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Optical Orientation and Twinning of Merwinite: A Restudy

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Abstract

Study of merwinite from the new locality of Killala, Ireland, and the type locality of Crestmore, California, shows that earlier reports on its optical orientation and twinning are erroneous. Merwinite is monoclinic, the new optical orientation is $Y = b$, $X{:}c = 13°$, O.A.P. is (010). The cleavages are : {100} poor and {010} very poor. Merwinite shows polysynthetic, parallel twinning on {100}, with twin axis $[0\bar{1}3]$; and on (611) and $(6\bar{1}1)$, with twin axes $[0\bar{1}3]$ and [013] respectively. The twin axes are inclined to the c axis at approximately 31° and to all the vibration directions of the host merwinite; therefore corresponding vibration directions of the twins bear a non-linear, angular relationship with those of the host. The chemical composition and X-ray data for the Killala merwinite compare favorably with the recent data on synthetic merwinite.

Introduction

Merwinite was first discovered by Larsen and Foshag (1921) from Crestmore, California, and was described as probably monoclinic, optical orientation, $Z = b$, $X{:}c = 36°$. The cleavage was stated to be perfect parallel to {010}, and the twinning was described as polysynthetic along {110} with c as the twinning axis, and along {100} twins with twinning axis unstated, but presumed to be the c axis. Tilley (1929) described merwinite from Scawt Hill as tabular on {010}, with optical orientation, cleavage, and twinning as for the Crestmore material. Yamaguchi and Suzuki (1967) determined the structure of synthetic merwinite crystals which were reported to be without cleavage or twinning but platy on {001}, which by Moore and Araki's (1972) orientation is {100}. They cite its optical orientation as $Y = b$ (in the plane of the plate) and X making a small angle with the a axis in this plane. Moore and Araki (1972) determined the crystal structure of synthetic merwinite and found that the tabular structure on {100} could not be reconciled with the tabular development and cleavage on {010} reported in the earlier literature. Consequently, they examined the Crestmore merwinite and found cleavage along {100} in accordance with the structure; they concluded that "misorientation in the earlier literature may have resulted from the near-orthogonal character of the crystal cell." Both these later studies imply misorientation of the optical indicatrix in the earlier literature, but the differences are not pointed out.

During the present study, the misorientation was detected when optical orientation and twinning of the Killala merwinite were examined with a universal stage. In sections normal to the Y vibration direction of the host, the traces of the alleged {110} twin lamellae were inclined to the traces of the poor cleavage and the X vibration direction was inclined to the traces of both the {100} twin lamellae and the cleavage. This indicated that the optical orientation and twinning of the mineral under study differed from those reported by Larsen and Foshag (1921) and Tilley (1929). The same conclusion was reached when a specimen of the Crestmore merwinite was re-examined.

The twinning described in this paper differs from that reported by Larsen and Foshag (1921) and Tilley (1929). In order to complete the description, the paper also includes X-ray and microprobe data on Killala merwinite.

Physical Properties

Cleavage

In thin section, the Killala merwinite grains have a rounded, hexagonal, or rectangular outline. The cleavage, earlier stated as perfect parallel to {010} (Larsen and Foshag, 1921; Tilley, 1929), is on {100}, in agreement with Moore and Araki (1972). It is poor and is never well developed but becomes more obvious in very thin sections where the grains show signs of disintegration. As described below, the O.A.P. is perpendicular to the plane of this cleavage, which makes a small angle with *X*. The platiness of Yamaguchi and Suzuki's (1967) synthetic crystals is probably also due to the potential {100} cleavage, which by their orientation is {001}. More rarely, the Killala merwinite shows a second very poor cleavage or parting parallel to {010}.

Specific Gravity

During heavy liquid separation of the Killala merwinite it became obvious that it is denser than first reported by Larsen and Foshag. Its specific gravity of 3.320, determined by means of a density column, is in agreement with the specific gravity of synthetic merwinite, reported by Yamaguchi and Suzuki (1967) and Moore and Araki (1972), and of the Crestmore merwinite reported by Neuvonen (1952).

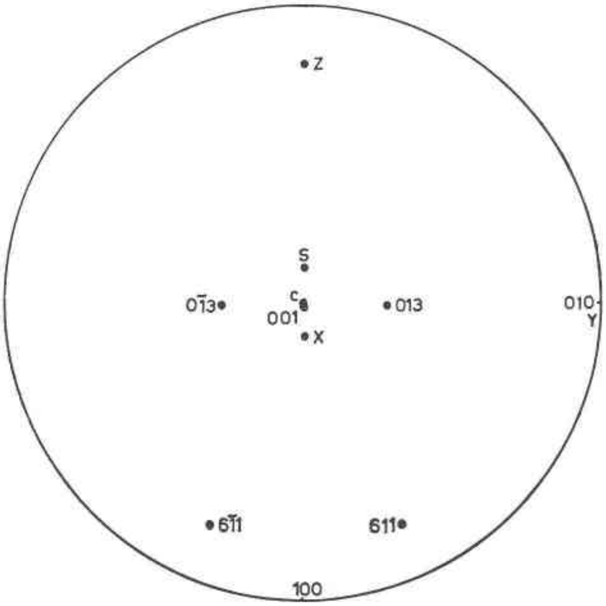

FIG. 1. The stereographic projection of the optical orientation and twinning of merwinite. Two of the twin planes, (611) and ($6\bar{1}1$), intersect at the point **S**, and their respective twin axes are [$0\bar{1}3$] and [013]. The third twin plane and cleavage on {100} are not shown. The angular separation between the *c* axis and [001] is nearly 2°.

Optical Orientation

The Killala merwinite shows intricate polysynthetic twinning, high relief, and low to medium birefringence. Its refractive index values are α 1.702, β 1.710, γ 1.723 (all ± 0.002), $2V_Z = 70 \pm 2°$. Its O.A.P. is parallel to (010), $Y = b$ and $X{:}c = 13°$. This optical orientation differs from that reported by Larsen and Foshag (1921) and by Tilley (1929) but is qualitatively similar to that for synthetic merwinite, reported by Yamaguchi and Suzuki (1967), if its *a* and *c* crystal-axes are interchanged.

Moore and Araki (1972) recognized the crystallographic misorientation in the earlier literature (as noted above), but did not comment on the optical orientation.

Twinning

The Killala merwinite contains three sets of twin lamellae. Two of the three sets, equivalent to the {110} twin lamellae reported by Larsen and Foshag, are well-developed and intersect at approximately 42°; the third, poorly-developed set, corresponding to their {100} twins, nearly bisects the acute angle between the first two sets and makes a very small angle with the poor {100} cleavage. The twinning axes neither coincide with the *c* axis nor with the intersection of the first two sets of twin lamellae, but are inclined to both of them (see below). The O.A.P. of the host bisects the obtuse angle between the first two sets of twin lamellae.

The exact location of the *c* axis is difficult to determine because both the {100} and {010} cleavages in merwinite are poorly-developed and consequently the measured angle, $X{:}c$, varies between 7° and 13°. The angle between *X* and the above intersection is $\sim 26°$; therefore the angle between the intersection and the *c* axis is $\sim 13°$ (using the highest value of $X{:}c = 13°$).

Plotting the results on a standard stereogram, based on Moore and Araki's crystallographic orientation, shows the first two sets of twin planes to be (611) and ($6\bar{1}1$) and their twin axes as [$0\bar{1}3$] and [013] (Fig. 1). From two observations, the composition plane of the third set appears to be practically parallel to {100} and the twin axis lies close to [$0\bar{1}3$]. Figure 1 shows that the twin axes make an angle of about 31° with the (010) plane and are inclined to *X*, to the *c* axis, and to the intersection of the first two sets of twin lamellae.

Because of the difficulty in accurately locating the *c* axis, the Miller indices of the twin planes and their twin axes are approximate. The twinning, however, is definitely not prismatic because the twin lamellae are inclined to the traces of the poor cleavage. Furthermore, the prismatic twinning would require the three entities (the intersection of the first two sets of twin lamellae, the twin axes, and the *c* axis) to be coincident and to make an angle of 36° with *X* (Larsen and Foshag's orientation). On the contrary, all the four entities show angular separation that is far larger than any errors of observation or measurement.

A discussion of the origin of the merwinite twinning must await a future thermal/X-ray study aimed at determining possible polymorphic changes in merwinite. Moore (1973) has suggested the possibility of a high-temperature merwinite polymorph with glaserite (hexagonal) structure and has demonstrated the correspondence between the pseudo-hexagonal *a* axis of merwinite and the *c* axis of glaserite. If polymorphism in merwinite does exist, then the twinning most likely results from inversion, just as Ca_2SiO_4, in the cement clinker, shows twinning due to inversion from α to α' to β forms (Ono, Kawamura, and Soda, 1968; Yamaguchi and Takagi, 1968).

Chemical Composition

The composition of the Killala merwinite was determined by electron probe microanalysis using a synthetic pyroxene standard. The total iron was calculated as Fe_2O_3. The analysis in weight percent is: SiO_2 34.93, Al_2O_3 0.13, Fe_2O_3 0.39, MgO 12.56, CaO 50.65.

The total (98.66) is low because the analysis is uncorrected and excludes minor constituents. Recalculated on the basis of 8 oxygens, the analysis leads to the formula, $Ca_{3.03}Mg_{1.04}Si_{1.96}O_8$, for the Killala merwinite.

X-ray Data

The X-ray powder pattern of the Killala merwinite, obtained on a diffractometer using nickel-filtered copper radiation, and Moore and Araki's (1972) pattern of synthetic merwinite (Table 2) are substantially similar to that of synthetic merwinite reported by Yamaguchi and Suzuki (1967). Moore and Araki's pattern was used for indexing the Killala pattern, which differs from it in some minor details. For example, Moore and Araki index their *d*-spacing at 2.672 Å as belonging both to $\bar{4}11$ and *013* reflections for which their calculated *d*-spacings are 2.709 Å and 2.680 Å. In the Killala pattern they are resolved; the $\bar{4}11$ reflection is a weak one and perhaps was not observed by them because of its proximity to the very intense *013* reflection. Similarly, the unresolved line at 2.273 Å (Moore and Araki) belonging to two reflections, *511* and *122*, is resolved in the Killala pattern into two lines. Corresponding to the line at 2.213 Å, which was indexed as $\bar{2}04$ by Moore and Araki, the Killala pattern contains two well-resolved lines; one of these is indexed as $\bar{2}04$ after Moore and Araki and the other as (600). The latter follows from Yamaguchi and Suzuki's (*h0l*) reciprocal net in which the reflections $\bar{2}04$ and *600* are of comparable intensity. This indexing is consistent with the space group and does not violate Moore and Araki's indexing because the calculated *d*-spacings for these reflections, 2.222 Å and 2.208 Å, bracket their observed line at 2.213 Å.

The sample used for the Killala pattern contained monticellite and spinel impurities, and the former acted as the internal standard. Because of the interference from the impurities and the uncertainties involved in accurately measuring weaker peaks, only lines that were sharp, sufficiently strong, free from impurity influence, and unambiguously indexed, were selected for calculating the unit cell dimensions. The 2θ values of the six peaks selected on this basis were measured with an accuracy of ±0.01°. The method of Smith (1956), modified for the monoclinic system, was used to construct six normal equations which were solved by Gauss-Jordan's direct numerical method, given in books on numerical analysis.

Although the input parameters were few, for the reasons already stated, the agreement between the observed and the calculated *d*-spacings is good. It is not fortuitous; rather, it testifies to the correct choice of the peaks for the unit cell refinement, and to the

TABLE 1. Unit Cell Dimensions of the Killala Merwinite

	Killala merwinite	Synthetic merwinite (1)	Synthetic merwinite (2)
a (Å)	13.283	13.254(21)	9.336
b (Å)	5.304	5.293(9)	5.301
c (Å)	9.317	9.328(17)	13.286
β	92.08°	91.90(15)°	92.13°

(1) *Moore and Araki (1972)*
(2) *Yamaguchi and Suzuki (1967)*

TABLE 2. X-ray Powder Diffraction Data for Merwinite

Synthetic merwinite†				Killala merwinite††		
I/I_o	d(Å) (obs)	d(Å) (calc)	hkl	I/I_o	d(Å) (obs)	d(Å) (calc)
20	9.27	9.32	001			
20	6.60	6.62	200	12	6.63	6.636
30	4.630	4.603	011	48	4.70**	4.608
			$\bar{1}11$	2	4.383	4.378
			111	3	4.326	4.329
20	3.865	3.873	$\bar{2}02$	11	3.878	3.878
20	3.740	3.751	211	1	3.742	3.747
30	3.310	3.312	400	24	3.315	3.318
30	3.128	3.160	311	10	3.164	3.163
20	3.032	3.062	212	23	3.032***	3.066
40	2.749	2.743	$\bar{4}02$	37	2.752*	2.750
100	2.672	2.709	$\bar{4}11$	15	2.715	2.716
		2.680	013	100	2.674*	2.679
30	2.650	2.646	020	47	2.652	2.652
			$\bar{2}13$	1	2.515	2.511
10	2.456	2.460	213	78	2.452**	2.457
30	2.326	2.321	$\bar{3}13$	21	2.321	2.324
30	2.311	2.313	$\bar{5}11$	17	2.318	2.318
20	2.273	2.280	511	13	2.282*	2.283
		2.261	122	6	2.262	2.263
40	2.213	2.222	$\bar{2}04$	11	2.227	2.222
			600	38	2.211*	2.212
30	2.169	2.176	204	13	2.175	2.172
			222	16	2.171	2.165
20	2.103	2.116	$\bar{1}14$	13	2.061	2.115
20	2.059	2.055	$\bar{3}22$	11	2.057	2.059
30	2.024	2.027	322	30	2.031**	2.029
		2.021	$\bar{6}02$		2.022	2.019
40	1.909	1.905	$\bar{4}22$	31	1.909*	1.913
30	1.875	1.877	404	26	1.874*	1.874
10	1.821	1.819	$\bar{4}14$			
10	1.757	1.752	$\bar{5}22$			
10	1.729	1.728	$\bar{6}13$			
30	1.636	1.656	800	17	1.659**	1.659

† *Moore and Araki, 1972*

†† *This study: calculated cell data a 13.283, b 5.304, c 9.317 (Å), β 92.08°*

* *Indicates peaks selected for cell refinement*

** *Indicates peaks influenced by spinel impurity*

*** *Indicates peak influenced by calcite impurity*

correct indexing by Moore and Araki of their powder pattern.

Moore and Araki have compiled an up-to-date list of reported unit-cell data on synthetic merwinite and have shown that all the previous data, with the exception of the Japanese data, are erroneous. The crystallographic orientation chosen by Moore and Araki (1972) is adopted for the purposes of this paper. The unit cell dimensions of the Killala merwinite (Table 1) are in substantial agreement with the results of both these studies. The differences may be attributed in part to the techniques used for calculating the unit cell dimensions from the powder data, and in part to ionic substitutions in the Killala merwinite.

Conclusions

Earlier data on the optical orientation and twinning of merwinite are incomplete or incorrect. The prominent cleavage in merwinite is on {100} and the O.A.P. is parallel to (010). The twinning is not on {110} but on {*hkl*} and the twinning axes are inclined to the *c* axis as well as to the principal vibration directions of merwinite. An electron probe microanalysis shows that the Killala merwinite composition is close to the theoretical formula $Ca_3Mg Si_2O_8$. The X-ray powder pattern and the unit cell dimensions of the Killala merwinite are similar to those of the synthetic material of Yamaguchi and Suzuki (1967) and Moore and Araki (1972).

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