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Property Improvement of Three-layer LDPE Shrink Film Using LDPE Recycled from Blown Film Production Waste

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Abstract

Low-density polyethylene (LDPE) is one of the most used plastics especially in packaging applications. This research attempted to reduce the use of virgin LDPE and effectively recycle industrial LDPE waste by utilizing LDPE scraps from blown film production line (re-LDPE) for property improvement of three-layer LDPE shrink film prepared by blown film co-extrusion. While the skin layers consisted only of virgin LDPE, the middle layer was a blend of virgin LDPE and re-LDPE mixed using an extruder. The LDPE:re-LDPE ratios in wt% were varied from 100:0, 85:15, 70:30, 55:45, 40:60 to 0:100. It was found that several properties of all shrink films containing re-LDPE in their middle layers were better than those of the film solely made of virgin LDPE. Thermal analysis using DSC revealed an increase in crystallinity percentage of these films upon increasing re-LDPE content that consequently improved their tensile strengths in both machine and transverse directions despite a slight decrease in their shrink percentages but still within typical range for shrink film applications. In addition to the enhancement of heat-seal ability in both directions, lower gloss and haze values were also observed as re-LDPE content was increased. The results indicated that the film prepared using re-LDPE 100 wt% had highest tensile and seal strengths while the film prepared using re-LDPE 60 wt% exhibited lowest haze with similar gloss to the film prepared without re-LDPE. These results suggests a possibility of using re-LDPE to improve the properties of the multilayer films.

1. Introduction

Due to its technical and economic feasibility, thermo-mechanical recycling by reprocessing is a popular method utilized for recycling plastic waste based on thermoplastic polymers including low density polyethylene (LDPE), one of the most used plastics in the world [1,2]. As previously reported [2], its production combining with other types of polyethylene in 2021 was at 26.9% compared to those of other plastics such as polypropylene (19.3%), poly(vinyl chloride) (12.9%) and polystyrene (5.3%). To reduce LDPE waste generated from both production and consumption, such waste is reprocessed into pellets or flakes which are then used for manufacturing certain products [2-4]. However, this method often causes thermal degradation of a recycled polymer which results in the deterioration of its properties especially the mechanical properties [3,4]. Therefore, instead of using a recycled polymer solely in manufacturing process of a certain product, blending with a virgin polymer is usually implemented to improve the properties of such product [5-8]. It has been found that the presence of a recycled polymer in the blend can significantly, slightly, or not affect the properties of the final product depending on its amount in the blend, the type of virgin polymer and the type of the product [8-11].

LDPE shrink films are of interest for using a recycled polymer in their productions due to their high consumption in packaging applications and necessity to reduce the use of virgin LDPE and its waste. These films are designed to tightly wrap and seal a variety of products with different shapes and sizes both individual and multipack for protection during storage and transportation. They can be manufactured into monolayer and multilayer structures by blown film extrusion and co-extrusion, respectively [1,12]. A monolayer film is normally produced using a single screw extruder whereas a multilayer film is made from the process of feeding two or more polymer melts together into a single die to form a layered melt which is then processed into a single film having a multilayer structure. As found from industrial applications [13,14], if the films are made of the same polymer, several properties of a film with three-layer structure are better than those of a monolayer film.

Previous works revealed that the blends of recycled and virgin LDPEs were used instead of just recycled LDPE to achieve the films with desirable properties [8-9,15,16]. Addition of some additives [8,15] or additional processes [9,16] to enhance those properties were often employed. Therefore, the main objective of this research was to maximize the recycled LDPE content in the production of a three-layer LDPE shrink film without adding any additives and additional processes by using proper type of recycled LDPE. Instead of using post-consumer waste as implemented by other works [3,8-11,16], LDPE scraps from blown film production line was used as a recycled polymer (re-LDPE) to avoid the property variation that can occur when post-consumer waste is used [10,11]. The effect of the recycle polymer content on the properties of a three-layer LDPE shrink film prepared by blown film co-extrusion was studied. While its skin layers comprised only virgin LDPE, its middle layer were made of a blend of recycled and virgin LDPEs. The blended compositions of re-LDPE and virgin LDPE were varied. Thermal, mechanical, and optical properties of the prepared films were investigated and compared to those of the film containing only virgin LDPE in the middle layer to determine suitable amount of re-LDPE for property improvement of this three-layer LDPE shrink film.

2. Experimental Procedures

2.1 Materials

Virgin LDPE resin (LDPE) (melt flow index: 0.23 g 10 min^{-1}) was purchased from TPI Polene Co., Ltd., Thailand. Recycled LDPE (re-LDPE) resin (melt flow index: 0.31 g 10 min^{-1}) was supplied by Sun Sea Plastic P.S. Co., Ltd., Thailand. This re-LDPE resin was made of LDPE film scraps obtained from blown film production line and pelletized by a single screw extruder.

2.2 Methods

The three-layer (LDPE/LDPE:re-LDPE/LDPE) shrink films were prepared using a Reifenhauser blown film co-extrusion machine. The processing temperatures were controlled at 185-195^{o}C whereas the output rate and the blow-up ratio (BUR) were fixed at 280 kg hr^{-1} and 2.73, respectively. The width and thickness of the obtained films were set at 1500 mm and 150 microns, respectively. The LDPE:re-LDPE ratios in weight percentage were varied from 100:0, 85:15, 70:30, 55:45, 40:60 to 0:100.

Thermal behaviours of the prepared films were characterized using Perkin Elmer Diamond differential scanning calorimeter (DSC) operating under nitrogen atmosphere using a flow rate of 50 ml min^{-1}. Two heating cycles were performed at a heating rate of 10^{o}C min^{-1} in the range from 50 to 200^{o}C. The percentage of crystalline fraction (X_c) can be calculated from $X_c = (\Delta H_m/\Delta H_m^0) \times 100$ where ΔH_m is the heat of fusion of the specimen observed from DSC and ΔH_m^0 is the heat of fusion for 100% crystalline polyethylene (ΔH_m^0 = 288 J g^{-1}) [17].

Tensile strength, seal strength, and heat shrink ability of the obtained films were measured in both machine and transverse directions followed ASTM D882, ASTM F88, and ASTM D2732, respectively. Their haze and gloss were characterized according to ASTM D1003 and D2457, respectively.

3. Results and Discussion

It can be seen from Fig. 1 that each DSC thermogram of the prepared three-layer shrink films exhibits a single sharp peak corresponding to one melting temperature (T_m) at around 109^{o}C. This observation suggests that all components of these films are miscible due to their similarities in the chemical structures and molecular chain branches which result in similar thermal behavior and crystallizing ability to form a co-crystallization [18]. In addition, because of these similarities, most of the blended samples exhibited only a slight decrease of T_m less than 1^{o}C or even higher T_m at proper re-LDPE content compared to those of the single-component films as seen from Table 1. This result is different from that of the films having blended components with less similarities where a notable decrease of T_m was observed [3]. However, Table 1 also shows the difference in crystallinity percentage between the prepared films. Like previous report [17], the crystallinity percentages increase as the values of their corresponding heat of fusion (ΔH_m) increase. This result suggests a possibility of the influence of re-LDPE molecules on the crystallization during the manufacturing process. According to Table 1, the ΔH_m and crystallinity percentage respectively increase from 45.36 J/g to 53.63 J/g and 15.75% to 18.62% with increasing the amount of re-LDPE in the middle layer from 0 to 100 wt%. This may be caused by a decrease in the average molecular weight of a recycled polymer due to thermal degradation during the reprocessing of LDPE scraps into re-LDPE pellets which can be indicated by higher melt flow index (MFI) of a reprocessed polymer compared to its virgin counterpart since the MFI of a polymer is inversely related to its molecular weight [1]. In this case, re-LDPE and LDPE resins exhibited the MFI at 0.31 g 10 min^{-1} and 0.23 g 10 min^{-1}, respectively. Hence, the former had lower average molecular weight than the latter. With shorter chain length due to its lower molecular weight compared to that of virgin LDPE molecules, re-LDPE molecules were less entangled and were easier to move and align together upon stretching and pulling up during blown film manufacturing process which resulted in better chain orientation. This enhanced the crystallization of the polymer chains as clearly seen from Table 1 for the film containing 100 wt% of re-LDPE in the middle layer whose crystallinity percentage is highest. In the case of the films containing the blends of LDPE and re-LDPE resins in the middle layer, the presence of re-LDPE can enhance the crystallization as previously described and possibly induce faster formation of the crystals. The firstly-formed crystals can consequently act as the source of the nucleation for further crystallization and crystal growth [19]; as a result, the crystallization is increasingly promoted with increasing the amount of re-LDPE as shown in Table 1.

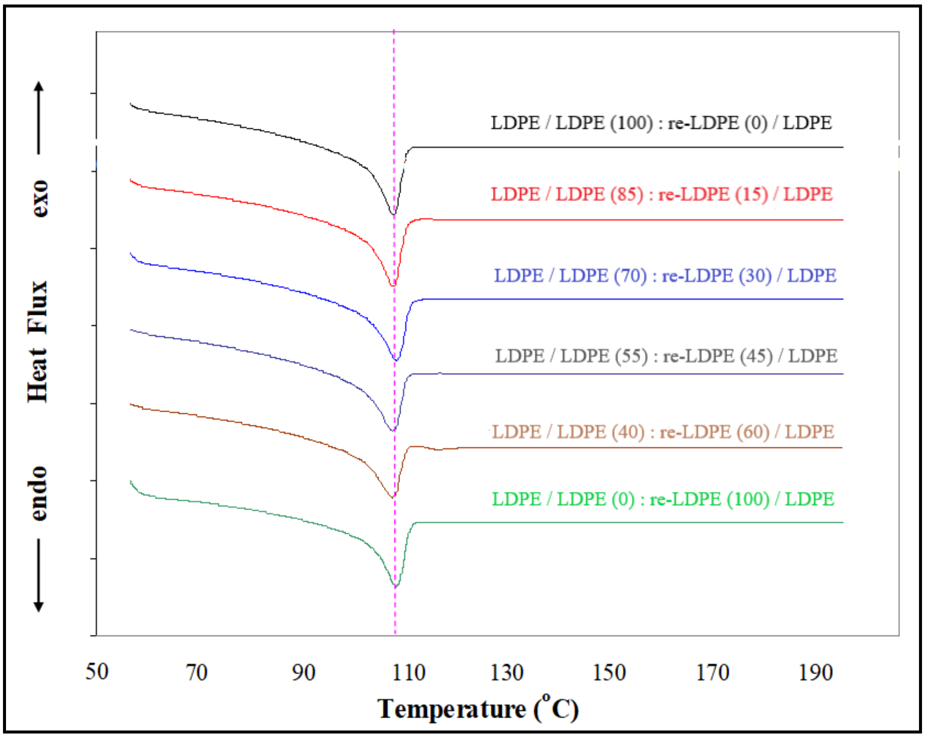

Fig. 1 *DSC thermograms of three-layer shrink films*

Table 1 *Melting temperature (T_m), the heat of fusion (ΔH_m) and crystallinity percentage of three-layer shrink films*

re-LDPE amount in the middle layer (wt%)	T_m (°C)	ΔHm (J g-1)	Crystallinity (%)
0	109.45	45.36	15.75
15	109.27	47.55	16.51
30	109.94	48.42	16.81
45	109.08	49.02	17.02
60	108.90	51.32	17.82
100	109.85	53.63	18.62

Due to the direction of the flow and blow-up mechanism of blown-film extrusion process, an extruded product normally exhibits the anisotropic properties [20] as seen in Fig. 2 where tensile strength of the films in machine direction (MD) is higher than that in TD. This is because the polymer chains are predominantly aligned in MD compared to transverse direction (TD) [1] indicating that there are more primary bonds (i.e., covalent bonds) that can withstand the applied tensile force in MD than in TD. In addition, since the crystalline regions can help the polymer chain withstand the applied force, the tensile strengths of the films in both directions increase with increasing the amount of re-LDPE in the middle layer from 0 to 100 wt% due to an increase in crystallinity percentage. As shown in Fig. 2, their tensile strengths are increased from 13.03±0.39 MPa to 14.79±1.03 MPa in MD and 11.23±0.72 MPa to 14.35±0.88 MPa in TD. At certain amounts of re-LDPE, the crystallinity percentage is enough to make the films more isotropic as seen from Fig. 2 that the tensile strengths in both directions are comparable when 60 wt% and 100wt% of re-LDPE were used. At 60 wt% of re-LDPE, the tensile strengths in TD and MD are at 14.02±0.33 MPa and 14.09±0.41 MPa, respectively. At 100 wt% of re-LDPE, the tensile strengths in TD and MD are at 14.35±0.88 MPa and 14.79±1.03 MPa, respectively.

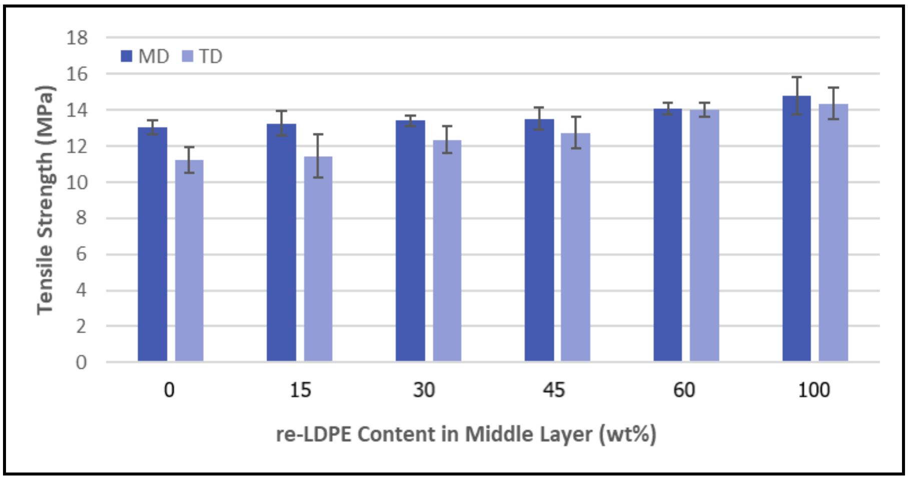

Fig. 2 *Tensile strength of three-layer shrink films*

Various packaging applications require the packaging films to have heat-sealing ability. To achieve an effective heat sealing for a semi-crystalline polymer, the molecular segments must melt upon heating, diffuse across the interface of the two layers and re-crystallize after cooling [21]. In this research, the influence of re-LDPE on heat seal ability of the three-layer shrink films was found. Fig. 3 shows that the heat seal strength can be increased from that of the film containing only virgin LDPE to around 22% in MD and 13% in TD when 100wt% of re-LDPE was used as the middle layer. This may be because re-LDPE, compared to virgin LDPE, has higher MFI (i.e., lower molecular weight) which can promote its ability to melt and diffuse across the interface under the sufficient time. Moreover, the re-crystallization of the polymer chains at the sealing interface after cooling may be easier to occur as mentioned earlier. As a result, the heat seal strength of the prepared three-layer shrink films was increased. Since the movement of the polymer chains in MD is more difficult than that in TD as previously described, any change to the components affecting such movement can result in greater improvement of the seal strength in MD than in TD.

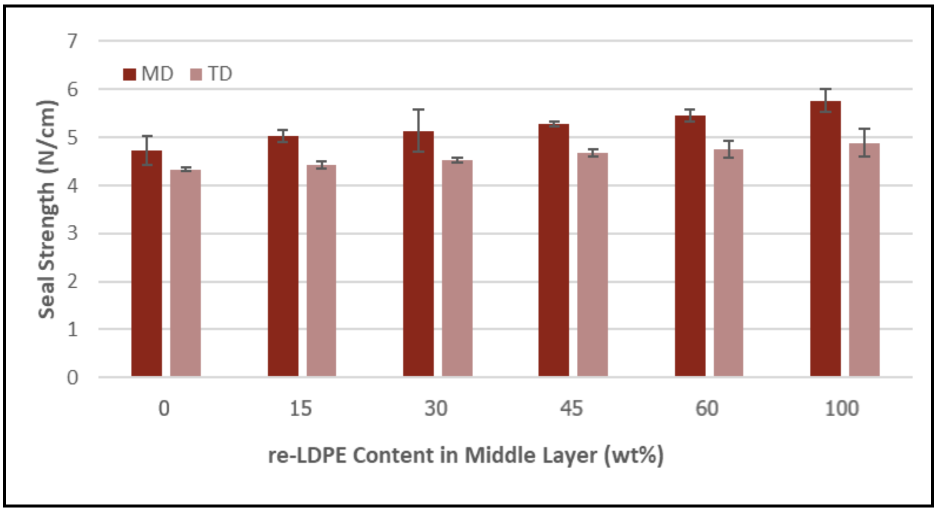

Fig. 3 *Seal strength of three-layer shrink films*

Shrink ability of a shrink film is affected by molecular behavior of the polymer chains. In general, the polymer chains in the film are randomly intertwined with no particular pattern. Upon heating during the processing, they are straightened and aligned to the direction of orientation. When the film is properly cooled, these chains remain in this stretched state until the film is re-heated and the polymer chains shrink back to their original form [22]. For a blown polyethylene film, it is usually shrinkable in MD because its molecular chains are normally stretched and oriented much more in this direction compared to TD as previously mentioned [1]. The shrink ability can be determined as a shrink percentage. From original length at 10 cm, after subjecting to shrinkage measurement according to ASTM D2732, the lengths of the test samples containing re-LDPE in the middle layer at 0, 15, 30, 45, 60 and 100 wt% in MD were 2.21±0.03, 2.26±0.02, 2.31±0.03, 2.39±0.02, 2.41±0.02 and 2.47±0.02 cm, respectively. On the other hand, the lengths of the samples in TD were 7.39±0.02, 7.41±0.02, 7.49±0.01, 7.53±0.02, 7.59±0.02 and 7.64±0.02 cm in TD, respectively. These values can be calculated into shrink percentages as shown in Fig. 4 where the shrink percentages are around 75-78% for MD and 23-26% for TD. Fig. 4 also displays that the shrink percentages slightly decreased with increasing the amount of re-LDPE in the middle layer. This may be due to the difficulty in the movement of polymer molecules caused by the increased crystalline regions upon increasing the re-LDPE amounts. Despite the slight decrease in the shrink ability, these results indicate that all prepared shrink films still exhibit the shrink percentages in a normal range of mono-oriented shrink films which are 60–80% in MD and 10–20% in TD [1].

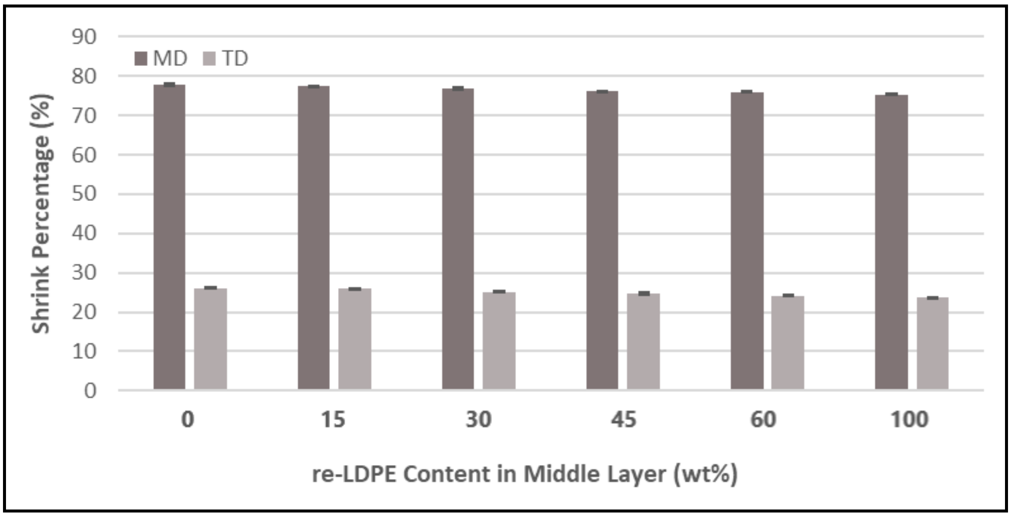

Fig. 4 *Shrink percentage of three-layer shrink films*

An increase in crystallinity percentage upon increasing re-LDPE content also affected the gloss and haze of the prepared films as seen from Fig. 5. In the case of gloss, a smooth surface of the film is required to reflect the light. The surface roughness caused by the crystals increasing formed when re-LDPE content was increased from 0 to 100 wt% can reduce the reflection and lower the gloss [1] from 71.28 to 67.42 GU with standard derivations less than 0.70 GU for all data. On the other hand, the crystals with certain sizes can scatter the light within the film bulk resulting in the haze. However, smaller crystals created by faster crystallization or the use of a nucleating agent can decrease the haze [1,19] which may be the case for this research since the haze shows a tendency to decrease despite an increase in crystallinity percentage. When the LDPE/re-LDPE blends or only re-LDPE were used as the material for the middle layer, re-LDPE molecules can enhance and induce the crystallization to create smaller crystals as previously mentioned. Therefore, haze values continue decreasing from 15.90±0.27% to 12.48±0.13% with increasing the amount of re-LDPE up to 60 wt%. When crystallinity percentage is higher at 100 wt% of re-LDPE, haze value minutely increased back to 13.42%. This may be because the crystalline regions became too high causing more light scattering within the film bulk. Therefore, re-LDPE content must be considered in film preparation since excessive gloss can reflect too much light and interfere with the appearance of the packed product and high haze can prevent the sight of the product inside the package [1].

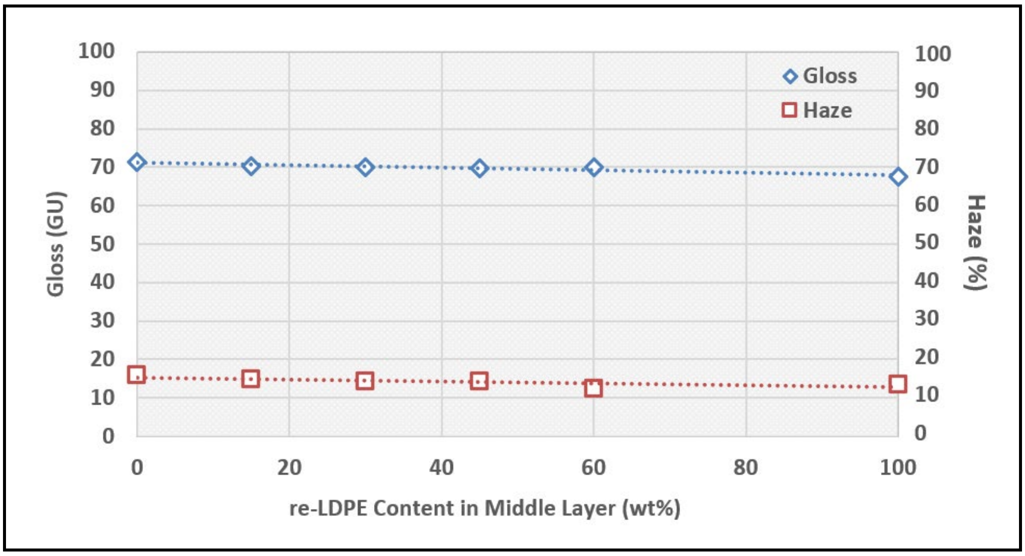

Fig. 5 *Gloss and haze of three-layer shrink films*

4. Conclusion

Several properties of a three-layer shrink film prepared by blown film co-extrusion were improved by the presence of recycled LDPE in the middle layer either by itself or as a miscible blend with virgin LDPE. The tensile and seal strengths of these shrink films in both machine and transverse directions were better than those of the film containing only virgin LDPE. Their gloss and haze values slightly decreased while their shrink percentages in both directions were in the typical range for mono-oriented shrink films. The property improvement was clearly affected by the amount of recycle LDPE. While the film prepared using re-LDPE 100 wt% had highest tensile and seal strengths, the film prepared using re-LDPE 60 wt% exhibited lowest haze with similar gloss to the film prepared without re-LDPE. The above results suggests that the recycle LDPE obtained from the scraps of blown film production line can be used to improve the properties of the multilayer films which can be controlled by using proper type of recycled LDPE and adjusting its content without the need to add any additives or use additional processes.

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Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *Omattaya Montrikul, Vimolvan Pimpan;* ***data collection:*** *Omattaya Montrikul;* ***analysis and interpretation of results:*** *Omattaya Montrikul, Vimolvan Pimpan;* ***draft manuscript preparation:*** *Omattaya Montrikul, Vimolvan Pimpan. All authors reviewed the results and approved the final version of the manuscript.*

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