HYDROTHERMAL AND ORE-FORMING PROCESSES CONNECTED TO THE EARLY CRETACEOUS VOLCANISM OF THE EAST-MECSEK MTS.

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Summary of PhD dissertation

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INTRODUCTION, OBJECTIVE

The origin of some mineralizations and ore indications in the Eastern Mecsek was not clearly defined by previous studies. However, a common feature of these mineralizations is their spatial connection with the Early Cretaceous volcanic activity can always be identified. One driving force behind the research was that only limited information is available on the origin of mineralizations and ore indications in the Eastern Mecsek which is not confirmed by analyses carried out with modern instruments. The main objective of the thesis was a more detailed field identification (compared to previous studies) of the outcrops of hydrothermal processes induced by Early Cretaceous volcanism and the related ore indications, the detailed description of the outcrops and their testing with modern analytical methods. As a result of observations and analysis results, it was possible to reconstruct the geological environment of hydrothermal processes and the geological and biogeochemical processes leading to the formation of mineralizations and ore indications.

In the Early Cretaceous, the area of the Eastern Mecsek, belonging to the Tisza Unit, was located in the zone of continental rifting leading to the opening of the Alpine Tethys. Rifting and tectonic movements resulted in an approximately 3-6% partial melting of the asthenosphere at about 60-80 km depth which led to widespread alkali magmatism (Harangi, 1994; Harangi et al., 2003). Sztrókay (1952) was the first to bring up the possibility of submarine volcanism in relation to the examination of the Zengővárkony iron ore mineralization; later Viczián (1966) and Bilik (1974) also published about the presence of submarine basalt, and Viczián (1966) even mentioned that volcanic islands could also be formed which was later confirmed by Császár and Turnšek (1996). The geological environment was similar to that of the Triassic rifting of the Western Tethys, whose submarine volcanism is also well known in the Bükk-Darnó Units and also in the Dinaric-Hellenic belt (Palinkaš et al., 2008; Kiss et al., 2008; Kiss et al., 2012). However, the first Lower Cretaceous alkali mafic magmas did not flow onto the seafloor, due to the higher density of the basaltic magma compared to that of the wet sediment; instead the magma penetrated into the soft, unconsolidated sediments and formed intrusive pillow basalt beds and hyaloclastites. This was first assumed by McBirney (1963), later DSDP (Deep Sea Drilling Project) boreholes drilled in the Gulf of California (Guaymas Basin) in 1979 proved this hypothesis. Einsele (1980, 1982, 1985) already described hypabyssal “sill-sediment” complexes in his publications where hydrothermal activity is also characteristic within the thick sediment series above the sills as a result of pore water escape. Based on the heat flow measurements above sill-sediment complexes, pore water escape can be followed by a slower, longer period hydrothermal circulation within the sediment series (Lonsdale and Becker, 1985). Hydrothermal fluid mobilization can also be assumed by studying seismic sections, such events were described in the sediments of the Norwegian Sea (Svensen et al., 2003; Planke et al., 2005). Since the Eastern Mecsek was also part of a rift system characterized by the accumulation of thick marine sediments and volcanism in the Early Cretaceous (Bilik, 1980; Harangi&Árváné, 1993; Harangi, 1994; Harangi et al., 2003), an analogy can be drawn between the hydrothermal processes induced by igneous bodies intruding the sediments of the Guaymas Basin and the hydrothermal events that can be identified in the Eastern Mecsek. With this approach, we can find explanations supported by evidence for the formation of most hydrothermal processes and ore indications related to Lower Cretaceous basic igneous bodies. To investigate the formation of these mineralizations, it was indispensable to clarify the formation conditions of the host rocks, as well as the analysis of process that occurred at the magma/sediment contact. Since biogenic components can be identified in several ore types in rock-forming quantity (crinoid remains, decapod coprolites, bacterial ichnofossils), their role in the mineralization received a strong emphasis in my research.

MATERIALS AND METHODS

The field identification/uncovering of Lower Cretaceous igneous and volcano-sedimentary formations and various ore indications outcropping in the Eastern Mecsek area was followed by detailed field observations, section logging and sampling. The following instruments were used in the testing of materials: Polarization microscopy examinations were carried out with Olympus BX-51 polarization microscope (ELTE University, Faculty of Science, Department of Mineralogy) and Nikon E600WPOL polarization microscope (PTE University, Faculty of Science, Department of Geology and Meteorology). Fluid inclusion analyses were done on Olympus BX-51 polarization microscope equipped with Linkam FTIR 600 heating-freezing stage (ELTE University, Faculty of Science, Department of Mineralogy). Trace element analyses were done by Agilent Technologies 725 inductively coupled atomic emission spectroscopy instrument (ICP-AES), PerkinElmer ELAN® 9000 inductively coupled plasma mass spectrometer (ICP-MS) (ALS Laboratory, Vancouver, Canada) and Jobin Yvon

ULTIMA 2C inductively coupled atomic emission spectroscopy instrument (Geological Institute of Hungary, Geochemical Laboratory). X-ray diffraction analyses were done with Bruker D8 Advance instrument (University of Miskolc, Faculty of Earth Science and Engineering) and Rigaku MiniFlex 600 instrument (Szentágothai Research Centre, Analytical Chemistry and Geoanalytical Laboratory, Pécs). Scanning electron microscopy analyses were done with Hitachi S-4800 electron microprobe (with Bruker XFlash 4010 detector) (FEG) (Bay-Nano Institute, Miskolc) and JEOL JXA-8600 electron microprobe (University of Miskolc, Faculty of Earth Science and Engineering, Institute of Mineralogy and Geology). In some rock samples, the determination of organic carbon was done with Analytik Jena HT1300 instrument (NDIR detector) (Szentágothai Research Centre, Analytical Chemistry and Geoanalytical Laboratory, Pécs). Raman spectroscopy analyses were done with Thermo Scientific DXR Raman microscope equipped with 532 nm wavelength Nd-YAG laser source. (University of Szeged, Department of Mineralogy, Geochemistry and Petrology). Sulphur isotope analyses were done in the laboratory of SUERC (Scottish Universities Environmental Research Centre) in Glasgow, the analytical work was done by Peter Koděra. SO_2 analysis was done by VG SIRA II mass spectrometer. K/Ar datings were done in the Institute for Nuclear Research of the Hungarian Academy of Sciences (Debrecen), the analytical work was done by Zsolt Benkó.

THESES

1. In the area of the Mecsek, significant tectonic, lithological and palaeontological evidence show that rifting took place from the Late Triassic until the Late Cretaceous shortening phase, based on previous research by Vörös (1993, 2001), Haas (1990), Haas and Péró (2004), and Császár (2005). From the Late Triassic, up to 5000 m thick sedimentary series accumulated until the Late Jurassic as a result of continuous sedimentation (Bilik, 1980), the Upper Jurassic - Lower Cretaceous beds of this series were largely in unconsolidated state during the alkali mafic magmatism which started in the Early Cretaceous. Based on my research, the alkali basalt of the first eruption period did not flow onto the seafloor due to its higher density than water-rich sediments. It intruded into the unconsolidated marine sediments where intrusive pillow basalt, hyaloclastite and peperite formed in considerable thickness. This is supported by the 134.1±8.0 Ma K/Ar age of the approximately 180 m thick Zengővárkony sill composed of intrusive pillow basalt in its central part, surrounded by hyaloclastite. This is the same as the age of the oldest alkali basalt magmatites, measured with similar radiometric dating method (Márévár Valley, plagioclase basalt: 134.6 ± 5.1 Ma (Harangi&Árváné Sós, 1993)). This age also sets the age of hydrothermal events coeval with the volcanism.

2. Based on the section near Jánosipuszta, the intrusion of the alkali basalt occurred in at least 3 eruption periods. In the first period, intrusive pillow basalt with blocky peperite and hyaloclastite formed in more than 40 m thickness, followed by the deposition of reworked hyaloclastite and conglomerate, mainly composed of basalt pebbles. The next stage was a lava flow of pillow basalt in approximately 76 m thickness, and finally the deposition of conglomerate composed of basalt and limestone pebbles which was intruded by intrusive pillow basalt again.

3. In the case of the intrusive pillow basalt and the hyaloclastite, where permeability conditions prevented the escape of vapour formed during the magma/sediment contact, we can find the few μm size globular, tabular, drop-shape or cuspate fragments of vitrified basalt in the sediments in random distribution, as a result of explosions. In this case the wet sediment / magma ratio (R_s) could be between the values of 0.1 and 1.0 (Wohletz, 1983; Wohletz, 1995). Hydraulic breccia and blocky peperite formed on the magma contact where the host rock was more consolidated, while water-saturated sediments fluidized and fluidal breccia, or rarely fluidal peperite formed where the magma intruded loose, water-rich sediments and the heated pore water could migrate away. Such fluidal breccia can be found in the outcrop near the Somos stream in the Vár valley and the cover limestone of the Zengővárkony iron ore. During fluidization, pyrite-marcasite bearing hydrothermal channels with tubular structure formed from the pyrite bearing calcareous mud between the intrusive pillow basalts of Zengővárkony. The sulphur isotope composition of these iron sulphide structures preserved their nearly original ratios which indicate anoxic sedimentary conditions and intensive bacterial sulphate reduction occurring in the open pore space. It is characteristic in the contact surface of the magma with both unconsolidated and consolidated sediment that the solidifying magma absorbed volatiles from the host rock which led to the formation of amygdales with calcite infill.

4. We can always find *Mo* anomaly in the limestone inclusions (containing pyrite-bearing coprolites) embedded in the space between intrusive pillow basalts, as well as in the intrusive hyaloclastites containing pyrite-calcite infills (Zobákpuszta-Hidas valley: up to 159 ppm, Zengővárkony: up to 511 ppm), in some cases high *Re* content (Zengővárkony: up to 0.106 ppm); as well as *Ag* (up to 0.38 ppm) and *U* (up to 24.5 ppm) anomaly (Zengővárkony).

These anomalies are caused by the pyritification and subsequent recrystallization (due to hydrothermal effect) of locally anomalous organic matter content in Lower Cretaceous carbonate sediments, composed of large quantity of terrestrial plant remains and mainly the coprolites of benthic decapods. Evidence for the hydrothermal alteration of anoxic sediments include the textural features of coprolites, the residual bitumen from the natural distillation of hydrocarbons formed by hydrothermal processes from coprolite sediments, the very similar, in many cases identical distribution of *Ag, Cd, Mo, Re, Se, Ta, Tl, U* and *V* trace elements in limestone inclusions containing pyritic coprolites and in hydrothermal iron sulphide formations with tubular structure between intrusive pillow basalts. The same sulphur isotopic composition of non-recrystallized limestone inclusions built by pyritic coprolites and tubular structure iron sulphides, as well as the “atoll” structure of recrystallized iron sulphides are further evidence for the recrystallization of these anoxic sediments as a result of hydrothermal effect.

5. The homogenization of primary fluid inclusions in the calcite cement of brecciated fragments of iron sulphide channels with tubular structure located between the intrusive pillow basalts of the Zengővárkony volcano-sedimentary series, yielded 115 and 145 °C maximum temperatures. The salt concentration of the fluid trapped in the inclusions was found to be nearly the same as the average salt concentration of sea water which proves a low temperature hydrothermal circulation of sea water within the volcano-sedimentary series. Based on calcite and quartz veins penetrating the peperite inclusions, peperitization and the fluidization of sediments were followed by this hydrothermal circulation. In line with this, the primary inclusions (of only liquid phase) in quartz which was the last phase to precipitate, indicate the presence of very low temperature (<50 °C) solutions (Goldstein, 2001).

In the case of real submarine pillow lavas, not influenced by the insulation effect of the sediment cover (such as the ones found near Jánosipuszta in 76 m thickness), there is no sign of a hydrothermal event forming iron sulphide / iron hydroxide, iron oxide mineralization, or an ore indication.

6. The celadonite infill with smectite intercalation formed in the fissures of intrusive pillow basalt at Zengővárkony and Jánosipuszta is also an evidence for low temperature, slightly oxidative fluid flow.

7. The organic carbon detectable from the carbonate-pyrite infills of the intrusive hyaloclastite of the Hidas valley near Zobákpuszta, as well as the sulphur isotope composition ($\delta^{34}S$= between -34.02 and -34.47), and the trace element ratios also indicate the recrystallization of unconsolidated, Upper Jurassic - Lower Cretaceous pyrite-bearing sediments. The high *Mo* content (~159 ppm) of the pyrite-bearing hyaloclastite can be compared with the *Mo* content (~511 ppm) of sulphide precipitates formed at the recrystallization of pyrite-bearing sediments near Zengővárkony due to hydrothermal effect. If we assume the same sedimentary sulphur origin for both occurrences, this shows the similarity of hydrothermal processes at the two localities.

8. The sulphur isotope composition ($\delta^{34}S$= between -11.10 and -12.65) in the pyritic zone formed within the tephrite on the igneous contact zone of tephrite dykes in the Réka valley (Pécsvárad), intruding Toarcian anoxic sediments containing pyrite (formed as a result of bacterial sulphate reduction) showed good correlation (or only slight difference) with the sulphur isotope composition of pyrites in the Toarcian black shale ($\delta^{34}S$= between -16.72 and -18.01) which also confirms that the sulphur released from anoxic sedimens propagated towards the solidifying magma during magma/sediment contact processes. Based on texture properties, the sediment-derived sulphur formed pyrite by entering into reaction with the glass and mafic component of tephrite. A similar process occurred on the contact of pyrite-bearing carbonaceous sediments and alkali basalt dykes intruding coal seams.

9. However, sulphur isotope analyses also detected sulphur of igneous origin from Lower Cretaceous alkali basalt ($\delta^{34}S$= -3.94 and -4.31) (Magyaregregy, Vár valley). At the same time, the analysis of pyrites found in the calcite veins between pillow lava-breccia beds in the Vár valley indicate pyrite formation as a result of bacterial sulphate reduction where the very high $\delta^{34}S$ values of pyrite ($\delta^{34}S$= 42.50 and 44.38) indicate Rayleigh fractionation taking place in closed space (Seal, 2006).

10. According to the analyses, the transport and accumulation of terrestrial plant fragments, in places in large quantity, was characteristic in the Mecsek area already in the very beginning of the Cretaceous (*Márévár Limestone F.*). The appearance of plant fragments in carbonate sediments (*Márévár Limestone F.*) in the beginning of the Early Cretaceous was favourable for the spread of benthic decapods ("ghost shrimp"). Within the coprolite-bearing sediments, the increased amount of organic matter resulted in local anoxic conditions and iron sulphide accumulation in certain basin parts. In these areas I also detected *Ag, Cd, Mo, Re, U, V* anomalies. *Mo* showed further anomalies in the recrystallized sulphide phases as a result of hydrothermal activity induced by the intrusion of Lower Cretaceous basalt dykes into these anoxic sediments. This could be identified in several areas of the Mecsek. Ferrihydrite formed by the oxidation of pyrite-bearing decapod coprolites recrystallized to form goethite which forms the bulk of the Zengővárkony iron ore and the other iron mineralization re-discovered in the frame of my research in the Hidas valley. The ferrihydrite released during the hydrolysis of intrusive hyaloclastite also precipitated as goethite in the carbonate sediment inclusions, and, to a lesser extent, along the cracks in the hyaloclastite.

11. The formation of smaller anoxic sediment patches was mainly driven by the fauna, with the dominance of presumably opportunistic benthic decapods which consumed plant fragments. This could also contribute to the increase of beta diversity. The connection between plant fragments and the decapods is indicated that terrestrial, carbonized plant remains were always found near the coprolites identified in Lower Cretaceous formations. Apart from the six ichnofossil species described by Palik (1965) (*Favreina dispentochetarius, F. hexaochetarius, F. octoochetarius, Palaxius decaochetarius, P. tetraochetarius*, *P. triochetarius*), I identified another seven different ichnofossil species *F.belandoi, P.huaricolcanensis, P.osaensis, P.azulensis, P.groesseri, P.salataensis, H.siciliana*). Among these, seven different coprolite ichnofossil species are found in the ore of the Hidas valley alonge, while six in the Zengővárkony iron ore, and three in the older rocks (probably representing deeper marine environment) of the Zengővárkony volcano-sedimentary series. The appearance of coprolites in formations of different facies shows that most adaptive to various conditions, thus the most widespread species was the host animal of *P.decaochetarius,* the coprolites of which were found in almost all formations containing decapod coprolites. According to the Mecsek occurrences, the coprolites of species favouring greater water depths are *F.belandoi, H.siciliana and P.osaensis*. A decapod fauna of such high diversity within one formation is unique in the world, with regard to both the Zengővárkony and the Hidas valley localities.

12. Non-metamorphic, carbonate neptunian dykes occur in along the planes mainly parallel to foliation in the metamorphites of the Goldgrund valley near Ófalu. These have micro-laminated structure and one can identify hematitic microfossils of iron oxidizing bacteria in rock-forming quantity, along with also hematitic foraminifera remains in them. These dykes were injected into the opening fissures of the metamorphic seafloor. Based on their microlamination parallel to the strike of dykes, the fine grained material and the smaller veins branching off larger dykes, these can be regarded as injection neptunian dykes. The NW dip of larger neptunian dykes corresponds to the dip direction of the Mecsekalja tectonic zone which also indicates the opening direction of metamorphites with foliated structure. The research of Dabi et al. (2011) estimates that these neptunian dykes formed in the Lower Cretaceous based on the intersecting calcite vein generations.

13. Conditions that led to the spread of iron oxidizing bacteria (*Gallionellaceae*), found in rock-forming quantity in the hematitic neptunian dykes of the Goldgrund valley near Ófalu, were most probably established by the fact that a Lower Cretaceous igneous body intruded the hypoxic sediments and created the slow pore water circulation which led to the slow, bacterial oxidation of the originally pyritic sediments. This is because the optimal metabolism of *Gallionella* requires slow circulation which supplies reductive iron. This mostly happens near low temperature hydrothermal upwellings and springs but a slow convection also starts within loose sediments in the environment of sill intrusions which can transport a sufficient amount of dissolved iron and may also enable the minimal oxygen supply necessary for microorganisms. This model is supported by the fact that iron oxide saturated Jurassic sandstones penetrated by nontronitic veins, and heavily goethitic Lower Cretaceous hyaloclastites are found within 1-3 km of the occurrence of neptunian dykes. According to another explanation, iron oxidizing bacteria could also spread during the slow fluid flow formed in the fissures after the injection of sulphide-bearing sediments into the fissures of metamorphites but in this case it is also most likely that fluid flow was coeval with Early Cretaceous volcanism and tectonic activity.

14. The Fe^{2+} oxidized by Galionella bacteria first precipitated as ferrihydrite which has several hundreds of m^2/g active surface (Hiemstra and Riemsdijk, 2009), so the high *Cu* (371 ppm), *Mn* (8620 ppm), *Pb* (75.4 ppm), *W* (7.34 ppm) concentration of the hematitic limestone also assumes adsorption occurring on the surface of ferrihydrite. The metastable ferrihydrite later recrystallized to form goethite and hematite below μm size, but the hematitized, bent protrusion with characteristic morphology, protruding from the cell of the bacterium can still be recognized easily.

15. Other structures of bacterial origin can also be found in the examined Lower Cretaceous igneous formations and ore indications. These are goethitic, branching channels with 1-2 μm constant thickness, formed along the cracks of the glassy matrix of intrusive hyaloclastite which were formed as a result of microbial decomposition of basalt glass. The traces of microorganisms decomposing the glass are found along the permeable zones of the hyaloclastite in all cases which indicates that the slow infiltration of sea water was necessary to maintain their metabolism. In some cases, we can find a large network of goethitic microstructures along the cracks of the glassy matrix which indicates that microbes had a significant role in the hydrolysis of basalt and in the release of iron. This was the first case where such structures of bacterial origin could be identified in basalts in Hungary.

16. Biomimetic structures formed by abiogenic processes are also frequent in the examined formations. These are periodic chemical precipitates: DLA structures and “Liesegang” precipitates (“chemical stromatolites”) (Toramaru et al., 2003; McLoughlin et al., 2008) which formed during the oxidation of iron sulphides and as a result of hydrothermal alteration of the basalt glass. Their size range and thickness are highly variable, the phases of their formation can be followed well in the micro-textural images of the rocks.

17. As the Early Cretaceous magmatism went on, basalt volcanoes formed which penetrated the marine sediments and already rose above the sea level. Apart from the basalt conglomerate and colonial coral remains accumulated along volcanic slopes (Császár& Turnšek, 1996), further evidence for this phenomenon is provided by the large number of carbonized terrestrial gymnosperm fragments in the clastic formations. A volcanic morphology rising above the sea level is also suggested by the presence of solutions with lower salt content than sea water, measured in the fluid inclusions of the calcite vein fills of the Hidas valley marl, which may indicate the presence of meteoric water.

18. The trigger of the hydrothermal event that formed the Pusztakisfalu hematitic iron ore was most probably one of the Lower Cretaceous intrusions. In my view, an evidence for this is that the ZV.35 borehole, less than 1 km WSW from the iron mine, intersected Lower Cretaceous basalt below the Neogene conglomerate (Hetényi et al., 1968). Similar to the tephrite dykes that intruded the Toarcian black shale of the Réka valley, it can be assumed that a thicker sill intruded the Aalenian formation or the underlying Toarcian pyrite-bearing formation which triggered significant pore water flow and groundwater flow in the surrounding sedimentary formations, including the crinoid limestone. Silicic acid can form in larger quantity during the resorption of silica fossils which later precipitates again (Fournier and Rowe, 1966). As such, with regard to its origin, the silica matrix could have formed during the recrystallization of sedimentary components. Hydrothermal effects on the rock are also indicated by the concentrations of trace element anomalies fluctuating within very broad ranges. The intense fluid flow formed in the Pusztakisfalu iron ore formation was most probably caused by the fact that this formation was located in the Mecsekalja tectonic zone which was active in the Early Cretaceous. The crinoid iron ore can be easily identified based on its highly characteristic texture. As such, the iron ore pieces found next to the stone axes, red-painted pot fragments, obsidian and chert blades found in the Zengővárkony Neolithic settlement, based on their thin section images are clearly from the Pusztakisfalu mine. This also means that the use of the Pusztakisfalu hematite (as a pigment material) already started in the Neolithic.

19. The mineralized samples taken from the smaller adits constructed for gold and silver mining in the Lower Cretaceous igneous rocks of the Eastern Mecsek (Magyaregregy, Barnakő; Pécsvárad, Réka valley), did not show any precious metal anomalies based on the analyses.

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