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## Solid bitumen in calcite veins from the Natih Formation in the Oman Mountains: Multiple phases of petroleum migration in a changing stress field



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#### ABSTRACT

Solid bitumen in calcite veins in Natih limestone on the southern flank of the Jebel Akhdar Anticline provide evidence for at least two previously unrecognized petroleum migration events in the Oman Mountains. We present field observation of bituminous calcite veins combined with microscopy using highly polished thin sections. This allows imaging of solid bitumen in reflected light and study of its microstructural context in transmitted light. Straight and en échelon, black impregnated, bedding-normal Natih A calcite veins strike 110°. They formed as one of the earliest structures in an extensional stress regime at the end of Cretaceous (Turonian–Santonian) either by flexure of the Arabian plate (Wasia-Aruma break) or by subsidence during subduction. The veins always contain planar arrays of small (<10 µm) intracrystal solid bitumen inclusions and may contain angular mosaic type solid bitumen associated with white, solid bitumen-free calcite and/or round, homogeneous type solid bitumen. The solid bitumen formed in multiple events and were most likely derived from the Natih B source rocks. During the first migration event, microfracturing during obduction of the Semail ophiolite and emplacement of the Hawasina thrust sheets in Santonian-Campanian times formed pathways for oil into the veins. Mosaic type solid bitumen formed during further vein growth. It has a higher solid bitumen reflectance ( $BR_r = 3.40-3.76\%$ ) combined with a high optical anisotropy compared to the underlaying thermally overmature Natih B source rock ( $BR_r = 3.10-3.14\%$ ). The higher reflectance of mosaic type solid bitumen is interpreted to result from deformation and does not reflect maximum burial temperatures which are inferred to have been about 225 °C. The second migration event is indicated by low-reflective homogeneous type solid bitumen with palaeotemperatures ( $BR_r = 0.86-0.92\%$ ; about 145 °C) lower than maximum burial. It occurs along pressure solution seams which cross-cut the veins showing that this is the latest event in the paragenetic sequence, interpreted to have formed after lateral intraformational oil migration from the active Natih oil kitchen located approximately 40 km SW of the study area during doming and uplift of the Oman Mountains in Oligocene to Pliocene times. © 2015 Elsevier B.V. All rights reserved.

#### 1. Introduction

Calcite veins on the southern flank of the Jebel Akhdar Anticline in the Oman Mountains have been used to study the geologic history of the area and infer paleostress directions and fluid properties (Arndt et al., 2014; Breton et al., 2004; Filbrandt et al., 2006; Gomez-Rivas et al., 2014; Hilgers et al., 2006; Holland and Urai, 2010; Holland et al., 2009a, b; Virgo et al., 2013). The predominantly white calcite vein networks occasionally comprise grey, bituminous calcite veins. In this paper we present results of an integrated analysis of field work, microscopy and organic geochemistry to unravel timing and mechanisms of a fossil oil migration system, which has undergone multiple major stress changes during its geologic history.

## 1.1. The use of solid bitumen as a hydrocarbon migration and paleotemperature indicator

The presence of solid bitumen in veins is diagnostic for hydrocarbon migration. Oils alter into solid bitumen after migration by various processes. The most important ones are (Blanc and Connan, 1994; Rogers et al., 1974): (1) thermal cracking, (2) biodegradation and (3) gas deasphalting. Thermal cracking of oil by increasing temperature leads to the release of gaseous and light hydrocarbons and precipitation of solid bitumen. According to Dahl et al. (1999) oils are only stable at

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temperatures <150 °C. Thermal cracking decreases the hydrogen index (HI) values and increases the aromaticity of solid bitumen similar to the maturation of vitrinite (Landis and Castaño, 1995; Robert, 1988). Biodegradation occurs at lower temperatures (<80 °C) due to microbial oxidation of oil whereas gas deasphalting is controlled by gas injection into the oil causing precipitation of solids (Blanc and Connan, 1994; Head et al., 2003; Lomando, 1992).

Reflectance of solid bitumen ( $BR_r$ ) as a maturity parameter can be used similar to vitrinite reflectance ( $VR_r$ ), which is widely applied in basin research. Pre-Devonian rocks, crystalline rocks and veins usually do not contain any vitrinite. In these cases, solid bitumen can be used instead as a paleothermometer. Occurrence of several generations of solid bitumen with different reflections may indicate multiple migration episodes (Gentzis and Goodarzi, 1990). After peak temperature, generation and migration of hydrocarbons will stop at a certain location. So, the bitumen with lower reflections compared to maximum burial temperatures postdate peak temperature and suggest arrival of hydrocarbons from an external, actively generating oil kitchen. In contrast, immobile vitrinite always experience maximum burial temperature.

Jacob (1989) and Landis and Castaño (1995) showed that there is a strong linear correlation between vitrinite reflectance and solid bitumen reflectance by measuring a large number of samples containing both types. However, these linear equations are not applicable over the complete maturity range (0.2-8.0% VR<sub>r</sub>) (Ferreiro Mählmann and Frey, 2012). Therefore, we used the cubic equation of Ferreiro Mählmann and Frey (2012), which consists of a combined dataset of Ciulavu et al. (2008), Ferreiro Mählmann and Giger (2012) and Ferreiro Mählmann (1994, 2001) (Eq. (1)):

$$\begin{split} BR_r[\%] &= -0.519 + 1.341 (VR_r[\%]) - 0.0977 (VR_r[\%])^2 \\ &\quad + 0.0151 (VR_r[\%])^3 \end{split} \tag{1}$$

Jacob (1989) subdivided solid bitumen ("migrabitumen") into different asphaltenes and impsonites (epi-, meso-. cata-impsonite) with impsonites having a reflectance >0.7% due to thermal alteration. Landis and Castaño (1995) classified solid bitumen based on their microstructures, namely: (1) homogeneous type, (2) granular type and (3) "coked" type. Homogeneous type solid bitumen shows a uniform reflectance and gives the most reliable results for peak burial temperatures in source rocks whereas the granular type has lower reflectance. The "coked" (with mosaic or flow structure) type solid bitumen has a strong optical anisotropy (Amijaya and Littke, 2006; Hwang et al., 1998; Landis and Castaño, 1995) that is interpreted to develop by high temperatures due to rapid heating (e.g. contact metamorphism) (Gize, 1986; Goodarzi et al., 1992; Hwang et al., 1998; Khavari-Khorosani and Murchison, 1978).

#### 1.2. Geological setting and study area

The study area is situated at the southern flank of the Jebel Akhdar Anticline in the Oman Mountains (Fig. 1). This mountain range trends NW–SE over 700 km from the Musandam Peninsula in the north of the Sultanate of Oman to the Batain coast in the southeast. The Oman Mountains were formed during several tectonic events. Most importantly for this study, SW directed obduction of the volcanicsedimentary Hawasina nappes and Semail ophiolite nappe started at the end of Cretaceous, during the Cenomanian (Breton et al., 2004; Glennie et al., 1974; Searle, 2007). After nappe emplacement the whole region underwent several episodes of extension and transpression that were followed by uplift, doming and erosion of the Jebel Akhdar Anticline (Al-Wardi and Butler, 2007; Breton et al., 2004; Filbrandt et al., 2006; Fournier et al., 2006; Gomez-Rivas et al., 2014; Hanna, 1990).

The Oman Mountains consist of three major units: Autochthonous A and B, Allochthonous nappes and Neo-autochthonous (Figs. 1 and 2A). The oldest unit, Autochthonous A, consists of a Neo-Proterozoic basement and Pre-Permian sediments (Roger et al., 1991). It is unconformably overlain by Autochthonous B, a 3 km thick succession of carbonates deposited on the passive margin of the Neo-Tethys ocean from late Permian to Late Cretaceous (Turonian). The uppermost part of this succession is the top of the Cretaceous Wasia group, the Natih Formation. It is subdivided after Clarke (1988) into seven members (A–G), from which this study focuses on the Natih A/B member



Fig. 1. Simplified geological map of the central part of the Oman Mountains and the foreland basin. The study area is situated on the southern flank of the Jebel Akhdar tectonic window where the Autochthon B is exposed. The map is simplified from Beurrier et al. (1986), Loosveld et al. (1996) and Breton et al. (2004). The cross-section below is modified from Searle (2007) and Boote et al. (1990).

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