

MANTLE GEOPHYSICS

Elasticity of davemaoite as a primary contributor to lower-mantle heterogeneities

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Geophysical detection of subducted mid-ocean ridge basalt (MORB) in the lower mantle is hindered by uncertainties in the elasticity of Fe,Al,Mg,Ti-bearing davemaoite, a key MORB component. Using Brillouin spectroscopy and x-ray diffraction, we determined the elasticity of a $\text{Ca}_{0.906(1)}\text{Fe}^{2+}_{0.027(1)}\text{Fe}^{3+}_{0.042(1)}\text{Mg}_{0.033(1)}\text{Al}_{0.072(1)}\text{Ti}_{0.020(1)}\text{Si}_{0.912(1)}\text{O}_3$ davemaoite up to 113 gigapascals and 2294 K. We found that it exhibited a shear wave velocity 10 to 20% slower than end-member davemaoite, making it the slowest phase among major lower-mantle minerals. Our models show that MORB, containing 20 to 25 volume percent davemaoite, potentially contributes to large low-shear-velocity provinces (LLSVPs), whereas a cumulate layer enriched in davemaoite crystallized from basal magma ocean may comprise ultralow-velocity zones (ULVZs). Davemaoite's ability to host incompatible and heat-producing elements possibly links LLSVPs and ULVZs to mantle plume initiation and geochemical signatures of ocean island basalts.

Recent seismic observations have revealed extensive heterogeneities within Earth's thickest layer, the lower mantle. Tomographic imaging has shown that some subducting slabs descend through the mantle transition zone and into the lowermost mantle; for example, the Farallon slab beneath North America (1). Additionally, relatively small-scale (several kilometers) seismic scatterers, known as midmantle scatterers, have been identified at depths between 700 and 1900 km (2). These scatterers exhibit three key characteristics: (i) they cluster below subduction zones, such as underneath the Japan Sea, at ~1000-km depth; (ii) they exhibit velocity anomalies ranging from -12% to +10%; and (iii) they are seismically sharp (2). Although midmantle scatterers largely disappear below 1900 km, additional heterogeneities exist deeper in the lower mantle. Seismic studies have identified two large low-shear-velocity provinces (LLSVPs) beneath the central Pacific Ocean and Africa that are the largest known lower-mantle heterogeneities. These structures cover ~30% of the core-mantle boundary (CMB) and rise as far as ~1000 km above it (3, 4). LLSVPs are characterized by shear wave velocity (V_s) and compressional wave velocity (V_p) reductions of >2 and >0.5%, respectively, along with ~1.0% positive density anomalies and sharp velocity gradients at their margins (3, 4). Additionally, dozens of ultralow-velocity zones (ULVZs), which are thin patches (5 to 40 km thick) with V_p and V_s reductions of up to 25 and 50%, respectively, have also been detected along the CMB (5). Thus, although structurally simpler than the upper mantle, the lower mantle remains a heterogeneous and dynamically complex region.

Multiple hypotheses have been proposed to explain these heterogeneities. For example, it has been proposed that they represent remnants of an ancient basal magma ocean, where Fe-rich minerals or melts may have accumulated at the base of the mantle, remaining compositionally distinct due to limited convective mixing over geologic time (6, 7). This mechanism has been invoked to explain LLSVPs and ULVZs because Fe-rich materials are both seismically slow and denser than surrounding mantle (8). Another hypothesis involves potential chemical reactions with or percolation of liquid metallic Fe at the CMB, enriching the lowermost mantle in Fe, and reducing V_p and V_s (7, 9). A third hypothesis attributes LLSVPs and midmantle scatterers to subducted oceanic crust (10–12), which is inherently enriched in Fe relative to ambient mantle (13). Phase transitions or spin-state changes in minerals comprising the subducted oceanic crust, such as stishovite and Fe-bearing bridgmanite, can also cause velocity reductions (11, 14). More recently, it has been proposed that lower-mantle heterogeneities could preserve fragments of Theia's mantle from the Moon-forming impact (15).

Despite different geologic explanations, most hypotheses invoke Fe-rich materials as the mineralogical cause of the lower-mantle heterogeneities. Elasticity data for Fe-bearing lower-mantle minerals are limited to bridgmanite and ferropericlase (14, 16–18), whereas Fe-bearing davemaoite remains poorly studied. Here, we focused on Fe-bearing davemaoite, which also contains small amounts of Al, Mg, and Ti and has a chemical composition representative of the subducted davemaoite expected in lower-mantle mid-ocean ridge basalt (MORB) ($\text{Ca}_{0.906(1)}\text{Fe}^{2+}_{0.027(1)}\text{Fe}^{3+}_{0.042(1)}\text{Mg}_{0.033(1)}\text{Al}_{0.072(1)}\text{Ti}_{0.020(1)}\text{Si}_{0.912(1)}\text{O}_3$). Although our primary goal was to investigate the seismic signatures of subducted oceanic crust, our results also offer insights into other proposed formation mechanisms of lower-mantle heterogeneities.

Compared with the ambient mantle, MORB is enriched in Si, Al, Ti, and Fe. Below 800-km depth, MORB consists of ~33 vol % bridgmanite, ~22 vol % Fe,Al,Mg,Ti-bearing davemaoite, ~22 vol % CF-phase (with calcium ferrite structure), and ~22 vol % stishovite (13). Its higher density enables continuous subduction down to the CMB (12). However, whether MORB is seismically slower or faster than the surrounding mantle remains controversial (10, 19), largely due to the lack of high pressure-temperature (high P - T) elasticity data for relevant lower-mantle minerals. Among these minerals, davemaoite presents the greatest challenge because of its unquenchable nature. Therefore, geophysical detection of subducted crust in the lower mantle is hindered by sparse experimental constraints on the elastic properties of key phases, particularly davemaoite, at relevant P - T conditions.

Previous studies have measured sound velocities (V_p or V_s) of end-member CaSiO_3 davemaoite under high-pressure/room-temperature conditions (32 to 133 GPa at 300 K) using Brillouin spectroscopy with diamond anvil cells (DACs) and moderate-pressure/high-temperature conditions (7 to 22 GPa at 300 to 1700 K) using ultrasonic interferometry coupled with multi-anvil press (MAP) (10, 20, 21). The room-temperature sound velocity data agree well across methods and with recent machine learning-based computational predictions (22), but they are consistently lower than earlier computational results (23). However, high-temperature experimental sound velocity data show considerable discrepancies (10, 21) that are also lower than computational predictions (22, 24). The presence of davemaoite in the ambient lower mantle remains debated (25, 26); if present, its composition likely approximates the end-member CaSiO_3 (27). By contrast, davemaoite formed in subducted MORB, basal magma ocean remnants, or other processes is expected to be enriched in Fe and Al. Hirose *et al.* (12) and Ishii *et al.* (13) have reported compositions of davemaoite formed in MORB through high P - T experiments in DACs and MAPs, respectively (table S1). The Fe, Al, Mg, and Ti contents determined in both studies are generally consistent within measurement uncertainties. We adopted the chemical composition reported in Ishii *et al.* (13) because of the slightly larger grain size of their recovered davemaoite,

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which yielded smaller analytical uncertainties. Nevertheless, we acknowledge the variations in reported mineral compositions in MORB (12, 13) arising from technical challenges of energy-dispersive x-ray spectroscopy analysis on ultrafine amorphous grains and possible dependence on the synthesis P - T conditions.

In this study, we synthesized davemaoite samples (end-member CaSiO_3 and Fe,Al,Mg,Ti-bearing davemaoite) using CO_2 laser-heated DACs, measured their elasticity using Brillouin spectroscopy and powder x-ray diffraction (XRD) (see the materials and methods), and modeled seismic velocities of davemaoite along mantle geotherms to demonstrate its key role in generating lower-mantle heterogeneities.

In situ synthesis and Brillouin spectroscopy experiments of davemaoite

We synthesized end-member CaSiO_3 davemaoite at high P - T conditions in DACs using CaSiO_3 glass and natural near-end-member wollastonite crystals as starting materials (table S1). Then, we measured these samples for their V_p and V_s at 20 to 30 GPa and 300 K using Brillouin spectroscopy (see the materials and methods). As shown in Fig. 1, our results align with previous Brillouin spectroscopy and ultrasound interferometry measurements on tetragonal end-member davemaoite (20, 21), confirming successful synthesis and reliable experimental procedures. The V_p and V_s values of tetragonal end-member CaSiO_3 davemaoite in this study match those reported by Kudo *et al.* (20) and Gréaux *et al.* (21), although V_p measurements from Kudo *et al.* (20), estimated using high-pressure XRD-derived isothermal bulk modulus and V_s , are slightly higher because they are subject to greater uncertainties. By contrast, Thomson *et al.* (10) reported systematically lower sound velocities, especially at high-temperature conditions (Fig. 1); this cannot be explained solely by compositional differences and highlights the need for further high-temperature studies on end-member CaSiO_3 davemaoite.

Following identical experimental procedures (see the materials and methods and Fig. 2), we also synthesized Fe,Al,Mg,Ti-bearing

davemaoite ($\text{Ca}_{0.906(1)}\text{Fe}^{2+}_{0.027(1)}\text{Fe}^{3+}_{0.042(1)}\text{Mg}_{0.033(1)}\text{Al}_{0.072(1)}\text{Ti}_{0.020(1)}\text{Si}_{0.912(1)}\text{O}_3$) and acquired V_p and V_s up to 103 GPa along a 300 K isotherm in its low-temperature tetragonal form (Fig. 1). These were found to be 10 to 20% lower than the V_p and V_s of end-member CaSiO_3 davemaoite (Fig. 1), whereas unit cell volumes, determined through synchrotron powder XRD, were slightly larger (figs. S1 and S2). We performed a joint fit of sound velocities and unit-cell volumes (tables S2 and S3) using Eulerian strain equations of state (28). This fit yielded the following aggregate elastic properties of the tetragonal Fe,Al,Mg,Ti-bearing davemaoite: $V_0 = 50.2(2)\text{Å}^3$, $K_{S0} = 129(5)$ GPa, $G_0 = 56.1(6)$ GPa, $K_{S0}' = 4.9(2)$, and $G_0' = 1.50(2)$ (see supplementary text 1). Although davemaoite's unquenchable nature precludes direct measurements of V_0 , our multiparameter fitting approach including V_p , V_s , and unit-cell volume provides robust constraints on V_0 (see supplementary text 4). The use of different NaCl pressure scales affects K_{S0} and K_{S0}' , but the calculated high-pressure sound velocities and densities using different parameter sets are similar because of the well-known trade-offs between them (table S6).

Chao *et al.* (29) suggested that replacing every 1 mol % Si with Ti in CaSiO_3 increases its V_0 by $\sim 0.4\%$, consistent with Ti's larger radius. Our results similarly indicate that Ti doping expands its unit cell. Substituting ~ 9 mol % Si with Ti and Al and ~ 10 mol % Ca with Fe and Mg collectively increased V_0 by 10%. Future experiments and/or first-principles calculations on compositionally controlled davemaoite samples are needed to isolate the effects of individual substituents.

Compared with end-member davemaoite, the K_{S0} and G_0 values of Fe,Al,Mg,Ti-bearing davemaoite are 40 to 50% lower (10, 21) (table S4). This increased compressibility and reduced sound velocities likely result from several factors. First, Al^{3+} substitution introduces oxygen vacancies ($\text{Si}^{4+} \rightarrow \text{Al}^{3+} + \text{vacancy}$) (18), softening the perovskite structure. Second, Fe^{3+} substitution ($3\text{Ca}^{2+} \rightarrow 2\text{Fe}^{3+} + \text{vacancy}$) creates additional A-site vacancies (30). Finally, incorporation of Fe^{2+} , Fe^{3+} , Mg^{2+} , Al^{3+} , and Ti^{4+} distorts the A (or dodecahedral) and B (or octahedral) sites because of their size mismatch with Ca^{2+} and Si^{4+} , leading

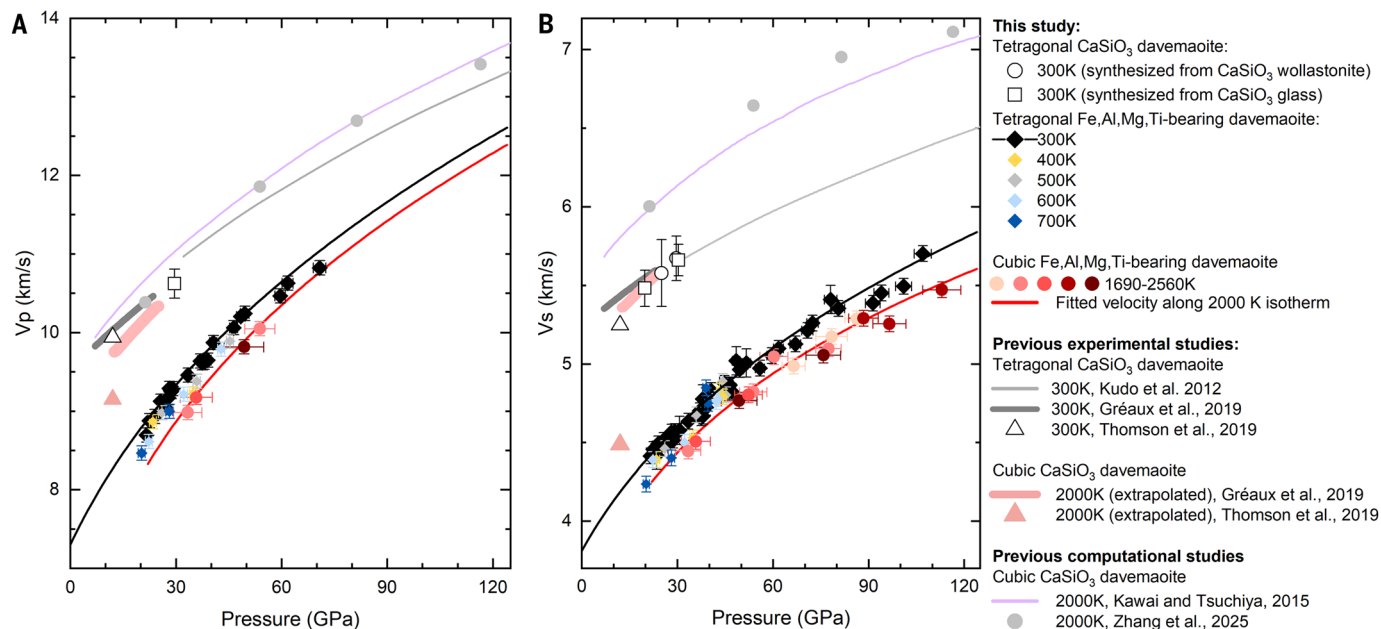


Fig. 1. Sound velocities of tetragonal and cubic davemaoite in the present study and previous work. (A) V_p of davemaoite. (B) V_s of davemaoite. Best-fit velocity curves for tetragonal and cubic davemaoite are along 300 and 2000 K isotherms, respectively. All numerical data from this study are listed in table S3. Vertical error bars represent 1 SD based on multiple sound velocity measurements on the same sample at each P - T condition. Horizontal error bars for 300 to 700 K data reflect pressure uncertainties derived from ruby fluorescence measurements before and after each run, as well as intrinsic ruby pressure scale uncertainty. For high-temperature data (>1600 K), horizontal error bars represent pressure uncertainties bracketed by isobaric and isochoic boundary conditions.

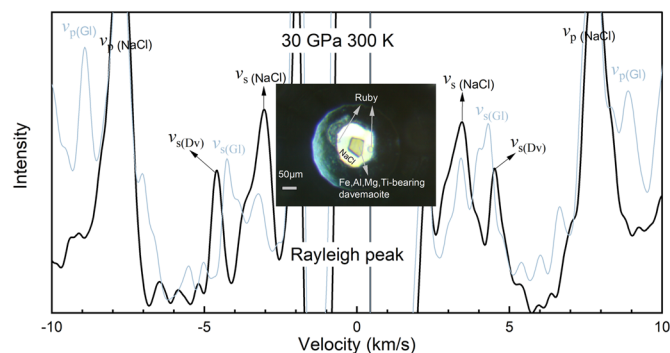


Fig. 2. Brillouin spectra of Fe,Al,Mg,Ti-bearing glass and davemaoite at 30(1) GPa and 300 K. Spectra of glass (Gl) and davemaoite (Dv) were collected before and after CO₂ laser heating, respectively. Disappearance of the glass peaks suggests complete transformation to crystalline davemaoite. The accompanying optical microscope image shows the sample chamber at ~30 GPa and 300 K containing the tetragonal Fe,Al,Mg,Ti-bearing davemaoite sample, ruby spheres, and pressure-transmitting medium.

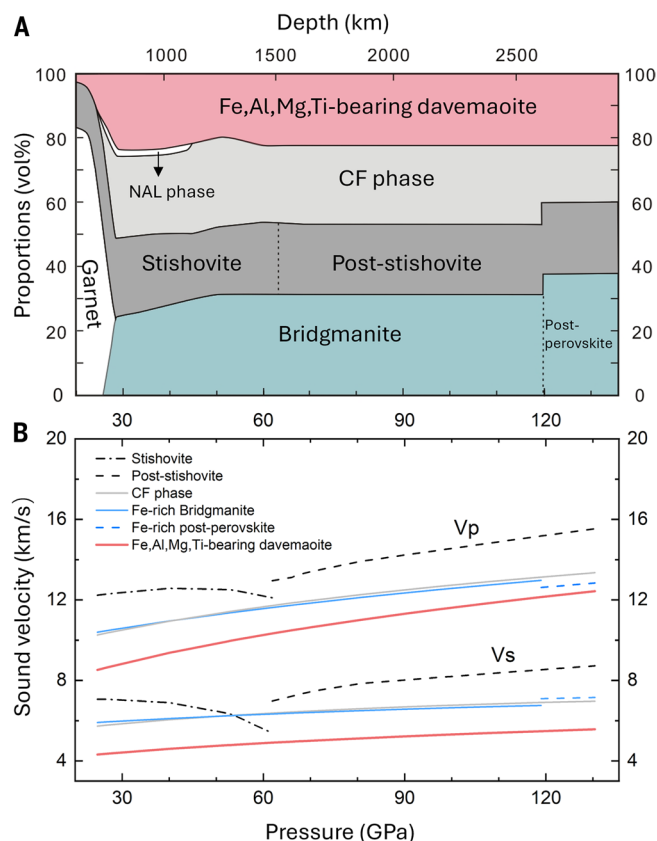


Fig. 3. Mineral proportions in MORB and sound velocities of constituent phases. (A) Phase diagram of MORB in the lower mantle (13). (B) Sound velocities of lower-mantle minerals along a 1600 K adiabatic geotherm (see tables S4 and S5 and supplementary text 2). NAL, new aluminum-rich phase; CF, aluminum-rich phase with a calcium ferrite structure.

to reduced incompressibility. Similar mechanisms have been proposed to explain the observed lower sound velocities and greater compressibility of Fe,Al-bearing bridgmanite compared with end-member MgSiO₃ bridgmanite (18, 30).

The V_p and V_s of Fe,Al,Mg,Ti-bearing davemaoite were also measured at 400 to 700 K and 20 to 45 GPa using resistively heated DACs (Fig. 1). We did not fit the 400 to 700 K data to obtain the temperature derivatives of the elastic moduli because of potential influence from the tetragonal-cubic phase transition (10, 21, 22, 31). For example, Thomson *et al.* (10) observed the gradual increase of V_p and V_s of tetragonal davemaoite with temperature at 12 GPa before the tetragonal-to-cubic transition was completed at 420 K. Gréaux *et al.* (21) similarly reported a continuous increase of V_p and V_s up to 600 K and did not fit the data across this transition. Although the transition in tetragonal Fe,Al,Mg,Ti-bearing davemaoite likely occurs at higher temperatures (12, 32), it remains unclear whether our 400 to 700 K sound velocity data were affected. Future high-resolution high P - T XRD and additional sound velocity measurements at finer P - T intervals are needed to constrain temperature derivatives of the elastic moduli and to capture the sound velocity change across this transition.

To determine the elasticity of cubic davemaoite, we conducted Brillouin spectroscopy measurements on cubic Fe,Al,Mg,Ti-bearing davemaoite at 33 to 113 GPa and 1600 to 2600 K in CO₂ laser-heated DACs (33). Similar to the tetragonal phase, V_p and V_s of cubic Fe,Al,Mg,Ti-bearing davemaoite are lower than those of the end-member CaSiO₃ davemaoite, likely also due to Fe, Al, Mg, and Ti incorporation (Fig. 1). The fitted elastic properties were: $K_{S0} = 138(3)$ GPa, $G_0 = 67(1)$ GPa, $G'_0 = 1.43(2)$ by fixing $V_0 = 50.2$ Å³, $K_{S0}' = 4.9$, $\theta_0 = 1000.0$ K, $\gamma_0 = 1.42$, $q_0 = 2.65$, and $\eta_{S0} = 1.54$ (14) (see supplementary text 1). Compared with its lower-temperature tetragonal polymorph, the cubic Fe,Al,Mg,Ti davemaoite exhibits a 6% higher K_{S0} and a 19% higher G_0 , which is qualitatively consistent with previous studies of end-member CaSiO₃ davemaoite (21, 23). For example, Stixrude *et al.* (23) proposed that G of the cubic phase is much larger than that of the tetragonal polymorph due to the softening of C_{44} and C_{66} in the tetragonal structure, although they found little difference in K_S . Gréaux *et al.* (21) later found that both K_{S0} and G_0 are ~9% higher than the tetragonal form.

It is worth noting that the Fe in Fe,Al,Mg,Ti-bearing davemaoite samples ($\text{Fe}^{3+}/\Sigma\text{Fe} = 0.6$ to 0.62) used in this study is dominantly ferric (see the materials and methods and fig. S6). Bridgmanite, the most abundant lower-mantle mineral, with a similar perovskite structure, can incorporate substantial ferric Fe ($\text{Fe}^{3+}/\Sigma\text{Fe}$ up to 0.98) (18). Frost *et al.* (34) has demonstrated that crystallization of bridgmanite from the early Earth's magma ocean likely involved charge disproportionation of ferrous iron (Fe^{2+}), producing metallic iron (Fe^0) that may have segregated into the core, leaving behind Fe^{3+} -rich bridgmanite. Given the structural similarity between bridgmanite and davemaoite, it is plausible that the ferric Fe-rich davemaoite used in this study realistically represents davemaoite in lower-mantle MORB.

Contribution of davemaoite to lower-mantle heterogeneities

Fe,Al,Mg,Ti-bearing davemaoite is seismically slower than not only end-member CaSiO₃ davemaoite, but also most major lower-mantle minerals. Thus, a reassessment of davemaoite's role in lower-mantle heterogeneities is needed. Among the four major mineral phases in lower-mantle MORB (Fig. 3A), cubic Fe,Al,Mg,Ti-bearing davemaoite exhibits the lowest seismic wave velocities (Fig. 3B). It is also the least dense phase below 50 GPa but becomes denser than stishovite or post-stishovite above 50 GPa (fig. S7). Using the new elasticity data of Fe,Al,Mg,Ti-bearing davemaoite and existing elasticity data of other lower-mantle minerals, we computed the updated seismic velocities of MORB, pyrolite, and chondritic lower-mantle composition and compared them with the Preliminary Reference Earth Model [PREM (35)] (for details, see supplementary text 2). We found that despite stishovite's ultrahigh velocities (11), MORB is seismically slower than pyrolite, the chondritic mantle,

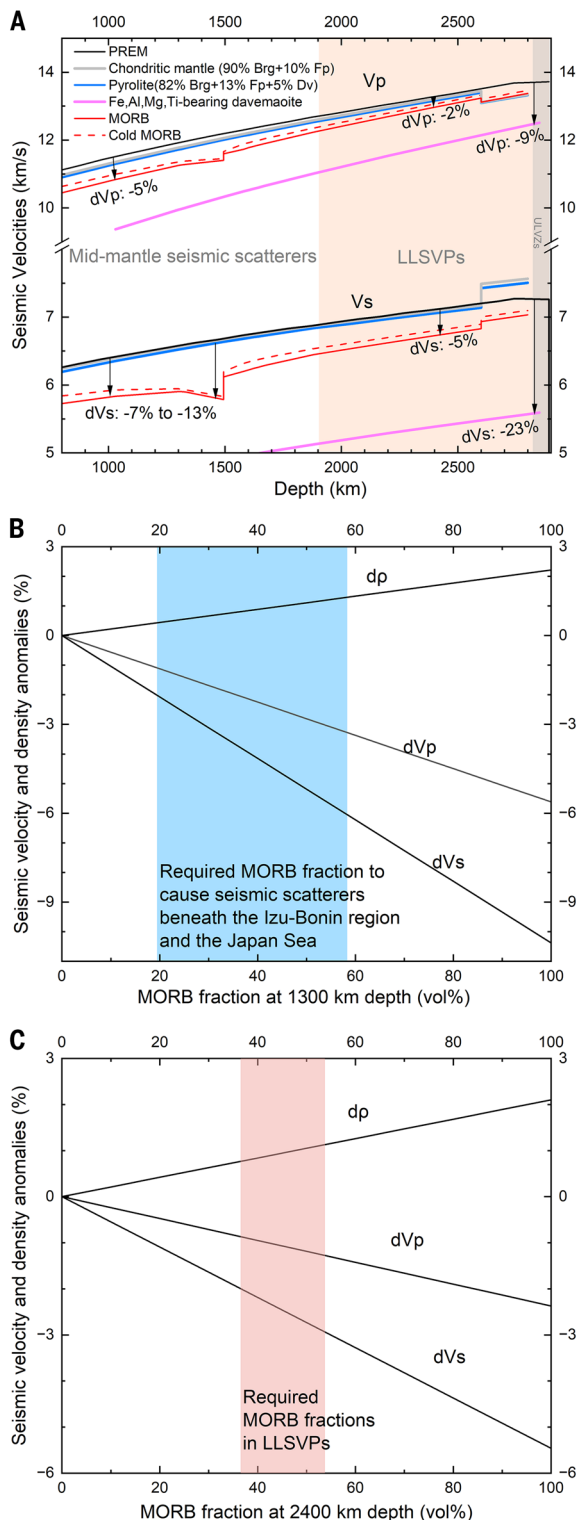


Fig. 4. Seismic velocity and density anomalies of MORB compared with other mantle petrologic compositions and mineral phases. (A) Seismic velocities of MORB, cubic Fe,Al,Mg,Ti-bearing davemaoite, the pyrolytic and chondritic mantle along a 1600 K adiabatic geotherm, and MORB along a 1100 K adiabatic geotherm (see tables S4 and S5 and supplementary text 2) compared with the one-dimensional seismic reference model PREM (35). (B) Seismic velocity and density anomalies induced by MORB at midmantle depths (1300 km). (C) Seismic velocity and density anomalies induced by MORB at lowermost mantle depths (~2400 km). Velocity differences (dV_p and dV_s) were calculated with respect to PREM.

and PREM at depths from 800 to 2900 km (Fig. 4A), highlighting MORB as a potential source of low-velocity rather than high-velocity anomalies in the lower mantle. Additionally, MORB remains denser than the surrounding ambient mantle at depths greater than ~800 km (fig. S8), supporting its continued subduction and long-term accumulation in the deep mantle.

Seismic heterogeneities are observed at various depths in the lower mantle, including seismic scatterers in the midmantle and LLSVPs and ULVZs in the lowermost mantle. For instance, seismic scatterers with -2 to -6% V_s anomalies observed beneath the Izu-Bonin region and Japan Sea at ~1000-km depth imply the presence of 20 to 60 vol % MORB (2) (Fig. 4B). Supporting evidence includes: (i) the thickness of most observed scatterers (~10 km) is consistent with subducted oceanic crust (2); (ii) many of them cluster near circum-Pacific subduction zones (36); and (iii) the presence of MORB can reduce V_s by up to 7% at ~1000-km depth (Fig. 4A). By contrast, the less frequently observed positive V_s anomalies, such as those beneath the Tonga and Solomon areas at 700-km depth (2), likely require alternative explanations, including cold harzburgite (21) or subducted stishovite-rich continental materials, given stishovite's high seismic velocities (11).

LLSVs show 2 to 3% V_s reductions and ~1% V_p reductions, along with ~1% positive density anomaly (3, 4), requiring the existence of 35 to 55 vol % MORB in the lowermost lower mantle (Fig. 4C). Assuming a MORB subduction flux of 10 km³/year into the lower mantle and the fact that LLSVPs occupy 2% of the entire mantle's volume (37), it would take ~800 million years for the accumulated MORB material to account for 35 to 55 vol % of the combined volume of the two LLSVPs. MORB remains denser than the ambient lower mantle without requiring thermal anomalies (fig. S8) (12), supporting its potential accumulation and contribution to LLSVPs. This interpretation contrasts with Wang *et al.* (19), who argued that MORB exhibits higher velocities than the ambient lower mantle and thus is an unlikely component of LLSVPs. Their assumption that Fe,Al,Mg,Ti-bearing davemaoite shares the same elastic properties with end-member CaSiO₃ is challenged by the results of the present study. Therefore, this study reaffirms the viability of MORB accumulation in the lowermost mantle as a plausible explanation for LLSVP formation without invoking additional mechanisms. Notably, MORB can exhibit chemical variations (e.g., FeO = 8 to 13 wt %; Al₂O₃ = 13 to 17 wt %) due to different mantle sources and degrees of melting (38), likely producing davemaoite with slightly variable chemical compositions. Elevated Al³⁺ and Fe³⁺ concentrations can increase vacancy densities in the crystal structure, which may further reduce sound velocities.

Seismically detected ULVZs exhibit extreme velocity reductions of 5 to 30% in V_p and 10 to 50% in V_s (5). MORB alone cannot explain these anomalies but may contribute to ULVZs. An alternative explanation is that ULVZs may represent remnants of a basal magma ocean (39). Previous studies attributed the seismic characteristics of ULVZs to Fe-rich melt or wüstite derived from a basal magma ocean (16), but the potential contribution of davemaoite has never been considered. As a major phase crystallizing during late-stage basal magma ocean cooling (40), a solid layer enriched in davemaoite cumulates may persist above the CMB (25, 41). Residual melts from the basal magma ocean would have become increasingly Fe-enriched during crystallization (42), and core-mantle interactions may have further enriched davemaoite in Fe (43). Consequently, the solid layer enriched in davemaoite cumulates would likely be Fe rich. The Fe,Al,Mg,Ti-bearing davemaoite measured in this study, containing 4 wt % FeO, is already 23% slower in V_s and 9% slower in V_p than the surrounding mantle. Although the maximum Fe content in davemaoite near the CMB remains unconstrained, we propose that an Fe-rich davemaoite layer could be a viable explanation for ULVZs. Although Fe incorporation is likely to lower the melting temperature, end-member CaSiO₃ davemaoite is suggested to have an extremely high melting point (42), implying that Fe-rich davemaoite may remain solid near the CMB. Future studies

exploring the melting behavior and elasticity of Fe-rich davemaoite are needed to assess its stability and quantify its contribution to ULVZs.

Our findings also have implications for the thermochemical evolution of the mantle. Compared with other lower-mantle minerals, davemaoite has a strong affinity for incompatible elements, including heat-producing elements (Th and U) and rare earth elements (44). If LLSVPs and ULVZs represent regions enriched in davemaoite, then they would serve as the deepest long-term geochemical reservoirs of these elements, facilitating plume initiation. Indeed, seismic observations show that several mantle plumes, including those in Hawaii, Iceland, and the South Pacific Superswell, originate from LLSVPs and ULVZs (45) that may be enriched in davemaoite. Furthermore, oceanic island basalts from these plumes, are known to exhibit high concentrations of incompatible elements (rare earth elements, Th, and U), such as those from the Austral Islands and Tristan-Inaccessible Island (25, 46). These correlations suggest that lower-mantle heterogeneities that are likely enriched in davemaoite may act as long-term localized heat sources and complex geochemical reservoirs in the deep Earth.

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SUPPLEMENTARY MATERIALS

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Elasticity of davemaoite as a primary contributor to lower-mantle heterogeneities

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Editor's summary

Seismic images of Earth's lower mantle revealed two continental-scale regions with anomalously slow shear-wave velocities rising from the core-mantle boundary. Understanding their origin depends on knowing their mineralogy, a challenge to measure under conditions at ~2500 kilometers depth. Zhou *et al.* used Brillouin spectroscopy and x-ray diffraction, with carbon dioxide laser heating, to synthesize and measure at lower-mantle conditions the elasticity of Fe-, Al-, Mg-, and Ti-bearing davemaoite, a common component of subducted oceanic crust. Their results showed that davemaoite is seismically slow, making it a likely contributor to low-velocity regions of the lower mantle, possibly through accumulation of subducted oceanic crust. —Angela Hessler

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