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Integrating Marine and Agro-Waste Biopolymers in Functional Syrup Formulation: Optimization of Fucoidan–Pectin Composite from Brown Seaweed and Watermelon Albedo

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Abstract

Developing functional food products from sustainable bio resources has gained growing interest, particularly in addressing lifestyle-related diseases such as dyslipidemia. This study aimed to formulate and optimize a composite syrup incorporating pectin extracted from watermelon (Citrullus lanatus) albedo and fucoidan isolated from brown seaweed (Sargassum crassifolium), two biopolymers with known lipid-lowering potential. Three formulation ratios of pectin to fucoidan (66.67:33.33, 83.33:16.67, and 100:0) were evaluated for their effects on the syrup's physicochemical (viscosity, total dissolved solids, pH, colour) and sensory (taste, texture, colour, and overall acceptability) properties. The results showed that increasing pectin content significantly enhanced viscosity and pH ($p<0.05$), while higher fucoidan content increased total dissolved solids and altered the colour profile. Sensory evaluation revealed that the formulation containing 83.33% pectin and 16.67% fucoidan achieved the highest overall acceptance ($p<0.05$). These findings highlight the potential of integrating agro-waste and marine-derived biopolymers in a functional syrup matrix, offering a sustainable and consumer-acceptable candidate for nutraceutical development. Nevertheless, the present study is limited to physicochemical and sensory assessments; further investigations on bioactivity, nutritional profile, microbiological stability, and long-term storage are required to substantiate its efficacy and practical application.

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Introduction

Dyslipidemia is still a major health problem in Indonesia. Dyslipidemia is a condition where the cholesterol level in the blood exceeds the standard limit (>200 mg/dl).[1] As many as 35% of Indonesians and 45% of the world's population have dyslipidemia,[2] which, if not treated properly, can lead to the development of atherosclerotic plaque and increase the risk of cardiovascular diseases such as stroke, coronary heart disease, and various other circulatory disorders.[3,4] Statins are widely used as first-line therapy; however, their long-term use can be limited by intolerance, resistance, and adverse effects such as hepatotoxicity and elevated diabetes risk.[5,6] Several studies have also shown potential side effects of long-term statin consumption, namely liver problems due to hepatotoxicity, increased risk of diabetes mellitus, and hair loss.[7] These drawbacks have prompted increasing interest in safe, natural alternatives that can be consumed regularly in the form of functional foods or nutraceuticals.

Fucoidan is known to have potential as an anti-dyslipidemia with its ability to reduce the expression of mature SREBP-2 protein and then reduce the expression of the HMG-CoA-R gene in the liver related to cholesterol synthesis.[8] The activity of fucoidan in lowering total cholesterol, triglycerides, and LDL levels in the blood has been shown.[8,9] Fucoidan is abundant in brown seaweed in the Indonesian Ocean.[10,11] Another compound that has potential as an anti-dyslipidemia is pectin which is abundant in fruit skin.[12] Pectin also plays a role in inhibiting cholesterol synthesis and preventing cholesterol absorption.[13] One potential source of pectin widely found in Indonesia is watermelon albedo, which contains 21.03%.[14] Kendalagung Village, Rembang Regency, Central Java, is one of the largest watermelon producers in Central Java.

Previous studies have extensively explored the utilization of pectin in various functional systems, including its role as a stabilizing agent in emulsion matrices and as a structural component in composite materials.[15] In biomedical research, pectin has frequently been combined with other biopolymers to enhance mechanical strength, biodegradability, and targeted delivery properties, underscoring its versatility beyond conventional food applications.[16] Fucoidan, on the other hand, has been predominantly examined as a single bioactive compound, with evidence supporting its individual roles in cardiovascular protection, anti-inflammatory modulation, immune-regulation, and anticancer activities across diverse pharmaceutical and nutraceutical platforms.[17,18]

While previous studies have investigated fucoidan- or pectin-based formulations individually, no prior research has examined their integration into a single composite matrix. The rationale is that combining biopolymers with distinct functional and structural properties may produce synergistic effects, enhancing physicochemical stability, sensory quality, and potential bioactivity. Such an approach could generate innovative nutraceutical beverages with improved acceptability and efficacy compared to single-polymer formulations. A composite is a mixed system of two or more types of materials with different properties.[19] Compared to a single formula, a composite formula can increase solubility, reduce toxicity in the body, increase pharmacological activity, increase stability, increase biodegradability, and provide superior properties to the preparation.[20] This study aims to determine the best pectin fucoidan composite syrup formulation based on its physical and sensory characteristics. This work highlights the novelty of integrating agro-waste-derived and marine-derived biopolymers into a functional syrup matrix and addresses the existing gap in the literature regarding composite hydrocolloid applications in nutraceutical development.

Materials and Methods

Pectin was extracted from watermelon (*Citrullus lanatus*) albedo collected from Central Java, Indonesia, following a modified acid extraction method by (Baghdadi *et al*., 2023).[21] Briefly, the albedo was dried, ground, and extracted in 0.05 M citric acid (solid-to-liquid ratio 1:20 w/v) at 85 °C for 90 min, followed by ethanol precipitation (95%), filtration, and freeze-drying. The extracted pectin was characterized and found to contain 6.42% methoxyl content and 47.27% galacturonic acid, indicating a moderately esterified pectin suitable for functional food formulation.

Fucoidan was isolated from brown seaweed (*Sargassum crassifolium*) obtained from Sulawesi, Indonesia, using hydrothermal-assisted extraction (HAE) with 0.1 N HCl at 120 °C for 62 min and a solid-to-liquid ratio of 1:30 (w/v). The crude extract was subsequently purified through a three-step process involving ethanol and $CaCl_2$ precipitation, ultrafiltration with a 10 kDa MWCO membrane, and solid-phase extraction to obtain a high-purity fucoidan fraction. The purified fucoidan was freeze-dried and stored at 4 °C until further use.[22] The extraction process produced a fucoidan yield of 0.88% (w/w) containing 18.2% of sulphate content, and ready to use as the syrup formulation.

Optimization of Pectin-Fucoidan Composite Formulation

Isolated watermelon albedo pectin (WAP) and brown seaweed fucoidan (BSF) are mixed into a composite with several percentage variations, which are shown in Table 1.

Table 1. Composite Formulation (total composite 3 gr)

Composite Formula	WAP (%)	BSF (%)
K1	66,67	33,33
K2	83,33	16,67
K3	100	0

Composite Syrup Making

Three composite formulations were prepared with varying pectin-to-fucoidan ratios (66.67:33.33, 83.33:16.67, and 100:0 w/w). The syrup base consisted of WAP-BSF composite (3 g), citric acid (0.9 g), propylene glycol (3 mL) to incorporate as a humectant and co-solvent to enhance the solubility of active components and maintain moisture stability in the Fucopect syrup,[23] carboxymethyl cellulose (0.2 g) as a viscosity modifier and stabilizing agent, improving the texture and preventing phase separation in the Fucopect syrup formulation,[24] honey (80 mL) as sweetener, lychee essence (5 mL), and distilled water (until 400 mL of solution). Pectin was dissolved in the aqueous phase at 60 °C, cooled to room temperature, and subsequently fucoidan was incorporated under constant stirring until homogeneity was achieved. The total volume of syrup prepared for each formulation was approximately 400 mL. All formulations were prepared in triplicate from independent batches to ensure reproducibility.

Viscosity Analysis

Analysing the viscosity of syrup using a Brookfield viscometer begins by ensuring the syrup is homogeneous and at the desired temperature (typically 25°C). Choose an appropriate spindle (usually LV or RV for syrup) and attach it to the viscometer. Calibrate the device as per the manufacturer's instructions. Submerge the spindle in the syrup without touching the container sides and set the desired RPM. Record the viscosity once readings stabilize, and express the results in centipoise (cP).[25]

Total Dissolved Solids

Analysing the total dissolved solids (TDS) in syrup using a Refractometer (Atago PAL-1). The analysis begins by calibrating the TDS meter as per the manufacturer's instructions. The syrup sample was homogenized and then centrifuged at 4,000 rpm for 10 minutes at room temperature to obtain a clear supernatant. The supernatant was carefully collected and used for °Brix determination using a digital refractometer previously calibrated with distilled water.[26]

Colour Analysis

Colour analysis was done by colour reader L*a*b*. First, calibrate the device with a white reference tile to perform a colour analysis of syrup using a colour reader in the Lab* colour space. Then, prepare the syrup by ensuring it is homogeneous and free of bubbles, and place it in a transparent container. Position the sensor of the colour reader on the container and take the reading. The device will display L* (lightness), a* (green to red), and b* (blue to yellow) values, which represent the syrup's colour. Record these values for further analysis or comparison.

Sensory Analysis

Sensory analysis was conducted using the hedonic method to see the level of panellist acceptance of the sensory characteristics of the composite syrup. The hedonic test was conducted on 25 untrained panellists. The test was conducted on the characteristics of taste, colour, texture (thickness), and overall with four scales (1: very dislike, 2: dislike, 3: like, 4: very like).

Statistical Analysis

Data will be analysed using the variance analysis (ANOVA) test, which uses a significance level of 5%. If the results of the analysis of ANOVA show the effect of the treatment, then further tests with

Duncan's multiple range test (DMRT) will be carried out to determine the differences between treatments. Data analysis was performed using the SPSS 26.0 application for Windows. Organoleptic test data from this study will be analysed using the Kruskal-Wallis test with a significance level of 5%. When the results of the Kruskal-Wallis test show the effect of the treatment, further tests will be carried out with the Mann-Whitney test.

Future Scope

Although the present study establishes the physicochemical and sensory basis of pectin–fucoidan syrup, future investigations should assess bioactivity through *in vivo* and clinical validation, microbiological stability, nutritional composition, solubility, swelling capacity, and advanced structural characterization of fucoidan–pectin interactions.

Results

Composite syrup's physical and chemical characteristics, including viscosity, total dissolved solids (TDS), pH, and colour, have been analyzed. The physical quality of syrup, especially viscosity and Total Dissolved Solids (TDS), must be tested in the syrup production process. These parameters determine the level of consumer acceptance. The pH parameter is related to the safety of the syrup, ensuring that the syrup is within a safe pH range and under the established standards.[27]

Table 2: Viscosity, Total Dissolved Solids (TDS), and pH of composite syrup

Composite Syrup Formula	Viscosity (cP)	Total Dissolved Solids (°Brix)	pH
K1	9.67±1.15^{b}	23.70±0.10^{a}	3.10±0.01^{b}
K2	11.00±1.00^{b}	23.37±0.15^{b}	3.13±0.01^{a}
K3	13.67±0.58^{a}	22.00±0.20^{c}	3.14±0.01^{a}

*Data are presented as mean ± standard deviation (n = 3). Different superscripts in the same column and row indicate significant differences ($p < 0.05$).

Syrup viscosity, which measures a liquid's thickness and flow, directly influences texture, taste, and consumer experience. Viscosity testing was performed using rotational viscometry with a Brookfield viscometer. Rotational viscometry is one of the most commonly chosen methods due to its accuracy in measuring the torsion required to overco (Ciursa and Oroian, 2021) instance.[28] Data are presented in Table 2.

The physicochemical analyses demonstrated that increasing the proportion of watermelon albedo pectin (WAP) significantly enhanced syrup viscosity ($p = 0.012$, $\eta^2 = 0.42$) and pH ($p = 0.037$, $\eta^2 = 0.28$), whereas higher concentrations of brown seaweed fucoidan (BSF) contributed to elevated total dissolved solids (TDS) ($p = 0.009$, $\eta^2 = 0.47$) and a darker colour profile (reduction in L* values, $p < 0.05$).

The viscosity of the composite syrup varied significantly ($p < 0.05$) among formulations, ranging from 9.67 ± 1.15 cP to 13.67 ± 0.58 cP (Table 2). The highest viscosity was recorded in formulation K3 (100% pectin), while K1 (66.67% pectin: 33.33% fucoidan) showed the lowest value. This increase in viscosity with higher pectin concentration indicates that pectin contributed more effectively to the formation of a cohesive and structured matrix through hydrogen bonding and water-binding interactions.[29] The total dissolved solids (TDS) or °Brix values of the composite syrup decreased with increasing proportions of watermelon albedo pectin (WAP) and decreasing brown seaweed fucoidan (BSF), demonstrating that the compositional ratio of polysaccharides affects the soluble solid fraction. This result aligns with the principle of refract metric measurement, where the °Brix value reflects the refractive index determined by the concentration of optically active solutes in solution.[30] The pH of the composite syrup ranged from 3.10 ± 0.01 to 3.14 ± 0.01, showing significant differences ($p < 0.05$) among formulations. The lowest pH was observed in K1 (3.10 ± 0.01) with the highest fucoidan content, while higher pectin levels in K2 and K3 slightly increased the ph. This trend indicates that pectin's weaker acidity compared to fucoidan's sulphate groups results in a less acidic environment.[31]

Table 3: Colour of composite syrup

Composite Syrup Formula	L*	a*	b*
K1	45.00±0,10[c]	1.47±0,15[a]	12.07±0,40
K2	49.23±0,25[b]	1.40±0,10[a]	12.83±0,11
K3	55.06±0,64[a]	1.07±0,15[b]	12.23±0,56

*Data are presented as mean ± standard deviation (n = 3). Different superscripts in the same column and row indicate significant differences ($p < 0.05$).

The colour parameters (L*, a*, b*) of the composite syrup formulations are presented in Table 3. The L* values, representing lightness, ranged from 45.00 ± 0.10 to 55.06 ± 0.64 and showed significant differences ($p < 0.05$) among treatments. Formulation K3 (100% pectin) exhibited the highest brightness (L* = 55.06 ± 0.64), whereas K1 (66.67% pectin: 33.33% fucoidan) showed the lowest (L* = 45.00 ± 0.10). The a* value, indicating the red–green spectrum, decreased slightly with increasing pectin concentration, from 1.47 ± 0.15 in K1 to 1.07 ± 0.15 in K3. Meanwhile, the b* values, representing the yellow–blue component, ranged narrowly from 12.07 ± 0.40 to 12.83 ± 0.11, showing no significant variation. These results indicate that higher fucoidan levels produced darker syrup with more intense red tones, while higher pectin concentrations yielded lighter-coloured formulations.

Table 4: Sensory hedonic test of composite syrup

Composite Syrup Formula	Colour	Taste	Viscosity	Over all
K1	2.72±0,73	2.60±0,82[b]	2.72±0,84	2.84±0,85[b]
K2	2.84±0,80	3.08±0,76[a]	3.04±0,73	3.08±0,70[a]
K3	2.96±0,61	3.08±0,76[a]	3.04±0,88	3.40±0,71[ab]

*Data are presented as mean ± standard deviation (n = 3). Different superscripts in the same column and row indicate significant differences ($p < 0.05$).

The sensory hedonic evaluation of the composite syrup formulations is summarized in Table 4. The attributes assessed included colour, taste, viscosity, and overall acceptability. The mean scores for colour ranged from 2.72 ± 0.73 to 2.96 ± 0.61, showing no significant difference ($p > 0.05$) among treatments. In contrast, significant variations ($p < 0.05$) were observed in taste and overall acceptability. The K1 formulation (66.67% pectin: 33.33% fucoidan) received the lowest scores for taste (2.60 ± 0.82) and overall preference (2.84 ± 0.85), while K2 (83.33% pectin : 16.67% fucoidan) achieved higher ratings for both taste (3.08 ± 0.76) and overall acceptability (3.08 ± 0.70). The K3 formulation (100% pectin) showed the highest overall mean score (3.40 ± 0.71), although the difference from K2 was not statistically significant. Viscosity scores ranged narrowly between 2.72 ± 0.84 and 3.04 ± 0.88, with no significant differences among samples. These results indicate that the syrup containing 83.33% pectin and 16.67% fucoidan (K2) provided the most balanced sensory profile and was generally preferred by panelists in terms of taste and overall acceptability.

Discussion

The overall findings of this study reveal that variations in the ratio of watermelon albedo pectin (WAP) and brown seaweed fucoidan (BSF) substantially influenced the physicochemical and sensory characteristics of the composite syrup (Table 2, 3, and 4). The interaction between WAP and BSF suggests possible synergistic effects on viscosity, where the combination of neutral polysaccharide backbones (pectin) and negatively charged sulphate groups (fucoidan) may enhance intermolecular hydrogen bonding and electrostatic interactions.[32,33] This could explain the observed improvements in textural consistency and the balanced sensory profile of the composite syrup, especially with the highest WAP content (K3). Similar synergistic behaviour has been documented in hydrocolloid composites, where structural complementarity contributes to improved rheological and functional properties.[34] These findings are also consistent with previous reports by Arioui *et al*., (2017),[35] which indicates that increasing pectin concentration results in higher viscosity in various food products, including syrups and gels.

Fucoidan, a sulphated polysaccharide composed of fructose, galactose, and bound sulphate groups, exhibits high solubility and ionic character, which increases the refractive index due to the presence of low-molecular-weight, light-interacting solutes.26 Consequently, formulations with higher fucoidan content (K1) display higher °Brix values. In contrast, WAP is rich in galacturonic acid and characterized by high hydrophilicity tends to form a polymeric network that binds water molecules and entraps some sugar components, reducing the concentration of freely dissolved solutes detectable by the refractometer.[36] The replacement of fucoidan with pectin therefore decreases ionic interactions and solute mobility, leading to lower TDS readings. Overall, the decrease in °Brix with increasing WAP content reflects not only reduced free sugar and soluble solid concentration but also differences in molecular hydration and optical properties, emphasizing that the balance between sulphated and carboxylated polysaccharides governs the physicochemical behaviour of the composite syrup system.

The more WAP added, the higher the composite syrup's pH value. This is related to the acidic condition of pectin-containing carboxyl groups (-COOH), which can release H+ ions in solution.[37] Studies have shown that the pH of pectin solutions plays a crucial role in affecting their viscosity and gel-forming properties, with lower pH levels typically leading to increased viscosity as gel networks are formed.[38] Meanwhile, the connection between pH and viscosity directly influences the texture and mouthfeel of syrups and their stability and shelf life. Research indicates that pectin solutions demonstrate shear-thinning behaviour, where the viscosity decreases with increasing shear rate, which is affected by pH levels.[39] Moreover, pH influences the electrostatic interactions among pectin molecules, further impacting syrups' viscosity and overall rheological properties.[40]

The colour analysis results showed that the difference in WAP and BSF concentrations significantly affected the composite syrup's brightness (L*) and red-green colour spectrum (b*). The more BSF added, the smaller the brightness value of the composite syrup. This is likely due to the dark-brown colour of BSF, which is getting darker. Research has also shown that adding darker ingredients can significantly alter the colour profile of food products, leading to lower brightness values.[41] The results showed that the higher the BSF concentration, the higher the intensity of the red colour. This is related to the higher red spectrum in the composite syrup with higher BSF. The red colour from the BSF raw material increases the intensity of the red colour spectrum of the composite syrup[42] This observation is consistent with studies indicating that darker syrups typically display lower brightness and greater red spectrum intensity due to the presence of phenolic compounds and other colour -contributing substances, especially on BSF.[43]

Hedonic sensory testing is crucial in evaluating consumer acceptance of functional syrups formulated with Brown Seaweed Fucoidan (BSF) and Watermelon Albedo Pectin (WAP) for dyslipidemia. This testing aims to assess various sensory attributes, including colour, taste, viscosity, and overall, significantly influencing consumer choice. The results of the study showed that overall, the composite syrup that the panellists most preferred was the composite syrup with the addition of PAS 83.33% and FRLC 16.67% of the total composite ($p = 0.021$, $r = 0.36$). The hedonic sensory values for colour and viscosity were not significantly different, suggesting that the amounts of WAP and BSF added were too low to be perceived distinctly by human senses. WAP and BSF do not have any specific sensory characteristics and tend to be bland.[44] The neutral characteristics of WAP and BSF likely contribute to their limited impact on sensory perception. Pectin, for example, is commonly described as having a mild, neutral flavour, making it less noticeable in formulations unless used in high concentrations.[45] Similarly, fucoidan, despite its functional benefits, lacks strong sensory traits that would significantly influence consumer preferences unless applied in more significant quantities.[46] This supports the idea that food products' sensory attributes are shaped by their specific ingredients' specific qualities, with neutral ingredients contributing little to enhancing the overall sensory experience.[47]

Overall, the results confirm that the integration of agro-waste-derived pectin and marine-derived fucoidan in a syrup matrix is technically feasible and produces formulations with acceptable physicochemical and sensory attributes. The identified optimal ratio (83.33% pectin and 16.67% fucoidan) offers a strong basis for subsequent investigations into bioactivity validation, nutritional profiling, stability testing, and benchmarking against commercial functional syrups.

Despite these promising outcomes, the present study was limited to physicochemical and sensory characterization. Critical aspects such as microbiological stability, nutritional composition, solubility dynamics, swelling, and advanced characterization of molecular interactions (e.g., FTIR, NMR, or rheological modelling) would further clarify the mechanisms underlying the synergistic effects observed.

Conclusion

This study demonstrated the feasibility of developing a functional syrup by integrating pectin from watermelon albedo and fucoidan from brown seaweed into a composite matrix. The ratio of 83.33% pectin to 16.67% fucoidan produced the most favourable physicochemical and sensory characteristics, with significant improvements in viscosity, pH, and overall consumer acceptance. These findings highlight the potential of combining agro-waste–derived and marine-derived biopolymers as sustainable ingredients for nutraceutical applications. However, the present work was limited to physicochemical and sensory evaluations, and therefore the product should be considered a promising candidate rather than a validated nutraceutical. Future studies are warranted to investigate bioactivity in in vivo and clinical models, assess microbiological stability and nutritional properties, and perform advanced characterization of fucoidan–pectin interactions.

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Conflict of Interest

The authors do not have any conflicts of interest.

Data Availability Statement

All data generated or analysed during this study are included in this published article. The datasets supporting the findings, including raw data for physicochemical measurements and sensory evaluations, are available upon reasonable request from the corresponding author. No proprietary or restricted-access data were used. This ensures complete transparency and allows other researchers to replicate or further explore the study's results.

Ethics Statement

This research does not involve any clinical trials that requires ethical approval.

Informed Consent Statement

This study did not involve human participants, and therefore, informed consent was not required.

Clinical Trial Registration

This research does not involve any clinical trials.

Permission to Reproduce Material from Other Sources

Not Applicable.

Author Contributions

- **Siti Susanti:** conceptualization, supervision, and manuscript writing.
- **Ulfah Amalia:** conducting the research and data collection
- **Ima Wijayanti:** data processing and analysis.
- **Yasmin Aulia Rachma:** writing and corresponding author

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