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Research article

Use calcium silicate filler to improve the properties of sago starch based degradable plastic

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Abstract: The addition of fillers or additives to improve the mechanical properties of degradable plastics such as sago starch has gained the interest of researchers, scientists, and academicians. This research aims to investigate the addition of calcium silicate as an additive filler on the properties of a sago starch-based degradable plastic. The calcium silicate fillers used were 2, 4, 6, and 8% by weight starch, and the gelatinization process temperature used was 70, 80, and 90 °C. The properties of these plastics were analyzed in terms of their strength, chemical composition, thermal stability, water absorption, and degradation rate. The optimum mechanical characteristics included a tensile strength of 28.04 MPa, 32.55 MPa of elongation at the break, and 70.02% of Young's modulus obtained with the addition of 8% calcium silicate and a gelatinization temperature of 90 ºC. Fourier Transform Infrared (FTIR) showed that there were O-H, C-H, and C=O groups that existed at wave numbers of 3795.91 cm^{-1}, 2927.94 cm^{-1}, and 1433.11–1616.35 cm^{-1}; moreover, these groups are hydrophilic, which bind water, so they can be degraded by the microbial activity in the soil. Differential Scanning Calorimetry (DSC) showed that the degradable plastic had a thermogram peak at 271.38 °C;

additionally an endothermic peak occurred at 309.30 °C. The maximum swelling value was 64.05% at 2% calcium silicate and a gelatinization temperature of 70 ºC. The addition of calcium silicate made the plastic more water-resistant. The degradation rate of the degradable plastic ranged from 12–15 days and conformed to the American Standard Testing and Materials (ASTM) D-20.96 (maximum 180 days of decomposition for degradable plastic).

1. Introduction

The ever-increasing problem of plastic waste needs a solution. Synthetic plastics such as polyethylene terephthalate (PET) take between 23 to 48 years to break down in the environment [1]. Global plastic use was approximately 359 million tons in 2018, which increased from 245 million tons in 2008, and will be three times greater by 2050. Since the 1950s, a mass production of plastics has occurred. However, an effective strategy has yet to be implemented to address the problem of increasing pollution issues of plastic waste in the environment [2]. Several methods exist to manage all plastic pollution, including recycling, incineration, and burials. However, incinerating plastic waste can result in the emission of toxic chemicals that are hazardous to life and the earth's environment. Recycling is not practical because plastic is not biodegradable [3]. The types of commercial plastics used today generate a lot of waste and are harmful to health and nature because even if they are recycled, they cannot decompose for several thousand years. The issue of plastic recycling is still a concern due to its impact on safe and healthy living and the high cost of production.

An appropriate alternative is making a degradable plastic using a starch base material that is easily degraded by bacterial activity. Starch is used from various plastic bases to provide degradable plastics and, thus, alternatives to fossil-fuel plastic. Starches are used in food and other industries as thickeners, emulsifiers, binders, and as a bonding material in grocery and non-grocery industries. Since starches are bio-degradable, inexpensive, renewable, and plentiful, they are considered as a "green route" toward feedstock to produce porous materials such as aerogels, biofoams, and bio-plastics [4]. Sago starch can serve as a base material for the production of degradable plastic. Sago is widely available in Southeast Asia and has a low price compared to similar starchs [5]. Sago has the potential to contribute to food security as a supplementary source of staple food, with a minimal competition for arable land with other food crops. Additionally, it can be utilized for other industrial purposes, including the production of bioplastics and bioethanol. Sago is cultivated in numerous countries, including Indonesia, Papua New Guinea, Malaysia, Thailand, the Philippines, Timor-Leste, and various Pacific Island nations. Despite its role as a traditional food source and a generator of off-farm and non-farm income for impoverished rural communities, the population of sago trees has significantly declined in recent decades due to the conversion of wetlands and swamps planted with sago for other purposes. This includes the expansion of industrial crops such as oil palm and rubber. To restore the population of sago trees, degradable plastics are manufactured using sago starch. This restores the added value of sago trees and reuses the trees that are left unused [6].

One of the weaknesses of degradable plastics from starch is the associated mechanical characteristics, which still need to be compatible with other plastics; therefore, technology is required to increase its behavior. As reported in a previous study, adding a 2.0 wt% silica compound and glycerol to a sago starch/polyvinyl alcohol (PVA) film showed an improved tensile strength and a decreased

elongation at break when compared to a film without added silica [7]. The effect of calcium carbonate additives on the mechanical characteristics of bioplastics made from cassava starch using glycerol as a plasticizer has been studied in previous research. The best tensile strength was found using a 0.4% calcium carbonate additive (22.88 ± 1.46 MPa). The tensile strength increased with additives between 0.5 and 1.0%. Elongation was measured at 0.8% calcium carbonate (27.57 ± 0.14%) [8]. The effect of sodium silicate derived from bagasse ash was studied utilizing biobased films from corn and potato starch. Sodium silicate stopped mold growth on corn and potato starch bioplastics. Sodium silicate from renewable sources can produce biodegradable packaging that stops mold growth [9]. Studies on the effect of mineral particle fillers such as talc, calcium carbonate, kaolin, and micas on the mechanical properties of nature-based plastics produced plastic materials with strong and rigid mechanics and elasticities. Still, adding mineral fillers to nature-based plastic polymers did not lead to bacterial degradation, although the fillers' nature affected the polymer biodegradation rate [10].

Sago has starch, fiber, protein, lipid, and ash ingredients. Sago starch consists of micronutrients such as cellulose [11]. Sago is a common ingredient in soups, cakes, and puddings in the southwestern Pacific region, where it is used as a thickener for puddings and sauces. In industry, it is used as a textile stiffener. A sago starch-based degradable plastic is an ecologically beneficial material. Sago starch is derived from natural resources, and its production can be considered safe. One of the components of sago starch is amylose, which can quickly bind with other substances. Sago starch becomes gelatinized when heated to 70–120 ℃ [12]. It can be used as a bioplastic material. Starch-based plastics lack good mechanical properties, making them unsuitable for some applications, including food packaging. Modifications are being explored to improve the properties of starch-based plastics. This is mainly done by using reinforcing agents, changing the chemical properties, and blending them with other polymers.

Kamali and Ghahremaninezhad [13] conducted a nanocomposite polymer study using calcium-silicate-hydrate to understand the morphology of the nanocomposite/polymer sheets obtained, which resulted in a particle density and low roughness of the nanosheets produced. Calcium silicate is one of the biggest promises of bio-ceramics because of its outstanding properties, such as bioactivities, bio-compatibilities, and osteoinductivities [14]. The novelty in this research of degradable plastics from sago starch uses calcium silicate as a filler. Previously, some researchers have developed bioplastics using mineral fillers such as mica, talc, kaolin, wollastonite, calcium carbonate, silica, and montmorillonite [15,16]. One way to improve the mechanical properties of degradable plastics is to use calcium silicate fillers. Calcium silicate makes degradable plastics more degradable. Fillers are needed to make bioplastics stronger and more flexible. Calcium silicate is a relatively inexpensive material with a wide range of sizes, from micro to nano, and has a predictable impact on the final product's properties. The primary advantages of its utilization are an enhanced stiffness and a diminished process shrinkage, in addition to its flammability—properties that serve as significant constraints on the deployment of unmodified thermoplastic polymers [17]. There are two main ways to make the film: wet and dry. The first method is called solvent casting, which involves pouring a polymer solution into a mold and then drying the solvent. The second method is based on the thermoplastic properties of some biopolymers, such as the extrusion process. Related processes include compression molding, injection molding, and blow molding. However, thermoplastic starch (TPS) is not yet common worldwide, and research is ongoing to improve both the properties and the large-scale manufacturing [18]. A previous study highlighed the production of degradable plastics with calcium silicate reinforcement using the casting method. The calcium silicate concentrations used were 2%, 4%, 6%, and 8%, and used starch gelatinization temperatures of 70 ℃, 80 ℃, and 90 ℃. Different calcium cilicate concentration variations were used to optimize the mechanical and thermal properties, functional group contents, water resistance, and biodegradability of the degradable plastics.

2. Materials and methods

The methodology involved three steps: sago starch preparation, degradable plastics synthesis, and testing the resulting plastics. The equipment used in this research included a blender, a 5 mL volumetric pipette (IWAKI Merck), a digital laboratory drying oven (PRIO Merck), a thermometer (10–100 °C), a 250 mL glass pyrex beaker, a hotplate stirrer (MS H280 Pro), an erlenmeyer flask , a digital balance 100 gr × 0.0001 gr (0.1 mg) (OSUKA Merck), a 25 mm magnetic stirrer ball, a pyrex grade A volumetric flask, a sieve, and a glass casting (20 cm × 20 cm) mold. This study used sago starch, which contained about 24.07–26.11% amylose and 73.89–75.93% amylopectin [19]. Sago stems were taken from the sago plantation of the North Aceh province, Simpang Keramat sub-district, and starch was processed from the harvest. An aquadest of calcium silicate ($CaSiO_3$) (99% CAS-1344-95-2 Qianfang Merck) was retrieved from PT Bratachem in Surabaya, Indonesia, and sorbitol 99% was retrieved from Sighma-Aldrich 240850 CAS 50-70-4.

2.1. Synthesis of degradable plastics

Sago starch was made by cutting the sago stems, washing them until they were clean, and then mashing them with a blender, after which water is added. Next, after it became sago pulp, it was filtrated with a filtering sheet, and the filtrate was precipitated within 24 hours. The residue was removed from the liquid and dried in an oven to produce the starch. Next, the degradable plastic was processed with the calcium silicate filler. Sago starch was taken in 10 grams and placed in a glass beaker, and 200 mL of the aquades were used. Sago starch, water, and sorbitol (30% of the starch weight) were mixed, heated, and stirred at 100 rpm until cooked so that it became a gel at gelatination temperatures with various variations (70 °C, 80 °C, and 90 °C) and a time about ±30 minutes. Calcium silicate was added at various doses (2%, 4%, 6%, and 8% by weight of starch) and stirred until the solution was gelled within 40 minutes, where the temperature did not exceed 85 °C. Next, the molding step used glass castings that were dried in an oven for 6 hours at a temperature of 60 °C.

2.2. Characterization and testing

The mechanical properties of sago starch-based degradable plastics with calcium silicate were measured using the tensile strength, the elongation at break, and the modulus of the elasticity. This test used the American Standard Testing and Materials (ASTM) D-638. ASTM D-638 is in the form of a dumbbell with a total length of 165 mm, a thickness of 3.2 mm (1/8 inch) and a gauge length of 50 mm (2 inches), and can be determined by Eqs 1 and 2:

$$\sigma = \frac{F_{maks}}{A} \tag{1}$$

where σ represents the tensile strength (MPa), $Fmaks$ represents the maximum stress (N), and A represents the cross-area of the film under stress (mm^2), and

$$\varepsilon = \frac{\triangle l}{l_0} \tag{2}$$

where ε represents the strain (MPa), l is utilized for the measuring length, which is the length measured by the sample test at elongation (mm^2), and l0 is utilized for the measured length of the initial sample (mm^2).

Fourier Transform Infrared Spectroscopy (FTIR) can be used to determine chemically linked organic materials, polymers, metals, and various materials. An FTIR analysis uses infrared radiation

to scan the sample and observe chemical characteristics. With infrared radiation, the absorbed IR is usually responsible for the molecule existing in a high-vibration state. The wavelength absorption by sampling is a characteristic of the structure of the molecule. In this study, the FTIR was analyzed at a 550–4000 cm^{-1} cm frequency. Differential Scanning Calorimetry (DSC) was used to study the thermal characteristics of degradable plastics and to measure the heat energy. It shows how the polymer reacts to heat during the melting process. This study used a DSC 8000 PerkinElmer DSC with ASTM D3418-15 results.

The swell test was used to analyze the degradable plastics' resistance to water absorption. ASTM D2765 determined the swelling rate. The plastic sample was weighed and put into a solvent for 24 hours. Then, it was reweighed when it swelled and dried, in which the final weight was found. The swelling rate was determined using Eq 3:

$$\text{Swelling Degree} \quad \frac{\text{Weight of extended sample}-\text{Weight of initial sample}}{\text{Initial sample}} \times 100\% \qquad (3)$$

The microbial degradation rate was analyzed by burying the plastic in the soil. Following ASTM G-21-70, a biodegradability analysis was performed by measuring the direct contact of the degradable plastic with the soil. A 5×2 cm plastic sample was cut and weighed to obtain the initial mass (M_0). Then, the sample was buried 30 cm deep in soil, and the inspections were conducted over 4 days. After this period, the sample was removed from the soil and cleaned of any remaining soil. The sample was weighed again to determine the final mass (M_1). The biodegradability of the plastic was calculated using Eq 4:

$$\text{Biodegradabillity (\%)} = \frac{M0 - M1}{M0} \times 100\% \qquad (4)$$

where M_0 represents the initial mass (grams), and M_1 represents the final mass (grams).

3. Results

3.1. Mechanical properties analysis with tensile strength, elongation at break and Young's modulus

The mechanical properties of the sago starch-based degradable plastics with various calcium silicate fillers (2%, 4%, 6%, and 8% by weight of starch) and gelatinization at 90 ℃ were tested. The tensile strength was inversely proportional to the elongation rate, and the Young's modulus was proportional to the elongation rate.
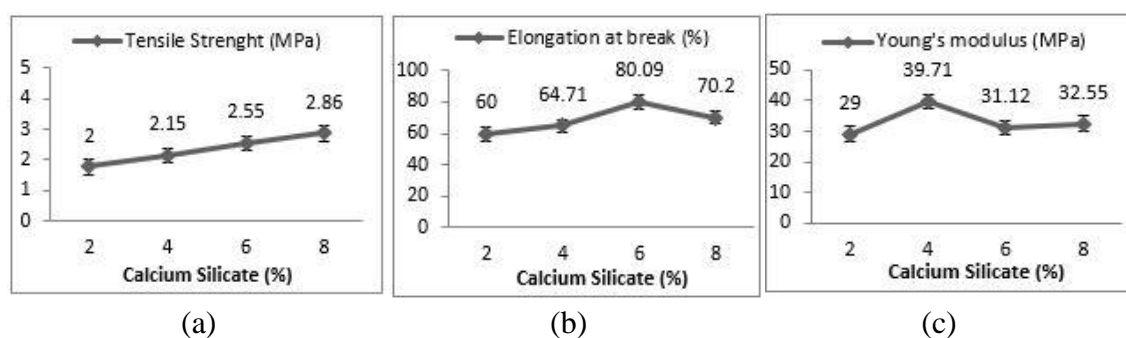

(a) (b) (c)

Figure 1. Effect of calcium silicate percentage on mechanical properties for degradable plastics: (a) Tensile strength (MPa), (b) elongation at break (%), and (c) Young's modulus (MPa).

3.2. Chemical properties analysis with FTIR

Figure 2 illustrates the results of the FTIR on the sago starch-based degradable plastic samples with the calcium silicate filler. The FTIR analysis was tested on plastic samples from the 8% calcium silicate variation with a starch gelatinization temperature of 90 °C.
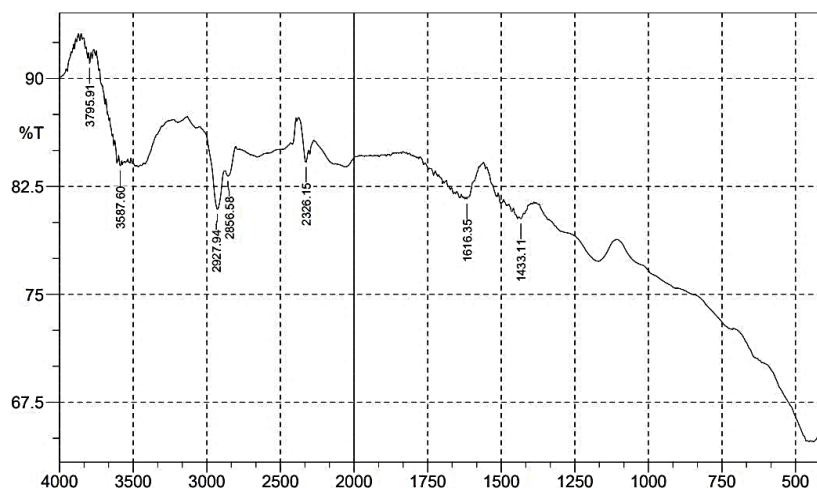

Figure 2. FTIR results on sago starch-based degradable plastics.

3.3. Energy absorption analysis with DSC

The results obtained from the test conducted on the sago starch-based degradable plastic using calcium silicate can be observed in Figure 3. The DSC analysis was tested on plastic samples from the 8% calcium silicate variation with a starch gelatinization temperature of 90 °C.
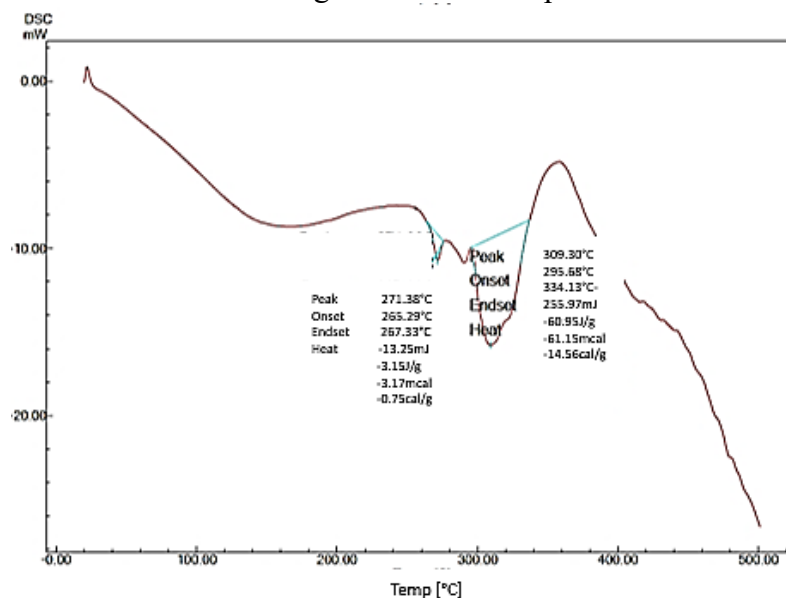

Figure 3. DSC results on sago starch-based degradable plastics.

3.4. Water absorption analysis with swelling degree

Figure 4 shows the water resistance test results of the sago starch plastic with a calcium

silicate filler (2%, 4%, 6%, 8%) using various gelatinization temperatures (70 °C, 80 °C, 90 °C) with swelling tests.
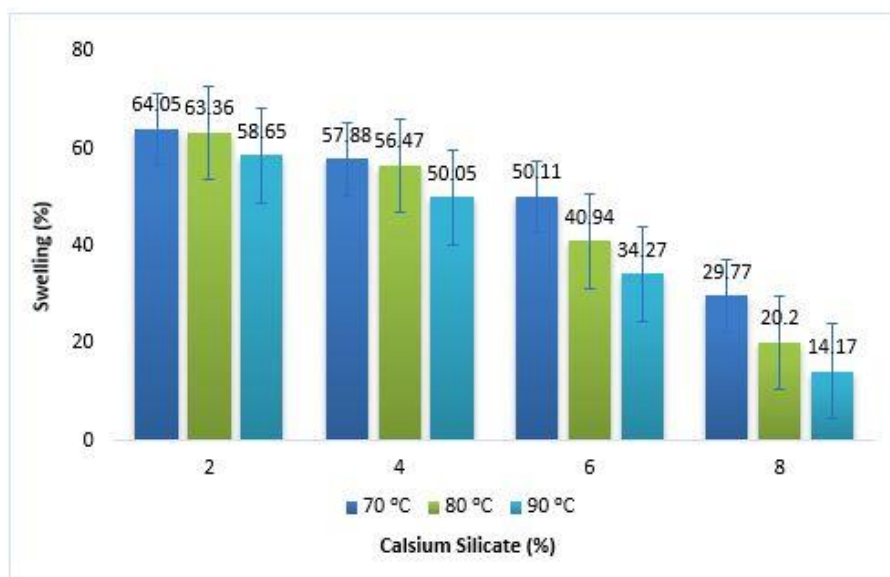

Figure 4. Water absorption of sago starch degradable plastic with variation of calcium silicate filler and gelatinization temperature.

3.5. Analysis of biodegradation rate by burial in soil

The results of the biodegradability test of the sago starch degradable plastic with a calcium silicate filler (2%, 4%, 6%, 8%) and various gelatinization temperature (70 °C, 80 °C, 90 °C) are listed in Table 1.

Table 1. Biodegradability test results on sago starch degradable plastics by soil burial.

| Temperature (°C) | Filler % | Biodegradability Test (%) Days | | | | |
		3	6	9	12	15
70	2	31.73	55.73	77.49	100	100
	4	29.45	42.81	83.22	100	100
	6	23.62	42.48	60.44	78.29	100
	8	23.64	31.93	77.45	87.54	100
80	2	30.47	52.37	89.46	100	100
	4	26.93	43.54	76.32	100	100
	6	20.71	36.76	59.87	84.61	100
	8	17.53	35.71	46.87	79.73	100
90	2	30.2	52.16	87.76	100	100
	4	27.72	46.66	74.19	87.61	100
	6	25.16	44.68	71.47	86.82	100
	8	22.38	42.84	70.7	86.28	100

4. Discussion

4.1. Mechanical properties

Figure 1(a) illustrates the tensile strength values of the sago starch-based degradable plastics with calcium silicate variations obtained in this study, which ranged from 17.35–28.04 MPa. Adding a calcium silicate filler can improve the mechanical characteristics of degradable plastics. The higher the calcium silicate concentrate used, the higher the tensile strength obtained. From the analyzed data, the mechanical characteristics were a tensile strength of 28.04 MPa, 32.55 MPa of elongation at the break, and 70.02% of Young's modulus obtained with the addition of 8% calcium silicate and a gelatinization temperature of 90 °C. In a previous study, bioplastics from cassava peels with variations of glycerol and calcium carbonate ($CaCO_3$) as fillers were prepared. Through tensile tests, it was shown that bioplastics with 20% glycerol and the addition of 0.2 g calcium carbonate showed the best results up to 5.82 ± 1.00 MPa [20]. Based on past research, a $CaCO_3$ filler was added at various mass percentages, ranging from 5% to 30%. A further addition of $CaCO_3$ tends to decrease the tensile strength up to 12.0 MPa for a 10% $CaCO_3$ addition. Even among the Low Linear Density Polyethylene/Thermoplastic Starch (LLDPE/TPS) composites, the 70:30 LLDPE/TPS composite with 10% $CaCO_3$ showed the best tensile strength [21]. It is thought that the greater the tensile strength value, the better the ability of the plastic to protect the product from mechanical factors such as mechanical force (drop and scrap), vibration, and impact, among others [22]. The tensile strength of the Moderate Properties group was between 10–100 MPa. The Standar Nasional Indonesia (SNI) standard concluded that the tensile strength for plastic is 24.7–302 MPa. The concentration of calcium silicate filler affects the tensile strength value; the higher the concentration of calcium silicate, the higher the tensile strength. This evidence indicates that the degradable plastic structure is amorphous. An amorphous molecular structure contains numerous branches but lacks a high degree of structural organization, resulting in a significant distance between the molecules and a relatively weak bond between them. The weak molecular bond in bioplastic reduces the force required to disrupt the plastic's structure. Therefore, the problem of filler agglomeration can be overcome if a good mixing technique is used, where the formation of stress concentration points can be avoided and the load distribution can be well spread [23]. The mechanical properties of the materials, including the strength, stiffness, ductility, and toughness, serve as the determining factors in their performance across a range of applications. The enhancement of these properties can facilitate the optimization of a product and the process reliability, durability, and efficiency. In a previous study [20], the bioplastic film exhibited an increased tensile strength with an increased concentration of bentonite used in taro starch-based bioplastics. This research is similar to previous studies . The potential of blending it with 5% to 20% polyethylene terephthalate (PET) has been previously explored. The report showed that the tensile strengths were 12.89, 12.65, 10.97, 12.07, and 13.4 MPa.

Figure 1(b) shows that the sago starch + calcium silicate filler-based degradable plastic had the highest elongation test values, with elongation values ranging from 59.56–70.2%. The concentration of calcium silicate filler affects the elongation result; the higher the concentration of calcium silicate, the higher the elongation percentage. Modifying silica was found to act as a filler that increased the tensile strength of sago starch-based bioplastics. The modified sago suggests that starch plastic films have a greater elongation at the break compared to unmodified plastic films. Modified sago starch may exhibit an enhanced compatibility and interaction properties. It is estimated that the SNI value for bioplastics elongation may be within a range of 21 to 220%. The elongation value obtained in this study aligns with the expectations set forth by the SNI standards. Additionally, it should be noted that the

elongation values obtained are almost comparable to PET, with a range from 15 to 165% [24].

Tian et al. (2024) reported that the sweet potato starch-oxalic acid/nanofibrillated cellulose (SPS-OA/NFC) composite film exhibited a notable enhancement in the tensile strength and tensile modulus, increasing from 1.46 MPa to 13.7 MPa and 6.04 MPa to 531.33 MPa, respectively, in comparison to the neat SPS film. The contact angle of the SPS/CA-NFC6 film was approximately 79% higher than that of the neat SPS film. In general, the optimal NFC addition is 6 wt%. This study provides guidance for the production of a new type of starch-based material with a high mechanical strength and transparency [25]. A study by Kong et al. (2023) showed that an esterified NFC can improve the mechanical properties and compatibility of highly filled poly(butylene adipate co-terephthalate) (PBAT) composite films. The effects of the NFC content and ball-milling treatment were studied. The amphiphilic esterified NFC acted as a bridge between the TPS and PBAT interfaces, thus improving the compatibility and mechanical properties [26].

When the pressure or stress is stopped, the material will return to its original position. The ratio of stress and strain is constant, and the same amount is referred to as the modulus of elasticity. The elastic modulus of a material may be similar to its elongation value. The modulus of elasticity measures how stiff a material can be stretched. This is related to the measure of stress and strain of a material. In Table 1 and Figure 1(c), the elastic modulus values of sago starch-based degradable plastics using a calcium silicate filler were between 29.16–32.55 MPa. An increase in the filler content resulted in an elevated value of the modulus of elasticity. In the absence of a modification to the adhesion between the polymer matrix and the filler, brittleness was observed despite an increase in stiffness, which was manifested in a reduction in the tensile strength and often a significant reduction in deformability [27]. However, the value obtained was the highest in degradable plastics using 4% calcium silicate. This could be a factor of uneven stirring and molding of the plastic samples [28]. The elastic modulus obtained was not comparable to that of a Polyethylene Bralen RA 2-63 foil (222.73 MPa to 298.24 MPa) [29]. The higher the elastic modulus, the stiffer the material. The fillers used affect the elastic modulus of the bioplastics. As previously reported [10], fillers have resilience to a strong and stiff biodegradable composite elasticity when utilizing lime and calcium carbonate. This indicates the balance of Young's modulus and the fracture strength for this series of composite polymers is delicate and results in industrially relevant mechanical characteristics.

4.2. Chemical properties

As depicted in Figure 2, the FTIR test was conducted on sago starch-based biodegradable plastic samples when a   calcium silicate filler was incorporated. As expected, the FTIR spectrum exhibited characteristic peaks indicative of sago starch and calcium silicate, which confirmed the concentration of the calcium silicate filler. Degradable plastics have precise wave number peaks with varying intensities. Based on Figure 2, it can be seen that the -OH group occured around the wave number 3587.60 cm^{-1}. The figure illustrates the hydroxyl group and -OH stretching vibration associated with the hydrogen bond in sago starch. The intensity of the -OH peak was observed to increase following the addition of calcium silicate at a wave number of 3795.91 cm^{-1}. Incorporating calcium silicate into the sago starch matrix indicates the interaction between the silicon group of calcium silicate and the hydroxyl group. It has been reported [20] that the same peak has a low intensity, attributed to C=O stretching in taro starch. This result indicates that the peak intensity was enhanced by adding bentonite to the taro starch matrix. This observation suggests a potential interaction between the aluminum (Al) groups of bentonite and the hydroxide (OH) groups, which could be responsible for the excellent biodegradability properties of taro starch-based films. The cluster content in the FTIR analysis of sago

starch degradable plastic with calcium silicate filler are organic groups. The plastic is hydrophilic, binds to water, and has ester groups such as O-H and C-O. With the presence of these groups, degradable plastics are easily degraded by soil. Furthermore, this implies that degradable plastics exhibit an elevated concentration of unbound hydroxyl (-OH) groups, which result from a reduction in the number of atoms capable of hydrogen bonding. A previous study [30] posited that the hydrogen bond between the OH group of starch and the COOH group of Cellulose Nano Crystals (CNC) exhibited a considerable tensile strength and a comparatively low elongation value. In contrast, the FTIR spectra of the degradable plastic from sago starch exhibited minimal variations with varying calcium cilicate ratios. This observation indicated the potential for interactions between the block copolymer and the calsium cilicate components [31].

4.3. Thermal properties

DSCs monitor the weight change as a function of temperature at a predetermined heating rate to evaluate the thermal stability of degradable plastics. The addition of extractive substances does not significantly affect the thermal properties of a degradable plastic-based sago starch, while the addition of cellulose improves the decomposition temperature of the degradable plastic. Figure 3 illustrates the reversible changes that occur in a hydrated substance after heating. Dehydration is the initial process indicated by the endotherm. A dehydrated substance undergoes a polymeric transmission, namely an endotherm, at elevated temperatures. Finally, the sample is melted, which results in a second endotherm. When cooled, the melted crystallizes, as indicated by the exothermic peak, and polymorphic changes also occur exothermically, though rehydration does not occur. If heating is endothermic, then cooling should be exothermic in this process [32]. The thermogram of the degraded plastic sample from sago starch with the addition of calcium silicate filler showed several peaks, namely at 271.38 °C, thereby showing a very sharp thermogram peak. This peak displayed physical changes, namely the loss of water groups, where water evaporates at 100 °C. The water molecules were no longer attached to the sample surface. At a temperature of 309.30 °C, the endothermic peak arised, which indicated a glass transition state. This is important when making composite grids using degradable plastics. A study by Lang et al. showed that adding microcrystalline cellulose (MCC) improved the thermal stability and resistance. For example, the T_{max} increased by 1 °C and T_{onset} increased by 2–8 °C. MCC can be used with PBAT and TPS if the MCC content is below 6%. Adding 6% MCC shifts the endothermic peak to a higher temperature. This is probably because hydrogen bonds form between the matrix and the MCC. The Xc was almost the same as CI, which showed that the composite could be easily processed. MCC had little effect on the composite melting transition [33].

Adding fillers to the matrix increased the melting point of the degradable resin, thus indicating crosslinking. More crosslinking occurs as the melting point increases. The hydrogen bonds in the plastic affect the melting point. The DSC analysis showed good thermal properties and thr stability of sago starch-based degradable plastics with calcium silicate as the filler and starch as the matrix. The more hydrogen bonds in the plastic, the higher the melting point. This is because the energy required to break the bond will also be greater [34]. Adding polyurethane increases the melting point of bioplastics because the melting point of the added polyurethane is higher than the melting point of the starch [35]. The addition of $CaCO_3$ improved the crystallinity and thermal stability for the poly(L-lactide)-b-poly(ethylene glycol)-b-poly(L-lactide)(PLLA-b-PEG-b-PLLA) matrix but not for the poly(L-lactide) (PLLA) matrix, thus confirming that $CaCO_3$ acts as a heterogeneous nucleating agent that enhances copolymer catalysis [31].

4.4. Water absorption

Degradable materials must absorb water and degrade. The growth and consumption of materials by microorganisms occurs when the materials have absorbed water, which serves as the microorganisms' source of energy [7]. Figure 4 shows the swelling value obtained in the sago starch degradable plastics with a potassium silicate filler at temperatures of 70 °C (29.77%–64.05%), 80 °C (20.2%–63.36%), and temperature 90 °C (14.17%–58.65%). The swelling value obtained from the difference in gelatinization temperature tended to be high. The calcium silicate filler concentration greatly influences the water-resistant characteristics of degradable plastics. A greater swelling percentage was obtained in the sago starch-based plastics with a 2% filler at all the gelatinization temperature ranges used. The starch's hydrophilic nature and high moisture flexibility lead to easy water movements into the starch-soaked area during soaking. Additionally, the use of plasticizers affects the water resistance of degradable plastics [36]. Previous research used calcium carbonate and glycerol in cassava-based bioplastics. The water absorption test showed that 40% glycerol absorbed the most, with a value of 28.64%. The lowest absorption rate was at 20% glycerol, with a $24.49 \pm 0.21\%$ value. As the glycerol concentration increased, so did the absorption. Cassava starch is hydrophilic because it contains hydroxyl, carbonyl, and ester groups, and $CaCO_3$ is hydrophobic, which means it does not let water in [37]. As in previous studies, the development of bioplastic films from taro starch with bentonite reinforcement showed an improved resistance to salt and acid, though it was susceptible to alkali and showed little swelling. Soil degradability tests showed that the film was degradable and could be substituted for conventional plastics [20]. The incorporation of fillers into bioplastics production is due to the renewable properties of such materials, beneficial biodegradability properties, and an abundant availability. Adding fillers in bioplastics can improve mechanical properties such as the stiffness, strength, gas resistance, melting resistance, thermal stability, and so on [38]. As stated by Ismail and Zaaba [7], modified sago starch plastic films have a decreased water absorption, thus making them more hydrophobic. This is due to the replacement of the hydroxyl groups present in starch molecules by phosphate groups, thus enhancing the film's resistance to water.

4.5. Biodegradability rate

Soil microbes help break down plastic when it is buried. To determine an appropriate burial period for a given material and application of a plastic product, the samples should first be buried in the soil of a known weight for a specified time interval. This research used sago starch as a base material with the addition of a calcium silicate filler (2%, 4%, 6%, 8%) and various gelatinization temperatures (70 °C, 80 °C, 90 °C) to see the effect of the resulting degradation rate. Table 1 shows that degradable plastic from sago starch with calcium silicate filler and a gelatinization temperature completely decomposed in the soil in 12–15 days. The addition of a calcium silicate filler in amounts of up to 2% and 4% by weight of the sago starch-based plastic resulted in a notable acceleration in the decomposition rate, which was observed to reach completion within 12 days. The more calcium silicate filler used, the slower the degraded. Variations in the gelatinization temperature did not affect the time it took plastic to degrade. The difference in the sample size and preparation meant that the claim that the addition of calcium silicate accelerated the biodegradation of degradable plastics. Other differences such as the variation of calcium silicate as a filler did affect the biodegradable characteristics of the overall degradable plastic sample [10].

The ASTM D-6002 standard, which is a guide for assessing the compostability of environmentally degradable plastics in conjunction with ASTM D-20.96 on environmentally

degradable plastics, defines the criteria for degradable plastics. Products comprised of a single polymer (homopolymer or random copolymer) must have at least 60% organic carbon converted to carbon dioxide within 180 days to meet the standard. Research conducted by [14] showed that using a bentonite filler in taro starch-based bioplastics affected the degradation rate. The more bentonite that was added, the slower the bioplastics degraded. The dosage of the filler material type affected the composite's overall biodegradability properties [9]. The material used in the polymer, the soil moisture, and the environmental conditions all affected the degradation rate of degradable plastics [39]. A recent study [40] demonstrated that adding either starch, cellulose, or cellulose derivatives to PVA resulted in an enhanced solubility and biodegradability of the plastic in soil. Lignin in cellulose fibers can stop them from breaking down. Lignin can be removed by using several lignin-degrading compounds, one of which is NaOH, which significantly affects lignin removal by delignification.

5. Conclusions

Sago starch-based degradable plastics with calcium silicate filler have good tensile strengths, elongation, and modulus of elasticity. A tensile strength test was conducted on the material in question, and the results ranged from 17.35 to 28.04 MPa. From the data, the mechanical characteristics were a tensile strength of 28.04 MPa, 32.55 MPa of elongation at the break, and 70.02% of Young's modulus obtained at the addition of 8% calcium silicate and a gelatinization temperature of 90 °C. The FTIR analysis of the sago starch-based plastic with a calcium silicate filler showed organic groups that made it hydrophilic and ester groups such as O-H and C-O. Thermal characterization by a DSC thermogram test showed that the sago starch degradable plastic with a calcium silicate filler had thermogram peaks at 271.38 °C and 309.30 °C; subsequently, the endothermic peak arose, which indicated a glass transition state. This point is of a significant importance for preparing compound lattices of degradable plastics. The swelling value obtained in sago starch degradable plastics was measured at temperatures of 70 °C (29.77%–64.05%), 80 °C (20.2%–63.36%), and 90 °C (14.17%–58.65%). The degradation of sago starch degradable plastics using a calcium silicate filler (2%, 4%, 6%, and 8%) completely decomposed at 12–15 days and is by ASTM D-20.96 for degradable plastics, which should be decomposed before 180 days. More calcium silicate fillers in the degradable plastic led to an increased degradation time in the soil.

The process technology for manufacturing degradable plastics via thermopressing offers optimal outcomes, thus enabling direct comparison with commercial/conventional plastics. These biodegradable plastics can subsequently be utilized in household products that are environmentally and human health-safe. The biodegradable plastic can be manufactured using a natural base material derived from the sago tree, specifically the trunk, which is capable of producing the starch. This has the potential to enhance the economic standing of sago farmers and traders, as it allows for the effective utilisation of sago trees that have not been fully exploited. This presents a promising avenue for the development of degradable plastic products that can replace commercial/conventional plastics, which have been identified as having an adverse impact on environmental hygiene and human health.

Use of AI tools declaration

The authors declare they have not used Artificial Intelligence (AI) tools in the creation of this article.

Acknowledgments

Conflict of interest

The authors declare no conflict of interest.

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