Available online: journal.ipb.ac.id/index.php/jphpi

SYNERGISTIC FUNCTIONAL PROPERTIES OF HYDROXYPROPYL-MODIFIED STARCH AND IOTA CARRAGEENAN FOR PLANT-BASED SOFT CAPSULE SHELLS

Taufik Hidayat[1,2*], Khaswar Syamsu[1], Titi Candra Sunarti[1], Mala Nurilmala[3], Lamhot Parulian Manalu[2], Dayu Dian Perwatasari[4], Ramlan[3,5], Renny Primasari Gustia Putri[4]

[1]Department of Agro-Industrial Engineering, IPB University, West Java, Indonesia 16680
[2]Research Center for Equipment Manufacturing Technology, National Research and Innovation Agency, South Tangerang, Banten, Indonesia 15314
[3]Department of Aquatic Product Technology, IPB University, West Java, Indonesia 16680
[4]Research Center for Processing Technology, National Research and Innovation Agency, South Tangerang, Banten, Indonesia 15314
[5]Department of Food Science and Technology, IPB University, West Java, Indonesia 16680

Submitted: 28 November 2025/Accepted: 8 April 2026
*Correspondence: taufikh22taufik@apps.ipb.ac.id

How to cite (APA Style 7th): Hidayat, T., Syamsu, K., Sunarti, T. C., Nurilmala, M., Manalu, L. P., Perwatasari, D. D., Ramlan, & Putri, R. P. G. (2026). Synergistic functional properties of hydroxypropyl-modified starch and iota carrageenan for plant-based soft capsule shells. *Jurnal Pengolahan Hasil Perikanan Indonesia,* 29(4), 270-290. http://dx.doi.org/10.17844/h6tbjw36

Abstract

Soft capsules are widely used oral dosage forms; however, the reliance on gelatin has raised safety, religious, and ethical concerns, driving the need for plant-based alternatives. Starch, a biodegradable and renewable polysaccharide, is a promising candidate; however, it suffers from poor solubility, retrogradation, and instability under acidic or thermal conditions. Chemical modification, particularly hydroxypropylation, has been employed to overcome these drawbacks, and iota carrageenan offers additional benefits as a gelling agent that enhances elasticity and stability. This study aimed to determine the best functional and thermal properties of hydroxypropyl derivatives from cassava, sago, corn, and potato, and to examine their potential for soft capsule application. This study characterized various hydroxypropyl starch modifications and determined the characteristics of the modified starch that meet the raw material requirements for soft capsule shells using the Zeleny method. The analysis parameters included degree of substitution, water holding capacity, oil holding capacity, swelling power, solubility, pasting properties, differential scanning calorimetry, scanning electron microscopy, Fourier Transform Infrared Spectroscopy, iota carrageenan, and rapid visco analysis. The results showed that hydroxy-propyl cassava starch exhibited selected mechanical properties, with a DS value of 0.104, WHC of 4.21%, OHC of 5.43%, swelling of 24.23%, and solubility of 21.54%. Hydroxypropylation improved the solubility, thermal resistance, and water retention, with cassava starch exhibiting the highest degree of substitution and favorable film-forming ability. Furthermore, incorporating iota carrageenan into hydroxypropyl cassava starch generated homogeneous composite films with enhanced mechanical strength and stability. These findings highlight the synergistic potential of hydroxypropyl starch and iota carrageenan, providing a scientific basis for developing sustainable, plant-based soft capsule shells as viable alternatives to gelatin.

Keywords: cassava, functional, gelatin, polysaccharide, thermal

http://dx.doi.org/10.17844/h6tbjw36

Sifat Fungsional Sinergis Modifikasi Pati Hidroksipropil dan Iota Karageenan untuk Kulit Kapsul Lunak Berbasis Tumbuhan

Abstrak

Kapsul lunak merupakan bentuk sediaan oral yang banyak digunakan, namun ketergantungan pada gelatin telah menimbulkan kekhawatiran akan keamanan, agama, dan etika, yang mendorong kebutuhan akan alternatif berbasis tumbuhan. Pati, polisakarida yang dapat terurai secara hayati dan terbarukan, merupakan kandidat yang menjanjikan, tetapi memiliki kelarutan yang buruk, retrogradasi, dan ketidakstabilan dalam kondisi asam atau termal. Modifikasi kimia, khususnya hidroksipropilasi, telah digunakan untuk mengatasi kelemahan ini, sementara iota karagenan menawarkan manfaat tambahan sebagai agen pembentuk gel yang meningkatkan elastisitas dan stabilitas. Penelitian ini bertujuan untuk menentukan sifat fungsional dan termal terbaik pati termodifikasi hidroksipropil yang berasal dari singkong, sagu, jagung, dan kentang, serta mengkaji potensinya sebagai aplikasi kapsul lunak. Penelitian ini dilakukan dengan mengkarakterisasi berbagai modifikasi pati hidroksipropil dan menentukan karakterisasi pati termodifikasi sesuai persyaratan bahan baku cangkang kapsul lunak dengan pendekatan metode Zeleny. Parameter analisis yang dilakukan meliputi derajat substitusi, kapasitas menahan air, kapasitas menahan minyak, daya mengembang, kelarutan, sifat perekatan, *differential scanning calorimetry*, *scanning electron microscope*, *fourier transform infra red*, iota karagenan, dan *rapid visco analyzer*. Hasil menunjukkan bahwa hidroksipropil singkong memiliki kekuatan mekanis yang terpilih, dengan nilai DS 0,104, WHC 4,21%, OHC 5,43%, sweling 24,23%, dan kelarutan 21,54%. Hidroksipropilasi meningkatkan kelarutan, ketahanan termal, dan retensi air, dengan pati singkong menunjukkan tingkat substitusi tertinggi dan kemampuan pembentukan film yang baik. Lebih lanjut, penggabungan iota karagenan ke dalam pati singkong hidroksipropil menghasilkan film komposit homogen dengan kekuatan dan stabilitas mekanis yang lebih baik. Temuan ini menyoroti potensi sinergis pati hidroksipropil dan iota karagenan, menawarkan dasar ilmiah untuk pengembangan cangkang kapsul lunak berbasis taumbuhan yang berkelanjutan sebagai alternatif yang layak menggantikan gelatin.

Kata kunci: fungsional, gelatin, polisakarida, singkong, termal

INTRODUCTION

Soft capsules are widely used oral dosage forms designed to deliver active pharmaceutical ingredients in liquid or semisolid forms. These units are encapsulated within an elastic, hermetically sealed shell that ensures content protection and enhanced bioavailability (Hidayat *et al.*, 2024); Benza & Munyendo, 2011). Gelatin, derived from animal collagen, is the primary material used in soft capsule production because of its film-forming ability, rapid dissolution, and effective barrier properties against oxygen and light (Gullapalli, 2010). Despite its functional benefits, the use of gelatin has drawn increasing scrutiny due to safety concerns, such as the transmission of animal-borne diseases, along with dietary restrictions rooted in religious and ethical beliefs. Bovine-sourced gelatin is unacceptable to Hindu consumers, and porcine-derived gelatin is not permissible for Muslims (Febriana *et al*., 2021). Additionally, the growing trend toward plant-based and vegan lifestyles is accelerating the search for alternative encapsulating materials of nonanimal origin.

Starch is a natural polysaccharide composed primarily of amylose and amylopectin, and is derived from a variety of botanical sources, including cassava, sago, corn, and potato. It is widely recognized for its biodegradability, renewability, and cost effectiveness (Deshmukh *et al*., 2017). In pharmaceutical applications, starch offers benefits such as thermal resistance, processability, and potential film-forming ability (Dagadiye *et al*., 2012). However, native starch suffers from several limitations, including retrogradation, syneresis, poor cold-water solubility, and instability under acidic or high-temperature conditions (Palimbong *et al.,* 2023; Rosida, 2024). These drawbacks hinder their performance and shelf stability when used in capsule shell formulations.

To overcome these limitations, starch can be chemically modified to enhance its functionality. One promising approach is hydroxypropylation, which involves the

substitution of hydroxypropyl ether groups onto the hydroxyl groups of the starch backbone through a reaction with propylene oxide under alkaline conditions (Wang *et al.,* 2024). This modification improves water retention, reduces retrogradation, enhances solubility and thermal stability, and contributes to better freeze-thaw performance (Yuniar, 2006). These improvements make hydroxypropyl starch a viable alternative to gelatin in soft capsule formulation. However, the degree of modification and functional enhancement may vary depending on the starch source because of differences in the amylose-to-amylopectin ratio, granule size, and molecular architecture.

In addition to starch modification, incorporating supporting agents capable of forming stable gel networks is essential for developing plant-based soft capsules. Iota carrageenan, a sulfated polysaccharide extracted from red seaweed (Rhodophyceae), is a promising candidate. It forms elastic, transparent, and flexible gels in the presence of calcium ions (Hunt & Park, 2012), and its interaction with modified starches results in stable and homogeneous gel matrices (Tischer *et al.,* 2006; Haida *et al.,* 2024). In capsule formulations, iota carrageenan has been shown to improve the mechanical strength and film stability during storage (Hidayat *et al*., 2025; Fauzi *et al.,* 2021; Liu *et al.,* 2025). Therefore, the combination of hydroxypropyl starch and iota carrageenan represents a promising strategy for formulating high-performance plant-based soft capsule shells.

This study aimed to determine and compare the functional and thermal characteristics of hydroxypropyl-modified starches from cassava, sago, corn, and potato, and identify the most suitable starch source for soft capsule applications. In addition, this study explored the synergistic interaction between hydroxypropyl cassava starch and iota carrageenan in composite film formation, providing a scientific basis for the development of plant-based soft capsule shells as alternatives to gelatin.

MATERIALS AND METHODS

Hydroxypropyl Starch Modification Stages

Hydroxypropyl starch was prepared following the procedure described by Paramitasari *et al*. (2024a). A homogeneous mixture was prepared by combining 250 g of native starch (NS) with 395 g of a 10% (w/w) aqueous sodium sulfate solution, yielding a starch suspension with a final concentration of 40% (w/w) and a total weight of 645 g. The pH of the suspension was adjusted to 12 using 1 M NaOH solution. Subsequently, 18 mL of a 7% (w/w) aqueous propylene oxide solution was added, and the mixture was stirred in a closed flask for 30 min. Hydroxypropylation was performed by maintaining the suspension at 40°C under constant agitation at 150 rpm for 3 h in a water bath. The reaction was terminated by cooling the mixture and neutralizing it with 0.5 M HCl until the pH reached a stable range of 6.5–7. The resulting suspension was filtered and washed repeatedly with distilled water thereafter. The solid residue ("cake") was then dried in an oven at 50°C overnight until the moisture content was reduced to below 12% (w/w). The dried hydroxypropylated starch (HPS) was milled and sieved through an 80 mesh screen to obtain a uniform powder.

Degree of Substitution of Hydroxypropylation (DSH)

The degree of substitution was analyzed following the method described by Permatasari *et al*. (2024a). Samples (0.1 g) were dissolved in 25 mL of 1 N H_2SO_4 in a 100 mL volumetric flask. The flasks were heated in a boiling water bath until the starch was fully dissolved, and then cooled to room temperature. The volume was adjusted to 100 mL using distilled water. Further dilution was performed if necessary to ensure that the concentration of the hydroxypropyl group remained below 0.04 mg/mL. A 10 mL aliquot of the prepared solution was transferred to a test tube, followed by the addition of 8 mL of concentrated sulfuric acid (98% w/w). The mixture was then heated in boiling water for 3 min and immediately cooled in an ice bath. Subsequently, 0.6 mL of

http://dx.doi.org/10.17844/h6tbjw36

ninhydrin reagent (3% w/w ninhydrin in 5% w/w sodium bisulfite solution) was carefully added along the inner wall of each test tube. After thorough mixing, the reaction mixture was allowed to stand at room temperature for 100 min. The final volume was adjusted to 25 mL using concentrated sulfuric acid, and the contents were gently mixed by repeated inversion. The absorbance of the resulting solution was measured at 590 nm using a UV-Vis spectrophotometer (Merck, Spectroquant Pharo 300, Germany) after transferring the solution to a 1 cm cuvette and allowing it to stand for 5 min. Calibration was performed using standard solutions of propylene glycol at concentrations of 10, 20, 30, 40, and 50 µg/mL.

The hydroxypropyl group content (HP%) and the degree of substitution (DSH) were calculated using the following equations:

$$HP\% = \frac{(PG \times 0{,}7763 \times 10 \times F)}{W}$$

$$DSH = \frac{162 \times HP}{(5800-(58 \times HP\%))}$$

where
PG = the amount of propylene glycol (µg/mL) determined from the calibration curve
F = the dilution factor (if applicable)
0.7763 = the conversion factor for the hydroxypropyl group from propylene glycol
W = the sample weight (mg)

Water Holding Capacity (WHC)

The water-holding capacity was analyzed following the method described by Permatasari *et al.* (2024a). A 0.2 g sample of dry starch was dispersed in 4 mL of distilled water and allowed to stand at room temperature for 10 min. The mixture was then centrifuged using a Hitachi 05P-21 centrifuge (Hitachi, Tokyo, Japan) at 900×g for 25 min. After centrifugation, the supernatant was discarded, and the remaining sediment was weighed again. The water-holding capacity (WHC) was calculated using the following equation:

$$WHC = \frac{W1}{W0}$$

where
W1 = weight of the sediment (g)
W0 = initial weight of the dry sample (g)

Oil Holding Capacity (OHC)

The oil-holding capacity was analyzed following the method described by Permatasari *et al.* (2024a). A 0.5 g portion of the dry sample was dispersed in 6 mL vegetable oil. The mixture was vigorously mixed using a vortex mixer for 1 min and then allowed to stand at room temperature for 30 min. The mixture was then centrifuged using a Hitachi 05P-21 centrifuge (Japan) at 900×g for 40 min. The supernatant was carefully removed, and the remaining sediment was weighed after removal. The oil holding capacity was calculated using the following equation:

$$OHC = \frac{W1}{W0}$$

where
W1 = weight of the sediment (g)
W0 = initial weight of the dry sample (g)

Swelling Power and Solubility

Swelling power and solubility analyses were conducted following the method described by Indrianti *et al.* (2019). In this procedure, 0.2 g of starch was mixed with 10 mL distilled water in a centrifuge tube. The sample was equilibrated at room temperature for 5 min and vortexed. The mixture was subsequently heated in a water bath at 95°C for 30 min. After heating, the starch suspension was cooled to room temperature for 30 min and centrifuged at 3,000 rpm for 15 min. The swelling power was assessed based on the weight of the gel formed after heating. The solubility was determined by transferring the supernatant into a pre-weighed dish and drying it in an oven at 105°C for 3 h.

$$\text{Swelling power} = \frac{W1}{W0}$$

$$\text{Solubility} = \frac{Ws}{W0} \times 100$$

where
W1 = weight of the sediment (g)
W0 = initial weight of the sample (g)
Ws = the weight of the dried supernatant (g)
W0 = the initial weight of the dry starch (g)

Pasting Profiles

Pasting profiles were analyzed using a rapid viscometer analyzer (RVA) following the method described by Permatasari *et al.* (2024a). A sample of hydroxypropyl starch (30 g, dry basis) was thoroughly suspended in 470 g of distilled water and transferred to a Brabender mixing bowl. The analysis was conducted using a rapid viscometer (Germany). The instrument was programmed to follow a heating and cooling cycle: initial heating from 30 to 93°C, holding at 93°C for 20 min, cooling to 50°C, followed by another 20-min holding period at 50°C. The experimental protocol included maintaining a constant stirring speed of 75 rpm, heating rate of 1.5°C/min, and applied force of 700 cm-g (equivalent to 1,000 Brabender units). The pasting profile parameters recorded were pasting temperature, peak viscosity (peak 1), minimum viscosity (trough 1), breakdown, final viscosity, setback, and peak time.

Differential Scanning Calorimetry (DSC)

The analysis of the differential scanning calorimetry in this study refers to the method by Permatasari *et al.* (2024b) the thermal behavior of hydroxypropyl starch samples was evaluated using differential scanning calorimetry (DSC 3+, Mettler Toledo, Switzerland). Approximately 3 mg of each sample was accurately weighed and placed in a sealed aluminum pan. To ensure measurement accuracy, 9 µL of distilled water was added using a syringe to the sample. The pan was then hermetically sealed and allowed to equilibrate at room temperature for 30 min. The sample was subsequently heated from 30 to 130°C at a rate of 10°C/min. An empty, sealed pan was used as the reference. The instrument's software recorded the DSC thermogram and provided values for the onset temperature (To), peak temperature (Tp), conclusion temperature (Te), and gelatinization enthalpy (ΔH).

Moisture Content

Moisture content was determined using the gravimetric method following the procedure outlined by the AOAC (2005). An aluminum dish was pre-dried in an oven at 105°C for 30 min, cooled in a desiccator for 15 min, and weighed. A 1 g sample was placed in the dish and weighed again. The samples were dried in an oven at 105°C for 3 h until a constant weight was achieved. After cooling in a desiccator for 15 min, the samples were reweighed. The drying process was repeated for an additional hour, if needed, to reach a constant weight. The moisture content was calculated based on the weight difference between the fresh and dry samples, with weight loss considered as evaporated water. The percentage moisture content was calculated using the following equation:

$$\text{Moisture content (\%)} = \frac{(W2-W3)}{(W2-W1)} \times 100$$

where
W1 = weight of the empty dish (g)
W2 = weight of the dish+wet sample (g)
W3 = weight of the dish+dried sample (g)

Ash Content

The ash content was analyzed using the dry ashing method described by Verem *et al.* (2021). Approximately 1 g of a finely ground sample was placed in a previously weighed porcelain crucible. The sample was incinerated in a muffle furnace at 550°C for 6 h until complete ashing was achieved. After cooling in a desiccator for 15 min, the crucible containing the ash was weighed again. The ash content was calculated on a dry basis using the following equation:

$$\text{Ash content (\%)} = \frac{(W2-W3)}{(W1)} \times 100$$

where
W1 = weight of sample (g)
W2 = weight of crucible+ash (g)
W3 = weight of empty crucible (g)

Water Absorption Capacity

The water absorption capacity (WAC) was measured following the method adapted from Rauf and Sarbini (2015). A sample (1 g) was mixed with 10 mL of distilled water and vortexed for 2 min. The mixture was allowed to stand for 15 min and then centrifuged at 3,000 rpm for 25 min. The supernatant was decanted, and the remaining hydrated sample was weighed again. The amount of absorbed

http://dx.doi.org/10.17844/h6tbjw36

water was calculated as the difference between the weights of the hydrated and initial dry samples per 100 g.

Fourier Transform Infrared (FTIR)

The Fourier transform infrared (FTIR) analysis in this study was performed according to the method described by Paramitasari *et al.* (2024b). FTIR spectroscopy (Bruker Alpha II, Germany) was used to determine the chemical bonds present in the sample molecules by evaluating the absorption intensities of the functional group peaks. Approximately 0.1–0.5 g of powdered sample was prepared and placed onto the diamond crystal of the ATR-FTIR system using a stainless-steel laboratory spatula or a similar instrument. Pressure was applied to the material using a pressure clamp. The sample was analyzed using FTIR system software over a wavenumber range of 400–4,000 cm^{-1} to characterize the molecular absorption of the sample.

Scanning Electron Microscope (SEM)

The scanning electron microscopy analysis in this study was performed as described by Paramitasari *et al.* (2024a). The morphological structures of the samples were examined using an environmental scanning electron microscope (ESEM) (Quattro S, Thermo Fisher Scientific, USA). The samples were mounted on a standard aluminum pin stub with a diameter of 12.5 mm, which was placed on a designated holder and inserted into the instrument chamber. The samples were scanned at an accelerating voltage of 20 kV and a magnification of 1500×.

Data Analysis

The data were analyzed using ANOVA with a confidence level of 95%. If there was a significant difference between the treatments, a least significant difference (LSD) test was performed. Data processing was performed using SPSS software version 29.

RESULTS AND DISCUSSION

Degree of Substitution of Modified Starch

Starch modification, particularly through hydroxypropylation, is influenced by the degree of substitution (DS), which indicates the number of hydroxypropyl groups from propylene oxide replacing hydroxyl groups on glucose units in starch molecules. The process is affected by factors such as the reagent concentration, reaction time, temperature, and pH. In this study, the DS ranged from 0.08 to 0.1, which remains within the safe limit for food applications as set by the US FDA (maximum 0.2) (Zehra *et al*., 2021). The results of the degree of substitution of cassava, sago, potato, and corn hydroxypropyl starch are presented in Table 1.

Cassava starch exhibited a better degree of substitution than sago, potato, and corn starch. This is attributed to its high amylopectin content (87%), which promotes better accessibility of hydroxyl groups for hydroxypropylation (Dewi *et al.,* 2023). In contrast, other starches have higher amylose content (23–30%) (Hartesi *et al.,* 2020; Syukri & Dina, 2022; Fadhilah & Nurhalimah, 2024). Denser crystalline regions are formed owing to the linear structure of amylose, which limits the penetration of reagents (Ulbrich & Flöter, 2020; Choi & Kim, 2022). The branched

Table 1 Characteristics of the degree of substitution of hydroxypropyl starch

Hydroxypropyl starch type	Degree of substitution
Cassava	0.104 ± 0.01^{ns}
Sago	0.093 ± 0.001^{ns}
Potato	0.1 ± 0.01^{ns}
Corn	0.087 ± 0.01^{ns}

Values followed by the letters 'ns' indicate no significant difference based on analysis of variance (ANOVA) at a 95% confidence level.

structure of amylopectin increases the surface area and creates amorphous regions that are more susceptible to chemical modification (Choi & Kim, 2022).

A higher DS was associated with improved functional properties, such as greater swelling power, solubility, and water-holding capacity, consistent with the findings of Maulani *et al.* (2013) and Liu *et al.* (2022). These enhancements are linked to increased hydrophilicity and disrupted crystallinity, which facilitate water interaction and lower gelatinization temperature (Ulbrich *et al.,* 2020; Choi & Kim, 2022). These properties improve the processing performance by enhancing the viscosity and gelation behavior during film formation.

DS also influences the mechanical quality of the capsule films. Starches with higher DS, such as hydroxypropylated cassava starch, tend to form more amorphous matrices with better flexibility and elongation, as reported by Wongphan *et al.* (2024). These films are less prone to recrystallization, easier to handle, and disintegrate faster at body temperature, which is essential for drug delivery. Almeida *et al.* (2024) further confirmed that higher DS improves hydration and gel strength by disrupting crystallinity and enhancing amorphous structure, critical for forming stable, flexible soft capsule shells.

Water and Oil Holding Capacity

This section presents the functional properties of hydroxypropyl starch, focusing on its water-holding capacity (WHC) and oil-holding capacity (OHC). These parameters are essential for evaluating the suitability of soft capsule formulation. The results of the WHC and OHC tests on hydroxypropyl starch are presented in Table 2.

The water-holding capacity reflects the ability of starch to absorb and retain water, which plays a key role in determining the swelling behavior, gelatinization characteristics, and film-forming potential of starch-based materials. In this study, cassava starch demonstrated the highest WHC, which correlated with its high degree of substitution and amylopectin content. A greater number of hydroxypropyl groups introduces more hydrophilic sites, enhancing the affinity of starch for water (Marseno *et al.,* 2022). Additionally, the branched structure of amylopectin offers more available regions for hydrogen bonding with water molecules (Gerçekaslan, 2020; Shang *et al.,* 2021). These properties are beneficial for the development of soft capsule shells, as a high WHC supports the formation of flexible, hydrated films with better processability and rapid disintegration. Although amylose generally forms compact crystalline regions that restrict water absorption, it may still contribute to WHC through capillary water entrapment in granule pores, as noted by Kamsiati *et al.* (2017).

Oil holding capacity (OHC), on the other hand, represents the starch's ability to bind lipophilic substances. Sago starch exhibited the highest OHC, likely due to its rough and porous granule surface, which promotes physical oil entrapment, as previously reported. Moreover, the presence of surface-active compounds, such as residual proteins, may aid in oil binding via amphiphilic interactions (Rizkiana, 2015; Muchlisyiyah *et al.,* 2016). Amylose may also enhance OHC by forming inclusion complexes with lipids (Gerçekaslan *et al.,* 2020; Shang *et al.,* 2021). In the context of capsule formulations, a high OHC can be advantageous for encapsulating oil-based active compounds or improving

Table 2 Characteristics of the degree of substitution, WHC, and OHC of hydroxypropyl starch

Hydroxypropyl starch type	Water holding capacity	Oil holding capacity
Cassava	4.21±0.11^{b}	5.43±0.01^{b}
Sago	4.16±0.07^{b}	7.20±0.03^{b}
Potato	1.83±0.2^{a}	2.20±0.02^{a}
Corn	1.97±0.1^{a}	1.20±0.3^{a}

Values followed by the different letter within the same column are significantly different according to the Least Significant Difference (LSD) test at the 5% significance level.

the uniformity of oil-plasticizer blends within the film matrix. Thermal treatments, such as heat–moisture treatment (HMT) and pregelatinization, have been reported to improve OHC by disrupting crystalline regions and creating more oil-accessible sites (Tanak *et al.,* 2016; Shahira *et al.,* 2023). However, excessive heat may lead to retrogradation and reduced oil retention.

Notably, the variations in WHC and OHC across different starches highlight their divergent suitability for soft capsule formulations. Cassava starch, with its high WHC and DS, favors rapid hydration and film plasticity, which are critical for fast disintegration and ease of processing. In contrast, sago starch, despite having a high OHC, showed lower WHC and DS, suggesting that it may be more suitable as a supporting matrix in oil-based formulations rather than as a standalone capsule film former. This distinction underscores the importance of balancing hydrophilic and lipophilic capacities when selecting modified starches for targeted drug delivery systems.

Swelling Power and Solubility

Swelling power and solubility were assessed to evaluate the hydration behavior and dispersion characteristics of the hydroxypropyl starches. These parameters provide essential information on the effects of hydroxypropylation on starch–water interactions. The results of the swelling power and solubility tests of the hydroxypropyl starch are presented in Table 3.

Swelling power and solubility are key indicators of the ability of starch to absorb water and disperse upon heating. These parameters are primarily influenced by the molecular structure of starch, particularly the amylose-to-amylopectin ratio and degree of crystallinity. Starches with high amylopectin content and loosely packed granule structures tend to exhibit greater swelling capacity because the branched structure of amylopectin facilitates water penetration and interaction within the matrix (Winarno, 2002; Chávez-Murillo *et al.,* 2008).

In this study, hydroxypropylated sago starch exhibited the highest swelling power (34.25 g/g), surpassing potato, cassava, and corn starch. This can be attributed to the high amylopectin content and the effect of hydroxypropylation, which disrupts intra- and intermolecular hydrogen bonding, facilitating water uptake (Gunaratne & Corke, 2007; Polnaya & Marseno, 2008; Shen *et al.,* 2019; Chen *et al.,* 2021; Jia *et al*., 2023). Cassava starch also exhibited a notable increase in swelling after modification owing to its inherently large granule size and open structure that allows easier water infiltration (Maulani & Hidayat, 2016; Tang *et al.,* 2023). In contrast, corn starch exhibited the lowest swelling capacity, likely due to its high amylose content and smaller, more compact granules, which limit expansion.

The solubility of starch also increased after hydroxypropylation. Cassava starch exhibited the highest solubility, followed by sago, potato, and corn starch. This trend is consistent with the degree of substitution (DS), as a higher DS introduces more hydrophilic groups that enhance water affinity and promote the disruption of crystalline domains (Senanayake *et al.,* 2014; Metta & Sahoo, 2024). This modification process leads to a more amorphous and water-permeable structure, particularly in starches with a lower amylose content.

Table 3 Characteristics of swelling power and solubility of hydroxypropyl starch

Hydroxypropyl starch type	Swelling power (g/g)	Solubility (%)
Cassava	24.23±0.11^{b}	21.54±0.08^{d}
Sago	34.25±0.13^{d}	16.55±0.08^{c}
Potato	28.23±0.40^{c}	15.39±0.10^{b}
Corn	17.04±0.30^{a}	1.42±0.50^{a}

Values followed by the different letter within the same column are significantly different according to the Least Significant Difference (LSD) test at the 5% significance level.

Enhanced swelling and solubility play crucial roles in the functionality of starch-based soft capsule shells. High swelling power facilitates water absorption and expansion, contributing to faster disintegration and improved flexibility of the capsule film. Similarly, increased solubility allows for better dispersion of the polymer network and more uniform hydration, improving the film integrity and active ingredient release. As emphasized by Wongphan *et al.* (2024), film-forming materials with higher hydration capacities tend to exhibit better elongation and less recrystallization. Liu *et al.* (2022) further demonstrated that swelling and solubility directly impact the gel structure and drug-release kinetics of starch-based delivery systems. Therefore, these parameters serve as critical indicators for selecting modified starches for targeted pharmaceutical applications.

Pasting Properties

The pasting properties of hydroxypropylated starches provide essential insights into their thermal behavior and processing performance. These parameters are particularly important for predicting starch functionality in various food applications. The results of the pasting property tests are presented in Table 4.

The pasting properties of the hydroxypropylated starches varied significantly depending on the botanical source and their structural characteristics. Cassava and potato starches exhibited higher pasting temperatures than sago and corn starches, indicating stronger resistance to initial gelatinization. This is likely due to their denser granule structures and lower responsiveness to hydroxypropyl substitution, which delays the onset of swelling (Gunaratne & Corke, 2007; Indrianti *et al.,* 2019). In contrast, sago starch exhibited a lower pasting temperature, attributed to its higher amylose content and more open granule structure, which allowed for earlier water infiltration and thermal disruption.

Hydroxypropylated cassava starch exhibited the highest peak viscosity, indicating its superior water-binding capacity and swelling behavior. This can be attributed to its large granule size and high amylopectin content, which facilitate extensive hydration. However, the high breakdown value observed in cassava starch suggests reduced thermal stability during continuous heating, indicating its vulnerability to structural collapse. In contrast, sago, corn, and potato starches exhibited lower breakdown values, suggesting greater resistance to shear and thermal stress. This may be due to the stabilizing effect of hydroxypropyl groups, which interfere with molecular reassociation and strengthen granule integrity during heating (Muhandri *et al.,* 2016; Wulandari *et al.,* 2019; Paramitasari *et al.,* 2024a).

The final viscosity of cassava starch was also the highest, suggesting its strong ability to form viscous gels upon cooling, a beneficial trait for capsule shell film formation, where cohesive gel strength is required. However, its higher setback viscosity also indicates a tendency toward retrogradation, possibly driven by residual amylose interactions, even after modification (Kartikasari *et al.,* 2016; Jia *et al.,* 2023; Paramitasari *et al.,* 2024a). Sago, corn, and potato starches exhibited

Table 4 Characteristics of pasting properties of hydroxypropyl starch

Hydroxypropyl starch type	Starch pasting profile				
	Pasting temp	Peak 1	Breakdown	Final visc	Seatback
Cassava	72.70±0.11^{c}	5,510.00±0.42^{d}	3,322.00±0.06^{c}	2,932.00 ±0.02^{c}	744.00±0.04^{b}
Sago	65.73±0.18^{b}	1,496.67±0.06^{c}	670.67±0.34^{b}	1,187.00±0.42^{b}	670.67±0.06^{a}
Potato	76.70±0.05^{d}	127.00±0.14^{b}	107.00±0.13^{a}	32.00±0.03^{a}	1,200.00±0.03^{c}
Corn	50.40±0.23^{a}	107.96±0.04^{a}	4,396.00±0.16^{d}	8,873.00±0.14^{d}	2,473.00±0.00^{d}

Values followed by the different letter within the same column are significantly different according to the Least Significant Difference (LSD) test at the 5% significance level.

http://dx.doi.org/10.17844/h6tbjw36

lower setback viscosities, indicating better retrogradation inhibition, which is crucial for enhancing storage stability and preventing film brittleness.

Overall, hydroxypropylation improves starch hydrophilicity, enhances water penetration, and lowers pasting temperatures. It promotes higher peak viscosities through increased granule swelling and contributes to a greater film-forming potential, particularly in starches such as cassava. However, balancing the peak and breakdown viscosities is essential to ensure thermal stability and mechanical integrity during capsule processing.

Differential Scanning Calorimetry (DSC)

DSC analysis was employed to evaluate the thermal properties of starch, including the onset (To), peak (Tp), conclusion (Tc) temperatures, and gelatinization enthalpy (ΔH), which reflect the thermal stability and crystallinity of starch granules. The results of the DSC analysis of the hydroxypropyl starch are presented in Table 5.

DSC analysis revealed significant differences in the thermal behavior of hydroxypropylated starches, reflecting the underlying variations in molecular organization and crystallinity. Corn and cassava starches exhibited the highest gelatinization temperatures (Tp=74–75°C; Tc=78°C), indicating more crystalline, thermally stable granules that require a higher energy input for disruption. In contrast, potato starch exhibited the lowest gelatinization temperature (Tp=60°C; Tc=68°C), consistent with a more amorphous structure that facilitates earlier water absorption and swelling. These differences are influenced by the amylose-to-amylopectin ratio, granule size, and degree of crystalline packing (Senanayake *et al.,* 2014).

The gelatinization enthalpy (ΔH) values further supported this trend. Sago starch exhibited the highest ΔH (7 J/g), suggesting a stronger crystalline order, whereas cassava and potato starches had lower values (4 J/g), indicating reduced structural regularity (Fuentes *et al*., 2019). These findings are critical because the energy required to break down crystalline regions directly affects the thermal and hydration responsiveness of starch during processing.

Hydroxypropyl modification is known to disrupt the semi-crystalline structure of starch by introducing hydrophilic groups, thereby lowering both the gelatinization temperature and ΔH. However, the extent of this reduction depends on the native granule architecture of the starch. As reported by Liu *et al*. (2022) and Zehra *et al*. (2021), starches with higher degrees of substitution (DS) tend to exhibit more pronounced decreases in both thermal parameters. Nonetheless, native crystallinity, particularly in starches such as corn, can mitigate this effect and maintain relatively higher thermal thresholds post-modification.

From a pharmaceutical perspective, particularly in the design of soft capsule shells, starches with low gelatinization temperatures and enthalpies are advantageous. These properties enable faster hydration and disintegration at physiological temperatures, thereby enhancing the release kinetics of encapsulated compounds (Liu *et al.,* 2022; Wongphan *et al*., 2024). Thus, cassava and potato starches, owing to their lower thermal transition values post-modification,

Table 5 DSC characteristics of hydroxypropyl starch

Hydroxypropyl starch type	T0 (°C)	Tp (°C)	Tc (°C)	ΔH (J/g)
Cassava	60±0.2^{b}	75±0.3^{c}	78±0.3^{c}	4±0.1^{a}
Sago	58±0.4^{a}	65±0.4^{b}	75±0.3^{b}	7±0.3^{c}
Potato	55±0.6^{a}	60±0.2^{a}	68±0.5^{a}	4±0.4^{a}
Corn	65±0.1^{c}	74±0.3^{c}	78±0.1^{c}	5±0.2^{b}

Values followed by the different letter within the same column are significantly different according to the Least Significant Difference (LSD) test at the 5% significance level.

offer a favorable balance of processability and functionality for gelatin-free capsule applications (Zehra *et al.,* 2021).

Selection of Optimal Hydroxypropyl Starch Type

In this study, the optimal hydroxypropyl starch was selected using the multiple attribute decision-making (MADM) method proposed by Zeleny (1992). The selection was based on calculating the degree of closeness and distance measures (L_1, L_2, and $L\infty$) across multiple evaluation parameters. The treatment with the lowest combined distance was considered the most preferable.

Among the four starch types evaluated (cassava, sago, potato, and corn), hydroxypropyl cassava starch was identified as the optimal treatment, exhibiting the lowest total distance value and ranking highest in the decision matrix. This selection was supported by superior performance in key parameters such as degree of substitution, water holding capacity (WHC), solubility, peak 1 in pasting properties, and enthalpy change (ΔH) in DSC analysis. While sago starch showed favorable swelling power and setback values, and potato starch excelled in breakdown, gelatinization temperatures (T_0, T_p, T*c*), and ΔH, cassava starch was determined to be the most balanced and effective overall. Therefore, it was selected for the subsequent soft capsule film formulation. The results of the selection process using the Zeleny-based multiple attribute decision-making (MADM) method are presented in Table 6.

Scanning Electron Microscope (SEM)

SEM analysis of hydroxypropyl cassava starch (Figure 1) revealed rounded to oval-shaped granules with a uniform particle size distribution and smooth surfaces, free from visible cracks. These features suggest that the hydroxypropylation process was homogeneous and conducted under mild reaction conditions, effectively introducing substitution without compromising the structural integrity of the starch granules.

In contrast, the morphology of hydroxypropyl sago starch, as reported by Paramitasari *et al.* (2024a), showed greater

Table 6 Results of optimal treatment selection using the MADM method (Zeleny)

Hydroxypropyl starch type	L1	L2	L∞	Total distance density	Ranking
Cassava	0.25	0.01	0.06	0.33	1
Sago	0.32	0.01	0.06	0.40	3
Potato	0.30	0.01	0.07	0.39	2
Corn	0.39	0.02	0.07	0.48	4

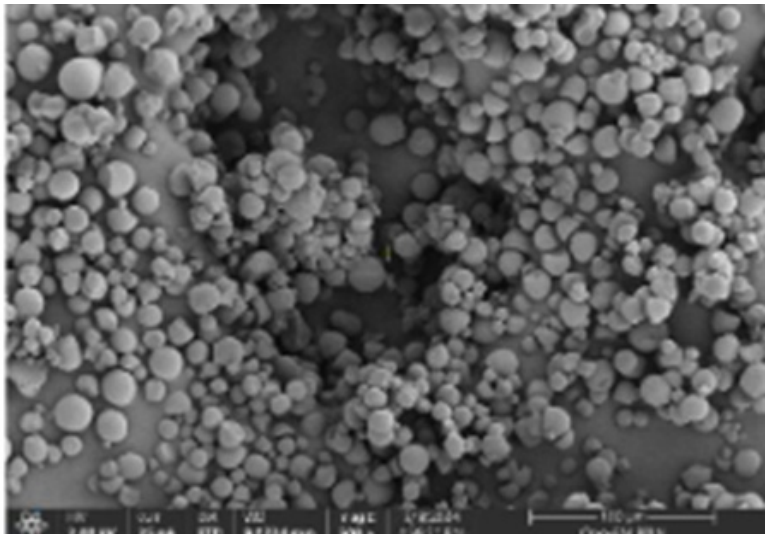

Figure 1 Morphology of hydroxypropyl cassava starch (2,000×)

http://dx.doi.org/10.17844/h6tbjw36

irregularity. The granules exhibited uneven shapes with surface indentations and deformations. This surface roughness was attributed to the processing stresses and heat exposure during film formation, especially when starch came into direct contact with a heated drum roller. The fibrous surface texture observed led to increased stiffness and enhanced moisture uptake during the storage period. Additionally, morphological variations in sago starch may result from its sensitivity to alkaline reactions and inconsistent degrees of substitution.

Similarly, the hydroxypropyl potato and corn starch films exhibited distinct morphological differences. According to Ock *et al.* (2020), these films contain small crystalline structures (2–15 μm) and exhibit non-uniform surfaces. The higher amylose content of potato (20.5%) and corn (22.8%) promotes crystalline aggregation during drying due to stronger intermolecular interactions among amylose chains. This crystalline structure contributes to increased water absorption and rapid structural degradation over time.

Although hydroxypropyl modification reduces retrogradation by enhancing hydration and weakening internal bonding, the final film morphology remains highly dependent on the amylose-to-amylopectin ratio (A/A ratio). Starches with lower amylose content, such as cassava, tend to form smoother and more stable films because amylopectin is more amorphous and less prone to crystallization.

In conclusion, based on the SEM observations and supporting literature, hydroxypropyl cassava starch demonstrates superior morphological properties for soft capsule film applications. Its intact granules, smooth surface, and consistent particle distribution offer clear advantages over sago, corn, and potato starch, which tend to exhibit rougher and more heterogeneous surface structures.

Fourier Transform Infra Red (FTIR)

FTIR spectra comparison between native cassava starch and hydroxypropylated cassava starch confirmed the success of the chemical modification via etherification. A broad absorption band at approximately 3,200–3,600 cm^{-1}, corresponding to O–H stretching, was observed in both samples, indicating hydrogen bonding within the starch structure. After modification, the sharper –OH band suggested changes in hydrogen bonding, consistent with the findings of Mudiaga-Ojemu *et al.* (2023).

The increased intensity of the C–H stretching band at 2,933.6 cm^{-1} indicates the presence of methyl and methylene groups introduced through hydroxypropyl chains. Although previous studies (Aminian *et al.* (2013); Rutkaitė *et al.* (2016) reported the appearance of a new asymmetric CH stretch near 2,976 cm^{-1} after hydroxypropylation, this peak was not prominently detected in the present sample.

The presence of a C–H bending band at 1,340.7 cm^{-1} corresponds to typical hydrocarbon-related deformations, supporting successful substitution. A notable increase in intensity at 1,150.2 cm^{-1} was attributed to C–O–C ether stretching, indicating the formation of new ether bonds, which is a key marker of hydroxypropylation. This observation is consistent with prior findings (Sukhija *et al.,* 2016), which reported enhanced C–O–C signals in the 1,076–1,068 cm^{-1} range after the modification.

In addition, the stability of the glycosidic C–O–C absorption at approximately 853–856 cm^{-1} suggests that the core anhydroglucose structure remained intact. Overall, the observed shifts and intensity changes in the –OH, C–H, and C–O–C regions provide strong evidence of successful hydroxypropylation. These chemical changes are expected to enhance the solubility, dispersion stability, and film-forming ability of starch, which are beneficial for food and pharmaceutical applications. The FTIR spectrum of the hydroxypropylated cassava starch is shown in Figure 2.

Characteristics and Morphology of Iota Carrageenan

The moisture content analysis of iota carrageenan showed a low value, indicating good storage stability. Low moisture content is crucial as it inhibits chemical degradation and microbial growth, thereby supporting

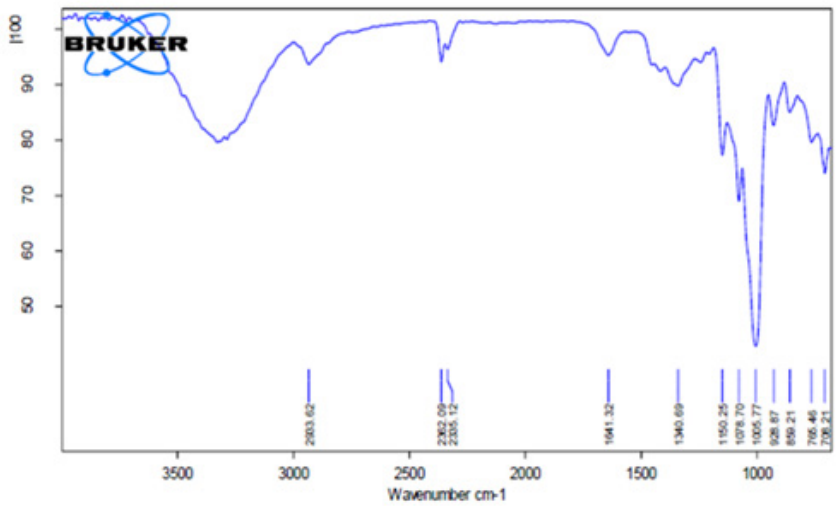

Figure 2 FTIR spectrum of hydroxypropylated cassava starch

a longer shelf life, especially in food and pharmaceutical products (Imeson *et al.,* 2009). Similarly, Campo *et al.* (2009) reported that iota carrageenan dried using conventional methods had a moisture content of 10.2±0.4%, which is comparable to the results of this study.

The ash content in iota carrageenan serves as an indicator of its purity and reflects its marine origin. High ash content corresponds to a high mineral content in iota carrageenan. The mineral composition of seaweed is strongly influenced by seasonal variations, geographical origin, and purification methods (Sari *et al.,* 2021). According to FAO standards, the acceptable ash content for commercial-grade carrageenan ranges from 15% to 40% (Praseptiangga *et al.,* 2018). In this study, the ash content of iota carrageenan was found to be 34.86±0.7%, which is within the FAO-recommended range. Wang *et al.* (2017) reported that after a purification process, the ash content of iota carrageenan could be reduced to 14.2±0.6%, indicating a high level of purity suitable for pharmaceutical applications, such as in soft capsule production.

A high swelling power indicates the ability of iota carrageenan to significantly swell and absorb water in solution. This property demonstrates that iota carrageenan is highly hydrophilic and has a strong potential to form stable and elastic gel matrices, making it highly suitable as a texture-forming agent in soft capsule formulations (Necas & Bartosikova, 2013). In the present study, the swelling power was higher, reaching 47.33±0.03 g, compared to the findings of Sriwahyuni *et al.* (2020), where iota carrageenan exhibited a swelling power of 12.1 g/g at 60°C. This suggests that the iota carrageenan used in this study had a superior water absorption capacity, possibly due to a looser gel network and higher porosity.

The low solubility of iota carrageenan indicates that it does not fully dissolve in water, but rather forms a gel or colloidal system. This limited solubility is influenced by the ionic interactions within the helical structure of carrageenan polysaccharides (Ferdiansyah *et al.,* 2023). According to Wang *et al.* (2017), iota carrageenan exhibits the highest solubility at neutral to slightly alkaline pH (pH 6–8), whereas its solubility decreases under extreme pH conditions owing to the degradation of the polymer chain. Additionally, solubility increases with temperature; iota carrageenan is insoluble in cold water but rapidly dissolves in hot water. Interactions with Ca^{2+} ions can further enhance solubility and promote gel formation.

The water absorption capacity of the capsules in this study was relatively high, indicating that iota carrageenan can effectively absorb and retain moisture. This property is crucial for formulation design to maintain the stability of the texture and moisture in the final product (Yuan *et al.,* 2020). According to Sriwahyuni *et al.* (2020), capsules formulated with iota carrageenan can absorb a significant

http://dx.doi.org/10.17844/h6tbjw36

amount of water, approximately 3.9–5.6 g of water per g of dry capsule, depending on the composition and concentration of plasticizers used (such as glycerol or sorbitol). Capsules with higher iota carrageenan content showed a linear increase in water absorption capacity due to the formation of a more porous matrix structure that allows easier water penetration. The results of the iota carrageenan analysis are presented in Table 7.

SEM analysis of iota carrageenan (Figure 3) revealed that the surface of the iota carrageenan particles exhibited a non-homogeneous morphology with a rough, cracked, and porous structure. Numerous structures resembling irregular crystal fragments were observed, indicating a fractured or brittle structure of the material. Some particles appeared agglomerated owing to adhesion, possibly caused by intermolecular interactions or the effects of the drying process. This suggests the presence of physical interactions between the components within the material system. The particle sizes varied significantly, ranging from very small to relatively large, with generally irregular shapes. However, some particles exhibited sharp edges resembling crystals, indicating the presence of partially crystalline structures.

This phenomenon is consistent with the findings of Campo *et al.* (2009), who reported that iota carrageenan possesses a rough, non-homogeneous, and porous surface structure. This surface morphology indicates the presence of aggregates and irregular microvoids, which are common characteristics of marine polysaccharides, such as iota carrageenan, after drying. The porous structure is attributed to the drying method (e.g., spray or freeze-drying), which creates voids within the material matrix. Iota carrageenan is known for its rapid disintegration and relatively low-to-moderate mechanical strength.

In contrast, the morphology of kappa carragenan, as reported by Dini *et al.* (2020), showed the smooth, dense, and homogeneous

Table 7 Characteristics of iota carrageenan

Analysis	Results
Moisture (%)	11.34±0.40
Ash (%)	34.86±0.70
Swelling power (g)	47.33±0.03
Solubility (%)	1.42 ±0.01
Water absorption capacity (WAC) (%)	80.00±0.20

Figure 3 Morphology of iota carrangenan (2,000×)

surface observed in the SEM images indicates that the gel structure formed by kappa carrageenan is compact and well-organized. Owing to its high mechanical strength, kappa carrageenan is more suitable for hard capsule formulations. In conclusion, based on the SEM observations and supporting literature, iota carrageenan demonstrates favorable properties for soft capsule film applications. It exhibits rapid disintegration and relatively low-to-moderate mechanical strength compared to kappa carrageenan, which tends to have higher mechanical strength, making it more appropriate for hard capsules.

Synergistic Role of Hydroxypropyl Starch and Iota Carrageenan in Soft Capsule Shell Formation

The soft capsule film solution was prepared by blending hydroxypropyl starch (HPS) with iota carrageenan, using sorbitol and glycerol as plasticizers. The formulation was adapted from a previous study (Paramitasari *et al.*, 2024a), with an upper limit composition consisting of 30% HPS, 2% iota carrageenan, 15% sorbitol, and 12% glycerol. The preparation process involved mixing iota carrageenan, sorbitol, glycerol, and water, followed by heating at 60°C. HPS was then gradually incorporated under continuous stirring at 3000 rpm until the temperature reached 95°C, maintained for 90 min, subjected to vacuum treatment for 3 min without agitation, and finally cast into 1 mm thick films.

The pasting and viscosity behaviors were evaluated using a rapid viscometer analyzer (RVA). The softgel 75 formulation exhibited a pasting temperature of 75°C with a final viscosity of 1,868 cP, indicating the development of a compact and stable gel network with limited retrogradation. Conversely, the softgel 95 formulation demonstrated a higher pasting temperature of 85°C but a lower final viscosity of 1,254 cP, reflecting weaker gel network formation and greater susceptibility to retrogradation.

These findings highlight that starch–iota carrageenan films require higher processing temperatures than gelatin-based systems, which can be attributed to their distinct thermal properties. Starch gelatinization is an endothermic transition associated with the loss of crystallinity in starch granules. A higher gelatinization enthalpy indicates greater crystallinity and more ordered double helices stabilized by long amylopectin chains, thereby requiring greater thermal energy to disrupt the crystalline structure (Shi *et al*., 2022*)*. During thermal processing, starch granules absorb water, swell, and rupture, resulting in gelatinization within a temperature range of 60–80°C, typically spanning 8–15°C rather than occurring at a discrete point (Briffaz *et al.,* 2012).

In contrast, gelatin exhibits lower melting and gelation temperatures owing to the susceptibility of its triple helical structure to thermal disruption. Upon heating, gelatin helices unravel into random coils, and during cooling, they reassociate into elastic gels through polypeptide networks that lack the crystalline order observed in starch (Lin *et al.* 2023). Gelatin dissolves in hot water above 40°C and forms gels upon cooling at 30–40°C (Rowe *et al.,* 2009). These fundamental differences underscore the higher thermal input required for starch–iota carrageenan films compared with that for gelatin systems.

Beyond thermal characteristics, the molecular interactions between HPS and iota carrageenan are critical to film performance. These interactions are primarily mediated by hydrogen bonding between the hydroxyl groups of starch and the sulfate groups of carrageenan, supplemented by partial exclusion and surface entrapment mechanisms (Lascombes *et al.,* 2017). These intermolecular associations enhance the compatibility of the composite matrix, yielding improved tensile strength, flexibility, and barrier properties compared to pure carrageenan films (Abdillah & Charles, 2021). Moreover, treatments such as high-pressure processing or starch modification further facilitate stronger bonding with carrageenan, leading to films with enhanced hydrophobicity and mechanical integrity (Shahbazi *et al.,* 2018). Collectively, these synergistic interactions explain the superior stability of starch–iota carrageenan films and substantiate their potential as robust plant-based alternatives to

conventional gelatin soft capsule shell.

Native starch is prone to retrogradation due to amylose and amylopectin reassociation, whereas hydroxypropyl modification reduces this tendency by improving paste stability. In contrast, iota carrageenan gels are flexible but mechanically fragile when used alone. Their combination produces a synergistic effect: iota carrageenan stabilizes the starch matrix and reduces retrogradation, whereas hydroxypropyl starch enhances the mechanical integrity of carrageenan films. This is consistent with the findings of Aliyatunnaim *et al.* (2022), who reported that starch–carrageenan blends exhibit superior physical properties compared to starch without hydrocolloids. This synergistic effect was reflected in the RVA results, where the softgel 75 formulation, with a lower pasting temperature and higher final viscosity, produced a more stable and compact network suitable for film formation and capsule sealing. In contrast, softgel 95, which had a higher gelatinization temperature but a lower final viscosity, formed weaker and more fragile gels, making it less suitable for soft capsule applications.

CONCLUSION

Modification of starch through hydroxypropylation has been shown to enhance the physical and functional properties of various native starches for application in plant-based soft capsules. Among the samples analyzed, hydroxypropylated cassava starch exhibited the most favorable characteristics, including the highest degree of substitution, superior water-binding capacity and solubility, and high peak viscosity, indicating excellent swelling and gel-forming abilities. Although sago starch demonstrated the highest swelling power and oil-holding capacity (OHC), its degree of substitution was comparatively lower. Overall, hydroxypropylated cassava starch is considered the most promising candidate for plant-based soft capsule formulations because of its balanced properties of stability, swelling ability, solubility, and ease of gelatinization.

ACKNOWLEDGEMENT

The authors are very grateful to the National Research and Innovation Agency (BRIN) for funding the housing program at the BRIN Research Agricultural and Food Organization.

REFERENCE

Abdillah, A. A., & Charles, A. L. (2021). Characterization of a natural biodegradable edible film obtained from arrowroot starch and iota-carrageenan and application in food packaging. *International Journal of Biological Macromolecules*, 191, 618–626. https://doi.org/10.1016/j.ijbiomac.2021.09.141

Aliyatunnaim, N. A., Septina, N. D. L., Ginting, D. F. A., & Maharani, F. (2022). Karakteristik pati biji nangka (*Artocarpus heterophyllus*) yang berpotensi sebagai pengganti gelatin pada pembuatan cangkang kapsul lunak. *Prosiding Seminar Nasional Sains dan Teknologi Fakultas Teknik*, 12(1), 9–12. https://doi.org/10.36499/psnst.v12i1.7184

Almeida, R. L. J., Monteiro, S. S., Santos, N. C., Rios, N. S., & Santos, E. S. D. (2024). Starch modification and its application in pickering emulsion stabilization: a review. *Journal of Food Measurement and Characterization*, 18(6), 4984–5003. https://doi.org/10.1007/s11694-024-02550-6

Aminian, M., Nafchi, A. M., Bolandi, M., & Alias, A. K. (2013). Preparation and characterization of high degree substituted sago (*Metroxylon sagu*) starch with propylene oxide. *Starch-Stärke* 65(7–8), 686–693. https://doi.org/10.1002/star.201200137

[AOAC] Association of Official Analytical Chemist. (2005). Official Methods of Analysis, 18th ed. Maryland, USA. AOAC International.

Benza, H. I., & Munyendo, W. L. (2011). A review of progress and challenges in soft gelatin capsules formulations for oral administration. *International Journal of Pharmaceutical Sciences Review and Research*, 10(1), 20–24.

Briffaz, A., Mestres, C., Escoute, J., Lartaud, M., & Dornier, M. (2012). Starch gelatinization distribution and peripheral cell disruption in cooking rice

grains monitored by microscopy. *Journal of Cereal Science*. 56(3), 699–705. https://doi.org/10.1016/j.jcs.2012.09.001

Campo, V. L., Kawano, D. F., Silva, D. B. J., & Carvalho, I. (2009). Carrageenans: biological properties, chemical modifications and structural analysis-a review. *Carbohydrate Polymers*, 77(2), 167–180. https://doi.org/10.1016/j.carbpol.2009.01.020

Chávez-Murillo, C. E., Wang Y. J., & Bello-Pérez, L. A. (2008). Morphological, physicochemical and structural characteristics of oxidized barley and corn starches. *Starch-Stärke*, 60(11), 634–645. https://doi.org/10.1002/star.200800016

Chen, P., Zhang, Y., Qiao, Q., Tao, X., Liu, P., & Xie, F. (2021). Comparison of the structure and properties of hydroxypropylated acid-hydrolysed maize starches with different amylose/amylopectin contents. *Food Hydrocolloids*, 110, 106134. https://doi.org/10.1016/j.foodhyd.2020.106134

Choi, H. W., Kim, H. S. (2022). Hydroxypropylation and acetylation of rice starch: effects of starch protein content. *Food Science and Biotechnology*, 31(9), 1169–1177. https://doi.org/10.1007/s10068-022-01106-y.

Dagadiye, R. B., Kajale, A. D., Mahajan, V. K., & Joshi, M. H. (2012). Advancement in manufacturing of non-gelatin capsule shell-a review. *International Journal of Advances in Pharmaceutical Research*, 3(10), 1178–1187.

Deshmukh, K., Ahamed, M. B., Deshmukh, R. R., Pasha, S. K., Bhagat, P. R., & Chidambaram, K. (2017). Biopolymer composites with high dielectric performance: interface engineering. *Biopolymer Composites in Electronics*, 27–128. https://doi.org/10.1016/B978-0-12-809261-3.00003-6

Dewi, S. R., Widyasanti, A., & Putri, S. H. (2023). Pengaruh konsentrasi pati singkong terhadap karakteristik *edible film* berbahan pati singkong dengan penambahan ekstrak daun belimbing wuluh. *Jurnal Keteknikan Pertanian Tropis dan Biosistem*, 11(2), 158–167. https://doi.org/10.21776/ub.jkptb.2023.011.02.05

Dini, A. F., Rochima, E., & Mulyani, O. (2020). Evaluation of hard capsule application from seaweed: gum Arabic-κ-carrageenan biocomposite films. *IOP Conference Series: Earth and Environmental Science*. 441.

Fadhilah, S., Nurhalimah, S. (2024). Analisis kimia pati sagu dari berbagai pati lokal. *Karimah Tauhid*, 3(10), 11726–11738. https://doi.org/10.30997/karimahtauhid.v3i10.15531

Fauzi, M. A. R. D., Pudjiastuti, P., Wibowo, A. C., & Hendradi, E. (2021). Preparation, properties and potential of carrageenan-based hard capsules for replacing gelatine: a review. *Polymers*, 13(16), 1-20. https://doi.org/10.3390/polym13162666

Febriana, L. G., Stannia, N. A. S., Fitriani, A. N., & Putriana, N. A. (2021). Potensi gelatin dari tulang ikan sebagai alternatif cangkang kapsul berbahan halal: karakteristik dan pra formulasi. *Majalah Farmasetika*, 6(3), 223–233. https://doi.org/10.24198/mfarmasetika.v6i3.33183

Ferdiansyah, R., Abdassah, M., Zainuddin, A., Rachmaniar, R., & Chaerunisaa, A. Y. (2023). Effects of alkaline solvent type and pH on solid physical properties of carrageenan from *Eucheuma cottonii*. *Gels*, 9(5), 1-14. https://doi.org/10.3390/gels9050397

Fuentes, C., Kang., I, Lee, J., Song, D., Sjoo, M., Choi, J., Lee, S., & Nilsson, L. (2019). Fractionation and characterization of starch granules using field-flow fractionation (FFF) and differential scanning calorimetry (DSC). *Analytical and Bioanalytical Chemistry*, 411(8), 3665–3674. https://doi.org/10.1007/s00216-019-01852-9

Gerçekaslan. (2020). Hydration level significantly impacts the freezable- and unfreezable-water contents of native and modified starches. *Food Science and Technology*, 41(2), 426–431. https://doi.org/10.1590/fst.04520

Gullapalli, R. P. (2020). Soft gelatin capsules (softgels). *Journal of Pharmaceutical Sciences*. 99(10), 4107–4148. https://doi.

org/10.1002/jps.22151.

Gunaratne, A., & Corke, H. (2007). Functional properties of hydroxypropylated, cross-linked, and hydroxypropylated cross-linked tuber and root starches. *Cereal Chemistry*, 84(1), 30–37. https://doi.org/10.1094/CCHEM-84-1-0030

Haida, N. R., Syahariza, Z. A., Fazilah, A., & Norsila, A. W. (2024). Preparation and characterization of sago/iota-carrageenan microgels as food thickener in texture modified food. *Food Research*, 8(6), 114–126. https://doi.org/10.26656/fr.2017.8(6).560

Hartesi, B., Andriani, L., Anggresani, L., Whinata, M. B., & Haflin, H. (2020). Modifikasi pati kentang secara pregelatinasi dengan perbandingan pati dan air (1:1,25). *Riset Informasi Kesehatan*, 9(2), 177–187. https://doi.org/10.30644/rik.v9i2.431

Hidayat, T., Syamsu, K., Sunarti, T. C., Nurilmala, M., & Manalu, L. P. (2024). Preliminary study effect of by-products of biodiesel glycerol in increasing the elasticity of film sheets for seaweed derivative-based soft capsules. In IOP Conference Series: Earth and Environmental Science (Vol. 1354, No. 1, p. 012019). IOP Publishing.

Hidayat, T., Syamsu, K., Sunarti T. C., Nurilmala, M., Manalu, L. P. (2025). Potential of tropical seaweed carageenan in applications of soft capsule as a replacement for gelatin: a review. *Science and Technology Indonesia*, 10(4), 1288-1300. https://doi.org/10.26554/sti.2025.10.4.1288-1300

Hunt, A., & Park, J. W. (2012). Alaska pollock fish protein gels as affected by refined carrageenan and various salts. *Journal of Food Quality*, 36, 51–58. https://doi.org/10.1111/jfq.12010

Imeson, A. (2009). Food Stabilisers, thickeners and gelling Agents. Wiley-Blackwell.

Indrianti, N., Afifah, N., & Sholichah, E. (2019). Pembuatan tepung komposit dari pati ganyong/garut dan tepung labu kuning sebagai bahan baku *flat noodle*. *Biopropal Industri*, 10(1), 49–63. https://doi.org/10.4172/2157-7110.1000713

Jia, R., Cui, J., Gao, L., Qin, Y., Ji, N., Dai, L., Wang, Y., Xiong, L., Shi, R., & Sun, Q. (2023). A review of starch swelling behavior: its mechanism, determination methods, influencing factors, and influence on food quality. *Carbohydrate Polymers*, 321, 121260. https://doi.org/10.1016/j.carbpol.2023.121260

Kamsiati, E., Herawati, H., & Purwani, E. Y. (2017). Potensi pengembangan plastik biodegradable berbasis pati sagu dan ubi kayu di Indonesia. *Jurnal Litbang Pertanian*, 36(2), 67–76. https://doi.org/10.21082/jp3.v36n2.2017.p67-76

Kartikasari, S. H., Sari, P., & Subagio. (2016). Karakterisasi sifat kimia, profil amilografi (RVA) dan morfologi granula (SEM) pati singkong termodifikasi secara biologi. *Jurnal Agroteknologi*, 10(1), 12–24.

Lascombes, C., Agoda-Tandjawa, G., Boulenguer, P., Garnec, C. L., Gilles, M., Mauduit, S., Baret, P., Langendorff, P. (2017). Starch-carrageenan interactions in aqueous media: role of each polysaccharide chemical and macromolecular characteristics. *Food Hydrocolloids*, 66, 176–189. https://doi.org/10.1016/j.foodhyd.2016.11.025

Lin, H. T. V., Tsai, J. S., Liao, H. H., & Sung, W. C. (2023). The effect of hydrocolloids on penetration tests and syneresis of binary gum gels and modified corn starch–gum gels. *Gels*, 9(8), 1-13. https://doi.org/10.3390/gels9080605

Liu, X. L., Chen, Y. F., Yang, J. J., Li, S. J., Xie, H. L., & Ma, T. L. (2022). Effect of molar substitution on the properties of γ-hydroxypropyl starch. *Molecules,* 27(7), 1-10. https://doi.org/10.3390/molecules27072119

Liu, Z., He, C., Yang, Z., Zhao, Q., Dong, Y., Ye, J., Zheng, B., Kankala, R. K., Zhang, X., & Xiao, M. (2025). κ/ι-Carrageenan blends in plant capsules: achieving harmony between mechanical and disintegration properties. *Marine Drugs*, 23(7), 1-24. https://doi.org/10.3390/md23070284

Marseno, D. W., *et al*. (2022). Teknologi modifikasi. UGM Press.

Maulani, R. R., Fardiaz, D., Kusnandar, F., & Sunarti, T. C. (2013). Characterization

of chemical and physical properties of hydroxypropylated and cross-linked arrowroot (*Marantha arundinacea*) starch. *Journal of Engineering and Technological Sciences*, 45(3), 207–221. https://doi.org/10.5614/j.eng.technol.sci.2013.45.3.1

Maulani, R. R., & Hidayat, A. (2016). Characterization of the functional properties of hydroxypropylated and cross-linked arrowroot starch in various acidic pH mediums. *International Journal of Technology*, 7(1), 176–184. https://doi.org/10.14716/ijtech.v7i1.1226

Metta, S., & Sahoo, S. K. (2024). Characterization and safety assessment of hydroxypropyl musa paradisiaca starch for pharmaceutical applications. *Journal of Applied Pharmaceutical Science*, 14(7), 111–123. https://doi.org/10.7324/JAPS.2024.174568

Muchlisyiyah, J., Prasmita, H. S., Estiasih, T., Laeliocattleya, R. A., & Palupi, R. (2016). Sifat fungsional tepung ketan merah pragelatinisasi. *Jurnal Teknologi Pertanian*, 17(3), 195–202. https://doi.org/10.21776/ub.jtp.2016.017.03.5

Mudiaga-Ojemu, B. O., Eraga, S. O., & Iwuagwu, M. A. (2023). Characterization of hydroxypropylated cassava and potato starches: functional and physicotechnical properties. *Journal of Science and Practice of Pharmacy*, 10(1), 489–498. https://doi.org/10.47227/jsppharm.v10i1.1

Muhandri, T., Nularif, P. S., & Hunaefi, D. (2016). Karakteristik amilografi tepung jagung termodifikasi dengan fermentasi spontan dan perendaman dalam larutan kalsium hidroksida. *Jurnal Mutu Pangan*, 3(2), 124–129. https://journal.ipb.ac.id/jmpi/article/view/26244

Necas, J., & Bartosikova, L. (2017). Carrageenan: a review. *Veteterinarni Medicina*, 58(4), 187–205.

Ock, Y. O., Lim, W. S., Park, G. D., Lee, M. H., & Park, J. H. (2020). Physical and mechanical properties of plant-derived soft-shell capsules formulated with hydroxypropyl starches from different botanical sources. *Polymer Testing*, 91, 106871. https://doi.org/10.1016/j.polymertesting.2020.106871

Palimbong, S., Nugroho, P., & Pratiwi, A. A. (2023). Modifikasi pati Suweg (*Amorphophallus paeoniifolius* var. companulatus) dengan menggunakan sodium tripolifosfat (STPP). *Jurnal Pengolahan Pangan*, 8(1), 47–54. https://doi.org/10.31970/pangan.v8i1.104

Paramitasari, D., Musa, M., Putra, O. N., Elisa, S., Suparman, S., & Pramana, Y. S. (2024a). Enhancing the functional characteristics of sago starch through dual chemical modification by hydroxypropylation and succinylation," Univ. Scientiarum 29(2), 146–167.

Paramitasari, D., Musa, M., Putra, O. N., Suparman, S., Pramana, Y. S., Elisa, S., & Supriyanti, A. (2024b). Hydroxypropylation for functional enhancement of sago starch: the effects of low propylene oxide concentration using response surface methodology. *Journal of Agriculture and Food Research*, 15, 1-14. https://doi.org/10.1016/j.jafr.2023.100933

Polnaya, F. J., & Marseno, D. W. (2008). Characteristics of hydroxypropylated and acetylated sago starches. *Sago Palm*, 16(2), 85–94. https://doi.org/10.1016/j.jafr.2023.100933

Praseptiangga, D., Giovani, S., Muhammad, D. R. A., & Manuhara, G. J. (2018). Development of edible film from semi-refined iota carrageenan for sustainable food packaging. *ARPN Journal of Engineering and Applied Sciences*, 13, 8907–8918.

Rauf, R., & Sarbini, D. (2015). Daya serap air sebagai acuan untuk menentukan volume air dalam pembuatan adonan roti dari campuran tepung terigu dan tepung singkong. *Agritech*, 35(3), 324–330. https://doi.org/10.22146/agritech.9344

Rizkiana, W. (2015). Produksi pati tapioka nanokristalin terasetilasi. [Skripsi]. Institut Pertanian Bogor.

Rosida, D. F. (2024). Buku ajar modifikasi pati dari umbi-umbian lokal dan aplikasinya untuk produk pangan. CV. Putra Media

Nusantara.

Rowe, R. C., Sheskey, P. J., & Quinn, M. E. (2009). Handbook of pharmaceutical excipients, 6th ed. Pharmaceutical Press.

Rutkaitė, R., Baranauskienė, R., Pečiulytė, L., Pukalskienė, M., & Venskutonis, P. R. (2016). Preparation and properties of propylene oxide and octenylsuccinic anhydride modified potato starches. *Journal of Food Science and Technology*, 53(12), 4187–4196. https://doi.org/10.1007/s13197-016-2403-9

Sandhu, K. S., Sharma, L., Kaur, M., & Kaur, R. (2020). Physical, structural and thermal properties of composite edible films prepared from pearl millet starch and carrageenan gum: process optimization using response surface methodology. *International Journal of Biological Macromolecules*, 143(15), 704–713. https://doi.org/10.1016/j.ijbiomac.2019.09.111

Sari, D. K., Kustiningsih, I., Heriyanto, H., Wijayanto, A. S., & Maulan, A. I. (2021). Physicochemical properties of semi-refined carrageenan by bleaching pretreatment. *Teknika: Jurnal Sains dan Teknologi*, 17(1), 8–14. https://dx.doi.org/10.36055/tjst.v17i1.9714

Senanayake, S., Gunaratne, A., Ranaweera, K. K. D. S., & Bamunuarachchi, A. (2014). Effect of hydroxypropylation on functional properties of different cultivars of sweet potato starch in Sri Lanka. *Int. J. Food Sci*. 1, 148982. https://doi.org/10.1155/2014/148982

Shahbazi, M., Majzoobi, M.., & Farahnaky, A. (2018). Physical modification of starch by high-pressure homogenization for improving functional properties of κ-carrageenan/starch blend film. *Food Hydrocolloids*. 85, 204–214. https://doi.org/10.1016/j.foodhyd.2018.07.017

Shahira, S. F., Subagio, A., & Diniyah, N. (2023). Pengaruh suhu pemanasan dan konsentrasi terhadap karakteristik kimia dan fungsional pada modifikasi pregelatinisasi MOCAF. *Jurnal Keteknikan Pertanian Tropis dan Biosistem,* 11(2), 207–219. https://doi.org/10.21776/ub.jkptb.2023.011.02.10

Shang, L., Wu, C., Wang, S., Wei, X., Li, B., & Li, J. (2021). The influence of amylose and amylopectin on water retention capacity and texture properties of frozen-thawed konjac glucomannan gel. *Food Hydrocolloids*, 113, 106521. https://doi.org/10.1016/j.foodhyd.2020.106521

Shen, Y., Zhang, N., Xu, Y., Huang, J., Yuan, M. A., Wu, D., & Shu, X. (2019). Physicochemical properties of hydroxypropylated and cross-linked rice starches differential in amylose content. *International Journal of Biological Macromolecules*, 128, 775–781. https://doi.org/10.1016/j.ijbiomac.2019.01.194

Shi, S., Pan, K., Yu, M., Li, L., Tang, J., Cheng, B., Liu., J., Cao, C., Jiang, Y. (2022). Differences in starch multi-layer structure, pasting, and rice eating quality between fresh rice and 7 years stored rice. *Current Research in Food Science*, 5, 1379–1385. https://doi.org/10.1016/j.crfs.2022.08.013

Sriwahyuni, E., Nugraheni, F. N., & Widyaningsih, T. D. (2020). Swelling power and solubility profile of different types of carrageenan for pharmaceutical capsule application. *Indones. J. Pharm*, 31(1), 45–51.

Sukhija, S., Singh, S., & Riar, C. S. (2016). Effect of oxidation, cross-linking and dual modification on physicochemical, crystallinity, morphological, pasting and thermal characteristics of elephant foot yam (*Amorphophallus paeoniifolius*) starch. *Food Hydrocolloids*, 55, 56–64. https://doi.org/10.1016/j.foodhyd.2015.11.003

Syukri, D., & Dina, M. (2022). Kumpulan informasi terkait penelitian dan pengembangan produk olahan berbahan baku jagung. Andalas University Press.

Tanak, Y. (2016). Modifikasi secara *heat moisture treatment* pada pati ubi jalar ungu untuk pangan fungsional. *Jurnal Sains dan Teknologi Tadulako*, 5(1), 39–48.

Tang, H., Jiang, P., Li, Y., & Liu, X. (2023). Effects of carboxymethylation and hydroxypropylation on properties and structure of different starches. *Cellulose*

Chemistry and Technology, 57(5-6), 501-503. https://doi.org/10.35812/CelluloseChemTechnol.2023.57.44

Tischer, P. C. S. F., Noseda, M. D., de Freitas, R. A., Sierakowski, M. R., & Duarte, M. E. R. (2006). Effects of iota-carrageenan on the rheological properties of starches. *Carbohydrate Polymer*, 65(1), 49–57. https://doi.org/10.1016/j.carbpol.2005.12.027

Ulbrich, M., Bai, Y., & Flöter, E. (2020). The supporting effect of ultrasound on the acid hydrolysis of granular potato starch. *Carbohydrate Polymers*, 230, 115633. https://doi.org/10.1016/j.carbpol.2019.115633

Ulbrich, M., Flöter, E. (2020). Modification of starches with different amylose/amylopectin ratios using the dual approach with hydroxypropylation and subsequent acid-thinning-impacts on morphological and molecular characteristics. *Starch-Stärke*, 72, 11–12. https://doi.org/10.1002/star.202000015

Verem, T. B., Dooshima, I., Ojoutu, B. E. M, Owolabi, O. O, & Onigbajumo, A. (2021). Proximate, chemical and functional properties of wheat, soy and moringa leaf composite flours. *Agriculgtural Sciences*, 12(1), 18–38. https://doi.org/10.4236/as.2021.121003.

Wang, L., Li, D., Ye, L., Zhi, C., Zhang, T., & Miao, M. (2024). Characterizations and film-forming properties of different fractionated high-amylose maize starches subjected to hydroxypropylation. *Food Chemistry*, 440, 1-10. https://doi.org/10.1016/j.foodchem.2023.138177

Wang, W., Zhu, L., Li, K., & Yang, C. (2017). Optimization of extraction and purification of iota-carrageenan from *Eucheuma denticulatum* for pharmaceutical use. *Int. J. Biol. Macromol*, 104, 1185–1193.

Winarno, F. G. (2002). Kimia pangan dan gizi. PT Gramedia Pustaka Utama.

Wongphan, P., Nerin, C., & Harnkarnsujarit, N. (2024). Modifying cassava starch via extrusion with phosphate, erythorbate and nitrite: phosphorylation, hydrolysis and plasticization. *Polymers*, 16(9), 2787. https://doi.org/10.3390/polym16192787.

Wulandari, E., Sukarminah, E., Mawdawati, E., & Furi, H. L. (2019). Profil gelatinisasi tepung sorgum putih termodifikasi amilase. *Jurnal Teknologi dan Industri Pangan*, 30(2), 173–179. https://doi.org/10.6066/jtip.2019.30.2.173

Yuan, Y., Gao, Y., Zhao, J., & Mao, L. (2020). Water interaction and functional properties of hydrocolloids in food applications–a review. *Carbohydrate Polymers*, 239, 116236.

Yuniar. (2006). Pengaruh hidroksipropilasi pati garut (*Maranta arundinacea* L) dan jenis pemlastis terhadap karakteristik film edible. [Tesis]. Institut Pertanian Bogor.

Zehra, N., Ali, T. M., & Hasnain, A. (2021). Combined effects of hydroxypropylation and alcoholic alkaline treatment on structural, functional and rheological characteristics of sorghum and corn starches. *International Journal of Biological Macromolecules,* 174, 22–31. https://doi.org/10.1016/j.ijbiomac.2021.01.096

Zeleny, M. (1992). Multiple criteria decision making. McGraw-Hill.