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Elasticity of davemaoite as a primary contributor to lower mantle heterogeneities

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Abstract

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 $\text{Ca}_{0.906(1)}\text{Fe}^{2+}_{0.027(1)}\text{Fe}^{3+}_{0.042(1)}\text{Mg}_{0.033(1)}\text{Al}_{0.072(1)}\text{Ti}_{0.020(1)}\text{Si}_{0.912(1)}\text{O}_3$ davemaoite up to 113 GPa and 2294 K. It exhibits V_s 10–20% slower than endmember davemaoite, making it the slowest phase among major lower-mantle minerals. Our models show that MORB, containing 20–25 vol% davemaoite, potentially contributes to large low-shear velocity provinces (LLSVPs), while a cumulate layer enriched in davemaoite crystallized from basal magma ocean may comprise ultra-low 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.

27 Main Text

28 Recent seismic observations have revealed extensive heterogeneities within the Earth's
29 thickest layer—the lower mantle. Tomographic imaging shows that some subducting slabs descend
30 through the mantle transition zone and into the lowermost mantle, such as the Farallon slab beneath
31 North America (1). Additionally, relatively small-scale (several kilometers) seismic scatterers,
32 known as mid-mantle scatterers, have been identified at depths between 700 and 1900 km (2).
33 These scatterers exhibit three key characteristics: 1) they cluster below subduction zones, such as
34 underneath the Japan Sea, at ~1000 km depth; 2) they exhibit velocity anomalies ranging from -
35 12% to +10%; 3) they are seismically sharp (2). While mid-mantle scatterers largely disappear
36 below 1900 km, additional heterogeneities exist deeper in the lower mantle. Seismic studies have
37 identified two large low-shear velocity provinces (LLSVPs) beneath the central Pacific and Africa,
38 which are the largest lower mantle heterogeneities. These structures cover ~30% of the core–
39 mantle boundary (CMB) and rise as far as ~1000 km above it (3,4). LLSVPs are characterized by
40 shear wave velocity (V_s) and compressional wave velocity (V_p) reductions of over 2% and 0.5%,
41 respectively, along with ~1.0% positive density anomalies and sharp velocity gradients at their
42 margins (3,4). Additionally, dozens of ultra-low velocity zones (ULVZs), which are thin patches
43 (5 to 40 km thick) with V_p and V_s reductions of up to 25% and 50%, respectively, have also been
44 detected along the CMB (5). Thus, although structurally simpler than the upper mantle, the lower
45 mantle remains a heterogenous and dynamically complex region.

46 Multiple hypotheses have been proposed to explain these heterogeneities. For example, it
47 has been proposed that they represent remnants of an ancient basal magma ocean, where Fe-rich
48 minerals or melts may have accumulated at the base of the mantle, remaining compositionally
49 distinct due to limited convective mixing over geologic time (6,7). This mechanism has been
50 invoked to explain LLSVPs and ULVZs, because Fe-rich materials are both seismically slow and
51 denser than surrounding mantle (8). Another hypothesis involves potential chemical reactions with
52 or percolation of liquid metallic Fe at the CMB, enriching the lowermost mantle in Fe, and
53 reducing V_p and V_s (7,9). A third hypothesis attributes LLSVPs and mid-mantle scatterers to
54 subducted oceanic crust (10-12), which is inherently enriched in Fe relative to ambient mantle (13).
55 Phase transitions or spin-state changes in minerals comprising the subducted oceanic crust, such
56 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 focus on Fe-bearing davemaoite that also contains small amounts of Al, Mg, and Ti, with a chemical composition representative of subducted davemaoite expected in lower mantle mid-ocean ridge basalt (MORB) ($\text{Ca}_{0.906(1)}\text{Fe}^{2+}_{0.027(1)}\text{Fe}^{3+}_{0.042(1)}\text{Mg}_{0.033(1)}\text{Al}_{0.072(1)}\text{Ti}_{0.020(1)}\text{Si}_{0.912(1)}\text{O}_3$). While our primary goal is to investigate the seismic signatures of subducted oceanic crust, the results also offer insights into other proposed formation mechanisms of lower mantle heterogeneities.

Compared to 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 P - T elasticity data for relevant lower mantle minerals. Among these minerals, davemaoite presents the greatest challenge due to its unquenchable nature. Consequently, 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 endmember CaSiO_3 davemaoite under high- P room- T conditions (32–133 GPa at 300 K) using Brillouin spectroscopy with diamond anvil cells (DACs) and moderate- P high- T conditions (7–22 GPa, 300–1700 K) using ultrasonic interferometry coupled with multi-anvil press (MAP) (10,20,21). The room- T sound velocity data agree well across methods and with recent machine-learning-based computational predictions (22), though they are consistently lower than earlier computational results (23). However, high- T experimental sound velocity data show considerable discrepancies (10,21), which 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 endmember CaSiO_3 (27). In 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), due to the slightly larger grain size of their recovered davemaoite, 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 ultra-fine amorphous grains, and possible dependence on the synthesis P - T conditions.

In this study, we synthesized davemaoite samples (endmember 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) (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 endmember CaSiO_3 davemaoite at high P - T conditions in DACs using CaSiO_3 glass and natural near-endmember wollastonite crystals as starting materials (Table S1). Then, these samples were measured for their V_p and V_s at 20–30 GPa and 300 K using Brillouin spectroscopy (Materials and Methods). As shown in Fig. 1, our results align with previous Brillouin spectroscopy or ultrasound interferometry measurements on tetragonal endmember davemaoite (20,21), confirming successful synthesis and reliable experimental procedures. V_p and V_s 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 from Kudo et al. (20), estimated using high- P XRD-derived isothermal bulk modulus and V_s , are slightly higher since they are subject to greater uncertainties. In contrast, Thomson et al. (10) reported systematically lower sound velocities, especially at high- T conditions (Fig. 1), which cannot be explained solely by compositional differences and highlights the need for further high- T studies on endmember CaSiO_3 davemaoite.

Following identical experimental procedures (Materials and Method, 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 300K isotherm in its low- T tetragonal form (Fig. 1). They were found to be 10–20% lower than the V_p and V_s of endmember CaSiO_3 davemaoite (Fig. 1), whereas unit cell volumes, determined through synchrotron powder XRD, were slightly larger (Fig. S1-S2). We performed a joint fit of sound velocities and unit-cell volumes (Table S2-S3) using Eulerian strain equations (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, $K_{S0}' = 4.9(2)$, $G_0' = 1.50(2)$ (Supplementary Text 1). Although davemaoite's unquenchable nature precludes direct measurements of V_0 , our multi-parameter fitting approach including V_p , V_s and unit-cell volume provides robust constraints on V_0 (Supplementary Text 4). Use of different NaCl pressure scales affect K_{S0} and K_{S0}' , but the calculated high- P sound velocities and densities using different parameter sets are similar due to 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 effects of individual substituents.

Compared to endmember davemaoite, K_{S0} and G_0 of Fe,Al,Mg,Ti-bearing davemaoite are 40–50% lower (10,21) (Table S4). This increased compressibility and reduced sound velocities likely result from several factors. Firstly, Al^{3+} substitution introduces oxygen vacancies ($\text{Si}^{4+} \rightarrow \text{Al}^{3+} + \text{vacancy}$) (18), softening the perovskite-structure. Secondly, 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 due to their size mismatch with Ca^{2+} and Si^{4+} , leading to reduced incompressibility. Similar mechanisms have been proposed to explain the observed lower sound velocities and greater compressibility of Fe,Al-bearing bridgmanite compared to endmember MgSiO_3 bridgmanite (18,30).

V_p and V_s of Fe,Al,Mg,Ti-bearing davemaoite were also measured at 400–700 K, 20–45 GPa using resistively heated DACs (Fig. 1). We did not fit the 400–700 K data to obtain the T derivatives of the elastic moduli due to 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 T 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 400–700 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 T derivatives of the elastic moduli and 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 33–113 GPa and 1600–2600 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 endmember CaSiO₃ davemaoite, likely also due to Fe, Al, Mg, and Ti incorporation (Fig. 1). The fitted elastic properties are: $K_{S0} = 138(3)$ 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, $\gamma_0 = 1.42$, $q_0 = 2.65$, $\eta_{S0} = 1.54(14)$ (Supplementary text 1). Compared to its lower- T tetragonal polymorph, the cubic Fe,Al,Mg,Ti-davemaoite exhibits a 6% higher K_{S0} and a 19% higher G_0 , qualitatively consistent with previous studies of endmember CaSiO₃ davemaoite (21,23). For instance, 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 ($\text{Fe}^{3+}/\Sigma\text{Fe}=0.6$ -0.62) used in this study is dominantly ferric (Fig. S6, Materials and Methods). Bridgmanite, the most abundant lower mantle mineral, with 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 endmember CaSiO_3 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/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 to PREM (35; details in Supplementary Text 2). We found that, despite stishovite's ultrahigh velocities (11), MORB is seismically slower than pyrolite, chondritic mantle, 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 mid-mantle seismic scatterers, 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 20–60 vol% MORB (2) (Fig. 4B). Supporting evidence includes: 1) the thickness of most of the observed scatterers (~ 10 km) is consistent with subducted oceanic crust (2); 2) many of them cluster near circum-Pacific subduction zones (36); and 3) the presence of MORB can reduce V_s by up to 7% at ~ 1000 km depth (Fig. 4A). In 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).

LLSVPs show 2–3% V_s reductions and $\sim 1\%$ V_p reductions, along with $\sim 1\%$ positive density anomaly (3,4), requiring the existence of 35–55 vol% MORB in the lowermost lower mantle (Fig. 4C). Assuming a MORB subduction flux of $10 \text{ km}^3/\text{yr}$ into the lower mantle, and that LLSVPs occupy 2% of the entire mantle's volume (37), it would take about 800 Myr for the accumulated MORB material to account for 35–55 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 ambient lower mantle, making it an unlikely component of LLSVPs. Their assumption that Fe,Al,Mg,Ti-bearing davemaoite shares the same elastic properties with endmember CaSiO₃ is challenged by this 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 = 8–13 wt%; Al₂O₃ = 13–17 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 5–30% in V_p and 10–50% in V_s (e.g., 5). MORB alone cannot explain these anomalies but may contribute to LLSVPs. 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% 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. Note, while Fe incorporation is likely to lower the melting temperature, endmember 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 (REEs) (44). If LLSVPs and ULVZs represent regions enriched in davemaoite, 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 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 (REE, Th, U), such as those from the Austral Islands (HIMU) and Tristan-Inaccessible Island (EM1) (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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Software: JSZ, MH, WZ

Visualization: WZ, JSZ, SS

Funding acquisition: JSZ

Writing—original draft: WZ, JSZ

269 Writing—review and editing: JSZ, MH, WS, TK, SC, SS, KA, PQN, DZ

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273 **Supplementary Materials:**

274 Materials and Methods

275 Supplementary Text

276 Figs. S1 to S6

277 Tables S1 to S5

278 References (47-70)

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462

463 **Fig. 1. Sound velocities of tetragonal and cubic davemaoite in this and previous studies.** A.
464 Compressional wave velocity (V_p) of davemaoite. B. Shear wave velocity (V_s) of davemaoite.
465 Best-fit velocity curves for tetragonal and cubic davemaoite are along 300 K and 2000 K isotherms,
466 respectively. All numerical data from this study are listed in Table S3. Vertical error bars represent
467 one standard deviation based on multiple sound velocity measurements on the same sample at each
468 P – T condition. Horizontal error bars for 300–700 K data reflect pressure uncertainties derived
469 from ruby fluorescence measurements before and after each run as well as intrinsic ruby P scale
470 uncertainty. For high-temperature data (> 1600 K), horizontal error bars represent pressure
471 uncertainties bracketed by isobaric and isochoric boundary conditions.

472

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

479

480 **Fig. 3. Mineral proportions in MORB and sound velocities of constituent phases.** A. Phase
481 diagram of MORB in the lower mantle (13). B. Sound velocities of lower mantle minerals along a
482 1600 K adiabatic geotherm (Table S4–5, Supplementary Text S2). NAL phase refers to new
483 aluminum-rich phase, whereas CF phase is the aluminum-rich phase with calcium ferrite structure.

484

485 **Fig. 4. Seismic velocity and density anomalies of MORB compared to other mantle petrologic**
486 **compositions and mineral phases.** A. Seismic velocities of MORB, cubic Fe,Al,Mg,Ti-bearing
487 davemaoite, pyrolitic and chondritic mantle along a 1600 K adiabatic geotherm, as well as MORB
488 along a 1100 K adiabatic geotherm (Table S4-5; Supplementary Text 2), compared with the 1-D
489 seismic reference model PREM (35). B. Seismic velocity and density anomalies induced by
490 MORB at mid-mantle depths (1300 km). C. Seismic velocity and density anomalies induced by

491 MORB at lowermost mantle depths (~2400 km). Velocity differences (dV_p , dV_s) are calculated
492 with respect to PREM