



High-pressure phases in shock-induced melt veins of the Umbarger L6 chondrite: Constraints of shock pressure

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Abstract—We report a previously undocumented set of high-pressure minerals in shock-induced melt veins of the Umbarger L6 chondrite. High-pressure minerals were identified with transmission electron microscopy (TEM) using selected area electron diffraction and energy-dispersive X-ray spectroscopy. Ringwoodite (Fa_{30}), akimotoite ($\text{En}_{11}\text{Fs}_{89}$), and augite ($\text{En}_{42}\text{Wo}_{33}\text{Fs}_{25}$) were found in the silicate matrix of the melt vein, representing the crystallization from a silicate melt during the shock pulse. Ringwoodite (Fa_{27}) and hollandite-structured plagioclase were also found as polycrystalline aggregates in the melt vein, representing solid state transformation or melting with subsequent crystallization of entrained host rock fragments in the vein. In addition, Fe_2SiO_4 -spinel ($\text{Fa}_{66}\text{-Fa}_{99}$) and stishovite crystallized from a FeO-SiO_2 -rich zone in the melt vein, which formed by shock melting of FeO-SiO_2 -rich material that had been altered and metasomatized before shock. Based on the pressure stabilities of the high-pressure minerals, ringwoodite, akimotoite, and Ca-clinopyroxene, the melt vein crystallized at approximately 18 GPa. The Fe_2SiO_4 -spinel + stishovite assemblage in the FeO-SiO_2 -rich melts is consistent with crystallization of the melt vein matrix at the pressure up to 18 GPa. The crystallization pressure of ~18 GPa is much lower than the 45–90 GPa pressure one would conclude from the S6 shock effects in melt veins (Stöffler et al. 1991) and somewhat less than the 25–30 GPa inferred from S5 shock effects (Schmitt 2000) found in the bulk rock.

INTRODUCTION

The Umbarger chondrite was found in 1954, 29 km west-southwest of Umbarger, Randall County, Texas. The single stone of 13 kg is highly oxidized with a remnant fusion crust (Dod et al. 1981). Umbarger was previously examined and classified as S4 chondrite (Stöffler et al. 1991). Deep blue ringwoodite clasts found in the shock-induced melt vein (Xie and Sharp 2000) and subsequently akimotoite and Fe_2SiO_4 -spinel found in the melt vein (Xie et al. 2001, 2002) suggest a shock stage of S6. The goal of this paper is to determine the shock conditions that produced the observed high-pressure minerals.

Shock metamorphism has been studied since the 1960s to understand the shock conditions and effects on terrestrial rocks and meteorites. Early work on shock metamorphism focused on high-pressure minerals in terrestrial rock, such as stishovite and coesite (Chao 1967), and on shock deformation features of major minerals in meteorites, such as olivine, pyroxene, plagioclase, and metal sulfides (Fredricksson et al. 1963; DeCarli and Milton 1965; van Schmus and Wood 1967;

Heymann 1967; Carter et al. 1968; van Schmus and Ribbe 1968; Taylor and Heymann 1969; Stöffler 1972, 1974; Dodd and Jarosewich 1979; Sears and Dodd 1988). Shock pressures have been calibrated by comparing shock-metamorphic effects in naturally shocked rocks to shock effects in experimentally shocked samples from known pressures (Kieffer et al. 1975; Stöffler 1972; Schmitt 2000).

Stöffler et al. (1991) developed a shock classification scheme and pressure calibration for ordinary chondrites based on the comparison of deformation and transformation effects between natural and experimentally shocked samples. Shock effects in olivine and plagioclase, as recognized by optical petrography, are used to define six stages of shock (S1 to S6). The shock pressure calibration defines the shock stage transitions S1/S2, S2/S3, S3/S4, S4/S5, and S5/S6 to be <5, 5–10, 15–20, 30–35, and 45–55 GPa, respectively, and the onset of whole rock melting to be 75–90 GPa. The phase transformation of olivine to ringwoodite in or adjacent to veins and pockets is an easily recognized indicator of shock stage S6 that is inferred to occur at pressures greater than 45–55 GPa (Stöffler et al. 1991). This shock classification system

is easy to apply and correctly represents the progressive shock pressure sequence from weak to strong. However, pressure calibration based on shock recovery experiments is problematic for some features, such as phase transformations that depend on kinetics. For example, ringwoodite, the high-pressure polymorph of olivine, is stable between 18 to 22 GPa in static high-pressure experiments (Katsura and Ito 1989; Ito and Takhashi 1989; Irifune 1993; Fei and Bertka 1999), but it has never been recovered from a shock experiment. Despite this, the presence of ringwoodite in shocked chondrites is used as one of the indicators of shock stage S6 (Stöffler et al. 1991), and is commonly used as evidence of local pressure excursions in excess of 45 GPa (Spray 1999). The transformation of olivine to ringwoodite during shock is both time and temperature dependent. Therefore, the duration of a shock experiment may not be sufficient for observable transformation.

Recent studies of shock-induced melt veins show that melt vein mineralogy is able to provide constraints on crystallization conditions and shock pressures, given that crystallization occurs before pressure release (Chen et al. 1996; Sharp et al. 1997, 2001; Langenhorst and Poirier 2000b; Xie et al. 2001). Close similarities in mineralogy, grain size, composition, and microstructures between the samples from static high-pressure melting experiments and the assemblages that crystallize from melt veins in highly shocked meteorites suggest that phase diagrams obtained from static high-pressure experiments can be used to constrain the conditions of melt vein crystallization (Chen et al. 1996; Sharp et al. 2000). Crystallization of the silicate melt during rapid quenching of a shock vein is likely to occur from a supercooled liquid (Langenhorst and Poirier 2000b), which can result in metastable crystallization of subliquidus and even subsolidus phases. Although supercooling and metastable crystallization can result in disequilibrium assemblages, the high-pressure minerals that crystallize, combined with their pressure stabilities, still provide constraints on the pressure of crystallization. Our approach is not to assume chemical equilibrium and determine exact crystallization pressure and temperature from phase equilibrium data, but rather to constrain the crystallization pressure from the overall pressure stabilities of the minerals present. These constraints are likely to be tighter than those provided by shock-induced solid-state phase transformations, such as olivine to ringwoodite or enstatite to majorite, because diffusion, nucleation, and growth are much faster in an ultramafic melt than in solids. Melt vein assemblages have the added advantage of recording the crystallization history of the melt vein as it cools from the melt vein edge to the center (Langenhorst and Poirier 2000b; Sharp et al. 2003). If the melt vein crystallization occurs during decompression, a crystallization sequence from high-pressure to low-pressure phases will be recorded in the melt vein from its margin to its center.

We examined shock-induced melt veins in Umbarger to see how the mineralogy of the veins is related to the shock stage and precursor mineralogy. The main goal is to constrain crystallization pressure, based on mineral assemblages in the melt veins, and compare this pressure range to the calibrated shock pressure range of the shock classification of Stöffler et al. (1991) and the modified calibration of Schmitt (2000). A second goal is to understand how melt vein mineralogy varies in the sample and to relate these variations to changes in crystallization conditions or melt composition.

In a previous publication (Xie et al. 2002), we reported the first occurrence of natural Fe_2SiO_4 -spinel along with stishovite, SiO_2 -rich glass, and disordered Fe-rich phyllosilicates in local regions of melt veins. Although the phyllosilicate is a secondary feature believed to represent chemical weathering of the sample, the Fe_2SiO_4 -spinel and stishovite crystallized from a FeO- SiO_2 -rich silicate melt at high pressure. At that time, we proposed that the FeO- SiO_2 -rich melt is either a product of fractional crystallization or shock melting of a FeO- SiO_2 -rich precursor. In the present paper, we present further evidence for shock melting of a FeO- SiO_2 -rich precursor.

EXPERIMENTAL METHODS

A 145.2 g piece of the Umbarger L6 chondrite was obtained from the Center for Meteorite Studies at Arizona State University (ASU sample #1217) and a 30 μm thin section was made using a thermal adhesive (Crystalbond) so that portions could be removed for transmission electron microscope (TEM) analysis. The sample was investigated with a combination of optical petrography using transmitted and reflected light, scanning electron microscopy (SEM) with thermal and field emission electron sources, electron microprobe analysis (EMPA), and analytical transmission electron microscopy (ATEM), also with thermal and field emission electron sources.

Petrography and SEM were used to locate melt veins, to study the shock features in the host rock, to identify large polycrystalline ringwoodite grains, and to characterize alteration zones. The textures of the melt veins and semi-quantitative analysis of minerals in the melt veins were investigated using a JEOL JSM-840 SEM and Hitachi S-4700 FESEM, both equipped with energy dispersive spectroscopy (EDS). Chemical analyses of melt veins and alteration veins were obtained using a JEOL-8600 electron microprobe (EMP) operated at 15 kV. Focused beam (2 μm) and broad beam (10–100 μm) analyses were used to measure the compositions of the single phases and bulk compositions of the mineral assemblages, respectively. We used San Carlos olivine for Mg and Si standard, anorthite for Al, wollastonite for Ca, and fayalite for Fe.

Since the grain sizes of the minerals in the shock melt veins range from hundreds of nanometers to several

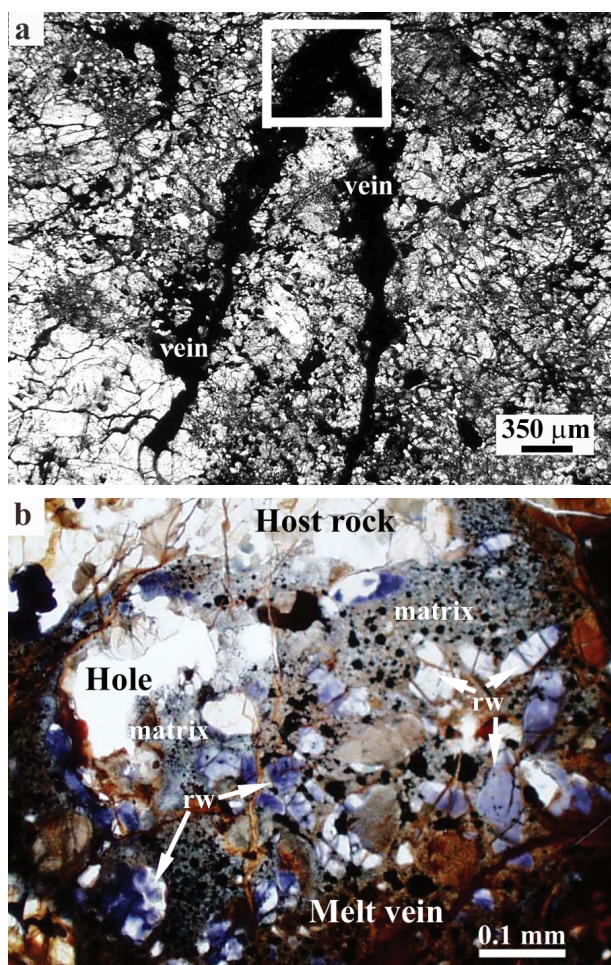


Fig. 1. a) Transmitted light photomicrograph of black melt veins and fractured host rock in Umbarger. The rectangular box represents the region studied by TEM; b) transmitted light photomicrograph of a thinned TEM sample. Abundant blue ringwoodite (rw) clasts are seen in the sample thinned to less than 10 μm . The black dots dispersed throughout the melt vein are metal troilite blebs.

micrometers, ATEM is an essential tool for their characterization. A sample disc 3 mm in diameter and containing a bifurcated melt vein (Fig. 1) was cored from the thin section and glued onto a copper TEM grid with a single 1 mm hole. The disc center was mechanically thinned to a thickness of $\sim 10\ \mu\text{m}$ using a Gatan dimple grinder and then ion milled using a Gatan precision ion polishing system (PIPS). The specimen was thinned to electron transparency using Ar ions accelerated to 5 keV at an angle of $4\text{--}6^\circ$ from the sample surface. ATEM data were recorded from the thin edges of ion-milled holes using JEOL JEM-2000FX and Philips CM200-FEG microscopes operated at 200 kV. Both of these instruments are equipped with KEVEX energy dispersive X-ray spectrometers and EmiSpec analytical systems for chemical analysis. The EDS detectors have ultra thin windows and can detect elements heavier than boron. However, elements lighter than Na are subject to strong absorption.

Conventional TEM imaging techniques were employed to characterize the textures of the veins and the microstructures of the minerals. Mineral phases were identified on the basis of quantitative EDS and selected area electron diffraction (SAED).

EDS analyses were quantified using the ratio technique of Cliff and Lorimer (1975). The K factors (relative to Si) for Mg, Fe, Ca, Al, and O were determined from standards using the parameterless correction method of van Cappellen (1990). The standards included fayalite for Fe and O; San Carlos olivine for Mg, Fe, and O; Johnstown orthopyroxene for Mg, Fe, and O; and Hakone anorthite for Al, Ca, and O. Although oxygen data was collected and reported (Table 2), oxygen data was not used to correct for absorption effects. Calculations of mineral formulas were based on the number of oxygen anions in the formula and assumed oxidation states of the cations.

RESULTS

Petrography

Umbarger consists of an assemblage of olivine ($\text{Fo}_{74}\text{Fa}_{26}$), orthopyroxene ($\text{En}_{77}\text{Wo}_2\text{Fs}_{21}$) (Table 1), and plagioclase, with minor amounts of metal (kamacite and taenite) and troilite. The chondrules are poorly defined. Coarse-grained olivine and pyroxene crystals have sizes up to hundreds of micrometers, while fine-grained olivine and pyroxene occur up to several micrometers in size. Both coarse-grained and fine-grained olivines and pyroxenes have abundant shock-induced irregular and planar fractures (Fig. 1a). Most grains also show pervasive mosaicism as indicated by a pronounced irregular optical extinction. A few grains show sharp extinction and few fractures, indicating re-crystallization or post-shock annealing (Rubin 2002). Most plagioclase grains are completely converted to maskelynite, a widely accepted indicator of high-pressure shock (Milton and DeCarli 1963; Binns 1967; Stöffler et al. 1991). Plagioclase grains near the melt vein show typical flow features, suggesting that the grains were molten during shock. Vein networks are pervasive throughout the thin section (Fig. 1a). Most of these are Fe-oxide-rich veins that probably formed by oxidation of metal sulfide veins during weathering, but some are shock-induced silicate melt veins.

Petrographic classification by Dod et al. (1981) implied that Umbarger has both type 3 and type 6 characteristics. The heterogeneity of the olivine compositions (Fa_{22} to Fa_{33}) was used to argue for type 3, yet abundant plagioclase and partial re-crystallization of the matrix suggest type 6 (Dod et al. 1981). Our electron microprobe data show that the host rock olivine compositions range from Fa_{24} to Fa_{28} with an average composition of Fa_{26} (Table 1) and a standard deviation of 1.27. This compositional range, along with poorly defined chondrules, partial re-crystallization of the matrix and coarse plagioclase, is consistent with type 6 (van Schmus and Wood

Table 1. Microprobe analyses of host rock, Fe oxide veins and FeO-SiO₂ veins.^a

	Ol (12)	Opx (2)	Fe-O vein (3)	Fe-Si-O vein (12)
Weight%				
Mg	22.66	16.61	0.19	0.55
Si	17.75	25.58	0.80	13.26
Ca	0.02	0.76	—	—
Fe	18.03	10.61	48.71	31.92
O	40.31	43.42	14.98	24.62
Total	98.77	96.97	64.68	70.36
Formula unit				
Oxygens	4	3	—	—
Mg	1.48	0.76	—	—
Si	1.00	1.01	—	—
Ca	0	0.02	—	—
Fe	0.51	0.21	—	—
Fe/(Fe + Mg)	25.74	—	—	—
Cation	3.00	1.99	—	—

^aThe numbers of microprobe analyses are in parentheses; O wt% is calculated by stoichiometry; Ol = olivine, Opx = orthopyroxene, Fe-O vein = Fe oxide vein, Fe-Si-O vein = FeO-SiO₂-rich vein.

Table 2. TEM EDS analyses of high-pressure phases in melt vein.^a

	P-rw (3)	M-rw (4)	Ak (11)	Ca-cpx (10)
Atom%				
O	57.14	55.83	58.53	59.05
Na	—	—	—	1.21
Mg	20.74	20.30	18.16	7.45
Al	0.00	0.29	1.79	1.73
Si	14.59	14.68	19.20	20.32
Ca	—	—	0.12	5.91
Cr	—	—	0.03	—
Fe	7.53	8.90	2.19	4.34
Total	100	100	100	100
Formula unit				
Oxygens	4	4	3	3
Na	—	—	—	0.06
Mg	1.45	1.39	0.88	0.36
Al	0.00	0.02	0.09	0.08
Si	1.02	1.00	0.94	0.99
Ca	—	—	0.01	0.29
Cr	—	—	0.00	—
Fe	0.52	0.59	0.11	0.21
Fe/(Fe + Mg)	0.27	0.30	0.11	—

^aThe number of EDS spectra are in parentheses; P-rw = polycrystalline ringwoodite; M-rw = matrix ringwoodite; Ak = akimotoite; Ca-cpx = Ca-rich clinopyroxene. The mineral formulas were calculated based on the number of oxygen anions in the formula and assumed oxidation states of the cations.

1967), but inconsistent with the Dod et al. (1981) type 3 classification. The large spread in Fa contents of the olivines measured by Dod et al. (1981) may be the result of shock and weathering, which can redistribute iron in the form of tiny sulfide droplets or oxide veins.

The heavily shocked Umbarger chondrite contains black shock veins, ranging from 0.1 mm to 0.5 mm in width (Fig. 1a). The shock melt feature described in this study has a highly irregular shape that could be described as either an irregular melt pocket or as two interconnecting melt veins. For simplicity, we will refer to this feature as a melt vein, but we have not inferred a specific formation mechanism for the feature described here. The black color of the shock melt is a result of fine-grained round metal and troilite blebs or metal troilite intergrowths up to greater than 10 µm in size, but commonly less than 5 µm in size throughout a fine-grained melt vein matrix (Fig. 1b). These metal sulfide blebs represent droplets of an immiscible metal sulfide melt that coexisted with silicate melt in the shock vein before quench. There are two distinct high-pressure silicate parageneses in the melt vein, corresponding to melt crystallization and solid-state transformation.

The first paragenesis consists of fine-grained crystals in the vein matrix coexisting with metal troilite droplets (Fig. 1b). This paragenesis results from crystallization of the chondritic silicate and immiscible metal-sulfide melts that make up the shock veins (Chen et al. 1996; Sharp et al. 1997; Langenhorst and Poirier 2000b). These veins represent localized melting during shock (Fredrickson et al. 1963), possibly by shear heating or pore collapse during shock loading. We use the silicate mineral assemblages of the vein matrix to infer crystallization conditions for the silicate melt during its quench (Chen et al. 1996; Xie and Sharp 2000; Sharp et al. 2001).

The second paragenesis consists of sub-rounded and unfractured host rock clasts entrained in the melt vein. Most of these are polycrystalline aggregates of ringwoodite, which are too small to be easily seen in a standard thin section. Only by using intense convergent light were we able to see the blue-purple color indicative of ringwoodite inclusions in the melt vein. Further thinning of the sample for TEM revealed many ringwoodite inclusions (Fig. 1b), which are generally smaller than the thickness of the thin section. Only the largest grains are large enough to be observed in standard 30 µm thick section. These rounded ringwoodite clasts have the same compositional range as the olivines in the host chondrite and occur only within or in contact with melt veins. We interpret these ringwoodite clasts as having formed by a solid-state transformation from olivine clasts that were entrained in the melt.

Interpretation of shock stage is complicated by heterogeneity in shock effects. Stöffler et al. (1991) assigned Umbarger a shock stage of S4. We observed olivine with strong mosaicism and abundant maskelynite in regions not adjacent to the melt veins, indicating a shock stage of S5. In and adjacent to melt veins, we find glassy plagioclase with flow textures and polycrystalline ringwoodite. Both of these features are characteristic of shock stage S6. Based on the shock classification scheme of Stöffler et al. (1991), one would infer that the melt veins experienced S6 conditions,

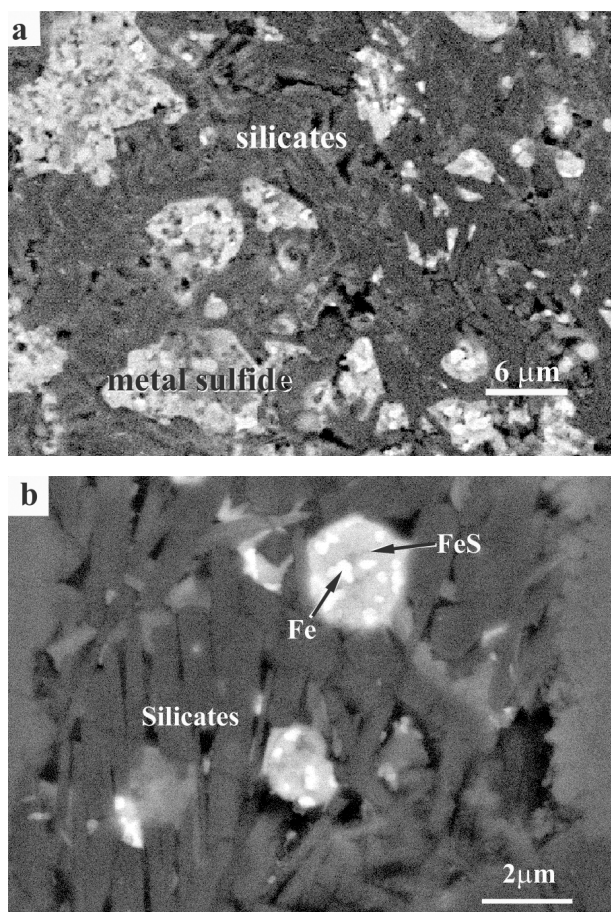


Fig. 2. a) FESEM image of the melt vein matrix showing elongate silicate crystals intergrown with metal sulfide droplets. Bright features in melt vein are metal troilite spheroids crystallized from the droplets of immiscible metal-sulfide melt; b) high magnification image of the vein matrix texture showing elongate silicate crystals and metal-sulfide droplets. The metal sulfide droplets consist of Fe-Ni metal (white) and Fe-sulfide (light gray).

whereas the chondrite host away from the melt vein experienced S5 conditions.

SEM Observations

Three distinct types of veins were characterized by SEM: silicate melt veins, FeO-SiO₂-rich zones or veins, and Fe oxide veins.

Backscatter electron (BSE) images (Figs. 2a and 2b) show quench textures typical of the melt vein in Umbarger, which consists of silicate crystals surrounding metal sulfide droplets. The silicate matrix consists of elongate minerals intergrown with more equant and irregular minerals (Fig. 2b). As discussed below, TEM confirms that these minerals are akimotoite and ringwoodite, respectively. Metal sulfide aggregate droplets show a clear eutectic texture, indicating crystallization of a Fe-Ni-S eutectic immiscible melt.

Some parts of the melt veins are rich in FeO and SiO₂ (Figs. 3a and 3b). The FeO-SiO₂-rich material associated with

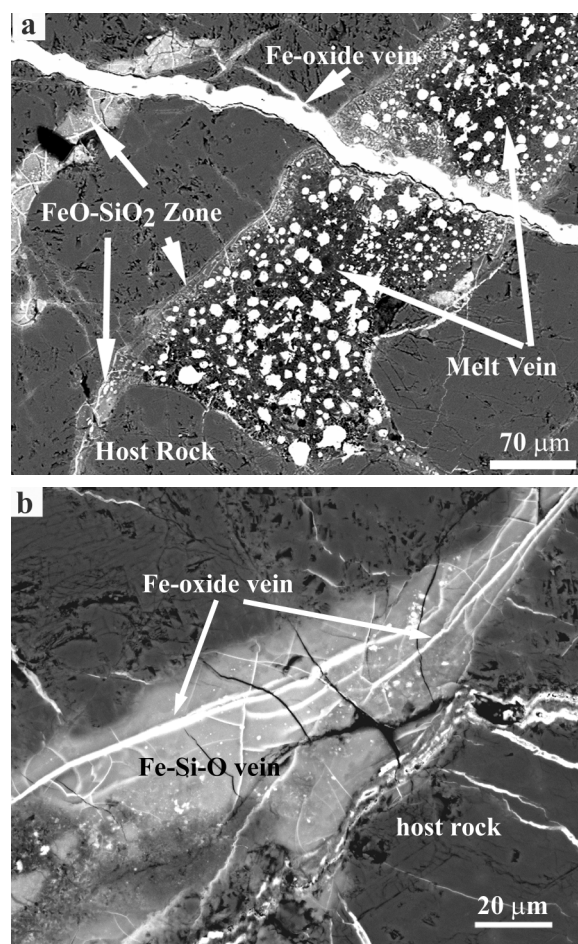


Fig. 3. a) BSE image of FeO-SiO₂-rich zones at the edge of a melt vein and as a separate vein near a melt vein. Note that the Fe-rich materials at the edge of the melt vein are connected to a narrow vein of FeO-SiO₂-rich material (left corner), which extends into Fig. 3b; b) BSE image showing a diffuse boundary between a FeO-SiO₂-rich vein (light gray) and the surrounding host rock (dark gray). Fe oxide veins (bright white) occur within the FeO-SiO₂-rich alteration vein.

melt veins commonly forms the edges of the melt veins (Fig. 3a), indicating that the shock-induced melt was compositionally heterogeneous and not a well-mixed chondritic melt as commonly described (Stöffler et al. 1991; Chen et al. 1996). FeO-SiO₂-rich zones were also found to extend beyond melt veins as FeO-SiO₂-rich alteration veins within the chondrite matrix (Figs. 3a and 3b). The alteration vein shown in Fig. 3b is a continuation of the FeO-SiO₂-rich zone that extends off the edge of the melt vein in Fig. 3a. Electron microprobe analyses of the FeO-SiO₂-rich zones yield low totals (~70%) (Table 1), suggesting that the material is probably porous or H₂O-rich. These FeO-SiO₂-rich veins have diffuse boundaries with the surrounding MgO-rich host chondrite (Fig. 3b), which suggests diffusive Fe-Mg exchange with the surrounding chondrite, and therefore, high temperatures during or after the alteration event. However, the diffuse interface could also reflect dispersed Fe-rich minerals in a region altered at low temperature.

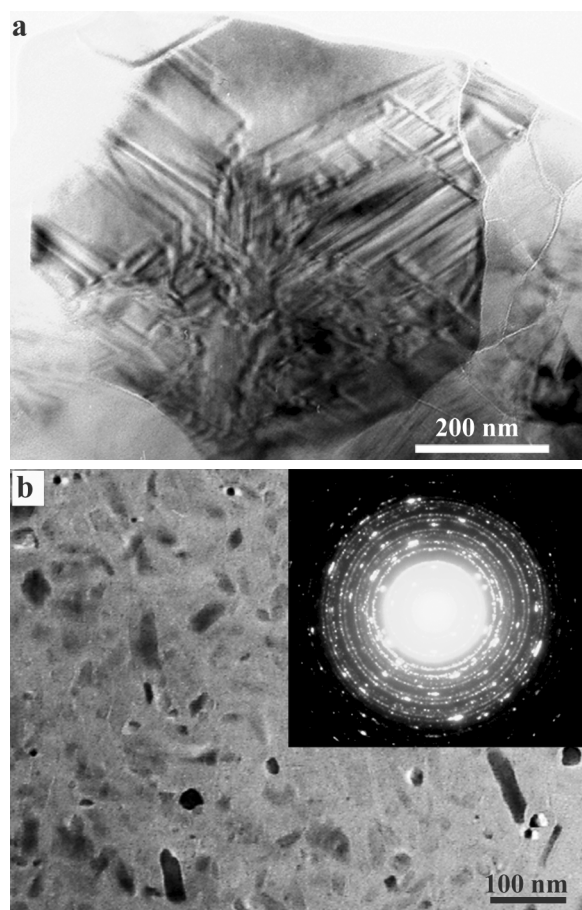


Fig. 4. a) Bright field TEM image of a polycrystalline ringwoodite aggregate showing the characteristic $\{110\}$ stacking faults in ringwoodite; b) bright field TEM image of a hollandite-structured polycrystalline aggregate after plagioclase, showing crystals ranging from 20 to 60 nm. The inserted SAED ring pattern is indexed as the hollandite structure.

Fe-oxide veins are also found throughout Umbarger (Figs. 3a and 3b). These oxide veins cross cut both shock-induced melt and the FeO-SiO₂-rich zones, and therefore, postdate both the shock metamorphism and the FeO-SiO₂ alteration event. These late-stage oxide veins have distinct sharp boundaries with the surrounding host rock, and therefore have not produced significant Fe-Mg exchange with the host, probably representing a low-temperature alteration event, such as weathering on Earth.

TEM Observations of Polycrystalline Aggregates

The ringwoodite grains that occur in the melt veins are polycrystalline aggregates that consist of tiny ringwoodite crystals ranging from 200 to 1000 nm in size (Fig. 4a). The characteristic $\{110\}$ stacking faults are abundant in these crystals (Fig. 4a), resulting in streaking along $\{110\}^*$ directions in SAED patterns, similar to those seen in ringwoodites from Tenham and Sixiangkou chondrites (Price

1979; Madon and Poirier 1983; Langenhorst et al. 1995; Chen et al. 1996). EDS analyses from ATEM reveal a stoichiometric olivine composition of approximately Fa₂₇, which is within the compositional range of the unaltered olivine away from the melt veins (Tables 1 and 2).

The melt veins also contain polycrystalline aluminosilicate clasts that have the hollandite structure. TEM imaging shows that these grains are not amorphous maskelynite or normal glass, but rather polycrystalline aggregates of ~20 nm nanocrystals (Fig. 4b). SAED ring patterns are indexed as the hollandite structure (Fig. 4b). ATEM EDS analyses of this material give a composition of Na_{0.43}K_{0.02}Ca_{0.25}Fe_{0.02}Al_{1.27}Si_{2.80}O₈. This composition is highly depleted in alkalis, presumably as a result of using a high current density in the TEM EDS analyses. It is likely that this highly unstable hollandite structure is more sensitive to alkali loss than plagioclase. Based on the relative abundance of Al and Si in the analyses, this composition corresponds to oligoclase (An₂₅). Plagioclase that has been transformed to hollandite has been reported in Sixiangkou (Gillet et al. 2000) and Tenham (Tomioaka et al. 2000; Sharp et al. 2000) L6-S6 chondrites and in the martian meteorite Zagami (Langenhorst and Poirier 2000a, 2000b). The hollandite may form by either a solid-state transformation or by crystallization from molten plagioclase (Sharp et al. 2000). In Umbarger, the polycrystalline texture of hollandite is consistent with both solid-state and crystallization mechanisms, but flow textures suggest crystallization from molten plagioclase during shock.

TEM Observations of Matrix Minerals

The principal mineral phases in the quenched silicate liquid of the melt vein matrix are akimotoite, ringwoodite, and Ca-rich clinopyroxene. The SEM imaging (Fig. 5a) and TEM imaging (Fig. 5b) show that akimotoite is the most abundant silicate in the melt vein, with ringwoodite as the second most abundant. However, the proportion of ringwoodite to akimotoite varies within the melt vein, suggesting heterogeneity of the bulk composition of the melt within the vein.

Akimotoite, a high-pressure polymorph of enstatite with the ilmenite structure, appears as idiomorphic elongate crystals up to 4 μm in length (Figs. 5a and 5b). Since we have not observed prismatic grains in end-on orientations, we have concluded that the akimotoite grains are platelets. Electron diffraction patterns from these crystals are only consistent with the structure of the akimotoite (Fig. 5c), and could not be indexed as any of the other enstatite polymorphs (orthoenstatite, clinoenstatite, majorite, and MgSiO₃-perovskite). Multiple diffraction patterns from single crystals were all indexed as akimotoite (space group of $R\bar{3}$) using d-spacings, interplanar angles, symmetry, and systematic absences for dynamical diffraction. Calculated cell parameters ($a = 0.48$ nm, $c = 1.38$ nm, $c/a = 2.86$, assuming a hexagonal

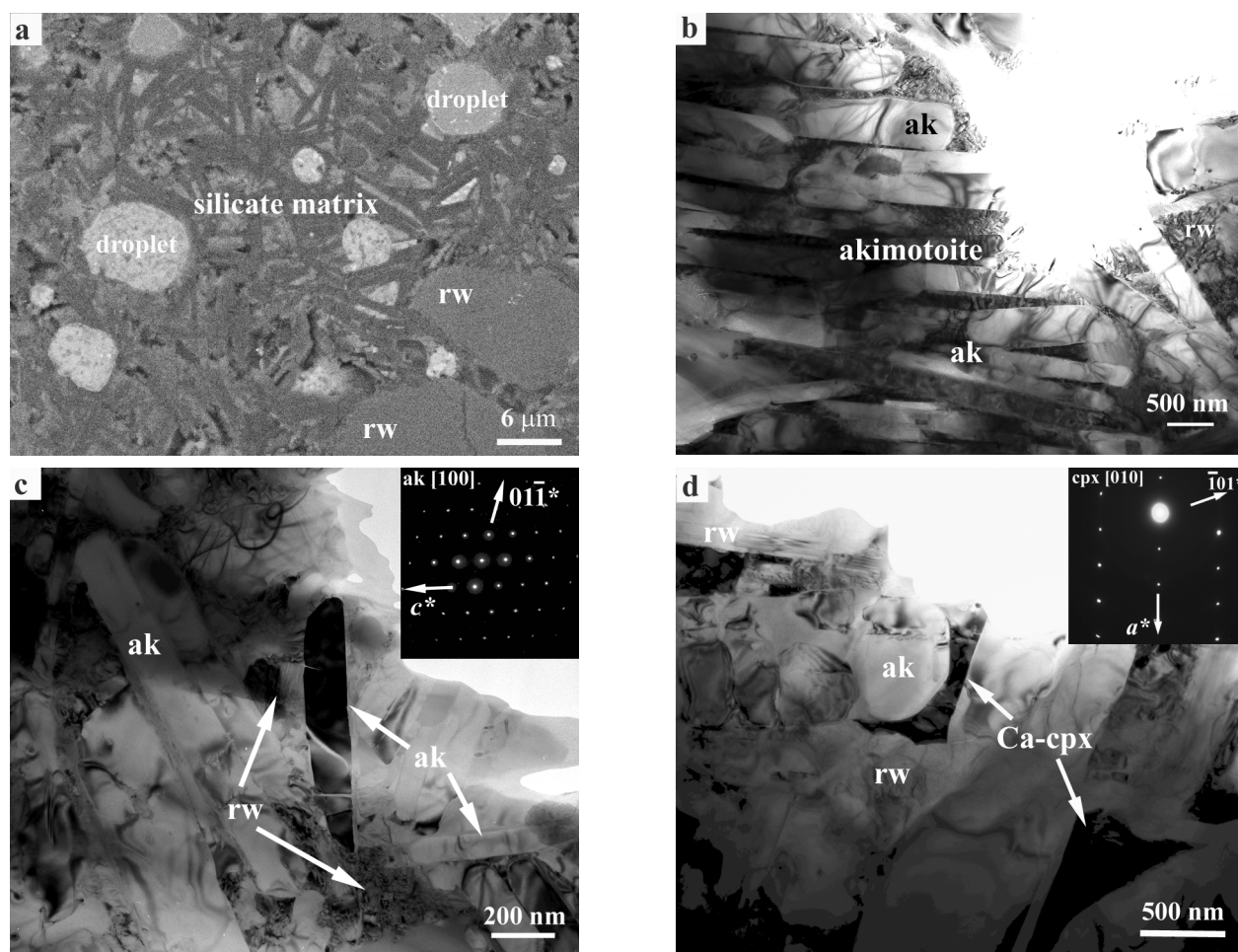


Fig. 5. a) FESEM image of the vein matrix texture showing elongate crystals intergrown with metal sulfide droplets. Polycrystalline ringwoodite (rw) grains are seen in the bottom right; b) bright field TEM image showing abundant elongate akimotoites with interstitial ringwoodites in the melt vein of Umbarger; c) bright field TEM image of intergrown akimotoite (ak) and ringwoodite (rw). The inset SAED pattern is the [100] zone axis pattern of the dark akimotoite in image center; d) bright field TEM image showing anhedral Ca-clinopyroxene (Ca-cpx) between akimotoite and ringwoodite. The inset SAED pattern is the (010) zone axis of the dark Ca-clinopyroxene in the image center.

structure) agree with those for synthetic akimotoite ($a = 0.473$ nm, $c = 1.356$ nm, $c/a = 2.867$, Horiuchi et al. [1982]) and those of akimotoite in Tenham chondrite ($a = 0.478 \pm 0.005$, $c = 1.36 \pm 0.01$ nm, $c/a = 2.85$, Tomioka and Fujino [1997]) within SAED accuracy. EDS analyses of akimotoite (see Table 2) give a formula of $(\text{Ca}_{0.01}\text{Mg}_{0.88}\text{Fe}_{0.11}\text{Al}_{0.03})_{1.03}(\text{Si}_{0.94}\text{Al}_{0.06})_{1.0}\text{O}_3$ with $\text{Fe}/(\text{Fe} + \text{Mg}) = 0.11$. The $\text{Fe}/(\text{Fe} + \text{Mg})$ ratio is slightly higher than the 10 at% upper limit of FeSiO_3 solubility determined experimentally (Ito and Yamada 1982). Compared with the host orthopyroxene, akimotoite is rich in Al_2O_3 (more than 9 at%), indicating that it did not form by a solid-state transformation from orthopyroxene, but rather crystallized from the chondritic melt, as did the akimotoite in Acfer 040 (Sharp et al. 1997).

The ringwoodite in the melt-vein matrix occurs interstitially between idiomorphic akimotoite platelets (Figs. 5a–5d). These ringwoodites can be equant or they can

be somewhat elongated with sizes ranging from about 200 nm to 1000 nm. They show a high degree of stacking disorder, indicated by high densities of stacking faults on $\{110\}$ planes that commonly occur as short (100–200 nm) segments. The average composition of melt vein matrix ringwoodites (Fa_{30}) is somewhat more fayalitic than the average composition of polycrystalline ringwoodite (Fa_{27}) and the host rock olivine (Fa_{26}) (Tables 1 and 2), which is consistent with the textural evidence for the melt vein matrix ringwoodite crystallizing from the melt rather than being a product of solid-state transformation from olivine.

The Ca-rich clinopyroxene occurs as small interstitial grains between idiomorphic akimotoite grains and anhedral ringwoodite grains (Fig. 5d). SAED patterns from these materials confirmed the structure of clinopyroxene with the space group C2/c (Fig. 5d). EDS analyses give a Ca-clinopyroxene composition of $(\text{Ca}_{0.58}\text{Na}_{0.12}\text{Mg}_{0.72}\text{Fe}_{0.42}\text{Al}_{0.15})_{1.99}(\text{Si}_{1.98}\text{Al}_{0.02})_{2.0}\text{O}_6$. Intergrowth textures suggest that

akimotoite crystallized first, followed next by ringwoodite, and then finally by clinopyroxene.

DISCUSSION

Origin of FeO-SiO₂-Rich Silicate Melt in Melt Veins

The presence of Fe₂SiO₄-rich spinel and stishovite crystals in a matrix of silica-rich glass in some regions of the melt vein in Umbarger (Xie et al. 2002) demonstrate that the melt vein contained both chondritic silicate melt and an FeO-SiO₂-rich melt when it crystallized at high pressure. In our previous paper (Xie et al. 2002), we discussed two possible origins: 1) the FeO-SiO₂-rich melt formed by fractional crystallization of MgO-rich silicates at high pressure, and 2) the FeO-SiO₂-rich melt formed by the shock melting of preexisting FeO-SiO₂-rich material in Umbarger.

The first possibility is ruled out by our current observation of FeO-SiO₂-rich alteration veins that have been melted and incorporated into shock melt veins. The similarity in composition between FeO-SiO₂-rich alteration zones outside the shock melted region and the FeO-SiO₂-rich shock melt is consistent with a common origin. The continuation of FeO-SiO₂-rich alteration veins from the unmelted matrix (Fig. 3b) into melt veins (Fig. 3a) confirms that these are the same materials and that shock loading caused some of this material to melt and be incorporated into shock-induced melt veins. The alteration zones and veins are distinct from late-stage oxide veins that cross cut both melt veins and FeO-SiO₂-rich zones and have sharp boundaries with the surrounding host rock. These observations demonstrate the formation of FeO-SiO₂-rich melt by shock melting of preexisting FeO-SiO₂-rich material. This indicates that an alteration event, which caused the local FeO-SiO₂ metasomatism of the host chondrite, occurred before the impact event that caused the shock metamorphism.

Estimating Crystallization Pressure

The pressure of melt vein crystallization generally can be constrained by the vein matrix assemblage using appropriate phase-equilibrium data (Chen et al. 1996; Sharp et al. 2001). In doing this, we do not assume equilibrium crystallization to determine a precise pressure and temperature, but rather we use the overall pressure stabilities of the minerals and mineral assemblages to constrain the crystallization pressure. In the case of Umbarger, the assemblage that crystallized is not an equilibrium liquidus assemblage for a chondrite. Akimotoite, which occurs throughout the melt veins in Umbarger, is not a liquidus phase in high-pressure melting experiments using the Allende CV3 chondrite (Agee et al. 1995) or KLB-1 peridotite (Zhang and Herzberg 1994). In both systems, the stable liquidus phase that occurs with ringwoodite is majorite garnet (Agee et al. 1995; Zhang and Herzberg 1994). In the

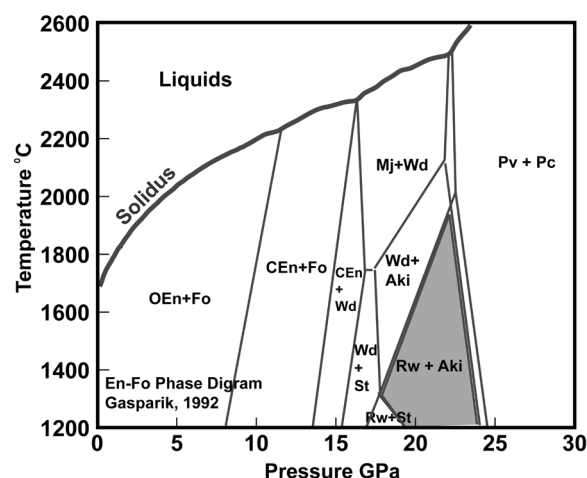


Fig. 6. Temperature-pressure phase diagram for the enstatite-forsterite join, after Gasparik (1992). Ringwoodite + akimotoite is subsolidus at pressures ranging from 18 to 25 GPa. Rw = ringwoodite, Aki = akimotoite, Wd = wadsleyite, Mj = majorite, Pv = perovskite, Pc = periclase, St = stishovite, CEn = clinoenstatite, OEn = Orthoenstatite, Fo = forsterite.

enstatite-forsterite system, akimotoite plus ringwoodite is stable as a sub-solidus assemblage between 18–24 GPa and >500° below the solidus (Fig. 6) (Gasparik 1992). The conditions under which akimotoite plus ringwoodite is stable in an L chondrite composition is unknown and there are insufficient subsolidus data for both Allende (Agee et al. 1995) and KLB-1 peridotite (Zhang and Herzberg 1994) compositions to constrain the akimotoite stability in these systems. By analogy to the enstatite-forsterite system, akimotoite is likely to be a subsolidus phase in an L chondrite, and therefore crystallized metastably from a supercooled chondritic melt in Umbarger. Assuming that metastable crystallization of akimotoite plus ringwoodite occurs under pressure conditions similar to the pressure stability of akimotoite in the enstatite-forsterite system, we infer that the chondritic melt in Umbarger crystallized at approximately 18–24 GPa. The presence of Ca-clinopyroxene is similarly problematic because, like akimotoite, Ca-clinopyroxene is metastable relative to majorite at high pressures and liquidus temperatures (Oguri 1997). Although Ca-pyroxene is not generally considered as a high-pressure mineral, Ca-rich clinopyroxene is stable up to about 18 GPa (Oguri 1997). The crystallization of two subsolidus phases, akimotoite and Ca-clinopyroxene, is consistent with metastable crystallization from a supercooled chondritic liquid where nucleation of majorite garnet was inhibited.

Interpreting the crystallization pressure for the Fe₂SiO₄-spinel-stishovite assemblage is complicated by the lack of high-pressure melting data in the FeO-SiO₂-system. In the Fe₂SiO₄ system (Fig. 7), Stishovite is the liquidus phase above 14 GPa whereas Fe₂SiO₄-spinel is the solidus phase from 14 to about 18 GPa (Otahni 1979). One possible

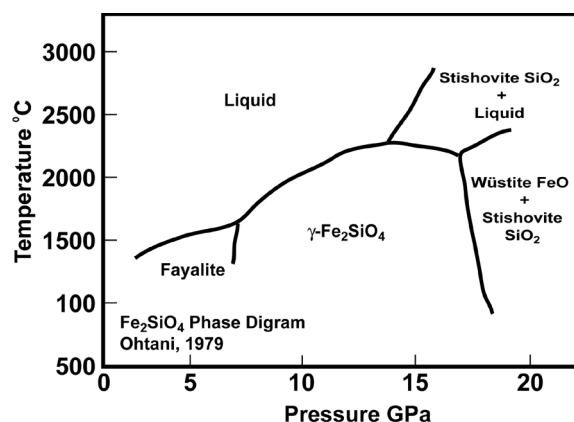


Fig. 7. Temperature-pressure phase diagram for the Fe_2SiO_4 system, after Ohtani (1979).

mechanism for producing spinel plus stishovite plus glass in Fe_2SiO_4 would be quench from super-liquidus conditions between about 14 and 17 GPa with stishovite crystallizing first, followed by Fe_2SiO_4 -spinel. One problem with trying to apply this data to Umbarger is that the FeO-SiO₂-rich zones in Umbarger are more SiO₂-rich than Fe_2SiO_4 . In addition, the Fe_2SiO_4 -spinel has a significant Mg_2SiO_4 component, which increases its pressure stability above 17 GPa. Therefore, the Fe_2SiO_4 -rich spinel + stishovite + glass assemblage that occurs in the FeO-SiO₂-rich zones of the melt veins can represent a quench at somewhat higher pressures than the 14–17 GPa range.

There are two interpretations of the observed mineral assemblages and the inferred crystallization sequence. First, crystallization occurred during pressure release, starting with akimotoite and ringwoodite at a pressure between 18–24 GPa and ending with crystallization of Ca-rich clinopyroxene and Fe_2SiO_4 -rich spinel at or below ~18 GPa. This is consistent with the textural evidence for pyroxene crystallizing after akimotoite and ringwoodite, but not with the sequence of minerals seen across the melt vein. An alternative interpretation is that all high-pressure phases crystallized, nearly isobarically, at an equilibrium shock pressure representing the common pressure stability of all the crystallizing phases. These two interpretations can be tested by the distribution of crystallization assemblages in the melt vein. Since melt veins represent local high-temperature anomalies, they quench primarily by conduction to the surrounding host chondrite (Langenhorst and Poirier 2000b; DeCarli et al. 2002; Sharp et al. 2003). Crystallization begins at the melt vein margins and progresses inward as heat is conducted outward. If a melt vein crystallizes during pressure release and the pressure change extends over a range of mineral stabilities, the mineral assemblage should change from the melt vein edge to the center reflecting the decrease in pressure during crystallization. However, if crystallization occurs at constant equilibrium shock pressure, crystallization

should be nearly isobaric, resulting in a constant mineral assemblage from the vein edge to center.

FESEM and TEM observations show that the akimotoite plus ringwoodite assemblage occurs throughout the melt vein. If Ca-clinopyroxene in the melt vein were a result of late crystallization at lower pressure during pressure release, the abundance of clinopyroxene should increase toward the center of the melt vein. Similarly, if the pressure decreases below akimotoite and ringwoodite stability during quench, then these phases should not occur in the center of melt veins. However, the crystallization assemblage in the melt vein of Umbarger does not vary from edge to center. This observation indicates that the crystallization of the melt vein in Umbarger occurred over a limited pressure range, implying that either the melt vein quenched at the equilibrium shock pressure or that crystallization occurred during very slow pressure release. The metastable crystallization assemblages and the lack of high-pressure melting data for L-chondrites preclude tight constraints on the crystallization pressure. However, if the akimotoite plus ringwoodite plus Ca-pyroxene assemblage represent crystallization over a narrow range of pressure, that pressure would correspond to the overlap in stability of these phases, which is approximately 18 GPa. The Fe_2SiO_4 -rich spinel + stishovite assemblage has an upper stability limit of 17 to 18 GPa or somewhat higher. Based on the pressure stabilities of all the crystallizing phases, combined with the evidence against a large pressure decrease during crystallization, we infer that the melt vein in Umbarger crystallized at approximately 18 GPa.

Different Estimates of Shock Pressure

There is a large discrepancy between the 45–90 GPa pressure that one would infer for Umbarger from the S6 classification of Stöffler et al. (1991) and the apparent crystallization pressure of ~18 GPa from the melt vein mineralogy. Based on shock effects in a separate piece of Umbarger, Stöffler et al. (1991) assigned a shock stage of S4, corresponding to the equilibrium shock pressure of 15–35 GPa. Greater metamorphic effects in our sample indicate a shock stage of S5 (30–55 GPa) for shock features in the bulk rock or S6 (45–90 GPa) if we use the presence of ringwoodite and “normal” plagioclase glass associated with melt veins.

Variations in shock effects within a given sample have been interpreted to result from pressure and temperature heterogeneities that occur as a shock wave passes through a heterogeneous sample. For example, the presence of unfractured quartz, quartz melt, coesite, and stishovite in shocked Coconino sandstone from Meteor Crater was interpreted as resulting from pressure heterogeneities during shock (Chao 1968; Kieffer 1975). Similarly, heterogeneous shock effects associated with melt veins in chondrites, such as local melting, olivine recrystallization, “normal” plagioclase glass, and phase transition of olivine to ringwoodite have

been interpreted to represent local pressure-temperature excursions (Stöffler et al. 1991; Spray 1999; Kenkmann et al. 2000, 2003; Schmitt 2000). Although pressure heterogeneities must occur during the shock loading of a heterogeneous sample such as a meteorite, large pressure heterogeneities would quickly decay because rocks are not strong enough to support stresses more than a few kilobars (Gault et al. 1968).

The pressure heterogeneities that occur during shock loading of a polyphase meteorite rapidly equilibrate by shock wave reverberations. Assuming a shock wave velocity of ~ 10 km/s, reverberations between 100 μm grains would occur on a 10^{-8} sec (10 ns) time scale. After several reverberations over a time of less than ~ 50 ns, the shock pressure homogenizes to the equilibrium shock pressure. The duration of the shock pulse, which is determined by the size of the projectile and the distance from the sample to a free surface (Melosh 1989), can be much longer than the pressure equilibration time in natural shock events between large impacting bodies. For example, for a 1 km projectile and a shock wave velocity of 10 km/s, the shock pulse duration is greater than 0.1 sec for a sample near the impact site. The initial pressure heterogeneities are damped out on a time scale $\sim 10^{-6}$ times the pulse duration for such an impact. If heterogeneities in shock effects are a result of local pressure variations caused by the variable shock impedances of the mineral constituents, they must be created in the very short period ($<10^{-7}$ sec) before the shock pressure equilibrates.

For a natural long duration impact, heterogeneous shock effects, such as phase transitions of olivine to ringwoodite, are more likely the result of the thermal heterogeneities than the very short-lived pressure heterogeneities that occur during shock loading. The transformation of olivine to ringwoodite is only observed within or in contact with melt veins and pockets (Fredriksson et al. 1963; Stöffler et al. 1991; Aramovich et al. 2003). The fine polycrystalline microstructure of the ringwoodite implies homogeneous nucleation of ringwoodite followed by interface-controlled growth. The rate of interface controlled growth is very slow at the low temperatures ($\sim 600^\circ\text{C}$) corresponding to the interiors of subducting slabs (Rubie and Ross 1994; Kerschhofer et al. 1996, 2000), but will be much faster at near solidus temperatures (~ 1900 – 2100 at 20 GPa) at the margin of a melt vein. It is commonly assumed that the duration of the shock pulse is too short for a nucleation and growth mechanism. This is clearly the case in shock recovery experiments in which ringwoodite has never been produced (Jeanloz 1980; Langenhorst et al. 2002). Natural impacts apparently provide sufficient time at pressure for these solid-state transitions to occur in locally heated regions (i.e., shock veins).

The use of ringwoodite in melt veins to infer an S6 shock stage and pressure of >45 – 55 GPa probably leads to an overestimate of the shock pressure. Ringwoodite is stable between about 18–23 GPa in static high-pressure experiments

(Katsura and Ito 1989; Ito and Takhashi 1989; Irifune 1993; Fei and Bertka 1999), but it is inferred to require >45 – 55 GPa for its formation during shock (Stöffler et al. 1991; Spray 1999). However, there is no experimental evidence to support this interpretation.

If we assume that the ringwoodite in Umbarger was formed at the shock pressure for the bulk rock rather than from a very short duration (<100 ns) pressure excursion during shock loading, the disparity in shock pressures between melt vein crystallization and other shock effects is not so large. The transformation of most plagioclase to maskelynite and the strong mosaicism in olivines within Umbarger indicate a bulk shock stage S5 and a shock pressure above 30–35 GPa (Stöffler et al. 1991). More recent shock experiments on the H chondrite Kernouvé have resulted in a slight recalibration of shock features in chondrites to lower pressure values (Schmitt 2000). Schmitt (2000) points out that most of the shock data used in calibrations of shock pressures was from single crystal and terrestrial rock samples that had little or no porosity whereas chondrites have porosities ranging from 5–20%. Porosity in a shocked sample provides a large variation in shock impedance that results in localized high temperatures and higher bulk temperatures, which enhance shock effects that are thermally activated such as the formation of maskelynite. For a starting temperature of 295 K, Schmitt (2000) observed the onset of strong mosaicism in olivine is 20 GPa and the transformation of oligoclase to maskelynite is 30 GPa, suggesting an onset of S5 effects at about 30 GPa. These shock effects may occur at even lower pressures in nature where single shock loading results in higher shock temperature (Bowden et al. 2000; Bowden 2002). The effect of higher shock temperatures is illustrated by Schmitt's (2000) 920 K starting temperature experiments, which formed maskelynite at pressures as low as 25 GPa. Based on Schmitt's (2000) calibration, the strong mosaicism in olivine and the formation of maskelynite may have formed at pressures as low as 25–30 GPa, which is still significantly higher than the ~ 18 GPa crystallization pressure. There are two possible solutions to this disparity. First, the crystallization pressure may represent crystallization during pressure release from about 25–30 GPa, but if this were the case the release rate was very slow. Alternatively, the pressure calibration of shock effects in chondrites is still too high because the near microsecond duration of shock experiments is much shorter than the milliseconds to seconds duration inferred from studies of naturally shocked meteorites.

CONCLUSIONS

Umbarger contains melt veins with a complex assemblage of high-pressure minerals that has not been previously described. The primary crystallization assemblage consists of ringwoodite, akimotoite, and Ca-clinopyroxene, which is metastable relative to a majorite-rich garnet

assemblage that commonly occurs in melt veins of highly shocked chondrites (Chen et al. 1996). In addition, Fe_2SiO_4 -spinel and stishovite crystallized from a FeO-SiO_2 -rich region of the silicate melt in the melt vein. This FeO-SiO_2 -rich silicate represents melting of material in Umbarger that was altered and metasomatized on the L-chondrite parent body prior to shock. Polycrystalline ringwoodite and hollandite-structured plagioclase represent transformations of olivine and plagioclase inclusions trapped in the melt vein.

The complex mixture of high-pressure minerals that crystallized in melt veins of Umbarger imply that the melt vein crystallization occurred at roughly 18 GPa. This value is much lower than the 45–90 GPa pressure one would conclude from the S6 shock effects in melt veins (Stöffler et al. 1991) and somewhat less than the 25–30 GPa S5 shock effects (Schmitt 2000) found in the bulk rock.

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