

**POLYTYPES OF 2:1 DIOCTAHEDRAL MICAS IN
SILICIFIED VOLCANICLASTIC SANDSTONES,
WARRAWOONA GROUP, PILBARA BLOCK,
WESTERN AUSTRALIA**

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ABSTRACT. Polytypes of dioctahedral 2:1 micas in pervasively silicified volcaniclastic sandstones of the early Archean Warrawoona Group, Western Australia, provide evidence bearing on the early alteration history of these rocks. Fine-grained "sericitic" aggregates occur as (1) pseudomorphs after feldspar framework grains and feldspar microphenocrysts in dacitic rock fragments and (2) irregular masses within the now silicified sandstones. Peak height ratios of the 2.80Å and 2.58Å illite-muscovite peaks for 12 samples from two widely spaced (35 km) localities were obtained by X-ray diffraction of random powder mounts. Relative percentages of 2M polytypes were determined by comparison to the 2.80Å/2.58Å ratio of pegmatitic muscovite, assumed to be 100 percent 2M.

Abundances of the 2M polytype [$2M/(2M + 1M)$] in the 12 samples range from 44 to 95 percent. Samples from the northern locality, collected from outcrops cut by silica- and barite-filled veins, have higher 2M percentages, ranging from 44 to 80 percent. The wide range in 2M percentages, both regionally and at the outcrop scale, coupled with field observations and thermal ranges for the observed petrographic assemblage, suggest that polytypism is a function of hydrothermal alteration and not regionally uniform low-grade metamorphism in these localities. The maximum temperature of alteration at the time of polytype equilibration is estimated at 200° to 350°C. The sampled localities represent some of the least-altered sequences in the Warrawoona Group, having undergone early, syndepositional alteration but probably escaping subsequent greenschist- to amphibolite-grade metamorphism, which has affected much of the Warrawoona Group.

INTRODUCTION

The fundamental structural motif of illite-muscovite crystal structures is the 2:1 dioctahedral layer. Stacking of the 2:1 layers parallel to the c-crystallographic axis results in the growth of mica crystals. Theoretically, there are six simple ways, with periodicities between one and six, in which 2:1 layers may be stacked, giving rise to the 1M, 2M₁, 2M₂, 20r, 3T, and 6H polytypes (Yoder and Eugster, 1955). In nature, however, only the 1M (1M and 1Md), 2M₁, and 3T polytypes have been commonly identified among dioctahedral muscovites (Bailey, 1980).

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Following the work of Yoder and Eugster (1955), many authors have shown that the 1M polytype prevails at low temperature, progressively reacting to form the 2M polytype at higher temperatures. Hence, dioctahedral mica polytypes are correlated with geologic occurrence and can be used to monitor diagenesis and very low-grade metamorphism: 1M varieties dominate in sedimentary and weathering environments (Bailey and others, 1962, Velde and Hower, 1963) whereas 2M polytypes become dominant in higher temperature magmatic and metamorphic mica associations (Reynolds, 1963; Maxwell and Hower, 1967; Karpova, 1969).

This report presents results of polytype determinations of 2:1 K-bearing dioctahedral micas from silicified volcanoclastic sandstones of early Archaean age. These rocks are now composed largely of replacement silica with subordinate dioctahedral mica. The 2:1 polytype distribution is coupled with other mineralogic and geologic criteria to help constrain the early alteration history of these rocks.

GEOLOGIC AND PETROGRAPHIC BACKGROUND

Micas analyzed in this study occur in silicified volcanoclastic sandstones near the top of the 3.3 to 3.5 Ga Warrawoona Group (Pidgeon, 1978; Jahn and others, 1981) in the granitoid-greenstone terrane of the eastern Pilbara Block, Western Australia. These sandstones are assigned to the Panorama and Duffer Formations and are believed (DiMarco, ms) to represent part of a regionally correlative unit of felsic lavas and volcanoclastic rocks in the eastern Pilbara Block. These and similar felsic volcanic and volcanoclastic rocks form a volumetrically minor component of the predominantly basaltic, 15+ km-thick Warrawoona Group. The Warrawoona Group has undergone extensive, locally variable, but generally low- to medium-grade alteration.

The present mineralogy of the altered sandstones, which includes replacement granular microcrystalline quartz and subordinate amounts of finely divided "sericite," bears little resemblance to the original compositions of the sandstones (DiMarco and Lowe, 1984; DiMarco, ms). X-ray analysis indicates that the "sericite" consists largely of K-bearing dioctahedral mica. Well-preserved pseudomorphs of primary minerals indicate that the sandstones were originally composed of felsic volcanic rock fragments, volcanic quartz, feldspar, sedimentary rock fragments, and mafic to ultramafic rock fragments. All rock fragments as well as original cement and matrix were replaced by microquartz and microcrystalline phyllosilicates. Detrital volcanic quartz, in many cases strain-free and having straight, well-defined grain margins, appears to have remained stable during alteration. Fine-grained micas or "sericitic" aggregates occur as (1) pseudomorphs after feldspar framework grains and feldspar microphenocrysts in volcanic rock fragments and (2) irregularly shaped domains without apparent pseudomorphous relations within replacement microquartz.

Initial results of whole-rock, major-element analyses (table 1) indicate that these altered sandstones are composed largely of SiO_2 ,

TABLE 1

*Major-element contents of Warrawoona altered volcanoclastic sandstones**

Location** Sample	NL P1-15-16	SL P1-16-5	NL P1-8-7	NL P1-8-14
SiO ₂	92.25	82.27	90.04	83.35
Al ₂ O ₃	4.57	11.69	6.59	9.52
Fe ₂ O _{3T}	0.32	0.32	0.12	0.25
MgO	0.00	0.02	0.07	0.13
CaO	0.03	0.08	0.02	0.03
Na ₂ O	0.02	0.01	0.04	0.06
K ₂ O	1.19	3.23	1.79	2.62
TiO ₂	0.13	0.42	0.10	0.16
P ₂ O ₅	0.03	0.28	0.02	0.03
MnO	0.01	0.01	0.03	0.20
S	0.01	0.01	0.03	0.20
	98.56	98.34	98.85	96.55

*Data by X-ray fluorescence (J. Veizer, personal commun., 1985).

**NL = northern locality, SL = southern locality.

Al₂O₃, and K₂O and have low concentrations of other major-element oxides. DiMarco (ms) showed that, compared to some modern felsic volcanoclastic sands, these rocks are enriched in SiO₂ and are depleted in Fe, Mg, Ca, and Na oxides. Because replacement mica is commonly concentrated at the sites of former feldspar grains, it appears that Al remained relatively immobile during alteration.

Electron microprobe analyses of individual mica flakes reveal a high within-sample variation and an overall potassium deficiency. Structural formulae calculated on the basis of 44 negative charges per 2:1 sheet illustrate the potassium-deficient character of the phyllosilicate assemblage and the aluminous nature of the octahedral layers. Results from sample P1-8-15 produced the ranges indicated below:

K _{0.67-1.77}	Na _{0.0-0.08}	Ca _{0.0-0.03}	interlayer
Al _{3.59-3.67}	Fe ³⁺ _{0.04-0.29}	Mg _{0.05-0.41}	octahedral layer
Si _{6.84-6.95}	Al _{1.05-1.16}		tetrahedral layer

The 2:1 micas could be termed hydromicas.

ALTERATION AND METAMORPHISM

Regional, early, low-grade alteration has affected all rocks within the Warrawoona Group. The very early timing of at least the initial stages of alteration is indicated by the good preservation of framework-grain textures and the loose packing of framework grains in these sandstones (DiMarco and Lowe, 1984; DiMarco, ms), the preservation of delicate evaporite and carbonate depositional habits by replacement silica or barite in associated facies (Lowe, 1983; Buick and Barnes, 1984), and argon-isotope work suggesting an age of at least 3200 Ma for regional metamorphism of the Warrawoona Group (Wijbrans and McDougall, 1987). Despite its widespread extent, a systematic and detailed picture of regional alteration and metamorphic trends is lack-

ing, perhaps because of the remoteness of the area as well as the paucity of suitable protolithologies, such as carbonates and mudrocks, within the Warrawoona Group.

Hickman and Lipple (1978) reported that the Archean succession was affected by pervasive greenschist-facies metamorphism, although amphibolite facies is locally present and prehnite–pumpellyite–meta-graywacke facies occurs in, among other places, the North Pole Dome (northern locality, fig. 1). Barley (ms), as part of a petrogenetic study of felsic rocks of the Warrawoona Group in the eastern Pilbara Block, made several points, including (1) the amphibolite grade is restricted to highly strained parts of the Warrawoona Group adjacent to gneissic parts of granitoid batholiths, (2) alteration, occurring during very low grade metamorphism, involved a fluid phase and resulted in the generation of sericite and carbonate replacement minerals, (3) alteration is particularly strong in originally porous rocks such as sandstones and tuffs, and (4) the lack of key lithologies such as pelites, marls, and dolomites precluded the regional mapping of detailed isoreaction grads based on important indicator minerals. Barley (ms) proposed the following metamorphic history for Warrawoona felsic volcanic rocks: early

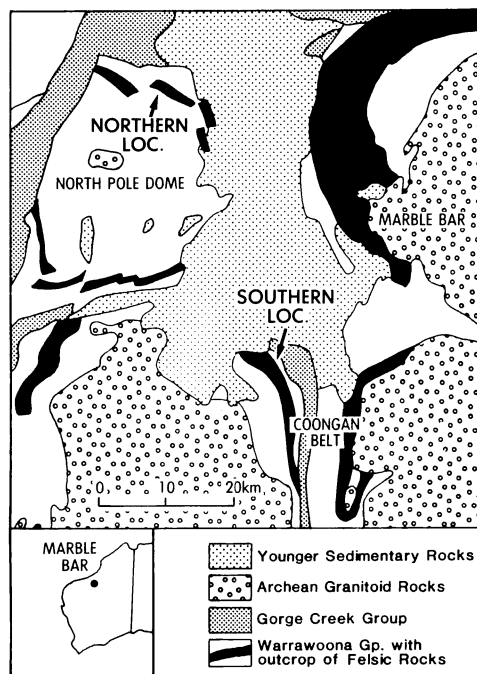


Fig. 1. Generalized geologic map of a portion of the eastern Pilbara Block showing outcrop extent of felsic rocks and approximate locations of sample collection areas.

seafloor alteration followed by burial diagenesis in an active volcanic environment accompanied by circulation of fluids and a high geothermal gradient. In a later paper, Barley (1984) suggested that the bulk of Warrawoona alteration, resulting in sericitization, carbonation, and silicification, stemmed largely from hydrothermal activity in a volcanic-related, shallow-water or subaerial system characterized by boiling.

Thus, it appears that early, syndepositional alteration, probably related to volcanic activity, affected much of the Warrawoona Group. Subsequent, but still early Archean, metamorphism of up to amphibolite grade then overprinted rocks in many areas. The lowest grades of alteration, perhaps escaping the later overprinting and representing the earlier, syndepositional phase, occur as low-strain sequences preserving many primary depositional and volcanogenic features. More highly altered rocks, particularly those adjacent to granitoid batholiths, show evidence of strain, the development of schistosity, and the progressive destruction of primary depositional features.

Analyzed samples were collected from two localities separated by about 35 km (fig. 1). Rocks at both localities show little evidence of strain and good preservation of sedimentary structures and textures. More highly strained rocks were not analyzed. Rocks from the northern locality, on the northern flank of the North Pole Dome, are cut by numerous silica- and barite-filled veins. Such veins are largely absent in the southern Coongan Belt locality. Samples at each locality represent no more than 100 m of stratigraphic thickness and were collected along traverses no more than 0.5 km wide.

ANALYTICAL METHODS

Unweathered rock chips were spalled from hand samples with a geologic hammer. Chip size was reduced to a maximum of approx 0.5 to 1.0 mm with a rotating disk pulverizer. Several runs of 5 to 10 sec duration were required for complete sample disaggregation. The fine fraction, less than 60 micrometers, which contained some rock flour, was removed after each run. This fraction was discarded to minimize the potential destruction of 2M polytypes of grinding to ultrafine sizes (Maxwell and Hower, 1967). Following pulverization, mica flakes were liberated from crushed rock using an ultrasonic generator. The <5 micrometer size fraction was then obtained by sedimentation through 5 cm for 22 min in a 0.1 percent solution of Na_3PO_4 . The suspensions were then concentrated by centrifugation and dried overnight in a 100°C oven prior to X-ray analysis.

X-ray diffractograms were obtained from randomly oriented powdered samples using a Philips APD-3500 diffractometer with Cu K_α radiation. Data were collected by step counting ($0.02^\circ 2\theta$ increments for 1 sec) from 25° to $40^\circ 2\theta$ with the chart full scale = 1600 counts. Random powder mounts were prepared using a side-loading Al sample holder. All peak intensity measurements, after background subtraction, were recorded as a percentage of the full scale.

The relative abundances of 2M and 1M micas were determined using the method of Maxwell and Hower (1967) by comparing the ratio of the height of the 2.80Å peak, which is unique to the 2M polytype, to that of the 2.58Å peak, which is of equal intensity for both polytypes. The technique does not distinguish 2M₁ and 2M₂ polytypes nor recognize 1Md forms, so the general designations 2M and 1M are used in this report. The 3.74Å peak used by Velde and Hower (1963) for mica polytype determination could not be used because of interference in some samples. It was assumed that the intensity ratio produced by the pegmatitic muscovite, used as a calibration standard, represents a sample of pure 2M polytype (Smith and Yoder, 1956). Intensity ratios from mixtures of the two polytypes are assumed to vary in direct proportion to the relative abundance of the 2M and 1M polytypes (fig. 2). In figure 3, proportions of two diffractograms are reproduced to illustrate the appearance of the 2.80 and 2.58Å peaks in a sample with a high relative proportion of 2M material in comparison to a sample that is about 60 percent 1M material. Reproducibility results based on five replicate analyses of sample PI-17-9 suggest a maximum standard deviation of ± 9 percent of the measured 2.80Å peak and a maximum experimental uncertainty in 2.80Å/2.58Å ratios of ± 10 percent of the reported value.

In order to check that the mechanical pulverizer did not destroy any 2M polytype that may have been present, the diffractogram for pulverized rock flour of sample PI-30-18 was compared to that of micas released during ultrasonic treatment of physically separated sand-sized

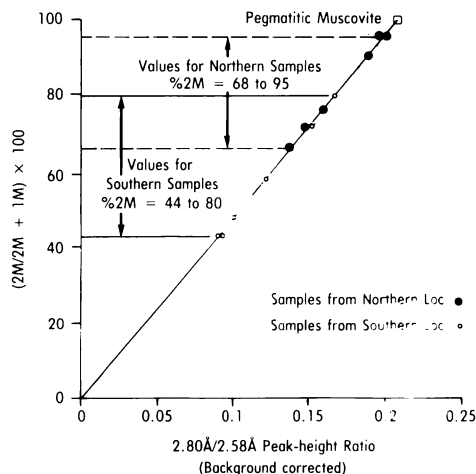


Fig. 2. Graph for determination of the percentage 2M dioctahedral mica in the mica fraction as a linear function of 2.80Å/2.58Å peak intensity ratios for volcanic sandstones of the Warrawoona Group. Muscovite from a pegmatite was assumed to be 100 percent 2M mica. Observed intensity ratios for the samples create two overlapping populations.

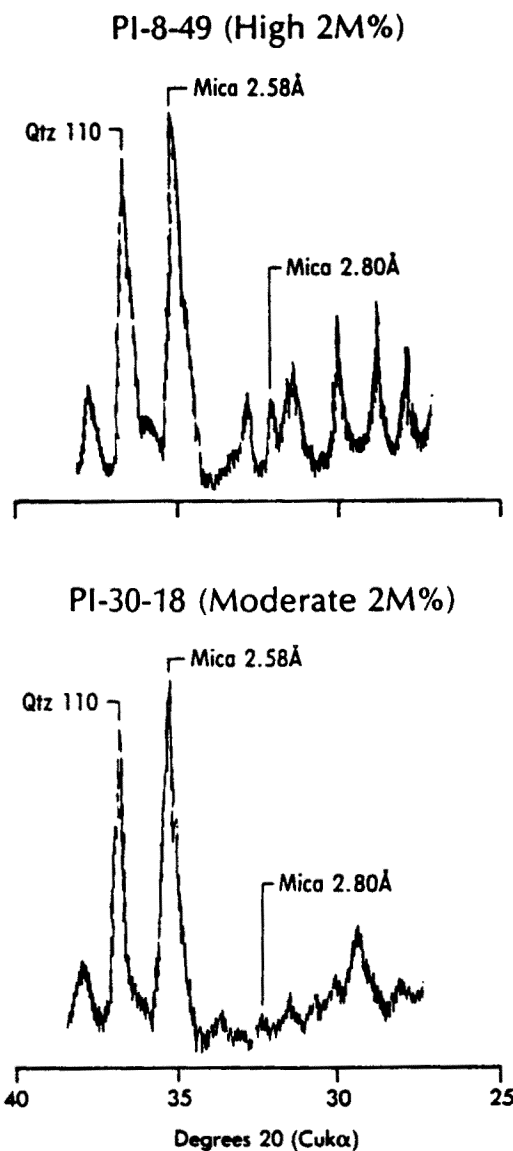


Fig. 3. Partial X-ray diffractograms of two random powder mounts illustrate the variation in peak intensities for samples with moderate (PI-30-18, 44 percent 2M) and high (PI-8-49, 85 percent 2M) quantities of 2M mica. Unlabeled peaks are attributed to mica and quartz.

fragments of the same sample. The 2.80Å/2.58Å ratios for both fractions are identical within the experimental error. Hence, there is no evidence that 2M polytypes were destroyed during sample preparation.

RESULTS

Heights of the mica 2.58Å and 2.80Å peaks and the 2.80Å/2.58Å peak ratios for analyzed samples are listed in table 2. There is a significant range in the 2.80Å/2.58Å ratios for the group as a whole (0.09–0.20) as well as for subset samples for each locality. The muscovite control sample, assumed to be 100 percent 2M, has the highest 2.80Å/2.58Å ratio. A mixture of 1M and 2M polytypes is present in the Warrawoona samples (fig. 2). The smallest percentages of 2M material occur in samples from the Coongan Syncline (44–80 percent). Samples from the North Pole Dome are enriched in the 2M polytype (68–95 percent).

Precise quantification of the percentage of 2M polytype present in each sample is difficult. Velde and Hower (1963) and Maxwell and Hower (1967) suggested that the curves of percentage 2M polytype versus 2.80Å/2.58Å ratio approach linearity. Figure 2 illustrates the calibration curve used in our study, but as noted by Reynolds (1963), such polytype percentage values have little absolute significance because of the uncertainty of the 2M content of the calibration material and the unverified assumption of a linear relation between 2.80Å peak height and actual percentage of 2M. Thus, values of percentage 2M polytype

TABLE 2

Peak heights for 2.80Å and 2.58Å peaks, silicified volcanoclastic sandstones, Warrawoona Group. Chart full scale = 1600 cts, peak height of 10 approx = 150 cts

Sample	2.80Å	2.58Å	2.80Å/2.58Å
Muscovite control	12.4	60.2	0.21
PI-8-4*	2.0	12.7	0.16
PI-8-9	interference	14.0	—
PI-8-15	4.0	22.0	0.18
PI-8-26	not detected	7.7	—
PI-8-35	1.8	12.0	0.15
PI-8-36	2.0	13.9	0.14
PI-8-49	4.9	24.7	0.20
PI-8-56	2.4	12.0	0.20
PI-17-9**	8.3	47.5	0.17
PI-30-13	2.7	30.7	0.09
PI-18-7	2.0	13.5	0.15
PI-30-18(1)	1.8	18.7	0.10
PI-30-18(2)	1.6	13.6	0.12
PI-30-18(3)	2.2	24.0	0.09

*Samples PI-8- collected at northern locality (North Pole Dome).

**Others are from southern locality (Coongan Syncline) illustrated in figure 1. Effects of sample preparation technique on analytical reproducibility was evaluated by preparing PI-30-18 three ways: (1) micas from sand-sized rock fragments, (2) micas from crushed rock flour, (3) micas from mixtures of rock fragments and crushed rock flour.

obtained from figure 2 are approximate and correctly should be termed "peak-height ratio percentages." However, for the sake of brevity, they will be termed "percentage 2M polytype" in this report.

DISCUSSION

Any model for the origin of mica polytypes in these altered volcanoclastic sandstones must be consistent with (1) characteristics of the polytype population present, (2) considerations inferred from the petrographic assemblage of the sandstones, and (3) known or inferred post-depositional events that could account for the presence of mica. For altered sandstones in the Warrawoona Group, the most likely origins for replacement mica are from (1) regionally uniform low-grade burial metamorphism and (2) hydrothermal alteration. A continuous and overlapping spectrum of processes exists between hydrothermal alteration and regionally extensive low-grade burial metamorphism. In this paper, we use the criterion of Henley and Ellis (1983) that low-grade metamorphism would be characterized by a tendency toward isochemical reactions with low water/rock ratios and, in many cases, higher temperatures, compared to hydrothermal alteration.

Because of the low values and wide range in percentage 2M polytype, even as observed at the scale of a single outcrop, an origin during uniform, regionally extensive, low-grade burial metamorphism is unlikely. Although we know of no detailed polytype analyses on predominantly sandstone compositions, mica polytypes in altered pelitic sequences suggest that uniformly pervasive low-grade metamorphism would provide a greater percentage of 2M than observed in the Pilbara sandstones. For example, Maxwell and Hower (1967) noted greater than 80 percent 2M in Belt Group shales, Idaho, which have undergone prehnite-pumpellyite (200°–300°C) and greenschist-grade (300°–400°C) metamorphism. The great lengths of time available for mineralogic equilibration during an episode of simple low-grade burial metamorphism might also lead to a more restricted range of 2M percentages than was observed, especially at the outcrop scale.

As suggested by the relatively wide range in percentage 2M polytype and pervasive, early silicification, it is more likely that the dioctahedral polytype distribution present in these samples was caused by early hydrothermal alteration. Unlike regional, low-grade burial metamorphism, which might be expected to produce uniform mineralogic trends in geologic space, hydrothermal fluids follow fracture systems, lithologic inhomogeneities, and stratigraphic paths of least resistance, often producing complex and tortuous alteration networks (Henley and Ellis, 1983). Geologic evidence for hydrothermal alteration at the Warrawoona outcrops where the samples were collected include the presence of silica- and barite-filled vein networks, especially at the northern locality, and higher degrees of silicification in originally more porous units such as sandstones in comparison to more massive lava flows. The pervasive silicification in these rocks is quite similar to silica

“self-sealing” of primary porosity observed in many modern hydrothermal systems (Keith, White, and Beeson, 1978). The wide range in 2M percentages observed in the Warrawoona samples, even over a few tens of meters, is compatible with highly complex alteration patterns associated with hydrothermal systems and other fluid-rich low-grade alteration processes. A similar range in 2M percentages (38–79 percent 2M polytype) was noted by Reynolds (1963, p. 1105) for hydrothermally altered Proterozoic rocks from Idaho that were collected from outcrops that are “abundantly silicified and mineralized.”

Precise temperature data on the transformation of 1M to 2M dioctahedral micas are unknown because of the sluggish nature of the laboratory reaction (Yoder and Eugster, 1955) and the possibility, for natural systems, that some of the 2M polytype measured might represent detrital, not in situ, material. Depositional mechanics for these sandstones (DiMarco, ms; DiMarco and Lowe, 1986) suggest that little clay-sized material was deposited with the sand in many of the sampled localities. Thus, a comparison of temperature estimates of the 1M–2M transition from other field and laboratory studies, combined with thermal information inferred from the observed petrographic assemblage, can constrain the temperature of hydrothermal fluids presumed to have produced the polytype population present in these samples. X-ray diffractograms indicate that no expandable clays are present. This places a lower limit of alteration at 175°C, the approximate temperature at which the conversion of smectite to illite is complete (Hower and others, 1976). In modern hydrothermal systems, illite is present over the range 150° to >300°C (Henley and Ellis, 1983). Rocks of the Belt Supergroup, which equilibrated at 200° to 300°C according to oxygen-isotope data (Eslinger and Savin, 1973), contain 80 to 100 percent 2M mica as do some samples from both Warrawoona localities. Weaver and others (1984, p. 86) suggested that the 1M–2M transformation in the southern Appalachians occurred between 280° and 360°C. Frey (1987), in a synthesis of data from numerous sources, suggested that the 1M–2M transition occurs between 200° to 350°C for laboratory runs of long duration and is completed in natural systems at the anchizone-epizone boundary (approx 350°C). Quartz framework grains in the Warrawoona sandstones remained physically and chemically stable during the alteration event. Hoagland and Elders (1978) found that detrital quartz is inert in hydrothermal systems up to about 300°C. These observations suggest that hydrothermal alteration and polytype generation in sandstones of the Panorama Formation probably took place between 200° and 350°C.

These temperature estimates probably represent the highest temperatures that affected these micas during the alteration episode. Clearly, alteration of the Warrawoona sequence began early. Data presented here might not reflect the temperature at which the mica began to form but rather the highest temperature of mica equilibration. It appears that at the localities sampled, low-temperature hydrothermal

alteration was the highest grade of alteration reached. Elsewhere in the Warrawoona Group, higher grades of metamorphism were achieved.

One additional origin for the observed pattern of polytype distribution has been considered. The spatial variability of the polytypes could be associated with present or paleo-weathering reactions, especially because modern silicification and silcrete formation are so common in the Australian desert. However, this origin is ruled out by the absence in these samples of expandable clays such as smectite.

SUMMARY

A wide range in percentage 2M polytype in dioctahedral 2:1 micas is present in pervasively silicified volcanoclastic sandstones of the early Archean Warrawoona Group, Western Australia. This wide range persists even at the scale of a single outcrop. These data, in conjunction with geologic and petrographic data, suggest that hydrothermal alteration, not regionally uniform low-grade burial metamorphism, generated the polytype population of K-bearing dioctahedral micas in these altered rocks. Based on temperature estimates for the 1M-2M transformation and thermal implications of the petrographic assemblage present in these rocks, the mica-altering hydrothermal fluids probably attained a temperature of 200° to 350°C.

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