

# 17 The nature and role of fluids in the upper mantle: evidence in xenoliths from Victoria, Australia

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## ABSTRACT

Mantle-derived xenoliths contain direct evidence of the nature of mantle melts (as veins of frozen basaltic melt in mantle wall-rock) and of mantle fluids (trapped in cavities and including fluid inclusions in mantle minerals). Volatile-bearing phases (amphibole, mica, apatite) in mantle Cr-diopside lherzolites provide structural sites for CO<sub>2</sub>, Cl, F, H<sub>2</sub>O and S; these phases are also generally enriched in incompatible elements. Microthermometry and Raman laser microprobe methods indicate that CO<sub>2</sub> is the dominant fluid now present in the fluid inclusions. However, OH-, CO<sub>2</sub>- and Cl-bearing phases lining fluid inclusion and cavity walls are inferred to have formed by reaction, implying the original presence of up to 15% H<sub>2</sub>O, and small amounts of Cl. Mass spectrometric analyses reveal the presence of small quantities of He, COS, SO<sub>2</sub>, N<sub>2</sub>, O<sub>2</sub>, Ar, H<sub>2</sub>S and hydrocarbons. The <sup>3</sup>He/<sup>4</sup>He ratios are 7-10 times atmospheric, indicating a He contribution from degassing of a deep primitive reservoir. <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>143</sup>Nd/<sup>144</sup>Nd ranges in volatile-rich mantle rocks also suggest the existence of at least one primitive reservoir. Sulphide globules in the clinopyroxenes of some mantle-derived xenoliths are associated with CO<sub>2</sub>-rich fluid inclusions. Their microstructure suggests that a sulphide fluid, a CO<sub>2</sub>/H<sub>2</sub>O fluid and silicate magma may coexist as immiscible liquids under certain mantle conditions.

Keywords: carbon dioxide, fluid inclusions, helium isotopes, mantle fluids, metasomatism, primordial fluids, sulphide immiscibility.

## INTRODUCTION

The occurrence of a fluid phase as a discrete component of the upper mantle is of crucial importance in interpreting the mechanisms and geochemical effects of mantle metasomatism and of anatexis in the mantle and lower crust (e.g. Mysen 1983; Olafsson & Eggler 1983; Andersen *et al* 1984). The nature of this fluid also has fundamental implications for the large-scale structure and dynamics of the Earth's mantle, its convection style, the consequences of mantle degassing, and the existence of a deep, primitive reservoir (Porcellii *et al*, in press; Craig *et al* 1978, Kurz & Jenkins 1981; Kyser & Rison 1982). The presence of a fluid component may also be a controlling factor in the incorporation of high-density xenoliths in basaltic melts (Andersen *et al* 1984) and in their continued suspension in lower-density host magma during transport to the surface.

Mantle-derived rock types (mainly high-pressure xenoliths entrained in basaltic and kimberlitic magmas) provide abundant evidence that there are two end-member type liquids active within the mantle. The first type includes silicate melts ± free or dissolved volatiles ± dissolved or immiscible sulphide melts (i.e. basaltic liquids). The second type includes free fluid phases (volatile fluxes). As silicate partial melts may contain varying proportions of volatiles, there is a complete spectrum between these two end-members.

## EVIDENCE FOR TRAPPED MELTS AND FLUIDS IN THE MANTLE

Evidence for the presence in the mantle of frozen partial melts is provided by composite xenoliths (Fig. 1a) where mantle wall-rocks (Cr-diopside spinel lherzolite) are veined by, or are in contact

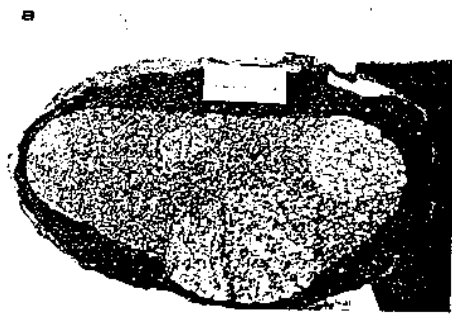


Fig. 1a Mantle xenolith from Mt Leura, western Victoria. Lighter Cr-diopside spinel lherzolite is veined by darker Al-augite pyroxenite. Scale bar intervals are 1 cm.



Fig. 1b Veins of Al-augite pyroxenite in Cr-diopside lherzolite in a tectonically-emplaced mantle-derived rock unit near Balmuccia in the Ivrea Zone, Italian Alps.

with, pyroxenites, wehrlites or some other modal variant of the Al-augite (Type II) xenolith series (Wilshire & Shervais 1975; O'Reilly & Griffin 1986). These latter xenoliths represent cumulates, or melts of basaltic liquids, which crystallized within the mantle (Irving 1980).

Further evidence of the entrapment of basaltic liquids within the mantle occurs in outcrops of upper mantle rocks tectonically emplaced in the upper crust. Examples of exposed complexes are the Ivrea Zone in the Italian Alps (e.g. Mehnert 1975; Rivalenti *et al.* 1984) and some terranes where peridotite is associated with eclogite pods or layers as in Norway (Griffin & Carswell 1985), Italy (Möckel 1969), and Czechoslovakia (e.g. Misar *et al.* 1984). Such outcrops have the advantage that large-scale rock-type relationships can be observed (Fig. 1b).

Evidence for the presence of a free fluid phase is provided directly in mantle-derived xenoliths by fluid inclusions trapped in minerals at high-pressure, and indirectly by volatile-bearing mineral phases formed by reaction of fluids with mantle rocks or by fluid infiltration and precipitation.

#### Fluid inclusions and vugs

High-pressure xenoliths from southwestern Victoria, Australia, contain abundant fluid inclusions with trapped fluids and cavities which were fluid-filled at high pressure. It is these xenoliths which provide the data for this study. However, xenoliths

from Hawaii, Spitsbergen, western USA, the Eifel (Germany), the Massif Central (France) and other regions in eastern Australia, show similar phenomena (authors' unpublished data).

Three types of xenoliths from western Victoria contain fluids trapped at high pressure: Cr-diopside spinel lherzolites, wehrlite series rocks and garnet and spinel pyroxenites. Cr-diopside lherzolites represent mantle wall-rock; wehrlite series rocks represent frozen basaltic liquids or cumulates; and garnet and spinel pyroxenites are recrystallized, metamorphosed wehrlite series rocks which have re-equilibrated by cooling from igneous temperatures to the temperature on the ambient geotherm appropriate to their depth. Full details of these rock types are given by O'Reilly & Griffin (1986).

Gas cavities in these rocks may constitute up to 3% by volume, and range upwards in size from typical fluid inclusions, commonly 10–30 microns across (Fig. 2c). These fluid inclusions show a range of morphologies. Many have negative crystal shapes and others have undergone decrepitation due to pressure release and/or heating and have haloes of secondary, low-pressure fluid inclusions. Small, dark-looking fluid inclusions without decrepitation haloes have the highest densities (determined by microthermometry) corresponding to a trapping pressure of 14–15 kbar (Andersen *et al.* 1984). These may represent the highest detectable trapping pressure as the mechanical strength of the host mineral is the limiting factor (Roedder 1984).

In addition to these 'normal' fluid inclusions,



Fig. 2a Irregularly-shaped vugs (arrowed) inferred to be fluid-filled at high pressure, in pyroxenite xenolith from Lake Bullenmerri, western Victoria.



Fig. 2b Amphibole lining fluid cavity in mantle-derived wehrlite xenolith from Lake Bullenmerri, western Victoria. Note euhedral crystal outlines in cavity. Width of photomicrograph is 1.3 mm.



Fig. 2c Fluid inclusions showing a variety of morphologies in a clinopyroxene grain in a Cr-diopside lherzolite xenolith from Lake Bullenmerri, western Victoria. Types visible include one- and two-phase inclusions with negative crystal outlines; small round, dark inclusions (densest); two-phase reticulate inclusions; and tiny secondary inclusions due to decrepitation. Width of photomicrograph is 0.13 mm.

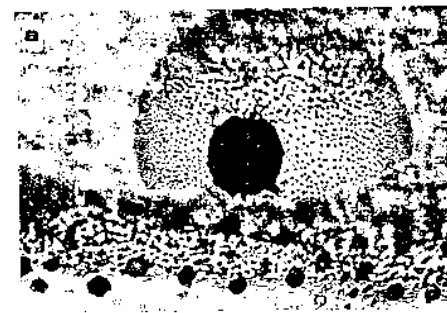


Fig. 3a Sulphide inclusions (originally fluid, now solid). Large inclusion is surrounded by a plane of reticulate and discrete inclusions. Compare with CO<sub>2</sub> inclusion morphologies in Fig. 2c. Width of photomicrograph is 0.35 mm. Transmitted light.

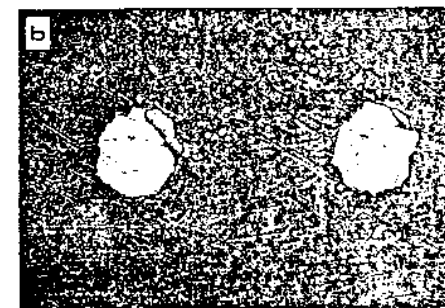


Fig. 3b Reflected light reveals the sulphides have been adjacent to a fluid phase now escaped and evident as holes in the polished surface, at upper right of both large sulphide inclusions. Width of photomicrograph is 0.35 mm.



Fig. 3c The sulphide inclusions are arranged in a grid-like pattern parallel to growing crystal surfaces in the host clinopyroxene. Width of photomicrograph is 2.8 mm.

there are large polygonally-shaped gas cavities which are situated at grain boundaries and form an integral part of the microstructure. There are also vugs up to 1.5 cm across which are inferred to have been filled with a fluid phase in the mantle (Andersen *et al* 1984; O'Reilly 1987) (Fig. 2a). These vugs are commonly lined with high-pressure minerals which are euhedrally terminated into the cavity (Fig. 2b).

#### Volatile-bearing mineral phases in mantle rocks

The most abundant upper mantle rock type in eastern Australia is anhydrous Cr-diopside spinel lherzolite (Wass & Irving 1976) consisting of the four-phase assemblage of olivine, orthopyroxene, clinopyroxene and spinel. The presence of volatile-bearing mineral phases in such rocks is generally considered to be the result of metasomatism by fluids or fluid-rich silicate melts (Menzies & Murthy 1980; Wass & Rogers 1980; Jones *et al* 1983; O'Reilly & Griffin 1984; Roden *et al* 1984). Volatile-bearing phases present in these lherzolites are amphiboles (pargasites and kaersutites), micas (phlogopites and biotites), apatite (carbonate-bearing hydroxylchlorapatite) and rare carbonate (dolomite which may be Fe-bearing). This is *a priori* evidence for the introduction of at least OH, F, Cl, P<sub>2</sub>O<sub>5</sub> and CO<sub>2</sub>.

Other mantle-derived xenoliths belonging to the amphibole/apatite series are almost totally composed of similar volatile-bearing minerals (Wass 1979; O'Reilly 1986; O'Reilly & Griffin 1986). These are considered to have crystallized within the mantle from a volatile-rich, differentiated kimberlitic magma. Such a magma may represent a metasomatizing agent rather than a product of metasomatism (Wass & Rogers 1980; O'Reilly & Griffin 1984).

#### COMPOSITION OF MANTLE FLUIDS

##### Evidence from fluid inclusions

##### Oxides, halogens and hydrocarbons

Fluid inclusions in the xenoliths studied are considered to represent primary fluids trapped within the mantle and uncontaminated by volatiles from the host basaltic magma. The density of primary CO<sub>2</sub> fluid inclusions is  $> 1.19 \text{ g/cm}^3$

(Andersen *et al* 1984). Some show partial decrepitation during ascent and heating. These observations, along with the great abundance of trapped high-pressure volatiles provide evidence that the fluid pressure within the xenoliths greatly exceeded the fluid pressure in the host magmas. Hence, these xenoliths have cegassed into the enclosing magma and are unlikely to be contaminated by introduction of volatiles from the magma. This contrasts with some xenoliths from the Eifel, Germany (E. Jagoutz, pers. comm.) which contain no visible primary fluid inclusions, but show dendritic cracks filled with fluid inferred to have been injected from the host magma. These xenoliths apparently did not have significant internal fluid pressure.

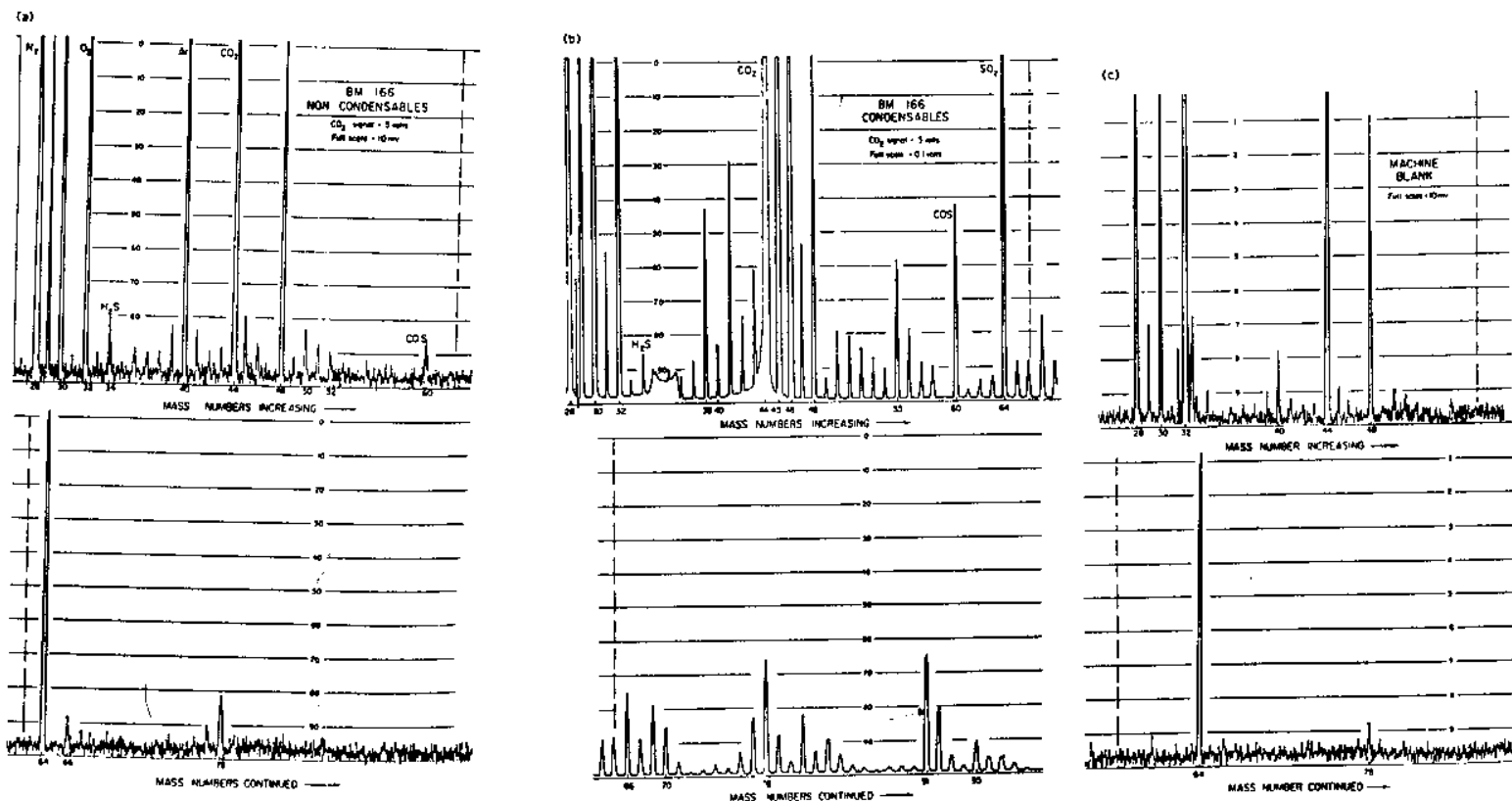
Microthermometry was carried out on about 60 fluid inclusions, and Raman spectroscopy on fluid inclusions from five rocks from western Victoria

(Andersen *et al* 1984). As is usual for mantle rocks (Roedder 1965; Murck *et al* 1978; Miller & Richter 1982), CO<sub>2</sub> is the dominant phase now present.

Scanning-electron microscope techniques were also used to show the detailed topography of the fluid inclusions. Combined use of SEM and energy-dispersive microprobe analyses showed that fluid inclusions on walls commonly are lined with euhedral crystals (usually Fe-rich and Ca-rich amphiboles with significant Cl contents) coated with Fe-rich dolomite (Andersen *et al* 1984). These crystals appear to have formed by reaction of the cavity wall (usually host clinopyroxene) with the original fluid trapped in the cavity. This means that the CO<sub>2</sub> observed now, is residual after post-entrapment reaction. This has resulted in removal of at least OH, Cl, F and possible CO from the original trapped fluid.

Calculations show that about 15% of H<sub>2</sub>O may have been present before reaction (Andersen *et al* 1984). Some of the gas-rich rocks were crushed under vacuum and the condensable and noncondensable fractions measured on a mass spectrometer. This crushing technique eliminates the possibility of synthesis of gases due to heating, which can occur if a thermal release method is used.

The mass-number peaks of the noncondensable fraction for a lherzolite are shown in Fig. 4a. Fig. 4b shows peaks for the condensable fraction. Gas species positively identified are N<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>O, CO<sub>2</sub>, Ar, SO<sub>2</sub> and COS and hydrocarbons. Some small peaks corresponding to the presence of HCl, NO and N<sub>2</sub> and O<sub>2</sub> may be due to air contamination in the spectrometer. Peaks at multiple mass-numbers for multiple C and H combinations are recorded in Fig. 4b (compare with machine blank in Fig.



Figs 4a-c See text for discussion.

4c). Gas chromatography on the condensable fraction of sample DR10162 (Table 1) showed no methane or ethane but the instrument sensitivity is low for these mass-members. However propane (i- and n-), butane (i- and n-), hexane (i-) and pentane (i- and n-) were all present at the 1–15 ppm level. All hydrocarbons detected are aliphatic and not aromatic, thus excluding contamination from surface or biological sources. The mass spectrometer traces (Fig. 4b) also confirm the presence of these hydrocarbon species, with more up to mass-number 100. This method of analysis could cause cracking of hydrocarbons on the filament so these results strongly imply the presence of hydrocarbons with mass-numbers greater than 100. As hydrocarbons other than methane and ethane are predicted to be unstable under mantle conditions (W.R. Taylor, pers. comm.), the higher mass numbers may be artifacts of the lower pressure environment but the presence of significant C and H is confirmed. This apparent conflict cannot be resolved with the data available, but highlights problems to be solved by further work. Hydrocarbons have been previously recorded in mantle-derived material such as diamonds (Giardini & Melton 1975). Such nonbiogenic hydrocarbons may be a significant source for some high-level oil and gas deposits (Giardini *et al.* 1982; Green 1985) if a given tectonic environment can tap and concentrate appropriate mantle fluids.

Isotopic data were obtained for CO<sub>2</sub> from three mantle xenoliths (Table 1). The  $\delta^{13}\text{C}$  values range from -2.9 to -8.7, but the value may reflect contamination from the very abundant hydrocarbons in that sample. These values are very close to the restricted 'heavy' range of  $\delta^{13}\text{C}$  values of -4 to -8 which characterizes the majority of mantle-derived material in which carbon is a major element, such as in CO<sub>2</sub>, diamonds and carbonates (Pineau & Javoy 1983). The  $\delta^{18}\text{O}$  values for the CO<sub>2</sub> are +36.1 to +51.5 and are essentially within the range of +8.9 to +41.4 shown to represent  $\delta^{18}\text{O}$  of the CO<sub>2</sub> (inferred to be primary) in mid-ocean ridge basalts (Moore *et al.* 1977; Pineau & Javoy 1983).

#### Helium isotopes

The isotopic composition of He was also obtained for these xenoliths (Porcelli *et al.* 1986). The selected xenoliths are so fluid-rich that they yield

TABLE 1 Summary of isotopic data and hydrocarbon abundances for three mantle xenoliths.

Sample	$\delta^{13}\text{C}_{\text{CO}_2}$	$\delta^{18}\text{O}_{\text{CO}_2}$
DR10162	-7.0	+46.5
BM166	2.9	+51.5
BM901	-8.7	+36.1
DR10162	Garnet pyroxenite, Lake Bullenmerri, western Victoria. Hydrocarbons not abundant at a CO <sub>2</sub> signal level of 0.8 V	
BM166	Garnet pyroxenite, Lake Bullenmerri, western Victoria. Hydrocarbons very abundant (Fig. 4) at a CO <sub>2</sub> signal level of 0.5 V, and may be a source of interference for the isotopic measurements.	
BM901	Cr-diopside spinel lherzolite with amphibole and apatite. Hydrocarbons abundant at a CO <sub>2</sub> signal level 0.5 V.	

Isotopic analyses were carried out on the Finnegan MAT 251 mass spectrometer by T.V. Segalstad at the Institute for Energy Technology at Kjeller, Norway.

Hydrocarbons were also detected on the above Finnegan MAT 251 mass spectrometer and by gas chromatography carried out by Einar Brevik (at the Institute for Energy Technology) on sample DR10162 using a Porapak Q column.

relatively large volumes of these gases ( $10^{-8}$  to  $10^{-6}$  atm.). The He appears to occur in the fluid inclusions rather than in structural sites in minerals as there is an empirical correlation between the presence of fluid inclusions and He abundance in both mineral separates and whole rocks.

<sup>4</sup>He is continuously produced by Uranium decay series reactions but <sup>3</sup>He represents primordial gas trapped during Earth's accretion (Clarke *et al.* 1969). The xenoliths measured yield <sup>3</sup>He/<sup>4</sup>He(R) ratios which are 7 to 10 times the present atmospheric level (Ra). Because of the young eruption ages of the host magma (O'Reilly & Griffin 1985), negligible <sup>4</sup>He would have evolved since eruption and thus these ratios reflect pristine mantle values.

The R/Ra values of 7 to 10 for these xenoliths (Fig. 5) coincide with the range for other xenoliths from continental areas and for mid-ocean ridge basalts (MORB) as shown in Porcelli *et al.* (1986). These results imply that He found in xenoliths from widely-distributed continental areas and from MORB, is derived from a similar well-mixed source with a significant component of primordial He. This further suggests that there must be a deep reservoir which has not totally

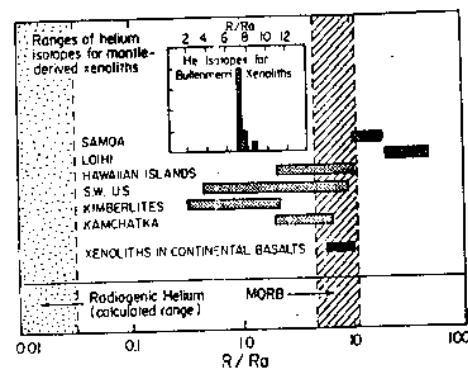


Fig. 5 Main diagram shows R/Ra ( $R = ^3\text{He}/^4\text{He}$  in sample,  $R_a = ^3\text{He}/^4\text{He}$  in the atmosphere) values of mantle-derived xenoliths from continental regions and oceanic islands. Sources are given in Porcelli *et al.*, 1986. Inset shows R/Ra values for xenoliths from western Victoria (Porcelli *et al.*, 1986).

degassed during the evolution of the upper mantle by partial melting.

#### Sulphides

Sulphides represent an additional fluid phase within the mantle. Spherical and cylindrical inclusions of sulphides are common in clinopyroxenes in upper mantle rocks (Andersen *et al.* 1987). They are especially abundant in, but not confined to, those high-pressure rocks that formed as frozen basaltic melts at high pressure. The sulphides are pyrrhotite and chalcopyrite, with rare pyrite (the latter being confined to some unusual relatively oxidised rock types). These sulphides show the same range of morphologies as CO<sub>2</sub>-rich fluid inclusions. They range from small spherical droplets to those with decrepitation haloes (Fig. 3a).

The compositions of the sulphide-bearing clinopyroxenes from different localities worldwide are remarkably uniform. They are Fe-rich and Cr-poor, suggesting crystallization from relatively evolved liquids. This is consistent with the interpretation that a sulphide liquid became immiscible at a certain stage of evolution of the parent magmas.

The sulphide droplets are commonly aligned in a grid-like pattern, parallel to growth planes in the host clinopyroxene (Fig. 3c). Some sulphide

droplets contain, or adjoin, trapped CO<sub>2</sub> fluid inclusions (Fig. 3b). All of these characteristics strongly suggest that under certain conditions, three immiscible fluids (CO<sub>2</sub>/H<sub>2</sub>O, sulphide, and silicate melt) may coexist in the upper mantle (Andersen *et al.*, in press).

#### Evidence from volatile-bearing minerals

Most data are available for the amphibole/apatite series xenoliths referred to above. They have high incompatible element abundances (especially Ti, K, P, REE), high volatile contents (mainly CO<sub>2</sub>, Cl, F, H<sub>2</sub>O, S). The Cl and F are largely accommodated in apatite, CO<sub>2</sub> in primary carbonate and S in sulphides (Wass 1979). Isotopically the amphibole/apatite xenoliths show a restricted range in <sup>87</sup>Sr/<sup>86</sup>Sr (0.70369–0.70398) and in <sup>143</sup>Nd/<sup>144</sup>Nd (0.512583–0.512716) (Wass & Shaw 1984; Menzies & Wass 1983). On a conventional <sup>143</sup>Nd/<sup>144</sup>Nd vs <sup>87</sup>Sr/<sup>86</sup>Sr diagram they plot to the left of the 'mantle array' and trend downwards parallel to the  $\epsilon$  Nd axis.

Preliminary isotopic data for metasomatized Cr-diopside lherzolites from eastern Australia (authors' unpublished data) suggest varying degrees of mixing of at least two components with mantle wall-rock. One component has high <sup>87</sup>Sr/<sup>86</sup>Sr up to 0.715, with nonradiogenic <sup>143</sup>Nd/<sup>144</sup>Nd around 0.51280.

The presence of amphibole  $\pm$  mica  $\pm$  apatite in these lherzolites also correlate with high incompatible element contents (especially Ba, Sr, Y and REE) and high volatile contents (e.g. P<sub>2</sub>O<sub>5</sub>, CO<sub>2</sub>, Cl, F, H<sub>2</sub>O).

#### SIGNIFICANCE OF MANTLE FLUIDS

The nature of mantle fluids is of fundamental significance to a wide spectrum of geological phenomena.

(a) Mantle fluids provide a mechanism for movement of heat in the upper mantle and lower crust. They may initiate partial melting in the upper mantle to yield basaltic liquids. They may invade the lower crust and promote granulite-facies metamorphism by dehydration and heat transfer (Touret 1974; Schilling & Kreulen 1979; Newton *et al.* 1980; Andersen *et al.* 1984).

(b) The presence of mantle volatiles may be an important factor in the fragmentation of some

mantle horizons, and hence the entrainment of selected xenoliths. Empirically, xenoliths characterized by abundant fluid contents commonly occur in explosively erupted host rocks (Andersen *et al.* 1984). Gas-rich mantle horizons may fragment due to degassing in contact with an ascending magma. This would result both in entrainment of fragments from these mantle horizons and in increased volatile content of the host magma. Increased volatile content will increase ascent velocity (Spera 1984) not by a propellant effect, but by determining the local magma pressure and the dynamics of advance crack propagation.

(c) Mantle fluids represent the mechanisms of transport and concentration of certain elements in the mantle. They are therefore the agents of mantle metasomatism and control its geochemical character. CO<sub>2</sub>/H<sub>2</sub>O fluids (Mysen 1983) and silicate melts (e.g. O'Reilly & Griffin 1984) determine the movement of REE and LIL elements, while sulphide liquids determine the distribution of chalcophile elements.

(d) Noble gas isotopic ratios (e.g. <sup>3</sup>He/<sup>4</sup>He) provide evidence for mantle evolution and convection models. The presence of significant <sup>3</sup>He suggests that degassing of a deep, primitive reservoir (the lower mantle?) is continually taking place (Porcelli *et al.* 1986). This also implies convective isolation of this deep reservoir from the upper mantle during the evolution of the upper mantle through partial melting events.

(e) The degassing of mantle fluids is the ultimate source of all the fluids and volatiles within the Earth's crust, on its surface and in its atmosphere.

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## NOTE ADDED IN PROOF

This paper was first submitted in 1985; interpretations are therefore four years out of date. No attempt has been made to update the manuscript.