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# EXPERIMENTAL STUDY OF THE ROLE OF WATER IN THE UPPERMOST MANTLE

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#### **SUMMARY**

The role of water in the uppermost mantle has been explored to 6 GPa (~190 km) by a novel experimental approach in which the silicate melting solidus, the stability of hydrous phases and the H<sub>2</sub>O-contents in nominally hydrous minerals (NAMs) were determined in each experiment. The experimental results are at variance with recent numerical models based in part on inferred high solubilities of water in NAMs. The vapor-saturated solidus (water-rich vapor) of fertile lherzolite increases in temperature (T) from a minimum of 970°C at 1.5 GPa to 1375°C at 6 GPa. Immediately above, and below the solidus the H<sub>2</sub>O-content in residual lherzolite is ~200 ppm retained in NAMs at 2.5 and 4 GPa. The hydrous mineral pargasite is the major H<sub>2</sub>Ostorage site in the uppermost mantle but pargasite is unstable at pressure (P) >3 GPa (>90-100 km depth) causing a sharp drop in water storage capacity from >2000 to ~200 ppm. For small H<sub>2</sub>O-contents (<2000 ppm approx., the solidus temperature of fertile upper mantle lherzolite decreases sharply at ~90 km depth. In oceanic intraplate settings, the geotherm passes from subsolidus pargasite-bearing lherzolite to garnet lherzolite with incipient melting, creating the rheological boundary between Lithosphere and Asthenosphere. The Asthenosphere becomes geochemically zoned with the 'enriched' intraplate basalt source (>500 ppm H<sub>2</sub>O) overlying the 'depleted' MORB source (200 ppm  $H_2O$ ). Water also plays a significant role at convergent margins where hydrous silicate melting in the mantle wedge is initiated at the vapour-saturated solidus. However, melting of lherzolite at or near the vapour-saturated solidus does not fully dehydrate residual lherzolite or harzburgite. Residual lithosphere returned to the upper mantle may carry ~100-200 ppm H<sub>2</sub>O.

The near-solidus melt (1-2%) with very low H<sub>2</sub>O-content at 2.5 GPa is transitional between sodic-dolomitic carbonatite and olivine melilitite. and is olivine-rich basanitic. With higher melt fraction at higher T or higher H<sub>2</sub>O-content it is olivine-rich basanite. At 2.5 GPa both high K/Na and low K/Na lherzolite compositions have similar vapour-saturated solidus temperatures and both K and Na are partitioned into the low melt fraction. At 4 GPa lherzolite with high K/Na has its vapoursaturated solidus at lower T than MORB-source mantle with low K/Na. However, a higher solidus temperature, the dehydration solidus for phlogopite-lherzolite, occurs if H<sub>2</sub>O-contents are lower than those for maximum phlogopite content. At 6 GPa the low K/Na mantle composition has normal melting behaviour whereas K-enriched mantle shows supercritical behaviour with continuous transition between solute-rich vapour (fluid) and H<sub>2</sub>O-rich silicate melt (fluid).

The experimental study also provides a useful data set of co-existing olivine, orthopyroxene, clinopyroxene and garnet in



'natural' lherzolite composition at pressures transitional to the diamond stability field. In using this study for geothermometry and geobarometry of natural garnet lherzolites, the presence of a  $H_2O$ -rich vapour or  $H_2O$ -rich silicate melt helps reaction rates and approach to equilibrium assemblages.

### **INTRODUCTION**

Water plays a crucial role in mantle processes, and thus influences partial melting as well as seismological and rheological properties. Water is a significant component in island arc or convergent margin volcanism and plays a major role in transfer of melts and/or fluids from the subducting slab into the overlying mantle wedge. The presence of aqueous vapour or  $H_2O$ -rich melt initiates melting close to the water-saturated peridotite solidus.

In contrast, the role of  $H_2O$  in intraplate magmas and mid-ocean ridge or divergent margin volcanism is debated. The rheology of the upper mantle, particularly the rheological distinction between the tectonically defined lithosphere and underlying asthenosphere, is considered to be very sensitive to the presence and roles of water. Models however, diverge in seeking to define the mechanically rigid and strong lithosphere and viscous or low strength asthenosphere respectively (Green et al., 2010; Hirth & Kohlstedt, 1996).

Consequently, the experimental determination of the water-saturated solidus of peridotite is essential in order to model P, T distributions in a particular subduction setting, and also to evaluate alternative models of the lithosphere/asthenosphere boundary.

## **BACKGROUND AND PREVIOUS WORK**

Early experimental determination of the water-saturated solidus of fertile lherzolite obtained a minimum of  $\sim 1000^{\circ}$ C at 1.5 to 2 GPa, with a positive dT/dP at higher P (Green, 1973;

Kushiro et al., 1968; Millholen et al., 1974). The Na+Ca-bearing amphibole, pargasite, is stable to the solidus and immediately above the solidus at P to 2.5 or 3 GPa in these three studies.

Against the background of these studies, Mysen & Boettcher (1975) and the recent studies by Grove et al. (2006) and Till et al. (2011) arrived at very different results. The water-saturated solidus of peridotite was interpreted to decrease to ~1000°C at 1 GPa, but to continue with negative dT/dP, reaching a minimum ~800°C at ~3.5 GPa and remaining below 820°C to >5 GPa. If correct, the result would mean that the water-saturated solidi for basalts, sediments and fertile lherzolite would be closely similar, within experimental uncertainty, at P around 4-6 GPa (Grove et al. 2006, Fig. 6 therein). Such a relationship would have considerable importance for understanding of relationships observed in ultra-high pressure metamorphic terranes and also for hypotheses of slab/mantle-wedge interactions in convergent margins.

## EXPERIMENTAL APPROACH AND STRATEGY

In order to test these differences in the observation and interpretation of the 'water-saturated solidus' of lherzolite (Green, 1973, 1976; Grove et al., 2006; Kushiro et al., 1968; Millholen et al., 1974; Mysen & Boettcher, 1975; Niida & Green, 1999; Till et al., 2011; Wallace & Green, 1991; Wendlandt & Eggler, 1989), this detailed experimental study aimed to clarify the effects of  $H_2O$  on the melting behaviour of lherzolite (Green et al., 2010, 2011, in review, in prep.; Kovacs et al., in review).

The investigation initially focused on the effects of varying amounts of  $H_2O$  (i.e. from nominally 'dry' to  $H_2O$ -concentrations up to 14.5 wt%) on the vapour-saturated solidus of fertile lherzolite. An essential first step is to distinguish between glass quenched from aqueous vapour



phase, from hydrous silicate melt, or from supercritical fluid.

Aqueous vapour dissolves silica, alkalis and other major elements and solubilities increase at greater mantle depth (i.e. higher P) in simple and more complex systems particularly at silica activities buffered by olivine and orthopyroxene (Green et al., 2010, in review; and references therein). In turn, quenching of vapour containing such highly dissolved components may give rise in the precipitation of siliceous glass. Thus, criteria such as textures of charges, identification of Feenriched quench phases and refractive indices of glass are used to disentangle subsolidus, vapourbearing experiments from above-solidus, silicate melt-bearing experiments as the appearance of quenched glass in high P experiments is insufficient to infer that conditions are above the silicate melt peridotite solidus. The criteria used in this study are as follows:

The presence within the lherzolite and 'melt trap' layers, of interstitial patches (interserts) of acicular or lath-shaped crystals of amphibole (and/ or quench clinopyroxene), mica and glass, all with Mg#  $\sim$ 70-85 and commonly showing Fe/Mg zoning in laths, is diagnostic for quenched hydrous silicate melt (Green, 1976; Fig. S2 in Green et al., 2010).

Conversely, the absence of such intersertal, iron-enriched quench phases, together with porous texture, planes of fluid inclusions in olivine, and stability of hydrous pargasite or phlogopite with Mg# similar to or greater than olivine, is diagnostic of subsolidus conditions and the presence of an aqueous vapour phase. The presence of glass from vapour-phase quench is particularly evident in experiments below the silicate solidus and with high H<sub>2</sub>O-contents (>5 wt%) (Green et al., 2010, 2011, in review, in prep.). Experimental charges are friable or disaggregated, freely pouring from the capsule after quenching. When observed with scanning electron microscope (SEM), these samples have euhedral olivine or pyroxene crystals coated with fragmented glass 'froth', or with films of silicate glass, and uncommon rosettes of quench carbonate (Fig. 1). These textures are indicative of vapourphase quench, the vapour containing significant dissolved oxides (Niida & Green, 1999; Green et al., 2010, 2011, in review; and references therein).



Fig. 1. Experiment O22 HZ2 lherzolite composition at 2.8 GPa, 840°C, 14.5 wt%  $H_2O$ . SEM image of broken surfaces of garnet lherzolite. Thin 'skin' of glass draped over euhedral olivine crystals. Interpreted as vapour-phase quench. See also Fig. S1 (H) in Green et al. (2010).

# EXPERIMENTAL AND ANALYTICAL METHODS

The reader is referred to Green et al. (2010, in review) regarding the description of experimental and analytical methods.

#### RESULTS

# Pargasite stability with or without an aqueous vapour phase

At 2.5 GPa, 1000°C pargasite is absent from experiments with 14.5 and 7.25 wt%  $H_2O$  but is present in all experiments with 2.9 wt% or lower  $H_2O$  contents including a nominally dry experiment. While clinopyroxene remains stable with increasing  $H_2O$ -content, the jadeite solid solution decreases and at the highest  $H_2O$ -content (14.5 wt%  $H_2O$ ) clinopyroxene has Na<sub>2</sub>O content

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d+0.2 wt. %. The absence of pargasite with 14.5 wt%  $H_2O$  reproduces Grove et al.'s (2006) observation at the same P, T condition. However with 1.45 wt%  $H_2O$  the pargasite has 3.05 wt% Na<sub>2</sub>O and 0.33 wt. % K<sub>2</sub>O. Co-existing clinopyroxene has 0.57 wt. % Na<sub>2</sub>O. The observations are consistent with the presence of a aqueous vapour phase in which the activities of  $H_2O$  increase and the activities of other components decrease (particularly Na and K which control the stability of pargasite and the jadeite solid solution in clinopyroxene).

### The vapour-saturated solidus of fertile lherzolite HZ1 + H,O (Model Upper Mantle)

Experiments with 1.45 wt%  $H_2O$  define the vapour-saturated solidus at 2.5, 4 and 6 GPa and at each P there is a clear distinction between above-solidus experiments in which interserts of quench amphibole, oxide and carbonate aggregates are present in the olivine layers, and subsolidus experiments in which such interserts are absent (Fig. S2 in Green et al., 2010).

The solidus of fertile lherzolite in the presence of a H<sub>2</sub>O-rich vapour has a negative dT/ dP from low P to a minimum between 950 and 1000°C at ~2 GPa based on our studies and earlier work, particularly Green (1973, 1976), Green et al. (2010, 2011), Millhollen et al. (1974), Niida & Green, (1999) and Wallace & Green (1991). The solidus then increases through ~1010°C at 2.5 GPa, ~1210°C at 4 GPa, and 1375°C at 6 GPa (Fig. 1 in Green et al., 2010). There is a clear distinction between hydrous silicate melt (~30 wt% H<sub>2</sub>O) above the solidus and aqueous vapour below the solidus. There is no evidence of a second critical end-point terminating the vapour-saturated solidus at 6 GPa or lower P in the HZ1 Lherzolite composition, although there is evidence that the aqueous vapour phase contains increasing solute components with increasing P. The detailed examination of run products shows co-variance of mineral and vapour phase compositions, and a



Fig. 2. Diagram at ~2.5 GPa expressing phase stability as a function of  $H_2O$ -content (note logarithmic scale) and T, based on the experimental results on HZ1 & HZ2 compositions and prior studies, particularly Niida & Green (1999) and Grove et al (2006). The experiments in which a  $H_2O$ -rich silicate melt was observed in HZ1 composition are shown by a triangle, those in HZ1 or HZ2 compositions with a  $H_2O$ -rich vapour (porous and with fluid inclusions or vapour phase quench) are shown by a circle, and 'dry' experiments (no added  $H_2O$ ) are shown by an ellipse.

transition in solidus temperature from the vapourabsent, dehydration solidus, to the vapoursaturated solidus. Increasing  $H_2O$ -contents beyond ~1.5 wt%  $H_2O$  result in dilution of dissolved components and corresponding decrease in the jadeite solid solution in clinopyroxene. Consequently as the condensed phases approach the simple CMAS system, the solidus increases in T to approx. 1100°C at 2.5 GPa (references in Green et al., in review) (Fig. 2).

### **Reconciliation of conflicting interpretations**

In investigating the 'water-saturated' melting of fertile lherzolite at high P we have duplicated the experiments and observations of Grove et al. (2006) including the observation of silica-rich glass and absence of pargasite at T of d+1000°C, and P of >2 GPa. However these relations are restricted to high  $H_2O$ -contents in experiments (>5wt%  $H_2O$  approx.). At lower  $H_2O$ -contents pargasite is stable and hydrous silicate melt is absent at 1000°C at 2.5 GPa. With increasing  $H_2O$ -contents in experiments, oxides, particularly Na<sub>2</sub>O, are leached from the solid phases into the



vapour phase. The H,O/Na,O ratio of an experimental charge is thus an important factor in determining the phase relations, including the composition and abundance of quench films of silica-rich glass (Fig. 1). If the earlier papers quoted in the introduction are examined, it is apparent that in two studies in which H<sub>2</sub>O/ Na<sub>2</sub>O>150 (mol. ratio) the authors inferred low solidus temperatures and pargasite breakdown at <2 GPa (Grove et al., 2006; Mysen & Boettcher, 1975). This interpretation was based on the observation of glass in experiments below 1000°C but the distinction between glass from vapourphase and hydrous melt phase was based on expectation 'that a quenched fluid phase would produce spherical blebs of an SiO<sub>2</sub>-rich component by unmixing from a dominantly fluid phase' (Grove et al., 2006). The glass was not analysable by electron microprobe, refractive indices were not measured and melt-trap techniques were not used. No analyses of glass or of quench aggregates below 1000°C were obtained. One 'melt' composition was reported (Grove et al., 2006) from an experiment at 1.2 GPa, 1020°C which is above the solidus in all previous work and not in dispute. For this experiment, the glass composition presented is silica-rich and low in MgO and FeO. The composition is similar to glasses analysed by Green (1976, and references therein) at 1-2 GPa. These glasses are not equilibrium melt compositions and were shown to be quenchmodified (by reversal experiments; Green, 1976). A similar test is warranted for the Grove et al. (2006) composition which should not be used as argument for production of silica-oversaturated melts by hydrous melting of lherzolite, particularly from the mantle wedge overlying subduction zones.

# The role of pargasite-bearing lherzolite in the uppermost mantle

Our results duplicate and confirm the

stability of pargasite, its composition and modal abundance in fertile lherzolite as described in detail by Niida & Green (1999) in the MOR Pyrolite composition. The experimental data, summarized in Fig. 2, also show that there is a restricted T field between 1025 and 1100°C approx. in which it is possible to pass through the solidus from subsolidus pargasite lherzolite to partially molten garnet lherzolite without change of P or T, by increasing H<sub>2</sub>O-content from <0.1 wt% H<sub>2</sub>O to ~0.5 wt% H<sub>2</sub>O. This process of *'isothermal hydrous melting'* occurs if additional H<sub>2</sub>O migrates into a pargasite lherzolite which is at T *below* the dehydration solidus but *above* the vapour-saturated solidus.

At P less than 3 GPa i.e. within the pargasite stability field, changes in P and/or increase or decrease in  $H_2O$ -activity, have complex effects on pargasite composition and modal abundance. The effect of increasing P (subduction) on pargasite lherzolite will release aqueous vapour as the modal abundance of pargasite decreases and the pargasite becomes more potassic and sodic. In the reverse process at P <3 GPa, an upwelling (from 3 to 2 GPa for example) of lherzolite containing incipient melt, will freeze with increased pargasite content if T is <1100°C. These complexities of melting vs. pargasite stability have applications in interpreting the mineralogy and geochemistry of upper mantle samples.

# Phlogopite stability field

If lherzolite is enriched in  $K_2O$  then phlogopite may crystallize in addition to pargasite at P below 3 GPa, and in this case pargasite has ~1.1 wt%  $K_2O$ , i.e. higher than in the absence of phlogopite. In K-enriched compositions phlogopite is stable to the vapour-saturated solidus at 2.5 GPa. For lower H<sub>2</sub>O-contents, phlogopite presence depends on  $K_2O$ , Na<sub>2</sub>O and H<sub>2</sub>O activity and at low H<sub>2</sub>O activity, pargasite + phlogopite lherzolite is stable to a higher T dehydration solidus (>1100°C at 2.5 GPa) (Mengel & Green,



1989). Phlogopite in lherzolite is stable to higher P than pargasite, i.e. up to P of <10 GPa, again depending on bulk lherzolite composition (e.g. Green, 1973; Green et al., in prep., Mengel & Green, 1989; Wendlandt & Eggler, 1980; and references therein).

## APPLICATIONS TO MAGMA GENESIS AND GEODYNAMICS

The lithosphere/asthenosphere boundary

The H<sub>2</sub>O-storage capacity of the model mantle composition may be defined as the amount of H<sub>2</sub>O which is structurally bound in stoichiometric components in hydrous minerals stable at the P, T of interest, together with that held in solid solution in crystal lattices of NAMs. The H<sub>2</sub>O-storage capacity, thus defined, is a useful indicator of whether an aqueous vapour phase, hydrous silicate melt or supercritical H<sub>2</sub>O-rich fluid will be present for a given lherzolite composition and H<sub>2</sub>O-content. In fertile lherzolite compositions the amphibole pargasite and the mica phlogopite are stable to solidus temperatures even in the presence of very small H<sub>2</sub>O-contents. Pargasite however is unstable at P greater than 3 GPa and phlogopite only occurs if K<sub>2</sub>O contents are enriched above those appropriate to MORBsource compositions (such as MOR Pyrolite or HZ1 Lherzolite). Thus the H<sub>2</sub>O-storage capacities of inferred mantle compositions are sensitively dependent on stabilities of hydrous phases i.e. on bulk composition, P and T. The H<sub>2</sub>O-storage capacity of HZ1 lherzolite decreases from ~3-4000 ppm at 1.5 GPa to  $\sim$ 2000 ppm at 2.5 GPa as the modal pargasite changes as a function of P (Niida & Green, 1999) but then drops abruptly to ~200 ppm at ~3 GPa as pargasite is destabilised by P.

Focussing attention on upper mantle compositions represented by the global MORBsource mantle, or modern, well-mixed upper mantle (i.e. compositions close to HZ1 Lherzolite or MOR Pyrolite), then estimated geothermal gradients in mature oceanic and young continental regions of lithospheric plates intersect the mantle solidus and pargasite breakdown reaction (Green et al., 2010). Such geotherms pass from subsolidus to partially molten lherzolite at ~90 km depth, provided that the intraplate upper mantle contains >2000 ppm H<sub>2</sub>O. Recent work (Faul & Jackson, 2007) on mantle rheology demonstrates that very small degrees of partial melting have a marked effect on dunite rheology, increasing strain rates by 2 orders of magnitude. It is inferred that the most plausible explanation for the rheological contrast between lithosphere and asthenosphere is the transition along the geotherm from subsolidus pargasite lherzolite to partially molten garnet lherzolite, the melt fraction present being determined primarily by the H<sub>2</sub>O-content (Green, 1973; Green et al., 2010, in review, and references therein).

Our data are also relevant to the suggestion (Hirth & Kohlstedt, 1996) that the asthenosphere is subsolidus H<sub>2</sub>O-bearing mantle and is rheologically weak because of intracrystalline H<sub>2</sub>O in olivine particularly. The overlying lithosphere was inferred to be 'dry' having lost intracrystalline H<sub>2</sub>O into a melt phase in the upwelling and melt extraction in lithosphere formation at mid-ocean ridge settings (Hirth & Kohlstedt, 1996). The evidence presented in Green et al. (2010) and companion papers, is that H<sub>2</sub>Ocontents in NAMs change little across the vapoursaturated solidus. It is suggested that high viscosity or rigidity of the lithosphere is not due to absence of H<sub>2</sub>O in NAMs but to the stability of hydrous phases pargasite and phlogopite when H<sub>2</sub>Ocontent exceeds that which can be held in NAMs i.e. ~200 ppm. Similarly, low viscosity in the asthenosphere is not due to higher H<sub>2</sub>O-content in NAMs but to the presence of incipient melt in mantle with >200 ppm H<sub>2</sub>O approximately. In our preferred model of the lithosphere and asthenosphere (Green et al., 2010, in review; and references therein) the MORB source lies at



deeper levels in the asthenosphere and has the character of garnet lherzolite depleted by extraction of small melt fraction(s) at the water-saturated solidus at >3 GPa.

# 'Wet mantle', island arc petrogenesis, recycled lithosphere and the depleted MORB source.

Accepting that the subduction process transports  $H_2O$  into the mantle wedge, the experimentally determined phase relations for lherzolite +  $H_2O$  can be superimposed on

geophysical models of P, T variation. This has been done previously using the phase relations for the model 'Pyrolite' composition (Niida & Green, 1999; Green et al., in review, and references therein) and, given the close similarity of HZ1 and MOR Pyrolite compositions, the detail of Fig. 6 in Niida & Green (1999) is equally applicable to the HZ1 composition as a model composition in the mantle wedge setting. The significance of the depth interval near 80-100 km i.e. pressure P of 2.5 to 3 GPa, is emphasized as a 'step' or rapid change in the H<sub>2</sub>O-storage capacity of the mantle wedge lherzolite. Immediately above the slab and at shallower depths, H<sub>2</sub>O water or hydrous silicate melt migrating up-temperature into the wedge from the slab can be 'captured' and stored in fertile lherzolite below ~1100°C up to ~0.5 wt% (20-30 % pargasite). Less modal pargasite and lower H<sub>2</sub>O-storage capacity apply to more refractory lherzolite (Wallace & Green, 1991). Pargasite-bearing lherzolite formed at <100 km and dragged downward by the subducting slab, will melt at ~100 km depth, possibly initiating mantle upwelling and consequent magmatism.

A very important result from this study (see also Green et al., 2010, 2011; Kovács et al., in review) is the recognition that if melting and melt extraction in subduction settings, does occur at or near the vapour-saturated solidus (aqueous vapour) then the residue from such melting is not 'dry' and if carried deeper into the mantle will

take up to  $\sim 200$  ppm H<sub>2</sub>O with it into the deeper mantle. Also, subduction of intraplate lithosphere containing pargasite will lead to pargasite breakdown to NAMs + vapour +/- phlogopite at the base, or in the deeper parts, of the slab at  $\sim 3$ GPa (Green 1973; Niida & Green, 1999; Green et al., 2010) but ~150-200 ppm H<sub>2</sub>O will remain for recycling into the mantle. It is probably not co-incidental that the source regions for MORB are inferred to contain ~100-200 ppm H<sub>2</sub>O (references in Green et al., 2010) i.e. an amount matching lherzolite residue from melting near the vapour saturated solidus, whether formed in the intraplate asthenosphere or mantle wedge. The LREE and LILE depleted character of the MORB source coupled with a H<sub>2</sub>O-content of ~100-200 ppm H<sub>2</sub>O is consistent with one or many events of near-solidus melting and melt migration. The upwelling of such depleted material from the deeper mantle at high T (mantle potential T ~1430°C) will yield parental MORB with ~0.1-0.2 wt% H<sub>2</sub>O and residual lherzolite to harzburgite will have <10 ppm H<sub>2</sub>O, reflecting high T of H<sub>2</sub>Opartitioning and the very low H<sub>2</sub>O-content of parental MORB (Kovács et al., in review). As the depths of segregation of primitive MORB from residual lherzolite, (i.e. the extraction of melt from a permeable flow regime or from an ascending diapir, and its movement in dykes or dunite channels) is at 1.5 to 2 GPa, new residual lithosphere formed at these depths is 'dry' (as in Hirth & Kohlstedt, 1996). The establishment of the pargasite-controlled Lithosphere/ Asthenosphere Boundary and thickening of the lithosphere with age are attributed to cooling and H<sub>2</sub>O access to the lithosphere as it moves away from the ridge setting.

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