

## ARE LABORATORY ELECTRICAL CONDUCTIVITY DATA RELEVANT TO THE EARTH?

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Before using laboratory data on the  $\sigma$  of rocks and minerals to interpret field electromagnetic data, one must be aware that the laboratory data may not be truly representative of conditions which generally prevail in the earth. Important conditions are oxidation state and time for solid-solid reactions to approach equilibrium. Data on the electrical conductivity ( $\sigma$ ) of rocks and minerals relevant to the crust and upper mantle of the earth are considered. Recent investigations which treat the time dependence of  $\sigma$  at high temperatures and the effect of oxidizing environments on  $\sigma$  are applied to the earth's upper mantle. The  $\sigma$  change at the olivine to spinel transition is unknown because of the effect of oxidation and contamination under the conditions of measurement. The  $\sigma$  of albite and basalt is observed to be time dependent below melting; thus, the assumption that  $\sigma$  increases with partial melting is questionable.

### I. Introduction

The electrical conductivity ( $\sigma$ ) of silicates has been studied extensively as a means for determining temperature vs. depth in the earth. In a recent review, SHANKLAND [41] has pointed out that  $\sigma$  is most affected by temperature, oxygen fugacity ( $f_{O_2}$ ), and transition-metal ion content. He indicates that the effects of orientation, twinning, and order-disorder have also been observed. The present review will discuss  $f_{O_2}$  effects, the importance of order-disorder phenomena, and the measurement of  $\sigma$  in polycrystalline materials.

When considering laboratory experiments on geological systems, an obvious question to ask is, "Do the experimental conditions differ significantly from those that prevail in the geological system of interest?" The point "differ significantly" is often missed. If one were asked to enumerate the important variables for the outer 400 km of the earth's mantle, he would probably respond that there are two whose limits are as follows: temperature up to about 2000 K, and pressure up to about 15 GPa. Perhaps he would enumerate  $f_{O_2}$ , but he most assuredly would not consider time as an important variable. In the real sense, time and  $f_{O_2}$  are probably not important variables in the mantle since time is usually sufficient for most chemical reactions to attain equilibrium, and  $f_{O_2}$  is controlled within fairly narrow limits by the phases present. *However, in the laboratory experiment, these two variables can be much more important than either temperature or pressure.* This situation arises because the  $f_{O_2}$  imposed in the laboratory is usually not controlled by the same phases

present in the mantle, and most experiments are performed in a time-frame of hours with little attention paid to kinetic effects. This paper will attempt to show where these two variables have led us astray regarding electrical conductivity measurements in the laboratory and their interpretation in terms of observations within the earth.

Electrical conductivity of silicate minerals is a thermally activated process which fits an equation of the form  $\sigma = \sigma_i \exp(-A_i/kT)$ . Here,  $\sigma_i$  is a constant depending on conduction mechanism,  $A_i$  is an "activation energy" which is the sum of the energy required to produce and to move a charge carrier in the structure. The term in the energy related to production vanishes at lower temperature where charge carrier concentration is extrinsically controlled. Despite much time and effort, no conduction mechanism has been satisfactorily defined for silicates likely to exist in the mantle. This arises from the difficulty of the measurement as in Hall effect studies [40], or from failure to control oxidation effects in thermoelectric measurements [6]. The  $\sigma$  of silicates shows a large dependence on temperature, and activation energies of 2 to 4 eV are common. However, the dependence of  $\sigma$  on pressure is small at least for olivine single crystals [14].

Studies of the variation of  $\sigma$  as a function of  $f_{O_2}$  have had a good deal of success in determining conduction mechanisms in simple oxides [29] and have been applied to very magnesian olivines with some success [34]. The  $\sigma$  of natural single crystals of olivine [15] containing about 10 weight% iron and pyroxene of about the same composition [13] cannot easily be interpreted in terms of the oxygen defect model. The large effect of oxidizing  $f_{O_2}$  on  $\sigma$  outside the olivine stability field [11], for example, shows that heed must be given to chemical stability when one attempts to use these simple oxygen defect models to explain conduction mechanisms in olivine of mantle composition. However, studies in the oxidizing regime of  $\sigma$  as a function of time and  $f_{O_2}$  may help in understanding the conduction mechanism within the stability field.

The other major effect to be discussed here is that of time. Studies in olivine below 1500 °C have indicated no time dependence of  $\sigma$  except where  $f_{O_2}$  was changed. However, there is some indication that time may be a factor above 1500 °C [14]. The  $\sigma$  of orthopyroxene also shows no rate effects so long as no phase transformations are induced by the pressure-temperature regime [17]. Once pyroxene is outside its  $P$ - $T$  stability field, it undergoes large, irreversible, time-dependent  $\sigma$  changes [13, 17].

Another phenomenon involving time is that of order-disorder in silicates. Plagioclase is a ubiquitous phase in crustal rocks such as granites and basalts and many comprise almost half the mineral fraction of rocks in the intermediate compositional ranges. The  $\sigma$  of albite, the sodic end-member of the plagioclase series increases with time and becomes equal to the  $\sigma$  of molten albite [33].



This is presumably the result of disorder, in the solid state. Thus, a previous observation [23] that the  $\sigma$  of albite increased several orders of magnitude on melting has to be qualified to take into account the temperature–time history of  $\sigma$ . This has implications for the discovery and delineation of partial melt zones in the earth.

## 2. Electrical conductivity and $f_{O_2}$

Figure 1 is the electrical conductivity of polycrystalline olivine of composition ranging from pure fayalite to pure forsterite as a function of temperature at various pressures. It is compared with the  $\sigma$  of a single crystal of Fo 90 composition from St. John's Island, Red Sea, Egypt [14]. Several features

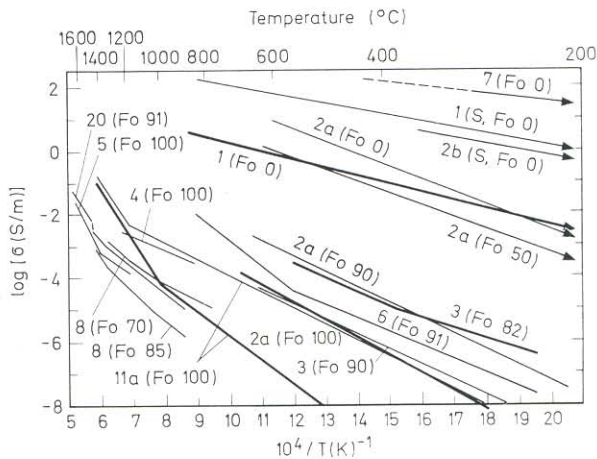


Fig. 1. The electrical conductivity of olivine. All measurements are powdered material except # 20. Composition and phase are identified in parantheses. S refers to the conductivity of spinel phase, composition ranges from pure fayalite (Fo 0) to pure forsterite (Fo 100). Numbers refer to studies identified below: 1) AKIMOTO and FUJISAWA [1]; 2a) BRADLEY et al. [6]; 2b) BRADLEY et al. [5]; 3) HAMILTON [19]; 4) JANDER and STAMM [21]; 5) PLUSCHKELL and ENGELL [34]; 6) SCHULT and SCHOBER [39]; 7) MAO and BELL [26]; 8) DUBA [10]; 11a) HUGHES [20]; 20) DUBA et al. [14]

of the figure are noteworthy. Two investigators [1, 5] report an increase in  $\sigma$  with the inversion of olivine (Fo 0) to the spinel phase. At about 800 °C the reported  $\sigma$  of pure fayalite is  $10^8$  times higher than that for pure forsterite. The  $\sigma$  reported for olivine of interest to the upper mantle (Fo 90) and pure fayalite (Fo 0) span about four orders of magnitude at about 800° and 300 °C, respectively, while the largest span for reported  $\sigma$  of pure forsterite is less than two orders of magnitude at 800 °C. These large differences may be attributed to the enormous number of experimental variables among these

studies. Pressure, sample purity and preparation methods, single crystal vs. polycrystal, and experimental atmosphere are obvious reasons to explain the differences observed. The question becomes one of choosing the data which were collected under experimental conditions most relevant to the interior of the earth. Polycrystallinity is an obvious consideration, and most reported values of the  $\sigma$  of polycrystals are higher than that of a single crystal under

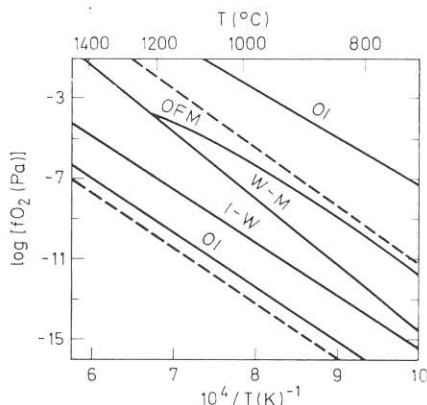
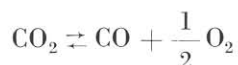
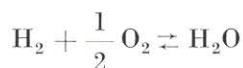


Fig. 2. Stability field of olivine as a function of temperature and  $f_{O_2}$ . Top line labelled O1 is for oxidation of olivine (Fo 90) NITSAN [28]; bottom line is for reduction of olivine (Fo 90) (DUBA and NICHOLLS [11]. QFM, W-M, and I-W are discussed in the text. Dashed lines represent the  $f_{O_2}$  as a function of temperature for the most oxidizing and reducing  $CO_2/CO$  mixes easily produced by our gas mixer

controlled  $f_{O_2}$  of similar forsterite content [compare polycrystal  $\sigma$ -lines 2a(Fo 90), 3(Fo 90, Fo 82), and 6 — with single crystal  $\sigma$ -line 20]. However, a polycrystal under controlled  $f_{O_2}$  is comparable in  $\sigma$  to a single crystal of similar forsterite content [compare polycrystal  $\sigma$ -line 8 (Fo 85) — with single crystal  $\sigma$ -line 20]. This observation is consistent with that of SCHOCK et al. [38] that the  $\sigma$  of polycrystals at high pressure is within one-half order of magnitude of that of the natural single crystals from which it was prepared and the difference is likely due to differing experimental  $f_{O_2}$ . Thus it is safe to assume that most  $\sigma$  results for polycrystals in Fig. 1 are not applicable because of oxidation problems during sample preparation and measurement in the laboratory [12]. There is a further issue that makes the measurements on polycrystals suspect when the heater is composed of graphite. It has been our experience that  $\sigma$  measured on alumina (DUBA, unpublished results) when graphite heaters were employed was several orders of magnitude higher than that measured when platinum foil heaters were used. This is because of the extreme mobility of carbon at high temperature which leads to contamination of the sample with highly conductive films of carbon. Because of oxidation

uncertainties, both demonstrations of the jump in  $\sigma$  associated with the fayalite to spinel transition [5, 1] are open to question. Assuming the observation of a  $\sigma$  jump to be valid (and it is plausible), the absolute  $\sigma$  is still in question of the unknown contribution of contaminants such as  $\text{Fe}^{3+}$  and/or carbon.

Figure 2 shows the olivine stability field for  $f_{\text{O}_2}$  vs.  $T^{-1}$  for olivine containing 10% fayalite (solid lines). The upper line is from NITSAN [28] and the bottom from DUBA and NICHOLLS [11]. Solid lines labelled QFM, W-M, and I-W are the solid buffers quartz-fayalite-magnetite, wüstite-magnetite, and iron-wüstite, respectively, and are taken from EUGSTER and WONES [18]. The dashed lines are the limits of  $f_{\text{O}_2}$  provided by our gas mixer and were calculated from thermodynamic data [7] for the reactions:



for various mixes [13]. The recent publication of DEINES et al. [8] provides a most convenient tabulation and display of  $f_{\text{O}_2}$  for various gas mixes which agrees with our calculations and is preferable and more reliable and convenient than calculations from thermodynamic data. The most useful feature of gas mixes for  $f_{\text{O}_2}$  control during experiments at high temperatures is that the  $f_{\text{O}_2}$  produced by constant gas mixes as temperature is increased, follows a path that is almost parallel to the olivine stability field and to the natural buffer systems—QFM, W-M, and I-W. This is significant for two reasons: as temperature is increased,  $f_{\text{O}_2}$  is maintained close to the  $f_{\text{O}_2}$  produced during heating of buffers which are likely to operate in the earth; and since the  $f_{\text{O}_2}$  of the experimental atmosphere is almost parallel to the olivine stability field, the number of oxygen defects produced during heating cycles is almost constant.

Figure 3 provides a demonstration of the large effect of  $\text{Fe}^{3+}$  on the  $\sigma$  of olivine. Line P1 is the  $\sigma$  of a peridotite measured in an unspecified atmosphere [31]; line 1 is the  $\sigma$  of olivine from San Carlos Indian Reservation, Arizona, measured in argon [9]; line 7a is  $\sigma$  for olivine from the same locality measured in air [11]. Slightly below 800 °C, the  $\sigma$  began to become irreversible. A gas mixer designed to control  $f_{\text{O}_2}$  was then attached to the sample assembly and a  $\sigma - T$  path was followed as indicated by the dashed line. The  $\text{H}_2/\text{CO}_2$  mix was changed to maintain a constant  $f_{\text{O}_2}$  of  $10^{-7}$  Pa up to about 1300 °C, at which point the mix was maintained constant and provided a  $f_{\text{O}_2} - T$  trajectory about one order of magnitude above and almost parallel to the bottom line marked "01" in Fig. 2. The  $\sigma - T$  path followed by the sample on heating and cooling cycles with this atmosphere is line 7b and is in good agreement with  $\sigma$  measured for gem quality, and  $\text{Fe}^{3+}$ -poor [12], olivine from



the Red Sea measured as follows: in argon (6a— [9]), in air (3— [20]), and in a mix of  $H_2$  and  $CO_2$  that is about one order below, and parallel to, the upper dashed line in Fig. 2 [14]. Admission of a small amount of air into the experimental atmosphere at 1000 °C causes an almost immediate increase in  $\sigma$  of several orders of magnitude. It then requires two to six hours for the

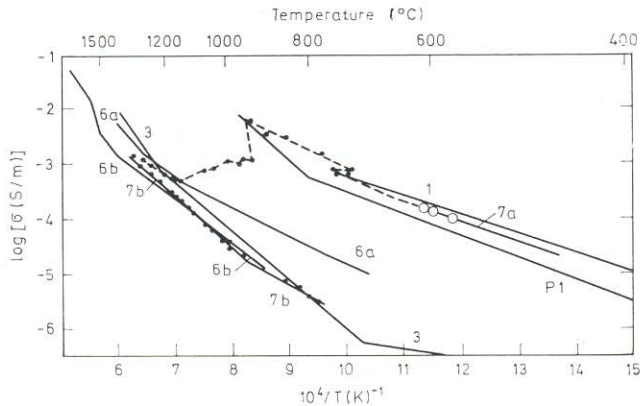


Fig. 3. The electrical conductivity of olivine single crystals of —10% fayalite composition. Lines are coded as follows. 1) Olivine from San Carlos Indian Reservation, Arizona, in argon, DUBA [9]; 3) olivine from St. John's Island, Red Sea, Egypt, in air, HUGHES, 1975; 6a) olivine from St. John's Island, Red Sea, Egypt, in argon, DUBA et al. [14]; 6b) same specimen as 6a, but under controlled  $f_{O_2}$ , DUBA et al. [14]; 7a) olivine from San Carlos Indian Reservation, Arizona, in air, DUBA and NICHOLLS [11]; 7b) same specimen as 7a, but under controlled  $f_{O_2}$ , DUBA and NICHOLLS [11]; P1) peridotite, locality and atmosphere of measurement unknown, PARKHOMENKO [31]

$\sigma$  to be re-equilibrated in a  $H_2/CO_2$  mix which is inside the olivine stability field as delineated by the solid lines marked "01" in Fig. 2. The effect of pressure on  $\sigma$  in olivine over an 800 MPa (8 kbar) range is less than a temperature change of  $\pm 5$  °C at temperatures between 1270° and 1440 °C [14].

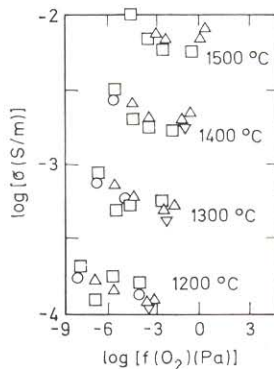


Fig. 4. Electrical conductivity of olivine from St. John's Island, Red Sea, Egypt, as a function of temperature and  $f_{O_2}$  (DUBA et al. [15])

Figure 4 demonstrates the effect of  $f_{O_2}$  on the  $\sigma$  of olivine from the Red Sea area [15] as a function of temperature to 1500 °C. As can be seen from Fig. 2, our gas mixer does not allow close approach to oxidation, but allows for reduction, of olivine. Data presented in Fig. 4 were obtained within the stability field of olivine and show a dependence that is approximately pro-

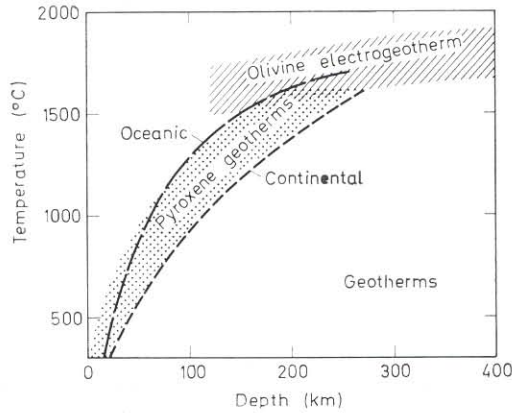


Fig. 5. The temperature vs. depth profile calculated from line 6a in Fig. 3, and the  $\sigma$  vs. depth data discussed in the text. The pyroxene geotherms (BOYD [3]; MACGREGOR and BASU [25]; and MERCIER and CARTER [27]) and postulated continental and oceanic geotherms (RINGWOOD [36]) are shown for comparison

portional to  $(f_{O_2})^{+1/6}$  at high  $f_{O_2}$  but approximately proportional to  $(f_{O_2})^{-1/12}$  at low  $f_{O_2}$ . From the work of SMYTH and STOCKER [42], this is consistent with a mechanism dependent on oxidation of ferrous to ferric iron at high  $f_{O_2}$  but the slope at low  $f_{O_2}$  does not fit a simple oxygen defect model. A similar interpretation for high  $f_{O_2}$  was made by SHANKLAND [41] for results which were obtained by PARKIN [32] for  $\sigma$  measured on Fe-doped synthetic forsterite under controlled  $f_{O_2}$ . The results of DUBA et al. [15], at low  $f_{O_2}$  are not consistent with SHANKLAND's interpretation of PARKIN's data for low  $f_{O_2}$ .

Figure 5 shows the geotherm calculated from line 6b of Fig. 3 and literature data for  $\sigma$  of the earth's mantle between 100 and 400 km [37, 22, 2, 30, 43]. The broad span in temperature is due to the range of  $\sigma$  values reported by these authors. The use of line 6b is reasonable if olivine of composition Fo 90 controls the  $\sigma$  of the earth's mantle since the  $\sigma$  of olivines of similar compositions and quite different histories (lines 6b and 7b, Fig. 3) agree very well under controlled  $f_{O_2}$ . Also shown in Fig. 5 are the limits of various values reported for geotherms calculated from pyroxene inclusions in nodules derived from the upper mantle [3, 27, 25] and the continental and oceanic geotherms proposed by RINGWOOD [36]. From this figure it is clear that a mantle whose  $\sigma$  is controlled by olivine is consistent with a reasonable geotherm at depths greater

than about 150 km. At shallower depths, however, the temperatures are considerably higher than expected. This suggests  $\sigma$  is controlled in the outer 150 km by other more conducting phases, perhaps interstitial water, partial melts, and other grain boundary impurities, or some other mineral species.

### 3. The effect of order—disorder and partial melt on $\sigma$

KHITAROV and SLUTSKII [23] have shown that the  $\sigma$  of albite, the Na-rich end-member of the plagioclase feldspar series, increases three to four orders of magnitude upon melting (solid and dashed lines, Fig. 6). PIWINSKII and DUBA [33], however, have shown that the  $\sigma$  of albite increases a similar

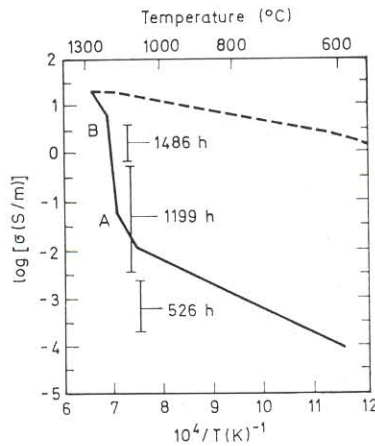


Fig. 6. The electrical conductivity of albite. Solid line is for polycrystalline albite prior to melting; dashed line is same sample during and after melting (KHITAROV and SLUTSKII [23]). The vertical lines are measured as a function of time as indicated at temperature below the solidus (PIWINSKII and DUBA [33])

amount below melting provided time is allowed for disorder to proceed. These data are shown as the solid vertical lines in Fig. 6.

More recent work [15] on basalt containing about 35% plagioclase indicates that  $\sigma$  increases with time subsolidus. These data are shown in Fig. 7 in which a comparison is made among the  $\sigma$  of samples from the same rock at various  $f_{O_2}$  and with differing time-temperature histories. The figure clearly demonstrates that the  $\sigma$  change upon partial fusion at 1050 °C (solidus temperature is  $1020 \pm 8$  °C) is dependent on the time spent near but below the beginning of melting by the sample. The  $\sigma$  is also dependent on  $f_{O_2}$  in this rock. Studies such as those of PRESNALL et al. [35] and WATANABE [44] and KHITAROV and SLUTSKII [23] on basalt and that of LEBEDEV and KHITA-



ROV [24] on granite must be used with caution since time was not a significant variable in their experiments. Hence, equilibrium was not attained. When time is considered in materials which show increases in  $\sigma$  upon disordering below melting, no significant electrical distinction may be made between a partial melt and a solid of similar composition which is slightly cooler.

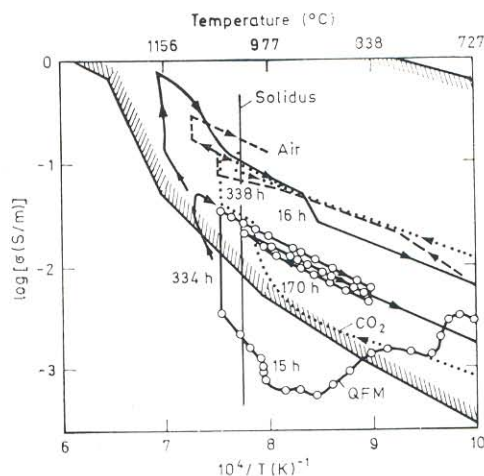


Fig. 7. The electrical conductivity of basalt (DUBA et al. [16]). Lines are coded for  $f_{O_2}$ : solid lines are for  $CO_2/CO$  gas mix near the QMF buffer, dotted lines for pure  $CO_2$ , and broken lines for air. Constant temperature portions of lines are indicated by heavy shading, with time, in hours, to the side. Normal heating rate is  $100\text{ }^\circ\text{C/h}$ . The solidus for this material is indicated by the vertical line. The heavy lines with almost vertical shading are the limits of literature  $\sigma$  data for basalt

However, partial melts such as basalt in a solid whose  $\sigma$  is controlled by olivine will show a conductivity contrast of two to four orders of magnitude from the data presented in Figs 3 and 7.

## 6. Conclusion

In summary, temperature and  $f_{O_2}$  are the most important variables to consider in the interpretation of laboratory  $\sigma$  measurements. In addition, kinetics becomes important if a time-dependent reaction such as order-disorder has a significant effect on  $\sigma$  as in albite and basalt. Pressure is not a significant variable except where it produces crack closure which limits water movement in rocks near the surface [4] and at depth where phase transitions may be involved.

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## ПРИМЕНЯЕМОСТЬ ЛАБОРАТОРНЫХ ДАННЫХ ЭЛЕКТРИЧЕСКОЙ ПРОВОДИМОСТИ ДЛЯ ЗЕМЛИ

А. ДУБА

РЕЗЮМЕ

Прежде чем использовать лабораторные данные горных пород и минералов для интерпретации электромагнитных измерений на местности, необходимо знать, что лабораторные данные не обязательно соответствуют условиям, господствующим на самом деле в Земле. Важными условиями являются состояние окисления и время, необходимое для достижения равновесия реакций между двумя твердыми телами. Рассматриваются данные электрической проводимости горных пород и минералов для верхней мантии и коры Земли. Новые исследования, рассматривающие зависимость  $\sigma$  от времени при большой температуре, а также оказанное средой окисления влияние на  $\sigma$  применяются для верхней мантии Земли. Вариация  $\sigma$  при переходе от оливина к шпинелю неизвестна из-за неизвестности окисления и загрязнения во время измерений. Находим, что  $\sigma$  альбита и базальта зависит от времени под точкой плавления; потому сомнительно предположение что  $\sigma$  возрастает в ходе частичного плавления.