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Original Article

Relative content of gallic acid over 5-galloylquinic acid as an index for the baking intensity of oolong teas

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ABSTRACT

Phenolic compounds in a series of old oolong teas prepared by baking annually were monitored and compared. The results showed that the relative content of gallic acid over 5-galloylquinic acid was subsequently elevated during this preparatory process. To reveal the effect was mainly resulted from baking or aging, two sets of oolong teas were collected and examined; one set was generated from fresh oolong tea via continually daily baking and the other set was composed of aged oolong teas with no or light baking in the storage period. The relative content of gallic acid over 5-galloylquinic acid was observed to be subsequently elevated when oolong tea was continually baked at 90, 100, 110, and 120 °C for 8 h day after day. In contrast, the relative contents of gallic acid over 5-galloylquinic acid in aged oolong teas with no or light baking were found to be similar to or slightly higher than that in fresh oolong tea. The results suggest that the relative content of gallic acid over 5-galloylquinic acid seems to be a suitable index for the baking intensity of oolong tea in different preparations.

1. Introduction

Tea is regarded as one of the most consumed beverage in the world [1–3]. In the past few decades, several phenolic compounds, such as epigallocatechin-3-gallate (EGCG), gallic acid, strictinin, and teaghrelin, in tea infusion have been demonstrated to provide a variety of beneficial functions to human health [4–11]. According to the different ways of processing, tea is mainly classified into three basic types, green tea (unfermented), oolong tea (partially fermented), and black tea (fully fermented), where "fermentation" refer to natural browning reactions caused by oxidative enzymes in the cells of tea leaves [12,13]. Oolong tea is produced predominantly in Fujian and Guangdong of China as well as in Taiwan. In the past century, different types of oolong teas have been innovatively produced via versatile processes in Taiwan, and are highly appreciated by Taiwanese due to their special taste and flavor [14].

Among different types of oolong teas, old oolong tea has become more and more popular in Taiwan as well as in China recently [15]. Old oolong tea is commonly referred to those

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oolong teas stored for a certain period of time, e.g., more than five years, and may be occasionally refined by baking during the storage [14]. Empirically, the longer oolong teas are stored and further oxidized gradually, the better they are in terms of taste. Furthermore, oolong teas with relatively high degrees of fermentation are thought to have a better chance to be converted into superior old oolong teas. A major type of old oolong teas commonly practiced in Taiwan utilized periodical baking refinement at least once a year during the aging process. Besides the fermentation process, long-term storage (aging) and professional baking are typically regarded as two major factors for the quality control in the preparation of old oolong teas [16]. As expected, different combinations of aging and baking processes have led to the commercial production of diverse old oolong teas with distinct tastes empirically [14]. According to liquid chromatography tandem mass spectrometry (LC/MS/MS) and gas chromatography/mass spectrometry (GC/MS) analyses, the phenolic and volatile compounds of oolong teas were evidently altered under the baking and aging processes [17,18].

As significant difference in chemical constituents was observed between fresh oolong teas and old oolong teas, we wondered if any alteration of chemical constituents in oolong teas could be assigned as a characteristic index for the effects of the baking and aging processes in the tea conversion. In this study, a series of old oolong teas prepared by baking annually were obtained from a local tea manufacturer in Taiwan, and their phenolic compounds were analyzed and compared. Strikingly, the relative content of gallic acid over 5-galloylquinic acid was found to be subsequently elevated during the conversion process of old oolong tea. To reveal the elevation of gallic acid over 5-galloylquinic acid was mainly resulted from baking or aging, two sets of oolong teas were collected and examined; one was prepared from fresh oolong tea by daily baking and the other was composed of aged oolong teas with no or light baking in the storage period. The results indicated that baking intensity seemed to be responsible for the elevation of gallic acid over 5-galloylquinic acid in old oolong teas.

2. Methods

2.1. Chemicals and materials

All chemicals were purchased from E. Merck Co. (Merck KGaA, Darmstadt, Germany) unless stated otherwise. High performance liquid chromatography (HPLC) grade acetonitrile and methanol were purchased from Fisher Scientific (Fair Lawn, NJ, USA). Acetic acid (99.7%) was obtained from J.T. Baker (Mallinckrodt Baker, Inc., Phillipsburg, NJ, USA). Purified water was afforded by a Millipore clear water purification system (Direct-Q, Millipore, Billerica, MA, USA).

2.2. Tea samples

Fresh Chin-shin oolong tea was prepared from tea plants (*Camellia sinensis* L.) grown in Mountain Ali, Chayi County, Taiwan in 2015. A series of old oolong teas prepared by baking annually and old oolong teas with no or light baking (a few times at temperature lower than 100 °C) in the storage period were gifts from local tea manufacturers. To obtain a set of baked oolong teas, fresh oolong tea was subjected to daily baking for 8 h at 90, 100, 110, and 120 °C subsequently by a drier composed of two parts, an electronic heater made of stainless steel (60 cm in diameter) and a tea container with a steel net at the bottom (58 cm in diameter) encircled with dry bamboo sheet (40 cm in height).

2.3. Preparation and HPLC analysis of tea infusions

Tea infusions were prepared by adding 20 mL of boiling water to 1 g of fresh, old or baked oolong teas for 15 min. After cooling to room temperature, the brew was filtered through a 0.22 μm polyvinylidene difluoride (PVDF) membrane filter (Pall Corporation, Glen Cove, NY) for the following analysis. Chemical constituents in oolong tea infusions were analyzed by HPLC system coupled to a 600E photodiode array detector (Waters Corporation, Milford, MA), and separation was performed on the Syncronis C18 column (4.6 × 250 mm inner diameter, 5 μm, Thermo Scientific, Waltham, MA, USA). The separated condition of HPLC analysis was modified according to Shih et al. [19]. The mobile phase consisted of (A) water containing 0.5% acetic acid and (B) acetonitrile. The program for gradient elution started at 95% solvent A and 5% solvent B, increased linearly to 77% solvent A and 23% solvent B in 70 min. The column was maintained at room temperature and the injection volume was 5 μL at a flow rate of 1 mL/min. The ultraviolet (UV) absorbance detection wavelength was set at 280 nm. Epigallocatechin-3-gallate (EGCG), caffeine, gallic acid (GA), and 5-galloylquinic acid (5GA) shown in the HPLC profiles of this study were identified according to the same procedure as described previously [10].

2.4. Detection of UV-Vis spectra of tea infusions

To detect UV–Vis spectra, tea infusions prepared as described above were cooled down to room temperature for 30 min and filtrated through the PVDF membrane filter. The filtrates of 200 μL were loaded onto a 96-well UV transparent plate (Corning, Corning, USA), and UV-vis spectra were scanned from 250 to 600 nm by the Infinite® M200 pro microplate reader (TECAN, Austria, Switzerland).

3. Results

3.1. Tea leaves and infusions of a series of old oolong teas baked annually

To examine the effects of preparatory processes on the outward appearance of old oolong teas baked annually, tea leaves and infusions of fresh, 10-year, 15-year, and 20-year old oolong teas were compared (Fig. 1). In comparison of the fresh oolong tea and old oolong teas, the dried tea leaves seemed to transform from dark yellow green to dark brown or light black after the preparatory processes. Tea infusion color of fresh oolong tea was yellow green while that of old oolong teas was brown red. Tea leaves of old oolong teas as well as those of fresh oolong tea could fully expand to their original sizes when they

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