Fracture related electromagnetic emission measurement and excess noise analysis of reinforced composites

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Abstract

Electrical properties of several types of reinforced composites were investigated. AC and DC resistivities were measured, and the frequency behavior along with parasitic components was put together in order to create equivalent electrical circuit. It was used for electrical noise measurement corrections. It turns out that samples produce high level Johnson-Nyquist noise and 1/f noise when current bias is applied. We have discovered that electrical contact metallization affects directly the 1/f noise and the bulk material is responsible for white-like Johnson-Nyquist noise. We managed to get information about the suitability of the various metals in view of noise generation and the silver conductive paint seems to be proper choice. Finally, the electromagnetic emission signal, which in turn depends on cracks, was measured and comparison with conventional acoustic emission was put forward.

Keywords: Reinforced composites; electromagnetic emission; excess noise; contact noise.

1. Introduction

The polymer based reinforced composites are very promising materials now. The reason is tailoring of its properties to the specific application with specific requirements, Greenhalgh (2009). Nevertheless, materials with wide variability in production require new methods for the quality control immediately after fabrication as well as...
during their application. It is also necessary to encourage fundamental material research in order to understand complex microstructure material properties. Our aim is put forward new methods or its combination for a fast scientific testing to determined degradation and live-time reduction. Very promising approach is the electromagnetic emission and the excess electrical noise measurement when the sample is kept at the constant physical conditions. The electrical noise is often used as an indicator of specific imperfections just by its level monitoring, Dutta (1981). This method is technically relatively easy to implement but it lacks information about the nature of imperfections and their causes or area of occurrence. For this reason, we use several different techniques and we observe the correlations between them. Methods that we use are acoustic emission (AE) and electromagnetic emission (EME) when sample is mechanically stressed, Koktavy (2009). Furthermore, we use electrical resistance measurement and electrical noise measurement when samples are without mechanical tension, Macku and Koktavy (2013).

2. Experimental methods and samples under investigation

The matrix used in this study is a vinyl ester, polyester and an epoxy polymer based on Bisphenol A resin. The lowest resin viscosity has a vinyl ester followed by polyester and epoxy. As reinforcement was used combination of E-glass, AR-glass, basalt and carbon fibres. The combination of matrix and reinforcement determine the interparticle contact, which affect the conductivity of the system and relating electrical properties. Carbon and E-glass fibres are approx. (300 – 700) nm in diameter (verified by SEM). Rectangular samples with varying dimensions were cut in order to distinguish between surface and bulk phenomena. The dimensions of samples are about (65, 50, 25) mm length, (50, 30, 20) mm width, and (20, 10, 5) mm thickness. Metallic contacts (cross-sectional areas) are prepared either as a full carbon layers with sheet resistance about 90 $\Omega$/sq, silver layers with sheet resistance about 100 $\Omega$/sq or copper foil. Each sample was cleaned by ethanol and stabilized by vacuum oven at 90 °C for 1 h to avoid moisture effects. The DC resistance temporal analysis was realized by the Keithley 6517B. The 2-terminal AC impedance spectroscopy was performed using an Atlas Sollich 0441 high impedance analyzer (frequency range from 10 $\mu$Hz to 100 kHz). The noise-based measurement has been done by means of the Agilent 35670A two-channel FFT dynamic spectral analyzer. The analyzer is equipped by the custom made two-channel ultralow noise preamplifier (LNA) with high input impedance of about 200 M$\Omega$ and input capacitance of about 15 pF. The noise background of LNA is about 2.4 nV/$\sqrt{Hz}$ at 100 Hz and the -3 dB bandwidth is 50 $\mu$Hz ÷ 10 kHz. The characteristics of the spectral densities as a function of the bias current has been done by means of the Keithley 6220 precision current source. The EME measurement has been done by means of the National Instruments sampling unit PCI 6111 in continual acquisition mode and the EME signal was detected by the capacitance sensor.

3. Results and discussion

3.1. DC resistance and AC impedance spectroscopy

The DC resistance as well as AC impedance of the fibre reinforced polymers generally depends on the moisture (hydrophilicity), impurities, chemical properties, material crystalline or amorphous nature internal defects and reinforcement to matrix internal contact, Sandler et al. (1999), Pathania and Singh (2009). This measurement bears diagnostically interesting information and fundamentally defines requirements on the relating experimental set-up. For the DC measurement must be strictly used high resistivity meter or combination of the electrometer and the constant voltage source (e.g. Keithley 6517B). To ensure sufficient measurement accuracy the test voltage in the range (10 ÷ 40) V must be applied. A relatively high electric field introduces the dielectric polarization and the relaxation process taking place in the material, Jonscher (1983). In any case, the material is in the different state compared with the noise measurement without electrical excitation as well as the EME measurement with mechanical stress. The long term monitoring of the sample resistance development was measured for the each sample under investigation. It turns out that the sample resistance increase monotonically just like relaxation processes disappears. Characteristics reach a maximum after 4.5 hour measurement interval. Let’s point out results related to samples mentioned before. We calculated DC volume resistivity from the resistance as is shown in Tab 1. Besides of that the AC impedance spectroscopy pointed out volume resistivity $\rho_{AC}(f)$ (it was calculated from real part of complex AC impedance). Measurement results are illustrated in fig. 1a.
Table 1. DC volume resistivities and dielectric constants of different reinforcement composites.

| Matrix and reinforcement composition | DC volume resistivity $\rho_{DC}$ / $\Omega$ cm | Dielectric constant $\varepsilon$ / - |
|-------------------------------------|-----------------------------------------------|------------------------------------------|
| Epoxy – carbon fibres              | $2.4 \times 10^9$                            | 4.70                                     |
| Epoxy – basalt fibres              | $5.3 \times 10^9$                            | 3.90                                     |
| Epoxy – E glass fibres             | $2.2 \times 10^{10}$                         | 5.85                                     |
| Epoxy – AR glass fibres            | $1.9 \times 10^{10}$                         | 5.71                                     |
| Polyester – E glass fibres         | $1.5 \times 10^{10}$                         | 5.64                                     |
| Vinyl ester – E glass fibres       | $9.1 \times 10^9$                            | 5.32                                     |

We can conclude that the AC volume resistivity is for low frequency comparable with DC resistivity and it is almost constant in the frequency range under inspection. This finding is not valid for epoxy-carbon and epoxy-basalt composites where the volume resistivity drops down rapidly (see fig. 1a). The imaginary part of complex resistivity pointed out uniform capacitive behavior (not presented here) and the dielectric constant $\varepsilon$ can be calculated, see Pathania and Singh (2009) and Tab. 1. The dielectric constant depends on reinforcement loading due to increased orientation and interfacial polarization, Greenhalgh (2009), Sandler et al. (1999). The dielectric constant also slightly depends on the frequency with decreasing tendency. This phenomenon seems to be insignificant for the noise measurement. In any case, the dielectric constant of composites increase with the addition of fibres with higher dielectric constant then base polymer matrix. Similar results were reported by Ounaies et al. (2003). Another experiment which was carried out relates to the measurement of the DC surface resistivity (it was done by means of modified Keithley 8009 test fixture). Results prove a similar behavior to the DC volume resistivity with an overall higher value. This important result indicates that the surface or edge related leakage current is not important and the electrical noise generation is really due to the cross-current (volume) conduction.

Fig. 1. (a) The log-log plot of the AC volume resistivity of different composites as a function of the frequency (carbon electrodes); (b) The model of the input circuits including sample, LNA input components and noise source under investigation. Typical values of individual components are $C_i = 15$ pF, $R_i = 200$ M$\Omega$, $C_k = 12$ pF, $C_l = 10$ nF, $C_f = 4$ pF; (c) Arrangement of samples electrical excitation and PSD measurement.

3.2. Excess electrical noise analysis and EME signal

The fundamental equivalent model of the input circuits is illustrated in fig. 1b. Here $i_n$ is internal current noise of the sample; $C_S$ is sample capacitance; $R_S$ sample resistance; $C_f$ fixture capacitance; $C_k$ cable capacitance; $C_C$ coupling capacitance and $R_{in}$, $C_{in}$ are amplifier input resistance and capacitance. Variable $u_n$ symbolizes input signal of the LNA. The EME signal along with the excess noise is modeled by means of the noise current source $i_n$. This is due to the physical nature of expected signals. The EME is created due to oscillation of coupled electrical charges in the form of electrical dipole (if crack is created). The peak value as well as total energy and pulse shape correlate...
with redistributed electric charge in the defect region, Koktavy (2009). In addition to the EME, we expect also two excess noise sources without direct relation to cracks. The first one is caused by the thermal energy of charge carriers and electric dipoles. These particles fluctuate in position and the macroscopic stochastic electric field is generated. Physical properties are defined on the basis of the fluctuation-deviation theorem and the Johnson-Nyquist noise is produced. Physical variable under consideration is the power spectral density (PSD) of current fluctuation given by well-known formula $S_i = \frac{4kT}{R}$ (here $k$ is Boltzmann constant, $T$ temperature and $R$ is sample static resistance), see Van Etten (2005). Nevertheless, the real experiments with the samples without electrical excitation prove that we have to look back to the equivalent model. The input components form modified first order band-pass filter and the EME along with the electrical noise is affected. Fortunately, the lower cut-off frequency is given by the coupling capacitance and it is lower than 100 mHz in our case. The upper cut-off frequency is given by the parallel capacitance combination and it is shifted slightly higher because of the input resistance $R_{in}$. Technically, the real amplifier input resistance and the sample capacitance along with the capacitance of cables and feed thought connectors are limiting factors for the composite materials noise and the EME signal measurement. On this account, the equivalent circuit frequency dependent function was derived in the differential form as follows.

$$(b + C_{in}R_{in}) \frac{d^2u_n}{dt^2} + \left( \frac{b}{C_c R_{in}} + \frac{R_{in}}{R_s} + 1 \right) \frac{du_n}{dt} + \frac{u_n}{C_c R_{in} A} = R_s \frac{di_n}{dt}$$

(1)

Here $b$ is time constant of the sample including parasitic capacitors $b = C_S C_f R_S$.

![Fig. 2. (a) The comparative plot of the current to voltage transition function based on eq. (1). Samples thickness is 10 mm, cross-section (50 x 30) mm; (b) The reconstructed PSDs of the samples without electrical excitation. Carbon electrodes, ambient temperature 295 K.](image)

This nonlinear function was symbolically solved by means of the MathWorks Matlab and the solution (input noise current $i_n$ to output noise voltage $u_n$ transition function) is used for reconstruction of affected noise PSDs in the frequency domain as well as electromagnetic signals in the time domain. Figure 2a illustrates the current to voltage transition functions based on eq. (1) for the each sample under inspection taking into account the frequency-dependent sample resistance. The samples capacities were calculated from the dielectric constants and the parasitic components were used as noted in fig. 1b. The transition functions were finally used for the reconstruction of the noise PSD measurements and results are illustrated in fig. 2b. The PSD pointed out white-like spectra and the volume resistivity can be calculated from Johnson-Nyquist formula. We get $2.41 \cdot 10^9 \Omega \cdot\text{cm}$; $5.28 \cdot 10^9 \Omega \cdot\text{cm}$; $2.18 \cdot 10^{10} \Omega \cdot\text{cm}$; $1.8 \cdot 10^{10} \Omega \cdot\text{cm}$; $1.28 \cdot 10^{10} \Omega \cdot\text{cm}$; $1.08 \cdot 10^{10} \Omega \cdot\text{cm}$ (values are in the same order as the data in the tab. 1). It corresponds with the data from the DC resistance measurement very well. This result supports the assumption of the Johnson–Nyquist noise caused by the free carriers rather than different white-like noise (polarization noise, shot noise) caused by the polarization effects or the metal-insulator potential barrier, Bittel (1976), Van Etten (2005).
The second excess electrical noise is produced if the external electric field is applied or the internal electric field arises by the crack creation. This noise results from the sample resistance fluctuation. That is why the external electric field is required and the electric current starts to flow in the same type. The PSD is inversely proportional to the frequency (so-called 1/f noise). For the homogeneous layers the PSD given by

$$S_R / R^2 = S_i / f^2 = \alpha_H / fN.$$  \hspace{1cm} (2)

Here $S_R$ is PSD of resistance fluctuation, $R$ is static sample resistance, $I$ is bias current flowing through sample, $\alpha_H$ is Hooge constant, $f$ is frequency and $N$ is number of the free carriers participating in the fluctuation process, Van Etten (2005). Generally, low dimension devices and dielectric materials with the low number of the free carriers produce high level of 1/f noise. The 1/f noise is usually supposed to be result of the bulk imperfections. Nevertheless, point metallic contacts and grainy layers produce 1/f noise, too, Hooge (2003). Arrangement of the experiment including electrical bias-current (20 nA) is illustrated in fig. 1c. Whenever the current bias was applied or changed the 4.5 hours settling time was used. By this way the relaxation phenomenon disappears and the background noise is invariant to the bias conditions. Experimental results of the individual samples with the same dimension are shown in fig. 3a. The current PSD is proportional to $f^b$ where $b$ belongs to the interval (1.2 ÷ 1.5) in case of the all samples under inspection. The poor quality exhibits the epoxy-carbon and the epoxy-basalt composites.

Fig. 3. (a) Current PSDs of different composites for the same bias current 20 nA. Samples thickness is 10 mm, cross-section (50 x 30) mm; contacts are made of carbon conductive paint; (b) Current PSDs vs. relative sample cross-section. Bias current 20 nA, frequency $f = 1$ Hz. Symbol $A_0$ denotes the smallest cross-section of the sample and $n$ is an sample index. Circles indicates copper based contacts, stars carbon contacts and crosses silver contacts; Settling time was 4.5 hours and ambient temperature $T = 295$ K.

We also provide another experiment to find the region where the 1/f noise is generated. We measured the PSD for the different thickness of the samples with the same cross-section (not presented here). The PSD is evidently invariant to the samples thickness. With increasing volume we naturally expect decreasing 1/f noise level because of the increasing number of the free particles $N$. Instead of this we conclude the invariant behavior. On the other hand, the PSDs are inversely proportional in case of samples with the different cross-sections and the uniform thickness. Figure 3b shows results of this measurement for three polymer composites with the different base-noise level. Now we can conclude that the region of the interest is contact – polymer interface. For this reason, measurement was done once again with the same group of the samples with different contacts. It turns out that type of the metallization affect results significantly. The lower noise level and the best results exhibits silver conductive paint followed by carbon paint and copper foil.

Finally we provide the EME measurement when a sample is mechanically stressed. We confine ourselves to the vinyl ester – E glass sample with the silver contacts as well as to small thickness and cross-section to ensure low 1/f and Johnson – Nyquist noise. Results are illustrated on fig. 4b together with the acoustic emission (AE) signals. The AE is well established tool for the material diagnostic and it is used for the results comparison here. It should be emphasized that the strong correlation exists even if the EME signal detection is much more complicated.
4. Conclusions

Electrical properties of the several types reinforced polymer composites were studied using the noise spectroscopy, the DC resistance and the AC impedance measurement techniques. The resistivity of the epoxy – carbon and the epoxy – basalt is relatively low and indicates good interparticle contact or fibres conductive paths. In addition, the AC impedance curves falls down rapidly which can, in principle, be important for the broadband noise measurement. It turns out, that all samples produce white-like Johnson-Nyquist noise. Nevertheless, the experimental data must be corrected. The PSDs were used and expected static volume resistivities were extracted. It proves assumption of the negligible polarization noise and the Johnson-Nyquist noise was confirmed. Another recognized noise is in the $1/f$ form. It develops with the bias current according to the square law. It turns out that the noise is invariant to the samples thickness change and it is inversely proportional to the sample cross-section. It indicates that the region responsible for the noise generation is the contact-polymer interface. Carbon and copper contacts are poor in view of the noise generation and the silver contacts may be recommended.

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