Mechanical Properties of Functionally Graded Carbon Black–Styrene Butadiene Rubber Composites: Effect of Modifying Gradation and Average Filler Loading

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ABSTRACT: The mechanical properties of functionally graded polymeric composites (FGPCs) with varying carbon black loading and the effect of stacking sequence in styrene butadiene rubber (SBR) matrix were studied. For a given average amount of nanofiller, the modulus of FGPCs for any given stacking sequence of layers is higher when compared with its uniformly dispersed polymeric composites (UDPCs) counterpart. Tensile strength, elongation at break, and tear strength either increase or decrease depending on the stacking sequence and average loading of the filler in FGPCs. In addition, the smoother gradation (i.e., lesser difference in the amounts of CB content in adjacent layers) and a wide gap of difference in CB content in a stack has a profound effect on the modulus and tensile strength of FGPCs. Dynamic mechanical analysis shows lesser damping in FGPCs than UDPCs. © 2012 Wiley Periodicals, Inc. J Appl Polym Sci 000: 000–000, 2012

Key words: functionally graded material; composites; mechanical properties; dynamic mechanical analysis

INTRODUCTION

Functionally graded materials (FGMs) are a category of relatively recent and promising materials that have emerged from the need to optimize the performance of a component in the target applications. In a homogeneous material the properties are constant, whereas in an FGM, the material composition, microstructure and hence properties vary continuously with position, usually in one coordinate direction.[1–3] The applications of metal/ceramic FGMs are exploited in air-craft and energy sectors.[4–6] But in polymer-based FGMs, the lack of awareness of different processing techniques hinders the exploration of its applicability in diversified areas. Still people have tried to make the gradation of properties in polymers like hardness, wear resistance, impact resistance, toughness, etc. and noticed majority of its applications in biomedical and optical fields.[1–3] Klingshirn et al. have studied the effect of gradation of SiC particles and glass fibers on the polymeric composites employing centrifuging technique before polymerization.[7] Akiyama et al. have analyzed the effect of gradation of crystalline phase on the semi-crystalline polymers.[8] Ikeda has prepared the functionally graded composites by layering method using styrene butadiene rubber (SBR) matrix with gradation of network chain density along thickness direction and compared them with homogeneous compounding sheets.[9,10] Free vibration analysis has been done on fly ash reinforced functionally graded rubber composite sandwiches to investigate damping ratio and natural frequency.[11]

There are various applications where controversial properties are required which suggests a need of a multifunctional material, e.g., slippers, rubber pads, etc. It was earlier elucidated that for a given volume fraction of nanofiller in polyisoprene (natural rubber) matrix, functionally graded polymeric composites (FGPCs) show much enhanced modulus than uniformly dispersed polymeric composites (UDPCs).[12,13] The major receding factor for the development of these FGPCs is the reduction in failure properties like tensile strength, elongation at break, tear strength, etc. when compared with its UDPC counterparts. Here, keeping in mind the importance of mechanical properties of polymer composites in polymer industry, an attempt is made to prepare FGPCs using synthetic polymer i.e., styrene butadiene rubber (SBR) as matrix and carbon black (CB) as grading material by construction-based layering method.[11] The article aims at the investigation of an impact of different stacking sequences on its

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TABLE I
The CB-Filled SBR Vulcanizate Formulation

Materials	phr
SBR (1502)	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, 15, 20, 30, 40… upto 100 phr.
[b] 1,2-Dihydro-2,2,4-trimethyl quinoline.
[c] 2-Mercaptobenzothiozole.

mechanical properties by comparing them with corresponding UDPCs. It is necessary to study the effects of average filler loading on the properties of FGPCs. With the use of synthetic polymers, modulus enhancement and the consequence on other properties is to be studied. The effect of smoother gradation by maximizing the number of layers in a stack as well as the change in the stacking sequence is to be investigated to enhance the failure properties of these FGPCs. It is worth to mention that there is hardly any literature available that will throw some light on gradation of CB in rubber-based composites.

EXPERIMENTAL

Materials, formulation, and mixing procedure

A typical composition of the UDPCs prepared, employing SBR matrix with uniform dispersion of carbon nanofillers throughout, is listed in Table I. SBR-1502 and CB (HAF-N330) were supplied by the M/S Kankani Brothers, India. Zinc oxide, polymerized TQ, stearic acid, SP oil, and sulfur were supplied by M/S Pragati Inst., India. These ingredients were homogeneously mixed with SBR on a two roll mixing mill at a temperature of 25–50°C and friction ratio of 1 : 1.1 according to the ASTM D 3182-89(R01)E01.[14]

FGPC specimen preparation

All the regular ingredients were homogeneously mixed in SBR and different mixes containing varied CB (from 0 to 100 phr) were made. A thin uncured layer from each mix was taken out from a two-roll mixing machine. All these uncured thin layers taken from different mixes were stacked sequentially with increasing/decreasing amount of CB in each layer as shown in Figure 1. The stack as a whole was kept in the mold. It was compression molded at 150°C, 4 MPa pressure, for 15 min in a hydraulic press to get a cured sheet with gradation of CB along thickness direction. The order of stacking of thin layers was also made in "ascending–descending–ascending" and "descending-ascending-descending" fashion.

Measurements

High resolution transmission electron microscopy (HRTEM)

The morphology and the structure of carbon nanoparticles were investigated using HRTEM on Tecnai G2 model operated at 200 kV.

Scanning electron microscopy (SEM)

SEM studies were carried out with FEI 2000 scanning electron microscope to study the variation of the filler in the matrix as well as to see the effects of loading–unloading on the stacking layers in FGPCs. Rubber samples were sputter-coated with Au-Pd with coating thickness ~ 1–2 nm. SEM micrographs were taken under high vacuum mode at 20 kV.

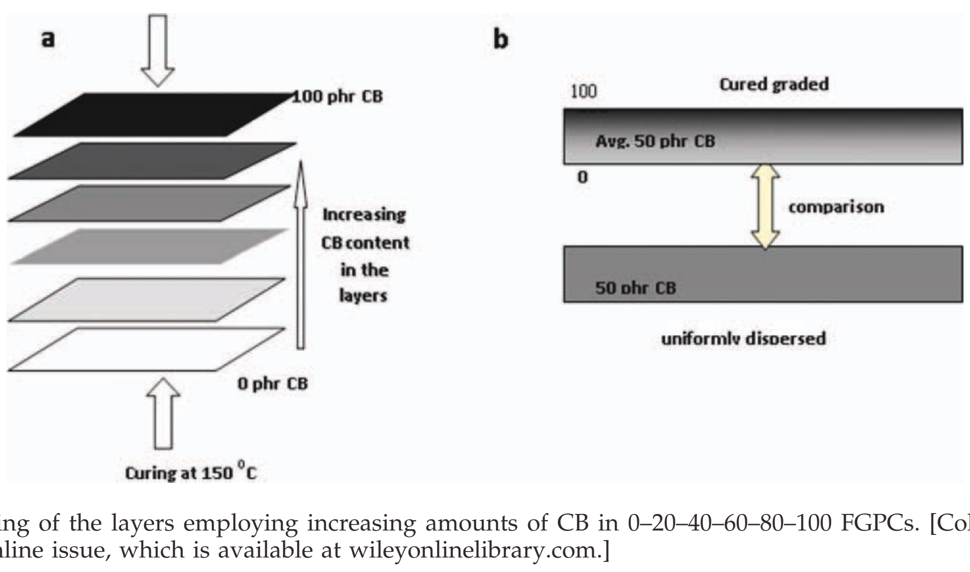

Figure 1 Stacking of the layers employing increasing amounts of CB in 0–20–40–60–80–100 FGPCs. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

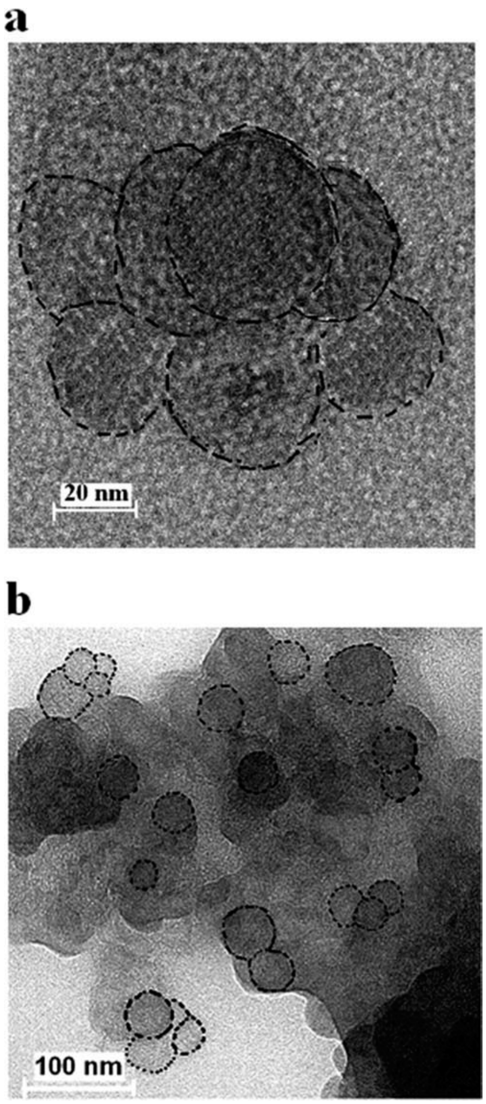

Figure 2 HRTEM images of carbon nanoparticles.

Hardness

The hardness of the UDPCs was measured according to ASTM D2240-04E01 with a shore durometer.[15] Specific gravity of the composite for respective filler-loading was calculated from the standard density values of the compounds.

Stress–strain properties

Tensile stress–strain measurements were employed for characterizing the performance of the UDPCs and FGPCs. Tensile strength, 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 crosshead speed of 500 mm min^{-1} according to ASTM D412 98(AR02)E01.[16] Tear strength was measured in Zwick/Roell Z010 UTM using unnotched 90° angled tear test specimen (Die C) as per ASTM D 624(00) E01.[17] The samples were punched out from molded sheets. The speed of crosshead was adjusted to 50 mm min^{-1}. For both FGPCs and UDPCs, an average reading of five different samples was taken from different sheets.

Dynamic mechanical properties

Dynamic mechanical properties, i.e., storage modulus, loss modulus etc., and loss tangent (a measure of hysteresis loss) of UDPCs and FGPCs were measured under a multiwave dynamic bending mode at the frequency of 100 Hz on Pyris Diamond DMA, Perkin Elmer Instruments, USA. Rectangular specimens 35 × 15 × 3 mm^3 ($l \times w \times t$) were subjected to sinusoidal loading and were heated from 30 to 120°C at a heating rate of 10°C min^{-1}.

RESULTS AND DISCUSSION

HRTEM/SEM analysis

The HRTEM images are shown in Figure 2. It can be seen that the nanofiller used shows a spherical morphology with size ranging from 20 to 30 nm.

Figure 3(a) shows SEM fractograph of FGPC wherein CB is varied from 0 to 100 phr in SBR. It corresponds to 0–20–40–60–80–100 (the numbers show phr values of CB and the sequence shows a stacking order of layers) stacking. The spatial variation in CB shows contrast at every layer-interface. The stack employing 80 and 100 phr layers are hardly distinguishable due to exorbitant amount of filler. At every layer interface, the fracture plane has changed. Figure 3(b) shows a SEM micrograph of 0–20–40–60–80–100 FGPC surface after elongating–relieving the sample to 100%. One can observe the serrations formed (gap between the projected areas) enlarge at the 100 phr end and close completely towards the 0 phr side. These serrations represent the roughness in the die-cut section. The surface roughness of the cut section also increases along thickness with increasing CB due to agglomerate formation. Severity of the formed serrations is more in the fractured sample (not shown here). Figure 3(c) corresponding to 70–45–30–15–15–30–45–70 FGPC shows the higher intensity of serrations at the outer ends (70 phr ends) of the sheet as this part is highly stressed. Higher number of stackings in this FGPC can be vaguely seen in the micrograph.

Effects of carbon black gradation

Three different stacking sequences were formed in preparing FGPCs to compare them with UDPCs. These are (a) "descending–ascending" stacking of the layers, (i.e., layers employing increasing CB were

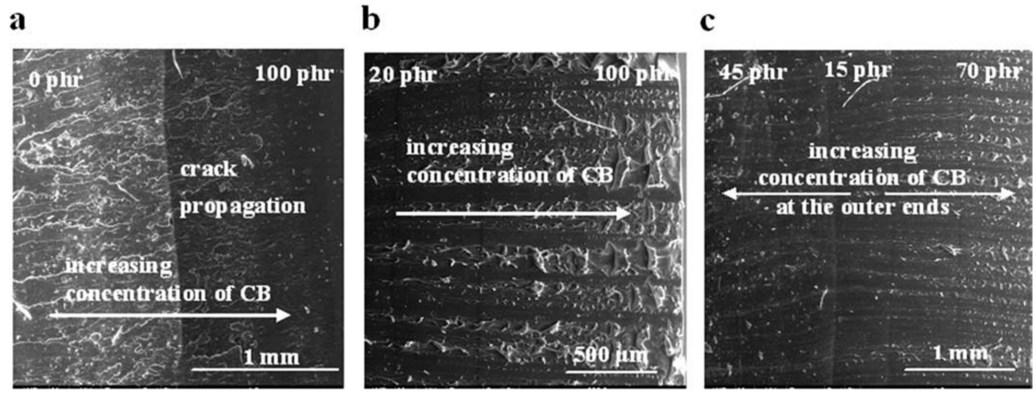

Figure 3 SEM micrographs showing (a) Fractured surface of 0–20–40–60–80–100 CB graded-SBR FGPC, (b) side surface of specimen elongated to 100% of 0–20–40–60–80–100 CB graded-SBR FGPC, and (c) a graded 70–45–30–15–15–30–45–70 stack showing thin layers.

stacked over each other e.g., 0–5–10–15–20 with average 10 phr CB, as representative), (b) "ascending–descending–ascending" stacking, (e.g., 80–60–40–20–0–0–20–40–60–80 stacking employing an average amount of 40 phr CB, as representative), and (c) "descending–ascending–descending" stacking, (e.g., 0–20–40–60–80–100–100–80–60–40–20–0 stacking employing 50 phr CB, as representative).

Effect on hardness and specific gravity

With increasing concentration of harder nanofiller in the softer SBR matrix as in case of "descending–ascending" stacking, hardness increases. For an average amount of filler, FGPCs display higher or lower hardness depending on the stacking sequence of the layers in FGPCs. For e.g., 100–80–60–40–20–0–0–20–40–60–80–100 FGPC shows higher hardness at the outer surface than 50 phr CB filled UDPC as shown in Figure 4. The FGPCs like 0–20–40–60–80 with "0 layer up" will show lower hardness. The specific gravity of UDPCs and all other FGPCs is the same as the overall volume fraction of nanofiller in the matrix is constant.

Effect of gradation/modifying gradation on modulus and tensile properties

With a given amount of nanofiller, FGPCs, for any given stacking sequence of the layers, display higher modulus than UDPCs. For an average 40 phr CB, 0–20–40–60–80 FGPCs show 43% enhancement in the modulus when compared with UDPCs as shown in Table II. Concurrently, this FGPC shows drop in the ultimate properties. Tensile strength and tear strength decreased by 40 and 26%, respectively. The decline in the tensile and tear strengths is probably ascribable to ineffective stress transfer at the interface of two layers with different carbon black loading. When stressed, crack initiates earlier at "0 phr layer" as this layer lacks strength and has no quality of strain induced crystallization as NR. The crack propagates abruptly along the thickness causing the total failure of the FGPC. The outer "0 phr layer" deteriorates the ultimate properties and so 20–30–40–50–60 and 20–20–30–30–40–50–60–70 sequences also tried. In these stackings, the ultimate properties show some improvement but this amendment in ultimate properties is compensated by reduction in modulus of FGPCs. Here 20–20–30–30–40–50–60–70 shows 20% enhancement in modulus while 0–20–40–60–80 demonstrates 43% increment. Wider span of CB variation (i.e., 80–0 = 80 phr) in 0–20–40–60–80 can be the cause of modulus enhancement compared to 20–20–30–30–40–50–60–70 grade (50 phr variation). The FGPCs employing an average 50 phr CB

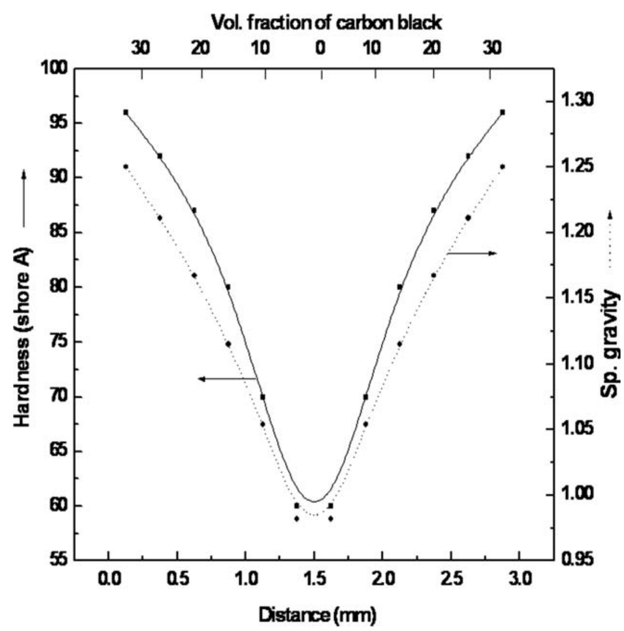

Figure 4 Change in hardness and specific gravity of 100–80–60–40–20–0–0–20–40–60–80–100 FGPC with changing volume fraction of nanofiller along thickness.

TABLE II
Comparison of Mechanical Properties of UDPCS and FGPCS ($T \approx 2.5$ mm) Employing Average 40 and 50 phr CB

Grade	Tensile strength (MPa)	Tear strength (kN m^{-1})	% Elongation at break	Modulus at 100% (MPa)	Modulus at 200% (MPa)
40 phr UDPC	*24.4*	*27*	*820*	*2.1*	*4.00*
0-20-40-60-80	14.5	20	450	2.9	5.7
20-30-40-50-60	20	22.4	675	2.2	4.3
20-20-30-30-40-50-60-70	16	24	510	2.4	4.8
50-40-20-40-50	22.2	26.7	750	2.2	4.2
60-40-0-40-60	17.6	24.8	620	2.4	4.61
70-45-30-15-15-30-45-70	12.2	20	400	2.9	5.8
80-60-40-20-0-0-20-40-60-80	13.4	19.8	400	3.2	6.2
100-67-33-0-0-0-0-33-66-100	8.8	17.8	240	3.6	7.3
50 phr UDPC	*25*	30.4	*770*	*2.4*	*4.4*
0-0-20-20-40-40-60-60-80-80-100-100	15.3	22.7	400	4.0	8.2
100-80-60-40-20-0-0-20-40-60-80-100	12.1	23.9	260	4.7	9.2
0-20-40-60-80-100-100-80-60-40-20-0	7.7	19.7	190	3.8	–

The values listed in the table show the deviation ±5% for UDPCS and ±12% for FGPCS. Modulus values of both are consistent within ±5%.

also demonstrate the similar kinds of results (see Table II). For an average 50 phr CB, modulus of 100–80–60–40–20–0–0–20–40–60–80–100 FGPC is 96% more than corresponding UDPCs. The key factors of the above experiments are—(a) The ultimate properties of FGPCs that employ "0 phr layer" at the outer ends show the least values among all the FGPC grades compared. (b) Maximizing the number of layers in a given FGPC sheet (i.e., smoother gradation of CB) entails enhancement of the ultimate properties without deterioration of modulus enhancement.

A limit for maximizing the number of stacking layers in a given FGPC sheet creates a hurdle in further enhancement of the modulus as well as drawing the ultimate properties closer to the corresponding UDPCs. Thus, "ascending–descending–ascending" and "descending–ascending–descending' stackings are preferred to check and compare mechanical properties of these FGPCs with the above FGPC grade as well as with UDPCs. All the grades of FGPCs show increased values of modulus while the tensile strength and other ultimate properties decrease. "Descending–ascending–descending" stacking of the FGPCs exhibits much lower values of ultimate properties like tensile strength, elongation at break, and tear strength compared with other two stackings as shown in Tables II and III. Hence that stacking is marginalized and other two were compared with the UDPCs. The outer "0 phr layer" at both the ends can be the cause of lowest ultimate properties amongst all the grades compared. Because of very less strength and modulus of this "0 phr layer" (i.e., neat SBR), crack can easily initiate at the surface, leading to the failure of the whole graded system. Here 50–40–20–40–50, 60–40–0–40–60 FGPC stackings are also tried. The 60–40–0–40–60 showed 18% increment in the modulus value while the ultimate properties started decreasing. With increasing CB content at the outer ends, FGPCs showed enhancement in modulus in compensation with their ultimate properties. Now 100–67–33–0–0–0–0–33–67–100 shows 82% increment in the modulus at 100% elongation while tensile strength and tear strength values decline by 64 and 34%, respectively when compared to their UDPC counterparts. For "ascending–descending–ascending" stacking, higher the departure of CB content from the average value, more will be the enhancement in the modulus of FGPCs than UDPCs and concurrently lower the ultimate properties of the given FGPCs will be.

Effect of the average filler loading

Effect of volume fraction of the nanofiller on mechanical properties of FGPCs is also investigated. With 40 and 50 phr average CB, FGPCs show much higher modulus that is compensated by the reduction in the ultimate properties for any given stacking sequence. With low CB content, FGPCs show slightly differing results from UDPCs. FGPCs employing average low filler content show enhanced tensile as well as tear strength along with modulus. In UDPCs, modulus as well as tensile strength increase with increasing CB content till the phr levels of this nanofiller reach to 50. With further increase in phr levels of CB, modulus increases but the ultimate properties go down.[18–20] In FGPCs, the grounds for higher values of ultimate properties concurrent with modulus enhancement can be—(a) Smoother gradation (i.e., the difference in CB content in the adjacent layers is less as the average amount itself is less) and (b) All the stacking layers forming FGPCs employ CB $\leq$ 50 phr.

Table III shows a comparative study of mechanical properties of UDPCs and FGPCs with average values of 10, 15, 20, and 30 phr CB. FGPCs with

TABLE III
Comparison of Mechanical Properties of UDPCs and FGPCs with Different Stacking Sequences ($t \approx 2.5$ mm) Employing Average 10, 15, 20, and 30 phr CB

Grade	Tensile strength (MPa)	Tear strength (kN m^{-1})	% Elongation at break	Modulus at 100% (MPa)	Modulus at 200% (MPa)
10 phr UDPC	5.1	18	520	1.1	1.8
0-5-10-15-20	5.75	18	525	1.3	2.2
15-10-0-10-15	6.9	17.4	580	1.4	2.4
0-15-20-15-0	4.7	17.6	425	1.3	2.2
15 phr UDPC	7.3	20	535	1.2	2.0
5-5-15-15-25-25	7.0	17	510	1.6	2.6
25-15-5-5-15-25	10.4	20	750	1.5	2.5
5-15-25-25-15-5	7.2	19	600	1.4	2.2
20 phr UDPC	13.1	22	700	1.4	2.4
0-10-20-30-40	10	25	600	1.8	3.3
30-20-0-20-30	10.6	26	555	1.7	3.2
30-20-10-10-20-30	13	25.6	750	1.5	2.7
0-30-40-30-0	8.5	26	515	1.5	2.8
30 phr UDPC	22	24.5	800	1.7	3.2
10-20-30-40-50	14	25.1	625	2.0	4.2
50-30-10-10-30-50	16	23.1	500	2.3	4.5
10-30-50-50-30-10	12.5	21.9	550	1.9	3.8

The values listed in the table show the deviation ±5% for UDPCs and ±10% for FGPCs. modulus values of both are consistent within ±5%.

"ascending–descending–ascending" stacking display improved results than other two grades. For average 10 phr CB, tensile strength and modulus of 15–10–0–10–15 FGPCs increased by ~ 35% while the tear strength is of both FGPCs and UDPCs are comparable. For an average 15 phr CB, tensile strength of 25–10–5–5–10–15 FGPC enhanced even more to ~ 42%, probably due to absence of "0 layer" present in 15–10–0–10–15, whereas the modulus increased by 25% only compared to 15 phr UDPC. For average 20 and 30 phr CB, most of the FGPCs show reduced values of tensile strength compared to their corresponding UDPC counterparts either due to enlarged difference of CB content between the adjacent layers or overall larger difference of amounts of CBs between the end layers. Comparatively, for average 20 and 30 phr CB, "ascending–descending–ascending" stacking outperforms in overall way. For 20 phr average, 30–20–10–10–20–30 stacking shows increased values of modulus and tear strength without affecting tensile strength.

Tensile stress–elongation curves corresponding to UDPCs and various FGPCs, employing an average 40 phr CB, are plotted in Figure 5. The values of tensile strength and elongation at break for UDPCs are 24 MPa and 820% while for a given sequence of 0–20–40–60–80 FGPCs, the values are ~ 14.5 MPa and 450%, respectively. With gradation of nanofiller, elongation at break has decreased by 55% and so the tensile strength also lowered to 60% of the respective UDPCs. With increasing roughness of gradation i.e., for the grade like 100–66–33–0–0–0–0–33–66–100 FGPC (change in phr values of CB in adjacent layers), elongation at break decreases further to 240% while tensile strength lowers to 8 MPa. In structural applications, the enhancement of modulus in elastomers is highly recommended. Figure 5 reveals that the roughness in the variation of CB in adjacent layers of the FGPCs increases the modulus. The number of layers and their stacking sequence can be optimized depending on the strength, modulus and surface properties required, if any.

The spatial variation of CB enhances the stress required to deform the FGPCs for a given elongation. In 0–20–40–60–80 FGPC, all the layers strain

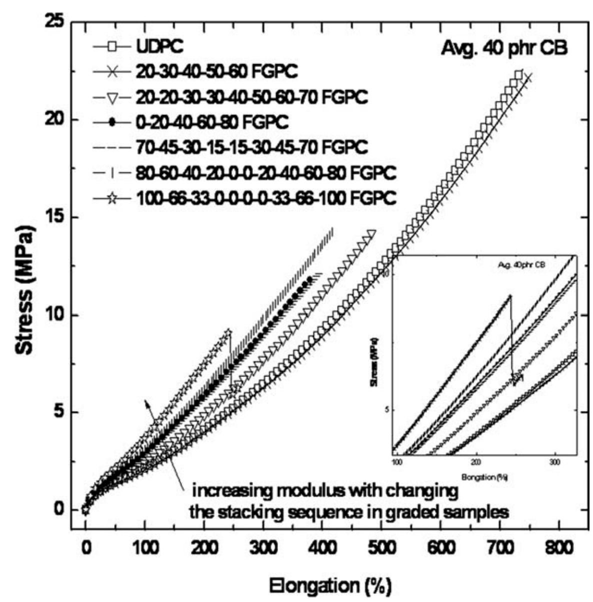

Figure 5 Comparison of stress–elongation curves of UDPCs and various FGPCs employing an average amount of 40 phr CB.

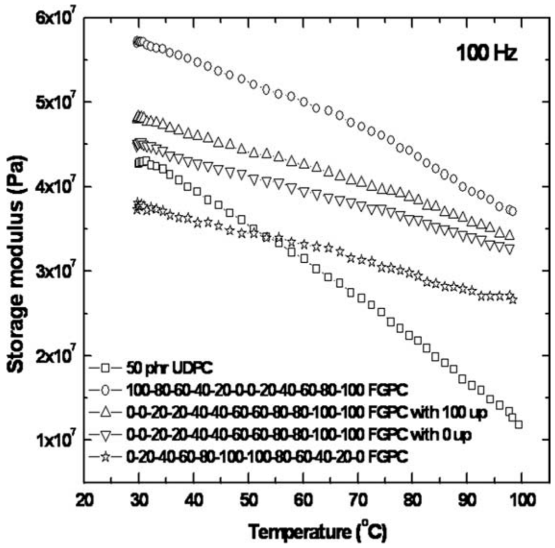

Figure 6 Comparison of storage modulus of 50 phr UDPC and different FGPC sheets employing the same average amount of filler content.

equally when stressed, but the amounts of loads shared by them are not equal. For a given elongation, 80 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 allows the crack to initiate in 80 phr layer.[19] Once this corner-initiated crack propagates, the stress required for further growth of the crack decreases. When this stress becomes equal to the stress required to initiate the crack in the next layer containing 60 phr CB, the crack opens up in that layer too and advances towards 0 phr layer as well. In 100–66–33–0–0–0–0–33–66–100 FGPC, crack propagates halfway till it reaches to 0 phr layer and again gets arrested (see amplified part of the Fig. 5). The other half of this FGPC takes up the load and increases the modulus again. Unable to bear the load, this part also fails without much extension.

Dynamic mechanical analysis

Most of the polymeric components deform dynamically, so these FGPCs also satisfy some specified dynamic properties. The addition of fillers has a substantial influence on the static as well as dynamic behavior of the polymer. Significant changes are observed in the dynamic mechanical properties of FGPCs when CB is dispersed spatially in the polymer. Storage modulus, loss modulus, loss tangent, etc., of UDPCs and FGPCs are measured under bending mode. Storage modulus (E') of both UDPCs and FGPCs is decreasing with increasing temperature as shown in Figure 6. CB loading enhances the effective crosslink density from entanglements that dies out with temperature and hence storage modulus decreases. The storage modulus of 50 phr CB filled UDPC is comparatively less at room temperature than its all graded counterparts having the same average amount of filler. The rate at which it decreases with temperature is also faster compared with the graded sheets. The probability of agglomerate formation in UDPCs may be higher as the CB is dispersed uniformly throughout the matrix and hence disruption of the agglomerate with temperature will also be higher. This leads to higher rate of decay of storage modulus of UDPCs than the corresponding FGPCs as shown in Figure 6. At 100 Hz, the values of storage modulus at room temperature for UDPC and for 100–80–60–40–20–0–0–20–40–60–80–100 FGPCs are 4.2×10^7 and 5.7×10^7 Pa, respectively. At 100°C, the values of storage modulus for UDPCs and 100–80–60–40–20–0–0–20–40–60–80–100 FGPC are 1.2×10^7 and 3.7×10^7 Pa, respectively. This FGPC shows 98% higher tensile modulus than 50 phr CB filled UDPC at 100% elongation. All the graded combinations with "100 layer up" in the dynamic bending mode show higher modulus than the "0 layer up."

At room temperature, the loss modulus (E'') of this UDPC is also higher than all FGPC grades and the rate at which it decreases with temperature is also higher (see Fig. 7) Particulate fillers hurdle the segmental mobility of the matrix causing an increase in the internal viscosity. This internal viscosity and hence loss modulus decreases with temperature as it enhances the segmental mobility. With higher agglomerate formation in uniformly dispersed filler, breakdown of these agglomerates will also be higher than FGPCs. At 100 Hz, the values of loss modulus

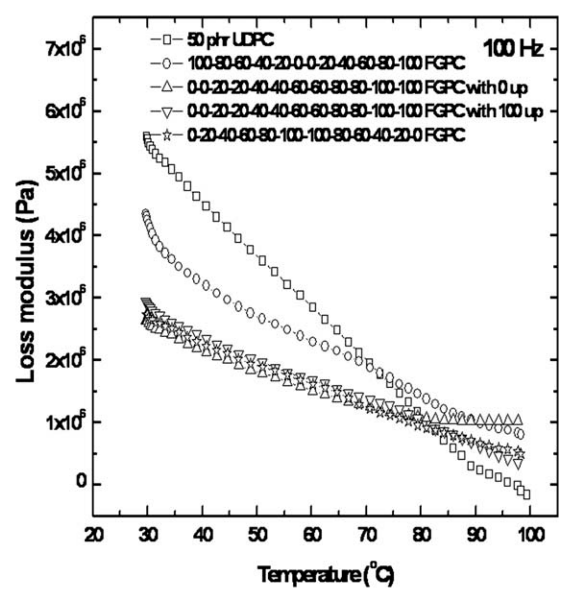

Figure 7 Comparison of loss modulus of 50 phr UDPC and different FGPC sheets employing the same average amount of filler content.

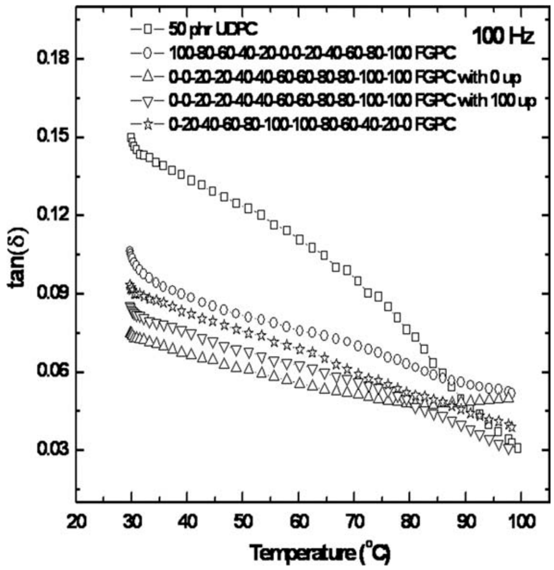

Figure 8 Comparison of loss tangent of 50 phr UDPC and different FGPC sheets employing the same average amount of filler content.

at room temperature for UDPCs and for 100–80–60–40–20–0–0–20–40–60–80–100 FGPC are 5.6×10^6 and 4.3×10^6 Pa respectively.

The tan (δ) of UDPC is also much higher at room temperature than its FGPC counterparts and also decreases at higher rate with temperature (see Fig. 8). At 100 Hz, the values of tan (δ) at 30°C for UDPC and for 100–80–60–40–20–0–0–20–40–60–80–100 FGPC are 0.15 and 0.09, respectively. Higher values of tan (δ) show higher hysteresis loss.[21] Hysteresis loss in UDPCs is higher than the FGPCs having any ascending/descending stacking sequence of the layers. The "0–0–20–20–40–40–60–60–80–80–100–100 with 0 up" FGPC shows the least value of tan (δ) and is 0.08.

CONCLUSIONS

The FGPCs were synthesized by the construction-based layering method using SBR as matrix and CB in graded form. These FGPCs were compared with those corresponding UDPCs that employ same average amount of nanofiller. The average amount of nanofiller in both UDPCs and FGPCs were varied and the effect of different stacking sequences on FGPCs was also ascertained to see its impact on the mechanical properties. When compared the mechanical properties of UDPCs and FGPCs, following conclusions were drawn.

- Hardness of FGPCs, a surface property of the component, varies from UDPCs depending upon the stacking sequence used to make these FGPCs. Specific gravity of UDPCs and FGPCs will remain same as an average amount of filler content is equal in both cases.
- "0 phr layer" at the outer ends in the stacking degrade the properties of FGPCs. For higher average amount of CB, modulus enhancement is compensated by the impairment of ultimate properties i.e., tensile strength, elongation at break. This ultimate properties deterioration problem can be worked out with a solution of maximizing the number of layers in a stack (i.e., smoother gradation).
- In an "ascending–descending–ascending" stacking, higher deviation of CB from the average value enhances the modulus. It correspondingly lowers the ultimate properties with increasing deviation.
- For lesser CB phr levels, the ultimate properties turned comparable or even higher than UDPCs along with the enhanced modulus values. Smoother gradation of the nanofiller (i.e., lesser difference between phr levels of CB in the adjacent layers of the stack) may be expected to be one of the reasons for this enhancement. All the layers employed in a stack contain less than 50 phr CB. This also may be the other reason for the enhancement of the ultimate properties.

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