CO_2 Storage in Novel CO_2-H_2O Phases at High Pressure

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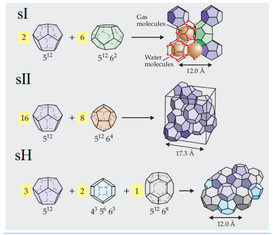
Abstract

Optical Raman spectroscopy was used to analyze the high-pressure behavior of a CO_2-H_2O system *in-situ* up to 54 GPa. Between 26.9 and 1.0 GPa, we observed a new CO_2-H_2O compound whose vibrational spectra differed dramatically from pure CO_2 and H_2O. When the pressure was decreased to <1 GPa, the CO_2 vibrons and OH stretch reverted to those expected for pure CO_2 and H_2O respectively, indicating a CO_2-H_2O mixture rather than a CO_2-H_2O molecular compound. Based on the difference in spectra seen upon pressure cycling, as well as the difference in spectra seen at multiple locations inside the sample, we concluded that multiple forms of the CO_2-H_2O compounds may exist.

Background

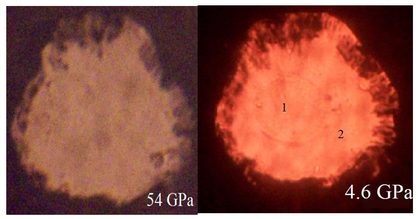
- Increasing level of atmospheric CO_2
- Climate change
- Proposed alternative methods:
  -Oceanic, underground, geologic
- Clathrate containment

What is a clathrate?
At low temperatures and/or high pressures, water can react with gas molecules to form crystalline inclusion compounds called clathrates. The water molecules form polyhedral cages that can accommodate differently sized guest molecules on the inside.



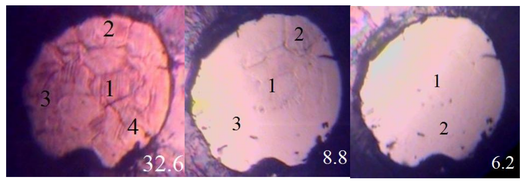
Results

Sample 1
-Mixture of CO_2 and H_2O
-54 to 1 GPa
-Lowered to 1, re-compressed to 10 GPa

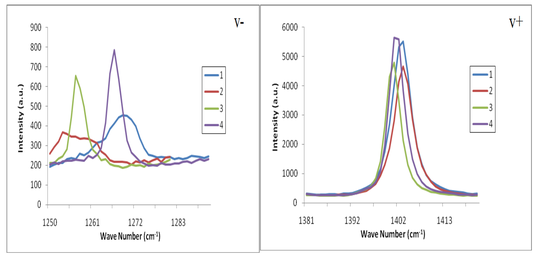


Distinction in physical characteristics was observed at below 4.6 GPa.

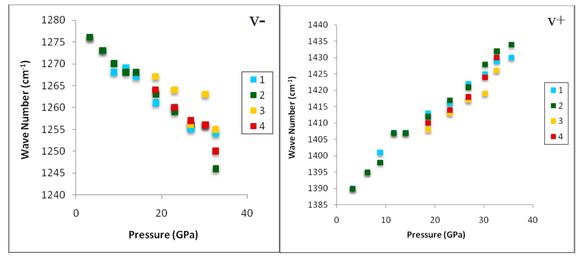
Sample 2
-Much more CO_2 than H_2O
-35 GPa to 5 Gpa
-Decrease in pressure



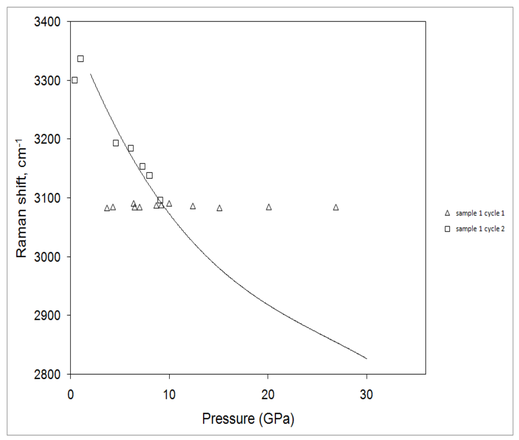
Crystallization, and gradual disappearance of the crystalline properties, occurred.



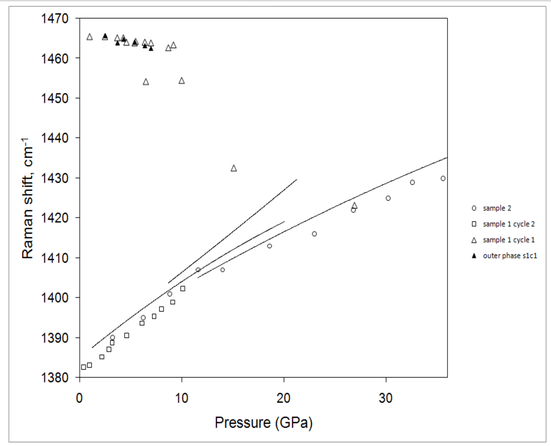
Spectra obtained at 11.6 GPa. The spectra differ depending on the location at which they were obtained



Location of the peaks, grouped by location inside the sample, as a function of pressure.



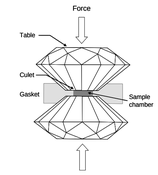
Expected water spectra vs pure water spectra.
In sample 1, the rate of change of the peaks is much less than that of the pure water.



Expected CO_2 vs pure CO_2.
In sample 1, the peaks differ drastically when pressure is lowered but revert to pure CO_2 when recycled.

Methods

- Tungsten gasket filled with water
- Ruby chips for pressure calibration
- CO_2 loaded using gas pressure vessel
- CO_2-H_2O mixture compressed inside diamond anvil cell



- Raman Spectroscopy
  -In situ probe for vibrational peaks
  -Various locations

Conclusions

- Variations in spectra by:
  -Location
  -Cycle
- Physical differences in sample
- Drastic differences compared to data for pure CO_2 and H_2O
- Dependent on initial composition/conditions
- Sample 2 was a CO_2-H_2O mixture while sample 1 was a CO_2-H_2O compound

Future Research

- Temperature
- Initial CO_2:H_2O composition
- Initial Pressure/Temperature conditions
- Structure