สภาพพิมพ์ได้ในการพิมพ์เฟล็กโซกราฟีของพลาสติกสลายตัวได้

นางสาวสมัชชา นวกิจวงศ์

วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญา วิทยาศาสตร์มหาบัณฑิต สาขาวิชาเทคโนโลยีทางภาพ ภาควิชาวิทยาศาสตร์ทางภาพถ่ายและเทคโนโลยีทางการพิมพ์ คณะวิทยาศาสตร์ จุฬาลงกรณ์มหาวิทยาลัย ปีการศึกษา 2555

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PRINTABILITY IN FLEXOGRAPHIC PRINTING OF COMPOSTABLE PLASTICS

Miss Samatcha Nawakitwong

A Thesis Submitted in Partial Fulfillment of the Requirements for the Degree of Master of science Program in Imaging Technology Department of Imaging and Printing Technology Faculty of Science Chulalongkorn University Academic Year 2012 Copyright of Chulalongkorn University

Thesis Title	PRINTABILITY IN FLEXOGRAPHIC PRINTING OF
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Ву	Miss Samatcha Nawakitwong
Field of Study	Imaging Technology
Thesis Advisor	Associate Professor Aran Hansuebsai, Ph.D.
Thesis Co-advisor	Hiromichi Noguchi, Ph.D

Accepted by the Faculty of Science, Chulalongkorn University in Partial Fulfillment of the Requirements for the Master's Degree

	Dean of the Faculty of Science
	(Professor Supot Hannongbua, Dr.rer.nat.)
THESIS COMMITTEE	
	Chairman
	(Assistant Professor Chawan Koopipat, Ph.D.)
	Thesis Advisor
	(Associate Professor Aran Hansuebsai, Ph.D.)
	Thesis Co-advisor
	(Hiromichi Noguchi, Ph.D.)
	Examiner
	(Associate Professor Pontawee Pungrassamee)
	External Examiner
	(Associate Professor Khemchai Hemachandra, Ph.D.)
	External Examiner
	(Associate Professor Phietoon Trevijitkasem, Dr.Ing)

สมัชชา นวกิจวงศ์: สภาพพิมพ์ได้ในการพิมพ์เฟล็กโซกราฟีของพลาสติกสลายตัวได้ (PRINTABILITY IN FLEXOGRAPHIC PRINTING OF COMPOSTABLE PLASTICS) อ.ที่ปรึกษาวิทยานิพนธ์หลัก : รศ.ดร.อรัญ หาญสืบสาย, อ.ที่ปรึกษาวิทยานิพนธ์ร่วม : HIROMICHI NOGUCHI, Ph.D., 111 หน้า.

งานวิจัยนี้มุ่งศึกษาสภาพพิมพ์ได้ ของพลาสติกสลายตัวได้ทางชีวภาพประเภท PLA ผสม PBAT และ PBATผสมแป้งข้าวโพด ด้วยระบบการพิมพ์เฟล็กโซกราฟีหมึกฐานน้ำ โดยวิเคราะห์ คุณภาพงานพิมพ์ รวมถึงสมบัติด้านกายภาพของพลาสติกและการพิมพ์ ที่ได้จากตัวแปรต่างๆ ดังนี้ ค่าความดำ, ค่าความเปรียบต่างทางการพิมพ์, โทนน้ำหนักสีของภาพที่เพิ่มขึ้น (TVI), ความ ทนทานต่อการขัดถู, การยึดติดของหมึกพิมพ์, ความขรุขระของผิวหน้าพลาสติกและ การเปียกผิว ของพลาสติก (วัดจากมุมสัมผัสของน้ำกลั่นและหมึกพิมพ์บนพลาสติก) ความละเอียดของลูกกลิ้ง แอนิล็อก 3 ระดับ 400,600,700 Ipi การปรับสภาพผิวด้วยวิธีคอโรนาดิสชาร์จทั้งหมด 4 ระดับ (0,500,1000,1500 วัตต์) ทั้งนี้ได้ทำการปรับสูตรหมึกพิมพ์ด้วยการเติมสารเติมแต่งให้สามารถ พิมพ์บนพลาสติกตัวอย่างได้ ผลการทดลองพบว่า ฟิล์มตัวอย่างทั้งสองชนิดสามารถพิมพ์งานที่มี รายละเอียดสูงได้ดีเมื่อมีการปรับสภาพผิว ค่าความดำ, การยึดติด, ความทนทานต่อการขัดถู และ การกระจายตัวของหมึกพิมพ์ที่สูงกว่าตัวอย่างที่ไม่ปรับสภาพผิว เนื่องจากผิวหน้าของพลาสติกทั้ง สองชนิดมีสมบัติความไม่ชอบน้ำและมีค่าพลังงานผิวที่ต่ำ แต่เมื่อปรับสภาพผิวมีส่วนช่วยให้ค่า พลังงานผิวและสภาพผิวหน้ามีความขรุขระมากขึ้น ทำให้สามารถรองรับหมึกและมีสมบัติการ พิมพ์ดีขึ้น ซึ่งสภาวะการพิมพ์ที่ให้คุณภาพงานดีที่สุด คือ ความละเอียดของลูกกลิ้งแอนิล็อกที่ 700 Ipi และระดับการปรับสภาพผิวที่เหมาะสมในการพิมพ์ คือ 500 วัตต์ โดยควบคุมความเร็วให้คงที่ 30 m/min เนื่องจากที่สภาวะการพิมพ์นี้ให้ค่าความดำของหมึกพิมพ์อยู่ในระดับที่เหมาะสม คือ ประมาณ 1.38 - 1.40 และมีค่า TVI น้อยที่สุด PBAT/Starch และ PLA/PBAT มีค่า print contrast อยู่ในช่วง 21 และ 18 ตามลำดับ ซึ่งอยู่ในช่วงที่ สามารถ แยกภาพน้ำหนักสี สว่าง (high light) และภาพส่วนน้ำหนักสีกลาง (mid-tone) ได้ดี แต่สำหรับสมบัติด้านการยึดติด ของหมึกบน พลาสติกนั้นอยู่ในระดับต่ำมากในทุกสภาวะการทดลอง

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KEYWORDS : COMPOSTABLE, FLEXOGRAPGY PRINTING, ANILOX, CORONA TREATMENT

SAMATCHA NAWAKITWONG: PRINTABILITY IN FLEXOGRAPHIC PRINTING OF COMPOSTABLE PLASTICS. ADVISOR : ASSOC. PROF. ARAN HANSUEBSAI, Ph.D., CO-ADVISER : HIROMICHI NOGUCHI, Ph.D., 111 pp.

This study was focused on printability compostable of plastic compounds which are PLA/PBAT and PBAT/Starch. Those two kinds of compostable plastic compounds were printed with water-based flexography printing ink. The print quality was observed in various variable parameter; density, print contrast, tone value increase (TVI), rub resistance, ink adhesion, surface roughness, and wettability (contact angle). The printing conditions were varied with three anilox line resolutions 400, 600 and 700 lpi and four levels of coronadischarge treatment 0, 500, 1,000 and 1,500 watt. Also, the water-based flexography printing ink was developed for this study with some additive in order to make the ink compatible for compostable plastic compounds. The results showed that two kinds of compostable plastic compounds could be printed with good high details but they were not suitable for printing without surface treatment because of its low adhesion, rub resistance and print uniformity. It was because the surfaces of those plastics compound films had hydrophobicity and low surface energy. However, the surface treatment increased the surface energy and roughness which made those plastics could be printed with better print result. The best printing condition was 700 lpi anilox line resolution and 500 watt corona treatment at 30m/min constant speed. Thorough this condition suitable ink density was gotten achieved in range of 1.38-1.40, and TVI was the lowest. The print contrast for PBAT/Starch and PLA/PBAT was about 21 and 18, respectively. It also was found that was no difference of tone rendering in high-light and mid-tone areas. While the ink adhesion property was in the lowest level in all condition.

Department : Imaging and printing technolog	<u>ay</u> Student's Signature
Field of Study: <u>Imaging Technology</u>	Advisor's Signature
Academic Year :	Co-advisor's Signature

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CHAPTER 1

INTRODUCTION

Recently, the packaging waste management becomes more serious problem in the many metropolitan around the world because of the increase of consumer. According to the state of packaging in the UK in 2008, the waste disposal was studied. The 65% of approximated 10.7 million tones of packaging waste was recovered and also approximated 4.7m tones of the household waste in 2008 relate to packaging. The packaging recycling has been concerned and increased over last decade. In order to decrease the packaging waste, the compostable packaging will be the good choice for waste management because it is not necessary to recovered or recycled. [1]

The increasing of domestic garbage and packaging waste results in environmental problem. So, the environmental friendly product has been promoted in order to replace those materials. 15 years ago, many countries in western try to produce degradable plastic which composes of petroleum product and corn starch. However, it did not give the satisfactory result. Because mixed corn starch in plastic will be intervened some part of whole polymer structure, when the structure of starch break down due to the aging, the polymer structure still remain but it becomes fragment. Instead of reduce environmental problem, this product becomes more serious problem in waste management. The tiny fragment of plastic does not be degraded. After this development, the advent of additive is used to be the catalyst for breaking down the plastic molecule unlike starch. However, this substance has limitation because it needs sunlight to initiate the chemical reaction. Thus, this process is called photodegradation. This is still not the practical method for degradation. If those plastic is disposed by land fill method, it will be not be degraded. So, it can be used in developing country where they left the garbage on the ground. However, that garbage is degraded into tiny particle and brings about the environmental problem; it causes the bad scenery and uncontrolled dust when the wind blows. After development by using heavy metal

component additive, this method can be used to catalyze for breaking down the polymer structure. It is called Oxodegradation. This method needs high temperature and oxygen enrichment condition to degrade plastic into small particle, then the microorganism can eliminate those particles. However, it is not compromised method because of remained heavy metal in the environment. [2]

After continuing development, the primary component from renewable resource such as plant, is the acceptable material to replace plastic material. Because its component can be compostable by biological process and become natural fertilizer too. Moreover, it have physical properties equivalent to LDPE and HDPE. Those LDPE and HDPE plastic are very popular to produce disposable bag for household usage. However, it causes the environmental pollution because those plastic cannot be recycled and they are disposed by incineration. Also it causes green house gas (GHG) which is the primary problem for climate change and affect to human health. So, the compostable plastics is recently a renewable material and the most environmental friendly product in present.

From the previous information, it can be categorized the bioplastics as this diagram.



Figure 1-1 The category of Bioplastic depends on produced material.

According to figure 1-1, it can be described that bioplastic has the two major categories;

- Compostable plastic which can be composted under appropriate condition. It can become the fertilizer in soil for plant growth. According to the biological process from micro-organism for degradation and composting in long time period, this compostable plastic becomes CO₂, water, inorganic compounds and biomass which has neither toxicity nor visible contaminants. [1]
- 2. Non-Compostable plastic is the product from plant which is used to replace petroleum products. It can be eliminated easier than ordinary plastic but its physical properties still equal to those petroleum plastics. Non-Compostable plastic cannot be composted by biological process. The commercial names are listed as follow.
 - Biodegradable plastics which is produced from petroleum product and it can be degraded in to small particle for disposal.
 - Photodegradation plastics which is produced from petroleum product and it can be degraded by sunlight.
 - Oxodegradation plastics which is produced from petroleum product and it has the heavy metal additive to catalyze the breakdown process in polymer structure instead of sunlight.
 - Green plastics plastic which is duplicated conventional molecular starchier and is produced from bio base material. It cannot be compostable but it can be categorized in bioplastics group. Because it made from plant material instead of petroleum product.

Because of different type of plastic, the symbol declaration shall be specified between compostable and non-compostable. This system is like the resin code which is used to categorize the type of plastic resin such as plastic bottle.



Figure 1-2. The samples of international standard certified symbol for compostable plastics declaration by biological process.

Because the above product declaration will relate to the printing process, type of printing ink shall be used according to ISO 17088. This international standard specified the limitation of compostable plastic and the printing ink must have the organic component which does not affect soil and plant growth such as water-based ink, soybased ink, sunflower oil-based ink, Palm oil-based ink, etc. However, those natural inks are not popular for industrial usage because of high cost and its properties. Comparison to the solvent-based ink, natural inks is time consuming in drying process but it is odorless and no toxic. It also it can be used as same as solvent-based ink.

Another benefit are that produced from vegetable oil, therefore it can be composted by biological process. When it was used to print on compostable plastic, all products can be composted. [3]

1.1 History and the important of problem

Recently, the compostable plastic packaging becomes the important product in market and gives more advantage unlike petroleum plastic which affects the environmental problem from manufacturing to waste management. The compostable plastic can be composted by enzyme and micro-organism in nature. After composted, it will produce water, carbon dioxide gas and biomass, these products are important for plant growth. In Thailand, there are resins which is used for this kind of product such as PLA (polylactic acid) and PBAT (polybutylene adipate-co-terephthalate). It has the physical properties similar to LDPE and HDPE [1] The compostable plastic compound

bag must has the "composible" symbol on it according to international standard such as EN 13432/14995 and ASTM D6400. Thus, the printing process deals with this compostable plastic. However, this kind of plastic has specific standard which shall be compostable by fermentation and inorganic component must less than 1% by weight of plastic. The overall inorganic component does not exceed 5% according to ISO 17088:2008. [3] This specification concerns the printing ink which is used to print on compostable plastic. In order to achieve this specification, the natural printing ink such as water-based ink or soy oil-based ink can be used. However, the drying process will be longer and it is hard to control the print quality compare to solvent-based ink. Because the flexographic printing use the minimum amount of toxic vapor, it is suitable for water-based and soy oil-based printing ink and also this printing process is easy to control. This thesis select this printing process for research and evaluate the actual print quality. However, print quality depends on many factors such as packing, type and properties of ink, print setting, anilox line resolution, etc. So, those factors must be controlled for appropriate parameter. This research aims to study the printability of compostable plastic compound especially Poly Lactic Acid (PLA) Poly(butylenes adipate-co-terephthalate) (PBAT) blend and PBAT compound starch with flexography printing in order to evaluate print quality and parameter effect.

1.2 Objectives

1.) To study the printability of compostable plastics films by flexographic printing system and

2.) To study physical property effect of corona treatment upon compostable plastic films.

1.3 Scope of study

This study is to analyze printability in flexographic printing of compostable plastics compound both Poly Lactic Acid (PLA)/ Poly(butylenes adipate-co - terephthalate) (PBAT) blend and PBAT compound starch which are produced carrier

bag. In this study, solely water-based inks are considered. Printing under the conditions consist of different three levels of anilox line resolution; 400, 600, and 700 line per inch (lpi), and different 4 levels of corona treatment; 0, 500, 1000, and 1500 watt are required. This investigation method will show the different results such as thickness, tensile strength, gloss, energy surface, the black, consistency of ink, and adhesion of ink. Getting results will be the reference to choose the suitable condition for printing on both of compostable plastics.

1.4 Basic agreement

1. Using two types of compostable plastics consist of PLA/PBAT blends from FKuR Kunststoff GmbH and PBAT/Corn Starch from Thantawan Industry PLC.

Using flexographic printing machine by Flexographic printer Nilpeter FB-LINE 3300 from KO Print and Sticker Tunjai Co., Ltd.

3. Using water-based flexo inks from Panorama Soy Ink Co., Ltd.

4. Two solution, Reverse Osmosis water (OR water) and black acrylic water base ink are used for contact angle test.

5. Using printed resolution at 133 lpi gradient tone area for print quality measurement in this experiment.

1.5 Expected outcome

Information about printability of compostable plastics by using flexographic printing.

1.6 Research methodology

- 1. Researching information and related documents
- 2. Sourcing compostable plastics
- 3. Testing properties of compostable plastics
- 4. Printing
- 5. Analysis of printing quality

- 6. Analysis and conclusions
- 7. Thesis

CHAPTER 2

THEORY AND LITERATURE REVIEW

THEORY

2.1 Plastics

Plastic is polymer material which composes of high molecular weight organic compound. [4]

The type of plastics can be divided into two categories.

Thermosetting: this kind of plastic can be melted one time only and it will crosslink among polymer chains. After that plastic will cool down until it hardens. Then it cannot be flexible again by heat and it will break apart when the temperature is reach at point. To form this kind of plastic as desire, it is necessary to use high temperature.

Thermoplastic: this kind of plastic has line or short chain structure and can be dissolved in some solvent. It can be melt and become viscous liquid in high temperature, after the temperature decrease it can be form into solid structure. This property makes this kind of plastic recyclable such as polyolefin (polypropylene and polyethylene) for boil-in-bag or ordinary bag.

In plastic manufacturing process, the additive will be used for increase specific properties such as flexibility, toughness, colorant, etc. The plastic resin is melted and flowed into template by extrusion, injection or blowing process depends on desire product.





This research use compostable plastics compound as substrate for printing. So, it will be described only in that kind of plastic.

2.2 Type of Bioplastics

Bioplastics is the environmental friendly product which reduce petroleum raw material and help in waste management. Bioplastics can be divided in two major categories. (Show in table 2-1)

Table 2-1 The types of bioplastics depends on compostable property and raw material.

[2]

BIOPLASTIC (Generic)										
NONCOMPOSTABLE				COMPOSTABLE						
PETRO-BASE	-	BIO-BASE		PETRO-BASE	BIO-BASE					
Compound		Green Plastic		PBS, PBSA,	PLA, PHA, PHB etc.					
PHOTO, OXO, BIO	D E G R A D A B L E	LDPE, HDPE, etc.	N O N - D E G R A D A B L E	PBAT, PCL						

From table 2-1, it can be summarized that compostable plastics was the raw material from bio-based or petro-based. It can be used for producing many kinds of product as same ordinary plastics. However, bioplastics is compostable through fermentation process and produce carbon-dioxide gas, water and biomass in the soil; it can be used as fertilizer for plant growth. Those process occur temperature above 50°C. Non-compostable plastics in Bio-base is the product from plant for petroleum replaceable product, the product is made from non-compostable plastics has the same toughness property. However, It is non degradable and compostable. Non-compostable plastics in petro-base is typical plastics but mixed additive for disintegrate property to from tiny particle which is called disintegration. It can be called compound plastic. There are attempt to understand that particle is small enough eliminate by micro-organism. This process is called biodegradation. However, this process is not compostable complete

2.2.1 Primary source of compostable plastics film

- Bio-based material

2.2.1.1 Polymer from plant or animal such as starch, cellulose, lignin, chitin, gelatin and silk.

2.2.1.2 Polymer from micro-organism or plant such as PHAs (PHA, PHB, PHV).

2.2.1.3 Polyester from biosynthesis such as PLA.

2.2.2 Petroleum-Based material

2.2.2.1 Aliphatic polyester: PGA, PBS, PBSA, PCL

2.2.2.2 Aromatic polyester: PBS, PBSA, PBST, PBAT

2.3 Poly Lactic Acid (PLA)

PLA made a bio-base, which is a hydrophobic and semi crystalline polyester, is a renewable material that can be utilized by microbes within 30-40 days. Its good physical properties and commercial availability make it very attractive, it have many applications. Example for applications in medicine and packaging. [5]

The synthesis of compostable polymer begins with the converting of agricultural products to sugar which is the primary source of monomer and polymer. Then the polymer will be developed the properties and formed into specific products for various usage. When those products are disposed into the appropriate condition, micro-

organism will degrade it to carbon dioxide gas, water and biomass which are the important products for photosynthesis in plant. [6]



Figure 2-2 The synthesis process of PLA compostable plastics [7]

Compostable polymer bio-base is the polymer which is mixed with starch. Starch is the polysaccharide which composed of monosaccharide, $(CH_2O)_n$. Generally, starch can be found in seed, root and stem of plant. Starch is small granule and has various form depends on plant species. Typically, starch is mixed with polyester from 10 to 90% depends on desired properties and usage. If there is more than 60% of starch content in polymer combination, it results in compostable property in biological process. In contract, if starch content is less than 60%, it can be degraded plastic into small particle. [8]



Figure 2-3 The structure of PLA [9]

2.4. Poly(butylenes adipate-*co*-terephthalate) (PBAT)

PBAT is a aliphatic polyester and aromatic polyester which is fexible and fully compostable within a few with the aid of naturally occringenzymes. There is no starch content. The reason why this polyester is used as primary material to produce bioplastics because those bioplastics it has low bonding strength and it can be degraded into small particle by hydrolysis process. PBAT could be considered as a good candidate for the toughening of rigid polymers such as PLA.

Figure 2-4 The structure of Aliphatic - Aromatic copolyester [4]

Recently, compostable plastics are developed for multipurpose in many application such as absorbable suture material, bag, sack, food container, wrap film and other packaging, etc.

2.5 International Standard of biodegradable and compostable plastics

All new compostable plastics must be tested its properties according to ISO17088:2008(E) standard before declare "Biodegradable and Compostable plastics" seal. [3,6] Those compostable plastics can be recovered for use. However, it must be tested the intensive aerobic composting process by simulating through the process.

- a) the ultimate level of aerobic biodegradation of the test material;
- b) the degree of disintegration obtained;
- c) and negative effects on the finished compost;
- d) the maximum concentration of regulated metals in the compost.

After the biodegradation process become stable or reach plateau phase, the test can be terminated in 45 days or continue up to six months. The desired properties of plastic:

- The degradation of plastic under conditioned fermentation process in 84 days has remained material on 2.0 mm strainer not exceed 10% of dry solid mass according to ISO 14855-1, ISO 14855-2 or ASTM D 5338 test method, under thermal fermentation process without carbon dioxide detector.

- The final stage of oxygen degradation of organic carbon in plastic, additive and filler must be converted into carbon dioxide for photosynthesis process more than 90% of organic carbon by plastic theory within 180 days. The micro-crystalline shall be used as positive reference material according to ISO 14855-1 ISO 14855-2 test method. The additive and filler must be tested in oxygen biodegradation process by following this condition. • In case of additive or filler is exceed above 1% of dry solid mass, it must be tested.

• In case of additive or filler is not exceed above 1% of dry solid mass, it is not necessary for testing. However, combined additive and filler must not exceed above 1% of dry solid mass. If it is not, it must be tested.

- No adverse effects on ability of compost to support plant growth and compliance with regional and/or national regulations

• In order to ensure that the composting of plastic products or materials does not have any harmful effect to environmental when the finished compost

- Sublimate solid in plastic shall not exceed 50% of dry solid mass according to ISO 20200 test method.
- Plant growth must be compared between plants which are grown

2.6 Flexography printing system

Flexography is a method of direct rotary printing which has resilient relief image plates of photopolymer material. The plates are affixed to plate cylinders and ink cover on relief image area. Flexography ink is liquid ink, it can be water-based or solventbased. The ink metering system is controlled by anilox roll. The anilox roll will transfer ink from ink chamber or ink try into plate. [10] Typical products which are printed by flexography are the flexible packaging or labels such as film plastics, sticker, paper, foil, multi-wall bags, corrugate board, labels, wallpaper and wrappers.



Figure 2-5 Relief image of plate on flexography printing system

The general flexography printing unit is composed of four basic parts:

- Fountain roll Ink-metering (anilox) roll
- Plate cylinder Impression cylinder



Figure 2-6 Principle of flexography printing [10]

At first, the process in print unit start from the ink-fountain pan supplies ink to a rubber ink-fountain roll and then transfer ink to the anilox roll. At this point, the doctor blade will remove the excess amount of ink. Ink still remains in anilox chamber which will be transfer a precise amount of ink onto the printing plate. This plate is mounted onto the printing cylinder and then ink will be transferred from the printing plate to substrate. The impression cylinder will impress at the back of substrate which is called nip at this point. It will make printed image shape and even. Because substrate has different

thickness, the nip of printing cylinder and impression cylinder must be adjusted depends on thickness of substrate.

2.7 Flexography printing machine

The flexography printing can print on various substrates, the machine must be designed for various job too. The types of flexography printing machine can be categorized into 4 main parts of printing unit; central impression, stack, in-line and sheefed. Each type has similar work process but different in its efficiency such as speed, number of printing unit substrate width, etc.

In this research, the in-line printing machine was selected. The in-line press has printing unit align in horizontal, each unit is independent and it can print both web and sheet. Because this press can print on thick substrate and it can operate at low speed, it is suitable for controlling the print condition.



Figure 2-7 Diagram of in-line press [10]

2.7.1 The anilox roll

The primary role of the anilox roll is to transfer a finite amount of ink through small engraved cells via the printing plate to the substrate. The engraved cells have a shape of a truncated hexagonal cone with a specific volume (ml/m²),each cell must be uniform and identical in both size and depth to ensure that a controlled uniform ink film thickness in transferred to print plate and are distributed in a regular screen pattern over the anilox roll with defined numbers of lines/cm. An example is an anilox roll with 120 l/cm and a nominal ink capacity of 8 ml/m². The cell count/cm should be 4-5 times greater on the anilox roll than the cell count on the plate to print a dot of as small as 4 %. Generally the anilox has a core of steel, which can be plated with a thin layer of chrome or it can also be coated with ceramic. [11]



Figure 2-8 The detail of cell in anilox [10]

The suitable anilox line should be used depends on print job and size of dot

screen on plate. If print job need high details, dot screen on plate must be small and high anilox line must be selected, otherwise it will cause print problem such as dot dipping. Dot dipping is the dot screen where it is submerged in the anilox cell and causes the loss in print detail, because ink covers all those area.



Figure 2-9 The position of dot dipping in flexography printing

2.7.2 Printing Plate

The most commonly used plate material is the solid-sheet photopolymer material and type of plate material has been used throughout this project. Photopolymer plates is the photosensitive plate which is produced through the polymerization process under UV light.[11] This plate is flexible and though to withstand the impression between cylinders in print unit, it can be compressed and recovered under impression and has wettability for water-based ink or solvent-based ink.



Figure 2-10 Structure of sheet photopolymer plates [10]

The plate build-up is chatacterised by multiple layers in a sandwich structure. It composed of 4 layers; polyester film cover sheet, Photopolymer layer, adhesive film and support base layer. Each layer has different function. Polyester film cover sheet is the protection layer for non-image area when the plate is exposure under UV light. Because the photopolymer is irradiated and hardened with a specific wavelength by UV-radiation, the non-exposure area will be washed with solvent. The adhesive layer was exposure at the first time to make the base of plate. Adhesive layers bone the photopolymer layer with a support base for dimensional stability. Fig 2-11, the photopolymer plate is exposed at the back under UV-A light to increase the strength of base, and then the laser beam will create the image at the top of plate. The reaction between photopolymer plate and laser beam will harden the image area. When the plate is washed, the non-image area is removed and the image area still remains. After dry the plate, it will be exposed under UV light again which is call post exposure to stop polymerization reaction in photopolymer plate. In the final stage, it will be exposure under UV-C light for surface treatment to eliminate sticky surface.


Figure 2-11 The procedure of sheet photopolymer plates making [11]



Figure 2-12 Completed sheet photopolymer plates [10]

Plate properties such as hardness can be controlled by the key ingredients and plasticizers. A common way to for a plate manufacturer to classify

plates, apart from used the process used and the field of application is by hardness (° shore A) and by thickness [11]

This kind of plate has various hardness from 31 to 88 ° shore A. The high number indicates high hardness. (It can be measured by durometer) The proper hardness of plate depends on substrate, also the print quality depends on hardness and thickness of plate too. Using suitable plate can reduce print problem such as dot gain and help in ink transfer for suitable substrate for print sharpness.



Figure 2-13 Selection of plate hardness and application

2.8 Flexographic water base ink

Printing ink is the color liquid which composed of various materials depends on viscosity, flexibility, flow and other properties. It can be transfer on many kinds of substrate. Water base ink, which now contains only small amounts of volatile organic compounds (VOC), has low viscosity like gravure printing ink and can flow into anilox cell and transfer on the substrate. Generally, the viscosity is about 20-25 second by measuring with Zahn cup No.2 at 25°C for 2-4 micron thickness of substrate printing. [12]

2.8.1 The basic elements in flexographic inks

- Colorant

To make the color for printing ink, it can be divided into 2 major groups; pigment and dye. Pigment is the non-dissolve particle which disperses in the vehicle. The vehicle will deliver pigment to adhere the surface of substrate. Dye is dissolvable particle in proper solution or vehicle. It can be adhered on the surface without binder. Most of flexographic water-based inks usually use organic pigment as colorant such as carbon blacks.

- Binder

It disperses colorant and delivers to adhere at the surface of substrate. The type of resin of binder affects strength, gross, adhesive drying and flexible property. For flexographic water-based inks usually use acrylic emulsion as binder.

- Solvent

Flexographic water-based inks usually use de-ionized water as solvent. It has no harm but affect on adhesive property on non-porous substrate.

- Additive

Additive is the substance which is used to increase desired properties of ink. For example, silicone is used to increase scratch resistant but it will reduce wet rub resistance too.[13] So, the proper amount of additive should be considered for all desired properties.

ingredient	Amount (%)
Pigment (Carbon blacks)	50
Acrylic solution polymer	10
Acrylic emulsion	30
Water	5
Organic amine	1
Polyethylene wax compound	3
Surfactant	0.5
Organic anti-foam	0.5

Table 2-2. The component of typical flexographic water-based Ink [10]

The formula of ink should be considered the printing process and substrate for good distribution, adhesion, transferring and wettability of ink.

2.8.2 Factors of flexographic water-based Ink

The factors of ink must be controlled for good printing quality otherwise it will cause print problem.

- pH control :

pH should be in range 8-9.5 for dispersion property. If it is too acidic, ink will become agglomerate and precipitate.

- volatility of water :

The rheological behavior can be controlled by printing speed and temperature. Because printing ink has pseudoplastic flow which is the rheological behavior that increasing in shear rate cause reducing in viscosity should be 20-40 second by measuring with Zanh cup No. 3 or 2.

- surface tension :

Surface tension of ink is the cohesion of molecule at the surface of substance. Surface tension indicates the interaction between ink and substrate due to wettability. If surface energy value of substrates is higher than surface tension of ink, ink can wet on the substrate. In contrast, if surface tension of ink is higher than surface energy value of substrates, it causes print problem. Ink cannot wet on the substrate. In practical, dyne pen usually use to measure the surface energy of print substrate which is non-absorbent such as plastic film, coated paper, can, etc. Another testing is contact angle which is the delicate technique and time consuming.

2.9 Wettability

Wetting is the replacement of air at surface substrate with liquid.





- (1) Cohesion of surface tension of liquid causes sphere shape of droplet.
- (2) Surface tension of substrate cause droplet spreading and wetting on the surface of substrate.

In homogenous substance, molecule or atom at the surface is different from interior. The average distance is controlled by cohesion force between molecules, in the same time, it has repulsive force too. So, the molecule of substance is in equilibrium between cohesion force and repulsive force, but the molecule at the surface of substance has cohesion force from internal molecule to reduce the number of molecule at the surface and it cause surface tension. The wettability is the important property for ink adhesion on substrate.

2.10 Contact angle

Contact angle is the angle of liquid droplet on the surface. It is measured from outside of droplet. It has tree-phase boundary; substrate-liquid, liquid-air and substrate-air, The contact angle of liquid and substrate at equilibrium, 0-180° (degree of wetting), see figure 2-15



Figure 2-15 Contact angle of droplet on substrate

Contact angle value indicates the wettability;

$$\theta = 0^{\circ}$$
 spreading

$0^{\circ} < \Theta < 90^{\circ}$	wetting
$90^\circ < \Theta < 180^\circ$	non-wetting
$\Theta = 180^{\circ}$	repellency

The measured angle is statistic number. It will be measured by dropping the liquid on surface of substrate under clear confined container, then measure the contact angle at equilibrium state. It contact angle value indicate both wettability of liquid and surface tension of substrate.

2.11 Surface energy

2.11.1 Surface Energy by Contact angle

The energy of a surface is most commonly quantified using a contact angle goniometer. To examine the surface energy of a material, drops of defined liquids (in Table 2-3.) on the material surface and the contact angles of these liquids on the surface are determined.

Table 2-3. Examples of surface tension of liquids with defined Lifshitz-van der Waals (LW) interaction and polar interaction components (γ^{P}_{l}) and (γ^{d}_{l}) [11]

	Surface tension [Dyne/cm]			
Liquids	γ	$\boldsymbol{\gamma}^{\scriptscriptstyle ext{LW}}$	γ^{P}	$\gamma^{{}^{\scriptscriptstyle d}}$,
Water	72.8	21.8	25.5	25.5
Ethylene glycol	48	29	1.92	47
Diiodomethane	50.8	50.8	0	0

The nature of a surface or the interfacial free energy for a solid and liquid

system is often explained in terms of apolar Lifshitz-van der Waals and polar forces.[11]



Figure 2-16 Contact angle of liquid on the solid surface; tree-phase boundary from Young's Equation

Figure 2-16 shows a droplet of a liquid on a plane surface. The contact angle (θ) is difined as the angle that is formed at the junction of solid-liquid interface (SL) and the liquid-vapour interface (LV). When the contact angle is > 90° the surface is said to be hydrophobic. The relationship of tree-phase boundary which is measured to contact angle can be writed as Young and Fowkes equation, see Eq.1 and Eq.2

$$\gamma_{LV} (1 + \cos \theta) = 2(\gamma_{l}^{d} \gamma_{s}^{d})^{1/2} + 2(\gamma_{l}^{p} \gamma_{s}^{p})^{1/2}$$
(Eq. 2-1)

When

 θ = contact angle

 $\gamma_{\scriptscriptstyle LV}$ = surface tension of liquid

 $\gamma^{\prime}{}_{\prime}$ = surface tension of non polar contribution of the liquid

 γ^{d}_{s} = surface energy of non polar contribution of the solid

 γ^{P}_{l} = surface tension of polar contribution of the liquid

 γ_{s}^{P} = surface energy of polar contribution of the solid

Total surface energy
$$(\gamma_{SL}) = \gamma^{\rho} + \gamma^{\sigma}$$
 (Eq. 2-2)

When γ^{d} = the dispersion component of total surface free energy

 $\gamma^{\scriptscriptstyle
ho}\,$ = the polar force component of total surface free energy

In this research, contact angle will be measured only RO water and water-based ink which are used in printing process in order to analyze the wettaility of compostable plastic sample.

2.11.2 Surface Energy by dyne solution test

This method will measure the surface energy according to ASTM D-2578 test method. It will draw the mixed solution which has different surface tension such as formamide and cellosolve in different ratio, see table 2. After drew the mixed solution on substrate and it can wet on that substrate, this indicate the similar surface energy of substrate and surface tension of mixed solution.

Table 2 -4. Ratio of concentration of solution between formamide and cellosolve for measuring the surface energy.

Volume of formamide (%)	Volume of cellosolve (%)	Surface tension (dyne/cm)
0	100.0	30
2.5	97.5	31
10.5	89.5	32
19.0	81.0	33
26.5	73.5	34
35.0	65.0	35
42.5	57.5	36

Volume of formamide (%)	Volume of cellosolve (%)	Surface tension (dyne/cm)
48.5	51.5	37
59.0	41.0	39
63.5	36.5	40
67.5	32.5	41
71.5	28.5	42
74.7	25.3	43
78.0	22.0	44
80.3	19.7	45
83.0	17.0	46
87.0	13.0	48
90.7	9.3	50
93.7	6.3	52
96.5	3.5	54
99.0	1.0	56

• Cellosolve is the commercial name of ethylene glycol monoethylether

• Surface tension was tested at 23±2°C temperature and 50±5% relative humidity.

In practical, dyne pen is used to test the surface energy of plastic film. The users can draw a line on the desired substrate and counts two second to evaluate the surface energy like dyne solution. The dyne pen has number indicate on the pen and it indicates the critical surface tension, it is very easy and simple to use with adequate confirmation result. However, dyne pen has shelf life only 6 months. After that the efficiency will decrease. [14]

Each type of plastic film has suitable specific surface energy for printing. Thus, the level of surface treatment should relate to the suitable surface energy.

Substrato	Surface Energy		
Substrate	(Dyne/cm)		
PE	38-44		
PP	38-44		
PVC	38-44		
PET	44-52		
PS	38-44		
PU	40-46		

Table 2-5. The suitable surface energy of plastic film for water-based ink

Compostable plastics which is used in this research composed of polyester and it has several physical properties like HDPE and LDPE. So, the dyne pen can be used to test the surface energy at 38-44 dyne/cm.

2.12 Corona discharge treatment

Corona discharge treatment is the surface treatment for plastic film before print. The surface of plastic film is non-polar which composes of carbon and hydrogen atom. So, the wettability of water-based ink is not good. When treat the surface with electricity, it will create oxygen atom and it will replace hydrogen atom on the surface of plastic film to make it more polar. It will increase the surface energy and wettability of plastic film. In this research, plastic film will be treated by corona-treatment unit on the In-line flexography press.



Figure 2-17 Diagram of corona discharge in the In-line flexography press Factors affect on surface treatment with corona discharge:

- Voltage Source
- Film speed through the Treater electrodes should be constant.
- Width of Treater electrodes to define the width of plastic film
- Number of electrode should be stable and have enough numbers per area.
- The gap between Treater electrodes and plastic film should be about 1.5 mm.

2.13 Print Quality Evaluation

Print Quality Evaluation can be achieved by visual assessment at quality control strip and densitometry measurement. The quality control strip is inserted at the top or bottom of printed matter, this tool can be used as represent of real image. By observing this quality control strip, the print problem can be analyzed quickly while the press is operating. However, using the quality control strip for visual assessment is not enough to be the standard reference for print quality control. The measuring equipment should be used with quality control strip for print quality control too. There are many types of quality control strip and also each printing process has different quality control strip depends on standard and organization.







(b)



Each step of dot screen at 3-10% is the representative of high light, mid-tone and shadow of image at the range of 40, 60% and 75%, respectively. (b) Control strip indicated values which are changed for the specific press as follows in each position of control patch; area produced for control or measurement purpose the example in patch

A ; ink tap patch of red, green, blue, patch

B and D;solid color patch hue and saturation

I; Characterization dot gain

2.14 Optical Density

Optical density is an important for every print. Optical density is roughly a measurement of how much ink that is transferred to the substrate and its appearance. Too low a optical density means that the print looks dull while too high a optical density means that half-tone dots tend to fill-in. The optical density is a measure of the contrast of a print and a logarithmic function is applied in accordance with the general experience that a psycho physiological perception is related to logarithm of the stimulus. Optical print density, D_s (solid tone) and D_T (half-tone) is defined as [11]

$$D_{s} = \log (R_{0} / R_{s})$$
 (Eq. 2-3)

$$D_{T} = \log (R_{0} / R_{T})$$
 (Eq. 2-4)

When : $\ensuremath{\mathsf{R}_{\text{T}}}$ is the reflectance factor of the half-tone,

 $\boldsymbol{R}_{\!\scriptscriptstyle 0}$ is the reflectivity of the unprinted substrate.

 R_{S} is the reflectance factor of the solid tone

Optical density is usually measured by a densitometer or spectrophotometer. The appropriate optical density for flexography printing is specified in table 2-6.

Table 2-6 Solid-Ink Density for film plastics [10]

	Optical density
Cyan	1.25
Magenta	1.20
Yellow	1.00
Black	1.40

2.15 Tone Value Increase (TVI)

Tone Value Increase is the tone value that increase more than the original file; unit %. TVI is caused by dot gain which the half-tone dot area increases more than the normal dot area from original file. Dot gain is not considered as print problem and it is hard to avoid because of the impression of printing. However, dot gain should not exceed too much and cause the loss of print detail. TVI can be measured from dot gain. Typically, there is two types of dot gain; physical dot gain and optical dot gain.



Figure 2-19 Type of dot gain.

2.15.1 Physical dot gain

Physical dot gain is the increase of dot area which is caused by impression force from printing. When compare the dot area on plate, this dot gain increase in all direction at the same level. The shape of dot gain is still the same as plate but it is bigger. If the shape is not the same, it may indicate other print problems, see figure 2-20. The level of dot gain depends on other factors such as impression force and screen line on plate.



Figure 2-20 The sample of print problem which looks like dot gain. (a) doubling problem (b) slur problem

2.15.2 Optical dot gain

Optical dot gain is the increase of dot area which caused by the light scattering inside of print substrate and it has less reflection. So, the dot area seems to increase more than the normal dot area.

Dot gain can be calculated from print density by measuring the dot area with densitometer on the control patch, then calculate with Murray-Davies equation (Eq.2-5) [11]

Dot gain (%) =
$$(1 - 10^{DT})/(1 - 10^{DS})$$
 (Eq.2-5)

When ; $\, \mathsf{D}_{_{\mathsf{T}}}\,$ is density value of the half-tone,

 D_{S} is density value of the solid tone

In case of spectrophotometer has both print density and %Dot gain function, the dot gain can be measured easily. Normally, it should be measured in high light, mid-tone and shadow at the control patches and then find the TVI, see figure 2-21. In range of mid-tone, dot gain occur at higher level than high light and shadow. Dot gain can be used to consider TVI in each color of print image.



Figure 2-21 TVI curve in each color of print image.

2.15.3 Print Contrast

Print contrast is the method for evaluating and optimizing the density of ink during printing. This method involved with the contrast of printed image by considering high light, mid-tone and shadow of image area. It can be calculated by measuring the ink density of a solid area and the ink density in a 75% or 80% on control patches [14]

When the ink film thickness reaches the high level, the density does not increase. The appropriate ink film thickness should be the lowest thickness but has the highest density. It will give good saturation and hue. Also, low ink film thickness results in less dot gain. The relationship between ink film thickness and dot gain can be considered from print contrast. Because print contrast indicate the appropriate ink density on specific substrate and appropriate condition for less dot gain.

The print contrast is calculated according to the formula.

Print contrast =
$$((D_{s} - D_{T})/D_{s}) \times 100\%$$
 (Eq.2-6)

When ; D_T is density value of the half-tone,

 D_s is density value of the solid tone

If the print contrast decrease, it indicate that condition indicate the loss of details in shadow or 75-95% screen area of image. It makes printed matter look darker than the original. In practical, it can be used the highest print contrast to select the condition for print and adjust the ink film thickness equal or slightly lower than those print density.

Literature reviews

Pradya Khiangprakhong. studied modification two type of biodegradable plastics, aliphatic-aromatic copolyester and aliphatic-aromatic co-polyester/polylactide blend by corona discharge. The surface treatment was performed at various power input and exposure time. Surface energy of the plastics was obtained by contact angle measurement of water and diiodomethane on the substrates. Adhesion of water-based flexographic ink and treated plastics was tested by tape test. After the corona treatment, the polar component of surface energy of aliphatic-aromatic co-polyester/polylactide blend was dramatically enhanced. The dispersive component of surface energy of aliphatic-aromatic co-polyester was higher compared to that of the blend. Increasing in exposure time also improved the surface polarity of the plastics. Tape test results suggested that that the water-based ink could adhere stronger on the treated aliphatic-aromatic copolyester/polylactide blend surface.[4]

Janejira Phunkoksung and Sroymanee Nimnuan studied the comparison of biodegradability and printing properties of biodegradable plastic which were printed by soy-bean lnk and solvent base ink. Biodegradable film was mixed between bio-based polymer and petro-based polymer in 50:50 ratios and then compare to HDPE. After that it was printed with red and green ink from both soy-bean Ink and solvent-based ink, and then buried it in soil for 93 days and then measured carbon dioxide in different time frame. Also observe the alteration of film surface, adhesion level, stretch, density, tone value, gross, adhesive property and rub resistance before and after bury. The result showed that biodegradable film without printing can be composted at the highest level. The biodegradable film which is printed by soy-bean Ink tend to composted better than the film which is printed by solvent- based ink. HDPE film has no different in degradation in both printing inks. The print quality of soy ink has higher gross than solvent-based ink on biodegradable film but the adhesion level, rub resistance, tone value are not different when compare to printed HDPE film.[6]

Johnson, J. studied about obtain further knowledge of some important mechanisms of flexographic printing and how they influence the print quality on board and liner but also on newsprint with water-borne ink using a full-scale flexographic Central Impression (CI) printing press and focus on the chemical interaction between the ink and substrate and the physical contact between the ink- covered printing plate and the substrate and study was emphasis was to investigate the relation between print quality and water-uptake of the paper surface with heat and water. Printing trials was carried out on substrates possessing a hydrophobic, and also a rather hydrophilic surface using a regular commercial water-borne ink. The favorable effect which water or surfactant solution on the hydrophobic substrate with regard to print mottle could depend had on its surface compressibility in combination with the hydrophobic nature of its surface that could affect the wetting properties. Conventional printing involves physical contact between plate and ink and between ink and substrate. A method for measuring the dynamic nip pressure using thin load cells is presented. Print quality was influenced by the plate material. A correction procedure taking into account the size of the sensor was developed in order to estimate the maximum dynamic pressure in the printing nip. An attempt was made to identify essential mechanical and chemical parameters, and also geometrical

properties of the plate that affected print quality. Laboratory printing trials were carried out and a multivariate analysis was applied for evaluation of print density data. The impact of the plate properties on print quality was evident. The essential properties of the plate that influence print quality were the small-scale roughness and long-scale roughness.[11]

Rentzhog, M. and Fogden, A. study was the performance of water-based acrylic flexographic inks laboratory printed on three different polymer-coated boards, LDPE OPP and PP analyses print quality and resistance properties obtained were related to varying ink formulation; emulsion polymer and presence silicone additive in vehicle each type of ink and varying level of corona pretreatment. Found that print mottle and adhesion were worst on PP, while wet (water) rub and scratch were worst on OPP and PE, respectively. However, these properties could be greatly influenced by the ink formulation more than corona level. In general addition of silicone improved scratch resistance, due to reduction in polar energy component of the print surface, but at the expense of worsened wet rub resistance. The emulsion polymer giving best resistance performance was generally found to give poorest optical properties, presumably due to more limited resolution on press.[13]

Gilberd, E.D and Frederick, L. studied print quality of both convertional and digital flexogeaphy and to analyze and statistically compare the two processed wanted to prove that digital imaged photopolymer flexographic plates produce lower dot gain, higher print contrast and longer range than conventional plate or not. By utilized the quasi experimental research design and an independent samples T-test. An alpha value 0.05. The result was no significant difference in dot gain between two plating system. However, significant improvement in print contrast values and tonal range.[16]

CHAPTER 3

MATERIALS AND METHODES

3.1 Materials and Equipments

This research used materials and equipments as following;

3.1.1 Materials

- Film Biodegradable and compostable plastic compound
 - PBAT compound starch; Mater $\mathrm{Bi}^{^{(0)}},40~\mu\mathrm{m}$ thickness from Thantawan Industry Plc.Co., Ltd.
 - PLA/PBAT blend (40:60); Bio Flex $^{\circ}$, 100 μ m thickness from FKuR Co., Ltd.
- Black color water base ink for print on plastic from Panorama soy ink Co., Ltd.
- Photopolymer plate Nyloflex ACE series; hardness 64°, shore A and 1.14 mm thickness from Flint group Co., Ltd.
- Backing adhesive tape, 0.5 mm thickness
- RO Water
- Carbon tape

3.1.2 Equipments

- Flexographic printer Nilpeter FB-LINE-3300 from KO Print and Sticker Tunjai Co., Ltd.
- Scanning Probe Microscope (SPM) series Nanoscope IV
- Scanning Electron Microscope (SEM) series JSM 6400
- Optical Microscope; Nikon camera Fiber-optic light
- Contact Angle Meter; Dataphysics series number SCA20 and application software for OCA and PCA

- Corona treat VETAPHONE Corona Plus inline of Flexographic printer Nilpeter FB-LINE-3300
- Tensile Tester LLOYD model LRX plus
- Rub tester SUTHERLAND 2000[™] of Danilee Co., Ltd.
- Spectrodensitometer X Rite 500 series
- Syringe 500 micro liter; Syringe Hamilton 750 NR
- Tape 3M number 500 for test adhesion
- Micrometer
- Poly-Test dyne pen solution number 38-58 dyne/cm
- Microscope; Traveler series SU 1071
- Zahn cup number 3
- pH meter Denver instrument, Model 225

3.2 Methods

3.2.1 Procedure

The procedure can be divided into three major stages;

- 1. Print ability testing of plastic film and ink properties testing
- 2. Flexography print testing
- 3. Print quality assessments

In second stage, Flexography print testing, the sample of plastic films will be printed without surface treatment and then analyze the print quality. If it give satisfactory results of print quality, the conclusion will be made. The reason why there is no surface treatment, because the structure of compostable plastics and its surface morphology fractured as molecular chain, more flexible and high porosity, it can be printed with solvent-based ink without surface treatment. Hence, the hypothesis will be assumed that water-based ink can be printed on that substrate without surface treatment too. However, if the print quality is not good, the surface treatment must be done by corona discharge method before print. The variation of electricity in corona discharge will be used as following; 500, 1000 and 1500 watt, respectively. Then make assessment for the best print quality.





Figure 3-1 The schematic of procedure in the experiment.

3.2.2 Print ability testing of plastic film and ink properties testing

3.2.2.1 Tensile strength and Elongation testing

Tensile strength and Elongation testing of 2 types of compostable plastics compound will be tested before and after print in order to analyze the difference and effect of printing by following ASTM D882-02 standard test method with Tensile tester. The width and length of sample will be recorded. Test sample both in machine direction (MD) and cross-machine direction (CD) with constant speed and load cell at 500 mm/min and 10N, respectively. Make 5 replication and record for average.

3.2.2.2 Surface energy testing

Surface energy testing was done by poly-test dyne solution (dyne pen) before and after surface treatment. Each dye pen has different ratio of solutions similar to dyne solution and it has specified number of surface tension of solution. This surface tension is surface energy of plastic film. Dyne pen was placed on plastic film and draw the line across film width. At first used dyne pen with assumed number which should be the surface energy of plastic film. If the solution still remain stable, does not create water drop on the film, this dyne pen indicate the right number of surface energy of plastic film. If the water drop occur, it indicate that used dyne pen has lower surface energy than plastics film. Suggest using higher number of dyne pen. See figure 3-2



Figure 3-2 Evaluation of dyne pen selection to measure surface energy of plastic film [14]

Dyne pen test was shown in Figure 3-2. It was found that for properly treated film, the solution in dyne pen lied evenly on the material in a continuous line. There is no ink reticulation. The surface energy of the material is at, or higher than the surface tension of the solution in dyne pen. In case of non treated film, the solution reticulates into droplets. The surface energy of the material is well below the surface tension of dyne pen tester. For partial treatment, the solution was partial reticulation from the edges. The surface energy of the material is just below the surface tension of the solution in dyne pen.

<u>1.Properly Treated:</u> The solution in dyne pen lies evenly on the material in a continuous line. There is no ink reticulation. The surface energy of the material is at, or higher than the surface tension of the solution in dyne pen. It is good for print.

<u>2.Not Treated:</u> The solution in dyne pen reticulates into droplets. The surface energy of the material is well below the surface tension of dyne pen tester.

<u>3.Partial Treatment:</u> The solution in dyne pen line is defined but there is partial reticulation from the edges. The surface energy of the material is just below the surface tension of the solution in dyne pen.

3.2.2.3 Surface treatment by corona-discharge

Place the roll of film into printing machine and operate the coronadischarge unit with 500, 1000 and 1500 watt, respectively. Operating speed will be 30 m/min. Then test the wettability by contact angle measurement.

3.2.2.4 Wettability testing of plastic film

The wettability of the plastic films was tested by contact angle measurement of RO water and black water-based ink.

PBAT/corn starch (30*0.5cm)

- Non-Corona
- Corona 500 watt
- Corona 1000 watt
- Corona 1500 watt

PAL/PBAT (30*0.5cm)

- Non-Corona
- Corona 500 watt
- Corona 1000 watt
- Corona 1500 watt

The contact angles of plastic films were measure with DataphysicsSCA20 instrument by randomly on the sample roll. The three sample pieces of each corona treatment condition were used. The samples were carefully cut to avoid touching the surface test area, because grease and sweat from finger will contaminate plastic film. Place plastic film evenly on sample holder in the middle. Fill the desired solution into 500 μ l syringes; specify each drop as 5 μ l with high speed injection. Measure contact angle in left and right of droplet 6 times and calculate the average.



Figure 3-3 Contact angle measured of solution.

3.2.2.5 Ink viscosity measurement

Measure the viscosity of ink by using Zahn cup No.3. Place the finger under the hold of Zahn cup and fill ink sample until the edge of cup. Move finger away to let ink flow, start timer quickly. Then stop the timer when flow disappears at first time. Make at least three replications and average value in second unit.

3.2.2.6 Measure pH of ink

Measure pH of ink with pH meter before print. The pH value should be 8-9.5 for dispersion property. If it is too acidic, ink will become agglomerate and precipitate. It causes the low print quality.

3.2.3 Flexography print testing

3.2.3.1 Printing condition

In the printing stage, the printing condition must be controlled as figure 3-4. Adjust printing machine with constant printing speed at 30 m/min, web tension in unwind (= 0) as same as rewind (= 0) and control drying unit at medium temperature



Figure 3-4 The controlled condition for flexography print testing

3.2.3.2 Test form design

Create test form file to evaluate all print quality. The halftone test patch 133 lpi resolution used to evaluate the print quality. Generally the higher resolutions is not used for flexographic printing. The test from is shown in figure 3-5. The test from composed of

- (A) Gradients tone which indicate the screen from 1 to 100 percent in order to measure dot gain and print density
- (B) Tool stripe to evaluate detail of micro text in each specified condition. It has both positive (printed text) and negative letter. (reverse in printed text)
- (C) Micro dot and micro line with thickness from 0.1 to 1.0 mm for evaluate the detail of smallest dot and line which can be printed in each condition.
- (D) Image area for assessment in high light, mid-tone and shadow.
- (E) Micro dot and micro line with thickness from 0.025 to 0.350 mm. It has the same function as C, but has smaller line.
- (F) Slur and doubling



Figure 3-5 Test form for printing and print quality assessment

3.2.3.3 Anilox line resolutions

The anilox line resolutions of 400, 600 and 700 line per inch (lpi) were used in order to compare print quality.

3.2.4 Print quality assessments

3.2.4.1 Optical density

The optical density of all plastic films were measure opcital density and evenness of printed ink on plastic film with Spectrodensitometer X Rite 500 series. The equipment was calibrated before taking the measure. The measure were taken on 9

positions of solid tone of each sample with paper making. Then the data from 3 samples of each treatment condition were average and recorded.

3.2.4.2 Print contrast

The print density at 75% dot area and 100% dot area were calculate to obtain the print contrast by the equation 3-1. [14]

Print contrast =
$$((D_{s} - D_{T})/D_{s}) \times 100\%$$
 (Eq. 3-1)

3.2.4.3 Tone value increase (TVI)

Measured tone value increase (TVI) with Spectrodensitometer X Rite 500 series by using the same condition as print density measurement. Measure on non-printed area of the plastic and then measure solid area, measure the screen area at 20, 40 and 80%, respectively. Calculate dot gain by equation 3-2.

3.2.4.4 Evaluate the thickness and ink film condition with SEM

Clean the surface of printed film and prepare the sample by cutting sample at 100% dot area. Then place into liquid nitrogen and cut into 1x1 cm. Make the cross section to evaluate the thickness by placing on stub with carbon adhesive tape. The samples gold coated for conductivity and observe the sample under SEM at 10,000x 3.2.4.5 Evaluate distribution of ink on film

Preparing the film by cutting sample at solid tone area of each surface treated level, 1×0.5 cm² size. After that capture the image by optical microscope at 10x with Nikon camera using Fiberoptic light source.

3.2.4.6 Roughness

Prepare an untreated film and a treated film at 1000 watt with 2x2 cm size. Capture the image under SPM microscope at 5000x and the roughness value are obtained.

3.2.4.7 Ink Adhesive test

Test adhesion of printed ink on substrate with ASTM D3359-09 standard test method. The samples must be conditioned in room temperature more than 48 hours before testing to let ink dry completely. The samples were cut at gradient tone 0-100% dot area. Use adhesive tape, 3M No. 500, with size 24×70 mm. and place on plastic film smoothly without trapped bubble. Peel tape at 180 degree within 90 second. Place adhesive tape on graph paper with 0.5 mm width. Evaluate amount of ink came off in 100 cycle of graph paper (1 cycle equal to 1%) to make the assessment. [6]

Score level	Evaluation criterion
5	non-peeled ink
4	5 % peeled ink
3	5-15 % peeled ink
2	15-35% peeled ink
1 35-65	% peeled ink
0	65 % peeled ink

Classification	Surface of cross-cul area from which flaking has occurred. (Example for six paralled cuts)
58	None
48	
38	
28	
18	
08	Greater than 65%

Figure 3-6 The evaluation of adhesive test [4]

3.2.4.8 Rub resistance test

Test rub resistance with ink rub tester with ASTM D5264-98 standard test method. Before test keep plastic film in room condition more than 48 hours to let ink dry completely. Use four different speed and 4 pound weight for testing. The samples was cut by 9×18 cm. and mount both on top of the Sutherland receptor base as well as to the bottom face of the detectable receptor block, each test area must be the same. Place white paper on the sample holder and tester. Place sample on the sample holder firmly and smoothly, then placed unprinted area film (same as printed film) on the tester. Make 10, 100 and 500 times of rub testing, respectively. Each times must be evaluated and scanned with Canon Scan LIDE 60 at 300 dpi. Evaluate the unprinted film for peeled ink by visual assessment as same as ink adhesion.

Score level	Evaluation criterion
5	non-peeled ink
4 5	% peeled ink
3	5-15 % peeled ink
2	15-35% peeled ink
1 35-65	% peeled ink
0 65	% peeled ink

3.2.4.9 Image resolution assessment

Prepare all printed test sample both treated and non-treated film. Capture image under 10x microscope at micro line, micro dot, 20, 40, 50 and 80% dot screen, then evaluated by visual assessment

CHAPTER 4

RESULTS AND DISCUSSION

4.1 The first printing with non- treatment

The first printing was done by non-treatment compostable plastic surface and print quality analysis that consisted of print contrast, density, TVI, rub resistance and adhesion test and considered about usability. Tensile strength, elongation and surface energy of compostable plastics were tested. The results of samples were shown in table 4-1. Properties of black water acrylic based flexo ink were measured before printing.

Table 4-1 Result of tensile strength, elongation and surface energy of compostable plastics before printing.

	Surface Energy (Dyne/cm)	Tensile strength (Kg/mm ²)		Elongation (%)	
		MD	CD	MD	CD
PBAT/Starch	38	2.30	1.42	461	744
PLA/PBAT	38	1.68	1.86	443	460

Table 4-1 shows the results of tensile strength, surface energy and elongation of compostable plastics before printing. We found the measured surface energy values of two plastic films were closed. This could expect to have similar behavior of ink wettability.

The measured surface energy of two plastic films is same value. From surface energy, both of sample compostable plastics were expected to show similar behavior for wetting and imaging by acrylic water-based flexographic ink. Tensile strength and elongation of PBAT/Starch along the direction of MD was higher than CD direction. Therefore, the tension setting should be careful for protecting film roll from being raged. Since the elongation was high, expansion of substrate should be concerned as this effect made the abnormal size. The tensile strength of PLA/PBAT along the MD and CD directions was not much different. But these values were lower than those of PBAT/Starch, including high thickness case. This could be said that PBAT/Starch could be proper for in-line web flexo press. The measurement of surface tension and viscosity of the acrylic water based flexo ink was 40 dyne/cm and 23 seconds respectively. pH was at 8.

4.1.1 Optical Density

Generally, the optical density of black on compostable plastic had value in range of 1.40 - 1.45. The density of printing was shown in figure 4-1. The optical density measured on 100% dot screen in gradient chart area. The density PBAT/Starch was higher than PLA/PBAT. Since anilox line resolution was adjusted higher, the density of both of compostable plastic trended to decrease. Because ink volume of anilox roller was decreased and structure of surfaces were different.



Figure 4-1 Print density of ink layer on PBAT/Starch and PLA/PBAT used three levels anilox line resolution 400, 600 and 700 lpi.
4.1.2 Print Contrast

Print contrast showed contrast of Mid-tone and Shadow area of printing. Print density was measured from 75% to 100% of screen dot area. Print contrast was shown in table 4-2. The result showed that print contrast of PBAT/Starch was higher than PLA/PBAT. Probability of PBAT/Starch had structure and surface that could contain ink better than PLA/PBAT. In case of PLA/PBAT, it was transferred to surface. That was why it could not be stable. Because spreading effected on detail of Mid-tone so it was shifted to be Shadow.

Table 4-2 Print contrast on PBAT/Starch and PLA/PBAT used three levels anilox line resolution 400, 600 and 700 lpi.

	Print contrast		
	Aniox res. Aniox res. Aniox res.		
	400 (Ipi)	600 (Ipi)	700 (Ipi)
PBAT/Starch	26.87	18.01	17.61
PLA/PBAT	15.51	9.33	1.03

In case of, print contrast and density with non-treatment of surface, anilox line resolution for PBAT/starch should be set at 600 or 700 lpi. Because of anilox line resolution at 400 lpi could dispense too high print contrast and tone could not be varied. And for PLA/PBAT should be set at 400 lpi.

4.1.3 Ink adhesion and Rub resistance of non-treatment compostable plastic samples

Ink adhesion testing could be done by using Tape test in region of continues tone in media. Rub resistance could be done by using Rub tester and tested region of people face in media. Number of rub was set to be 10, 100 and 500 times.

After 500 times was rubbed, quantities of ink that peeled from media were evaluated. The result was shown in Table 4-3. However, ink adhesion of both of samples was low and could not be inacceptable in reality. Rub resistance of PBAT/Starch could be satisfied but PLA/PBAT could not be accepted. Because just 10 times rub on PLA/PBAT could peeled off ink from substrate and the picture shown in figure 4-11.

Table 4-3 Quantities of peeled ink.

	Ink adhesion	Rub resistent
PBAT/Starch	0	2
PLA/PBAT	0	0

Score level	Evaluation criterion
5	non-peeled ink
4	5 % peeled ink
3	5-15 % peeled ink
2	15-35% peeled ink
1 35-65	% peeled ink
0	65 % peeled ink

Print contrast, Ink adhesion and Rub resistance showed that non-treatment was low quality or unusable. That is why we should to treat surface before printing. Moreover, this study had another part of experiments for evaluating qualities of non-treatment surface that was compared with treated surface by corona discharge before printing.

4.2 The second printing experiment with corona treatment

Corona-treatment varied to be three levels of corona discharge. All of three levels were analyzed for searching the level that gave desired quality and usable printing. Surface treatment was done by using corona unit and varied three level of discharge as 500, 1000, and 1500 watt and compared for selecting optimization of condition for printing.

This is the example printed before and after corona treatment of sample compostable plastics.





PBAT/Starch Non-corona





PBAT/Starch Corona

Figure 4-2 Examples image area printed on PBAT/Starch non-corona and corona treatment at 500 watt and used anilox line resolution at 700 lpi.



PLA/PBAT Non-corona





PLA/PBAT Corona

Figure 4-3 Examples image area printed on PLA/PBAT non-corona and corona treatment at 500 watt and used anilox line resolution at 700 lpi.

Figure 4-2 and 4-3 show the test form printed images on non-corona and corona treated PBAT/Starch and PLA/PBAT at 500 watt and anilox line screen 700 lpi. It should be noted that this chosen printing condition gave optimum print quality based on highest print contrast.

4.2.1 Tensile strength and Elongation

Tensile strength and elongation showed that both of samples trended to decrease after treatment by high level of discharge. The results were shown in Table 4-4 and 4-5. Since, high charge was shot on surface that made roughness deeply and some bond of polymer was destroyed and strength decreased.

Table 4-4 Tensile strength of MD and CD of PBAT/Starch and PLA/PBAT before and after surface treatment at 1500 watt

	Tensile strength (Kg/mm ²)				
Sample	Before corona treat		After corona treat		
	MD	CD	MD	CD	
PBAT/Corn starch	2.30	1.42	1.89	1.41	
PLA/PBAT	1.68	1.86	1.24	1.07	

Table 4-5 Elongation of MD and CD of PBAT/Starch and PLA/PBAT before and after surface treatment at 1500 watt

	Elongation (%)				
Sample	Before corona treat		After corona treat		
	MD	CD	MD	CD	
PBAT/Corn starch	461	744	414	715	
PLA/PBAT	443	460	324	395	

4.2.2 Surface energy

Surface energy was tested by using poly-test dyne pen. When surface was treated, surface energy would increase. This experiment was limited because poly-test dyne pen had surface energy at 38-56 dyne/cm. So surface energy of samples before corona-treatment, might be lower than 38 dyne/cm.

 Table 4-6
 Surface energy of compostable plastics before surface treatment and treatment with different level.

		Surface Ener	gy (dyne/cm)	
Sample	Non-corona	Corona 500	Corona 1000	Corona 1500
		(watt)	(watt)	(watt)
PBAT/Starch	38	44	48	52
PLA/PBAT	38	40	44	46

4.2.3 Wettability

Wettability was done by measuring of contact angle of OR Water and acrylic water base ink. Through this method, we could simulate and analyze wettability of plastic. If contact angle decreased that means wettablility of surface got better.



Figure 4-4 Contact angle of OR water on surface of compostable plastic by 4 levels of corona treatment

Contact angle of OR water in case of corona treat at 1000 and 1500 watt was abnormal and disagreed. The theory mention to corona treatment with higher electric power was good wettability property. However, the result showed contact angle that was unstable so we could not analyze. We assumed that might caused surface was touched too much.



Figure 4-5 Contact angle of black acrylic water base ink on surface of compostable plastic by 4 levels of corona treatment

4.2.4 Optical density

Density of ink on compostable plastic was measured in area of 100 %. In figure 4-6 and figure 4-7, The optical density measured on 100% dot screen in gradient chart area. Optical density had low resolution at 400 lpi that means its density was higher than 600 and 700 lpi. Considered level of corona treatment by using higher discharge, optical density significantly increased at any anilox line resolution. It followed the theory of anilox line solution that refer to high resolution effect to film thickness was decreased. So optical density decreased. Optical density showed that anilox line resolution at 700 lpi and corona at 500, 1000 watt should be selected.



Figure 4-6 Optical density of ink on PBAT/Starch at anilox line solution 400, 600 and



700 lpi on different surface treatment levels.

Figure 4-7 Optical density of ink on PLA/PBAT at anilox line solution 400, 600 and 700 lpi on different surface treatment levels.

4.2.5 Tone value increase on compostable plastic samples

Figure 4-8 and Figure 4-9 shows that corona treatment with increased charges, TVI increased as well. Because properties of well ink contain, extension and

distribution of dot increased. Level of corona treatment that suited with PBAT/Starch was 500 watt and PLA/PBAT suited with both of 500 and 1000 watt. Considered to varied anilox line resolution at 700 lpi optimized for printing.





Figure 4-8 Comparision of TVI printing on PBAT/Starch with different anilox line resolution at 400, 600 and 700 lpi





Figure 4-9 Comparision of TVI printing on PLA/PBAT with different anilox line resolution

at 400, 600 and 700 lpi.

4.2.6 Print contrast of compostable plastics with corona treatment

Print contrast was normally prior considered to choose the proper printed sheet with right amount of ink. The obtained print quality may be called as an optimum quality. It was found that surface treatment levels that suited with PBAT/Starch and PLA/PBAT could be at both 500 and 1000 watts and anilox line screen 700 lpi. Highest print contrast of PBAT/Starch was achieved only at 21 and PLA/PBAT 18-19 as given in Table 4-7 and 4-8. This property could identify the deviation of shadow rendering between two samples which showed that PBAT/Starch seemed to give better quality.

Table 4-7 Print Contrast on PBAT/Starch after surface treatment with different level and using anilox line resolution at 400, 600 and 700 lpi

	Non-corona	Corona 500	Corona 1000	Corona 1500
		(watt)	(watt)	(watt)
400 lpi	26.87	20.85	17.90	14.21
600 lpi	18.01	16.51	9.43	20.52
700 lpi	17.61	21.38	21.86	22.02

Table 4-8 Print contrast on PLA/PBAT after surface treatment with different level and using anilox line resolution at 400, 600 and 700 lpi

	Non-corona	Corona 500	Corona 1000	Corona 1500
		(watt)	(watt)	(watt)
400 lpi	15.51	3.85	4.43	2.13
600 lpi	9.33	7.33	14.15	17.61
700 lpi	1.03	17.99	19.14	17.51

4.2.7 Print uniformity

This parameter relates to substrate uniformity and wettability of ink. In case of non-corona treatment, wettability of ink was not good. This resulted in ink aggregation and peeled off easily. However, after corona treatment at 500 and 1000 watts, improved print uniformity was existed. However, it was noted that thinner substrate or lower density had limitation in corona treatment level. Thus the PBAT/Starch film sample could not be treated over 1000 watts as the surface profile would be damage. This affected losing of ink adhesion. For PLA/PBAT, its behavior was similar to PBAT/Starch. Thus, 500 watts could be the satisfied power level of surface treatment of compostable films. Figure 4-10, 4-11 show the appearance of print uniformity on PBAT/Starch and PLA/PBAT respectively, of solid tone area with different surface treatment level: (A) anilox line screen 400 lpi (B) anilox line screen 600 lpi and (C) anilox line screen 700 lpi.

 \rightarrow ink aggregation - - \rightarrow no ink aggregation









Figure 4-10 Print uniformity on PBAT/Starch sample with different surface treatment level: (A) anilox line screen 400 lpi (B) anilox line screen 600 lpi (C) anilox line screen





(E)



Figure 4-11 Distribution of ink on PLA/PBAT in 100% dot with different surface treatment (D) Anilox line resoultion at 400 lpi (E) Anilox line resoultion at 600 lpi (F) Anilox line resoultion at 700 lpi

4.2.8 Roughness

PBAT/Starch was lower than PLA/PBAT. We considered that it was because charges effected the vibration of molecule. Table 4-9 shows the result of roughness on surface PBAT/Starch before corona treatment compared with those after corona treatment by using electric 1000 watts.

	PBAT/cor	n starch	PLA/PBAT	
	Before corona	After corona	Before corona	After corona
	Treatment	Treatment	Treatment	Treatment
Roughness : Rms (Rq)	207.78	236.66	435.3	540.98
Mean roughness (nm)	171.30	181.8	344.13	417.90
Max height (nm)	2.012	2.593	2.446	3.550
Surface area (µm²)	2575.1	2690.9	2547.2	2645.3

Table 4-9 Result of roughness on surface PBAT/Starch before corona treatment compared with after corona treatment by using electric 1000 watt

Considered figure 4-12 shows the SPM images of substrate surfaces before and after surface treatment. It was found that RMS surface roughness value of PBAT/Starch was higher than that of PLA/PBAT. This probably caused PBAT/Starch well contained and cohesion on surface.



(A)



Figure 4-12 Roughness of two films surface in 3D at magnification 50 μm (A) surface before corona treatment, (B) surface after corona treatment

4.2.9 Evaluation of thickness and ink film condition by using SEM

Figure 4-13 shows aggregation of ink. Thickness of ink on PBAT/Starch was about 1.2 μ m and PLA/PBAT was 2.0 μ m approximately.



PBAT/Starch



PLA/PBAT

Figure 4-13 Aggregation of PBAT/Starch and PLA/PBAT in 100% dot area and treatment at 1000 watt were observed by using SEM at 10,000x magnification.

4.2.10 Ink adhesion test

Table 4-10 Adhesion of ink water-based on PBAT/Starch and PLA/PABT

	Ink adhestion on	Ink adhesion on
	PBAT/starch	PLA/PABT
Non-corona	0	0
Corona 500 watt	0	0
Corona 1000 watt	1	0
Corona 1500 watt	2	0

<u>S</u>	core level	Evaluation criterion
	5	non-peeled ink
	4	5 % peeled ink
	3	5-15 % peeled ink
	2	15-35% peeled ink
1 35-65		% peeled ink
	0	65 % peeled ink

4.2.11 Rub resistance

Rub resistance was divided to be three level as 10, 100 and 500 times. Evaluation of rub resistance was estimated after 500 times rubbed. Through evaluation criterion of rub resistance was same as ink adhesion that was shown in Table 4-8. The result showed that PBAT/Starch was good rub resistance after treatment at 500 watt as shown in figure 4-14. Table 4-11. Rub resistance of ink water-based on PBAT/Starch and PLA/PABT with anilox line resolution at 600 lpi and 500 times rubbed.

	Rub resistance of	Rub resistance of
	PBAT/ starch	PLA/PABT
Non-corona	3	0
Corona 500	5	1
Corona 1000	5	1
Corona 1500	5	2

PBAT/Starch Non-corona treatment			
No Ru	b Tester	10 times	
100 times		500 times	

PBAT/Starch Corona treatment 500 watt			
No Rub Tester		10 times	
100 times		500 ti	mes

PBAT/Starch Corona treatment 1000 watt			
No Rub Tester		10 times	
100 times		500 times	



Figure 4-14. Result rub tester on PBAT/Starch at anilox line resolution 600 lpi with nontreatment and every level of corona-treatment

PLA/PBAT got better properties after treatment but peeling off ink still was problem as shown in figure 4-15.

PLA/PBAT Non-corona treatment			
No Rub Tester		10 times	
100 times		500 ti	mes

PLA/PBAT Corona treatment 500 watt			
No Rub Tester		10 times	
100 times		500 t	imes

PLA/PBAT Corona treatment 1000 watt			
No Rub Tester		10 times	
100 times		500 times	

PLA/PBAT Corona treatment 1500 watt			
No Rub	o Tester	10 tim	nes
100	times	500 tin	nes

Figure 4-15 Results of Rub Tester on PLA/PBAT at anilox line resolution 600 lpi with noncorona treat and every level of corona-treatment

This experiment showed some error from PBAT/Starch. Low thickness effected to friction between film and plate that rubbing was unstable on all surfaces. So some region could not be attached by ink.

4.2.12 Image resolution

This experiment was done by observing by using optical microscope on each part of platform of printing. Resolution of screen dot was considered how relate with three levels of anilox line resolution and 4 levels of corona treatment. Actually, images from each condition didn't show difference of screen dot so much. However, the image that showed difference was from anilox line resolution at 700 lpi and coronatreatment level at 500 watt. Figure 4-16 and figure 4-17 shows PBAT/Starch and PLA/PBAT that detailed resolution of image in the smallest positive line and negative line at 0.025 mm and 0.1 mm, respectively. Sizes of the smallest dot positive and dot negative could be printed by size 0.15 mm and 0.2 mm, respectively.



Figure 4-16 Negative micro dot tap on both of compostable plastics, corona treatment at 500 watt and used anilox line resolution at 700 lpi



PBAT/Starch





Figure 4-17 The smallest negative and positive micro line that could be printed after corona-treatment at 500 watt and used anilox line resolution at 700 lpi

Dot in area of dot screen at 20, 40, 50 and 80% dot area were shown for comparing dot gain. For supporting of anilox line resolution at 700 lpi that was the optimized condition, the result was shown in figure 4-18 and 4-19. They were comparison between dot gain at 40% and 50%. The result showed that shape of dot gain at 700 lpi was more beautiful than 400 and 600 lpi. Both films were the best

substrates by which they were photographed by anilox line resolution at 700 lpi and the optimization of surface treatment level was 500 or 1000 watt







Figure 4-18 Dot gain on PBAT/Starch in %dot area of 20, 40, 50 and 80% after

treatment at 500 watt







Figure 4-19 Dot gain on PLA/PBAT in %dot area of 20, 40, 50 and 80% after treatment at

500 watt

CHAPTER 5

CONCLUSIONS

5.1 Conclusions

This study showed that both compostable film samples could be printed by flexographic printing, using acrylic water base ink. However, it was found that ink adhesion and rub resistance were insufficient. This included their obtained reflectance solid tone densities. It were because these compostable films were hydrophobic and low surface energy. Thus, to achieve good printability, their surfaces should be treated to increase surface energy.

Printing with non-treatment surface on PBAT/Starch and PLA/PBAT film could be satisfied about TVI and print contrast at anilox line resolution at 700 lpi and 400 lpi, respectively. Value of print density, ink adhesion, rub resistance and ink distribution of two film compostable plastics compounds could not be accepted. Because of compostable plastic surfaces were hydrophobicity and low surface energy. In case of printability and usability, their surface should be treated by corona to increase surface energy.

Surface treatment not only improved the surface energy, but also increased surface roughness. These gave the tendency of ink acceptability and print quality such as print density, Ink rub resistance and print uniformity. However, level of corona discharge of surface treatment should be determined as the surfaces of such substrates could be easily damaged. In case of too high discharge, it affected the property of film strength such as tensile strength and elongation. In addition, print uniformity at 1500 watts corona discharge became worse as the occurrence of surface damage

The both of films was the best substrates when they were treated by Anilox line resolution at 700 lpi and the optimization of surface treatment level was 500 or 1000 watt where stable velocity at 30 m/min. Nevertheless, treatment at 500 watt should be selected because electricity was saved.

By those conditions, Quality of PBAT/Starch was better than PLA/PBAT but the TVI and ink adhesion still needs to be improved.

5.2 Suggestions

- Comparison of print quality of compostable plastic that is printed by using water base ink and solvent base ink.

- Comparison of quantities of water base ink and solvent base ink that are printed on compostable plastic compound for according to ASTM 17088:2008.

- Comparison of quantities of ink that is used on compostable plastic by Flexography printing and Gravure printing for according to ASTM 17088:2008.

- Studying of penetration of ink in compostable plastic compound starch.

- Acrylic water base ink formula should be improved in order to increase ink adhesion for improving printability on compostable plastics.

- Comparison of print quality of round dot and shape dot about hardness by using different kinds of plate.

- In case of Flexo-graphy printing, print velocity should be faster than 30 m/min.

- Evaluation of wettability can be done by measuring of contact angle and solution should be OR water, Diiodomethane or Ethylene glycol for calculating surface energy replace of Poly-Test Dyne pen.

REFERENCE

- [1] Association for Oraganics Recycling. Concise gide to compostable products and packaging. <u>UK Local Authority Guidance</u> (2011): 2-5.
- [2] Phietoon Trevijitkasem. Green Packaging. <u>Journal of Printing, Packaging and</u> <u>imaging Technology (Thailand)</u> vol.24 No.97 (2011): 18-26.
- [3] ISO17088:2008(E). Specifications for compostable plastics. 2008: 5.
- [4] Pradya Khiangprakhong. <u>Surface Modification of Biodegradable Plastics by Corona</u> <u>Discharge</u>. Bachelor of Science. Department of Photographic Science and Printing Technology. Chulalongkorn University, 2008.
- [5] Jie, Ren., and other. Pretaration, characterization and properties of binary and ternary blends with thermoplastic starch, poly(lactic acid) and poly(butylenes adipate-co-terephthalate). <u>Carbohydrate Polymer</u> 77 (2009) : 576-582
- [6] Janejira Phunkoksung and Sroymanee Nimnuan. <u>Biodegradability and Printing</u> <u>Properties of Soy-bean Ink on Biodegradable Film</u>. Bachelor of Science. Department of Printing and Packaging Technology. Industrial Education and Technology King Mongkut's University of Technology Thonburi, 2552.
- [7] Amorrat Lasvarasirikhun. Poly(lactic acid):Polyester from Renewable Resources. <u>Risk Assessment of Traction Battery Charging System in a warehouse.</u> 24 (July-September 2011): 99-110
- [8] Phietoon Trevijitkasem. Standard and determination of Compostable plastic in Thailand, <u>BioPlastics</u> vol.4 No.2 (April-June 2012): 6-7.

- [9] Faculty of Pharmacy, Silpakorn University. <u>PLA (Polylactic Acid)</u>. [online]. Available
 from : http://www.pharm.su.ac.th/cheminlife/cms/index.php/product-name/
 product-name-english/662-polylactic-acid.html [2012,Jun 2]
- [10] Foundation of Flexographic Technical Association, Inc. <u>Flexography: principles</u> <u>and practices</u>. 5th Edition. United State of America: Foundation of Flexographic Technical Association, Inc, 1999.
- [11] Johnson, J. <u>Aspect of flexographic print quality and relationship to some printing parameters</u>. Faculty of Technology and Science Chemical Engineering. Karstad University, 2008.
- [12] Kipphan. technologies and production methods. <u>Handbook of print media</u>. Germany: springer-Veriag Berlin Heidelberg, 2001.
- [13] Rentzhog, M. and Fogden, A. Print quality and resistance for water-based flexography on polymer-coated boards: Dependence on ink formulation and substrate pretreatment. <u>Progress in organic coatings</u>. 57 (2006): 183-194.
- [14] Marc Nolan Converting Supply Inc. <u>Dyne Testing Why Test In The First Place?</u>, 20/07/2012.
- [15] <u>Print contrast</u>. [online]. Available from : http://www.printwiki.org/Print_Contrast.
 [2012,Jun25]
- [16] Gilberd, E.D and Frederick, L. Flexography Plate Technology: Conventional solvent plates versus Digital solvent plates. <u>Journal of industrial technology</u> V24 (3) July (2008).

APPENDICES

Appendix A

Materials and Equipments



Figure 1. Sample compostable plastics film PBAT/Starch (left); 40 µm thickness and PLA/PBAT (right); 100 µm thickness substrate for printed in this experiment.



Figure 2. Photopolymer plate hardness 64°, shore A and 1.14 mm thickness from Flint group Co., Ltd.



Figure 3. Acrylic water-base ink Panorama soy ink Co., Ltd.



Figure 4. Flexographic printer Nilpeter FB-LINE3300; KO print and sticker Tunjai.Co.Ltd.


Figure 4. Corona treatment inline of Flexographic printer Nilpeter FB-LINE3300



Figure 5. Zahn cup number 3 for measured viscosity of ink in this experiment.



Figure 6. Micrometer for measured thickness sample of compostable plastics.



Figure 7. Tensile tester LLOYD model LRX plus for measured Tensile strength and Elongation.



Figure 8. Rub tester SUTHERLAND 2000[™] for rub resistance test.



Figure 9. Poly-Test dyne pen solution number 38-58 dyne/cm for test surface energy on both of sample compostable plastics.



Figure 10. Contact Angle Meter; Dataphysics series number SCA20 and application software for OCA and PCA used measure contact angle of OR water and acrylic water base ink.



Figure 11. Scanning Electron Microscope (SEM) series JSM 6400

Appendix B

Printed line resolution of image on sample compostable plastics

The result aside from chapter 4. This is the picture printed image line resolution varied anilox line resolution three levels of anilox line resolution and 4 levels of corona treatment. Capture by optical microscope.



Figure 12. Negative micro dot tap on PBAT/starch used anilox line resolution at 400 lpi





Figure 13. Negative micro dot tap on PBAT/starch used anilox line resolution at 600 lpi



Figure 14. Negative micro dot tap on PBAT/starch used anilox line resolution at 700 lpi



Figure 15. Negative micro dot tap on PLA/PBAT used anilox line resolution at 400 lpi





Figure 16. Negative micro dot tap on PLA/PBAT used anilox line resolution at 600 lpi



Figure 17. Negative micro dot tap on PLA/PBAT used anilox line resolution at 700 lpi



Figure 18. Image positive line and negative line 0.025 mm (left) and 0.1 mm. (right) on PBAT/Starch used anilox line resolution at 400 lpi Non-corona, Corona 500, 1000 and 1500 watt



Figure 19. Image positive line and negative line 0.025 mm (left) and 0.1 mm. (right) on PBAT/Starch used anilox line resolution at 600 lpi Non-corona, Corona 500, 1000 and 1500 watt



Figure 20. Image positive line and negative line 0.025 mm (left) and 0.1 mm. (right) on PBAT/Starch used anilox line resolution at 700 lpi Non-corona, Corona 500, 1000 and 1500 watt



Figure 21. Image positive line and negative line 0.025 mm (left) and 0.1 mm. (right) on PLA/PBAT used anilox line resolution at 400 lpi Non-corona, Corona 500, 1000 and 1500



Figure 22. Image positive line and negative line 0.025 mm (left) and 0.1 mm. (right) on PLA/PBAT used anilox line resolution at 600 lpi Non-corona, Corona 500, 1000 and 1500



Figure 23. Image positive line and negative line 0.025 mm (left) and 0.1 mm. (right) on PLA/PBAT used anilox line resolution at 700 lpi Non-corona, Corona 500, 1000 and 1500

Appendix C

Sample of compostable plastics

PBAT/Starch

PBAT/Starch print on plate resolution 133 lpi

PLA/PBAT

PLA/PBAT print on plate resolution 133 lpi

VITA

Ms. Samatcha Nawakitwong was bone in 16 Aug, 1986 in Bangkok, Thailand. She received a Bachelor's Degree of Department of Printing and Packaging Technology, Faculy of Industrual Education and Technology from King Mongkut's University of Technology Thonburi in 2008. Her Senior project about Paper making from Used Chopsticks. She worked about flexible packaging after graduated Bachelor's Degree around 1 years. She entered the Department of Imaging and Printing Technology, Faculty of Science, the Graduate School, Chulalongkorn University in 2012.