Evaluation of Radiation-induced Compounds in Irradiated Raw or Cooked Chicken Meat during Storage

A.S. Leaflet R2764

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Summary and Implications

The concentrations of hydrocarbons, 2-alkylcyclobutanones, and sulfur volatiles in irradiated (0, 5 kGy) chicken meats (raw, pre-cooked, and irradiated-cooked) were analyzed after 0 and 6 months of frozen storage (-40 ^{o}C) under oxygen permeable packaging conditions. Two hydrocarbons [8-heptadecene ($C_{17:1}$) and 6,9-heptadecadiene ($C_{17:2}$)], two 2-alkylcyclobutanones [2-dodecylcyclobutanone (DCB) and 2-tetradecylcyclobutanone (TCB)], and dimethyl disulfide were determined as radiation-induced detection markers in the irradiated raw and cooked chicken meats. Although, irradiated-cooked samples produced less hydrocarbons and 2-alkylcyclobutanones than pre-cooked irradiated ones, the amount of individual hydrocarbons or 2-alkylcyclobutanones was still sufficient enough to detect radiation treatment even after 6 months of storage at -40 ^{o}C. Among sulfur volatiles, only dimethyl disulfide were found in meat after 6 months of storage indicating it has potential to be used an irradiation detection marker for frozen-stored meats under oxygen permeable packaging conditions.

Introduction

Irradiation improves the safety and shelf life of food products by controlling microorganisms. Irradiation of raw meat or poultry has potential to enhance community health by preventing food-borne diseases. Electrons with energies up to 10 MeV, X-rays with energy up to 5.0 MeV, and gamma rays from cobalt-60 and cesium-137 are all allowed by the U.S. Food and Drug Administration (FDA) for food irradiation. Many other countries have also approved irradiation as an efficient technology to control pathogens and parasites, and to extend the shelf-life of products. However, most countries have various regulations with mandatory labeling requirements for irradiated foods. In this scenario, effective and validated detection methods for irradiated food products are very important for the enforcement of laws and regulations and to facilitate international trades.

The formation of long-chain hydrocarbons and 2-alkylcyclobutanones (2-ACB) in irradiated food products were standardized by the European Committee for Normalization (CEN) as reference methods for the detection of irradiated foods. Irradiation of fats or oils causes loss of an electron from acyl-oxygen bond in fatty acids, followed by a rearrangement process to produce 2-alkylcyclobutanones such as 2-dodecylcyclobutanone (DCB) and 2-tetradecylcyclobutanone (TCB) specific to their parent fatty acids. 2-Dodecylcyclobutanone (DCB) and 2-tetradecylcyclobutanone (TCB) could be used as radiation-induced markers because of their absence in non-irradiated foods. However, some studies found a higher concentration of 2-(tetradec-5'-enyl) cyclobutanone specific to oleic acid in irradiated foods.

In fat-containing foods, the cleavage of chemical bonds in fatty acids upon irradiation generates hydrocarbons. Hydrocarbons produced by irradiation have one carbon atom less than the parent fatty acid (Cn-1) or two carbon atoms less and an additional double bond at position one (Cn-2, 1-ene), which are predominately generated by the breakage of fatty acid moieties of triglycerides, mainly at the alpha and beta positions of carbonyl groups.

Irradiation treatments produce very reactive hydroxyl radicals in aqueous or oil emulsion systems that can initiate lipid oxidation to form a lipid radical by removing a hydrogen atom from fatty acyl chain of a polyunsaturated fatty acid (PUFA). After a series of reactions, the lipid radicals produce lipid hydroperoxides that breaks into various volatile compounds including aldehydes, ketones, hydrocarbons, and sulfur compounds. Sulfur-volatile compounds in irradiated meat are mainly formed by the radiolytic degradation of sulfur-containing amino acids (e.g., cystein and methionine).

The objective of this study was to determine irradiation-induced chemical changes in irradiated raw or cooked chicken meat during frozen storage. The irradiation-induced compounds were also evaluated as possible detection markers for irradiated chicken meat.

Materials and Methods

Fresh chicken thighs were ground and vacuum-packaged in an oxygen-impermeable nylon/polyethylene bags (O_2 permeability, 9.3 mL O_2/m^2/24 hr at 0 ^{o}C). Five treatments were prepared depending on cooking and irradiation conditions: 1) nonirradiated raw chicken meat (uncooked-0 kGy), 2) irradiated raw chicken meat (uncooked-5 kGy), 3) nonirradiated cooked chicken meat (cooked-0 kGy), 4) precooked irradiated chicken meat (cooked-5 kGy), and 5) irradiated cooked chicken meat (5 kGy-cooked). Uncooked meat samples were packaged in oxygen permeable nylon bags (2,300 mL/m^2/24 h). For cooked meat, samples were vacuum-packaged in nylon/polyethylene bags (O_2 permeability, 9.3 mL O_2/m^2/24 h at 0 °C), and then cooked. Cooking was done in an 85 ^{o}C water bath to an internal temperature of 75 ^{o}C. Meat

samples were irradiated at 5 kGy using a Linear Accelerator (Circe IIIR). Samples were analyzed at 0 day and after 6 months of storage at -40 ^{o}C under oxygen permeable packaging conditions. The fatty acid composition was analyzed using a gas chromatograph. A gas chromatograph/mass spectrometer (GC/MS) was used to analyze hydrocarbons and 2-alkylcyclobutanones.

Statistical Analysis

Data were analyzed using the generalized linear model procedure of SAS software (SAS Institute Inc., 1995). The Student-Newman-Keul's multiple range test was used to compare the mean values of the treatments. Mean values were reported ($p < 0.05$).

Results and Discussion

The fat of raw chicken meat had 38.64% oleic acid, 25.24% palmitic acid, 16.89% linoleic acid, 5.32% stearic acid, and 1.00% arachidonic acid. The 5 kGy irradiation before or after cooking had no significant change on the fatty acid composition of chicken meat. 8-Heptadecene ($C_{17:1}$) and 6,9-heptadecadiene ($C_{17:2}$) are derived from oleic acid and linoleic acid, respectively, and are found in irradiated fat-containing foods. Palmitic, stearic, and oleic acids are the major fatty acid precursors that generate radiolytic products of 2-DCB, 2-TCB, and 2-(tetradec-5'-enyl) cyclobutanone, respectively, upon irradiation.

Two hydrocarbons (8-heptadecene and 6,9-heptadecadiene) were found only in irradiated samples (Table 1). Therefore, 8-heptadecene and 6,9-heptadecadiene could be used as markers for irradiated chicken meat. 1-Hexadecene was found not only in irradiated but also cooked chicken meat. Thus, 1-hexadecene cannot be used as an irradiation marker for chicken meat.

New long-chain hydrocarbons could be produced in vegetable oils and in animal products such as roasted chickens. However, 8-heptadecene and 6,9-heptadecadiene were not detected in any of the non-irradiated chicken meat samples. 8-heptadecene ($C_{17:1}$) showed more storage stability than 6,9-heptadecadiene ($C_{17:2}$) in all the irradiated samples. The concentration of 6,9-heptadecadiene ($C_{17:2}$) declined in raw and irradiated-cooked samples during the storage, but these were still detectable even after 6 months of storage. 2-Dodecylcyclobutanone (2-DCB) and 2-tetradecylcyclobutanone (2-TCB) were found in all irradiated chicken meat samples (Table 2). The concentration of 2-DCB (derived from palmitic acid) was higher than that of 2-TCB (derived from stearic acid) because of the relative concentrations of the parent fatty acids in the samples. 2-DCB concentration in gamma-irradiated beef (5.1 kGy) was about 0.45 μg/g fat (0.068 mg/kg. 2-DCB only found in irradiated meats, and 2-DCB and 2-TCB were used as irradiation markers in liquid whole eggs. 2-(5'-Teradecenyl) cyclobutanone (2-TeCB) was detected in non-irradiated samples and irradiation had little effect on its concentration. In irradiated raw meat samples, the amounts of 2-ACBs showed great stability during the storage. The concentration of 2-DCB and 2-TCB declined over time, but detectable amounts of them were still remaining in meat after 6 months of frozen storage.

Sulfur volatile compounds were detected only in irradiated meat samples, where pre-cooked samples showed the highest amounts of all (Table 3). The changes in a relative composition of sulfur volatiles in a meat during storage also affect the overall sensory characteristics of meat samples. Sulfur volatiles in irradiated meats are mainly dependent upon the storage conditions and easily disappeared under aerobic storage conditions. In this study, only dimethyl disulfide exhibited long-term stability, showing the potential of using it as an irradiation detection marker for chicken meat stored for 6 months under oxygen permeable packaging conditions.

Table 1. Concentration of irradiation-induced hydrocarbons in beef during storage at -40 °C.

Hydrocarbons	Storage time (mo)	Raw meat 0 kGy	Raw meat 5 kGy	Cooked before IR 0 kGy	Cooked before IR 5 kGy	Cooked after IR 5 kGy
		---------- (μg/g fat) ----------				
1-Tetradecene($C_{14:1}$)	0	-[k)]	9.30^{bx}	-	12.23^{ax}	8.06^{cx}
1-Tetradecene($C_{14:1}$)	6	-	6.37^{by}	-	7.35^{ay}	7.58^{ay}
Pentadecane($C_{15:0}$)	0	1.61^{cx}	7.12^{bx}	1.35^{cx}	14.39^{ax}	7.86^{bx}
Pentadecane($C_{15:0}$)	6	1.46^{dx}	5.10^{cy}	1.04^{ex}	9.9^{ay}	5.78^{by}
1-Hexadecene($C_{16:1}$)	0	0.86^{dx}	6.74^{cx}	1.33^{dx}	9.91^{bx}	19.61^{ax}
1-Hexadecene($C_{16:1}$)	6	-	4.47^{cy}	0.56^{dx}	5.97^{by}	15.80^{ay}
6,9-Heptadecadiene($C_{17:2}$)	0	-	2.51^{ax}	-	2.54^{ax}	1.73^{bx}
6,9-Heptadecadiene($C_{17:2}$)	6	-	1.82^{ax}	-	1.98^{ay}	1.49^{ax}
8-Heptadecene($C_{17:1}$)	0	-	7.00^{ax}	-	6.94^{ax}	4.53^{bx}
8-Heptadecene($C_{17:1}$)	6	-	5.05^{by}	-	5.75^{ay}	3.07^{cy}
n-Heptadecane($C_{17:0}$)	0	2.88^{bx}	5.43^{ax}	3.44^{bx}	6.18^{ax}	6.17^{ax}
n-Heptadecane($C_{17:0}$)	6	0.37^{cy}	3.83^{by}	0.38^{cy}	5.24^{ax}	5.51^{ax}

[a-d]Different superscript letters within a column of the same storage day are significantly different ($p < 0.05$); n = 3.

[k]Not detected.

[x,y]Different superscript letters within a row are significantly different ($p < 0.05$).

Table 2. Concentration of irradiation-induced hydrocarbons in pork during storage at -40 °C.

Hydrocarbons	Storage time (mo)	Raw meat 0 kGy	Raw meat 5 kGy	Cooked before IR 0 kGy	Cooked before IR 5 kGy	Cooked after IR 5 kGy
				(μg/g fat)		
1-Tetradecene($C_{14:1}$)	0	0.42^{bx}	5.63^{ax}	0.3744^{bx}	5.50^{ax}	5.10^{ax}
1-Tetradecene($C_{14:1}$)	6	-[k])	4.81^{ay}	-	4.23^{cy}	4.47^{bx}
Pentadecane($C_{15:0}$)	0	0.59^{cx}	6.13^{ax}	1.41^{cx}	4.50^{bx}	5.15^{bx}
Pentadecane($C_{15:0}$)	6	-	5.35^{ax}	1.33^{dx}	2.97^{cy}	4.03^{by}
1-Hexadecene($C_{16:1}$)	0	-	3.19^{ax}	1.93^{bx}	3.22^{ax}	3.12^{ax}
1-Hexadecene($C_{16:1}$)	6	-	2.89^{ax}	1.80^{bx}	3.25^{ax}	1.06^{by}
6,9-Heptadecadiene($C_{17:2}$)	0	-	3.37^{ax}	-	3.51^{ax}	3.09^{ax}
6,9-Heptadecadiene($C_{17:2}$)	6	-	1.17^{cy}	-	1.82^{ay}	1.55^{by}
8-Heptadecene($C_{17:1}$)	0	-	6.25^{ax}	-	5.57^{ax}	6.62^{ax}
8-Heptadecene($C_{17:1}$)	6	-	4.57^{ay}	-	4.11^{ax}	4.20^{ay}
n-Heptadecane($C_{17:0}$)	0	0.86^{cx}	4.13^{ax}	1.34^{cx}	1.02^{cx}	2.16^{bx}
n-Heptadecane($C_{17:0}$)	6	0.20^{cy}	4.12^{ax}	0.37^{bcy}	0.62^{bcx}	0.70^{by}

[a-d]Different superscript letters within a column of the same storage day are significantly different ($p < 0.05$); n = 3.

[k]Not detected.

[x,y]Different superscript letters within a row are significantly different ($p < 0.05$).

Table 3. Concentration of irradiation-induced 2-Alkylcyclobutanones in beef during storage at -40 °C.

Cooking treatment	IR dose (kGy)	2-DCB 0 month	2-DCB 6 months	2-TCB 0 month	2-TCB 6 months	2-TeCB 0 month	2-TeCB 6 months
				(μg/g fat)			
Raw meat	0	-[k])	-	-	-	1.28^{cx}	0.55^{cy}
Raw meat	5	1.93^{ax}	0.96^{by}	0.51^{ax}	0.14^{ay}	5.81^{ax}	2.97^{ay}
Cooked before IR	0	-	-	-	-	0.67^{cx}	0.42^{cy}
Cooked before IR	5	1.63^{ax}	1.06^{ax}	0.25^{bx}	0.05^{bx}	4.39^{bx}	2.76^{ax}
Cooked after IR	5	0.76^{bx}	0.23^{cx}	0.13^{bx}	0.10^{abx}	2.09^{cx}	1.77^{bx}

[a-c]Different superscript letters within a column of the same storage day are significantly different ($p < 0.05$); n = 3.

[k]Not detected.

[x,y]Different superscript letters within a row are significantly different ($p < 0.05$).

Table 4. Concentration of irradiation-induced 2-alkylcyclobutanones in pork during storage at -40 °C.

Cooking treatment	IR dose (kGy)	2-DCB		2-TCB		2-TeCB	
		0 mo	6 mo	0 mo	6 mo	0 mo	6 mo
		---------- (μg/g fat) ----------					
Raw meat	0	-[k)]	-	-	-	0.25^{ax}	0.22^{cx}
Raw meat	5	0.41^{ax}	0.08^{by}	0.44^{ax}	0.11^{ay}	0.82^{ax}	0.40^{cy}
Cooked before IR	0	-	-	-	-	0.30^{ax}	0.27^{cx}
Cooked before IR	5	0.45^{ax}	0.25^{ax}	0.65^{ax}	0.42^{ax}	0.88^{ax}	0.68^{bx}
Cooked after IR	5	0.46^{ax}	0.23^{ay}	0.38^{ax}	0.19^{ay}	0.54^{ax}	0.95^{ax}

[a-c]Different superscript letters within a column of the same storage day are significantly different ($p < 0.05$); n = 3.

[k]Not detected.

[x,y]Different superscript letters within a row are significantly different ($p < 0.05$).

Table 5. Effect of irradiating beef before or after cooking on sulfur compounds during storage at -40 °C.

Cooking treatment	IR dose (kGy)	Dimethyl sulfide		Dimethyl disulfide		Dimethyl trisulfide	
		0 mo	6 mo	0 mo	6 mo	0 mo	6 mo
		---------- (Total ion counts × 10^4) ----------					
Raw meat	0	721	0	0^{b}	0c	0^{b}	0
Raw meat	5	1064	0	2833^{ab}	2054^{b}	0^{b}	0
Cooked before IR	0	790	0	0^{b}	0^{c}	0^{b}	0
Cooked before IR	5	0	0	2872^{ab}	2255^{b}	0^{b}	0
Cooked after IR	5	662	0	5933^{a}	3835^{a}	485^{a}	0

[a,b]Different superscript letters within a column of the same storage day are significantly different ($p < 0.05$); n = 3.

Table 6. Effect of irradiating pork before or after cooking on sulfur compounds during storage at -40 °C.

Cooking treatment	IR dose (kGy)	Dimethyl sulfide		Dimethyl disulfide		Dimethyl trisulfide	
		0 mo	6 mo	0 mo	6 mo	0 mo	6 mo
		---------- (Total ion counts × 10^4) ----------					
Raw meat	0	907	0	0^{b}	0	0^{b}	0
Raw meat	5	2135	0	3394^{ab}	136	0^{b}	0
Cooked before IR	0	573	0	0^{b}	0	0^{b}	0
Cooked before IR	5	249	0	4317^{a}	44	489^{a}	0
Cooked after IR	5	815	0	3405^{ab}	690	535^{a}	0

[a,b]Different superscript letters within a column of the same storage day are significantly different ($p < 0.05$); n = 3.