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Thermally Produced Flavor Components in the Aroma of Meat and Poultry

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Meat flavor (taste and aroma) is developed by heating the raw product. Thermal degradation of the many classes of chemical compounds in meat results in flavor formation. Conditions influencing the chemical composition of meat, heating procedures, and products formed on heating various

The desirable flavor of meat is developed by the application of heat. Raw meat in general has a salty, metallic, bloody taste and a sweet aroma resembling serum. Raw pork may also have a "sour" smell, and the aroma of lamb or mutton may contain a "basic" note. Application of the proper amount of heat, however, results in the formation or release of odorous reaction products. Animal tissue is composed of a large variety of chemicals that may be involved in flavor development and, although the chemical composition of the tissue may remain essentially constant, small quantitative and qualitative changes could affect flavor. Thus, in regarding the chemistry of meat flavor, we should note some factors influencing potential flavor precursors.

Species differences in flavor precursors must be a major factor in view of the characteristic flavor of beef, pork, lamb, and chicken meat, and some differences in chemical composition among these meats have been noted by many investigators. Within a species flavor characteristics may vary with the strain of the animal and even among individual animals. Gross differences in total nitrogen and fat, in the acid composition of fat, occur in some instances, but the relation of these factors to flavor has not been studied. Various organs meat components are reviewed. None of the compounds identified in meat aroma has been described as uniquely meaty. Recent identifications of interesting boiled beef flavor fractions, however, suggest that progress in elucidating meat flavor may be more rapid than in the past.

and tissues within an animal also vary in flavor produced by heat, and while considerable information is available on the chemical composition at this level, correlation between composition and flavor still has not been achieved. Diet and nutritional status of the animal influence the chemical composition of the tissues. Animals on a high nutritional plane of feeding have more intramuscular fat which is more highly saturated (Lawrie, 1966). Muscle from starving animals, or those on a low nutritional plane, has a lower pH and produces more H<sub>2</sub>S on heating (Johnson and Vickery, 1964). The flavor of beef from animals fattened on grain was scored significantly higher than that from animals finished on grass (Meyer et al., 1960); this experiment, however, did not include chemical analyses of the tissues. Unsaturated fats in the diet induce deposition of unsaturated intramuscular fat in the pig (Lawrie, 1966) and turkey (Klose et al., 1952), but in ruminants (cows and sheep) the unsaturated fats are hydrogenated in the digestive tract (Lawrie, 1966). The digestion of ruminants relies on the action of bacteria, and fatty acids produced by these organisms may modify the lipids of the animal. Harris et al. (1968) reported that meat from conventionally raised chickens had a more characteristic flavor than meat from chickens born and raised in a germ-free environment.

The emotional state of the animal at slaughter affects the chemical composition of the tissue. In an animal that has been exercised or excited prior to slaughter, the glycogen reserve is depleted, and the muscle pH may rise from 5.5, nor-

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mally found in meat to 6.5. There will undoubtedly be changes in the balance of other catabolic products which, to date, have not been determined.

Aging, particularly of beef, is important in the development of flavor precursors. Microbiological and enzymic changes occur in the tissue. Glycogen undergoes glycolysis to form lactic acid and the pH drops to 5.5. Other metabolic acids and compounds are also formed at this time. The proteins are denatured, becoming liable to attack by tissue enzymes with the formation of peptides and free amino acids. Ammonia, H<sub>2</sub>S, and various carbonyls are also produced. Nucleotides are degraded with the eventual increase in the concentration of hypoxanthine and ribose or ribose-phosphate. Caul (1957) reported on the flavor profiles of broiled steaks from aging beef. Hot meat, prepared about 3 hr after slaughter, was sour, metallic, and astringent and had no recognizable beef flavor. Green meat cooked 24 hr after slaughter tasted and smelled more like beef, but the flavor was still metallic and astringent, and the characteristic aroma had not developed. After about 8 days of aging, the final flavor was a blend of sweet and serum, with brothiness and bouquet typical of a desirable steak.

The method of applying heat to meat affects its flavor. The aroma of stewed or braised meat heated at  $100^{\circ}$ C differs from that of the same meat roasted with dry heat at  $190^{\circ}$ C. However, it is still not known whether different compounds are produced under the two conditions of heating or whether the differences are only quantitative. It is interesting to note that when meat is roasted at  $190^{\circ}$ C the interior temperature varies from about  $60^{\circ}$ C for rare meat to  $80^{\circ}$ C for well done meat. The flavor of roast meat, therefore, is derived essentially from the surface.

A series of physical and chemical changes occur during the heating of meat (Hamm, 1966). With the temperature as low as 30°C protein degradation is initiated, and small fragments may be split off of some proteins. Between 35 and 50°C actomyosin fibers unfold, increasing the number of available imidazolium groups, which results in a pH increase from 5.5 to about 6.0. The number of -SH groups also increases as a result of the protein unfolding. Most of the proteins are coagulated between 55 and 80°C. The -SH groups are oxidized to disulfide bonds above 80°C, and at temperatures above 90°C H<sub>2</sub>S is released from the -SH groups of the myofibrillar protein.

Browning begins at about 90°C and increases with time and temperature. The amount of browning is related to the concentration of sugar in the meat (Pearson *et al.*, 1962). On exposure to dry heat evaporation of water from the surface of the meat concentrates tissue constituents, creating conditions that are more favorable for the occurrence of the browning reaction. Adipose tissue and lipids are also affected by the heating process; fat is liquified and, as cell membranes rupture, spreads throughout the tissue, becoming more readily available for chemical reaction.

Although some of the physical effects of the heat applied to meat can be related, chemically, to the aroma components produced, the effects of biological changes are still largely unknown. However, studies have been made on the effect of heat on some of the major components of meat

#### AMINO ACIDS

The thermal degradation of amino acids has not been studied extensively at temperatures attained in cooking meat, but some observations have been reported at temperatures of 300°C and above. Decarboxylation can occur with the formation of the appropriate amine at 300-400°C, while at higher temperatures more varied but less complex substances appeared. Merritt and Robertson (1967) listed the major components formed on pyrolyzing various common amino acids. Several of these, 3-methyl- and 2-methylbutanol from leucine and isoleucine, and 2-methylpropanal from valine, have also been reported as browning reaction products; several of the others have been reported in meat aroma. Benzene, toluene, and ethylbenzene were obtained on pyrolyzing phenylalanine and the corresponding -OH compounds were derived from tyrosine. Imidazole derivatives were formed from histidine (Shulman and Simmonds, 1968) and a variety of nitriles have also been reported (Völlmin et al., 1966). Most interesting, however, is Merritt's observation that the compounds formed on pyrolyzing a peptide depended on the sequence of amino acids. Although only dipeptides were used, this presumably holds true for longer peptides and may explain aromas not obtained on heating model systems of single amino acids.

#### CARBOHYDRATES

Thermal degradation of carbohydrates is important in food processing and has been studied extensively. Gravimetric thermoanalysis of sugars indicated that at 100 to  $130^{\circ}$ C bound water was lost without alteration in molecular structure. With increasing temperature (150–180°C) a molecule of water splits from sugar forming the anhydride, and from about 190 to 220°C a second molecule is removed with formation of furfural from pentoses and hydroxy-methylfurfural from hexoses. These compounds degrade further at higher temperatures (Lorant and Boros, 1965).

Heyns *et al.* (1966) reported that more than 130 compounds were formed from glucose at 300°C and identified 56 of these. Furans were the principle components; a variety of alcohols, carbonyls, and aromatic hydrocarbons was also found. Some compounds contained more than six carbons, and Heyns suggested that these came from polymerization products which formed readily and extensively on heating sugars (Sugisawa and Edo, 1966). Fagerson (1969) has compiled a list of almost 100 volatile compounds identified from heated glucose.

The effect of pH on the composition of thermal degradation products of fructose has been investigated (Shaw *et al.*, 1967, 1968). Thirteen compounds were identified in an acid-catalyzed reaction and 15 compounds were identified as a result of alkali-sugar degradation. While eight furans were formed under acidic conditions, only three (furfuryl alcohol, 5-methyl-2-furfuryl alcohol, and 4-hydroxy-2,5-dimethyl-3(2*H*)-furanone) were formed during alkaline fructose thermal degradation. None of these three were formed under acidic conditions. Acetol and 3-hydroxy-2-butanones were also found in the fructose-base degradation products; the authors demonstrated how cyclic diketones could be formed as secondary reaction products.

Ascorbic acid, a vitamin found in animal tissue as a coenzyme for metabolic activity, is degraded by heat. Tatum *et al.* (1969) identified ten furan derivatives, two lactones, three acids, and 3-hydroxy-2-pyrone. Six of these compounds were also found during the thermal degradation of fructose under acidic or basic conditions.

#### AMINO ACID-CARBOHYDRATE REACTIONS

Although amino acids and carbohydrates react individually to form odorous products, in meat they are in close proximity and interact with each other when heated to form a variety of compounds. Pyrazines have been identified in the aroma of peanuts (Mason *et al.*, 1969; Koehler *et al.*, 1969), cocoa (van Praag *et al.*, 1968), coffee (Bondarovich *et al.*, 1967), and other foods exposed to higher temperatures during processing as well as in model systems in which amino acids were heated with sugars or carbonyl compounds (Koehler and Odell, 1970; Hodge *et al.*, 1969). Pyrazines have been found in meat aroma only recently, however, Wasserman and Zaika (1971) tentatively identified methyl, 2,3-dimethyl, 3-ethyl, 2,5-dimethyl, and 2-methyl-5-propenyl derivatives of pyrazine in pyrolyzed dialyzed aqueous extracts of beef, pork, and lamb. Watanabe and Sato (1971) further identified the 2,6-dimethyl-, 2-ethyl-5-methyl-, 2,5-dimethyl-, and tetramethyl-pyrazines, as well as several pyridine compounds, from shallow-fried beef.

Tonsbeek *et al.* (1968, 1969) recently identified 4-hydroxy-5-methyl-3(2H)-furanone as a component of the flavor of beef broth. This compound can be formed from the reaction of ribose-5-phosphate with pyrrolidone carboxylic acid or taurine.

Browning is a complex interaction between amino acids and dicarbonyl compounds or their precursors, such as the carbohydrates. It may occur through the Amadori reaction pathway and may involve the Strecker degradation of the amino acid to an aldehyde with one less carbon atom. The reactions have been reviewed by Reynolds (1963, 1965) and Hodge (1967). A number of studies on model systems have been reported in which amino acids were reacted with sugars under several conditions of pH, temperature, or sugar classes. Rothe and Voight (1963) reacted xylose with various amino acids and concluded there was no direct relation between color and flavor development. Casey et al. (1965) studied the reaction of glucose, fructose, and ribose and reported more intense and rapid browning with pentose than with hexose sugars. While some interesting odors were obtained, no meat aromas were described. El-Ode et al. (1966) varied pH, temperature, and sugars in the reaction with selected amino acids and reported that at 180°C several of the sugaramino acid combinations had a beef broth or meaty aroma.

Wasserman and Spinelli (1970), working with dialyzed meat extracts and model systems composed of approximately the same concentrations of amino acids and sugars found in the extract, reported that during 30 min of boiling very few changes occurred in the amino acid concentrations of meat extract diffusate; only arginine declined substantially. Among the sugars, the concentration of ribose decreased. When the diffusate dried to a brown residue with a meaty aroma, the amino acid concentrations declined 40-60%; but there was no pattern of disappearance that could be correlated with aroma. In a model system of amino acids alone no color or aroma developed, even after drying at 125°C and the concentration of most of the amino acids was 80-90% of the initial concentration. Addition of glucose to the amino acid solution led to large losses of amino acids and the development of a sweet grainy aroma which was not like meat aroma at all. Four moles of glucose disappeared per mole of amino-N disappearing in this study Reynolds (1965) reported ratios of 6:1 for glucose to amino-N. The amino acids may act as catalyst for sugar disappearance.

The character of the aroma developed on heating meat extracts varied with the amount of water present and the composition of the extract. In general, the first notes of the aroma from an aqueous extract heated at 125 °C were brothy, becoming sweet and like roasted grain as the amount of liquid decreased, then like roast or broiled meat and, finally, at complete dryness, a harsh burnt meat odor. Superimposed were acid, sour, green or plant-like, and other notes produced from the different compounds present (Wasserman and Gray, 1965; Zaika *et al.*, 1968).

There is no doubt that browning reactions occur on heating meat, but the relationship of the compounds formed to aroma remains to be established.

# SULFUR-CONTAINING COMPOUNDS

Sulfur compounds are involved in the flavor of cooked meats. Bouthilet (1951), Pippen (1967), and Kazeniac (1961) have reported on the role of sulfur compounds in chicken aroma. Minor *et al.* (1965) demonstrated that removal of sulfur components from chicken volatiles resulted in a loss of meaty aroma from the total odor.

Hydrogen sulfide has an obnoxious odor and is toxic when present in high concentrations, but appears to be a necessary component of meat aroma in low concentration.  $H_2S$  is produced from protein on heating as shown by Hamm (1966), increasing concentrations being formed at higher temperatures. In chicken meat heated for 1 hr about 92% of the  $H_2S$  was produced in the temperature range of 70–125°C (Parr and Levett, 1969). At acid pH values more  $H_2S$  was produced than at pH 6.15, thus supporting the report of Johnson and Vickery (1964) that beef and mutton from starved animals, with a lower pH than meat from normal animals, yielded more  $H_2S$  on heating.

The odor threshold of  $H_2S$  in water is 10 ppb (Pippen and Mecchi, 1969). The concentration of  $H_2S$  in freshly cooked chicken meat is 20–100 times greater than the threshold and leg meat produces two to three times more  $H_2S$  than breast.

Hydrogen sulfide may also contribute to cooked meat aromas by combining with other components to form new aromatic sulfur compounds. Pippen and Mecchi (1969) suggested the sym-trithiane reported by Minor *et al.* (1965) can come from the reaction of  $H_2S$  and HCHO. When these authors (Pippen and Mecchi, 1969) reacted  $H_2S$  with acetaldehyde in fat they demonstrated the disappearance of  $H_2S$ and formation of an odorous compound.

A number of interesting sulfur compounds have been reported in heated meat preparations. In roast chicken leg meat Swoboda (1970) identified dimethyl trisulfide, at a concentration of 10 ppb, as a very strong aroma component. An ether extract of beef broth contained 2-acetyl-2-thiazoline (Copier *et al.*, 1970) and Herz (1968) and Herz and Chang (1970) identified four sulfur-containing compounds in meaty-smelling fractions from boiled beef preparations. These compounds are 5-thiomethylfurfural, thiophencarboxy-2-aldehyde, 3,5-dimethyl-1,2,4-trithiolane, and 2,4,5-trimethyl-3-oxazoline.

From our present state of knowledge of meat aroma, sulfur compounds appear to have both desirable and undesirable effects on the aroma. A most undesirable compound has been reported in some samples of canned beef. While it is not a component of meat aroma, it is produced by the reaction of  $H_2S$  from cooked meat with mesityl oxide (Spencer, 1969). The compound, 4-mercapto-4-methylpentan-2-one, has an unpleasant cat urine odor and is definitely a flavor defect.

Von Sydow (1971) described unpublished work in which he related the importance of some sulfur compounds to canned beef off-flavor by stepwise regression analyses. The compounds, in order of decreasing importance, were: 2-methyl-thiophene, dimethyl sulfide, dimethyl disulfide, thiophene, and methyl mercaptan.

Thiamine is a vitamin whose loss as a result of heating has

been of concern primarily from a nutritional aspect. However, odorous compounds are formed on the thermal degradation of this vitamin. Arnold *et al.* (1969) reported that, while thiamine was stable at pH 3.5, at pH 6.7 2-methylfuran, 2-methylthiophene, 4,5-dihydro-2-methylthiophene, and H<sub>2</sub>S were formed. Methylthiophene compounds have a sulfury, onion-like aroma. Nonaka *et al.* (1967) identified 2-methylfuran and 2-methylthiophene in an odorous fraction isolated from boiling chicken meat.

A number of patents for simulated meat aroma are based on the reaction of a sulfur-containing compound, amino acids, and carbonyl compounds. A few patents have specified heating thiamine with various amino acids (Yamamoto *et al.*, 1970; Giacino, 1968). In many instances, however, hydrolyzed protein, meat or vegetable, is also included, making it difficult to evaluate the role of the pure materials in aroma development.

## LIPIDS

The role of lipids in meat aroma is being studied in a number of laboratories. It has been suggested that, although the basic meaty aroma is the same for beef, pork, and lamb species, differences are determined by the components contributed by the fat (Hornstein and Crowe, 1960). The term "fat," however, has not been carefully defined in many studies. Intramuscular fat, extracted from lean meat, may differ substantially from depot adipose tissue insofar as the quantity of free fatty acids and fatty acid composition of phospholipids are concerned. Adipose tissue, most often referred to as "fat," contains amino acids, proteins, sugars, salts, and the other components present in all tissues. In many studies these factors are not taken into consideration.

Adipose tissue, which yielded the characteristic aroma of the species on heating, was extracted with chloroform-methanol (2:1), and the extract was water-washed (Wasserman and Spinelli, 1970). The original extract retained the speciesspecific aroma but in the water-washed residue the characteristic aroma was either missing or greatly reduced. The watersoluble material from the lipid contained amino acids and at least glucose in the carbohydrate fraction. A typical lean meaty aroma developed on heating.

Lipids may contribute to flavor in several ways. They may act as solvent, trapping aroma components produced elsewhere. Thus, the aroma of water-washed pork lipid extract still had a "piggy" note when heat was first applied (Wasserman and Spinelli, 1970). This may be due to  $5,\alpha$ -androst-16en-3-one, the boar odor identified by Patterson (1968), which is dissolved in the lipids of the animal.

Lipid components or degradation products may react with compounds produced in the lean meat. Pippen and Mecchi (1969) demonstrated the possibility of this option. Lipid obtained from chicken adipose tissue by pressure and waterwashed to remove polar compounds had no chicken aroma on heating. However, lipid from chicken that had been boiled or fried contained increasingly more aroma. Increasing concentrations of sulfur compounds were also found. They showed that  $H_2S$  reacted with acetaldehyde to form an odorous compound and suggested other components may be formed in a similar fashion.

Flavor components are formed as a result of thermal oxidation of lipid constituents. The number of compounds arising from lipids and free fatty acids is fairly large, in keeping with the size of the molecules, degree of unsaturation, and type of substitution. Thermal autoxidation of lipids can occur at  $60^{\circ}$ C in the presence of a few free radicals, but most of the degradation occurs at 200–300 °C. At higher temperatures, ca. 600 °C, pyrolysis occurs with formation of bitter and acrid components. Lactones, alcohols, ketones, and lower fatty acids are associated with the thermal oxidation of the lipids.

Lipids have been investigated intensively in the flavor of dairy products and other foods; major studies on meat lipids in relation to aroma have been carried out by Hornstein and coworkers (Hornstein and Crowe, 1960, 1963; Hornstein *et al.*, 1961). Hydrolysis of triglycerides to free fatty acids occurred to greater extent when the fat was heated at  $100^{\circ}$ C in air instead of in a vacuum, and the concentration of carbonyl compounds was also greater on heating in air. The composition of free fatty acids in lamb differs from that of pork and beef, possibly accounting for the quantities of 2-enals and absence of 2,4-dienals found in heated lamb fat. The aroma of 2,4-decadienal was described by Patton *et al.* (1959) as "deep fat fried."

Lactones have been isolated from beef depot fat melted at 60°C for 1 hr (Watanabe and Sato, 1968). At least 19 compounds were present and could be separated into  $\gamma$ - and  $\Delta$ -lactones from C<sub>6</sub> to C<sub>15</sub>. The lactones are odorous compounds and may contribute either desirable or undesirable notes to the meat aroma. This class of compounds originates in the  $\gamma$ - or  $\Delta$ -hydroxy fatty acids that are known to be present in fat. Watanabe and Sato (1969) also isolated lactones from heated pork fat and identified  $\gamma$ -C<sub>5</sub> to C<sub>12</sub> and  $\gamma$ -C<sub>9,10,12,14</sub>.

A number of ketones and aldehydes were reported by Sanderson *et al.* (1966) from lean beef heated under  $N_2$  in water or beef fat for 8 hr. Fat heated alone gave insignificant quantities of the carbonyls, but heating the lean beef in fat yielded about  $3 \times$  as much carbonyls as lean beef heated in water, although qualitatively they were similar. Methyl-substituted aldehydes also present may have come from amino acids rather than from lipids.

Yamato *et al.* (1970) identified three aliphatic ketones, eight alkanals, six 2-alkenals, a 2,4-dialkenal, two  $\alpha$ -dicarbonyls, and an aromatic aldehyde from heating what they called beef fat. However, they used "beef leaf fat" which is adipose tissue, so the origin of some of the components may be in doubt.

Essentially no pentanal or hexanal appeared in the volatile carbonyls produced on heating cured hams to 70°C internal temperature, although substantial quantities appeared on heating uncured or fresh ham in the same manner. Cross and Ziegler (1965) suggested these are oxidation products of unsaturated fatty acid residues, probably linoleate, and that treatment with nitrite interferes with the oxidation of these unsaturated lipids.

Off-flavors developed in beef and mutton tallow on heating at 160 °C under  $N_2$  were attributed by Hoffman and Meijboom (1968) to 4-*cis*- and 4-*trans*-heptanal (with a "green" odor becoming butterscotch-like on dilution) and 2-*trans*,6-*cis*- and 2-*trans*,6-*trans*-nonadienal (green cucumber-like and tallowy odors).

Thus, although a considerable number of compounds have been identified in the aroma of meat products exposed to heat, none has been described as having a uniquely meaty odor. Continuous investigation of the components produced on heating the various classes of compounds constituting meat tissue and relating these to the overall meat aroma may eventually lead to elucidation of this problem.

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# Review of Literature on Chicken Flavor and Report of Isolation of Several New Chicken Flavor Components from Aqueous Cooked Chicken Broth

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Fifty liters of broth from 80 lb of stewing chickens cooked at 90°C under atmospheric pressure was filtered, subjected to continuous centrifugation to remove fat, and the volatiles were vacuum stripped. The distillate was saturated with sodium chloride and extracted with methylene chloride. The concentrated volatiles, analyzed by combined gasliquid chromatography/mass spectrometry, consisted of 68 components, of which 47 were identified

The majority were saturated and unsaturated alcohols, aldehydes, and ketones, of which those reported for the first time in chicken were 1-octanol, 2-methyl-3-buten-2-ol, 1-penten-3-ol, 3-penten-2-ol, 1-octen-3-ol, linalool, trans-2-octenol, a-terpineol, 4-hexen-3-one, 2,4-nonadienal, and piperonal. Suggested explanations for their occurrence are offered.

by their mass spectra and relative retention indices.

uring the last 20 yr many investigators, employing varied approaches, have identified a total of 178 components in cooked and raw chicken. These are summarized in Table I.

It was felt that a more complete picture of the flavor vola-

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tiles might be obtained by examining chicken broth from which most of the fat had been removed after cooking. In this way it was hoped to increase the probability of identifying the more water-soluble components.

### EXPERIMENTAL

Preparation of Broth. Eighty pounds of locally procured, eviscerated, stewing chickens were stored at  $-28^{\circ}$ C in poly-