

Neutron diffraction, excess sorption and infrared study of CO₂ interaction with Na-rich montmorillonite at CCS P-T conditions

ELIZABETH KRUKOWSKI^{1*}, ANGELA GOODMAN², GERNOT ROTHER³, EUGENE ILTON⁴, GEORGE GUTHRIE², ROBERT BODNAR¹

^{1*} Virginia Tech, Blacksburg, VA, USA, egk@vt.edu*, rjb@vt.edu

² National Energy Technology Laboratory, U.S. Department of Energy, Pittsburgh, PA, USA, angela.goodman@netl.doe.gov, george.guthrie@netl.doe.gov

³ Oak Ridge National Laboratory, Oak Ridge, TN, USA, rotherg@ornl.gov

⁴ Pacific Northwest National Laboratory, Richland, WA, USA, eugene.ilton@pnl.gov

The interaction of CO₂ with Na-rich montmorillonite clay (Na-mont) as an analog for a clay-rich caprock was studied to better understand how CO₂ might interact with caprock at P-T conditions relevant to carbon capture and storage (CCS). Neutron diffraction, excess sorption and Attenuated Total Reflectance – Fourier Transform Infrared Spectroscopy (ATR-FTIR) analyses of Na-mont at 35°C and 50°C, and pressures relevant to CCS, were conducted.

Neutron diffraction and excess sorption measurements were conducted on Na-mont at CO₂ pressures from 0-200 bars. Neutron diffraction measurements show a shift in the d(001) spacing from 12.10 Å to 12.55 Å and a decrease in the intensity of the d(001) peak, both of which are consistent with CO₂ entering the interlayer region of the clay. Excess sorption isotherms were determined gravimetrically to provide a better understanding of changes in the density of CO₂ near clay surfaces. Maxima in the excess sorption isotherms were observed at a bulk density ≈0.15 g/cm³ and pressures of 58 bars (35°C) and 64 bars (50°C). As the bulk density of the CO₂ increases, the amount of CO₂ sorbed to the clay decreases.

To better understand the specific structural locations of CO₂ interaction with the Na-mont, the same clay sample examined by neutron diffraction and excess sorption was studied using ATR-FTIR. Measurements were conducted on both dried and hydrated Na-mont from 1-82 bars at 35° and 50°C. ATR-FTIR data show that the asymmetric stretch and bending mode of sorbed CO₂ is impacted by the presence of interlayer water, but the absorption bands due to adsorbed water between 3564 and 2975 cm⁻¹ are not affected by the presence of CO₂. Specifically, the frequency of the asymmetric sorbed CO₂ band is at 2339 cm⁻¹ in dehydrated clay, but at 2344 cm⁻¹ in hydrated clay. The sorbed CO₂ bands increase in intensity with increasing CO₂ pressure. The stretching mode for the isolated inner hydroxyl groups at 3623 cm⁻¹ is present in both the hydrated and dehydrated clay and is not affected by the presence of CO₂.

Analysis of the data indicates that sorbed CO₂ enters the interlayer space and potentially sorbs onto the edges of octahedral sheets in the Na-mont structure. If CO₂ does enter the Na-mont interlayer it could affect the rheological properties of the caprock, but further work is needed to determine if this might lead to a degradation or enhancement of seal quality.

Origin of Q-gases in pristine meteorites: an experimental study

M. KUGA^{1*}, B. MARTY¹, Y. MARROCCHI¹, L. TISSANDIER¹ AND L. ZIMMERMAN¹

¹Centre de Recherche Pétrographique et Géochimique - CNRS, Nancy, France (* correspondence : mkuga@crpg.cnrs-nancy.fr)

Most of heavy noble gases (Ar, Kr and Xe) trapped in primitive meteorites are hosted in the so-called phase Q which is commonly thought to be part of the Insoluble Organic Matter (IOM) of primitive meteorites [1]. This enigmatic phase is found in many classes of meteorites, implying that it was widespread in the early solar system [2]. Q-gases are characterized by an important fractionation relative to solar composition in favor of the heavy elements and isotopes (up to 1.3%/amu for xenon isotopes, the least-prone to isotopic fractionation) [2]. Despite many effort, the origin of Q-gases and the nature of their carriers are still poorly understood. Several experiments performed in the past decades showed that Q-gas characteristics could be reproduced to some extent by trapping ionized noble gases in various solids [3-7]. It has been interpreted by [6-7] as evidence for anomalous adsorption of xenon on growing or crumbled surfaces. This process takes place at low ion energy (<10 eV) and involves chemical bonds between ionized xenon and reactive sites onto the surface. Such a process could have occurred in the outer part of the protoplanetary disk or in the parent cloud, where gases and organics are subjected to intense UV light from the young Sun or near-by stars [8]. However, it remains unclear whether this process controls the xenon isotopic fractionation and if it can account for the light noble gas fractionation reported in phase Q [2].

The goal of this study is to better constrain the trapping and the isotopic fractionation of noble gases under ionizing conditions. Thus, we investigated the synthesis of carbonaceous particles in presence of noble gas ions by dissociating CO in a microwave plasma, at low pressure and ambient temperature. Solid particles were characterized by SEM, TEM, Raman Microscopy and Infrared Spectroscopy. The abundances and the isotopic ratios of noble gases were analyzed by static Mass Spectrometry.

Xenon trapped in carbonaceous particles is enriched in heavy isotopes by a factor of 0.2 to 1.7%/amu, depending mostly on the duration of the experiment. Assuming Henry's law for xenon trapping in carbonaceous particles, the trapping yield of xenon ions is 12000 mbar.(cm³.g⁻¹)⁻¹ in average, which can largely account for the amount of xenon trapped in phase Q [2,7]. Preliminary element ratios Ar/Xe and Kr/Xe are in the range of those measured in meteorites [2]. Future investigations will focus on the role of plasma parameters on the trapping efficiency and isotopic fractionation of Ar, Kr and Xe.

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