

MINERALOGY AND PARAGENESIS OF AMBLYGONITE - MONTEBRASITE
WITH SPECIAL REFERENCE TO THE TANCO (CHEMALLOY) PEGMATITE,
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ABSTRACT

This thesis describes the results of a study of about 219 specimens of the amblygonite - montebrasite series, LiAlPO_4F - $\text{LiAlPO}_4(\text{OH})$, with 157 specimens from the Tanco pegmatite and the remainder from other world localities. The investigation had two more or less separate objectives, one being a study of the variations of certain physical properties with variable F/OH ratios within the series, and the other being a study of the chemical variation in amblygonite - montebrasites from different zones within the Tanco pegmatite.

The variations of physical properties with F/OH ratio were studied on 15 members of the amblygonite - montebrasite series varying from 1.4 to 11.8% F. Specific gravity increases with F-content, following closely a straight line connecting the end-members values of 2.98 for montebrasite and 3.11 for amblygonite. The refractive index n_f increases from 1.586 for amblygonite to 1.644 for montebrasite. Differential thermal analysis showed a regular variation with F-content in the positions of the principal endotherms on the DTA curves. The products after heating to 1000° C have been identified as $\text{Li}_4\text{P}_2\text{O}_7$, Li_3PO_4 and three forms of AlPO_4 , the mutual portions of which vary with the F-contents of the original phase. Unit cell dimensions showed non-linear variations from F-rich to F-poor members. A chart for the indirect estimation of the F-contents in amblygonite - montebrasites was constructed using a group of six powder reflections whose $2\theta^\circ$ $\text{CuK}\alpha$ angles vary most conspicuously; the accuracy is regarded as $\pm 0.5\%$ F. Of these physical properties, X-ray powder diffractograms are considered to yield the most accurate estimate of F-contents.

For the second part of the study, 157 specimens of the amblygonite - montebrasites in the Tanco pegmatite were X-rayed and their F-contents estimated, twelve were analyzed for F and 35 were analyzed for Na_2O , K_2O and CaO . All of the last 35 specimens were found to contain negligible amounts of the three oxides. The amblygonite - montebrasite occurs mainly in three zones of primary crystallization, and none has been found in presumed metasomatic units. In the spodumene - alkali feldspar - albite - quartz intermediate zone, white, pink and yellow amblygonite occurs with F from 6.8 to 4.0%. In the later spodumene - quartz - (alkali feldspar) intermediate zone, white amblygonite - montebrasite is richer in F, containing 7.1 - 5.4% F. In the still later quartz core, which is gradational from the previous zone, the F-content decreases gradually to 6.4 - 4.5%. Within each individual zone, single blocks of amblygonite show a decrease of up to 1.9% in F-content during their growth; this decrease is usually accompanied by a colour variation from pink through white to yellow. In all zones, the primary amblygonite - montebrasite is frequently replaced by a polysynthetically twinned, F-poor montebrasite (1.8 - 4.0% F), the loss in F-content from the parent mineral being as much as 4.0%.

PART I. GENERAL INTRODUCTION

CHAPTER I A. INTRODUCTION AND ACKNOWLEDGEMENTS

I A - 1. Introduction

This study begun as an investigation of the amblygonite -
- montebrasite minerals $((\text{Li},\text{Na})\text{AlPO}_4(\text{F},\text{OH}))$ in the Tanco (Chemalloy)
pegmatite at Bernic Lake, S. E. Manitoba, with the primary goal of
investigating their occurrence and distribution in this pegmatite,
and to establish the changes in the chemical composition of these
minerals, especially with reference to F : (OH) ratios, during the
evolution of the pegmatite. This research is a part of a general
mineralogical study of the Tanco pegmatite deposit, being carried out
at the Department of Earth Sciences, University of Manitoba.

From the very beginning, a need for indirect estimates of the
(F,OH) content had been felt because of the large amount of material
which needed to be examined. The X-ray powder diffraction work was
very promising in this respect, since the powder patterns showed
striking differences in d-spacings of some reflections due to different
F-contents. The paper published by A. A. Moss, E. E. Fejer, and P. G.
Embrey in the fall 1969 fully confirmed this opinion. This paper provided
the basic idea which has been worked on in this study, but did not
render the present work useless. In the present investigation the X-ray
powder diffraction study has been continued on a more detailed and
wider basis, and accompanied by single-crystal X-raying, determination

of specific gravity, and differential thermal analysis.

Thus there were two main objectives of this study: (1) to correlate the physical properties quoted above with F-contents and to establish a simple reliable method for estimating the F-contents, and (2) to study the distribution of amblygonite - montebrasite in the Tanco pegmatite and to establish the changes in the chemical composition of these phases during the evolution of the pegmatite.

The correlation of physical properties with the F-contents is introduced in the following Chapter I A, and discussed in Part II. Part III contains the paragenetical and geochemical treatment of amblygonite - montebrasite in the Tanco (Chemalloy) pegmatite, with a general introduction to this problem presented in Chapter I B.

I A - 2. Acknowledgements

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Messrs. K. Ramlal, research chemist, and R. M. Hill, chemical technician, from the Department of Earth Sciences, University of Manitoba, did much of the chemical work. Dr. P. Povondra, Geological Institute of

the Czechoslovak Academy of Sciences, analyzed five specimens. Dr. E. H. Nickel from the Mines Branch, Department of Energy, Mines and Resources, Ottawa, drew our attention to the possibility of obtaining fluorine determinations by neutron activation in the Mines Branch, and all other fluorine determinations have been performed by Mr. J. L. Dalton through the courtesy of Mr. W. A. Haw and Dr. A. H. Gillieson from the same institution.

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CHAPTER I B. MINERALOGY OF THE AMBLYGONITE - MONTEBRASITE GROUP

Amblygonite and montebrasite are phosphates of lithium and aluminium with respective formulae LiAlPO_4F and $\text{LiAlPO}_4(\text{OH})$. Table 1 gives a summary of the characteristics of all the minerals of this group known to date, to provide a quick reference to their properties.

The first mineral of this group, amblygonite, was investigated and named in 1817 by Breithaupt by coupling the Greek words amblys (blunt) and goni (angle). Montebrasite was first described considerably later, in 1871, by Des Cloizeaux, and named after its type locality in Montebras, France. The name hebronite has been used for members with an intermediate composition, that is, with an F/OH ratio close to 1, but the term seems to be rather unnecessary.

Several names have been applied to sodium-rich montebrasite, namely natronamblygonite, fremontite and natro montebrasite. The nomenclature proposed by Palache et al. (1951), accepted by Strunz (1957), and shown in Table 1, seems to be the most widely accepted, whereas the later proposal by Strunz (1966) seems to be oversimplified and burdened by an obsolete term: he uses amblygonite for the whole (F,OH) series, and natronamblygonite for the sodium-rich montebrasites - a term rejected already by Schaller in 1914.

Tavorite, a rare ferric member of the group, has been found relatively recently (Lindberg and Pecora, 1954, 1955; Strunz and Fisher 1957), and since its occurrences, paragenesis and mode of origin are quite different from those of the other members of the group important

TABLE 1

MINERALOGICAL CHARACTERISTICS OF THE AMBLYGONITE - MONTEBRASITE - TAVORITE GROUP

(summarized from Palache et al.(1951) and Vlasov et al. (1964) if not indicated otherwise)

LiAlPO_4F	amblygonite					
$\text{LiAlPO}_4(\text{OH})$	montebrasite					
$(\text{Na,Li})\text{AlPO}_4(\text{F,OH})$	natrc_montebrasite (natronamblygonite, fremontite) (1)					
$\text{LiFePO}_4(\text{OH})$	tavorite					

Crystal system and Class : triclinic, pinacoidal $\bar{1}$

Unit cell dimensions	$a_o(\text{\AA})$	$b_o(\text{\AA})$	$c_o(\text{\AA})$	α	β	γ
Amblygonite	5.15	7.21	5.06	114.0°	98.6°	67.3°
Montebrasite	5.19	7.17	5.04	112.5°	97.9°	67.8°
Tavorite	not determined					

Structure :

The unit cell contains 2 molecules of $\text{LiAlPO}_4(\text{F,OH})$. Both aluminum atoms are octahedrally coordinated to 4 oxygens and 2 $(\text{F,OH})_4$ atoms and lie in two special positions, one in the corners of the unit cell, and the other in $0, \frac{1}{2}, \frac{1}{2}$. P is tetrahedrally coordinated, surrounded by 4 oxygens. The position of Li can be considered as in the centre of a trigonal dipyramid formed by 4 oxygens and 1 (F,OH) atom.

Table 1 - continued

Habit :	equant and short prismatic [010] crystals rare, large cleavable masses, columnar aggregates		
Crystal forms :	at least 19 forms known		
Twinning :	common on $\bar{1}11$, rare lamellae on 111		
Cleavage :	100 perfect, 110 good, $0\bar{1}1$ distinct, 001 imperfect; fracture uneven to conchoidal; brittle		
Hardness :	$5\frac{1}{2}$ - 6		
Specific gravity :	3.11 amblygonite	2.98 montebrasite	(6)
	3.04 - 3.11 natronamblygonite	3.30 tavorite	
Lustr ^e _A :	vitreous to greasy		
Colour :	waterclear to milky white, creamy white, yellow, beige, salmon pink, greenish, pale violet, gray, brownish, always in pale shades; transparent to translucent. Tavorite - greenish to yellow (6).		

Table 1 - continued

Optical properties :	α	β	γ	sign	2V
Amblygonite ⁽⁴⁾	1.572	1.587	1.589	(-)	medium
Montebrasite ⁽⁴⁾	1.605	1.621	1.636	(+)	large
Natronmontebrasite ⁽⁵⁾	1.594	1.603	1.615	(+)	large

Optical orientation varies with changes in chemical composition but is not known for the whole amblygonite - montebrasite series.

Chemistry : as indicated by the formulae, a continuous series seems to exist between amblygonite and montebrasite; natro_montebrasite never approaches the end-member; tavorite does not show any appreciable solid solutions with others.

Occurrences : mainly in Li-bearing granitic pegmatites; also in high-temperature tin veins, greisens, and in special amblygonite - Sn-sulphide deposits; tavorite rare in pegmatites.

(1) For details and history see Palache et al. (1951), Heinrich and Corey (1955), Strunz (1957), Strunz (1966)

(2) Moss et al. (1969)

(5) Palache et al. (1951, p.824)

(3) Palache et al. (1943)

(6) Lindberg and Pecora (1955), Strunz and Fisher (1957)

(4) Extrapolated from Winchell and Winchell (1951)

for this study, it has not been investigated here.

In a mineral species or series showing ionic substitutions, the relation between the chemical composition and physical properties is always of primary interest to mineralogists, and these relationships have been studied in the amblygonite - montebrasite group. The substitution of fluorine and hydroxyl is of primary importance in this group, but the sodium percentage in lithium sites can also attain considerable values. However, specimens with appreciable amounts of sodium (over about 1.5% Na_2O) are rare; moreover, they occur usually in small grains insufficient for complete investigation, and are usually partly altered. Thus the investigation of this substitution and its effect on physical properties has not been studied during this investigation. (With the exception of specific gravity and refractive indices which increase with the Na content of montebrasite, other properties do not seem to be appreciably influenced according to Palache et al. (1951), and Moss et al. (1969)).

The variation of refractive indices and $2V$ with the F/OH ratio has been studied by several investigators, the last and most widely used graph being that by Winchell and Winchell (1951), from an earlier paper by A. N. Winchell in 1926. However, it is based mainly on old data, it shows a considerable scatter of the plotted points, and it has raised some objections from other authors (Ginzburg 1950). It is evident that this graph needs updating, and a study of changes in the optical orientation with F/OH ratio is also desirable, the present data being disseminated through the literature, and being only fragmental in nature

(Palache et al., 1943; Nel, 1946; Volborth, 1951; Boruckij, 1966). A detailed optical study was not feasible during the present study because of time restrictions, although some measurements of the refractive indices γ have been performed, as a form of check on the specific gravity values (Chapter II B - 2, II B - 3). However, the material analyzed here is suitable for such an investigation, and it would provide a valuable tool for compositional estimates, particularly because of the very small amount of material necessary.

Specific gravity has been also related to the F/OH substitutions (Chapter II B - 2). Determination of the specific gravity is not as valuable for an indirect estimate of a F/OH ratio as the refractive indices, since the amount of material necessary is much larger, the method is not so sensitive, and the determination is affected by alterations and/or primary inclusions. The specific gravities measured during this study are presented and discussed in Chapter II B - 2.

Distinction within the amblygonite - montebrasite series by X-ray diffraction has been used by Pough and Henderson (1945), Fisher (1958), and more recently by Quensel (1962) and Kittrick and Hope (1967), on the basis of both powder and single crystal data. However, the data published by these authors did not cover the whole substitutional series, they often involved intensity estimates, and some have not been based on chemically analyzed specimens. The full potential of the X-ray powder diffraction method has been utilized only recently by Moss et al. (1969).

The X-ray powder study needs an even smaller amount of material

than does the determination of the specific gravity, and the results are not influenced by inclusions and slight alterations. Thus X-raying can be applied more widely than the specific gravity, and it may be preferred even to optical methods for badly clouded specimens. The results obtained here are presented in Chapter II B - 5.

Differential thermal analysis of amblygonites has been described by only a small number of authors (Ginzburg in Vlasov et al., 1964; Manly, 1950; Boruckij, 1966; Ivanova, 1961), and no systematic investigation of the F/OH substitutional series has been attempted to date. The results published by the quoted authors have shown considerable variations in thermal behaviour can be expected in the amblygonite - montebrasite group. With appropriate equipment, the d.t.a. method can be used with very small amounts of material. However, several important factors such as heating rate, grain size, and the character and quantity of admixed material influence the resulting curves so substantially that any attempts to use them for the F/OH estimates should be based on comparison with a set of reference records made on well-characterized specimens analyzed under standardized conditions. All specimens chemically analyzed for F-content in this study have been examined on d.t.a. and the products after heating to 1000° C were X-rayed. Details of the method and the results are presented in Chapter II B - 4.

CHAPTER I C. PEGMATITE OCCURRENCES OF AMBLYGONITE - MONTEBRASITES

Lithium-bearing granitic pegmatites are the most important rocks in which amblygonite - montebrasite minerals occur. In some instances, they can serve also as lithium ore when accumulated in large quantities. Their occurrence within lithium-bearing pegmatites has been shown to be restricted to several characteristic zones and assemblages (Ginzburg 1959, 1960):

They are absent in the external border and wall zones (aplitic and graphic) as well as in the blocky-feldspar + quartz zones. In pegmatites particularly rich in lithium and phosphorus, amblygonite - montebrasites originate for the first time in the pegmatite evolution in the margins of the quartz core, together with giant K-feldspars and spodumene. According to Ginzburg this assemblage is thought to belong to the last products of primary crystallization proceeding directly from the magma and supercritical solutions. The next generations of amblygonite - montebrasite occur in various metasomatic assemblages: in albitized parts of the parent pegmatite, and with lithium micas. Natro-montebrasite is characteristically found with late albite.

This general scheme of amblygonite - montebrasite generations seems to be confirmed in all well-studied localities (Ginzburg, 1950; Quensel, 1937; Heinrich and Corey, 1955; Staněk, 1965) although the detailed views of some authors may show some deviations (Volborth, 1951; Quensel, 1956).

Alterations of the amblygonite - montebrasite minerals are also well known and rather widespread. Besides being replaced by other

amblygonite - montebrasite types, they give rise to lithium micas, cookeite, clay minerals, and mixtures of those during late stages of pegmatite evolution (Ginzburg, 1950; Landes, 1925; Quensel, 1937; Staněk, 1965). In many geological situations when the pegmatites have been exposed to long and/or deep surficial weathering, the sensitivity of amblygonite to atmospheric agents and its readiness to decompose under their influence are the main obstacles in its study.

The fact that several varieties of amblygonite - montebrasite may occur in a single pegmatite body was shown first by Des Cloizeaux in 1873. Heinrich and Corey (1955, p.1144) list several earlier authors who have confirmed it, including Landes (1925), Quensel (1937), and Palache et al. (1943); more recently Ginzburg (1950), Volborth (1951), Heinrich and Corey (1955), Boruckij (1966), Gallagher (1967), and Moss et al. (1969) have found the same in other localities. The differences in F/OH ratio and/or the Li/Na ratio are the principal ones among different generations found in different pegmatite units and mineral assemblages. As to the replacement of amblygonite - montebrasite by another member of this group, it has been observed by Ginzburg (1950) and explained as a consequence of proximity to late albite, causing a replacement of the earlier montebrasite by natro-montebrasite. Staněk (1965) accepts this idea, too.

Studies of changes in the F/OH and Li/Na ratios in amblygonite - montebrasite from different zones in a lithium-bearing pegmatite are rare. This is not surprising because of many difficulties such a study faces: it can be carried out only in well-opened and thoroughly studied pegmatites containing appreciable amounts of fresh amblygonite, and it

is necessary to have a suitable direct or indirect method for determining the fluorine content of large numbers of specimens. The only papers found in the literature that deal with the problem are those by Quensel (1937, 1956) and Boruckij (1966).

Quensel (1937) described three varieties of montebrasite from the Varutrask pegmatite (in the sequence of formation): small crystals disseminated in quartz and muscovite (4.42%F, 0.95% Na₂O), large rounded blocks in coarse quartz, K-feldspar and spodumene (2.10%F, 0.38% Na₂O), and rare salmon-pink grains in cleavelandite and petalite (1.80% F, 2.17% Na₂O). However, in a later paper Quensel (1956) changed this sequence, claiming that the second type is the first formed, in the last stages of primary crystallization of the core; the first fluorine-richest type was allocated to the high-temperature lithium metasomatism, and the third sodium-rich type to the low-temperature sodic metasomatism assemblage.

Boruckij (1966) described amblygonite - montebrasite from small intensely albitized pegmatite veins from Siberia. This amblygonite is of an early generation forming large blocks in the quartz core, and has a composition with F/OH very close to 1. Later varieties from fine-grained metasomatic and recrystallization units show higher refractive indices increasing towards the margins. This indicates that the late montebrasite is progressively enriched in hydroxyl and its F-content decreases.

Boruckij's study shows considerable variations in the F/OH ratio in very small pegmatite veins. This suggests that the few specimens examined by other authors could hardly be taken as representative

for all amblygonite - montebrasite in their parent assemblage. A very broad statistical basis is evidently necessary for such an investigation, particularly in large pegmatite bodies.

In the Tanco (Chemalloy) mine, only a part of the pegmatite has been accessible but included all zones and replacement units composing the whole body. Thus the results of this investigation can be taken as representative of the complete pegmatite evolution at this locality. The sampling, laboratory examination, distribution of the amblygonite - montebrasite minerals and their composition are presented in Chapters III A to III C, and the genetic conclusions are discussed in Chapters III D and III E.

PART II. PHYSICAL PROPERTIES AND FLUORINE CONTENT

OF AMBLYGONITE - MONTEBRASITE

CHAPTER II A. SOURCES AND DESCRIPTIONS OF SPECIMENS

For this whole research project 157 samples collected at the Tanco pegmatite and 62 amblygonite - montebrasite specimens from the collections of various institutions were used. Appendix I describes all the specimens sampled in the Bernic Lake pegmatite, and Appendix II lists those obtained from other sources. Brief descriptions and X-ray estimated fluorine contents in weight percentages accompany each entry, as well as the determined fluorine contents when analyzed for.

After a preliminary X-ray check of the F-contents of all specimens, twelve covering the widest possible range of F-contents were chosen for detailed study. Five samples analyzed prior to the beginning of this study by Dr. P. Povondra, Geological Institute, Czechoslovak Academy of Sciences in Prague, were coupled with this series. In most instances, specimens were found, that yielded a large amount of fresh, homogeneous-looking material from a single hand specimen. In some cases, however, even small and clouded fragments have had to be used because of the lack of better specimens with a given composition, particularly for compositions close to the high-fluorine end-member.

Despite the careful examination of all these specimens prior to the detailed study, later investigation showed that some specimens

are not homogeneous. Besides this, more specimens became of doubtful suitability for the correlation of physical properties and F-content, because of the later necessity to use 10g samples for fluorine determination by neutron activation instead of the more homogeneous 3g samples prepared for chemical analysis by the usual methods and for other examination. For these reasons a detailed description of all 17 specimens used for correlating physical properties and F-contents is given in Table 2, to illustrate the material used in this study as well as to show the difficulties with separation of homogeneous material that may be encountered in any examination of the amblygonite - montebrasite minerals. The agreements and anomalies among the physical properties and F-contents of individual specimens are also discussed in Table 2; this involves the use of some results and combinations described later which cannot be avoided without confusing dispersing of this condensed review over all sections of the Chapter II B.

TABLE 2

DESCRIPTIONS OF SPECIMENS ANALYZED FOR FLUORINE

AND DISCUSSION OF THEIR CORRELATIONS WITH PHYSICAL PROPERTIES

- Specimen A - 1 Pale pink zone of a big zoned crystal. Complete analysis shown in Table 3. Fairly homogeneous with low amount of fluid inclusions as seen in immersion liquids. However, thin sections reveal very fine veinlets of higher-refracting phase with uniform optical orientation which slightly differs from that of the matrix. This is most probably a F-poorer secondary montebrasite. Thus the specific gravity agrees well with the F-content, although it varies with the amount of inclusions, but the unit cell dimensions and the group of characteristic reflections favour a slightly higher F-content, being based on X-raying which is governed by the predominant matrix. The same is indicated by the refractive index γ' .
- $C \approx 5.56\% \text{ F}$
- Specimen A - 2 Yellow outer zone of the same crystal as A-1. Fluid inclusions rather numerous, the specific gravity seems to be lowered by them. Unit cell dimensions show good agreement, as well as the refractive index γ' . The DTA pattern fits the continuous changes in neighbouring specimens.
- $C = 3.65\% \text{ F}$
- Specimen A - 3 Waterclear crystal. No inhomogenities found under microscope. Unit cell dimensions, refractive index γ' , specific gravity and the group of characteristic reflections agree very well with the content of 6.30% F determined by J. L. Dalton; 5.63% F found by I. Hrejsová is evidently too low.
- $N \approx 6.30\% \text{ F}$
 $C = 5.63\% \text{ F}$
- Specimen A - 4 White material surrounding and veining A-5. Low amount of fluid inclusions is the only inhomogeneity found. Unit cell dimensions, group of characteristic reflections, refractive index γ' and specific gravity compare well
- $N = 4.05\% \text{ F}$
 $C = 3.65\% \text{ F}$
- * C - wet chemical determination
N - neutron activation

with 4.05% F found by J. L. Dalton, the content of 3.65% F determined by I. Hrejsová is most probably lower than the actual percentage. The DTA pattern fits the continuous changes in neighbouring specimens.

Specimen A - 5 Yellow material in the core of the same specimen as A-4.

C = 4.46% F

Specific gravity is distinctly higher than appropriate for 4.46% F but the X-ray data and refractive index γ' seem to favour a slightly lower F-content. The amount of fluid inclusions is very low, no other inhomogeneities have been observed and the content of Na_2O and other cations is negligible (Table 3). No explanation can be found for these discrepancies.

Specimen A -13 White to pinkish cleavable mass. The X-ray data, refrac-

N = 6.43% F

tive index γ' and particularly the specific gravity favour lower F-content than the analyzed value of 6.43% F. However, the amount of fluid inclusions is very low, and the individual measurements of specific gravity are grouped so well that they seem to exclude any chances of accidental imperfections in some grains. No explanation available.

Specimen A -22 Pale brownish material with poor cleavage and dull luster,

N = 1.40% F

secondary after a white, F-rich phase. Good agreement of specific gravity and refractive index γ' with the F-content of 1.40% but X-ray data prefer a slightly higher F-content. This can be explained by ^{different} degrees of purity of the material used in individual measurements. The character of the DTA record shows that this specimen is the F-poorest sample in the analyzed series.

Specimen A -29 White cleavable mass similar to A-3 but somewhat milky.

N = 6.30% F

X-ray data, specific gravity, DTA records and products favour a slightly lower F-content than the determined value, particularly in direct comparison with the sample A-3. However, the difference would be lower than 0.5% F.

The content of fluid inclusions is rather high, and this could influence the most differing value of specific gravity.

Specimen A -60 Milky white cleavable fragment. X-ray data agree well with the analyzed F-content but refractive index γ' , differential thermal behaviour and products would fit better a slightly higher F-percentage. On the other hand, the specific gravity values seem to suggest a lower F-content. However, the grains showing the most deviating low values are particularly rich in fluid inclusions that are much less abundant in heavier grains. Thus the majority of physical properties favours a higher F-content than that of the sample A-29. This may be well possible since the difference between them is only 0.13% F.

Specimen A -72 Beige fragment with barely perceptible variation in luster, milky clouding and character of cleavage. Only after finding out the discrepancies between the analyzed F-content of 3.17 and the physical properties, a detailed optical check revealed the presence of two phases with the respective values of $\gamma' = 1.616$ and 1.634. These values correspond, when compared to other specimens studied here, to minerals containing about 2.2 and 6.2% F, respectively. The unit cell dimensions would fit approximately 2.5% F, the DTA pattern and products are close to those of A-22 and AF-43. Thus these experiments were undoubtedly carried out with material containing predominantly the higher refracting F-poor phase.

Specimen A -98 Pale yellow translucent mass with indistinct cleavage, very clear with few fluid inclusions under microscope. The X-ray data, refractive index γ' , DTA record and specific gravity would correspond better to a mineral with slightly higher F-content than the analyzed value of 3.44%

but the possible difference seems to be well inside the analytical error.

Specimen A-126 Pale pink zone of the same big crystal which yielded the specimen A-1. The F-content of 5.66 by J. L. Dalton matches perfectly with that of 5.56% F determined on A-1 by I. Hrejzová. Also the optical check gave the same results. No other data have been measured on this specimen.

Specimen AF-1 Milky white opaque mass. Stuffed with anisotropic inclusions that could not be determined and are probably the main cause of very low specific gravity. Nevertheless, the X-ray data fit the analyzed F-content, and the refractive index γ^1 would favour even higher F-percentage. This can be explained only by a very large difference between the specific gravities of the amblygonite as such and its inclusions.

Specimen AF-43 Grayish-white cleavable fragment. It contains some fluid inclusions and an unidentified anisotropic phase in very small particles. X-ray data and optical characteristics agree well with the determined 1.88% F, the specific gravity favours seemingly a higher fluorine content. The DTA pattern fits the continuous changes in neighbouring specimens.

Specimen AF-47 Waterclear equant crystals with bluish tint, etched on crystal faces, with platy albite in pegmatite cavities. Because of lack of material, the F-content could not be determined to date. The X-ray data and the γ^1 -index correspond to a montebrasite with about 1.5% F but the specific gravity seems to indicate approx. 4% F. May be it is influenced by a higher Na_2O -content; this can be solved only by a complete chemical analysis.

Specimen AF-48 Clear platy crystals from a pegmatite cavity. The determination of the F-content has not been feasible to date

because of lack of material. X-ray data and refractive index γ' indicate about 1.4% F, the specific gravity suggests again a higher value of 2% but the discrepancy is much smaller than in the previous case.

Specimen AF-55 Colourless cleavable mass with numerous veinlets of milky white material. The colourless mineral shows low $\gamma' = 1.596$ and irregularly disseminated fluid inclusions and anisotropic particles. White veinlets consist of a similar phase with higher $\gamma' = 1.605$ and with different optical orientation passing gradually into colourless material, and is stuffed with undetermined anisotropic inclusions. The white phase is undoubtedly a F-poorer alteration product of the primary colourless amblygonite. No wonder that the unit cell dimensions and the group of characteristic reflections, determined on small and pure amount of the colourless phase, indicate a higher F-content than the analyzed value of 10.17%. The best fit would be reached at 11.0% F. Specific gravity shows a broad scatter of individual measurements, caused undoubtedly by various degree of contamination of the grain used. Lack of specimens in the F-rich half of the series does not allow any correlations with the neighbouring specimens based on DTA data.

Specimen AF-65 Translucent fragments with faint bluish tint, part of the specimen Z of Moss et al. (1969). Some of the grains used for specific gravity determination suggest a lower F-content but the refractive index γ' and X-ray data agree well with the analyzed value.

11.8% F
(Moss et al., 1969)

CHAPTER II B. EXPERIMENTAL METHODS, RESULTS AND DISCUSSION

II B - 1. Chemical composition

Of the twelve specimens chosen for detailed examination 3g of each were prepared for complete chemical analyses and X-ray powder diffraction study. Due to technical difficulties, the complete analyses were not available prior to writing this thesis, but it was possible to analyze large 10g samples for fluorine. Thus the present study is based only on weight percentages of F, which is together with (OH) the chief variable component. However, all the results will be re-evaluated once the complete analyses become available, and the 5 specimens from the Tanco mine, analyzed independently prior to the beginning of this study, will be also included.

In the twelve specimens, fluorine contents were determined by neutron activation by J. L. Dalton in the Mines Branch, Department of Energy, Mines and Resources, Ottawa and the results are shown in Appendix I and II and in Table 2. The above-mentioned five complete analyses have been performed by Dr. P. Povondra and I. Hrejsová in the Geological Institute, Czechoslovak Academy of Sciences, Prague. Potentiometric titration was used to determine alumina, and colorimetric methods were used for iron, manganese and magnesium. Lithium, sodium and calcium were determined by flame photometry, and phosphorus and water gravimetrically. Fluorine was extracted by distillation and determined colorimetrically. These five complete analyses are shown in Table 3.

Besides this R. M. Hill of the Department of Earth Sciences,

TABLE 3

COMPLETE CHEMICAL ANALYSES OF FIVE SPECIMENS OF AMBLYGONITE-MONTEBRASITE

FROM THE TANCO PEGMATITE

	A - 1	A - 2	A - 3	A - 4	A - 5
Al_2O_3	34.53 %	34.53 %	34.63%	34.45 %	34.86 %
Fe_2O_3	0.11	0.014	0.007	0.020	0.028
Li_2O	9.86	10.07	10.22	10.14	10.29
Na_2O	0.054	0.025	0.039	0.055	0.044
CaO	0.17	0.049	0.22	-	0.074
MgO	0.005	0.002	0.008	-	0.002
P_2O_5	49.32	49.21	49.22	49.48	49.26
$\text{H}_2\text{O} +$	3.39	4.09	2.70	3.85	3.30
F_2	5.56	3.65	5.63	3.65	4.51
$\text{H}_2\text{O} -$	0.10	0.04	0.08	0.08	0.08
	103.099	101.68	102.754	101.725	102.44
$\text{O}\equiv\text{F}$	2.341	1.563	2.370	1.536	1.898
Total	100.758	100.117	100.384	100.189	100.542

Analysts: Dr. P. Povondra, I. Hrejsová, 1969.

University of Manitoba, analyzed 32 specimens for Na_2O , K_2O and CaO by atomic absorption. These specimens were chosen to represent all types of amblygonite and montebrasite in the Tanco pegmatite and are discussed in Part III dealing with the paragenesis and geochemistry of these minerals.

II B - 2. Specific gravity

II B - 2a. Methods and results

The specific gravity of the 17 specimens has been measured by means of the Berman torsion balance, using toluene checked for temperature as a suspension medium. The purest grains available have been selected from each specimen for this determination. However, in some cases only milky, clouded grains were available. Microscopic examination has shown that liquid inclusions were the main impurity in all cases, with clay-like and/or mica-like particles observed only occasionally. Both these admixtures should decrease the specific gravity of amblygonite - montebrasite.

Table 4 gives the results obtained in this study. All of them fall between the end-member values 2.98 and 3.11 quoted in Palache et al. (1951), showing a range of mean values 2.992 - 3.094. The refractive indices and homogeneity of the measured grains were checked, particularly of those showing larger variations in specific gravity of a single sample. In most cases the γ' index was found to be constant in all grains of one specimen, suggesting a constant composition for the amblygonite - montebrasite as such, but the content of impurities was mostly different

TABLE 4. SPECIFIC GRAVITIES AND REFRACTIVE INDICES γ'

Specimen	F% wt.	Sp.gr.	ϕ	Comments	γ'
A - 1	5.56	3.040	3.037		1.615
		3.032	+ .010	fluid inclusions	1.615
		3.029	-	fluid inclusions	1.615
A - 2	3.65	2.998	3.008	fluid inclusions	1.625-6
		3.013	+ .008	fluid inclusions	1.625-6
		3.013	-	fluid inclusions	1.625-6
A - 3	6.30	3.030	3.048	clear	1.614
		3.071	+ .020	clear	1.614
		3.043	-	clear	1.614
A - 4	4.05	3.022	3.022	few fluid inclusions	1.624
		3.019	+ .002	few fluid inclusions	1.624
		3.024	-	few fluid inclusions	1.624
A - 5	4.51	3.042	3.037	few fluid inclusions	1.626
		3.046	+ .008	few fluid inclusions	1.626
		3.022	-	few fluid inclusions	1.626
A - 13	6.43	3.042	3.032		1.616
		3.025	+ .009	few fluid inclusions	1.616
		3.024	-	few fluid inclusions	1.616
		3.039			1.616
A - 22	1.40	2.987	2.992	few fluid inclusions	1.638
		2.997	+ .005	abundant fluid inclusions	1.638
		2.991	-	abundant fluid inclusions	1.638
A - 29	6.30	3.031	3.039	abundant fluid inclusions	1.616
		3.042	+ .007	abundant fluid inclusions	1.616
		3.043	-	abundant fluid inclusions	1.616
A - 60	6.17	3.045	3.031		1.614
		3.030	+ .012	fluid inclusions	1.614
		3.022	-	abundant fluid inclusions	1.614
		3.043		abundant fluid inclusions	1.614
		3.016		abundant fluid inclusions	1.614

Table 4 - continued

Specimen	F% wt.	Sp.gr.	ϕ	Comments	γ'
A - 72	3.17	3.015	3.008	few fluid inclusions	1.634
		2.993	+ .003	few fluid inclusions	1.634
		3.020	-	few fluid inclusions	1.634
		3.004		few fluid inclusions	1.634
A - 98	3.44	3.029	3.022	clear with few bubbles	1.626
		3.019	+ .005	clear with few bubbles	1.626
		3.024	-	clear with few bubbles	1.626
AF - 1	7.85	3.000	3.007	very abundant anisotrop.incl.	1.605
		2.997	+ .010	very abundant anisotrop.incl.	1.605
		3.018	-	very abundant anisotrop.incl.	1.605
		3.004		very abundant anisotrop.incl.	1.605
		3.019		very abundant anisotrop.incl.	1.605
AF - 43	1.88	3.011	3.005	few fluid and anisotrop.incl.	1.636
		3.017	+ .014	few fluid and anisotrop.incl.	1.636
		3.008	-	few fluid and anisotrop.incl.	1.636
		2.985		few fluid and anisotrop incl.	1.636
AF - 47	1.5*	3.025	3.024	clear	1.636-7
		3.028	+ .010	clear	
		3.033	-	clear	1.636-7
		3.009		clear	1.636-7
AF - 48	1.4*	3.003	3.004	clear	1.637-8
		3.006	+ .001	clear	1.637-8
		3.003	-	clear	1.637-8
AF - 55	10.17	3.054	3.086	very abundant opaque incl.	1.595-6
		3.077	+ .022	very abundant opaque incl.	1.595-6
		3.109	-	very abundant opaque incl.	1.596
		3.084		very abundant opaque incl.	1.596
		3.105		very abundant opaque incl.	
AF - 65	11.8	3.090	3.094		1.592-3
		3.107	+ .012		1.592-3
		3.083	-		1.592-3

* - F-contents estimated

in different grains. The detailed discussion of the refractive index γ is given in the next section II B - 3.

II B - 2b. Discussion

The relationship between specific gravity and fluorine content is illustrated in Fig. 1. Generally, the averaged points fall close to the line which connects the specific gravities of montebrasite (2.98) and amblygonite (3.11) end-members (according to Palache et al. 1951): eight samples out of 14 analyzed lie within ± 0.005 (or $\pm 0.5\%$ F), and five other specimens within ± 0.01 (or $\pm 1\%$ F). The only exception deviating remarkably is the sample AF-1.

It should be noted, however, that, as Table 4 shows, measurements of different grains from a single specimen show considerable spread, and the averaging of values of three to five grains seems to be the only way of obtaining a satisfactory value. In many cases, the measurement of a single grain was repeated several times and the differences were always negligible, within ± 0.0015 . Thus the considerable variations among different grains appear to be real, caused most probably by fine air-filled cracks, various inclusions, and/or chemical inhomogeneity.

The writer's experience appears to confirm the views of A. Nicolas (1968) who proved experimentally the important role of inclusions and other crystal imperfections in slight variations of specific gravity, predominant over the possible variations of chemical composition of the host phase. The data obtained here support the relation between the specific gravity and fluorine contents observed by earlier authors, and

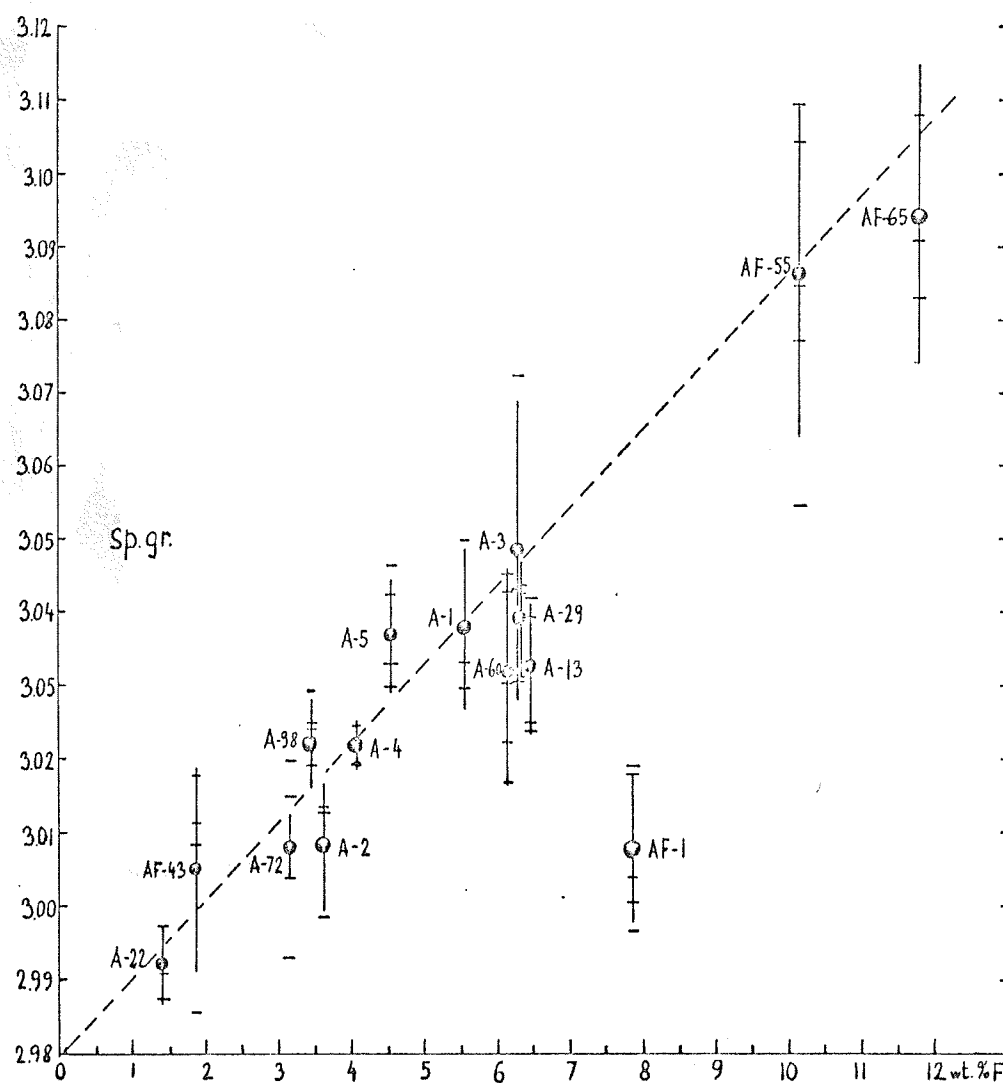


Fig.1. Relation of specific gravity of amblygonite-montebrazite to its fluorine content (in wt.% F). Horizontal bars - individual determinations; dots - averages; vertical lines - standard deviations; straight line connects the end-member values of 2.98 and 3.11 after Palache et al.(1951).

the writer's observations show that multiple measurements of carefully selected material should be made in any attempt to estimate F-contents of amblygonite - montebrasite from its specific gravity.

II B - 3. Refractive index γ'

II B - 3a. Method and results

The refractive index γ' has been measured in white light by the Becke method, using a set of immersion liquids spaced at 0.002 and 0.004. The refractive index of the mixed liquids was measured by means of a Zeiss refractometer. The γ' index has been determined on crushed grains immersed in a liquid, as the highest index measured on fragments showing highest birefringence. Thus the measured values were not obtained on exactly oriented grains, and the γ' index could be in some cases slightly higher (up to about 0.002). The use of white light in microscopic work may cause some error in the measured values because of the dispersion of refractive indices, particularly noticeable in the F-rich specimens. However, repeated measurements and observations in different closely spaced liquids have shown that this error should be lower than 0.002.

The measured values are presented in Table 4, and plotted against the F-content in Fig. 2.

II B - 3b. Discussion

Despite the preliminary character of these measurements and use of white light, the data show a good linear relationship of the two

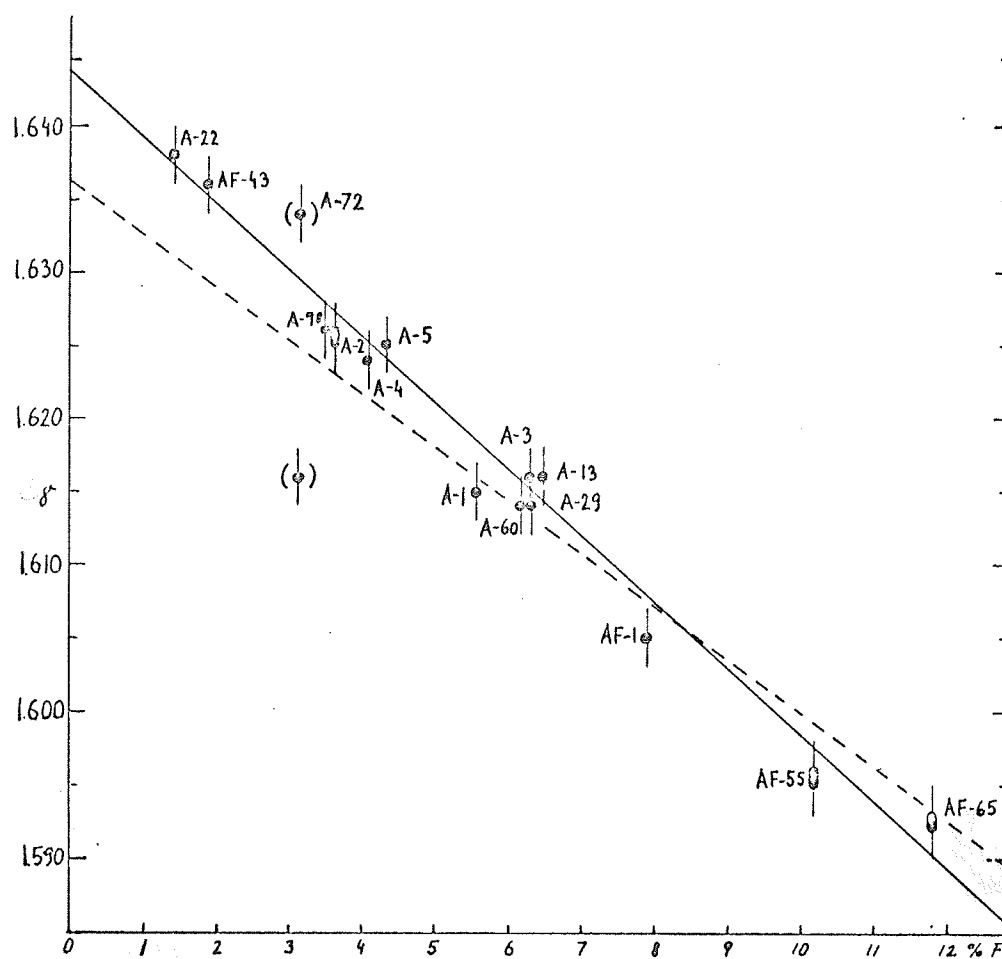


Fig.2. Relation of refractive index n_y of amblygonite-montebrazite to its fluorine content (in wt.% F). Vertical lines - estimated maximum errors; dashed line - after Winchell and Winchell (1951); full line - estimated linear correlation of the present data. Plots in brackets belong to two phases of the specimen A-72.

variables. The extrapolated values for the end-members are 1.644 for montebrasite and 1.586 for amblygonite.

The plotted points show much smaller spread around the estimated linear relationship than data used by Winchell and Winchell (1951) in their graph. Their averaged line, shown also in Fig. 2, shows a smaller slope than that derived here and indicates considerably lower indices for the OH-rich part of the series, with their $\gamma^i = 1.6365$ extrapolated for pure montebrasite. This difference could not be caused by some errors in the present measurements, since the only possible systematic error may slightly decrease the indices obtained here. Besides this, the γ^i values for the OH-rich members which differ most from Winchell's are supported by recent data of Gallagher (1967). His montebrasite from Rhodesia containing 1.29% F shows the refractive index $\gamma = 1.638 \pm 0.001$, which is the same as for sample A-22 with 1.40% F.

This preliminary plot should be made more precise in the future by measurements on perfectly oriented grains in monochromatic light, and all optical properties should be checked in the same way on recently analyzed specimens. The example of the indices γ shows that the optics of the amblygonite - montebrasite group need a review.

II B - 4. Differential thermal analysis

II B - 4a. Methods and results

The thermal behaviour of 11 specimens has been studied using the automatic apparatus DTA - 13M, R. L. Stone Co., Austin, Texas. Samples were prepared by crushing and separating the 0.104 - 0.177 mm

fraction between the 150 and 80 mesh sieves. About 0.1 g of this fraction was placed on a platinum dish which was in a wire loop in the heating chamber. Al_2O_3 was used as a reference standard, mounted in the same way in another wire loop, the two loops forming a thermocouple. The rate of heating in air was approximately $100^\circ\text{C}/10$ min. but the actual variations were negligible. The recorded curves have been corrected for unbalance in the weight of sample and standard which causes general vertical shifts, and for the changes in the conductivity of the sample during heating by deducting the record of the second run from that of the previously analyzed natural specimen.

The records are illustrated in Fig. 3, where the peak heights have been reduced to one-half the original to enable a close-packed presentation on a single page; the temperatures of the peaks are summarized in Table 5. In both Fig. 3 and Table 5, the thermal analyses are arranged according to decreasing F-content, to facilitate the discussion.

The products of the differential thermal treatment at 1000°C of nine of the specimens have been X-rayed and identified as mixtures of $\text{Li}_4\text{P}_2\text{O}_7$, high-temperature form of Li_3PO_4 , trigonal AlPO_4 (berlinite), and high-temperature cristobalite-type and tridymite-type AlPO_4 , in various proportions in different samples.

II B - 4b. Discussion

The differential thermal curves obtained in this study agree roughly with those published by Ivanova (1961) and Boruckij (1966), but

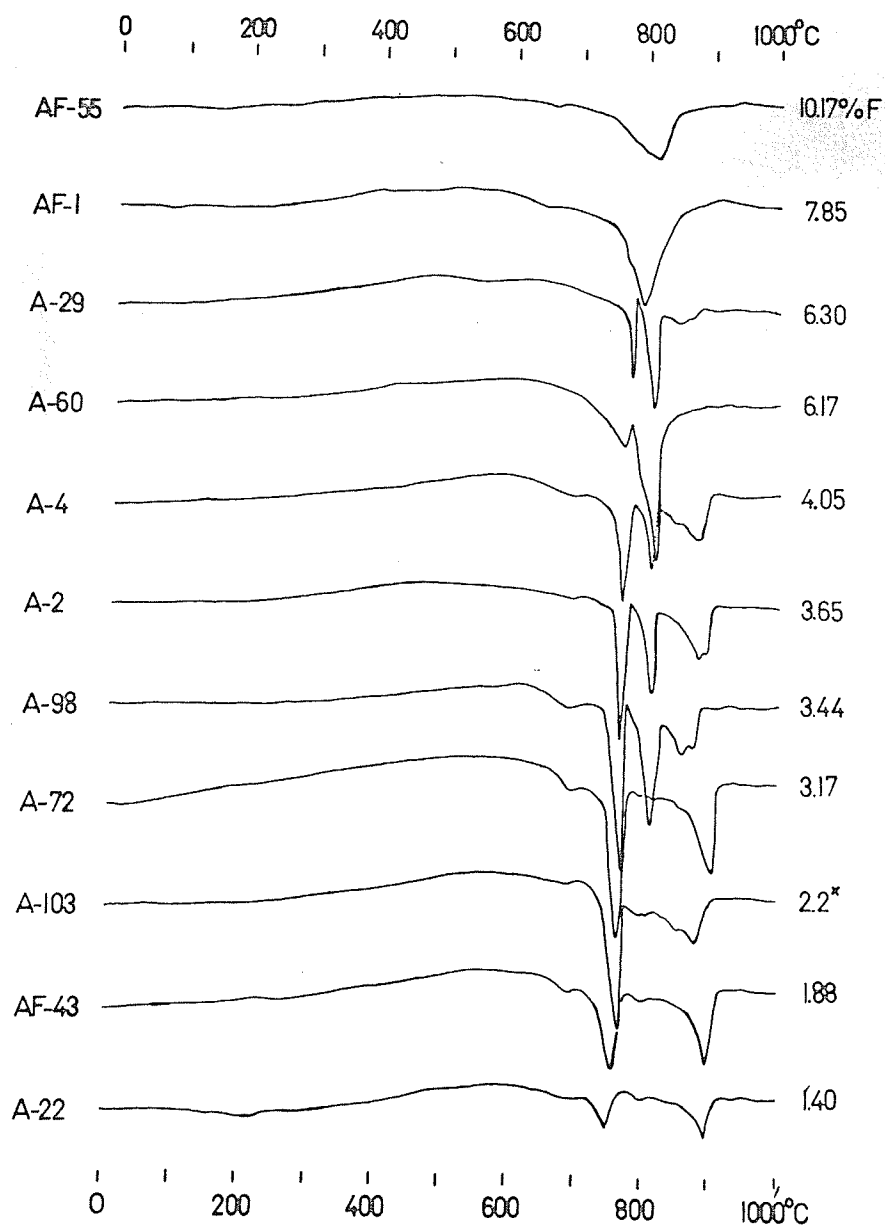


Fig.3. Differential thermal curves of selected amblygonite-montebra-site specimens arranged according to the decreasing F-content.

x - F-content estimated by X-ray powder diffraction method

TABLE 5

TEMPERATURES OF DIFFERENTIAL THERMAL REACTIONS

Specimen	F% wt.	700°C	800°C	900°C
AF- 55	10.17	(657)	(793) <u>811</u>	
AF- 1	7.85	(636)	(780) <u>797</u>	
A - 29	6.30		780 <u>810</u> (854)(870)	
A - 60	6.17		777 <u>805</u>	
A - 4	4.05	(698)	<u>764</u> 807 (850) 879	
A - 2	3.65	(688)	<u>761</u> 808 884 895	
A - 98	3.44	(689)	<u>760</u> 812 862 879	
A - 72	3.17	(690)	<u>755</u> (797)(806) (850) 895	
A -103	2.2*	(690)	<u>760</u> (795)(805) (850)881	
AF- 43	1.88	(688)	<u>757</u> (800)	897
A - 22	1.40	(684)	751 (800)	896

* F-content estimated by X-ray powder diffraction method

() weak reaction

_____ strong reaction

show a wider variation in the number and temperature of reactions.

The curves in Fig. 3 show several characteristic changes in the reactions, all of them endothermic:

(1) A very weak reaction at 636 to 698° C, sometimes absent.

(2) The first major reaction at 751 to 793° C forms only a broad shoulder on a higher-temperature endotherm in the two F-richest specimen, then increases in intensity up to around 3% F, and finally decreases again for the F-poorest samples. The temperatures of this endotherm shift from higher to lower temperatures with decreasing F-content.

(3) A reaction at 795 - 811° C is the most prominent for the F-richest specimens, and rather broad. It sharpens and increases in intensity in the 7 - 4% F range, then fades out rapidly. In the range 3.3 - 2.5% F it becomes perceptibly doubled and then is united again.

(4) A closely-spaced doublet, absent in the two F-richest specimens, develops gradually in the 6.5 - 3.4% F range in the 850 - 879° C region. From 3.4% F downwards, the first reaction becomes reduced to a mere shoulder and broad slope on the higher-temperature reaction, which becomes much more intense for specimens with the F-content lower than 3% and occurs at 896° C.

There seems to be only one exception from the above-described trends: the sample A-60 (6.17% F) and A-29 (6.30% F) would fit the general scheme better if their positions in Fig. 3 are interchanged.

The same applies also to the phase composition of heating products discussed below (Fig.4). It is possible this changed sequence may

correspond to the true F-content of these specimens, as discussed in Chapter II A.

The products of the d.t. treatment after heating to 1000° C have been identified by X-ray powder diffractograms as berlinite AlPO_4 , the high-temperature form of Li_3PO_4 , $\text{Li}_4\text{P}_2\text{O}_7$, and tridymite- and cristobalite-type forms of AlPO_4 . The two last phases always occur in nearly equal amounts, their most prominent powder diffraction peaks being roughly of the same intensity; furthermore, these two strongest peaks are close together and for this reason the two phases are represented together in Fig. 4. This figure shows the relative amounts of individual phases as estimated from the relative intensities of their most prominent peaks, within each single X-ray diffractogram. The samples are arranged according to their decreasing F-content. The heights of columns are purely schematic, without any absolute values, and they are intended to show only the changes in mutual proportions among the four phases in different specimens.

The following observations can be made about the individual phases: (1) The high-temperature Li_3PO_4 seems to be present in roughly equal, relatively low amounts in all specimens. (2) The $\text{Li}_4\text{P}_2\text{O}_7$ compound is absent in the F-richest specimens, then it suddenly appears as the most abundant phase for intermediate compositions, and then it decreases slightly in the F-poorest minerals. (3) The cristobalite- and tridymite-type AlPO_4 phases are very subordinate to absent in amblygonite heating products, but they increase in the F-rich montebrasites and prevail in the F-poorest specimens. (4) The berlinite form of AlPO_4 is most abundant in the F-richest specimens, it then disappears in the intermediate

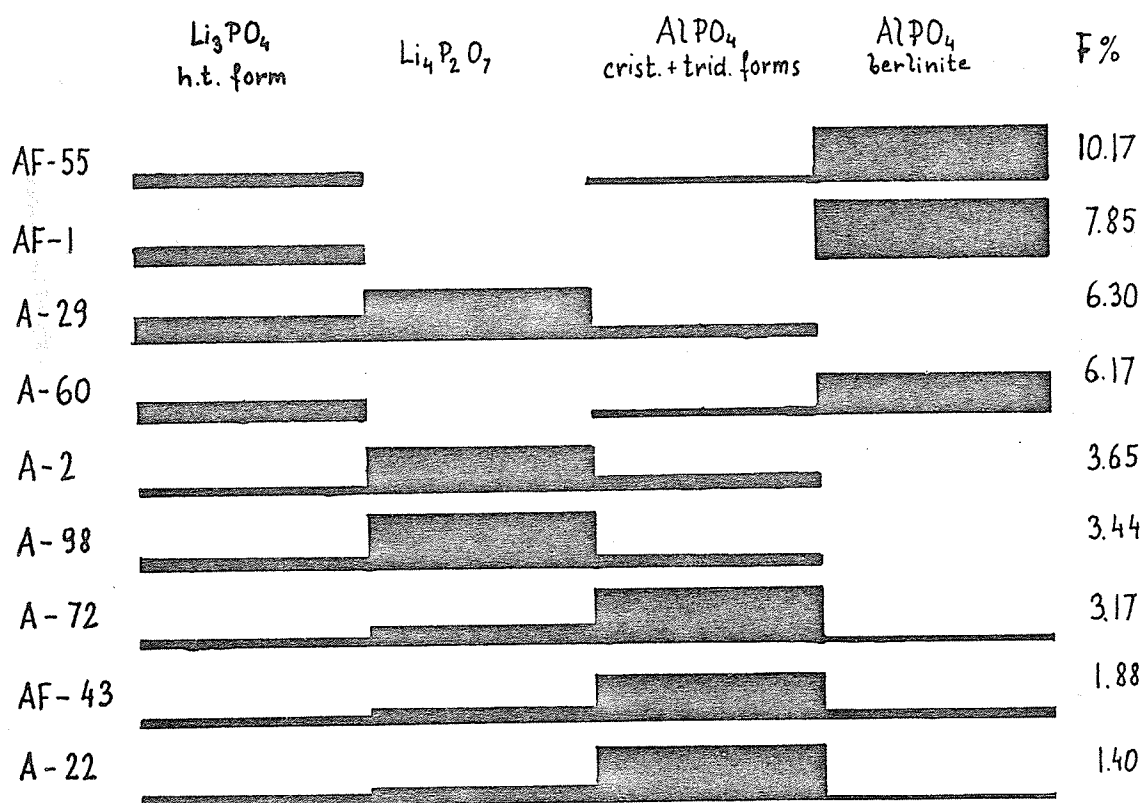


Fig.4. Heating products of selected amblygonite-montebrasite specimens after differential thermal treatment to 1000°C. Heights of columns indicate relative proportions of individual phases, estimated from relative intensities of their strongest X-ray powder reflections in individual samples.

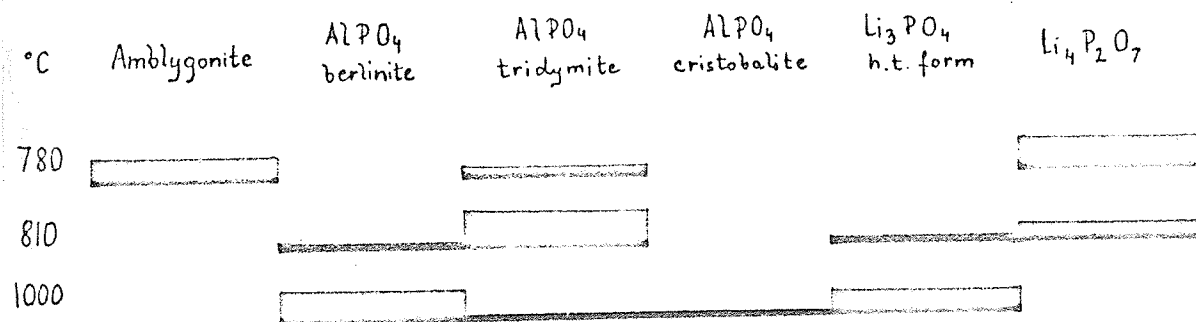


Fig.5. Heating products of sample A-60 after subsequent thermal reactions. Heights of columns indicate relative proportions of individual phases, estimated from relative intensities of their strongest X-ray powder reflections in individual samples.

compositions, and it forms again in very subordinate amounts in the F-poorest phases.

The heating products after each endotherm were X-rayed, identified, and semiquantitatively estimated for the sample A-60 with 6.17% F. The results are presented schematically in Fig. 5, and they show the complicated reactions that take place during the heating of an amblygonite - montebrasite with intermediate composition. The phase assemblage obtained at 1000° C after the differential treatment is not equilibrated. For example, a second differential thermal treatment of the d. t. a. product of sample AF-43 obtained at 1000° C showed again weak high-temperature reactions, and berlinite, which was very subordinate after the first run, appeared as the most abundant product. This suggests that the mutual phase abundances could be expected to be quite different from the author's not only after equilibrium heating at constant temperature, but also under different experimental conditions of the differential thermal analysis itself.

This is clearly documented by the d. t. a. results published by Manly (1950) and Boruckij (1966). Manly reported fusion of his sample at 850° C. Boruckij has found much of his charge unchanged at 900° C, with only a subordinate amount of a heating product optically different from amblygonite - montebrasite. Unfortunately, no details about the equipment and experimental conditions are given by these authors.

Besides the possible errors in the determination of the individual members of the amblygonite - montebrasite series, the evident sensitivity of the d. t. behaviour of the amblygonite - montebrasite

site minerals to the experimental setting presumably explains the difference between the results obtained here and those published in the literature. The curve given by Boruckij (1966) for a hebronite-type mineral with $F/OH \doteq 1$ shows a prominent reaction at $680^{\circ} C$. Manly's (1950) record of montebrasite with approximately 2.5% F corresponds more closely to our curve for a mineral with approx. 4% F, except that his sample fused at $850^{\circ} C$. The montebrasite record by A. I. Ginzburg (in Vlasov et al., 1964) agrees well with the present curve for a hebronite-type amblygonite - montebrasite phase. Ivanova (1961) published a montebrasite record comparable with those in Fig. 3, but her amblygonite curve corresponds rather to a hebronite phase with $F/OH = 1$.

Thus the d. t. a. method proves to be too sensitive to experimental setup to be useful for determination purposes. It can be safely used only under a set of constant conditions, with a series of records of well-known specimens analyzed under the same conditions as reference standards. Nevertheless, the striking differences in d. t. a. heating products and the complicated reactions during the d. t. a. treatment suggest that there may be some significant differences in the equilibrium heating assemblages, depending on the F/OH ratio of the original mineral, and the products can be identified with certainty. The phase changes during the differential thermal process and the equilibrated heating products deserve further study.

II B - 5. X-ray diffraction study

II B - 5a. Methods and results

Powdered amblygonite - montebrasite specimens were X-rayed on a Philips Norelco diffractometer. The rotating sample holder was used for several specimens but later all samples were run on glass slide mounts, since the amount of material available was not sufficient for filling the rotating holder, and preference was given to the method which could be applied uniformly to all specimens examined. X-raying with the rotating sample holder would probably have yielded better intensities and shapes of peaks, but, as shown below, the accuracy of the results obtained with glass slide mounts is quite satisfactory. In all cases, the X-rayed material was taken out of the sample prepared for the chemical analysis.

Ni-filtred Cu-radiation was used, with a scanning speed of $\frac{1}{4}^\circ$ per minute and a chart driving setting of 10, yielding approximately 1 inch per $\frac{1}{2}^\circ 2\theta$, and with 400 counts per second, time constant 4 sec., zero suppression 0. After X-raying the pure sample, the same powder was re-run three-times (remounting the powder for each run) with an approx. $\frac{1}{4}$ admixture of pegmatitic quartz from Bernic Lake as internal standard. All the peaks were measured as close to the top as reasonable, and 2θ values were then corrected using the theoretical $\text{CuK}_{\alpha 1}$ -values for quartz calculated in Frondel (1963). The amblygonite patterns averaged from the three calibrated runs have been indexed according to the powder data published by Haapala (1966) and Moss et al. (1969).

In all patterns, at least 25 sharp and uniquely indexed

reflections could be used for refinement of the unit cell dimensions; in the majority of cases around 30 reflections were suitable, and in a few cases up to 37. The unit cell dimensions have been calculated from the 2θ values of these reflections using the Fortran IV least squares refinement program by Evans et al. (1963), modified later by D. E. Appleman and D. B. Stewart, in the computing centre of the University of Manitoba.

Table 6 presents the data for three complete X-ray powder diffractometer patterns of a fluorine-rich, an intermediate, and a fluorine-poor specimen. Table 7A and 7B contain the unit cell dimensions and their standard errors of all X-rayed specimens for direct and reciprocal cell respectively.

The computer-calculated standard errors are mostly very small and even the doubled values, taken as more reasonable for feldspars by Wright and Stewart (1968), would be still smaller than the precision in other studies (Haapala, 1966). An additional check proved that this high accuracy is true: refinement of single patterns taken out of three averaged records gave unit cell dimensions only rarely falling outside of the standard errors of the averaged patterns, and furthermore the same relation was found for unit cell dimensions refined for other portions of the sample prepared for chemical analysis. Thus the caution concerning computer-calculated standard errors, expressed by Moss et al. (1969) does not seem to apply to amblygonite - montebrasites under the present experimental conditions. The large number of uniquely indexed and well shaped peaks obtainable from amblygonite - montebrasite likely accounts for this high accuracy of the refined unit cell dimensions.

TABLE 6

X-RAY POWDER DIFFRACTION DATA FOR AMBLYGONITE, INTERMEDIATE AMBLYGONITE - MONTEBRASITE, AND MONTEBRASITE

$2\theta^\circ$ given for $\text{CuK } \alpha_1$, corrected against quartz internal standard using the data of C. Frondel (1963).
 Intensities recalculated to $I = 100$ for the strongest reflections after deducting background.

Specimen AF - 55 (10.17% F)					Specimen A - 3 (6.30% F)					Specimen A - 22 (1.40% F)				
$a_o = 5.1510 \overset{\circ}{\text{\AA}}$					$a_o = 5.1668 \overset{\circ}{\text{\AA}}$					$a_o = 5.1896 \overset{\circ}{\text{\AA}}$				
$b_o = 7.2062 \overset{\circ}{\text{\AA}}$					$b_o = 7.1807 \overset{\circ}{\text{\AA}}$					$b_o = 7.1685 \overset{\circ}{\text{\AA}}$				
$c_o = 5.0596 \overset{\circ}{\text{\AA}}$					$c_o = 5.0482 \overset{\circ}{\text{\AA}}$					$c_o = 5.0407 \overset{\circ}{\text{\AA}}$				
$\alpha = 113^\circ 53.6'$					$\alpha = 113^\circ 4.7'$					$\alpha = 112^\circ 27.1'$				
$\beta = 98^\circ 34.9'$					$\beta = 98^\circ 8.8'$					$\beta = 97^\circ 55.9'$				
$\gamma = 67^\circ 15.3'$					$\gamma = 67^\circ 37.3'$					$\gamma = 67^\circ 48.5'$				
$V = 158.354 \overset{\circ}{\text{\AA}}^3$					$V = 159.322 \overset{\circ}{\text{\AA}}^3$					$V = 160.464 \overset{\circ}{\text{\AA}}^3$				

h	k	l	I	d meas.	2 θ meas.	2 θ calc.	h	k	l	I	d meas.	2 θ meas.	2 θ calc.	h	k	l	I	d meas.	2 θ meas.	2 θ calc.
0	1	0				14.400	0	1	0				14.342	0	1	0				14.288
1	0	0	19	4.744	18.690	18.664	1	0	0	16	4.774	18.570	18.556	1	0	0	12	4.791	18.500	18.449
1	1	0	68	4.657	19.040	19.039	1	1	0	57	4.662	19.020	18.987	1	1	0	58	4.671	18.980	18.917

Table 6 - continued

Specimen AF - 55					Specimen A - 3					Specimen A - 22				
h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.
0-11	73	4.640	19.110	19.106	001	75	4.640	19.110	19.094	001	53	4.659	19.030	19.035
001				19.176	0-11				19.196	0-11	b21	4.578	19.370	19.271
-1-11	15	3.867	22.980	22.967	-1-11	13	3.855	23.050	23.043	-1-11	8	3.858	23.030	23.057
101				26.769	101				26.627	101				26.530
-101	30	3.301	26.990	26.997	-101	59	3.313	26.890	26.868	-101	30	3.335	26.710	26.727
-1-21	42	3.246	27.450	27.427	-110	52	3.255	27.380	27.364	-110	35	3.273	27.220	27.207
-110	34	3.239	27.515	27.539	-1-21	38	3.227	27.620	27.619	-1-21				27.723
0-21	71	3.176	28.070	28.056	011	53	3.187	27.970	27.965	011	72	3.213	27.740	27.772
011	73	3.161	28.210	28.203	0-21	56	3.163	28.190	28.177	120	94	3.163	28.190	28.176
120	71	3.152	28.290	28.316	120	59	3.156	28.250	28.256	0-21				28.267
020	3	3.071	29.050	29.034	020	b5	3.082	28.950	28.914	020				28.805
1-11	100	2.954	30.230	30.239	1-11	100	2.961	30.160	30.161	1-11	100	2.966	30.100	30.113
111	23	2.900	30.800	30.808	111	b8	2.920	30.590	30.578	111	7	2.938	30.400	30.401
210	11	2.545	35.240	35.231	210	18	2.557	35.070	35.077	210	14	2.569	34.900	34.903
0-12	24	2.500	35.885	35.877	0-12	23	2.499	35.900	35.911	0-12	22	2.496	35.950	35.952
-111	8	2.426	37.015	37.014	-111	10	2.446	36.710	36.721	-111				36.454
-1-31				37.661	200	36	2.390	37.600	37.622	200				37.398
-2-11	24	2.384	37.700	37.686	-2-11				37.622	-2-11	32	2.397	37.490	37.488

Table 6 - continued

Specimen AF - 55					Specimen A - 3					Specimen A - 22				
h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.
2 0 0	13	2.376	37.840	37.847	-1-3 1	7	2.376	37.830	37.856	-1-3 1	7	2.368	37.970	37.961
-2-2 1				38.044	-2-2 1	3	2.359	38.120	38.128	-2-2 1				38.105
-1-1 2				38.261	-1-1 2				38.298	-1-1 2				38.290
-1-2 2	b 4	2.342	38.400	38.382	2 2 0	10	2.335	38.530	38.521	2 2 0	8	2.343	38.380	38.377
2 2 0	5	2.327	38.660	38.631	-1-2 2	8	2.329	38.620	38.628	0 0 2				38.621
0-2 2	7	2.320	38.783	38.770	0 0 2				38.746	-1-2 2				38.780
0 0 2				38.917	0-2 2	6	2.310	38.960	38.958	0-2 2	9	2.300	39.130	39.116
1-2 1				39.448	1-2 1	12	2.285	39.400	39.379	1-2 1	13	2.288	39.350	39.330
-1 2 0				40.258	-1 2 0				40.015	1 2 1				39.786
1 2 1	b 4	2.235	40.320	40.348	1 2 1	4	2.249	40.050	40.039	-1 2 0	6	2.265	39.770	39.803
0-3 1				40.780	0 2 1	2	2.219	40.620	40.610	0 2 1	b 3	2.232	40.380	40.321
0 2 1	b 3	2.203	40.940	40.956	0-3 1				40.865	0-3 1				40.917
1 3 0	5	2.189	41.205	41.187	1 3 0	7	2.194	41.110	41.082	1 3 0	10	2.203	40.940	40.962
2 0 1	5	2.128	42.440	42.611	2 0 1				42.360	2 0 1				42.158
-2 0 1	17	2.105	42.920	42.915	-2 0 1	17	2.116	42.690	42.680	-2 0 1	21	2.129	42.420	42.421
2 1 1				43.002	2 1 1				42.721	2 1 1	9	2.124	42.520	42.489
1-1 2				43.098	1-1 2				43.002	1 0 2				42.993

Table 6 - continued

Specimen AF - 55					Specimen A - 3					Specimen A - 22				
h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.
1 0 2	6	2.085	43.355	43.342	1 0 2	6	2.095	43.150	43.128	1-1 2	11	2.102	42.990	43.000
-1 0 2				43.641	-1 0 2				43.444	-1 0 2				43.252
-2-3 1				43.873	0 3 0				43.984	0 3 0				43.813
-1-3 2	7	2.058	43.960	43.967	-2-3 1	3	2.054	44.050	44.038	-2-3 1	5	2.053	44.070	44.076
0 3 0				44.172	-1-3 2	5	2.042	44.330	44.332	-1-3 2 b 4		2.031	44.570	44.573
-2 1 0	3	1.987	45.610	45.602	-2 1 0	3	2.001	45.280	45.299	-2 1 0				45.019
0-3 2				46.668	0 1 2	20	1.949	46.560	46.560	0 1 2	18	1.960	46.270	46.275
0 1 2				46.921	2 3 0	7	1.938	46.840	46.839	2 3 0	6	1.943	46.700	46.693
-2-2 2	25	1.934	46.927	46.927	0-3 2				46.925	2-1 1	6	1.936	46.880	46.896
2 3 0				46.945	2-1 1	16	1.929	47.070	47.089	-2-2 2	8	1.929	47.070	47.120
2-1 1	7	1.918	47.343	47.331	-2-2 2				47.090	0-3 2				47.128
1-2 2	21	1.898	47.897	47.911	1-2 2	13	1.896	47.940	47.931	2 2 1				47.805
2 2 1	3	1.880	48.370	48.405	2 2 1				48.081	1 1 2				47.959
1 1 2				48.585	1 1 2				48.222	1-2 2	25	1.894	47.990	47.980
-2-1 2				49.085	-2-1 2				49.028	-2-1 2				48.892
-2-3 2	3	1.837	49.585	49.603	-1 2 1				49.553	-1 2 1				49.183
-1 2 1				49.975	-2-3 2	3	1.827	49.940	49.933	-2-3 2				50.090
-1-4 1	4	1.798	50.743	50.740	-1-4 1	12	1.792	50.900	50.895	-1-4 1	10	1.790	50.960	50.965

Table 6 - continued

Specimen AF - 55					Specimen A - 3					Specimen A - 22				
h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.
1-3 1	1	1.762	51.840	51.856	1-3 1				51.758	-2 1 1				51.605
-2 1 1				52.372	-2 1 1				51.982	1-3 1				51.676
1 3 1	8	1.728	52.943	52.949	1 3 1	10	1.740	52.560	52.558	1 3 1				52.229
-1 1 2				53.179	-1 1 2				52.773	-1 1 2	19	1.743	52.460	52.414
-1-4 2				53.647	-2-4 1	3	1.698	53.940	53.927	3 1 0				53.738
-2-4 1				53.745	3 1 0				54.042	-2-4 1	3	1.697	53.990	53.978
3 1 0				54.324	-1-4 2				54.051	-3-2 1				54.142
-3-2 1				54.387	-3-2 1				54.321	-1 3 0				54.178
3 2 0				54.770	-1 3 0				54.457	3 2 0				54.295
-1 3 0	3	1.674	54.800	54.777	3 2 0	3	1.682	54.520	54.554	-1-4 2	2	1.687	54.320	54.317
2 0 2				55.159	2 0 2				54.847	2 0 2	6	1.678	54.650	54.633
-1-2 3				55.306	0 3 1	6	1.667	55.050	55.057	0 3 1				54.677
0-4 1				55.311	0-4 1				55.341	-2 0 2	14	1.666	55.070	55.066
0 3 1	7	1.654	55.510	55.507	-2 0 2	21	1.658	55.360	55.375	0-4 1				55.340
0-2 3				55.537	-1-2 3	11	1.654	55.520	55.505	1 4 0				55.464
0-1 3				55.621	0-1 3				55.569	0-1 3				55.549
-2 0 2	8	1.650	55.653	55.659	1 4 0				55.641	-1-2 3				55.629

Table 6 - continued

Specimen AF - 55					Specimen A - 3					Specimen A - 22				
h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.
1 4 0				55.809	0-2 3	4	1.649	55.690	55.690	-3-1 1				55.663
-3-1 1				56.162	2-2 1	6	1.643	55.910	55.912	2-2 1	7	1.649	55.700	55.703
2-2 1	4	1.636	56.180	56.170	-3-1 1				55.950	0-2 3				55.831
-2-4 2	5	1.625	56.600	56.605	-2 2 0	3	1.628	56.480	56.466	-2 2 0	4	1.638	56.110	56.120
1-3 2	7	1.619	56.810	56.802	2-1 2				56.751	2-1 2				56.605
-2 2 0				56.853	1-3 2	6	1.617	56.880	56.864	1 2 2				56.905
-3-3 1				56.979	-3-3 1	20	1.614	57.010	57.027	-3-3 1				56.933
2-1 2	11	1.615	56.985	56.980	-2-4 2				57.030	1-3 2				56.936
-1-1 3	5	1.604	57.407	57.405	2 1 2				57.254	2 1 2	17	1.616	56.940	56.952
-1-3 3	3	1.600	57.540	57.529	1 2 2				57.294	2 3 1	8	1.613	57.060	57.041
2 1 2				57.666	-1-1 3				57.361	-2-4 2	11	1.608	57.240	57.260
2 3 1				57.754	2 3 1	5	1.601	57.500	57.373	-1-1 3	14	1.606	57.300	57.296
1 2 2				57.797	0 2 2				57.796	0 2 2				57.368
0-4 2	9	1.589	57.997	57.998	3 0 0	14	1.592	57.850	57.851	3 0 0	24	1.602	57.480	57.488
3 0 0	16	1.583	58.223	58.216	-1-3 3				57.926	-1-3 3				58.201
0 2 2				58.324	0-4 2	10	1.582	58.270	58.266	2 4 0	12	1.583	58.250	58.265
2 4 0	6	1.574	58.580	58.574	2 4 0	10	1.578	58.430	58.440	0-4 2	15	1.577	58.460	58.467

Table 6 - continued

Specimen AF - 55					Specimen A - 3					Specimen A - 22				
h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.
3 3 0				59.491	3 3 0	3	1.557	59.310	59.313	3 3 0	3	1.563	59.070	59.077
0-3 3	8	1.547	59.730	59.718	0 0 3				59.680	3 1 1				59.367
0 0 3				59.958	3 1 1	2	1.547	59.700	59.707	0 0 3				59.476
3 1 1	5	1.538	60.100	60.093	0 4 0				59.908	0 4 0				59.664
0 4 0				60.176	0-3 3				60.026	0-3 3	9	1.534	60.290	60.284
1-1 3	5	1.519	60.940	60.956	1-1 3	6	1.522	60.810	60.822	1-1 3	9	1.523	60.760	60.770
-3-2 2				61.524	3 0 1				61.319	3 0 1	5	1.519	60.960	60.980
3 0 1				61.710	-3-2 2				61.542	-3 0 1				61.283
-3-3 2				61.884	-3 0 1				61.688	-3-3 2				61.407
-3 0 1				62.061	-3-3 2				62.089	3 2 1				61.830
-2-2 3				62.138	3 2 1				62.187	-3-3 2				62.095
-2 3 3				62.334	-2-2 3				62.288	-2-2 3				62.310
3 2 1	3	1.483	62.600	62.586	2-2 2	13	1.480	62.710	62.712	2-2 2	12	1.482	62.610	62.604
1-2 3				62.781	-2-3 3				62.713	1 0 3				62.689
2-2 2	8	1.477	62.883	62.889	1-2 3				62.804	1-2 3 b 6		1.478	62.820	62.875
1 0 3	4	1.470	63.190	63.209	1 0 3				62.891	-2-3 3				62.911

Table 6 - continued

Specimen AF - 55					Specimen A - 3					Specimen A - 22				
h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.	h k l	I	d meas.	2 θ meas.	2 θ calc.
-1 0 3				63.555	-1 0 3				63.256	-1 0 3				62.987
-3-4 1				63.593	2 2 2	8	1.460	63.660	63.656	2 2 2	7	1.469	63.240	63.256
-1-4 3				63.787	-3-4 1				63.710	-3-4 1				63.666
2 2 2				64.179	-2 2 1				64.256	-2 2 1				63.764
-2 2 1				64.795	-1 3 1	2	1.447	64.310	64.304	-1 3 1	3	1.457	63.820	63.826
-1 3 1				64.854	-1-4 3				64.319	-1 2 2				64.553
-3-1 2				65.197	-3-1 2				65.000	-1-4 3				64.694
-1 2 2	5	1.421	65.660	65.661	-1 2 2				65.065	-3-1 2 b 6		1.441	64.610	64.704
-1-5 1				65.719	-2 1 2	3	1.428	65.290	65.267	-2 1 2 b 6		1.439	64.720	64.791
-2 1 2				65.766	-3 1 0				65.498	-3 1 0				65.064
-2-1 3	7	1.415	65.935	65.950	-1-5 1				65.821	-2-1 3	7	1.420	65.670	65.662
-3 1 0				65.958	-2-1 3	4	1.418	65.820	65.840	-1-5 1				65.840

TABLE 7A

DIRECT UNIT CELL DIMENSIONS OF AMBLYGONITE - MONTEBRASITE DERIVED FROM X-RAY POWDER DIFFRACTION DATA

Specimen	F% wt.	a_o (Å)	b_o (Å)	c_o (Å)	α	β	γ	V (Å ³)
AF - 65	11.8	5.1483 17*	7.2071 20	5.0601 12	114° 0.3' 1.3	98°38.3' 1.6	67°14.5' 1.6	158.155 .054
AF - 55	10.17	5.1510 10	7.2062 12	5.0596 8	113°53.6' 0.7	98°34.9' 0.8	67°15.3' 0.9	158.354 .032
AF - 1	7.85	5.1606 11	7.1879 13	5.0499 17	113°23.7' 1.3	98°17.3' 1.7	67°32.0' 1.4	158.861 .048
A - 13	6.43	5.1705 15	7.1799 14	5.0509 11	113° 4.9' 1.0	98° 9.6' 1.4	67°36.3' 1.2	159.480 .046
A - 3	6.30	5.1668 11	7.1807 9	5.0482 7	113° 4.7' 0.6	98° 8.8' 0.9	67°37.3' 0.9	159.322 .038
A - 29	6.30	5.1691 12	7.1782 11	5.0513 9	113° 5.8' 1.1	98°10.7' 1.3	67°35.4' 1.0	159.379 .042
A - 60	6.17	5.1694 19	7.1815 12	5.0507 10	113° 9.1 0.8	98°11.2' 1.5	67°32.6' 1.2	159.321 .052
A - 1	5.56	5.1681 10	7.1816 10	5.0501 9	113° 5.6' 0.7	98°11.2' 0.9	67°35.6' 0.7	159.392 .031

Table 7A - continued

Specimen	F% wt.	a_o (Å)	b_o (Å)	c_o (Å)	α	β	γ	$V(\text{Å}^3)$
A - 5	4.46	5.1792 9	7.1736 14	5.0451 10	112°45.5' 0.9	98° 0.5' 1.0	67°43.6' 1.0	159.944 .039
A - 4	4.05	5.1783 7	7.1770 12	5.0468 9	112°49.3' 0.7	98° 4.8' 0.9	67°42.5' 0.7	159.952 .031
A - 2	3.65	5.1798 8	7.1685 13	5.0438 7	112°41.9' 1.0	98° 0.4' 0.9	67°45.1' 0.7	159.908 .030
A - 98	3.44	5.1801 7	7.1755 11	5.0448 6	112°43.7' 0.7	98° 0.1' 0.7	67°43.8' 0.6	160.044 .027
A - 72	3.17	5.1888 9	7.1715 13	5.0424 7	112°28.4' 0.7	97°56.0' 0.8	67°49.2' 0.9	160.548 .031
AF - 43	1.88	5.1924 9	7.1689 10	5.0422 8	112°26.7' 0.7	97°57.3' 1.0	67°47.3' 0.7	160.595 .032
A - 22	1.40	5.1896 8	7.1685 10	5.0407 7	112°27.1' 0.7	97°55.9' 0.7	67°48.5' 1.0	160.464 .029
AF - 47	1.5**	5.1957 9	7.1681 10	5.0397 10	112°19.4' 0.9	97°53.3' 1.2	67°48.6' 0.9	160.763 .033
AF - 48	1.4**	5.1946 10	7.1712 14	5.0430 12	112°21.5' 1.0	97°54.8' 1.3	67°51.6' 1.0	160.926 .039

* - standard error

** - estimated from X-ray powder data

TABLE 7B

RECIPROCAL UNIT CELL DIMENSIONS OF AMBLYGONITE - MONTEBRASITE DERIVED FROM X-RAY POWDER DIFFRACTION DATA

Specimen	F% wt.	a_o^* (Å)	b_o^* (Å)	c_o^* (Å)	α^*	β^*	γ^*
AF - 65	11.8	0.2106	0.1628	0.2163	67°30.6' 1.2 *	90°29.3' 1.5	111° 8.5' 1.5
AF - 55	10.17	0.2105	0.1627	0.2162	67°36.6' 0.7	90°30.0' 0.9	111° 8.9' 1.0
AF - 1	7.85	0.2097	0.1623	0.2158	68° 2.3' 1.1	90°30.7' 1.7	110°57.7' 1.5
A - 13	6.43	0.2092	0.1621	0.2152	68°19.7' 0.9	90°30.0' 1.3	110°55.9' 1.0
A - 3	6.30	0.2093	0.1620	0.2153	68°19.7' 0.7	90°30.4' 1.0	110°55.2' 0.9
A - 29	6.30	0.2093	0.1622	0.2152	68°19.2' 1.0	90°29.4' 1.3	110°56.4' 1.1
A - 60	6.17	0.2093	0.1622	0.2153	68°15.7' 0.9	90°31.5' 1.6	110°59.4' 1.3
A - 1	5.56	0.2093	0.1621	0.2153	68°19.5' 0.6	90°28.8' 0.9	110°56.0' 0.9

Table 7B - continued

Specimen	F% wt.	a_o^* (Å)	b_o^* (Å)	c_o^* (Å)	α^*	β^*	γ^*
A - 5	4.46	0.2087	0.1618	0.2150	68°37.2' 0.9	90°29.5' 1.6	110°51.5' 1.1
A - 4	4.05	0.2087	0.1618	0.2150	68°34.7' 0.8	90°26.5' 1.0	110°51.0' 0.8
A - 2	3.65	0.2086	0.1618	0.2149	68°41.0' 0.6	90°27.5' 0.8	110°49.8' 0.8
A - 98	3.44	0.2086	0.1617	0.2149	68°39.1' 0.7	90°29.0' 0.7	110°51.3' 0.7
A - 72	3.17	0.2081	0.1614	0.2146	68°54.0' 0.6	90°25.2' 0.6	110°47.0' 0.9
AF - 43	1.88	0.2080	0.1615	0.2146	68°56.3' 0.7	90°23.8' 1.0	110°48.5' 0.7
A - 22	1.40	0.2081	0.1615	0.2147	68°55.4' 0.8	90°25.0' 0.7	110°47.7' 1.0
AF - 47	1.5**	0.2079	0.1613	0.2145	69° 2.7' 0.7	90°25.0' 0.9	110°48.7' 0.8
AF - 48	1.4**	0.2078	0.1612	0.2144	69° 1.1' 0.8	90°22.8' 1.1	110°44.8' 0.9

* - standard error

** - estimated from X-ray powder data

Single-crystal X-ray diffraction photographs of cleavage fragments of four specimens have been taken on the O. von der Heyde precession instrument, using Zr-filtred Mo-radiation from a Philips X-ray generator. Two specimens (AF-55 and A-22) were mounted with the (100) plane normal to the dial axis and thus with x^* as rotation axis, and precession photographs taken around the y and z axes. One other specimen (AF-47) was mounted with the (001) plane normal to the dial axis, making z^* the rotation axis, and with precession photographs taken around the x and y axes. A fourth sample, A-3, was mounted in both positions; prints of the three principal 0-level precession photographs of this specimen are shown in Figs. 6, 7, and 8.

Zero-level and 1-level photographs were taken to determine the unit cell dimensions, and to check for a possible supercell which could escape unnoticed in X-ray powder patterns. Unit cell dimensions were obtained from the h00, 0k0 and 00l rows of 0-level photographs, using 3 to 7 orders, and from the directly measured angles. No superstructure and/or substructure was found; all specimens examined have the classic unit cell determined by Palache et al. (1943) despite the widely varying chemical composition and mode of origin: AF-55 F-rich, A-22 OH-rich, A-3 intermediate ($F/OH \approx 1$), AF-47 OH-rich from pegmatite cavities. Table 8 contains the reciprocal cell dimensions found by the single-crystal X-ray work compared to the values found by the refinement of the powder data.

II B - 5b. Unit cell dimensions refined from powder data versus F-contents

The unit cell dimensions quoted in Table 7A are plotted

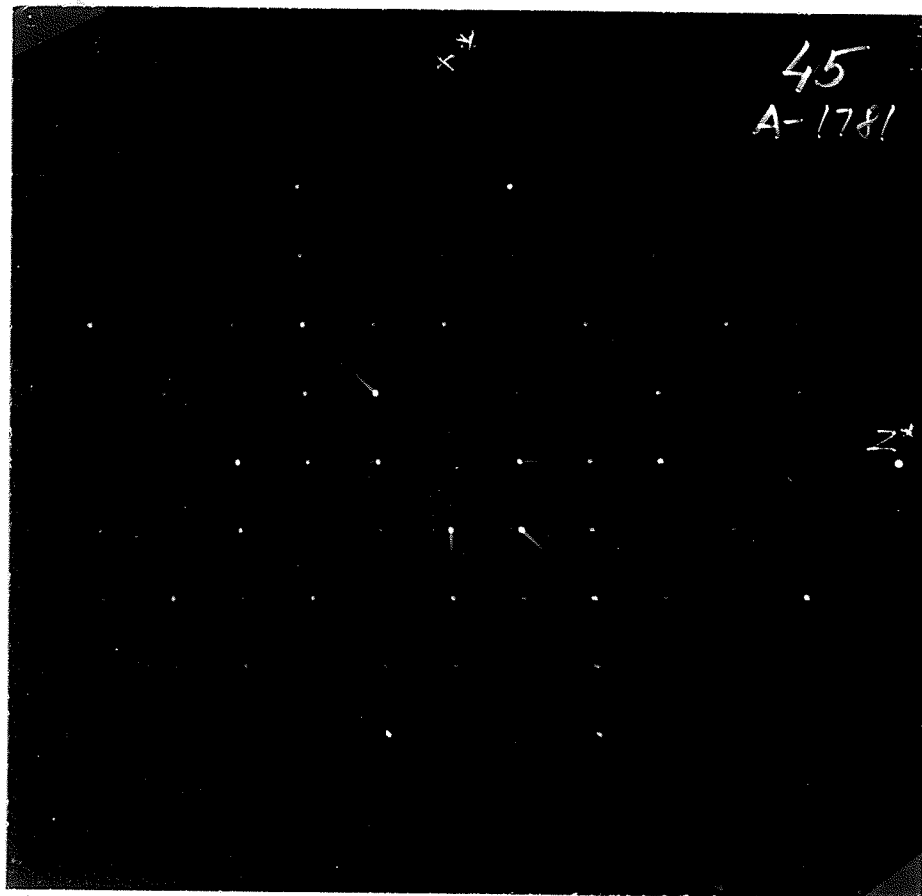


Fig. 6. Zero level precession photograph of sample A-3; rotation axis z^* , precession around y , showing the $z^* - x^*$ plane.



Fig.7. Zero level precession photograph of sample A-3; rotation axis z^* , precession around x , showing the plane $z^* - y^*$.



Fig.8. Zero level precession photograph of sample A-3; rotation axis x^* , precession around z , showing the plane $x^* - y^*$.

against the F-contents of the respective specimens in Figs. 9 and 10; the four specimens examined by Moss et al. (1969) are included, as well as two low-temperature, cavity filling crystals from Newry, Maine, plotted against their F-contents estimated by the X-ray method described later in section II B - 5c.

The present data plot very well along flat arcs, with the exception of the c_o plot which is probably truly linear. The data of Moss et al. (1969) show mostly a broader spread. Since the F-rich half of the F% range is represented by only 3 samples no attempt was made to calculate statistically based curves. The estimated curves are regarded as satisfactory until more data on F-rich amblygonites become available and a reasonable calculation can be performed.

Even the best fitting plots (as those of c_o and all the angles) show a spread around the curves which exceeds several times the predominant standard error of the cell dimensions. These deviations are probably caused by differences between the samples X-rayed and those chemically analyzed as discussed in Chapter II A and Table 2; another cause might be slight errors in the fluorine determinations.

It should be noted here that the F-poor, low-temperature montebrasites from pegmatite cavities at Newry, Maine and the specimen A-22 which is an alteration product of a F-rich montebrasite, plot as close to the averaged curves as do the other amblygonites- montebrasites that are primary higher temperature phases. This is discussed further in section II B - 5e.

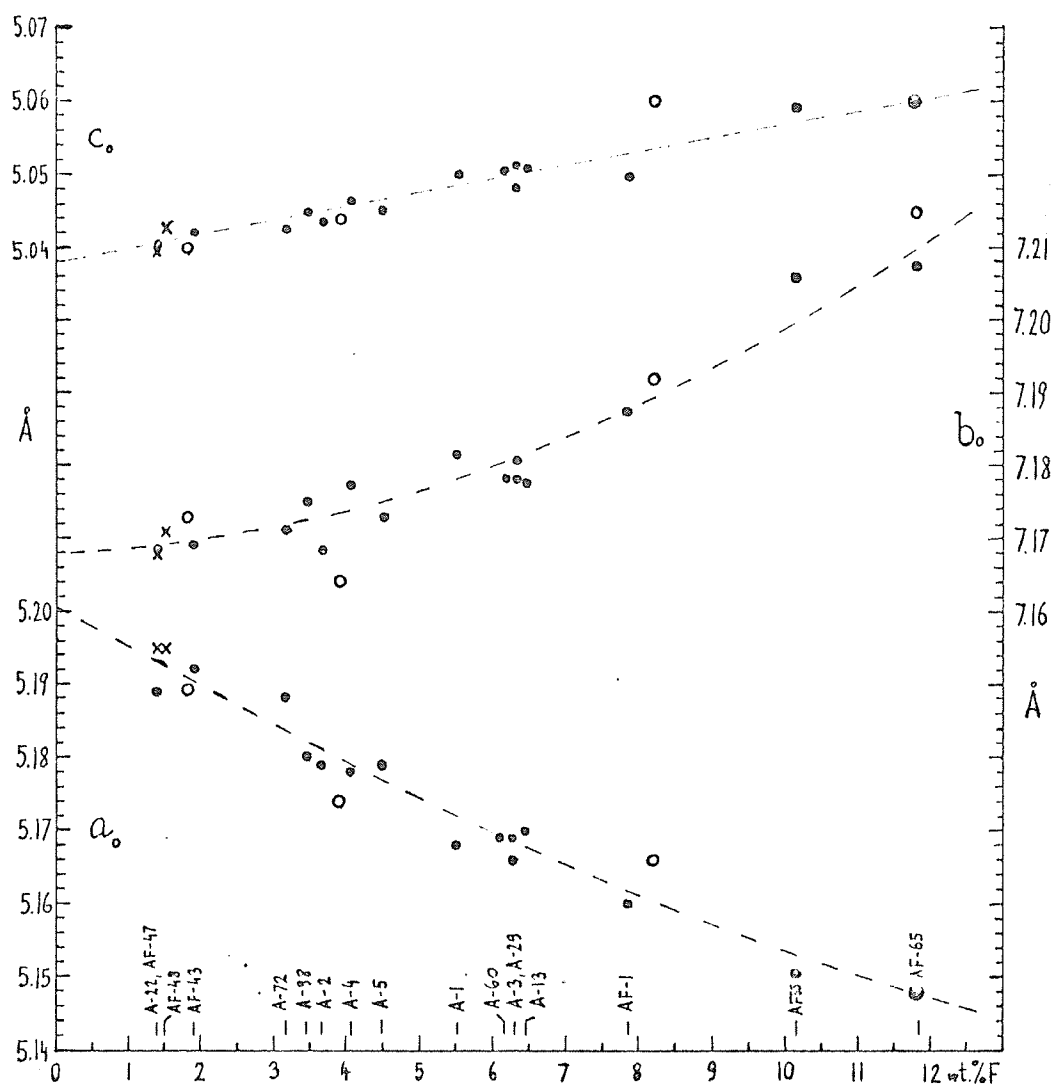


Fig.9. Variation of a_0 , b_0 , and c_0 of amblygonite-montebrazite with its fluorine content (in wt.% F). Dots - plots of the present data, mostly exceeding the standard errors in size; empty circles - data from Moss et al.(1969); large dots - overlap of the data by Moss et al. with present values for the same specimen; crosses - plots based on fluorine contents estimated from physical properties; dashed lines - estimated best correlations.

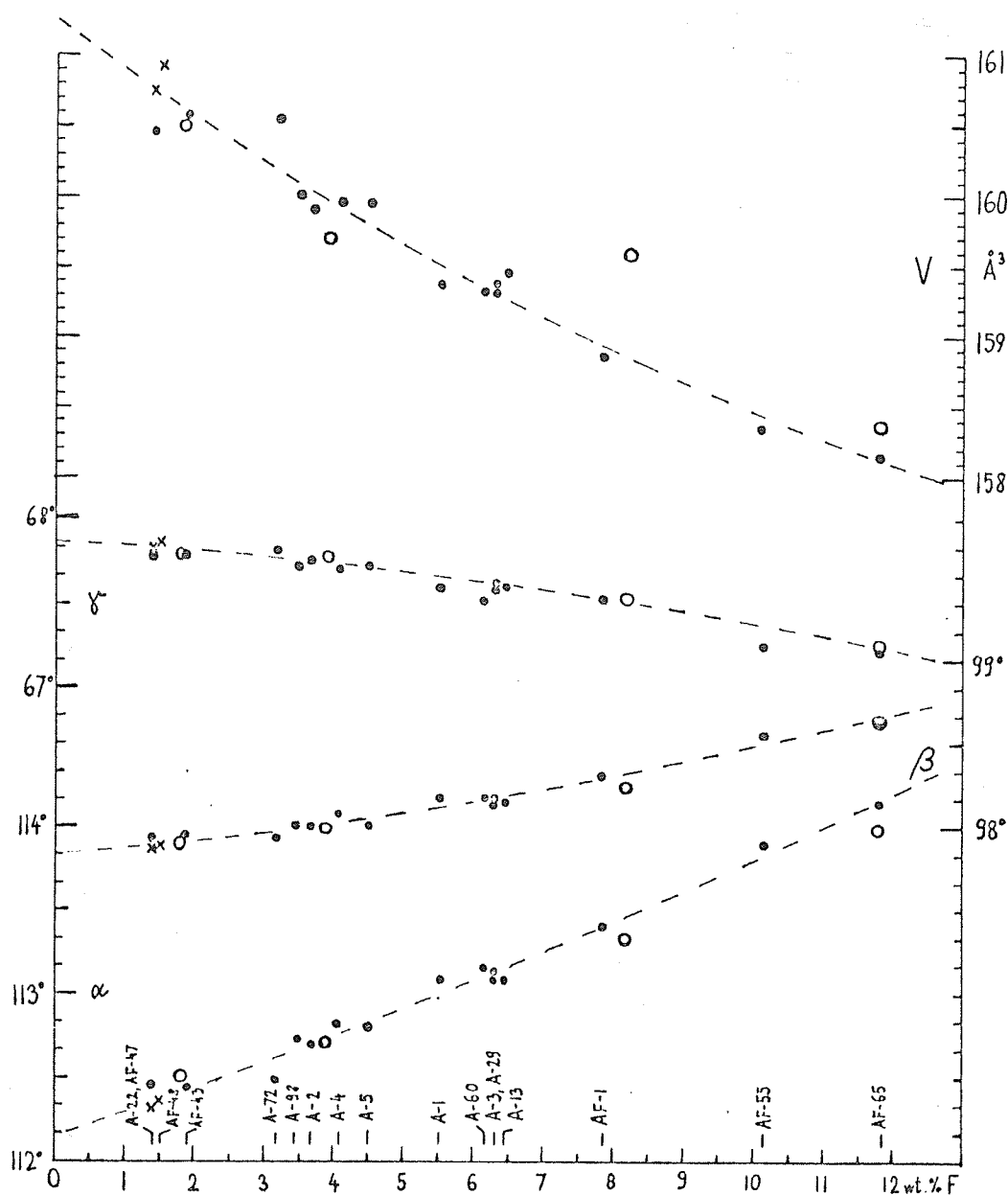


Fig.10. Variation of α , β , γ , and V of amblygonite-montebrazite with its fluorine content (in wt.% F). Dots - plots of the present data, mostly exceeding the standard errors in size; empty circles - data from Moss et al.(1969); large dots - overlap of the data by Moss et al. with present values for the same specimen; crosses - plots based on fluorine contents estimated from physical properties; dashed lines - estimated best correlations.

II B - 5c. Estimates of the F-contents from the X-ray powder data

The plots of cell dimensions vs. F-contents illustrated in Figs. 9 and 10 indicate that reasonable estimates of the F-contents can be obtained from those graphs when accurate unit cell data are available, obtained by the same method as those on which the graphs are based. The accuracy of such estimates would be approximately $\pm 0.5\%$ F. However, even with computerized calculations the refinement of unit cell data is still time-consuming, and may become quite lengthy if large numbers of specimens are to be checked.

Moss et al. (1969) proposed that the spacing of reflection (131) be used for the indirect determination of the F-contents. The authors note, however, that a one-line graph is influenced by the zero error of the diffractometer, and thus calibration of the pattern is necessary and despite their use of an internal standard and the high correlation coefficient, many of their plots deviate by 1 - 1.5% F from the line calculated from them.

In the present study, a determinative graph has been constructed which is based on six reflections -1 0 1, -1 1 0, -1-2 1, 0 1 1, 0-2 1, and 1 2 0 that show the most prominent shifts with changes of the F-content. They have been illustrated by Moss et al. (1969, Fig.2) and used by those authors for an arbitrary division of the amblygonite - montebrasite series into five groups with characteristic patterns in the range $26 - 29^\circ 2\theta \text{ CuK}\alpha$.

In this investigation all the analyzed samples have been run several times through the range $26 - 29^\circ 2\theta \text{ CuK}\alpha$, from lower to higher

2θ angles, at 400 c.p.s., time constant 4, goniometer speed $\frac{1}{4}^\circ/\text{min.}$, chart driving speed set at 10. To avoid the necessity of calibrating the diffractometer patterns, the mutual distances of these reflections were plotted from an origin taken to be the position of the $-1\ 0\ 1$ peak. After plotting the measured values, averaged lines were drawn for each reflection. Then the theoretical 2θ angles calculated from the refined unit cell dimensions were plotted, too, using the $-1\ 0\ 1$ reflection as an origin again. With few exceptions, these plots coincided very well with the curves averaged from the measured data. The theoretical points confirmed also the course of the individual curves in those regions where the paths of two lines cross and only one peak is observable.

The plotted data with a light tracing of the computed curves are demonstrated in Fig. 11. and a working graph with areas of probable coincidence of two closely spaced peaks indicated as stippled areas is shown in Fig. 12. The frequent overlapping of peaks is the main disadvantage of this particular group of peaks in using them for estimating the F-percentages. However, this is not too serious when proper care is taken with X-raying, and particularly when a large number of specimens is studied. In good diffractometer records, peaks $0.1^\circ\ 2\theta$ or more apart should be resolved, and with some practice the correct evaluation of intensity, width and shape of composite peaks is rather easy.

Generally, the estimates from this chart should be within approximately $\pm 0.5\%$ F, with the exception of the F-rich end of the series where the basic data for the curves are scarce, and of the OH-rich part where the overlap in two pairs of reflections and the resulting

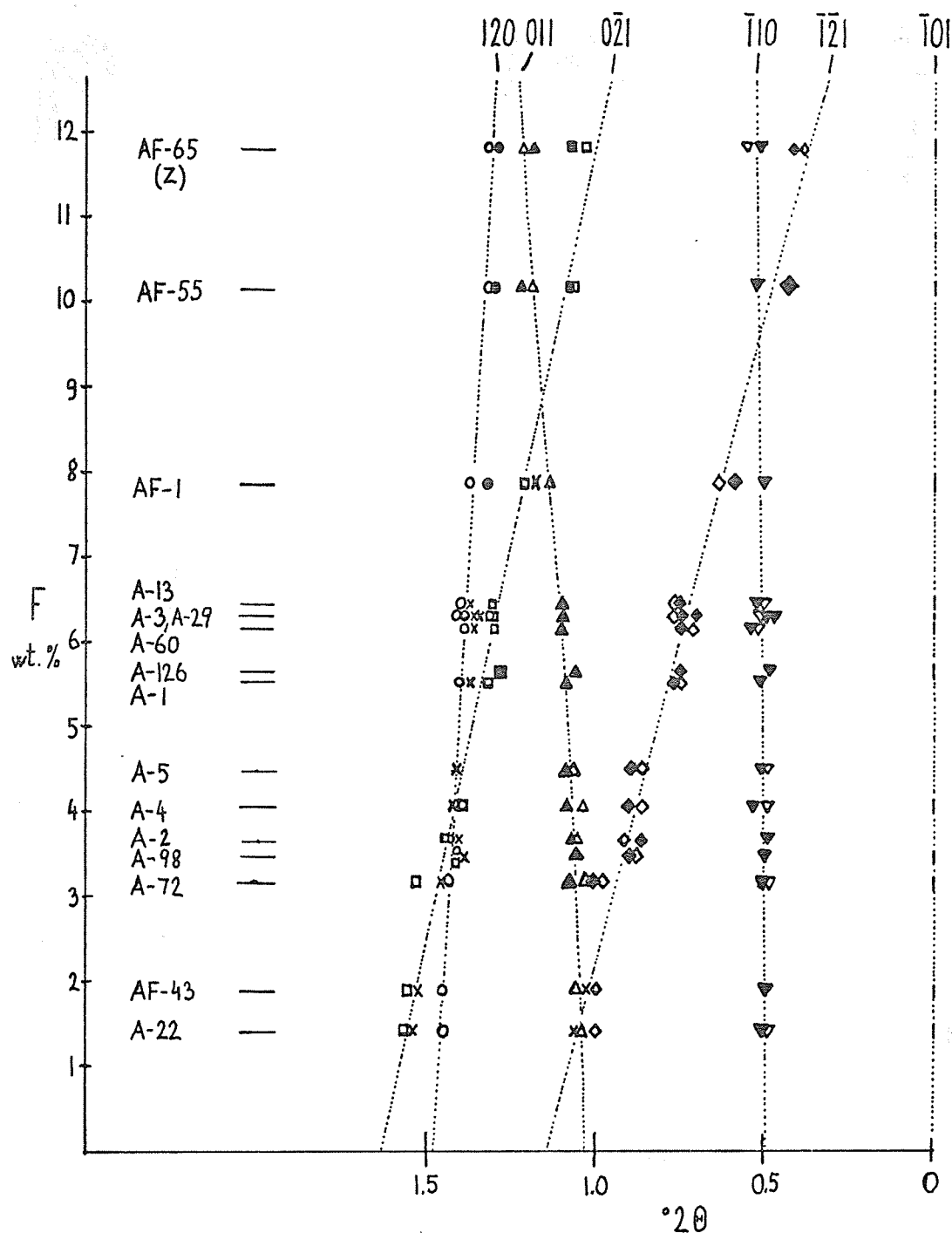


Fig.11. Plots of 2θ ($\text{CuK}\alpha$) values of six X-ray powder reflections of amblygonite-montebbrasite against its fluorine content (in wt.% F), with the value of 101 taken as zero. Full symbols - measured values; empty symbols - theoretical values calculated from unit cell dimensions refined from powder data; crosses - measured values for overlapping reflections; dotted lines - estimated best correlations.

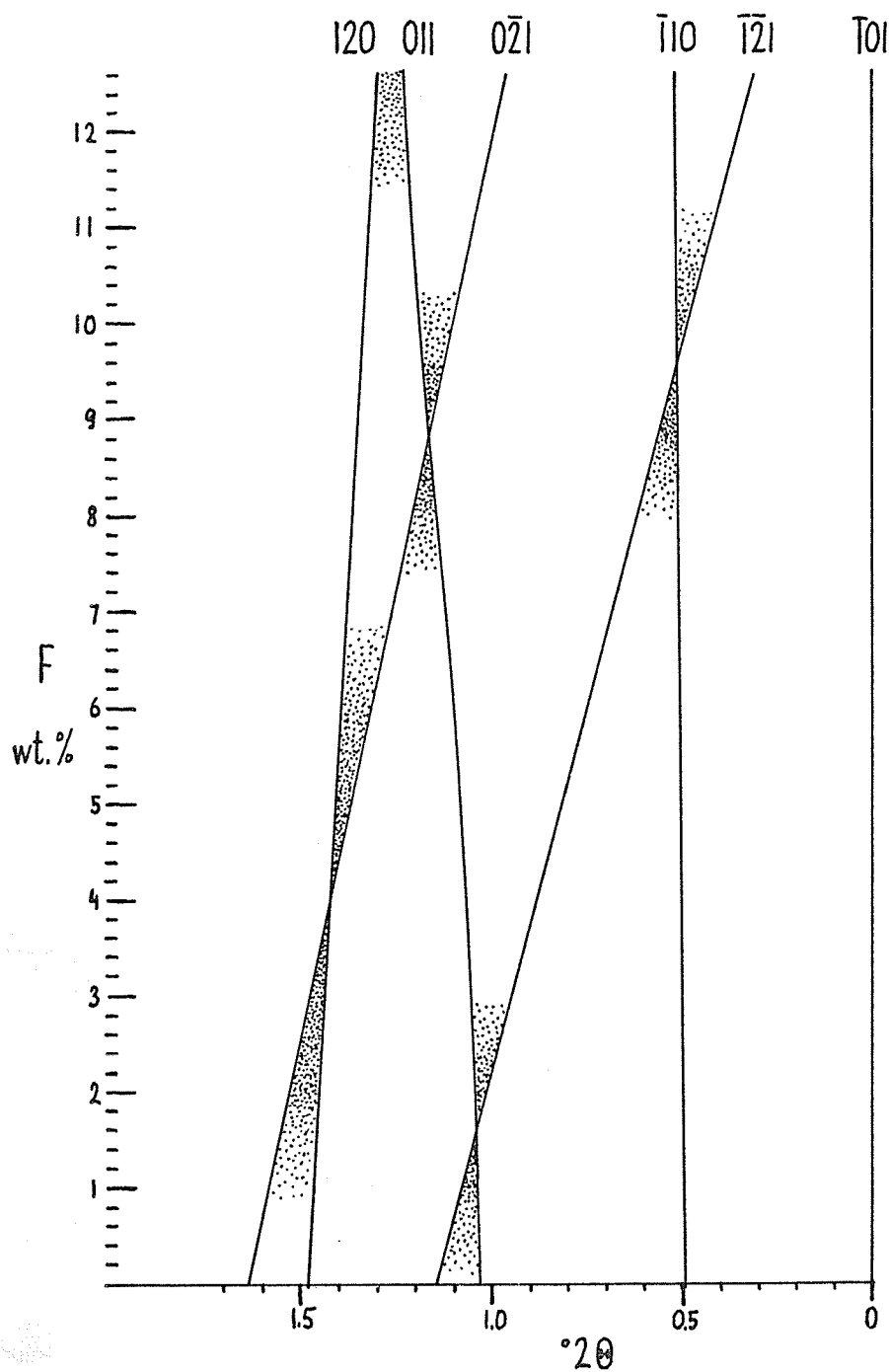


Fig.12. Chart for estimating the fluorine content of amblygonite-mon-tebrasite derived from Fig.11. The dotted areas denote ranges of possible overlap of two X-ray powder reflections.

decrease in changes of the peak positions may cause difficulties. It is desirable to take several X-ray records, preferably under the same conditions as those used for constructing the diagram and described above (or similarly detailed records on other diffractometer types), and to use averaged values or the best shaped record for the determination. Whenever possible, the contents of Na, K and Ca should be checked although low amounts of these elements do not seem to influence the diffraction patterns of amblygonite - montebrasite noticeably; however, the F-estimates should be treated cautiously if higher percentages of these elements are present.

II B - 5d. Unit cell dimensions from the powder and single-crystal

X-ray work

The unit cell dimensions derived from the single-crystal precession data are compared with corresponding values refined from X-ray powder records in Table 8. The table shows that the two sets of data generally match very well, similarly to those quoted by Haapala (1966, Table 14). This does not mean, however, that the unit cell dimensions based on the less accurate single-crystal work are sufficiently accurate for deriving F-contents.

There is a difference of almost one order between the powder- and single-crystal-based standard deviations of both axial lengths and interaxial angles in Haapala's data; single-crystal data by Baur (1959) also show high standard deviations. No standard errors are given by Simonov and Belov (1958), and they have not been calculated in the present study for the single-crystal data. Nevertheless, Haapala's and Baur's

TABLE 8

COMPARATIVE UNIT CELL DIMENSIONS
FROM SINGLE-CRYSTAL PRECESSION PHOTOGRAPHS AND FROM POWDER DIFFRACTOGRAMS

Specimen	a*	b*	c*	α or α^*	β^*	γ or γ^*
AF - 55	s 0.2102	0.1618	0.2143	113°50'	90°25'	111°10'
	p 0.2105	0.1627	0.2161	113°53'	90°29'	111°08'
A - 3	s 0.2112	0.1613	0.2157	68°00'	90°37'	111°15'
	p 0.2093	0.1620	0.2153	68°19'	90°30'	110°55'
A - 22	s 0.2080	0.1606	0.2127	112°25'	90°20'	110°50'
	p 0.2081	0.1614	0.2146	112°27'	90°25'	110°47'
AF - 47	s 0.2075	0.1608	0.2138	68°00'	90°40'	67°40'
	p 0.2078	0.1613	0.2145	68°19'	90°24'	67°48'

s - data from single-crystal work

p - values from powder data

precession-based figures show that the unit cell dimensions derived by single-crystal X-ray methods, unless special techniques are applied, are inferior to those refined from powder data in their precision.

The same fact is evident from Table 8. The differences between the two sets of figures exceed substantially the reliable standard errors calculated for the powder-data-based unit cell dimensions (Table 7 A,B) sometimes by two orders. Besides the different X-ray methods, the possible actual difference between the chemical composition of the precessed crystal fragment and the much larger powdered sample may also contribute to this difference. Since the changes in unit cell dimensions through the whole amblygonite - montebrasite range are rather small ($0.02 - 0.04\text{\AA}$, $0.5 - 1.5^\circ$), the lower precision of data based on standard single-crystal methods makes them unsuitable for estimates of the F-content, particularly when they are to be compared with a graph derived by another method.

II B - 5e. High- and low-temperature phases in the light of present results

In 1958, Simonov and Belov resolved for the first time the structure of amblygonite (without specifying the composition of the examined crystal). Baur (1959) confirmed the principal findings of the previous authors, studying a crystal from a pegmatite cavity; his crystal had a negligible Na-content and an alleged F/OH ratio of 1, derived from the structure determination. However, he discovered some differences in unit cell dimensions; some of his Al- and P-anion distances are significantly different; and he found that Li occupies one well-defined position in the crystal but two positions spaced $0.5\text{\AA}$ apart within the same cavity as did Simonov and Belov.

Baur supposes that, besides the possible differences in chemical composition, the two samples used for the structure determinations may differ in origin and structural state, the Russian material being a high-temperature, partly disordered form, and his crystal from a pegmatite cavity a low-temperature ordered type.

The unit cell dimensions given by Simonov and Belov (1958) and Baur (1959) for their crystals cannot be plotted in Figs. 9 and 10, since the real F-contents are not known, and the values are not accurate enough for its indirect estimate. In relation to the present authors' curves, Baur's data vary between 3% F and a negative, out-of-scale value, and the Russian sample shows a wide spread from pure amblygonite to F-rich montebrasite. Two gem-quality montebrasites from pegmatite cavities at Newry, Maine, probably very similar to Baur's low-temperature crystal, have been X-rayed by the author, and the results are plotted against the X-ray estimated F-contents in Figs. 9 and 10. The unit cell dimensions of those crystals coincide very well with the general trend of unit cell dimension changes in the whole amblygonite - montebrasite series, and fall in the approximate center of the range covered by Baur's unit cell dimensions. This indicates that if the high- and low-temperature forms of amblygonite - montebrasite really exist, they cannot be distinguished by the unit cell dimensions; and these results also suggest that the crystal examined by Baur was a F-poor montebrasite, rather than a hebronite-type phase with $F/OH \approx 1$, as he indicated. The present authors' work thus suggests that the structural differences between Baur's and the Russian specimens may be caused only by differences in chemical composition, since the crystal studied by them seems to be a F-rich amblygonite.

PART III. AMBLYGONITE - MONTEBRASITE IN THE TANCO (CHEMALLOY) PEGMATITE

CHAPTER III A. GEOLOGY OF THE TANCO PEGMATITE

The Tanco pegmatite is located under the northern shore of Bernic Lake, S. E. Manitoba, 100 miles ENE of Winnipeg. According to Wright (1963), it has been known since the late 1920's from a single drilling hole. Extensive drilling and shaft sinking programs have been carried out by several mining companies since 1954. At present, the pegmatite is being mined by the Tantalum Mining Corporation of Canada, Ltd. for tantalum, being one of the world's largest deposits of this metal; in addition, it contains the largest known reserves of cesium in the form of pollucite in the western world, and large amounts of iron-low spodumene suitable for refractory ceramics.

The first detailed reports on this pegmatite are those by Davies (1957) and Hutchinson (1959). Nickel (1961) and Nickel et al. (1963) have dealt with the mineralogy of the pegmatite; high lithium contents in amblygonite have been found in the first of these studies, as well as a predominance of the montebrasite (from refractive indices). The last geological report is that by Wright (1963), based on numerous drill cores and about 500 m. (metres) of underground workings.

Other studies being currently carried out on minerals from this pegmatite are the Li-Rb-Cs-micas by R. Rinaldi, the Ta, Nb-oxides by J. D. Grice, and the feldspars, petalite, spodumene and other minerals by P. Černý, all in the Mineralogical Laboratory at the University of Manitoba, under the direction of Professor R. B. Ferguson. Absolute age

determinations are being carried out by G. S. Clark and A. Penner, also at the University of Manitoba, and the geochemistry of K and Rb is being investigated by P. Marshall under the direction of R. W. Hutchinson and A. D. Edgar at the University of Western Ontario, London, Ontario.

The Tanco pegmatite is a flat elliptical body elongated in the E - W direction, dipping northwards up to 80m; at present it is known to be at least 800m long. It crosscuts an almost vertically foliated band of amphibolites located between a granitic body in the south and a metag^arywacke series in the north. The pegmatite as well as the neighbouring geological units are of Archean age.

According to Wright (1963), the Tanco pegmatite consists of six zones of primary crystallization: (1) quartz - albite border zone, (2) perthite - quartz - plagioclase - muscovite wall zone, (3) spodumene - perthite - plagioclase - quartz intermediate zone, (4) spodumene - quartz (-perthite) intermediate zone, (5) microcline - quartz intermediate zone, (6) quartz core. Generally the structural relationship of the consequent zones is that of concentric shells, from the margins of the pegmatite towards its center. These zones are partly replaced by three metasomatic units: (I) Aplitic albite is supposed to replace lower portions of the spodumene - bearing zones; (II) fine-grained lepidolite replaces parts of the microcline - quartz intermediate zone; and (III) pollucite bodies under the roof of the pegmatite are^{q/sv} supposed to be of metasomatic origin.

The metasomatic units of Wright may be subject to reinterpretation, and the sequence of crystallization of the primary zones may

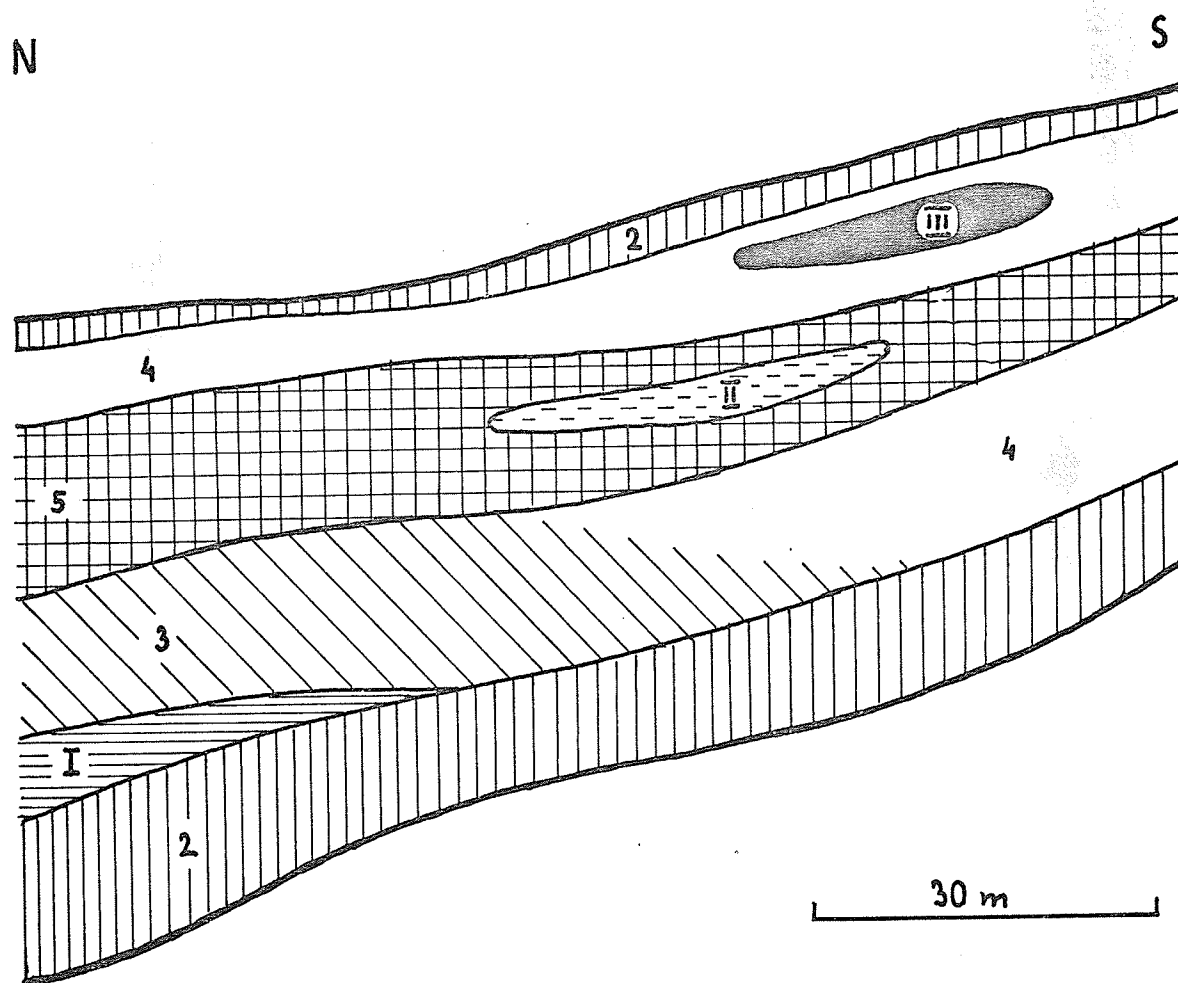


Fig.13. Schematic cross-section of the Tanco pegmatite, close to the sampled area. From section AA', Plate 1, in Wright (1963). Numbers correspond to individual zones and units as distinguished by Wright. 2 - Kf-qtz-plg-ms zone, 3 - spod-Kf-plg-qtz zone, 4 - spod-qtz-(Kf) zone, 5 - Kf-qtz zone, I - aplitic albite, II - lepidolite, III - pollucite.

undergo minor changes, too (P. Černý, personal communication, 1970). However, from a descriptive viewpoint, his subdivision of the Tanco pegmatite is satisfactory, and the new underground workings have not changed the general relations of the zones and units as illustrated by Wright (1963, Plate I). A part of his N - S cross-section, close to the part of the pegmatite body sampled here is shown in Fig.13.

CHAPTER III B. SAMPLING AND LABORATORY STUDY

The sampling of the amblygonite - montebrasite in the Tanco mine was done in as detailed a manner as possible, the limited time for laboratory examination of the specimens being the main restricting factor. Figs. 14, 15, 16 show the location of samples in the 1st level shaft station, the main drift, the west drift, the pollucite sublevel, and the ladder stope connecting it with the first level. This^{is} the amblygonite-bearing area in that part of the mine that was accessible in the summer of 1969. At the present time (mid-1970), however, the major part of the 1st level main drift between the shaft station and the junction with west drift has been mined out.

Within each petrographic unit of the pegmatite containing amblygonite - montebrasite minerals, numerous specimens were collected of each colour variety, particularly from individuals showing colour zoning. Attention was given to the immediately neighbouring phases, and specimens adjacent to K-feldspar, albite, spodumene, pollucite, etc., were sampled wherever possible. The large blocks that occur in some zones were sampled at several spots, either irregularly or along

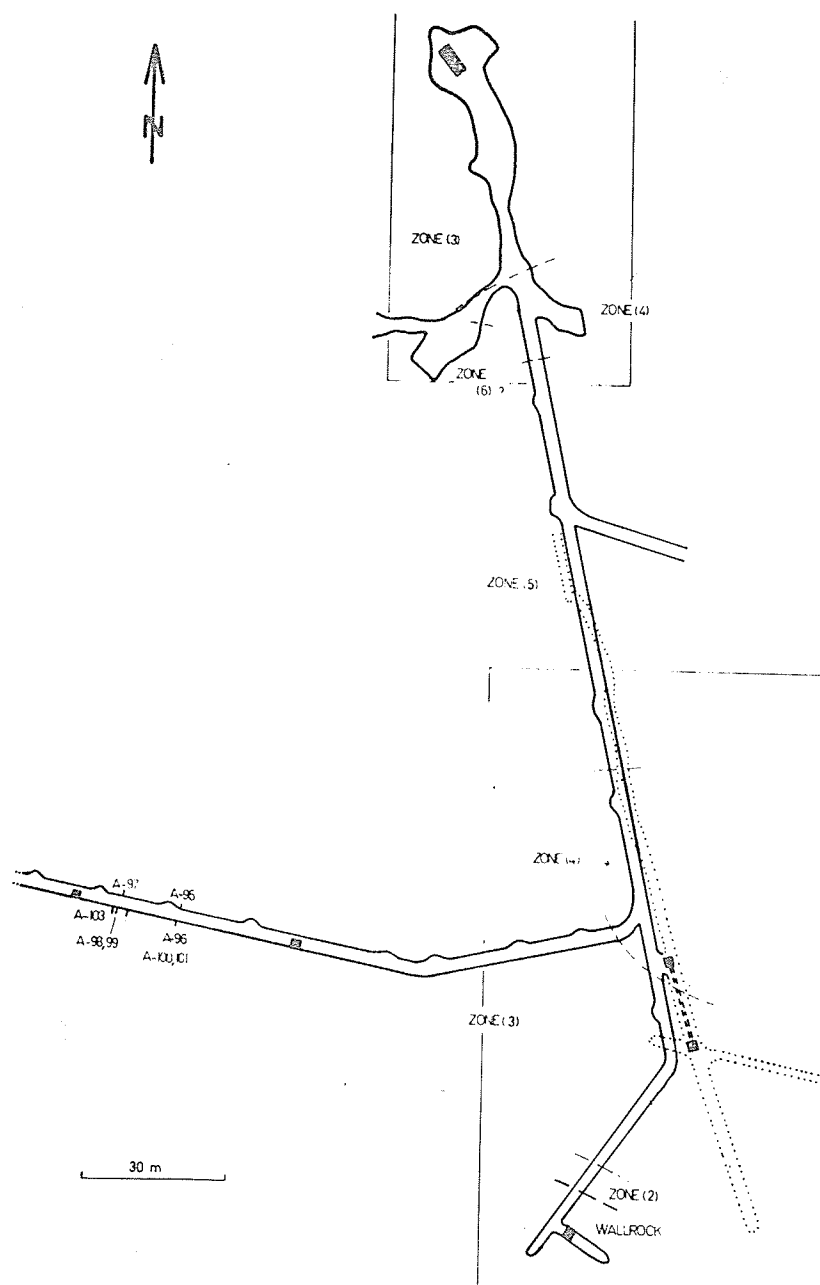


Fig.14. Map of the sampled area in the 1st level of the Tanco mine, with the overlying pollucite sublevel dotted. Dashed lines - schematic boundaries of pegmatite zones; location indicated for specimens taken in the west drift.

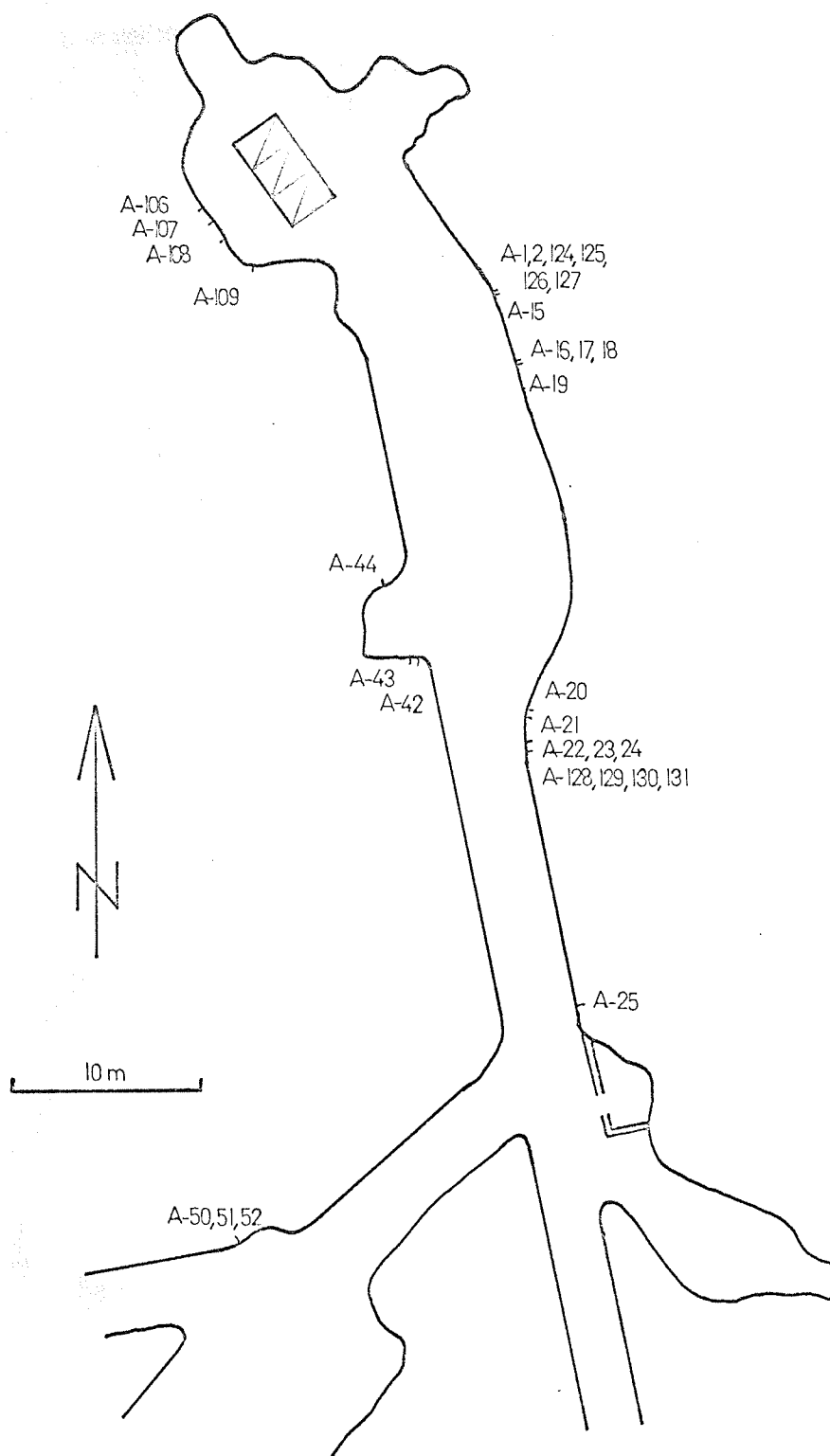


Fig.15. Map of the surroundings of the 1st level shaft station, showing location of specimens sampled in this area.

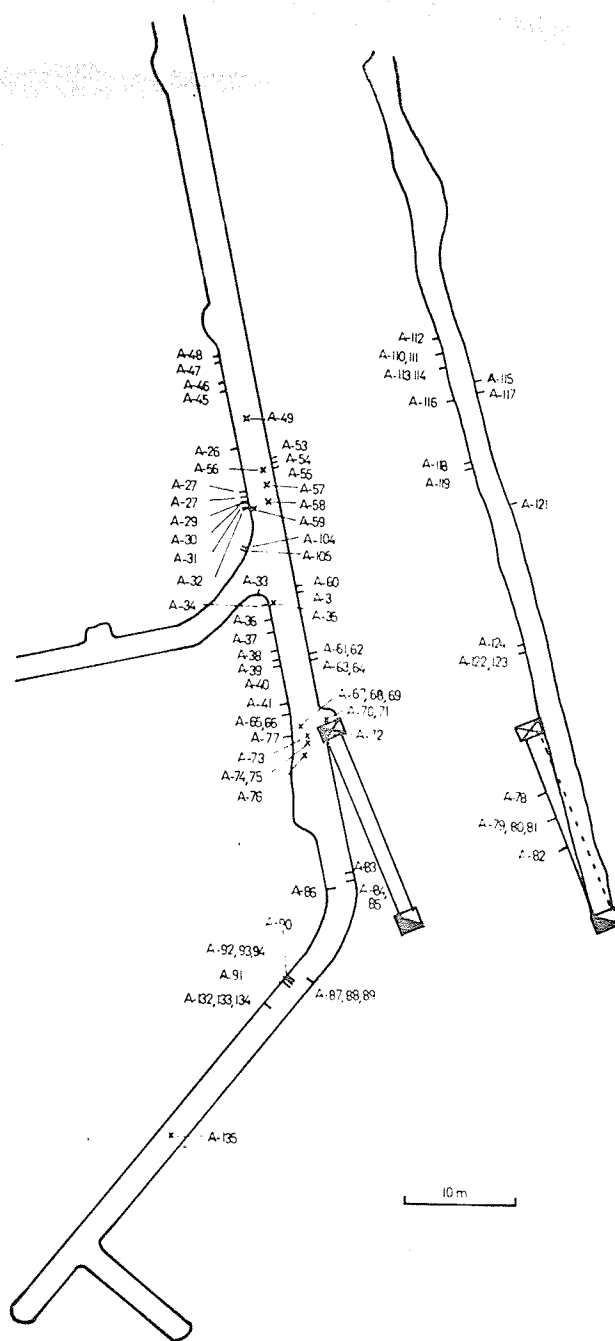


Fig. 16. Map of the pollucite sublevel, adjacent parts of the 1st level main drift, and of the connecting ladder, showing location of specimens taken in this area; the separate plan of the pollucite sublevel is shifted eastwards, as indicated by the ladder repeated in both drawings, to avoid overlap.

traverses. Both primary and altered material were collected in numerous crystals showing changes in colour along margins and cross-cutting fissures.

All specimens collected were X-rayed to estimate their F-contents, and 33 samples representing different types from different zones were chemically analyzed for Na_2O , K_2O and CaO (Table 9). Thin sections were prepared when necessary, and several suitable specimens were included in the series of 12 specimens analyzed for F and examined in detail, for their physical properties. The list of specimens from the Tanco mine is given in Appendix I, along with F-estimates and other available information.

CHAPTER III C. DISTRIBUTION OF THE AMBLYGONITE - MONTEBRASITE,

ITS TYPES AND ALTERATIONS

The portion of the pegmatite sampled was rather small but it contained all the zones and units found in the whole body (with the possible exception of the true quartz core). Thus the results should be characteristic of the whole pegmatite, although shifts in the general pattern cannot be excluded between lower and upper parts of the gently dipping body.

Wright (1963) indicated that amblygonite occurs in three of his zones: (3) the spodumene - perthite - plagioclase - quartz intermediate zone; (4) the spodumene - quartz - (perthite) intermediate zone; and (6) the quartz core. In the present study, amblygonite - montebrasite was found in the same zones, and in addition one specimen was located

TABLE 9

THE CONTENTS OF Na_2O , K_2O AND CaO IN 35 SELECTED SAMPLES

OF AMBLYGONITE - MONTEBRASITE FROM THE TANCO PEGMATITE

Specimen	Zone	Na_2O	K_2O	CaO
A-135	Wall zone (2)	0.167	0.039	0.029
A-125	Zone (3)	0.094	0.012	0.017
A-126c	Zone (3)	0.267	0.019	0.067
A- 1	Zone (3)	0.054	n.d.	0.17
A-126a	Zone (3)	0.068	0.007	0.023
A-126b	Zone (3)	0.043	0.005	0.023
A- 2	Zone (3)	0.025	n.d.	0.049
A-127b	Zone (3)	0.141	0.012	0.049
A- 24	Zone (3)	0.077	0.004	nil
A- 42a	Zone (3)	0.139	0.009	0.005
A- 98	Zone (3)	0.020	nil	nil
A-101a	Zone (3)	0.050	0.003	0.015
A-101c	Zone (3)	0.227	0.005	nil
A-100a	Zone (3)	0.022	0.042	0.032
A-100b	Zone (3)	0.052	0.007	nil
A-42b	Zone (3)	0.178	0.032	0.173
A-99	Zone (3)	0.025	0.013	nil
A- 33	Zone (4)	0.064	0.007	0.016
A- 34	Zone (4)	0.035	0.007	0.015
A- 36	Zone (4)	0.061	0.007	0.015
A- 37	Zone (4)	0.024	0.007	0.030
A- 38	Zone (4)	0.058	0.007	0.065
A- 39	Zone (4)	0.046	0.007	0.017
A- 3	Zone (4)	0.039	n.d.	0.22
A- 68c	Zone (4)	0.032	0.116	0.002
A- 67b	Zone (4)	0.034	0.108	nil
A- 67a	Zone (4)	0.038	0.036	0.050
A- 68b	Zone (4)	0.070	0.023	0.002
A-117	Zone (6)	0.086	0.009	0.028
A-118	Zone (6)	0.022	0.007	0.032
A-112	Zone (6)	0.066	0.023	0.077
A-116	Zone (6)	0.024	0.007	0.012
A-110b	Zone (6)	0.039	0.015	nil
A-123	Zone (6)	0.076	0.003	nil
A-122	Zone (6)	0.029	0.020	nil
A-110a	Zone (6)	0.034	0.014	n.d.

in Wright's zone (2), the perthite - plagioclase - quartz - muscovite wall zone.

There are, however, some doubts about the identity of the quartz core in the area investigated here: the spodumene - rich zone (4) passes gradually into large quartz-rich portions with only minor spodumene, amblygonite - montebrasite, and alkali feldspar that fit Wright's description of the quartz core (zone 6) but could not be separated from zone 4 in the course of pegmatitic crystallization by the perthite - quartz zone (5), as proposed by Wright (1963).

Despite these uncertainties, whose investigation is beyond the scope of this study, we can summarize that amblygonite - montebrasite occurs only in the zones of primary crystallization, and that the crystallization sequence of these amblygonite - montebrasite - bearing zones corresponds to that proposed by Wright, disregarding the uncertainty in the identity of the quartz core and the possible "interstratified" amblygonite-free zone (5). In the following discussion the amblygonite - montebrasite - carrying zones are quoted as zones (2), (3), (4), and (6), to relate them to the only detailed description of the pegmatite published to date. The boundaries between these zones in the sampled area are shown in Figs. 14, 15, 16.

In the following paragraphs, the primary amblygonite - montebrasite of the individual zones is discussed first, and the secondary montebrasite and alteration products of the amblygonite - montebrasite are discussed afterwards.

The wall zone (2) consists of medium - grained albite and quartz

with disseminated coarse nests and blocks of alkali feldspar, quartz, muscovite and beryl. Montebrasite was found in one of these nests as irregular, white crystal, 4cm in diameter and with thin rusty coatings on contacts with other minerals. The wall zone was exposed in a rather small area in the end of the main 1st level drift (Fig.16), and it was quite difficult to distinguish montebrasite from alkali feldspars and beryl here. Montebrasite may be more widespread in this zone than is suggested by the single find, but it seems to be only an accessory mineral here; the fact that Wright (1963) does not report it from the zone (2) supports this conclusion.

The intermediate zone (3) consists mainly of alkali feldspars, albite, quartz, lithian muscovite and amblygonite - montebrasite. The textures seem to be rather irregular and complicated, but the most frequent pattern seems to be clusters of large alkali feldspars with fine-grained albite, muscovite, and quartz, rimmed by cleavelandite and muscovite against quartz pods with amblygonite - montebrasite. Spodumene occurs in both alkali feldspar-rich aggregates and in quartz. The orientation of the wedge-shaped crystals of amblygonite - montebrasite normal to the cleavelandite/quartz contacts, and their colour zoning suggests that this mineral was formed after the feldspathic masses, and grew from their surface towards the space occupied by quartz (Fig.17). Irregular blocks of amblygonite - montebrasite attain a size of 60cm in this zone; smaller crystals up to 20cm in diameter tend to have distinct crystal faces.

The predominant colours of amblygonite - montebrasite are white and yellow, the latter being characteristic of the outer shells of amblygonite - montebrasite crystals, or the last formed a symmetric zones.



Fig.17. Large crystals of amblygonite-montebrazite (white) in quartz, surrounding an albite - alkali feldspar - mica aggregate rimmed by a band of radial cleavelandite and mica. Zone (3), close to the 1st level shaft station.

Pale - pink amblygonite is rather rare; it never occurs in isolated crystals but is always accompanied by younger white and yellow zones, forming the earliest part of the grains (Fig.18).

Zone (4) contains the largest lithium concentration among the zones and units of the Tanco pegmatite, because of very abundant spodumene (Wright,1963). Large, flat blocks of spodumene + quartz intergrowths and quartz are the main constituents, accompanied by subordinate amblygonite - montebrasite and minor amounts of alkali feldspar and albite. Amblygonite - montebrasite is white throughout this zone, and its masses reach up to 1.5m in maximum dimension (Fig.19). These large accumulations are usually irregular, but small crystals 5 - 15cm in size are frequently well bounded by crystal faces. Generally, amblygonite - montebrasite "floats" freely in quartz or is attached to spodumene + quartz aggregates. It is euhedral against them in some cases, but it also surrounds euhedral blocks of spodumene + quartz; the crystallization of these phases was evidently overlapping. Direct contact with feldspars is less frequent.

The above description fits also the amblygonite - montebrasite from around the ladder to the pollucite sublevel (Fig.16); this area is transitional to zone (6).

Zone (6) seems to be, as explained earlier, rather a quartz-rich variant of the previous zone, overlying it and being overlain by pollucite. Its contact with the latter is very sharp, but a gradual transition connects zones (4) and (6). Quartz predominates considerably over spodumene and amblygonite - montebrasite in zone (6), and alkali feldspar

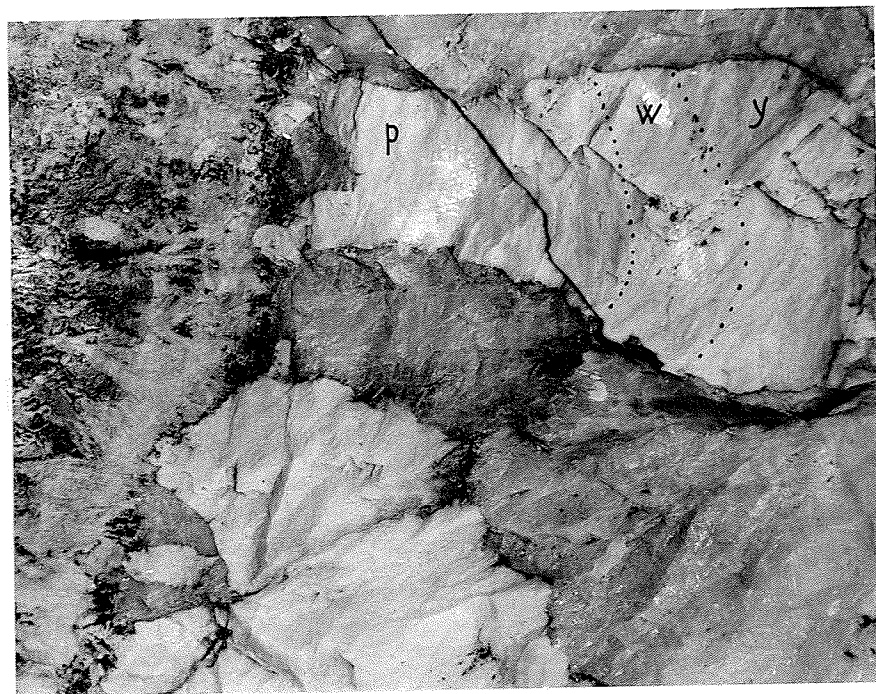


Fig.18. Detail from Fig.17, showing colour zonation typical of amblygonite-montebbrasite in zone (3); p - pink, w - white, y - yellow.



Fig.19. Part of a large block of white amblygonite-montebbrasite, surrounded by grayish quartz containing small crystals of this mineral. Zone (4), close to pollucite ladder.

and albite are very subordinate; dark blue Mn-apatite is a characteristic constituent at the contact with pollucite. Amblygonite - montebrasite forms irregular blocks up to 50cm in length, without any distinct tendency to crystal shape. It is white or yellow, the yellow material forming either separate crystals or outer zones around the white cores. Amblygonite - montebrasite is imbedded usually in quartz, but adjoins also spodumene, feldspars, and pollucite (Fig.20).

In all four zones, the primary amblygonite - montebrasite described above is frequently rimmed or penetrated by late, secondary montebrasite. This secondary phase is always darker, brownish, gray or greenish in colour. Its luster is rather dull and the cleavage not so conspicuous as in the primary minerals. The secondary montebrasite always forms at the expense of the primary phases, without penetrating the adjacent quartz, feldspar or spodumene, and in most instances the replacing montebrasite maintains a crystallographic continuity with the primary amblygonite - montebrasite (Fig. 21). Besides this, the alteration is always accompanied by extensive twinning which seems to develop without mechanical strain or cataclastic effects (Fig.22). The altered areas are usually surrounded by haloes of the primary mineral penetrated by a very fine network of the secondary phase (Fig.23). The alteration is often difficult to observe in hand specimens when the difference in colours is not conspicuous. It should be stressed that in most cases the secondary montebrasite is not accompanied by other late phases except for some microscopic veinlets of micaceous material (Fig.21).

Besides this "autopseudomorphism", amblygonite - montebrasite is frequently altered to other minerals. These secondary assemblages were

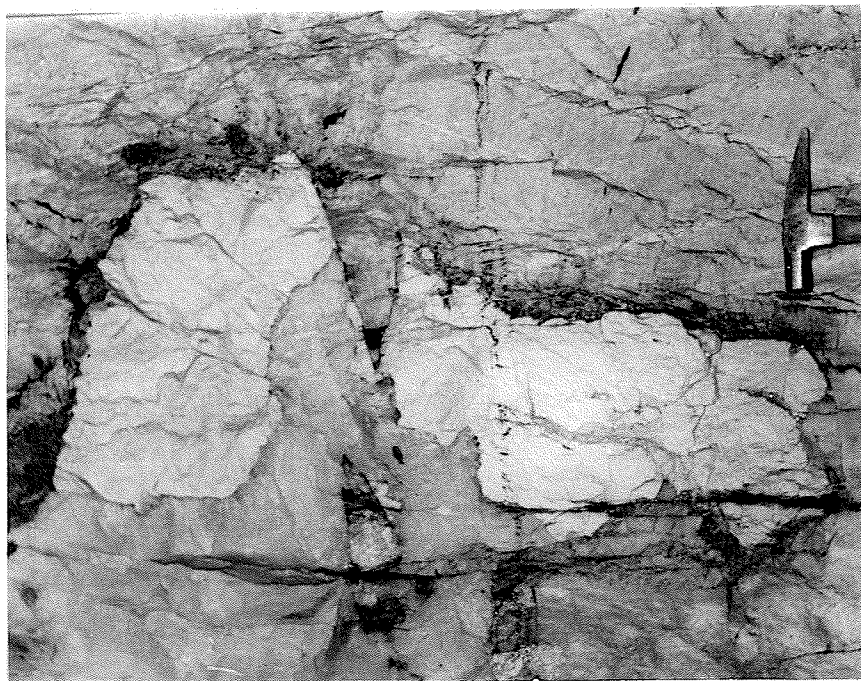


Fig.20. Amblygonite-montebbrasite crystals (white) at the contacts of pollucite (top) and quartz (bottom) of zone (6). Few spodumene + quartz aggregates (dark, lower center) in quartz.

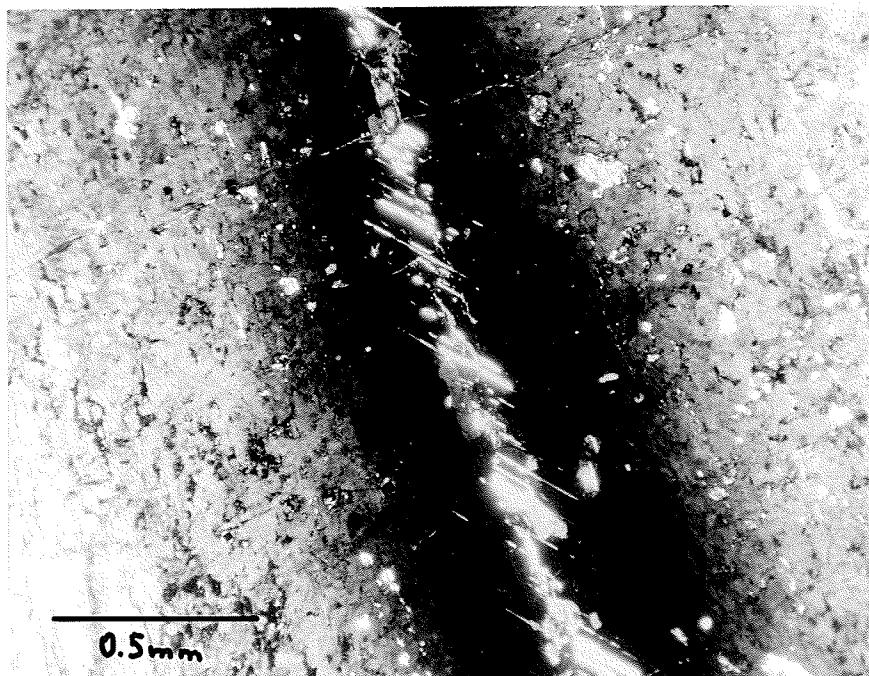


Fig.21. Veinlets of secondary montebbrasite (dark) with few twinning lamellae (white) in a slightly altered primary amblygonite-montebbrasite (grayish, *left* and *right*). Gradual darkening from the primary to the secondary mineral caused by gradual change in optical orientation. Sample A-46, crossed nicols.

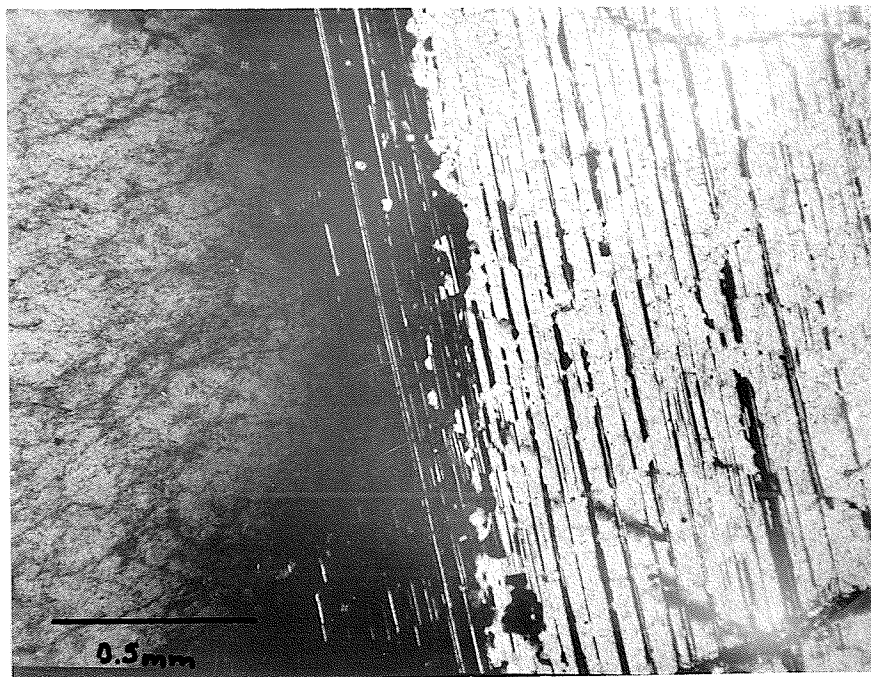


Fig.22. Primary amblygonite-montebbrasite (grayish, left side) passes gradually into secondary montebbrasite (center, black, fully extinct) which becomes intensely twinned (black and white, right side). Sample A-22,23, crossed nicols.

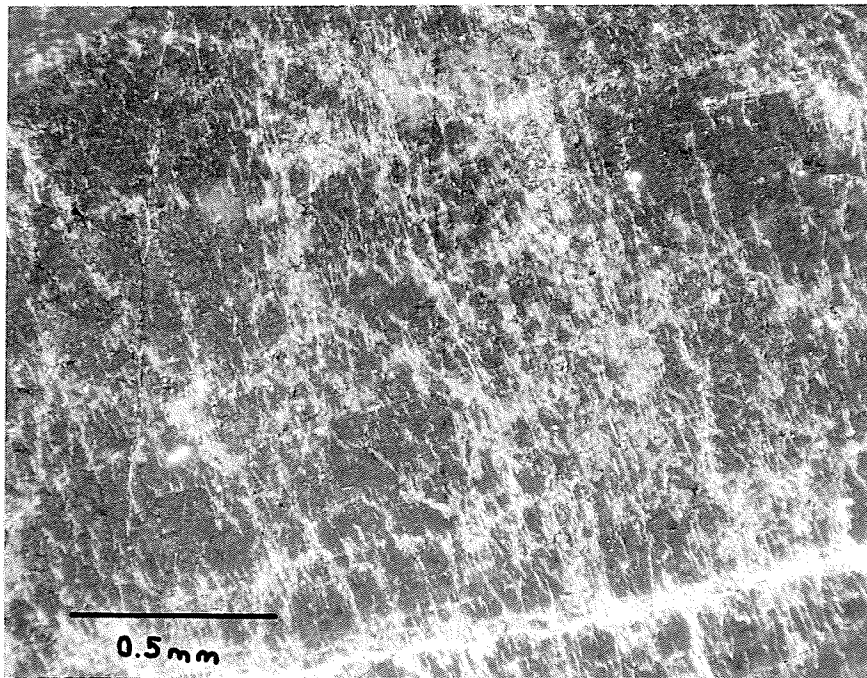


Fig.23. Irregular fine network of secondary montebbrasite (white) penetrating its parent amblygonite-montebbrasite phase (gray). Sample A-42, crossed nicols.

not investigated systematically and the results may be subject to later corrections. At present two characteristic associations are well established : F-apatite + muscovite + quartz and lithian muscovite + quartz. The first of these often covers the outer surface of the secondary montebrasite rims; the second does not create any changes in the replaced amblygonite - montebrasite.

CHAPTER III D. CHEMICAL COMPOSITION OF AMBLYGONITE - MONTEBRASITE

Besides the few chemical determinations of fluorine, the content of this element has been estimated by the X-ray method described in section II B - 5c in all 157 collected specimens which are listed in Appendix I. The possible interference from substituting ^{ti} cations should be negligible, since a series of 35 specimens, representing all types from all zones, did not show any appreciable contents of Na₂O, K₂O and CaO. For better comparison, the F-percentages of all specimens have been plotted in Fig. 24, in separate groups for each zone. Within one zone, the specimens are arranged from the F-richest at the top to the F-poorest at the bottom, against a horizontal scale of wt.% F. The colours of the primary phases and the secondary minerals are distinguished by different symbols.

The single specimen found in zone (2) is not included in the diagram and will not be discussed at all. According to its general character, it may be somewhat altered, and even if a true primary phase, it cannot tell anything general and reliable about the composition of the amblygonite - montebrasite in its parent zone.

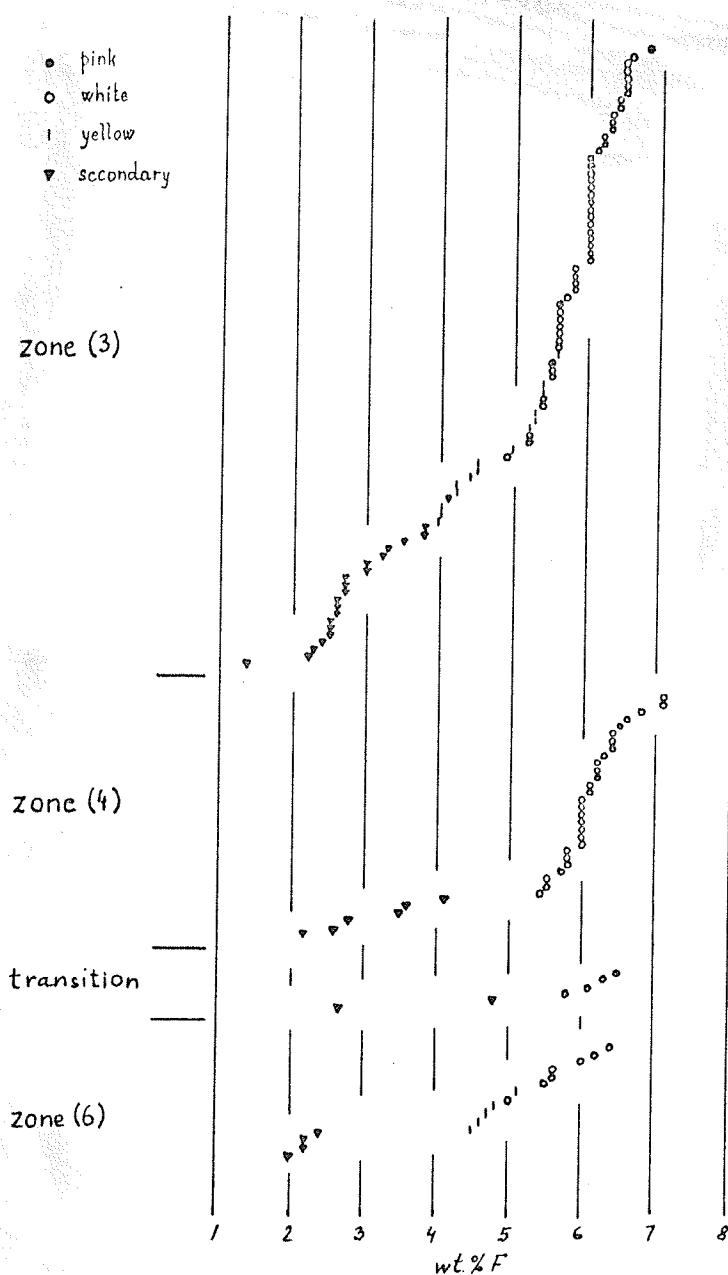


Fig.24. Fluorine content and colour of amblygonite-montebbrasite in the Tanco pegmatite; all specimens arranged according to the decreasing fluorine content in each separate zone. Empty circles - white; solid circles - pink; vertical bars - yellow; triangles - secondary montebrasite. Zones numbered according to Wright (1963).

As shown in Fig.24, the primary amblygonite - montebrasite shows a remarkable range of F-contents within each zone : 6.8 - 4.0% in zone (3), 7.1 - 5.4% in zone (4), and 6.4 - 4.5% in zone (6). Within each of zones (3) and (4), the most frequent compositions are those around the centre of the respective range, 5.5 - 6.0% F and 6.0 - 6.2% F, respectively. In zones (3) and (6) that contain early white and late yellow phases, the white material is generally richer in F than the yellow phase. In pairs of white and yellow samples coming from a single crystal, the yellow younger material is always F-poorer of the two. The few pink specimens occurring in zone (3) are disseminated over the range of F-contents covered only by white amblygonite - montebrasite in this zone - 7.1 to 5.4%. However, the F-content of each of these pink specimens is always higher than that of the overgrowing white and yellow material. In these pink specimens the sequences are: 6.0, 5.6 (pink) - 5.2 (white) - 4.4% F (yellow); 6.0 (pink) - 5.5% F (yellow); 5.56 (pink) - 3.65% F (yellow); 6.8 (pink) - 5.6, 5.4% F (white).

As to the secondary phases, their compositions group quite separately from the primary minerals at the montebrasite end of the F-scale, in each pegmatite zone. This shows that the alteration of early amblygonite - montebrasite always produces F-poorer montebrasite. Inspection of individual pairs of "parent" and "daughter" phases has shown that the difference between their F-contents often reaches as much as 4%.

The Na_2O , K_2O and CaO contents of a selected group of amblygonite - montebrasite specimens (Table 9) are generally very low, and show no evident differences among different zones, colour types, and generations. The very subordinate percentages of these oxides vary within the

same ranges in all types of amblygonite - montebrasite, found in the Tanco pegmatite.

There seems to be an evident relation between the F-content and colour of the white and yellow phases. The F-content of yellow specimens is always lower than 5.5%, and that of white material mostly higher than this value with only a few specimens reaching 4.9% F. This can be observed in both zones containing white and yellow phases, (3) and (6) (Fig.24). In contrast, the F-percentages of the few pink specimens are distributed over the whole range of white samples. (It is interesting to note, that the only pink montebrasite found at Varutråsk shows the lowest F-content, 1.8% F, of all three specimens analyzed from this locality.) It is unlikely that the white and yellow colour could be caused by the different F/OH ratios; some trace elements or structural effects may be the cause of varying colours, but no data are available from the literature, and this study did not reveal any possible explanation.

CHAPTER III E. CHANGES IN CHEMICAL COMPOSITION OF AMBLYGONITE-MONTEBRASITE

DURING THE CRYSTALLIZATION OF THE PEGMATITE

As shown in Chapter III C, amblygonite - montebrasite occurs only in the zones of primary crystallization. It was not found in the units designated as metasomatic by Wright (1963). The late, secondary montebrasite replacing the primary amblygonite - montebrasite is not related to the origin of the supposed metasomatic units.

The primary amblygonite - montebrasite originated during a relatively long period of the pegmatite solidification, and it shows

remarkable changes in the F/OH ratio during its crystallization in four zones. On the contrary, the Na_2O , K_2O and CaO contents are negligible and quite uniform in amblygonite - montebrasite from all four zones. In zone (3) the primary amblygonite - montebrasites exhibit a very wide range of F-contents, 6.8 - 4.0% (Fig. 24). In zone (4), the compositions cover the upper half of the range (7.1 - 5.4% F) characteristic of the white and pink amblygonite - montebrasite of zone (3). Thus the average F-percentage is distinctly higher in zone (4) than in zone (3) (6.1 and 5.6%, respectively). As Figs. 16 and 24 show, the range of the F-contents and their average decrease gradually from the examined part of zone (4), through the transitional area around the pollucite ladder towards zone (6) below the pollucite body, where the range is 4.5 - 6.4% F, and the average is the lowest at 5.4% F.

Within individual pegmatite zones, the crystallization of primary amblygonite - montebrasite always proceeds from higher to lower F-contents. This is most conspicuous in zone (3) where the zoned crystals with pink, white and yellow material show a span up to 1.9% F, and furthermore the same order of decrease in F during crystallization of a single crystal is attained in the white - yellow sequences of zone (6).

We may conclude that the average F-content of primary amblygonite - montebrasite in the Tanco pegmatite increases from 5.6% in zone (3) to 6.1% in zone (4), and then gradually decreases to 5.4% F in zone (6). The same trend is also followed by the ranges of F-contents in these zones. Within each single zone, the individual crystals frequently show a decrease in the F-content during their growths, reaching up to 1.9%.

The F-contents of the secondary montebrasites are generally much lower than those of their parent phases. The averages in individual zones seem to change roughly parallel to the changes in averages of the primary phases: from 2.7 in zone (3) to 3.2 in zone (4) and 2.2% F in zone (6). Thus the late alteration seems to decrease the F-content of the primary amblygonite - montebrasite by about the same amount in all zones.

The uniform and negligible percentages of Na_2O , K_2O and CaO seem to be normal for amblygonite - montebrasite crystallizing in the primary pegmatite zones because alkali-rich specimens are characteristic of metasomatic units, according to Heinrich and Corey (1955) and Quensel (1956).

It is difficult to compare these results with the few data found in the literature (see Chapter I B). There is a general agreement between the situation in the late primary zones of the Tanco pegmatite and the statement of Boruckij (1966) that amblygonite - montebrasite becomes F-poorer during pegmatite crystallization. No comparison can be made with the results of Quensel (1937, 1956), since two out of three of his analyzed specimens are claimed to come from metasomatic units. In other occurrences, only one specimen was investigated (e.g. Volborth, 1951) and no data on amblygonite - montebrasite from different pegmatite units are available.

APPENDIX I

LIST OF SPECIMENS FROM THE TANGO PEGMATITE

No.	Location	Description	DTA	Sp.gr.	UCD	Na,K Ca	F% wt. estim.	F% wt. anal.
1	Zone 3	Pink		x	x	x		5.56
2	Zone 3	Yellow	x	x	x	x		3.65
3	Zone 4	Colourless		x	x	x		6.30
4	Dump	White	x	x	x	x		4.05
5	Dump	Yellow		x	x	x		4.51
12	Zone 6	White					5.2	
13	Not localized	White		x	x			6.43
14	Not localized	White					5.5	
15	Zone 3	White					5.4	
16	Zone 3	White					5.4	
17	Zone 3	Pink					6.8	
18	Zone 3	White					5.6	
19	Zone 3	White					5.6	
20	Zone 3	Colourless					6.0	
21	Zone 3	Colourless					6.0	
22	Zone 3	Brownish	x	x	x	x		1.40
23	Zone 3	Brownish					1.4	
24	Zone 3	White					6.0	
25a	Zone 3	White					6.0	
25b	Zone 3	Colourless					6.5	
27	Zone 4	Colourless					6.4	
28	Zone 4	White					6.2	
29	Zone 4	White	x	x	x			6.30
30	Zone 4	White					5.5	
31	Zone 4	White					6.2	
32	Zone 4	Colourless					6.3	
33	Zone 4	White					6.0	
34	Zone 4	White					5.8	

No.	Location	Description	DTA	Sp.gr.	UCD	Na,K Ca	F% wt. estim.	F% wt. anal.
35a	Zone 4	Colourless					6.4	
35b	Zone 4	Pale beige					4.1	
35c	Zone 4	White					6.0	
35d	Zone 4	Brownish					2.8	
36	Zone 4	White				x	6.2	
37	Zone 4	White				x	6.0	
38	Zone 4	White				x	7.1	
39	Zone 4	White				x	6.0	
41	Zone 4	White					6.0	
42a	Zone 3	White				x	5.6	
42b	Zone 3	Brownish				x	2.5	
42c	Zone 3	Brownish					1.4	
43	Zone 3	White					5.6	
44a	Zone 3	White					4.5	
44b	Zone 3	Gray					2.0	
44c	Zone 3	Gray					3.8	
45	Zone 4	White					6.1	
46a	Zone 4	White					5.8	
46b	Zone 4	White					5.6	
47	Zone 4	White					6.0	
48a	Zone 4	White					5.8	
48b	Zone 4	Brownish					2.0	
49	Zone 4	White					5.4	
50	Zone 3	Yellow					5.4	
51a	Zone 3	Yellow					5.6	
51b	Zone 3	Yellow					5.3	
52	Zone 3	Yellow					5.3	
53	Zone 4	White					6.8	
54	Zone 4	White					5.8	
55	Zone 4	White					6.6	
56	Zone 4	Pale beige					3.6	
57	Zone 4	White					6.1	

No.	Location	Description	DTA	Sp.gr.	UCD	Na,K Ca	F% wt. estim.	F% wt. anal.
58	Zone 4	Grayish white					2.2	
59	Zone 4	White					3.5	
60	Zone 4	White	x	x	x			6.17
61	Zone 4	White					6.1	
62	Zone 4	White					6.0	
63	Zone 4	White					5.8	
64	Zone 4	Colourless					6.4	
65	Zone 4	White					6.5	
66	Zone 4	White					5.7	
67a	Zone 3	Gray				x	2.6	
67b	Zone 3	Yellow				x	4.2	
68a	Zone 3	White					5.2	
68b	Zone 3	Gray				x	2.5	
68c	Zone 3	Yellow				x	5.4	
69	Zone 3	Yellow					4.5	
70	Zone 3	Colourless					6.5	
71a	Zone 3	Greenish gray					3.0	
71b	Zone 3	White					6.6	
72	Zone 3	Beige	x	x	x			3.17
73	Zone 3	Brownish					2.6	
74a	Zone 3	White					6.0	
74b	Zone 3	Beige					2.5	
75a	Zone 3	White					6.0	
75b	Zone 3	Beige					2.5	
76	Zone 3	Colourless					6.5	
77	Zone 3	White to beige						
78	Zone 6	Gray					2.7	
79	Zone 6	White					6.1	
80	Zone 6	White					6.5	
81a	Zone 6	Colourless					6.3	
81b	Zone 6	White					4.8	
82	Zone 6	White					5.5	

No.	Location	Description	DTA	Sp.gr.	UCD	Na,K Ca	F% wt. estim.	F% wt. anal.
83	Zone 3	Colourless					6.0	
84	Zone 3	Colourless					6.2	
85	Zone 3	White					5.8	
86	Zone 3	Brownish					2.7	
87	Zone 3	Colourless					6.3	
88a	Zone 3	Colourless					6.4	
88b	Zone 3	White					2.7	
89	Zone 3	White					5.7	
90	Zone 3	White					6.5	
91	Zone 3	Colourless					6.0	
92	Zone 3	White					6.4	
93	Zone 3	White					5.5	
94	Zone 3	Colourless					6.2	
95	Zone 3	White					5.8	
96	Zone 3	White					6.0	
97a	Zone 3	White					6.5	
97b	Zone 3	Grayish					6.1	
98	Zone 3	Yellow	x	x	x	x		3.44
99	Zone 3	Gray				x	2.2	
100a	Zone 3	Yellow				x	3.8	
100b	Zone 3	Gray				x	3.5	
100c	Zone 3	Yellow					4.2	
100d	Zone 3	Yellow					4.0	
101a	Zone 3	White				x	6.3	
101b	Zone 3	White					6.0	
101c	Zone 3	Yellow				x	4.9	
103	Zone 3	Greenish gray	x				2.2	
104	Zone 4	White						
105	Zone 4	White					5.5	
106	Zone 3	White					6.3	
107a	Zone 3	White					5.5	
107b	Zone 3	Pink					6.0	

No.	Location	Description	DTA Sp.gr. UCD	Na,K Ca	F% wt. estim.	F% wt. anal.
108	Zone 3	Gray			3.2	
109a	Zone 3	Gray			3.3	
109b	Zone 3	White			5.6	
110a	Zone 6	Gray		x	2.2	
110b	Zone 6	Yellow		x	4.6	
111a	Zone 6	Yellow			5.1	
111b	Zone 6	Gray			2.4	
112	Zone 6	Yellow		x	4.7	
113	Zone 6	White			5.6	
114	Zone 6	Yellow			4.5	
115	Zone 6	Beige			2.2	
116	Zone 6	Yellow		x	4.8	
117	Zone 6	White		x	5.5	
118	Zone 6	White		x	6.0	
119	Zone 6	White			5.0	
121	Zone 6	White			5.6	
122	Zone 6	Brownish		x	2.0	
123	Zone 6	White		x	6.2	
124	Zone 6	White			6.4	
125	Zone 3	Pink		x	6.0	
126a	Zone 3	White		x	5.2	
126b	Zone 3	Yellow		x	4.4	
126c	Zone 3	Pink		x		5.66
127a	Zone 3	Gray			2.6	
127b	Zone 3	Yellow		x	4.0	
128	Zone 3	Gray			2.3	
129	Zone 3	White			6.0	
130	Zone 3	White			5.5	
131	Zone 3	White			6.0	
132	Zone 3	Yellow			4.0	
133	Zone 3	Yellow			4.7	
134	Zone 3	White			6.7	
135	Zone 2	White		x	3.7	

APPENDIX II

LIST OF SPECIMENS FROM OTHER LOCALITIES THAN THE Tanco Pegmatite

No.	Locality	Source	Source No.	DTA	UCD	Sp.gr.	F% wt. estim.	F% wt. anal.
AF- 1	Montebras, France	Dept. Geol. U of M	41.5.5.1-2	x	x	x		7.85
AF- 2	Merrit Mine, Paris, France	Dept. Geol. U of M	41.5.5.1-4				4.7	
AF- 3	Mount Mica, Maine	Dept. Geol. U of M	41.5.5.1-1				5.3	
AF- 4	Port Darwin, Australia	Dept. Geol. U of M	41.5.5.1-5				5.0	
AF- 5	Port Darwin, Australia	Dept. Geol. U of M	41.5.5.1-3				2.2	
AF- 6	Bikita, Rhodesia	Prof. E. Wm. Heinrich					6.0	
AF- 7	Hugo pegm., Black Hills, S. Dakota	Prof. E. Wm. Heinrich					4.0	
AF- 8	Meyer's Quarry pegm., Eight Mile Park, Fremont Co., Colorado	Prof. E. Wm. Heinrich					5.6- 4.6	
AF- 9	Strickland Quarry, Portland, Conn., U.S.A.	ROM*, Toronto	M 21312				5.1	
AF-10	Tin Mountain Mine, Custer, S. Dakota	ROM, Toronto	M 24009				5.0	
AF-11	Varuträsk, Sweden	ROM, Toronto	M 19587				4.8	
AF-12	New Ross, Lake Ramsay, N.S.	ROM, Toronto	E 3307					
AF-13	Mt. Mica, Paris, Maine, U.S.A.	ROM, Toronto	M 6746				4.7	
AF-14	Dobrá Voda, Moravia, CSR	ROM, Toronto	M 26992				1.6	
AF-15	Peru, Maine, U.S.A.	ROM, Toronto	M 6744				5.2	

*ROM - Royal Ontario Museum

Appendix II - continued

No.	Locality	Source	Source No.	DTA UCD	Sp.gr.	F% wt. estim.	F% wt. anal.
AF-16	Coolgardie, W. Australia	ROM, Toronto	M 14139			6.1	
AF-17	China	ROM, Toronto	M 26733			3.2	
AF-18	Canon City, Colorado, U.S.A.	ROM, Toronto	M 23420			5.6	
AF-20	Vernéřov near Aš, ČSR	ROM, Toronto	M 25929			4.2	
AF-21	China	ROM, Toronto	M 12963			6.2	
AF-23	Chursdorf, Saxony	Dept. of Mineralogy Charles Univ., Praha	10824			11.0	
AF-24	Přibyslavice, ČSR	Dr. F. Čech				6.1	
AF-25	Kynžvart, ČSR	Dr. F. Čech				2.4	
AF-26	Vernéřov, ČSR	Dr. F. Čech	Spec. A			5.3	
AF-27	Varuträsk, Sweden	Dr. F. Čech				3.1	
AF-28	Ubini, W. Australia	Dept. of Mineralogy Charles Univ., Praha	9816-Ch. U.			5.0	
AF-29	Vernéřov, ČSR	Dr. F. Čech	Spec. B			5.8	
AF-30	Vernéřov, ČSR	Dr. F. Čech	Spec. C			5.3	
AF-31	Rožná, ČSR	Dr. F. Čech				7.5	
AF-32	Jeclov, ČSR	Dr. F. Čech				5.5	
AF-33	Nová Ves, ČSR	Dr. F. Čech				2.2	
AF-34	Bikita, S. Rhodesia	Dr. F. Čech				3.5	

Appendix II - continued

No.	Locality	Source	Source No.	DTA	UCD	Sp.gr.	F% wt. estim.	F% wt. anal.
AF-35	Peru.Maine,U.S.A.	Dr.H.Winchell	2023-3				5.2	
AF-36	Greenwood,Maine,U.S.A.	Dr.H.Winchell	3025-3				6.8	
AF-37	Dobrá Voda,ČSR	Dr.J.Staněk	1				6.3	
AF-38	Dobrá Voda,ČSR	Dr.J.Staněk	2				4.6	
AF-39	Dobrá Voda,ČSR	Dr.J.Staněk	3				6.0	
AF-40	Etta Lithium Mine,Keystone,S.Dakota	U.S.N.M.,Washington	93313				4.4	
AF-41	Hill City,S.Dakota,U.S.A.	U.S.N.M.,Washington	5327				4.8	
AF-42	Tinton,S.Dakota,U.S.A.	U.S.N.M.,Washington	102855				2.6	
AF-43	Nesbitt Spodumene Dike,Gunnison County, Colorado,U.S.A.	U.S.N.M.,Washington	117775	x	x	x		1.88
AF-44	Moguk,Upper Burma	U.S.N.M.,Washington	10734				1.8	
AF-46	Karibib,South Africa	U.S.N.M.,Washington	105914				7.6	
AF-47	Plumbago,Newry,Maine,U.S.A.	U.S.N.M.,Washington	5841		x	x	1.5	
AF-48	Newry,Maine,U.S.A.	U.S.N.M.,Washington	5906		x	x	1.4	
AF-49	Fremont County,Colorado,U.S.A.	U.S.N.M.,Washington	114040				6.1	
AF-50	Coolgardie,West Australia	U.S.N.M.,Washington	10432				7.1	
AF-51	Mt.Mica,Paris,Maine,U.S.A.	U.S.N.M.,Washington	20234				5.3	
AF-52	Mt.Mica,Paris,Maine,U.S.A.	U.S.N.M.,Washington	13736				4.5	
AF-53	Maine,U.S.A.	U.S.N.M.,Washington	96516				1.6	

U.S.N.M. - United States National Museum

Appendix II - continued

No.	Locality	Source	Source No.	DTA	UCD	Sp.gr.	F% wt. estim.	F% wt. anal.
AF-54	Hugo Mine, Black Hills, S. Dakota, U.S.A.	U.S.N.M., Washington	117837				3.0	
AF-55	Hebron, Oxford County, Maine, U.S.A.	U.S.N.M., Washington	62576	x	x	x		10.17
AF-56	Emmons Quarry, Greenwood, Maine, U.S.A.	U.S.N.M., Washington	R 16729				5.7	
AF-57	Buranga, Rwanda	U.S.N.M., Washington	117129				5.0	
AF-58	Lapinha Mine, Brazil	U.S.N.M., Washington	104855				6.2	
AF-59	W.A. Ware farm, N. Carolina, U.S.A.	U.S.N.M., Washington	103591				1.6	
AF-60	Pointe Du Bois, Manitoba, Canada	U.S.N.M., Washington	96000				2.1	
AF-61	Canon City, Colorado, U.S.A.	U.S.N.M., Washington	C 4163				2.4	
AF-62	Pala, California, U.S.A.	U.S.N.M., Washington	93863				4.6	
AF-63	Viitaniemi, Eräjärvi, Finland (I)	U.S.N.M., Washington	11816				4.7	
AF-64	Viitaniemi, Eräjärvi, Finland (II)	U.S.N.M., Washington	11816				5.2	
AF-65	Chursdorf, Saxony, Germany	Dr. P.G. Embrey	BM 14274 (Z in Moss et al. 1969)		x	x		11.8

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