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Ferric iron dependence of the electrical conductivity of the Earth's lower mantle material

BY J. P. POIRIER, A. GODDAT AND J. PEYRONNEAU

Département des Géomatériaux (Unité Associée CNRS 1734), Institut de Physique du Globe de Paris, 4 Place Jussieu, 75252 Paris Cedex 05, France

We measured the DC electrical conductivity of samples of magnesiowüstite–perovskite assemblage, prepared by laser heating at 400 kbar in a diamond–anvil cell, from San Carlos olivine equilibrated at various oxygen partial pressures. The conductivity was measured as a function of temperature between room temperature and 400 °C.

The conductivity obeys the Arrhenius law, compatible with a conduction by small polarons. The activation energy is about 0.3 eV atm^{−1}, and decreases with increasing pressure and increasing oxygen fugacity of equilibration. The effect of the oxygen fugacity on the pre-exponential factor is negligible.

We suggest that the effect of oxygen fugacity is essentially due to the compression of the lattice accompanying the increase in trivalent iron ions content. The effect of oxygen partial pressure is therefore comparable to the effect of pressure. The present results confirm our previous estimate that the electrical conductivity of the lower part of the mantle is of the order of 1–10 S m^{−1}.

1. Introduction

The Earth's mantle screens the iron alloy core, the seat of the geodynamo, and acts as a filter for the variations of the geomagnetic field observed at the surface. The electrical conductivity of the lower mantle controls the characteristics of the filter. Also, whether the core transfers angular momentum to the mantle by electromagnetic coupling or some other mechanism, depends on the lower mantle electrical conductivity. There is no compelling reason to believe that the Earth's lower mantle has a chemical composition widely different from that of the upper mantle (Weidner 1986). It is therefore reasonable (and simpler) to accept that it essentially consists of an assemblage of silicate perovskite (Mg,Fe)SiO₃ and magnesiowüstite (Mg,Fe)O.

In previous studies, we have measured the DC electrical conductivity of the lower mantle material in an externally heated diamond–anvil cell, and investigated its temperature, pressure and iron content dependence (Peyronneau & Poirier 1989; Poirier & Peyronneau 1992; Shankland *et al.* 1993). Our results were compatible with the assumption that the conduction mechanism was hopping of holes from Fe³⁺ to Fe²⁺ ions, and should be dependent on the concentration of Fe³⁺, and hence on oxygen fugacity. However, we had only used San Carlos olivine, naturally equilibrated in the conditions of the upper mantle, as a starting material. In the present work, we complement the earlier studies by investigating the effect of equilibration of the

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same olivine at various oxygen fugacities. We thus obtain better constraints on the extrapolation of the experimental results to the conditions of the lower mantle.

2. Experimental procedure

Olivine crystals from a San Carlos peridotite xenolith (SC 8303), with a Fe/(Fe + Mg) ratio equal to 16.3 at. %, were ground in an agate mortar, under alcohol, down to a grain size of less than 5 μm . The powder was then equilibrated at 1200 °C, for 5 h (Wanamaker & Duba 1993), in a controlled atmosphere. Three batches of powder were treated in gas mixtures with CO/CO₂ volume ratios of 10, 1 and $\frac{1}{10}$, corresponding to oxygen partial pressures of 1.34×10^{-13} atm, 8.48×10^{-12} atm and 1.01×10^{-9} atm, respectively. Samples were then loaded in a diamond-anvil cell, in the 100 μm gap between two ribbons of tungsten, acting as gasket and electrical leads at the same time. Pressure was increased up to 40 GPa and a 60–90 μm -wide sample of the high-pressure assemblage, extending from one lead to the other, was transformed by laser heating. The cell was heated by an external resistive heater, and the DC electrical conductivity of the sample was then measured at temperatures between room temperature and 400 °C. The apparatus and the experimental procedure have been described in more detail by Poirier & Peyronneau (1992). The electrical conductivity is determined with an uncertainty of about a factor of two (Poirier & Peyronneau 1992).

One sample was recovered after the measurements were made; it was ion-beam thinned, and examined in transmission electron microscopy. It consisted, as expected, of an assemblage of silicate perovskite and magnesiowüstite, with equiaxed grains less than 10 μm in size. Two series of measurements were made.

(i) The effect of oxygen fugacity was investigated by measuring the electrical conductivity of samples prepared from olivine equilibrated at 1200 °C, under oxygen partial pressures of 1.34×10^{-13} , 8.48×10^{-12} and 1.01×10^{-9} atm. The measurements were made at 40 GPa, and temperatures between room temperature and 400 °C, in cycles of increasing and decreasing temperature. It must be emphasized that the measurements of conductivity as a function of temperature were made on samples prepared from olivines equilibrated at various oxygen fugacities, which is different from measurements made at constant oxygen fugacity. What matters really is not the oxygen fugacity but the corresponding atomic fraction of Fe³⁺ ions. Equilibration of olivine results in a given atomic fraction of Fe³⁺ ions, and it is reasonably assumed that the amount of Fe³⁺ ions thus formed is conserved in the high-pressure assemblage, prepared in the confined environment of the diamond cell.

(ii) The effect of pressure was investigated by measuring the electrical conductivity of a sample equilibrated at 1.01×10^{-9} atm oxygen partial pressure, at 40 GPa and temperatures between room temperature and 400 °C. The sample was then decompressed to 2 GPa and the same measurements were repeated.

3. Results

(a) *Effect of equilibration at various oxygen fugacities*

The experimental data are given in table 1 and the Arrhenius plots in figure 1. Electrical conductivity increases with oxygen partial pressure. At room temperature, the conductivity of samples prepared from olivine equilibrated at 1.01×10^{-9} atm is

Table 1. *Electrical conductivity, at $P = 40$ GPa, of samples prepared from olivine (16.3 at. % Fe) equilibrated at various oxygen fugacities*

$f_{\text{O}_2} = 1.34 \times 10^{-13}$ atm		$f_{\text{O}_2} = 8.48 \times 10^{-12}$ atm		$f_{\text{O}_2} = 1.01 \times 10^{-9}$ atm	
$1000/T$	σ (S m $^{-1}$)	$1000/T$	σ (S m $^{-1}$)	$1000/T$	σ (S m $^{-1}$)
3.38	0.000 109	3.38	0.000 51	3.38	0.003 95
3.01	0.000 349	3.16	0.000 887	2.98	0.009 26
2.72	0.001 01	2.78	0.002 61	2.66	0.0277
2.30	0.0051	2.51	0.008 46	2.35	0.0637
2.07	0.0136	2.34	0.0134	2.07	0.140
1.80	0.0306	2.15	0.0286	1.81	0.297
1.62	0.0818	1.95	0.0693	1.67	0.465
1.49	0.194	1.84	0.0822	1.58	0.628
		1.72	0.128	1.50	0.838
3.38	0.000 192	1.61	0.189		
2.99	0.000 682	1.52	0.260		
2.79	0.00126				
2.64	0.00224				
2.46	0.00412				
2.30	0.00714				
2.11	0.0137				
1.96	0.0268				
1.85	0.0417				
1.74	0.0625				
1.69	0.0765				
1.62	0.0921				
1.56	0.112				

almost two orders of magnitude greater than that of samples prepared from olivine equilibrated at 1.34×10^{-13} atm. However, the effect decreases as temperature increases and the Arrhenius plots of samples equilibrated at 1.34×10^{-13} , 8.48×10^{-12} and 1.01×10^{-9} atm converge for $1/T = 0$ (figure 2). This means that the pre-exponential factor σ_0 , in Arrhenius law: $\sigma = \sigma_0 \exp(-E/kT)$, does not depend on the oxygen fugacity, within experimental error; in other terms, only the activation energy E is affected.

The values of σ_0 and the activation energy E for the three oxygen fugacities are given in table 2. The activation energy dependence on oxygen fugacity (figure 2) can be expressed as

$$E \text{ (eV)} = 0.0445 - 0.0227 \log f_{\text{O}_2} \text{ (atm)}. \quad (3.1)$$

(b) Effect of pressure

The experimental data are given in table 3 and the Arrhenius plots in figure 3. The electrical conductivity of the same sample, equilibrated at 1.01×10^{-9} atm, measured at room temperature at 40 GPa, is greater by one order of magnitude than

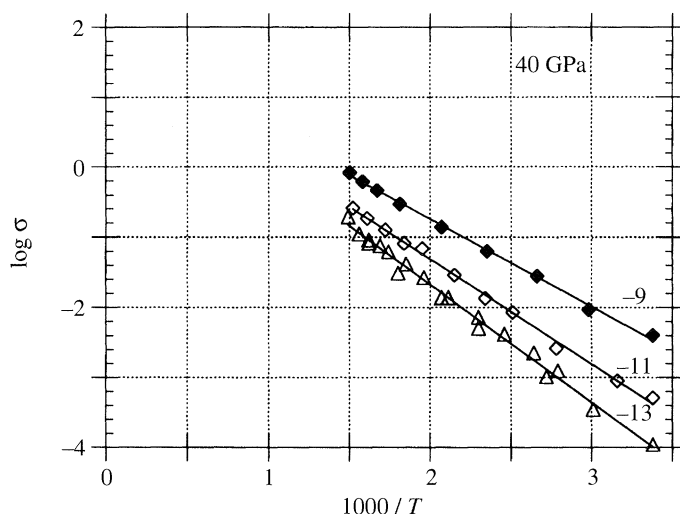


Figure 1. Arrhenius plots of the electrical conductivity (in S m^{-1}), at 40 GPa, of perovskite-magnesiowüstite assemblages prepared from San Carlos olivine (16 at. % Fe) equilibrated at 1200 °C, at oxygen partial pressures of 10^{-13} , 10^{-11} , 10^{-9} atm (the curves are labelled with the logarithm of the equilibration partial pressure). The equations of the linear fits to the experimental points are respectively: $\ln \sigma = 1.6807 - 1.679(1/T)$ ($r = 0.996$); $\ln \sigma = 1.6776 - 1.495(1/T)$ ($r = 0.998$); $\ln \sigma = 1.7566 - 1.250(1/T)$ ($r = 0.999$).

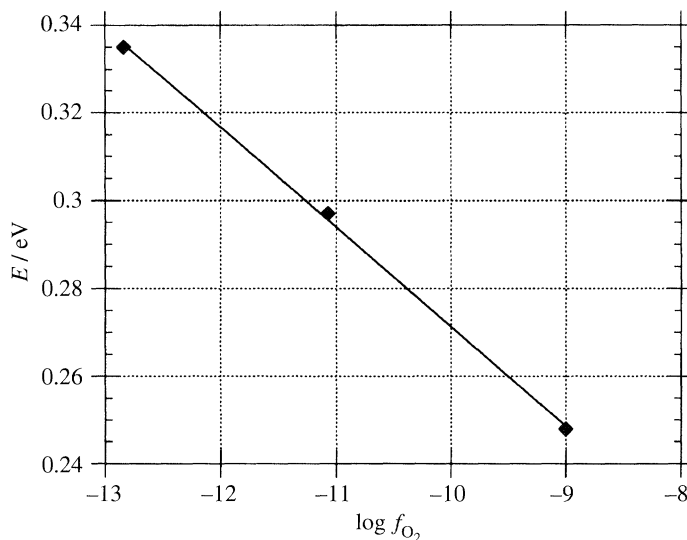


Figure 2. Dependence of the activation enthalpy E (eV atm^{-1}) on the oxygen fugacity at which the olivine was equilibrated, for the Arrhenius curves of figure 2. The equation of the linear fit is E (eV) = $0.0445 - 0.02269 \log f_{\text{O}_2}$ (atm) ($r = 1$).

when measured at 2 GPa. As expected, the pressure dependence resides only in the activation enthalpy: $E = E_0 + P\Delta V$, where ΔV is the activation volume, and the pre-exponential term σ_0 is constant within experimental error. We have

$$E \text{ (eV)} = 0.29124 - 0.00127P \text{ (GPa)}. \quad (3.2)$$

Table 2. Values of the pre-exponential factor σ_0 and the activation energy E for samples prepared from olivine equilibrated at $f_{\text{O}_2} = 1.01 \times 10^{-9}$ atm, 8.48×10^{-12} atm and 1.34×10^{-13} atm ($P = 40$ GPa)

$\log f_{\text{O}_2}$	σ_0 (S m^{-1})	E (eV)
-12.84	47.9	0.34
-11.07	47.6	0.30
-9	57.1	0.25

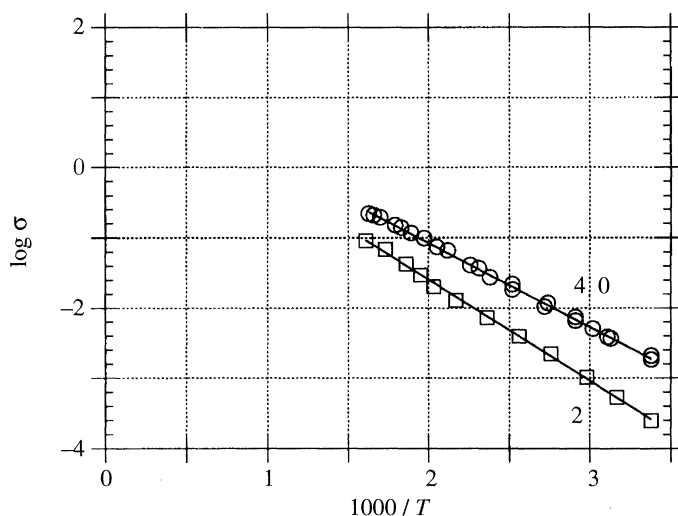


Figure 3. Arrhenius plots of the electrical conductivity (in S m^{-1}) of perovskite–magnesiowüstite assemblages prepared from San Carlos olivine (16 at. % Fe), equilibrated at 1200°C at an oxygen partial pressure of 10^{-9} atm, and measured at pressures of 40 and 2 GPa (the curves are labelled with the values of the pressure in GPa). The equations of the linear fits to the experimental points are respectively: $\ln \sigma = 1.3331 - 1.201(1/T)$ ($r = 0.999$); $\ln \sigma = 1.2876 - 1.442(1/T)$ ($r = 0.999$).

At 40 GPa, $E = 0.24 \text{ eV atm}^{-1}$ (in good agreement with the value found when investigating the oxygen fugacity dependence $E = 0.25 \text{ eV atm}^{-1}$), and at 2 GPa, $E = 0.29 \text{ eV atm}^{-1}$. The activation volume is $\Delta V = -0.12 \text{ cm}^3 \text{ mol}^{-1}$, and the pre-exponential factor is $\sigma_0 = 20.5 \pm 1 \text{ S m}^{-1}$. These values are comparable, within experimental error, to the values previously found for samples prepared from the same, untreated, olivine (Shankland *et al.* 1993): $\Delta V = -0.18 \text{ cm}^3 \text{ mol}^{-1}$ and $\sigma_0 = 18 \text{ S m}^{-1}$.

Conductivity data obtained at 40 GPa, on samples treated at various oxygen fugacities, and at 40 and 2 GPa, on samples treated at an oxygen fugacity of 10^{-9} atm, were simultaneously fitted to the equation

$$\sigma = \sigma_0 f_{\text{O}_2}^m \exp - \left(\frac{E_0 + P\Delta V + A \log f_{\text{O}_2}}{kT} \right), \quad (3.3)$$

using a nonlinear Levenberg–Marquardt method (Press *et al.* 1986).

We find: $\sigma_0 = 16.6 \pm 0.07 \text{ S m}^{-1}$, $m \approx 0$; $E_0 = 0.12 \pm 0.03 \text{ eV atm}^{-1}$; $\Delta V = -0.15 \pm 0.008 \text{ cm}^3 \text{ mol}^{-1}$; $A = -0.020 \pm 0.003 \text{ eV atm}^{-1}$.

Table 3. *Electrical conductivity of samples prepared from olivine (16.3 at. % Fe) equilibrated at $f_{\text{O}_2} = 1.01 \times 10^{-9}$ atm ($P = 40$ GPa and $P = 2$ GPa).*

$P = 40$ GPa		$P = 2$ GPa	
$1000/T$	σ (S m $^{-1}$)	$1000/T$	σ (S m $^{-1}$)
3.38	0.001 82	3.38	0.000 25
3.11	0.003 84	3.17	0.000 536
2.91	0.007 30	2.98	0.001 02
2.74	0.0118	2.76	0.002 19
2.52	0.0218	2.56	0.003 87
2.31	0.0376	2.36	0.007 38
2.12	0.0659	2.17	0.0130
1.97	0.0994	2.03	0.0204
1.83	0.140	1.95	0.0298
1.70	0.197	1.86	0.0432
1.63	0.225	1.73	0.0689
		1.61	0.0922
1.66	0.214		
1.79	0.152		
1.89	0.118		
2.05	0.0735		
2.26	0.0410		
2.38	0.0273		
2.52	0.0182		
2.72	0.0105		
2.91	0.006 62		
3.02	0.005 02		
3.13	0.003 68		
3.38	0.002 07		

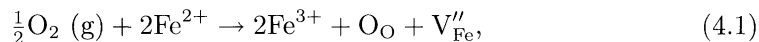
The values found for the coefficients are in good agreement with the values previously determined by separately fitting the data for pressure dependence and oxygen fugacity dependence: $\Delta V = -0.12$ cm 3 mol $^{-1}$ and $A = -0.0023$ eV atm $^{-1}$.

4. Discussion

Electrical conduction in the perovskite–magnesiowüstite assemblage is believed to be controlled by magnesiowüstite (Wood & Nell 1991), and due to hopping of holes from Fe $^{3+}$ to Fe $^{2+}$ ions (Shankland *et al.* 1993). An alternative, not fundamentally different, mechanism is one of extrinsic semi-conduction, with acceptor Fe $^{3+}$ ions generating holes in the valence band (Li & Jeanloz 1991, 1994).

The values of the activation energy and activation volume, and the fact that σ_0 is pressure independent, are compatible with a hopping mechanism (Shankland *et al.* 1993). However, one would expect the conductivity to depend on oxygen fugacity through the pre-exponential term. Indeed, all models of hopping conduction con-

sider that conductivity is proportional to the atomic fraction of trivalent iron ions $[\text{Fe}^{3+}]$ (see, for example, Tannhäuser 1962; Hirsch *et al.* 1993). In the simplest case, oxidation of Fe is controlled by the reaction



where V''_{Fe} represents iron vacancies, created to preserve electrical neutrality. Noting that $[\text{Fe}^{3+}] = 2[V''_{\text{Fe}}]$, the law of mass action gives

$$[\text{Fe}^{3+}]^3 = 2K f_{\text{O}_2}^{1/2}. \quad (4.2)$$

The conductivity, being proportional to $[\text{Fe}^{3+}]$, should therefore be proportional to $f_{\text{O}_2}^{1/6}$. More complicated cases have been analysed by Hirsch & Shankland (1991). In fact, the $f_{\text{O}_2}^{1/6}$ dependence is seldom observed, but a power-law dependence is at least expected. Wood & Nell (1991), for instance, in magnesiowüstite at ambient pressure, find $\sigma \propto f_{\text{O}_2}^{0.33}$.

The diamond cell under pressure is well sealed, and we can safely assume that the sample is self-buffered. The Fe^{3+} content of the powders equilibrated at various oxygen partial pressures is therefore assumed not to change during preparation of the sample. We also assume that Fe^{3+} preferentially partitions into magnesiowüstite in the same way as Fe^{2+} . The atomic fraction of trivalent iron $[\text{Fe}^{3+}]$ is equal to $1 - x$, where $x = \text{Fe}/\text{O}$ expresses non-stoichiometry in the wüstite component Fe_xO of magnesiowüstite. Increasing the oxygen fugacity f_{O_2} increases $[\text{Fe}^{3+}]$ and decreases x . It also increases electrical conductivity, but we find that the effect resides in the activation energy rather than in the pre-exponential factor, as currently believed. This has to be explained.

(i) First of all, we must note that our results do not really preclude an effect of f_{O_2} on the pre-exponential factor σ_0 . We can only say that the effect is less than the uncertainty of the measurements: i.e. about a factor of two. Now, if the conductivity is proportional to $[\text{Fe}^{3+}]$, we can estimate its variation with f_{O_2} , if we know the dependence of x on f_{O_2} . We can reasonably assume that this dependence is the same as for pure wüstite, and use the experimental relation given by Giddings & Gordon (1973) for wüstite equilibrated at 1200 °C:

$$\log f_{\text{O}_2} = 24.155/x - 37.298. \quad (4.3)$$

We find that when $\log f_{\text{O}_2}$ varies from -13 to -9 (as it does in our experiments), x varies from 0.99 to 0.85. It follows that $[\text{Fe}^{3+}]$, and hence the conductivity, increases by about 15%. This variation is well within our experimental error, and therefore undetectable.

(ii) To our knowledge, models of hopping conduction take into account the concentration of charge carriers $[\text{Fe}^{3+}]$ and $[\text{Fe}^{2+}]$, but tacitly assume that the activation energy for the jump of the hole from Fe^{3+} to Fe^{2+} is independent of $[\text{Fe}^{3+}]$, although it depends on pressure, in the usual way, through an activation volume (see, for example, Hirsch *et al.* 1993). However, we find that the hopping activation energy depends both on pressure and on the value of the oxygen fugacity at which the starting material was equilibrated. It can be written

$$E = E_0 + P\Delta V + A \log f_{\text{O}_2}, \quad (4.4)$$

where ΔV is the activation volume and A is an experimentally determined coefficient. The dependence of E on $\log f_{\text{O}_2}$ can be justified if E is written in the form

$$E = E_0 + P\Delta V + \mu[\text{Fe}^{3+}], \quad (4.5)$$

where $\mu = kT \ln[\text{Fe}^{3+}]$ is the chemical potential of the Fe^{3+} species, and $[\text{Fe}^{3+}]$ is proportional to f_{O_2} .

We suggest that the physical reason why the hopping energy depends in the same way on pressure and oxygen fugacity is due to the fact that the lattice parameter of wüstite Fe_xO decreases with x , i.e. the specific volume decreases as non-stoichiometry (or equivalently $[\text{Fe}^{3+}]$) increases (Hayakawa *et al.* 1972; Simons 1979). This effect can be put in parallel with the effect of pressure, and we can speculate that whatever the physical reason for an effect of pressure on the activation energy of the jump of a hole (e.g. shorter distance between ions, narrowing of the gap, etc.), increasing the non-stoichiometry of the wüstite component, by increasing the oxygen fugacity, should have the same effect as increasing pressure. Indeed, this reasoning is similar to that used by Birch (1961), when he successfully speculated that increasing the density of a mineral by changing its chemical composition should have the same effect on its elastic properties as increasing the density by compression.

We can compare the effect of pressure to the effect of oxygen fugacity in our experiments, by comparing experimental relations (3.1) and (3.2). We find that increasing f_{O_2} by one log unit has the same effect on the activation energy as increasing pressure by 17.2 GPa. An increase in $\log f_{\text{O}_2}$ from -13 to -9 would therefore be equivalent to a 69 GPa increase in pressure. We can check that the compression of the lattice due to pressure is similar to that due to the increase in Fe^{3+} content: using the bulk modulus of 26% Fe magnesiowüstite, $K = 167$ GPa (Jackson *et al.* 1978), we find that increasing pressure by 69 GPa causes a compression $\Delta V/V = -0.41$. Now, using relation (4.2), we find that increasing $\log f_{\text{O}_2}$ from -13 to -9 results in $\Delta x/x = -0.14$. As the lattice parameter a varies linearly with x (Hayakawa *et al.* 1972), we see that $\Delta a/a = -0.14$ and $\Delta V/V = 3(\Delta a/a) = -0.42$, in good agreement with the value resulting from application of the equivalent pressure.

The value of the agreement must not, of course, be overstated. Indeed, one may find fault with many of the assumptions underlying it. In particular, it has been pointed out that the olivine structure can accept only a very low concentration of Fe^{3+} ions; at the oxygen partial pressures and temperature conditions at which our olivine samples were equilibrated, the $\text{Fe}^{3+}/\Sigma\text{Fe}$ ratio should not exceed 10^{-4} (H. O'Neill, personal communication). It is therefore difficult to imagine how magnesiowüstite could pick up high concentrations of Fe^{3+} , unless by dismutation of Fe^{2+} into Fe^{3+} and Fe during laser heating. No iron was observed in electron microscopy, but that does not preclude the possibility of it being present in small dispersed particles (which anyway, would have no influence on the conductivity, because of their dispersion). Also, the experiments show a strong and very reproducible dependence of the conductivity on the oxygen partial pressure at which the olivine sample were treated, and even though temperature during laser heating cannot be perfectly controlled, one would expect such a consistent and reproducible effect as was observed.

Much, therefore, remains to be understood. However, the idea that Fe^{3+} concentration may play the same role as pressure in decreasing the activation energy of hopping, by promoting a contraction of the lattice, seems to be a promising lead.

5. Conclusions

The results of the experiments presented here complement our previous studies (Peyronneau & Poirier 1989; Poirier & Peyronneau 1992; Shankland *et al.* 1993), by investigating the effect of equilibrating the starting material at various oxygen fu-

gacities. Within experimental error, the pre-exponential factor σ_0 is independent of pressure and of oxygen fugacity. As we have equilibrated the olivine in a large range of oxygen fugacities, it seems reasonable to suggest that at the high temperatures of the lower mantle, electrical conductivity will be very little dependent on the (unknown) oxidation state. Extrapolation of our results to temperatures between 2000 and 4000 K yield conductivities of less than a few tens of S m^{-1} .

However, the present experiments have been carried out on a mineral assemblage prepared from olivine richer in iron (16 at. %) than the lower mantle, which probably does not contain more than 12 at. % Fe. Taking into account the effect of iron concentration on conductivity (Poirier & Peyronneau 1992; Shankland *et al.* 1993), these estimates should be somewhat lowered. We are therefore led to the conclusion that the electrical conductivity of a lower mantle composed of perovskite and magnesiowüstite is likely to be of the order of a few S m^{-1} : $1 < \sigma < 10 \text{ S m}^{-1}$.

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