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Effect of Cellulose-to-Starch Ratio and Sorbitol Concentration on the Mechanical, Physical, and Biodegradation Properties of OPEFB-Based Bioplastics

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ABSTRACT

The extensive use of conventional plastics, which are difficult to degrade, has driven the development of environmentally friendly bioplastics derived from oil palm empty fruit bunches (OPEFB). The independent variables were the OPEFB Cellulose-to-Starch ratio (1:2, 1:1, and 2:1) and sorbitol concentration (5%, 10%, 20%, and 30% w/w) in OPEFB-derived cellulose matrices reinforced with cellulose nanocrystals (CNC) and polyvinyl alcohol (PVA). The bioplastics were prepared using the solution casting method. The results indicate that the optimal formulation for water resistance was achieved at a 2:1 cellulose-to-starch ratio with 5% sorbitol, yielding the lowest water absorption of $2.56\% \pm 0.12\%$ and an outstanding water resistance of 97.44%. Conversely, the highest tensile strength of $44.16 \pm 1.45 \text{ MPa}$ was obtained at a 1:2 ratio with 30% sorbitol. The highest biodegradability in the soil burial test was observed at 30% sorbitol, with a maximum weight loss of $66.16\% \pm 2.11\%$ within 14 days. FTIR analysis confirmed the typical preservation of polysaccharide functional groups. At the same time, SEM observations at higher magnification revealed a well-distributed nanostructure with a controlled porous morphology that effectively balances mechanical performance and accelerated microbial degradation.

1. Introduction

Plastic waste has become a serious global environmental problem due to its extensive use and resistance to natural degradation. Conventional petroleum-based plastics persist in the environment for decades, leading to severe soil, water, and marine pollution [1]. These challenges have intensified research efforts toward developing biodegradable and renewable alternatives, particularly bioplastics derived from natural lignocellulosic biomass. Bioplastics are commonly produced from polysaccharides such as starch, cellulose, and chitosan because of their biodegradability, renewability, and environmental compatibility [2]. Among these materials, cellulose is considered a promising bioplastic component due to its high tensile strength, thermal stability, and ability to form strong intermolecular hydrogen bonds [3]. Indonesia has abundant lignocellulosic biomass resources, including oil palm empty fruit bunches (OPEFB), a major by-product of the palm oil industry. Oil palm empty fruit bunches (OPEFB) are an abundant industrial by-product in Indonesia, with a high cellulose content ranging from 23.7% to 65.0%, making them a highly sustainable raw material [4].

However, cellulose-based bioplastics often exhibit limitations, including brittleness and low flexibility. To overcome the inherent brittleness of pure cellulose-based films, plasticizers like sorbitol are commonly incorporated to reduce intermolecular forces and improve flexibility. Sorbitol is a polyol plasticizer that has been widely reported to improve flexibility and elongation by reducing intermolecular forces between polymer chains [5]. Nevertheless, excessive sorbitol addition may reduce tensile strength and water resistance due to increased hydrophilicity.

Additionally, components such as polyvinyl alcohol (PVA) and cellulose nanocrystals (CNC) are frequently added as reinforcing agents to achieve high interfacial adhesion and efficient stress transfer within the polymer matrix. Polyvinyl alcohol (PVA) is known for its good film-forming ability and compatibility with cellulose and starch matrices, resulting in improved homogeneity and mechanical stability. Meanwhile, CNC particles exhibit an exceptionally high aspect ratio and act as efficient stress-transfer agents that crosslink within the blended polymer network via strong, extensive hydrogen bonding [6]. Previous studies have reported that incorporating CNC and PVA into bioplastic formulations can increase tensile strength to approximately 32.5 MPa while maintaining biodegradability [7]. Despite extensive research on cellulose-based bioplastics, studies focusing on the combined effects of cellulose–starch ratio and sorbitol concentration in OPEFB-derived cellulose bioplastics reinforced with CNC and PVA remain limited. Most existing studies examine these components individually, with limited comparative analysis of their synergistic effects on mechanical properties, water absorption, and biodegradation behavior.

Therefore, this study aims to evaluate the effects of varying cellulose–starch ratios (1:2, 1:1, and 2:1) and sorbitol concentrations (5-30%) on the physical, mechanical, and biodegradation properties of OPEFB-based bioplastics reinforced with CNC and PVA. The bioplastics were prepared using the solution casting method and characterized through tensile strength, water absorption, biodegradation, FTIR, and SEM analyses. This research is expected to contribute to the development of optimized OPEFB-based bioplastics with balanced mechanical strength, water resistance, and biodegradability, thereby advancing sustainable plastic alternatives.

2. Research Methodology

2.1. Materials

The materials used in this study included oil palm empty fruit bunches (OPEFB) as a cellulose source, sodium hydroxide (NaOH) for delignification and bleaching, starch (tapioca) as a supporting material, sorbitol as a plasticizer, cellulose nanocrystals (CNC) as a reinforcing agent, polyvinyl alcohol (PVA) as an additional matrix, and distilled water as a solvent [8].

2.2. Procedures

The study began with the preparation of oil palm empty fruit bunches (OPEFB), which were washed, dried for 24 hours, ground, and sieved using a 100-mesh sieve to obtain fine powder. The materials used included OPEFB as the cellulose source; sodium hydroxide (NaOH) for delignification; hydrogen peroxide (H_2O_2) for bleaching; tapioca starch, sorbitol, CNC, PVA, and distilled water. Cellulose extraction involved three stages: pre-delignification (2% NaOH, 50°C, 1 h), reflux delignification (12% NaOH, 80-90°C, 1 h), and bleaching (2% H_2O_2, 60 °C, 2 h). The final purified cellulose powder was dried at 100°C for 6 hours [9].

Bioplastic film preparation was performed by heating a mixture of tapioca starch and CNC at 80°C until gelatinization occurred, then mixing it with a previously dissolved PVA solution. OPEFB cellulose was added according to the specified ratios (1:2, 1:1, and 2:1), along with sorbitol at concentrations of 5-30%. The mixture was then heated at 70°C with stirring at 320 rpm for 60 minutes. The resulting solution was cast onto glass plates and dried for 12 hours to form bioplastic films. The films were subsequently characterized through tensile strength testing, water absorption testing, and biodegradability testing using the soil burial method. Biodegradability was measured using a controlled soil burial method over a strict 14-day duration inside compost soil at a stabilized moisture level (60-70%).

3. Results and Discussion

3.1. Characteristics of Cellulose Extracted from OPEFB

Cellulose extraction from oil palm empty fruit bunches (OPEFB) was carried out via delignification with a NaOH solution, followed by bleaching. During the delignification stage, cleavage of lignin bonds caused the color of OPEFB to change from dark brown to a lighter shade. The systematic removal of non-cellulosic components was visually validated by the distinct transition in fiber color from a dark brown raw state to a highly purified, clean, light-brown color post-bleaching. These changes in color and texture indicate that the separation of lignin and hemicellulose proceeded effectively. This finding is consistent with the study by [4], which reported that achieving this high purity is critical; removing brittle, hydrophobic amorphous lignin patches allows the native crystalline cellulose domains to form unhindered, continuous, and highly compatible networks with the surrounding starch and PVA macromolecules during blending. Cellulose purity plays a crucial role in determining the mechanical properties of bioplastics. Cellulose that still contains lignin tends to produce brittle, nonhomogeneous bioplastic films. Therefore, the presence of clean, smooth, and lightly colored cellulose indicates that the extraction process was successful and met the requirements for bioplastic raw materials.

Fig. 1. OPEFB before delignification and bleaching

Fig. 2. OPEFB after delignification and bleaching

3.2. Effect of Sorbitol Variation on the Mechanical Properties of Bioplastics

Sorbitol acts as a plasticizer that reduces intermolecular forces between polymer chains, resulting in more flexible bioplastics. In formulations with higher sorbitol content, the bioplastics exhibited increased flexibility and elongation. This behavior occurs because sorbitol penetrates the polymer structure, increasing the distance between polymer chains and thereby facilitating chain mobility and deformation. However, excessive sorbitol addition can reduce tensile strength. This is attributed to the weakening of internal cellulose–starch interactions due to excessive sorbitol, which makes the structure softer and less stable. This finding is consistent with the report by [5], which emphasized that an optimum sorbitol concentration is required to maintain a balance between flexibility and tensile strength. Overall, sorbitol variation significantly affects the mechanical properties of bioplastics. Moderate sorbitol concentrations produce bioplastics with the best combination of flexibility and tensile strength.

3.3. Effect of CNC and PVA Addition on Tensile Strength

Cellulose nanocrystals (CNC) and polyvinyl alcohol (PVA) were added as reinforcing agents to enhance the mechanical properties of the bioplastics. CNC has a nanometer-scale size and a highly crystalline structure, thereby enhancing tensile strength by forming strong hydrogen bonds with cellulose. The incorporation of CNC into the formulation resulted in bioplastics with a smoother surface and a more compact structure.

PVA acts as a compatible co-polymer with cellulose and starch. It enhances the homogeneity of the blend and improves the dispersion of CNC within the polymer matrix. The combination of PVA and CNC has been shown to increase tensile strength significantly. This finding is consistent with the study by [6], which reported a tensile strength of up to 32.5 MPa in bioplastics containing a combination of PVA and CNC.

The highest tensile strength (44.16 MPa) was achieved by formulation A4 (1:2 ratio with 30% sorbitol). Mechanical profiles were determined using a Universal Testing Machine on standardized specimens (10 cm x 2.5 cm x 0.2 cm). This stands out compared to traditional unreinforced starch films, which typically fall below 15 MPa [6], and to PVA/CNC-based bioplastics, which achieved a tensile strength of approximately 32.5 MPa. The high mechanical strength is due to the synergistic interaction between the starch matrix, PVA, and highly crystalline CNC filler.

Fig. 3. Tensile strength analysis

Table 1. Tensile Strength Test Results of Biodegradable Plastics

No	Cellulose: Starch (g)	Sorbitol (%)	Tensile Strength Test (MPa)
A1	1: 2	5	20.41
A2	1: 2	10	37.47
A3	1: 2	20	41.02
A4	1: 2	30	44.16
B1	1: 1	5	29.15
B2	1: 1	10	25.87
B3	1: 1	20	26.96
B4	1: 1	30	23.53
C1	2:1	5	16.47
C2	2:1	10	36.48
C3	2:1	20	16.39
C4	2:1	30	25.12

3.4. Water Absorption

Water absorption is an important parameter in packaging applications. Cellulose and starch possess inherent hydrophilicity, resulting in relatively high water absorption capacity. The addition of sorbitol can further increase hydrophilicity due to its multiple hydroxyl groups. However, the incorporation of PVA and CNC can reduce water absorption, as both materials enhance intermolecular bonding density and promote the formation of a more compact polymer structure. A denser film structure decreases pore spaces, thereby limiting water penetration into the bioplastic matrix.

Fig. 4. Water absorption analysis

Formulations with low sorbitol content, high CNC loading, and moderate PVA levels generally exhibit the lowest water absorption values. This finding indicates that the composition of reinforcing materials plays a significant role in controlling water permeability.

Table 2. Water Absorption Test Results of Biodegradable Plastics

No	Cellulose: Starch (g)	Sorbitol (%)	Initial Weight (g)	Final Weight (g)	Water Absorption (%)	Water Resistance (%)
A1	1: 2	5	0.1088	0.1143	4.81	95.19
A2	1: 2	10	0.1059	0.1135	6.70	93.30
A3	1: 2	20	0.1071	0.1178	9.08	90.92
A4	1: 2	30	0.1012	0.1144	11.54	88.46
B1	1: 1	5	0.0956	0.0993	3.73	96.27
B2	1: 1	10	0.0982	0.104	5.58	94.42
B3	1: 1	20	0.0995	0.1087	8.46	91.54
B4	1: 1	30	0.0979	0.1101	11,08	88.92
C1	2: 1	5	0.095	0.0975	2.56	97.44
C2	2: 1	10	0.0901	0.0935	3.64	96.36
C3	2: 1	20	0.0937	0.0995	3.64	94.17
C4	2: 1	30	0.0998	0.111	10.09	89.91

Overall, the results of this study demonstrate a clear relationship: higher starch content and greater sorbitol addition increase water absorption in bioplastics. Starch contributes inherent hydrophilicity, while sorbitol loosens intermolecular interactions among polymer chains, facilitating water diffusion into the structure. In contrast, higher cellulose content results in more compact bioplastics with improved water resistance. In general, bioplastics with a higher cellulose ratio and lower sorbitol concentration are optimal for water resistance, whereas combinations with high starch and high sorbitol content yield bioplastics with the highest water absorption.

The lowest water absorption value (2.56%) was observed in bioplastics with a cellulose–starch ratio of 2:1 and 5% sorbitol. This value is lower than those reported in starch-based bioplastics without reinforcement, which typically exhibit water absorption above 10% [10]. The improved water resistance observed in this study can be attributed to the higher cellulose content and the presence of CNC and PVA, which together contribute to a denser, more compact polymer network.

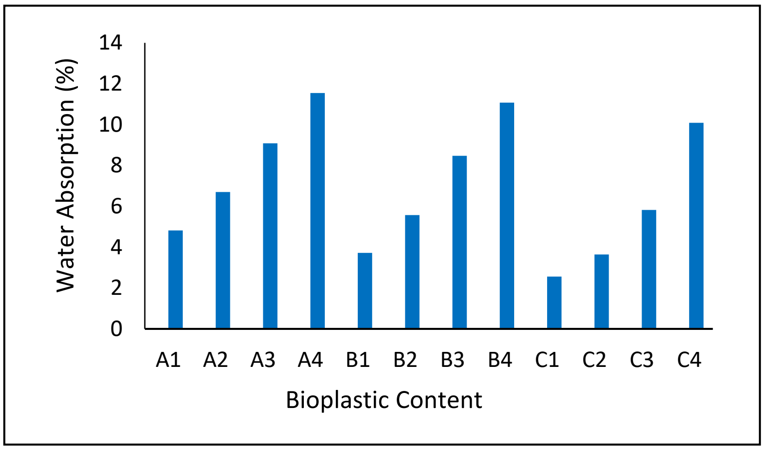

Fig. 5. Water absorption test results of biodegradable plastics

3.5. Biodegradation Test

The biodegradation test was conducted using the soil burial method, in which bioplastic samples were buried in moist soil for several days or weeks. Cellulose- and starch-based bioplastics exhibited a faster degradation rate than conventional plastics because microorganisms readily decompose both materials.

Bioplastics with high cellulose and starch contents tend to undergo color changes, mass reduction, and surface cracking during soil burial. Formulations with higher sorbitol content generally degrade more rapidly due to their softer structure, which is more susceptible to microbial attack. In contrast, the presence of PVA and CNC slightly slows the degradation rate by enhancing material strength. Overall, the bioplastics exhibit good biodegradability, indicating that the produced materials meet the research objective of developing environmentally friendly plastic alternatives.

Fig. 6. Biodegradation test analysis

Table 3. Biodegradability Test Results of Biodegradable Plastics

No	Cellulose: Starch (g)	Sorbitol (%)	Initial Weight (g)	Final Weight (g)	Weight Loss (%)
A1	1: 2	5	0.1240	0.06	51.61
A2	1: 2	10	0.0973	0.037	61.97
A3	1: 2	20	0.1004	0.034	66.14
A4	1: 2	30	0.1250	0.0423	66.16
B1	1: 1	5	0.0952	0.049	48.53
B2	1: 1	10	0.0951	0.0375	60.57
B3	1: 1	20	0.0994	0.035	64.79
B4	1: 1	30	0.0996	0.0337	66.16
C1	2:1	5	0.0841	0.0497	40.90
C2	2:1	10	0.1084	0.056	48.34
C3	2:1	20	0.1162	0.0466	59.90
C4	2:1	30	0.1221	0.042	65.60

Overall, the results of this study demonstrate that cellulose- and starch-based bioplastics have strong potential for rapid degradation in soil environments, particularly when sorbitol is added as a plasticizer at higher concentrations. The best biodegradability performance was observed in formulations A4, B4, and C4, which exhibited the highest weight loss values of approximately 66%, indicating excellent degradation capability. Bioplastics with higher sorbitol content showed greater mass loss during soil burial tests, reaching approximately 66%. This result is comparable to the biodegradation behavior reported by [11], where increased plasticizer content accelerated microbial degradation. The higher degradation rate observed in this study is likely due to increased hydrophilicity and porous morphology, which facilitate microbial penetration.

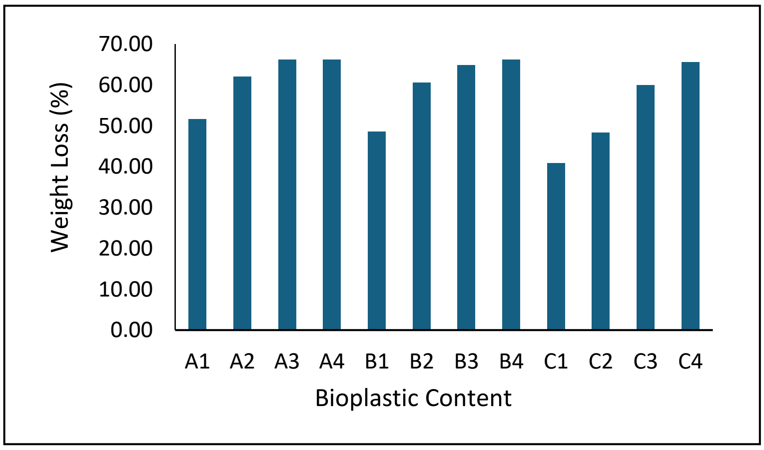

Fig. 7. Biodegradability test results of biodegradable plastics

3.6. FTIR Analysis

FTIR analysis was performed to identify the functional groups present in bioplastics derived from Oil Palm Empty Fruit Bunches (OPEFB) and to confirm the chemical characteristics of the bioplastic components. Chemical functional groups were verified through FTIR spectroscopy in the range 4000–500 cm^{-1}. Fig 8. FTIR analysis results of OPEFB-based bioplastic.

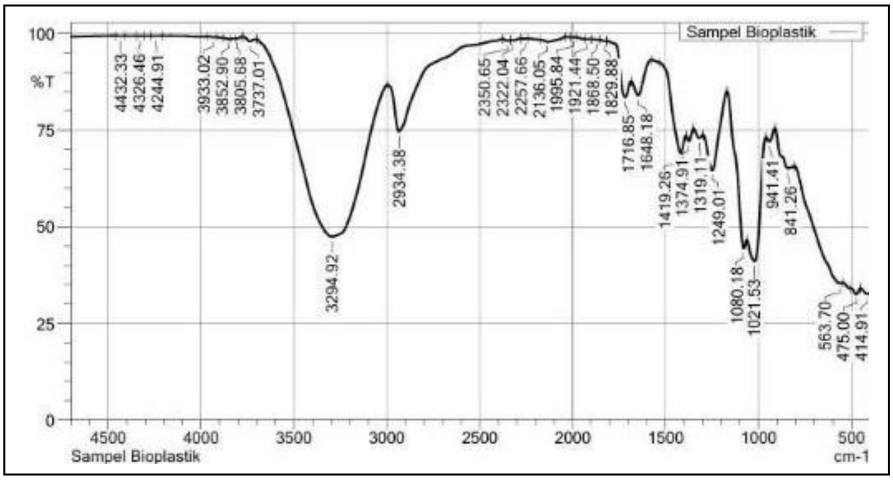

Fig. 8. FTIR analysis results of OPEFB-based bioplastic

The FTIR spectrum shows typical absorption bands of biomass-based bioplastics. The broad peak at around 3294 cm^{-1} indicates –OH stretching from cellulose and starch. The peak at 2934 cm^{-1} corresponds to C–H stretching vibrations. Absorption bands in the 1700–1600 cm^{-1} region are associated with C=O and O–H vibrations, while peaks at 1249–1021 cm^{-1} represent C–O and C–O–C stretching, confirming the presence of polysaccharide structures.

3.7. SEM Analysis

Scanning Electron Microscopy (SEM) analysis was conducted to observe the surface morphology and cross-sectional structure of the bioplastic based on Oil Palm Empty Fruit Bunches (OPEFB). Fig 9. Morphological characteristics were examined using Scanning Electron Microscopy (SEM) at both 250x and high-magnification 1000x levels. Shows the SEM micrograph of the sample.

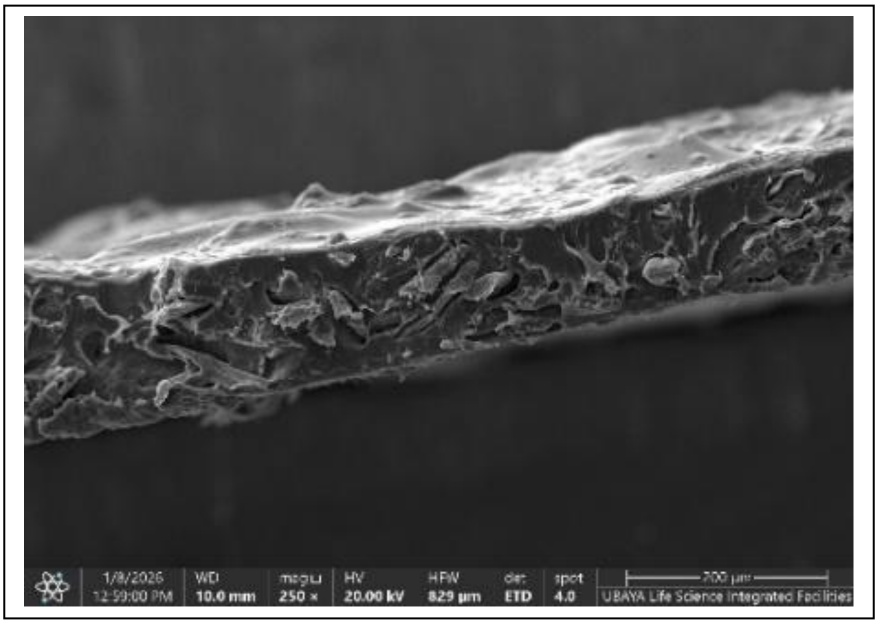

Fig. 9. SEM micrograph

The SEM micrograph at 250× magnification shows a relatively rough and heterogeneous cross-sectional morphology of the OPEFB-based bioplastic. The presence of pores and irregular cavities indicates incomplete packing of the polymer matrix and filler particles. These voids may be associated with the dispersion of cellulose fibers and plasticizer within the bioplastic matrix. Such morphological features can influence the mechanical strength and water absorption behavior of the material. Overall, the SEM results demonstrate that the bioplastic's microstructure is porous, which may enhance biodegradability but can reduce mechanical performance if voids are excessive. SEM observations revealed a porous, heterogeneous microstructure that supports biodegradation but may reduce mechanical strength if voids are excessive. Similar porous morphologies have been reported in cellulose–starch bioplastics by [12], who noted that pore formation enhances biodegradability by increasing the surface area exposed to microorganisms.

Table 4. Comparison of OPEFB test results after treatment with previous studies.

Raw material/Formulation	Tensile strength (MPa)	Water absorption (%)	Biodegradation (%)	Ref
Commercial plastic (LDPE)	8-12	<1	<5	[13]
Commercial PVA film	25-35	6-10	40-55	[14]
Lignocellulosic bioplastic (OPEFB-based)	36.2	5.4	63	[15]
OPEFB–Cellulose/Starch + Sorbitol + PVA + CNC	44.16	2.56	6.16	This Study

Table 4 compares the performance of the developed OPEFB-based bioplastic with commercial plastic materials. Conventional plastics such as LDPE exhibit low water absorption but extremely poor biodegradability, making them environmentally problematic. Commercial PVA films show better biodegradability but still suffer from relatively high water absorption and moderate tensile strength. In contrast, the OPEFB-based bioplastic developed in this study demonstrated superior tensile strength (44.16 MPa) and significantly lower water absorption (2.56%) while maintaining high biodegradability (66.16%). These improvements are attributed to the synergistic effect of cellulose reinforcement, CNC addition, and PVA compatibility within the polymer matrix. The results indicate that the developed bioplastic not only outperforms conventional plastics in environmental compatibility but also exhibits greater mechanical stability than commercial PVA films.

4. Conclusion

OPEFB-based bioplastics were successfully produced with varying cellulose–starch ratios and sorbitol contents. Sorbitol influenced flexibility and tensile strength, while the addition of CNC and PVA improved mechanical performance. Higher starch and sorbitol contents increased water absorption, whereas higher cellulose ratios enhanced water resistance. All samples showed good biodegradability, especially at higher sorbitol concentrations. FTIR and SEM analyses confirmed the presence of polysaccharide functional groups and a porous structure that supports biodegradation, indicating the potential of OPEFB-based bioplastics as environmentally friendly materials.

Notation

Sample Code Description

A = Cellulose: Starch (1:2)
A1 = Sorbitol 5%
A2 = Sorbitol 10%
A3 = Sorbitol 20%
A4 = Sorbitol 30%
B = Cellulose: Starch (1:1)
B1 = Sorbitol 5%
B2 = Sorbitol 10%
B3 = Sorbitol 20%
B4 = Sorbitol 30%
C = Cellulose: Starch (2:1)
C1 = Sorbitol 5%
C2 = Sorbitol 10%
C3 = Sorbitol 20%
C4 = Sorbitol 30%

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