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Traces of hydrocarbon migration in the Central Mecsek Mountains

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Abstract

Hydrocarbon-bearing fluid inclusions in fracture-filling calcite and void-filling dolomite and quartz cement phases in Middle Triassic carbonates at Árpádtető, in the Central Mecsek Mountains indicate that hydrocarbon migration occurred in the area. Petrographic observations suggest the presence of at least two hydrocarbon fluid generations in the host void-filling quartz crystals at Árpádtető representing multiple events of hydrocarbon migration of different composition and/or different source rocks.

Keywords: hydrocarbon-bearing fluid inclusions, microthermometry, hydrocarbon migration, source rocks, Mecsek Mountains

Introduction

Recently, traces of hydrocarbon migration were found near Pécsvárad in the Eastern Mecsek Mountains, in fracture-filling calcite in the Hosszúhetény Calcareous Marl Formation (LUKOCZKI et al. 2012). Recent investigation of Middle Triassic dolostones at Árpádtető (*Figure 1, A–B*) by LUKOCZKI & HAAS (2013) revealed the presence of hydrocarbon-bearing fluid inclusions (HCFI) in fracture-filling calcite and void-filling dolomite and quartz cements, as well as in the crystals of the host dolostone indicating that hydrocarbon migration occurred also in the Central Mecsek Mountains. Further occurrences of HCFIs were found in fracture-filling quartz in the Mecsek Coal Formation in outcrops at Pécs-Vasas (JÁGER V., pers. comm.) and in fracture-filling calcite in the Kantavár Formation at Kantavár (SZIGETI 2013). This paper presents a short description of the recently found HCFI occurrences at Árpádtető.

Middle Triassic limestone and dolostone outcrops can be found in an abandoned quarry, in a road-cut and in several small excavations at Árpádtető. The first detailed description of the dolostone outcropping in the quarry was given by NAGY (1968), who suggested that the dolomitization of the thick bedded limestone advanced along fractures at elevated temperatures probably during deepest burial or related to hydrothermal events after the Cretaceous, but before the Tertiary deformation events.

VETŐ-ÁKOS (1978) studied fluid inclusions hosted in void-filling idiomorphic quartz crystals collected from the same quarry as studied by NAGY (1968). VETŐ-ÁKOS (1978) distinguished a primary and a secondary fluid inclusion generation in the studied quartz crystals: The primary fluid inclusions contain two phases (liquid+vapour) with colourless liquid phase and homogenize to the liquid phase between 130–150 °C. The secondary inclusions contain two or three phases. The liquid phase in the two-phase (liquid+vapour) secondary inclusions is usually yellowish brown. The secondary two-phase inclusions homogenize to the liquid phase between 80–120 °C. The three-phase secondary inclusions contain two liquid and a vapour phases and are colourless. VETŐ-ÁKOS (1978) did not describe the presence of HCFIs. Based on homogenization temperature (T_h) measurements VETŐ-ÁKOS (1978) concluded that the quartz crystals formed during maximum burial and/or related to magmatic events not sooner than Early Cretaceous.

Traces of hydrocarbon migration were revealed by the presence of hydrocarbon bearing HCFIs at three localities besides Árpádtető. Secondary HCFIs found in calcite veins in the Early Jurassic Hosszúhetény Calcareous Marl Formation near Pécsvárad are colourless in plane polarized light and show yellowish blue, light blue and vivid blue fluorescence under UV-light, homogenize to the liquid phase between 30–100 °C and do not contain solid phases (LUKOCZKI et al. 2012). Primary, green fluorescent HCFIs in quartz crystals found in the Pécs-Vasas coal pit at the contact of magmatic dykes and silty sandstones (Mecsek Coal Formation, Early Jurassic) contain gas-dominant two-phase fluid inclusions (gas+liquid), which homogenize to the gas phase between 93–97 °C suggesting entrapment from gas phase (JÁGER V., pers. comm.). Blue fluorescent HCFIs were also found in calcite veins in a quarry at Kantavár (Kantavár Formation, Late Triassic), but their study has not been performed yet (SZIGETI M., pers. comm.).

Five Mesozoic formations can be considered as potential source rocks for the hydrocarbons entrapped in fluid inclusions in the Mecsek Mountains. The Late Triassic Kantavár Formation, the Early Jurassic Mecsek Coal and Vasas Marl Formations could have been gas source rocks, while the Early Jurassic Hosszúhetény Calcareous Marl Formation could have been a source rock for liquid hydrocarbons (BADICS & VETŐ 2012; LUKOCZKI et al. 2012). The Early Jurassic Óbánya Siltstone Formation in the Réka Valley contains immature organic matter (RAUCSIK et al. 2002), thus could not have been a source rock; however, all organic matter-rich Early Jurassic formations reached the oil window in the northern forelands of the Mecsek Mountains and thus could have generated hydrocarbons (BADICS & VETŐ 2012).

Hydrocarbon-bearing fluid inclusions at Árpádtető

Hydrocarbon-bearing fluids are present in fluid inclusions in the planar-s dolomite crystals of the medium crystalline, fabric destructive host dolostone, as well as in fracture-filling calcite, and in void-filling saddle dolomite and quartz cements. The fracture-filling, medium–coarsely crystalline blocky calcite appears as thin (few mm) veins cross-cutting the dolostone, but not affecting the other cement phases. The medium–coarsely crystalline saddle dolomite occurs as lining voids, where the medium–extra coarsely crystalline subhedral quartz is the final cement phase, if present. Based on textural relationships, calcite→ dolomite→quartz cement paragenesis can be established.

In the rock-forming dolomite blue fluorescent HFCIs occur in trails cross-cutting several crystals, which refers to secondary origin. The fluid inclusions are very small (<5 µm) thus are not suitable for further analysis.

In the fracture-filling calcite the fluid inclusions are small (<10 µm) and contain two phases ($liquid_{aq/hc}$+vapour/gas; aq= aqueous, hc=hydrocarbon). Sparse, blue-fluorescent HCFIs are scattered in the calcites, which makes the determination of their primary or secondary origin difficult and their small size impedes their detailed investigation.

In the void-filling saddle dolomite small (<10 µm) HCFIs seem to be arranged mostly along growth zones (*Figure 1, C*) suggesting primary origin. Both the aqueous and the HCFIs contain two phases ($liquid_{aq/hc}$+vapour/gas) and the hydrocarbon-bearing liquid shows blue fluorescence in UV-light. Due to their small size, further investigations of these fluid inclusions could not be performed.

Three fluid inclusion generations were distinguished in the void-filling quartz crystals: one aqueous and two with hydrocarbon-bearing fluids. Phase volume ratios were estimated visually.

Primary, two-phase (liquid+vapour) aqueous fluid inclusions (5–20 µm) occur with constant volume ratios, the liquid phase being the dominant. The shape of the fluid inclusion vacuole is usually angular.

Secondary one-, two- and three-phase HCFIs ($liquid_{aq/hc}$, $liquid_{aq}$+$liquid_{hc}$, $liquid_{aq/hc}$+gas and $liquid_{aq}$+$liquid_{hc}$+gas) (5–30 µm) occur with varying volume ratios. The fluid inclusion vacuoles have irregular shapes (*Figure 1, D–E)*, in many cases with the presence of necks (*Figure 1, D*), suggesting necking-down. The hydrocarbon-bearing liquid is colourless in plane polarized light and shows blue fluorescence under UV-light (*Figure 1, D–E)*.

Large (50–200 µm), variously shaped, three- or four-phase secondary HCFIs ($liquid_{hc}$+solid+gas, $liquid_{aq}$+$liquid_{hc}$+ solid+gas) have yellowish brown colour in plane polarized light (*Figure 1, F*) and show blue fluorescence under UV-light. The solid phase occurs seemingly attached to the inclusion walls, has brown colour and show no fluorescence under UV-light. The $liquid_{hc}$–solid–gas volume ratios are constant in the three-phase HCFIs. A rim of aqueous liquid is discernible under UV-light in some of the HCFIs. Cogenetic two-phase ($liquid_{aq}$+vapour) aqueous fluid inclusions could not be distinguished.

Homogenization temperature measurements were carried out with a Linkam THMSG 600 heating-freezing stage at the Department of Mineralogy, Geochemistry and Petrology, University of Szeged. Synthetic fluid inclusions were used for calibration at –56.6 °C, 0.0 °C and 374.0 °C. Data accuracy is ~ ±0.5 °C.

T_h measurements were performed on two fluid inclusion assemblages (FIA) in the void-filling quartz: on two-phase (liquid+vapour) primary aqueous fluid inclusions (FIA-1) and on three-phase (liquid+solid+gas), yellowish brown HCFIs (FIA-2). The aqueous fluid inclusions (FIA-1) homogenized to the liquid phase between 129–156 °C (n=11) (*Figure 1, G*) and the vapour phase did not reappear upon cooling. T_h measurements were performed on three-phase HCFIs with constant liquid-solid-gas volume ratios (FIA-2). The HCFIs (FIA-2) homogenized to the liquid phase between 76–114 °C (n=76) (*Figure 1, G*) and the solid phase did not show any phase changes during heating up to 200 °C.

Based on the similar petrographic features and the T_h values, FIA-1 of the current study probably corresponds to the primary fluid inclusion generation described by VETŐ-ÁKOS (1978). The colourless secondary fluid inclusion generation with variable phase ratios suggest entrapment from a heterogeneous fluid containing hydrocarbon and aqueous liquids. This fluid inclusion generation might correspond to those secondary fluid inclusions studied by VETŐ-ÁKOS (1978), which are colourless and/or contain three phases. Based on the yellowish brown colour and the similar T_h values presented in this study and in that of VETŐ-ÁKOS (1978) it can be supposed that the yellowish brown secondary fluid inclusions in her study correspond to the yellowish brown secondary HCFI generation of the current study; however, VETŐ-ÁKOS (1978) did not observe the presence of solids in the yellowish brown inclusions.

Although aqueous phase is present in a small amount in some of the fluid inclusions in this fluid inclusion generation, the constant liquid–solid–vapour volume ratio suggests the presence of a homogeneous fluid during entrapment: a small amount of H_2O was probably dissolved in the HC fluid, which exsolved upon cooling (GOLDSTEIN & REYNOLDS 1994). The solid phase is probably a hydrocarbon daughter phase that might have formed from the hydrocarbon liquid through irreversible processes (GOLDSTEIN 2003).

VETŐ-ÁKOS (1978) proposed in her study that the secondary inclusions contain CO_2 and/or aqueous liquid and CO_2 gas phases. Based on the blue fluorescence of both secondary fluid inclusion generations, it can be established that the fluid inclusions contain HC-bearing fluids (BURRUSS 1991); however, the presence of CO_2 cannot be excluded. The blue fluorescence might refer to the presence of mature hydrocarbons in the fluid inclusions, although other factors can also be responsible for the blue fluorescence of hydrocarbon fluids (OXTOBY 2002, and references therein).

Conclusion

Primary HCFIs at Árpádtető occur in saddle dolomite cements, which formed at elevated temperature (LUKOCZKI & HAAS 2013) suggesting that hydrocarbon migration occurred during deep burial and/or related to hydrothermal events, most probably during the Early Cretaceous when the Middle Triassic carbonates were deeply buried and magmatic events occurred in the Mecsek Mountains (VETŐ 1978, VETŐ-ÁKOS 1978).

Differences of the hydrocarbons entrapped in fluid inclusions at Árpádtető (colourless and yellowish brown HCFI generations) and at other localities (Pécsvárad, Pécs-Vasas, Kantavár) suggest multiple events of hydrocarbon migration and/or different source rocks. Chemical alteration of the hydrocarbons due to thermal or biogenic processes during migration or during/after entrapment could also have led to the differences currently observable at each occurrence. It remains an open question which of the five organic matter rich Late Triassic – Early Jurassic formations served as the source rock(s) for the studied HCFIs.

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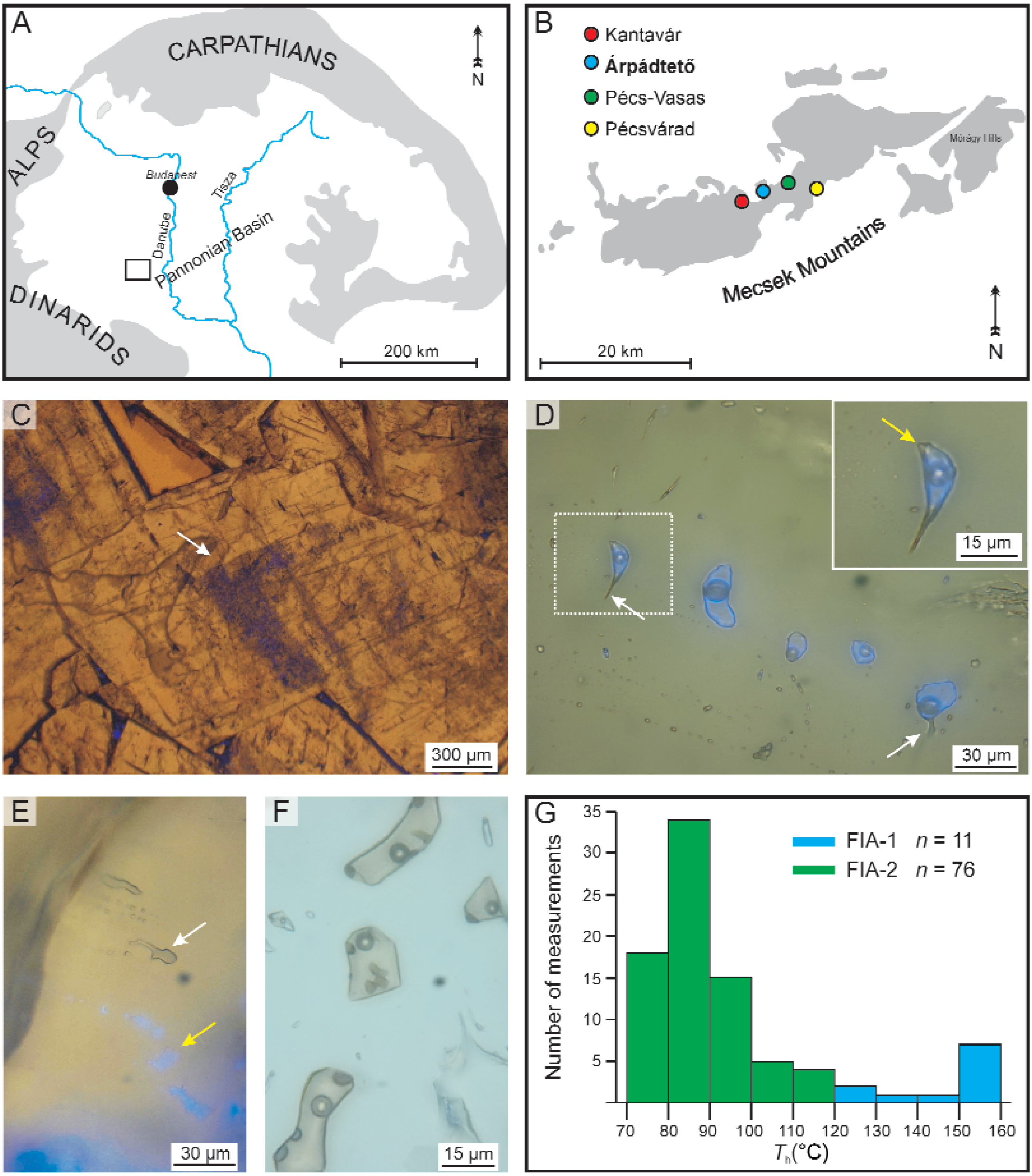
A
CARPATHIANS
ALPS
Budapest
Tisza
Danube
Pannonian Basin
DINARIDS
200 km
N
B
Kantavár
Árpádtető
Pécs-Vasas
Pécsvárad
Mórágy Hills
Mecsek Mountains
20 km
N
C
300 µm
D
15 µm
30 µm
E
30 µm
F
15 µm
G
FIA-1 n = 11
FIA-2 n = 76
Number of measurements
T_h(°C)