

## FLUID INCLUSION STUDY OF THE GNEISS FROM THE BOREHOLE NAGYATÁD K—1, 11, SW TRANSDANUBIA [HUNGARY]

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### ABSTRACT

Fluid inclusions in synkinematic quartz lenses, segregations and those in the matrix quartz were studied from the 11-th core of the Nagyatád-K-1 borehole. This one is bored in the Somogy—Dráva Basin, SW-Hungary. It explored a polymetamorphic staurolite-kyanite-garnet gneiss mass. The studied fluid inclusions can be divided into three groups according to their composition: (1)  $H_2O + 1.7—14.3$  NaCl equivalent weight % salt, (2)  $CO_2 + / - CH_4$  and/or  $N_2$  [ $X_{CH_4-N_2} \cong 0.1$ ], (3) mixed  $CH_4-N_2-CO_2$ .

The peak P—T conditions of two metamorphic phases were inferred from geothermobarometric data of ÁRKAI [1984] and the obtained fluid inclusion data. The inferred P—T conditions of the first metamorphic phase were about 890—900 MPa and 550 °C. The fluid, existed at these P—T conditions, was a  $CO_2$  rich one. The maximum pressure and temperature, reached during the second stage of metamorphism was determined by the method of intersecting isochores of two immiscible fluids, trapped simultaneously. On the basis of this method the obtained pressure was 240—260 MPa and the temperature about 540 °C. All the three compositional types were present during the second metamorphic event. The third metamorphic stage was a very low-low grade one with a simple, dilute aqueous solution.

### INTRODUCTION

The studied Nagyatád-K-1-11. sample derives from a hydrocarbon exploratory well of the National Oil and Gas Industrial Trust. The exposed staurolite-kyanite-garnet gneiss belongs to the crystalline basement of the Somogy Dráva Basin. The exact location of the borehole is shown on the *Fig 1* after ÁRKAI [1984].

The aim of this paper is to provide further data on the polymetamorphic P—T history and fluid evolution of the crystalline basement of the Somogy—Dráva Basin on the basis of fluid inclusion study. This paper is a first attempt in this topic in Hungary.

The obtained fluid inclusion data and the available geothermo-barometric results made it possible to construct a P—T path for the polymetamorphic history of the basement.

### GEOLOGY AND PREVIOUS DATA

The Somogy—Dráva Basin is situated in south western Transdanubia, as it is shown on the *Fig. 1*. Its crystalline basement is known only by hydrocarbon exploratory wells, because it is covered by thick neogene-quaternary sediments.

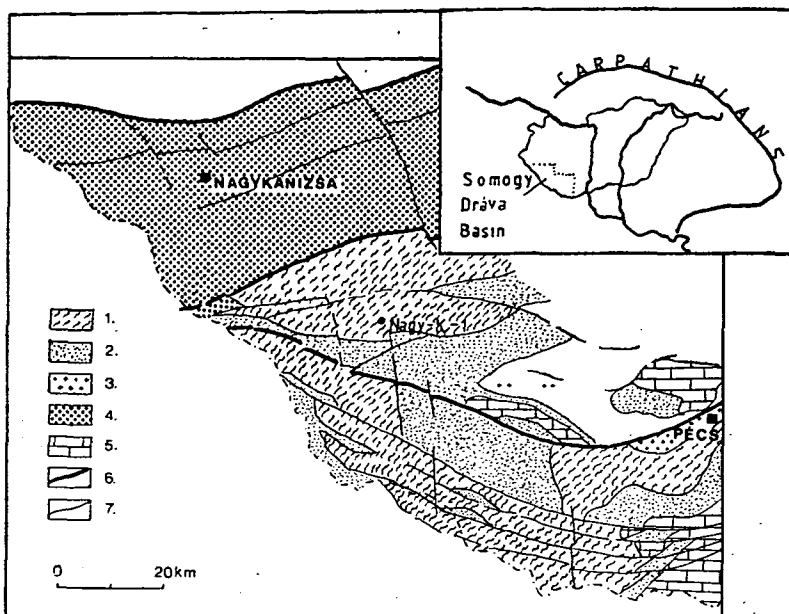


Fig. 1. Simplified geologic sketch of the pre-Tertiary formations of the Somogy—Dráva Basin with the locality of the Nagyatád-K-1 borehole taken from ÁRKAI [1984]. 1. = polymetamorphic basement, 2. = Paleozoic in general, 3. = granitoid rocks, 4. = Permian-Mesozoic sedimentary rocks of the northern part of the Somogy—Dráva Basin, 5. = Mesozoic in general, 6. = major tectonic lines, 7. = subordinate tectonic lines

The crystalline basement of the Somogy—Dráva Basin is connected with the Slovenian Papuk—Jankovac migmatite-metamorphite and the Psunj—Katjevo granite-metamorphite complexes towards south-southwest. On the opposite direction it is in connection with the Transylvanian Midmountains through the Göröcsöny—Mórág complex and the polymetamorphic basement of the Hungarian Great Plain [SZEDERKÉNYI, 1974; JANTSKY, 1979; ÁRKAI ET AL., 1985].

The most frequent rock types are micaschist, gneiss or milonite, blastomilonite originated from the rocks mentioned above. The amphibolite and amphibole-gneiss is subordinate. The detailed description of the different rock types and a more detailed geologic map of the crystalline basement is published by ÁRKAI [1984], see this study for details.

The rock forming minerals of the studied Nagyatád-K-1, 11 staurolite-kyanite-garnet gneiss are plagioclase, quartz, biotite, muscovite. The rock contains garnet, kyanite, staurolite, carbonate minerals, chorite, zircon, rutile, apatite, pyrite, and dispersed graphite as accessory minerals.

The case history of the Somogy—Dráva Basin and the adjoining areas can be summarized according to SZEDERKÉNYI [1974, 1976], JANTSKY [1979], LELKES—FELVÁRY and SASSI [1981], and ÁRKAI [1984], ÁRKAI *et al.* [1985].

The parent rock of the different micaschist and gneiss types may have been pelitic-psammitic sediment. However, in case of some gneiss types of mainly potash feldspar-plagioclase-quartz composition, the magmatic origin cannot be excluded either. The precursor of the amphibolite and amphibole-gneiss may have been a

certain kind of basic-neutral magmatic rock. The exact age of the parent rocks is unknown. According to suppositions it might be precambrian or early paleozoic.

The first metamorphic event was a Barrovian type one with kyanite-staurolite-garnet-biotite-muscovite-plagioclase [sometimes with sillimanite] mineral assemblage. ÁRKAI [1984], ÁRKAI *et al.* [1985] determined the peak P—T conditions of this event from 5 samples, belonging to the crystalline basement of the Somogy—Dráva Basin, using a plagioclase-garnet-biotite-muscovite geothermobarometer [GHENT and STOUT, 1981] and a hornblende-plagioclase geothermobarometer presented by PLJUSNINA [1982]. He obtained 590—890 MPa pressure and 550—600 °C for the micaschist and gneiss samples and 750 MPa and 507 °C for the amphibolite. The age of the first metamorphic event is thought to be Precambrian, Caledonian or early Hercynian.

The second stage of the metamorphism was a low pressure, medium grade one which can be characterized by andalusite-staurolite-garnet-biotite-muscovite mineral assemblage. During this event a migmatitisation and a granitisation took place in the Mórógy unit and in some places of the crystalline basement of the Hungarian Great Plain. The age of the second stage is defined by isotope geochronological data [BALOGH *et al.* 1981]. The inferred age of the metamorphism and the granitisation is Hercynian. BUDA [1972] reported late hercynian potash metasomatism.

The first two metamorphic events were followed by a low-very low grade one which locally caused milonitisation of the preexisting rocks. The typical mineral assemblage of this phase is quartz-sericite-chlorite and carbonate minerals. The age of the third event may have been late Hercynian.

#### SELECTING OF SAMPLES, SAMPLE PREPARATION

The fluid inclusion studies were carried out on synkinematic quartz segregations, which are concordant with the schistosity. Besides these inclusions 20 aqueous inclusions were also measured from the matrix quartz grains. Fluid inclusions in other minerals than quartz were not observed. The selection and preparation of the samples were carried out in accordance with the method proposed by HOLLISTER *et al.* [1981].

All measurements were conducted on doubly polished 100—150 $\mu$  thick samples, using a Chaixmeca heating-freezing stage [PORY *et al.* 1976] at the Department of Mineralogy of the Eötvös Loránd University. The reproducibility of the measurements was  $\pm 0.1$  °C in the negative and  $\pm 0.2$  °C in the positive temperature interval. All microthermometric measurements were made during the heating cycles. The correction of the obtained data was carried-out using the Inc. calc- computer program of GATTER and MOLNÁR [unpubl.].

The Raman spectroscopic analyses were run in Novosibirsk at the Institute of Geology and Geophysics of the Siberian Branch of Academy of Sciences of the Sovietunion by A. SHEBANIN on a Jobin Yvon instrument.

#### MODE OF OCCURENCE AND TRAPPING SEQUENCE]

The trapping sequence and the following grouping of inclusions were deduced from textural observations on the basis of textural criteria, published by SWANENBERG [1980], TOURET [1981], ROEDDER [1984] and OLSEN [1987]. The fluid inclusions were grouped in four categories from the oldest [group A] to the youngest ones

[group D]. The trapping sequence of the inclusions is not always clear; there are overlaps between the four groups. An example for the mode of occurrence of the A, B and C type CO<sub>2</sub> inclusions is presented on the Fig. 2.

*Type A:* Solitary inclusions and those in small groups both in the synkinematic quartz lenses and in the matrix quartz grains are considered to belong to this group. Both CO<sub>2</sub> and H<sub>2</sub>O inclusions belong to the type A. The observed clusters of inclusions in all cases were intragranular. The inclusions are rounded or have negative crystal shape. The negative crystal shape is more characteristic for the CO<sub>2</sub> inclusions than for the H<sub>2</sub>O rich ones. The CO<sub>2</sub> inclusions are between 5 μm and 10 μm in size, while the H<sub>2</sub>O-rich ones range from 8 μm to 12 μm in longest dimension.

*Type B:* Both CO<sub>2</sub> and H<sub>2</sub>O rich inclusions belong to this group. The inclusions are similar in size to those being in the group A. The shape of the inclusions tends to be more spherical or ellipsoidal and there are less negative crystal like inclusion than are found in the group A. The inclusions form trails along healed microcracks which do not cross grain boundaries. There are no CO<sub>2</sub> rich and aqueous inclusions together along the same trail.

*Type C:* All of the compositional groups are observed in this type. There are CH<sub>4</sub>—N<sub>2</sub>—CO<sub>2</sub>, aqueous and CO<sub>2</sub> rich inclusions along healed fractures which cross the grain boundaries and show the features of a mature fracture OLSEN [1987].

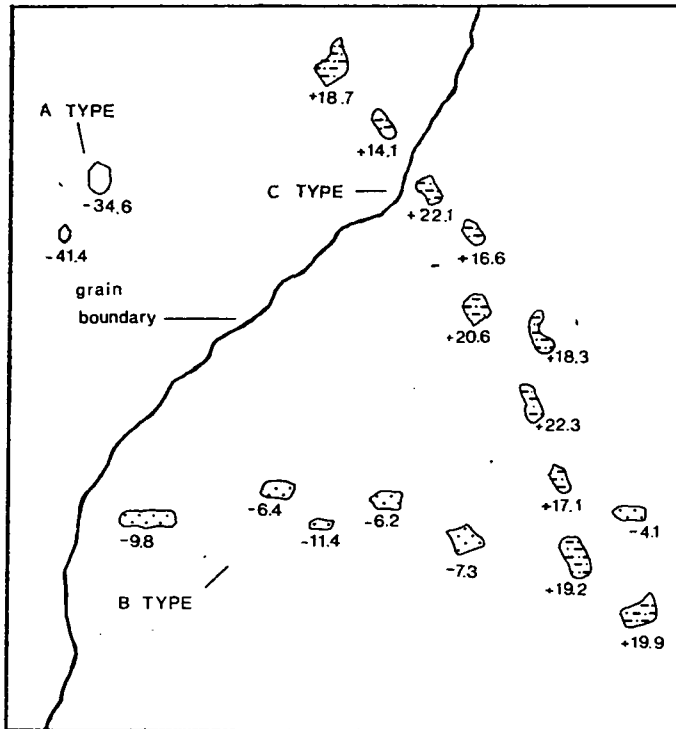


Fig. 2. An example for the mode of occurrence of the A, B, and C type CO<sub>2</sub> inclusions. The numbers indicate the homogenisation temperature of the inclusions in °C

O = A type CO<sub>2</sub> inclusions,  
 .... = B type CO<sub>2</sub> inclusions,  
 -.-.- = C type CO<sub>2</sub> inclusions.

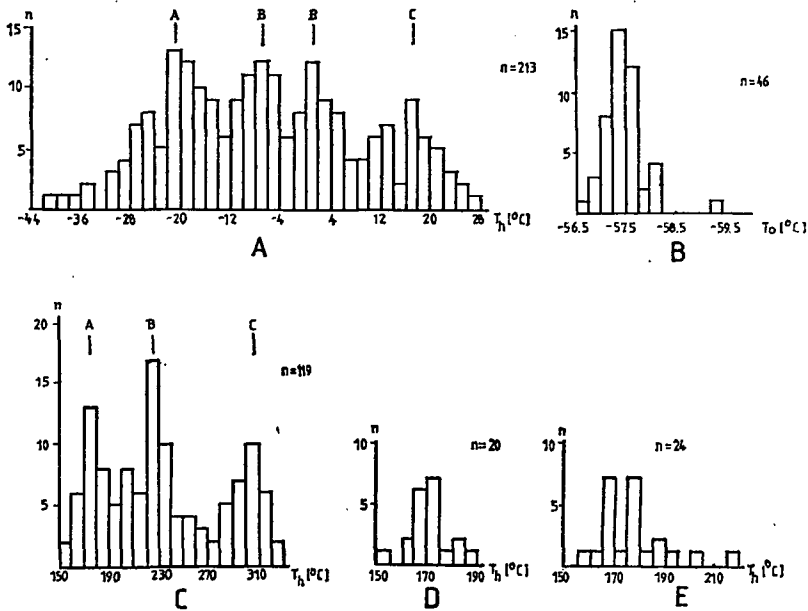
The inclusions are ellipsoidal or irregular in shape and their size ranges from 9  $\mu\text{m}$  to 16  $\mu\text{m}$ . The C type  $\text{CO}_2$  rich inclusion trails crosscut the B type ones at the angle of  $50^\circ$ – $70^\circ$ .

*Type D:* There were only  $\text{H}_2\text{O}$ -rich inclusions observed in this group. The trails of inclusions along immature healed fractures [OLSEN 1987] crosscut the grain boundaries and contain sometimes hundreds of well oriented, irregular or amoeba-like inclusions of various size, up to 50  $\mu\text{m}$ .

#### FLUID INCLUSION DATA

##### *$\text{CO}_2$ -rich inclusions*

The  $\text{CO}_2$  rich inclusions were identified by melting temperatures close to  $-56.6^\circ\text{C}$ . Most of the inclusions show freezing point depression to a minimum of  $-59.4^\circ\text{C}$  [Fig. 3b]. They may contain a minor amount of either  $\text{CH}_4$  or  $\text{N}_2$  up to 0.1 mol% [BURRUSS, 1981; KERKHOFF, 1988]. However, the highest density  $\text{CO}_2$  inclusion measured by Raman spectroscopy, does not contain any  $\text{CH}_4$  or  $\text{N}_2$  [Table 1]. The homogenisation temperature of the  $\text{CO}_2$ -rich inclusions range from  $-41.4^\circ\text{C}$  up to  $+26.5^\circ\text{C}$  [Fig. 3a]. All inclusions homogenized to liquid phase. There are no  $\text{CO}_2$ -rich inclusions observed in the matrix quartz, they seem to be restricted to the quartz segregations where they tend to separate from the  $\text{H}_2\text{O}$ -rich ones.



*Fig. 3.* Distribution diagrams:  $n$  — number of measurements,  $T_h$  — homogenisation temperature,  $T_0$  — melting temperature. A:  $T_h$  distribution diagram of A, B and C type  $\text{CO}_2$  inclusions. B: Distribution diagram of melting temperatures of  $\text{CO}_2$  inclusions. C: Distribution diagram of homogenisation temperatures of A, B and C type aqueous inclusions in the quartz segregations. D: The distribution diagram of  $\text{H}_2\text{O}$  inclusions in the matrix quartz. E:  $T_h$  distribution diagram of D type aqueous inclusions

Results of the laser Raman spectroscopic analyses.  
Analyst: A. SHEBANIN

Sample	$T_h(^{\circ}\text{C})$	$T_o(^{\circ}\text{C})$	$\text{CO}_2$	$\text{CH}_4$	$\text{N}_2$
N-2/8	-41,4	-56,9	100	n.d.	n.d.
N-II/1	-63,9C	-75,0	25	39	36

$T_h$  = homogenisation temperature,  $T_o$  =  $\text{CO}_2$  melting temperature, C = critical homogenisation, n.d. = not determinable. All compositional units are given in mole percent.

A computer program, "CO<sub>2</sub>", published by NICHOLLS and CRAWFORD [1985], utilizing the equation of state of ANGUS *et al.* [1976], was used to determine the densities and the isochores of the CO<sub>2</sub>-rich inclusions. The isochore of the most dense CO<sub>2</sub> inclusion and those of belonging to the maxima of the histogram [Fig. 3a] are presented on the Fig. 4. Though there are overlaps in the homogenisation temperatures of the different inclusion types, but the maxima of the histogram roughly correspond to the types listed above.

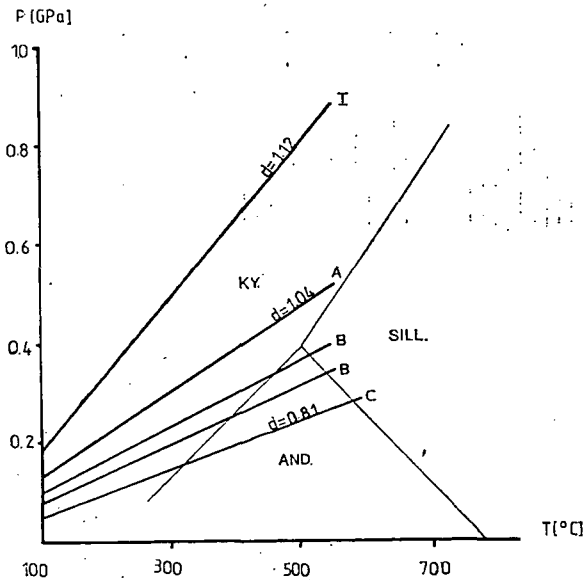


Fig. 4. Representative isochores of CO<sub>2</sub> inclusions. I = isochore of the most dense CO<sub>2</sub> inclusion, A, B, C = isochores, calculated from the adequate peaks of the distribution diagram [Fig. 3a]. ky. = kyanite, and. = andalusite, sill. = sillimanite. The Al<sub>2</sub>SiO<sub>5</sub> stability fields were drawn after HOLDAWAY [1971]

## H<sub>2</sub>O-rich inclusions

The H<sub>2</sub>O-rich inclusions are less common in the quartz segregations than the CO<sub>2</sub>-rich ones. The homogenisation temperatures [T<sub>h</sub>] of the A, B, C type aqueous inclusions range from +154.8 to +325.8 °C with three maxima [Fig. 3c]. These inclusions contain more or less dilute solutions with 1.7–14.3 wt.% NaCl equivalent salt content. There is no correlation between the salinity and the homogenisation temperature of the inclusions.

The maximum of the homogenisation temperatures of the aqueous inclusions in the matrix quartz gives a good coincidence with that of the A type H<sub>2</sub>O inclusions in the quartz segregations. The salinity of the matrix quartz H<sub>2</sub>O inclusions ranges from 3.2 to 4.6 NaCl equivalent wt% and the homogenisation temperature is between +155 °C and +188 °C [Fig. 3d]. The H<sub>2</sub>O inclusions found in the matrix quartz grains may represent a fluid originated from metamorphic reactions taking place soon after the peak P–T conditions of the first metamorphic stage. The relative scarcity of H<sub>2</sub>O inclusions, as compared with the quartz segregations, may be due to the H<sub>2</sub>O consuming mineral reactions, such as the formation of biotite and muscovite during different stages of metamorphism on one hand and to the possible destruction of inclusions by later recrystallisation on the other. The homogenisation temperature and salinity range of H<sub>2</sub>O inclusions in the matrix quartz is less broad than those of the type A aqueous inclusions in quartz segregations, which may refer to the fact that these fluids were closer to the equilibrium with the host rock than the aqueous fluids trapped in the quartz segregations [YARDLEY, 1983]. The lack of CO<sub>2</sub> inclusions in the matrix quartz may be due to the lack of CO<sub>2</sub> producing reaction during the metamorphism. The CO<sub>2</sub>-rich fluids were probably introduced along the quartz segregations.

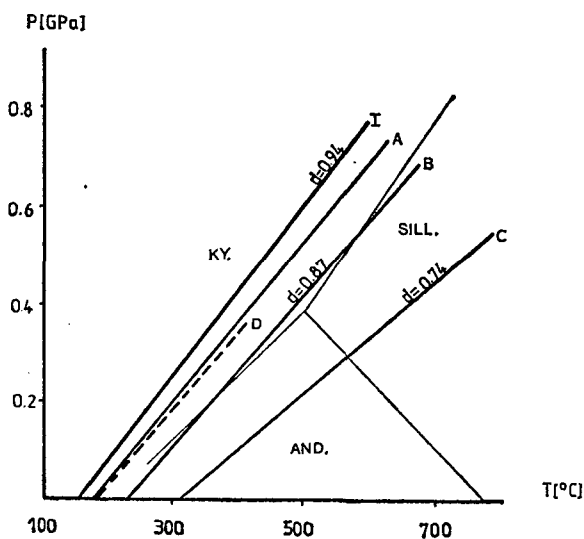


Fig. 5. Representative isochores of the aqueous inclusions. I = isochore of the most dense H<sub>2</sub>O inclusion, A, B, C, D — Isochores, calculated from the adequate peaks of the distribution diagram [Fig. 3c and 3e]. ky = kyanite, and. = andalusite, sill. = sillimanite. The Al<sub>2</sub>SiO<sub>5</sub> stability fields are after HOLDAWAY [1971]

Most of the  $T_h$  values of the D-type, late aqueous inclusions fell within the interval between 160 °C and 180 °C, but the whole measured  $T_h$  range is from 155.4 °C up to 217.2 °C [Fig. 3e]. The salinity of the D-type aqueous inclusions ranges from 1.9 to 4.2 wt% NaCl equivalent.

Calculation of the isochores was made by computer program of GATTER and MOLNÁR [unpubl.] based on the equation of state published by TALANTCHEV [1978]. Pressure results, exceeding 300 Mpa were obtained by extrapolation. The representative isochores of the most dense A-type aqueous inclusion [I] as well as those belonging to the three maxima of the histogram [Fig. 3c] and to the D-type  $H_2O$ -rich inclusions are presented on the Fig. 5.

#### $CH_4-N_2-CO_2$ inclusions

The  $CH_4-N_2-CO_2$  inclusions were observed along distinct healed mature fractures which always cross grain boundaries. All these inclusions belong to the C-type considering textural criteria. They were identified by critical homogenisation between -83 °C and -63.9 °C. The measured melting temperatures of the solid  $CO_2$  range from -75 °C up to -73.6 °C. The exact composition of a  $CH_4-N_2-CO_2$  inclusion was determined by Raman spectroscopy [Table 1]. The measured intermediate composition between these three fluids is quite rare. According to KERKHOF [1988], most of the metamorphic fluid inclusions in the  $CH_4-N_2-CO_2$  system can be approached as binary  $CO_2-N_2$ ,  $CO_2-CH_4$ , or  $CH_4-N_2$  systems, though TOMILENKO and CHUPIN [1983] also report mixed  $CH_4-N_2-CO_2$  inclusions with various proportions of the three fluids in high grade metamorphic rocks of the Aldan shield.

Besides the inclusion types listed above, some one phase inclusions could be observed that failed to show any phase change in the temperature interval of -180 °C and +350 °C. These inclusions may contain some kind of low pressure gas or metastable fluid [HOLLISTER *et al.*, 1981]. Decrepitated inclusions of all compositional types were observed as well.

#### DISCUSSION

If we want to evaluate the nature of the fluid existed at peak metamorphic conditions we have to consider the followings: (1) The isochore of the most dense  $CO_2$  inclusion marks higher pressure than that of the most dense aqueous inclusion, (2) In spite of thorough study of several samples, mixed  $CO_2-H_2O$  inclusions were not observed at all, (3) The earlier published miscibility-immiscibility relations in the system  $CO_2-H_2O-NaCl$  [see TAKENOUCI and KENNEDY, 1965; GEHRING *et al.*, 1979; HENDEL and HOLLISTER, 1981; SISSON *et al.*, 1981; BOWERS and HELGESON, 1983a, b; among others].

Considering the observations and available literature data, the existence of a pure or almost pure  $CO_2$  fluid is the most probable at peak P-T conditions of the first metamorphic stage. However,  $H_2O$  may be present in minor amount that cannot be observed optically in the  $CO_2$  inclusion up to 20 mol% [CRAWFORD and HOLLISTER, 1986]. The presence of more than 20 mol% water cannot be proved because of the lack of mixed  $CO_2-H_2O$  inclusions. The causes of separated  $CO_2$  and  $H_2O$  inclusions can be as follows:



- (1) Decrepitation of preexisting mixed  $\text{CO}_2\text{—H}_2\text{O}$  inclusions and redistribution of the fluid under P—T conditions where the mixed  $\text{CO}_2\text{—H}_2\text{O}$  fluid was no more miscible.
- (2) Sampling problems, due to the little amount of core sample.
- (3) The immiscibility may account for distinct  $\text{CO}_2$  and  $\text{H}_2\text{O}$  inclusions as well, but only under lower P—T regions of the metamorphism. The two fluids should have been miscible at higher metamorphic grades regarding the data published in the studies listed above.
- (4) Physical separation of the two fluids at higher P—T regions of metamorphism [CRAWFORD and HOLLISTER, 1986].
- (5) The recrystallisation of the host quartz due to the later stages of the metamorphism also may have contributed to the elimination of the mixed  $\text{CO}_2\text{—H}_2\text{O}$  inclusions.
- (6) The fluids were introduced separately along distinct fractures.

The peak P—T conditions of the first stage of the metamorphism, marked by the most dense  $\text{CO}_2$  isochore [see isochore 1 on the *Figure 4*] are in good agreement with those, determined by ÁRKAI [1984],  $P=890\text{ MPa}$ ,  $T=550\text{ }^\circ\text{C}$ , on the basis of plagioclase-garnet-biotite-muscovite geothermobarometer [GHENT and STOUT, 1981]. After the peak of the metamorphism, the fluid enriched in  $\text{H}_2\text{O}$ , but the two fluids trapped separately. The representative isochores of the A and B-type  $\text{CO}_2$  and  $\text{H}_2\text{O}$  inclusions [Fig. 4 and 5] show a continuous drop in the pressure and temperature. The aqueous inclusions in the matrix quartz formed during the early stage of the retrogression at the same time with the A-type aqueous inclusions.

There were three different fluids,  $\text{CO}_2$ ,  $\text{H}_2\text{O}$ , and  $\text{CH}_4\text{—N}_2\text{—CO}_2$  present during the second, low pressure metamorphic stage. On the basis of intersecting isochores

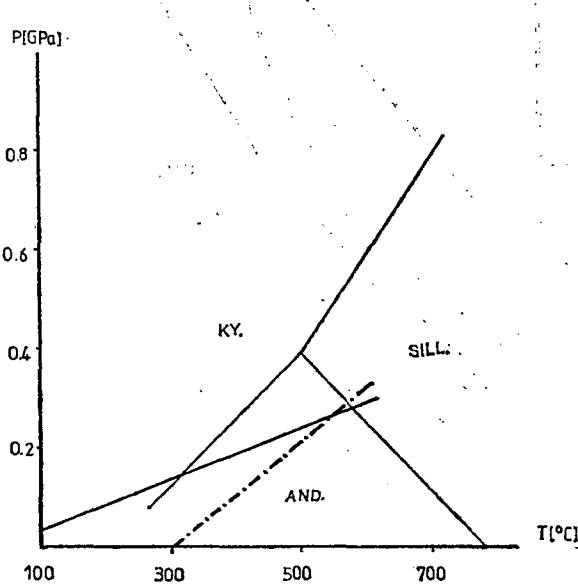


Fig. 6. Intersection of isochores of representative C type aqueous and  $\text{CO}_2$  inclusions, taken from Fig. 4 and 5.

— — — representative C type  $\text{H}_2\text{O}$  isochore,  
 — — — representative C type  $\text{CO}_2$  isochore

of two immiscible fluids trapped simultaneously, the P—T conditions of the second metamorphic stage can be determined. The intersecting isochores of the C-type H<sub>2</sub>O and CO<sub>2</sub> inclusions, calculated from the adequate maxima of the histograms [Fig. 3a and 3c], show 240—260 MPa pressure and approximately 540 °C temperature [Fig. 6]. The pressure and temperature inferred for the second metamorphic phase fall within the stability field of the andalusite [HOLDAWAY, 1971]. Though andalusite was not found in this sample, but it is present in several localities of the crystalline basement of the Somogy—Dráva Basin, giving an evidence on the existence of the second, low pressure metamorphic phase [LELKES—FELVÁRI and SASSI, 1981, ÁRKAI 1984].

The D-type aqueous inclusions, sealed along immature healed fractures, mark a late fluid influx after the second, low pressure metamorphic stage. They are probably due to the third, very low-low grade metamorphic phase. The representative isochore of the D-type aqueous inclusions [Fig. 5] show a similar temperature gradient as the A-type aqueous inclusions do. According to ÁRKAI [1984], the P—T conditions of this metamorphic event did not reach those of the biotite isograd, that means, the temperature remained below 450 °C [WINKLER, 1976]. If the temperature is considered to be about 300—400 °C, the pressure should have increased during this metamorphic stage.

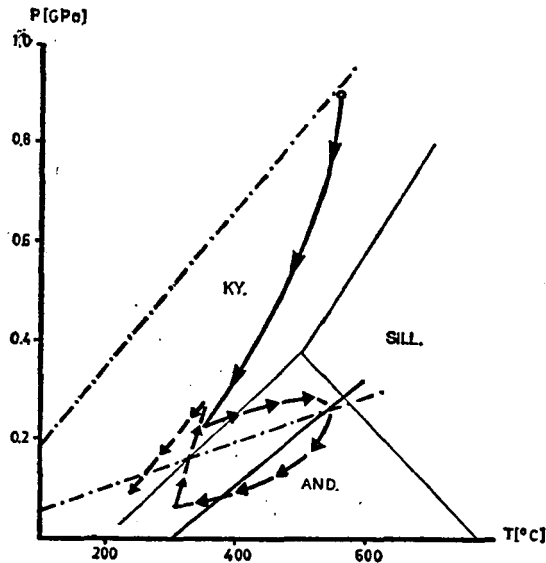


Fig. 7. A possible metamorphic P—T history of the Nagyatád K—1 gneiss on the basis of geothermobarometric data of ÁRKAI [1984, 1985] and fluid inclusion data.

- O — peak P—T conditions of the first metamorphic stage,
- = peak P—T conditions of the first metamorphic stage,
- = P—T path of the second stage,
- = P—T path of the third stage,
- · — = CO<sub>2</sub> isochores [isochore of the most dense CO<sub>2</sub> inclusion and the representative isochore of the C type ones are taken from the Fig. 4.],
- · — = representative isochore of the C type H<sub>2</sub>O inclusions taken from the Fig. 5.

## CONCLUSIONS

The peak metamorphic P—T conditions, marked by the densest CO<sub>2</sub> inclusion, are in good agreement with those, which are calculated from plagioclase-garnet-biotite-muscovite geothermobarometer [GHENT and STOUT 1981] by ÁRKAI [1984], ÁRKAI ET AL. [1985]. The fluid, existed at the peak of the first metamorphic stage was a CO<sub>2</sub>-rich one, perhaps with a minor amount of aqueous solution. After the peak P—T conditions CO<sub>2</sub>-rich and aqueous fluids existed together and formed separate inclusions during the whole period of metamorphism. The decreasing density of A and B-type fluid inclusions is due to the pressure and temperature drop during the retrogression. When the temperature decreased as low as 300—400 °C at approximately 200—300 MPa pressure, due to uplift, the temperature began to rise again, but the pressure did not change considerably. This increase in the temperature was probably due to a heat influx from below. The pressure and temperature of the second low pressure metamorphic event, inferred from the method of intersecting isochores of two immiscible fluids are about 240—260 Mpa and 540 °C. Besides the temperature increase, the fluid composition became more difficult than it was previously. Three different kinds of fluid existed during this metamorphic stage: a CO<sub>2</sub>-rich-, an aqueous- and a mixed CH<sub>4</sub>—N<sub>2</sub>—CO<sub>2</sub> one. After the peak of the second metamorphism the temperature dropped faster, than the pressure did. During the third phase, the thermal gradient decreased again, i.e. the pressure increased quicker than the temperature did. The fluid composition became simple, only a dilute aqueous solution was present during the third metamorphism. A possible uplift path, concluded from the obtained data, is presented on the *Figure 7*.

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