

LIQUID ILMENITE OR LIQUIDUS ILMENITE: A COMMENT ON THE NATURE OF ILMENITE VEIN DEPOSITS

J. C. DUCHESNE

Laboratoires associés de Géologie, Pétrologie et Géochimie, Université de Liège, B 4000 SART TILMAN, Belgium

ABSTRACT

The interstitial character of ilmenite in common gabbroidic rocks has led to the idea that the last drop of liquid to crystallize in a rock could be pure ilmenite. When injected into enclosing rocks this liquid could give rise to ilmenite-pure veins. Fractional crystallization or immiscibility cannot produce a pure ilmenite melt. Actually, ilmenite is a liquidus mineral, which is the first or among the first mafic minerals to crystallize in Fe-Ti rich magmas. This is clearly shown by the sequence of crystallization in the Bjerkreim-Sokndal layered intrusion, by quantitative modelling of the liquid line of descent of jotunitic (hypersthene monzodioritic) magmas, and by experimental work. The interstitial character of ilmenite is therefore acquired subsolidus. Deformation can enhance the process. The synemplacement deformation linked to the diapiric uprising of anorthosite massifs is a suitable environment for the formation of layers of Cr- and Mg-rich ilmenite and their subsequent deformation and mobilization in discordant veins of pure ilmenite.

1. Introduction

Massive anorthosites are hosting the most important ilmenite deposits in the world. Recent advances in the petrology of anorthosites (see Ashwal, 1993; Wiebe, 1992) have notably improved the knowledge of the mechanism of intrusion and of the nature of the parent magmas. Deformation during diapiric emplacement of a plagioclase crystal mush is now generally admitted as a common mechanism. Fe and Ti rich magmas, such as the jotunitic (hypersthene monzodiorites, monzonorites), are now widely recognized as intimately linked to the anorthosite formation. It is therefore interesting to investigate or to reconsider the origin of the ilmenite deposits in this new tectonic and magmatic framework. A synthesis of the problematic of the Rogaland deposits has been published recently (Duchesne, in press). The present paper focuses on a well-known feature of ilmenite in common rocks (its interstitial character) and discusses the origin of some ilmenite orebodies.

2. Interstitial character of ilmenite

Perhaps the most enigmatic character of the Fe-Ti oxide deposits is the occurrence of veins of pure ilmenite in massive anorthosite, as is the case with the Lac Tio deposit in Quebec (Hargraves, 1962) or the Jerneld deposit in Rogaland (Hubaux, 1960; Duchesne, 1973).

This character, together with the observation that ilmenite is generally interstitial in plutonic rocks, as for instance in the Bjerkreim-Sokndal cumulates (Michot, 1965; Duchesne, 1970, Duchesne, 1972) or in jotunitic dyke rocks (Duchesne et al., 1985; Michot, 1965), have forced people to consider that the Fe-Ti oxides were the last minerals to crystallize, in other words, that the last drop of liquid to crystallize should be pure ilmenite.

This character of ilmenite is often used to identify protoclastic structure: "structure qui traduit l'action des déformations sur les agrégats cristallins encore imprégnés de liquide magmatique résiduel" (Michot, 1965, p. 970). Ilmenite is considered to be able to inject into fractures in broken or granulated crystals.

Bateman (1951) has forged a mechanism to explain the late-magmatic injection of oxide minerals. It was called late-gravitational liquid enrichment. According to that author, crystallization of silicate minerals enriches the residual liquid in Fe and Ti. This residual liquid percolates down due to its high density and accumulates in the lower part of the system. A slight deformation can squeeze it out of the crystal framework to form late-magmatic injection in the enclosing rock.

3. Liquid ilmenite?

There are however a number of difficulties in accepting this concept of a pure ilmenite late-liquid. It is well known from phase diagrams that in a crystallization process the number of minerals coexisting with the liquid increases as the crystallization proceeds. Therefore one should expect a multi-saturated liquid at the end of the

Postprint (Author's version)

crystallization and not a liquid crystallizing a single phase such as ilmenite.

Excellent examples of interstitial crystallization are provided by orthocumulates (Wager and Brown, 1968). In Rogaland, the best case was described by DemaiFFE and Hertogen (1981) in the Hidra anorthosite, where a granophyric assemblage made up of plagioclase, K-feldspar, quartz and some mafic minerals develops between euhedral plagioclases.

Since a late stage liquid made up of pure ilmenite cannot be produced by fractional crystallization, we are justified to examine immiscibility as an alternative mechanism. Philpotts (1981) has proposed that evolved magmas related to anorthosites, enriched in Fe and Ti by previous crystallization dominated by plagioclase, could enter an immiscibility field and split into a silica-rich liquid (mangerites) and an Fe-Ti-rich liquid (orebodies). These ideas were supported by several experimental data (see Roedder, 1979).

However, field evidence in Rogaland has not validated this process. Several cases in which immiscibility was expected failed to give supporting evidence. Firstly, liquids represented by fine-grained dyke rocks -such as the ferrodiorites of the Vettaland dyke (Duchesne et al., 1985)- have the adequate composition to produce immiscibility: their compositions plot right within the immiscibility field (Fig. 1), but the rocks remain homogeneous.

Secondly, pairs of rocks intimately associated in the field and with compositions close to the immiscibility field have been studied in the Bjerkreim-Sokndal intrusion (Duchesne et al., 1987). They belong to the transition zone between the Layered series and the acidic upper part of the massif: in the region of Örsland, layers of mangerites are interleaved with layers of ultramafic rocks made up of titanomagnetite, ilmenite, olivine and clinopyroxene. Partitioning of trace elements such as P, REE and Ba between the two rock types is however not at all consistent with experimental data on the partitioning between immiscible liquid (Watson, 1976) nor with theoretical considerations on the structure of the liquids (Ryerson and Hess, 1978). It has therefore been concluded that these rocks were not liquids, but cumulates (Duchesne et al., 1987).

Thirdly, another negative argument against an immiscibility process can be found in the composition of cumulate rocks from the Bjerkreim-Sokndal intrusion. Cumulus rocks from the Layered series systematically display in Harker diagrams two linear trends, each one connecting a plagioclase component to a mafic component (Fig. 2). The first mafic component is made up of orthopyroxene and hemoilmenite and characterises the anorthosite and leuconorite of macrocyclic unit II (MCUII), the lower part of MCU III and IV (Wilson et al., 1996), the so-called leuconoritic phase of Duchesne (1978). The second mafic component comprizes ortho- and clinopyroxene, ilmenite, magnetite and apatite and suits the clinopyroxene norites of the upper part of MCU III and IV. Cumulate rocks thus appear as mixtures of plagioclase and mafics. The interesting feature being that each mafic components have a constant composition. This feature is somewhat unexpected in cumulate rocks where sorting processes between cumulus minerals are usually considered to be active during accumulation (Duchesne, work in progress). It is obvious here that the oxides always remain together with the rest of the mafics. Such a behaviour precludes the existence of an oxide liquid which would have concentrated, due to its high density, and possibly segregated as conformable layers or cross-cutting veins. No structure of that type has ever been observed in this intrusion.

From an experimental point of view, the existence of ilmenite-rich liquids has also received severe criticisms. The temperature (around 1400°C) of the apatite-bearing immiscible liquids of Philpotts (1967) is outside the magmatic range. In these experiments, the role of apatite as a fluxing agent has been questioned by Lindsley et al. (1988) but carbon has been considered as a possible candidate (Weidner, 1982).

On the other hand, interesting results have been obtained by Lindsley (1991) suggesting that leucotroctolite can produce two conjugate rocks, one rich in oxide and the oilier one similar to jotunite. The liquid character of the oxide-rich material is however difficult to assess. Microtextural criteria are the only ones that have been used, and attempts to sink a small platinum ball in the oxide-rich phase have failed (Lindsley, oral comm.).

Whatever the final issue of these experiments, it should however be recalled that a 100% ilmenite-pure liquid cannot be obtained by immiscibility. The shape of the immiscibility field is such that, at the blocking temperature of the process, the ilmenite-enriched liquid will always contain a certain proportion of the other liquid (see e.g. Fig.1).

Postprint (Author's version)

Fig.1 Triangular diagram showing the immiscibility field of Roedder (1979) (dotted) and Firestone (1978) (dashed). Ferrodioritic rocks (inverted triangle) from the Vettaland dyke (Duchesne et al., 1985) plot inside the immiscibility field.

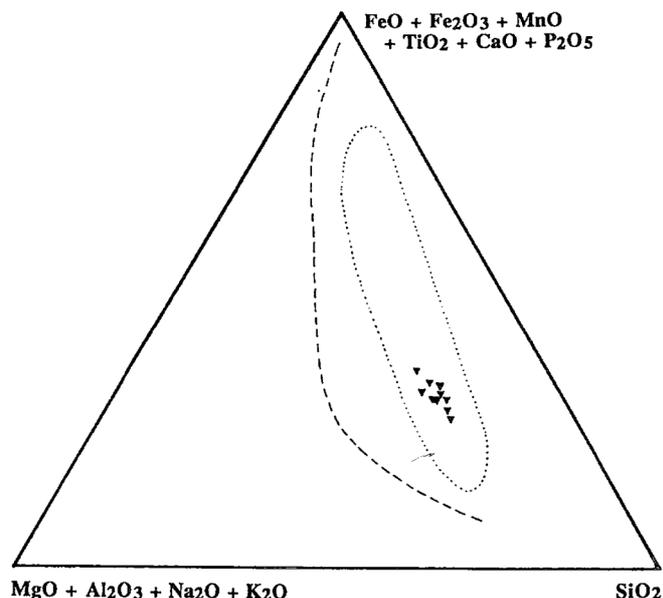
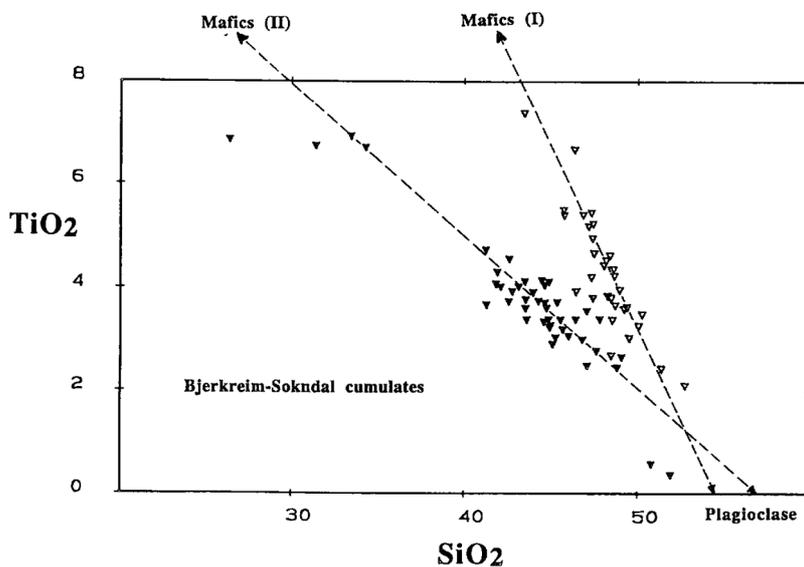


Fig. 2 Harker diagram (TiO_2 vs SiO_2) showing the composition of cumulate rocks from the Layered Series of the Bjerkreim-Sokndal layered intrusion (Duchesne, work in progress). Two trends can be defined by tie lines connecting a plagioclase pole to a mafic pole. Legend: Open triangle: anorthosite and leuconorite from the leuconoritic phase; dark triangle: clinopyroxene norites and leuconorites.



4. Liquidus ilmenite

Since neither fractional crystallization nor immiscibility are able to produce an ilmenite-pure liquid, what is the status of ilmenite in gabbroic and anorthositic rocks and particularly in the cumulus rocks of the Bjerkreim-Sokndal intrusion?

The answer is straightforward: ilmenite has the same status as the other minerals: it is a liquidus mineral. Several lines of evidence support this view:

In the Bjerkreim-Sokndal layered intrusion, which displays a cumulus mineral sequence, refined mapping of the

Postprint (Author's version)

Bjerkreim lobe by Robins and his group (Wilson et al., 1996) has clearly shown that besides plagioclase which is the first mineral to crystallize in the sequence at the base of the intrusion, ilmenite is the first mafic mineral to appear, slightly before hypersthene (Fig. 3).

In the series of jotunitic liquids which form dykes and small intrusions in massive anorthosite, major element geochemistry has revealed a continuous decrease of TiO₂ with the evolution towards high SiO₂ contents (Duchesne et al., 1989). This behaviour is explained by continuous subtraction of a cumulus assemblage containing ilmenite. Wilmart et al. (1989) have calculated by least square modelling that to evolve from a jotunitite to a mangerite, the subtracted cumulate must contain 43% An₃₈ + 20% En₅₆ + 9% Wo₄₄En₃₇Fs₁₉ + 11% Magnetite (5% TiO₂) + 9% Ilm₉₆Hem₄. The composition of the minerals was taken from cumulates in the Bjerkreim-Sokndal intrusion and the Fe# (FeO/FeO + MgO) of the orthopyroxene was constrained by using the Fe# of the olivine obtained by the calculation of Ford et al. (1983).

Experimental data also confirm the early appearance of ilmenite. (Vander Auwera and Longhi, 1994) have used a fine-grained chilled jotunitite from Tjörn to determine the liquidus minerals in various conditions of total pressure and fugacity of oxygen. In the range of fO₂ normally found in these rocks, the Fe-Ti oxides and particularly ilmenite are the first or among the first mafics to crystallize (Table 1).

5. Solid stage processes

Though ilmenite is a liquidus mineral in the cumulates of the Bjerkreim-Sokndal intrusion it does not display an euhedral shape but is interstitial to the other minerals. It must therefore have acquired this character after accumulation of the minerals.

Several lines of evidence indicate that this cannot be due to intercumulus crystallization because most Bjerkreim-Sokndal cumulates are adcumulates, as shown by well-defined trace element trends (Duchesne, 1978), absence of intercumulus minerals, and lack of zoning in non granulated plagioclase. One must look for solid stage processes in which interfacial energy (Hunter, 1987) and not crystal-liquid relationship, controls the structure.

Quantitative data on the wetting properties of ilmenite are not available (see e.g. Spry, 1969), though the ability of this mineral to wet the surface of the silicate grains seems obvious in thin sections. A systematic measure of the angles between grain boundaries of ilmenite and the other minerals might provide some interesting results.

Mobility of ilmenite can be greatly enhanced by deformation. It has been observed that ilmenite is preferentially localized in pressure shadows around pyroxenes in some deformed Bjerkreim-Sokndal cumulates (Paludan et al., 1994). A similar observation is also reported here (Fig. 4) in a cumulate from the Hogstad layered body (Vander Auwera and Duchesne, this book). The ability of ilmenite to fit the interstices between the granulation products of plagioclase is a common character, frequently invoked in favour of protoclasis (Michot, 1965). In a pressure shadow, the probability that an ilmenite grain could have crystallized contiguously with the neighbouring pyroxene has of course not a zero value. If it is the case, several plastic deformation mechanisms can explain its arrow-head shape. On the other hand, if ilmenite has migrated from any position in the rock to the low stress zone of the pressure shadow, this suggests that the deformation was migration creep (diffusional flow), following Cobble or Nabarro-Herring processes. Deformation-mechanism maps of polycrystalline ilmenite are not available, but Atkinson (1977) has provided data on magnetite and hematite, the latter mineral being structurally close to ilmenite. These data show that hematite at about 670°C has a much larger migration creep domain than magnetite. For mm-sized grains and strain rates between 10⁻¹² and 10⁻¹⁴ sec⁻¹, deformation would be at the limit between migration and dislocation creeps and would imply deviatoric stress of 10-100 MPa, i.e. rather plausible deformation conditions.

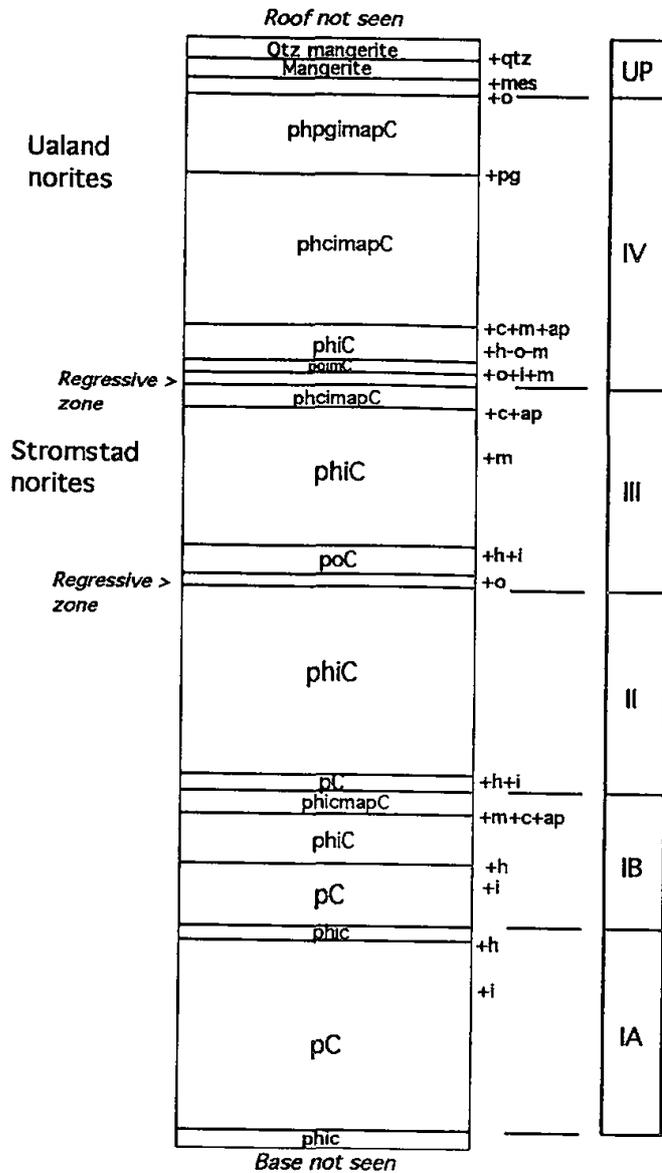
Table 1 Experiments on the Tjörn jotunitite (after Vander Auwera and Longhi, 1994)

Experim. #	T(°C)	P	Time (h)	fO ₂	Products(*)
TJ-37	1190	1 bar	16	NNO	gl86, pl6, ilm 1, mt7
TJ-31	1160	1 bar	18	NNO	gl75, pl15, ilm 1, mt9
TJ-45	1124	1 bar	93	FMQ-1	gl69, pl25, ol5, ilml

(*) Abbreviations: gl: glass; pi: plagioclase; ol: olivine; ilm: ilmenite; mt magnetite; the figures are modal proportions in per cent. **Table 1**

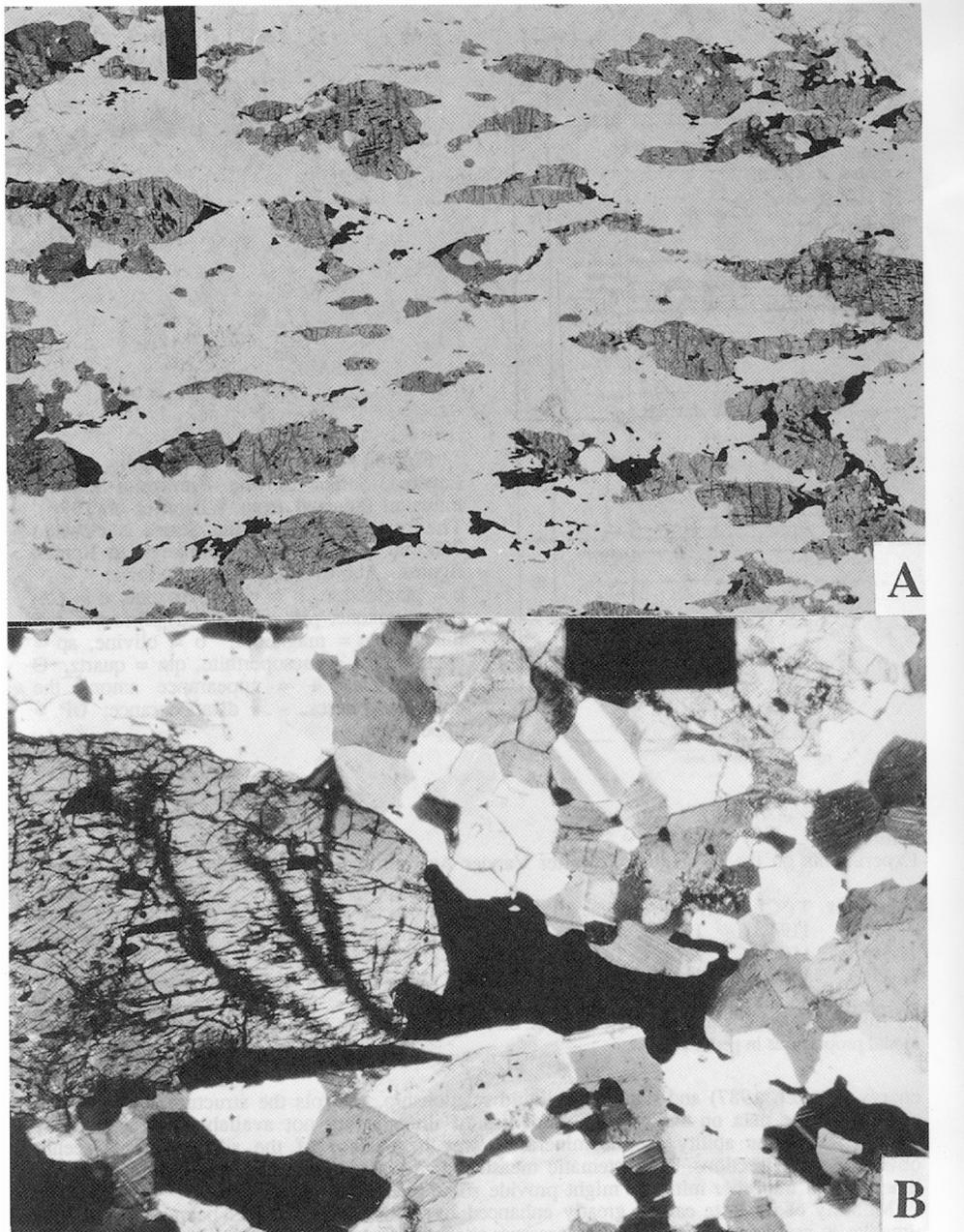
Postprint (Author's version)

Fig. 3 Generalised stratigraphy of the Layered Series of the Bjerkreim-Sokndal intrusion (adapted from Wilson et al. 1996). The total thickness of the Series is close to 7000 m. The MCU are numbered in Roman figures. Abbreviations are as follows: p = plagioclase, h = orthopyroxene, c = Ca-rich pyroxene, pig = (inverted) pigeonite, i = ilmenite, m = magnetite, o = olivine, ap = apatite, mes = mesoperthite, qtz = quartz, C = cumulate; + = appearance among the cumulate phases, - = disappearance; UP = upper part.



Postprint (Author's version)

Fig. 4 Slightly deformed leuconoritic cumulate from the Hogstad layered body (Rogaland) (specimen #JCD73-02). **A.** Overview of the structure showing subhedral orthopyroxene crystals defining the igneous lamination. Note the preferential localization of the ilmenite grains in the prolongation of the pyroxene grains. The black object is 1 mm-thick. **B.** Detailed view showing arrow-shaped ilmenite grain localized in a pressure shadow of a pyroxene grain. Note the granulated texture of the plagioclase and the bent shape (ondulous extinction) of the pyroxene. Slightly uncrossed nicols.



6. Ilmenite deposits

The ability shown by ilmenite to deform and migrate suggests that similar creep mechanisms can explain the pure ilmenite vein deposits. There is however a big jump from the mm scale of the interstitial ilmenite to the meter scale vein deposits and many uncertainties need to be clarified. The geological environment of the massive anorthosites certainly favours creep mechanisms. Deformation of these massifs occurs during emplacement and takes place at very high temperature, close to the solidus. Dynamic recrystallization is a possible mechanism.

The origin of the ilmenite can be found in layered bodies where, in the early stages of the evolution of the parent magmas, ilmenite has crystallized ahead of the other mafic minerals, thus forming layers of pure ilmenite. Subsequent deformation can deform the layers to produce veins of pure ilmenite. Much evidence in favour of

Postprint (Author's version)

such a general process is found in Rogaland (Duchesne, in press). Firstly, sill-like magma chambers of relatively small dimensions, such as the Løyning layered intrusion (Ernst and Duchesne, 1991), occurred in the Egersund-Ogna diapiric intrusion. Secondly, many small highly deformed deposits, made up of pure ilmenite, show evidence of primary igneous layering, still preserved at places. It is the case e.g. in the Swannes and Kydlandsvatn deposits (Duchesne, in press). Thirdly, the ilmenite from these occurrences is very rich in Cr and Mg, in agreement with the fact that they have crystallized at the very beginning of the magmatic evolution (Duchesne, in press). From that point of view these deposits are very similar to the vein-type deposit of Jerneld.

7. Conclusions

Since it is not possible to produce ilmenite-pure liquids by any igneous process (fractional crystallization or immiscibility), one has to admit that ilmenite vein type deposits were formed by mobilization through migration and recrystallization of highly deformed ilmenite layers. The latter were cumulated in small magma chambers at the beginning of the evolution of the anorthosite parent magma.

The interstitial character of ilmenite in common rocks does not result from a late stage crystallization, but from solid stage processes. To remain valid the concept of protoclasis should not be defined on the basis of the interstitial character of the opaques.

ACKNOWLEDGEMENTS

The author wishes to thank J. Vander Auwera and J. Longhi for continuous discussions on the jotunitic magmatism. J. Verkaeren and M. Ohnenstetter are thanked for their reviews. This work has been supported by the Belgian Fund for Collective Research.

REFERENCES

- Ashwal, L.D. (1993) Anorthosites. Springer, Berlin, Heidelberg, 422 p.
- Atkinson, B.K. (1977) The kinetics of ore deformation: its illustration and analysis by means of deformation-mechanism maps. *Geologiska Föreningens i Stockholm Förhandlingar*, 99, 186-197.
- Bateman, A.M. (1951) The formation of late magmatic oxide ores. *Economic Geology*, 46, 404-426.
- Demaiffe, D. and Hertogen, J. (1981) Rare earth element geochemistry and strontium isotopic composition of a massif-type anorthositic -charnockitic body: the Hydra massif (Rogaland, SW. Norway). *Geochimica et Cosmochimica Acta*, 45, 1545-1561.
- Duchesne, J.C. (1970) Microstructures of Fe-Ti oxide minerals in the South Rogaland anorthositic complex (Norway). *Annales de la Société Géologique de Belgique*, 93, 527-544.
- Duchesne, J.C. (1972) Iron-titanium oxide minerals in the Bjerkrem-Sogndal massif, Southwestern Norway). *Journal of Petrology*, 13, 57-81.
- Duchesne, J.C. (1973) Les gisements d'oxides de Fe et Ti dans les roches anorthositiques du Rogaland (Norvège méridionale). In *Les roches plutoniques dans leur rapport avec les gîtes minéraux*, p. 241-248. Masson, Paris.
- Duchesne, J.C. (1978) Quantitative modeling of Sr, Ca, Rb and K in the Bjerkrem-Sogndal layered lopolith (S.W. Norway). *Contribution to Mineralogy and Petrology*, 66, 175-184.
- Duchesne, J.C, Demaiffe, D., Roelandts, I. and Weis, D. (1985) Petrogenesis of monzonoritic dykes in the Egersund-Ogna anorthosite (Rogaland, S.W. Norway): trace elements and isotopic constraints. *Contribution to Mineralogy and Petrology*, 90, 214-225.
- Duchesne, J.C, Denoiseux, B. and Hertogen, J. (1987) The norite-mangerite relationships in the Bjerkreim-Sokndal layered lopolith (SW Norway). *Lithos*, 20, 1-17.
- Duchesne, J.C, Wilmart, E., Demaiffe, D. and Hertogen, J. (1989) Monzonorites from Rogaland (Southwest Norway): a series of rocks coeval but not comagmatic with massif-type anorthosites. *Precambrian Research*, 45, 111-128.
- Duchesne, J.C. (in press) Fe-Ti deposits in Rogaland anorthosites (South Norway): geochemical characteristics and problems of interpretation. *Mineralium Deposita*.

- Ernst, G.J. and Duchesne, J.C. (1991) Evidence of ultra-small layered intrusion : the Loyning lens, Egersund, Norway (abstract). *Terra abstracts*, 3, 426-427.
- Ford, C.E., Russel, D.G., Craven, J.A. and Fisk, M.R. (1983) Olivine-liquid equilibria: temperature, pressure and composition dependence on the crystal/liquid cation partition coefficients for Mg, Fe²⁺, Ca and Mn. *Journal of Petrology*, 24, 256-265.
- Hargraves, R.B. (1962) Petrology of the Allard Lake anorthosite suite, Quebec. *Geological Society of America (Buddington Volume)*, 163-190.
- Hubaux, A. (1960) Les gisements de fer titané de la region d'Egersund (Norvège). *Neues Jarbuch für Mineralogie (Festband Ramdohr)*, 94, 926-992.
- Hunter, R.H. (1987) Textural equilibrium in layered igneous rocks. In I. Parsons, Ed., *Origins of igneous layering*, p. 473-504. Reidel, Dordrecht.
- Lindsley, D.H., Epler, N.A. and Bolsover, L.R. (1988) Nature and origin of the Sybille Fe-Ti oxide deposit, Laramie anorthosite complex, Wyoming (abstract). *Penrose conference: The Origin and evolution of anorthosites and associated rocks, Wyoming, 14-19 August 1988*.
- Lindsley, D.H. (1991) Origin of Fe-Ti oxide deposits in the Laramie anorthosite complex (abstract). *IGCP 290, Adirondack meeting*, 13-19 Sept. 1991.
- Michot, P. (1965) Le magma plagioclasique. *Geologisch Rundschau*, 54, 956-976.
- Paludan, J., Hansen, U.B. and Olesen, N.6. (1994) Structural evolution of the Precambrian Bjerkreim-Sokndal intrusion, South Norway. *Norsk Geologisk Tidsskrift*, 74, 185-198.
- Philpotts, A.R. (1967) Origin of certain iron-titanium oxide and apatite rocks. *Economic Geology*, 62, 303-315.
- Philpotts, A.R. (1981) A model for the generation of massif-type anorthosites. *Canadian Mineralogist*, 19, 233-253.
- Roedder, E. (1979) Silicate liquid immiscibility in magma. In H.S. Yoder, Ed., *The evolution of the igneous rocks. Fiftieth Anniversary Perspectives*, p. 15-57. Princeton University Press, Princeton.
- Ryerson, F.J. and Hess, P.C. (1978) Implications of liquid-liquid distribution coefficients to mineral-liquid partitioning. *Geochimica et Cosmochimica Acta*, 42, 921-932.
- Spry, A. (1969) *Metamorphic textures*. Pergamon, Oxford, 350 p.
- Vander Auwera, J. and Longhi, J. (1994) Experimental study of a jotunite (hypersthene monzodiorite): constraints on the parent magma composition and crystallization conditions (P, T, fO₂) of the Bjerkreim-Sokndal layered intrusion *Contribution to Mineralogy and Petrology*, 118, 60-78.
- Vander Auwera, J. and Duchesne, J.C. (this book) *Petrology and geochemistry of the noritic Hogstad layered body (Rogaland, SW Norway): implications for the nature of the andesine anorthosite parent magma*.
- Wager, L.R. and Brown, G.M. (1968) *Layered igneous rocks*. Oliver and Boyd, London, 588 P.
- Watson, E.B. (1976) Two-liquid partition coefficients: experimental data and geochemical significance. *Contribution to Mineralogy and Petrology*, 56, 119-134.
- Weidner, J.R. (1982) Fe oxide magma in the system Fe-C-O. *Canadian Mineralogist*, 20,555-566.
- Wiebe, R.A. (1992) Proterozoic anorthosite complexes. In K.C. Condie, Ed., *Proterozoic crustal evolution (Development in Precambrian Research vol. 10)*, p. 215-262. Elsevier, Amsterdam.
- Wilmart, E., Demaiffe, D. and Duchesne, J.C. (1989) Geochemical constraints on the genesis of the Tellnes ilmenite deposit (S.W. Norway). *Economic Geology*, 84, 1047-1056.
- Wilson, J.R., Robins, B., Nielsen, F., Duchesne, J.C. and Vander Auwera, J. (1996) The Bjerkreim-Sokndal layered intrusion, Southwest Norway. In R.G. Cawthorn, Ed., *Layering in Igneous Complexes*, p. 231-256. Elsevier, Amsterdam.