Thesis of PhD dissertation

New results on the miocene volcanic activity of Bükk Foreland – physical volcanological and geochemical approach

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Introduction and goals

The Bükk Foreland volcanic area is the biggest and best exposed outcrop of the Miocene silica-rich pyroclastits which were deposited in the internal part of the Carpatho-Pannonian region. In some places the total thickness of the entire pyroclastic succession exceeds 750 meters in the Vatta-Maklár Trough (SZAKÁCS, A. et al. 1998). The age of the pyroclastic succession covers 8 Ma. The oldest pyroclastit is 21 Ma old and the Harsány Ignimbrite Unit, which is considered to be the youngest shows 15,66±0,66 and 13,35±1,01 Ma K/Ar and 14,0±0,3 Ma U/Pb ages (MÁRTON, E.–PÉCSKAY, Z. 1998; LUKÁCS, R. et al. 2007, 2015). Three main units can be distinguished within the succession based on physical volcanology, petrography, radiometric ages and paleolmagnetic declination, which can be related to the "lower", "middle" and "upper rhyolite tuff" (NOSZKY, J. 1926; SZAKÁCS, A. et al. 1998). The pyroclastit complexes, which can be readily discriminated via radiometric ages and paleomagnetic declinations (MÁRTON, E.–PÉCSKAY, Z. 1998) are supposed to contain layers from various volcanic centers (LUKÁCS, R. et al. 2007; LUKÁCS R. et al. 2015). The pyroclastit complexes comprise layers from various volcanic processes. Pyroclastic density current deposits, or more precisely thick ignimbrites are dominants, but some minor fall deposits from magmatic and phreatomagmatic eruptions and epiclastits are also present.

Thus, the Bükkalja is important in the investigation of tectonic and petrogenetic features of the Miocene, intrabasinal silica-rich pyroclastits. Moreover, these pyroclastits are good candidates for the entire examination of layers from caldera-related explosive volcanic eruptions.

In this thesis two aspects of the pyroclastic succession of the Bükkalja were investigated: 1) examination of structural hydroxyl content of quartz phenocryst fragments of ignimbrites (Topic I) and 2) investigation of physical volcanological features of phreatomagmatic fall deposits (Topic II).

Measurements on the structural hydroxyl content of quartz crystals from silica-rich lava flows, metamorphic bodies and sedimentary formations have been already performed (MÜLLER, A.–KOCH-MÜLLER, M. 2009; STALDER, R.–NEUSER, R.D. 2013; STALDER, R. 2014). Data on the structural hydroxyl content of quartz from granite + H_2O and from quartz + albite + H_2O runs are also available (STALDER, R.–KONZETT, J. 2012; FRIGO, C. et al. 2016). On the contrary, such measurements on quartz-bearing pyroclastits were not yet performed up to now. For such measurements the Bükkalja is an excellent candidate, because various quartz-bearing pyroclastits from different eruptions are presented in the succession. In Topic I

the main goals were the following: 1) to measure the characteristic structural hydroxyl content of >300 quartz crystals from four different ignimbrites and 2) to determine the most important factors that have major influence on it.

The presence of phreatomagmatic pyroclastits within the succession of the Bükkalja was known long ago (CAPACCIONI, B. et al. 1995; SZAKÁCS, A. et al. 1998). The hydromagmatic origin is believed to be evident from the fine-bedded structure and the accretionary lapilli content of the pyroclastits. However, the detailed volcanosedimentological and granulometrical investigation of these pyroclastits are still lacking. Thus, in Topic II the detailed field volcanological and granulometrical analysis of the pyroclastit succession near Bogács and Tibolddaróc villages were performed, in order to reconstruct the features of the volcanic processes.

Methods

- ***Topic I***

At Eger – Bolyki pincészet, Noszvaj, Tibolddaróc and Harsány samples were collected from the internal part of ignimbrites with >20 m in thickness. At Eger – Tufakőbánya and Bogács, samples were elaborated from different heights above the base (Eger: 0,05; 0,5; 1,2; 10,0 m a. b.; Bogács: 0,05; 0,5; 1,0; 6,5 m a. b.) of two different ignimbrites. Quartz phenocrysts were separated from pumice clasts and from the matrix too. Doubly polished "wafers" are 50-300 μm in thickness were elaborated. Single and transect unpolarized micro-FTIR (Fourier Transform Infrared) measurements were performed on the wafers. Measurements according to the unpolarized IR methodology of SAMBRIDGE, M. et al. (2008) and KOVÁCS, I.J. et al. (2008) were done on 325 randomly oriented quartz crystal. 20-35 crystals from each sample were measured. The homogeneity of intra- and intergranular AlOH structural hydroxyl defect contents were evaluated by comparing the measured and the theoretical distributions of AlOH structural hydroxyl defect contents. Theoretical distributions were computed according to the unpolarized indicatrix of AlOH hydroxyl defects. Trace element content of quartz phenocryst fragments were measured at the Mining and Geological Survey of Hungary by using LA-ICP-MS.

- ***Topic II***

First, volcanosedimentological features (thickness, color, bedding, sorting, componentry) of the pyroclastic succession which crops out near Bogács and Tibolddaróc villages

were documented on the field. Granulometrical analysis were performed on unit II, which is the best-preserved part of the succession. Granulometrical investigations were only possible on layers, which were not cemented and could be easily crushed by hand. At Bogács, 11 and 7 samples from A and B subunits and 4 from C subunit, thus 22 samples for granulometry in total were collected. At Tibolddaróc, 8 samples from subunit A, 1 from subunit B and 2 samples from subunit C, thus 11 sample in total were analyzed for granulometry. Granulometrical analyses were conducted in two steps. Fractions that are >63 micrometers (μm) in diameter were measured via dry sieving and the smaller ones by laser diffractometry at the Hungarian Academy of Sciences, RCAES, Institute of Geography. Grain size distributions were characterized by the median diameter (Mdϕ) and sorting (σϕ) values of INMAN, D.L. (1952) and by the F1/F2 ratio of WALKER, G.P.L. (1983). To determine the relative degree of shearing during deposition anisotropy of magnetic susceptibility were measured from 7 layers at the Paleomagnetic laboratory of Mining and Geological Survey of Hungary. These measurements can be used to decide whether a layer was deposited as fall or pyroclastic density current deposit.

Thesis

TOPIC I. – STRUCTURAL HYDROXYL CONTENT OF QUARTZ PHENOCRYSTS FROM IGNIMBRITES

- **Results on the structural hydroxyl content of ignimbrite-hosted quartz phenocrystals and on the factors that have major influence on it:**

- ***Thesis 1: Structural hydroxyl content of ignimbrite-hosted quartz phenocrysts is principally controlled by dehydration during slow cooling, however pre-eruptive zonation within the magma chamber, pre-depositional thermal history, incorporation of xenocrystals during pyroclast transportation as well as microcrack and inclusion content of crystals may also have influence on it.*** (chapter II.6.3.)
  - Structural hydroxyl content of ignimbrite-hosted quartz phenocrysts exhibit structural hydroxyl contents between 2.8±0.7 and 12.8±3.2 ppm and show systematic decrease with height above the base of the ignimbrite.
  - The lowering of structural hydroxyl concentration with height above the base is more gradual at Eger – Tufakőbánya (0.05 m:12.1±3.0; 0.5 m: 11.4±2.85; 1.2 m: 6.1±1.53; 10.0 m: 3.3±0.83 ppm) and more abrupt at Bogács (0.1 m:12.8±3.3; 0.5 m: 9.6±2.4; 1.0 m: 2.8±0.7; 10.0 m: 2.8±0.7 ppm).
  - Structural hydroxyl content of quartz phenocryst fragments from the internal part of thick ignimbrites (Eger – Bolyki pincészet, Noszvaj, Harsány sampling sites) is <3±0.8 ppm. At Tibolddaróc, 4.0±1.0 ppm structural hydroxyl content was obtained from a sample, which is elaborated ~6 m above the base of the ignimbrite.

- Thus, after hot deposition, the slow cooling induced diffusional dehydration is the main factor which cause lowering structural hydroxyl concentrations in quartz phenocrystals with height above the base of the ignimbrites.
- Dehydration of quartz crystals during cooling at a particular height above the base of the ignimbrite can be modelled, if information on heat conductivity of the ignimbrite, emplacement temperature, thickness, average diameter of quartz crystals and temperature dependence of diffusional coefficient of H in quartz is available.
- Systematic difference between measured and modelled structural hydroxyl concentrations of E-1 (0.05 m a. b.) and E-2 (1.2 m a. b.) level at Eger and BG-1 (0.05 m a. b.) and BG-2 (1.0 m a. b.) level at Bogács was observed in all model runs are based on various emplacement temperatures (Eger: T_0 = 300, 400, 500, 600°C; Bogács: T_0 = 400, 500, 600, 700°C) and thicknesses (Eger: z1 = 30, 40, 50 m; Bogács: z1 = 20, 30, 40 m).
- Thus, observed lowering of structural hydroxyl concentrations with height above the base of the ignimbrite is principally, but not exclusively controlled by different cooling rates and different average crystal sizes at different heights above the base. In the model runs uniform initial structural hydroxyl concentrations and effective diffusional speeds were used for each height above the base. In effect, these variables can be different between particular heights above the base due to P, T, X zonation within the magma chamber, different pre-depositional thermal histories, contrasting microcrack and silicate melt inclusion density of quartz crystals.
- Original magmatic, or only slightly lower structural hydroxyl concentration could only be preserved just above the lower contact of the ignimbrite, where cooling was instantaneous, thus prohibited effective dehydration.

- ***Thesis 2: By using the unpolarized IR methodology of SAMBRIDGE, M. et al. (2008) and KOVÁCS, I. et al. (2008) on n > 20 phenocryst fragments it is possible to recognize the homogeneity/inhomogeneity of structural hydroxyl concentration even in such an anisotropic mineral as quartz.*** (chapter II.6.2.1.; II.6.3.)
  - Distribution of unpolarized integrated absorbances are measured on randomly oriented quartz phenocryst fragments in samples from just above the lower contact of ignimbrites are not identical to the theoretical distribution from the unpolarized indicatrix of AlOH structural hydroxyl defects in quartz. On the contrary, measured and theoretical distribution of unpolarized integrated AlOH-related absorbances are identical in samples are several meters above the base (=from the internal part) of ignimbrites.
  - This difference at the base can be the consequence of the presence of incorporated xenocrystals with contrasting structural hydroxyl concentration, which were incorporated during eruption and PDC movement besides the cogenetic quartz population. Just above the base of the ignimbrite, original, or slightly lower structural hydroxyl concentrations were retained due to fast cooling. In the internal part of the ignimbrites slow cooling erased the initial intergranular heterogeneity of structural hydroxyl content.

➢ **Results on the unpolarized IR absorbance of quartz as a nominally anhydrous mineral:**

- ***Thesis 3: Effective sample thickness can be determined by using unpolarized FTIR measurements without independent measurement on sample thickness.*** (chapter II.4.4.1.)
  - Based on measurements on 306 randomly oriented quartz phenocryst fragment the following strong connection (R^2 = 0.86) is evident between integrated unpolarized absorbance between 400-2100 cm^{-1} (A_{SiO}) and the sample thickness:

$$A_{SiO} = 3.282^{\,x}\ Thickness$$

  - Thus, effective thickness of quartz grains or fragments can be determined quickly and in a cost-effective way by using unpolarized IR spectroscopy regardless of crystal shape and orientation.
- ***Thesis 4: The absorbance feature at 3200 cm^{-1} is related to water bonded on sample surface and not to overtone of Si-O frame vibration.*** (chapter II.6.1.)
  - On each unpolarized IR spectrum a weak absorbance feature with large FWHM is visible at 3200 cm^{-1}.
  - This absorbance feature was related to the overtone of the Si-O lattice vibration at 1680 cm^{-1} and not to structural hydroxyl by KATS, A. (1962). In contrary, BAYLY, J.G. et al. (1963) and YE, S. et al. (2000) interpreted this absorbance feature as a result of symmetric stretching vibration of O-H on the sample surface.
  - While, the integrated absorbance of AlOH structural hydroxyl defects at 3378 cm^{-1} show strong thickness dependence, the integrated absorbance of the peak at ~3200 cm^{-1} is independent from sample thickness.
  - The absorbance feature at 3200 cm^{-1} was also presented after heat treatment of the samples at 100 and 150°C. The integrated absorbance was not changed 15 and 30 minutes after opening the hermetically sealed sample holders.
  - Thus, it is plausible, that the absorbance feature at 3200 cm^{-1} is related to hydroxyls are bonded strongly to the crystal surface.

TOPIC II. – PHREATOMAGMATIC PYROCLASTITS WITHIN THE SUCCESSION ON THE BOGÁCS IGNIMBRITE UNIT

- ***Thesis 5: The pyroclastic successions near Bogács and Tibolddaróc can be correlated. After the deposition of Bogács Ignimbrite Unit explosive volcanic activity was going on in four main periods, which formed several tens of fall pyroclastits. Well-developed paleosols were formed in the lull of volcanic activity.*** (chapter III.4.3)
  - Near Bogács and Tibolddaróc a ~20 m thick layered pyroclastic succession cropping out, which was deposited on the Bogács Ignimbrite Unit. The two outcrops can be correlated. The entire succession is only visible at Tibolddaróc. The lowermost 3 m thick part of the formation is not visible at Bogács. The succession is subdivided into four main units (I-IV) by well-developed paleosols.
  - The presence of paleosol horizons imply that the succession was aggraded in three main periods. Between the periods were characterized by pyroclastic deposition, sufficient time for soil formation, thus at least several thousand years were elapsed. The

best-preserved part of the succession is unit II, which consists of several layers are 1-80 cm in thickness and can be subdivided into subunit A, B and C.

- The succession is dominated by fall layers, which is evident from the following features: general constant thickness and well-sorting of the layers, very weak magnetic fabric of subunit C in unit II and of unit III. Layers from pyroclastic density currents are subordinate.
- Emplacement from pyroclastic density currents is possible in subunit B of unit II. These layers are characterized by weak magnetic fabric and similar physical volcanological features than ignimbrites from dilute, turbulent, low energy, water-rich currents from the A Phase of the 26.500 y old Oruanui formation at New Zealand (WILSON, C.J.N. 2001).

- ***Thesis 6: The significance of water/magma interaction increases upwards in unit II. Initially pyroclast sedimentation from dry magmatic eruptions (subunit A), after that, from phreatomagmatic eruptions (subunit B and C) were taken place in unit II, which imply significant amount of water at the vent.*** (chapter III.4.1.; III.4.2.)
  - Unit II, which is the best-preserved part of the succession contains >30 layer which are 1-80 cm in thickness. Unit II can be subdivided into three subunits (A, B and C) based on the frequency of different lithofacies (LF).
  - Well-sorted ($\sigma\phi$ = 1-2) tuffs and coarse tuffs ($Md\phi$ < 1) which are 1-25 cm in thickness and contain >90% pumices and are characterized by constant thickness were deposited in subunit A. On the contrary, 10-80 cm thick, poorly sorted ($\sigma\phi$ >2) fine tuffs ($Md\phi$ > 3; F2 > 40%), which contain abundant accretionary lapilli are typical in subunit B and C, which deposited as phreatomagmatic fall deposits and from low energy, dilute, wet pyroclastic density currents.
  - Unit II comprises pyroclastits from dry magmatic Plinian eruptions (sensu lato) and highly fragmented pyroclastits from phreatomagmatic eruptions, thus it is supposed to be similar to the the Oruanui formation.
  - Unit II is supposed to provenance from a caldera, which was initially dry during the formation of subunit A and after that was filled with high amount of water due to caldera collapse which resulted high fragmentation in the case of subunit B and C.

Conclusions

Structural hydroxyl content of quartz phenocryst fragments is principally controlled by post-depositional slow cooling-induced diffusional dehydration at initially high T and low P. This finding highlights the potentional role of nominally anhydrous minerals in the investigation of thermal histories during eruption, pyroclastic transportation and time frame of volcanic processes. Although, for determining the original structural hydroxyl concentrations, that were equilibrated within the magma chamber one must investigate crystals from fall pyroclastits or from the contact zone of pyroclastic density current deposits and lava bodies, where cooling was fast enough, and only subtle dehydration was taken place.

Between the deposition of Harsány and Bogács Ignimbrite Units several tens of explosive volcanic eruption was taken place, which resulted mainly pyroclastic fall sedimentation at the Bükkalja. Regarding the number of eruptions, such explosive eruptions, that produced distal fall pyroclastits were much more significant, than the ignimbrite-forming eruptions in the volcanic history of the Bükkalja, which led to the aggradation of thick ignimbrites. Within the bedded succession, which crops out near Bogács and Tibolddaróc the influence of water on the eruptions ranging between extreme values. The succession contains pyroclastic fall deposits from dry magmatic eruptions and fall and pyroclastic density current deposits from phreatomagmatic eruptions. Thus, the succession is identical to the Oruanui formation at New Zealand. Such a pyroclastic formation has been hitherto unrecognized in the Carpatho-Pannonian region.

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