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b_0 of muscovite in low and high variance assemblages from low grade Verrucano rocks, Northern Apennines, Italy.

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Abstract

The regional distribution of the muscovite b_0 parameter as function of metamorphic grade and assemblages in the Verrucano rocks of the Northern Apennines has been investigated. The equation relating b_0 to R.M. (MgO/M.W.+FeO_{tot}/M.W.) for the Verrucano muscovite is $b_0 = 8.982 + 0.499$. R.M. with $r = 0.88$. The mean b_0 values increase from pyrophyllite or kyanite-bearing rocks (9.002 Å) to K-feldspar-bearing rocks (9.056 Å and 9.042 Å) for pyrophyllite and kyanite grade respectively. In the pyrophyllite or kyanite as well as in the K-feldspar-bearing rocks (low variance assemblages) the mean muscovite b_0 values are very similar in all Verrucano areas and their standard deviations are small. On the contrary, in muscovites from pyrophyllite and K-feldspar-free samples (high variance assemblages) the mean b_0 values vary within the different Verrucano areas and show a high standard deviation.

The muscovite b_0 values in Al-rich low variance assemblages of Verrucano and other metamorphic areas with different P/T regime of metamorphism are discussed in relation to their use as a potential qualitative geobarometer.

Keywords: b_0 of muscovite, mineral assemblages, geobarometer, Verrucano rocks, Northern Apennines.

Introduction

Over the last twenty years many studies have shown that the b_0 parameter of K-white mica is particularly useful for characterizing metamorphism in the low grade region. SASSI and SCOLARI (1974) first attempted to calibrate the muscovite b_0 parameter as a qualitative geobarometer in ms + ab + qtz ± chl ± carb ± grp (mineral abbreviations as in KRETZ, 1983; other abbreviations used in the text: carb = carbonates, grp = graphite, k-fs = K-feldspar, su = sudoite) high variance assemblages for low grade metamorphic rocks. Even though it was suggested a long time ago (GUIDOTTI and SASSI, 1976), very little effort has been spent on calibration of a geobarometer based on the b_0 of muscovite from low variance assemblages.

This paper describes the variation of the muscovite b_0 parameter in low grade metamorphic rocks of the Verrucano metasediments (Northern Apennines, Italy) as function of metamorphic grade, mineralogical assemblage and muscovite composition.

The aim of this study is to establish some constraints on the variation of b_0 in a kyanite-type low temperature metamorphism and to lay the foundations for an empirical b_0 reference scale for the muscovite from low variance assemblage in the model system KNaASH (K_2O-Na_2O-Al_2O_3-SiO_2-H_2O), GUIDOTTI and SASSI (1976).

The Verrucano rocks are particularly suitable for this purpose because they show different degrees of Al-saturation and contain muscovite showing a wide range of chemical variation mainly in the phengite and ferrimuscovite components (FRANCESCHELLI et al., 1986).

Regional Setting

The Verrucano metasediments crop out in discontinuous patches along the entire length of the Northern Appenines (Fig. 1). For details of the geology and stratigraphy of the Verrucano sequences refer to GELMINI (1969), RAU and TONGIORGI (1974), TONGIORGI et al. (1977), CASSINIS

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et al. (1979), COSTANTINI et al. (1985). Mineralogy, geochemistry and general aspects of the Verrucano metamorphism have recently been discussed by FRANCESCHELLI et al. (1986, 1987, 1989).

From those papers, some general features useful for the present discussion, are summarized below.

The Verrucano sequence is mainly composed of quartzites, metapsammites, metaconglomerates and minor metapelites metamorphosed and deformed during the Alpine orogeny. The rocks show weak to moderate schistosity. In thin section the texture is dominated by the superimposition of two main schistosities (S1 and S2, FRANCESCHELLI et al., 1986). Generally only S1 and subordinately S2 schistosity is associated with mineral crystallization.

The regional distribution of Al-silicates such as kaolinite, pyrophyllite and kyanite were taken as a basis for the metamorphic zonation of the Verrucano rocks in the Northern Apennines. Four metamorphic zones have so far been distinguished: ka, (Perugia II boreholes), ka + prl + qtz transition (Monte Argentario, Monticiano-Roccastrada ridge and Monti Leoni pro-parte), prl + qtz (Monti Leoni, Monticiano-Roccastrada ridge, Monti Pisani and Punta Bianca areas) and ky + qtz zones (Massa area).

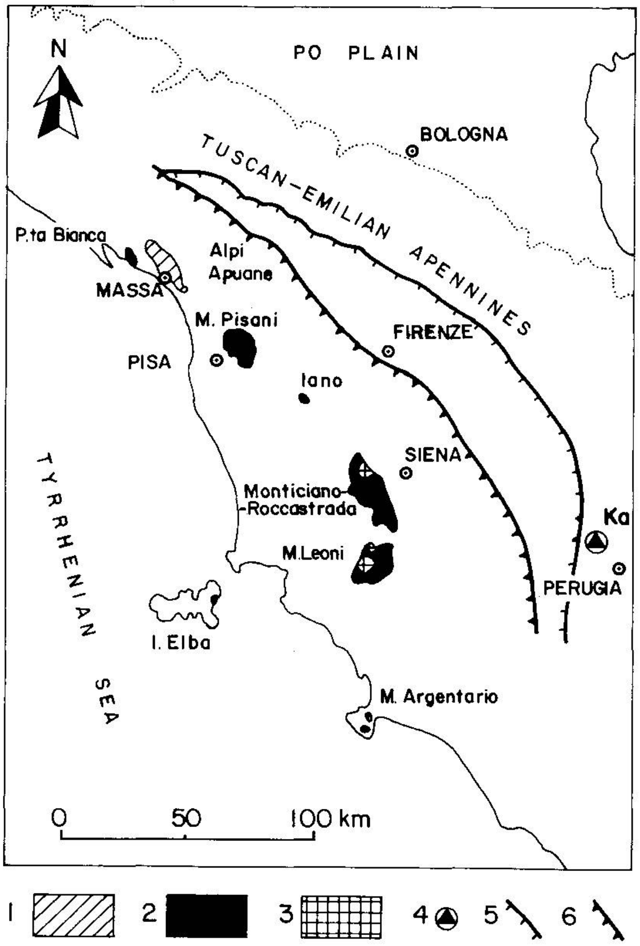

Fig. 1 Sketch map showing the metamorphic zonation of the Verrucano of the Northern Apennines and the geographical distribution of the localities sampled. (1) ky + qtz zone, (2) prl + qtz zone, (3) ka + prl + qtz zone, (4) ka + qtz zone boreholes, (5) Front of the Tuscan Nappe, (6) Front of the Cervarola flysch.

In these zones the pressures and the temperatures of metamorphism have been estimated by FRANCESCHELLI et al. (1986) as ranging from 3 to 5 kb and from 300° to 450°C respectively.

Sample selection and analytical techniques

This study covers rocks characterized by a great variety of lithology and bulk chemistry in order to assess the variation of b_0 of muscovite with mineral assemblages and metamorphic grade. The samples studied have been subdivided into three assemblage groups showing different degrees of Al-saturation. They are:

Al-rich:
pyrophyllite (or kyanite)-bearing assemblages
ms + qtz + prl ± su ± cld
ms + qtz + ky ± chl ± pg ± cld

Al-intermediate:
pyrophyllite (or kyanite) and K-feldspar free-assemblages
ms + qtz ± chl ± pg ± ab ± ctd

Al-poor:
K-feldspar-bearing assemblages
ms + qtz + k-fs ± ab ± chl

Accessory minerals are magnetite, Ti-oxides, ilmenite, hematite, pyrite, zircon, epidote.

The d 060 spacing of muscovite was measured using a Philips PW 1793 diffractometer. The d 060 was determined on polished slabs of rock cut parallel to the cleavage using quartz as an internal standard, and b_0 was calculated as 6 · d 060 for Cu K_α or $K_{\alpha 1}$. The standard deviation (1σ) of a single d 060 measurement is estimated to be ± 0.0005 Å.

Most of the muscovites examined show the d 060 reflection being strongly asymmetric or partially split into two peaks. The asymmetry or splitting degree was more pronounced in celadonite-rich muscovite. As shown in Fig. 2, when the Cu K_β radiation is used the d 060 peak loses its partial splitting. This is the best evidence to demonstrate that the asymmetry or splitting of the 060 peak is due to the $K_{\alpha 1}/K_{\alpha 2}$ doublet.

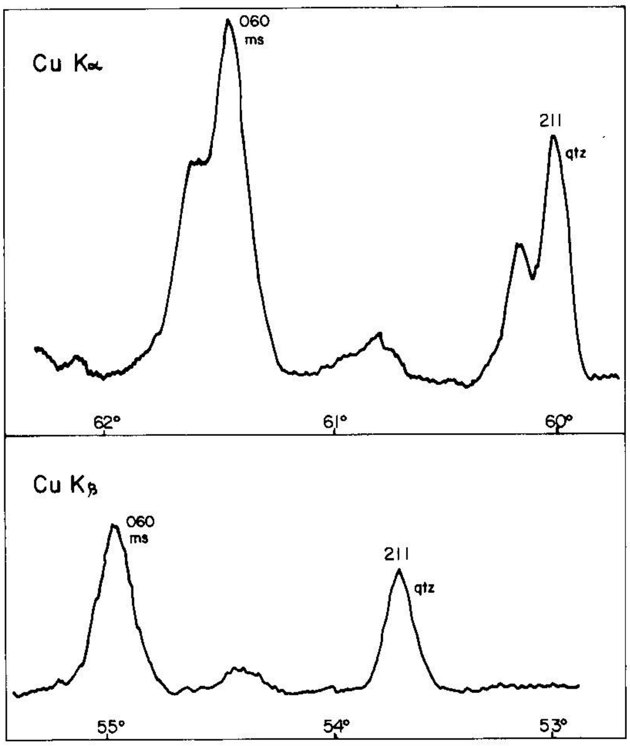

Fig. 2 XRD traces of Cu K_α and Cu K_β of 060 peak of muscovite and 211 peak of quartz. See text for explanation.

Analytical results

REGIONAL DISTRIBUTION OF b_0 VALUES

The regional distribution of the muscovite b_0 values in the Verrucano rocks is shown in Fig. 3. The data are presented as a function of metamorphic grade.

For the pyrophyllite zone histograms representative for Monti Leoni, Monticiano-Roccastrada, Iano, Monti Pisani and Punta Bianca outcrops are also given. In all the histograms a further subdivision was made according to the three groups previously defined: pyrophyllite-bearing, pyrophyllite and K-feldspar free and K-feldspar bearing assemblages.

In the ka + prl + qtz transition zone (Monte Argentario area) the muscovite b_0 values range from 8.990 to 9.045 Å with two maxima value densities at 9.005 and 9.040 Å (BERTAGNINI and FRANCESCHELLI, 1982).

The muscovite b_0 spacing in samples of the prl + qtz zone range from 8.995 to 9.061 Å, with three maxima value densities around 9.008, 9.025 and 9.055 Å. Within this zone the range of b_0 values of each outcrop is as follows: Monti Leoni 9.000 to 9.045 Å, Monticiano-Roccastrada area 8.990 to 9.050 Å, Punta Bianca 9.005 to 9.045 Å, Monti Pisani 8.995 to 9.065 Å, Iano 8.990 to 9.040 Å.

In the Verrucano rocks of the ky + qtz zone (Massa Unit) the muscovite b_0 values range from 8.995 to 9.050 Å with a maximum density value close to 9.005 Å.

In each metamorphic zone or outcrop a close relationship between the b_0 values and assemblage type was found. The b_0 values in the pyrophyllite or kyanite-bearing samples range from 8.990 to 9.010 Å and no significant variations of the mean values in the three metamorphic zones seem to exist. In K-feldspar-bearing samples the values range from 9.045 to 9.065 Å and from 9.035 to 9.050 Å in the prl + qtz (Monti Pisani) and ky + qtz (Massa Unit) zones respectively.

K-feldspar bearing rocks, in the ky + qtz zone, occur in the Paleozoic sequence of the Massa Unit. However during the Alpine orogeny these rocks experienced the same metamorphic history as the Triassic Verrucano sequence.

Furthermore we observed that b_0 values from pyrophyllite and K-feldspar-bearing assemblages never overlap and that muscovites from pyrophyllite and K-feldspar-free assemblages shows widely varying intermediate b_0 values.

b_0 AND R.M. RELATIONSHIP IN MUSCOVITE

The b_0 and R.M. (R.M. = MgO/M.W. + FeO_{tot}/M.W.; M.W. = molecular weight) relationship for the Verrucano muscovites is shown in Fig. 4. The R.M. values were calculated assuming all the iron to be in the divalent state.

Table 1 gives the MgO/M.W., FeO/M.W., 2 Fe_2O_3/M.W., R.M. and b_0 values of some selected Verrucano muscovites from M. Pisani and Monticiano-Roccastrada ridge. Chemistry of these muscovite will be discussed in detail in an other paper (BALDELLI et al. in preparation).

On the assumption of a linear dependence of b_0 on R.M., the following equation was computed by a least square method: b_0= 8.982 + 0.499 R.M. with a linear correlation coefficient r = 0.88. FREY et al. (1983) found a similar relationship directly relating the d (060+$\bar{3}$31) spacing to the R.M. values (d [060+$\bar{3}$31] = 1.4976 + 0.0819 R.M.). Expressing the R.M. as a function of b_0, FREY's equation transforms into b_0 = 8.986 + 0.491 R.M. The equation relating the b_0 to R.M. (defined as [MgO/M.W. + FeO/M.W. + 2 FeO/M.W.] by CIPRIANI et al. (1968) was: b_0 = 8.990 + 0.327 R.M. The slight differences between CIPRIANI's, FREY's and our own equations may be due to various factors.

Apart from the different method of calculating the R.M. values, it is well known that the b_0 musco-

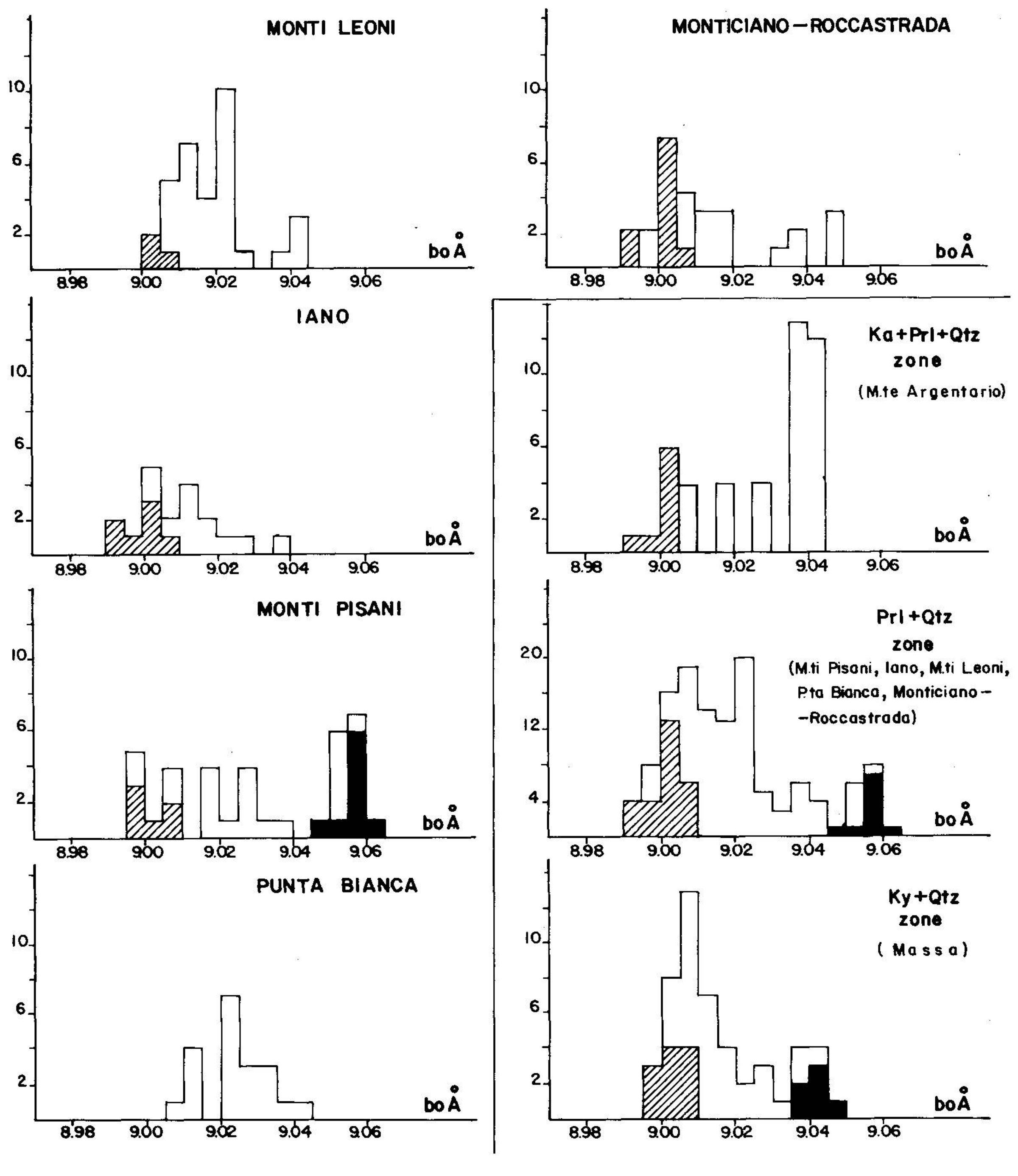

Fig. 3 Histograms showing the regional distribution of muscovite b_0 values in the Verrucano of the Northern Apennines; hatched: pyrophyllite(or kyanite)-bearing samples; white: pyrophyllite (or kyanite) and K-feldspar-free samples; black: K-feldspar-bearing samples. On the right the b_0 spacing of muscovite are arranged according to the metamorphic grade.

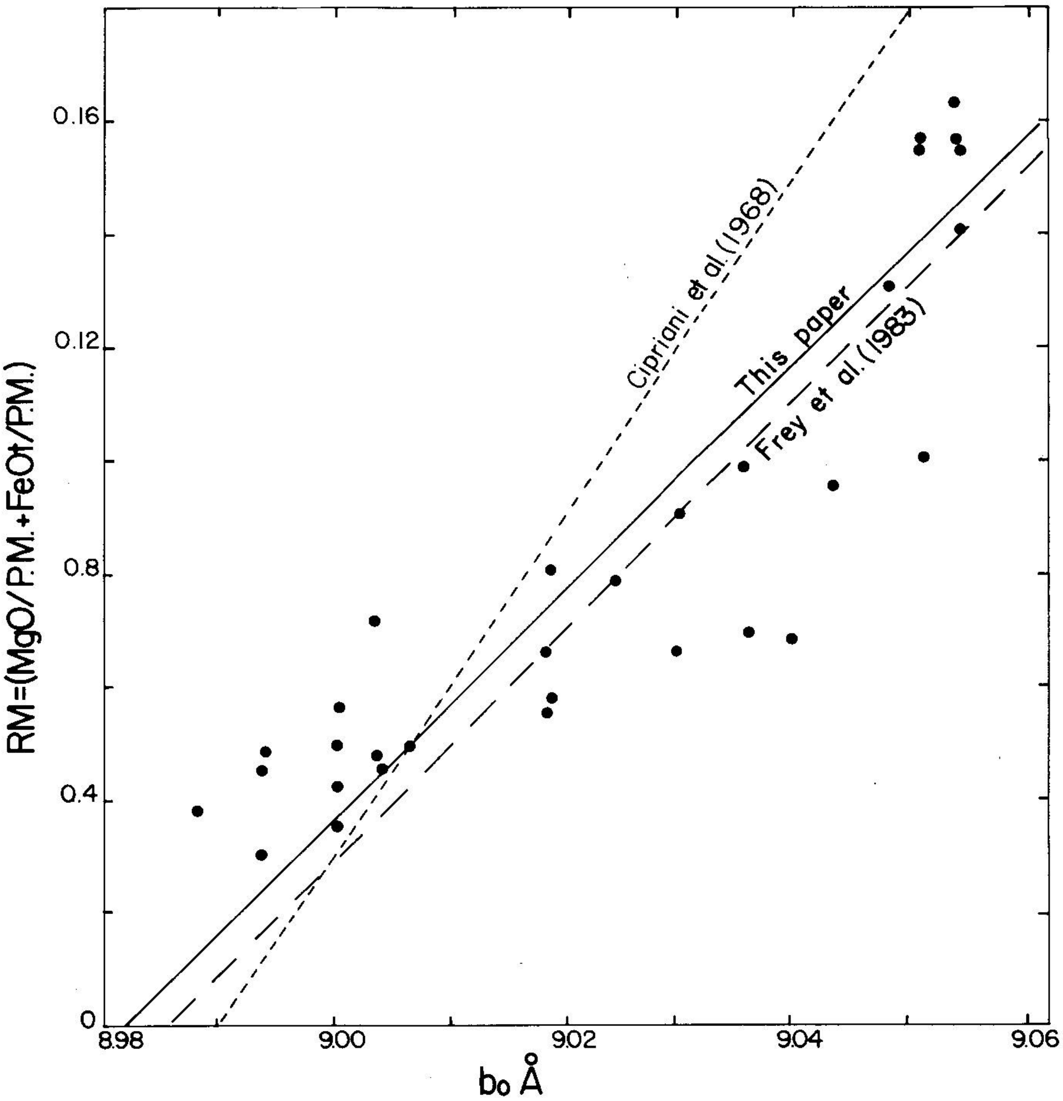

Fig. 4 Correlation of b_0 and R.M. Dotted and broken lines represent the equation of CIPRIANI et al. (1968), FREY et al. (1983) respectively. The best fit line to 32 data of Verrucano muscovite is also shown.

vite parameter is not only (Mg + Fe_{tot}) dependent, but is also somewhat influenced by the Na content (CIPRIANI et al., 1968) and by the ordering status (VELDE, 1980). Besides, despite their different ionic radius, usually no distinction among Mg, Fe^{2+} and Fe^{3+} cations is made.

For twelve muscovites whose analyses include Fe^{2+} and Fe^{3+} determinations (Tab. 1) (BALDELLI et al., in prep.) the contribution of the (Mg + Fe^{2+}) (celadonite) and Fe^{3+} (ferrimuscovite) contents were separately evaluated. The linear equation computed by a least square fitting method was: $b_0 = 8.982 + 0.548\ (Mg + Fe^{2+}) + 0.522\ (Fe^{3+})$. This regression has been calculated assuming only (Mg + Fe^{2+}) and Fe^{3+} as variables; 8.982 is the value resulting from the previous equation. This equation, compared with the one previously reported, seems to suggest that, in the range of composition considered here (see Tab. 1), the celadonite and ferrimuscovite components similarly influence the b_0 parameter.

The collected data indicate that the b_0 parameter of the K-white mica may be considered a good measure of the octahedral sheet composition (the R.M. value of CIPRIANI et al., 1968) for the Verrucano muscovites.

Discussion

b_0 IN LOW AND HIGH VARIANCE ASSEMBLAGES

In Tab. 2 the mean b_0 values and standard deviation of muscovite from low and high variance as-

Tab. 1 b_0, MgO/M.W., FeO/M.W., 2 Fe_2O_3/M.W. and R.M. values of some selected muscovites from Monti Pisani and Monticiano-Roccastrada ridge. Chemical data and mineral assemblages from BALDELLI et al. in prep.) and FRANCESCHELLI et al. (1986).

Samp.	2 Fe_2O_3/P.M. x 10^{-3}	MgO/P.M. x 10^{-3}	FeO/P.M. x 10^{-3}	R.M. x 10^{-3}	bo (Å)	Mineral assemblages
1	6.2	36.0	15.3	57.5	8.998	ms+qtz+prl+su+pg?
2	20.2	31.7	10.2	62.1	9.000	ms+qtz+prl+cld
3	10.8	27.0	13.2	51.0	8.998	ms+qtz+prl+su
4	30.0	46.6	10.7	87.3	9.000	ms+qtz+prl
5	26.0	40.7	6.9	73.4	9.000	ms+qtz+su+pg?
6	30.2	47.6	14.3	92.1	9.030	ms+qtz+chl
7	21.0	44.1	4.1	69.2	9.040	ms+qtz+chl+ab
8	48.4	56.6	3.4	108.4	9.052	ms+qtz
9	82.4	70.9	4.2	157.5	9.054	ms+qtz+ab
10	72.0	72.7	15.1	159.8	9.054	ms+qtz+chl?+ab+k-fs
11	82.0	76.4	5.6	164.0	9.054	ms+qtz+chl+ab+k-fs
12	77.6	69.0	8.4	155.0	9.055	ms+qtz+ab+k-fs

semblages (in the KNaASH system) are given for each metamorphic zone of the Verrucano complex.

The mean b_0 values of muscovite from high variance assemblages are 9.032, 9.022 and 9.015 Å for the Ka + qtz + prl, prl + qtz and Ky + qtz zones respectively. According to SASSI and SCOLARI (1974) these values are typical of a medium-pressure type metamorphism despite the fact that many samples lack the peculiarity required for a correct application of the b_0 geobarometer (absence of albite, presence of magnetite and/or hematite, etc., according to GUIDOTTI and SASSI, 1976). For muscovites from high variance assemblages the most relevant features of the b_0 data reported in Tab. 2 are the wide range of the mean b_0 value variation and the great dispersion of values within the same zone or outcrop. In the pyrophyllite zone the mean b_0 values of Al-intermediate samples ranges from 9.013 Å (Iano) to 9.018 Å (Monti Leoni), 9.020 Å (Monticiano-Roccastrada ridge), 9.024 Å (Punta Bianca), 9.040 Å (M. Pisani).

Such a variation cannot be explained by invoking different metamorphic conditions but rather by the wide range of muscovite composition. This in turn is dependent upon the bulk chemistry variation of the Verrucano metasediments. In such a case, in fact, the mean b_0 value and the associated standard deviation for a given outcrop are largely determined by the bulk chemistry variations of a certain lithotype, by the representativity of the lithotype in the field and, finally, by the frequency of sampling.

In the Verrucano metasediments the mean muscovite b_0 value in the Al-intermediate group, as shown in Fig. 3, is actually obtained by averaging values covering the entire b_0 range from low to high pressure-type metamorphism (SASSI and SCOLARI, 1974).

On the contrary, when the influence of bulk chemistry is reduced (i.e. in low variance assemblages), the mean muscovite b_0 values are very similar in all the Verrucano areas and their standard deviations are considerably smaller.

Tab. 2 Average b_0 values of muscovite from Al-rich, Al-intermediate and Al-poor groups for Verrucano rocks as a function of metamorphic grade. Figures in brackets represent the estimated standard deviation in terms of least units cited for the value to their immediate left.

Type of assemblage	Key mineral	bo av.value	Metamorphic zone	locality
Al -rich	prl	9.001(8)	ka+prl+qtz.	Mt. Argentario
	prl	9.001(8)	prl+qtz	(M.Pisani, Monticiano -Roccastrada, Iano, Punta Bianca, Monti Leoni)
	ky	9.001(4)	ky+qtz	Massa
Al-intermed	-	9.032(14)	ka+prl+qtz	Mt.Argentario
	-	9.022(16)	prl+qtz	(M.Pisani, Monticiano-Roccastrada, Iano, Punta Bianca, Monti Leoni)
	-	9.015(11)	ky+qtz	Massa
Al-poor	k-fs	9.056(5)	prl+qtz	Monti Pisani
	k-fs	9.042(6)	ky +qtz	Massa

In the prl + qtz zone, where it is possible to compare data from different outcrops, the mean b_0 values are substantially the same, about 9.002 Å for all the outcrops.

For the Al-poor low-variance assemblages, b_0 values from different outcrops of the same metamorphic zone are not available. In the prl + qtz (Monti Pisani) and ky + qtz zone (Massa Unit) the mean b_0 values of muscovites from Al-poor assemblages are 9.056(5) and 9.042(6) Å respectively.

In this case the slight decrease in the b_0 values may be related to the increase in metamorphic grade. It is well known that an increase in temperature reduces the ($Mg + Fe^{2+}$) solid solution field in muscovite (GUIDOTTI and SASSI, 1976).

INTERREGIONAL COMPARISON OF b_0 FROM LOW VARIANCE ASSEMBLAGE

It is interesting to analyze the muscovite b_0 values from low variance assemblages in low, medium and high pressure metamorphism.

Unfortunately, few studies mention the assemblages present in samples for which muscovite b_0 data are given. We have compiled (Tab. 3) some of the available data from Iberian Massif and Sierra Baza (Spain), Grande Kabylie (Algery), Calabria, Northern Apennines and Ligurian Alps (Italy), Glarus Alps (Central Swiss Alps), Crete (Greece). From the data of Tab. 3 it can be observed that:

i) In Al-rich low-variance assemblages (pyrophyllite or kyanite-bearing rocks) the mean b_0 values vary from 8.954(5) (Hercynian metamorphites of Iberian Massif, Spain) to 8.996(6) Å (Hercynian metamorphites of Calabria, Italy and Sierra Baza, Spain) in the low-pressure type metamorphism.

ii) In the medium pressure type metamorphism (Verrucano metamorphites of the Northern Apennines and anchimetamorphic black shales of Glarus Alps) the mean b_0 values are 9.002(4) and 8.982(7) respectively. As commented by FREY, "The reason

Tab. 3 Comparison of b_0 muscovite mean values from Al-rich low variance assemblages from several metamorphic areas with different P/T metamorphic facies series. Figures in brackets represent the estimated standard deviation in terms of least units cited for the value to their immediate left. N = number of analyzed samples.

locality	facies series	age of metamorphism	Key mineral	bo	N	source
Iberian M.	low	Hercynian	prl	8.954(5)	30	BRIME (1985)
Iberian M.	low	Hercynian	prl	8.961(4)	10	BRIME (1985)
G.Kabylie	low	Hercynian	prl	8.981(8)	6	BOSIERE et al.(1982)
Calabria	low	Hercynian	prl	8.996(6)	57	COLONNA et al.(1979)
Sierra Baza	low-medium	Hercynian	prl	8.996(6)	28	GOMEZ-PUGNAI-RE et al.
Glarus Alps	medium	Alpine	prl	8.982(7)	38	FREY (1978) and written comm.
Northern Apennines	medium	Alpine	prl	9.002(3)	43	this paper
Northern Apennines	medium	Alpine	ky	9.002(4)	11	this paper
Ligurian Alps	high	Alpine	prl	9.010(7)	4	this paper
Crete	high	alpine	prl	9.013(7)	2	this paper

for the rather low b_0 values in shales from Glarus Alps could be due to the paragonite content in muscovite of these rocks".

iii) In the pyrophyllite-bearing rocks from high pressure type metamorphism of the Ligurian Alps and Crete the $b_0$mean value is 9.010(7) and 0.013(7) Å respectively. SEIDEL (1977) gives an average value of about 9.024 Å for Al-rich rocks (chloritoid schists) from Crete.

The data presented in Tab. 3 clearly show that the mean b_0 values in the Al-low variance assemblages tend to increase hand in hand with the P/T regime of metamorphism.

One could derive the following relation between the b_0 average values in Al-low variance assemblages and the character of the metamorphism:

$b_0 < 9.000$ Å	low pressure
$9.000 < b_0 < 9.010$ Å	medium pressure
$b_0 > 9.010$	high pressure

This is a first step towards the calibration of the b_0 geobarometer scale in ms + prl (ky) + qtz (plus a Mg,Fe silicate) assemblages. Nevertheless they cannot yet be used as a reference scale; other data are necessary to control its general validity especially in high pressure rocks.

The data given here seem to encourage the study of b_0 in low variance assemblages (either in Al-rich or Al-poor) as a workable qualitative geobarometer even if the b_0 variations due to pressure in Al-rich low variance assemblages appear to be too modest and comparable with the analytical error.

Conclusion

In the Verrucano metamorphites of the Apennines the mean b_0 values increase in the following order: Al-rich, Al-intermediate, Al-poor assemblages. In both Al-rich and Al-poor low variance assemblages the muscovite b_0 spacing forms two distinct groups. In these assemblages the mean b_0 values change only slightly in different outcrops and/or metamorphic zones of the Verrucano and the values are characterized by a small standard deviation. Muscovites from Al-intermediate high variance assemblages, on the other hand, show highly variable b_0 values. The mean values in each outcrop or metamorphic zone have a high standard deviation.

The pattern of b_0 variation in muscovites, as it results from this study, seems to suggest that the use of this parameter to evaluate the P/T gradient of metamorphism, referring to high variance assemblages only, requires a great deal of attention. From the data given here, it clearly emerges that the mean b_0 values of muscovite from high variance assemblages may be very different even in rocks metamorphosed under the same P/T gradient, therefore any comparison among different areas may prove to be thwarting.

On the other hand, when low variance assemblages are considered the b_0 method of estimating the P/T gradients in metamorphic rocks from low grade regions is more reliable.

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