Fe-Ti OXIDE MINERALS IN TRANSDANUBIAN (WESTERN HUNGARY) BASALTS

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SUMMARY

In samples taken from Transdanubian basalt profiles, the following Fe-Ti oxide minerals have been identified: ulvöspinel, magnetite, titanomagnetite, titanomaghemite, hematite and iron hydroxide varieties. New in Hungarian magmatites are ulvöspinel and titanomaghemite. Analysis of the pressure and temperature relations of lava flows has revealed some relationships between quantity, quality and size of the crystalline phases. Results are compared with similar investigations of the author on a lava flow in the South Caucasus (Georgia, Soviet Union) and with internationally published results.

Hungarian literature did not discuss so far in any detail the iron-titanium oxide minerals of the Transdanubian basaltic rocks. Observations were limited to recording of the phenomena visible in thin sections. Some authors described magnetite, ilmenite and iron hydroxide types, without observing their solid solutions in each other, which, as a matter of course, requires the application of ore microscopy.

As in other mineralogical problems concerning Transdanubian basalts, it was Hermann (1875–78) who first revealed a remarkably sharp insight into the problems of the opaque minerals. He described the magnetite as chiefly octahedral, rarely xenomorphic. He stated ilmenite to be a rather scarce mineral occuring as thin lamellae. He noted that "surely magnetite too, as is normal in basaltic rocks, has a large Ti content" (!), although the presence of titanomagnetite (or of the dissolution of titanium minerals in magnetite) was proved only much later by ore microscopical investigations. According to Mauritz and Harwood (1937) the smaller magnetite grains are mostly idiomorphic; growth forms – skeletons, indented bars, lattices – are common. These authors call the translucent variety of ilmenite of 12 to 20 microns in diameter and 1–2 microns in thickness a titanium-iron mica. From the matrix of the basalt, Jugovics (1955–56) described magnetite, ilmenite and ilmenite mica (probably the same as the titanium-iron mica observed by Mauritz and Harwood). He observed also microlites of ore minerals in the matrix, which he considered as secondary. Vörös (1963), investigating the basalt area of the Kabhegy (Transdanubia, Hungary) described the oriented unmixing of an opaque material in olivine and suggests – on the basis of his studies on lapilli of pyroclastic basalts – that unmixing had chiefly taken place in the last phases of crystallization, after the lava-flows had risen to the surface.

In international literature, the investigation of the Fe-Ti-oxide minerals of basic magmatites (mostly of intrusives) looks back on a past of several decades. A complete presentation of this literature is therefore out of question; let us emphasize only those important and recent results, which facilitated the interpretation of the results. After Mogensen's (1946) and Ramdohr's fundamental publications, it was Vincent (1960) who first gave lattice constants measured on natural phases of ulvöspinel ($a_0 = 8,431$ Å); on the other hand the most recent among investigation on synthetic ulvöspinel is that of Forster and Hall (1965): according to these authors "a" ranges from 8,521 Å to 8,538 Å; Ti^{4+} is in eightfold coordination; the degree of inversion of the spinel structure is 0,92. According to several authors, the Curie point of pure ulvöspinel is at $-150°$ centigrade (!); hence, ulvöspinel unmixed in magnetite under normal pt conditions will, as it were, magnetically dilute the magnetite. Of the minerals of the Fe-Ti oxide triangle, magnetite is most abundant; its study is most important also in research into paleomagnetism. According to the published results it always contains a little Ti, and unmixing products are common in it (ilmenite and/or ulvöspinel); according to several authors (Vincent et al. 1957), if both are present it is the ilmenite that unmixes first. Already Newhouse (1936), later Ade-Hall and Wilson (1963) observed that the morphology of magnetite in the Scottish basalts suggests a crystallization subsequent to that of the plagioclases. In the abundant literature on titanomagnetite some authors do not restrict this term unequivocally to optically and roentgenographically homogeneous titanium-bearing magnetite but extend it to any chemical compound situated on the $Fe_3O_4 - FeTiO_3$ line (or near to it) in the Fe-Ti-oxide triangle. The danger of this is that magnetite with ulvöspinel and ilmenite in solid solutions is – because of its chemical composition – also qualified as titanomagnetite. Titanomagnetite literature includes profound investigations by Vincent et al. (1954, 1957), the Japanese authors Katsura, Kushiro, Akimoto et al. (1960, 1961, 1962) and Wright (1959), Basta (1959), Buddington et al. (1955, 1963), Meitzner (1963) and many others. In intrusive rocks the independent ilmenite phase is common, which is easily investigated because of its grain size. One often observes ilmenite dissolved in magnetite (titanomagnetite), too. Several authors, e.g. Katsura et al. (1962) have detected the ilmenite-hematite unmixing in Hawaiian soils. Separate mention is due to Buddington's and his co-authors' terminological achievements: they cleared the jungle of terms in several publications, their most important merit being the emphasizing of the rarity of pure phases in the iron-titanium-oxide triangle, and of the abundance of transitional types. The name titanomaghemite is due to Basta (1959), who measured an *a* of 8,342 Å. According to Akimoto and Kushiro (1960), and later to Katsura and Kushiro (1961), this mineral is fairly common, in volcanic rocks: it can readily be demonstrated particularly by correct ore-microscopical and chemical procedures. They found this mineral to have in most cases an irregular shape, either on the rim of titanomagnetite or along its fissures. As to the origin of titanomaghemite, opinions are different: Basta did not discuss the primary or secondary character of the processes of oxidation to which titanomaghemite is due; Akimoto and Kushiro mentioned decomposition, or weathering, but they not enter upon the particulars of this subject. Katsura and Kushiro made a step forward: according to their findings, titanomaghemite can develop under the influence of hydrotermal solutions, or of vapors (as witness the titanomaghemite in rocks decomposed in such a way), but the fact that its quantity increases towards the surface of the lava flow suggests an origin by weathering (secondary formation). Katsura et al. (1962) put forward arguments in favour of primary origin. Much fewer papers deal with the role of hematite; only Basta and Katsura et al. investigated it in connection with the titanomagnetite problem. The latter have proved by X-ray methods the presence of a trigonal ilmenite-hematite phase in the samples. Publications on pseudobrookite playing a role in the iron-titanium-oxide triangle are few and far between. One reason for this may be the absence of pseudobrookite in the investigated intrusive rocks, another one may be the instability of the pseudobrookite lattice: this Fe-Ti phase of maximum oxidation is rare even in effusive rocks. In 1957 Akimoto and his co-workers observed a pseudobrookite-like substance on examining a synthetic material between $Fe^{2+}Ti_2O_5$ and $Fe_2^{3+}TiO_5$ in composition; in 1959, Wright, applying heating methods, proved the presence of pseudobrookite in samples of the Skaergaard intrusion. Among the recent publications, Frenzel's paper (1954–55) is remarkable, which discusses the pseudobrookite problem in its relation to the more important Fe-Ti minerals. The pre-cited literature does not examine the role of iron hydroxides; their presence is attributed to secondary processes due to supergene effects.

As it is evident from the above citations, international literature discusses in great detail the iron-titanium oxide minerals. Every author takes pains to explain genetically the development or evolution of the examined phase. Still, it is remarkable that the papers – aside from some praiseworthy exceptions – are characterized by an effort towards the most perfect description, while genetics plays a more subordinate role, being limited at best to the examined phase, or the association of a few phases. A striking example is the excellent paper on titanomaghemite in basaltogenic soils by Katsura et al. (1962) which includes no examination in detail of the original basaltic rock. Another case in point is one recent French publication (Colin-Poyet 1965) which, using the microsonde methode, makes no attempt at all at the investigation and interpretation of the geological profile. On the other hand, Vincent and his co-workers investigating the Skaergaard intrusion performed an exemplary set of observations and reached geological conclusions by studying their object in geological space rather than on a few rock samples taken at random.

Methods of investigation

Investigations into the iron-titanium oxide minerals of basaltic rocks have included ore microscopy, X-ray analysis, chemical and magnetic methods.

Ore-microscopy: Traditional methods have been used; the polished sections have been examined also after etching (with cc HF) to assure the good observation of structural relations and intergrowth of magnetite and to enable a certain distinction between titanomagnetite and titanomaghemite.

X-ray methods: diffractograms made on some original basalt samples have reliably identified magnetite only, which is the most common opaque mineral. To improve the situation, the author has hand-picked samples under the stereo-microscope; Debye–Scherrer analysis has then been performed on two samples. Even so one sample has been found to be insufficient: only the magnetite could be reliably identified.

Chemical analyses: similarly to the X-ray methods, analyses performed on the original basalt-samples were irrelevant as to the characteristics of the Fe-Ti-oxide minerals. In the future it will be desirable to use microanalytical methods for which the samples prepared for X-ray investigations are sufficient.

Magnetic methods: to prepare the samples for X-ray and chemical analysis, magnetic separation has been used after pulverizing and settling, but manual separation, mentioned above, has proved more succesful. In the course of the paleomagnetic study of Transdanubian basaltic rocks Márton (1963) examined in detail the magnetic features of these rocks by different magnetic methods.

Results of the investigations

In the Transdanubian basaltic rocks the following iron-titanium oxide minerals have so far been identified: ulvöspinel, magnetite, titanomagnetite, ilmenite, titanomaghemite, hematite, pseudobrookite, and iron-hydroxide varieties.

Ulvöspinel: Of micron size, it was very difficult to identify ore-microscopically. Nor was it observed as yet as an individual mineral, only as an intergrowth in magnetite, which often included ilmenite too (Plate I, fig. 1–2). Consequently, the author had to lean heavily on bibliographic data. Ulvöspinel seems to be parallel to the octahedral faces of magnetite. In many cases, it is only the presence of these intergrowths that reveals the polycrystalline character of magnetite. Etching shows up on the homogeneous magnetite surface two kinds of field: one bears segregations of ulvöspinel, the other is truly homogeneous. This suggests the continued oriented growth of magnetite at lower temperatures. Ulvöspinel occurs – in keeping with its composition – in the most reductive environment, where the more oxidized types of the iron-titanium-oxide triangle are absent. In the geological profile ulvöspinel occurs in the middle of the lava-flow, or somewhat below. In some samples (Tótihegy) certain grains of typically titanomaghemitic titanomagnetite are seen to include segregations of ulvöspinel.

Magnetite is the most common opaque mineral of Transdanubian basalts. In some samples it is fairly idiomorphic; in other cases the xenomorphic type, which at first sight seems to be corroded, is frequent. However, this is not the result of a secondary process, but depends on the timing of magnetite crystallization: it is obvious in many cases that this magnetite type fills the interstices between the minerals formed earlier (olivine, pyroxene, felspars). The triangular or square shapes of idiomorphic grains and their skeletal intergrowths and growth forms in some basalts set the crystallization of magnetite at the beginning of the crystallization of the lava-flow on the surface: it cannot originate from the magma, because these thin plates could not resist friction still strong in the molten lava; skeletal growth indicates superficial crystallization, a relatively quick cooling of the lava-flows (this is why this type of magnetite is common in the vitreous – i.e. quickly cooled – basalts). Magnetite is often platy, which may also be due to the limited space for crystallization: magnetite developing in the last phase of felspar crystallization finds place between the felspar laths only: in there it can still be idiomorphic, however. On the basis of ilmenite segregations the magnetite platelets are parallel to (111).

Titanomagnetite: ore-microscopically determined titanomagnetite which term shall refer in the following to chemically, roentgenographically homogeneous titaniferous magnetite) is common enough in the Transdanubian basalts. In the geological profile of the lava-flow, it occurs higher up than magnetite, chiefly in the upper third of the lava-flow, or indeed at its surface. As opposed to magnetite, it cannot be demonstrated in all Transdanubian basalts. Its size and shapes are similar to those of magnetite. Titanomagnetite but rarely includes segregations of ulvöspinel; on the other hand, segregated ilmenite is not abundant, but common enough in it. In the upper parts of the lava-flows, the rim of titanomagnetite is altered more or less deep into titanomaghemite, the smaller grains even completely so.

Ilmenite: common in every Transdanubian basalts, it is identified ore-microscopically more as a segregation in magnetite, titanomagnetite and hematite than a separate mineral (Table I, fig. 1–2–3–4). Skeletal idiomorphic ilmenite occurs chiefly in the upper level of the lava-flow. Segregation may be oriented, mostly parallel to the octahedral plates of magnetite; in this way beautiful triangular forms come to exist. The segregated grains are sometimes very dense, in which case the quantity of ilmenite approaches or exceeds that of magnetite. The reflexion intensity of densely segregated ilmenite is in some cases higher than average; also its reflexion is lighter, whitish, sometimes slightly bluish-white: it is thus likely, that during the segregation of abundant ilmenite, some trivalent iron enters into the ilmenite (ferri-ilmenite). In other cases, ilmenite segregates in the form of parallel laths, or wider bands, apparently parallel to the (100) directions of magnetite. Finally, segregation can develop in irregular spots in the magnetite, or on its rim: in this latter case, magnetite-ilmenite-intergrowths or continued growth is also possible. The reflexion of parallel-oriented or irregularly segregated ilmenite is a dull white, as is that of the types of segregation parallel to (111). Its anisotropy, readily observable in the thicker slabs or in the larger spots is bright, the colour-effect is a deep brown. This shows that between these types of segregation the difference is in composition as well as in orientation. Ilmenite is an ore microscopically obvious segregated phase also in hematite: segregation occurs in all three types mentioned above, but the reflexion invariably suggests ferri-ilmenite. Thin sections show in many cases transparent or semi-transparent membrane – like form of ilmenite of micron size, described already by Hofmann (1875–78), Mauritz – Harwood (1937) and Jugovics (1955–56). The more exact determination of these is beyond the scope of the present work.

Titanomaghemite: occurs in the upper parts of some lava-flows or cones, always together with titanomagnetite: the rims or fissures of the larger titanomagnetite grains are altered into titanomaghemite with sharp, but not straight limits: the boundary is labyrinthine (Plate II, fig. 1). In the smaller titanomagnetite grains only the core is fresh, or even that is altered into titanomaghemite. Even without etching titanomaghemite is easily demonstrated, but it is much more evident on etching with cc HF. Up till now the author could not identify any segregations in it.

Hematite is common in the upper, or superficial part of the lava-flows, mostly as thin, semi-transparent plates. Its size is considerably smaller than that of the opaque minerals in the deeper parts of the lava-flows. Ilmenite (ferri-ilmenite) segregations are frequent in it (Plate I. fig. 4).

Pseudobrookite: up till now pseudobrookite has not been demonstrated by ore-microscopic methods; its presence is suggested only by the X-ray diffractogramm of a sample from Mt. Gulács.

Iron-hydroxide varieties are common in the highest levels of lava-flows, often around the hematite grains. The determination of these has not been envisaged in the present paper. The material is apparently chiefly goethite.

Genetic evaluation

Author's observations concerning the dimensional, abundance and quality distribution of iron-titanium oxide minerals in the Transdanubian basaltic rocks and in dolerites from Georgia (Dmanisi, South Caucasus, Soviet Union, examining for the sake of a genetic comparison) have led to the following results (Table 1):

1. The abundance distribution of Fe-Ti-oxides shows some differentiation: the opaque minerals of greater specific gravity are condensed at the lower levels of the ancient lava-flows. A good example for this is the uppermost lava-flow of the Kabhegy. However, considering its dimensions, this enrichment probably is the result of a small-scale differentiation of 1 or 2 m depth. In the thicker lava sheets (Uzsabánya) and in the basalt cones it is already more difficult to prove any regularity, because repeated fluctuations presumably disturbed more often the uniformity of distribution until the lava solidified; this effect was evidently much weaker in the investigated lava-flow of the Kabhegy. According to the author's observations, the superficial crystallization of most of the opaque material, as well as the prolongation of its crystallization up to the last phases of solidification of the lava-flow does not leat any possibility for differentiation by gravity of the opaque grains developing in this phase.

A

thickness of lava-flow ↓	1.	2.	3.
3,8 – 5,0 m	not measurable owing to small size		takes no polish
5,0 – 6,0 m			
6,0 – 7,2 m			hematite-ilmenite
7,2 – 9,6 m			hematite-ilmenite
9,6 – 12,4 m	3,2%	34	hematite-ilmenite
12,4 – 14,6 m	1,6%	50	magnetite-ilmenite (-ulvöspinel ?)
14,6 – 16,0 m	2,6%	90	magnetite-ilmenite-ulvöspinel
16,0 – 17,5 m	2,3%	40	magnetite-ilmenite-ulvöspinel (abundant)
17,5 – 19,0 m	3,7%	65	magnetite-ilmenite-ulvöspinel (scarce)
19,0 – 21,0 m	5.0%	63	magnetite-ilmenite-ulvöspinel (scarce)

B

thicknes of lava sheet ~ 30 m ↓	1	2.	3.
	3,0%	60	magnetite-ilmenite-ulvöspinel
	3,6%	60	magnetite-ilmenite-ulvöspinel
	2,4%	50	magnetite-ilmenite-ulvöspinel
	2,8%	66	titanomagnetite-titanomaghemite
	3,6%	39	magnetite-ilmenite-ulvöspinel (abundant)
	3,6%	75	magnetite-ilmenite-ulvöspinel (scarce)
	2,1%	51	magnetite-ilmenite-ulvöspinel
	3,5%	57	magnetite-ilmenite-ulvöspinel
	3,5%	83	magnetite-ilmenite-ulvöspinel
	3,0%	51	magnetite-ilmenite-ulvöspinel

C

thicknes of lava-flow ~ 10 m ↓	3.
	Magnetite skeleton, hematite
	magnetite-ilmenite skeleton, titanomaghemite (?)
	hematite and magnetite-ilmenite skeleton
	magnetite-ilmenite skeleton, hematite
	magnetite-ilmenite, hematite
	magnetite-ilmenite (-ulvöspinel ?), hematite-ilmenite
	magnetite-ilmenite-ulvöspinel
	magnetite-ilmenite skeleton

Table 1: Distribution of Fe-Ti-oxide minerals in geological profile of lava-flow (sheet) *A:* Kabhegy, upper lava-flow (drilling no. 5). *B:* Uzsabánya, basalt-quarry. *C:* dolerite, Dmanisi, South-Caucasus, Georgia, SU. 1: weight percent, 2: mean diameter in micron, 3: quality distribution.

2. The dimensional distribution follows the isothermal isobaric surfaces of the ancient lava-flow (sheet, pipe): the size of Fe-Ti-oxide grains decreases towards the surface in many cases, e.g. in the well-preserved profile of the Kabhegy: at the top, the grains are so small as to escape measurement. Hence, dimensional distribution alone may indicate in a profile the ancient lava-surface, or, in the same manner, the levels of minimum temperature – or certain surfaces in the fluctuating lava of a thick sheet, or of a pipe. A good example is the occurrence of several grain size minima in the Uzsabánya profile.

3. The quality distribution of Fe-Ti-oxide minerals in the examined profiles shows the development of different crystalline phases from the melted lava in the same lava-flow (sheet, pipe) depending on redox and pt-relations: in the inner, deeper part of the lava-flow, where decrease of temperature was slowest, pressure of the lava was greatest and strong external oxidative effects were absent, there crystallized the least oxidized Fe-Ti-oxides: magnetite with segregations of ilmenite and ulvöspinel. Authorities disagree as to whether ilmenite or ulvöspinel crystallizes at a higher temperature; however, ulvöspinel is the more likely one. The segregation of ulvöspinel in basalt parallel to the octahedron of magnetite as compared to (100) in gabbro – can be the result of pt differences (similarly to the platy development of magnetite parallel to (111)). Author's investigations did not cover the question whether or not ilmenite can be a product of secondary oxidation of ulvöspinel (see Vincent et al. 1954). It is a more or less accepted fact that the composition of first-crystallized magnetite and ilmenite is far from ideal: magnetite contains more or less Ti, ilmenite can sometimes contain Fe^{3+} (ferri-ilmenite). Proceeding upward in the geological profile, the segregations of ulvöspinel disappear first, as the oxidative effect increases and the pt parameters decrease. After this the magnetite-ilmenite association is replaced rather abruptly by hematite containing segregated ilmenite. This sudden change indicates presumably a significant redox and pt boundary in the profile of the lava-flow; the interpretation of this phenomenon will require further detailed investigations on both ancient and recent lava-flows. Of the examined profiles, the Kabhegy is of this type as well as the Georgian dolerite profile, in which the individual phases are "telescoped" into each other owing to their thinness and to the fluctuation of the lava-flow. In other cases, as in the profiles of Mt. Gulács, Mt. Tóti and possibly Uzsabánya (the highest part of the basalt sheet was not examined in the present work), above the ulvöspinel-ilmenitic magnetite of the lower levels, titanomagnetite is observed in the upper, most oxidized level, where the decrease of temperature most rapid; the titanomagnetite may include segregated ilmenite; moreover the surface of grains may be more or less deeply altered into titanomaghemite. The presence of typically reductive ulvöspinel in some titanomagnetites contradicts this assumption: the elucidation of this problem will necessitate further research. The branching in two of the crystallization sequence after the magnetite-ilmenite phase of the lower and middle levels may be due either to differences in the Fe-Ti-ratio, and/or to differences in pt and redox relations and/or geochemical factors (e.g. chemical and mineralogical catalysts etc.). Already Vincent and Phillips (1954) pointed out, that different pt conditions tend to displace the equilibrium of the various $FeO - Fe_2O_3 - O_2$-systems.

Consequently according to the author's investigations, several facts prove that the Fe-Ti-oxide minerals of basaltic lava-flows crystallized partly or entirely after the uprising of the lava to the surface and crystallized as different mineral phases in different periods of solidification, depending on pt and redox-relations. Previous investigations have proved (1963) that the more rapidly cooled pyroclastics, crystalline bombs or lapilli, contain little opaque material of microscopic size, but many microlites (at the limit of microscopic visibility). This is why these and the vitreous basaltic rocks are darker of colour than the other basaltic rock-types. The larger grains may originate from the magma; the microlites are already the results of the rapid cooling on the surface.

Comparing the author's results and genetic conclusions with the relevant international literature justifies the following statements:

1. It is necessary to continue the investigations on the Transdanubian basalts and to extend them to the basalts of North-Hungary as well as to the best-preserved Hungarian andesites. The small grain size of opaque minerals in the eruptives is a great handicap for X-ray and chemical studies; this is why data concerning this point are scarce in the entire literature. Also, it is necessary to develop investigations using micro- and semi-microanalytical methods.

2. It is needed to subordinate further investigations even more to the geological viewpoint. However perfect the interpretation of the presence or genesis of a phase in a rock-sample may be, as long as a full understanding of the interrelations and genetic implications of the various phases in the Fe-Ti system requires by all means a profound consideration of the geologic setting.

3. The employment of electron microscopy could further the investigations, as has been proved by a few successful experiments; however, the author has not had the occasion to pursue this subject any farther.

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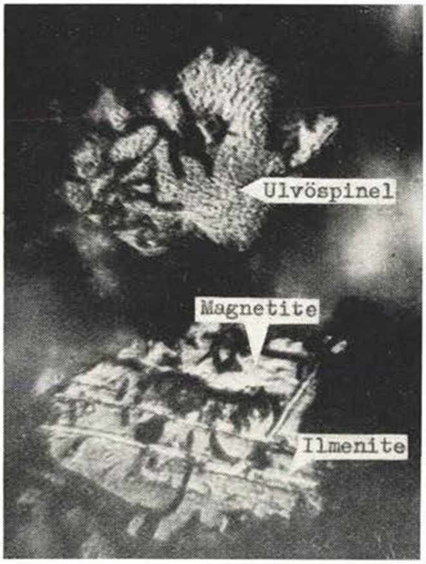

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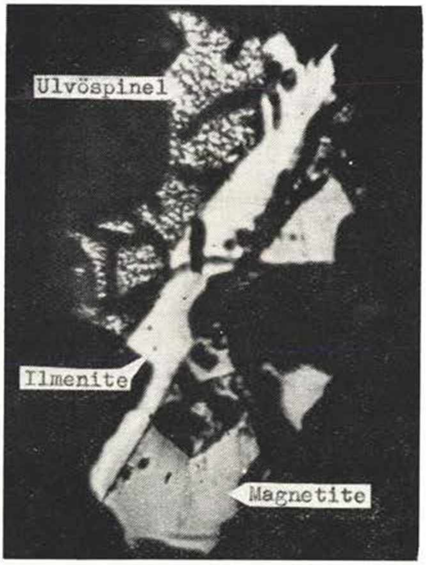

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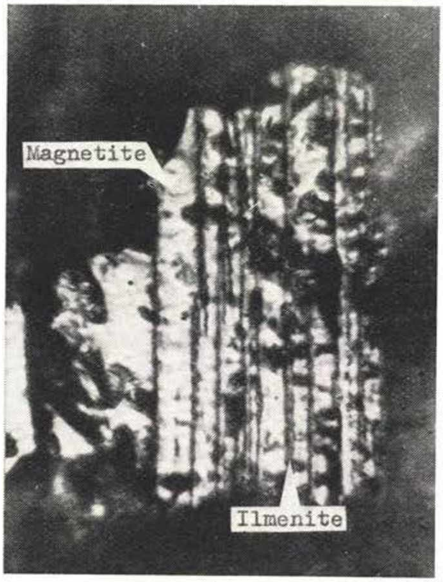

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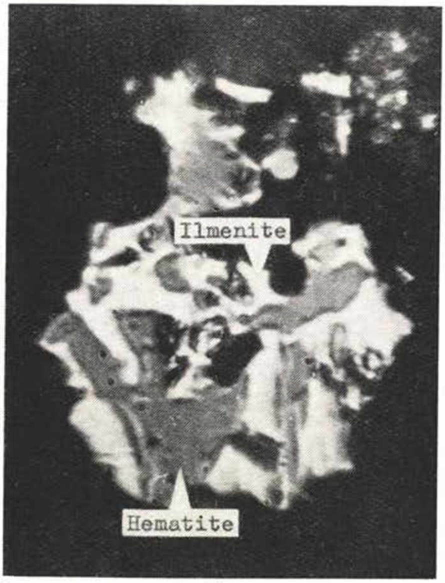


Plate I.

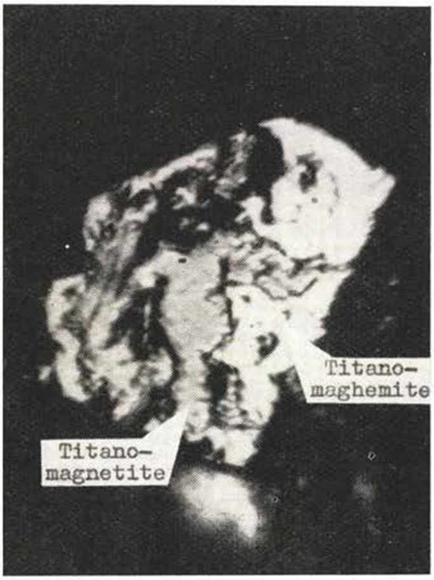
4.

Fig.1. Segregation of ulvöspinel and ilmenite in magnetite. Etching with ccHF. Size: 40 × 50 microns. Nicols//. – Mt. Gulács (sample G – 16)

Fig. 2. Segregation of ulvöspinel and ilmenite in magnetite. Etching with cc HF. Size: 30 × 60 microns. Nicols // – Uzsabánya (sample Uzsa – 71).

Fig. 3. Ilmenite segregation in magnetite, parallel to (100). Etching with cc HF. Size: 40 × 45 microns. Nicols //. – Mt. Gulács (sample G – 16).

Fig. 4. Hematite-(ferri-) ilmenite segregation. Size: 30 × 40 microns. Nicols //. – Kabhegy, upper lava-flow (drilling Öcs – 5).



1.

Plate II.
Fig. 1. Titanomaghemite on titanomagnetite. Etching with cc HF. Size: 30 × 40 microns. Nicols //.
— Mt. Tóti (sample T – 21).