Refinement of the Kudo-Weill Plagioclase Thermometer and Its Application to Basaltic Rocks

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Abstract. The Kudo-Weill plagioclase thermometer is applied to basaltic rocks. Comparison of measured and calculated crystallization temperatures shows that the latter are generally higher than measured values. The plagioclase thermometer predicts non-ideal behavoir of plagioclase solid solution and is in close agreement with experimental work at lower temperature. New equations are generated that relate plagioclase and melt chemistries to plagioclase crystallization temperatures; and these, combined with activity coefficient corrections, result in a refinement of the plagioclase thermometer.

Introduction

In his classic account of the melting behavior of plagioclase, Bowen (1913) demonstrated that albite ($NaAlSi_3O_8$) and anorthite ($CaAl_2Si_2O_8$) form a continuous solid solution series at high temperatures. He interpreted the experimental data in terms of an ideal solution model. In ideal solutions, melting temperatures can be related to mole fractions in coexisting phases by the Van't Hoff equation from enthalpies and fusion temperatures of the end members. However, attempts to relate plagioclase-liquid equilibria to plagioclase crystallization temperatures in complex multicomponent systems, such as magmas, require a knowledge of the activities of plagioclase constituents (Na, Ca, Al, Si) in the magmatic liquid. Such data are not available.

Using an empirical approach, Kudo and Weill (1970) derived an expression for the activity coefficients of these constituents in some granite and simple synthetic systems for which requisite compositional and melting data are available. They suggested that their resulting equations yield "reasonable" plagioclase crystallization temperatures when applied to magmatic systems representing a wide range of composition. In subsequent work, plagioclase crystallization temperatures calculated according to the Kudo-Weill plagioclase thermometer (Stormer and Carmichael, 1970; Brown and Carmichael, 1971) are almost invariably higher than the expected values. Similar disagreement has been found in this paper for rocks of basaltic composition. Based upon this application, this study offers an empirical refinement of the plagioclase thermometer which produces more realistic temperature estimates.

Thermodynamic Basis of the Plagioclase Thermometer

The thermodynamic basis of the Kudo-Weill thermometer is briefly summarized below. Reference should be made to the original work for a more detailed account (Kudo and Weill, 1970).
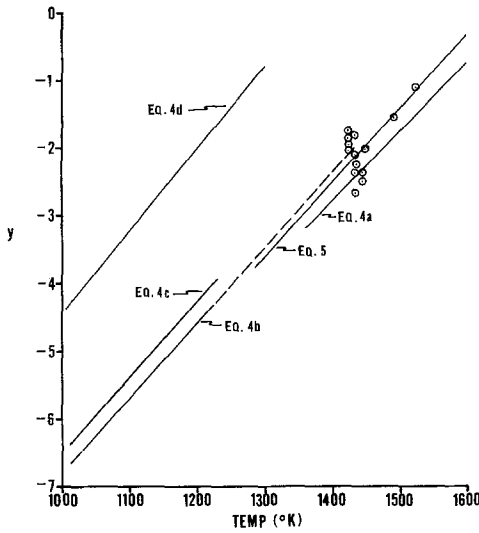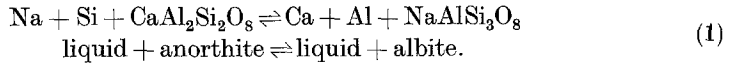
Fig. 1. Kudo and Weill (1970) linear regression curves for $y = \ln \lambda/\sigma + 1.29 \times 10^4 \, \phi'/T$ versus T for different conditions of crystallization. Eq. (5) is a graphical fit of the plotted data of Table 3 and assumes the slope of Eq. (4 b)

Plagioclase crystallizing from a melt can be represented by the exchange reaction

$$Na + Si + CaAl_2Si_2O_8 \rightleftharpoons Ca + Al + NaAlSi_3O_8$$
$$\text{liquid} + \text{anorthite} \rightleftharpoons \text{liquid} + \text{albite.} \tag{1}$$

Assuming plagioclase ideality, the equilibrium expression for this reaction is

$$-\frac{\Delta\mu^0}{RT} = \ln \lambda/\sigma + \ln \gamma_{Na} \, \gamma_{Si}/\gamma_{Ca} \, \gamma_{Al} \tag{2}$$

where $\mu^0 = \mu^0_{An} - \mu^0_{Ab} + \mu^0_{Na} + \mu^0_{Si} - \mu^0_{Ca} - \mu^0_{Al}$, $T = °K$, $\lambda = X_{Na} \, X_{Si}/X_{Ca} \, X_{Al}$, $\sigma = X_{Ab}/X_{An}$, $\gamma = $ activity coefficient, and subscripts denote the atomic fraction (X) of an element in the melt or mole fractions of Ab and An in plagioclase. It is asumed that the activity coefficients of Eq. (2) take the form of a regular solution model; that is, $\ln \gamma = C \phi / T$, where C is a constant empirically evaluated at $1.29 \times 10^{-4} \, T^{-1}$ and ϕ is the atomic fraction of the element in the melt. Substitution into Eq. (2) yields

$$y(T) = \ln \lambda/\sigma + 1.29 \times 10^4 \, \phi'/T \tag{3}$$

where ϕ' becomes $X_{Ca} + X_{Al} - X_{Si} - X_{Na}$. A plot of the right-hand side of Eq. (3) against T for experimental data yields a linear regression curve that may be used to calculate plagioclase crystallization temperatures from magma and coexisting plagioclase compositions. (Fig. 1) shows such a plot, and Kudo and Weill (1970) gave the following set of linear regression curves for different conditions of crystallization:

$$\ln \lambda/\sigma + 1.29 \times 10^4 \, \phi'/T = 10.34 \times 10^{-3} \, T - 17.24 \text{ (dry)} \tag{4a}$$
$$\ln \lambda/\sigma + 1.29 \times 10^4 \, \phi'/T = 11.05 \times 10^{-3} \, T - 17.86 \; (P_{H_2O} = 0.5 \text{ kb}) \tag{4b}$$
$$\ln \lambda/\sigma + 1.29 \times 10^4 \, \phi'/T = 11.14 \times 10^{-3} \, T - 17.67 \; (P_{H_2O} = 1.0 \text{ kb}) \tag{4c}$$
$$\ln \lambda/\sigma + 1.29 \times 10^4 \, \phi'/T = 12.18 \times 10^{-3} \, T - 16.63 \; (P_{H_2O} = 5.0 \text{ kb}). \tag{4d}$$

Table 1. Basaltic and andesitic rocks with their corresponding plagioclase crystallization temperatures calculated according to the Kudo-Weill (Eq. 4a) and refined (Eq. 7a) thermometer

(Sample numbers are those of the original references.)

Sample No.	Reference	Calculated plagioclase Crystallization temperature (°C)	
		Eq. (4a)	Eq. (7a)
—	[1, 2]	1205	1164
225	[3, 4]	1208	1161
190	[3, 4]	1232	1150
168	[3, 4]	1243	1157
116	[3, 4]	1246	1171
7	[3, 4]	1221	1152
F6	[5, 6]	1234	1163
6	[7]	1191	1201
8	[7]	1117	1149
10	[7]	1176	1188
14	[7]	1224	1216
16	[7]	1197	1213
1	[8]	1171	1148
2	[8]	1231	1200
3	[8]	1192	1202
5	[8]	1297	1213
7	[8]	1228	1210
8	[8]	1297	1212
D1	[9, 10]	1398	1290
26	[11]	1380	1281
28	[11]	1357	1256
42	[12]	1301	1208
31	[12]	1220	1222
45	[12]	1293	1200
78	[12]	1263	1205
65	[12]	1204	1204
44	[12]	1205	1212
70	[12]	1170	1183
71	[12]	1183	1177
F14	[5, 6]	1200	1143
S1	[5, 6, 13]	1181	1163
KI186	[14]	1170	1153
48589	[15]	1179	1154
57374	[15]	1166	1162
57364	[15]	1151	1159
57358	[15]	1129	1169
111549-7	[16]	1448	1353
111547-5	[16]	1315	1221
111547-13	[16]	1242	1154
P29166	[17]	1306	1208
P17171	[17]	1151	1109
P29114	[17]	1247	1169

Sources: [1] Wright and Fiske (1971); [2] Wright and Weiblen (1967); [3] Moore and Evans (1967); [4] Evans and Moore (1968); [5] Thompson and Tilley (1969); [6] Richter and Murata (1966); [7] Aoki (1967); [8] Muir and Tilley (1964); [9] Engel and Engel (1964); [10] Bottinga et al. (1966); [11] Carmichael (1964); [12] McBirney and Williams (1969); [13] Murata and Richter (1966); [14] Richter and Moore (1966); [15] Yoder and Tilley (1962); [16] Bryan et al. (1972); [17] Ewart (1969).

Test of the Plagioclase Thermometer

Forty-two basaltic and andesitic rocks for which published whole-rock chemistries and plagioclase compositions exist (Table 1) have been selected for use because they meet the following criteria:

1. Plagioclase compositional determinations are by electron microprobe or refractive index techniques. The most calcic plagioclase composition of each rock is reported and is the one used in the following analysis.

2. The rocks crystallized at or near the surface, either subaerially or in a submarine environment, so rapid cooling rates minimized reequilibration of initial crystals with successive liquid fractions. Some of the rocks are porphyritic, and in these cases, plagioclases probably began nucleating in a deeper environment. However, the effect of limited load pressure on plagioclase crystallization temperatures is probably minimal (Kudo and Weill, 1970).

3. No rock is of cumulate origin.

Seven rocks are calc-alkaline andesites or icelandites. The basalts exhibit wide chemical variation, from alkaline (hawaiites, etc.) to tholeiitic varieties. Plagioclase crystallization temperatures in lunar basalts have not been calculated because of the possibility of Na vaporization during crystallization (O'Hara *et al.*, 1970; Naughton *et al.*, 1971).

In order to test the Kudo-Weill thermometer, plagioclase crystallization temperatures for these rocks were calculated from Eq. (4a). Their distribution is presented in Fig. 2 A. The scatter of calculated temperatures may in part reflect errors in input data because the plagioclase thermometer is particularly sensitive to plagioclase composition (Kudo and Weill, 1970). Fig. 2 B gives the temperature distribution for 108 basalts and andesites for which measured temperatures are available. The corresponding data are listed in Table 2. In addition, measured or reliable estimates of plagioclase crystallization temperatures exist for 12 of the rocks in Table 1. These are listed in Table 3, along with their calculated and measured (or estimated) temperatures. Comparisons of the measured and calculated temperature data both in Fig. 2 A and B and in Table 3 show that the calculated temperatures are generally higher than the experimental values.

Divergence of Calculated Temperatures

Divergence of calculated from measured temperatures may result because Eq. (4a) is, in part, generated from plagioclase-liquid equilibria data from the Ab-An system. It is likely that the activity coefficients of melt components entering plagioclase in complex liquids differ from those in the simple binary system; thus, these data should not be used to calibrate the plagioclase thermometer.

It is appropriate, therefore, to calculate a new "dry" equation from the data of Table 3, which are plotted on Fig. 1. Their distribution does not allow rigorous definition of a new linear regression curve; however, if the slope of a new curve is taken as that of Eq. (4b), its y intercept can be derived graphically. The resulting equation is

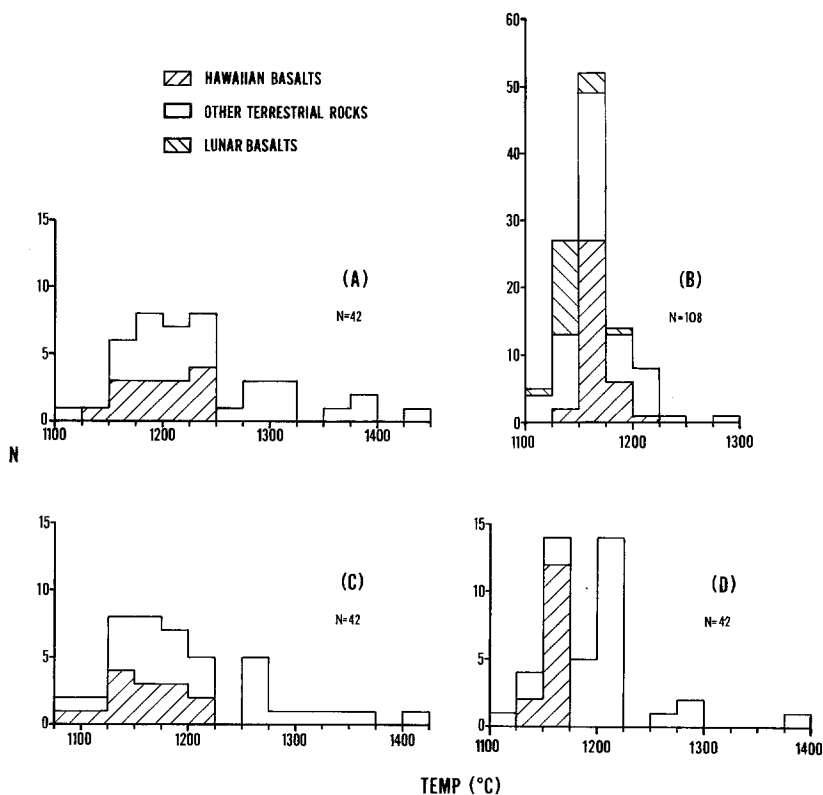$$\ln \lambda/\sigma + 1.29 \times 10^4 \, \phi'/T = 11.05 \times 10^{-3} \, T - 17.98. \tag{5}$$

Fig. 2A—D. Frequency distribution of temperatures for the beginning of crystallization of plagioclase for (A) rocks of Table 1, where T is calculated according to Eq. (4a); (B) rocks of Table 2, where T is measured; (C) rocks of Table 1, where T is calculated according to Eq. (5); and (D) rocks of Table 1, where T is calculated according to Eq. (7a)

The range of plagioclase crystallization temperatures calculated according to Eq. (5) (Fig. 2C) is in closer accord with experimental data than that calculated from Eq. (4a).

Refinement of the Plagioclase Thermometer; Non-Ideality of Plagioclase

The Kudo-Weill thermometer assumes that plagioclase behaves as an ideal solid solution at high temperatures. But there is no *a priori* reason to suggest that this should be so. Indeed, a recent investigation of plagioclase in chloride solutions (Orville, 1972) has shown that the solid solution exhibits a positive deviation from ideality at 700° C, 2000 bars P_{H_2O} in the presence of quartz. If plagioclase exhibits non-ideal behavior at higher temperatures, values of γ_{Ab}/γ_{An} for the data of Table 3 can be calculated by replacing σ with $(X_{Ab}\gamma_{Ab})/(X_{An}\gamma_{An})$. This assumes that the constants of Eq. (5) are correct and that the relatively narrow range of temperature

Table 2. Experimental temperatures for the beginning of plagioclase crystallization in basaltic and andesitic rocks
(Sample numbers are those of the original references.)

Sample No.	Reference	Experimental plagioclase crystallization temperature (°C)	Sample No.	Reference	Experimental plagioclase crystallization temperature (°C)
Terrestrial rocks			N	[8]	1215
40452	[1]	1165	Ha	[8]	1150
86120	[1]	1162	1801	[8]	1170
50035*	[1]	1165	X	[8]	1187
86186	[1]	1175	1	[8]	1185
AJ32	[2]	1167	2	[8]	1187
AJ35	[2]	1152	3	[8]	1187
AJ168	[2]	1157	4	[8]	1160
AJ33	[2]	1148	5	[8]	1140
AJ68	[2]	1145	6	[8]	1140
AJ102	[2]	1148	Ac	[8]	1180
AJ175	[2]	1156	St	[8]	1197
AJ99	[2]	1132	mb	[8]	1165
AJ76	[2]	1118	m	[8]	1130
AJ75	[2]	1122	bm	[8]	1110
AJ98	[2]	1132	t114	[8]	1145
AJ123	[2]	1119	t6	[8]	1170
—	[3]	1175	t364	[8]	1150
225[a]	[4]	1150	t125	[8]	1145
190[a]	[4]	1150	t230	[8]	1195
168[a]	[4]	1150	t627	[8]	1170
7[a]	[4]	1150	66123dcg	[9]	1180
F14	[5]	1159	6752mb3	[9]	1170
S1	[5]	1163	6751mb2	[9]	1165
48589	[5]	1170	111	[9]	1130
57374	[5]	1160	97	[9]	1130
57358	[5]	1160	95	[9]	1170
57364	[5]	1170	15	[9]	1155
65998	[5]	1240	1	[9]	1165
65992	[5]	1160	3	[9]	1125
60464	[5]	1185	S	[9]	1180
1(C)	[6]	1200	5	[9]	1155
F6	[7]	1159	324	[9]	1165
1959a	[7]	1159	B1	[10]	1208
1960L	[7]	1159	HAB	[10]	1220
1959b	[7]	1159	2	[10]	1198
M1	[7]	1157	14	[10]	1203
1960e	[7]	1159	19	[10]	1197
KIpb	[8]	1150	2	[10]	1210
KIg	[8]	1155	3S	[11]	1155
1921f	[8]	1155	5S	[11]	1155
1921ph	[8]	1155	7S	[11]	1155
1921t	[8]	1155	9S	[11]	1213
1920ph	[8]	1155	16K	[11]	1280
			20L	[11]	1245
			27V	[11]	1215

Table 2 (Continued)

Sample No.	Reference	Experimental plagioclase crystallization temperature (°C)	Sample No.	Reference	Experimental plagioclase crystallization temperature (°C)
Lunar basalts			Synthetic lunar basalts		
10017[b]	[12, 13]	1133		[17]	1180
12018[b]	[14, 15]	1149		[18]	1140
12020[b]	[14, 15]	1149		[19]	1130
12009	[16]	1140		[19]	1115
12021[b]	[14, 15, 16]	1133		[20]	1160
12022[b]	[15, 16]	1134		[21]	1140
12038[b]	[14, 15]	1154		[21]	1140
12040[b]	[14, 15, 16]	1146		[22]	1168
12052[b]	[14, 15]	1140			
12064[b]	[14, 15]	1140			
12065[b]	[14, 15, 16]	1135			

[a] Reliably estimated values (see original work for method).
[b] Average of values listed by several workers.

Sources: [1] Upton (1972); [2] Thompson and Flower (1971); [3] Wright and Fiske (1971); [4] Evans and Moore (1968); [5] Yoder and Tilley (1962); [6] Kudo and Weill (1970); [7] Thompson and Tilley (1969); [8] Tilley et al. (1966); [9] Tilley et al. (1967); [10] Tilley et al. (1968); [11] Brown and Schairer (1968); [12] Akimoto et al. (1971); [13] O'Hara et al. (1970); [14] Biggar et al. (1971); [15] Muan et al. (1971); [16] Green et al. (1971); [17] Muan and Schairer (1970); [18] Tuthill and Soto (1970); [19] Ringwood and Green (1972); [20] Smith et al. (1970); [21] Kushiro et al. (1970); [22] Newton et al. (1971).

Table 3. Rocks for which measured or reliable estimates of plagioclase crystallization temperatures (T_{exp}) exist and for which sufficient data are available to calculate temperatures. These also appear in Tables 1 and 2

Sample No.[a]	T_{exp}	Calculated temperatures (°C)		
		Eq. (4a)	Eq. (5)	Eq. (7a)
—	1175	1205	1169	1164
225	1150	1208	1174	1161
190	1150	1232	1196	1150
168	1150	1243	1206	1157
7	1150	1221	1195	1152
F6	1159	1234	1197	1163
F14	1159	1200	1165	1143
S1	1163	1181	1149	1163
48589	1170	1179	1144	1154
57374	1160	1166	1134	1162
57364	1160	1129	1100	1159
57358	1170	1151	1120	1169
Ab-An-Di[b]	1216	1246	1211	1257
Ab-An-Sp[b]	1248	1287	1249	1250

[a] See Tables 1 and 2.
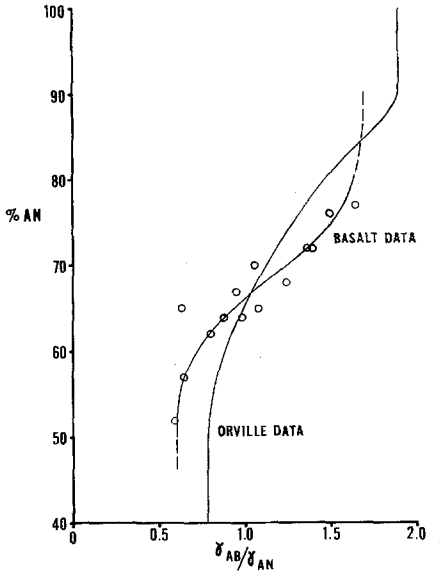[b] Data for the albite-anorthite-diopside and albite-anorthite-spinel systems are from Kudo and Weill (1970).

Fig. 3. Comparison of values of γ_{Ab}/γ_{An} as a function of plagioclase composition calculated for the data of Table 3, using Eq. (6) and T_{exp}, with the data of Orville (1972, p. 265)

for the onset of plagioclase crystallization in basalts does not significantly affect γ_{Ab}/γ_{An}. Rearrangement of Eq. (5) yields

$$\ln \frac{\gamma_{Ab}}{\gamma_{An}} = \ln \frac{\lambda}{X_{Ab}/X_{An}} + \frac{1.29 \times 10^4 \phi'}{T} - 11.05 \times 10^{-3} \, T + 17.98. \qquad (6)$$

Fig. 3 compares values of γ_{Ab}/γ_{An} calculated in this manner for the experimental data of Table 3 with similar values calculated from the data of Orville (1972). *Clearly, Eq. (6) predicts the same type of non-ideal behavior of plagioclase as indicated by Orville's measured data.* The ratios obtained from the basalt curve of Fig. 3 are here termed "activity coefficient corrections," and these are used below to generate new equations relating crystallization temperatures to compositions of coexisting plagioclase and melts. First, several points require consideration.

The shape of the basalt curve of Fig. 3, although not specific values of γ_{Ab}/γ_{An}, is essentially independent of the regression coefficients of Eqs. (5) and (6); that is, a similar curve, shifted either to the left or right, results by substituting the coefficients of Eq. (4a) or Eq. (4b), for example, into Eq. (6).

Secondly, it is conceivable that chemical characteristics of melts other than those elements involved in the exchange reaction of Eq. (1) exercise some control on plagioclase composition during crystallization. Or, more precisely, activity coefficients of Eq. (2) are assigned incorrect values in the Kudo-Weill analysis. However, a systematic search for some relationship between major elemental abundances and plagioclase compositions and values of λ/σ for the rocks of Table 1 revealed no bulk chemistry dependencies.

Thirdly, the above approach assumes that values of γ_{Ab}/γ_{An} are independent of variables, such as T and P_{H_2O}. Thus, calculations from Eq. (6) are not strictly quantitative. However, there is close agreement between the curves in Fig. 3 calculated for data at 700° C and 2000 bars P_{H_2O} and those where $T \approx 1125° \, C - 1225° \, C$
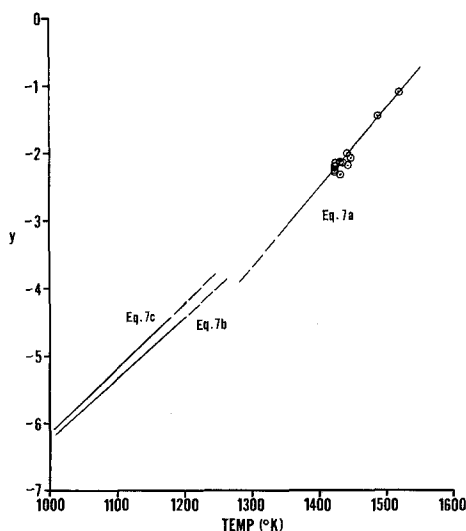
Fig. 4. Linear regression curves for $y =$ $\ln \lambda/\sigma' + 1.29 \times 10^4 \ \phi'/T$ versus T, where $\sigma' = (X_{Ab} \gamma_{Ab})/(X_{An}\gamma_{An})$. The plotted data are from Table 3

and P_{H_2O} is small. This implies that neither T nor P_{H_2O} significantly affect γ_{Ab}/γ_{An} or else that the effects of these variables oppose each other.

In the generation of the refined equations below, the differences in the curves of Fig. 3 are accepted as real and likely a reflection of the temperature difference. Thus, the activity coefficient corrections from the basalt curve are applicable to basaltic or andesitic rocks, where crystallization temperatures are high, whereas the Orville corrections may be applied to more acid systems. Therefore, Eq. (7a) is generated from the experimental data of Table 3 and the basalt curve of Fig. 3, and the granite data of Kudo and Weill (1970) for $P_{H_2O} = 0.5$ kb and 1 kb and the Orville corrections are used in the generation of Eqs. (7b) and (7c). These new equations of linear regression relating y and T (Fig. 4) are

$$\ln \lambda/\sigma' + 1.29 \times 10^4 \ \phi'/T = 11.76 \times 10^{-3} \ T - 19.01 \ \text{(basalt)} \qquad (7a)$$

$$\ln \lambda/\sigma' + 1.29 \times 10^4 \ \phi'/T = 9.87 \ \times 10^{-3} \ T - 15.21 \ (P_{H_2O} = 0.5 \text{ kb}) \qquad (7b)$$

$$\ln \lambda/\sigma' + 1.29 \times 10^4 \ \phi'/T = 9.60 \ \times 10^{-3} \ T - 15.76 \ (P_{H_2O} = 1.0 \text{ kb}). \qquad (7c)$$

The required input data are $\lambda = X_{Na}X_{Si}/X_{Ca}X_{Al}$ and $\phi' = X_{Ca} + X_{Al} - X_{Si} - X_{Na}$, where $X =$ atomic fraction of the element in the melt; and $\sigma' = X_{Ab}\gamma_{Ab}/X_{An}\gamma_{An}$, where X_{Ab}/X_{An} is the plagioclase composition and γ_{Ab}/γ_{An} the corresponding activity coefficient correction from the curves of Fig. 3.

Fig. 2D shows that the range of plagioclase crystallization temperatures in basalts calculated from Eq. (7a) is in close agreement with that of measured data. Furthermore, the general effect of applying Eqs. (7a)–(7c) instead of those of Kudo and Weill (Eqs. 4a–4c) is to lower calculated T. Thus, the difficulty alluded to by Stormer and Carmichael (1970) and Brown and Carmichael (1971) that the Kudo-Weill thermometer yields high T estimates is at least partially resolved.

Although their ranges are similar, the distributions of Fig. 2B and D are clearly different. Several points are pertinent here. Firstly, the basalts of Table 3, on which the activity correction is based, are all from Hawaii, and it may be that
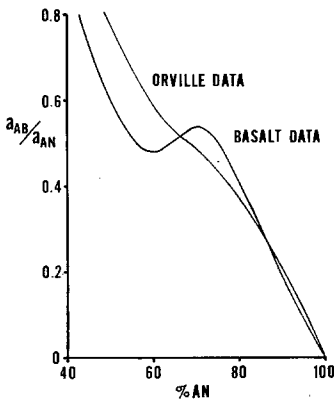
Fig. 5. Values of a_{Ab}/a_{An} corresponding to the curves of Fig. 3 plotted against plagioclase composition

12 rocks from one petrographic province do not precisely define the curve of Fig. 3. Secondly, 42 basalts may not be strictly representative of some larger population. This is indicated by Fig. 2 A and C, which suggest a bimodal distribution similar to that exhibited by Fig. 2 D. Thirdly, the activity coefficient corrections themselves predict a bimodal distribution. This is best illustrated by Fig. 5, in which activities are plotted against plagioclase composition. Such a plot is not a predictable result from the solidus of the Ab-An phase diagram of Bowen (1913). However, Smith (1972) has recently emphasized the lack of knowledge accompanying this system, and his suggested phase diagram exhibits several high-temperature complications. Fig. 5 may reflect the reequilibration statistics of metamorphosis between the high-temperature weakly disordered anorthite structure and that of intermediate plagioclase solid solution.

Conclusion

Application of the plagioclase thermometer to basaltic rocks predicts the same sort of non-ideal behavior for plagioclase solid solutions as that indicated by experimental work at 700° C. On this basis, an activity coefficient correction is calculated, and a new set of equations are defined that relate initial plagioclase crystallization temperature to plagioclase and melt compositions. Temperatures calculated according to these equations are in the same range (1125° C–1225° C) as measured values.

Attention is drawn to the fact that the refined plagioclase thermometer is strictly empirical. It may well suffer from the assumption of a regular solution model for the melt; and in view of the paucity of data on which it is based, it can only be said at present that it yields more reasonable temperature estimates than those calculated from the original equations of Kudo and Weill (1970). However, its apparent empirical success indicates that it will be of use until more data from natural rock systems become available and until the thermodynamics of plagioclase solid solutions and character of melts become better understood.

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