Research Article

Changes in Electrokinetic Coupling Coefficients of Granite under Triaxial Deformation

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1. Introduction

Electrokinetic phenomena occur when an electrolyte flows along charged solid surfaces. For several decades, these phenomena have been of interest to geophysicists in many subfields. Observed self-potential has been associated with geothermal fields (e.g., [1, 2]), volcanic activity and topography (e.g., [3–6]), and shallow ground water flow (e.g., [7, 8]). In numerical modelings, quantitative interpretation of self-potential observed in geothermal and volcanic areas and modelings in hydrogeophysics have been studied (e.g., [9–13]). Electrokinetic phenomena are also believed to be the most likely origin of the observed electromagnetic signals preceding or accompanying earthquakes. Mizutani et al. [14] first proposed a model: during dilatancy stage, which is assumed to precede earthquakes [15, 16], pore pressure in the dilatant region decreases and water flows into this region from the surrounding area, generating electromagnetic precursors to earthquakes due to electrokinetic phenomena.

To provide an appropriate interpretation of field observations, a better understanding of the physics of electrokinetic effect at the level of the rock-fluid interface and at the level of the rock sample is required. In laboratory experiments, zeta potential and streaming potential coefficients, fundamental quantities that characterize the electrokinetic effect, were measured for crushed rocks (e.g., [5, 17–21]) and for natural intact rocks (e.g., [22–29]) to determine the electrokinetic parameters as a function of pH, resistivity, permeability, or temperature. Jouniaux and Pozzi [23] measured the streaming potential coefficients of Fontainebleau sandstones under triaxial stress up to failure. They reported a large increase of the streaming potential coefficient beginning at about 75% of the yield stress. Yoshida [27] measured electric current and electric potential during rock deformation and found that the streaming current flowed before main failure, showing good correlation with dilatancy rate and water flow rate. In his study, however, changes of coupling coefficient (streaming potential coefficient or streaming current coefficient) during deformation were not measured.

Jouniaux and Pozzi [23] suggested that the increase of the streaming potential coefficient is due to an increase of zeta potential. An increase of the streaming potential coefficient,
however, is also caused by an increase of bulk resistivity. In this study, by measuring not only the streaming potential coefficient but also the streaming current coefficient which has different dependence on bulk resistivity, we investigate what causes changes in the coupling coefficient during rock deformation.

2. Electrokinetic Phenomena

It is well known that when in contact with an electrolyte, the surface of rock-forming minerals are charged and surrounded by an equivalent amount of ionic charge of opposite sign from the electrolyte. The overall arrangement of the electric charge on the solid surface together with the balancing charge in the bulk liquid phase is often referred to as an electric double layer. Electrokinetic phenomena are induced by the relative motion between the fluid and the rock which develops an electric double layer. Electrokinetic phenomena are induced by the relative motion between the fluid and the rock which develops an electric double layer. When the fluid in such a system moves due to a pressure gradient, the charges in the fluid are transported in the direction of fluid motion, resulting in an electric current. In a porous medium the current density $i$ (in A/m$^2$) and fluid flux $j$ (i.e., flow velocity, in m/s) are described by the following relations [30, 31]:

$$
\begin{align*}
    i &= -\frac{\sigma_f + \sigma_s}{F} \text{grad} \phi + \frac{\epsilon \zeta}{\mu F} \text{grad} P_p, \\
    j &= \frac{\epsilon \zeta}{\mu F} \text{grad} \phi - \frac{k}{\mu} \text{grad} P_p,
\end{align*}
$$

where $\sigma_f$ and $\sigma_s$ are the electrical bulk and surface conductivities, $\epsilon$ is the dielectric constant of the fluid, $\zeta$ is the zeta potential (the potential at the slipping plane near the boundary), $k$ is the permeability, $\mu$ is the viscosity of the fluid, $P_p$ is the pressure of the fluid, and $\phi$ is the streaming potential. The first term of (1) represents Ohm’s law, and the second term represents streaming current which can be derived by considering the product of the charge density (proportional to $\epsilon \zeta$) with the flow velocity of the viscous fluid (proportional to $\text{grad} P_p/\mu$). The first term of (1) shows the macroscopic conductivity of rock (reciprocal of bulk-resistivity), which is expressed as

$$
\sigma_R = \frac{\sigma_f + \sigma_s}{F}. 
$$

Considering the capillary model [17, 32], we define formation factor $F$ as

$$
F = \frac{\sigma_{\text{eff}}}{\sigma_R} = \frac{T^2}{\eta},
$$

where $\eta$ is the porosity, $T$ is the tortuosity, and $\sigma_{\text{eff}}$ is the effective conductivity defined as

$$
\sigma_{\text{eff}} = \sigma_f + \sigma_s.
$$

The surface conductivity $\sigma_s$ (in S/m) is related to the specific surface conductance $\Sigma_s$ (in S) by $\sigma_s = 2 \Sigma_s/m$, where $m$ is the hydraulic radius for the capillary model. The permeability of the capillary model is represented as

$$
k = \frac{\eta m^2}{T^2},
$$

where $b$ is a constant related to the shape of pore; $b = 8$ for capillaries with a circular cross-section. We refer to $-i/\text{grad} P_p$ under grad $\phi = 0$ in (1) as the streaming current coefficient $C_e$:

$$
C_e = \frac{\epsilon \zeta}{\mu F} = \frac{\eta \epsilon \zeta}{T^2 \mu}.
$$

Both $C_e$ and $k$ are functions of the fluid path network, and the dependence of $C_e$ and $k$ on $\eta$ and $T$ are the same. However, dependencies on $m$ are different for $C_e$ and $k$. The difference by $m^2$ can be understood if we note that the volume flow rate of a viscous fluid through a tube is proportional to the square of the cross-sectional area of the tube, while the amount of the transport electric charges distributed along the boundary are proportional to circumference length (i.e., proportional to the radius) and the flow velocity around the boundary is also proportional to the radius.

If there are no external current sources and no leaking current, the streaming current (due to $\text{grad} P_p$) would be balanced by the conduction current (due to $\text{grad} \phi$), so

$$
\Delta \phi = \frac{\epsilon \zeta}{\sigma_{\text{eff}} / \mu} \Delta P_p,
$$

which is the Helmholtz-Smoluchowski equation. The ratio $\Delta \phi / \Delta P_p$ is referred to as the streaming potential coefficient,

$$
C_p = \frac{\Delta \phi}{\Delta P_p} = \frac{\epsilon \zeta}{\sigma_{\text{eff}} / \mu} = \frac{C_e}{\sigma_{\text{eff}} / F} = \frac{C_e}{\sigma_R}.
$$

In general geometry, divergence of the total current is zero, but the zero total current condition (7) is not always satisfied.

3. Experimental Methods

In this study, we measured the streaming current (or streaming potential), permeability, and dilatancy of the rock specimen simultaneously and continuously during rock deformation test. We used the triaxial apparatus which was specially designed to investigate the electrical behavior during rock deformation and failure [27]. In this apparatus, the rock specimen is electrically isolated from the surroundings by inserting alumina plates. The pore fluid tubes of stainless steel inside the vessel are also isolated from the outside fluid tubes by using insulating tubes through the vessel closure as shown in Figure 1. This apparatus has two options for force loading: hydraulic loading with servo valves and a screwed pump with a servo motor. In the present experiment, we used the screwed pump for deformation test at a strain rate of approximately $10^{-7}$/s. This apparatus is equipped with up to 11 feedthroughs that Nishizawa [33] developed on the basis of Bridgeman’s self-sealing mechanism. During the
Figure 1: (a) Schematic diagram of apparatus. A rock specimen is electrically isolated by inserting alumina plates and by using insulating tubes of pore water through vessel closure. (b) Picture of the specimen after experiment. A failure plane is indicated by black arrows.

In the present study, we used coarse grained Inada granite (from a locality in Ibaraki, Japan), which has been often used as a standard specimen in rock mechanics (e.g., [27, 29, 34]). We used two specimens which were cored cylindrically 24 mm in diameter and 60 mm in length. One specimen was used in the electric potential measurement and denoted G01. The other was used in the electric current measurement and denoted G02. The porosity of both specimens was approximately 1%. The rock specimens were air-dried and degassed under a vacuum for 12 hours before being saturated with $10^{-3}$ M KCl solution for 2 days. The conductivity of the fluid was 14 mS/m. Then, the specimens were placed between the stainless steel end plugs and jacketed in a Teflon sleeve.

To measure the axial strain $\varepsilon_a$ and the circumferential strain $\varepsilon_c$, strain gauges were mounted at four positions on the cylindrical surface of the Teflon sleeve. A volume change of the specimen $\Delta$Vol is estimated as $\Delta$Vol = $\text{Vo}_0 (2\bar{\varepsilon}_c + 2\bar{\varepsilon}_a)$, where $\bar{\varepsilon}_c$ and $\bar{\varepsilon}_a$ are averaged strains for four positions, and $\text{Vo}_0$ is the initial volume of the specimen. The cross-section of the specimen is assumed to remain circular. This assumption is not satisfied when a fault plane is formed and large localized deformation occurs. A volume change due to dilatancy is obtained by subtracting elastic deformation from the volume change.

To measure the permeability continuously during the deformation experiments [35], we adopted the sinusoidal oscillation method [36–39]. The method is based on the measurement of an attenuation and a phase retardation of an oscillation of the pore-fluid pressure as it propagates through the specimen. In its application, a sinusoidal pressure oscillation is imposed at one end of the specimen and a pressure response is monitored at the other end as illustrated in Figure 2. The permeability is calculated from the measured attenuation factor $R$ between downstream and upstream pore-fluid-pressure sinusoidal waves, and the phase lag $\delta$, using the following relation (calculations detailed in [37]),

$$P_{p1} = RP_{p2} \sin(\omega t - \delta),$$

$$R \cdot \exp(i\delta) = \frac{1}{\cosh[\psi(1+i)] + i \psi(1+i) \sinh[\psi(1+i)]},$$

where $P_{p1}$ and $P_{p2}$ are upstream and downstream pore fluid pressure, respectively, $R$ is the attenuation factor, $\delta$ is the
The streaming current coefficient involves bulk resistivity and the source potential. By reading the amplitude of the sinusoidal variation of the electric current (ΔI) and the amplitude of the sinusoidal variation of the fluid pressure difference (ΔPp2 = Pp1 − Pp2), we evaluated the generated current per unit pressure change in the fluid pressure ΔI/ΔPp. The streaming current coefficient Cc was obtained using the relation

\[ C_c = \frac{\Delta I}{S \Delta P} = \frac{\Delta I}{L \Delta P} \cdot \frac{1}{S} \quad [\text{A/mPa}]. \tag{11} \]

Similarly, from measurements of the amplitude of electric potential variation (ΔΦ), we estimated the streaming potential coefficient as

\[ C_p = \frac{\Delta \Phi}{\Delta P} \quad [\text{V/MPa}]. \tag{12} \]

Frequency effect on the coupling coefficient [40–42] for the Inada granite with the present experiment system has been reported in [27]. Coupling coefficient does not depend on frequency in such a low frequency range (0.01–1 Hz) for the intact Inada granite. Although we cannot rule out the possibility of changes of frequency dependence on the coupling coefficient during deformation, we focus on the continuous measurement during deformation to fix the frequency to 0.01 Hz and do not discuss the frequency dependence in the present study.

4. Results

Here, we show the results of the two experiments. Experimental conditions for G01 and G02 were the same. The confining pressure Pc was kept at 15 MPa. The pore-fluid pressure at each end of the specimen was set to 5.3 MPa at the beginning of the experiment. Then, pore fluid pressure of the bottom face of the specimen was sinusoidally oscillated at a frequency of 0.01 Hz with an amplitude of 0.5 MPa throughout the rest of the experiment. Experiments were conducted under room temperature (25 ± 1°C).

Figure 3 shows the results of the run G01, in which electric potential was monitored. The axial loading rate was 5.1 × 10⁻²/s. A shear failure plane was found in the postexperimental sample (Figure 1(b)). From Figure 3(a) showing the differential axial stress and the displacement, it can be seen that dynamic failure occurred at t = 37, 323 s. Some small releases of axial stress occurred around t = 6,000 and 17,000 s. These small stress releases may be due to poor initial setting of the apparatus. Thus, we do not analyze the data before these stress changes (t ∼ 20,000 s). The pore-fluid pressure Pp1 and Pp2 are shown in Figure 3(b). The pore-fluid pressure of the bottom face of the specimen appears to be a thick line in this scale because it is sinusoidally oscillated at a frequency of 0.01 Hz and amplitude of 0.5 MPa. Figure 3(e) shows the volume change of the specimen obtained from strain measurements and the dilatancy calculated by subtracting the elastic deformation from the volume change. In Figure 3(b), the volume change
Figure 3: Result of deformation test for initially intact Inada granite (G01) at $P_c = 15$ MPa, $P_p = 5$ MPa. (a) Differential stress and displacement. (b) Volume change of the specimen obtained from the average of the strain measurements at four positions, dilatancy, pore fluid pressures, and water volume. (c) Streaming potential (SP) and pore-fluid pressure difference between bottom and top faces of the specimen. (d) Axial stress versus axial displacement. In this experiment, failure stress was 324 MPa and Young’s modulus was 32 MPa. (e) Volume change of the specimen and dilatancy versus the axial stress. The elastic deformation is indicated by the thick black line in this figure.
and the dilatancy are plotted. When dilatancy began (around \( t = 27,000 \) s), the downstream pore pressure \( P_{p1} \) began to drop, indicating that the pore pressure in the specimen dropped and water flowed into the specimen. From the change of \( P_{p1} \), we calculated the water volume \( Q_1 \), which flows into the specimen from the downstream reservoir, as \( Q_1 = -\beta d (P_{p1} - P_{p0}) \), where \( P_{p0} \) is an initial pore pressure. Although we attempted to estimate the water volume \( Q_2 \), which flows into the specimen from the upstream side, using the displacement of the piston of the pore water.
intensifier, we could not estimate $Q_2$ due to the leakage of the water at the upstream side. Therefore, we show only $Q_1$ in Figure 3(b). In Figures 3(b) and 3(c), we can see good correlation among the trends of dilatancy, the pore pressure difference, and the streaming potential (SP). Details of this “DC” relation are discussed later. Here, we focus on the results of “AC” measurement based on the imposed sinusoidal oscillation of the pore pressure.

Figure 4 shows the other run (G02) in which the electric current was measured. A similar result with the run G01 was obtained, showing good correlation among dilatancy, pore pressure difference, and the streaming current (EC). The axial loading rate was $5.5 \times 10^{-7}/s$. As in G01, a discrete shear plane was found in the postexperimental sample, indicating that the main failure (around $t = 35,788$ s) involved the formation of such a failure plane. When dilatancy began (around $t = 28,650$ s), the pore pressure of the downstream $P_{p1}$ began to decrease. Some small stress releases were also observed at about $t = 6,000$ and $22,000$ s due to the setting of the apparatus similarly to the former experiment.

Figure 5 shows the permeability of G01 and G02. The permeability was initially of the order of $10^{-18}$ m$^2$. With the increase of the axial loading, the permeability decreased to $10^{-19}$ m$^2$. Then, just before the failure, the permeability increased to $\sim 10^{-18}$ m$^2$ in the both experiments. There are a lot of studies dealing with permeability-porosity relationship and permeability-stress relationship (e.g., [43–45]). In our experiment, permeability reduction is approximately an exponential function of effective mean stress [45] and mainly attributed to elastic crack closure [43]. The permeability increase indicates enhanced connection of cracks. Although the dilatancy should involve the creation of microcracks, the permeability continued to decrease with progressive loading, indicating that microcracks were not fully interconnected or not fully saturated with pore fluid.

5. Discussion

The coupling coefficients $C_p$ (or $C_c$) were calculated from amplitude of pore pressure difference and potential (or current). The polarity of obtained coupling coefficients were negative in the present experiments, indicating negative zeta potential, as expected for granites. The values of the coupling coefficients are shown in absolute values hereafter. Figure 6 shows the coupling coefficients of G01 and G02. To remove the data which are not suitable for calculating streaming potential coefficient, we evaluated a signal quality by $P(0.01 \text{ Hz})/\sum P(f)$, where $P(f)$ is the power spectrum of potential variation. The data with the signal quality lower than 0.98 were not used. We can see variations of $C_p$ around
Figure 7: Normalized stress, permeability, dilatancy, and coupling coefficient versus time. The gray, red, and blue lines indicate the stages A, B, and C, respectively. Three stages which are divided by the beginning of dilatancy (G01 \( t = 27,000 \) s, G02 \( t = 28,650 \) s) and the beginning of permeability increase (G01 \( t = 36,000 \) s, G02 \( t = 34,750 \) s).

\( t = 6,000 \) and \( 17,000 \) s in Figure 6(b). These variations were due to small stress releases resulting from the setting of the apparatus as mentioned before. A fluctuation of \( C_c \) around \( t = 22,000 \) s was due to the same reason. The streaming potential coefficient \( C_P \) decreased with loading (Figure 6) and then increased by a factor of two at the onset of dilatancy (around \( t = 27,000 \) s). Note that \( C_P \) did not continue to increase with dilatancy increase but \( C_P \) increased just at the onset of the dilatancy. In contrast, the streaming current coefficient \( C_c \) continued to decrease during the loading until the time of failure, not particularly affected by dilatancy. It is noted that the observed change in \( C_c \) indicates that the source current density did not increase during the deformation, and therefore observed increase in \( C_P \) is attributed to bulk resistivity (see (8)).

The estimation of the zeta potential is done from the measured streaming current coefficient. The streaming current coefficient \( C_c \) of Inada granite before loading is approximately \( 7 \mu A/\text{mMPa} \). The formation factor \( F \) of Inada granite under atmospheric pressure was estimated to be 1100 from the measurement of the resistance of the rock sample saturated with KCl solution with a high conductivity (0.2–1.1 S/m). Inserting these values into (6), we obtained the zeta potential as \(-11 \) mV, which is slightly smaller than the previously reported value of granite [17, 18, 29].

To investigate the evolution of the coupling coefficients in detail, we divide experiments into three stages; stage A: from the start of experiment to the beginning of dilatancy, stage B: from the beginning of dilatancy to the beginning of permeability increase, stage C: from the beginning of permeability increase to the failure. Figure 7 indicates these stages in different colors. We defined the normalized stress as the stress normalized by the failure stress. Figure 8(a) shows the streaming potential coefficient \( C_P \) and the dilatancy of G01 as a function of the normalized stress. We can see that the dilatancy and increase of streaming potential coefficient began at 47% of yield stress. Figure 8(b) shows the relation between the streaming potential coefficient and the dilatancy. Increase of the streaming potential coefficient (5 V/MPa to 10V/MPa) occurred at the onset of the dilatancy. Figure 8(c) shows \( C_c \) and the dilatancy of G02 as a function of the normalized stress. We can see the dilatancy began at 58% of the failure stress. The streaming current coefficient \( C_c \) continued to decrease at an approximately constant rate unrelated to the dilatancy. Relation between the \( C_c \) and the dilatancy is shown in Figure 8(d). When the permeability increase (stage C) began, \( C_c \) stopped to decrease and remained roughly constant during the stage C up to failure.

Figure 9 shows the relation between the streaming current coefficients and the permeability. The streaming current
coefficient $C_c$ was approximately proportional to the square root of the permeability. This dependence can be explained by assuming that $m^2$ is proportional to $1/F = \eta/T^2$ in (5) and (6) for the capillary model [17, 27, 32]. This assumption is supported by the experimental results that $\log k$ is linearly related to $\log F$ with slope of $\sim -2$ for granite reported by Walsh and Brace [32].

The fact that the streaming current coefficient did not increase indicates that the zeta potential did not increase throughout the deformation test. Furthermore, there is a possibility of decrease of the zeta potential, because $C_c$ does not increase with the permeability increase in stage C. If bulk resistivity increases at the onset of the dilatancy, the streaming potential coefficient $C_p$, which is the product of $C_c$ and bulk resistivity (see (8)), will increase. Figures 3(b) and 4(b) show the volume of water flow from the downstream $Q_1$, which is much smaller than dilatancy. The ratio of $Q_1$ to dilatancy is approximately 0.1 to 0.2, indicating the possibility of the undersaturation of the pore, although the water flow from the upstream is not included. We mention the possibility of bulk-resistivity change here. To understand the observed change of $C_p$, we would require measurements of the bulk-resistivity changes during the deformation. On the basis of recent studies (e.g., [46–48]), however, the magnitude of $C_p$ decreases with decreasing water saturation $S_w$ in most situations even though substantial increase of bulk-resistivity takes place at the same time.
Figure 10: The streaming potential (SP) and the pore pressure difference (Pdiff). Long-term trend of these values are used in the “DC” measurements of electrokinetic phenomena (a). Amplitude and phase of the sinusoidal variation caused by imposed pore pressure oscillation are used for the “AC” measurements of electrokinetic phenomena (b). The DC coupling coefficients and the dilatancy as a function of the pressure difference for G01 (c) and G02 (d). Blue and red lines indicate coupling coefficients and dilatancy, respectively.
The $S_v$ dependence of the coupling coefficients is important also for modeling of field self-potential data related to unsaturated flow in volcanic areas (e.g., [49, 50]) and shallow groundwater systems. We need further study to clarify the $S_v$ change during dilatancy stage and its effect on the coupling coefficients of low-permeability rocks such as granite used in this study.

Here we compare the coupling coefficients obtained from the AC measurements and those obtained from the DC measurements. We can see the DC electrokinetic effect in Figure 10(a). Figure 10(b) shows the AC electrokinetic effect discussed earlier. The DC coupling coefficients and the dilatancy are shown as a function of the pressure difference in Figures 10(c), 10(d). The dilatancy showed a linear relation with the pressure difference in both experiments. The magnitude of the DC streaming current coefficient $C_c$ was approximately 6 $\mu$A/mMPa before dilatancy began and then decreased to 2 $\mu$A/mMPa after dilatancy began. These values agree well with the $C_c$ obtained from the AC measurement (see Figure 7). On the other hand, the magnitude of the DC streaming potential coefficient $C_p$ during dilatancy was approximately 30 mV/MPa, which was three-times as large as that from the AC measurement (see Figure 7). At the present stage, we do not fully understand the discrepancy between AC and DC streaming potential coefficients. We need further study including a frequency dependence of the specific resistivity of the rock specimen.

6. Conclusions

Jouniaux and Pozzi [23] suggested that the onset of increase in the streaming potential coefficient corresponded to the onset of shear localization and that this increase was due to an increase of the zeta potential in the shear zone as new surfaces were created and connected. Although Jouniaux and Pozzi [23] suggested a possibility of the enhancement in the streaming potential coefficient, which was the product of the streaming current density and the zeta potential of the specimen. Therefore, one cannot deny a possibility that the observed increase of the streaming potential was due to an increased bulk resistivity rather than an enhanced zeta potential.

In our experiment, the $C_p$ increased but the $C_c$ did not increase, indicating that the source current density did not increase during the deformation. Such an increase in $C_p$, due to increase in bulk resistivity cannot be the source of the electric signals unless the increase in bulk resistivity occurs broadly in the observation field. Variation of the zeta potential according to the deformation stage makes it difficult to interpret the self-potential data quantitatively. Results of the present experiments, however, indicate the zeta potential does not vary so much throughout all the deformation stage of the rock up to failure.

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