

## Ion microprobe mass analysis of exsolution lamellae in labradorite feldspar

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### Abstract

A method for *in situ* quantitative microanalysis of exsolution lamellae in labradorite (“Bøggild intergrowth”) has been developed, using an ion microprobe mass analyzer. Ion probe analyses for two kinds of exsolution lamellae of low-temperature intermediate plagioclases showing various iridescent colors led to the calculation of the following determinative curves:

Mol % An =  $97.0[9.4] - 142.1[27.5](I_{\text{Na}^+}/I_{\text{Al}^+})_{\text{corr}}$ , where  $(I_{\text{Na}^+}/I_{\text{Al}^+})_{\text{corr}}$  is the observed  $(I_{\text{Na}^+}/I_{\text{Al}^+})$  multiplied by an An correction factor  $f_{\text{An}}$ .

Mol % Or =  $0.73[0.28] + 104.8[33.0](I_{\text{K}^+}/I_{\text{Al}^+})_{\text{corr}}$ , where  $(I_{\text{K}^+}/I_{\text{Al}^+})_{\text{corr}}$  is the observed  $(I_{\text{K}^+}/I_{\text{Al}^+})$  multiplied by an Or correction factor  $f_{\text{Or}}$ .

From trace analysis of a micron area (“line analysis”) and three-dimensional analysis of lamellar multilayer including the direction of depth (“in-depth analysis”), the mean chemical compositions of An-poor and An-rich lamellae are  $\text{An}_{44(\pm 4)}\text{Or}_{3(\pm 1)}$  and  $\text{An}_{58(\pm 6)}\text{Or}_{2(\pm 1)}$ , respectively. The mean composition difference between adjacent lamellae is 15 percent An and 1 percent Or.

### Introduction

Determination of the composition of individual exsolution lamellae of intermediate plagioclases ( $\text{An}_{\sim 40}$ – $\text{An}_{\sim 60}$ ) has been one of the objectives of many mineralogical studies since the observation by Laves *et al.* (1965) that a cooling intermediate plagioclase unmixes, forming lamellae with compositional differences.

At present, there are four reports of compositions of Bøggild lamellae:

1. From the ratio of lamellar thicknesses from micrographs for fourteen labradorites, Nissen (1971) concluded that the chemical compositions of the two lamellae are  $\text{An}_{35-40}$  and  $\text{An}_{60-65}$ .

2. From microanalysis of lamellae with an analytical electron microscope (EPMA), Nissen *et al.* (1973) and Cliff *et al.* (1976) confirmed that in a sample of bulk composition  $\text{An}_{55.1}\text{Or}_{2.9}$  the compositional difference between An-rich and An-poor lamellae is at least 12 percent An.

3. From the change of spacing and orientation of superlattice fringes between lamellae, McConnell (1974) found that in a sample of composition  $\text{An}_{54.1}\text{Or}_{2.1}$  the extremes of composition difference between adjacent lamellae may be 10–12 percent An.

4. From the intensity maxima calculated from the computer simulation model of labradorescence, Miura *et al.* (1974, 1975) found that labradorescence may occur in the composition range from  $\text{An}_{47}$  to  $\text{An}_{60}$ , and that lamellar microstructure in the 100 nm scale may disappear in both bulk compositions of  $\text{An}_{42(1)}$  and  $\text{An}_{60(1)}$ .

Unfortunately, these results are based on indirect estimates or direct analysis, without detecting sodium and potassium, of compositions of the lamellae in a sample. Whether or not these results are valid cannot be known until further systematic and quantitative research is done. Therefore, it is necessary to directly analyze lamellae on a few samples using another high-resolution device.

### Analytical technique

The ion microprobe analyzer (IMA) is an instrument for analysis of a small volume at a solid surface which has been bombarded by the primary ion beam (*cf.* Nishimura and Okano, 1969; Andersen and Hinthorne, 1972). The production of secondary ions, sputtered from the surface by a high-energy ion beam in a mass spectrometer, can be maintained at a higher level by bombarding the lamellar surface with

oxygen ions rather than with inert gas ions such as argon. When an insulator, such as a mineral, is bombarded by positive ions, the sample becomes positively charged. In order to prevent charge accumulation on the surface, two methods are available:

- (1) Use of negative ions as the primary ions (Andersen *et al.*, 1969).
- (2) Electron spray method (Nakamura *et al.*, 1972).

In the first method, the surface potential increases with time until a critical value is reached, at which time the emitted secondary electrons are drawn back to the sample by the surface potential; this process causes charge compensation. Consequently, the surface potential eventually attains a constant value. The electron spray method, developed by Nakamura *et al.* (1972) and Nakamura *et al.* (1974), eliminates charge on the sample surface by spraying the target area with a low-energy electron beam overlapping the primary ion beam.

The electron probe microanalyzer (EPMA) detects elements of low atomic number with difficulty and is not suitable for making precise analyses of sub-micrometer width lamellar layers. The IMA, however, with its superior sensitivity can detect all the elements and their relative isotopic ratios in lamellar layers of widths down to tens of angstroms. Therefore, lamellar layers can be quantitatively analyzed with a resolution of tens of angstroms using an IMA.

In quantitative analysis by an IMA, two different methods have been employed. One is the working curve method (Hinthorne and Andersen, 1975; Hinthorne and Ribbe, 1974) and the other is a thermodynamic approach based on the Local Thermal Equilibrium (LTE) method (Andersen and Hinthorne, 1973; Shimizu *et al.*, 1974). Although the latter method still leaves some fundamental questions unsolved, the former method is recognized as a valuable technique, especially for exsolution lamellae which have similar compositions and crystal structures (Hinthorne and Andersen, 1975).

We have established IMA analytical procedures for quantitative analysis of exsolution lamellae by the working-curve method and for determining the compositions of lamellae in a Bøggild intergrowth.

#### Analytical procedure

Figure 1 is a schematic diagram of the Ion Microprobe Analyzer (IMA), IMA-2, as presently installed at Naka Branch, Central Research Laboratory, Hitachi Ltd., Ibaragi, Japan (see Nakamura *et al.*, 1972,

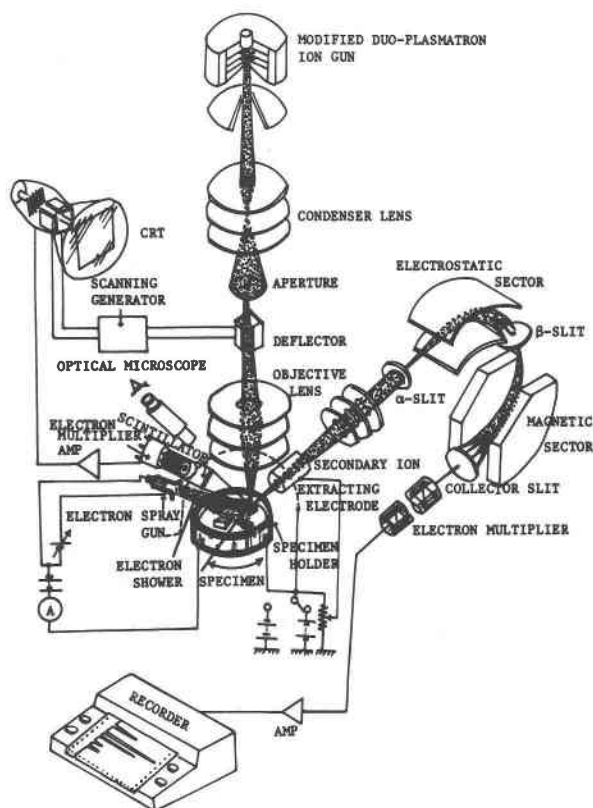


Fig. 1. Schematic diagram of the ion microprobe analyzer, IMA-2, as presently installed at Naka Branch, Central Research Laboratory, Hitachi Ltd., Ibaragi, Japan. The construction of the electron spray gun is also illustrated.

1974, 1976). The construction of the electron spray gun is also illustrated in Figure 1. The accelerating potential ( $\pm 3\text{kV}$ ) for the secondary ions is applied to the sample holder, and the hemispherical shield covering it is made of wire gauze (Nakamura *et al.*, 1976). The surface charge accumulation is effectively compensated by spraying low-energy electrons over the lamellar surface.

Samples of iridescent labradorite from Nain, Labrador, showing uniform characteristic color with no color zoning were prepared by cutting cubes of approximately 1 cm with a diamond cutter. Two surfaces of the cubes were cut along planes closely parallel to known lamellar plane; these surfaces were buff-polished with carborundum, and finished with diamond paste C, 0.25  $\mu\text{m}$  in size. These samples were analyzed by electron probe microanalyzer (EPMA) JXA-50A at 15kV (see Table 1) before IMA analysis.

Before taking the secondary ion intensities using the IMA, the sample surface was ion-etched for one minute by approximately 500  $\mu\text{m}$  so as to remove surface contamination. The samples were then bom-

Table 1. EPMA data on iridescent labradorite feldspars from Labrador, Canada

	LB-1	LG-1	LY-1	LY-2	LR-1
SiO <sub>2</sub>	55.45	54.64	54.36	55.14	54.33
Al <sub>2</sub> O <sub>3</sub>	29.54	31.66	29.92	28.33	29.45
CaO	9.78	9.51	10.36	11.09	11.06
Na <sub>2</sub> O	4.94	4.58	4.83	4.56	4.05
K <sub>2</sub> O	0.45	0.46	0.43	0.89	0.53
Total	100.16	100.85	99.90	100.01	99.42

Ab (mol %)	46.4(1)	45.2(1)	44.6(2)	40.4(2)	38.6(2)
An (mol %)	50.8(2)	51.8(2)	52.8(2)	54.4(2)	58.1(2)
Or (mol %)	2.8(1)	3.0(1)	2.6(1)	5.2(1)	3.3(1)

\* Estimated standard deviations are in round brackets.

barded with a positively-charged primary beam of oxygen (<sup>16</sup>O<sup>+</sup>) at 12keV, using a beam diameter of approximately 50 μm, a beam current of 5 × 10<sup>-8</sup>A, an electron spray gun current of 4 μA, and an electron beam diameter of 1 millimeter. The wall effect created by cratering can be eliminated by the electronic aperture method (Andersen *et al.*, 1974). The point being analyzed can be viewed through an optical microscope while under bombardment.

Typical spectra of minor and major lamellae of the blue-iridescent labradorite (An<sub>50.8</sub>Or<sub>2.8</sub>) are shown in Figure 2, where the estimated compositions were calculated from the following method.

The analytical method is based on the observation that the ratio of different sputtered ion species from a given matrix has been observed to be constant (Hinthorne and Andersen, 1975). The data necessary to determine the An and Or contents are secondary ion intensities of the <sup>23</sup>Na<sup>+</sup> and <sup>39</sup>K<sup>+</sup> isotopes and the secondary ion intensity of a major-element isotope for intersample normalization. We have chosen to use <sup>27</sup>Al<sup>+</sup> for the internal standard, because it requires no correction for natural isotopic abundance and it has the highest relative yield of all elements in the sample. It was found that mole percent An of a lamella should be calculated from the ratio (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>), because it is more sensitive to changes in lamellar compositions than other possible ratios.

To determine working curves, the measured ion intensity ratio, (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>obs</sub> and (I<sub>K</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>obs</sub>, corrected for natural isotopic abundance in the case of <sup>39</sup>K<sup>+</sup>, were obtained and multiplied by the factors *f*<sub>An</sub> and *f*<sub>Or</sub>. The factor *f*<sub>An</sub> is the average atomic percent

Al, as determined by electron microprobe analyses, of several well-characterized standard samples of composition An<sub>50.8</sub>Or<sub>2.8</sub>(LB-1), An<sub>52.8</sub>Or<sub>2.8</sub>(LY-1), and An<sub>58.1</sub>Or<sub>3.3</sub>(LR-1). Similarly the factor *f*<sub>Or</sub> is the average atomic percent Al for samples LB-1, LG-1 (An<sub>51.8</sub>Or<sub>3.0</sub>), and LY-2 (An<sub>54.4</sub>Or<sub>5.2</sub>). These mean values determined by EPMA are accurate to approximately 0.4 percent in An (or Ab) content and 3 percent in Or content (*cf.* Table 1). The corrected ion intensity ratios (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>corr</sub> and (I<sub>K</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>corr</sub> are equal to (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>obs</sub> × *f*<sub>An</sub> and (I<sub>K</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>obs</sub> × 1.074 × *f*<sub>Or</sub>, respectively. These values are listed in Table 2 and plotted against the known bulk An and Or con-

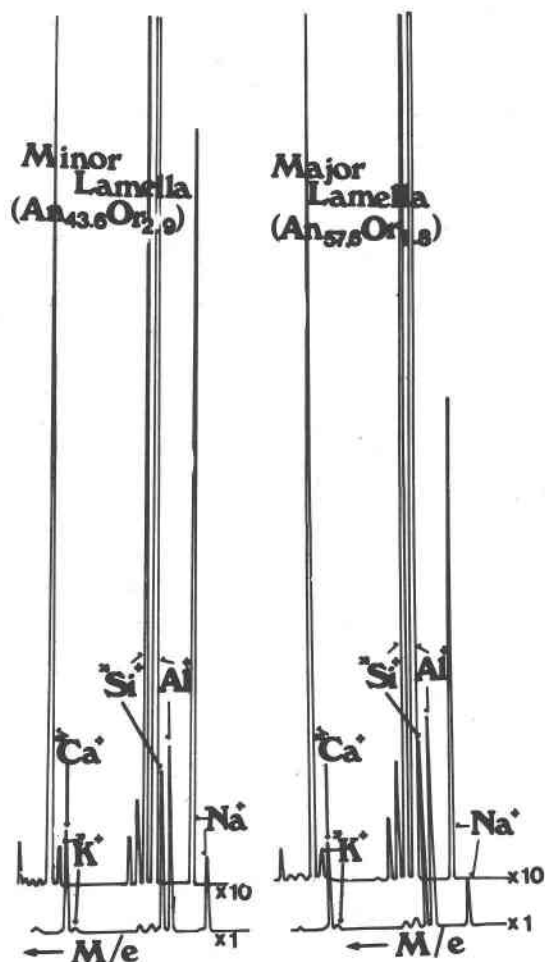


Fig. 2. Mass spectra of minor and major lamellae of blue-iridescent labradorite (LB-1) from ion microprobe analysis. Primary beam is <sup>16</sup>O<sup>+</sup>; beam energy, 12 keV; secondary ion accelerating voltage, 3 kV; vacuum, 4 × 10<sup>-7</sup> torr; spot size of diameter 50 μm. M represents mass (g) and e electric charge (cgs esu). The estimated compositions of the two lamellae were calculated from the An and Or contents working curves (*cf.* Figs. 3 and 4; Table 3).

Table 2. The observed and corrected ion intensity ratios and An and Or correction factors for the establishment of working curves for determining An and Or contents

Specimen	Iridescence Color	(I <sub>Na</sub> <sup>+</sup> /I <sub>Al</sub> <sup>+</sup> ) <sub>obs</sub>	f <sub>An</sub> <sup>*</sup>	(I <sub>Na</sub> <sup>+</sup> /I <sub>Al</sub> <sup>+</sup> ) <sub>corr</sub>	(I <sub>K</sub> <sup>+</sup> /I <sub>Al</sub> <sup>+</sup> ) <sub>obs</sub>	K <sub>100</sub> <sup>**</sup>	f <sub>Or</sub> <sup>***</sup>	(I <sub>K</sub> <sup>+</sup> /I <sub>Al</sub> <sup>+</sup> ) <sub>corr</sub>
LB-1	Blue	0.32 <sup>##</sup>	1.00 <sup>#</sup>	0.320	0.015	0.017	1.00 <sup>#</sup>	0.017
LG-1	Green	(0.32) <sup>##</sup>	(1.01)	(0.318)	0.025	0.027	0.98	0.026
LY-1	Yellow	0.31	1.02	0.316	(0.018)	(0.019)	(1.00)	(0.019)
LY-2	Yellow	(0.30)	(1.02)	(0.306)	0.043	0.046	0.88	0.041
LR-1	Red	0.26	1.05	0.273	(0.022)	(0.024)	(0.99)	(0.024)

\* f<sub>An</sub> = (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>corr</sub> / (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>obs</sub>.

\*\* K<sub>100</sub> = 1.074 x (I<sub>K</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>obs</sub>.

\*\*\* f<sub>Or</sub> = (I<sub>K</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>corr</sub> / K<sub>100</sub>.

# It is assumed that f<sub>An</sub> = f<sub>Or</sub> = 1.00 in the well-characterized specimen (LB-1).

## The values in round brackets were not used to establish the working curves.

tents in Figures 3 and 4, where Ab(mol percent) = 100 - An - Or. The equations for the regression lines relating (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>corr</sub> and (I<sub>K</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>corr</sub> to An (or Ab) and Or contents, respectively, are given in Table 3.

**Working curves for determining An and Or contents**

The working curves for estimating An (or Ab) and Or contents of lamellae (Figs. 3 and 4) show good correlations between values of An (or Ab) and Or contents determined by the EPMA and the normalized <sup>23</sup>Na<sup>+</sup> and <sup>39</sup>K<sup>+</sup> intensities as measured with the IMA. Thus the compositions of lamellae can be determined using the working curves. In general, the sputtering rate depends on the matrix, especially the binding energy of an atom to the surface, and the current density of the primary ion beam. Thus the sputter rate will remain constant with a stable primary ion beam and a sample of uniform composition (Hinthorne, personal communication). In the analysis of lamellar layers the sputtering rate of the beam can be obtained by measuring the distance between the boundary of lamellae obtained by ion intensity ratio, and by the known analyzing conditions (Nakamura *et al.*, 1976). In this study the mean sputtering rate of the beam on the lamellar layer was 6 nm/min. This indicates that the lamellar layer can be quantitatively analyzed in depth with a resolution of tens of angstroms.

Figures 5 and 6 show typical results from the analyses of blue- and yellow-iridescent labradorites. An (or Ab) contents of the blue (Fig. 5) and yellow

(Fig. 6) samples can be determined using the working curve in Figure 3 and Table 3. Or contents in these samples can also be determined using Figure 4 and Table 3. The error bars shown in Figures 5 and 6 are those calculated from the errors in the counts for the two elements, 67/√N (%) where N is the observed count. Figure 6 shows distinct dips in the sodium (or calcium) and potassium content between the two lamellae. This shows that there is a local uneven distri-

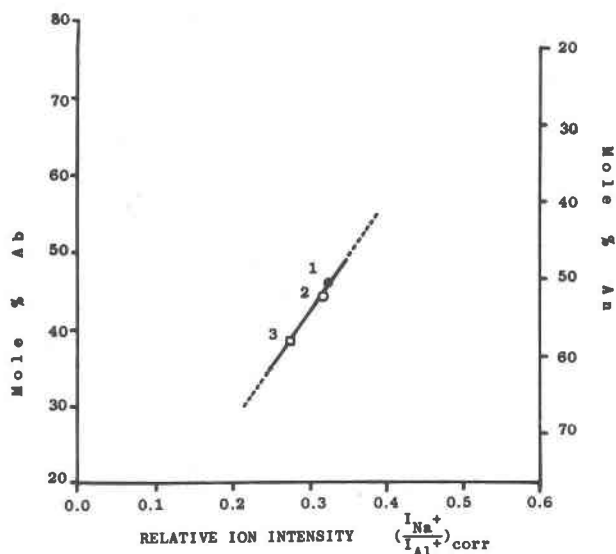


Fig. 3. Working curve for mole percent An (or Ab) in iridescent labradorite feldspars. Ab(mol %) = 100 - An - Or. The corrected ion intensity ratio (I<sub>Na</sub><sup>+</sup>/I<sub>Al</sub><sup>+</sup>)<sub>corr</sub> is obtained from the equation in Tables 2 and 3. Standard samples are 1:LB-1(Ab<sub>46.4</sub>An<sub>50.8</sub>Or<sub>2.8</sub>), 2:LY-1(Ab<sub>44.6</sub>An<sub>52.8</sub>Or<sub>2.6</sub>), and 3:LR-1(Ab<sub>38.6</sub>An<sub>68.1</sub>Or<sub>3.3</sub>).

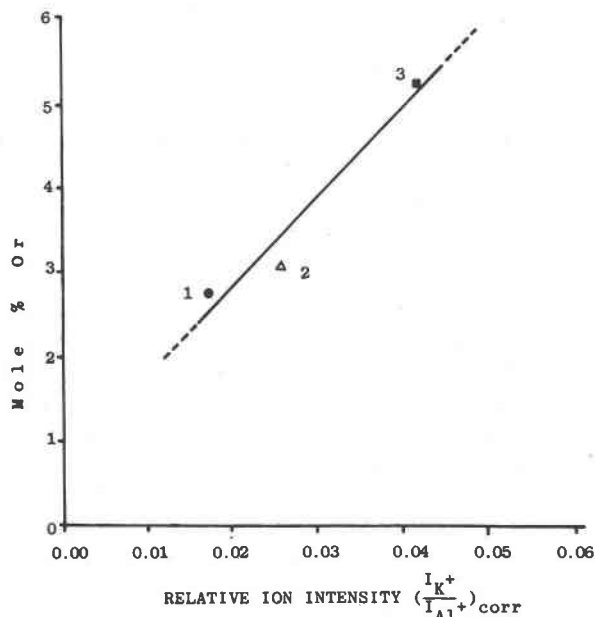


Fig. 4. Working curve for mole percent Or in iridescent labradorite feldspars. The corrected ion intensity ratio  $(I_{K^+}/I_{Al^+})_{corr}$  is obtained from the equation in Tables 2 and 3. Standard samples are 1:LB-1, 2:LG-1( $Ab_{45.2}An_{51.8}Or_{3.0}$ ), and 3:LY-2 ( $Ab_{40.4}An_{54.4}Or_{5.2}$ ).

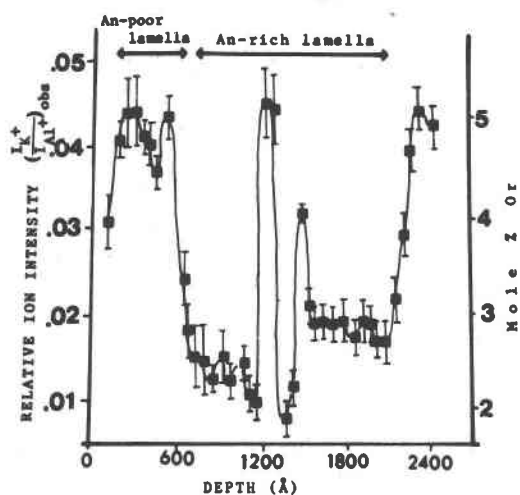
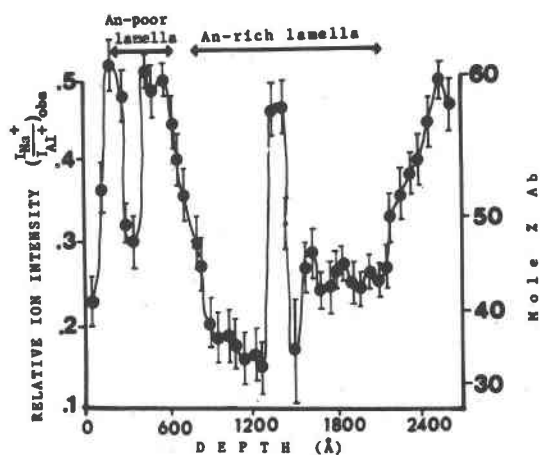


Fig. 6. The variations in calculated mole percent Ab and Or in the in-depth analysis of lamellae of yellow-iridescent labradorite, LY-1( $Ab_{44.8}An_{52.8}Or_{2.8}$ ). The error bars shown are obtained from the same calculation in Fig. 5.

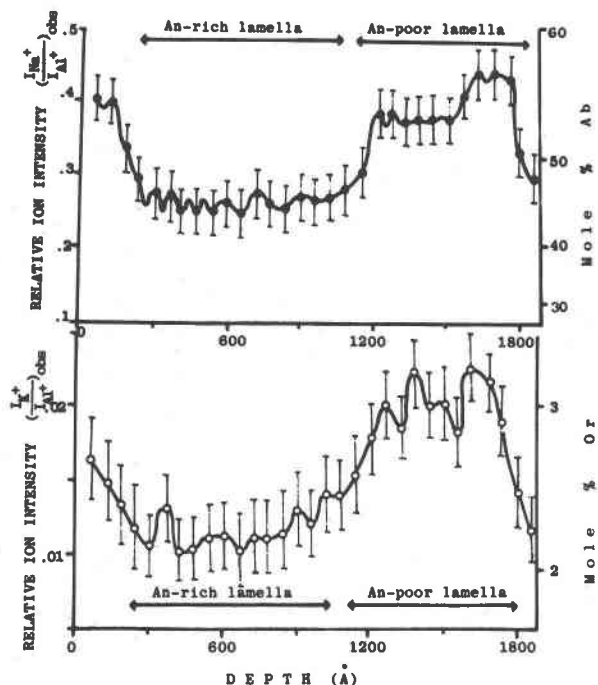


Fig. 5. The variations in calculated mole percent Ab and Or from the in-depth analysis of lamellae of blue-iridescent labradorite, LB-1( $Ab_{46.4}An_{50.8}Or_{2.8}$ ). The error bars shown are those of  $1\sigma$  calculated from the errors in the counts for the elements.

bution of composition, such as more sodic lamellar domains in the An-rich lamellae of yellow-iridescent samples.

In the analysis of lamellar compositions the wall effect becomes increasingly important in successive analyses. The effects of inclusions and non-planar lamella are also important. Line analyses over the stepped lamellar surface were also made by carefully traversing the probe  $60\ \mu\text{m}$  across them, using the same analyzing conditions as in the lamellar analysis. The results of analyses in the blue, yellow, and red samples are listed in Table 4. The calculated bulk An and Or contents were obtained as follows (see Table 4):

$$C_{\text{calc}} = C_A N_A + C_B N_B,$$

Table 3. Equations for regression lines relating  $(I_{Na^+}/I_{Al^+})_{corr}$  and  $(I_{K^+}/I_{Al^+})_{corr}$  to An and Or contents, respectively

Equation for least-squares regression line	Range of residuals	Correlation coefficient
$An(\text{mol } \%) = 97.0 - 142.1(I_{Na^+}/I_{Al^+})_{corr}$ [9.4] [27.5]	-0.68 ~ 0.74	-0.98
$f_{An}^{**} = 1.25 - 0.76(I_{Na^+}/I_{Al^+})_{obs}$ [0.07] [0.20]	-0.01 ~ 0.01	-0.97
$Or(\text{mol } \%) = 0.73 + 104.8(I_{K^+}/I_{Al^+})_{corr}$ [0.28] [33.0]	-0.46 ~ 0.29	0.95
$f_{Or}^{***} = 1.07 - 4.43(I_{K^+}/I_{Al^+})_{obs}$ [0.04] [0.92]	-0.01 ~ 0.02	-0.98

\* The numbers in square brackets below the regression coefficients are estimated standard deviations.

\*\*  $f_{An}$  is correlation factor for observed  $(I_{Na^+}/I_{Al^+})_{obs}$ , and

$$f_{An} = (I_{Na^+}/I_{Al^+})_{corr} / (I_{Na^+}/I_{Al^+})_{obs}$$

\*\*\*  $f_{Or}$  is correlation factor for observed  $(I_{K^+}/I_{Al^+})_{obs}$ , and

$$f_{Or} = (I_{K^+}/I_{Al^+})_{corr} / 1.074(I_{K^+}/I_{Al^+})_{obs}$$

Table 4. The results of in-depth and line analyses in the blue-, green-, yellow-, and red-iridescent labradorites from Nain, Labrador. The estimated standard deviations are given in brackets.  $An_{calc}$  and  $Or_{calc}$  are the calculated compositions from the measured lamellar thicknesses and bulk compositions

Method	Minor lamellae			Major lamellae			Specimen				
	An(mol %)	Or(mol %)	No. of lamellae measured	An(mol %)	Or(mol %)	No. of lamellae measured	$An_{calc}$	$Or_{calc}$	An(mol %)	Or(mol %)	Iridescence Color
<b>In-depth Analysis</b>											
	44.3[2.2]	2.8[0.2]	20	57.0[1.3]	1.9[0.1]	27	51	2	50.8	2.8	Blue
	42.7[5.7]	4.4[0.3]	8	57.8[8.4]	2.7[0.8]	21	53	3	52.8	2.6	Yellow
Mean	43.8(3.6)	3.3(0.8)	28	57.3(5.6)	2.3(0.7)	48					
<b>Line Analysis</b>											
	38.9[5.3]	2.7[1.2]	6	60.9[5.2]	1.7[0.5]	11	51	2	50.8	2.8	Blue
	45.9[0.0]	3.2[0.1]	2	56.6[1.0]	2.1[0.0]	2	52	3	52.8	2.6	Yellow
	45.7[4.0]	3.7[1.3]	15	61.1[4.1]	2.5[1.1]	11	58	3	58.1	3.3	Red
Mean	44.0(5.2)	3.4(1.3)	23	60.7(4.7)	2.1(0.9)	24					
<b>Mean values</b>											
	43.7(4.4)	3.3(1.1)	51	58.4(5.6)	2.2(0.8)	72					

\* Estimated standard deviations are in brackets.

where  $N_A$  and  $N_B$  are numbers of minor and major lamellae measured, respectively, and  $C$  is An or Or content. The bulk An and Or contents calculated using the above equation correspond to bulk compositions determined by EPMA within 0.9 percent in An content and 16 percent in Or content.

### Discussion

The data in Table 4 are consistent with the difference in composition of the two lamellae being at least 15 mole percent An and 1.2 mole percent Or. This composition difference is consistent with the results of Nissen *et al.* (1973), Cliff *et al.* (1976), and Miura *et al.* (1974, 1975). In this study the mean compositions of the two lamellae are  $An_{44(\pm 4)}Or_{3(\pm 1)}$  and  $An_{58(\pm 6)}Or_{2(\pm 1)}$  (Table 4).

Lamellar compositions, however, are likely to vary from one labradorite to another, in accordance with the report by Cliff *et al.* (1976). Variation in lamellar compositions of samples from the same locality and with similar cooling history depends mainly on the initial composition of the feldspar before exsolution. Table 4 shows that the An contents of the two lamellae are shifted slightly to larger An contents as the overall An content of the sample increases. This is consistent with the result of analytical electron microscopy by Cliff *et al.* (1976) that the thicker lamellae have more calcic compositions, and that the compositional differences of the lamellar pairs remain nearly constant through an area of a few square micrometers. The detection of potassium was also successful in this study. The sodic lamellae are a little enriched in potassium in all samples (Table 4).

In summary, the combination of IMA (IMA-2) and EPMA (JXA-50A) provides a technique that is valuable for determining the compositions of lamellar pairs.

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