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The Effect of Glycerol as Plasticizer on Density and Water Uptake Capacity of Polystirene-Polycaprolactone Bioblend Plastic Film

Rika Sari Lalfari[1], Syukri Arief[2] and Akmal Djamaan[3*]
[1]*Dept. of Chemistry, Fac. of Sciences, Universitas Andalas, Padang, Indonesia*
[2]*Dept. of Chemistry, Fac. of Sciences, Universitas Andalas, Padang, Indonesia*
[3]*Dept. of Pharmaceutical Chemistry, Fac. of Pharmacy, Universitas Andalas, Padang, Indonesia*

Abstract
Background: *Polystyrene (PS) in the form of foam known as styrofoam, is widely used as a wrapping material for food, beverages, medicines and even as protective materials for electronic devices. However, styrofoam wastes are very difficult to decompose in nature and has caused some serious damages to environmental ecosystem. The purpose of this research was to study the effect of using glycerol as a plasticizer on the density and water uptake capacity of plastic film in the form of PS bioblend by mixing it with a biodegradable polymer, which was polycaprolactone.*
Methods: *The density were calculated by dividing the weight of the plastic film by its volume and the water uptake capacity which were determined by using the swelling test method.*
Results: *The results showed that the addition of 0.1-0.4 ml of glycerol into the PS-PCL bioblend mass affected the density and water uptake capacity of the resulting plastic film layer. In the PS-PCL bioblend plastic film (99-1% w/w) the density increased from 1.12-1.28 g /cm^2 in addition of 0.1-0.3 ml glycerol and decreased in value to 0.95 g / cm^2 with the addition of 0.4 ml glycerol. Likewise, the water uptake capacity increased from 0.23-0.45% w/w in the addition of 0.1-0.3 ml glycerol and decreased in value to 0.32% w/w in the addition of 0.4 ml glycerol. In the PS-PCL bioblend plastic film (95-5% w/w) the density decreased from 1.20-1.07 g/cm^2 at the addition of 0.1-0. 3ml glycerol and increased its value to 1.31 g/cm2 at addition of 0.4 ml glycerol. On the other hand, the water uptake capacity increased from 0.21 to 0.40% w/w in the addition of 0.1-0.3 ml glycerol and decreased in value to 0.31% w/w at the addition of 0.4 ml glycerol. On the PS-PCL bioblend plastic film (90-10% w/w) the density decreased from 1.33-1.14 g / cm^2 at the addition of 0.1 ml glycerol, then increased to 1.40 g / cm^2 at the addition of 0.2 ml glycerol and decreased to 1.15 with the addition of 0.4 ml glycerol. The water uptake was decreased from 0.38% w/w to 0.18% w/w with the addition of 0.4 ml glycerol.*
Conclusion: *In general, it can be stated that the addition of 0.1-0.2 ml of glycerol will increase the density value of the water uptake capacity of the PS-PCL bioblend film, and the addition of 0.3-0.4 ml glycerol will reduce the density and water uptake value.*
Key Words: *bioblend, polistiren, styrofoam, glycerol, polycaprolactone*

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I. Introduction

In the last few decades, the use of synthetic plastics has expanded and entered almost all activities of everyday human life. The advantages of the synthetic plastics includes; light, flexible, strong, not easily broken, transparent, water proof and economical. Along with these conditions, the petrochemical-based plastic (polymer) industry has been growing rapidly and due to the increasing demand, the industry has produced more than 300 million tons of plastic every year. The uncontrolled use of synthetic plastics caused serious environmental pollution, because these plastics are not biodegradable in nature. In the United States and Japan it was reported that more than 30 percent of the waste produced is plastic waste. Thousands of tons of plastic waste are dumped into the sea every year, causing pollution and damage to marine life. It is estimated that more than one million of marine animals are die each year as a result of being entangled or ingested the plastic debris in the sea water[1].

Polystyrene (PS) is one type of polymer that is widely used, which is rigid, hard, and has a pure white color. In daily use, polystyrene is known in the form of foam, namely styrofoam. Styrofoam is widely used, including as a material for food wrappers, disposable drinking containers, medicine wrappers, protective materials and vibration barrier for fragile items, such as electronic devices. After using styrofoam, most people just throw it away into garbage. Styrofoam waste is reported to be able to disrupt environmental ecosystem, because Styrofoam cannot decompose naturally. Used polystyrene is a waste that is difficult to recycle, because the cost of treating used polystyrene waste is higher than the price of pure polystyrene[2].

Previous research on the reuse of styrofoam waste, included: the production of bioblend plastic film from polystyrene by mixing it with polycaprolactone (PCL), a biodegradable synthetic polymer. It was also reported that the PS-PCL bioblend can be used as a coating material for urea and NPK granules to produce slow release fertilizer[3]. However, from the previous research it was reported that the plastic film produced from the PS-PCL bioblend still needed to be improved for its physical properties, because the plastic films produced were fragile and less flexible. Bioblend PS-PCL which was used as a coating for urea and NPK fertilizers turned out to be less stable over a long period of time, because the coating cracked and broken in a dry storage environment[4].

Based on these problems, this research has developed a plastic film formula in the form of a PS-PCL bioblend with the use of glycerol as a plasticizer. The addition of this plasticizer is expected to improve the physical properties of the PS-PCL bioblend produced. From the literature study conducted, there have been no reports of the use of glycerol as plasticizer in PS-PCL bioblend plastic film, however glycerol is widely used as plastic film plasticizer for food packaging, such as agar-based edible films[5].

In this article, we reported the effect of glycerol as a plasticizer on the physical properties of PS-PCL bioblend-based plastic film, especially on the density and water uptake capacity (swelling test).

II. Materials and Methods

Plastic films production from the PS-PCL bioblend with the addition of glycerol as plasticizers

The composition for making polymer films from the PS-PCL bioblend with the addition of glycerol as plasticizers is shown in Table 1.

Table 1. The plastic film composition of PS-PCL bioblend with the addition of glycerol as a plasticizer

No.	Sample Code	Composition PS:PCL (% w/w)	PS (g)	PCL (g)	Glycerol (ml)
1.	A0	100:0	2.00	0.00	0.0
2.	A1	100:0	2.00	0.00	0.1
3.	A2	100:0	2.00	0.00	0.2
4.	A3	100:0	2.00	0.00	0.3
5.	A4	100:0	2.00	0.00	0.4
6.	B0	99:1	1.98	0.02	0.0
7.	B1	99:1	1.98	0.02	0.1
8.	B2	99:1	1.98	0.02	0.2
9.	B3	99:1	1.98	0.02	0.3
10.	B4	99:1	1.98	0.02	0.4
11.	C0	95:5	1.90	0.10	0.0
12.	C1	95:5	1.90	0.10	0.1
13.	C2	95:5	1.90	0.10	0.2
14.	C3	95:5	1.90	0.10	0.3
15.	C4	95:5	1.90	0.10	0.4
16.	D0	90:10	1.80	0.20	0.0
17.	D1	90:10	1.80	0.20	0.1
18.	D2	90:10	1.80	0.20	0.2
19.	D3	90:10	1.80	0.20	0.3
20.	D4	90:10	1.80	0.20	0.4

The process was started by carefully weighing the PS and PCL powders according to the formula. Then, it put into a beaker with a volume of 50 ml. Furthermore, 20 ml of chloroform was added and heated on a heating device (hot plate) until it dissolved completely. The glycerol was added according to the formula, then stirred evenly until the volume of the solution become 10 ml. The solution was poured into a glass Petri dish (9 cm in diameter) and shaken until it evenly distributed. It was left for 1-2 days at room temperature (30-35^{o}C), until the plastic film formed in the Petri dishes were released itself from the bottom of the Petri dishes[6].

Then, the plastic film of bioblend PS-PCL was removed, allowed to dry at room temperature, and weighed until a constant weight was obtained. These steps were also carried out for other PS-PCL polymer film compositions. The plastic film composition of the PS-PCL bioblend was prepared by adding glycerol as a plasticizer.

Determination of the density of PS-PCL bioblend plastic film

The plastic film samples made from the PS-PCL bioblend were cut to the size of 6x1 cm for 3 specimens. The thickness of plastic film was measured using a Digital Micrometer (Krisbow KW06-85) at 3 (three) different places around the plastic film and the average was calculated. To determine the density value, each sample was weighed using a digital scale, its height or thickness was measured using a digital caliper. Then the volume value was calculated from the length, width and height data. The density value was calculated by dividing the weight of the sample by the volume according to Equation-1 below[5]:

$$\text{Density (g/cm}^3\text{)} = \frac{\text{Weight (g)}}{\text{Volume (cm}^3\text{)}} \qquad \text{(Equation-1)}$$

Swelling test of the PS-PCL bioblend plastic film

The PS-PCL bioblend plastic film of each composition was cut into 2 x 1 cm sizes, then weighed initially (W0). Furthermore, the plastic film was immersed in water for 10 seconds. Then, the plastic film was removed from the water, dried in a desiccator until a constant weight (W1) was obtained. The water uptake by the plastic film was calculated by equation-2, as follows:

$$\text{Water uptake (\%)} = \frac{W_1 - W_o}{W_1} \times 100 \qquad \text{(Equation-2)}$$

where : W_o = initial weight of plastic film
W_1 = sample weight after put into water and dried in a dessicator

The percentage of water uptake of each PS-PCL bioblend plastic film composition, with or without the addition of glycerol as a plasticizer, was determined. Each measurement sample was repeated 3 times[5].

III. Results and Discussion

In this experiment, a thin, clear, slightly bluish-white plastic film with a smooth and flat surface has been produced. In general, the physical form or morphology of all the films produced did not show any significant differences in the shape and morphology of the plastic films produced. The plastic film was dried and rinsed using a tissue to remove the remaining glycerol that adheres to the surface of the resulting plastic film. The plastic films from the PS-PCL bioblend added with glycerol as plasticizers were made in the laboratory using the solvent casting method. The morphology of the formed plastic films were as shown in Figure 1, which was in the form of a thin film, with a clear whitish color, smooth and shiny surface. It has observed that, there is a slight difference in the appearance of the plastic film with added glycerol (B) compared to the plastic film without glycerol (A). The plastic film added with glycerol plasticizer look smoother and more flexible.

The technique of making plastic films using this solvent casting method has been developed because it is relatively simple and does not require a lot of equipment. The manufacture of small-scale plastic films using this method can use glass Petri dishes, in which the hot and molten plastic mass was poured out and allowed to solidify and evaporate at room temperature.

However, for large-scale production, Petri dishes cannot be used due to their limited volume and size. At present, at the industrial level, machines based on this solvent casting method have been developed, mainly used for the manufacture of plastic packaging and edible films for food wrapping[7].

A B

Fig. 1. The examples of plastic films made by PS-PCL bioblend without the addition of glycerol (A) and with the addition of glycerol (B).

Beside the solvent casting method, currently in modern industry other methods are also being developed for the manufacture of thin plastic films, including the holt melt extrusion method, semi-solid casting, and rooling solvent casting. The solvent casting method is a method that dissolves all plastic material in a volatile solvent, then the solution is poured and allowed to dry at room temperature or an oven at 40-50°C. The holt melt extrusion method uses the materials in the form of solids, which are homogenized and dried in a screw extruder machine. The temperature in this process is set at 100°C until all the ingredients become liquid, then pressed to form a thin film. Furthermore, the semi-solid casting method uses a polymer that are insoluble in acid. First a water-soluble polymer solution was prepared, then the solution is added to the acid-insoluble polymer. Next, a plasticizer is added between the two processes to form a gel mass. The solution is poured into a mold film with an adjusted thickness. The ratio of water insoluble polymers and acidic polymers in this method was 1:4. Finally, the rooling solvent casting method, which is the process of making a thin plastic film by suspending their constituent materials into water or a water-alcohol mixture and rolling them with a certain tool. Furthermore, the plastic film was dried on a roller and shaped and cut to the desired size[7,8].

Table 2. The measurements results of the density and water uptake of bioblend PS-PCL plastic film made in various compositions, with and without the addition of glycerol plasticizer

No.	Sample Code	Composition PS-PCL (% w/w)	Glycerol (ml)	Density* (g/cm^2)	Water uptake* (% w/w)
1.	A0	100-0	0.0	0.14	0.07
2.	A1	100-0	0.1	0.81	0.20
3.	A2	100-0	0.2	1.17	0.35
4.	A3	100-0	0.3	1.17	0.53
5.	A4	100-0	0.4	0.83	0.16
6.	B0	99-1	0.0	1.11	0.26
7.	B1	99-1	0.1	1.12	0.23
8.	B2	99-1	0.2	1.18	0.34
9.	B3	99-1	0.3	1.28	0.45
10.	B4	99-1	0.4	0.95	0.32
11.	C0	95-5	0.0	1.20	0.00
12.	C1	95-5	0.1	1.08	0.21
13.	C2	95-5	0.2	1.11	0.31
14.	C3	95-5	0.3	1.07	0.40
15.	C4	95-5	0.4	1.31	0.31
16.	D0	90-10	0.0	1.33	0.00
17.	D1	90-10	0.1	1.14	0.38
18.	D2	90-10	0.2	1.40	0.18
19.	D3	90-10	0.3	1.23	0.18
20.	D4	90-10	0.4	1.15	0.18

*Data was the average of 3 replications

Table 2 shows the results of measurement of the density and water uptake capacity of the PS-PCL bioblend-based plastic films with various composition ratios, without and with the addition of glycerol plasticizers. Sample A0-A4 was a plastic film made from PS with a content of 100% (w/w), without and with the addition of glycerol as plasticizer, the density varies between 0.14-1.17 g/cm^2. The percentage value of water uptake of A0-A4 samples ranged from 0.07-0.53% (w/w). Meanwhile, sample B (PS-PCL composition 99-1% w/w) had a percentage value of water uptake between 0.23-0.45% (w/w), sample C (PS-PCL composition 95-5% w/w) had a water uptake percent value of 0.00-0.41% (w/w) and sample D (PS-PCL composition 90-10% w/w) had a water uptake percent value of around 0.00-0.38 % (w/w).

In Figure 2, the effect of the amount of glycerol added on the density of the plastic film made from the PS-PCL bioblend was shown. In general, this increase in density value lasts until the volume of added glycerol was 0.3 ml, after which it tended to decrease, except for sample C4 which occured a slight increase in the volume of glycerol by 0.4 ml. This was due to the possibility of a slight inaccuracy of the sample weighing data. However, the overall trend for all other samples were to increase in density until the addition of glycerol reached 0.3 ml, afterwards there was a slight decrease.

According to Birley *et al.* (1988), determining the density of plastic film layers is very important, because this value can indicate the structure of plastic in general. Its application is the ability of plastics to protect products from several substances such as water, O_2 and CO_2. Plastics with low density value indicate that the plastic has an open structure, meaning that it is easy to penetrate or can be penetrated by water, O_2 or CO_2. In contrast to paper, the value of plastic density is very important in determining the properties of plastic associated with its use. In commerce it is possible to use the units of grams, because these units are sufficient to represent the molecular weight and area of the plastic[9].

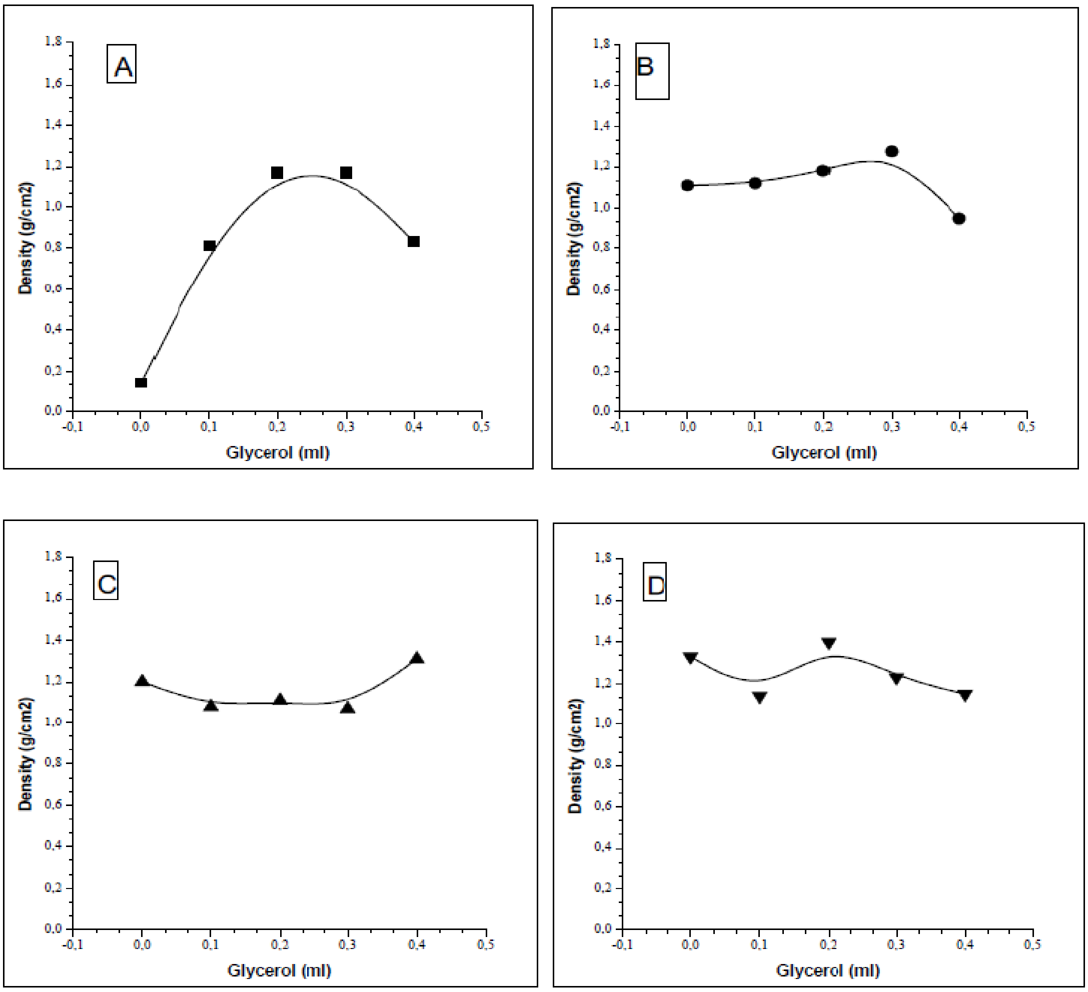

Fig. 2. The effect of adding glycerol as plasticizer on density values of plastic film made from bioblend PS-PCL with various composition ratios : PS-PCL 100-0 % w/w (A), PS-PCL 99-1 % w/w (B), PS-PCL 95-5 % w/w (C), PS-PCL 90-10 % w/w (D)

Table 2 also shown the results of the water uptake (swelling test) of plastic films made from bioblend PS-PCL with the addition of glycerol as a plasticizer. The purpose of this swelling test was to determine the water uptake capacity of the plastic film that has been made. Theoretically, a lower percentage of swelling of a plastic film means low water uptake, which means there is a high possibility that the plastic can be used as a wrapper or packaging for aqueous materials. According to Fathanah *et al.* (2017), the greater the water uptake capacity of a plastic film, the less plastic is able to protect the product from water, which can cause the product to quickly break down or degrade in quality[10].

The effect of adding glycerol on the water uptake capacity of the bioblend plastic film tested was clearly shown in Figure 3. The water uptake value of plastic films made from 100% PS or PS without the addition of glycerol or other polymers, was 0.07% (w/w), a very low water absorption, so that PS is widely used as a packaging for various water-containing products. Meanwhile, if PS was mixed with PCL as a bioblend, there was an increase in the percentage value of water uptake. In samples E1-E4, F1-F4 and G1-G4 there was an increase in the percentage of water uptake to 0.45; 0.40; and 0.38% (w/w). This proves that the water uptake value of the bioblend in this study was also influenced by other polymer components, in this case, PCL.

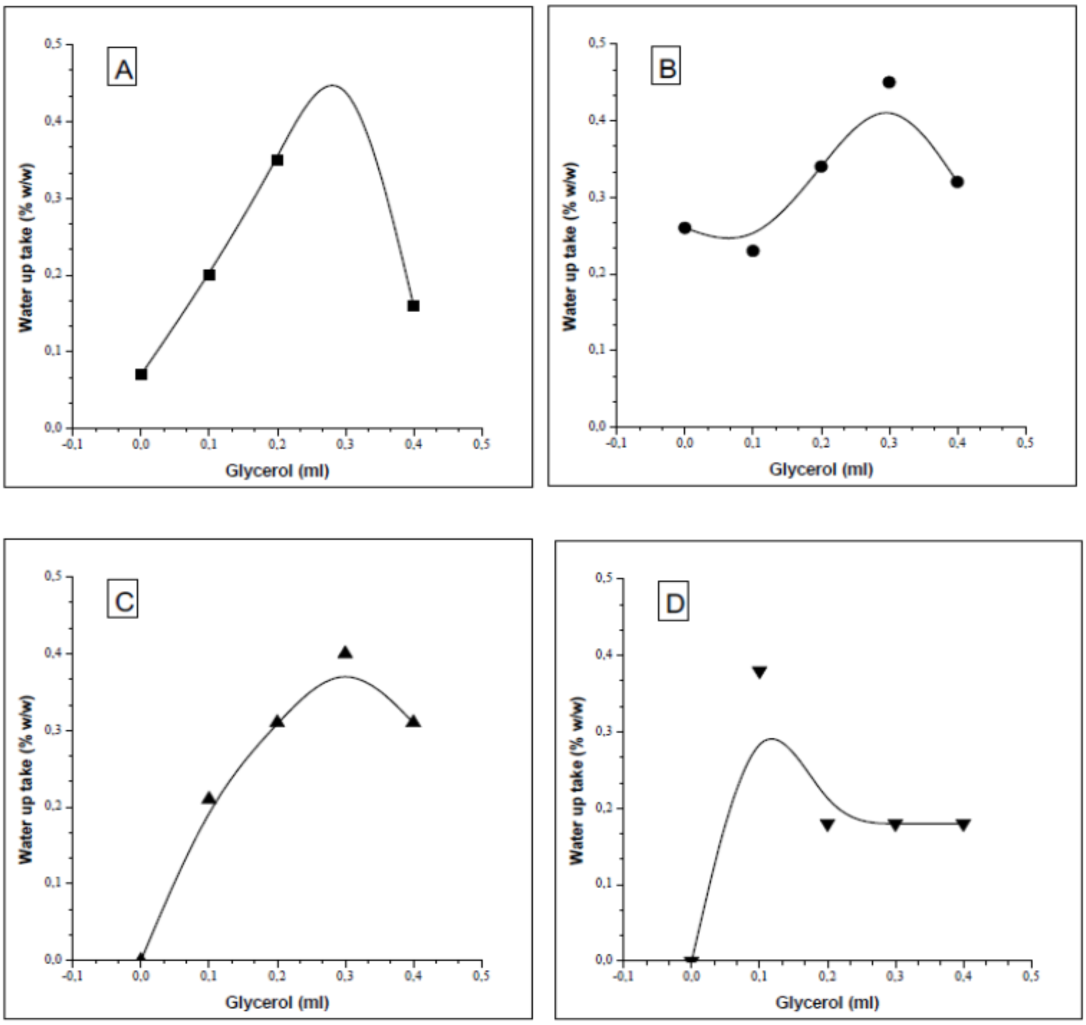

Fig. 3. The effect of glycerol addition as plasticizer on the water uptake capacity of plastic film made from PS-PCL with various composition: PS-PCL 100-0 % w/w (A), PS-PCL 99-1 % w/w (B), PS-PCL 95-5 % w/w (C), PS-PCL 90-10 % w/w (D)

Overall from this study it was found that, on the PS-PCL bioblend plastic film (99-1% w/w) the density increased from 1.12-1.28 g/cm^2 in the addition of 0.1-0.3ml glycerol and decreased in value to 0.95 g/cm^2 on addition of 0.4 ml glycerol. Likewise, the water uptake power increased from 0.23-0.45% w/w in the addition of 0.1-0.3 ml of glycerol and decreased in value to 0.32% w/w in the addition of 0.4 ml of glycerol. In the PS-PCL bioblend plastic film (95-5% w/w) the density decreased from 1.20-1.07 g/cm^2 at the addition of 0.1-0 ml glycerol and increased its value to 1.31 g/cm^2 at addition of 0.4 ml of glycerol. On the other hand, the water uptake capacity increased from 0.21 to 0.40% w/w in the addition of 0.1-0.3 ml of glycerol and the value decreased to 0.31% w/w at the addition of 0.4 ml of glycerol. In the PS-PCL bioblend plastic film (90-10% w/w) the density decreased from 1.33-1.14 g/ cm^2 at the addition of 0.1 ml glycerol, increasing its value to 1.40 g/cm^2 at the addition of glycerol 0,2 ml, and decreased to 1.15 with the addition of 0.4 ml glycerol. Likewise, the water uptake capacity decreased from 0.38% w/w at the addition of 0.1 ml glycerol to 0.18% w/w at the addition of 0.4 ml glycerol.

These results are in accordance with what was stated by Donhowe and Fennema (1993) who stated that the increase in water uptake of a polymer film was also caused by the addition of other large polymer components[11]. In this experiment, PCL is a large compound (macromolecule), because it was a polymer composed of repeating monomers so it has a large molecular weight. The use of glycerol as a plasticizer in this experiment also increased the percentage of water uptake. As shown in Figure 3, it can be seen that there is an increase in the percentage of water uptake starting from 0.1 ml to 0.3 ml glycerol was added and tends to decrease when 0.4 ml glycerol was added. This increase is probably due to the hygroscopic nature of glycerol, so that the increasing volume of glycerol added to the production of bioblend, will of course also increase the amount of water it absorbs during the water swell test. This finding was confirmed by the statement of Grenby *et al*. (1994), who stated that glycerol and polyethylenglycol (PEG) added as plasticizers can cause moisture balance with their own hygroscopic properties[12].

The increase in water uptake capacity of the plastic film tested occured along with the increase in the volume of glycerol added. The exception to the data was the addition of 0.4 ml glycerol which caused a decrease in the percentage of water uptake in some of the test samples. This can occur because the addition of glycerol to the polymer mass of 0.4 ml has passed its absorption capacity, so that some of the glycerol were left in the Petri dishes which will be lost during washing.

An interesting finding in this experiment was that, in fact, the average percent water uptake value of the bioblend studied, are mostly lower in the PS value. The mean of PS water uptake value was 0.26% (w/w), while the mean water uptake rate of samples C and D were 0.24 and 0.18% (w/w), respectively. Only one sample has a percentage value of water uptake higher than PS, which was sample B of 0.32% (w/ w). These data indicated that, compared to PS, the water uptake value of the PS-PCL bioblend was better. The lower the percent deflation of a plastic film, the better it is used for packaging materials and bottles for storing water-containing materials.

This research still needs a further process to obtain a comprehensive characteristics of the bioblend produced, including looking at the effect of glycerol addition on mechanical properties, thermal properties, crystallinity and biodegradation of the PS-PCL bioblend, and will be reported in the next article.

IV. Conclusions

From this research, the following conclusions can be drawn:

1. The addition of 0.1-0.4 ml glycerol to the PS-PCL bioblend mass affected the density and water uptake capacity of the resulting plastic film.
2. On the PS-PCL bioblend plastic film (99-1% w/w) the density increased from 1.12-1.28 g / cm^2 when glycerol is added 0.1-0.3 ml and the value decreased to 0.95 g/cm^2 in the addition of 0.4 ml glycerol. Likewise, the water uptake capacity increased from 0.23-0.45% w/w in the addition of 0.1-0.3 ml glycerol and decreased in value to 0.32% w/w in the addition of 0.4 ml glycerol.
3. On the PS-PCL bioblend plastic film (95-5% w/w) the density decreased from 1.20-1.07 g/cm^2 at the addition of 0.1-0.3ml glycerol and increases its value to 1.31 g/cm^2 on the addition of 0.4 ml glycerol. On the other hand, the water uptake capacity increased from 0.21 to 0.40% w/w in the addition of 0.1-0.3 ml glycerol and decreased in value to 0.31% w/w at the addition of 0.4 ml glycerol.
4. On the PS-PCL bioblend plastic film (90-10% w/w) the density decreased from 1.33-1.14 g/cm^2 at the addition of 0.1 ml glycerol, the value increased to 1.40 g/cm^2 on addition of 0.2 ml glycerol, and decreased to 1.15 with the addition of 0.4 ml glycerol. Likewise, the uptake power of water decreased from 0.38% w/w at the addition of 0.1 ml glycerol to 0.18% w/w at the addition of 0.4 ml glycerol.
5. In general, it can be stated that the addition of glycerol 0.1-0.2 ml will increase the density value of the water uptake capacity of the PS-PCL bioblend film, and the addition of glycerol 0.3-0.4 ml will decrease the density and water uptake value.

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