



## Preface

## CO<sub>2</sub> geological storage: Integrating geochemical, hydrodynamical, mechanical and biological processes from the pore to the reservoir scale

The trend in global warming observed since the beginning of the industrial revolution is attributed to a great extent to the accumulation of anthropogenic carbon dioxide (CO<sub>2</sub>) in the Earth's atmosphere (IPCC, 2007). The capture and storage of CO<sub>2</sub> in geological formations is considered to be one of the most promising approaches to reduce further emissions and to minimize the existing climatic risk until alternatives to fossil fuels become available (e.g. Gunter et al., 1997; Holloway, 1997; Bachu, 2000; Gunter et al., 2000; IPCC, 2007, and thereafter).

During the last decade, fundamental research has focused increasingly on the short and long term effects of the massive injection of anthropogenic carbon dioxide in various geological environments, such as depleted oil and gas reservoirs, saline aquifers, coal beds and (ultra) basic rocks, in order to assess the feasibility of storing large amounts of CO<sub>2</sub> in the subsurface. Sequestration processes involve different trapping mechanisms according to the physico-chemical and biological conditions encountered in the host reservoir. Hydrodynamics involves the storage of CO<sub>2</sub> as a free-phase (i.e. gas, liquid or supercritical CO<sub>2</sub>) in structural or stratigraphic traps. Solubility trapping consists of dissolution of CO<sub>2</sub> as aqueous carbonated species in formation water. Mineral trapping involves the incorporation of CO<sub>2</sub> in a stable mineral phase, for example via the precipitation of secondary carbonate minerals. This last mechanism appears the most desirable approach as it ensures a permanent and secure sequestration of CO<sub>2</sub>.

There are still significant fundamental research needs that must be addressed before geologic formations can be widely used for carbon sequestration. Experimental and numerical modelling of the fate of injected CO<sub>2</sub> and the assessment of the sealing capacity of the wellbores and caprocks must be conducted. Appropriate monitoring methodologies relying both on geophysics and geochemistry need also to be validated on pilot sites. Additionally, because bacteria can be involved in mineral dissolution and formation, the potential effects of subsurface microbial ecosystems on the fate of injected CO<sub>2</sub> must be evaluated.

Since the first Chemical Geology special issue on CO<sub>2</sub> sequestration (Oelkers and Schott, 2005), a growing scientific community has contributed to the better understanding of the coupled biological, geochemical, mechanical and hydrodynamical processes that result from the strong disequilibrium induced by injection of large amounts of CO<sub>2</sub> into wellbores and reservoirs and the accompanying modification of the pH of the formation waters. New concepts are now emerging to study the consequences and kinetics of these effects, using well-controlled laboratory experiments and field observations that are aimed at improving and validating numerical codes for CO<sub>2</sub> trapping. In particular, experimental studies are now focusing on the thermodynamic stabilities and the kinetics of dissolution and precipitation of important rock-forming minerals in order to determine the extent and

rate of formation of stable carbonate minerals in injection environments. In addition, the modification of deep biosphere metabolisms potentially induced by the injected CO<sub>2</sub> and the complex kinetics associated with biologically-induced precipitation of carbonates are now thought to represent key aspects of the mineralization processes. In parallel, dissolution and precipitation can modify the hydrodynamical and mechanical properties of a reservoir, inducing permanent deformation and eventually failure, and significant modification of the storage volume and transport properties of the reservoir. However, experiments sometimes fail to reproduce natural processes due to the imposed time and scale constraints, and investigations on natural analogues or on pilot sites are also needed in order to unravel coupled complex processes.

This special volume includes studies of the bio-mechano-hydro-geochemical processes that arise from the interactions between CO<sub>2</sub> and minerals, wellbore cement or reservoir rocks. This collection of papers provides an overview of the current experimental work aimed at characterizing some of these research needs: the solubility and dissolution/precipitation rates of carbonate minerals in the laboratory at temperatures and CO<sub>2</sub> partial pressures representative of storage sites (Bénézech et al., 2009-this issue; Golubev et al., 2009-this issue; Pokrovsky et al., 2009a-this issue); the effects of organic and inorganic ligands (Pokrovsky et al., 2009b-this issue); the role of microbial activity (Dupraz et al., 2009a-this issue, b-this issue); and the nature and extent of mineral trapping of CO<sub>2</sub> associated with carbonation of silicates (Daval et al., 2009-this issue; Dufaud et al., 2009-this issue; Hangx and Spiers, 2009-this issue). Complementary to investigations at the mineral scale, batch experiments at the core scale allow assessment of the reactivity of reservoir rocks (Sterpenich et al., 2009-this issue) or cement (Regnault et al., 2009-this issue; Wigand et al., 2009-this issue) upon CO<sub>2</sub> injection and the induced geomechanical effects (Liteanu and Spiers, 2009-this issue). Hydrodynamic effects are investigated using reactive transport experiments and modelling (Luquot and Guze, 2009-this issue; Noiriél et al., 2009-this issue). Numerical modelling also enables the assessment of the interactions of CO<sub>2</sub> with rocks both at the pore-scale (Flukiger and Bernard, 2009-this issue) and at the reservoir-scale (Cantucci et al., 2009-this issue). For this purpose, new estimates of accessible surface areas of reactive minerals, based on image analysis are proposed (Peters, 2009-this issue). Field studies, including geological analogue (Boschi et al., 2009-this issue) and monitoring (Assayag et al., 2009-this issue) investigations, provide new constraints on the extent and timing of fluid-rock interactions in the presence of CO<sub>2</sub>.

The starting point for this special volume stems from a session of the EGU General Assembly 2007 in Vienna, Austria, co-organized by Pascale Bénézech, Bénédicte Ménez and Catherine Noiriél, entitled:

“CO<sub>2</sub> Geological Sequestration: bio–mechano–geochemical processes from the pore-scale to the reservoir-scale”. The compilation of papers in this special issue highlights the efforts that are made toward our better understanding of CO<sub>2</sub>–fluid–mineral interactions. The guest editors wish to thank all the reviewers for their contributions, their time and their insightful comments. Lastly, we would like to thank the Editor-in-Chief Jeremy Fein for his support of this special issue, and the Editorial Managers for Chemical Geology, Peggy Tan and Tim Horscroft for their assistance in all editing aspects of this project.

## References

- Assayag, N., Matter, J., Ader, M., Goldberg, D., Agrinier, P., 2009. Water–rock interactions during a CO<sub>2</sub> injection field-test: Implications on host rock dissolution and alteration effects. *Chem. Geol.* 265 (1–2), 227–235 (this issue).
- Bachu, S., 2000. Sequestration of CO<sub>2</sub> in geological media: criteria and approach for site selection in response to climate change. *Energy Convers. Manag.* 41 (9), 953–970.
- Bénézech, P., Dandurand, J.-L., Harrichoury, J.-C., 2009. Solubility product of siderite (FeCO<sub>3</sub>) as a function of temperature (25–250 °C). *Chem. Geol.* 265 (1–2), 3–12 (this issue).
- Boschi, C., Dini, A., Dallai, L., Ruggieri, G., Gianelli, G., 2009. Enhanced CO<sub>2</sub>-mineral sequestration by cyclic hydraulic fracturing and Si-rich fluid infiltration into serpentinites at Malenrata (Tuscany, Italy). *Chem. Geol.* 265 (1–2), 209–226 (this issue).
- Cantucci, B., Montegross, G., Vaselli, O., Tassi, F., Quattrocchi, F., Perkins, E.H., 2009. Geochemical modeling of CO<sub>2</sub> storage reservoirs: the Weyburn Project (Canada) case study. *Chem. Geol.* 265 (1–2), 181–197 (this issue).
- Daval, D., Martinez, I., Corvisier, J., Findling, N., Goffé, B., Guyot, F., 2009. Carbonation of Ca-bearing silicates, the case of wollastonite: experimental investigations and kinetic modeling. *Chem. Geol.* 265 (1–2), 63–78 (this issue).
- Dufaud, F., Martinez, I., Shilobreeva, S.N., 2009. Experimental study of Mg-rich silicates carbonation at 400 and 500 °C and 1 kbar. *Chem. Geol.* 265 (1–2), 79–87 (this issue).
- Dupraz, S., Ménez, B., Gouze, P., Leprovost, R., Bénézech, P., Pokrovsky, O.S., Guyot, F., 2009. Experimental approach of CO<sub>2</sub> biomineralization in deep saline aquifers. *Chem. Geol.* 265 (1–2), 54–62 (this issue).
- Dupraz, S., Parmentier, M., Ménez, B., Guyot, F., 2009. Experimental and numerical modeling of bacterially induced pH increase and calcite precipitation in saline aquifers. *Chem. Geol.* 265 (1–2), 44–53 (this issue).
- Flukiger, F., Bernard, D., 2009. A new numerical model for pore scale dissolution of calcite due to CO<sub>2</sub> saturated water flow in 3D realistic geometry: Principles and first results. *Chem. Geol.* 265 (1–2), 171–180 (this issue).
- Golubev, S., Bénézech, P., Schott, J., Dandurand, J.-L., Castillo, A., 2009. Siderite dissolution kinetics in acidic aqueous solutions from 25 to 100 °C and 0 to 50 atm pCO<sub>2</sub>. *Chem. Geol.* 265 (1–2), 13–19 (this issue).
- Gunter, W.D., Perkins, E.H., Hutcheon, I., 2000. Aquifer disposal of acid gases: modelling of water–rock reactions for trapping of acid wastes. *Appl. Geochem.* 15 (8), 1085–1095.
- Gunter, W.D., Wiwchar, B., Perkins, E.H., 1997. Aquifer disposal of CO<sub>2</sub>-rich greenhouse gases: extension of the time scale of experiment for CO<sub>2</sub>-sequestering reactions by geochemical modelling. *Mineral. Petrol.* 59 (1–2), 121–140.
- Hangx, S., Spiers, C.J., 2009. Reaction of plagioclase feldspars with CO<sub>2</sub> under hydrothermal conditions. *Chem. Geol.* 265 (1–2), 88–98 (this issue).
- Holloway, S., 1997. An overview of the underground disposal of carbon dioxide. *Energy Convers. Manag.* 38, S193–S198.
- IPCC, 2007. Climate change 2007: synthesis report. In: Pachauri, R.K., Reisinger, A. (Eds.), Contribution of working Groups I, II and III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. IPCC, Geneva, Switzerland. 104 pp.
- Liteanu, E., Spiers, C.J., 2009. Influence of pore fluid salt content on compaction creep of calcite aggregates in the presence of supercritical CO<sub>2</sub>. *Chem. Geol.* 265 (1–2), 134–147 (this issue).
- Luquot, L., Gouze, P., 2009. Experimental determination of porosity and permeability changes induced by massive injection of CO<sub>2</sub> into carbonate rocks. *Chem. Geol.* 265 (1–2), 148–159 (this issue).
- Noiriel, C., Luquot, L., Madé, B., Raimbault, L., Gouze, P., van der Lee, J., 2009. Changes in reactive surface-area during limestone dissolution: an experimental and modelling study. *Chem. Geol.* 265 (1–2), 160–170 (this issue).
- Oelkers, E.H., Schott, J., 2005. Geochemical aspects of CO<sub>2</sub> sequestration. *Chem. Geol.* 217 (3–4), 183–186.
- Peters, C.A., 2009. Accessibilities of reactive minerals in consolidated sedimentary rock: an imaging study of three sandstones. *Chem. Geol.* 265 (1–2), 198–208 (this issue).
- Pokrovsky, O.S., Golubev, S., Schott, J., Castillo, A., 2009a. Calcite, dolomite and magnesite dissolution kinetics in aqueous solutions at acid to circumneutral pH, 25 to 150 °C and 1 to 55 atm pCO<sub>2</sub>: new constraints on CO<sub>2</sub> sequestration in sedimentary basins. *Chem. Geol.* 265 (1–2), 20–32 (this issue).
- Pokrovsky, O.S., Golubev, S., Jordan, G., 2009b. Effect of organic and inorganic ligands on calcite and magnesite dissolution rates at 60 °C and 30 atm pCO<sub>2</sub>. *Chem. Geol.* 265 (1–2), 33–43 (this issue).
- Regnault, O., Lagneau, V., Schneider, H., 2009. Experimental measurement of portlandite carbonation kinetics with supercritical CO<sub>2</sub>. *Chem. Geol.* 265 (1–2), 113–121 (this issue).
- Sterpenich, J., Sausse, J., Pironon, J., Géhin, A., Hubert, G., Perfetti, E., Grgic, D., 2009. Experimental ageing of oolitic limestones under CO<sub>2</sub> storage conditions. Petrographical and chemical evidence. *Chem. Geol.* 265 (1–2), 99–112 (this issue).
- Wigand, M., Kaszuba, J.P., Carey, J.W., Hollis, W.K., 2009. Geochemical effects of CO<sub>2</sub> sequestration on fractured wellbore cement at the cement/caprock interface. *Chem. Geol.* 265 (1–2), 122–133 (this issue).

Pascale Bénézech  
*Université de Toulouse; UPS (OMP-CNRS-IRD); LMTG;*  
*14 Avenue Edouard Belin, 31400 Toulouse, France*  
 Corresponding author.  
*E-mail address: benezech@lmtg.obs-mip.fr.*

Bénédicte Ménez  
*Centre de Recherches sur le Stockage Géologique du CO<sub>2</sub>*  
*(IPGP/TOTAL/SCHLUMBERGER/ADEME); 4 place Jussieu,*  
*75005 Paris, France*  
*Equipe Géobiosphère Actuelle et Primitive, IPGP/IMPIC;*  
*75005 Paris, France*  
*E-mail address: menez@ipgp.jussieu.fr.*

Catherine Noiriel  
*Laboratoire Géosystèmes, Université de Lille 1, CNRS,*  
*59655 Villeneuve d'Ascq, France*  
*E-mail address: catherine.noiriel@univ-lille1.fr.*