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THE OCCURRENCE OF NICKEL-
ARSENIDES AND NICKEL-ANTIMONIDE
AT IGD LÚNGUAQ, IN THE
ILÍMAUSSAQ ALKALINE MASSIF,
SOUTH GREENLAND

BY

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WITH 13 FIGURES IN THE TEXT
AND 4 PLATES

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Abstract.

Ore minerals are rare in the alkaline rocks of the Ilimaussaq massif in SW. Greenland. At the locality of Igdlúnguaq on the North coast of the Tunugdliarfik fjord accessory amounts of Ni-arsenides and Ni-antimonide have been found in natrolitized naujaite adjacent to an acmite-arfvedsonite vein. The latter is later than the naujaite but earlier than or contemporaneous with the neighbouring lujavrites. A brief description of the field occurrence and petrography of the ore-bearing naujaite and the acmitite-arfvedsonite vein is given.

The ore minerals in the naujaite occur in very small and fine-grained aggregates, which consist of galena, skutterudite, niccolite, breithauptite, maucherite, löllingite and gudmundite. Diablastic intergrowths are common. Niccolite-maucherite network or cell textures, skutterudite-breithauptite and niccolite-breithauptite myrmekites, maucherite-breithauptite myrmekites and breithauptite-maucherite myrmekites and bladed intergrowths, breithauptite-löllingite network or cell textures, maucherite-löllingite bladed intergrowths, breithauptite-gudmundite bladed intergrowths, as well as certain zonal and other textures are described and their significance discussed.

It is concluded that the observed assemblages of ore minerals and their characteristic textures have resulted from the modification at temperatures around 400° C of earlier formed ore minerals, which are thought to have included galena, skutterudite, niccolite and breithauptite. The acmite-arfvedsonite vein is believed to have been formed by replacement of the naujaite along a fracture or fault plane. It is thought that the modifications of the earlier ore minerals in the naujaite are genetically related, in both time and space, with the formation of the acmite-arfvedsonite vein which is believed to be connected with the emplacement of the neighbouring lujavrites. It is suggested that fracturing of rocks, recementation of cracks and fractures, and the circulation of pneumatolytic or hydrothermal fluids having differential dissolving power for As, Sb, S and other components in the system of ore minerals, may have caused significant changes in the partial vapour pressures of these components. Such changes in partial vapour pressures of As, Sb and S are regarded as the main factors controlling the mineralogical and textural modifications of the ore minerals. The zeolites and other low-temperature minerals in the acmite-arfvedsonite vein were formed at a late stage of the vein formation, when temperature had already declined appreciably.

With regard to the origin of the ore minerals no conclusive arguments can be presented. Field evidence seems to favour the possibility that the components of the ore minerals have been completely or partly introduced into the naujaite as a result of hydrothermal action connected with the emplacement of the lujavrites and with the associated acmite-arfvedsonite veins. However, the possibility cannot be excluded that the ore minerals were present in the naujaite as primary magmatic minerals, which were subsequently modified during the emplacement of the lujavrite.

I. INTRODUCTION

In the summer of 1957 one of the authors (H.S.) found small spots of niccolite and other ore minerals in the Ilímaussaq alkaline massif, South Greenland. These ore minerals have been found only at one locality, Igdlúnguaq on the north coast of the Tunugdliarfik fjord. They are so scarce that no more traces of ore minerals have been found during further visits to the locality in 1960 (O.I.S.) and in 1962 (H.S.).

The niccolite and associated ore minerals present so intricate structural and textural relations that we feel justified in presenting a description of these minerals notwithstanding their extreme rarity.

Oen Ing Soen has undertaken the ore microscopical examination of the material, while working for »Grønlands geologiske Undersøgelse« in Copenhagen; H. Sørensen the field work and petrographic examinations. The discussion of the paragenesis and origin of the ore minerals is a co-operative effort.

We are indebted to Mrs. M. DANØ, Mr. Ib SØRENSEN, Mr. Chr. HALKIER, Mrs. R. LARSEN and Miss G. HANSEN, all of the Mineralogical Museum of the University of Copenhagen, respectively for X-ray examination of some minerals, for spectrographic examinations, for the microphotographs, for drawing the diagrams and for typing the manuscript. Miss Else BREVAL assisted in the field. Mr. J. WATTERSON kindly corrected the English of the manuscript.

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II. GENERAL GEOLOGY

The Ilimaussaq alkaline massif is situated on the Tunugdliarfik fjord a few kilometres to the east of Narssaq. The massif was described in great detail by USSING (1912). A summary of the geology of the massif has been given by SØRENSEN (1958). A new geological map of the Ilimaussaq massif and a description of the map will be published by FERGUSON (1964).

The massif is composed of an older unbanded series of common rock types (augite syenite, essexite, nordmarkite and alkali granite) and of a younger banded series of uncommon per-alkaline rock types, foyaite and alkali granite. The per-alkaline rocks comprise sodalite foyaite, naujaite, lujavrite and kakortokite (USSING, op.cit.). These per-alkaline rocks form a peculiar sub-group of the nepheline syenites which has been termed agpaitic by USSING (cf. SØRENSEN, 1960).

The agpaitic rocks are very probably derived from an augite syenitic magma enriched in volatiles (SØRENSEN, 1958 and FERGUSON, 1964). They were formed in two stages. In the first stage sodalite foyaite and naujaite crystallized in the upper part of the magma chamber while the kakortokites were formed in the lower part of the chamber. The lujavrite intruded these rocks during the second stage and according to FERGUSON (op.cit.) it occupies an intermediate position between naujaite and kakortokite. Veins of lujavrite cut the naujaite and fragments of the latter rock are enclosed in the larger masses of lujavrite.

Only the naujaite and the lujavrite will be considered in the present paper.

The *naujaite* is a very coarse-grained nepheline syenite characterized by peculiar poikilitic textures. Small crystals of sodalite are enclosed in large anhedral of microcline, eudialyte, ægirine and arfvedsonite. The sodalite crystals are often so closely spaced that the above-mentioned minerals are reduced to a thin interstitial network. Nepheline occurs as stout prisms. The naujaite may display an irregular banding. Accessories are ænigmatite, rinkite, sphalerite and others; secondary minerals are analcime and natrolite.

The *lujavrite* is a fine-grained melanocratic rock which often shows a pronounced lamination and even schistosity. The mafics make up

30–50 % of the rock. Two varieties have been distinguished by USSING (op.cit.), a green one rich in ægirine and a black one rich in arfvedsonite. The light coloured minerals are microcline and/or albite, nepheline and sometimes sodalite. Small crystals of eudialyte make up 2–10 % of the lujavrite. The acicular grains of arfvedsonite and ægirine, the laths of feldspar, the nepheline prisms and the eudialyte plates lie within the planes of lamination. Accessories are sphalerite, schizolite, steenstrupine, monazite and others; secondary minerals are analcime, natrolite, ussingite and katapleite.

The naujaite contains zoned pegmatites rich in eudialyte and other more irregular pegmatitic masses.

The naujaite and its pegmatites are cut by several types of *late veins*: green veins composed of felted ægirine, brown and black veins rich in acmite and arfvedsonite, and light coloured veins composed of one or more of the minerals albite, analcime, natrolite, sodalite and ussingite. These late veins are often rich in steenstrupine, britholite, sphalerite, schizolite, lepidolite, epistolite, pyrochlore and other rare minerals. Eudialyte is generally lacking. The late veins have never been observed in the lujavrite but they may be contiguous with lujavrite veins in fractures in the naujaite. According to SØRENSEN (1962) the green veins are slightly earlier than the lujavrite, marking the opening of the second phase of agpaiteic magma activity in Ilímaussaq. The brown and black veins are contemporaneous with or slightly earlier than the lujavrite. The light coloured veins are generally younger than the lujavrite being connected with the latest stages of crystallization of the latter rock.

III. ORE MINERALS IN THE ROCKS OF THE ILÍMAUSSAQ MASSIF

Ore minerals are common in the unbanded rocks of the complex. Thus iron-oxides, in part titaniferous, are common in the essexite and augite syenite, while pyrite and chalcopyrite have been observed along fractures in the essexite.

The agpaitic rocks are poor in ore minerals. Iron-oxides are practically absent and this in spite of the high iron content of some of the rocks, for instance about 10 % $\text{FeO} + \text{Fe}_2\text{O}_3$ in the lujavrite. The iron is, however, contained in silicate minerals.

Rust coloured spots in the naujaite are quite common, but have so far been regarded as weathered silicate minerals. However, in the summer of 1961 blasting was undertaken in the naujaite at the head of Kangerdluarssuk in order to obtain unweathered specimens. At this locality it was seen that the rust coloured spots are caused by the alteration of small aggregates of sulphides up to half a centimetre across. The aggregates of sulphides are composed mainly of pyrrhotite but small amounts of stannite and polybasite-pearceite occur in association with the pyrrhotite. The stannite shows a finely developed cross-hatched microcline-like twinning, which according to RAMDOHR (1960, p. 510–512) indicates that the mineral originally crystallized in its high-temperature cubic form.

Traces of löllingite have been reported by FLINK (1898, p. 245) from Naujakasik.

Small flakes of molybdenite have occasionally been observed in the naujaite and in the naujaite pegmatites.

The kakortokites locally contain small amounts of pyrrhotite and galena, which appears from an examination by John FERGUSON and one of the writers (O.I.S.).

Small grains of galena, less than one millimetre across, occur in some of the late analcime-rich veins. These grains are associated with steenstrupine and pseudomorphs after eudialyte (composed of katapleite, pyrochlore, analcime and other minerals).

Sphalerite is the only common ore mineral in the agpaitic rocks. It is a common accessory mineral in the lujavrites and also occurs in

the naujaite, naujaite pegmatites and the late veins. The pegmatites and the albite-rich late veins contain yellow or light brown grains of sphalerite which are several centimetres across. A sample of sphalerite from the late albititic vein at Tugtup agtakórfia contains 5.40 mol per cent FeS which corresponds to a temperature of formation of 170°C (the FeS-ZnS thermometer, KULLERUD, 1953, p. 129). However, it should be pointed out that this sphalerite has not been formed in equilibrium with other iron-bearing minerals; consequently, the temperature of formation indicated by this method should be regarded with some reservation. In some cases, the sphalerite is partially altered into a fine-grained aggregate of sheaf-like hemimorphite.

IV. THE OCCURRENCE OF NICCOLITE AND OTHER ORE MINERALS AT IGDLÚNGUAQ

The small point of Igdlúnguaq on the north coast of the Tunugdliarfik Fjord is situated in the *breccia zone* of USSING (1912, p. 36), that is, a zone composed of lujavrite with inclusions of naujaite (cf. fig. 3). Some naujaite inclusions are rotated and tilted and are cut by lujavrite veins. The breccia zone may therefore be regarded as an intrusion breccia on the border between the upper naujaite and the lower lujavrite (cf. SØRENSEN, 1962).

The naujaite of this locality is rather light coloured and contains some analcime and natrolite. The poikilitic texture is generally well developed. A banding consisting of an alternation of dark ægirine-rich bands, reddish-coloured eudialyte-rich bands and light coloured microcline-rich bands is locally developed.

The lujavrite is of the black, arfvedsonite-rich type and contains inclusions of naujaite in all stages of digestion. Lujavrite in contact with naujaite, as for instance where thin lujavrite veins cut the naujaite, is enriched in analcime and arfvedsonite and may contain poikilitic grains of steenstrupine.

The marginal zones of the large inclusions of naujaite are often strongly analcitized. Eudialyte has disappeared from these zones, and steenstrupine occurs instead. The analcime-rich parts of the naujaite inclusions have in some cases been mobilized, forming analcime-steenstrupine veins which cut the enclosing (and younger) lujavrite.

The naujaite is traversed by a number of thin veins (cf. fig. 2 and SØRENSEN, 1962). These veins do not cut the lujavrite but are occasionally contiguous with lujavrite veins occupying the same fissures in the naujaite. Three types of veins may be distinguished:

1. *Green veins* composed of felt-like ægirine, microcline, steenstrupine and a few other minerals. These veins often occur in zones of deformation in the naujaite. The veins are in some cases partially replaced by lujavrite and they are thus earlier than that rock.

2. *Brown or black veins* rich in acmite and/or arfvedsonite. They may have coarse-grained patches rich in analcime, natrolite, sodalite,

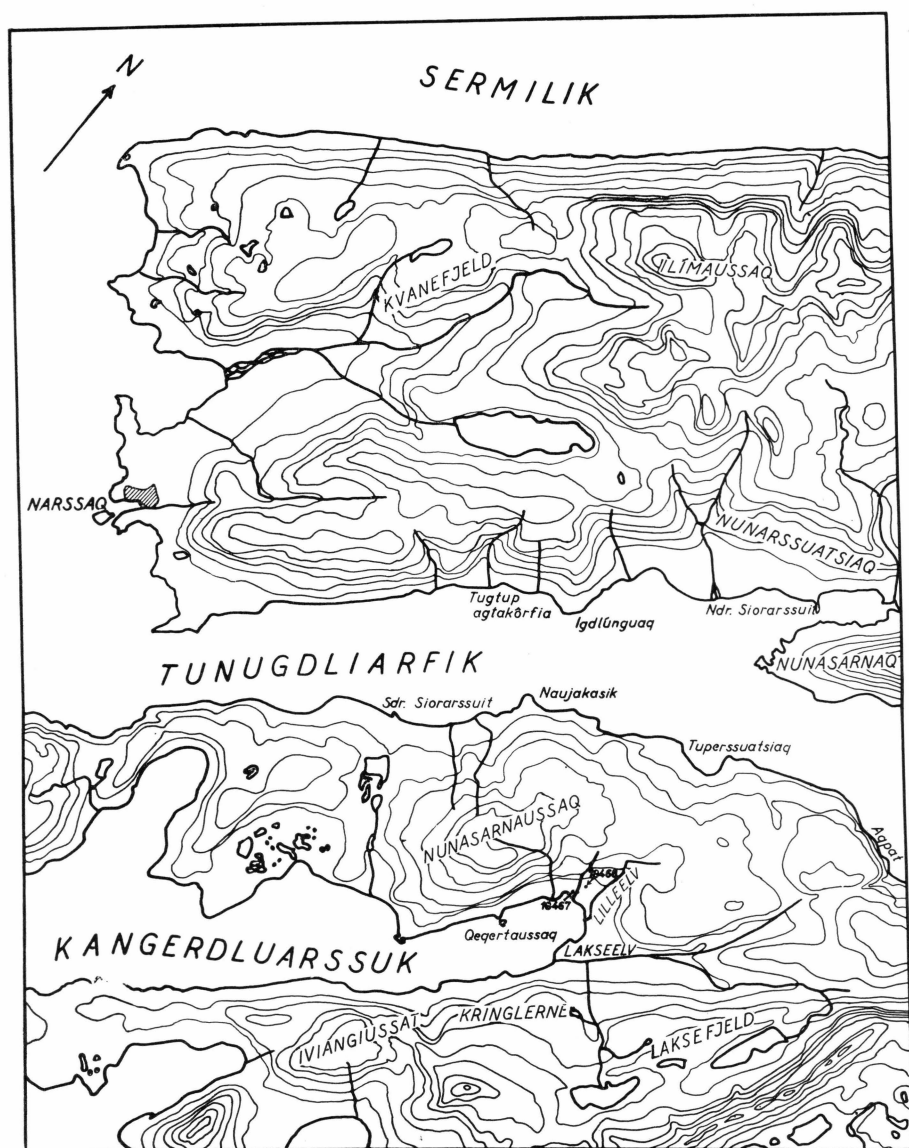


Fig. 1. Map showing the localities mentioned in the text. Scale 1:143,000; equidistances 100 m. The map is based on a preliminary 1:50,000 map kindly supplied by the Geodetic Institute, Copenhagen (Copyright the Geodetic Institute). Geological maps are to be found in USSING (1912), SØRENSEN (1958) and FERGUSON (1964).

steenstrupine and a number of other minerals. The veins are sometimes contiguous with lujavrite veins. Naujaite bands cut by these veins may be slightly displaced and slickensides are sometimes developed in the border zones of the veins. The acmite and arfvedsonite of these veins

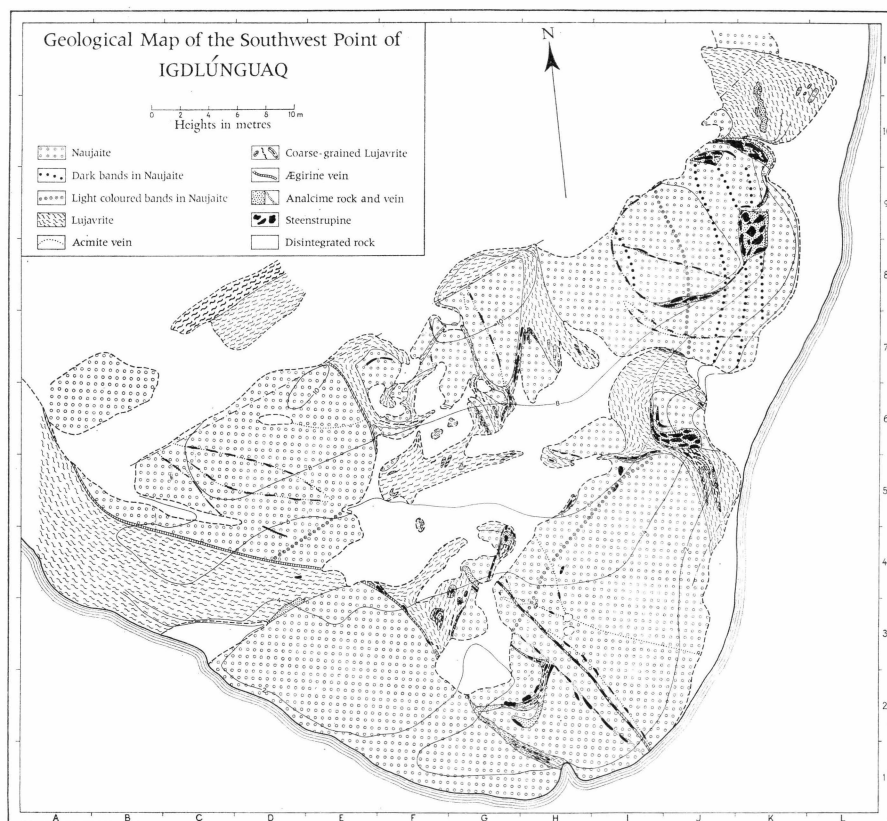


Fig. 2. Geological map of the Southwest point of Igdlúnguaq. (Reproduced from SØRENSEN, 1962).

are regarded as having been formed simultaneously with the *mise en place* of the lujavrite. The patches containing analcime, etc. are most probably slightly later than the *mise en place* of the lujavrite and are formed partly from the residual liquids expelled from the lujavrite.

3. *Light coloured veins* composed of the minerals found in the coarse-grained patches of the above-mentioned brown and black veins. The light coloured veins often have concentrations of acmite and steenstrupine along the borders and are regarded as special cases of the light coloured, coarse-grained patches of the brown and black veins.

The following rare minerals have been found in the late veins: steenstrupine, sphalerite, schizolite, monazite, pyrochlore, igdloite, neptunite, chkalovite, britholite, lepidolite and thorianite. Eudialyte does not occur but pseudomorphs after it are fairly common, and consist of katapleite, pyrochlore, neptunite, ægirine/acmite, monazite, analcime and other minerals.

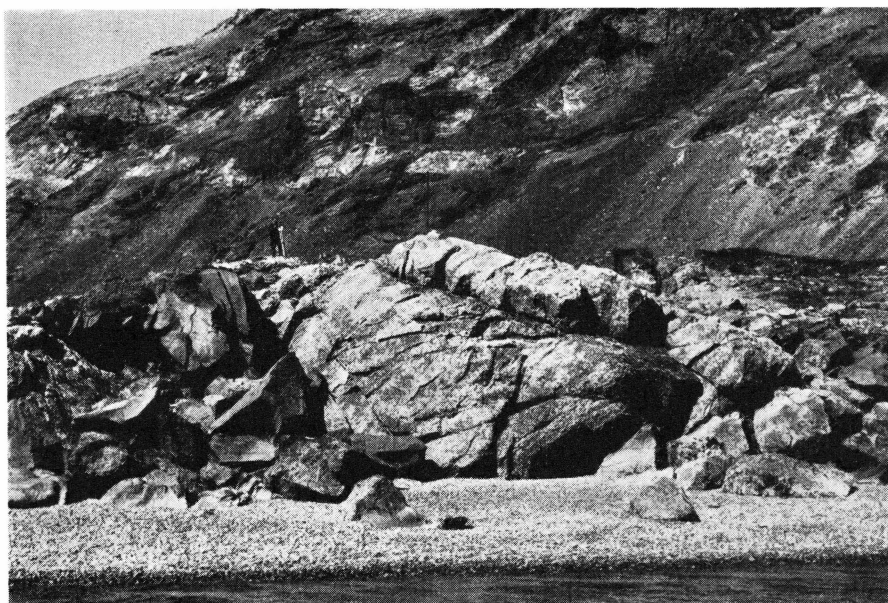


Fig. 3. The southwest point of Igdlunguaq as seen from the east. The distinct slightly inclined fracture in the centre of the photograph (dipping towards the lower right corner) is the brown vein adjacent to which the ore minerals described in the text occur. (Reproduced from SØRENSEN, 1962).

The mineral associations of the late veins indicate that the veins were formed under pegmatitic to hydrothermal conditions. A detailed description and discussion of these veins has been given by SØRENSEN (1962).

The niccolite and associated ore minerals occur in naujaite adjacent to an acmite-arfvedsonite vein at the coordinate position J-K, 8-9 in fig. 2. The vein is flat-lying with an easterly dip of about 30° . It is for a short distance parallel to the surface of the ground which explains its apparent thickness in fig. 2 (cf. figs. 3 and 4). The naujaite is banded and the vein is in contact with a eudialyte-rich band.

The acmite-arfvedsonite vein is a few centimetres thick and is composed mainly of black and brown fine-grained aggregates of acmite and/or arfvedsonite with light coloured, coarse-grained patches.

The brown parts of the vein often have a greenish tint and are composed of tiny needles of acmite with a subordinate amount of ægirine. The grains are sometimes arranged with their longest dimension parallel to the strike of the vein, but generally no preferred orientation can be seen. A few large black prisms of arfvedsonite, up to a few centimetres in length, are aligned parallel to the strike of the vein.



Fig. 4. The brown vein seen in fig. 3. The acmite-arfvedsonite vein rock has been partly removed by erosion so that white areas of the underlying naujaite are seen.

The black parts of the vein, in addition to the above-mentioned large arfvedsonite prisms, contain streaks of fine-grained arfvedsonite intergrown with acmite. The streaks are arranged approximately perpendicular to the walls of the vein.

The light coloured, coarse-grained patches often show an outer zone of small crystals of steenstrupine. In places the acmite-arfvedsonite matrix of the vein is lacking and the steenstrupine crystals then occur in direct contact with the naujaite. Generally, however, a thin rim of acmite and/or arfvedsonite separates the steenstrupine from the naujaite.

Inside this marginal steenstrupine zone the coarse-grained patches have a thin zone of yellow sodalite, which surrounds a coarse-grained core composed of large grains of analcime and natrolite with a minor amount of the rare mineral chkalovite (SØRENSEN, 1962). The analcime is white, grey or green and it has a distinct cubic cleavage. The large prisms of natrolite are white or light blue. White masses of fine-grained natrolite also occur. Other macroscopic constituents of the coarse-grained patches are: arfvedsonite, patches of yellow pyrochlore, white streaks

of fine-grained igdloite associated with dark red neptunite and fine flakes of lepidolite.

Steenstrupine also occurs as small brown patches in the acmitic matrix of the vein.

Concentrations of small brown crystals of eudialyte locally occur along the contact between naujaite and vein. This eudialyte differs in colour from the red coloured eudialyte of the naujaite. The large eudialyte grains of the naujaite may also occur in direct contact with the vein. They are then brown in colour and are generally separated from the steenstrupine of the vein by a rim of acmite. Where steenstrupine forms the border of the vein the naujaite is rich in natrolite and devoid of eudialyte. Small impregnations of steenstrupine occur locally in the eudialyte-poor naujaite adjacent to the vein.

The nickel-arsenides and -antimonide occur as very small streaks and patches, a few millimetres long and generally less than one millimetre thick. They have been found only within a restricted area a few square centimetres in size. The ore minerals occur in cracks and along the cleavages of the sodalite and ægirine of the naujaite adjacent to the acmite-arfvedsonite vein. Patches of ore minerals also occur in a thin natrolite zone between the naujaite and vein.

V. PETROGRAPHY

A petrographic description of the various rock types found at Igdlúnguaq has been given by SØRENSEN (1962). In the present paper only the rocks which are of direct importance for the discussion of the ore minerals will be considered.

The Naujaite.

The naujaite is composed mainly of poikilitic grains of microcline, ægirine and eudialyte with numerous inclusions of sodalite.

The crystals of sodalite, which are up to one centimetre across, are rich in microlites of arfvedsonite and ægirine. They are generally unaltered but sometimes partly replaced by aggregates of fine-grained natrolite ("spreustein").

The eudialyte occurs as large grains, several centimetres across, and crowded with inclusions of sodalite, sometimes to such an extent that the eudialyte is reduced to a fine interstitial network between the sodalite grains. The grain boundaries of the eudialyte are generally irregular, but crystal faces are locally developed. In the latter case the eudialyte may be zoned with alternating zones of eudialyte and mesodialyte parallel with the crystal faces. Some secondary katapleite occurs in the eudialyte.

Ægirine forms large poikilitic grains which show evidence of deformation in the form of "columns" of slightly different optical orientation. The colour in thin section is green, but parts of the grains are brown and then consist of acmite (cf. SØRENSEN, 1962). Large areas composed of aggregates of fine-grained acmite enclose sodalite crystals in a poikilitic manner. Arfvedsonite is sometimes enclosed in the acmitic parts of the ægirine grains. Small individual prisms of arfvedsonite may have inclusions of acmite.

The large platy grains of microcline display the albitic chess board twinning which is so characteristic of the microclines found in agpaitic rocks (cf. SØRENSEN, 1962).

Large grains of nepheline with small inclusions of microcline and arfvedsonite are surrounded by fine-grained natrolite.

The Contact Zone between the Naujaite and the Acmite-Arfvedsonite Vein.

This zone is composed mainly of natrolite and analcime with inclusions of sodalite.

The natrolite occurs as large prismatic crystals, up to one centimetre long, and as aggregates of many small grains which are fairly equigranular.

Adjacent to the vein the large poikilitic ægirine grains of the naujaite are often deformed. Where the grains happen to occur at approximately right angles

to the vein walls the broken-off parts of the grains appear to have been dragged into a position parallel to the vein. In other cases the ends of the ægirine crystals abutting the vein pass into acmite crowded with small flakes of a brown mica. The ægirine and acmite parts of these grains show a uniform optical orientation.

The ægirine of the natrolitic contact zone and of the adjoining naujaite is intergrown with arfvedsonite in a way indicating that it has been partially replaced by the latter mineral. Furthermore, large grains of arfvedsonite of this border zone contain small inclusions of acmite which, in restricted areas, all have the same optical orientation. This indicates that the arfvedsonite replaces aggregates of acmite. This acmite can have green cores.

Concentrations of small eudialyte crystals occur in the contact zone along the vein. This eudialyte is of the same type as that of the lujavrites having irregular, vein-like areas of mesodialyte. The crystals are generally altered into rust pigmentation, katapleite and other secondary minerals which are the cause of the brown colour seen in the hand specimen.

Small rust coloured grains of steenstrupine are occasionally seen in the contact zone.

In one place aggregates of small rounded grains of an apatite mineral of britholitic type (cf. DANØ and SØRENSEN, 1959) have been observed in a groundmass rich in analcime. These grains are associated with small prisms of ægirine and arfvedsonite, the latter often forming homoaxial rims around the former.

The Acmite-Arfvedsonite Vein.

The major part of the vein is composed of acmite and arfvedsonite.

The acmite occurs in aggregates of small grains which often show well-developed prism faces. The grains generally show no preferred orientation but, locally, they may parallel the vein walls. The acmite often has central green patches of ægirine and contains abundant small flakes of a brown biotitic mica. The acmite aggregates have interstitial grains of arfvedsonite.

Arfvedsonite occurs as large prisms arranged parallel to the vein, as streaks composed of small needles and as large, apparently crushed grains of an appearance intermediate between that of the large grains and the streaks. The large prisms are in some cases seen to grade into crushed grains which again grade into the streaks. The latter are orientated at right angles to the walls of the vein and they were formed by deformation of the large prisms.

The large arfvedsonite prisms contain inclusions of acmite with flakes of the brown mica; the latter mineral also occurs in the arfvedsonite adjacent to the acmite inclusions. The streaks of fine-grained arfvedsonite have small interstitial fibres of acmite. Between the "units" of the crushed arfvedsonite grains there are also aggregates of acmite fibres.

The relationship between the acmite and the arfvedsonite of the vein may be interpreted as follows: Along a zone of deformation in the naujaite the ægirine has been deformed, crushed and partially substituted by acmite. This acmite forms the matrix of the vein. Large grains of arfvedsonite have partly replaced the acmite and are orientated parallel to the walls of the vein. Subsequently these prisms have been crushed and rolled-out into streaks almost perpendicular to the walls of the vein. The interstitial acmite in these streaks may have been formed by crushing of the acmite inclusions in the large arfvedsonite prisms, but there may also have been a new formation of acmite at this stage. Probably connected with the deformation in the zone now occupied by the vein is the formation of micro gash joints in

the sodalite adjacent to the vein. These gash joints are arranged along fractures approximately parallel to the vein.

In the *natrolite zone* between the acmite-arfvedsonite vein and the adjoining naujaite there are clusters of small grains of a brown mineral. It appears to form prismatic crystals with a distinct cleavage at right angles to the elongation. The mineral is optically negative with a very small optic axial angle. The absorption scheme is of the type seen in tourmaline and astrophyllite: the darkest colour (brownish green to dark) appears when the shortest dimension of the crystals is orientated parallel to the vibration direction of the polarizer, while in the reverse position lighter colours (brown or reddish brown) appear. The "prisms" are connected with stages of transition into the above-mentioned flakes of a biotitic mica and the mineral is therefore most probably biotite.

The acmite-arfvedsonite rocks of the vein have a minor amount of interstitial analcime and natrolite. Accessories are steenstrupine, neptunite, britholite and, in rare cases, small eudialyte crystals of lujavritic type. There may also be pseudomorphs after eudialyte which are rich in plates of katapleite.

The coarse-grained, light coloured parts of the vein are made up of large grains and aggregates of small grains of natrolite, large analcime grains, large sodalite grains, schizolite, lepidolite, arfvedsonite, acmite, neptunite, pyrochlore, igdloite, chkalovite and steenstrupine.

The large grains of natrolite may display an irregular extinction.

The sodalite, which is yellow in the hand specimen, is free of dark microlites and thus differs from the sodalite of the naujaite. It contains 0.55% S and may be termed hackmanite (SØRENSEN, 1962).

Igdloite and pyrochlore occur along fractures and are associated with neptunite and a red micaceous mineral. They form fine-grained streaks and aggregates. The pyrochlore may occur as rims to the steenstrupine crystals.

Steenstrupine occurs in the contact between the acmite-arfvedsonite rock and the coarse-grained rock. It has irregular boundaries with the arfvedsonite and acmite, whereas crystal faces are developed towards the analcime, natrolite and sodalite of the patches. The steenstrupine contains inclusions of acmite and arfvedsonite. The arfvedsonite adjacent to and enclosed in the steenstrupine is often separated from the latter mineral by a rim of acmite.

The steenstrupine crystals have yellowish grey, isotropic cores and strongly coloured anisotropic marginal zones. The steenstrupine may be associated with remnants of pseudomorphs after eudialyte which are mainly composed of a pigimentary substance in a groundmass of analcime.

The coarse-grained rock has white, fine-grained patches composed of very fine-grained natrolite and some analcime with inclusions of strongly deformed microcline.

The ore minerals to be described in this paper occur in the natrolite-rich contact zone between naujaite and acmite-arfvedsonite vein. The natrolite penetrates the sodalite of the naujaite and contains inclusions of that mineral.

Several small nests of ore minerals occur as lines of inclusions in the natrolite. They also occur along the cleavages of ægirine and arfvedsonite and in cracks in the sodalite. The minerals surrounding the ore grains are rust coloured.

The ore in the fractures in sodalite is often separated from the latter by a thin rim of natrolite. Analcime may also replace the sodalite.

The ore-bearing natrolite rock contains small grains of britholite, schizolite, arfvedsonite, ægirine, steenstrupine and remnants of large grains of naujaitic ægirine.

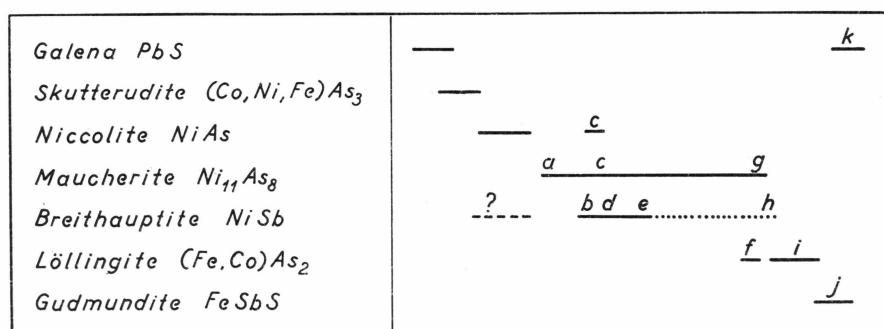


Fig. 13. Diagrammatic representation of the mineralogical and textural sequence of Ni-arsenides and associated ore minerals in a naujaite of the Ilimaussaq complex, South Greenland.

a. Beginning of partial dissociation of niccolite yielding maucherite and As-vapour. a–c. Niccolite-maucherite networks and zonal textures; recrystallization of niccolite and probably also of some skutterudite (not indicated in the figure). b–d. Crack-fillings of columnar breithauptite; skutterudite-breithauptite and niccolite-breithauptite myrmekites; cementation of cracks and other openings with breithauptite and possibly other minerals. c. Niccolite reaction zones along skutterudite-maucherite boundaries; some fine-grained maucherite idiomorphs and beginning of formation of maucherite-breithauptite myrmekites. d–e. Coarsely granular breithauptite aggregates. c–g. Maucherite-breithauptite myrmekites and breithauptite-maucherite myrmekites and bladed intergrowths. e–h. Local recrystallizations of breithauptite into finer-grained aggregates inside the maucherite-breithauptite myrmekites and around breithauptite-löllingite intergrowths. f. Segregation of löllingite from breithauptite, breithauptite-löllingite network and cell textures. i. Segregation of löllingite from maucherite, maucherite-löllingite bladed intergrowths. j. Segregation of gudmundite from breithauptite, breithauptite-gudmundite bladed intergrowths and gudmundite rims. k. Rims of late galena.

to increase the volume of the reacting system in the ore nests (radial expansion cracks), whilst the volume of the solid phases decreased or remained constant (concentric cracks).

According to HEWITT (1948, p. 415) partial dissociation of niccolite occurs around 450°C, when NiAs is heated in sealed tubes. YUND (1959, pp. 148–150), reporting on the Ni-As system, has made no mention of the dissociation of niccolite, but has confirmed earlier records of the incongruent melting of maucherite into niccolite plus liquid at 829°C. These data suggest that temperatures between roughly speaking 450° and 800°C have been reached during the formation of the ore minerals. However, a consideration of the mineral paragenesis in the associated zeolite-bearing acmite-arfvedsonite vein does not indicate such high temperatures. The possibility that the dissociation of niccolite in the rocks considered might have occurred at lower temperatures than as suggested above may be conceived on the following grounds:

i) The experiments by HAWLEY and HEWITT were carried out over a limited time. It may be expected that with sufficient time available the partial dissociation of niccolite may be accomplished at lower temperatures when other conditions are favourable.

ii) In analogy with results obtained by KULLERUD and YODER (1958), according to which the temperature at which pyrite breaks down to pyrrhotite and sulphur is lowered by the presence of water, a similar lowering of the dissociation temperature of niccolite due to a water-rich environment may be suspected.

iii) It is possible that fracturing and the circulation of water-rich fluids capable of dissolving arsenic, may have caused a lowering of arsenic vapour pressures, which would accelerate the breakdown of niccolite into maucherite (HAWLEY and HEWITT's experiments refer to a closed system).

Experiments by VIGOUROUX (1908) and HOLMES (1947, p. 318) suggest that in an open system the As-content of Ni-arsenides decreases with increasing temperatures, so that the skutterudite-niccolite-maucherite sequence, as shown, e. g., by the niccolite reaction zones along the skutterudite-maucherite boundaries, may well be explained by a formation under conditions of increasing temperature. However, the effect of a variation in pressure should also be considered since a decrease in total pressure most probably will favour the formation of the most As-poor minerals.

ROSEBOOM (1959) has indicated that at lower temperatures in the Co-Ni-As system the pair niccolite-skutterudite seems to be the stable one relative to the pair safflorite-rammelsbergite. The lack of the latter pair in the present paragenesis of ore minerals may indicate that the temperatures at which this pair is stable were not attained before and during the formation of niccolite and skutterudite.

The complete preservation of the many intricate intergrowths of the ore minerals indicate that they have not been subjected to modifying influences after the formation of these textures. As mentioned above, the formation of the acmite-arfvedsonite vein commenced at temperatures of the order of 400°C and at rather high total pressures. The subsequent precipitation of analcime, natrolite, steenstrupine, etc. took place at lower temperatures and pressures. Two different ways of ore formation may then be possible:

i) The ore minerals were present as primary constituents of the naujaite. These ore minerals became modified by pneumatolytic-hydrothermal processes which accompanied the emplacement of the lujavrite.

ii) The ore minerals were deposited in fractures in the naujaite by late fluids expelled from the crystallizing lujavrite magma. As discussed by SØRENSEN (1962) several stages of expulsion of fluid phases may have occurred. Following FERSMAN it may then be suggested that the phases expelled at the highest temperatures and pressures were enriched in the most volatile components of the magma such as, for instance, arsenic and antimony (cf. also LOVERING, 1961). It is therefore not unreasonable to assume that there has been an early precipitation of nickel arsenides in a fracture in the naujaite. The later phases of fluids expelled from the lujavrite deposited the steenstrupine, analcime, natrolite, etc. in the fractures. During this process the first-formed ore minerals were modified. According to the experimental data available this may have taken place at not very low temperatures, i. e. earlier than the final crystallization of natrolite.

In the case that the ore minerals are assumed to have been present as original constituents of the naujaite, the early paragenesis presumably consisted of galena, skutterudite, niccolite and breithauptite, while other minerals such as gersdorffite may also have been present (HAWLEY, STANTON and SMITH, 1961, have described niccolite-maucherite intergrowths resulting from the breakdown of gersdorffite). Subsequent pneumatolytic-hydrothermal activity connected with the intrusion of the lujavrite caused the partial dissociation of higher arsenides into lower arsenides and arsenic vapour. Skutterudite and niccolite apparently recrystallized *in situ*; the niccolite yielded some maucherite, which is partially held in the niccolite-maucherite network intergrowths and partially segregated as rims around these intergrowths, while the skutterudite was partially replaced by niccolite. At approximately the same stage breithauptite began to recrystallize and the mineral was redeposited as columnar aggregates in the cracks formed as a consequence of the volume changes attending the partial dissociation of niccolite. The latter crack-fillings, as well as the formation of some skutterudite-breithauptite myrmekitic intergrowths as a result of replacement by breithauptite, indicate that, unlike the arsenides, which recrystallized *in situ*, the breithauptite was at least partially remobilized during its recrystallization.

If, on the other hand, it is assumed that the ore minerals have crystallized from the late fluids expelled from the lujavrite magma, an overlapping subsequent deposition of galena, skutterudite and niccolite may be conceived during the first stages of a crystallization at rather high temperatures and pressures. The partial dissociation of

the early-formed higher arsenides took place as soon as the appropriate temperatures and pressures were reached. The crystallization of breithauptite commenced at about this time.

Both of the possibilities considered above allow for the assumption that after the formation of skutterudite and niccolite and their partial breakdown and recrystallization *in situ*, the breithauptite, being still in the process of crystallization or recrystallization, behaved as a relatively more mobile (soluble) substance than the arsenides. A removal of parts of the antimony from the system at this stage is consistent with the preponderance of new arsenides over antimonide during the subsequent development.

It is suggested that after the cementation of the cracks with breithauptite and possibly other minerals (silicates), further (re)crystallization in the ore nests took place in a partly isolated system, permitting only very slow escape of volatiles. It is thought that under these circumstances the liberation of arsenic vapour, the partial removal of antimony and the (re)crystallization of breithauptite may cause a significant increase in the partial pressure of arsenic, so that with temperatures beginning to fall, the partial pressure of arsenic became in excess of that required for equilibrium conditions. As a consequence of this, arsenic tended to enter into the solid phases, accelerating the formation of maucherite at the expense of breithauptite. The formation of some maucherite idioblasts had presumably already been initiated contemporaneously with the (re)crystallization of breithauptite. The development of niccolite reaction zones at the expense of skutterudite along boundaries of the latter mineral with some maucherite-breithauptite myrmekites may indicate that the initially high temperatures still prevailed during the first stages of maucherite blastesis. However, the bulk of the abundant maucherite-breithauptite myrmekites, as well as the breithauptite-maucherite myrmekites and bladed intergrowths, apparently developed by replacement of breithauptite at a relatively late stage, presumably when temperatures had decreased appreciably. It is believed that the same critical conditions of excess arsenic pressures, persisting during further cooling, also caused the segregation of iron in the form of the diarsenide löllingite from the breithauptite (breithauptite-löllingite networks or cell textures) and from the maucherite (maucherite-löllingite bladed intergrowths).

During the modifications of the earlier formed minerals, a large proportion of the galena and possibly other sulphides were presumably dissolved and removed, so that galena is now found only as a few rounded, relic inclusions, mostly in (re)crystallized breithauptite. How-

ever, a part of the dissolved galena was apparently redeposited at a late stage of cooling as a late generation of galena, whilst the late formation of breithauptite-gudmundite bladed intergrowths and gudmundite rims may also be interpreted as a manifestation of the increasing sulphur concentrations in the residual phases.

The inferences made from a consideration of the characteristic ore textures may be briefly summarized as follows:

i) The present paragenesis of ore minerals and the ore textures are the result of a modification of an early-formed ore mineral assemblage.

ii) Changes in the partial vapour pressures of As, Sb, S and possibly other components were important factors controlling the mineralogical modifications at elevated temperatures as well as during the subsequent cooling.

iii) Fracturing of the rocks, later recementation of cracks, the circulation of hydrothermal fluids having differential dissolving powers for As and Sb and other elements, may be suggested as possible causes for the occurrence of changes in partial vapour pressures of the different elements.

iv) The ore minerals were formed earlier than the low temperature minerals of the acmite-arfvedsonite vein.

5. On the origin of the ore minerals.

The ore minerals described in the present paper occur in the border zone between naujaite and a late acmite-arfvedsonite vein which cuts the naujaite. The close association of the ore minerals with natrolite and small ægirine prisms, which occur in this border zone, suggests a relation between the ore minerals and the formation of the late vein. Textural evidence discussed in the preceding section indicates that the observed assemblage of ore minerals has been formed by recrystallization, partial mobilization and further modification of an earlier formed assemblage of ore minerals, which probably comprised galena, skutterudite, niccolite, breithauptite and possibly other minerals. A combined consideration of the geologic, petrologic and ore microscopic observations allow the following suggestions regarding the origin of the ore minerals:

i) The original assemblage of ore minerals crystallized as primary magmatic constituents of the naujaite. Later hydrothermal and pneumatolytic (?) action occurring in connection with the formation of the lujavrite and its accompanying late veins, caused modifications of the original ore minerals and their textures. Such an origin of the ore mine-

erals is supported by the local occurrence of minerals such as pyrrhotite, stannite and polybasite in the naujaite. These minerals are not alien to parageneses of Ni-Co-Fe-arsenides. However, it is not yet known whether the pyrrhotite, etc. are primary minerals in the naujaite or of later origin. If these ore minerals are primary they have not been subjected to late modifications of the type seen in the nickel-arsenides at Igdlúnguaq. For example, the stannite found in the naujaite shows a finely developed lamellar twinning parallel to (001) and (100). According to RAMDOHR (1960, p. 510–512) this cross-hatched microcline-like twinning in stannite is formed by the conversion of a high-temperature cubic form of stannite into its normal low-temperature form. Although the experimentally determined conversion temperature of 600°C is to be considered as improbably high (RAMDOHR, 1960, p. 512), it is obvious that the mineral must have crystallized at appreciably elevated temperatures, approaching those of magmatic conditions, and that the preservation of the conversion lamellae indicates the absence of further modification subsequent to the gradual cooling. Thus it seems that if the stannite is a primary magmatic mineral no marked temperature fluctuations occurred during the post-magmatic cooling stages of the naujaite. The modifications in the nickel-arsenides must then be ascribed to later actions in connection with the formation of the acmite vein.

ii) The ore minerals have been introduced into the naujaite as a result of pneumatolytic-hydrothermal action connected with the emplacement of the lujavrite. Hitherto, nickel-arsenides have not been found in the lujavrites of the complex, but, as stated on p. 39, the lujavrite is certainly nickel-bearing.

iii) Since it is known that nickel and cobalt are present in small traces in some of the silicate minerals of the naujaite, the possibility that the nickel, cobalt and iron of the ore minerals have been derived from these sources should also be considered. It is conceivable that these trace elements may have been leached out of the naujaite minerals adjacent to the fractures through which the late fluids percolated. The latter fluids may have introduced the arsenic and antimony of the ore minerals.

In conclusion it may be stated that the ore microscopic observations are consistent with all the three above suggested interpretations. The many diablastic and symplectitic intergrowths seem to characterize the metasomatic alteration of earlier formed ore minerals. The field observations are best accounted for by a combination of hypotheses 2 and 3, but as hypothesis 1 also falls within the possibilities, the authors prefer to postpone a final statement until a systematic study of the ore minerals in the rocks of Ilimaussaq has been made.

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PLATES

Plate I.

Fig. 1. Aggregates of ore minerals consisting mainly of maucherite-breithauptite myrmekites and breithauptite. The ore minerals occur enclosed in natrolite and sodalite. (Black area). (1 nicol, 80 x).

Fig. 2. Network of very fine, intergranular maucherite veinlets and stringers in a finely granular niccolite aggregate. In this example the maucherite zone around the niccolite is surrounded by a breithauptite rim (appearing in the same shades of grey as niccolite), which do not show intergrowths with maucherite. The straight boundary between the niccolite-maucherite aggregate and the breithauptite rim in the lower part of the picture suggests that the granular texture of the niccolite is due to recrystallization of a formerly subhedral or euhedral niccolite crystal. (1 nicol, 430 x).

(Chr. HALKIER phot.).

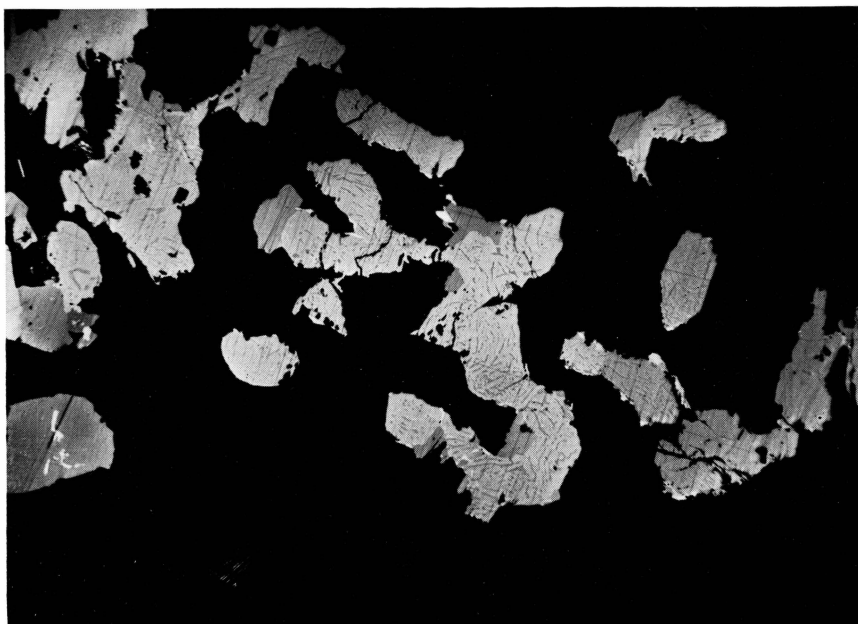


Fig. 1.



Fig. 2.

Plate II.

Fig. 1. Rims and veinlets of breithauptite with columnar textures (ruled in fig. 7) developed around an aggregate consisting of niccolite and maucherite (stippled area in fig. 7). The latter aggregate consists mainly of granular, apparently recrystallized niccolite (appearing in the photograph in shades of light and dark grey to black), veined by very fine intergranular maucherite veinlets and surrounded by a narrow, irregular maucherite zone. The maucherite appears in an intermediate tint of grey, contrasting very poorly with the niccolite. However, the textural relations are visible in that part of the photograph marked with a cross in fig. 7; here the niccolite is almost white, while the maucherite veinlets appear darker grey. Note the typical outline of the niccolite-maucherite aggregate, strongly suggestive of the former existence of a euhedral niccolite crystal. (2 nicols, 240 x).

Fig. 2. Subhedral maucherite-breithauptite myrmekites. The maucherite grains M_1 and M_2 (see key on fig. 9) probably have a twin relationship. The breithauptite inclusions in M_1 have the same optical orientation as that of the adjoining breithauptite grain B_1 , which appear dark and therefore cannot be distinguished in the photograph (compare fig. 9); those in M_2 , M_3 and M_4 have orientations which are obviously related to that of the breithauptite "superindividual" Bs. The latter consists of a group of breithauptite grains, the orientation of which varies so little that in the present case it may be suggested that they formerly formed a single homogeneous grain. The myrmekite M_5 has few inclusions, which are not easily seen in the photograph owing to the unfavourable optical orientation of the grains. The black inclusions in the ore minerals are transparent gangue material. Note the straight boundaries and clean rims of the myrmekitic crystals, suggesting that no corrosion of maucherite by breithauptite has taken place. (2 nicols, 160 x).

Fig. 3. Rims and radial veinlets of breithauptite with columnar texture (ruled in fig. 8). The central part of the ore nests consists of fine-grained niccolite (stippled in fig. 8). An intermediate zone of maucherite is well developed, but it does not appear clearly on the photograph; the maucherite zone is shown in fig. 8 (crosses). (2 nicols, 160 x).

Fig. 4. The maucherite-breithauptite myrmekite in the central part of the figure tends to occur interstitially in a comparatively coarse-grained breithauptite aggregate (various shades of grey). The breithauptite inclusions in the myrmekite have the same optical orientation as the breithauptite grain just below the myrmekite. Löllingite blades (white) appear as parallel lamellae along two directions in the maucherite and concentrated along its borders. Small, dark grey to black spots in the upper right hand part of the picture are inclusions of galena, which occur as rounded drops when enclosed by breithauptite, but the rounded outlines of which appear to have been destroyed by corrosion when occurring in contact with maucherite or löllingite (as in this picture); these relations suggest the formation of the latter minerals to be later than that of the breithauptite. (1 nicol, 160 x).

(Chr. HALKIER phot.).

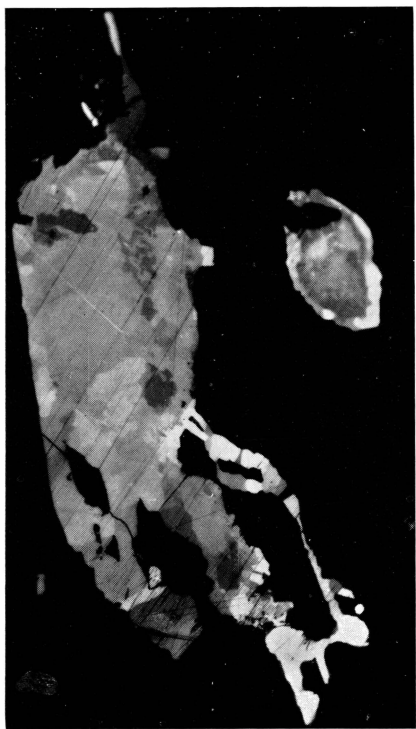


Fig. 1.

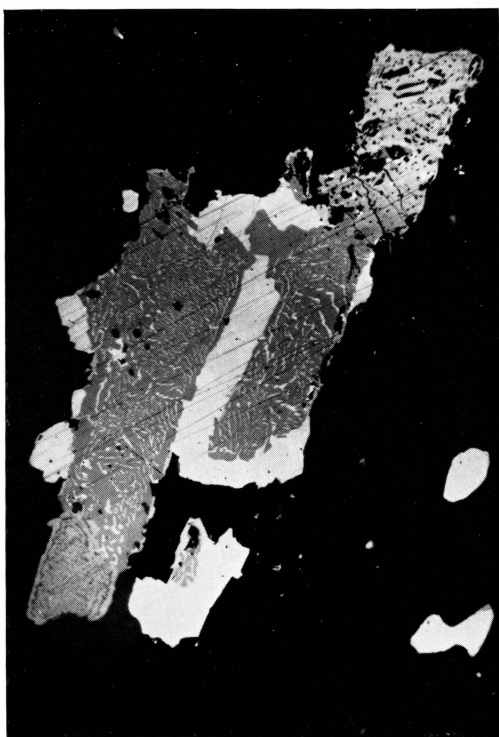


Fig. 2.

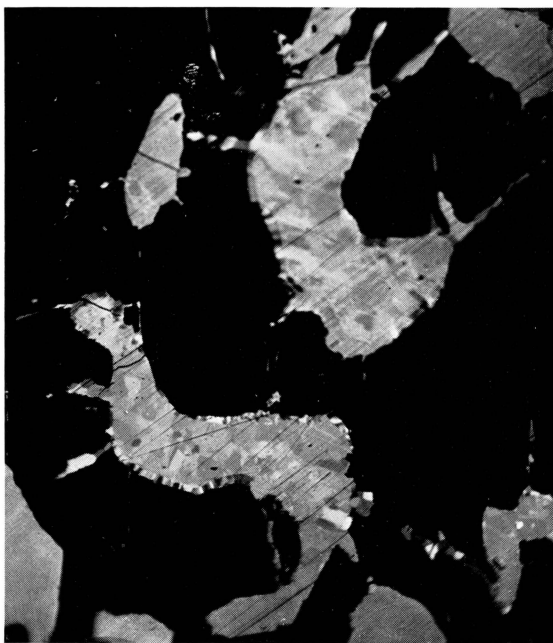


Fig. 3.

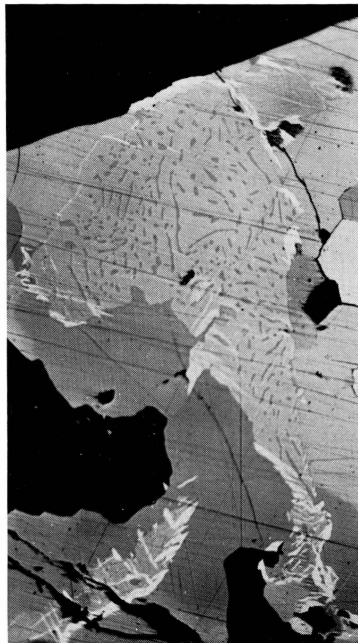


Fig. 4.

Plate III.

Fig. 1. Maucherite-breithauptite myrmekites and a bladed intergrowth of breithauptite and maucherite. In the photograph the breithauptite appears dark grey to black, the maucherite light grey to white. The maucherite blades (left of the centre of the photograph) occur enclosed within a breithauptite grain, the optical orientation of which differs so little from that of neighbouring breithauptite grains, that a "superindividual" is defined. The breithauptite inclusions within each of the maucherite-breithauptite myrmekites are generally uniformly oriented and an adjoining breithauptite grain showing the same or approximately the same orientation can usually be seen. Some of the myrmekites show two groups of optically and spatially related inclusions (see fig. 10; upper right hand corner). The oriented löllingite blades in maucherite are sometimes faintly visible in the photograph. In fig. 10 the different directions of the striation indicate different optical orientations of breithauptite grains. "Superindividuals" are uniformly striated. The direction of broken lines in the maucherite-breithauptite myrmekites indicates the optical orientation of the breithauptite inclusions in the myrmekite. A relation between the orientation of the inclusions and those of breithauptite grains is clearly indicated (see also figs. 11 and 12). (2 nicols, 160 x).

Fig. 2. Longitudinal and cross section of euhedral maucherite-breithauptite myrmekites. The cross section is presumably of a twinned crystal. Note that in one part of the twinned crystal the breithauptite inclusions appear as black spots on the picture, whereas in the other part the inclusions appear as light coloured bars, showing the same optical orientation as the surrounding breithauptite grain. Note also the clean borders of the myrmekites and, furthermore, the faintly visible löllingite blades occurring in three directions in the twinned maucherite crystal. (2 nicols, 160 x).

Fig. 3. Breithauptite-maucherite myrmekite (large grey coloured grain with white inclusions) and maucherite-breithauptite myrmekite (white grain with grey inclusions in upper left side of picture) occurring closely associated. (2 nicols, 160 x).

Fig. 4. The upper part of the picture shows a breithauptite-maucherite myrmekite, in which the maucherite inclusions have an optical orientation similar to that of the skeletal grains of maucherite shown about the central part of the figure (maucherite appears black to dark grey in the photograph, breithauptite is white to dark grey). The lower part of the figure shows an aggregate of maucherite-breithauptite myrmekites. (2 nicols, 80 x).

(Chr. HALKIER phot.).



Fig. 1.

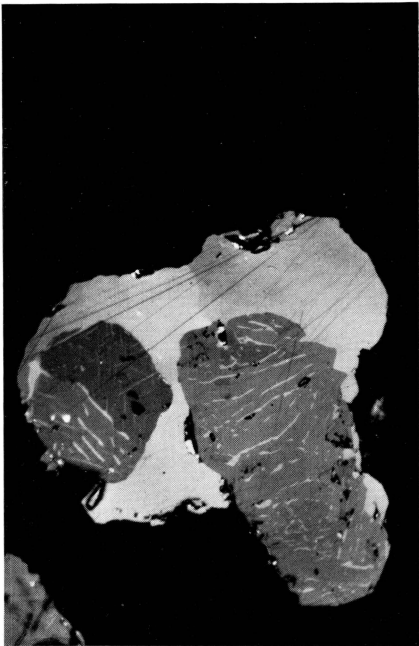


Fig. 2.



Fig. 3.



Fig. 4.

Plate IV.

Fig. 1. An aggregate of maucherite-breithauptite myrmekites in which the maucherite contains abundant oriented blades of löllingite. The optical orientations of the breithauptite inclusions and the directions of the löllingite blades change with the orientation of the maucherite host. Areas of granular breithauptite sometimes occur partly or completely enclosed in the myrmekitic aggregate. (2 nicols, 160 x).

Fig. 2. Gudmundite crystals (white) forming rims around very finely granular, presumably recrystallized breithauptite aggregates. Fine-grained interstitial löllingite occurs in the recrystallized breithauptite areas, where it forms a network or cell texture. (1 nicol, 160 x).

Fig. 3. Breithauptite-löllingite network or cell textures consisting of very fine-grained breithauptite with intergranular löllingite. The networks are surrounded by a clean rim of coarser-grained breithauptite. Grains of maucherite-breithauptite myrmekites occur enclosed in or adjacent to the breithauptite-löllingite networks. The maucherite commonly contains löllingite blades. (1 nicol, 160 x).

Fig. 4. Bladed intergrowths of gudmundite (white) and breithauptite (light grey). Darker grey myrmekitic crystals (left-hand and lower part of photograph) are maucherite-breithauptite myrmekites. (1 nicol, 160 x).

(Chr. HALKIER phot.).

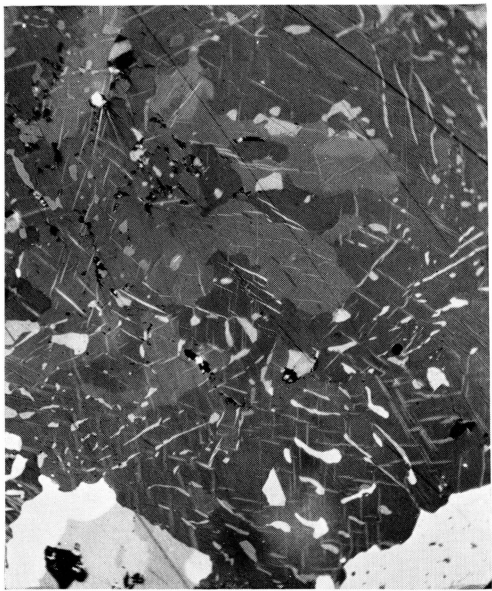


Fig. 1.

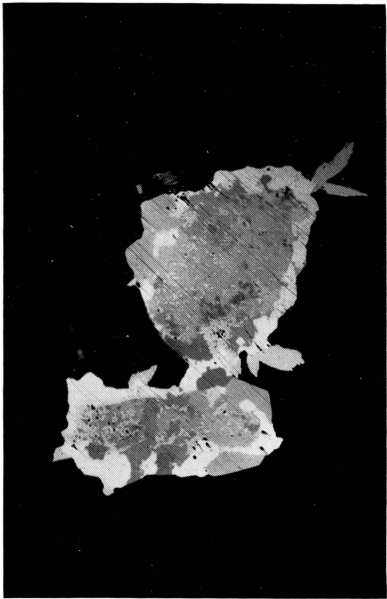


Fig. 2.



Fig. 3.

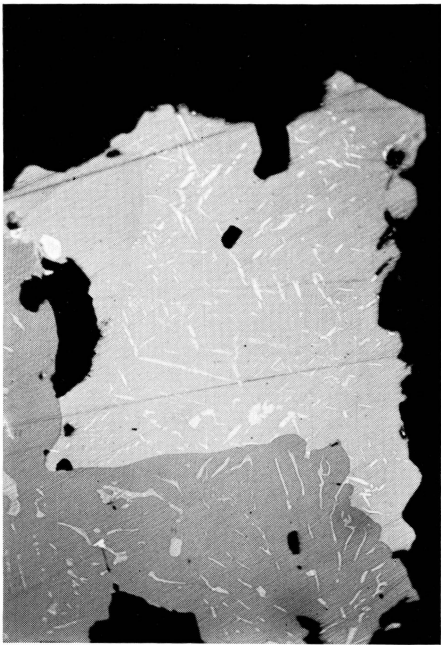


Fig. 4.