



Research Article

ESTIMATION OF OIL CHARACTERISTICS: RESULTS BASED ON FLUORESCENCE PHENOMENON AND FLUID INCLUSION SYNTHESIS AND ANALYSIS

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ARTICLE INFO

Article History:

Received 17th November, 2016

Received in revised form 18th December, 2016

Accepted 4th January, 2017

Published online 28th February, 2017

Key words:

Fluid Inclusions, Hydrocarbons, Petroleum, Fluorescence, Microscopy

ABSTRACT

The accurate characterisation of the petroleum fluid entrapped in fluid inclusions in minerals is a considerable challenge for the researcher. Hydrocarbon bearing fluid inclusions (HCFI) are useful for learning about natural processes (in that e.g., migration of hydrocarbon fluids in petroleum basins), composition of fluids and p-T conditions in geological systems. Natural processes may be studied by creating synthetic inclusions. The research performed has three stages combining fluorescence features of oils and hydrocarbon fluids, synthesis of inclusions in salts as well. At the first stage – chemical properties and fluorescence features of oils were studied. The second stage was the synthesis of hydrocarbon(HCFI), aqueous (AQFI) and mixed inclusions in salts in temperatures below 60 °C. At the third stage, synthetic HCFI were analysed from the fluorescence point of view. A simple tool for estimation of oil characteristics in inclusions has been proposed as the result of the experiment, analyses and data compilation. Using this tool, an estimation of oil characteristics in some natural rock samples from petroleum reservoirs was conducted by use of a certain diagramme, and presented based on fluorescence colours of inclusions in uncovered samples from the Baltic Sea (Polish sector, onshore and offshore) and the Carpathians. The limitation of the results correspond to numerous factors that may influence fluorescence.

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INTRODUCTION

Hydrocarbon – bearing fluid inclusions (HCFI) are useful for learning about the processes, compositions, and temperature and pressure conditions in geological systems (Roedder 1984). They may give an explanation on migration of hydrocarbon fluids in petroleum basins (Stasiuk, 2002; Balitsky *et al.*, 2007). The study on HCFI offers a unique insight into the formation and development of petroleum basins (Bodnar, 1990; Nedkvitne *et al.*, 1993; Munz, 2001; Pironon and Bourdet, 2008; Blamey and Ryder, 2009). The use of fluorescence phenomenon in a wide variety of methodologies for the characterisation and analysis of HCFI is generally well established (Thiéry *et al.*, 2002).

A synthesis of inclusions represents also a good tool for understanding different natural fluid processes. The composition of mixtures in the inclusions must be known and cannot be changed since the trapping (Roedder, 1984). The synthetic inclusions are often used to calibration of analytical techniques. Their synthesis occurs in the laboratory conditions and in different combination of pressures and temperatures. For example Shelton and Orville (1980), Sterner and Bodnar (1984) and Zhang and Frantz (1987) obtained synthetic inclusions in quartz in high temperatures (220-850 °C) and pressures (0.5-7 kbar) for different CO₂, CH₄ and aqueous chlorine systems. They either healed fractures or form

overgrowths on the original crystals. Fang Lin and Bodnar (2010) made their experiments with determinations of PVTX properties of aqueous – methane synthetic inclusions. Some researchers synthesised inclusions in gypsum (Sabouraud-Rosset, 1969) and halite (David *et al.*, 1989) below 100°C under the atmospheric pressure. A reconnaissance study on synthetic saline and hydrocarbon fluid inclusions in calcite has been recently presented by Yong Chen *et al.* (2016). Pironon (1990) made a synthesis of hydrocarbon inclusions in low temperatures. McLimans (1991) mentioned the aqueous – hydrocarbon inclusions in calcite, without methodological details, however. Jarmołowicz-Szulc (1998) made her own synthetic experiment using different oils and solutions of chlorides. The importance of the synthetic inclusions has been proved by Teinturier *et al.* (2003).

The present paper based on that fore-last author's cited above, tries to combine both the fluorescence and synthesis tools and deals with the experiment of the synthesis of petroleum inclusions in low temperatures and at the atmospheric pressure in salts, studies of their fluorescence and evaluation of the maturity and density of oils both in synthesised samples and hydrocarbon inclusions in natural samples. As the final result, based on the fluorescence features and laboratory steps, a simple tool for some estimations of character of hydrocarbons in fluid inclusions trapped in the minerals/rocks is provided. The idea of using saline aqueous solutions and crude oils to the synthesis of inclusions is similar to that by

Yong Chen *et al.* (2016), the experimental conditions, however, differ a lot both in temperatures and pressures.

METHODOLOGY

The experimental procedure occurred in three stages. They are as following:

1. a determination of hydrocarbon composition of oils used further to the experiment;
2. the experiment of the synthesis of inclusions in salts in low temperature conditions;
3. a characterization of fluorescence of synthetic inclusions.

Fig. 1

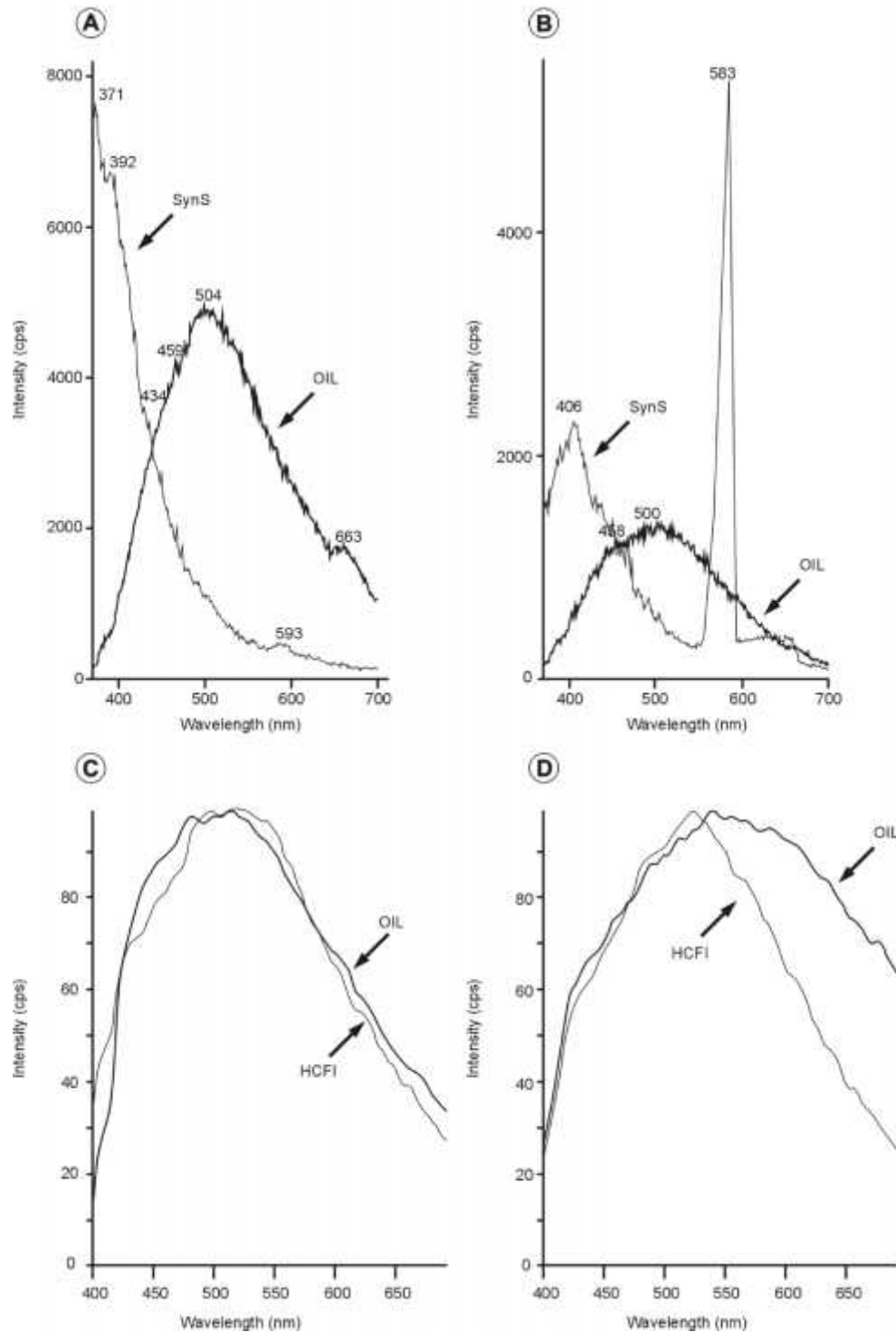


Fig.1 Fluorescence spectra of oils and hydrocarbon fluid inclusions (HCFI) – examples

A. Fluorescence spectra of oil from the a4 well and the synthetic sample B 2-21.

B. Fluorescence spectra of oil from the B 8-1 well and the synthetic sample B 8-1.

C. Fluorescence spectra of oil and dominant HCFI in Hibernia K-18. After Stasiuk&Snowdon (1997), modified.

D. Fluorescence spectra of oil and dominant HCFI population in Hebron J-13. After Stasiuk&Snowdon (1997), modified.

At the first stage, features of oils used further to the synthesis were determined. Analyses were performed by use of the chromatography method. The saturated hydrocarbon fraction was analyzed by use of the gas chromatograph with a mass spectrometer (GC/MSD, Hewlett-Packard). The following studies were conducted: - n-alkane and isoprenoid determinations by application of the external standard; - a qualitative analysis of biomarkers in fraction I. At the second stage, solutions of halogens and sulphates were used. The temperature corresponded to the values below 60°C under atmospheric pressure (Jarmołowicz-Szulc, 1998). The third stage comprised studies on samples obtained experimentally. The fluorescence phenomenon of synthetic inclusions was observed using a Nikon polarising microscope with a fluorescence device (e.g., Jarmołowicz-Szulc and Dudok, 2005; Jarmołowicz-Szulc, 2010). Results were combined with other data taken from bibliography (Pironon, 1990; Stasiuk and Snowdon, 1997) and they were furthermore used to characterisation of natural inclusions.

Fluorescence studies

Fluorescence studies of oils were conducted prior to the experiment of synthesis and salt crystal crystallisation. These analyses were performed by M. Czaja from the Silesian University in Sosnowiec in JobinYvon-Spex. Spectrofluorometer “Fluorolog” FL 3-12. Oil spectra were obtained.

The fluorescence studies on inclusions followed the experiment of synthesis. They were conducted by means of the Nikon microscope with the fluorescence device and ultra violet and blue light filters (365 nm and 480 nm, respectively). The synthesized salt crystals were placed under the microscope in the transparent light to fix inclusion position and to observe a fluid character. When found, each inclusion was observed in the reflected light, using both ultraviolet and blue filters. Diagnostic was the fluorescence in ultraviolet, while the blue light fluorescence, yellow in colour, is only a proof of hydrocarbon presence (Jarmołowicz-Szulc, 2001; Jarmołowicz-Szulc *et al.*, 2012).

An attempt was also made at this stage to analyse spectra of synthetic oil inclusions in crystals. Due to the technical reasons, however, it was impossible to focus on inclusions themselves. The spectrofluorometer was not equipped in the microscope. The crystals were, therefore, crushed and analysed “en bloc”.

Description of the synthesis

The solutions of halogens may be used for the synthesis of hydrocarbon – aqueous inclusions in low temperatures (Li and Mai, 1992). The halogens display a high solubility in low temperatures as it was presented by David *et al.* (1989). The experiment was conducted based on the procedure proposed by Pironon (1990). Two types of chlorides were used for the experiment – the potassium and sodium chlorides. These chlorides have a solubility of 25.5 g/100g of the saturated solution and 26.4/100 g, respectively. The adequate weight amount of the chloride was dissolved in water aiming at a solution oversaturated at room temperature. This solution was further heated until saturation point with a constant monitoring of the temperature. Close to the saturation point, a portion of oil was injected into the solution. The conditions were generally below 60°C, so that the hydrocarbons

remained stable. The mixture of both liquids was further cooled aiming at crystallization of salt crystals with fluid inclusions. Crystals were formed at the contact of both phases.

Five types of oils were used to the experiment – three from the Baltic Sea wells, one from the arnowiec region (N Poland, onshore) and one from the Gotland Island. Four first oils mentioned were colourless, while the last was brown in colour.

The experimental part was conducted twice for each oil type and the crystallisation time varied from days to a month. The synthetic crystals were further observed under binocular, and manually selected. Crystals rich in inclusions were separated for further studies.

RESULTS

Three stages of the analyses performed have resulted at least in three types of data obtained, namely: analytical spectra, salt crystals and fluid inclusions therein. The crude oils used for the synthesis origin from different wells. The reservoir rocks for four of them are of the Cambrian age and Silurian for one. By means of chromatographic petroleum analysis prior to the fluorescence studies it was attempted to observe a relation between visual fluorescence and chemical comparison of oils entrapped as hydrocarbon inclusions. The selected fluorescence spectra for oils are shown in figure (Fig. 1), while other parameters characteristic for such spectra as: the spectrum range, L_{max} value and Q parameter (Li and Mai 1992) are compiled in table 1.

Table 1 Compilation of characteristic parameters

Sample	L max	Q	API	Source
Oil B8-1	500	0.22		Present paper
Oil B3-17	492	0.18		Present paper
Oil a-4	504	0.35		Presentpaper
Oil Go	500	0.24		Presentpaper
Oil a-4 SFI	458	0.14		Presentpaper
Oil B3-17 SFI	487	0.13		Presentpaper
1	463	0.35		PirononandPradier (1992)
9	536	1.44		PirononandPradier (1992)
20166	560	0.65	31	Stasiuk andSnowdon (1997)
20188	580	0.75	25.9	Stasiuk andSnowdon (1997)
20656	595	0.95	17.5	Stasiuk andSnowdon (1997)
20121	460	0.10	44.5	Stasiuk and Snowdon (1997)

SFI – synthetic fluid inclusions in the synthetic sample (SynS in Fig.1).

The range of the wave spectra is generally 370-700 nm at the extinction level of 350 nm. The fluorescence intensity is high and reaches 6000 counts per second. The shape of the emission spectra of oils is rather homogeneous with a distinct maximum at about 500 nm, although other less distinct peaks occur, too. They may be strengthened in other conditions of spectrum measurements (Maria Czaja, University of Silesia, oral information). In the first approximation of interpretation, this fact may be connected with the extinction of other compounds in the oil (or inclusion). It demands more detailed studies, however, the range of which is wider than the limits of the present paper.

As it is seen in the figure (Fig. 1, A, B) and in the table (Table 1) – at same counting conditions – the spectra for oils are generally similar (B 3-17 and a- 4 shown). They resemble those present in bibliography (e.g., Stasiuk and Snowdon, 1997). Other parameters are as follows: maximum wave

length L_{max} (between 450 and 600 nm, corresponding to fluorescence colours limits white – red, respectively), a ration red/green Q (between 0 and 1), API gravity (between 17.5 and 44.5 API). The oils in synthetic fluid inclusions (SFI) display somehow lower and diversified spectrum values. That is because the analyses were performed on whole samples, not on inclusions themselves. In contrary to that the spectra by Stasiuk and Snowdon (1997) represent exactly crude oils and HCFI (Fig. 1, C, D).

Salt crystals synthesized are typical in shape, i.e., they are regular. They display a variable size changing from some tenths of millimeter to some millimeters (2-3 mm). These crystals contain the following types of inclusions (Fig. 2): - aqueous (AQFI) – Fig. 2.A; - mixed hydrocarbon – aqueous (Fig. 2 B); - oil (HCFI) – Fig. 2 A, C, D. The size of the HCFI oscillates between 1 and 100 μm . The AQFI are generally larger than those HCFI and more abundant. Mixed aqueous – hydrocarbon inclusions contain two liquids in different mutual proportions, and display diversified size and pseudo two phase character.

Visual fluorescence features are documented by microphotographs and shown as examples in figures (Fig. 2; Fig. 3). The colour and a refractive index of inclusions are close to those of sylvite crystals. The detection of the colourless oil inclusions in crystals in the transmitted light is therefore difficult. They are well seen due to their fluorescence in the reflected light.

The distinctly coloured Gotland oil inclusions could be easier to follow. Identification of these inclusions in the transmitted light is already possible (dependent on crystal size, however), being confirmed in the fluorescence, evident both in ultraviolet and blue lights, as it has been already mentioned above (Fig. 3).

Fig. 2

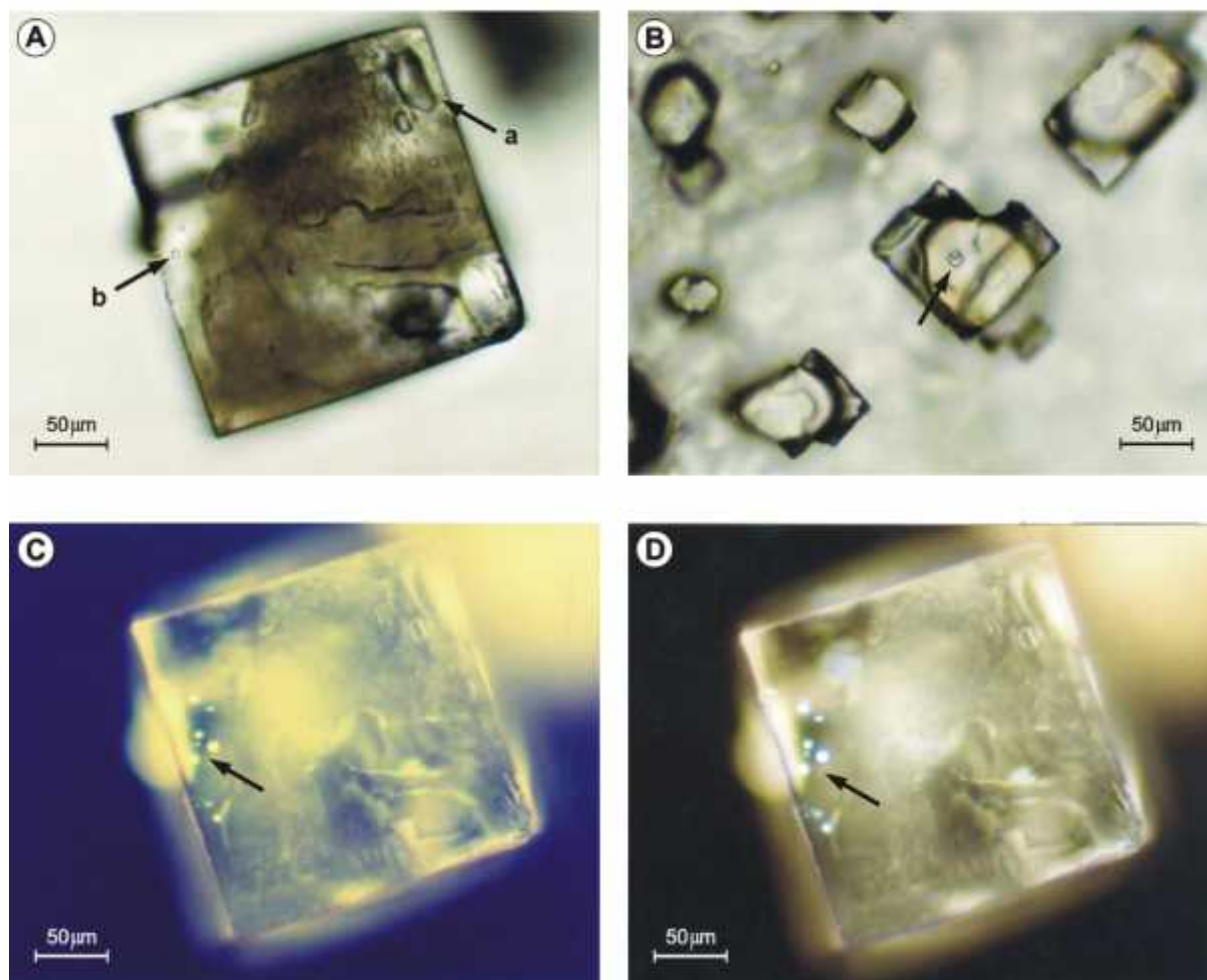


Fig.2 Fluid inclusion types after the synthesis experiment

1. Aqueous (a) and hydrocarbon (b) inclusions in sylvite crystal. Polarising microscope, transmitted light. Arrows point the inclusions.
2. Two-phase fluid inclusion aqueous – hydrocarbon in sylvite crystal. Polarising microscope, transmitted light. An arrow points the inclusion.
3. Yellow fluorescence of hydrocarbon inclusions in blue light Objects shown by an arrow – same as (b) in figure A. Reflected light.
4. White-blue fluorescence of hydrocarbon inclusions. Objects shown by an arrow – same as (b) in figure A. Reflected light, ultraviolet.

Fig. 3

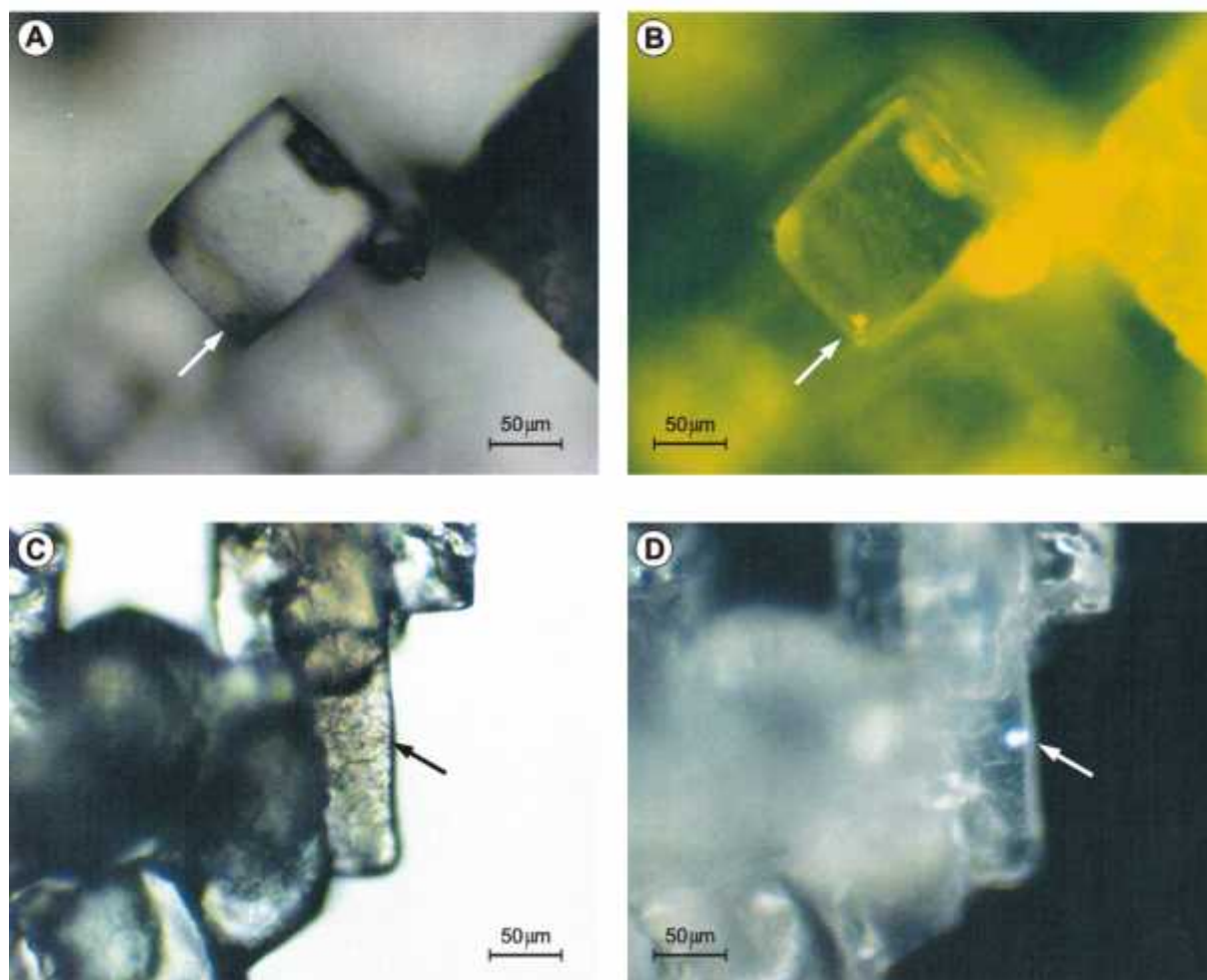


Fig.3 Hydrocarbon fluid inclusion types (HCFI) in synthetic sylvite crystals

- A. Hydrocarbon fluid and aqueous inclusions. An arrow points the inclusions. The arnowiec oil was used for synthesis. Polarizing microscope, transmitted light.
- B. Yellow fluorescence of hydrocarbon inclusion in blue light. Objects shown by an arrow – same as in figure A. Reflected light.
- C. Hydrocarbon inclusions in sylvite crystal. Polarizing microscope, transmitted light. An arrow points the inclusions. Polarizing microscope, transmitted light.
- D. White fluorescence of hydrocarbon inclusions. Objects shown by an arrow – same as in figure C. The Gotland oil was used for synthesis. Reflected light, ultraviolet.

RESULTS - PRACTICAL ASPECTS OF SYNTHESIS OF HCFI AND FLUORESCENCE

The present experiment is strictly connected with the problem of fluorescence of aromatic compounds, and oil maturity and density. Using the fluorescence spectra and data on composition obtained, a special diagram for estimation of characteristics oil in inclusions can be constructed (Jarmolowicz-Szulc, 2001). It may be applied to the estimation of the oil maturity in inclusions (HCFI) and the approximate fluid density in the hydrocarbon inclusions based on the visual microfluorescence colors (Fig. 4). However, the filling of the inclusion must not be biograded. Knowing one of the fluorescence components (e.g., fluorescence colour), it is possible to find out estimates of other parameters.

The two-phase HCFI from the Baltic Sea sandstone which show a white fluorescence seen in microphotographs (A) in Fig. 4 contain a high mature oil, light (density of about 41-45 API).

The HCFI associations (HCFI 1, HCFI 2) from the Carpathian quartz veins which show a fluorescence in different colors seen in microphotographs (B) in Fig. 4 contain different oils:

a high mature oil, light (density of about 41-45 API) which displays a white luminescence; less mature, naftene in chemical composition, medium in gravity oil which has yellow luminescence, respectively.

The two-phase HCFI from the Carpathians which show a white and orange fluorescence seen in microphotographs (C) in Fig. 4 contain a high mature oil, light (density of about 41-45 API) and traces of very heavy hydrocarbons, very low in maturity.

A huge naturally coloured fluid HCFI with solid phases is shown in transmitted light (upper figure, 4 D) and in fluorescing mode (white fluorescence – lower figure, 4D). It comes from the Marmarosh diamond from the Carpathians in Ukraine. According to the results compiled in Fig. 4, the oil filling this inclusion is highly mature, of paraffine character, with Q close to 0.1 and API gravity of about 44.5 – 45, i.e. light.

Based on the proposed method, conclusions may be drawn on the character of other natural samples (Fig. 5), both for HCFI associations in dolomite from the deep well in the Polish Lowlands (the BMB zone) and secondary and primary

inclusions from the Baltic Sea quartz arenite (the B 8-1 well, Fig. 5, C, D).

Hollerbach (1981), the hydrocarbons fluoresce and their fluorescence colour may be used as a maturity factor

Fig. 4

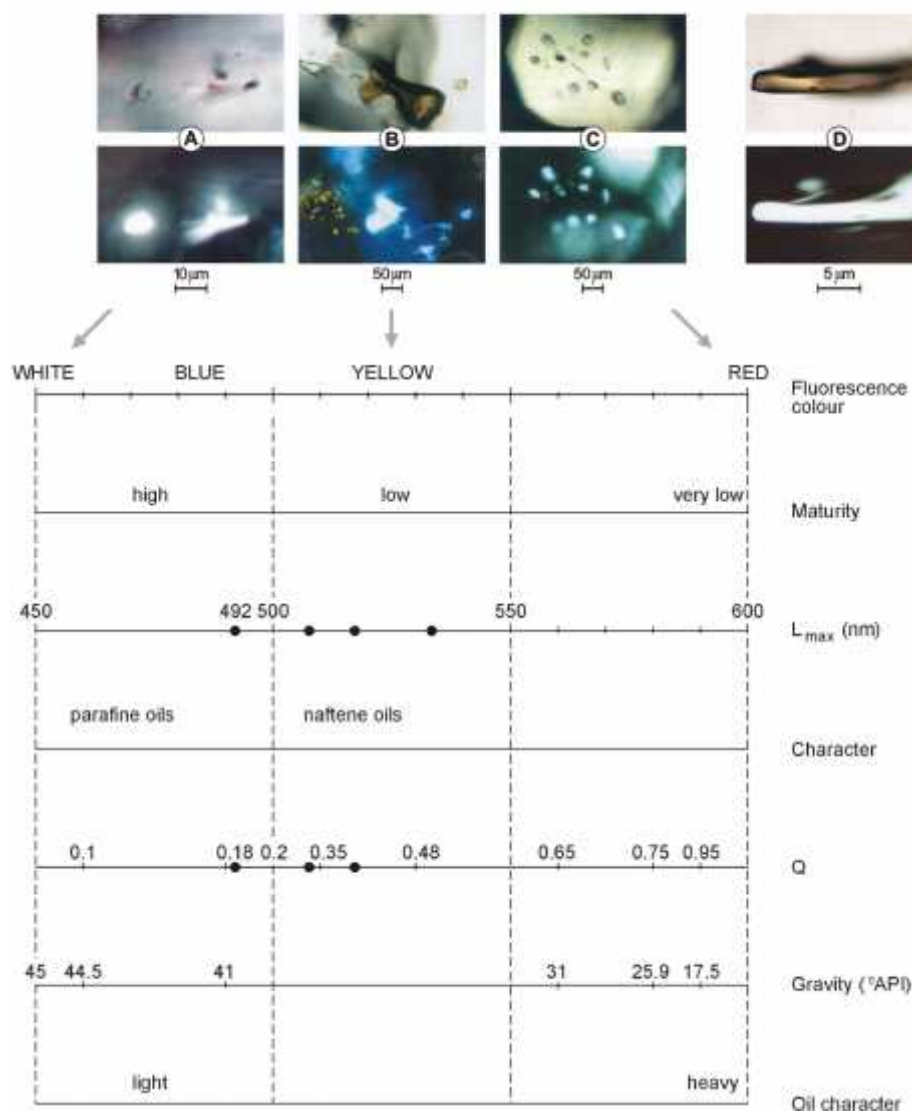


Fig.4 Correlation diagramme of fluorescence colours, maturity, Lmax, Q and oil gravity in inclusions

- A. Hydrocarbon fluid inclusions in transmitted light (upper figure) and in fluorescing mode (white fluorescence – lower figure). The quartz arenite from the arnowiec well.
- B. Hydrocarbon fluid inclusions in transmitted light (upper figure) and in fluorescing mode (yellow luminescence of an association of small inclusions and white fluorescence of a part of a huge inclusion – lower figure). The Marmarosh diamond, Carpathians.
- C. Methane– rich inclusions in the quartz with a small volume of heavier hydrocarbons in transmitted light (upper figure) and in fluorescing mode (bluish fluorescence for methane and orange-red fluorescence for other HC - in lower figure). Quartz, Carpathians.
- D. A huge naturally coloured fluid HCFI with solid phases in in transmitted light (upper figure) and in fluorescing mode (white fluorescence – lower figure). The Marmarosh diamond, Carpathians, Ukraine.

Based on the fluorescence microphotographs of inclusions enclosed in other papers (e.g., Stasiuk, 2002; Jarmolowicz-Szulc, 2005; Blamey and Ryder 2009) a potential character of oils entrapped may be estimated there, with a full understanding of a lot of factors which can influence these estimations.

DISCUSSION

Hydrocarbon and aqueous inclusions may be trapped in the crystal during its growth and/or due to the healing of microfissures in the rock history in the sedimentary basin (Roedder, 1984). As it was defined by Hageman and

(McLimans, 1987; Videtich *et al.*, 1988; Guilhaumou *et al.*, 1990; Blamey and Ryder, 2009). The intensity of fluorescence is proportional to the concentration of fluorescing compounds called chromophores in their low accumulation and it decreases with the increase in the chromophores number (Atkins, 1982). There exist some types of chromophores that influence the emission in natural oils. The polycyclic aromatic hydrocarbons are intensively fluorescent. The intensity of this luminescence increases towards longer wave values with the increase of aromatic chains in the molecule (Burrus, 1991). The fluorescence spectra are used to the classification of the organic matter (Robert, 1979; Stasiuk, 2002) and for the

characterization of its chemical composition and/or maturity (Hageman and Hollerbach, 1981; Bertr and *et al.*, 1986). The fluorescence microscopy is often used for a differentiation of intracrystalline fissures filled in with oil (e.g., hydrocarbon fluid inclusions versus aqueous fluid inclusions; Burrus, 1981; Karlson *et al.*, 1993; Jarmołowicz-Szulc, 2001; Munz, 2005; Jarmołowicz-Szulc *et al.*, 2012). Some authors correlate the fluorescence colours with API degree which is referred to oil density (McLimans, 1987; Bodnar, 1990; Tsui, 1990; Jarmołowicz-Szulc, 1998, 2001; Stasiuk, 2002; Nandakumar and Jayanthi, 2016). The independent control is, however, needed since a lot of factors can influence the fluorescence, and the results of parameter estimation must be treated really as the first approximation (Jarmołowicz-Szulc, 1998, 2001, 2016). Barres *et al.* (1987) and Pironon and Pradier (1992), for example, are of opinion that the determination of the chemical composition of fluorescing inclusions based on microfluorescence spectrometry is impossible. The color of fluorescence is influenced by many factors that make a composition diagnosis very risky (Blamey and Ryder, 2009). The precise diagnosis of chemistry and maturity is almost impossible.

However, the presented correlations enable some preliminary, non-destructive and quick estimations. The proposed method is quick and easy, much less complicated and spectrometer demanding than that reported by Przyjalowski *et al.* (2005). The results must be treated as an approximation, as it has been already underlined. Still, it can be concluded that the higher the API degree, the larger a shift towards white-blue color emission in fluorescence (Fig. 4). Some trends may be also observed both from the present results and bibliography (Pironon and Pradier, 1992; Stasiuk and Snowdon, 1997; Jarmołowicz-Szulc 2001). For inclusions – a decrease in L_{max} and Q occurs with an increase of total hydrocarbons and concentration of saturated fractions. A progressive increase in L_{max} and Q is correlated with an increase in aromatic hydrocarbons and concentrations of resins and asphaltenes. Recently Nandakumar and Jayanthi (2016) have also presented an empirical tool for predicting APIG of oils and documented it on the samples from the well from Mumbai offshore basin, India. Fluorescence emission of oil in HCFIs was recorded by these authors in the region of 406–720 nm, i.e., wider than reported in the present paper, and they give calculated spectral emission ratios at F620/F560 in the plot of API gravity known oils. The APIG of unknown samples they propose to infer from the algebraic expression linking emission spectra to APIG for known crude oil samples.

FINAL CONCLUSIONS

Based on bibliography and studies conducted the following final conclusions may be drawn:

1. It is possible to produce synthetic hydrocarbon fluid inclusions in salt crystals from crude oils and saturated salt solutions;
2. L_{max} and Q factor values for synthetic inclusions (SHCFI) in salt crystals are in ranges of 480 – 550 nm and 0.1 – 0.3, respectively.
3. A compilation of different fluorescence parameters with chemical composition and thermal maturity of oils results in a simplified estimation features of hydrocarbon fluids entrapped as inclusions.

4. The fluorescence colours exhibit a positive correlation with the API gravity, which gives some limits for the estimation of natural hydrocarbon fluids.
5. Fluorescence features of synthetic hydrocarbon inclusions display a progressive shift toward a longer wave length with an increase of aromatic hydrocarbon content and decrease in maturity.
6. Fluorescence features of HCFI may give some information on composition and density of oils entrapped. Best fit is obtained for not altered and hermetic closed inclusions.

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