

การปรับปรุงคุณสมบัติของกระดาษรีไซเคิลจากกล่องลูกฟูกโดยวิธีผสมกับเยื่อใหม่และการเคลือบด้วยไคโตซาน

Property Enhancement for OCC Paper by Virgin Pulps and Chitosan Coating

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บทคัดย่อ

งานวิจัยนี้จัดทำขึ้นเพื่อศึกษาการปรับปรุงคุณสมบัติเชิงกลของกระดาษรีไซเคิลจากกล่องลูกฟูกด้วยวิธีการผสมกับเยื่อใหม่ที่ได้จากเยื่อฟางข้าว เยื่อชานอ้อย และเยื่อปอสา ด้วยวิธีการเติมสารเคมี และด้วยวิธีการเคลือบกระดาษด้วยสารละลายไคโตซาน จากผลการทดลองปรากฏว่ากระดาษรีไซเคิลที่เตรียมจากการผสมของเยื่อปอสา 30% และเยื่อรีไซเคิลจากกล่องลูกฟูก 70% สามารถเพิ่มคุณสมบัติเชิงกลของกระดาษรีไซเคิลได้ และพบว่าคุณสมบัติเชิงกลของกระดาษรีไซเคิลที่ได้จากการผสมของเยื่อปอสาและเยื่อกล่องลูกฟูกนี้ดีขึ้นหลังจากเติมสารเติมเต็ม โดยงานวิจัยนี้เน้นศึกษาผลของปริมาณ dry strength resin ต่อคุณสมบัติเชิงกลของกระดาษรีไซเคิล ผลปรากฏว่าคุณสมบัติเชิงกลของกระดาษรีไซเคิลไม่เปลี่ยนแปลงเมื่อปริมาณของ dry strength resin มากกว่า 0.45% ของน้ำหนักเยื่ออบแห้ง จากนั้นจึงทำการศึกษาการปรับปรุงคุณสมบัติเชิงกลของกระดาษรีไซเคิลโดยวิธีการเคลือบด้วยไคโตซาน จากผลการทดลองปรากฏว่าความเข้มข้นของสารละลายไคโตซานที่เหมาะสมคือ 0.3 %w/v ซึ่งทำให้ค่าความต้านทานแรงดึงและความต้านทานน้ำของกระดาษรีไซเคิลเพิ่มขึ้น 22% และ 69% ตามลำดับ

ABSTRACT

In this study, mechanical property enhancement of recycled old corrugated container (OCC) paper was examined. The studies included the mechanical property improvement of the recycled OCC paper by mixing with virgin pulps from rice straw, sugarcane bagasse, and paper mulberry, by adding chemical additives, and by coating with chitosan solution. The results shows that the recycled paper made from 30% of virgin paper mulberry pulp and 70% of recycled OCC pulp provided the improvement of paper strength properties. The overall mechanical properties of the recycled (OCC/paper mulberry) paper showed an improvement after adding chemicals. This work focuses on the effect of dry strength resin amount on the paper mechanical properties. The addition amount of dry strength resin over 0.45% of oven dry weight of pulp didn't show any improvement of paper mechanical properties. Then, the mechanical property enhancement by chitosan coating was studied. The results show that the optimal chitosan concentration was 0.3 %w/v which improved tensile strength and water resistance of the recycled paper by 22% and 69%, respectively.

Key Words: recycled paper, paper mechanical properties, old corrugated container (OCC), chitosan, coating

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INTRODUCTION

Corrugated box is a highly useful, cost-efficient, versatile packaging material and also the most-recycled packaging material on earth, with a recovery rate over 78 percent. Corrugated box has been recycled for decades, mostly by commercial users such as your neighborhood grocery store. The recycling of corrugated boxes, called old corrugated container (OCC), helps decrease solid waste disposal in landfills, saves fiber resources, and requires less total energy. However, the recycling of OCC faces a potential barrier. It is known that paper strength properties decrease after recycling (Horn, 1995; Howard and Bichard, 1992). The main factor contributing to paper strength deterioration is a reduced bonding ability of the fibers caused by an irreversible hornification (Nazhad and Paszner, 1994; Weise and Sedlachek, 1998). Many studies have reported the ways to increase the bonding ability of fibers after they already have been recycled. These approaches are such as applying additional refining (De Ruvo and Htun, 1983), adding dry strength agents (Smith, 1992), and subjecting the fibers to conditions that favor rehydration (Ogden, 1999). One approach taken in this study was to coat the recycled OCC paper by chitosan solution to overcome strength deficiencies of recycled OCC paper.

Chitosan is a high molecular weight linear carbohydrate composed of β -1,4-linked-2-amino-2-deoxy-D-glucose units. Chitosan is prepared by hydrolysis of the N-acetyl groups from the natural polymer chitin which can be isolated in commercial quantities from shells of crustaceans (crab, shrimp, etc). While chitin is insoluble in common solvent, chitosan containing basic amino group is soluble in acidified water. Chitosan possesses good mechanical properties, as has been reported to use as a wet end additive in paperboard to improve mechanical properties of the product (Laleg and Pikulik, 1991). Moreover, studies of chitosan coating on paper, paperboard, and cellophane have been reported. These reports, however, focus mainly on antimicrobial, medical, or barrier applications (Ho *et al.*, 2003; Vartiainen *et al.*, 2004; Gallstedt *et al.*, 2005; Kjellgren *et al.*, 2006). The improvement of some mechanical properties by chitosan coating has also been reported. For example, Kuusipalo *et al.* (2005) reported that barrier properties and bending strength of the paper was improved significantly after coating with chitosan. Tear index and tensile index of chitosan coated paper were also improved but only slightly. Furthermore, chitosan coating of paper-based packaging materials offer benefits because this polymer can be applied to the paper in line as an aqueous solution using conventional coating techniques and could be as an environmentally friendly coating material. Since chitosan possesses those good properties, in this work, we chose chitosan solution as a coating material to improve mechanical properties of the recycled OCC paper.

In this study, we first investigated the optimal virgin pulp (eg. rice straw, sugarcane bagasse, and paper mulberry pulp) that can mix with OCC pulp and improve the mechanical properties of the recycled OCC paper. The recycled paper made from the chosen virgin pulp and OCC pulp was used

as based paper for the further studies in this work. The present work focuses on the effect of chemical additives and chitosan coating on the mechanical properties of the prepared (virgin pulp/OCC) paper.

MATERIALS AND METHODS

Pulp

Four types of pulp, including recycled OCC pulp, paper mulberry pulp, rice straw pulp, and sugarcane bagasse pulp, were used for making handsheet paper in this study. Recycled OCC pulp was prepared by repulping OCC with water in Hollander beater. Paper mulberry pulp was prepared by pulping paper mulberry bark with 8% NaOH (% by oven dry weight of paper mulberry bark) using 8:1 of liquor to bark ratio at 100 °C for 3 hours. Rice straw pulp was prepared by pulping rice straw with 15% NaOH (% by oven dry weight of rice straw) using 14:1 of liquor to straw ratio at 100 °C for 3 hours. Sugarcane bagasse pulp was obtained by pulping sugarcane bagasse with 29% NaOH (% by oven dry weight of sugarcane bagasse) using 8:1 of liquor to straw ratio at 100 °C for 3 hours, and then bleaching with 8% H₂O₂, 2% NaSi₂O₃, and 1.5% NaOH (% by oven dry weight of sugarcane bagasse) using 8:1 of liquor to straw ratio at 100 °C for 2 hours.

Preparation of recycled paper

Laboratory paper (handsheets) of the recycled OCC pulp was prepared by using 70:30 of recycled OCC pulp to the other pulps ratio. Before making handsheets the pulp was dispersed in disintegrator for 10 minutes. Then, chemicals, including 1% aluminium sulfate octadecahydrate (supplied from Riedel-de Haën), a desired amount of polyacrylamide resin, 0.5% sizing agent, and 0.5% wet strength resin (supplied from Seiko PMC Corporation), were added to the pulp slurry and mixed with stirring at 5% consistency. Addition amounts of chemicals were based on oven dry weight of the pulp. Then, the handsheets were made according to TAPPI Standard Method T 205. The basis weight of handsheets was 60±5 g/m². The prepared handsheets were then conditioned at 23 °C and 50% relative humidity for at least 24 hours before physical testing or kept at room temperature before coating with chitosan solution.

Preparation of chitosan coated recycled paper

A desired amount of chitosan flake, 0.1%, 0.3% or 0.5% (%w/v), was mixed in 0.3% acetic acid solution. The solution was stirred at room temperature until chitosan flake was completely dissolved, and then filtrate through 100 mesh screen to obtain chitosan solution. Then, the prepared handsheets were coated with chitosan solution by using brush coating method. The coated handsheets were then conditioned at 23 °C and 50% relative humidity for at least 24 h before physical testing.

Measurement of mechanical properties of the prepared paper

The mechanical properties of the prepared handsheets were measured. Basis weight, thickness, tensile strength, tearing resistance, burst strength, wet tensile strength, and water

absorptiveness (cobb test at 60 seconds), were determined according to TAPPI Standard Methods T 410, T 411, T 494, T 414, T 403, T 456, and T 441, respectively. The testing data were statistically analyzed by using SPSS program, and the statistical difference of mean was determined by using Duncan's new multiple rang test (DMRT).

RESULTS AND DISCUSSION

Preliminary study of the effect of pulp type on the mechanical properties of recycled OCC paper

Since the paper strength properties have been known to decrease after recycling, the first approach in this study was to find the suitable type of virgin pulp that can combine with recycled OCC pulp and increase the mechanical properties of the recycled paper. In this work, we chose virgin pulp produced from agricultural waste/crops in Thailand, including paper mulberry (fiber length = 9.29 mm, width = 0.0161 mm), rice straw (fiber length = 0.72 mm, width = 0.0103 mm), and sugarcane bagasse (fiber length = 2.06 mm, width = 0.0177 mm). The laboratory papers (handsheets) were produced from 70% of recycle OCC pulp and 30% of virgin pulp without adding chemicals. The mean thickness of the handsheets was 0.143 ± 0.06 mm. The mechanical properties of these handsheets were summarized in Figure 1.

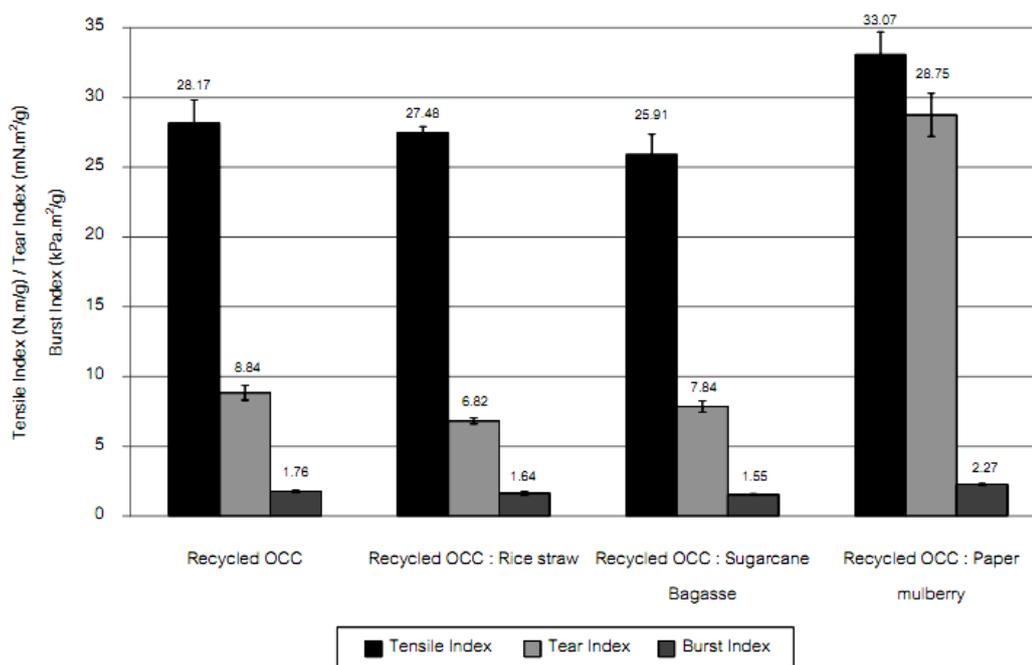


Figure 1. Mechanical properties of recycled OCC handsheets.

The results show that the mechanical properties of handsheets made from 70% of recycled OCC pulp and 30% of rice straw pulp (or sugarcane bagasse pulp) were not much different from those of handsheets made from recycled OCC pulp alone. In contrast, the mechanical properties, i.e. tear, tensile, and burst index, increased tremendously when 30% of paper mulberry pulp was combined to 70% of recycled OCC pulp to form the handsheets. The increase of paper strength properties attributed

to high strength of the individual paper mulberry fiber which has very long length and good flexibility, when compared to the shorter fibers from rice straw and sugarcane bagasse. Therefore, in this study, the addition of virgin paper mulberry pulp can enhance the strength of recycled OCC paper.

Effect of dry strength resin on the mechanical properties of recycled OCC paper

From the previous experiments, the paper made from 70% of recycled OCC pulp and 30% of paper mulberry pulp was chosen to study the mechanical property enhancement by using chemical additives. Herein, we focused on the effect of dry strength resin, polyacrylamide resin, on paper mechanical properties. The procedure used for making recycled OCC paper (handsheets) was described in the materials and methods section. The amount of chemicals used in handsheet forming was 1% aluminium sulfate octadecahydrate, 0.5% sizing agent, 0.5% wet strength resin, and a desired amount of polyacrylamide resin. Addition amounts of chemicals were based on oven dry weight of the pulp.

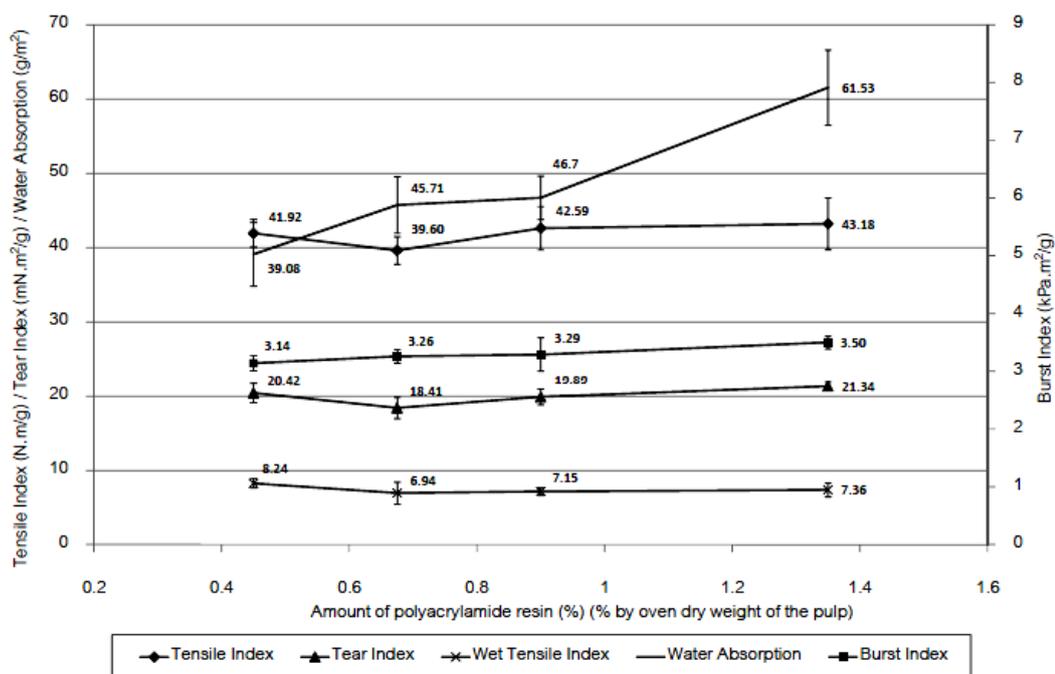


Figure 2. Effect of addition amount of polyacrylamide resin on mechanical properties of recycled (OCC/paper mulberry) paper.

First, we used 0.45% of polyacrylamide resin for preliminary study of the paper mechanical properties after chemical addition. The results show that tensile and burst strength of recycled (OCC/paper mulberry) paper improved after the addition of chemical additives in comparison to control recycled (OCC/paper mulberry) paper from the previous experiment. However, the addition of chemicals diminished tearing resistance of the paper. Moreover, the addition of sizing agent improved water resistance property of the recycled (OCC/paper mulberry) paper, since the water absorption value of the recycled paper reduced from $>100 \text{ g/m}^2$ of control recycled paper to about 39 g/m^2 after

adding sizing agent. Next, the effect of the amount of dry strength resin, polyacrylamide resin, on the paper mechanical properties was determined. In this study, we varied the amount of polyacrylamide resin which was 0.45%, 0.675%, 0.9%, and 1.35% of oven dry weight of the pulp. The effect of the amount of polyacrylamide resin on the mechanical properties of recycled (OCC/paper mulberry) paper was illustrated in Figure 2. As you can see, burst and tear index of the recycled paper increased only a small value after increasing the addition amount of polyacrylamide resin from 0.45% to 1.35%. In addition, the increase of dry strength resin amount showed no effect on tensile and wet tensile strength of the paper ($p>0.05$). Therefore, adding more dry strength resin did not give satisfactory results in the improvement of paper strength properties as expected. Moreover, water absorption of the recycled paper increased from 39.08 g/m² to 61.53 g/m² when the addition amount of polyarylamide resin increased from 0.45% to 1.35%, respectively. Therefore, water resistance property of the recycled paper was diminished by adding more polyacrylamide resin.

Effect of chitosan coating on the mechanical properties of recycled OCC paper

The previous section showed that the addition of polyacrylamide resin at 0.45% improved overall mechanical properties of the recycled paper. However, the addition of polyacrylamide resin more than 0.45% did not show any improvement of the mechanical properties of the recycled paper. Since the enhancement of paper strength properties by chitosan has been reported (Kuusipalo *et al.*, 2005), in this step, we studied whether the mechanical properties of recycled (OCC/paper mulberry) paper can be improved by chitosan coating or not. The recycled (OCC/paper mulberry) paper was prepared by using the same method as in previous section, and the amount of chemicals used was 1% aluminium sulfate octadecahydrate, 0.5% sizing agent, 0.5% wet strength resin, and 0.45% polyacrylamide resin. The prepared recycled paper was then coated with chitosan solution at different concentration, i.e. 0.1%, 0.3%, and 0.5% (w/v). Coating weight of chitosan was 3.88±0.28, 4.29±0.24, and 5.20±0.69 g/m² for chitosan solution at 0.1%, 0.3%, and 0.5% (%w/v), respectively. An increase of coating weight was in accordance with an increase in concentration of chitosan coating solution. The mechanical properties of chitosan-coated recycled paper were illustrated in Figure 3. From statistical analysis using DMRT method, concentration of chitosan coating solution affected on mechanical properties of the recycled (OCC/paper mulberry) paper ($p\leq 0.05$). As you can see in Figure 3, the paper strength properties, such as tensile, wet tensile, and burst index, increased after chitosan coating. The increase in these strength properties can be explained by the formation of hydrogen bonding between chitosan and cellulose fibers that strengthens bonding between fibers. These properties sharply increased until the concentration of chitosan coating solution reached 0.3%, and then slightly increased when the chitosan concentration increased from 0.3% to 0.5%. In contrast, tear index of the recycled paper slightly decreased after chitosan coating. Moreover, coating recycled paper by chitosan appears to slow down water absorption when the absorption is being evaluated with the Cobb method.

Therefore, chitosan coating improved water resistance of the paper. The water resistance improved tremendously after coating the paper with 0.3% chitosan solution but didn't show any improvement after increasing the chitosan concentration from 0.3% to 0.5%. Therefore, the overall mechanical properties of the recycled (OCC/paper mulberry) paper improved by chitosan coating, and in this study, the chitosan concentration of 0.3% was enough to use as paper coating solution, and to improve mechanical properties of the recycled paper.

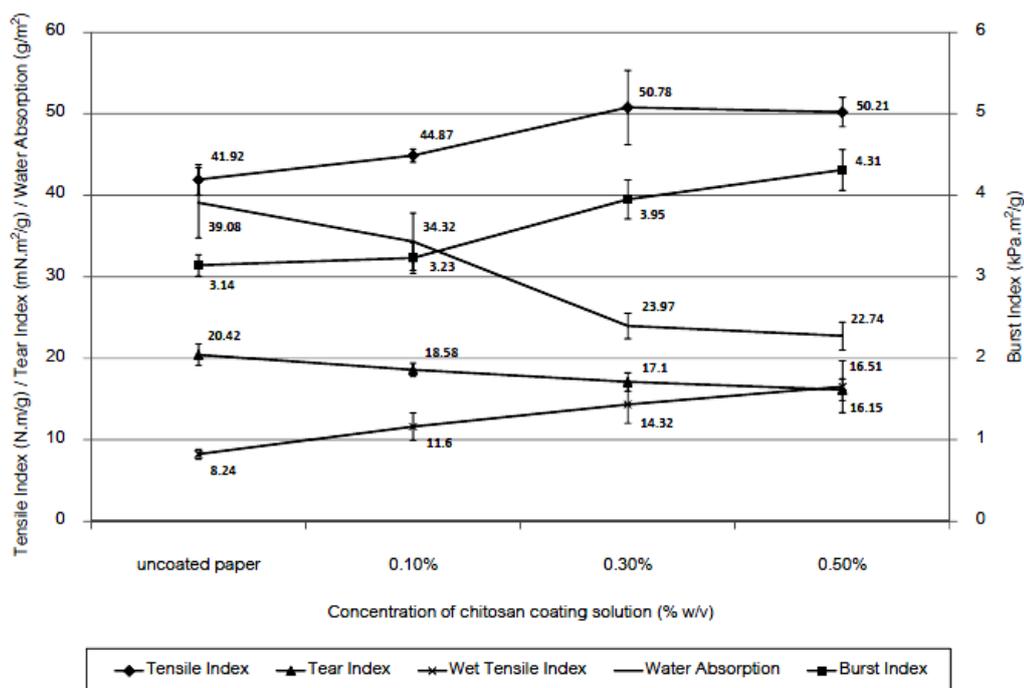


Figure 3. Effect of chitosan concentration on mechanical properties of recycled (OCC/paper mulberry) paper.

CONCLUSIONS

The mechanical property enhancement of recycled OCC paper was examined in this study. The results show that the strength of recycled OCC paper can be improved by forming the recycled paper with 30% of virgin paper mulberry pulp and 70% of recycled OCC pulp, without adding any chemicals and coating. In addition, the overall mechanical properties of the recycled (OCC/paper mulberry) paper can be enhanced by adding chemical additives. However, there was a limit of using chemical additives. The addition amount of dry strength resin over 0.45% (% of oven dry weight of pulp) didn't show any improvement of paper strength properties, and the water resistance property of the paper decreased when using dry strength resin higher than 0.45%. Moreover, in this study, the mechanical property enhancement of recycled (OCC/paper mulberry) paper by chitosan coating was also examined. The results show that the overall mechanical properties of the recycled (OCC/paper mulberry) paper

improved tremendously by chitosan coating, and in this study, the optimal chitosan concentration for paper coating was 0.3% (%w/v).

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