

The Effect of Castor Oil Composition as a Plastisizer on Bioplastic with Spider Silk Reinforcement

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Abstract

The increasing amount of plastic waste demands the development of sustainable and environmentally friendly alternative materials. This study aims to develop bioplastics reinforced with spider silk as a natural reinforcement and to analyze the effect of varying concentrations of castor oil as a bio-based plasticizer on the characteristics of the resulting bioplastics. The novelty of this study lies in the combination of using spider silk as a reinforcement and castor oil as a natural plasticizer to produce a more sustainable material. Bioplastics were made with varying concentrations of castor oil of 0%, 10%, and 20%, then tested for their mechanical properties and biodegradability. The results showed that increasing the concentration of castor oil was directly proportional to the increase in biodegradability and elongation at break of the bioplastics. However, this increase in concentration actually decreased the tensile strength of the material. Furthermore, observations showed that spider silk fibers did not decompose completely and tended to form granular aggregates within the bioplastic matrix, resulting in uneven distribution of the reinforcement. Overall, the resulting bioplastics did not meet national quality standards (SNI), particularly in terms of tensile strength. Nevertheless, this research makes a significant contribution to the development of bio-based materials and aligns with Sustainable Development Goal (SDG) 12 on responsible consumption and production. Further research is recommended to optimize the material composition to achieve a more homogeneous reinforcement distribution and improved mechanical properties.

Keywords: bioplastic; castor oil; plasticizer; spider silk; biodegradability

Abstrak

Meningkatnya jumlah limbah plastik menuntut pengembangan material alternatif yang berkelanjutan dan ramah lingkungan. Penelitian ini bertujuan untuk mengembangkan bioplastik yang diperkuat dengan sutra laba-laba sebagai bahan penguat alami serta menganalisis pengaruh variasi konsentrasi minyak jarak (castor oil) sebagai plasticizer berbasis hayati terhadap karakteristik bioplastik yang dihasilkan. Kebaruan penelitian ini terletak pada kombinasi penggunaan sutra laba-laba sebagai penguat dan minyak jarak sebagai plasticizer alami untuk menghasilkan material yang lebih berkelanjutan. Bioplastik dibuat dengan variasi konsentrasi minyak jarak sebesar 0%, 10%, dan 20%, kemudian diuji sifat mekanis dan kemampuan biodegradasinya. Hasil penelitian menunjukkan bahwa peningkatan konsentrasi minyak jarak berbanding lurus dengan peningkatan kemampuan biodegradasi dan elongasi saat putus (regangan pada patahan) bioplastik. Namun, peningkatan konsentrasi tersebut justru menurunkan

kekuatan tarik material. Selain itu, pengamatan menunjukkan bahwa serat sutra laba-laba tidak terurai sepenuhnya dan cenderung membentuk agregat granular di dalam matriks bioplastik, sehingga menyebabkan distribusi bahan penguat menjadi tidak merata. Secara keseluruhan, bioplastik yang dihasilkan belum memenuhi standar mutu nasional (SNI), khususnya dalam hal kekuatan tarik. Meskipun demikian, penelitian ini memberikan kontribusi signifikan terhadap pengembangan material berbasis hayati dan sejalan dengan Tujuan Pembangunan Berkelanjutan (SDG) nomor 12 mengenai konsumsi dan produksi yang bertanggung jawab. Penelitian lebih lanjut disarankan untuk mengoptimalkan komposisi material guna mencapai distribusi penguat yang lebih homogen serta meningkatkan sifat mekanisnya.

Kata Kunci: bioplastik; minyak jarak; pemlastis; sutra laba-laba; biodegradabilitas

Introduction

Biodegradable plastics offer an environmentally preferable alternative to petroleum-based plastics because they can be degraded by microorganisms under suitable conditions, whereas petroleum-based plastics such as PET exhibit minimal degradation and persist longer in the environment (Nachod et al., 2021). A key additive in their production is the plasticizer, which enhances flexibility and workability (Halim et al., 2023; Tan et al., 2022). Plasticizers may be derived from glycerol, animal oils, or plant-based oils. Previous studies have explored various plant oils as plasticizers, such as soy protein isolate containing glycerol and glycols (Aguilar, 2020), seed oil byproducts like sunflower and cottonseed oil (Yadav & Ali, 2022), and even waste cooking oil in carrageenan or corn starch bioplastics (Mangaka et al., 2024).

Despite these advances, research on castor oil as a plasticizer remains limited, even though its physical and chemical properties are similar to soybean oil. Castor oil contains around 46% unsaturated fatty acids, dominated by oleic acid (Putri, 2023), making it a promising candidate for bioplastic formulation. Unsaturated fatty acids are widely used in bioplastic formulations because they contain carbon-carbon (C=C) double bonds that serve as reactive sites for various chemical modifications, such as epoxidation, polymerization, and cross-linking. These reactions enable the formation of a more stable polymer network, thereby improving the mechanical and functional properties of bioplastics (Miao et al., 2014; Perez-Puyana et al., 2024).

In addition to plasticizers, reinforcement materials are essential for improving bioplastic strength. Natural fibers such as cottonseed (Yue et al., 2020), water hyacinth (Pratama et al., 2020), chitosan from shrimp shells (Jahiding & Khaeri, 2022), seaweed (Adam et al., 2022), and rice husk silica (Srivastava et al., 2023) have been tested, but their performance remains suboptimal.

An untapped yet highly promising reinforcement material is spider silk, one of the strongest natural fibers known (Mi et al., 2023). Produced by the major ampullate and flagelliform glands (Römer & Scheibel, 2008), spider silk is composed primarily of the amino acids glycine and alanine (Fatimatuzzahro, 2021). Alanine-rich regions form highly ordered β -sheet nanocrystals that provide exceptional tensile strength and stiffness, while glycine-rich regions form amorphous domains that impart flexibility and extensibility. The hierarchical arrangement of these crystalline and amorphous structures enables spider silk to simultaneously exhibit high strength, elasticity, and toughness (Scheibel, 2004; Keten & Buehler, 2010), making it an excellent reinforcement material for biodegradable bioplastics.

This study investigates the effect of varying castor oil concentrations as a plasticizer in spider silk-reinforced bioplastics. The novelty of this research lies in (1) the

use of spider silk as a natural reinforcing material, (2) the incorporation of castor oil as a bio-based plasticizer, and (3) determining the optimal castor oil composition. This research contributes to the development of sustainable biodegradable materials with improved properties by utilizing renewable resources as both plasticizer and reinforcement. As the accumulation of non-biodegradable plastic waste continues to threaten terrestrial and aquatic ecosystems, the development of environmentally friendly materials is becoming increasingly important. Therefore, this research aligns with Sustainable Development Goal (SDG) 12: Responsible Consumption and Production by promoting renewable materials, encouraging more sustainable resource utilization, and supporting efforts to reduce plastic waste through sustainable material innovation.

Materials and methods

This research was conducted over a period of three months at two locations: the Research Laboratory of Universitas Pendidikan Indonesia and the iLab BRIN Laboratory in Cibinong. These sites were selected due to their complete research facilities and the availability of instruments necessary for bioplastic fabrication and testing, particularly for tensile strength and biodegradability assessments.

The instruments used in this study included an analytical balance, beakers, measuring cylinders, drop pipettes, stirring rods, a hot plate, magnetic stirrer, bioplastic acrylic molds, and a thermometer. The materials used consisted of castor oil (as a plasticizer), glycerol, spider silk extract, liquid nitrogen, acetic acid, cornstarch, and distilled water. These materials were selected based on their specific roles in bioplastic formation and degradation potential.

The research variables were adapted and modified from previous studies. The independent variable in this study was the concentration of castor oil used as a plasticizer, with variations of 0%, 10%, and 20%. The controlled variables were the composition of glycerol, cornstarch, and acetic acid, maintained in a fixed ratio of 1:1:1, as told by Anggraeni et al. (2021). The dependent variables were the biodegradability and tensile strength of the resulting bioplastics. These variables were selected to assess the impact of castor oil concentration on the mechanical and environmental properties of the bioplastics.

The research procedure began with the extraction of spider silk. The silk was first rinsed with sterile distilled water and then air-dried at room temperature (25°C). Once dried, it was ground using a mortar with the addition of liquid nitrogen until it formed a fine powder. This process aimed to obtain protein molecules from the spider silk without degradation, allowing for homogeneous blending with the other bioplastic components. This technique followed the liquid nitrogen-based freeze-drying approach by Bhatta *et al.* (2020), which has been shown to preserve bioactive compounds and structural integrity.

The next step was the formulation of the bioplastics. The spider silk extract was dissolved in 50 mL of distilled water, followed by the addition of glycerol, cornstarch, acetic acid, and castor oil based on the specified variations. The mixture was then heated to 60°C to induce gelatinization. After reaching a uniform consistency, the bioplastic solution was poured into acrylic molds designed according to ASTM D638 ISO 527-1 and ISO 527-2 tensile test standards, as well as into petri dishes. The samples were then left to dry at room temperature. This procedure was adapted from Dewi et al. (2021) and Pella et al. (2020) with several modifications.

After the formulation process, the mechanical properties of the bioplastics were tested through a tensile strength test using a Universal Testing Machine (UTM) 1000 KN

at iLab BRIN. This test evaluated the material's resistance to tensile force and its ability to withstand stress before breaking, in line with the methodology described by Salindeho (2013). The results served as a reference for assessing the material's mechanical performance as a sustainable plastic alternative.

The final stage was the biodegradability test. This test involved immersing bioplastic samples in 10 mL of water for seven days, following the method by Nandiyanto et al. (2020). After immersion, the samples were dried and weighed again to measure mass loss. Biodegradation was evaluated by calculating the percentage of mass loss using the formula:

$$\text{Mass loss (\%)} = \frac{W_0 - W_1}{W_0} \times 100\%$$

where W_0 is the initial weight and W_1 is the final weight after immersion. This test was conducted to determine the environmental degradability of the bioplastics under aqueous conditions.

Results and discussion

Morphological Analysis

Figure 1a shows the morphology of the bioplastic that has been made. The bioplastic was molded according to tensile test mold standards (ASTM D638 ISO 527-1 & ISO 527-2). The dimensions of the bioplastic are 165 mm long, 19 mm wide, and 3.2 mm thick. The resulting bioplastic has a transparent color. The spider web fibers in the bioplastic do not achieve perfect homogeneity, resulting in coarse granular aggregates that fill the bioplastic polymer. Figure 1b shows the surface of the bioplastic observed under a microscope at 40X magnification. The spider web fibers used in this study do not break down completely. The fine fibers from the spider web that do not break down are observed. The spider silk fibers used in this study did not break down completely. The fine fibers from the spider silk that did not break down were observed to fill the bioplastic polymer. Most of the spider silk fibers formed brown aggregates and were scattered throughout the bioplastic polymer. Some air bubbles were observed trapped within the polymer. This could occur during bioplastic production, where incomplete mixing causes air to become trapped within the bioplastic mixture. Additionally, the process of pouring the mixture into the mold could also cause air bubbles to become trapped within the polymer. Figure 1c shows the bioplastic after drying following the 7-day biodegradation test. The brown aggregates, which are spider web aggregates, appear unchanged. Soaking in water does not degrade the spider web. However, water enters the cracks of the bioplastic, creating large pores on the surface and inside the bioplastic. Microorganisms in the water, such as fungi and bacteria, play a role in breaking the polymer bonds in the bioplastic, thereby supporting the biodegradability of the bioplastic (Nanda et al., 2022; Paloyan et al., 2025).

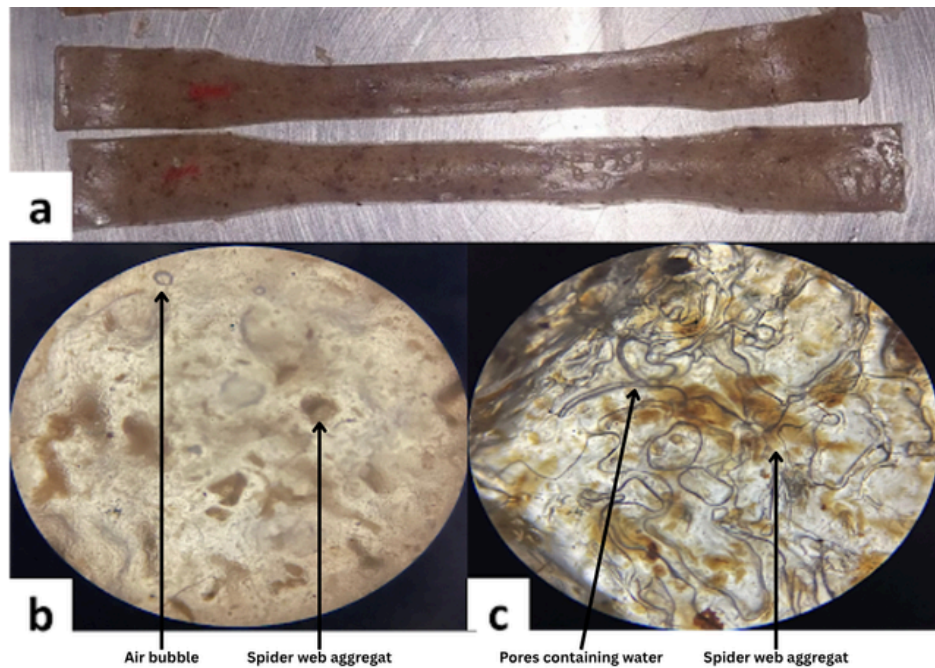


Figure 1. (a) Morphology of bioplastics, (b) Surface of bioplastics before biodegradation testing, (c) Surface of bioplastics after biodegradation testing

Biodegradation Test

The biodegradability of bioplastic samples with different castor oil contents was tested using the immersion method (Nandiyanto et al., 2020). Table 1 shows the weight loss of bioplastics during the biodegradability test. The biodegradability test was conducted over 7 days. All samples became lighter, the color of the bioplastics turned white, the morphology of the bioplastics once immersed became porous, and they were easily crushed. Figure 1c shows the pores formed post-immersion. Analysis indicated that the sample with 2 mL of castor oil had the best biodegradability, as evidenced by a weight loss of 86% to 96% during one week. The weight loss during one week of immersion was likely due to the outer surface of the bioplastic dissolving into the water.

Table 1. Weight Loss of Bioplastic during Immersion in Biodegradation Test

Variation	Day	Initial dimension (cm ²)	Initial mass (g)	Mass after soaking (g)	Mass loss (%)	Dimension of decay (g/cm ²)
0 mL	1	2.68	0.210	0.130	38	0.030
	2	2.68	0.190	0.100	47	0.034
	3	2.68	0.230	0.095	59	0.050
	4	3.20	0.290	0.090	69	0.062
	5	3.20	0.290	0.060	79	0.072
	6	3.20	0.300	0.055	82	0.077
	7	3.20	0.300	0.045	85	0.080
1 mL	1	2.68	0.210	0.075	64	0.050
	2	2.68	0.200	0.060	70	0.052
	3	2.29	0.180	0.050	72	0.057

Variation	Day	Initial dimension (cm ²)	Initial mass (g)	Mass after soaking (g)	Mass loss (%)	Dimension of decay (g/cm ²)
	4	2.42	0.190	0.040	79	0.062
	5	2.62	0.230	0.035	85	0.074
	6	2.52	0.180	0.025	86	0.062
	7	2.62	0.230	0.020	91	0.080
2 mL	1	2.56	0.220	0.060	73	0.062
	2	2.78	0.220	0.050	77	0.061
	3	2.56	0.230	0.040	83	0.074
	4	2.32	0.230	0.030	87	0.086
	5	2.56	0.210	0.010	95	0.078
	6	2.78	0.240	0.010	96	0.083
	7	2.78	0.270	0.010	96	0.094

Table 2. Average Percentage of Mass Loss in Biodegradation Test Based on the LSD Test

Comparison of Variations	Mean Difference (%)	Sig. (p-value)	Interpretation
0 mL - 1 mL	12.571	0.090	Not significantly different ($p > 0.05$)
1 mL - 2 mL	8.571	0.238	Not significantly different ($p > 0.05$)
2 mL - 0 mL	21.143*	0.007	Significantly different ($p < 0.05$)

Based on the results of the Least Significant Difference (LSD) in Table 2, variation in castor oil concentration between 2 mL and 0 mL showed a statistically significant difference compared to the variations 0 mL - 1 mL and 1 mL - 2 mL. This results indicates that increasing the concentration of castor oil as a plasticizer leads to an increase in the average mass loss of the bioplastic during the biodegradation test process. In addition, during the biodegradation test process, samples containing 2 mL of castor oil showed enhanced brittleness and softness in texture compared to other samples on the same immersion days. The castor oil content affects the ester bonds between starch in the bioplastic. Thus, the presence of castor oil in bioplastic facilitates microorganisms in breaking the carbohydrate ester bonds in bioplastic (El Feky et al., 2024). Additionally, the starch used in this study was cornstarch (maizena). Triawan et al. (2020) reported that microorganisms can easily break the polymer chains in cornstarch-based bioplastics. The

ester bonds between cornstarch and castor oil are relatively weak and easily broken (Triawan et al., 2020; El Feky et al., 2024). This means that the addition of castor oil can accelerate crack propagation in bioplastics. The presence of cracks in bioplastics allows for an increase in the contact area between water and the bioplastic. Contact between bioplastics and microorganisms in water can degrade the polymer bonds in the bioplastic. The breakdown of polymer bonds in bioplastics is often accompanied by the growth of fungi, which further accelerates the degradation of the bioplastic (Tiawan et al., 2020).

Tensile Test

Table 3. Average of Tensile Test

Code	Ingredients			Responses	
	Castor oil (mL)	Spider web (g)	Maximum time (s)	Tensile strength (MPa)	Elongation (%)
MJ0	0	0.4	12	0.053	3219
MJ1	1	0.4	35	0.102	8578
MJ2	2	0.4	40	0.044	10851

Overall, the bioplastic samples produced are still far from meeting bioplastic quality standards. According to SNI 7188.7:2016, the tensile strength of bioplastics is 24.7–302 MPa, while the elongation at break is 21–220% (Simarmata et al., 2023). Based on the average tensile test results presented in Table 3. The increase in castor oil concentration is directly proportional to elongation (%) and inversely proportional to tensile strength (Mpa). This can be seen from the elongation at break value, which increases as the amount of castor oil added increases. The addition of castor oil as a plasticizer can reduce the energy required for molecules to move, thereby lowering the stiffness of the material. This causes the tensile strength to decrease as the concentration of plasticizer increases (Putra et al., 2017). This is also revealed in the research of Putri et al. (2021), which states that the addition of castor oil plasticizer concentration increases the elongation at break value and decreases the tensile strength value. The use of plasticizers can reduce intermolecular forces, thereby increasing the elasticity of bioplastic materials. Therefore, as the concentration of castor oil increases, the percentage of elongation also increases (Budiman et al., 2018). The increase in elongation at break is caused by the excess molecular weight of the plasticizer, which causes the distance between starch chains to widen, thereby facilitating molecular movement (Dewi et al., 2023). Thus, in this study, the elongation at break value is inversely proportional to the tensile strength value.

But, conversely, the addition of castor oil lowers the value of tensile strength that can be seen in Table 3. The decrease in tensile strength value along with the increase in the amount of plasticizer is probably because the plasticizer can cause its molecules to be located between biopolymer bonds and interact to form hydrogen bonds in the bond chain between polymers so that the interaction between biopolymers decreases and the tensile strength value decreases (Purwanti, 2010; Abotbina et al., 2021). High concentrations of plasticizer have been reported to reduce tensile strength of

biopolymers by weakening the intermolecular bonds between cornstarch and the plasticizer through hydrogen bond formation (Sanyang et al., 2015). The effect is driven by the hydrophilic nature of the plasticizer which exhibits high water absorption and consequently enhances hydrogen bond interactions (Vieira et al., 2011).

Conclusion

Based on the results of the study, bioplastics with the addition of 2 mL of castor oil as a plasticizer exhibited the highest level of biodegradability compared to other volume variations. The incorporation of castor oil serves to increase the flexibility of bioplastics by reducing intermolecular interactions within the polymer structure, thereby making it more susceptible to microbial degradation in the environment. As the amount of castor oil increases, both the biodegradability and the elongation at break of the bioplastic also tend to increase. This indicates that castor oil effectively enhances the ductility and flexibility of the bioplastic material, promoting its potential as an environmentally friendly alternative.

However, the increased amount of castor oil does not universally enhance all mechanical properties of the bioplastic. A negative correlation was observed between the volume of castor oil added and the tensile strength of the bioplastic. As the concentration of castor oil increases, the tensile strength tends to decrease. This can be attributed to the disruption of the polymer's cohesive interactions, leading to a weaker structural integrity.

On the other hand, the elongation at break improves with higher castor oil content indicating that the material becomes more stretchable and resilient before breaking. However the addition of castor oil conversely reduces the tensile strength value, as shown in Table 3. The decrease in tensile strength with increasing plasticizer content is likely due to the plasticizer molecules positioning themselves between the biopolymer chains. These molecules interact by forming hydrogen bonds within the polymer network, thereby weakening the interactions between biopolymer chains and ultimately lowering the tensile strength.

Furthermore, variations in the amount of castor oil used significantly affect other physical and mechanical characteristics of the bioplastic, including thickness, surface texture, homogeneity, and degradation time under natural conditions. These findings highlight the importance of optimizing the plasticizer concentration in bioplastic formulations to achieve a balanced combination of mechanical strength, elasticity, and biodegradability. Thus, castor oil can be regarded as a promising natural plasticizer in the development of environmentally friendly bioplastics suitable for various applications as alternatives to conventional plastics.

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