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Effect of the Addition of Orange Peel Powder on the Physical and Mechanical Properties of Polylactic Acid Cellulose Acetate Film Composites

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Abstract

Conventional plastics are already widely used in society, despite the fact that these materials have been demonstrated to be environmentally hazardous. As a result, renewable polymers generated from nature and biodegradable alternatives to typical plastics are required, one of which is plastic made from polylactic acid (PLA), cellulose acetate (CA), Tween 80, and orange peel powder (OPP). The objective of this study was to determine the best ratio of OPP added to PLA/CA with the compatibilizer Tween 80 that could improve the physical and mechanical properties of the composite film as well as its antibacterial activity. The method used is solvent casting with a mass ratio of PLA/CA 6:4 with the addition of Tween 80 2% v/v and the addition of OPP, with variations of 2, 4, 6, and 10% w/w, to the weight of PLA. The product was then characterized by FTIR, SEM, XRD, DSC-TGA, mechanical tests (tensile strength and elongation at break), swelling tests, and antibacterial tests. The results showed that the PLA/CA/OPP film composite has a fairly smooth surface with a small cavity, capable of degrading to a temperature of 546.57°C. Regarding the mechanical properties, the best film composites were identified at a variation of 6% w/w with a tensile strength of 4.05 MPa, elongation at break of 11.2%, and a fairly small water absorption. However, this variation has no antibacterial activity.

Keywords: cellulose acetate, composite film, orange peel powder, PLA, solvent casting

1. Introduction

Conventional plastic is still the main packaging material in the food industry because of its characteristics and properties, which include being lightweight, resistant to water, having the potential to be a good insulator, being anti-corrosion, and being low in price. Data submitted by the Indonesia Packaging Federation (IPF) in 2020 stated that the use of plastic in Indonesia as a packaging material in food is 44 percent. This was also conveyed by the Central Statistics Agency (BPS) in 2021: the plastic waste in Indonesia reached 66 million tons per year, with most of the waste generated by food packaging plastic. The use of conventional plastics is already very high, adversely affecting the environment and the economy. Conventional plastics take a very long time to decompose, causing a buildup of waste that pollutes the environment. Efforts to reduce this problem include the search for biodegradable alternatives to conventional plastic for food packaging, such as bioplastic.

Natural polymers that have potential as bioplastic raw materials are polylactic acid (PLA) because they are biodegradable, biocompatible, non-toxic, easy to process, and environmentally friendly (Gomaa et al., 2017). PLA, however, has disadvantages, such as not being able to withstand high temperatures, a slow crystallization process, and being brittle and fragile, which affect its use as a food packager. One way to improve PLA's mechanical properties is to combine it with other types of polymers as fillers (Zuo et al., 2014). A polymer that can be used as a filler and can be combined with PLA is cellulose acetate (CA). CA is a cellulose esterification product (wood, cotton, and rice husks) that can be utilized as a reinforcing filler since it is biodegradable, non-toxic, thermoplastic, hydrophilic, easily soluble in organic solvents, has good chemical and mechanical stability, and is inexpensive to produce (Ali & Hameed, 2022). The formation of homogenous films in PLA and CA composites is problematic due to differences in characteristics between PLA (hydrophobic) and CA (hydrophilic). The inclusion of compatibilizer agents can increase the homogeneity of PLA and CA composites. A compatibilizer is a substance that, through intramolecular bonding, may unite incompatible polymers into a stable mixture.

The synthesis of PLA/CA film composites utilizing maleic anhydride (MA) compatibilizers yielded the best film composite mechanical properties at the addition of 1.5% MA, with tensile strength values of 7.38 MPa and elongation at break of 18.11%, according to Kholiq et al., (2022). Witri (2021) manufactured PLA/CA film composites with the addition of Tween 80 compatibilizer to produce the best film composite mechanical properties with the addition of Tween 80 2%, with a tensile strength value of 9.40 MPa and elongation at break of 23.06%. This demonstrates that using Tween 80 as a compatibilizer can improve PLA/CA mechanical qualities more than MA.

The disadvantage of adding compatibilizer to the resulting composite film when used as food packaging is its lack of antibacterial properties. Antibacterial characteristics can reduce the activity of microbes, thus extending the shelf life of food. Chitosan (Garavand et al., 2020), essential oils (Khazaei et al., 2021), and orange peel powder (OPP) (Terzioğlu et al., 2021) are examples of natural antibacterials utilized in film composites, whereas ZnO, MgO, and TiO_2 metals (Ghozali et al., 2020) are examples of unnatural antibacterials. These materials have a good ability to suppress microbial development, but they must be improved to produce a film composite that meets packaging material specifications and improves food shelf life.

Natural antibacterial agents are safer to use in food packaging since they are non-toxic and environmentally benign, allowing them to be employed as additives in food packaging systems (Shao et al., 2022). OPP, a natural chemical that contains phenol groups, has antioxidant and antibacterial properties. In the previous research, antioxidant content was tested (Terzioğlu et al., 2021), but no antibacterial tests were carried out. Based on the above description, research on the synthesis of PLA/CA/Tween 80 film composites with the inclusion of natural components in the form of OPP is required to produce packaging materials with good physical and mechanical qualities as well as antibacterial capabilities. This study's findings are expected to provide an alternative method for reducing the usage of traditional plastics in food packaging. The goal of this study is to estimate the concentration of OPP required to manufacture composite films with good physical, mechanical, and antibacterial qualities in accordance with packaging regulations.

2. Methodology

2.1. Materials

Several materials will be employed in this research, including distilled water, acetone (Merck), OPP (a *commercial product by* Ryou Indonesia), CA, glycerol, chloroform, PLA, and polysorbate 80 (Tween 80).

2.2. CA modification with Tween 80

The mass ratio of 6:4 is used in the production of PLA/CA composites. CA powder is dissolved in acetone at a 1:10 w/v ratio and swirled with a magnetic stirrer at a steady speed (400 rpm) until homogenous. The plasticizer, glycerol (10% v/v of CA), and the compatibilizer Tween 80 with variations (2% v/v) are then added to the CA solution. Mixing is carried out at a temperature of 50°C for 2 hours so that a homogeneous and viscous CA solution is formed.

2.3. Synthesis of PLA/OPP

Ten grams of PLA were dissolved in chloroform at a 1:10 w/v ratio, and OPP was dissolved in 10 mL of chloroform and then dissolved into the PLA solution at 2, 4, 6, and 10% w/w variations. To obtain the dispersed phase, the PLA solution and OPP were sonicated using ultrasonic waves for 30 minutes.

2.4. Synthesis of PLA/CA/Tween 80 Film Composite with the addition of OPP

The homogenous Tween 80 modified by CA solution was slowly injected into the PLA solution and OPP using a magnetic stirrer at a rotating speed of 1500 rpm at 50°C for two hours. The PLA/CA/Tween 80 solution with OPP was then irradiated for 30 minutes using ultrasonic waves. The solvent casting method is used to create film composites by pouring a suspension into a 9-cm-diameter petri dish to form a thin layer (film) with a maximum thickness of 5 mm. Then it was dried for 24 hours at ambient temperature (25-30°C). The dried film composite can then be taken out of the petri dish and subjected to the characteristic tests.

2.5. Characterization of Film Composite

Composite film obtained is characterized by the following methods:

2.5.1. Functional Group

Analysis of functional groups in film composites was carried out using FTIR (Perkin-Elmer UATR Spectrum Two, USA). The films with a size of 1x1 cm were dried in an oven at 40°C for 2 hours. Furthermore, the films were left at room temperature for approximately 2 hours, and the absorbances of the infrared spectrum were recorded between wavenumbers 500 and 4000 cm^{-1}.

2.5.2. Morphology

The composite film morphology test was carried out through SEM (SEM-EDX JEOL JSM-6510LA, USA) with a diameter of 0.5 cm at a 20 kV voltage. The surfaces of composite films were mounted on metal channels and then coated with osmium powder for 30 minutes. The samples were then mounted on SEM imaging and photographed at 5,000 and 10,000x magnification.

2.5.3. Crystallinity

The degree of crystallinity of the composite film was tested using XRD (PANalytical AERIS), with a sample size of 3x3 cm. The analysis was conducted in a range of 2θ angles (10–80°), at a voltage of 40 kV and current of 30 mA. X-ray radiation wavelength was 1.5406 Å. The crystallinity degree was calculated as the ratio between the area corresponding to the crystalline phase and the total area under the XRD curve using the equation 1 (Rahmayetty & Sulaiman, 2023).

$$C=\frac{\text{Crystalline area (5-80°)}}{\text{Total area (5-80°)}} \quad (1)$$

2.5.4. Thermal Degradation

A thermal degradation test on composite films was carried out using DSC-TGA (NEXTA STA, Hitachi STA200RV, Japan) with a sample size of 0.5 cm. The DSC test was conducted based on ASTM standard D3418-08, using Perkin-Elmer DSC-6, and the TGA test was conducted based on Perkin Elmer's TGA 7. The samples were weighed at around 8.78 mg, then placed and sealed in an aluminum pan. The samples were heated until 550°C. The heating rate for DSC-TGA test was 10°C/min, and the flow rate of nitrogen was 100 ml/min. The glass transitions and the melting temperatures of the material were recorded.

2.5.5. Mechanical Properties

The tensile strength mechanical properties test was performed using the Tensile Strength Tester UT-3500 with a sample size of 5x1 cm by pressing the sample on a testing machine. The thickness and initial length of the sample were recorded. Then the sample was withdrawn at a speed of 1 mm/minute until the sample broke. The elongation value is calculated from the comparison between the gap distance at break and the initial length of the sample and is expressed as a percentage (%).

2.5.6. Swelling Characterization

The swelling test was carried out by soaking a composite film sample, 4 cm in diameter, in a petri dish filled with water for 2 hours and then weighing the final sample after soaking. Samples were weighed as the initial weight (W_0), then the sample was soaked in distilled water for 2 hours, then weighed again to obtain the final weight (W). The swelling test can be calculated using the equation 2.

$$\text{Swelling (\%)}=\frac{W-W_0}{W_0} \quad (2)$$

2.5.7. Antibacterial Characterization

The antibacterial test was carried out using the diffusion method by placing a sample of composite film with a diameter of 5 mm into a petri dish that had been smeared with *E. coli* bacteria and then storing it at room temperature for 24-48 hours.

3. Results and Discussion

3.1. Film Composite's Functional Groups

Figure 1 shows an image of the FTIR test spectrum pattern produced from a PLA/CA composite sample with a Tween 80 compatibilizer. The composite of PLA and CA, based on the spectral pattern, makes a good and strong bond between the components by gaining the same binding and not establishing new bonds.

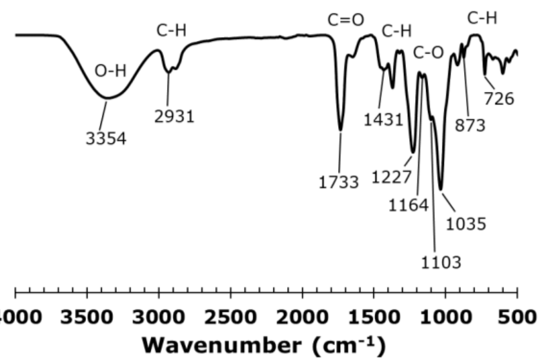

Figure 1. PLA/CA and Tween 80 film composite FTIR pattern

Table 1 depicts the number of strong interactions and good dispersion between components; changing the arrangement increases the peak of functional group bonds (Rihayat et al., 2020; Popa et al., 2017).

Table 1. Functional groups of PLA/CA and Tween 80 film composite

Group	Wavenumber cm^{-1}	
	This Research	Literature
O-H	3354	3294 (Hakim, 2018)
C-H	2931	2897 (Suryani et al., 2018)
C=O	1733	1735 (Ren et al., 2012)
C-H	1431	1454 (Lasprilla et al., 2011)
C-O	1227	1220 (Hadinata et al., 2019)
	1164	1168 (Ikhtiarini et al., 2022)
	1103	1103 (Witri, 2021)
	1035	1030 (Witri, 2021)
C-H	873	870 (Hakim, 2018)
	726	755 (Hakim, 2018)

The addition of OPP to the PLA and CA films is designed to improve the qualities of the final film. Antibacterial film composites are applied in 2%, 4%, 6%, and 10%, each of which has an effect on the intensity of infrared absorbance from the specific functional group in the film composite, as shown in Figure 2.

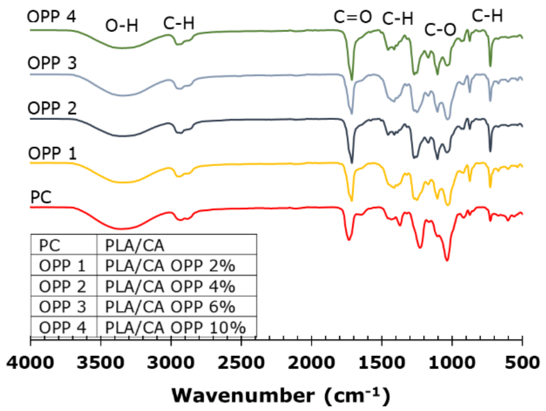

Figure 2. PLA/CA film composite FTIR pattern with the addition of orange peel powder

Table 2 shows the FTIR test spectral pattern produced from a PLA/CA composite sample with a Tween 80 compatibilizer and OPP addition, which has the same peak as a PLA/CA film composite sample.

However, other peak changes, such as an increase in peak intensity in the O-H group, suggest that secondary forces, such as strong hydrogen bonds, are developing between PLA, CA, and OPP, as seen in Table 2. According to Pan et al., (2021), the C=O ester functional group in OPP is defined by a peak intensity in wavenumber 1734 cm^{-1}. As a result, the intensity of peaks decreases, indicating an increased interaction of PLA/CA and OPP (Lan et al., 2020). Similarly, some C-H and C-O groups exhibit intensity variations due to the responses of PLA and CA when OPP is applied (Terzioğlu et al., 2021).

Table 2. Functional groups of PLA/CA/Tween 80 film composite with the addition of OPP

Group	Wavenumber cm^{-1}				
	(Terzioğlu et al., 2021)	OPP 1	OPP 2	OPP 3	OPP 4
O-H	3278	3344	3349	3345	3349
C-H	2918	2940	2929	2933	2952
C=O	1734	1713	1712	1713	1712
C-H	1417	1412	1411	1412	1412
C-O	1300-	1250	1252	1248	1252
	1000	1169	1169	1169	1168
		1104	1103	1104	1103
C-O	1034	1030	1034	1033	1030
C-H	842	873	873	874	874
	727	728	727	727	727

3.2. Film Composite's Morphology

The resultant sample is made up of PLA, CA, and Tween 80, as well as a combination of PLA and CA with OPP, as seen in Figure 3. Figures 3 (a) and (b) show that the cavities on the surface of the final film composite are smaller and narrower after the inclusion of OPP variants.

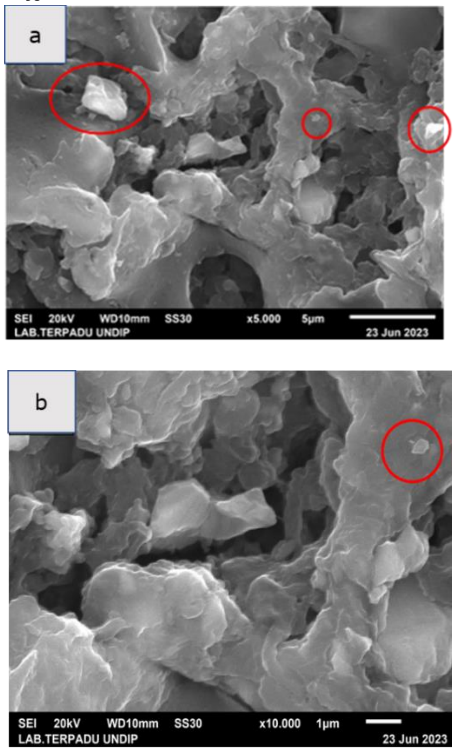

Figure 3. SEM PLA/CA/Tween 80 and OPP film composite test results with magnification of (a) 5,000x, (b) 10,000x

Terzioğlu et al. (2021) claim that OPP does not dissolve entirely in polymer solutions. As a result of the insoluble particles on the surface of the composite film, numerous clumps form. Thus, the morphology of this OPP variation seems slightly hollow, and OPP adheres to the PLA/CA matrix.

3.3. Film Composite's Crystallinity

The XRD test results are shown in Figure 4. Figure 4 depicts some of the highest diffraction peaks obtained. In general, the higher the material's crystallinity, the higher the peak formed will be, which means the pure crystal's peak is sharper (Yuant, 2016).

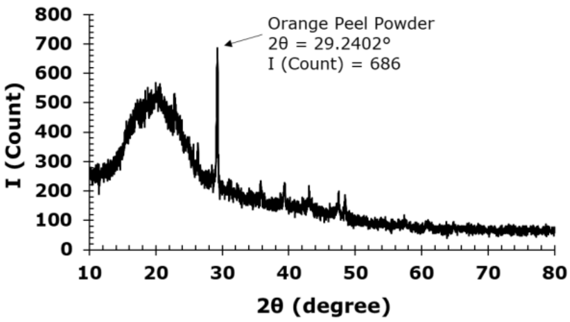

Figure 4. X-Ray Difraction (XRD) test results on PLA/CA film composite with the addition of OPP

According to Witri, 2021, PLA has peaks at an angle of 2θ, which are 23.11°, 28.65°, 29.44°, 35.95°, and 39.45°. The diffraction pattern of PLA/CA OPP is relatively comparable to the previous study, with the peak at an angle of 2θ = 29.2402° and an I (count) of 686. By comparing the XRD test findings to earlier studies, it can be concluded that PLA and CA were successfully composited. According to Pamela et al. (2016), the crystalline composition of the resultant film composite affects the tensile strength response. The crystallinity value of the film composites containing OPP was 22.8221%.

3.4. Thermal degradation properties of film composites

The DSC-TGA test results are shown in Figure 5. Figure 5 (a) shows the trend of increasing energy or heat capacity with incre nnasing temperature. At 49.50 minutes, the highest peak addition of OPP with 10% heat capacity was 8827.16 μW/mg at a temperature of 534.86°C. According to the findings of Zhao et al. (2013), the largest peak in the DSC test graph indicates the presence of melting in the sample. In this situation, as the melting process begins, the sample's energy or heat capacity decreases. The composite film was heated for 52 minutes, yielding a heat energy capacity of 1174.73 μW/mg and a final temperature of 546.57°C.

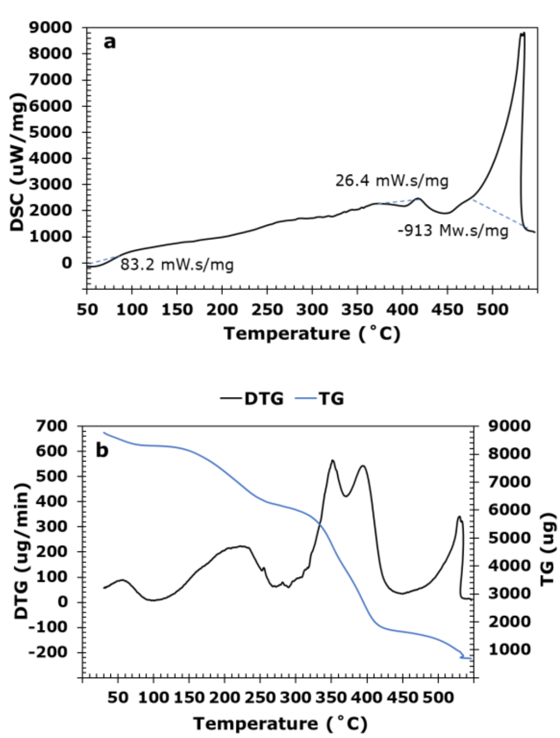

Figure 5. Test results (a) DSC and (b) TGA PLA/CA film composite with the addition of OPP

After 52 minutes and reaching the final temperature of 546.57°C, the sample weight decreased to 692.22 μg, or 0.692 mg. According to Terzioğlu et al. (2021), bio composite films containing OPP will decompose in four phases at a temperature of 35-240°C due to the evaporation of water that is absorbed physically and then chemically. The polymer chain is then thermally degraded. Furthermore, the final disintegration of the combined components occurs at temperatures ranging from 250 to 420 °C. According to the DSC-TGA test results, the PLA/CA/Tween 80 film composite with the presence of OPP has thermal stability and can degrade pretty well through a substantial reduction in sample weight with a comparison of data from previous investigations.

3.5. Mechanical Properties of Film Composites

PLA material films have higher tensile strength and break elongation features in general. The addition of CA as a filler to the PLA matrix reduces tensile strength and lengthens the breaking of the film. The mechanical properties of film composites will be degraded because hydrophobic polymer matrices are poorly matched to CA's hydrophilic qualities. As a result, the inclusion of Tween 80 as a compatibilizer and glycerol as a plasticizer enhances mechanical properties and lowers tension in film composites while also offering good dispersion (Paramitha & Sitompul, 2020). The composite mechanical characteristics of the modified PLA/CA film Tween 80 resulted in a 2.77 MPa improvement in tensile strength and a 9.5% increase in elongation at break.

Table 3. Mechanical properties of the PLA/CA film composite with the addition of orange peel powder

No.	OPP (%b/b)	Tensile Strength (MPa)	Elongation at Break (%)
1.	0	2.77	9.5
2.	2	2.08	8.8
3.	4	2.78	9.5
4.	6	4.05	11.2
5.	10	3.3	10.0

OPP was added to the PLA/CA composite film as a filler to improve the composite qualities of the film as a food packager. Table 3 shows the tensile strength and elongation results obtained from each variation: 2%, 4%, 6%, and 10% of OPP. Mechanical qualities improve as more OPP is applied to the PLA/CA film combination. However, when 2% (v/v) OPP was added, the mechanical characteristics of the PLA/CA composite film decreased due to a decrease in the traction force between chains in the polymer matrix (Terzioğlu et al., 2021).

The addition of 6% OPP resulted in the greatest improvement in mechanical characteristics, with a tensile strength of 4.05 MPa and elongation at break of 11.2%. Based on these findings, adding OPP to the PLA/CA composite improves its mechanical properties by boosting the resulting tensile strength and breaking point. These findings are reinforced by research conducted by Kevij et al., (2021), who found that adding OPP to the film improved the mechanical properties of the accomplished film. This is due to the presence of natural fibers in OPP, which can improve tensile strength and elongation qualities in film composites. The fibers serve to inhibit relative movement of the film, which boosts tensile stress resistance (Leites et al., 2021; Fadzil & Othman, 2021).

Furthermore, the PLA matrix has a CA filler, which is a polysaccharide. Therefore, it interacts well with OPP, which is a good source of complex polysaccharides. As a result, it can create intramolecular connections with CA in PLA, increasing the strength and strain resistance of the resultant film (Kevij et al., 2021; Adilah et al., 2018). However, the addition of 10% OPP (v/v) reduced tensile strength and break length. This is due to the fact that OPP has an optimal mass for inclusion in the PLA/CA film composition. Excess OPP addition weakens the tensile tension between chains in the polymer matrix, which in the film forms additional hydrogen bonds between PLA, CA, and OPP, lowering the mechanical characteristics of the film composite (Terzioğlu et al., 2021).

3.6. Film Composite's Absorption Properties

Water absorption tests are performed to assess how much water the material absorbs. Bioplastics have the property of readily absorbing water. The ability of bioplastics to absorb water is best compared to their resistance to water; the higher the absorption capacity of bioplastics, the lower their resistance to water. It will lower the qualities of bioplastics that are readily damaged due to their high absorption (Coniwati et al., 2014). The goal of composite PLA with CA is to increase the malleability and strength of the PLA's hydrophilic characteristics, thereby enhancing the strength of the PLA matrix as a film. Tween 80 compatibilizer and glycerol plasticizer increase the material absorption quality of the resulting film. The mixture of hydrophilic and hydrophobic polymers improves degradation qualities while not reducing brittleness (Satriyatama et al., 2021; Rahmayetty et al., 2018). As a result, the percentage of positive results obtained from the swelling test after 2 hours is 11.804%.

Table 4. Absorption properties of PLA/CA film composite with the addition of orange peel powder

No.	OPP (%w/w)	Absorption (%)
1.	0	11,804
2.	2	8,754
3.	4	5,46
4.	6	2,326
5.	10	2,341

Table 4 demonstrates that adding OPP to the Tween 80 modified PLA/CA film composite improves the film's water absorption capabilities greatly. The more powder added to the film composite, the less absorption happens in the film. OPP has a reasonably strong water resistance and certainly has an effect on the composite of hydrophobic and hydrophilic components, which influences the film's absorption capabilities (Kevij et al., 2021; Nur Hanani et al., 2018). Research conducted by Leites et al. (2021), supporting this statement that the decrease in water absorption qualities is produced by hydrogen bonds generated between the PLA/CA matrix and the bioactive chemicals of OPP, which limit the ability to absorb water and contribute to the film's degrading properties. The best results achieved in the samples with the appropriate addition of OPP were 6% and 10%, resulting in absorption for 2 hours of 2.326% and 2.341%.

3.7. Antibacterial Properties of Film Composites

The addition of OPP to the PLA/CA film solution aims to improve film quality and limit bacterial development in food packaging. The inhibitory impact is calculated based on the clear zone that circles the film disc. Orange peel extract, according to (Kanmani & Rhim, 2014), has a strong antibacterial action against gram-negative bacteria (*E. coli*). Samples containing OPP were tested for antibacterial activity against gram-negative bacteria (*E. coli*). Figure 6 depicts the antibacterial activity of PLA/CA film composites without and with antibacterial additions.

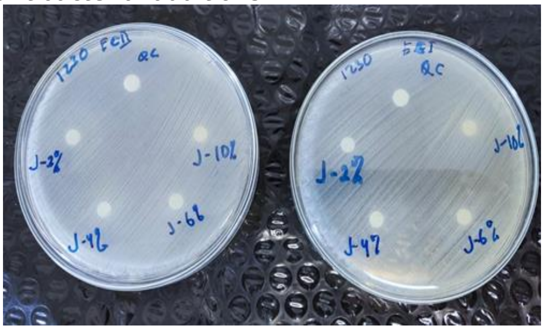

Figure 6. Antibacterial activity of PLA/CA composite film with addition of 2%, 4%, 6%, and 10% OPP

Figure 6 shows the absence of an inhibitory zone between the PLA/CA/Tween 80 film composite and the inclusion of various natural antibacterial substances, OPP, and scratches caused by the addition of *E. coli* bacteria. Several factors, including the inhomogeneous distribution of OPP particles in the film composite solution, confirm the absence of an inhibitory zone between bacteria and film composite samples. A modest sample size is used in this test. The lack of an inhibitory zone in the antibacterial test is most likely owing to the uneven distribution of OPP particles in the test sample. Thus, antibacterial agents are unable to penetrate into the bacterial cell wall. Therefore, no antibacterial activity was identified. Additionally, according to Zhang et al., (2017), particle size or particle distribution of OPP, form or morphology, and change of the composite surface of the film can all play a role in its antibacterial efficiency. Furthermore, the inhibitory zone, including the type of metal oxide, composition, and method of growth media, can be affected because the method used must be adjusted to the type of material tested for antibacterial activity and the physicochemical properties of the antibacterial agent (Nafchi et al., 2012).

4. Conclusion

The addition of OPP to the PLA/CA/Tween 80 composite film increases the physical and mechanical properties, with the best results obtained at a 6% (w/w) OPP ratio. With 6% OPP (w/w), the tensile strength is 4.05 MPa, the elongation at break is 11.2%, and the water absorption is 2.326%. According to the DSC-TGA test results, the composite film PLA/CA/Tween 80 with OPP inclusion is also thermally degradable. An antibacterial test employing the barrier zone method for PLA/CA composite film with OPP yielded no barrier zones. However, from the results obtained, the composite film of PLA CA OPP can be an alternative solution to reduce the use of conventional plastic in food packaging.

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