Carbohydrate Polymers 142 (2016) 133–140

Contents lists available at ScienceDirect

Carbohydrate Polymers

journal homepage: www.elsevier.com/locate/carbpol

# Microcrystalline-cellulose and polypropylene based composite: A simple, selective and effective material for microwavable packaging

S. Ummartyotin*, C. Pechyen*

*Department of Physics, Faculty of Science and Technology, Thammasat University, Klong Luang 12120, Patumtani, Thailand*

ARTICLE INFO

*Article history:*
Received 29 September 2015
Received in revised form 4 January 2016
Accepted 9 January 2016
Available online 12 January 2016

*Keywords:*
Microcrystalline cellulose
Polypropylene
Composite
Microwavable packaging

ABSTRACT

Cellulose based composite was successfully designed as active packaging with additional feature of microwavable properties. Small amount of cellulose with 10 μm in diameter was integrated into polypropylene matrix. The use of maleic anhydride was employed as coupling agent. Thermal and mechanical properties of cellulose based composite were superior depending on polypropylene matrix. Crystallization temperature and compressive strength were estimated to be 130 °C and 5.5 MPa. The crystal formation and its percentage were therefore estimated to be 50% and it can be predicted on the feasibility of microwavable packaging. Morphological properties of cellulose based composite presented the good distribution and excellent uniformity. It was remarkable to note that cellulose derived from cotton can be prepared as composite with polypropylene matrix. It can be used as packaging for microwave application.



## 1. Introduction

Due to the environment and sustainable issues, this century has witnessed remarkable achievements in green technology in the field of materials science through the development of bio-based composite (Faruk, Bledzki, Fink, & Sain, 2012; Ummartyotin & Manuspiya, 2015a,b). The development of high performance materials from bio-based material has been increased worldwide. It exhibited the performance of use in many areas of research. The example of bio-based composite was infrastructure, automotive part, electronic device as well as medical and pharmaceutical research (Akil et al., 2011; Sengupta, Bhattacharya, Bandyopadhyay, & Bhowmick, 2011; Abdul Khalil, Bhat, & Ireana Yusra, 2012; Koronis, Silva, & Fontul, 2013). The evidence of bio-based resource was cellulose and its derivative, chitin–chitosan, poly lactic acid, starch, poly butylene based materials (Hankermeyer & Tjeerdema, 1999; Ravi Kumar, 2000; Averous, 2004; Elsabee & Abdou, 2013; Soroudi & Jakubowicz, 2013; Balan & Verestiuc, 2014). Up to the present time, numerous efforts have been extensively conducted on both bulk structure and surface properties. The activity on modification can be endorsed on binary blend preparation, surface modification by physical and chemical routes as well as insertion of small amount of nano-scale particle for being as composite. From our long term achievement on the academic and industrial researches of cellulose of our research group, cellulose has been studied from natural resource to many feasibilities of application. The role of cellulose for industrial commercialization was versatile in many areas of research. From the fundamental point of view, it was remarkable to note that cellulose was considered as one of the most abundant naturally occurring bio-based polymer. It was commonly found in the cell walls of plants and certain algae. Cellulose can be found from some types of bacterial specie. It can be produced from bacteria. It was considered as a product of its primary metabolism and forms a protective coating and the most effective specie was called "*Acetobacter xylinum*" (Fiedler, Fussel, & Sattler, 1989; Cheng, Wang, Chen, & Wu, 2002; Kongruang, 2008; Hong et al., 2012; Fu, Zhang, & Yang, 2013). Recently, there has been a great deal of research interest in utilizing cellulose for various applications such as composite manufacturing due to its remarkable reinforcing capability, excellent mechanical properties, low density and environmental benefits. One of our excellent projects related to the development of bacterial cellulose based composite has successfully gained many interests on electronic device substrate (Ummartyotin, Juntaro, Sain, & Manuspiya, 2012; O-Rak et al., 2014). The benefit of bacterial cellulose was involved on mechanical properties. The Young's modulus of its single fibril was measured to be as high as 114 GPa. It also has attractive feature of high degree of crystallinity of 89%, high degree of polymerization of 14,400 as well as high specific surface area of 37 $m^2$/g. Although, bacterial cellulose was successfully developed and it provided numerous advantages including purity and

* Corresponding authors. Tel.: +6625644490; fax: +6625644458.
*E-mail addresses:* sarute.ummartyotin@gmail.com (S. Ummartyotin), chiravoot.p@gmail.com (C. Pechyen).

http://dx.doi.org/10.1016/j.carbpol.2016.01.020
0144-8617/

homogeneity, the use of bacterial cellulose was still limited on cost effectiveness if mass production was developed for industrial commercialization.

One of effective routes for raw material on cellulose preparation was related to cotton. It was considered due to availability, ease of processing, low cost benefit as well as valued-added concept from agricultural waste (Vasconcelos & Paulo, 2006; Meftahi et al., 2010; Oun & Rhim, 2015). Cellulose prepared from cotton should be recommended on purification from lignin, hemicellulose and any impurities. To use cellulose from cotton in industrial commercialization, it gained many interests on active and intelligent food packaging application. It can be integrated into commodity plastic and subsequently provided significant enhancement on thermal resistance, mechanical properties as well as environmental benefit. To insert small amount of cellulose into commodity plastic can be tailored significant effort on many areas of research. From the fundamental point of view, the use of transparent commodity plastic was extensively considered as packaging. It offered numerous advantages including specific design, low cost and effective processing as well as commercially available for mass production.

Contrary, although it was important to note that mass production process on plastic based packaging was successfully emerged, it was still limited in particular categories of food. The variation of food quality was relied on fresh food, dry food and semi-dry food. To maintain the quality of food with higher efficiency, the design on food packaging should be extremely considered in order to serve on the variation of food quality. One of strategic approaches on the design of food packaging based on plastic research was focused on plastic based composite. To integrate on the small amount of cellulose derived from cotton into plastic was one of the effective routes in order to design food packaging based on microcrystalline cellulose based polymer composite. From the fundamental point of view, active packaging can be designed by means of many routes. The example of modification was therefore involved on controllable atmosphere, high absorption of water and gas, as well as antimicrobial properties (Barbosa-Pereira, Angulo, Lagaron, Paseiro-Losada, & Cruz, 2014; Han, 2014; Mihindukulasuriya & Lim, 2014). In order to reach the objective of modification on transparent and flexible plastic as active packaging, the incorporation on cellulose as filler into polymer matrix was emphasized to develop. It was important to note that cellulose exhibited many benefits due to high porosity, high chemical and thermal resistance, cost effectiveness, ease of processing as well as availability.

The objective of this research was involved on the development of cellulose from cotton. Chemical modification on cellulose was investigated. Small amount of cellulose was integrated into polypropylene matrix. The role of maleic anhydride was investigated as coupling agent in composite formation. Characterization on structural, thermal, mechanical and morphological properties of microcrystalline cellulose based polypropylene composite was evaluated. After that, preliminary investigation on microwave irradiation of microcrystalline cellulose and polypropylene composite was determined for active packaging.

## 2. Materials and methods

### 2.1. Materials

PP (EL-PRO™ P600F) was obtained from SCG Chemicals (Thailand). PP-*g*-MA (DuPont™ Fusabond® P613), as a reactive compatibilizer, was obtained from Chemical Innovation (Thailand). Cotton was donated as a gift from SCG paper, Thailand. Analytical grade of hydrochloric acid was purchased from Sigma Aldrich, Thailand. All of chemical reagents were used as received without further modification.

**Table 1**
Information on microcrystalline cellulose and polypropylene composite sample.

| Sample | Condition | | |
|---|---|---|---|
| | *i*-PP (wt%) | MA-*g*-PP (wt%) | MC (wt%) |
| 1 | 100 | 0 | 0 |
| 2 | 100 | 0 | 5 |
| 3 | 100 | 0 | 10 |
| 4 | 100 | 0 | 20 |
| 5 | 100 | 10 | 5 |
| 6 | 100 | 10 | 10 |
| 7 | 100 | 10 | 20 |

### 2.2. Methods

#### 2.2.1. Cellulose preparation from cotton

Smaller size of cotton was cut by hand. Then cotton (80 g) was then soaked in a solution containing 1% sodium hydroxide (NaOH), and heated at 60 °C for 1 h. After complete removal of the supernatant, the waste cotton was treated with DI water for 1 h at 60 °C. Then the liquid was removed by filter to separate pure cellulose. The cellulose was washed with distilled water until neutralization. The cotton was then subjected to acid hydrolysis with 2 M hydrochloric acid (HCl) at 80 °C for 4 h. The acid solution was removed by filtration and the powder was washed with distilled water several times until neutral condition (pH 6–7). The powder was dried at 60 °C overnight and passed through 140 mesh sieve and stored under controlled humidity and temperature.

#### 2.2.2. Cellulose based composite preparation

Small amount of cellulose was integrated into polypropylene and polypropylene-*g*-maleic anhydride, respectively. The role of maleic anhydride was involved on coupling agent. Cellulose was incorporated into polymer matrix by using a Brabender internal mixer. The process was set at 200 °C of screw temperature and the screw rotation was set at 60 rpm for 5 min. After that, the composite was cooled in air to ambient temperature. Then, it was pressed by compression molding technique at a temperature of 200 °C under 15 t of pressure for 10 min. Only polymer matrix was employed for comparison. All of samples were conditioned by keeping them in a desiccator at 25 °C and relative humidity of 75 ± 2%, maintained using a saturated solution of sodium chloride (NaCl), prepared following the ASTM E 104 standard. The amount of cellulose and polymer matrix is exhibited in Table 1.

#### 2.2.3. Microwave irradiation evaluation of cellulose based composite

In order to determine the feasibility of microcrystalline cellulose and polypropylene composite for active packaging, microwave irradiation test were employed to investigate. Evaluation can be categorized into two different parts. The one was focused on microwave irradiation test; microcrystalline cellulose based polypropylene composite was investigated by microwave system technique. The power of microwave test was set to be 340, 650 and 1300 W. The investigation time was observed for 2.5, 5 and 10 min, respectively. Additional information is exhibited in Table 2.

### 2.3. Characterization technique

#### 2.3.1. Differential scanning calorimetry

DSC of the cellulose based composite was performed in the temperature range of −50 °C to 300 °C at the heating rate of 10 °C/min (TA-10000 DSC, TA Instruments, DE, USA). The crystallization and melting temperature were determined from the heat flow curve.