PETROGRAPHY AND MINERAL CHEMISTRY OF RHÖNITE IN OCELLI OF ALKALI BASALT FROM VILLÁNY MTS, SW HUNGARY

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ABSTRACT

In ocelli of alkali basalts from the Villány Mts (SW Hungary) rhönite has been found recently. This is among the first reports of rhönite from ocelli in basaltic dykes, as well as from the southern part of the Pannonian Basin. This new occurrence of rhönite has been analysed and described in this paper, in light of the hypotheses of formation of ocelli and crystallisation conditions of rhönite. On the basis of textural observations and mineral chemistry, we suggest that the ocelli was formed in the late stage of the crystallisation of magma, near the Earth's surface, at relatively high temperature but low pressure. Rhönite in ocelli crystallised as a primary phase in these conditions.
Key words: rhönite, ocelli, basalt, Villány Mts, Hungary.

INTRODUCTION

Rhönite is a relatively rare accessory constituent of magmatic rocks, because due to its minor grain size and semi-opaque character, its exact optical determination is difficult. Nevertheless, recently it is thought to be more frequent than it was described before. In the last decades it has been found in very different magmatic environments from being an accessory phase in groundmass of undersaturated volcanic rocks (e.g. Downes et al., 1995; Prestvik et al., 1999; Fodor and Hanan, 2000) to being a constituent in silicate melt inclusions and pockets of ultramafic mantle xenoliths (Kóthay and Szabó, 1999; Kóthay et al., 2001, 2003; Sharygin et al., 2003) and meteorites (Nazarov et al., 2000). To the best of our knowledge, there is only one occurrence known from ocelli (Sabatier, 1999), so, any new description may have a great importance that can contribute to specify the crystallisation conditions of both ocelli and rhönite.

The main aim of this paper is to present the textural and chemical circumstances of rhönite from ocelli. This also is the first report of rhönite from the southern part of Tisza Unit; namely from the Villány Mts alkali basalts. We also attempt to interpret the formation of ocelli and rhönite crystals in light of the previous descriptions and experimental studies of both,

giving in this way some contributions to the hypotheses about formation of ocelli, as well as to the crystallisation conditions of rhönite.

GEOLOGICAL BACKGROUND

In the Villány Mts, Mesozoic dykes of basaltic composition are present sporadically as small bodies (Fig. 1). Altered basalt dykes, containing high amount of ocelli crosscut Aptian – Albian limestones near Beremend and Máriagyűd. They are alkali basalts

containing several mafic and felsic xenocrysts, as well as ultramafic spinel lherzolite xenoliths of upper mantle origin (Nédli and M.Tóth, 1999a). Based on whole rock K-Ar data (Molnár and Szederkényi, 1996; Harangi and Árváné Sós, 1993) and the geochemical characteristics, these dykes are thought to have formed in the Late Cretaceous by intraplate magmatism (for details see Nédli, 1999; Nédli and M.Tóth, 1999a, b, 2003).
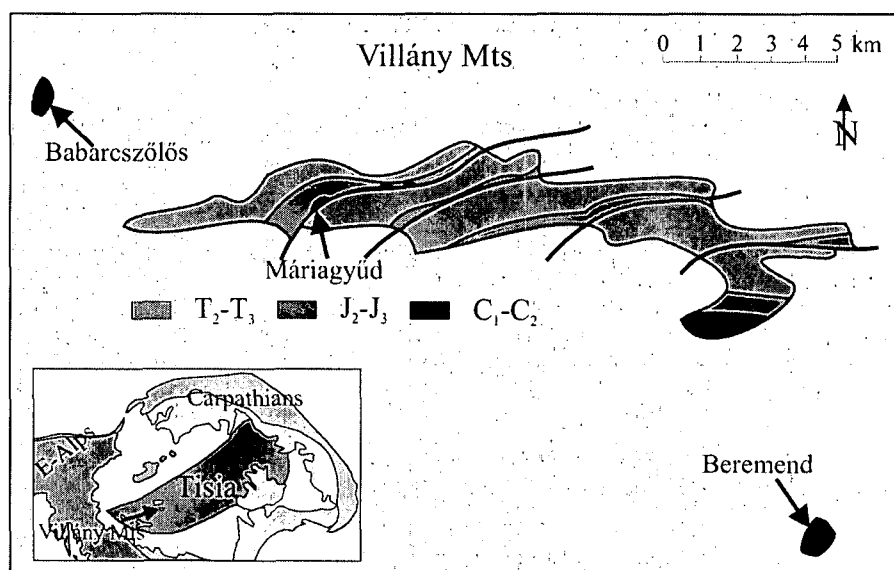
Fig. 1. Geological sketch map of the Villány Mts (after Fülöp, 1966). Surface outcrops of alkali mafic dykes at Máriagyűd and Beremend as well as the sill at Babarcszőlős are highlighted. Inset: Position of the Tisza Block in the Alp-Carpathian-Dinaric system (T_2-T_3: Diverse Triassic carbonate formations; J_2-J_3: Szársomlyó Limestone; C_1-C_2: Nagyharsány Limestone).

Petrography

The Villány Mts dyke group consists of porphyritic basalt with partially or entirely altered olivine and clinopyroxene phenocrysts, at places overgrown by amphibole rims, settled in a fine-grained, pyroxene-rich groundmass. Plagioclase is present only in the groundmass, where it occurs together with euhedral amphibole and biotite crystals, Fe-Ti-oxide grains and apatite needles. Secondary minerals (chlorite, clay minerals and calcite) compose variable amounts of groundmass, up to 20-30 vol%.

Felsic, globular ocelli frequently occur in the dykes. They are mostly ellipsoidal or spherical, rarely irregular in shape (Fig. 2A) of 1-5 mm in diameter. Most ocelli contain anhedral carbonate core surrounded by a complex rim of silicate minerals (Fig. 2B). The rim contains two types of microphenocrysts; the 0.3-0.5 mm long, greenish, euhedral amphibole prisms usually overgrow a clinopyroxene core (Fig. 2B, C). The rhönite grains are small (0.1-0.2 mm), semi-opaque or dark brown in colour (Fig. 2C, D). There is no textural reason for assuming any post-magmatic reaction between the amphibole and rhönite phases; they are regarded co-existing minerals. In addition to microphenocrysts, apatite needles occur in the weakly altered plagioclase-rich or glassy matrix. Plagioclase grains in the ocelli are significantly larger (0.5-1 mm) than those in the groundmass. Ocelli usually have a sharp contact with the groundmass; occasionally the groundmass pyroxene is aligned tangentially to the rim of the ocelli (Fig. 2A, B.). A few ocelli are composed only of carbonate minerals.

High amount of spinel lherzolite xenoliths of upper mantle origin and at some places resorbed quartz xenocrysts are present in the Beremend dyke (Nédli and M.Tóth, 1999a;2003). Significant textural variation can be observed in the hand specimens studied; those with high amounts of ultramafic xenoliths contain a few ocelli and felsic xenocrysts, whereas the highest amount of ocelli occurs mainly in the xenolith-free samples.

ANALYTICAL METHODS

Quantitative mineral chemical analyses were performed on representative rhönite, amphibole and pyroxene grains of ocelli on an upgraded *ARL-SEMQ 30* electron microprobe equipped with 4 wavelength-dispersive spectrometers (WDS) with TAP, LiF and PET diffraction crystals at
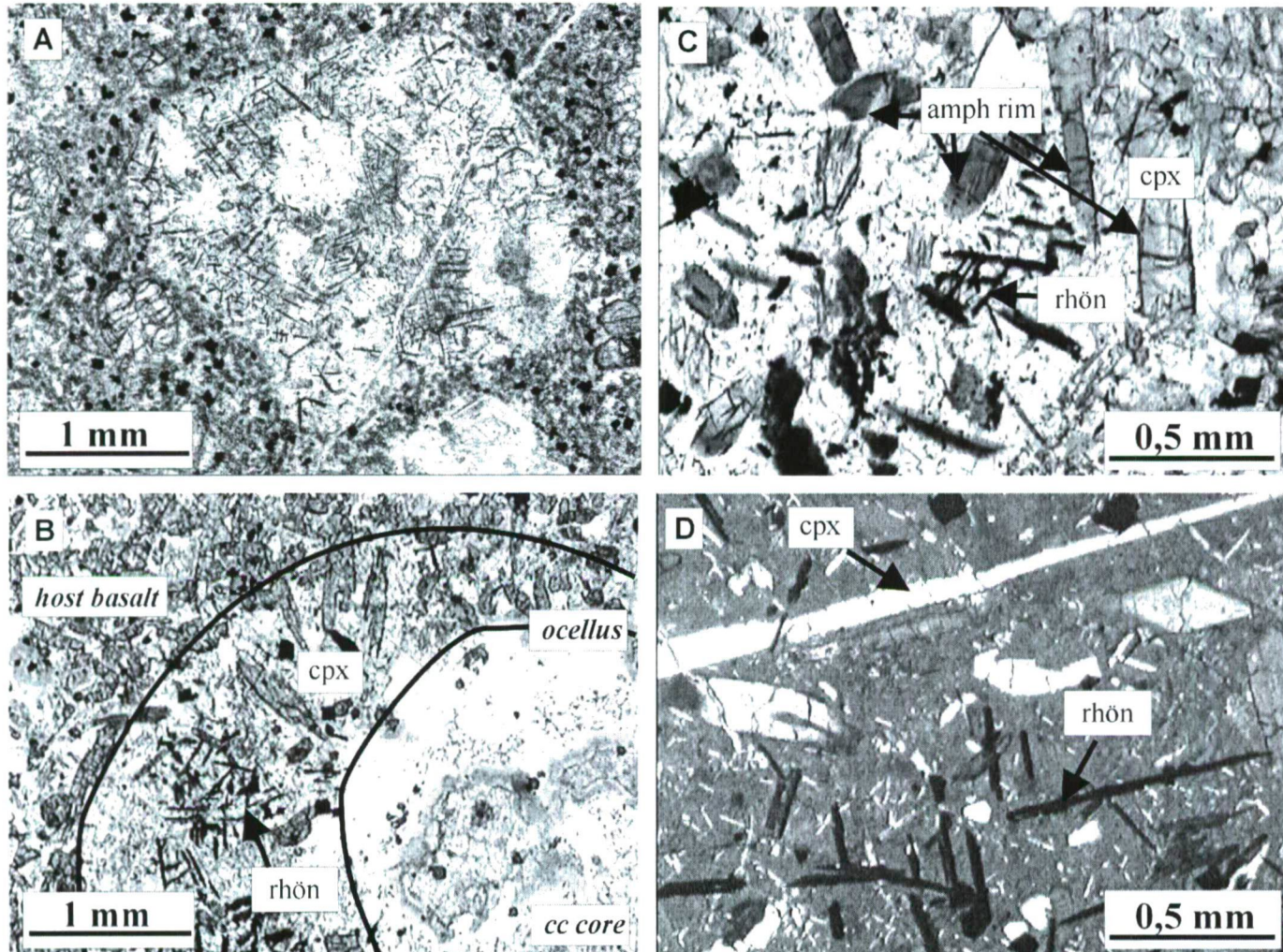
Fig. 2. Microphotographs of ocelli and rhönite in the Villány basalt samples. *(A)* ocellus with minor opaque rhönite sceletals (Máriagyűd basalt dyke, 1N). *(B)* typical occelus with carbonate core surrounded by a complex rim with clinopyroxene and rhönite microphenocrysts (Beremend basalt dyke, 1N). *(C)* opaque and semi-opaque rhönite crystals in ocellus with amphibole rimmed clinopyroxene and amphibole grains (Beremend basalt dyke, 1N). *(D)* clinopyroxene and rhönite microphenocrysts in ocellus (Beremend basalt dyke, +N).

Montanuniversität, Leoben (Austria) with 15 kV accelerating voltage, 20 nA beam current. For calibration synthetic and natural standards were used.

MINERAL CHEMISTRY

Compositions of selected pyroxene, amphibole and rhönite grains from the ocelli are listed in Table 1. Single pyroxene grains (cpx I.) have similar compositions to those rimmed by amphibole (cpx II.). Amphibole is maximal in Al^{IV}, and also is rather high in Ti and $(Na+K)^A$, while the most important pressure indicators Na^{M4} (Thieblemont et al., 1988) and Al^{VI} (Leake, 1965, 1971; Raase, 1974; Spear, 1981) are remarkably high too. The optically dark brown, small, elongated microphenocrysts are rhönite. Data suggest a high-silica variety with a clear change in compositions from the core to the rim; Si and Mg decrease rimwards, whereas Ti, Na and Fe tend to increase.

DISCUSSION

Origin of ocelli

Ocelli are characteristic textural elements of mafic rocks with high volatile contents. They principally occur in lamprophyres (Cooper, 1979; Foley, 1984; Rock, 1991). Their origin is still debated; in fact they have been interpreted either as amygdales, or nucleation cores of leucocratic minerals, or vesicles filled with late phase crystallisation minerals, or as products of immiscibility or pegmatitic immiscibility (Cooper, 1979; Foley, 1984; Philpotts, 1976; Sabatier, 1999).

Ocelli found in dykes in analysis are rounded with sharp boundaries. They contain coarser grains than the surrounding matrix; tangentially arranged grains around the ocellus and a zoned structure with spherical, carbonatic core are common. They are very similar to ocelli in lamprophyre dykes from the Alcsútdoboz-2 borehole (Szabó et al., 1993), whereas the ocelli described from the Mecsek Mts (SW Hungary) miss the silicate rim around the carbonate core and have different mineralogical composition (Demény and Harangi, 1996) being less similar to those in analysis. The ocelli of Villány Mts are also similar to the type II globular structures found in the sannaite dykes from Aillik Bay, Labrador (Foley, 1984) and to those

Table 1. Selected microprobe analysis of rhönite, pyroxene and amphibole in ocelli in Beremend basalt dyke.

| | Beremend basalt, ocellus | | | | | |
| | rhönite | | cpx-I/1 | cpx-I/2 | cpx-II. | tschermakite |
	core	rim			core	rim
SiO$_2$	28,09	27,74	45,54	44,35	41,97	39,59
TiO$_2$	6,66	7,30	2,55	2,62	4,13	3,40
Al$_2$O$_3$	14,33	13,85	9,03	9,30	11,79	15,17
Cr$_2$O$_3$	0,03	0,00	0,01	0,00	0,00	0,00
Fe$_2$O$_3$	0,00	0,00	0,00	0,00	0,00	0,00
FeO	25,67	26,25	9,97	9,87	9,30	15,43
MnO	0,28	0,21	0,29	0,20	0,13	0,40
MgO	12,07	10,87	9,23	9,26	9,06	9,17
CaO	10,35	10,44	20,73	21,20	21,68	10,26
Na$_2$O	1,34	1,69	0,91	0,91	0,70	2,41
K$_2$O	0,00	0,00	0,03	0,01	0,00	0,89
Total	98,82	98,35	98,29	97,72	98,76	96,72
Si	3,91	3,90	1,748	1,718	1,611	6,25
Al-IV	2,35	2,30	0,252	0,282	0,389	1,75
Al-VI			0,157	0,143	0,145	1,08
Ti	0,70	0,77	0,074	0,076	0,119	0,40
Cr	0,00	0,00	0,000	0,000	0,000	0,00
Mg	2,50	2,28	0,528	0,535	0,518	2,16
Fe+2	2,99	3,09	0,320	0,320	0,299	2,04
Mn	0,03	0,03	0,009	0,007	0,004	0,05
Ca	1,54	1,57	0,852	0,880	0,892	1,74
Na	0,36	0,46	0,068	0,068	0,052	0,74
Cation	14,40	14,40	4,01	4,03	4,03	16,20
O	20	20	6	6	6	24

Rhönite structural formulae based on 20 oxygen atoms (after Anthony et al., 1995). In the calculations Fe_2O_3/FeO ratio was set to ~ 0.5 following suggestions of Prestvik et al. (1999).

described by Philpotts (1976) in lamprophyres of the Monteregian alkaline province of Quebec, as well as others described from the Massif Central (Sabatier, 1999). The formation of such globular structures has been explained by segregation of a late-stage melt into bubbles after much of the groundmass had crystallised (Smith, 1967; Foley, 1984). The internal, carbonate core can be interpreted as a bubble that originally was occupied by a gas phase. The outer zones may have derived during solidification of a late-stage silicate melt. The origin of the core from a gas bubble is based on its common spherical shape (Smith, 1967; Foley, 1984), which is well shown by the ocelli from the dyke in analysis.

The high amount of ocelli in the studied subvolcanic bodies points to a high volatile content of the injected magma. The systematic appearance of the ocelli in the xenolith-free samples of the dyke provides evidence of

gravitational differentiation between the denser, xenolith-rich and the lighter, volatile-rich melt during crystallisation.

Chemical zoning of ocellus forming minerals is well described for some ocellar lamprophyres. In type II ocelli from Labrador (Foley, 1984), the common trends show a rimward increase of acmite component in clinopyroxenes coupled with a drop in Mg and Ca. In micas decrease of Mg, Al, Ti and rise in Ca and Mn is also observed (Foley, 1984). Zoned kaersutites of ocelli are progressively enriched form core to rim in Fe, Ca and Na and depleted in Mg in lamprophyres form New Zealand (Cooper, 1979). Clinopyroxenes from ocelli of lamprophyres in Hungary (AD-2) show normal zoning with rimward increase of Ti, Al and Fe (Szabó et al., 1993). These trends are in agreement with the interpretation of these ocelli as they represent late stage segregation melts (Foley, 1984).

Pyroxene crystals of the ocelli in question are identical to those appear in the host basalt suggesting an identical origin. Single amphibole grains and amphibole overgrowth (tshermakite, according to Leake et al., 1997) around pyroxene grains occur rarely outside the ocelli and so must have formed from the segregated volatile-rich material simultaneously with rhönite laths.

Based on the petrographic similarities between the Villány Mts ocelli and those from lampyrophyre dykes wordwide (Philpotts, 1976; Cooper, 1979; Foley, 1984; Szabó et al., 1993) we think that the common complex ocelli in the studied Villány Mts basalts derived by segregation of the volatile-rich segment of the magma in the late phase of the crystallisation and solidified after separation of carbonate-rich and silicate-rich parts into the carbonate-bearing bubble and the silicate mantle. Pyroxene grains rarely develop in the ocelli, rather they are inherited from the basalt matrix, based on their similar composition and on the fact that they crosscut the ocellus-host basalt boundary.

Crystallisation of rhönite

Rhönite is a rare aluminosilicate belonging to the aenigmatite group minerals. The general crystal-chemical formula of rhönite may be written as $A_2B_6T_6O_{20}$, where A = Ca, B = Mg, Fe^{2+}, Fe^{3+}, Ti, T = Si, Al. Rhönite has been described from different magmatic environments; mostly from groundmass of undersaturated alkali volcanics and intrusives e.g. melilite, nepheline basalt, basanite, alkali syenite (Kylie and Price, 1975; Magonthier and Velde, 1976; Johnston and Stout, 1985; Fodor and Hanan, 2000; etc.) In groundmass it occurs as small, opaque or semi-opaque, dark brown, euhedral grains, together with plagioclase, pyroxene and opaque minerals. Rhönite is known from other conditions as well; from meteorites as rounded grains included in fassaite crystals in melilite-fassaite paragenesis, considered as a metastabile phase in the melts (Nazarov et al., 2000; references therein) and from silicate melt inclusions of olivine phenocrysts as small, dark brown crystals together with augite, minor Al-spinel, pure CO_2 and sulfide blebs (Kóthay and Szabó, 1999; Kóthay et al., 2001, 2003). Secondary formation of rhönite from breakdown of Ti-bearing amphiboles is also reported (Kunzmann, 1999). Presence of rhönite in ocelli is rather infrequent worldwide; rhönite may afford important information about the crystallisation conditions of the ocelli, but its exact formation conditions are still lesser known.

There are only a few measurements available of rhönite worldwide (Kyle and Price, 1975; Magonthier and Velde, 1976; Johnston and Stout, 1985; Kunzmann, 1989, 1999; Downes et al., 1995; Prestvik et al., 1999; Fodor and Hanan, 2000; Grapes et al., 2003) but no one from ocelli. Comparing the rhönite from the Villány Mts basalt ocelli (Table 1, Fig. 3A, B) to those described from the more similar geological environment (groundmass), we can find some compositional differences. The rhönite in analysis is of higher Si, Fe and Na content while lower in Ti, Al and Ca than the samples described in several mafic magmatics (Downes et al., 1995; Prestvik et al., 1999; Fodor and Hanan, 2000). Fig. 3A shows the Ti vs. Na + ^{IV}Si plot of the Villány Mts rhönite and some published rhönite data from alkali mafic magmas, according to the nomenclature of Kunzmann (1999). The Villány Mts

samples fall within the rhönite range, but they are slightly higher in Na + ^{IV}Si than the other samples. On the (Na + ^{IV}Si) vs.(Ca + ^{IV}Al) diagram (Fig. 3B) the Villány Mts samples also are well separated from the other data due to their higher Na + ^{IV}Si and lower Ca + ^{IV}Al values; only one sample from Kyle and Price (1975) falls near to them. Comparison of terrestrial rhönite data (Grapes et al., 2003) shows that systematically high Na + ^{IV}Si and low Ca + ^{IV}Al is characteristic of rhönite formed as a breakdown product of kearsutite in mantle xenoliths or in gabbroic rocks, whereas rhönite phenocrysts and microphenocrysts or groundmass consituents normally have lower Na + ^{IV}Si, but higher Ca + ^{IV}Al. Such a chemical classification, however, depends essentially upon the bulk composition (Grapes et al., 2003), and so does not define clear genetic classes.

Several experiments have aimed to reveal the exact formation conditions of rhönite. According to conclusion of Kunzmann (1989), its formation under magmatic conditions requires pressure below 600 bars, temperature between 840 and 1200°C and presence of a hydrous fluid phase, however there is no limit of oxygen fugacity. Huckenholz et al. (1988)
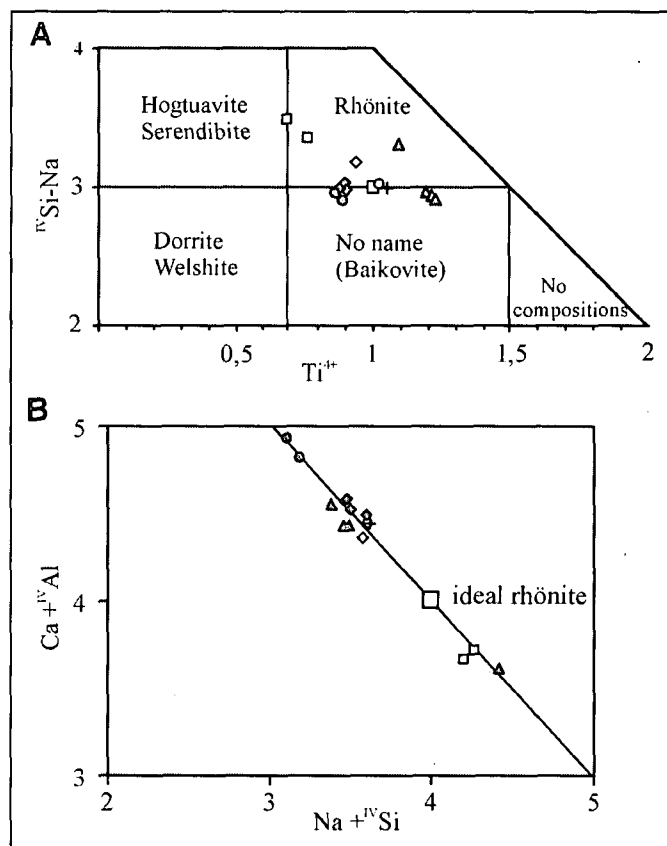

Fig. 3. (A) ^{IV}Si-Na vs. Ti^{4+} plot of rhönite from Villány Mts basalt in comparison with published data of rhönite as groundmass constituent (after Kunzmann, 1999). *(B)* Ca+^{IV}Al vs. Na+^{IV}Si plot of Villány Mts rhönite in comparison with some published data for rhönite as groundmass consituent. Black filled squares: Villány Mts; grey filled diamonds: Magonthier & Velde (1976); grey filled triangles: Kyle & Price (1975); grey filled circles: Prestvik et al. (1999); open circle: Downes et al. (1995); open diamond: Camerun et al. (1970); cross: Boivin (1980); open square: "ideal" $Ca_2(Mg_3Fe^{2+}Fe^{3+}Ti)(Si_3Al_3)O_{20}$ rhönite composition (Kunzmann, 1999).

determined the stability field for coexistence of rhönite and amphibole under magmatic conditions in an even narrower range of temperature and pressure: between 0.5 kbar, 1050°C and 0.2 kbar, 1000°C. They suggested that in ascending basalt, the formerly crystallised amphiboles leaving their stability field breakdown into an assemblage of rhönite + plagioclase at low pressure (below 0.5 kbar) and high temperature (1050-1140°C), and rhönite coexists with amphibole in the reaction assemblage only in the above mentioned very narrow PT range.

Experimental study of Boivin (1980) aimed to determine the chemical controls of the formation of rhönite. According to this experiment rhönite formation is independent of fO_2 conditions, but significantly depends on the chemical composition of the magma, especially on Ca and Ti activity of the liquid; TiO_2 content above 2.5 w% is required for the crystallisation of rhönite. If the Ca-content of the melt exceeds 11 mol%, rhönite coexists with plagioclase in groundmass, otherwise rhönite is the only crystallised phase. Rhönite normally occurs below 1 kbar, at about 1170-1100°C, accompanied by plagioclase.

Petrographic studies on rhönite in meteorites (Nazarov et al., 2000) and several recent microthermometric analyses on silicate melt inclusions (Kóthay and Szabó, 1999; Kóthay et al., 2001, 2003; Sharygin et al., 2003) have, however, proved that rhönite can form as a primary phase at rather high PT conditions.

CONCLUSIONS

The evolution of Villány Mts basalts, according to Nédli (1999), Nédli and M.Tóth (1999a, b, 2003) and unpublished data, can be outlined as follows. The magma was generated from a metasomatised spinel lherzolite mantle source, by a low degree (approx. 1-5%) partial melting. Ascending from the mantle it included fragments of the heterogeneous, metasomatised mantle, while at shallow depth the magma contaminated the crustal material, as well. Previous estimations (Kovács et al., 2001; Nédli and M. Tóth, 2003) showed that the ascending velocity of the magma was high enough (max.10-30 cm/s) to arrive near surface to form dykes. When intruding Mesozoic carbonates, the temperature could drop rapidly causing the separation of volatile-rich constituents from the crystallised basalt matrix. At the end of the crystallisation history the segregated melt solidified in ocelli.

Comparing the experimentally determined data and petrographical observations about the formation of rhönite to the evolution of the studied samples contradictory results may be got. Data suggest that the ocelli were formed in the late stage of the crystallisation of the basalt, certainly below 1000°C and 8 kbar, which are the crystallisation conditions of the clinopyroxene phenocrysts, according to method of Lindsley (1983). According to the method of Otten (1984), amphibole in ocelli records formation temperature around 980 °C as a minimum estimation through co-existing with another Ti-phase (rhönite). Taking also the fact into account that high ascent velocity of magma is supposed (Kovács et al., 2001; Nédli and M.Tóth, 2003), ocelli probably formed near the Earth's surface, at relatively low pressure but high temperature, like in the experiments of Kunzmann (1989) and Boivin (1980). Composition of co-existing amphiboles

of the ocelli confirms formation at high temperature through high Ti (Raase, 1974; Ernst and Liu, 1998), Al^{IV} (Bard, 1970; Blundy and Holland, 1990) and $(Na+K)^A$ values, significant Al^{VI} and especially Na^{M4} data, however exclude crystallization under extremely low pressures (Hammarstrom and Zen, 1986; Ernst and Liu, 1998). Several recent studies on rhönite in silicate melt inclusions (Kóthay and Szabó, 1999; Kóthay et al., 2001, 2003; Sharygin et al., 2003) also doubt crystallization of rhönite at very low pressures.

As a conclusion we can state that rhönite in the ocelli of the Villány basalt dykes are of primary igneous origin, which crystallized from the late, segregated volatile-rich part of the magma. Based on petrographic observations and composition of the co-existent amphibole, rhönite certainly developed under rather high temperature, but at significantly higher pressure than most experiments suggest.

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