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COMPARATIVE GEOTHERMOMETRY OF SOME MAGNETITE–ILMENITE–ORTHOPYROXENE–CLINOPYROXENE ASSOCIATIONS FROM VOLCANIC ROCKS

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ABSTRACT

Four geothermometers [magnetite–ilmenite (Buddington & Lindsley 1964, Powell & Powell 1977), orthopyroxene–clinopyroxene (Wood & Banno 1973, Wells 1977), orthopyroxene–ilmenite and clinopyroxene–ilmenite (Bishop 1980)] have been applied to 120 pyroxene–Fe–Ti-oxide parageneses from volcanic rocks. The first two geothermometers yield "reasonable" magmatic temperatures, though discrepancies between them may reach 200°C. Temperatures obtained from the magnetite–ilmenite equilibrium, by various methods of calculation of the end members, differ by less than 30°C. Wells's formulation of the orthopyroxene–clinopyroxene geothermometer yields temperatures noticeably higher (60–80°C) than those given by the equation of Wood & Banno, which are in better agreement with the magnetite–ilmenite results. In their present form, the orthopyroxene–ilmenite and clinopyroxene–ilmenite methods do not seem adequate to estimate the temperatures of crystallization of volcanic rocks.

Keywords: Pyroxene (ortho-, clino-), Fe–Ti oxide (magnetite, ilmenite), mineralogical equilibrium, geothermometry, volcanic rocks.

SOMMAIRE

Quatre géothermomètres [magnétite–ilménite (Buddington & Lindsley 1964, Powell & Powell 1977), orthopyroxène–clinopyroxène (Wood & Banno 1973, Wells 1977), orthopyroxène–ilménite et clinopyroxène–ilménite (Bishop 1980)] ont été appliqués à 120 paragenèses à pyroxènes et oxydes de fer–titane de roches volcaniques. Les deux premiers géothermomètres permettent de calculer des températures plausibles pour des roches magmatiques, en dépit de divergences atteignant souvent 200°C. Les températures déduites de l'équilibre magnétite–ilménite, selon diverses méthodes de calcul des pôles, diffèrent entre elles de moins de 30°C. La formulation selon Wells du géothermomètre orthopyroxène–clinopyroxène aboutit à des températures notablement plus élevées (de 60 à 80°C) que celles obtenues selon l'équation de Wood & Banno; ces dernières sont en meilleur accord avec les résultats de l'équilibre magnétite–ilménite. Dans leur formulation actuelle, les géothermomètres orthopyroxène–ilménite et clinopyroxène–ilménite ne paraissent pas utilisables pour l'estimation des températures de cristallisation des roches volcaniques.

Mots-clés: Pyroxènes (ortho-, clino-), oxydes de fer–titane (magnétite, ilménite), équilibre minéralogique, géothermométrie, roches volcaniques.

INTRODUCTION

The association magnetite (titanomagnetite) – ilmenite (hemoilmenite) is common in volcanic rocks, especially in the TiO_2-rich alkali basalt series. Since the study of Buddington & Lindsley (1964), the Fe–Ti oxide geothermometer has generally been considered the most precise method available for estimating magmatic temperatures and oxygen fugacities (*e.g.*, Carmichael 1967, Anderson 1968, Wright & Weiblen 1968, Helz 1973). Considerable use has also been made of the orthopyroxene–clinopyroxene geothermometers (Wood & Banno 1973, Wells 1977), which have been successfully applied to volcanic rocks. Bishop (1980) recently studied the distribution of Fe^{2+} and Mg between coexisting ilmenite and pyroxene, and formalized their partitioning for use as geothermometers applicable to ilmenite–orthopyroxene and ilmenite–clinopyroxene associations. It is thus possible to test simultaneously four geothermometers in rocks containing titanomagnetite (Mt), hemoilmenite (Il), orthopyroxene (Opx) and clinopyroxene (Cpx) presumed in equilibrium. In this paper, 120 pyroxene–Fe–Ti-oxide associations have been considered for the purpose of evaluating the mutual consistency of these geothermometers and the constraints on their petrological applications.

METHOD OF INVESTIGATION

Samples selected

Table 1 gives a brief account of the sources of the analytical data used. Most analyses have been made with an automated Camebax microprobe and are unpublished results; others are from French theses. The results of analyses are

TABLE 1. ORIGIN OF THE MINERAL ASSOCIATIONS STUDIED

Series	Rock type	Geographical Location	Mt-Il temperatures (°C)	References
Alkali Basalt Series	Alkali Basalts and Basanites	French Massif Central	1032; 939; 889;	Maury 1976 , Maury et al. 1980
		French Polynesia	1025; 1123; 1064;	Maury et al. 1978, unpublished data
		Gulf of Guinea	961; 944;	Cornen & Maury 1980
	Hawaiites Mugearites Benmoreites	French Massif Central	1015; 997; 996; 974; 973; 966; 963; 952; 928; 915; 901; 883; 882; 877; 865; 857; 853; 844; 843; 836; 833; 832; 830; 816; 793; 787; 739; 738; 735; 710; 698;	Maury 1976 , Maury & Brousse 1978 ; Maury et al. 1980 , Vatin-Pérignon et al. 1980 , Bourdier 1980
		French Polynesia	1025; 966; 813;	Maury et al. 1978
		Gulf of Guinea	1078; 1058;	Cornen & Maury 1980
		Virunga (Central Afr.)	1028; 943; 853; 850; 838;	Pouclet (1976), unpublished
	Trachytes	French Massif Central	942;	Maury et al. 1980
		French Polynesia	1101; 875;	Maury et al. 1978 , Blanchard 1978
	Rhyolites	French Massif Central	954; 949; 939; 919; 905; 873; 867; 856; 831; 820;	Ménard 1979
Orogenic Series (Island arc tholeiitic, calcalkalic or shoshonitic)	Basalts	Mexico	1072; 1070; 962; 882; 808;	Silva Mora 1979
	Andesites	Mexico	1035;1012;984; 956; 919; 667; 657;	Silva Mora 1979
		New Hebrides	855; 812;	Unpublished
	Dacites	Lesser Antilles	1033;1024;925;919; 869;769;766;744; 742;729;708;706;683;665;664;662;647	D'Arco et al.(in prep.), Semet et al. (in prep.)
		Mexico	862; 857; 758; 727; 724;	Silva Mora 1979
	Shoshonitic Lavas	Argentina	1074; 1026; 1008; 980; 952; 889;	Deruelle 1979

Mt-Il temperatures are calculated using Anderson's (1968) method of evaluating proportions of end members.

available on request from the authors or from the Depository of Unpublished Data, CISTI, National Research Council of Canada, Ottawa, Ontario K1A 0S2. All sets of data include measurements of Si, Ti, Al, Cr, Fe, Mn, Mg, Ca, Na and K concentrations. In the microprobe analyses, working conditions used were 15 kV, 10–12 nA, counting time 6 seconds; contents lower than 0.1 wt. % are not considered representative. The mineral associations belong to rocks from the alkali basalt series of continental areas (Massif Central in France, Central Africa) and oceanic islands (French Polynesia, Gulf of Guinea) and from orogenic series of island arcs (Lesser Antilles, New Hebrides) or continental margins (Mexico, Argentina). The chosen associations fulfil the following requirements: (1) The presence of magnetite (titanomagnetite) and ilmenite (hemoilmenite) coexisting with preferably two but at least one pyroxene. (2) Accuracy of analytical results: the totals vary from 98.5 to 101.5 for pyroxenes (Fe expressed as FeO) and for oxide analyses recalculated following the method of Carmichael (1967). (3) Minerals belong to the same stage of crystallization of their host rock (groundmass, neighboring inclusions in phenocrysts, or phenocrysts presumed in equilibrium). Groundmass and phenocryst associations are considered separately. (4) Fe-Ti oxides must be described as fresh and unexsolved, yielding "reasonable" magmatic temperatures (Table 1) when calculated following the method of Buddington & Lindsley (1964). Of course, the possibility of subsolidus re-equilibration cannot be ruled out in some cases. (5) If minerals show noticeable compositional inhomogeneity and if there is an ambiguity in selecting the most appropriate composition, several combinations must be tried for a single rock.

A total of 120 associations were selected: 70 groundmass minerals and 50 phenocrysts or inclusions in phenocrysts. They include 34 magnetite (Mt) – ilmenite (Il) – orthopyroxene (Opx) – clinopyroxene (Cpx) associations, 16 Mt–Il–Opx and 70 Mt–Il–Cpx parageneses. The dominance of the last type is due to the fact that the greatest number of analyses available to the authors were from the alkali basalt series of the Massif Central (intermediate and differentiated lavas from the Chaîne-des-Puys, Mont-Dore and Cantal volcanoes), in which orthopyroxene is usually lacking.

Calculation procedures

A Fortran IV program was used to calculate the equilibration temperatures, following the equations given in Table 2, and to prepare graphical plots of the results. For Mt–Il geothermometry, the formulation of Powell & Powell (1977) was used. Fe^{3+} and Fe^{2+} were estimated following the method of Carmichael (1967). The proportions of Fe_2TiO_4 and Fe_3O_4 in titanomagnetite (Mt) and of Fe_2O_3 in hemoilmenite (Il) were calculated by both the methods of Carmichael (1967) and Anderson (1968). For Opx–Cpx geothermometry, the formulations of Wood & Banno (1973) and Wells (1977) were used (Table 2). The effect of total pressure (P) on the Opx–Il and Cpx–Il geothermometers (Bishop 1980) was purposely neglected.

TABLE 2. EQUATIONS USED FOR GEOTHERMOMETRY

Equilibria	Equations (T°C)	References
Magnetite Ilmenite	$T = \frac{8155}{4.59 + \mathrm{Ln}\ (X^{Mt}_{Fe_2TiO_4} X^{Il}_{Fe_2O_3}) / (X^{Mt}_{Fe_3O_4} X^{Il}_{FeTiO_3})} - 273$	Powell & Powell (1977)
Orthopyroxene	$T = \frac{-10202}{\mathrm{Ln}\ \frac{a^{Cpx}_{Mg_2Si_2O_6}}{a^{Opx}_{Mg_2Si_2O_6}} - 7.65 \left\lvert \frac{Fe^{2+}}{Fe^{2+}+Mg} \right\rvert^{Opx} + 3.88 \left\lvert \left\lvert \frac{Fe^{2+}}{Fe^{2+}+Mg} \right\rvert^{Opx} \right\rvert^{2} - 4.6} - 273$	Wood & Banno (1973)
Clinopyroxene	$T = \frac{7341}{3.355 + 2.44 \left\lvert \frac{Fe^{2+}}{Fe^{2+}+Mg} \right\rvert^{Opx} - \mathrm{Ln}\ \frac{a^{Cpx}_{Mg_2Si_2O_6}}{a^{Opx}_{Mg_2Si_2O_6}}} - 273$	Wells (1977)
Orthopyroxene Ilmenite	$T = \frac{1646 + 1634 \left\lvert \frac{Fe^{2+}}{Fe^{2+}+Mg} \right\rvert^{Il} + 0.0124P}{\mathrm{Ln}\ \frac{(Mg/Fe)^{Opx}}{(Mg/Fe)^{Il}}} - 273$	Bishop (1980)
Clinopyroxene Ilmenite	$T = \frac{748 + 2745 \left\lvert \frac{Mg}{Mg+Fe^{2+}} \right\rvert^{Cpx} + 817 \left\lvert 2 \left\lvert \frac{Fe^{2+}}{Fe^{2+}+Mg} \right\rvert^{Il} - 1 \right\rvert + 0.0124P}{\mathrm{Ln}\ \frac{(Mg/Fe)^{Cpx}}{(Mg/Fe)^{Il}}} - 273$	Bishop (1980)

a : activities, T : temperatures (°C).

This assumption is roughly valid in the case of volcanic groundmass associations, which usually are considered to have equilibrated under $P = 1$ bar for thermodynamic calculations (Carmichael *et al.* 1974). In the case of phenocrysts, a systematic error is introduced by neglecting P. However, the coefficients affected by P in Bishop's (1980) equations are very small (0.0124 and 0.0106) with respect to the usual values (3–4) of the terms ln $[(Mg/Fe)^{Opx}/(Mg/Fe)^{Il}]$ and ln $[(Mg/Fe)^{Cpx}/Mg/Fe)^{Il}]$ (Table 2). Thus an increase in pressure from 0 to 5 kbar (the latter value being a relatively high pressure for the crystallization of phenocrysts from volcanic rocks) produces a corresponding temperature increase of less than 20°C, which is negligible compared with the discrepancies found among the geothermometers (see below). The corollary is that Opx–Il and Cpx–Il equilibria cannot be used as geobarometers in their present formulation.

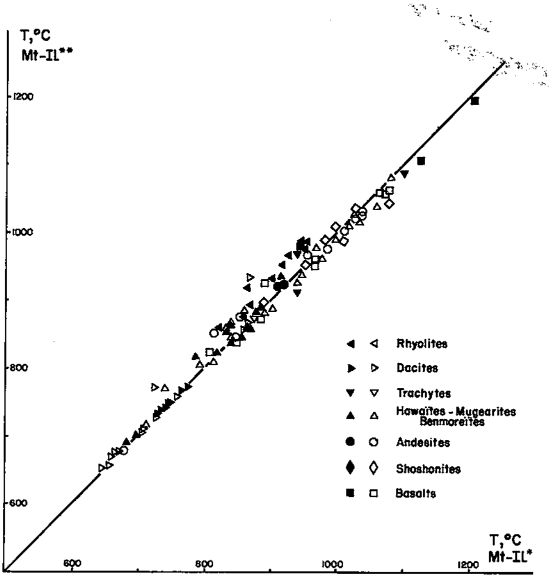

FIG. 1. Mt–Il geothermometry. Temperatures (**) following Carmichael (1967) *vs.* temperatures (*) after Anderson (1968). Solid symbols, phenocrysts; open symbols, groundmass.

RESULTS

Mt–Il geothermometry

The results (labeled C and A, respectively), obtained using the methods of Carmichael (1967) and Anderson (1968) of calculating the molar fractions of ulvöspinel in Mt and hematite in Il, are compared in Figure 1. There is excellent agreement between them. The temperatures obtained for a given pair usually differ by less than 30°C from one another: this is the limit of precision of the geothermometer (Buddington & Lindsley 1964, Powell & Powell 1977) for minerals departing only slightly from

TABLE 3. TYPICAL ANALYSES AND CORRESPONDING CALCULATED TEMPERATURES

PHENOCRYST	K - MUGEARITES				K - BENMOREITES			
	Cpx	Opx	Il	Mt	Cpx	Opx	Il	Mt
SiO_2	51.63	52.78	*	*	51.63	53.24	*	*
TiO_2	1.04	0.36	50.09	17.37	1.04	0.34	49.86	16.83
Al_2O_3	2.68	1.03	0.26	2.35	2.68	1.08	0.20	7.81
Fe_2O_3			6.14	29.11			6.34	29.28
Cr_2O_3	*	*	*	*	*	*	*	*
FeO	9.09	17.09	40.59	49.90	9.09	16.83	41.41	41.77
MnO	0.31	0.41	0.64	0.43	0.31	0.47	0.52	0.54
MgO	15.61	25.59	2.24	0.93	15.61	25.77	1.76	3.72
CaO	19.43	1.76	*	*	19.43	1.72	*	*
Na_2O	0.28	*	0.19	0.38	0.28	*	0.27	0.32
K_2O	*	*	*	*	*	*	*	*
TOTAL	100.07	99.02	100.17	100.49	100.07	99.45	100.28	100.29
T °C	Mt - Il^1 : 837				Mt - Il^1 : 859			
	Mt - Il^2 : 843				Mt - Il^2 : 830			
	Opx - Cpx^3 : 1039				Opx - Cpx^3 : 1038			
	Opx - Cpx^4 : 1069				Opx - Cpx^4 : 1067			
	Cpx - Il : 729				Cpx - Il : 669			
	Opx - Il : 643				Opx - Il : 588			
GROUNDMASS	K - MUGEARITES				K - BENMOREITES			
	Cpx	Opx	Il	Mt	Cpx	Opx	Il	Mt
SiO_2	44.89	52.82	*	*	52.90	50.78	*	*
TiO_2	4.11	0.17	44.09	7.92	0.38	0.19	47.90	7.36
Al_2O_3	7.37	0.55	0.13	0.64	1.28	0.25	0.15	1.08
Fe_2O_3			14.45	51.92			9.00	53.90
Cr_2O_3	0.33	*	*	0.17	*	*	*	0.09
FeO	7.03	20.65	36.28	36.07	7.51	25.41	38.19	36.30
MnO	0.13	1.04	0.72	0.72	0.78	1.67	1.14	0.61
MgO	11.94	22.41	1.53	0.75	15.77	20.32	2.15	0.77
CaO	22.95	1.57	*	*	22.16	1.74	*	*
Na_2O	0.40	*	*	*	0.59	*	*	*
K_2O	*	*	*	*	0.05	0.17	*	*
TOTAL	99.15	99.21	97.20	98.19	101.42	100.48	98.53	100.16
T °C	Mt - Il^1 : 802				Mt - Il^1 : 715			
	Mt - Il^2 : 793				Mt - Il^2 : 710			
	Opx - Cpx^3 : 752				Opx - Cpx^3 : 894			
	Opx - Cpx^4 : 720				Opx - Cpx^4 : 925			
	Cpx - Il : 630				Cpx - Il : 604			
	Opx - Il : 697				Opx - Il : 816			

These analyses are from the French Massif Central (Maury 1976). Number beside geothermometer association indicated the method used : 1 : Carmichael's, 2 : Anderson's, 3 : Wood & Banno's, 4 : Wells'.
* below limit of sensitivity.

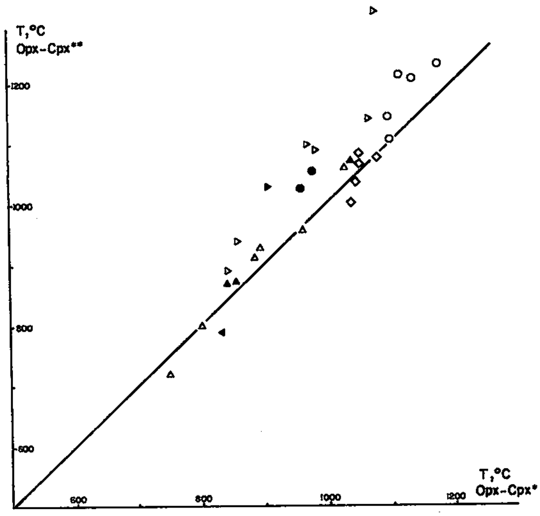

FIG. 2. Opx–Cpx geothermometry. Temperatures (**) following Wells (1977) *vs.* temperatures (*) after Wood & Banno (1979). Symbols as for Figure 1.

the system FeO–Fe_2O_3–TiO_2. In Tables 1 and 3 and in the corresponding diagrams (Figs. 3, 4, 5 and 6), Anderson's method has been chosen because it discards the minor elements to reduce natural compositions to pure Fe–Ti oxide, and because the influence of Mn, Mg and Al on the calibration of Buddington & Lindsley (1964) is incompletely understood.

It appears from Figure 1 and Table 1 that the range of temperatures encountered is wholly compatible with the generally accepted temperatures of crystallization of volcanic rocks. However, some values appear to be low (particularly those lower than 700°C for andesites and dacites). They can be taken to indicate probable re-equilibration during subsolidus cooling. The data plotted in Figure 1 show a fairly good correlation between temperatures and rock types. The highest temperatures (> 900°C) are observed for basalts, shoshonites, andesites and trachytes; associations from rhyolites and dacites indicate temperatures usually below 900°C. This should be considered when examining Figures 3 to 6, where Mt–Il temperatures are plotted as a function of those given by other geothermometers.

Opx–Cpx geothermometry

The agreement between the results obtained by the methods of Wood & Banno (1973) and Wells (1977) is reasonably good (Fig. 2). The latter gives temperatures systematically higher than the first. The discrepancy commonly reaches 60–80°C, especially for temperatures above 900°C. The difference appears to be greater for groundmass minerals than for phenocrysts. The range of temperatures encountered (800–1200°C) is reasonable for the volcanic rocks under consideration. Among other possible origins, these discrepancies may be chemical, reflecting the fact that the Wood–Banno geothermometer has been calibrated mostly by reference to basaltic compositions and that of Wells predominantly by reference to ultramafic compositions.

Comparison of Mt–Il and Opx–Cpx thermometers

The correlations between the results of these two geothermometers are not good (Figs 3, 4). The Opx–Cpx temperatures are usually higher than Mt–Il temperatures. The differences observed commonly reach 200°C, and are larger if Wells's equation is used (Fig. 4) than with that of Wood & Banno (Fig. 3). Again, greater discrepancies are found for groundmass minerals, perhaps owing to the important compositional variations in groundmass pyroxenes. In fact, comparisons between the two-pyroxene and two-oxide data are difficult. A substantial part of the discrepancy may reflect the fact that pyroxenes begin to crystallize at higher temperatures than oxides. So we must temper our conclusions concerning the differences observed for phenocrysts, which may be recording successive stages of crystallization. However, the groundmass pyroxene–Fe–Ti-oxide association unambiguously corresponds to the same stage of the rock's history. It is suggested that the more important discrepancies (100°C or more) observed in this last case may well reflect subsolidus re-equilibration (specially for oxides, which re-equilibrate more readily than the pyroxenes); it cannot be proved that the accuracy of the geothermometers is responsible for these differences.

Comparison of Cpx–Il and Mt–Il thermometers

A very rough positive correlation is apparent (Fig. 5), but the Cpx–Il temperatures are, in most cases, lower than the Mt–Il ones; the discrepancy commonly reaches 200°C or more. About one third of the Cpx–Il temperatures are lower than 650°C and thus cannot be considered plausible for volcanic rocks. Discrepancies exceeding 400°C are found in several cases; they are considerably higher than the differences observed between Mt–Il and Opx–Cpx results for the same parageneses, indicating problems of accuracy with the Cpx–Il method.

Comparison of Opx–Il and Mt–Il thermometers

Results obtained using these two methods do not show a systematic relationship (Fig. 6). Approximately one third of the Opx–Il temperatures are lower than 650°C; discrepancies equal to or higher than 400°C with respect to the Mt–Il temperatures are common and can be interpreted as in the previous case.

Other correlation diagrams (Opx–Il *vs.* Cpx–Il, Cpx–Il *vs.* Opx–Cpx, Opx–Il *vs.* Opx–Cpx) are not presented here; they show a lack of correlation between the geothermometers, though some rare values may be in mutual agreement.

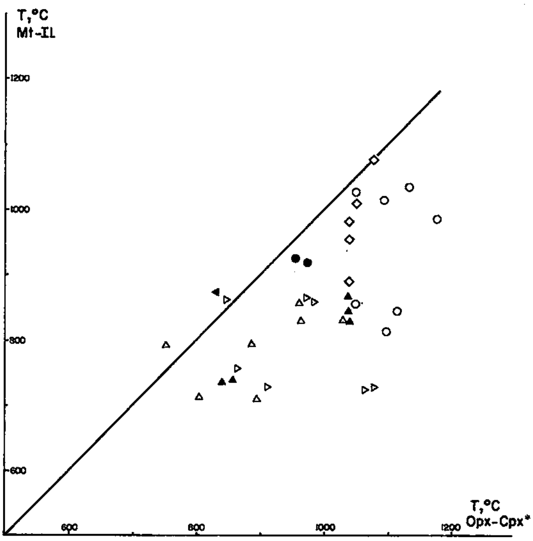

FIG. 3. Plot of Mt–Il temperatures (Anderson's method) *vs.* Opx–Cpx according to Wood & Banno (1973). Symbols as for Figure 1.

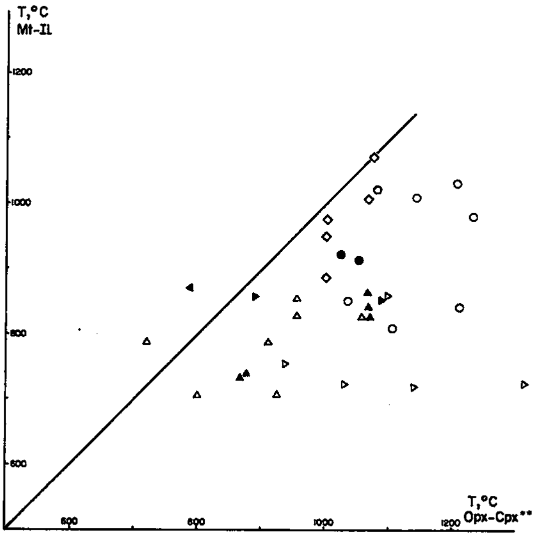

FIG. 4. Plot of Mt–Il temperatures (Anderson's method) *vs.* Opx–Cpx temperatures (Wells's formulation). Symbols as for Figure 1.

DISCUSSION

The results presented above are disappointing and raise doubts as to the practical applications of geothermometers to natural parageneses. The formulations of the geothermometers cannot be considered as the unique causes of the observed discrepancies. Many errors can originate in questionable equilibria between mineral phases, especially with respect to the presum-

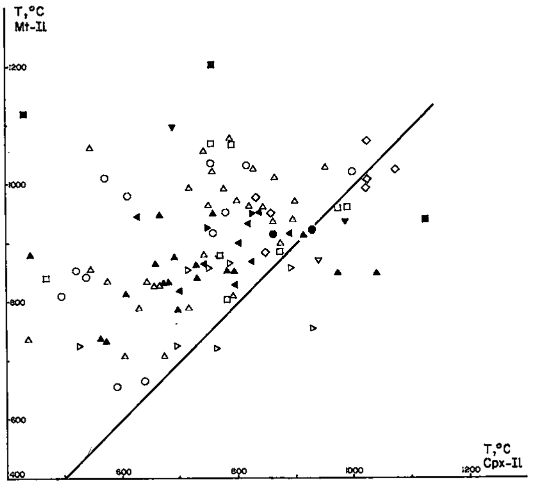

FIG. 5. Plot of Mt–Il temperatures (Anderson's method) *vs.* Cpx–Il temperatures. Symbols as for Figure 1.

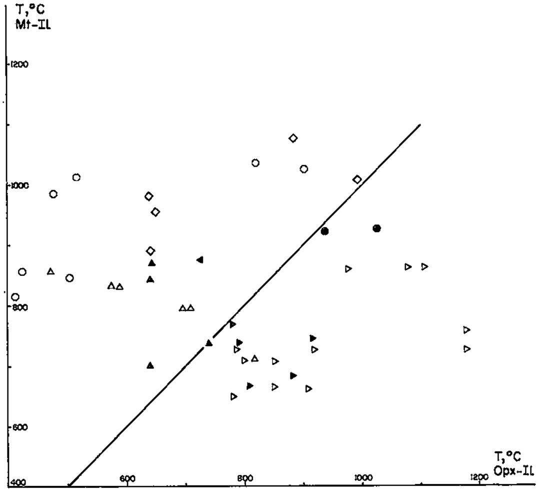

FIG. 6. Plot of Mt–Il temperatures (Anderson's method) *vs.* Opx–Il temperatures. Symbols as for Figure 1.

ably different temperatures at which equilibration ceases for pyroxenes and Fe–Ti oxides during cooling. Another source of error may be the choice of the analyses considered, in view of the usual compositional variations. An important source of error can also result from the poorly understood role of minor components in solid-solution models, especially in the case of the Fe–Ti oxides. In his application of the Cpx–Il and Opx–Il geothermometers to natural assemblages, Bishop (1980) also found significant discrepancies (reaching 200°C) with evaluations based on the Mt–Il or Opx–Cpx thermometers.

Table 3 presents some results obtained with the four geothermometers considered. Even if the temperatures obtained are in mutual agreement in some cases, it appears that the Opx–Il and Cpx–Il geothermometers (Bishop 1980) cannot be considered reasonable indicators of magmatic temperatures. Therefore, their extensive use in petrological studies cannot be recommended. However, they could be possible indicators of equilibrium where they give results similar to those obtained from other geothermometers. Mt–Il and Opx–Cpx methods give "reasonable" results, in good mutual agreement (discrepancies smaller than 100°C) in about one third of the cases studied. The Wood & Banno (1973) formulation of the Opx–Cpx equilibrium shows the best agreement with the Mt–Il data, better than that of Wells (1977). Some Mt–Il temperatures appear to be low with respect to the commonly accepted temperatures of crystallization of lavas; re-equilibration during subsolidus cooling may have played an important role, even where no petrographic indication of their occurrence has been found. Even in favorable cases, it is difficult to determine the temperature of crystallization of a given mineral phase with a precision better than ± 50°C. This conclusion implies serious limitations on the applications to natural rocks of thermodynamic calculations based on geothermometry (*e.g.*, Carmichael *et al.* 1974, Luhr & Carmichael 1980), for which a precise knowledge of temperature is required.

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