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### Citation

HAREA, Evghenii, Sanjoy DATTA, Jaroslav MALOCH, and Radek STOČEK. Novel perspectives on self-monitoring internal rubber failure using piezoresistivity. *International Journal of Fatigue* [online]. vol. 175, Elsevier, 2023, [cit. 2025-02-11]. ISSN 0142-1123. Available at <https://www.sciencedirect.com/science/article/pii/S0142112323003262>

### DOI

<https://doi.org/10.1016/j.ijfatigue.2023.107825>

### Permanent link

<https://publikace.k.utb.cz/handle/10563/1011611>

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# Novel perspectives on self-monitoring internal rubber failure using piezoresistivity

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## ABSTRACT

Carbon black (*CB*) fillers are often used to increase the ultimate strength and durability of rubber products. Incidentally, *CB*s are also electrically conductive and this property at low filler concentrations was successfully exploited by several researchers to monitor changes in mechanical properties of rubber by piezoresistance evaluation.

In the present work, piezoresistive response of deformed natural rubber (*NR*) filled with higher concentrations of *CB*, which are often encountered in rubber products under dynamic loading conditions like tires, conveyor belts, vibration dampers and the like was investigated. Unfortunately, high carbon black content significantly decreased piezoresistive signal, severely limiting the self-monitoring capability. Moreover, unexpectedly high and undesirable for electromechanical application such as precursor of shoulder peak phenomenon (*PSPP*) and shoulder peak phenomenon (*SPP*) were observed.

However, these otherwise undesirable phenomena in a noble way was used for self-monitoring the rubber integrity during cyclic deformation. The gradual increase in residual strain, indicating rubber matrix fatigue, was clearly related to changes in experimentally detectable electrical resistance as a function of *PSPP* and *SPP*.

The study is optimistic of a new concept for self-monitoring and forecasting rubber structure degradation under long-term cyclic loading predictable from simple obtainable piezoresistive response.

**Keywords:** Piezoresistivity, shoulder peak phenomenon, self-sensing monitoring, natural rubber, cyclic loading

## 1. Introduction

Rubbers are lowly cross-linked polymeric materials. Due to high internal flexibility, rubbers exhibit large deformations even under weak external forces above the glass transition temperature. These materials are usually filled with hard nano-structured particles in order to enhance different mechanical properties. However, the strength of rubber also depends on its chemical structure and the viscoelastic behaviors.

Over the years, numerous rubber fatigue prediction methods [1-3] based on mechanical parameters and self-monitoring procedures [4-6] mainly focused on electrical measurements have been developed. The advantages of electrical data usage over the mechanical approach are the undebatable lower cost of equipment, faster measurements and data processing. Thus, the self-monitoring of rubber degradation with in-situ appreciation of fatigue and remaining life time by measuring the piezoresistance of rubber product could be highly appreciated by stakeholders.

Clearly, high deformation of rubber-based products causes material degradation, decreasing the mechanical properties and simultaneously decreasing the lifetime.

Such degradation appears as a considerable stress reduction (so called stress softening) resulting from the rubber chain scission, sulfur bond breakage and destruction of inherent structures such as filler-filler and filler-polymer interfaces [7] followed by residual strain explained by rubber network reorganization and permanent internal rupture [8]. All the aforementioned processes directly influence the filler distribution within the rubber matrix. Thus, a direct link can be expected between the material degradation and electric properties. However, such a link has not yet been intensively scientifically discussed. On the other hand, it is important to mention that as the content of electrically conductive filler increases, the piezoresistive response of the deformed material decreases, i.e., the material becomes less electromechanically sensitive.

Generally, the electrical resistivity of a filled rubber is mainly influenced by the statistical average distance between the conductive filler particles and the probability of electrons flow to cross these distances. Therefore, a change in the distance between the filler particles, caused by the deformation of the filled rubber matrix is the reason for piezoresistance [9,10] confirmed experimentally by composites containing carbon nanotubes [11], carbon black [12 -15], carbon fiber [16], graphite [17], ceramics [18] and metals [19].

Unfortunately, most of these studies are limited to rubbers filled with conductive particles just above percolation threshold. Though this is an excellent approach for reproducible analysis of electrical properties, yet there are no studies dealing with highly filled rubbers that much more suit the real-world applications such as truck tires, conveyor belts, seals, bushing elements, and the like. The main reason is the drastic decrease in piezoresistive sensitivity, and an increase in electrical response nonlinearly. An increase in filler content increases the probability of an alternative conductive path formation governed mainly by hardly predictable internal (local) stress produced during the deformation of the sample.

The incorporation of necessary quantity of reinforcing filler in rubbers increase tear strength and reduce fatigue crack growth rates [20-23], whereas an increase in *CB* loading produces a slight decrease in fatigue threshold [24-26]. However, despite the considerable reinforcing effect, cavities, cracks and crack propagation occur during both quasi-static and cyclic dynamic loading [27-30], leading to failure of the integrity of the filler network [31]. From this point on, it will be considered that a change in repeatedly measured electrical resistance during cycling loading of rubber can detect changes in the structure of the filler network [32,33].

In terms of piezoresistance, it is necessary to distinguish between positive and negative piezoresistance for filled polymers, where a positive piezoresistance represents an increase and a negative a decrease in electrical resistance [34]. A unique situation occurs with rubber, where a positive piezoresistive effect is followed by a negative one during the same unloading process, which is manifested through the appearance of a shoulder peak phenomenon (*SPP*) [35-37].

Mersch et al. [38] notify that SPP may appear during the release of strain or when the imposed strain is completely released, independent of the filler material such as *CB*, carbon nano-tubes (*CNT*), or graphene. A largely accepted explanation states that *SPP* are caused by the simultaneous destruction, reconstruction, and re-formation of the conductive network [35-38]. Conclusively there is no doubt about the *SPP* being an integral part of piezoresistive properties of highly filled rubber and thus should be carefully analyzed.

The present work reports findings on piezoresistivity of natural rubber (*NR*) vulcanizates filled with 50, 60 and 70 phr of *CB*, all of which are unusually high concentrations for electro-sensitive but otherwise usual for many other industrial rubber applications. In order to understand the effect of the *CB* concentration on the evolution of piezoresistive effect, the rubbers were subjected to a monocyclic mechanical loading of a specific trapezoidal testing protocol.

Further to this, a second part of the study shows the possibility of applying the piezoresistive effect for self-monitoring and predicting the fatigue life of highly filled rubber under cyclic loading following a simplest triangular testing protocol.

## 2. Experimental section

### 2.1. Materials

The formulations for the carbon black (*CB*) filled elastomer composites, tested within this study are listed in **Table 1**. Natural rubber (*NR*) was supplied by the Astlett Rubber Inc (type *SMR 20 CV/BP1*). Sulphur used as the curing agent, and zinc oxide (*ZnO*) and stearic acid used as activators were supplied by Sigma-Aldrich. The reinforcing filler used in all the compounds was high abrasion furnace (*HAF*) (*N330*) carbon black (*CB*) supplied by Cabot Corporation, Boston, MA, USA.

**Table 1** Rubber formulation for the variously compounded batches.

	NR	Oil	Carbon Black	CBS	Sulfur	ZnO	Stearic Acid
	Content in phr <sup>*</sup>						
NR50	100.00	10.00	50.00	1.00	2.50	5.00	2.00
NR60			60.00				
NR70			70.00				

<sup>\*</sup>Parts per hundred of rubber by mass.

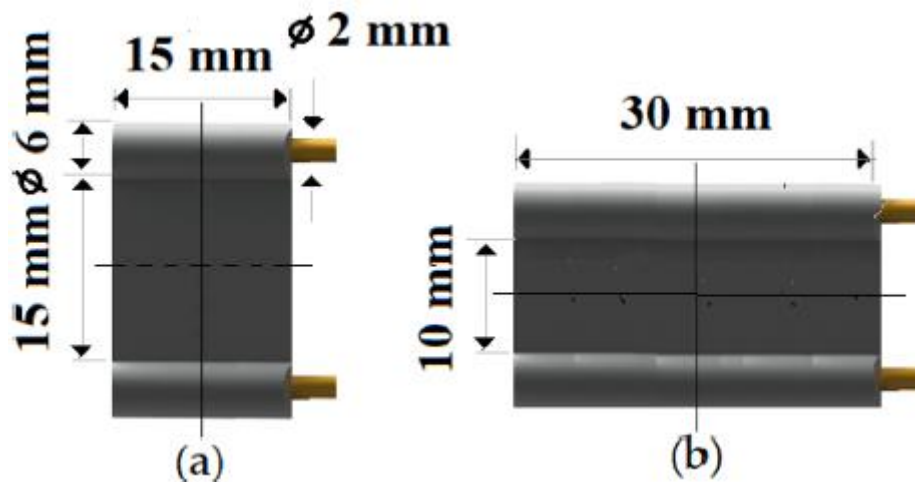
Moreover, Naphthenic oil NYTEX (Nynas AB, Sweden) was used as an extender for the efficient incorporation of *CB* filler and finally, *CBS* (N-cyclohexyl-2-benzothiazolesulfenamide) was employed as the curing accelerator.

Compounding of the various batches was accomplished in an internal mixer (Brabender Plastograph) at 60 °C at a rotor speed of 50 rpm at a fill factor of 80 %. The rubber and the compounding ingredients were successively added and mixed as follows: *NR* was masticated for 3 min followed by the addition and mixing of *ZnO* and followed by stearic acid activators, each for 1 min. The filler was added simultaneously with the extender oil in parts of three steps and mixed for another 3 min. Finally, *CBS* and Sulphur were added and mixed for another 2 min, so that the total mixing time was 10 min,

maintained to be identical for all the compounding batches. After 24 h conditioning at an ambient temperature of 23 °C, the compounds were molded using electrically heated hydraulic press (LabEcon, The Netherlands) at 160 °C and 200 kN into samples of specific geometry, defined generally with dimensions of  $15 \times 15 \times 2 \text{ mm}^3$  with cylindrical shoulders of 6 mm in diameter at the both ends. The optimum cure time for molding at 160 °C for each batch was determined using a moving die rheometer (MDR 3000 MonTech, Germany) according to ISO 3417. A brass tube (outer diameter 2 mm and inner diameter 1.4 mm) was vulcanized into each cylinder to implement electrical contacts for testing, as shown in **Fig. 1a**. In each compounded batch, three samples were prepared and tested. For the fatigue cyclic test, the formulation containing 70 phr *CB* with a deformable part of the sample having dimension of  $10 \times 30 \times 2 \text{ mm}^3$  was selected (see **Fig. 1b**).

## 2.2. Experimental setup

The customized experimental setup for direct current (*DC*) resistance measurements used in this study has been described in details in a previous work [13] (see **Fig. 2**). All data were recorded with digital multichannel oscilloscope RIGOL MS05104 (RIGOL TECHNOLOGIES, Co Ltd, China). In order to achieve accurate time synchronization of the sampled data, each of the physical quantities (force (*F*), displacement (*Δl*), current (*I*) and voltage (*V*)) was recorded from one of the 4 channels of the oscilloscope mentioned above. That fact gave the possibility to have a perfect fit of all the data with the internal timer of the oscilloscope. This was absolutely necessary, especially in the case of cyclic fatigue analyses for precise synchronization of values between the recording of mechanical quantities from the tensile device and electrical quantities from the electrical setup. Otherwise, in an unguaranteed synchronization, there would have been a risk of an increase in the temporal deviation of the values with the number of applied cycles.



**Fig. 1.** Geometries of tested samples: (a) for monocyclic testing protocol, (b) for cyclic test.

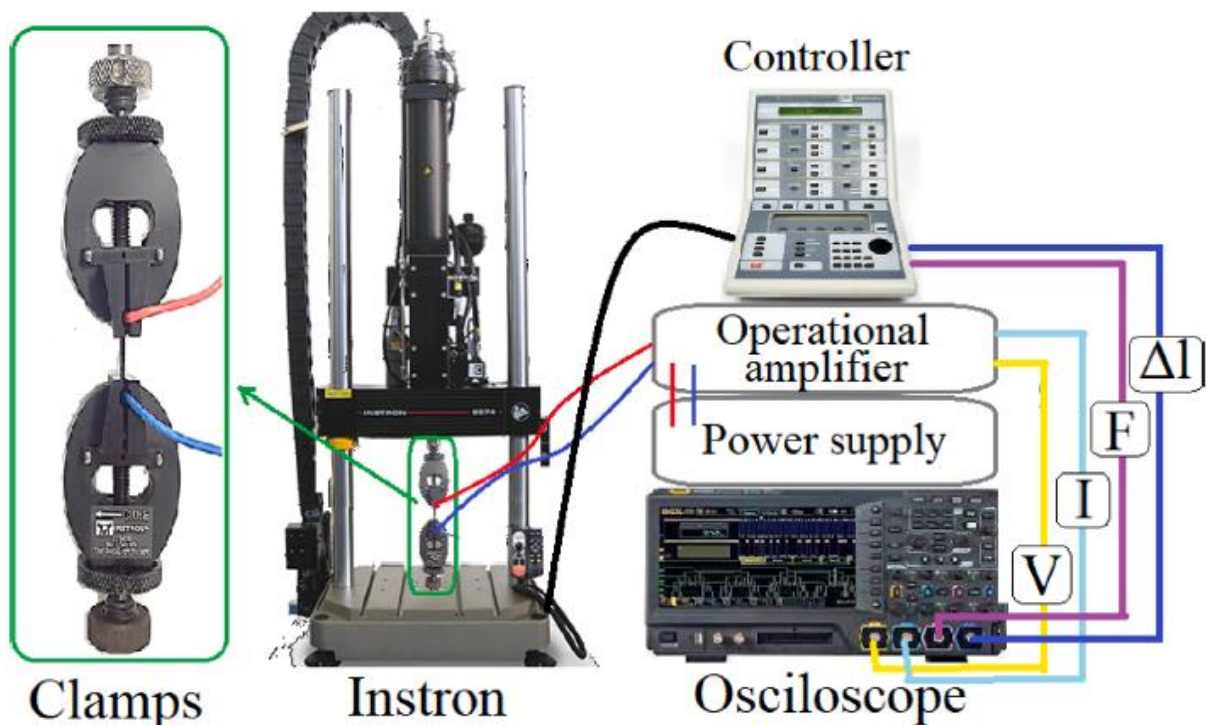


Fig. 2. The sketch of experimental setup.

The mechanical deformation of samples was done in a servohydraulic testing equipment, Instron 8871 (Instron, UK), equipped with modified non-conductive clamps as has already been described in [13], which were used to fix the lateral cylindrical shoulders of the tested samples. Due to the incorporation of the cylindrical shoulders, the additional stress commonly induced with simple fixing, was efficiently reduced. The conductive wires were mechanically crimped into the brass tubes, avoiding metal-rubber interface overheating as is otherwise disadvantageously encountered in the case of soldering.

### 2.3. Methodology

The applied testing protocol used for single test, which is schematically depicted in Fig. 3a has been based on three consecutive steps with an identical time of 8 s for each step. Firstly, the sample was strained up to 4 mm (26.7 % strain) at a constant strain rate of  $0.5 \text{ mm}\cdot\text{s}^{-1}$  followed by a holding phase at the maximum strain (total relaxation time of 8 s) and finally unloaded at the strain rate of  $0.5 \text{ mm}\cdot\text{s}^{-1}$  up to the basic position of the clamps.

The results of electrical resistance were interpreted in the form of a commonly used unitless ratio  $(R - R_0)/R_0$  termed as the relative resistance, where  $R$  is the measured resistance at a defined deformation and  $R_0$  is the initial resistance of the undeformed sample.

The testing protocol applied for the fatigue cyclic test is presented in Fig. 3b. A tensile deformation of a triangular waveform with a constant amplitude of 50 % strain for 10,000 cycles at a frequency of 1 Hz was applied. The initial position of the sample was fixed at zero force. Nearly twice the strain amplitude (50 %) compared to that of the single-cycle test protocol (26.7 %) was chosen to achieve faster destruction of the internal bonds of the samples. Moreover, the shape of the specimen for fatigue cyclic tests (Fig. 1b), was chosen to exhibit a more rapid strength degradation [30,39] compared to that of the specimen intended for monocyclic test (Fig. 1a). For an easy appreciation of the real

magnitude in the variation of electric resistance during the cyclic test, the results were shown in kilohm ( $k\Omega$ ) instead of the unitless presentation.

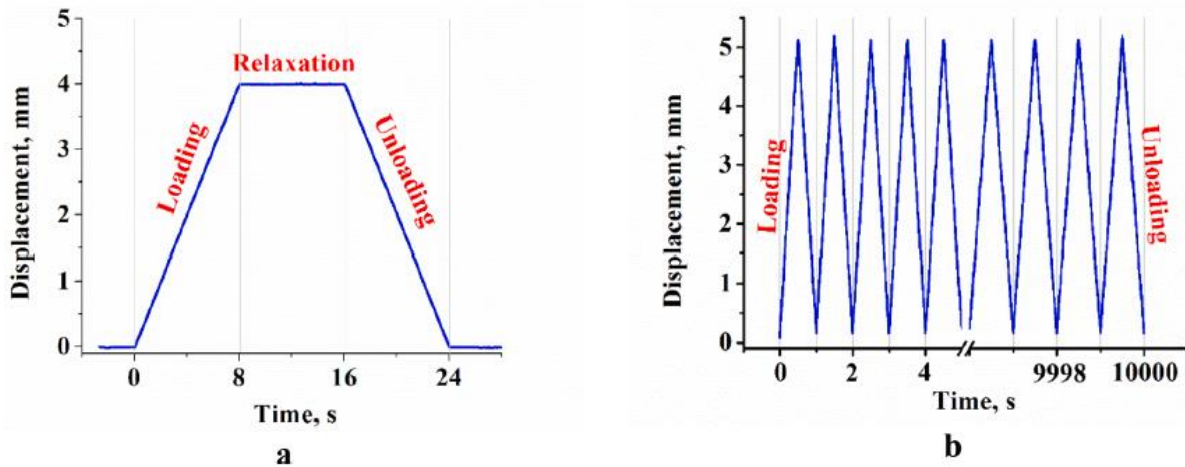


Fig. 3. (a) Monocyclic testing protocol, (b) cyclic testing protocol.

### 3. Results and discussion

Fig. 4 shows the representative curves of (a) applied force as functions of time and (b) electric resistance during all the three phases of the monocyclic testing protocol, for all filler concentrations. From the mechanical point of view, the relation of force versus time (Fig. 4 a) reflected the well-known and expected trend of an increase in force with an increase in the filler content in all loading phases. For all the tested samples, a transition to negative force just before the clamps turned back to their initial position was observed. This force was used to suppress the effects of increased sample length (residual strain) and lasted up to the complete relaxation of the rubber molecules and accommodation of the new entropy state.

From Fig. 4b, it can be seen that as the filler concentration increased, the electrical resistance decreased in all loading phases, following a highly nonlinear pattern. This nonlinearity is quite undesirable for elastomeric sensors. The linear character of the dependence of the resistance on the applied deformation can be traced only during the initial loading phase in the first step and only in the deformation region below 5 %. Further elongation of the samples up to maximum strain, as well as during the relaxation and unloading step, led to nonlinear electric response for the rest of the testing protocol. Relaxation was characterized by non-linear decrease in electrical resistance, and unloading was fully dominated by SPP. It should be mentioned that the complete recovery of the electrical conductivity lasted from tens of minutes to hours, which did not matter in the case of monocyclic test, but certainly influenced the cyclic test.

During the unloading phase, depending on the CB content of the three samples, a clear difference in SPP evolution was observed. Fig. 4b shows a broad shoulder peak for the material containing 50 phr of CB, which started and ended in the first half of this phase. The amplitude of SPP was lower than that of the electrical resistance reached just at the end of the loading stage when the sample experienced maximum strain. After the tensile machine reached its zero-position, a non-monotonous decrease in electrical resistance was present.

In the sample containing 60 phr *CB*, the character of the *SPP* changed significantly compared to that of the sample containing 50 phr *CB*. The electrical resistance reflected a continuous slight increase throughout the strain release time. The amplitude of the *SPP* was more or less the same as the value of the electrical resistance at maximum strain. Such a shape of the *SPP* has not been reported or discussed in any previous scientific study so far. In fact, it could be classified as a monotonic slightly increasing shoulder peak which lasted for the entire unloading phase. Immediately thereafter, the electrical resistance began to drop down, which can be considered a process of long-term recovery of electrical conductivity.

The addition of high 70 phr *CB* to the tested samples challenged all the existing scientific knowledge based on publications with lower filler content. A distinct slope of resistance during loading process was observed for all the tested samples. It is noteworthy that for a sample with 70 phr of *CB*, this slope changed twice, behaving like a material with positive, then negative and again positive piezoresistance. Moreover, the maximum increase in electrical resistance over the entire loading phase did not exceed 20 % of the initial resistance. Such samples are completely useless for sensorial applications. This hitherto apparent uselessness was later converted to a very useful phenomenon where lies the novelty of present work and discussed in details later.

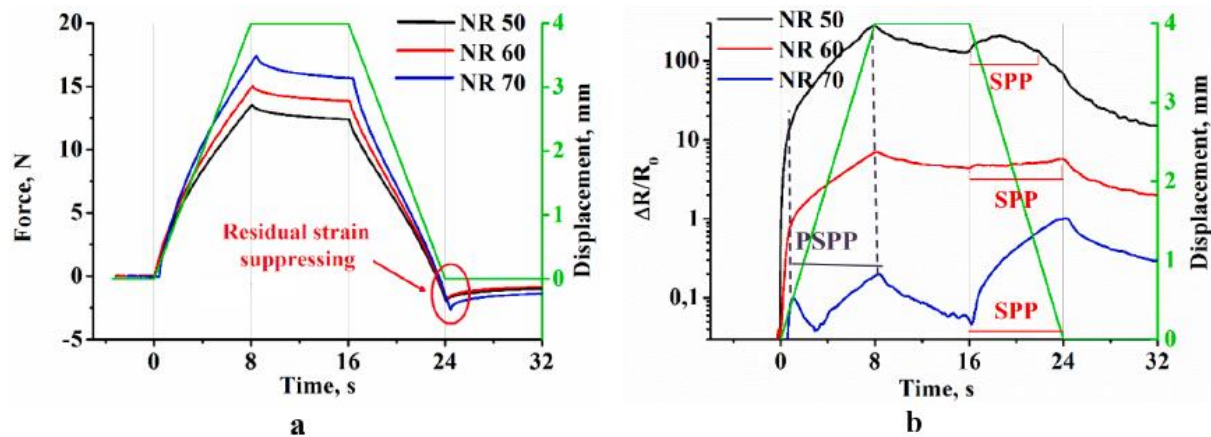


Fig. 4. (a) The force registered during all phases of testing protocol; (b) the relative electric resistance of samples.

During the holding phase, the rubber with 70 phr *CB* showed the most significant decrease in electrical resistance compared to those of 50 and 60 phr. The *SPP* was so pronounced that it exceeded 5 times (500 %) the value of the electrical resistance at maximum strain. To the best of the authors' knowledge, previous scientific studies have never reported an *SPP* amplitude greater than the sample resistance at maximum strain.

It should be noted that the giant *SPP* observed during unloading was somehow signaled by an alternating resistance slope variation during the loading stage (see Fig. 4b). Thus, an evident change in the slope in the figure can be regarded as a newly coined term - precursor of the shoulder peak phenomenon (*PSPP*) and a similarity of the origin of *PSPP* and *SPP* could be expected. The generally accepted explanation of the origin of *SPP* is the competition between network breakdown and network reformation including the orientation effects, such as the alignment and rotation of the *CB* aggregates [40,41]. The detailed explanation of the origin of *SPP* and *PSPP* is beyond the scope of this study. However, in this study, the giant *SPP* as well as *PSPP* were used as parameters to continuously monitor changes in the strength of the highly filled NR under cyclic fatigue test.

As has been mentioned, *NR* samples with the highest filler content exhibited a weak piezoresistive effect and thus, generally cannot be used to self-monitor their integrity by the methods developed earlier for composites with low filler content, as for example in [4]. For the reason mentioned above, special shaped samples (Fig. 1b) containing 70 phr of *CB* were subjected to cyclic fatigue loading. The samples were loaded with a triangular waveform at a loading frequency of 1 Hz, under a constant deformation 50 % up to 10,000 cycles. Cyclic fatigue loading usually simulates the long-term service life of rubber products, and transformation in electrical properties of the material are attributed to changes in filler network [32]. Therefore, in general, a change in piezoresistive response under fatigue loading corresponds to and indicates the mechanical degradation of a rubbery material.

Fig. 5a and 5b show the dependence of force and electrical resistance of the analyzed *NR* sample containing 70 phr *CB* under cyclic loading. For convenience and easier understanding of the degradation trends, only the results obtained at the beginning (Fig. 5a) and the final cycles (Fig. 5b) of the experiment are shown.

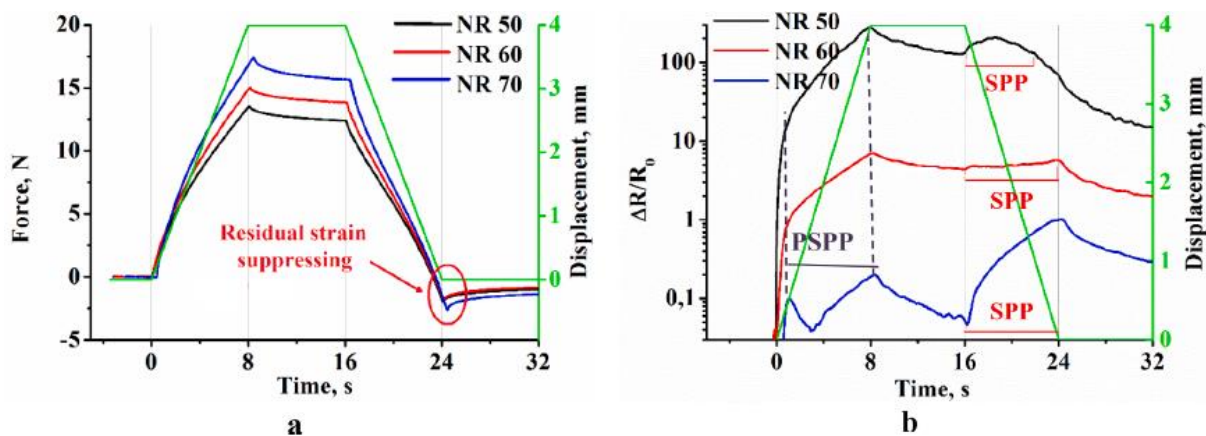
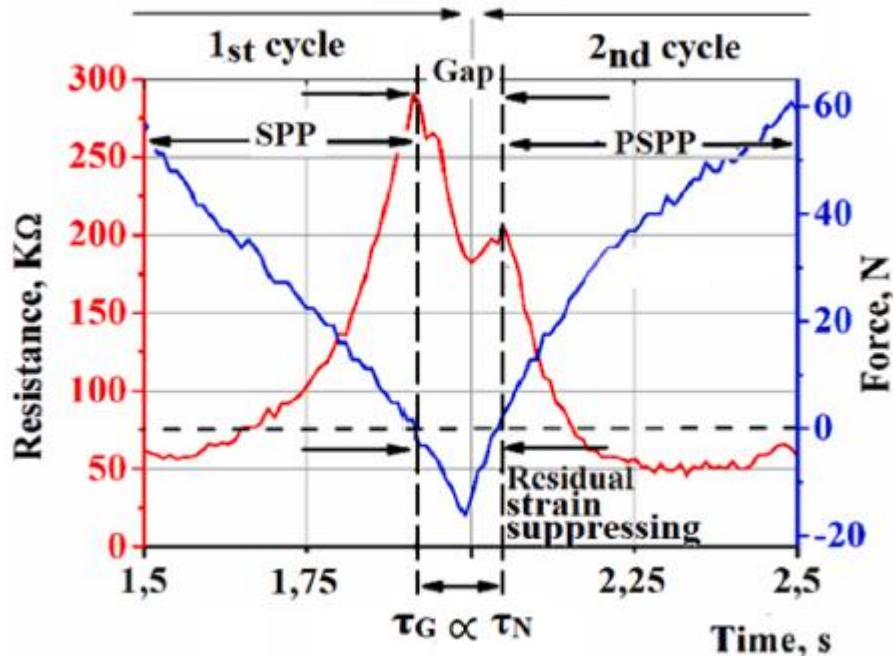


Fig. 5. Electric resistance and force in dependence on loading cycles: (a) for the few initial loading cycles, and (b) for the last few cycles.

In the first, and all subsequent cycles, the maximum deformation of the sample did not result in the maximum resistance as was encountered and described in articles dedicated to low *CB* concentrations. A giant *SPP* is followed by little bit lower, but still immense peak, now termed precursor of shoulder peak phenomenon *PSPP* (see Fig. 5a). To avoid confusion, it should be noted that the period of interest was the interval between the end of the *SPP* and the beginning of the *PSPP* of each two consecutive cycles. This transition time was characterized by a resistance Gap which lasted seconds, and had a similar duration as residual strain suppression time - clearly delimited by "negative" force apparition on loading-unloading curves.

Residual strain suppression actually means the effort of the tensile testing machine (Instron) to fight against the forces developed by "compressed" sample. Due to an increased length attributed to the residual strain, the sample needs to be forcefully fitted in compression until the clamps reach their initial position. For obtained electric signals denoted as Gap, it is immaterial to what happens with the sample during this forceful fitting in various forms such as compression, bending, or twisting, Gap is showing the beginning and the end of fight, where it can be conclusively inferred that an increase in the duration of fight is directly related to an increase in the residual strain. This reflects the beauty of this method.

As has been mentioned, the residual strain fitting is perfectly signaled by two prominent peaks on the electrical resistance curve (**Fig. 6**). The conversion of the cycles into the time easily allows to determine the and for any two consecutive cycles.



**Fig. 6.**  $\tau_G$  and  $\tau_N$  within two consecutive cycles.

The duration of Gap -  $\tau_G$ , as well as the duration of residual strain suppression  $\tau_N$ , gradually increased with the number of cycles. Taking into consideration that an increase in (directly related to residual strain) quantify the degradation of tested material during the cyclic test, it can be stated that Gap duration can indirectly show the same degradation of the material.

This is now explained in some details. Any residual strain reflects to some degree, the amount of detachment between filler-filler particles, rubber-filler particles and also rubber chain scission, all of which are predecessors to the material degradation.

The degradation of material was confirmed by the softening of the tested sample in cyclic test (**Fig. 7**). The force necessary to strain the sample up to set amplitude gradually decreased. Simultaneously, the negative force, suppressing the residual strain exhibited a gradually increasing trend.

The correlation of  $\tau_G$  and  $\tau_N$  in the cyclic test was carefully analyzed. **Fig. 8a** exhibits the experimental results and corresponding fitting curves. Although, the result from **Fig. 5a** and **b** shows a quite negligible deviations between  $\tau_G$  and  $\tau_N$  in the framework of any two consecutive cycles, the small deviations are still there. More pronounced, the fluctuations are visible when  $\tau_G$  and  $\tau_N$  are plotted for whole duration of cyclic test. Such situation is quite normal and often encountered during any experimental measurements. Many measurement set-ups have their build up filtering software to make the results smother. The simple logarithmic fitting gave us a perfect correlation between  $\tau_G$  and  $\tau_N$  as exhibited in **Fig. 8b**. In such a way, both of them can be considered as the indicators of mechanical degradation of material and taken as a new damage parameter in fatigue simulations.

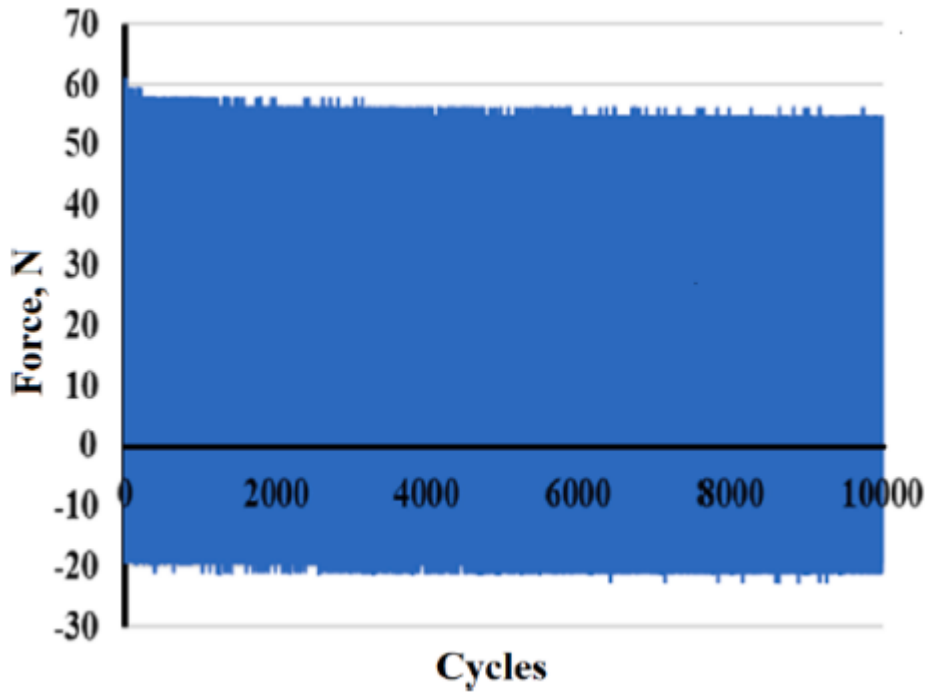


Fig. 7. The cyclic evolution of force necessary to strain the sample up to set value.

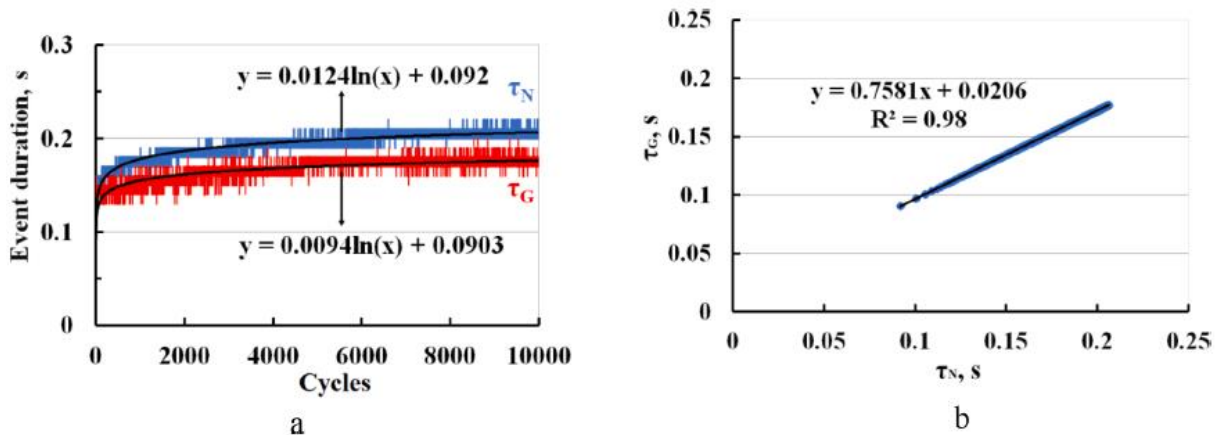


Fig. 8. The cyclic evolution of (a)  $\tau_N$ ,  $\tau_G$  and corresponding fitting curves; (b) The correlation between fitted curves of  $\tau_N$  and  $\tau_G$  and corresponding linear fit.

This fact opens up a real possibility to track negative changes in the mechanical properties of highly filled rubbers, which usually possess an inexpensive piezoresistive effect, by analyzing the evolution of the Gap duration.

#### 4. Conclusions

In the present study, the effects of filler loading at 50, 60 and 70 phr *CB* in a *NR* matrix on piezoresistance were investigated. The increase of filler content showed a high non-linearity in resistance variation as a function of the applied strain, and a continuous decrease in piezoresistive sensibility, which is a major obstacle for self-monitoring of mechanical properties by existing methods.

The undesirable processes for electromechanical applications, such as unexpected drop of resistance under the tension of the samples (*PSPP*), exponential decrease of resistance under constant strain and the subsequent increase in resistance stated as *SPP* during the unloading of the samples, were found. Thus, samples containing 70 phr *CB* exhibited a huge and unusual *SPP* during the unloading process and reached an increase of up to 500 % compared to the resistance at maximum load.

A first attempt was made to convert a giant *SPP* and *PSPP* of two consecutive cycles into the new Gap-based method to successfully monitor the process of mechanical degradation of material in cyclic loading. The duration of Gap - was found to be very close to the duration of residual strain fitting process -  $t_n$ , when the tensile testing machine clamps undergo the transition from positive to negative and from negative to positive forces while moving toward and away the initially set position. If it is assumed that the residual strain is an indicator of internal integrity, an increase in the duration of Gap reflected the state of mechanical degradation and we believe it could be propounded further as a damage parameter for fatigue prediction.

The work proposed a novel perspective for self-monitoring of rubber degradation based on piezoresistivity. This method can be recommended for highly filled rubber products, subjected to cyclic dynamic loading conditions, such as tires, conveyer belts, *V*-belts and the like where strong mechanical reinforcement of the products is achieved only after incorporation of an appropriate high amount of carbon black filler.

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