



# Dissolution of arkose in dilute acetic acid solution under conditions relevant to burial diagenesis



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## ARTICLE INFO

### Article history:

Available online 20 January 2015

Editorial handling by Mats Åström

## ABSTRACT

The organic acid produced during diagenesis can cause mineral dissolution, leading to the formation of secondary porosity zones. Arkose dissolution in organic acid solution is important in the evolution of reservoir quality, and is influenced by various factors. To investigate the effects of acetic acid on arkose dissolution under different burial diagenetic conditions, seven batch experiments were conducted over a wide range of reaction conditions. Medium-grained lithic arkose from Songliao Basin, northeastern China, was selected for the study. Four primary factors were considered: temperature (80 °C, 120 °C, and 160 °C), pH (2.5, 4.0, and 5.5), initial water/rock mass ratio (10:1 and 15:1) and grain size (1–3 mm, 3–5 mm). Each experiment was performed for 312 h and sampled six times in parallel. Results indicate that these four factors have significant effects on arkose dissolution. In a certain temperature range, feldspar dissolution increased, but declined when the temperature exceeded a certain value. Different minerals/elements were affected differently by temperature. Silicon was the element most sensitive to temperature. Under acidic conditions, the dissolution rates of most minerals increased as the pH decreased. Grain size and water/rock mass ratio affected the degree of reaction in the early period and then the form of mineral transformation in the late period. The experimental results are generally consistent with previous field research outcomes. Understanding the factors that control sandstone dissolution under organic acid conditions will allow the easier detection of secondary porosity zones, which may improve the precision of oil exploration.

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## 1. Introduction

Previous studies have shown that large quantities of organic acids can be produced during diagenesis (Surdam et al., 1989; Barth et al., 1990; Ehrenberg and Jakobsen, 2001; Feng, 2008). With a mature hydrocarbon source rock, much organic acid can intrude into the sandstone reservoir, causing a series of chemical reactions (Manning et al., 1992; Welch and Ullman, 1996; Cama and Ganor, 2006; Fan et al., 2009). Organic acids can change the kinetics and reaction mechanisms of mineral dissolution via its surface complexation with metal ions (Huang et al., 1995, 2009). Undoubtedly, they greatly influence reservoir quality (e.g., porosity and permeability) and the secondary pores of sedimentary rocks, which are highly significant in oil and gas exploration (Surdam et al., 1989; Barth et al., 1990; White and Brantley, 1995; Wei et al., 2006; Zeng et al., 2006; Ganor et al., 2009; Pokrovsky et al., 2009; Vandeginste et al., 2009; Liermann et al., 2011; Shao et al., 2011; Ma et al., 2012; Declercq et al., 2013).

However, much exploratory research has shown that secondary pores only occur at specific depths and with specific periods of diagenesis. Thus, the diagenetic environment controls the development and evolution of secondary pores. Although organic acids have positive effects on mineral dissolution, especially on arkose, this dissolution is influenced by many factors, including temperature (Liu et al., 2003; Cama and Ganor, 2006; Ptáček et al., 2010; Gong et al., 2012; Rosenbauer et al., 2012), pH (Bevan and Savage, 1989; Stillings et al., 1996; Welch and Ullman, 1996; Köhler et al., 2003; Daval et al., 2013; Yang et al., 2014), and the solution (Manning et al., 1992; Blake and Walter, 1999; Allan et al., 2011). These factors significantly control the dissolution behaviors of feldspars in different stages of diagenesis. Gong et al. (2012) determined the kinetic data for the dissolution of arkosic sandstone powders in deionized water at different temperatures (50–350 °C). Their results showed that Na, K, Ca, Al, and Mg were the major ions in solution at low temperatures, whereas Si was the major ion retained at higher temperatures. Therefore, there exists some kind of functional relationship between temperature and the dissolution rates of arkosic sandstone powders. Allan et al. (2011) carried out a series of kinetic experiments at 25 °C and 80 °C using 0.01, 0.1, and 1 M NaCl solutions at pH 3 to investigate

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the dissolution rates of common sandstone minerals in response to the acidification of pore water. Their results revealed that chemical affinities differed under different conditions and that reservoir rocks rich in feldspar, illite, and/or smectite were likely to react most rapidly with acidified pore water.

In this study, dilute pure acetic acid solution was used to avoid the interference of initial ions. The effects of four factors (temperature, pH, the water/rock mass ratio, and grain size) were considered in designing experiments for this sensitivity analysis. Seven groups of experiments were conducted over a wide range of conditions. Understanding the main factors controlling arkose dissolution in organic acid solution should help to explain the occurrence of zones of anomalously high porosity and secondary pores.

## 2. Experimental materials and methods

### 2.1. Materials

Songliao Basin is located in northeastern China and hosts several oil and gas combinations. Yingcheng Formation is one of the main targets of deep natural gas exploration in Songliao Basin (Feng et al., 2013; Zhang et al., 2013b). A large number of studies have shown that there are two main types of sandstone in the Yingcheng Formation of Songliao Basin, lithic feldspar sandstone and feldspar lithic sandstone (Shao, 2005; Li, 2009; Liu et al., 2012). In different diagenetic stages, organic acid intrudes into the reservoir at different rates. This triggers dissolution of silicate and carbonate minerals and produces secondary porosity, which is very important in oil and gas exploration (Shao, 2005; Tian et al., 2007; Meng et al., 2010b, 2011; Liu et al., 2012).

In sedimentary basins, the reservoir porosity is usually controlled by sedimentary facies and diagenesis. In Songliao Basin, porosity is mainly influenced by sedimentary facies in the early period, whereas diagenesis influences it in the late period (Qu et al., 2005; Liu et al., 2012). Therefore, with the development of many secondary pores, the correlation between porosity and depth is poor. Previous studies have shown that several vertical secondary porosity zones exist and many geologists consider that they were formed during the dissolution of feldspar and other mineral cements by organic acids (Meng et al., 2010a; Mi et al., 2010; Jia et al., 2013; Sun et al., 2013a,b; Zhang et al., 2013a). The secondary porosity zone correlates well with the intrusion of organic acid, and the degree of secondary porosity development correlates positively with the organic acid concentration (Shao, 2005; Liu et al., 2010; Meng et al., 2010b, 2011).

In this study, a block of rock of Lower Cretaceous age, about 50 cm × 50 cm × 50 cm, was obtained from an outcrop of Yingcheng Formation in Songliao Basin. The rock was mainly composed of feldspar, quartz, and lithic fragments, with small amounts of mica, identified with thin-section identification, X-ray Diffraction (XRD) and X-ray Fluorescence (XRF). The mineral composition of the arkose rock analyzed with XRD is shown in Table 1. To avoid the influence of mineral heterogeneity, six randomly selected parts (called samples here) from the rock block were measured. Each mineral was given a value range, and feldspar was dominant. Thin-section imaging revealed that most of lithic fragments were volcanic and most mineral grains were of medium size (Fig. 1). By SEM, authigenic albite with regular plate-like crystal can be observed. The rock was designated a medium-grained lithic arkose, by definition.

Acetic acid is very common in oil field water (Barth and Riis, 1992; Mayumi et al., 2011). Therefore, dilute acetic acid was chosen as the reaction solution. Dilute pure acetic acid at different acidities was produced by adding distilled water, to avoid any change in the original ionic composition.

**Table 1**  
Modal mineralogy of the arkose sample.

	Quartz	Plagioclase	Alkali-feldspar	Calcite	Muscovite	Kaolinite
Wt.%	14–17	30–40	34–38	10–15	0–7	0–4

### 2.2. Methods

The experiments were conducted in several uniform high-temperature airtight reaction kettles, with liners made of polytetrafluoroethylene. The advantages of the kettles are their resistance to acid and alkali corrosion with a maximum tolerable temperature of 260 °C. The volume of reaction kettle is 100 mL. Other instruments, such as the incubator chamber, electronic scales, and pH meters, were also used.

Because four factors (temperature, pH, water–rock mass ratio, and grain size) were considered, the experiments were designed with different reaction conditions (Table 2). Experiment A (Ext. A) was the baseline experiment and the other experiments were derived from it. In Ext. B, the grain size was altered from 3–5 mm to 1–3 mm to investigate the effect of grain size on the reaction. Because the effects of acidic conditions were examined in this study, the pHs were altered in Exts. C and D, and in both, the organic acid was below neutral pH 7.0. The differences in the reactions at three different acidities were determined by comparing the results of Exts. A, C, and D. In Exts. E and F, the temperature was changed from 120 °C to 80 °C and 160 °C, respectively. The different temperature conditions (80 °C, 120 °C, and 160 °C) roughly represent the conditions from early burial diagenesis to late burial diagenesis and the stages of organic matter evolution from immature to highly mature. Groundwater flows slowly in a sedimentary basin, especially in deep formations, and mineral transformation processes proceed for millions of years. The laboratory cannot simulate such a large time scales. To improve the reaction rate, the capacity of the solution was increased. In Ext. G, the water/rock mass ratio was increased from 10:1 to 15:1. For a solution of approximately 70 g (71.4 mL), 7 g of rock were required when the water/rock mass ratio was 10:1, whereas a 90 g (91.7 mL) solution and 6 g of rock were required when the ratio was 15:1.

According to the water/rock mass ratio in Table 2, a certain quality of sample and dilute acetic acid were put into the kettle. After being sealed, the kettle was put into the incubator chamber with specific temperature. The static batch experiment began. The saturated vapor pressure was the reaction pressure. These experiments were conducted for 13 days (312 h), with sampling on the 2, 4, 6, 8, 11, 13 days. The experiments were performed in parallel, and six high-temperature airtight reaction kettles were used in each experiment, corresponding to six samples (2, 4, 6, 8, 11, 13 days). After experiment, all rock samples are tested by XRD with each sample of 1 g and XRF with each sample of 2 g after they were dried and weighed. Scanning electron microscopy (SEM) was used to analyze mineral dissolution/precipitation. The concentrations of conventional ions (K, Ca, Na, Cl, and Si) in each solution were measured with atomic absorption spectroscopy, inductively coupled plasma mass spectrometry, and ultraviolet spectrophotometry. The initial concentrations of the conventional ions were almost zero, so the changes in the ion concentrations effectively reflect the dissolution and/or precipitation of the minerals.

## 3. Results

### 3.1. Different temperature

The results of Exts. A (120 °C), E (80 °C), and F (160 °C) were analyzed comparatively to examine the different changes that

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