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Research paper

Fluid inclusion and microfabric studies on Zechstein carbonates (Ca2) and related fracture mineralizations — New insights on gas migration in the Lower Saxony Basin (Germany)

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ABSTRACT

New petrographic and fluid inclusion data from core samples of Upper Permian dolomitic limestone (Hauptdolomit, Zechstein group, Stassfurt carbonate sequence) from a gas field located at the northern border of the Lower Saxony Basin (LSB) essentially improve the understanding of the basin development. The gas production at the locality is characterized by very high CO_2 concentrations of 75–100% (with CH_4 and N_2).

Samples consist of fine grained, mostly laminated and sometimes brecciated dolomitic limestone (mudstone/wackestone) from the transition zone between the shallow water zone (platform) and the upper slope. The study focuses on migration fluids, entrapped as fluid inclusions in diagenetic anhydrite, calcite, and fluorite, and in syn-diagenetic microfractures, as well as on the geochemistry of fluorite fracture mineralizations, obtained by LA-ICP-MS analysis. Fluid inclusion studies show that the diagenetic fluid was rich in H_2O–NaCl–$CaCl_2$. Recrystallized anhydrite contains aqueous inclusions with homogenization temperatures (T_h) of ca. 123 °C, but somewhat higher T_h of ca. 142 °C was found for calcite cement followed by early Fluorite A with T_h of 147 °C. A later Fluorite B preserves gas inclusions and brines with maximum T_h of 156 °C. Fluorite B crystallized in fractures during the mobilization of CO_2-bearing brines. Crossing isochores for co-genetic aqueous-carbonic and carbonic inclusions indicate fluid trapping conditions of 180–200 °C and 900–1000 bars. $\delta^{13}C$-isotopic ratios of gas trapped in fluid inclusions suggest an organic origin for CH_4, while the CO_2 is likely of inorganic origin.

Basin modelling (1D) shows that the fault block structure of the respective reservoir has experienced an uplift of >1000 m since Late Cretaceous times.

The fluid inclusion study allows us to, 1) model the evolution of the LSB and fluid evolution by distinguishing different fluid systems, 2) determine the appearance of CO_2 in the geological record and, 3) more accurately estimate burial and uplift events in individual parts of the LSB.

1. Introduction

The evolution of the Lower Saxony Basin (LSB) as southern part of the Central European Basin has been subject to petroleum geological investigations for many decades with a main focus on the generation of oil and gas resources and their migration within the basin (e.g. Boigk, 1981; Boigk and Stahl, 1970; Plein, 1985). During petroleum exploration, major CO_2 occurrences were tapped, with CO_2-concentrations exceeding 75% (Brand et al., 1982) and accompanied by other gases like methane, nitrogen, and hydrogen sulfide (e.g. Doornenbal and Stevenson, 2010; Krooss et al., 2008; Lüders et al., 2005, 2012). Although high CO_2 concentrations are characteristic for reservoir rocks within the Zechstein units of the LSB, the source of CO_2 is still under debate. Apart from thermal decomposition of organic material, degassing of magmatic bodies may have caused CO_2-generation (Lokhorst et al., 1998); therefore, plutonic intrusions during the Cretaceous into the basement were discussed by various authors (Bilgili et al., 2009; Bruns et al., 2013; Hahn and Kind, 1971; Kus et al., 2005; Petmecky et al., 1999; Stadler and Teichmüller, 1971; Ziegler, 1990). While some authors favor the

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existence of plutonic intrusions in order to explain high vitrinite reflectance values between 3.0 and 4.5%VRr in some parts of the LSB (Teichmüller and Teichmüller, 1985; Bartenstein et al., 1971), other possible reasons for supporting those levels of thermal maturity include deep burial followed by basin inversion (Adrisola-Muñoz, 2007; Adrisola-Muñoz et al., 2007; Petmecky et al., 1999), and the circulation of hydrothermal fluids along faults during rifting (Stadler and Teichmüller, 1971; Reutel and Lüders, 1998). Basin modelling based on new seismic and thermometric data (e.g. fluid inclusion analysis, apatite/zircon fission track analysis) has revealed, that anomalous thermal maturity values in different parts of the LSB (e.g. anomalies of Bramsche, Vlotho, Uchte, Apeldorn) can be explained by deep burial during the Late Jurassic/Early Cretaceous and subsequent rapid uplift during the Late Cretaceous (e.g. Adrisola-Muñoz, 2007; Brink, 2005; Bruns et al., 2013; Fischer et al., 2006; Petmecky et al., 1999; Senglaub et al., 2005).

Fluid inclusion studies of cement phases and mineralized fractures can help to better understand burial and uplift events and their effect on the mobilization of fluids and gases in the LSB. Several studies in the North German Basin suggest a considerable variability of fluid inclusions with respect to chemical composition and gas content as a result of water-rock interaction (formation of Na-/Ca-rich brines due to the presence of evaporites and volcanic rocks), decomposition of organic matter (CH_4/CO_2-formation), thermochemical sulfate reduction (TSR), and hydrothermal activity (Fischer et al., 2006; Huttel, 1989; Lüders et al., 2005, 2012; Reutel and Lüders, 1998; Schmidt Mumm and Wolfgramm, 2002, 2004; Siemann and Ellendorff, 2001; Steuer, 2008; Zwart and Touret, 1994). Few fluid inclusion studies are focussed on the source and migration of CO_2 in the LSB (Fischer et al., 2006; Lokhorst et al., 1998; Lüders et al., 2005, 2012).

Fluids within the Zechstein limestone are dominated by $CaCl_2$–NaCl-rich solutions and commonly show salinities between 16 and 23 wt% (eq) NaCl and 7–15 wt% (eq) $CaCl_2$, respectively (e.g. Zwart, 1995; Zwart and Touret, 1994); other salts like LiCl and KCl are present as well, but they mostly occur in low concentrations and usually together with $CaCl_2$. Significant changes in brine salinity are often explained with fluid migration from one fluid system to the other (e.g. Huttel, 1989), triggered by extensional fracturing. Aqueous fluid inclusions can contain traces of dissolved gases like CO_2, CH_4, N_2, and H_2S, while pure gaseous inclusions are rare. CO_2- and CH_4-rich gaseous inclusions are reported from fracture mineralizations that relate to rifting (extensional fractures) or reverse faulting (shear fractures) (Fischer et al., 2006; Lüders et al., 2005, 2012; Reutel and Lüders, 1998; Schmidt Mumm and Wolfgramm, 2002, 2004) and thus may provide information about timing and P-T conditions during gas migration.

In order to relate changes in fluid and mineral composition to specific geological events special emphasis is given to fluorite geochemistry. Numerous studies on fluorite have shown (Bau and Dulski, 1995; Bau and Möller, 1991, 1992; Jacob, 1974; Lüders, 1991; Möller et al., 1976, 1980; Schulz, 1980; Schwinn and Markl, 2005) how distributions of REE and Y in fluorite may help identifying the environment of crystallization, i.e. distinguish between primary crystallized and recrystallized fluorite (Möller et al., 1976). Thus differences in fluorite chemistry may reveal the importance of hydrothermal and magmatic fluids during basin evolution.

Information on fluid migration in sedimentary rocks can be obtained from fluid inclusions in healed micro-fractures, which provide the pressure and temperature of fluid trapping as well as the fluid composition (Goldstein, 2001; Goldstein and Reynolds, 1994). Steep fractures are supposed to form in extensional regimes during rifting and simultaneous subsidence, when vertical stress prevails (extensional fracturing or extensional-shear fracturing, see Sibson, 1998); brittle fracturing is favoured also by fluid overpressure. In order to define the burial depth and tectonic activity fracture-hosted fluid inclusions have been studied and compared with geothermal and geostatic gradients from the LSB (e.g. Nollet et al., 2009). Anhydrite and fluorite vein mineralizations, which follow fracture propagation due to extensional-shear during burial, allow us to calculate pressure and temperature conditions in the reservoir close to maximum burial (P-T calculated from co-existing fluids). This information is used to calibrate the 1D model together with the relative ages of mineral precipitation as obtained from standard optical microscopy and fluid inclusion studies.

Fluid migration and mineral precipitation within the LSB are subjected to changing tectonic regimes (e.g. Strohmenger et al., 1996). For example, transpressional forces during reactivation of older normal faults located at the northern margin of the LSB supported the forming of positive flower structures and fault block structures along a ESE-WNW striking thrust belt (Kockel, 1994; Ziegler, 1990). By investigating core samples from an individual fault block, which is limited by a ENE-WSW striking reverse fault, we aim to reconstruct the evolution of the fluid phases at the northern margin of the LSB, and mark the time of CO_2 migration and trapping. Our fluid inclusion, micro-structural and petrographic study complements existing evolution models of the LSB based on vitrinite reflectance (e.g. Adrisola-Muñoz, 2007; Buntebarth, 1985; Petmecky et al., 1999; Senglaub et al., 2005; Uffmann, 2013) and fission track analyses (Adrisola-Muñoz, 2007; Petmecky et al., 1999; Senglaub et al., 2005).

2. Geological setting

The study area is located at the north-eastern border of the Lower Saxony Basin, which is part of the North German Basin (NGB). The NGB forms the southern extension of the Central European Basin; it is an intra-cratonic basin that extends from the North Sea in the northwest to Poland in the southeast. The basin is mainly composed of thick sequences of Paleozoic and Mesozoic sediments that filled a complex WNW-ESE oriented graben system (Baldschuhn et al., 2001; Betz et al., 1987; Boigk, 1968; Kley and Voigt, 2008; Kockel, 1994; Kockel et al., 1994) (Fig. 1). During the Late Carboniferous – Early Permian extensional tectonics caused crustal rifting which resulted in the thinning of the crust and the rise of magmas from the mantle (Ziegler, 1990; Brink et al., 1992). In the eastern part of the NGB volcanic sequences have formed continuous layers of several hundreds of meters covering Carboniferous sediments, while to the west of the basin, e.g. in the LSB, Permian volcanic rocks form patch-like spots due to erosion or missing deposition (Lokhorst et al., 1998; Mempel, 1962). Sediments were unconformably deposited on folded Variscan basement rocks (Füchtbauer, 1962; Richter-Bernburg, 1955). During Early Triassic a system of NNE-SSW trending horst and graben structures formed due to the evolution of the North Sea rift system (Betz et al., 1987; Doornenbal and Stevenson, 2010; Lokhorst et al., 1998). From Mid to Late Jurassic thermal doming affected the NGB leading to a period of uplift and thermal subsidence (Ziegler, 1990). During the Late Jurassic – Early Cretaceous an extensional stress regime resulted in the formation of the ENE – WSW trending LSB (Betz et al., 1987; Brink et al., 1992) when the Permo – Carboniferous fracture system was reactivated (Betz et al., 1987).

The LSB is bounded to the north by the Pompeckj Block, and to the south by the Münsterland platform (Brink et al., 1992; Glennie, 1990). The main subsidence occurred during the Late Jurassic and Early Cretaceous (Boigk, 1981; Doornenbal and Stevenson, 2010) and continued until Mid-Cretaceous (Betz et al., 1987; Doornenbal and Stevenson, 2010). Subsidence was subsequently followed by three major tectonic inversion phases, likely related to