Tectonophysics 637 (2014) 198–206

Contents lists available at ScienceDirect

Tectonophysics

journal homepage: www.elsevier.com/locate/tecto

A combined complex electrical impedance and acoustic emission study in limestone samples under uniaxial loading

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ARTICLE INFO

Article history:
Received 31 March 2014
Received in revised form 3 October 2014
Accepted 8 October 2014
Available online 18 October 2014

Keywords:
Electrical impedance spectroscopy
Acoustic emissions
Electrical conductivity
Pressure
Limestone

ABSTRACT

In the present work, complex electrical impedance measurements in the frequency range of 10 mHz to 1 MHz were carried out in conjunction with acoustic emission monitoring in limestone samples subjected to linear and stepped-like uniaxial loading, up to ultimate failure. Cole–Cole plots of the complex impedance during the stepped loading of limestone have been used to discriminate the contributions of grains interior, grain boundaries and electrode polarization effects to the overall electrical behavior. The latter is well-described with an equivalent-circuit model which comprises components of constant phase elements and resistances in parallel connection. Electrical conductivity increases upon uniaxial loading giving rise to negative values of effective activation volume. This is a strong experimental evidence for the generation of transient electric signals recorded prior to seismic events and may be attributed to charge transfer (proton conduction) due to cracks generation and propagation as a result of the applied stress. The time-series of ac-conductivity at two distinct frequencies (10 kHz, 200 kHz) during linear loading of limestone samples exhibits a strong correlation with the acoustic emission activity obeying the same general self-similar law for critical phenomena that has been reported for the energy release before materials fracture.

1. Introduction

The application of mechanical stress in rocks and in brittle materials in general may be accompanied by various fractoemission phenomena such as emission of charged particles, atoms, molecules, electromagnetic radiation (including infrared radiation) and acoustic emission (AE) activity (Cress et al., 1987; Dickinson et al., 1981, 1991; Freund, 2002; Frid et al., 2003; Lavrov, 2005; Lockner, 1993; Mori et al., 2009; Yoshida and Ogawa, 2004). In each case, the underlying physical mechanisms which are responsible for the occurrence of one or more of these fracture-induced processes have been widely investigated, mainly due to the fact that the plethora of all these phenomena constitutes the basis of searching precursory signals in mechanical damage as well as in earthquake prediction (Bleier et al., 2010; Eberhardt et al., 1999; Freund et al., 2006; Park, 1997; Turcotte et al., 2003; Varotsos, 2005).

For example, transient weak electric currents (known better as pressure stimulated currents or PSCs) which have been experimentally recorded before and during the failure of uniaxially-loaded rock specimens have also been observed in field measurements and discussed in theoretical models, i.e., seismic electric signals (SES) generated before the occurrence of seismic events (Stavrakas et al., 2004; Vallianatos et al., 2004; Varotsos and Alexopoulos, 1984a,b, 1987; Varotsos and Lazaridou, 1991; Varotsos et al., 1986; Varotsos et al., 1991). Various models have been proposed to explain the generation of these weak electric signals in certain cases, including the piezoelectric effect in rocks containing piezoelectric minerals, electrokinetic phenomena in water-saturated rocks, the motion of charged edge dislocations (MCD model), the presence of point defects including positive holes (defect electrons in the O^{2-} sublattice of silicate minerals), etc. (Enomoto, 2012; Freund, 2002; Vallianatos and Tzanis, 1998; Vallianatos et al., 2004; Varotsos, 2005; Yoshida et al., 1998).

The electric transport which is responsible for the recorded transient electric signals in geomaterials may be investigated by means of electrical impedance spectroscopy (EIS) since the application of an ac-electric field lead the pressure-induced charges and thus will cause a permanent variation to the measured electrical properties. In such a way, the electrical and dielectric properties may be analyzed in terms of the physical parameters related to the damage evolution in the material under investigation during its mechanical stress (Glover et al., 1997, 2000; Gómez et al., 1997; Nover et al., 2000).

To the best of our knowledge, detailed studies on the frequency-dependent electrical properties in conjunction with acoustic emissions during the application of mechanical stress in materials are rather limited and are restricted only to the investigation of phase transitions at high temperatures in relaxor ferroelectric compounds (Dul'kin et al., 2009, 2011).

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http://dx.doi.org/10.1016/j.tecto.2014.10.011
0040-1951/

So, in the present work, measurements of complex electrical impedance were carried out simultaneously with AE records during the uniaxial mechanical loading of limestone samples and different formalisms of representation were investigated in order to reveal any possible interrelations between acoustic parameters and the electrical and mechanical properties of samples.

2. Experimental setup

A light brown-beige limestone sample (polycrystalline $CaCO_3$ mainly with crystal forms of calcite and aragonite) of Mesozoic age originated from Ioannina, Greece, was supplied from a marble company and was sawed in disks of cylindrical shape (12–15 mm height and 33 mm diameter). This length-to-diameter ratio of the limestone samples is inevitable in order to compromise the requirement of a homogenous electrical field in the sample for ac-electrical measurements with the capacitance method and the necessity of enough adjacent space to mount properly the AE sensors and the strain gauge (Jonscher, 1996; Papathanassiou et al., 2010, 2011). In this way, AE activity which can be recorded for any sample geometry cannot be correlated with published data in the field of rock mechanics where length/diameter ratios from 2:1 to 3:1 are used, but can be further correlated in perspective to the measured electrical measurements. The samples dried in an oven at 105 °C for sufficiently long time in order to remove free water, until no weight loss could be measured and the electrical impedance spectrum remained unchanged. The high density limestone (2662 kg/m^3 in dried condition) has a Young's modulus of 37 GPa, its porosity is 1.1% and its water content at saturation is 0.42% (values provided to supplier by IGME, www.igme.gr/grlith.htm). The measurement of the longitudinal ultrasonic velocity of the samples was measured with a pair of AE sensors in pulse-receiver mode and gave an average value of 4.65 km/s.

The experimental setup is shown schematically in Fig. 1. The specimen was mounted between two parallel stainless-steel electrodes forming a cylindrical capacitor. Conductive silver paint was applied at both sides of the cylindrical sample in order to achieve a stable electrical surface contact with the stainless-steel electrodes during the uniaxial loading. The electrodes were electrically isolated from the load platens by thin Teflon plates (Stavrakas et al., 2004 and references there in). A miniature strain gauge (1 mm length) was mounted on the lateral surface of the specimen and a load cell was placed at the bottom side of the capacitor for continuous stress–strain monitoring. Signals from both load cell and strain gauge were recorded with a signal-conditioning module (SCXI-1520 with SCXI-1314 terminal block, National Instruments) and a DAQ card (PCI-6221, National Instruments), through an appropriate program developed in LabVIEW graphical programming environment.

Acoustic emissions were detected through 4 miniature piezoelectric sensors (PICO sensors, 200 kHz–1 MHz, MISTRAS Group, SA) mounted on the sample's lateral surface and recorded in an integrated multichannel system of Physical Acoustics Corporation. A pre-amplification of 40 dB was used in each channel and the sampling frequency of signals was 5MSPS. The threshold of detection was determined with the load machine turned on and the specimen in contact with the load platens at a minimum load of 2–3 kN and thereby was settled at 40 dB in order to eliminate the background noise. The AE activity is represented in time series of detected signals (hits), events (single count for hits detected to different sensors into a predefined time window from a hit) and amplitude (signal peak in dB).

Uniaxial stress was applied by a 3000 kN loading machine (ALPHA S-3000, Form + Test GmbH) equipped with a digital controller (Digimaxx-21) for the automated control of the servo-hydraulic system.

Complex conductivity measurements were carried out by means of a high-resolution broadband spectrometer (Novocontrol Alpha-N Analyzer). The frequency of the applied ac electric field was varied between 10^{-2} Hz and 10^6 Hz. At this frequency range, the accuracy of impedance measurements is 0.2–1%, depending on the measured resistance of the sample (10^3–10^{11} Ω). Proper electromagnetic shielding consisting of a Faraday cage with copper sheets was implemented to the whole apparatus, in order to diminish noise problems which are common at low frequencies (Fig. 1b). The WinDeta and WinFit software of Novocontrol Technologies were used for acquisition and modeling of the dielectric experimental data.

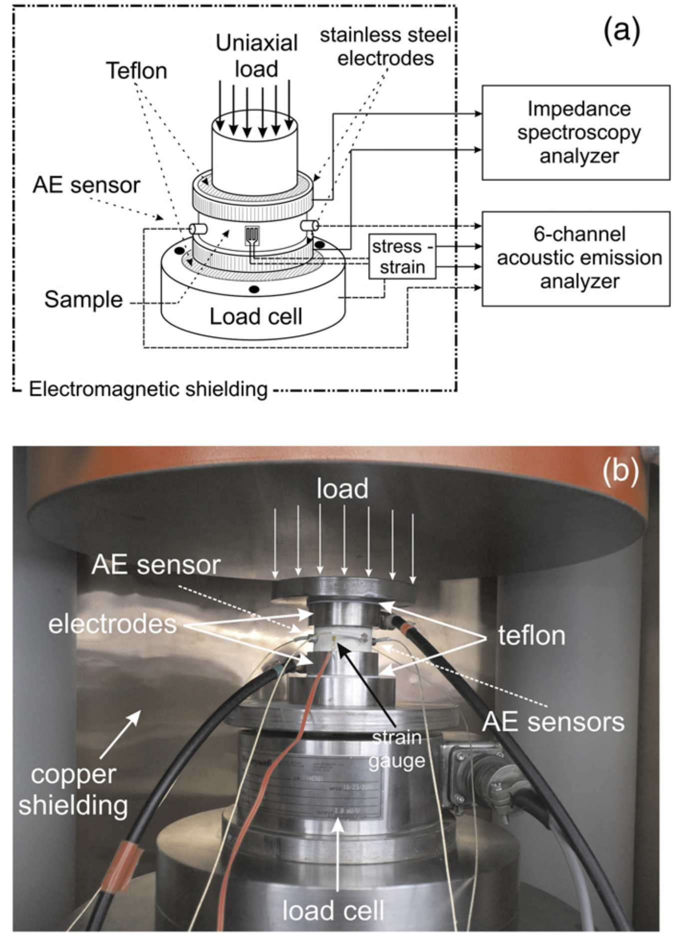

Fig. 1. (a) Schematic representation of the experimental setup used for the simultaneous measurement of complex electrical impedance and acoustic emissions during the uniaxial loading of limestone samples. (b) Photograph of the sample inside the load frame which is covered by copper sheets for EM shielding.

Assuming a parallel connection of the capacitance C(ω) and the resistance R(ω) which are the output values of the dielectric analyzer, the complex impedance Z*(ω), the complex conductivity σ*(ω) and the dielectric permittivity ε*(ω) are interrelated through the following relations:

$$\frac{1}{Z^*(\omega)} = \frac{1}{R(\omega)} + j\omega \cdot C(\omega) \tag{1}$$

$$\varepsilon^*(\omega) = \varepsilon' - j\varepsilon'' = \frac{C(\omega)}{C_o} - j\frac{1}{\omega \cdot C_0 \cdot R(\omega)} = -\frac{j}{\omega \cdot C_0 \cdot Z^*(\omega)} \tag{2}$$

and

$$\sigma^* = \sigma' - j\sigma'' = j\omega\varepsilon_o(\varepsilon^* - 1) = \frac{\varepsilon_o}{C_o R(\omega)} - j\omega\varepsilon_o\left(\frac{C(\omega)}{C_o} - 1\right) \tag{3}$$

where $C_0 = \varepsilon_o \cdot \pi \cdot r^2/d$ is the capacitance of the empty cylindrical capacitor, with distance d between the electrodes and r their radius, ω = 2πf is the angular frequency and ε_o is the permittivity of the vacuum.