

On the X-ray identification of amblygonite and montebrasite

A. A. MOSS, E. E. FEJER, AND P. G. EMBREY

Department of Mineralogy, British Museum (Natural History), London S.W. 7

SUMMARY. Previous investigations have been confined to individual members or to small compositional ranges of the amblygonite–montebrasite series. The present study is a survey based on twenty-two specimens in the range $Amb_{13}Mon_{86}$ to $Amb_{92}Mon_8$ (1.8–11.8 % fluorine), and indicates that the fluorine content may be related both qualitatively and quantitatively to differences in the X-ray powder patterns. Fully indexed powder data to $d = 1.68 \text{ \AA}$ and derived cell parameters are given for four members of the series. There is no evidence of a break in the continuity of the series. The first British occurrence of both amblygonite and montebrasite is reported from a pegmatite vein in aplite at Meldon, Okehampton, Devonshire.

IN the course of an examination of a number of specimens of presumed amblygonite and montebrasite collected by the late Mr. A. W. G. Kingsbury at the old aplite quarry, Meldon, Devonshire, we noted marked differences between the X-ray powder patterns. It was by no means clear from published data that all the specimens belonged to the series, and variations in powder patterns of different specimens of montebrasite (Quensel, 1962) were less than we observed. Other workers (Fisher, 1958; Kittrick and Hope, 1967) distinguish between amblygonite and montebrasite on the basis of spacing and intensity differences of the three strongest lines of the patterns, and on comparisons of single crystal diffraction patterns with powder patterns, but their reference samples are inadequately characterized and their data too scanty. The powder patterns figured by Pough and Henderson (1945) are indistinct, and are not supported by chemical analyses.

Twenty-two specimens of amblygonite and montebrasite were selected for apparent homogeneity, and powder prepared from each was used for partial chemical analysis (table I) and both photographic and diffractometer records of the powder patterns. Careful examination of the photographic powder patterns showed that it is possible to distinguish four general types of pattern, here designated A, AB, B, and C (fig. 1). Diffractometer records, mentioned below, permit division of the AB pattern-type into two (fig. 2).

For chemical analysis, between 15 and 70 mg of the powder were fused with five times their weight of sodium carbonate, extracted with 40 ml of 1:1 sulphuric acid, transferred to a distillation flask, and the fluorine steam-distilled. Fluorine was determined on the distillate by the zirconium–eriochrome-cyanine method of Megregian (1954). Alkalis were determined by flame photometry on a sulphuric acid solution.

Comparison of the chemical and X-ray results shows that there is a correlation between pattern-type and fluorine content (fig. 4). The replacement of lithia by soda

has no obvious effect on the pattern type; specimen S, although near the upper edge of the AB group, retains AB affinities as firmly as specimen R which has a much lower soda content. No specimen of true natromontebasite was studied. Fig. 4 also shows that there is a tendency for high-fluorine samples to contain more soda, but this may be a function of the geological environment rather than the crystal chemistry.

TABLE I. Localities, partial chemical analyses, and values of $2\theta_{131}(\text{Cu-K}\alpha)$. The specimens are lettered from E to Z, in order of increasing fluorine content. The values of $2\theta_{131}$ were obtained from diffractometer traces; 0.1° has been subtracted throughout to obtain better agreement with the photographic data of table II

Spec.	% F	% Na ₂ O	% K ₂ O	% Li ₂ O	$2\theta_{131}$	Spec.	% F	% Na ₂ O	% K ₂ O	% Li ₂ O	$2\theta_{131}$
E†	1.8	0.3	< 0.1	9.1	52.22°	P	4.2	tr.	tr.	9.8	52.45°
F	2.0	0.7	< 0.1	8.6	52.25	Q	4.5	1.2	tr.	9.6	52.56
G	2.2	0.1	tr.	9.9	52.32	R	5.1	0.6	tr.	8.8	52.45
H	2.3	0.2	< 0.1	9.1	52.20	S	5.2	3.2	0.6	6.9	52.40
I	2.7	0.1	< 0.1	9.6	52.25	T	5.4	0.2	0.3	9.7	52.47
J	2.7	< 0.1	tr.	9.8	52.45	U	5.5	< 0.1	tr.	10.0	52.60
K	3.2	1.7	< 0.1	9.3	52.45	V	6.4	1.7	< 0.1	9.0	52.65
L	3.2	0.5	0.5	9.5	52.35	W†	8.2	1.0	tr.	9.5	52.70
M	3.5	0.1	0.2	10.4	52.42	X	8.9	1.9	tr.	8.4	52.90
N†	3.9	0.1	tr.	9.4	52.43	Y	9.7	1.3	0.1	9.2	52.97
O	4.1	tr.	tr.	9.8	52.49	Z†	11.8	1.2	tr.	8.8	52.91

E (BM 45956), W (BM 44421), X (BM 1908,344): Montebas, Creuse, France.
 F (BM 1947,75): Tip Top mine, Custer, Custer Co., South Dakota, U.S.A.
 G (BM 1946,67): Varuträsk, Skelleftea, Västerbotten, Sweden.
 H (BM 87153): Carmelita mine, Caceres, Spain.
 I (BM 1929,1504): 12 miles E. of Pte du Bois and 1 mile S. of Winnipeg river, Manitoba, Canada.
 J (BM 1921,491): Amblygonite mine, Caceres, Spain.
 K (BM 1925,961): West Arm, King's Table, Northern Territory, Australia.
 L (BM 1961,341): Verněřov, nr. Aš, Bohemia, Czechoslovakia.
 M (BM 1963,221): Nevel mine, Newry, Oxford Co., Maine, U.S.A.

N (BM 50270), T (BM 46004): Mt. Mica, Paris, Maine, U.S.A.
 O (BM 1905,5): Pala, San Diego Co., California, U.S.A.
 P (BM 1961,2): Bikita mines, Southern Rhodesia.
 Q (BM 1929,95): Black Hills, South Dakota, U.S.A.
 R (BM 1929,147): Ubini, Coolgardie, Western Australia.
 S (BM 1912,116): Canon City, Fremont Co., Colorado, U.S.A.
 U (BM 1934,11): Greenwood, Oxford Co., Maine, U.S.A.
 V (BM 1926,508): Lady Don tin mine, Euriovie, New South Wales, Australia.
 Y (BM 96902), Z (BM 14274^x): Chursdorf, nr. Penig, Saxony, Germany.

† Specimens E, N, W, and Z were used for calculation of cell parameters and the indexed powder data (tables II and III).

Photographic methods, including Guinier records (fig. 1), failed to resolve satisfactorily a prominent group of powder diffraction lines between 26° and $29^\circ 2\theta$. (Cu-K α), but a Philips diffractometer gave good resolution at the following settings: Cu-K α radiation, 40 kV, 20 mA; scanning speed $1^\circ 2\theta/\text{min}$, chart speed 1600 mm/h (approx. $1''/1^\circ 2\theta$); ratemeter 200 counts/sec, time constant 1 sec; slits 1° , 0.1 mm, and 1° . The reflections were indexed (see below) by means of the calculated data in table II, and grouped into five pattern types broadly referable to the visual groupings already mentioned; visual pattern-type AB, with the largest number of samples

TABLE II. Indexed powder patterns, with observed and calculated spacings and observed intensities. I_{rot} intensities are very approximate, and were used to eliminate very weak or absent reflections. Spacings in italics are out of strict numerical sequence to retain indexed reflections in the same line

hkl	I_{rot}	Specimen E (patt. A)			Specimen N (patt. AB)			Specimen W (patt. B)			Specimen Z (patt. C)		
		d_c	d_o	I_o	d_c	d_o	I_o	d_c	d_o	I_o	d_c	d_o	I_o
010	w	6.20	6.19	w	6.18	6.19	vw	6.17	6.16	vw	6.15	6.12	vww
100	w	4.805	4.816	w	4.790	4.816	vw	4.776	4.776	w	4.747	4.758	w
110	s	4.688			4.673			4.668			4.657		110
001	s	4.656	4.659	vs	4.653	4.652	vs	4.646	4.640	vs	4.624	4.642	vs
011	m	4.604			4.608			4.635			4.645		011
111	w	3.853	3.850	mw	3.853	3.850	w	3.864	3.867	mw	3.870	3.866	mw
101	s	3.358			3.350			3.345			3.326		101
101	s	3.330	3.329	ms	3.325	3.327	ms	3.315	3.317	m	3.299	3.300	m
110	s	3.275	3.273	m	3.266	3.261	mw	3.255	3.246	ms	3.236	3.237	ms
121	s	3.216			3.216	3.218	m	3.235			3.252		121
011	s	3.208	3.211	vs	3.200	3.181?	m	3.183			3.161		011
120	s	3.165			3.156			3.154	3.167	vvs	3.151	3.151	vvsb
021	s	3.157	3.159	vvs	3.156			3.173			3.181		021
020	w	3.098			3.090	3.089	vww	3.084	3.081	vw	3.075	3.078	vw
111	s	2.966	2.967	vs	2.962	2.960	vs	2.964	2.964	s	2.953	2.955	vs
111	w	2.937			2.929			2.918	2.910	w	2.899	2.901	w
210	m	2.568	2.565	m	2.560	2.560	w	2.555	2.555	mw	2.544	2.543	w
012	s	2.496	2.496	ms	2.497	2.500	mw	2.504	2.503	m	2.501 _s	2.502	mw
111	w	2.461	2.462	vw	2.455	2.457	vw	2.443	2.441	vw	2.426	2.426	vw
200		2.402			2.395			2.388			2.374		200
211	s	2.396	2.397	s	2.392	2.393	m	2.390	2.391	ms	2.384	2.384	m
131	w	2.370	2.365	vww	2.368	2.368	vww	2.379			2.389		131
221		2.360			2.356			2.361			2.364		221
112	w	2.348			2.348			2.352			2.351		112
220	w	2.344	2.341	vw	2.337	2.338	vw	2.334	2.337	w	2.328 _s	2.325	w
002		2.328			2.326			2.323			2.312		002
122	w	2.320			2.323			2.336			2.345		122
022	m	2.302			2.304	2.304	vww	2.317	2.316	vw	2.322		022
121	m	2.290	2.297	mw	2.286	2.286	w	2.289	2.284	mw	2.283	2.284	w

AMBLYGONITE AND MONTEBRASITE

417

121	2:263	2:259	vw	2:256	2:253	vww	2:246	2:246	vww	2:234	2:237	vww	121
120	2:263			2:257			2:250	2:250		2:239			120
021	2:234			2:228			2:216			2:202			021
031	2:205			2:203			2:210			2:213			031
130 m	2:202	2:201	w	2:196	2:189	w	2:194	2:195	w	2:119	2:194	vw	130
201	2:142			2:136			2:132			2:105			201
201 s	2:128	2:129	m	2:123	2:123	mw	2:116	2:115	m	2:101	2:106	mw	201
211 w	2:126			2:119			2:113			2:097			211
112 vw	2:102	2:101	w	2:101			2:104			2:085	2:084	vw	112
102 m	2:102			2:099	2:098	w	2:096	2:095	w	2:072			102
102	2:088			2:087			2:081			2:050			102
030	2:065			2:060			2:056			2:063	2:062	w	030
231 mw	2:053	2:057	vw	2:051	2:051	vww	2:057	2:054	w	2:060			231
132	2:032			2:034			2:048			2:060			132
210 vw	2:012			2:006	2:024	vww	1:999	2:000	vw	1:987	1:986	vw	210
012 s	1:959	1:960	m	1:956	1:956	mw	1:947	1:947	ms	1:934	1:935	ms	012
230 mw	1:944			1:938			1:937			1:934			230
211 vw	1:936			1:931			1:929	1:929	w	1:918	1:918	vww	211
032 vw	1:928	1:930	w	1:929	1:928	w	1:941	1:930	w	1:946			032
222 w	1:926			1:926			1:932			1:935			222
221 vw	1:901			1:895			1:888			1:878			221
122 s	1:896	1:895	m	1:894s	1:896	m	1:900	1:898	m	1:898	1:899	mw	122
112 vw	1:895			1:891			1:884			1:872			112
212	1:860			1:859			1:858			1:854			212
121	1:850			1:845			1:836			1:823			121
232 w	1:820	1:818	vww	1:820	1:817	vw	1:829s	1:830	vw	1:837	1:838	vw	232
141 s	1:791	1:791	mw	1:789	1:791	w	1:795	1:796	mw	1:800	1:799	mw	141
211 w	1:769	1:770	vww	1:764	1:766	vww	1:756	1:756	vww	1:745	1:745	vww	211
131 w	1:768			1:765			1:766			1:762s	1:763	vww	131
131 s	1:750	1:749	m	1:744	1:746	mw	1:737	1:737	m	1:728	1:728	m	131
112	1:743			1:740			1:732			1:720			112
310 vw	1:704			1:699			1:695			1:686			310
241 m	1:698	1:697	vw	1:695			1:701	1:701	vw	1:705s	1:710	vw	241
142	1:68			1:689			1:700			1:709			142

involved, may be divided into two. Representations of the traces typical of each group are shown in fig. 2; the subsidiary diagram below the traces shows the range of movement of the six reflections, and it will be seen that the change in pattern is largely caused by two reflections ($12\bar{1}$ and $0\bar{2}1$) moving in the opposite direction to the other four (011 , $1\bar{1}0$, 120 , and $10\bar{1}$) with changing fluorine content.

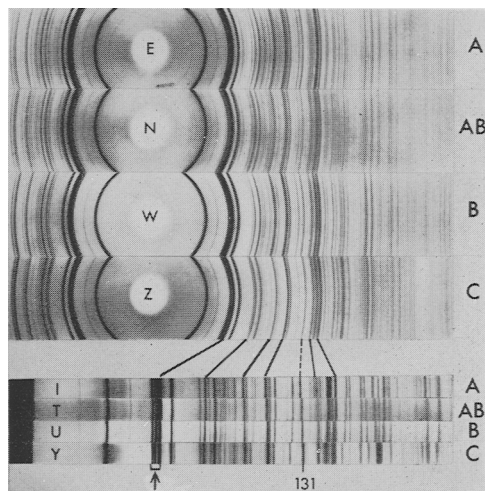


FIG. 1. Philips (114.6 mm dia.) and Guinier powder patterns of specimens in the montebrasite (A)-amblygonite (C) series, Cu- $K\alpha$ radiation. Letters A-C refer to pattern types, E-Z to the specimens (table I). The 131 reflection and the group resolved by diffractometer (fig. 2) are marked.

Pattern A, representing specimens EFGHI, corresponds to about $1\frac{1}{2}$ -3% fluorine; pattern A-AB, specimens JKLMN, to 3-4%; pattern AB-B, specimens OPQRST, to 4- $5\frac{1}{2}$ %; and pattern C, specimens XYZ, to more than $8\frac{1}{2}$ % fluorine. The boundary between patterns B and C is the least certain, and lies in the range 7% (by diffractometer) and $8\frac{1}{2}$ % (by film). Unfortunately pattern B includes the halfway point of the series, and since so few of our specimens lie in this region we cannot be certain of its limits.

It must be emphasized that our grouping into pattern types is subjective, and the placing of a particular pattern as being closer to one or other types is a matter for personal judgement. There is no evidence whatever in the present study to suggest any discontinuity in the series, and our criteria for the grouping into pattern types are presented in figs. 1 and 2.

Amongst various diffraction lines whose positions vary appreciably with composition of sample, 131 (fig. 1) moves fairly uniformly and has the advantages of good intensity and a spacing well clear of the strongest lines of likely contaminants, notably apatite. A pair of lines would have been preferable, since measurement of their angular separation would have eliminated zero errors, but none were found. Values of

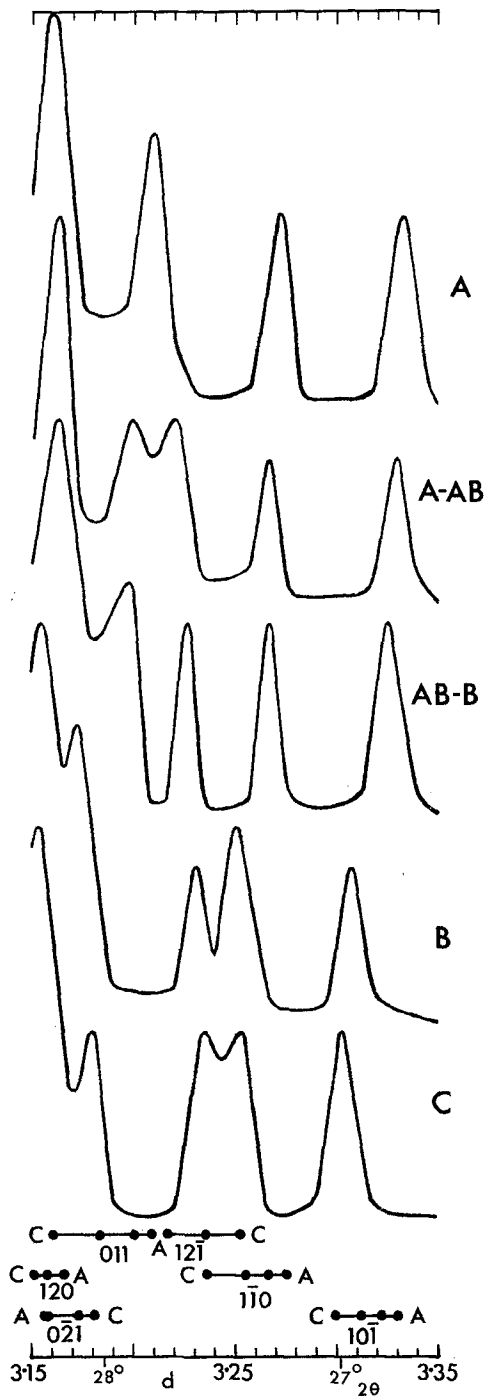
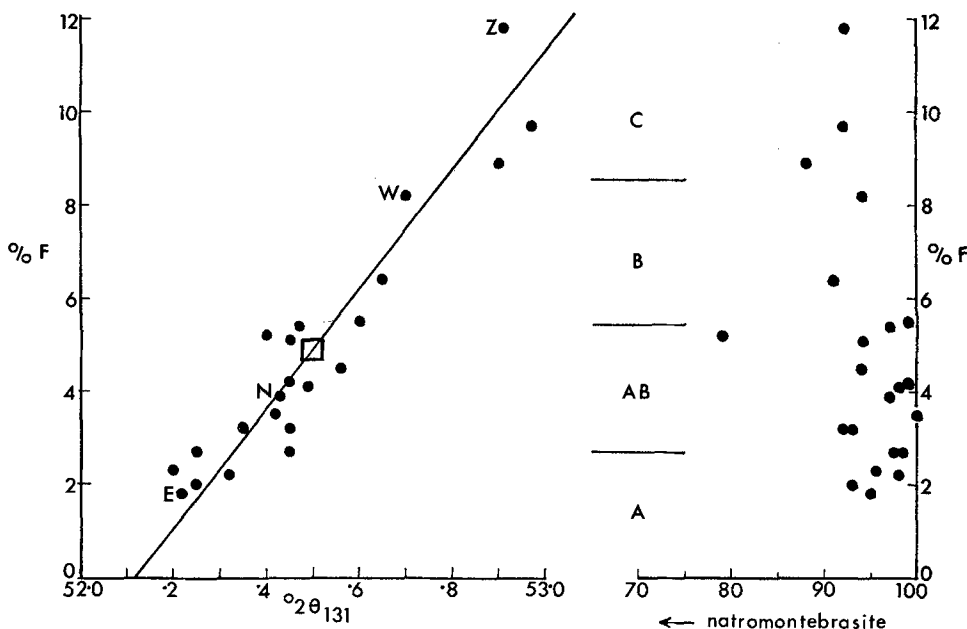


FIG. 2. Drawings of representative diffractometer traces of the six reflections, $10\bar{1}$, $1\bar{1}0$, $12\bar{1}$, 011 , 120 , and $02\bar{1}$, related to powder pattern type. The horizontal lines show the movements of the peaks from pattern A to pattern C; $1\bar{1}0$ changes places with $12\bar{1}$, 120 with $02\bar{1}$.

$2\theta(\text{Cu-K}\alpha)$ for 131 are listed in table I, and shown plotted against fluorine content in fig. 3. The line of best fit (the 'Kummell line', Hey, 1969) passes through the mean at 4.84% fluorine and $52.49^\circ 2\theta$, giving the relation: wt % fluorine = $(12.91 \pm 1.05)(2\theta_{131} - 52.12^\circ)$. The calculated correlation coefficient, $r = 0.94$, is high. The constant term (52.12) may require minor adjustment for the zero error of the diffractometer.



FIGS. 3 and 4: FIG. 3 (left). Plot of $2\theta_{131}$ against fluorine content, Cu-K α radiation; numerical values are given in table I. The hollow square marks the mean, and has sides equal to the estimated errors (0.5% fluorine, $0.05^\circ 2\theta$). FIG. 4 (right). Plot of fluorine content related to soda content. The abscissa indicates the soda content in terms of the natromontebasite-montebasite series. The indications of pattern type are common to both figures.

The indexed powder pattern in the ASTM file stops at a d -spacing of 2.334 \AA , but thanks to the kind co-operation of our colleague Dr. R. J. Davis we have been able to extend the indexing to 1.68 \AA for each of four patterns (table II) using his line-enumeration computer programme. We have also used his programme to obtain cell parameters for the same specimens, one at each end of the composition range and two intermediates (specimens E, N, W, and Z, tables I and III). Table III also gives four sets of cell parameters from the literature, of which only one is associated with a chemical analysis (Haapala, 1966); the data given by Simonov and Belov (1958) have been recalculated to the unit cell of Palache, Richmond, and Wolfe (1943) which, although not a reduced cell, is the one most commonly in use and is employed throughout the present paper. Fig. 5 is a plot of the cell parameters against fluorine content, together with the corresponding regression lines calculated for the four specimens of

TABLE III. Cell parameters for amblygonite–montebrasite series. E, N, W, and Z are the specimens studied in this paper (localities in table I); estimated errors are 0.1% in dimensions and 0.1° in angles. Specimen 1: Palache, Richmond, and Wolfe (1943). Specimen 2: Baur (1959). Specimen 3: Haapala (1966). Specimen 4: Simonov and Belov (1958)

		a(Å)	b(Å)	c(Å)	α	β	γ	V(Å ³)
Montebrasite	1	5.18	7.11	5.03	112.04°	97.83°	68.13°	—
	2	5.18	7.15	5.04	112.11	97.80	67.89	—
	E	5.189	7.173	5.040	112.50	97.90	67.81	160.5
	N	5.174	7.164	5.044	112.70	98.02	67.78	159.7
Amblygonite	3	5.175	7.173	5.047	112.89	98.08	67.67	—
	W	5.166	7.192	5.060	113.35	98.24	67.60	159.6
	Z	5.148	7.215	5.060	113.97	98.64	67.25	158.4
	4	5.16	7.21	5.06	113.20	97.90	67.53	—

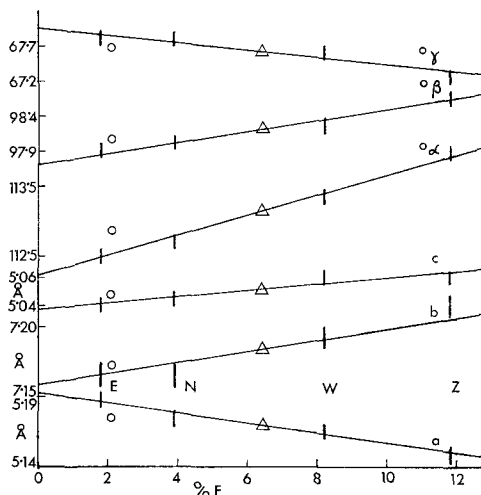


FIG. 5. Cell parameters plotted against fluorine content. Vertical bars indicate the estimated parameter errors ($\pm 0.1\%$ dimensions, $\pm 0.1^\circ$ angles). Calculated regression lines drawn through the means (hollow triangles). Data of Haapala (1966), hollow circles.

this paper; Haapala's data, plotted for the determined fluorine content 2.06%, would lie almost exactly on the lines of all six parameters at a fluorine content of 5%. The remaining three sets of literature data fit less closely and have not been plotted on fig. 5, but it would appear that two (Baur, 1959; Palache, Richmond, and Wolfe, 1943) lie well over towards montebrasite while the other set (Simonov and Belov, 1958) shows a scatter on the amblygonite side of the halfway mark. It is possible that this represents a real compositional difference and is related to the differences between the structures determined by Simonov and Belov (1958) and Baur (1959), which were commented on by Pabst (1961).

Rotation photographs of a single crystal of montebrasite were taken with Cu- $K\alpha$ radiation around [100], [110], and [001] and superposed on powder photographs taken under the same conditions. Values of h , $(k+l)$, and l were thus obtained for each powder line (or group of unresolved lines), and in conjunction with the computer-enumerated line positions the relative magnitudes of the contributions of several reflections to the same line could be deduced. On this basis it was possible to eliminate many reflections as too weak to make a significant contribution to the powder intensity, and to extend a largely unique set of line indices well beyond the limits of unique indexing based on line positions alone. Dr. Davis informs us that experience with examples where cell dimensions can be more accurately determined by other methods shows that least-squares estimates of errors are unreliable, and that cell dimensions and angles are accurate only to 0.2 % and about 0.1°.

Most of the specimens that we have studied were labelled 'amblygonite', but less than a quarter of the total contain the 6.5 % or more of fluorine to qualify for the name. It may not be a valid generalization to state that amblygonite is less common than montebrasite, but Roberts and Rapp (1965) have expressed the opinion that 'Upon further investigation, many of the reported occurrences of amblygonite [in the Black Hills, South Dakota] will, in all probability, prove to be montebrasite'. We have deliberately omitted optical studies from the present survey, and hope that our work will stimulate further investigations of this series.

The amblygonite and montebrasite specimens from Meldon have not been chemically analysed, and are, therefore not included in this survey; their identifications are based on pattern types only. Unfortunately, Mr. Kingsbury left no notes about the occurrence apart from the fact that he had picked the pieces from a fragmented pegmatite vein in the old aplite quarry. We must, therefore, be content with a bare record until further specimens are found.

Acknowledgements. We thank Mr. I. M. Hodgson for taking the diffractometer records, Dr. R. J. Davis for the photographic prints in fig. 1 and his help in the use of his computer programmes, and Dr. M. H. Hey for our introduction to the use of the Kummell line.

REFERENCES

- BAUR (W. H.), 1959. *Acta Cryst.* **12**, 988 [M.A. 15-264].
 FISHER (D. J.), 1958. *Amer. Min.* **43**, 196-7 and photograph 3D [M.A. 15-342].
 HAAPALA (I.), 1966. *Bull. Comm. géol. Finlande*, **224**, 1 [M.A. 18-120].
 HEY (M. H.), 1969. *Min. Mag.* **37**, 83.
 KITTRICK (A.) and HOPE (W.), 1967. *Amer. Min.* **52**, 286 [M.A. 18-236].
 MEGREGIAN (S.), 1954. *Anal. Chem.* **26**, 1161.
 PABST (A.), 1961. *Min. Abstr.* **15**, 264.
 PALACHE (C.), RICHMOND (W. E.), and WOLFE (C. W.), 1943. *Amer. Min.* **28**, 39 [M.A. 9-142].
 POUGH (F. H.) and HENDERSON (E. P.), 1945. *Ibid.* **30**, 572 [M.A. 9-186].
 QUENSEL (P.), 1962. *Geol. Foren. Förh.* **84**, 318 [M.A. 16-455].
 ROBERTS (W. L.) and RAPP (G., JR.), 1965. *Mineralogy of the Black Hills, South Dakota School of Mines Bull.* **18** [M.A. 17-553].
 [SIMONOV (V. I.) and BELOV (N. V.)] СИМОНОВ (В. И.) и БЕЛОВ (Н. В.), 1958. *Кристаллография*, **3**, 428; transl. as *Soviet Physics: Crystallography*, 3-429 [M.A. 15-264].

[Manuscript received 12 February 1969]