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34. The first and second acid dissociation constants of sulfurous acid are $10^{-7.2}$ and $10^{-1.85}$, respectively, whereas those of carbonic acid are $10^{-10.3}$ and $10^{-6.3}$.
35. Pre-industrial atmospheric pSO_2/pCO_2 is ~3×10^{-7}. However, sulfite minerals do not precipitate because Earth's ocean is not in equilibrium with these atmospheric concentrations. As soon as it dissolves, sulfite is either rapidly oxidized in the aqueous phase to sulfate or reduced by bacteria to sulfide.
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Coupled ^{142}Nd-^{143}Nd Isotopic Evidence for Hadean Mantle Dynamics

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The oldest rocks—3.85 billion years old—from southwest Greenland have coupled neodymium-142 excesses (from decay of now-extinct samarium-146; half-life, 103 million years) and neodymium-143 excesses (from decay of samarium-147; half-life, 106 billion years), relative to chondritic meteorites, that directly date the formation of chemically distinct silicate reservoirs in the first 30 million to 75 million years of Earth history. The differences in ^{142}Nd signatures of coeval rocks from the two most extensive crustal relicts more than 3.6 billion years old, in Western Australia and southwest Greenland, reveal early-formed large-scale chemical heterogeneities in Earth's mantle that persisted for at least the first billion years of Earth history. Temporal variations in ^{142}Nd signatures track the subsequent incomplete remixing of very-early-formed mantle chemical domains.

Isotope data for short-lived decay schemes such as the ^{146}Sm-^{142}Nd system [half-life ($T_{1/2}$), 103 million years (My)] obtained from samples of the Moon, Mars, and meteorites are revealing the complexities of early planetary differentiation that occurred soon after accretion [e.g., (*1–4*)]. Attempts to demonstrate ^{142}Nd variations in Earth were initiated in the 1990s (*5*), with recent measurements of some ancient rocks [dating from >3.6 billion years ago (Ga)] (*6–10*) now firmly establishing variable ^{142}Nd/^{144}Nd ratios in Earth relative to modern terrestrial compositions. This is important because detectable ^{142}Nd isotopic variations can only be generated from Sm/Nd fractionation during the largely unknown first ~300 My of Earth history, while ^{146}Sm is still actively decaying. Previous studies focused largely on the ~3.7- to 3.8-billion-year (Gy)–old regions of the Isua supracrustal belt of West Greenland, representing largely one, albeit important, spatial and temporal point in Earth evolution. Interpreting the large range of reported ^{142}Nd/^{144}Nd ratios [0 to +17 parts per million (ppm) higher than modern terrestrial compositions] in terms of early planetary processes is complicated because many of the analyzed rocks were either metasedimentary mixtures of eroded terranes (*7–9*) or metabasalts (*6*, *9*, *10*) from areas that experienced widespread secondary chemical alteration [e.g., (*11*, *12*)].

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Tantalizing hints of Hadean Era (>4.0 Ga) Earth dynamics come from the recent recognition that all crust and upper mantle rocks today have a ^{142}Nd/^{144}Nd excess of ~20 ppm compared with primitive chondritic meteorites (*2*, *13*, *14*), which were the building blocks of Earth. This requires not only that chemically distinct domains with high and low Sm/Nd, evolving to high and low ^{142}Nd, respectively, formed during or soon after accretion but also that these domains must have persisted to the present in order to account for the continued isotopic offset between chondrites and modern terrestrial rocks. Additionally, the more extreme ^{142}Nd excesses measured in some Archean rocks (*6*, *7*, *9*, *10*) require the early existence of an older, or more severely depleted (higher Sm/Nd) mantle, whose extent and longevity is unknown. To track the origin, distribution, and interaction of these global chemical domains requires precise ^{142}Nd data for ancient rocks with a range of ages and from a variety of localities.

Here, we present high-precision ^{142}Nd data (Table 1) (*15*) combined with ^{143}Nd data from samples of the two most aerially extensive >3.6-Gy-old terranes: the 3000 km^2 Itsaq Complex (*16*) of southern West Greenland, of which the Isua supracrustal belt is one component, and the Narryer Gneiss Complex of the Yilgarn craton, Western Australia. In contrast to previous studies, the emphasis is on analysis of the oldest tonalites, a juvenile granitic rock type typically representing the earliest formed continental crust in a region and from which direct age information in the form of U-Pb zircon ages can be obtained. Archean tonalites are melts of young oceanic crust [e.g., *17*)], derived from the upper mantle. The intermediate basalt stage is likely short compared with the time scale of ^{147}Sm decay and with Sm/Nd similar to that of the mantle source.

The Itsaq samples span a 210-My age range (3.64 to 3.85 Gy old) and include crystalline rocks from newly recognized localities of homogeneous 3.85-Gy-old tonalites (*18*) and >3.85-Gy-old mafic rocks (*15*). These samples are some of the oldest terrestrial rocks yet discovered. The two 3.73-Gy-old Narryer Gneiss Complex tonalitic gneisses are the oldest rocks from the Australian continent (*19*). All rocks are from our field collections and represent the most geologically pristine materials, with well-defined crystallization ages having minimal secondary chemical alteration (table S1) (*15*).

Homogenized powdered samples weighing from 0.1 to 0.3 g were dissolved and processed to isolate >500 ng Nd. The ^{142}Nd/^{144}Nd isotopic compositions were measured as Nd^+ on a Triton thermal ionization mass spectrometer using a multidynamic data collection scheme (*9*). Measurements of a standard Nd solution run interspersed with the samples yielded an external reproducibility (2 SD) of ±3.5 ppm (fig. S1) (in-run precisions were from 1.0 to 2.5 ppm, 2 SE). Replicate analyses of 14 modern rocks yielded the same external precision as the standard data (fig. S2), which demonstrates the validity of this precision for chemically processed samples. The ^{147}Sm-^{143}Nd data were obtained on separate powder samples using standard isotope-dilution methods (*15*).

Fifteen samples from the Itsaq Complex all have well-resolved ^{142}Nd excesses of 9 to 20 ppm relative to the modern terrestrial reference composition (Table 1). The six basalts with ages of 3.8 Gy and >3.85 Gy have slightly higher, and more variable, ^{142}Nd than the similar age tonalites. In contrast, the four oldest (~3.85 Gy) tonalites collected from four separate localities have a narrow range of compositions, with mean ^{142}Nd excess of +15.3 ± 1.2 ppm (2 SD), and the three youngest (3.64 Gy) samples have a ^{142}Nd excess = +12.0 ± 0.9 ppm (2 SD).

High ^{142}Nd relative to modern rocks now appears to be a pervasive feature in the vast Archean terranes of southwest Greenland. The regional extent of this signature was likely much greater in the early Archean, as the analyzed rocks from the northern and southern parts of the Itsaq Complex, now >150 km distant, represent at least two different terranes, each with a distinct Eoarchean geologic history, that were later juxtaposed (*20*). In contrast to all of the Greenland samples, the two granitic samples from the Yilgarn craton have identical ^{142}Nd ≈ +5 ppm. The lack of ^{142}Nd excesses in a few previously analyzed Itsaq rocks (*8*, *10*) raises the question of how much confidence to place in these two samples representing the Yilgarn craton. We emphasize first that all granitic rocks (not metabasalts or metasediments) from the diverse Archean terranes of West Greenland give consistent results, with 100% of the analyzed >3.6-Gy-old granitic rocks (13 samples total) (Table 1) (*9*) having well-resolved ^{142}Nd excesses. Second, the two Western Australian samples were selected specifically to be equivalent to the Greenland samples in having similar juvenile tonalitic compositions, reflecting the earliest known granitic rocks in each terrane, with a similar degree of good preservation, such that they can be expected to yield reliable isotopic records.

Although this demonstration of global heterogeneity is powerful evidence for early differentiation on Earth, ^{142}Nd variations alone cannot be modeled uniquely in terms of time or extent of mantle depletion. However, concordant data from the two Sm-Nd decay schemes measured from the same sample can be combined to yield both the formation age of the mantle reservoir and the degree of Sm/Nd fractionation of this mantle (*5*), which is indicative of the extent of chemical differentiation. The calculated differentiation ages are most accurate using data from the oldest sam-

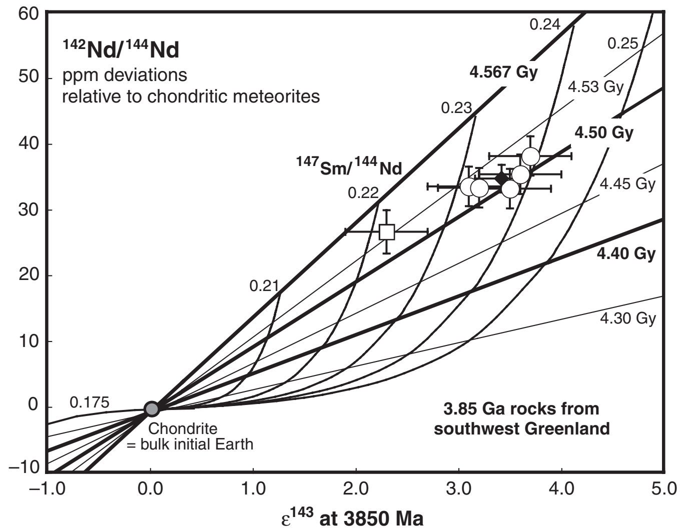

Fig. 1. Two-stage Sm-Nd evolution model (*5*) for the terrestrial mantle starting at the time of solar system origin (T_0 = 4567 Ma) (*30*), with a silicate Earth having a chondritic Sm/Nd ratio and Nd isotopic compositions. The second stage starts at the formation of a Hadean mantle source with high Sm/Nd. This mantle continues to evolve and is ultimately sampled by partial melting to form the ~3.85-Gy-old rocks studied here, which carry and preserve the elevated ^{142}Nd and ε^{143}Nd signatures. The straight lines represent the loci of equal ages for the formation of variable Sm/Nd mantle sources. The curved lines are the loci of increasing ^{147}Sm/^{144}Nd ratios representing varying levels of mantle differentiation. The combined ^{143}Nd and ^{142}Nd data from the oldest (3.85 Gy old), least altered tonalites and amphibolites (circles, with mean indicated by solid diamond) are self-consistent and indicate formation of a chemically fractionated, high Sm/Nd mantle within the first 30 My to 75 My of Earth history. Bulk silicate Earth compositions (*21*); decay equations (*15*); initial ^{146}Sm/^{144}Sm = 0.0075 ± .0025 (*31*). Decay constants: $\lambda_{147} = 6.54 \times 10^{-12}$ yr^{-1} and $\lambda_{146} = 6.74 \times 10^{-9}$ yr^{-1}. Error bars are ± 2 SD external reproducibility (Table 1).

Table 1. Summary of ^{142}Nd and ^{143}Nd isotopic data from two Eoarchean terranes. ^{142}Nd data are presented as the ppm difference from the terrestrial reference, calculated using the average value of terrestrial standards measured during the course of this study. Data sources for ε^{143}Nd(0) (*21*) are given in table S1. The errors on ε^{143}Nd(*t*) are ≤ ±0.5 epsilon units. The ages of the mafic rocks are determined from U-Pb zircon dating of associated cross-cutting felsic rocks and are thus minimums. Complete data sets, analytical methods, and geologic locality and age information are in (*15*).

Sample No.	Age (My)	Lithology	^{142}Nd/^{144}Nd	±2σ	^{142}Nd (ppm)	ε^{143}Nd(0)	ε^{143}Nd(*t*)
			Itsaq complex, West Greenland				
Islands near Nuuk							
G01/113	3849 ± 6	Tonalite	1.1418565	4.2×10^{-6}	15.8	−45.06	+3.1
G93/07	3852 ± 12	Tonalite	1.1418563	2.2×10^{-6}	15.6	−37.73	+3.2
G99/22	3862 ± 16	Tonalite	1.1418561	3.0×10^{-6}	15.5	−36.93	+3.5
G01/36	3849 ± 6	Tonalite	1.1418547	2.8×10^{-6}	14.2		
G97/112	3640 ± 11	Ferrodiorite	1.1418520	2.8×10^{-6}	11.9		+1.7
G97/112	Replicate		1.1418517	2.5×10^{-6}	11.6		
G97/111	3642 ± 3	Augen gneiss	1.1418527	2.4×10^{-6}	12.5		+1.7
JG03/11	>3850	Amphibolite	1.1418586	2.2×10^{-6}	17.6	12.02	+3.6
JG03/12	>3850	Amphibolite	1.1418618	1.0×10^{-6}	20.4	1.68	+3.7
JG03/36	>3850	Amphibolite	1.1418486	3.0×10^{-6}	8.9	−31.52	+2.3
G01/91	>3850	Metagabbro	1.1418509	3.0×10^{-6}	10.9		
Isua supracrustal belt and vicinity							
G06/3.7	3698 ± 8	Tonalite	1.1418544	2.6×10^{-6}	14.0	−51.85	+1.2
JG03/52	>3803 ± 3	Pillow basalt	1.1418526	2.6×10^{-6}	12.4	−6.73	−0.2
JG03/48	>3803 ± 3	Pillow basalt	1.1418605	2.9×10^{-6}	19.3	−31.70	+4.2
G97/98	3795 ± 3	Tonalite	1.1418578	3.1×10^{-6}	16.9	−45.86	+2.1
			Yilgarn craton, Western Australia				
88/28	3731 ± 4	Tonalite	1.1418444	3.2×10^{-6}	5.2	−41.47	+1.7
88/173	3730 ± 5	Tonalite	1.1418438	3.0×10^{-6}	4.6	−48.88	+1.8
Nd standard average (this study); 2σ external precision							
n = 13			1.1418385	4.0×10^{-6}	0.0		

ples, in this case the ~3.85-Gy-old samples from Greenland, because these are less likely to contain mixed-age crustal components. The ^{146}Sm-^{142}Nd scheme requires no corrections for in situ decay in Archean rocks, eliminating a potential source of error, and there is no process that can generate positive ^{142}Nd variations after ^{146}Sm has largely decayed by ~ 4.2 Ga. Alteration or open-system behavior can only move ^{142}Nd signatures toward modern compositions. The samples analyzed here have a wide range of present-day ε^{143}Nd = +12 to –52 (*21*), which after corrections for in situ decay yield a narrow range of initial ε^{143}Nd (Table 1).

The tonalites and mafic samples indicate similar ages and, assuming the five samples with the highest ^{142}Nd represent the same mantle source, they define a mean mantle differentiation age of 4.51 ± 0.02 Gy (1 SD), which suggests that the Hadean high Sm/Nd mantle formed within the first 35 to 75 My of Earth history (Fig. 1). This is similar to the time of metallic core formation on Earth obtained from the ^{182}Hf-^{182}Hf chronometer ($T_{1/2}$ = 9 My) (*22*). Early massive melting of Earth is predicted in many accretion scenarios [e.g., (*23*)], and these coupled ^{142}Nd -^{143}Nd results provide a temporal linkage between formation of major silicate reservoirs and early catastrophic events during planet formation (*10*). Such differentiations may point to magma ocean scenarios [e.g., (*24*)], whereby distinct chemical domains formed by crystal fractionation in a molten Earth rather than by progressive continental crust extraction over time. The ^{147}Sm/^{144}Nd of the Hadean mantle was ≈0.236 (Fig. 1), which is 20% higher than the chondritic (bulk Earth) value of 0.1966 but similar to compositions calculated for lunar highlands crustal rocks (*25*). In contrast to the tonalite suite, ^{142}Nd -^{143}Nd data from 3.7- to 3.8-Gy-old Isua metabasalts show large variations (*6*, *9*, *10*), with many samples falling outside of all possible mantle isochrons generated using two-stage models (fig. S3). This reflects a more complex petrogenesis for these samples [see (*11*, *12*)].

The differences between the contemporaneous Greenland and Australian Eoarchean terranes is highlighted by the coeval ~3.7-Gy-old samples from both regions, with the Narryer tonalites having markedly lower excesses of ^{142}Nd. These contrasting ^{142}Nd signatures demonstrate fundamental, very-early-formed chemical heterogeneities in the terrestrial mantle. Two general scenarios to explain these observations are possible (Fig. 2). The first is formation of a global mantle with uniformly high Sm/Nd [termed here Itsaq-DM (Depleted Mantle)] within ~50 million years. This mantle was transient but survived locally until at least 3.6 Ga, that is, long enough to generate the youngest Archean crustal rocks with high ^{142}Nd in southwest Greenland. In other regions, Itsaq-DM partially remixed with enriched low Sm/Nd, low ^{142}Nd-ε^{143}Nd domains, which existed as a complementary reservoir, or with undifferentiated mantle, to form the mantle source that produced the Narryer Complex gneisses. The second scenario (Fig. 2B) that will satisfy the ^{142}Nd constraints is that two (or more) mantle domains with variable Sm/Nd formed in the first ~50 My of Earth history and retained their distinct identities until at least 3.7 Ga.

What is the fate of the highly depleted Hadean mantle domain? Tracking of ^{142}Nd compositions in West Greenland samples through time shows an apparent decrease in the most positive ^{142}Nd signatures with younger sample crystallization ages (Fig. 3). In contrast, the mantle sampled by the Eoarchean Narryer Complex gneisses and in other somewhat younger Archean regions [i.e., Barberton, South Africa (*9*)] shows minor, or no, ^{142}Nd excess compared with the modern mantle. The trend of decreasing ^{142}Nd in the Itsaq Complex suggests that even the compositions of the oldest samples from 3.85 Ga may already reflect partial destruction of a highly fractionated Hadean mantle domain that formed >4.5 Ga and that the original Sm/Nd may have been even higher, perhaps as

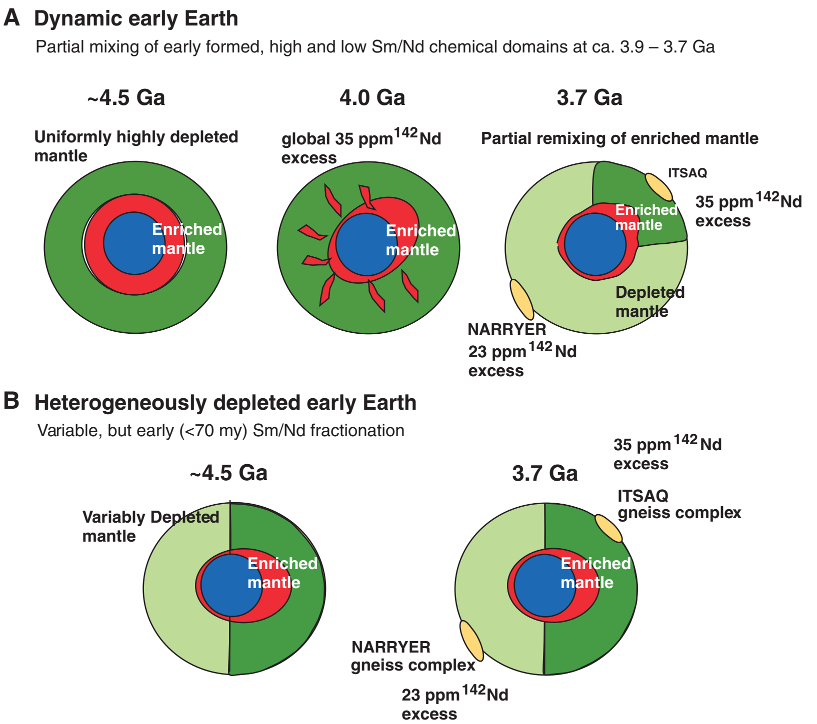

Fig. 2. Two scenarios to account for the distinctive ^{142}Nd signatures of Archean rocks from coeval early crustal terranes. (**A**) Formation of uniform mantle (dark green) with high Sm/Nd early in Earth's history. Most, but not all, of this mantle is subsequently remixed with complementary low Sm/Nd material (red), depicted here as residing in the deep mantle. The Itsaq Complex rocks, southwest Greenland, sample a relict of the early high Sm/Nd domain. The Narryer Gneiss Complex rocks, Western Australia, sample a mantle source with more modest Sm/Nd. (**B**) Early-formed heterogeneous mantle. These chemically distinct mantle source regions persisted for at least 1 Gy, to be sampled by the formation of 3.6- to 3.9-Gy-old Archean rocks in the two regions.

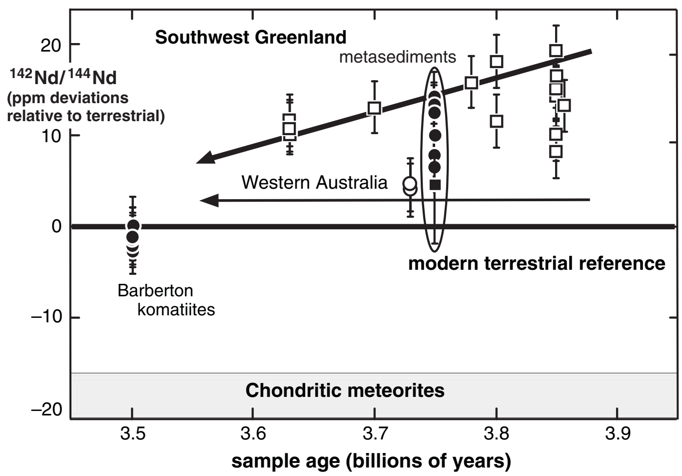

Fig. 3. ^{142}Nd excess (ppm) relative to modern terrestrial compositions as a function of rock crystallization age. Open symbols (this study), with error bars ±3.5 ppm (2 SD). Filled circles (*9*); filled squares (*8*), with errors as reported in each study. The decreasing ^{142}Nd through time suggests progressive remixing of early-formed, highly fractionated mantle sources. The range of data apparent in the mixed provenance metasediments may reflect input from various Archean ^{142}Nd sources (*9*), further supporting the existence of mantle chemical heterogeneity.

extreme as some portions of the Martian mantle [e.g., (*26*)]. Although the Itsaq-DM seems to have been obliterated, or at least not sampled at present, the source mantle of the Narryer Complex crust persists to the present. These observations require an explanation for the differing behavior of the two depleted mantle components, the Narryer-DM able to retain its identity, despite ongoing mantle dynamics for >4.5 Gy, as compared with the transient Itsaq-DM. If all or much of the mantle was initially as highly fractionated as Itsaq-DM, then why was this mantle only partially, but apparently homogeneously, remixed such that all modern terrestrial rocks yield precisely the same ^{142}Nd, distinct from chondrites? The difference supports models of "hidden reservoirs" where part of the complementary, low Sm/Nd domain is locked in a region of Earth, where it is both never sampled at the surface [e.g., (*27–29*)] and isolated from remixing. More speculative suggestions are that part of the low Sm/Nd component may have been lost from Earth during accretion, with the missing material accounting for the present-day high ^{142}Nd, or that Earth accreted with a nonchondritic Sm/Nd ratio. In contrast, the Itsaq-DM mantle source persisted for at least a billion years after its formation, as recorded by compositions of 3.6-Gy-old Greenland samples, but was able to communicate with other less fractionated mantle reservoirs and eventually lost its distinct signature through remixing.

An enduring tenet of geology is that Earth started from a well-mixed homogeneous body and evolved progressively over geologic time to a more differentiated chemical state through observable processes such as plate tectonics and continental crust formation. The ^{142}Nd data presented here, however, provide strong evidence that terrestrial planets such as Earth were affected by non-uniformitarian processes early in their histories, resulting in locally extreme chemical differentiation. Furthermore, some of the chemical effects of these events appear to persist in silicate domains to the present day. Thus, an emerging challenge for understanding the Earth system is determining the relative roles of early planetary processes versus progressive differentiation in shaping Earth's chemical architecture.

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Supporting Online Material

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Materials and Methods
Figs. S1 to S3
Tables S1 to S3
References

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High-Pressure Creep of Serpentine, Interseismic Deformation, and Initiation of Subduction

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The supposed low viscosity of serpentine may strongly influence subduction-zone dynamics at all time scales, but until now its role could not be quantified because measurements relevant to intermediate-depth settings were lacking. Deformation experiments on the serpentine antigorite at high pressures and temperatures (1 to 4 gigapascals, 200° to 500°C) showed that the viscosity of serpentine is much lower than that of the major mantle-forming minerals. Regardless of the temperature, low-viscosity serpentinized mantle at the slab surface can localize deformation, impede stress buildup, and limit the downdip propagation of large earthquakes at subduction zones. Antigorite enables viscous relaxation with characteristic times comparable to those of long-term postseismic deformations after large earthquakes and slow earthquakes. Antigorite viscosity is sufficiently low to make serpentinized faults in the oceanic lithosphere a site for subduction initiation.

Subduction zones, in which slabs of oceanic lithosphere sink into the mantle, are active zones where frequent large earthquakes cause considerable human and material damage. Such events are triggered by stress buildup or strain localization, the understanding of which relies on identifying the materials involved and their rheology. On top of slabs of many subduction zones, a layer with low seismic velocity and high Poisson ratio (>0.29) is interpreted as extensively serpentinized mantle material (*1*, *2*), and may accommodate most of the deformation at the slab/mantle wedge interface. Serpentinites form by peridotite hydration either during hydrothermal alteration of the oceanic lithosphere before subduction or by percolation of the fluids released by mineral dehydration within the downgoing slab through the overlying mantle wedge (*3*). The high-pressure variety of serpentine, antigorite, can remain stable down to ~180 km depth in cold subduction zones (*4*). Serpentinites are highly deformed as compared to other exhumed materials in paleosubduction zones (*5*), which points to their crucial mechanical role. The expected low strength or viscosity of serpentinite

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