OXYANDESITIC PETROFACIES IN THE WESTERN AND CENTRAL PARTS OF THE MÁTRA MOUNTAINS

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INTRODUCTIO N

In young volcanic territories, more or less altered rocks occur very frequently. During the mapping of such rocks one meets with some difficulties, on the one hand because of the insufficiency of available data, on the other band because of the great, variety of alterations, the right interpretation of the latter being problematical, too. These rocks have been referred to as "hydrothermally altered rocks", and chiefly observed in ore districts. Clearing up of genetic relationship between variosuly altered rocks and fresh rock" was lacking, too.

SZÁDECZKY-KARDOSS, examining the fundamental problems of volcanic mountains [11, 12], studied the genetical processes of the rocks hitherto considered as "hydrothermally" altered, and has established the system of altered rock's. Starting from the general regularities of hypomagmatites and metamagmatites, the formation, appearance, mineral composition of each type of altered rocks i's easily interpretable in the above-mentioned system.

As it is well-known, the temperature of crystallization of hypomagmatites is 1200 to 50° C, their volatile-content may be considerable. These rocks are characterized by the presence of primary low temperature minerals rich in volátiles (clay-mineralis, chlorite, etc.), and they are formed in a hygroscopic environment. In the case of metamagmatites, a partial substitution by volatiles takes place. Inside the magmatic mass, they chiefly appear along irregularly ramifying zones around tectonic lines. The great majority of epigenetically altered magmatic rocks belongs to this group: oxyvolcanites, hydrovolcanites, chlorovolcanites, etc. Oxyvolcanites may occur among hypomagmatites and metamagmatites, too.

FORMATION OF OXYANDESITES

After SZÁDECZKY-KARDOSS, the characteristic features of the different types of altered rocks have been described by PANTÓ [8]. As for the oxyvolcanites, PANTÓ has pointed out that they always represented rock-alterations taking place in oxidizing environment. In order to establish a uniform interpretation,

the products of both endometamagmatic and exometamagmatic processes are classed here. Great bodies of oxyandesites are not frequent. Metaoxyandesites of irregular shape and smaller mass are more wide-spread. Though the mapping of the latter — because of their small size — meets with difficulties, it is necessary to examine them from both volcanological and tectonical points of view.

It is difficult to give the liminal value of oxyvolcanites. The red colour of the rock is not always proportional to the O_{Fe} value. It is proved by serial examinations carried out on rocks of the Mátra Mountains, that it is impossible to unequivocally infer the degree of oxidation from the increase of O_{Fe} . Previously, PANTÓ, examining the andesites of Tokaj-Hegyalja [8], has pointed out that the reddish brown colour of the rock depended first of all on the respective modification and fine texture development of $Fe₂O₃$.

In igneous rocks $Fe^{2+} > Fe^{3+}$, in sediments it is the very reverse. The same holds true of oxyvolcanites. Considering the degree of oxidation, it is

1,4–1,6 $\left(\frac{2 \text{ Fe}_2 \text{ O}_3}{\text{ FeO}}\right)$ the same value being higher than 5,0 in the case of oxy-

volcanites. It would not be right, however, to characterize oxyandesites merely by the higher degree of oxidation, since this higher degree of oxidation does not always imply the distinct red or reddish brown colouration of the rock, whereas, in the field, oxyandesites are mostly recognized by colour.

To give a good example of the fact that the increase of the degree of oxidation does not necessarily imply the red colouration of the rock, we mention the rock of the andesite-quarry of the entrance of the Csevice valley at Tar; this rock belongs to a Lower Helvetian series. In its fresh state, this rock is dark grey, compact. At the fissures, the rock is partly altered by infiltrating waters: limonite occurs very frequently along the fissures, and it is forming incrustations or filling up thin fissures. A's pyritic, quartzy veins are to be found in the rock, too, not only exometamagmatic but also endomecamagmatic alteration should be present. Pseudoagglomerate formations, appearing here and there, show the same. Thus ferric iron does not precipitate, here, in the form of haematite, but in that of limonite, which generally does not take part in the colouration of the whole rock. Colouration produced by limonite is limited to the fissures.

It is well-known that, under subaerial conditions, when p_H is 4 to 8, iron precipitates in the form of $Fe(OH)_{3}$, wherefrom limonitic minerals are formed. Therefore, in this case, the colouration of the rock can undergo the effect of limonite minerals formed in the course of diagenesis. Upon the effect of hydrothermal solutions or higher temperature, ferrous iron can turn into ferric iron, at similar p_H in case of medium oxidation-reduction potential, therefore it is very difficult to distinguish oxyvolcanites produced by hydrothermal processes from those produced by weathering [11].

At this occurrence, the following data have been given by the analyses. $1 =$ weight percents of the fresh rock, $2 =$ weight percents of the altered rock. Analyst: L. JANKOVICH)

There is a considerable difference between the respective degrees of oxidation. In the fresh rock, the value of O_{Fe} is as low as 0,05 which indicates thatconsolidation did not take place on the surface, but near the surface where conditions of oxidation did not exist. Consolidation taking place near the surface is also indicated by the amount of the groundmass which is much smaller than in the neighbouring andesites, while its degree of crystallization is higher.

The difference between the values of Al_2O_3 is conspicuous pointing to the fact that weathering took place at higher p_H , thus it might have been at least partly eliminated in solution. The appearance of calcite indicates higher value of p_H too.

Examining the possibilities of oxyvolcanite formation, SZADECZKY-KAR-DOSS [13] has come to the conclusion that, in case of lower oxidation-reduction potential, at p_H 6 to 8, orthovolcanites are stable, while opacitization appears at increasing values of oxidation-reduction potential $(+0,2)$ to O,3 V). In the latter case, the degree of oxidation will be considerably higher, moreover, in the case of oxyvolcanites the value of oxidation-reduction potential is as high as $+1,0$ V. In case of higher water-content, in similar conditions as for the rest, it is leucovolcanite which will be stable. The formation of oxyvolcanites is considerably affected by p_H , since in case of higher p_H it is montmorillonitization, at lower p_H it is kaolinization which can be observed. In these cases, of course, the volatile-content should be higher, too.

After all, in the field work we can subscribe to PANT6'S opinion, according to whom *,,oxyvolcanite"* is a collective term for rocks, the alteration of which characteristically produces $Fe₂O₃$ minerals. This phenomenon becomes visible in the colour of the rook: various nuances of red are observable, even macroscopically. As a matter of course, the degree of oxidation of these rocks will be higher, often as high as 8 or 10.

FORMS OF $Fe₂O₃$ AND $Fe₃O₄$

Now, one can raise the question in what form $Fe₂O₃$ is present. Many varieties have been described. Two forms certainly exist, namely α -Fe₂O₃ i. e.

57

haematite and γ-Fe₂O₃ *i. e. maghemite.* Both forms may be considered as polymorphic modifications of the $Fe₂O₃$ compound. According to the examination it is quite probable that modification α is stable, while γ is the metastable one. MASON [6] refers to several authors according to whom modification α turns to modification γ in the interval of 200 to 700 (800)° C. This interval depends on the conditions of formation of the mineral in question, on the impurities, etc. Modification γ turns into modification α by increase in temperature. As, at ordinary temperature, the turning of modification γ to modification α is slow, the former one occurs in nature, too. Natural γ -Fe₂O₃, *i. e.* maghemite is probably produced in the oxidation zone.

Martite also shows the $Fe₂O₃$ composition, but this is pseudomorphous after magnetite. Martitization always .follows crystallographical directions; mostly along octahedral faces, whereas conversion to maghemite is independent from any crystallographic form.

GRUNER [3] has pointed out that magnetite was very slowly oxidizing into haematite, in the open air, at 150 to 200° C. Oxidation starts along the octahedral faces, as well as it has been observed in the case of martitization. In accordance with the above-mentioned phenomenon, natural turning of magnetite into haematite ta'kes place near the surface of the Earth, exclusively in the oxidation zone. This process is a sort of weathering. According to SHEPERD $[9]$ the volatiles of the lavas and magmatic rocks always contain reducing agents (H₂, CO, SO₂, etc.), thus the effect of magmatic fluidal solutions on $Fe₂O₃$ may be expressed by following equations:

$$
3 Fe2O3 + CO = 2 Fe3O4 + CO2
$$

$$
3 Fe2O3 + H2 = 2 Fe3O4 + H2O
$$

It has been proved by experimental evidence th'at, in this reactions, equilibrium shifted to the right to such a degree that these processes might be practically considered as irreversible. The equations also show that one weight fraction of CO can reduce seventeen times as much haematite to magnetite, while H_2 reduces 240 times as much. This is why primary haematite lacking or playing a minor part in magmatic rocks, except for granite.

 $SO₂$ and less oxidized sulfur compounds reduce ferric iron at a relatively low temperature, this effect is reflected by the presence of magnetite in some sulfide deposits. It can be stated, therefore, that haematite turns into magnetite on the effect of reducing agents, at least a considerable part of the transformation haematite/magnetite is due to the reducing effect of C, H and S-compounds, the source of which can be a magmatic one.

The alteration of magnetite into haematite is a general and wide-spread phenomenon. It follows from the foregoing that magnetite cannot turn into haematite but in case of absence of reducing agents or in conditions favourable to oxidation.

Magnetite can turn into haematite, as a result of descendent processes. The p_H of the medium in which the reaction takes place, plays an important part in the oxidation of ferrous iron into ferric iron. Oxidation takes place much more easily in an alkaline medium than in a neutral or acidic one. It is quite probable that alkaline solutions, even in the absence of atmospheric oxygen, have an oxidizing effect on magnetite [11]. Chemical factors in themselves, however, cannot completely determine the degree of replacement. Physical properties of tihe rock, porosity, grain-size, direction of the fissures are factors promoting the change or retarding it. The change of haematite into magnetite implies decrease in volume, while the turning of magnetite into haematite is accompanied by increase in volume. This is the result of the difference between specific gravities calculated from the accepted unit-cell dimensions of magnetite and haematite. In the case of magnetite and haematite, there is an increase of 2,5 percent in the respective volumes.

According to SCHMIDT and VERMAAS [10], there are two, distinctly exothermal peaks on the d. t. a. curve of magnetite. The first one appears at 360 to 375° C, the second one at 580° C. The value of the first peak is slightly modified by varying grain-size. The authors have come to the conclusion that magnetite, heated in air, undergoes two stages of oxidation. The first one, is a surface oxidation into haematite, the other one being a complete oxidation into haematite. They have also pointed out that in the course of the heat treatment, no trace of γ -Fe₂O₃-formation was observable. According to LEPP [5] superficial oxidation $-$ considered as first stage $-$ seems to be in connection with specific surface. Examining synthetic magnetites, GEITH [2] has interpreted the result of d. t. a. investigations as follows: the first exothermal peak should represent the initial oxidation into γ -Fe₂O₃, and this is followed by the complete alteration from modification γ into modification α . According to LEPP, too, the first exothermal peak should represent the oxidation into $Fe₃O₃$.

In order to investigate the nature of the first exothermal peak, LEPP pulverized natural magnetite to the grain-size of 0,061 mm (—250 mesh), a'nd heated it to 430, then to 500° C. Both temperatures are above that of the first exothermal peak and below that of the second one. The X-ray diagram of this product has given a very 'strong magnetite pattern. Very few of the even strongest haematite-lines were to be seen. This phenomen probably indicates that only a very small amount of magnetite has oxidized. The examination of the ferrous iron-content, however, indicates a quite different situation. While the composition of the original substance approximated to the theoretical composition of magnetite, after having been heated to 430° C the Fe₂O₃/Fe₃O₄ ratio was 34 : 66 in it, in the substance heated up to 500^o C the ratio was 45 : 55. This would indicate, on the other hand, that the few and weak haematite lines of the X-ray diagrams of the samples that underwent heat treatment, would not represent complete oxidation, and the oxidation taking place below 500° would represent a change into γ -Fe₂O₃. The broadening of the above mentioned lines on the X-ray diagrams seems to be due to the fact that the dimensions of the unit-cell of γ -Fe₂O₃ differ but slightly from those of magnetite.

As a result of his d. t. a. investigations, LEPP infers the following phases in the course of the rapid oxidation of magnetite:

4 Fe₃O₄ + O₂
$$
\longrightarrow
$$
 6 γ -Fe₂O₃

The reaction starts at about 200° C and its maximum is at 350 or 400° C.

$$
\gamma\text{-}Fe_2O_3\text{-}\text{-}\text{-}\text{-} \text{-} \alpha\text{-}Fe_2O_3
$$

The reaction starts at about 375° C and ends at 525 to 550° C.

4 Fe₃O₄+O₂
$$
\longrightarrow
$$
 66 \times Fe₂O₃

the reaction starts at 550 to 570° C.

After all, the rate of magnetite oxidation, at given pressure and temperature may be considered as a function of the specific surface of the matter and of the order in the crystal lattice.

DESCRIPTION OF THE OCCURRENCES

In the western and central parts of the Matra, oxyandesitic petrofacies is frequently observable along andesite dikes. Though these occurrences are not very thick, they represent, however, a characteristic part of the massive andesite [4, 7].

Between Tar and Hasznos on one hand, as well as Kékes and Határpataktető on the other hand, andesitic lava flowed sometimes on andesite tuff or on lava-bond agglomerate. In these places, the surface of the lava-bond agglomerate and andesite-tuff was somewhat more mature, its water-content was higher than that of the lava. In ordinary facies, the colour of the lava is either medium grey or yellowish grey (pumiceous). If the lava flows on such a surface, a volatile mass of higher pressure appears below the lava, because of the higher water-content of the floor. It is one of the effects of the abovementioned phenomenon that the lower part of the lava-flow becomes scoriaceous. *Scil.* after the escape of volatiles, the lava becomes viscous to such a degree, that the porosity produced by volatiles remains.

In the valley of the Csevice-brook, near the Tar we could observe an occurrence of this kind. There, the grey, yellovish grey andesite-tuff crops out with a 45—225° strike and 23° SE dip, near point 279. The succession of . strata is as follows:. Down below, one finds grey, fine-grained, andesite tuff of sandy appearance. This is overlain by a 60 cm thick stratum of yellowish grey andesite tuff with lapillis, which occurs very frequently in this territory, then about 40 cm thick, red oxyandesite tuff. The latter is in contact with the andesitic lava, the lower 20 to 40 cm thick part of which consists of red oxyandesite, overlain by the normal, medium grey, more compact andesite.

The chemical composition of the normal, yellowish grey andesite tuff (1) and that of the average oxyandesite tuff (2) are as follows (analyst: M. EMSZT):

If one compares the data of the two analyses, one finds no considerable difference but in the change in $Fe₂O₃ - FeO$ -content and in $-H₂O$ -content.

The yellowish grey andesite tuff has been formed in an oxidizing environment where FeO-content had been lower. In the oxyandesite tuff, however, the whole FeO oxidized into $Fe₂O₃$, in conformity with the new conditions of equilibrium produced by the heat effect of the lava, consequently a considerable change took place in the ferrous/ferric ratio. The decrease in $-H₂O$ -content is evident, too, since $-$ on the effect of heat $-$ a part of the water, namely adsorbed, unstably combined water, easily passes into the vapour phase producing the scorification of the overlying lava. The andesite-tuff zone turning into oxyandesite tuff is thin in this case, because the overlying lava flow was very thin, too.

The thin lava flow, lying over the andesite tuff, has been broken up by erosion *(Fig. 1).* The porosity of the rock has secondarily developed over the scoriaceous oxyandesite. Cavities are filled up with chlorite, chalcedony, and yellowish, rhombohedral calcite.

l'ig. 1. **Andesite-lava broken up by erosion; in the lower part oxyandesite. Tar, Csevicevalley.**

The chemical composition of both rock types is following (Grey, porous andesite (1), oxyandesite (2), analyst: L. JANKOVICH):

If one compares the chemical analyses of both rocks, considerable differences are to be found, as a matter of fact, only in the $Fe₂O₃ - FeO-values$. While the value of O_{Fe} is approximately 3 in the case of the lava flow — indicating the subaerial congealation of the latter $-$ that of the oxyandesite is 11,3. This difference becomes visible in the colour of the rocks.

A similar phenomenon was observable in the Central Mátra, too. Along the northern ski-track of the Kékes, red oxyandesite appears in a clastic form at the higher levels and in massive occurrences at the lower levels. The latter can be considered as a thinner, intercalated lava flow under the younger andesite of the Kékes. As this formation is covered in a large measure, it is very difficult to determine more exactly its position. The rock *in situ* rises to 5 or 6 m over the surroundings, at the western side of the ski-track.

Under the microscope, particularly in case of considerable magnification, the rock is red-translucent. The groundmass is completely covered with haematite grains of the order of magnitude of some microns, consequently the microlithe of the rock-forming minerals are not discernible. Porphyritic felspars are well distinguishable and contain, among others, haematite inclusions, too. Around the inclusions, one can sometimes observe ochreous colouration which is already an effect of colouring by limonite. The clay-mineralization of the felspars is not considerable. Pyroxenes are often surrounded by an opacitic border, which indicates a higher degree of oxidation, too. Ore inclusions frequently occur in the latter.

To the North of the Határpatak-tető, one finds likewise oxyandesite on the surface, overlain by grey andesite *in situ.* The situation is the same as in the Csevice valley, *i. e.* the andesite tuff is overlain by a thin lava flow, the very thin lower part of which altered into oxyandesite.

On the western side of the Kozbérc, at 500 to 550 m above sea-level, on an almost N—S ridge consisting of grey andesite, the upper part of the andesite tuff — under the massive rock — changed into oxyandesite tuff, the clastics of which can be followed at great length, on the western slope of the mountain. Thus one finds once more the interrelation between andesite tuff and lava.

At the middle course of the Csonka brook one also finds oxyandesite on the agglomeratic andesite tuff. It is well observable, on this occurrence, that coloured minerals are surrounded with ore border, *i. e.* there appears opacitization.

On the southern slope of the mount Várhegy at Hasznos, there is an intercalation of banked, lamellar andesite, in the lava-bond agglomerate. The lower part of the andesite is not opened up, thus we do not know the conditions in this part, while the banked, grey, augitic hypersthene-andesite gradually passes into oxyandesite *(Fig. 2).* This zone can be followed along the exposure. From the appearance of the oxyandesite one can infer that the alteration into oxyandesite of the upper part of the already congealed rock was facilitated By the still hot, lava-bond agglomerate which flowed on the andesite. The narrow zone is a necessary consequence of the relatively small amount of lava-

hig. 2. **Banked andesite-lava overlying by oxyandesitic and hydroandesitic varieties. Hasznos, Várhegy.**

bond agglomerate, its effect is observable only on a narrow stripe. The increase in haematite-content of the rock is limited to the same stripe, too. Volatiles probably did not play any important part, here, in the basement of the massive rock. This is probably why scorification, appearing elsewhere, is not observable, here. The results of the chemical analyses of fresh, grey andesite (1) and red •oxyandesite (2) are following (analyst: M. BARANYI):

Thus, while the first analysis shows the composition of the characteristic andesitic lava of the environs, where the degree of oxidation is low [3, 15], too, there is no considerable difference, in the case of oxyandesite, but in the ferrous/ferric ratio, in consequence of which there is an increase in the degree •of oxidation (6,56).

The oxyandesite tuff appearing on the ridge running to the NE of the watch-house of Zsillő, at Hasznos, is also interesting. There, the red andesite tuff with lapillis or sandstone inclusions is to be found in the upper part of the .Helvetian andesite tuff appearing on the schlieric formation.' Since sandstone inclusions in the tuff show a weak contact effect $[3]$, one can infer that there was a jet of high-temperature pyroclastics which, having a loose structure on the surface, altered into oxy-tuff in the oxidizing environment. To the East, there is andesitic lava on the surface, but for the moment it is difficult to say wether it had any affect on the formation of the oxyandesite tuff.

In this point of view, it was interesting to observe the contact zone of the dikes. When the andesite dike passed in schlier sandstone, a very weak contact effect appeared on the sandstone and a very slight discolouration on the andesite. The same was observed by Bognár and Póka [1], as for the andesite -dikes at Nagybátony. In the latter dikes, calcite appears very frequently, which is probably in connection with the marly facies of the schlieric formation. The pyroxene-content of this andesites often shows a slight chloritization.

The other dikes run in andesite tuff. The phenomena observable here are much the same as in lava flow, on andesite tuff. Since, in the case of dikes, there is a still smaller amount of lava, the effect will be relatively less intense.

.64

This phenomenon can be followed along the valley "Szakadás-gödre", one of the northern branches of the Csevice valley. Here, in the upper part of the valley, a roughly 6 m large, almost perpendicular dike, with 170—350° strike, appears in the agglomeratic andesite tuff. On the contact of the two rocks, both in andesite and andesite tuff, the development of the oxy-character is observable. On a narrow stripe, tuff and dike take a red colour, the dike becoming porous, with slight chloritization.

Results of analyses of both types (1. fresh, grey andesite, 2. red, scoriaceous andesite; analyst: L . JANKOVICH) are following:

Thus, the difference is not considerable, there either, in the degree of oxidation (3,2 and 5,8). In the red oxyandesite O_{Fe} is over 5, but this value is not an extraordinary one. No opacitization takes place here, as the rock is practically free from coloured minerals. The increase in $Fe₂O₃$ -content represents, here too, haematite.

Taking into consideration the trend of the $Fe₂O₃$ ratio, one can point out that only the increase in numerical values is regular in connection with the changes in degree of oxidation, but it was impossible to determine any limit value allowing the delimitation between oxyandesites and other types. There are considerable differences in the $Fe₂O₃/FeO$ ratio between the normal and oxy-varieties of each type:

The determined weight-percent quantities of $Fe₂O₃$ and FeO are recalculated to 100 percent.

Comparing the data of analysis of oxyandesites to those of the fresh rock, one could generally point out that the extreme values of $\rm SiO_2,\ \rm Al_2O_3$ decreased in the oxy-rocks; there is a trend towards equalization in some measure, in oxy-rocks, except for the value of $Fe₂O₃$. Besides, CaO- and MgOcontent also decreased in oxyandesites. The behaviour of the alkalis has not proved regular.

Tu sum up, it was proved that, on the contact of andesite lava or andesite dike with andesite tuff, oxyandesite with considerable haematite-content has been formed, the amount of wich depended on the mass of the lava. The

Fig. J. **Microphotograph of a sandstone inclusion with contact border in oxyandesite-tuff. East from the watch-house of Zsilo, Hasznos. Crossed nicols, magnif. 40 X .**

seemingly capricious 'appearance of red oxyandesite or oxy-tuff actually shows some regularities, and it is due to the different degrees of denudation that we often find, on the surface, normal grey andesite tuff and oxy-tuff, or various varieties of andesite, side by side. In these oxyandesites, there is always a considerable amount of haematite which plays an important part in the red colouration of the rock, too.

When the lava flows on the surface of the tuff, or it forms a dike in the andesite tuff, the temperature of the latter is still over the formation temperature of α -Fe₂O₃, consequently magnetite already present in the lava directly altered into α -Fe₂O₃, and produced the red colour of the rock. The same holds for the magnetite-content of the tuff. The volatile-content of the tuff, on the other hand, primarily facilitates scorification, while secondarily enables the formation of volatile minerals produced by the decomposition of the mineral (chlorite, clay minerals, etc.)

In andesite tuff, the iron-content often appears in the form of $Fe(OH)_{3}$, in that of goethite or, in case of lower p_H , as lepidocrocite. At higher temperature, this will alter into α -Fe₂O₃, too. Consequently the red colouration of oxyandesite tuff is due, first of all, to α -Fe₂O₃.

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