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**HYDROUS CORDIERITE IN THE HIGH-GRADE CRUST:
IMPLICATIONS FOR MAGMA GENERATION**

G. STEVENS, J.D. CLEMENS and G.T.R. DROOP

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ABSTRACT

In experiments on biotite fluid-absent partial melting in metasediments, cordierite proportion is inversely correlated with melt fraction. At constant P and T , this variation in melt proportion is a function of bulk-rock Mg# ($100\text{Mg}/(\text{Mg}+\text{Fe})$) and $a\text{Al}_2\text{O}_3$. However, melt compositions are essentially unaffected and melt fractions are a function of melt:cordierite H_2O partitioning. At 0.5 GPa, the calculated H_2O contents of cordierite decrease from ~ 1.4 to ~ 0.9 wt% as T increases from 850 to 1000°C. These observations have important implications for the partial melting of aluminous metasedimentary rocks and the production of crustally derived granitoid magmas. In such compositions, the cordierite stability field expands dramatically with increasing Mg# and $a\text{Al}_2\text{O}_3$, and considerably overlaps with the conditions of fluid-absent melting in the middle to lower crust. Under these circumstances, the fluid-absent melting of biotite occurs with a crystalline hydrate in both the reactant and product assemblages, severely restricting melt productivity. Consequently, metapelites may be much less fertile sources of granitic magma than metagreywackes, in which garnet and orthopyroxene are the main ferromagnesian residues of partial melting under similar conditions.

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INTRODUCTION

Experiments on fluid-saturated systems show that Mg-cordierites have almost complete solid solution from the anhydrous ($\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$) to the fully hydrated end-member ($\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18} \cdot 1.2\text{H}_2\text{O}$) with 2.99 wt% H_2O (Mirwald and Schreyer, 1977; Johannes and Schreyer, 1981).

Cordierites from high-grade metamorphic terranes, in which fluid-absent conditions are likely (Stevens and Clemens, 1993), also have significant H_2O contents (e.g. Droop, 1989; Vry et al., 1990). However, the volatile contents of natural high-grade cordierites are rather variable. Cordierites in metasedimentary, granulite-grade migmatites from Prydz Bay, East Antarctica, contain between 0.1 and 0.5 wt% H_2O , while CO_2 contents in the same crystals vary from 0.1 to 1 wt% (Fitzsimons, 1994). In a study of volatile contents of cordierites from 33 different granulite exposures, Vry et al. (1990) documented H_2O contents varying from 0.33 to 1.75 wt%. Also, some high-grade cordierites are zoned in volatile content, suggesting they may be reset during cooling.

The possibility of retrograde resetting, as well as the large variations in measured natural cordierite H_2O content, make it impossible to quantify H_2O partitioning between cordierite and other hydrous phases, including melt, in the fluid-absent high-grade environment. Consequently, the H_2O contents of high-grade cordierites have been dealt with inconsistently in petrological modelling exercises. Waters (1988) and Hensen and Harley (1990) discussed cordierite stability in the context of crustal melting and petrogenetic grids, and Bhattacharya (1986) and Waters (1988) modelled hydrous cordierite equilibria. All of this work would have benefited from better constraints on cordierite-melt H_2O partitioning. Even more serious errors are introduced when studies assume anhydrous high-grade cordierite, or treat cordierite as having a constant hydration state. To regard cordierite as an anhydrous mineral always seems to underestimate its stability range, as illustrated by Newton (1972) and Newton and Wood (1979).

A potential hydrous component in high-grade cordierite also has implications for crustal anatexis. In high-grade metasedimentary sources voluminous and hot granitic magmas, capable of rising through the crust, are generated through fluid-absent, incongruent melting reactions involving biotite (Clemens, 1990). In theoretical and experimental investigations of melt fractions produced during such fluid-absent partial melting events, the melt is typically regarded as the only hydrous product phase (e.g. Clemens and Vielzeuf, 1987; Vielzeuf and Holloway, 1988; Le Breton and Thompson, 1988). Melt fraction is thus seen as a function of the amount of decomposing hydrous phase (biotite), its H_2O content, and the H_2O solubility in the melt. Field investigations of migmatites (e.g. Ashworth, 1976; Phillips, 1980; Stevens and Van Reenen, 1992; Fitzsimons, 1994) indicated that cordierite is an important product of biotite melting reactions in metapelites from diverse tectonic environments. Under these circumstances, the cordierite produced is also likely to be hydrous, and potentially influencing the amount of melt produced.

If, during fluid-absent breakdown of stoichiometric phlogopite, the hydrous Mg-cordierite in the product assemblage were to contain exactly 0.67 H_2O pfu (i.e. 44 % free

"cages"), the reaction would be H₂O conserving: $2\text{KMg}_3\text{AlSi}_3\text{O}_{10}(\text{OH})_2 + 9\text{SiO}_2 + 6\text{Al}_2\text{SiO}_5 = 3[\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18} \cdot 0.67\text{H}_2\text{O}] + 2\text{KAlSi}_3\text{O}_8$ with no melt produced. Thus, if the H₂O content of cordierite is unknown, prograde dehydration, retrograde dehydration, and water-conserving reactions, are all possible. The actual H₂O contents of natural high-grade cordierites are thus of considerable interest. While this paper was in review, Harley (1994) published a single data point from an experiment (at 900°C, 0.7 GPa) on a model metapelite in the Ca- and Na-free analogue system K₂O-MgO-FeO-Al₂O₃-SiO₂-H₂O. This result indicates a melt-cordierite H₂O partitioning coefficient ($\text{H}_2\text{O}_{\text{melt}}/\text{H}_2\text{O}_{\text{Crd}}$) of 3 ± 0.3 confirming the potential importance of cordierite H₂O content in anatexis processes. Unfortunately, though the experiment was meant to be fluid-absent, the sample is thought to have been contaminated with H₂O and to have been fluid-present. The melt H₂O content was not reported.

In this study, it is shown how hydrous cordierite restricts the fertility of metapelites as magma sources. At temperatures up to 900°C the data allow good estimates of cordierite H₂O content and melt-cordierite H₂O partitioning. This is higher than the maximum *T* attained in most high-grade rocks. Above this, uncertainties in the H₂O content of the melts produce large errors. However, even here, modal data illustrate the significance of H₂O-bearing cordierite in restricting melt fractions.

STARTING MATERIALS AND EXPERIMENTAL TECHNIQUES

Three "metapelite" compositions were prepared by mixing natural quartz, plagioclase (An₃₅) and sillimanite with synthetic biotites in the proportions Qtz 28%, Pl 15%, Sil 28%, Bt 28% (mineral abbreviations are those of Kretz, 1973). The three biotites are all Ti-free, with the same Al and H₂O contents, and vary only in their Mg#s. This results in three otherwise almost identical whole-rock compositions, with Mg#s fixed at 49 (AS), 62 (BS) and 78 (CS) and *a*Al₂O₃ buffered at a relatively high level by coexistence of sillimanite and quartz. A "metagreywacke" composition (A), with lower *a*Al₂O₃, (Al₂SiO₅ - free) was created by mixing quartz and plagioclase with the most Fe-rich biotite, in the proportions Qtz 39%, Pl 21%, Bt 39%. Mineral powder starting mixtures were prepared by grinding to an average grain-size of $\leq 5 \mu\text{m}$. Aliquots of 100 to 200 mg of starting material were loaded into gold tubes and dried overnight at 110°C, prior to immediate crimping and arc welding. Phase identification in the run products was primarily by SEM on polished sections.

Experiments were conducted at 0.5 GPa, in the temperature range 800 - 1000°C, using a Holloway-type, small-volume, internally heated argon gas vessel. Pressure measurement was by means of a manganin cell and bridge, believed accurate to ± 5 MPa. Temperature was controlled to $\pm 1^\circ\text{C}$ and measured using type-K (chromel-alumel) thermocouples, believed accurate to $\pm 3^\circ\text{C}$. Run times varied from 240 hours (at 850°C) to 120 hours (at 1000°C). Product assemblages for experiments at 850, 900, 950 and 1000°C are listed in Table 1.

Phase proportions in the run products (Fig. 1) were calculated by least-squares mixing. The average of the sum of the squared residuals for the 16 mixing calculations is less than 0.15. The accuracy of these data was tested by comparison with orthopyroxene, garnet, quartz and spinel proportions derived from computer analysis of BSE images of the run products. This technique cannot be used reliably to derive the entire mode of a specimen, due

Table 1: Product assemblages for 0.5 GPa experiments at 850, 900, 950 and 1000°C. Mineral abbreviations are those of Kretz (1973). In addition, Gl = glass (quenched melt). Mg#s of the FeO- and MgO-bearing phases are given in subscript

Comp.	T (°C)	Assemblage
AS	850 ± 2	Qtz, Pl, Sil, Gl ₂₇ , Grt ₃₆ , Crd ₆₅ , Kfs
"	900 ± 2.5	Qtz, Pl, Sil, Gl ₂₁ , Grt ₃₈ , Crd ₆₉ , Kfs, Sp ₁₈
"	950 ± 3	Qtz, Pl, Sil, Gl ₂₁ , Grt ₄₁ , Crd ₇₁ , Sp ₂₀
"	1000 ± 4	Qtz, Sil, Gl ₁₉ , Crd ₆₅ , Sp ₂₃
BS	850 ± 2	Qtz, Pl, Sil, Gl ₂₅ , Grt ₄₀ , Crd ₇₀ , Kfs
"	900 ± 2.5	Qtz, Pl, Sil, Gl ₃₃ , Grt ₃₇ , Crd ₇₃ , Kfs
"	950 ± 3	Qtz, Pl, Sil, Gl ₃₁ , Crd ₇₀ , Sp ₂₁
"	1000 ± 4	Qtz, Pl, Sil, Gl ₂₆ , Crd ₇₀ , Sp ₂₆
CS	850 ± 2	Qtz, Pl, Sil, Gl ₃₃ , Crd ₈₃ , Kfs
"	900 ± 2.5	Qtz, Pl, Sil, Gl ₃₆ , Crd ₈₀ , Kfs
"	950 ± 3	Qtz, Pl, Sil, Gl ₄₁ , Crd ₇₇
"	1000 ± 4	Qtz, Pl, Sil, Gl ₃₂ , Crd ₈₂
A	850 ± 2	Qtz, Pl, Gl ₄₄ , Grt ₃₉ , Opx ₄₈ , Crd ₇₃ , Kfs
"	900 ± 1	Qtz, Pl, Gl ₃₅ , Grt ₃₄ , Opx ₅₀
"	950 ± 3	Qtz, Pl, Gl ₃₁ , Grt ₃₅ , Opx ₄₈
"	1000 ± 2	Qtz, Gl ₂₈ , Opx ₅₅

to similarities in the BSE grey scales of cordierite, plagioclase and K-feldspar from experiments at 850 and 900°C. Comparison of the mixing derived modes with the image analysis results indicates that the calculated phase proportions are accurate to better than ± 2.5 wt%. Phase compositions used in the calculations were determined by energy-dispersive X-ray analysis on the SEM (for analytical details see Vielzeuf and Clemens, 1992). To avoid alkali counting losses during analysis, the glasses were cooled to -196°C by a liquid nitrogen cryo-stage. Bulk-rock H₂O contents were measured by thermogravimetry (TG), in argon, on the prepared and dried starting materials. TG results for standard materials suggest an absolute accuracy of better than 0.05 %.

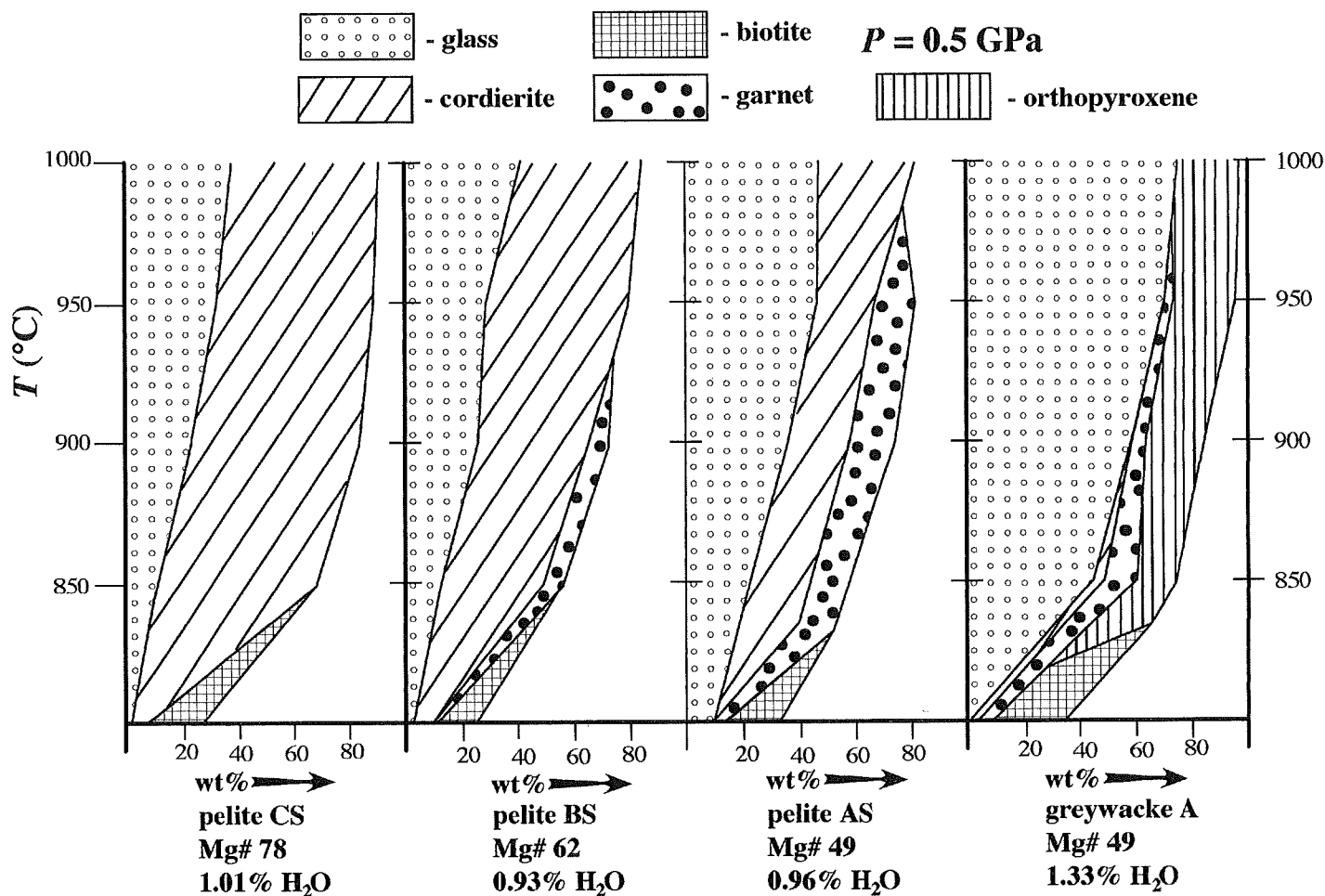


Figure 1: Variation in the proportions of the major products of biotite fluid-absent melting in the four bulk compositions as a function of T . Note the strong bulk-rock Mg# and $a\text{Al}_2\text{O}_3$ (pelite vs greywacke) controls on metasediment fertility and the inverse correlation between melt and cordierite abundance.

MELT PROPORTION AND H_2O PARTITIONING

In the metapelite samples, the fluid-absent biotite breakdown reaction can be expressed qualitatively as $\text{Bt} + \text{Sil} + \text{Qtz} + \text{Pl} = \text{Crd} + \text{Kfs} \pm \text{Grt} + \text{Melt}$. In the metagreywacke the reaction is $\text{Bt} + \text{Qtz} + \text{Pl} = \text{Opx} + \text{Grt} + \text{Kfs} + \text{Melt}$. Cordierite is a major phase in all metapelite run products between the solidus (at $\sim 800^{\circ}\text{C}$) and 1000°C , but is never present in more than trace quantities in the metagreywacke. Cordierite and garnet proportions in the metapelites vary systematically with bulk-rock Mg# (Fig. 1), probably in response to the divariant equilibrium $\text{Grt} + \text{Sil} + \text{Qtz} = \text{Crd}$, which is located at progressively higher pressure with increasing Mg# (Hensen and Green, 1973; Currie, 1974; Hutcheon et al., 1974). This accounts for the differences in garnet and cordierite proportions between the different metapelites, at temperatures below the disappearance of garnet (Fig. 2). In AS and BS, garnet does not seem to have been consumed by this reaction at high T . Rather, garnet disappears through an incongruent melting reaction that produces cordierite and hercynitic spinel.

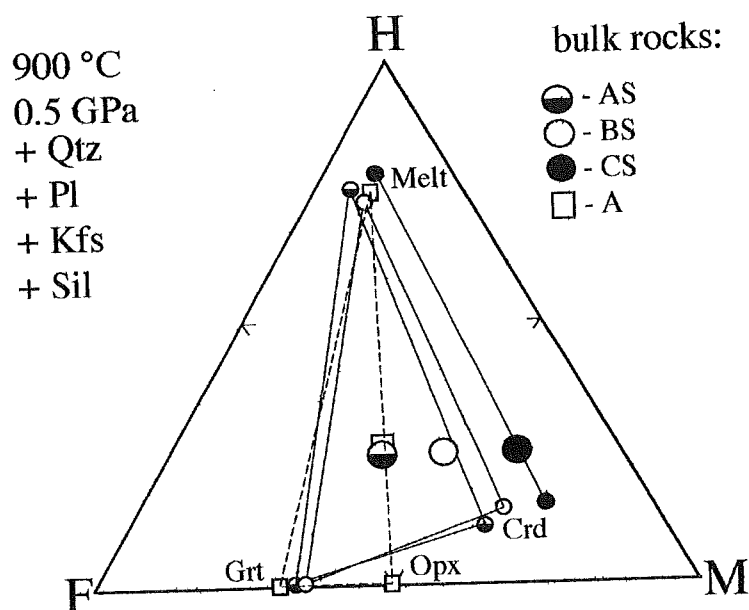


Figure 2: Molecular HFM (H_2O -FeO-MgO) projection, from K-feldspar, plagioclase and quartz, illustrating the projected glass and cordierite compositions, as well as the bulk-rock Mg# control on the equilibrium $\text{Grt} + \text{Sil} + \text{Qtz} = \text{Crd}$ in the metapelites.

The marked inverse correlation between melt proportion and bulk-rock Mg# in the metapelites is not accompanied by any large shifts in melt composition (Table 2) and all the metapelite melts in the 850 and 900°C runs coexisted with quartz, plagioclase and K-feldspar. Thus, this relationship must be a function of H_2O partitioning between cordierite

Table 2: Representative glass (quenched melt) compositions, normalized to 100 wt% anhydrous, from experiments at 900°C and 0.5 GPa

	AS	BS	CS	A
SiO₂	74.49	74.24	74.26	74.25
Al₂O₃	14.29	14.18	14.45	13.94
MgO	0.25	0.48	0.41	0.52
FeO	1.65	1.78	1.30	1.76
CaO	0.64	0.63	0.74	1.09
Na₂O	2.30	2.28	2.16	2.31
K₂O	6.39	6.39	6.68	6.13

and melt. To determine the cordierite H₂O content, and make an estimate of cordierite-melt H₂O partitioning, it is necessary to know the melt H₂O content. This can be estimated, with some confidence, for melts coexisting with quartz, plagioclase and K-feldspar. The H₂O contents of these melts should approximate those of modelled eutectic melts in the Qtz-Ab-Or-H₂O system (Nekvasil and Burnham, 1987). This is demonstrated by the 850°C metagreywacke-derived melt, where the calculated melt H₂O content (wt% H₂O in melt = wt% H₂O in the bulk rock/melt fraction), in this essentially cordierite-free composition, is very close to that of the modelled eutectic melt in Qtz-Ab-Or-H₂O (Table 3). In all three metapelite compositions the melts coexisted with quartz, plagioclase and K-feldspar at 850 and 900°C. The H₂O contents of these melts could thus be confidently predicted from the eutectic compositions (Table 3). The melt H₂O contents for the runs at 950 and 1000°C are more difficult to estimate accurately. In the metapelites, K-feldspar is consumed prior to 950°C. As a result, the melts at these conditions do not approximate the eutectic composition and will be significantly more hydrous (Table 3). However, melt H₂O contents in these experiments must be lower than the ~ 1.7 wt% estimated for those at 900°C (the highest *T* at which Qtz, Pl, and Kfs coexist with melt). It has been assumed in this study that the rate of decrease in melt H₂O content is similar to that for the cordierite-free metagreywacke over the *T* interval 900 to 1000°C. This results in calculated melt H₂O contents of 1.5 and 1.3 wt% at 950 and 1000 °C, respectively.

Table 3: Calculated H₂O contents of melts (in italics) and cordierites (in parentheses) as a function of temperature and bulk-composition. Data listed under EM are H₂O contents of eutectic melts, calculated using the solubility model of Nekvasil and Burnham (1987). Data for A (in bold) are melt H₂O contents derived via a single stage calculation from the modes of the cordierite-free metagreywacke. Where these melts co-existed with quartz, plagioclase and K-feldspar (shaded blocks), the calculated H₂O contents should closely approximate that predicted by the Nekvasil and Burnham model. Data for (AS), (BS) and (CS) are assumed melt H₂O contents and calculated cordierite H₂O contents for the metapelites. Where these melts co-existed with quartz, plagioclase and K-feldspar (shaded blocks) the model eutectic melt H₂O contents (EM) can be adopted with confidence. At higher temperatures the melt H₂O contents are more difficult to constrain, introducing larger uncertainties in the calculation of cordierite H₂O content

<i>T</i> °C	AS	BS	CS	A	EM
1000	<i>1.3</i> (0.92)	<i>1.3</i> (0.90)	<i>1.3</i> (0.97)	1.81	<i>0.3</i>
950	<i>1.5</i> (1.30)	<i>1.5</i> (0.97)	<i>1.5</i> (0.89)	1.88	<i>1.1</i>
900	<i>1.7</i> (0.63)	<i>1.7</i> (0.96)	<i>1.7</i> (1.05)	2.18	<i>1.7</i>
850	<i>2.9</i> (1.30)	<i>2.9</i> (1.50)	<i>2.9</i> (1.30)	2.96	<i>2.9</i>

The cordierite H_2O content in the metapelites can be calculated from the phase proportions in Figure 1 and the bulk-rock and melt H_2O contents ($wt\% H_2O$ in Crd = $[wt\% H_2O$ in bulk rock - $(wt\% H_2O$ in melt \times melt fraction)] / cordierite weight proportion). The data indicate that the H_2O contents of cordierites, coexisting with water-undersaturated granitic melts, decrease from ~ 1.4 wt% at $850^\circ C$ to ~ 1 wt% at $900^\circ C$, then remain almost constant to $1000^\circ C$ (Table 3). In Table 3, note that the cordierite H_2O contents calculated for AS are substantially more variable than for BS and CS. Experimental data suggest that Mg# exerts no control on the H_2O content of cordierite (Weisbrod, 1973). Thus, this variability is probably due to larger errors propagated through the two-stage calculation process. This is a particular problem with composition AS, as relatively high melt-cordierite ratios result in higher potential propagated errors. Consequently, the data from BS and CS are preferred in the following discussion of cordierite-melt H_2O partitioning. Melt-cordierite H_2O partition coefficients were calculated using the relation: $D_w = H_{2O_{melt}} / H_{2O_{Crd}}$, and the variation with T is illustrated in Figure 3.

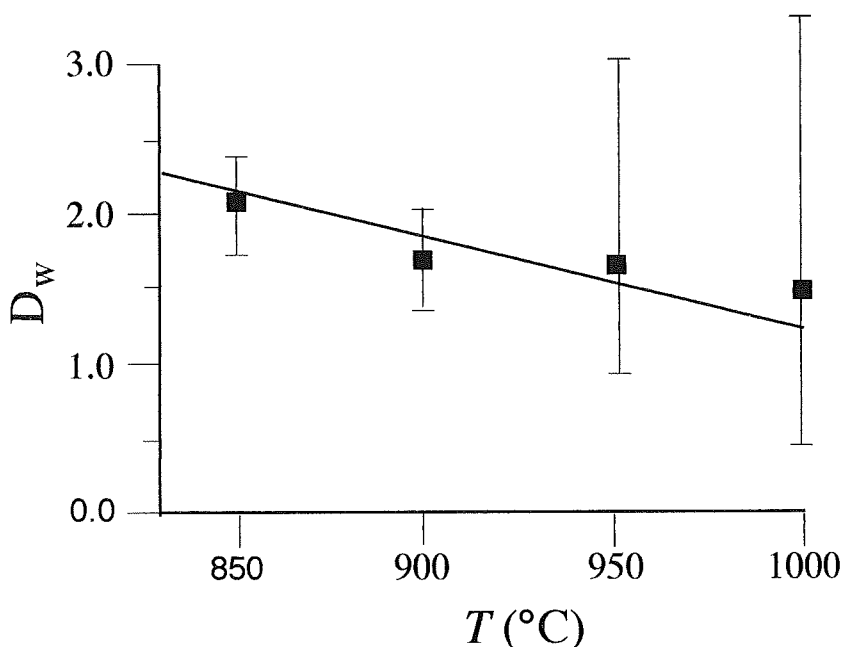


Figure 3: Calculated melt-cordierite D_w values for data averaged from compositions BS and CS. Error bars on the values at 850 and $900^\circ C$ assume negligible uncertainties for all major element microprobe totals, 3 wt% uncertainties on melt and cordierite modal proportions, and 0.2 wt% uncertainties on the melt H_2O contents. The much larger error brackets at higher T reflect an estimated 25% and 50% potential uncertainty in the melt H_2O contents at 950 and $1000^\circ C$.

DISCUSSION

During fluid-absent partial melting in the crust, H_2O is the chemical component with the greatest influence on melt fraction. The data in Table 3 show that cordierite H_2O content can strongly influence source-rock fertility, across the entire temperature range of high-grade

metamorphism. This effect will be most marked in rocks containing high modal proportions of cordierite, including aluminous metasediments metamorphosed at pressures low enough to stabilize cordierite. For average metapelite compositions ($Mg\# \approx 40$) this equates with $P \leq 0.5$ GPa, a common range for mid- to lower-crustal metamorphism and granitoid magma genesis.

These findings have significant implications for some long-held geological beliefs: (1) metapelites are typically regarded as more fertile magma sources than less aluminous metagreywackes. Indeed, S-type granites are commonly conceived as being pelite-derived; (2) high-grade cordierite is still commonly thought of as an anhydrous phase; and (3) decompression melting is regarded as a mechanism that will enhance melt productivity in metapelites. The data from this study indicate that all of these ideas are misconceptions.

During biotite fluid-absent melting, the cordierite produced is sufficiently hydrous to restrict the melt proportion severely (Fig. 4). Consequently, magnesian metapelites are poor

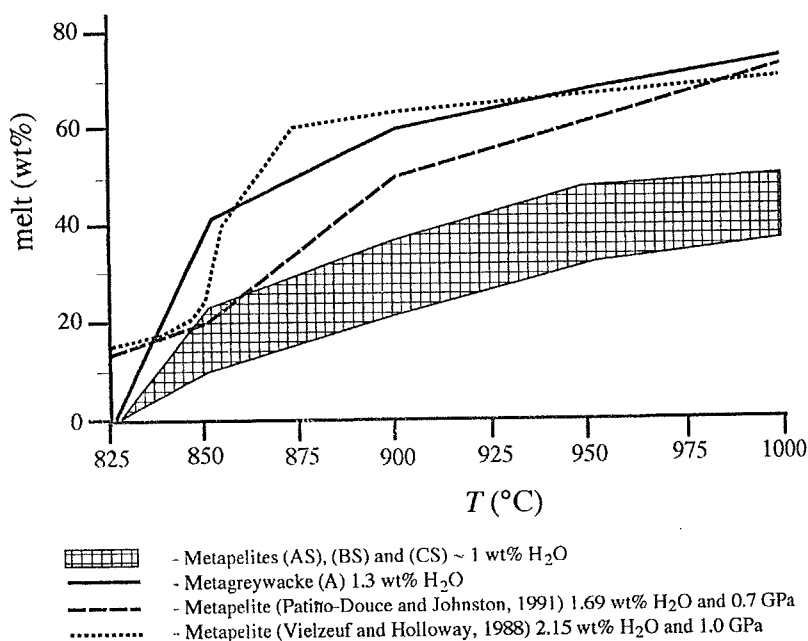


Figure 4: Plot of melt fraction versus T , from the present study. Note the metapelite CS (defining the lower limit of the hatched area contains only 25% less H_2O than the metagreywacke (A), yet produced less than half the amount of melt. This is a function of higher bulk-rock $Mg\#$ and aAl_2O_3 , resulting in formation of a high proportion of cordierite. Data from two similar studies, at 0.7 and 1 GPa on more Fe-rich bulk compositions, appear reasonably consistent with the results from the cordierite-free metagreywacke in this study. Patino-Douce and Johnston (1991) calculated melt H_2O contents, varying between 3.45 wt% (at 825°C) and 2.9 wt% (at 1075°C) for H_2O -undersaturated melts derived by 0.7 GPa partial melting of a natural metapelite. At this pressure, H_2O solubility in the melt would have been significantly higher than in the present study. Data from a similar study at 1.0 GPa (Vielzeuf and Holloway, 1988) indicated melt H_2O contents of 4.3 and 3.3 wt%, at 900 and 1000°C respectively.

candidates for the protoliths of large volumes of S-type magma unless the melting takes place at a P too high for cordierite stability. However, many voluminous S-type magmas seem to have been generated at only moderate crustal depths, within the cordierite stability field (e.g., Clemens and Wall, 1981; 1984, Phillips et al., 1981, Wyborn et al., 1981). Decompression of the anatectic magma source will certainly result in lower melt H_2O contents, favouring higher melt proportions. However, for all but very Fe-rich pelites, it will also favour higher modal proportions of cordierite relative to garnet. In suitable compositions this may completely counteract the effect of the lower solubility of H_2O in the melt. Thus, crustal differentiation by upward migration of granitic melts is most likely to occur when granulite-facies metamorphism affects greywacke-dominated terranes, or those with relatively Fe-rich pelites (see Vielzeuf and Montel, 1994).

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