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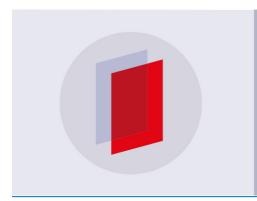


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### Study on the biodegradability and water adsorption of ldpe/sago starch blend

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Abstract. The aim of this study is to develop starch incorperated with low density polyethylene (LDPE) composites with enhanced degradable properties. The degradable test has been carried out by hydrolysis. The water uptake of the composites has been determined. The degradability of the compatible thermoplastic LDPE/starch was investigated by weight loss percent (WLP). Simple and alkaline hydrolysis of the composites apparently involves surface starch granules; embedded granules are not easily accessible because of the poor moisture absorption of the matrix. Exposure of the composites to hydrolysis resulted in massive deterioration typified by turbidity, discoloration, embrittleness and dimensional changes. Moisture uptake increased with increased starch content and immersion time. The time taken for the composites to equilibrate was about three months even when they were immersed completely in water. The mechanical properties of the composite also dropped with increasing moisture uptake. SEM imaging was also carried out on the samples before and after degradation. High content of starch also was found to increase the rate of biodegradability of LDPE/sago starch mould.

#### 1. Introduction

During the latter of this century, synthetic polymers have become major new materials for a wide range of applications, such as packaging, industrial, medical etc. Low density polyethylenes (LDPE) are the most commonly used synthetic plastics [1]. They are slow in degradability in natural environments, causing serious environmental problems. In this regard, there is growing interest in non-degradable synthetic polymer biodegradation using starch [2]. The addition of starch as a filler for producing biodegradable composites has been studied by many researchers. Starch is identified as a key natural product for the production of biodegradable plastic. Lowering the amount of synthetic polymer by blending with starch is a strategy widely adopted for research. [3]

Degradation is irreversible change in chemical and physical properties of a polymer with time generally leading to deterioration in mechanical properties. This is normally measured as decrease in tensile, toughness and impact resistance. The time taken for the properties of polymer to become unacceptable is

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the durability of the polymer [4][5]. The process of degradation describes the chain scission process during which polymer chains are cleaved to form oligomers and finally to form monomer [5]. The increased outdoor use of plastic has created a need for a better understanding of the effect of the environment factor has significant effect on appearance and properties of composites. Th severity of the damage depends largely on factors such as the nature of environment, types of polymeric material, and duration of exposure. The effect can be anywhere from discolouration, embrittlement, and cracking to a complete breakdown of the polymer structure [5][6]. The main objectives of this paper were to investigate effect of hydrolysis studies on the biodegradability of LDPE/sago starch blend at certain period of time.

#### 2. Experimental

#### 2.1 Materials

Sago starch as filler used in this study was food grade stuff. Sago starch was purchased from G-far keropok enterprise (Malaysia). The moisture content of the starch was 13% and the granule sizes ranged from 9  $\mu$ m to 35  $\mu$ m [8]. Low density Polyethylene (LDPE) with melting temperature of 140°C, was collected from Petlin Malaysia Sdn Bhd (Malaysia). Maleic Anhydride Grafted Polypropylene (MAPP) was chosen as a coupling agent which supplied by MTBE (M) Sdn Bhd.(Malaysia). *Aspergillus niger* for degradation studies was purchased from National Pharmaceutical Control Bureau (Malaysia).

#### 2.2 Sample preparation for sago starch/LDPE blend

The starch and LDPE were dried before sample preparation in an oven at 70 °C for 24 h and then cooled in a desiccator for 1 h to ensure it was moisture free. The sample was mixed and placed in a airtight plastic container to equilibrate for 12 h at 25 °C and then extruded. Table 1 represents the different composition of the blend. The compounding of LDPE/starch blend was done using Brabender twin screw extruder. The compounding process was carried out at a speed of 80 rpm and the temperature was set at 150°C,150°C,140°C for nozzle, front and middle part, respectively [7][9]. The sample was fed manually as much as the extruder would process. The extrudets will be palletized using a pelletizer machine for each formulation and were stored in an airtight plastic container.

#### 2.3 Injection molding

After the storing process, the pallets were injection moulded with injection moulding machine equipped with a diameter 35 mm screw. The injection moulding pressure was 1400 bars, and the holding pressure was varied from 600 to 1000 bars. The injection moulded specimens were stored in an airtight plastic container. The detail information of composition of LDPE/Sago starch blend is shown in table 1.

No	Samples	LDPE	Sago
		(w/w %)	Starch
			(w/w %)
1	Native LDPE	100	-
2	LDPE/SS:95/5	95	5
3	LDPE/SS:90/10	90	10
4	LDPE/SS:85/15	85	15
5	LDPE/SS:80/20	80	20
6	LDPE/SS:75/25	75	25
7	LDPE/SS:70/30	70	30

**Table 1.** Composition (w/w) of LDPE/Sago starch blend.

#### 2.4 Water absorption

Water absorption test was carried out according to ASTM Standard D750-95. The molded specimen, in the size of 2.0 x 3.5 cm, were dried for 6 hours at 50°C before they were cooled and weighed. The samples were soaked in distilled water at  $23\pm1$ °C. The samples were periodically weighted every 2 days to record any change in their weights. The percentage of the water absorption by films was then calculated using the following formula:

$$\% Wf = \frac{Ww - Wc}{Wc} \times 100 \tag{1}$$

Where  $W_f$  is the final increased weight percentage,  $W_w$ , the wet weight and  $W_c$ , the conditioned weight of the testing samples.

#### 2.5 Biodegradation studies

#### 2.5.1 Hydrolysis

2.5.1.1 Simple hydrolysis The method used was according to the one proposed by Arvanitoyannis *et al.* (1997) [1]. The samples in the size of dumbbell shapes were soaked in 20 ml distilled water at  $70^{\circ}$ C. The changes in the weight were measured every 3 hours in order to record the weight losses versus time.

2.5.1.2 Alkali hydrolysis The method used was that of Arvanitoyannis *et al.* (1997). The samples with the size of dumbbell shape were soaked in 20 ml 0.1 sodium hydroxide at  $70^{\circ}$ C. The changes in weight were measured every 3 hours to record the weight losses versus time.

#### 2.6 Weight Loss Percent (WLP)

The weight loss percent was determined as the following method: Prior to the experiment, the molded specimen were accurately weighed. After the mould samples were placed at the degradable environments for 18 hours, they were also accurately weighed. The ratio of the weight reduction of the mould after degradation to the original weight of the mould prior to degradation was defined as weight loss percent. The formula of WLP was as follows:

Weight loss (%) = 
$$\frac{Wo-We}{Wo} \times 100$$
 (2)

Where WLP (%) refers to the weight loss percent of the degradable mould;  $W_e$  refers to the weight of the mould after degradation;  $W_o$  refers to the weight of the mould before degradation.

#### 2.7 Morphological properties

Morphological studies were carried out on the tensile fracture surface of test specimen. The morphological surface of the samples before and after biodegradation was studied by using EVO 50 (ZEISS brand) scanning electron microscope (SEM) at 300KV. Test samples were first sputter-coated with gold to prevent any electrical discharge during testing. The coated sample was then observing the microstructures of the specimen.

#### 3. Result and discussion

#### 3.1 Water absorption

Starch content influences the water absorption of LDPE/Sago starch blend. This phenomenon can be observed through figure 1 that shows the variation in water absorption with time of immersion. Water

absorption increased with immersion time and increasing sago starch content. Rapid water absorption was observed for all samples within the first few days of immersion, but this decreased slowly with time. This phenomenon has been observed at different relative humidity. The absorption of water is related to its rate of diffusion into the composites. Starch based synthetic materials tend to absorb water because the hydroxyl group in starch can form a hydrogen bond with water. Since the starch is hydrophilic, it has a highly tendency to attract water molecules [7][8][9]. There is also high amylopectin in sago starch (73%) that influenced the water absorption. researcher reported that the starch blends containing high amylopectin content absorbed more water.

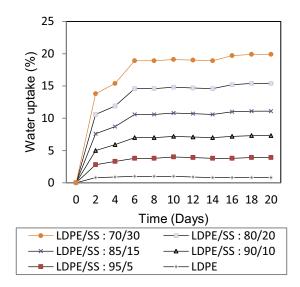
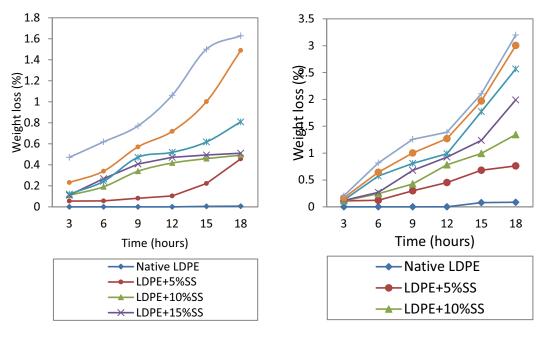


Figure 1. Percentages of water absorption for LDPE/Starch blend with time of Immersion.

#### 3.2 Biodegradation studies

*3.2.1 Hydrolysis* The data obtained from the simple hydrolysis test are shown in figure 2(a). Native LDPE significantly experienced a slower weight loss as compared to LDPE filled sago starch blend. LDPE with higher amount of starch showed higher weight losses. This might be due to the loss of starch which got dissolved in water.



(a)

(b) Figure 2. Percentage weight loss versus degradation time on LDPE/Sago starch (5wt% to 30wt%) a) Simple hydrolysis b) Alkaline hydrolysis.

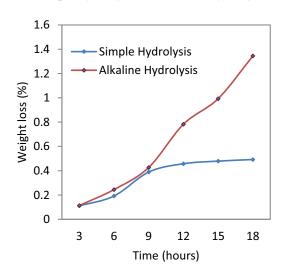
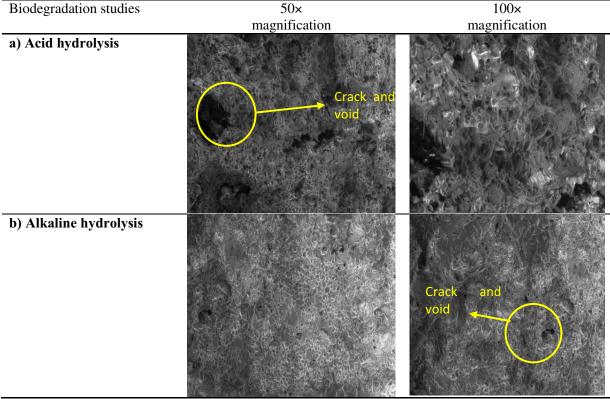


Figure 3. Percentage weight loss versus degradation time of LDPE/SS:90/10 starch blend on hydrolysis.

Figure 2(b) illustrates the results of the alkali hydrolysis test. All the LDPE/SS blend showed a drastic weight loss versus hydrolysis time, compared with the native LDPE without sago starch. As seen in a figure 3, the alkali hydrolysis caused a higher weight loss as compared to the simple hydrolysis. Thus, NaOH was found to enhance the solubilization of gelatinized starch in the polymer blend [1]. The purpose of using these methods for assessing the biodegradability of blend was to prove that the synthesized composites could be degraded in water with different values of pH. Higher pH values could accelerate the degradability of starch based mould.

Figure 4 is a SEM micrograph of LDPE/Sago starch blend (containing 10 w/w% starch) after 18 hour of hydrolysis. SEM photographs demonstrated degradation of starch particles within the LDPE. Starch particles in all specimens degraded regardless of degradable environments. Large crack and void appear on the surface of the matrix which apparently resulted in its collapse, thus exposing the embedded starch granules. Fracture surface of alkaline hydrolysis show many crack, void and holes compare to fracture surface of simple hydrolysis. This proves that higher pH value can affect the degradation of the composites.



**Figure 4.** SEM micrograph fracture part of LDPE/SS blend with 10wt% loading at (50× and 100×) magnification after 18-hour hydrolysis testing.

#### 4. Conclusion

Starch is blended with synthetic polymer to induce degradability of synthetic polymers. Addition of sago starch to LDPE will enhanced biodegradability of the composites. Sago starch filled-LDPE composites absorb moisture in proportion to the amount of starch added. The ability of the composites to absorb moisture not only reduces their mechanical properties but also renders them susceptible to fungal attack, thus limiting their use. Degradation studies show that the composites exhibit varying degrees of degradation. Alkaline hydrolysis produced high dramatic changes and conduct fast degradation compared to simple hydrolysis. The sample resulted in weight loss and the production of void on the sample surfaces. Consumption of sago starch as a biodegradable agent initiates the biodegradation process. According to this research, sago starch based low density polyethylene is a microbial biodegradable polymer.

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#### 6. Acknowledgements

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