Applied *Nanomaterials and smart Polymers,*
Online ISSN: 2981-0434
Vol. 2, No. 5

ANSP

Thermal and Morphology Properties of Polypropylene Composites Reinforced with nSBR and n-GO

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https://doi.org/10.82428/ANSP.2025.1208550

Abstract

The purpose of this research is to investigate the Rheological, Thermal and Morphological properties of polypropylene with Styrene-Butadiene nano elastomer (nSBR) and Graphene nano Oxide (nGO). From the nanocomposites homogenized in the Internal Mixer based on the test design table, SEM morphology, DSC thermal properties were tested. SEM images showed that uniform dispersion and proper interactions between nanoparticles and PP were established. SEM observations of the fracture surface of the composites show that the dispersion of nGO and nSBR in the PP/nSBR/nGO composite happened homogeneously. DSC analysis shows that the crystallinity peaks shift to higher temperatures and the crystallinity of pure PP changes from 117.2 °C and 117.94 °C at the optimal values of PP/nGO/nSBR composite. By adding optimal amounts of nanoparticles to PP, the maximum melting temperature (Tmp) and maximum crystallization temperature (Tcp) increase by 2.5% and 0.8%, respectively, compared to pure PP. The results of thermal analysis show that PP/nSBR/nGO composite has higher thermal stability than pure PP.

Keywords: Polypropylene, Styrene-Butadiene nano elastomer, Graphene nano Oxide, Thermal analysis.

1. Introduction

Polymer nanocomposites reinforced with nano-sized particles (with at least one dimension below 100 nm) exhibit superior thermal and mechanical properties, making them highly attractive for advanced applications across various industrial fields [1]. Among these polymers, polypropylene (PP), as one

of the most widely used polyolefin, is extensively employed in industries such as construction materials, furniture manufacturing, automotive engineering, and toy production [2]. However, a major limitation of PP is its poor impact resistance, particularly at low temperatures. To address this issue, PP is traditionally modified with elastomers to enhance its impact strength [3]. For instance, the toughness of PP can be enhanced using SBR/PP blends by incorporating an elastomeric component. However, this modification simultaneously alters other properties, including thermal characteristics, meaning that changes in thermal behavior will also influence mechanical performance. Additionally, the incorporation of nanoparticles has proven to be an effective method for simultaneously improving the stiffness and toughness of the polymer matrix [4]. Nevertheless, challenges such as the difficulty in achieving uniform nanoparticle dispersion within the polymeric matrix, due to their strong tendency to agglomerate, result in low filler loading efficiency [5]. To address these issues, many researchers have focused on surface modification techniques (chemical or physical treatments) to enhance nanoparticle compatibility and dispersion. Wang and colleagues investigated the mechanical and morphological properties of a synergistically toughened-stiffened polypropylene composite system incorporating styrene-butadiene rubber (SBR) and graphene oxide nano sheets. Their study involved the chemical reduction of graphene oxide to produce reduced graphene oxide (RGO), which was then used to prepare PP composites co-enhanced with both SBR elastomer and RGO nano fillers. Through melt blending processing, the researchers achieved significant improvements in the mechanical properties of PP, as demonstrated by DSC analysis, impact testing, and tensile measurements [6]. Mirzaee research team employed response surface methodology to model and optimize the toughness-strength balance in SBR/PA6 blends using compatibilizers. Their work statistically analyzed and optimized both the grafting degree of glycidylmethacrylate (GMA) and the gel content of GMA-g-SBR to enhance material performance [7]. In 2010, Yi-Li et al. investigated the flame-retardant properties of polypropylene (PP) modified with nano sized elastomer powder [8]. Their study incorporated fully vulcanized powdered rubber (UFPR) into the PP matrix, followed by comprehensive morphological and thermal characterization of the resulting composite. Prachayawasin et al. subsequently examined the effects of nano-scale styrene-butadiene rubber (SBR) as a nucleating agent on the thermal properties, crystallization behavior, and physical characteristics of isotactic polypropylene (iPP). More recently [9], Manli Zhang's research group conducted a systematic study on the influence of elastomeric nanoparticles on the mechanical properties and crystallization kinetics of PP. Their work confirmed that while elastomer nanoparticle incorporation effectively enhances PP toughness, this improvement typically occurs at the expense of reduced modulus [10].

The present study aims to investigate the thermal and morphological properties of polypropylene (PP)/nano-styrene-butadiene rubber (nSBR) composites reinforced with nano-graphene oxide (nGO). Specifically, this research focuses on evaluating the synergistic effects of these nano fillers on the thermal behavior of the polymer matrix.

2. Materials and Methods

2.1. *Materials*

The base polymer matrix consisted of polypropylene (commercial grade L440 EP PP) supplied by Jam Polypropylene Petrochemical Company. This grade was selected for its balanced mechanical

properties and process ability characteristics. Nano-graphene oxide (nGO) was procured from Kara Pazhouh Research Institute (Amirabad University of Technology), with the material originating from Beijing Petrochemical Corporation. The nGO exhibited a specific surface area of 150-200 m^2/g and oxygen content >30%. Nano-scale styrene-butadiene rubber (nSBR) was obtained from the same supplier, with the white powdered elastomer showing the following characteristics: Styrene content: 23.5 ± 2%, Particle size distribution: 80-120 nm, Volatile content: <0.5 wt%.

2.3. *Preparation of PP/nGO/nSBR Nanocomposites*

To prepare the nanocomposites, the required amounts of materials were precisely weighed according to Table 1, using a digital balance. The pre-weighed components were then homogeneously mixed in an internal mixer to ensure uniform dispersion (The temperature was set at 190 °C and the processing time was 8 minutes for each sample, with a rotation speed of 60 rpm.). For the DSC analysis, the samples, after being compounded in an internal mixer, were heated from room temperature to 220 °C at a heating rate of 20 °C/min to ensure complete melting. After reaching the target temperature, the samples were held isothermally for 5 minutes. They were then cooled down to room temperature at the same rate and kept for another 5 minutes. Subsequently, a second heating cycle up to 220 °C at the same rate (20 °C/min) was performed to erase the thermal history of the polymer. For the TGA analysis, the samples were heated from room temperature to 800 °C at a constant heating rate of 10 °C/min.

Table 1. Compositional Formulation of PP/nGO/nSBR Nanocomposites

Run.	Factor 1 nGO(phr)	Factor 2 nSBR(phr)	Factor 3 PP(phr)
1	1.4	2	70
2	0.7	0	70
3	0.7	1	70
4	0.7	2	70
5	0.7	1	70
6	0.7	1	70
7	1.4	1	70
8	0	2	70
9	0.7	1	70
10	0.7	1	70
11	1.4	0	70
12	0	1	70
13	0	0	70

2.4. *Characterization*

The surface morphology of the nanoparticles and nanofibers was examined using field emission scanning electron microscopy (SEM, AIS2100, Seron Technology, Korea). To ensure optimal

imaging quality, all specimens were sputter-coated with a 10-nm gold-palladium layer prior to analysis under high vacuum conditions (10^-5 Torr) at an accelerating voltage of 15 kV.

The thermal behavior of the nanocomposites was evaluated using differential scanning calorimetry (DSC, model Q2000, TA Instruments). Measurements were conducted under a nitrogen atmosphere with a heating rate of 10°C/min across a temperature range of 25°C to 230°C.

The thermal stability of the nanofiber membranes was assessed using thermogravimetric analysis (TGA, Q600 model, TA Instruments, USA). Measurements were carried out under an argon (Ar) atmosphere with a heating rate of 10°C/min over a temperature range of 25–500°C.

For mechanical test, we consider the impact test, because this property is important for PP, V-shaped specimens for the impact test were prepared using a hot press at 190 °C for 5 minutes, followed by cold pressing at 20 MPa. The impact test was then conducted according to ASTM D256:2023. The specimen dimensions were selected as 10 × 1.5 mm². Prior to testing, the samples were conditioned for 40 hours under standard laboratory conditions (23 ± 2 °C and 50 ± 5% relative humidity). According to the standard, the impact test must be performed on at least five specimens..

3. Results

3.1. *Rheological Analysis*

To evaluate the rheological behavior of the samples, nine specimens were prepared according to Table 1, and their mixing process was examined. As illustrated in Figure 1, all samples were successfully mixed and are suitable for subsequent analyses. Since the torque-time diagram serves as an indicator of the rheological behavior of nanocomposites, their rheological characteristics can also be observed during the mixing process.

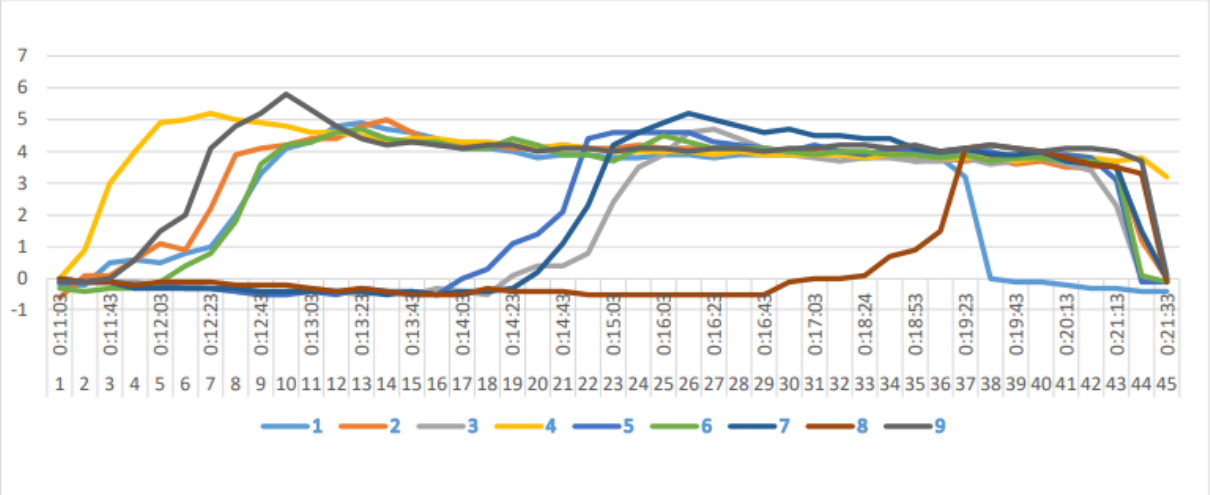

Figure1. Torque Meter of samples in internal mixer during mixing.

3.1. *Morphological Analysis*

Uniform dispersion of fillers within the polymer matrix is a critical parameter significantly influencing the enhancement of mechanical and morphological properties in nanocomposites. Therefore, SEM imaging was employed to analyze the distribution of nanoparticles (GO and SBR) within the polypropylene matrix.

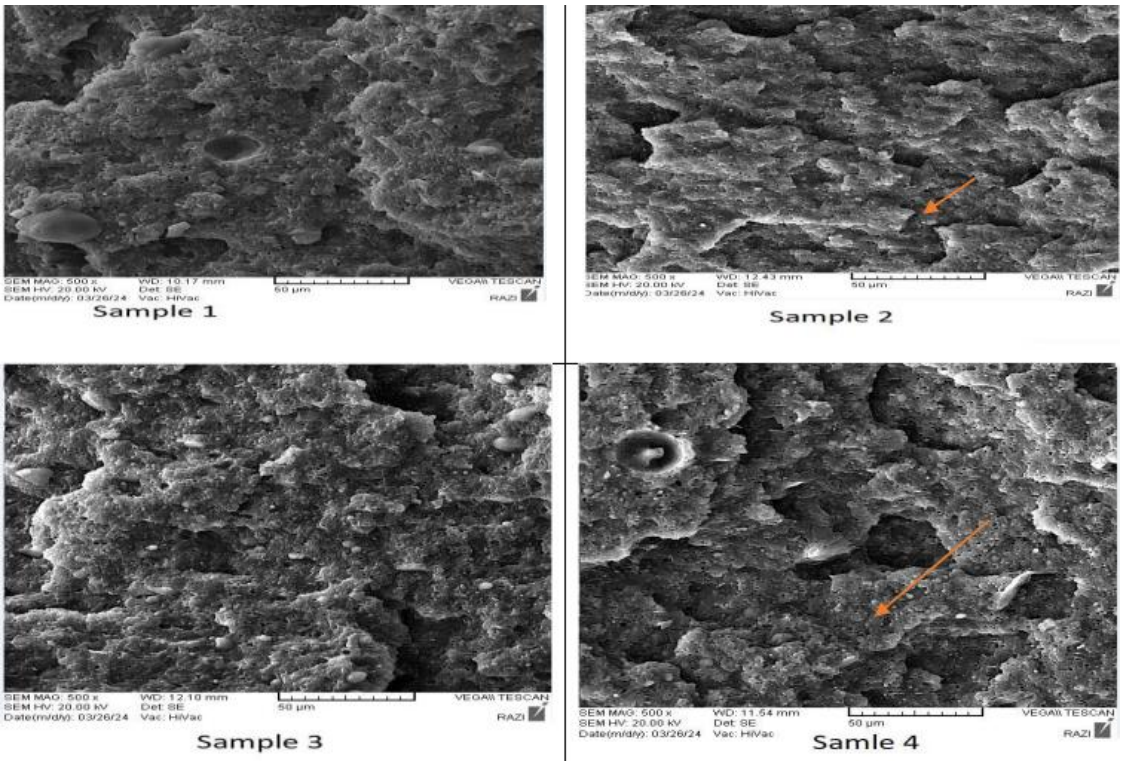

Figure 2. SEM of Some Samples based on Table 1.Sample 1(PP 70+nSBR 2+Ngo 0) & 2(PP 70+nSBR 0+nGO 1.4) & 3(PP 70+nSBR 2+nGO 0.7) & 4(PP 70+nSBR nGO 1.4)

Figure 2, shows the SEM images of the surface and fracture surface of polypropylene containing nanoparticles. Careful examination of the images reveals a uniform distribution of nanoparticles within the polypropylene matrix. The presence of nano-graphene reduces the domain size of SBR nanoparticles.

3.2. Impact Izod Test

Polypropylene (PP) exhibits limited impact resistance at low temperatures, restricting its practical applications. Modification with elastomers, such as styrene-butadiene rubber (SBR), has been commonly employed to enhance the toughness of PP/SBR blends. To further improve mechanical performance, graphene oxide (nGO) can be incorporated as a nano filler due to its high strength and aspect ratio. The combined use of SBR and nGO in PP matrices is expected to synergistically increase impact resistance, offering a promising strategy for developing PP-based composites with superior low-temperature toughness. Result of impact izod present in table3:

Table 3. Impact Izod (Kj/m^2), test for samples prepared based on Table 1

Run	1	2	3	4	5	6	7	8	9	10	11	12	13
Impact izod	14.16	16.65	14.92	11.62	14.20	15.30	17.49	13.38	15.39	15.32	14.94	12.52	13.16

As observed, the neat PP sample exhibited an impact strength of 13.16 kJ/m² (Sample 13), while the PP/nGO sample showed an increased value of 16.65 kJ/m² (Sample 2). The PP/nSBR sample displayed a slightly lower impact strength of 12.52 kJ/m² (Sample 2). Notably, Sample 7, containing PP, nGO, and nSBR, demonstrated the highest impact strength of 17.49 kJ/m², indicating the synergistic effect of both nano fillers on the mechanical properties of the composite.

3.3. *Thermal Properties*

In this method, the samples were first heated from room temperature to 230°C at a rate of 20°C/min and maintained at this temperature for 5 minutes to erase their thermal history. Subsequently, the samples were cooled to room temperature at the same rate of 20°C/min, Figure 3.

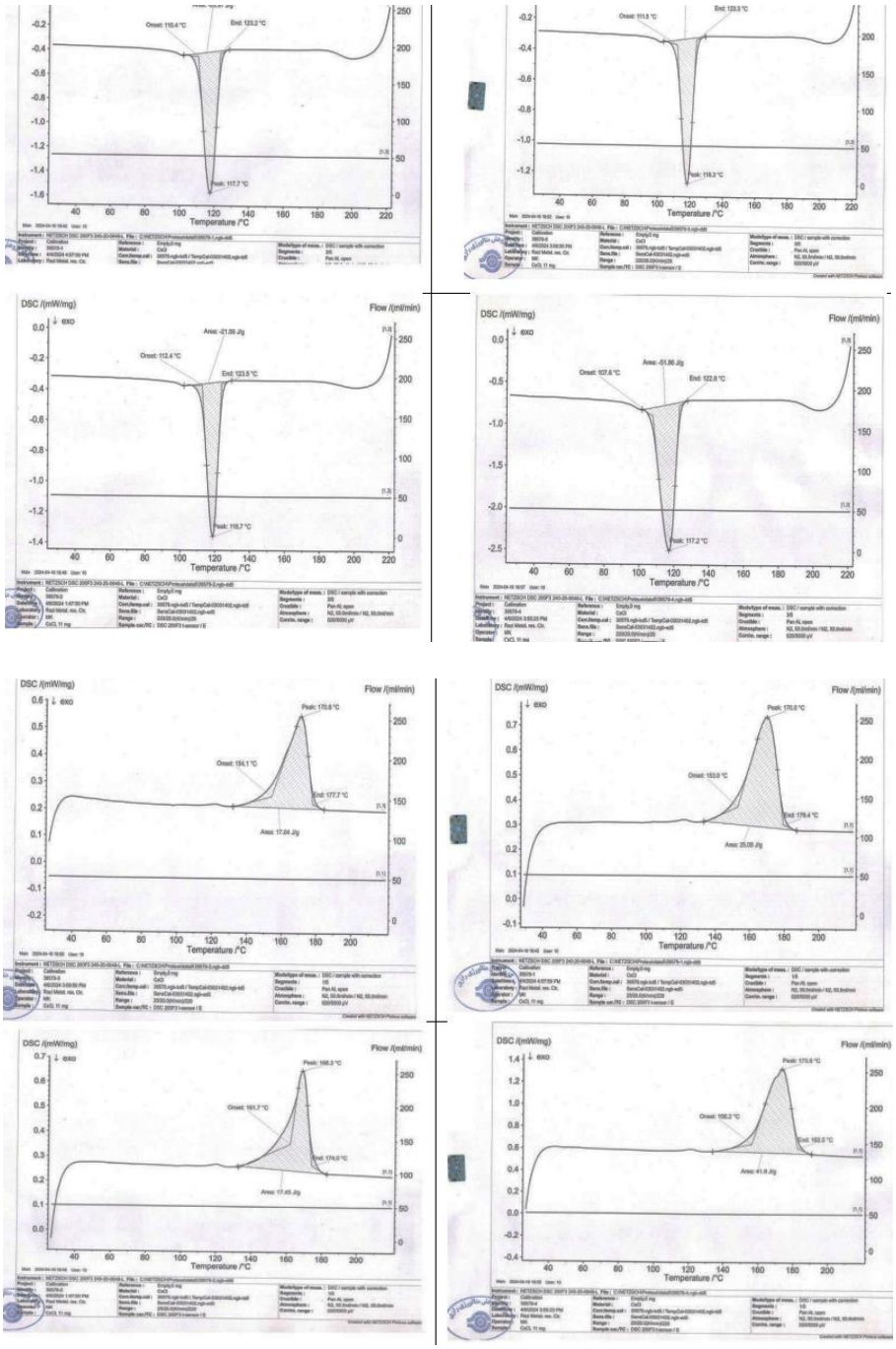

Figure 3. DSC of Some Samples based on Table 1.Sample 1(PP 70+nSBR 2+Ngo 0) & 2(PP 70+nSBR 0+nGO 1.4) & 3(PP 70+nSBR 2+nGO 0.7) & 4(PP 70+nSBR nGO 1.4)

All results were recorded in Table 2. The heating cycle was then repeated to determine the crystallization temperature (Tcp) and melting temperature (Tmp).

Table2. The Crystallinity Temperature (TCP), crystallization enthalpy (ΔHc), peak melting temperature (T_mp), melting enthalpy (ΔH_m), Relative crystallinity (Xc) and relative crystallinity were characterized as functions of both nGO (A) and nSBR (B), variables.

Run	A:GO(phr)	B: nSBR (phr)	TCP(°C)	ΔHC (jg^{-1})	T_{mp} (°C)	ΔHC (jg^{-1})	Xc (%)
1	1.4	2	117.5	39	172	23.7	18.7
2	0.7	0	118	27.6	169	2.5	12.2
3	0.7	1	118.2	32.2	170	40.7	15.4
4	0.7	2	118.3	17.04	171	22.08	8.15
5	0.7	1	118.6	34.1	171	41.61	16.31
6	0.7	1	118.2	33.2	176	48.2	15.88
7	1.4	1	117.3	41.9	174	51.86	20
8	0	2	117.7	35.08	170	28.97	12
9	0.7	1	118.7	33.5	169	50.5	16.02
10	0.7	1	117.9	32.6	172	40.8	15.6
11	1.4	0	118.7	17.45	168	21.56	8.35
12	0	1	117.1	28	168	22.7	13
13	0	0	117.2	20	168	28	9.57

3.4. *Analysis of Variance (ANOVA) for the Relative Crystallinity (Xc)*

Analysis of Variance (ANOVA) for the Relative Crystallinity (Xc) of PP/nSBR/nGO Table 3 presents the results of the ANOVA for the relative crystallinity (Xc). The correlation coefficient (R^2) was found to be 0.6872, indicating that 68.72% of the variation in the improvement of relative crystallinity can be explained by the model. The remaining 31.28% of the variation in the enhancement of relative crystallinity of the nanocomposite cannot be accounted for by the model.

Table 3. ANOVA Results for the Relative Crystallinity (Xc) of PP/nSBR/nGO

Source	**Sum of Square**	**Degree of Freedom**	**Mean of Square**	**F-value**	**p-value**	
Model	109.36	5	21.87	3.08	0.0876	Not Significant
A-nGO	25.96	1	25.96	3.65	0.0977	
B-nSBR	9.96	1	9.96	1.40	0.2753	
AB	15.68	1	15.68	2.21	0.1811	
A^2	4.51	1	4.51	0.6346	0.4518	
B^2	57.10	1	57.10	8.03	0.0253	
Pure Error	0.0000	4	0.0000			
Sum	159.14	12				

The comparison between the predicted and actual values can be observed in the plot presented in Figure 4. A good agreement between the results indicates a strong correlation. Moreover, the data points are scattered around a 45° line, demonstrating the high capability of the model in predicting the responses accurately.

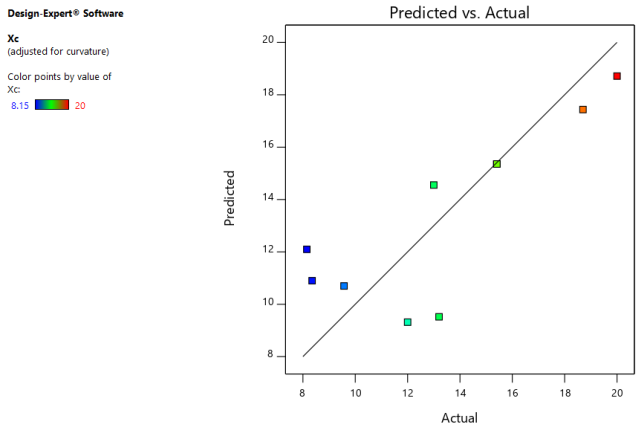

Figure 4. Predicted vs. Actual Plot of the Relative Crystallinity (Xc) for PP/nSBR/nGO

Figure 5, illustrates the simultaneous effect of two parameters, nGO and nSBR, showing a significant interaction of factor A at higher percentages and a pronounced interaction of factor B at intermediate levels.

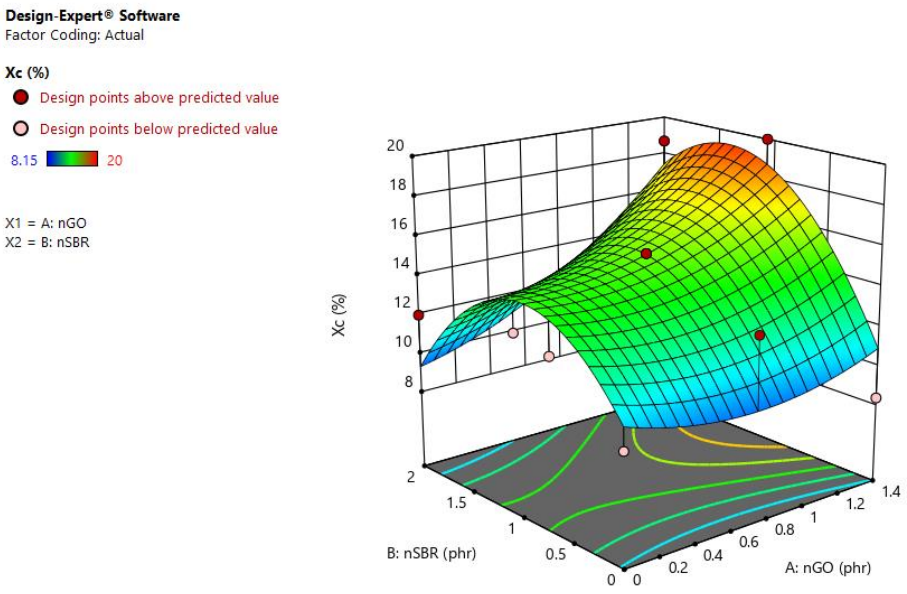

Figure5. Three-Dimensional Plot of the Relative Crystallinity (Xc) for PP/nSBR/nGO

4. Discussion

Morphological analysis of the nanocomposites using scanning electron microscopy (SEM) reveals a homogeneous and well-dispersed distribution of nanoparticles within the polypropylene matrix. Furthermore, the addition of nSBR to the composite improves compositional homogeneity. The rough surface morphology with visible pores in the following image suggests a higher graphene content. As shown in the figure, the composite containing a higher nSBR content appears white in color, exhibits fewer voids, and demonstrates better structural integrity and formation.

According to results the peak melting temperature (Tmp) and peak crystallization temperature (Tcp) of polypropylene containing 1.4 wt% nano-GO and 0.5 wt% nano-SBR increased by 2.5% and 0.8%, respectively, compared to pure polypropylene.

5. Conclusions

This study investigated the rheological, thermal, and morphological properties of polypropylene (PP) reinforced with Styrene-Butadiene nano elastomer (nSBR) and Graphene Nano Oxide (nGO). Nanocomposites were prepared in an internal mixer based on a specified test design, and their morphology and thermal properties were analyzed using SEM and DSC, respectively. SEM images confirmed uniform dispersion and strong interactions between the nanoparticles and the PP matrix. Fracture surface analysis indicated homogeneous distribution of nGO and nSBR within the composite. DSC results revealed that the crystallinity peaks shifted to higher temperatures, with the crystallinity of pure PP increasing from 117.2 °C to 117.94 °C in the optimal nanocomposite formulation. The addition of nanoparticles led to increases in the maximum melting temperature (Tmp) by 2.5% and the maximum crystallization temperature (Tcp) by 0.8% compared to pure PP. Overall, thermal analysis demonstrated that the PP/nSBR/nGO nanocomposite exhibits higher thermal stability than pure PP.

Acknowledgments

The authors gratefully acknowledge the scientific support from the Nanotechnology Research Center and Iranian Islamic Fashion and Clothing Research Center, Islamic Azad University, South Tehran Branch.

Nomenclature

Symbols:
Tcp = Crystallinity Temperature (°C)

Tmp= Melting Point (°C)

Abbreviations:
GO = Graphene Oxide

n-SBR= Nano SBR

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