



# Properties of thermoplastic rice starch composites reinforced by cotton fiber or low-density polyethylene

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## ABSTRACT

Biodegradable polymer was prepared from thermoplastic rice starch (TPRS) plasticized by glycerol. In order to improve poor tensile properties and high water absorption of the TPRS, cotton fiber or low-density polyethylene (LDPE) were added into the TPRS matrix. The effect of maleic anhydride-grafted-polyethylene (MAPE) and vinyltrimethoxy silane (VTMS) compatibilizers on properties of the TPRS/LDPE specimens were also studied. The TPRS/cotton fiber, TPRS/LDPE, TPRS/LDPE/MAPE and TPRS/LDPE/VTMS samples were analyzed for tensile and morphological properties. The results showed that the incorporation of either cotton fiber or LDPE into the TPRS matrix caused the considerable improvement of tensile strength and Young's modulus. Moreover, water absorption of the TPRS samples was clearly reduced by the inclusion of cotton fiber or LDPE. In addition, phase morphology, thermal stability and biodegradability were carried out for different TPRS samples.

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## 1. Introduction

Many efforts have been made to develop biodegradable materials based on starch due to environmental problems resulting from petroleum-derived plastics. Starch is an important productive polysaccharide in plants. Due to its low cost, availability as a renewable resource, biodegradable and innocuous degradation products, it has already been widely researched as an important raw material for packaging, agricultural and biomedical applications.

The possibility of transforming native starch into a thermoplastic starch (TPS) has gained considerable interest (Curvelo, Carvalho, & Agnelli, 2001; Córdoba, Cuéllar, González, & Medina, 2008; Ma, Yu, & Kennedy, 2005; Yang, Yu, & Ma, 2006). Starch is not a true thermoplastic but in the presence of plasticizers at high temperature and under shear, it can readily melt and flow, allowing for its use as extruded or injected material, similar to most conventional synthetic thermoplastic polymers (Forssell, Mikkilä, Moates, & Parker, 1997). Thermoplastic process involves the transformation of the semi-crystalline starch granule into homogenous materials with the destruction of hydrogen bonds between the macromolecules under shear and pressure. In this process, plasticizer is added to the native starch and blended thoroughly and then plasticized, new hydrogen bonds between plasticizer and starch are formed synchronously with the destruction of hydrogen bonds between starch molecules; thus the starch is plasticized (Hulleman, Janssen, & Feil,

1998). Plasticizers increase starch flexibility due to their ability to reduce internal hydrogen bonding between polymer chains while increasing molecular space. Traditional plasticizers are polyols such as glycerol, glycol, sorbitol, sugars and ethanolamine (Huang, Yu, & Ma, 2005; Ma, Yu, & Wan, 2006; Róz, Carvalho, Gandini, & Curvelo, 2006; Teixeira, Róz, Carvalho, & Curvelo, 2007), but the main plasticizer used in thermoplastic starch is glycerol (Curvelo et al., 2001; Ma et al., 2005; Róz et al., 2006; Teixeira et al., 2007). The proportion of plasticizer and its chemical nature strongly influence physical properties of TPS (Róz et al., 2006; Ma et al., 2006; Huang et al., 2005).

Starch from various sources has been studied as TPS, including corn starch (Curvelo et al., 2001; Córdoba et al., 2008; Ma et al., 2005; Róz et al., 2006), potato starch (Thuwall, Boldizar, & Rigdahl, 2006), cassava starch (Müller, Laurindo, & Yamashita, 2009; Teixeira et al., 2007) and wheat starch (Rodríguez-Gonzalez, Ramsay, & Favis, 2004). However, thermoplastic starch prepared from rice starch (TPRS) has not yet been prepared. Rice is the most widely consumed basic food in the world. Each year over 500 million tons of rice is harvested, providing sustenance to many countries and people throughout the world. Rice starch and its major component, amylose and amylopectin, are biopolymers, which are attractive raw materials used in packaging materials.

Generally, TPS still has two main disadvantages, compared to most plastics currently in use, i.e. poor mechanical properties and high water solubility. One approach to increase mechanical properties and reduce water absorption is that the use of natural fibers to reinforce TPS (Curvelo et al., 2001; Ma et al., 2005; Müller et al., 2009). It was reported that (Curvelo et al., 2001) tensile strength and

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modulus of glycerol plasticized corn starch increased by the reinforcement of Eucalyptus pulp. Winceyette fibers were also used to reinforce corn starch plasticized by urea and formamide (Ma et al., 2005). Another way to overcome these drawbacks of TPS is to blend TPS with synthetic polymers (Pierre, Favis, Ramsay, Ramsay, & Verhoogt, 1997). However, TPS and synthetic polymers tend to separate from each other due to incompatible chemical structures.

In this present article, we prepared TPS from rice starch and modified the TPRS matrix by reinforcing either with natural cellulose fibers, i.e. cotton fiber or with synthetic polymer, i.e. LDPE. In the latter case, two different compatibilizers, i.e. maleic anhydride-grafted-polyethylene (MAPE) and vinyltrimethoxy silane (VTMS) were introduced into the TPRS/LDPE samples in order to improve phase compatibility. The effect of cotton fiber, LDPE, MAPE and VTMS contents on mechanical properties, determined from tensile testing, of the TPRS was investigated. In addition, water absorption, morphology, thermal stability and biodegradability of different TPRS samples were examined using percentage weight change, Scanning Electron Microscope (SEM), TG (Thermogravimetric Analyzer) and soil burial test, respectively.

## 2. Experimental

### 2.1. Materials

Rice starch (11.5–13.0% moisture) was obtained from Bangkok Interfood (Bangkok, Thailand), containing 17 wt% amylose and 83 wt% amylopectin. Glycerol (plasticizer) and stearic acid (processing aid) were purchased from Lab System Co. Ltd. (Thailand). Cotton fiber with the aspect ratio of 500:1 were obtained locally and used as-received. Low-density polyethylene (LDPE, LD1905F) with MFI of 5.2 g/10 min (tested at 21.6 N and 190 °C) was obtained from Thai Polyethylene, Co. Ltd. (Bangkok, Thailand). MAPE (MB 100D) and VTMS (A-171) compatibilizers were purchased from Chemical Innovation, Co. Ltd. (Thailand) and Momentive Performance Materials, Co. Ltd. (Thailand), respectively.

### 2.2. Sample preparation

Rice starch and glycerol were pre-mixed in polyethylene bags overnight. The weight ratio of rice starch and glycerol was maintained at 1:1. Each TPRS sample contained 2 wt% of stearic acid by weight of starch. Blending was carried out using a high-speed mixer (Lab-Tech Engineering, Thailand) at the temperature of 170 °C at the speed of 40 rpm for 5 min. in order to obtain a homogeneous material. The processed samples were compressed at 170 °C into 2 mm thick plates. Preliminary results showed that the TPRS could be mixed and processed at the 1:1 rice starch: glycerol ratio and at the processing condition (Patthanapongnun, Gatesuda, & Thammiga, 2007).

The property modification of the TPRS was carried out as followed:

1. By the addition of cotton fiber, the cotton fiber was added into the TPRS at 5 wt%, 10 wt% and 15 wt% by weight of starch and glycerol.
2. By the addition of LDPE, the LDPE was added into the TPRS at 5 wt%, 10 wt%, 15 wt%, 20 wt%, 25 wt% and 30 wt% by weight of starch and glycerol.
3. By the addition of compatibilizers, the MAPE or VTMS compatibilizer was added into the TPRS/LDPE at 1 wt%, 3 wt% and 5 wt% by weight of LDPE.

### 2.3. IR spectroscopic study

FTIR spectra of different TPRS samples were recorded on a Spectrum 2000 GX spectrometer (PerkinElmer, USA) using KBr disk technique with a resolution of 4 cm<sup>-1</sup> in a spectral range of 4000–600 cm<sup>-1</sup> using 16 scans per sample.

### 2.4. Tensile properties

Tensile tests were conducted according to ASTM D-638 at the temperature of 23 ± 1 °C and relative humidity of 60 ± 5%. The tensile measurements from dumbbell specimens were carried out using Universal Testing Machine (LLOYD Instrument, LR 5K, UK) operated by WINDAP software with 100 N load cell and a crosshead speed of 40 mm/min. It should be noted that the mechanical property results of the TPRS samples were obtained by averaging from ten independent tested specimens.

### 2.5. Morphology

A LEO 1455 VP scanning electron microscopy (Carlzeiss, Germany) was employed to study the morphology of the TPRS with and without the modification by cotton fiber or LDPE. The samples were immersed in a nitrogen liquid before fractured. After that the samples were sputter-coated with a thin layer of gold to prevent electrical charging during the observation.

### 2.6. Water absorption

Newly prepared samples were dried at 105 °C for 12 h and then stored at 50% relative humidity at the temperature of 30 ± 2 °C. The 50% relative humidity was obtained using a saturated solution of CaCl<sub>2</sub> in a closed vessel. The amount of water absorbed by the samples was measured until the constant weight was reached. The percentage water absorption was calculated as followed:

$$\text{water absorption} = \frac{W_2 - W_1}{W_1} \times 100 \quad (1)$$

where  $W_2$  and  $W_1$  were the final weight and the dried weight of the sample, respectively.

### 2.7. Thermal property

TG thermograms were recorded by Thermogravimetric analyzer (PerkinElmer, Pyris 1, USA). The TPRS samples were tested under nitrogen atmosphere using a temperature range of 50–600 °C and a heating rate of 10 °C/min. Thermal degradation temperature ( $T_d$ ) was reported by the onset degradation temperature where the weight loss started to occur. Percentage weight loss was obtained from the onset decomposition temperature determined only for the starch decomposition.

### 2.8. Soil burial test

The TPRS samples with the dimension of 20 mm × 50 mm were burial under soil surface of approximately 10 cm. The pH and temperature of the soil were maintained at 7 and 32 ± 2 °C, respectively. The water content of the soil was in the range of 10–20% and the weight change was recorded for 21 days. Average percentage weight change was recorded from three independently tested samples.

### 3. Results and discussions

#### 3.1. IR spectroscopic study

The modification of the TPRS matrix with cotton fiber and LDPE reinforcing agents including MAPE and VTMS compatibilizers can be examined using IR technique. It can be seen in Fig. 1(a) that the pure TPRS exhibits IR main peak positions in the range of  $3500\text{--}3250\text{ cm}^{-1}$ ,  $3000\text{--}2800\text{ cm}^{-1}$ ,  $1475\text{--}1450\text{ cm}^{-1}$ ,  $1275\text{--}1070\text{ cm}^{-1}$ , and  $1200\text{--}1000\text{ cm}^{-1}$ , representing O–H stretching, C–H asymmetric stretching of  $-\text{CH}_2-$ ,  $-\text{CH}_2-$  deformation, C–O–C stretching and C–O–H stretching, respectively (Bower & Maddams, 1989). As expected, both the TPRS and cotton fiber

composed of the same cellulose chemical structures displayed these overlapped IR wave numbers. Furthermore, the peak position in the range of  $1950\text{--}1600\text{ cm}^{-1}$  (assigned to C=O stretching) obtained from stearic acid component. The difference in IR spectra between the TPRS and TPRS/cotton fiber is that the O–H stretching peak is narrower in the TPRS/cotton fiber, possibly due to the rearrangement of hydrogen bonds between the TPRS and cotton fiber. The wave number in the range of  $1000\text{--}850\text{ cm}^{-1}$  in the TPRS was attributed to C–C stretching; whereas, the  $722\text{ cm}^{-1}$  in the TPRS/cotton fiber was assigned to  $-\text{CH}_2-$  rocking (Bower & Maddams, 1989).

Fig. 1(b) shows IR spectra of the TPRS/LDPE, TPRS/LDPE/MAPE and TPRS/LDPE/VTMS. The peak position of  $1646\text{ cm}^{-1}$ , assigned

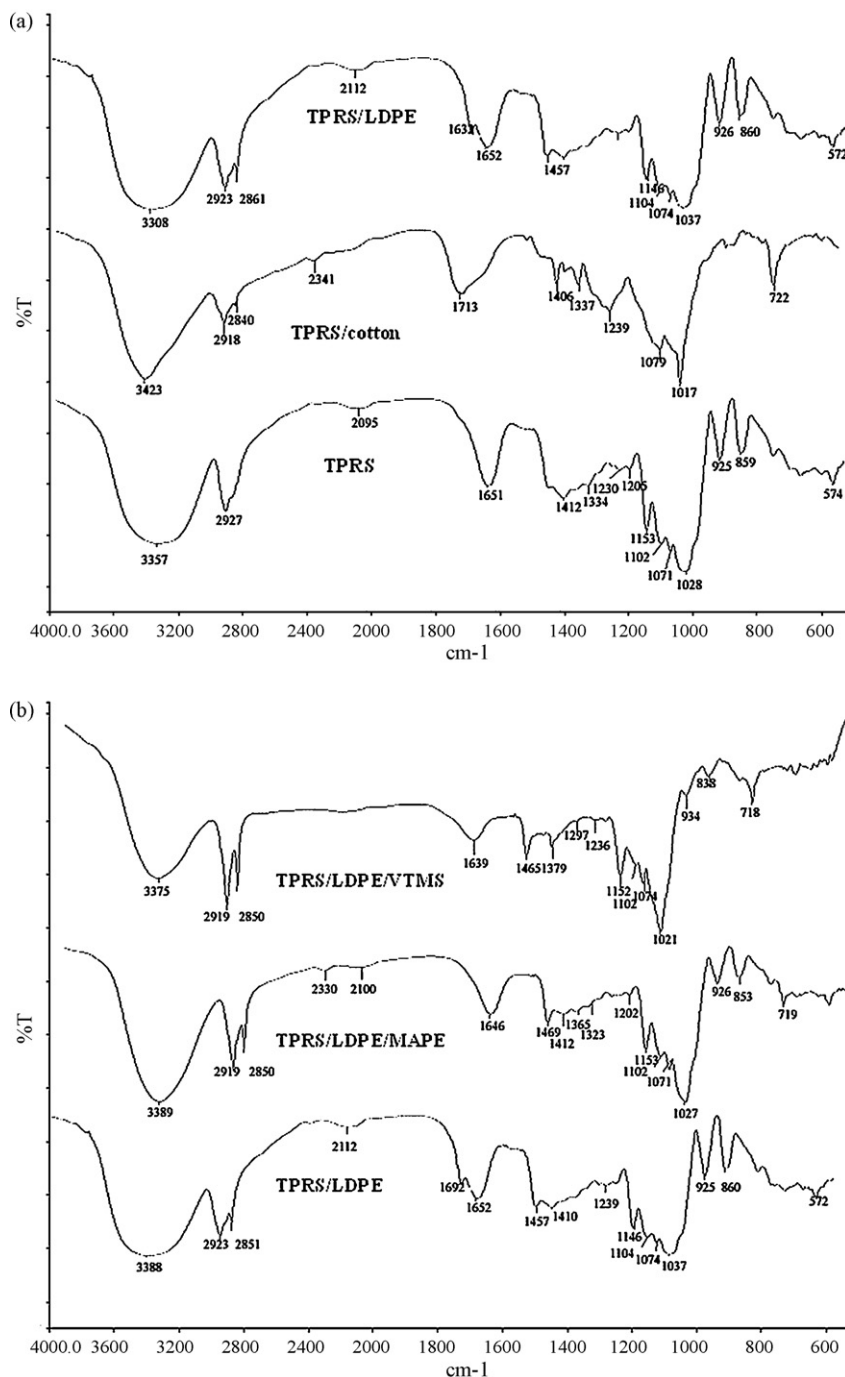


Fig. 1. (a) IR spectra of TPRS, TPRS/cotton fiber and TPRS/LDPE samples. (b) IR spectra of TPRS/LDPE, TPRS/LDPE/MAPE and TPRS/LDPE/VTMS samples.

for C=O stretching and appeared from not only stearic acid but also ester group formation between the TPRS and MAPE, was overlapped. Nevertheless, the ester bond formation could be identified by the wave number at  $1027\text{ cm}^{-1}$  represented C–O stretching from ester group (Bower & Maddams, 1989). In the case of VTMS compatibilizer, new peak position at  $1021\text{ cm}^{-1}$ , indicating of Si–O–C stretching (Bower & Maddams, 1989) was found, demonstrating of new chemical bond formed by the addition of VTMS compatibilizer.

### 3.2. Tensile properties

#### 3.2.1. Effect of cotton fiber and LDPE

A biodegradable polymer must withstand normal stress encountered during its application. It is expected that the TPRS and cotton fiber are compatible due to the similarity of the main cellulose structures composed of hydroxyl functional groups; however, LDPE chemical structure is clearly different from those of starch and cotton fiber.

It can be seen in Fig. 2(a) that tensile strength of the pure TPRS was approximately 0.3 MPa; however, tensile strength of the TPRS clearly increased twice by the incorporation of 10% cotton fiber. The results suggest that cotton fiber can reinforce the TPRS matrix due to the fiber structure and also the phase compatibility. On the other hand, greater weight % of cotton fiber (15%) resulted in the decline of tensile strength, possibly due to the discontinuity of the TPRS matrix. On the contrary, the gradual improvement of tensile strength was found when the contents of LDPE were in the range of 0–30%, possibly because LDPE can flow easier than the TPRS. The tensile strength of the TPRS/LDPE could probably resulted from more dominant property of LDPE since LDPE shows higher tensile strength than that of the TPRS.

Fig. 2(b) shows the relationship between strain at break of the TPRS with the addition of different amounts of cotton fiber or LDPE. It can be observed that cotton fiber caused the decrease in strain at break of the TPRS matrix gradually; however, LDPE continually increased strain at break of the TPRS. The former case could be due to the high crystallinity of the fibers; whereas, the rise of strain at break in TPRS/LDPE sample could be affected by the LDPE flexibility and toughness.

Young's modulus of the TPRS can also be improved by the incorporation of cotton fiber or LDPE as presented in Fig. 2(c). Similar to tensile strength, Young's modulus of the TPRS enlarged and it reached the maximum value at 10% cotton fiber; then, it tended to clearly drop. By the introduction of LDPE, Young's modulus enhanced greatly with the increased content of LDPE.

A considerable increase in tensile strength and Young's modulus of the TPRS/5% cotton fiber and TPRS/10% cotton fiber indicates that the TPRS is suited as the matrix for natural cellulose fibers. This is due to the remarkable adhesion of the fiber-matrix interface caused by the chemical similarity of rice starch and cotton fiber. The tensile results were in accordance with Curvelo et al. (2001) who studied the thermoplastic corn starch-cellulose composites. It was found that tensile strength and Young's modulus of the composites greatly increased with the addition of the Eucalyptus bleached wood pulp. It should be noted that the addition of 5% or 10% cotton fiber gives higher values of tensile strength and Young's modulus of the TPRS than those of 5% or 10% LDPE. This implies better reinforcement by cotton fiber in the TPRS matrix.

#### 3.2.2. Effect of MAPE and VTMS compatibilizers

Due to the incompatibility between the hydrophilic rice starch and the hydrophobic LDPE, MAPE and VTMS compatibilizers were introduced into the TPRS/LDPE system. Fig. 3 shows the relationship between tensile properties of the TPRS/LDPE samples with different contents of MAPE. It can be seen in Fig. 3(a) that tensile strength of the TPRS/LDPE increases with the addition of MAPE compatibilizer.

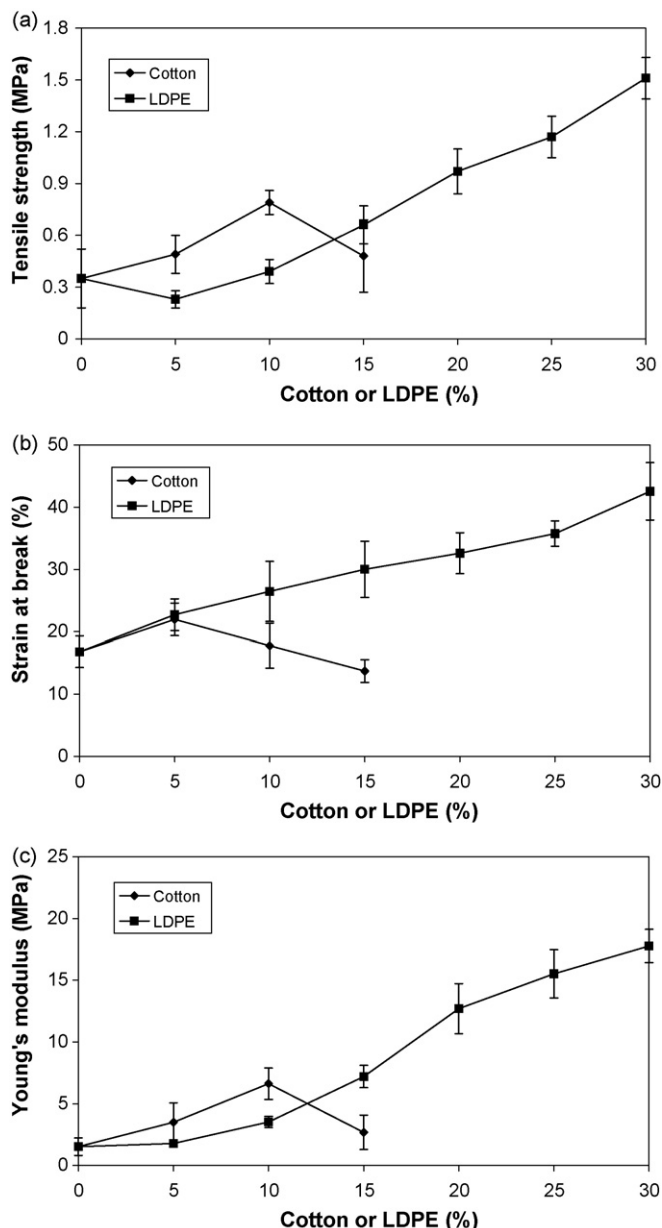


Fig. 2. Mechanical properties of TPRS/cotton fiber and TPRS/LDPE (a) Tensile strength (b) %Elongation at break and (c) Young's modulus.

The maximum tensile strength could be obtained when 3% MAPE was used for different contents of LDPE. On the contrary, strain at break in Fig. 3(b) decreased when 1% of MAPE was added into the TPRS/LDPE samples; after that strain at break tended to be maintained by the use of 3% and 5% MAPE. Similar to tensile strength, Young's modulus of the TPRS enlarged with the increased amount of MAPE and the maximum value could be obtained with 3% MAPE as presented in Fig. 3(c). The improvement of tensile properties could be due to the chain entanglement between LDPE and polyethylene part of MAPE molecule as presented in Fig. 4. In addition, strong ester linkage could form between maleic anhydride part of MAPE and hydrophilic rice starch (as previously shown in Fig. 1(b)) so that the incompatible part between the hydrophilic rice starch and the hydrophobic LDPE could be linked and strengthened (Fig. 4).

By the use of VTMS compatibilizer, it can be seen in Fig. 5 that tensile strength and Young's modulus of the TPRS/LDPE samples could be improved. However, strain at break tended to be constant with different amounts of the VTMS compatibilizer. Similar

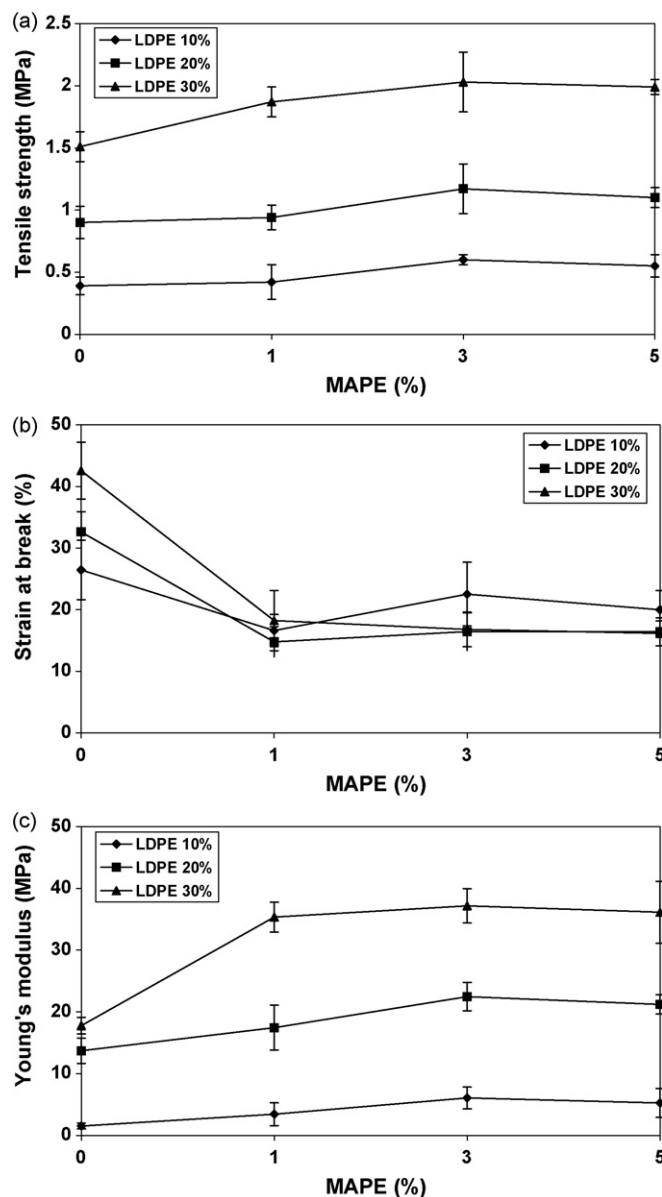


Fig. 3. Mechanical properties of TPRS/LDPE with different amounts of MAPE compatibilizer.

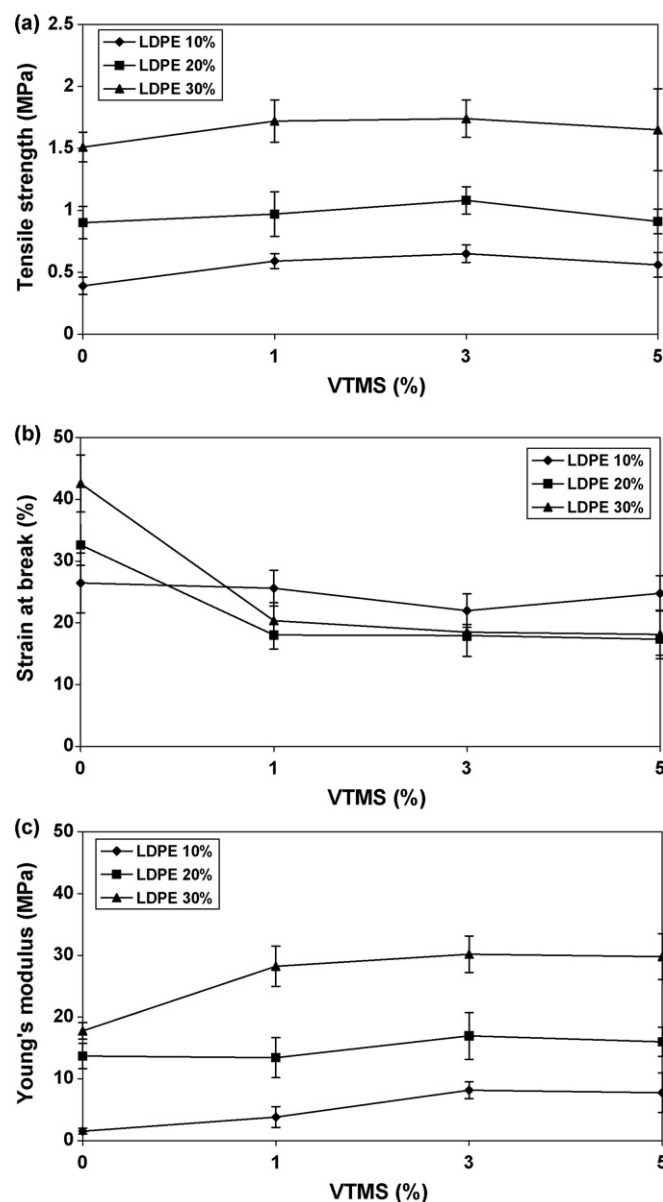


Fig. 5. Mechanical properties of TPRS/LDPE with different amounts of VTMS compatibilizer.

to MAPE compatibilizer, 3% VTMS provided the maximum tensile properties of the TPRS/LDPE. The schematic diagram of the interactions between the TPRS and LDPE with the use of VTMS is presented in Fig. 6. The improvement of tensile properties of the TPRS/LDPE/VTMS samples was due to Si–O–Starch chemical

bond formed between the silanol group of VTMS and hydroxyl group of rice starch (Fig. 1(b)). Besides, double bond in vinyl part in the VTMS molecule could probably be dissociated at high processing temperature so this possibly led to chemical bond between the vinyl part of the VTMS and LDPE molecule. Comparison

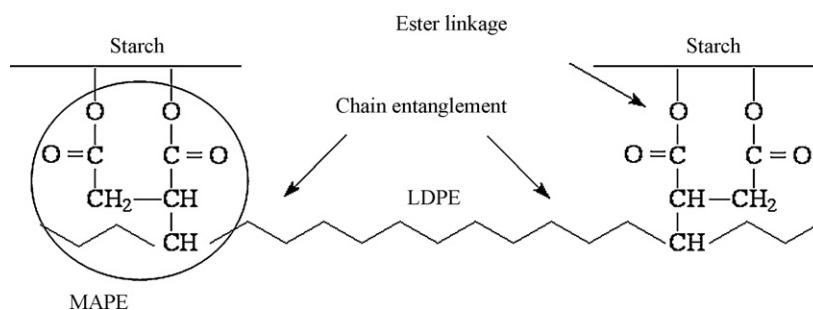


Fig. 4. Schematic diagram of interaction in TPRS/LDPE/MAPE.



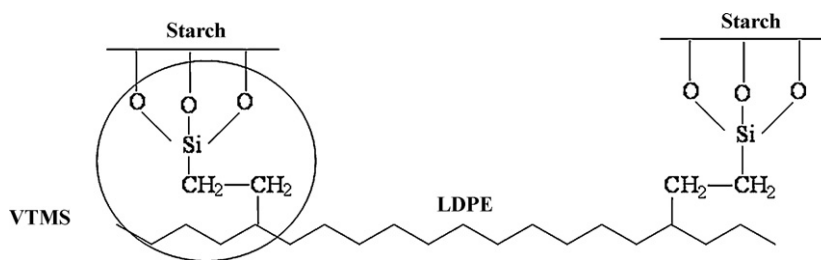


Fig. 6. Schematic diagram of interaction in TPRS/LDPE/VTMS.

between the TPRS/LDPE samples added with either MAPE or VTMS compatibilizer, it was found that the use of MAPE led to greater mechanical properties of the TPRS samples than those using VTMS and the best mechanical properties were obtained in the TPRS/30%LDPE/3%MAPE. However, when the modification of the TPRS with 10% cotton fiber or with 10%LDPE/3%MAPE was compared, it was found that the highest mechanical properties were obtained in the TPRS/10% cotton fiber (Figs. 2 and 3).

### 3.3. Morphology

Morphological structure of polymer is a very important characteristic because it ultimately determines many polymer properties. SEM micrographs of cryofractured surfaces of fragile test specimens are shown in Fig. 7. Rough surface of the glycerol plasticized TPRS could be observed as presented in Fig. 7(a). For the TPRS reinforced by cotton fiber, the SEM micrograph in Fig. 7(b) showed long cotton fiber embedded into the TPRS matrix and the fiber surface was wetted by the TPRS matrix.

On the contrary, the addition of LDPE into the TPRS matrix resulted in the poor phase distribution between TPRS and LDPE because of the differences in polarity. It can be seen in Fig. 7(c) that the hydrophilic TPRS phase tends to separate from the hydrophobic LDPE phase, representing by the smoother surface region in the micrograph.

Fig. 7(d) and (e) present the SEM micrographs of the TPRS/LDPE samples with MAPE and VTMS compatibilizers, respectively. It can be observed that similar phase morphology could be obtained with the use of either MAPE or VTMS compatibilizers. Phase separation between the TPRS and LDPE could not be observed as can be seen in the TPRS/LDPE specimen in Fig. 7(c). This phase morphology is the good evidence of phase compatibility of the TPRS and LDPE by the use of MAPE or VTMS compatibilizers.

### 3.4. Water absorption

Water sensitivity is another important criterion for many applications of starch products. The results of water absorption of different TPRS specimens performed at the temperature of  $30 \pm 2^\circ\text{C}$

and 50% RH are shown in Fig. 8. It can be seen that it took about 3 days for different TPRS samples with or without modification to reach absorption equilibrium. The equilibrium water contents for the pure TPRS, TPRS/10% cotton and TPRS/10% LDPE were approximately 380%, 305% and 225%, respectively.

It was found that water absorption of the TPRS was clearly reduced with the incorporation of cotton fiber because of the less hydrophilic character of the fibers in comparison to starch hydrophilic property (Fig. 8). Moisture absorption of cotton fiber is approximately 7.0–8.0%; whereas, it is in the range of 11.5–13.0% for the rice starch (Li, Shoemaker, Ma, Moon, & Zhong, 2008; Morton & Hearle, 1997).

As expected, the incorporation of LDPE into the TPRS matrix could dramatically restrain the water absorption of the TPRS matrix and reduced the water sensitivity of the TPRS due to the hydrophobic characteristic of LDPE. Greater content of LDPE caused the decline of water absorption of the TPRS/LDPE specimens. It should be noted that; although, cotton fiber shows more hydrophilic nature than LDPE, water absorption of the TPMBS can be decreased by the incorporation of cotton fiber.

When MAPE or VTMS compatibilizer were added into the TPRS/30% LDPE samples, it was found that water absorption tended to increase slightly due to the more hydrophilic nature of the MAPE and VTMS compatibilizers, compared with LDPE.

### 3.5. Thermal properties

In thermogravimetric analysis, the loss in mass due to volatilization of the degradation products is monitored as a function of temperature. The thermogravimetric (TG) and derivative thermogravimetric (DTG) curves for different TPRS samples are shown in Table 1 and Fig. 9. The weight loss of the samples in the first step was mainly referred to water loss.

It can be also seen in Table 1 and Fig. 9 that the TPRS shows the onset decomposition temperatures ( $T_d$ ) at  $178^\circ$  and  $301^\circ\text{C}$  due to the decomposition temperatures of glycerol and rice starch, respectively. The modification of the TPRS with the cotton fiber causes the improvement of  $T_d$  of the TPRS (Table 1).  $T_d$  values of the TPRS were  $305^\circ\text{C}$  and  $303^\circ\text{C}$  by the use of 5% and 10%

Table 1

Onset degradation temperatures and percentage weight losses of different TPRS samples obtained from TG and DTG thermograms.

Samples	Onset degradation temperatures ( $^\circ\text{C}$ )				Weight loss (%)
	Zone 1	Zone 2	Zone 3	Zone 4	
TPRS	178.2	300.9	–	–	52.2
TPRS/5% cotton	190.3	304.6	397.6	–	31.7
TPRS/10% cotton	184.9	303.2	407.8	–	28.0
TPRS/10% LDPE	162.9	293.1	–	448.4	50.6
TPRS/20% LDPE	156.8	294.7	–	429.4	45.5
TPRS/30% LDPE	154.9	300.6	–	447.3	39.9
TPRS/30% LDPE/5% MAPE	155.9	301.0	–	448.3	38.2
TPRS/30% LDPE/5% VTMS	167.5	306.1	–	440.6	29.8

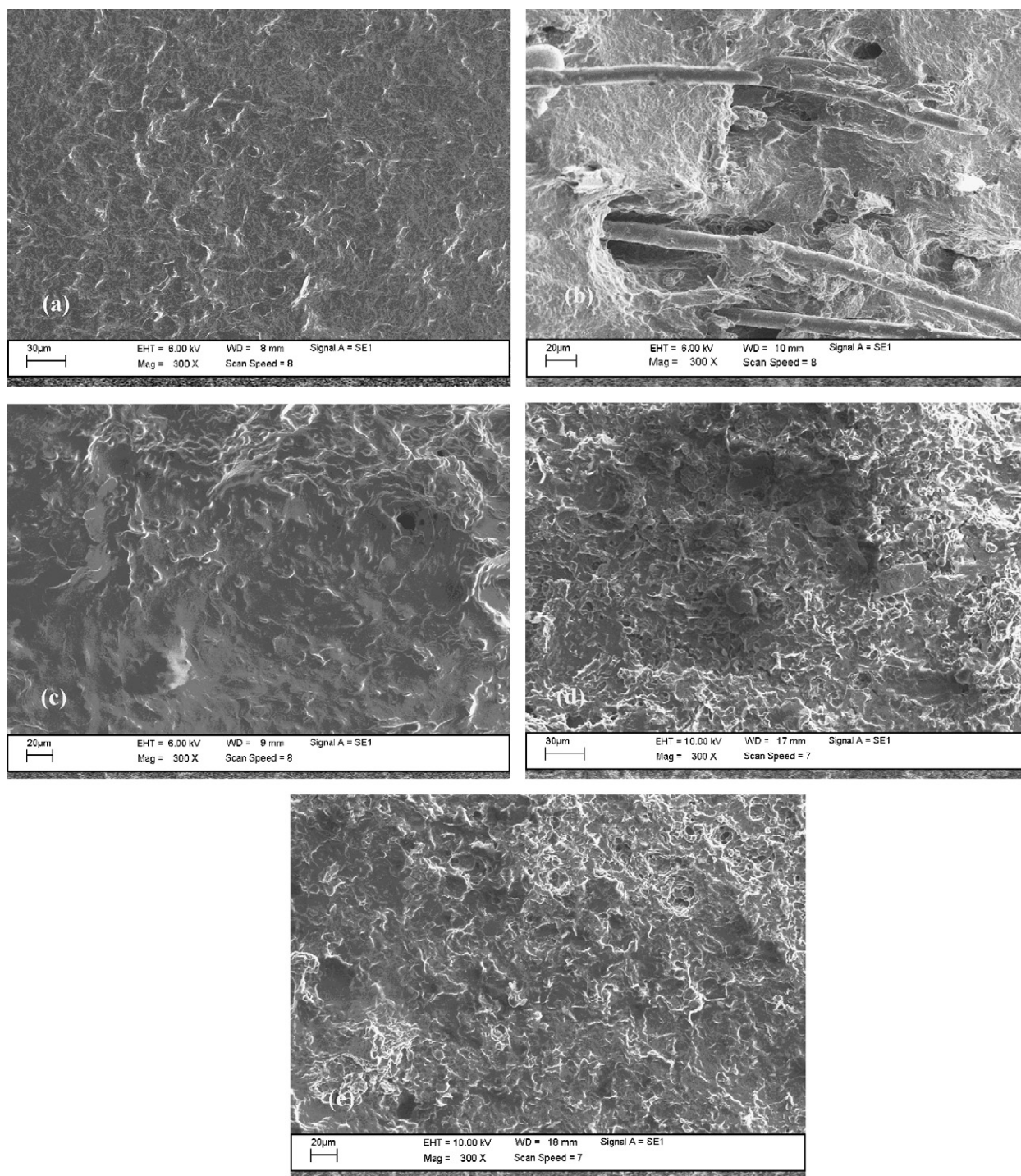


Fig. 7. SEM micrographs of (a) TPRS (b) TPRS/cotton fiber (c) TPRS/LDPE (d) TPRS/LDPE/MAPE and (e) TPRS/LDPE/VTMS.

cotton fiber, respectively. This could be because the TPRS is phase compatible with cotton fiber by the hydrogen bond linkages. In addition,  $T_d$  of cotton fiber appeared approximately in the range of 398–408 °C as the third step in the thermograms. However, the weight loss at onset  $T_d$  was found to be 52.2%, 31.7% and 28.0% for the TPRS, TPRS/5% cotton and TPRS/10% cotton, respectively. The decrease in percentage weight loss implies that thermal stability of the TPRS/cotton samples tend to increase, due to good adhesion between the TPRS and cotton fiber. Similar decrease in weight loss at onset temperature by the incorporation of fibers into TPS was also reported (Curvelo et al., 2001; Ma et al., 2005).

Nevertheless, the incorporation of LDPE into the TPRS tended to decrease  $T_d$  of the rice starch.  $T_d$  values of the TPRS/LDPE were found to be slightly lower than  $T_d$  of the pure TPRS. This could be possibly because of the different phase incompatibility between TPRS and LDPE. Besides, the  $T_d$  of LDPE component was obtained in the range of 429–448 °C. It was also found in Table 1 that percentage weight losses were 52.2%, 50.6%, 45.5% and 39.9% for the TPRS, TPRS/10% LDPE, TPRS/20% LDPE and TPRS/30% LDPE, respectively. The results clearly indicate that the thermal stability of the TPRS is improved by the addition of LDPE. Nevertheless, the incline in percentage weight loss or the enhancement in thermal stability

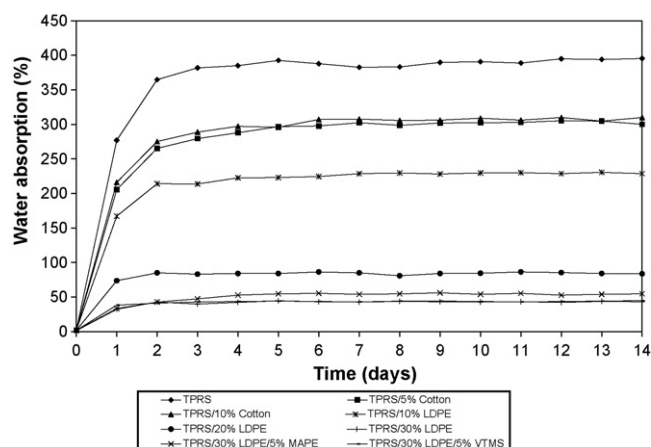


Fig. 8. The relationship between water absorption and time in different TPRS samples at 50% RH.

in TPRS is dominated by the addition of cotton fiber than those of LDPE.

By the use of MAPE and VTMS compatibilizers, it can be seen in Table 1 and Fig. 9 that  $T_d$  of the TPRS/LDPE samples slightly

is increased. It can be due to the improved phase compatibility between the TPRS and LDPE as previously shown in Fig. 7. Similar to the TPRS/cotton fiber samples, the slightly increase in  $T_d$  shows the increase in thermal stability with the presence of MAPE or VTMS compatibilizers in the TPRS/LDPE samples. Percentage weight loss was found to be 39.9%, 38.2% and 29.8% for the TPRS/LDPE, TPRS/LDPE/MAPE and TPRS/LDPE/VTMS, respectively. The decrease in percentage weight losses shows the slight improvement in thermal stability with the addition of MAPE or VTMS compatibilizers.

### 3.6. Soil burial test

Biodegradable properties of different TPRS specimens can be determined by the soil burial test. It can be seen in Fig. 10 that all of the TPRS samples, especially the TPRS and TPRS/cotton fiber specimens, show the increment in weight after a few days of soil immersion due to the water uptake from the soil; after that, the samples weights tend to decrease and then the samples start to degrade. The pure TPRS sample degraded within 6 days of soil burial test. It should be noted that the TPRS/cotton fiber samples can rapidly absorb water because of the hydrophilic nature of not only the TPRS but also cotton fiber. Percentage weight change in the

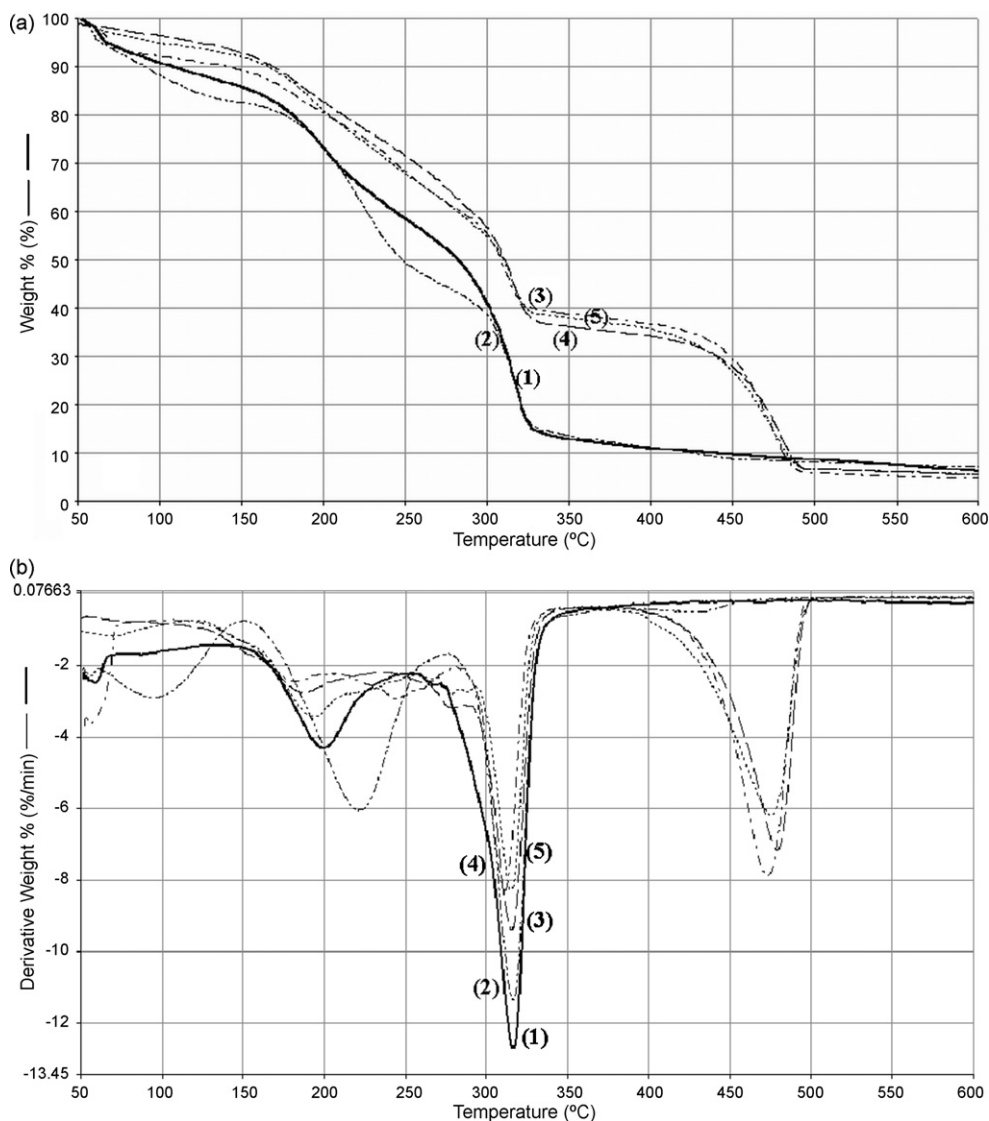


Fig. 9. (a) TG and (b) DTG thermograms of (1) TPRS (2) TPRS/cotton fiber (3) TPRS/LDPE (4) TPRS/LDPE/MAPE and (5) TPRS/LDPE/VTMS.



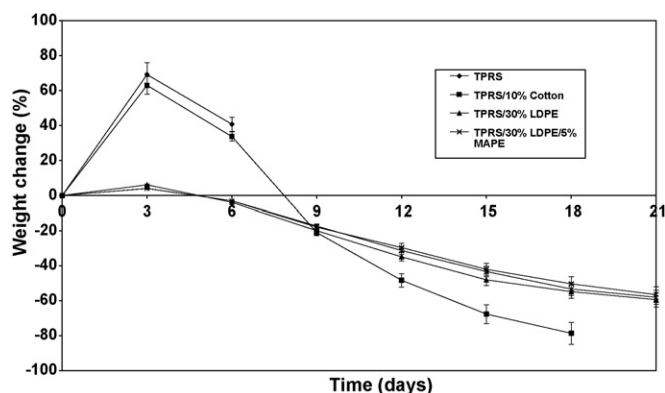


Fig. 10. Percentage weight changes of TPRS samples under soil burial test

TPRS/cotton fiber sample seems to lower than that of the pure TPRS. However, the maximum decrease in weight change was found in the TPRS/cotton fiber sample, presenting the ease of degradability, i.e. the sample can degrade within 18 days of testing. On the contrary, more difficult degradable samples can be clearly obtained in the TPRS/LDPE specimens due to the hydrophobic characteristic of LDPE part. However, the TPRS/LDPE samples with the addition of the MAPE or VTMS compatibilizers caused less percentage weight change than that of the TPRS/LDPE sample without the compatibilizer due to the improved phase compatibility between the TPRS and LDPE.

#### 4. Conclusions

Due to the poor tensile properties and high water absorption of the TPRS, the modification of the TPRS was carried out using cotton fiber or LDPE. It was found that the TPRS/cotton fiber and the TPRS/LDPE presented greater mechanical properties and lower water uptake. By the use of MAPE and VTMS compatibilizers in the TPRS/LDPE, the TPRS/LDPE/MAPE and TPRS/LDPE/VTMS specimens also illustrated the improved tensile properties but slight increase in water absorption, as compared to the TPRS/LDPE. From SEM micrographs, cotton fibers were embedded and wetted by the TPRS matrix; however, the TPRS with LDPE showed the considerable phase separation. However, the phase separation could be reduced by the addition of the MAPE or VTMS compatibilizers. Besides, thermal stability of the TPRS increased significantly with the application cotton fiber. As expected, the faster biodegradability was found in the TPRS/cotton fiber than in the TPRS/LDPE. From this study, the optimum mechanical, thermal, water absorption and

biodegradable properties were obtained from the TPRS with the addition of 10% cotton fiber.

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