

Electrical Conductivity of Olivine at High Pressure and Under Controlled Oxygen Fugacity

A. DUBA, H. C. HEARD, AND R. N. SCHOCK

Lawrence Livermore Laboratory, University of California, Livermore, California 94550

Electrical conductivity σ in the [100] direction has been determined for the Red Sea olivine (Fo 91) to 1440°C and 8 kbar in argon. No systematic variation of σ with pressure was observed. The effect of an 8-kbar variation in pressure over the 1270°–1440°C range is equivalent to a temperature uncertainty of $\pm 5^\circ\text{C}$. We have also determined σ on the same sample up to 1660°C with controlled oxygen fugacity f_{O_2} at 1 bar of total pressure. By using published σ -depth profiles and assuming olivine as the major phase in the earth's upper mantle with $f_{\text{O}_2} = 10^{-6}$ – 10^{-8} bar, temperatures of the upper mantle are calculated as a function of depth. The temperature uncertainty due to possible pressure effects is 2–5 times smaller than that resulting from the ambiguity in published σ -depth profiles.

It is now generally accepted that the major phase in the earth's upper mantle is olivine with an approximate composition $\text{Mg}_{1.8}\text{Fe}_{0.2}\text{SiO}_4$ [Fujisawa, 1968]. Published values of electrical conductivity σ of olivine of this composition (either single crystal or polycrystalline) at high temperatures show very poor agreement [Duba and Lilley, 1972]. In many instances the difference can be attributed either to trace cation impurities or to different $\text{Fe}^{3+}/\text{Fe}^{2+}$ ratios [Duba, 1972]. Calculations of geothermal profiles are often based upon the combination of σ values derived from magnetic field measurements with laboratory σ data. Since the oxygen fugacity f_{O_2} with which olivine is in equilibrium has a significant effect on σ [Duba and Nicholls, 1973], such calculations require laboratory data obtained under controlled f_{O_2} .

The pressure dependence of σ must also be considered in estimating temperature-depth profiles for the earth's upper mantle. Previous attempts to measure the effects of pressure on σ of olivine have yielded ambiguous results and with the exception of the work of Hughes [1955], Schober [1971], and Duba [1972] have been restricted to temperatures less than 1000°C. For polycrystalline samples prepared from powders it is difficult to separate intrinsic pressure effects from changes in sample dimensions due to compaction [Bradley *et al.*, 1964; Hamilton, 1965]. The possibility of slight oxidation of the olivine during σ measurement, as well as thermocouple deterioration [Duba, 1972], and uncertainty due to the high leakage conductance of the sample assembly [Hughes, 1955] make the pressure derivatives obtained from those single-crystal measurements suspect.

In this paper we report σ values measured over a wide range of conditions on a single sample of olivine from the Red Sea area (St. John's Island). This olivine was selected because of its composition ($\text{Mg}_{1.81}\text{Fe}_{0.19}\text{SiO}_4$), which is near that assumed for olivine in the mantle, and its low $\text{Fe}^{3+}/\text{Fe}^{2+}$ ratio [Duba *et al.*, 1973]. Measurements were made up to 1440°C and 8 kbar of argon [Duba *et al.*, 1972] and to 1660°C under controlled f_{O_2} (1 bar total pressure). Results from this study are compared with available single-crystal data. Finally, by assuming an f_{O_2} variation with depth and using available σ -depth profiles, we construct temperature-depth profiles for the upper mantle.

EXPERIMENTAL PROCEDURE AND RESULTS

The Red Sea olivine (Fo 91) sample was cut normal to the *a* axis and polished, and Pt electrodes were deposited under vacuum as previously described [Duba, 1972]. For the high-pressure measurements the experimental procedure of Duba [1972] was modified by replacing the Cr-Al thermocouples with Pt-Pt 10% Rh thermocouples and by constructing a filter [Duba *et al.*, 1972] to remove most of the foreign material that plagued earlier experiments. Any hydrocarbons that passed the filtration system were deposited as a thin film in the secondary heater only and did not affect σ measurements, which were performed in the primary heater. The partial pressure of oxygen during the high-pressure experiments is not known.

Electrical conductivity was calculated from conductance measured using either the capacitance bridge or the resistance network described by Duba [1972]. Measurements of conductance were made first at 8.0 kbar under conditions of both increasing and decreasing temperature. Gas was then released from the pressure vessel for the 5.0- and 2.5-kbar measurement cycles, followed by repressurization to 8.0 kbar in order to check reproducibility. Constant pressure was maintained by releasing gas from the pressure vessel during the temperature increase portion of the cycle (600°–1440°C) and by pumping during the temperature decrease portion of the cycle. Temperatures were corrected for the effect of pressure on thermocouple emf by using the values of Getting and Kennedy [1970]. The temperature corrections for the pressure-temperature range of this study do not differ significantly from those indicated by Hanneman *et al.* [1971] and Lazarus *et al.* [1971].

Figures 1a–1c include the σ data collected at the three pressures at temperatures above 560°C. Below this temperature, leakage conductance in the sample assembly affected the measurement. Inspection indicates a negligible pressure effect over the range 2.5–8.0 kbar. The scatter for any given pressure encompasses the scatter for all other pressures. However, the σ obtained on temperature decrease at each pressure appears to be consistently higher than that measured for temperature increase. The reasons for this slight hysteresis are possibly related to a reaction between olivine and the high-pressure atmosphere. The decreased conductivity during the second 8-kbar cycle (Figure 1a) may be the result of the slightly different atmosphere (i.e., a change in f_{O_2}) in the vessel

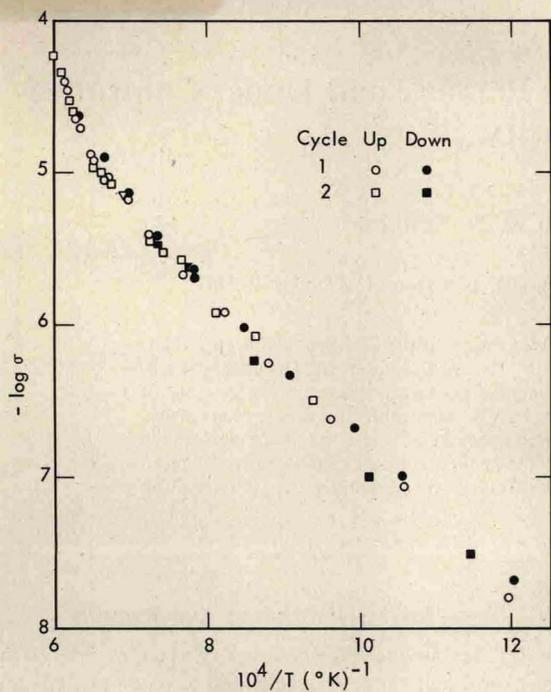


Fig. 1a.

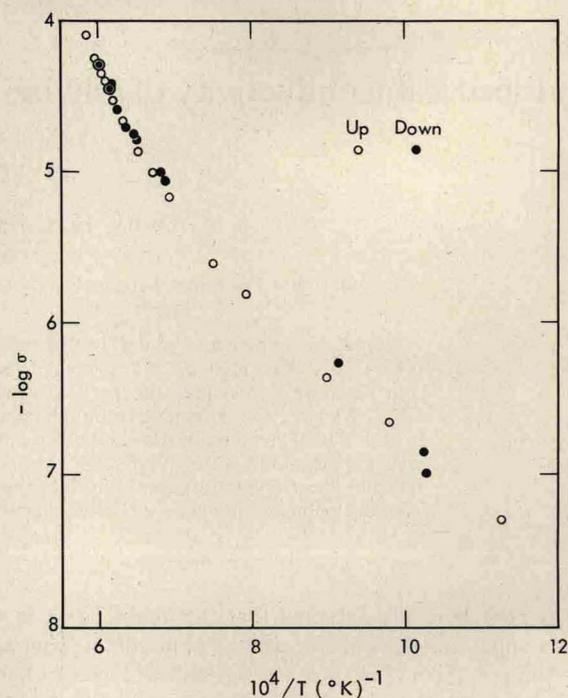


Fig. 1c.

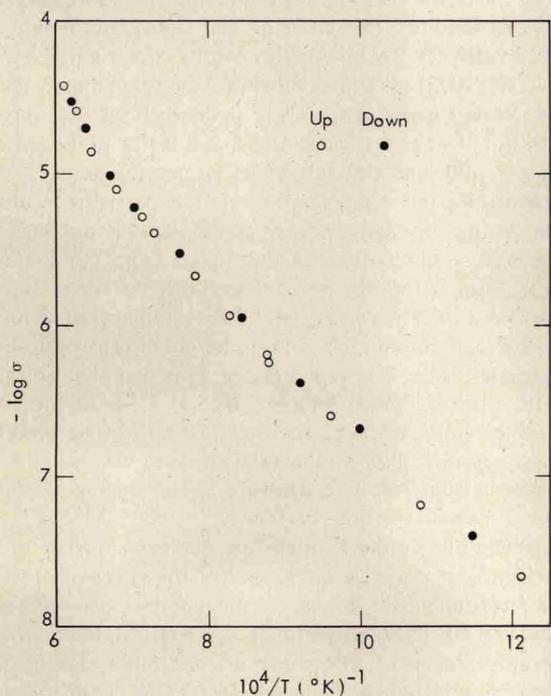


Fig. 1b.

Fig. 1. Conductivity ($\Omega^{-1} \text{cm}^{-1}$) of Red Sea olivine [100] to 1440°C at various pressures: (a) 8.0 kbar, cycle 1, 2 (second cycle data taken after 5.0- and 2.5-kbar cycles), (b) 5.0 kbar, and (c) 2.5 kbar.

after repressurization or some change in the sample. After the four cycles were completed and the sample was removed from the pressure vessel it was observed to have one central fracture. It is not known when the sample was fractured or how the fracture affected σ . All of the data in Figures 1a-1c are replotted in Figure 2 along with a linear regression fit of the data to (1) in the high- and low-temperature regions. Analyses of each data set shown in Figures 1a-1c are summarized in Table 1.

Six months later we measured the σ of the same sample under controlled f_{O_2} using mixtures of H_2 and CO_2 as described by *Duba and Nicholls* [1973]. Below 1200°C, σ was initially as much as 10 times greater than that determined at high pressure. However, σ decreased rapidly with time and approached the high-pressure values previously determined at temperatures above 1200°C (Figure 3). We infer that this low-temperature behavior is the result of adsorbed water in the crack. Similar observations have been reported for cracked pyroxenes [*Duba et al.*, 1974]. After the sample was allowed to remain at $1252^\circ \pm 10^\circ\text{C}$ for about 15 hours at $f_{\text{O}_2} = 10^{-8.4}$ bar (referenced to 1200°C), σ had decreased about one-half order of magnitude to point A indicated in Figure 3. When no further change in σ was noted at $1244^\circ \pm 3^\circ\text{C}$ for 1.5 hours, data were collected at the lower temperatures as shown. Heating and cooling rates during cycling were 50°-100°C per hour.

On temperature increase, f_{O_2} was readjusted to 10^{-8} bar (1200°C). This mixture of CO_2 and H_2 follows a T - f path (indicated at the top of Figure 3) close to the quartz-fayalite-magnetite equilibrium curve and within the olivine stability field [*Duba and Nicholls*, 1973]. After 30 hours at $1060^\circ \pm 10^\circ\text{C}$ a small decrease in σ was observed as indicated in Figure 3 (point B). Following σ measurement to 1475°C, equilibration of the sample with the experimental atmosphere was indicated by the absence of any change in σ after 15 hours at approximately 1400°C. During the next 6 hours, σ was measured as the temperature was increased from 1400°C to 1660°C and then decreased to 1440°C. The data on the temperature decrease portion indicate an irreversible increase in the conductivity of the sample below 1500°C. Repeat measurements over an identical cycle during the succeeding 5 hours yielded a curve displaced to higher σ by about 0.1 order of magnitude from the previous temperature increase portion and practically identical with the decrease portion. After 15 hours at about 1400°C, no change in σ was observed. Repeat of the above temperature cycles a third time yielded conduc-

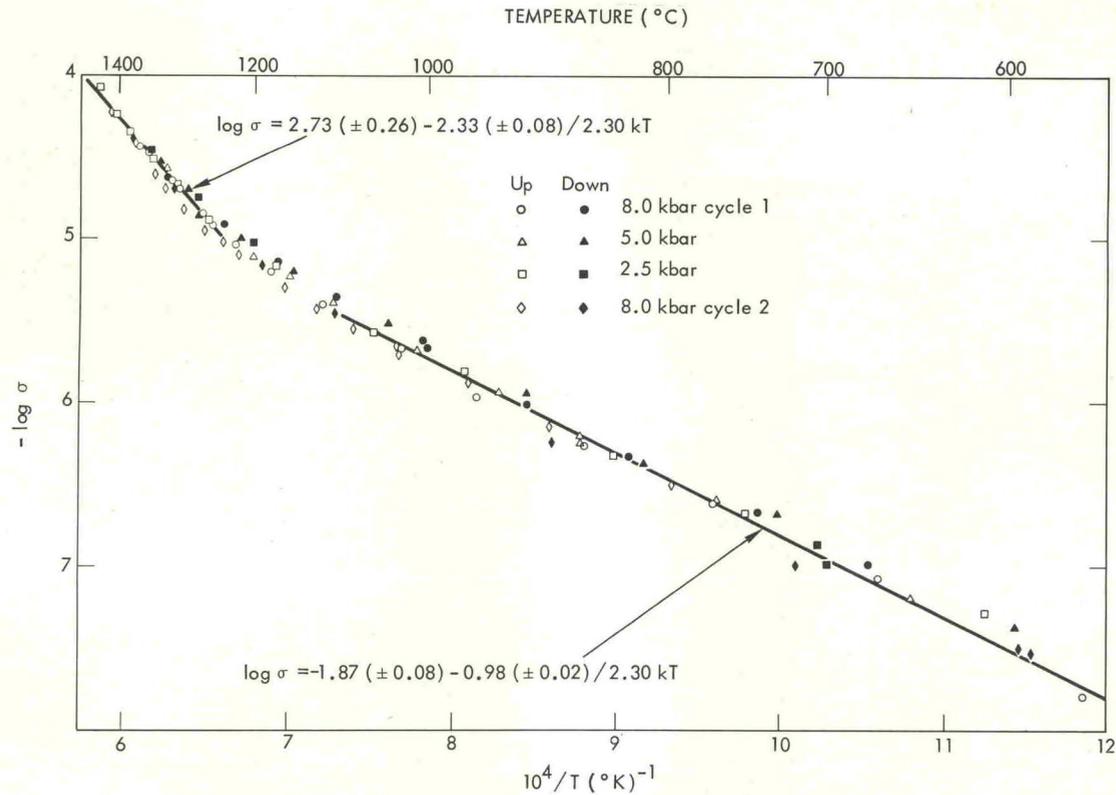


Fig. 2. Compilation of the data in Figure 1 with linear regression fit to (1).

TABLE 1. Results of Linear Regression Analyses of σ Data for Red Sea Olivine [100]

Experimental Conditions	Temperature Range, °C	$\log \sigma_x$	A_x , eV	Number of Observations
8.0 kbar in argon (Fig. 1a)	Cycle 1			
	560 to 1120	-1.77 ± 0.11	1.00 ± 0.02	14
	Transition region	4
	1270 to 1370	2.46 ± 0.35	2.24 ± 0.11	6
8.0 kbar in argon (Fig. 1a)	Cycle 2			
	600 to 1120	-1.85 ± 0.12	0.99 ± 0.03	17
	Transition region	6
	1270 to 1410	3.66 ± 0.34	2.64 ± 0.11	9
5.0 kbar in argon (Fig. 1b)	600 to 1100	-1.76 ± 0.17	0.99 ± 0.04	12
	Transition region	4
	1270 to 1370	2.18 ± 0.82	2.15 ± 0.26	6
2.5 kbar in argon (Fig. 1c)	610 to 1060	-2.12 ± 0.16	0.92 ± 0.04	7
	Transition region	4
	1310 to 1440	2.36 ± 0.23	2.20 ± 0.08	13
8.0, 5.0, and 2.5 kbar in argon (Fig. 2; Fig. 4, curve 7)	560 to 1120	-1.87 ± 0.08	0.98 ± 0.02	50
	Transition region	18
	1270 to 1440	2.73 ± 0.26	2.33 ± 0.08	34
$f_{O_2} \sim 10^{-8}$ bar at 1200°C* (Fig. 3; Fig. 4, curve 4)	Cycle 1 (up)			
	890 to 1390	-0.18 ± 0.05	1.51 ± 0.01	31
	1400 to 1500	3.42 ± 0.28	2.74 ± 0.10	15
	1510 to 1540	18.01 ± 0.85	7.89 ± 0.30	5
$f_{O_2} = 10^{-8}$ bar at 1200°C (Fig. 3; Fig. 4, curve 6)	1550 to 1660	5.07 ± 0.17	3.23 ± 0.06	19
	Cycles 1 (down), 2, 3			
	870 to 1320	-0.76 ± 0.05	1.34 ± 0.01	43
	1380 to 1450	2.41 ± 0.22	2.36 ± 0.08	19
$f_{O_2} = 10^{-8}$ bar at 1200°C (Fig. 4, curves 4 and 6)	1450 to 1490	11.48 ± 0.87	5.46 ± 0.30	11
	1490 to 1640	5.21 ± 0.16	3.26 ± 0.03	48
	1490 to 1660	4.82 ± 0.15	3.13 ± 0.06	67

*Includes $f_{O_2} = 10^{-8.4}$ bar at 1200°C.

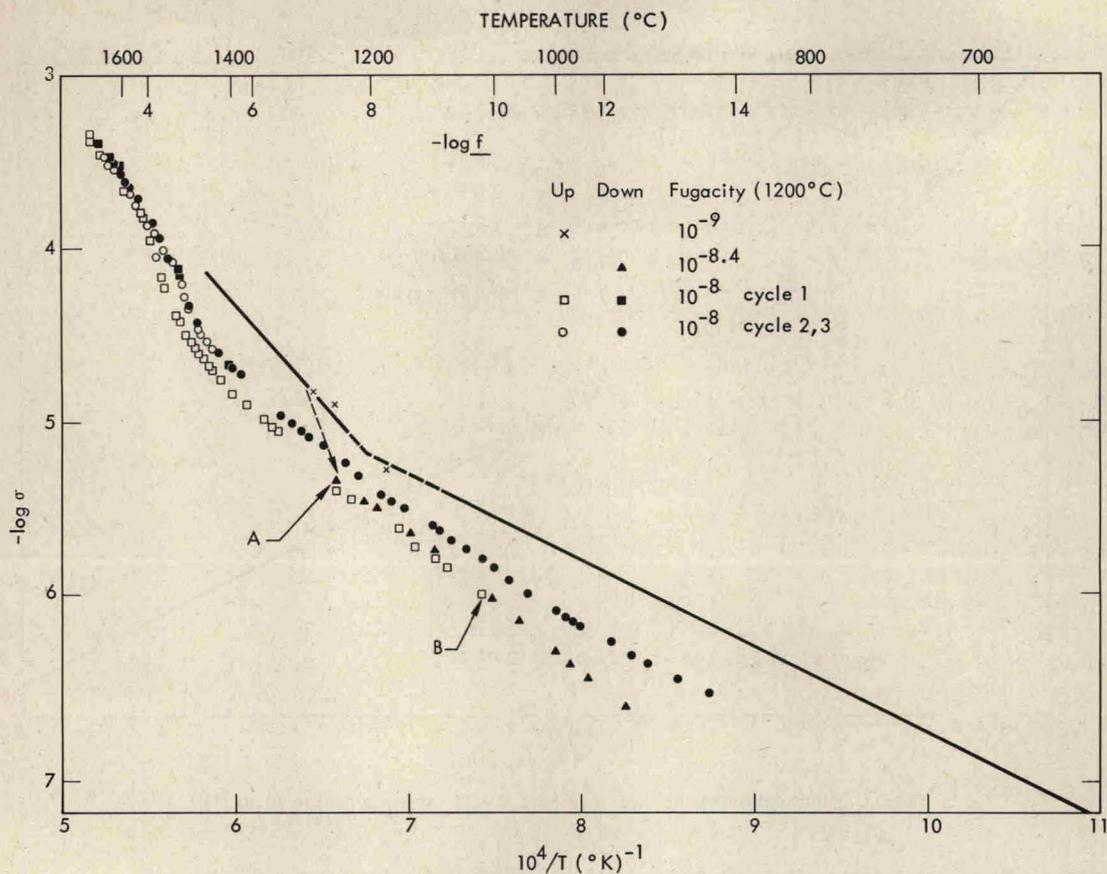


Fig. 3. Conductivity ($\Omega^{-1} \text{ cm}^{-1}$) of Red Sea olivine [010] to 1660°C. The linear regression fit for high- and low-temperature regions was replotted from Figure 2. Point A was measured after 15 hours at $\sim 1200^\circ\text{C}$. Point B was measured after 30 hours at $\sim 1000^\circ\text{C}$. For a H_2/CO_2 mix yielding $f_{\text{O}_2} = 10^{-8}$ bar at 1200°C , $-\log f_{\text{O}_2}$ is plotted along the upper abscissa.

tivity data identical with those for the second cycle. Analyses of the data from the various cycles are summarized in Table 1. After removal from the assembly several additional fractures were noted, and the sample was somewhat paler than when originally inserted.

The irreversible increase in σ below 1500°C , which was observed after the temperature increase portion of the first cycle, could be the result of fracturing or contamination of the sample with impurities released from the furnace parts at high temperature [Osburn and Vest, 1971]. Besides the increase in σ , which occurred after the first temperature increase cycle to 1660°C , the most remarkable feature of the higher-temperature data is the discontinuous change in σ between temperatures of 1460°C – 1500°C . This completely reversible discontinuity was observed in all three cycles to high temperature.

DISCUSSION

The data of Figures 1, 2, and 3 indicate that the temperature variation of the σ of olivine may be described by the equation

$$\sigma = \sum_x \sigma_x \exp(-A_x/kT) \quad (1)$$

where σ_x is a constant dependent on mechanism x , A_x is an activation energy for the conduction mechanism, k is the Boltzmann constant, and T is temperature. Table 1 lists σ_x and A_x at several values of pressure and f_{O_2} for various temperature ranges. Figures 1 and 2 and Table 1 demonstrate that pressure

to 8 kbar has no measurable effect on the σ of the present sample. For olivine from the same locality, Hughes [1955] reported a pressure effect equivalent to about a 14° change in temperature over an 8.2-kbar range between 1060° and 1240°C . This observation lies within the scatter of our data and is less than the temperature gradient across his sample. Hughes determined his pressure effect with measurements at only three temperatures: 1060° , 1160° , and 1240°C , each at six different pressures. Between 1060° and 1240°C we typically have 10 data points at each of three pressures.

The lack of a measurable pressure effect, distinct from experimental uncertainty, in the present study may be due to a number of factors. The error associated with the σ measurement is 1–2%. The error in temperature measurement is more difficult to assess. Although the temperature differences across the 0.37-mm-thick sample were 10°C or less, the thermocouples would undergo 5° – 15°C fluctuations with time, evidently in response to convection in the argon. Thus a good proportion of the uncertainty is due to our inability to determine adequately the temperature of the sample in the pressure vessel. Another potential contributor to a change in σ is a partial equilibration of the olivine with the high-pressure atmosphere during the measurement, since no annealing at high temperature and high pressure was performed prior to the σ measurement. On the other hand, the effect of pressure on σ is expected to be instantaneous in comparison with any chemical reaction. Thus any appreciable pressure effect should dominate initially. We conclude that the maximum

effect of 8-kbar pressure on the σ of the Red Sea olivine is equivalent to less than $\pm 5^\circ\text{C}$ temperature change (within the 95% confidence limit) between 1270° and 1440°C.

The A_x and σ_x given in Table 1 for σ in the Red Sea olivine may be compared with those from other studies on σ in olivine single crystals (Table 2). Because of uncertainties associated with grain boundaries, degree of compaction, impurity levels, and oxidation state of the cations in the various samples, most of the σ data on natural or synthetic monomineralic aggregates are difficult to interpret. Also, since most previous studies of σ of olivine single crystals have been performed on relatively highly conductive, partially oxidized samples [Duba et al., 1973], we shall limit our discussion to the present results and the data of Shankland [1969], Duba [1972], Hughes [1953, 1955], and Duba and Nicholls [1973]. All of these studies present data on highly resistive, presumably unoxidized, forsterite-rich olivine with activation energies greater than 1 eV. The results of these studies are plotted in Figure 4, and the experimental details are compiled in Table 2.

Hughes [1953] measured the σ of olivine from the same locality as the present olivine up to 1400°C in air. He reported no detectable difference in σ between the temperature increase and temperature decrease portions of the measurement cycles. In contrast, most olivines rapidly become better conductors at

TABLE 2. Values of A_x and σ_x for σ in Single-Crystal Forsterite

Description	T Range, °C	log σ_x	A_x , eV
Hughes [1953]; Red Sea olivine (Fo 90); in air; Fig. 4, curve 5; [010]	700 to 1100 1100 to 1400	0.33 5.16	1.66 3.01
Hughes [1955]; Red Sea olivine (Fo 90); in nitrogen; Fig. 4, crosses and circles; direction unknown	1060 to 1240	5.60*	2.70
Shankland [1969]; synthetic Fo 100; in air; Fig. 4, curve 1; direction unknown	200 to 1150 1150 to 1400	-3.17 6.53	1.00 3.83
Duba [1972]; Red Sea olivine (Fo 91); in argon at 7.5 kbar; Fig. 4, curve 8; [010]	400 to 1100 1100 to 1200	-0.62 20.06(?)	1.21 7.12(?)
Duba [1972]; synthetic Fo 100; in argon at 7.5 kbar; Fig. 4, curve 9; direction unknown	750 to 1100 1100 to 1200	-0.71 21.97(?)	1.70 7.99(?)
Duba and Nicholls [1973]; San Carlos olivine (Fo 92); $f_{O_2} \sim 10^{-12}$ bar at 1200°C; Fig. 4, curve 3; [001]	800 to 950 950 to 1350	-2.33 0.81	1.07 1.82
Duba and Nicholls [1973]; San Carlos olivine (Fo 96 containing metallic Fe); $f_{O_2} \sim 10^{-12}$ bar at 1200°C; Fig. 4, curve 2; [001]	900 to 1350	2.20	2.32

*Hughes' [1955] reported σ_x yields a higher σ than is indicated by his Figure 1 or the present Figure 4.

about 700°C in an oxidizing atmosphere; olivine from the Red Sea is apparently a notable exception [Duba, 1972]. Hughes' [1955] measurement of the σ of olivine from this

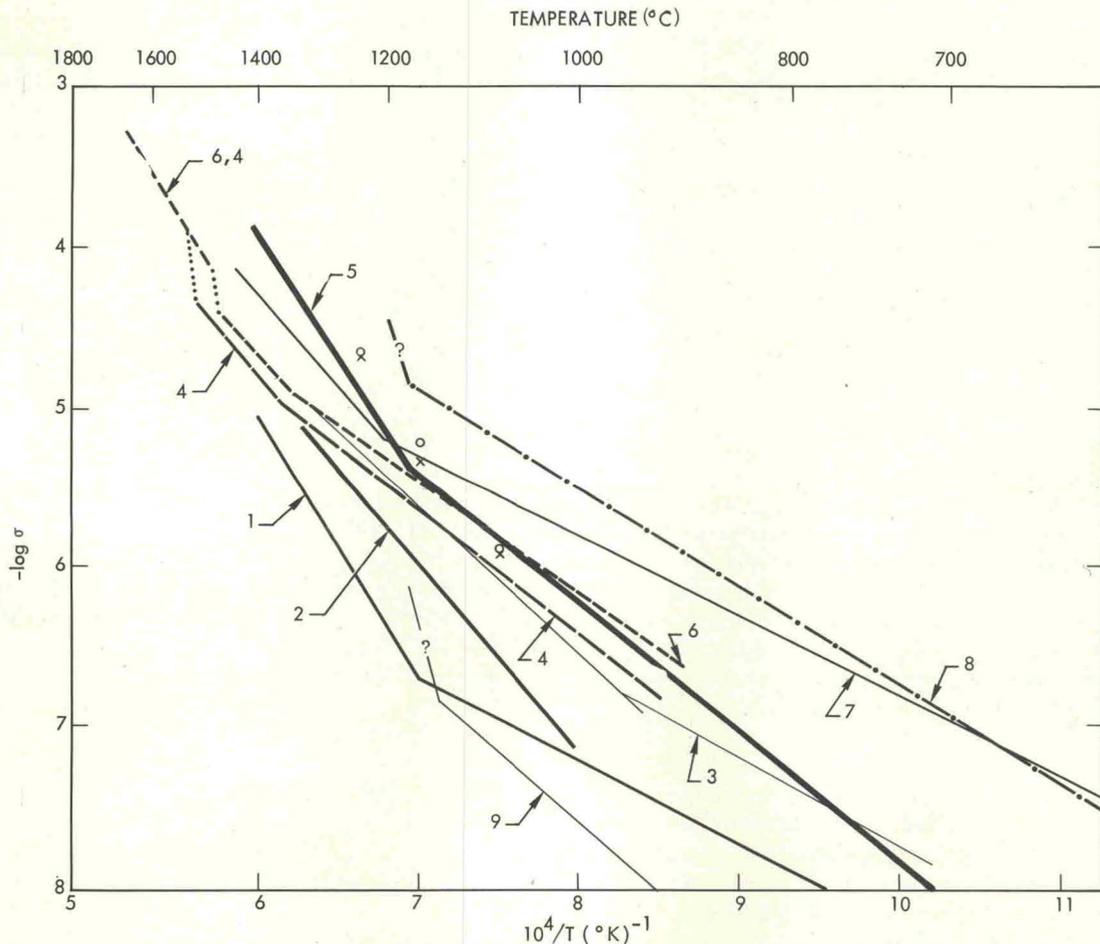


Fig. 4. Summary of σ data ($\Omega^{-1} \text{ cm}^{-1}$) for low-conductivity forsterite single crystals. The following numbers and symbols refer to data in Tables 1 and 2: curve 1, Fo 100 (~0.2 wt % Fe) [Shankland, 1969]; curve 2, Fo 96 [Duba and Nicholls, 1973]; curve 3, Fo 92 [Duba and Nicholls, 1973]; curve 4, cycle 1 (up), $f_{O_2} \sim 10^{-8}$ bar at 1200°C (this work); curve 5 [Hughes, 1953]; curve 6, cycles 1 (down), 2, and 3, $f_{O_2} = 10^{-8}$ bar at 1200°C (this work); curve 7, linear regression analysis of high pressure data (this work); curve 8, Fo 91 [Duba, 1972]; curve 9, Fo 100 (~0.2 wt % Mn) [Duba, 1972]; crosses, 8.2 kbar Fo 90 [Hughes, 1955]; circles, 2.5 kbar Fo 90 [Hughes, 1955].

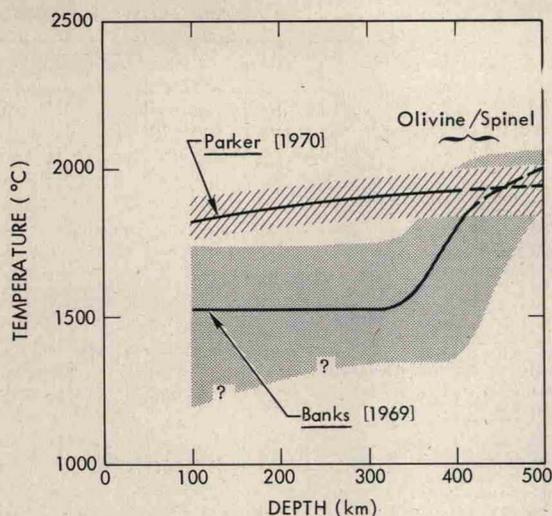


Fig. 5. Temperatures calculated for the mantle from conductivity-depth profiles and $\log \sigma_x = 4.82$ (0.15), $A_x = 3.13$ (0.06) as reported in Table 1. Uncertainties are indicated by stippling [Banks, 1969] and cross-hatching [Parker, 1970].

locality in nitrogen to 8.2 kbar does not differ significantly from his 1953 results. However, his reported σ_x is not consistent with the data in his Figure 1, which are plotted here in Figure 4. The data of Duba [1972] for the Red Sea olivine to 7.5 kbar in argon are similar to those of Hughes [1953, 1955] and those of the present study in absolute σ below 1100°C (Figure 4), although there is some variation in A_x and σ_x as indicated in Tables 1 and 2.

The Red Sea olivine is the most resistive naturally occurring olivine [Duba and Lilley, 1972]. However, the conductivity of the San Carlos olivine was reduced by about 3 orders of magnitude after annealing at $f_{O_2} = 10^{-18}$ bar at about 1340°C [Duba and Nicholls, 1973]. This f_{O_2} is within an order of magnitude of that necessary to reduce Fe^{2+} in fayalite to metallic iron. The ensuing σ is nearly coincident with that from the Red Sea [Duba and Nicholls, 1973]. The reported σ for the four studies on the Red Sea olivine and the annealed San Carlos olivine differ by less than an order of magnitude above 1100°C if the very high A_x (8 eV) data [Duba, 1972] are excluded. We believe that value to be a result of deterioration in the Cr-Al thermocouples. The variation in σ for these olivines (Fo 90–92) reflects not only experimental uncertainties in the measurements of conductivity, temperature, and experimental atmosphere but also sample differences such as impurity contents. The latter point is illustrated by the data for the synthetic forsterite single crystals plotted in Figure 4.

The sample Shankland [1969] measured (curve 1) was doped with ~0.2 wt % Fe, while that of Duba [1972] (curve 9) contained ~0.2 wt % Mn. Also of interest is the decrease in σ of the San Carlos olivine (curve 2) after half of its iron and all of its nickel had been reduced to the metallic state [Duba and Nicholls, 1973]. The decrease in σ is less than half an order of magnitude, but the A_x is significantly higher, indicating perhaps a change in the mechanism of conduction. The data of Figure 4 suggest that below 1400°C the σ of natural olivine is extrinsic (i.e., defect controlled), either ionic or electronic.

If the conduction mechanism is ionic, then data at the higher temperatures of this study ($T > 0.9$ of the melting temperature) may provide the activation energy for intrinsic ionic conduction [Lidiard, 1957]. Unfortunately, the picture is confused by the peculiar behavior noted at 1460°–1500°C,

where a high activation energy is apparently superseded by a lower energy. This behavior could indicate a change in the defect structure or a chemical change in the olivine resulting from the higher f_{O_2} ($\geq 10^{-5}$ bar) at temperatures above 1460°C (Figure 3). For example, Buening and Buseck [1973] propose cation vacancies to explain the dependence of diffusion in olivine on f_{O_2} . Discontinuities in σ could also be due to a phase change; however, existing thermodynamic data to 2000°K [Robie and Waldbaum, 1968] fail to indicate higher-order phase transitions. A reversible change in the ordering of the olivine structure (low-order transition) with increasing temperature as hypothesized by Virgo and Hafner [1972] could also give rise to the observed discontinuity.

Other processes that might be considered include exsolution of impurity defects at high temperatures, as proposed to explain similar behavior in BeO [Condit and Hashimoto, 1967], or an interaction between impurities and vacancies, which may decrease vacancy concentration at high temperatures as in doped NaCl [Dreyfus and Nowick, 1962]. Both mechanisms have been proposed to explain an observed decrease in activation energy at high temperatures, similar to that which is observed here. Clearly, more work needs to be done before an exact mechanism of conduction may be specified for olivine.

GEOPHYSICAL IMPLICATIONS

Figure 5 shows two temperature-depth profiles for the earth between 100 and 500 km that were calculated using σ -depth data for the earth's upper mantle [Banks, 1969; Parker, 1970], (1), and the present σ data (Figure 3) under controlled f_{O_2} at temperatures of $>1500^\circ\text{C}$. Except for the addition of a lower limit to Banks' solution (indicated by the question mark in Figure 5), each profile is shown bounded by regions that reflect the temperature uncertainty resulting from the σ error and depth error that each author attributes to his determination.

The temperature-depth profiles calculated from the data of Banks [1969] and Parker [1970] are also shown in Figure 6, along with the melting curves of several relevant materials

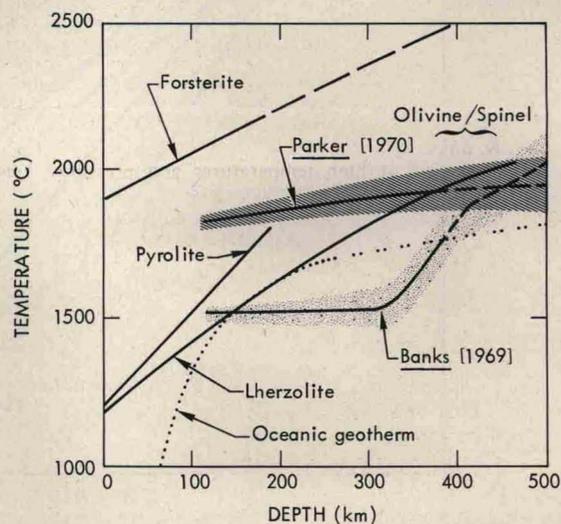


Fig. 6. Temperature-depth profile replotted from Figure 5. Uncertainty bands are based on measured pressure effect on σ (Figures 1a–1c) and σ_x and A_x values (Figure 2). Also shown are the lherzolite solidus of Ito and Kennedy [1967], as extended by using the Simon equation [Griggs, 1972], the forsterite solidus [Davis and England, 1964], the pyrolite solidus, and a proposed oceanic geotherm [Ringwood, 1966].

and an oceanic geotherm. The temperature uncertainties are now based on the estimated error in our σ data. Implied in the results in Figures 5 and 6 is the requirement that the σ of the earth's mantle at these depths is controlled by olivine. Olivine with a composition near Fo 90 is believed to undergo a phase change to spinel, perhaps via an intermediate phase, at a depth near 380 km, the transformation being likely to be complete by about 450 km [Fujisawa, 1968; Ringwood and Major, 1970; Ringwood, 1972]. Other assumptions in the calculations are that there is no further change in conduction mechanism in olivine above 1660°C and that the pressure effect at higher temperatures is similar to that observed below 1440°C. Furthermore, the temperatures calculated from the present data are correct only if the oxygen fugacity in the mantle is near the values indicated at the top of Figure 3. Comparison of Figures 5 and 6 indicates clearly that although the uncertainty in σ due to the maximum possible pressure effect is large, it is at most only 25% of the total uncertainty due to lack of resolution of σ with depth for the earth at depths to 400 km. Thus improvements must be made in the resolution of the σ -depth data in order to improve the temperature profiles shown in Figures 5 and 6. It would also be helpful to be able to set realistic limits on the oxygen fugacity in the mantle.

Figure 3 demonstrates that the oxygen fugacity in the experimental atmosphere affects the electrical conductivity of olivine. Other structure-sensitive properties (i.e., elastic constants, creep properties, diffusion, optical absorption, thermal diffusivity) may also be affected at high temperatures by the atmosphere in which they are measured. We suggest therefore that more care be exercised in specifying the experimental conditions at which these properties are determined.

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