Bioplastic from Pectin of Dragon Fruit (Hylocereus polyrhizus) Peel

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1. Introduction

The use of plastic has become part of daily life because of the versatile function of plastics in various life sectors. However, plastic also poses a significant threat to the environment where plastic causes soil pollution because it occupies about 25% of the total volume of land on earth [1]. Plastics are made from polymer compounds derived from cracking or breaking of long chains of petroleum hydrocarbons. Usually, plastics are made of polyethylene terephthalate (PET), low and high-density polyethylene (LDPE and HDPE), polystyrene (PS), and high-density polyethylene (PE) [2, 3]. The use of plastic derived from petroleum endangers the environment as these plastic products survive in the environment and are difficult to degrade, causing environmental problems [3, 4].

The negative effects of the use of plastics that have been synthesized from petroleum have led to the production of plastics that originate from the use of easily degraded materials. These materials can come from vegetables, animals, or microbial materials [5]. Bioplastics are defined as plastics that have characteristics derived from biological creatures, are biodegradable, or contain both of these characteristics [6, 7]. Starch-containing plants can be used as ingredients to make bioplastics [8]. These plants include corn, potatoes, wheat, tapioca, and rice, in which the most widely used material in making bioplastics is corn. However, challenges also arise because corn is a food commodity, resulting in competition occurs between the corn used to be a food source and a bioplastic source [5].

Food waste or agricultural waste has the potential to be used as material for bioplastics. According to the Economist Intelligence Unit, Indonesia produced food waste nearly 300 kilograms of food waste per person in 2016 [9]. Food waste is mostly organic waste from fruit and vegetable peels, expired foodstuffs, or uneaten dishes. Parts such as peel, seeds, stems, and parts that are not edible are usually discarded and no longer utilized [10]. Several papers reported the potential use of food waste, especially waste materials from fruit and vegetables, to become bioplastics. Food waste such as citrus peel and apple pomace can be used for bioplastic and biofilm production [11, 12, 13]. Citrus peel and apple pomace are by-products obtained from juice or cider production. Food waste is generated every year in the world, including Indonesia. One of the fruits which have
high demand in Indonesia is dragon fruit. Studies show that dragon fruit contains anthocyanin and other minerals such as potassium, magnesium, calcium, and vitamin, suitable for the body [14, 15, 16, 17]. The production of dragon fruit is 737.5 tons annually, with a demand of around 1475 tons [18]. It is expected that dragon fruit peel will be produced every year and can be utilized for bioplastic production.

In general, fruit peels contain polysaccharides, i.e., pectin, and cellulose, to be used as material for making biodegradable plastics. Fruit peel waste also contains pectin [19, 20], a complex polysaccharide compound with the main component of D-galacturonic acid. Pectin is naturally contained in ripe fruits that are often used in the pharmaceutical, cosmetic, and food fields as a counterweight, thickener, and emulsifier. Dragon fruit peel weights of 30-35% of the fruit’s weight contain pectin ± 10.8%. Dragon fruit peels have not been used maximally and are discarded as agricultural waste [21, 22]. Several studies have reported pectin extraction from dragon fruit [15, 22, 23, 24]. There is also increasing interest in using pectin for biofilms for fresh vegetable coating [25]. Moreover, the utilization of food waste and agricultural waste into bioplastic products can add economic value to the material [26].

Making of bioplastics from agricultural waste is carried out to utilize waste into more useful products. This study aims to investigate the extraction of pectin and to make bioplastic from dragon fruit peel pectin.

2. Methodology
2.1. Materials and Tools

The dragon fruit peel was obtained from the traditional market in Yogyakarta. Other materials were concentrated HCl (Merck), ethanol (Merck), 2 M HCl solution, 2 M NaOH (Merck) solution, distilled water, pectin (CV. Chemix Pratama Yogyakarta) and ethylene glycol (CV. Chemix Pratama Yogyakarta). The tools used in the study were beaker glasses, stirring rods, spoons, hotplates, universal indicators, pipettes, drop, ovens, centrifuges, filters, gauze, mortar & pestle, blenders, stopwatches, labels, watch glass and Fourier Transform Infrared Spectrophotometer (IRSpirit-T Shimadzu) with DLATGS detector.

2.2. Preparation of Dragon Fruit Peel

Dragon fruit peel waste was washed and heated in an oven at 55°C for 48 hours. Dragon fruit peel was mixed with a blender so that the powder was produced. The powder was then filtered with a ten-mesh filter and stored in a tightly closed container.

2.3. Pectin Extraction

10 g of dragon fruit peel powder was added with 150 mL of distilled water. Pectin extraction of pectin was carried out using hot dilute mineral acid at pH ~ 2 [27]. The mixture was added with 2 M HCl to get a pH of 2.0. The mixture was stirred and heated at 50°C until it becomes a homogeneous mixture for 60 min [27, 28, 29]. Extraction using dilute HCl and heating in 60 minutes caused protopectin to hydrolyze into pectin to get more pectin precipitated. The filtrate was filtered and added with 150 mL of ethanol and left for 60 minutes at room temperature. The pectin was precipitated and centrifuged at 1500 rpm for 20 minutes. The precipitated pectin was then washed with ethanol with composition 1:3 (pectin: ethanol). The mixture was centrifuged at 1500 rpm for 30 min. The pectin obtained was then heated in an oven at 50°C for 24 hours. The dried pectin was milled and filtered with a ten mesh filter. The pectin yield was calculated as follows.

\[
\text{Pectin yield (\%)} = \frac{\text{Extracted pectin}}{\text{Dried dragon fruit powder (g)}} \times 100\%
\]

2.4. Preparation of Bioplastics

0.23 g dragon fruit pectin was added with 4.5 mL of distilled water. The mixture was added with 1 M NaOH solution to neutral pH. Two experiments were carried out in which mixture 1 (without plasticizer), mixture 2 (ethylene glycol 0.5 mL/g pectin). The mixture was smeared on the glass and dried in an oven at 50°C for 2 hours.

2.5. Determination of Moisture Content

Bioplastics were weight and heated in an oven at 100 °C with variation time 30 minutes, 60 minutes, 120 minutes, 24 hours and 48 hours until the weight is constant [30, 31]. The moisture content was defined as mass loss during heating and calculated as follows [11, 30, 32].

\[
\text{Moisture content (\%)} = \frac{W_1 (g) - W_2 (g)}{W_1 (g)} \times 100\%
\]

where \(W_1\) = weight of sample before heating (g), \(W_2\) = weight of sample after heating (g) [33, 34].

2.6. FT-IR Spectroscopy Analysis

Bioplastics were analyzed by the Fourier Transform Infrared Spectrophotometer (IRSpirit-T Shimadzu) with a DLATGS detector in the Chemistry Education Study Program Laboratory of Universitas Sanata Dharma. The spectra were recorded at absorbance mode from 4000 to 400 cm⁻¹. Also, the commercial pectin bioplastic (CV. Chemix Pratama Yogyakarta) was prepared using Method 2.4 mentioned above. Three samples of bioplastics, i.e., commercial pectin bioplastic, bioplastics of dragon fruit peel pectin, and bioplastic of dragon fruit peel pectin plus ethylene glycol, were analyzed using FT-IR spectrophotometer.

3. Results and Discussion

3.1. Extraction and Preparation of Bioplastics

The pectin yield was ~11%, and this result is not high because of the longer duration of hot water extraction influences the pectin yield [15]. This result is in line with the study of [15], which obtained 16.20–20.34% of pectin yield using an extraction duration of 20, 40, 60, and 80 min. However, the study of [15] was not used to dilute acid in extraction, which caused pectin yield to differ slightly from this study.

Bioplastics are synthesized with and without the addition of plasticizer. Ethylene glycol is a plasticizer
used for improving the elasticity of bioplastics [6, 22].

The surface bioplastic morphologies of the dragon fruit peel pectin are presented in Figure 1. Figure 1(a) is a bioplastic of dragon fruit peel pectin without ethylene glycol, which is rigid and difficult to be separated from the printing glass. Whereas Figure 1(b) is a bioplastic of dragon fruit peel pectin with ethylene glycol, which is easily separated from the printing glass.

![Figure 1](image)

**Figure 1.** Bioplastics (a) of dragon fruit peel pectin; (b) dragon fruit peel pectin and ethylene glycol morphology of the surface.

Although several studies have reported pectin extraction from dragon fruit peel, the studies on bioplastic production from dragon fruit pectin are still limited [15, 22, 23, 24]. In general, plasticizers are added to improve intermolecular interactions and bioplastic properties. Ethylene glycol decreases the density of the bioplastics. The formation of hydrogen bonds in the polymer chain causes a decrease in bioplastic density. Bioplastic stiffness is reduced because ethylene glycol lowers the polymer chain [35].

### 3.2. Moisture Content of Bioplastics

Bioplastic moisture content is shown in Table 1. Moisture content in bioplastics indicates the amount of water in the bioplastic. The bioplastics show constant mass after 60 minutes of heating. The moisture content of bioplastics of dragon fruit peel pectin is 5.71–12%, whereas dragon fruit peel pectin and ethylene glycol is 2.86–5.71%.

| Sample                          | Moisture content (%) |
|---------------------------------|----------------------|
| Bioplastics of dragon fruit peel pectin | 5.71                 |
|                                 | 12                   |
|                                 | 12                   |
| Bioplastics of dragon fruit peel pectin and ethylene glycol | 2.86                 |
|                                 | 5.71                 |
|                                 | 3.33                 |

Heating bioplastic at 100°C evaporates water in bioplastics. The loss of water results in bioplastic to be more rigid. Bioplastic moisture content is connected with its capability to absorb water from the surrounding environment. Moisture content depends on ambient conditions [11]. Bioplastic tends to absorb water, which can result in lower elasticity. The moisture content of bioplastic may alter the elasticity of the resulting plastic [36]. The moisture content in bioplastic of dragon fruit peel pectin and ethylene glycol is due to ethylene glycol absorbing ambient moisture.

### 3.3. FT-IR Spectroscopy

Bioplastics are analyzed to identify the functional groups. The pectin functional groups are usually present in the region between 1000 and 2000 cm⁻¹ in the FTIR spectrum [23]. FTIR spectra of pure commercial pectin plastics, dragon fruit skin pectin bioplastics, and dragon fruit pectin bioplastics and ethylene glycol are presented in Figure 2.

![Figure 2](image)

**Figure 2.** Bioplastics FT-IR spectra: pure commercial pectin; dragon fruit peel pectin; dragon fruit pectin and ethylene glycol

Analysis of the bioplastic functional group of dragon fruit peel pectin using FT-IR is presented in Table 2. The presence of carbonyl groups is at 1636 cm⁻¹ for bioplastic of dragon fruit pectin (without plasticizer) and 1628 cm⁻¹ for bioplastic of dragon fruit peel with ethylene glycol, which indicates that the sample contains pectin group. This follows with the FTIR results for pure pectin, which shows absorption at 1625–1604 cm⁻¹. The presence of the C-O stretching group at 1098 cm⁻¹ in bioplastic samples from dragon fruit peels (without plasticizers) and bioplastics from dragon fruit peel with ethylene glycol shows that the samples contain pectin groups. This is consistent with the results of pure pectin FT-IR, which shows adsorption at 1098 cm⁻¹. The results of the three samples follow previous studies [22, 23, 37].

The addition of ethylene glycol in pectin bioplastics of dragon fruit increases the intensity of the carboxyl - OH functional group [37, 38]. The FT-IR spectrum of dragon fruit pectin bioplastics without plasticizer or ethylene glycol has the same wavenumber as the bioplastic FT-IR spectrum for pure commercial pectin. Agustin and Padmawijaya [39] reported that the bioplastic FT-IR spectrum has the same wavenumber as the FT-IR of the bioplastic's main ingredient, namely pectin. Bioplastics' making by the addition of chemical additives (plasticizers) is through a physical mixing process [37, 39, 40]. FT-IR results show no new functional groups; in other words, there is no chemical change, which implies that bioplastic production is merely physical mixing [37].
Table 2. Function groups of dragon fruit peel pectin bioplastic

| Wavenumber (cm⁻¹) | Pure commercial pectin | Bioplastics of dragon fruit peel pectin | Bioplastic of dragon fruit peel pectin and ethylene glycol | Functional Group | References |
|------------------|-------------------------|----------------------------------------|----------------------------------------------------------|----------------|-----------|
| 3383–3256        | 3370–3350               | 3395–3256                              | –OH carboxyl                                              |                | [37]      |
| 2923             | 2930                    | 2936 C-H alkane                        | RCOOR                                                    | [22, 23]       |           |
| 1740             | 1748–1732               | 1734                                   | C-O carbonyl                                             | [22, 23]       | [37]      |
| 1612–1604        | 1636                    | 1628                                   | C-O stretch                                              | [22, 23]       | [37]      |
| 1098             | 1098–1101               | 1098                                   | C-O stretch                                              | [22, 23]       | [37]      |

4. Conclusion

Dragon fruit peels contain pectin, which can be extracted with HCl solution and yield an 11% pectin. Extracted pectin can be used for bioplastic synthesis mixed with ethylene glycol plasticizer. The moisture content of pectin bioplastic of dragon fruit peels is 5.71-12%, while the pectin and ethylene glycol of dragon fruit peels is 2.86-5.71%. FT-IR spectra showed that bioplastics belong to the pectin group, which is indicated by carbonyl absorption in 1636–1628 cm⁻¹ and the C-O stretching group at 1098–1101 cm⁻¹.

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207
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