Biodegradable containers from rice husk as substitutes for single-use plastic products

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Abstract:

Vietnam ranks among the top four nations for single-use plastic (SUP) waste, trailing only China, Indonesia, and Philippines. The abundant rice husk (RH) by-product of rice cultivation presents an opportunity to fabricate biodegradable containers, potentially supplanting single-use disposables. This study focuses on creating biodegradable containers from RH via thermo-pressing. Utilising the Taguchi orthogonal array method, we examined the impact of the RH to modified starch (MS) ratio (w/w %), pressing temperature (°C), and time (minutes) on the trays' hardness, colour variation, and density, aiming to refine the manufacturing process. Additionally, variations in adhesive concentrations and glycerol were assessed for their influence on the aforementioned properties. The identified optimal conditions for fabricating viable biodegradable containers were a RH and MS composition of 80% (w/w), with a pressing duration of 3 minutes at 150°C. The resulting product exhibited a hardness of 5.26±0.22 kgF. Moreover, the thickness, density, and colour on the front and back sides were measured at 2.05±0.01 mm, 0.97±0.004 g/cm³, 50.87±0.96, and 48.05±0.87, respectively. Consequently, RH-based materials have emerged as promising candidates for crafting consumer-safe, environmentally benign biodegradable containers.

Keywords: **biodegradable containers, hardness, rice husk, thermo-pressing.**

Classification numbers: **2.3, 5.3**

1. Introduction

SUP based on expanded polystyrene (EPS) are prevalent in food packaging due to their low density, superior thermal insulation, high tensile strength, and affordability. However, SUPs derived from petroleum are chemically inert and non-biodegradable, posing significant challenges in recycling and potentially generating toxic byproducts during decomposition [1]. With global plastic production reaching 390.7 million tonnes in 2021 [2], SUPs not only exacerbate environmental issues but also deplete petroleum resources. Commonly, SUP waste is either buried or incinerated, compromising the quality of land, air, water, oceans, and human health [3].

In response, biodegradable composites are being developed as alternatives to SUPs. Research on biodegradable products has largely focused on natural materials. Proteins such as soy protein [4], whey protein [5] and polysaccharides (chitosan, cellulose, starch) [6] have been utilised as raw materials for biodegradable products. Amongst these, starch-based materials are extensively studied due to starch's abundance, biocompatibility, non-toxicity, renewability, and cost-effectiveness. Moreover, the polyhydroxy structure of starch facilitates the modification of its functional and structural properties through chemical or enzymatic treatments [3]. To enable the use of these materials in container applications, reinforcement with cellulose is necessary to enhance tensile strength and load-bearing capacity. Cellulose fibres can form a dense network structure with starch molecules, thanks to the numerous hydroxyl groups on their surfaces [7]. The addition of cellulose fibres is shown to improve thermal properties, tensile strength, and hydrophobicity [8]. Additionally, cellulose is recognised for its excellent biocompatibility, biodegradability, non-toxicity, abundance, and affordability [9]. Agricultural waste, such as banana trunk [10], bagasse [11], RH [12], peanut skin [13], has been repurposed to create green products to supplant SUPs.

In Vietnam, according to the Ministry of Agriculture and Rural Development, the rice yield exceeded 43.86 million tonnes in 2021. Consequently, about 20% of this harvest, equating to 8.772 million tonnes, becomes RH agricultural residue [14]. Typically utilised for animal feed, biomass, and organic fertiliser production, RH with its high cellulose, silica, lignin, and moisture content, and a bulk density ranging from 90-150 kg/m³ presents an opportunity for creating environmentally friendly materials [14].

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To streamline the experimental process, various design and optimisation methods have been explored. The Taguchi method, a well-known approach, utilises statistical analysis to assess the impact of different parameters on the means and variance of a production or operational process [15]. The Taguchi method employs statistical analysis to determine how various parameters influence the means and variance of the production or operation process [16]. The system design procedure aims to establish optimal test levels for design parameters, while the factor design procedure identifies parameter levels that enhance the performance of the product in question.

This study focuses on producing biodegradable trays from RH and starch via a thermo-pressing process. Using the Taguchi method, we investigated how operational factors such as the RH to starch ratio, pressing time, and pressing temperature affect the mechanical properties of the container. Additionally, the influence of additives such as glycerol and adhesive on the colour difference, hardness, and density of the trays was examined.

2. Materials and methods

2.1. Preparation of RH trays

RH was sourced from rice milling operations in Dong Thap province, Vietnam. The husks were ground to a 0.25 mm granularity. Cationic MS VN1605, with a moisture content of 14%, was supplied by Thuan Phat Hung Company. Varied ratios of RH to modified starch (MS) were employed: 20/80, 35/65, and 50/50. Polyvinyl acetate (PVA) from China (87%) and glycerol from Merck (99.5%) were added prior to the pressing process and thoroughly blended using a mixer. For the process, a stainless-steel mould of 150 mm diameter and 2 mm thickness was utilised (Fig. 1) [10]. The thermo-pressing was performed at 130, 150, and 170°C, for durations of 3, 4, and 5 minutes, respectively. Post thermo-pressing, the trays were cut into shape. The process followed a prescribed flowchart (Fig. 2).

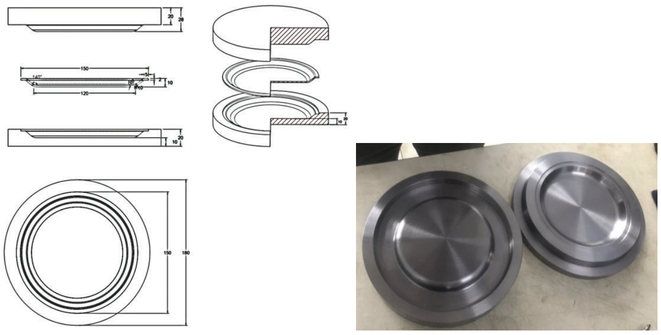

Fig. 1. Mould for the thermo-pressing process.

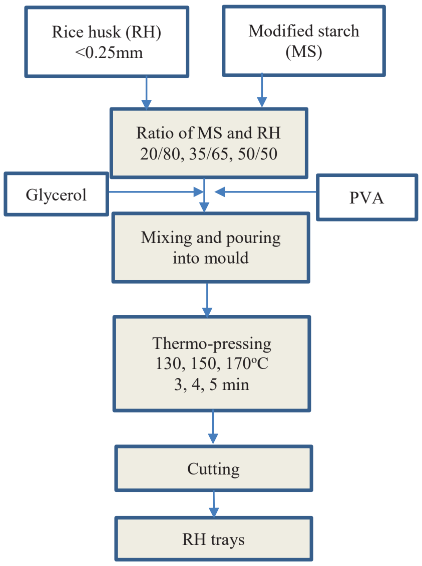

Fig. 2. Flowchart of the RH trays production process.

2.2. Design of experiments for biodegradable RH trays

A preliminary study identified factors impacting product quality, focusing on hardness, colour difference, thickness, and density. Factors examined included concentrations of glycerol, PVA, pressing temperature, time, and the RH to MS ratio. Glycerol and PVA were found to have a negligible effect on the RH tray quality, leading to the selection of three influential factors for the Taguchi method: the ratio of MS to RH (A), pressing temperature (B), and pressing time (C). An orthogonal array was designed for the experiment, requiring nine tests (3^2=9) to economise on time and resources. Each test was conducted thrice (Table 1).

Table 1. The Taguchi method design for RH tray experimentation.

Independent factors	Level 1	Level 2	Level 3
The ratio of MS and RH (A) (w/w)	20/80	35/65	50/50
Pressing temperature (B) (°C)	130	150	170
Pressing time (C), (min)	3	4	5

2.3. Samples analysis

2.3.1. Hardness test

Hardness is defined as a product's resistance to permanent deformation such as indentation, abrasion, wear, and scratch. In this study, a hardness tester (IC-FR5105) was used to determine the load of the container.

2.3.2. Thickness and density

A manual micrometre (Mitutoyo, Japan) was used to measure the thickness of the RH container. The density was calculated using the weight-thickness relationship. The reported value is the mean of three calculations.

2.3.3. Colorimetry

A colour spectrophotometer (CR-400/CR 410) was used and the 0% colour was calibrated with white and 100% with black standards. The colour parameters L*, a*, and b* were recorded for each product. Each product was analysed at six different points on the upper and lower surfaces of the product. The comprehensive colour difference, ΔE, was calculated using the formula:

$$\Delta E = \sqrt{(\Delta L)^2 + (\Delta a)^2 + (\Delta b)^2}$$

where ΔL represents the brightness difference (lightness and darkness); Δa denotes the red-green difference; Δb denotes the yellow-blue difference.

3. Results and discussion

3.1. Effect of additives in RH container process

The role of additives in product processing is crucial and cannot be overstated. Additives may take various forms, contingent upon the desired properties of the product. They can enhance fibre hardness, bonding strength, and resistance to moisture and oil. Additionally, additives can influence fibre and production costs, whiteness, brightness, colour stability, and energy consumption [17]. While additives can augment the natural bonding between fibres, their effect is secondary to that achieved through fibre refining and beating [18]. In this study, the impact of additives was examined using a fixed MS/RH ratio of 50/50 (w/w), a pressing temperature of 170°C, and a pressing time of 3 minutes. We analysed all formulations to optimise tray quality without the presence of cracks

3.1.1. Effect of PVA

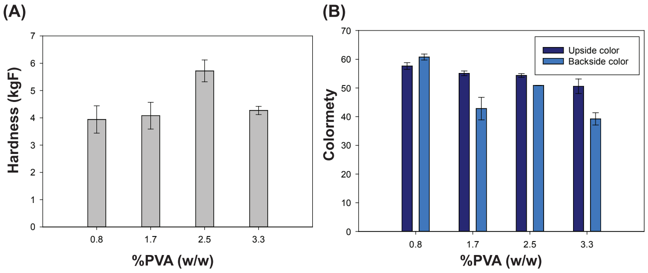

Fig. 3. Effect of poly(vinyl alcohol) on the hardness (A) and colour (B) of biodegradable trays.

Poly(vinyl alcohol) (PVA) is a biodegradable, biocompatible, and hydrophilic synthetic polymer [19]. The trays were hot-pressed for 3 minutes at 170°C and 1 MPa, with PVA concentrations varying from 0.8% to 3.3% (w/w) (Fig. 3). PVA served as an adhesive, bonding the MS and RH fibres. The tray with 2.5% PVA (w/w) exhibited the highest hardness. Beyond this concentration, an increase in PVA reduced the hardness of the product and caused it to adhere more to the mould. The colorimetric differences (ΔE) were measured on both the front and back of the products, revealing a slight fading of colour with increasing PVA concentration. The thickness and density of the products remained consistent at 2.05±0.01 mm and 0.93±0.03 g/cm³, respectively, indicating that the variation in PVA concentration did not significantly impact these parameters. Additionally, the high lignin content within RH fibres contributed to the adhesion of product components and reduced porosity within the trays when subjected to elevated temperatures and pressing force [20].

3.1.2. Effect of glycerol

Glycerol, a prevalent plasticiser in starch and fibre-based biodegradable material production, significantly affects mechanical properties. Its presence can enhance the flexibility of biodegradable containers by reducing their brittleness and permeability to water vapour and oxygen [21]. In this study, glycerol concentrations varied from 3% to 6% (w/w). The hardness of the product increased with glycerol concentrations up to 5% (w/w) but decreased when the glycerol level exceeded 5% (Fig. 4). E. Basiak, et al. (2018) [21] observed that higher glycerol content in starch structures diminishes tensile strength and elastic modulus.

The colour difference values on both sides of the products changed with glycerol concentration. At 3% (w/w), the colour difference value was low and remained stable at higher concentrations. However, the colour did not change markedly with increasing glycerol levels. Glycerol concentration had no significant effect on product thickness or density, with thickness around 2.05±0.01 mm and density approximately 0.9±0.07 g/cm³.

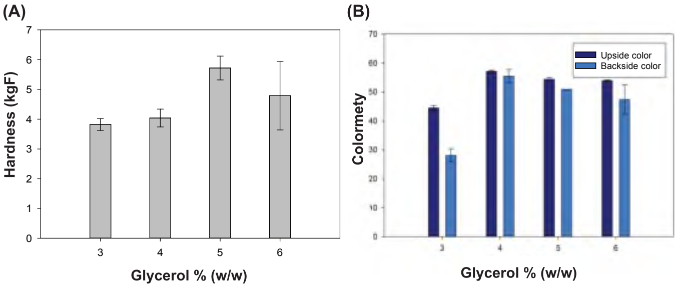

Fig. 4. Effect of glycerol on the hardness (A) and colour (B) difference of biodegradable trays.

3.2. Optimum analysis by Taguchi approach

Given that the additives did not impact the product's quality significantly, the main factors influencing the tray production process were the MS/RH ratio, pressing temperature, and pressing time (Table 2). The Taguchi method was employed to assess the effects of these variables, recommending a signal-to-noise (S/N) ratio to optimise the quality of the biodegradable container. Maximising the S/N ratio implies minimising the variability and effects of noise factors. In this context, biodegradable trays with higher hardness, lower density, and lighter colour were preferred.

Thus, a 'larger-is-better' approach was used for hardness analysis, while a 'smaller-is-better' approach was adopted for analysing tray density and colour (Tables 3-5).

Table 2. L9 orthogonal array design for three factors at three levels.

No.	A	B	C	The ratio of MS/RH (w/w)	Pressing temperature (°C)	Pressing time (min)	Hardness (kgF)	Density (g/cm^3)
1	1	1	1	20/80	130	3	4.92	0.954
2	1	2	2	20/80	150	4	4.02	0.980
3	1	3	3	20/80	170	5	5.68	1.062
4	2	1	2	35/65	130	4	4.49	0.926
5	2	2	3	⇨ 35/65	150	5	4.44	0.951
6	2	3	1	35/65	170	3	4.55	0.971
7	3	1	3	50/50	130	5	3.58	0.879
8	3	2	1	50/50	150	3	5.72	0.923
9	3	3	2	50/50	170	4	3.55	0.926

Table 3. Response table for signal-to noise ratios and means of the hardness of the product.

Level	The ratio of starch and RH	Pressing temperature (°C)	Pressing time (mins)
Response table for signal to noise ratios larger-is-better			
1	13.67	12.65	14.05
2	13.05	13.40	12.04
3	12.41	13.09	13.04
Delta	1.26	0.74	2.01
Rank	2	3	1
Response table for means			
1	4.874	4.329	5.064
2	4.492	4.728	4.019
3	4.284	4.594	4.568
Delta	0.590	0.399	1.046
Rank	2	3	1

Density is a crucial physical property for the tray's practical use. In the response table for product density, the MS/RH ratio is the most influential factor. As the MS/RH ratio increases, the tray's density decreases. The pressing temperature and time have lesser effects on product density. The best results, combining low density with good incorporation of RH fibre in the polymeric matrix, were achieved with a 50/50 MS/RH ratio, a pressing temperature of 130°C, and a pressing time of 4 minutes. These conditions, however, are in contrast to those for optimal hardness. The better the tray in terms of lower density, the lower its hardness. An increased MS/RH ratio results in softer products (Fig. 5). The addition of more RH fibres makes the tray harder due to the dense network structure created by the hydroxyl groups on the fibres' surfaces [7]. It has been demonstrated that adding RH fibres can improve the thermal properties, tensile strength, and hydrophobicity of biodegradable trays [22]. Nonetheless, the density variations were small, ranging from 0.87 to 1.06 g/cm^3, and the product's hardness was prioritised when selecting the optimum starch-to-rice-husk ratio of 20/80 (w/w). A higher proportion of RH implies a longer biodegradation time since cellulose degrades more slowly than starch [8].

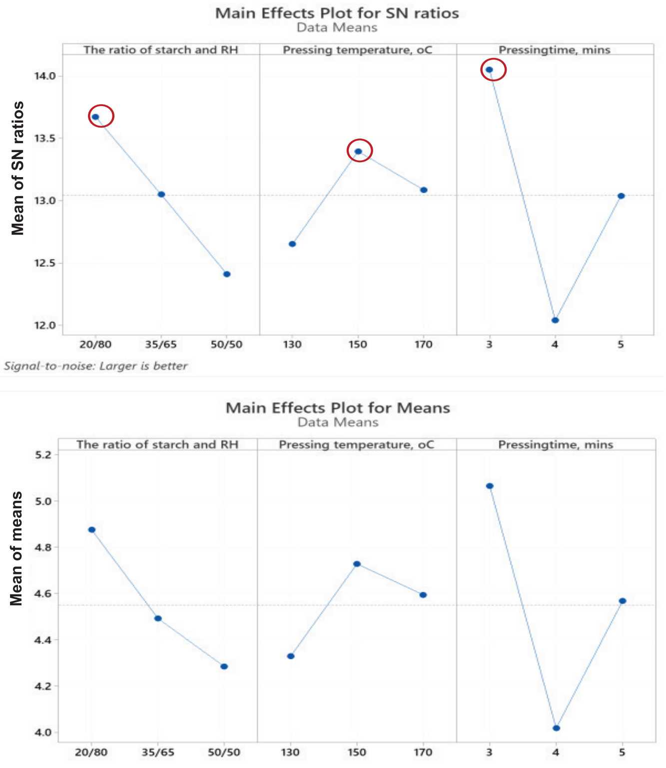

Fig. 5. Effect plots for the hardness of biodegradable trays.

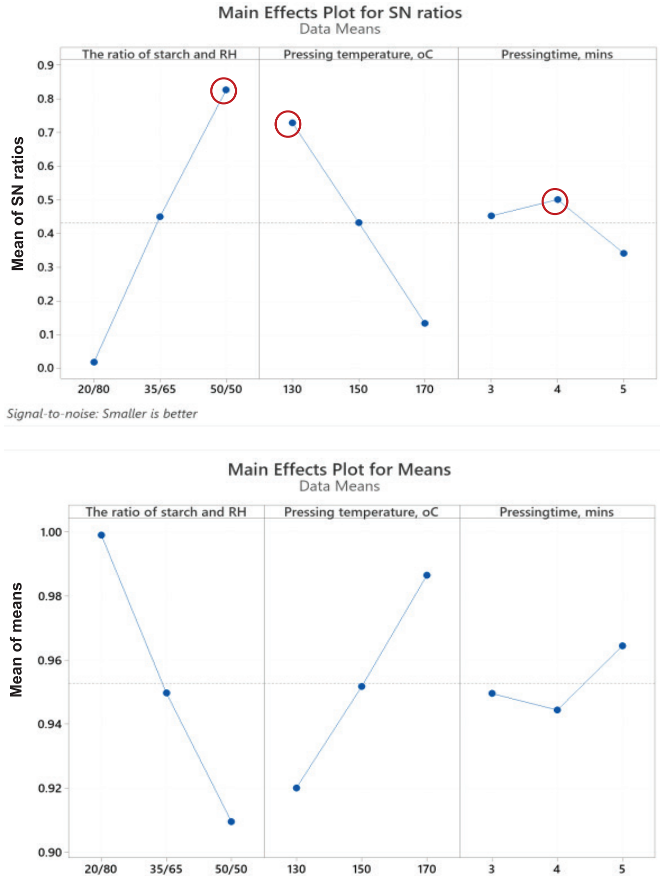

Fig. 6. Effect plots for the density of biodegradable trays.

The results for the biodegradable trays indicate that a pressing temperature of 150°C and a pressing time of 3 minutes yield the highest hardness (Fig. 6 and Table 4). Conversely, the most effective density is achieved at a pressing temperature of 130°C with a pressing time of 4 minutes. Nonetheless, this condition also results in the lowest hardness S/N values and means. While pressing temperature marginally affects hardness, it significantly impacts product density. Higher moulding temperatures, such as 170°C, enhance the crystallinity index of cellulose, reduce its hygroscopic nature, and create a stronger, less porous network resistant to moisture absorption. This is corroborated by N. Suderman, et al. (2016) [23], who noted the impact of drying temperature and time on the mechanical properties of biodegradable containers.

Table 4. Response table for signal-to-noise ratios and means for the density of the product.

Level	The ratio of MS and RH	Pressing temperature (°C)	Pressing time (mins)
Response table for signal to noise ratios (smaller is better)			
1	0.01796	0.72831	0.45216
2	0.44994	0.43263	0.50051
3	0.82599	0.13296	0.34123
Delta	0.80803	0.59535	0.15928
Rank	1	2	3
Response table for means			
1	0.9990	0.9201	0.9495
2	0.9497	0.9517	0.9443
3	0.9095	0.9864	0.9644
Delta	0.0894	0.0663	0.0200
Rank	1	2	3

Table 5. Response table for signal-to-noise ratios and means for the colour differences.

Level	The ratio of MS and RH	Pressing temperature (°C)	Pressing time (mins)
Response table for signal to noise ratios (smaller is better)			
1	-33.76	-33.57	-33.81
2	-33.78	-33.92	-32.73
3	-33.04	-33.10	-34.05
Delta	0.74	0.82	1.32
Rank	3	2	1
Response table for means			
1	48.70	47.46	48.83
2	48.62	49.68	43.06
3	44.99	45.18	50.43
Delta	3.71	4.50	7.37
Rank	3	2	1

The colour difference (ΔE) of a biodegradable tray, detailed in Table 5, shows that trays with the highest MS/RH ratio (20/80) exhibit the highest ΔE values. Conversely, a decrease in the MS/RH ratio leads to a significant reduction in ΔE as the RH content increases (Fig. 7). The addition of RH fibres to the starch matrix results in an increased ΔE value, giving the trays a brownish and yellowish hue, likely influenced by the natural colour of the RH fibres and their high lignin content [24]. Moreover, the high-temperature and pressure conditions cause the degradation of lignocellulosic materials, releasing compounds such as furfural and glycolaldehyde, which contribute to the brown colouration of the product.

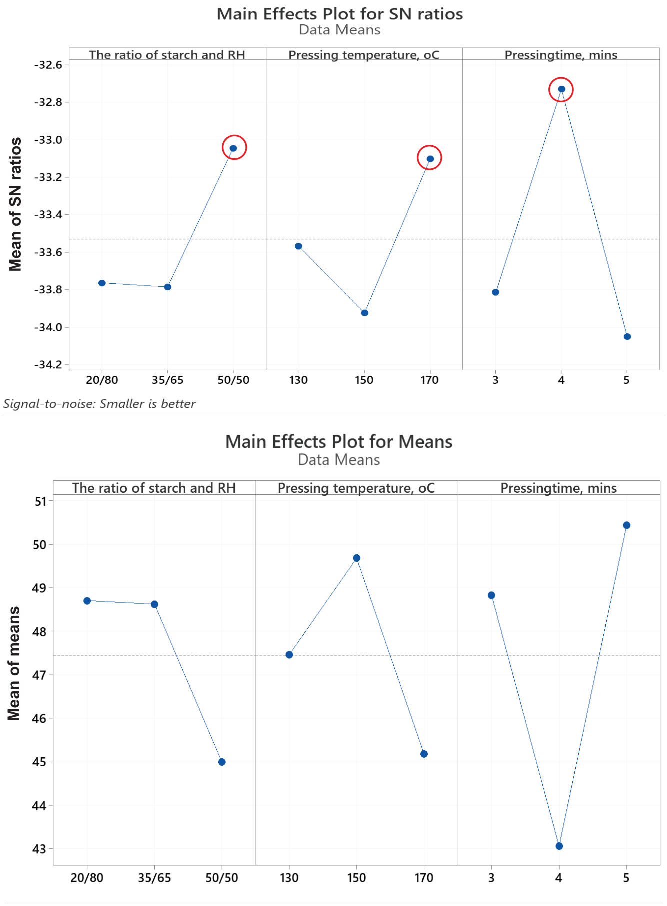

Fig. 7. Effect plots for the colour difference of biodegradable trays.

For optimal product quality, biodegradable trays were produced with an MS/RH ratio of 20/80, a pressing temperature of 150°C, and a pressing time of 3 minutes (Fig. 8). The colourimetry, hardness, and density were evaluated, with results showing hardness (5.26±0.22 kgF), frontside colour difference (50.87±0.96), backside colour difference (48.05±0.87), and density (0.97±0.004 g/cm³).

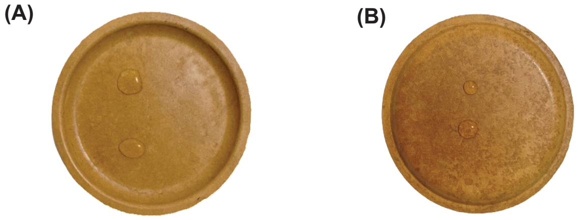

Fig. 8. Biodegradable trays based on RH and starch. (A) Frontside; **(B)** Backside.

4. Conclusions

The thermo-pressing process has been successful in manufacturing biodegradable trays from MS and RH fibre. The optimal trays are characterised by an appealing off-white brown colour, robust hardness, and low density. The ideal starch to RH ratio is 20/80, with a pressing temperature of 150°C and a pressing time of 3 minutes. The mechanical properties of the trays include hardness (5.26±0.22 kgF), frontside colour difference (50.87±0.96), backside colour difference (48.05±0.87), the thickness (2.05±0.01 mm) and density (0.97±0.004 g/cm^3). The Taguchi method identifies pressing time as the most significant factor affecting the mechanical properties (hardness and colour) of the product, with the starch/RH ratio being paramount for density criteria.

CRediT author statement

Nhung Thi-Tuyet Hoang: Conceptualisation, Resources and Data curation, Writing - Reviewing and Editing; Anh Thi-Kim Tran: Writing original draft preparation, Writing - Reviewing and Editing.

COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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