

Intergrowths in ilmenite of the beach sands of Kerala

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[Taken as read 14 January 1965]

Summary. A detailed study of ilmenite concentrates from Chavara and Manavalakurichi deposits has brought to light several interesting textures traceable to exsolution, eutectic crystallization, and replacement and alteration. Magnetite-ilmenite, magnetite-ilmenite-picotite, ilmenite-picotite, ilmenite-hematite, and ilmenite-rutile exhibit exsolution intergrowths. Part of the ilmenite seems to have been formed either by oxidation of ulvöspinel or from a cubic phase of ilmenite by monotropic inversion. Some of the ilmenite-hematite intergrowths display either an emulsoid or a triangular type of texture. Subgraphic intergrowth is observed between ilmenite-magnetite and ilmenite-rutile, suggesting an eutectic relationship between them.

Magnetite-hematite, magnetite-goethite, and magnetite-maghemite intergrowths are the result of alteration of magnetite. Sphene also partly replaces some ilmenite grains.

The proportion of ilmenite grains with and without intergrowths and the probable origin of the intergrowths and their effect on the marketability of ilmenite concentrates are discussed in the paper.

INDIA is one of the largest producers of ilmenite sand and the beach sand deposits of Kerala are world-famous for their high heavy-mineral content. With a view to determining the constituents responsible for the deleterious ingredients, especially chromium, a microscopic examination of the ilmenite concentrate has been made. Polished sections of ilmenite from Chavara and Manavalakurichi have been prepared, and their study has brought to light several interesting textures, which are described below.

The following minerals are present in intricate textural relationships: ilmenite, magnetite, hematite, rutile, picotite (chrome spinel), goethite, maghemite, sphene, and leucoxene. Three different types of textures are observed, which are due to exsolution, eutectic crystallization, and replacement and alteration. The exsolution textures are the most important among these.

Exsolution textures

Magnetite-ilmenite-spinel and magnetite-ilmenite. Ilmenite lamellae occur in the octahedral planes of magnetite forming the characteristic triangular texture. All transitions exist from minute to broad lamellae

extending from one end to the other of the grains. In addition to the normal octahedral lamellae of ilmenite, there are in some cases two other sets of ilmenite lamellae, occupying the dodecahedral planes of magnetite, and not infrequently parallel to the cubic planes (fig. 1). Even here all transitions from minute to large lamellae occur. Most of the magnetite-ilmenite intergrowths also contain two sets of mutually

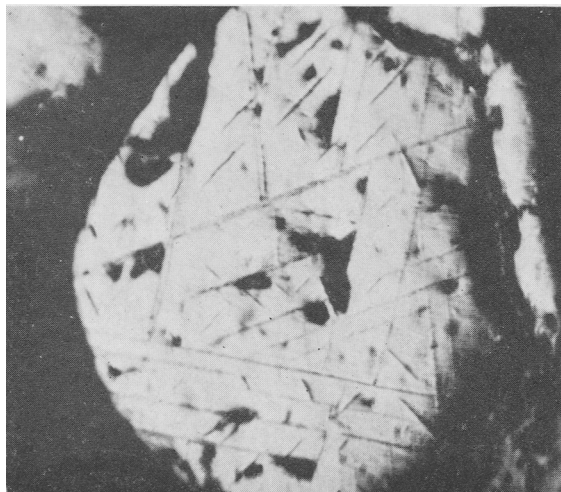
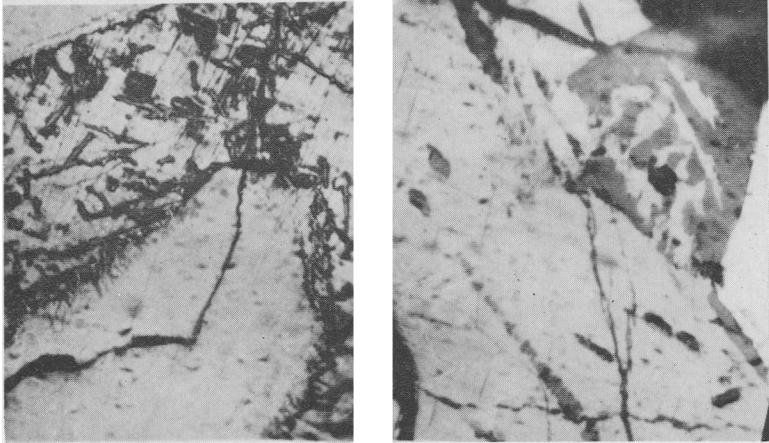


FIG. 1. Exsolution intergrowth of ilmenite lamellae (large medium-grey lamellae) and spinel (small medium-grey lamellae) in magnetite (light grey). $\times 200$.

perpendicular spikes of a transparent spinel, oriented parallel to the cubic planes of magnetite. The spinel spikes invariably exhibit a slight vermicular distortion. Spinel spikes parallel to the octahedral planes of magnetite are also not uncommon, in which case the ilmenite lamellae occur parallel to them, forming an intricate triangular texture. Most of the ilmenite lamellae also contain myriads of minute spinel granules, generally forming a beaded edge at the junction of magnetite and ilmenite lamellae. Broad ilmenite lamellae with two or three such rows of spinel granules are not uncommon.

Occasionally, the ilmenite lamellae themselves are irregular, localized by grain boundaries and cracks. Minute subhedral growth of ilmenite often joins two or more lamellae of ilmenite; sometimes from a single lamella such growth takes place on either side into magnetite; in all these cases, however, a crystallographic control of the growth is evident,

being guided by the octahedral planes of magnetite. When localized by cracks, the ilmenite resembles replacement veins, but no other characteristic suggestive of replacement is observed. In one case, where the ilmenite is localized by a crack in magnetite, the spinel spikes project from the ilmenite boundary into magnetite, almost at right angles. Most, if not all, of these irregular lamellae of ilmenite have a stronger



FIGS. 2 and 3. FIG. 2 (left). Granular intergrowth of ilmenite (medium grey) in magnetite (light grey, etched). At the junction of the two is the myrmekitic growth of spinel (dark grey). $\times 300$. FIG. 3 (right). Pseudoeutectic intergrowth of ilmenite (light grey) and spinel (dark grey) at the grain boundary of magnetite (medium grey). $\times 300$.

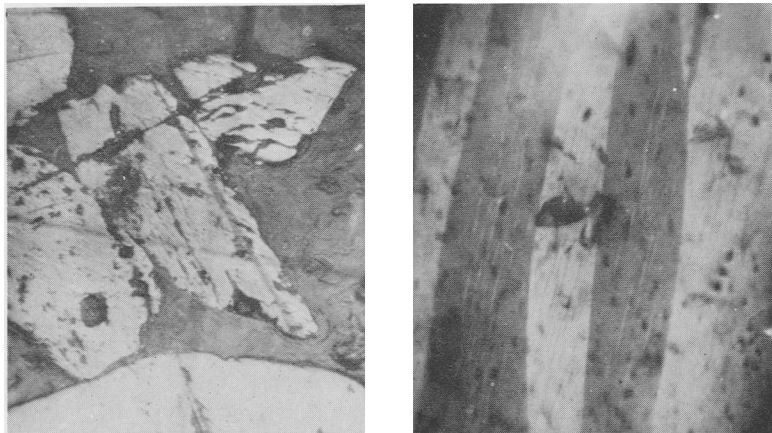
reflection pleochroism and a sharp anisotropism, unlike the regular lamellae described earlier, and, in general, are devoid of the granules of the transparent spinel.

Granular intergrowth between magnetite and ilmenite is also frequently present, and in all such associations the junction between the two is smooth and gently curving and is of the type called a sealed boundary (Vincent and Phillips, 1954). The grain contact between the two is frequently characterized by either a pseudoeutectic (fig. 3) or a myrmekitic (fig. 2) intergrowth of spinel in ilmenite; the spinel is invariably confined to the ilmenite side of the contact. Rarely, grain boundaries of magnetite are characterized by such ilmenite-spinel graphic intergrowth.

The spinel has a reflectivity slightly higher (nearly 8 % in air in green

light) than the other associated transparent minerals like quartz. It is dark, bluish grey in colour, and is negative to all standard etching reagents including HF. It is thought to be the chrome spinel, picotite, since it contains chromium in its composition.

Ilmenite-spinel. A few ilmenite grains contain oriented lamellae of spinel (picotite), occurring parallel to the 0001 planes of ilmenite



FIGS. 4 and 5. FIG. 4 (left). Exsolution lamellae of spinel (medium grey in the central grain) in ilmenite (light grey). $\times 150$. FIG. 5 (right). Seriate, bladed exsolution intergrowth between ilmenite (dark grey) and hematite (light grey). Oil immersion. $\times 800$.

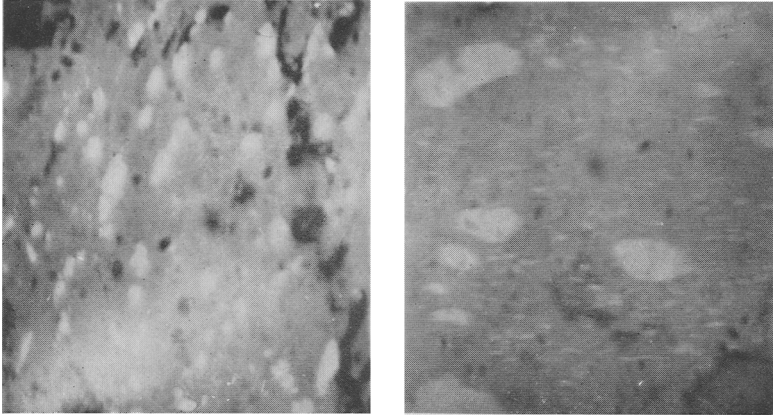
(fig. 4). The spinel has a higher reflectivity than that occurring elsewhere, and appears to be nearer to chromite in composition.

Ilmenite-hematite. In typical cases of this intergrowth, elongated exsolution bodies of hematite occur in ilmenite, with their long axes parallel to each other and to the [0001] direction of ilmenite. An ordered or seriate arrangement of the exsolution bodies is common, in which case broad blades of ilmenite and hematite alternate with each other, each containing smaller bodies of the other, and all elongated parallel to each other (fig. 5).

In addition, many other patterns of intergrowth between ilmenite and hematite are seen. In some, the hematite exsolution bodies are nearly rounded or oval, but all arranged in linear rows (fig. 6); in others, large rounded blebs of hematite bodies are surrounded by numerous smaller ones (fig. 7); while in yet others the hematite blebs are irregular,

of all sizes and distributed irregularly throughout the ilmenite base. All transitions from these emulsion or mottled textures to the typical bladed type are seen.

In one grain at a very high magnification ($\times 1250$), a very interesting type of intergrowth of hematite in ilmenite is seen. The hematite occurs as fine lamellae forming a typical triangular texture, with another series



FIGS. 6 and 7. FIG. 6 (left). Emulsoid exsolution intergrowth of hematite (light grey) in ilmenite (dark grey). Oil immersion, $\times 800$. FIG. 7 (right). Seriate emulsoid exsolution intergrowth of hematite (light grey) in ilmenite (dark grey). Oil immersion, $\times 800$.

of lamellae, slightly irregular, overlapping the former. The latter lamellae appear to be elongated parallel to the [0001] direction of ilmenite. The net result is a fine reticulate type of intergrowth. At first sight the lamellae appear to be that of rutile, but they lack the characteristic anisotropism and polarization colours of rutile; they are faintly anisotropic and have a much higher reflectivity than ilmenite. The ilmenite grain as a whole is itself strongly anisotropic. The hematite constitutes nearly 40 % of the area of the grain.

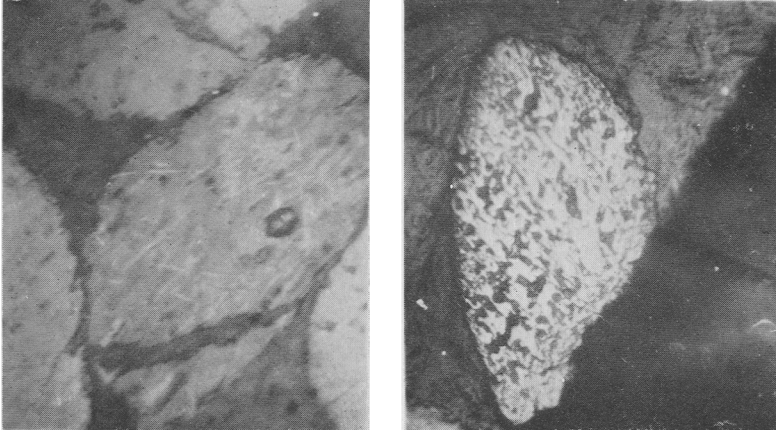
Ilmenite-hematite-rutile. In grains with seriate lamellar ilmenite-hematite intergrowth, the ilmenite base, in places, contains fine needles of rutile crisscrossing, mostly near the junction of hematite blades with ilmenite. The rutile needles occur in the rhombohedral planes of ilmenite, making an angle of nearly 60° with the direction of elongation of the hematite exsolution bodies.

Ilmenite-rutile. A few ilmenite grains that are free from hematite

exsolution bodies contain blades of rutile occupying the rhombohedral planes of ilmenite and forming a Widmanstätten-type pattern (fig. 8).

Eutectic textures

Magnetite-ilmenite. One grain showed an interesting eutectic intergrowth of magnetite and ilmenite. The intergrowth is of graphic type,



FIGS. 8 and 9. FIG. 8 (left). Exsolution lamellae of rutile (light grey) in the rhombohedral planes of ilmenite (dark grey). $\times 150$. FIG. 9 (right). Eutectic intergrowth of ilmenite (light grey) and magnetite (dark, etched with HCl). $\times 150$.

and is so intimate that only when magnetite is etched with HCl does it become clear (fig. 9). It is quite unlike the granular magnetite-ilmenite intergrowth described above, in which well-defined grains of ilmenite could easily be distinguished. The intimacy of the intergrowth is such that it could result only from a simultaneous crystallization at the eutectic temperature.

Rutile-ilmenite. A few rutile grains contain subgraphic intergrowths of ilmenite suggesting an eutectic relationship between the two.

Replacement and alteration textures

Ilmenite-sphene-rutile. Crystals of sphene associated with rutile occur intergrown in some of the ilmenite grains. Rutile and sphene may form discrete crystals occurring adjacent to each other, or may have an apparent granular intergrowth relationship between them. The direction of elongation of the crystals is either parallel or perpendicular to

each other, but apparently bears no relationship to crystallographic directions in the ilmenite. The rutile crystals show clearly their characteristic twinning, the twin plane being normal to their direction of elongation.

Magnetite-hematite. Martitization of magnetites along their octahedral planes has resulted in the formation of a Widmanstätten-type texture consisting of lamellae of hematite elongated parallel to their [0001] direction and the [111] direction of the magnetite. The concentration of hematite at the grain boundaries and adjacent to cracks indicates replacement origin of the texture. However, in a few grains the hematite blades appear to be distributed uniformly and also form border zones to ilmenite lamellae, and as such the possibility of an exsolution origin of part of the hematite cannot be excluded.

Magnetite-goethite and magnetite-maghemite. Goethite forms irregular veins traversing magnetite grains, and they end abruptly at the junctions of ilmenite lamellae. It is typically dark bluish-grey in colour, porous, and has red internal reflection. Hematite is also seen partly altering to goethite, and not infrequently the whole of a magnetite grain is replaced by hematite and goethite, in which case the ilmenite lamellae are sharply defined. The occurrence of maghemite is very rare. It is bluish-grey in colour, reflecting slightly higher than magnetite, but much higher than ilmenite, and clearly has a replacement relationship to magnetite. It is seen associated with goethite veins, or bordering ilmenite lamellae, or as segregated patches and veins in magnetite.

Ilmenite-leucoxene. Ilmenite is seen to alter along grain boundaries or fractures or as patches into leucoxene. All the three stages of alteration recognized by Bailey *et al.* (1956) have been observed. The colour of the leucoxene is variable—white, brown, and red. A detailed study of the alteration is in progress and will be described elsewhere.

Discussion: Exsolution

The nature of the various intergrowths described under this head indicates that they were formed as a result of exsolution from homogeneous solid solutions that existed at higher temperatures. The criteria for recognition of exsolution textures are summarized by Edwards (1954) and Ingerson (1955). Thus the crystallographic control of the formation of the intergrowths, absence of enlargements where the blades or lamellae cross each other, as well as lack, in general, of relationship of included minerals to grain boundaries and mineral contacts, are all indicative of exsolution.

The homogeneous solid solutions occur as a result of substitution of a cation in the structure by another cation of similar valency, ionic size, and chemical nature (Shaw, 1953). The latter concept includes the electronic structure, ionization potential, and electronegativity of the cations.

Magnetite-ilmenite. In magnetite, Fe_3O_4 , which has a spinel structure Ti^{4+} (0.68 Å) can replace Fe^{3+} (0.64 Å) positions. However, Fe^{3+} and Ti^{4+} diadochy is not extensive at low temperatures. The fall in temperature therefore, results in the exsolution of ilmenite giving rise to the characteristic Widmanstätten texture, in which the ilmenite lamellae, elongated parallel to (0001) planes, occupy the octahedral, or sometimes cubic or dodecahedral planes of magnetite. However, the exact nature of solid solution and exsolution does not appear to be so simple, as heating of magnetite-ilmenite intergrowths has failed to homogenize even at 1300° C (Vincent *et al.* 1957; Wright, 1959).

Extensive solid solution is known to occur between magnetite and ulvöspinel, on the other hand, solid solution between magnetite and ilmenite is very limited (Hier, 1956; Verhoogen, 1962). Though ulvöspinel itself has not been observed in the present case, the occurrence of minute lamellae of ilmenite observable only under high magnifications ($\times 1000$), as well as the occurrence of ilmenite lamellae occupying the (100) and (110) planes of magnetite indicate the probable existence of this phase (Ramdohr, 1953), which has subsequently been oxidized to ilmenite. The irregularity of some of the ilmenite lamellae as well as the growth of ilmenite linking adjacent lamellae may be the result of such oxidation (see Vincent, 1960). It may be mentioned here that some heating experiments have shown that magnetite-ilmenite intergrowths could be homogenized only when ilmenite is reduced to ulvöspinel (Wright, 1959).

The other feature that gives some indication as to the nature of magnetite-ilmenite exsolution is the manner of association of the transparent spinel with ilmenite grains and lamellae intergrown in magnetite. The occurrence of minute spinel granules distributed in ilmenite lamellae, as well as forming a beaded edge to the same, has also been observed elsewhere (Dunn, 1937; Vincent *et al.*, 1957). In addition a pseudo-eutectic and myrmekitic relationship has been observed between ilmenite and spinel in the present case. Mention may be made that the occurrence of spinel spikes enclosed within ulvöspinel 'boxes' in some magnetite-ulvöspinel intergrowths observed elsewhere has been attributed by Ramdohr (1953) to exsolution. A similar solid-solution

relationship might have existed in this case also, which would explain the ilmenite-spinel relationship. The intergrowth is not the crystallographic oriented type, but appears to have been produced due to exsolution from, and segregation in, a phase now changed to ilmenite, as a result of diffusion out of the now unfavourable structure to the grain and lamella boundaries. The changes that occurred may be either an oxidation of ulvöspinel or a monotropic inversion from a cubic phase of ilmenite (Nichollas, 1955; Vincent *et al.*, 1957), the result of both of which is the formation of rhombohedral ilmenite. The absence of crystallographically oriented spinel, and its segregation at the grain boundaries of rhombohedral ilmenite indicate that the unmixing took place at a comparatively high temperature (Edwards, 1954).

Ilmenite-spinel. The crystallographically oriented spinel lamellae occurring in some of the ilmenite grains are probably the result of exsolution of ilmenite-spinel (picotite) solid solution. The latter can be explained as due to replacement of some of the Ti^{4+} ions in the ilmenite structure by Cr^{3+} (0.63 Å), Al^{3+} (0.51 Å), and Fe^{3+} (0.64 Å) and of Fe^{2+} (0.74 Å) by Mg^{2+} (0.66 Å) ions. Electrical neutrality may be achieved by the appearance of vacant cation sites or by the entering of some of the trivalent cations into Fe^{2+} ionic position. Exsolution due to the lowering of temperature would result in the formation of picotite, $(Mg,Fe)(Al,Fe,Cr)_2O_4$ elongated in [111] directions, and oriented parallel to the [0001] planes of ilmenite, for in these planes the oxygen positions practically coincide in spinel and ilmenite structures (Edwards, 1954). It may be mentioned here that a limited solid solution between ilmenite and chromite has been postulated to explain the lamellae of chromite in ilmenite in the chromite ores of Nausahi, Bihar (Mukherjee, 1961).

Ilmenite-hematite. The solid-solution phenomenon between ilmenite and hematite is well known. Ilmenite and hematite have similar structures of the corundum type (Berry and Mason, 1959). Extensive diadochy takes place between Fe^{3+} and Ti^{4+} ions. Homogeneous solid solutions exist only above about 600° C (Edwards, 1954; Ingerson, 1955). Below this temperature, it unmixes into a ferriferous ilmenite and a titaniferous hematite. The exsolution bodies, hematite in the ilmenite base and ilmenite in the hematite base, are elongated parallel to the (0001) plane, in which plane the oxygen ions have similar spacings.

Since the exsolution bodies grow by diffusion, the earlier formed ones are comparatively large and the later ones smaller, giving rise to a regular seriate texture. Slow diffusion during the low-temperature stage of

exsolution results in the formation of small exsolution bodies surrounding a few larger ones (fig. 7), which were formed earlier when diffusion was fast. The emulsion type of texture is probably the result of rapid cooling of the ilmenite-hematite solid solution. The irregularity of some exsolution bodies indicates that they have grown *in situ*, by absorbing exsolving hematite from the adjacent ferri-ilmenite.

The triangular texture observed in one grain between ilmenite and hematite is interesting. In fineness and arrangement it resembles a magnetite-ulvöspinel eutectoid intergrowth of the 'cloth-woven pattern' observed elsewhere (Vincent and Phillips, 1954). The possible existence of a γ -FeTiO₃- γ -Fe₂O₃ solid solution (Vincent *et al.*, 1957) can explain the occurrence of this type of texture. The lamellae forming the triangular pattern are probably exsolved before the formation of the rhombohedral ilmenite; the lamellae cutting across the former are exsolved subsequent to the inversion of the cubic phase to the rhombohedral phase.

Ilmenite-rutile. The rutile occurring in the rhombohedral planes of ferri-ilmenite evidently represents the excess of TiO₂ present at this stage, over that required to form the FeTiO₃-Fe₂O₃ system. The existence of a solid-solution series between ilmenite and rutile is also inferred by the presence of rutile lamellae confined to the rhombohedral planes of a few ilmenite grains (which are free from hematite exsolution bodies). The solid solution must be very limited as very few rutile lamellae are observed. Further, it is confined only to the ilmenite end of the ilmenite-rutile binary series, as none of the several hundred grains of rutile examined showed any exsolved lamellae of ilmenite; on the other hand, a few of them showed eutectic relationship with ilmenite. The limited nature of the solid solution can be attributed to structural reasons.

Eutectic textures

Magnetite-ilmenite. At a lower temperature the magnetite-ilmenite relationship is an eutectic one, and according to Malyshev (in Evrard, 1949) the eutectic composition lies between 20 and 30 % of ilmenite, though the presence of such oxides as Cr₂O₃, Al₂O₃, MgO, &c. may considerably influence the eutectic point (Evrard, 1949). The grains showing the eutectic intergrowth of ilmenite-magnetite may possibly be related to the titanian magnetites described earlier. For, as Evrard has pointed out, in a magma poor in Ti, magnetite-ilmenite may reach an eutectic point on progressive crystallization, with the lowering

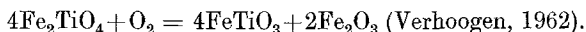
temperature, by the progressive separation of titanian magnetite, with the consequent enrichment of TiO_2 relative to $\text{FeO} + \text{Fe}_2\text{O}_3$.

Rutile-ilmenite. As already pointed out, the subgraphic intergrowth of ilmenite in a few rutile grains indicates an eutectic crystallization of the two.

Replacement textures

It is known that during the main stage of crystallization sphene may replace partly some ilmenite grains (Rankama and Sahama, 1950) and it is interesting that rutile is generally seen associated with sphene.

The replacement of magnetite by hematite, goethite, and maghemite is the result of alteration of magnetite. The occurrence of hematite blades bordering some of the ilmenite lamellae observed by Dunn (1937) elsewhere was thought by him to be due to the presence of excess Fe_2O_3 in the system magnetite-ilmenite. The available evidence in this case leads the authors to conclude that they are simply the result of martitization along octahedral planes of magnetite, which were already occupied by the exsolution lamellae of ilmenite. This is further supported by the fact, that, not uncommonly, such hematite blades are connected to hematite concentrations at the grain boundaries and cracks that are evidently the result of martitization. Mention should be made, however, that they could be also the result of oxidation of ulvöspinel to ilmenite at high temperatures according to the equation



The question arises whether the grains showing the two important types of exsolution intergrowths, namely the magnetite-ilmenite and ilmenite-hematite intergrowths, are genetically related or are derived from different sources. It should be emphasized here that in the case of placer deposits it is generally difficult to find with certainty the relationship of the different minerals and textures. The difficulty is all the more augmented when the source rocks are varied. The hinterland of the Kerala beach deposits consists of both igneous and metamorphic rocks with a very wide range of composition.

Evrad (1949), from a study of a number of analyses of titaniferous ores, differentiates a titanian magnetite with a TiO_2 molar % of less than 30 and an ilmenite of molar % above 30. In the former the evolution of the composition is governed by the magnetite-ilmenite binary series, while the latter follows the ilmenite-hematite binary system.

Only around 30 molar % of TiO_2 do both the types coexist. This observation holds good, according to him, not only for titaniferous ores, but also for opaque minerals included in eruptive rocks. Should the titanian magnetite and the ferri-ilmenite, in the present case, be derived from the same source, the number of grains of titanian magnetite expected would be much more than that observed. Further, the probable presence of ulvöspinel in this deposit together with ferri-ilmenite rules out the possibility of a common source for these two minerals, since ulvöspinel cannot exist in the presence of free Fe_2O_3 (Rankama and Sahama, 1954).

The grains having the various types of intergrowths described (except the ilmenite-leucoxene alteration) form but a small fraction of the ilmenite concentrates, and the majority of the ilmenite grains are pure, devoid of any type of intergrowths. In spite of this the following general conclusions may be drawn regarding the source rocks, which may be classified as: Those from which were derived the titanian magnetites and magnetite-ilmenite eutectic intergrowths; those that contributed the ferri-ilmenite grains; those that provided the bulk of pure ilmenite grains; and those that were the source of the bulk of the rutile, the ilmenite-rutile exsolution intergrowths, and the rutile-ilmenite eutectic intergrowths.

Source of Cr_2O_3 . Part of the chromium in the ilmenite concentrates was contributed by free chromite grains (unpublished report), and another part is derived from the picotite intergrowths in ilmenite and titanian magnetites. Another probable source is unexsolved chromium occurring in the ilmenite structure. A comparison of the characters of Fe^{3+} , Ti^{4+} and Cr^{3+} ions would show that Cr^{3+} ions can diadochically replace either Ti^{4+} or Fe^{3+} . Chromium in igneous rocks is strongly enriched in the early crystallates, and when present in trace amounts it is always camouflaged by Fe^{3+} (Ringwood, 1955). When Ti^{4+} is also present, Ti can still easily camouflage Cr because of the closeness of their electro-negativity and ionization potential. It should be expected, therefore, that Cr would be more concentrated in titanomagnetites and early crystallized ilmenites, rather than the bulk of the ilmenite grains, which were formed later.

Conclusion

The grains of ilmenite that have intergrowths form an insignificant minority of the ilmenite concentrates. Among the textures the most prominent ones are the intergrowths with magnetite and hematite. The diverse types of intergrowths and associations point to a variety of source rocks. Even though the features studied are mostly of academic

interest, they may be useful in pointing out the sources of some deleterious ingredients, especially chromium, in the final ilmenite concentrates.

Acknowledgements. The authors' thanks are due to Dr. K. K. Majumdar, Head, Ore Dressing Section, A.E.E.T. for his sustained interest in the work presented herein, and to Dr. Brahm Prakash, Director, Metallurgy Group, A.E.E.T., for his kind permission to publish this paper.

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[*Manuscript received 22 November 1963.*]
