

# **Research Article**

# **Development of Multifunctional Film for Greenhouse Applications in Tropical Regions**

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Single-purpose greenhouse films such as UV-blocking, NIR-blocking, or ultrathermic films are commonly developed in various climate regions. However, multifunctional films of combined functions are rarely explored, especially in the tropical regions. In this research, a multifunctional film having high UV filtration, high NIR reflection, and good light diffusion was developed for a greenhouse cover application in tropical regions. Effects of type, quantity, and particle size of additives on optical properties (280–2500 nm) and mechanical properties of 3-layer laminated films comprising 90% LLDPE/10% EVA polymer matrix and additives were studied. Results show that properties of those films are adjustable by varying types, particle size, and content of additives. The UV transmission of the film was ranged from 13.7 to 32.7 %T, NIR reflection from 12.1 to 19.8 %R, and %haze diffusion from 39.5 to 72.3 where photosynthetically active radiation (PAR) transmission was in the range of 62.6–78.9 %T. Those films exhibit tensile strength of 18–24 MPa, modulus of elasticity of 200–280 MPa, and elongation at break of 610–810%. A field test of the newly developed films as a cover for a greenhouse of 6 m wide ×24 m long ×4.3 m high with double roof design showed a better quality of plant growth in terms of weight, height, and bush width compared to a 7% UV absorber commercial film.

#### 1. Introduction

Greenhouse films of a single function such as ultrathermic, UV-blocking films have been extensively developed, especially in the cold climates. In tropical climate regions where the weather is described as hot, high humidity and heavy rain, a suitable greenhouse film for these climates has not been established. In tropical regions, not only the greenhouse structure should have a special roof design to reduce the air temperature inside the greenhouse but also the greenhouse cover should also provide a cooling effect. One of the most effective ways to reach the cooling effect within the greenhouse is to keep out the near infrared radiation, NIR (700-2500 nm), which is accountable for enormous heating effect inside the greenhouse. NIR radiation can be reduced by employing absorption, reflection, or interference pigment additives. Polyethylene- (PE-) based film with those additives has been reported earlier in the literatures. For instance, titanium dioxide pigment (TiO<sub>2</sub>) with different particle sizes and phases was frequently used as absorption/reflection

materials [1–4]. In addition, titanium dioxide in the rutile phase was reported to be capable of reflecting infrared radiation to a high degree [5]. In the recent year, WO patent no. 2015/052319 A1 disclosed the preparation of greenhouse cover containing commercial TiO<sub>2</sub> powder including Iriodin®SHR 9870, Altiris®, and Tipure R103-07 as NIR filtering materials. Results showed that the Altiris® (particle size of 0.04–2.28  $\mu$ m) provided the best properties as the NIR can be reflected out about 30% while the PAR transmission was kept at 70% [6]. Effect of cooling was also reported in TiO<sub>2</sub>-coated multilayer "Kool Lite/Astrolux" film developed by Hyplast/ Klerk's and Merck KGaA. It was revealed that Kool Lite Plus film could reduce the temperature inside the greenhouse by 4-5°C, and the PAR transmission was still comparable to that of the standard film [4].

Whitewash  $(Ca(OH)_2)$  solution was employed by Hemming et al. to reduce the NIR radiation [7]. It was found that the whitewash had adverse effect on PAR transmission more than that of the NIR-reflecting materials. This result agreed with Lopez-Martin et al. in which 24% and 15% of PAR reductions were found in the whiten cover and in the NIR-reflecting cover, respectively [8]. Nonetheless, the NIR transmission of the whitewash and the NIR-reflecting covers was reduced by 21.5% and 19.2%, respectively, and the inside temperature could be reduced up to 3°C. The PE greenhouse cover coated with Reduheat® pigment was developed by Merck Inc., and the university of Hannover was found to reduce the inside temperature by 4°C and reduced the NIR transmission (700-1500 nm) by 28% while the PAR transmission was reduced only by 17%. However, this coated cover could repeatedly wash out by rain [9]. Recently, the greenhouse cover using silica as an addition to serve as a basis for radiation cooling was reported [10]. It was found that, under 35°C ambient conditions, the inside temperature of the simulated greenhouse with the 1% SiO<sub>2</sub> double layer films was 3 to 5°C less than that of the greenhouse with the commercial agricultural polyethylene (PE) film.

Other NIR absorption/reflective materials that have been developed for greenhouse cover include black iron oxide, blue cobalt oxide, coated copper powder, lanthanum hexaboride, and metal compounds such as CoAl, NiSbTi, CrSbTi, CoNiZnTi, and MnSbTi. Those films could reduce the UV transmission to 42–53% and reflect NIR radiation by 8–14%. It was also found that, by adding 0.5% CrSbTi, the most reflection of NIR radiation could be obtained at about 14%. However, the PAR transmission was also reduced to 43% [11].

The light diffusion is another important property of the greenhouse cover aside from high PAR transmission, NIR reflection, and UV filtration. This property will evenly distribute sunlight within the greenhouse resulting in an enhancement of the effectiveness of plant photosynthesis and prevent the leaves burning [12-14]. Many studies had been conducted on the light diffusion of greenhouse cover [15-18]. The US patent no. US5585418 prepared multilayered greenhouse film having a variable light diffuse surface which was substantially clear with high light transmission when wetted but reduced light transmission when dry with a surface haze greater than 65% [19]. The WO patent no. 2007063240A1 disclosed the preparation of greenhouse covering film comprising natural fillers based on calcium carbonate, chosen from among chalks, calcites, or marbles as a diffusing additive [20].

Noticeably, the previous developments of the greenhouse cover were carried out in the direction of a single function or cooperative functions of either NIR reflection, UV filtration, or light diffusion and mostly for the coldclimate regions. However, the multifunctional greenhouse cover suitable for tropical climates is almost unexplored. Therefore, it is a challenge to develop a new generation of covering materials that have multiple functionalities including UV filtration, NIR reflection, and light diffusion in a single film. To achieve all these requirements, the multifunctional film with the combined additives of UV absorber and NIR-reflecting material was prepared in this study. Effects of type and amount of additives on mechanical properties and optical properties in terms of transmittance and reflectance in the wavelength of 280-2500 nm were evaluated. In addition, the effect of additive's location in

different layers and the amount of additive in the 3-layer laminated film on those properties were also examined. Last but not least, a practical implementation of those newly developed multifunctional films was field-tested as a greenhouse cover. Studies on plant growth and air temperature inside the greenhouse were compared to a commercial film.

## 2. Experimental

2.1. Materials. A film grade LLDPE pellet (MFI 0.85 g/ 10 min, density 0.92 g/cm<sup>3</sup>) was purchased from the Dow Chemical Company (USA). Ethylene-vinyl acetate polymer resin (EVA; 18%VA, MFI 2.3 g/10 min., density 0.94 g/cm<sup>3</sup>) was obtained from TPI Polene Public Co., Ltd. (Thailand). Solvent and reagents for surface modification of additives such as isopropyl alcohol and triethylamine (99.95% purity) were purchased from RCI Labscan (Thailand). Trimethoxy(propyl)silane (97% purity) was purchased from Sigma Aldrich (Singapore). Commercial additives were selected from the previous studies [4–7, 21] and were obtained from major suppliers, and their descriptions are listed in Table 1.

2.2. Modification of UV Absorption Additives. To increase the compatibility and prevent the aggregation of additives in the polymer matrix, which will result in the good mechanical properties and the effectiveness of the additive, surface of additives was firstly modified by organosilane as published elsewhere [22]. In brief, predetermined amount of UV1 or UV2 was dried in the oven at 120°C for 24 h before adding into a reactor that contained isopropanol under nitrogen atmosphere. Triethylamine and trimethoxy(propyl)silane were then added at a ratio of 1:3 whereas a weight ratio of triethylamine to TiO<sub>2</sub> was set at 1:36. The reaction was carried out at 80°C for 6 h. The final product was then separated by filtration and washed with isopropanol before drying in the oven at 100°C for overnight.

2.3. Compounding. LLDPE (90%) and EVA (10%) with various types and contents of additives were compounded in a twin-screw extruder (Labtech LTE-20-32, Labtech Engineering, Thailand), where the screw diameter was 20 mm and the L/D ratio was 32:1. The content of UV absorber and NIR reflective additives was varied from 0.1 to 1.5 wt.%. The blend temperature was set at 170–210°C with the screw speed of 100 rpm. The extruded polymer was cooled and cut into pellets by a pelletizer.

2.4. Fabricating. All compounds were blown into films using a single screw blown film extruder unit (Thermo Scientific HAAKE PolyLab, Germany) with die size of 34 mm inner diameter and 35 mm outer diameter and die gap of 0.5 mm. The screw speed was set at 60 rpm, and take off speed was controlled at 2.5 m/min with a blow up ratio at 2.77 and a draw down ratio of 6.02. The obtained pellet was melted at 170–210°C and blown into film with the thickness of 30  $\mu$ m.

TABLE 1: Composition and particle size of additives.

Additives	Compositions	Particle size (µm)	Suppliers
UV1	TiO <sub>2</sub> -78% anatase	0.02	Evonik Degussa GmbH, Germany
UV2	TiO <sub>2</sub> -100% rutile	0.23	Sigma Aldrich, USA
NIR1	Al flake	77	Sigma Aldrich, USA
NIR2	A1 flake	19	Aldoro, Brazil

The 3-layer laminated film was prepared by hot compressing 3 layers of  $30 \,\mu\text{m}$  films at  $140^{\circ}\text{C}$  for 6 min with the pressure of 1500 psi. Each layer of the film might contain individual or mixed additives of the UV absorber and NIR reflective material.

2.5. Characterizations. The chemical structure of the surface-modified additive was characterized by nuclear magnetic resonance (solid-state <sup>13</sup>C-NMR, Bruker DRX400, Germany) and FT-IR/FT-Raman (Perkin Elmer System 2000) spectroscopies. Polarity test of modified additives was observed in mixed solvent of DI water and hexane. Thermal properties of compounds and films were measured by differential scanning calorimeter (DSC) on a DSC822<sup>e</sup> Mettler Toledo (Switzerland). The samples were scanned twice from 0 to 200°C with a heating rate of 20.0°C/min. Mechanical properties of polymer films were measured on an Instron tensile testing machine (Model 55R4502, USA), according to ASTM D882-2012, with a crosshead speed of 500 mm/min and load cell of 100 N, using rectangle specimens with 50 mm gauge length and 10 mm width. Dispersion of additives in polymer matrix was characterized by a scanning electron microscope (FE-SEM with EDS; Hitashi SU5000). Optical properties including transmission and reflection in the range of 200-2500 nm were examined on UV-VIS/NIR spectrophotometer (Lambda 950 by PerkinElmer) according to the JIS R 3106 standard. The UV and PAR transmissions were reported as average values in the range of 280-400 nm and 400-700 nm, respectively. However, the NIR reflection data were obtained from the average value in the range of 700–2500 nm. Haze diffusion of film was obtained through the measurement of the %haze on haze meter (BYK Gardner, model 107821) according to ASTM D-1003. The % haze was acquired via the light transmission of the film at wide angle (>2.5°), as described in equation (1). The "good light diffusion" was defined when more than 30% haze was attained [23]:

% haze = 
$$\frac{T_{\rm dif}}{T_{\rm t}}$$
, (1)

where  $T_{\text{dif}}$  is the light transmission of the sample at wild angle (>2.5°) and  $T_{\text{t}}$  is a total light transmission of the sample.

2.6. Field Test. The newly developed films of two selected formulas were industrial blown into three layers of  $120 \,\mu\text{m}$  total thickness for a field test. The greenhouse of 6 m wide  $\times 24 \,\text{m} \log \times 4.3 \,\text{m}$  high with double roof design was covered with the obtained films compared with a 7% UV absorber

commercialized greenhouse film. The greenhouse was located in the north eastern part of Thailand, and the experiment was carried out from June 2017 to April 2018 (rainy season to summer). The microclimate data inside the greenhouses such as air temperature, humidity, and light intensity were monitored. The empirical statistical data of plant growth were collected and analyzed using randomized complete block design (RCBD).

# 3. Results and Discussion

3.1. Modification of UV-Absorption Additives. The structural compositions of commercial additives UV1 and UV2 were characterized by XRD before performing surface modification (data not shown here). It was found that composition of UV1 consisted of 2 phases: anatase (78%) and rutile (22%) while UV2 showed only rutile phase. Figure 1 shows the chemical structure of unmodified and modified UV1 additives, which was examined by FTIR. Both additives showed characteristic peak at 3407 cm<sup>-1</sup>, which associated with O-H stretching whereas peak at 1633 cm<sup>-1</sup> represented bending vibration of O-H, indicating the water as a moisture [24]. The intensity of both peaks was decreased after modification. The additional peaks at 2875 cm<sup>-1</sup> and 2960 cm<sup>-1</sup> corresponding to asymmetric and symmetric C-H stretching of methylene segments, respectively, were observed in the modified UV1. The presence of those bands confirmed that the organosilane was covalently attached to the additive surface [25]. The new peak at 1216 cm<sup>-1</sup> was assigned to Si-C stretching [26].

The chemical structure of the modified UV1 additive was confirmed by <sup>13</sup>C-NMR where the chemical shift at 16.4 ppm, associated with carbon atom that connected to silicon atom (-O-Si-C-) [26], was observed, as shown in Figure 2. This implied the occurrence of the reaction between titanium dioxide and silane coupling agent. It should be noted that the observed FTIR and <sup>13</sup>C NMR spectra of the modified UV2 (not shown here) were resembled to that of the modified UV1. The UV1 additive after modification was found to locate more in the hexane layer (upper layer) than in the DI water layer (lower layer) implying the increase of the hydrophobicity of the modified additive, as seen in Figures 3(a) and 3(b). Same result was observed in the modification of UV2 additive (Figures 3(c) and 3(d)).

3.2. Thermal Properties of Polymer Films. Thermal properties of polymer films were measured using DSC and are summarized in Table 2. Melting  $(T_m)$  and crystallization  $(T_c)$  temperatures of all polymer films before and after compounding were almost the same, implying that additives had no effect on  $T_m$  and  $T_c$  of the polymer. However, enthalpy of melting  $(\Delta H_m)$  of all additive films was decreased, indicating the decrease in crystallinity probably because those additive particles obstructed molecular chain movement during the crystallization process. This result agreed with the previous study in the preparation of PE/TiO<sub>2</sub> nanocomposites, where TiO<sub>2</sub>

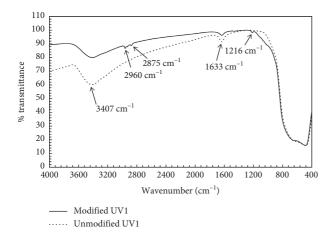


FIGURE 1: FTIR spectra of unmodified and modified UV1.

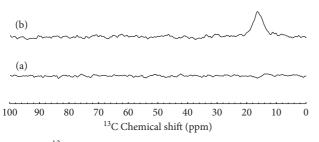


FIGURE 2: <sup>13</sup>C-NMR spectra of (a) unmodified UV1 and (b) modified UV1.

nanoparticle affected the crystalline size and the degree of crystallinity [27, 28].

3.3. Scanning Electron Microscopy (SEM). The dispersion of additives in the polymer matrix was illustrated by SEM. It was found that the dispersion of the UV1 and UV2 additive particles was quite uniform, as seen in Figures 4(a) and 4(b). However, the distribution of the NIR1 and NIR2 platelets was randomly dispersed due to relatively large size and low amount of the additive (Figures 4(c) and 4(d)).

3.4. Optical Properties of Polymer Films. Effects of additives on optical properties of the polymer film were studied by varying type and content of additives in polymer compounds. The additives in this study could be added individually or cooperatively. The compound was fabricated into blown films of monolayer ( $30 \,\mu$ m) and compressed into 3-layer laminated film ( $90 \,\mu$ m) as following details.

3.4.1. Monolayer Films. By adding 1.0 wt.% of various additives in S1–S4 (Table 3 and Figure 5), different effects on light transmission and NIR reflection were observed when compared with 90%LLDPE/10%EVA polymer matrix film (B). UV1 additive had a great effect on UV absorption with slight reduction in PAR radiation while UV2 greatly affected both UV as well as PAR radiations. NIR reflection of the UV2 additive film was slightly increased from 8.3%R to 13.2%R compared to the polymer matrix film (B). The most

prominent property of UV2 additive was the increase of % haze diffusion of the film up to 73%. On the other hand, the significantly increase of NIR reflections from 8.3%R to 31.2% R and 34.8% R was observed in NIR1 and NIR2 additive films, respectively. As a result, by combining the UV absorber and NIR reflective material, the optimization of the UV filter, NIR reflection, and light diffusion of the film was achieved. For the combination of UV1 and UV2 (S5-S8), although the reduction of UV radiation and the %haze diffusion were significantly improved, the PAR transmission was also notably dropped except for S5, where the total amount of both additives was lower (less than 1.5 wt.%). It was found that the combination of UV1 and NIR1 (S9-S11) gave the most promising results as the total amount of both additives was kept below 1.25 wt.% where the PAR transmission was maintained at more than 70%T, the %haze diffusion at more than 40, the UV transmission at less than 30%T, and the NIR reflection at more than 13%R. Those numbers are more or less comparable to that of the whitewash and Reduheat® films, where the 30%T UV transmission, 28%R NIR reflection, and 59% PAR transmission were reported [4, 9, 29]. An increase amount of UV1 from 0.5 wt.% to 1.0 wt.% resulted in an increase of UV filtering by 18-21% whereas an increase amount of the NIR1 from 0.25 wt.% to 0.5 wt.% resulted in an increase of NIR reflection by 21-25% which also accompanied by the reduction of the PAR transmission by 5-6%.

3.4.2. Three-Layer Laminated Films. Multilayer coextrusion films have been widely used for the commercialized greenhouse cover due to their durability and high strength [30]. In addition, additives can be selectively added in each layer resulting in the optimal dosage and function of the additives. Thus, 3-layer laminated film was also investigated in this study. To study effects of the additive's location in different layers and content of additive on mechanical and optical properties, the 3-layer laminated films (90  $\mu$ m) were prepared by hot compression of three monolayer films having  $30 \,\mu m$ thickness. Each layer contained different additives, either UV1, NIR1, or NIR2, in different locations (outer, middle, or inner layer). Table 4 and Figure 6 show light transmission, light reflection, and %haze diffusion of the prepared 3-layer laminated films with different types and amounts of additives in different layers. It was found that the optimum concentration of each additive (UV1, NIR1, and NIR2) was still in the range of 0.3-1.0 wt.%, as observed in the monolayer film. An increasing amount of UV1 (M2-M3) resulted in a decrease of UV transmission and slightly increased NIR reflection. However, an increasing amount of NIR1 from 0.3 wt.% to 0.5 wt.% (M6 vs. M2 and M7 vs. M3) led to an increase of the NIR reflection while PAR transmission was still sufficiently high. At the same amount of NIR1 additive, the increase of the NIR reflection might be additionally contributed from the increased amount of UV1 (M2 vs. M3 and M6 vs. M7). The switching between the outer and the middle layer of the additive in M2 and M4 had no effect on the transmission and reflection of the laminated film. This implied that each additive could perform their functionality regardless of the position of outer, middle, inner layer. For NIR2 additive

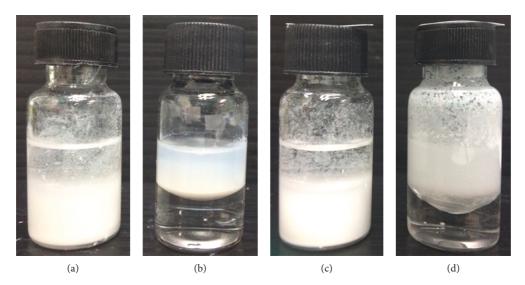


FIGURE 3: Dispersion of additives in mixed solvents of DI water and hexane after 1 hour of (a) unmodified UV1, (b) modified UV1, (c) unmodified UV2, and (d) modified UV2.

TABLE 2: Thermal properties of polymer films.

Samples	$T_{\rm m}$ (°C) (2nd scan)	$\Delta H_{\rm m}~({\rm J/g})$	$T_{\rm c}(^{\circ}{\rm C})$ (1st scan)	$\Delta H_{\rm c}$ (J/g)
LLDPE	121.0	111.4	107.3	-113.8
90%LLDPE/10%EVA	120.0	112.7	106.7	-112.2
90%LLDPE/10%EVA/1%UV1	121.0	109.1	107.0	-108.5
90%LLDPE/10%EVA/1%UV2	122.7	100.1	104.7	-94.1
90%LLDPE/10%EVA/1%NIR1	122.3	95.6	105.0	-99.1
90%LLDPE/10%EVA/1%NIR2	119.0	110.8	106.0	-110.3

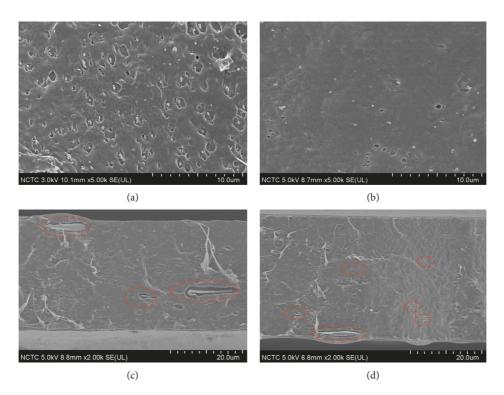


FIGURE 4: SEM images of (a) 90%LLDPE/10%EVA/1%UV1, (b) 90%LLDPE/10%EVA/1%UV2, (c) 90%LLDPE/10%EVA/1%NIR1, and (d) 90%LLDPE/10%EVA/1%NIR2.

Sample no.	Compositions	UV (%T) 280-400 (nm)	PAR (%T) 400–700 (nm)	NIR (%R) 700–2500 (nm)	Haze (%H)
Commercial GH film	LDPE + 7% UV absorber	$30.4 \pm 2.3$	$89.2 \pm 0.3$	$8.9 \pm 1.3$	$29.1 \pm 2.2$
В	90% LLDPE + 10% EVA	$90.3 \pm 0.8$	$91.0 \pm 0.0$	$8.3 \pm 0.1$	$22.6\pm0.1$
S1	B+1.0% UV1	$28.4 \pm 1.5$	$80.0 \pm 0.5$	$10.1 \pm 0.3$	$54.3 \pm 0.1$
S2	B+1.0% UV2	$44.6\pm0.7$	$69.5\pm0.4$	$13.2 \pm 0.1$	$72.9 \pm 1.6$
S3	B+1.0% NIR1	$63.3 \pm 1.6$	$77.5 \pm 0.8$	$31.2 \pm 0.5$	$28.5 \pm 0.2$
S4	B+1.0% NIR2	$45.1 \pm 1.6$	$37.7 \pm 1.9$	$34.8 \pm 0.5$	$51.4 \pm 1.3$
S5	B+0.5% UV1+0.5% UV2	$29.0 \pm 0.4$	$74.3 \pm 0.1$	$11.8 \pm 0.0$	$58.7 \pm 0.1$
S6	B+1.0% UV1+0.5% UV2	$16.0 \pm 1.1$	$68.1 \pm 1.0$	$12.0 \pm 0.2$	$68.5 \pm 0.3$
S7	B+1.0% UV1+1.0% UV2	$13.6 \pm 0.2$	$63.6 \pm 0.4$	$14.4 \pm 0.2$	$78.9 \pm 0.3$
S8	B+1.5% UV1+1.5% UV2	$9.0 \pm 0.2$	$57.9 \pm 0.2$	$30.5 \pm 0.1$	$86.4 \pm 0.5$
S9	B+0.5% UV1+0.25% NIR1	$36.3 \pm 1.3$	$77.3 \pm 0.5$	$13.0 \pm 0.0$	$41.2 \pm 0.1$
S10	B+0.5% UV1+0.5% NIR1	$34.6 \pm 0.3$	$72.6 \pm 0.1$	$16.3 \pm 0.3$	$46.8 \pm 0.1$
S11	B+1.0% UV1+0.25% NIR1	$22.8 \pm 1.3$	$71.2 \pm 0.9$	$13.2 \pm 0.5$	$44.6 \pm 0.0$
S12	B+1.0% UV1+0.5% NIR1	$23.1\pm0.3$	$67.9\pm0.4$	$16.0\pm0.4$	$50.3\pm0.1$

TABLE 3: Optical properties of monolayer films and polymer matrix film (B).

which had a smaller size than that of NIR1, results showed that the smaller size had more impact on the NIR reflection than the larger one (M5 vs. M2 and M8 vs. M6). This might be possible because that size was a suitable size for reflection of the radiation in the NIR range (700–2500 nm) [20].

PAR transmission of a greenhouse film is a significant parameter and should be sufficiently high for plant photosynthesis (usually more than 70 %T). Therefore, this factor was taken into consideration for the newly developed film. In our case, by adding UV absorber more than 0.5 wt.% caused the reduction of PAR transmission to less than 70% except for M5 where the contribution from NIR2 was prominent. Therefore, the amount of the UV absorber was kept at less than 0.5 wt.%. For the UV blocking, NIR reflection, and %haze diffusion, the higher percentage would be more desirable. Accordingly, a high potential film for a greenhouse application could be either M2, M4, or M8. Since M8 formula showed better haze diffusion where an even light was anticipated, and M8 film was then chosen as the most promising formula for greenhouse production for a field test. These film formulas showed better optical properties than that of the previous NIR reflective films, which could reduce the UV transmission to 42-53%, reflect NIR radiation at most 14%, but the PAR transmission was significantly reduced to 43% by adding 0.5% of the NIR reflective material [11].

3.5. Mechanical Properties of 3-Layer Laminated Films. Mechanical properties of monolayer and 3-layer laminated films were evaluated through tensile strength, modulus of elasticity, and elongation at break and are summarized in Table 5. The tensile strength of 3-layer laminated films was lower than that of the monolayer films whereas the tensile modulus was higher. These unexpected results might be affected from the preparation techniques. It was found that all 3-layer laminated films showed lower tensile strength, elongation, and toughness than that of laminated 90% LLDPE/10% EVA polymer matrix film. Nonetheless, these mechanical properties of 3-layer laminated film were comparable to a general purpose LDPE greenhouse film

where the tensile strength, tensile modulus, and elongation at break were in the range of 11–38 MPa, 113–230 MPa, and 300–600%, respectively [31, 32].

3.6. Applications. To demonstrate the value of the newly developed multifunctional film in greenhouse applications, two formulas of the greenhouse films (T2 and T3 in Tables 6 and 7) were industrial blown into 3-layer films for a field test. Mechanical properties of the industrial blown film, T2 and T3 compared with the 7% UV commercial blown film, are summarized in Table 6. Although the thickness of the newly developed film was lower than that of the commercial film, the tensile strength is higher whereas other properties were comparable. Table 7 summarizes optical properties of the newly developed industrial blown films (T2 and T3) compared with the commercial greenhouse film (T1). T2 greenhouse film had UV filtration and light diffusion effects whereas T3 possessed three functions of UV filtration, light diffusion, and NIR reflection. Figure 7 shows the SEM micrographs (Figures 7(a)-7(d)) and the SEM-EDS Ti-elemental distribution maps (Figures 7(e) and 7(f)) of T2 and T3 greenhouse films. The distribution of additive particles in SEM images, especially in the middle layer, was quite uniform regardless the small amount of additives. This uniform distribution of Ti particles (red dots) in the middle layer was also confirmed by the SEM-EDS elemental distribution maps.

The greenhouse of 6 m wide  $\times 24$  m long  $\times 4.3$  m high with double roof design was covered with those developed films compared with a 7% UV absorber commercialized greenhouse film. The greenhouse was located in the north eastern part of Thailand, and the experiment was done during June 2017 to April 2018 (rainy season to summer). Data of the microclimates inside the greenhouses such as air temperature, relative humidity, and light intensity (not shown) as well as empirical statistical data of plant growth were monitored. The preliminary results showed that the maximum air temperature inside the T3 was lower than that of T1 by 1–3°C (Table 8), probably due to NIR reflective effect of the multifunctional film that reflects some of the NIR

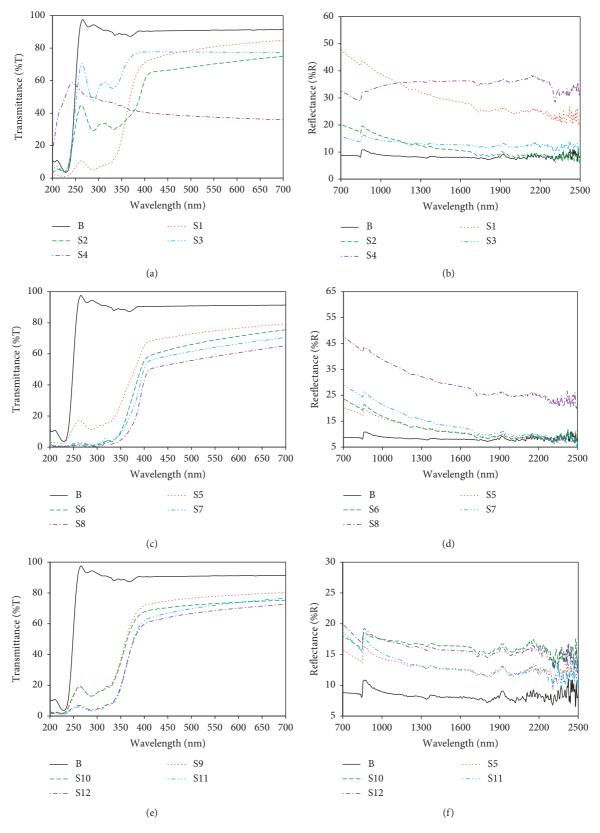


FIGURE 5: Light transmission and reflection of polymer films in the range of 200–2500 nm compared with polymer matrix film (B) of (a) light transmission of S1–S4, (b) light reflection of S1–S4, (c) light transmission of S5–S8, (d) light reflection of S5–S8, (e) light transmission of S9–S12, and (f) light reflection of S9–S12.

	TABLE 4. Optical properties of 5-layer laminated mins.						
Sample no.	Compositions (outer/middle/inner)	UV (%T) 280–400 (nm)	PAR (%T) 400–700 (nm)	NIR (%R) 700–2500 (nm)	Haze (%H)		
S10	B+0.5% NIR1+0.5% UV1	$34.6 \pm 0.3$	$72.6 \pm 0.1$	$16.3 \pm 0.3$	$46.8 \pm 0.1$		
M1	B/B/B	$46.8 \pm 1.6$	$88.8\pm0.1$	$8.5 \pm 0.1$	$39.5\pm0.1$		
M2	0.5% NIR1/0.5% UV1/B	$28.7\pm2.4$	$74.6 \pm 1.5$	$14.5 \pm 0.4$	$58.2 \pm 0.1$		
M3	0.5% NIR1/1.0% UV1/B	$13.7 \pm 2.1$	$62.6 \pm 2.4$	$16.2 \pm 1.9$	$72.3\pm0.8$		
M4	0.5% UV1/0.5% NIR1/B	$29.9 \pm 2.2$	$75.2 \pm 1.4$	$14.3\pm0.8$	$55.9\pm0.4$		
M5	0.5% NIR2/0.5% UV1/B	$27.3 \pm 2.4$	$68.6 \pm 1.9$	$19.8\pm0.8$	$43.3\pm0.5$		
M6	0.3% NIR1/0.5% UV1/B	$32.7 \pm 3.0$	$78.9 \pm 1.3$	$12.1 \pm 0.4$	$58.1 \pm 0.1$		
M7	0.3% NIR1/1.0% UV1/B	$13.7 \pm 1.3$	$66.9 \pm 3.9$	$14.0 \pm 2.8$	$70.5 \pm 0.7$		
M8	0.3% NIR2/0.5% UV1/B	$30.6 \pm 2.5$	$73.8 \pm 2.3$	$17.5 \pm 0.8$	$42.6\pm0.1$		

TABLE 4: Optical properties of 3-layer laminated films.

B = 90%LLDPE + 10% EVA.

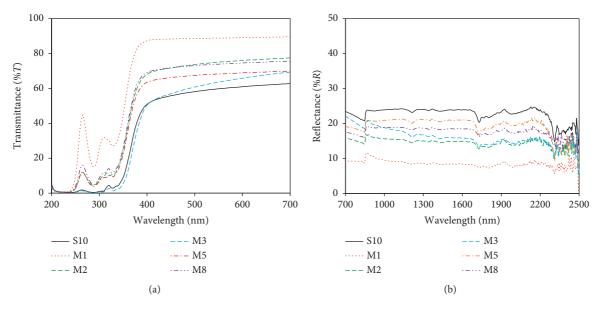


FIGURE 6: Light transmission and reflection of 3-layered laminated films compared to the monolayer film (S10) in the range of 200–2500 nm of (a) light transmission of M1–M8 and (b) light reflection of M1–M8.

TABLE 5: Mechanical properties of monolayer films (S9-S10) and 3-layer laminated films (M1-M8).

Sample no.	Compositions (outer/middle/inner)	Tensile strength (MPa)	Modulus (MPa)	Elongation at break (%)	Toughness (mJ/mm <sup>3</sup> )
S9	B+0.5% UV1+0.25% NIR1	$30.4 \pm 1.0$	$126.2 \pm 5.4$	$726.1 \pm 56.6$	$116.9 \pm 11.7$
S10	B + 0.5% NIR1 + 0.5% UV1	$32.1 \pm 0.6$	$175.5 \pm 18.6$	$850.5 \pm 16.2$	$134.0\pm4.0$
M1	B/B/B	$25.0 \pm 1.5$	$298.7 \pm 18.0$	$716.3 \pm 11.7$	$99.3 \pm 5.3$
M2	0.5% NIR1/0.5% UV1/B	$18.6 \pm 2.0$	$207.4\pm20.1$	$647.6 \pm 49.6$	$77.4 \pm 8.4$
M3	0.5% NIR1/1.0% UV1/B	$23.3 \pm 3.4$	$234.5\pm24.0$	$806.6 \pm 63.1$	$104.7\pm13.5$
M4	0.5% UV1/0.5% NIR1/B	$21.4 \pm 0.7$	$257.9 \pm 16.1$	$671.4 \pm 17.1$	$84.8\pm2.5$
M5	0.5% NIR2/0.5% UV1/B	$23.1 \pm 1.3$	$232.1 \pm 14.4$	$739.1 \pm 7.6$	$93.4\pm4.0$
M6	0.3% NIR1/0.5% UV1/B	$17.8 \pm 1.6$	$262.1\pm25.1$	$610.9 \pm 32.6$	$70.9 \pm 6.2$
M7	0.3% NIR1/1.0% UV1/B	$20.0 \pm 2.8$	$278.9 \pm 11.0$	$647.8 \pm 39.1$	$80.5\pm8.1$
M8	0.3% NIR2/0.5% UV1/B	$24.1 \pm 1.9$	$242.6 \pm 18.6$	$747.4 \pm 42.4$	$98.1\pm8.5$

B = 90%LLDPE + 10% EVA.

radiation out resulting in the lowering of air temperature inside the greenhouse. Comparing to the previous studies of greenhouse film containing Reduheat® pigment where the inside temperature was reduced by 4°C [4], although the decrease of the air temperature inside the T3 greenhouse was lower, the reduction of the PAR transmission was almost the same. The empirical statistical data of plant growth were collected and analyzed using randomized complete block design (RCBD). Experimental data (Table 9) were obtained from the observation of Chinese cabbage growing under the newly developed films (T2 and T3) compared to that of the commercial greenhouse cover (T1). These Chinese cabbages were grown under all treatments for 25 days during June

GH	Compositions	Thickness (µm)	Tensile strength (MPa)	Modulus (MPa)	Elongation at break (%)	Toughness (mJ/mm <sup>3</sup> )
T1	7% UV commercial	170	$27.2\pm0.9$	$311.2\pm10.1$	$1236.2 \pm 51.3$	$186.4 \pm 12.3$
T2	B/0.5 UV1/B	120	$34.4\pm1.9$	$308.8 \pm 5.5$	$1107.2\pm28.8$	$176.9 \pm 10.3$
Т3	0.3%NIR2/0.5%UV1/ B	120	$35.9 \pm 1.6$	$306.6 \pm 4.4$	$1039.3 \pm 24.8$	$181.0\pm9.9$

TABLE 6: Mechanical properties of field-tested greenhouse films.

TABLE 7: Optical properties of field-tested greenhouse films.

GH	Compositions	Thickness (µm)	UV (%T)	PAR (%T)	NIR (%R)	Haze (%H)
T1	7% UV commercial	170	$30.4 \pm 2.3$	$89.2 \pm 0.3$	$8.9 \pm 1.3$	$29.1 \pm 2.2$
T2	B/0.5 UV1/B	120	$46.0 \pm 1.5$	$83.5 \pm 0.3$	$9.6 \pm 0.0$	$65.6 \pm 0.8$
Т3	0.3%NIR2/0.5%UV1/B	120	$33.4\pm0.8$	$62.0\pm0.7$	$22.3\pm0.2$	$67.9\pm0.3$

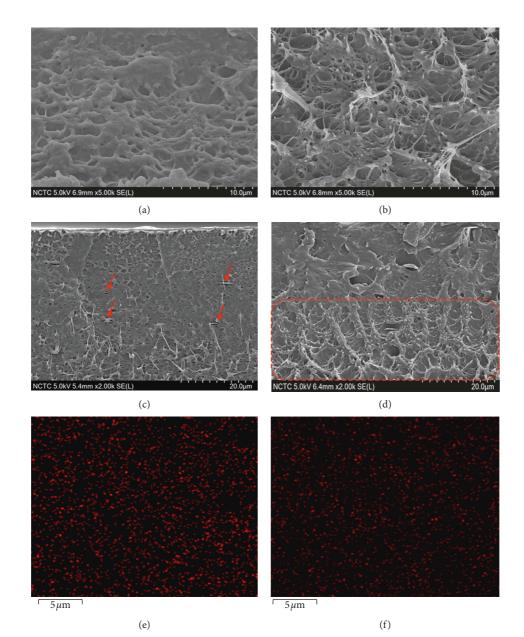


FIGURE 7: SEM images (a-d) and the SEM-EDS Ti-elemental distribution maps (e-f) of T2 and T3 greenhouse films: (a) T2 top layer, (b) T2 middle layer, (c) T3 top layer, (d) T3 middle layer, (e) T2 middle layer, and (f) T3 middle layer.

Date	Max. temperature outside (°C)	Max. temperature inside the greenhouse (°C)			
		T1	Τ2	Т3	
Sept. 2017	34.3	38.0	39.0	37.6	
Oct. 2017	28.4	30.4	31.0	30.2	
Nov. 2017	32.3	34.2	34.1	33.4	
Dec. 2017	30.2	32.8	32.5	30.4	
Jan. 2018	28.9	31.8	31.2	30.5	
Feb. 2018	35.5	38.8	37.7	37.2	
Mar. 2018	32.8	36.8	37.6	35.1	
Apr. 2018	31.9	37.6	37.2	35.4	

TABLE 8: Maximum air temperatures inside the greenhouses compared with the outside temperatures.

TABLE 9: Statistic data of Chinese cabbage growth in greenhouses covered with different films.

			Parameters				
GH		Leaves (no.)	Bush width (cm)	Weigh	Weight (g)		
	Height (cm)	Leaves (no.)	) Bush width (Cff)	Before trimming	After trimming		
T1	$15.4 \pm 3.0^{\circ}$	$13.7 \pm 1.0$	$16.7 \pm 1.6^{\circ}$	$42.0 \pm 14.9^{\circ}$	$35.5 \pm 14.6^{b}$		
Τ2	$18.9 \pm 1.0^{\mathrm{b}}$	$14.0 \pm 0.0$	$18.5 \pm 1.1^{\rm b}$	$65.8 \pm 16.5^{b}$	$55.6 \pm 16.2^{a}$		
Т3	$20.3\pm1.9^{\rm a}$	$14.0 \pm 0.0$	$20.5 \pm 1.2^{a}$	$81.6 \pm 28.7^{a}$	$66.7 \pm 27.6^{a}$		
Block	ns	ns	ns	ns	ns		
F-test	* *	ns	**	**	* *		
CV (%)	11.8	_	7.2	33.1	38.1		

\*, \*\*, and ns: significant at 5%, 1% probability levels, and nonsignificant. Values in each column expressed as means in each blocks  $\pm$  standard deviations (n = 4). Means followed by the same letter are not significantly different at the 0.05 level. CV (%) is defined as the standard deviation divided by the mean multiplied by 100 percent.

19th to July 29th, 2017. It was revealed that, on the harvest day, Chinese cabbage grown under different treatments showed different height, bush width, and weight at 1% probability level. The Chinese cabbages grown in the newly developed greenhouse film, especially T3, showed taller, larger, and heavier than those grown in the greenhouse covered with the commercial film (T1). This was probably because the multifunctional greenhouse film (T3) had proper light conditions including low UV and NIR transmission, high PAR transmission, and good light diffusion. Surprisingly, it was also observed that the growth of Chinese cabbages grown under T2 was better than those under the T1 despite the lower PAR transmission and higher UV transmission (Table 7). The only significant difference between T1 and T2 was the %haze diffusion. This implied that the light diffusion is a significant parameter on the plant growth. T2 might provide an even distribution of high light intensity inside the greenhouse.

# 4. Conclusions

We have demonstrated that the multifunctional film of UV filtration, NIR reflection, and light diffusion could be prepared by adding combined additives of UV absorber and NIR reflective material in LLDPE/EVA polymer matrix. Optimized quantities, of each additives, were in the range of 0.3–0.5 wt.%, for the optimum UV filtration, NIR reflection, and PAR transmission. Both monolayer and 3-layer laminated films were investigated. To be used as a greenhouse cover, a 3-layer laminated film with a formula of 0.3% NIR2/ 0.5%UV1/B is recommended. This film formula allowed 74% PAR transmission, 30% UV transmission, and 17.5% NIR reflection and showed 43% haze diffusion. The film exhibited tensile strength of 24 MPa, modulus of 243 MPa, and elongation at break of 747%. Preliminary results on the field test of industrial blown film of this formula showed that Chinese cabbages grew better and larger in a greenhouse covered with this newly developed film than those in the greenhouse covered with a commercial film.

# **Data Availability**

No data were used to support this study.

#### **Conflicts of Interest**

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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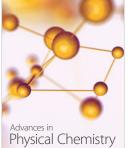
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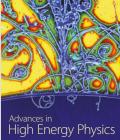
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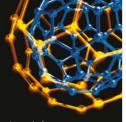
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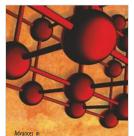




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