

Development and Characterization of Polypropylene/Polyethylene Vinyl Acetate/Micro Cellulose Trays as a Prototype for Chilled Food Packaging Application

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Abstract

Cellulose and polypropylene (PP) were successfully designed as a composite material. To obtain higher efficiency, ethylene vinyl acetate (EVA) with vinyl acetate (VA) content 28 % was integrated as a coupling agent. The presence of EVA improved the composite compatibility and polypropylene matrix toughness under low temperatures. The composite was compounded by twin screw extrusion (TSE) with 30 part of EVA per hundred parts of resin (phr) and varying ratios of cellulose 5, 10, 20 and 30 phr, respectively. The effects of cellulose and EVA on properties of the composite were investigated. Thermal degradation temperature of PP/cellulose composites with and without EVA was higher than pure PP. The impact strength of the composite with EVA was increased compared to composite without EVA. PP/EVA/cellulose-10 phr is the best condition which showed a very high elongation at break and $\tan \delta$. In conclusion, the role of cellulose and EVA could improve the thermal stability and flexibility of thermoplastic composites and would be useful in packaging chilled food.

Keywords: Cellulose, polypropylene, polyethylene vinyl acetate; chilled food packaging

Introduction

In recent year, the growth of population worldwide has been exponentially increasing; the requirement on food and its related industry are in high demand of production. Chilled food is one of the great convenient and a high-quality product that continues to grow and evolve respond to the demand of consumers. As the high demand for product and the expenditure of processing, polyolefin gained many interests to be developed as food packaging. One of the most effective materials to use as a tray for various chilled food meals is Polypropylene (PP) due to its many suitable properties such as microwave safe, good processability, good chemical resistance and good mechanical properties [1]. In order to develop polyolefin as a sustainable and renewable food packaging, bio-based materials were used. The application of bio-based materials plays an important role in many commercialisations such as building material, automotive part, electronic component, drug delivery system, medical device as well as food contact material. Among other filler, cellulose fiber become an attractive choice for being a reinforcing materials because they can enhance many properties of polymeric matrix such as biodegradability, mechanical properties, dimensional stability and thermal stability [2-5].

However, the drawback such as incompatibility between cellulose fiber and hydrophobic as well as polypropylene matrix the brittleness of polypropylene at a temperature below glass transition temperature (T_g) greatly reduce the ability of cellulose-based packaging while using in the application area. In order to use cellulose and polypropylene composite with higher efficiency, development of cellulose and

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polypropylene composite with a small amount of ethylene vinyl acetate (EVA) has been investigated. From the structural point of view, the structure of EVA monomer is composed of ethylene and vinyl acetate. It was easy to be attached to propylene unit and the hydroxyl group of cellulose to form a chemical linkage. Moreover, the introduction of an elastomer EVA can simultaneously enhance the toughness and stiffness relative to neat polypropylene [6-8].

The objective of this research work is to focus on the development of EVA and cellulose extracted from Kraft paper incorporate into PP matrix. The composite was compounded by twin screw extrusion (TSE) with 30 part of EVA per hundred parts of resin (phr) and varying ratios of cellulose 5, 10, 20 and 30 phr, respectively. Effects of cellulose loading and EVA on mechanical properties, morphology properties, thermal properties and chemical properties of the composite were investigated as a prototype of chilled food packaging application.

Materials and methods

Materials

Polypropylene (PP, EL-Pro PXXXXS) was obtained from SCG Chemicals (Public) Co., Ltd., Thailand with a density of 0.963 g/cm³, a melting point of 200 °C and melt flow index of 2.8 g/10 min (230 °C, 2.16 kg). Cellulose from Kraft process was obtained from SCG Packaging (Public) Co., Ltd., Thailand. An analytical grade of hydrochloric acid was purchased from Sigma-Aldrich for further preparation of micro-cellulose (MC). Polyethylene vinyl acetate (EVA, MV1055) with 28 % vinyl acetate (VA) content was obtained from TPI Polene (Public) Co., Ltd., Thailand, has a density of 0.953 g/cm³, a melting point of 70 °C and melt flow index of 8 g/10 min (190 °C, 2.16 kg).

Methods

Modification of cellulose from Kraft paper

Cellulose was extracted from Kraft paper. Briefly, a small amount of Kraft paper was milled in order to reduce its size prior to acid hydrolysis. A 1500 cm³ of 1.5 M hydrochloric acid (HCl) solution was poured into an 80 g of product and then stirred at room temperature for 48 h. The resulting product was washed with DI water, following up with the air-drying process before being grinded into a fine powder using a general electric blender.

Preparation of cellulose-based composite

The cellulose-based composite was prepared by melt mixing method using twin screw extruder (Collin model T-20). The ratio between polypropylene, ethylene vinyl acetate, and modified cellulose is presented in **Table 1**. The temperature for extruder was set between 140 - 170 °C with the screw rotation speed of 80 rpm. After that, the composite was cooled down in a water bath and it was dried at 60 °C for 24 h.

Table 1 Composition of composites.

Samples	Amount of materials (phr)*		
	PP	EVA	Cellulose
Neat PP	100	0	0
Neat EVA	0	100	0
PP/cellulose-5	100	0	5
PP/cellulose-10	100	0	10
PP/cellulose-20	100	0	20
PP/cellulose-30	100	0	30
PP/EVA	100	30	0
PP/EVA/cellulose-5	100	30	5
PP/EVA/cellulose-10	100	30	10
PP/EVA/cellulose-20	100	30	20
PP/EVA/cellulose-30	100	30	30

*Per hundred resins (the weights of EVA and cellulose were calculated based on 100 % weight of PP).

Characterization and testing

Scanning Electron Microscope (SEM)

The morphological properties were investigated by JEOL JSM-6400 SEM with 15 kV accelerating voltage. The fracture surfaces of samples were mounted on adhesive tape and sputter coated with Au to enhance the electrical conductivity.

Fourier transform infrared (FTIR)

FTIR spectra were examined by Perkin Elmer Universal ATR Sampling Accessory (Massachusetts, USA) and recorded over the range of 650 to 4000 cm^{-1} at a resolution of 4 cm^{-1} with a scan frequency of 32 times.

Universal testing machine

Tensile test specimens were prepared with a dumbbell shape following the ASTM D638 standard and then oven dried at 40 °C for 24 h before testing. The tensile strength, elastic modulus, and elongation at break values were recorded with a Hounsfield Tensile Testing Machine at room temperature with a crosshead speed of 50 mm/min and load cell 25 kN. Five replicates were tested to obtain an average value for each composition.

Impact testing

Impact testing was performed according to ASTM D256 Izod impact method. The sample dimension was 60×12×3 mm^3 with 2 mm notch dept. The Impact energies were determined for 5 test specimen of each composition using CEAST impact tester at room temperature.

Dynamic mechanical analysis (DMA)

Dynamic mechanical properties were investigated by NETZSCH DMA 242 in compression mode. Rectangular specimen sample was heated from -50 to 100 °C at a heating rate of 5 °C/min under flowing nitrogen and frequency of 1 Hz. The damping factor ($\tan \delta$) was calculated from storage modulus (E') and loss modulus (E'') as following Eq. (1).

$$\tan \delta = E'' / E' \quad (1)$$

Differential scanning calorimetry (DSC)

The DSC measurement was performed by Mettler Toledo DSC3+ (Leicester, England) to obtain crystallization temperature and melting temperature. The samples were heated from -50 to 200 °C at a heating rate of 10 °C/min to dispose of thermal history, cooled to -50 °C and reheated with the same condition under 20 mL/min flowing nitrogen. The percentage of crystallinity is related to the melting enthalpy of polymer (ΔH_m) and 100 % crystalline polymer (ΔH_m°). It was calculated using Eq. (2).

$$\% \text{ Crystallinity} = (\Delta H_m / \Delta H_m^\circ) * 100 \% \quad (2)$$

where ΔH_m° is 207 J/g and 293 J/g for the heat fusion of 100 % crystalline PP [9] and PE [10].

Thermogravimetric analysis (TGA)

The thermogravimetric behavior of cellulose-based composites was investigated by Mettler Toledo TGA 2 (Leicester, England). Samples with approximately 10 mg were heated from 25 to 600 °C at a heating rate of 10 °C/min under 20 mL/min flowing nitrogen.

Statistical analysis

Data were analyzed to investigate the influence of cellulose and EVA to PP matrix, using a one-way analysis of variance (ANOVA), followed by independent-sample t-test (Statistics programming software SPSS 19.0, Chicago, U.S.A.). A p-value < 0.05 was considered statistically significant.

Results and discussion

Morphological properties

Scanning electron microscope was used to evaluate the uniformity of size and shape of cellulose and the compatibility between cellulose and polymer matrix. **Figure 1** exhibits the morphology of cellulose derived from Kraft paper using chemical and mechanical treatment. The size of cellulose fibers are 80 to 150 micrometer in length and the diameter are approximately 10 micrometers.

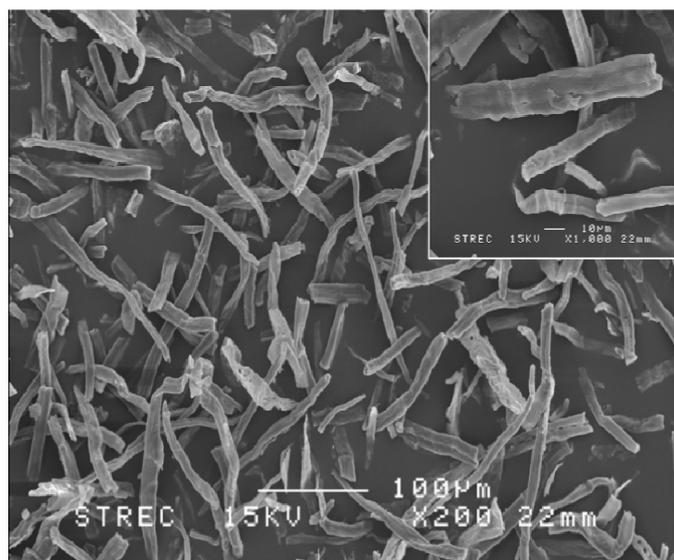


Figure 1 SEM images of cellulose extracted from Kraft paper.

The addition of cellulose and EVA influenced the morphology of PP composites that were shown in **Figure 2**. The morphology of the PP/cellulose-10 composite (**Figure 2(b)**) exhibited a part of cellulose fiber on the fracture surface of the composite. On the other hand, that did not clearly see the part of cellulose on the fracture surface of PP/EVA/cellulose-10 composite (**Figure 2(d)**). Moreover, the SEM pictures of PP/EVA composite show the 2-phase morphology of EVA distributed in PP matrix. As reported by Aghjeh *et al.* [11], Maciel *et al.* [7] and Mihaylova *et al.* [12], who employ SEM to examine the structure of PP/EVA blend with various ratio. Their SEM micrographs of PP/EVA blends show that in PP-rich blends, PP was in a continuous phase, while EVA formed droplets within the blend. In addition, the image of EVA-rich blends shows the dispersed PP particles in the EVA matrix. For the blends with composition 50/50 wt. %, co-continuous type structure was observed.

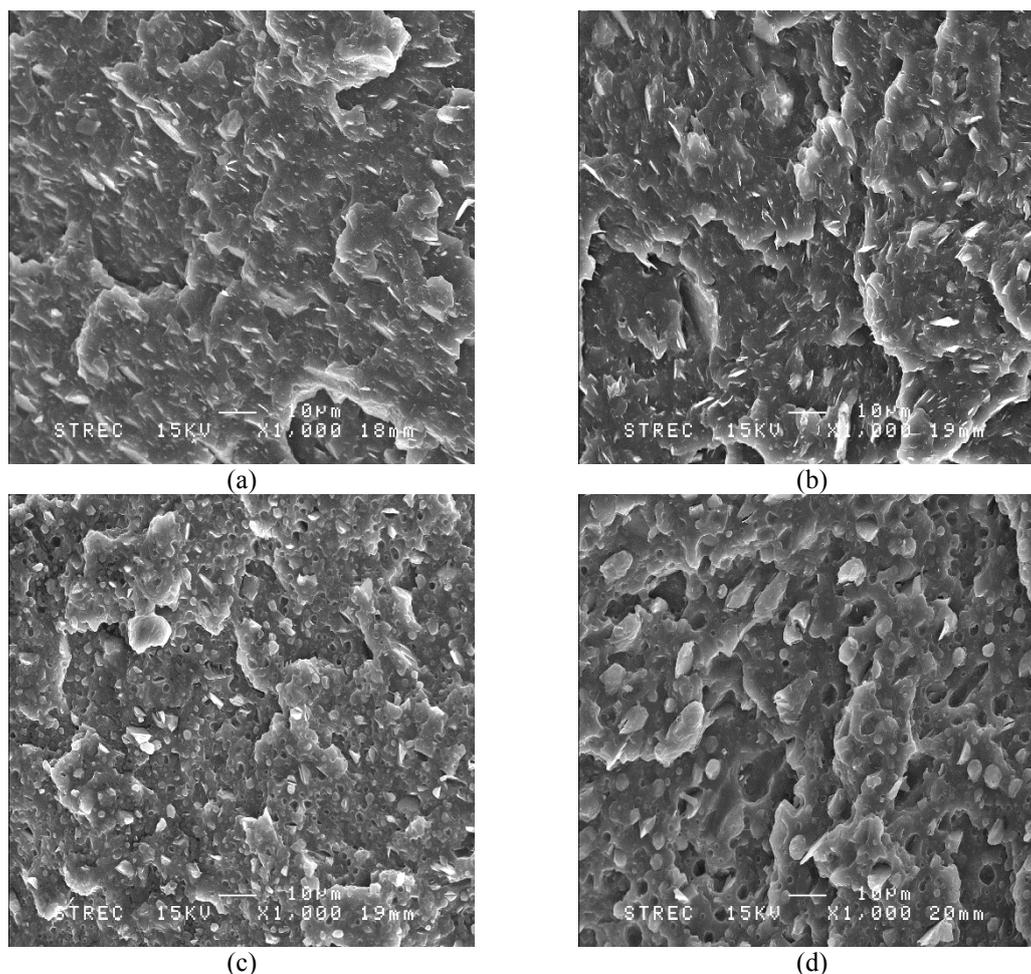


Figure 2 SEM photos of the fracture surface of (a) PP, (b) PP/cellulose-10 phr, (c) PP/EVA and (d) PP/EVA/cellulose-10 phr (1000× magnification)

Chemical properties

FTIR pattern of cellulose, PP, EVA, PP/EVA and composite with and without EVA was shown in **Figure 3(A)**. In the cellulose spectrum, there is a broad peak of the hydroxyl group at 3334 cm^{-1} , a stretching and bending of C-H in alkane group at 2897 and 1428 cm^{-1} respectively, and a strong absorption vibration from 1081 to 1160 cm^{-1} which refers to C-O stretching of the primary and secondary alcohol.

The FTIR spectrum of PP shows a strong C-H stretching vibration band of alkane from 2838 to 2950 cm^{-1} , and the peak at 1376 and 1455 cm^{-1} is represented by C-H bending of the alkane. Similar to PP spectrum, EVA has a C-H stretching at 2849 and 2917 cm^{-1} , and also C-H bending at 1371 and 1465 cm^{-1} . In addition, the peak at 1737 cm^{-1} represents C=O stretching of the ester group and the peak at 1238 cm^{-1} belong to C-O stretching of the ester group in EVA. The PP/EVA blend shows all spectrum of pure PP and pure EVA, no new peak was detected, which means they have no interfacial reaction or interaction between them. Focusing on PP/EVA/cellulose composite, the peak at 1737 cm^{-1} which represent C=O stretching of EVA was less intense compared to PP/EVA blend. This suggested that carbonyl group of

EVA was interacted with the hydroxyl group of cellulose and formed a hydrogen bond [13] as shown in **Figure 3(B)**.

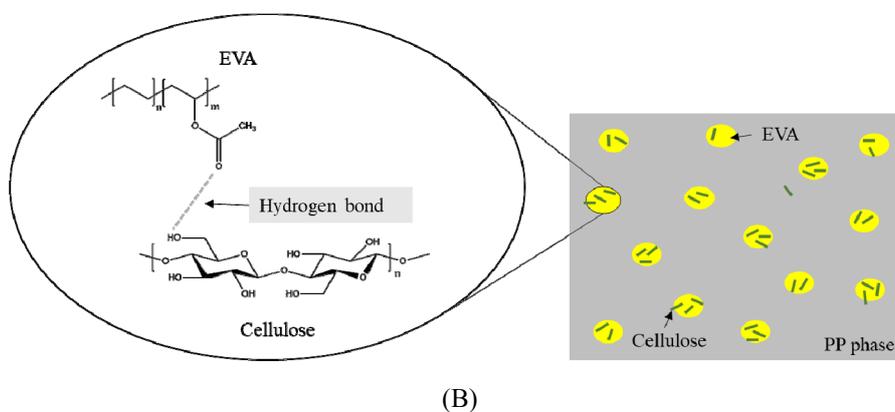
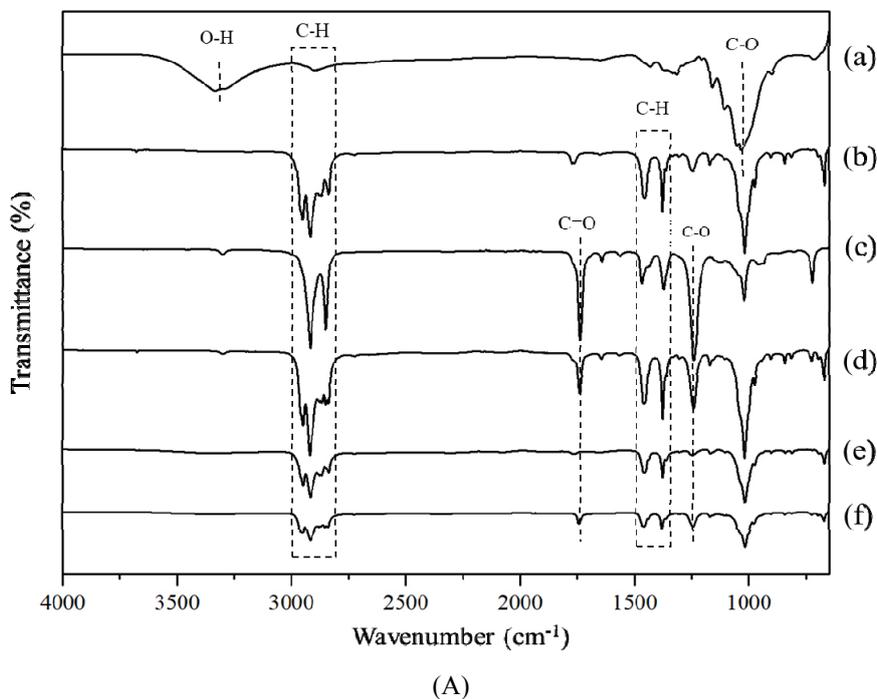


Figure 3 (A) FTIR spectra of (a) cellulose, (b) PP, (c) EVA, (d) PP/EVA, (e) PP/cellulose-10 phr, (f) PP/EVA/cellulose-10 phr and (B) Chemical interaction of cellulose and EVA dispersed on PP matrix phase

Mechanical properties

Mechanical properties of composites were observed by the tensile testing. **Table 2** exhibits the tensile strength, elongation at break, elastic modulus and impact strength of composites. As reported in morphological properties, the blend of PP and EVA is likely to be immiscible. Therefore, the mechanical

properties of materials depend on those of PP polymer but they are going to be weaker than neat PP. The tensile strength and elongation at break of PP/cellulose composite further decreases with increase in cellulose content due to the poor interfacial interaction between PP matrix and cellulose. Therefore, Cellulose cannot act as a mechanical reinforcement to be a means of stress transfer, but it acted as a defective point [14,15]. On the other hand, the addition of EVA resulted in an improvement in the elongation at break of materials of up to 5.4 %, which was the composite with 10 phr of cellulose content. This was affected by the softening of the matrix and the flexible interface around the cellulose in the presence of EVA [16]. However, the tensile strength and elastic modulus of PP/EVA/cellulose were lower than the composite without EVA, indicating that the presence of EVA elastomer decreases the strength and stiffness of the composite materials. The elastomeric properties of the dispersed phase EVA were effective to improve the toughness of the PP and allow the absorption of energy of greater impact. However, the impact strength of PP/EVA/cellulose was decreased drastically with increasing cellulose content nearly to the same value of PP/cellulose composites. Focusing on PP/EVA/cellulose-5, the highest impact strength of the composites was seen (11.52 J/m²). This was because EVA was able to embed cellulose fiber, leading to a reduction in stress concentration between cellulose and PP interface, resulting in higher impact strength but lower elastic modulus [8,17].

Table 2 Mechanical properties of PP, PP/EVA, and PP/cellulose composite with and without EVA.

Samples	Tensile strength (MPa)	Elongation at break (%)	Young's Modulus (GPa)	Impact strength (J/m ²)
Neat PP	28.7±2.2 ^a	17.8±9.7 ^{a,b}	2.0±0.2 ^{a,b}	16.53±1.16 ^{a,b}
PP/EVA	20.3±0.4 ^b	17.3±7.7 ^{a,b}	1.4±0.1 ^{a,c}	21.49±5.47 ^{a,b}
PP/cellulose-5	18.3±3.5 ^{a,b}	2.8±0.6 ^{a,c}	1.5±0.1 ^{a,b}	6.19±1.55 ^{a,b}
PP/cellulose-10	17.6±3.8 ^{a,b}	2.6±0.5 ^{a,b}	1.7±0.3 ^a	4.96±1.16 ^a
PP/cellulose-20	16.0±1.7 ^{a,c}	2.7±0.3 ^a	1.5±0.2 ^{b,c}	5.44±0.86 ^a
PP/cellulose-30	15.2±3.2 ^c	2.2±0.7 ^{a,b}	1.7±0.3 ^{a,c}	4.05±0.85 ^{b,c}
PP/EVA/cellulose-5	13.6±1.1 ^{a,c}	4.3±1.7 ^{a,c}	1.1±0.1 ^b	11.52±4.08 ^{b,c}
PP/EVA/cellulose-10	15.0±3.8 ^{b,c}	5.4±2.5 ^{a,b}	1.2±0.3 ^b	5.28±1.27 ^c
PP/EVA/cellulose-20	11.6±1.9 ^{a,b}	4.1±0.8 ^{a,c}	1.0±0.1 ^{b,c}	5.08±1.85 ^c
PP/EVA/cellulose-30	10.1±1.3 ^{a,b}	3.2±1.2 ^{b,c}	0.9±0.1 ^c	6.67±0.94 ^{b,c}

*EVA can't be analyzed with the same conditions with others because of elastomeric properties.

Thermo-mechanical properties

One of the most important properties of composites for chilled food packaging is dynamic mechanical properties. DMA is a useful technique to study the viscoelastic behavior as a function of stress, temperature, time and frequency to determine the processibility and end-user performance. **Figures 4 and 5** show the influence of EVA to the storage modulus (E') and damping factor (tan δ) of PP/cellulose composites. Throughout the sample, the drastic change in the modulus from glassy high modulus region to rubbery region is shown, especially for the EVA curve that represents the T_g of the materials. Focusing on 0 °C, which is the lowest storage temperature for chilled food packaging, the PP/cellulose composite

storage module was much lower than the pure PP indicating a poor interfacial interaction between PP and cellulose. The incorporation of EVA in composites with cellulose 10 and 30 phr exhibited much higher storage modulus than neat PP and composite without EVA. Its further increases with increase in cellulose content (except 20 phr of cellulose) attributing that the interfacial interaction was improved which allowed the stress transfer at the interface and increased in stiffness [18].

The $\tan \delta$ data is determined by the ratio of loss and storage modulus. So, it can reflect the relaxation behavior of materials more intensely than loss modulus and represents the softness of specimen. The EVA exhibits relaxation peak at $-30\text{ }^{\circ}\text{C}$ while PP display peak at $15\text{ }^{\circ}\text{C}$ which is the glass-rubber relaxation (β relaxation) of amorphous portions of EVA and PP [19]. The $\tan \delta$ curve of the blend shows 2 separate T_g peak confirming the immiscibility of the blend [20]. At $0\text{ }^{\circ}\text{C}$, PP/cellulose-10 composite display the lowest damping data influence by the obstacle of cellulose as mention above, while the $\tan \delta$ curve of the composites with EVA was higher than the composite without EVA, This leads to the conclusion that the existence of EVA in 10 and 30 phr of cellulose composite can enhance the relaxation softness and also stiffness of composite at the chilled food storage temperature.

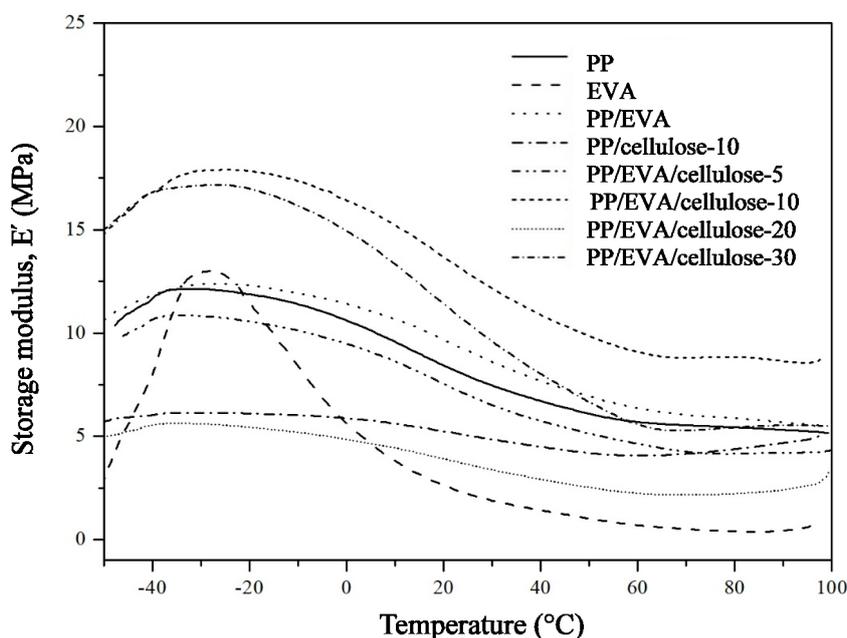


Figure 4 Storage modulus curves of PP, EVA, PP/EVA and PP/cellulose composites with and without EVA.

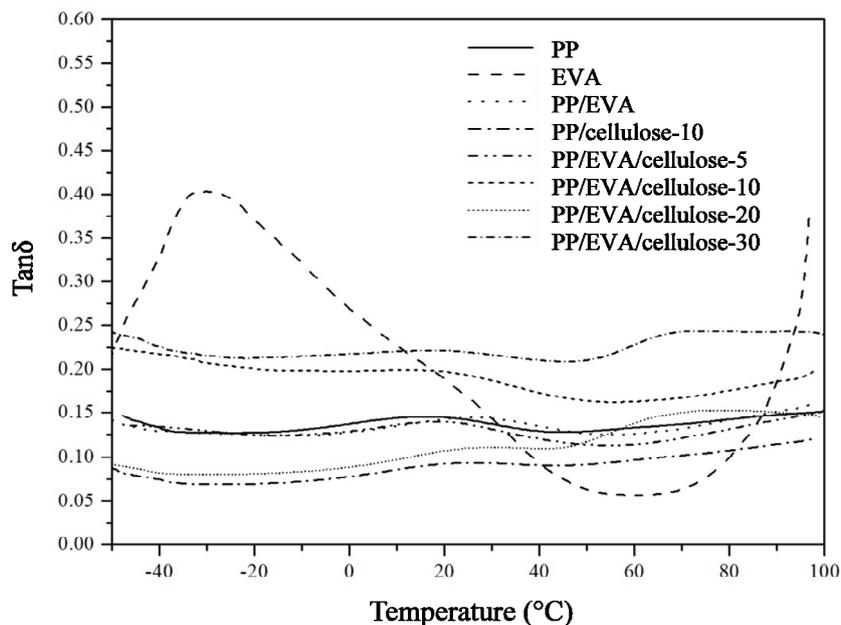


Figure 5 Tan δ curves of PP, EVA, PP/EVA and PP/cellulose composites with and without EVA.

Thermal properties

The DSC melting data of composites are summarized in **Table 3**. The composites with cellulose display slightly lower T_m compare to neat PP. This was due to the existence of cellulose changed, the crystallization of PP matrix around the cellulose reinforcement phase and promoted a weak bond between cellulose and PP [15]. PP/EVA blend exhibits two melting point at 71 °C for EVA and 164 °C for PP which is not significantly different from pure EVA and PP, indicating that EVA does not influence the crystallinity of PP [21]. The melting enthalpy is related to the amount of polymer content in the sample. As expected, the melting enthalpy of composites was decreased with decreasing amount of PP and EVA. The crystal melting temperature is an important characteristic to deciding the processing temperature of materials [22]. The T_c and T_m of the composites with EVA were slightly different from the neat PP. However, it has less effect on the temperature of operation of extrusion and injection molding process.

Table 3 DSC melting data of PP, EVA, PP/EVA and PP/cellulose composites with and without EVA.

Samples	T _c (°C)		T _m (°C)		ΔH _m (J/g)		X _c (%)	
	EVA	PP	EVA	PP	EVA	PP	EVA	PP
Neat PP	-	125.8	-	165.8	-	96.1	-	46.4
Neat EVA	52.0	-	72.2	-	69.4	-	23.7	-
PP/cellulose-5	-	126.5	-	165.8	-	93.3	-	45.1
PP/cellulose-10	-	127.0	-	165.2	-	88.5	-	42.8
PP/cellulose-20	-	126.8	-	165.8	-	63.6	-	30.7
PP/cellulose-30	-	127.3	-	164.2	-	80.2	-	38.7
PP/EVA	53.8	126.0	71.0	164.5	5.8	73.5	2.0	35.5
PP/EVA/cellulose-5	53.8	124.2	71.2	163.8	9.6	58.3	3.3	28.2
PP/EVA/cellulose-10	53.5	123.3	72.5	164.5	5.6	64.5	1.9	31.1
PP/EVA/cellulose-20	54.2	123.7	71.3	164.3	5.6	62.4	1.9	30.1
PP/EVA/cellulose-30	54.5	124.8	71.3	163.7	3.6	57.9	1.2	27.9
PP/EVA/WP (45/45/10) [21]	-	-	85.5	167.5	10.2	35.8	-	-
MAPP/EVA/WP (45/45/10) [23]	-	-	84.8	164.2	9.8	36.3	-	-
PP/SBR/WF (80/10/10) [19]	-	115.4	-	158.7	-	59.4	-	31.2

The degradation step of PP, EVA, cellulose, PP/EVA, PP/cellulose-10 and PP/EVA/cellulose-10 composite were shown in **Figure 6**. The TGA curve of cellulose and neat PP showed degradation temperature at 345 and 450 °C, respectively. The influence of cellulose-reinforced PP on degradation temperature was observed. The PP/cellulose composites showed 2 degradation steps of cellulose and PP decomposition. The degradation temperature of PP in PP/cellulose composites was higher (470 °C) compared to the neat PP. This is probably because of the crystallinity of PP is changed in the presence of cellulose [24]. The EVA curve shows a degradation of vinyl acetate group at 350 °C and a complete decomposition of EVA backbone at 470 °C. The PP/EVA/cellulose composites also display 2 degradation steps. The first weight loss process is combined with the degradation of cellulose and vinyl acetate group of EVA. The second weight loss process is attributed to the depolymerization of PP and EVA backbone chains. The temperature of the second step was higher than neat PP and slightly increases with increased cellulose content, suggesting that cellulose can enhance the thermal stability of PP/EVA/cellulose composites. The more thermally stable the materials are, the harder it is to decompose during melt processing or microwave heating.

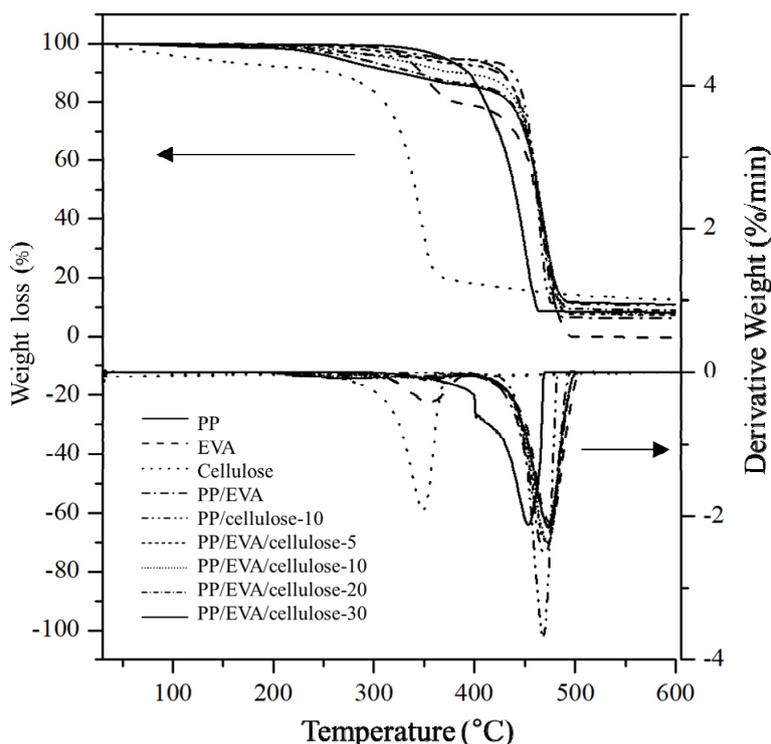


Figure 6 TGA and DTG curves of PP, EVA, PP/EVA and PP/cellulose composites with and without EVA.

Conclusions

In this work, PP/EVA/cellulose composites were prepared and the influence of EVA on their properties was investigated. The 2-phase morphology of EVA distributed in PP matrix were observed by SEM. The existence of EVA can enhance the softness of PP/cellulose composites confirmed by the increased in elongation at break, Impact strength and damping factor of PP/cellulose composite. The storage modulus of composites with EVA was higher than that without EVA. This was influenced by the interfacial interaction between EVA and cellulose. Moreover, the PP/EVA/cellulose composites are more thermally stable than neat PP. The optimum condition of PP/EVA/cellulose was achieved at 10 phr of cellulose which has the elongation at break 5.4 % and the tensile strength value is 15 MPa. The crystallization behavior of all composition composites was not significantly different from neat PP so it can be used at the same processing temperature of PP. In conclusion, PP/EVA/cellulose composite has proven to be a good candidate for chilled food packaging.

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