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APPLICATION OF PALM FATTY ACID DISTILLATE AS COMPATIBILIZER ON THERMOPLASTICIZED CASSAVA FLOUR-LLDPE COMPOSITE FILM

SUGIARTO[1], TITI CANDRA SUNARTI[2], ANI SURYANI[3], SUTRISNO[4] & INDAH YULIASIH[5]

[1]Doctoral Student, Agroindustrial Technology, Bogor Agriculture University, Indonesia

[2,3,5]Department of Agroindustrial Technology, Faculty of Agricultural Technology, Bogor Agricultural University, Indonesia

[4]Department of Machinary and Biosystem Engineering, Faculty of Agricultural Technology, Bogor Agricultural University, Indonesia

ABSTRACT

Composite plastic is plastic which made from synthetic polymer and biopolymer blends with the addition of some additives. The objective of this study was to produce composite plastic bag from linear low density polyethylene (LLDPE) resin and cassava flour blends with the addition of additives such as water and glycerol as plasticizer and palm fatty acid distillate (PFAD) as compatibilizer. The result showed that the best formulation to make plastic bag was LLDPE and cassava flour with ratio of 7:3, glycerol 30%, and PFAD 5%. This formulation made composite plastic which had specific gravity 0.916 and melt flow index 4.45 gr/10 min. Composite plastic film had thickness of 250 micron and the texture of its surface was relative smooth. Tensile strength and elongation value of composite plastic film at each orientation were 3.71 MPa and 396.18% for machine direction orientation, 3.06 MPa and 126.29% for transverse direction orientation, and 3.11 MPa and 137.06% for heat sealing orientation. The value of whiteness index, yellowness index and opacity of composite plastic film was 6.36, 16.28 and 18.13, respectively.

KEYWORDS: Plastic Bag, Composite, Cassava Flour, Glycerol, LLDPE, PFAD

INTRODUCTION

Plastic bag is one kind of packaging made from plastic. Most of conventional plastic bags are made from polyethylene. This material was chosen for several advantages i.e. ability for blowing film processing, high tensile strength and elongation, light weight and low cost. However, polyethylene is difficult to degrade. Therefore, significant and continue utilization of polyethylene in producing plastic bags bring serious impact to environment.

Composite plastic bag is one of alternatives that could overcome the problem mentioned above. In the production of composite plastic bag, biopolymer is used during the process. Therefore, it is relatively more environmentally friendly material. Composite plastic is plastic made from biopolymer and synthetic polymer added with additive materials. Numerous researches concerning on plastic which made from biopolymer and synthetic polymer had been conducted before. Permatasari (2010) had conducted research concerning on composite plastic made from thermoplastic cassava-dried cassava with compatibilized polyethylene.

Cassava flour can be used as raw material in the manufacturing process of composite plastic as it contains biopolymer such as starch. Cassava flour is regarded as potential material that has some advantages including low cost,

abundant availability, and renewable resource. Nevertheless, the weakness relies on non-elastic material and can't be printed because the starch component is rigid. Material which has rigid property is mainly due to high glass transition temperature and crystalline molecule structure (Wade, 1991). Modifying the starch component contained in the cassava flour through thermoplastic process is required to obtain more plastic and elastic starch. One of the materials used for plasticizer is glycerol. As a plasticizer material, glycerol is not easily evaporated as glycerol has high boiling point of 290°C.

Geng (2005) explained that biopolymer and synthetic polymer are two materials that are not mutually compatible. This because of the biopolymer is polar(hydrophilic) while the synthetic polymer is nonpolar (hydrophobic). This condition will produce weak bonding interface of final product which formed between two different phases. To overcome this condition, additive materials used for compatibilizer are required in the mixing process of synthetic and biopolymer. One of the materials than can be used is stearic acid. Stearic acid is one of fatty acids compound in vegetable oil or fat. In the previous research, Enriquez *et al.* (2010) used stearic acid as compatibilizer to produce composite plastic from a mixture of high density polyethylene and coconut fiber. If stearic acid could be used as compatibilizer in the manufacturing process of composite plastic, then this study was also expected to determine the ability of PFAD which known as a mixture of fatty acid for compatibilizer material.

The objectives of the research was to 1) make composite plastic bag made from LLDPE resin and cassava flour added with additive materials such as water and glycerol as plasticizer and PFAD as compatibilizer; 2) determine the characteristic of the composite plastic bag.

METHODS

Tools and Materials

Materials used in this research were cassava flour sized 100 mesh, linear low density polyethylene (LLDPE) resin UF1810 and UI2420 from PT. Chandra Asri Petrochemical, glycerol and water as plasticizer, and PFAD from PT SMART Tbk. as compatibilizer. Tools used in this research were compression-type kneading and mixing machine model ML-5L for plasticizer and compounding, crusher FRB-7.5, and blowing film line with dies for LLDPE film. Analysis instruments used in this research were Universal Testing Machine from Lloyd Instrument, Gretagmacbeth color i5 spectrophotometer, Melt Flow Indexer Frank, and moisture analyzer AND MS-70.

METHODS

Characterization of Cassava Flour

Proximate analysis was conducted to determine the characteristic of cassava flour sized 100 mesh. The observed parameters were moisture content, ash content, fat content (AOAC 1999), protein content and crude fiber content (AOAC 1995), the levels of starch and amylose content (AOAC 1994).

Preparation of Composite Pellet Film and Analysis

The first step was cassava flour plasticizer using glycerol and water. Glycerol was mixed with the flour at doses of 30 and 40% of the total weight, while water was added to increase the moisture content up to 25%. Plasticizer was conducted using kneader with a rotary speed of 52 rpm for 15 minutes at 90 ^{o}C. Granule as a yield of mixing was then crushed using crusher to obtain pellets of 6-8 mm. Thermoplastic cassava flour was mixed with LLDEP resin and stearic

acid to obtain composite pellet. The mixing ratio of cassava flour: LLDEP resin was 2:8 and 3:7. PFAD doses were 5 and 7% of the total weight of LLDEP resin. LLDEP resin used in this research was LLDEP UF1810 mixed with LLDPE UI2420 with ratio 1:1. Compounding was conducted at 190^{o}C with a speed of 52 rpm to obtain homogenous compound. Granule compound was then crushed to obtain 6-8 mm size. The moisture content of the composite pellet was then analyzed (ISO 787-2, 1995). Composite pellet was then dried using hopper dryer at 110^{o}C to obtain moisture content less than 0.3%. After that, the MFI (ASTM D-1238, 1991) and specific gravity were analyzed.

Dried composite pellet was then proceed using blowing film to obtain film tube with a screw speed of 800 rpm and the temperature of the fourth extruder zone was 150^{o}C. The mechanical characteristics of the resulting film were analyzed. The observed parameters were tensile strength, elongation value, film, seal strength (ASTM D-882, 1991), FTIR, SEM (ASTM E-2015, 1991), and color (*yellowness* and *opacity*) (ASTM E-313, 1991).

Experimental Design

The experimental design used in this research was a complete randomized design with 3 experimental factors and 2 repetitions. Factors used in the research were the ratio of cassava flour and LLDPE which consisted of two treatments i.e. 3:7 and 4:6, glycerol doses at 30 and 40% of the total weight of cassava flour, and PFAD doses at 5 and 7% of the total weight of LLDPE.

RESULTS AND DISCUSSIONS

The Characteristic of Cassava Flour

The characteristic of cassava flour was conducted after the size of the material was reduced up to 100 mesh. The result of the analysis is shown in Table 1.

Table 1: The Characteristic Result of Cassava Flour Sized of 100 Mesh

Parameter	Value
Moisture content (% bb)	15.87
Ash content (% bb)	1.48
Protein content (% bb)	2.83
Fat content (% bb)	0.11
Crude fiber content (% bb)	0.23
Starch content (% bb)	78.53
Amylose ratio (% weight of starch)	27.07

Table 1 shows that that the moisture content of cassava flour was relatively high that vulnerable for long storage but essential for plasticizer process and producing composite. The content of fat and protein were appropriate for producing composite. Lee (2009) stated that material with lower content of protein and fat was good for thermoplastic process. In addition, Corradini *et al.* (2007) stated that fiber give better quality of mechanical characteristic of the resulting composite plastic. However, the crude fiber content of the plastic resulted in this research was low. The amylose content of cassava flour is relatively high which produced stronger film (Thomas and Atwell, 1999).

Characteristic of Palm Fatty Acid Distillate

PFAD used in this research contained fat acid content as shown in Table 2. The fat acid content was dominated by palmitic acid and oleic acid with more than 60% of the total weight of PFAD.

Table 2: The Analysis Result of PFAD

Parameter	Value (% b/b)
Capricacid	0.02
Lauricacid	0.09
Tridecanoicacid	0.92
Pentadecanoicacid	0.04
Palmiticacid	36.78
Palmitoleicacid	0.11
Heptadecanoicacid	0.07
Stearicacid	3.43
Oleicacid	27.54
Linoleicacid	7.72
Arachidonicacid	0.3
cis-11-eicosenoicacid	0.09
Linoleicacid	0.26
Lignocericacid	0.08
Total(%ofPFAD)	77.45

Physical and Thermal Properties of Composite Plastic

Moisture content was not influenced by the composition of the composite. However, the MFI and specific gravity were influenced by the composition of the material. The MFI decreased along the increasing amount of cassava flour. This decreasing was due to the flowing properties of thermoplastic cassava flour was lower than LLDPE resin. Meanwhile, the addition of glycerol and PFAD increased the value of the MFI composite. Glycerol as plasticizer increased the elasticity of cassava flour so the addition of glycerol caused the flour easier to flow. Characteristic of dispersing agent and lubricating agent in PFAD as a mixture of fatty acid increased the flowing capacity of the composite.

The physical and thermal characteristic of LLDPE/cassava a lour is shown in Table 3.

Table 3: Composite Characteristic

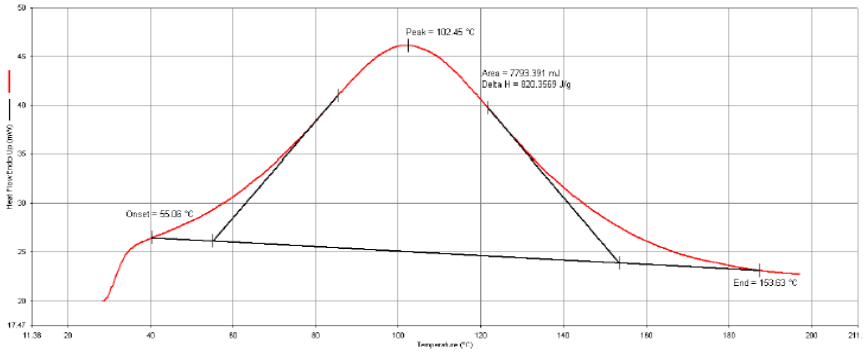
Formulation	Moisture Content(%)	MFI(g/10 Minute)	Specific Gravity(g/cm^3)
T30R70G30PFAD5	1.215a	4.420d	0.924a
T30R70G30PFAD7	1.096a	5.034f	0.917a
T30R70G40PFAD5	2.356a	5.451g	0.964b
T30R70G40PFAD7	2.138a	5.778h	0.942b
T40R60G30PFAD5	2.523a	3.424a	0.922a
T40R60G30PFAD7	2.567a	4.148b	0.926a
T40R60G40PFAD5	3.081a	4.367c	0.961b
T40R60G40PFAD7	2.277a	4.689e	0.955b

Note: the same letter in the same column shows no significant different a ta level of confidence 95%

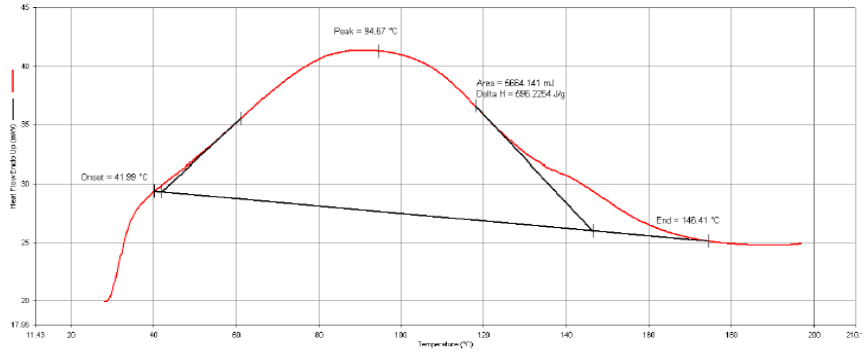
The specific gravity of the composite was influenced by the amount of cassava flour and the doses of glycerol. The increasing amount of cassava flour and glycerol increased the specific gravity of the composite. This was due to the specific gravity of cassava flour and glycerol was higher than LLDPE resin.

Figure 1 shows that the melting point of cassava flour was around 153^{o}C and decreased to 146^{o}C with a decrease of enthalpy changes around 225 J/g after plasticizer process. The addition of glycerol as plasticizer caused intermolecular force among polymer chains that could decrease the melting point and enthalpy changes (McHugh and Krochta, 1994).

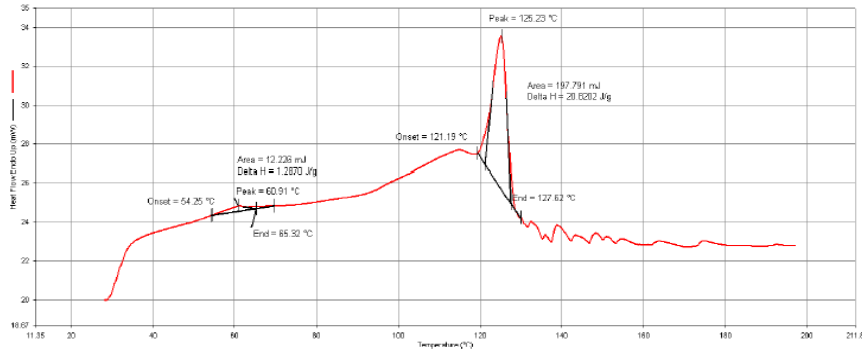
In this research, the melting point of the composite was around 127^{o}C with enthalpy changes was 20 J/g lower than thermoplastic cassava flour. A mixing with LLDPE which had lower melting point than cassava flour was the reason of the decreasing.



Cassava flour



Thermoplastic cassava flour



Cassava flour-LLDEP composite

Figure 1: DSC Chart

Morphology of Composite Film

The surface morphology of LLDPE/cassava flour composite film is shown in Fig. 2. It can be seen that the thermoplastic cassava flour was spread evenly at LDPE matrix. It shows that PFAD roles as compatibilizer during the mixing process of thermoplastic cassava flour and LLDPE. The film surface resulted from 7% of PFAD was smoother than the used of 5% of PFAD. PFAD could improve the interface interaction and adhesion force on thermoplastic cassava flour mixed LLDPE as shown by other compatibilizers used by various researchers. Some of researches were maleic anhydride grafted polyethylene (Waryat *et al.,* 2013), vinyltrimethoxy silane (Prachayakorn et al., 2010), anhydride grafted polypropylene (Mengeloglu and Karakus, 2008), and dibutyl maleate grafted polyethylene (Wang *et al*., 2002).

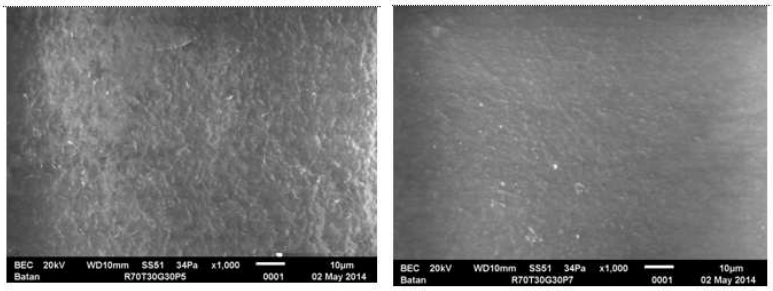


Figure 2: Surface Morphology of LLDPE/Cassava Flour Composite Film

FTIR Spectrum

FTIR spectra of cassava flour and cassava flour/LLDPE composite are shown in Fig. 2. It can be seen that the FTIR spectra of the composite (wave number around 3000-3750/cm) shows many peaks of primary, secondary and tertiary alcohol with strong and well-strong bonds. It also shows well-strong bonding between carboxylate acid groups. Meanwhile, cassava flour only shows one peak of carboxylate acid and alcoholic group. From the various peaks of carboxylate acid groups found in the composite, it can be assumed that the binding of fatty acid content of PFAD at LLDPE chain didn't occur at the carboxylic group but occurred at the carbon chain. Therefore, it was expected that fatty acid bounded by LLDPE would have carboxylic group with hydrophilic characteristic. This group was the reason why compatibilized LLDPE could be well mixed with cassava flour which has hydrophilic characteristic.

The FTIR spectra of composite film showed an increase of peak at wave number around 1000-1200/cm which indicated a presence of ester groups. The ester groups was presumably formed between the carboxylic group of fatty acid and alcohol groups of starch and fiber contained in thermoplastic cassava flour. This assumption relied on the condition that peak of carboxylic acid and alcohol group decreased (wave number around 3000/cm).

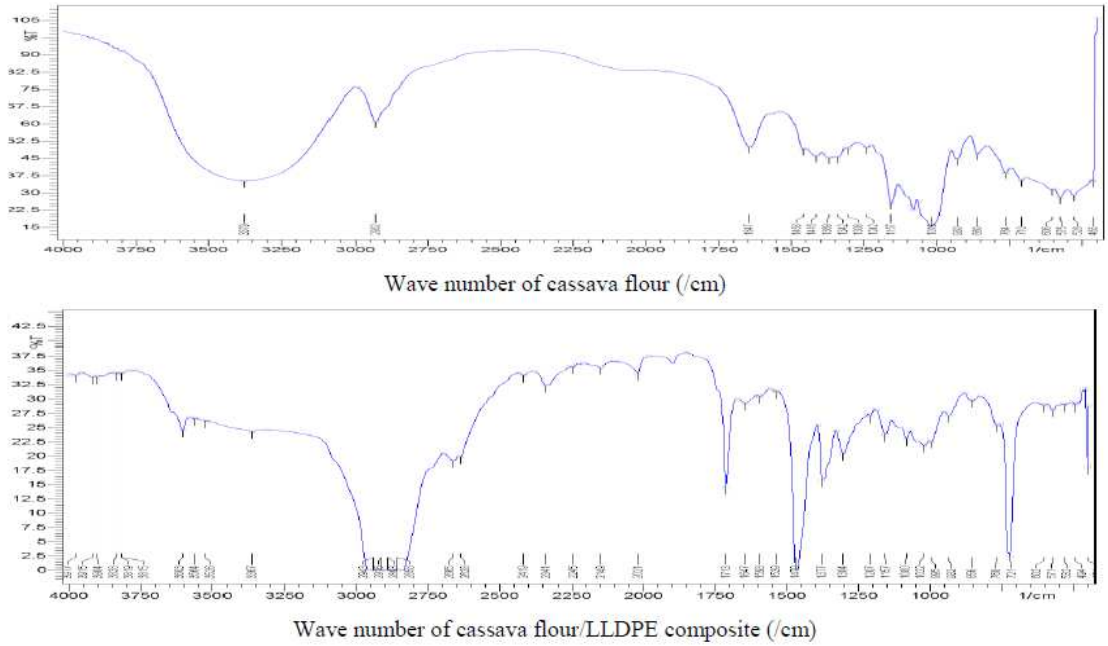


Figure 3: FTIR Spectra of Cassava Flour and Cassava Flour/LLDPE Composite

Mechanical Properties

The mechanical properties of the composite film are shown in Table 4

Table 4: The Result of Tensile Strength and Elongation of Composite Plastic Film

Treatment			Machine Direction		Transverse Direction		Heat Sealing	
Ratio LLDPE : Flour	Glycerol (%)	PFAD (%)	Tensile Strength (MPa)	Elongation (%)	Tensile Strength (MPa)	Elongation (%)	Tensile Strength (MPa)	Elongation (%)
7:3	30	5	3.71	396.18	3.06	126.29	3.11	137.06
		7	3.63	290.83	2.70	87.67	2.89	111.61
	40	5	3.18	217.23	2.65	76.73	2.51	81.50
		7	2.75	178.79	1.21	45.89	1.85	70.66
6:4	30	5	3.30	173.83	1.74	32.40	-	-
		7	3.73	111.49	2.01	29.09	-	-
	40	5	3.93	111.01	2.37	25.28	-	-
		7	4.94	97.69	2.54	21.90	-	-
LLDPE			34.85	891.44	24.73	890.44	13.34	419.57

Note: film was not tested because film couldn't be sealed with heat

The result showed that composite plastic film had lower magnitude of tensile strength and elongation compared to LLDPE film at any orientation. Overall, film with mixing ratio of 7:3 had better mechanical properties than mixing ratio of 6:4. Higher amount of starch contained in mixing ratio of 6:4 gave negative impact to the mechanical properties. Christianty (2009) stated that the higher amount of starch used in the process of composite plastic would reduce the mechanical properties. Variant analysis showed that mixing ratio significantly influenced the tensile strength and elongation at machine direction and transverse direction orientation.

The result of tensile strength test under machine direction orientation showed that mixing ratio with 6:4 was higher than 7:3. Film with mixing ratio 6:4 had thicker size so that the film was harder. However, this ratio created more fragile film which indicated by lower elongation magnitude compared to film with mixing ratio 7:3. Film testing at transverse direction orientation showed that the tensile strength and elongation was smaller than those resulted from machine direction orientation. According to the mixing ratio, film resulted from ratio 7:3 had higher tensile strength and elongation compared to 6:4.

At the heat sealing orientation, tensile strength test was only conducted upon mixing ratio of 7:3. Film produced from mixing ratio 6: 4 cannot be tested for tensile strength and elongation orientation because of the size of heat sealing film is too thick so that the film is difficult to be sealed with heat.

The variant analysis showed that the doses of glycerol was significantly influenced the elongation magnitude at machine direction and transverse direction orientation. The increasing doses of glycerol reduced the elongation magnitude both in the mixing ratio of 7:3 and 6:4. Sun *et al.* (2008) stated that the magnitude of elongation will increase along the increasing concentration of plasticizer. However, this study found that the increasing doses of glycerol decreased the elongation magnitude. This was presumably due to less homogenous mixing process which resulted that plasticizer material was not evenly distributed onto composite plastic matrix that negatively impacted the elongation.

The variant analysis showed that the doses of PFAD significantly influenced the film's tensile strength and elongation at machine direction and transverse direction orientation. The increasing doses of PFAD reduced the film's

tensile strength and elongation. Pritchard (1998) stated that the tensile strength and elongation will increase along the increasing amount of compatibilizer. The increasing magnitude of film's tensile strength and elongation in this research was presumably occurred due to the increasing doses of PFAD. As explained before, this condition was due to the thicker size of film that made the film was harder.

Optical Characteristic of Composite Plastic Film

Yellowness index indicates the level of sample color from white to yellow. Opacity indicates the clarity of a sample ranging from clear to opaque or dark. The result of yellowness index and opacity of composite plastic film is shown in Table 5.

Table 5: The Result of Whiteness, Yellowness Index and Opacity of Composite Plastic Film

Treatment			Yellowness Index	Opacity
Ratio LLDPE: Flour	Glycerol (%)	PFAD (%)		
7:3	30	5	16.28	18.13
		7	16.66	20.24
	40	5	17.12	21.83
		7	22.26	23.34
6:4	30	5	29.56	33.51
		7	30.32	44.97
	40	5	43.97	52.98
		7	44.14	59.26
LLDPE			14.025	15.435

The presence of flour attached to the matrix of LLDPE decreased the clarity of the film. Glycerol also affected the color of composite plastic film. At initial stage, the increasing doses of glycerol resulted dark color of thermoplastic starch product. This condition also occurred at the resulted composite plastic film. Another material that affected the color particularly the yellow color of composite plastic film was PFAD. This result was indicated by the increasing level of yellow color along the increasing doses of PFAD as shown in Table 8. According to the variant analysis, the treatment of mixing ratio, doses of glycerol and doses of PFAD significantly affected the film's whiteness, yellowness index and opacity.

CONCLUSIONS

The best formulation to produce composite plastic bag was LLDPE resin and cassava flour with mixing ratio of 7:3, glycerol 30% and PFAD 5%. The resulted composite and film were suitable for blowing film processing. The specific gravity and MFI value of the composite plastic film were 0.916 and 4.45 g/10 minutes, respectively. The highest tensile strength and elongation occurred at machine direction orientation with a value of 3.71 MPa and 396.18%, respectively. Meanwhile, at transverse direction orientation, the tensile strength was 3.06 MPa and the elongation value was 126.29%. At heat sealing orientation, the tensile strength was 3.11 MPa. The yellowness index and opacity of composite plastic film were 16.28 and 18.13, respectively.

SUGGESTIONS

A further research which studies a technique to produce clearer composite plastic film should be conducted. Blowing film technique should be more explored to produce composite film with higher amount of cassava flour.

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