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Study of the Degradation Rate of Polymeric Biodegradable Composites PLA (Powder) – PCL Produced by Solvent Casting in 0.3% NaCl Medium

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***Abstract* - The treatment of long-standing fractures is influenced by trauma, resorption, and bone pathology. Additional issues arise with the use of non-biodegradable metal implants, requiring additional procedures for removal, increasing the risk of infection, complications, and prolonging recovery time. The solution being studied is the use of biodegradable materials such as Polylactic Acid (PLA) - Polycaprolactone (PCL) polymers for bone repair and fixation, which can reduce the risk of complications and infection while accelerating healing. This research aims to analyze the effect of PLA powder percentage on the degradation rate of PCL mixtures using the solvent casting method. The specimen preparation process in this study utilizes the solvent casting method by mixing PLA and PCL using acetone as a solvent. The PLA-PCL compositions used are 10 PLA: 90 PCL, 30 PLA: 70 PCL, and 50 PLA: 50 PCL. The specimens are then characterized through density and degradation testing by immersing them in a 0.9% NaCl infusion solution. The density testing results indicate that the 50 PLA: 50 PCL composition has the highest density, at 0.97 g/cm³. In degradation testing, the 10 PLA: 90 PCL composition exhibits the highest degradation rate, with a weight loss percentage of 22.01% in the fourth week and a degradation rate of 0.0128 mmpy in the first week.**

Keywords: Biodegradable Polymer, Degradation, Polycaprolactone, Polylactic acid, Solvent Casting.

I. INTRODUCTION

Men and women under 45 years old have a higher risk of fractures. Furthermore, the most common fractures are those located in the upper extremities and vertebrae. For patients experiencing fractures, there are several factors contributing to the prolonged treatment process. Trauma usually causes fractures or bone breaks. If not promptly addressed, fractures can lead to various issues, such as nerve trauma, vascular trauma, bone complications, and bone emboli [1]. The need for new materials with characteristics tailored to meet the biochemical and biomechanical requirements of tissue engineering in bone is the primary motivation in ongoing research to develop biodegradable materials. The basic concept is that replacement biomaterials act as scaffolds for surrounding cells or tissues to infiltrate, grow, and initiate tissue regeneration, leading to the formation of new bone [2].

Biodegradation is a process of breaking down complex compounds into simpler ones, such as water and carbon dioxide [3]. This degradation process alters molecular integrity through microorganism activity. In recent years, composites of polylactic acid (PLA) and polycaprolactone (PCL) have been extensively studied for biomedical applications due to their promising characteristics, including their biodegradability. PLA is a suitable material for implants due to its biocompatibility, biodegradability, and non-toxic nature. One member of the biodegradable aliphatic polymer family is polycaprolactone (PCL) [4].The synthesis process yields PLA polymers. PLA synthesis begins with the production of lactic acid (LA) and then polymerization with the formation of lactide [5]. Polycaprolactone (PCL) is a biodegradable and biocompatible polyester. PCL is considered an ideal material because it is non-toxic, can be absorbed after implantation, and has good mechanical properties [6].As biopolymers, polycaprolactone (PCL) and polylactic acid (PLA) have been widely used in the medical field. One reason is that the United States Food and Drug Administration (USFDA) has designated PLA and PCL as the most researched polyesters due to their ease of processing. However, both still exhibit good crystallinity, mechanical properties, and thermal transitions [7].

The solvent casting method is a process of dissolving active ingredients and additives in a volatile solvent, such as water or ethanol. After the solution mixture is combined, it is formed into a film and dried in an oven at a temperature of 45–50 degrees Celsius. Subsequently, pieces of the solution are cut for packaging and sealing [8].

II. MATERIALS AND RESEARCH METHOD

The materials used in this study include Polylactic acid (PLA), Polycaprolactone (PCL), and acetone solvent. The

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PLA material used in this research is PLA produced by NatureWorksin America, categorized as medical-grade PLA 2002d specifically designed for medical applications. The PCL used is CAPA-6800 produced by Perstop, in granular form with a size of 2-3 mm. The solvent used to dissolve PCL is acetone. The acetone used is pure acetone obtained from PT. Kurnia Makmur Abadi Jaya.

The method used in mixing biodegradable PLA/PCL polymers in this study employs the solvent casting method, also known as solution casting or wet processing technique, which has been favored for years due to its simplicity and the relatively non-specialized equipment required. Essentially, solvent casting is a manufacturing process involving the continuous mechanical mixing of polymer dissolved with fillers. SCPL is a simple and cost-effective procedure that controls porosity and pore size. The achievable pore size and porosity range from 20% to 50%, and between 30 and 300 µm, respectively [9].

The variables used in this study involve variations of the Straight Blade Turbine agitator design with PLA:PCL compositions (10:90, 30:70, 50:50), and the stirring time used per variation is 15 minutes. The specimen preparation stages begin with preparing 5 grams of PLA and PCL materials. Then, PCL is dissolved using 100 ml of acetone, and in the first 5 minutes, the hotplate stirrer temperature is heated to 200°C, then reduced to 150°C, and in the last 5 minutes, it is raised back to 180°C. The solution is poured into prepared Molds. Ensure that the Molds are cleaned and dried before use. Allow the solution to evaporate slowly in the mold; the drying process can be done at room temperature.

Once the specimens are made, material testing is conducted to determine the density value of the test specimens and degradation testing to assess the extent of specimen degradation using the immersion method, aided by macro photos visualizing the physical changes of the specimens each week. After all data from density values and degradation rates are collected, regression analysis is performed on the data using Statistical package software.

The purpose of density testing is to understand the mass density value of a material. This testing can also be done using buoyancy force methods, Archimedes' method, or the pycnometer method. The results of density testing are also used to compare the density values of different materials and to predict physical properties such as strength, stiffness, and dimensional stability [10]. Archimedes' principle states that when an object is submerged in a fluid, it experiences an upward force equal to the weight of the displaced fluid.

Many factors can influence the degradation rate of a specimen, such as physical and environmental factors, concentration factors, and comparison of existing hydrocarbon structures. Additionally, microorganisms have the ability to break down hydrocarbon structures. These factors include: 1) chemical factors related to nutrient availability, absence of growth-supporting compounds, and required enzyme inducers; 2) environmental factors, which relate to extreme physical conditions (such as pH, temperature, and redox potential); 3) microorganism factors related to the absence of populations of pollutant-degrading microorganisms.

Macrographic evaluation is conducted to determine how the specimen's shape changes after testing. Visual photos made using a digital camera with a macro lens are called macro photos. After several tests, macro photos will show whether the specimen has changed.

III. RESULTS AND DISCUSSIONS

Table 1shows the results of density test. The density of the composites is in the range of 0.95% to 0.96%. Figure 1 shows the comparison of density test results and the theoretical density calculation. It can be shows that all of the specimens have density are lower than theorical density. It indicates that all of specimens have porosity. Porosity can occur due to trapped air in the liquid during stirring or pouring into a mold.

Table 1: Density Test Result

Ratio PLA : PCL	Specimen 1		Specimen 2		Specimen 3		Average
	$\rho_{specimen}$ (gr/cm^3)	Ave.	$\rho_{specimen}$ (gr/cm^3)	Ave.	$\rho_{specimen}$ (gr/cm^3)	Ave.	
10 : 90	0,97	0,94	0,97	0,96	0,97	0,94	**0,95**
	0,95		0,94		0,90		
	0,89		0,98		0,94		
30 : 70	0,95	0,95	0,99	0,96	0,95	0,96	**0,96**
	0,97		0,94		0,95		
	0,93		0,96		0,98		
50 : 50	0,98	0,96	0,98	0,97	0,98	0,98	**0,97**
	0,91		0,98		0,98		
	0,98		0,95		0,97		

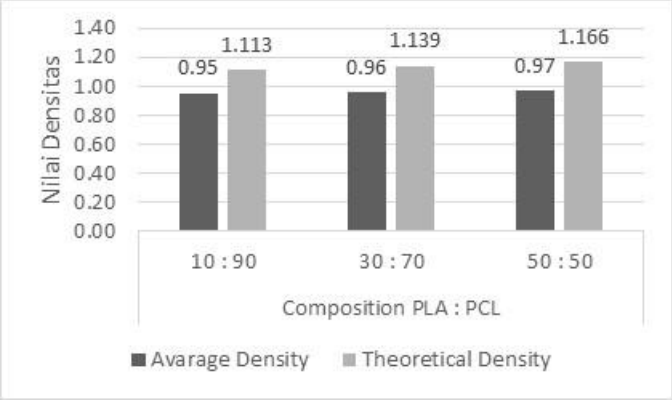

Figure 1: Comparison Chart of Density Measurement with Actual Density

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Degradation testing serves to observe the degradation rate of the PLA-PCL specimens and to understand the influence of PCL-PLA composition on their degradation rate. In this study, the immersion method was employed, conducted routinely every week for up to four immersion cycles, with immersion periods in the first, second, third, and fourth weeks, respectively. This immersion process was monitored weekly. The data from the degradation testing results using the immersion method can be seen in Table 3.

Table 2: Changes in mass of PLA-PCL specimens

Ratio PLA : PCL	Mass loss (g) in week:			
	1	2	3	4
10 : 90	0,1217	0,1259	0,1298	0,1238
	0,2033	0,2316	0,2363	0,2167
	0,2088	0,2268	0,2292	0,2178
30 : 70	0,1187	0,1255	0,1402	0,1208
	0,1618	0,1682	0,1758	0,1643
	0,1348	0,1437	0,1558	0,1378
50 : 50	0,0807	0,0896	0,0919	0,0877
	0,0685	0,0756	0,0777	0,0717
	0,0550	0,1626	0,1677	0,0782

Figure 2 show the percentage of the mass loss every week of observation. In the first week, the percentage of weight loss is greater than in the following weeks. This indicates that at the beginning of immersion, the specimen surface dissolves more quickly, while in the subsequent weeks, the dissolution decreases. The decrease in weight loss is caused by the formation of a protective layer that hinders the dissolution rate.

Figure 3 shows the degradation rate of the specimens in every week of observation. It can be seen that the degradation rate at the beginning of immersion is very fast and begins to decrease dramatically in the following weeks. PLA and PCL have different dissolving speeds. When immersed in the solution, PLA will dissolve faster. The dissolving process occurs on the surface of the specimen. PLA on the surface starts to dissolve first. The dissolving process of the specimen starts to decrease when the surface of the specimen starts to contain a lot of PCL. This causes the degradation rate to drop. The decreased degradation rate is also due to the fact that on the surface, the degradation products are still attached. The partial covering of the surface by the degradation products also contributes to the decrease in the degradation rate. Figure 4 shows a schematic of the degradation mechanism in the PLA-PCL composite.

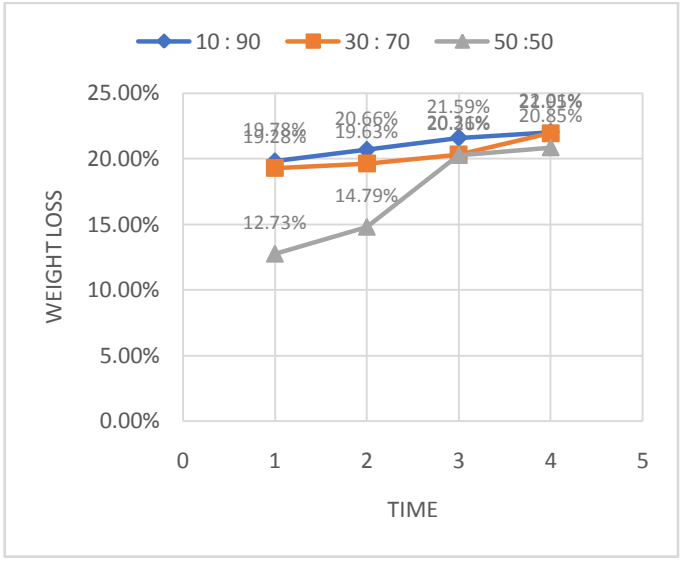

Figure 2: Weight Loss

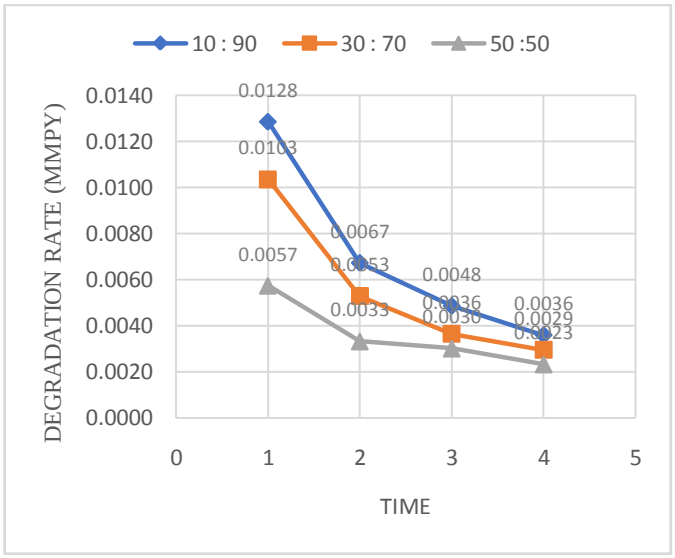

Figure 3: Degradation Rate

For the composition variations, the one with the highest degradation rate is the 10 PLA:90 PCL composition, with a degradation rate of 0.0128 mmpy in the first week, 0.0067 mmpy in the second week, 0.0048 mmpy in the third week, and 0.0036 mmpy in the fourth week. This is followed by the 30 PLA:70 PCL composition, which has a degradation rate of 0.0103 mmpy in the first week, 0.0053 mmpy in the second week, 0.0036 mmpy in the third week, and 0.0029 mmpy in the fourth week. Lastly, the composition with 50 PLA:50 PCL has the lowest degradation rate, with values of 0.0057 mmpy in the first week, 0.0033 mmpy in the second week, 0.0030 mmpy in the third week, and 0.023 mmpy in the fourth week.

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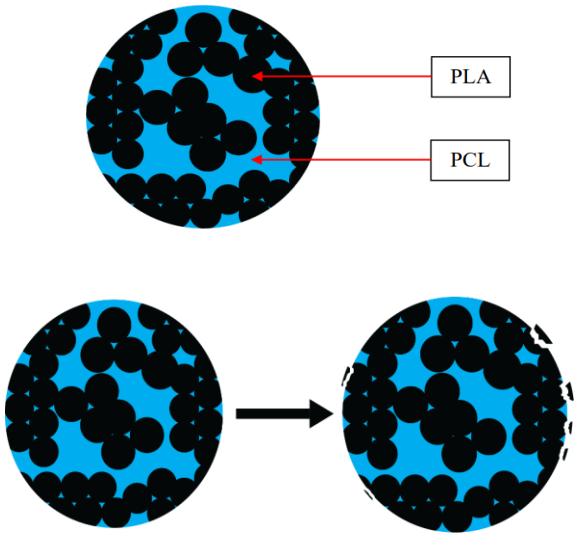

Figure 4: PLA and PCL Degradation Scheme

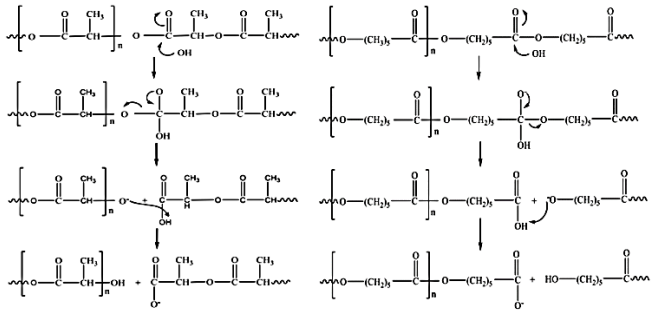

Figure 5: PLA and PCL Hydrolysis Reaction Mechanism

It can be observed from Figure 5 that hydrolysis reactions can also affect the degradation process. There is water present in the infusion solution used, leading to hydrolysis reactions on the specimens. When the PLA-PCL polymer blend, especially PLA (Polylactic Acid), is exposed to water or water-based solutions like infusion solutions, they tend to undergo hydrolysis. The possible mechanism of ester bond hydrolysis reactions that may occur during the degradation process of PLA-PCL.

IV. CONCLUSION

The highest density value was found in the composition variation of 50 PLA:50 PCL, with a density of 0.97 g/cm3. However, in the theoretical density testing, the highest density value was obtained in the composition variation of 50 PLA:50 PCL, with a density of 1.66 g/cm3. The theoretical density measurements also revealed that as the amount of PLA in the mixture increased, the density value also increased.

In the degradation testing using the immersion method every week, the results showed that with increasing immersion time, the percentage of weight loss also increased when calculated using weight loss method. The composition variations showed an increase in weight loss percentage as well. The composition variation of 10 PLA:90 PCL had the highest weight loss percentage, with a value of 22.01% in the fourth week of immersion. In the degradation rate calculations, it was found that with longer immersion time, the degradation rate decreased or even tended to stabilize. The composition variation of 10 PLA:90 PCL had the highest degradation rate value, with a value of 0.0128 mmpy in the first week of immersion.

REFERENCES

[1] Permatasari, C., Yuhertiana, I., & Kirana, N. W. I. (2022). Persepsi Konsumen dalam Melakukan Pembayaran BPJS Kesehatan dengan Menggunakan E-Wallet di Masa Pandemi Covid-19. *Journal of Management and Bussines (JOMB)*, *4*(2), 1029–1037. https://doi.org/10.31539/jomb.v4i2.4502.

[2] Sheikh, Z., Najeeb, S., Khurshid, Z., Verma, V., Rashid, H., & Glogauer, M. (2015). Biodegradable Materials for Bone Repair and Tissue Engineering Applications. *Materials*, *8*(9), 5744–5794. https://doi.org/10.3390/ma8095273

[3] Fadlilah, F. R., & Shovitri, M. (2014). Potensi Isolat Bakteri Bacillus dalam Mendegradasi Plastik dengan Metode Kolom Winogradsky. *Jurnal Teknik Pomits*, *3*(2), 40–43.

[4] Serfandi, D. N., Setyarini, P. H., Purnami, P., & Sulistyono, S. (2022). Karakterisasi Sudut Kontak dan Biodegradasi Pada Komposit Polylactide Acid (PLA) – Polycaprolactone (PCL) Untuk Aplikasi Biomaterial. *Prosiding Sains Nasional Dan Teknologi*, *12*(1), 124–130. https://doi.org/10.36499/psnst.v12i1.7243

[5] Saputra, R. A., Savetlana, S., & Sukmana, I. (2023). Komposit PLA (Poly Lactic Acid) untuk Aplikasi Biomaterial Scaffold Mampu Terdegradasi. *Jurnal Energi Dan Manufaktur*, *15*(1), 7–18. https://doi.org/10.24843/jem.2022.v15.i01.p02

[6] Warastuti, Y., & Suryani, N. (2013). Karakteristik Degradasi dari Biomaterial Poli-(kaprolakton-kitosan-hidroksiapatit) Iradiasi Dalam Larutan Simulated Body Fluid. *Jurnal Ilmiah Aplikasi Isotop Dan Radiasi*, *9*(1), 11–22.

[7] Assalam, M. R., Ismail, R., Bayuseno, A. P., & Fitriyana, D. F. (2023). Pengaruh Suhu Pembuatan Filamen Biokomposit (PCL/PLA/HA) Menggunakan Mesin Screw Extruder. *Jurnal Teknik Mesin S-1*, *11*(3), 11–16.

[8] Hijriawati, M., & Febriana, E. (2016). Review: Edible Film Antimikroba. *Jurnal Farmaka*, *14*(1), 8–16.

[9] Fell, E. V., & Lukianova, N. A. (2015). British Universities: International Students' Alleged Lack of

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Critical Thinking. *Procedia - Social and Behavioral Sciences*, *215*(June), 2–8. https://doi.org/10.1016/j.sbspro.2015.11.565

[10] Mohazzab, P. (2017). Archimedes' Principle Revisited. *Journal of Applied Mathematics and Physics*, *5*(4), 836–843. https://doi.org/https://doi.org/10.4236/jamp.2017.54073

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