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Short communication

Effect of far-infrared radiation and heat treatment on the antioxidant activity of water extracts from peanut hulls

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Abstract

The antioxidant activities of peanut (*Arachis hypogaea* L.) hull extracts were evaluated after far-infrared (FIR) radiation or heat treatment. Peanut hulls in petri dishes were FIR-irradiated or heat-treated (150 °C) for 5, 10, 15, 20, 40 or 60 min. The water extracts (300 mg/10 mL) of peanut hulls (WEPH) were prepared and their total phenol contents (TPC), radical scavenging activity (RSA), and reducing power were determined. The antioxidant activities of WEPH increased as the time of heating or FIR-radiation increased. When peanut hulls were FIR-irradiated at 150 °C for 60 min, the values of TPC, RSA, and reducing power of WEPH increased from 72.9 to 141.6 μM, 2.34% to 48.83%, and 0.473 to 0.910, respectively, compared to the untreated controls. Heat treatment of peanut hull under the same conditions (150 °C for 60 min) also increased the TPC, RSA, and reducing power of WEPH from 72.9 to 90.3 μM, 1.90% to 23.69%, and from 0.471 to 0.718, respectively. The result indicated that FIR-radiation or heat treatment on peanut hulls increased the antioxidant activities of WEPH.

Keywords: Peanut hulls; Extracts; Far-infrared radiation; Heat treatment; Antioxidant activity

1. Introduction

Several synthetic antioxidants, such as butylated hydroxyanisole, butylated hydroxytoluene and *tertiary*-butylhydroquinone have been widely used in foods to prevent oxidation. The use of synthetic antioxidants in foods, however, is discouraged because of their toxicity and carcinogenicity (Ito et al., 1986). Therefore, special interests have been focused on the use of natural antioxidants that can remove free radicals causing various diseases, carcinogenesis, and aging (Pokorny, 1991). Natural antioxidant compounds such as flavonoids, tannins, coumarins, curcuminoids, xanthons, phenolics, and terpenoids are found in the fruits, leaves, seeds, and oils of various plant products (Larson, 1988), and some of these are as much effective as synthetic antioxidants in model systems (Deshpande, Sathe, & Salunkhe, 1984; Duthie & Crozier, 2000).

Peanut (*Arachis hypogaea* L.) is one of most widely used nuts due to its nutrition and taste. Peanut kernel has been reported to contain antioxidant flavonoids, dihydroquercetin (Pratt & Miller, 1984), and ethyl protocatechuate was identified as antioxidant component in peanut seed (Huang, Yen, Chang, Yen, & Duh, 2003). Antioxidant properties of methanolic extracts from peanut hulls (PH) were investigated (Yen & Duh, 1993), and luteolin was identified as a major flavonoid in mature peanut to show high antioxidant activity (Duh, Yeh, & Yen, 1992).

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Many antioxidant phenolic compounds in plants, however, are most frequently present as covalently bound form with insoluble polymers (Niwa & Miyachi, 1986; Peleg, Naim, Rouseff, & Zehavi, 1991). Therefore, it is necessary to find effective processing methods to liberate the natural antioxidant compounds from plant sources. Previously we reported that far-infrared (FIR) radiation or simple heat treatment could liberate and activate low-molecular-weighted natural antioxidants in plants (Jeong et al., 2004; Jeong, Kim, Kim, Jo et al., 2004; Lee et al., 2003). The objective of this research was to determine the antioxidant activities of FIR-radiated or heat-treated peanut hull extracts.

2. Materials and methods

2.1. Materials

Peanuts (*A. hypogaea* L.) were purchased from a local market (Masan, Korea) and divided into hulls and edible parts. The peanut hulls (PH) were stored at 4 °C until used. Tannic acid, and 1,1-diphenyl-2-picrylhydrazyl (DPPH) were purchased from Sigma Chemical Co. (St. Louis, MO, USA) and Folin–Ciocalteu reagent from Wako Pure Chemical Industries, Ltd. (Osaka, Japan). Other reagents were all of analytical grade and used as received.

2.2. Far-infrared radiation or heat treatment

PH were dried under room temperature and finely ground using a blender (MC-811C, Novita, Korea). The milled PH (1 g) was placed as a thin layer in a Pyrex petri dish (8.0 cm diameter) and irradiated using a FIR heater (35 × 10 cm, output 300 W, Hakko Electric Machine Works Co., Ltd., Nagano, Japan), which emitted radiation at the wavelength range from 2 to 14 μm in a FIR-dryer (A-Sung Tester, Korea). A sample holding tray in the middle of FIR-dryer was placed to face the FIR heater with rotation for even irradiation. FIR-radiation was carried out with separate batches for 5, 10, 15, 20, 40 or 60 min at a controlled temperature of 150 °C. In case of heat treatment, PH was roasted in an electric muffle furnace (Model DMF-802, Daeil Engineering, Korea) under the same conditions of FIR-radiation. After heating, PH was allowed to cool to ambient temperature before extraction.

2.3. Preparation of water extracts from PH

FIR-irradiated or heat-treated PH (0.3 g) was extracted with 10 mL of distilled water in a shaking incubator (100 rpm) at room temperature overnight. Then the extracts were centrifuged at 1000*g* for 15 min and the supernatants were filtered through a Whatman No. 1 filter paper. The distilled water extract of PH (WEPH) was used to determine the antioxidant activities.

2.4. Total phenolic contents

The total phenolic contents (TPC) of WEPH was determined according to the method of Gutfinger (1981). One mL of WEPH was mixed with 1 mL of the 50% Folin–Ciocalteu reagent and 1 mL of 2% Na_2CO_3, and then centrifuged at 13,400*g* for 5 min. After 30 min incubation at room temperature, the absorbance was measured with a spectrophotometer (Shimadzu UV-1601, Tokyo, Japan) at 750 nm. TPC were expressed as tannic acid equivalents.

2.5. DPPH radical scavenging activity

The DPPH radical scavenging activity of WEPH was estimated according to the method of Blois (1958). After mixing 0.1 mL of WEPH with 0.9 mL of 0.041 mM DPPH in ethanol for 10 min, the absorbance of the sample was measured at 517 nm. Radical scavenging activity was expressed as percentage according to the following formula:

$$\%\text{DPPH radical scavenging activity} = (1 - \text{sample OD}/\text{control OD}) \times 100$$

2.6. Reducing power

The reducing power of WEPH was determined according to the method of Oyaizu (1986). WEPH (1 mL), phosphate buffer (1 mL, 0.2 M, pH 6.6) and potassium ferricyanide (1.0 mL, 10 mg/mL) were mixed together and incubated at 50 °C for 20 min. Trichloroacetic acid (1.0 mL, 100 mg/mL) was added to the mixture and centrifuged at 13,400*g* for 5 min. The supernatant (1.0 mL) was mixed with distilled water (1.0 mL) and ferric chloride (0.1 mL, 1.0 mg/mL), and then the absorbance was measured at 700 nm.

2.7. Gas chromatography/mass spectrometry analysis of PH extracts

Extracts from untreated and treated (150 °C for 60 min) PH were dissolved in ethanol (200 mg/mL) and centrifuged at 13,400*g* for 5 min to precipitate undissolved materials. The supernatant was mixed with 4 volumes of BSA [*N*,*O*-bis(trimethylsilyl)acetamide] and derivatized in a water bath (70 °C) for 15 min (Du & Ahn, 2002). The compounds in PH extracts were identified using a gas chromatograph/mass spectrometer (GC6890/MS5973, Hewlett–Packard Co., Wilmington, DE). A split inlet (99:1) was used to inject sample (5 μL) into an HP-5 column (30 m, 0.32 mm i.d., 0.25

μm film; Hewlett–Packard Co., Wilmington, DE). A ramped oven temperature was used (100 °C for 2 min, increased to 270 °C at 10 °C/min, and held for 6 min). The inlet temperature was 250 °C and the carrier gas was helium at 1.5 mL/min. The ionization potential of mass selective detector was 70 eV and the scan range was 19.1–400 *m*/*z*. Identification of compounds detected was achieved by comparing mass spectral data of samples with those of the Wiley library (Hewlett–Packard Co.).

2.8. Statistical analysis

All measurements were done in triplicate, and analysis of variance was conducted by the procedure of general linear model using SAS software (SAS Institute, 1995). Student–Newman–Keul's multiple range tests were used to compare the differences of the mean values among treatments ($P < 0.05$).

3. Results and discussion

Phenolic compounds are the most active antioxidant derivatives in plants (Bors, Michel, & Stettmaier, 2001). They are known to act as antioxidants not only because of their ability to donate hydrogen or electrons but also because they are stable radical intermediates (Cuvelier, Richard, & Berset, 1992; Maillard, Soum, Boivia, & Berset, 1996). Generally, the outer layers of plant such as peel, shell, and hull contain large amount of polyphenolic compounds to protect inner materials. A number of phenolic acids are linked to various cell wall components such as arabinoxylans and proteins (Harris & Hartley, 1976; Hartley, Morrison, Himmelsbach, & Borneman, 1990). Duh et al. (1992) identified luteolin, a flavonoid which has a strong antioxidant activity, from methanolic extract from PH.

The TPC in WEPH increased significantly by FIR-radiation or heat treatment and the increase was time-dependent (Table 1). The TPC of WEPH was increased from 72.9 μM in unheated control to 141.6 μM by FIR-radiation and to 90.3 by heating at 150 °C for 60 min. Therefore, FIR radiation of PH was more efficient to increase phenolic contents from WEPH than simple heating. FIR rays are defined as electromagnetic waves which have wavelengths longer than 4 μm but shorter than microwave ($\lambda > 0.1$ cm). FIR rays are biologically active (Inoue & Kabaya, 1989) and transfer heat to the center of materials evenly without degrading the constituent molecules of surface (Niwa, Kanoh, Kasama, & Neigishi, 1988). FIR may have capability to cleave covalent bonds and liberate antioxidants such as flavonoids, carotene, tannin, ascorbate, flavoprotein or polyphenols from repeating polymers (Niwa et al., 1988). Our previous study (Lee et al., 2003) showed that simple heat treatments could not cleave covalently bound phenolic compounds from rice hulls but FIR treatments could. On the other hand, simple heat treatments increased the TPC of defatted sesame meals (Jeong et al., 2004) and citrus peels (Jeong, Kim, Kim, Jo et al., 2004). This indicated that phenolic compounds of plants should be present in different binding status depending on plant species. Thus, effective processing steps to liberate antioxidant compounds from different plants may not be the same.

Radical scavengers were evaluated by their reactivity toward a stable free radical, DPPH. DPPH radical scavenging activities (RSA) of WEPH also significantly increased with FIR-radiation (Table 2). After FIR radiation for 60 min, the percentage of RSA increased from 2.34% to 48.33%. Though simple heating also increased the RSA to 23.69%, the increasing activity was lower than that of FIR-radiation. The increase of RSA was also dependent upon the exposure time to FIR-radiation or heat.

The power of certain antioxidants is associated with their reducing power (Jayaprakasha, Singh, & Sakariah, 2001), which is associated with the presence of reductones (Duh, 1998). The reducing power of WEPH increased significantly by FIR-radiation or heat treatment (Table 3). The reducing power of WEPH increased from 0.473 to 0.910 by FIR radiation for 60 min at 150 °C and to 0.718 by heating at the same heating time and temperature conditions.

Methanolic extract from PH showed a strong antioxidant activity in fried potato chips (Rehman, 2003). However, to use the extracts of PH as an antioxidant

Table 1
Effect of FIR-radiation and heat treatments on total phenolic contents of water extract from peanut hulls (WEPH) (unit: μM)

	Time (min)							SEM[A]
	0	5	10	15	20	40	60	
FIR-radiation	72.9^{e}	79.3^{de}	88.6^{d}	99.4^{cx}	107.8^{cx}	124.1^{bx}	141.6^{ax}	0.3
Heat treatment	72.9^{c}	79.8^{b}	79.5^{b}	78.6^{by}	78.5^{by}	86.7^{ay}	90.3^{ay}	0.1
SEM[A]	0.2	0.2	0.3	0.2	0.4	0.3	0.2	

[a–e] Different letters within a row are significantly different ($P < 0.05$), $n = 3$.
[x–y] Different letters within a column are significantly different ($P < 0.05$).
[A] Standard error of the means.

Table 2
Effect of FIR-radiation and heat treatments on the radical scavenging activity of water extract from peanut hulls (WEPH) (unit: %)

	Time (min)							SEM[A]
	0	5	10	15	20	40	60	
FIR-radiation	2.34[f]	6.25[f]	14.68[e]	26.21[d]	33.42[cx]	43.39[bx]	48.83[ax]	1.73
Heat treatment	1.90[c]	11.32[b]	18.62[a]	19.35[a]	16.13[aby]	19.78[ay]	23.69[ay]	1.87
SEM[A]	0.51	1.35	1.90	1.98	2.51	2.35	1.14	

[a–f] Different letters within a row are significantly different ($P < 0.05$), $n = 3$.
[x–y] Different letters within a column with same color value are significantly different ($P < 0.05$).
[A] Standard error of the means.

Table 3
Effect of FIR-radiation and heat treatments on the reducing power of water extract from peanut hulls (WEPH) (unit: Absorbance)

	Time (min)							SEM[A]
	0	5	10	15	20	40	60	
FIR-radiation	0.473[e]	0.460[ex]	0.558[d]	0.695[cx]	0.704[cx]	0.794[bx]	0.910[ax]	0.005
Heat treatment	0.471[d]	0.493[cdy]	0.533[c]	0.554[cy]	0.552[cy]	0.659[by]	0.718[ay]	0.015
SEM[A]	0.007	0.007	0.007	0.009	0.007	0.019	0.015	

[a–e] Different letters within a row are significantly different ($P < 0.05$), $n = 3$.
[x–y] Different letters within a column with same color value are significantly different ($P < 0.05$).
[A] Standard error of the means.

in foods, methanol should be substituted with some harmless solvent. Although water is not as effective as organic solvents to extract useful compounds from plants, it can be a good candidate when an appropriate processing such as FIR-radiation or heating is combined.

Different types of phenolic compounds were detected in water extracts of peanut hulls by gas chromatograph/mass spectrometer (Table 4). Compared to unheated extract, the extract from heat-treated or FIR-radiated PH had more versatile phenolic compounds. 4-hydroxy-3-methoxy benzaldehyde, 4-hydroxy-3-methoxy benzoic acid, and 4-hydroxy-3-methyl benzenacetic acid were newly detected in roasted peanut hull extract. The amounts of 2-methoxy phenol increased in the order of control, roasted, and FIR-radiated samples. FIR-radiated peanut extract had greater number and higher amounts of phenolics than heat-treated ones. The results are very credible when they were compared with the antioxidant activities shown in Tables 2 and 3. There-

Table 4
Representative phenolic compounds of water extract from peanut hulls (WEPH) affected by FIR-radiation and heat treatments at 150 °C for 60 min

RT	Compound	Total ion counts ($\times 10^4$)
Untreated		
4.23	2-Methoxy phenol	217
7.24	2-Methoxy-5-vinyl phenol	55
10.98	4-Hydroxy-3-methoxy benzenacetic acid	143
Roasted		
7.24	2-Methoxy-5-vinyl phenol	253
9.12	4-Hydroxy-3-methoxy benzaldehyde	197
10.95	4-Hydroxy-3-methoxy benzenacetic acid	168
11.77	4-Hydroxy-3-methoxy benzoic acid	93
14.02	4-Hydroxy-3-methyl benzenacetic acid	120
FIR-radiated		
4.29	2-Methoxy phenol	1736
4.33	2-Hydroxy-4-methoxy benzoic acid	1617
7.24	2-Methoxy-5-vinyl phenol	143
9.12	4-Hydroxy-3-methoxy benzaldehyde	795
9.52	2,4-Bis(1,1-dimethylethyl) phenol	446
10.98	4-Hydroxy-3-methoxy benzenacetic acid	115
11.06	Vanillyl alcohol	75
14.40	Methyl cinnamate	152

fore, it was concluded that FIR-radiation was more effective way than simple heat treatments in making the antioxidant phenolic compounds in peanut hulls active.

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