



Source of diagenetic fluids from fluid-inclusion gas ratios



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ABSTRACT

Alteration of marine carbonates, such as those from the Mesoproterozoic and Lower Ordovician, during burial history is carried out by diagenetic fluids that could potentially have different sources. During precipitation diagenetic minerals may trap tiny amounts of those solutions in fluid inclusions, thus providing fossil archives and aliquots that offer valuable record about the origin of fluids. The analysis of various fluid-inclusion gases (e.g., CH₄, CO₂, N₂, He and Ar) may provide reliable information which allows the discrimination among different fluid sources. Generally speaking, the meteoric cements should have N₂/Ar ratio around 38 (the value for air-saturated water). However, the gases in inclusions of the current study, trapped by diagenetic carbonates (cements), from independent depositional basins of different ages (Mesoproterozoic, Early Ordovician, and Modern) and locations (Canada, USA, Brazil and Java Sea), have N₂/Ar ratios (~92) which match non-magmatic (modified marine or meteoric) fluids. Also, the Ar/He ratios (~3) in the studied fluid inclusions are significantly lower than those of calc-alkaline magmatic fluids (≥20), thus excluding magmatic fluids as a potential source. On the other hand, some N₂/Ar and Ar/He ratios of the studied samples approach values reported for basinal fluids from the Hansonburg MVT deposit in New Mexico (68 and 1, respectively), suggesting that both meteoric/mixed marine–meteoric (evolved meteoric) and basinal fluids may be involved during carbonate diagenesis.

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1. Introduction

Marine carbonates are generally precipitated in metastable phases such as aragonite (A) and high magnesium calcite (HMC), which are stabilized into the diagenetic low magnesium calcite (LMC) through water–rock interaction during exposure to meteoric or deep burial conditions (Tucker and Wright, 1990). Also, dolomitization is a common diagenetic process, which may influence the original marine carbonates or the previously altered phase (LMC) and it occurs at early (e.g., near-surface dolomitization) and/or late (e.g., hydrothermal dolomitization) stages during the burial history of sediments (e.g., Lonnee and Machel, 2006). Traces of the diagenetic waters are trapped in the form of fluid inclusions in the crystals of the end product carbonate. Those inclusions work as time capsules that retain the chemical characteristics of the diagenetic fluid and the temperature of precipitation of the diagenetic carbonate phase (Goldstein and Reynolds, 1994). Also, those inclusions may trap traces of the gases that existed during diagenesis. The relative concentrations of these gases reveal significant information about the nature of the diagenetic fluid (e.g., basinal vs. magmatic) and environment to provide an additional verification tool of the conditions that dominated during the sediment burial history (e.g., Norman and Musgrave, 1994).

The origin of fluids that drive diagenesis may at times remain debatable. Meteoric waters and basinal brines are potential fluids (e.g., Lavoie et al., 2005; Azmy et al., 2006; Lonnee and Machel, 2006; Azmy et al., 2008c, 2009; Conliffe et al., 2010) in addition to magmatic fluids (cf. Conliffe et al., 2010). Resolving the issue of the nature of diagenetic processes requires a multifaceted approach such as petrography, stable isotopes and trace element geochemistry, microthermometric studies and quantitative fluid-inclusion volatile analysis by mass spectrometry.

Fluid inclusions may be regarded as microscopic aliquots of a fluid that comprise possible combinations of liquid, gas or solid, trapped during mineral growth or by annealing of secondary fractures (Roedder, 1984). The most common inclusions in the current study are water with dissolved gases and salts. Salinity reported in diagenetic fluids of the investigated coarse-grained dolomite is up to 25 wt.% NaCl equivalent (e.g., Azmy et al., 2008c, 2009; Conliffe et al., 2009, 2010). However, no microthermometric measurements were obtained from the dolomicrites because of the crystal near-micritic size and submicron inclusions. Petrography of the investigated dolomites and their 2-phase fluid inclusions have been recently studied and discussed in detail by Azmy et al. (2001, 2006, 2008a, 2008b, 2008c, 2009), Conliffe et al. (2009, 2010) and Azmy and Conliffe (2010), which makes these dolomites ideal candidates for a fluid-inclusion gas ratio investigation. Origin of dolomitizing fluids have been the focus of studies particularly because of the development of porosity associated with dolomites, which constitute a significant

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global hydrocarbon reservoirs. Earlier studies of gas ratios in fluid inclusions provided ranges of compositions characteristic of magmatic fluids and non-magmatic (possibly meteoric or mixed marine-meteoric) fluids (e.g., Moore et al., 1997; Blamey and Norman, 2002), which can be applied in the identification of the origin of dolomitizing fluids through the diagenetic history of basins. The main objective of the current study is to investigate the composition of gases in fluid inclusions of non-altered and altered carbonate phases (aragonite, calcite and dolomites) from different basins (Canada, USA, Brazil and Java Sea) and diagenetic settings (shallow and deep burial) in an attempt to discriminate the fluid sources for the examined carbonates, particularly those from the St. George Group hydrothermal dolomites, and test the reliability of ratios of dissolved gases in fluid inclusions for the reconstruction of their diagenetic environments. This study is fairly new and it utilizes a suite of samples that was already extensively screened with multitechniques (petrography and geochemistry). The samples cover a wide spectrum of depositional and diagenetic settings.

2. Methods

There are four principle ways to gain information about fluid inclusion gases (Blamey, 2012), which are by microthermometry, Raman spectroscopy, gas chromatography and mass spectrometry. Microthermometry and Raman spectroscopy require relatively large fluid inclusions and the gases to be in relatively high abundance and these methods provide information on a limited species range. However, both methods lack the reliable detection limits and ability to measure a larger species range. Gas chromatography has an excellent species range but requires a 5–10 g sample, thus limiting its ability to resolve sample heterogeneities. Fluid-inclusion gas analysis by mass spectrometry has a broad species range, analyzes for noble gases, produces quantitative data, and meanwhile has detection limits around 0.2 ppm for a typical incremental crush. The ability to analyze samples as small as 25 mg, through the procedure utilized in the current study, has allowed minimizing problems with sample heterogeneities.

Fluid inclusion volatile analysis was done in vacuum using the CFS (crush-fast scan) method (Norman and Moore, 1997; Norman and Blamey, 2001; Parry and Blamey, 2010; Blamey, 2012) at the New Mexico Institute of Mining and Technology (NMT) Fluid Inclusion Gas laboratory. Unlike the conventional bulk thermal decrepitation method whereby all the inclusions are liberated to produce one analysis, the incremental crush method produces smaller bursts and hence increases information of potential sample heterogeneities if multiple fluid inclusion assemblages are present. Samples were first cleaned with NaOH to reduce surface organic contamination, then rinsed several times with deionized water, and dried at room temperature. Approximately 0.2 g of each sample was incrementally crushed under a vacuum of $\sim 10^{-8}$ Torr yielding five to eight crushes per sample. The vacuum is the equivalent of $\sim 10^{-11}$ atm and therefore the potential for air contamination is unlikely and opening inclusions in this high vacuum is orders of magnitude above background.

The analyses were performed by two Pfeiffer-Vacuum Prisma quadrupole mass spectrometers operating in fast-scan, peaking-hopping mode (Norman and Blamey, 2001). The system routinely reports the following gaseous species: H₂, He, CH₄, H₂O, N₂, O₂, Ar, and CO₂. The instrument was calibrated using synthetic inclusions filled with Scott Gas mixtures, and three in-house fluid inclusion gas standards as described by Norman and Blamey (2001), Norman et al. (2002), Parry and Blamey (2010), and Blamey (2012). The concentration of each species was calculated by proprietary software to provide a quantitative analysis. Precision is better than 5% for major gaseous species and 0.2% for water/gas ratios based on natural fluid inclusion standards. The 3 σ detection limit varies for most species and is dependent on burst size, interference from other species, and instrument alignment but is approximately 0.2 ppm for He (Blamey, 2012; Blamey et al., 2012),

which is comparable to the femtomole detection limit reported for gas chromatography (Bray et al., 1991).

The analyzed samples cover a wide spectrum of carbonates (Table 1) including well-preserved marine carbonates (modern aragonitic coral), altered sediments at near surface conditions (e.g., lime mudstones and dolomicrite) and late hydrothermal (fracture-filling) dolomite (Azmy et al., 2006, 2008a, 2008b; Azmy et al., 2009). The samples were globally selected from different sedimentary basins of different ages (Table 1) to examine the influence of age and burial conditions on the results. The purpose of including samples from the Mesoproterozoic Vazante dolomites is mainly to examine the influence of age on the investigated gas ratios since some of the investigated gases (e.g., He) can be produced by radiogenic decay of some elements such as U and other radiogenic elements although the concentrations of those elements is significantly lower in carbonates (≤ 0.5 ppm) compared with their counterparts in igneous rocks (cf. Norman and Musgrave, 1994). Also, the samples include organic-rich dolomicrites that had microbial (stromatolitic) lime mud precursor to investigate the influence of organic material on the fluid-inclusion gas ratios.

For comparison of fluid-inclusion gas ratio results, the fluorite (F) fluid inclusion laboratory standard HF-1 available at New Mexico Tech was analyzed as a regular sample (example of an MVT deposit) and 28 measurements were performed. This standard from the Hansonburg Mississippi-Valley type deposit (MVT) in New Mexico was originally collected, described, and analyzed using the thermal decrepitation method of gas extraction by Norman et al. (1985). The origin of MVT deposits is known to be amagmatic but from a basinal brine fluid and/or highly modified seawater component (Chi and Savard, 1997; Stoffel et al., 2008). By reanalyzing the HF-1 standard using the newer incremental crush fast scan method, we can certainly compare the diagenetic carbonate data to material that hosts basinal fluids acquired by a similar mechanism.

The detailed petrography of the carbonate samples is described by Azmy et al. (2006, 2008a, 2008b, 2009) and summarized in Table 1. The crushed material, from each sample, was intentionally selected to contain only a single carbonate phase to avoid mixing of fluid-inclusion gases released from different carbonates phases.

3. Geologic setting and lithostratigraphy

The nature of the sediments and diagenetic history have been well established through earlier studies of their detailed petrographic, microthermometric and geochemical attributes (e.g., Azmy et al., 2006; Knight et al., 2007; Azmy et al., 2008a, 2008c; Edinger et al., 2008; Knight et al., 2008; Azmy et al., 2009; Conliffe et al., 2009; Azmy and Conliffe, 2010; Conliffe et al., 2010, 2012; Conliffe et al., 2012). They include unaltered Modern aragonitic coral (A) from the Java Sea at Point Teluk Awur, near Jepara, Central Java (Azmy et al., 2008a; Edinger et al., 2008) and altered phases of marine carbonates, including lime mudstones (C1), late fracture-filling calcite (C3), dolomicrites (D1) of early diagenesis and late hydrothermal dolomites (D2 and D3, Table 1). The altered phases (hosted in lime mudstones and dolomites) were collected from two sequences (Lower Ordovician St. George Group in western Newfoundland, Canada and Mesoproterozoic Vazante sequence in east central Brazil) of different ages and from different sedimentary basins (Azmy et al., 2006; Knight et al., 2007; Azmy et al., 2008a, 2008b, 2008c; Knight et al., 2008; Azmy and Conliffe, 2010; Azmani et al., 2013).

The St. George Group consists of early Ordovician (Tremadoc–Arenig) platform carbonates (~ 500 m-thick) which, from bottom to top, include the Watts Bight, Boat Harbour, Catoche, and Aguathuna formations (Fig. 1). The upper boundary of the St. George Group (Aguathuna/Table Point formations contact) is marked by the major regional St. George Group Unconformity (SGU, Fig. 1). The St. George Group can be divided into two sedimentary megacycles separated by the Boat Harbour Disconformity (BHD, Fig. 1). Each megacycle is

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