



Experimental nucleation and growth of smectite and chlorite coatings on clean feldspar and quartz grain surfaces



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ABSTRACT

Laboratory experiments were performed to investigate high-temperature (100–150 °C) authigenic formation of clay coatings (smectites, chlorites) on clean feldspar and quartz surfaces. Artificial formation waters were used, with Mg-concentrations in the range of North Sea brines. Experiments were run for 21–50 days in brines composed of magnesium chloride and sodium carbonate adjusted to circum-neutral pH. The experiments suggest that the silica activity is the main factor determining if grain coating smectites or chlorite form. The clay minerals form easily on both clean quartz and feldspar surfaces, but chlorite coatings were formed only on feldspar surfaces while smectite coatings were formed on both feldspar and quartz. The chlorite morphology varies between honeycomb, edge-to-face and rosette patterns, while all smectite formed with honeycomb-like textures. The clay coatings produced in this study are morphologically similar to naturally occurring diagenetic clay minerals. In natural sediments and sedimentary rocks, the formation of clay coatings is promoted by pre-existing clay drapings on mineral grains. The growth substrates used in this study were not coated with such a natural precursor materials. This suggests that the nucleation of clay coatings in nature may be possible on clean quartz and feldspar surfaces, but pre-existing clay minerals may impact the allowed levels of supersaturations and thereby the formation of the grain-coating phases.

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1. Introduction

Quartz cementation is one of the main porosity reduction mechanism in siliciclastic reservoir sandstones (Bjørlykke, 1979; Bjørlykke and Egeberg, 1993; Houseknecht, 1987; Leder and Park, 1986; McBride, 1989; Worden and Morad, 2000). Grain coatings of chlorite, smectite, mixed-layer illite/smectite, illite, mixed-layer chlorite/smectite, and micro quartz may however inhibit quartz formation and thereby preserve reservoir quality to great burial depths (Aase et al., 1996; Bloch et al., 2002; Ehrenberg, 1993; Heald and Larese, 1974; Pittman et al., 1992; Storrø, 2002). The above mentioned scientific literature documents the inhibition of quartz overgrowth due to clay and micro quartz coatings and its effect on petro-physical parameters such as porosity and permeability in siliciclastic hydrocarbon reservoir rocks.

The extent and mode of the clay-coat coverage is very crucial for the preservation of porosity. With higher coat coverage, less quartz cement forms (Heald and Larese, 1974; Walderhaug, 1996). Coatings

do not always prevent formation of quartz overgrowths on sand grains with unique features such as fracturing of sand grains at coatings (Heald and Larese, 1974). Such type of interruption of the continuity of coatings will expose quartz surfaces for nucleation to form new quartz overgrowths. Hence, understanding and characterizing these features may give important information on how the coating of quartz grains preserves high porosity at deep burial depth. The occurrence of clay coats, the extent of the coating, and its relevance to preservation of porosity in sandstones, are discussed in detail by Pittman et al., 1992 and Doney et al., 2012. They show that there is a depositional environment control on the occurrences of clay coats and the formation of different types of authigenic clays (Pittman et al., 1992; Doney et al., 2012). For example, authigenic chlorite coats were observed in fluvial channels in Berea sandstones, corrensites coats were observed in distributary channel and mouth-bar facies and smectite was observed as a dominant clay in various near shore marine facies (Almon et al., 1976; Pittman et al., 1992). Moreover, Pittman et al. (1992) noted that quartz cementation may be hindered well if the clay coats are thick, well-developed and continuous. One possible explanation for inhibition of quartz cement is that grain-coating chlorites and smectites have structures and orientations that imposes an energy barrier for quartz

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nucleation, while the energetically favorable detrital quartz surfaces are coated and not available for nucleation (Aagaard et al., 2000; Jahren et al., 1998; Pittman et al., 1992; Walderhaug, 2000). Clay minerals that exist in sedimentary rocks may be detrital or authigenic. Authigenic clay minerals are formed *in situ* while detrital clay minerals are transported from a given source area as a result of weathering and erosion and deposited in its present day environmental setting. Authigenic clay minerals in sandstones may be formed as a result of direct precipitation from solution due to dissolution of very labile mineral grains and/or transformation of some precursor clay minerals (Hillier, 1995; Wilson and Pittman, 1977). The factors controlling the formation of clay mineral coatings on detrital sediments nevertheless still remain poorly understood. Precursor clay minerals in sandstones may have originated from mechanically infiltrated clays, inherited clay rims, bioturbation clays or adhesion clays (Matlack et al., 1989; McLroy et al., 2003; Needham et al., 2004, 2005; Wilson, 1992; Wilson and Pittman, 1977). Clay minerals may be introduced into the sediments just shortly after deposition as a result of bioturbation, deep infiltration of muddy waters as a result of water table fluctuations, tidal pumping in tidal dominated environments, partial saturation and dry out in arid climates (Al-Ramadan et al., 2013; Herringshaw and McLroy, 2013; Houseknecht, 1992; Walker, 1976; Walker et al., 1978; Worden and Morad, 2003; Worden et al., 2006). Effective superpositioning of mechanically infiltrated clays on sand grains are common in tidal estuaries, fluvial channels, point bars and delta plain deposits. Inherited clay rims may stick on framework grain surfaces before reaching the depositional environment. Such type of clay rims are common in eolian settings and marine shelf environments (Wilson, 1992). In the eolian environments inherited clay rims may be formed due to muddy-water infiltration or adhesion of clays on wetted sand grain surfaces. Depositional facies highly controls the abundance, distribution and presence of bioturbated, inherited and infiltrated clays in sediments. Therefore, including the depositional environment is very crucial to have comprehensive understanding of how clay coats form.

The origin of authigenic clay mineral coatings can be due to direct precipitation from aqueous solution (neof ormation) and without the aid of precursor clay materials. Such a process requires unstable rock fragments, biogenic silica, biotite, detrital heavy minerals etc. (Anjos et al., 2003; Bahlis and De Ros, 2013; Humphreys et al., 1989; Pittman et al., 1992; Thomson, 1979). Feldspar, used as a substrate and source of Al and silica in this study, commonly transforms into illitic clay and kaolinite in diagenetic environments. However, despite being an important diagenetic process, chloritization of feldspar is rarely reported in the literature and thus little is known about it (Morad, 1984; Moraes, 1992; Walker et al., 1978). However, Morad and Aldahan (1987) documented the transformation of detrital feldspar into chlorite in sandstones of Visingsö (Upper Proterozoic, southern Sweden).

Hydrothermal reactions performed on natural sandstone samples from the Oseberg and Veslefrikk fields offshore Norway produced chlorite overgrowths on reservoir sand originally coated with berthierine precursors (Aagaard et al., 2000). Aagaard et al. (2000), however, did not investigate the mechanism of the conversion of berthierine into chlorite, and the detailed mechanism still remains elusive (Morad et al., 2003). Previous findings, in hydrothermally formed chlorites (Cathelineau and Nieva, 1985) and diagenetically formed chlorite (Jahren and Aagaard, 1989) support a temperature controlled chemical variations in chlorites. A second-order related growth mechanism was proposed based on chlorite particle size distribution profiles for coarsening of the chlorite crystals as a function of increasing burial depth (Jahren, 1991).

In this study clay coatings formed in batch reactors were compared to minerals formed in natural systems based on amount,

surface coverage, crystal morphology, crystal chemistry, etc. The experiments were done at 100–150 °C with feldspar and quartz as growth substrates. The produced clay coatings were compared to clay coats formed in Intra-Draupne Sandstones from the South Viking Graben. Feldspar and quartz were selected as substrates in this study because they are diagenetically significant minerals in siliciclastic sandstone reservoirs. Additionally, they are the most abundant minerals in the earth's continental crust.

The purpose of this study was:

- 1) To study the effect of changing the bulk solution composition and temperature upon the formation of clay-coatings on feldspar and quartz surfaces
- 2) To investigate the role of silica activity on the formation of clay coatings (smectites, chlorites)
- 3) To acquire data on clay coating formation rate for use as input to modeling of clay-coatings in siliciclastic reservoir sandstones

2. Methods and materials

2.1. Experimental setup

In this study, pure quartz grains and large perthitic feldspar grains with cleavage planes was used as substrates for secondary clay nucleation and growth. The compounds used to prepare the reactive solutions were reagent grade $MgCl_2$, $CaCl_2$, Na_2CO_3 , and HCl dissolved in high purity Milli-Q water. Na_2CO_3 was added to provide CO_2 pressures of 0.2–1 bars, which is within the range encountered in sedimentary systems (Aagaard and Egeberg, 1998). The initial experimental solutions had Mg contents in the range of high-temperature North Sea formation waters (Fig. 2). High NaCl background electrolyte concentrations were not used in order to avoid problems in the post-experiment analyses of the reacted solutions. The batch experiments were run for 21–50 days at temperatures of 100 and 150 °C at vapour pressure conditions ($P_{H_2O} + P_{CO_2}$) and with different bulk solution compositions (Table 1). A polypropylene non-stirred batch hydrothermal reactor

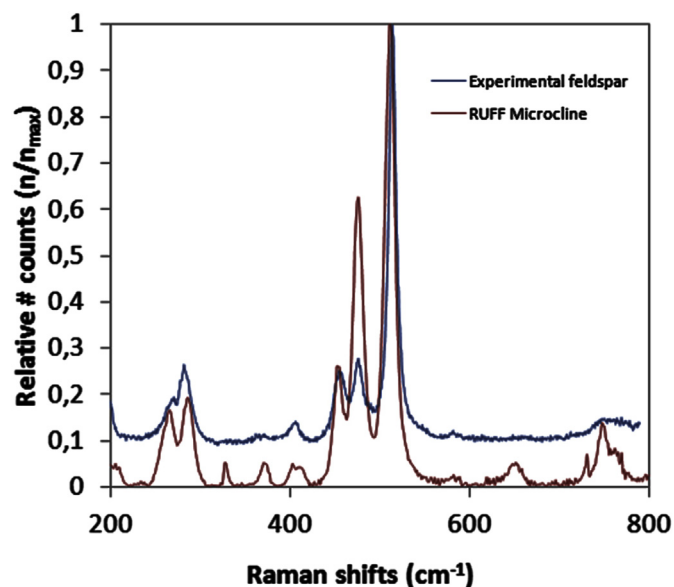


Figure 1. Raman spectrum of feldspar well fitted with microcline spectrum found from RRUFF data base.

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