ISSN : 2249 - 8877 Volume 3 Issue 1

Trade Science Inc.

Research & Reviews On Polymer

Full Paper

RRPL, 3(1), 2012 [23-31]

Conductive rubber based nanocomposites applicable as pressure sensors

Omar A.Al-Hartomy[1,2], Ahmed A.Al-Ghamdi[2], Nikolay Dishovsky[3*], Petrunka Malinova[3], Rashko Dimitrov[3], Farid El-Tantawy[4]

[1]Department of Physics, Faculty of Science, University of Tabuk, Tabuk 71491, (SAUDI ARABIA)
[2]Department of Physics, Faculty of Science, King Abdulaziz University, Jeddah 21589, (SAUDI ARABIA)
[3]Department of Polymer Engineering, University of Chemical Technology and Metallurgy, 1756 Sofia, (BULGARIA)
[4]Department of Physics, Faculty of Science, Suez Canal University, Ismailia, (EGYPT)
E-mail : dishov@uctm.edu

Received: 8th September, 2011 ; Accepted: 8th October, 2011

ABSTRACT

This work reports on the preparation of composites based on different in chemical nature elastomers (natural rubber and siloxane) filled with different amounts of fillers (furnace carbon black, titanium diboride and hybrid combination of furnace carbon black and nanosized nickel powder) and investigation of the changes in their volume resistivity depending on applied pressure on the composites in the regions 0-35 kPa (low pressures) and 0-100 MPa (high pressures) to evaluate their pressure sensitive properties. It has been established that when applied pressure is higher than 10 MPa (in the 0÷100 MPa range) the volume resistivity of all composites studied depends slightly or almost does not depend on pressure no matter what the nature of the elastomer matrix and filler is. It means that the composites are not suitable for pressure sensors at high pressures. In the 0÷35 kPa range all composites are sensitive to pressure changes. In most cases the character of the dependences is linear and reversible. The increase in filler concentration and pressure applied leads to a volume resistivity decrease, i.e. at low pressures some of the composites could be used as pressure sensors in orthopaedics and in other fields of medicine. The composites based on siloxane rubber and filled with carbon black are the most suitable for sensors of all studied.

KEYWORDS

Natural rubber;
Siloxane rubber;
Nanocomposites;
Pressure sensors.

INTRODUCTION

Electrically conductive polymers are subject of intensive studies due to the great interest in their technological applications. On the other hand, being of low stability restricts their practical application to few cases. Alternatively, electrically conductive composites and nanocomposites, have been successfully prepared by adding conductive fillers (such as carbon black or metal powders) to a polymeric material. This kind of materials could be used as sensing elements of flexible pressure sensors, applicable in different fields[1-9], especially in monitory the rock pressure upon the support structures in mines and civil engineering tunnels aiming at prevention of caveins and breakdowns. Running of such activities requires to install a great number of sensors both along the gallery (tunnel) and on its cross section. Having in mind the high price of the sensors used currently, such monitoring is often omitted due to financial reasons. Since the production of pressure sensors based on elastomers involves only technological operations traditional for rubber processing industry, it can be expected to produce sensors at a price much lower than that of the sensors used nowadays. The long lasting stability of the characteristics of those sensors opens possibilities for their usage in devices for long exploitation as control sensors for prevention of accidents, caused by changes in rock pressure. Such sensor could be also used to register the strain-stress state of dam walls and other important facilities subject to different pressure[10]. In the cases above the pressure applied on the elastomer sensor is in the megapascal (MPa) range. There are data of using elastomer based pressure sensors in medicine and orthopedics in particular[3]. In those cases the pressure is much lower – it is within the range of kilopascals (kPa) up to 1MPa. A new tire concept has been also developed. It is based on measuring the pressure inside the wheel by an integrated in the tire pressure sensor. The main innovation in this concept is incorporating the sensor into the tire material. This technology opens the opportunity of producing smart tires in the near future[11].

The major requirements that the materials for elastomer based pressure sensors should meet are:

1. The dependence between the volume resistivity and the applied pressure should be linear in a wide range of pressure changes and reversible, without hysteresis;
2. The change in the volume resistivity with pressure changing should be great enough in order to guarantee that the changed value is not in the error range of the adopted method for measuring the pressure;
3. The volume resistivity of the composite should not change with time, i.e. the properties of the dielectric matrix, as well as those of the conductive phase, should remain constant for a long time; it means that the effects of rubber creep should be kept in mind.
4. The change in the environment temperature should have a negligible effect on the volume resistivity of the sensor material compared to that of pressure.

The study of the relation between external pressure and the composite volume resistivity is the key to fabricating this kind of sensors.

The working thesis of this study is that the introduction of a suitable conductive phase into the rubber matrix at an appropriate amount could ensure that the volume resistivity of the vulcanizates thus obtained will meet the requirements for sensor materials and may be used successfully for production of pressure sensors.

This work reports on the preparation of composites based on different in chemical nature elastomers (natural and siloxane rubber) filled with different types of fillers (furnace carbon black, titanium diboride and hybrid combination of furnace carbon black and nanosized nickel powder) and investigations on their volume resistivity changes depending on external pressure applied with regard to use such of composites as pressure sensors.

EXPERIMENTAL

Materials

Natural rubber/carbon black/nickel powder nanocomposites

NR was purchased from North Special Rubber Corporation of Hengshui, Hebei Province, China. Carbon black, namely N-220, of particle size about 20 nm was used as a reinforcing filler. Other ingredients such as zinc oxide (ZnO), stearic acid (SA), N(1,3 Dimethylbuthyl)-N'-Phenyl-p-Phenylenediamine (Vulkanox 4020, produced by Lanxess), 1,2-mercaptobenzothiazole (MBT) (Vulkacit Merkapto, produced by Lanxess), and sulfur (S) were commercial grades and used without further purification. Nickel powder supplied by Wako Chemical Company, Tokyo, Japan with starting particle size of 10 μm was used as a conductive ferromagnetic filler.

Preparation of nickel nanoparticles

The starting material for ball milling was nickel powder of above 99.9% purity. The nickel powder was placed in a stainless steel vials with stainless steel balls of 10 mm diameter. The ball-to-powder ratio 20:1 was used in the planetary mill (Marconi MA 350 ball mill) at 400 rpm under argon atmosphere for 6 h to obtain nickel nanoparticles of 20 nm average particles size.

Preparation of rubber nanocomposites

Typical formulations of natural rubber composite compounds are presented in TABLE 1. First, mixtures of carbon black with conducting nickel nanoparticles were prepared by grinding them together in a grinding machine for 1 h. The filler mixing with rubber was accomplished in an open two-roll mill under identical conditions of time, temperature, and nip gap, with the same sequence of mixing all compounding ingredients, in order to avoid the effect of processing on the properties. The vulcanization process of the rubber compounds was carried out in an electrically heated hydraulic press using a special homemade mold at 153°C, under pressure of 150 kN/m^2 for 30 min.

TABLE 1 : Formulations of natural rubber based nano composites.

Ingredients, phr	Sample				
	1	2	3	4	5
Natural Rubber (NR)	100	100	100	100	100
ZnO	4	4	4	4	4
Stearic Acid (SA)	1	1	1	1	1
Processing Oil	10	10	10	10	10
Carbon Black / Nickel nanosized powder	0/0	12.5/12.5	25/25	37.5/37.5	50/50
MBT	2	2	2	2	2
Vulkanox 4020 (6PPD)	1	1	1	1	1
Sulfur	2	2	2	2	2

Siloxane rubber/TiB$_2$ nanocomposites

Materials

Siloxane rubber namely ELASTOSIL ® R 401/30 produced by Wacker Chemie AG with high temperature cure, density at 25°C - 1,1 g/cm^3 and hardness of 30 Shore A was purchased from North Special Rubber Corporation of Hengshui, Hebei Province, China. TiB$_2$ with particle size of about 50 nm used as conductive filler was received from Wako Chemical Company, Tokyo, Japan.

Preparation of rubber nanocomposites

The mixing was accomplished in an open two-roll mill under identical conditions of time, temperature, and nip gap, with same sequence of mixing of all compounding ingredients to avoid the effect of processing on physical properties. Several batches with different concentration of TiB$_2$ 0, 10, 15, 20, 30 and 40 phr, respectively, were prepared.

The vulcanization process of the siloxane rubber based compounds was carried out in an electrically heated hydraulic press using a special homemade mold at temperature 195°C and under pressure 150 kN/m^2 for 60 min.

Siloxane rubber / carbon black nanocomposites

Materials

Siloxane rubber namely ELASTOSIL ® R 401/30 produced by Wacker Chemie AG with high temperature cure, density at 25°C - 1,1 g/cm^3 and hardness of 30 Shore A was purchased from North Special Rubber Corporation of Hengshui, Hebei Province, China. Furnace carbon black namely N-220 with particle size of about 20 nm used as conductive filler was received from Alexandria Trade Rubber Company, Alexandria, Egypt.

Preparation of rubber nanocomposites

The mixing was accomplished in an open two-roll mill under identical conditions of time, temperature, and nip gap, with same sequence of mixing of all compounding ingredients to avoid the effect of processing on physical properties. Several batches with different concentration of carbon black 0, 10, 20, 30 and 40 phr respectively were prepared. The vulcanization process of the siloxane compounds was carried out in an electrically heated hydraulic press using a special homemade mold at temperature 195°C and under pressure 150 kN/m^2 for 60 min.

Characterization and measurements

Volume resistivity measurement

Volume resistivity (ρ_v, Ω.m) of the obtained flat rubber based samples, was measured using two electrodes (2-terminal method) and calculated by the equation:

$$\rho_v = R_v \cdot S / h \qquad (1)$$

where:

R_v - measured ohmic resistance between the electrodes, Ω;

S – cross sectional area of the measuring electrode, m^2;

h – sample thickness between the electrodes, m

Determining the volume resistivity at low pressures (0-35 kPa)

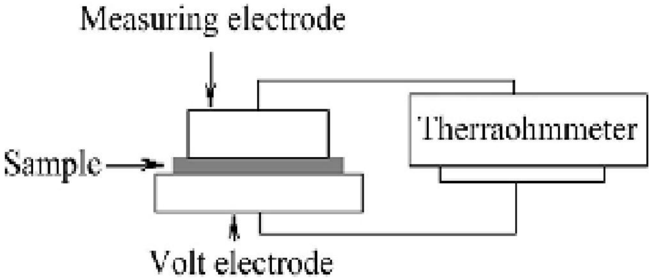

Figure 1 : Scheme of laboratory equipment for volume resistivity measurement at low pressures

The equipment presented schematically on Figure 1 was used to determine the volume resistivity as a function of the pressure applied. The pressure in the given range was changed by using measuring electrodes which different mass was exactly calculated in advance, so that the sample was subjected to 5, 10, 15, 20, 25, 30 and 35 kPa, respectively.

The resistance was measured by a therraohmmeter Teralin III (produced in Germany), using direct current.

The current voltage was 100 V. The resistance values were measured 1 min after the current switching on, when the resistance values was already stabilized. The measurements were carried out at ambient temperature increasing and decreasing the pressure in order to determine the presence or absence of hysteresis. The samples used had a form of a circle with diameter of 90 mm. Their thickness was 2 mm.

Determining the volume resistivity at high pressures (0-100 MPa)

The volume resistivity was determined by the equipment shown in Figure 2. To avoid diameter deformation at high pressures the samples (in the form of cylinders with diameter of 15 mm and height of 25 mm) were placed into a special steel measuring cell (Figure 3) upon which the desired pressure was applied, using the dynamometer shown in Figure 2. The changes in the height of the samples were measured by micrometer and the correct values were used in equation 1.

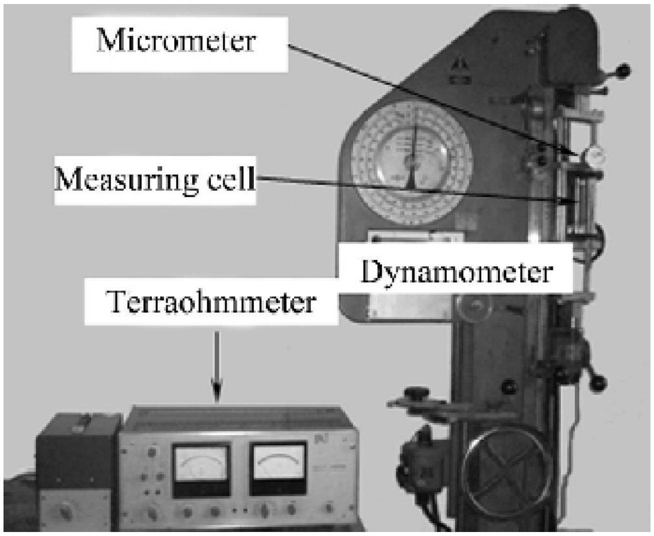

Figure 2 : Laboratory equipment for ρ_v measurements at high pressures

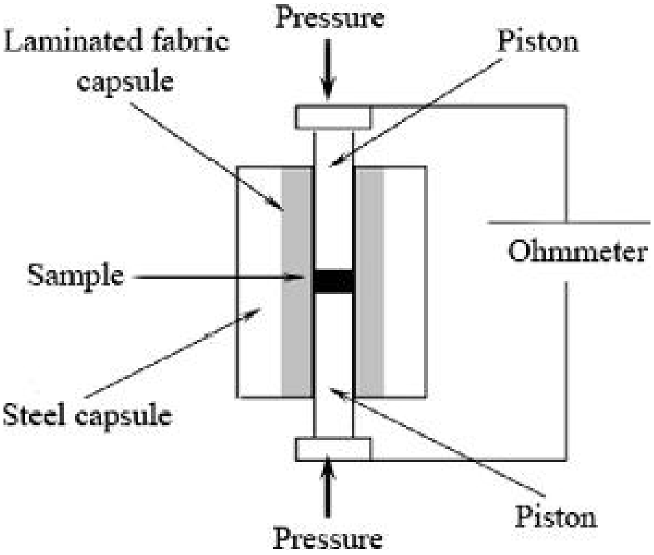

Figure 3 : A measuring cell for determining ρ_v at high pressures

RESULTS AND DISCUSSION

High pressure

Figure 4 shows the dependence of the specific volume resistivity (ρ_v) of a series of natural rubber based vulcanizates as a function of the external pressure applied.

The volume resistivity of the non-filled vulcanizates decreases drastically from 10^{13} to 10^3 Ω.m. in the 0÷30 MPa interval. After that up to 130 MPa its value remains constant. Having in mind that the electro conductivity of the nonfilled elastomers is often realized by so called hopping mechanism, the higher pressure short-

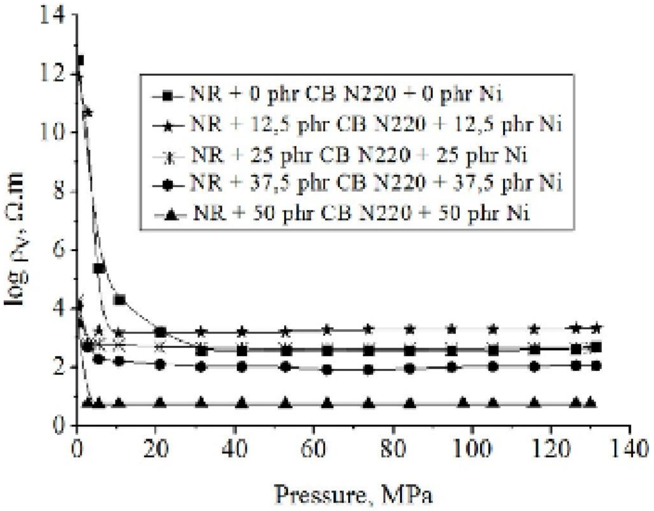

Figure 4 : Volume resistivity (ρ_v) of natural rubber based vulcanizates as a function of the external pressure applied

ens the distance among the macromolecules and as a result less activation energy is needed for electrons hopping, which decreases considerably volume resistivity values in the range 0-30 MPa. However, a further pressure increase stabilizes the electro conductive structures (and so called conductive paths) leading to formation of a more dense change resistant structures under the condition of increasing compression (from 30 till 130 MPa).

From the other hand the structure densification, when 12.5 phr of fillers are added may be connected with filling the free volumes in the rubber matrix which hinder the current carriers transport. For that reason the resistivity of the composite, containing 12.5 phr of fillers increases in comparison to one of the non filled vulcanizates, but the initial volume resistivity (without external pressure) does not change.

The obtained resistivity values are due to the addition of 12.5 phr carbon black and nickel powder to the vulcanizates. Most probably at the given concentration the number of filler particles is small, hence they are just traps for the charge carriers. For that reason the resistivity increases in comparison to the one of the non-filled vulcanizate.

When the vulcanizate is filled at 25 phr, its volume resistivity without external pressure decreases drastically. to 10^4 Ω.m. On one hand the higher filler amount introduces additional charge carriers. On the other hand, the higher filler concentration and the reached percolation threshold are parted by the formation of associates with the macromolecules and new additional conductive paths. The favourable conditions are on account of the finer elastomer membranes resulting from the macromolecules distribution over a larger number of filler particles. The resistivity of this vulcanizate is considerably lower and is affected by the new structure formed. Its initial value of $1.9.10^4$ Ω.m drops to 5.10^3 Ω.m at 5 MPa. The increase in the pressure up to 130 MPa does not change the density of the membranes separating the particles. That is why the resistivity remains unchanged. The higher filler amount of 37.5 phr decreases considerably the initial resistivity value. It decreases to 10^2 Ω.m at 10 MPa and does not change upon greater external pressures. This result allows the assumption that volume resistivity is controlled also by the elastomer membranes. The rigidity of those layers is too high and ensures a constant resistivity at any higher pressure applied.

When the vulcanizate is filled at 50 phr, ρ_v decreases from $3.2\,10^3$ to $5.6.10^0$ Ω.m. in a relatively small pressure range from 0 to 5 MPa. The results obtained about this vulcanizate are due to the wrapping of filler particles in even finer films.

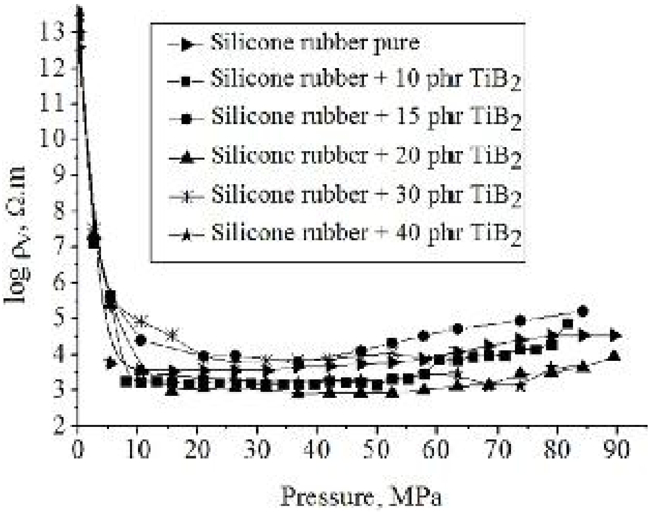

Figure 5 : Pressure dependence of the volume resistivity (ρ_v) for siloxane rubber vulcanizates filled with titanium diboride

All the results show that the increase in the number of conductive particles in the composites increases the number of elastomer-filler associates favouring the current carriers transport. The higher the number of elastomer-filler associates the lower the vulcanizates resistivity due to the thinning of the membranes when their number increases. The fact that the resistivity remains constant after a certain value regardless of the increasing pressure allows the assumption that the structure elements are extremely stable and cannot be broken to let a direct contact between the filler particles. The changes in the resistivity of siloxane rubber vulcanizates filled with titanium diboride caused by compression (Figure 5) follow the pattern of those for natural rubber ones.

The resistivity of those vulcanizates drops drastically in the 0÷5 MPa interval. As seen from the plot the specific volume resistivity of silicone rubber vulcanizates does not change considerably with the increase in the titanium boride amount. The pattern of the curves overlaps with that for the non-filled vulcanizate. The pressure increase up to 10 MPa does not change the resistivity. Obviously the electrical properties of that type of composites are determined chiefly by the elastomer matrix, wherein the electroconductivity is realised by ions and electrons. Under normal conditions the electrons are not a factor determining the resistivity values. But under higher pressure when the dense structure is a dominating factor, the ions transport is hampered. In the case the electrical properties are determined by the electrons movement which is favoured by the better contact between the macromolecules. The considerable drop of resistivity at applied pressure up to 10 MPa is due to this mechanism. When pressure is increased up to 60 MPa the structure does not change its density, therefore the resistivity does not change either. In the 60÷100 MPa interval the density increases considerably and the access to the electron exchange sites is hampered. The result is a resistivity increase.

The composites of siloxane rubber filled with N 220 carbon black have a significantly lower resistivity (Figure 6)

The percolation threshold for those vulcanizates is reached at 20 phr carbon black, determining the specific volume resistivity of 10^3 Ω.m. The non-filled sample and the one filled at 10 phr carbon black reach the percolation threshold at about 10 MPa. Under pressure the resistivity of the non-filled sample and of the ones filled at 10 and 20 phr carbon black increases. The vulcanizates structure becomes denser and hampers the charge carriers transport. This effect reveals the electric charges in the composites to be predominantly ions, despite of their comprising carbon. When the filler amount is increased by 30 and 40 phr the initial resistivity is higher and increases with pressure.

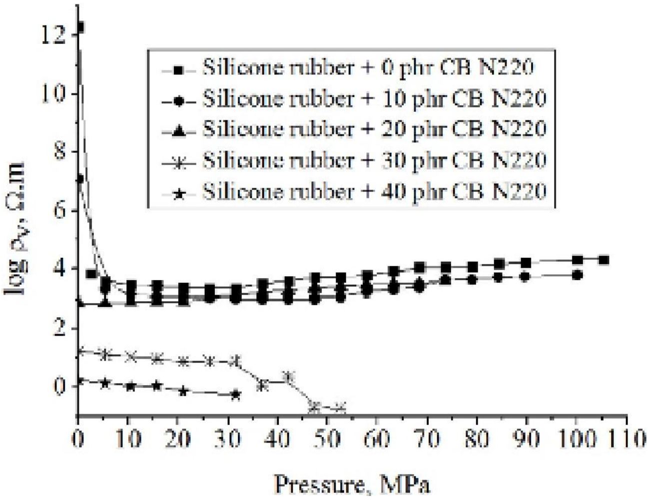

Figure 6 : Pressure dependence of the specific volume resistivity (ρv) for siloxane rubber vulcanizates filled with N220 carbon black

The curves pattern changes into a descending one and demonstrates the transformation of the electric charge from ionic and electronic into only electronic, as the denser structure favours the electrons transport.

The application of high pressure upon those samples is limited by their low tensile strength. They break under pressure of 50 and 30 MPa, respectively. According to one of the theories on elastomer reinforcement[12] the high electrconductivity and the unsatisfactory tensile strength of silicone rubber filled with carbon black is due to the fact that the cohesion interaction between the macromolecules is greater than the adhesion towards carbon black particles. Those interactions do not lead to the formation of elastomer/carbon black associates which are the active structure of enforced rubber. This gives rise to domains of carbon black agglomerates which get into 'dry' contacts between each other. The elastomer barrier membranes do not facilitate those contacts. That favours to a great extent the charge carriers transport. Regarding the tensile properties such a structure is not favourable and is the reason for the low tensile strength of the composites.

The experiments carried out under pressure higher than 10 MPa reveal that the composites investigated are unsuitable for pressure sensors, because the pressure dependence of specific volume resistivity is not linear. Moreover, the resistivity becomes irresponsive to pressure in a wide range.

Low pressure

Figures 7-9 show the specific volume resistivity dependence on pressure in the 0÷35 kPa for the composites studied.

As seen from Figures 7 - 9 in all the cases studied the increase in pressure and in filler amount leads to a decrease of specific volume resistivity, although it happens in a different way for each elastomer and filler. With pressure increase the specific volume resistivity increases very slightly, hardly noticeable, only in the case of non-filled siloxane rubber. It is probably due to the lack of charge carriers and conductive pathways, as well as to the additional densification of the structures. At very low amounts of filler the same effect has been observed by other authors[13]. It can be assumed that at low pressure and low amounts of filler the electroconductivity of the composite is determined by the quantum mechanical tunnelling[14]. With the increasing pressure and filler amount the rubber layers between the highly conductive particles get thinner. That brings them closer and even to the possibilities of contact, hence to lower resistivity. The effect is more pronounced for the samples comprising higher filler amount (20, 30 and 40 phr) wherein under the given pressure the filler particles are able to build denser conductivity pathways, what leads to a more considerable resistivity decrease[15]. In some cases (eg. in composites comprising TiB_2) that could also be an evidence of the transformation of ion conductivity into electron conductivity promoted by the presence of TiB_2. Supposedly TiB_2 is an emitter of electrons in the process of charge carrier transport[16]. As the Figures show the siloxane rubber composites filled with carbon black are most sensitive to the changes in pressure and filler amount what has been also stated by other authors[13]. The effects are less pronounced for the natural rubber based composites filled with a combination of furnace carbon black and nanosized nickel powder. The least effect is observed for the composites comprising TiB_2. In a number of cases at applied low pressure the function resistivity *vs.* pressure for the composites filled with carbon black is linear or

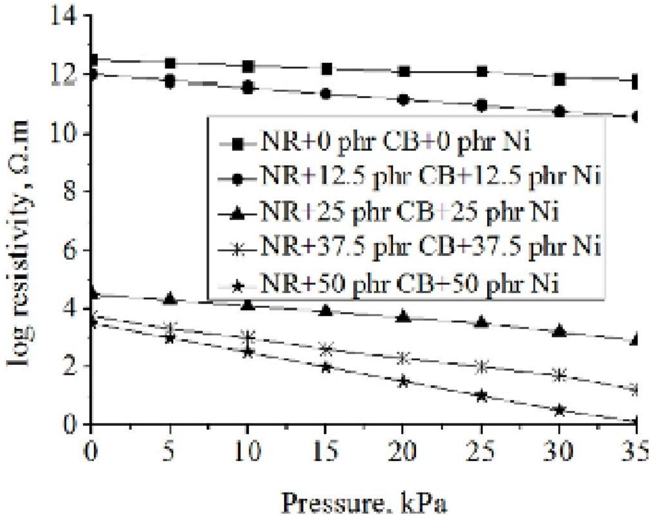

Figure 7 : Volume resistivity (ρ_v) dependence on pressure for natural rubber based vulcanizates filled with a combination of furnace carbon black and nanosized nickel powder

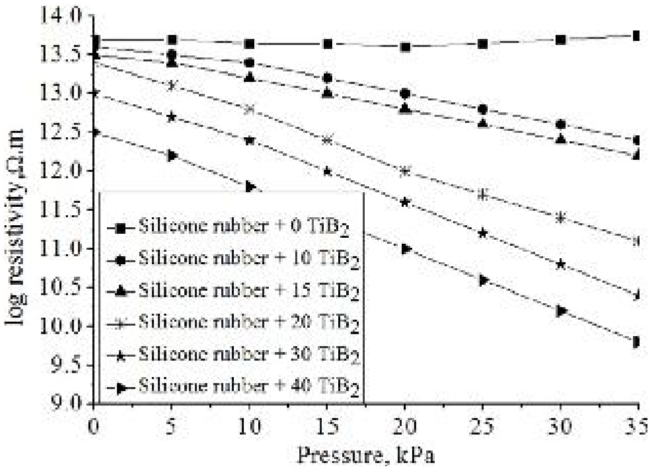

Figure 8 : Specific volume resistivity (ρ_v) dependence on pressure for siloxane rubber based vulcanizates filled with titanium diboride

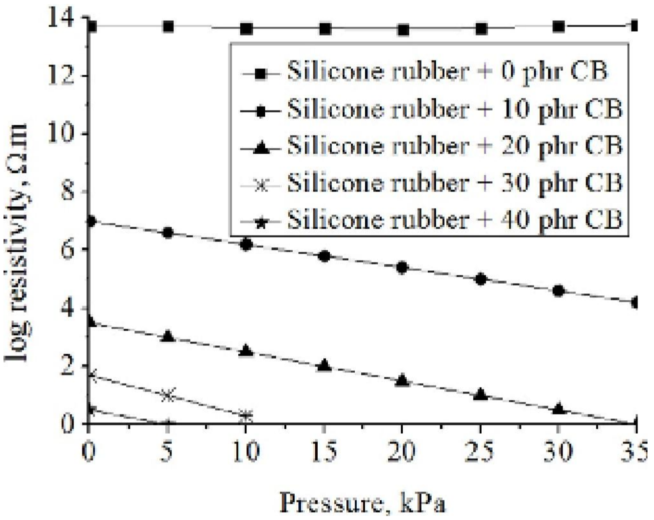

Figure 9 : Specific volume resistivity (ρ_v) dependence on pressure for siloxane rubber based vulcanizates filled with furnace carbon black

very close to linear. The siloxane rubber based composites filled with carbon black are most sensitive to pressure changes. The specific volume resistivity of that type of composites comprising 20 phr carbon black changes about in 4 orders at a pressure increase from 0 to 35 kPa. A similar case are the natural rubbers based composites filled with a combination of furnace carbon black and nanosized nickel powder but at 50 phr. The effect is less pronounced for the composites comprising titanium diboride even at a higher degree of filling. The changes in ρ_v have been studied both at increasing and decreasing pressure but due to the absence of a difference in ρ_v values we present only a case of a siloxane rubber based composite filled with carbon black at 20 phr (Figure 10).

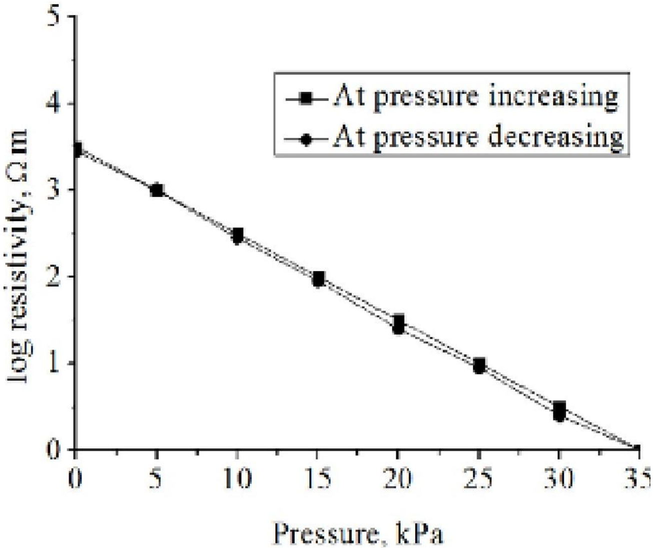

Figure 10 : Specific volume resistivity as a function of gradually increased and decreased pressure for siloxane rubber based composite filled with carbon black at 20 phr

Having in mind the absence of a hysteresis at pressure increase and decrease, it is obvious that some of the composites studied could find application as pressure sensors.

We can consider, that fillers used are incompressible compared with natural and siloxane rubber. Therefore, the external pressure can induce translation and rotation of fillers. To explain the complicated above mentioned phenomena three kinds of changes in the effective conductive paths of the composites as a result of the external pressure applied may be considered[13]:

(1) Change in gap size in existing effective conductive path: The compression makes the gaps between two adjacent conductive particles smaller, leading to the decrease of the electrical resistance of existing effective conductive path.

(2) Formation of new effective conductive paths: The compression and the increased filler concentration make the gaps between carbon black particles smaller, leading to the formation of new effective conductive paths. This effect contributes to the increase of the number of effective conductive path(s).

(3) Destruction of effective conductive paths: The transverse slippage of carbon black, caused by compression, leads to the destruction of effective conductive paths. This effect contributes to the decrease of the number of effective conductive path(s).

The changes in effective conductive paths aforementioned concur during compression. The change (1) and (2) contribute to the decreasing tendency of the composite resistance. The change (3) contributes to the increasing tendency of the composite resistance.

In the investigations, described above, the existence of creep in the material and its effect on the results is not studied entirely. Creep is the change in the deformation of rubber over time from an applied constant force or load. It is also called strain relaxation[17]. When the strain or the strain rates are sufficiently small, the creep response is linear[12]. It is known that the rubber compounding has a great effect on the creep. Some elastomers give better creep resistance than other. The chemical nature and concentration of the filler, the type of crosslinks and the crosslink density have a profound effect on the creep[17]. In our investigations at high pressures as was already described in the experimental part we use a special measuring cell, made of steel, in which the rubber sample was placed to eliminate the effects of creep. At low pressures the effect of creep is not observed.

As a whole the effect of creep on the exploitation properties of rubber based pressure sensors is not discussed in the literature. It will be a subject of our future investigations.

CONCLUSIONS

The changes in volume resistivity of natural rubber and siloxane rubber based composites filled with different amounts of carbon black, titanium diboride and hybrid combination of carbon black and nickel powder depending on applied external pressure in the regions 0-35 kPa and 0-100 MPa have been studied to evaluate their pressure sensitive properties.

1. It has been established that when applying pressure higher than 10 MPa (in the 0÷100 MPa range) the specific volume resistivity of all filled composites studied depends slightly or almost does not depend on pressure no matter what the nature of the elastomer matrix and filler is. That is why the composites of the types studied are not suitable for pressure sensors. The character of the dependences is determined by the fact whether the respective filler concentration is bellow or above the percolation threshold.
2. In the 0÷35 kPa range all composites studied are sensitive to pressure changes. In most cases the character of the dependences is linear. The increase in filler concentration and pressure applied leads to a resistivity decrease, i.e. in this pressure range some of composites could be used as sensors in orthopaedics and in other fields of medicine.
3. The composites based on siloxane rubber and filled with titanium diboride are the least sensitive to pressure changes and are the most unsuitable for sensors. The composites based on siloxane rubber and filled with carbon black are the most sensitive, next come those based on natural rubber filled with a combination of carbon black and nanosized nickel powder. Those composites, especially the former are the most suitable for sensors of all studied. That is confirmed by the linear and reversible dependencies (lack of hysteresis) in their case.
4. When choosing composites for pressure sensors one should take account of the fact that the most suitable are those whose filler amount is about the percolation threshold, preferably a little bit bellow it. At filler concentrations quite bellow the percolation threshold the pressure sensitivity of the composite lowers which lessens its prospects for sensor usage. Reaching and passing the percolation threshold is a crucial factor determining the suitability of the composite to be used as a pressure sensor. The results of our studies reveal that the composite based on siloxane rubber comprising carbon black at 20 phr is the most suitable for sensor applications.

ACKNOWLEDGEMENT

The present research is a result of an international collaboration program between University of Tabuk, Tabuk 71491, Kingdom of Saudi Arabia and the University of Chemical Technology and Metallurgy, 1756 Sofia, Bulgaria. The authors gratefully acknowledge the financial support from the University of Tabuk.

REFERENCES

[1] W.E.Mahmoud, A.M.Y.El-Lawindy, M.H.El Eraki, H.H.Hassan; Sensors and Actuators A, Phys., **136(1)**, 229-233 **(2007)**.
[2] T.H.Ding, L.H.Wang, P.Wang; J.Polym.Sci.Part B, Polym.Phys., **45**, 2700–2706 **(2007)**.
[3] E.E.Job, F.A.Oliveira, N.Alves, J.A.Giacometti, L.H.C.Mattoso; Synthetic Metals, **135**, 99-100 **(2003)**.
[4] L.H.Wang, T.H.Ding, P.Wang; Sens Actuators A, Phys., **135(2)**, 587-592 **(2007)**.
[5] A.C.Clark, S.P.Ho, M.LaBerge; Tribology, **39(11)**, 1327-1335 **(2006)**.
[6] M.Knite, V.Teteris, A.Kiploka, J.Kaupuzs; Sens Actuators A, Phys., **110(1-3)**, 142-149 **(2004)**.
[7] M.H.El Eraki, A.M.Y.El Lawindy, H.H.Hassan, W.E.Mahmoud; Polym.Degrad Stab., **91(7)**, 1417-1423 **(2006)**.
[8] X.J.Wang, D.D.L.Chung; Sens Actuators A, Phys., **71(3)**, 208-212 **(1998)**.
[9] L.H.Wang, T.H.Ding, P.Wang; J.Polym.Sci., Part B, Polym.Phys., **46(11)**, 1050-61 **(2008)**.
[10] T.Blythe, D.Bloor; Electrical Properties of Polymers, Cambridge, University Press, **(2008)**.
[11] L.Valenta, J.Hallas; Tire Technology International, Annual Review, 14-1 **(2010)**.
[12] Ed.J.Mark; The Science and Technology of Rubber, Elsevier, Amsterdam, **(2005)**.
[13] W.Luheng, D.Tianhuai, W.Peng; Carbon, **47**, N14, 3151-3157 **(2009)**.
[14] R.D.Sherman, L.Middelman, S.Jacobs; Polymer Engineering and Science, **23**, N1 321-327 **(1983)**.
[15] Y.Ishigure, S.Iijima, H.Ito, T.Ota, H.Unuma; Journal of Materials Science, **34**, 2979-2985 **(1999)**.
[16] Z.Todorova, F.El-Tantawy, N.Dishovsky, R.Dimitrov; Journal of Elastomers and Plastics, **39**, N1 69-80 **(2007)**.
[17] Ed.J.S.Dick; Rubber Technology, Hanser Publishers, Munich, **(2001)**.