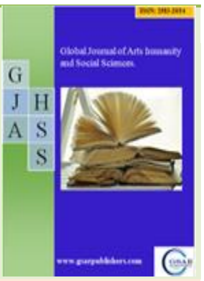
Global Journal of Arts Humanity and Social Sciences
ISSN: 2583-2034

Global Journal of Arts Humanity and Social Sciences
ISSN: 2583-2034
Abbreviated key title: Glob.J.Arts.Humanit.Soc.Sci
Frequency: Monthly
Published By GSAR Publishers
Journal Homepage Link: https://gsarpublishers.com/journal-gjahss-home/



Volume - 6 | Issue - 1 | January 2026 | Total pages 125-133 | DOI: 10.5281/zenodo.18413834

OBTAINING A BINARY SYSTEM BASED ON STYRENE–BUTADIENE–STYRENE POLYMER USING CARBON NANOTUBES AND DETERMINATION OF THE FLOW

By

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Article History
Received: 22- 01- 2026
Accepted: 26- 01- 2026
Published: 29- 01- 2026

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Abstract

This work is devoted to the preparation and investigation of a composite based on styrene–butadiene–styrene (SBS) polymer using carbon nanotubes**.** Nanocomposites based on styrene–butadiene–styrene polymer (SBS) and carbon nanotubes were prepared by the emulsion mixing method, while the vulcanization process was carried out in accordance with standard test methods used for SBS evaluation.

The incorporation of carbon nanotubes (CNTs) into the rubber compound improved the compatibility of the components, and the resulting nanocomposite exhibited high thermal stability. The nanomaterial enhanced the processability of SBS rubber–based composites. Compared with rubber reinforced solely with carbon black, the use of nanomaterials led to a 27% increase in stress at 100% permanent elongation. In addition, partial replacement of carbon black resulted in an increase in the elongation at break of the vulcanizate from 424.5% to 554.0%**.**

The mechanical properties of the obtained rubbers demonstrated that they can be regulated by varying the ratio of carbon nanotubes and carbon black. Based on this, the mass fraction of carbon black was reduced in order to shorten the mixing time at the final stages of mechanical compounding, and SBS/CNT nanocomposites were prepared.

The incorporation of carbon nanotubes into polymers increases the material's hardness, elasticity**,** wear resistance, and resistance to thermal effects. In this regard, the preparation of SBS–CNT composites and the systematic study of their physical, mechanical, and morphological properties are of significant scientific and practical relevance. This research may play an important role in the development of new materials and the optimization of their properties.

Due to their excellent electrical conductivity, the use of carbon black (CB) and carbon nanotubes (CNTs) in a 3:1 ratio as conductive fillers has been shown to enable the production of materials exhibiting both high electrical conductivity and excellent mechanical strength**.**

Keywords: *Styrene–butadiene–styrene rubber, carbon black, CNT, modification, vulcanization, rheology.*

INTRODUCTİON

Styrene–Butadiene–Styrene (SBS) is a triblock copolymer belonging to the class of thermoplastic elastomers**,** and it is one of the most important materials combining both elastic and thermoplastic properties [1–4]. Its structure consists of a linear PS–PB–PS block chain, where polystyrene (PS) blocks are located at both ends and a polybutadiene (PB) segment is positioned in the middle. This unique architecture provides SBS with rubber-like elasticity as well as thermal form stability. The polybutadiene segment imparts flexibility and elongation capability, while the polystyrene domains act as rigid phases that ensure mechanical strength. Consequently, SBS behaves as an elastomer under ambient conditions, whereas upon heating it softens like a thermoplastic and becomes moldable [5–8].

SBS is typically synthesized via anionic polymerization, which enables the production of polymers with high molecular weight and narrow molecular weight distribution. During synthesis, styrene is polymerized first, followed by the addition of butadiene, and finally another styrene polymerization step, resulting in the formation of the triblock structure. The type of catalyst used and the monomer ratios directly determine the polymer's properties such as elasticity, stiffness, and thermal stability. An increase in butadiene content results in a softer and more elastic material, whereas a higher styrene fraction leads to increased rigidity and mechanical strength [9–14].

The chemical structure of SBS contains both unsaturated carbon chains and aromatic rings**,** making it readily identifiable by Fourier Transform Infrared Spectroscopy (FTIR) through characteristic absorption bands. In the spectrum, deformation and stretching vibrations of C–H bonds appear at approximately 1450 cm^{-1} and 2920 cm^{-1}, respectively, while absorption bands characteristic of butadiene and styrene groups are observed around 966 cm^{-1} and 700 cm^{-1}**.** These peaks serve as key indicators for monitoring the polymer's chemical structure and changes occurring due to oxidation. During aging, C=C bonds react with oxygen, leading to the formation of carbonyl (C=O) and hydroxyl (–OH) groups, which manifest as additional bands in the spectrum. The analytical method based on the PB/PS calibration curve developed by Canto et al. allows for the accurate determination of composition ratios in SBS copolymers [15–21].

The physical and mechanical behavior of SBS can be explained by its two-phase morphology, where polystyrene domains function as rigid phases and the polybutadiene matrix serves as the elastic phase. At low temperatures, the polystyrene blocks remain solid, reinforcing the elastic matrix, while at elevated temperatures these domains soften, resulting in thermoplastic behavior. This characteristic makes SBS a recyclable polymer capable of maintaining high performance under various conditions [22–28].

SBS possesses a wide range of applications. One of its most significant uses is in asphalt modification**,** where its incorporation significantly enhances elasticity, resistance to cracking, and thermal stability. In addition, SBS is widely used in the production of footwear and sports equipment, where it is considered an ideal material for manufacturing flexible, soft, and wear-resistant soles. In the automotive industry, SBS-based thermoplastic elastomer (TPE) compounds are employed in the production of seals, protective coatings, steering wheel grips, and similar components. The mechanical durability and dimensional stability of SBS ensure reliable performance of these parts under both low- and high-temperature conditions [29–30].

Other important applications of SBS include hot-melt adhesives, mastics, and elastic coatings. Such adhesives are widely used in the packaging, furniture, and textile industries due to their high adhesion strength, temperature resistance, and flexibility. Furthermore, SBS is blended with polystyrene for the production of high-impact polystyrene (HIPS)**,** which is suitable for household appliances, toys, and electronic housings. In the packaging sector, SBS is utilized in the manufacture of transparent, elastic, and mechanically durable films [31–34].

One of the main drawbacks of SBS is its sensitivity to oxidation and ultraviolet (UV) radiation**.** Unsaturated C=C bonds readily degrade under exposure to oxygen and UV light, leading to yellowing and deterioration of mechanical properties. To mitigate this issue, antioxidants, UV stabilizers, and nanoscale additives (such as SiO_2, TiO_2, and clay nanoparticles) are incorporated during synthesis and processing. These additives enhance the aging resistance of SBS and extend its long-term stability and service life [35–37].

Overall, styrene–butadiene–styrene copolymer is a multifunctional and high-value material in modern polymer technology. Its molecular architecture combines thermoplastic and elastomeric characteristics, enabling extensive industrial applications. From asphalt modification to footwear, automotive manufacturing, adhesives, and packaging technologies, SBS occupies an indispensable position among engineering materials due to its mechanical strength, elasticity, and ease of processing. Moreover, its chemical stability and heat resistance make SBS an ideal material for construction applications and high-temperature industrial environments.

The electromechanical performance of SBS–CNT composites strongly depends on the styrene/butadiene ratio, the CNT loading level, and the matrix architecture (linear or radial structure). SBS with higher styrene content exhibits increased stiffness and reduced elasticity but improved electrical conductivity; conversely, higher butadiene content provides greater elasticity at the expense of conductivity. By selecting an optimal composition, a balance between mechanical integrity and conductive behavior can be achieved.

Such composites demonstrate promising application potential not only in sensor technologies, but also in electromagnetic shielding coatings**,** thermal management systems, energy storage devices, and flexible electronic components. For instance, SBS–CNT-based composites can achieve electromagnetic absorption efficiencies of up to 99%**,** making them highly relevant for communication and military technologies. Additionally, these materials are used in vibration-damping layers, flexible battery components, elastic conductors, and sensing elements of smart wearable devices [38–40].

Future development of SBS–CNT nanocomposites is primarily focused on improving **CNT** dispersion, enhancing the chemical stability of interfacial bonding, and maintaining long-term mechanical flexibility. If surface functionalization of CNTs and matrix selection are further optimized, these materials may become key components in future industrial and high-tech applications, including robotics, automotive engineering, power transmission systems, and smart structural materials [41–42].

Our literature survey indicates that SBS rubber is one of the most important polymers used worldwide. In order to utilize this valuable polymer more efficiently, numerous researchers have conducted extensive scientific investigations. Nevertheless, despite these efforts, the full potential of SBS applications has not yet been realized. Therefore, intensive research is currently underway to expand and diversify the application areas of SBS.

METHOD

Preparation and Mixing Technology of SBS-CNT Composites

For the preparation of the nanocomposites, multi-walled carbon nanotubes (MWCNTs) were first dispersed in a predetermined amount of toluene and subjected to ultrasonication in a Bandelin Sonorex Super RK106 ultrasonic bath for 6 hours. This step was intended to reduce nanotube agglomeration and to ensure good dispersion within the solvent. Subsequently, SBS polymer was added to the dispersion at a defined ratio (5.5 mL of toluene per 1 g of SBS) and mechanically stirred until complete dissolution was achieved. The resulting homogeneous solution was cast as a thin layer onto a clean glass surface and allowed to evaporate at room temperature, yielding thin, elastic, and homogeneous composite films.

The distribution and morphology of MWCNTs within the SBS matrix were examined using scanning electron microscopy (SEM, Philips X230 FEG) based on both surface and fracture images. Prior to SEM analysis, the samples were cryogenically fractured in liquid nitrogen and subsequently gold-coated by sputter deposition to prevent surface charge accumulation and to enhance image quality.

The electrical conductivity and resistance of the composites were measured using a Keithley 487 automated picoammeter/voltage source. During the measurements, the applied voltage was varied from −10 V to +10 V. Based on the obtained current–voltage (I–V) characteristics, the electrical resistance (R) was determined, after which the volume resistivity (ρ) was calculated using the following equation:

$$\rho = RLA \qquad (1)$$

RESULTS OF EXPERIMENTAL RESEARCH

Study of rheological properties

Determination of the melt flow index (MFI) of polymer composite systems is of great importance for establishing their processing parameters. Taking these considerations into account, a composite based on SBS + CNT was prepared according to the formulation given below (Table 1).

Table 1. Formulation of the CNT-based composite.

Names of Components		Sample K-1 Component amount, gr	Sample K-2 Component amount, gr	Sample K-3 Component amount, gr	Sample K-4 Component amount, gr	Sample K-5 Component amount, gr
Styrene–butadiene–styrene rubber		100	100	100	100	100
Carbon Nanotube		1	2	3	4	5
Total		101	102	103	104	105

The binary mixture was prepared in an extruder at a temperature of 190 **°C** for 3 minutes.

For the binary mixtures, the flow time of the composite melts through a 2 mm capillary over a fixed distance (S = 0.002 cm) was determined (in seconds) at different temperatures (150, 170, 190, and 210 °C) under four different applied loads. The obtained results are presented in Table 2.

Table 2. Time required for the flow of the melts of the SBS/CNT binary system (in seconds)

145℃				
Loads / Samples	G_1	G_2	G_3	G_4
K-1	43.18 40.12 41.35	10.92 18.85 11.24	8.47 7.74 8.09	5.29 4.62 4.95
K2	44.86 46.12 45.58	13.78 13.45 13.22	9.61 10.38 9.97	6.67 6.15 6.34
K3	47.32 48.05 47.61	16.71 13.64 13.41	13.18 13.64 13.41	8.76 7.94 8.33
K4	49.42 53.18 51.09	17.94 16.86 17.85	14.48 14.91 14.65	8.18 9.48 8.83
165℃				
K1	31.02 29.85 30.21	8.62 9.19 8.41	6.23 5.48 5.79	2.41 3.28 2.89
K2	31.58 31.12 30.89	9.15 9.57 9.31	6.74 5.48 6.35	3.21 3.15 3.18
K3	33.42 33.65 33.29	11.08 8.42 9.74	7.29 7.48 7.36	3.81 4.08 3.96
K4	33.89 31.05 32.41	9.62 10.15 9.88	7.58 7.76 7.69	4.23 4.36 4.29
185℃				

K1	19.92 19.15 19.48	6.48 6.01 6.24	4.79 4.23 4.51	1.74 2.04 1.88
K2	21.54 18.72 20.05	6.64 5.98 6.29	4.45 4.20 4.33	1.72 1.54 1.62
K3	20.56 18.74 19.52	4.51 4.65 4.58	5.17 6.59 5.92	2.27 3.25 2.78
K4	23.94 22.45 23.08	6.47 6.02 6.25	5.63 6.98 6.31	3.24 3.46 3.35

The applied loads were, respectively: G1 = 11.75 kg, G2 = 20.85 kg, G3 = 26.85 kg, and G4 = 34.85 kg. The pressure generated on the sample surfaces due to the applied loads was calculated primarily using the following equation (2).

$$P=\frac{G}{\frac{\pi D^2}{4}}=\frac{G}{\frac{3.14\cdot(0.954)^2}{4}}=\frac{G}{0.7144} \qquad (2)$$

$P_1=\frac{G_1}{0,8321}=\frac{11,75}{0,8321}=16,9701946\ \text{kg/sm}^2$ $P_3=\frac{G_3}{00,8321,}=\frac{26,85}{0,8321}=32,267756\ \text{kg/sm}^2$

$P_2=\frac{G_2}{0,8321}=\frac{20,85}{0,8321}=25,0570844\ \text{kg/sm}^2$ $P_4=\frac{G_4}{0,8321}=\frac{34,85}{0,8321}=\ 41,881985\ \text{kg/sm}^2$

After modification of the SBS polymer with carbon nanotubes, the prepared samples were extruded through a capillary. For this purpose, the shear stress was calculated (Equation (3)).

$$\tau=\frac{P\cdot r}{2l}=\frac{P\cdot 0.05}{2\cdot 0.8} \qquad (3)$$

Here, P is the pressure generated on the sample surface as a result of the applied loads; τ is the shear stress; r is the radius of the capillary used; and l is the length of the capillary. The calculations were performed as follows:

$\tau_1=\frac{P_1\cdot 0,05}{1,6}=\frac{16,9701946\ .0,05}{1,6}=0,530318\frac{kq}{sm^2}\cdot 9,806\cdot 10^4=5,20029833\cdot 10^4\ Pa$

$\tau_2=\frac{P_2\cdot 0,05}{1,6}=\frac{25,0570844\cdot 0,05}{1,6}=0,783656\frac{kq}{sm^2}\cdot 9,806\cdot 10^4=7,68453\cdot 10^4 Pa$

$\tau_3=\frac{P_3\cdot 0,05}{1,6}=\frac{32,267756\cdot 0,05}{1,6}=1,008365\frac{kq}{sm^2}\cdot 9,806\cdot 10^4=9,888027\cdot 10^4 Pa$

$\tau_4=\frac{P_4\cdot 0,05}{1,6}=\frac{41,881985\cdot 0,05}{1,6}=2.094095\frac{kq}{sm^2}\cdot 9,806\cdot 10^4=120,5346955\cdot 10^4 Pa$

To calculate the volumetric flow rate of the melts of the composites modified with carbon nanotubes at different compositions, Equation (4) was used.

$$Q=\frac{S}{t}\text{x}\frac{\pi D^2}{4}=\frac{0,02}{t}\text{x}\frac{3,14x(0,954)^2}{4}=\frac{0,014288}{t} \qquad (4)$$

The values calculated based on the given equation and their decimal logarithms are presented in Table 3.

Table 3. Calculated volumetric flow rate (Q) values of the SBS+CNT composite (cm³/s)

145℃				
Samples	G1	G2	G3	G4
K1	0,0003625	0,0013289	0,0018426	0,0030184
K2	0,0003341	0,0011423	0,0015089	0,0023547
K3	0,0003184	0,0009628	0,0011297	0,0018126
K4	0,0002961	0,0008796	0,0010364	0,0017068
165℃				
K1	0,0004986	0,0017894	0,0025867	0,0053182
K2	0,0004862	0,0016128	0,0023795	0,0047429
K3	0,0004528	0,0015489	0,0020696	0,0038247
K4	0,0004685	0,0015297	0,0019823	0,0035069
185℃				
K1	0,0007765	0,0024158	0,0033486	0,0080943
K2	0,0007569	0,0023947	0,0034972	0,0093986
K3	0,0007751	0,0032864	0,0025698	0,0054681
K4	0,0006589	0,0024183	0,0023875	0,0044862

In this study, the volumetric flow rate of the composite (log Q) was calculated, and the obtained results are presented in Table 4.

Table 4. Calculated volumetric flow rate of the SBS/CNT mixture (log Q values).

150℃				
Samples	G1	G2	G3	G4
K1	−3.4682	−2.9014	−2.7586	−2.5449
K2	−3.5039	−2.9687	−2.8468	−2.6543
K3	−3.5261	−3.0395	−2.9756	−2.7698
K4	−3.5586	−3.0859	−3.0134	−2.7957
170℃				

K1	−3.3296	−2.7718	−2.6119	−2.2974
K2	−3.3403	−2.8169	−2.6487	−2.3476
K3	−3.3728	−2.8334	−2.7105	−2.4419
K4	−3.3871	−2.8398	−2.7282	−2.4806
190℃				
K1	−3.1354	−2.6298	−2.4986	−2.0345
K2	−3.1471	−2.6431	−2.5098	−2.0492
K3	−3.1665	−2.6649	−2.6147	−2.2853
K4	−3.2098	−2.6895	−2.6446	−2.3719

Based on the calculations corresponding to the data presented in the tables, the log γ̇–log τ dependence (shear rate and shear stress) for the SBS/CNT binary system at 150 °C, 170 °C, and 190 °C was constructed, and the resulting curves are shown in Figures 1, 2, and.3.

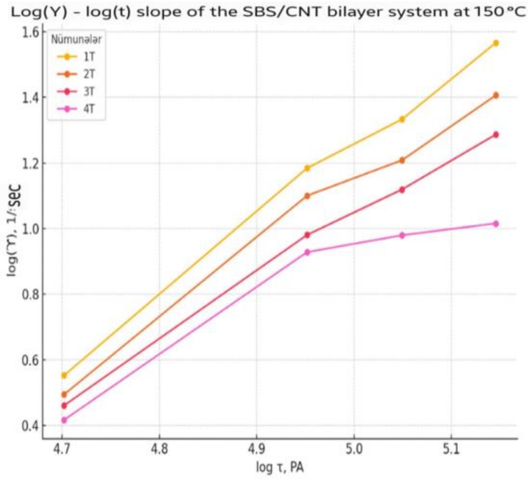
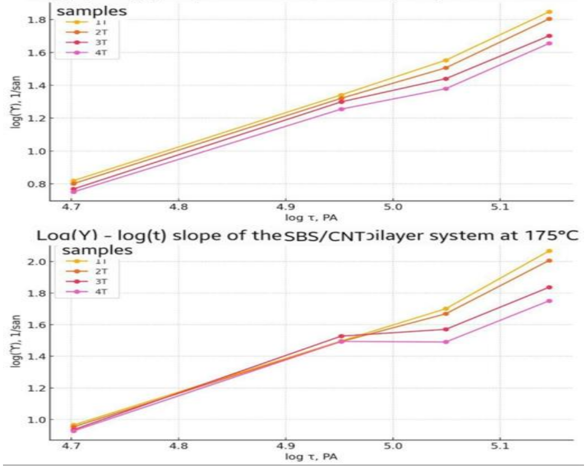


Fig. 1. Dependencies of log(γ̇) and log(τ) for the SBS/CNT composite at 150 °C



Figs. 2–3. Dependences of log(γ) and log(τ) for the SBS/CNT composite at 170 °C and 190 °C

Analysis of log(γ)–log(τ) curves at 140 °C:
The graphical curves obtained at 140 °C indicate that, in the SBS/CNT binary system, an increase in shear stress leads to a corresponding increase in the logarithmic value of the shear rate. This behavior suggests that the viscoelastic properties of the material become softened under the influence of temperature, accompanied by increased molecular mobility within the structure. As the temperature rises, the freedom of movement of chain segments increases, thereby enhancing the flowability of the system. Among the obtained curves, sample 1T exhibits the highest log(γ̇) values, indicating lower internal resistance and higher SBS/CNT This behavior can be attributed to fewer structural constraints and weaker intermolecular interactions within this sample. In contrast, sample 4T shows relatively lower log(γ̇) values, suggesting the presence of a higher fraction of rigid phases and stronger intermolecular interactions in its structure. In such systems, flowability is limited because high structural density and molecular entanglements reduce material mobility. Therefore, depending on temperature and composition, the factors influencing shear rate in the SBS/CNT system can be associated not only with mechanical effects but also with changes occurring at the molecular level.

Analysis of log(γ)–log(τ) curves at 160 °C:
The curves constructed at 160 °C exhibit a more pronounced increasing trend. At this temperature, enhanced thermal activity in the polymer system allows molecular chains to move more freely, directly affecting flow behavior. The analysis shows that, for all samples, log(γ̇) values increase in parallel with log(τ). However, the intensity of this increase varies depending on the compositional characteristics of the samples.

The highest log(γ) values are again observed for sample 1T, indicating that this blend maintains its flow ability and mechanical flexibility even at elevated temperatures. Conversely, sample 4T displays lower log(γ) values, suggesting the dominance of structural stability and internal resistance. This sample demonstrates lower sensitivity to temperature effects and exhibits more stable and mechanically rigid behavior.

Analysis of log(γ)–log(τ) curves at 175 °C
The log(γ̇)–log(τ) curves obtained at 175 °C reflect the maximum flow ability of the system. Under this temperature regime, a noticeable increase in log(γ) values is observed for all samples, indicating a more pronounced response of the material to elevated temperature. In particular, the rapid increase in log(γ̇) values for samples 1T and 2T suggests that these compositions possess lower structural stability.

In contrast, samples 3T and 4T exhibit a weaker increase in log(γ), indicating a more stable structure and higher resistance to thermal effects. Overall, the observed changes at this stage demonstrate that, with increasing temperature, the viscoelastic behavior limits of the system expand, and thermal stability is directly related to the composition of the composite.

Observations of the melt flow index (g/10 min) of the SBS/KN binary blend indicate that this parameter is significantly influenced by both temperature and the SBS content in the composition. Graphical analysis reveals that the melt flow index increases with rising temperature, indicating that the material transitions to a more fluid state under thermal influence.

The general trend shows that the flowability of the SBS/KN system is simultaneously governed by temperature and the proportion of SBS in the blend. In particular, within the temperature range of 160–175 °C, variations in SBS content exert a stronger influence on flow behavior. However, above 175 °C, the intensity of this effect decreases and the values tend to stabilize, indicating that the system approaches a saturated flow behavior regime.

These results provide an important scientific and practical basis for determining the optimal processing temperature and effective compositional ratios of the SBS/KN system (Figure 4).

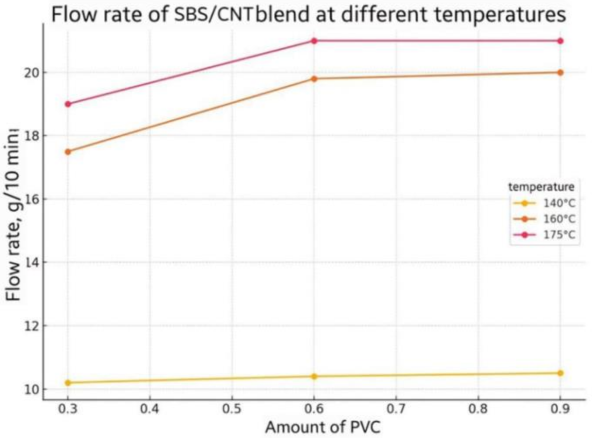


Fig. 4. Determination of the flow index of the SBS/KN blend at different temperatures under constant stress

For the purpose of studying rheology, the effective viscosity was ultimately calculated based on the true values of displacement stress and displacement rate, determined as functions of temperature and applied loads.

The effective viscosity values of the mixtures under the influence of various temperatures and stresses were calculated using the formula (5) given below.

$$\eta_{ef} = \frac{\tau}{\dot{\gamma}} \qquad (5)$$

Here, η_ef is the value of the effective viscosity under the influence of various temperatures and pressures. At a temperature of 140 °C, under load G1

$$\eta_1 = \frac{5{,}0403 \cdot 10^4}{3{,}56820} = 14125{,}61; \ \log 14125{,}61 = 4{,}1505$$

$$\eta_2 = \frac{5{,}0403 \cdot 10^4}{3{,}121} = 28654{,}598; \ \log 28654{,}598 = 4{,}4571$$

$$\eta_3 = \frac{5{,}0403 \cdot 10^4}{2{,}889} = 38752{,}1634; \ \log 38752{,}1634 = 4{,}5887$$

$$\eta_4 = \frac{5{,}0403 \cdot 10^4}{2{,}611} = ; \ \log = 53555{,}726; \ \log 53555{,}726 = 4{,}7244$$

The corresponding results of the effective viscosity obtained from the calculations performed are presented in Table 5., after which the logarithmic values were determined and graphical curves were constructed. The logarithms of the effective viscosity of the melts of the SBS/KN binary system under the influence of various temperatures and loads are given in Table 6:

Table 5. Effective viscosity of the melts of the SBS/KN binary system under the influence of various temperatures and loads

140℃				
Samples	1st load	2nd load	3rd load	4th load
1T	14820.4	3521.8	2489.6	1425.3
2T	29710.2	7425.9	5789.4	3668.1
3T	40190.6	12145.7	8820.3	6035.9
4T	55280.9	17110.6	15120.4	13890.2
160℃				
1T	7985.6	2410.3	1624.7	742.9
2T	14785.2	4489.1	2938.4	1476.2
3T	19940.8	5895.6	4250.7	2338.9
4T	25890.3	8125.4	6020.8	3195.6
175℃				
1T	5710.8	1680.5	1054.3	458.6
2T	10340.9	3015.2	2010.8	924.1
3T	13490.6	3495.7	3120.4	1695.8
4T	17210.5	4695.8	4680.9	2589.4

Table 6. Logarithms of the effective viscosity of the melts of the SBS/KN binary system under the influence of various temperatures and loads.

140℃				
Samples	1st load	2nd load	3rd load	4th load
1T	4.1823	3.5486	3.3928	3.1674
2T	4.4895	3.8839	3.7721	3.5798
3T	4.6218	4.1027	3.9624	3.7946
4T	4.7569	4.2512	4.1983	4.1538
160℃				
1T	3.9127	3.3896	3.1768	2.8824
2T	4.1786	3.6598	3.4729	3.1754
3T	4.3094	3.7782	3.6351	3.3786
4T	4.4238	3.9197	3.7923	3.5214
175℃				

1T	3.7649	3.2335	3.0287	2.6619
2T	4.0261	3.4876	3.3109	2.9728
3T	4.1368	3.5497	3.5061	3.2408
4T	4.2473	3.6809	3.6828	3.4236

After calculating the effective viscosities of the SBS/KN binary system at different temperatures, graphs of the dependence of the effective viscosity (log η_ef) on the displacement stress (log τ) were constructed (Fig. 5).

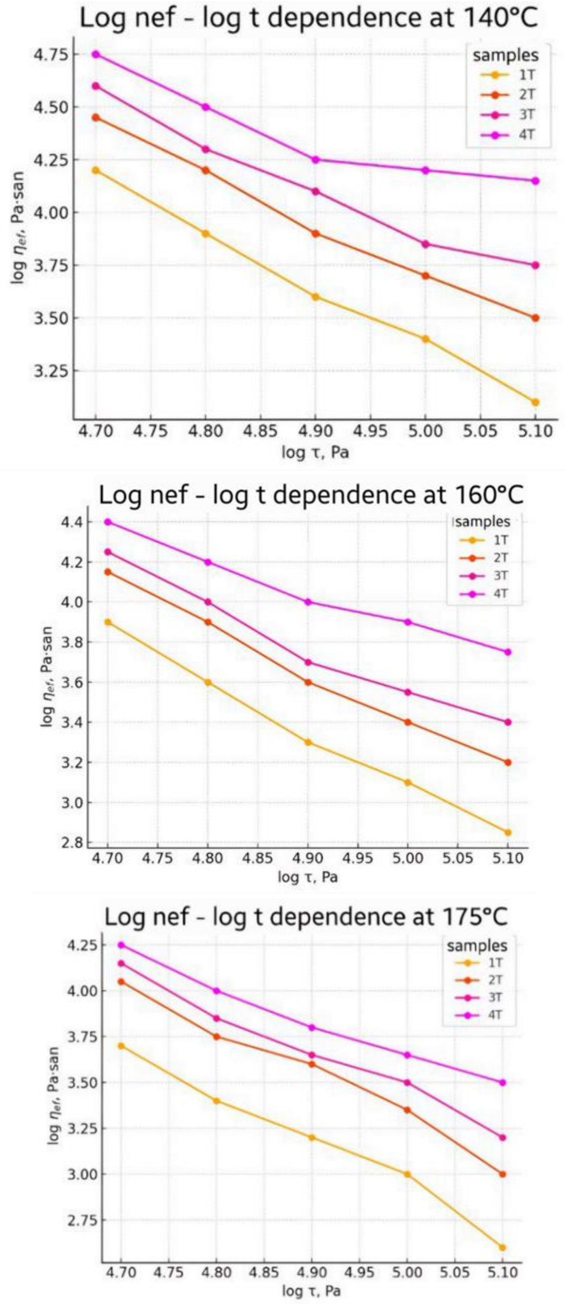


Fig. 5. Dependence of the effective viscosity of the SBS/KN binary mixture on displacement stress at 140 °C, 160 °C, and 175 °C.

RESULTS

To improve the photostability of SBS-based composites, 3–5 parts by weight of carbon nanotubes (CNTs) were added. These additives enhance the stability of the material under solar radiation, ensuring long-term durability.

The results show that nanomaterials such as CNTs and carbon black scatter and reflect UV radiation, preventing its penetration into the polymer structure and reducing macromolecular degradation. Their combined use increases surface area and improves resistance to photodegradation.

The addition of 3□4% CNTs and carbon black nanoparticles to SBS composites increases resistance to photodegradation by approximately 50%, making these materials promising for durable coatings in high solar radiation environments while also improving mechanical strength and thermal stability.

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