MINERALOGY AND PETROGRAPHY OF SOME TIN, LITHIUM AND BERYLLIUM BEARING ALBITE-PEGMATITES NEAR DOADE, GALICIA, SPAIN

BY

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ABSTRACT

The petrography and mineralogy of some Hercynian albite-pegmatites near Doade, Galicia, Spain is described. The mineral assemblage consists of albite, K-feldspar, quartz, muscovite, spodumene, petalite, cassiterite, beryl, columbite-tantalite, montebasite, apatite, eosphorite-childrenite, zircon, tourmaline, garnet, pyrite, bertrandite, ferrisicklerite, hetersite and cookeite. A tentative paragenetical sequence of these minerals is given. From the relationships between petalite and spodumene approximate initial temperature-pressure conditions of 600 °C and 2.5—3.5 kb. can be derived. The possible cause of the high sodium content and of the fine grainsize of the bulk of the pegmatites is discussed. A genetical relationship of the pegmatites with the medium to coarse grained Hercynian two-mica granites is suggested.

CONTENTS

Introduction .................................. 249
The country rock ............................ 250
Petrography of the pegmatites ............... 251
Summary of paragenetic relationships ...... 256
Genesis of the pegmatite .................. 257
Conclusions ................................ 259
Acknowledgements .......................... 259
References ................................ 259

INTRODUCTION

Fieldwork was carried out by the author in the summer of 1964 on some pegmatites occurring near the border of the provinces Pontevedra and Orense in Galicia, N.W. Spain. Topographical maps 186 and 135 of the Instituto Geográfico y Catastral de España were used on an enlarged scale of 1 : 25.000.

The pegmatites studied occur as veins in garnet-bearing quartz-mica schists that have a vertical NNW-SSE striking schistosity. The veins, which run parallel to the schistosity, are not continuous and are of variable thickness. All veins are confined to a zone 600 m. wide and about 15 km. long (Fig. 1) and have thicknesses ranging between 0.5—6 m. At approximately one km. to the W. of this zone an Hercynian two-mica granite crops out. Its contact is roughly parallel to the schistosity of the mica schists. The relationship between this granite and the pegmatites is discussed below.

Most of the veins have been mined for cassiterite and some for beryl. Mining developments are mainly open cut but some shallow underground workings are present.

Most of the study was confined to the pegmatite bodies near Doade (Fig. 2) where detailed sampling and subsequent mineralogical investigations were carried out. Two mines have been developed in this part of the pegmatite zone, namely Parabellon in the N. and Alcebedo in the S.

In the N. open cut a vertical section reveals the shape of the main pegmatite vein (Plate I Fig. 1). In this section a vertical dyke-like vein suddenly changes its dip to almost horizontal and thickens considerably and further W. it steepens again losing most of its thickness (Fig. 3). The pattern is similar in the underground workings of the Alcebedo mine according to the former manager of this mine. Thus, a stepwise dip to the W. seems to be general.

The contacts with the wallrock are sharp. Inclusions of schist in the pegmatite are rare. Some large patches
of schist which are completely or partly enclosed in pegmatite have not been rotated or otherwise displaced to any extent. Irregular apophyses of pegmatite in the schist are relatively rare but seem to be slightly more abundant in the thick, irregular flat-lying parts of the pegmatite veins (Plate I, fig. 2).

In both the N. and the S. a vein of milky quartz is found in the immediate vicinity of the main pegmatite vein. The dimensions of this quartz vein are 2 m. thick and several tens of metres long (Fig. 2).

THE COUNTRY ROCK

The quartz-mica schists that constitute the country rock have a vertical NNW-SSE striking schistosity. This schistosity has been observed in some places to be an axial plane to small scale folds that have subhorizontal axes. Numerous quartz bands occur generally forming boudins parallel to the schistosity and occasionally following the above mentioned small scale folding. Close to the contact with the pegmatites the schists are very rich in tourmaline.

Mineralogy

Quartz. — Quartz is always a major constituent. Invariably it shows undulose extinction. Different types of quartz, some of them probably belonging to different generations, have been found. Small quartzes with their long dimension oriented parallel to the schistosity occur in the mica-rich bands while in the quartz bands bigger equidimensional grains occur. However, in some thin sections of the mica schists these grains have an elongated habit, the elongation being parallel or at a small angle to that of the smaller grains in the mica-rich bands, and in others they are strongly undulose, showing amoeboid intergrowth produced by recrystallisation of crushed zones between the grains.

Muscovite. — This mineral is relatively fine-grained and has a preferred orientation parallel to the schistosity. Some recrystallised crenulation cleavages are observed, showing that the present foliation was preceded by an earlier schistosity. A small proportion of the micas indicate a third ill-defined plane which is parallel to the direction of elongated amoeboid quartz
Petrography of the Pegmatites

The pegmatites are leucocratic. Albite is the main constituent. K-feldspar, quartz and muscovite are found in varying amounts. Petalite, spodumene and most of the accessory minerals are difficult to recognize macroscopically because of their white colour and fine grainsize. Exceptions are cassiterite, columbite-tantalite and beryl.

The pegmatites vary in grainsize. Only K-feldspar occurs in relatively large crystals up to 5 cm. None of the other minerals exceeds the grainsize of 2 cm. Furthermore, the grainsize distribution is inhomogeneous. In places a layering parallel to the wall is marked by variations in grainsize and changes in mineralogical composition (Plate I, fig. 3). The layering, however, cannot be traced for any great distance. Irregular variations in grainsize occur even within a thin section. Fine fractions have grainsizes of 0.05—0.15 cm., coarser parts have 0.3—1.5 cm. While the K-feldspar-rich bands and patches vary from 1.5—4 cm.

A contact zone of 2—10 cm. thick is generally present. It consists of fine-grained muscovite and quartz. Very fine grains of cassiterite and apatite have been found in this zone. The first albite crystals adjoining the contact zone are often oriented with their longest axis normal to the wall.

Mineralogy

Albite. — Albite is the most abundant constituent of the pegmatites, which occasionally contain up to 80% of this mineral. It is white to colourless and the longest dimensions of the generally lath shaped crystals do not exceed 1 cm.

Microscopically, several varieties of albite may be recognised. Crystals of all varieties have large negative extinction angles (in sections perpendicular to X the angle between Y and the traces of (010) vary between 14 and 17°, indicating a nearly pure to pure albite composition). Albite twinning is common but Carlsbad-albite and pericline twinning are relatively rare. The varieties are as follows:

i) Small albite crystals with random orientation are included in K-feldspar and show corroded outlines. These probably crystallised at a very early stage.

ii) Relatively big crystals that often display graphic intergrowth with non-perthitic K-feldspar are of later origin. The K-feldspar is usually about 5—10%, but in rare cases may reach 50% (Plate I, fig. 4). As indicated below this albite variety commonly accompanies petalite and spodumene. The subhedral to euhedral crystals are lath shaped and exhibit irregular twinning, various patches having lamellae of different thickness. Wedge shaped twins are common, especially in crystals that are bent.

iii) The fine grained (0.2—7 mm.) sugary type of albite is quantitatively the most important one.

described above. Late cross-muscovites occur. These also show deformation in the form of bending and kinking, similar to that observed in all of the above mentioned varieties.

Biotite. — This mineral is less abundant than muscovite. Its pleochroism is red-brown (Z, Y) to yellow (X). The fabric arrangement is the same as for muscovite with which it is frequently intergrown. Cross-biotite has not been observed.

Garnet. — Small poikiloblastic crystals (1—3 mm.) of garnet are present. They contain small elongated quartzes that are mainly parallel to the external foliation. Mica-rich bands bend around the crystals.

Tourmaline. — Small, euhedral, needle shaped crystals (1 mm.) have a pleochroism of yellow-brown (O) to colourless (E). When zoned the pleochroism of the cores is blue-green (O) to colourless (E). The c-axes of the crystals are parallel to the schistosity, being randomly oriented in this plane. In thin sections in which the quartzes show amoeboid intergrowth, slight deformation of the tourmaline has occurred in the form of bending accompanied by fracturing at right angles to the c-axis.

Accessories. — Zircon, apatite, anatase, unidentified opaque mineral (lath shaped parallel to the schistosity).
The euhedral to subhedral crystals are lath-shaped and display a fine lamellar albite twinning. Together with muscovite and quartz the sugary albite corrodes and replaces K-feldspar phenocrysts along crystal boundaries, cleavage planes and irregular veins. The albite crystals in these veins have random orientation. This process leads in places to the almost complete replacement of the K-feldspar, a few patches of K-feldspar with parallel orientation indicating the former presence of that mineral.

Granophyric intergrowth with quartz is locally very abundant in both the bigger albites and particularly the saccharoidal type. Separate swarms of vermicular inclusions within one grain may be distinguished by their different optical orientations. The presence of quartz inclusions normally does not influence the habit of the albite (Plate II, fig. 1). Sometimes the granophytic quartz is confined to the rims of crystals, which have corroded outlines. Mostly, however, it occurs irregularly, showing no preference for either core or outer parts of the albite crystals. Frequently, only part of an albite crystal contains the vermicular quartz and this area is separated from the normal albite by a sharp boundary. Although the granophytic intergrowths with quartz may be due to simultaneous crystallization of albite with that mineral as described by Maijer (1965), the observed relationships seem to indicate that some of the myrmekitic intergrowths are due to the replacement of albite by quartz.

Veins of normal sugary albite in a granophytic-rich rock do not favour a time relationship as described by Maijer in which granophytic albite crystallizes later than the sugary type. It is thought that the apparent contradictory observations, also between the second and third varieties, are caused by the fact that the paragenetic relationships not only vary in time but also in space.

A possible fourth variety of irregularly twinned, cloudy albite which occurs together with adularia and euhedral quartz is probably formed under hydrothermal conditions as proposed by Maijer (1965). This type was only found in one thin section. All varieties show signs of deformation in the form of bending and undulose extinction.

**K-feldspar.** — The percentage of K-feldspar in the pegmatite probably does not exceed 15 but is difficult to estimate because of its mode of occurrence.

Three varieties can be distinguished. The first occurs in white to light pinkish crystals of up to 5 cm. in length. Euhedral crystals are not common but when present are mainly concentrated in patches and bands. Crosshatch twinning is very rarely observed in portions of the crystals and Carlsbad twinning is absent. Perthite may occur either as fine strings and patches which are probably due to exsolution of Na-feldspar, or as a coarser albite twinned type in veins and patches which might be due to replacement by albite. Replacement by albite, muscovite and quartz ± spodumene is common. Only albite and quartz have been noticed as inclusions.

The second variety, interstitial K-feldspar, commonly shows crosshatching and Carlsbad twinning and is not perthitic. It is only found in small quantities. This K-feldspar seems to have crystallised at the same time or later than the sugary albite.

Both varieties have a cloudy appearance in thin section owing to alteration to kaolinite.

Adularia is the third variety and abundant in a few hand-specimens collected in the western pegmatite near Parabellum. Euhedral crystals occur in small cavities together with quartz but the crystals in the solid rock have a hemimorphic appearance showing euhedral wedgeshaped outlines on one side only. Diagonal simple twinning is common. Very fine irregular crosshatch twinning (?) can be observed in parts where the alteration to kaolin is less pronounced. $2V_x \approx 70-80^\circ$. Together with adularia aggregates of fine greenish muscovite, idiomorphic quartz and bertrandite occur.

**Quartz.** — Quartz is a major constituent whose percentage in thin sections varies from 25 to 60. The anhedral grains commonly show undulose extinction. Quartzes from the Alcebedo area are strongly undulose and in some places amoeboid intergrowth of elongated grains occurs indicating deformation. No euhedral pseudomorphs after high temperature quartz have been detected.

Apart from the small parallel oriented euhedral quartzes that occur with cloudy albite and adularia, replacing earlier formed minerals, prismatic quartz crystals have been observed in veins and small cavities.

As indicated above, milky quartz occurs in two large quartz veins near the Parabellum and Alcebedo open cuts.

**Muscovite.** — Fine-grained (0.5—5 mm.) muscovite is found throughout the pegmatite. The average percentage is estimated to be 5 to 15. Especially in spodumene-rich specimens in which quartz predominates both muscovite and albite are rather rare. Most muscovite occurs with saccharoidal albite and quartz.

One mica was observed to have a central part with $2V_x = 0^\circ$ and a marginal part with $2V_x = 40^\circ$. This could mean that the mica is composed of different structural polymorphs (Deer, Howie and Zussman, 1962).

X-ray powder patterns of micas from both Parabellum and Alcebedo were compared with values given by Levinson (1953). They show some resemblance to the monoclinic 2-layer Li-muscovite, but the LiO content appears to be relatively low.

Trace-element analysis of Sn, Mn, Li and B contents are given in Table 1. Muscovite from the Parabellum mine has a LiO of 0.15 %. Micas from Campanarias have LiO = 0.05 % and those from Alfonsin LiO = 0.23 %.

Graphic intergrowth with quartz is occasionally
and encountered. K-feldspar is often replaced by muscovite in particular along grain-boundaries. Very fine-grained, greenish aggregates of muscovite occur in the adularia-rich rocks. They are probably of a late origin.

Deformation of muscovite in the form of bending and kinking is common. In the more strongly deformed parts of the pegmatites muscovite is concentrated in trails that follow a twisting course between the feldspar crystals.

In hand-specimen these rocks have a coarse phyllonitic texture similar to that found in parts of the Hercynian two-mica granites (Plate IV, fig. 4). The rocks part along irregular planes which are mica-rich and display slickensides.

Spodumene. (LiAlSi₂O₆) — Spodumene occurs only in the veins of the Alcebedo area where it may constitute 25—35 % of the rock. The white to colourless crystals are fine-grained (0,5 — 3 mm.) and have a prismatic or platy habit. Euhedral, eight-sided basal sections with simple (100) twinning are numerous although commonly sub- to anhedral outlines are encountered. In one thin section sheaf-like aggregates radiating from a vein have been noticed (Plate II, fig. 2). 2V° ~ 65—70° and Z/c ~ 23—25°.

Spodumene often shows graphic intergrowth with quartz. The relative proportions of the intergrown minerals vary from some quartz droplets in spodumene to small isles of spodumene surrounded by quartz. The quartzes are often relatively big and strongly undulose, not infrequently displaying amoeboid intergrowths. Several spodumene grains with different orientations often occur in individual quartz grains giving "sieve structure" (Plate II, fig. 3).

Spodumene replaces K-feldspar, although a fine graphic intergrowth observed locally on the boundary between the two minerals might indicate simultaneous crystallization. Anhedral, symplectic spodumene (spodumene + albite?) together with albite and muscovite, that is encountered in veins of K-feldspar, has been formed later (Plate II, fig. 4).

Against albite, and rarely against K-feldspar, the spodumene displays symplectic rims (Plate III, Fig. 1). Wholly symplectic, anhedral grains as described above occur interstitial to the first generation albite, slightly corroding it along grain-boundaries. The second mineral in the symplectites often appears to be quartz. Only in a few instances in rims against albite grains, albite could be proved to be the intergrown phase. In one thin section this type was found as dispersed grains in an albite-rich rock. Finely granulated, cloudy zones containing spodumene and quartz, that occasionally cut across euhedral spodumene crystals, are most likely due to deformation. Offseting along these zones has not been observed. Spodumene is not altered.

Petalite. (Li(Na)Si₄AlO₁₀) — In contrast with spodumene, this mineral was encountered only in very few thin sections of specimens collected in the eastern vein of the Alcebedo mine. Macroscopically its colour is white. The anhedral grains with grainsizes varying from 0,05—1,5 cm. occur in albite-rich rocks. Alteration along cleavage planes to yellowish-brown, fibrous montmorillonite is a typical feature. Lamellar twinning is rare. Near the margin of a large crystal small albite inclusions occur exhibiting thin, myrmekitic rims. This crystal also encloses differently oriented petalite grains, that become more abundant towards the margin, grading into a mosaic of small (0,5—2 mm.) petalite grains. Along grain boundaries replacement by graphically intergrown spodumene and quartz is observed (Fig. 4). The breakdown of petalite to spodumene and quartz has been demonstrated experimentally by Stewart (1963).

The textural relationships seem to indicate that petalite was formed before and together with the albite. Spodumene was formed later.

The pink clay mineral that had been noticed previously by Ypma and others (personal communication) proved to be the alteration product of petalite. It occurs in small nests in the E. vein and was found in greater quantities in nests up to 10 cm. in diameter in a mine near Presqueira (Fig. 1). A thin section from this locality contains petalite. An X-ray powder photograph proved that the mineral was dioctahedral.
thus belonging to the montmorillonite group. (See chemical analysis in Table 2).

Plate III, fig. 2 shows a thin section of a pink montmorillonite aggregate in which relics of petalite, displaying lamellar twinning, occur. The pink colour of the mineral is dependent upon its moisture content because when dried it loses its colour.

### TABLE 2. Chemical analysis of montmorillonite

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<table>
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<tr>
<td>Li₂O</td>
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</tr>
<tr>
<td>Loss on ignition</td>
<td>22.34</td>
</tr>
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</table>

Analyst: Miss A. Giesbert

Deer, Howie and Zussman (1962) report this alteration to be typical for petalite occurrences. To date petalite has not been recorded from Galicia or N. Portugal. The identification of petalite was confirmed by X-ray powder photography.

**Cassiterite.** — Mining values indicate that the cassiterite weight-percentage of the pegmatite does not exceed 0.2. Cassiterite is found dispersed through the pegmatite as anhedral, subhedral and occasionally euhedral crystals up to 0.5 cm. in size. Macroscopically the mineral is black. In the heavy mineral concentrate dipyrismatic crystals with occasional small prism faces were observed. In thin section lozenge-shaped sections with diagonal twin planes are common.

Pleochroism is red-brown (E) to yellow to colourless (O). A zoning marked by bands showing an oscillating intensity of pleochroism is common. The zoning is often irregular, outer zones not being parallel to inner zones and sometimes cutting them off.

Inclusions of quartz, muscovite and albite are found, rarely in large quantities, giving the cassiterite a poikiloblastic habit.

The paragenetic assemblage consists of both albite types, especially the sugary type, quartz and muscovite. In the spodumene-rich rocks cassiterite is rare or absent. It does not occur as inclusions in K-feldspar. In the contact-zone very small grains of cassiterite are found together with muscovite and quartz.

**Beryl.** — Beryl is a common and often rather abundant accessory constituent. Its percentage equals and not infrequently exceeds that of cassiterite. In hand-specimen it has a pale green to yellowish colour. Crystals are mainly prismatic and have subhedral to anhedral outlines. Grainsizes vary from 0.3 to 3 mm. Beryl occurs together with sugary albite, quartz, muscovite and cassiterite.

In a thin section of the hydrothermally altered part of the W. vein of the Azevedo area, a grain was encountered that vaguely exhibited two cleavages at right angles. An X-ray powder photograph of this grain revealed a distortion of the normal beryl pattern. The cell parameters were calculated using the d-values of the (304) and (431) reflections and are a = 9,159 Å and c = 9,142 Å. Compared with values given by Deer, Howie and Zussman (1962) i.e. a = 9,188 Å, c = 9,189 Å and the A.S.T.M. X-ray, powder data file (card 9-430), i.e. a = 9,215 Å, c = 9,192 Å, a considerable shrinkage of the lattice is indicated.

In the same thin section and in others made from the same hand-specimen, beryl is found only in very small amounts while the mineral bertrandite (Be₃Si₃O₉(OH)₉) is rather abundant.

As indications of alteration of beryl to this mineral have been found it seems possible that the occurrence of the anomalous cell parameters is due to incipient alteration of beryl to bertrandite. By the loss of Al and Si and the addition of H₂O the beryl lattice might slightly shrink before it loses its hexagonal symmetry and becomes bertrandite, that has a rhombic pyramidal symmetry (Solovyeva and Belov, 1961).

Three other alteration products have been found, the first two listed below also occurring with bertrandite. All three minerals are mostly fibrous and occur in radial aggregates. They occur along grain-boundaries and occasionally completely replace beryl. These are:

i) A biotite-like mineral with a pleochroism of dark brown or green-brown (Y, Z) to pale yellow (X), 2Vₓ = 20° approximately, r<v and moderate birefringence,

ii) a chlorite-like mineral with an orange-yellow colour, 2Vₓ = 40° approximately, low birefringence and anomalous interference colours, and

iii) a clay mineral with very low birefringence and 2Vₓ = 30° approximately.

**Columbite-tantalite.** — In the heavy mineral concentrates subhedral to euhedral platy and prismatic crystals of columbite-tantalite have been found. The crystal faces are striated parallel to their length and have a black lustre with a purple tinge. The streak is dark brown to black.

Microscopically the mineral is opaque, probably due to the fact that it is Fe-rich. Very small inclusions of columbite-tantalite were noticed in the margin and on the outside of cassiterite crystals. Intergrowth of bigger grains with cassiterite has been observed macroscopically.

In the spodumene-rich specimens from the S., columbite-tantalite has been found to be more abundant than cassiterite in a heavy mineral concentrate, but the total percentage of heavy minerals was very low in this case.

The X-ray pattern is in accordance with that of columbite-tantalite.
Montebrasite. — (LiAl(OH)PO₄) — Montebrasite occurs in very small quantities both in the N. and S. veins. The colourless grains are 0,1 to 1 mm. in size and have subhedral, platy or anhedral habit. Lamellar twinning is typically developed. One grain displayed two systems of twinning approximately at right angles to each other (Plate III, fig. 3).

In the southern veins the mineral occurs together with spodumene, in which it is occasionally included. In the Parabellon veins it is found together with sugary albite, muscovite, quartz, cassiterite and beryl, all of which may replace K-feldspar.

According to Winchell and Winchell (1956) the measured value of \(2V_Z = 71\), 72° indicates that the mineral is the F-free end member of the isomorphous amblygonite-montebrasite series. Similar montebrasite has been described by Maijer (1965). Three alteration products described by Maijer can be recognized.

i) A light green to colourless mineral with very low birefringence,

ii) a fibrous chlorite-like mineral. Both i) and ii) occur along grain-boundaries and

iii) a mineral with a higher birefringence than montebrasite, occurring along cleavage-planes.

As the alteration of the montebrasite is not very pronounced no determination of the optical properties of these materials could be made.

Apatite — This mineral is only found in small amounts. It forms small, seldom euhedral crystals often with a cloudy appearance.

It has probably formed at different times during the crystallization of the pegmatite. It occurs in the wall zone and in the albite-rich rocks. It is slightly more common in the eastern hydrothermally altered rocks, where it is associated with adularia.

Eosphorite-childenite. — (MnFe)AlPO₄(OH).H₂O) — This mineral has been encountered in some minute clusters of anhedral grains in thin sections of the veins of the Parabellon mine. Microscopically, the mineral is colourless and has a moderate birefringence and \(2V_Z = 44°\) (measured). The optical plane is parallel to the rather poor (100) cleavage. The relief is high and axial dispersion \(r < v\) strong. These properties indicate that the composition lies near the eosphorite end of the series.

The mineral has been reported from two other localities in Galicia (Avé Lallemant, 1965 and Hensen, 1965). Both of these occurrences are in Hercynian leucocratic muscovite-granite.

Zircon. — Zircon is found in heavy mineral concentrates as small, brown-yellow, euhedral crystals, having tetragonal-bipyramidal symmetry. In thin section they are metamict (cloudy) and show decomposition haloes. X-ray powder pattern of these crystals confirmed them to be zircon.

Tourmaline. — Tourmaline is very rare. It was only observed once in a narrow vein containing very little cassiterite and no other accessories. The small prismatic crystals show a pleochroism of yellowish brown (O) to colourless (E).

Garnet. — A few grains of this mineral have been found in the heavy mineral concentrate of the northern mine. It forms very small, pinkish, euhedral crystals. \(a_h = 11.57\) Å, \(n = 1.81\). These properties indicate a garnet with high spessartite and almandite content, having spessartite > almandite. This is in accordance with the values given by Schneiderhöhn (1961) for garnets in pegmatites.

Pyrite. — In the heavy mineral concentrate of the southern mine very small amounts of fine-grained pyrite were found. The identity of this mineral was verified by X-ray photography.

Bertrandite. — (Be₂Si₂O₇(OH))₂ — Bertrandite was only recognized microscopically. It forms small colourless crystals with sub- to euhedral outlines. It exhibits two perfect cleavages at right angles and a typical heart-shaped, simple twinning (Plate III, Fig. 4). These heart-shaped (001) twins are mentioned by Tröger (1959). Relief and birefringence are very similar to those of muscovite. \(2V_X = 70, 71°\) (measured) and \(r < v\) weak.

Direct alteration of beryl to bertrandite could only be found in a few places. Generally the bertrandite occurs in clusters of euhedral, hemimorphic crystals, indicating that the mineral probably crystallized from hydrothermal solutions. It is almost invariably found together with adularia, both in the W. veins, where this mineral is very abundant locally and in the E. veins, where only a very small vein containing adularia and bertrandite was observed in an albite-rich specimen. Bertrandite is not altered.

X-ray powder photographs verified the identity of this mineral.

Ferrisicklerite. — (Li(Fe,Mn)PO₄) — Ferrisicklerite only occurs in the Alcebedo mine. This mineral, that has been described from several Li-pegmatites, is probably a hydrothermal alteration product of triphyllite (Mason, 1964).

Colour is very dark brown to black. In thin section a dark red brown to orange brown pleochroism is observed. The anhedral grains show one good and one poor cleavage at right angles to each other. The grains have a straight extinction. An axial figure could not be obtained. An X-ray analysis provided values very similar to those given by Björling and Westgren (1938) for this mineral in the Varuträsk pegmatite (Sweden). See Table 3. Note that values under \(d = 1.35\) Å had not been recorded by these authors. The anhedral grains are dispersed through the spodumene-rich parts of the pegmatite. Grain size is from 0,05 to 0,5 cm. It is most abundant in the W. vein of the Alcebedo mine. The mineral readily alters to heterosite if exposed to the atmosphere.

Triphyllite which has not been found in this pegmatite has been reported by the author in an aplite near Bueu (Hensen, 1965). Optical properties are \(2V_Z = 5-10°\), axial dispersion \(r > v\) marked, high relief and low birefringence (quartz). The X-ray powder data are consistent with those given in the A.S.T.M. X-ray powder data file for sicklerite (card 13-338).
TABLE 3. X-ray powder data for Ferri-sicklerite and Heterosite.
Fe-Kα radiation, camera diam. 90 mm.

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<tr>
<td>2,49 vs</td>
<td>2,43 s</td>
</tr>
<tr>
<td>2,43 m</td>
<td>2,42 w</td>
</tr>
<tr>
<td>2,33 w</td>
<td>2,32 w</td>
</tr>
<tr>
<td>2,24 w</td>
<td>2,25 wv</td>
</tr>
<tr>
<td>2,17 w</td>
<td>2,21 w</td>
</tr>
<tr>
<td>1,86 w</td>
<td>2,16 m</td>
</tr>
<tr>
<td>1,75 w</td>
<td>1,97 wv</td>
</tr>
<tr>
<td>1,67 w</td>
<td>1,84 wv</td>
</tr>
<tr>
<td>1,61 m</td>
<td>1,81 w</td>
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<tr>
<td>1,48 w</td>
<td>1,72 w</td>
</tr>
<tr>
<td>1,46 w</td>
<td>1,68 w</td>
</tr>
<tr>
<td>1,35 m</td>
<td>1,63 wv</td>
</tr>
<tr>
<td>1,28 w</td>
<td>1,37 w</td>
</tr>
<tr>
<td>1,25 w</td>
<td>1,35 w</td>
</tr>
<tr>
<td>1,16 vw</td>
<td>1,51 m</td>
</tr>
<tr>
<td>1,15 vw</td>
<td>1,46 m</td>
</tr>
<tr>
<td>1,13 vv</td>
<td>1,41 w</td>
</tr>
<tr>
<td>1,12 vv</td>
<td>1,33 w</td>
</tr>
<tr>
<td>1,10 vv</td>
<td>1,30 vv</td>
</tr>
<tr>
<td>1,09 w</td>
<td>1,24 vv</td>
</tr>
<tr>
<td>1,04 vv</td>
<td>1,23 vv</td>
</tr>
<tr>
<td>1,02 vv</td>
<td>1,21 vv</td>
</tr>
<tr>
<td>1,01 vvv</td>
<td>1,19 vv</td>
</tr>
<tr>
<td>0,99 w</td>
<td>1,10 w</td>
</tr>
<tr>
<td>0,98 vv</td>
<td>1,09 w</td>
</tr>
<tr>
<td></td>
<td>1,07 w</td>
</tr>
</tbody>
</table>

**Heterosite.** — (Fe, Mn)PO₄ — This mineral is the weathering product of ferrisicklerite. It shows, after a thin black coating has been removed, a bright purple colour. In places two cleavages, at approximately 90°, could be observed. The hardness of the mineral is low and it has a purple streak. Under the microscope thin cleavage fragments have a purple colour and are weakly birefringent. Pleochroism as described by Quensel (1957) has not been observed. According to Quensel the above mentioned black coating is an H₂O-rich, Fe-Mn phosphate.

X-ray analysis of heterosite provided values that are slightly different from those given by Björling and Westgren (1938). The d-values are given in Table 3. Around the small masses of heterosite an orange-brown powdery decomposition material is found. An X-ray photograph showed only a small number of very vague lines in part corresponding to those of heterosite.

**Cookeite.** — (LiAl₄(OH)₆Si₂AlO₁₈) — The mineral cookeite has been encountered in a ground fraction (< 0.4 mm.) of two hand-specimens, from the Alcebedo mine. It could not be detected in thin sections of these rocks. It occurs as transparent, light green, euhedral crystals of 0.5 to 2 mm. in size, that have acute, wedge-shaped forms and striated crystal faces (Plate IV, fig. 1).

Optical properties are the same as those given by Winchell and Winchell (1956). 2V₉ varies from 0 to 70°. Axial dispersion r > v is distinct. nₙ ≈ 1.57. A basal section divided into six bi-axial sectors around an uni-axial centre as described by the above named authors was observed in one of the crystals. In other crystals either this phenomenon is only partly developed or fan-like extinction patterns are exhibited.

X-ray data are in accordance with those given by the A.S.T.M. X-ray powder data file (card 10-397).

With a crystal reflection goniometer two zones of relatively weak reflecting faces were detected in one crystal (Plate IV, fig. 2). The small number of measured crystal faces did not supply an answer to the crystallographic symmetry of the crystal. However, an X-ray rotation photograph, rotating the crystal around its most prominent zone-axis (zone of 4 faces) gave the result presented in Plate IV, fig. 3. An almost random orientation of probably fibrous components is indicated by the photograph. Some preferred orientation is expressed by slightly thicker and more blackened parts of the lines, which show a symmetrical pattern. Part of the crystals are composed of a different colourless mineral. Striations on the crystal faces continue uninterruptedly across mineral boundaries. The fact that this mineral appears to be quartz, makes it very probable that the cookeite crystals are partial pseudomorphs after quartz. None of the measured interfacial angles are found among the normal crystal faces of quartz, listed by Frondel (1962).

**Mn (Fe) oxide.** — Black coating on surfaces and along cleavage planes of albite and muscovite are very abundant in parts of the pegmatite. Microanalysis proved Mn as the most important element. Iron is only of minor importance. One exception of the Alcebedo mine gave Fe positive, Mn trace and P positive. This is similar to the results of heterosite. These gave Fe positive, Mn positive and P positive.

**SUMMARY OF PARAGENETIC RELATIONSHIPS**

On the basis of the microscopic investigations, described above, several consecutive phases in the crystallisation history of the pegmatite can be distinguished. Although the age relationships between the different phases could not definitely be established and some of the phases may largely overlap each other, the paragenetic assemblages as given below are regarded to be significant. The phases are listed in the order of formation.

**Pegmatitic phase**

a) K-feldspar + minor amounts of albite (variety (i)) and quartz that are included in the former
b) albite (variety (ii)) + petalite
c) spodumene + quartz ± albite (variety (ii))
d) sugary albite (variety (iii)) + muscovite + quartz ± cassiterite, beryl + accessory minerals.
This assemblage constitutes the bulk of the pegmatites.

**Hydrothermal phase**
- adularia, bertrandite, ferri-sicklerite, cookeite, albite (variety (iv)).

**Supergene phase**
- clay minerals, heterosite.

These paragenetic relations are summarised in Table 5.

### Table 5. Summary of paragenetical relationships

<table>
<thead>
<tr>
<th>Pegmatitic phase</th>
<th>Hydrothermal phase</th>
<th>Supergene phase</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Albite</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>K-feldspar</strong></td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td><strong>Quartz</strong></td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td><strong>Muscovite</strong></td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td><strong>Spodumene (A)</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Petalite (A)</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Casiterite</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Beryl</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Columbite-tantalite</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Montebrasite</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Apatite</strong></td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td><strong>Eosphorite-childrenite</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Zircon</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Tourmaline</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Garnet</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Pyrite</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Bertrandite</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Ferri-sicklerite (A)</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Heterosite (A)</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Cookeite (A)</strong></td>
<td>x</td>
<td></td>
</tr>
<tr>
<td><strong>Chlorite(-like) mineral</strong></td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td><strong>Biotite-like mineral</strong></td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td><strong>Pink montmorillonite (A)</strong></td>
<td>x</td>
<td>x</td>
</tr>
<tr>
<td><strong>Mn, Fe oxide</strong></td>
<td>x</td>
<td></td>
</tr>
</tbody>
</table>

The pegmatitic phase is subdivided into four subphases that coincide with those mentioned in the text. (A) indicates found in Alcebedo area only.

**Genesis of the Pegmatite**

Recent experimental work on the crystallization behaviour of Li-containing melts by Stewart (1960, 1963) indicates the following:

i) the saturated feldspar-silica liquidus is lowered by the presence of Li-bearing components,

ii) the bulk compositions of a number of large, zoned and unzoned pegmatites lie in a region of minimum melting temperatures of the system albite-quartz-spodumene-water. The Li₂O content of these rocks varies from 1.2—1.8 %. An homogeneous magma of above named composition can exist at temperatures below 625 °C.

iii) The pressure-temperature conditions of the system influence the formation of either petalite or spodumene independently of the composition of the melt. It has been found that the petalite-feldspar assemblage is the high temperature-low pressure equivalent of the feldspar-spodumene-quartz assemblage. Under equilibrium-conditions petalite is stable between 550°—660° C. at 2 kb. and 635°—680° C. at 4 kb. Below both minimum temperatures petalite reacts to spodumene + 2 quartz.

It would seem then that the presence of both petalite and spodumene and the reaction rims of spodumene and quartz around the petalite in the rocks under discussion indicate that the temperature must have been close to the phase boundary. Stewart (1963) further found that the solid dissolved in the gas phase coexisting with the melt contains more silica than the melt. As other indications for an escape of gas from the system exist (see below), the quartz veins occurring close to the pegmatite veins may be explained as having been formed from these solutions.

Jahns and Tuttle (1963) elaborate on the genetic relationships between pegmatites and aplites. They emphasize the relative enrichment in sodium of the fine-grained parts of layered pegmatites. Both the difference in grain size and in chemical composition of aplitic and pegmatitic phases is explained by a segregation process in which potassium in preference to sodium is extracted from the liquid by a vapour. Experimental evidence has given proof for this process and also for the supposition that phases crystallized from the vapour are much coarser than those crystallized directly from the melt.

Several phenomena that have been observed during the present study fit their category “Bodies of aplite or fine-grained pegmatite with very large phenocrysts or megacrysts” remarkably well. The following properties have to be noted:

i) Anhedral to euhedral phenocrysts of perthite and other minerals.

ii) Groundmass consisting mainly of quartz and albite, with a sugary appearance and grain sizes from 0.2—5.0 mm.

iii) Fine layering of bands with different composition (probably flow-layering). The layered groundmass is commonly traceable into homogeneous rock both laterally and in directions normal to the layering. The layering tends to be parallel to the walls.

Although the phenocrysts in this case are relatively small (up to 5 cm.) and merely consist of perthitic K-feldspar, it must be kept in mind that only a small vertical section is accessible for investigation.

Majer (1965) reports bigger phenocrysts of K-feldspar and also certain other minerals from the otherwise, mineralogically and texturally very similar pegmatites, from the Amarante area, N. Portugal. He refers to a
study by Cotela Neiva (1954) of pegmatites near Cabraçao, N. Portugal. Cotela Neiva showed that chemical analyses on specimens taken at different intervals between 0 and 70 m. below the surface indicate a rapid decrease of potassium with depth. However, the apparent uniform, fine grain size in all outcrops visited in the pegmatite zone, which have maximum differences in elevation of approximately 100 m., does not indicate any differentiation of this kind within these limits.

It is possible that in this case the mechanism of "pressure quenching" (Jahns and Tuttle, 1963) that results from the escape of the vapour phase from the system, has played an important role in the crystalization history of the melt.

The escape of potassium into the wall rocks does not seem to have taken place to any great extent as no K-feldspar and only minor amounts of late cross-muscovite have been found in the schists within the pegmatite zone.

The Li-pegmatites from Lalin (Fig. 1) described by Parga-Pondal and Cardoso (1948) are coarse-grained and have prismatic spodumene crystals up to 30 cm. in size. It is probable that in that case the gas pressure during crystallization has been higher. No petalite is reported from these pegmatites. An analysis of a bulk sample of this pegmatite gave Na₂O = 6.56%, K₂O = 0.92%, Li₂O = 1.20%, SiO₂ = 72.61%, Al₂O₃ = 17.68% (Parga-Pondal and Cardoso, 1948). Although the K₂O content of this sample is probably not representative for the whole pegmatite, the high Na₂O percentage seems to be significant.

The absence of fluoride and topaz in the pegmatites is thought to indicate a deficiency in F. The occurrence of the mineral montebraziel instead of ambygonite and the absence of lepidolite which is a stable Li-silicate under 640° C. in the presence of K and H₂O and F are most likely due to this deficiency.

Schneiderhöhn (1961) mentions the fact that only very few topaz occurrences are known in Spain. He concludes that F must have been relatively rare in pegmatic rest melts, while B was very abundant. In this case most of the B has escaped into the wallrock to form tourmaline. These very tourmaline-rich contact zones are known from the contacts of many other Hercynian granite and pegmatite occurrences in Galicia and also appear to be common in Portugal (Maijer, 1965).

For several reasons given below, a genetic relationship with the two-mica granite to the W. is suggested. This granite is a medium to coarse grained two-mica granite. It is an homogeneous, intrusive granite of Hercynian age (Floor, 1966). Quite frequently the granites of this type have undergone a late cataclastic deformation, which gives them a phyllonitic texture (Avé Lallemant, 1965; Hensen, 1965). The coarse-grained, biotite granites which crop out to the S. do not show this type of deformation and could also, by means of intrusive relationships, be proved to be younger than the former.

**Mineralogy**

The mineralogy of this two-mica granite is very similar to that given by Floor (1966) and is as follows: K-feldspar. — (30—35%) — The K-feldspar is perthitic and has small inclusions of plagioclase that show decalcification rims. It has a marked microcline cross-hatching.

Plagioclase. — (25—30%) — Albite composition (An₂⁵—An₁₈). Although zoning is inconspicuous, sericitized cores seem to indicate that a zonary distribution with a more lime rich core originally existed.

Quartz. — (40%) — In places strongly undulose.

Muscovite. — (10%) — Fairly thick lepidoblasts having irregular intergrowths with biotite.

Biotite. — (5—7%) — Slightly lower percentage than muscovite. Pleochroism: red-brown (X, Z) to light yellow (Y).

Both minerals show signs of deformation in the form of kinking and bending.

**Accessories.** — zircon, apatite, indeterminate opaque mineral.

The following observations seem to indicate a relationship between this granite and the pegmatites:

i) The stepwise dip of veins towards the granite.

ii) The occurrence of phyllonitic texture in the pegmatite. This feature indicates that the emplacement of pegmatite took place before that of the coarse-grained biotite-granites to the south (Fig. 1). The emplacement of the pegmatite veins is thought to have taken place at about the same time as the intrusion of the granite. The "shouldering aside" of the countryrocks by the intrusion may have facilitated the uprise of the pegmatic magma along a dilation zone in the schist, which runs parallel to the contact.

A chemical relationship cannot be proved. Trace analyses of a small number of samples of micas from the granite are given in Table 4. The sample numbers correspond with those on Fig. 1. The Li₂O content of the biotite is not very high and does not seem to provide a proof for a genetic relationship between pegmatites and granite.

**TABLE 4. Trace analyses of biotite and muscovite from 2-mica granite**

<table>
<thead>
<tr>
<th>Sample no.</th>
<th>Mineral</th>
<th>Li₂O %</th>
<th>MnO %</th>
<th>Sn %</th>
<th>B %</th>
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</thead>
<tbody>
<tr>
<td>1</td>
<td>musc.</td>
<td>0.13</td>
<td>0.007</td>
<td>0.009</td>
<td>0.01</td>
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<tr>
<td>2</td>
<td>musc.</td>
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<td>0.37</td>
<td>0.115</td>
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<tr>
<td>3</td>
<td>musc.</td>
<td>0.51</td>
<td>0.39</td>
<td>0.013</td>
<td>0.01</td>
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<tr>
<td>4</td>
<td>musc.</td>
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<td>0.007</td>
<td>0.007</td>
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<tr>
<td>5</td>
<td>musc.</td>
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<tr>
<td>6</td>
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<td>0.001</td>
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</tr>
<tr>
<td>7</td>
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<td>0.007</td>
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<tr>
<td>8</td>
<td>musc.</td>
<td>0.34</td>
<td>0.27</td>
<td>0.022</td>
<td>0.01</td>
</tr>
</tbody>
</table>

Analyses by H.M.I.B. Arnhem.
CONCLUSIONS

From the above facts the following conclusions are derived:

i) The pegmatitic assemblages most likely crystallized from a melt genetically related to the two-mica granites outcropping to the W.

ii) The initial temperature of crystallization was probably around 600°C at a pressure between 2.5 and 3.5 kb.

iii) A segregation process that preferentially removed potassium from the melt led to the high albite content of the pegmatites.

iv) The fine, aplitic, grain size of the bulk of the pegmatite is either due to the escape of the gas phase from the system producing a "pressure quench" or to the fact that albite crystallized from a melt rather than a vapour phase. Possibly both processes operated to give the observed result.

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I am much indebted to Dr. P. J. M. Ypma, Dr. G. L. Krol and Mr. L. J. Fick for supervision of the work which was carried out for the second M. Sc. thesis in the Dept. of Petrology, Mineralogy and Crystallography of Leiden University. Dr. P. Hartman is acknowledged for his kind interest and help with the X-ray investigations and Mr. R. O. Felius for the X-ray determination of ferri-sicklerite. To Professor B. Nashar the author wishes to express his gratitude for critically reading and correcting the English manuscript.

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Last but not least I want to acknowledge the invaluable help of the technical staff of the Leiden Geological Institute in completing this study.

REFERENCES


PLATE I

Fig. 1 Vertical vein with irregular flat-lying body (centre). Parabellon open cut mine looking N.N.W.

Fig. 2 Irregular apophysis of pegmatite in schist, occurring in the flat-lying part of the pegmatite (Parabellon)

Fig. 3 Layering in fine-grained albite-quartz-muscovite rock, marked by changes both in mineralogy and grainsize. The layering is cut off by a small vein containing a coarser-grained albite (nat. size).

Fig. 4 Graphic intergrowth of albite (2nd variety) and K-feldspar. Spodumene (lower part of the picture) corrodes the albite along its grain boundaries (25 ×).
Fig. 1 Granophyric albite. The quartz veins cutting across several albite grains appear to have had a replacement origin and the vermicular quartz seems to have had a replacement origin (25 x).

Fig. 2 Sheaf-like aggregates of spodumene radiating outwards from a thin vein (25 x).

Fig. 3 Graphic intergrowth of spodumene and quartz. Several spodumene grains with different orientations occur per quartz individual (60 x).

Fig. 4 Veinlet of symplectic spodumene with albite and muscovite in perthitic K-feldspar (25 x).
Fig. 1  Symplectitic spodumene in contact with albite (2nd variety). At the contact between the two minerals a thin symplectitic rim occurs. Dark patches in albite are K-feldspar (60 ×).

Fig. 2  Relics of petalite, exhibiting lamellar twinning, in pink montmorillonite (50 ×).

Fig. 3  Montebrasite with fine lamellar twinning. Dark grey is quartz. Spodumene shows high relief. Note grain with two sets of twins normal to each other in upper right hand quadrant (40 ×).

Fig. 4  Euhedral hemimorphic crystals of bertrandite, showing heart shaped simple twinning. Also muscovite (left) and adularia (upper right) (60 ×).
Fig. 1  Typical cookeite crystals with striated crystal faces. Most crystals appear, on close examination, to consist partly of quartz (15 ×).

Fig. 2  Cookeite crystal with two zones of crystal faces. Milky white part is quartz having one crystal face in common with the cookeite (30 ×).

Fig. 3  X-ray rotation photograph of crystal pictured in fig. 2. The crystal was rotated around a zone axis (zone of 4 faces). The symmetrical arrangement of thicker parts of the lines may be caused by some preferred orientation of flakes within the crystal. Cu Kα (Ni) radiation.

Fig. 4  Thin crosscutting pegmatite vein in schist showing penetrative phyllonitisation. Horizontal section.