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Effect of *Parinari curatellifolia* Peel Flour on the Nutritional, Physical and Antioxidant Properties of Biscuits

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Abstract: This study investigated the impact of *Parinari curatellifolia* peel flour on the nutritional, physical and antioxidant properties of formulated biscuits. Biscuits enriched with 5%, 10%, 15% and 20% of *Parinari* (*P. curatellifolia*) peel flour were formulated and characterised. Thermal, physico-chemical, polyphenolic compounds and antioxidant properties of flour and biscuits were determined. The incorporation of *P. curatellifolia* peel flour significantly increased ($p < 0.05$) thermal properties (onset, peak and conclusion temperatures) of flour. However, enthalpy of gelatinisation, viscosity and pH of flour samples decreased. Nutritional analysis revealed an increase in ash (0.74% to 2.23%) and crude fibre contents (0.39% to 2.95%) along with an increase of *P. curatellifolia* peel flour levels. Protein content and carbohydrates decreased while moisture content was insignificantly affected by the addition of *P. curatellifolia* peel flour. The L^* , b^* values and whiteness index of formulated biscuits decreased while parameter a^* value (10.76 to 21.89) and yellowness index (69.84 to 102.71) decreased. Physical properties such as diameter (3.57 mm to 3.97 mm), spread ratio (2.67 to 3.45) and hardness (1188.13 g to 2432.60 g) increased with the inclusion levels of peel flour while weight and thickness decreased. The inclusion of *P. curatellifolia* improved the polyphenolic compounds and antioxidant activity of biscuits with values of total flavonoids content ranging from 0.028 to 0.104 mg CE/g, total phenolic content increasing from 20.01 mg to 48.51 mg GAE/g, ferric reducing antioxidant power (FRAP) increasing from 108.33 mg to 162.67 mg GAE/g and DPPH (2,2-diphenyl-1-picrylhydrazyl) from 48.70% to 94.72%. These results lead to the recommendation of the utilisation of *P. curatellifolia* peel flour to enhance the nutritional value, polyphenolic compounds and antioxidant activity of bakery products such as biscuits.

Keywords: composite flour; proximate composition; polyphenolic compounds; antioxidant activity; textural properties

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1. Introduction

Improved living standards, as well as lifestyles, have resulted in consumers not just preferring food which meets their daily nutrient requirements but food with nutraceutical and functional characteristics [1,2]. Therefore, the baking industry should produce products added with bioactive components such as phenolic acid, flavonoids, dietary fibre, protein, vitamins and minerals [3,4]. Biscuits are popular bakery products consumed worldwide as a ready-to-eat snack with different appealing properties such as a wide consumption base, reasonable cost and convenience due to shelf-life stability [5]. Therefore, biscuits can supply essential nutrients. Moreover, there is a shift towards producing functional biscuits prepared from wheat flour and bioactive components from plant-based materials [6].

Wild fruits are important because of their use as food or medicines and their potential for generating income when processed into alcoholic drinks and juices [7]. *Parinari curatellifolia* is an indigenous tree bearing fruits found in most parts of southern Africa [8]. It is known by various names such as *mobola* in Sepedi, *grysappel* in Afrikaans, *muvhula* in

Venda, *umkhuna* in Ndebele, *muchacha* and *muhacha* in Shona, *mbura* in Swahili and *mula* in Tongan [9]. It belongs to the family *Chrysobalanaceae*, found over a great range of places such as South Africa, Malawi, Zimbabwe, Botswana and Nigeria. It is found in the forest along streams and enduring alone in areas of cleared up woodland [10]. The fruit resembles plums, and its peel and edible flesh are yellow with grey speckles when ripened and is ± 50 mm long with yellow edible flesh. The fruit has a sweet taste when fully ripened, and it is consumed without a peel [7]. The utilisation of *P. curatellifolia* peels might improve the yield of raw materials and eventually reduce the large waste disposal problems faced by the food industry [11]. Therefore, the objective should be recovery of the peels through technological processes to obtain natural-derived functional products.

Parinari curatellifolia (*P. curatellifolia*) fruit is rich in carbohydrates (84.95%), dietary fibre (4.71%), protein (3.90%) and ash (2.46%) [7]. The fruit is usually eaten fresh as a snack or dried into powder which is added to products such as beverages, porridge and fritters for feeding young children [12]. In view of the above nutrients, *P. curatellifolia* can be used as a functional ingredient in bakery products for nutritional improvement and fortification. This is because consumers nowadays are interested in food with high nutritional properties such as dietary fibre, antioxidants, vitamins and polyphenolic compounds. Moreover, these health-promoting compounds aid in the maintenance of health and prevention of diseases such as cancer, cardiovascular and other chronic diseases [13].

The fruit peels are generally discarded, used for livestock feeding or to improve soils. The correct usage of fruit peels might reduce the problem of waste disposal and become a source of polyphenolic compounds and antioxidants [14]. In addition, fruit peels are rich in bioactive components such as phenolic acid, flavonoids, antioxidants vitamins and have nutraceuticals properties [15]. Different authors have partially substituted wheat flour with fruit peel flour to produce bakery products such as bread and biscuits due to new consumption styles and trends, for economic reasons and as required by businesses [16–18]. For example, prickly pear and potato peels improved the phenolic compounds, antioxidant activity and dietary fibre of crackers [19]. The incorporation of guava peel flour revealed a high amount of total polyphenols and β -carotene content in biscuits, and it also affected colour, flavour, texture and appearance parameters [20]. Furthermore, biscuits samples enriched with banana peel and prickly pear peels improved crude fibre, phenolic compounds and flavonoids content. The diameter and spread ratio of the biscuits increased with a decrease in height [21]. Therefore, enriching biscuits with peel flour from *P. curatellifolia* fruit fits the needs of health-conscious consumers.

The utilisation of *P. curatellifolia* fruit peels in biscuits requires research because of the growing demand for bakery products with improved nutritional composition. In this regard, utilisation of peels such as of *P. curatellifolia* fruit as a functional ingredient in composite flour is one of the novel ways to modify the nutritional as well as quality properties of the biscuits. Most composite biscuits are produced by combining various flours of cereals and legume or root crops which improves the functional properties and nutrients composition [22]. Moreover, there is a scarcity of relevant information about the utilisation of *P. curatellifolia* fruit peels as a functional ingredient for biscuits. Therefore, this study utilised *P. curatellifolia* peel flour at different ratios in biscuits-making and determined its influence on the nutritional, physical and antioxidant properties in order to evaluate its usefulness as a functional ingredient.

2. Materials and Methods

2.1. Preparation of *P. curatellifolia* Peel Flour

The *P. curatellifolia* fruits at their ripening stage were harvested from the University of Venda's experimental farm in Thohoyandou, South Africa. The selected fruits were washed with clean tap water and the peels were separated from the pulp using a stainless-steel knife. Afterwards, the peels were oven-dried for 4 h at 60 °C and ground into fine flour using a miller (Retsh ZM 200 miller, Haan, Germany). The flour was then passed through

250 μm sieve mesh to standardise the particle size and packaged in an airtight polyethylene plastic bag and stored at 4 °C for subsequent use in biscuit formulations.

2.2. Preparation of Biscuits

The biscuits were prepared with different ratios of *P. curatellifolia* peel flour. The control biscuits were prepared from 100 g of wheat flour, 120 g of powdered sugar, 250 g of margarine (fat), 10 g of liquid milk, 4 g of baking powder and 1 g of salt and vanilla essence. The *P. curatellifolia* flour was incorporated into the baking mixture at levels of 5, 10, 15 and 20% based on the wheat flour weight. Ingredients used to produce biscuits were manually mixed for 5 min. Gauge strip was used to roll the dough with moisture content between 55 and 60% to the correct thickness, poured into greased pans and baked for about 150 °C for 20 min in an electric stove (Defy Kitchenaise 621, Midrand, South Africa). The formulated baked biscuits (Figure 1) were cooled at room temperature (25 °C) for 30 min and packed in an airtight plastic container for subsequent analysis.

Figure 1. Biscuits formulated using different levels of wheat flour replacement with *P. curatellifolia* fruit peel flour. Control (100% wheat biscuit), BPC1 (5%), BPC2 (10%), BPC3 (15%) and BPC4 (20%) *P. curatellifolia* peel flour.

2.3. Thermal Properties of Wheat–*P. curatellifolia* Composite Flours

The gelatinisation temperature of composite flour was determined through Differential Scanning Calorimetry (DSC, DSC 4000, Perkin-Elmer, Shelton, CT, USA). Indium was used to calibrate the instrument. Dry flours of 4 mg were weighed into aluminium pans and a micro-syringe was used to add distilled water in order to obtain the starch-water suspension. The wheat–*P. curatellifolia* peel flour was hermetically sealed and incubated for 1 h at 25 °C and humidity of 35 to 50% before heating. The rate of 10 °C/min was used to heat the pan from 20 to 125 °C. An empty pan (Perkin-Elmer, Shelton, CT, USA) was used as a control, thermal analyses (onset, end set, peak temperature and enthalpy of gelatinisation) were carried out using the software provided with the equipment. Onset temperature (T_O), peak temperature (T_P), conclusion temperature (T_C) and enthalpy of gelatinisation (ΔH_{gel}) were automatically calculated [23].

2.4. Viscosity of the Composite Flour (Cold and Hot) Pastes

About ten (10) g of composite flour was added to 90 mL of distilled water at 30 °C and allowed to moisten for 30 min with intermittent stirring. The Brookfield viscometer (Model RV, Middleboro, MA, USA) was used to measure the viscosity of the cold paste in cP using spindle number 03 rotating expressed as 54 g. Afterwards, the cold paste was heated to boiling in a water bath for 20 min at 95 ± 1 °C, cooled to 30 °C and the viscosity of hot paste was also measured in cP. The paste temperature was kept constant by monitoring temperature using thermometer until it reaches 75 °C.

2.5. pH of the Composite Flours

The pH values of wheat-*P. curatellifolia* peel flours were measured using a Crison digital pH meter (Crison Instrument, SA, Midrand, South Africa). Before using the pH, it was calibrated at a controlled temperature ranging from 20 to 25 °C at a relative humidity of 35% to 50% with three (3) different buffers pH meter 4, 7 and 9. The condition of the electrode was checked and cleaned before calibration. Thirty millilitres of each buffer pH 4, 7 and 9 was poured into a clean, dry beaker and the pH meter probe was immersed in the buffer solution. The pH meter could read the buffer, and a stable reading was recorded. The meter was automatically stopped as soon as the reading was stable, and then the pH reading was recorded. The electrode was removed from the beaker and rinsed with distilled water for further use. About ten (10) grams of flour was added in a beaker with 100 mL distilled and deionised water and stirred for 15 min to blend the flour. The resulted suspension was allowed to rest for 15 min, the pH level was read in the suspension liquid and the readings were taken in triplicates.

2.6. Colour Attributes of Composite Flours and Biscuits

Hunter Lab (Hunter Lab, Mini Scan XE Plus and Reston, VA, USA) was used to measure the colour attributes of composite flour biscuits using an illuminant D65. Differences in colour were measured in CIE $L^*a^*b^*$ scale with regard to lightness (L^*) and colour (a^* —redness; b^* —yellowness). The hue angle (H°), chroma (C^*), the total colour difference (ΔE), whiteness (WI) and yellowness (YI) indexes were calculated using the following equations:

$$\text{Hue } (H^\circ) = \tan^{-1} \left\{ \frac{b^*}{a^*} \right\}$$

$$\text{Chroma} = \sqrt{(a^*)^2 + (b^*)^2}$$

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

$$\text{YI} = \frac{142.86b^*}{L^*}$$

$$\text{WI} = \sqrt{(100 - L^*)^2 + (a^*)^2 + (b^*)^2}$$

2.7. Proximate Composition of Biscuits

The proximate composition was determined using AOAC [24] in which the moisture content was measured using an oven-drier method (945.32), crude protein using the Kjeldahl method (978.02), fat content using Soxhlet extraction method (945.38), ash content using muffle furnace method (923.03) and crude fibre was determined by fibre-tech method (985.33). The carbohydrate content was measured by subtracting the total ash, fat, fibre, protein and moisture content from 100%, and total energy value was calculated by multiplying carbohydrate and protein contents by 4 and fat content by 9.

2.8. Polyphenolic Compounds and Antioxidant Activities of Composite Biscuits

2.8.1. Extraction

The extract was obtained as described by Moussa-Ayoub et al. [25], whereby 100 mg of biscuits sample was added to 50 mL of water in a test tube which was kept in the dark for 10 min with occasional stirring. The solution was centrifuged (Zentrifuge 380 R, Labotec, Midrand, South Africa) at $2086 \times g$ for 10 min at 4 °C and the suspension was filtered by a 0.20 µm pore size membrane filter (Millipore® Burlington, MA, USA) and stored at 4 °C. The *P. curatellifolia* extract was used for the subsequent determinations of polyphenolic compounds and antioxidant activity.

2.8.2. Total Phenolic Compounds (TPC)

The TPC of the extract was determined using the slightly modified method of Kapcum et al. [26], where 0.5 mL of the biscuit sample extracts were poured into test tube, 1.5 mL of Folin–Ciocalteu reagent (1:2 v/v) was mixed with the extract and the mixture was allowed to rest for 5 min at laboratory temperature. After 5 min, 2 mL of 7% sodium carbonate was added and incubated for 45 min in the dark and a blue colour developed. Distilled water (10 mL) was used to dilute the colour since it was too deep. The absorbance was read with a UV-Visible spectrophotometer (Zenyth 200rt Biochrom, Cambridge, UK) at 725 nm. TPC was expressed as milligram gallic acid equivalent per gram (mg GAE/g) of extract and the calibration curve for gallic acid was obtained as $R^2 = 0.999$.

2.8.3. Total Flavonoids Compounds (TFC)

The method of Shen et al. [27] was used to determine the TFC of biscuits, whereby 0.3 mL of extracts was poured into a test tube and 3.4 mL of 30% methanol was added, followed by 0.15 mL of NaNO_2 (0.5 M). Approximately 0.15 mL (10% w/v) AlCl_3 was added after 5 min, followed by 1 mL of 1 M NaOH after 6 min. Distilled water was used to make the volume up to 10 mL and vortexed, and a UV spectrophotometer microplate reader (Zenyth 200rt Biochrom, Cambridge, UK) was used to read the absorbance of the mixture at 506 nm. TFC was expressed as milligram catechin equivalent per gram (mg CE/g), and the calibration curve for catechin was obtained as $R^2 = 0.998$.

2.8.4. DPPH (2,2-Diphenyl-1-picryl-hydrazyl) Free Radical Scavenging Activity

The antioxidant capacity (DPPH Assay) of biscuit samples was measured following the method described by Nsabimana et al. [28]. Each sample of 2 mL was mixed with 2 mL of 0.1 mM DPPH in 95% ethanol. The mixture was vigorously shaken and allowed to rest for 30 min under subdued light at 25 °C. The absorbance of the mixture was measured by UV spectrophotometer microplate reader (Zenyth 200rt Biochrom, Cambridge, UK) at 517 nm. A gallic acid solution was used to prepare a standard curve, and results were expressed as percentage inhibition of the DPPH radical. The equation for calibration curve was: $y = 3.6574x + 0.0363$; $R^2 = 0.9986$.

2.8.5. Ferric-Reducing Antioxidant Power (FRAP)

The reducing power assay was determined using the method of Radenkors et al. [29]. Briefly, 100 µL of the extract was added to a test tube, methanol 2.5 mL 0.2 M phosphate buffer (pH 6.6) was used to make the volume to 1 mL and 2.5 mL 1% potassium ferricyanide was added to the tube and vortexed. The mixture was incubated in a water bath (WBH 601 Labcon, Krugersdorp, South Africa) for 20 min at 50 °C. Afterwards, the mixture was added with 2.5 mL of 10% (w/v) trichloroacetic acid and centrifuged (Zentrifuge 380 R, Labotec, Midrand, South Africa) for 20 min at 5000 rpm. About 2.5 mL distilled water and 0.5 mL 0.1% (w/v) ferric chloride were added to 2.5 mL supernatant in a test tube, and UV spectrophotometer microplate reader (Zenyth 200rt Biochrom, Cambridge, UK) was used to measure the absorbance of the mixture at 700 nm. The standard curve was produced using 50 g gallic acid dissolving in 2 mL ethanol and diluting the mixture with

1 L of distilled water. Results were expressed in mg gallic acid equivalents (GAE) per gram of sample.

2.9. Physical Properties of the Composite Biscuits

2.9.1. Thickness, Weight, Diameter and Spread Ratio

Biscuits were analysed for thickness, weight and diameter and spread ratio. The weight of the biscuits was measured by a digital weighing balance [30], while a Vernier calliper was used to measure the diameter and thickness. The spread ratio was calculated by dividing diameter with thickness using the formula (W/T), where W was the diameter, and T was the thickness of the biscuit. All measurements were carried out in triplicates.

2.9.2. Texture Measurement

TA-XT Plus Texture Analyzer (Stable Micro System Ltd., Surrey, UK), a cylindrical probe P/2 fitted with a 5 kg cell load was used to measure the hardness of the biscuits. The settings of TA consisted of: test mode, compression; pre-test speed, 1.00 mm/s; test speed, 0.50 mm/s; post-test speed, 10.0 mm/s; distance, 2.00 mm; trigger force, auto, 5 g.

2.10. Statistics

Mean value standard deviations are recorded in the tables, and all measurements were completed in triplicate. An IBM SPSS Statistics computer program (version 26) was used to statistically evaluate the results. One-way ANOVA was used to determine the significant effect of different levels of *P. curatellifolia* by evaluating the statistical significance at the level of $p < 0.05$. Duncan's multiple range tests were applied to identify any significant differences among the samples at a 95% confidence level ($p \leq 0.05$).

3. Results and Discussion

3.1. Thermal Properties of Composite Flours

Differential scanning calorimetry measures the minimum amount of energy required to destruct the starch order available in a food product. Granule type and starch concentration are some of the factors that influence starch [31]. The thermal properties of flour samples are presented in Table 1. Inclusion of *P. curatellifolia* peel flour increased the onset temperature (T_o) with values ranging from 55.28 to 56.77 °C and peak temperature (T_p) from 61.62 to 62.73 °C and conclusion temperature (T_c) from 68.70 to 68.57 °C. The control and composite flours were significantly different ($p < 0.05$) in terms of onset, peak and conclusion temperatures. The high values in composite flours might be attributed to more crystallisation which influenced the structural strength by forcing the starch granules to resist gelatinisation [32,33]. Moreover, high gelatinisation temperatures are related to low amylose content. Competition for water utilisation between starch and other components such as dietary fibre in the composite flours likely contributed to variations of results in onset and peak temperatures. In addition, other parameters such as starch purity, the nature of association within the amorphous and crystallisation region [34] also played a part. Moreover, the distribution of amylopectin chains inside starch granules might also affect the gelatinisation properties of starch [35]. On the other hand, low values of gelatinisation temperatures of the control sample might be due to amylose-lipid complexes since they require a higher onset temperature to melt. The values of gelatinisation temperatures of composite flours are within the range reported by other authors [34,36].

There was a significant difference ($p < 0.05$) between control and composite flours with values of enthalpy gelatinisation ranging from 5.66 to 4.48 J/g. The low values of enthalpy gelatinisation might be attributed to various structures and particle size of fibres, and the amount of water used in composite flours. Fibre might interchange with starch to facilitate the generation of a more coherent structure, thereby decreasing enthalpy gelatinisation. The low values obtained mean that less thermal energy is required to gelatinise starch in composite flours [37]. Moreover, variation in enthalpy gelatinisation values might be due

to differentiation in the degree of association between the double helices that form the crystallisation region of the composite flours [38].

Table 1. Thermal properties of wheat–*P. curatellifolia* composite flours.

Sample	T ₀ (°C)	T _p (°C)	T _c (°C)	ΔH (J/g)
Control	55.28 ± 0.63 ^a	61.62 ± 0.21 ^a	68.70 ± 0.79 ^a	5.66 ± 0.07 ^d
BPC1	56.33 ± 0.87 ^b	62.27 ± 0.36 ^b	68.80 ± 0.62 ^b	4.65 ± 0.70 ^c
BPC2	56.38 ± 0.18 ^b	62.54 ± 0.05 ^c	68.83 ± 0.43 ^b	4.63 ± 0.18 ^c
BPC3	56.56 ± 0.55 ^c	62.61 ± 0.40 ^d	69.36 ± 0.78 ^c	4.52 ± 0.07 ^b
BPC4	56.77 ± 0.35 ^d	62.73 ± 0.14 ^e	69.57 ± 0.52 ^d	4.45 ± 0.17 ^a

Results are expressed as mean ± standard deviation. Different superscripted letters within columns are significantly different at $p < 0.05$. T₀ = onset temperature. T_p = peak temperature. T_c = conclusion temperature. ΔH = enthalpy of gelatinisation. Control (100% wheat biscuit), BPC1 (5%), BPC2 (10%), BPC3 (15%) and BPC4 (20%) *P. curatellifolia* peel flour.

3.2. Viscosity and pH of Composite Flours

Table 2 shows results for cold and hot paste viscosity of control and composite flours. The inclusion of *P. curatellifolia* peel flour reduced the viscosity of cold and hot paste, with results ranging from 24.00 to 30.00 cP and 241.67 to 321.00 cP. There was a significant difference ($p < 0.05$) between control and composite flours with regard to cold and hot paste viscosities. Low values of viscosity in composite flours might be due to protein which protects starch granules from swelling and breaking down [39].

Table 2. Viscosity and pH analysis of composite flours.

Sample	Viscosities (cP)		pH
	Cold Paste	Hot Paste	
Control	30.00 ± 1.00 ^b	282.67 ± 3.12 ^c	6.09 ± 0.13 ^c
BPC1	24.00 ± 1.03 ^a	258.33 ± 2.08 ^b	5.71 ± 0.06 ^b
BPC2	25.00 ± 1.10 ^a	251.67 ± 2.08 ^a	5.48 ± 0.03 ^a
BPC3	24.00 ± 1.09 ^a	240.33 ± 1.53 ^c	5.49 ± 0.02 ^a
BPC4	24.00 ± 1.00 ^a	229.00 ± 3.61 ^d	5.45 ± 0.09 ^a

Results are expressed as mean ± standard deviation. Different superscripted letters within columns are significantly different at $p < 0.05$. Control (100% wheat flour), BPC1 (5%), BPC2 (10%), BPC3 (15%) and BPC4 (20%) *P. curatellifolia* peel flour.

Usman et al. [40] indicated that carbohydrates reduce viscosity given that they have a low water-absorption capacity and might be easily digested and absorbed as desired by infants. Therefore, composite flours can be beneficial in the process of weaning infants. The results for pH of flours ranged from 5.45 to 6.09 and showed a significant decrease ($p < 0.05$).

The pH values of all composite flours were <4.5, and flour samples are therefore low-acid food. pH is an important factor since it influences the flavour of biscuits [41]. The results for pH obtained from this study imply that the composite flours will have a more stable shelf life than the control sample. Similar results were reported by Ogunjobi and Ogunwolu [41], whereby the inclusion of cashew apple powder decreased the pH value of cassava flour.

3.3. Colour Attributes of the Composite Flours and Biscuits

Colour is an essential quality characteristic of flour since it influences appearance and consumer acceptance of products [42]. Colour attributes of composite flour blends and biscuits are depicted in Table 3. The colour of flour blends as well as their biscuits is determined by the amount of natural pigments, proteins, fibres and the presence of impurities [43]. From the five flour samples, the control sample was lighter ($L^* = 89.68$) than composite flours. The L^* values of flour samples ranged from 72.13 to 89.68, and the control sample had a higher value. The degree of lightness of composite flours decreased

with the addition of *P. curatellifolia* peel flour. This might be attributed to natural pigments of *P. curatellifolia* peels and thermal processes that the flour went through, such as drying and milling. The higher L* value of the control sample might be due to the removal of bran or endosperm during the milling of wheat [42].

Table 3. Colour attributes for composite flours and biscuits.

Flour	L*	a*	b*	C	H°	ΔE	WI	YI
Control	89.68 ± 0.01 ^e	0.67 ± 0.02 ^a	09.87 ± 0.01 ^a	9.51 ± 0.01 ^a	85.92 ± 0.03 ^e	-	25.94 ± 0.25 ^e	08.88 ± 0.01 ^a
BPC1	78.40 ± 0.03 ^d	3.68 ± 0.02 ^b	10.26 ± 0.01 ^b	10.90 ± 0.00 ^b	70.74 ± 0.05 ^d	77.12 ± 0.02 ^d	25.50 ± 0.12 ^d	08.94 ± 0.00 ^b
BPC2	75.63 ± 0.21 ^c	4.21 ± 0.02 ^c	10.60 ± 0.04 ^c	11.41 ± 0.03 ^c	68.34 ± 0.11 ^c	74.23 ± 0.03 ^c	22.42 ± 0.36 ^c	08.99 ± 0.03 ^c
BPC3	72.41 ± 0.01 ^b	5.23 ± 0.02 ^d	11.81 ± 0.03 ^d	12.92 ± 0.03 ^d	66.35 ± 0.05 ^b	70.68 ± 0.00 ^b	21.04 ± 0.32 ^b	09.20 ± 0.00 ^d
BPC4	72.13 ± 0.01 ^a	5.42 ± 0.02 ^e	12.39 ± 0.02 ^e	13.52 ± 0.02 ^e	66.10 ± 0.07 ^a	70.29 ± 0.01 ^a	16.95 ± 0.03 ^a	09.98 ± 0.00 ^e
Biscuits								
Control	42.98 ± 0.50 ^e	10.76 ± 0.69 ^a	24.77 ± 1.13 ^e	28.69 ± 0.63 ^e	59.71 ± 1.97 ^e	-	50.09 ± 3.19 ^e	69.84 ± 3.31 ^a
BPC1	41.25 ± 3.22 ^d	11.34 ± 0.62 ^b	23.37 ± 0.19 ^d	25.79 ± 0.73 ^c	58.12 ± 1.02 ^d	25.41 ± 0.89 ^c	48.24 ± 0.54 ^d	73.73 ± 4.12 ^b
BPC2	32.57 ± 1.73 ^c	13.62 ± 0.67 ^c	15.47 ± 1.20 ^c	27.85 ± 0.42 ^d	57.10 ± 1.78 ^c	24.94 ± 1.18 ^b	41.67 ± 1.63 ^c	74.51 ± 1.29 ^c
BPC3	29.97 ± 1.70 ^b	15.13 ± 0.94 ^d	14.45 ± 0.49 ^b	17.87 ± 1.59 ^b	52.90 ± 1.39 ^b	15.71 ± 1.44 ^a	32.68 ± 2.39 ^b	83.87 ± 2.62 ^d
BPC4	29.13 ± 1.65 ^a	21.89 ± 0.39 ^e	14.27 ± 1.50 ^a	19.18 ± 1.33 ^a	53.72 ± 0.64 ^a	15.66 ± 0.53 ^a	34.15 ± 2.04 ^a	102.71 ± 6.12 ^e

Results are expressed as mean ± standard deviation. Different superscripted letters within columns are significantly different at $p < 0.05$. Control (100% wheat flour), BPC1 (5%), BPC2 (10%), BPC3 (15%) and BPC4 (20%) *P. curatellifolia* peel flour. YI = yellowness index, WI = whiteness index. The yellowness index (YI) of the flours ranged from 16.95 to 25.94, and there was a significant increase ($p < 0.05$) in the yellowness index of the composite flours with the inclusion of *P. curatellifolia* peel flour. This was due to natural pigmentation of the fruit peels.

The addition of *P. curatellifolia* peel flour significantly increased ($p < 0.05$) the redness (a*) and yellowness (b*) of the flour with values ranging from 0.67 to 5.42 and 09.87 to 12.39. This was likely caused by the long exposure to thermal treatment (drying), which could have added a darker colour to the peels. The yellowness values, on the other hand, are likely due to natural pigment carotenoids found in *P. curatellifolia* peels [44]. Moreover, the higher values of redness and yellowness are an indication that the composite flours had more appealing colours as well as various amounts of red and yellow pigments than the control sample [45]. Therefore, the incorporation of *P. curatellifolia* peel flour into wheat flour improves the colour of baked products [42]. Weng et al. [16] obtained similar results after including passion fruit peel flour, which had a positive impact on a* and b* values and decreased the L* values of wheat flour.

The colour of composite flours was more concentrated than that of the control sample, as shown by its high chroma values, which ranged from 9.51 to 13.52. An increase in chroma is always associated with the concentration of pigment, while a decrease in the lightness index is linked with high chroma values [42,45], and the results of this study corroborate this claim. The high colour saturation was observed in the control sample (85.92) while sample BPC4 (66.10) showed the least H°. The total colour difference (ΔE) results showed a significant decrease with an increase in *P. curatellifolia* peel flour; the results varied from 77.12 to 70.29 for composite flours. As more peel flour was added, there was an increase of ΔE based on the categories of differences in observable colour [46]. The colour of composite flours was very distinct from one another.

The whiteness index (WI) of composite flours was significantly different ($p < 0.05$) from the control sample, with results ranging from 08.88 to 09.98. Furthermore, the incorporation of *P. curatellifolia* peel flour at all levels affected the WI index. The decreasing trends are due to the yellowish colour of the peels.

The biscuits incorporated with *P. curatellifolia* peel flour had lower L* values ranging between 29.13 and 42.25 compared to the control, with 42.98. The low L* values show that the composite biscuits were darker in colour with the increase of peel flours. This might be due to the development of the brown colour of *P. curatellifolia* peel flour because of thermal treatment during drying, Maillard reaction and caramelisation of the sugars and the unequal subjection of the surface area of biscuits to heat during the baking process [21]. Borrelli et al. [47] indicated that carbohydrates (glucose) and proteins are the chief components that contribute to browning reactions and influence the sensory properties of baked products. Furthermore, the colour of biscuits depends on the level of sugar added to the dough since it influences the initiation of Maillard reactions during baking.

The a^* value of biscuits displayed a significant ($p < 0.05$) increase as levels of *P. curatellifolia* peel flour increased, suggesting that protein content was adversely related to the lightness of biscuits. This shows that the Maillard reaction played a significant role in the generation of colour. The Maillard reaction and caramelisation of sugar are likely responsible for the generation of brown colour during baking [48].

Moreover, b^* values of the biscuits were significantly different ($p < 0.05$) across each other and it was observed that the yellowness (b^*) showed a significant increase with an increased level of peel flour. The results for yellowness ranged from 14.27 to 24.77, and similar results were observed by Ho and Abdul Latif [49], whereby the incorporation of pitaya fruit peel flour increased the b^* value of biscuits from 17.08 to 28.83.

The chroma (C^*) values of biscuits varied from 19.18 to 25.79, and there was a significant increase ($p < 0.05$) with the addition of *P. curatellifolia* peel flour. This result could be elucidated by both C^* and H° depending on a^* and b^* , while values for H° (colour saturation) ranged from 53.72 to 59.71 with the control having the highest H° and sample E showing the least H° value. There was an increasing trend as the peel flour increased in biscuit formulations. The control biscuit had a pure yellow colour (90°), while the composite biscuits had H° that inclined toward pure red (0°). Similarly, Mahloko et al. [21] reported an increase in H° angle values.

The total colour difference (ΔE) of the biscuit samples showed a significant decrease ($p < 0.05$), and there was a low trend in the colour of biscuits as more *P. curatellifolia* peel flour was added to wheat flour. Sample BPC1 had a maximum value of 25.41, while BPC4 had a minimum value of 15.66. The variations in ΔE might be attributed to flours' and biscuits' exposure to several thermal technological processes such as drying, milling, sieving as well as baking [44]. Zouari et al. [50] reported similar results after sesame peel flour decreased the ΔE of the biscuits.

The whiteness index (WI) and yellowness index (YI) had a negative relationship. However, the coefficients of association between whiteness and yellowness index relied on hue, particularly yellowness–blueness. The results for WI and YI ranged from 34.15 to 50.09 and 69.84 to 102.71. The WI showed a significant decrease as the level of *P. curatellifolia* peel flour increased. The WI shows the degree of discolouration during the drying process and is associated with the general degradation of food products by either light, chemical exposure or processing [51]. A significant increase was observed in the YI of the biscuits with the increase of the level of peel flour. This could be due to the addition of other baking ingredients such as shortening, which impart the characteristic yellow colour, as well as the Maillard reaction that occurs as a result of reducing sugars, heat and amino acids after baking [21].

The inclusion of *P. curatellifolia* peel flour positively influenced the a^* , b^* , C and YI of the composite flour and biscuits. These increases were at a higher level in biscuits than in flours and are attributable to the addition of the leavening agent, that likely increased parameters such as the YI and chroma as well as baking time. High temperatures possibly caused a significant increase to a^* and b^* values of the biscuits. Additionally, low b^* values of composite biscuits than that of flour blends can also be attributed to the breakdown of unstable yellow compounds during baking [49].

There was a significant decrease in the L^* , H° , ΔE and WI of flours and biscuits. There was a decreasing trend in L^* for biscuits compared to the composite flours. The darkening in biscuits could be attributed to thermal treatment during the baking of biscuits which degraded wheat flour's white colour. The colour change of composite biscuits from light grey to dark yellow might be attributed to an increase in the amount of *P. curatellifolia* peel flour. The dark colour of composite biscuits was also reported by Obafaye and Omoba [14] after incorporating orange peel powder.

3.4. Nutritional Composition of Biscuits

Table 4 shows the nutritional composition of control and composite biscuits. The moisture content of the biscuits varied from 3.93 to 4.01%, and there was no significant

difference ($p < 0.05$) between composite biscuits and the control sample. However, sample BPC3 had a slight increase in moisture content, and this might be due to the percentage of moisture intake during cooling of the biscuits [52]. However, the value was still less than 10%, and this invigorates longer shelf life as reported by Obafaye and Omoba [15]. The values of moisture content of all biscuit samples were within the prescribed limits ($<10\%$). Reduced moisture content is beneficial to the shelf life of biscuits because most of the unwanted microbes might struggle to survive [53]. Moreover, the shelf stability of bakery products has a direct relationship with the moisture content. Similar results were observed by Bertagnolli et al. [20] for biscuits incorporating guava peel flour with values ranging from 2.7 to 4.9%.

Table 4. Proximate composition of biscuits in 100 g/g.

Samples	Moisture	Ash	Fat	Crude protein	Crude Fibre	Carbohydrates	Energy (kcal)
Control	3.95 ± 0.00 ^a	0.74 ± 0.12 ^a	23.52 ± 0.37 ^d	2.46 ± 0.11 ^c	0.39 ± 0.08 ^a	68.94 ± 0.48 ^d	497.29 ± 0.00 ^d
BPC1	3.93 ± 0.02 ^a	1.38 ± 0.11 ^c	23.12 ± 0.49 ^c	2.32 ± 0.05 ^b	1.23 ± 0.13 ^b	68.12 ± 0.55 ^c	489.49 ± 0.03 ^c
BPC2	4.01 ± 0.00 ^a	1.25 ± 0.05 ^b	23.03 ± 0.69 ^c	2.24 ± 0.06 ^{ab}	1.21 ± 0.37 ^b	68.10 ± 0.99 ^{bc}	490.88 ± 0.01 ^c
BPC3	3.95 ± 0.01 ^a	1.30 ± 0.22 ^c	22.84 ± 0.12 ^b	2.17 ± 0.01 ^a	1.72 ± 0.27 ^c	67.98 ± 0.41 ^b	483.13 ± 0.00 ^b
BPC4	3.95 ± 0.02 ^a	2.23 ± 0.10 ^d	21.50 ± 0.19 ^a	2.14 ± 0.01 ^a	2.95 ± 0.20 ^d	66.76 ± 0.36 ^a	457.47 ± 0.09 ^a

Results are expressed as mean ± standard deviation. Different superscripted letters within columns are significantly different at $p < 0.05$. Control (100% wheat flour), BPC1 (5%), BPC2 (10%), BPC3 (15%) and BPC4 (20%) *P. curatellifolia* peel flour.

The ash content of the biscuits differed significantly ($p < 0.05$). The ash content of the formulated biscuits increased with the incorporation levels of *P. curatellifolia* peel flour, with values varying from 0.74 to 2.03%. Sample BPC4 had the highest ash content, and this shows that the peels contain essential minerals, and the addition of peel flour could boost the mineral content of the biscuits. Gondim et al. [54] indicated that fruit peels have more minerals than edible parts, which explains the high ash content of composite biscuits in this study. Ho and Abdul Latifa [49] reported similar results whereby the incorporation of pitaya peel flour enhanced the ash content of biscuits.

The inclusion of *P. curatellifolia* peel flour significantly decreased ($p < 0.05$) the fat content of the biscuits. The decrease in fat content of composite biscuits could be due to related size particles of both wheat and *P. curatellifolia* peel flour, which facilitate the same level of oil removal from the biscuits. A low fat content is desirable for keeping quality because biscuits with a high fat content are prone to lipid oxidation. Bertagnolli et al. [20] reported similar results of a low fat content of biscuits incorporated with guava peel flour.

Protein content showed a significant decrease across the samples ($p < 0.05$), ranging from 2.46 to 2.17%. Low protein content in composite biscuits might be due to a reduction in the amount of wheat flour used to produce biscuits, since wheat flour contains high gluten protein content. Moreover, a significant decrease in protein content might be due to the thermal treatment that was applied during the preparation of peel flour and biscuits. High temperature denatures protein and breaks down the amino acid chains. The addition of water during dough development could have also caused the protein content to decrease. Chatepa et al. [55] reported protein content of 4.17 and 3.4% for fresh peels and pulp of *P. curatellifolia* fruit, respectively. Similar results of a decrease in protein content of biscuits added with orange peels powder were reported by Rani et al. [56].

The crude fibre content of biscuits showed a significant increase ($p < 0.05$) with an increase in the level of *P. curatellifolia* peel flour, with results ranging from 0.39 to 2.85%.

The significant increase in crude fibre content could be due to a higher percentage of dietary fibre in the peels than that of the control sample. Mawula [57] reported that the peels of *P. curatellifolia* have crude fibre of 19.4%. Moreover, other components available in *P. curatellifolia* peels might have contributed to the development of extra crude fibre during baking [58]. Oxidative enzymes such as peroxidase and phenolase catalyse the development of cross-links between carbohydrates such as arabinoxylans, also between carbohydrate and side chains of amino acids in protein through phenolic molecules such as ferulic acid [59,60]. The high fibre content of biscuits is advantageous since crude fibre

have many health benefits, including prevention of certain cancers and lowering the risk of developing haemorrhoids. Youssef and Mousa [61] also reported similar results where the inclusion of citrus peel powder increased the crude fibre content of biscuits.

The carbohydrate content of the biscuits ranged from 64.86% to 68.94%, and there was a significant decrease as the level of *P. curatellifolia* peel flour increased. The low values of carbohydrate content in composite biscuits might be due to the dilution of the flour with the incorporation of peel flour. The carbohydrate content was lower in comparison to that reported by Bertagnolli et al. [20] in biscuits added with guava peel flour (77.5%). Pravin and Sanita [62] reported that the fruits' by-products had low digestible carbohydrates and were high in fibre. Moreover, Ayo et al. [63] observed a similar decreasing trend in carbohydrates content as the level of orange peel flour was increasing in the study of *acha*-orange peel flour blend biscuits.

The total energy of the biscuits ranged from 497.29 to 457.47 kcal/g and had a decreasing trend with a high inclusion of *P. curatellifolia* peel flour. The decreasing trend of total energy is likely caused by the high fibre content of peel flour. The low energy values of these biscuits have some nutritional and health benefits. For example, regular intake could aid in reducing weight for overweight/obese persons or in controlling sugar diabetes.

3.5. Polyphenolic Compounds and Antioxidant Activity of Biscuits

Table 5 presents the results of polyphenolic compounds and antioxidant properties of biscuits. Total phenolic compounds (TPC) of the biscuits varied from 20.01 to 40.01 mg GAE/g. There was an increasing trend with the incorporation level of *P. curatellifolia* peel flour. The increase in the TPC of the biscuits may be related to more bound phenolic acid from the disintegration of cellular components during the baking process [64]. Moreover, the increase of TPC of biscuits might also be attributed to the Maillard reaction during baking [65]. Prithwa and Sauryya [66] reported similar results where the incorporation of juice and peels powder of fresh pomegranate increased bioactive compounds of the biscuits.

Table 5. Polyphenolic compounds and antioxidant activity of the composite biscuits.

Samples	TFC (mg CE/g)	TPC (mg GAE/g)	DPPH %	FRAP (mg GAE/g)
Control	0.028 ± 0.01 ^a	20.01 ± 0.42 ^a	48.70 ± 1.69 ^a	108.33 ± 5.80 ^a
BPC1	0.061 ± 0.05 ^b	24.76 ± 0.50 ^a	78.74 ± 0.66 ^b	142.33 ± 4.04 ^b
BPC2	0.073 ± 0.05 ^c	32.37 ± 3.82 ^b	90.21 ± 0.48 ^c	161.67 ± 3.51 ^c
BPC3	0.086 ± 0.03 ^d	46.49 ± 2.35 ^c	90.79 ± 0.27 ^c	162.67 ± 2.11 ^d
BPC4	0.104 ± 0.05 ^e	48.51 ± 2.58 ^c	94.72 ± 0.46 ^d	162.67 ± 2.11 ^d

Results are expressed as mean ± standard deviation. Different superscripted letters within columns are significantly different at $p < 0.05$. Control (100% wheat flour), BPC1 (5%), BPC2 (10%), BPC3 (15%) and BPC4 (20%) *P. curatellifolia* peel flour. TPC = total phenolic content; TFC = total flavonoid content.

The total flavonoids content (TFC) of the composite biscuits significantly increased with high levels of incorporation. The TFC of biscuits varied from 0.028 to 0.108 mg CE/g. This increase in TFC of biscuits might be attributed to the formation of brown pigments (meladins) that are Maillard reaction products that take place during baking [67]. Similar results of an increase in TFC were observed by Mahlako et al. [43] for biscuits added with banana and prickly pear peel flour.

The values of percentage inhibition DPPH ranged from 48.70 to 94.72%. The DPPH values of composite biscuits significantly increased ($p < 0.05$) with the increment of levels of incorporated *P. curatellifolia* peel flour. The generation of melanoidins during baking might be responsible for the increase in DPPH values since they have antioxidant capacity [68]. Moreover, high DPPH values might also be due to high TPC and TFC because of the inclusion of peel flour. Higher DPPH values are associated with stronger antioxidant activity, while lower values are related to a weaker antioxidant activity [69]. The inhibition of DPPH follows the same order as TPC, i.e., where the concentration of phenolic compounds

increases, the DPPH also shows an increase. Similar results of an increase in DPPH were reported by Ajila et al. [58] for biscuits added with mango peels powder.

The FRAP results for biscuits ranged from 108.33 to 162.67 mg GAE/g, and there was a significant increase ($p < 0.05$) with the incorporation level of peel flour. The increase of FRAP values might be due to the generation of the compound during Maillard browning since the soluble part of the compounds possesses a metal-chelating ability [68].

Chen and Kitts [70] indicated that the thermal process of food material produces Maillard browning pigments that are known to possess substantial antioxidant activity. Results are in line with a report of Mahloko et al. [21], where the inclusion of banana and prickly pear peel flour enhanced the FRAP values of biscuits. Therefore, the inclusion of *P. curatellifolia* fruit peel flour improves the health benefits by enhancing the antioxidant properties of biscuits.

3.6. Physical Properties of Biscuits

Results for thickness, weight, diameter, spread ratio and hardness are displayed in Table 6. The thickness values ranged from 1.10 to 1.23 mm and decreased with the inclusion of *P. curatellifolia* fruit peel flour, and the dilution of gluten might have contributed to the decrease [71]. On the other hand, the high thickness value of the control sample might be due to the lower hygroscopy of wheat flour, which allowed more water to be present for gluten proteins to produce a network and to increase the height of biscuits [72,73]. Similar results of decrease in thickness were reported by Ho and Abdul-Latifa et al. [49] for biscuits incorporated with pitaya peel flour.

Table 6. Physical properties of composite biscuits.

Samples	Thickness (mm)	Weight (g)	Diameter (mm)	Spread Ratio	Hardness (g)
Control	1.23 ± 0.06 ^c	9.97 ± 0.11 ^c	3.57 ± 0.12 ^a	2.67 ± 0.12 ^a	1188.13 ± 2.01 ^a
BPC1	1.10 ± 0.10 ^a	9.43 ± 0.15 ^b	3.63 ± 0.06 ^b	3.95 ± 0.31 ^c	1758.49 ± 1.20 ^b
BPC2	1.13 ± 0.06 ^a	9.43 ± 0.64 ^b	3.63 ± 0.12 ^b	3.13 ± 0.12 ^b	1764.24 ± 1.43 ^c
BPC3	1.10 ± 0.10 ^a	8.57 ± 0.06 ^a	3.73 ± 0.06 ^c	3.41 ± 0.31 ^d	1995.67 ± 1.05 ^d
BPC4	1.17 ± 0.06 ^b	9.30 ± 0.44 ^b	3.97 ± 0.12 ^d	3.45 ± 0.13 ^d	2432.60 ± 2.08 ^e

Results are expressed as mean ± standard deviation. Different superscripted letters within columns are significantly different at $p < 0.05$. Control (100% wheat flour), BPC1 (5%), BPC2 (10%), BPC3 (15%) and BPC4 (20%) *P. curatellifolia* peel flour.

The weight of the control and composite biscuits varied from 8.57 to 9.97 g and significantly decreased ($p < 0.05$) with the inclusion level of peel flour. This might be attributed to the lower solubility of peel flour during baking which allowed more free water to be absorbed by the fibre.

The diameter of biscuits increased with the inclusion level of *P. curatellifolia* peel flour, with values ranging from 3.57 to 3.97 mm. The low viscosity of composite flours might have contributed to the decrease in diameter since it makes the dough have a high flow rate and thereby increases the diameter of composite biscuits [49]. The incorporation of *P. curatellifolia* peel flour decreased the gluten protein of composite flours, and this caused a low viscosity of the dough. Similar results were observed by Nassar et al. [74], where the inclusion of citrus peel flour improved the diameter of biscuits.

Results for the spread ratio of the biscuits varied from 2.67 to 3.41 and significantly increased ($p < 0.05$) with the inclusion of peel flour; this might be attributed to low gluten content. The spread ratio shows the ability of biscuits to rise, and it is controlled by the viscosity of the dough since low viscosity makes biscuits spread faster and vice versa [49,50].

The availability of more water in the dough causes more sugar to be dissolved during mixing. This reduces the initial viscosity of the dough, which results in biscuits spreading faster during heating [75]. On the other hand, the low spread ratio of the control sample suggests that its starches are more hydrophilic. Zouari et al. [50] reported similar results, whereby the addition of sesame peel flour decreased the spread ratio of biscuits.

Values of the hardness of biscuits varied from 1188.13 to 2432.60 g, and the control sample had a lower value. The increase of hardness in composite biscuits can be associated with the hardness of the fibres in composite flour. In addition, the dilution of wheat protein with *P. curatellifolia* peel flour also contributed to the increase since the interchange of the two proteins makes biscuits compact, thereby increasing the hardness [76]. The formation of the gluten network contributes to the hardness of biscuits since gluten stimulates the development of the network by attracting water molecules [73]. Therefore, incorporation of *P. curatellifolia* peel flour did not contribute to the development of the gluten network, hence the high levels of hardness. Pareyt and Delcour [77] observed that the utilisation of flour with high gluten content could reduce the weight, hardness, density and stickiness of the dough. Mahloko et al. [21] reported similar results where the inclusion of prickly pear and banana peel flour increased the hardness of biscuits.

4. Conclusions

The utilisation of *P. curatellifolia* peel flour improves the nutritional, physical and antioxidant properties of bakery products such as biscuits. The formulated biscuits were characterised by higher contents of ash and crude fibre. Other important benefits of *P. curatellifolia* peel flour incorporation included improved polyphenolic compounds and antioxidant activity of biscuits. This is important since consumers are demanding food with nutraceutical and functional properties beyond basic nutrition. In terms of physical properties, the enriched biscuits were harder and had darker colours compared to the control. Overall, *P. curatellifolia* peel flour is a potential raw material for the manufacturing of functional bakery products such as biscuits. Further studies should be carried out to assess the influence of incorporation of *P. curatellifolia* on the functional and pasting properties of flour as well as parameters such as minerals, microstructural properties and consumer acceptance of biscuits.

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