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Supporting Information for

Reduced thermal conductivity of hydrous aluminous silica and calcium ferrite-type phase promote water transportation to Earth's deep mantle

Wen-Pin Hsieh[1,2]*, Takayuki Ishii[3], Frédéric Deschamps[1], Yi-Chi Tsao[1], Jen-Wei Chang[1], and Giacomo Criniti[4]

[1]Institute of Earth Sciences, Academia Sinica, Taipei 11529, Taiwan

[2]Department of Geosciences, National Taiwan University, Taipei 10617, Taiwan

[3]Institute for Planetary Materials, Okayama University, 827 Yamada, Misasa, Tottori 682-0193, Japan

[4]Earth and Planets Laboratory, Carnegie Institution for Science, Washington, DC, USA

Corresponding author: Wen-Pin Hsieh (wphsieh@earth.sinica.edu.tw)

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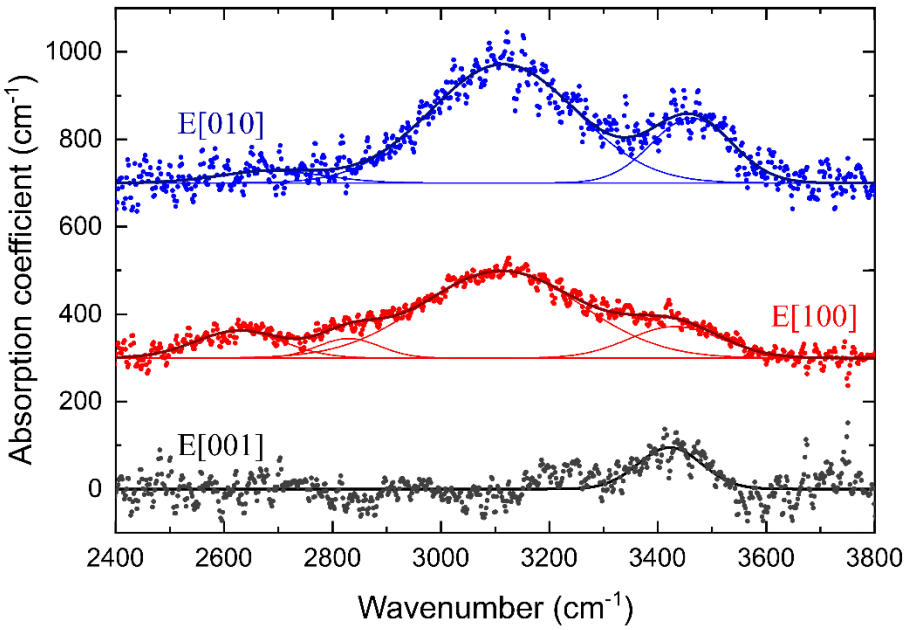
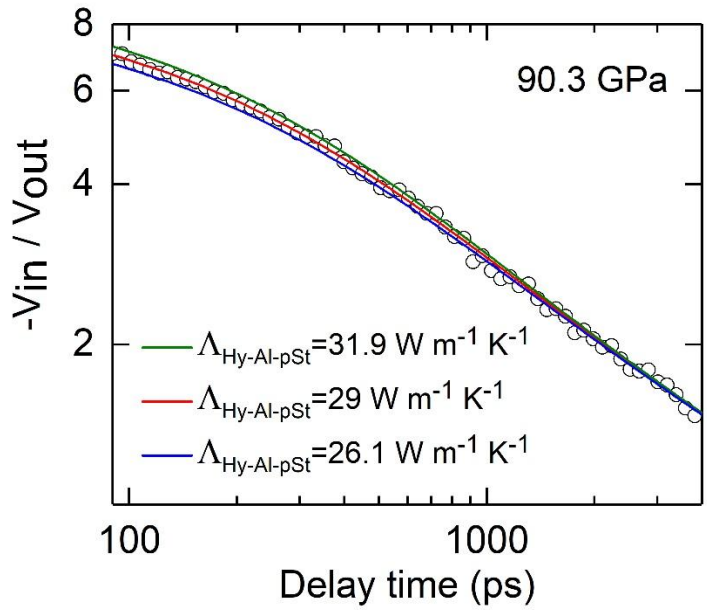
Introduction

There are four figures and one table that show supporting information to the present study. Figure S1 plots the polarized Fourier transform infrared (FT-IR) spectra for hydrous aluminous post-stishovite. Figure S2 shows a set of raw TDTR spectrum fitted by the thermal model. Figure S3 presents analysis of data uncertainty caused by uncertainty of each model parameter. Table S1 lists a set of input parameters used in the thermal model

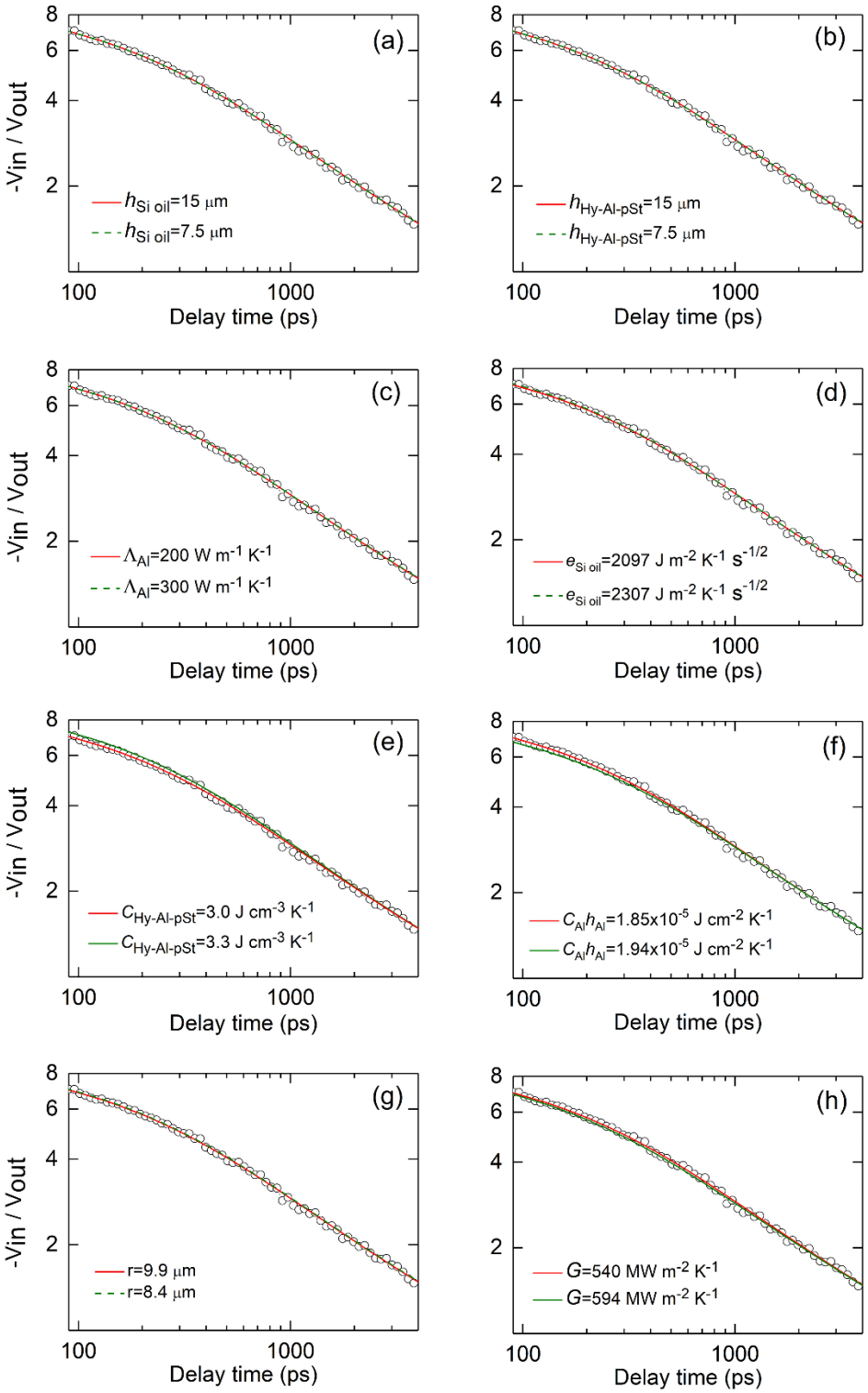
for hydrous aluminous post-stishovite at 90.3 GPa and 300 K. Figure S4 shows the Raman spectrum of hydrous aluminous post-stishovite at high pressures.



Supplementary Figure S1. Example polarized Fourier transform infrared (FT-IR) spectra for hydrous aluminous post-stishovite at ambient conditions oriented along the (010) and (100) crystallographic planes, respectively, as indicated in the top two sub-plots. The spectrum along (001) at the bottom was plotted for comparison.

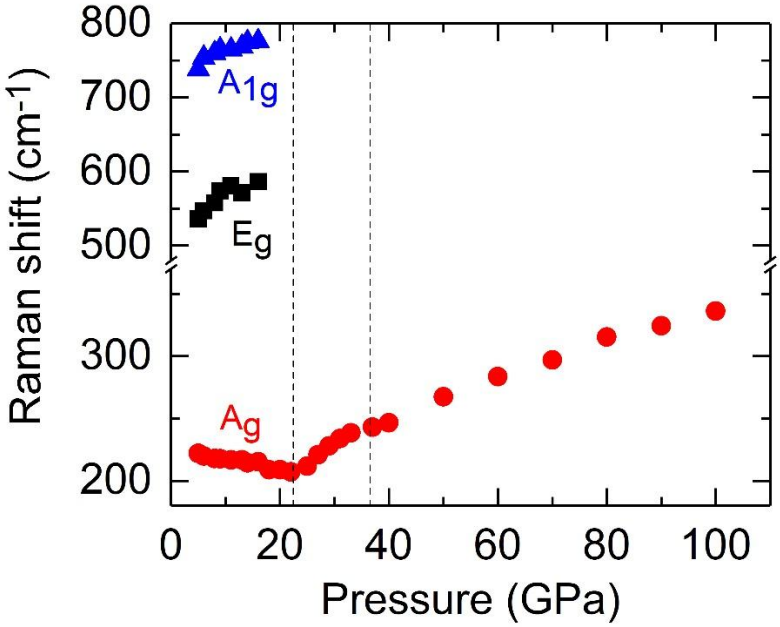


Supplementary Figure S2. Representative TDTR data (open circles) for hydrous aluminous pSt (Hy-Al-pSt) at 90.3 GPa and room temperature fitted by thermal model simulations (color solid curves). The raw TDTR data is best-fitted with $\Lambda_{Hy\text{-}Al\text{-}pSt}$=29 W m^{-1} K^{-1} (red curve) using a set of input parameters listed in Supplementary Table S1. The data for $-V_{in}/V_{out}$ is most sensitive to the sample's thermal conductivity at delay time of few hundred picosecond (ps), see (Cahill & Watanabe, 2004; Zheng et al., 2007) for details. When fitting the data with a $\Lambda_{Hy\text{-}Al\text{-}pSt}$ that is 10% difference (green and blue curves) than the best-fitted $\Lambda_{Hy\text{-}Al\text{-}pSt}$=29 W m^{-1} K^{-1}, the thermal model simulation yields a poor fitting, which indicates that our data analysis is highly sensitive and precise due to our high-quality data.



Supplementary Figure S3. Sensitivity tests of the thermal model simulation to several key input parameters for hydrous aluminous pSt (Hy-Al-pSt) at 90.3 GPa. Here we fix the thermal conductivity of Hy-Al-pSt, $\Lambda_{Hy\text{-}Al\text{-}pSt}$, at 29 W m^{-1} K^{-1} as derived in Supplementary Fig. S2. **(a)** and **(b)** The model simulations remain essentially the same, even if the

thicknesses of the pressure medium silicone oil ($h_{\text{Si oil}}$) and Hy-Al-pSt sample ($h_{\text{Hy-Al-pSt}}$) are changed by 50%, respectively. These indicate that their individual uncertainty does not affect the $\Lambda_{\text{Hy-Al-pSt}}$. (**c)** Again, the model simulation remains the same even if the high thermal conductivity of Al film increases at high pressures, i.e., its uncertainty has no effect on the $\Lambda_{\text{Hy-Al-pSt}}$. (**d)** If there is an example 10% uncertainty from the thermal effusivity of the pressure medium silicone oil, $e=(\Lambda_{\text{Si}}C_{\text{Si}})^{1/2}$, the model simulation is hardly changed, i.e., its uncertainty has very minor effect on the $\Lambda_{\text{Hy-Al-pSt}}$. (**e)** If there is a 10% uncertainty in the volumetric heat capacity of Hy-Al-pSt, $C_{\text{Hy-Al-pSt}}$, the data can be re-fitted by a slightly lower $\Lambda_{\text{Hy-Al-pSt}}$=27 W m^{-1} K^{-1}, i.e., minorly propagating ~7% uncertainty. (**f)** The uncertainty in the heat capacity of Al film per unit area (product of Al's volumetric heat capacity and thickness, $C_{\text{Al}}\,h_{\text{Al}}$, see (Zheng et al., 2007)) is the major uncertainty of our data analysis. If there is an example 5% uncertainty, a slightly higher $\Lambda_{\text{Hy-Al-pSt}}$=31 W m^{-1} K^{-1} (~6.8% change) re-fits well the data. (**g)** A 15% uncertainty in the laser spot size and (**h**) a 10% off in the thermal conductance of Hy-Al-pSt and Al/silicone oil interfaces, G, respectively, does not significantly influence the model simulations. These results indicate that their uncertainties have very little effect on the $\Lambda_{\text{Hy-Al-pSt}}$.



Supplementary Figure S4. Raman shift of the Hy-Al-pSt as a function of pressure. The characteristic A_g mode at ~230 cm^{-1} softens with initial compression, while its pressure slope becomes small around 22 GPa, after which the frequency increases with higher pressure.

Supplementary Table S1. Input parameters in the thermal model for Hy-Al-pSt at 90.3 GPa and 300 K in TDTR measurements

P (GPa)	$C_{Hy\text{-}Al\text{-}pSt}$ (J cm^{-3} K^{-1})	C_{Al} (J cm^{-3} K^{-1})	h_{Al} (nm)*	$e=(\Lambda_{Si}C_{Si})^{1/2}$ (J m^{-2} K^{-1} $s^{-1/2}$)	r (μm)	$h_{Hy\text{-}Al\text{-}pSt/Si\ oil}$ (μm)	Λ_{Al} (W m^{-1} K^{-1})	G (MW m^{-2} K^{-1})
90.3	3	2.683	69.0	2097	9.9	15/15	200	540

*In this experimental run, the Al thickness at ambient pressure is 88.2 nm.
$C_{Hy\text{-}Al\text{-}pSt}$: Hy-Al-pSt heat capacity, C_{Al}: Al heat capacity, h_{Al}: Al thickness, e: silicone oil thermal effusivity, r: laser spot size, $h_{Hy\text{-}Al\text{-}pSt}$: Hy-Al-pSt thickness, $h_{Si\ oil}$: silicone oil thickness, Λ_{Al}: Al thermal conductivity, G: thermal conductance of Al/Hy-Al-pSt and Al/silicone oil interfaces.

Supplementary References

Cahill, D. G., & Watanabe, F. (2004). Thermal conductivity of isotopically pure and Ge-doped Si epitaxial layers from 300 to 550 K. *Phys. Rev. B*, *70*(23), 235322. https://doi.org/10.1103/PhysRevB.70.235322

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