

Intergrowths of hexagonal and monoclinic pyrrhotites in some sulphide ores from Norway

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Abstract

Pyrrhotites in polished sections from more than twenty stratabound massive sulphide and magmatic nickel-copper deposits in Norway were studied under the microscope using the magnetic colloid method. In both types of deposits, two distinct styles of intergrowths between monoclinic and hexagonal pyrrhotites were found: crystallographically-controlled lamellar intergrowths and fissure-controlled irregular intergrowths.

Lamellar intergrowths consist of crystallographically oriented monoclinic lamellae occurring in a hexagonal matrix and were produced originally by exsolution from hexagonal pyrrhotite on cooling. Irregular intergrowths comprise blades and patches of monoclinic pyrrhotite occurring along fissures and grain boundaries of hexagonal pyrrhotite, and were formed by interactions between hexagonal grains and sulphur-rich hydrothermal solutions.

Increase in lamella thickness and spacing, development of lamella zonations, wedge-shaped composite ends, boxworks and composite lamellae were caused by progressive lamellae coarsening and maturation during natural annealing, which could have been promoted by anisotropic stress. Metamorphic recrystallization and annealing tend to homogenize pyrrhotite and erase preexisting exsolution lamellae.

KEYWORDS: annealing, exsolution, magnetic colloid, metamorphism, pyrrhotite.

Introduction

SINCE monoclinic pyrrhotite, Fe_7S_8 , was first recognized by Byström (1945), much research has been carried out on hexagonal and monoclinic pyrrhotites (hpo and mpo) in terms of their composition, structure, physical properties and intergrowth (Carpenter and Desborough, 1964; Arnold, 1967; Yund and Hall, 1969; Bennett *et al.*, 1972) their stability and equilibrium relations to each other and to other iron sulphides (Arnold, 1962, 1969; Toulmin and Barton, 1964; Desborough and Carpenter, 1965; Clark, 1966; Yund and Hall, 1970; Kissin and Scott, 1982; Lusk *et al.*, 1993) and their occurrence in ore deposits (Vaughan *et al.*, 1971; Carpenter, 1974; Zhang Zhengen *et al.*, 1976; Kübler and Lindqvist, 1979; Gu *et al.*, 1988, 1993). In Norway, however, little work has been published

on the internal textures of pyrrhotite grains in sulphide ores although volcanite-hosted massive sulphides and magmatic Ni-Cu ores were, respectively, once the main copper and nickel sources of the country and have given rise to an enormous volume of literature. The present contribution aims at describing intergrowth styles of different varieties of pyrrhotite from a selection of Norwegian ores and subsequently discussing their origin.

In spite of the fact that various superstructures have been found for the pyrrhotite group (Desborough and Carpenter, 1965; Nakazawa and Morimoto, 1970, 1971), we have chosen to adopt the traditional classification of these iron monosulphides, except troilite, into the two general varieties of monoclinic and hexagonal (Arnold, 1962, 1967, 1969; Yund and Hall, 1969; Vaughan and Craig, 1978) in order to fit the magnetic colloid method (see Craig and Vaughan, 1981) used in this work.

Most of the samples used in this research form part of the ore mineralogy collection of the Norwegian Institute of Technology, University of Trondheim,

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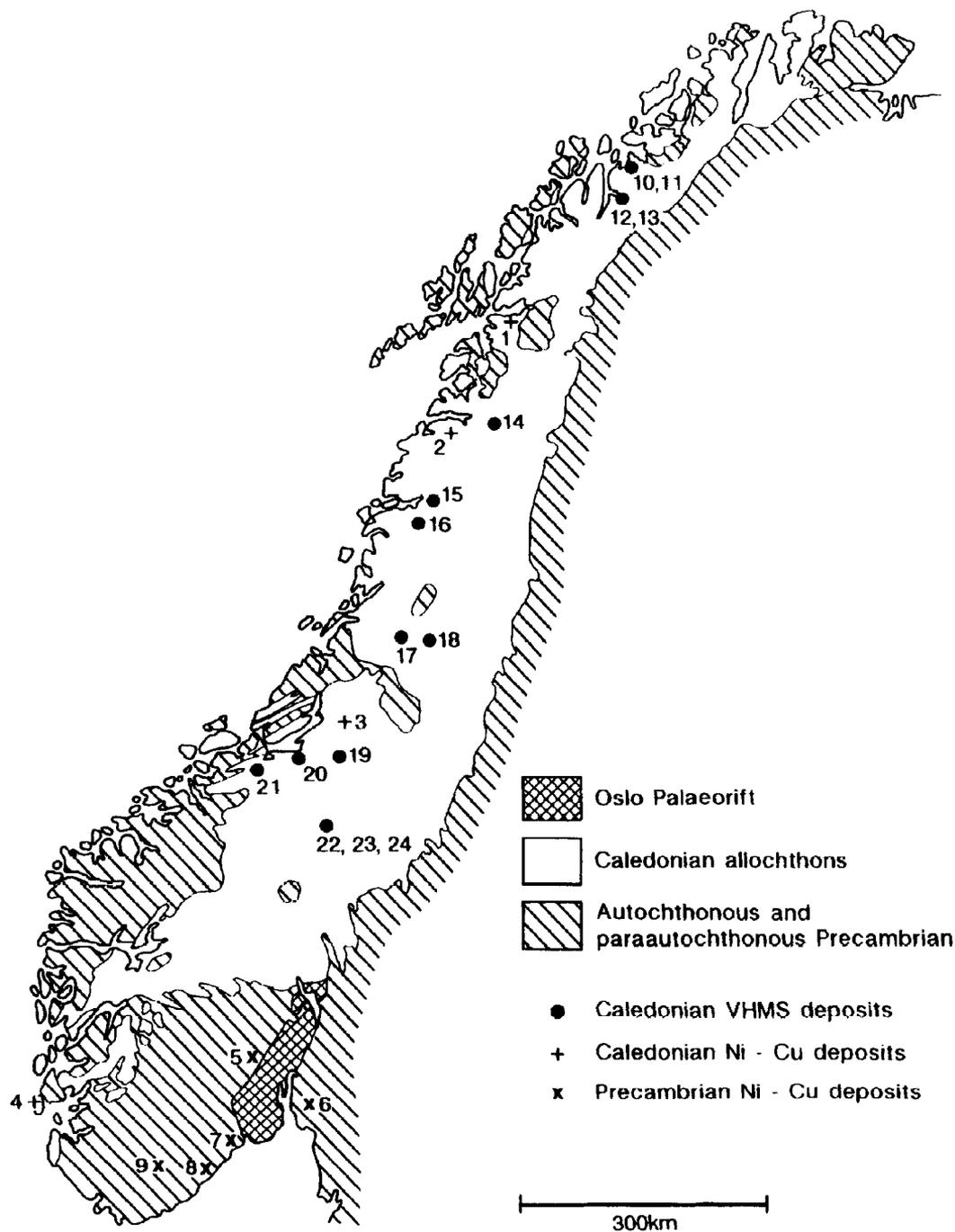


FIG. 1. Locations of sampled nickel-copper and stratabound massive sulphide deposits in Norway (see Tables 1 and 2 for deposit names).

TABLE 1. Geological features of sampled Precambrian and Caledonian Ni-Cu deposits in Norway (See Fig. 1 for locations)

No.	Deposit	Region	Age	Host rock	Ore minerals ¹	References
1	Bruvann	Råna	L. Paleozoic	peridotite	pn+cp+po	Boyd and Nixon 1985
2	Beiam	Salten	L. Paleozoic	gabbro	pn+cp+po+mt	Boyd and Nixon 1985
3	Skjækerdalen	Trondheim	L. Paleozoic	gabbro	pn+cp+po	Boyd and Nixon 1985
4	Fæøy	Karmøy	L. Paleozoic	sheeted dykes	pn+cp+py+po	Boyd and Nixon 1985
5	Ertelien	Ringerike	Proterozoic	norite	pn+cp+po+py	Boyd and Nixon 1985
6	Svinndal	Østfold	Proterozoic	metagabbro	pn+cp+po	Rekstad 1921
7	Skogen	Bamble	Proterozoic	metanorite	pn+cp+po+py+mt	Boyd and Nixon 1985
8	Brattåsen	Arendal	Proterozoic	pyroxenite	pn+cp+po	Brickwood 1986
9	Flåt	Evje	Proterozoic	ultram./amphib.	pn+cp+po+py	Boyd and Nixon 1985

¹ Abbreviations for minerals: cp – chalcopyrite; mt – magnetite; po – pyrrhotite; pn – pentlandite; py – pyrite.

Norway. Polished and polished thin sections from more than 20 deposits (Fig. 1, Tables 1 and 2) have been investigated. The samples are distributed unevenly throughout these deposits, partly due to the rare occurrence or absence of pyrrhotite in some deposits, partly due to lack of suitable material. However, we feel that the deductions and conclusions of this paper do not depend too much upon the representativity of the samples from the individual deposits, but instead upon the samples themselves.

Ore geological background

The main pyrrhotite-bearing ores of Norway comprise: (1) orthomagmatic Ni–Cu ores of varying ages, palaeotectonic origins and present

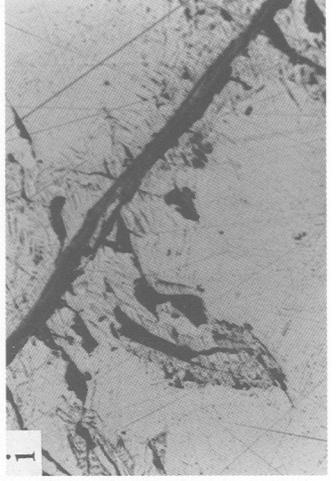
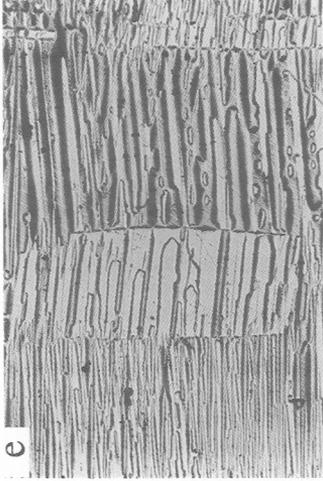
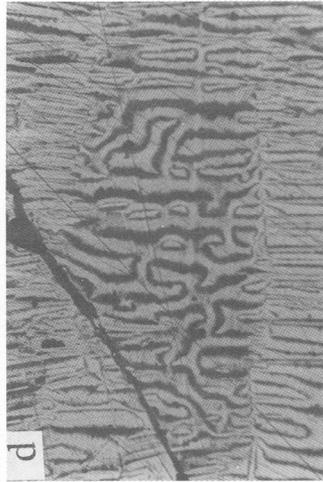
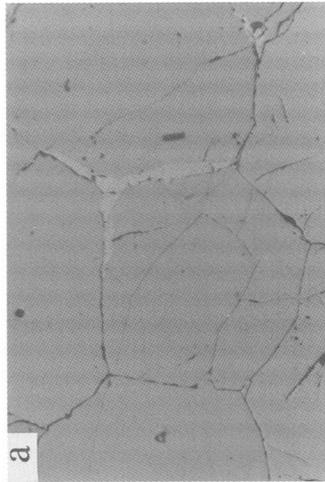
tectonostratigraphic environment; (2) stratabound, often stratiform, base-metal bearing ores of the volcanite-hosted and sediment-hosted massive sulphide (VHMS and SHMS) types. Both types of ores have been deformed and metamorphosed to varying degrees since their initial formation.

Nickel mineralizations in Norway occur in mafic/ultramafic igneous rocks ranging in age from Archaean to Lower Palaeozoic (Caledonian). Historically the main production came from deposits in the Upper Proterozoic (Sveconorwegian or Grenvillean) orogenic belts, especially in southern Norway. In recent years more interest has accrued to nickel possibilities in the Caledonides and the only primary Norwegian nickel producer at the present time is situated in a Caledonian synorogenic intrusive

TABLE 2. Geological features of sampled Caledonian massive sulphide deposits in Norway (see Fig. 1 for locations)

No.	Deposit	Region	Age	Metal	Ore minerals ¹	Metamorphic facies	References
10	Rieppe	Vaddas	Ordov.	Zn-Cu	sph+cp+po	amphibolite	Lindahl, 1974
11	Gresdalen	Vaddas	Ordov.	Cu-Zn	cp+sph+po+py	greenschist	Lindahl, 1974
12	Moskogaissa	Birtavarre	End-Ordov.	Cu	cp+po	amphibolite	Vokes, 1957
13	Sabetjok	Birtavarre	End-Ordov.	Cu	cp+po+py	amphibolite	Vokes, 1957
14	Sulitjelma	Sulitjelma	Ordov.	Cu-Zn	sph+cp+py+po	amphibolite	Cook <i>et al.</i> , 1990, 1993
15	Sæterdalen	Rana	Cambrian?	Zn	cp+po	amphibolite	
16	Bleikvassli	Rana	Precamb. to Camb.	Pb-Zn	sph+gn+py+po	amphibolite	Vokes, 1963
17	Gjersvik	Grong	E. Ordov.	Cu-Zn	sph+cp+py+po	greenschist	Reinsbakken, 1986a
18	Joma	Grong	E. Ordov.	Cu-Zn	sph+cp+py+po	greenschist	Reinsbakken, 1986b
19	Meraker	Trondheim	L. Ordov.	Cu	cp+sph+py+po	greenschist	Foslie, 1926
20	Leksdal	Trondheim	E. Ordov.	Fe-S	py+po+mt	greenschist	Sand, 1986
21	Løkken	Trondheim	E. Ordov.	Cu-Zn	cp+sph+py+po	greenschist	Grenne <i>et al.</i> , 1980
22	Lergruvebakken	Røros	M. to L. Ordov.	Zn-Cu	sph+cp+py+po	amphibolite	Bugge, 1978
23	Mugg	Røros	End-Ordov.	Cu	cp+po+mt	amphibolite	Bugge, 1978
24	Olavs	Røros	End-Ordov.	Cu	cp+sph+gn+po	amphibolite	Bugge, 1978

¹ Abbreviations for minerals: cp – chalcopyrite; gn – galena; mt – magnetite; po – pyrrhotite; py – pyrite; sph – sphalerite



complex. The reader is referred to the review by Boyd and Nixon (1985) for further details.

Base-metal bearing, pyritic/pyrrhotitic, stratabound massive sulphide ores, mainly of VHMS class, are from an historical-economical point of view the most important of the deposit types of the Scandinavian Caledonides in Norway (Foslie, 1926; Vokes, 1976; Bjørlykke *et al.*, 1980). On the basis of their metal contents these ores may be described as either Cu, Cu-Zn or Zn-Pb-(Cu) types though the majority fall into the Cu-Zn type. They were generated in several different palaeotectonic environments during the course of the Caledonian orogenic cycle and deformed/metamorphosed during this cycle (Vokes, 1968, 1988; Stephens *et al.*, 1984).

A variety of stratabound sulphide mineralization which has been included in the present investigation — of restricted occurrence and no present economic importance — is the so-called 'vasskis', found in parts of the Caledonian orogen. The term has been used historically in Norway to denote relatively thin stratigraphical units of base-metal free iron formation of different mineralogical facies. Interest in the present case has been focused on the sulphidic facies of the 'vasskis', (see, e.g. Carstens, 1923; Sand 1986).

Tables 1 and 2 summarise the main features of the deposits sampled during the present study.

Pyrrhotite in nickel-copper deposits

Nickel-copper ores hosted by mafic and ultramafic rocks (Table 1), whether in the Precambrian terranes

or in the Caledonides in Norway, occur as massive to disseminated sulphides. They are composed principally of pyrrhotite, chalcopyrite, pentlandite with or without pyrite and/or magnetite. Pyrrhotite is by far the most abundant sulphide in practically all the samples examined during the present work. Magnetite occurs, in most cases, as rounded to polygonal grains of variable size. Pyrite is absent or occurs as a minor constituent in the ores. Chalcopyrite is usually present as irregularly dispersed anhedral polycrystalline aggregates and veinlets. Pentlandite was observed in three distinct textures: (1) oriented blebs and 'flames' inside pyrrhotite grains, (2) granular polycrystalline veinlets interstitial to pyrrhotite grains, (3) coarse grains larger than 1 mm in size. Graphite is reported to occur within sulphide disseminated in peridotite of the Råna deposit (Boyd and Mathiesen, 1979).

Pyrrhotite occurs in most cases as anhedral to subhedral grains with mutually curved boundaries and a size range of 2–6 mm. Occasionally, grains exceeding 10 mm in size are found. Deformation bands are not uncommon. In some deposits, pyrrhotite is completely annealed and hence exhibits typical 120° dihedral angles (Fig. 2a).

Magnetic colloid coating indicates that the majority of the samples exhibit intimate intergrowths of monoclinic and hexagonal pyrrhotites. Two distinct styles of intergrowths are observed: crystallographically-controlled lamellar intergrowths and fissure-controlled irregular intergrowths.

Lamellar intergrowth involves monoclinic pyrrhotite (mpo) occurring as roughly parallel lamellae in a

FIG. 2a–i Pyrrhotite textures in nickel-copper deposits. (a) Triple junction texture of hexagonal pyrrhotite in equilibrium with intergranular aggregates of pentlandite (white). No mpo lamellae are seen after coating with magnetic colloid. Råna, plane reflection, field width = 1.8 mm. (b) Crystallographically oriented intergrowth of mpo lamellae in mpo grains. Granular aggregates of pentlandite (white) occur along boundaries between pyrrhotite grains. Flåt, coated with magnetic colloid, plane reflection, field width = 0.45 mm. (c) Thickness zonation of mpo lamellae. A zone with fine, closely-spaced lamellae is intercalated between two zones with coarser and wider-spaced lamellae. In the fine lamella zone, two neighbouring lamellae approach mutually and meet together to form sharp wedge-shaped ends. Svinndal, coated with magnetic colloid, plane reflection, field width = 0.45 mm. (d) Lamellar intergrowth with thickness zonation and boxwork textures. A zone with coarse, wider-spaced and boxworked lamellae is intercalated between two zones with thin and closely-spaced lamellae. Typical boxwork textures can be seen in the coarser zone. Skogen, coated with magnetic colloid, plane reflection, field width = 0.45 mm. (e) Lamellar zonation between kink-bands in a pyrrhotite grain; monoclinic lamellae are also aligned along the kink boundaries. Boxworks are developed to different degrees in different zones. Svinndal, coated with magnetic colloid, plane reflected light, field width 0.45 mm. (f) Mpo lamellae aligned in the same direction as exsolved pentlandite flames. Flåt, coated with magnetic colloid, plane reflection, field width = 0.45 mm. (g) Two sets of composite lamellae. Secondary lamellae in the same set have the same orientation. Composite lamellae grade into a normal lamella zone towards the bottom of photograph. A coarse W-E trending lamella branches at both ends. Ertelien, coated with magnetic colloid, plane reflection, field width = 0.45 mm. (h) Lamellar-intergrown pyrrhotite cut by anastomosing gangue-mineral veinlets. Each fragment has a lamellae-free rim. Beiarn, coated with magnetic colloid, plane reflection, field width = 1.8 mm. (i) Mpo as selvages to quartz veinlet. Lamellae intergrowths are absent in the hpo on either side of the selvage. Brattåsen, coated with magnetic colloid, plane reflection, field width = 0.5 mm.

matrix of hexagonal pyrrhotite (hpo). The thickness of the mpo lamellae varies from less than 0.2 to 20 μm with the majority less than 10 μm , and they terminate or change direction abruptly at grain boundaries of the host (Fig. 2b). The relative proportions between mpo and hpo are highly variable from sample to sample. Lamellae of various thickness in the same pyrrhotite grain normally show zonal distribution, i. e. zones of fine lamellae alternate with zones of coarser ones (Fig. 2c).

Lamellae thinner than 2 μm are commonly straight, smooth, evenly spaced and parallel to each other (Fig. 2c). They vary between 0.03 and 0.5 mm in length with a lamella spacing of 3–4 μm . Some of these lamellae thicken at one end and then grade into the coarser ones, whereas others meet the neighbouring lamellae at their ends after approaching mutually, resulting in sharp wedge-shaped composite ends (Fig. 2c).

Lamellae coarser than 2 μm regularly show wider spacing (Fig. 2c). A coarse lamella normally joins a fine lamella at either end. Two fine lamellae that join the same coarse one at each end can be the neighboring ones in the same zone or those in opposite zones across the coarser zone (Fig. 2d). Typical boxwork textures can be seen in the coarser zones where the lamellae either change direction or close on themselves to form circles, triangles, rectangles or irregularly-shaped patterns (Fig. 2d,e).

Careful scrutiny shows that there are often slight differences in extinction positions between different lamellar zones and that boundaries between lamellar zones often coincide with subgrain or kink boundaries (Fig. 2e).

In spite of the zonations, boxworks and local irregularities, the lamellae are generally crystallographically oriented parallel to the long axes of exsolution blebs or 'flames' of pentlandite in the same pyrrhotite grains (Fig. 2f). Mpo lamellae in different pyrrhotite grains or even in different twin individuals or kink bands in the same grains normally trend in different directions (Fig. 2b,e). Occasionally, mpo lamellae are seen aligning along kink boundaries (Fig. 2e), indicating that the formation of these lamellae post-dates plastic deformation.

In the samples from Ertelien, Flåt and Brattåsen, what may be termed composite lamellae are encountered in many grains (Fig. 2g). These lamellae normally have thin lensoid or spindle shapes, with secondary lamellae inside. The proportion of mpo to hpo within a composite lamella varies between 1:1 and 9:1. Such lamellae can either stem out like bamboo shoots from the normal lamellar parts of the grains and then thin out, or thin out at both ends to form thin lenses (Fig. 2g). Composite lamellae in one area are often oriented in two sets with secondary lamellae in the same set trending in the same

direction; some composite lamellae have branches at their ends (Fig. 2g). Areas with composite lamellae often show slight optical discontinuity with the surroundings.

Of particular interest in this context is the texture in the samples from the Beiarn deposit where the pyrrhotite grains are intersected by anastomosing veinlets of gangue minerals. Most of the pyrrhotite fragments have a lamellar core and a lamellae-free clear margin (Fig. 2h). The width of the margin varies considerably from grain to grain and even in the same fragment. There is hardly any mpo core left in some grains. Such lamellae-free margins could be the result of an influx of iron-rich fluid which acted upon lamellar-intergrown pyrrhotite grains, donated iron to the lattice vacancies of the original mpo at grain or fragment margins and converted them to hpo.

The above delicate textures have not been well-described in the literature, although, microphotographs of composite lamellae of mpo have already been published from nickel-copper deposits elsewhere in the world (Arnold, 1967; Kissin and Scott, 1982).

No lamellae were found in any of the eight sections from the Bruvann (Råna) deposit except for one where small patches of mpo lamellae with boxworks occur inside the disseminated hpo grains. The pyrrhotite in the samples from this deposit has been recrystallized or annealed, and in some sections exhibits typical triple junctions (Fig. 2a). As a result of the annealing of the pyrrhotite, the numbers of blebs and flames of pentlandite were noticeably reduced and coarse grains with well-developed octahedral cleavages developed.

The term irregular intergrowth refers to mpo occurring as irregular patches, blobs and blades along grain boundaries, fissures or veinlets. Typical irregular intergrowth is well developed in the samples from Brattåsen and Sæterdalen (Fig. 2i).

Pyrrhotite in stratabound massive sulphide deposits

The ores of the Caledonian massive sulphide deposits (MSD) in Norway are characterized by various combinations of chalcopyrite, sphalerite and in places, galena, together with high contents of pyrite and/or pyrrhotite (Vokes, 1962). Troilite has been observed occurring as exsolution lamellae in pyrrhotite grains at Moskogaissa, Birtavarre (Vokes, 1957), where the ores are characterized by the near-absence of pyrite. The yellow colour with reflectivity higher than pyrrhotite and the strong anisotropism of this mineral agree well with the description of troilite in other districts (Fleet and Macrae, 1969; Gribble and Hall, 1992).

Ore textures and mineral parageneses of the Caledonian MSD deposits have been well documented (see, e.g. Craig and Vokes 1992). Pyrrhotite textures are in good harmony with the metamorphic grade of the hosting ores. In weakly deformed and metamorphosed ores like the 'vasskis' of the Leksdal and Løkken areas, most of the pyrrhotite occurs as fine- to medium-sized, anhedral to subhedral grains with mutually curved boundaries. The coexistence of pyrrhotite in 'vasskis' with large numbers of unmodified pyrite framboids (Fig. 3a), the lack of replacive textures in coexisting pyrite and the general absence of minerals of higher metamorphic grades, indicate that these pyrrhotites are primary precipitates that formed during sedimentary–diagenetic processes and survived later-stage low-grade regional metamorphism without marked recrystallization and annealing. With progressive deformation and metamorphism, pyrrhotite in MSD ores tends to show some straight boundaries due to recrystallization or incipient annealing. The development of elongated, thin, oriented and strainless grains in some of the samples from the Sulitjelma deposit may have resulted from recrystallization and grain growth in kinked matrices. Further metamorphism and annealing results in roughly equant grains with 120° dihedral angles at their boundaries (Stanton, 1972; Craig and Vokes, 1992) as can be seen, e.g. in Fig. 3b. Post-annealing plastic deformation which results in lattice distortion, grain elongation and kinkbanding of annealed pyrrhotite is also evident in some samples like those from the Sulitjelma and Moskogaissa deposits. Lattice distortion of annealed grains is discernible due to their distorted basal partings revealed by plucking pits. Deformation twinning (Clark and Kelly, 1976; Craig and Vaughan, 1981) is not so common as kinkbanding in the Norwegian massive sulphide ores. Most interestingly, second phase annealing might have occurred at the Moskogaissa and Mugg deposits, as indicated by the fact that smaller, strainless grains occur in a matrix of relatively coarser, annealed and subsequently deformed, grains (Fig. 3c).

Retrogressive and metahydrothermal pyrrhotites are also present in some of the massive sulphide deposits. Such later generations of pyrrhotite often strongly corrode pyrite porphyroblasts (Fig. 3d) or fill cracks (Fig. 3e), and are characterized by anhedral to subhedral grains with relatively weaker deformation and recrystallization.

As in the case of the nickel–copper deposits, the intergrowth styles of mpo with hpo in the MSD deposits also fall into two groups: lamellar and irregular.

Lamellar intergrowths can be found in two genetic types of pyrrhotites: (1) primary (sedimentary–diagenetic) pyrrhotites with weak recrystallization

and annealing, like those in the Gjersvik and Løkken deposits; (2) retrogressive and metahydrothermal pyrrhotites like those in the Sabetjok and Leksdal deposits.

Mpo lamellae in pyrrhotites of both the above genetic types have a common orientation in the same, undeformed grain, but show different orientations from grain to grain and even between different twin individuals or kink bands inside the same grain (Fig. 3a,d,e,f,g). They can either be present across the whole grain of hosting hpo, or can be terminated half way. Lamella thickness and spacing are highly variable from grain to grain and even inside a single hexagonal crystal. Local coarsening of and the resultant interconnection between lamellae may also give rise to blebs or irregular patches of mpo in the background of hpo (Fig. 3d,f,g).

Although pyrrhotite is extremely rare in the main base-metal ore at Løkken, mpo lamellae were found in the sample we examined (Fig. 3g). In some samples of 'vasskis' from the Løkken deposit, anhedral to subhedral primary pyrrhotite, 0.05–0.15 mm in grain size and in coexistence with framboidal pyrite, also shows well-developed mpo lamellae (Fig. 3a).

Lamellar zonations, wedge-shaped composite ends, boxworks and composite lamellae have been found locally in the Sabetjok, Gressdalen and Løkken deposits but are not so well developed as in the magmatic nickel–copper deposits.

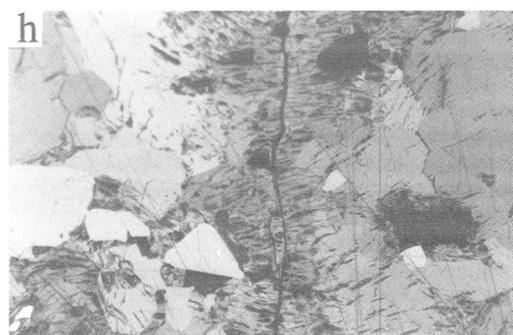
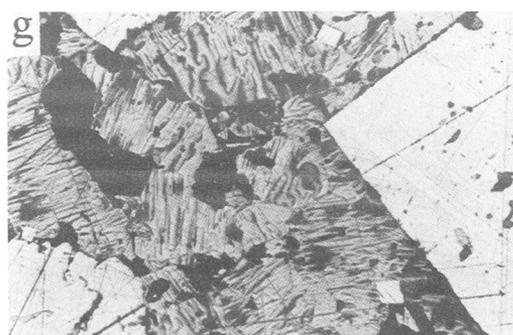
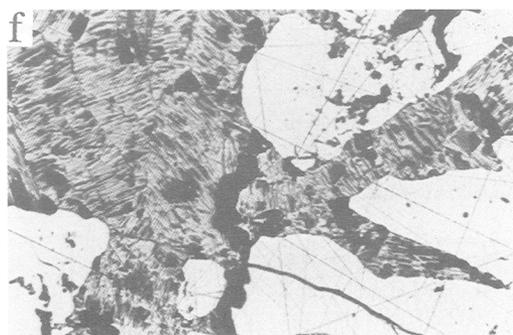
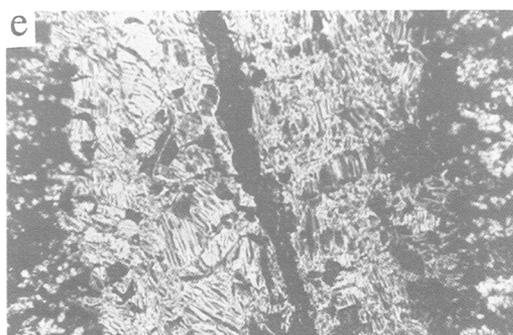
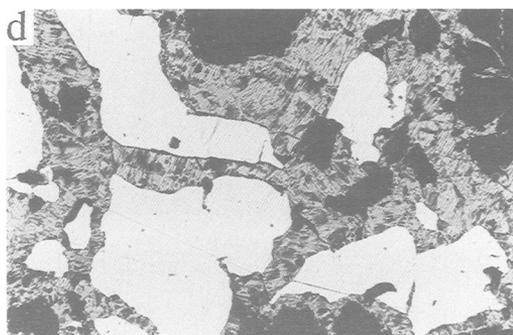
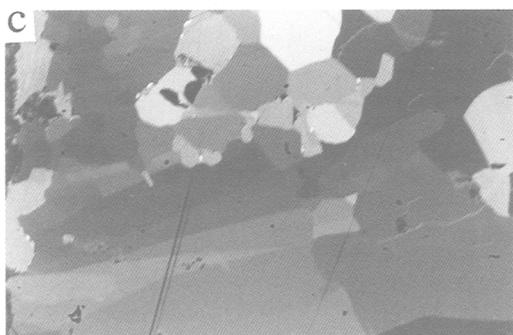
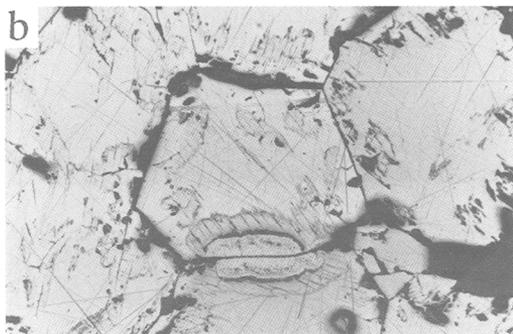
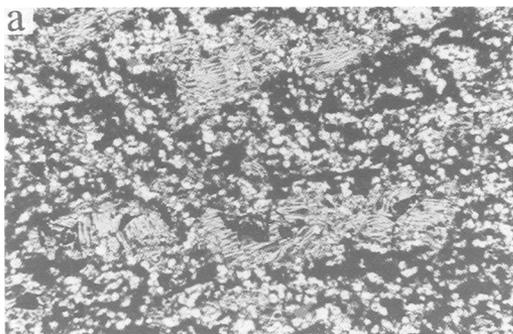
In general, intensely recrystallized and annealed pyrrhotite lacks lamellar intergrowth as in the deposits at Bleikvassli, Joma, Lergruvebakken, Meråker (Fig. 3b), Moskogaissa, Mugg (Fig. 3c), Olavs, Rieppe, Sæterdalen and Sulitjelma. Typically in the Gjersvik mine, two sorts of pyrrhotites may be distinguished on the basis of their distinctive grain size, crystal form and the existence or absence of mpo lamellae. Lamellar intergrowth is only found in anhedral to subhedral, non-annealed grains (Fig. 3f).

In contrast to annealed samples, non-annealed retrogressive or metahydrothermal pyrrhotite often shows mpo lamellae, as in the samples from Sabetjok and in the 'vasskis' ores of both Leksdal and Løkken (Fig. 3d,e).

Irregular intergrowths along grain boundaries, fissures or veinlets are commonly observed in the samples from Bleikvassli, Gjersvik, Joma, Leksdal, Lergruvebakken, Olavs and Sabetjok. Mpo blades in such intergrowths are often aligned at large angles to the cracks and veinlets (Fig. 3h).

Discussion and conclusions

As has been shown in the foregoing sections, there are two contrasting intergrowth styles, lamellar and irregular, between monoclinic and hexagonal



pyrrhotites in both the stratabound massive sulphides and the magmatic nickel-copper deposits. These two styles of intergrowths are different in origin.

Origin of lamellar intergrowths

The lamellar intergrowths can be well explained in terms of exsolution. The pyrrhotite solvus (Fig. 4) obtained from literature (Arnold, 1962; Kissin and Scott, 1982) implies that hexagonal pyrrhotite can exsolve pyrite during cooling at temperatures higher than 254°C and that the composition of the hosting pyrrhotite will move continuously along the solvus down to 254°C (Kissin and Scott, 1982). In the case where the final composition of hexagonal pyrrhotite lies between 47.4 and 46.67 atomic percent iron (Arnold, 1967; Yund and Hall, 1969), it will exsolve mpo as temperature drops rapidly across the upper boundary of the mpo+hpo two-phase field. Mpo actually exsolving from hexagonal grains has been observed in experiments (Von Gehlen, 1963; Arnold, 1969). Arnold (1969) also provided a series of photomicrographs of the products of his experiments. The exsolved monoclinic phase in Arnold's experiments is crystallographically oriented within its hexagonal host. Such an orientation can be accounted for by the common a and c axes of the two phases (Fleet and Macrae, 1969). The lamellar intergrowths of mpo in a hexagonal matrix in the Norwegian deposits are reminiscent of the exsolving monoclinic pyrrhotite in Arnold's (1969) experiments. Therefore, this kind of intergrowth in these deposits could also have been formed by exsolution during a period of decreasing temperature.

As part of the ore-forming processes for magmatic nickel-copper deposits, the separation of an immis-

cible sulphide melt from a sulphur-saturated magma, the succeeding cooling of the melt, and the crystallization and exsolution of monosulphide solid solution, have been fully documented by previous authors (Naldrett *et al.*, 1967). Mpo lamellae in pyrrhotite should be the direct product of post-magmatic exsolution of this monosulphide, taking place when temperature fell below the upper boundary of the hpo+mpo field. In view of the fact that pentlandite has a lower sulphur:metal ratio than pyrrhotite (Naldrett *et al.*, 1967; Kelly and Vaughan, 1983) and can continue to be exsolved at temperatures down to 100°C (Naldrett *et al.*, 1967; Craig and Vaughan, 1981), the exsolution of mpo lamellae could be enhanced by and accompany the exsolution of pentlandite 'flames' and blebs; these two sorts of precipitates will be retained in the same crystallographical directions of the host pyrrhotite. (Fig. 2f).

Primary pyrrhotite in the Caledonian massive sulphide deposits has undergone at least two phases of thermal processes since it was precipitated, viz: a submarine geothermal process and a metamorphic heating process. Direct determinations on ocean ridge hydrothermal systems, fluid inclusion studies and isotopic differences between mineral phases indicate that volcanogenic massive sulphide deposits can form at temperatures higher than 300°C and even up to 350°C (MacDonald *et al.*, 1980; Cathles, 1993; Lalou *et al.*, 1993). Due to the presence of what they term 'isochalcopyrite' exsolution lamellae in the so-called intermediate solid solution (iss), Missack *et al.* (1989) suggested that the copper-iron sulphides in the Atlantis II deep of the Red Sea formed above 450°C. Such temperatures are favourable for the crystallization of hpo and may be attained

FIG. 3a-h. Pyrrhotite textures in massive sulphide deposits. (a) Lamellar intergrowth of mpo in fine-grained anhedral-subhedral pyrrhotite grains coexisting with pyrite frambooids (round white spots). Løkken vasskis ore, coated with magnetic colloid, plane reflection, field width = 0.5 mm. (b) Annealed pyrrhotite with 120° dihedral angles and the 'bird's eye' texture induced by weathering. The 'bird's eye' texture is composed of a rim of marcasite laths and a core of minute pyrite grains. No mpo lamellae are seen after coating with magnetic colloid. Meråker, plane reflection, field width = 0.5 mm. (c) Two phases of pyrrhotite annealing. Deformed grains in the sample show straight boundaries, indicating first phase annealing. Second phase annealing occurs locally on post-annealing deformation bands. No mpo lamellae are seen after coating with magnetic colloid. Mugg, plane reflection, field width = 0.5 mm. (d) Lamellar intergrowth in pyrrhotite grains that corrode pyrite (white). Sabetjok, coated with magnetic colloid, plane reflection, field width = 0.5 mm. (e) A pyrrhotite veinlet cutting frambooidal pyrite ore. All the pyrrhotite grains have well-developed mpo lamellae. Løkken vasskis ore, coated with magnetic colloid, plane reflection, field width = 0.5 mm. (f) Lamellar intergrowth of pyrrhotite. Mpo lamellae are crystallographically oriented. Irregular grains are pyrite (white). Gjersvik mine, coated with magnetic colloid, plane reflection, field width = 0.5 mm. (g) Lamellar intergrowth of mpo in anhedral-subhedral pyrrhotite coexisting with pyrite porphyroblasts (white). Boxworks are seen in some grains. Løkken main orebody, coated with magnetic colloid, plane reflection, field width = 0.5 mm. (h) Irregular intergrowth of mpo, where mpo occurs along cracks and grain boundaries in an annealed hpo sample. 120° dihedral angles between annealed hpo grains can be seen on either side of the major crack. Gjersvik, coated with magnetic colloid, partially crossed nicols, field width = 0.5 mm.

particularly in the case of proximal deposits. When the temperature of the submarine hpo falls below the transformation boundary between hpo and hpo+mpo, it will exsolve mpo if its bulk composition plots in the hpo+mpo region. Typical exsolution from primary pyrrhotite can be seen in the samples from the main orebodies of the Løkken and Gjørsvik deposits, both of which have been documented as proximal ones (Grenne, 1989; Reinsbakken, 1986a). The Løkken ore is thought to have been formed at about 300–350°C (Grenne, 1989). The 'vasskis' mineralization of the Løkken deposit, which exhibits delicate mpo lamellae, is located stratigraphically above and laterally to the main ore body and could also be proximal. Two phases of pyrrhotite have also been reported from the Broadlands geothermal field (Kissin and Scott, 1982) and the Atlantis II deep (Missack *et al.*, 1989).

Most of the stratabound massive sulphide deposits in the Norwegian Caledonides were metamorphosed to greenschist or amphibolite facies (Table 2). Pyrrhotite in these deposits has been deformed, recrystallized and annealed during prograde metamorphism (Table 2 and Vokes, 1968; Cook *et al.*, 1993). Annealing of pyrrhotite is generally thought to be important only above 450°C, which is well above 254°C, the transformation temperature from mpo to hpo (Fig. 4) according to Kissin and Scott (1982). As a consequence, metamorphic annealing will convert lamellar mpo into hpo to form a uniform hpo grain (Kissin and Scott, 1982). Slow cooling following peak-metamorphism will give sufficient time for such homogenized pyrrhotite to exsolve pyrite (Arnold, 1962; Yund and Hall, 1970) with the

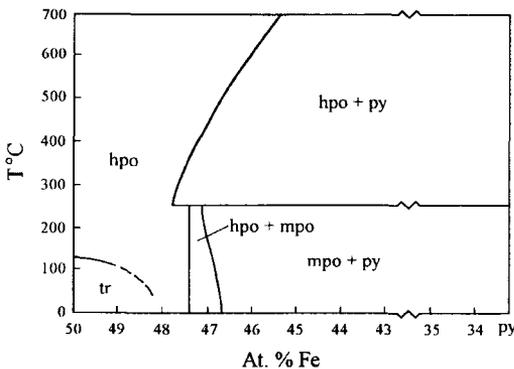


FIG. 4. Highly schematic diagram of the Fe-S system (data from Arnold, 1962, 1967; Desborough and Carpenter, 1965; Kissin and Scott, 1982). Mineral phases: hpo = hexagonal pyrrhotite; mpo = monoclinic pyrrhotite; py = pyrite; tr = trolite.

formation of retrogressive pyrite porphyroblasts rather than to exsolve mpo at lower temperatures (cf. Craig and Vokes 1993).

Gu and McClay (1992) studied ore samples from the Sullivan massive sulphide deposit in Canada, which has been strongly deformed and metamorphosed to upper greenschist facies. The pyrrhotite in that deposit was strongly annealed and occurs in association with retrogressive, porphyroblastic pyrite cubes. No mpo lamellae were observed by the magnetic colloid method. Scott *et al.* (1977) investigated the sulphide ores of the Broken Hill region, where the ore-hosting Willyama Complex has been subjected to granulite facies regional metamorphism (Markham and Stevens, 1982). No primary mpo lamellae were reported by these workers as a result of etching of pyrrhotite samples. The absence of mpo in the Ducktown deposit, Tennessee, could also be related to annealing and subsequent slow cooling as is suggested by Lusk *et al.* (1993). All these results support a general idea that metamorphism and annealing tend to erase earlier exsolved mpo lamellae in hpo. This idea also holds true for the Ni-Cu ores, as is shown by the Bruvann deposit where small patches of boxworked mpo lamellae inside hpo grains in one sample can be regarded as the remnants that survived annealing.

In addition to the effect of temperature, the processes of recrystallization, annealing and exsolution are also significantly controlled by strain (Spry, 1969; Stanton, 1972). Recent research tends to recognize anisotropic stress as an accelerating factor for exsolution (Kübler and Lindqvist, 1979; Lusk *et al.*, 1993). These two parameters and the resultant extent of pyrite-pyrrhotite re-equilibration are highly variable over small distances in a deposit or even in the same samples (Kübler and Lindqvist, 1979). With this in mind, we are able to account for the coexistence of lamellae-free, annealed pyrrhotite with primary fine-grained and subhedral to anhedral pyrrhotite in samples from the same deposit, as at Gjørsvik and at Løkken.

The good preservation of mpo lamellae in retrograde or metahydrothermal pyrrhotite grains from the Sabetjok copper ores and the vasskis of the Leksdal and Løkken deposits could have been made possible by the absence of late-stage, high-temperature geothermal events. This is in good agreement with the research on pyrrhotite from the Mashan massive sulphide deposit in South China. Pyrrhotite ores at Mashan were derived from Middle Carboniferous pyritic sediments by contact metamorphism induced by a Mesozoic quartz diorite intrusion. The faster cooling and the lack of post-pyrrhotite metamorphism resulted in mpo lamellae being well-preserved in almost every pyrrhotite grain from that deposit (Gu *et al.*, 1988, 1993).

Lamellar coarsening

Mpo lamellae in the Ni–Cu deposits are very distinct from those in the massive sulphide deposits in their typical zonations, boxworks and composite internal textures. These special features can be accounted for in terms of lamellae annealing and grain coarsening.

It has been shown that lamellar exsolution will produce two forms of free energy, i.e. exsolution strain free energy and interfacial free energy ($+\Delta G_{\text{strain}}$ and $+\Delta G_{\text{interfacial}}$), which are opposite in sign to the change in the Gibbs or volume free-energy ($-\Delta G_{\text{vol}}$) accompanying exsolution (Robin, 1974; Yund and Davidson, 1978; Kelly and Vaughan, 1983). That means that the exsolution precipitates (lamellae) will not have reached final equilibrium and that these kinds of energy will become the driving force for the coarsening of precipitates during annealing.

Experiments have been carried out on the coarsening of exsolution lamellae of metals and minerals (Yund *et al.*, 1974; Joesten, 1991; Durazzo and Taylor, 1982). Annealing during slow cooling or slow heating will cause exsolution lamellae to increase in thickness and spacing, to be reduced in number and to become irregular in shape (Yund *et al.*, 1974; Yund and Davidson, 1978).

With respect to the zonal distribution of mpo lamellae in the Ni–Cu ores, zones with coarser and well-spaced lamellae may represent a more advanced stage of annealing than the finer and more closely-spaced lamellae zones. The coarser lamellae could have grown at the expense of their neighbouring ones by volume diffusion of the material through the intervening hpo matrix (Brady, 1987). When two adjacent source lamellae have diminished to the degree that the driving force for diffusion is no longer able to remove material from them to the coarsening lamellae nearby, the ends of the two diminished lamellae will approach and finally join each other to form wedge-shaped composite ends so that the interfacial free energy and the strain energy at their terminals would be reduced to a minimum. (Fig. 2c.)

In the samples that were subjected to annealing at the highest temperature and for the longest duration in the experiments of Yund *et al.* (1974), exsolution lamellae became distorted to give fairly irregular configurations, to a certain extent similar to the boxworks of mpo in the Norwegian Ni–Cu deposits. Consequently, the boxwork textures, which are exclusively composed of coarser lamellae, may mark the more advanced stage of annealing. Composite lamellae, which appear sometimes in a background of boxworks and are often accompanied by local homogenization of the pyrrhotite, may therefore indicate the further maturation of the lamellae. Slight optical discontinuity between areas

of different internal structures suggests that strain variations over a pyrrhotite grain are the possible cause of lamella zonation as well as of the uneven distribution of boxworks and composite lamellae. In other words, annealing and maturation of lamellae could be promoted or accelerated by deformation or anisotropic stress in a similar way to the effects of such stress on mpo exsolution as advocated by previous authors (Kübler and Lindqvist, 1979; Kissin and Scott, 1982; Lusk *et al.*, 1993).

Lamellar zonations, wedge-shaped composite ends, boxworks and composite lamellae, although present at Sabetjok, Gressdalen and Løkken, are less well-developed in the massive sulphide deposits. The reason for this could be the smaller grain size of their pyrrhotite relative to the nickel copper deposits; small grains are less sensitive to anisotropic stress (McClay and Ellis, 1983).

Origin of irregular intergrowths

In contrast to exsolution from hpo, hydrothermal alteration has been suggested to be another mechanism for mpo formation (Desborough and Carpenter, 1965). Japanese workers (Sugaki *et al.*, 1977) reported the synthesis of monoclinic pyrrhotite by reacting an aqueous solution of sodium or ammonium sulphide with hexagonal pyrrhotite at temperatures below 272°C. This implies that monoclinic pyrrhotite can be formed as a result of hydrothermal alteration. Monoclinic pyrrhotite that occurs along grain boundaries, cracks and veinlets should at least partly be a result of replacement of earlier hexagonal pyrrhotite by sulphur-rich and/or iron-poor hydrothermal (or metahydrothermal) fluids. Such replacive mpo can coexist with exsolution products in the same samples, as in the case of the Brattåsen, Sæterdalen and the Gjersvik deposits.

Carpenter (1974) interpreted the monoclinic rim around hexagonal grains from southeastern Tennessee and southwestern North Carolina to represent incipient weathering. However, the present authors do not think that weathering processes are essential to the formation of replacive mpo in the Norwegian sulphide ores because the samples showing typical replacive textures were taken from drill cores in Brattåsen and from fresh road cuts in Gjersvik and there is no sign of weathering on these samples.

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