



Adhesive forces between two cleaved calcite surfaces in NaCl solutions: The importance of ionic strength and normal loading

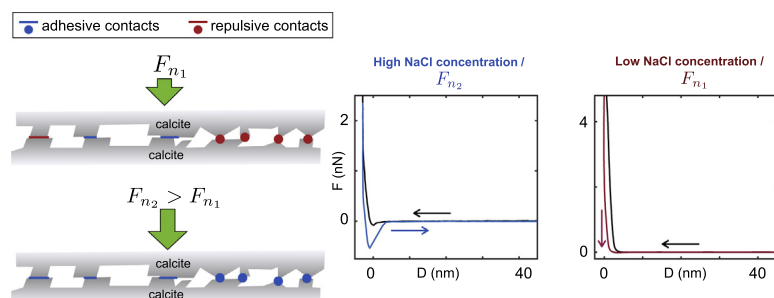
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GRAPHICAL ABSTRACT



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ABSTRACT

The mechanical strength of calcite bearing rocks is influenced by pore fluid chemistry due to the variation in nano-scale surface forces acting at the grain contacts or close to the fracture tips. The adhesion of two contacting surfaces, which affects the macroscopic strength of the material, is not only influenced by the fluid chemistry but also by the surface topography. In this paper, we use Atomic Force Microscope (AFM) to measure the interfacial forces between two freshly cleaved calcite surfaces in CaCO_3 -saturated solutions with varying NaCl concentration. We show that calcite contacts become stronger with increasing NaCl concentration (>100 mM), as a result of progressively weaker secondary hydration and increasing attraction due to instantaneous ion-ion correlation. Moreover, we discuss the effect of normal applied force (F_n) and surface roughness on the measured adhesion forces (F_{ad}). We show that the measured pull-off force (adhesion) is linearly correlated with the magnitude of F_n , where an increase in applied force results in increased adhesion. This is attributed to a larger number of contacting surface asperities and thus increase in real contact area and the contact-bond strength. We discuss that the possible variation in local topography at contacts, together with strong dependence on ionic strength of the solution, can explain the inconsistent behavior of calcite rocks in NaCl solutions.

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1. Introduction

Calcite is an abundant mineral in nature. It is a crystalline polymorph of calcium carbonate with a cleavage plane along the $(10\bar{1}4)$ direction [1–3]. Calcite plays a key role in biomineralization and it is a constituent of shells and skeletons of many marine

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invertebrates [4]. Moreover, it is one of the most common rock-forming minerals of importance to hydrocarbon recovery, CO₂ sequestration [5,6] and nuclear waste storage [7]. Calcite is the main constituent mineral of chalk (>99%). Chalk deposits form many of the world's oil and gas reservoirs, such as the North Sea oil reserves, where they alone account for 25 million barrels of oil since the 1970s [8].

Chalk reservoirs are prone to strong compaction, due to water injection associated with Enhanced Oil Recovery (EOR) projects [9–13]. The mechanical behavior of chalk and calcite-bearing rocks is known to be influenced by the pore fluid chemistry [9,14–17], an effect that is often referred to as water-weakening (the significant loss of mechanical strength of chalk in water-saturated rocks [12,18]). Several mechanisms have been proposed to describe this phenomenon, such as pressure solution [19,20], chemical influences [19], invading the capillary bridges/menisci by water flooding [21], time-dependent water adsorption on calcite surfaces [11,22] and subcritical crack growth at the grain boundaries [23]. In the early 2000s, Risnes et al. [12,18] proposed that water activity is a key parameter behind the strength loss in chalk in aqueous solutions. Hellmann et al. [13] suggested that water-weakening may be also related to the repulsive forces due to adsorbed water molecules on adjacent calcite surfaces. These two hypotheses were further supported by atomic force microscope (AFM) experiments by Røyne et al. [24], in which adhesion between two surfaces depended on water activity, with strong repulsion measured in pure water.

At the nano-scale, repulsive and attractive forces operate between two calcite surfaces that are separated by a thin fluid film. Surface forces between two charged surfaces in an electrolyte solution can be described by the Derjaguin-Landau and Verwey-Overbeek (DLVO) theory [25,26] that includes van der Waals (vdW) and Electrical Double Layer (EDL) forces [27,28]. However, at separations significantly shorter than the Debye length, and for solutions with high ionic strength (>0.1 M) [28,29], where specific ion interactions and hydration effects become exceedingly important, [30–36] the DLVO theory cannot accurately describe the interaction between surfaces in aqueous solutions. The AFM direct force measurements by Røyne et al. [24] shows that the observed repulsion in water is due to hydration forces acting between two hydrophilic calcite surfaces. A similar experiment by Pourchet et al. [37] indicates that attractive forces act between calcite surfaces in high pH and higher ionic strength solutions (0.12 M), which were attributed to the ion-ion correlation forces. Both hydration and ion-correlation forces are not included in the DLVO theory.

Several studies have shown that the degree of water weakening is also affected by the salinity of the pore fluid [12,18,38]. The salinity of the solution affects both the EDL component of the DLVO forces, and the water activity [39,40]. It also changes the calcite dissolution kinetics in aqueous solutions [41–44].

In general, calcite and other natural mineral surfaces display some degree of roughness at a molecular scale. Several studies have shown that surface roughness affects the water wettability of calcite [45,46] and oil desorption from calcite surfaces [47]. It, also, influences the interfacial forces between mineral surfaces in molecular scale. This is because of the actual contact area is always smaller than the nominal surface area (see Fig. 1) [48–57]. For rough surfaces, contacting surface asperities give rise to an exponentially decaying repulsive force upon loading [58], which can potentially be interpreted as hydration repulsion, as in the past studies [58,59].

Calcite surfaces are dynamic in aqueous solutions, with continuous dissolution and recrystallization on the timescale of hours even in saturated solutions, as shown by Stipp et al. [60,61]. We, therefore, expect the distribution and geometry of surface asperi-

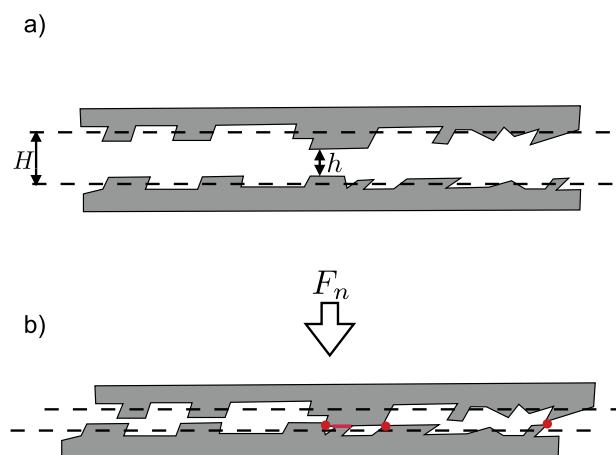


Fig. 1. A simple sketch of two opposing, cleaved calcite surfaces with nano-scale roughness characterized by steps and terraces on the (1 0 1 4) surface. (a) Dotted lines represent the midline of surface asperities with H as the surface separation, and “ h ” is the distance between highest asperities. (b) When two surfaces pushed into contact by an applied normal load (F_n), a discrete number of asperities are forced into contact, as represented by the red dots and lines. The sum of these discrete areas of contact are referred to the actual area of contact for rough, contacting surfaces. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

ties to change with time, which in turn influence the magnitude of repulsive mechanical effects due to asperity deformation. Recently Dziadkowiec et al. [62] performed force measurement experiments between two rough calcite surfaces (with nm-scaled asperities), using the Surface Force Apparatus (SFA). They observed repulsive forces with a decay length that increases with time, which was explained by repulsive hydration forces combined with continuous recrystallization and roughening of the calcite films in saturated CaCO₃ solutions.

The effect of NaCl on the interaction between calcite surfaces is of interest because both Na⁺, and Cl[−] are the two most abundant monovalent ions in seawater. The influence of NaCl on the strength of carbonate rocks [15,16,63] and EOR systems has been extensively investigated in surface science and reservoir engineering, e.g. [64–66]. Liu et al. [64] observed that injection of NaCl solution (low and high concentrations) into a carbonate rock increases the oil desorption rate from calcite surfaces. They explained this result by increased solubility of calcite in high concentration NaCl solution, which in turn increases the local pH, leading to more negatively charged calcite surfaces and hence repulsive forces between the calcite and the oil. For low NaCl concentrations, they relate the high-rate oil desorption to the EDL repulsive forces between oil and calcite surfaces. Interestingly, in contrast, Fathi et al. [65,66] showed that oil recovery improves when the NaCl (named as non-active salt) is removed from the seawater. This effect was attributed to a high population of Na⁺ and Cl[−] near the calcite surfaces that prevents the potential determining cations/anion (Mg²⁺, Ca²⁺ and SO₄^{2−}) to reach to the surface. As a result, a more positively charged calcite surface attracts oil to a higher extent. However, they show that surface reactivity and ultimately wettability of the surface varies with the temperature as well as the solution ionic strength. This shows the increased complexity of the calcite-brine-calcite system once the oil is present. In fact, the type and history of mineral surfaces, and the components of oil and brine are inevitable parameters and shall thus be considered when investigating such a system.

In this study, we aim to understand the role of ionic strength in compaction of calcite-bearing rocks, and its potential relation to the nm-range forces between calcite surfaces. To achieve this, we use the colloidal probe AFM with a calcite probe against a freshly

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