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Valorization of Banana Peels to Obtain a Bioplastic from Acetylated Starch

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Abstract

Global solid waste generation will increase by 70% by 2050. Within this projection, organic waste will constitute 44% of the total. Among these organic wastes are banana peels, which can be valorized to be transformed into bioplastic. The objective of this research is to develop a bioplastic film on a laboratory scale, from the starch extracted from banana peels using the acetylation method. To evaluate the quality of the bioplastic, six tests were carried out modifying the residence time, the ripening of the banana peels, the pH and the concentration of glycerin. To evaluate the quality of the bioplastic obtained, biodegradability tests, tensile tests and Fourier transform infrared spectroscopy (FTIR) were carried out. The results obtained revealed an 80% efficiency in the conversion of banana peels to bioplastic. The percentage of biodegradability reached a maximum percentage of 47%, highlighting the influence of the amount of glycerin used in the process. In terms of tensile strength, values of 3.169 MPa were recorded, while the Young's modulus was 0.486 MPa. Fourier Transform Infrared Spectroscopy (FT-IR) analysis indicated the presence of functional groups characteristic of starch molecules such as Carbon - Hydrogen (CH), Hydroxyl Radical (OH), Carbon Monoxide (CO) characteristic of starch molecules.

Keywords
Banana peels, Acetylation, Bioplastics, Biodegradability, FTIR

1. Introduction
One of the most important challenges of the 21st century, environmental pollution, is being faced worldwide. In that sense, we think about the amount of waste produced by industries at a global level which are not properly managed, specifically, the plastic waste generated. Today, more than 300 types of plastic are produced (Chen et al. 2021). Each year, 11.2 billion tons of solid waste are generated, with organic decomposition contributing to 5% of greenhouse gas emissions (United Nation Organization, 2023). Between 2000 and 2019, global plastic waste production doubled to 353 million tons, highlighting that only 9% of this waste is recycled, according to the Organization for Economic Cooperation and Development (2022), intensifying the environmental problem. Despite the excellent characteristics of plastic in terms of its usefulness, a major disadvantage is that it is not biodegradable because it remains in the environment for many years, causing problems when deposited in landfills and the sea. This is due to the Biological Oxygen Demand that causes negative effects on biodiversity (Vinodh et al. 2021). However, the problem is not only in the plastic as waste after its useful life ends and they are discarded, but in the production of this compound since 95-99% of the synthetic plastic in the world is made from non-renewable resources that come from the petrochemical industry (Shaikh et al. 2021) and using fossil resources in industrial processes increases CO2 in the atmosphere (European Bioplastics 2023).

Thus, due to the aforementioned problems and the multiple research that continues to be carried out, bioplastics are born in the world. "Bioplastics are plastics derived from renewable biomass sources, such as vegetable fats and oils, corn starch, straw, wood chips, food waste, agricultural by-products, also from used plastic bottles and other packaging using microorganisms" (Beevi et al. 2020). The main characteristic of bioplastics is that they are biodegradable being a great advantage compared to synthetic plastics and another of their characteristics is that they have the ability to be compostable (Veena and Rani 2022). For the production of bioplastics, polysaccharides, proteins, polyesters and lipids, which are present in vegetables and fruits, are used as raw materials (Chandrasekar et al. 2023). Thus, since polysaccharides are one of the macromolecules found in the greatest quantity in fauna and flora in the form of starch, they are an excellent option (Shafqat et al. 2021).

Other authors have elaborated bioplastic films using various organic wastes; for example, in one study, white potato peel starch was synthesized with glycerol and it was concluded that increasing the amount of glycerol from 1.5 to 2.5 ml increases the tensile strength up to a value of 0.56 ± 0.05 MPa (Imoisili and Jen 2023). In another study a bioplastic based on cassava starch which was modified by sialylation method with 3-aminopropyl trimethoxy silane was produced, in this the tensile strength they obtained was 9.29 MPa (Yang et al. 2023). Kharb and Sahara fabricated a bioplastic material from cucumber peel and corn starch which was synthesized with glycerol and sorbitol; and in the FTIR test they found OH functional groups characteristic of starch and CH due to the interaction between starch and acetic acid (Kharb and Saharan 2023).

Banana peels will be used as raw material because world banana production is growing at a compound annual rate of 0.80% during the forecast period from 2023 to 2028, with the largest market being Asia and the Pacific (Mordor Intelligence Research 2023). The total production in 2021 was approximately 163 million tons, and since the weight of the peel is about 35% of the total weight of bananas (Karne et al. 2023), there would be an amount of 57 million tons of banana peels per year. In addition, this residue has a large source of starch in percentage approaching 18.5% but this will depend on the level of maturity, since the higher the maturity the amount of starch decreases (Kader and Wan 2017).

Bioplastics present differences in their attributes and properties depending on the manufacturing process and production conditions. Among the characteristics sought are chemical composition, evaluating the presence of functional groups; mechanical strength, measured by tensile tests to assess its ability to withstand stress, strength, and elasticity; biodegradability, highlighting the material's ability to decompose naturally; and moisture resistance (Chocano 2019).In this study, the acetylation method was used to perform the chemical modification of native starch, which consists of the formation of an esterification reaction of starch with acetic acid by which the hydroxyl (OH) group of starch is replaced by the acetyl group of acetic acid (Cardona 2019). This is done since the modified starch presents better physical characteristics such as greater resistance and biodegradability taking into account that it will later be the basis for the formation of the bioplastic film (Kumari and Sit 2023).

1.1 Objectives

The objective of the present research is to obtain a bioplastic at a laboratory scale from banana peels through the acetylation process. Six tests will be carried out to evaluate biodegradability, tensile test, and FTIR analysis. The research problem question is: Can bioplastics be obtained from banana peels by the acetylation process?

Our main focus is to offer a valuable proposal focused on environmental preservation. We seek to give an additional value to waste not used in the food industry such as banana peels, thus presenting a different alternative to conventional plastic and contributing to the care of the planet.

2. Literature Review

Type 1: Comparison of the physical characteristics of conventional plastic with biodegradable plastic

The article entitled “Banana peel starch to biodegradable alternative products for commercial plastics” (Arjun et al. 2023) states that its object of study is to demonstrate that starch extracted from banana peels could be used to produce biodegradable plastics. In addition, degradation and elongation tests were carried out on biodegradable plastic and compared with those of conventional plastic. It is therefore concluded that the material produced can be used as packaging and in the manufacture of transport bags.Likewise, in “Bioplastic synthesis using banana peels and its characterization” (Noorjahan et al. 2022) a comparison is made of the properties of commercial plastic with biodegradable plastic based on banana peels. It also adds that the produced material should be considered a promising solution since its use can reduce the environmental pollution caused by commercial plastic.

Type 2: Development of different characterization tests for the bioplastic obtained

In the article "Bioplastic from fruit waste" (Oo et al. 2019), characterization tests are developed for bioplastic based on banana peels. Water absorption, biodegradability, swelling, solubility, and Fourier transform infrared spectroscopy (FTIR) tests were performed. The latter with the purpose of knowing the functional groups and discovering any chemical alteration after adding the different reagents used in the process of making the bioplastic film.

Also, in the article "Production of Biodegradable Plastic from Banana Peel" (Kadam and Datta 2020), the elongation test is performed on biodegradable plastic, which aims to determine the amount of tension that the material resists before failing and the elongation distance when failure occurs.In "Development of a Bioplastic from Banana Peel" (Alcivar et al. 2022) the procedure and standards to be followed to evaluate the characteristics of the bioplastic obtained are described in detail. In this article, characteristics such as water vapor permeability for which the ASTM E96-80 standard is used, biodegradability which is measured by the weight loss of the bioplastic over time and the tensile test for which the modified ASTM D-882 standard is used were evaluated.
In this segment, the analysis of two articles will be carried out, describing the motivation, contribution, process and results of each one:

Article: Utilization of Banana (Musa Paradisiaca) Peel as Bioplastic for Planting Bag Application (Huzaisham and Marsi 2020)

Motivation: The author points out that he has found multiple investigations where tapioca starch is produced, which when making a bioplastic film shows discouraging characteristics since it is a fragile bioplastic.
Contribution: The author proposes to make a bioplastic based on the starch of banana and tapioca peels that can then be used to manufacture planting bags.
Process: The process begins with the pretreatment of the banana peels, which consists first of removing the dust or dirt that is attached to the banana peels, then the peels are cut into a size in the range of 2 ± 0.5 cm. These are dried in the oven for 24 hours, then crushed, starch is extracted and the bioplastic is made.
Results: Characterization tests were carried out with the aim of determining the physical and mechanical properties of the final product; that is, the biodegradable plastic. A mechanical test was carried out to measure the tear resistance, the biodegradability test was carried out using the soil burial technique for which the ASTM D5988-18 standard was used.

Article: Biodegradable Packaging Made from Banana Peels (Lora et al. 2022)

Motivation: The author points out that the development of this project is due to the growing interest of several researchers around the world in bioplastics, due to the excessive production of plastic materials, their difficult elimination process and the impacts it causes on the environment. Contribution: The author proposes to use banana peels to make a bioplastic that is characterized by being favorable for both living beings and the entire planet.
Process: The process begins with washing the banana peels with drinking water to remove dirt. They are then weighed, cut and left to rest in a bowl with a mixture composed of 2% citric acid and 98% drinking water. Afterward, only the peels are liquefied and mixed with other additives, filtered, and molded.
Results: After the biodegradability test, it was determined that after 30 days there were no signs of the presence of microorganisms in the form of mold. In addition, when compared to sheets made from mango peels, banana sheets are less brittle.

3. Methods

To obtain the bioplastic based on banana peels, the acetylation method will be used, which is a chemical modification of starch. This acetylation process occurs through esterification, in which the acetyl groups replace the hydroxyl groups (OH) and bind to the starch molecule. The chemical modification of this starch extracted from banana takes place because the acetyl groups prevent the linear structure of amylose, causing the starch chains to regroup into an ordered structure after gelatinization and cooling (retrogradation). This esterification confers improved physicochemical properties to banana starch, such as hydrophobicity and thermoplastic (León et al. 2020). Starches can be altered by physical, chemical, or enzymatic methods. Chemical modification excels in improving properties such as molecular stability, shear strength, viscosity, and retrogradation. Common procedures include acid hydrolysis, cross-linking, starch grafting, dual modification, oxidation, and acetylation/esterification. The latter, esterification, involves replacing hydroxyl groups with acetyl groups in the starch chains, generating covalent bonds that increase hydrophobicity and give a firmer texture than native starch (Teixeira Garcia et al. 2020).

The following figure illustrates the starch acetylation reaction.

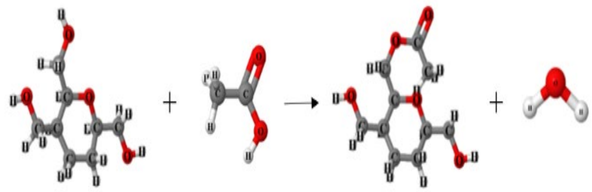

Figure 1. Starch Acetylation Reaction

In relation to the organic wastes frequently used in the production of bioplastics, potato, banana and corn stand out. In the present research, we chose to use starch extracted specifically from banana peels as raw material. This choice is based on the high levels of amylose and amylopectin, two molecules that make up starch. In the following, we will present the percentages of amylose and amylopectin and research carried out with other raw materials.

Table 1. Percentages of Amylose and Amylopectin

Starch	Amylose (%)	Amylopectin (%)
Banana	18	83
Rice	15	65
Potato	23	77

3.1 Materials and Equipment

The raw materials used for each test were five banana peels weighing approximately 300 grams, containing starch and cellulose, 400 ml of acetic acid, an inorganic compound with antioxidant and preservative properties, 3 ml of hydrochloric acid, a liquid reagent that will be used to achieve greater hydrolysis of the starch, 3 ml of sodium hydroxide or caustic soda, to regulate pH levels, 2 ml of glycerol, which will act as a plasticizer, providing elasticity and resistance to the bioplastic film, and 600 ml of distilled water (Arjun et al. 2023). In addition, utensils were used to perform the cutting, mixing and decanting operations: knife, gauze, beakers, filter paper, Petri dish, decanter, the use of each one is detailed in the procedure for obtaining the bioplastic from banana peels (Li et al. 2018).

3.2 Preparation of Raw Material and Starch Extraction

At the beginning, a sample of 300 g of banana peels is taken and then conditioned to homogeneously reduce the size down to 1 cm in order to facilitate processing. Next, the conditioned banana peels and 400 ml of an acetic acid solution (0.2 M) are fed to a stirred mixing tank (800 ml beaker) with a residence time of 45 minutes with the objective of slowing down the biodegradation period of the banana peel pieces (Kader and Wan 2017).

Subsequently, an 800 ml beaker was filled with distilled water, which was arranged on a Hotplate. The banana peels were introduced in the 800 ml beaker to start the heating operation, being subjected to a time of approximately 30 minutes to bring it to a boiling temperature of 100°C. After this stage, the beaker was removed from the hotplate and the shells were separated from the water by the decantation process. They were then covered with gauze and allowed to dry for a period of 30 minutes at room temperature, between 21 and 25°C. This procedure was carried out for the purpose of preparing the banana peels for the next phase of the experimental process (Kader and Wan 2017).

Finally, the aforementioned husks are subjected to a homogenization operation by grinding in a blender, resulting in the formation of a uniform paste called "slurry". Subsequently, this slurry is filtered using 100-micron mesh. The resulting filtrate is left to stand for 20 minutes in a 500 ml beaker, followed by the application of the decanting technique. The supernatant is carefully removed, culminating in obtaining 30 ml of starch (Kader and Wan 2017).

3.3 Production of Bioplastic Film

First, 30 ml of banana peel-derived starch were placed in a 100 ml beaker, followed by the addition of 5 ml of acetic acid. The solution was homogenized and allowed to rest for 1.5 days. Subsequently, 3 ml of hydrochloric acid were introduced, followed by continuous stirring using a stirring rod for a period of five minutes. Next, 3 ml of 15% glycerol were incorporated, and the mixture was stirred again. Once the mentioned composition was obtained, 3 ml of sodium hydroxide were added to neutralize the pH to a value of 7, indicative of a neutral level (Kader and Wan 2017).

The resulting mixture was subjected to additional stirring for 15 minutes, maintaining a constant temperature of 220°C in a Hot Plate equipped with magnetic stirring. Simultaneously, the temperature was monitored using a thermometer. Immediately afterwards, the mixtures obtained were poured into a glass mold and molded with a spatula. The resulting product was subjected to a polymerization process in an oven at a temperature of 130°C for a period of 1 hour. After a rest period of two days, the bioplastic film formed on the surface of the material was removed. The final bioplastic, with a thickness of 1.5 - 2 mm, was successfully prepared following the steps described in the procedure (Kader and Wan 2017). The aforementioned husks are subjected to a homogenization operation by grinding in a blender, resulting in the formation of a uniform paste called "slurry". Subsequently, this slurry is filtered using 100-micron mesh. The resulting filtrate is left to stand for 20 minutes in a 500 ml beaker, followed by the application of the decanting technique. The supernatant is carefully removed, culminating in obtaining 30 ml of starch (Kader and Wan 2017).

4. Data Collection

With the purpose of carrying out a more exhaustive experimental analysis, three tests of bioplastic films were initially produced, followed by the performance of three more tests, with the aim of expanding the information for the characterization tests, thus providing a more comprehensive investigation. solid for the evaluation of the properties of the bioplastics obtained.

In the first trial, 30 ml of starch was obtained, in the second trial 19.56 ml was obtained and in the third 26.37 ml. The tests were carried out with the same number of banana peels, that is, five, but with different ripening characteristics. To guarantee that what was obtained was starch, a test was carried out using a little sample and a few drops of potassium iodine were added, which has a reddish color, but when poured into the supposed starch it should change color to one blackish (Kader and Wan 2017).In each test, the starch obtained is poured into a 40 ml beaker and acetic acid is added to produce the acetylation reaction. The amount of acetic acid poured in the first test and third test is 5ml and in the second the starch was mixed with 2ml. A stirring rod was used to mix in all three cases (Kader and Wan 2017).

The glycerol was added to the acetylated starch. In the first trial, 3ml of glycerol was added, in the second trial 1ml and in the third 2ml. Glycerol is added with the aim of incorporating some characteristics of conventional plastic into the bioplastic; For example, it provides shine and also improves its physical characteristics since it reduces the rigidity of the bioplastic obtained so that it can be molded and has the appearance of a bioplastic (Kader and Wan 2017).

Table 2. Experimental Units

Test	Starch (ml)	Glycerin (ml)	Acetic acid (ml)
T1	30.00	3	5
T2	19.56	1	2
T3	26.37	2	5

Regarding the first difference mentioned, regarding the ripening period of the raw material, it is important to mention that by having a longer ripening period, the amount of starch extracted will be less than when banana peels with a shorter period of ripening is used. maturation. (approximate % ripening of banana peel). When carrying out the test with potassium iodine, it turned out that what was obtained was starch, since the reddish color of the aforementioned reagent transformed into a blackish one when it encountered the supposed starch at that moment (Kader and Wan 2017).

Concerning the difference in the amount of acetic acid that was mixed with the starch, results with some different characteristics were obtained. First, in the second test, when the acetylation reaction occurred and as it contained a smaller amount of acetic acid, there were some signs of degradation, which is due to the fact that acetic acid works as a preservative agent. Secondly, since the acetylation reaction involves the release of H2O molecules, in the second test a greater amount of liquid was found on the starch than in the first test. Consequently, the separation of the liquid from the acetylated starch was carried out, taking small samples to calculate the percentage of moisture by drying in an oven at 60°C for 30 minutes in each test (Kader and Wan 2017).

Thus, it was determined that the second was the most humid. (% starch degradation/oxidation; % moisture of acetylated starch). Formula applied to tests:

$$\%Humidity = (Weight\ 1 - Weight\ 2)/Weight\ 1 \times 100$$

Weight_1: Pre-test weight; Weight_2: Weight after test

Table 3. Humidity Percentages

Test	Initial weight	Final Weight	% Moisture
T1	3.6	2.3	36.11 %
T2	3.7	1.6	56.76 %
T3	3.8	2.6	31.57 %

5. Results and Discussion

Six bioplastic film trials were carried out using banana peels as raw material.

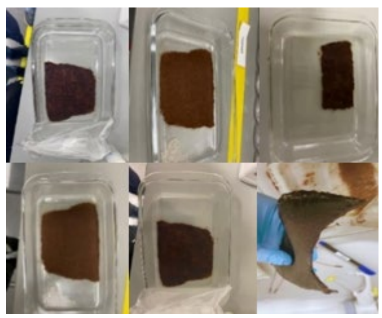

Figure 2. Six samples

5.1 Numerical Results

Biodegradability Test

The test was extended over 21 days in the Docimasia and Microbiology Laboratory. During this period, small samples were extracted from the containers and subjected to a washing process using deionized water, to separate the maximum amount of soil residues accumulated during burial. Subsequently, the samples were dried in an oven at 120°C for 30 minutes. The evaluation of biodegradation will be carried out by measuring the percentage of weight loss of the samples (C. M. Noorjahan et al 2022). Three weeks after carrying out the biodegradability test, the following results were obtained:

Table 4. Percent biodegradation over time

Sample	**1 days**	**7 days**		**14 days**		**21 days**		**Glycerin (ml)**
	Weight 1	**Weigh 2**	**Percent 1**	**Weigh 3**	**Percent 2**	**Weigh 4**	**Percent 3**	
S1	0.58	0.49	16%	0.4	31%	0.31	47%	3
S2	0.59	0.53	10%	0.47	20%	0.39	34%	1
S3	0.56	0.49	13%	0.43	23%	0.36	36%	2

It is verified that the amount of glycerin used exerts a significant influence, and the relationship between the two is directly proportional. This implies that the biodegradation capacity over time of the bioplastic film will increase with the increase in the amount of glycerin used. By comparing our results with those obtained in the article titled "Synthesis of biodegradable material from banana peel" (H. U. Karne et al. 2023), we confirmed our observations. In this article, sample 5, which contains a greater amount of glycerin (12 ml), achieved the maximum percentage of biodegradation, reaching 97%, in the same period of time.

5.2 Graphical Results

Tensile Test

In this test we use the bioplastic film with the best physical characteristics, in order to obtain optimal results for the resistance, elongation at break and Young's modulus tests. Precise measurements of the length, width and thickness of the bioplastic film were carried out, using measuring instruments such as a Vernier and a ruler to determine the longitudinal and lateral dimensions. The results revealed that the film had a length of 130 mm and a width of 45 mm. To measure the thickness accurately, a micrometer was used, obtaining a value of 0.64 mm. The results showed a tensile strength of 3.169 MPa, while the Young's modulus presented a value of 0.486 MPa. It is important to note that the test was carried out at a constant speed of 2 mm/min, using a caliper of specific dimensions, with a length of 54 mm by 45 mm (N. A. Azieyanti et al. 2020).

After analyzing the results of this research, where the tensile strength reached 3,169 MPa, this value was compared with that recorded in the research article "Development of a Bioplastic from Banana Peel" (M. G. Alcivar et al. 2022) indicated a resistance of 2,400 MPa. As a result of this comparison, it is concluded that the film developed in our research exhibits greater resistance. This is attributed to the use of a greater amount of starch, specifically 2.3 g in our film preparation, in contrast to the 1.25 g used in the other study. Regarding Young's modulus, the value obtained in our research was 0.486 MPa. When contrasted with the result of 1.88 MPa indicated in the article "Bioplastic from Banana Peel" (J. Vinodh et al. 2021), it is determined that the film made in our research presents a lower elasticity, indicating greater rigidity compared to the film derived from banana peel from the previous study.

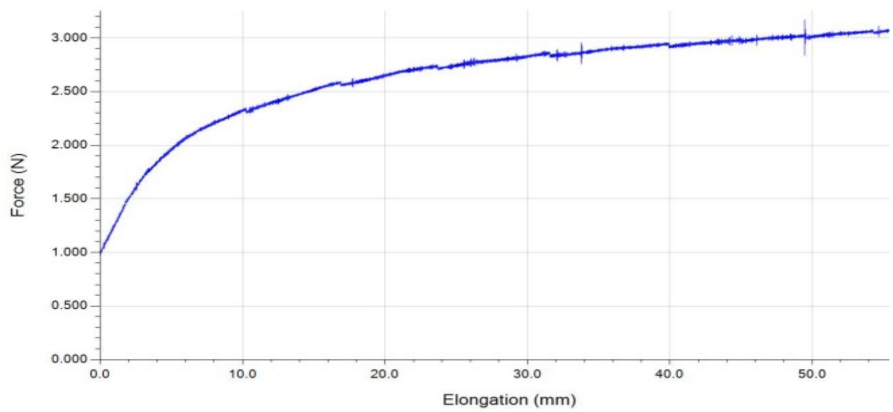

Figure 3. Test Chart on ASTM Standard Machine

Fourier Transform Infrared Spectroscopy (FTIR) Test

In the Figure 4. generated for the three samples of the bioplastic films, various peaks were obtained. The first peak observed between the wavelength 3100 to 3700 cm-1 in the three spectra corresponds to the OH group, in this sample it is observed that the peak decreases taking into account the amount of glycerin, since sample 1 has a lower stretching of OH, having a greater amount of glycerin, follows sample 3 with an amount of 2ml of glycerin and then sample 2 which, having a smaller amount of glycerin, has a greater stretch of OH. The peak located in the wavelength range between 2800 to 3000 cm-1 denotes the stretching of the CH functional group which is characteristic in the structure of starch. The peak with a wavelength between 1500 and 1650 cm-1 is due to the OH group of water and the difference in the spectra refers to the amount of water absorbed by each sample. The peak with a wavelength between 1000 and 1250 cm-1 corresponds to the stretching of the CO functional group (N. Fatimah Kader Sultan and W. Lutfi Wan Johari 2017)

This is consistent with the results presented in the article titled The Development of Banana Peel/Corn Starch Bioplastic Film (N. Fatimah Kader Sultan and W. Lutfi Wan Johari 2017). However, the differences are observed in the elongation of the peaks since in this article the peaks are more elongated, due to the fact that the starch concentration is higher compared to the present research carried out.

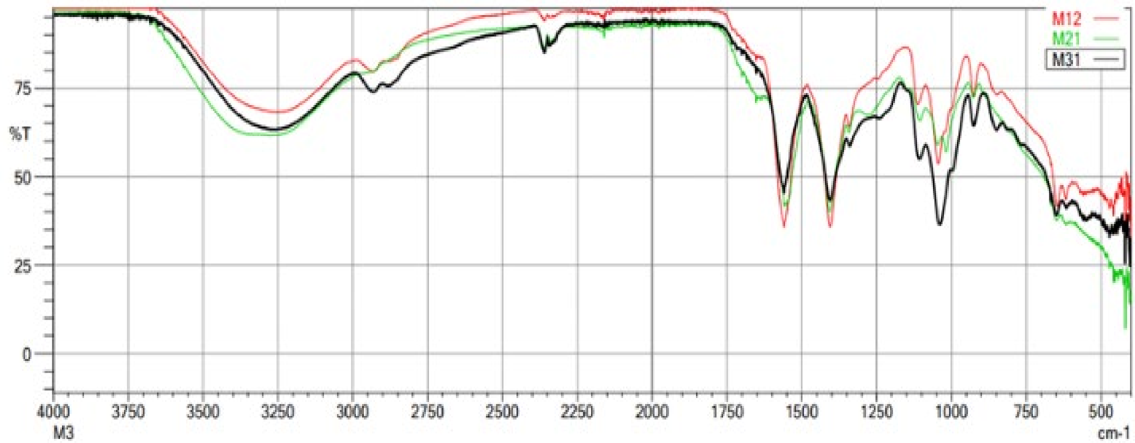

Figure 4. Results of the three FTIR samples

6. Conclusion

The bioplastic film made from the starch of banana peels by applying the acetylation method meets the quality criteria such as tensile strength and Young's modulus with a result of 3.169 MPa and 0.486 MPa respectively, with a percentage of biodegradation of 47%.It was proven that the quantities of glycerin and acetic acid used influence the physical characteristics of the bioplastic film such as tensile strength and percentage of biodegradation over time. This, because sample 1 had a higher percentage of biodegradation due to a higher glycerin content and lower acetic acid content and that the film obtained in this research project has a higher tensile resistance with a value of 3.169 MPa.

It was possible to verify that after 21 days of carrying out the biodegradation test using the soil burial method, the maximum percentage of biodegradation obtained was 47%; That is, the film obtained is biodegradable, which makes it a sustainable alternative. Furthermore, with respect to the Young's modulus obtained, the biodegradable film is less flexible than another film made in other research. In conclusion, the peaks identified at wavelengths from 3100 to 3700 cm-1 indicated variations in the amount of glycerin, affecting the stretching of the OH group. Likewise, the peaks in the ranges of 2800 to 3000 cm-1 and 1500 to 1650 cm-1 highlighted the stretching of the CH and OH functional groups of water, respectively. Although the results coincide with the reference article, the differences in the length and shape of the peaks point to differences in starch concentrations.

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