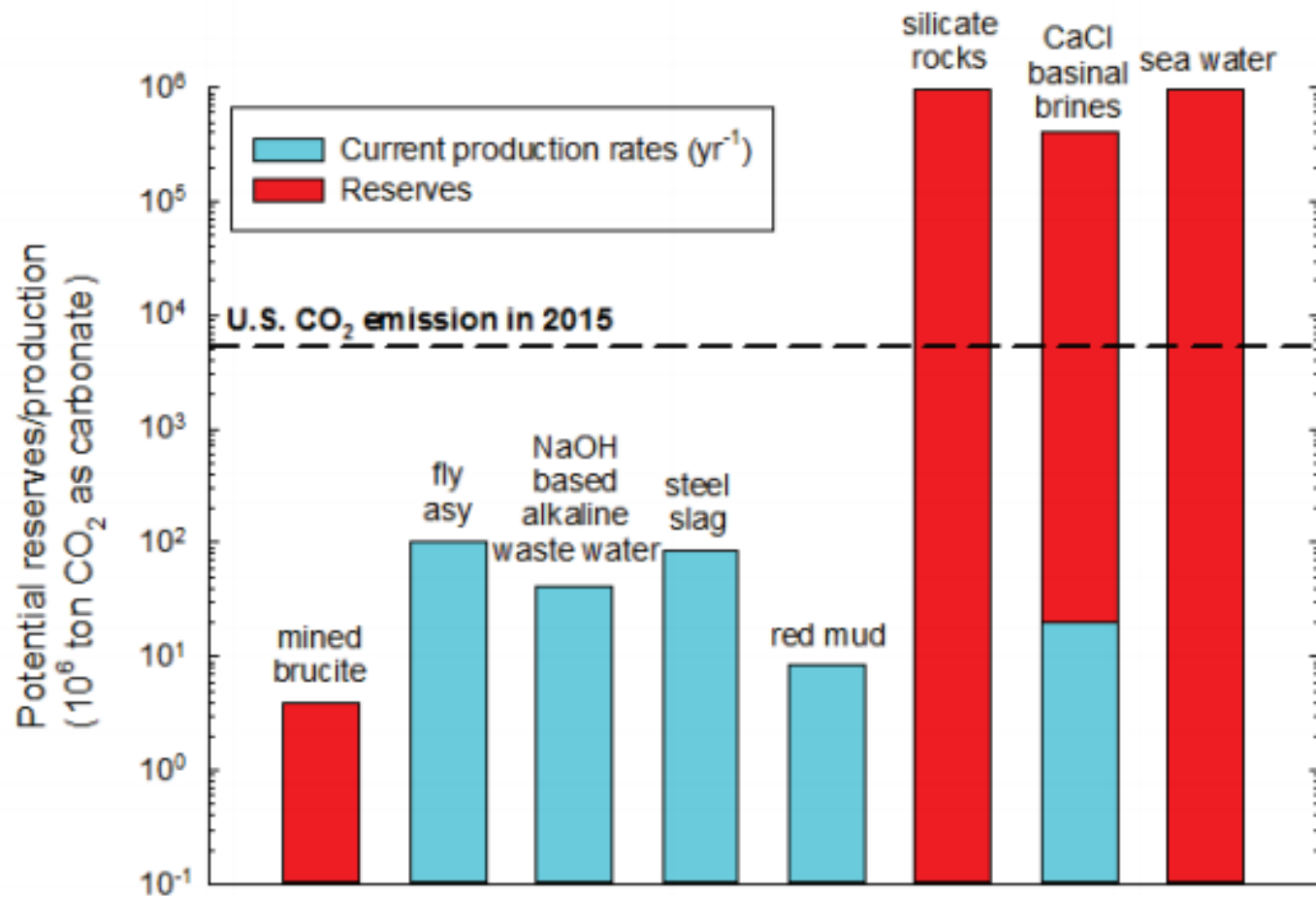


On average the production of one ton of steel results in two tons of CO<sub>2</sub> emissions and 400 kg of blast and basic oxygen furnace slag, and 200 kg of electric arc furnace slag. Globally, more than 400 Mt of steel slag are produced annually.

*Said et al, 2016*



# Feedstocks for carbon mineralization



MI Expert workshop on CCUS report, 2017



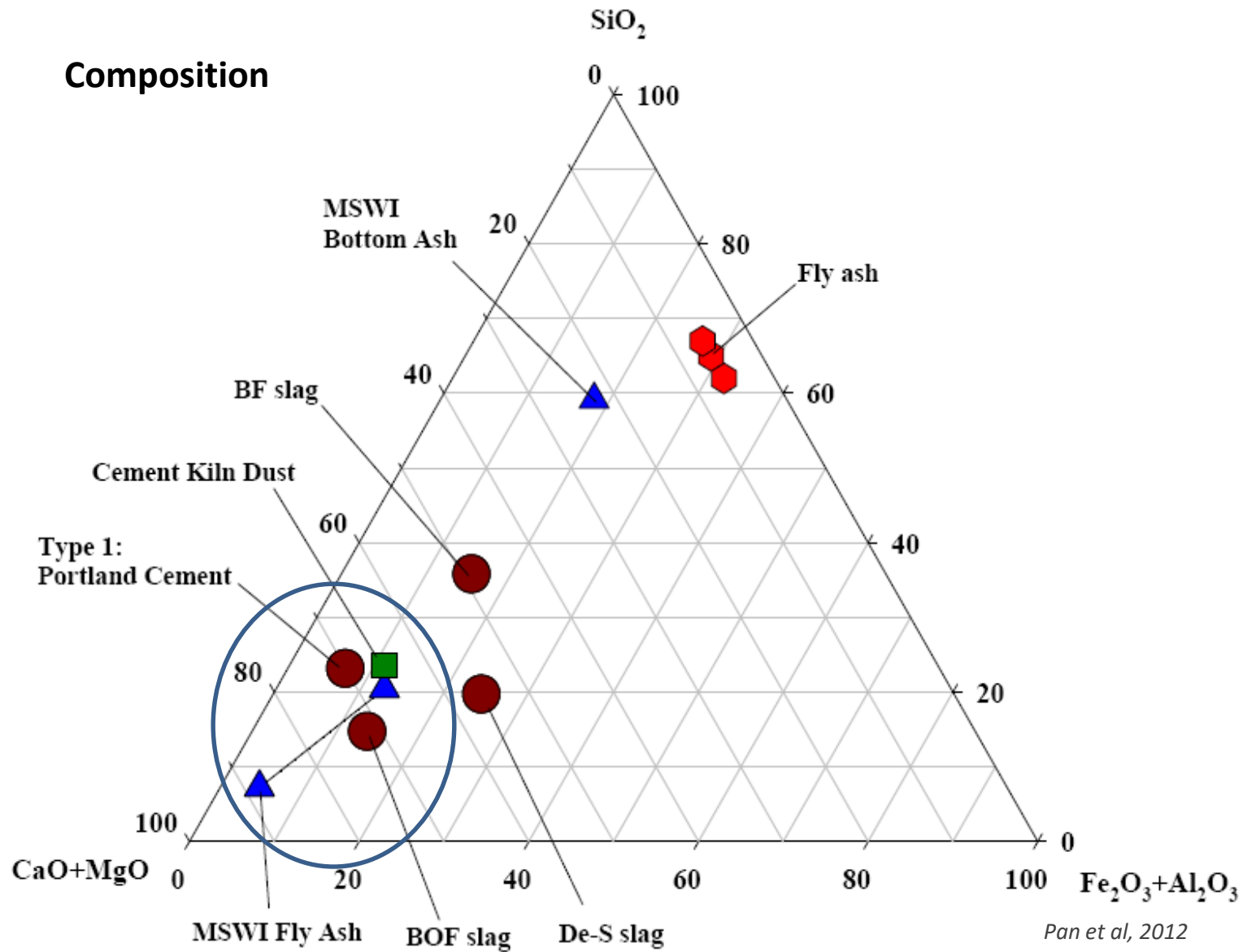
## Feedstocks for carbon mineralization: residues versus minerals

---

- ✓ Often associated with CO<sub>2</sub> point source emissions and under used (or landfilled)
- ✓ No need for mining or quarrying
- ✓ Tend to be more unstable than geologically derived materials
- ✓ Require a lower degree of pre-treatment and less energy intensive conditions
- ✓ The product may present improved environmental and technical properties compared to the untreated residues
- ✗ Niche application with regard to CO<sub>2</sub> storage
- ✗ Composition and mineralogy may vary as a function of the industrial process



# Feedstocks for carbon mineralization: residues



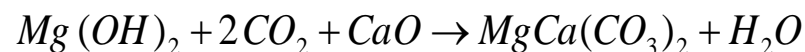
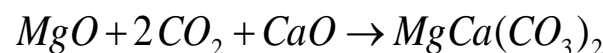
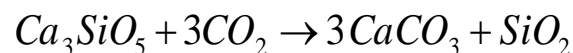
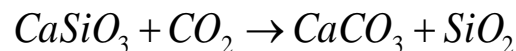
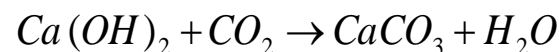
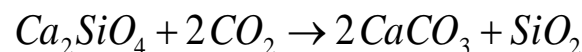
# Feedstocks for carbon mineralization: residues

## Mineralogy

	AOD1	EAF1	APC1	BOF1	BOF2	BOF3
Ca <sub>2</sub> SiO <sub>4</sub>	92.6%	75.5%	---	30.9%	---	12.1%
MgO	2.7%	4.2%	---	---	---	7.9%
CaCO <sub>3</sub>	4.6%	3.6%	10.8%	5.9%	7.7%	4.0%
Fe <sub>3</sub> O <sub>4</sub>	0.1%	4.6%	---	19.6%	---	22.1%
CaF <sub>2</sub>	0.0%	0.0%	---	---	---	---
SiO <sub>2</sub>	---	2.9%	---	---	---	---
Ca(OH) <sub>2</sub>	---	---	24.4%	26.6%	---	17.1%
Mg(OH) <sub>2</sub>	---	---	---	17.0%	---	---
CaSiO <sub>3</sub>	---	---	---	---	56.8%	---
FeO	---	---	---	---	35.5%	---
NaCl	---	---	13.8%	---	---	---
CaOHCl	---	---	51.0%	---	---	---
Ca <sub>3</sub> SiO <sub>5</sub>	---	---	---	---	---	32.1%
Cr <sub>2</sub> O <sub>3</sub>	0.1%	5.1%	---	---	---	---
MnO	---	---	---	---	---	4.6%
Al <sub>2</sub> O <sub>3</sub>	---	4.0%	---	---	---	---

Zingaretti et al, 2014

AOD1 and EAF1 Baciocchi et al., *Greenhouse Gases: Sci Technol.* 2012, 4: 312-19,  
BOF1 and BOF3 Baciocchi et al., *Proc Crete 2012*  
BOF 2 Huijgens et al., *Environ. Sci. Technol.*, 2005, 39: 9676-9682  
APC1 Baciocchi et al., *Waste Manage* 2009, 29: 2994-3003



## Carbon mineralization products

---

Depending on the treatment route applied a wide range of products and by-products can be obtained.

Process adaptation or post-processing can be employed to further expand the range of products to include higher-value ones.



*Aggregates (Carbon8)*



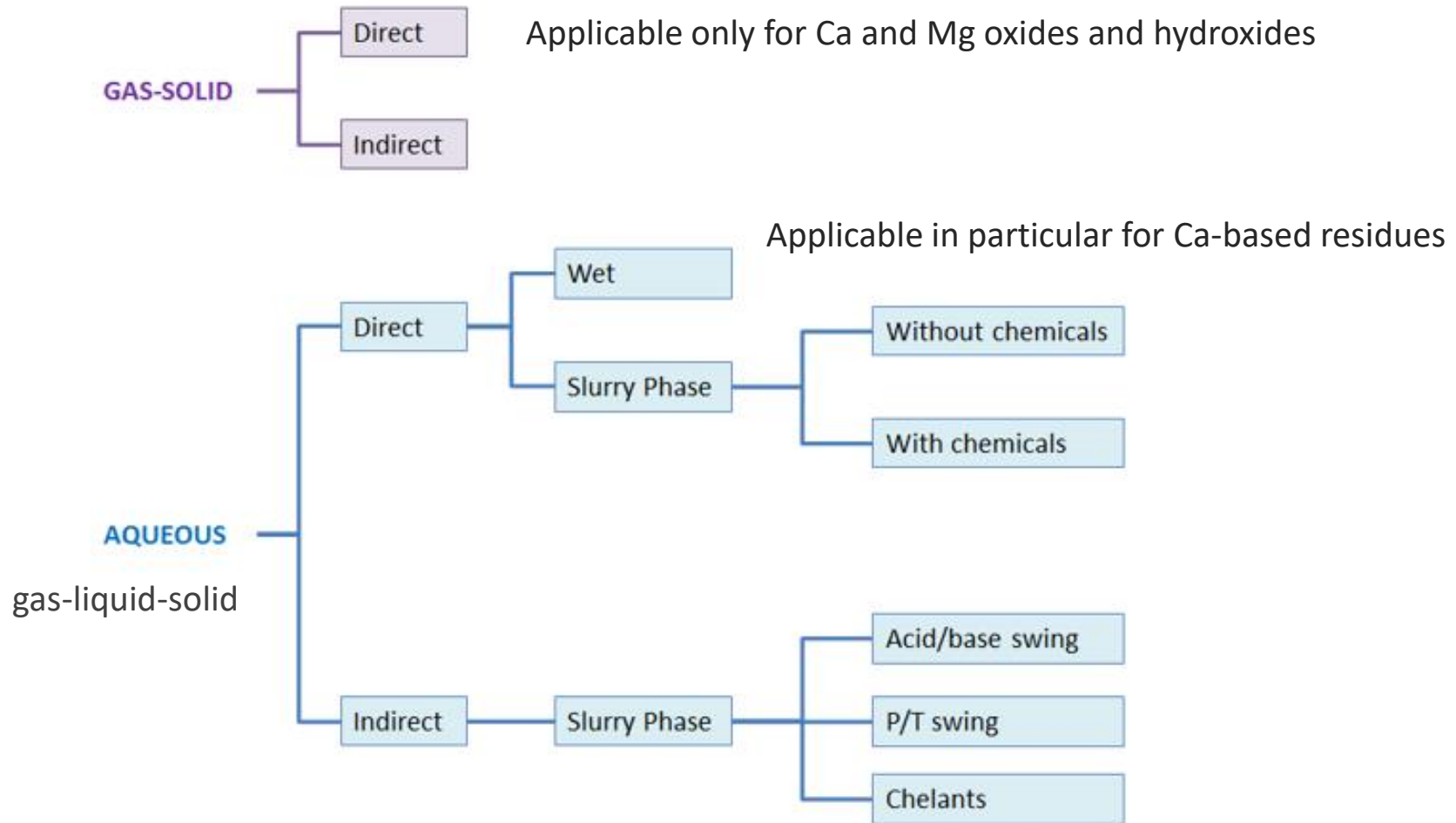
*Blocks (Carbstone-innovation)*



*Precipitated calcium carbonate for paper coating applications as well as mineral fillers for plastics*



# Carbon mineralization process routes



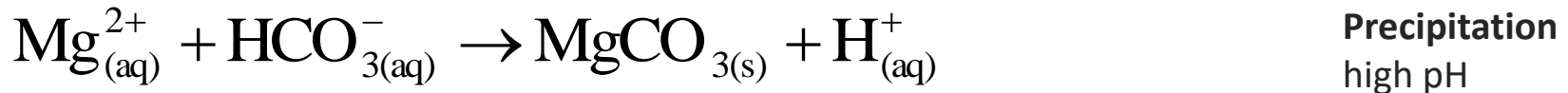
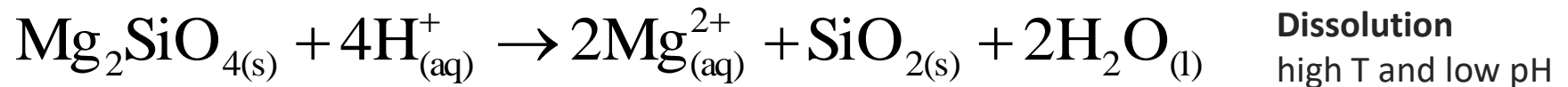
Baciacchi et al, 2014





## Aqueous route: dissolution and precipitation

---



**Single-step:** dissolution + precipitation (in a single reactor)

**Multi-step:** dissolution and precipitation are separately optimized

Two or more steps are needed when dissolution conditions are too far from those required for precipitation or when a high purity product is required

Additives for the direct step: Bicarbonate/Salt mixture ( $\text{NaHCO}_3/\text{NaCl}$ ) proposed to accelerate the reaction

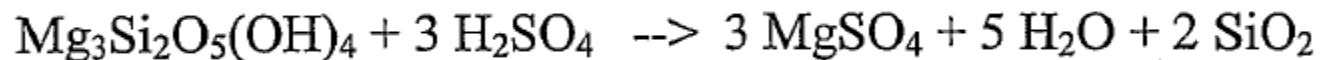


## Example of indirect process routes

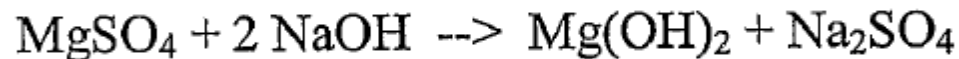
---

**Maroto-Valer et al., (Penn State Research Fundation,US) patents 2004 (WO),2005 (US)**

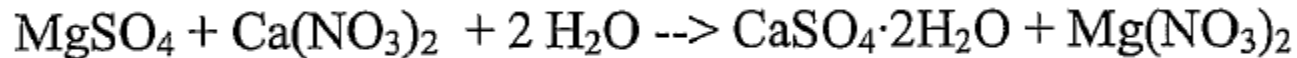
- Aqueous mineral carbonation / Two step process
- Materials: silicate-based Ca/Mg minerals or materials (olivine, serpentine, waste cement)
- Particle size: 70-150  $\mu\text{m}$
- Step 1: chemical activation with an acid:  $T=15 - 75^{\circ}\text{C}$ ; 3-12 hours time



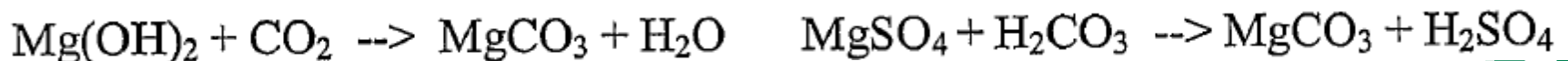
- Step 2a: Use of a base (KOH, NaOH,  $\text{NaHCO}_3$ , Acetates, Formates (pH=7-14)



- Step2b: addition of  $\text{Ca}(\text{NO}_3)_2$



- Step 3: Mg carbonate precipitation and acid recovery (reaction with  $\text{CO}_2$ )



## Example of indirect process routes

### Maroto-Valer et al., (Penn State Research Fundation, US) patents 2004 (WO),2005 (US)

- Step 1: dissolution of the mineral (serpentine)

Treatment	% Mg <sup>2+</sup> in solution	Activation Conditions
Sulfuric Acid	71	25°C, 12 hours
Hydrochloric Acid	21	25°C, 12 hours
Phosphoric Acid	25	25°C, 12 hours
Acetic Acid	48*	60°C, 4 hours*

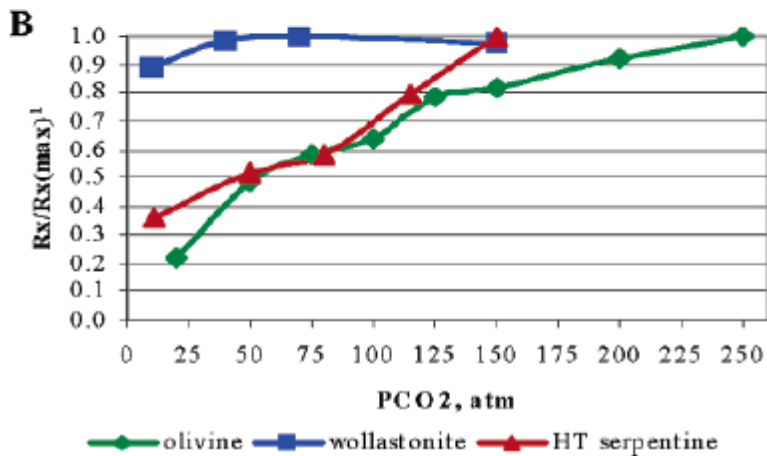
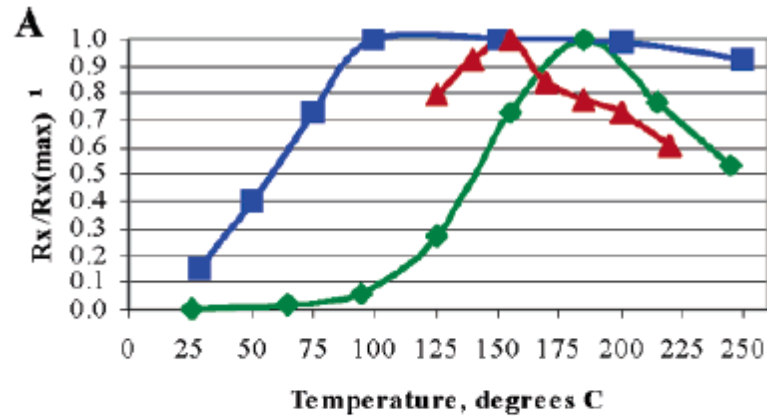
\* Maximum extraction yield reported in the literature using acetic acid (Kakizawa et al., 2001).

- Step 2: conversion of Mg(OH)<sub>2</sub> /MgSO<sub>4</sub> to MgCO<sub>3</sub>

Sample	Conversion	Carbonation Conditions
Parent, untreated	8 %	155°C, 1850 psig, 1 hour
Steam treated	70%	155°C, 1850 psig, 1 hour
Mg(OH) <sub>2</sub>	>55%	20°C, 650 psig, 3 hours
MgSO <sub>4</sub>	54%	20°C, 650 psig, 6 hours
Mg Acetate	30%	20°C, 650 psig, 3 hours



# Single step aqueous carbon mineralization



## Optimum carbonation conditions

mineral	carbonation conditions		
	$T, ^\circ C$	$P_{CO_2}$ , atm	carrier solution
olivine	185	150	0.64 M $NaHCO_3$ , 1 M $NaCl$
wollastonite	100	40	distilled water
HT serpentine	155	115	0.64 M $NaHCO_3$ , 1 M $NaCl$

Samples ground to  $< 75\mu m$

HT: Heat treatment at  $630^\circ C$  for 2 hours

$R_x$  = extent of reaction in 1h

$R_x(\max)$  = maximum extent of reaction in 1h

olivine: 49.5%

wollastonite: 81.8%

HT serpentine: 73.5%

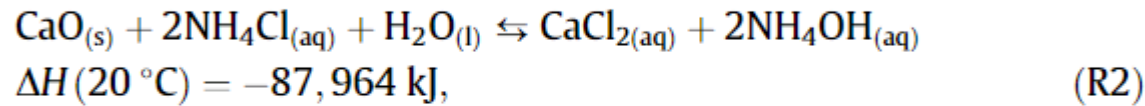
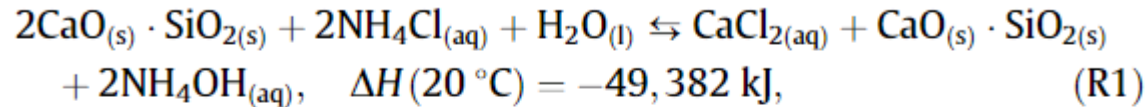
Gerdemann et al. (2007)



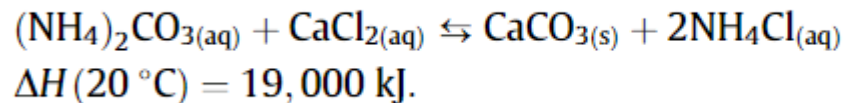
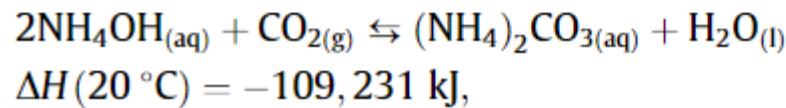
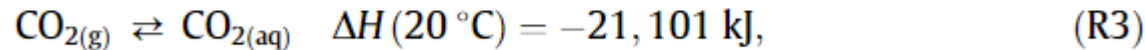
## Example of an indirect process route for residues

Teir et al., (Abo Academy Finland, patent 2013 (WO))

The calcium extraction stage



and the carbonation reactions



Slag 2 PCC

Ammonium salts ( $\text{NH}_4\text{Cl}$ ,  $\text{H}_4\text{NO}_3$  or  $\text{CH}_3\text{COONH}_4$ )  
Dissolution at  $20^\circ\text{C}$

Pilot plant in operation

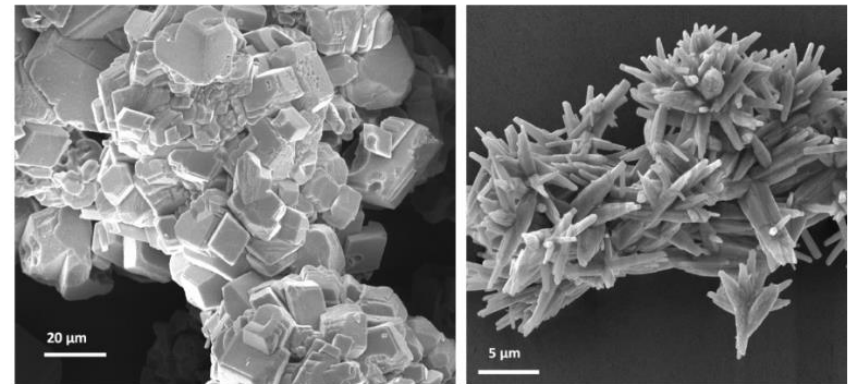


Fig. 13. SEM micrographs of PCC produced from the Slag2PCC pilot plant: (a) Test 1 at  $20^\circ\text{C}$ , rhombohedral calcite and (b) Test 2 at  $45^\circ\text{C}$ , aragonite.

Carbonation  $T < 60^\circ\text{C}$  to avoid ammonia emissions

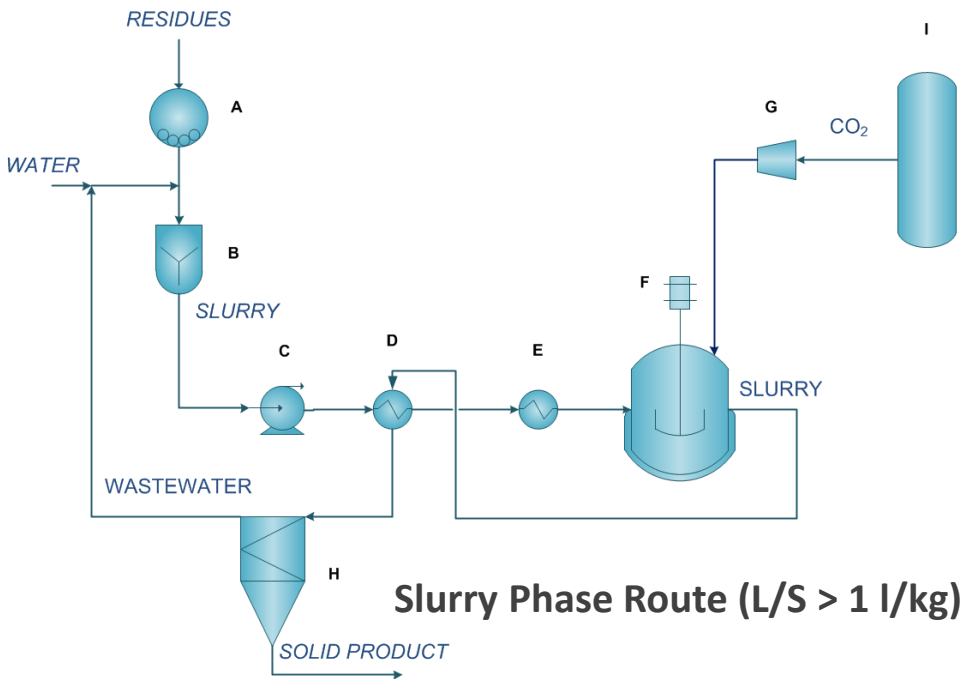
Said et al 2016



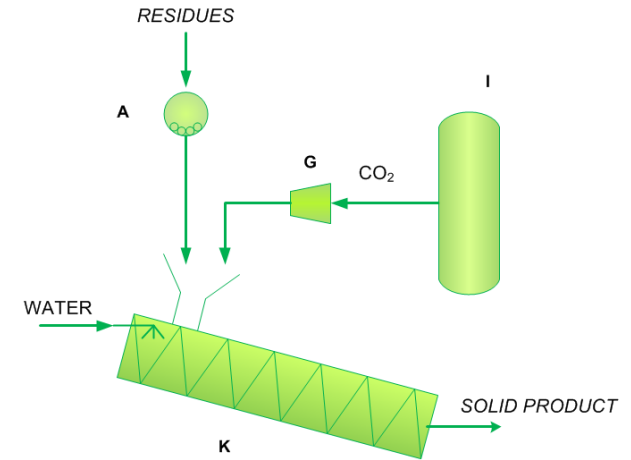
# Direct aqueous carbonation of residues

**SLURRY PHASE ROUTE** ( $L/S > 2 \text{ l/kg}$ ):  $T = 50\text{--}150 \text{ }^\circ\text{C}$  and  $p_{\text{CO}_2} = 1\text{--}20 \text{ bar}$

**WET ROUTE** ( $L/S < 1 \text{ l/kg}$ ):  $T = 20\text{--}50 \text{ }^\circ\text{C}$  and  $p_{\text{CO}_2} = 1\text{--}10 \text{ bar}$



**Slurry Phase Route ( $L/S > 1 \text{ l/kg}$ )**



**Wet route ( $L/S < 1 \text{ l/kg}$ )**

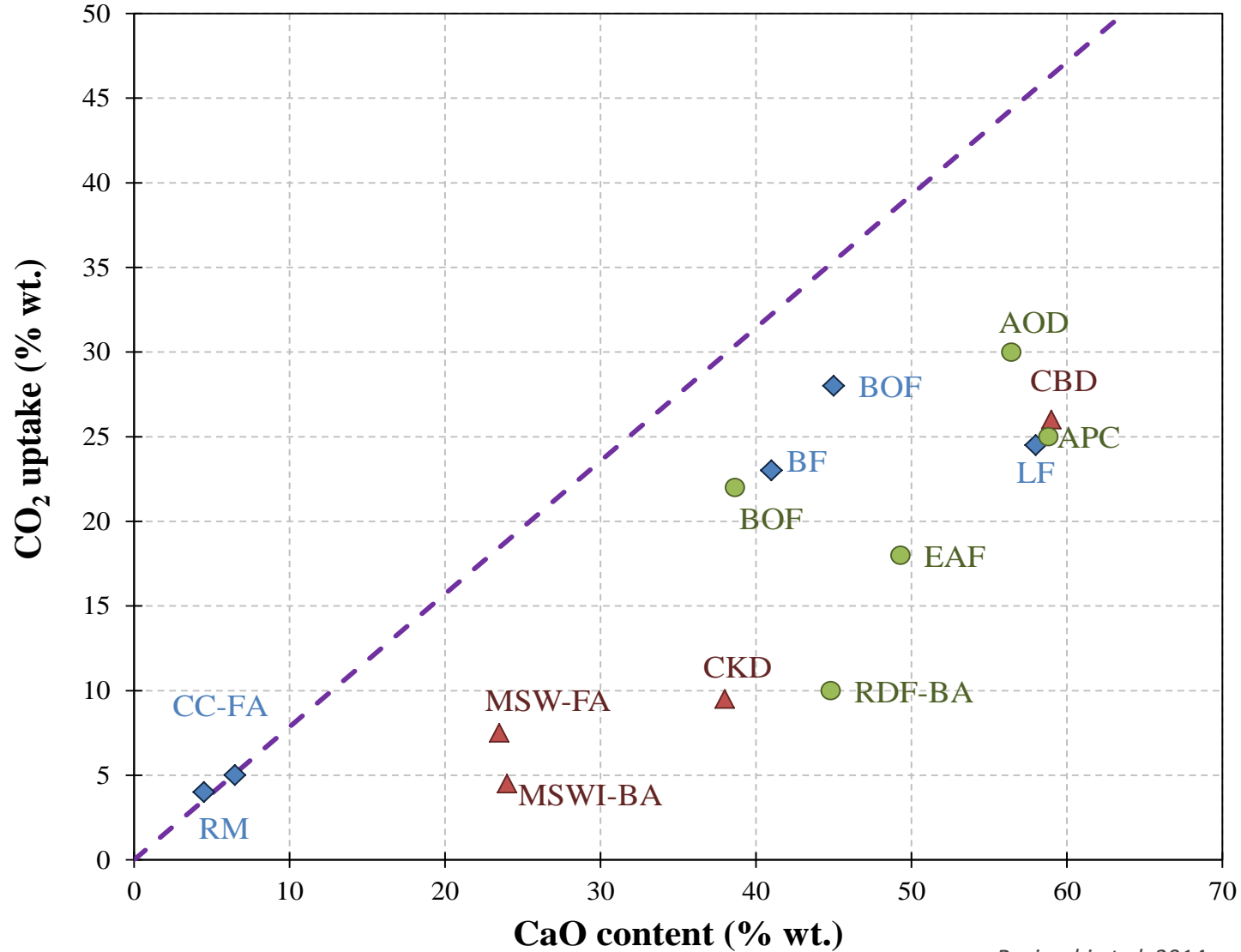
*Zingaretti et al, 2014*

- A Mill
- B Mixer
- C Slurry Pump
- D Heat Exchanger
- E Heater

- F Carbonation Reactor
- G Compressor
- H Clarifier
- I CO<sub>2</sub> Absorber
- K Rotary Drum Reactor



# CO<sub>2</sub> uptake as a function of waste composition



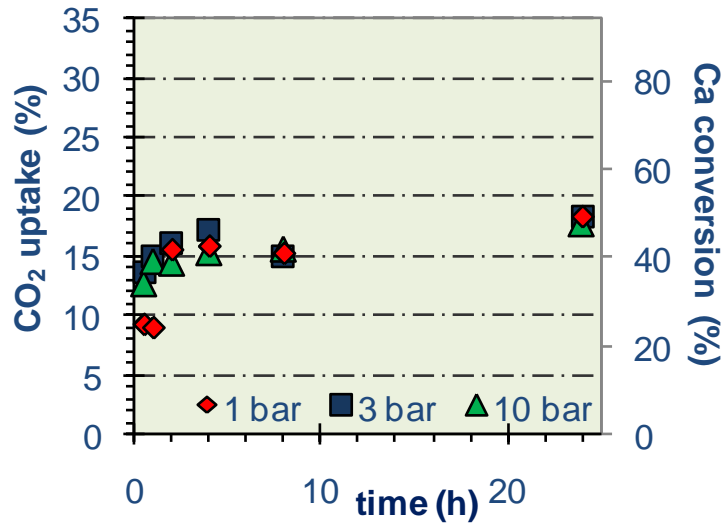
Bacocchi et al, 2014



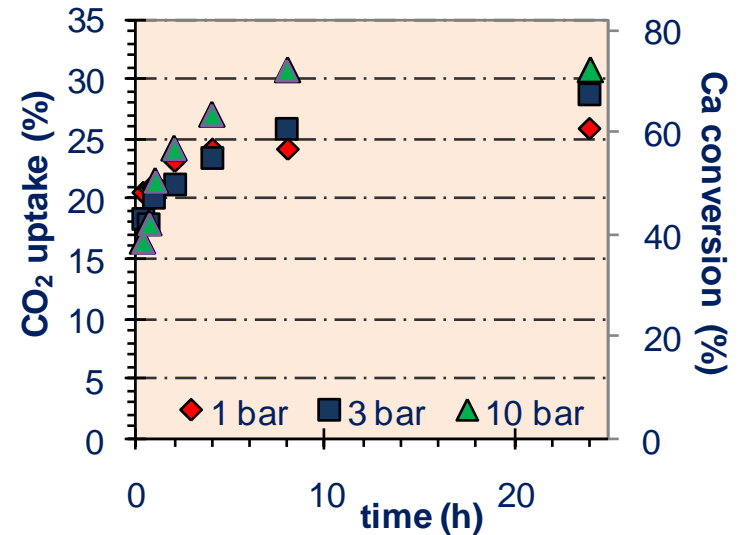


# CO<sub>2</sub> uptake as a function of waste mineralogy

EAF slag



AOD (ladle) slag



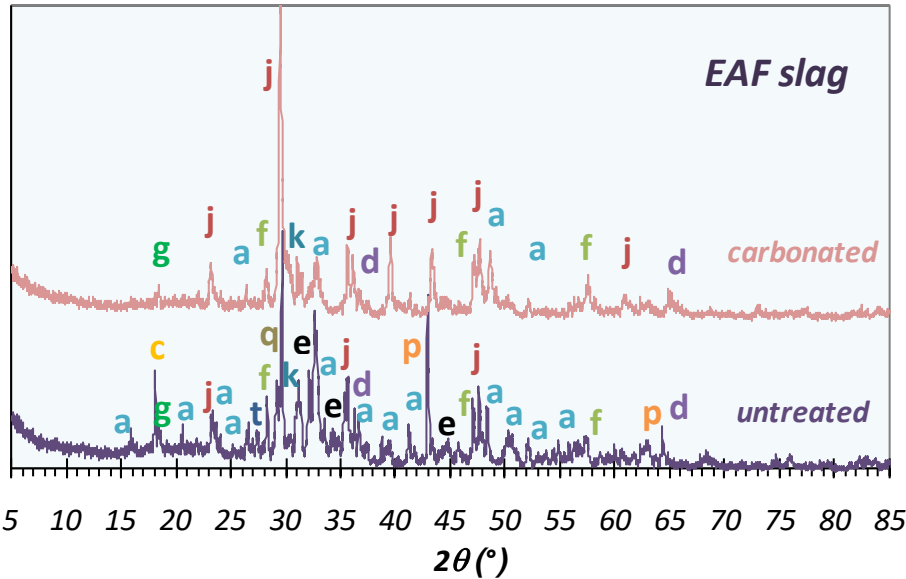
Baciacchi et al, 2012

For residues with high contents of Ca(OH)<sub>2</sub> (e.g. APC residues), Ca conversions of over 60% were found for  $t < 30$  min at ambient T and  $p\text{CO}_2 = 1$  bar for L/S ratios of 0.2 l/kg

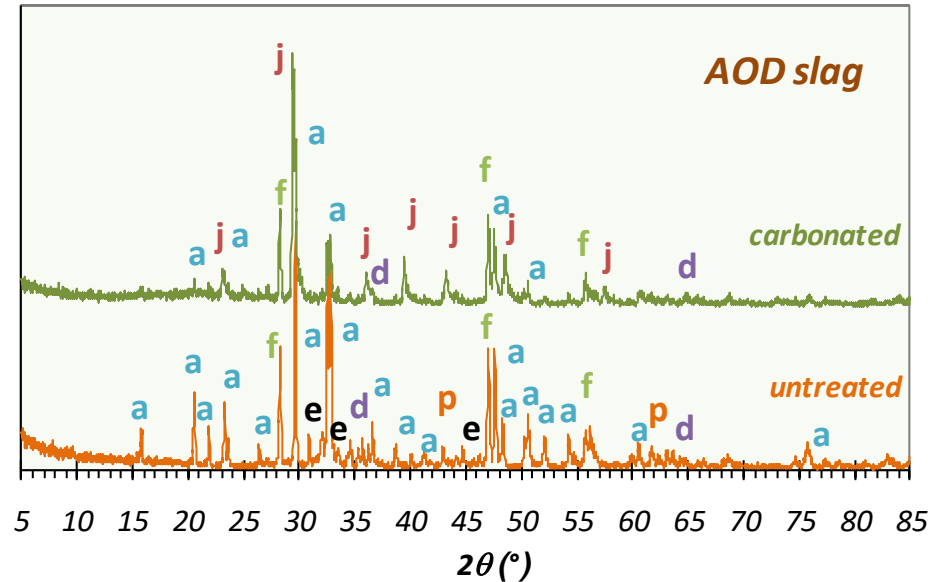


# CO<sub>2</sub> uptake as a function of waste mineralogy

Carbonation conditions: 4 h and 10 bar



Carbonation conditions: 2 h and 1 bar



a) dicalcium silicate ( $\text{Ca}_2\text{SiO}_4$ )

c) Ca-Al oxide ( $\text{Ca}_{12}\text{Al}_{14}\text{O}_{33}$ )

d) magnetite ( $\text{Fe}_3\text{O}_4$ )

e) chromium oxide ( $\text{CrO}$ )

f) calcium fluoride ( $\text{CaF}_2$ )

g) Cr-Mg oxide ( $\text{MgCr}_2\text{O}_4$ )

j) calcite ( $\text{CaCO}_3$ )

k) gehlenite ( $\text{Ca}_2\text{Al}_2\text{SiO}_7$ ) or akermanite ( $\text{Ca}_2\text{MgSi}_2\text{O}_7$ )

p) periclase ( $\text{MgO}$ )

q) cuspidine ( $\text{Ca}_4\text{Si}_2\text{O}_7(\text{F},\text{OH})$ )

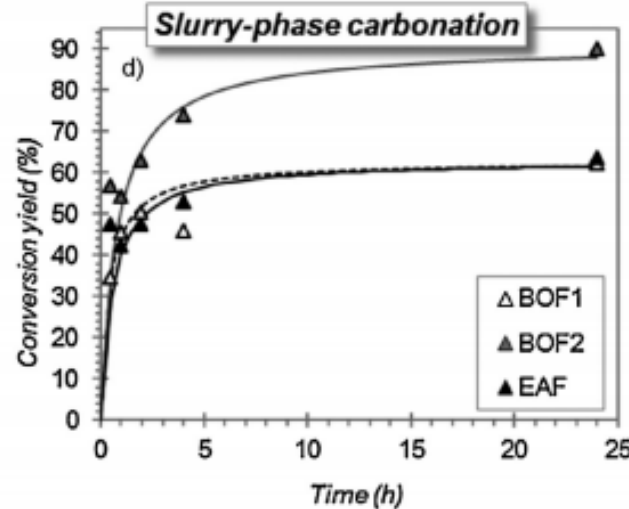
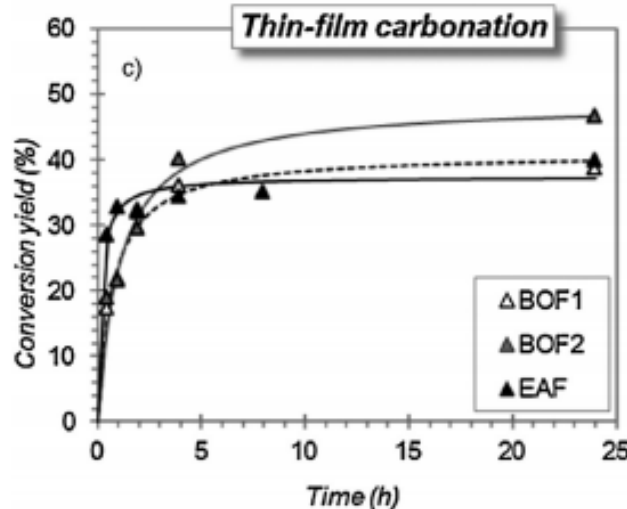
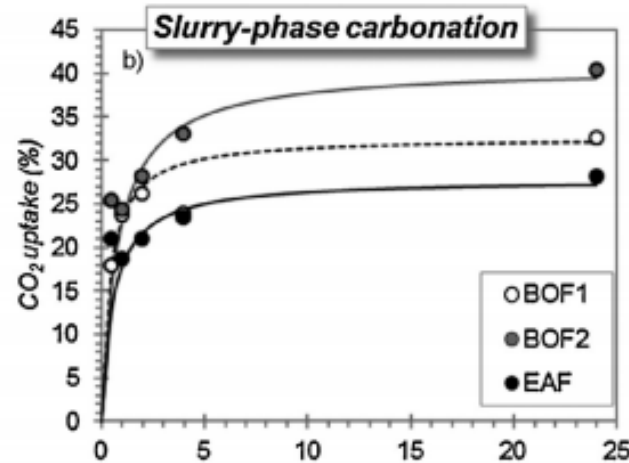
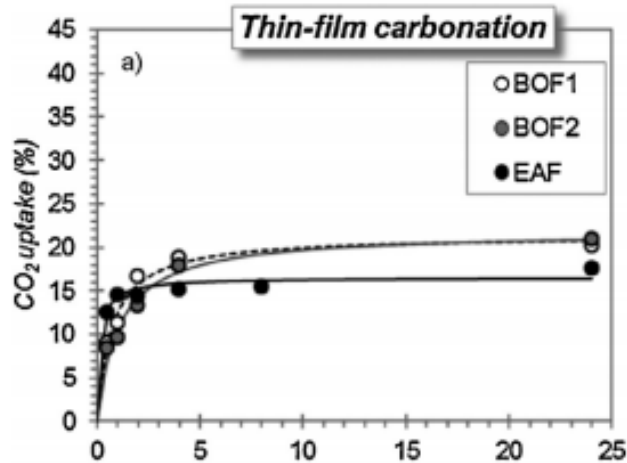
t) quartz ( $\text{SiO}_2$ )

Bacocchi et al, 2012



# CO<sub>2</sub> uptake as a function of process route and operating conditions

50 °C  
10 bar  
L/S=0.3 l/kg  
100% CO<sub>2</sub>



100 °C  
10 bar  
L/S=5 l/kg  
100% CO<sub>2</sub>

For the slurry route dolomite was found in the product so Mg was considered in the conversion yield

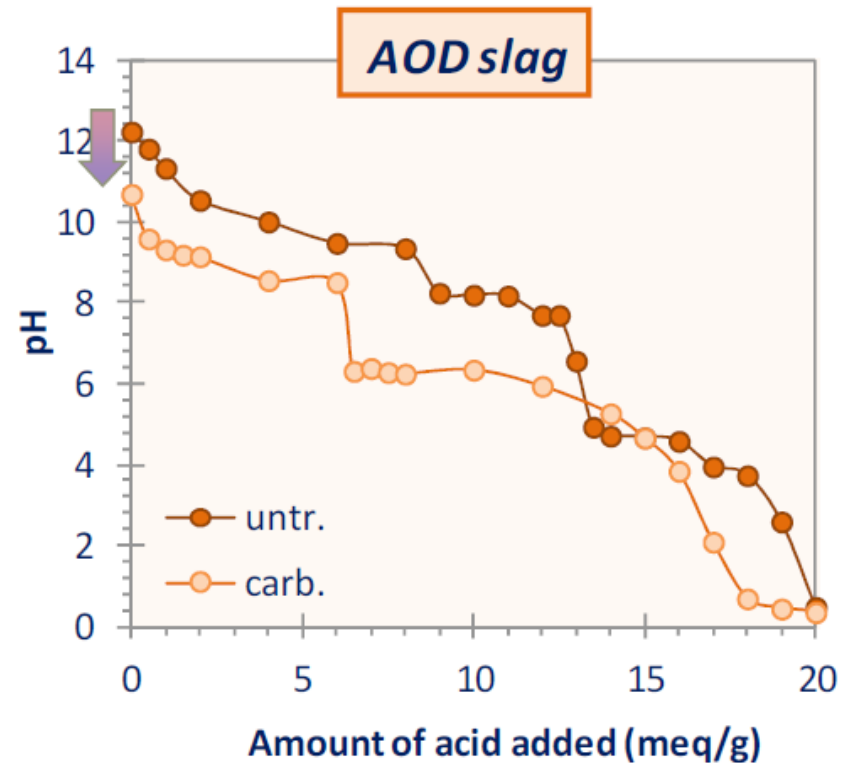
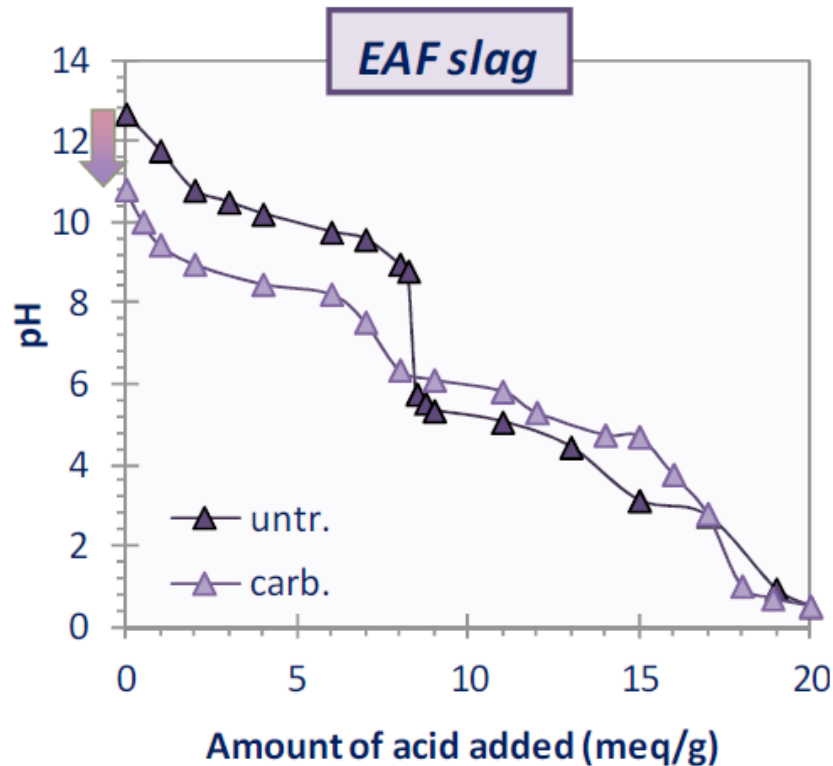
Samples milled to below 150 μm

*Bacocchi et al, 2015*



# Effects on leaching behavior for wet route carbonation

## ➤ Acid neutralization capacity

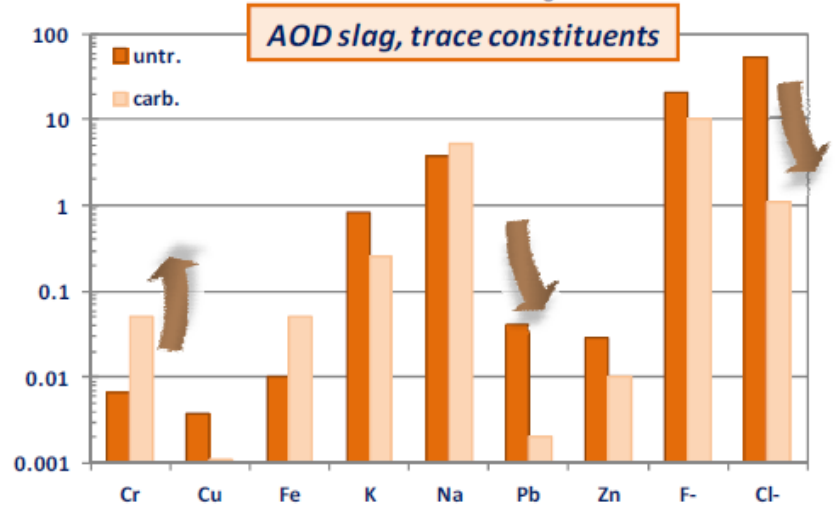
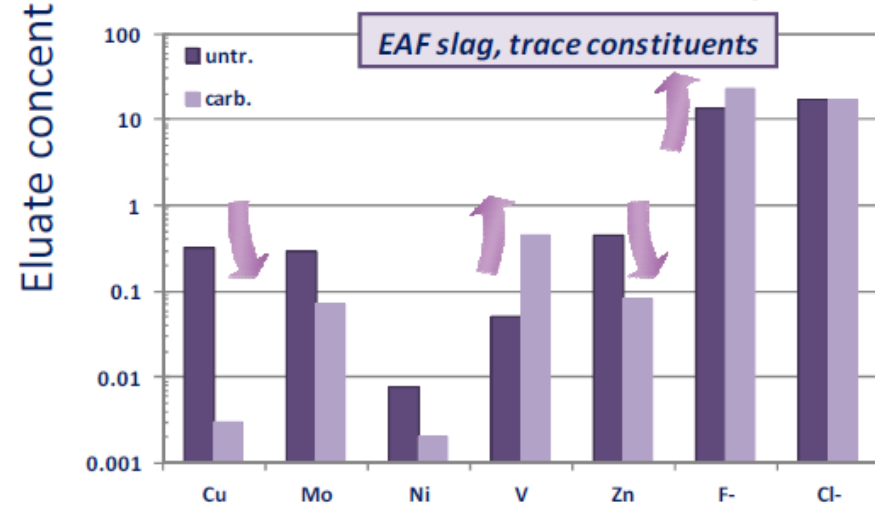
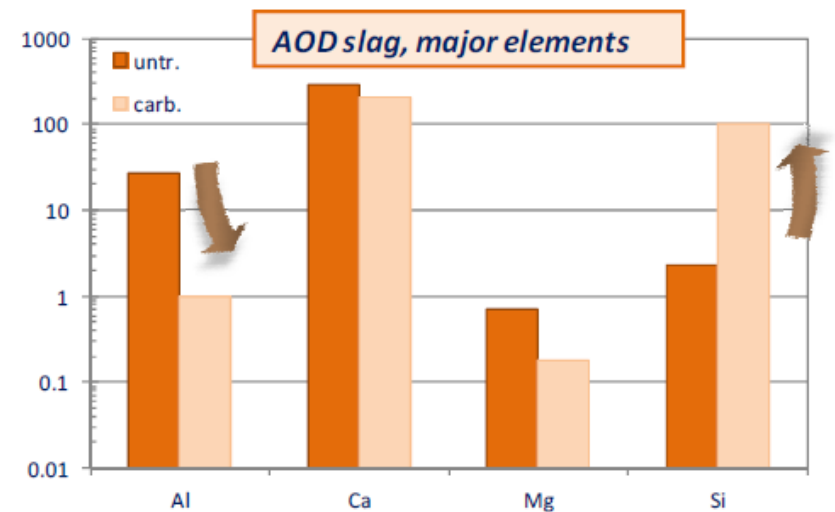
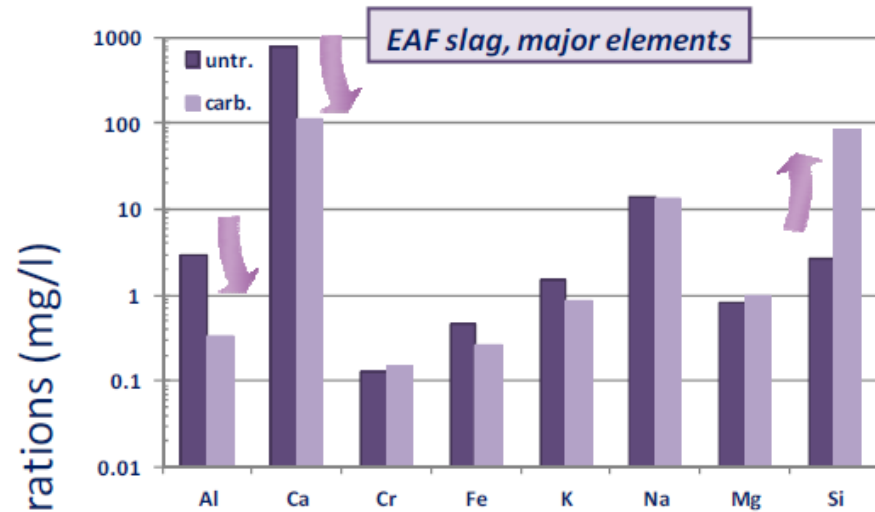


Carbonation conditions: 50 °C, L/S=0.4 l/kg, 100% CO<sub>2</sub>

Baciacchi et al, 2012



# Effects on leaching behavior for wet route carbonation



pH: 12.6 → 10.8

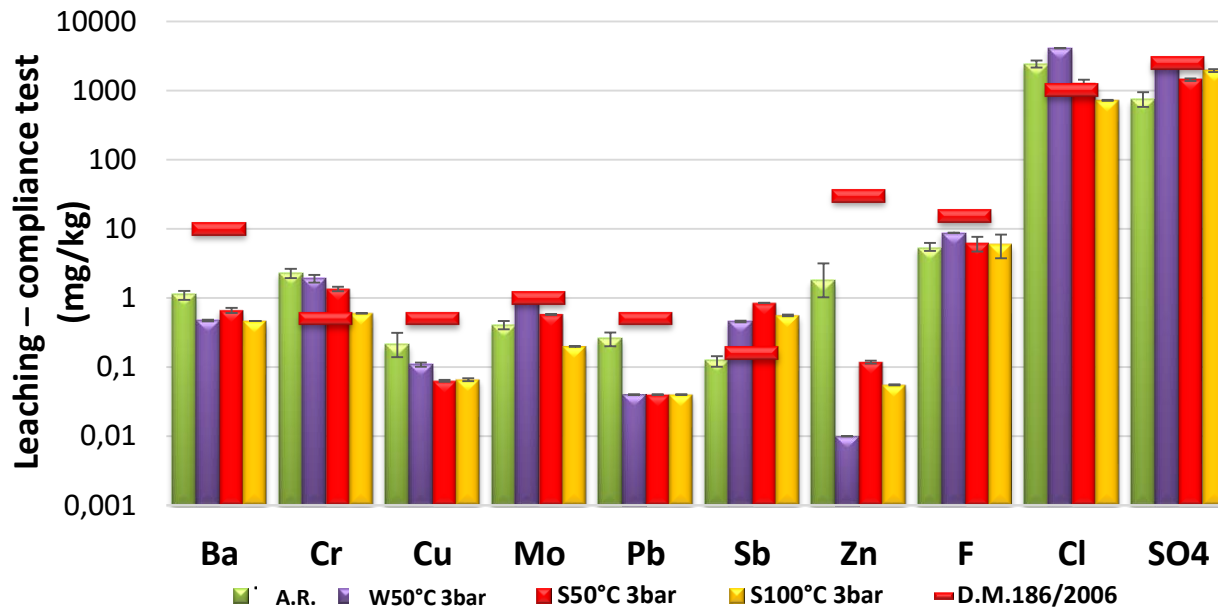
Bacocchi et al, 2012

pH: 12.5 → 10.5



# Effects on leaching behavior: wet vs. slurry route carbonation

## Tests performed on a sample of BA from dry waste incineration



Slurry carbonation at 100 °C

✓ pH, Cr and Cl

✗ Sb

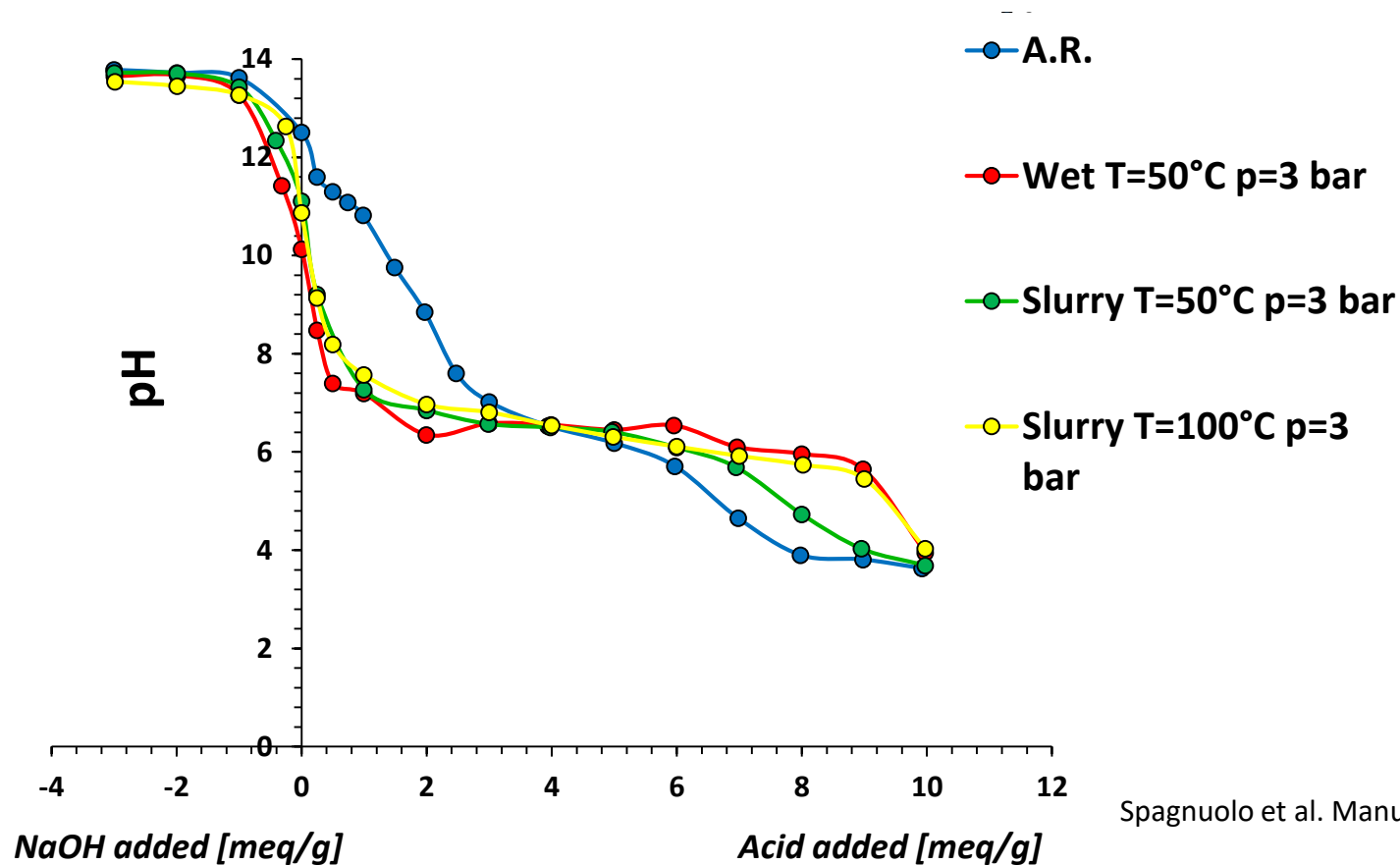
	pH	CO <sub>2</sub> uptake(%)
A. R.	12.5	-
W50°C 3bar	9.8	11.31
S50°C 3bar	10.4	9.95
S100°C 3bar	10.7	9.38

Spagnuolo et al. Manuscript in preparation



# Effects on leaching behavior: wet vs. slurry route carbonation

Tests performed on a sample of BA from dry waste incineration: ANC behaviour



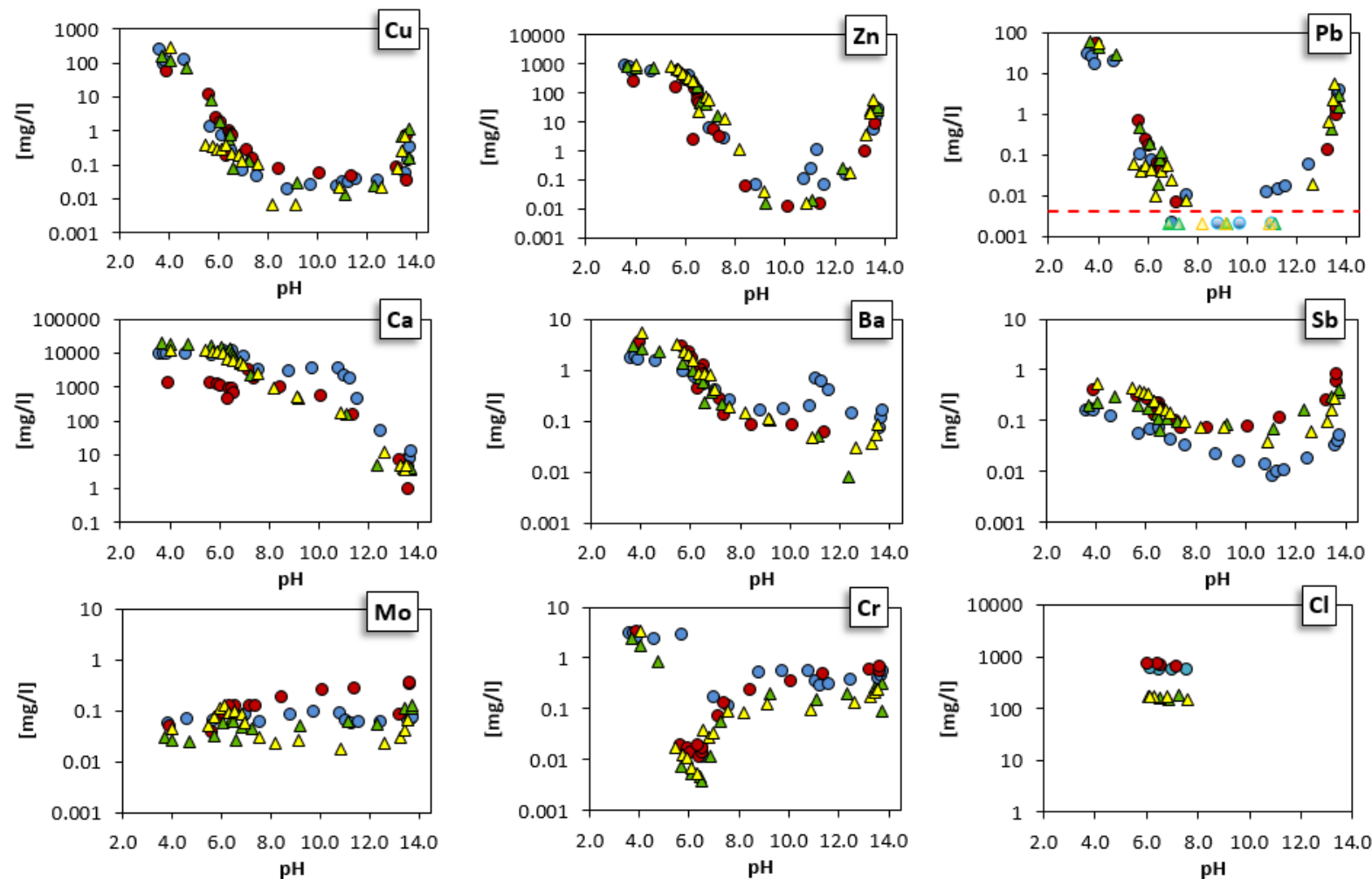
Spagnuolo et al. Manuscript in preparation





# Effects on leaching behavior: wet vs. slurry route carbonation

## Results: Release as a function of pH

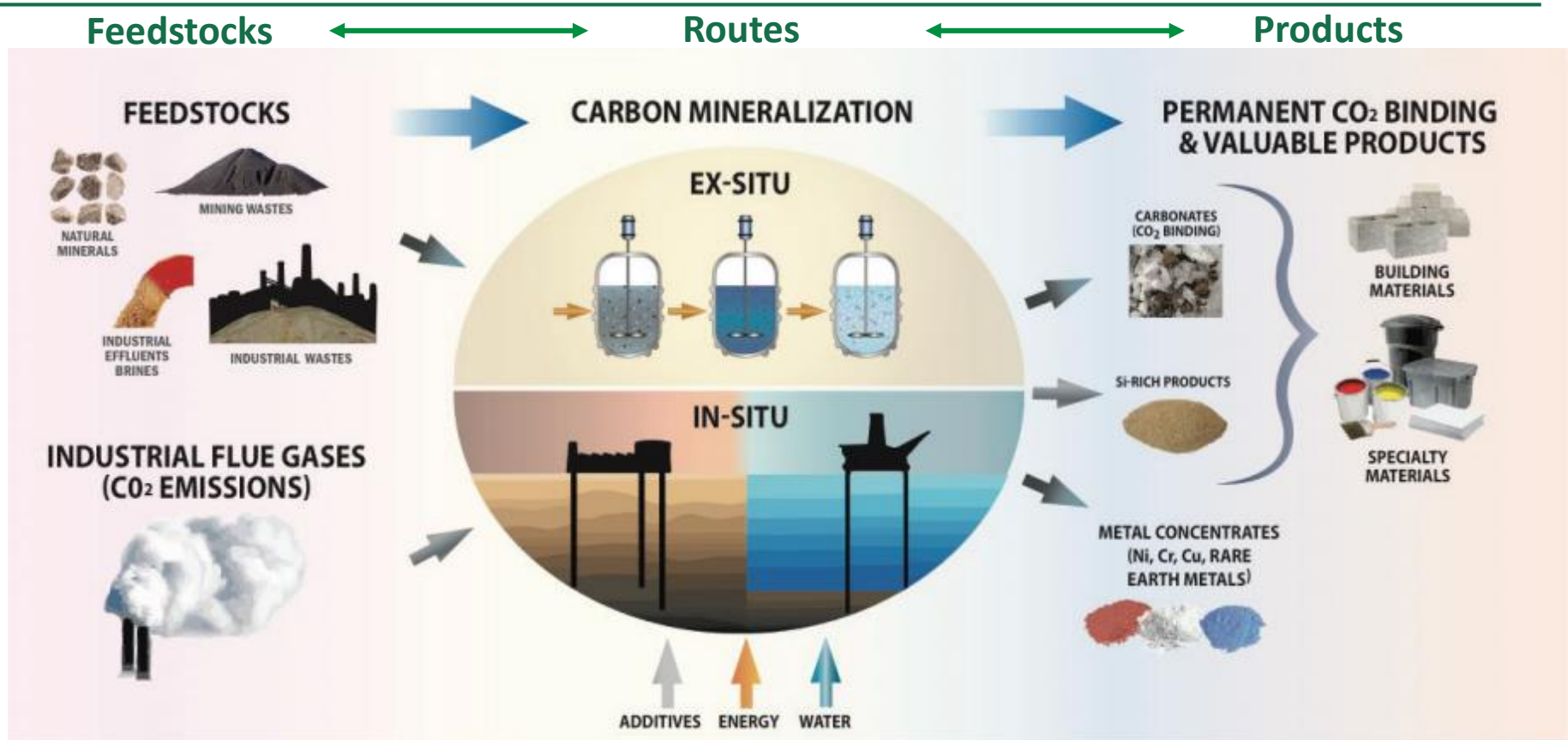


Spagnuolo et al. Manuscript in preparation

● A.R. ● Wet T=50°C, p=3 bar ▲ Slurry T=50°C, p=3 bar ▲ Slurry T=100°C, p=3 bar --- LOQ



# Applications of carbon mineralization

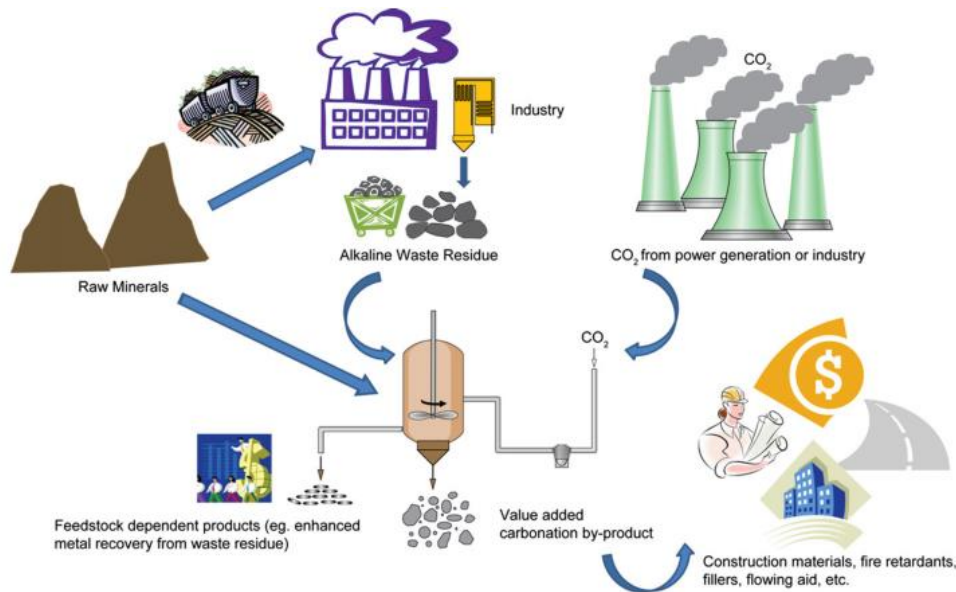


MI Expert workshop on CCUS report, 2017

- **Ex situ engineered processes (minerals & industrial residues)**
- In-situ passive weathering (mine tailings)
- Ex situ treatment or curing step (cements/concrete)



### Ex situ carbonation of minerals (Mg-bearing silicate rocks) and alkaline waste residues

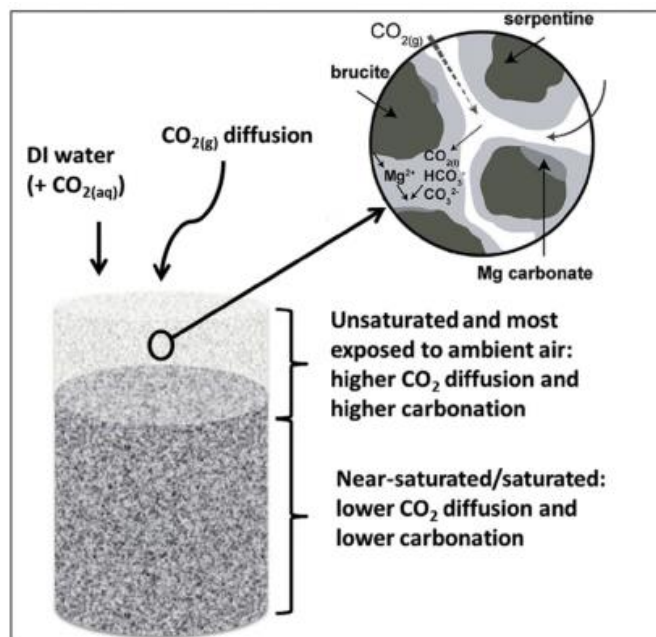


*Bobicki et al. 2011*

- CCS and/or CCU
- Many types of process routes tested
- Multi-step processes for minerals (serpentine)
- Energy-intensive pre-treatment usually required
- Need for recycling additives employed for enhancing dissolution kinetics
- Need to produce a product with specific technical properties for utilisation (e.g. purity, particle size, morphology)
- Issues on effects of treatment on environmental behaviour of waste materials
- Direct use of diluted CO<sub>2</sub> sources to avoid capture would be very important



### Passive mineralization of atmospheric CO<sub>2</sub> in mine waste/tailings



Kandji et al 2017

- Direct air capture or permanent storage (or use) of carbon point sources
- Stabilization of mining waste and improvement of environmental properties (e.g. chrysotile)
- Monitoring uptakes with different methods (e.g. carbon footprinting)
- Need to accelerate CO<sub>2</sub> gas dissolution and hydration processes at ambient temperature to increase uptakes
- Need to engineer the process
- Need to monitor and predict the leaching behaviour of the waste (geochemical modelling)

Serpentine Tailings



## MINERAL CARBONATION AT A DIAMOND MINE

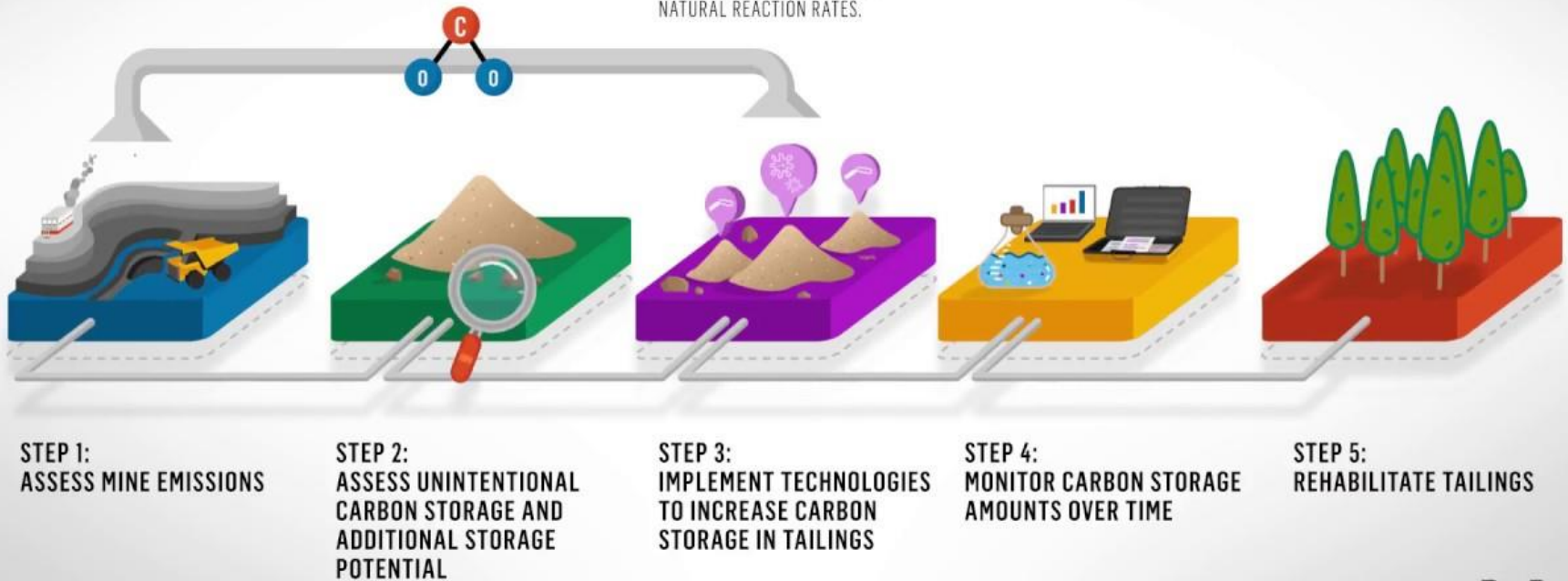
CO<sub>2</sub> EMISSIONS AT A MINE, LARGELY RESULTING FROM ELECTRICITY AND VEHICLES, ARE QUANTIFIED BEFORE AN EMISSIONS OFFSET TARGET IS SET.

ANY UNINTENTIONAL CARBON STORAGE IN MINE TAILINGS IS IDENTIFIED. TOTAL STORAGE CAPACITY AND TECHNOLOGIES BEST SUITED TO FACILITATE STORAGE ARE THEN ASSESSED.

POTENTIAL METHODS TO INCREASE CARBON STORAGE INCLUDE CO<sub>2</sub> INJECTION, PHYSICALLY CHANGING THE TAILINGS LAYOUT TO INCREASE FLOW OF ATMOSPHERIC CO<sub>2</sub>, AND USING MICROBES TO INCREASE NATURAL REACTION RATES.

VARIOUS TOOLS AND APPROACHES ARE USED TO MONITOR THE AMOUNT OF CO<sub>2</sub> BEING STORED AND CONFIRM THAT IT IS ATMOSPHERIC CO<sub>2</sub>, RATHER THAN FROM ANOTHER SOURCE.

AS CO<sub>2</sub> STORAGE IS A FAST PROCESS, TAILINGS CAN BE REHABILITATED AS NORMAL, USUALLY BY COVERING THEM WITH SOIL AND VEGETATION.

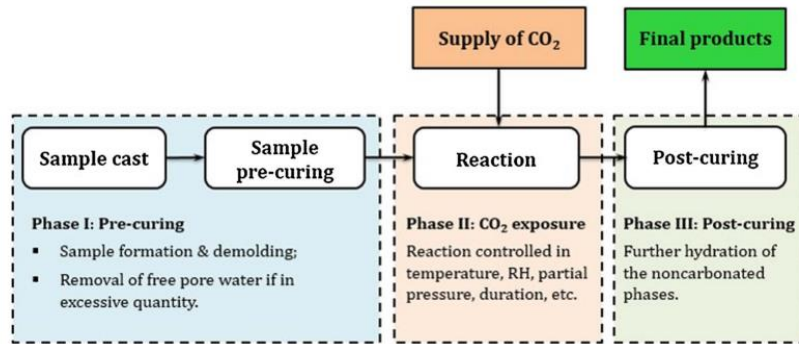


DE BEERS  
GROUP OF COMPANIES





## CO<sub>2</sub> utilization for concrete production



Typical lab-scale procedure (Zhang et al. 2017)



CarbonCure

- Natural weathering is well known to cause detrimental long term effects on concrete (cracking and strength loss) but early stage concrete curing under a CO<sub>2</sub> atmosphere can improve product properties and store CO<sub>2</sub>
- Cement phases, Ca(OH)<sub>2</sub> and C-S-H, react with CO<sub>2</sub> forming micro-sized calcite crystals that can fill pores, even repair micro-cracks and improve compressive strength
- Other applications investigated: use of carbonated aggregates for concrete production, enhance hydraulic properties of silicate phases found in residues, facilitate 3D printing of cement based materials
- Long term stability and durability of products needs to be investigated as well as the evolution of product-layers and diffusion mechanisms in the system





## Carbonation Workshop

Other material for roundtable discussion: energy requirements and open issues identified at MI workshop

**Giulia Costa**

*Laboratory of Environmental Engineering  
Department of Civil Engineering and Computer Science Engineering  
University of Rome "Tor Vergata", Italy*



**TOR VERGATA**  
UNIVERSITÀ DEGLI STUDI DI ROMA



# Energy requirements

Several studies have addressed the issue of the energy penalties associated to carbonation processes in view of the assessment of the feasibility of their application at full scale

	MATERIALS	ROUTE	PENALTIES
ALBANY RESEARCH CENTER (2005)	• Minerals	Slurry Phase	28 %
HUIJGENS ET AL. (2006)	• Minerals	Slurry Phase	25 %
	• Steel Slags		31 %
KELLY ET AL. (2011)	• Minerals	Slurry Phase	55-100 %
	• Steel Slags		> 100 %
KIRCHOFER ET AL. (2012)	• Minerals	Slurry Phase	25-75 %
	• Steel Slags, Coal Fly Ash	Slurry Phase	34-45 %
	• Cement Kiln Dust	Wet	14 %

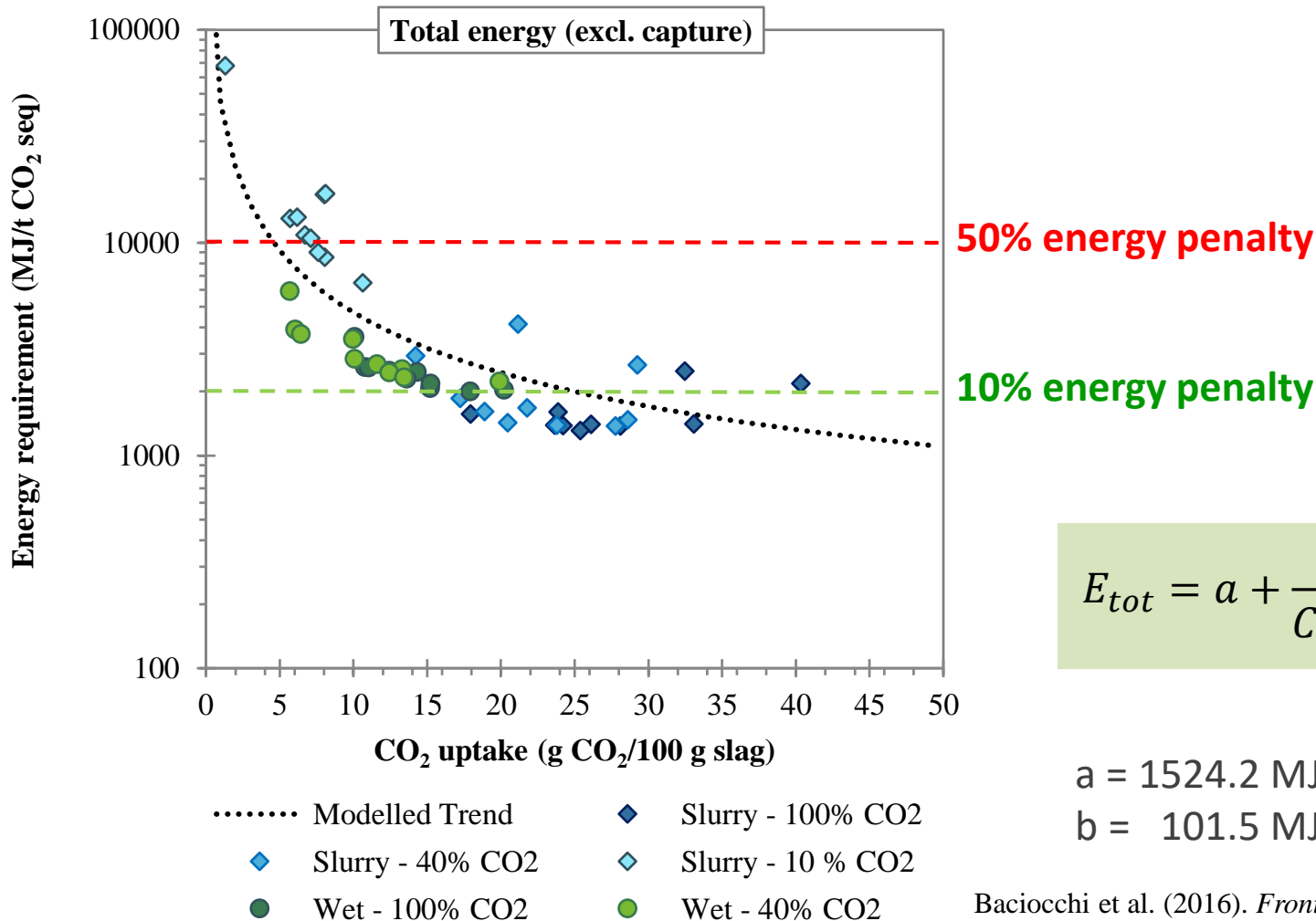


- Large difference in the assumptions and in the resulting total energy penalties
- Lack of a specific wet route process layout



# CO<sub>2</sub> uptake and energy requirements

## ➤ Total energy requirement vs CO<sub>2</sub> uptake (excluding capture!!)

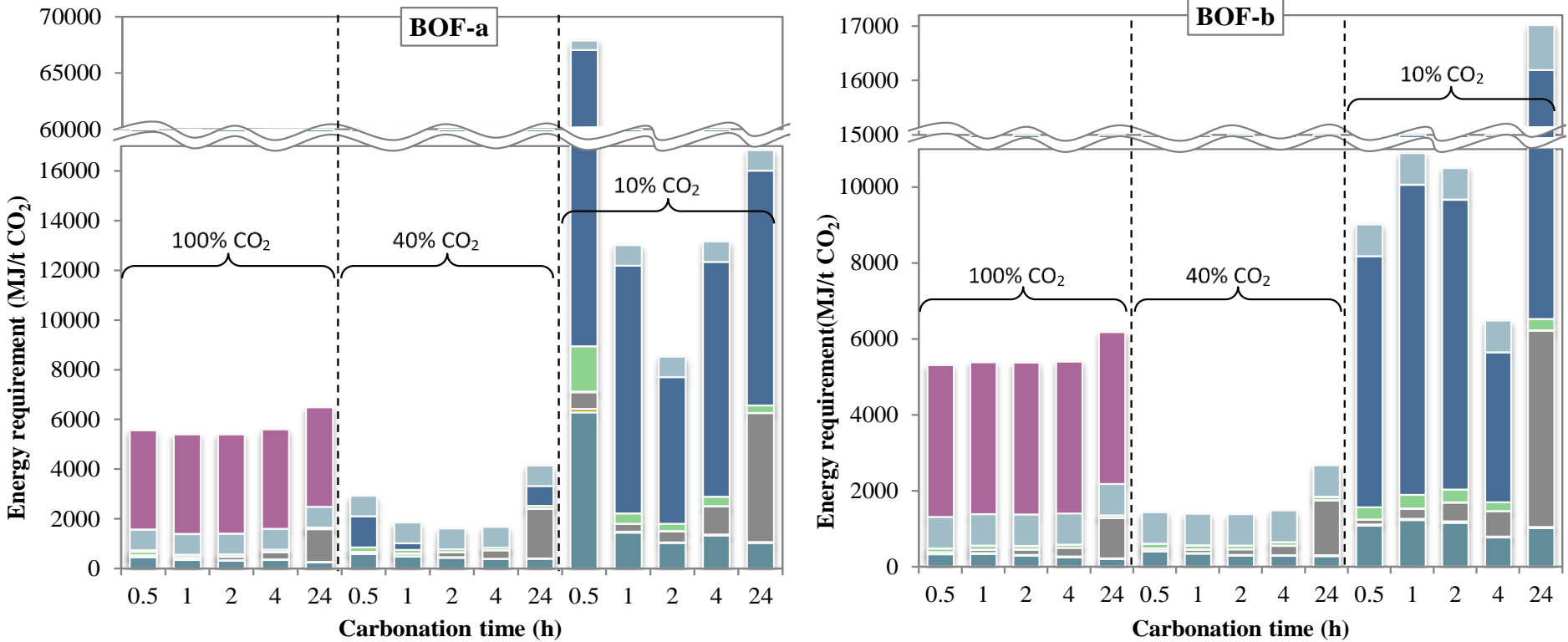


Baciacchi et al. (2016). *Frontiers in Energy research*



# Energy requirements for the Slurry Phase Route

## ➤ Energy requirement of each unit operation

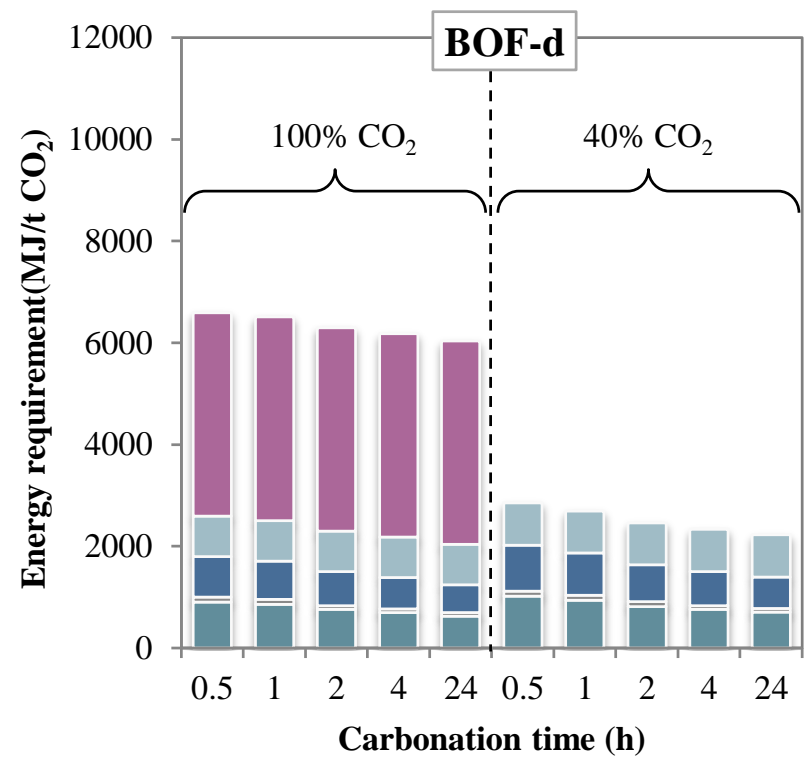
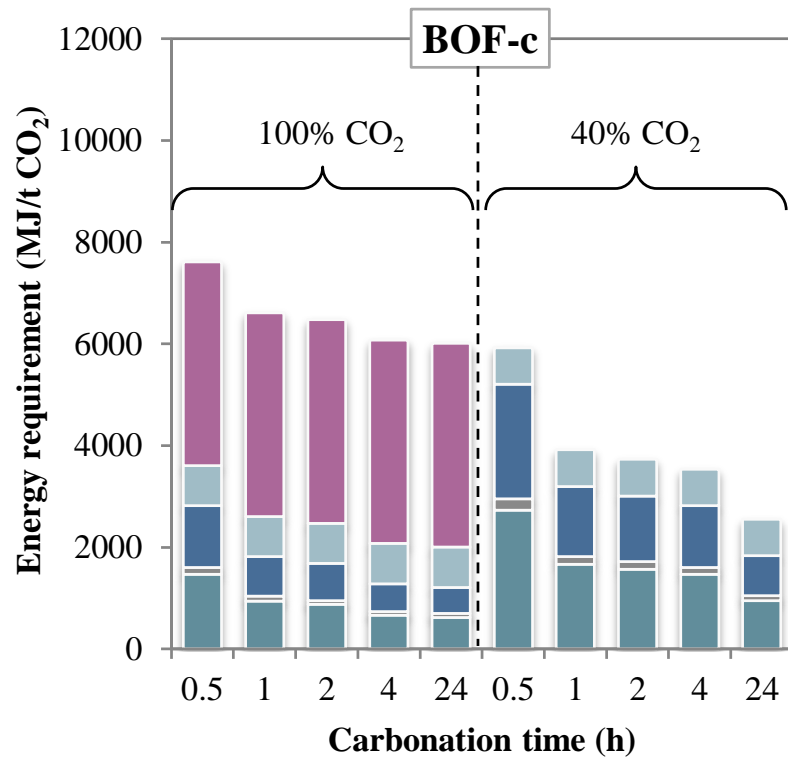


Baciacchi et al. (2016). *Frontiers in Energy research*



# Energy requirements for the wet route

## ➤ Energy requirement of each unit operation



Baciacchi et al. (2016). *Frontiers in Energy research*

■ Capture ■ Compression ■ Heating ■ Reactor mixing ■ Size reduction



# Feedback from MI workshop participants: open issues

---

- Low energy activation & enhanced feedstock reactivity under lean CO<sub>2</sub> conditions
- Complete or near to complete utilisation of mineral feedstock
- Optimization of passive carbon mineralization process
- Molecular-scale simulation of carbonation reactions
- Thermodynamic properties of alkaline minerals for geochemical modeling
- Reusable carbonation additives/reagents
- Making use of carbonation exothermic heat
- Synthetic minerals for carbonation
- In-situ monitoring of carbonation reactions
- Material Reactivity: Flue gas conditions material behavior, kinetics enhancement, pre-treatment options
- Process integration: Energetic modeling, cost modeling
- Cost effective catalytic enhancement of dissolution, separation and carbonation of silicate materials (ligands, CA, seeding, etc.)
- Innovative system integration for recycle and reuse of energy (heat) and materials



# Feedback from MI workshop participants: open issues

---

- Combining various industrial waste materials for CO<sub>2</sub> carbonation technology (i.e. producing NaHCO<sub>3</sub> from desalination waste water)
- Role of the curing step on extent of carbonation and products' properties (e.g. in situ measurements, carbonation tests with model mineralogical phases)
- Scale up issues and data for LCA. Standardization of CO<sub>2</sub> life cycle analysis for carbonation processes
- Effects of operating conditions on the leaching behaviour of the product & solutions for reducing mobility of specific elements (metalloids) after carbonation
- New MC-based pathways for the transition to a circular economy of construction materials
- Innovative MC-based mining & mineral processing pathways for base metals production
- Breakthrough MC-based production of energy carriers
- Dry or low water consuming MC processes



# Feedback from MI workshop participants: open issues

---

- Effect of further water curing after CO<sub>2</sub> curing on the hydration products and microstructure of CO<sub>2</sub>-cured concrete
- Effect of carbonated recycled aggregate on the properties and microstructure of recycled aggregate concrete
- Evolution of different carbonation product-layers (calcium silicates, lime, calcium hydroxide and C-S-H gel) and diffusion mechanism of gas and ions through the layer
- Restructure process of cementitious system during carbonation in the bulk solid phase
- Nucleation and growth mechanisms for carbonation/de-carbonation
- High CO<sub>2</sub> density precursors for *in situ* carbonation
- Ultra-fast carbonation
- Cheap CO<sub>2</sub> sources
- Creating realistic time-lines for technology adoption and creating a climate model that evaluates its effectiveness

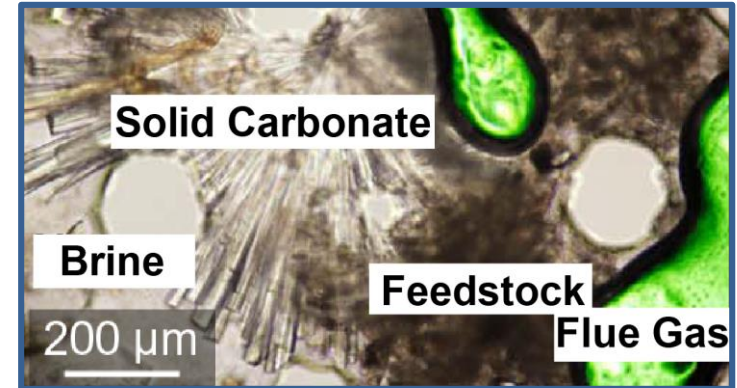


## • Scientific Challenges

- ✓ Carbon mineralization, a slow phenomenon, has been observed to proceed at rates orders of magnitude faster than predicted by conventional models.
- ✓ Predicting the dynamic speciation and product properties of solid feedstock/fluid/CO<sub>2</sub> systems is the key to unlock carbon mineralization to control reaction pathways and rates.

## • Research Directions

- ✓ Broad-spectrum collection of thermodynamic and kinetic data associated with carbon mineralization of feedstocks, focusing on reactions and mass transfer at solid-fluid interfaces.
- ✓ Development of broadly available on-line high-resolution investigative tools for local mass transfer measurements and phase characterization.
- ✓ Mechanistic understanding and predictive phenomenological modelling of carbon mineralization processes at the molecular level.



*Microfluidics carbon mineralization experiment  
(after Harrison et al., 2017. Chem. Geol.)*

## • Impacts

- ✓ Fast-tracked novel market ready net positive energy carbon mineralization processes for permanent CO<sub>2</sub> binding.
- ✓ Provide a ranked list of high potential carbon mineralization feedstocks.
- 
- ✓ Provide internally-consistent and validated thermodynamic, transport and kinetic data for carbon mineralization systems.
- ✓ Improved understanding of solid-liquid-CO<sub>2</sub> reaction pathways under a broad range of conditions.





## PRD 2: Tailoring material properties to enable carbon storage in products

### Scientific Challenges

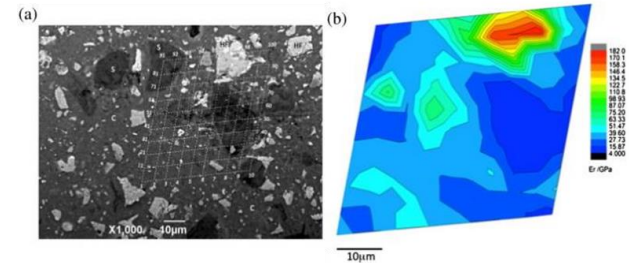
- ✓ To realize the potential of carbon mineralization for the built environment we must predict and control processes to consistently meet product performance criteria.
- ✓ Linking product performance to micro-structure, geochemistry and mineralogy is the key to achieve consistency.

### Research Directions

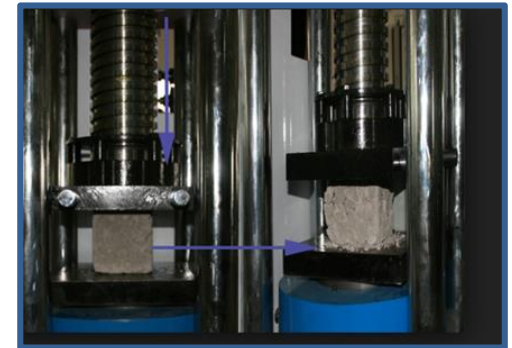
- ✓ Develop and apply real time monitoring of carbon mineralization product properties during reaction.
- ✓ Exploit nano-characterization for deriving insight into solid and aqueous speciation and improve control on bulk physical and chemical properties.
- ✓ Understand and model structure-property-performance relationships.
- ✓ Develop pre/post processing treatments to improve product performance criteria.
- ✓ Predict and control long term durability for applications.

### Impacts

- ✓ Reliable cost-effective processes for generating construction materials that meet performance criteria while permanently storing CO<sub>2</sub> on a Gigaton scale.
- ✓ Provide a better understanding of process conditions on product properties (e.g. strength, leaching and durability).
- ✓ Insights applicable to the carbonation of industrial waste streams and sub-surface mineral trapping.



Nano-indentation grid testing  
(Mo et al., *Cement and Concrete Research*, 2016)



Carbonated building block performance testing  
(Columbia Dept. of Civil Engineering)



Concrete forest (The Who's Who in *Building & Construction*, Houston Spring 2016 Ed.)



# References

---

- MI experts workshop on CCUS full report, US DOE, May 2018. Carbon mineralization chapter pages 3-19 to 3-30 <https://www.energy.gov/fe/downloads/accelerating-breakthrough-innovation-carbon-capture-utilization-and-storage>
- Zevenhoven et al, *Catal Today* 115 (2006) 73–79
- Gerdemann et al, *Env Sci & Tech.* 41 (2007) 2587–2593
- Said et al, *Applied Energy*, 177 (2016) 602-611
- Teir et al., *Patent US 8603428 B2 Dec. 10 2013*
- Pan et al, *Aerosol Air Qual Res* 12 (2012) 770–791
- Zingaretti et al 2014 *Industrial & Engineering Chemistry Research*, 53 (2014) 9311-9324
- Baciocchi et al in: Bhanage B M, Arai M, *Transformation and Utilization of Carbon Dioxide. Green Chemistry And Sustainable Technology*, 263-302 (2014)
- Maroto Valer et al., (Penn State Research Foundation, US) patents 2004 (WO), 2005 (US)
- Baciocchi et al., *Greenhouse Gases: Sci Technol.*, 4 (2012) 312-19
- Baciocchi et al, *J Hazard Mater*, 283 (2015) 302-313
- Bobicki et al, *Prog Energy Combust*, 38 (2012) 302–320
- Kandji et al, *Applied Geochemistry* (2017), 84
- Zhang et al, *Journal of CO<sub>2</sub> Utilization* 21 (2017) 119–131
- Baciocchi et al. *Frontiers in Energy research* (2016)

