Color zoning in labradorescence

YASUNORI MIÚRA

Department of Mineralogical Sciences and Geology,
Yamaguchi University, Yamaguchi 753

ABSTRACT

Labradorescence in the *color zoning*, in which remarkable change of lamellar thickness and An content are observed, is discussed. A wide-area macro-analysis in the region of color zoning is performed by using "X-ray contour mapping" with computer controlled electron probe microanalyzer, which has hitherto been infeasible with conventional automated electron probe microanalyzer.

The result of mapping analysis in a wide area agrees with the Bøggild rule between An content and interference color reported by Miúra *et al.* (1975). In the region of *color zoning*, it is found that thickness of alternate lamellae and composition change mutually and remarkably. For the specimen showing color zoning in *smaller* region, it is extremely difficult to find a one-to-one correlation between An content and iridescence color which agrees with the Bøggild rule. In the region of *smaller* color zoning it is possible to find that blue region contains only 2-3% less An content than the red one. In color zoning through a few millimeters area, however, the compositional difference between blue and red regions is about 7 mole % An. An content obtained by electron probe microanalysis in the *smaller* region of red-iridescence shows lower value than that in the *wider* red region showing uniform iridescence color.

Introduction

Iridescent labradorite showing labradorescence always has the bulk compositions from about An_{44} to An_{61} which are known as "Bøggild range" (Ribbe, 1975), and shows lamellar structure called "Bøggild lamellae" or "Bøggild intergrowth". The relation between iridescence color and An content, which is called "Bøggild rule", holds for the iridescent labradorite as a whole.

As discussed by Miúra *et al.* (1975), the *exact* Bøggild rule, i.e., characteristic color changes equally from U.V. to Near IR as An content increases linearly from An₄₂ to An₆₀, can be obtained for labradorescence.

Miúra *et al.* (1974) determined the An content of typical iridescent labradorite for the wider region showing uniform iridescence and having regular lamellar periodicity. From the experimental results for iridescent labradorites obtained by using an electron microscope and an electron probe microanalyzer, the following linear relationship were established between An content (mol. %) and mean thickness of lamellar pairs d (nm) (Miúra *et al.*, 1975):

$$\text{An} = 8.6 \times 10^{-2} \cdot (d) + 36.083 \quad (\sigma = 0.945), \quad (1)$$

between thickness percent of "a"-lamella, which corresponds to acidic lamella, and An content (mol. %):

$$100 \cdot d_a/d = 3.440 \cdot (\text{An}) - 119.89 \quad (\sigma = 0.844), \quad (2)$$

and between wavelength maximum λ_{max} and mean d :

$$\lambda_{\text{max}} = 3.105 \cdot (d) - 21.178 \quad (\sigma = 0.999). \quad (3)$$

The bulk compositions reported by Miúra *et al.* (1974) are the results of point analyses in the wider region showing uniform iridescence color and having regular lamellar periodicity. However, there are also iridescent specimens showing color zoning in the smaller area (cf. Ribbe, 1975), in which remarkable changes of lamellar thickness and An content are observed. It is therefore necessary for such specimens to apply two- or three-dimensional analysis.

In this paper, the Bøggild rule and the changes of An content and lamellar thickness in both the transverse and depth directions of iridescent labradorite are discussed quantitatively.

Experimental

To perform wide-area marco-analysis, the writer has taken an entirely new analyzing technique of two-dimensional analysis using "X-ray contour mapping" with computer controlled electron probe microanalyzer, JCXA-50A, which has hitherto been infeasible with conventional automated electron probe microanalyzer (EPMA).

In the techniques of conventional two-dimensional EPMA analysis, the elemental distribution is displayed on a CRT by detecting characteristic X-rays generated from the specimen surface scanned with a finely focussed electron probe. In the new analyzing method X-ray intensities of up to three elements are simultaneously measured at each analyzing point by shifting the specimen stage in a stepwise two-dimensional way, with electron probe position fixed. The X-ray intensities thus obtained are classified into 10 preset levels (0-9) by a built-in computer, and these level values are two-dimensionally displayed on a character display. The software, a special conversational computer language named the "JECASS-X" which is the abbreviation of JEOL Computer Automation System Software, is applied for crystal position displacement, crystal exchange for up to three channels of spectrometers, counter and timer operation, X-, Y- and Z-axes position displacement for the specimen stage, an electron beam shutter operation and probe current digital readout (Okudera *et al.*, 1976; Onuma *et al.*, 1976).

In the case of quantitative analysis, the probe current is estimated by a Farady cage before each peak intensity measurement of the standard specimen. Each standard specimen positioning is followed by peak searching sequence, and the result after instrumental corrections for dead time, background and probe currents are typed out. After standard specimen measurements, the sequence goes into unknown specimen measurements. For quantitative correction method, the on-line Bence and Albee method is used.

Results and discussions

Fig. 1 shows a sketch of the color zoning of iridescent labradorite from Nain, Labrador. The direction of the boundary of the color zoning is indicated by scar for identification of boundary in two-dimensional EPMA analysis. The Ca, Na and K characteristic X-ray contour mappings in the same region of color zoning are shown in Fig. 2. These contour mappings are given by the level-values 0-9. Level-values smaller than 0 are all given by “-” and those larger than 9 are given by “X”. The X-ray intensity distribution of Na and Ca are fairly non-uniform over the color zoning. These contour mappings show subtle differences of X-ray intensity in each of the

Fig. 1. Sketch of the relation between interference color and composition in iridescent labradorite from Nain, Labrador.

Fig. 2. Ca, Na and K characteristic X-ray contour mapping in the same region of the wider color zoning in Fig. 1 by using computer controlled EPMA (JCA-50A). The contour mappings are given by the level-values 0-9. Level-values smaller than 0 are all given by “-”, and those larger than 9, by “X”.

phases, which are indicated by light and shade in Fig. 3. The numerical tables under the contour mappings represent a sort of mode analysis. The total number of analyzing points in the analyzing area is 816. The second column gives the frequency of appearance, namely, the numbers of analyzing points at each level-values. The third column represents their percentages. For example, 123 analyzing points which are 15.07% of the total have the level-value 7 for Ca content. It is also found in Fig. 3 that Na is rich in the upper part; Ca, rich in the lower part, and K distributes uniformly. This result agrees well with bulk An content obtained for the same region by

quantitative EPMA analysis (cf. Fig. 1).

The result of mapping analysis agrees with the exact relation of Bøggild rule by Miúra *et al.* (1975), who has established the relationship among An content, lamellar thickness, iridescence color and its wavelength maximum in the specimen showing typical uniform iridescence color (Tables 1 and 2). The lamellar thickness, its iridescence color and its wavelength maximum in the regions of wider and smaller color-zonings are shown in Table 1, where *mean* values of An content are listed because compositions in color zoning are changeable. Table 2 shows chemical composition, mean values of *total* and *individual* thicknesses of alternate lamellae, and iridescence color in the Bøggild intergrowth. Of two types of lamellae, it is assumed that "a"-lamella corresponds to An-poor (or acidic) lamella, and "b"-lamella, An-rich (or basic) one, respectively.

Fig. 4 shows the relationship among bulk An content, thicknesses of alternate lamellae, iridescence color and its wavelength maximum in the Bøggild range, in which the compositions of Bøggild lamellae are plotted. Solid curves show the results obtained from equations (1) and (2). This shows that the mean composition difference between

Table 1. Lamellar thickness, its iridescence color and its wavelength maximum in the Bøggild intergrowth showing both wider and smaller color zonings.

Lamellar thickness $d(=d_a+d_b)$ (nm)	Iridescence color	Wavelength maximum (nm)	An content (mol. %)	An content in smaller color zoning (mol. %)
80~125	Near U.V.	270~380	44~47	44~49
~145	violet	~445	~49	} 48~53
~160	blue	~495	~50	
~180	green	~555	~52	} 51~56
~190	yellow	~585	~53	
~205	orange	~630	~54	} 53~59
~260	red	~800	~59	
~310	Near IR	~960	~64	~64

Table 2. Composition, mean values of total and individual thicknesses of alternate lamellae, and iridescence color in the Bøggild intergrowth.

An and Or contents (mol. %)	Total lamellar thickness (nm)	Mean a-lamella thickness (nm)	Mean b-lamella thickness (nm)	Iridescence color (nm)
An _{48.9} Or _{3.3}	125	70	55	violet
An _{50.0} Or _{2.7}	137	57	80	blue
An _{50.8} Or _{2.8}	160	77	83	blue
An _{51.8} Or _{3.0}	180	87	93	green(∼yellow)
An _{52.8} Or _{2.6}	174	71	103	yellow
An _{53.2} Or _{5.4}	180	55	125	yellow
An _{54.2} Or _{5.2}	200	73	127	yellow(∼orange)
An _{54.7} Or _{4.7}	214	62	152	orange
An _{58.1} Or _{3.3}	245	55	190	red(∼near IR)

adjacent lamellae is 10-25 mole percent An, which agrees well with those by Nissen (1971) and Miúra *et al.* (1975) and Miúra and Tomisaka (1978) shown in the lower part of Fig. 4. And it is found that Near U.V.-iridescence is observed from about An₄₄ to An₄₉, and Near IR-iridescence, from An₅₆ to An₆₃. The experimental values of these lamellar thicknesses are plotted in Fig. 4, which shows the data by Nissen *et al.* (1967) as open circles, those obtained by Bolton *et al.* (1966) as cross marks, and those by Miúra *et al.* (1974, 1975) as filled circles. Since Bolton *et al.* (1966) did not report the compositions of their iridescent labradorites, the bulk An contents of their specimens estimated by the author were used to plot the data in this figure. The typical lamellar thicknesses of each iridescence shown in Fig. 5 are referred to as the filled circles in Fig. 4. It is also found from Fig. 4 that lamellar microstructure in the 100 nm scale disappear in both bulk compositions of An_{42(±1)} and An_{60(±1)}.

Although it is shown in Fig. 4 as solid curves that the thicknesses of “a”- and “b”-lamellae change unequally in the Bøggild range, the

Fig. 4. Relationship among An-content, two kinds of thicknesses of alternate lamellae, iridescence color and its wavelength maximum in the Bøggild intergrowth. Solid curves are obtained from equations (1) and (2). Their limits of error are shown by fine curves. The experimental values of lamellar thicknesses are plotted by open circles (Nissen *et al.*, 1967), cross marks (Bolton *et al.*, 1966), filled circle (Miura *et al.*, 1974, 1975) and squares (Nissen, 1971). Heavy lines M show the limit of labradorescence to be observed by the naked eyes. The lower part of the figure shows the compositions of the Bøggild lamellae reported by Miura *et al.* (1974, 1975), Miura and Tomisaka (1978), McConnell (1974), Ribbe (1975) and Nissen (1971). Distribution of iridescence color in both wider and smaller color zonings are also shown apart in the figure.

(a)

(b)

(c)

(d)

(e)

Fig. 5. Electron micrographs of Bøggild lamellae in Nain labradorites. These data are those shown as the filled circles in Fig. 4, and listed in Table 2. (a) : Violet-iridescence, mean thickness of lamellar pairs $d(=70+55)=125(\text{nm})$. Replica image. (b) : Blue-iridescence, $d(=57+80)=137(\text{nm})$. Replica image. (c) : Yellow-iridescence, $d(=55+125)=180(\text{nm})$. Replica image. (d) : Orange-iridescence, $d(=73+127)=200(\text{nm})$. Replica image. (e) : Red-iridescence, $d(=55+190)=245(\text{nm})$. Replica image. (f) : Blue-iridescence, $d(=77+83)=160(\text{nm})$. Bright-field image. (g) : Blue-iridescence, $d=160(\text{nm})$. Replica image cut diagonally. (h) : Replica image and lamellar flake to distinguish between An-poor and An-rich lamellae. Blue-iridescence, $d=137(\text{nm})$.

Bøggild rule holds for the *mean* thickness of lamellae pairs. However, it is often difficult to obtain the Bøggild rule in the *smaller* color zoning, where the ratio of alternate lamellar thicknesses d_b/d_a remarkably changes in both the transverse and depth directions of the specimen. And it is also difficult to take an electron micrograph showing color zoning including both blue and red regions (cf. Fig. 6). Fig. 7, therefore, shows schematic diagram of changes of lamellar thicknesses in both directions. The change in ratio of lamellar thicknesses is produced not directly by increasing the lamellar thickness parallel to lamellar layer, but by branching the adjacent lamellae. It is twin (Fig. 6) and tweed textures (Fig. 8) mutually to intersect lamellar structures at acute and right angles, respectively.

Fig. 6. Electron micrograph of Bøggild lamellae showing uniform thickness and albite twin (cf. Miúra *et al.*, 1974) in blue-iridescent labradorite from Nain.

Fig. 7. Schematic diagram of the color zoning mainly in the transverse direction, where composition and thicknesses of alternate lamellae change remarkably. The change in lamellar thicknesses is produced by branching the adjacent lamellae through several millimeters area.

Fig. 8. Electron micrograph of Nain labradorite showing tweed texture cross $(1.\bar{1}2.1)$ and $(10\bar{3})$ lamellae in replica image. Yellow-iridescence. $d=180(nm)$.

For such minor specimen as smaller color zoning, it is extremely difficult to find the one-to-one correlation between composition and iridescence color which agrees with the exact Bøggild rule obtained in the specimen showing uniform color in the area of several centimeters. An content in smaller color zoning shows lower value than that in wider zoned region consisting of uniform colors. It is satisfactory to consider that An content is the value in depth direction of about one micrometer, which corresponds to thickness of a few ten lamellar layers. And iridescence color is produced by a few thousand lamellar layers. In the smaller color zoning, it is possible to find that blue region contains 2-3% less An content than red one (Ribbe, 1975). But in the color zoning through *several millimeters* area, the composition difference between blue and red regions is about 7 mole % An (Table 1, Fig. 4).

Generally in the region of color zoning, it is found that thicknesses of alternate lamellae change remarkably and that An content changes mutually. The exact relation of Bøggild rule obtained from equations (1) and (2) holds in labradorite showing color zoning through a few centimeters area, but exact relation between bulk composition and iridescence color is not obtained in smaller color zoning in a few ten to hundred micrometers scale. The minor color zoning can be observed at random around inclusions, such as FeO-TiO system minerals up to 0.2 mm across, often in twin individual.

Therefore, Bøggild rule is a relation in typical region of wider color zoning because the linear relationship between composition and iridescence has been established in well-characterized specimens having uniform iridescence and regular lamellar periodicity (Miúra *et al.*, 1974, 1975). To avoid confusion of bulk compositions in color zonings, description of the *lamellar thickness*, also with iridescence color, is indispensable to understand the exact relation of Bøggild rule in both wider and smaller color zonings.

Ribbe (1975) pointed out that red region of An_{58} surrounded by

blue region of An_{61} (Miúra *et al.*, 1974) was produced by difference in order of interference color. Result of computer simulation of labradorescence (Miúra *et al.*, 1974, 1975; Miúra, 1976), however, shows that red-iridescence color is produced only from first order of interference color. The higher orders of interference color, especially in red-iridescence, are not produced by lamellar microstructure in the 100 nm scale. Periodic layer-structures in the micrometer scale should be required for red color of higher orders of interference color.

Therefore, the compositions in color zoning should be obtained by EPMA analysis under these conditions that (1) surface of specimen is cut in a direction parallel to lamellar planes, and (2) analyzing points are the place having no complex structures, such as twin and tweed texture and inclusion minerals of FeO-TiO system, alkali feldspar and pyroxene.

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