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INVESTIGATE THE PROPERTIES OF DIFFERENT IRRADIATED STARCH BIOPLASTIC FOR PACKAGING APPLICATION

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Abstract: Plastics have become the preferred materials for use due to their barrier properties and strength. This study aimed to determine the wettability, contact angle, and polymer chemical properties by comparing pure and mixed starch bioplastics with different plasticizers. Gamma radiation and electron beams are used in industrial applications. Gamma radiation is used because it is extreme in penetration, and Cobalt-60 is often used for sterilization. Most bioplastics with citric acid as the plasticizer produced a high contact angle and achieved hydrophobicity. Materials used in constant amounts are distilled water and glycerol. Others manipulate variables based on the presence of starch, such as corn, potato, rice, or a mixture of the two starches. The Samples were characterized using tensile and elongation tests, water contact angle, Fourier Transform Infrared (FTIR) analysis, topography using Atomic Force Microscopy (AFM), moisture content tests, and biodegradability tests. Potato and corn-rice bioplastics have low moisture contents. Meanwhile, the corn bioplastic degraded faster at 80.17%. Potato rice with citric acid lead tensile test with 4.095 MPa. However, potatoes with sorbitol lead with 34.57% for the elongation case. FTIR analysis identified the functional groups of the normal polymeric OH and C-H stretching of the methylene group at wavenumbers of 3280–3300 cm^{-1} and 2920–2935 cm^{-1}, respectively. According to FTIR analysis, corn rice with the presence of citric acid bioplastic was chosen to undergo AFM to survey roughness in case to determine whether crosslinking might happen; the different average roughness between pre-irradiated and post-irradiated samples is 8.82 nm. Based on these findings, bioplastics may contribute tremendous benefits, especially in smart packaging applications.

Keywords: *Waterproof, irradiated, packaging, bioplastic, cobalt-60*

1. Introduction

Plastics are everywhere, including the market, ocean, streets, and in the body. None degrade well because the plastic compounds are hard to break down. Plastics have become popular due to their barrier properties and strengths. We can obtain them anywhere; they are cheap, resistant to water, and flexible. Currently, polymers can be molded into various shapes. Most people are obsessed with plastic, and the beloved planet becomes dirty and unorganized, negatively affecting lands, waterways, and the ocean. This is because plastics have been used for more than 50 years. It was created in the late 19th and early 20th centuries. Besides, plastic bombing occurred during World War II (Clunies-Ross, 2019).

Carbon emissions cause environmental issues during manufacturing because they cause harm rather than positive effects. An estimated 33 billion tons of plastic are produced annually due to the increasing rate of plastic production. Single-use plastics accounted for approximately one-third of the total plastic production in 2018, with an estimated 359 million tones. Many efforts have been made to reduce the amount of plastics, but they have not been successful, including the 3R program, reduction, reuse, and recycling. This affects but not on a large scale. Thus, bioplastics are created to educate people about making better choices to save the earth.

Many people are unfamiliar with bioplastic production, but only a few have used it in their lives. Bioplastics are now widely known in society and their awareness has occasionally increased. Therefore, biodegradable materials are preferred to overcome fossil fuel consumption and plastic accumulation (Tokiwa et al., 2009). Natural polymers are more environmentally friendly and easy to dispose of (Marichelvam et al., 2019). Their chemical and physical properties must meet the ideal characterization to construct a beautiful structure to evaluate the biodegradability of solid polymers. The term “bioplastic” can be confusing because some petroleum-based plastic also degrades or bio-based plastic, which is synthesized from biomass or renewable sources. Some bio-based plastics, like Nylon 11, has been produced as non-biodegradable. Unlike acetyl cellulose, biodegradability refers to the degree of acetylation.

However, fossil fuel is at an endangered level. Hence, plastics synthesized from biomass are preferable. Bioplastics have many kinds because the molecules can be carbohydrate-based or fat and oil-based polymers to reduce dependency on fossil fuels. However, carbohydrate-based polymers are the most popular because they are derived from starch, cellulose, lactic acid, lactide, polyhydroxyalkanoates, and chitosan, which are easier to obtain, especially starch-based polymers because they are the

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cheapest among others. Bioplastics are growing in market niches, such as packaging, agriculture, and automotive parts due to cost and trends (Aranda-Garcia et al., 2015).

Domestic and municipal composting are the preferred end-of-life disposal options for these materials (Mostafa et al., 2018) instead of landfills, which is the worst disposal option. Starch and water cannot compete with polyethylene (PE) or polypropylene (PP). Therefore, plasticizers must be added to enhance the water barrier and mechanical strength. Plasticizers, such as glycerol, sorbitol, citric acid, and beeswax (Byun et al., 2014), have been tested in several studies to reduce water permeability. However, not all studies will derive the same result because each plasticizer has weaknesses. Therefore, the wettability of bioplastics must be determined by studying their moisture content and water contact angle.

When it comes to radiation exposure to products, people make a fuss, and myths about the products emerge over time. People fear ionizing radiation because it involves radioactive substances or handling equipment. Most people believe that radiation exposure is equivalent to death. Many studies have been conducted on handling and using radiation as a progress to produce a product. A few incidents show how evil radiation can be, but scientists were more careful after that incident, and standard operations and policies were enhanced to ensure that workers faced less health risks. The incident became a story that served as a reminder to prioritize safety. "The benefits hidden behind a negative image are finally revealed."

Packaging material is widely used in markets, industries, and households. Over 67 million tons of packaging have been reported (Maulida et al., 2016), leading to severe environmental complications. In a landfill, the dumps of packaging material increase yearly as online shopping becomes a trend among shoppers. This results in a poor situation because most packaging is polymer-based and degrades poorly. Some studies have started to apply bioplastic in the packaging sector, but due to its short-term shelf-life nature, it does not spread well, especially when dealing with water resistance. Many studies have been conducted on handling and using radiation as a progress to produce a product. A few packaging materials are listed in Title 21 of the Code of Federal Regulations (CFR) (Paquette et al., 2004) and irradiated under regulation 179.45(d) with a maximum dose of 60 kGy. This shows that packaging material has already been introduced to radiation for years.

Bioplastics have many properties that must be enhanced to achieve a good packaging standard and can replace petroleum-based plastics in the future. Environmentally friendly materials have a shorter shelf life, as they degrade faster than petroleum-based plastics, removing carbon dioxide, methane, water, wood, humic matter, and other natural substances (Ibrahim et al., 2017). Starch-based bioplastics are mixtures of amylose/amylopectin ratios, depending on their botanic origin (Jiang et al., 2017). Starch may have many weaknesses, and plasticizers help maintain their chemical and physical robustness. Since bioplastic's biodegradability is faster than petroleum-based plastics, their lifelong has been questioned many times. They can be an excellent choice for degradation, but wearing them for a long time is not recommended.

Barrier properties have focused on providing an effective and cost-effective packaging system and maximizing the shelf life of packaged foods and beverages. Hydrophobicity mostly depends on the material used or the concentration of the material. Bioplastics must achieve a level where they can be picked as desired products. Bioplastics production must beat petroleum-based plastic standards because, through composition, all these biomaterials are hydrophilic but quite crystalline, messing things up, specifically regarding the packaging of wet products. For decades, many studies have experimented with radiation toward polymers. The radiation process is difficult because it can cause significant damage and may break the bonds of the plasticizer and other compounds as the polymer chains are broken and/or cross-linked, allowing the mechanical, thermal, and physicochemical properties to change if exposure over the limit of the substance can be handled (Zygoura et al., 2011).

In this study, mixed- and pure-starch bioplastics were compared. Rice, potato, and corn starches were used to produce the samples. Examples of mixed starches include potato rice, corn rice, and corn potato. Additional materials, including glycerol and distilled water, were used at a constant weight. Plasticizers such as sorbitol and citric acid were compared to analyze their wettability and mechanical properties. Then, bioplastic samples undergo a characterization test and irradiation treatment effect test.

The wettability of the samples under characterization can be determined by measuring the contact angle with a sessile drop and studying their moisture content. Fourier Transform Infrared (FTIR) spectroscopy was used to determine the polymer behavior and molecule concentration to compare pre- and post-irradiation bioplastic samples. Pure and mixed starch morphology were compared to common plastic using atomic force microscopy (AFM). Gamma radiation was used to irradiate samples at doses ranging from 0.5 kGy to 2.5 kGy.

2. Methodology

Chemicals

In this study, 5 g of corn flour, 5 g of rice flour, and 5 g of potato flour were combined to prepare a bioplastic. However, 1.5 g of glycerol, 1 g of sorbitol, 0.3 g of starch, 0.5 g of citric acid, and 1 mL of distilled water were mixed with each type of bioplastics. Low-density polyethylene (LDPE) plastic was used for comparison. All the chemicals were purchased from Emory.

Table 1. Abbreviations used for Bioplastics with Different Types of Plasticizers

Types of Bioplastics		Types of Plasticizers	
		Sorbitol	Citric acid (CA)
Starch (S)	Corn (C)	CS	CCA
	Rice (R)	RS	RCA
	Potato (P)	PS	PCA
	Corn-Potato (CP)	CPS	CPCA
	Corn-Rice (CR)	CRS	CRCA
	Potato-Rice (PR)	PRS	PRCA

Sample Preparations

Different solution mixtures were stirred until the starches achieved the gelatinization phase, and the solution's liquid consistency turned from watery to a thick gel. The temperature was regularly checked to prevent overheating of the molecular mixtures, as it might decrease the function or properties of the bioplastics. When the solutions thickened, it was poured into the proper mold. The solution was dried for three–four days before turning into the solid phase. Bioplastic films were made in several pieces for characterization tests. LDPE and bioplastic pieces were produced for tensile strength testing. All plastics were exposed to five doses of gamma radiation: 0.5, 1.0, 1.5, 2.0, and 2.5 kGy. Afterward, all the post-irradiated sample conditions were compared to the pre-irradiated sample conditions with water contact angle reading and Fourier-Transform Infrared (FTIR) spectroscopy.

Sample Characterizations Water Contact Angle

Hydrophilicity has become a problem for bioplastic production. The bioplastic hydrophilicity was determined by measuring the wettability of the bioplastic surface. The contact angle is the most sensitive surface analytical technique because wettability is influenced by the top of the surface nanometer. The contact angle was determined by checking the intersection angle. The test was run in a temperature- and humidity-controlled room to reduce random errors when reading was performed. Moreover, sessile droplets were used to interact with polymer molecules. The contact angle test was performed using Video Contact Angle (VCA) optima.

Moisture Content Test

This test has a similar goal to the water contact angle test. However, in this case, glycerol played a significant role in determining the moisture content percentage, which was obtained using the following equation:

$$\text{Moisture Content} = \frac{(W_1 - W_2)}{W_1} \text{x}100 \qquad (1)$$

with W_1 = initial weight before drying, W_2 = final weight after drying

W_2 can be obtained by drying the bioplastic sample at 100C - 110 °C in an oven to obtain a fixed reading as the final weight.

Biodegradability Test

The soil burial method was chosen to consider landfill conditions for at least 15 days, and the time could be lengthened if necessary. The weight loss percentage was calculated using the following formula:

$$\text{Weight loss percentage} = \frac{(W_0 - W)}{W_0} \text{x}100 \qquad (2)$$

with W_0 = weight of the sample before burial , W = weight of sample after burial.

The samples were buried for 15 days before being weighed. The biodegradability test was conducted in a plastic bag to avoid confusion with other foreign materials. Each bag was filled with approximately 300 g of soil, and the sample was buried deeply to ensure that the soil covered the sample. Each sample had its bag and was labeled with stickers.

Tensile Strength Test

All samples were set to the same length and height. All samples were placed vertically, with a tensile grip probe gripping both ends and measuring 1 cm wide and 7 cm long. A texture analyzer (Stable Micro System machine) was used for the assisted tensile strength test. Fixed parameters for the test were chosen under the packaging film standard test, with a test speed of 60 mm/min and tension mode.

FTIR Spectroscopy

FTIR spectroscopy is an analytical machine to ease the research journey. FTIR is well known for characterization analysis due to its accuracy and sensitivity by affecting atomic vibration with infrared radiations. There are several wavenumber divisions: far, mid, and near spectra. However, the mid-region of the wavenumber was used to analyze the samples because this spectrum region was divided into four more categories: single bond ranging from 2500 to 4000 cm^{-1}, triple bond ranging from 2000 to 2500 cm^{-1}, double bond labeled ranging from 1500 cm^{-1} to 2000 cm^{-1}, and fingerprint ranging from 600 cm^{-1} to 1500 cm^{-1}. Approximately 1 g of different types of bioplastics were prepared for FTIR characterization.

AFM

AFM was used to determine the roughness of sample surfaces. A laser beam was produced to light behind the cantilever and was reflected towards the photodiode. There are three modes: contact, tapping, and lateral. This experiment was performed in tapping mode, where the contact between the tip and samples is discontinuous due to the sensitive sample surface. Moreover, tip contamination may result in a convex fold related to the contaminant size relative to the surface size features to be measured.

Gamma Irradiation Treatment

In this study, gamma and ultraviolet (UV) irradiation treatments were used to apply to the samples. However, a photosensitizer was required to complete the UV irradiation treatment process because it was feared that increasing the barrier properties would cause the sample to become completely hydrophobic. Therefore, gamma radiation was used in this study. The samples were treated with five doses of 0.5, 1.0, 1.5, 2.0, and 2.5 kGy.

3. Results and Discussion

Water Contact Angle

LDPE is the control sample, showing 97.70° for the contact angle and 99.60° for the right after irradiation; it decreased to 96.40° for the left and 95.30° for the right. Irradiation treatment can degrade and reduce the contact angle in the presence of free radicals (Gorna *et al*., 2003). Crosslinking may occur in some samples as the contact angle values increase. However, it maintained a hydrophobic state after losing 1°. For sorbitol in pure starch, the contact angle for the pre-irradiated potato bioplastic was 0°; however, after irradiation treatment, it increased to 49.40° for the left and 56.10° for the right. The same applies to the corn bioplastic case, where the contact angle increased. After treatment, the rice bioplastics experienced much lower contact angles of 13.30° on the left and 17.60° on the right. Unlike when citric acid is involved, all pure starch bioplastics do not experience a significant difference after treatment. Corn-potato bioplastic reading angles were unique in the mixed bioplastic cases because when sorbitol was present, the contact angle decreased, whereas citric acid produced the opposite result. It increases from approximately 30° to 60°. Simultaneously, the corn-rice bioplastic contact angle differed significantly before and after irradiation, starting at approximately 30° and becoming hydrophobic at approximately 106°.

Plasticizers in bioplastics help reduce brittleness/fragility and increase flexibility in films, making them simpler to handle and prevent cracks and pores. Plasticizers are classified into two types, hydrophilic and hydrophobic, based on their chemical properties. When hydrophilic plasticizers are applied to bioplastics at greater concentrations, they can lead to increased water diffusion in the plastic. Hydrophobic plasticizers can reduce water uptake by plugging the micro-voids into the films (Varee and Bhaswati, 2019). These factors influence the wettability of bioplastics. Different bioplastics exhibit varying degrees of wettability.

Table 2. Contact Angle of Bioplastic with Presence of Sorbitol or Citric Acid

Contact Angle (°) (Left, Right) Types of Bioplastic	Types of Plasticizers			
	Sorbitol		Citric Acid	
	Before Radiation	After Radiation	Before Radiation	After Radiation
Corn	41.10, 37.60	67.10, 67.90	103.90,100.60	99.70, 98.90
Rice	22.10, 23.80	13.30, 17.60	101.90, 99.80	109.10, 106.70
Potato	0.00, 0.00	49.40, 56.10	33.10, 25.70	33.70, 37.90
Corn-Rice	96.70, 94.50	85.50, 89.50	28.70, 29.00	106.70, 105.20
Corn-Potato	43.40, 44.20	27.40, 28.70	34.10, 39.70	61.90, 60.90
Potato-Rice	29.10, 12.80	20.80, 19.30	100.10, 98.60	103.80, 100.90
	Before		After	
LDPE	97.70, 99.60		96.40, 95.30	

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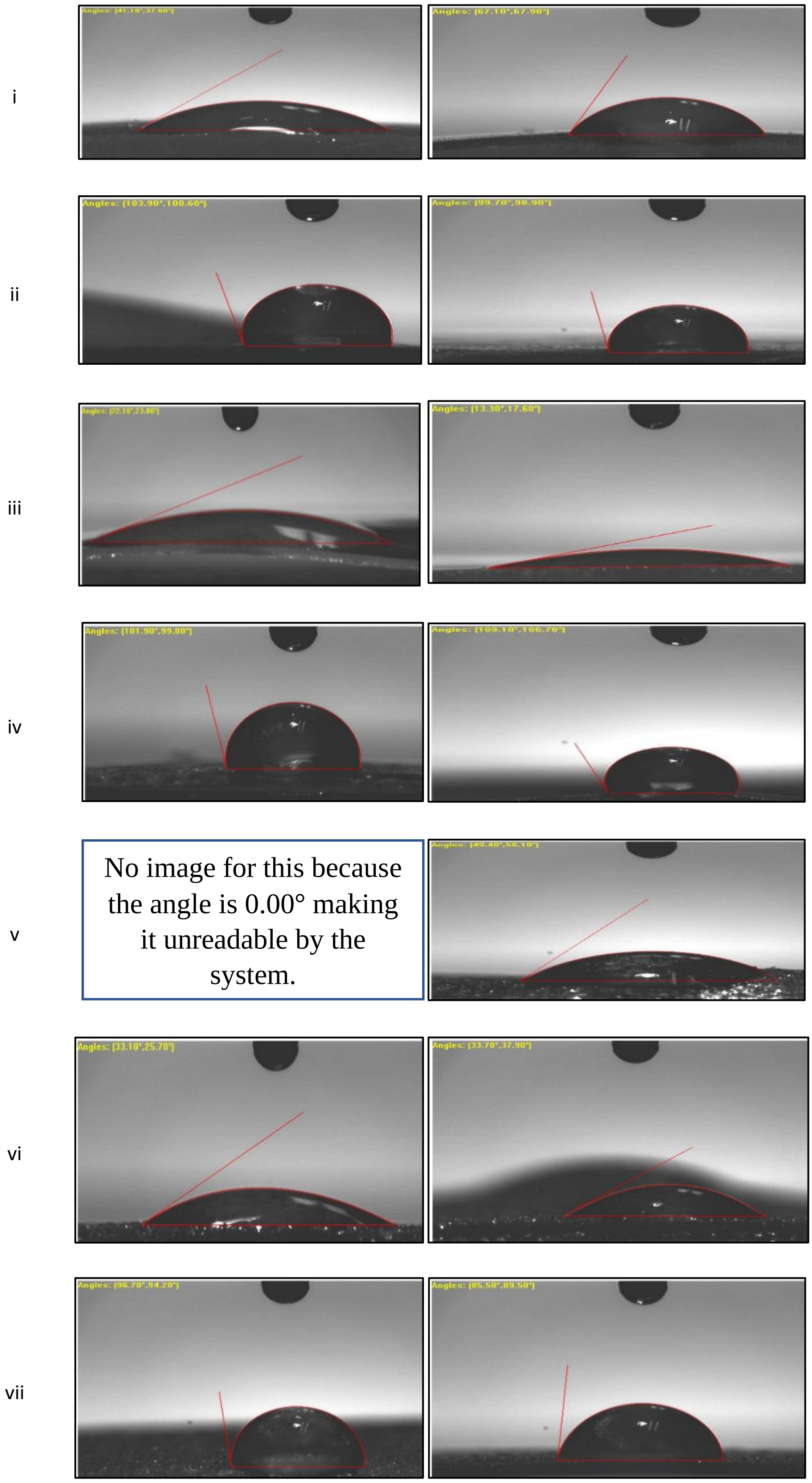

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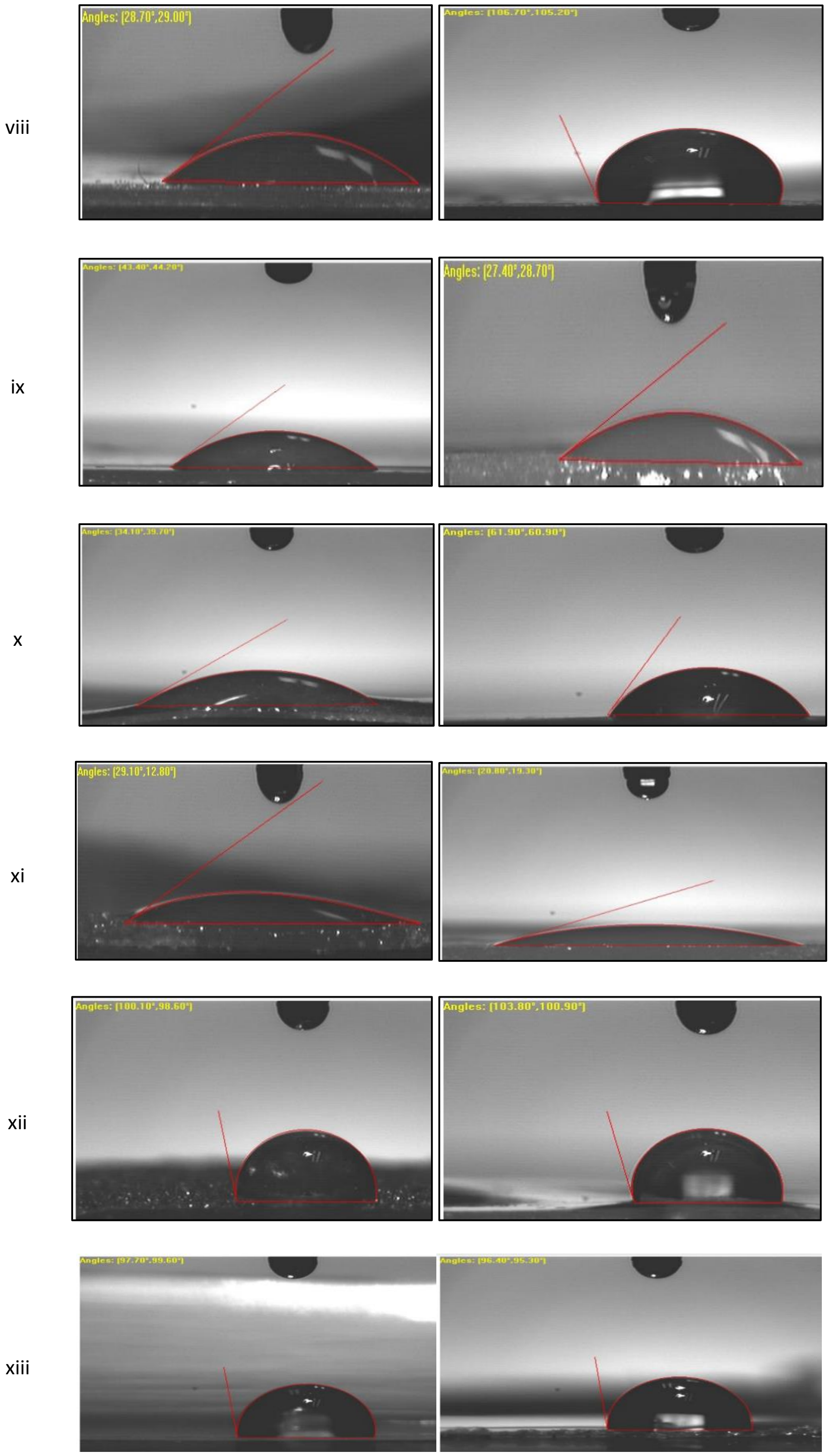

(a) (b)

Figure 1. Contact angle of bioplastic with presence of sorbitol or citric acid (a) Pre-irradiation (b) Post-irradiation for (i) CS (ii) CCA (iii) RS (iv) RCA (v) PS (vi) PCA (vii) CRS (viii) CRCA (ix) CPS (x) CPCA (xi) PRS (xii) PRCA (xiii) LDPE samples

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Content of Moisture Test

When all the bioplastics were dried in an oven from 100 °C to 110 °C for the first 30 min, their weights did not differ considerably. Thus, several samples showed weight changes for the second time, but all samples had a difference in weight for the third time. All the samples were placed in an oven for the fourth time to ensure a fixed weight of the dried samples. Some remained at the same weight as before, while others decreased and reached a fixed weight after the fifth 30 min. The duration of this test was approximately 2 h and 30 min. Table 3 shows that rice-sorbitol bioplastic has a high moisture content of 18.49% in the pure starch category, whereas corn has 21.21% in the citric acid category. Regarding the mixed starch category, potato-rice bioplastics had a high moisture content of 16.68%. However, when citric acid replaced sorbitol, the corn-potato bioplastic had the highest moisture content (22.11%). The moisture content of the bioplastics is related to their wettability. It is defined as the tendency of a fluid to spread or adhere to a material surface (Tarek Ahmed, 2019). Therefore, it alters the three-dimensional network of the polymer chains, resulting in increased mobility by increasing the free volume. Plasticizers aid in bioplastic resistance to migration and extraction in water or solvents (Sothornvit and Krochta, 2005).

Table 3. Weight of Samples for Moisture Content of Bioplastic with Presence of Sorbitol or Citric Acid

Weight	Types of Plasticizers					
	Sorbitol			Citric Acid		
Type of Bioplastic	Initial Weight (g)	Final Weight (g)	Moisture Content (%)	Initial Weight (g)	Final Weight (g)	Moisture Content (%)
Corn	0.3654	0.3114	17.34	0.2372	0.1957	21.21
Rice	0.3582	0.3023	18.49	0.2005	0.1714	16.98
Potato	0.5586	0.4750	17.60	0.1481	0.1287	15.07
Corn-Rice	0.4704	0.4105	14.59	0.2844	0.2455	15.85
Corn-Potato	0.9380	0.8558	9.60	0.3407	0.2790	22.11
Potato-Rice	0.2203	0.1888	16.68	1.4673	1.2555	16.87

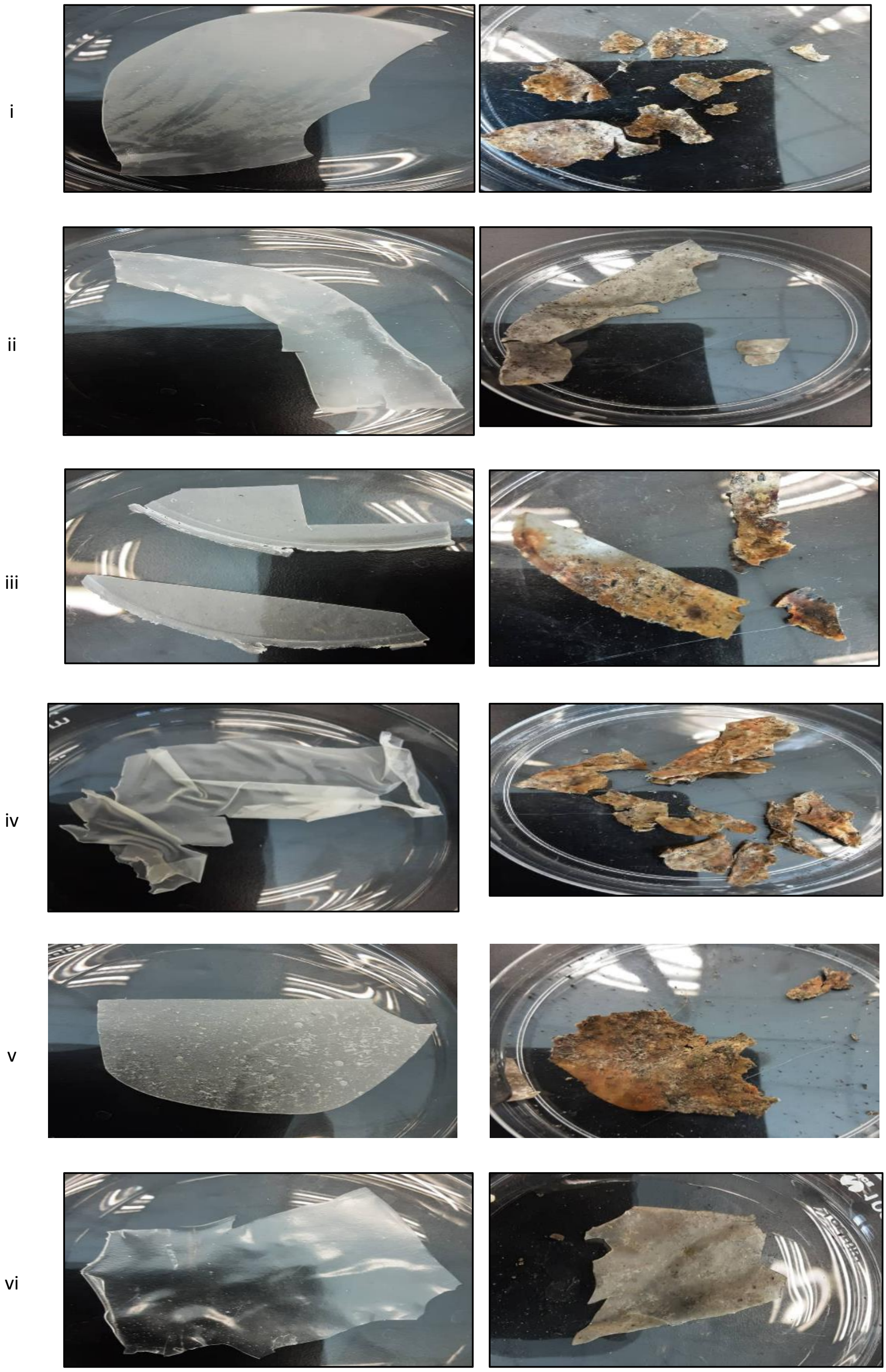
Biodegradability Test

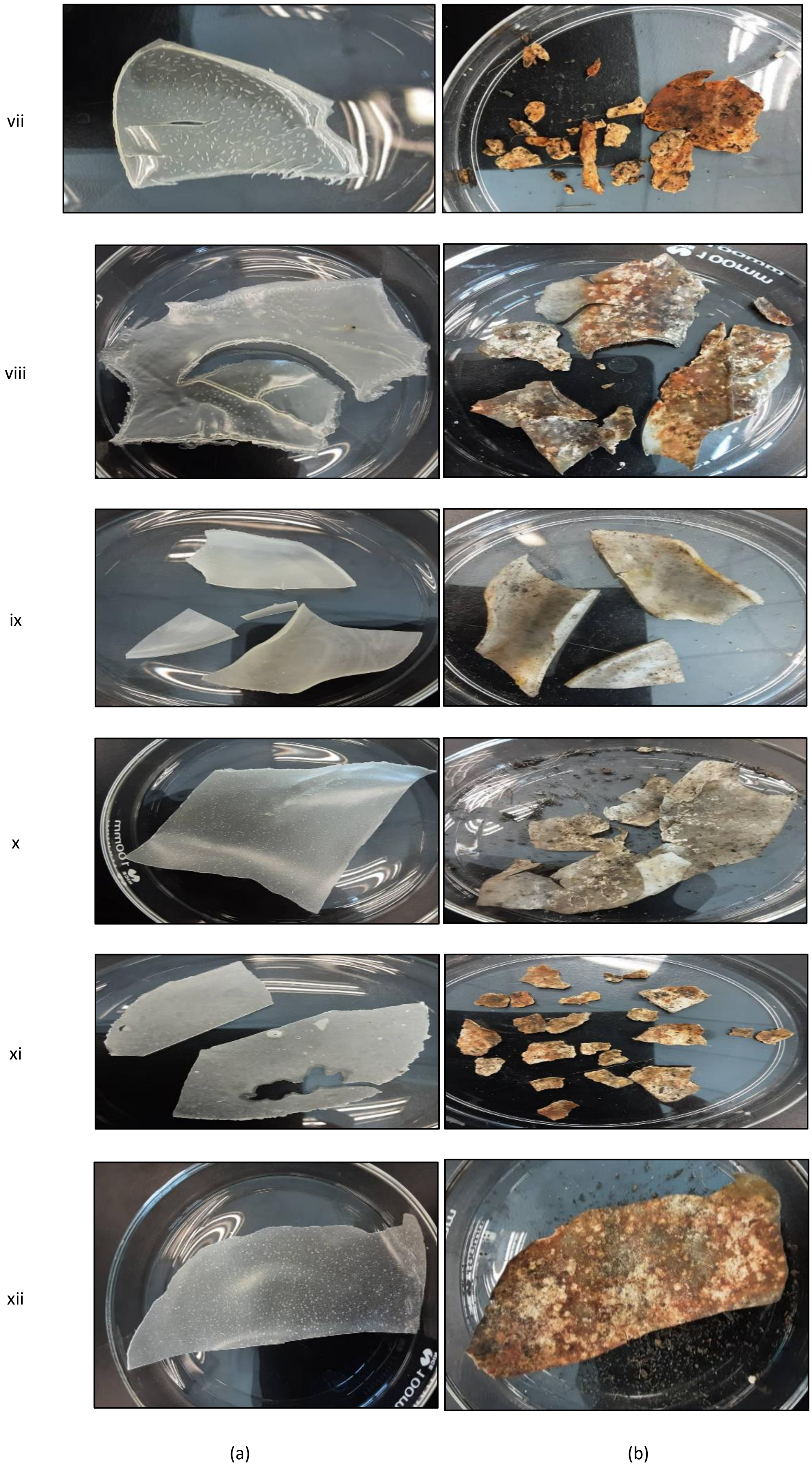
Degradation is required to dispose of waste without negatively affecting environmental conditions, and bioplastics should pass this test. Consequently, the duration of degradation varies depending on the mixture of molecules. When hydrophobicity increases, the chance of degradability may decrease; however, all bioplastics beat the challenge. The lowest weight loss percentage was 9.9% for the corn-potato bioplastic (Table 4). Moreover, all bioplastics involving sorbitol as a plasticizer experience a great weight loss percentage, from 47.36% to 80.17%. Unlike when citric acid played the role of sorbitol, only rice bioplastics had a high weight loss of 53.15%, while the others only achieved a range of 11.34% to 21.28%. The primary process generated by irradiation is degradation, which is accompanied by cross-linking. Irradiation-induced changes in film characteristics can be attributed to changes in the film surface properties (particularly surface oxidation) and structures. The chain scission process causes degradation to reduce the molecular weight of all irradiated samples (IAEA, 2019). Adding plasticizers to pure starch may increase its workability and reduce film brittleness. Plasticizers aim to improve starch flexibility and processibility by lowering the strong intermolecular interactions between starch molecules. Therefore, the polymeric chain mobility increases, improving the flexibility, extensibility, and ductility of plasticized films (Sanyang *et al.,* 2015).

Table 4. Weight of Samples for Biodegradability of Bioplastic with Presence of Sorbitol or Citric Acid

Weight of Samples (g)	Types of Plasticizers					
	Sorbitol Biodegradability			Citric Acid Biodegradability		
Types of Starch	Initial Weight (g)	Final Weight (g)	Weight Loss (%)	Initial Weight (g)	Final Weight (g)	Weight Loss (%)
Corn	0.7840	0.1555	80.17	0.2918	0.2587	11.34
Rice	0.5109	0.2102	58.86	0.8972	0.4203	53.15
Potato	1.0268	0.5045	47.36	0.7229	0.6311	12.69
Corn-Rice	1.2442	0.5920	52.42	2.3591	1.8570	21.28
Corn-Potato	1.8327	1.6512	9.90	1.5723	1.2878	18.09
Potato-Rice	1.0188	0.3980	60.93	1.9409	1.5718	19.02

Figure 2. Images of Bioplastic with Presence of Sorbitol or Citric Acid During Soil Burial Process (a) before (b) after for (i) CS (ii) CCA (iii) RS (iv) RCA (v) PS (vi) PCA (vii) CRS (viii) CRCA (ix) CPS (x) CPCA (xi) PRS (xii) PRCA samples.




vii
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ix
x
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xii
(a)
(b)

Tensile Strength and Elongation Test

From the maximum force, F_{max} (kg), the tensile strength of the bioplastics was calculated using the equation for the force per initial area of the samples. Simultaneously, the elongation percentage was estimated from the difference between the initial and elongated length of the samples. Table 5 shows that for bioplastics with sorbitol as a plasticizer, the highest tensile strength would be the corn-rice bioplastic with 2.916 MPa, but the potato bioplastic had the highest percentage (34.57%). Meanwhile, when citric acid replaced the sorbitol, potatoes experienced 3.189 MPa. However, corn bioplastics had a high elongation percentage (28.98%). Bioplastics containing citric acid experience higher tensile strength due to their crosslinking characteristic, resulting in high rigidity in the polymer. The covalent intermolecular interactions between the hydroxyl and carboxyl groups may strengthen the bonds between the plasticizer and starch compounds (Azeredo *et al.*, 2016). However, the samples' tensile strength is inconsistent with the elongation percentage reading.

Table 5. Tensile (MPa) and Elongation (%) of Bioplastics with Presence of Sorbitol or Citric Acid

Tensile and Elongation	Types of Plasticizer Involved					
	Sorbitol			Citric Acid		
Types of Bioplastic	F_{max} (kg)	Tensile strength (MPa)	Elongation (%)	F_{max} (kg)	Tensile strength (MPa)	Elongation (%)
Corn	1.0300	1.4430	31.150	1.058	1.4820	28.980
Rice	0.5970	0.8364	16.120	0.538	0.7537	14.410
Potato	1.1930	1.6710	34.570	2.276	3.1890	18.980
Corn-Rice	2.0810	2.9160	10.350	1.198	1.6780	23.150
Corn-Potato	1.2160	1.7040	13.780	1.592	2.2300	16.980
Potato-Rice	0.6350	0.8896	6.240	2.923	4.0950	8.297
Type of Plastic	F_{max} (kg)		Tensile strength (MPa)		Elongation (%)	
LDPE	1.558		2.183		84.31	

FTIR Spectroscopy

FTIR spectroscopy was used to investigate the molecules that appeared with a certain transmittance percentage. Figure 3(a) indicates that each bioplastic experienced a curve ranging between 2800 and 2930 cm^{-1} and at wavenumbers of 3200–3400 cm^{-1} in the pre-irradiated samples. However, PRCA achieved the highest transmittance (73.62%) with a wavenumber of 3287 cm^{-1}. In contrast, RPS exhibited the lowest transmittance (50.78%) at 3292.96 cm^{-1}. The LDPE graph narrowed down to 2916 cm^{-1} and 2848 cm^{-1} with transmittances of 69.97% and 72.35%, respectively. The CS curve exhibits a transmittance of 77.95% at 2929 cm^{-1}. This shows that LDPE and CS have a highly functional group of methylene C-H asymmetric/symmetric stretching, whereas RPS has a highly normal polymeric OH stretching.

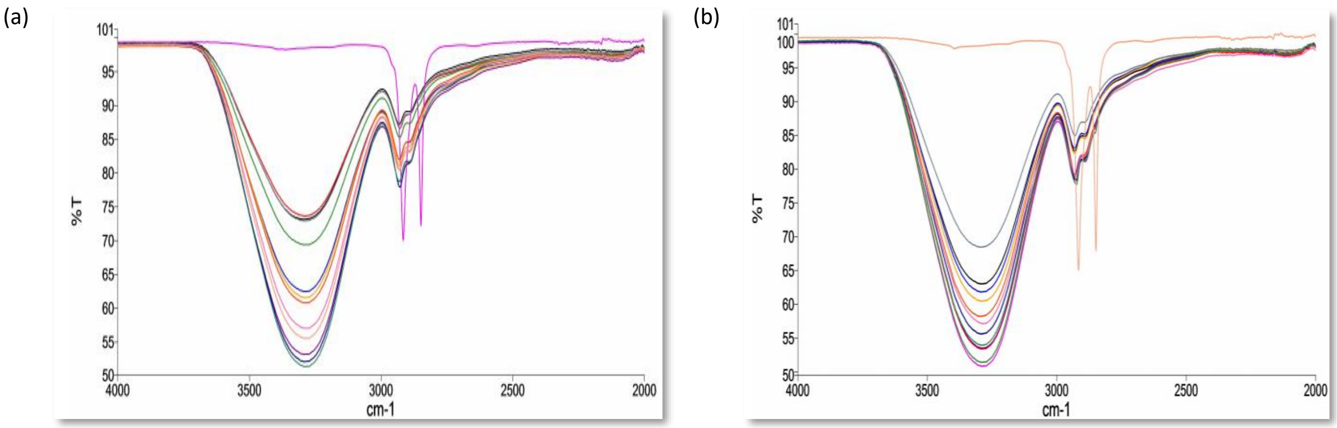

Figure 3. Graph of FTIR Analysis of (a) Pre-Irradiated samples (b) 0.5kGy Irradiated Samples

Figure 3(b) shows that the PRS has a low transmittance percentage at 3292 cm^{-1} (50.78 %), followed by RS (51.36 %). However, in the range of 2800 cm^{-1} to 3000 cm^{-1}, RS had the lowest transmittance percentage of 78.56% at 2931.39 cm^{-1}, although there was not much difference with the PRS transmittance percentage at 78.63%. LDPE maintained the same wavenumber but decreased the transmittance percentage by 65.81% and 71.765%, respectively. Figure 4(a) shows that almost all bioplastics have a similar transmittance percentage, but the lowest is PRS, with 51.20% at 3293 cm^{-1}, followed by CS at wavenumber 3291 with 53.07%, and the lowest is bioplastic at 2923 cm^{-1} with 77.34%. The LDPE curves in three different regions, 3393 cm^{-1} with 95.93%, at 2916 cm^{-1} with 66.098%, and 2848 cm^{-1} with 67.99%. The result exposed 1.5 kGy gamma radiation, which resulted in a slightly lower transmittance percentage than the 1.0 kGy (Figure 4b). PS had the lowest 50.77% at 3285 cm^{-1}, followed by CS with 50.89% at 3283.9 cm^{-1}. At wavenumber 2931 cm^{-1}, it has an equal transmittance percentage with CRCA at 2930 cm^{-1} with 78.59%.

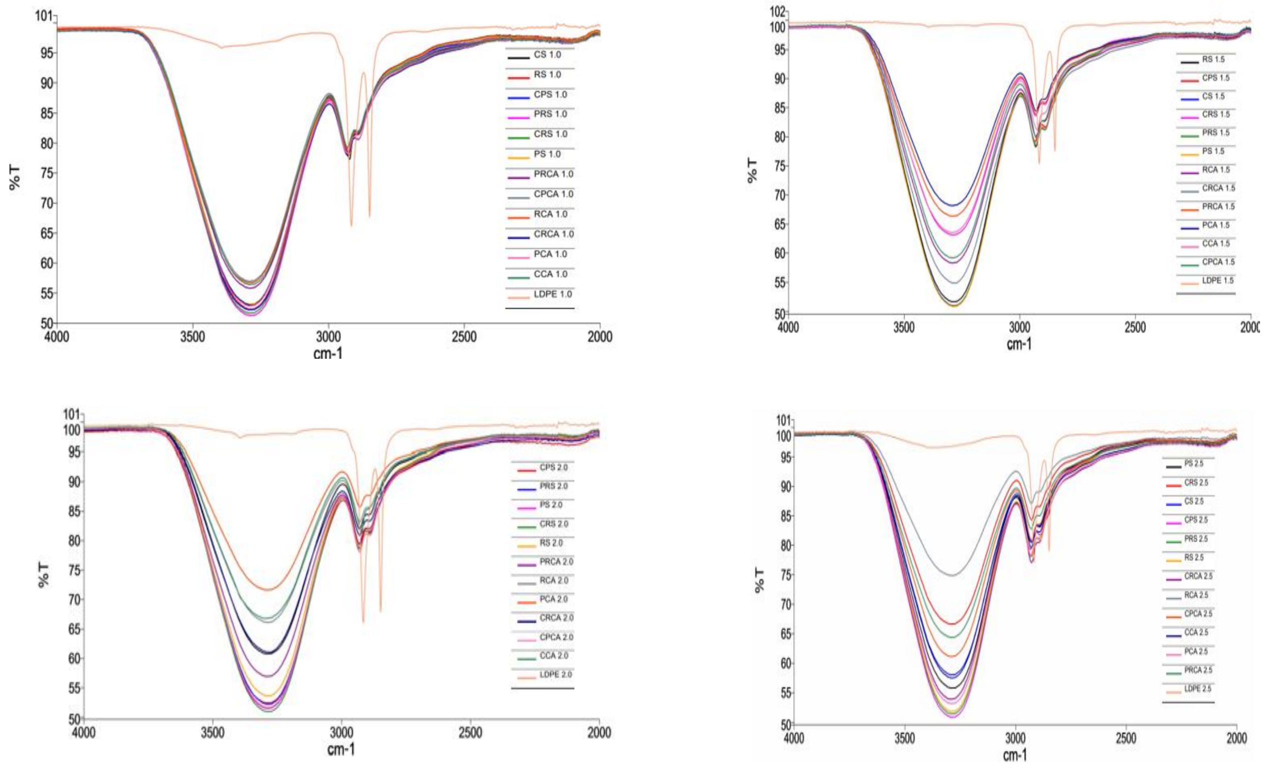

Figure 4. Graph of FTIR Analysis of (a) 1.0 kGy Irradiated Samples (b) 1.5 kGy Irradiated Samples (c) 2.0 kGy Irradiated Samples (d) 2.5 kGy Irradiated Samples

Figure 4(c) shows the results of the sample treated under gamma radiation at a dose of 2.0 kGy. CRS achieved the lowest transmittance of 50.89% at a wavenumber of 3285 cm^{-1}. The LDPE remained at the same wavenumber, but the transmittance percentage changed to 66.03% at 2916 cm^{-1} and 68.18% at 2848 cm^{-1}. PS achieved low transmittance percentages of 51.68% at 3266 cm^{-1} and 78.62% at 2931 cm^{-1}. Figure 4(d) shows that the CPS percentage was 50.75%, making it have the lowest transmittance at 3293 cm^{-1}, followed by CRCA at 3294 cm^{-1} with 53.81%. However, CRCA leads with 77.17% at 2928 cm^{-1} than CPS with 79.38% at 2933 cm^{-1}. LDPE again obtained three readings at wavenumbers of 3361 cm^{-1} with 96.73%, 2915 cm^{-1} with 77.29%, and 2848 cm^{-1} with 79.03%.

Most bioplastics experienced decreasing transmittance percentages as exposure radiation doses increased due to interactions via the hydroxyl group of bioplastics. Furthermore, bonding significantly enhances the mechanical strength and degradation of bioplastics for polymeric materials (Wang *et al.,* 2017). In contrast, RCA had the highest percentage when exposed to a dose of 2.5 kGy for the pure starch bioplastic. However, CRCA had the lowest percentage of mixed-starch bioplastics. All bioplastics show two huge curves, with the first curve at wavenumbers in the range of 3280–3300 cm^{-1}, followed by the second curve at wavenumbers in the range of 2920–2935 cm^{-1} from all six graphs of FTIR analysis due to similarities of functional groups (Hindi *et al*., 2017). The first curve shows the normal polymeric OH stretch group and the second curve defines the asymmetric C-H stretch of the methylene group (Coates, 2006).

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Table 6. Functional Groups from FTIR Analysis Spectrum

Functional group	Wavenumbers (cm^{-1})
OH stretch group	3280- 3300
C-H stretch of methylene group	2920- 2935

AFM

To analyze the roughness, an image of the sample topography in the tapping mode was used to reduce contact between the tip and the samples, as shown in Figure 6.

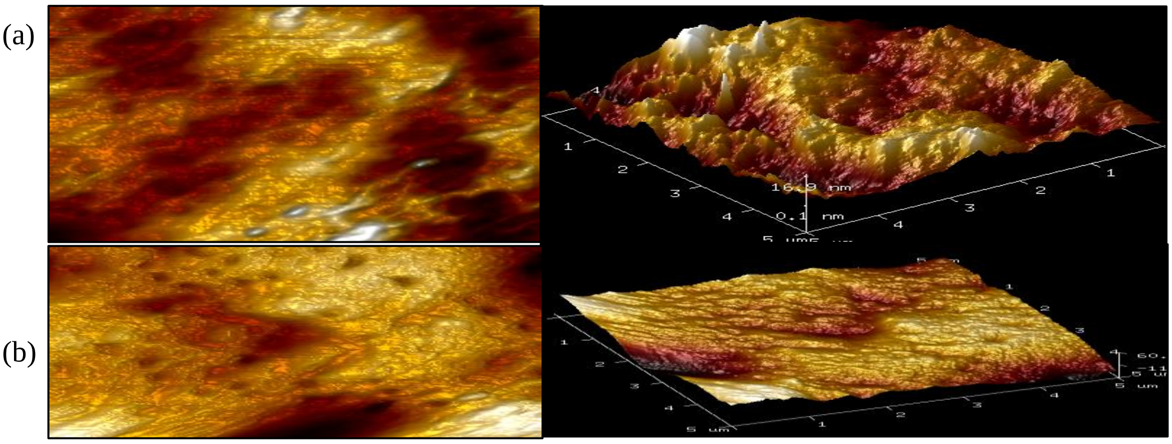

Figure 5. CRCA roughness conditions (a) before and (b) after irradiation exposure

The pre-irradiation sample had a peak-to-peak distance of 2.14011 nm. The minimum peak depth was 17.7510 nm and the maximum peak depth was 19.9 nm. The maximum depth at the histogram was 19.8911 nm, with 157 peaks. The difference in the image surface area was 0.0647%. The root means square image was 4.64 nm with an average roughness of 3.68 nm. The maximum image roughness was detected at 34.7 nm. When the samples underwent irradiation treatment, the peak-to-peak distance became 2.74845 nm, with a minimum peak depth of 44.1834 nm. The maximum peak depth was recorded at 46.9 nm, with 162 peaks and a maximum histogram depth of 46.9318 nm. The roughness result was described with an image surface area difference of 0.738%, with a root mean square of 17.1 nm, average roughness equal to 12.5 nm, and maximum image roughness was detected at 122 nm. According to Oleyaeia *et al*. (2016), surface roughness is related to water permeability. However, irradiation treatment might indicate surface etching, leading to increased roughness.

4. Conclusions

Regarding wettability, mixed starches were confirmed to deal with hydrophilicity, but pure starch could handle it with citric acid as a plasticizer. However, CRS was the highest for sorbitol, while PRCA was the best angle for citric acid as a plasticizer, with readings of 100.10 ˚and 103.8˚, respectively. The water contact angle test revealed that many bioplastics containing citric acid act as plasticizers to achieve hydrophobicity. Regarding moisture content, RS (21.21%) led to pure starch, but corn potato with citric acid (22.11%) was present in the mixed starches. The highest biodegradability percentages were for CS (80.17%), PRS (60.93%), and RCA (53.13%). The tensile strength cases were enclosed by the CRS (2.916 MPa) and PRCA (4.095 MPa) results. Meanwhile, for elongation, PS (34.57%), and CCA (28.98%). CRCA was subjected to AFM topography to survey its surface roughness because it has a high transmittance percentage from FTIR analysis under dose radiation-exposed 2.5 kGy. The hydrophobicity levels for bioplastics can achieve LDPE standards, but other physical properties are far from the target. However, only the C-H stretch methylene group could achieve the same transmittance percentage for chemical bonding. Therefore, this study meets the objectives and can serve as a reference to other researchers to enhance the properties of existing and future bioplastics, especially in smart packaging applications.

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