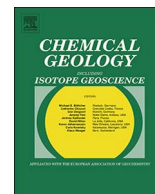




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Solute transport in porous media during drying: The chlorine isotopes point of view

I. Bernachot^{a,b,*}, B. Garcia^a, M. Ader^b, Y. Peysson^a, E. Rosenberg^c, G. Bardoux^b, P. Agrinier^b

^a IFPEN, Geosciences Division, GeoFluids and Rocks Department, 1-4 Avenue Bois Préau, 92852 Rueil-Malmaison, France

^b Institut de Physique du Globe de Paris, Equipe Géochimie Isotopes Stables, Sorbonne Paris Cité, UMR 7154 CNRS, F-75238 Paris, France

^c IFPEN, Physics and Analysis Division, 1-4 Avenue Bois Préau, 92852 Rueil-Malmaison, France

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ABSTRACT

Drying-out phenomena during injection of dry gas (CO₂, CH₄, H₂, ...) in geological reservoirs can be damaging for industrial processes as it can alter the transport properties and the injectivity of a reservoir due to salt precipitation. The distribution of salt precipitate, which may result in clogging of the formation, depends on two competing transport processes. Brine capillary-driven flows, which provide a continuous supply of dissolved salt towards the drying zone, and molecular diffusion, which tends to rehomogenize dissolved salt concentrations. In this study, we experimentally investigated the possibility that chlorine isotopes could be used as a geochemical tool to constrain the interplay of salt precipitation and solute transports process in drying saline porous media. Drying experiments were carried out on Lavoux carbonate plugs initially saturated with a 100 g L⁻¹ NaCl brine and dried in an oven at controlled temperature (60 °C) with vapor evacuation. Cl-content and δ³⁷Cl profiles along the length of the plugs were obtained for different evaporation stages ($S_w = 1$; $S_w = 0.82$; $S_w = 0.68$). The results show a clear Cl-ion redistribution during drying, with Cl-accumulation near the plug evaporative surface together with the growth of salt efflorescence on the surface. A “u”-shaped profile of δ³⁷Cl values is observed in the drying porous media. After 30 h of drying, salt crystals as well as the upper and lower parts of the plug present higher isotope ratios than the initial brine, while the center of the plug presents lower ratios. These experimental results suggest, as predicted by previous experimental and theoretical studies, that ³⁵Cl and ³⁷Cl are transported without fractionation by upward capillary flow to the evaporative surface, where salt precipitation occurs with a small preference for the heavier isotope. The resulting concentration gradient drives backward diffusion. Because ³⁵Cl moves faster than ³⁷Cl by diffusion this leads to more negative δ³⁷Cl towards the center of the plug. Cl-isotopes transport modeling results suggest that backward advection may also have occurred, as indicated by a relatively high effective diffusion coefficient and a low Cl-isotope fractionation factor. This study provides new insights regarding the use of Cl-isotopes to characterize drying of porous media.

1. Introduction

Water evaporation from saline porous media is of major concern in many cases such as soils salinization (e.g. Scanlon et al., 1997; Nachshon et al., 2011), preservation of building materials (Scherer, 1999; Gupta et al., 2014) and CO₂ geological sequestration (Pruess and Garcia, 2002). In geological storage context, injection of dry CO₂ can lead to complete water desaturation of the near wellbore region by evaporation of the formation water into the CO₂ phase (Pruess and Müller, 2009). This dry-out can precipitate salts from the solute initially dissolved in the brine, which may lead to gas injectivity impairment by reducing formation porosity and permeability, thereby threatening the viability of the industrial process (Peysson et al., 2010; Ott et al., 2015)

as reported for gas production wells in reservoirs of high salinity brine in Northern Germany (TDS from 150 to 340 g L⁻¹, Kleinitz et al., 2001; Kleinitz et al., 2003).

Because of the limited number of field-scale CO₂ sequestration projects, the behavior of salt precipitation during CO₂ injection have mostly been investigated through experimental and numerical studies. Several experiments of gas injection on cores under controlled drying conditions have shown that salt precipitation in the pore network can induce severe porosity and permeability reduction (Bacci et al., 2011; Peysson, 2012; Peysson et al., 2014; Roels et al., 2014). Numerical simulation of CO₂ storage in saline aquifers has also been used to assess the extension of the drying zone in the near wellbore region and to estimate the amount and distribution of salt precipitated (André et al.,

* Corresponding author at: IFPEN, Geosciences Division, GeoFluids and Rocks Department, 1-4 Avenue Bois Préau, 92852 Rueil-Malmaison., France.
E-mail address: i.bernachot@gmail.com (I. Bernachot).

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2014; Giorgis et al., 2007; Pruess and Müller, 2009; Zeidouni et al., 2009). For example, André et al. (2014) have shown that salt can be deposited within a radius up to a dozen meters around the well with significant consequences on porosity and permeability of the geological formation, depending on various parameters such as brine salinity, reservoir structure but also gas injection rate. In these studies, the mechanisms affecting desiccation of porous media are usually described as follows: when dry supercritical CO₂ is injected in an initially saturated porous medium, the brine is displaced away from the injection zone resulting in a drying front moving into the medium, while the residual trapped water evaporates precipitating salts in the dry zone. These processes are counter-balanced by capillary flows initiated by the water saturation gradient, which transport the brine towards dryer zones. This results in a continuous advective flow of dissolved salt, which precipitates in the near wellbore zone as water is evaporated by the ongoing injection of CO₂ (Giorgis et al., 2007; André et al., 2014). Salt precipitation can therefore be substantial not only in high salinity reservoirs but also in lower salinity reservoirs due to salt accumulation (Pruess and Müller, 2009). Several studies have shown that the amount and distribution of precipitated salt depends on brine salinity, water transfer into the CO₂ phase and solute transport. For example, Ott et al. (2011) have identified a gas injection velocity threshold above which salt precipitates homogeneously inside the porous medium, whereas for lower velocities, capillary-induced back flow of the remaining brine transports solutes towards the injection zone, where salt eventually precipitates. More recently, Peysson et al. (2014) and André et al. (2014) have highlighted the importance of the evaporation rate on salt distribution. Indeed, when evaporation rate is low, solutes accumulate close to the injection point but capillary fluxes are weak so that backward diffusion tends to homogenize their distribution throughout the porous medium. When the evaporation rate is high, solutes are transported by massive capillary flows towards the gas injection zone where they locally precipitate when saturation concentration is reached as diffusive flows are weaker.

These observations are similar to the processes described in studies focusing on solute transport in “static” evaporating systems (without gas injection), such as soils or building materials for example, where salt was found to precipitate preferably at or in a region near the surface of the porous material in contact with air (Huinink et al., 2002; Sghaier et al., 2007; Guglielmini et al., 2008; Shokri and Or, 2011). In several studies, models based on the advection-diffusion equation have been applied to understand solute transport within porous structures. They have shown that under common evaporative conditions, convective capillary flows can induce greater concentration at the surface of the porous medium where salt can precipitate when solubility is reached (Eloukabi et al., 2011). Non-destructive techniques such as Nuclear Magnetic Resonance (NMR) (Pel et al., 2000; Faure and Coussot, 2010; Peysson et al., 2011b) and imaging techniques, such as neutron radiography (Shokri et al., 2008) and synchrotron X-ray tomography (Shokri, 2014), have been used to investigate the spatial liquid, solute and salt distributions in porous media during evaporation.

Drying behavior of porous media has therefore been well conceptualized through a large number of experimental studies and numerical models. Relying on this knowledge, it can be considered using tracers as indicators of drying process in natural systems. In this study, we investigate experimentally the possibility that chlorine isotopes could be used as a geochemical tracer to constrain dynamics of solute transports and salt precipitation during desiccation of porous media, with further application to CO₂ industrial storage. Chlorine, occurring as chloride, is indeed widely assumed to be unreactive in surface and subsurface aqueous environments, and $\delta^{37}\text{Cl}$ is already used as a tracer of solute transport, especially diffusion (Desaulniers et al., 1986; Eggenkamp and Coleman, 2009; Beekman et al., 2011) and salt precipitation (Eggenkamp et al., 1995; Eastoe et al., 1999; Luo et al., 2014; Eggenkamp et al., 2016) so that it is well

designed for characterising complex solute transport processes in drying porous media. Diffusion in aqueous systems is known to fractionate chlorine isotopes by up to 2‰, while advection produces no isotopic fractionation (e.g. Eggenkamp and Coleman, 2009). Chlorine isotopes have thus been used in several studies to characterize the hydrodynamics in natural systems (e.g. Coleman et al., 2001; Lavastre et al., 2005; Beekman et al., 2011). As to salt precipitation, a few experimental studies investigated equilibrium fractionation between salts and their saturated solutions (Eggenkamp et al., 1995; Luo et al., 2012, 2014; Eggenkamp et al., 2016). But to our knowledge, the complexity of the interplay between solute transport and salt precipitation remains to be investigated.

As the same solute transport mechanisms are implied when water evaporates in the air by vapor diffusion or in the injected dry gas (Peysson et al., 2011a), experiments consisting in drying brine saturated carbonate plugs in controlled conditions were carried out in the present study. Different saturation stages were investigated in order to assess chloride content and isotopic profiles inside the plug samples during drying. The results show a clear evolution of chloride distribution and stable isotope composition inside the plug during water evaporation. A simplified numerical model was used to identify the respective influences of transport processes (advection and diffusion) and salt precipitation in the observed variations. The first order consistency between analytical results and modeling suggests that indeed chlorine isotopes have high potential for the tracing of hydrodynamics in drying porous media. Provided that more detailed solute transport models can be performed to take into account the complexity of the processes occurring during evaporation from porous media, chlorine isotopes can be very efficient tracers of salt precipitation during dry gas (CO₂, CH₄, H₂, ...) industrial injection.

2. Experimental methods

2.1. Rock samples description

Samples of Lavoux carbonate were used in this study to represent a reservoir rock for gas storage. Lavoux carbonate is a Callovian (Middle Jurassic) white oolitic limestone from Paris Basin, France. It was selected for these experiments because of its particularity of being homogeneous and mostly composed of calcite (99%) (Auffray et al., 2016). Previous High Pressure Mercury Injection (HPMI) analyses have shown that Lavoux carbonate samples appear to have dual porosity (macro- and microporosity) and dual pore throat size distribution, with peaks at 0.26 μm and 10 μm (e.g. Grgic, 2011). Permeability is about 100 mD.

The experiments were performed on four cylindrical plugs (Lx-1 to Lx-4) of diameter 2 cm and length 10 cm cored from a same quarry block in the direction perpendicular to the horizontal bedding of the material. Sample porosity was comprised between 23% and 26%, as measured by water content at saturation (Table 1). As our plugs were extracted from the same homogeneous block, we assume here that they share the same petrophysical properties.

Table 1
Porosity measured by weight of dry and 100 g L⁻¹ NaCl brine-saturated samples ($\phi = \frac{M_{\text{sat}} - M_{\text{dry}}}{\rho \times V_{\text{tot}}}$).

Sample	Dry mass (g)	Saturated mass (g)	$M_{\text{sat}} - M_{\text{dry}}$ (g)	Calculated porosity
Lx-1	61.61	69.89	8.28	0.23
Lx-2	61.76	70.10	8.34	0.25
Lx-3	61.93	70.45	8.52	0.26
Lx-4	61.71	69.90	8.19	0.25

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