Dielectric characteristics of marble rocks after the application of various stress modes before fracture

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Abstract: - Studies of dielectric relaxation and ac conductivity of marble samples collected from Mt. Penteli (Attica) have been carried out in the frequency range of 150Hz to 1MHz. Initially reference measurements were conducted under atmospheric pressure for all of the samples. Subsequently the samples were subjected to various compressive stresses of approximately 300s duration each. Then, the stress was removed and dielectric constant and loss measurements were conducted. The experimental results indicate a large permanent differentiation a long relaxation time if the applied stresses are beyond the elasticity limit and the material is stressed in the plastic region.

Key-Words: - Dielectric Spectroscopy, marble rocks, permittivity, conductivity, axial stress.

1 Introduction

Marble is a rock, which from a geological point of view belongs to the metamorphic rocks. Imperfections in its structure are usually due to either internal or external factors such as mechanical strain, chemical or physical processing and play an important role in the behaviour of the material. Mechanical stresses upon rocks create microscopic or macroscopic discontinuities resulting in changes in the mechanical behaviour of the material[1-3].

Dielectric properties relate to the ability of a material to polarise under the influence of an electromagnetic field. The polarisability of a material depends on the structure and molecular properties and therefore dielectric measurements can provide information in this respect. The technique for measuring dielectric properties is known as Dielectric Spectroscopy[4-7].

The objective of this work is to investigate microscopic and macroscopic discontinuities and cracks using dielectric spectroscopy. This method was applied on marble samples collected from Mt. Penteli, Attica, Greece.

Dielectric measurements have proved to be a valuable tool in the study of relaxation behaviour. One of the advantages of the technique is ease of frequency variation during measurements. Dielectric spectroscopy measures complex relative permittivity of materials over a wide frequency range. Complex relative permittivity ε^* (hereafter, referred to as complex permittivity for convenience) is defined as:

$$\varepsilon^* = \varepsilon' - j\varepsilon'' = \varepsilon' + \frac{\sigma}{j\varepsilon_0\omega} \tag{1}$$

where ε' and ε'' are the real and imaginary parts of ε^* respectively, σ is conductivity, ε_0 is vacuum permittivity, ω is the angular frequency ($\omega = 2\pi f$) and $j = \sqrt{-1}$. The real part, ε', of the complex permittivity and conductivity, σ, of materials, though not always, show dielectric relaxation or dielectric dispersion in which ε' decreases and σ increases with increasing frequency. The ratio of the imaginary part of ε^* to the real part $\varepsilon''/\varepsilon'$ is defined as the loss angle tangent $\tan\delta$ or dielectric loss :

$$\tan\delta = \varepsilon''/\varepsilon' \tag{2}$$

Jonscher suggested[8] that the widely observed power law frequency dependence of ac conductivity is a “universal” property of materials. In general ac conductivity $\sigma(f)$ is found to vary with frequency f as

$$\sigma(f) = \sigma(0) + Af^n \tag{3}$$

at frequencies well below the lattice vibrational frequency. In equation (3), $\sigma(0)$ is the dc limit of conductivity, A is a parameter depending on temperature and pressure and the exponent n takes values between 0 and 1.

2 Materials and experimental technique.

Marble samples collected from Mt. Penteli, Attica (Dionysos marble) is the kind of marbles that have been typically used since the ancient times for the construction of artifacts and monuments. It is mainly composed of calcite (98%) and other minerals depending on the variety of the marble, such as muscovite, sericite and chlorite[9]. Its content in quartz is very low, about 0.2%. Its density is 2.7 gr/cm^3 and its porosity is approximately 0.4%. This fact suggests that Penteli marble is a geomaterial of low porosity. Calcite crystals are equisized and polygonic, sometimes exhibiting twinning. Their texture may be characterized as quasi-homoblastic. The rock is white with a few thin parallel ash-green colored veins containing silver-shaded areas due to the existence of chlorite and muscovite. In this work the marble samples were provided in the form of specimens of thickness t=4-5mm and cross-section A=400mm^2 approximately.

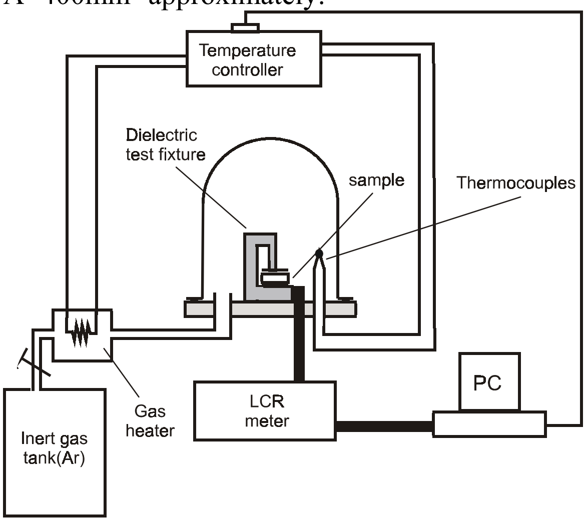

Fig. 1: Experimental setup

The experimental setup is shown in Figure 1. The dielectric measurements were conducted using an LCR meter (Agilent model 4284A), accompanied by the dielectric test fixture (Agilent model 16451B) and further supported by a computer for data recording, storage and analysis. The dielectric test fixture that was used to hold the specimen was protected by a cabin providing constant temperature (298K), inert atmosphere by continuous effusion of inert gas and also low humidity. It is important to mention that although the specific rock is characterised by its low porosity the existing ac conductivity mechanisms depend determinately on the water content of the specimen[10].

For all dielectric measurements, the amplitude of a sinusoidal ac voltage source was kept constant at 2V so that the average electric field across the sample was about 5V/cm. The instrument directly provided the values of capacitance C and dielectric loss $\tan\delta$. Other parameters such as the real part ε' of complex permittivity and ac conductivity σ are calculated using the relations:

$$\varepsilon' = \frac{4tC}{\varepsilon_0 \pi d^2} \tag{4}$$

and

$$\sigma = 2\pi \cdot f \cdot \varepsilon_0 \cdot \varepsilon' \cdot \tan\delta \tag{5}$$

where t is the sample thickness, d is the diameter of the electrodes, $\varepsilon_0 = 8.856x10^{-14}\ F \cdot cm$ is the permittivity of free space and f is the frequency of the applied ac field. The experimental procedure can be described as follows: Initially dielectric measurements were conducted on unstressed samples. Afterwards, the samples were subjected to mechanical axial stress S, using an axial hydraulic load machine (Enerpac–RC106). The stress S was applied on the sample for time $t_S = 300s$ and then the sample was removed and remained unstressed for time $t_r = 6ks$ before repeating the measurements. The value of the applied stress S varied between $0.1S_{\max}$ and $0.84S_{\max}$, where $S_{\max}$ stands for the upper limit of the recorded stress before fracture.

The stress S on the material is given as a function of the strain Є. For the linear elasticity region it can be stated that:

$$S = Y_o \cdot Є \tag{6}$$

where Y_o is the Young modulus of the undamaged material which is constant in the elastic region. For the rock samples this region corresponds up to $0.5S_{\max}$ approximately. When the stress takes values that lead further than the (linear) elastic region (i.e $S > 0.5S_{\max}$) then microcracks occur. In this region, where the sample is plastically deformed, the strain Є *is* greater than the value given by equation (6).

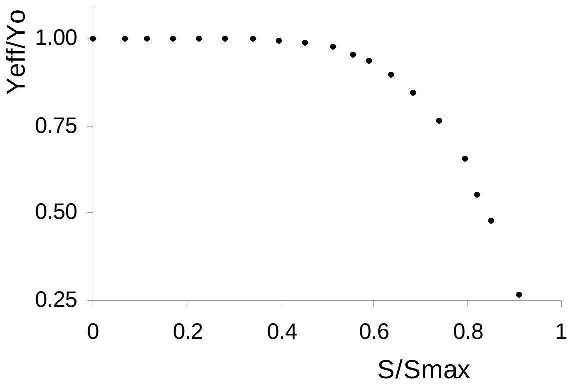

Fig. 2: The relation between Young modulus and stress on normalized axes.

Accordingly[3]:

$$S=Y_{eff}\cdot \epsilon \quad (7)$$

where Y_{eff} is the effective Young modulus and it is no longer considered as constant. In the plastic region the Young modulus becomes progressively smaller while stress increases.

Figure 2 depicts how the Young modulus is related to stress when both quantities are normalised by their maximum values. The curve was built using data provided in other scientific works[9,11] and correspond to Penteli marble samples.

3 Experimental results and discussion

A three-dimensional plot of the measured real part ε', of the complex relative permittivity, versus frequency and ratio $S/S_{\max}$ for marble sample is shown in Figure 3. Studying in detail the diagram several results can be extracted concerning the behaviour of the sample depending on the frequency or stress level. The real part of the dielectric permittivity approaches asymptotically a constant value, ε_∞ at high frequencies. This value is not related to the hopping dynamics of the mobile ions but instead is the result of much more rapid polarization processes occurring in the material[12]. When the frequency is lower than 10kHz ε' takes values with a strong dispersion confirming the universal law[13]:

$$\varepsilon'(f)-\varepsilon_\infty \propto f^{\,n-1}$$

where the exponent n lies in the range $0.6 < n < 1$. However, for applied stress $0 < S < 0.55S_{\max}$, no major changes are observed in the values of ε'. A slight distinct change is detected in low frequency regions of less that 1kHz. In the range $0.55S_{\max} < S < 0.84S_{\max}$, where the strain lead to the initiation of microcracking, the values of ε'

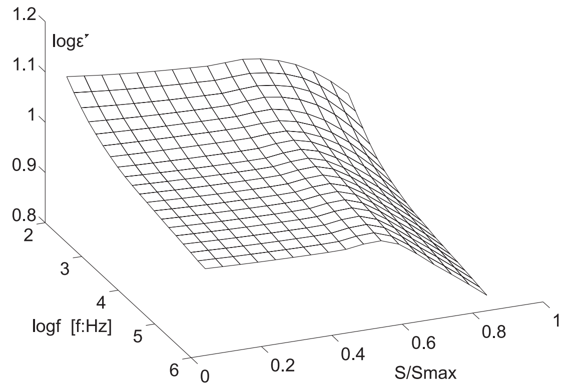

Fig. 3: Presentation of the behaviour of the real part of ε^* and its dependence on frequency and stress for marble samples.

decrease slightly. Especially when the strain of the samples due to the uniaxial stress S applied, reaches the value $0.84S_{\max}$, thus causing significant macro-cracks to the sample, the decrease in the values of ε' at all frequencies from 150Hz to 1MHz varies between 25% and 30%. For minor strains corresponding to $0.7S_{\max}$ the relevant decrease of the observed ε' values ranges from 10% to 15 %.

A three-dimensional plot of the measured dielectric loss $\tan\delta$ versus frequency and the ratio S/S_{max} for marble sample is shown in Figure 4. From the above curves it is obvious that for applied stress up to $0.5S_{\max}$ no loss peaks are recorded in the studied frequency range (150Hz-1MHz) and at the

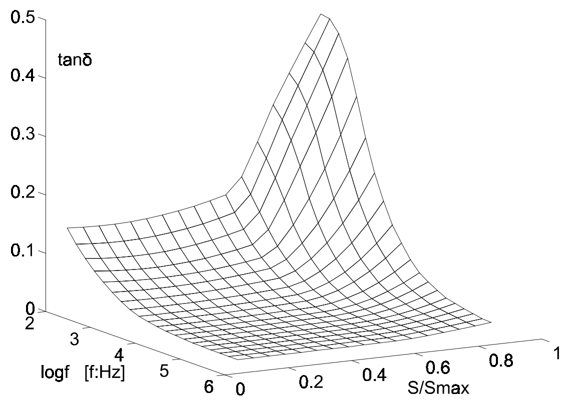

Fig. 4: Presentation of the dielectric loss $\tan\delta$ dependence on frequency and stress in marble samples

same time a very strong dispersion at low frequencies (*f*<5kHz) is noticed. This behaviour can be attributed to the response of hopping charge carrier systems which is characterized by the absence of loss peaks and which may show a very strong dispersion at low frequencies (*f*<5kHz).

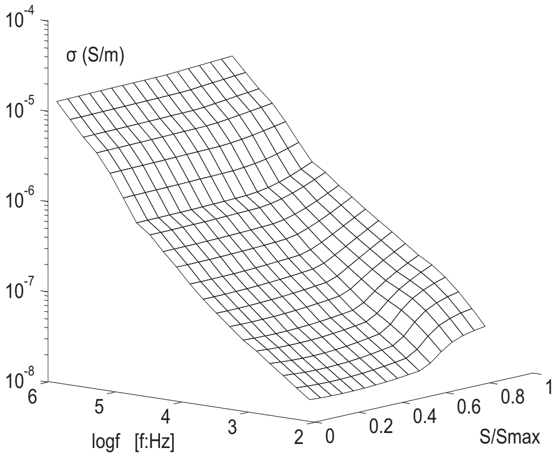

Fig. 5: Presentation of the dependence on frequency f and ratio pressure S/Smax of ac conductivity in marble samples.

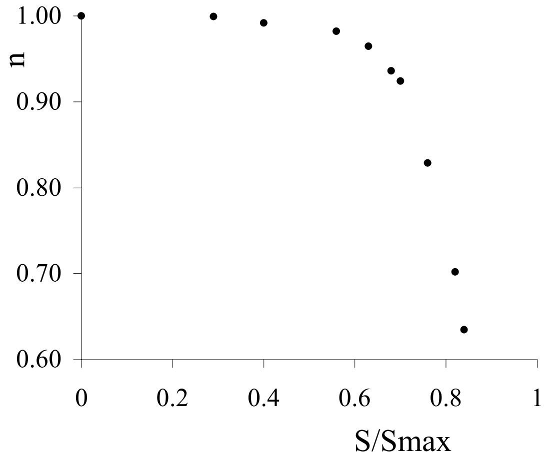

Fig. 6: Plot of the exponent n vs normalized stress.

From the shape of $\tan\delta$ curves, when the applied stress to the samples is greater than $0.5S_{\max}$, a tendency of loss peak formation can be noticed, at frequencies lower than 150Hz. Especially in the case of an applied stress $S = 0.84S_{\max}$ the existence of a loss peak is obvious in the range of 200-400Hz.

Using the expression (5) the ac conductivity σ is calculated. A three-dimensional plot of the measured ac conductivity versus frequency and the ratio $S/S_{\max}$ for marble sample is shown in Figure 5. As can be seen at the surface plot (figure 5) when stress is lower than $0.5S_{\max}$ there is no obvious change of the shape of the curves with respect to frequency. The above means that regarding ac conductivity the samples behave identically when they are not stressed and when the applied stress is in the elastic region.

When the applied stress becomes greater than $0.6S_{\max}$, the value of conductivity reached is definitely greater than that of the corresponding value at a lower stress $S < 0.5S_{\max}$. In the low frequency region, strong changes of the ac conductivity are recorded, compared to those at high frequencies. Especially, at frequencies $f > 100kHz$ such changes are even smaller.

In the high frequency region $f > 50kHz$ ac conductivity follows the power law $\sigma = Af^n$ once the component $Af^n >> \sigma(0)$. With the use of fitting techniques in the above mentioned frequency range the exponent n can be calculated quite accurately once the estimated error never exceeded ±0.03. The exponent n depends greately on the ratio $S/S_{\max}$ when the sample has been subjected to stress greater than $0.5S_{\max}$ and more precisely when the ratio increases, n decreases. When the sample is subjected to a stress $S < 0.55S_{\max}$ in the elastic region, no important variations of the factor n are calculated; it varies between 0.90 to 0.95. Figure 6, presents the variation of the exponent n with respect to the values of the ratio $S/S_{\max}$.

4 Conclusions

When the samples were subjected to stress S below the plastic region (as far as the mechanical behaviour of the material is concerned), no important variations were observed in the values of the real part of the complex permittivity, the dielectric loss and the ac conductivity of marble samples.

When the marble sample had been subjected to stress adequate to lead it to the plastic region, the values of ε' were clearly different and smaller than those measured when the sample was subjected to stress corresponding to the elastic region. This shows that the appearance of a crack micro-structure and the total crack volume prevented the reversible displacement of electric charges in the geomaterial. On the contrary, the values of $\tan\delta$ and conductivity increase rapidly. In this case, the appearance of crack micro-structure as well as the further appearance of the total crack volume caused significant increase to the ohmic losses due to the free charge movement in the material.

The exponent n that participated in the relation $\sigma = Af^n$ that describes ac conductivity in the high frequency region (i.e $f > 50kHz$) gets low values when the samples have suffered a stress that caused the creation of crack micro-structures.

From the above mentioned results it can be concluded that dielectric spectroscopy is a promising non-destructive testing method that can be easily applied to the investigation of micro-cracks and structural imperfections in marble that have been caused by aging, earthquake preparation mechanisms or various other factors to be determined.

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