



CO₂ Capture and Storage –
Response to Climate Change
13-14 April 2011, Vilnius, Lithuania

Prospects of CO₂ sequestration in serpentinites of South Lithuania

Stasiulaitiene, Inga¹, Denafas, Gintaras¹, Zevenhoven, Ron²

¹Department of Environmental Engineering, Kaunas University of Technology, Radvilenu pl. 19, LT50254
Kaunas, Lithuania, e-mail: inga.Stasiulaitiene@ktu.lt

²Thermal and Flow Engineering Laboratory, Abo Akademi University, Biskopsgatan 8, FI-20500, Abo/Turku, Finland



Introduction

Lithuania, similarly to many countries worldwide, has ratified the Kyoto protocol and has to fulfill its commitments on CO₂ emissions. Although presently Lithuania meets Kyoto protocol requirements, its contribution to the world's CO₂ emissions combined with the fact that geological conditions are not suitable for storage of the CO₂ in the structural traps of saline aquifers, encourage an assessment of mineral carbonation. Utilization of the natural local resources, such as serpentinites, for storing captured CO₂ could provide safe and permanent storage option inside of the country. These natural local resources should be investigated as potential raw materials for use in mineral carbonation processes.

Serpentinites are distributed in the southeastern part of Lithuania within the Varena Geological Province of the East Lithuanian Domain. More than a dozen of serpentinite bodies were identified in southern Lithuania. Owing to their association with the iron ore deposits these bodies have been extensively studied by the drilling. They are covered by platform sediments of 280 - 500 m thick. The depth of the basement increases towards the northwest. The depth of the serpentinites layer varies from 360 m and continues to 1 – 1.5 km.

Indirect gas/solid carbonation has been performed with Lithuanian serpentinites in cooperation with Abo Akademi University (ÅA).

The rock samples were taken from different mining holes of various depths, the one (Fig.1) with the highest MgO (12.48 %) concentration has been chosen for further experiments.

Production of magnesium in the form of Mg(OH)₂ from serpentinite rock, followed by conversion into MgCO₃ using a pressurised fluidised bed reactor (PFB) at ÅA operating at 490 °C and elevated pressures (20-50 bar) has been performed (Fig.2).

The method of Mg(OH)₂ extraction from Lithuanian serpentinite has been used as shown in Fig.3.

The experimental setup (Fig.4) consisted of a heated vertical reactor, operated as a fluidized bed with a sintered plate at the bottom (internal diameter 1.4 cm, height 40 cm), a CO₂ preheater to assure the right reaction temperature, a cyclone for solid particle removal from the exiting fluid and the necessary measuring/controlling equipment. As a source of CO₂, a helium pressurized CO₂ bottle was used, allowing for pressures up to 120 bar (CO₂ becomes super-critical above 73.9 bar and 31.1°C). The pressure of CO₂ gas was maintained at ~20 bar.

Methods



Fig. 1. Serpentinite sample used in the experiment.

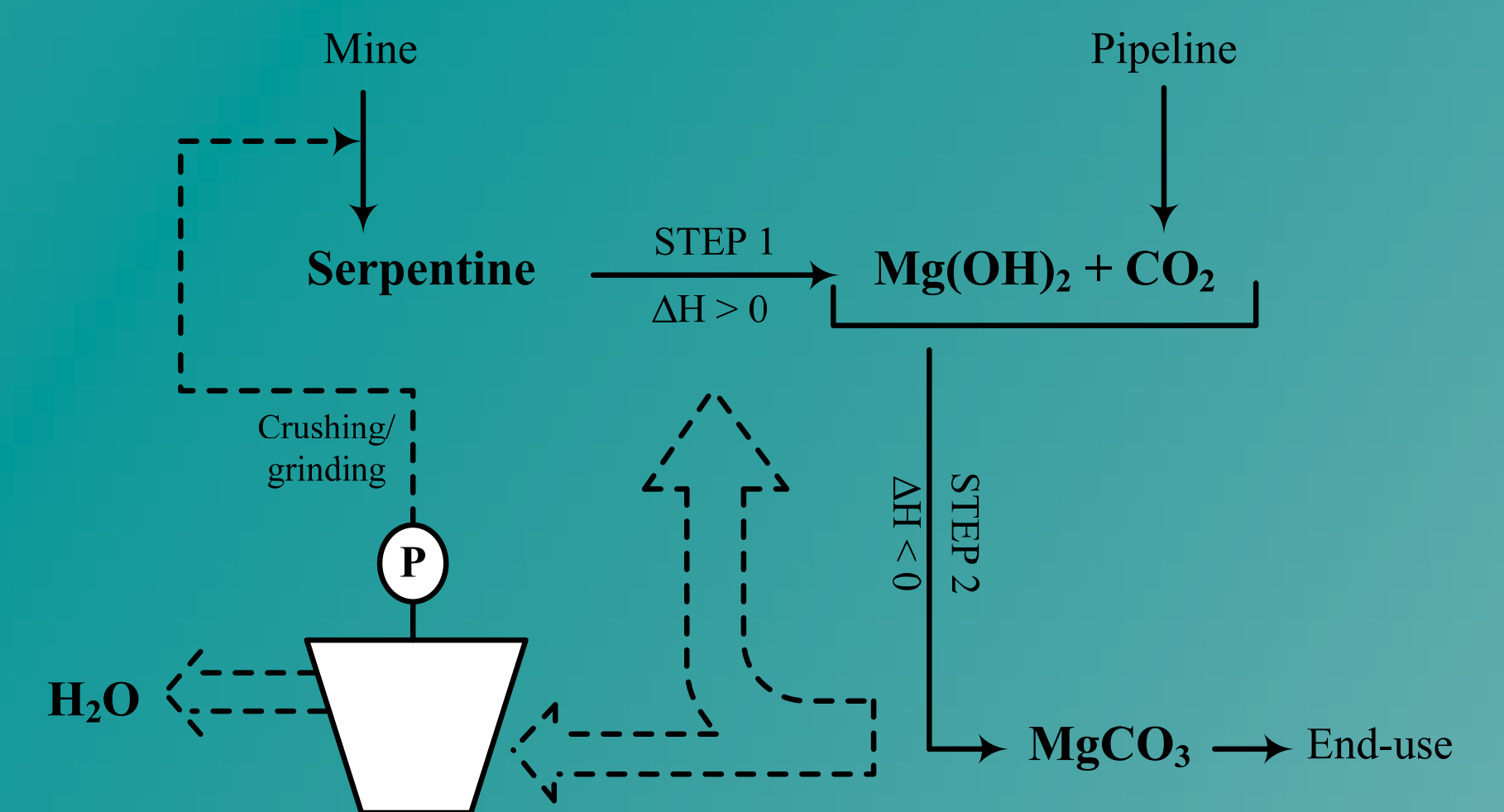


Fig.2. Schematic illustration of a staged gas-solid mineral carbonation process.

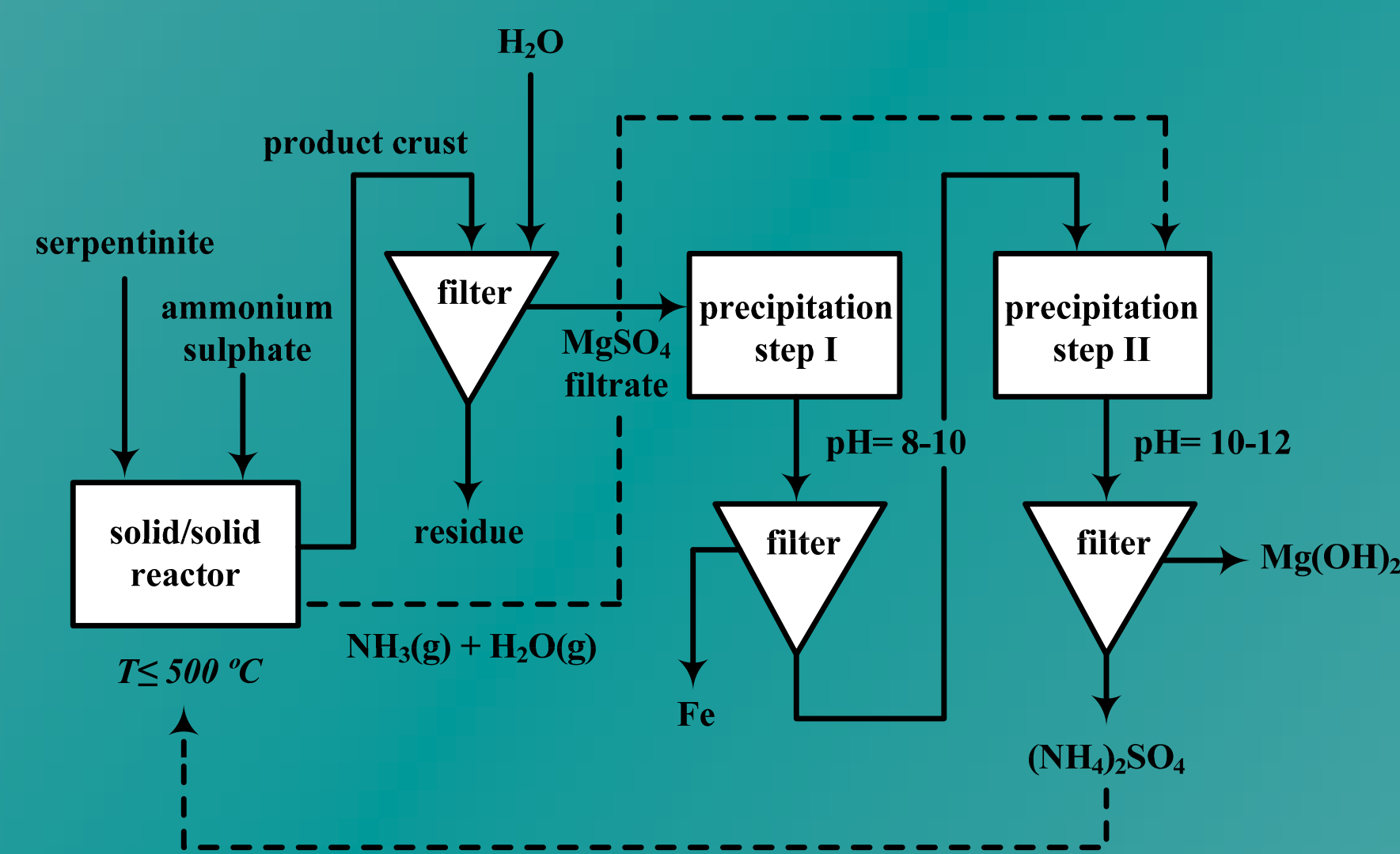


Fig.3. Schematic picture of Mg(OH)₂ production from serpentinite rock.

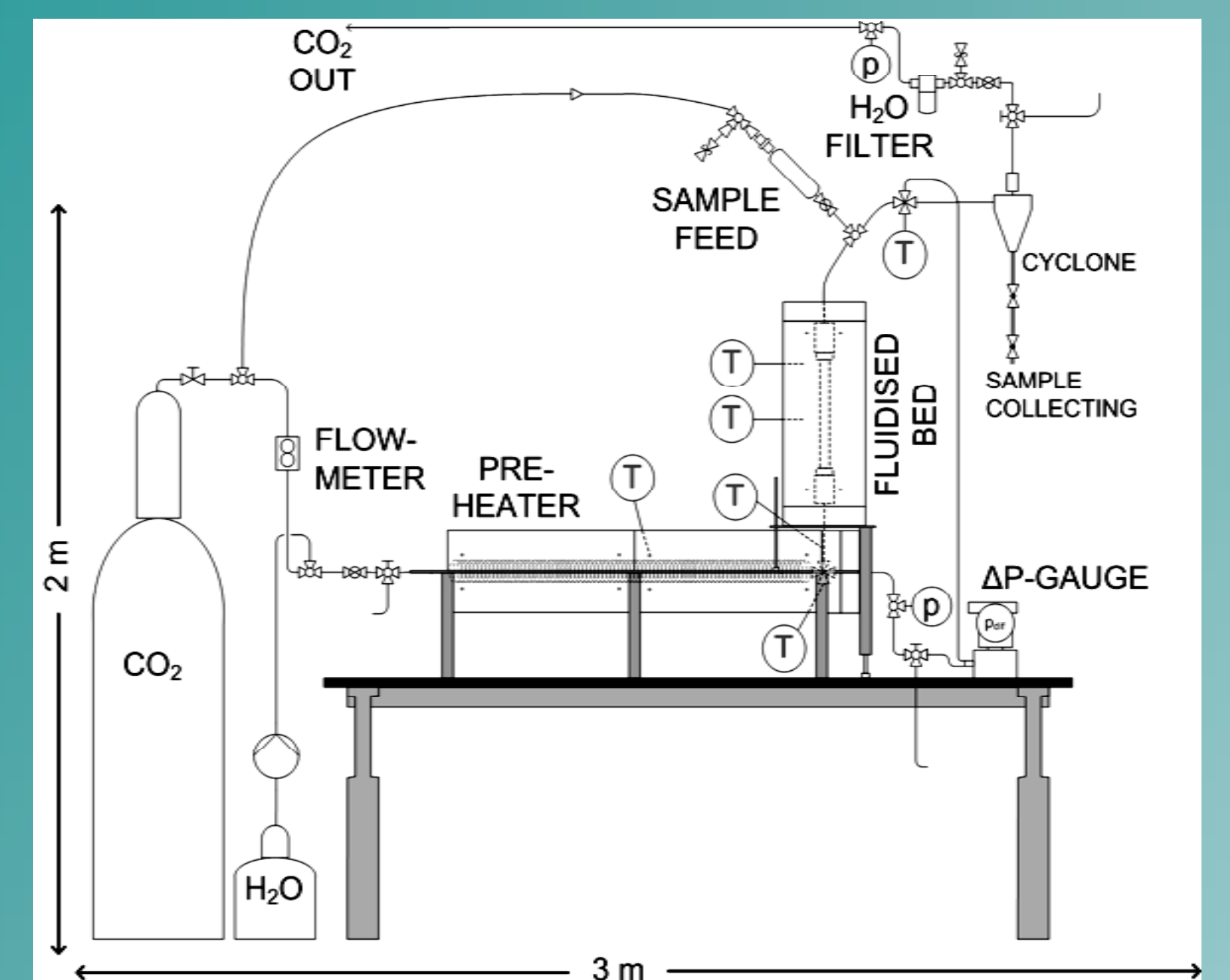


Fig.4. Experimental setup for mineral carbonation of Mg(OH)₂ obtained from serpentinite.

Results and Discussion

Table 1. Concentrations of extracted Mg and Fe from Lithuanian serpentinite in reaction mixture produced at different temperatures.

t (min)	T (°C)	Serpentinite (g)	Ammonium sulphate (g)	Mg/Fe	Mg wt-%	Fe, wt-%
10	430	2	3	1.5	10.83	5.69
10	480	2	4	1.5	13.16	4.28
10	480	2	4	1.5	11.62	3.98
10	520	2	3	1.5	19.63	10.12
20	500	2	3	1.5	16.64	2.36
20	535	2	3	1.5	16.82	2.17
23	535	2	3	1.5	23.42	3.57

Table 2. Pressurised fluidized bed carbonation results using Lithuanian Mg(OH)₂. 1-relatively pure sample prepared at KTU, 2- Fe and Si containing sample prepared at KTU, 3,4 – relatively pure samples prepared at AAU

Sample of Mg(OH) ₂	Sample size (g)	Mean particle size (μm)	Experiment time (min)	Mean pressure of CO ₂ (bar)	Fluid velocity (cm/s)	Carbonation degree (%-wt)
1	3.8	254.5	9	19.9	15.3	52
2	4.2	255.9	9	19.8	16.0	45
3	4.0	168.3	9	20.4	12.2	54
4	4.0	168.3	3	20.3	12.7	53

The first step of indirect carbonation experimental results (Table 1) revealed that an increase in both the reaction time and the temperature plays important role for the efficiency of Mg (and Fe) extraction. The highest level of Mg extraction (23.42 wt-%) was achieved at temperature of 535 °C, and reaction time of 23 min.

The temperature of the carbonation reaction was 490 °C, pressure was 20 bar. Carbonation degrees (Mg(OH)₂ carbonation to MgCO₃, which is determined by comparing the amount of CO₂ released from the sample, to that which would be obtained for a 100 % pure carbonate of the same weight) appeared to be very similar among the samples. The sample containing iron and silicate impurities (#2 in Table 2) gave the lowest carbonation degree (45 %).

The indirect carbonation experiments performed with the step of producing Mg(OH)₂ allowed achieving relatively high carbonation degree of 52 %, after 9 minutes, provided that Mg(OH)₂ was efficiently separated from impurities. For the carbonation efficiency the Mg(OH)₂ production showed little dependence on the type of reaction chamber, although it appeared quite sensitive to the conditions of the chemical precipitation steps.