

Effect of PEG on the biodegradability studies of Kenaf cellulose -polyethylene composites

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Abstract: Several blends of cellulose derived from bast part of kenaf (*Hibiscus cannabinus L.*) plant, with different thermoplastics, low density polyethylene (LDPE) and high density polyethylene (HDPE), were prepared by a melt blending machine. Polyethylene glycol (PEG) was used as plasticizer. Biodegradability of these blends was measured using soil burial test in order to study the rates of biodegradation of these polymer blends. It was found that the cellulose/LDPE and cellulose/HDPE blends were biodegradable in a considerable rate. The bio-composites with high content of cellulose had higher degradation rate. In addition, biodegradability of the bio-composites made up using PEG was superior to those of the bio-composites fabricated without PEG, due to the improved wetting of the plasticizer in the matrix polymer. The results were also supported by the scanning electron microscopy (SEM).

Keywords: Kenaf cellulose, polyethylene, biodegradation, PEG, soil burial test

Introduction

Polyethylene (PE) is a non-degradable polymer being used extensively in many applications. It is also the first commodity plastic to be used for food packaging, came into general use in the 1950s. Since then it has achieved its dominant position as a packaging material for a wide range of foods and beverages because of its relatively low cost, its versatile properties, and the ease with which it can be manufactured and converted. The main end forms of PE plastics used for food packaging are films, made by both cast and oriented processes, bottles and other containers made by thermoforming and blow molding processes. Recently, there is a growing interest in replacing some or all of the synthetic polymers, like PE, to become biodegradable materials due to their environmental impacts (Arvanitoyannis *et al.*, 1998).

Despite of good biodegradability properties of some natural polymers such as 3-hydroxyl butyrate (PHB) and its copolymers and aliphatic polyesters, their higher cost compare to petroleum-based commodity plastics prevents them from a larger

commercial usage and finds applications only in niche sectors (Avella *et al.*, 2005).

The best-known renewable resources able to create biopolymer and biodegradable plastics are starch and cellulose (Chandra and Rustgi, 1998; Selke, 2000). There are a lot of studies about using starch in biopolymer production but fewer for cellulose. Cellulose obtained from kenaf (*Hibiscus cannabinus L.*), an annual plant with many environmental advantages, is a good source of biodegradable polymer (Nishino, 2004).

Biodegradation occurs with enzymatic action and involves living microorganisms. Almost all microbial degradations are carried out by both fungi and bacteria. There are four biodegradation environments for polymeric products namely soil, aquatic, landfill and compost. Each environment contains different microorganisms and has its special conditions for degradation. In soil, fungi are mostly responsible for degradation of organic matter including polymers (Baker and Mead, 2000; Chandra and Rustgi, 1998; El-Hadi Abdel Ghaffar, 2002; Hodzic, 2004; Sridach *et al.*, 2006).

The biodegradation rate in biopolymers depends on a number of factors including fiber content, the biodegradability of each component and the quality of the interface. The fiber addition generally increases the degradation rate of composites and alkaline treatment of fibers produce a slightly higher degradation rate than pure matrix (Plackett and Vazquez, 2004). Other than that, the additives used (e.g. plasticizers, fillers, etc.) are important in biodegradation kinetics as well as the type of polymer reflected in molecular weight, structure and crystallinity (Guilbert and Gontard, 2005). In addition, biodegradability and the rate of biodegradation depend in general on the substrate structure, the substrate composition and the existing microorganisms. Also, the rate of biodegradation decreases with the progress of the reaction (Sridach *et al.*, 2006).

The objective of this study is to investigate the kenaf cellulose-PE biocomposites biodegradability. Soil burial test is used to determine the biodegradability on the polymer blends. The effects of plasticizer, PEG, in biodegradation of these polymers were also studied.

Materials and Methods

Materials

The cellulose used in this study was extracted from kenaf, variety V36, planted in Malaysian Agricultural Research and Development Institute (MARDI). LDPE with density 0.923 g/cm³, melt flow index 6.0 g/10 min and HDPE with density 0.945 g/cm³, melt flow index 20.0 g/10min were supplied from TITAN PETCHEM (M) SDN. BHD. PEG with average molecular weight 8000 was supplied from SIGMA. The soil used for this experiment was topsoil from Seri Serdang, Selangor, Malaysia with pH 6.50.

Cellulose extraction

The cellulose processing was carried out according to Standard Test Method, ASTM No.D1103-60 and using method of Han and Rowell (1996) from kenaf fiber.



Figure 1. Part of specimens before filling by soil

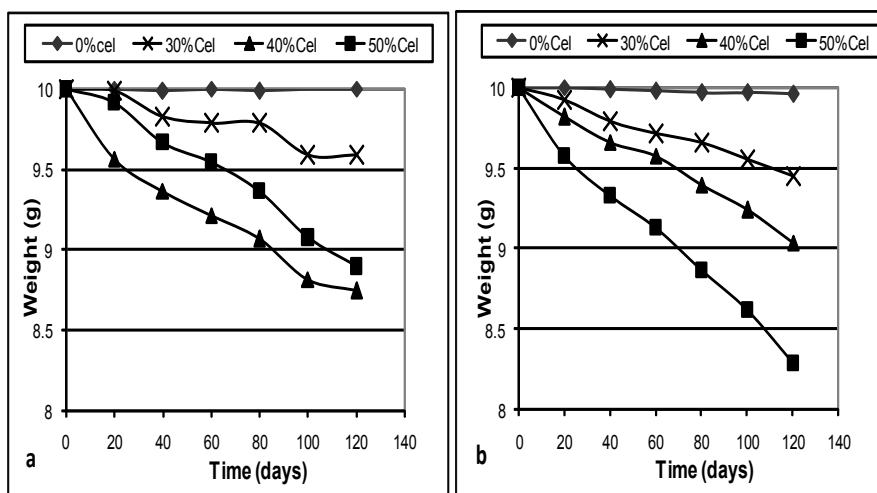


Figure 2. Biodegradability of (a) LDPE/cellulose and (b) HDPE/cellulose composites during burial time

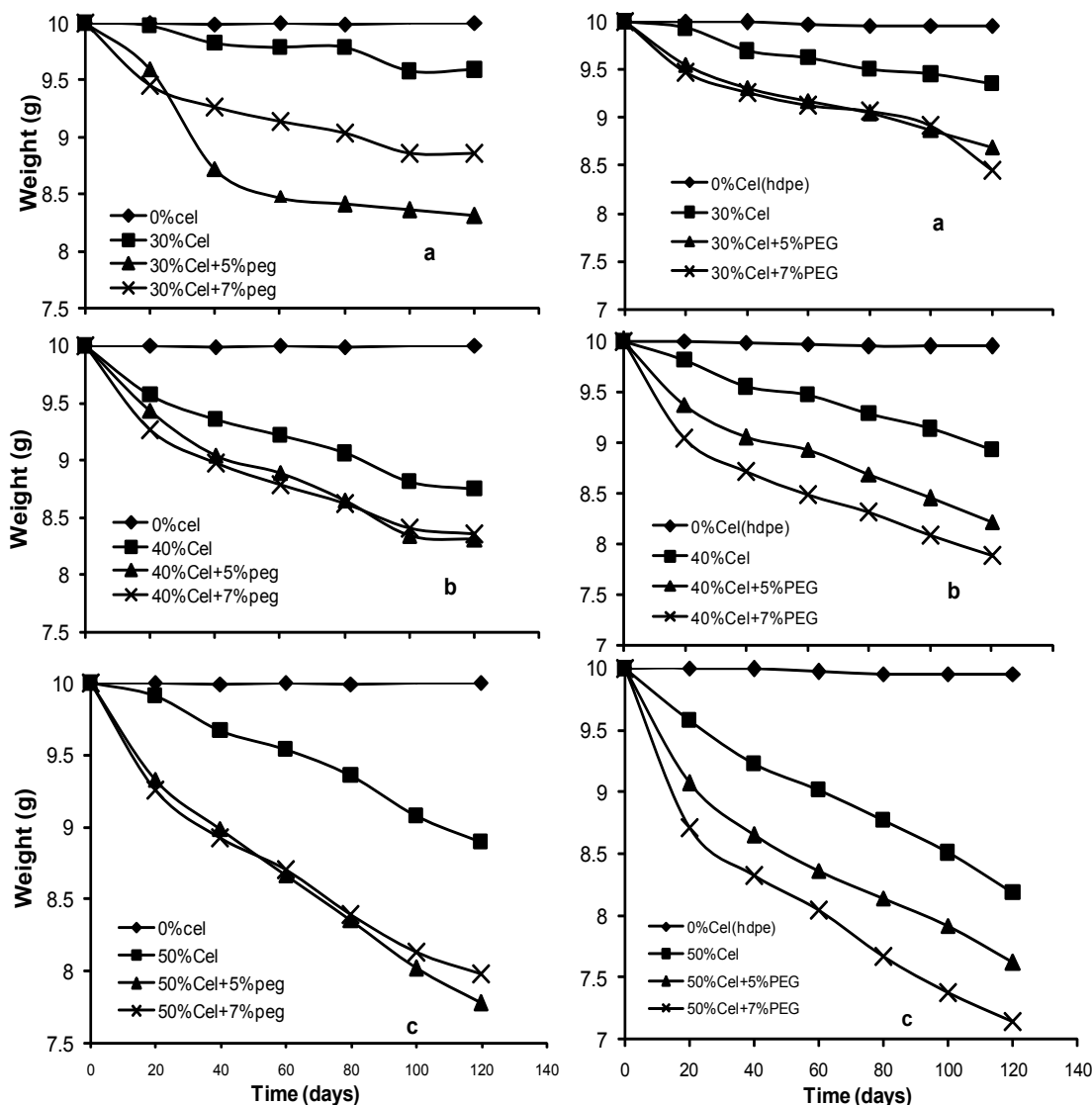


Figure 3. Biodegradability of LDPE/PEG (Left) and HDPE/PEG (Right) composites with 30% (a), 40% (b) and 50% (c) cellulose during burial time

Blending

PE/Cellulose/PEG formulations were processed using two different types of PEs (LDPE, HDPE) with 0, 30, 40 and 50% of cellulose and 0, 5 and 7% of PEG, respectively. Predetermined amount of materials were mixed using Thermo Haake PolyDrive R600/610 blending machine. Blending was performed for LDPE at 125°C with rotor speed 30 rpm in 10 minutes and 145 °C for HDPE at 30 rpm in 12 minutes.

Soil burial test

A soil burial test was carried out on a laboratory scale to examine the biodegradability. First, rectangular sheets (about 10 cm × 15 cm × 1 mm thickness) of specimens were buried in the soil by random pattern (Figure 1). The pot containing the

soil and samples were incubated at almost constant temperature of 26°C for four months. The moisture content was maintained at 40-50% of the soil’s maximum water holding capacity. This humidity is optimal for microbial activity (Chandra and Rustgi, 1998). Also, the pots were covered with plastic film to avoid water evaporation from the soil surface.

Biodegradation was estimated by monitoring changes in weight as a function of burial time. The samples were removed from the soil every 20 days. The debris on the specimens was removed by washing with water. The samples were then dried in an oven at 100-105°C for 24 hours. After drying, they were weighed using an electronic balance with a precision of 0.1 mg. For comparative reason, the weight of treatments was normalized to 10 g in the first measurement.

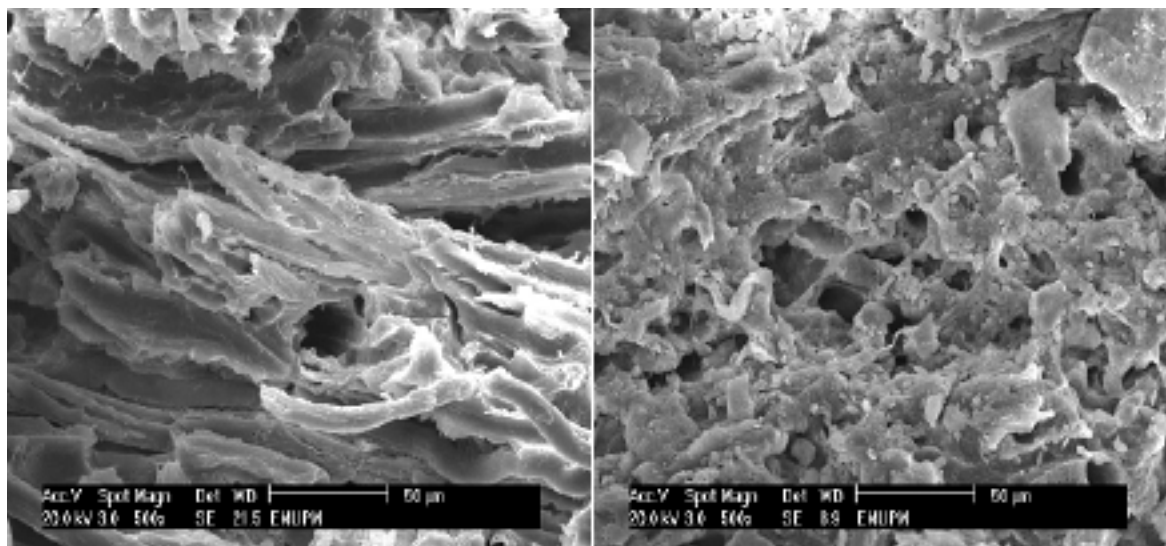


Figure 4. Scanning electron micrographs (Mag. \times 500) of the fracture surface of 50%cellulose+5%PEG for LDPE (Left) and 50%cellulose+7%PEG for HDPE (Right) composites after 4 months in the soil

Results and Discussion

Figure 2 shows the biodegradation test experimental data for the blends of LDPE and HDPE with cellulose. The dispersion of integrity and breakup of samples was observed after around 40 days and increased during burial time. It can be noted that higher content of cellulose had lead to higher degradation. These observations were in agreement with the findings reported in the works by Oldak *et al.* (2005) and Kaczmarek and Oldak (2006). They found that with a very small amount of cellulose (5-15%) in polyethylene (PE) composite, it may not improve its biodegradability. The biodegradability in PE films will only have the pronounced effect if it contains 30% cellulose and above.

Figure 3 presents the effect of PEG content on the biodegradation of LDPE and HDPE/PEG composites. The compounds show some degree of biodegradability with increasing PEG. The degradation is about 20% for LDPE and 25% for HDPE compounds after 120 days, respectively.

From the data in Figures 2 and 3, it shows the effect of different cellulose and PEG loading on biodegradability of LDPE and HDPE/Cellulose/PEG compounds. With additional PEG, all PE/cellulose samples show substantial decrease in their weight in the first 30 days. It was noted that for LDPE/cellulose compounds, addition of 5% PEG shows higher degradation rate after 120 days. And for HDPE/cellulose compounds, addition of 7% PEG shows the highest degradation rate. It was also noted that 50%

cellulose and 5% PEG for LDPE treatments and 50% cellulose and 7% PEG presented highest degradation rate among all treatments.

Figure 4 illustrates the Scanning electron micrograph (SEM) results for LDPE treatment with 50% cellulose + 5% PEG and HDPE treatment with 50% cellulose and 7% PEG. From these figures, it is apparent that cellulose is almost completely degraded from the surface of the samples due to soil burial.

Conclusion

As the biodegradability test progressed over time up to 120 days, the composites biodegradability enhanced with increasing cellulose content because the cellulose is easily attacked by microorganisms. It was observed that for LDPE/cellulose composites, with addition of 5% PEG, it shows better biodegradability rate. As in the HDPE/cellulose compounds, higher degradation rate was achieved with the addition of 7% PEG. It is due to good interactions between molecular structure of matrix (PE), cellulose and PEG in each case. The results suggested that it is possible to blend the non-degradable polyethylene polymer with kenaf cellulose in order to improve its biodegradability. The successful biodegradability test can be continued with more percentage of cellulose and PEG; however, it may be weaken the mechanical properties of the composites. These results were supported by surface degradation of biocomposites observed through morphological study.

References

- Arvanitoyannis, I., Biliaderis, C.G., Ogawa, H. and Kawasaki, N. 1998. Biodegradable films made from low-density polyethylene (LDPE), rice starch and potato starch for food packaging applications: Part 1. *Carbohydrate Polymers* 36(2-3): 89-104.
- Avella, M., De Vlieger, J.J., Errico, M.E., Fischer, S., Vacca, P. and Volpe, M.G. 2005. Biodegradable starch/clay nanocomposite films for food packaging applications. *Food Chemistry* 93(3): 467-474.
- Baker, A.-M.M. and Mead, J. 2000. Thermoplastics. In *Modern plastics handbook*, (Harper, C.A., Ed.), pp. 1.1-1.92. McGraw - Hill.
- Chandra, R. and Rustgi, R. 1998. Biodegradable polymers Program Polymer science. *Progress in Polymer Science* 23: 1273- 1335.
- El-Hadi Abdel Ghaffar, A.M. 2002. Development of a biodegradable material based on poly (3-hydroxybutyrate) PHB. Dissertation, Ph.D Thesis. In. Martin-Luter University, Halle-Wittenberg.
- Guilbert, S. and Gontard, N. 2005. Agro-polymers for edible and biodegradable films: review of agricultural polymeric materials, physical and mechanical characteristics. In *Innovations in Food Packaging*, (Han, J.H., Ed.), pp. 263-276. Elsevier Academic Press.
- Han, J.S. and Rowell, J.S. 1996. Chemical composition of fibers. In Paper and Composites from Agro-Based Resources. In *Paper and Composites from Agro-Based Resources* (Rowell, R.M. and Rowell, J., Eds.), pp. 83-130 CRC Press.
- Hodzic, A. 2004. Re-use, recycling and degradation of composites. In *Green Composites, Polymer Composites and the Environment*, (Baillie, C., Ed.), pp. 252-271. CRC Press.
- Kaczmarek, H. and Oldak, D. 2006. The effect of UV-irradiation on composting of polyethylene modified by cellulose. *Polymer Degradation and Stability* 91(10): 2282-2291.
- Nishino, T. 2004. Natural fiber sources. In *Green Composites, Polymer Composites and the Environment*, (Baillie, C., Ed.), pp. 49-80. CRC Press.
- Oldak, D., Kaczmarek, H., Buffeteau, T. and Sourisseau, C. 2005. Photo- and bio-degradation processes in polyethylene, cellulose and their blends studied by ATR-FTIR and Raman spectroscopies. *Journal of Materials Science* 40(16): 4189-4198.
- Plackett, D. and Vazquez, A. 2004. Natural polymers sources In *Green Composites, Polymer Composites and the Environment*, (Baillie, C., Ed.), pp. 123-153. CRC Press.
- Selke, S.E. 2000. Plastics recycling and biodegradable plastics. In *Modern plastics handbook*, (Harper, C.A., Ed.), pp. 12.11- 12.108. McGraw - Hill.
- Sridach, W., Hodgson, K.T. and Nazhad, M.M. (2006). Biodegradation and recycling potential of barrier coated paperboards. *BioResources* 2(2): 179-192.