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Research Progress on the Transformation of Primary Constituents in Aged Tea and Its Impact on Flavor Quality

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Abstract. The aging of aged tea at long term storage is a continuous quality evolution process leading to systematic transformation of main internal substances, thus determining the development and evolution of its flavor quality. However, the current work does not extensively investigate the dynamic transformation rules of key components in the aging process, their synergistic effects, and their intrinsic relationship to flavor quality, nor does the related knowledge appear sufficiently comprehensive or systematically established. In this article, we comprehensively review the transformation pathways and reaction properties of core internal substances including tea polyphenols, amino acids, and alkaloids in aged tea during long term storage; discuss in detail the specific impact of transforming different components on key flavor characteristics such as aroma and taste; and further study the complex network relationship model of synergistic effect between various components, aiming to provide more solid theoretical support and precise scientific guidance for targeted control of flavor and sharp quality improvement of aged tea.

Keywords: Vintage Tea, Flavor, Transformation Patterns, Influencing Factors.

1. Introduction

Tea is one of the most popular non-alcoholic beverages [1]. Tea plays a fundamental role in human culture due to its cultural importance and health benefits.Tea cultivation and use have evolved over centuries to a complex cultural system comprising natural science, humanities, arts, and socio-economics [2]; for instance, flavor quality plays a major role in tea value. From fresh leaves' biochemical composition, and enzymatic change during processing, all steps are oriented towards influencing particular features. While green tea seeks "fresh and crisp", certain tea types aging under specific storage conditions. Their primary ingredients and flavors always evolve, leading to increased sensitivity and market interest over time. Teas whose value varies over time are referred to as aged tea. Based on primary processing and microbial participation, aged tea is mainly classified into two categories: First, post-fermented teas (e.g. Yunnan Pu'er tea [3], and Hunan black tea, whose aging processes are based on the metabolism of dominant microbial communities; Second, aged teas, e.g., Fujian white tea and roasted oolong tea, which mainly yield flavor changes by oxidative polymerization reactions and water activity regulation.

Ageing is a time-dependent phenomenon. Fresh tea is typically very sweet and tannic but its flavour will be systematically altered as a whole: bitterness and stress greatly diminish and soluble

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sugars, pectins, and water soluble pectin-like chemicals are slowly accumulating, providing mellow smooth and smooth, and velvety tactile and gustatory characteristics of the brew. At the same time, aged aromas will gradually appear, such as woody, herbal, or datey, with distinct differences from fresh leaves.Different tea flavors vary after aging: Pu'er tea has an "aged charm", deep red, translucent liquor, and mellow flavour [4]; White tea will turn sour while honey leaves will gradually fade, and it is found that the "herbal fragrance" is the key identifier of aged white tea [5] while black tea can be a special "fungal floral flavor" and woody flavor by microbial activity [6].

Research on key ingredient transformations in aged teas under different types of aging offers insights into their impact on flavor quality. Hence, this article studies how key flavor compounds change during aging, analyzes and summarizes previously published works on how these compounds affect aged tea, providing objective notions about how internal substance transformation affects aged tea flavor quality, seeking to investigate the relationship between aging and flavor, by quantifying physicochemical parameters during aging through quantitative adjustment. The theory supports the standardization and economic development of aged tea industry.

2. Transformation patterns of tea polyphenols and their role in flavor regulation

Tea polyphenols represent an aggregate term for polyphenol compounds in tea leaves with about 20%-35% dry weight of tea leaves. They are the fundamental ingredients of tea and its functional components. Depending on their chemical properties, they can be divided into catechins, flavonoids, or flavanols, which usually occur in the largest percentage of catechin [7]. Tea polyphenolic compounds are responsible for the astringent response of salivary proteins in the mouth that leads to the astrringency effect. Their concentration, which is the relation of percentage of ester-type catechines, determines the bitterness intensity and astringency of tea liquor and is important in the flavor of aged tea.

2.1. Transformation pathways and content changes of tea polyphenols during aging

At aging time, tea polyphenols convert to ester-type catechins and to non-ester-type Catechins as well as simple phenolic compounds. Their effects are strongly dependent on aging time and the physicochemical conditions (like humidity and temperature). Early studies suggest that oxidation and degradation of tea polypholecules (catechins) in green tea are accelerating with increasing relative air humidity, because higher moisture content promotes the converting of insoluble phenolic compound into soluble tea polyPholecules and consequently rapid oxidative reactions [8]. Chen studied Bai Mu Dan tea cakes at different aging times [9]. They found that non-enzymatic oxidation of polyphenole in aging convert them into other substances, and their content decreases. They found in Bai MuDan tea cakes that polyphenol content decreases with aging age.

Hence, the polyphenol content of aged teas drops in storage as a consequence because in storage the polypholic compounds undergo chemical reactions like auto-oxidation and polymerization, leaving non-esterified catechins and simple phenolic compounds.

2.2. Effects of tea polyphenol transformation on astringency, mouthfeel, and color of tea infusion

The above results suggest that aging reduces the content of tea polyphenols and produce nonester-type catechins and simple phenolic compounds leading to the flavor of aged tea: tea poly-phenols give their aroma when dissolved in water and the solution is pale yellow as well as greenish-brown

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[10]. This lower content of the ester-type Catechins (leading astringent compounds) considerably diminishes the harsh astringency, while milder phenolic acids and other transformed products increase the mellow content of this tea liquor.At the same time, by oxidising the polyphenolic compounds during aging, the polymerization contributes to the brownened tea liquor to increase the color of the tea liquor and reduce its flavor. This chemistry is one of the ingredients behind the transformation from greenish--yellow to reddish-green tea liquor [11]. Thus, oxidation, polymerization and degradation of the teaphenol during aging contribute to the tea flavor evolution of aged, which is darker and more mellow.

3. Metabolic conversion of amino acids and theanine and their synergistic effects on flavor

Amino acids are a significant component of nitrogen-containing tea leaves, responsible for around 1%-4% of dry weight and for free leaf cell vacuoles. Twentysix amino acids have been found in tea leaves with the largest proportion being 50%-70% of all tea amino acids; the remaining major amino acids are glutamic acid, leucine, serine and phenylalanine, proteinogenic amino acids [12]. As the main flavor sources in tea, variations of different free amino acid components directly affect the flavor of tea and they are one of the major factors for the quality [13].

3.1. Aging metabolic pathways and content changes of free amino acids and theanine

Free amino acids such as theanine commonly drop in aging and act as substrate of other flavour components.For Bai Mu Dan tea, Chen et al. [9] estimated that total free amino acid content decreases over aging years as a result of conversion, polymerization, or degradation of free amino acids in aging. For black tea, Zhao et al. [14] found that free amino amino acid contents decreased when storing more time. They proposed that when water soluble proteins degrade faster at higher storage time amino acids react with soluble sugars and polyphenols with certain temperature and humidity and gradually decrease total amino acid level. For Black tea and green tea, Yang et al. [15] calculated that amino acid reaction with indan-3-one reagent increases with storage time (within 10 years). They also found that tea leaves have higher amino acid levels than broken leaves and hence their earlier findings could come from initial fluctuations in amino acid type and concentrations between tea types.

Huang et al. [16] investigated the amino acid anabolism and transformation processes for tea leaves, indicating that the amino acids were initially formed by protein hydrolysis, oxidative polymerization with quinones, microbial action, or Maillard reaction with carbonyl compounds (e.g. reducing sugars). L-theanine is an example of the growth of an amino acid in a fresh tea leaf: Liu et al. [17] found that their expression levels for genes CsTS2, CsGS1, and CsGDH2 were correlated with the initial L-thinal in fresh tea leaves. During long storage periods amino acids will remain decreased due to Strecker degradation by oxidation of catechin quinone. High temperatures and humidity lead to higher deterioration [18]. Also, theanine content typically decreases after 10 years of storage. This may be related to N-ethyl-2-pyrrolidone-substituted flavanol from catechin derivatives [19].

3.2. Contribution of amino acid metabolism to tea's freshness, aged aroma, and bioactivity

Free amino acids are highly important in the flavour and texture of tea and the free amino acids contribute to the flavor of brewed tea [20]. Theanine gives the impression of umami, which can be soluble complexes of other amino acids by hydrogen bonds and hydrophobic processes. During

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aging, the complexes merge or aggregate and develop phenolic-amino complexes that lead to freshness. Chen's study of Bai Mu Dan tea suggests them may be quinone-like substances [9], that contribute to tea liquor color. For most aged teas, although their total free amino acid content is decreasing, their degradation products contribute positively to the flavour of aged tea liquor. Additionally, melanoidins of amino acids and soluble sugars by moderate Maillard reactions, as well as oxidation products like catechins, form the foundation of "age aroma", which indirectly enhances the harmony and roundness of tea liquor [13]. However, Zhang et al..'s study on free amino amino acids in tea showed that these in aged tea offer human health advantages such as anti-tumor, antioxidant, neuroprotection, and sleep [13]. This suggests that free aminoacid in aged Tea not only contribute to flavor but also have multiple beneficial biological activities for human health.

4. Stability of alkaloids during aging and their fundamental role in flavor development

Alkaloids are four-fold words for nitrogen-containing heterocyclic secondary metabolites in tea leaves, which represent ~3–5% of the dry weight. Caffeine is the largest one, accounting for more than 90% of alkaloids; small amounts of theobromine, theophylline, and methylxanthine are also present [21]. Caffeines are mainly produced by tea plants through the purine nucleotide degradation process. It is produced in the leafs and buds of new shoots as an important chemical indicator for tea grading.

4.1. Characteristics of caffeine content stability during aging

Contemporary studies suggest that caffeine is relatively stable in aging and does not change much when stored on long time intervals. From aged white tea, black tea, red tea, and green tea stored for longer time periods, no change has been observed. Zhao et al. [15] suggest that this can be due to the stable chemical properties, its preservation from degradation and oxidation.

4.2. Caffeine's regulation of vintage tea's bitter undertones and sweet aftertaste

Caffeine is bitter and a favorite flavor ingredient in aged tea. Its strong chemical structure remains constant throughout aging and is a source of the "bitter base" flavour in aged Tea [22]. The degradations of strongly astringent catechins, and the decrease of umami enhancing chemicals such as theanine help to mask the umami effect and to make coffee bitterness more distinctly present.

5. Degradation and transformation of sugars and their contribution to sweetness and mellow flavor in tea infusion

Sugars constitute 20% to 25% of the dry matter in tea leaves. Beyond cell walls, they include free sugars, bound glycolipids, glycoproteins, and other polysaccharides responsible for imparting sweetness to tea [23]. With extended storage, the gradual degradation of polysaccharides like cellulose and starch in aged teas may increase soluble sugar content, contributing to a "sweet and mellow" sensation. This phenomenon was validated in Huang et al..'s study on aged Fuzhuan tea [24].

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6. Transformation network of key components in vintage tea and synergistic mechanisms of flavor development

6.1. Multi-component interaction transformation network

The final flavor of aged tea is not simply a linear transformation of a single substance; it also forms complex interactions between core ingredients (e.g., tea polyphenols, amino acids and caffeine) at temporal and spatial scales that form a transformation network between major components. For example, when tea is aging, free amino acids auto-oxidation with polyphenol to produce quinone intermediates forming polymers of amino acids or catechins (N-ethyl-2-pyrrolidone-substituted flavanol molecules) after further oxidation [25], that become high-molecular weight brownish yellow pigments that lead to the color of tea liquor [26]. Water soluble substances (the bulk of all soluble ingredients in tea) are therefore regarded as a quality indicator of tea, which in turn reflects the flavor of the tea liquor.

6.2. Flavor synergy: cross-modal sensory experience of taste and smell

This transformation network then plays an important role in the simultaneous flavor changes. For example, catechin hydrolysis and oxidative polymerization reduce astringency intensity, while amino acids reduce umami by deamination and decarboxylation processes.These two effects decreases freshness and crispness of tea liquor, which in turn enhance aged tea.The bitter flavour of caffeine, the light mouthfeel due to oxidized tea polyphenol products, and the sweetness due to amino acid degradation products and soluble sugars combine the distinct flavor of aged tea to produce hybrid flavor features.Therefore, aged tea is seen to share a cross-modal flavour with aged-aroma volatile compounds.As a taste factor, bitterness activates the reward circuitry of the brain as well as the smell perception of aged aroma and woody notes which forms aged tea's distinct multidimensional sensory experience of "richness-bitterness-sweetness-aged aroma" [27,28].

7. Conclusion

In the presence of aging, tea is transformed from "fresh, crisp, stimulating" to "mellow, harmonious and aged". This perception feature is fundamentally motivated by the dynamic chemical structure of three major external factors: tea polyphenols, amino acids and alkaloids, and other ingredients and mechanisms in combination with time and microenvironment. In the way of transformation direction: Tea polyphenol controls astringency and color; Amino acids control freshness and aged flavor; Caffeine balances the bitter basis. By the transformation networks and synergy among the components, the three major components form the diverse perception of aged tea with "smellow, smooth, sweet, aged'.

Although recent studies have shown basic component transformation patterns in aged tea, there are still limitations: First, we do not understand the molecular mechanism underlying synergistic transformation among components, which hinders targeted regulation of flavor compounds. Second, aging mechanisms between different tea types (post-fermented teas, aged teas) are still poorly understood. In future work, multi-omics technologies (e.g., metabolomics, microbiomics) should be used to fully understand transformation networks. Inspired by carbon-nitrogen stable isotope ratio-based Pu'er tea certification systems, standardized aged tea quality assessment and control should be developed. Exploring the role of microorganisms in aging process (a new research direction) and

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thoroughly studying these dynamic transformation mechanisms will form the basis of controllable aging and scientific quality evaluation of aged tea.

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