# Preparation and Some Mechanical Properties of Composite Materials Made from Sawdust, Cassava Starch and Natural Rubber Latex

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*Abstract*—The composite materials were prepared by sawdust, cassava starch and natural rubber latex (NR). The mixtures of 15%w/v gelatinized cassava starch and 15%w/v PVOH were used as the binder of these composite materials. The concentrated rubber latex was added to the mixtures. They were mixed rigorously to the treated sawdust in the ratio of 70:30 until achive uniform dispersion. The batters were subjected to the hot compression moulding at the temperature of 160°C and 3,000 psi pressure for 5 min. The experimental results showed that the mechanical properties of composite materials, which contained the gelatinized cassava starch and PVOH in the ratio of 2:1, 20% NR latex by weight of the dry starch and treated sawdust with 5%NaOH or 1% BPO, were the best. It contributed the maximal compression strength ($341.10 \pm 26.11$ N), puncture resistance ($8.79 \pm 0.98$ N/mm$^2$) and flexural strength ($3.99 \pm 0.72$N/mm$^2$). It is also found that the physicochemical and mechanical properties of composites strongly depends on the interface quality of sawdust, cassava starch and NR latex.

*Keywords*—Composites, Sawdust, Cassava starch, Natural rubber (NR) latex, Surface chemical treatments.

## I. INTRODUCTION

THE composite materials in these experiments are based mainly on natural polymer, e.g. natural rubber latex, cellulose and starch. They have a much lower undesirable impact on the environment since they are made from renewable resources. Sawdust and wood flour is the most common wood filler used in wood-plastic composites and other wood-alternative material composites has made major advances in material performance. It is typically a post-industrial source consisting of wood shavings, chips, and sawdust produced by secondary wood product [1]. The composites of sawdust and adhesive from gelatinization of cassava starch and natural rubber (NR) latex are polymeric material displaying excellent physical properties. However the mixtures of starch, NR latex and sawdust when prepare composite by hot compression moulding have a high degree of hardness and are brittle suitable strength.

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The performance and stability of sawdust-reinforced composite materials depends on the development of coherent interfacial bonding between sawdust and matrix. The general components of sawdust are cellulose, hemicellulose, lignin, pectin, waxes, and water-soluble substances [2], [3]. Cellulose, hemicellulose, and lignin are the main components contributed the strength, flexural, and impact properties of the composites. Moreover the bonding between sawdust and the hydrophobic matrix has effected to the mechanical properties of the composite material.

The major problems of wood composites are associated with the hydrophilic character of the cellulose structure of fiber. To overcome these problems, it is necessary to modify the fiber surface by the physical and chemical methods to reduce the hydrophilic nature of the sawdust and improve the sawdust–matrix bonding. In general, the surface modification by using suitable chemical treatments of the fiber [4]; A significant increase in the strength of composites after treatment of the sawdust have shown that the interface quality of sawdust, cassava starch and NR latex composites could be enhanced through different surface modifications, such as dewaxing, alkali treatment, cyanoethylation, and benzolation. The alkalization and benzoylation were then applied to sawdust and their influence on the chemical composition and surface structure of the sawdust and matrix adhesion.

The goal of the present work was to studies the use of a hot compression moulding process to prepare the composites based on cassava starch, NR latex and sawdust. It was also aimed to assess the effect of varying the proportion of these three components on the physicochemical and mechanical properties of composites, and the relationship between these mechanical properties and morphology in composites.

## II. MATERIAL AND METHODS

### a. Material and Chemical

Cassava starch, also known as tapioca starch (13.01% moisture) was obtained from Tapioca Development Co. Ltd, Bangkok, Thailand. Natural rubber latex (High ammonia concentrated natural latex, HA TZ type 60% solids) was purchased from Chalong Latex Industry Co. Ltd, Songkhla, Thailand.

Polyvinyl alcohol; PVOH (Fully hydrolyzed PVOH No.26) (CCP brand) was 98.5-99.2% hydrolyzed, with molecular weight of 112,000-120,000 (BF-26) daltons. It was purchased from Chang Chun Petrochemical Co. Ltd, Taipei, Taiwan. Nonidet P40 (a non-ionic detergent/surfactant) was purchased from Fluka, Switzerland. Sawdust (Mesh No.10-20) was obtained from Plan Creations Co. Ltd, Trang, Thailand.

*b. Preparation of sawdust surface treatments by Alkalization*

The untreated sawdust, screened with sieve mesh no. 10–20, was placed in a glass vessel containing NaOH solution of various concentrations (1, 3 and 5% w/v). The sawdust was soaked in NaOH solution at room temperature with continuous stirring for 18 hours. The sawdust was then washed several times with acetone and distilled water to eliminate the adsorbed alkali. The alkali-treated sawdust was later dried in a hot-air oven at 70°C and kept it in a dessicator for subsequent analysis.

*c. Preparation of sawdust surface treatments by Benzoylation*

The untreated sawdust (46 g) was mixed with 1 wt % of sawdust BPO (0.46 g) dissolved in toluene (2 L) in a round-bottom glass vessel. The amounts of BPO used varied from 1, 3, and 5 wt% of fiber. The reaction was carried out at 80°C with stirring for 1 hour under $N_2$ atmosphere. The fibers were removed from the reactor and washed thoroughly with toluene. They were dried at 70 °C.

*d. Preparation of Sawdust, Cassava starch and Natural rubber latex blends*

The non-ionic surfactant was added to NR latex and stirred at room temperature for a few seconds to obtain a stabilised natural rubber latex. Cassava starch with 15% by weight of water in combination was stirred and heated until clarity. PVOH (BF 26) with 15% by weight of water was mixed with starch with 15% by weight of water at the ratio of 1:0, 1:2, 1:1, 2:1 and 0:1 homogeneously and then mixed with the natural rubber latex and sawdust in the container. The composites of all the blend preparation was showed in Table

1. The mixtures were mixed at room temperature for 15–20 min to obtain a good dispersion of the mixtures.

*e. Natural rubber latex blends*

Cassava starch, natural rubber latex (NR), non-ionic surfactant (Nonidet P40) [5], and treated sawdust with NaOH and BPO, were mixed according to the Table 2. Gelatinised starch mixed with PVOH adhesive at the ratio of 2:1 was then mixed with the latex and sawdust in a container equipped with a stirrer. The mixture was stirred at room temperature for 15–20 min to obtain a good dispersion of the mixtures.

*f. Composite Forming by Hot compression moulding*

The mixtures, according to Table 1 and Table 2, were introduced into the mould and compressed at the temperature of 160°C. The moulding time and pressure of forming procedure were 5 min and 3,000 psi, respectively.

*g. Analyses*

Density of composites materials were calculated by the relationship between weight and volume. Reported values were averages of five determinations for each formulation.

Mechanical properties of composite materials were determined as compression strength, puncture resistance and flexural strength. Compression strength was examined according to ASTM D642 by Universal Tester, LLOYD Instruments (Model LR30K). Puncture resistance measurements were performed in a statistic testing procedure according to ASTM D781 and flexural strength according to ASTM D790-92. For the flexural testing, a three point bending with a span of 125 mm. was used and the crosshead speed was set at 2.5 mm/min.

*h. Statistical analysis*

A completely randomized experimental design was used to characterize the composites. Analysis of variance (ANOVA) was used to compare mean differences of the samples. If the differences in mean existed, multiple comparisons were performed by using Duncan Multiple Range Test (DMRT).

TABLE I
FORMULATION OF SAWDUST, CASSAVA STARCH AND
NATURAL RUBBER LATEX BLENDS

| Sample | Sawdust (% of the mixtures) | Starch and PVOH adhesive (% of the mixtures) | NR latex (% of starch) | Nonidet P40 (% of NR latex) |
|---|---|---|---|---|
| 1 | 30 | 70 | 20 | 1.5 |
| 2 | 30 | 70 | 35 | 1.5 |
| 3 | 30 | 70 | 50 | 1.5 |

TABLE II
FORMULATION OF TREATED SAWDUST, CASSAVA STARCH AND NATURAL RUBBER LATEX BLENDS

| Sample | Concentration of NaOH (% w/v) | Amount of BPO (wt % of fiber) | Sawdust (% of the mixtures) | Starch and PVOH adhesive (% of the mixtures) | NR latex (% of starch) | Nonidet P40 (% of NR latex) |
|---|---|---|---|---|---|---|
| 1 | 1 | - | 30 | 70 | 20 | 1.5 |
| 2 | 3 | - | 30 | 70 | 20 | 1.5 |
| 3 | 5 | - | 30 | 70 | 20 | 1.5 |
| 4 | - | 1 | 30 | 70 | 20 | 1.5 |
| 5 | - | 3 | 30 | 70 | 20 | 1.5 |
| 6 | - | 5 | 30 | 70 | 20 | 1.5 |

## III. RESULTS AND DISCUSSION

### a. Effect of Adhesive ratio between Starch and PVOH on The Mechanical Properties of Composites

The solid content and viscosity of adhesive were presented in Table 3. The results showed that the viscosity of adhesive was increased with increasing the starch content. However, the solid content of adhesive were not much different. One reason for this may be the swelling and solubility characteristics of starch and PVOH in water. Starch granule could swelling higher than PVOH with the same solid content. The higher solid content of adhesive shows the higher bonding strength [6]. and the viscosity of adhesive. The higher viscosity of adhesive resisted the flow and sheared the lower of adhesive dispersion in composite materials.

TABLE III
SOLID CONTENT AND VISCOSITY OF ADHESIVE

| Starch : PVOH | Solid content (%) | Viscosity (cp) |
|---|---|---|
| 1 : 0 | 13.43 | 40,050 |
| 1 : 1 | 14.37 | 19,310 |
| 2 : 1 | 14.22 | 21,900 |
| 1 : 2 | 14.83 | 16,810 |
| 0 : 1 | 15.21 | 16,150 |

The effects of adhesive composition between starch and PVOH on the mechanical properties of composites were showed in Table 4. The compression strength and the puncture resistance of the composites made from the starch and PVOH in the ratio of 2:1 was the highest (1044.98 N and 164.30 N/mm respectively). On the other way, the composites made from the starch and PVOH in the ratio of 1:1 contributed the lowest compression strength (475.63 N) and the composites made from the starch and PVOH in the ratio of 0:1 contributed the lowest puncture resistance (11.81 N/mm). However, the density of composites was not much different.

TABLE IV
EFFECT OF ADHESIVE RATIO BETWEEN STARCH AND PVOH ON THE MECHANICAL PROPERTIES OF COMPOSITES

| Starch : PVOH | Density[a] (g/cm$^3$) | Compression strength[a] (N) | Puncture resistance[a] (N/mm) |
|---|---|---|---|
| 1 : 0 | 0.60±0.01a[b] | 832.67±2.76c | 55.48±0.49a |
| 1 : 1 | 0.59±0.01a | 475.63±13.66a | 105.10±1.38b |
| 2 : 1 | 0.61±0.01a | 1044.98±7.49d | 164.30±0.84e |
| 1 : 2 | 0.59±0.01a | 656.20±13.07b | 12.05±0.96d |
| 0 : 1 | 0.60±0.01a | 1461.67±6.03e | 11.81±0.66c |

[a]Mean ± SD from five determinations.
[b]The different superscripts in the same column indicate the significant differences ($p<0.05$).

The composites contributed the appropriate compression strength and puncture resistance thus it can be as a package.

The composites made from starch and PVOH in the ratio of 2:1 showed the highest compression strength and puncture resistance. The interaction between hydroxyl group (-OH) of starch and hydroxyl group (-OH) of PVOH adhesive - The hydrogen bonding occurred between starch and PVOH have enhanced the strong adhesion so the composite products contributed the higher mechanical properties[7].The adhesive that have a starch content higher than a PVOH content could increase the hydrogen bonding of adhesive. So the composite prepared from adhesive that higher starch content, it will shown the higher compression strength and puncture resistance.

### b. Effect of Natural rubber latex concentration on The Properties of Composites made from Sawdust and Cassava starch

Table 5 showed the results of density, compression strength, puncture resistance and flexural strength of composites.

TABLE V
EFFECT OF NATURAL RUBBER LATEX CONCENTRATION ON THE PROPERTIES OF COMPOSITES MADE FROM SAWDUST AND CASSAVA STARCH

| NR latex, % of dry starch | Density* (g/cm$^3$) | Compression strength* (N) | Puncture resistance* (N/mm) | Flexural strength* (N/mm$^2$) |
|---|---|---|---|---|
| 0 | 0.62±0.01a** | 1044.98±7.49c | 164.3±0.84c | 1.65±0.41a |
| 20 | 0.72±0.01b | 341.08±26.11b | 8.79±0.98b | 3.24±0.84b |
| 35 | 0.72±0.01b | 315.72±35.93a | 7.83±0.27b | 3.02±0.34b |
| 50 | 0.73±0.01b | 344.88±30.51b | 6.39±0.96a | 1.76±0.30a |

[a]Mean ± SD from five determinations.

[b]The different superscripts in the same column indicate the significant differences (p<0.05).

These results showed that the addition of NR latex concentration of 20, 35 and 50 % by weight of dry starch to the composites. The density of the composites with and without the NR latex was not much different. The compression strength of the composites with 50% NR latex was the highest (344.88 N) and the composites with 35% NR latex was the lowest compression strength (315.72 N). The puncture resistance and the flexural strength of the composites made by added 20% NR latex was the highest (8.79 N/mm and 3.24 N/mm$^2$ respectively) and decrease with increasing the NR latex. Puncture resistance of composites with 50% NR latex was the lowest (6.39 N/mm and 1.76 N/mm$^2$ respectively). However, the compression strength and the puncture resistance of the composites have decreased when the composites had the NR latex in the composites. On the other way, the density and the flexural strength of the composites have increased with having the NR latex as the additive.

Considering the characteristics of the composites, it can be explained that NR latex contributed the elastic property to the composites. When it was added to the composites, it tended to reduced the brittle failure of the blend [8]. It can be observed by the increasing of the flexural strength when the composites have the NR latex. However, the NR particles are smaller than the cassava starch particles and could probably coagulate and cover the starch granules to generate a mechanical resistance, which slows the gelatinization of the starch granules [5]. When the NR latex content exceeded 20 by weight of dry starch, NR latex particles inhibited starch gelatinization and might be coagulation and could not be dispersed in the gelatinized starch in the mixing process and it had effected to the decreasing of the flexural strength. Thus suitable amount of NR latex concentration 20% by weight of dry starch, was used.

Despite the fact that the contrast between starch and NR latex was poor, it was possible to identify the two distinct phases [9]. Adding NR latex to starch-based materials without any external plasticizer does not result in any improvement of the materials properties [10]. A non-ionic surfactant (Nonidet P40) was selected to stabilise the NR latex in the gelatinised starch. The poly(ethylene glycol) part of Nonidet P40 protruding into the water boundary provided steric stabilization to the suspended NR particles[5]. It should be emphasised that the minimum amount of Nonidet P40 used to prevent the coagulation of NR latex and to disperse NR latex in gelatinised starch was 1·5% by weight of the NR latex.

*c. Effect of surface treatment of sawdust on The Properties of Composites made from Sawdust, Cassava starch and Natural rubber latex*

The chemical treatment of sawdust aimed to improve the adhesion between the sawdust surface and the hydrophobic polymer matrix in this experiment, sodium hydroxide and benzoly peroxide were carried out to improve the bonding at the sawdust -polymer interface.

### 3.3.1 Alkalization

Alkaline treatment or mercerization is one of the most popular chemicals using for treatment of natural fibers. The important modification done by alkaline treatment is the disruption of hydrogen bonding in the network structure, thereby increasing surface roughness. This treatment removes a certain amount of lignin, wax and oils covering the external surface of the fiber cell wall, depolymerizes cellulose and exposes the short length crystallites. Addition of aqueous sodium hydroxide (NaOH) to natural fiber promotes the ionization of the hydroxyl group to the alkoxide, as in (1).

$$\text{Fiber} - \text{OH} + \text{NaOH} \rightarrow \text{Fiber} - \text{O} - \text{Na} + \text{H}_2\text{O}. \qquad (1)$$

Thus, alkaline processing directly influences the cellulosic fibril, the degree of polymerization and the extraction of lignin and hemicellulosic compounds [2].

### 3.3.2 Benzoylation

Benzoylation is an important transformation in organic fiber treatment. Benzoyl chloride includes benzoyl ($\text{C}_6\text{H}_5\text{C}=\text{O}$) which is attributed to the decreased hydrophilic nature of the treated fiber and improved interaction with the hydrophobic matrix. The reaction between the cellulosic hydroxyl group of the fiber and benzoyl chloride is shown in (2) and (3).

$$\text{Fiber} - \text{OH} + \text{NaOH} \rightarrow \text{Fiber} - \text{O}^-\text{Na}^+ + \text{H}_2\text{0}. \qquad (2)$$



$$(3)$$
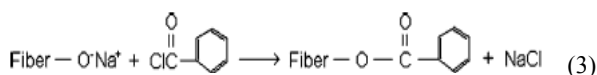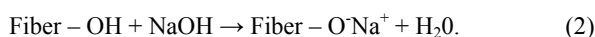
Benzoylation of fiber improves fiber matrix adhesion, thereby considerably increasing the strength of composite, decreasing its water absorption and improving its thermal stability [2].

The effect of chemical modifications on the sawdust treatments was related to the mechanical properties of composites. The flexural strength, compression strength and puncture resistance of composites treated with NaOH and BPO were presented in Table 6.

TABLE VI
DENSITY AND FLEXURAL STRENGTH OF COMPOSITE CONTAINING DIFFERENT PERCENTAGE OF NaOH-TREATED SAWDUST AND BPO-TREATED SAWDUST

| Sample | Concentration of NaOH, % w/v | Amount of BPO, wt % of fiber | Density (g/cm$^3$) | Flexural strength (N/mm$^2$) |
|---|---|---|---|---|
| Untreated | - | - | 0.72±0.01 | 3.24±0.84 |
| 1 | 1 | - | 0.72±0.01 | 2.12±0.62 |
| 2 | 3 | - | 0.72±0.01 | 3.83±0.33 |
| 3 | 5 | - | 0.72±0.01 | 3.99±0.72 |
| 4 | - | 1 | 0.72±0.01 | 3.26±0.84 |
| 5 | - | 3 | 0.72±0.01 | 2.07±0.44 |
| 6 | - | 5 | 0.72±0.01 | 2.26±0.31 |

The flexural strength of composites was showed in Table 6. The flexural strength of the composites was the highest when treared the sawdust with used NaOH solution in the concentration of 5% w/v (3.99 N/mm$^2$) or BPO in the concentration of 1%wt of fiber (3.26 N/mm$^2$). NaOH solution and BPO improve the bonding between sawdust and polymer

matrix. However, the density of composites was not different. This is because of poor adhesion between the untreated fiber and the NR latex. The fracture surfaces in the cases of rubber composites containing 1, 3 and 5% NaOH-treated fibers show the better adhesion between the fibers and rubber matrix, as indicated by the presence of rubber phase adhering to the fibers. The results obtained from the various concentrations of NaOH solution showed that the 5% NaOH solution produced the best mechanical properties. On the other way, The composites reinforced with BPO-treated fibers (1, 3, and 5 wt % of fiber) showed the higher flexural strength comparing to untreated fiber composites. It was due to the increasing of hydrophobic property on these fiber surfaces and making the surfaces more compatible with natural rubber. Table 6, showed the flexural strength of composites rapidly increase when using 1% BPO. However, as the amount of BPO is further increased, the flexural strength began to fall off. The 1% BPO-treated fiber composite improved the flexural strength, when compared to those of untreated fiber composites. The slight reduction in mechanical properties of the composites with BPO was greater than 1% warrants further study. The flexural strength of untreated and treated fibers should be measured to explain the mechanical behavior of BPO-treated fiber composites. The rubber matrices of treated fiber composites have the higher bonding when compared with untreated fiber composites. From these experimental results, the optimal concentrations of alkali solution and BPO using for fiber surface treatments are 5% (w/v) and 1% (by weight of fiber), respectively.

## IV. CONCLUSION

The composite materials can be prepared from sawdust, cassava starch and natural rubber latex on various proportions. The composite materials which contained the gelatinized cassava starch and PVOH in the ratio of 2:1, natural rubber latex 20% by weight of the dry starch and treated sawdust with 5% NaOH or 1% BPO were the best. It contributed the maximal compression strength ($341.10 \pm 26.11$ N), puncture resistance ($8.79 \pm 0.98$ N/mm) and flexural strength ($3.99 \pm 0.72$ N/mm$^2$). It is also found that the mechanical properties of composites strongly depend on the interface quality of sawdust, cassava starch and natural rubber latex.

## ACKNOWLEDGMENT

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