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## FLAVOR OF TOMATO AND TOMATO PRODUCTS

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### Abstract

The characteristic sweet-sour taste of tomato and its overall flavor intensity are due to the following components: reducing sugars (fructose and glucose), free acids (mainly citric acid), their ratio, as well as some volatile substances, not identified so far, and the interplay between the above-mentioned groups of compounds. Of the minerals, potassium (by influencing the free acid content) and phosphate (due to its buffering capacity) indirectly affect the taste. The effect of the free amino acids (glutamic acid, glutamine, gamma-aminobutyric acid, and aspartic acid) is not unambiguously proven.

The characteristic tomatolike flavor is predominantly determined by volatile substances. Volatile substances develop partly during ripening, partly during the comminution of the ripe fruit, as an effect of the enzymes activated. Volatile substances characterizing fresh tomato flavor are derived mainly from fatty acids and amino acids. Differences between the flavors of different varieties, the less characteristic, weaker flavor of

hothouse-grown tomatoes or artificially ripened ones, are explained by the different quantitative proportions of the volatile substances. So far no key substance playing a decisive role in the flavor of tomato has been detected. Of the approximately 400 volatile compounds so far identified, several were found, however, which have an important role in the flavor of fresh tomato (cis-3-hexenal, trans-2-hexenal, 2-isobutylthiazole, hexanal, cis-3-hexen-1-ol, 2E,4E-decadienal, 6-methyl-5-hepten-2-one, etc.).

In the course of tomato processing the flavor substances undergo substantial qualitative and quantitative change, which may or may not be favorable. Depending on the extent of heat treatment and its duration, the reducing sugar and amino acid content decreases while the acid content (by the formation of pyrrolidone carboxylic acid) increases. A part of the volatile substances (mainly those providing "green" smell) evaporates and at the same time—traceable back to the breakdown of sugars and carotenoids—new components, among them those which characterize the "cooked" smell of tomato products, are formed (e.g., dimethyl sulphide, acetaldehyde). Although our knowledge as to the qualitative and quantitative composition of tomato flavor, the factors affecting it, and the effect of certain compounds on it has substantially increased, we are still not in the position to consciously develop the flavor during cultivation and processing. To achieve this further systematic research work is necessary.

## INTRODUCTION

The tomato (*Lycopersicon esculentum*), a member of the potato (Solanaceae) family, is very important as a food source. Within the family of Solanaceae, including about 1500 tropical and subtropical species, the genus *Lycopersicon* consists of relatively few (eight) species and is subdivided into two subgenera—*Eulycopersicon* and *Eriopersicon*. The species generally cultivated belong in the *Eulycopersicon* subgenus. When ripe they are characterized by an attractive red or yellow color and a pleasing flavor (1).

From the botanical aspect the tomato is a berry fruit, but it is cultivated and used as a vegetable (2).

The tomato probably originated in Central and South America, and its wild species spread most widely in Peru. Domestication of the plant presumably

started in Mexico, and it was brought from there to Europe soon after the discovery of America (1).

Quite a long time elapsed before the nutritional value of the tomato was realized. Reluctance to consume the tomato was caused mostly by its relationship to several poisonous plants (e.g., *Atropa belladonna*) (2). Britain was the last to accept it as a dietary item, in the middle of the eighteenth century.

Large-scale cultivation of the tomato did not begin until about a century ago, and it became generally cultivated only after the First World War. These days it is known all over the world and is one of the most important food plants (3).

The yearly output of tomatoes grown the world over increased between 1974 and 1983 from 38m tons to nearly 54m tons, amounting to an increase of 43% (4,5). The distribution of production, however, remained at almost the same level and is at present the following:

Europe (without U.S.S.R.)	21.8%
Asia (without U.S.S.R.)	23.5%
United States	14.3%
U.S.S.R.	13.3%
Africa	9.9%
South America	5.5%
Northern and Central America (without U.S.A.)	4.9%
Oceania	0.5%

Production per capita in the most important tomato-growing countries is summarized in Table 1.

Of cultivated vegetables, the proportion of tomato ranges from 20 to 30% on most continents, with the exception of Oceania (13–16%) and Asia (without U.S.S.R.) (6–7%).

Compared with other kinds of fruit, tomato production is third behind grapes and citrus fruits (5).

The unique popularity of the tomato is unrelated to its nutritive value. According to a survey carried out at the University of California at Davis, in which the main fruit and vegetable crops were ranked on the basis of their content of 10 vitamins and minerals, the tomato occupied only sixteenth place. At the same time, taking into account the amount consumed, its contribution to the vitamin and mineral requirement put it in first place. In this hierarchy broccoli, formerly occupying first place, was put in twenty-first place, spinach, formerly occupying second place, got eighteenth place, and Brussels sprouts moved from third place to thirty-fourth place (2).

*Table 1. Yearly per Capita Yield (kg) in Important Tomato-Producing Countries (5,6)*

Country	1980	1982	1984
Greece	231.3	195.9	200.0
Bulgaria	94.6	97.0	73.4
Italy	79.9	74.1	79.1
Turkey	78.4	77.9	76.0
Libya	75.9	73.0	71.7
Syria	71.7	81.5	74.4
Israel	65.8	85.3	92.7
Romania	64.0	83.3	88.5
Egypt	58.8	60.3	55.4
Spain	57.7	59.6	59.1
Jordan	50.2	55.9	58.1
Portugal	46.5	43.1	49.7
Tunisia	44.1	39.0	52.7
Hungary	40.2	35.7	34.6
Iraq	35.2	33.4	32.5
U.S.A.	29.8	33.8	32.3
The Netherlands	28.0	31.8	32.2
U.S.S.R.	24.7	26.7	26.6
Mexico	20.9	11.5	14.3
Argentina	20.2	20.7	21.8

The tomato's important role in nutrition can be explained by its hedonic value and versatility. Its consumption in the raw state is increasing all over the world, and in addition it is widely used in soups, meat and pasta dishes, and in the manufacture of soft drinks as the main raw material or as a flavoring additive.

Taking this into consideration, it is not surprising that the literature on this subject is enormously rich. Between 1969 and 1985 more than 3000 scientific papers were published on this subject, on the basis of data obtained from AGRIS (International Information System for the Agricultural Sciences and Technology). A great number of data are also available on the chemical composition of tomatoes.

The composition of the tomato is, however, greatly affected by the species, stage of ripeness, year of growth, climatic conditions, light, temperature, soil, fertilization, irrigation, and other conditions of cultivation, and, thus, data published in related literature fluctuate within wide limits. Davies and Hobson

*Table 2.* Composition of Dry Matter  
Content of Tomato (1)

Constituent	%
Fructose	25
Glucose	22
Saccharose	1
Citric acid	9
Malic acid	4
Protein	8
Dicarboxylic amino acids	2
Pectic substances	7
Cellulose	6
Hemicellulose	4
Minerals	8
Lipids	2
Ascorbic acid	0.5
Pigments	0.4
Other amino acids, vitamins, and polyphenols	1
Volatiles	0.1

(1), based on the critical analysis of papers publishing data on tomato composition, found the average dry matter content of the ripe fresh fruit to be 5.0–7.5%. They characterized the constituents of dry matter based on partly modified data published by Stevens in 1974 and brought up to date in the light of recent work, as seen in Table 2.

In the formation of the characteristic flavor of tomato, almost all the constituents listed in Table 2 may play some role. Their effect may be direct (flavor substances) or indirect (e.g., they provide an appropriate medium for the chemical or biochemical reactions leading to the formation of the flavor; they catalyze the above reactions; they are precursors of the flavor substances; they absorb to some extent the volatile substances; they affect the tension of the aroma components in the headspace, etc.).

In this survey we summarize the present knowledge only of the so-called flavor substances having a direct bearing on the sensory quality of tomato.

As with every foodstuff, the agreeable, characteristic flavor of tomato is the complex effect of the many volatile and nonvolatile components in which, beyond the flavor of the individual components, their interaction plays an important part. Although taste and odor are perceived by different senses, the

nearness of the sensory organs and their connection through the pharynx renders the separate analysis of taste or odor difficult. The recognition of such complicated systems can be carried out only step by step, and this always involves a certain inaccuracy. Taking this into consideration, the survey of the literature on flavor substances should begin with a short summary of the compounds affecting taste.

## SUBSTANCES AFFECTING TASTE

The pleasant sweet-sour taste of tomatoes is mainly due to their sugar and organic acid content. The character and intensity of taste is greatly affected by the salts present as well as the buffer effect of the various cations and anions. The free amino acids may also affect—partly by their own taste, partly by their buffer effect—the character and intensity of taste.

Bitter taste is not characteristic of tomato and is rarely encountered. If it is accidentally present it is caused by phenolics—long-chain organic compounds and alkaloids—however, these are not treated in this survey. The quality and quantity of proteins, fats, pectins, pigments, vitamins, and cellulose are also not discussed, since these components affect primarily the color, texture, and consistency of tomato; their effect upon flavor is only secondary.

### Sugars

About 50% of the dry matter in cultivated tomatoes is made up of sugars, primarily glucose and fructose. There is frequently saccharose as well, but its quantity rarely exceeds 0.1% of the fresh mass (8–10).

There are occasionally also minute quantities of other sugars to be found in tomato, such as raffinose (11), arabinose (12), xylose (12), galactose (13), an unidentified ketoheptose (14), and the sugar alcohol, myoinositol (10).

In the initial stage of development the tomato berry contains a relatively small amount of sugar (<1%) with glucose dominating (glucose:fructose ratio = 1.8). During growth and on ripening, the sugar content increases significantly and gradually decreases the glucose:fructose ratio. The total sugar content of the ripe tomato is between 1.7 and 4.7%, depending on the variety; the glucose:fructose ratio is around 1, and the amount of fructose in most varieties exceeds that of glucose (10,15,16).

Out of the environmental factors, light has the most profound effect on sugar concentration. The more sunlight that reaches the fruit, the higher the sugar content. It is a consequence of this that hothouse tomatoes grown during the winter months contain substantially less sugar (17,18).

During processing the sugar content decreases depending on the extent and time of heat treatment. The loss can be explained by caramelization,

browning reactions between sugars and amino acids, and by the formation of 5-hydroxymethyl furfural in the acid medium upon the loss of  $H_2O$ . According to El Miladi and co-workers (12), in processed tomato juice the breakdown of glucose was almost the same as that of fructose. The total loss of sugar amounted to about 16%. Alpári (19) found a 5% loss of sugar in the course of manufacturing powdered tomato from puree.

### Organic Acids

Organic acids, among them mostly citric and malic acids, form more than 10% of the dry content of tomatoes (8,20-22). In addition to the two main acid components, the presence of a number of other acids was detected in substantially lower concentrations. According to Davies and Hobson's survey (1) of the aliphatic monocarboxylic acids, formic, acetic, lactic, mevalonic, and pyruvic acid, of the aliphatic di- and tricarboxylic acids, trans-aconitic, dihydroxy tartaric, fumaric, malonic, oxalacetic, oxalic, 2-oxoglutaric, succinic, and tartaric acids, of the sugar acids, galacturonic acid, and of the alicyclic monocarboxylic acids, quinic acid, were detected.

The acid content of tomato changes in the course of ripening. During growth of the berry and during ripening, up to the breaker stage, the acid content increases, later to decline. Initially malic acid is predominant; in the mature green stage, the malic acid: citric acid ratio is above 1; after this the amount of citric acid exceeds that of malic acid; and in the ripe red tomato their ratio is 0.5 or below (23). The malic acid: citric acid ratio as against the glucose: fructose ratio is highly dependent on the variety. In the fully ripe fruit, citric acid accounts for 45-66% of the total acidity in English cultivars (23), 40-90% in American cultivars (24), and 60-85% in Hungarian cultivars (8).

Of the environmental factors the potassium content of the soil most affects the total acid content in the fruit. Davies and Winsor (25) found a positive logarithmic correlation between the potassium level in the soil and the acid content of the fruit. The quality and quantity of organic acids is important not only in determining flavor but in view of technology, because below pH 4.3 the butyric, thermophilic, and putrefactive anaerobic microorganisms cannot thrive (26).

### Free Amino Acids

On the average, free amino acids form about 2-2.5% of the total dry matter content of tomato. This is, however, an estimated value, the more so as there are only few comparable quantitative data available on the free amino acid



content of tomatoes. These fluctuate within wide limits due to environmental factors, cultivar, and, to a not negligible degree, to the analytical method used.

Most of the authors agree that glutamic acid forms the major part of the free amino acids present in tomato (12,28,32-35). Glutamine, gamma-amino-butyric acid, and aspartic acid are present in substantial amounts, too. These four amino acids form about 80% of the total free amino acid content (34).

Beyond the above-enumerated amino acids, all the other amino acids usually present in protein hydrolysates were detected in tomato, even if in minute quantities. Six to ten ninhydrin-positive, unidentified components were also detected (35,36).

During the ripening period a substantial growth of the glutamic acid content and some increase in aspartic acid content was observed (23,37-39). There are only few data available on the change of glutamine during ripening. Liu and Luh (35) found an increase at the beginning, with a decrease later on. The data on gamma-aminobutyric acid seem to be contradictory. Some of the authors found that its amount increased (35,39) while others found it to decrease (37, 40), and some authors did not observe any important change (41).

Quantitative distribution of amino acids among cultivars shows characteristic differences (33,35,39,42).

The glutamic acid and aspartic acid concentration in tomato is highly dependent on the nitrogen and phosphate content in the soil. A high nitrogen content and a low phosphate content substantially increases the concentration of these amino acids (43). It was observed also that tomatoes grown in the field had a substantially higher glutamic acid content than those grown in a hot-house (40).

The amino acid concentration is also important from the point of view of processing and storage stability, because a high amino acid concentration promotes browning reactions. As to the effect of heat upon the quantity of amino acids opinions differ; the authors, however, agree that during heat treatment the glutamic acid content diminishes, while pyrrolidone carboxylic acid is formed (28,36).

Stadtman (36) observed a similar substantial decrease in the gamma-amino-butyric acid content, but he makes no attempt at explanation. Upon a milder treatment the hydrolysis of glutamine may result in glutamic acid formation (28), but continued heat treatment reduces the amount of this amino acid, too (36). Only El Miladi and co-workers (12) found a significant increase of glutamic acid, but Stadtman ascribes this to an error in calculation (36).

During storage the amino nitrogen content of tomato juice and puree diminishes rapidly, particularly at higher temperatures (27,44).

## Minerals

Minerals constitute, on average, about 8% of the dry matter content of tomatoes. Since their effect on the pH and titratable acid content is important, they highly influence the taste of tomatoes, too. Of the cations, potassium, forming about 80–90% of the total amount of cations, and of the anions, phosphate, forming about 50–60% of the anions, are of highest importance (1,25,42,45).

In addition to the above, the presence in minute or trace amounts of a number of other cations and anions in tomatoes has been shown (Na, Mg, Ca, Fe, Cu, Mn, Zn, Co, Ni, Cr, Se, Si, Rb, Al, B, Cd, Pb, SO<sub>4</sub>, Cl, NO<sub>3</sub>) (1,25,42,46–48).

In the course of development, growth, and ripening of the fruit, the mineral content shows an increase in absolute value, but the concentration related to dry matter content remains unchanged (47).

The total mineral content and composition of the fruit is determined by the mineral content of the soil and by fertilization—cultivar plays a much lesser role (25,35,45).

During processing the mineral content related to the dry matter content shows practically no change. The increased Ca, Mg, NO<sub>3</sub>, and SO<sub>4</sub> content found occasionally in tomato juice is due to defective processing or pollution by sand, dilution of concentrates with tap water, etc. (49).

## FLAVOR OF TOMATO AS AFFECTED BY COMPOUNDS STUDIED

The rapid growth of the popularity of tomatoes induced breeders to produce new cultivars with good appearance, high yield, good mechanical properties, and thus suitable to mechanized harvesting and resistant to diseases.

However, in the course of improvement, not much attention was paid to the study of flavor characteristics. In consequence the number of consumer complaints rapidly increased and many newspaper articles were published on the characterless, weak flavor of new cultivars and even on off-flavor. Although the researchers did not attribute the quality deterioration to the introduction of new cultivars—rather to picking the fruit in an unripe stage and to the postharvest maturation—they started intense research activity to find out the actual causes of deterioration. The aim was to discover the chemical factors causing the sensory differences between varieties and throw light on the correlations between sensory characteristics and composition. It is due to this that

since the 1960s the number of publications on this subject has substantially increased (9,16,24,34,42,50-59).

As a result of these investigations the knowledge on the role of the main chemical components in the characteristic flavor of tomato has substantially increased. We are, however, still far from knowing all the correlations between the components and from the quantification of these effects.

In the formation of the characteristic sour-sweet taste and of differences in flavor of the genotypes an important role is attributed to the concentration and ratio of sugars and organic acids (9,16,50,51).

Some of the authors observed a high positive correlation between sweetness and reducing sugar content (53,54,56,60) and between sweetness and water-soluble dry matter content (56,57,60). The influence of saccharose upon the sweetness of tomato seems to be negligible because of its very low concentration (9).

Of the reducing sugars fructose is considered to be of greater importance because its sweetening power is twice that of glucose (9,54).

Stevens and co-workers (54) have proven experimentally the greater contribution of fructose to the sweetness of tomato. Using a cultivar of relatively low sugar, acid, and volatile substance content ("Cal Ace"), they added fructose or glucose to it and studied their respective effects upon sweetness. The stepwise regression analyses indicated that about 50% of the variation in sweetness was related to changes in fructose concentration. They did not attribute, however, greater practical importance to this fact, since glucose and fructose are present in different cultivars in about the same proportion.

The sensation of sweetness, although probably highly dependent on the sugar content, cannot solely be explained by this.

Jones and Scott (58) investigated  $F_1$  hybrids of relatively high sugar and acid content and did not find close correlation between their reducing sugar or dry matter content on the one hand and their sweetness on the other. One of the possible explanations of this may be that above a certain threshold of sugar levels the impact of additional increases in sugars on perceived sweetness may plateau. Even in the case of these hybrids of high sugar content, they considered reducing sugars only to be 50% responsible for perceived sweetness. Watada and Aulenbach (59) did not find correlation between sweetness and dry matter content either. They made a comparative analysis with variety "Walter" using vine-ripened tomatoes and tomatoes picked at different stages of maturity and ripened postharvest. The absence of correlation in this case was caused by the insignificant differences in the dry matter content. Their observation, however, that fruit ripened on the vine was sweeter in spite of equal dry matter content shows that, in addition to sugars, other components affect perceived sweetness.

Of the nonvolatile components of tomato, in addition to sugar, only the influence of citric acid was investigated. Stevens and co-workers (54), in their addition experiments mentioned above, found an interesting relationship between the sensation of sweetness and the glucose-citric acid interaction. In the case of high citric acid content, they found glucose to affect sweetness more than fructose. Diminishing citric acid content, however, diminished the importance of glucose. In samples where the total sugar content was low, citric acid reduced perceived sweetness. On the other hand, in samples of high sugar content, citric acid had a sweetness-increasing effect.

Based on their studies, they established that inclusion of fructose and glucose, interactions between glucose and citric acid, and interactions between glucose and fructose accounted for about 80% of the variation in sweetness. When studying the influence of pH they found that it affected most flavor characteristics. As much as 86% of the variation in sweetness was ascribed to an interaction between sugar content and pH.

The effect of salts upon perceived sweetness was not studied in detail. Only deBruyn and co-workers (9) mention that the presence of salts intensifies sweetness. In the course of their addition experiments they established also that panel members favored tomato sample to which, beside sugars and acids, salts (K citrate) were added. It was not detailed, however, whether this advantageous judgment was based on more intense sweetness or other sensory characteristics. This paper was the first to mention that volatile substances, in spite of their minute quantity, may influence the sensation of taste. This was later confirmed experimentally by several authors (54,59).

Stevens and co-workers (54) found among the volatile substances, separated by gas chromatography, several which affected sweetness. Thus, when beside the interaction between sugars and pH, they took into consideration the effect of two volatile substances, separated by gas chromatography, not identified but measured (marked No. 43 and 60), they found the interaction even closer (about 95%).

Watada and Aulenbach (59) found four volatile substances, i.e., four peaks separated and measured by gas chromatography, which had a significant effect upon sweetness [variable: peaks 1, 6 (11 + 12),  $R^2 = 0.48$ —significant at the 1% level].

The sour taste of tomato can be ascribed mainly to the organic acids in the fruit. The researchers found unanimously close correlation between the extent of sourness and the titratable acidity (53,54,56-60). However, the same authors did not find such an unanimous correlation between sourness and pH. Some of the authors observed a close correlation (53,54,56,60) while others found only a very loose correlation or none at all (57-59).

This definitely shows that in tomato the sensation of sourness is more dependent on the free acid content than on the hydrogen ion concentration (57). This seems to be proven by the observation in model solutions according to which at the same pH value acetic acid is substantially more sour than hydrochloric acid. This shows that in some way acids not dissociated may also affect the sensation of taste. This assumption can be explained well by the Richard theory, according to which a reaction of hydrogen ions at the receptor surface causes the sour taste, and, as hydrogen ions are used up, more appear from the dissociation of the acids (61).

The loose correlation observed occasionally between sourness and pH may be explained, apart from the organic acids present in different proportions and having different dissociation constants, by the strong buffer effects as observed in tomato (16,24).

Of the publications finding close correlation between titratable acidity or pH on the one hand, and sour taste on the other, two attribute more importance to pH, the other two to titratable acidity.

The differing, sometimes contradictory results show the difficulties involved in the study of complex biological systems and the indefiniteness of the anomalies caused by complicated interactions. In many cases this situation is aggravated by methodological problems, rarely mentioned in the papers. For instance, we observed that the exact determination of titratable acidity of tomato is not at all a simple task because of the presence of different poly-functional organic acids and of the indistinct equivalence point in consequence of strong buffering effect. In addition, the pH range of tomatoes is restricted to relatively narrow limits (4.0–4.5), and if the slight differences are measured with an instrument not sensitive enough, the results cannot be evaluated. If the differences in the routine and sensitivity of the sensory panels are also taken into consideration, it is obvious how difficult it is to come to unambiguous conclusions. Further, it is also obvious that conclusions drawn by mathematical statistical evaluation are highly influenced by the substantial differences in compositional or sensory characteristics as measured in tomatoes compared within a single experiment. In the case of more marked differences relationships are easier to discover, however, if differences are smaller they may be difficult to detect, partly because of an error in measurements, partly because of the interplay between compositional characteristics.

The research of Jones and Scott (60) is a good example of these difficulties. They carried out two series of experiments: In one, no correlation was found between sourness and pH or titratable acidity, while in the other series, a close correlation was observed with both parameters.

Gould (65) found that panel members were not able to discover differences in the pH of tomato juices below 0.16, while 0.08% differences were readily detected in the titratable acidity. This experiment shows that the correlation

between perceived taste and titratable acidity is easier to detect than that between pH and sourness.

While the correlation between titratable acidity and pH, on the one hand, and sourness of tomato on the other, was investigated by a number of researchers, there are only very few references to the estimation of the contribution of individual acids to taste.

Investigating the taste of different acids in model solutions, deBruyn and co-workers (9) found that malic acid is more sour than citric acid. Their results were in agreement with findings of Gardner (62), who found malic acid about 14% more sour than citric acid. These authors agreed, however, that malic acid had less influence on tomato taste than citric acid because of its lower concentration. These opinions are supported by the investigations of Stevens (16), according to which close correlation was found between the titratable acidity and the total citric acid content ( $r = 0.09$ ), while the correlation between titratable acidity and total malic acid content was less close ( $r = 0.11$ ). Stevens and co-workers (64) found on the basis of their addition experiments that citric acid was responsible for the sour taste of tomato to about 60%. They investigated the effect of sugars on sourness, too, in addition experiments. It was established that in the case of low citric acid and glucose content, fructose reduced sour taste; with high citric acid and glucose content, however, this effect was not valid. Notwithstanding, they found the effect of sugars on sourness much less intense than the effect of acids on sweetness.

Kader and co-workers (53) found the pH, the interaction between titratable acidity and pH, in addition to the fructose content, 84% responsible for perceived sourness.

The possibility that, beside malic and citric acids, the free amino acids may also affect the taste of tomato was first observed by Davies (42).

Investigations of Kirimura and co-workers (63) revealed that the taste threshold concentration of pure amino acids is 0.03–3.0 g/L. Amino acids affect taste by their own taste, by their taste-enhancing capacity, or by their buffer effect. The taste-enhancing effect of L-glutamic acid (61,63,64), of the amino acids present in tomato in the greatest amount, is well known. On the other hand, deBruyn and co-workers found (9) that when the amino acid content of tomato juice was increased (with glutamic acids, aspartic acid, glutamine, and asparagine jointly) three to four times, the taste of tomato was unfavorably affected.

On searching for the reason of off-flavor, occasionally observed in tomatoes picked when unripe, then after-ripened, Kader and co-workers (34) established that during after-ripening the quantity of only four of the amino acids (glutamic acid, asparagine, glutamine, and alanine) increased. The sweetish glutamine, asparagine, and alanine were present in vine-ripened tomato in greater amount than in artificially ripened ones, however, because of their small

quantity, their contribution to taste was considered negligible. Glutamic acid, however, was present in a much higher quantity in the after-ripened tomato than in the vine-ripened, and its quality was correlated with the off-flavor. Other experiments have shown that glutamic acid had no direct role in the development of off-flavor. At the same time it was established that differences in amino acid content have less bearing on taste than other factors. Stevens (16), studying the buffer systems in tomato, found that differences in buffer activities of amino acids are of minor importance.

Results in related literature, however, do not exclude the possibility of participation of amino acids in the formation of the character of tomato flavor.

Of the minerals, potassium affected titratable acidity, while phosphate, having a buffer effect, influenced significantly the hydrogen ion concentration—titratable acid ratio (16). Thus, indirectly, both play a role in the development of sour taste.

The same research group treated in two papers (53,54) the correlation between volatile substances and sourness. They found a number of chromatographically separated peaks, the substance of which influenced significantly the acid taste. Most of them showed negative correlation with sourness. The peak, marked 43 and having the closest correlation, reveals the presence of a substance that affects significantly the sweetness, too.

Taste and the overall flavor intensity showed a close correlation with the reducing sugar, or soluble solid content and the titratable acidity (9,53,54,56, 60). In the development of intensity the titratable acid content appears to have a more substantial role (9,53,56,60).

In the addition experiments of Stevens and co-workers (54), significant correlation was found between overall flavor intensity and citric acid and fructose concentration as well as the glucose-citric acid interplay. These are to about 70% responsible for intensity. The sensation of flavor was the most intense when the sample contained substantial fructose and citric acid but very little glucose. In this case citric acid was of greater effect upon intensity than fructose. With high glucose content, however, there was no difference in their effect.

The highest overall flavor intensity was observed in samples where both sugar and titratable acid content were high. They found also some volatile compounds to affect intensity. The substances revealed in gas chromatographic peaks marked 53 and 77-79, if only to a small extent, had a significant effect upon intensity.

The flavor of heat-treated tomato products, because of compositional changes, as discussed earlier, differs from that of fresh tomato. In order to intensify the taste of tomato juice it is generally permitted to use common salt. However, this single additive is not sufficient to eliminate the quality differences between tomato juices. In the knowledge of the strong effect of the

sugar-acid ratio upon complete aroma intensity, Gould (65) attempted to find out the sugar-acid ratio that lends the highest sensory value to tomato juice. He found that panel members could detect slight differences in the sugar-acid ratio ( $<3$ ). The highest values were assigned to juices with sugar-acid ratios between 10:1–18:1. Based on his experiments he concluded that tomato juices of more stable quality could be achieved by occasionally adding saccharose and/or citric acid.

The off-flavor occasionally appearing in heat-treated vegetable products has been ascribed by several authors to the formation of pyrrolidone carboxylic acid (5-oxo-2-pyrrolidone-carboxylic acid) (29,66–71).

Shallenberger and Moyer (66) found pyrrolidone carboxylic acid to have, particularly in neutral solutions (pH 5–6), an unpleasant medicinal, phenolic, or bitter flavor.

Mahdi and co-workers (68) observed that the taste of tomato juices having different pyrrolidone carboxylic acid concentrations, dependent on sterilization periods of different length, differed significantly. When the pyrrolidone carboxylic acid content was set at a definite concentration (0.14% w per v) panelists could not differentiate between samples and found them uniformly bitter, medicinal, or acidic.

Luh and co-workers (27,44) studied the changes occurring in tomato products during storage (juice, puree, pasta) and established that the most sensitive index of quality deterioration during storage, particularly at temperatures above 25°C, were changes in the flavor. Deterioration of color appears substantially later.

## VOLATILE COMPONENTS

While there was a close correlation between the sweet-sour taste of tomato and the intensity of this and of the overall flavor on the one hand and the sugar and acid content and the sugar-acid ratio on the other, the same parameters showed very weak (53,54,56,57) or no correlation (9,58,60) with the tomato-like flavor. Therefore, to acquire knowledge of the compounds and groups of compounds responsible for the tomatolike flavor, it is necessary to thoroughly study the composition of the volatiles.

The volatile substances first detected in tomato were acetaldehyde and ethanol (72). Soon afterwards Sándor (73) determined all the volatile acids present in tomato but did not identify individual acid constituents. In the next 30 years the number of identified substances increased only slightly (to 30–40 compounds) and most of them were carbonyls and alcohols. In the mid-1960s (primarily due to the development of the gas chromatograph and various other instruments suitable for structure identification) the number of detected and



identified volatile constituents of tomato and tomato products showed an abrupt increase, and has steadily increased ever since. At present the number of the compounds known amounts to ca. 400. These compounds, classified according to their chemical structure, are listed in Table 3. In listing the constituents only the original publications were taken into account so as to promote the estimation of the probability of their presence on the basis of the number of references after each compound. The list contains all the compounds found in the collected literature, independent of the method of identification.

This course of action is justified upon the consideration that even the most reliable method of identification may lead to false results (e.g., if some foreign matter gets into the sample during or prior to analysis) whereas the simplest method of identification may yield lasting results (as shown later in relation to *cis*-3-hexenal).

The exact number of compounds detected cannot be given because complete identification of the structure was not always carried out. Thus, in the case of compounds where all the possible isomers were not identified, it is not possible to establish whether it is one of the known structures or not.

Another problem arises in relation to the border line between volatile and nonvolatile substances, as there are very slightly volatile compounds that have intense smells. Therefore, all we can say is that the number of substances detected so far in tomato considered to be volatile is between 400 and 412.

### Development of Volatile Components

The free amino acids and fatty acids in tomato seem to be of much greater importance as possible precursors of volatiles than are the carbohydrates forming more than 50% of the dry matter content (26,108,135).

A great number of alcohols, carbonyls, acids, and esters can be derived from amino acids. With the aid of  $C^{14}$  labeled precursors, it was established that in the first step the amino acid is enzymatically deaminated into the appropriate keto acid, then, bound to thiaminepyrophosphate, turns into so-called activated aldehyde under decarboxylation, as a result of which various volatile esters are formed. By enzymatic reduction alcohols may also be formed from the aldehydes (136).

Yu and co-workers (137-140) considered that, of the free amino acids present in tomato, alanine, leucine, and valine were the most important aroma precursors. Examining the activity of enzyme preparations extracted from tomatoes grown under various conditions (in the field, hothouse) and picked at different stages of ripeness, they found that the activity of the enzyme extract of field-grown ripe tomatoes was higher than that obtained from hothouse-grown ripe tomatoes or of the extract obtained from field-grown

Table 3. Volatiles in Tomato and Tomato Products

ACYCLIC AND ISOCYCLIC COMPOUNDS	
<u>HYDROCARBONS</u>	
heptane	113
octane	113
nonane	113
decane	103
undecane	103, 113
pentadecane	113
ethylene	93
camphene [2,2-dimethyl-3-methylenebicyclo [2.2.1] heptane]	134
3-carene	103, 132
limonene [1,8-p-menthadiene]	82, 103, 111, 113, 123, 134
myrcene [7-methyl-3-methylene-1,6-octadiene]	103, 132
$\alpha$ -phellandrene [1,5-p-menthadiene]	113
$\beta$ -phellandrene [1,7/2/-p-menthadiene]	121
$\alpha$ -pinene [2-pinene]	82, 92, 97, 103, 121, 134
$\beta$ -pinene [2/10/-pinene]	124
sabinene [4/10/-thujene]	132, 134
terpinolene [1,4/8/-p-menthadiene]	132, 134
triisobutylene [2,5,8-trimethyl-1,4,7-nonatriene]	103
benzene	103, 132, 134
toluene	103, 112, 113, 118, 123, 124, 125, 132, 134
ethylbenzene	112, 113, 118, 123, 125, 132, 134
styrene [vinylbenzene]	124, 125, 132, 134
propylbenzene	113, 124, 125, 132, 134
cumene [isopropylbenzene]	103, 112, 113, 134
butylbenzene	134
o-xylene [1,2-dimethylbenzene]	112, 113, 123, 124, 125
m-xylene [1,3-dimethylbenzene]	113, 118, 123, 124, 125, 132, 134
p-xylene [1,4-dimethylbenzene]	103, 112, 113, 118, 123, 124, 125, 132, 134
l-ethyl-4-methylbenzene	124, 125
diethylbenzene (unknown structure)	124, 125
cymene (unknown structure)	112
p-cymene	113, 125, 132, 134
trimethylbenzene (unknown structure)	124, 125
hemimellitene [1,2,3-trimethylbenzene]	103, 113, 132, 134
pseudocumene [1,2,4-trimethylbenzene]	103, 113, 132, 134
mesitylene [1,3,5-trimethylbenzene]	113
biphenyl	113, 123
naphtalene	113
<u>ALCOHOLS</u>	
methanol	77, 81, 85, 95, 101, 103, 108, 113, 116, 127, 134
ethanol	72, 74, 81, 84, 85, 86, 93, 94, 95, 101, 103, 105, 108, 111, 113, 116, 117, 119, 125, 132, 134
1-propanol	84, 88, 94, 95, 101, 103, 111, 116, 117, 125, 132, 134

(Continued)

Table 3. (Continued)

ACYCLIC AND ISOCYCLIC COMPOUNDS	
<i>ALCOHOLS</i>	
2-propanol	84, 86, 93, 95, 102, 116, 134
2-methyl-1-propanol	81, 84, 85, 86, 92, 97, 101, 103, 108, 111, 113, 116, 117, 120, 121, 123, 132, 134
2-methyl-2-propanol	101
2-propen-1-ol	132, 134
1-butanol	81, 84, 85, 86, 95, 102, 103, 108, 111, 112, 113, 116, 117, 118, 120, 121, 123, 125, 132, 134
2-butanol	101, 111, 132, 134
buten-1-ol (unknown structure)	112
2-methyl-1-butanol	81, 84, 85, 86, 99, 101, 102, 103, 104, 111, 113, 123
3-methyl-1-butanol	81, 84, 85, 86, 92, 93, 95, 97, 99, 100, 101, 103, 105, 108, 111, 112, 113, 117, 118, 119, 120, 121, 123, 124, 125, 132, 134
3-methyl-2-buten-1-ol	113, 123
2-methyl-3-buten-2-ol	108, 111, 113, 123, 132, 134
2,3-butanediol	113
1-pentanol	81, 84, 85, 86, 95, 99, 101, 108, 111, 112, 113, 116, 117, 118, 119, 120, 123, 124, 125, 132, 134
2-pentanol	113, 117, 120
3-pentanol	102, 109, 113
cis-3-penten-1-ol	108
1-penten-3-ol	103, 104, 111, 112, 113, 117, 123, 125, 132, 134
2-methyl-1-pentanol	117, 120, 121
3-methyl-1-pentanol	113, 117, 120
2-methyl-2-pentanol	112, 125
1-hexanol	81, 84, 85, 86, 92, 95, 97, 100, 101, 104, 108, 111, 112, 113, 116, 117, 118, 119, 120, 121, 123, 125, 132, 134
2-hexanol	101
hexenol (unknown structure)	121
2-hexen-1-ol (unknown structure)	124
cis-2-hexen-1-ol	112, 124
trans-2-hexen-1-ol	112, 117, 118, 119, 120, 123, 125, 132, 134
3-hexen-1-ol (unknown structure)	100, 102
cis-3-hexen-1-ol	84, 86, 99, 103, 104, 105, 108, 111, 112, 113, 117, 118, 119, 120, 123, 124, 125, 132, 134
trans-3-hexen-1-ol	112, 117, 120, 124
cis-4-hexen-1-ol	117, 132, 134
methylhexanol (unknown structure)	103
2-methyl-3-hexanol	92, 97, 102
5-methyl-1-hexanol	116
1-heptanol	134
2-heptanol	112, 125
4-heptanol	112, 125
6-methyl-5-hepten-2-ol	108, 111, 113, 123, 125, 132, 134
1-octanol	116, 134

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 ACYCLIC AND ISOCYCLIC COMPOUNDS
 

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ALCOHOLS

- 1-octen-3-ol 108, 116, 123, 132, 134  
 7-methyl-1-octanol 116  
 1-decanol 116  
 8-p-cymenol 113  
 citronellol [3,7-dimethyl-6-octen-1-ol] 125, 132, 134  
 farnesol [3,7,11-trimethyl-2,6,10-dodecatrien-1-ol] 132  
 geraniol [trans-3,7-dimethyl-2,6-octadien-1-ol] 112, 113, 123, 125, 132, 134  
 linalool [3,7-dimethyl-1,6-octadien-3-ol] 104, 108, 111, 113, 116, 117, 123, 125, 127, 132, 134  
 nerol [cis-3,7-dimethyl-2,6-octadien-1-ol] 113, 117  
 nerolidol [3,7,11-trimethyl-1,6,10-dodecatrien-3-ol] 132  
 terpinen-4-ol [1/7/-p-menthen-4-ol] 113  
 $\alpha$ -terpineol [1-p-menthen-8-ol] 111, 113, 117, 123, 125, 132, 134  
 benzyl alcohol 104, 112, 113, 117, 118, 123, 125, 132, 134  
 2-phenylethanol 88, 104, 108, 111, 112, 113, 116, 117, 118, 120, 123, 124, 125, 132, 134  
 4-isopropylbenzyl alcohol 124, 125  
 menthol 127

PHENOLS

- phenol 104, 113, 132, 134  
 o-cresol [2-methylphenol] 104, 132, 134  
 p-cresol [4-methylphenol] 132  
 4-ethylphenol 104, 111, 113, 123  
 4-vinylphenol 112, 113, 118, 123, 132, 134  
 guaiacol [2-methoxyphenol] 104, 108, 111, 113, 117, 118, 123, 125, 132, 134  
 4-ethyl-2-methoxyphenol 113, 132  
 2-methoxy-4-vinylphenol 104, 111, 113, 117, 123, 132, 134  
 eugenol [4-allyl-2-methoxyphenol] 96, 104, 108, 111, 112, 113, 118, 123, 125, 132, 134  
 3,4-xylene 132

ETHERS

- diethyl ether 117, 127  
 1,1-dipropoxyethane 103  
 1-ethoxy-1 (3-methylbutoxy)-ethane 103  
 1-ethoxy-1-pentoxylethane 103  
 1-methoxy-4-methylbenzene 113  
 isopropyl-methoxybenzene (unknown structure) 112, 118, 125  
 2-methoxy-biphenyl 132

ALDEHYDES

- formaldehyde 90, 130, 131, 132  
 acetaldehyde 72, 74, 76, 80, 81, 84, 90, 93, 94, 95, 101, 103, 104, 108, 114, 116, 127, 130, 131, 132, 134

(Continued)

Table 3. (Continued)

## ACYCLIC AND ISOCYCLIC COMPOUNDS

ALDEHYDES

propanal	90, 93, 94, 95, 103, 114, 116, 117, 120, 127, 130, 131
acrolein [2-propenal]	130, 131, 132, 134
2-methylpropanal	108, 116, 130, 131
butanal	103, 108, 116, 134
2-butenal	130, 131
2-methylbutanal	103
3-methylbutanal	76, 80, 81, 84, 86, 92, 93, 94, 97, 101, 102, 103, 104, 108, 111, 113, 117, 121, 130, 131, 132, 134
2-methyl-2-butenal (unknown structure)	121, 124, 125
tiglaldehyde [trans-2-methyl-2-butenal]	103, 113
pentanal	90, 103, 113, 116, 123, 130, 132, 134
2-pentenal (unknown structure)	103, 119, 130, 131
trans-2-pentenal	108, 134
3-pentenal (unknown structure)	103
methylpentenal (unknown structure)	103
hexanal	80, 81, 84, 86, 88, 90, 92, 93, 97, 100, 101, 102, 103, 104, 108, 111, 113, 114, 116, 117, 118, 120, 121, 122, 123, 124, 125, 130, 131, 132, 134
hexenal (unknown structure)	112, 118
2-hexenal (unknown structure)	80, 88, 90, 100, 116, 130, 131
cis-2-hexenal	103, 119
trans-2-hexenal	81, 84, 86, 103, 104, 108, 111, 113, 117, 119, 120, 121, 122, 123, 124, 125, 132, 134
cis-3-hexenal	104, 108, 111, 117, 121, 122, 123
2E,4Z-hexadienal	123
2E,4E-hexadienal	104, 123, 132, 134
heptanal	103, 116, 130, 132
heptenal (unknown structure)	112, 118
2-heptenal (unknown structure)	96, 103, 119, 131
trans-2-heptenal	111, 123, 132, 134
2E,4Z-heptadienal	104, 111, 123
2E,4E-heptadienal	104, 108, 111, 123, 132, 134
octanal	116, 130, 131, 132, 134
2-octenal (unknown structure)	125, 131
trans-2-octenal	108, 111, 123, 132, 134
nonanal	90, 102, 109, 116, 130
2-nonenal (unknown structure)	130
trans-2-nonenal	111, 123, 132, 134
2E,4E-nonadienal	111
decanal	102, 109, 131, 132, 134
2-decenal (unknown structure)	130, 131
2,4-decadienal (unknown structure)	96, 125
2E,4Z-decadienal	111, 123, 132, 134
2E,4E-decadienal	104, 108, 111, 123, 132, 134
undecanal	116
dodecanal	102, 109

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 ACYCLIC AND ISOCYCLIC COMPOUNDS (Continued)
 

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ALDEHYDES

citral [3,7-dimethyl-2,6-octadienal] 76, 82  
 neral [cis-3,7-dimethyl-2,6-octadienal] 102, 110, 111, 123, 132, 134  
 geranial [trans-3,7-dimethyl-2,6-octadienal] 108, 110, 111, 113, 123, 125, 132, 134  
 citronellal [3,7-dimethyl-6-octenal] 76, 82, 132  
 farnesal [3,7,11-trimethyl-2,6,10-dodecatrienal] 110  
 benzaldehyde 84, 86, 92, 97, 102, 103, 104, 108, 111, 113, 116, 117, 118, 121, 123,  
 124, 125, 127, 130, 132, 134  
 3-methylbenzaldehyde 113  
 4-methylbenzaldehyde 125, 127, 132, 134  
 salicylaldehyde [2-hydroxybenzaldehyde] 104  
 4-hydroxybenzaldehyde 113, 123  
 3-methoxybenzaldehyde 132  
 anisaldehyde [4-methoxybenzaldehyde] 112, 118, 125  
 2-phenylacetaldehyde 88, 104, 108, 111, 118, 123, 124, 125, 132, 134  
 3-phenylpropanal 81, 83, 134  
 cinnamaldehyde 81, 83, 116, 134

KETONES

acetone 79, 80, 93, 94, 95, 101, 103, 113, 116, 127, 130, 131, 132, 134  
 2-butanone 81, 83, 84, 90, 93, 94, 101, 103, 116, 117, 120, 134  
 3-hydroxy-2-butanone 81, 104, 112, 113, 116, 118, 123, 125, 132  
 3-buten-2-one 119, 125  
 2-pentanone 81, 83, 90, 93, 94, 101, 103, 113, 116, 117, 120, 123, 134  
 3-pentanone 103, 117, 119, 120  
 1-penten-3-one 103, 111  
 3-penten-2-one 132, 134  
 cyclopentanone 132, 134  
 2-ethylcyclopentanone 124  
 2-methyl-3-pentanone 113  
 mesityl oxide [4-methyl-3-penten-2-one] 113  
 4-hydroxy-4-methyl-2-pentanone 113  
 2,4-dimethylpentan-3-one 113  
 2-hexanone 103, 116, 123, 132, 134  
 2-hydroxy-2,6,6-trimethylcyclohexanone 104  
 2-heptanone 113, 116, 123, 132, 134  
 4-methyl-3-heptanone 119  
 methylheptenone (unknown structure) 103  
 6-methyl-5-hepten-2-one 81, 83, 96, 100, 103, 104, 108, 110, 111, 112, 113, 116, 117,  
 118, 119, 120, 121, 123, 124, 125, 130, 132, 134  
 6-methyl-3,5-heptadien-2-one (unknown structure) 104, 111, 113, 123, 132, 134  
 2-octanone 116, 131  
 3E,5E-octadien-2-one 132, 134  
 2-nonanone 116, 117, 120, 130  
 trans-2-nonen-4-one 111

(Continued)

Table 3. (Continued)

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 ACYCLIC AND ISOCYCLIC COMPOUNDS
 

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KETONES

undecanone (unknown structure)	132
2-dodecanone	116
pseudo-ionone [6,10-dimethyl-3,5,9-undecatrien-2-one, $\Psi$ -ionone]	107, 111, 123
geranylacetone [6,10-dimethyl-5,9-undecadien-2-one]	96, 100, 104, 107, 108, 110, 111, 113, 118, 123, 125, 132, 134
farnesylacetone [6,10,14-trimethyl-5,9,13-pentadecatrien-2-one]	100, 110, 111, 123, 132, 134
$\alpha$ -ionone [4-(2,6,6-trimethylcyclohex-2-enyl-1)-but-3-en-2-one]	110
$\beta$ -ionone [4-(2,6,6-trimethylcyclohex-1-enyl-1)-but-3-en-2-one]	104, 108, 110, 111, 113, 116, 123, 125, 132, 134
$\gamma$ -ionone [4-(2-methylene-6,6-dimethylcyclohexyl-1)-but-3-en-2-one]	113
epoxy- $\beta$ -ionone [4-(1,2-epoxy-2,6,6-trimethylcyclohexyl-1)-but-3-en-2-one]	104, 113, 123
carvone [6,8-p-menthadien-2-one]	125
acetophenone	111, 113, 123, 127, 130
4-methylacetophenone	113
2-hydroxyacetophenone	104, 132, 134
4-methoxyacetophenone	132, 134
4-methyl-4-phenyl-2-pentanone	113
1-phenyl-2-propanone	124, 125
1-phenyl-2-butanone	124, 125

DICARBONYL COMPOUNDS

glyoxal	81, 83, 116, 130
methylglyoxal [2-oxopropanal]	81, 83, 116, 130
biacetyl [2,3-butanedione]	81, 82, 83, 88, 101, 102, 108, 111, 112, 116, 119, 130, 132, 134
2-oxo-3-butenal	103
2,3-pentanedione	88, 111, 130
2,3-heptanedione	113

ACIDS

formic	20, 75, 78, 89, 106
acetic	12, 20, 29, 46, 75, 78, 89, 104, 113, 114, 115, 132, 134
propanoic	104, 113, 114, 132, 134
2-methylpropanoic	113, 114, 132, 134
butanoic	113, 132, 134
2-methylbutanoic	104
3-methylbutanoic	104, 113, 114
pentanoic	113, 114, 132, 134
4-methylpentanoic	113
hexanoic	113, 114, 132, 134
4-hexenoic (unknown structure)	113
heptanoic	113, 132, 134

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 ACYCLIC AND ISOCYCLIC COMPOUNDS (Continued)
 

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ACIDS

octanoic	104, 113, 132, 134
geranic [trans-3,7-dimethylocta-2,6-dienoic]	113
nonanoic	132, 134
myristic [tetradecanoic]	132
pentadecanoic	132
palmitic [hexadecanoic]	91, 113, 132, 134
stearic [octadecanoic]	91, 132
oleic [cis-9-octadecenoic]	91, 132
linoleic [9Z,12Z-octadecadienoic]	91, 114, 132
linolenic [9,12,15-octadecatrienoic]	91, 114
benzoic	113, 132, 134
salicylic [2-hydroxybenzoic]	113
2-phenylacetic	113
cinnamic	113
4-hydroxycinnamic	113

ESTERS

methyl formate	93
ethyl formate	103, 132, 134
pentyl formate	113
phenethyl formate	113
methyl acetate	101, 132, 134
ethyl acetate	84, 86, 93, 94, 95, 101, 103, 113, 116, 117, 120, 132, 134
propyl acetate	95, 102, 109
butyl acetate	103, 112, 116, 117, 118, 134
2-methylbutyl acetate	113
isopentyl acetate	92, 93, 95, 97, 103, 113, 117
pentyl acetate	116, 134
hexyl acetate	116, 117, 120
trans-2-hexenyl acetate	134
3-hexenyl acetate (unknown structure)	103, 117, 120
cis-3-hexenyl acetate	132, 134
trans-3-hexenyl acetate	132
heptyl acetate	116
6-methylheptyl acetate	116
nonyl acetate	116
phenethyl acetate	117, 120
citronellyl acetate [3,7-dimethyl-6-octen-1-yl acetate]	125
geranyl acetate [trans-3,7-dimethyl-2,6-octadien-1-yl acetate]	102, 109
linalyl acetate [3,7-dimethyl-1,6-octadien-3-yl acetate]	102, 109, 124, 125
ethyl propanoate	93, 94
isopentyl propanoate	117
citronellyl propanoate [3,7-dimethyl-6-octen-1-yl propanoate]	102
methyl butanoate	112

(Continued)



Table 3. (Continued)

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 ACYCLIC AND ISOCYCLIC COMPOUNDS
 

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ESTERS

2-butyl butanoate	112
isopentyl butanoate	92, 97, 102, 116
citronellyl butanoate [3,7-dimethyl-6-octen-1-yl butanoate]	102, 109
geranyl butanoate [trans-3,7-dimethyl-2,6-octadien-1-yl butanoate]	102, 109
isobutyl 3-methylbutanoate	117
2-methylbutyl 3-methylbutanoate	113
isopentyl 3-methylbutanoate	92, 97, 102, 113, 117
isobutyl pentanoate	117
isopentyl pentanoate	116, 117
methyl hexanoate	103, 132, 134
ethyl hexanoate	95, 116
butyl hexanoate	92, 97
isopentyl hexanoate	116
hexyl hexanoate	92, 97
isopentyl heptanoate	116
methyl octanoate	132, 134
propyl nonanoate	116
isopentyl nonanoate	116
propyl decanoate	116
isopentyl decanoate	116
methyl myristate [methyl tetradecanoate]	132
ethyl myristate [ethyl tetradecanoate]	132
methyl pentadecanoate	132, 134
methyl palmitate [methyl hexadecanoate]	132, 134
ethyl palmitate [ethyl hexadecanoate]	132
methyl oleate [methyl cis-9-octadecenoate]	132
methyl linoleate [methyl 9Z,12Z-octadecadienoate]	132, 134
ethyl linoleate [ethyl 9Z,12Z-octadecadienoate]	132
methyl linolenate [methyl 9,12,15-octadecatrienoate]	132
ethyl linolenate [ethyl 9,12,15-octadecatrienoate]	132
methyl salicylate	84, 86, 92, 97, 104, 108, 111, 113, 123, 132, 134
ethyl salicylate	132, 134

LACTONES

$\gamma$ -butyrolactone	104, 111, 113, 123, 125, 134
2-methyl-4-butanolide	113
3-methyl-2-buten-4-olide	113
4-pentanolide	113, 123
3-methyl-4-pentanolide	113
4-hexanolide	104, 113, 123
6-hydroxy-5-hexanolide	112
4-octanolide	104
5-octanolide	125
4-nonanolide	104, 132

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 ACYCLIC AND ISOCYCLIC COMPOUNDS (Continued)
 

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LACTONES

- 2,4-dimethyl-2-nonen-4-olide 134  
 dihydroactinidiolide (2,2,6-trimethyl-7-oxabicyclo [4.3.0] non-9-en-8-one) 104, 113,  
 123, 132, 134  
 phthalide 113

SULFUR COMPOUNDS

- hydrogen sulfide 87, 101  
 dimethyl sulfide 87, 93, 94, 98, 101, 111, 113, 132  
 ethylmethyl sulfide 132  
 dimethyl disulfide 103, 113, 127, 132, 134  
 methylpropyl disulfide 113  
 methanethiol 127  
 2-(methylthio)ethanol 111, 113, 123  
 3-(methylthio)-1-propanol 111, 113, 123  
 5-(methylthio)-1-pentanol 113  
 2-(methylthio)-acetaldehyde 111  
 3-(methylthio)-propanal 88, 108, 111, 123  
 methyl-methanethiosulfonate 113

NITROGEN COMPOUNDS

- methylamine 126, 128  
 ethylamine 126, 128  
 dimethylamine 126, 128  
 trimethylamine 128  
 propylamine 126, 128  
 butylamine 126  
 isobutylamine 126, 128  
 dimethylethylamine 128  
 diethylamine 126, 128  
 2-methylbutylamine 128  
 pentylamine 126  
 isopentylamine 126, 128  
 diphenylamine 132  
 3-methylbutanal-oxime 113  
 butanenitrile 103  
 3-methylbutanenitrile 111, 123  
 pentanenitrile 117, 120, 121  
 benzyl cyanide 104, 111, 113, 123, 132, 134  
 3-methylnitrobutane (unknown structure) 113  
 3-hydroxy-3-methylnitrobutane (unknown structure) 113

HALOGEN COMPOUNDS

- trichloromethane 103, 125  
 trichloroethylene 103  
 1,2-dichlorobenzene 103

(Continued)

Table 3. (Continued)

HETEROCYCLIC COMPOUNDS	
<u>OXYGEN-CONTAINING HETEROCYCLIC COMPOUNDS</u>	
furan	127
2-methylfuran	113, 132, 134
2-ethylfuran	103, 132, 134
2-propylfuran	129
2-isobutenylfuran (unknown structure)	129
2-isopropyl-5-methylfuran	129
2-isopropenyl-5-methylfuran	132, 134
2-methyl-5-propenylfuran	129
2-pentylfuran	111, 129, 132, 134
2-hexylfuran	129
2-heptylfuran	129
acetylfuran (unknown structure)	112
2-acetylfuran	111, 113, 117, 118, 123, 124, 125, 132, 134
furfural	79, 88, 92, 97, 101, 108, 111, 112, 113, 116, 117, 118, 123, 124, 125, 127, 129, 130, 132, 134
5-methylfurfural	111, 112, 113, 118, 123, 124, 125, 127, 132
2-acetyl-5-methylfuran	113, 118, 123, 124, 125, 129, 132
2-acetonyl-5-methylfuran	112
methyl-2-furancarboxylate	129
dibenzofuran	132
furfuryl alcohol	112, 113, 118, 123, 124, 125, 129, 132, 134
2-furancarboxylic acid	113
2-methyltetrahydro-3-furanone	112, 123
linalool oxide I. or II. (unknown structure)	129
linalool oxide I. [cis-5-(1-hydroxy-1-methylethyl)-2-methyl-2-vinyl-tetrahydrofuran]	113, 132
linalool oxide II. [trans-5-(1-hydroxy-1-methylethyl)-2-methyl-2-vinyl-tetrahydrofuran]	111, 113, 132, 134
linalool III. or IV. [5-hydroxy-2,6,6-trimethyl-2-vinyl-tetrahydropyran] (unknown structure)	129
1,4-dioxane	132
2,2,4-trimethyl-1,3-dioxolane	103
2,7-dioxo-1,3,3-trimethylbicyclo [2.2.1.] heptane	113
6,8-dioxo-1,5-dimethylbicyclo [3.2.1.] octane	113
<u>SULFUR-CONTAINING HETEROCYCLIC COMPOUNDS</u>	
2-formylthiophene	113, 129
3-formylthiophene	113
2-formyl-5-methylthiophene	113
2-acetylthiophene	113
2-thiophenecarboxylic acid	113
<u>NITROGEN-CONTAINING HETEROCYCLIC COMPOUNDS</u>	
pyrrole	132
2,5-dimethylpyrrole	132

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HETEROCYCLIC COMPOUNDS

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NITROGEN-CONTAINING HETEROCYCLIC COMPOUNDS

2-formylpyrrole	132
2-acetylpyrrole	132
pyridine	88, 113, 132, 134
2-formylpyridine	132
methylpyrazine (unknown structure)	113
2-methylpyrazine	88, 123, 132
2,6-dimethylpyrazine	88, 113, 123, 132
2-ethyl-6-vinylpyrazine	125
2-isopropyl-3-methoxypyrazine	115, 123
indene	132

SULFUR- AND NITROGEN-CONTAINING HETEROCYCLIC COMPOUNDS

2-propylthiazole	113
2-isobutylthiazole	16, 104, 108, 111, 113, 117, 119, 120, 121, 123, 124, 125, 129, 132, 134
2-sec-butylthiazole	113
2-isopropyl-4-methylthiazole	129
benzothiazole	117, 132

NITROGEN- AND OXYGEN-CONTAINING COMPOUNDS

4-butyloxazole	129
5-pentyl-4-ethyloxazole	129
4,5-dimethylisoxazole	113

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mature-green tomatoes. They observed that from alanine mainly carbonyls and from leucine and valine mostly alcohols originated. The  $\alpha$ -alanine aminotransferase was more active than leucine aminotransferase from the same source.

Several authors examined the formation of  $C_6$  aldehydes and alcohols from unsaturated fatty acids (119,141,142). Since free fatty acids do not accumulate in healthy plant tissues, Galliard and co-workers (143) tried to find the explanation for the liberation of these precursors. They investigated the intact tomato tissues for lipids and the composition of lipids. About 50% of the total lipid content was found to be phospholipid/phosphatidyl choline and phosphatidyl ethanolamine (1:1), with free and acylated-sterol glycosides (ca. 18%) and galactosyldiglycerides (about 16%) as the two other most important groups of components. Investigating the fatty acid composition of lipids, the polyunsaturated fatty acids were found to prevail (linoleic acid, 40%; linolenic acid, 26%). On maceration of the tissues, enzymatic decomposition of the lipids proceeds very rapidly. The pathway of complete decomposition is shown in Fig. 1.

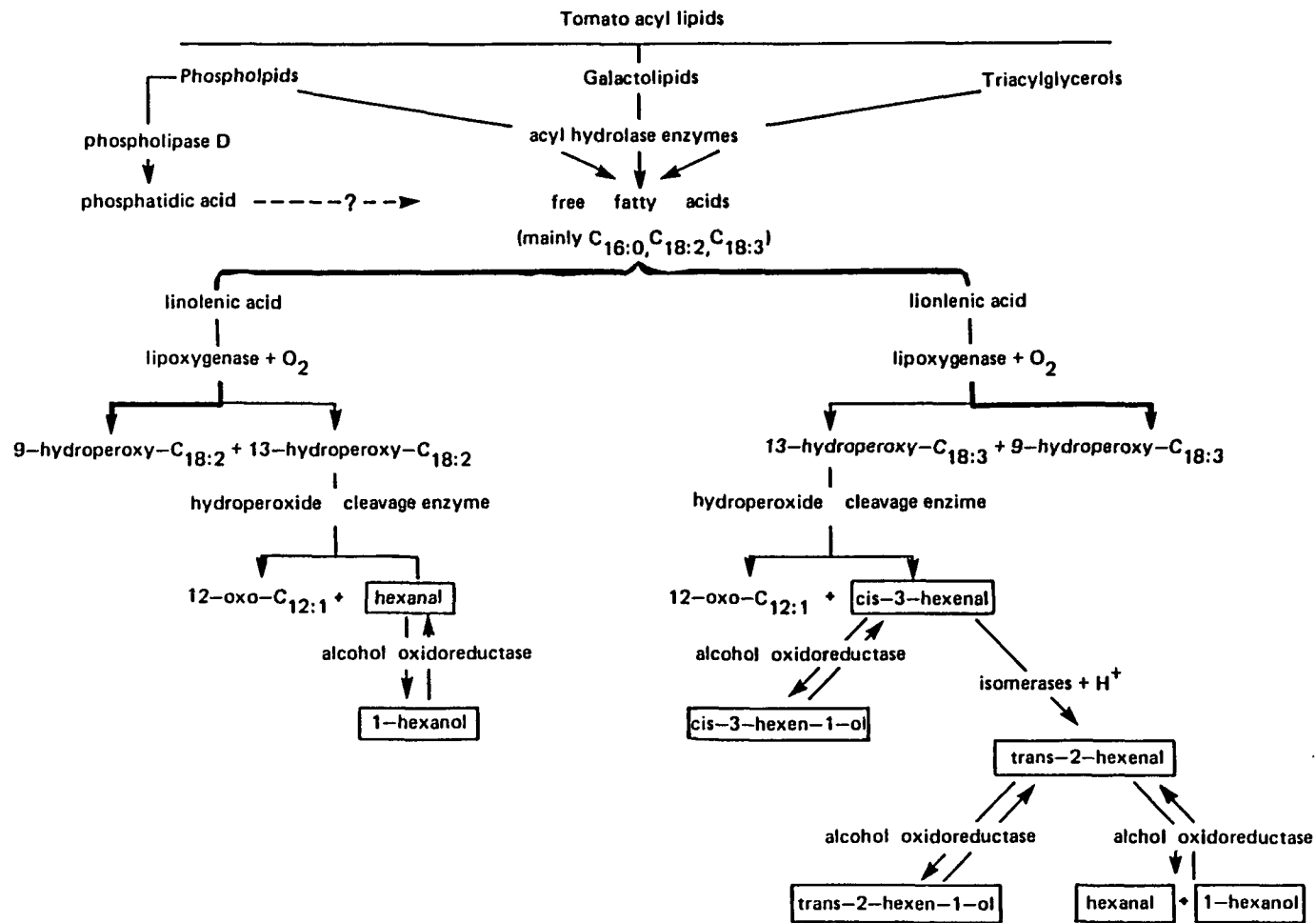


Figure 1. Pathway of the enzymic degradation of tomato acyl lipids (119, 142, 143).

According to experimental results the main product of this decomposition process was *cis*-3-hexenal. The enzyme catalyzing *cis*-3-*trans*-2 transformation is either not present in tomato or is of very low activity.

Investigations of Kazeniac and Hall (108) have shown that *cis*-3-hexenal was not a stable compound. In acidic medium under the influence of heat it is rapidly transformed into the much more stable *trans*-2-hexenal isomer. This finding was supported by the observations of Winter and Sundt (144). From the above it is evident that the distribution of C<sub>6</sub> atom aldehydes and alcohols according to quality and quantity is highly affected by the conditions of sample preparation (whether enzyme activity was inhibited, the amount of oxygen in the sample, extent of comminution, etc.), by the isolation procedure (the extent of heat treatment, if any, etc.).

Schormüller and Grosch (85) observed that among the volatile components of tomato the alcohols tallying with almost all the aldehydes are present and attributed this to the reducing effect of the diphospho-pyridinium nucleotide dependent alcohol dehydrogenase. Meigh and co-workers (145) on the other hand found that the tomato tissue produces acetaldehyde, propionaldehyde, and acetone from the appropriate alcohols by enzyme activity. Thus, an equilibrium is formed between carbonyls and alcohols, depending on the activity of the enzymes present and on other factors influencing the reactions (metal ions, oxygen, etc.).

In addition to the enumerated, many more precursors as well as the transformation of these into volatile substances is described in related literature, without direct observations in experiments related to tomato. The origin of some of the volatiles is not known or only partly known. Thus, Kazeniac and Hall (108) consider thiamine as sulphur precursor of 2-isobutylthiazole, hitherto detected only in tomato. However, Schutte (146) thinks it more probable that it is formed in a biochemical pathway from cysteamine and isovaleraldehyde in the unimpaired fruit.

Partly because of the limits of the experimental methods, partly because of the enzymic procedures instantaneously occurring upon the maceration of the fruit, it is difficult to establish which of the volatile components were present originally in the ripe fruit and which developed during comminution and homogenization. It is, however, probable that some of them are already forming during ripening, among them 2-isobutylthiazole, considered important in the determination of the character of aroma (108). In any case, while for the essential oil content isolated in an inert atmosphere with inhibited enzyme activity at no or very little heating, values of 2-13 ppm were published in the literature (76,84,147), when extraction was carried out in the presence of oxygen under heating and without inhibiting the enzymes, the yield was higher

by one or several orders of magnitude. Buttery and co-workers (111) found about 10 times higher essential oil content using the latter isolation technique.

Of the tomatoes grown, only a smaller percentage is eaten fresh in the raw state; the rest is processed. Therefore it is not without interest how the essential oil composition changes during processing. Depending on the final product, the time and extent of heat treatment is a varying requirement of processing. In the course of processing in the acidic medium reactions take place resulting partly in characteristic and desirable, partly in unpleasant smelling volatile substances. Among the precursors of these, beside the above-mentioned amino acids and fatty acids, the pigments and sugar components of tomato play an important role.

The oxidative decomposition of carotenoids leads to the formation of terpenes and terpenelike compounds. Since the pigments of the fully ripe fruit consist of 50–80% lycopene and 2–7%  $\beta$ -carotene (1,3), the decomposition products of these were detected. In the course of lycopene thermal breakdown Cole and Kapur (148) observed the presence of 6-methyl-5-heptene-2-one, glyoxal, methyl glyoxal, levulene aldehyde, acetone, and levulenic acid. Buttery and co-workers (100) derived from lycopene, too, farnesylacetone (6,10,14-trimethyl-5,9,13-pentadecatrien-2-one), geranylacetone (trans-6,10-dimethyl-5,9-undecadien-2-one), and 6-methyl-5-hepten-2-one. Kazeniac and Hall (108) explained the presence of  $\beta$ -ionone among the volatile components of tomato with the decomposition of  $\beta$ -carotene. They drew attention also to the observation that, in the presence of oxygen, decomposition is more extensive. Onyewu and co-workers (149) surveyed derivatives of carotenoids (particularly  $\beta$ -carotene) formed upon heat treatment of various length and extent. One of them was toluene, detected by several authors in tomato, also meta and para-xylone,  $\alpha$ - and  $\beta$ -ionone, as well as dihydroactinidiolide.

As an effect of the nonenzymatic browning reaction between reducing sugars and amino acids upon heating (Maillard reaction), volatile carbonyl and sulphur compounds are formed, and these may highly affect the character of aroma (80,150–155). Typically these decomposition products are the different furan, pyrrole, and pyrazine derivatives (113,132). 2-Methylmethionine-sulfonium ion was found by Wong and Carson (156) to be the precursor of dimethyl sulphide, considered one of the most important volatile substances in heat-treated tomato products. As an effect of heat, 2-methyl-methionine-sulfonium ion breaks down into homoserine and dimethyl sulphide.

It is evident from the above that fresh tomato, the raw juice prepared from it, and the different—practically additive free—products (sterilized juice, puree, pasta, powder) have different volatile composition. The final composition is determined by the variety used, its stage of ripeness, and the conditions of processing.

### Characteristic Aroma Substances

The rapid increase in the number of volatile substances incited researchers to select from the multitude of detected and identified compounds those considered most important. The less important components participating in the formation of the complete aroma may then be selected and gradually the role of the other volatile components can be determined. Beyond the chemical analysis of the aroma components, this necessitates a psychophysical approach—the study of the aroma properties of individual components and of their interplay.

On looking for the reason for differences in the aroma between varieties, all the researchers agreed that the qualitative composition of essential oils is the same in every variety; differences are to be found only in the quantity of essential oils and in the relative ratios of individual components (95,99,101, 117,134).

Simultaneously, Johnson and co-workers (99) drew attention to the fact that great differences may exist in ratios of the volatile substances within the same variety depending on the time of picking. Thus it is extremely difficult to find the components and quantities characteristic of the variety. When analyzing the volatiles in several descendants of two tomato varieties, Stevens (110) established inheritable differences in the respective concentrations of 2-isobutylthiazole, methylsalicylate, and eugenol. On setting in the samples the sugar-acid ratio and the 2-isobutylthiazole concentration at the same level, sensory judges could not significantly differentiate between them. He concluded from this that in the difference of varietal aromas, 2-isobutylthiazole has a decisive role. He did not exclude, however, the possibility that methylsalicylate and eugenol also affect the aroma, since the sensory threshold value of both compounds is rather low.

Many authors investigated the differences in the aroma of field- and hot-house-grown tomatoes and observed that, with the exception of some compounds (mainly alcohols), the volatile content is substantially higher in field-grown fruit (92,97,117). The compounds of relatively increased concentration (3-hexen-1-ol, 1-hexanol, and isovaleraldehyde) possess a generally "green" odor, thus they may be responsible for an undistinctive, grasslike, weak aroma.

Because of occasionally inevitably extended transport and storage periods, tomatoes are often picked when not fully ripe. Artificial ripening, however, may cause much lower quality, and sometimes off-flavor. Shah and co-workers (102) observed that artificially ripened tomatoes are enriched in short carbon chain ( $C_4$ – $C_6$ ) compounds, while longer carbon chain compounds ( $C_9$ – $C_{12}$ ) diminish. Chung and co-workers (157) came to the same conclusion. Hayase



and co-workers (134) found tomatoes picked in the unripe stage and after-ripened to have a less sweet, more sour taste, and beside the occasionally appearing off-flavor their characteristic tomato flavor was substantially weaker, too. In field-grown, vine-ripened tomatoes terpene-esters dominate, therefore, they concluded that these compounds may be of importance in the fresh-tomatolike aroma.

Dalal and co-workers (92) found increased quantity of 2-butanol, 2-phenyl-ethanol, 6-methyl-5-hepten-2-one, geranylacetone, farnesylacetone, and 2-isobutylthiazole in artificially ripened tomatoes.

Johnson (107) ascribed the appearance of a musty hay smell to  $\Psi$ -ionone. Kader and co-workers (55) found that by exchanging thoroughly the air in the ripening room the appearance of off-flavor can be avoided. Watada and Aulenbach (59) found tomatoes picked in the table-ripe stage much sweeter, their fruity-floral flavor much stronger, and the sensory judges considered it much more desirable than artificially ripened tomatoes.

It was proven by mathematical statistical methods that the desirability and the fruity-floral flavor of tomato are more closely correlated ( $r = 0.83$ ) than desirability and sweetness ( $r = 0.47$ ). Of the substances separated by gas chromatography, five showed close correlation with the fruity-floral flavor. The correlation of the peak areas of the five unidentified substances and the pH of desirability was expressed in mathematical form, too.

On the basis of the above-described investigations much information was obtained on the desirability of individual compounds in the fresh tomato aroma and on the compounds of which enrichment should possibly be avoided.

Following is a summary of those experimental results aimed at the identification of the "key" aroma substances. Although up to the present no such single chemical compound has been found which has in itself a tomato aroma, the researchers have established a number of volatile components to which they contribute the aroma of fresh or processed tomato. With a view to improved lucidity information hitherto published is discussed in relation to individual compounds.

## Aroma Components of Fresh Tomato

### Cis-3-hexenal

In 1954 Spencer and Stanley (76) found a fraction among the volatile components extracted by vacuum distillation that could be characterized as a pleasant "green" aroma and therefore was considered important in the formation of fresh tomato aroma. Although they did not succeed in exactly identifying the compound, they assumed it to be cis-3-hexenal. The same compound was found by Winter and Gautschi (158) as having fresh "green" aroma, while

Hoffmann (159) thought it resembled the aroma of stringbeans. However, 15 years passed until its presence in tomato was proven (104). The explanation for this is obtained in the experiments of Kazeniak and Hall (108), who found that *cis*-3-hexenal is rapidly transformed into *trans*-2-hexenal, which is much more stable. Buttery and co-workers (111) studied thoroughly the aroma properties of the isomer. They determined its sensory threshold concentration and found it extremely low (0.25 ppb). A solution of 1 ppm *cis*-3-hexenal in water resembled the aroma of a freshly cut green tomato. Gaudagni and co-workers (160) studied its aroma profile in a solution of 2 ppm in water and found it decisively of tomato character (76–85%), at the same time slightly reminiscent (10%) of green paprika. On examining the odor of some commercially available tomato flavor products, they were found to differ significantly from that of *cis*-3-hexenal. Added to powdered tomato, its aroma improved substantially. The optimum concentration in tomato juice was found to be 2 ppm, while in powdered tomato it was 1 ppm.

Kazeniak and Hall (108) carried out a similar experiment and found that, added to heat-treated tomato juice or paste, it improves with its “green” character the aroma of these products. They stressed also the desirable blending and mouth-feel properties. In contrast to Guadagni and co-workers (160), they found the optimum concentration to be between 0.3–0.5 ppm, and found that at concentrations of 1 ppm and above, *cis*-3-hexenal produced strongly “green” rancid-type flavor. From the above it appears that *cis*-3-hexenal is an important component of fresh tomato aroma. In contrast Dirinck and co-workers (117), in an aroma concentrate from the headspace of fresh tomato, found a small amount of *cis*-3-hexenal, however, the effect of this upon the aroma of tomato they found to be insignificant: The two latest publications do not even mention it (132,134).

### Trans-2-hexenal

This compound was first detected among the volatiles of tomato by Schormüller and Grosch (80). Subsequently it is mentioned in almost all publications treating the volatiles of tomato, and in most of them it is judged an important aroma component (see Table 3). Although Kazeniak and Hall (108) find it less fresh “green” in character than *cis*-3-hexenal and less intense, its effect on the desirable blending or mouth-feel properties cause it to be similar. The desirable range in heat-treated tomato juice was 0.5–2.0 ppm, however, at higher levels it gave an unpleasant, rancid aroma. It was pointed out that this desirable range was well below the amount found in fresh tomato homogenates (3–10 ppm). They explained this by the fact that the processed, canned tomato juice was low not only in *trans*-2-hexenal but also in many of the other volatile compounds, and when it was added to the juice and flavor interactions with

the other volatiles were less intense. Buttery and co-workers (111) determined the threshold concentration of trans-2-hexenal and found it in aqueous solution to be 17 pp. They found its aroma weaker than that of cis-3-hexenal and less characteristic, not like tomato.

### 2-Isobutylthiazole

The presence of this compound in tomato was first described by Viani and co-workers (104) and was characterized with a tomato leaf green aroma. It is worth noting that so far it has been identified only in tomato.

Kazeniak and Hall (108) found the aqueous solution of the compound to have a spoiled vinelike, slightly horseradish-type flavor. In spite of this, when it was added to canned tomato juice or paste, it intensified the aroma and the harsh, unpleasant notes were weakened, and thus the mouth-feel properties improved. This advantageous effect asserted itself in the 25–50 ppb concentration range. At higher levels, however, the aroma became objectionable and rancid, and medicinal and metallic off-odors became dominant. The threshold concentration in aqueous solution was found to be 2 ppb. It was noticed also that upon slicing, firmly ripe tomatoes containing 2-isobutylthiazole at a high level had the characteristic aroma. In the pulps and juices manufactured from these varieties this unique flavor was also evident. Buttery and co-workers (111) established a threshold concentration of 3.5 ppb, which does not differ substantially from the value observed by Kazeniak and Hall (108). Because of its low threshold value and relatively high concentration, its contribution to the aroma was judged important. Potter (161), however, declared that if somebody would utilize 2-isobutylthiazole as a component of tomato aroma just because he succeeded to detect its presence by gas chromatography would reach catastrophic conclusions.

Chung and co-workers (132), observing that the amount of 2-isobutylthiazole diminished in the course of puree and paste manufacture, considered its role in the aroma of fresh tomato rather important.

Hayase and co-workers (134), sniffing the components eluted in the course of gas-chromatographic separation of the essential oils extracted from tomato, found the aroma of 2-isobutylthiazole grassy and sweet-fruity. Thus, they did not list it as one of the most important aroma components, but based on the publication of Kazeniak and Hall (108) did not exclude its possible importance.

### Hexanal

The presence of this compound in tomato was first described by Schormüller and Grosch (80). According to Kazeniak and Hall (108) it improved with its "green" flavor in the concentration range of 0.1–0.5 ppm the aroma of tomato

juice, although they did not find it as effective as *cis*-3- or *trans*-2-hexenal. Above 0.5 ppm concentration, because of a flavor similar to rancid vegetable fats, its aroma was impaired. Buttery and co-workers (111) found the threshold value in aqueous solution to be 4.5 ppb. Due to the low threshold value and fresh "green" smell it is generally considered an important component in the development of fresh tomato aroma (117,122,134).

#### Cis-3-hexen-1-ol

This compound was detected among the volatile substances in tomato in 1954 and was described as an important component because of its "green" odor (76). This was later supported by several authors (84,108,117,134). The sensory threshold concentration was found to be 70 ppb in water (111).

#### 2E,4E-Decadienal

The presence of this compound in tomato was first described by Viani and co-workers (104). According to the investigations of Kazeniak and Hall (108), it has rather unique flavor properties. When added to tomato juice or diluted paste within concentration limits of 2.5–10.0 ppb it advantageously affected the aroma. The mouth-feel or blending properties became very desirable, the harsh, acidic-type notes were smoothed out. At higher concentration levels, however, the objectionable, rancidlike notes became dominant. Buttery and co-workers (111) considered it, because of the low threshold concentration (0.07 ppb), an important component of the aroma of fresh tomato.

#### 6-Methyl-5-hepten-2-one

This compound's presence in tomato has been described by several authors (Table 3). The first author to identify it was Grosch (12). As to the aroma properties, opinions are divided. Kazeniak and Hall (108) found it to possess fruitlike aroma. In spite of this, when added to tomato juice or paste in the highest concentration it decreased their tomatolike aroma and resulted in a flat, insipid flavor. At a concentration of about 0.75 ppm, the product acquired a cooked, stewed tomato flavor. Sieso and Crouzet (162) considered it significant in the formation of the aroma of heat-treated products. Dalal and co-workers (92) have shown that in after-ripened tomato its amount was increased. In the opinion of Guadagni and co-workers (160), 6-methyl-5-hepten-2-one did not have an important effect upon the aroma of tomato juice. Dirinck and co-workers (121) on the other hand, considered it an important aroma substance in fresh tomato.

Following are listed the compounds not investigated as thoroughly as the previous ones but considered at least by one researcher to be an important

component of the aroma of fresh tomato: citronellal, citral (82); l-hexanol, isovaleraldehyde (97); trans-2-pentenal (108); valeronitrile (121); geranylacetone, farnesylacetone, trans-2-hexen-1-ol (134); 4-butyloxazole, 4-ethyl-5-pentyloxazole (129).

### Aroma Substances in Heat-Treated Tomato Products

In the course of processing tomatoes important changes occur in the volatile substance composition. Several authors explain the appearance of the "cooked" aroma as a result of the reduction of "green" aroma components (108,163). Guadagni and co-workers (98), however, found that panelists gave preference to heat-treated tomato juice over fresh juice. They concluded from this that upon short heat treatment, compounds are formed which lend a characteristic desirable aroma to the product. These compounds are listed below.

#### Dimethyl Sulphide

Dimethyl sulphide was first detected in tomato products by Miers (87) while he searched for the compounds responsible for the so-called "cooked" aroma. Later Nelson and Hoff (101) established that this compound is not present in the raw tomato but is abundant in tomato products. The investigations of Kazeniac and Hall (108) have shown that, depending on its concentration and the food product in which it was used, dimethyl sulphide produced rather unique flavor notes. Guadagni and co-workers (98) found dimethyl sulphide to be forming in the first 20 minutes of heat treatment in linearly increasing quantity. Subsequently, the rate of increase slows and approaches a limit value. The absolute quantity depends on many factors (variety, season, etc.). The maximum quantity measured in tomato juice was between 4.2 and 9.8 ppm. The role it plays in the aroma of heat-treated products was considered important. Buttery and co-workers (111) found its threshold value as measured in water 0.3 ppb and attributed decisive significance to its contribution in the aroma of cooked tomato.

Guadagni and co-workers (160) studied the aroma profile of the 1 ppm solution and found it 44–50% characterized by tomato flavor, however, several panelists found it reminiscent of onion or cabbage flavor. In other experiments they did not find it effective in itself; if, however, added to cis-3-hexenal, it significantly improved the flavor of tomato powder. They established further that the flavor of several commercial aroma products did not differ significantly from that of dimethyl sulphide.

Chung and co-workers (132) detected the presence of dimethyl sulphide in raw tomato in minute quantity when using a sulphur-sensitive detector.

In canned tomato juice the concentration of the compound increased substantially.

### Acetaldehyde

It is noteworthy that this was one of the first aroma substances detected in tomato (72). Its sensory quality and its effect on tomato flavor was investigated only much later. Katayama and co-workers (95) found its quantity highly increased in heat-treated tomato juice. When the juice was prepared from different tomato varieties under uniform conditions, the concentration of acetaldehyde differed substantially. In the varieties investigated by Nelson and Hoff (101) the acetaldehyde concentration varied in the range of 0.2–0.9 ppm. In the heat-treated juice this value increased two-fold. According to Kazeniac and Hall (108) if the quantity of acetaldehyde increased substantially the aroma became characteristically “cooked.” The author of this paper found the threshold value to be 2.5 ppm in aqueous solution and the aroma reminiscent of green plants (164).

In addition to dimethyl sulphide and acetaldehyde, the quantity of other components also increased upon heat treatment. It was also assumed that some of these participate in the formation of the “cooked” aroma. However, different authors considered different compounds important. Thus Kazeniac and Hall (108), doubling the average concentration of 0.3–0.5 ppm of geranylacetone, found tomato juice to acquire heated paste-type flavor notes, with slightly haylike character. If beside geranylacetone, 6-methyl-5-hepten-2-one and 6-methyl-5-hepten-2-ol were also added, tomato juice acquired the typical flavor notes of heated, canned tomato pastes. This is more interesting as each of these compounds had in itself a fruitlike aroma. If the amount of geranylacetone added was further increased in the juice the aroma became objectionable, reminiscent of spoiled watermelon. The 3(methylthio)-propanal accumulating in the heat-treated products had unusual flavor characteristics. When added in concentrations of 1 ppm or above, it masked the typical flavor of tomato juice and produced an insipid flavor. In the opinion of these authors, in addition to the above components, furfural, 2,4-heptadienal, and phenylacetaldehyde contributed to the “cooked” aroma. Their flavor properties, however, were not studied thoroughly.

Buttery and co-workers (111) because of the low sensory threshold concentration, attributed importance to linalool (6 ppb) and possibly to  $\alpha$ -terpineol (350 ppb). Sieso and Crouzet (162) attached importance to furfural and linalyl acetate. Chung and co-workers (132) drew attention to the possible effect of 2-acetylpyrrole and 2-formylpyrrole because of their “burnt” flavor. In addition they mention the probable importance of 2-methylpyrazine and 2,6-dimethylpyrazine.

On studying the flavor properties of the volatile substances found in tomato, Kazeniac and Hall (108) found several compounds that may cause off-flavor in fresh tomato or any heat-treated product. Thus, if 2-octenal was added to tomato juice or diluted paste in 0.1 ppm or about twice the amount found in volatiles from fresh tomatoes produced cardboard-type flavor. Methyl salicylate at 0.03 ppm, the highest concentration found in tomatoes, decreased the tomatolike character of tomato juice and gave flavor notes of canned tomatoes opened and stored refrigerated for a few days. Phenylacetaldehyde at a concentration of 0.5 ppm lost its typical floral note and developed undesirable flavors in tomato juice.

## SUMMARY

Relatively much is known about the formation, chemical, and sensory properties of the compounds causing the fresh pleasant "green" and the characteristic "cooked" flavor of heat-treated tomatoes. It is also probable that the compounds discussed exert their advantageous effect in definite concentration ranges. Below the limit values their effect is less important; above it they affect the aroma in an undesirable direction. This optimum concentration range, however, is difficult to determine for individual compounds because strong interactions assert themselves between the volatiles and other components.

Substantially less is known about the importance of the compounds discussed and their characteristic aroma properties. Reference to this problem is found only in relation to dimethyl sulphide. Guadagni and co-workers (98) found a close correlation between aroma intensity and dimethyl sulphide content in canned tomato juice and concluded that the contribution of dimethyl sulphide to its characteristic flavor is more than 50%.

According to the literature it is evident, too, that a "key" aroma substance solely responsible for the characteristic aroma does not exist. Several authors are of the opinion that compounds responsible for ripe tomato aroma are to be found among compounds of medium or low volatility. Pyne and Wick (84) found that the relatively high-boiling components are major contributors to typical ripe tomato aroma. Dirinck and co-workers (117) also drew attention to the importance of compounds of medium volatility. On separating by gas chromatography the aroma substances found a fraction eluted between carbohydrates  $C_{12}$  and  $C_{16}$  of ripe tomato aroma, but they did not identify individual components. The author of the present paper found (127), on the other hand, that the fraction of highest volatility had fresh ripe tomato aroma. GC-MS analyses enabled the identification in this fraction of acetaldehyde, methanol, methanethiol, diethyl ether (not as contamination), furan, propionaldehyde, acetone, and dimethyl sulphide. The measurement of the aroma

properties of each detected compound was carried out; interactions and their effect upon the aroma is a task for further experiments.

Chung and co-workers (132) detected the compounds of characteristic aroma in the neutral aroma fraction of every product (fresh tomato, tomato juice, puree, and paste). They did not investigate the sensory character of the detected and identified more than 150 volatile substances, thus, it is not possible to establish which of them are responsible for the aroma of ripe tomato.

In spite of the intense research work carried out with up-to-date instruments and the several hundreds of volatile substances detected and identified, the totality of the compounds responsible for the characteristic tomato aroma is still not known.

It is possible that these substances are on the list of known components, but because of insufficiently detailed sensory tests, their actual effect on the aroma has not been established yet. It is, on the other hand, also possible that the most important aroma substances are present in such minute quantities that the available techniques are not sufficiently sensitive to detect them.

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