Processing and Mechanical Behavior of Carbon Black Graded Rubber Compounds

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ABSTRACT: Functionally graded rubber compounds (FGRCs) were prepared by construction based method. The matrix used was natural rubber (NR). Amorphous carbon black (N-330) was used as grading material. The gradation of nanoparticles in a rectangular geometry comprised the variation of particle volume fraction along thickness direction. Its performance was evaluated for structural application through various mechanical and surface properties like tensile strength, modulus, tear strength, elongation at break, hardness, fracture surface by scanning electron microscopy, etc. At the same percentage of nanofiller loading, FGRCs show enhanced properties, i.e., modulus and tear strength (in some grades) compared to uniformly dispersed rubber compounds (UDRCs). Modulus of FGRCs, for a given particular stacking sequence of the layers, increases as much as by 275% compared to UDRCs. The ultimate properties like tensile strength and elongation at break made up for the modulus enhancement that decreases to as minimum as 50 and 80%, respectively. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 115: 3146–3154, 2010

Key words: functionally graded; rubber; carbon black; mechanical properties

INTRODUCTION

Functionally graded materials (FGMs) are the composites wherein the compositions of two or more components vary continuously to optimize the performance of a material for a specific application. Compositional gradient brings in the improvement of properties of the isotropic materials when compared with the same average composition of the components.[1,2] In polymer based FGMs, the cognizance of different processing methods is very limited as compared to ceramic and metal based systems.[3] In polymer composites, the gradation effect using SiC particles and glass fibers as the reinforcing materials is studied by Klingshirn et al. by centrifuging prior to polymerization.[4] He has observed that the filler concentration after centrifugation in the outer portions of the specimen is increased up to 27% for the glass fibers and up to 45% for SiC particles, for an average filler content of 20%. Variation in the molecular weight, i.e., polydispersity index alters the properties of the polymers. For multiphase polymers like multiblock copolymers, blends, etc., morphology can be altered by varying the processing conditions. Akiyama et al. have analyzed the gradation effect of crystalline phase on the semicrystalline polymers.[5]

Mixing of the filler is one of the crucial parameters generally focused as the homogeneity of mixing significantly affects the properties of the vulcanizates.[6] Even with the homogeneous mixing of the compounds, technologists have observed that the crosslink density of the vulcanizate is not uniform along the thickness due to non-uniform temperature distribution from the outer to inner portions of the mold cavity. Contrarily, Ikeda has used this conception and intentionally varied the curing agent along the thickness to study the effects on the dynamic mechanical properties of the vulcanizate.[7] Zhao et al. have cured polyurethane elastomer in graded temperature field.[8] Ahankari and Kar have introduced the gradation of crosslink density as well as the gradation of glass transition temperature and have compared these graded compounds with conventional vulcanizates using equivalent amounts of the ingredients.[9,10] There was hardly any literature available that has envisaged the effects of the heterogeneity of the filler content on the mechanical properties of the filled composite. A plenty of questions that come to authors mind, like, what will be the consequence of variation of the filler on mechanical properties of such a composite? will that variation affect the modulus of the composite as the rubber specialists really look for? if the UDRCs and FGRCs are compared for

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a given average loading of the filler, will there be a marginal or really a substantial effect on the properties? need to be answered.

The concept of non-homogeneous, spatial filler distribution in an elastomer assists the gradation of other properties like hardness, modulus, tensile strength, tear strength, hysteresis loss, etc.[11] Carbon black (CB) is a crucial ingredient for the most rubber formulations. It enhances the modulus as well as the ultimate properties of the soft elastomer. Ultimate properties let in tensile strength, tear strength, fatigue resistance, abrasion resistance, etc., and have a considerable influence on the performance of the final product.[12] Even though the reinforcement of rubber shows the pronounced increment in the tensile strength, tear strength, abrasion resistance, and modulus far beyond the values expected by Einstein-Guth and Gold theory considering the hydrodynamic effect and the occlusion of rubber,[13] it is also observed that the above mentioned properties vary to a large extent with the way fillers are dispersed within the rubber matrix.[11] The smooth variation of CB along the thickness of the vulcanizate alters the other properties accordingly.

The present work is focused on the processing and mechanical characterization of FGRCs comprised of NR as a matrix and CB in a graded form. For a given average volume fraction of the filler, these FGRCs are compared with UDRCs by physical and mechanical characterizations and an attempt is made to reach to the answers of the questions that invoked during the discussion. An attempt is made to enhance the modulus and tear strength of the FGRCs than the corresponding UDRCs at the expense of tensile strength and percentage elongation at break.

EXPERIMENTAL DETAILS

Materials, formulation, and mixing procedure

Typical composition of NR vulcanizates with varied filler content is listed in Table I. NR (RMA-4) and CB (HAF-N330) supplied by the M/S Kankani Brothers, India, were selected for the study. Polymerized TQ, zinc oxide, stearic acid, SP oil, and sulfur were supplied by M/S Pragati, India. The ingredients were mixed with NR on a two-roll mill at the temperature range of 40 to 50°C and a friction ratio 1 : 1.1 according to the ASTM D 3182-89(R04)E01.

Specimen preparation

NR with regular ingredients was mixed and different homogeneous mixes containing varied CB (0–100 phr at the interval of 10) were made. A thin uncured layer of thickness 0.2–0.3 mm was prepared by pressing it between the mylar sheets in the hydraulic press at room temperature. The inner surfaces of the mylar sheets were coated with PVA for easy removal of the layer. All these uncured thin layers taken from different mixes were stacked sequentially with increasing/decreasing amount of CB in each layer. The stack as a whole was kept in the mold maintaining 150°C. The mold was then promptly compression molded at 4 MPa for 15 min in a hydraulic press to get a cured sheet with gradation of CB along thickness direction. At such high curing temperatures and pressures, the stacking layers get interpenetrated in the adjacent layers. This ensures the spatial gradation of carbon black along the thickness of the compound. This spatial variation of carbon black can be seen as the change in the contrast of Figure 1(a,c) along the thickness direction. The samples for testing of mechanical properties at required depth in FGRCs were made by using buffing wheel. A special care was taken to avoid crack generation on the samples during buffing. In addition to this, the amount of heat generated during buffing was dissipated to the atmosphere by using a fan. The concentration (phr) of carbon black was determined by thermo-gravimetric analysis. To measure the mechanical properties of vulcanized FGRCs and UDRCs, thickness of the sheet is maintained $\sim$ 3 mm.

TABLE I
Formulation of NR Vulcanizates for UDRCs

Materials	Phr
NR (RMA-4)	100
Carbon black (N330)[a]	Variable
TQ[b]	1.5
Stearic acid	2.0
Zinc oxide	5.0
MBT[c]	0.8
SP oil	2.0
Sulfur	2.5

[a] Used at 0, 10, 20, 30, 40, 50, 60, 70, 80, and 100 phr.
[b] 1,2-Dihydro-2,2,4-trimethyl quinoline.
[c] 2-Mercaptobenzothiozole.

Measurements

Hardness

The hardness of UDRCs and FGRCs was measured according to ASTM D-2240 with a shore A durometer. Specific gravity of the composite for respective filler-loading was calculated from the standard density values of the compounds and compared with the experimental values measured by specific gravity meter.

Stress–strain properties

Tensile stress–strain measurements were used for characterizing the performance of the UDRCs and

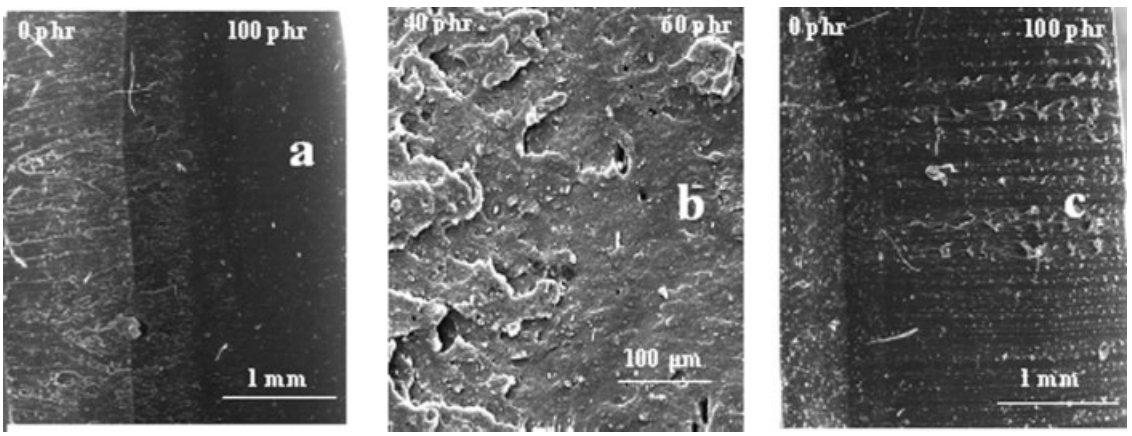

Figure 1 SEM micrographs showing (a) fractured surface of 0-20-40-60-80-100 carbon black graded-NR FGRC, (b) micrograph at the interface of 40-60 layer, and (c) side surface of specimen elongated to 100% of 0-20-40-60-80-100 carbon black graded-NR FGRC.

FGRCs. Tensile strength, strain energy at break, modulus, and elongation at break were measured on dumb-bell specimens (cut from the sheets using type-A die) and tested on a Zwick/Roell *Z010* model at a cross-head speed of 500 mm/min according to ASTM D412-83. The testing software *textXpert* was used to control the testing parameters like cross-head displacement, cross-head speed, applied load/strain, etc. This software was also used to measure the above material properties. For FGRCs, an average reading of properties of five different samples taken from different sheets is mentioned in the Table II. For UDRCs, an average reading of four samples is placed.

Tear strength

Type C tear strength was measured by applying a tearing strain to a test specimen by means of a *Zwick/Roell-Z010* Universal Testing Machine. It was operated without interruption at a constant rate of cross-head traverse until the specimen is completely torn according to ASTM D624.

RESULTS AND DISCUSSION

SEM and photographic analysis

Figure 1(a) shows the SEM fractograph of the FGRCs wherein the CB is varied from 0 to 100 phr in NR. It corresponds to 0-20-40-60-80-100 (the numbers stand for the phr values of the CB and the sequence shows the stacking order of the layers) gradation. One can see that the ligaments are stretched at 0 phr end, broken and shrank in between and the fracture looks planar towards 100 phr end. At every layer interface, the fracture plane has changed. Stretching of ligaments end at the interface of 40 and 60 layers as shown in Figure 1(b). Stacking of the layers might be leaving the small tiny air-pores trapped in the adjacent layers. Small tiny pores formed at the interface are stretched and can easily be seen in Figure 1(b). Figure 1(c) shows the SEM micrograph of the

TABLE II
Comparison of Modulus and Tensile Properties of UDRCs and FGRCs with Gradation of Carbon Black in NR Vulcanizates

Grade	Avg. phr carbon black	Tensile strength (MPa)	% Elongation @ break	Tear strength (kN/m)	Mod. @ 50% (Mpa)	Mod. @ 100% (MPa)	Mod. @ 150% (MPa)
30 phr	30	12	570	28	1.02	1.67	2.4
0-20-40-60	30	9.4	388	17	1.2	2.24	3.29
40 phr	40	13	485	65	1.23	2.12	3.14
0-20-40-60-80	40	6.5	200	18	1.9	3.34	4.61
50 phr	50	13.5	420	24	1.52	2.71	4.03
0-20-40-60-80-100	50	7.1	184	34	2.41	4.27	5.98
0-50-100-100-50-0	50	6.0	80	25	4.12	–	–
100-50-0-0-50-100	50	7.5	75	36	5.67	–	–
0-0-50-50-100-100	50	7.4	75	32	5.15	–	–
100 phr	100	9.3	133	25	4.8	7.85	–

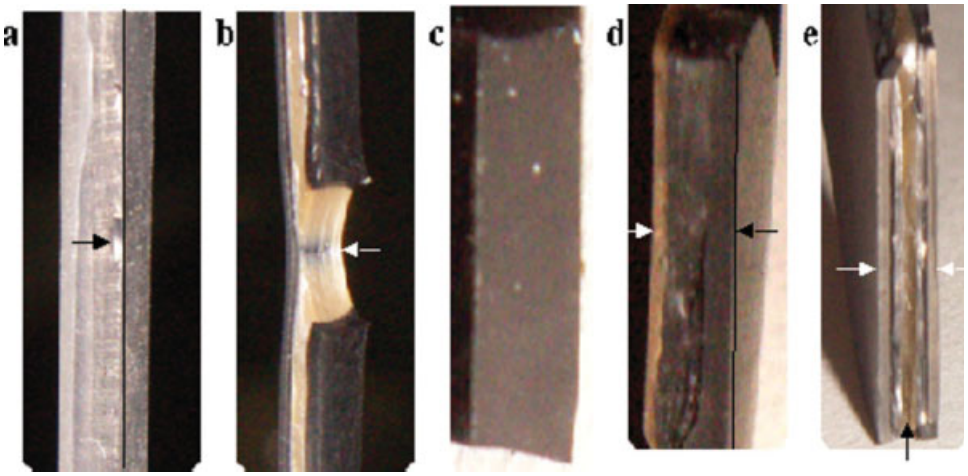

Figure 2 Photograph showing (a) the initiation of crack at 100 phr end in 0-20-40-60-80-100 FGRC during deformation, (b) fractured half portion of 100-50-0-0-50-100 FGRC, and the other half is about to (c,d,e) fractured surfaces of UDRC, 0-20-40-60-80-100 and 100-50-0-0-50-100 FGRCs correspondingly, all containing an average 50 phr carbon black. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

side surface after 100% elongation of the sample. One can observe the corrugations/serrations formed (the gap between the projected areas) enlarge at the 100 phr end and close completely towards the 0 phr side. Severity of the formed corrugations is more in the fractured sample (not shown here).

To visualize the fracture behavior, few digital snaps also are attested. Figure 2(a) shows it clearly that for 0-20-40-60-80-100 FGRCs, fracture initiates at 100 phr end (arrow marked at the interface of width and thickness zone) and not along the whole thickness at once. For 100-50-0-0-50-100 FGRCs, as shown in Figure 2(b), crack initiates at one of the ends and that side propagates through width completely, leaving the other half layers ready to sustain the loads. Figures 2(c,d,e) stand sequentially for the fracture surfaces of UDRC (smooth fracture), 0-20-40-60-80-100 FGRC (golden color with marked of white arrow stands for 0 phr carbon black loading, and black color with marked of black arrow stands for 100 phr carbon black loading) and 100-50-0-0-50-100 FGRC (golden color with marked of black arrow stands for 0 phr carbon black loading, and white arrow stands for 100 phr carbon black loading), all containing an average amount of 50 phr CB. It clearly depicts that both the FGRCs have highly rougher macroscopic fracture surfaces than UDRCs.

Effects of carbon black gradation

Effect on hardness and specific gravity

In 0-20-40-60-80-100 FGRCs, hardness and specific gravity both increase with increasing volume fraction of the CB along thickness, as the hardness and specific gravity of CB is much higher than the softer matrix. The volume fraction of CB increases from 0 to 0.33 from one surface to the other along the sheet thickness. Hardness increases from 35 to 92 shore A while the specific gravity lifts up from 0.97 to 1.24, respectively, as shown in Figure 3. The percentage increase in hardness and specific gravity is 160 and 27, respectively. For other FGRCs (prepared with different stacking sequence and with different step variation), these properties vary within the range mentioned above and can be easily altered by changing the stacking sequence depending upon the material properties required for a given particular application (Figures are not shown here).

Effect on modulus and tensile properties

Tensile stress–strain measurements were carried out to understand the reinforcement mechanism.[14] The

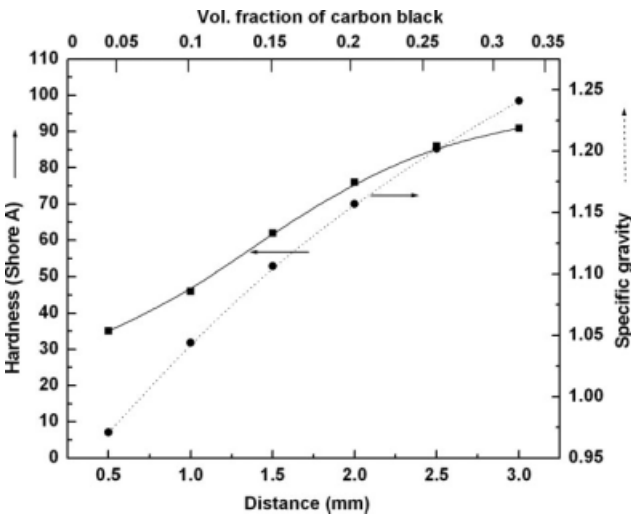

Figure 3 Hardness (■) and specific gravity profiles (●) of 0-20-40-60-80-100 FGRC.

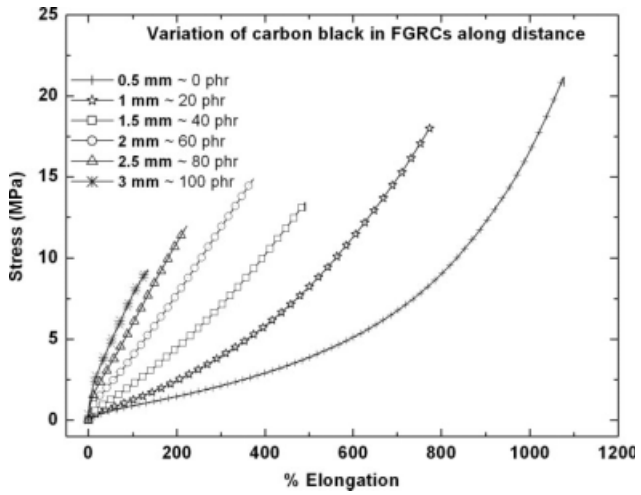

Figure 4 Variation of stress-elongation behavior of different layers of 0-20-40-60-80-100 FGRC.

ultimate properties, i.e., tensile strength, strain energy at break, elongation at break, etc., were measured to characterize the UDRCs and various FGRCs that envelope different stacking sequences of the layers. These properties vary as the composition (i.e., loading of nanofillers) and hence microstructure varies along thickness. The variation of stress-elongation percentage behavior of individual layers of the 0-20-40-60-80-100 FGRC is shown in Figure 4. The gum compound (0 phr), which is present at the periphery of low hardness zone to a distance of 0.5 mm of the aforementioned FGRCs extends by more than 1000% with a stress value of $\sim$ 20 MPa, which is much more than the filled vulcanizates. This attributes to strain induced crystallization of the NR gum compound. With increasing amount of filler content or moving towards the inner surface from the low hardness zone, the tensile strength and

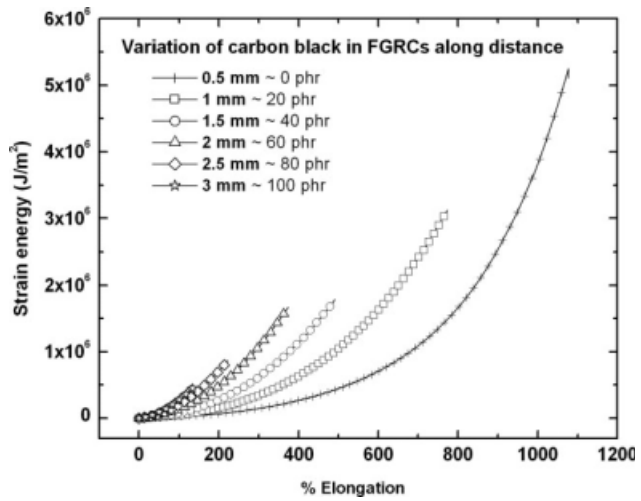

Figure 5 Variation of strain energy-elongation behavior of different layers of 0-20-40-60-80-100 FGRC.

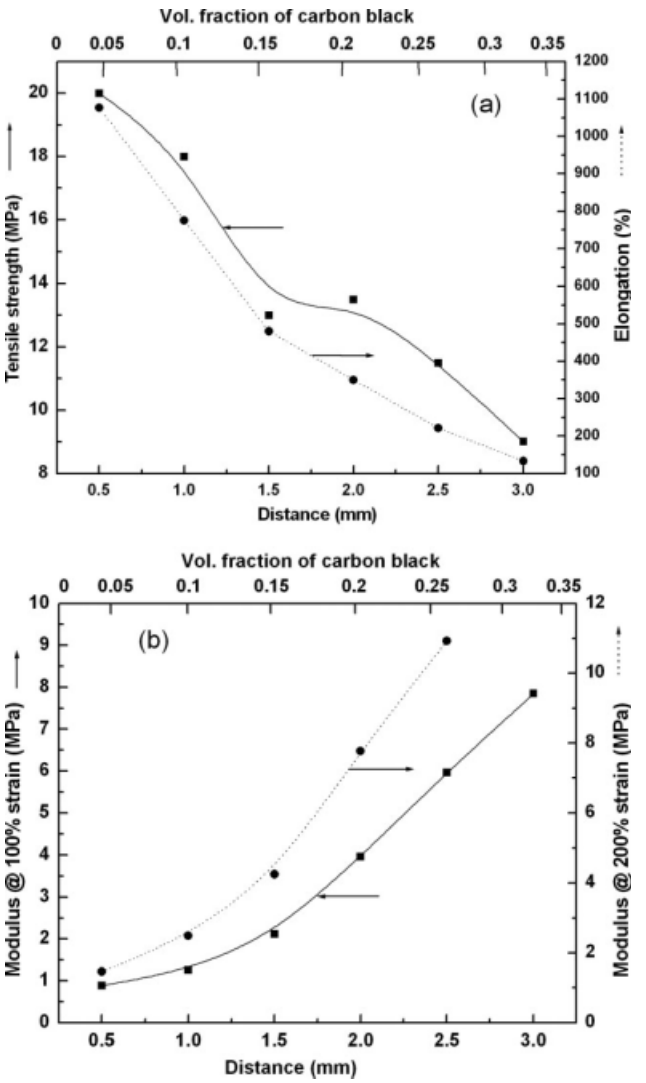

Figure 6 (a) Profile of tensile strength (■) and elongation at breaking point (●) of 0-20-40-60-80-100 FGRC. (b) Profiles of modulus at 100 and 200% strain of 0-20-40-60-80-100 FGRC.

percentage elongation at break drops down. NR with 100 phr CB loading or the surface which is present at the periphery of high hardness zone shows the least values of tensile strength and elongation at break of 9 MPa and 133%, respectively, among mentioned. The strain energy of the FGRCs also decreases with the increasing filler content or moving from low hardness zone to the high hardness zone as shown in Figure 5.

Figure 6(a) displays the variations in tensile strength and elongation at break with sheet thickness along which the volume fraction of the nanoparticles increases from one end to the other. For 0-20-40-60-80-100 FGRC, tensile strength drops down linearly from 20 to 9 MPa along the span of 3.5 mm while elongation at break also descends from 1100 to 150% with increasing concentrations of the filler. Tensile strength and elongation at break decrease by

nearly 55 and 87%, respectively. With increasing filler content, slope of the curve, i.e., modulus also increases (see Fig. 4). Filler aggregates have a tendency to form network structures. Elastic modulus of the filled-compound increases significantly due to the trapped rubber in the network as the effective volume of the nanofiller increases by the presence of occluded rubber. Amount of trapped rubber, and hence, modulus increases with loading of the filler. Modulus of 100 phr vulcanizate at 100% elongation is nine folds higher than the modulus of gum compound. The values are 7.85 and 0.88 MPa, respectively. The change in the modulus at 100% elongation is also plotted with the variation in the volume fraction of CB along the distance and shown in Figure 6(b). The increment in the modulus is steep in the first half (i.e., upto volume fraction of 0.15) along the thickness and then increases rapidly in the next half. At lower concentrations, nanofillers can not form the three-dimensional network structures as the inter-aggregate distance is higher. This might be the cause of small increment in the modulus in the first half along thickness. Modulus at 100% elongation increases by 600% in the span of 3.5 mm whereas modulus at 200% elongation lifts up nearly by 800% in the same span [Fig. 6(b)].

A comparison of properties between various UDRCs and FGRCs with gradation of CB in NR vulcanizates is shown in Table II. In this context, modulii at 50% (amount of stress required for 50% deformation), 100% (similarly the stress required for 100% deformation), and 150% elongations are measured for these UDRCs and FGRCs. For a given average amount of CB, FGRCs exhibit higher modulus at a given percentage elongation corresponding to their UDRC counterparts. For an average of 50 phr CB, the modulus of the FGRCs at 50% elongation is enhanced by maximum up to 275% than the corresponding UDRCs for a given particular sequencing of the layers. Modulii of 50 phr UDRC and for a given sequence of 100-50-0-0-50-100 FGRC at 50% elongation are 1.52 and 5.67 MPa, respectively. Figure 7(a) demonstrates a bar plot comparing the modulus values of UDRCs and various FGRCs using different stacking sequences for an average of 50 phr CB. 0-20-40-60-80-100 FGRCs show the modulus value to be 2.4 MPa at 50% elongation, which is 58% more than its UDRC counterpart. Enhancement of the modulus in all FGRCs is credited to the spatial variation of filler in the matrix. Table II shows that for any possible combination of stacking sequences that govern the spatial or stepwise variation of the filler demonstrates the enhancement in the modulus. For an average amount of 30 as well as 40 phr CB, the graded specimens also show the enhancement in the modulus. The respective percentage enhancement of the modulus in FGRCs for 30, 40, and 50 phr nanofiller is highlighted in Figure 7(b) in the form of a bar plot.

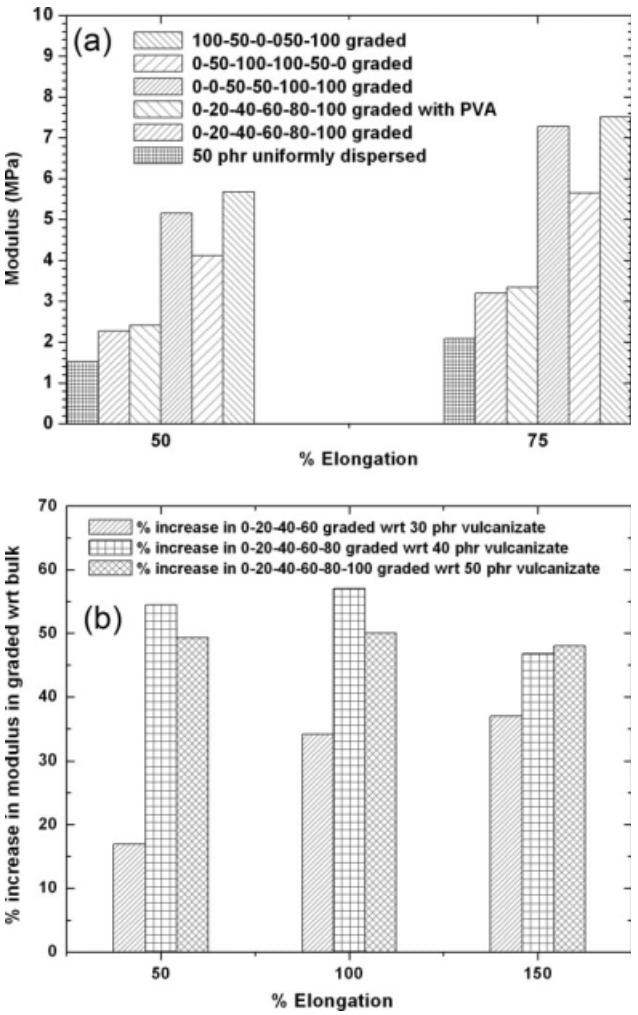

Figure 7 (a) Comparison of modulus values of UDRCs and various FGRCs employing an average amount of 50 phr carbon black. (b) Percentage increase in the modulus of FGRCs w.r.t. to the corresponding UDRCs.

Tensile stress-elongation curves corresponding to UDRCs and various FGRCs, using an average of 50 vphr CB, are also plotted and shown in Figure 8(a). Figure 8(b) is the amplified part of the same curve. For an average 50 phr CB, the values of tensile strength and elongation at break for UDRCs are 13.5 MPa and 420% while for a given sequence of 0-20-40-60-80-100 FGRCs, the values are $\sim$ 7 MPa and 185%, respectively. With gradation of nanofiller, elongation at break has decreased by 56% and so the tensile strength has also dropped down to 50% of the respective UDRCs. With increasing roughness of gradation, i.e., for the grades like 100-50-0-0-50-100, 0-0-50-50-100-100, and 0-50-100-100-50-0 FGRCs, elongation at break decreases further to 75%, maintaining nearly the same tensile strength. The values of tensile strength and elongation at break for 100-

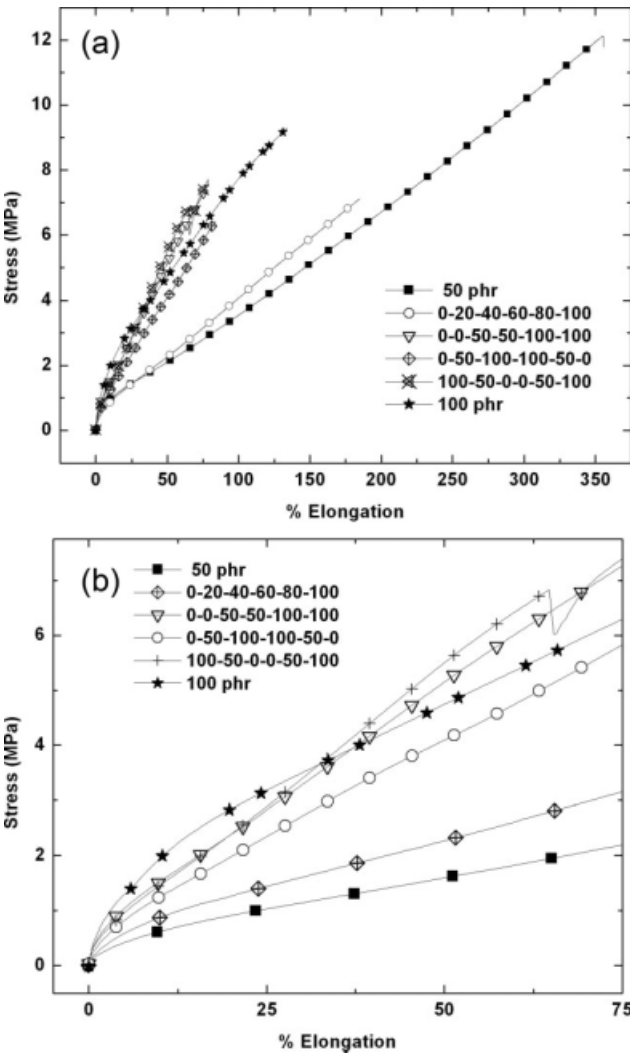

Figure 8 (a) Comparison of stress-elongation curves of UDRCs and various FGRCs employing an average amount of 50 phr carbon black. (b) Magnified plot of Figure 8(a).

50-0-0-50-100 FGRCs are 7.5 MPa and 75%, respectively. For structural applications, the failure parameters are least interested by the designers whereas the enhancement of modulus in elastomers is highly recommended. Figure 8 reveals that the roughness in the variation of the CB in the adjacent layers of the FGRCs increases the modulus. Depending on the strength, modulus, and surface properties required, design can be optimized and layers can be stacked accordingly. The UDRC containing 100 phr CB has higher initial modulus than all the FGRCs [see Fig. 8(b)] that incorporate average 50 phr nanofiller. During the initial deformation of this UDRC, network structure has not broken down and the formation of the filler network has significantly increased the effective volume fraction of the filler due to trapped rubber in the agglomerates causing higher initial modulus than FGRCs. At higher elongations, weak agglomerates can easily get broken by the applied forces. Hence, modulus decreases at higher elongation. The stress required to deform the FGRCs for a given percentage elongation is obviously much higher than its UDRCs counterpart due to spatial variation of the CB along thickness. In 0-20-40-60-80-100 FGRC, all the layers strain equally when stressed, but the amounts of loads shared by them are not equal. For a given percentage elongation, 100 phr layer takes the maximum load while 0 phr layer takes the least as the modulus increases with increasing nanofiller content. This unbalanced load bearing of the layers allow the crack to initiate in 100 phr layer [Fig. 2(a)]. This corner-initiated crack propagates along the width and once the cut starts to grow, the stress required for further growth of the crack decreases.[15] When this stress becomes equal to the stress required to initiate the crack in the adjacent layer containing 80 phr CB, the crack opens up in the direction of thickness too and progresses towards 0 phr layer as well. Non-uniform distribution of the loads taken up by the contiguous layers containing spatial variation of the CB is ascribable to the reduction in elongation and hence tensile strength of FGRCs. Decrease in the elongation may also be attributed to the increase in the modulus of the FGRCs. The UDRC using 100 phr CB elongates to much lesser extent as the probability of formation of bigger sized flaw increases with increasing concentration of CB in the matrix.[16] With increasing flaw size, elongation at break and hence tensile strength decreases. So, accordingly, crack initiates in 100 phr layer of the 0-20-40-60-80-100 FGRCs making it to fail at 185% elongation only. UDRC containing 50 phr CB elongates to 420%. The layering sequence containing a difference of 50 phr CB loading in a single step like 0-50-100-100-50-0 or 100-50-0-0-50-100 or 0-0-50-50-100-100 elongates to ~75% only. Higher is the non-uniformity in the filler distribution, lesser is the observed elongation of the FGRCs. For an average 50 phr CB, the FGRCs containing any layer sequencing show nearly the same tensile strength value (see Table II) but the modulus of every combination is different. With higher difference in the filler content in the adjacent layers, modulus of the FGRCs will be higher. When the difference between filler content of the adjacent layers increases, unequality of load contribution of the layers increases. With increasing this unequality, modulus increases while elongation at break comes down. Any of the FGRCs containing 0 and 50 phr CB layers show higher modulus than 0-20-40-60-80-100 FGRCs and much higher modulus than UDRCs for a given percentage elongation.

Effect on tear strength

The UDRCs and FGRCs are characterized by another mechanical property, i.e., tear strength (the amount

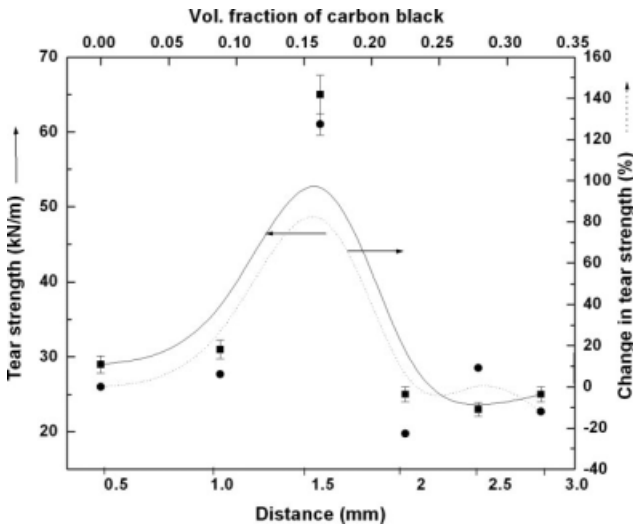

Figure 9 Variation of tear strength and its percentage change for FGRCs.

of stress required to break the specimen having a pre embedded crack as per ASTM standard) and is shown in Figure 9. The variation of tear strength along the thickness direction is not linear as of tensile strength, elongation, and modulii curves. Higher tear energy is needed in filled UDRCs than unfilled NR vulcanizates as CB assists in tear deviation leading to stick-slip or knotty tear in the compound. Tear deviation makes the fracture surface larger that correspondingly increases tearing energy. CB particles create barriers by forming the strands of aggregates along the direction of stretching and change the path of tear. In these aforementioned modes, path of tear deviates and again force accumulates to initiate the new tear path. And this process requires more tearing energy. Thomas deduced that the force required to initiate tearing increases with the unstrained diameter of the tip of the tear.[17] Tear strength of the few FGRCs with an average 50 phr CB are compared with the materials wherein the nanofillers are dispersed uniformly in the matrix, i.e., UDRCs and are shown in Table II. The tear strength of few graded compounds shows higher values than their corresponding UDRCs. Table II displays various forms of FGRCs showing the tear strength values of nearly 32–36 kN/m that are comparatively higher than the corresponding UDRCs having 24 kN/m. With the inhomogeneous distribution of the nanofiller along thickness, tear force gets unbalanced along the crosssection. This unbalanced force initiates the crack in the higher CB content layer. In 0-20-40-60-80-100 FGRCs, crack opens up at one end (the end that contains 100 phr CB) along thickness. Force needs to build up for every opening of the crack. And as the layers are stacked sequentially with gradual decrease in the filler content, at every layer crack deviates and highly macroscopically irregular torn surface can be seen. As tear path takes different ways at every layer, larger fracture surface area appears that correspondingly increases the tear energy in FGRCs. In 0-0-50-50-100-100 FGRC, the gradation is not smooth and hence the way the tearing crack propagates is slightly different. It propagates completely in 100 phr layer and then it initiates in 50 phr layers. This FGRC shows more irregular fracture surface and hence higher tearing energy than even 0-20-40-60-80-100 FGRC. For a given FGRC, crack initiates at 100 phr end along thickness and that propagates along the width either partially or fully, depending upon the smoothness of gradation, and again opens up in the adjacent layer along thickness and likewise. In 0-50-100-100-50-0 gradation, crack opens up in the middle along thickness and propagates along width as well as towards 0 phr layers along thickness. The UDRC using 40 phr nanofiller has much higher tear strength (naughty tear) compared with its graded counterpart, i.e., 0-20-40-60-80 FGRC. The respective values for the UDRCs and FGRCs are 65 and 18 kN/m. The reason for the exorbitant increase in the tear strength of this UDRC is adverted by Gluclich and Landel.[18] They reported that the unstrained tip diameter increases 10 times with the addition of N330 CB (for $v_f = 0.25$) and the corresponding average tearing force increases 30 times for the knotty tear.

CONCLUSION

FGRCs were made using a simple construction based layering method using NR as a matrix and CB in a graded form. These prepared FGRCs were compared with UDRCs for a given average amount of CB. Following conclusions were made based on the above experimental investigation.

- Modulus of FGRCs for any given possible combinations of sequential gradation has increased (max. up to 275%) compared to UDRCs, both using the same average volume fraction of the filler. With increasing roughness in gradation, modulus of the FGRC increases whereas percentage elongation at break decreases. Tensile strength has hardly affected the roughness of gradation.
- Tensile strength and percentage elongation at break of FGRCs, the failure parameters, have decreased as low as by $\sim$ 50% and by 81% of the UDRCs, respectively. Tear strength of few FGRCs, for a given 50 phr CB, has increased max. up to 50%.
- For a given 0-20-40-60-80-100 FGRC, hardness varied from 35 to 92 shore A whereas specific gravity lifted up from 0.97 to 1.24.

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