Downward infiltration of fluid into the high-grade basement of South Norway II: Composition and phase-behaviour of petroleum fluids at high temperature

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The high-grade Modum Complex, situated in the Precambrian basement ca. 0-30 km west of the Permian Oslo Rift, has experienced multiple events of retrograde and postmetamorphic fluid infiltration (Munz et al., 1994, 1995; Gleeson et al., 1998). This has resulted in the formation of complex vein systems and alteration of the high-grade rocks. Open veins with coarse, euhedral quartz represent the youngest generation, and they are often associated with albite and calcite veins. The quartz veins crosscut all metamorphic structures, but are older than N-S trending diabase dykes, which are presumably of Permian age. Abundant hydrocarbon inclusions occur within the quartz veins. Bitumen has been found in an albite-calcite breccia at one of the localities (Eakin, 1989). A δ^{13} C composition of -30.3% PDB suggest a biogenic origin for the bitumen (Eakin, 1989). This paper describes the occurrence and compositions of the hydrocarbon inclusions in the Modum Complex, and discuss possible mechanisms of their formation.

Classification of hydrocarbon inclusions

The vein quartz crystals show well developed growth zoning. Aqueous fluids occur as primary, pseudose-condary and secondary inclusions (Munz *et al.*, 1995; Gleeson *et al.*, 1998). The hydrocarbon inclusions mostly occur as secondary trails in the quartz veins. A small minority of the hydrocarbon inclusions may however be of primary or pseudosecondary origin. Secondary trails of hydrocarbon inclusions also occur in the high-grade gneisses.

A large range in hydrocarbon fluid compositions are present. On basis of petrography, microthermometry and preliminary gas chromatography (GC) results a classification into three groups of hydrocarbon inclusions can be made:

Group I inclusions occur as one-phase inclusions at room temperature, and show no fluorescence. The homogenisation temperature fall in the range -90 to -55° C, mostly to the liquid phase. The freezing/ melting of a solid occur in some of the samples, and the homogenisation of the samples, where a solid appear, is both to the liquid and vapour phase. The composition of these inclusions range from pure CH₄ (=C1), CH₄-CO₂ mixtures to C1-C2-C3 compositions.

Group II inclusions also occur as one-phase inclusions at room temperature. The inclusions show a weak dark blue fluorescence, when illuminated with ultraviolet light (wavelength = 365 nm). In some of the inclusions a colourless coating or viscous/semi-solid? patches occur. These show a bright yellow fluorescence. A black solid is also sometimes present in these inclusions. The homogenisation temperatures vary between -75 and -15° C. Hydrocarbons up to C9 have been detected by GC analyses.

Group III inclusions occur as multiphase (two-, three- or four-phase) inclusions at room temperature. Both vapour and liquid hydrocarbon phases are present. The liquid hydrocarbon phase shows a deep blue fluorescence. An aqueous phase is common in these inclusions. The volumetric proportions between the hydrocarbons and the aqueous phase are highly variable. A black solid is commonly also present in these inclusions. Homogenisation of the vapour and liquid hydrocarbon phases is always to the liquid phase at temperatures mostly between +20 and +60°C. Compositional data on this group of inclusions have not yet been achieved, but phase calculations show that the composition must be significantly more dominated by higher hydrocarbons than group II inclusions.

Gasoline range hydrocarbons

The preliminary GC results for group II hydrocarbon inclusions show fluid compositions consisting of n-

alkanes, branched alkanes, naphtenes and aromatic hydrocarbons in proportions typical for mature petroleum in sedimentary basins. Some selected component ratios in the gasoline range are represented in Table 1.

Discussion

Isochores for group I and group II inclusions have been calculated by combining information from microthermometry and GC analyses. The relatively large variation in homogenisation temperatures within each group, can be a result both from variation in composition and density, which can not be resolved on the level of individual inclusions. Considering this problem of resolution, the isochore calculations for group I and II inclusions show similar results with pressure in the range of 400-1200 bar at temperatures of $200-300^{\circ}$ C.

Munz et al. (1995) suggested that equilibrium between the one-phase hydrocarbon and aqueous fluids was not achieved. The presence of easily water soluble components like benzene and toluene in samples containing group II inclusions support this. Extensive water washing has not occurred.

Hydrocarbons in the gasoline range are not direct derivatives of biological precursors. It is generally accepted that gasoline range hydrocarbons originate by cracking of higher hydrocarbons. Cracking from biogenic, multicyclic naphtenes (sterane-, and terpane-related kerogen) has been suggested as mechanisms for formation of naphtenes in the C6-C7 range (Thompson, 1979), although this mechanism has been questioned (Mango, 1997). The similarities in composition between the group II inclusions and mature sedimentary petroleums would however suggest an ultimate biogenic source. Ongoing work on the group III inclusions may finally settle this question. The similar results for isochore calculations for group I and II inclusions suggest that the two groups (and most probably also group III inclusions) are linked in time to the same process. According to kinetic models for cracking of oil to gas at temperatures around 200°C 50-100% will be

TABLE. 1. Selected component ratios (weight based) for sample DOV3 (group II inclusions)

benzene/nC6	0.20
toluene/nC7	0.76
nC7/M-Cy-C6	0.98
(nC6+nC7)/(Cy-C6+M-Cy-C6)	1.76
Cy-C6/M-Cy-C5	0.97

nC6 = normal hexane nC7=normal heptane M-Cy-C5 = methylcyclopentane Cy-C6=cyclohexane M-Cy-C6 = methylcyclohexane

converted to gas. We propose that the variation in hydrocarbon fluid compositions at Modum, ranging from pure methane to liquid hydrocarbons, is a result from disequilibrium during fluid migration at high temperatures (>200°C). Local variations/fluctuations in pressure and temperature during the dynamic process of fluid migration, crack opening and sealing may produce the observed variation in fluid chemistry, due to the rapid kinetics of cracking at high temperatures. The final result of the process is the formation of $CH_4 \pm CO_2$ and bitumen. The high thermal maturity and carbon isotope composition of the bitumen (Eakin, 1989) is consistent with this interpretation.

References

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