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Comments on the phenomena underlying pressure stimulated currents in dielectric rock materials

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Abstract It has been observed that there is a systematic detection of weak pressure stimulated electric currents (PSC) in polycrystalline and amorphous solids during or after an application of mechanical stress upon them. An interpretation of the mechanisms governing the electrical behaviour of solid structures when they undergo uniaxial stress tests is presented. In numerous experiments, repetitive stress loadings and unloadings have been conducted, the corresponding PSC have been recorded, and the behaviour of the material is interpreted with respect to PSC emission. The dominant conclusion is that the behaviour of the solid depends on the overall stress it has suffered and that PSC is related with a memory effect which associates current emission with the previous history of the sample.

Introduction

The material fracture phenomena, particularly those concerning inhomogeneous materials, such as geomaterials, in association with transient electric phenomena, have attracted the interest of the scientific community for a long time. The main reason for this interest is that such phenomena can be associated with probable earthquake prediction techniques and methodologies [1–3]. During the deformation process of a geomaterial and particularly in the range where the material shows a deviation from linearity in terms of mechanical behaviour, microcracking occurs. In this range, the material appears damaged and irreversible deformations take place. Experiments conducted under laboratory conditions have shown that similar deformations produce electrical phenomena. Electric signal generation and emission mechanisms appear and a number of researchers acknowledge such mechanisms as related to crack generation and propagation in the Earth's crust [4, 5].

For a number of years, scientists worked to establish a well accepted relation between mechanically induced major disturbances in solids and the variation of electrical parameters. [1, 2, 6–9].

A large number of laboratory experiments have been conducted on minerals and rocks (both dry and water-saturated) in order to understand and interpret the nature of the mechanisms responsible for the production of such electrical signals [10–12]. The most recent of them employed an experimental arrangement suitable for recording the pressure-stimulated current (PSC) emitted by geomaterial samples during the application of either an abrupt uniaxial stress increase or a monotonically increasing stress up to sample fracture [11, 13–16]. It was confirmed that marble and cement mortar samples under the above conditions produced weak electric currents (PSC). The experimental procedure followed is described by the term PSC technique.

Furthermore, there are numerous studies and recordings of acoustic emissions due to microcrack opening in

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rocks and other materials during mechanical stress application as well as of memory effects [17–22].

In the present work, a reference is made to the numerous experiments of repetitive stress loadings and unloadings which confirmed that the behaviour of the solid with respect to stress depends on the overall stress it has suffered, i.e. there is a memory effect which associates current emission with the history of the sample. This observation could be of significant importance due to its possible applications in material fatigue testing.

Sample characteristics and experimental setup

In this experiment, Dionysos marble samples collected from Mt. Penteli, Attica, were used. Dionysos marble is mainly composed of calcite (98%) and other minerals, such as muscovite, sericite and chlorite. Its content in quartz is very low, about 0.2%. Its density is 2.7 g/cm^3 and its porosity is approximately 0.4%. The geometric dimensions of the prismatic samples were 69.6 mm × 49.0 mm × 51.2 mm while the average fracture limit was measured to be in the range from 50 MPa to 60 MPa. The experiment was conducted in a Faraday shield to eliminate electrical noise. The stressing system comprised a uniaxial hydraulic load machine (see Fig. 1) that applied compressional stress to the sample which was placed on a stainless steel base. The marble sample was placed between two thin teflon plates in the direction of stress to provide electrical insulation. For conducting the PSC measurements, two electrodes were attached in a direction perpendicular to the axis of the applied stress and a sensitive programmable electrometer Keithley 617 was used.

Experimental results and discussion

In the present work repetitive equilasting but not necessarily equispaced ramps of uniaxial compressional stress were applied to marble samples of the form depicted in Fig. 2a. The magnitude of the applied stress peak was approximately 45 MPa. The frequency of repetitive loading—unloading was such as to permit a considerable time to elapse between two successive loadings in order that the material could have an adequate time to relax. Preliminary measurements indicated that the time required by the PSC of marble samples to get to quiescent levels comparable to noise is shorter than 200 s. Thus, stress ramps corresponding to triangular pulses were applied to the samples. Between the final edges of the fall of each stress pulse and the rise threshold of the next pulse there is a quiescent time of approximately 250s for relaxation at 12 MPa. As a result of this triggering, PSC peaks were recorded as shown in Fig. 2b.

During the application of repetitive equilasting abrupt steps of uniaxial compressional stress deep in the plastic range of the stress–strain curve of the material, the phenomenon of pressure stimulated current (PSC) has been observed several times. In

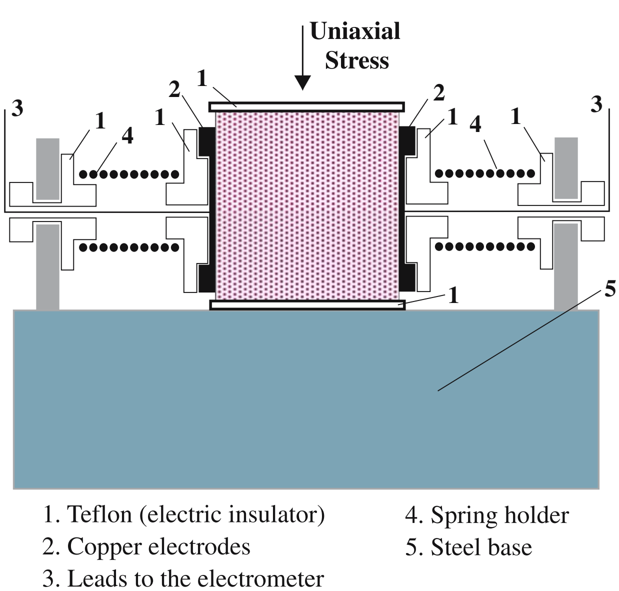

Fig. 1 Diagram of the hydraulic load machine with the peripheral connections

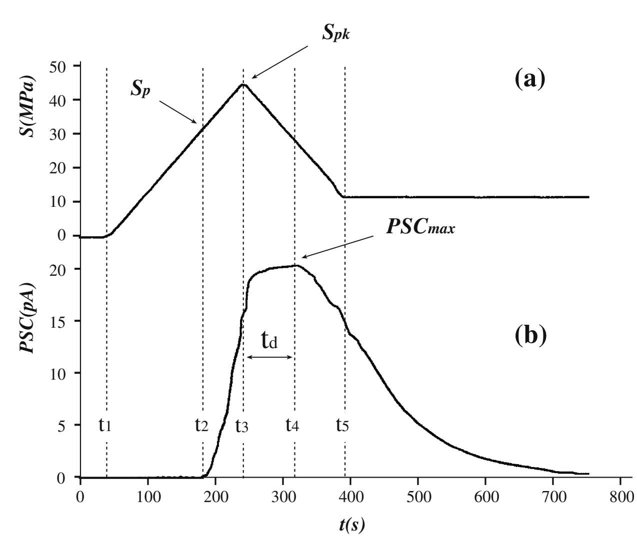

Fig. 2 Diagram showing the correlation of the initial mechanical loading pulse (**a**) and the corresponding PSC peak (**b**)

the case of the initial smooth progressive loading, four characteristic observations based on Fig. 2b should be mentioned:

While the stress ramp is applied at t_1, the PSC makes its initial appearance after a considerable time lag at S_p (t_2), i.e. until after the stress has reached a value ≈ 0.65 S_{max} where S_{max} is the maximum applied stress before sample fracture [11].

The maximum compressional stress S_{pk} appears at t_3 while the PSC continues increasing between t_3 and t_4 at a much lower rate. The PSC peak appears at PSC_{max} with a delay $t_d = t_4 – t_3$ from the compressional stress peak (S_{pk}) during the gradual removal of the stress. The PSC_{max} occurs at the instant t_4.

The PSC discharge current continues its gradual relaxation although stress has been stabilized to a final minimum value of 12 MPa.

These observations may be explained in the following manner: When a material sample suffers uniaxial compressional stress its crystalline or amorphous lattice undergoes a major disturbance which, if the stress persists will ultimately lead the whole system to irreversible changes and destruction. In the beginning some electron trajectories may be affected and become reorientated, resulting in multiple local polarizations. However when adequate energy depending on the kind of material-is transferred to the lattice the chemical bonds break, dislocations are introduced and the system collapses. During the initial compression, when stress reaches values corresponding to the early plastic range of the material at t_2, i.e. ≈ 0.65 S_{max}, microcracks occur. Those microcracks occur at the weakest points of the material which undoubtedly are point defects and discontinuities such as grain boundaries. In rock-like materials such as marble and amphibolite, discontinuities of both of these kinds are the rule rather than the exception.

In any case, the whole process of compression results through various channels in mass displacement of charged microparticles, which automatically constitutes electric current and is recorded as PSC. If groups of particles (molecules) change their state of motion, thus consuming energy, in order to become separated from other groups of particles and cause the formation of microcracks, they do so under the influence of an externally applied force (compressional stress). From the above it becomes easily accepted that the number of activated charge carriers is a measure of the energy conveyed to the material sample for microcrack generation.

The sudden separation of two grains in contact destroys the electrical equilibrium, producing electrostatic charges at the grain boundaries which are separated by an elementary potential difference. Deeper within the material this fact corresponds to a lattice disturbance which may result in temporary local polarization due to reorientation of the electron trajectories and consequent charge motion, namely PSC generation.

After a relatively short time some charges move irrespectively of the mechanical state of the material sample in order to re-establish the lost equilibrium, thus producing a weak electric current in addition to the initially recorded PSC and which is emitted to establish a new equilibrium. This electric relaxation, from the mechanical point of view, corresponds to partial closing or—to be more exact—to an infinitesimal approach of the edges of the cracks, which establishes the new equilibrium state.

In addition to such particle motions, the material is subjected to continuing strain at a very low rate, as stress is still present. The few new microcracks that go on appearing produce new microcurrents and result in conserving PSC at relatively high values that do not permit a direct relaxation to noise level. The slight increase in PSC following S_{pk} can be attributed to a superposition of the above described currents.

After 750s of activity of stress application several courses of repetitive stress ramps were carried out with the results depicted in Fig. 3, where four sequential current peaks can be distinguished and the following new observations can be mentioned:

The generated PSC seems to decrease gradually after each stress cycle, and this phenomenon is so characteristic that although six loading/unloading cycles were performed, the emitted PSC peaks corresponding to the 5th and 6th loading ramps were

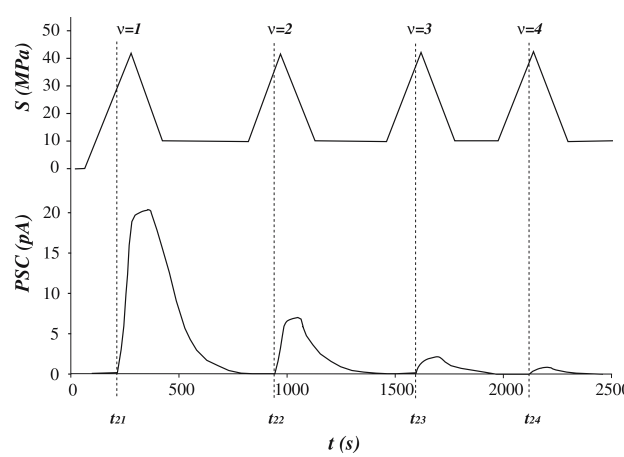

Fig. 3 The relationship between PSC and uniaxial compressional stress ramps applied in repetitive cycles upon a geomaterial sample. The initial appearance of PSC in each cycle corresponds to a larger stress

negligible and are not presented in the diagram. It becomes clear that the sample exhibits a decreasing sensitivity in producing PSC after each stress cycle. Thus, it is shown experimentally that there is a measurable change in the behaviour of the material depending on its mechanical stress history, in other words a "memory effect" is present. Figure 4 depicts the experimentally recorded PSC peak exponential decrease rate. After computer fitting the results, it seems that the depicted curve follows an exponential law of the form:

$$PSC_{\max}(v) = C \cdot \exp\left(-\frac{v-1}{\lambda}\right), v = 1, 2 \ldots, \kappa \tag{1}$$

where λ is a relaxation constant, v is the number of loading/unloading cycles and C is a pre-exponential constant approaching the value of PSC_{max} (1).

The application of a new compressional stress cycle of the same characteristics causes the removal of the edges to their previous excitation positions resulting in a smaller number of new microcracks to which the smaller peak values of PSC may be attributed. At the same time, some of the existing dislocations are replaced by partially grown neighbouring dislocations corresponding to smaller energy values in order to meet the requirements of thermodynamics. If no new microcracks were to be formed during each reloading, then, the solid would not suffer fatigue and it would not be aged, so, it would never be fractured. It would be elastic even in the plastic range and this is clearly a false conclusion.

The initial appearance of the PSC (at the instant t_2 in Fig. 3) corresponding to the next and the following stress cycles takes place for a greater stress magnitude during the rise of the ramp, i.e. the value of S_p becomes larger for each new cycle. It may also be seen in Fig. 3 that the starting points of the current curves following the initial tend to approach temporally the corresponding stress peaks.

Figure 5 depicts the expected stress S_p values for the PSC to appear with respect to reloading cycles. After

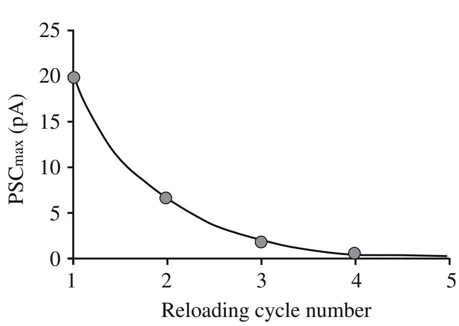

Fig. 4 Decrease rate of PSC peaks

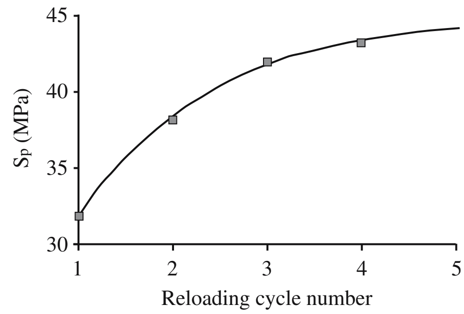

Fig. 5 Expected stress S_p values for the PSC to appear initially with respect to reloading

computer fitting an exponentially increasing relation appears having the form:

$$S_p(v) = S_{p1} + S_{p2}\left[1 - \exp\left(-\frac{v-1}{\mu}\right)\right], v = 1, 2 \ldots, \kappa \tag{2}$$

where μ is a relaxation constant and the parameters take the following values:

S_{p1} = 31.9 MPa, S_{p2} = 12.8 MPa and μ = 1.4

There is a time delay in the appearance of current after a mechanical excitation of the material sample by a stress ramp. In the second and following cycles of reloading it becomes obvious that there is a delay in the initial appearance of the PSC with respect to the previous cycles as can be read from the magnitude of stress applied. There is also a delay in the appearance of PSC_{max} with respect to the maximum compressional excitation. This is due to the fact that distortions in the solid are neither created nor do they propagate instantly but take some time. The structure and consequently the strength of the material will facilitate or make it difficult for the distortions to travel within its lattice, thus it is very probable that such a hysteresis is characteristic of the nature of the material.

These facts are interpreted as follows: Upon re-application of stress, the edges of the existing cracks tend to separate for one more time but no current flows since no new microcracks are created initially. As the procedure goes on in the same way and the same stress magnitude is reached, fewer new microcracks appear resulting in a new cycle of current flow which is weaker than before.

Conclusions

Prismatic marble samples were subjected to sequential uniaxial compressional stress ramps of identical magnitude and weak electric current emissions originating from the lattice distortion were detected.

It was found that material samples that had been subjected to stress previously emit weaker PSC than unstressed samples which are initially stressed, thus, PSC emissions can give a measure of the fatigue and ageing of a material sample.

Memory phenomena (Kaiser effect) that have appeared in acoustical emissions are also present in the processes of loading and unloading material samples through pressure stimulated current emissions.

The experimental results indicate that the values of PSC peaks during loading–unloading procedures follow an exponential decrease law as the number of loading–unloading cycles increases. This could be a substantial criterion of ageing and fatigue of the sample because the used and damaged samples emit smaller PSC than those that have never suffered stress.

This is due to the fact that for every new equiloading cycle, fewer microcracks are created than those created during the previous one. This indicates an ambiguous extension of the elastic range of the material.

In general, such results show up a measurable differentiation in the behaviour of the material both electrical and mechanical after subjection to stress. The fact that the appearance of PSC with respect to work-hardened material requires more and more stress following a seemingly exponential decrease rule as indicated in Fig. 5, indicates that the material sensitivity to external excitations decreases i.e. its history is memorized and one of the keys to the decryption code is the PSC measurement.

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