

Research Article

Evaluation of PE Films Having NIR-Reflective Additives for Greenhouse Applications in Arid Regions

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Linear-low-density-polyethylene- (LLDPE-) based formulations with several near-infrared- (NIR-) reflective pigments were prepared by melt blending technique and their subsequent films were prepared by blown film extrusion process. Thermal properties of these films were evaluated using differential scanning calorimetry (DSC). The results showed that the melting and crystallization temperatures (T_m and T_c , resp.) of these formulations were almost similar to that of control resin. The melt viscosity was measured by stress-controlled rotational rheometer and melt flow index (MFI) instruments. Rheological measurements indicated that the blend formulations with NIR-reflective additive have similar melt viscoelastic behavior (storage modulus and dynamic viscosity) to the control resin. The mechanical test performed on NIR-reflective films showed similar values of tensile strength for blend samples as that of control resin. The spectral radiometric properties of the blend films were evaluated in the solar wavelength range of 200–1100 nm and found to be improved over the control sample without having NIR-reflective pigment.

1. Introduction

Climate in arid regions is characterized by hot, dusty, dry, and long summer. The maximum ambient temperature in these regions is usually well above 45°C, solar radiation flux is quite high (up to 1200 Wm⁻²), and the relative humidity is as low as 10%. Moreover, the water resources are scarce and brackish [1]. Such harsh weather conditions negatively affect the overall production of the crops. One of the most common ways to protect the crops from excessive solar radiation is to use the greenhouse coverings [2]. They protect crops from adverse weather conditions, such as rain, wind, heat burn, insects, and diseases [3]. Nevertheless, the harsh weather conditions also affect the shelf life of greenhouse covering material and rapidly degrade their optical and mechanical properties. Some of the practical methods adopted to overcome the heat buildup inside greenhouse are ventilation and evaporative cooling systems [4]. Ventilation can suitably be adopted in the

regions where ambient temperature is around 30°C. However, in the extreme summer conditions, it is uncommon to use a ventilation method alone, since it replaces the overheated greenhouse air with a very hot ambient air and cannot provide adequate cooling [5–8]. Evaporative cooling (wet pad-fan system or fogging system) is alternatively used to provide cool and moist air for plant growth during summers. The most essential requirement for such system to operate efficiently in an arid climate is the availability of pure and fresh water resource that can be used for continuous wetting of the pad and pumping it through the nozzles of a fogging system. However, in arid regions such as Saudi Arabia, the water resources are saline and brackish. The use of such saline water causes fast deterioration in the cooling performance of the wet pad-fan systems. The clogging by the salt buildup on the pad surfaces restricts the air flow leading to an increased electrical energy consumption as well as temperature rise inside the greenhouse [9, 10]. For these

reasons, ventilation and evaporative cooling techniques are not practically suitable to adopt in this region. A greenhouse covering that can reduce the solar irradiative load on crops by preventing a portion of unwanted incident radiation would be ideal for agriculture applications in arid regions.

Among the choice of greenhouse covering materials, plastics are more suitable than glass principally for their cheapness, lightness, and large size features [11]. Mechanical properties of a greenhouse covering film are quite an important factor, as it undergoes severe weathering conditions. The film's response to stresses (mainly due to thermal variations and weathering) evolves with time since the polymeric materials degrade under ultraviolet radiation, heat, and microorganism [12]. Due to their low cost and availability, polyethylene- (PE-) based monolayer films are quite popular in the greenhouse covering application. PE films with the thickness of about 140–200 μm are often characterized by a shelf life of as long as 2-3 years [1, 13].

The optical properties of chosen plastic films are vital as well, as they largely govern the crop yield. Solar radiation induces photosynthesis, which is essential for the plant growth and provides energy to the greenhouse system [14]. The spectral distribution of solar radiation reaching earth surface has about 40% of the total energy emitted in the near-infrared (NIR) radiation range (700–2500 nm) and about 39% in the photosynthetically active radiation (PAR) range (400–700 nm) [11, 15]. A provision that can allow the PARs to reach the crops and inhibit NIR radiations (i.e., 50% of global solar radiation) would be ideal for the agriculture production in arid regions. This can be accomplished by using a radiation-filtering greenhouse cover that can reflect the NIR radiations and transmit selectively the PARs to the crops.

PE-based films and sheets with several NIR-reflective additives have been prepared earlier and reported in the literature [16, 17]. These films are found to reduce the solar radiation across the whole solar spectrum including the PAR region. The PAR transmittance for these films was found to be in the range from 62% to 72%, while their NIR reflectance was in the range from 37% to 54% [16, 17]. However, the presence of NIR-reflective additives adversely affected the lifetime of covering films thus making them unsuitable for the greenhouse application. Verlodt and Verschaeren have reported the development of a novel NIR-reflective film having stronger reflection of NIR capacity combined with a higher PAR transmission [18]. The performance of this new film was evaluated against a standard PE film for its application as a greenhouse cover. Results showed that the PAR transmittance of the developed film was quite good (almost the same as that of the standard PE film). However, its efficiency to reflect NIR radiation was quite low (though 4.3% higher than that of the standard PE film). López-Marín et al. have also examined the effectiveness of a PE film having NIR-reflective pigment as a greenhouse cover [19]. Their results also showed that the use of NIR-reflective additive caused a shift in the whole spectrum to a lower transmission side resulting around 15–20% loss in the PAR transmission as compared to the standard PE cover. Impron et al. studied two PE films incorporating NIR-reflective pigments with varying concentrations [20]. The measured PAR transmittance for

TABLE 1: Important physical parameters of polymer resins used in this study.

Sr. no.	Sample	Density (gm/cc)	MFI (g/10 min)	Strength (MPa)	Elongation (%)
1	LLDPE 6821	0.921	0.80	13	730
2	EVA 218	0.941	1.7	39	500

such films was about 77–80%, while NIR reflectance was about 21–26%. Mutwiwa et al. studied the effects of coating the PE roof of a naturally ventilated greenhouse with NIR-reflective pigment on the greenhouse microclimate [21]. According to their results, the PAR transmittance of the coated film was estimated to be around 72%, which is quite encouraging. However, their NIR reflectance was estimated to be merely 22%. The coating of NIR-reflective pigment, however, has its own limitations to be practically used for greenhouse covering applications.

Based on studies reported in the literature, it can be stated that the development of PE films with NIR-reflecting properties for greenhouse covering application is still under the developmental stage. The objective of this project was to satisfy the need for a new generation of greenhouse covering materials with a good shelf life (2-3 years) that can reflect the NIR portion of the solar spectrum, yet allowing the PARs that are essential for plant growth to pass through. In this study, PE-based formulations using different NIR-reflective pigments were prepared by the blown film extrusion technique. These films were characterized thoroughly to investigate the influence of additives on the film processing and its mechanical properties. The radiometric properties of these films were also evaluated in terms of NIR reflectance and PAR transmittance to study the influence of these pigments on the spectral irradiative behavior of the film.

2. Materials and Methods

A film grade LLDPE resin was selected from a local polyolefin producing industry (SABIC, Saudi Arabia). Ethylene-vinyl acetate polymer resin used in this study was obtained from ExxonMobil Chemicals, USA (Escorene Ultra FL 00218). Some of the important physical parameters for these raw materials used in preparing blend formulations are listed in Table 1.

Various NIR-reflective additives used in the present work were supplied by Shepherd, Cincinnati, USA. Table 2 shows their compositions and other important properties such as total solar reflectance (TSR) and heat buildup (HBU). Total solar reflectance is the percentage of the total solar energy reflected by the pigment. Heat buildup is the temperature raised due to solar radiations being absorbed by the pigment and is measured according to ASTM D4803 method.

Commercially available UV additives such as antioxidants and UV stabilizers used in this study were obtained from their major suppliers, and their description is listed in Table 3. These additives were used in different concentrations and combinations.

TABLE 2: Composition and important properties of NIR-reflective additive used in this study.

Sample no.	Pigment code	Composition	TSR*	HBU*
NIR1	Blue 424	CoAl	42%	31°C
NIR2	Yellow 10P110	NiSbTi	69%	20°C
NIR3	Orange 10P225	CrSbTi	63%	22°C
NIR4	Green 223	CoNiZnTi	25%	35°C
NIR5	Brown 10P850	MnSbTi	35%	26°C

3. Experimental

3.1. Compounding. A blend of LLDPE 6821 (80%) and EVA 218 (20%) was prepared and used as a base resin (control sample) to prepare all other formulations reported in this study. Composition of control material and different blends prepared are listed in Table 4. The control resin (S1) was dry-blended with different additives (NIR reflective, UV stabilizer, antioxidants, etc.) in different ratios. The dry-blend was pelletized using an intermeshing and corotating twin screw extruder, Farrell FTX20 (screw dia 26 mm; l/d ratio 35). The screw has got both the dispersive and distributive mixing elements. The dry-blends were fed to the extruder that was operating at a screw speed of 12 rpm and an average processing temperature of 235°C. The melt pressure was about 7 bars. The extrudate was cooled in a water bath, dried, and pelletized for further use.

3.2. Blown Film Preparation. Thin PE films of various thicknesses (60–200 μm) were prepared by blown film extrusion techniques, using the above-mentioned formulations (Table 4). Films were drawn at 30°C through a ribbon die by a takeoff machine (model Shouman FPE200, Egypt) at a varying speed of 0.5–2 m/min in order to obtain the films of desired thickness. The temperature was controlled with a water-cooled thermostatic system connected with the takeoff machine.

3.3. Differential Scanning Calorimetry. Differential scanning calorimetry (Shimadzu DSC-60) was used for the thermal analysis of control and blend resins. Specimens of about 6–7 mg each were prepared by “shaving off” a thin layer (<1 mm thickness) of the pellets to minimize the thickness effect. The specimen was placed in an aluminum pan without being sealed then placed in the DSC oven in air at an atmospheric pressure. Samples were heated at 5°C/min from room temperature to 200°C, held there for 10 min, and then cooled back to room temperature at the same rate as of heating. The peak temperature of the melting curve was recorded as the melting temperature (T_m) of these materials. The onset crystallization temperature (T_c) was determined at the beginning of the crystallization peak (at the intersection of the peak slope with the baseline). The melting and crystallization enthalpies, ΔH_m and ΔH_c , were determined from the corresponding peak areas in the heating and cooling DSC scans, respectively.

3.4. Melt Flow Index. The melt flow rates of the samples were determined by using a Dynisco Polymer Test Melt Indexer,

USA, at $190 \pm 1.0^\circ\text{C}$ using a 2.16 kg load and a dwell time of 300 seconds. ASTM 1238 was used as the guideline for this procedure.

3.5. Viscoelastic Properties. The viscoelastic properties of control and blend resins were characterized using AR-G2 rheometer (TA instruments, USA). The samples were compression molded at 190°C under pressure with required diameter disks to fit the rheometer circular plates. The linear viscoelastic functions were measured using the parallel plate geometry (diameter of 25 mm and gap of 1000 micron). Frequency sweep was performed between 0.1 to 100 rad/s at 190°C. Strain sweeps were performed prior to ensure that the frequency sweep tests were done in the linear viscoelastic region. Additionally, time sweeps were also performed to make sure that the polymer is stable (e.g., no degradation is taking place during the length of measurement). The temperature was stable within 0.5°C over the range used in this study. All tests were performed in the atmospheric air.

3.6. Tensile Test. The tensile properties of the film specimens were measured according to ASTM D882 using a Hounsfield H100 KS series tensile testing machine. The samples were conditioned prior to testing at room temperature for 24 hours. The tensile tests were performed at a crosshead speed of 20 inch/min. The reported measurements for all of the above tests represent the median of three experiments.

3.7. Radiometric Properties. The spectral transmittance and reflectance of the samples were measured at normal incidence using Black-Comet (StellarNet Inc., USA) spectrophotometer, scanning between 200 and 1100 nm at 0.5 nm intervals in the UV-Vis range (200–750 nm) and at 1 nm intervals in the NIR range (750–1100 nm). The measured data were averaged at each 20 nm interval.

4. Results and Discussion

4.1. DSC Analysis. Figure 1 shows the dynamic thermogram (heating and cooling) of control sample (S1) and blend composites having NIR-reflective additives (S2 to S6). Figure 2 depicts the thermograms of control (S1) and the blend composites having mixture of various NIR and other UV additives (S1 and S7 to S9). The thermograms were shifted vertically for the ease of the presentation. The DSC data including melting and crystallization temperature (T_m and T_c , resp.) are summarized in Table 5.

The endothermic melting peaks of these samples appear to be quite similar. There was no significant change in the T_m of these formulations compared with the control resin (S1). Therefore, it can be said that the incorporation of these additives (NIR reflective, UV absorber, antioxidants, etc.) does not significantly affect the melting characteristics of the neat polymer blend. Likewise, the shapes of the exothermic crystallization peak for control and blend samples were also quite similar to each other. However, deviation was found in the T_c which is slightly higher for the blend samples than

TABLE 3: List of UV additives used in this study.

Sample no.	Functionality	Description
UVA1	UV stabilizer masterbatch	It is a concentrate of light stabilizers and thermal stabilizers in pellet form. It contains UV stabilizers and IR thermal stabilizers in polyethylene carrier.
UVA2	Antioxidant masterbatch	It is a concentrate of antioxidant agents in pellet form. It contains a combination of antioxidants in polyethylene carrier.
UVA3	UV stabilizer masterbatch	It is a UV stabilizer specifically developed for polyolefins. It is suitable for all applications using LDPE, LLDPE, and PP.

TABLE 4: List of the samples prepared and used this study.

Sample no.	Composition
S1	LLDPE 6821 (80%) + EVA 218 (20%)
S2	S1 (99.5%) + NIR1 (0.5%)
S3	S1 (99.5%) + NIR2 (0.5%)
S4	S1 (99.5%) + NIR3 (0.5%)
S5	S1 (99.5%) + NIR4 (0.5%)
S6	S1 (99.5%) + NIR5 (0.5%)
S7	S1 (98%) + UVA1 (2%)
S8	S3 (99.5%) + UVA2 (0.25%) + UVA3 (0.25%)
S9	S5 (99.5%) + UVA2 (0.25%) + UVA3 (0.25%)

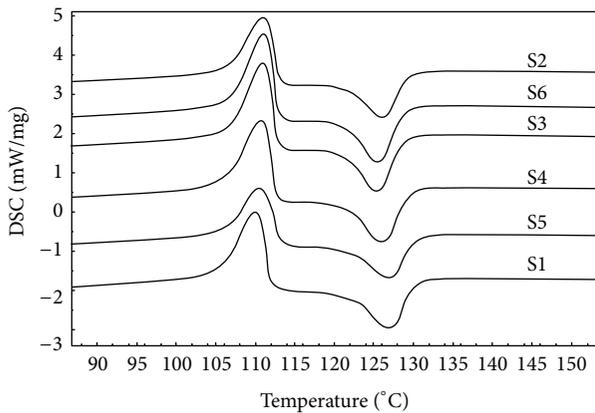


FIGURE 1: Heating and cooling thermograms of control and blend samples with NIR-reflective pigments.

control (S1). This finding might be attributed to the heterogeneous nucleating effect induced by the presence of additives (NIR-reflective pigments, UV absorber, antioxidants, etc.) in blend samples [22].

4.2. Tensile Properties. The tensile data for control and blend samples is shown in Table 6. Tensile strength of blend samples is usually lower than the control resin (except in the case of S2 and S7). This indicates that there is no orientation of the polymer chains as a result of introducing various NIR and UV additives in control resin, and, hence, the mechanical strength of blend samples is reduced [23]. Percent deformation at the break of blend samples is also shown to be decreased by the incorporation of these NIR and UV additives since they restrict the motion of the polymer chains. The additives might

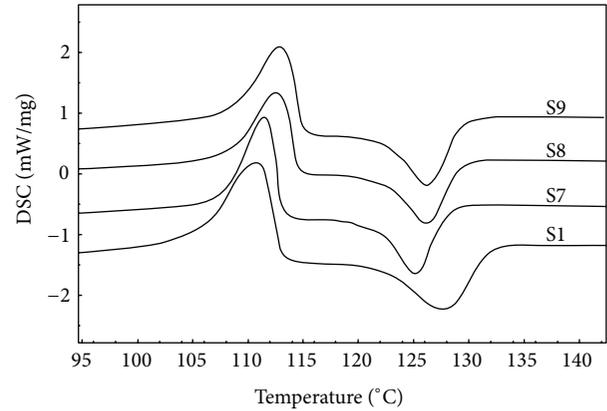


FIGURE 2: Heating and cooling thermograms of control and blend samples with NIR reflective and other UV additives.

TABLE 5: Thermodynamic properties of control resin and blend samples.

Sample	T_c (°C)	ΔH_c (J/g)	T_m (°C)	ΔH_m (J/g)
S1	109.82	46.17	127.54	33.40
S2	110.86	36.64	125.95	27.75
S3	110.82	45.42	125.13	33.87
S4	110.65	44.29	125.89	37.13
S5	110.53	35.67	126.83	28.62
S6	110.94	46.52	125.37	36.58
S7	111.50	28.35	125.36	27.72
S8	111.46	34.12	125.01	22.52
S9	112.82	31.16	126.19	24.08

reduce the flexibility by perturbing the chain motion of the control resin [24].

4.3. MFI and Rheological Properties. The MFI is a simple, easily obtainable viscosity parameter that indicates the physical properties and molecular structure of the polymer [25]. The effect of the incorporation of additives on the melt flow of control and blend samples is shown in Table 6. It is evident that there is no significant change in the melt flow index of these blend samples as compared to the control. This confirms that the blend formulations have similar flow characteristic and processing profile as the control resin [26].

The analysis of the viscoelasticity at lower frequency range can provide information about the microstructure of the final blend morphology that would be helpful to

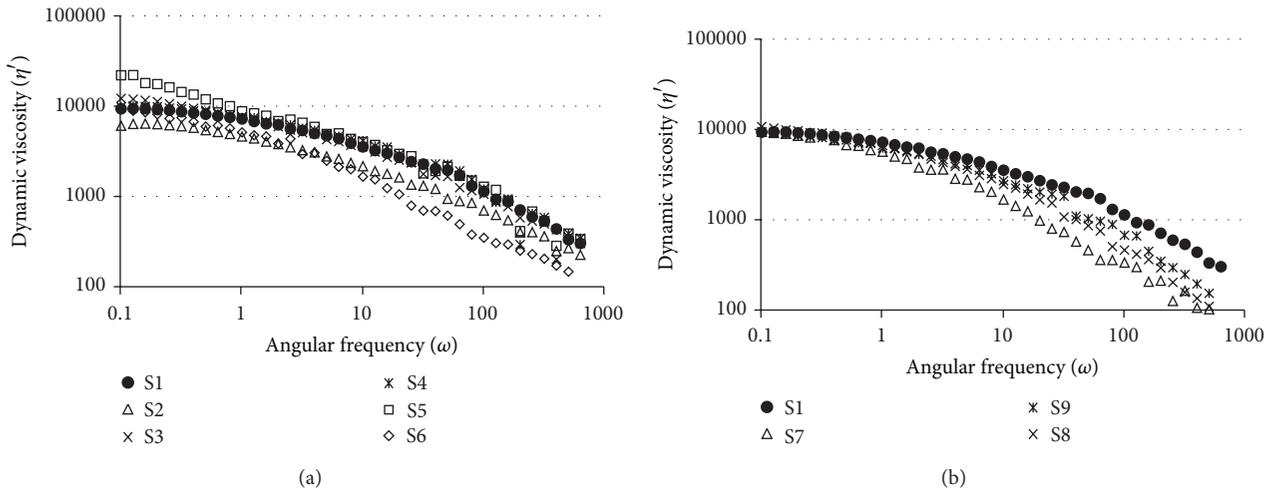


FIGURE 3: Frequency sweep rheogram for (a) control and blend samples with NIR-reflective pigments and (b) control with NIR reflective and other UV additives.

TABLE 6: MFI and tensile data for control and various blend samples.

Sample no.	MFI (g/10 min)	Tensile strength (MPa)	Elongation (%)
S1	0.824	13.11	870
S2	0.862	14.75	716
S3	0.801	12.09	852
S4	0.812	11.25	690
S5	0.823	12.75	836
S6	0.781	12.74	600
S7	0.821	14.09	644
S8	0.790	12.01	606
S9	0.836	12.71	620

evaluate the interfacial interaction between the different phases of polymer blends. At low frequency, the effect of the flow-induced molecular orientation on viscoelasticity becomes more or less negligible and is mainly governed by the polymeric microstructure rather than the flow-induced molecular artifacts [27]. Frequency sweep profiles (plot of the dynamic viscosity (η') versus the angular frequency (ω)) for the control sample (S1) and blends having NIR-reflective additives (S2 to S6) are shown in Figure 3(a). Figure 3(b) depicts the frequency sweep profiles for blend samples having NIR and other UV additives (S7 to S9).

The effect of oscillation frequency on the dynamic viscosity of these samples measured at 190°C shows that as the frequency increases, the viscosity decreases due to the increased number of polymeric chain entanglements being broken than the number of new entanglements being reformed [26–28]. The η' of these various blend samples is more or less similar to that of control which is an indicative of the fact that these blend samples are rheologically similar to the control one. However, it is interesting to note that loss in viscosity (η') with increased angular frequency (ω) is

more prominent for the samples having NIR and other UV additives.

The storage modulus (G') of control (S1) and blend resins with NIR-reflective additives (S2 to S6) are plotted against the loss modulus (G'') as shown in Figure 4(a), while similar plot for blends having NIR and other UV additives is shown in Figure 4(b).

It can be observed that the slopes of the curves for blend samples are slightly shifted towards the y -axis. It manifests that blend samples exhibit better elastic properties than control. This might be due to the nature of additives (NIR reflective, UV stabilizer, antioxidants, etc.) that are contributing to the elasticity of polymeric microstructure.

4.4. Radiometric Properties. The solar irradiative characteristics of the prepared films were measured at normal incidence in the wavelength range of 200–1100 nm and illustrated in Figures 5(a) and 5(b). Figure 5(a) shows the reflectance spectra for control (S1) and blend samples having NIR-reflective pigments (S2 to S6), while Figure 5(b) shows the same for blend samples containing NIR-reflective pigment and other UV stabilizers. The transmittance profile for these two sets of samples is shown in Figures 6(a) and 6(b), respectively. Average transmittance and reflectance values in various spectral regions (UV, PAR, and NIR) are summarized in Table 7.

From the results, it is evident that the blend film samples show an improvement in NIR reflectance as compared to the control sample S1. It is observed that S9 (having combination of NIR-reflective pigment and UV additives) shows the highest reflective capacity as compared to other blend samples. Those blend samples which contain additional UV stabilizers (S7 to S9) show a relatively lower transmittance values in UV region. The PAR transparency (an essential requirement for plant growth) is sufficiently high for control and most of blend samples (except S6 and S9). These results are an indicative of the right selection of the NIR-reflective pigments,

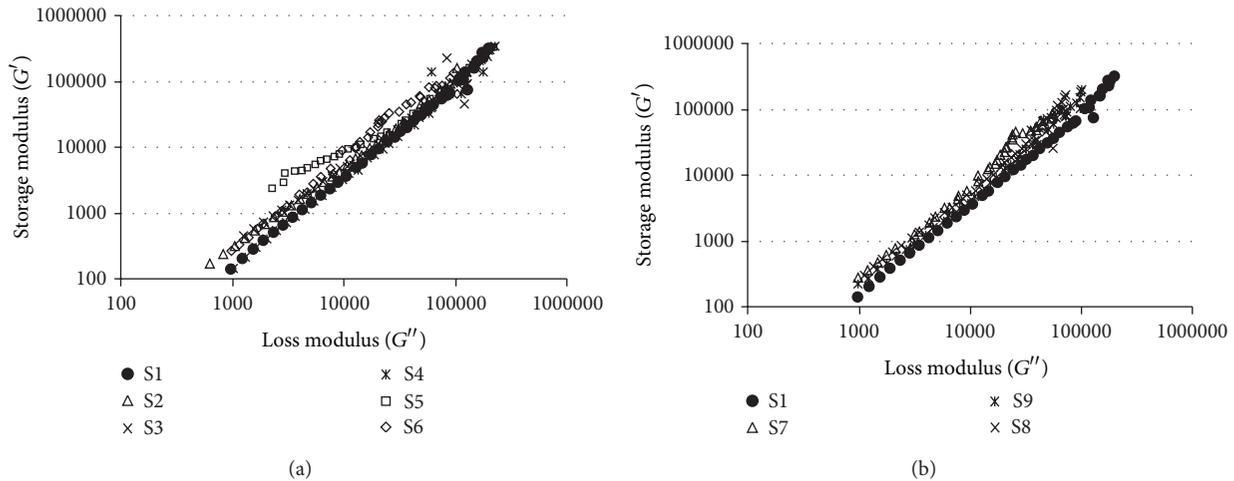


FIGURE 4: G' versus G'' curve for (a) control and blend samples with NIR-reflective pigments and (b) control with NIR reflective and other UV additives.

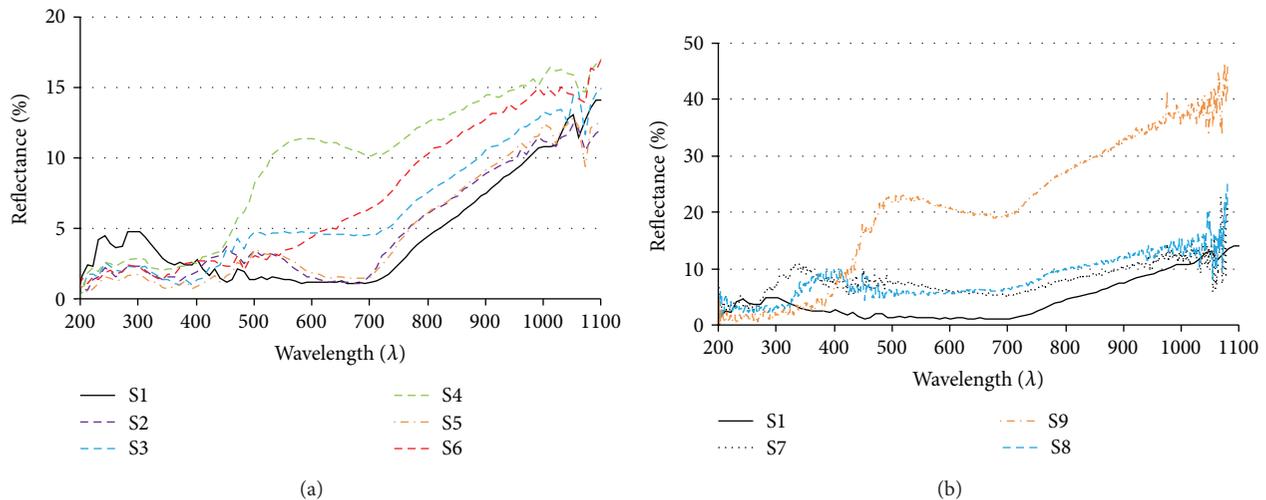


FIGURE 5: Spectral reflectance of (a) control and blend samples with NIR-reflective pigments and (b) control with NIR reflective and other UV additives.

and the formulations prepared thereof can be potentially tested for their use as a prototype greenhouse covering agent. However, these studies are still in the primitive stage since the reflectance properties of these films did not reach the ultimate goal. There is still a need to enhance the NIR reflectance of these formulations. Further work in this direction is currently under progress to improve the reflectance (by varying the loading ratio of NIR-reflective pigments, using different types of NIR-reflective pigments with higher TSR values, etc.).

5. Conclusion

A series of blend formulations has been prepared and tested for their suitability as a greenhouse covering film in this study. The polymer blends were successfully prepared by melt blending technique using several additives (NIR reflective, UV stabilizer, antioxidant, etc.), and their subsequent films

with specific thickness were prepared by blown film extrusion technique. The DSC analysis and MFI data revealed that the thermal and flow characteristics of new formulations having NIR and UV additives are not very different from control, which is a positive attribute for the cost-effective processing of these formulations. The rheological characterization of these blends showed that their viscoelastic properties are quite similar to that of the control. However, the incorporation of various additives in control resin resulted in increased elastic modulus. The measurement of mechanical properties showed that the tensile strength of blend samples was not greatly affected by the incorporation of these additives, and these films can withstand the aggressive environment during their use as greenhouse covering films in arid region. The radiometric properties were found to be improved for these formulations. However, there is a need to further improve the NIR reflectance of the current formulations. To achieve

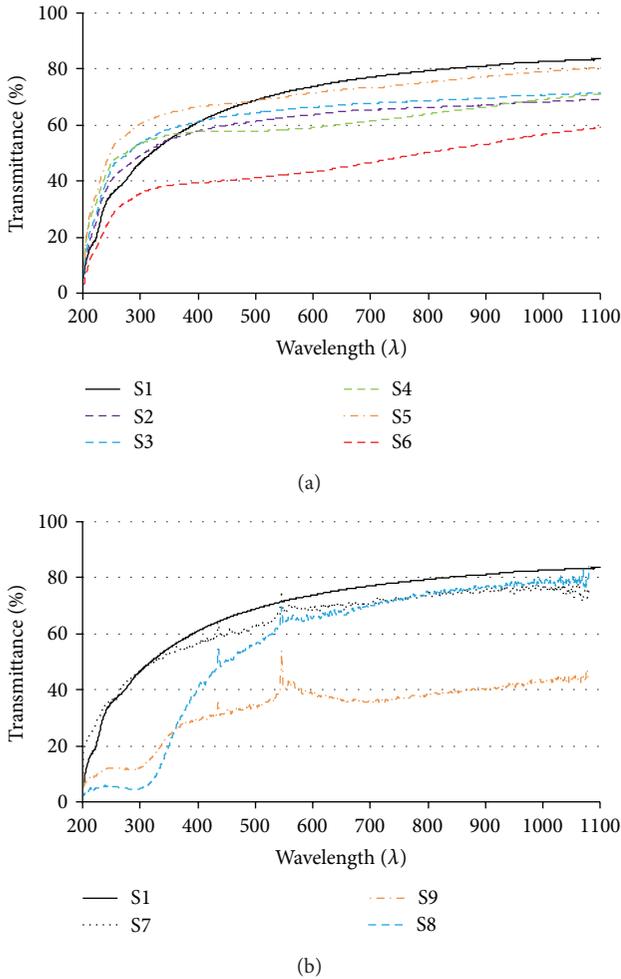


FIGURE 6: Spectral transmittance of (a) control and blend samples with NIR-reflective pigments and (b) control with NIR reflective and other UV additives.

TABLE 7: Average values in percentage for transmittance and reflectance in UV, PAR, and NIR regions for control and various blend samples.

Sample	T (%) UV	T (%) PAR	T (%) NIR	R (%) NIR
S1	41.2	70.8	81.0	7.6
S2	43.1	62.4	67.3	8.3
S3	46.5	65.3	69.7	10.1
S4	46.7	58.8	66.5	13.9
S5	52.6	70.1	77.2	8.5
S6	42.2	42.5	53.3	12.2
S7	29.9	65.7	74.6	9.7
S8	12.4	60.1	75.9	11.6
S9	16.0	35.8	40.3	31.5

this, a series of experiments are going on to improve the NIR reflectance by optimizing the percentage of loading and the type of NIR-reflective pigments used in the formulation.

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