

Antioxidant, physical, and sensory properties of dried fresh onion (*Allium cepa* L.) leaves-added cookies

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Summary

Fresh onion leaves (FOL), which are usually discarded, were used to make functional cookies. FOL was dried, and dried FOL-added cookies were made by replacing 5% to 20% of the wheat flour with dried FOL powder. The antioxidant, physical, and sensory properties of dried FOL-added cookies were investigated. With increasing amounts of dried FOL powder added to cookies, the total oxygen radical absorbance capacity value, total phenolic content, and total vitamin C concentration increased. The surface color of the cookie was changed to dark green using dried FOL powder. Furthermore, the taste, appearance, and other sensory properties of the cookies indicated that replacing up to 10% of the wheat flour with dried FOL powder was acceptable.

Key words

Fresh onion leaves (FOL), Cookies, Antioxidant activity, Surface color, Sensory property

Introduction

Onions (*Allium cepa* L.) are a popular vegetable that is eaten worldwide. Onions are high in quercetin¹⁾, potassium, and dietary fiber²⁾. In Japan, onions are harvested from summer to autumn and again in spring. The early-season variety is harvested from April to May, whereas the very early-season variety is harvested before April³⁾. These early-season onions are known as “fresh onion” in Japan. However, fresh onion leaves (FOL) are discarded before shipment to Japanese markets because of their short shelf life. As such, they are considered food waste in Japan. Phenolic compounds, quercetin, vitamin C, and β -carotene are among the antioxidants found in FOL^{4–7)}. These findings suggest that FOL is a good dietary source of antioxidants and may be a useful new food resource. However, FOL's cooking and processing methods were not addressed.

Currently, developing a sustainable food-supply system is required in accordance with the Sustainable Development Goals (SDGs). Attempts to reuse food loss and/or food waste in the food industry and on farms are one such approach. For example, Ishida *et al.*⁸⁾ proposed that sweet potato leaf powder can be mixed into cookie dough. Pomace from carrots^{9,10)}, pumpkins¹⁰⁾, roselle (*Hibiscus sabdariffa* L.) seeds¹¹⁾, raspberries and blueberries¹²⁾, sour cherries¹³⁾, rose hips, rowanberries, blackcurrants, and elderberries¹⁴⁾, one of the food wastes, was added in cookie doughs in previous studies. Additionally, vegetable and fruit pomace or peel was added to baked goods, such as biscuits¹⁵⁾ and rice flour-based gluten-free cakes¹⁶⁾. According to these studies, the addition of food losses and/or waste increased the levels of phenolic compounds, micronutrients, and dietary fiber in baked goods. Thus, the addition of food losses and/or wastes is beneficial for reuse and increasing the function of baked goods.

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In this study, we added FOL to cookies to develop the cooking method of FOL and to increase the function in cookies based on previous research. As a result, we assessed the antioxidant, physical, and sensory properties of cookies containing FOL. Previous studies dried and powdered vegetables and fruits before adding to baked goods^{8,14,17} because their water content was high. In this study, the FOL was dried and mixed into cookie dough.

Materials and Methods

Dried fresh onion leaves preparation

The cultivar of fresh onion (*A. cepa* L.) was the Ebisudama, and these were purchased with leaves intact from the Shima-to-kurasu K.K. (Awaji city, Hyogo prefecture, Japan) in February 2019. The leaves were collected from the bulbs of fresh onion, and then these were washed with detergent and disinfected by sodium hypochlorite. Samples were washed extensively in tap water, and were dried using a paper towel. Fresh onion leaves were dried under freeze-drying (FD) and low-temperature drying (LD) methods. In the previous study, phenolic compounds concentration in vegetables was changed between FD and LD¹⁸, thus we selected these drying conditions. For FD, samples were first frozen at -20°C for 24 h, before drying for 4 days using the freeze-dryer FDU-1200 (TOKYO RIKAKIKAI CO, LTD., Tokyo, Japan) with dry-chamber DRC-3L (TOKYO RIKAKIKAI CO, LTD., Tokyo, Japan) and oil-sealed rotary vacuum pump GCD-051X (ULVAC KIKO, Inc., Miyazaki, Japan). The cold trap was cooled to -45°C ; vacuum was under 20 Pa absolute pressure. For LD, samples were dried using the hot-air food dryer (Petit Marengi DX TTM-440 N, Tohmei Tech Co., Ltd. Osaka, Japan) at 55°C for 24 h. Dried FOL samples were powdered, and moisture concentrations of these powders for FD and LD were 7.5% and 8.4%, respectively. Dried FOL powders passed through a 500 μm sieve. Dried FOL powders were stored at -30°C until further use.

Cookie preparation

Composition of dried FOL-added cookies was presented Table 1. In dried FOL (FD)-added cookies, 5%, 10%, and 20% of wheat flours were replaced with the dried FOL (FD) powder, respectively. In the dried FOL (LD)-added cookie, 10% of wheat flour was replaced with the dried FOL (LD) powder.

The unsalted butter (Yotsuba Milk Products Co., Ltd., Hokkaido, Japan) was stirred, and the sugar (Fuji Nihon Seito Corporation., Tokyo, Japan) was added and mixed. Subsequently, the beaten egg (Marusan Co., Ltd., Hiroshima, Japan) was added and stirred, and the vanilla essence (Meidi-Ya Co., Ltd., Tokyo, Japan) was added and stirred. Then, the wheat flour (Tomizawa Shouten, Tokyo, Japan) and dried FOL powder were added and stirred, and this dough was cooled at -18°C for 50 min. The dough was flattened out to a thickness of 3 mm, and it was cut out to a diameter of 38 mm with the cookie cutter. Then, these were baked at 170°C for 15 min, and these were cooled down to room temperature. Cookies were packed and sealed in a vacuum package, and these were stored at 4°C until further analysis.

Oxygen radical absorbance capacity (ORAC) value

The ORAC values in the dried FOL powder and dried FOL-added cookie were measured for the hydrophilic and lipophilic fractions using the OxiSelect ORAC Activity Assay Kit (Cell Biolabs, Inc., CA, USA). The hydrophilic and lipophilic fractions were prepared from the dried FOL powder and dried FOL-added cookie, as previously described⁶. The ORAC value was measured for the hydrophilic fraction (hydrophilic (H)-ORAC value) and the lipophilic fraction (lipophilic (L)-ORAC value). The values were expressed as μmol 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox) equivalents (TE) per 100 g of sample weight.

Total phenolic content (TPC)

TPC in the dried FOL powder and dried FOL-added

Table 1 Composition of dried FOL-added cookies

Ingredients (g)	Control	FD5%	FD10%	FD20%	LD10%
Wheat flour	50.0	47.5	45.0	40.0	45.0
Sugar	20.0	20.0	20.0	20.0	20.0
Unsalted butter	30.0	30.0	30.0	30.0	30.0
Egg	15.0	15.0	15.0	15.0	15.0
Vanilla essence	2 drops	2 drops	2 drops	2 drops	2 drops
Dried FOL (FD) powder	0.0	2.5	5.0	10.0	0.0
Dried FOL (LD) powder	0.0	0.0	0.0	0.0	5.0

FOL, fresh onion leaves; FD, freeze-drying; LD, low-temperature drying.

cookie were determined using the Folin-Ciocalteu method, as previously described⁶. The values were expressed as mg gallic acid equivalents (GAE) per 100 g of sample weight.

Total vitamin C

Total vitamin C in the dried FOL powder and dried FOL-added cookie were measured based on the dinitro-phenylhydrazine method, and using a Prominence Ultra-Fast Liquid Chromatography system (Shimadzu Co., Ltd., Kyoto, Japan), as previously described⁶. The analysis conditions were as follows: mobile phase, ethyl acetate: hexane: acetate (50: 40: 10 by vol.); injection volume, 10 μ L; flow rate, 1.5 mL/min; column oven temperature, 40°C; and UV/Vis monitoring wavelength, 495 nm. This method is measuring total amount of ascorbic acid and dehydroascorbic acid. The values were expressed as mg per 100 g of sample weight.

Rupture measurement

The rupture measurement of the dried FOL-added cookie was performed using the RE2-33005B creep meter (Yamaden Co., Ltd., Tokyo, Japan). The cookie was ruptured using a wedge-shaped plunger (No. 49, W13 \times 30°, Yamaden Co., Ltd., Tokyo, Japan) with a load cell of 200 N, and speed of 1.0 mm/s. The samples were measured at 95% strain. The rupture strain, rupture stress (kPa), rupture energy (kJ/m³), and brittle stress (kPa) of samples were evaluated.

Surface color

The surface color of the dried FOL-added cookie was measured using a color-difference meter CR-200 (Konica Minolta, Inc., Tokyo, Japan). The color scale was used to measure the L* (dark to light), a* (green to red), and b* (blue to yellow) parameters. ΔE was calculated as a color difference using the following formula:

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

where ΔL^* , Δa^* , and Δb^* are differences between the brightness, redness, and yellowness intensity difference from the control cookie, respectively. The perception of the color difference ΔE varies according to the sensitivity of the human eye and the observed color. The human eye only distinguishes color difference if ΔE is larger than 1–3. ΔE values of 1 can be detected for some colors (mainly blues), but the same ΔE may not be perceptible for other colors (e.g. red)¹⁹.

Sensory evaluation

The sensory properties of the dried FOL-added cookies

were evaluated by sensory evaluation. The cookies were prepared as described in “*Cookie preparation*”. Five cookies (Control, 5%FD, 10%FD, 20%FD, and 10%LD) were placed into white dishes and randomly tasted by blinded subjects. The welsh onion flavor and sweetness (−3 [weak] to +3 [strong]) and the texture (−3 [moist] to +3 [crispy]) were examined as the intensities. The aroma, color and appearance, sweetness, aftertaste, taste, and overall judgment (−3 [dislike] to +3 [like]) were examined as the preferences. The evaluation was conducted by 25 untrained non-expert Japanese male and female students between the ages of 19 and 24 who were undergoing an education course.

Statistical analysis

Values are shown as the mean \pm standard deviation (SD) or mean. Statistical analysis was performed using Excel 2019 (Microsoft Japan Co., Ltd., Tokyo, Japan) and EZR software (Saitama Medical Center, Jichi Medical University, Saitama, Japan), which is a graphical user interface for R (The R Foundation for Statistical Computing, Vienna, Austria, version 4.0.3)²⁰. The differences between the groups were compared using the Welch's *t*-test and Tukey's honestly significant difference (HSD) test. A *p*-value < 0.05 was considered significant.

Results and discussion

Antioxidant activities and antioxidant concentrations of dried FOL powders and dried FOL-added cookies

The hydrophilic ORAC value, lipophilic ORAC value, total ORAC value (μ mol Trolox equivalent/100 g), TPC (mg gallic acid equivalent/100 g), and total vitamin C concentration (mg/100 g) of dried FOL (FD) powder were 12016.23 \pm 1801.78, 675.66 \pm 145.97, 12691.89 \pm 1798.93, 549.05 \pm 38.00, 351.08 \pm 9.28, respectively (Table 2), whereas those of dried FOL (LD) powder were 12018.46 \pm 1702.44, 674.48 \pm 128.69, 12692.94 \pm 1706.11, 733.77 \pm 134.53, and 295.25 \pm 2.62, respectively (Table 2). According to our findings, dried FOL powders for both FD and LD are an excellent dietary source of antioxidants. Although the hydrophilic, lipophilic, and total ORAC values of dried FOL powders did not differ between FD and LD, TPC was lower (*p* < 0.05) in FD and total vitamin C content was lower (*p* < 0.05) in LD (Table 2). TPC levels in dried vegetables and fruits decreased as drying temperature increased^{18, 21, 22}. TPC in dried tomatoes was higher in hot-air-drying than in FD²³ because higher temperatures deactivate oxidative and hydrolytic enzymes for phenolic compounds, preventing TPC loss in hot-air-drying.

Previous research found that heating treatments above 45°C reduced polyphenol oxidase (PPO) activities^{24,25}. As a result, in the current study, deactivating these enzymes with heating treatment resulted in a high level of TPC in dried FOL (LD) powder. Conversely, vitamin C concentrations in vegetables and fruits decreased with increasing drying temperature^{23,26}, and this finding was confirmed in dried FOL powders in this study.

The hydrophilic ORAC value, lipophilic ORAC value, total ORAC value, TPC, and total vitamin C concentration in cookies added with dried FOL (FD) increased with an increasing amount of dried FOL powder ($p < 0.05$) (Table 2). According to our findings, adding dried FOL to cookies improves their antioxidant activity. Conversely, the hydrophilic and total ORAC values of the 10%LD cookie were higher than those of the 10%FD cookie ($p < 0.05$). This finding could be explained by the 10%LD cookie having a higher total vitamin C content than that of the 10%FD cookie ($p < 0.05$). Plant tissue damage releases ascorbate oxidase, which reduces the vitamin C content of raw vegetables and fruits^{27,28}. Because ascorbate oxidase in dried FD powder is activated during the cooking process, vitamin C may oxidize and decrease in dried FOL (FD)-added cookies. Furthermore, Leong and Oey²⁹ claimed that a 3-h

heating treatment at 55°C reduced more than 90% of the ascorbate oxidase activity. As a result of the LD process deactivating this enzyme in dried FOL, the total vitamin C concentration in the 10%LD cookie was higher than that in dried FOL (FD)-added cookies.

Vegetables with total ORAC values of 265–357 μmol Trolox equivalent/100 g include Japanese radish, Chinese cabbage, lettuce, Welsh onion, and tomato³⁰. Furthermore, vegetables high in phenolic compounds, such as spinach, sweet pepper, eggplant, and broccoli, contain 49–62 mg gallic acid equivalent/100 g³⁰. All of the dried FOL-added cookies had nearly identical values for these parameters (Table 2), implying that dried FOL-added cookies could be an excellent dietary source of phenolic compounds. Conversely, the total vitamin C concentrations (mg/100 g) in 5%FD, 10%FD, 20%FD, and 10%LD cookies were theoretically 8.4, 16.7, 33.4, and 14.0, respectively, but these values in cookies were practically lower than theoretical values. This observation was made as a result of the baking process and the degradation of vitamin C during cookie preparation.

Table 2 Antioxidant activity and antioxidants in dried FOL powders and dried FOL-added cookies

		ORAC values (μmol Trolox equivalent/100 g)			Total phenolic content (mg Gallic acid equivalent/100 g)	Total vitamin C (mg/100 g)
		Hydrophilic	Lipophilic	Total		
Dried FOL powder (n=5)	Freeze-drying (FD)	12016.23 \pm 1801.78	675.66 \pm 145.97	12691.89 \pm 1798.93	549.05 \pm 38.00	351.08 \pm 9.28
	Low-temperature drying (LD)	12018.46 \pm 1702.44	674.48 \pm 128.69	12692.94 \pm 1706.11	733.77 \pm 134.53 [*]	295.25 \pm 2.62 [*]
Dried FOL-added cookies (n=3)	Control	109.03 \pm 10.86 ^a	19.77 \pm 5.07 ^a	128.80 \pm 12.12 ^a	34.08 \pm 2.38 ^a	LOD ^a
	5%FD	296.81 \pm 28.80 ^b	31.67 \pm 2.09 ^b	328.48 \pm 30.23 ^b	55.05 \pm 6.32 ^b	2.15 \pm 0.10 ^b
	10%FD	298.81 \pm 35.16 ^b	37.19 \pm 5.35 ^{bc}	335.38 \pm 31.97 ^b	66.66 \pm 4.04 ^{bc}	4.09 \pm 0.16 ^c
	20%FD	794.79 \pm 116.60 ^c	55.77 \pm 3.47 ^d	850.57 \pm 115.91 ^c	107.98 \pm 5.66 ^d	6.95 \pm 0.81 ^d
	10%LD	455.20 \pm 27.75 ^d	44.86 \pm 2.31 ^c	500.06 \pm 27.56 ^d	77.38 \pm 9.88 ^c	9.69 \pm 0.26 ^e

FOL, fresh onion leaves; FD, freeze-drying; LD, low-temperature drying; ORAC, oxygen radical absorbance capacity; Trolox, 6-hydroxy-2, 5, 7, 8-tetramethylchroman-2-carboxylic acid; LOD, limit of detection. Values are the mean \pm SD. * $p < 0.05$ (Welch's *t*-test), compared between FD and LD in dried FOL powder. ^{a-e} Values with different superscripts indicate a significant difference ($p < 0.05$, Tukey's HSD test), compared between FOL-added cookies.

Table 3 Rupture properties of dried FOL-added cookies

	Rupture strain	Rupture stress (kPa)	Rupture energy (kJ/m ³)	Brittle stress (kPa)
Control	0.06 \pm 0.03	543.74 \pm 119.54 ^a	17.27 \pm 10.73 ^a	190.84 \pm 54.00
5%FD	0.07 \pm 0.02	745.89 \pm 235.88 ^a	30.44 \pm 16.77 ^{ab}	204.03 \pm 92.84
10%FD	0.05 \pm 0.01	735.11 \pm 137.08 ^a	21.29 \pm 5.61 ^{ab}	192.22 \pm 40.13
20%FD	0.08 \pm 0.03	1097.73 \pm 227.81 ^b	49.99 \pm 24.28 ^b	238.04 \pm 134.97
10%LD	0.06 \pm 0.02	694.76 \pm 131.91 ^a	27.08 \pm 17.81 ^{ab}	219.47 \pm 94.04

FD, freeze-drying; LD, low-temperature drying. Values are the mean \pm SD (n=5). ^{a-b} Values with different superscripts indicate a significant difference ($p < 0.05$, Tukey's HSD test), compared between FOL-added cookies.

Rupture properties and surface colors of dried FOL-added cookies

The rupture stress and rupture energy were higher in the 20%FD cookie than in the control cookie ($p < 0.05$) (Table 3). Rupture strains and brittle stress did not differ between cookies. These results indicate that replacing more than 20% of the wheat flour with dried FOL powder (FD) hardens the texture of the cookie. The increase in hardness of the 20%FD cookie might be related to the amount of dietary fiber and its high water-holding capacity. Similar studies indicated that doughs have higher moisture content due to increase dietary fiber and produce an extensive gluten structure and harder cookies and biscuits^{10, 15}. Previous research and our findings suggest that adding dried vegetable or fruit powder to the dough may harden the texture of the cookie by increasing dietary fiber and its water-holding capacity.

The L^* and b^* values of dried FOL-added cookies decreased as the amount of dried FOL powder added to the cookie dough increased ($p < 0.05$) (Table 4). The a^* values of 5%FD, 10%FD, 20%FD, and 10%LD cookies were significantly lower than that of the control cookie ($p < 0.05$) (Table 4). ΔE values of dried FOL-added cookies increased as the amount of dried FOL powder added to the cookie

dough increased (Table 4). As a result, by adding dried FOL powder to the dough, the surface color of the cookie is changed to dark green.

Sensory properties of dried FOL-added cookies

An increasing amount of dried FOL powder added to the cookie dough increased the Welsh onion flavor and decreased the sweetness ($p < 0.05$), but the texture did not change between cookies (Fig. 1). Although the rupture forces in the 20%FD cookie increased, the cookie texture remained virtually unchanged by adding dried FOL powder to the dough. An increasing amount of dried FOL powder added to the cookie dough reduced the aroma, surface color and appearance, sweetness, aftertaste, taste, and overall judgment in preferences scores ($p < 0.05$) (Fig. 1). Our results suggest that the taste, aroma, surface color, and appearance of cookies are changed by adding dried FOL powder to the dough, but not their texture. However, the overall judgment score in the 10%FD dried FOL-added cookie was 0.79, ranging from 0 (neither) to 1 (lightly like). By contrast, the score in the 20%FD dried FOL-added cookie was -0.49, ranging from -1 (lightly dislike) to 0 (neither) (Fig. 1). As a result, our finding indicated that replacing up to 10% of the wheat flour with

Table 4 Surface colors of dried FOL-added cookies

	L^*	a^*	b^*	ΔE
Control	73.46 \pm 2.14 ^a	5.18 \pm 1.79 ^a	38.36 \pm 1.06 ^a	—
5%FD	54.07 \pm 4.18 ^b	-4.70 \pm 2.49 ^b	35.43 \pm 1.98 ^{b,c}	21.96
10%FD	49.00 \pm 2.63 ^{cd}	-9.29 \pm 2.24 ^b	34.63 \pm 1.35 ^c	28.66
20%FD	40.44 \pm 2.34 ^e	-6.72 \pm 5.16 ^b	27.24 \pm 2.45 ^d	36.81
10%LD	50.73 \pm 1.16 ^{bd}	-6.95 \pm 2.39 ^b	34.70 \pm 1.32 ^c	26.02

FD, freeze-drying; LD, low-temperature drying. Values are the mean \pm SD (n=5). ^{a-e} Values with different superscripts indicate a significant difference ($p < 0.05$, Tukey's HSD test), compared between FOL-added cookies.

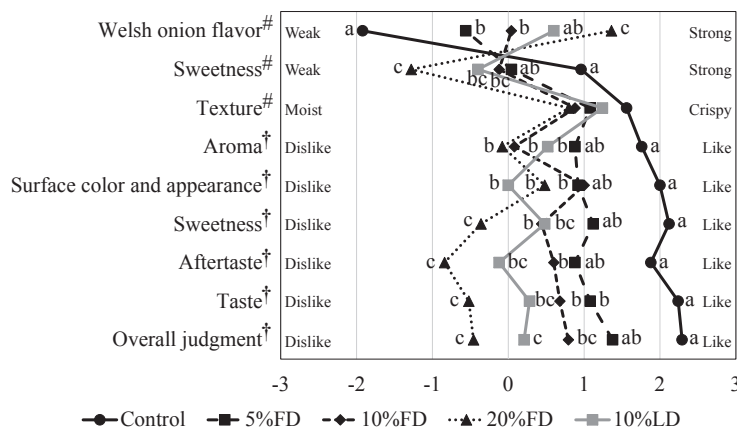


Fig. 1. Sensory properties of dried FOL-added cookies. #Intensity; †Preference. FD, freeze-drying; LD, low-temperature drying. Values are the mean (n=25). ^{a-c} $p < 0.05$ (Tukey's HSD test), compared between dried FOL-added cookies.

dried FOL powder was acceptable in the dried FOL-added cookie.

Conclusion

In this study, we found that adding dried FOL powder to cookies improves their antioxidant activity and TPC. By adding the dried FOL powder to the dough, the surface color of the cookie changes to dark green and the preference for appearance and taste decreases. However, replacing up to 10% of the wheat flour with dried FOL powder was acceptable in the dried FOL-added cookie. When adding dried FOL powder to cookies, LD powder is preferred over FD powder because the antioxidant activity and vitamin C content of the 10%LD cookie were higher than those of the 10%FD cookie. Our findings suggested that drying FOL and replacing wheat flour in cookie dough with dried FOL powder are beneficial for reusing fresh onion leaves.

Authors' contribution

AT and MT conceived and designed the study. MY, AT, RS, and MT performed the experiments. MY, RS, and AT analyzed the data. MY prepared figure and tables, and wrote the paper.

Declaration of competing interest

The authors declare that they have no conflict of interest.

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