

## Effect of different loading low density polyethylene (LDPE) on thermal and mechanical properties of Tapioca starch-based plastic composite

Norin Zamiah Kassim Shaari\*, Mazlan Bairik, Junaidah Jai

Faculty of Chemical Engineering, Universiti Teknologi MARA, Selangor, Malaysia

\*Corresponding email: [norinzamiah@uitm.edu.my](mailto:norinzamiah@uitm.edu.my)

### Abstract

Effect of incorporating different loading of low densities polyethylene (LDPE) on the mechanical and thermal properties of the plastic composite made from tapioca starch was investigated. The compound formulation was done in the internal mixer and the lump produced was crushed. Then, the crushed sample was compressed to form a dumbbell-shaped in a compression molding equipment, heated at 140°C. The sample was subsequently analyzed for tensile property, thermal stability and functional groups. Results show that the incorporation of LDPE improves thermal and mechanical stabilities of the plastic composite. However, too much LDPE causes lower ductility of the composites. LDPE-15 was found as the best formulation since it has moderate tensile strength with good elongation.

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### 1.0 Introduction

Due to the global concern on the use of biodegradable and compostable materials, the search of material to replace partially or fully the petroleum-based plastics is widely investigated (Seligra et al., 2016). Petroleum-based plastics with its high resistance to microorganism and other natural degradation forces have made them become sources of environmental problem and also contribute to a shortage of land for landfills (Shah et al., 2008). To overcome this problem, the use of natural biopolymer such as starch has become an attractive raw material for plastic composite because it is an inexpensive and biodegradable material (Yunos & Rahman, 2011). However, due to it contains multi hydroxyl group, modification of the structure by incorporating plasticiser should be conducted. It will make the starch as true thermoplastic, where it can be heated at high temperature (90–80°C), and can be used as an injection, extrusion, or blow moulding material (Ma et al., 2007).

Polyols such as glycerol are the most widely used plasticizer. Plasticizer increases the mobility of polymer chains in starch films to make the polymer structure more elastic, and the material's intermolecular forces and glass transition temperature also increases (Bilck et al., 2015).

However, the incorporation of glycerol causes recrystallization (retrogradation) of the thermoplastic starch on storage. In order to prevent retrogradation, a nontoxic plasticizer such as citric acid is used (Shi et al., 2007). Shi et al. (2007) has conducted a study by incorporating both glycerol and citric acid as coplasticizers. Result showed that the increase concentration of citric acid causes improvement on the thermal stability of the plastic film. Citric acid as the cross linker reduces the tendency of retrogradation of the film by forming strong hydrogen and ester bonds with starch molecules (Yu et al., 2005). But the incorporation of citric acid could reduce the tensile strength of the film but improves the elongation at break (Yu et al., 2005). It is therefore it must be accompanied with plasticizer such as glycerol in the plastic composite's formulation.

Plastic composite from starch with the presence of glycerol and citric acid alone still inferior in properties as compared to plastics made from petroleum. This is due to its large affinity to water that makes it unsuitable to various applications including straw and food containers (Ogunrinola & Akpan, 2018). Therefore, a blend with petroleum-derived material such as polyethylene (LDPE) is conducted in this study. This blend polymer is expected to not jeopardize the biodegradability of the starch film. LDPE (low density

polyethylene) is an essential material to be used for power transmission, food packaging, consumer goods, electronics, household goods, industrial storage and transportation industries (Mahl et al., 2017).

As reported by Chandra & Rustgi (2008), when biodegradable component (e.g. starch) present in a composite starch is removed by microorganism upon disposal, the inert component in the LDPE would disintegrate and disappear. Thus, the composite which contains high percentage of starch as compared to LDPE is more environmentally friendly and biodegradable.

The aim of this study is to evaluate the thermal and mechanical properties of starch-LDPE composite at various loadings of LDPE. The optimum loading of LDPE is used for subsequent study on the biodegradability of the plastic composite.

## 2.0 Methodology

### 2.1 Material

The starch used for the preparation of the composites was the tapioca starch (Brand: Cap Kapal ABC). Glycerol, citric acid and analytical grade of low densities polyethylene (LDPE) were purchased from Sigma Aldrich Malaysia.

### 2.2 Mixing process

Fifty grams of starch, 10 g LDPE (20% wt./wt. starch), 20 g glycerol, and 3 mL citric acid were premixed manually. Then the plasticization and melting of the mixture were carried out in the twins screw internal mixer, model: Polylab DS System, brand: Thermo Haake, Germany). The temperature was set at 140 °C and screw velocity of 50 rpm. The sample was mixed for 15 min in the mixer. Five different formulations were prepared as shown in Table 1 by varying the LDPE loading in each formulation.

**Table 1:** Formulations of plastic composite

Code	Glycerol (g)	Citric acid (mL)	Starch (g)	LDPE (g)	% LDPE (wt./wt. starch)
LDPE-10	20	3	50	10	20
LDPE-15	20	3	50	15	30
LDPE-20	20	3	50	20	40
LDPE-25	20	3	50	25	50
LDPE-30	20	3	50	30	60

### 2.3 Compression moulding

After the mixing process, the lump of plastic composite was then crushed using a crusher. Then, test specimens were moulded to a dumbbell-shaped by using a hot press machine, (Cometech: QC602). The moulding temperature was set at 140 °C, for 3–5 min preheating time and 15 min compression time.

### 2.4 Characterisation

#### 2.4.1 Tensile test

This test was made according to the technical standard method ASTM D638 using a universal testing machine (Tinius Olsen: H50KT). The load cell used of 2.5 kN and crosshead speed was set to 20 mm/min. For each sample, 3 specimens were tested and the average value was recorded.

#### 2.4.2 Thermogravimetric Analysis (TGA)

This analysis was carried out using Mettler Toledo TGA instrument under nitrogen atmosphere with flow rate of 50 mL/min and heating rate of 10 °C/min. The heating temperature was from 30 to 650 °C.

#### 2.4.3 Fourier-transform infrared spectroscopy (FTIR)

FTIR analysis was conducted using Fourier transform infrared (FTIR) spectroscopy with model Spectrum One, manufactured by Perkin Elmer. The measurement was in the range from 500 to 4000  $\text{cm}^{-1}$ .

## 3.0 Result and Discussion

### 3.1 FTIR

The IR spectrum for five samples is shown in Fig.1. By increasing the content of LDPE in the composite's formulation, the intensity of spectra at 2917  $\text{cm}^{-1}$  and 2847  $\text{cm}^{-1}$  increase. The same finding was observed by Pushpadass et al. (2010), where the functional group related with LDPE appeared in the same wavelength that obviously the LDPE is dispersed uniformly in the starch matrix. The peak at 3307  $\text{cm}^{-1}$ , which correspond to O–H bond from glycerol, is not much affected with the concentration of LDPE. Another significant band which shows an increasing in intensity with the increase in LDPE is at 1020  $\text{cm}^{-1}$ , which corresponds to the amorphous content of the composite. It shows that the crystalline structure of the starch was destroyed during the mixing process (Pushpadass et al., 2010).

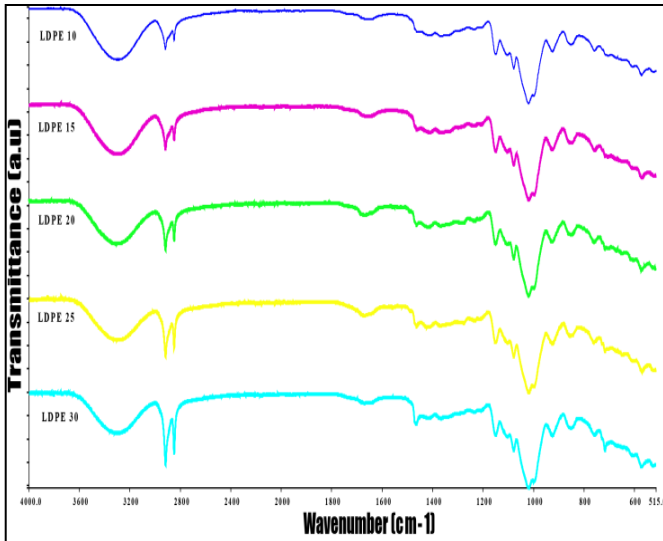


Fig. 1: FTIR spectra for various formulations of plastic composites

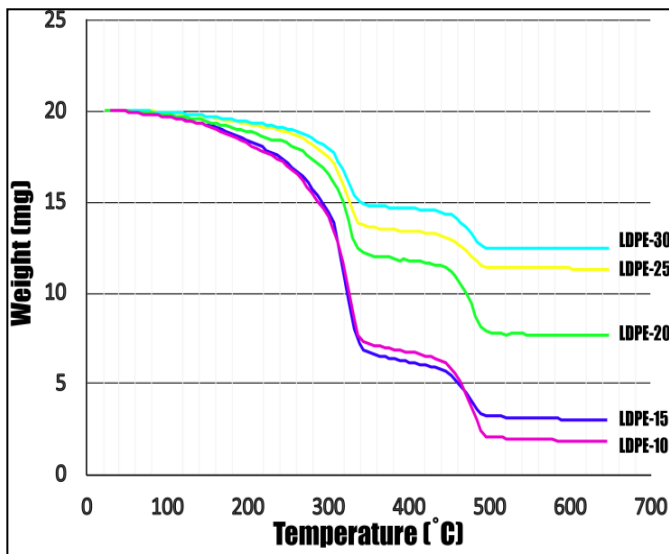


Fig. 2: TGA curves for various formulations of polymer composites

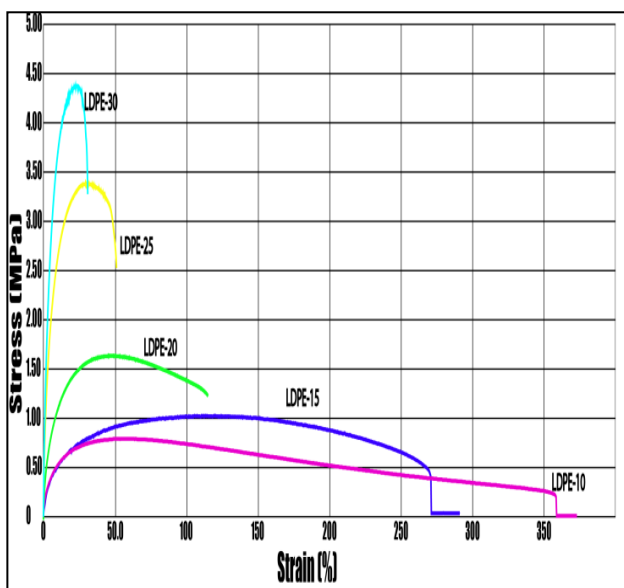


Fig. 3: Stress vs. strain curves of plastic composites

### 3.2 Thermogravimetric analysis (TGA)

Fig. 2 shows the TGA curves for all samples with various loading of LDPE. All samples exhibit four weight loss profiles. From 30 to 160 °C, the weight loss was attributed to the evaporation of free water. The second weight loss profile from 160 to 300 °C corresponds to loss of water. The third weight loss from 300 to 450 °C is attributed to the decomposition of the starch (Mendes et al, 2015). The last weight loss profile from 450 to 500 °C corresponds to the LDPE (Yusof et al, 2018). Based on Fig. 2, plastic composites with higher loading of LDPE (40, 50, 60% wt./wt. starch) have better thermal stability due to decompose at slightly higher temperature in the last profile. Furthermore, the decomposition process was not rapid to leave higher weight residues as compared to the composites with LDPE 20 and 30% wt./wt. starch.

### 3.3 Stress-strain diagram

Fig. 3 shows the stress-strain curves for all plastic composites, and the extracted data was displayed in Table 2. Although the tensile strength of composites with LDPE 40, 50, 60% wt./wt. starch are higher as compared to that with 10 and 15% LDPE, the composites are slightly brittle and easily break. This phenomenon was observed through lower strain exhibited by the composites. Composites with LDPE 20 and 30% wt./wt. starch, on the other hand, have high elongation, which show ductility behavior. Ductility is important criteria of the plastic composite because it will make the processing ability of the composite into certain shapes easier (Reddy & Yang, 2010).

Table 2: Tensile properties of plastic composites

Code	Tensile strength (MPa)	Strain (%)
LDPE-10	0.792	374
LDPE-15	1.030	263
LDPE-20	1.640	115
LDPE-25	3.380	51
LDPE-30	4.370	31

### 4.0 Conclusion

A series of plastic composite from tapioca starch, glycerol, citric acid and LDPE was successfully prepared and tested. The increase loading of LDPE into the composites' formulation has improved the

thermal stability of the composites but contributed to the brittleness of the structures. Therefore, it was observed that LDPE-15 with 30% wt./wt. starch is the optimum amount as the plastic composite yield moderate tensile strength with better elongation. This result will be used for the subsequent study on the biodegradability of the plastic composite.

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