# Significance of illite crystallinity and $b_o$ values of K-white mica in low-grade metamorphic rocks, North Hill End Synclinorium, New South Wales, Australia

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ABSTRACT. A study of low-grade metamorphism in late Silurian to early Carboniferous rocks in the North Hill End Synclinorium and adjacent anticlinoria has been made by the determination of illite crystallinity and  $b_o$ values of K-white mica in eighty slates and phyllites. Illite crystallinity values vary from 0.40  $\Delta^{\circ}2\theta$  on the Molong Anticlinorium to 0.12  $\Delta^{\circ}2\theta$  within the axis of the synclinorium, suggesting anchizonal to epizonal metamorphic conditions. This is in agreement with previous observations on Ca-Al-hydrosilicate assemblages which indicated a change from prehnite-pumpellyite facies in the anticlinoria adjacent to the synclinorium to middle greenschist facies in the axis. Local variations in crystallinity are attributed to variation in  $a_{\rm K}^+$  in fluids migrating along cleavage zones.

The mean  $b_o$  value obtained from the pelites is 9.017 Å ( $\sigma n = 0.008$ ; n = 80) which is in close agreement with that obtained from part of the adjacent Capertee Anticlinorium ( $\bar{x} = 9.019$  Å;  $\sigma n = 0.007$ ; n = 52). However, 't' tests indicate that two  $b_o$  populations are present in the synclinorium ( $\bar{x} = 9.010$  and 9.022 Å), with the lower values concentrated in the southern portion of this structure. The two populations are considered to be the result of slightly different metamorphic conditions prevailing during the deformation of the rocks in the synclinorium. A higher geothermal gradient affecting rocks giving the lower  $b_o$  values is attributed to the presence of granitoids at shallower depths than elsewhere in the synclinorium.

KEYWORDS: illite, mica, metamorphic rocks, North Hill End Synclinorium, New South Wales, Australia.

THIS study has been carried out on a sequence of marine and terrestrial sediments and intermediate to silicic volcanic rocks of late Silurian to early Carboniferous age which occur in the north-eastern portion of the Lachlan Fold Belt (fig. 1). Part of the sequence was deposited in the Hill End Trough, an ensialic graben active from the late Silurian until the (?)early Middle Devonian. Subsequently, these rocks were mildly deformed during the Middle Devonian Tabberabberan Orogeny and overlain unconformably by a late Devonian to early Carboniferous succession. A major cleavage-forming and metamorphic event followed in the Carboniferous, producing the major structures in the area, the Hill End Synclinorium and the Molong and Capertee Anticlinoria (Cas, 1983; and references therein; fig. 1).

The metamorphic rocks produced during this event have been the subject of earlier studies. Smith (1969) delineated five metamorphic zones in the Molong Anticlinorium and Hill End Synclinorium, which in order of increasing grade are; albite-quartz (Z1), prehnite (Z2), prehnite-pumpellyite (Z3), actinolite (Z4) and biotite (Z5). He showed that the highest grade rocks (Z5) were located in the axis of the synclinorium. The position of the biotite zone boundary was later modified by Prendergast (1981) who found evidence for biotite appearing at a slightly lower grade in quartzo-feldspathic rocks than in pelitic rocks. Textural studies by Vernon and Flood (1979) and Prendergast (1981) indicated that biotite grew during the early and late stages of deformation and after deformation had ceased.

Subsequently, a metamorphic study involving illite crystallinity (Kubler, 1966; Kisch, 1980) and  $b_o$  measurements (Sassi and Scolari, 1974; Padan *et al.*, 1982) on K-white micas, was carried out in the Windamere Dam-Cudgegong area (fig. 1; Offler



FIG. 1. Location of Hill End Synclinorium and adjacent anticlinoria and study area; shaded area, Windamere Dam to Cudgegong area. Inset Location of synclinorium in the Lachlan Fold Belt.

and Pemberton, 1983). The illite crystallinity (IC) studies indicated upper anchizonal to lower epizonal conditions and  $b_o$  values ( $\bar{x} = 9.019$  Å,  $\sigma n = 0.007$ , n = 52) suggesting baric conditions higher than that proposed by Smith (1969) for rocks in the Molong Anticlinorium. As a result of this work, the  $b_o$  study was extended to the Hill End Synclinorium and the Molong Anticlinorium to ascertain whether: (a) the pattern in these areas was similar to that in the Cudgegong-Windamere Dam area; (b) the  $b_o$  values reflect the high heat flow that should have existed in this area where rifting, extensive silicic volcanism and post tectonic granitoid emplacement took place.

IC studies were also carried out to show whether they could be used to delineate zones of progressive metamorphism in an area containing epizonal, greenschist-facies rocks. Adequate data could be obtained from the synclinorium but not from the Molong Anticlinorium, because of the lack of K-white-mica-bearing rocks. All rocks that were studied contain abundant metamorphic white mica and in some, detrital muscovite is a minor phase. Microprobe analyses indicate that in most instances this phase has chemically re-equilibrated during metamorphism (Prendergast, 1981). The results and interpretation of this investigation are the subject of this paper.

#### Experimental methods

The illite crystallinity (IC) measurements were made on polished slabs of slate and phyllite cut parallel to cleavage and on the <2  $\mu$ m fraction. The <2  $\mu$ m fraction was obtained from approximately 10 g of sample crushed in a siebtechnik disc mill for 20 sec and dispersed in sedimentation cylinders. After the appropriate settling period, the <2  $\mu$ m fraction was drained off and sedimented on to a glass slide. Scans were carried out over the  $2\theta$  range 7.5 to 10° at  $\frac{1}{2}$ °  $2\theta$ /min; divergence and scatter slits of 1° and receiving slits of 0.2 mm were used. Peak width was calculated in terms of  $\Delta$ °2 $\theta$  and samples from the lower grade zones were glycolated and heated at 550 °C to ascertain the presence of other components interlayered with illite.

The rocks used for  $b_{o}$  measurements were of pelitic composition and contained the non-limiting assemblage quartz-albite-muscovite-chlorite (Guidotti and Sassi, 1976). Carbonaceous material was present in most samples. Rocks containing excessive amounts of chlorite or quartz were not analysed as  $b_0$  values obtained from K-white micas in these rocks can result in misleading interpretations of the baric conditions (Guidotti and Sassi, 1976). The analyses were made on polished rock slabs cut perpendicular to cleavage or on powdered samples packed into aluminium mounts of the same design as that of Robinson (1981). The range 58-63° was scanned at  $\frac{1}{4}$ ° 2 $\theta$ /min and  $b_0$  determined from the (060) peak using the (211) quartz reflection as an internal standard. Mean  $b_o$  was calculated from six determinations for each specimen. The settings were as follows: slits 2°, 0.1 mm, 2°, goniometer speed 0.5°  $2\theta$ /min, chart speed 20 mm/min and time constant 3.

All IC and  $b_o$  analyses were carried out on a Philips automated PW 1732/10 X-ray diffractometer using Cu-K $\alpha$  radiation, graphite monochromator and 40 kV, 30 mA. Machine drift was not taken into account for the IC study (cf. Primmer, 1983). However, drift appears not to be a problem, as tests carried out with standards, subsequent to the completion of the study, showed it to be insignificant.

#### Results and discussion

Illite crystallinity (IC). The analysis of the  $< 2 \mu m$  fraction of the samples reveals that the highest IC values (and therefore the lowest crystallinities) occur in the anticlinorial regions (fig. 2) where prehnite-pumpellyite facies assemblages have been noted (Smith, 1969; Offler and Pemberton, 1983), and lowest values within the biotite zone delineated by Prendergast (1981). If 0.21  $\Delta^{\circ}2\theta$  is taken as the lower boundary of the epizone (Kisch, 1980), then most IC values are epizonal. The results obtained from rock slices of the same specimens are



FIG. 2. Location of samples and their IC values ( $< 2 \mu m$  fraction) in  $\Delta^{\circ}2\theta$ . Full heavy lines delineate metamorphic zones (Z3-4) of Smith (1969); heavy dashed lines, biotite zone boundaries of Prendergast (1981).



FIG. 3. Histograms of IC values from rock slices and  $< 2 \mu m$  fraction of specimens from the actinolite zone (Z4) and biotite zone (Z5). Dashed lines, samples east of biotite zone, full lines, west of biotite zone.

similar but a greater concentration of low IC values is apparent in the biotite zone (fig. 3). Thus the IC results are in accord with the petrographic observations of Smith (1969) and Prendergast (1981) which indicated that grade increased towards the axial zone of the synclinorium.

It is clear, however, that the IC pattern in fig. 2 is too irregular to allow the delineation of zones of decreasing IC values. Much of the irregularity may have been reduced had further samples been taken. However, closer sample spacing has shown that this feature still exists. This is exemplified by the IC values obtained from rocks SSW of Hill End (230 894, fig. 2) and in the Windamere Dam to Cudgegong area ( $\bar{x}_{IC} = 0.23$ ,  $\sigma n = 0.04$ , n = 42). Why this irregularity exists, even on a small scale, is not completely understood. Temperature perturbations over distances small enough to explain the local variations in IC, are unlikely. Similarly, changes in IC due to interlayered smectite or paragonite, which are known to affect IC (Kisch, 1980), can be discounted because contents of these components in the white micas are insignificant. There remain three further possible explanations, tectonic strain, permeability and chemistry of the host rock (Frey et al., 1980; Kisch, 1980). Although several attempts have been made to determine whether there is any relationship between illite crystallinity and tectonic strain, no conclusive evidence has been produced (Frey et al., 1980). However, recent microstructural studies on cleavage formation have shown that cleavage domains are highly permeable and that fluid flow is concentrated at the contact between the cleavage P domains and the poorly oriented Q domains (Stephens et al., 1979; White and Johnston, 1981; Etheridge et al., 1983). Since porosity and permeability are factors which control illite crystallinity (Kisch, 1983), conditions appropriate for the white micas to develop a better 'crystallinity' occur, therefore, in the P domains. Thus tectonic strain is important in the development of 'crystallinity' in the white micas. In the study area, there is little doubt that it has played a role because all of the rocks examined are cleaved. However, it has not been the only factor because slates collected from closely spaced locations contain white micas with different IC values (230 894 fig. 2). The most likely explanation for this variability is that more  $K^+$ -rich fluids existed in some specimens than in others at the time of the formation of the micas. This interpretation is based on the observation that IC is dependent on the supply of K in the rock, high K contents favouring increased crystallinity (Kisch, 1980, 1983). Possible evidence for fluid chemistry controlling IC has been noted in some specimens. It is exemplified by data obtained from a rock slice of sample GO8 which shows contrasting IC values in

psammitic, white mica-rich bands  $(0.10 \ \Delta^{\circ} 2\theta)$  and pelitic, chlorite-white-mica bands  $(IC = 0.18 \ \Delta^{\circ} 2\theta)$ .

Another feature noted in the IC study which deserves comment is that IC (rock slice) is generally less than IC ( $< 2 \mu$ m) for the same specimen. This is seen in fig. 4 which shows that the majority of analyses occur above the 1:1 line. These results are similar to those obtained by Weber (1972) who attributed the higher IC values in the  $< 2 \mu$ m fraction, particularly in lower grade samples, to the concentration of very fine-grained illites with a lower degree of crystallinity than in the coarser size fractions. This results in IC from the  $< 2 \mu$ m fraction being greater than in the rock slice where all fractions together give an intermediate value.



FIG. 4. IC values obtained from  $< 2 \,\mu$ m fraction and rock slices of individual specimens.

This may be one possible explanation. However, the difference in IC values may also be attributed to the greater permeability of the P domains than the Q domains in the slates and phyllites. The basis for this suggestion is that other authors have found that substantial changes in chemistry occur in P domains during their formation (Stephens et al., 1979) and that chemically, white micas in P domains are of a 'higher' grade than in Q domains (White and Johnston, 1981). If the same processes have operated during the formation of the slates and phyllites in this area, then white micas with better crystallinity would grow in the P rather than the O domains. Thus when X-rays impinge on the rock slices, they will be diffracted mainly by the micas in the cleavage where they are in abundance.

By contrast, on  $a < 2 \mu m$  fraction sedimented slide, less crystalline white micas from the Q domains will be mixed up with those from the P domains, thus a higher IC value results.



FIG. 5. Histogram of  $b_o$  values obtained in the Hill End Synclinorium and Windamere Dam to Cudgegong areas.

 $b_{o}$  values. The  $b_{o}$  values obtained in this study are similar to those reported by Offler and Pemberton (1983) in the Windamere Dam to Cudgegong area (fig. 5). However, an examination of the spatial distribution of the  $b_0$  values (fig. 6) shows a concentration of low  $b_0$  values in the central and southern part of the area and a less well-defined small group at the eastern margin. The mean value for the main concentration is 9.010 Å, which is much lower than in the surrounding area ( $\bar{x} =$ 9.022 Å). According to a 't' test these two populations could not have arisen by chance and are significant. This being so, it implies that metamorphic conditions were different in the two areas, one akin to the low-medium P metamorphism of New Hampshire, the other, the medium-P metamorphism of the Dalradian (Sassi and Scolari, 1974). This is confirmed by the Si<sup>4+</sup> contents of white micas in specimens containing the limiting assemblage quartz-K-feldspar-chlorite-muscovite ('low'  $b_0$  zone,  $\bar{x} = 3.21$ , n = 3; 'high'  $b_0$  zone,  $\bar{x} = 3.34$ , n = 4) and in those without K-feldspar ('low'  $b_0$  zone, maximum Si<sup>4+</sup> = 3.21, 'high'  $b_0$  zone, 3.33). These contents indicate pressures of approximately 2 kbar and 3 kbar using the experimental work of Velde (1967) and assuming temperatures of 400 and 350 °C respectively. The former temperature is chosen for the low  $b_0$  samples because they are close to the biotite zone boundary, the latter because the samples are near the greenschist- to prehnite-pumpellyite-facies boundary. The value



FIG. 6. Location of samples and their  $b_0$  values. Stippled area contains low  $b_0$  values. Metamorphic zone boundaries as in fig. 2. A  $b_0$  value of 9.007 Å is shown as 7 on the figure.

of 400 °C suggested for the low  $b_o$  samples must be considered as tentative because equilibria involving biotite-forming reactions have not been satisfactorily calibrated (Turner, 1981). However, the value for the high  $b_o$  samples is considered reasonable since the temperatures at which greenschist-facies assemblages first appear in basic rocks is well established (Plyusina and Ivanov, 1979; Schiffman and Liou, 1980).



FIG. 7. AKF diagram showing muscovite (full square), chlorite (heavy bar) and pelitic rock analyses (filled circle with slash 'low'  $b_o$  zone, filled circle 'high'  $b_o$  zone). Analyses from Prendergast (1981);  $b_o$  values are shown adjacent to each analysis.

It might be argued that much of the  $b_0$  variation in this area is a function of host rock chemistry. Thus more aluminous rocks should crystallize low  $b_{0}$  (celadonite-poor) white micas. A plot of a representative suite of pelitic rocks, muscovites, and chlorites in terms of A  $[Al_2O_3 + Fe_2O_3 (Na_2O + K_2O + CaO)$ , K (K<sub>2</sub>O) and F (FeO + MgO + MnO) in molecular proportions, clearly shows that this is not so. Aluminous rocks contain micas with high  $b_0$  and vice versa (fig. 7). A similar result can be obtained by plotting  $Al_2O_3/(Al_2O_3 +$  $SiO_2$ ) (molecular proportions) of the rock against the same ratio for the white mica (fig. 8). Therefore the conclusion is made that P/T conditions rather than host rock chemistry have been the major factor in defining white mica chemistry.

Assuming that the  $b_o$  variations reflect P/T conditions, two questions arise. First, what is the explanation for the marked decrease in  $b_o$  values in the south and second, are the conditions suggested



FIG. 8.  $Al_2O_3/(Al_2O_3 + SiO_2)$  rock (mol. prop.) versus  $Al_2O_3/(Al_2O_3 + SiO_2)$  muscovite (mol. prop.). Filled circle, 'high'  $b_0$  zone, filled circle with slash, 'low'  $b_0$  zone, empty circle with slash, 'low'  $b_0$  and within biotite zone.

by the  $b_o$  values compatible with the ensialic rift setting proposed for this area (Cas, 1983)? A Bouguer anomaly map of the area appears to provide the answer to the first question. A strong negative anomaly overlaps with the main concentration of low  $b_o$  values (fig. 9), suggesting granitoid bodies at depth. If these bodies existed during deformation, they may have been deeper in the crust but were able to provide sufficient heat to increase locally the geothermal gradient. This resulted in the development of less celadonitic white micas. A more extensive biotite zone should have formed if the P/T conditions are as suggested but this has not occurred because rocks of the appropriate composition are absent.

With regard to the second question, Cas (1983) has drawn analogies between the Lachlan Fold Belt, of which the Hill End Synclinorium is part, and the Basin and Range Province, a rift setting in continental crust in the western USA. According to Lachenbruch and Sass (1977), the geothermal gradient in this province varies from 23 to 30 °C/km. Such gradients are comparable with those determined for the medium P Dalradian metamorphism (28-33 °C/km, Turner, 1981), the type of metamorphism recorded in the 'high'  $b_o$  zone. These authors have also noted subregions and subprovinces of higher heat flow (35-43 °C/km). They suggest that these zones of higher heat flow occur



FIG. 9. Bouguer anomaly map (BMR 155/B2-8-0, 155/B2-4) showing late Carboniferous granites (+) and zone of low b<sub>o</sub> values (stippled pattern).

where extension of the lithosphene is locally more rapid. If such zones were present in the Lachlan Fold Belt, then the low  $b_o$  area may represent a subprovince in which slightly more rapid local extension took place. Thus the metamorphic conditions suggested by the pattern are compatible with the ensialic rift setting of Cas (1983), a setting that may have been the result of slow upwelling of mantle material beneath thick continental crust (Åberg *et al.*, 1984).

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