Revision 1

A new interpretation of decomposition products of serpentine under shock compression

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ABSTRACT

Dense hydrous magnesium silicates (DHMSs) may play an important role in water transport during planetary accretion and as water reservoirs in the Earth's deep mantle. We show that the dynamic decomposition products of antigorite, $Mg_3Si_2O_5(OH)_4$, can be interpreted to contain the newly discovered, dense hydrous silicate, phase H ($MgSiO_4H_2$). The Hugoniot for phase H was calculated based on the Hugoniots for its constituent oxides and the equation of state data derived from first principles calculations. The measured antigorite Hugoniot, previously suggested to decompose into high-pressure phases without generating fluid H_2O, was compared with those derived from calculations involving phase H. Sound velocity data were also compared to confirm that the dynamic breakdown product of antigorite at pressures above ~40 GPa is most likely phase H plus MgO without formation of fluid H_2O.

Keywords: Dense hydrous magnesium silicates, Phase H, High pressure, Hugoniot, Decomposition, Serpentine

INTRODUCTION

Due to their stabilities at high pressures, the dense hydrous magnesium silicates may provide important insights into deep-focus earthquakes, water sources for the Earth's interior, and formation of the primitive atmosphere and oceans (e.g., Tyburczy et al. 1990; Meade and Jeanloz 1991; Ulmer and Trommsdorff 1995; Peacock 2001; Drake 2005; Kawakatsu and Watada 2007; Sekine et al. 2012). Phase D ($MgSi_2O_6H_2$) previously was thought to be the only possible dense hydrous magnesium silicate (DHMS) present in the lower mantle (Irifune and Tsuchiya 2007). It has a wide stability field up to 40–50 GPa in pressure, at temperatures to ~1800 K, dehydrating to form an assemblage containing perovskite (Pv) and magnesiowustite (Mw) at higher temperatures (Shieh et al. 1998). Recently, using first principles methods Tsuchiya (2013) predicted a new high-pressure hydrous phase with composition $MgSiO_4H_2$ as a product of a high-pressure phase transition of phase D. Subsequently, Nishi et al. (2014) observed this phase (designated phase H) experimentally at ~50 GPa and 920 ^oC in both quench experiments and *in situ* X-ray diffraction measurements using multi-anvil apparatuses. At 0 K, phase H is theoretically predicted to be stable up to ~52 GPa before it dissociates into Pv plus H_2O (ice VIII) (Tsuchiya 2013). Phase H forms a solid solution with δ-AlOOH and the stability field of the resulting aluminous phase H (Al) expands to higher pressures and temperatures, extending into the lower mantle to depths of ~2000 km (Nishi et al. 2014). Shock wave experiments play an important role in understanding the dynamic behavior of hydrous minerals and their stability during impact process and can be used to study

their potential ability to be a water carrier.

METHOD AND RESULTS

From theoretical calculations (Tsuchiya 2013), phase H is characterized by the zero-pressure density, bulk modulus, and its first derivative of ρ_0 = 3.412 g/cm^3, K_0 = 185.8 GPa, K'_0 = 4.20, respectively, although hydrogen bond symmetrization can be expected to occur above ~30 GPa. A Hugoniot ($U_s = C_0 + su_p$, U_s shock velocity, u_p particle velocity, constants of C_0 and s) for phase H can be estimated using the equations of $C_0 = \sqrt{K_0/\rho_0}$ = 7.38 km/s and $s = (K_0' + 1)/4$ = 1.30, respectively. Moreover, phase H is compositionally a mixture of the phases brucite $Mg(OH)_2$ (Br) plus stishovite SiO_2 (St) which are stable at our pressures of interest. Therefore, the Hugoniot for phase H can also be calculated based on the known Hugoniots of Br and St using the additive volume law. This approch is applicable to estimate Hugoniots for an isochemical mixture of minerals with known Hugoniots (Al'tshuler and Sharipdzhanov 1971; Kalashnikov et al. 1973; Telegin et al. 1980). At pressure P, the specific volume of the mineral mixture $V(P)$ can be computed by means of the relation:

$$V(P) = \sum_{i=1}^{n} \alpha_i V_i(P) \quad (1)$$

Here, α_i is the weight fraction of mineral i, and $\sum_{i=1}^{n} \alpha_i = 1$. $V_i(P)$ is the specific volume of mineral i, and can be described as

$$V_i(P) = \left(1 - \frac{B_i - \sqrt{B_i^2 - 4A_i}}{2A_i}\right) V_{i,0} \quad (2)$$

Therein, $A_i = s_i^2$, $B_i = 2s_i + C_{i,0}^2/(V_{i,0}P)$, $V_{i,0}$, $C_{i,0}$, and s_i are the initial specific volume, the bulk sound velocity, and the slope of the shock Hugoniot of mineral i at

zero pressure, respectively. Hugoniot parameters calculated for phase H based on both the results of first principles calculations and the additive volume law, are listed in Table 1. The calculated Hugoniots are very close to each other for P > ~40 GPa. A predicted Hugoniot for phase D (ρ_0 = 3.49 g/cm^3) is also calculated from theoretical equation of state data (Tsuchiya et al. 2005), and listed in Table 1.

The Hugoniot for natural antigorite, $Mg_3Si_2O_5(OH)_4$, containing small amounts of Al_2O_3, FeO and Fe_2O_3, has been determined up to a pressure of 140 GPa (Sekine et al. 2012; Zhang et al. 2014). These results indicate that antigorite shows different Hugoniots depending on compression duration as shown in Fig. 1. A low-pressure phase (LPP) exists up to ~40 GPa. A metastable extension phase above ~40 GPa was observed in experiments with short compression durations (Fig. 1, open squares for antigorite in the metastable region ME). In shots with long compression durations, a decomposition of antigorite occurs above ~40 GPa to form high-pressure phase(s) (HPP). It is consistent with the lizardite data from Tyburczy et al. (1991) (Fig. 1, solid circles and squares for lizardite and antigorite, respectively). The decomposition is accompanied by a decrease in density and an increase in sound velocity along the Hugoniot relative to the metastable extension of the Hugoniot. However, the nature of the breakdown reactions has not yet been determined fully. Taking into account both phase D and phase H, possible reactions are as follows;

$$Mg_3Si_2O_5(OH)_4 = 2\ MgSiO_4H_2\ (H) + MgO\ (Pe) \qquad (3.44\ g/cm^3) \qquad (3)$$

$$Mg_3Si_2O_5(OH)_4 = MgSiO_3\ (Pv) + 1.5\ Mg(OH)_2\ (Br) + 0.5\ MgSi_2O_6H_2\ (D)$$

$$(3.19\ g/cm^3) \quad (4)$$

$Mg_3Si_2O_5(OH)_4$ =2 $Mg(OH)_2$ (Br) + $MgSiO_3$ (Pv) + SiO_2 (St) (3.16 g/cm^3) (5)

$Mg_3Si_2O_5(OH)_4$ = 2 $Mg(OH)_2$ (Br) + MgO (Pe) + 2 SiO_2 (St) (3.13 g/cm^3) (6)

$Mg_3Si_2O_5(OH)_4$ = $MgSi_2O_6H_2$ (D) + 2 MgO (Pe) + H_2O (3.03 g/cm^3) (7)

$Mg_3Si_2O_5(OH)_4$ = 2 $MgSiO_3$ (Pv) + MgO (Pe) + 2 H_2O (2.88 g/cm^3) (8)

$Mg_3Si_2O_5(OH)_4$ = 3 MgO (Pe) + 2 SiO_2 (St) + 2 H_2O (2.83 g/cm^3) (9)

The numbers in parentheses are the calculated zero-pressure densities for the mixtures on the right-hand side of each reaction. Among these possible breakdown reactions, we have compared the pressure-density, P-ρ, relations and sound velocity data to determine which reaction is the most probable in different pressure intervals.

We assumed reactions (5) and (6) with brucite for the high-pressure breakdown of antigorite (Sekine et al. 2012; Zhang et al. 2014) according to the estimate by Tyburczy et al. (1991). The zero-pressure densities calculated for the products of reactions (5) and (6) are 3.16 and 3.13 g/cm^3, respectively, while the estimated zero-pressure density is between 3.3 and 4.0 g/cm^3 for HPP of serpentine (lizardite) (Tyburczy et al. 1991). This suggests we should look for reactions to form more dense products than reactions (5) and (6). We propose reaction (3) with products of the zero-pressure density 3.44 g/cm^3 as the most probable decomposition reaction of antigorite. Hugoniot for decomposition product (3) was calculated based on the additive volume law using the data listed in Table 1. The results are shown in Figure 1. As descripted previously, Hugoniots for phase H were calculated using two independent methods (Table 1). Predicted Hugoniots for reaction (3) were also calculated by using the two approaches (shown by symbols + and * in Fig. 1) and they

129 are located closely. The measured Hugoniot data for antigorite as well as lizardite are
130 almost on the predicted Hugoniot at pressures of ~40–70 GPa as shown in Fig. 1. So,
131 in light of the newly discovered phase H, the high-pressure phase of antigorite and
132 lizardite can be re-interpreted most likely as the product of reaction (3). Further, the
133 measured Hugoniot data indicate that there appears a gap in the P-ρ plot at P > 70
134 GPa, suggesting that phase H may dissociate into Pv and H_2O (as fluid on the H_2O
135 phase diagram (Stewart and Ahrens 2005)), consistent with theoretical calculations
136 (Tsuchiya 2013). Serpentine may dehydrate by reaction (8) along the Hugoniot, as
137 indicated by static experiments (Shieh et al. 1998). The calculated Hugoniot for
138 reaction (8) is plotted in Figure 1 (symbols x) and shows good agreement with the
139 measured Hugoniot in the pressure range of ~80–125 GPa. The measured Hugoniot is
140 not consistent with reaction (4) (dash line in Fig. 1), which includes phase D,
141 probably because the Hugoniot temperature is too low in the stability field of phase D,
142 or because the reaction is kinetically inhibited. Moreover, phase D is only stable
143 below ~45 GPa (Irifune and Tsuchiya 2007), thus phase D may not be involved.

144 First principles calculations report similar longitudinal, shear, and bulk sound
145 velocities for phase D and phase H (Tsuchiya and Tsuchiya 2008; Tsuchiya 2013).
146 The calculated longitudinal sound velocities of phases D and phase H are 12.2 km/s
147 and 12.5 km/s at a pressure of 50 GPa, respectively. At ambient pressure, perovskite
148 and stishovite have high sound velocities, but serpentine, brucite and water have
149 lower ones. Under ambient conditions, the longitudinal sound velocity of serpentine is
150 6.76 km/s (Bezacier et al. 2010) and that for the products of reaction (3) is calculated

151 to be 9.6 km/s using the mixing model. The ambient condition values of 9.38 km/s for
152 phase H (Tsuchiya 2013) and 9.71 km/s for MgO (Zha et al. 2000) were used for
153 longitudinal sound velocities. This suggests that the reaction product (3) may have a
154 greater longitudinal velocity than the metastably compressed antigorite. Zhang et al.
155 (2014) observed an increase in the sound velocity for shocked antigorite above the
156 decomposition pressure of ~40 GPa. So, the sound velocity increase supports the
157 occurrence of reaction (3), although they did not measure the sound velocity above 80
158 GPa.

159 Serpentine (antigorite) is stable up to ~7 GPa at 540 ^{o}C (Ulmer and Trommsdorff
160 1995), and it decomposes or transforms into a series of phases with increasing
161 pressure. The products vary as a function of pressure and temperature: phase A +
162 enstatite (En) and phase clinohumite (chu) + En (Stalder and Ulmer 2001), phase D +
163 superhydrous phase B (sB) ± St, phase D + Pv ± St (Shieh et al. 1998), and phase H +
164 Pe. The schematic phase boundaries are shown in Figure 2. The stability for phase H
165 is known to extend substantially in the presence of Al_2O_3 by forming a solid solution
166 with δ-AlOOH (Nishi et al. 2014). Hugoniot temperature (T_H) of antigorite was
167 calculated by Zhang et al. (2014) using the relation (Tyburczy et al. 1991): $T_H =$
168 $T_0 exp\left(-\int_{V_0}^{V} \gamma/V\, dV\right) + (E_H - E_{tr} - E_S)/C_V$ (therein, T_0 the initial temperature, γ
169 the Grüneisen parameter, C_V the specific heat, E_H, E_{tr}, E_S the Hugoniot energy, phase
170 transition energy, and isentropic energy, respectively). The calculated T_H for the
171 antigorite HPP (Fig. 2) falls within the stability field for aluminous phase H (Al)
172 above ~40 GPa. Note that the phase boundary for phase H (Al) may have a negative

slope. With increasing shock pressure, the temperature will decrease if reaction (3) proceeds along the boundary. The calculated Hugoniot temperature for antigorite HPP (Zhang et al. 2014) indicated that it is relatively lower than the calculated temperature for the metastable extension. Therefore, shock temperature calculation also may support reaction (3). All the available data prefer the decomposition reaction (3) for antigorite at pressures of 40–70 GPa, although the decomposition may form amorphous material of which behaviors are similar to those of the reaction product (3).

GEOPHYSICAL IMPLICATIONS

We conclude that the new high-pressure dense hydrous phase H, $MgSiO_4H_2$, may be involved as the decomposition product of serpentine at pressures of ~40–70GPa and that the reaction does not produce fluid water. It suggests that serpentine can carry water as phase H to depth of the lower mantle below ~2000 km before dehydration, and that aluminous phase H (Al) would survive in violent impact events and at deeper mantle. This implies that serpentine plays a more important role than previously thought in primitive meteorites and in the Earth's mantle. Although phase H has never been found in nature, it will have to be recognized as the most stable hydrous silicate under extreme conditions. If even a trace of water is carried by phase H near the mantle-core boundary, the water reduces significantly the solidus of silicates (Nomura et al. 2014) and the temperature estimation at the mantle–core boundary also will be affected. Under impact conditions, it is unclear whether phase H formed by decomposition of serpentine is quenchable in the process of pressure

195 release or not. However, serpentine itself has been known to survive in Murchison
196 carbonaceous chondrite shocked up to a pressure of ~30 GPa (Tomioka et al. 2007).
197 Therefore, serpentine can be a potential water reservoir to carry water inward the
198 snowline during the planetary formation. The high pressure and temperature stability
199 of serpentine and phase H helps us to understand how water is carried to deeper and
200 hotter areas in the planetary interiors and to the planets near the Sun, although such
201 water carriers may have been subjected to heat and collision during migration and
202 accretion.
203

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208

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Figure captions

FIGURE 1. Comparison of pressure-density relations for the measured Hugoniot of antigorite (squares, not including experimental data based on scaling or "link factors" calculation, Sekine et al. 2012; Zhang et al. 2014) and lizardite (solid circles, Tyburczy et al. 1991), and Hugoniots of mixtures of 2 H + Pe (pluses and asterisks), 2 Pv + Pe + 2 H_2O (crosses), and Pv + 1.5 Br + 0.5 D (dash line). Curves with pluses (+) and asterisks (*) represent the calculated Hugoniots for 2 H + Pe by the additive volume law and by the data derived from the first principles calculations, respectively. The parameters and references used in the present study are listed in Table 1. LPP and HPP are the low pressure phase and high pressure phase of antigorite, respectively (Sekine et al. 2012). Metastable extension (ME) means the Hugoniot of antigorite in short compression durations and it moves to the HPP in long compression durations (Sekine et al. 2012). Symbols; H, phase H; D, phase D; Pv, perovskite; Pe, periclase; Br, brucite; St, stishovite.

FIGURE 2. Schematic illustration of phase boundaries for antigorite in pressure-temperature plot. Data are after Ulmer and Trommsdorff (1995), Shieh et al. (1998), Stalder and Ulmer (2001), and Nishi et al. (2014). The calculated Hugoniot temperature of antigorite is after Zhang et al. (2014). Symbols: A, phase A; Chu, clinohumite; sB, superhydrous phase B; others, same as in Fig. 1.

321 **TABLE 1.** Summary of Hugoniot parameters (density ρ_0, bulk sound velocity C_0, and
322 constant s at zero pressure) for minerals used in the present study.

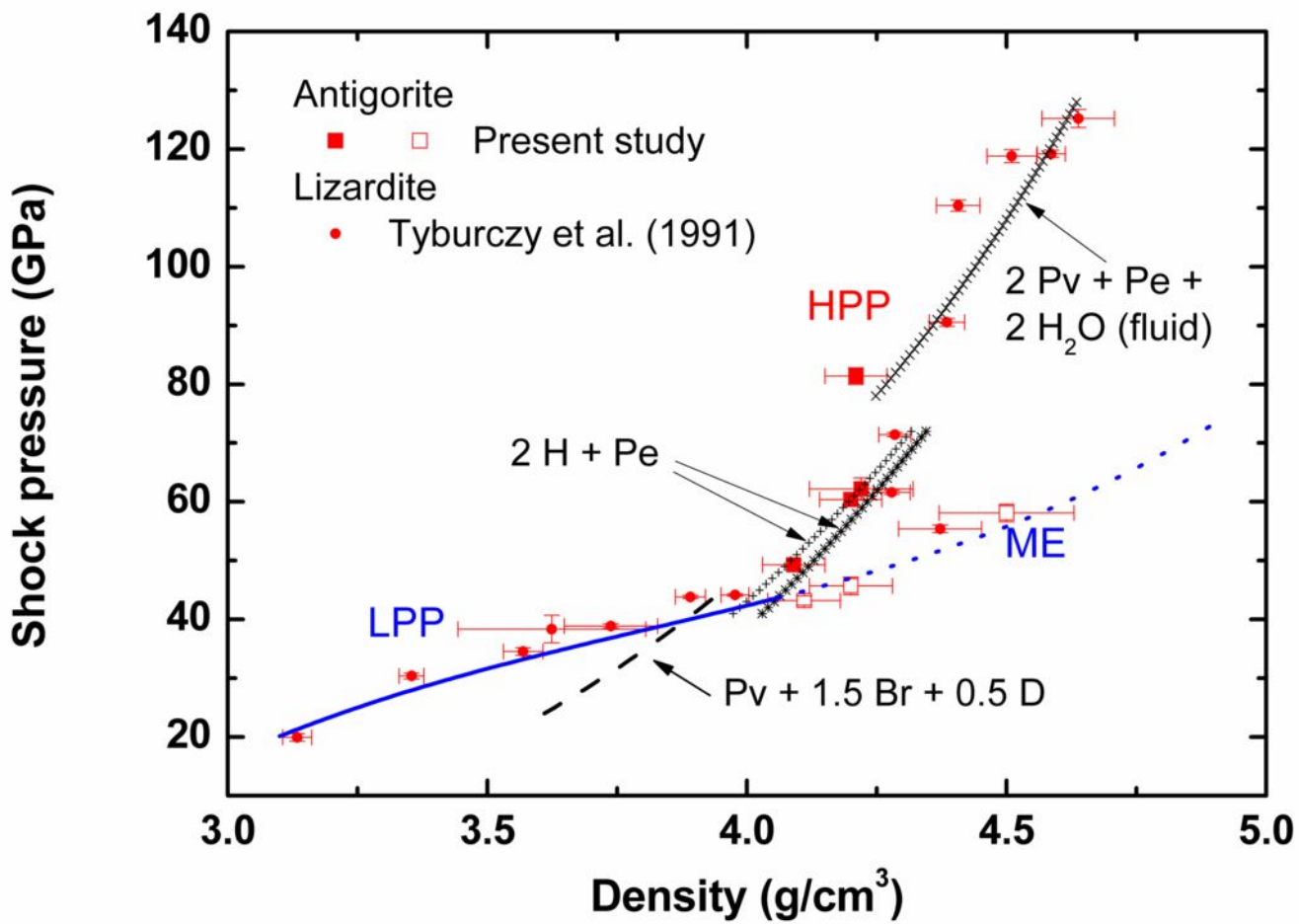
323

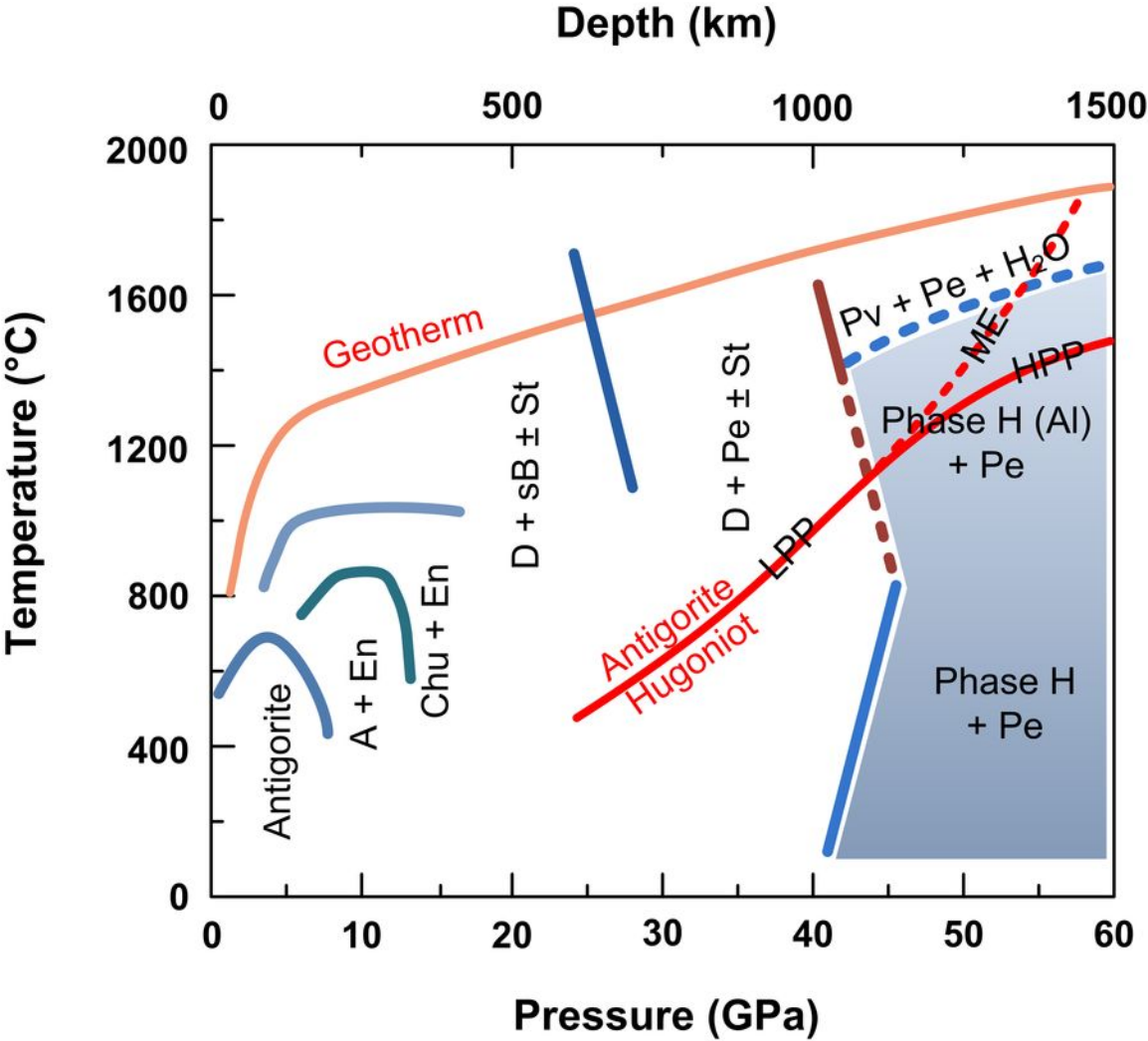
Minerals	Composition	ρ_0 (g/cm^3)	C_0 (km/s)	s	Pressures (GPa)	Sources
Brucite (Br)	$Mg(OH)_2$	2.382	4.76	1.35	<~100	(Simakov et al. 1974; Duffy et al. 1991)
Periclase (Pe)	MgO	3.584	6.61	1.36	<~200	(Marsh 1980; Vassiliou and Ahrens 1981)
Stishovite (St)	SiO_2	4.31	9.08	1.23	<~235	(Luo et al. 2002)
Enstatite* (En)	$MgSiO_3$	3.06	3.76	1.48	<~140	(Gong et al. 2004)
Water	H_2O	0.998	2.393	1.333	<~100	(Mitchell and Nellis 1982)
Phase D	$MgSi_2O_6H_2$	3.49	7.25	1.30	<~40	(Tsuchiya et al. 2005 + This work)†
Phase H	$MgSiO_4H_2$	3.41	7.38	1.30	~40–70	(Tsuchiya 2013 + This work)†
		3.08	5.76	1.31	~40–70	(This work)‡

324 * Perovskite (Pv) above ~40–50 GPa.

325 † calculated on the results of first principles calculations, respectively.

326 ‡ calculated using the Hugoniots of Br and St by the additive volume law.


Antigorite
Present study
Lizardite
Tyburczy et al. (1991)
HPP
2 Pv + Pe +
2 H2O (fluid)
2 H + Pe
ME
LPP
Pv + 1.5 Br + 0.5 D
Shock pressure (GPa)
Density (g/cm3)
140
120
100
80
60
40
20
3.0
3.5
4.0
4.5
5.0


Depth (km)
0
500
1000
1500
2000
1600
1200
800
400
0
Temperature (°C)
Geotherm
D + sB ± St
D + Pe ± St
Pv + Pe + H2O
ME
HPP
LPP
Phase H (Al)
+ Pe
Phase H
+ Pe
Antigorite
Hugoniot
Antigorite
A + En
Chu + En
0
10
20
30
40
50
60
Pressure (GPa)