



Dielectric properties of granodiorite partially saturated with water and its correlation to the detection of seismic electric signals

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ABSTRACT

Transient electric signals emitted prior to earthquake occurrence are recorded at certain sites in the Earth's crust termed sensitive. These field observations enforce the laboratory investigation of the dielectric response of rocks forming these localities. The dielectric relaxation of granodiorite rock coming from such a sensitive locality (Keratea, Greece) reveals, through complex impedance spectroscopy, that the activation volume for relaxation of this rock is negative that so far has been reported only rarely. This result, however, supports a theoretical model on the pre-seismic electric signals and is likely to be correlated with the sensitivity of the site and hence with the selectivity.

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1. Introduction

Seismic electric signals (SES) are transient electric signals recorded before an earthquake occurrence (Varotsos and Alexopoulos, 1984a, 1984b). Signals are collected by stations (consisting of electrode arrays of certain arrangement and scale) situated at “sensitive localities” in the Earth's crust. In these sensitive localities, SES signals are considered to be enhanced. Stations located at sensitive positions collect SES stemming from restricted seismic areas that may be far from the station (selectivity effect). The latter can be related with both large-scale (or “regional”) and small-scale (or “local”) heterogeneous properties of the Earth's crust (Varotsos and Lazaridou, 1991). Field observations and theoretical models indicate that in a strongly inhomogeneous area some sub-areas may provide localities, which may be sensitive to SES. One way to make a site sensitive may be that one or both electrodes are located within a structure in which SES are significantly enhanced. A typical example of such a case is Keratea station, Greece (Varotsos and Lazaridou, 1991). This station lies in a place with dykes of granodiorite, with surface width of the order of a few meters. Field observations showed that when one or both electrodes are installed on the granodiorite dyke, strong SES are observed (Varotsos and Lazaridou, 1991).

The sensitivity effect of a SES station might reasonably be related to some specific physical characteristics of the rocks underlying the station. So, it was considered highly desirable to investigate the dielectric properties of granodiorite of Keratea station, to shed light on one of

the parameters that probably constitute the sensitivity phenomenon, i.e., the role of the type of the sustrating rock that constitutes the site. In the present work, we study the dielectric relaxation properties of the granodiorite from the Keratea station, under different states of hydrostatic pressure and temperature in the laboratory by employing complex impedance measurements in the frequency domain. As it will be evident in this work, this granodiorite belongs to the class of solids that are characterized by negative activation (relaxation) volume, which is reported in the literature very rarely.

2. Effect of pressure on the dielectric relaxation

A consequence of the presence of aliovalent impurities in crystalline ionic solids is the formation of extrinsic defects, such as vacancies and interstitials, which are produced for reasons of charge compensation (e.g., see Varotsos (2007) and references therein). A portion of these vacancies and/or interstitials form electric dipoles with the aliovalent impurities (defect dipoles). The dynamics of these dipoles, when triggered by an external ‘force’ (e.g., an electric field), yield dielectric relaxation, which is determined (within the frame of the rate process theory) by the relaxation time:

$$\tau(P, T) = (\lambda\nu)^{-1} \exp\left(\frac{g^{act}}{k_B T}\right) \quad (1)$$

where P and T denote the pressure and temperature, respectively, ν is the vibration frequency of the relaxing charge carrier, λ is a geometrical constant, g^{act} is the Gibbs activation energy for relaxation and k_B is the Boltzmann's constant. Note that all thermodynamic

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quantities appearing in this paper are related to relaxation or, alternatively, to the localized motion of the electric charge species that form the relaxing ‘dipoles’. By differentiating Eq. (1) with respect to pressure at constant temperature, we get:

$$(\partial \ln \tau / \partial P)_T = -\frac{\gamma}{B} + \frac{v^{act}}{k_B T} \quad (2)$$

where $\gamma \equiv -(\partial \ln \nu / \partial \ln V)_T$ is the Grüneisen constant (V denotes the volume), $B \equiv -(\partial P / \partial \ln V)_T$ is the isothermal bulk modulus and

$$v^{act} \equiv \left(\partial g^{act} / \partial P \right)_T \quad (3)$$

is the activation volume (Varotsos and Alexopoulos (1986)).

Structural heterogeneity of rocks due to porosity and distribution in the size of grains or crystallites, combined with compositional heterogeneity stimulates interfacial polarization phenomena. Strong polarization phenomena appear when porosity is filled (partially or fully) with water (Endres and Knight, 1991; Nettelblad, 1996; Papathanassiou and Grammatikakis, 2000; Papathanassiou, 2000, 2001a, 2001b; Varotsos, 2005). Additionally, water in rocks (which is abundant in the Earth’s crust) plays a dual role: (i) it enhances the dielectric properties of the rock and (ii) induces the so-called *dilatancy* when the rock is compressed. In particular, the latter occurs in the hypocenter area of an earthquake that is surrounded by water saturated porous rock with fluid-filled pore channels and the pre-earthquake stage is assumed to be accompanied by the appearance of fresh cracks in the fracture zone (Surkov et al., 2002). The time variation of the stress field prior to an earthquake results in the dynamic compression of water-filled rocks (Morgan and Nur, 1986; Morgan et al., 1989). On account of these reports, we presume that the dielectric response of a rock results from different dynamic processes, such as defect dipole rotation, interfacial polarization, double layer polarization, etc., (Papathanassiou and Grammatikakis, 1997) which can be regarded generally as effective electric ‘dipoles’. These electric ‘dipoles’ in rocks, tend to orient themselves under the influence of polarizing fields such as the stress field, which plays a role similar to that of an external field (Varotsos, 2005). When they undergo a monotonic compression, their characteristic relaxation time is modified with increasing pressure. Eq. (2) indicates that the percentage variation of the relaxation time caused by an isothermal change in pressure is determined by v^{act} . A negative activation volume implies that the rotational mobility of electric ‘dipoles’ is enhanced on increasing pressure, i.e., the characteristic relaxation time becomes shorter on pressurization. Negative activation volumes were experimentally found earlier in a few materials, by studying the pressure dependence of the electrical conductivity (e.g., in NAFION hydrogels (Fontanella et al., 1996)) or the pressure dependence of the dielectric relaxation (e.g., in β -PbF₂ doped with lanthanum (Fontanella et al., 1982), and in semi-conducting polypyrrole (Papathanassiou et al., 2006, 2007)). Recently, negative activation volumes were found in rocks filled with water, such as leukolite (polycrystalline MgCO₃) and kataclastic limestone (polycrystalline CaCO₃) (Papathanassiou et al. (2010)). The latter finding for a natural rock is particularly important for the following reason: Varotsos, Alexopoulos and Nomicos (Varotsos et al., 1982, see also Varotsos and Alexopoulos, 1986) in order to explain the generation of seismic electric signals (SES) (i.e., the electric signals that are observed (Varotsos, 2005; Varotsos et al., 2006a, 2006b; Varotsos and Alexopoulos, 1984a, 1984b; Varotsos et al., 2003a, 2003b, 2005; Varotsos et al., 2002) before earthquakes, regarded the Earth’s crust as a solid containing ‘dipoles’ in a polarizing field (i.e., the mechanical stress field). The monotonic increase of pressure exerted on rocks characterized by negative activation volume for relaxation, during the earthquake preparation, reduces the relaxation time of these ‘dipoles’. Hence, at a certain critical pressure (smaller than that at which

rupture occurs) the relaxation time of ‘dipoles’ becomes too short and then the ‘dipoles’ undergo a transition from random-orientation state to an oriented one. The time variation of the polarization of the rock yields the emission of a transient polarization current, which is SES. This is known in solid state physics as pressure stimulated currents (PSC) (for a brief review of PSC see also Varotsos et al. (1998)).

3. Experimental setup and methodology

Granodiorite rock samples from Keratea (Greece) of thickness ranging from 1 to 2 mm and parallel surfaces of 1–2 cm² were placed inside the pressure vessel, a Novocontrol (Germany) high pressure apparatus operating from ambient pressure to 0.3 GPa and at various temperatures ranging from room temperature to 393 K. The pressurization fluid was silicone oil. Metallic electrodes were pasted on to the parallel surfaces of the specimen and a very thin insulating layer of epoxy covered the specimen to prevent contamination from the pressure transmitting fluid (Papathanassiou, 1998, 2001a, 2001b, 2002; Papathanassiou and Grammatikakis, 1996a, 1996b). Note that the use of epoxy layer to jacket the sample was also suggested independently by the manufacturer of the pressure apparatus. The pressure system was connected with an HP impedance analyzer operating from 10⁻² to 10⁶ Hz. Measurements in the frequency domain were monitored by a computer. The rock samples were partially saturated with water by immersing (and keeping) them in distilled water at 373 K for a time duration of about 3 months. The elevated temperature yield expansion of the specimen and opening of the pore cavities and, at the same time, diffusion of water molecules into the rock samples is enhanced. By weighting the sample before and after the wetting we found that the (partially) saturated rock accommodates about 0.5 wt.% water.

The tangent loss angle function $\tan \delta \equiv \text{Im}(\epsilon) / \text{Re}(\epsilon)$, where $\text{Im}(\epsilon)$ and $\text{Re}(\epsilon)$ are the imaginary and the real parts of the (relative) complex permittivity ϵ (reduced to its value of free space), was selected to study a dielectric relaxation spectra in the frequency domain. For Debye processes, the latter exhibits a maximum at a frequency

$$f_{\max, \tan \delta} = \sqrt{\epsilon_s / \epsilon_\infty} / (2\pi\tau) \quad (4)$$

where τ the dielectric relaxation time (Eq. (1)), ϵ_s and ϵ_∞ denote the static and high frequency (relative) permittivity, respectively. These values can be estimated by extrapolating the available broadband complex impedance data to the low and high frequency limits, respectively. Differentiating the natural logarithm of Eq. (4) with respect to pressure at constant temperature, we get:

$$(\partial \ln \tau / \partial P)_T = \frac{1}{2} (\partial \ln(\epsilon_\infty / \epsilon_s) / \partial P)_T - \left(\partial \ln f_{\max, \tan \delta} / \partial P \right)_T. \quad (5)$$

From Eqs.(2) and (5), we get the following formula, which permits the evaluation of the activation volume from the pressure variation of $f_{\max, \tan \delta}$ and the pressure variation of $\epsilon_\infty / \epsilon_s$:

$$v^{act} = k_B T \left\{ \frac{\gamma}{B} + \frac{1}{2} (\partial \ln(\epsilon_\infty / \epsilon_s) / \partial P)_T - \left(\partial \ln f_{\max, \tan \delta} / \partial P \right)_T \right\}. \quad (6)$$

We note that the above mentioned formalism is an approximation for qualitative analyses of the experimental data, based on the Debye approach.

4. Experimental results and discussion

A typical plot of $\tan \delta$ vs. frequency of wetted granodiorite at constant temperature and pressure is presented in Fig. 1, where a relaxation mechanism appears below the kHz region. The latter is sensitive

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