# METASOMATIC PHENOMENA IN THE MÁTRA MOUNTAINS

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#### INTRODUCTION

In areas of orthomagmatism one may repeatedly encounter rock alteration phenomena characterized by devitrification or possibly metamorphism of the groundmass and by the usual conversion of rock-forming minerals into hydrous neoformations. Also, potassium may happen to show considerable secondary enrichments. Thus the original, orthomagmatic, rock changes into a hypomagmatite and the andesite is replaced by various meta-rocks (hydroandesite, etc.), eventually by K-trachyte. It is a K-magmatism-like phenomenon rather than real potash-magmatism that one has to do with in the latter case.

Near the land surface, orthomagmas may acquire some potash character if the sedimentary rock traversed by the magma was rich in potassium. In such cases, transvaporisation takes place. When hot-water solutions migrate across an already-developed orthomagmatite, then dissolution and replacement are more considerable. Accordingly, this mobilization of potassium takes place close to the surface already. Potassium concentration may also increase when this element is bonded to mixed-layer clay minerals in a near-surface, acid or intermediary environment, while sodium and calcium are leached from the original rock.

In the last analysis, since hydrothermal solutions, as a rule, do not contain alkalis in substantial amount, in ascendent solutions they can be enriched solely by the exsolution of the alkali content of earlier-formed alumosilicates. Comparatively great amounts of alkalis can be introduced into ascendent solutions primarily during the weathering of feldspars.

#### THE ROLE OF ASCENDENT SOLUTIONS IN METASOMATISM

During metasomatic processes, in dependence on the quantity and quality of components dissolved in the ascendent solutions, the original rock can be differently altered. The solutions will either seep through the pores, or migrate upwards via fissures and joints. At Mátraszentistván locality, this latter is the case, so that infiltration metasomatism can be spoken of.

During the processes of the volcanic aftermath the original composition of the ascendent solutions may substantially change and the replaced paragenesis may differ considerably from the resulting, new mineral composition. According to KORZHINSKY [1959], the hydrothermal solutions show two extreme tendencies. One is the evolution of the solutions, the other the evolution taking place in the magma chamber, where a fractional distillation of residual solutions occurs. In both cases hydrothermal parageneses of different composition will develop.

In the course of infiltration metasomatism the ascendent solutions rise via joints and fissures produced by tectonic movements. Considering that the effect of the solutions is felt even farther away from the fissures, the influence of the country rock cannot be disregarded. Accordingly, if an ascending hydrothermal solution is taken to be of definite composition and governed by either of the above two tendencies, it is merely the ions exsolved from the country rock that will provoke additional substantial changes. Along with the formation of new, usually volatile-rich, minerals, the chemical composition of the rock may also change substantially. Therefore it is not uninteresting to study the rock sequence crossed by the solutions.

Considering the geological structure of the western Mátra Mountains, the following formations can be reckoned with. The crystalline basement seems to be overlain by Oligocene rocks which, though in reduced thickness, are known to be exposed near Nagybátony, in the northern foreland of the Mátra Mountains. In these Oligocene (Chattian) sandstones the share of glauconite can attain even  $10^{0}/_{0}$ . Since glauconite is a potassium-containing hydrosilicate, so under proper conditions this mineral may become one of the sources of potassium accumulation. According to well-known chemical analyses, the K<sub>2</sub>O content of the Burdigalian "Lower Rhyolite Tuff" varies between 3.22 and 3.47%, this value being considerably higher than the average  $K_2O$  content of the andesites of that region. The 40- to 60-m-thick dacite tuffs of the Helveto-Tortonian boundary have a potassium content similar to that of the rhyolite tuffs of the Burdigalian sequence. The K2O contents of the Helvetian and Lower Tortonian andesites vary between 1.2 and 2.6%, being always lower than the value of Na<sub>2</sub>O. Inasmuch as a hydrothermally altered rock is considered, the  $K_2O$  content may be as high as 5.5% (near the KIOSZ mansion at Mátraszentistván). In this case mainly illitization seems to be responsible for the high K<sub>2</sub>O content [MEZŐSI, 1968]. On the basis of the boreholes Gyöngyösoroszi–2 [Kubovics, 1964] and Mátraszentimre–2 [Mrs. Csillag, 1968] it seems probable that these formations are available beneath the andesite complex throughout the western Mátra Mountains. Therefore, in this territory abundant sources of potassium can be reckoned with.

As regards the geological structure of the territory, it could be stated that the area affected by metasomatism belongs to a NW-SE-trending system of faults, forming a part of the heavily block-faulted sequence occurring in the vicinity of Nagybátony [MEZŐSI, 1969]. Along fissures and faults, alteration is more pronounced.

On the basis of the results of air-spectrometric surveying, WÉBER and GÉRESI [1970] showed the vicinity of Mátraszentistván to be an area where the potassium content is comparatively higher, K-metasomatism associated with volcanism being more pronounced there. These authors also showed the highest concentrations of potassium to be controlled by tectonic zones. Thus an earlier statement of the present writer [1968] has been confirmed by investigations of this kind.

The ample variety of the manifestations of metasomatism in the area under consideration can also be explained by the fact that the front of metasomatism does not shift with constant velocity. On one hand, this is due to the extremely irregular variation of the texture, composition and porosity of the rock owing to the random distribution of thin lava flows and interbedded tuff layers; on the other hand, it is due to the presence of fracture lines. Therefore there is no real zone of replacement in which the same mineralization could develop.

Because of the low concentration of ions in the migrating hydrothermal solutions the rock-dissolving effect of the solutions will be considerable. Solution is also promoted by the comparatively higher temperature — a factor which can always be reckoned with in the course of hydrothermal metasomatism.

Grain size also influences the dissolution of minerals. In general, solubility increases with decreasing grain size. This can be explained by the fact that on the grain surface the number of ions of unbalanced charge increases greatly as compared to the unbalanced ions inside the crystal. Thus if the dissolution of the mineral grains of a clastic rock were examined in comparison to the crystals of different size of a magmatic rock, one would find that in the clastic rock the same mineral is dissolved more easily, i. e. an alkali is leached from the sanidine fragments of rhyolite tuffs or from the glauconite of sandstones more easily than from a relatively intact crystal component of magmatites.

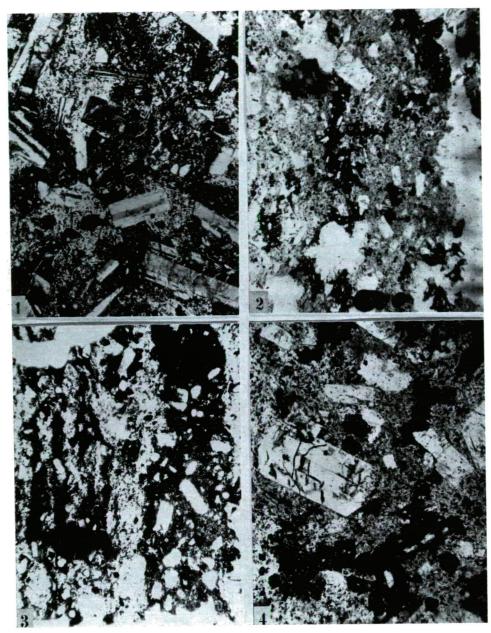
### THE PROCESS OF ZONATION

During dissolution, reactions may set in, provoking a change in the concentration of  $H^+$ -ions, reactions playing an important geological role in the solubility behaviour of mineral compounds. In addition to this, the variation of pH influences the valency, i. e. the degree of ionization. On the other hand, an increase in pH will promote oxidation — an important geochemical factor during metasomatism.

As shown by KORZHINSKY's investigations [1959], the solutions first show an increasing acidity as manifested by autometasomatic reactions of comparatively higher temperature. In the course of their slow migration the solutions will react with the country rock and their acidity will thus gradually decrease. The leached substances and/or the precipitated components will exhibit a zonation. In the direction of migration, i. e. away from the fissure or joint, the following zones can be distinguished: 1. a zone of increasing acidity, where the rocks are exposed to leaching and where precipitation is insignificant, 2. a zone of maximum leaching with mineral precipitations, and finally, 3. a zone of decreasing acidity with a decline of leaching and an increase of mineral precipitations. In the last analysis, the development of zonation is a natural process.

In his classification of infiltration metasomatism SZADECZKY-KARDOSS [1958] preserved the above tripartite division. Proceeding from the ore body towards the magmatite, he distinguished: 1. an ore-bearing zone, 2. a silico-magmatite zone and 3. a hydromagmatite zone. Under special conditions this zonal pattern may change.

The occurrence of zonation has been observed in Hungary, too. Thus V. Széky-Fux [1964] showed the presence of kaolinization with a low con-



Figs. 1-4.

centration of precious metals in a reduced vertical, and greater lateral, extension close to the surface at Telkibánya, North Hungary. At 400 to 500 m depth the function of kaolinite is taken over by montmorillonite containing somewhat richer ore mineralization. Wherever the fissures or joints are closely spaced, the original ortho-rock has been totally altered into andesitogenic propylite. Where spacing is wider, there relics of fresh ortho-rock may occur between the fissures or joints.

Zonation can also be shown to occur in a dike at Mátraszentimre [NAGY, BARBÁCSI, 1966]. On both sides, the dike is similarly altered in a width of some 50 m. The plagioclases of the country rock are completely sericitized, the pyroxenes are chloritized or they have been totally dissolved out of the rock. In the cavities, precipitations of silica gel can often be observed. Close to the dike, the country rock is heavily silicified. Away from the boundary of this zone, over a distance of about 80 m, the feldspars are still sericitized, but the pyroxenes have remained fresh.

A zonation can also be caused by the variation of both the temperature and composition of the fluctuating solutions. On Mt. Hidegkút, western Mátra Mountains, KUBOVICS [1966] discovered K-metasomatism of comparatively higher temperature as manifested by the formation of sanidine, adular, glauconite, seladonite and zeolite. The lower-temperature phase has produced mainly SiO<sub>2</sub> modifications.

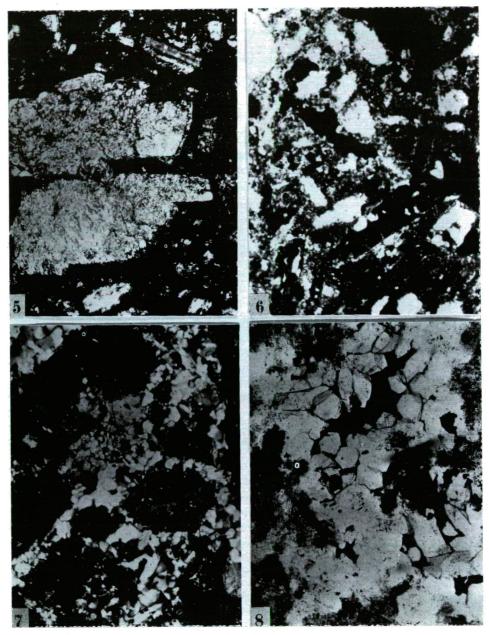
# EXAMINATION OF THE ROCK WITH MICROSCOPE AND DERIVATOGRAPH

In the metasomatic alteration of the andesites of Mátraszentistván a kind of regularity, developmental sequence, can be recognized. The fresh or just slightly altered rock is of dark-grey colour with red ochreous bands of just a few mm width running parallel to the surface or along the joints. The rock of this kind is compact, the feldspars in it being fresh, with bright, glistering joint planes. As shown by the microscopic image of the rock, the groundmass has acquired a mottle-patterned habit in the initial stage and the lighter parts have lost their magnetite content. At the same time, the groundmass has been more or less devitrified (Fig. 1). Using just one nicol, one can readly observe that the magnetite grains have been dissolved and the ore has been concentrated along the joints (Fig. 2) or, eventually, that it has stained the groundmass in a zonal pattern (Fig. 3). At this stage the feldspars are still fresh, twinning and zoning being quite distinct upon them. However, the alteration of pyroxene does set in. In fact, pyroxene has been totally replaced by seladonite, fibrous, slightly pleochroic, forming pseudomorphs in many cases (Fig. 4). The alkali content of a rock of this kind does not change yet essentially, the increase of potassium content may still be quite insignificant (Table I). The feldspars of such rocks may be completely sericitized either partly as a result of the effect of surface agents, or under the influence of hydrothermal solutions (Fig. 5). This is partly responsible for the higher potassium content observable in such cases.

Fig. 1. Devitrified, mottled groundmass, + N,  $80 \times$ 

Fig. 2. Ore concentration in the mottles of the mottlepatterned groundmass, 1 N,  $32 \times Fig.$  3. Zonal ore accumulation, 1 N,  $32 \times$ 

Fig. 4. Fresh feldspar associated with seladonitic pyroxene, + N,  $80 \times$ 



Figs. 5-8.

Silicification is first manifested when the feldspars are still comparatively fresh and when the femic components already show an advanced ore mineralization (Fig. 6). Quartz either shows an irregular mode of occurrence, often dissecting the groundmass in a net-like pattern (Fig. 7), or it forms crystals of hexagonal cross-section grown on the walls of cavities in the rock and filled up additionally by limonite (Fig. 8). Leaching may lead to an increase of porosity, i. e. to the formation of cavities of different size. First of all, limonite precipitated on the walls of the cavities and then their inner space was filled up by fine-grained chlorite which has been identified as pennine. In such cases the pennine aggregate is surrounded by an ore frame (Fig. 9). Sometimes, not only the femic mineral, but also the altered feldspar is oreframed (Fig. 10).

TABLE 1

Sample number	rock habit	Na <sub>2</sub> O <sup>0</sup> /0	K <sub>2</sub> O <sup>0</sup> /0
1.	ochre-yellow, slightly porous	0,72	11,50
2.	pinkish to ochre-coloured, porous	0,10	. 10,80
3.	reddish to ochre-coloured, porous	0,58	10,09
4.	dark-grey, compact	2,25	2,18
5.	greyish-pink, slightly porous	0,40	11,30
6.	pinkish to light-grey	0,05	10,10
7.	reddish to purple, slightly porous	0,10	10,57
8.	grey, compact	2,89	2,53
9.	pink, slightly porous, with yellow and white mottles	0,10	9,65
10.	purple to pink, compact	0,30	10,99
11.	pink, slightly porous, with white and yellow mottles	6,10	6,10
· 12.	dark-grey, compact, with ochreous bands	2,90	3,36
13.	dark-grey, compact	2,65	2,66
14.	dark-grey, compact, with a thin oxide coating	1,25	8,53
15.	grey, slightly porous, with pink bands	0,35	. 12,00

In orthomagmatites the degree of oxidation increases under the influence of hydrothermal solutions and magnetite develops into  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> which is known from earlier investigations [MEZŐSI, 1967]. The magnetite lattice develops into a hematite lattice rather easily. In fact, with additional metasomatic effect, the iron may be precipitated as iron(III)-hydroxide in fissures or joints at the level of the oxidation zone. Iron(III)-hydroxide may, on its turn, develop into goethite. Accordingly, on account of the behaviour of iron the bleaching, discolouring of the rock corresponds to the final phase of metasomatism. Also, this is the reason why the femic minerals, if any, are often subordinate in such rocks.

The nature of the iron oxide phase precipitated under metasomatic effect is also indicated by the following results of analyses. Of the powder of 'a porous, reddish to ochre-coloured rock a derivatogram was made. The total iron content of the rock was found to be 4.58%, of which the share of Fe<sup>2+</sup> was as low as  $0,7^{0}/_{0}$  (Fig. 11). Between 70 and 200°C the DTA curve showed

Fig. 5. Sericitized feldspar pseudomorph, + N,  $80 \times$ Fig. 6. Ore-mineralized, thermal mineral with accumulation of quartz, + N,  $80 \times$ Fig. 7. Groundmass impregnated by quartz in a net-like pattern, + N,  $80 \times$ Fig. 8. Idiomorphic quartz accumulated in cavities, 1 N,  $80 \times$ 

some weight loss which on the TG curve corresponded to a weight loss of  $0.45^{\circ}/_{0}$ . In correspondance with this, an endothermic peak appeared on the DTA curve as a result of water lost partly by clay minerals, partly by adsorption. At  $310^{\circ}$ C on the DTG curve, again, a pronounced weight loss

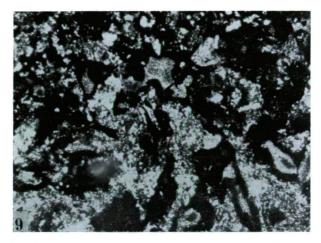


Fig. 9. Rhytmical precipitation of limonite and pennine in cavities, + N, 80imes

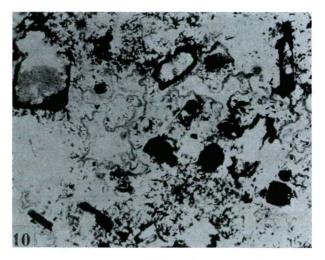


Fig. 10. Ore-framed, altered feldspar in porous rock, 1 N, 80imes

presents itself which corresponds to  $0.78^{\circ}/_{\circ}$  as measured on the TG curve. However, on the exothermic peak appearing at 350°C on the DTA curve an inflexion appears at 310°C. Properly, here an endothermic reaction is concealed by an exothermic one. The exothermic reaction is the alteration of goethite into  $\gamma$ -Fe<sub>2</sub>O<sub>3</sub> showing two exothermic peaks — one at 310°C, one at about 540°C, a phenomenon indicating the alteration of this modification.

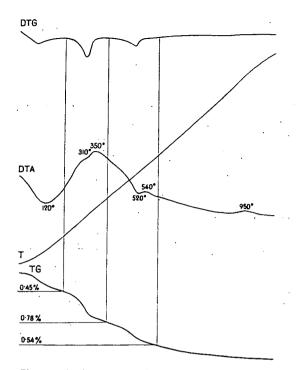


Fig. 11. Derivatogram of metasomatized andesite

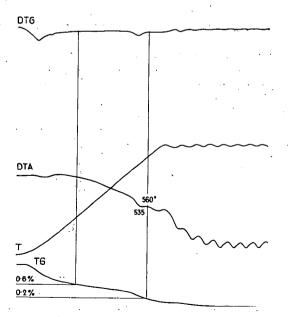


Fig. 12. Derivatogram of metasomatized andesite held at a temperature of 400°C

This could also be proved by that the curve, obtained for the sample at 400°C, showed nothing else than the higher exothermic peak of the alteration  $\gamma \rightarrow a$ , i. e. a change in modification no longer accompanied by any change in weight. However, a weight loss of  $0.2^{0/0}$  due to water released during the break-down of the illite structure could still be observed within the 480° to 560°C range (*Fig. 12*).

As referred to earlier, when plagioclase is replaced, the mineral gets first mottled, then it is deprived of both the zonal structure and twin intergrowth characteristic of orthoandesites of normal development.

Since in the course of andesite metasomatism the species of feldspar corresponds more or less to the labradorite series, let us quote LAGACHE's [1965] experiments in a few lines. He examined the dissolution of labradorite, albite and adular at different temperature and  $CO_2$  pressure under the conditions of identical specific surface area.

During his experiments LAGACHE came to the following conclusions. Alkalis are the most easily soluble elements. The concentration of alkalis in solution increases as a function of time, being proportional to the surface area of the crystal exposed to solution. With a rise in temperature and CO, pressure, the rate of dissolution will increase. Silicium behaves differently. Initially the rate of its dissolving is higher than that of alkalis, but after attaining saturation, it does not show any substantial change anymore. The saturation limit is slightly influenced by the species of feldspar, by temperature and by the pressure of CO<sub>3</sub>. Testing albite and adular, he used also such a method in which at the end of a given time unit the solution was decanted from the sample being tested and the sample was again diluted with distilled water. After fivefold repetition no substantial difference in the quantity of dissolved substance could be observed (in case of albite 28.6 - 31.0 ppm Na<sub>2</sub>O and 159–183 ppm SiO<sub>2</sub>, in case of adular 24–29 ppm  $K_2O$  and 112–140 ppm SiO<sub>2</sub>). In connection with this change, it should be borne in mind, that in the fourth and fifth cases the surface was in a heavily dissolved state — hence the lower value. Aluminium, practically, was not dissolved at all.

With labradorite, the same could be observed as in the case of albite or adular in respect of the dissolution of sodium, aluminium and silicium. Calcium, however, precipitated in the form of calcite which was quite logical under the given experimental conditions.

As shown by quantitative analyses, adular was dissolved most slowly, albite most rapidly. The rate of dissolution first shows an exponential rise, later a marked decline.

The plagioclases of the labradorite series do not represent a stable state, as notified also by LAVES [1960] who drew the conclusion that no feldspar of stable phase could be expected to occur between  $An_{30}$  and  $An_{70}$ .

As confirmed experimentally by WYART and SABATIER [1956], at a relatively low temperature (500°C and at a pressure of 600 bar), an orthoclase, treated by an aqueous solution of NaCl, will change into albite. However, in the case of anorthite a replacement of this kind was impossible. Albite, in turn, did alter into microline at 350°C upon treatment by aqueous KCl solution. Accordingly, Na and K are easily interchangeable, but for an exchange of Ca, the Si/Al.ratio must change, too. Consequently, the above investigations confirmed the assumption that during metasomatism, under proper conditions, labradorite alters into albite rather easily and that albite, on its turn, does so into sanidine.

In the course of K-metasomatism, when plagioclase was replaced, a zonal structure rather than a mottled pattern could be observed in certain cases. Investigating the causes of the two different forms of appearance, the present writer has come to conclude that at certain orientation the labradorite replaced by sanidine will always become zoned, at some other orientation, mottled. Precisely, when the cross-section was perpendicular or subperpendicular to crystal axis b, the zonal structure was observed, at orientations other than this the mottled pattern appeared (*Fig. 13*). The zones were always

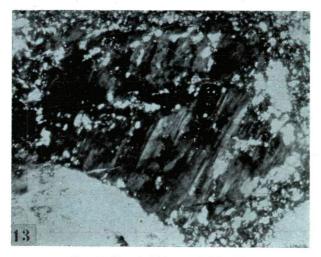


Fig. 13. Zoned feldspar. + N, 200imes

parallel to face 001. As for feldspar structures, it is well-known that in the direction of crystal axis "a" there are chains in which the cations of the second order are situated. Hence, it looks probable that the zones exhibit structural elements connected with metasomatic replacement, elements which can be visualized optically.

BAMBAUER and LAVES [1960] observed a zonal structure in adular. This, however, was due to slow growth and the individual zones showed different degrees of arrangement.

## X-RAY ANALYSES

Numerous X-ray diffractograms were made of bulk rock and selected samples (26 kV, 18 mA, CuK $\alpha$  radiation). The interpreters of these had to answer the following questions: Can the degree of alteration be traced in the bulk samples of differently metasomatized rocks? Which of the characteristic lattice planes are expected to occur on the diffractogram of a comparatively fresh rock? Which planes of the lattice can be considered characteristic of a metasomatized rock.

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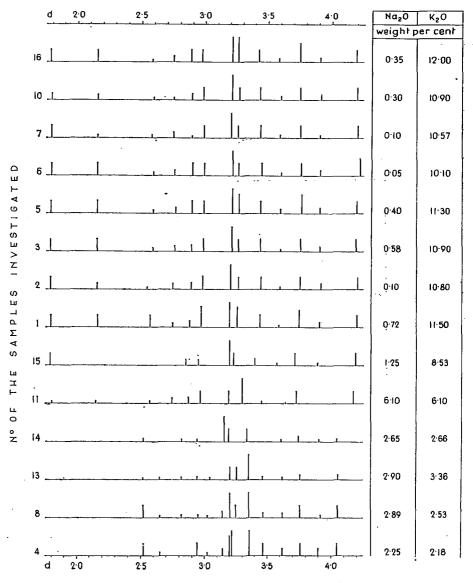


Fig. 14. d values of test samples as referred to feldspar

Considering the results of CORLETT and EBERHARD [1967], one can point out that lattice planes 131 and 131 were always present on the X-ray diffractograms of those rock samples which were of dark-grey colour, which contained fresh feldspar and which could be considered to represent an ortho-rock even from the point of view of alkali content. On the basis of the difference in the 2  $\Theta$  value of the two lattice planes a plagioclase containing An in 50 to 60% could be identified. With the appearance of sanidine, plane 131 will vanish. According to CALLERGI and DE PIERI [1967], the value of  $d_{201}$  can be of use for the determination of the sodium content of sanidine. In the test samples the corresponding *d* value varied between 4.15 and 4.23, a fact hinting at the presence of a transition between sodium- and potasium-rich members, with 10 to 20% *Ab*. The results of BOWEN and TUTTLE [1950] led to similar interpretation.

. Fig. 14 shows the d values of a few samples as referred to feldspar. The size of the lines is proportional to the value of intensity. The numerals are serial numbers of test samples. In the tabulation the Na<sub>2</sub>O and K<sub>2</sub>O contents of the samples have also been shown.

During the interpretation of X-ray diffractograms the degree of Si/Al arrangement could also be assessed. If this value was about 1.6 there was just an inversion point among the elements of symmetry. In this case the planes  $d_{hkl}$ of higher intensity were split up into planes of lower intensity. Consequently, the line corresponding to the value of 131 and  $1\overline{31}$  d, or the lines  $24\overline{1}$  and  $\overline{241}$ , respectively, made their appearance. However, because of the statistical degree of Si/Al arrangement the plane of symmetry parallel to face 010 appeared with 6%  $K_2O$  already (the angle formed by faces 001  $\wedge$  010 being then as high as 90°), and the splitting of the planes  $d_{hkl}$ ,  $d_{hk0}$  and  $d_{0kl}$  ceased. Consequently, the planes 131 and 241 must also have missed. Since sanidine shows an excellent jointing along planes 0k0 and 001, because of the plane of symmetry, the planes 040, 060 and 001 have come even more into prominence and their intensity too was comparatively high. Still absent in labradorite, the high-intensity line 060 is always present in the case of sanidine. In this latter the plane 204 was always represented by a high intensity, whereas it was absent in labradorite. It could also be observed that the increase of symmetry was accompanied by an increase in the d value of lattice plane 201.

In the last analysis, it can be concluded that the metasomatic replacement of feldspars can be analysed by X-ray methods. Since higher potassium content is often indicative of ore accumulation, the above results are also important from the practical point of view.

### SUMMARY

The variation of the composition of ascendent solutions is greatly influenced by the geological sequence intersected. In the western Mátra Mountains, glauconitic sandstones, rhyolite tuffs and dacite tuffs are of interest from this point of view. Along fracture lines, the ascendent solutions provoke more or less heavy alteration of the rock, bringing about an infiltration metasomatism. During metasomatism, along fracture lines a zonation corresponding to the degree of alteration will develop.

In the process of metasomatic alteration of the Mátraszentistván andesites some regularity can be observed. At the initial stage the groundmass of the fresh, dark-grey andesite becomes mottle-patterned, being more or less devitrified. At the same time, the magnetite grains are dissolved. At this stage, feldspars are still fresh (intact), whereas pyroxenes are replaced by sanidine, fibrous, slightly pleochrous. Silicification usually sets in when the femic components already show a high degree of metallization. As for quartz, it is either irregularly scattered, or dissects the groundmass in a net-like pattern. On account of leaching, porosity may increase and in such cases quartz may appear grown freely on the walls of cavities. During K-metasomatism, plagioclase is sometimes zoned rather than mottle-patterned. These zones run parallel to face 001.

As observed by the present writer while interpreting X-ray diffractograms, with an increase of potassium content the conditions of symmetry also changed, the splitting of the  $d_{hkl}$  planes ceased and lattice planes 040, 060 and 002 manifested themselves with high intensity.

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