------Raf. Jour. Sci., Vol. 19, No.1, pp 88-97 , 2008------

The Effect of Silica Filler and Aging on Breakdown Voltage and Some Mechanical Properties of Styrene-Butadiene Rubber(SBR)/Silica

Sanaa M. Al-Delaimy
Department of Physics
College of Education
Mosul University

(Received 15/1/2007, Accepted 16/7/2007)

ABSTRACT

The effect of silica filler percent on breakdown voltage(V_{BD}) and some mechanical properties (Tensile strength (T.S),Ultimate elongation (E%)and Equilibrium swelling (Q%)) of Styrene–Butadiene rubber filled with (20,40,60,80,100) phr (part per hundred rubber) of silica have been investigated. It was found that breakdown voltage and tensile strength were increased, while ultimate elongation and equilibrium swelling were decreased with increasing of silica filler percent. The effect of thermal aging on these properties was investigated. It was found that breakdown voltage, equilibrium swelling and ultimate elongation were slightly decreased while tensile strength was increased with aging time. The largest effect during aging appeared on samples with smaller filler loading. The same tests have been done for all samples after2 years (i.e Natural aging). It was found that breakdown voltage, ultimate elongation and tensile strength were slightly decreased while equilibrium swelling was increased compared to thermal aging results, although that the effect of both agings is the same on each property for samples with different silica filler percents.

تأثير حشوة السليكا وزمن التعتيق على جهد الانهيار وبعض الخواص الميكانيكية لمطاط الستايرين بيوتادايين/سليكا

الملخص

استخدم مطاط الستايرين ـ بيوتادايين المحشو بمادة السليكا بواقع phr (20,40,60,80,100) جـزء لكل مائة جزء من المطاط ، لدراسة تأثير نسبة الحشوة على الخواص (جهد الانهيار ، الشد ، الاستطالة عند القطع والانتفاخ المتزن). وجد أن كل من جهد الانهيار والشد يزداد مع ازدياد نسبة حشوة السليكا، أما كل من الاستطالة والانتفاخ المتزن فانه يقل مع زيادة الحشوة.

درس أيضا تأثير زمن التعتيق الحراري (thermal aging) على الخواص نفسها ولـنفس النـسب ووجد بأن كل من جهد الانهيار، الاستطالة عند القطع والانتفاخ المتزن يقل أما الشد فانه يزداد ، مع زيادة

الزمن وبمقادير تختلف مع اختلاف نسبة حشوة السليكا ويبدأ التأثير الأكبر عند نسبة الحشوة الأقل (20phr) وهكذا.

اختبرت المادة أيضا تحت تأثير زمن التعتيق الطبيعي (natural aging) ووجد بأن الخواص (جهد الانهيار، الاستطالة عند القطع والشد) تقل بشكل طفيف أما الانتفاخ المتزن فانه يزداد مقارنة بتأثير زمن التعتيق الحراري مع بقاء تأثير زمن التعتيق للحالتين نفسه على جميع الخواص للنماذج ولجميع نسب الحشوة.

INTRODUCTION

Synthetic polymers have found extensive applications as electrical insulators, for example, cables, they have wide applications ranging from power to control cables. A good combination of flexibility and strength is an essential requirement of cable insulator.

A continuous effort is being made to improve the mechanical and thermal endurance properties of cable insulators and also the electrical properties (Coran,1989).Styrene-Butadiene Rubber (SBR) is now extensively employed for insulating and sheathing where it has an economic advantage over natural rubber (Lee et al., 1994).

Fillers play an important role in improving the physical properties of elastomers. Filling rubber with various fillers is an easy way to obtain a wide spectrum of applications.The physical properties of filled rubber can be easily varied by changing the type, quantity and mixture composition of filler.

Carbon black and silica are the most widely used fillers in the rubber industry (Donnet, 1998, Bishai et al., 2003). Recently, silica was used as partial, or even complete replacement for carbon black in many products (Schaal et al., 2000). It is also the most important material used as a filler to increase the insulation of rubber (Turov et al.,1995).

Materials commonly considered to be insulators can breakdown under high electrical voltage. Breakdown occurs only when extremely high voltage gradients are encountered A very strong electric field can be sufficient to disrupt the induced dipoles in the insulator, and when the strength of the field exceeds the strength of the dipole, rupture may occur (Van Vlack, 1970).

Dry mineral fillers like tricomponent system HRH (Hydrated silica, Resorcinol , Hexamethylene tetraminel) are acceptable for achieving good electrical properties such as resistivity, breakdown voltage, permitivity and dielectric. However, some fillers are excluded from electric applications due to water absorption from the humidity of the air (Coran et al.,1971). A strong hydrogen bonding between water and hydroxyl groups on the mineral surface compete with any possible bond between the polymer and the filler. Even if the composite is prepared with perfectly dry filler, water will be able to reach the interface by diffusion through the polymer (Ahmad and Mark,1998). This interfacial water is an important subject in composite technology, as it seriously affects the mechanical and the dielectric properties of these materials. Interfacially absorbed water weakens the filler matrix interaction, and may in some cases even cause complete debonding due to hydrolysis, in which case the mechanical properties are strongly degraded (Ahmad and Mark, 1998, Mark et al.,1994).With respect to dielectric properties an increase of the permitivity and the dielectric loss of composites, after absorption of water, have been reported by several authors (Woo and Piggot,1988, Ward el al., 2001, 2003) .

The effect on the mechanical and/or electrical properties of aging on natural and synthetic rubber loaded with either black(Carbon or Graphite)or white (Kaolin, Quartz, PVC, or Talk) fillers have been studied (El-Nashar and Turky, 2003, Ismail and Turky, 2001, Shanmugharaj and Bhowmick, 2004 , Ding et al.,1996) .

The aim of this work is to study the effect of silica filler concentration on breakdown voltage and on equilibrium swelling of SBR. Moreover, the effect of thermal and natural aging on the mentioned and some other mechanical properties has been investigated .

Experimental Work

Material used in this study was supplied by Jabber-Bin-Hayyan company. It is Styrene-Butadiene Rubber (SBR) filled with (20,40,60,80,100) phr (part per hundred rubber)of silica filler. Sheets of this product were cut into samples with dimensions depends on the kind of test to be done .

Breakdown voltage was measured for the samples with different silica percents and (2 x 10 x 10) mm dimension using [D.C. Test Set, Model T(103) BICC - Test - Instruments].

The equilibrium swelling in toluene (Q%) for all samples was carried out according to the standard method (Shavarts, 1957). It can be calculated according to the equation :-

$$Q\%=[(W_s-W_d)/W_d]\times 100 \qquad (1)$$

Where Ws is the weight of the swelled sample, and Wd is the weight of the dried sample .

Tensile strength and elongation at break were determined using an electronic ZWICK testing machine (model 1425)-Germany .The samples preparation was mentioned before (Al-delaimy,2007) .

All the previously investigated samples were exposed to normal weathering conditions for two years , and all the mentioned properties remeasured again .

A new set of samples from SBR with (20,40,60,80,100) phr silica were thermally aged at 90±1°C using the oven method (ASTM,1957) for different successive periods up to 7-days , and a new measuring for the mentioned properties has been done after aging .

Results and Discussion

The relation between the breakdown voltage and the composite SBR with different silica filler percent is displayed in Figure (1).It is clear that the breakdown voltage increases with the increasing of filler percent within the studied percentages. This behavior is attributed to the filling material which is insulating material, with resistivity equal to 10^{20} Ω- cm while SBR resistivity is equal to 10^{14}Ω-cm (Van Vlack,1970), therefore increasing its concentration in the SBR composite will increase the insulation of the material as a whole .

The equilibrium swelling (Q%) in toluene was determined and illustrated graphically in Figure (2) versus silica percent in SBR .The Figure shows that (Q%) decreases with an increasing of silica filler percent in SBR. This is due to the ability of interaction of the filler particle with SBR composite. So, by increasing the concentration of silica in SBR there will be no enough space for toluene molecules to enter in-between the two materials .This is in agreement with the literatures that silica fillers have a good interparticle intractions with rubber (Coran ,1989 ,Zheng et al., 1997 and Zhang et al., 2001) .

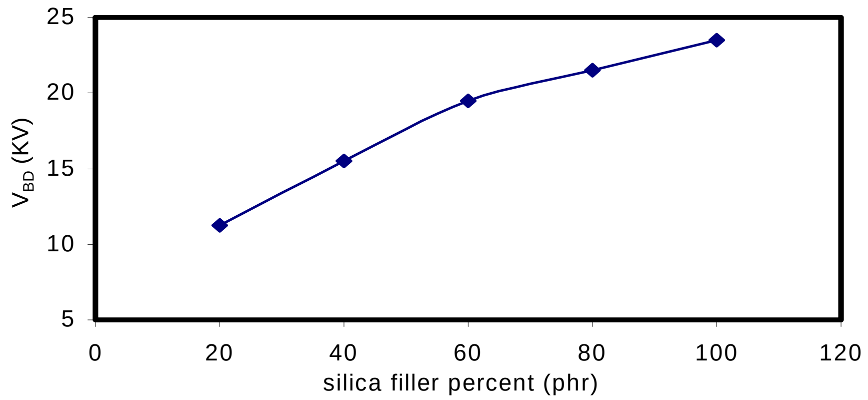

Fig1: Break down voltage versus silica filler percent in SBR

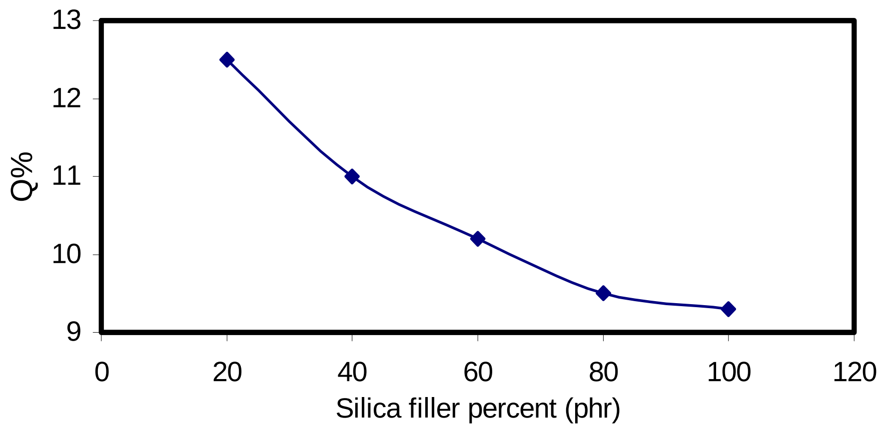

Fig 2: Equilium swelling in toluene verses silica filler percent in SBR

Tensile strength and elongation at break versus silica percent in SBR were displayed in Figures (3&4) respectively. It is clear in Figure (3) that tensile strength increases with the increase of silica filler percent, while elongation at break is sharply decreased up to (60) phr, then a slight decrease occur with a further increase of silicon percent . This is due to the change in rubber concentration percentage which is higher in the upper part than the lower part of the curve (Fig. 4).

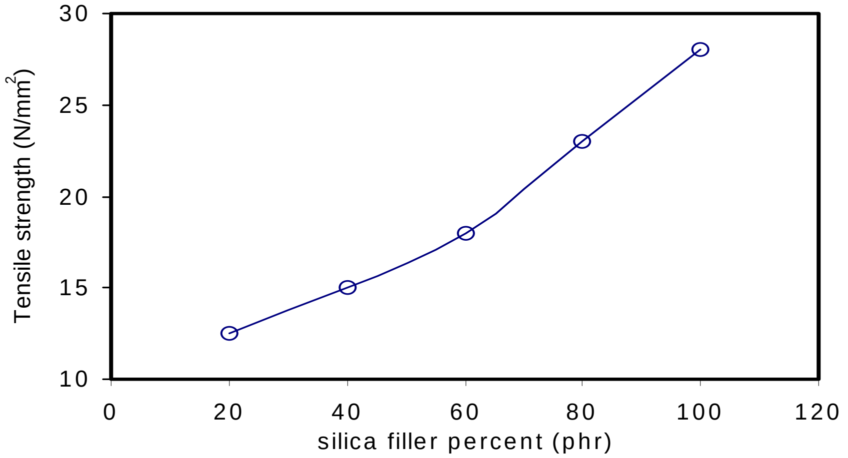

Fig 3: Tensile strength versus silica filler percent in SBR

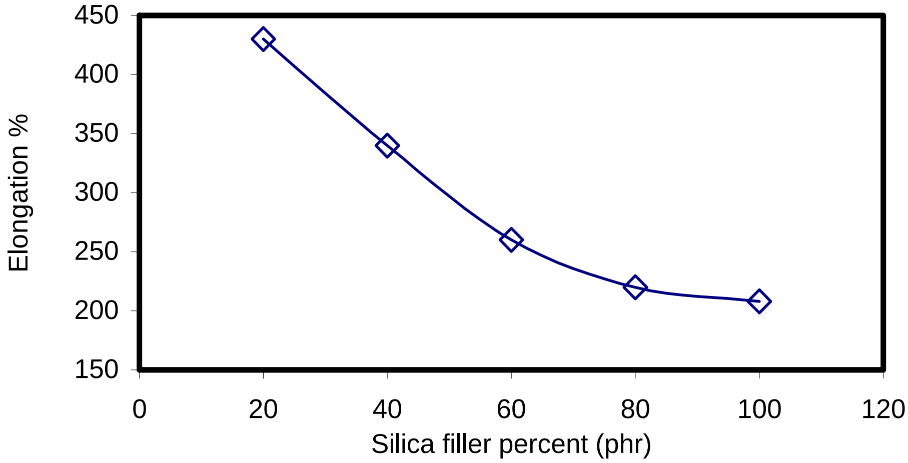

Fig 4: Elongation versus silica filler percent in SBR

The effect of thermal aging on the properties of a new set of samples with the same silica filler percent was investigated and illustrated graphically as follows :

Figure (5) represent the effect of thermal aging on breakdown voltage. It is clear that the voltage slightly decreases with the increasing of aging time for SBR loaded with silica concentration up to 60 phr which may attributed to the formation of cross linked molecules during thermal aging.

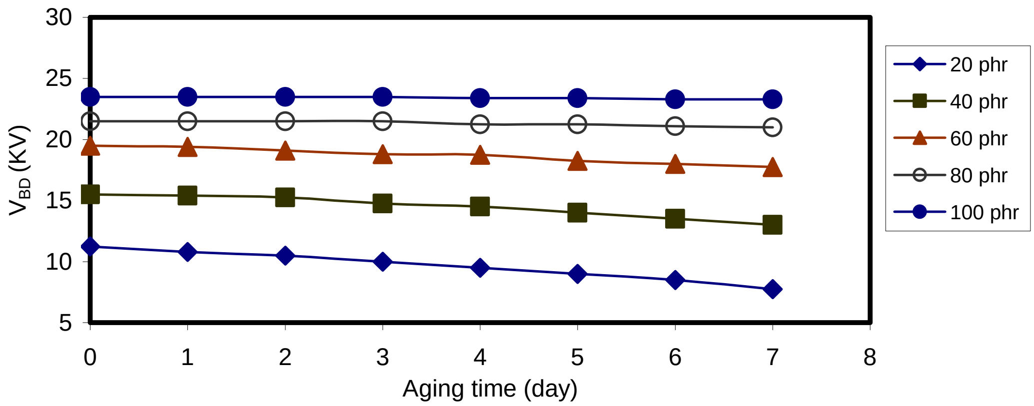

Fig 5: Break down voltage versus thermal aging time for SBR/ silica

The decrease in breakdown voltage, through aging time depend on rubber percent which is seem to be the most affected constituent by aging.

Figure (6) display the equilibrium swelling (Q%) of SBR composite filled with different percent of silica filler versus aging time. The figure shows that (Q%) decreases with time. The decrease of (Q%) is higher for low filler percent samples than that with high percent during aging period. This may be attributed to the reduction of water molecules those exit by heat process during aging.

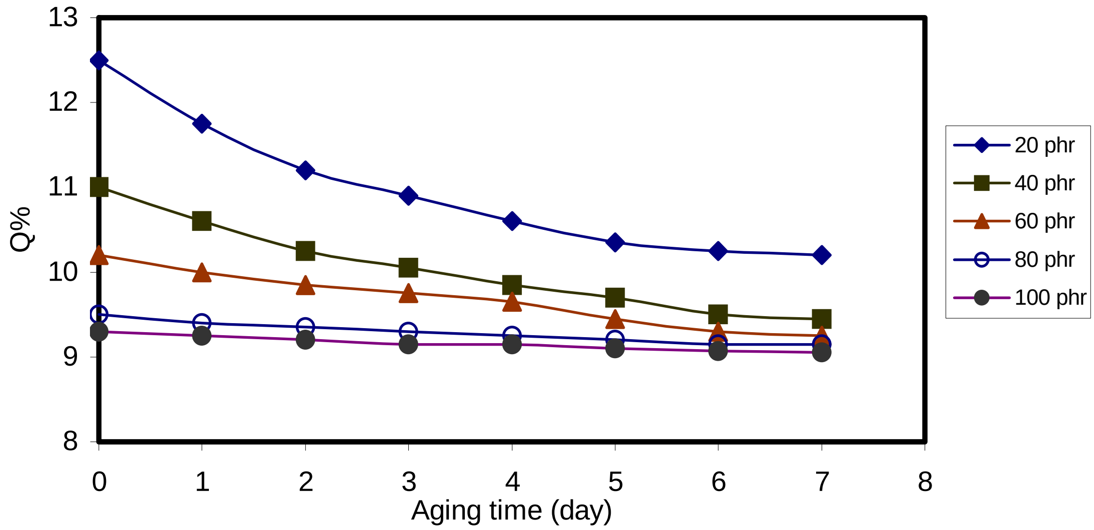

Fig 6: Equilibrium swelling versus thermal aging time for SBR/ silica

Tensile strength versus thermal aging time was illustrated in Figure (7). It is clear that tensile strength slightly increases with time especially at the high silica percent . This could indicate that the interparticle interaction power increases with the increased aging time, this may be attributed to formation of more cross linked molecules which improves the forces between SBR and silica. This phenomenon agree with the results of (Zheng et al., 1997, Bishai et al., 1976).

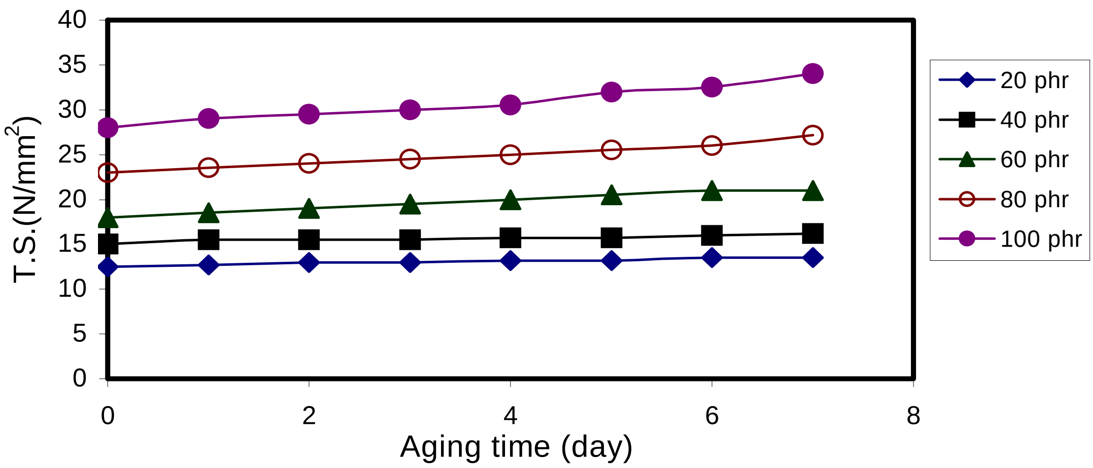

Fig 7: Tensile sreangth versus thermal aging time for SBR/ silica

Figure (8) shows the relation between ultimate elongation and aging time for different silica filler percent . Elongation decreases with aging time at low silica percent because of the high concentration of rubber material which represent the main constituent affected by heat. While elongation is nearly steady with aging time at high silica percent in-which further formation of cross-linked molecules balance the degradation occurs during aging .

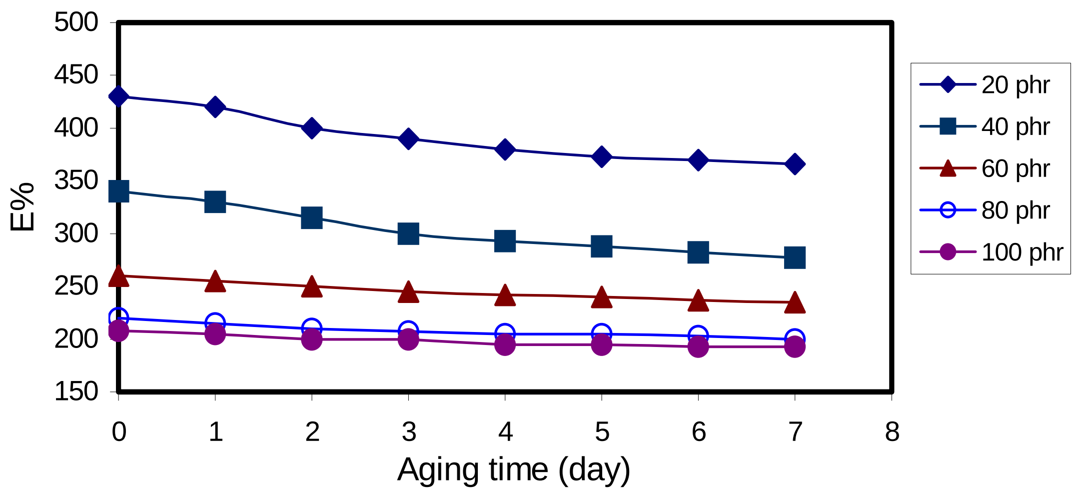

Fig 8: Elongation at break versus thermal aging time for SBR/ silica

All properties for the firstly investigated samples were remeasured after exposing the samples to normal weathering conditions for about two years (natural aging). The data obtained before aging ,after thermal aging and natural aging for SBR/silica composite were listed in tables (1),(2) and (3) respectively .

The compare son between thermal and natural aging , shows that there is a little increment in breakdown voltage , ultimate elongation and tensile strength in thermal aging and a decrement in equilibrium swelling, although the effect of both aging is the same on all properties, this is due to humidity of the weather which effect the natural aging and not exist in the thermal one. Similar behavior has been noticed before in case of SBR-PE (Polyester Short-Fiber) (Bishai et al.,2003) and others (Woo and Piggot,1988) .

Table1:The properties of SBR/silica before aging

Silica percent	20phr	40phr	60phr	80phr	100phr
Breakdown voltage(Volt)	11.25	15.5	19.5	21.5	23.5
Equilibrium swelling (%)	12.5	11	10.2	9.5	9.3
Ultimate elongation (%)	430	340	260	220	208
Tensile strength (N/mm^2)	12.5	15	18	23	28

Table2 : The properties of SBR/silica after thermal aging (at the 7^{Th} day)

Silica percent	20phr	40phr	60phr	80phr	100phr
Breakdown voltage(Volt)	7.75	13	17.75	21	23.3
Equilibrium swelling (%)	10.2	9.45	9.25	9.15	9.05
Ultimate elongation (%)	366	277	235	200	193
Tensile strength (N/mm^2)	13.5	16.2	21	27.2	34

Table 3: The properties of SBR/silica after natural aging (for 2 years)

Silica percent	20phr	40phr	60phr	80phr	100phr
Breakdown voltage(Volt)	6.8	11.5	16.5	20.5	23
Equilibrium swelling (%)	10.7	9.7	9.5	9.3	9.1
Ultimate elongation (%)	360	272	231	198	190
Tensile strength (N/mm^2)	13	15.5	20.5	26.5	33

CONCLUSIONS

1-Breakdown voltage and tensile strength of SBR composite filled with silica, increase, while elongation at break and equilibrium swelling decrease, with the increase of silica filler percent .

2-Tensile strength increases while breakdown voltage, equilibrium swelling and elongation at break decrease with aging time for SBR /silica during thermal aging process.

3-The effect of aging depends on the percent of silica filler. It was found that SBR sample with lower silica percent was the most affected one while the highest silica percent is less affected .

4- All the investigated properties were behaved in natural aging the same way as in thermal aging but there is a slight decrease in tensile strength, ultimate elongation and breakdown voltage, and increase in equilibrium swelling compared to results of those properties by thermal aging .

5-The studied properties of SBR composite as insulating material were enhanced by adding silica filler material within the studied circumstances.

المصادر العربية

الدليمي، سناء محمود حسين، 2007، دراسة تأثير نسبة حشوة السليكاعلى بعض الخواص الميكانيكية والتوصيلية الكهربائية لمادة مطاط الستايرين-بيوتادايين/سليكا، مجلة التربية والعلم، المجلد 19، العدد (1)، ص155-165.

REFERENCES

Ahmad, Z., Mark, J.E., 1998, Study of Hydrated Structures on the Surface of Polymeric Materials, Mater. Sci. Eng. ,C., Vol.(2-3), pp. 183-192.

ASTM-D 573-53 ,1958, For Thermal Oxidative Aging .

Bishai, A.M., Ghoneim, A.M., Ward, A.A.M. ,and Younan, A.F., 2003,Electrical Conductivity of SBR/Polyester Short Fiber Reinforced with Different Type of Carbon Black ,J. Polymer- Plast. Technol. and Eng. ,Vol.42, pp. 1257-11265 .

Bishai, A.M., Hakim, I.K., and Hanna, F.F., 1976, Electrical Conductivity of Styrene-Butadiene Rubber /Polyester ,Plaste and Kautshcuk ,Vol.24(8), pp. 585-592.

Coran, A.Y.,1989, New Components For Rubber Compounds, Plaste and Kautshcuk ,Vol.26 ,pp. 10-19 .

Coran,A.Y.,Boustany, K., and Hamed, P., 1971, Unidirectional Fiber-Polymer Composite: Swelling and Modulus Anisotropy, J. Appl. Polymer Sci.,Vol.15, pp. 2471-2480 .

Ding ,R., Leonov, A.I.,and Coran, A.Y., 1996, A Study of the Valcanization of the Kinetics of an Accelerated – Sulfer Compound, Rubber Chem. Technol., Vol.69, pp. 81-89 .

Donnet,J.B., 1998, Black and White Fillers and Tire Compound ,Rubber Chem. Technol., Vol.71, pp. 323-330 .

El-Nashar, D.E., and Turky, G.M., 2003, The Effect of Mixing Conditions and Chemical Cross-Linking Agents on the Physicomechanical and Electrical Properties of Rubber, Polymer-plastics Technology and Engineering , Vol.42, pp. 269-279.

Ismail, M.N., and Turky, G.M., 2001, Effect of Filled and Valcanizing systems on the Physicomechanical and Electrical Properties of Epdm Valcanizates, Polymer-Plastics Technology and Engineering ,Vol.4 , pp.158-164.

Lee, S., Pawlowski, H., and Coran, A.Y., 1994, Method For Estimating the Chemical Cross-Link Densities of Cured Natural Rubber and Styrene-Butadiene Rubber , Rubber Chemical. Technol.,Vol.67, pp. 854-862 .

Mark, J.E., Erman, B., Eirich, F.R., 1994, Science and Technology of Rubber , 2nd ed., Academic Press: New York, 688 p.

Schaal, S., Coran, A.Y., and Mowdood, S.K., 2000, The Effects of Certain Recipe Ingredients and Mixing Sequence on Rheology and Pocessability of Silica and Carbon Black Filled Tire Compound, Rubber Chem. Technol., Vol. 73, pp. 240-256 .

Shanmugharaj, A.M., and Bhowmick , A.K., 2004, Rheological Properties of SBR Filled with Electron Beam Modified Surface Treated During Phase Fillers, Radiation Physics and Chemistry ,Vol. 69, pp. 91-104.

Shavarts, A., 1957 , Kautshcuk – I Resina, Vol. 7 ,pp. 31-47 .

Turov, V.V., Lebodaa, R., Bogillo,V.I., Skubiszewaska, Z., 995, Structure of Adsorption Center on a Cabosil Surfaces , J. Langmuir, Vol. 11, pp.931-938.

Van Vlack , 1970, Elements of Materials Science , Addison–Wesley Comp. , 445 p.

Ward, A.A.M., Ghoniem, A.M., Younan, A.F., Bishai, A.M., 2001, Mechanical and Dielectric Properties of SBR/Polyester Short-Fiber Composite, International J. of Polym. Material, Vol..48, pp. 355-369 .

Ward, A.A., Stoll, B., Soden, W.V., Haus, S.H., Bishai, A.M., Hanna, F.F., 2003, Studies on Dielectric Behavior of Silica Filled Butyl Rubber, J. of Macromolecular Science, Part B: Physics, Vol. 42, pp. 1265-1274.

Woo, M, and Piggot, R., 1988, Study of Moisture Absorption by Organic Materials, J. Composites, Vol. 10, pp.16-32 .

Zhang,Y., Ge, S., Tang, B., Koga, T., Rafailovich, M.H., Sokolov, J.C., Peiffer, D.C. ,Li, Z., Dias, A.J., McElrath, K. O., Lin , M. Y. , Satija, S. K. , Urguhart, S. G., Ade, H.,and Nyuyen, D., 2001, Effect of Carbon Black and Silica Fillers In Elastomer Blends ,J. Macromolecules ,Vol. 34, pp. 7056-7069.

Zheng, X., Rafailovich, M.H., Sokolov, J., Strzhemechny, Y., Schwarz , S.A., Sauer, B., Rubinstein , M. ,1997, Long-Range Effects on Polymer Diffusion Induced by Bounding Interface , Phy. Rev. Lett ., Vol. 79, pp. 241-258.