

MINERALOGY OF THE WOLLASTONITES FOUND IN FINLAND ¹

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ABSTRACT

The physical and chemical properties and the powder X-ray data of the wollastonites in calcitic limestones of Finland are presented. The wollastonites investigated are very pure calcium metasilicates. Their optical properties and X-ray diffraction data indicate a triclinic structural symmetry.

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INTRODUCTION

According to present-day views, there are three modifications of calcium metasilicate:

pseudowollastonite		α -CaSiO ₃
parawollastonite	monoclinic	} β -CaSiO ₃
wollastonite	triclinic	

Pseudowollastonite is an artificial mineral representing the stable form of calcium metasilicate above 1180°C. In Nature, calcium metasilicate occurs in two modifications. Parawollastonite belongs to the monoclinic system, and wollastonite belongs to the triclinic system. The properties

¹ Received November 20, 1952.

of parawollastonite and wollastonite are closely related and, according to Peacock (1935 a, p. 528), »wollastonite is distinguished from parawollastonite by a completely different asymmetric series of terminal forms, a perceptible inclination of the optic axial plane to the plane normal to the b-axis, and an asymmetric system of X-ray interference spots». Parawollastonite is very rare and has been found in limestone blocks ejected from volcanoes. The normal triclinic wollastonite is characteristic of large bodies of contact metamorphic limestones and is the type of the triclinic minerals of the wollastonite group listed below (see Peacock, 1935 a, b):

wollastonite	$6 [\text{CaO} \cdot \text{SiO}_2]$
bustamite	$6 [(\text{Ca}, \text{Mn})\text{O} \cdot \text{SiO}_2]$
vogtite	$6 [(\text{Ca}, \text{Fe}, \text{Mn}, \text{Mg})\text{O} \cdot \text{SiO}_2]$
pectolite	$\text{H}_2\text{O} \cdot \text{Na}_2\text{O} \cdot 4\text{CaO} \cdot 6\text{SiO}_2$
schizolite	$\text{H}_2\text{O} \cdot \text{Na}_2\text{O} \cdot 4(\text{Ca}, \text{Mn})\text{O} \cdot 6\text{SiO}_2$

Wollastonite is a common mineral in many Archean metamorphic calcitic limestones in southern Finland. A great number of these localities have been mentioned by Eskola, Hackman, Laitakari, and Wilkman (1919). The physical and chemical properties of the wollastonites in Finland are, however, inadequately known, and no study on their symmetry has been made. The present author selected ten wollastonite specimens for detailed examination. Their physical and chemical properties and X-ray data will be briefly described.

NOTES ON THE SPECIMENS

The wollastonite in all the specimens examined occurred in calcitic limestones. A preliminary investigation of the specimens in the collections of the Institute of Geology, University of Helsinki, already showed that Finnish wollastonites are extremely pure calcium metasilicates and that no other members of the wollastonite group occur. The wollastonites selected for detailed examination are listed in Table 2.

In the specimens investigated wollastonite usually occurs as tablets or needles varying from a few mm up to 10 cm in length. Equidimensional grains, 1—3 mm in diameter, occur only in the sample from Piukkala, Parainen. The crystal faces (100), (001), (101), and $(\bar{1}02)$ in the [ac] zone are sometimes well-developed, but terminal faces important for the determination of the monoclinic or triclinic symmetry by the morphological data do not occur. The cleavages parallel to (100), (001), and $(\bar{1}02)$ are predominant, and the axis of the cleavage zone (b-axis) is commonly parallel to the elongation of the wollastonite crystals (see p. 13).

The color of the studied wollastonites is white, but some specimens show, especially on the weathered surface, a slight greyish, pinkish, or brownish tinge.

The wollastonites of the calcitic limestones are often associated with other calcium silicates. In the limestones of the Parainen area, the wollastonite paragenesis contains, according to Laitakari (1921), for example, the following lime-bearing silicates: diopside, grossularite, vesuvianite, and scapolite. The white-colored diopside has been found to be the most common associated silicate mineral in the specimens investigated. The refractive indices of the diopside in some specimens are as follows:

	α	β	γ
Tytyri, Lohja	1.675	1.682	1.703
Remonmäki, Pusula	1.666	1.675	1.695
Martinkylä, Sipoo	1.691	1.699	1.719

The values obtained show that the diopside is usually an extremely pure Ca-Mg-silicate.

In addition to diopside small amounts of zoisite and garnet occur in the wollastonite specimen from Tytyri, Lohja. The zoisite is a pink thuleite and forms small spots in the crystalline limestone. The zoisite fraction was separated with centrifuging in Clerici solution and was analyzed by Mr. H. B. Wiik, M. A. The chemical composition and optical properties (Table 1) correspond very well to those of pure zoisite. The white garnet occurs only

Table 1. Chemical composition of zoisite

Constituent	1		2
	%	Mol. prop.	%
SiO ₂	39.20	.6527	39.67
TiO ₂	0.08	.0010	
Al ₂ O ₃	32.01	.3140	33.66
Fe ₂ O ₃	0.76	.0048	
FeO	0.54	.0075	
MnO	0.05	.0007	
MgO	0.20	.0050	
CaO	25.68	.4579	24.69
H ₂ O+	2.03	.1127	1.98
H ₂ O—	0.00		
Total	100.55		100.00
α	1.694 $\pm$ 0.002		
β	1.696 $\pm$ 0.002		
γ	1.702 $\pm$ 0.002		

1. Zoisite. Tytyri, Lohja.

2. Theoretical composition of zoisite, Ca₂Al₂(OH)Si₃O₁₂.

very sparsely in the wollastonite specimen from Tytyri, and it was not possible to separate enough of a pure fraction for chemical analysis. It was expected that the garnet closely associated with the zoisite would be a hydrogarnet, because, according to Yoder (1950), dry grossularite cannot exist in the presence of water at elevated temperatures. The refractive index ($n = 1.736$) and the unit cell size ($a_0 = 11.85 \text{ \AA}$, calculated from the X-ray diffraction data) of the garnet are, however, values typical of grossularite containing no hydroxyl groups. The refractive index of the hydrogarnet is much lower than that of the dry grossularite, and the unit cell of the hydrogarnet is greater than that of the grossularite (see Yoder, 1950).

The wollastonite from all specimens mentioned in Table 2 was separated with centrifuging in Clerici solution. The purity of the material used for determination of physical and chemical properties was tested under the microscope by means of an immersion liquid with approximately the same refractive index as that of the mineral itself. The amount of impurities in all the separated wollastonite fractions was less than 1 %.

OPTICAL PROPERTIES

The refractive indices, axial angles, and extinction angles β/b of wollastonites are presented in Table 2.

Table 2. Optical properties of the wollastonites from Finland

Locality	α	β	γ	$\gamma - \alpha$	$2V_\alpha$	β/b
Pettiby, Parainen	1.621	1.632	1.634	0.013	38°	6° ± 1°
Piukkala, Parainen	1.620	1.632	1.634	0.014	40°	3° ± 1°
Skräbböle, Parainen	1.619	1.631	1.633	0.014	39°	5° ± 1°
Storgård, Parainen	1.620	1.632	1.634	0.014	39°	2° ± 1°
Lammala, Vestanfjärd	1.618	1.630	1.632	0.014	38°	1° ± 1°
Tytyri, Lohja	1.620	1.632	1.634	0.014	39°	4° ± 1°
Remonmäki, Pusula	1.619	1.630	1.632	0.013	40°	0° ± 1°
Martinkylä, Sipoo	1.618	1.631	1.633	0.015	40°	0° ± 1°
Sääksjärvi, Iitti	1.619	1.632	1.634	0.015	39°	4° ± 1°
Ihalainen, Lappeenranta	1.620	1.632	1.634	0.014	39°	5° ± 1°

The refractive indices were determined by means of the immersion method in sodium light at 20°C. The error is ± 0.001 . No variations indicating notable differences in the chemical composition were observed. The refractive indices of Finnish wollastonites vary as follows:

$$\alpha = 1.618 - 1.621$$

$$\beta = 1.630 - 1.632$$

$$\gamma = 1.632 - 1.634$$

The axial angle was measured on the universal stage. It varies only from 38° to 40° .

It was commonly believed before the X-ray studies that the wollastonite is monoclinic, with the plane of optic axes normal to the axis of the cleavage zone. The triclinic structural symmetry of the wollastonite was emphasized for the first time in the X-ray studies by Warren and Biscoe (1931), and soon afterward the small deviations from the monoclinic optics were also observed in some wollastonites. Peacock (1935 a) pointed out, on the basis of careful morphological studies, that there exist both a monoclinic and a triclinic modification of wollastonite. The optical properties of the two modifications were very similar, but »the optic axial plane was found to be strictly normal to the axis of the cleavage zone in monoclinic wollastonite, in triclinic wollastonite this plane was found to be inclined to the axis of the cleavage zone at 4° ». The inclination of the optic axial plane is, so far known, the only optical method to distinguish the triclinic wollastonite from the monoclinic parawollastonite. Many determinations of the extinction angle β/b in Finnish wollastonites were made on the universal stage, turning the wollastonite plates or needles around the horizontal b-axis. The results of these determinations are presented in Table 2. Most of the studied wollastonites are triclinic, with a distinct inclination of the optic axial plane. In some instances, however, the inclination is so small that it is not possible to decide by this method with certainty the symmetry of the wollastonite. The monoclinic optical properties found in the wollastonites from Pusula and Sipoo are probably caused by a very fine polysynthetic twinning of the triclinic modification, because the structural data of these wollastonites (see p. 14) agrees completely with those of the distinctly triclinic modification.

The elongation of the wollastonite is commonly parallel to the b-axis, and the sign of elongation varies when the needle-shaped wollastonite crystal is turned around the axis of the cleavage zone. Positive elongation, however, occurs in all sections of the wollastonite from Storgård, Parainen. This peculiar deviation from the common rule is caused by the fact that the elongation is parallel to the cleavage plane $10\bar{2}$ as also Winchell (1951) found in the wollastonite from Bingham, Utah.

CHEMICAL COMPOSITION

Three new chemical analyses of Finnish wollastonites were made by Mr. H. B. Wiik, M. A., at the request of the present author. Material centrifuged in Clerici solution was used for the analysis. The amount of impurities in the analyzed specimens was found to be less than 0.5 %. The specific gravity was determined by the pycnometer at 20°C .

Table 3. Chemical composition of Finnish wollastonites

Constituent	1		2		3	
	%	Mol. prop.	%	Mol. prop.	%	Mol. prop.
SiO ₂	50.82	.8462	50.74	.8448	50.76	.8452
FeO.....	0.18	.0025	0.36	.0050	0.72	.0100
MnO.....	0.03	.0004	0.03	.0004	0.08	.0011
MgO.....	0.22	.0055	0.50	.0124	0.14	.0035
CaO.....	48.16	.8588	47.92	.8545	48.72	.8688
Na ₂ O.....	0.12	.0019	0.00	—	n. d.	—
K ₂ O.....	0.07	.0007	0.00	—	n. d.	—
H ₂ O+.....	0.08	.0044	0.08	.0044	0.00	—
H ₂ O—.....	0.00	—	0.00	—	0.02	—
Total.....	99.68		99.63		100.44	
Sp. gr.	2.91		2.91		2.92	

1. Wollastonite. Remonmäki, Pusula.

2. Wollastonite. Sääksjärvi, Iitti.

3. Wollastonite. Storgård, Parainen.

The chemical composition of the analyzed wollastonites (Table 3) agrees very well with the theoretical composition of calcium metasilicate. The content of FeO, MnO, and MgO is always less than 1 %. The greatest variation occurs in the content of FeO, but this variation is not sufficiently strong to cause observable differences in the physical properties. The low alkali and water content in the wollastonite from Pusula suggests the presence of the pectolite component.

X-RAY DIFFRACTION DATA

An attempt to resolve the structural symmetry of Finnish wollastonites by means of the powder X-ray diffraction data was made. The powder X-ray diagrams were taken by means of a Geiger Counter Recording X-Ray Spectrometer (Norelco) of the North American Philips Co., Inc., with CuK-radiation and Ni-filter. The X-ray spectrometer was calibrated with a quartz standard, and it will reproduce angles (2θ) measurable to 0.025° .

The powder patterns of the wollastonites in Finland (Table 4) agree completely with one another, thus suggesting the same group of structure symmetry for all studied specimens. It may be emphasized that the diffraction patterns of the wollastonites from Pusula and Sipoo having monoclinic optical properties (see Table 2) are rather similar to those of the optically triclinic wollastonites.

Table 4. Powder diffraction data for wollastonites from Finland (1) and for triclinic wollastonite from Crestmore (2)

1		2		1		2	
d	I	d	I	d	I	d	I
3.850	6	3.82	vw	2.210	$\frac{1}{2}$	—	—
3.524	6	3.50	w	2.185	$\frac{2}{2}$	2.18	vw
3.440 n. o.	$\frac{1}{2}$	—	—	2.165	1	—	—
3.324	8	3.29	m	2.093 n. o.	$\frac{1}{2}$	—	—
3.245 n. o.	$\frac{1}{2}$	—	—	2.023	1	2.01	vvw
3.200 n. o.	$\frac{1}{2}$	3.19	vvw	1.984	1	1.972	vvw
3.132 n. o.	$\frac{1}{2}$	—	—	1.920	$1\frac{1}{2}$	—	—
3.090	5	3.09	w	1.854 n. o.	$\frac{1}{2}$	—	—
2.985	10	2.97	s	1.835	$1\frac{1}{2}$	1.823	m
2.923 n. o.	$\frac{1}{2}$	—	—	1.809 n. o.	$\frac{1}{2}$	1.788	vvw
2.815 n. o.	$\frac{1}{2}$	—	—	1.759	$\frac{2}{2}$	1.750	vw
2.731	$2\frac{1}{2}$	—	—	1.719	$\frac{2}{2}$	1.711	vw
2.561	$\frac{2}{2}$	—	—	1.607 n. o.	$\frac{1}{2}$	1.596	vw
2.481	3	2.46	vw	1.532	1	1.526	vvw
2.422 n. o.	$\frac{1}{2}$	—	—	1.516 n. o.	$\frac{1}{2}$	—	—
2.349	3	2.34	vvw	1.471 n. o.	$\frac{1}{2}$	1.471	vvw
2.337	2	—	—	1.457	$\frac{2}{2}$	1.449	vvw
2.304	4	2.30	vvw	1.360	2	1.353	vvw
2.297	1	—	—				

Explanation of symbols: d, interplanar spacings in kX units; I, intensity; s, strong; m, medium; w, weak; v, very; n. o., not observed in all specimens. $I = I(hkl)/I_{10}$.

The diffraction patterns of the wollastonites from Finland are closely related to those obtained by Clark (1946) for the wollastonite from Crestmore (see Table 4), whose triclinic symmetry was demonstrated by Peacock (1935a). Furthermore, the diffraction patterns of Finnish wollastonites are related to the pattern of pectolite. According to Peacock (1935b), pectolite is triclinic and belongs to the group of minerals whose type is triclinic wollastonite.

The diffraction data suggests a triclinic structure symmetry for the wollastonites in Finland. An accurate check of the triclinic symmetry may be obtained, if it is possible to observe marked differences between the diffraction patterns of the wollastonites studied and of monoclinic parawollastonite.

Unfortunately, the present author had no material for producing a powder X-ray diagram of the monoclinic parawollastonite. Furthermore, the d-spacings of the accurately monoclinic wollastonite have not been published¹. On the basis of the structural data for the monoclinic wollastonite ($a_0 = 15.33\text{\AA}$; $b_0 = 7.28\text{\AA}$; $c_0 = 7.07\text{\AA}$; $\beta = 95^\circ 25'$; space group C_{2h}^5) given by Barnick (1935), the d-spacings for h0l's were calculated from the formula

¹ O'Daniel and Tscheischwili (1948) published the diffraction data for a natural wollastonite supposed to be monoclinic belonging to the space group C_{2h}^4 . No accurate check relative to the monoclinic structure symmetry of this wollastonite is available, and its diffraction pattern agrees almost completely with the patterns of triclinic wollastonites presented in Table 4.

$$d_{(hkl)} = \frac{1}{\sqrt{\frac{h^2}{a^2} + \frac{l^2}{c^2} - \frac{2hl \cos \beta}{ac} + \frac{k^2}{b^2} \sin^2 \beta}}$$

All calculated d 's from $d = 4.000$ kX to $d = 2.000$ kX are seen in Fig. 1, where the d -spacings for $h0l$'s, permitted in the space group of the parawollastonites are marked by solid lines and the d -spacings not permitted are shown by lines of dashes. For comparison of structural data, the X-ray diagrams were produced from oriented plates and needles of Finnish wollastonites rotating around the b -axis with the X-ray beam normal to the axis of the cleavage zone. These d -spacings are also presented in Fig. 1.

The comparison of the d -spacings for the $[ac]$ zone (Fig. 1) shows that most of the interplanar spacings observed in Finnish wollastonites are also possible in the monoclinic parawollastonite, but the wollastonites from Finland give some diffraction lines not permitted in the monoclinic modification of wollastonite. The most «critical» d -spacings, which indicate for Finnish material, a symmetry different from that of parawollastonite, are:

$$d = 2.349 \text{ kX}; d = 2.337 \text{ kX}; d = 2.304 \text{ kX}; d = 2.297 \text{ kX}.$$

These d -spacings represent the surfaces $(\bar{1}03)$, (003) , (601) , $(\bar{2}03)$, and (103) , which do not give reflections in the space group C_{2h}^5 of the monoclinic wollastonite, but the d -spacings for these $h0l$'s are permitted in the triclinic structural symmetry.

The diffraction patterns of the monoclinic and triclinic modifications of the wollastonite seem to be closely related, as would be expected also on the basis of the similar physical properties. The structural differences are found in the reflections from the $[ac]$ zone, but it is probable that these same differences are observable also in the powder X-ray diagrams.

CONCLUSION

The physical and chemical properties show that the wollastonites in Finnish limestones are very uniform in all localities investigated. The observed small inclinations of the optic axial plane to the axis of the cleavage zone suggest a triclinic symmetry for most wollastonites, but in some instances the optical properties suggest a monoclinic symmetry. The powder X-ray diffraction patterns show, however, that the structural symmetry of all Finnish wollastonites agrees with that of the triclinic wollastonite from Crestmore. The triclinic structure symmetry of the wollastonites from Finland was checked also by the structural differences obtained in the

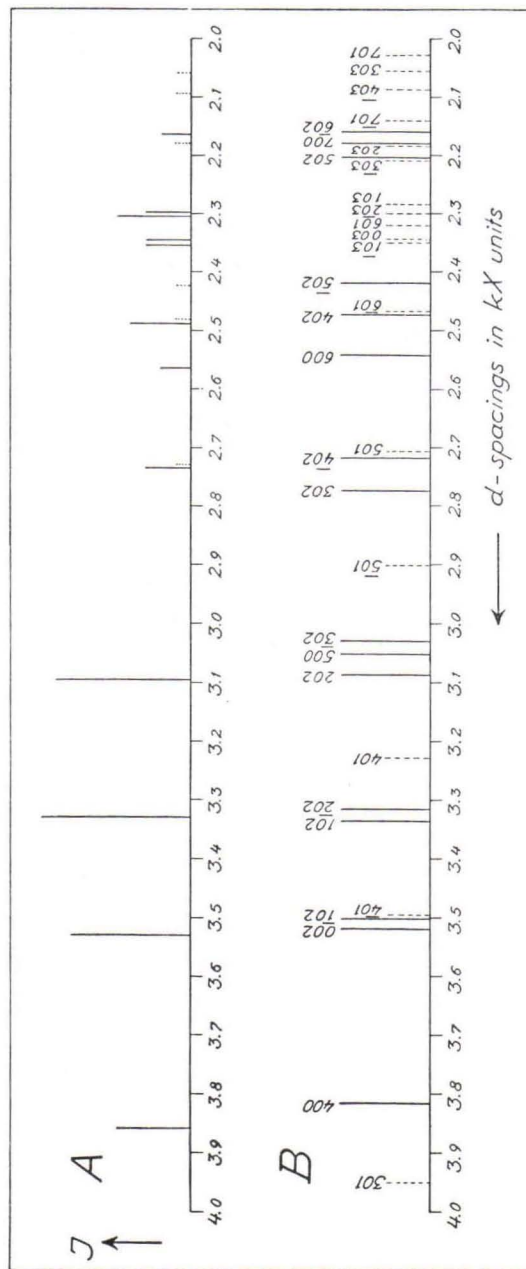


Fig. 1. Interplanar spacings for the [ac] zone. A, Finnish wollastonites; dotted lines, not observed in all specimens. B, Monoclinic parawollastonite (calculated); lines of dashes, d-spacings not permitted in the space group C_{2h}^2 .

diffraction patterns of the natural wollastonite modifications. It appears from this investigation that the apparently monoclinic optical properties of some wollastonites are caused by a very fine polysynthetic twinning of the triclinic modification.

Ito (1950), on the basis of structural data, regarded also monoclinic parawollastonite as a twinned modification of triclinic wollastonite. Applying his theory of twinned lattices, Ito presented many alternative ways of combining two unit cells of triclinic wollastonite to obtain the monoclinic unit cell, and he suggested that both wollastonite types probably might be derived from another monoclinic wollastonite type, called protowollastonite. This type, however, has not been found in Nature.

Acknowledgments — In the course of this work the facilities of the Geochemical Laboratory at the Institute of Geology, University of Helsinki, were placed at author's disposal through the kindness of Professor Th. G. Sahama. Advice in the X-ray diffraction technique was obtained from Dr. K. J. Neuvonen. The chemical analyses were made by Mr. H. B. Wiik, and Mrs. Toini Mikkola assisted in the optical determinations. The author wishes to express his sincere gratitude to all these persons.

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