Mekanika: Majalah Ilmiah Mekanika

Effect of Holding Time and Temperature of Hot Pressing on Tensile Strength of Biodegradable Plastic Made of Carrageenan

Fitrian Imaduddin[1], Palgunadi Sastra[2], Wijang Wisnu Raharjo[2*], Putri Wullandari[3], Ridwan Ridwan[4]

1 Department of Mechanical Engineering, Islamic University of Madinah, Madina, Saudi Arabia

2 Department of Mechanical Engineering, Universitas Sebelas Maret, Surakarta, Indonesia

3 Research Institute for Fisheries Postharvest Mechanization, Ministry of Marine Affairs and Fisheries, Bantul, Indonesia

4 Department of Mechanical Engineering, Universitas Merdeka Madiun, Madiun, Indonesia

*Corresponding Author's email address: m_asyain@staff.uns.ac.id

Keywords:
Biodegradable plastic, hot press, carrageenan

The most significant environmental problem was caused by plastic. One way to handle plastic waste is biodegradable plastic because it decomposes naturally faster. Biodegradable plastic can be made from carrageenan by the hot press method. This method can make biodegradable plastic with large dimensions. However, the plastic quality depended on the holding time, temperature, and pressure selected during the hotpressing process. Therefore, this research is conducted to determine the effect of holding time and temperature in the hot press process on the tensile strength of biodegradable plastic made from carrageenan. The composition of the biodegradable plastic material used was 35% carrageenan, 35% polyvinyl alcohol (PVA), and 30% glycerol. In the manufacture of composites, the holding time was varied: 10, 20, 30, and 40 minutes, as well as the process temperature, varied from 100, 110, 120, and 130 °C. The tensile strength of the composite was observed through tensile testing using a Universal Testing Machine. FTIR, XRD, and SEM tests were also conducted to sharpen the analysis. The addition of holding time led to an increase in the tensile strength of biodegradable plastics. The highest tensile strength was obtained at a holding time of 30 minutes with a value of 4.45 MPa. After 30 minutes, the tensile strength of the biodegradable composite decreased. Meanwhile, the addition of process temperature caused a decrease in the tensile strength of biodegradable plastics. The highest tensile strength was obtained at a process temperature of 100 °C with a value of 5.28 MPa.

1 Introduction

In recent decades, plastic consumption has increased. The recent COVID-19 pandemic in 2020 has increased the need for single-use plastic. More than 8,000,000 tons of pandemic-associated plastic waste have been generated globally, with more than 25,000 tons entering the global ocean [1]. Plastic is a

https://dx.doi.org/10.20961/mekanika.v21i2.64292

Revised 12 August 2022; received in revised version 22 August 2022; Accepted 25 August 2022
Available Online 30 September 2022

2579-3144

Group of synthetic or natural materials produced from a molecular chain consisting of carbon as the main element. There are about 98% of food products packaged using plastic. However, conventional plastics take 500 – 1,000 years to decompose naturally by microbes. Using biodegradable plastic as a packaging material is one way to overcome the difficulty of traditional plastics biodegrading in nature [2-3]. Analysis of volatile organic compounds produced during incineration of non-degradable and biodegradable plastics has been conducted in the study by Jang et al. [4]. In a recent study, Jin et al. [5] found that biodegradable plastics can be degraded by anaerobic digestion.

Furthermore, carrageenan-based bio nanocomposites for food packaging applications showed a crucial role in maintaining food quality and prolonging its shelf life [6]. Carrageenan is one of the known materials used for biodegradable plastic packaging. This material is extracted from seaweed. The existence of Indonesia as one of the largest seaweed-producing countries in the world makes carrageenan very potential to be developed as a biodegradable plastic material. In 2019, approximately 10,000,000 tonnes of global carrageenan seaweed were produced [7].

Forming thermoplastic composite can be done by hot press forming or injection molding. The hot press process is more advantageous than injection molding because the fiber undergoes one heating cycle, reducing fiber degradation. In the hot press method, process parameters significantly influence the character of the resulting thermoplastic composite. Therefore, the proper process parameters must be selected to produce an optimal composite product. Temperature, pressure, and holding time are essential parameters when making plastic composites using a hot press (Figure 1). The increase in temperature until the melting temperature causes the matrix to melt, allowing the matrix to wet the reinforcement and fill the voids in the composite. However, increasing the temperature above the melting temperature can cause fiber degradation [8]. The optimum temperature for manufacturing unidirectional kenaf/PLA composites is 160 ^{o}C. The addition of processing pressure increases the mechanical properties of green composites. The increased time and pressing temperature can lead to a better bond between the reinforcement and the matrix leading to higher stiffness [9]. Holding time allows the matrix to melt and fill the composite voids [10]. The tensile properties of the PLA/jute fiber composites increased when the heating time was reduced. Therefore, the impact of process parameters on the mechanical properties of plastic composites needs to be carried out.

The research aims to study the effect of process parameters on the tensile strength of biodegradable plastic made from carrageenan. The investigated process parameters include process temperature and heating time. Additional tests such as XRD, FTIR, and SEM were conducted to deepen the analysis.

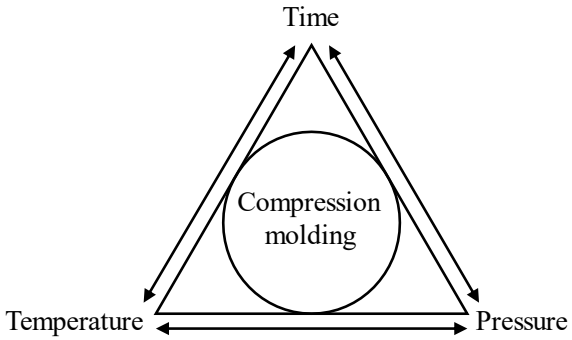

Figure 1. Parameters of the hot press method

2 Experimental Methods

2.1 Materials

The materials used are carrageenan, polyvinyl alcohol (PVA), and glycerol. The carrageenan was obtained from the Indonesian Carrageenan, Semarang. Carrageenan is a natural carbohydrate (polysaccharide) acquired from edible red seaweeds. Carrageenan is susceptible to depolymerization

through acid-catalyzed hydrolysis [11]. Carrageenan serves as a reinforcing material in the manufacture of biodegradable plastics. The specifications of carrageenan are having a grain size of 80 mesh, viscosity of 35 mPa·s, a melting point of 175 ºC, and a moisture content of 8%. The PVA was obtained from Bratachem, Yogyakarta. PVA functions as a matrix that aims as an adhesive so that the resulting composite can adhere well. The PVA used is in the form of a white powder. Glycerol was obtained from General Labora, Yogyakarta. Glycerol functions as a plasticizer to increase the matrix's flexibility and the elongation value of biodegradable plastics.

2.2 Biodegradable Composite Fabrication

Biodegradable plastic composites were made by mixing carrageenan, PVA, and glycerol using a food processor. The mass fraction of carrageenan: PVA glycerol used was 35%:35%:30%. The mixture of materials was converted into pellets using a single screw extruder with a process temperature of 130 °C and a rotational speed of 27 rpm [12]. Then, the extruded pellets were cut along 5 mm using a pelletizer and molded into biodegradable plastic using the hot press method, Figure 2. The holding time and process temperature vary in the hot press process. The process pressure of the hot press is 10 MPa [13] and the holding time changed by 10, 20, 30, and 40 minutes at a process temperature of 130 °C. The holding time that produces the highest tensile strength is selected as a fixed variable in the temperature variation of the hot press process. The process temperature varies from 100, 110, 120, and 130 °C.

2.3 Tensile Strength Test

Tensile testing of biodegradable plastic is carried out to know the tensile strength. Tensile tests were conducted using a Universal Testing Machine (JTM UTS 510). The biodegradable plastic was prepared in ASTM D882 for tensile testing at 50 mm/minute [14]. The tensile strength value is determined from the average tensile strength value of 5 specimens.

Figure 2. Hydraulic Hot Press Machine Fabrication

2.4 X-Ray Diffraction (XRD) test

The X-ray diffraction analysis of raw and treated powder CHF was carried out on Panalytical XRD (model PANalytical Xpert highscore plus) at a current of 30 mA and an accelerating voltage of 30 kV. The Segal method calculated the crystallinity index (CI) [2] by comparing crystalline intensity with the crystalline and amorphous intensity sum.

$$CI(\%) = \frac{I200 - Imin}{I200} x100 \quad (1)$$

2.5 Fourier Transform Infrared Spectroscopy (FTIR) test

JASCO FT/IR 4000 was used to obtain the structural analysis of the biodegradable plastic samples. The FTIR was used in the transmission mode, with a resolution of 4 cm^{-1} scans in the range of 4000–400 cm^{-1}. In addition, FTIR was used to detect molecular vibration to analyze the distribution of functional groups of the samples.

2.6 Scanning Electron Microscopy (SEM) test

SEM testing was carried out using the JEOL JSM-6510LA model tool (Japan), which aims to see the fracture surface of the specimen obtained from the tensile test. Each sample was mounted on the aluminum holder of the microscope using double-sided electrical conduction carbon adhesive tabs. The acceleration voltage used is 10 kV. The SEM analyses of the different specimens were observed at 100 and 1000 magnification.

3. Result and Discussion

3.1 Variation of pressing time

3.1.1 SEM analysis

SEM observations were carried out with 100x magnification in the cross-section position of the tensile test results of biodegradable plastic. SEM observations were performed with a magnification of 100x on the cross-section of the tensile test fracture of biodegradable plastic. The increase in pressing time allows carrageenan to have time to bond with one another. Meanwhile, less pressing time causes cavities to appear in biodegradable plastics. Carrageenan pores become a medium for spreading PVA and glycerol in biodegradable plastics. It results in a good bond between biodegradable plastic components [15].

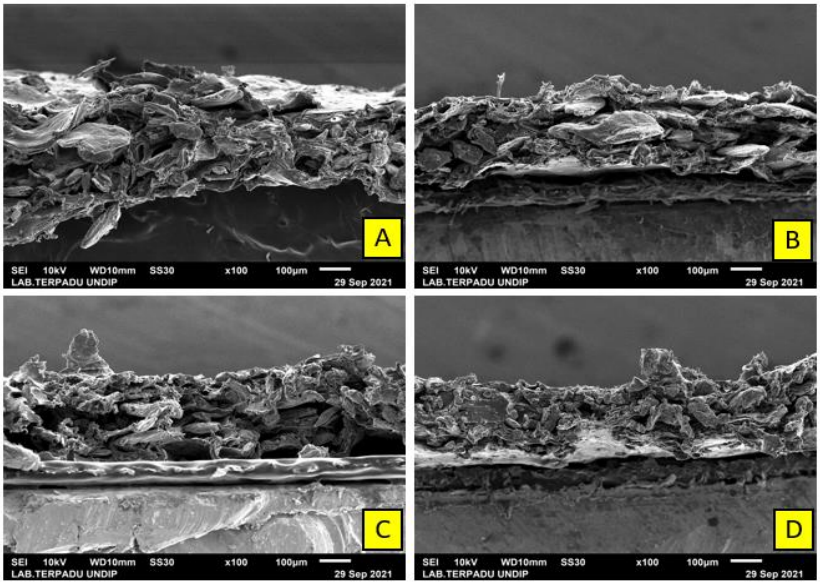

Figure 3. SEM observations of biodegradable plastic with variations in pressing time (a) 10 minutes; (b) 20 minutes; (c) 30 minutes; (d) 40 minutes

3.1.2 XRD analysis with variations in pressing time

The peak at position 20º has a lattice plane (1 0 3). The peak that is formed has a wide shape. It indicates that biodegradable plastic has an amorphous structure [16]. The increase in holding time from 10 minutes to 30 minutes caused a rise in the degree of crystallinity of biodegradable plastics from 23.13% to

25.62%. Conversely, the addition of holding time above 30 minutes decreased the degree of crystallinity to 25.62%.

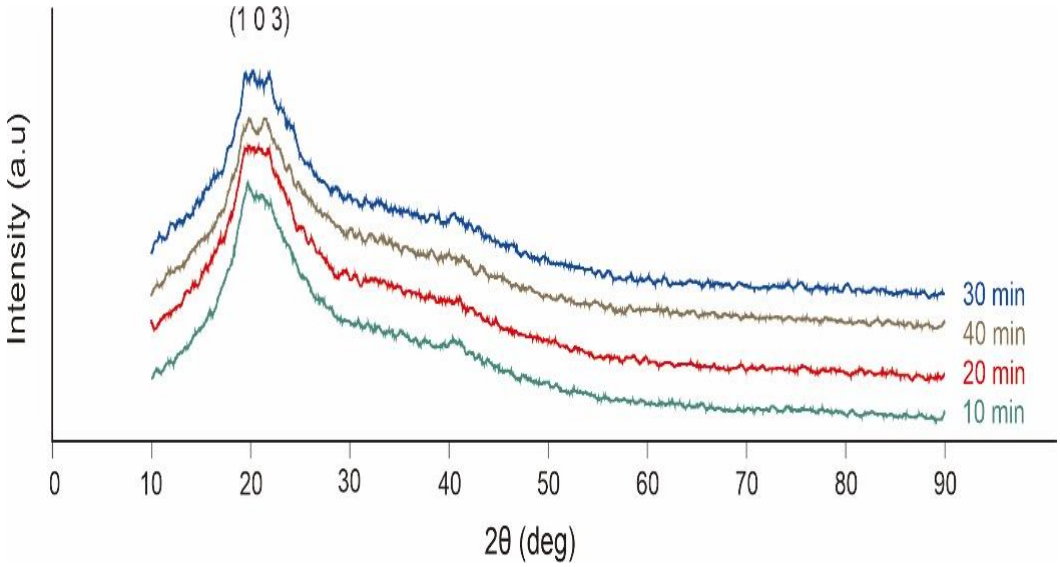

Figure 4. XRD observations of biodegradable plastics with variations in pressing time

The increase in crystallinity was caused by mixing PVA and glycerol with carrageenan, which made carrageenan more integrated with other carrageenans, as evidenced by SEM observations in Figure 3. In addition, the excellent bond between carrageenan with PVA and glycerol can increase hydrogen bonding so that there will be an increase in the degree of crystallinity.

3.1.2 Tensile test analysis with variations in pressing time

The highest tensile strength and elongation of biodegradable plastic occurred at a pressing time of 30 minutes, Table 1. Because PVA and glycerol can spread evenly and cover the cavities, a good bond is formed between carrageenan with PVA and glycerol. It is in line with the results of the XRD test, as shown in Figure 4. The pressing time of 30 minutes resulted in the highest degree of crystallinity due to the formation of good bonds between carrageenan, PVA, and glycerol. However, after 30 minutes, the tensile strength and elongation decreased. This decrease was due to the thermal degradation of carrageenan [17].

Table 1. The results of the tensile test of biodegradable plastic with variations in pressing time

Time (min)	Tensile Strength (MPa)	Elongation (%)	Modulus Young (MPa)
10	3.43	142.43	2.58
20	3.70	163.34	2.31
30	4.58	245.02	1.90
40	3.80	228.06	1.70

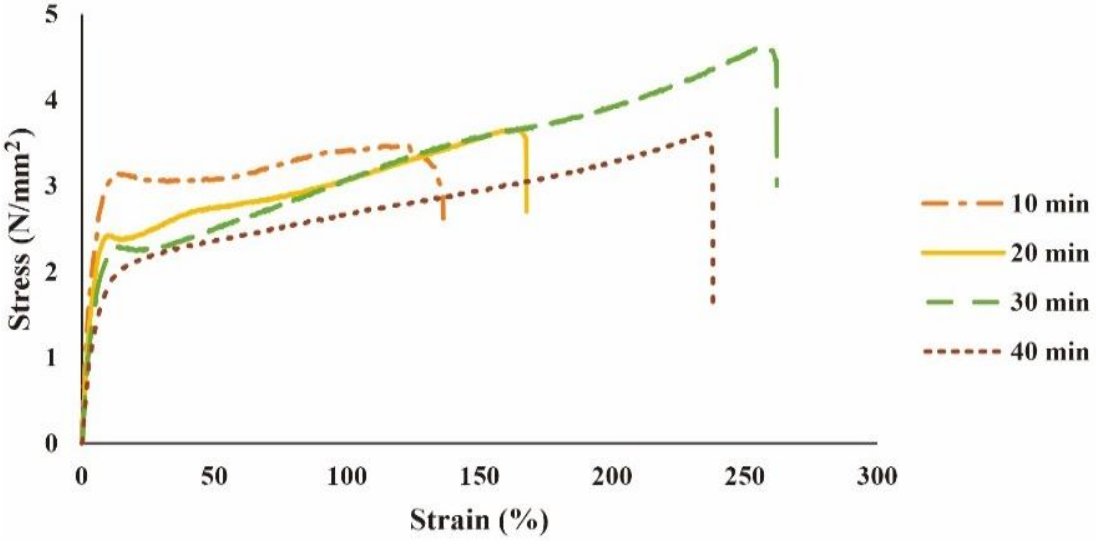

Figure 5. Stress-strain of biodegradable plastic with variations in pressing time

Increasing the pressing time can increase the ductility of biodegradable plastics. The increase in strain value evidences the increase in the ductility of biodegradable plastics, Figure 5. The increase in ductility in biodegradable plastics is directly proportional to the increase in the tensile strength value.

3.2 Variation of Pressing Temperature

3.2.1 SEM analysis with variations in pressing temperature

SEM observations were carried out with 1000x magnification in the cross-section position of the tensile test results of biodegradable plastic, Figure 6. Increasing the pressing temperature can cause the carrageenan to lose its shape due to the melting of the carrageenan due to the release of the carrageenan layer, which forms flakes on the surface of the carrageenan [18]. Increasing the pressing temperature can make the wrinkles on the carrageenan more visible because the carrageenan is getting damaged.

3.2.2 FTIR analysis with variations in pressing temperature

FTIR spectroscopy showed powerful light absorption at 1256 cm^{-1} for S=O bonds (ester sulfate) and 1041 cm^{-1} for C–O bonds (glycosidic), Figure 7. The peak at 924 cm^{-1} corresponds to the C=O=SO of galactose D-galactose4-sulphate, and 846 cm^{-1} corresponds to the C=O of 3,6 anhydrous-D-galactose. The peak at 3389 cm^{-1} is associated with the O- H stretching of PVA intermolecular and intramolecular hydrogen bonds. It corresponds to the stretching of polysaccharides and free hydroxyl groups. Furthermore, the reduction in the intensity of the O–H peak in glycerol mixed with PVA indicates the possibility of random chain rearrangement of PVA physical crosslinks with glycerol to destroy the crystals.

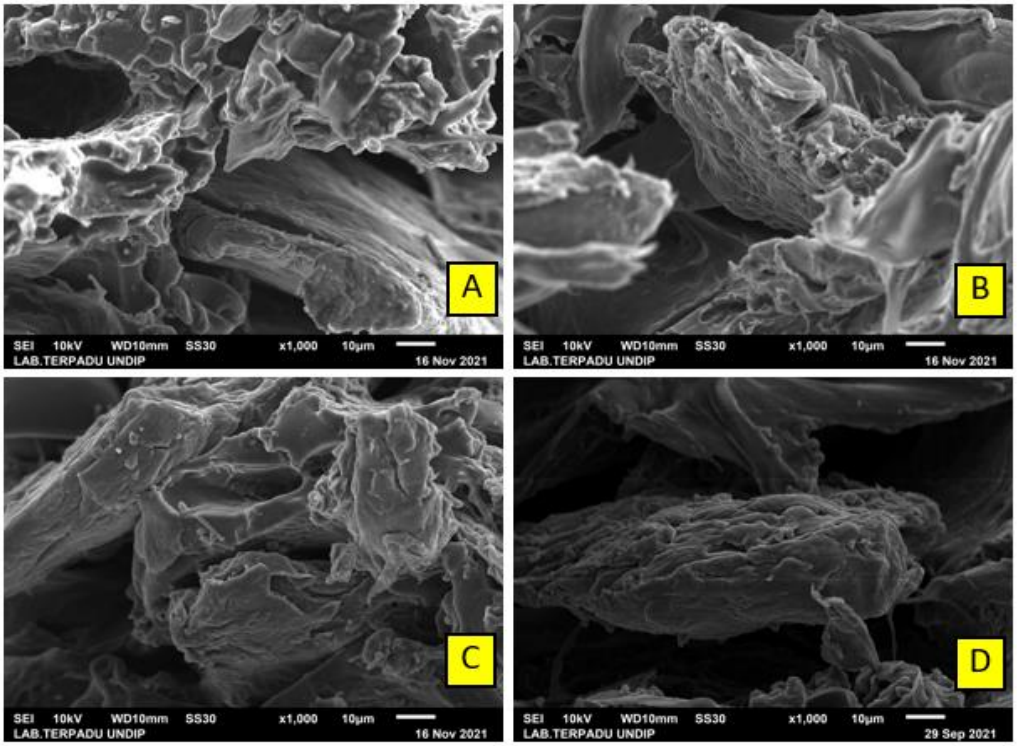

Figure 6. SEM observations of biodegradable plastic with variations in pressing temperature (a) 100 °C; (b) 110 °C; (c) 120 °C; and (d) 130 °C

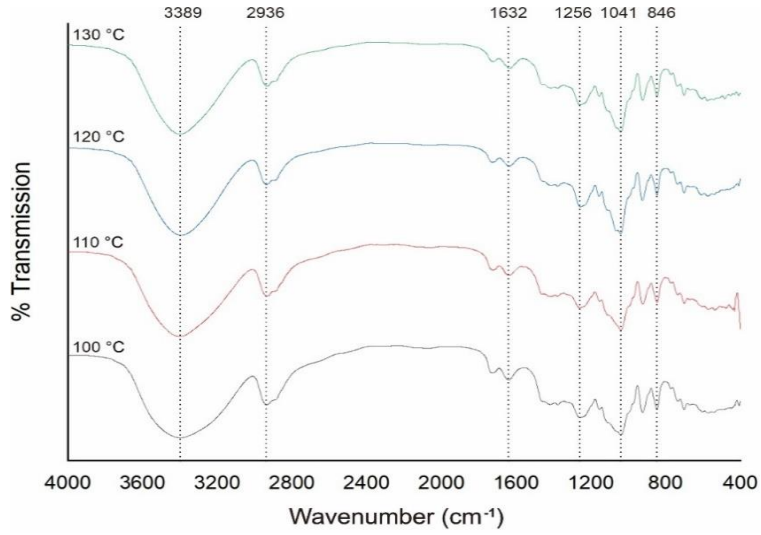

Figure 7. FTIR observations of biodegradable plastics with variations in pressing temperature

Furthermore, it can reduce the tensile strength due to an amorphous structure between glycerol and PVA where the polymer chains begin to open. The peak of 2936 cm^{-1} was associated with a moderate C–H functional group. In addition, the appearance of peaks at 1632 cm^{-1} and 1726 cm^{-1} indicates the C=O (ester) region.

3.2.3 XRD analysis with various pressing temperatures

The peak of 2 thetas is formed at 20°, so it has a lattice plane (1 0 3). The peak has a wide shape. The graph shows the degree of crystallinity of biodegradable plastics. The degree of crystallinity at a temperature of 100 °C was 28.62%, a temperature of 110 °C was 27.19%, a temperature of 120 °C was

26.90%, and a temperature of 130 °C was 25.62%. The increase in pressing temperature makes the degree of crystallinity decrease.

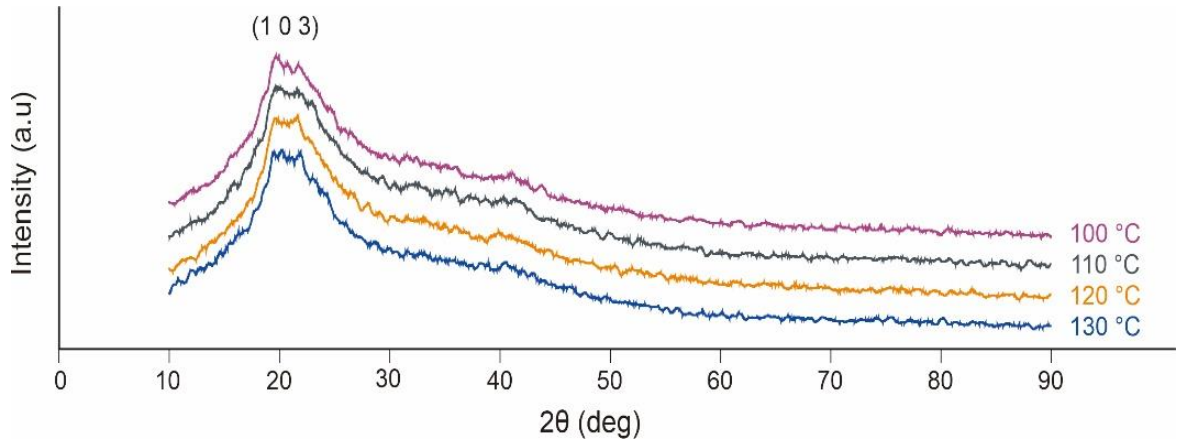

Figure 8. XRD observation of biodegradable plastic with variations in pressing temperature

The decrease in the degree of crystallinity is caused by the pressing temperature that exceeds the carrageenan's gelatinity temperature, as evidenced by the SEM observations in Figure 6. Therefore, the higher the press temperature when the liquid material is cooled, the material maintains a gel state, thereby reducing the crystallization process [19].

3.2.1 Analysis of tensile strength with variations in pressing temperature

The increase in pressing temperature causes a decrease in the tensile strength of biodegradable plastics, Table 2. The increase in press temperature that is too high can cause the molecular chains in carrageenan to break to reduce the tensile test results. The pressing temperature causes the breaking of the molecular chain to exceed the gelatinity temperature, causing decomposition [20]. Carrageenan undergoes decomposition, evidenced by carrageenan's many wrinkles as in the SEM observations in Figure 6. This decreases the degree of crystallinity in biodegradable plastics, as in XRD observations in Figure 8.

Table 2. The results of the tensile test of biodegradable plastic with variations in pressing temperature

Temperature (°C)	Tensile Strength (MPa)	Elongation (%)	Modulus Young (MPa)
100	5.28	113.97	4.85
110	4.91	161.98	3.38
120	4.73	179.53	2.64
130	4.58	245.02	1.89

Increasing the pressing temperature causes the biodegradable plastic to be more ductile, Figure 9. The increase in ductility is inversely proportional to the tensile strength of biodegradable plastic because the tensile strength value decreases. Strength assessment of a structure such as a ship structure and public transportation [21-26], tube under axial loading [27], and ballistic impact [28] can be seen in related literature. Further advanced development of sensors' roles in maritime-based industry and research can be seen in [29].

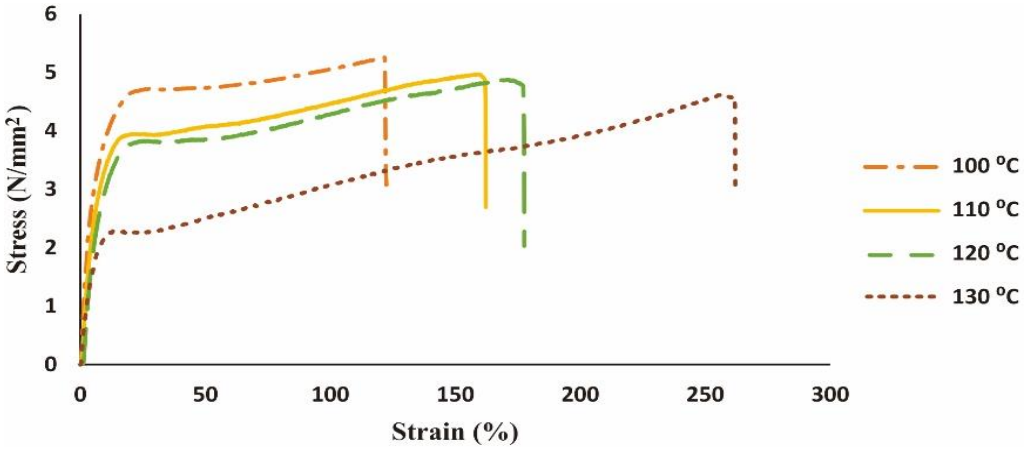

Figure 9. Stress-strain of biodegradable plastic with variations in pressing temperatures

4. Conclusion

Based on the results of tests that have been carried out on biodegradable plastics made from carrageenan, PVA, and glycerol, it can be concluded that:

a. The tensile strength of biodegradable plastic has increased up to 30 minutes, and the tensile strength of biodegradable plastic will decrease after 30 minutes.
b. The tensile strength of biodegradable plastic decreases due to the addition of temperature, i.e., 100 °C, 110 °C, 120 °C, and 130 °C in the hot press process.

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