# Labelling refractive index curves for mineral series

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# Abstract

When presenting curves to designate the changes in the three principal refractive indices for biaxial members of a solid-solution or order-disorder series, it may be advantageous to designate these indices according to the crystal axes nearest which the corresponding principal vibration axes lie. Thus,  $n_c$ ,  $n_a$ , and  $n_b$  would represent refractive indices for principal vibration axes along the **c**, **a**, and **b** crystal axes for orthorhombic crystals (or closest to these for monoclinic or triclinic crystals). Any crossing of the lines for  $n_a$ ,  $n_b$ , or  $n_c$  then indicates, for the series, a change in optical orientation and its nature. The andalusite-kanonaite series, the zoisite series, an Al-rich chlorite series, and the high-sanidine to low-microcline series provide examples.

#### Introduction

The principal refractive indices for individual biaxial crystals are conventionally denoted as  $\alpha$ ,  $\beta$ , and  $\gamma$  ( $\alpha < \beta < \gamma$ ) or as  $n_x$ ,  $n_y$ , and  $n_z(n_x < n_y < n_z)$ . For members of a mineral series, however, these principal refractive indices may sometimes be better denoted as  $n_a$ ,  $n_b$ , or  $n_c$  according to whether their associated vibration axis coincides with or *lies closest to* the **a**, **b** or **c** crystallographic axes. For an orthorhombic solid solution series involving triphylite, the convention has already been used by Chapman (1943) and by Winchell and Winchell (1951). Hewlett (1959) extended the convention to a non-orthorhombic series, the alkali feldspars, when he used the symbol "b"  $[=n_b]$  to denote "the refractive index of the ray that vibrates most nearly parallel to the *b* crystallographic axis."

The advantages of better maintaining the structural identity of the refractive indices within a mineral series can be illustrated by a few examples.

## Andalusite-kanonaite series

The changes in refractive indices of andalusite (Al<sub>2</sub>SiO<sub>5</sub>) as Mn<sup>3+</sup> and/or Fe<sup>3+</sup> substitute for Al have been conventionally plotted by connecting the  $\alpha$  values, the  $\gamma$  values and the  $\beta$  values. (Fig. 1A,B). However, Gunter and Bloss (1982), using spindle stage techniques, maintained the structural identities of the refractive indices for this series. Thus, they could plot curves for  $n_a$ ,  $n_b$ , and  $n_c$  (Fig. 1C). These clearly explained the change in optic orientation and the consequent change in the sign of elongation of needles with  $\{110\}$  cleavage from (-) for relatively pure and alusites to (+) for Fe- and Mn-rich specimens. Maintaining structural identities of the refractive indices also permitted a meaningful crystal-chemical analysis of the relationship between optical properties, composition, and crystal structure for this series. The results also predicted the likely existence of nearly isotropic andalusites, which have since

been observed by J. A. Grambling (pers. comm.) of the University of New Mexico.

# **Zoisite series**

Myer (1966) used the symbols  $n_a$ ,  $n_b$ , and  $n_c$  when plotting refractive indices versus Fe<sup>3+</sup> content for five zoisites that he had measured plus two from the literature. The compositions coupled with these indices were derived from chemical analyses of zoisite concentrates deemed 98% or 99% pure, rather than from electron microprobe analyses of the same crystal as measured optically. Despite this and a relative paucity of data, Myer demonstrated that the n<sub>a</sub> and  $n_b$  lines crossed (Fig. 2) and that Termier (1898, 1900) erred in concluding that  $\beta$ -zoisite [optic plane perpendicular to the perfect cleavage  $\{100\}$  contained more Fe<sup>3+</sup> than  $\alpha$ -zoisite [optic plane parallel (100)]. Accordingly, Myer suggested that  $\beta$ -zoisite be called *zoisite*, because it approached the end-member composition Ca<sub>2</sub>Al<sub>3</sub>  $[O/OH/SiO_4/Si_2O_7]$  more closely than  $\alpha$ -zoisite, which he called ferrian zoisite.

Table 1 compares this terminology to that suggested by Holdaway (1972) and that used in the Troger (1979) tables. The refractive indices cited in these tables—namely, 1.701, 1.702, and 1.707 for  $\alpha$ -zoisite and 1.695, 1.695, and 1.702 for  $\beta$ -zoisite—lend support to Myer's conclusion that  $\alpha$ -zoisite contains more Fe<sup>3+</sup> than  $\beta$ -zoisite. Thus, substitution of Fe<sup>3+</sup> for Al<sup>3+</sup> usually increases the refractive indices of minerals as already shown for zoisites (Myer, 1966) and epidotes (Hormann and Raith, 1971). Accordingly,  $\beta$ -zoisite [O.P. (010)] probably represents a purer zoisite than  $\alpha$ zoisite [O.P. (100)] and to call it pseudozoisite (Table 1) seems inadvisable.

#### **Al-rich chlorites**

Conventional labels for the refractive indices of an Alrich chlorite series, circled in Figure 3A, convey no ad-



Fig. 1. Depictions of the variation in principal refractive indices produced by substitution of  $Fe^{3+}$  and/or  $Mn^{3+}$  in the andalusite (A1<sub>2</sub>SiO<sub>5</sub>) structure: (A) From Troger (1979); (B) From Vrana et al. (1978); (C) From Gunter and Bloss (1982). In (B) "kanonaite" represents the  $Mn^{3+}$  and  $Fe^{3+}$  end members combined.

ditional information. Mineralogists know that  $\gamma > \beta > \alpha$  or  $n_z > n_y > n_x$  and thus that the uppermost part of each line represents  $n_z$  and the lowermost  $n_x$ . On the other hand, if we replace these labels—as done in Fig. 3A—by the symbols  $n_a$ ,  $n_b$  and  $n_c$  as previously defined, the changes in optic orientation and thus in the sign of elongation relative to the {001} cleavage become readily understood. An obvious question, previously obscured, arises. What structural changes occur across the series to cause the  $n_a$  and  $n_c$  curves to cross? The question merits investigation. The change in optical orientation of the chromium chlorites (Fig. 3B) indicates that a plot of their refractive indices versus chromium content would resemble Figure 3A.

#### High-sanidine to low-microcline series

The potassium feldspar series ranges from relatively complete Al,Si disorder (high sanidine, HS) to complete Al,Si order (low microcline, LM). For a single crystal, degree of Al,Si order is expressed by  $(t_1 o + t_1 m)$ , which is the amount of Al occupying the  $T_1$  sites, and by  $(t_1 o - t_1 m)$ .

To determine how refractive indices (and cell edges)



Fig. 2. Variation of 2V and principal refractive indices with substitution of Fe<sup>3+</sup> in the zoisite  $[Ca_2AI_3(O/OH/SiO_4/Si_2O_7)]$  structure (From Myer, 1966). Note Myer's early use of the symbols  $n_a$ ,  $n_b$ , and  $n_c$ . The symbols clearly indicate that for Fe<sup>3+</sup>-poor specimens  $n_a = \alpha$  and  $n_c = \gamma$  so that (010) is the optic plane whereas, for the more Fe<sup>3+</sup>-rich specimens,  $n_b = \alpha$  and  $n_c = \gamma$  so that the optic plane is (100).

change with Si,Al ordering for K feldspars, Su et al. (1984) plotted, against  $(t_1 o + t_1 m)$ , the refractive indices and cell edges for (a) an almost fully disordered high sanidine using data extrapolated from J. V. Smith (1974, p. 386) and for (b) a low microcline using data reported for the Pellotsalo microcline by Brown and Bailey (1964). Rather than identify the refractive indices as  $\alpha$ ,  $\beta$  and  $\gamma$  for each of these two end members, Su et al. identified these principal indices as  $n_a$ ,  $n_b$  and  $n_c$  depending upon whether their associated principal vibration direction was closest to crystal axis **a**, **b** or **c**. Hewlett (1959) had earlier symbolized  $n_b$  as "b". Because the crystals are non-orthorhombic, the associated principal vibration axes were sometimes at appreciable angles to the

Table 1. Zoisite terminology

OPTIC	TERMIER (1898)	MYER (1966)	HOLDAWAY (1972)	TRÖGER (1979)
(010)	β-zoisite	zoisite	Al-zoisite	pseudozoisit
(100) <sup>1</sup>	a-zoisite	ferrian zoisite	Fe-zoisite	zoisite

<sup>1</sup> The orientation now in use-a = 16.2; b = 5.6; c = 10.0Å -indexes zoisite's perfect cleavage as (100).



Fig. 3. (A) Effect of Fe content on the principal refractive indices of Al-rich chlorites as depicted by Troger (1979) from data by Albee (1962). For the symbols Troger used (encircled), the symbols  $n_a$ ,  $n_b$ , and  $n_c$  have been substituted. These new symbols were assigned by assuming that the change in optical orientation, which occurs when Fe exceeds ca. 52 mole %, resembles that which occurs in chromium chlorites (B) when  $Cr_2O_3$  exceeds (ca.) 6 wt.%. Conversely, the change in optic orientation shown in (B) for chromium chlorites indicates that a plot of their principal refractive indices versus chromium content would resemble (A).

nearest crystal axis. The indices  $n_a$ ,  $n_b$  and  $n_c$  were assumed to vary linearly with Si,Al ordering so that the  $n_b$  and  $n_c$ lines crossed where shown in Figure 4. This linear model was used to calculate  $2V_x$  to obtain the sigmoidal upper curve. Data from the literature for 49 of 50 cases fell along the sigmoidal curve, thereby confirming the linear refractive index model, or a nearly linear one. As Su et al. note, measurement of 2V from point-to-point on a crystal in a thin section or from grain-to-grain will now suffice to determine the Al,Si ordering parameter ( $t_1o + t_1m$ ). However, some unusual potassic feldspars (Bambauer and Laves, 1960) remain at odds with the model illustrated in Figures 4.

### Sturmanite hypothesis

Peacor et al. (1983) describe the new mineral sturmanite—uniaxial (+) with  $\omega = 1.500$  and  $\varepsilon = 1.505$ —as a ferric iron, boron analogue of ettringite. They note, without explanation, that some sturmanite crystals possess uniaxial (-) cores with  $\omega = 1.499$  and  $\varepsilon = 1.497$ . Currently, because it has a sulfur: boron ratio similar to sturmanite,

they are also studying a uniaxial (-) "ettringite-like mineral from Franklin, New Jersey" for which Hurlbut and Baum (1960) determined the refractive indices  $(\pm 0.001)$  as  $\omega = 1.491$  and  $\varepsilon = 1.470$ . A plot of these indices suggests that the uniaxial cores of the sturmanite crystals may represent solid solutions between sturmanite and an ettringite compositionally similar to that from Franklin, New Jersey (Fig. 5). This hypothesis, at present mostly conjecture, can be tested if chemical analyses can be obtained for sturmanite's (-) cores. If it holds, isotropic members of the series may occur naturally.

Taylor (1973) notes that if "ettringite is heated at  $110^{\circ}$ C most of the molecular water is lost and lattice shrinkage occurs in directions normal to" the c-axis. Hurlbut and Baum (1960) previously observed that such heating changed ettringite from uniaxial (-) to uniaxial (+) and increased its average refractive index by ca. 0.05. This increase appears consonant with the heating-produced shrinkage of the unit cell observed by Bannister et al. (1936). However, the change from uniaxial (-) to uniaxial (+) appears to be at odds with the observation that shrinkage normal to c greatly exceeds that along c. Such greater



Fig. 4. A model by Su et al., (1984) for the change in  $n_a$ ,  $n_b$ ,  $n_c$  with Al,Si ordering ( $\Sigma t_1$ ) in K-feldspars ranging from complete disorder (HS or high sanidine) to complete order (LM or low microcline). The model assumes that refractive index values derived from the literature (large circles) vary linearly with  $\Sigma t_1$ . However, rather than connect 1.5238 to 1.5247 ( $\gamma$  to  $\gamma$ ) and 1.5228 to 1.5217 ( $\beta$  to  $\beta$ ), Su et al. connect the structurally more comparable indices 1.5238 to 1.5217 ( $n_c$  to  $n_c$ ) and 1.5228 to 1.5247 ( $n_b$  to  $n_b$ ). The model gains credence because it indicates that, following principles stated by Gunter and Bloss (1982),  $n_c$  decreases as cell edge c increases whereas  $n_b$  increases as cell edge b decreases. Moreover, calculations of  $2V_x$  from this linear model for  $n_a$ ,  $n_b$ , and  $n_c$  yielded a sigmoidal curve along which, as shown here, 49 of 50 data points from the literature (small triangles and circles) closely followed. The equation for this curve, and another derived from it, permitted Su et al. to predict a K-feldspar's Al,Si ordering from a simple measurement of  $2V_x$  with about the same precision as the more laborious method of measuring the b and c cell edges. The model also explains the change in orientation of the optic plane from parallel to (010) for high sanidine (A) to perpendicular to (010), or nearly so, for some orthoclases and microclines (B).



Fig. 5. The refractive indices determined by Hurlbut and Baum for an ettringite from Franklin Furnace, New Jersey—before and after its dehydration by heating at  $110^{\circ}$ C—are here plotted along with the refractive indices measured for sturmanite by Peacor et al. (1983). The abscissa represents the mole % of unheated Franklin ettringite versus either sturmanite or dehydrated Franklin ettringite. The light curves assume a simple linear change in refractive indices for solid solutions between ettringite and sturmanite. The square indicates mixed crystals whose optical properties would resemble those reported by Peacor et al. for the uniaxial negative cores they observed in some sturmanite crystals.

shrinkage normal to c would presumably favor an increase of  $n_a(\omega)$  relative to  $n_c(\varepsilon)$  and not the reverse. Combined structural and optical studies of ettringite, before and after heating, should prove enlightening.

# Suggestions and conclusion

In compendia of optical properties of minerals, the symbols  $n_a$ ,  $n_b$  and  $n_c$  should be used for biaxial crystals whenever possible. Thus, the statement that celestite is orthorhombic with  $n_a = 1.631$ ,  $n_b = 1.624$ , and  $n_c = 1.622$  concisely states—because 1.631, 1.624 and 1.622 must represent  $\gamma$ ,  $\beta$ , and  $\alpha$  respectively—that  $Z = \mathbf{a}$ ,  $Y = \mathbf{b}$  and  $X = \mathbf{c}$  so that the optic (XZ) plane must be (010).

For monoclinic and triclinic crystals, the symbols  $n_a$ ,  $n_b$ and  $n_c$  represent refractive indices for vibration axes that do not necessarily coincide with crystal axes **a**, **b** and **c**. A more specific but cumbersome symbol for, say, actinolite might be:  $n_{a,6^\circ} = 1.688$ ;  $n_b = 1.699$ ;  $n_{c,10^\circ} = 1.704$  where the angluar values represent the angle of the principal vibration axis relative to the nearest crystallographic axis. For monoclinic crystals, if standard conventions could be agreed on as to the signs of the angles, the angle subscripts would disclose the value of the angle  $\beta$  (=106° for actinolite).

In particular, the curves relating refractive indices to composition for a biaxial mineral series should be labelled  $n_a$ ,  $n_b$  and  $n_c$ . The symbols  $\alpha$ ,  $\beta$ , and  $\gamma$  (or  $n_x$ ,  $n_y$ , and  $n_z$ ), really convey no information not already conveyed by the position of one curve versus the others.

Adoption of the convention should add impetus to attempts to relate changes in optical properties to composition and crystal structure within mineral series.

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