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In the project: Carbonate production by sequestration of industrial CO₂: revalorization of mine and industrial waste

Motivation

Greenhouse Gas (GHG) emissions

- In 2011, around 31,780 million metric tonnes CO₂e added to the earth's atmosphere through fossil fuel combustion (U.S.EPA, 2013, U.S. inventory on GHG emission 1990-2011)
- Canadian emissions in 2011 were 702 Mt CO₂e (Environment Canada, 2013, National inventory report on GHG emission)
- In 2010, Quebec has an emission of 82.5 Mt CO₂e of which 33% was from industry (MDDEFP, 2013, GHG emission Inventory report)



Carbon Capture and Storage (CCS) technologies

- Geological Storage, Oceanic Storage and Mineral Storage
- Mineral carbonation**
- Naturally occurring weathering process in which alkaline or alkaline earth metal compound react with CO₂ to form stable and environmentally benign products



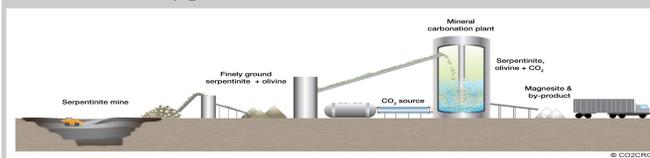
Challenges and Goals

Challenges

- Increasing atmospheric CO₂ concentration due to industrial activity
- Challenge in revalorization of mine residues to give useful products
- Develop a simple, environmental friendly, energetically and economically efficient process for industrial CO₂ sequestration
- Very slow reaction kinetics of dry mineral carbonation process

Goals

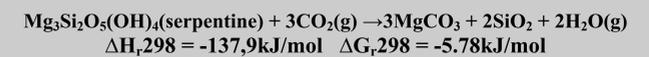
- Develop a simple and promising process for the treatment of industrial flue gas (CO₂) using direct dry gas-solid carbonation of magnesium silicates
- Use different waste residues as feed stock material and revalorize into valuable by products



Strategy

Direct dry gas-solid mineral carbonation

- Magnesium bearing minerals reacts with CO₂ to form magnesium carbonate:



- Enhance the reaction kinetics through different pre-treatments such as grinding, heat and magnetic separation
- Lower the temperature, pressure and reaction time for a viable process.

Magnesium silicates source

- Québec has two principal mine regions which are abundant in serpentine minerals (Thetford Mine and neighboring towns of Black Lake and mine Asbestos).



Originality

- Gas mixture of approximate industrial flue gas composition used to avoid the need for carbon capture processes
- Use of mineral waste residue with different mineral phases will reduce the mining and purification cost
- Revalorization of a waste residue into useful products



Material

Sample name and location

- Serpentine mining residue from Black Lake, Québec

Sample preparation

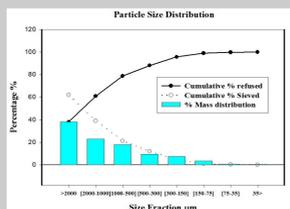
- Mixing, crushing and grinding

Sample characterisation

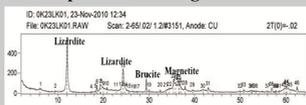
- BET specific surface area (<75 μm) = 12 m²/g
- Average pore diameter = 14 nm

ICP-AES after fusion

Size fraction (mm)	LOI*	MgO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	CaO
	(%)	(%)	(%)	(%)	(%)	(%)
>2	12.01	41.46	37.01	11.18	0.39	0.08
[1-2]	12.02	36.89	37.73	9.02	0.61	0.24
[0.5-1]	11.95	41.25	37.98	10.12	0.46	0.26
[0.3-0.5]	11.9	39.39	36.46	11.88	0.47	0.27
[0.15-0.3]	11.8	34.83	35.97	12.16	0.64	0.37
[0.075-0.15]	10.81	36.94	34.51	18.59	0.52	0.47
[0.035-0.075]	10.42	31.91	32.39	21.95	0.69	0.55
Average	11.57	37.53	35.99	13.42	0.54	0.32
Std	0.67	0.47	1.96	4.52	0.11	0.16



Particle size distribution Black lake serpentine mining residue



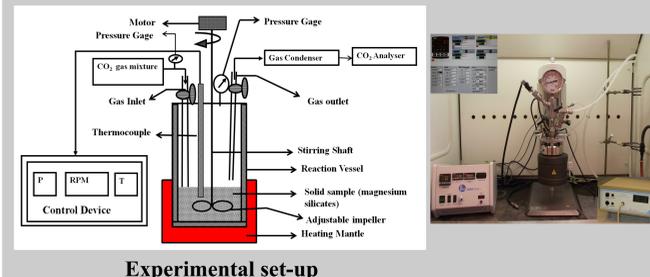
XRD of Black lake serpentine mining residue

- Black lake mine residues constitute mainly lizardite and magnetite with minor presence of brucite and chromite

Method

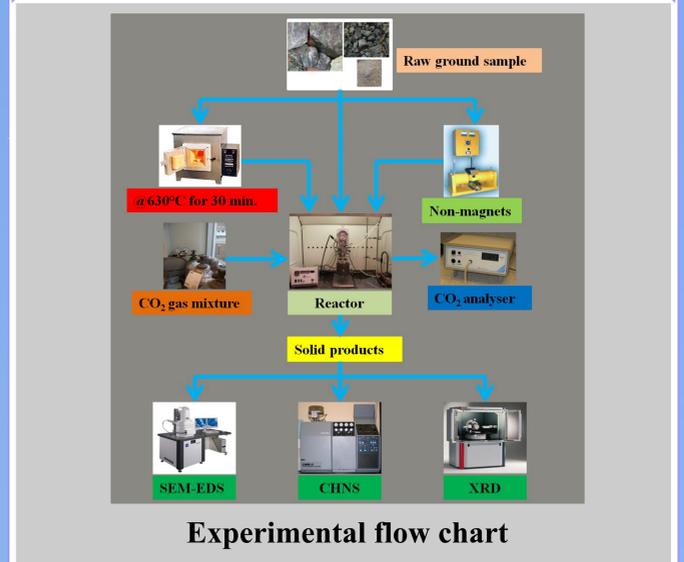
Conditions

- Gas Mixture composition = N₂/CO₂/O₂/78/18/4 % (v/v)
 - Device: 300 mL Parr stirred reactor (Parr 5560 Mini Bench Top Reactor)
 - Reaction type: Dry gas-solid carbonation
- Study Range:**
- Temperature: 25 to 300°C
 - Pressure: 5 to 100 atm
 - Reaction Time: 15 to 360 min
 - Solid Size: 1 to 25 g
 - Particle size: <75 μm



Experimental set-up

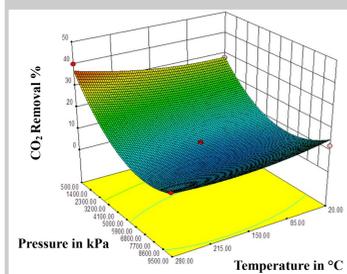
Process flow



Experimental flow chart

Results

- Optimisation tests were carried out with raw and pre-treated (heat treated and non-magnetic) serpentine mining residues.
- Samples are ground to a size <75 μm
- Tests were carried out in two ways:
 - Conventional single variable at a time method
 - Box-Behnken analysis



Optimum conditions
258°C, 5.6 atm for 15 min with 40% CO₂ removal.

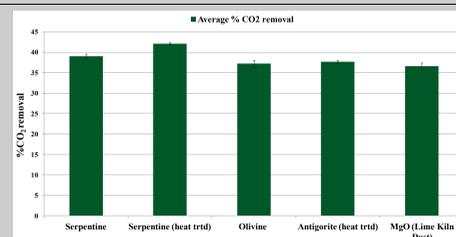
97.5% of the variable responses are explained

Response surface graph showing the interaction between temperature and pressure in %CO₂ removal

Results

Box-Behnken Validation

Sample type	Temperature (°C)	Pressure (atm)	Time (min)	CO ₂ removal (%)
Raw	258	5.6	15	36.3±2.7
Heat treated	258	5.6	15	37.1±0.8
Nonmagnetic	258	5.6	15	35.7±2.7



Direct dry gas-solid carbonation with different Mg bearing materials

- All the results presented are calculated based on the CO₂ analyser value
- Preliminary results reveal that the removal is more due to adsorption rather than carbonation

Conclusion & Acknowledgment

Conclusions

- Primary studies indicate that serpentine milling residue can be used for direct dry gas-solid CO₂ capture
- Grinding of the sample is required for better efficiency
- It is working at low pressure and mild temperature
- Preliminary results indicates that removal is due to adsorption rather than real carbonation
- Process has to be improved for better efficiency

Acknowledgment

