



Tracing the evolution of seep fluids from authigenic carbonates: Green Canyon, northern Gulf of Mexico



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ABSTRACT

Authigenic carbonates from hydrocarbon seeps are unique long-term archives of past fluid flow. The studied samples were collected from Green Canyon block 140 at a water depth of 260 m in the Gulf of Mexico. Petrography, X-ray diffraction, stable isotopes and ¹⁴C dating were applied to assess the evolution of seep activity and potential driving forces. The carbonates are dominated by high-Mg calcite (HMC) and aragonite, with a minor amount of low-Mg calcite (LMC) and dolomite. Petrographically, peloids, clotted microfabric, acicular aragonite and a variable content of bioclasts were observed. Three types of carbonates are recognized. Structure I carbonates, with ¹⁴C ages from 46.5 ka to 25.8 ka BP, are characterized by $\delta^{13}\text{C}$ values from -23.2‰ to 5.1‰ suggesting multiple carbon sources that include thermogenic methane, biodegraded crude oil, seawater and residual CO₂ from methanogenesis at greater depth. In contrast, Structure II carbonates formed between 17.6 ka and 11.7 ka BP and have $\delta^{13}\text{C}$ values varying from -22.2‰ to -8.8‰ , suggesting carbon sources similar to those of Structure I carbonates but with a negligible influence of residual CO₂ from methanogenesis. In addition, the presence of LMC in this type of carbonate may be associated with brine seepage. Structure III carbonates among the youngest of the samples analyzed with ¹⁴C ages of 1.2 ka BP. These carbonates have the most negative $\delta^{13}\text{C}$ values ranging from -36.1‰ to -26.8‰ , suggesting that thermogenic methane is the primary carbon source. The majority carbonates of both Structure I and II are slightly ¹⁸O-enriched, which is most likely related to the incorporation of water from dehydration of clay minerals. The considerable range of mineralogical and isotopic variations of the studied carbonates highlights the local control of the seepage flux. It is proposed that factors affecting the activity of hydrocarbon seeps are sea level changes and salt movement. The combination of petrography, stable isotopes, and dating approach used here, highlights that these are valuable tools to assess the variability of past fluid flow at hydrocarbon seeps.

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1. Introduction

Cold hydrocarbon seepage is a frequently observed phenomenon in marine settings worldwide (e.g., Campbell, 2006; Judd and Hovland, 2007 and references therein). The key process at cold seeps is anaerobic oxidation of methane, conjointly operated by consortia of anaerobic methane-oxidizing archaea and sulfate-reducing bacteria (Boetius et al., 2000; Valentine and Reeburgh, 2000). This process leads to an increase of pore water alkalinity

by the production of bicarbonate that favors the precipitation of authigenic carbonates (Baker and Burns, 1985; Berner, 1980), the so-called seep carbonates or methanogenic carbonates. Seep carbonates, thus, provide excellent long-term archives of past seepage and associated environmental parameters (Aloisi et al., 2000; Birgel et al., 2011; Bohrmann et al., 1998; Chen et al., 2006; Feng et al., 2009a, 2010b; Greinert et al., 2001, 2002; Greinert and Derkachev, 2004; Mazzini et al., 2004, 2008; Naehr et al., 2007; Peckmann et al., 1999, 2001a,b, 2002, 2009; Tong et al., 2013).

Seep carbonates are known to show complex mineralogical as well as carbon and oxygen isotopic compositions (Campbell, 2006; Gieskes et al., 2005; Mazzini et al., 2005; Naehr et al., 2007; Peckmann et al., 1999, 2001a,b; Peckmann and Thiel, 2004;

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Roberts et al., 2010; Roberts and Aharon, 1994). Carbonate minerals in seep carbonates are primarily high-Mg calcite (HMC), aragonite and dolomite (protodolomite; Greinert et al., 2001; Naehr et al., 2007; Peckmann et al., 2001b; Roberts et al., 2010; Roberts and Aharon, 1994). Many factors influence the type of carbonate minerals that precipitate at hydrocarbon seeps. In general, high levels of carbonate supersaturation in combination with a high sulfate concentration seem to favor aragonite precipitation over HMC and dolomite (Greinert et al., 2001; Moore et al., 2004; Naehr et al., 2000, 2007; Peckmann et al., 2001b). The occurrence of low-Mg calcite (LMC) may be attributed to a brine-rich environment, which is relatively Mg-depleted compared to normal marine settings that usually produce HMC and aragonite (cf. Aloisi et al., 2000; Davis, 2000; Feng and Roberts, 2011; Judd and Hovland, 2007).

The $\delta^{13}\text{C}$ values of seep carbonates reflect the isotopic signature of the dissolved inorganic carbon pool at the time of carbonate precipitation (Mazzini et al., 2004, 2008; Naehr et al., 2007; Peckmann and Thiel, 2004; Roberts and Aharon, 1994). At seeps, the possible carbon sources include: 1) biogenic methane ($\delta^{13}\text{C} < -65\text{‰}$; Whiticar, 1999) and thermogenic methane ($\delta^{13}\text{C} = -50\text{‰}$ to -30‰ ; Sackett, 1978; Whiticar et al., 1986), 2) oil fraction ($\delta^{13}\text{C} = -35\text{‰}$ to -25‰ ; Roberts and Aharon, 1994), 3) seawater ($\delta^{13}\text{C} = 0 \pm 3\text{‰}$; Anderson and Arthur, 1983), and 4) residual CO_2 from methanogenesis ($\delta^{13}\text{C}$ as high as 26‰ ; Gieskes et al., 2005; Naehr et al., 2007; Paull et al., 2007). The stable oxygen isotopic compositions of a specific carbonate mineral phase depend on the formation temperature and isotopic composition of the source water where the precipitation occurred (Friedman and O'Neil, 1977; Naehr et al., 2000, 2007). Anomalously positive $\delta^{18}\text{O}$ values may be derived from decomposition of gas hydrate or dehydration of clay minerals (e.g., Bohrmann et al., 1998; Davidson et al., 1983; Hesse, 2003; Matsumoto, 1994). Samples that are ^{18}O -depleted may indicate the influence of the local presence of meteoric water or residual fluids from gas hydrate formation

during carbonate precipitation (Dahlmann and de Lange, 2003; Greinert et al., 2001).

Seafloor observations indicate that the physical, chemical and biological processes at hydrocarbon seeps can change significantly with time (Chen et al., 2004; Greinert, 2008; Roberts, 2001; Roberts and Carney, 1997; Schneider von Deimling et al., 2011; Solomon et al., 2008; Torres et al., 2002; Tryon, 2004; Tryon and Brown, 2001), and this temporal variability can be expressed by the difference in geochemical and mineralogical signatures of the associated seep carbonates (Bayon et al., 2007, 2011; Campbell et al., 2010; Díaz-del-Río et al., 2003; Feng et al., 2009a,b, 2010a; Ivanov et al., 2010; Magalhães et al., 2012; Mazzini et al., 2006, 2008; Naehr et al., 2007; Peckmann et al., 2001b, 2009; Pierre et al., 2012; Rongemaille et al., 2011). Here, we investigated authigenic carbonate rocks from the subsurface sediment of the hydrocarbon seeps at Green Canyon block 140 (GC140) of the Gulf of Mexico (GOM) continental slope. Petrography, mineralogy, stable isotopes and ^{14}C dating were applied to constrain the conditions under which the carbonates formed. The obtained data were also used to constrain the long-term evolution of the seep fluids and the possible driving forces. This study aims at tracing the evolution of seep fluids by a multidisciplinary study of authigenic carbonates.

2. Geological setting and sampling

The Gulf of Mexico is a hydrocarbon-bearing basin with various types of salt structures. Faults resulting from subsurface salt movement serve as pathways for deep hydrocarbon migration. The hydrocarbon seepage produces brine pools, gas hydrates and authigenic carbonates (MacDonald et al., 2010; Milkov and Sassen, 2001; Roberts et al., 2000, 2010; Sassen et al., 1999; Weimer et al., 1998). The GC140 seep site is located in the northern upper continental slope of the GOM (Fig. 1). This area is characterized by an abundance of faults activated by salt diapirism. Geophysical data, submersible observations, and samples retrieved clearly indicate

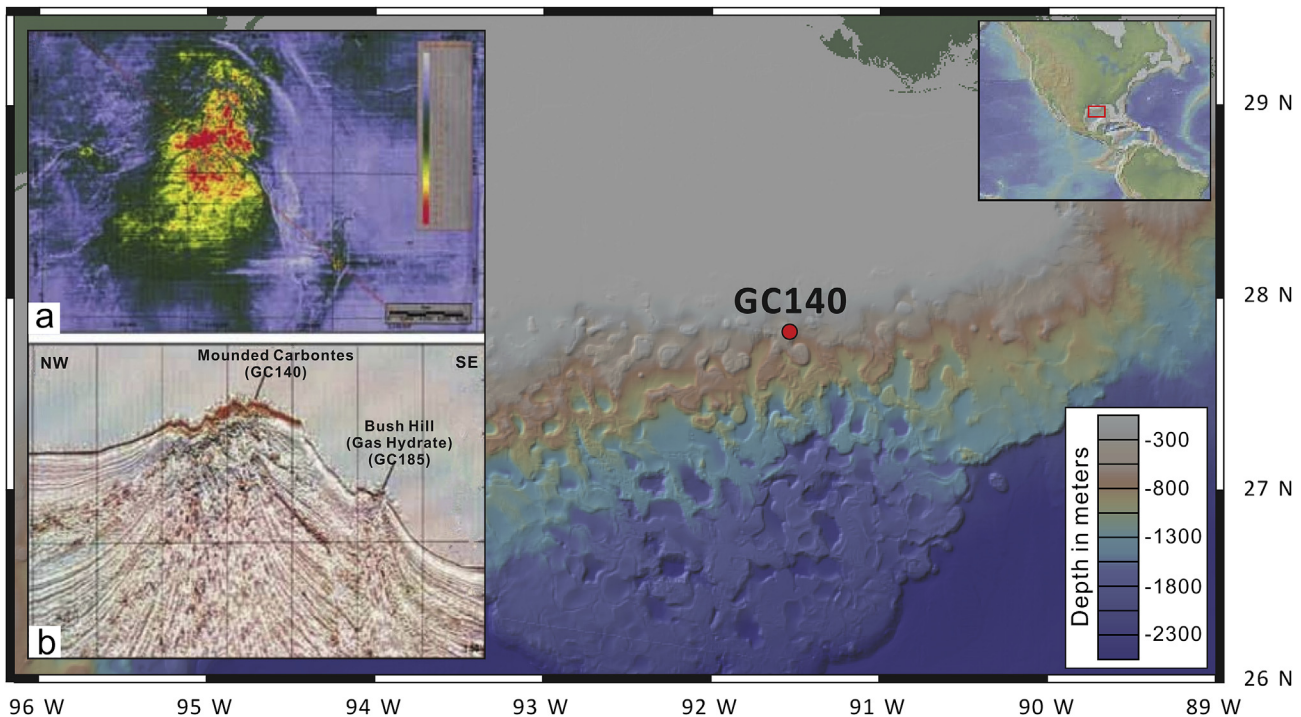


Figure 1. Location of the Green Canyon block 140 (27°48'N/91°32'W) study site, northern Gulf of Mexico. The inset 3-D seismic surface amplitude illustrates the extent of the hard bottom (a) and the profile view (b), showing that the dome top has an irregular surface and that the seafloor reflector is a strong, positive reflector, indicating a hard bottom (Roberts, 2011).

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