The Study of Eco-friendly Printing Ink Composed of Starch/PVA Blend for Printing On Cardboard

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Keywords: starch, PVA, flexography, printing ink, cardboard

Abstract

Cardboard cartons such as corrugated board are secondary packaging that holds together the individual units of a good to deliver mass quantities of the good. Many trashes of cartons are disposed to environment, or brought for making recycled paper. Purpose of this research was to identify natural resource materials to develop an environmental friendly flexographic ink that can be safety recycled or decomposed in landfills. Because starch was a resin derived from natural resource, non-toxic, and less pollution, so it was used as a natural binder for making water based printing ink. The solution of starch was blended with polyvinyl alcohol (PVA) solution by shear mixing. PVA was mixed into starch solution to improve flowing behavior, film formation and flexibility of the ink.

This work presents the study of water based flexographic printing inks composed of solution blending of starch and PVA. Blending variance of different starches were evaluated for the best printed quality and ink performances, against a typical synthetic water based flexographic ink. The starch/PVA inks prepared from cassava, rice, or corn starches were compared on their properties and print density. Next, the cassava starch/PVA ink was printed on a corrugated board as a substrate, printing by a flexographic printing machine. Print quality was compared to that of a typical flexographic ink composed of a synthetic binder. These studies were concluded that the ink properties: viscosity, pigment dispersion, and fastness of the starch/PVA inks were suitable for flexographic printing on corrugated cardboard. Print quality showed good print density, good sharpness of printed text and line, but % dot gain of halftone screen on the printed samples was more than that of the typical flexographic ink.

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Introduction

Corrugated paperboard box, or carton is used as a secondary packaging for collecting individual units to transfer goods to customer. Currently, this kind of box packaging had high demand for B to C, C to C, and C to B of e-commerce. It is used promptly before throwaway as household waste. However, it made from an environmental friendly material; kraft paper with recycled paper. Unlike plastic, paper can be bio-degraded in nature. (Dixson-Hardy, 2009; Fishbein, 1996; Geueke et.al., 2018; Landi et al., 2020; Antonio et al., 2021). There is a report that in year 2007, Thailand is in the top ranks in generating huge amount of organic waste in the developing countries. 40% of the components are plastic, paper, metal, and glass (Chinda et al, 2012).

Printing process for a corrugated paperboard is not costly with not much of design. Flexographic printing is commonly a printing process on the corrugated paperboard. A flexible rubber printing plate of flexography is suitable to impress and ink-transfer on rough, uneven surface, and flutes of the corrugated paperboard. Flexographic platemaking methods are divided into rubber-cutting, rubber-molding, and UV light exposure on a photopolymer. A printing ink is commonly composed of colorants, binders, solvents, and additives. Usually, an ink component called binder is a polymeric resin made from chemicals. A flexographic ink for printing on a corrugated paperboard is commonly a water-based ink with drying on paper by penetration mechanism. (Thomson, 1998; Todd, 1994)

Starch is available in renewable resources, as well as being used in various applications, and it can be used as a binder in the paper and textile industries. (Pizzi et al., 2003) It is a natural polymer, which is polymeric carbohydrate consists of two types of molecules: amylose and amylopectin. Starch mainly found in seeds, roots, and tubers. The presence of minor constituents of lipids, phosphate ester groups, and proteins is usually found in starch. These starch components depend on botanical source, and determine the functionality as well as end-use of starch. Usually, native starches present inappropriate characteristics for most industrial applications and, hence, they must be modified by chemical and/or physical methods in order to broad their usage possibilities (Villar, 2018). Starch is extracted from flour. Gelatinisation of starch is an initial stage to prepare starch paste as a process of breaking down the intermolecular bonds of starch molecules in the presence of water and heat, allowing swelling phenomenon. Amylose and amylopectin contents consist in starch mainly effect to gelatinisation with difference in starch varieties. Chemical treating of starch with NaOH is a process that to lower cooking temperature to accelerate gelatinisation of the starch granule as the Stein-Hall process. (BeMiller et al, 2009; Bajpai, 2018; Emblem, 2012; Biduski et al, 2018; Chisenga et al., 2019, Chaiwanichsiri et al, 2012; Emengo et al, 2002; Imam et al, 2001; Pereira, 2011)

Polyvinyl alcohol is a water-soluble synthetic polymer with biodegradable property under both aerobic and anaerobic conditions. This polymer is widely used by blending with other biopolymers and other hydrophilic polymers including starch. There are many reports about mechanical properties of the blended polymers enhanced by polyvinyl alcohol because of compatible structure and hydrophilic properties (Halima et al, 2016; Chiellini, 2003; Jain et al., 2017; Follain et al, 2005)

It is suggested as this work that both materials of a corrugated box, or carton, and a printing ink are eco-friendly materials. So, an ink printed on this secondary packaging could also made from a natural-derived to benefit on environment, e.g., safety decomposed in a landfill, and harmless substance contaminated in a recycled pulp. There are many of documents previously published involving an eco-friendly printing ink, and materials. (Karmaus et al., 2018; Liu et al., 2021; Ding et. al., 2020; Alam et al., 2014; Bener et al., 2020; Ha et al., 2012, Pereira et al, 2011)) This work was to study on a natural derived material as a natural binder to develop an environmental friendly flexographic ink for printing on a corrugated paperboard. Three starches of cassava, rice, and corn were studied at variant amount blended to polyvinyl alcohol (PVA). Since starch unfortunately did not provided long length for ink transferring in a flexographic printing press, therefore blending of PVA was to improve length of ink. These starch/PVA blends were compounded in a water based flexographic ink, and then properties of the inks were identified to determine an ink formulation for printing on a corrugated paperboard.

Experimental

Study on starch/PVA blends of cassava, rice, and corn starches suitable for making a flexographic ink

Preparation of starch/PVA blend: Food grade starches of corn as well as cassava starch, and rice flour were used as purchased from Thai Flour Industry Co., Ltd, Thailand. Firstly, aqueous starch solution was prepared by cooking in distilled water using an overhead stirrer, heating in a range of 70-80◦C. Gelatinisation temperature of starch was accelerated with NaOH, 1M conc. An amount of sodium hydroxide added in the starch dispersion as shown in Table 1 was depended on starch types.

Polyvinylalcohol (PVA), molecular weight of 93000 Da, 98-99% hydrolysis, pH 5-7 (5%) from Ajax Finechem, was used as purchased. PVA was dissolved into distilled water, stirring with heating at 70◦C until the PVA solution was complete. PVA solution was blended into the corn, rice, or cassava starch solutions at three various ratios as in Table 1.

Ingredients	% weight (amounts of NaOH)		
Cassava starch (NaOH 1M) or, Rice starch (NaOH 1M) or, Corn starch (NaOH 1M)	5 (15ml) 5 (45ml) 5 (1.2ml)	3 (3ml) 3 (10ml) 3 (5ml)	7 (5ml) 7 (30ml) 7 (15ml)
PVA	5	7	3
Water	90		

Table 1. *Recipes of starch/PVA blends*

Blending of the starch and PVA solutions was prepared by shear mixing using an overhead stirrer at stirring speed in a range of 1000-1500 rpm. Then, the starch/PVA blends were brought to measure their viscosity using a Brookfield rotational viscometer (DV-E) measuring as function of shearing rate in a range of 10-100 rpm. The mixture of starch and PVA blend was observed visually.

Microstructures of starch/PVA blend films: Microstructures of starch/PVA blends were observed. Photomicrograph was captured using a scanning electron microscope (SEM) at an operating voltage of 10kV on a gold metallized coated specimen of the starch/PVA blend film.

Properties of a water based flexogarphic ink composed of the starch/PVA blend

Ink preparation: Water based flexographic printing ink was prepared as following ingredients: a cyan pigment (Fastogen blue 5380, with hydrophilic surface treatment for water-based printing ink) supported by Dainippon Ink and Chemicals, Japan, a nonionic surfactant (alcohol ethoxylate, HLB 12). At first step, the cyan pigment at 10%wt of total ink was mixed into the starch/PVA as well as other ingredients as in Table 2. Second step, these ingredients were grinded in a milling container with glass beads (Ø 1.00-1.25 mm, Sigmund Lindner Gmbh, Germany) using a shearing mixer, stirring at a speed of 3000 RPM for 3 hours. Pigment dispersibility in the starch/PVA vehicle was identified by plots of fineness of grind against grinding time duration.

Ingredients	Amount (%wt)
Cyan pigment (Fastogen blue 5380)	10
Starch/PVA blend (3:7, 5:5, 7:3 %wt)	10
Nonionic surfactant	0.9
Water	79.1

Table 2: *Compositions in flexographic ink*

Testing of ink property: Properties of the starch/PVA inks were compared among starch types. Fineness of grind, viscosity, flow behavior, print density, fastness to light, water, and abrasion of the inks were found out. Fineness of grind of the milled ink was tested using a grind-o-meter according to ASTM D1316. Viscosity dependency on shearing rate was measured using a rotational viscometer (Brookfield DV-E model). The starch/PVA inks were printed using K printing proofer (RK model) at a printing speed scale of 2.5, pressure scales of doctor blade, rubber roller, and metal roller at 2.1, 1.35, and 1.0 mm, respectively, printing on a plain white paper of 80 gsm. Lightfastness of the printed samples were examined by sunlight exposure for 21 hours, then color changing of the exposed sample was evaluated by color difference (ΔE, CIELAB) to the unexposed sample. Rub resistance was tested using Ugra abrasion tester. A printed sample was rubbed with a metal block having a surface area of 50x50 mm, an abrasive pressure of 0.2 N/cm^2, rubbing for 20 cycles. Evaluating scale of abrasion resistance was divided into 5-step scales (Ugra, 2017) as shown in Figure 1. Water resistance of the ink was tested by means of soaking a printed sample in water for 8 hrs. The soaked sample was dried, and weighted to find weight losing of the ink from the printed sample (%weight-loss). A lower value of %weight-loss implied better water resistance. Print density of a printed sample after completely dried was measured using a densitometer, Vipdens. An average value of print density on the printed sample was reported.

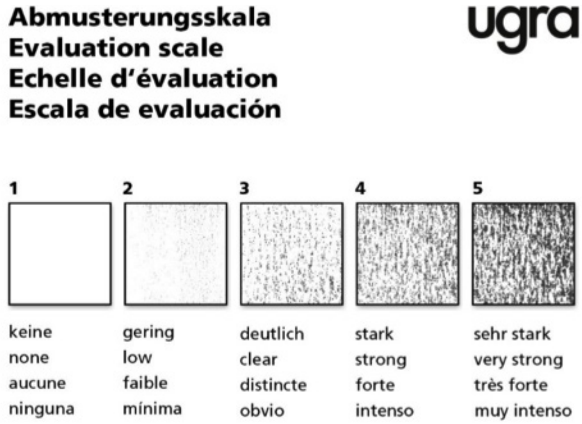


Figure 1. *Rub resistance evaluation scales by UGRA (Ugra, 2017)*

a b

Figure 2. *(a) starch/PVA flexographic inks of cassava starch (left), rice starch (middle), and corn starch (right), (b) ink proofing on paper substrate using K-proofer*

Application of the starch/PVA blend in ink formulation for corrugated paperboard printing

Applying starch/PVA ink to a flexographic printing press and evaluation on print quality: A cassava starch/PVA based ink was applied to a formulation of water based flexographic ink for corrugated paperboard printing as shown in Table 3. Viscosity of the prepared inks was measured using a Zahn flowing cup (#4) prior printing in a flexographic printing machine on a corrugated paperboard: KI kraft liner on B flute. Comparing to a typical flexographic ink, the starch/PVA ink and the typical flexographic ink were printed with the same condition in the printing press: impression, printing speed, and ink temperature. Before printing, a flexographic photopolymer plate was normally exposed and developed by UV light, and chemical developer, respectively; a plate of print testform consisted of dot screening 0-100% at a resolution of 30 LPI, different sizes of texts and lines. After printing, printed densities as well as % dot gain/tone value increase (TVI) were measured on areas of % dot screen using a colorimeter (GretagMacbeth, Spectro Eye). Printed sample of the starch/PVA ink was visually noticed comparing to a printed sample of the typical flexographic ink. In addition, both of the inks: the starch/PVA, and the typical flexographic inks, were demonstrated wetting ability on paper substrate by means of contact angle of the ink onto substrate surface. An ink was dropped onto a KI kraft paper was determined a degree of contact angle (°), which was the angle at the interface where liquid, air, and solid meet (Thomson et al, 1998).

Ingredients	Amount (%wt)	Functions
Pigment, Fastogen blue 5380	12.4	Colorant
Cassava starch solution (10%wt)	5.1	Binder
PVA (10%wt)	35	Binder
Non-ionic surfactant	1.9	Dispersing agent, wetting agent
Water	28.3	Solvent and diluent
Isopropyl alcohol	7.6	Drying accelerator,
Wax	0.5	Slipping agent
Latex additive	9.2	Waterfastness promoter

Table 3. *Formulation of a cassava starch/PVA flexographic printing ink for corrugated paperboard*

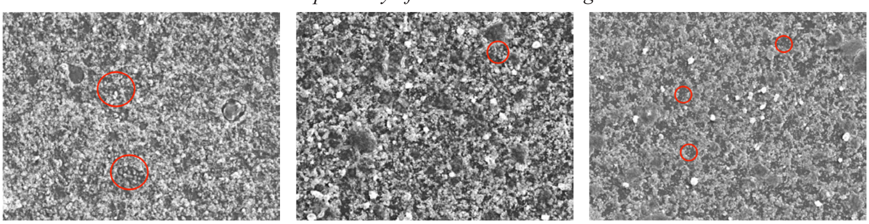
Results and Discussion

Properties of starch/PVA blends

At cooking stage, the amounts of NaOH added for gelatinized the starch as in Table 1 was different between various types of starches. Cassava starch paste was translucent due to high amylopectin composition of tuber-derive starched, whereas grain-derived starches have a higher amylose content. Corn starch used in this work was possible a high amylopectin starch as gelatinization was fast (BeMiller et al., 2009). There were reports that rice starch granule had swelling power and solubility lower than those of cassava and corn. (BeMiller et al, 2009; Hamza et al, 2010). NaOH induced rapid swelling affected to decrease gelatinisation temperature in starch cooking process. In starch molecules, NaOH was added hydroxyl group (starch-OH) transfer into Sodium cation salt (Starch-ONa^{+}), consequently there was repulsion of the same charge promoting swelling and dissolution (Roberts et al, 2002). NaOH possibly altered rheology of aqueous starch solution. Many factors effect to starch gelatinization such as starch granule size, crystallinity, morphology, and minor contents e.g., protein, lipid, ash, mineral, etc. It was that the mixture of starch and PVA solution was miscible. Table 4 showed the cassava starch/PVA mixture at the content ratio of 5%wt to 5%wt had good performance as a distinct layer between two solutions did not form. It was depended on molecular weight and % degree hydrolysis of PVA, as well as starch retrogradation, and molecular composition including amylose and amylopectin (Park et al., 1994; Zanela et al, 2016).

starch : PVA (%wt)	Blending of starch/PVA solution	Stability of starch/PVA blended
5:5		
Cassava Rice Corn	Miscible	Good
3:7		
Cassava Rice Corn	Miscible	A distinct layer formed after left for 3 days
7:3		
Cassava Rice Corn	Miscible	A distinct layer formed after left for 3 days

Table 4. *Compatibility of starch/PVA blending solution*



(a) cassava starch/PVA (b) rice starch/PVA (c) corn starch/PVA

Figure 3. *SEM photomicrograph (x1000) of starch/PVA blends (a), (b), and (c)*

Figure 3 represented photomicrographs of the starch/PVA blend films by SEM to consider on microstructures of the blends. All images showed clusters of starch granules embedded in the blend matrix. It was observable that the starch and PVA was uniformly mixed, appeared the same in all part as in figures of micrographs. However, the rice starch/PVA blend showed rougher surface than those of the cassava and corn starch.

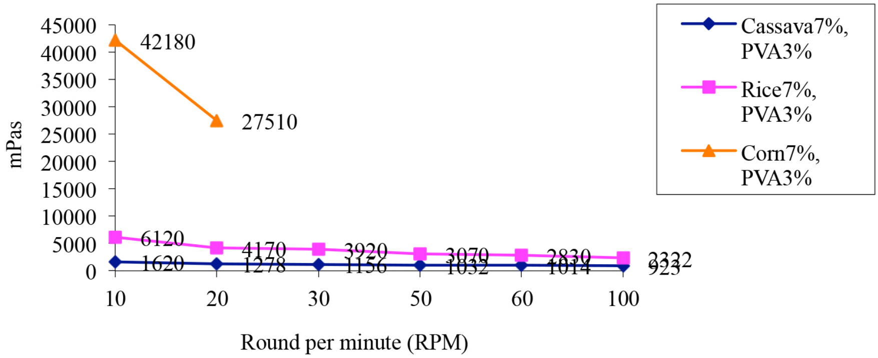


Figure 4. *viscosity dependence on shear rate of starch/PVA blend at 7%wt starch to 3%wt PVA*

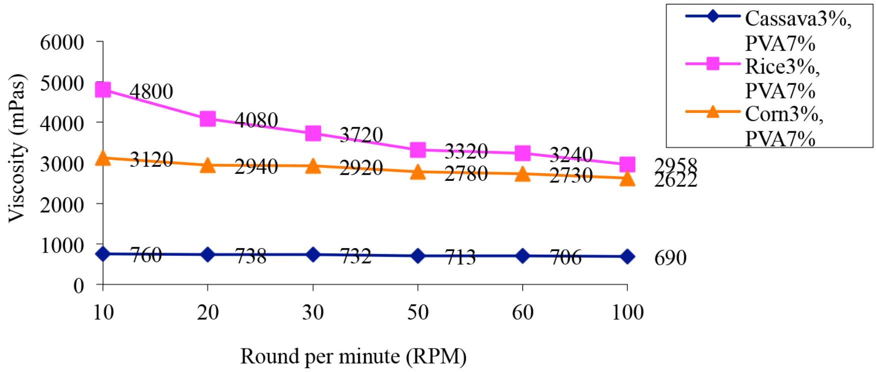


Figure 5. *viscosity dependence on shear rate of starch/PVA blend at 3%wt starch to 7%wt PVA*

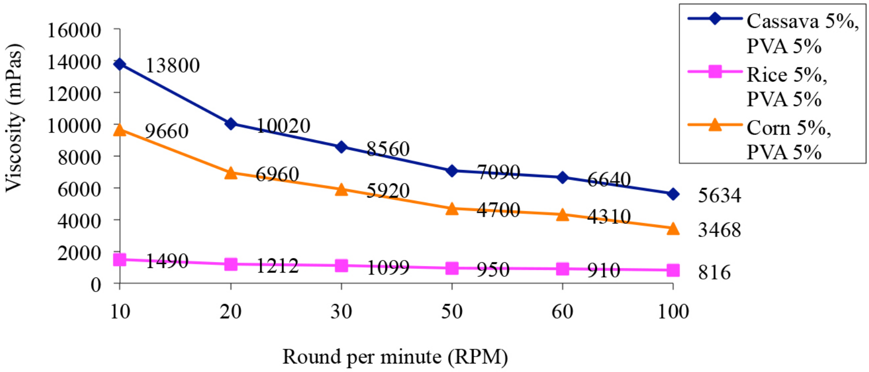


Figure 6. *viscosity dependence on shear rate of starch/PVA blend at 5%wt starch to 5%wt PVA*

The viscosity dependency on shear rate was different among starch types as well as content ratio of the starch/PVA blend. However, minor contents e.g., protein, lipid, ash, mineral, etc., obtained in rice flours, cassava, and corn starches derived from variety resources and productions possibly affected to their own viscosity and rheology. Besides, physicochemical properties of starch obtained from different botanical sources vary due to difference in their amylose content, grain size and chain length distribution of amylopectin molecules (Tako et al, 2014). It was same as flow behavior of printing ink rheology, that viscosity dependency on shearing rate of the starch/PVA blended solution was shear thinning of non-Newtonian flowing as shown in Figure 4, 5, and 6 (Leach and Pierce, 1993). The results of viscosity of the blends were considered on gelatinization and retrogradation properties, blending with PVA performed different characters from nature of pure starches as well. At 5%wt of starch (Figure 6) showed normal character as it was reported that the cassava, corn, and rice starch had high, medium, and low viscosity, respectively. When the blend solutions consisted of a high starch amount of 7%wt, it tended to reflect starch nature. Nevertheless, the corn starch blend showed very high viscosity on the curve of Figure 4, it was possibly caused by high retrogradation of the corn starch, but cassava starch has low retrogradation (BeMiller et al, 2009). Furthermore, viscosity values were not proportional to starch amount of the blends as well as starch-to-PVA ratio, which it can be studied further.

Properties of the inks composed of starch/PVA blend

It was found that a ratio of starch to PVA blending of 5%wt to 5%wt was an appropriate content to make a flexographic ink, those were it had non-Newtonian flowing as shear thinning behavior, it had good compatible of blending, and the blend was not too thick for making a flexographic ink. The blending ratio at a lower content of 3%wt of starch blended in a higher content of 7% wt of PVA was not chosen to make the eco-friendly ink because PVA was a synthetic polymer even though it was bio-gradable. Therefore, the content ratio of 5% to 5%wt of starch and PVA blend was selected to make the ink following recipe of Table 2.

a) Fineness of grind

Figure 7 showed a fineness of grind of the starch/PVA blends against grinding time duration. The cassava starch/PVA blend gave the best capability of pigment dispersion, since the minimum size was comparatively the finest size about 2.5 μm with the shortest time duration of grinding. However, all starch blends had the minimum size of fineness of grind lesser than 5 μm (cassava 2.5, corn 3, and rice 4 μm).

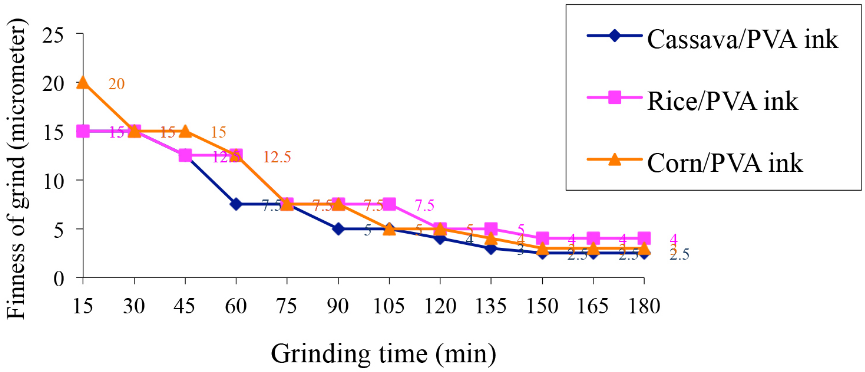


Figure 7. *Fineness of grind of the starch/PVA flexographic inks*

b) Viscosity behavior

Considering on Figure 8, the inks made from the starch/PVA blends behaved shear thinning of non-Newtonian flow, which was one of nature of a printing ink (Leach and Pierce, 1993). Viscosity values were lower than those of the viscosity values of the 5%wt starch/PVA blend as reported in Figure 6. And, non-Newtonian flow behaviors of all inks (Figure 8) were weaker than those of the starch/PVA blends (Figure 6). The reason was possible that there was interrupting in chain entanglement of starch and PVA blend by the other ink compositions e.g., pigment and nonionic surfactant as well as shearing force in grinding process.

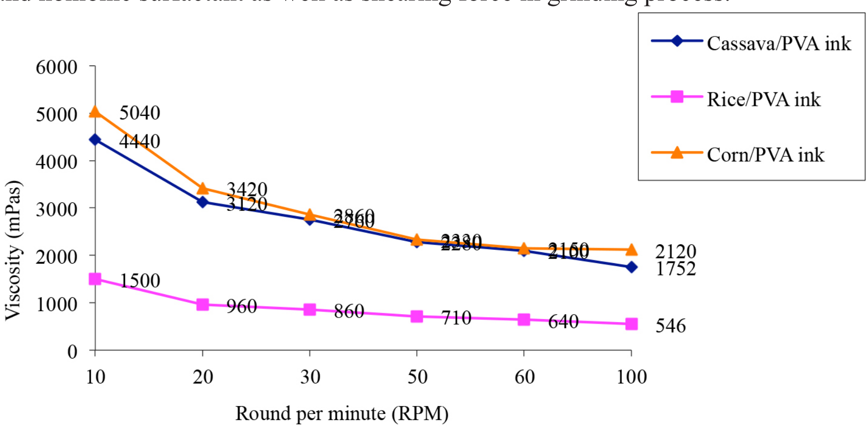


Figure 8. *Viscosity of the starch/PVA flexographic inks*

c) Print density by printability tester

To compare among starch types, a value of print density by K proofer as shown in Table 6 was that the blend of rice starch gave comparatively the highest density. Whereas, those of the blends of cassava and corn starches were almost the same value. This result was possibly related to the resultes of both ink viscosity behavior, microstructure of the starch/PVA blend, and clarity of the blend. Generally, pastes of potato, tapioca, and waxy maize starches are transparent. Corn and wheat starches, in contrast, give opaque pastes (Chen, 1990). As clarity of the rice starch blend film was not transparent, but opaque, so it gave higher reflected density to the printed density (1.13) than the other starch blends.

Starch based ink	Print density
Cassava	1.02
Rice	1.13
Corn	1.08

Table 6. *Print density on plain white paper by K proofer*

d) End-use property

Results showed that an ink made from a starch/PVA blend performed property closed to the typical flexographic ink (Table 5). Considering on a value of %weight-loss, the starch based ink had higher water resistancy than that of the typical flexographic ink due to crytallility of the polysaccharide molecule.

Starch based inks	Scale of rub resistance [a]	Water resistance (% weight-loss of the ink)	Lightfastness (ΔE exposed-unexposed by sunlight)
Typical flexographic ink (synthetic binder)	3-clear stain	40	8
Cassava/PVA ink	2-low stain	5	7.9
Rice/PVA ink	3-clear stain	6	6.7
Corn/PVA ink	3-clear stain	11	9.9

[a] According to Ugra scale guidance

Table 5. *Resistance properties of the starch/PVA inks*

Print quality of the cassava starch/PVA ink printing on the corrugated board with a flexographic press

The cassava/PVA blend was applied into an ink formulation of water based flexographic ink for corrugated paperboard printing. The cassava starch/PVA ink with higher viscosity performed a higher value of print density on the printed sample than that of the typical flexographic ink (Table 7). Dot gain/TVI of the printed sample as shown in Figure 9 showed that the cassava starch/PVA ink gave more than double (x2) of TVI value on the printed sample of the typical flexographic ink.

Starch based ink	Print density
Cassava/PVA ink	1.58[a]
Typical flexographic ink	1.32[b]

Ink flowing time using Zahn flow cup #4 was 17 sec (a), and 14 sec (b)

Table 7. *Print density on the corrugated paperboard*

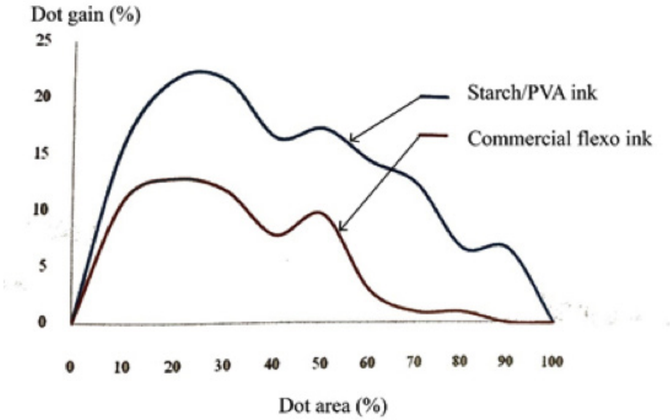
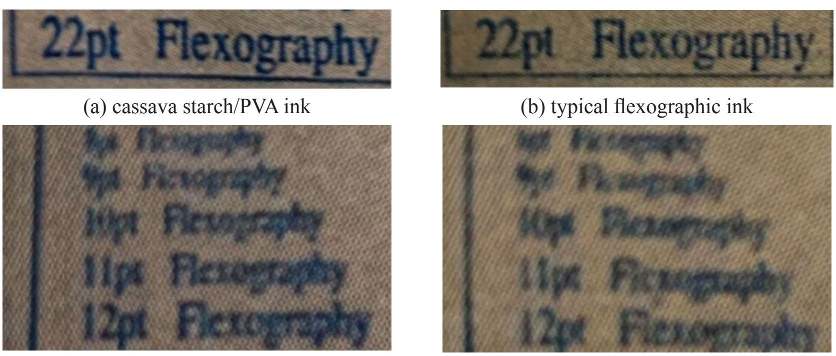


Figure 9. *Dot gain of the printed sample*

Figure 10 showed printed samples of the cassava starch /PVA ink compared to that of the typical flexographic ink. At the same printing condition as mentioned above, the printed texts of the cassava starch /PVA ink were readable better than that of the typical flexographic ink. It was easier to read on a small size character in a range of 8-10 pt. Evenness on the printed texts of the cassava starch/PVA ink were observable, but looked bold from high ink spreading. Print quality of TVI and printed texted related to surface interaction between ink and substrate. The contact angle of the cassava starch/PVA ink was 58°, whereas a contact angle of the typical ink was a higher value of 86°. Since lesser degree of contact angle demonstrated spreading and adhering on the surface of the ink, therefore the cassava starch/PVA ink could wet on, and spread along paper surface more than the typical ink. Contact angle of surface wetting had a relation with surface tension of the inks. Surface tension of the ink was a key property of wetting, spreading, and adhering on the printing substrate.



(a) cassava starch/PVA ink (b) typical flexographic ink

(c) cassava starch/PVA ink (d) typical flexographic ink

Figure 10. *Printed characters of a cassava starch /PVA ink (a), (c).*
a typical flexographic ink (b), (d)

Conclusion

This study showed some differences between cassava, rice and corn starches in the starch/PVA blend. PVA blending in the starch improved rheology as well as film formation of the printing ink. Proportion of starch and PVA was important to rheology behavior. The starch/PVA had good fastness, and high wetting on paper substrate. All cassava, rice, and corn starches can be blended with PVA for making a water based flexographic ink with a proper proportion in an ink formulation to carry out ink performance and print quality.

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