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ARTICLE

Effect of Charcoal Broiling on the Formation of Volatile Compounds in Gamma-Irradiated *Dakgalbi*, a Korean Chicken-Based Food

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Abstract

The purpose of this study is to evaluate the change of volatile compounds in *Dakgalbi* cooked by charcoal broiling. Fresh deboned and marinated chicken meat was cooked by electric-pan frying or charcoal broiling and subsequently irradiated with a dose of 0, 10 and 20 kGy. Volatile components from *Dakgalbi* were analyzed using solid phase micro-extraction gas chromatography - mass spectrometry (SPME GC-MS) and identified through the comparison of mass spectrum database. SPME GC-MS analysis shows that a total of 32 volatiles were identified. Among them, aldehydes such as hexanal and octanal, which have relevance to off-flavors such as green, paint, metallic, bean and rancid were detected in *Dakgalbi* cooked by both methods. However, the contents were less detected in the *Dakgalbi* cooked by charcoal broiling than in the *Dakgalbi* cooked by electric-pan frying. Gamma-irradiation caused the change in the formation of these aldehydes in cooked *Dakgalbi*. The irradiation significantly increased the contents of hexanal and octanal in *Dakgalbi* cooked by electric-pan frying and a similar increase was found in *Dakgalbi* cooked by charcoal broiling. However, the contents of the off-flavors were much less in the latter. The results suggest that charcoal broiling might be more effective than electric-pan frying for the reduction of the contents of off-flavor such as hexanal and octanal increased in *Dakgalbi* by gamma-irradiation.

Key words: *Dakgalbi*, gamma-irradiation, hexanal, octanal, off-flavor

Introduction

Dakgalbi is a popular chicken-based food in Korea. This food is made by cooking marinated chicken with lots of vegetables such as cabbage, sweet potatoes, scallions, onions; rice cake (*Tteok*); Korean chili paste (*Gochugang*). The common way to prepare *Dakgalb*i is stir-frying and charcoal broiling. Stir-frying is to cook marinated *Dakgalbi* with those ingredients on an iron pan and charcoal broiling is directly to grill *Dakgalbi* with the ingredients on charcoal fire. However, as *Dakgalbi* is commercially prepared, stir-frying is becoming more general than charcoal broiling. As *Dakgalbi* is prepared using high-protein chicken, and lots of high-dietary fiber and low-calorie vegetables, it is known to be a healthy food.

However, despite its popularity, *Dakgalbi* has a short shelf life as a cooked food. One of these reasons is due to the initial bacterial contamination level of the chicken and red pepper paste. Korean chili paste in particular has been reported to be contaminated with a high level of thermo-resistant bacteria such as *Bacillus* spp. (over 6 Log CFU/g of red pepper paste) (Yoon *et al.,* 2009). Therefore, the control of bacterial contamination on preparing *Dakgalbi* becomes a critical factor affecting its quality and hygienic safety.

Ionizing radiation is an effective food processing technology that can prevent food poisoning against harmful pathogens (Rajkowski and Thayer, 2000; Serrano *et al*., 1997). Unlike heat treatment, this technology can expand the shelf life of foods by eliminating bacteria present in foods without changing the nutritional value (Ahn *et al*., 2004; Farkas, 2006; WHO, 1999). Numerous studies using chicken demonstrate that ionizing radiation can improve the shelf life with no negative effects on their nutritional and sensory properties (Gomes *et al*., 2003; Javanmard *et al*., 2006; Mckay *et al*., 1997). Recent research performed by our group also showed that gamma-irradiation enhances the shelf life of cooked *Dakgalbi* (Yoon *et al*., 2012). However, as ionizing radiation can induce the generation

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of radiation-specific off-flavor in foods (Yook *et al*., 1998), it requires a combination with other food processing methods to alleviate the formation of the off-flavor.

In this study, the effect of gamma-irradiation on the change of volatile compounds in *Dakgalbi* cooked by charcoal broiling and the effects of the cooking condition on the formation of off-flavors in *Dakgalbi* were investigated.

Material and Methods

Sample preparation

Deboned fresh chicken was purchased from Harim Co. (Korea). As materials for the preparation of *Dakgalbi* sauce, *Gochugang* (CJ Co., Korea), crushed garlic (Dadam, CJ Co., Korea), soy sauce (Monggo Co., Korea), oligo-saccharides (CJ Co., Gyeonggido), saccharide (CJ Co., Gyeonggido), curry powder and turmeric powder (E-mart, Korea) were procured from a local market in Jeongeup-si in South Korea. Carboxen-polydimethylsiloxan (CAR/PDMS) fiber for SPME GC-MS analysis was obtained from Supelco Co. (USA).

Preparation of *Dakgalbi*

The chicken was diced into small pieces (2×5 cm) and cooked after marinating with *Dakgalbi* sauce composed of 45% (w/v) red pepper paste, 22.5% (w/v) water, 10% (w/v) corn syrup, 5% (w/v) garlic, 5% (w/v) soy sauce, 5% (w/v) sugar, 4% (w/v) ginger, 2.5% (w/v) turmeric powder and 1% (w/v) curry powder. To be exact, 1,200 g of diced chicken was seasoned with 300 g of *Dakgalbi* sauce for 2 h at 4°C. The marinated *Dakgalbi* was then divided into 2 aliquots. One was used for the electric-pan frying and other was for the charcoal broiling. For the electric-pan frying (NU-VUE-3 Cooker, USA), the aliquot was cooked for 12 min until its central temperature achieved 70°C. The other aliquot was charcoal-broiled on a spit (DK-E8-003; Daeil Co., Korea) for 12 min until its central temperature achieved 70°C. The cooked *Dakgalbi* samples were cooled under room temperature and then immediately aseptically vacuum-packed in sterilized aluminum-laminated vacuum bags (low-density polyethylene; meting point, 120°C; density, 0.92 g/cm^3) and followed by freezing at -20°C in refrigerator. The electric-pan-fried *Dakgalbi* sample was used as a control for comparing only the contents of hexanal and octanal formed in charcoal-broiled *Dakgalbi* sample.

Gamma irradiation

Gamma irradiation of cooked *Dakgalbi* was carried out using a cobalt-60 irradiator (IR-221; MDS Nordion International Co. Ltd., Canada) at the Advanced Radiation Technology Institute of the Korea Atomic Energy Research Institute (Korea). The frozen *Dakgalbi* samples were irradiated with absorbed doses of 10 and 20 kGy (10 kGy/h) in a dry-ice packed box. The determination of the absorbed dose of irradiated cooked *Dakgalbi* was performed using a 5 mm diameter alanine dosimeter (Bruker Instruments, Germany), calibrated against an international standard set by the International Atomic Energy Agency (Austria). The error of the total adsorbed doses in the *Dakgalbi* samples was within 2%. The irradiated samples were kept at -20°C again prior to SPME GC-MS analysis.

Extraction of volatile compounds from *Dakgalbi*

Dakgalbi samples were finely chopped with a knife, and 2 g of each sample was placed into a 20 mL of headspace vial, and sealed with a septum and an aluminum cap. Carboxen-polydimethylsiloxan (CAR/PDMS, 75 mm) fiber was used for the extraction of volatile components. The fiber was preconditioned before the analyses according to the instructions by the manufacturer (Supelco Co., USA).

Volatiles were extracted by exposing CAR/PDMS fiber to the headspace of the sample vial maintained at 50°C for 20 min. For the thermal desorption, the fiber was immediately inserted into GC injector, and a desorption time of 1.5 min at 250°C was used in split mode (10:1).

Analysis of GC-MS

Volatile components from *Dakgalbi* were analyzed using GC (Varian Star 3400 CX; Agilent Technology Inc., USA) fitted with an HP-5S capillary column (crosslinked 5% diphenyl and 95% dimethylpolysiloxane; 30 m, 0.32 mm inner diameter, 0.32 mm film thickness; Agilent Technology Inc., USA). The GC oven temperature program consisted of 32°C for 2 min, 32-70°C at 2°C/min, 70-200°C at 9°C/min, and 200°C for 2 min. Helium was used as a carrier gas with a constant column flow rate of 1 mL/min. The mass spectrometric detector (Varian Saturn 2000; Agilent Technology Inc., USA) was operated in electron impact (EI) mode at 70 eV in a range of 15-210 amu. The tentative identifications were based on comparing the mass spectra of the unknown compounds with those in the Wiley mass spectral library (Registry of mass spectral data, 6^{th} edition, USA).

Statistical Analysis

All *Dakgalbi* samples were repeated five times for GC-

MS analysis. Data sets of the contents of hexanal and octanal were analyzed by the Student's *t* test. Differences between the contents according to cooking methods considered significant at $p<0.05$.

Results and Discussions

Changes of volatile compounds in gamma-irradiated charcoal-broiled *Dakgalbi*

To investigate the effect of charcoal broiling on the changes of volatile compounds in *Dakgalbi* according to gamma irradiation, *Dakgalbi* was charcoal-broiled and irradiated with absorbed doses of 0, 10 and 20 kGy. SPME GC-MS analysis revealed that a total of 32 volatile compounds are identified as the major flavoring components of *Dakgalbi* (Fig. 1), including sulfur compounds such as allyl mercaptan, 3,3'-thiobis-1-propene, 4-thia-1,6-heptadiene and allyl disulfide, aromatic compounds such as toluene, benzaldehyde and *o*-cymene, low molecular compounds such as pellandrene, carene, cryophillene, cedr-8-ene and tumorone, degraded compounds of lipids such as methylbutanal and hydroxybutanone, and hydrocarbons such as hexane, nonane, heptanal, octanal, nonanal, terpinene and tetradecene formed from lipids by the adsorption of energy during cooking.

Gamma-irradiation increased or decreased these volatiles formed in charcoal-broiled *Dakgalbi*. As the absorbed doses increased from 0 kGy to 20 kGy, the volatiles such as octanal, n-nonanal, (E)-3-tetradecene, n-tetradecene and (Z)-cyclodecene proportionally increased, while the volatiles such as allyl mercaptan, 3-hydroxy-2-butanone, 4-thia-1,6-heptadiene, gamma-terpinene and allyl disulfide decreased (Table 1). Cha and Cadwallader (1995) has reported that gamma-irradiation applied to chicken enhanced the generation of hydrocarbons, aromatics, aldehydes, ketones and other volatile low molecules, and among these volatiles, hydrocarbons were the most abundant. However, these hydrocarbons do not affect food flavor as they have a high odor threshold. Our results also demonstrated that gamma-irradiation increased the hydrocarbons such as hexane, nonane, heptanal, octanal, nonanal, teripnene, tetradecene and cyclodecene formed in charcoal-broiled *Dakgalbi* (Table 1). This is attributed to the generation of hydrocarbons by high ionizing energy

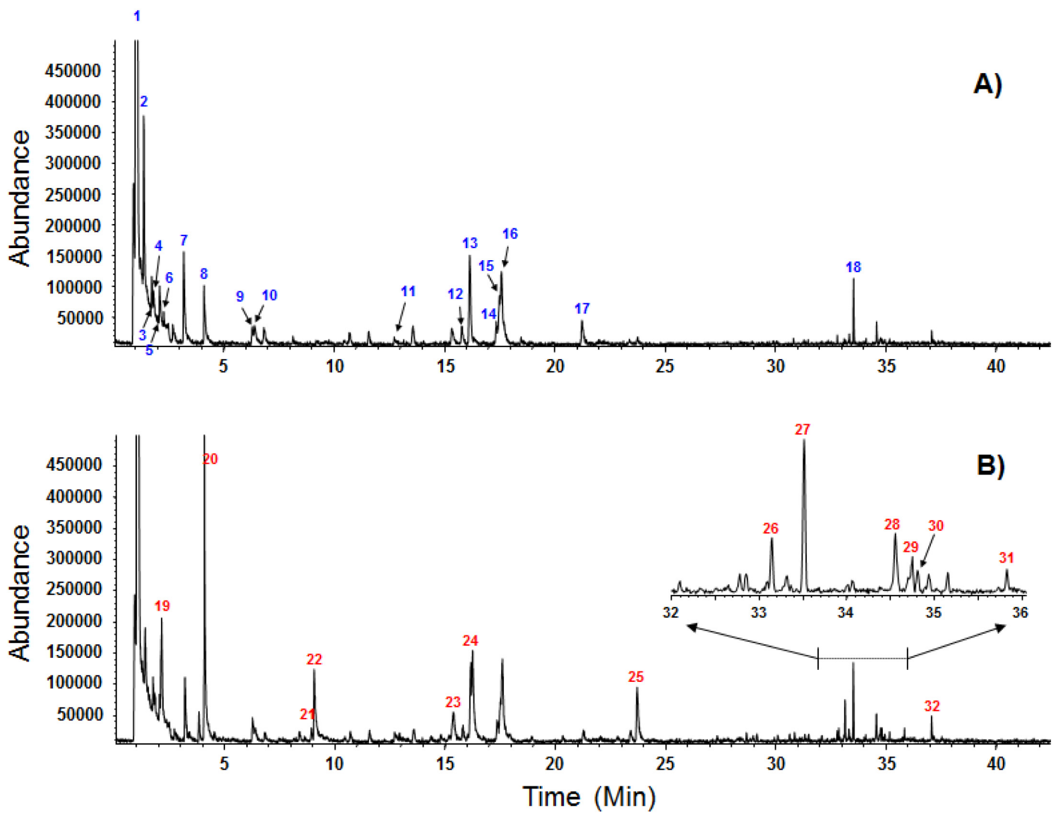

Fig. 1. SPME-GC chromatograms of cooked *Dakgalbi* by charcoal broiling after irradiation. (A) Non-irradiated *Dakgalbi* cooked by charcoal broiling, (B) 20 kGy gamma-irradiated *Dakgalbi* cooked by charcoal broiling.

Table 1. Changes of volatile compound contents in cooked *Dakgalbi* by charcoal broiling after irridiation by SPME GC-MS analysis

Peak No.	Compound	Peak Area[a]	kGy[b]			Remark
			0	10	20	
1	Ethanol	45,943,601 ± 8,421,570	1.0	1.00	1.30	
2	Allyl mercaptan	10,944,533 ± 4,006,218	1.0	0.79	0.89	
3	3-Methyl-butanal	2,720,635 ± 317,024	1.0	0.96	1.53	↑
4	2-Methyl-butanal	2,336,992 ± 242,394	1.0	1.09	1.40	↑
5	Acetoin	3,293,222 ± 197,508	1.0	1.19	0.86	
6	3-Hydroxy-2-butanone	33,387,627 ± 5,619,084	1.0	0.32	-	↓
7	Toluene	3,495,281 ± 521,730	1.0	1.36	1.30	
8	2-3-Butandiol	4,985,634 ± 361,085	1.0	0.76	0.17	↓
9	3,3'-Thiobis-1-propene	765,745.1 ± 81,007	1.0	2.51	2.62	↑
10	4-Thia-1,6-heptadiene	630,278 ± 74,806	1.0	2.80	2.25	↑
11	Benzaldehyde	391,419.3 ± 49,567	1.0	2.79	3.34	↑
12	1-Phellandrene	1,588,554 ± 380,176	1.0	1.19	1.10	
13	Delta-3-carene	5,794,759 ± 842,721	1.0	1.3	1.18	
14	*o*-Cymene	1,358,148 ± 188,064	1.0	1.41	1.60	↑
15	Gamma-terpinene	3,840,498 ± 251,094	1.0	1.28	1.38	
16	DL-limonene	7,592,047 ± 541,231	1.0	1.41	1.63	↑
17	Allyl disulfide	1,746,814 ± 218,022	1.0	0.48	0.70	↓
18	*Trans*-cryophyllene	376,360.9 ± 91,874	1.0	1.49	1.53	↑
19	1-(methythio)-(E)-1-propene	3,293,222 ± 618,041	1.0	1.28	0.79	
20	Hexanal	2,147,523 ± 117,029	1.0	5.71	10.1	↑
21	N-Nonane	-[c]	-	New[d]	New	↑
22	Heptanal	576,388 ± 81,054	1.0	4.75	9.42	↑
23	1-Decene	1,746,618 ± 381,050	1.0	1.83	2.38	↑
24	Octanal	1,304,408 ± 90,745	1.0	2.93	6.27	↑
25	N-Nonanal	-	-	New	New	↑
26	(E)-3-tetradecene	-	-	New	New	↑
27	Caryophyllene	1,973,637 ± 310,861	1.0	1.79	1.74	↑
28	AR-curcumene	829,692 ± 88,107	1.0	1.74	2.07	↑
29	Cedr-8-ene	406,914 ± 32,170	1.0	1.68	1.24	
30	N-Tetradecane	177,000 ± 18,940	1.0	1.36	2.41	↑
31	(Z)-cyclodecene	-	-	-	New	↑
32	AR-tumerone	532,321 ± 11,811	1.0	1.70	2.17	↑

[a]Peak area detected in non-irradiated charcoal-broiled *Dakgalbi*.
[b]Relative peak area fold ratio of irradiated *Dakgalbi* to non-irradiated *Dakgalbi*.
[c]Not detected.
[d]Detected as a peak.

such as gamma-ray, which have one (Cn-1) or two (Cn-2:1) carbon atoms less than the predominant fatty acids present in foods (Spiegelberg *et al.*, 1994).

In general, fat present in meats contains various fatty acids such as palmitic acid, stearic acid, oleic acid and linoleic acid (Kim *et al.*, 1999), and ionizing radiation can generate hydrocarbons having variable carbon chain lengths. Therefore, hydrocarbons have been suggested as an index substance for discriminating irradiated foods (Kim *et al.*, 2002). However, as hydrocarbons can be generated by the cleavage of acyl-oxygen linkage or acyloxy-methylene linkage of glycerol moiety ester bonded with fatty acids, or by the cleavage of covalent linkage between carbon atoms in fatty acids (LeTellier and Nawar, 1972; Nawar, 1978; Patterson and Stevenson, 1995), the mechanism of forming hydrocarbons in fat by the adsorption of energy is presumed to be rather extensive.

Irradiation can induce the formation of aromatic compounds such as alkyl-benzenes, naphthalenes and phenols (Cha *et al.,* 2000). Although the mechanism of forcming alkyl-benzenes is not clear, Watanabe and Sato (1971) has shown that various alkyl-benzenes was formed in swallow fried beef, and toluene, xylene and benzene derivatives have been reported to be formed from carotenoids (Borenstein and Bunnell, 1966; Pippen *et al.,* 1969). Jo and Ahn (2000) have also suggested that bovine serum albumin, gelatin and myofibril proteins might be precursors of the benzene derivatives. Supporting this, our results have shown that toluene, benzaldehyde and *o*-cymene were detected in non-irradiated charcoal-broiled *Dakgalbi*, and gamma-irradiation increased the generation of benzaldehyde (Table 1). However, toluene was not affected by gamma-irradiation.

Ho and Chen (1993) and Hansan *et al.* (1987) have shown that the irradiation of fresh chicken meats induced the formation of aldehydes. These aldehydes are known to be major volatile components formed by the auto-oxidation of unsaturated fatty acids and off-flavors such as green, paint, metallic, bean and rancid (Cha *et al.,* 2000). The cause of the formation of aldehydes in irradiated charcoal-broiled *Dakgalbi* might be due to the acceleration of the oxidation of unsaturated fatty acids by free radicals generated during irradiation (Yook *et al*., 1998) and the increase of lipid rancidity by the cooking temperature and the time of charcoal broiling (Park *et al*., 2005). In this study, gamma-irradiation increased the aldehydes such as 3-methyl-butanal, 2-methyl-butanal, 3-hydroxy-2-butanone, hexanal, heptanal, octanal, 1-decene, and n-nonane in charcoaled-broiled *Dakgalbi* (Table 1). Among the aldehydes, hexanal is known as an index substance in the oxidation and deterioration of foods and is formed by the auto-oxidation of 13-hydroperoxide and 15-hydroperoxide derived from unsaturated linoleic acid and arachidonic acid (Ho and Chen, 1993).

Volatiles such as caryophyllene, tumorone, curcumene, cymene and delta-3-carene, which were identified in this study, are the compounds originating from seasonings used for the preparation of charcoal-broiled *Dakgalbi*. In particular, caryophyllene and delta-3-carene are volatile compounds of black pepper (Plessi *et al*., 2002), and tumorone and curcumene are the principle ingredients of tumoric, which is a base of curry powder (Park *et al*., 1991). Cymene is a volatile derived from plants (Block *et al*., 1992).

Effects of cooking method on the formation of hexanal and octanal in irradiated *Dakgalbi*

Next, to investigate the effect of the cooking method on the formation of volatile compounds in irradiated *Dakgalbi*, the samples were stir-fried in an electric-pan or

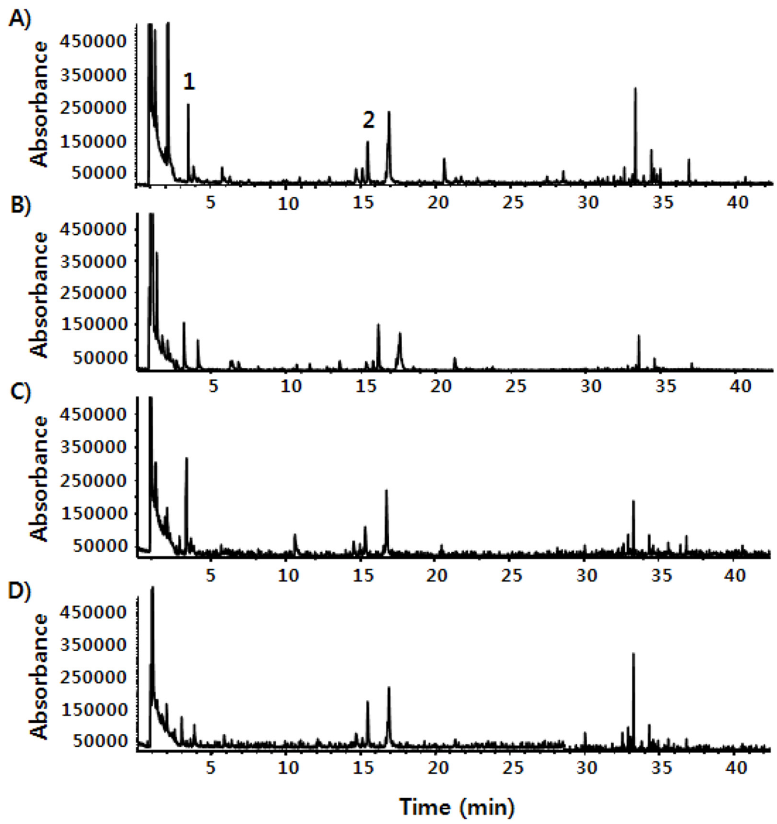

Fig. 2. SPME-GC chromatograms of volatile compounds from cooked *Dakgalbi* by electric-pan frying or charcoal broiling after irridiation. (A) Non-irradiated *Dakgalbi* cooked by electric-pan frying, 1. Hexanal, 2. Octanal, (B) Non-irradiated *Dakgalbi* cooked by charcoal broiling, (C) 20 kGy gamma-irradiated *Dakgalbi* cooked by electric-pan frying, (D) 20 kGy gamma-irradiated *Dakgalbi* cooked by charcoal broiling.

Table 2. Changes of volatile compound contents in cooked *Dakgalbi* by electric-pan frying or charcoal broiling after irridiation by SPME GC-MS analysis

PeakNo.	Compound	Cooking Methods[a]		Remarks[b]
		Electronic-pan frying	Charcoal broiling	
A) non-irradiated *Dakgalbi*				
1	Hexanal	4,007,907 ± 280,176[A]	2,832,296 ± 118,972[B]	0.71
2	Octanal	25,477,272 ± 987,526[A]	10,787,822 ± 446,397[B]	0.42
B) 20 kGy gamma-irradiated *Dakgalbi*				
1	Hexanal	42,674,782 ± 1,897,624[A]	5,514,941 ± 882,591[B]	0.13
2	Octanal	69,438,262 ± 2,591,428[A]	38,418,351 ± 3,948,627[B]	0.55

[a]Peak area detected in irradiated cooked *Dakgalbi*.
[b]Relative peak area fold ratio of charcoal broiling to electric-pan frying.
[A-B]Mean values within a column follow by the different letter are significantly different (p<0.05).

charcoal-broiled, and gamma-irradiated with an absorbed dose of 20 kGy. Among the volatile compounds identified using SPMC GC-MS analysis, the significant change of aldehydes such as hexanal and octanal were detected in both non-irradiated electric-pan-fried and charcoal-broiled *Dakgalbi* and the contents were 0.71- and 0.42-times less in the charcoal-broiled *Dakgalbi* than in the electric-pan-fried *Dakgalbi*, respectively (Fig. 2, Table 2A).

Gamma-irradiation caused a dramatic change in the contents of these volatiles in *Dakgalbi*. Gamma-irradiation increased the contents of hexanal and octanal in electric-pan-fried *Dakgalbi*, and a similar effect was also observed in charcoal-broiled *Dakgalbi* (Table 2A and 2B). However, the contents of hexanal and octanal were 0.13- and 0.55-times less in the charcoal-broiled *Dakgalbi* than in the electric-pan-fried *Dakgalbi*, respectively (Table 2B). This indicates that although gamma-irradiation causes the increase of the contents of these volatiles in *Dakgalbi*, a charcoal broiling method can reduce increasing range of aldehydes formed during cooking. The reason for this might be increasing the removal of lipid content in broiled *Dakgalbi*, which is cooked directly above charcoal fire, or facilitating the rapid decomposition of aldehydes such as hexanal and octanal formed during the cooking, as the temperature of charcoal broiling is much higher than that of electric-pan frying.

Recently, Yoon (2012) reported that the volatiles like aldehydes formed during gamma-irradiation can influence the sensory quality of *Dakgalbi*. The results showed that gamma-irradiation deteriorated the sensory quality of electric-pan-fried or charcoal-broiled *Dakgalbi*, and the effect was irradiating dose-dependent. However, the worsening of the sensory quality was significantly much less in charcoaled-broiled *Dakgalbi* than in electric-pan-fried *Dakgalbi*. Among the tested sensory evaluations, off-flavor was found to mostly affect the overall acceptability of *Dakgalbi* (Yoon, 2012), implying the possible involvement of aldehydes on its sensory quality. This indicates that charcoal broiling can be a better cooking method than electric-pan frying for the improvement of the organoleptic property of *Dakgalbi*.

As a conclusion, these results suggest that charcoal broiling might be more effective than electric-pan frying for the relative reduction of the contents of hexanal and octanal increased by gamma-irradiation.

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