

# DEVELOPMENT OF BIODEGRADABLE PELLETS FROM ECUADORIAN STARCH AND A RICE SUB-PRODUCT

Sandra Acosta-Davila, PhD<sup>1\*</sup>, C. Moreira, PhD<sup>2</sup>, J. Amaya-Pinos, PhD<sup>3</sup>, and, Augusta Jiménez-Sánchez, MSc.<sup>1</sup>

<sup>1</sup>Faculty of Chemical Engineering, University of Guayaquil, Cda. Universitaria Salvador Allende Malecón del Salado entre Av. Delta y Av. Kennedy, Guayaquil 090510, Ecuador, [sandra.acostad@ug.edu.ec](mailto:sandra.acostad@ug.edu.ec); [dolores.jimenezs@ug.edu.ec](mailto:dolores.jimenezs@ug.edu.ec)

<sup>2</sup>ESPOL Polytechnic University, Facultad de Ingeniería en Mecánica y Ciencias de la Producción, Campus Gustavo Galindo Km. 30.5 vía Perimetral Guayaquil, Ecuador, [cmoreir@espol.edu.ec](mailto:cmoreir@espol.edu.ec)

<sup>3</sup> Salesian Polytechnic University, UPS. Carrera de Ingeniería Ambiental – Calle Vieja 12-30 y Elia Liut – Cuenca, Ecuador; [jamaya@ups.edu.ec](mailto:jamaya@ups.edu.ec)

*Abstract— The single-use plastic items derived from oil are considered an environmental burden, worldwide. Efforts have been exerted to replace oil-based plastic with biodegradable polymers. However, it is unsustainable depending on a single material, so developing countries with plenty of biomass should develop their solutions. This study proposes developing biodegradable polymer pellets based on cassava or plantain starch using rice bran as a source of fiber, different palletization temperatures, and glycerol as a plasticizer. A degradation test followed the pellet manufacture to decide the most promising formula developed. Cassava starch seems to be a better polysaccharide when working with rice bran, and 50 °C palletization temperature.*

*Keywords—cassava starch, plantain starch, rice bran, pellet, biodegradable polymer*

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**Abstract**– *The single-use plastic items derived from oil are considered an environmental burden, worldwide. Efforts have been exerted to replace oil-based plastic with biodegradable polymers. However, it is unsustainable depending on a single material, so developing countries with plenty of biomass should develop their solutions. This study proposes developing biodegradable polymer pellets based on cassava or plantain starch using rice bran as a source of fiber, different palletization temperatures, and glycerol as a plasticizer. A degradation test followed the pellet manufacture to decide the most promising formula developed. Cassava starch seems to be a better polysaccharide when working with rice bran, and 50 °C palletization temperature.*

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## I. INTRODUCTION

Biodegradable polymers (BP) could be substitutes for fossil polymers (FP) which are considered an environmental burden. On the other hand, the residues (sub-products) generated by agroindustry processing usually require appropriate disposal due to their content of minerals, starch, sugar, and proteins [1]. Ecuador possesses a wide variety of sub-products; for example, Guayas and Los Rios provinces, where it is cultivated 95% of the country's rice production, release significant amounts of rice bran (a sub-product of the rice milling), rice husks, etc. [2].

Starch is an example of a biodegradable polymer (BP); however, its characteristics vary depending on where it comes from [3] notably, during plasticization, the semi-crystalline starch changes to an amorphous material because of the partial substitution of the amylopectin and amylose macromolecule hydrogen bonds with newly formed molecules.

The result of this process is known as thermoplastic starch, which disadvantage is being hydrophilic due to the presence of hydroxylic groups (OH-) bonded to the glycosidic units of this polysaccharide [3]. Another important disadvantage of BP is its mechanical properties; many studies incorporate natural fibers to reinforce them. Lately, the use of natural fibers with

BP has been rising. Nowadays, fiber is an ingredient in most thermoplastic formulas, usually added in a range of 5 – 30% (w/w). The fiber addition improves the plastic matrix's characteristics, i.e., better thermal stability, less water absorption, and prevents starch recrystallization after processing [4].

The plastic industry commercializes BP or FP in its pellet form, called resin. Pellets are sphere-ovoid in shape, with sizes ranging from 1 to 5 mm; their color depends on the chemical composition and the plastic fate. In general, thermoplastics' structure is linear or poorly branched; as a result, they can flow at pressures over their fusion temperatures. Also, they can be molded or remolded using heat as often as required, easing their recycling [5].

This study aims to generate a BP pellet using local raw materials, i.e., casava starch (*Manihot sculenta*), green plantain starch (*Musa paradisiaca*), and rice bran. Each of the selected materials possesses appealing characteristics. For example, cassava starch is a known stabilizer in the food industry due to its content of amylose and amylopectin, jellification temperature, texture and gel consistency, viscosity, and thermal properties [6] [7]. Plantain starch is a biopolymer known for generating films with sound mechanical properties and water vapor barrier (WVP). However, it is recommended the addition of nanocomponents, such as fiber, in its film formulae since its lipids, ashes, and protein content could affect optical and physical-chemical properties in the film manufacturing process [8]. Finally, rice bran is a good fiber source, improving the film's mechanical properties, thermal stability, functional properties, and WVP [9] [10] [11].

## II. MATERIALS AND METHODS

The following raw materials were used,

- 1) Plantain starch (AP) 9.7% moisture (RH), 20.2% amylose, 79.8% amylopectin, from a farmers market located in Manabí province.
- 2) Local “La Pradera” cassava starch (AY), 12.3 RH, 31.2% amylose, 68.2% amylopectin.
- 3) Rice bran (P) 9% RH, 3.8% amylose, 96.2% amylopectin, from a local mill located in Samborondón, Guayas.

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- 4) Amylose and amylopectin kit assay (Amylose/Amylopectin, Assay Procedure, K-AMYL 04/06, Megazyme, Wicklow, Ireland)
- 5) Glycerol (G) from Merck &Co US.

### Pellet elaboration

As described in Table 1, six formulas were developed using the available raw materials, i.e., AP, AY, P, and G. Therefore, 100 g of AY and P and 30 g of G were weighed using the analytical balance (BOECO BWL61, US) for formulas 1, 2, and 3; cassava starch (AY) was used, and for formulas 4, 5, and 6 plantain starch (AP) was used. The homogeneous mass of starch and rice bran with glycerol were mixed at room temperature (25 °C) until a homogeneous mass (mix); then the procedure followed fed the mix in a laboratory-size vertical matrix pellet machine at room temperature for molding (feedstock); the length depended on the matrix speed. The mix was subjected to this treatment two times; then the feedstock was fed, varying the temperature inside the pelletizer to 50, 60, 70, and 80 °C. After the treatment, the pellets were taken to a convection oven (CBX 4T/6040 Tagliavini, Italy) and maintained at 60 °C for an hour. The manufactured pellets were stored inside desiccators containing a saturated solution of magnesium nitrate hexahydrate.

TABLE 1  
formulas developed for biodegradable pellets.

Formula	Ingredients	Temperature °C			
		50	60	70	80
1	AY(75%) + P(25%)	50	60	70	80
2	AY(50%) + P(50%)	50	60	70	80
3	AY(25%) + P(75%)	50	60	70	80
4	AP(75%) + P(25%)	50	60	70	80
5	AP(50%) + P(50%)	50	60	70	80
6	AP(25%) + P(75%)	50	60	70	80

### Pellet Characterization

#### Yield

The pellets were collected using a 100-micrometer sieve and weighed using the analytical balance (BOECO BWL61, US). The yield was calculated using equation 1.

$$\% Y = \frac{\text{mass pellets}}{\text{mass formula mix}} \times 100 \quad (1)$$

#### Moisture

The pellets' moisture content was determined using a thermobalance (Ohaus MB120 Smartguide, US). Two grams of each formula were weighed and placed in the cited equipment for this effect. The reported results are the median of two determinations.

#### Statistical analysis

The method applied in this study's results was the analysis of variance (ANOVA) with 95% of confidence, coupled with Tukey analysis for multiple comparisons. The software used was Minitab ® 17.1.0

### Degradation rate

The degradation rate was determined using the method described by [12]. Therefore, 2 g of the pellet was placed inside a 150 ml Pyrex beaker containing 100 ml of distilled water with a magnetic stirrer. The setup was then set on a heating stirring plate (HOTPLATE STIRRER BS – 4HC, US). The speed of the plate was set to 400 – 440 rpm without heating. The time taken for the pellet to disappear visually was registered.

## III. RESULTS AND DISCUSSION

The data corresponding to the initial characteristics of the pellet are described in Table 2; the yield obtained for each formulation after going through the pelletizer is shown in Table 3. When using AY and P, the results suggest that formula 2, with a temperature of 70 °C gave the highest yield, 90.08%, followed by a yield of 43.23% corresponding to formula 3, with a temperature of 50 °C. However, when using AP and P, the highest yield, 87.54%, was registered when using formula 5 with a temperature of 70 °C, followed by a yield of 46.62%, corresponding to formula 4 with a temperature of 70°C.

TABLE 2  
Data collected at the pelletizer, (F) feedstock introduced to the pelletizer, (PF) mass of pellets formed, (RPM) pelletizer speed.

FORMULA	T(°C)	F (G)	PF(G)	RPM
1	50	74,00	66,70	424
	60	100,90	97,20	414
	70	113,80	94,10	410
	80	116,20	72,10	414
2	50	96,70	90,00	405
	60	94,60	90,30	426
	70	121,40	117,10	420
	80	91,50	81,30	416
3	50	57,60	56,20	420
	60	113,80	108,10	416
	70	73,30	70,40	426
	80	97,50	94,40	435
4	50	82,30	81,50	424
	60	108,30	105,90	414
	70	73,80	60,60	410
	80	101,80	99,60	414
5	50	114,30	113,80	405
	60	95,30	94,30	426
	70	99,40	97,50	420
	80	102,10	100,90	416
6	50	98,90	98,40	420
	60	81,00	79,70	416
	70	111,10	109,20	426
	80	82,60	80,60	435

A direct relationship exists between the fiber content and the yield in both scenarios, i.e., AY and AP. The higher the fiber content of the formula, the higher the yield. The

observed results agree with [13] [14]. Especially, in the part regarding the behavior of organic natural fibers such as rice straw, which says, it presents linearity and hydrophilicity, which contribute to the formation of a solid bridge in the pellet structure, giving strength and resistance to the pellet. Importantly, the gelatinization temperature of cassava starch is in the range of 58.5 – 70 °C; therefore, this material shows better resistance and viscosity at temperatures over 50 °C [15]. Besides, [16] [17] concludes that plantain starch gelatinization occurs in the range of 77.7 – 80 °C, which agrees with the results of this study. Indeed, the effects of moisture and temperature are mentioned in [18] study, which concludes that they affect the formation of polymeric matrices. In this study, the moisture of the pellet was analyzed statistically, and the results are shown in Table 3. The only pellets presenting significant differences in their moisture content resulted from formula 1. The pellet with the lowest moisture content comes from formula 2 (4.06%). Conversely, the pellet with the highest moisture content results from formula 6 (7.48%). Seems that the behavior of the pellet varies depending on the fiber content, which is in agreement with the study of [19], who reported that when adding sugar cane bagasse to starch matrices decreases the pellet moisture absorption [19].

TABLE 3  
Pellet manufacture parameters: pelletizing temperature (T), yield (Y), Moisture (M), and degradation time (t).

FORMULA	T(°C)	Y (%)	M (%)	t (min)
1	50	51,31	5,63 (0,63)a	12,44
	60	74,77	4,67 (0,18)ab	18,00
	70	72,38	4,86 (0,21)b	7,34
	80	55,46	6,87 (0,16)b	6,35
2	50	69,23	4,06 (0,46)a	11,37
	60	69,46	5,77 (0,73)a	10,00
	70	90,08	6,17 (0,76)a	18,00
	80	62,54	5,09 (0,47)a	10,00
3	50	43,23	5,81 (0,35)a	18,00
	60	83,15	5,77 (0,50)a	18,00
	70	54,15	6,20 (0,15)a	16,00
	80	72,62	4,96 (0,07)a	18,00
4	50	62,69	7,12 (0,32)a	10,00
	60	81,46	5,49 (0,85)a	10,00
	70	46,62	6,40 (0,43)a	9,50
	80	76,62	6,66 (1,22)a	10,00
5	50	87,54	6,64 (0,10)a	16,00
	60	72,54	6,46 (0,33)a	15,00
	70	75,00	5,90 (0,69)a	15,00
	80	77,62	5,74 (0,23)a	18,00
6	50	75,69	6,56 (0,91)a	18,00
	60	61,31	5,93 (0,10)a	16,00
	70	84,00	7,48 (0,08)a	16,00
	80	62,00	5,57 (0,93)a	15,00

\*a, b, ab refers to significant differences among treatments

Also, the used starch affects the moisture content, which agrees with the study of [20] [21], who reported an increment in the content of plantain starch results in pellets with increased moisture [20] [21]. Overall, the pellet moisture of this study remains under 10% which is below the threshold given by [22], for having a pellet with good mechanical properties.

All the formulas were subjected to a degradation test. Formula 1 pellets are shown if Fig 1. They registered a speed of degradation of 12.44 minutes when testing pellets pelletized at 50 °C; the speed of degradation time of the pellet increased to 18 minutes when testing pellets pelletized at 60 °C, reaching a maximum value. For the rest of pelletizing temperatures, the speed of degradation time diminishes, as shown in Fig 2.

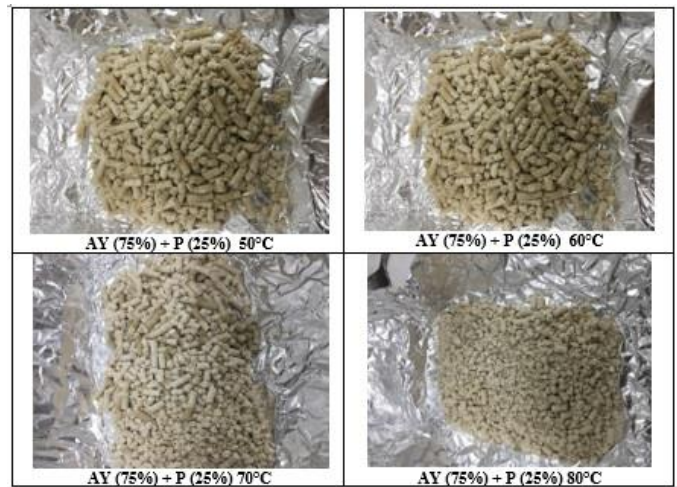


Fig. 1 Pellets manufactured under Formula 1 conditions.

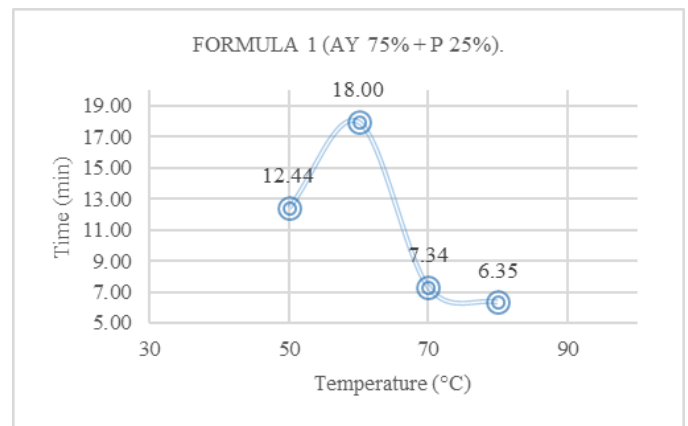


Fig. 2 Time of the speed of degradation for formula 1

Formula 2 pellets are shown in Fig 3. They registered a speed of degradation of 11.37 minutes when testing pellets pelletized at 50 °C; the speed of degradation time was reduced to 10 minutes when testing pellets pelletized at 60 °C. Then



the speed of degradation time increases to 18 minutes, reaching a maximum when testing pellets pelletized at 70 °C. Finally, the speed of degradation time comes back to 10 minutes when testing pellets pelletized at 80°C, as shown in Fig 4.

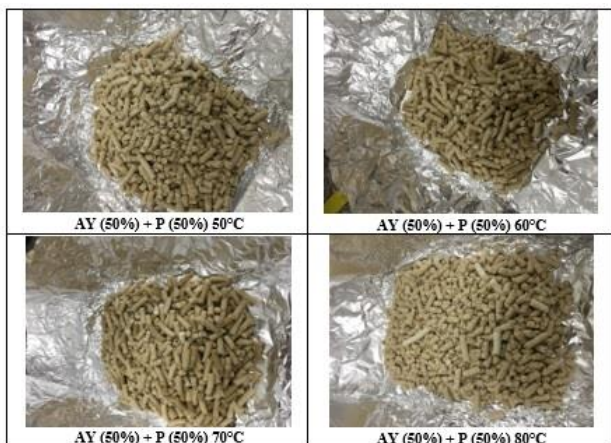


Fig. 3 Pellets manufactured under Formula 2 conditions.

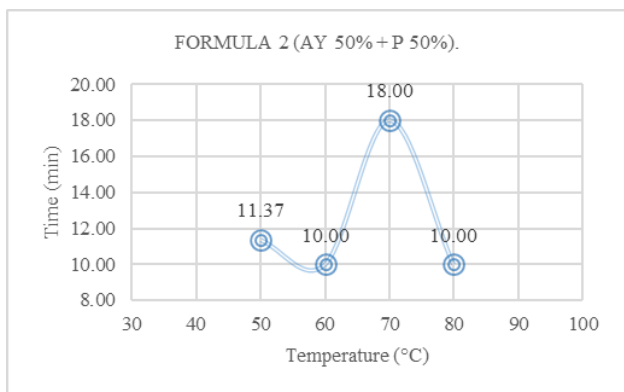


Fig. 4 Time of speed of degradation for formula 2

Formula 3 pellets are shown in Fig. 5. They registered a speed of degradation of 18 minutes when testing pellets pelletized at 50 °C; the speed of degradation time was maintained at 18 minutes when testing pellets pelletized at 60 °C. Then the speed of degradation time diminishes to 16 minutes, reaching a minimum when testing pellets pelletized at 70 °C. Finally, the speed of degradation time comes back to 18 minutes when testing pellets pelletized at 80°C, as shown in Fig 6.

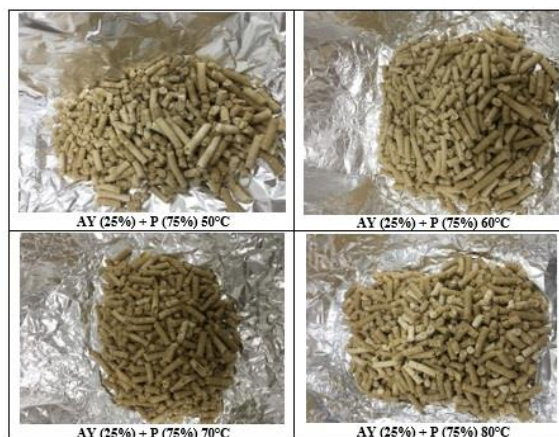


Fig. 5 Pellets manufactured under Formula 3 conditions.

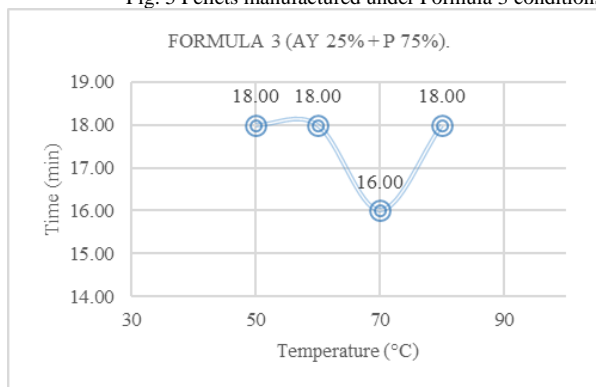


Fig. 6 Time of speed of degradation for formula 3

Formula 4 pellets are shown in Fig. 7. They registered a speed of degradation of 10 minutes when testing pellets pelletized at 50 °C; the speed of degradation time was maintained at 10 minutes when testing pellets pelletized at 60 °C. Then the speed of degradation time diminishes to 9.5 minutes, reaching a minimum when testing pellets pelletized at 70 °C. Finally, the speed of degradation time comes back to 10 minutes when testing pellets pelletized at 80°C, as shown in Fig 8.



Fig. 7 Pellets manufactured under Formula 4 conditions.

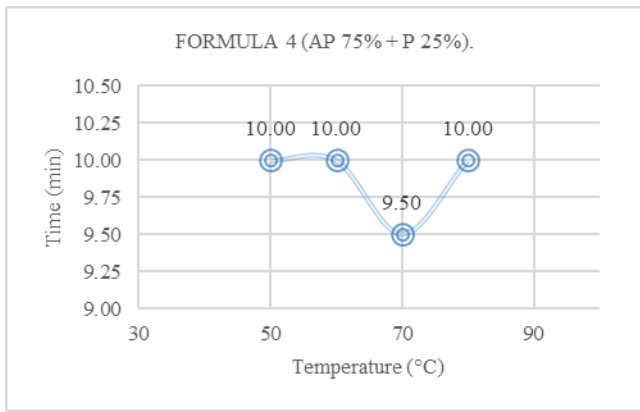


Fig. 8 Time of speed of degradation for formula 4

Formula 5 pellets are shown in Fig. 9. They registered a speed of degradation of 16 minutes when testing pellets extruded at 50 °C; the speed of degradation time was reduced to 10 minutes when testing pellets pelletized at 60 °C, a time that was kept when testing pellets pelletized at 70 °C. Finally, the speed of degradation time increases to 18 minutes when testing pellets pelletized at 80°C, as shown in Fig 10.



Fig. 9 Time of speed of degradation for formula 5.

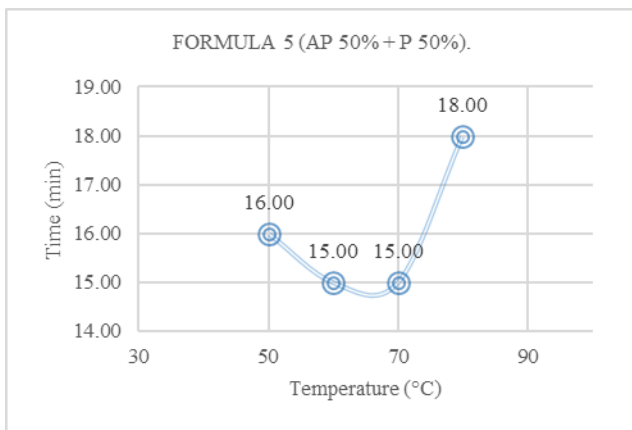


Fig. 10 Time of speed of degradation for formula 5

Formula 6 pellets are shown in Fig. 11. They registered a maximum speed of degradation of 18 minutes when testing pellets pelletized at 50 °C; the speed of degradation time was reduced to 16 minutes when testing pellets pelletized at 60 °C, a time that was kept when testing pellets pelletized at 70 °C. Finally, the speed of degradation time decreases to a minimum of 15 minutes when testing pellets pelletized at 80°C, as shown in Fig 12.

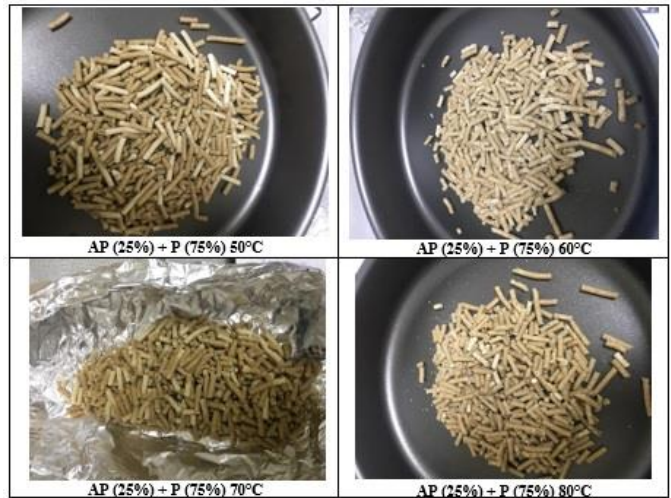


Fig. 11 Time of speed of degradation for formula 5.

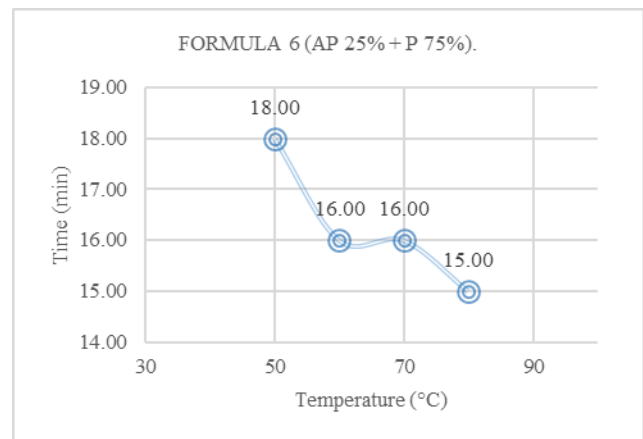


Fig. 12 Time of speed of degradation for formula 6

The time of the speed of degradation varies according to the characteristics of the pellet. The data has been summarized in Table 3. From it, we can conclude that formula 1 pelletized at 80 °C required the minimum amount of time to degrade, i.e., 6.35 minutes. Meanwhile, formulas 1, 2, 3, 5, and 6 pelletized at 60, 70, 80, 80, and 60 °C, respectively, share the maximum amount of time registered, 18 minutes. It can be inferred that pellets made of cassava starch at higher pelletizing temperatures and rice bran concentrations resulted in dryer pellets that could be degraded faster. Meanwhile, the pellets

made from plantain starch (AP) registered a higher time of speed of degradation when increasing the rice bran content. Therefore, the increment in the concentration of rice bran (rich in fiber) seems to slow the degradation. An observation that agrees with the study of [23], when working with sugar cane bagasse reported that the higher the content of this fiber (5, 10, 15%) in starch matrices, the improved mechanical properties, i.e., traction, strength, and stability, properties of the pellet [23] [24]. The behavior regarding the pelletizing temperature is different between cassava starch and plantain starch, which agrees with the study of [25], who conclude that the pellet strength and hardness depend on the polymeric component of its formulation and the pelletizing temperature. Noticeable, the study of [26], concludes traction stress of matrices vary directly proportionally with its moisture content, similar behavior to the observed in this study.

#### IV. CONCLUSIONS

Formulas 2 and 5, composed of equal amounts of starch and rice bran, subject to a temperature of 70°C in the pelletizer, are the more efficient, therefore, possess the best conditions for pellet manufacturing, i.e., starch gelatinization temperature and fiber content. It is inferred that adding rice bran reduces the degradation of the manufactured pellet. The maximum time of speed of degradation rate was 18 minutes for both of the used starches, i.e., plantain and cassava. However, the more stable pellet resulted from formula 3, i.e., 25% cassava starch and 75% rice bran.

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