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Effect of Defatted Rice Bran Content on Physicochemical and Sensory Properties of Edible Cutlery Made from Rice Flour Green Composites using Compression Molding

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Abstract

The optical properties, moisture content, water activity, water absorption, mechanical properties, morphological observations, and sensory properties of edible spoons made from rice flour and rice flour green composites were investigated. The rice flour green composites were prepared with different weight ratios of rice flour and defatted rice bran as 100:0, 75:25, 50:50, and 25:75 by compression molding at 150°C at a fixed pressure of 9 bar for 7 min. Increasing the weight proportion of defatted rice bran in rice flour green composites resulted in higher lightness values, yellowness values, and water absorption, but redness values and moisture content decreased. Weight proportions of defatted rice bran in rice flour green composites up to 50 improved mechanical properties (both Izod impact and flexural strength). The cross-sectional surface after flexural testing of rice flour spoons was smooth, whereas rice flour green composite spoons were rough. Sensory properties of rice flour green composite spoons showed reduced appearance scores, color scores, texture scores, and overall preference scores but odor scores increased compared with rice flour spoons. Maximum compression force of rice flour green composite spoons increased with increasing weight proportion of defatted rice bran. An increase in maximum compression force measured by the mechanical testing equipment was related to a decrease in texture scores. Spoons made from rice flour and rice flour green composites showed promise as single-use edible cutlery and packaging.

Keywords: *green composites; edible cutlery; rice flour; defatted rice bran; compression molding; physicochemical properties; sensory properties*

1. Introduction

In 2019, the European Union (EU) adopted the EU directive 2019/904 which bans single-use products (SUPs) such as plastic bags, plastic bottles, plastic lids, takeaway and fast-food packaging (including disposable coffee cups and lids), plastic cutlery and plates (including plastic coffee stirrers and straws), plastic packets and wrappers, cotton buds with plastic rods, sanitary towels, tampons, wet wipes, and cigarette butts. The main purpose of the directive was to prevent and reduce the impact of non-biodegradable products on the natural environment and human health. Plastic cutlery comprises SUPs used for a short time before being thrown away (Dybka-Stępień et al., 2021; Kiessling et al., 2023).

Disposable cutlery is widely used in fast-food restaurants, take-out restaurants, catering establishments, and food delivery businesses. Disposable cutlery is normally made from non-biodegradable materials, such as polypropylene (PP), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polyvinyl chloride (PVC), polyethylene terephthalate (PET), and polystyrene (PS) (Dybka-Stępień et al., 2021; Kiessling et al., 2023). These petroleum-based plastics pollute the natural environment. Biodegradable plastics are one alternative to overcome these problems through reduction of greenhouse gas emissions, reduction of cost for plastic waste management, and increased soil fertility.

Biodegradable plastics have now infiltrated everyday life such as thermoplastic starch (TPS), poly(lactic acid) (PLA), poly(butylene adipate-*co*-terephthalate) (PBAT), and polybutylene succinate (PBS) (Jariyasakoolroj et al., 2018). TPS or starch-based plastics demonstrate renewability, non-toxicity, and biodegradability as low-cost raw materials (Jariyasakoolroj et al., 2018; Yokesahachart, & Yoksan, 2011). Starch is normally sourced from plant-based crops such as rice, wheat, corn, sorghum, cassava, mungbean, and soybean. In 2010, Bakey's, an Indian company, began manufacturing edible cutlery made from rice, wheat, and sorghum. They were the first edible cutlery manufacturing startup company (Dybka-Stępień et al., 2021; Roy, & Morya, 2022). Agricultural waste has emerged as a potential source for the production of edible cutlery because it is naturally produced in large quantities and is biodegradable (Dybka-Stępień et al., 2021).

Rice is a vital economic crop in Thailand, both for consumption as the main food and as a major agricultural export product. Kraithong et al., (2018) reported Thai rice exports of 7.4 million metric tons, generating US$ 3.3 million. Thai rice can be classified into two main types as white rice and pigmented rice (Kraithong et al., 2018; Rungsardthong et al., 2023). White rice is a cheap rice species variety and the most widely traded on global markets (Saleh et al., 2019). White rice is classified by grading into different price brackets depending on its moisture content and the proportion of broken rice (Mohd Ramli et al., 2021). A higher proportion of broken rice results in a lower price. Domestic consumption is mainly as rice flour (Saleh et al., 2019), rice-based snacks (Saleh et al., 2019), animal feed (Lazova-Borisova, & Markov, 2023), biomass-based electricity (Alengebawy et al., 2023), and bioethanol (Alengebawy et al., 2023). Defatted rice bran is a major byproduct of the rice bran oil industry and is not yet efficiently utilized for human consumption due to its taste (Zhuang et al., 2019). Defatted rice bran is a rich source of nutrients and nutraceuticals such as starch, protein, non-starch polysaccharides, γ-oryzanol, and polyphenols (Zhuang et al., 2019). Blending rice flour and defatted rice bran to produce edible cutlery has not been reported in the literature. Nevertheless, defatted rice bran was employed as a cost-effective natural filler to reinforce cellular natural rubber vulcanizate (Moonchai et al., 2016). They discovered that the incorporation of defatted rice bran led to a more homogenous dispersion in cellular natural rubber vulcanizate than the addition of calcium carbonate and clay. The cellular natural rubber vulcanizate loaded with defatted rice bran exhibited the highest water absorption value, tensile strength, and modulus, but the lowest hardness value.

Some researchers have studied the properties of edible cutlery, such as biodegradable spoons made by mixing grape seed flour, wheat flour, and sorghum flour using a baking process (Dordevic et al., 2021), and spoons made from natural soy protein isolate added with 5-20 wt% crude morning glory using compression molding (Choeybundit et al., 2022). Dordevic et al., (2021) studied the production and properties of biodegradable spoons by mixing different weight ratios of flour from grape seeds, wheat, and millet using water as a plasticizer. The biodegradable spoons were baked at 180°C and 240°C and then dried in a fruit dehydrator. Increasing amounts of grape seed flour resulted in increased spoon antioxidant properties but decreased hardness. The hardness of samples baked at 240°C was higher than that of spoons baked at 180°C, while the addition of xanthan gum increased spoon hardness. Adding palm oil resulted in decreased hardness, but the spoons had a more attractive appearance.

Choeybundit et al., (2022) prepared spoons using soy protein isolate (SPI) mixed with 5-20 wt% crude morning glory stem fiber (MGSF) and 30 wt% glycerol as a plasticizer. The spoons were fabricated by hydraulic hot press molding at 160°C and pressure of 100 bars for 5 min. Incorporation of MGSF led to the reduction of lightness value, stiffness, impact strength, compression load, degree of swelling, water absorption, and water solubility but an increase in deflection (measured in mm) at a force of 0.05 N. The flexural modulus of SPI increased significantly when 5% MGSF was added, and the distribution of 5 wt% MGSF in the SPI matrix was uniform. The researchers determined that the addition of 5 wt% MGSF maintained the physical and mechanical characteristics of an edible and biodegradable SPI spoon, making it suitable for use with food. However, the sensory characteristics have not yet been fully examined to assess customer perception.

2. Objectives

This research studied the effect of defatted rice bran (DRB) content on the physicochemical properties of rice flour (RF) green composites, including optical properties, moisture content, water activity, water absorption, mechanical properties, and microstructure. Sensory properties were also investigated for customer perception.

3. Materials and methods

3.1 Materials

Rice flour was prepared by milling Soa Hai rice (kung koo thong kham brand) acquired from Lorpoonphol Rice Mill Co., Ltd. (Thailand). The defatted rice bran was purchased from Good Thai Feed Co., Ltd. (Thailand). A chemical examination of defatted rice bran revealed moisture, protein, ash, crude fiber, and lipid contents of 11.1 wt%, 14.5 wt%, 14.0 wt%, 15.0 wt%, and 0.9 wt%, respectively. All other ingredients such as baking powder, gum arabic, salt, sugar, and soybean oil were food-grade.

3.2 Preparation of edible spoons from rice flour green composites

Rice and defatted rice bran were pulverized with a SHARP model EM-ICE2 blender (Thailand) and then sieved using a No. 120 sieve (125 microns). The rice flour and defatted rice bran powders were stored in ziplock bags before making the edible spoons. The weight proportions of rice flour to defatted rice bran powder were 100:0, 75:25, 50:50, and 25:75, corresponding to defatted rice bran percentages of 13.9 wt%, 27.8 wt%, and 41.8 wt%, respectively. The quantities of all the ingredients remained unchanged. The composition included 1.7 wt% baking powder, 1.7 wt% gum arabic, 0.9 wt% salt, 5.6 wt% sugar, 6.7 wt% soybean oil, and 27.8 wt% water. The rice flour and defatted rice bran powders and all the components were combined to form a dough. Doughs weighing approximately 7 g were compressed using an LP-30B compression molding machine from Labtech Engineering (Thailand). The process was conducted at 150°C with a heating duration of 7 min and a constant pressure of 9 bar. The samples were stored in a desiccator with a saturated magnesium nitrate salt solution at 25±5°C and a relative humidity of 50±5% before testing. The mixtures were abbreviated as RF/DRB 100/0, RF/DRB 75/25, RF/DRB 50/50, and RF/DRB 25/75. Figure 1A displays the dimensions of the spoon after compression molding.

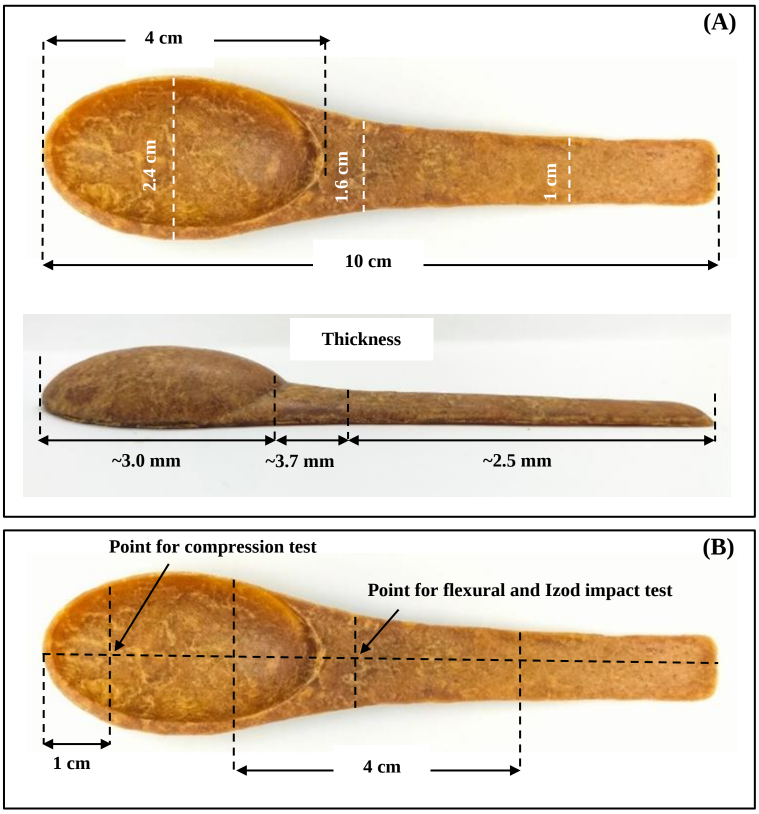

Figure 1 Images of (A) spoon dimensional measurements, and (B) points for mechanical testing of rice flour green composites

3.3 Characterization and property testing of edible spoons from rice flour green composites

Optical properties were examined for color to measure L^*, a^*, b^*, and total color difference (ΔE^*) of the samples using an UltraScan VIS colorimeter (USA) in reflectance mode, based on the CIELab classification system and illuminant D65 (daylight) (HunterLab, 2001). Those parameters were used to determine color with the scales varying from $L^* = 0$ (black) to $L^* = 100$ (white), $a^* = -$ (greenness) to $a^* = +$ (redness), and $b^* = -$ (blueness) to $b^* = +$ (yellowness) (Dalrymple, & Smith, 2018). Initially, the colorimeter was calibrated with a standard plate with white and black before measurement. Each sample was measured five times. The total color difference (ΔE^*) was calculated according to Equation (1) (rice flour spoons as control).

$$\text{Total color difference } (\Delta E^*) = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{0.5} \quad (1)$$

The moisture content of the samples was assessed using the oven-drying method as outlined in AOAC (2000). The water activity of the samples was measured using an AquaLab 4TEV DUO (USA). Ground samples weighing approximately 3 g were placed in a silicone cup and the amount of available water related to microbial growth (van der Hoeven-Hangoor et al., 2014) was determined. All samples for measuring moisture content and water activity were replicated three times. Water absorption determination followed the method of Nugroho et al., (2022). The samples were weighed and recorded for initial weight (W_0) before being immersed in 300 mL of distilled water at room temperature for 60 s. Subsequently, the samples were wiped with tissue paper and then weighed to determine the final weight (W_1). The water absorption percentage was calculated according to Equation (2).

$$\text{Water absorption percentage} = [(W_1-W_0)/W_0] \times 100 \quad (2)$$

The Izod impact strength test was conducted following the standard procedure of ASTM D256-02 (ASTM International, 2002). Samples were placed in a closed chamber with a saturated magnesium nitrate salt solution at 50±5% RH and 25±5°C for 48 hours before testing. The preconditioned samples were tested using a QC-639 mechanical impact tester (Cometech Testing Machines Co., Ltd., Taiwan) with an impact energy of 3 J. Each sample was tested using three specimens. Flexural strength measurement was performed using a TA-XT plus texture analyzer (Stable Micro Systems Ltd., UK) equipped with a three-point bending rig (HDP/3 PB) according to the method of Kouhsari et al., (2022). The test parameters were as follows: load cell capacity 5 kN, support span 40 mm, pre-test speed 60 mm/min, test speed 120 mm/min, post-test speed 600 mm/min, and compression distance 6 mm. All measurements were repeated three times. Figure 1B shows the locations of flexural and Izod impact testing. Cross-sectional morphological observation after flexural testing was performed using a JSM-6610V scanning electron microscope (JEOL, Japan) at an acceleration voltage of 15 kV. The samples were directly mounted on a stub using double-sided carbon tape and then coated with a thin layer of gold. Sensory testing was conducted by academic staff and students (n = 50). All spoon samples were served in hermetically sealed polypropylene bags with random 3-digit codes. Each spoon sample was evaluated for appearance, color, odor, taste, texture, and overall preference using a 9-point hedonic scale (1 = dislike extremely, 2 = dislike very much, 3 = dislike moderately, 4 = dislike slightly, 5 = neither like or dislike, 6 = like slightly, 7 = like moderately, 8 = like very much, 9 = like extremely) (da Rocha Lemos Mendes et al., 2021). Compression testing was measured using a 6 mm diameter cylindrical probe on a TA-XT plus texture analyzer (Stable Micro Systems Ltd., UK). The test settings included load cell capacity 5 kN, pre-test speed 50 mm/min, test speed 50 mm/min, post-test speed 600 mm/min, and compression distance 6 mm. Each sample was replicated three times. The compression testing position was measured at the point of the first bite, which was located 1 cm below the tip of the spoon, as shown in Figure 1B.

4. Results and discussion

Figure 2 illustrates the visual characteristics of the samples. Rice flour spoons were yellow-colored (Figures 2Aa and 2Ba), while rice flour green composite spoons were brown-colored (Figures 2Ab-2Ad and 2Bb-2Bd). The brown color was attributed to the formation of compounds resulting from the Maillard reaction under high heat and pressure, as well as the presence of residual color pigments from lignin in the defatted rice bran fiber. Sharma et al., (2004) discovered that when rice bran was processed by extrusion at a pressure of 275.80 kPa and temperature range of 135-140°C, it turned dark brown. By contrast, rice bran that was heat treated at 120°C for 30 min had a light tan color. The color changes were related to the Maillard reaction during the extrusion process (Sharma et al., 2004) and natural color pigments from rice bran (Sharma et al., 2004; Feng et al., 2023).

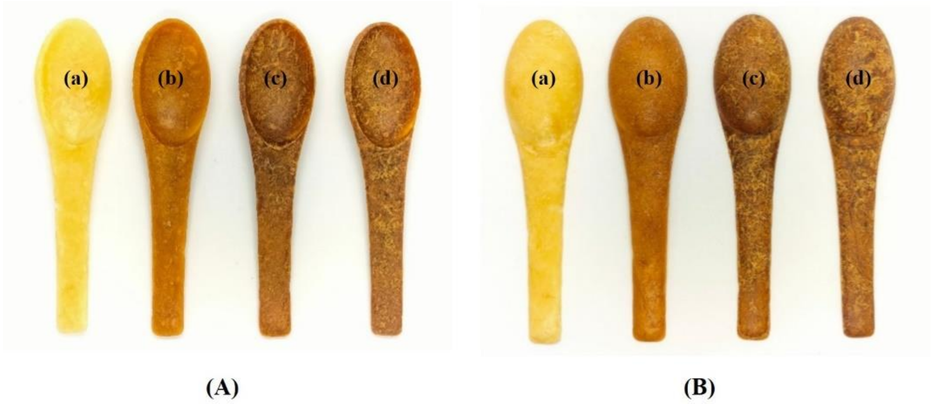

Figure 2 Appearances of (A) concave side, and (B) convex side of different rice flour green composites: (a) RF/DRB 100/0, (b) RF/DRB 75/25, (c) RF/DRB 50/50, and (d) RF/DRB 25/75

Table 1 Optical properties, moisture content, water activity and water absorption of different rice flour green composites

Sample	Optical properties				Moisture content (%)	Water activity (a_w)	Water absorption (%)
	L^*	a^*	b^*	ΔE^*			
RF/DRB 100/0	55.88±3.05^a	3.47±0.42^a	20.66±1.65^a	-	4.16±0.11^c	0.60±0.00^a	8.19±0.40^a
RF/DRB 75/25	48.78±2.22^b	9.00±0.89^b	18.65±2.65^{ab}	9.32±1.23^c	6.82±0.68^b	0.56±0.02^a	6.58±0.56^b
RF/DRB 50/50	40.35±1.37^c	9.75±0.85^b	16.48±0.38^b	17.29±2.08^b	7.23±0.35^b	0.61±0.05^a	6.50±0.25^b
RF/DRB 25/75	36.87±0.84^d	9.26±0.40^b	12.86±1.40^c	21.37±2.01^a	8.01±0.08^a	0.57±0.03^a	6.93±0.34^b

Data are reported as mean±SD (n = 3-5). Different superscripts indicate significant differences at $p < 0.05$ (Duncan's new multiple range test)

Table 1 demonstrates the optical properties (L^*, a^*, and b^* values), moisture content, water activity (a_w), and water absorption percentages. L^*, a^*, and b^* values show lightness, greenness/redness, and blueness/yellowness, respectively. The L^*, a^*, and b^* values of the rice flour spoon were 55.88, 3.47, and 20.66, respectively, whereas those of rice flour green composites ranged from 36.87-48.78, 9.00-9.75, and 12.86-18.65, respectively. Increasing the proportion of defatted rice bran resulted in lower L^* and b^* values, while a^* values increased, indicating that rice flour green composite spoons were darker with redness and yellowness, possibly due to the natural pigments in defatted rice bran and concurring with Feng et al., (2023). They also found that L^* and a^* values of extruded rice noodles decreased but b^* values increased with increasing 3-11 wt% rice bran content, indicating that rice noodles turned darker and more yellowish due to various natural pigments such as polyphenols and carotenoids (Feng et al., 2023). When compared to rice flour spoons, the total color difference (ΔE^*) of the samples was found to be within the range of 9.32-21.37, which indicates that the observers were able to see color difference through their individual eyes. The moisture content of rice flour spoons significantly increased with increasing defatted rice bran content, suggesting that the more hydrophilic rice flour green composites caused hydrophilic chemical compositions in defatted rice bran. Ajmal et al. (2006) also found that bread added with defatted rice bran ranging from 5-20 wt% showed increased moisture content because defatted rice bran contains more cellulose and other non-starch polysaccharides (Ajmal et al., 2006). The a_w of rice flour spoons was 0.60, while the a_w of rice flour green composites ranged 0.57-0.61, and was not significantly different. The moisture absorption of the samples was also investigated. This is governed by the interaction between water molecules and the chemical structures. The water absorption percentage of rice flour spoons was 8.19%, whereas values of rice flour green composite spoons ranged 6.50-6.93%. Increasing defatted rice bran content resulted in significantly decreased water absorption of 0.15-0.21 fold, implying more difficult interaction with water molecules. This was explained by interference among the hydrophilic groups in rice flour by interaction via hydrogen bonding with defatted rice bran. A similar effect of defatted rice bran content on the starch structure was observed by Charunuch et al., (2014). Their results indicated that the presence of more bran in the mixture reduced the availability of the starch granules for gelatinization. This reduced the viscosity and water absorption index because of the replacement of the starch by fiber components.

Table 2 shows the Izod impact strength and flexural strength of the spoon samples. Impact and flexural testing of composite materials are important for mechanical properties. Impact testing measures the impact strength of the composites which is the ability to resist fracture under a sudden impact, whereas flexural testing measures flexural strength which is the ability to resist deformation under flexural loading (Saba et al., 2019). Flexural testing was performed on one side of the composite faces, while the other side underwent compression loading (Saba et al., 2019). Izod impact strength and flexural strength of the rice flour spoons significantly increased at higher weight proportions of defatted rice bran, ranging from 25-50, possibly due to the reinforcing effect of defatted rice bran, similar to the result from thermoplastic wheat flour composites added with 20-50 wt% wheat bran prepared via an extrusion process (Dominici et al., 2020). The Izod impact strength and flexural strength decreased when the weight proportion of defatted rice bran increased to 75 due to fiber agglomeration (as observed from the SEM results). Compared with PP, Izod impact strength and flexural strength values of spoons from rice flour and rice flour green composites were lower than those of PP at around 0.94 and 0.64-0.76 fold, respectively (Izod impact strength and flexural strength of PP spoons were 5.29 kJ/m^2 and 45.03 MPa, respectively).

Figures 3A (100X) and 3B (1,000X) exhibit cross-sectional morphologies after flexural testing of samples at different magnifications. Figures 3Aa and 3Ba show smooth fracture surfaces of rice flour spoons, whereas Figures 3Ab-3Ad and 3Bb-3Bd demonstrate rough fracture surfaces. Adding weight proportions of defatted rice bran ranging from 25-50 demonstrated a uniform distribution of defatted rice bran in the rice flour matrix (Figures 3Ab-3Ac and 3Bb-3Bc), indicating reinforcement by defatted rice bran (Dominici et al., 2020). Formation of fiber agglomeration was observed when the weight proportion of defatted rice bran increased to 75 (Figures 3Ad and 3Bd).

Table 2 Izod impact strength and flexural strength of different rice flour green composites

Sample	Izod impact strength (kJ/m^2)	Flexural strength (MPa)
RF/DRB 100/0	0.30 ± 0.01^{b}	0.94 ± 0.03^{c}
RF/DRB 75/25	0.32 ± 0.00^{a}	1.14 ± 0.04^{b}
RF/DRB 50/50	0.32 ± 0.00^{a}	1.36 ± 0.05^{a}
RF/DRB 25/75	0.30 ± 0.00^{b}	1.17 ± 0.05^{b}
PP spoons	5.29±0.00	45.03±1.55

Data are reported as mean±SD (n = 3). Different superscripts indicate significant differences at $p < 0.05$ (Duncan’s new multiple range test)

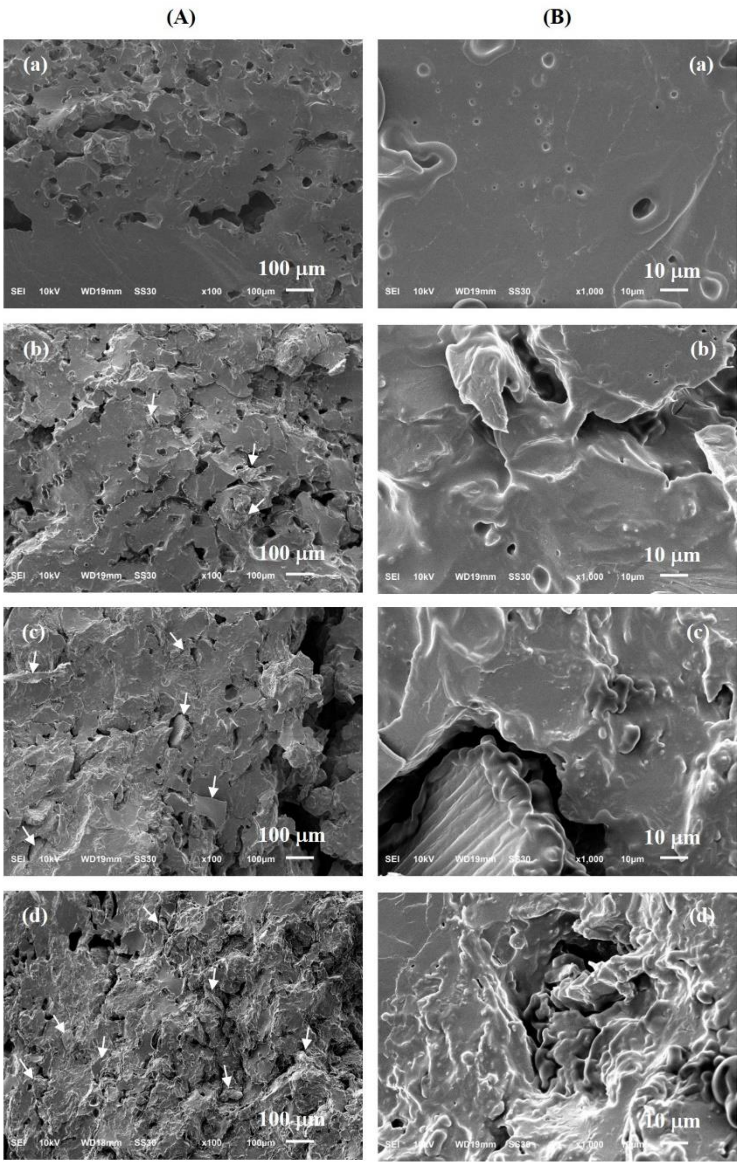

Figure 3 SEM micrographs (A) 100X and (B) 1,000X of different rice flour green composites: (a) RF/DRB 100/0, (b) RF/DRB 75/25, (c) RF/DRB 50/50, and (d) RF/DRB 25/75

Table 3 Sensory properties and compression force of different rice flour green composites

Sample	Sensory properties (minimum value = 1 and maximum value = 9)						Maximum compression force (N)
	appearance	color	odor	taste	texture	overall preference	
RF/DRB 100/0	6.90±1.47[a]	6.94±1.54[a]	5.70±1.31[b]	6.12±1.52[a]	6.08±1.59[a]	6.28±1.44[a]	47.73±1.91[a]
RF/DRB 75/25	6.92±1.28[a]	7.12±1.24[a]	6.62±1.52[a]	6.12±1.69[a]	6.08±1.91[a]	6.46±1.75[a]	63.70±2.23[b]
RF/DRB 50/50	6.74±1.26[a]	6.66±1.30[a]	6.52±1.49[a]	6.50±1.67[a]	5.90±1.58[a]	6.44±1.80[a]	66.30±2.17[bc]
RF/DRB 25/75	5.14±1.93[b]	5.10±1.89[b]	6.14±1.88[a]	5.90±1.69[a]	4.96±2.04[b]	5.12±1.99[b]	68.63±3.23[c]

Data are reported as mean±SD (n = 3 for compression testing and n = 50 for sensory testing). Different superscripts indicate significant differences at $p < 0.05$ (Duncan's new multiple range test)

Table 3 exhibits the sensory properties of different rice flour green composites. Sensory scores of rice flour spoons were 6.90, 6.94, 5.70, 6.12, 6.08, and 6.28 for appearance, color, odor, taste, texture, and overall preference, respectively. Increasing the weight proportion of defatted rice bran resulted in significantly decreased scores of appearances, color, texture, and overall preference but increased scores for odor. Results indicated that rice flour green composite spoons were less acceptable to consumers by appearance and texture compared with rice flour spoons that were darker and had more rigidity. Odor scores of rice flour spoons improved when incorporating small amounts of defatted rice bran, possibly due to chemical components in defatted rice bran, such as lignin. Ajmal et al., (2006) found that bread made with wheat flour added with 5 wt% rice bran showed higher aroma scores (5.91) than bread made from wheat flour and wheat flour added with 10-20 wt% rice bran, possibly because low levels of rice bran improved the aroma by restricting rancid aromas in the final product (Ajmal et al., 2006). However, the incorporation of defatted rice bran ranging from 25-75 did not impact the taste scores. The maximum compression force at the point of the first bite was also measured. The maximum compression force of rice flour spoons was 47.73 N, while those of rice flour green composites were 63.70-68.63 N. Similarly, Dordevic et al., (2021) also found that the hardness of biodegradable spoons made from wheat and millet ranged from 52.04 to 59.51 N. In contrast, Choeybundit et al., (2022) discovered that spoons made from soy protein isolate (SPI) mixed with 5-20 wt% crude morning glory stem fiber (MGSF) had a compression load ranging from 0.08 to 0.1 N, depending on the spoon's composition. Maximum compression force directly related to the sensory property texture scores and increased with the increasing proportion of defatted rice bran from 25-75. The materials became more rigid, corresponding to a decrease in texture scores.

5. Conclusions

Edible spoons from rice flour green composites with weight proportions of defatted rice bran ranging from 25 to 75 were prepared using a compression molding machine at 150°C and a constant pressure of 9 bar for 7 min. Increasing defatted rice bran content resulted in decreased L^* values, b^* values, and water absorption but increased a^* values and moisture content, suggesting that the materials became darker with less water absorption. Izod impact strength and flexural strength increased with increasing weight proportion of defatted rice bran up to 50 due to the reinforcing effect. Sensory properties i.e. appearance, color, odor, taste, texture, and overall preference were also observed for consumer perception. Sensory scores for appearance, color, texture, and overall preference significantly decreased, whereas scores of odors increased when the weight proportion of defatted rice bran increased. Texture scores of spoons with higher weight proportions of defatted rice bran decreased and were directly related to the increased maximum compression force, implying more rigidity. The shelf life of spoons from rice flour and rice flour incorporated with defatted rice bran requires further study in future investigations. The optimal weight proportions of rice flour and defatted rice bran was 75 and 25, respectively. This proportion corresponds to a defatted rice bran percentage that does not exceed 13.9 wt%, which is acceptable to consumers and is utilized appropriately.

6. Acknowledgements

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