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Analysis of lipophilic compounds of tea coated on the surface of clay teapots

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ABSTRACT

The surface of a clay teapot tends to be coated with a waterproof film after constant use for tea preparation. The waterproof films of two kinds of teapots (*zisha* and *zhuni*) used for preparing oolong tea and old oolong tea were extracted and subjected to gas chromatography–mass spectrometry analysis. The results showed that comparable constituents were detected in these films; they were primarily fatty acids and linear hydrocarbons that were particularly rich in palmitic acid and stearic acid. To explore the source of these two abundant fatty acids, the fatty acid compositions of fresh tea leaves, granules, infusion, and vapor of infusion were analyzed by gas chromatography. Fresh tea leaves were rich in palmitic acid (C-16:0), unsaturated linolenic acid (C-18:3), linoleic acid (C-18:2), and oleic acid (C-18:1), which were presumably from the phospholipid membrane. During the process of manufacturing oolong tea, the three unsaturated fatty acids may be substantially degraded or oxidized to stearic acid (C-18:0), which was enriched with palmitic acid in the tea granules and in the infusion. The vapor of the tea infusion is primarily composed of palmitic acid and stearic acid. Thus, the coated films of teapots mostly originated from the lipophilic compounds of the tea infusions.

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1. Introduction

Tea is a very widely consumed beverage around the world, and many compounds in tea infusions have been identified with beneficial health functions [1–3]. Various teas are produced by versatile processes, and primarily classified as green tea (i.e., unfermented), oolong tea (i.e., partially fermented), and black tea (i.e., fully fermented), in which "fermentation" refers to natural browning reactions caused by oxidative enzymes in the cells of tea leaves [4]. Oolong tea, which possesses a taste and color between green tea and black tea, is manufactured primarily in Fujian and Guangdong in China, as well as in Taiwan. In the past few decades, different types of oolong teas with a fermentation degree in the range of 20–80% have been highly appreciated by Taiwanese people because of their special taste and flavor [5].

Old oolong tea generally refers to oolong tea that has been stored for >5 years and refined annually by a professional drying process at various desired temperatures [6]. Long-term storage (i.e., aging) and professional baking in a specialized oven at various desired temperatures are generally regarded as two major factors for the quality control of old oolong teas [7]. The longer oolong tea is stored and further oxidized gradually, the better it is empirically in taste and beneficial effects to human health. According to analyses by liquid chromatography tandem mass spectrometry (LC/MS/MS) and gas chromatography–mass spectrometry (GC/MS), the phenolic and volatile compounds of teas are significantly altered under the baking and aging processes [8–10].

To enhance the aroma, color, and taste of oolong tea, an infusion is traditionally prepared by immersing oolong tea granules with hot water in clay teapots. Clays such as *zisha* and *zhuni* are used to make teapots in Yixing, Jiangsu Province, China, and are fired at 1100–1200°C in an oxidizing atmosphere. In general, the major mineral composition of clay is silicon dioxide (SiO_2), followed by aluminum oxide (Al_2O_3) and iron (III) oxide (Fe_2O_3); however, iron influences the color of clay after firing. Of course, the mineral ratio varies substantially in clays dug from different areas [11]. Further observation of the clay teapot microstructure using a scanning electron microscope reveals continuous and discontinuous cavities of approximately 0.01–0.02 mm in the sections of teapots; they occupy approximately 20% of the clay volume. The presence of cavities in clay teapots seems to prevent the latent risk of breakage induced by the drastic thermal expansion during tea preparation with boiling water. Furthermore, water vapor and volatile compounds of the tea infusion in clay teapots probably pass through these cavities.

After frequent tea preparations, the inside of a clay teapot tends to be precipitated with dark brown scum while its surface is coated with a waterproof film. For tea consumers, it is part of enjoyment to observe the gradual change of the cumulative scum precipitation and surface coating in clay teapots, and the practice of this teapot maintenance is regarded as an art in tea culture called "Yang Hu." It has been proposed that the tea scum develops via the formation of complexes with tannin polymers and calcium (Ca^{2+}) ions in the tea infusion [12–15]. By contrast, the chemical constituents of the waterproof film coated on the surface of teapot are largely unknown.

In this study, we aimed to analyze the chemical constituents of the waterproof films coated on the surface of teapots. Because volatile compounds of tea infusions may pass through the cavities of the clay teapots, the waterproof films of *zisha* and *zhuni* teapots used for preparing oolong tea and old oolong tea were extracted and subjected to GC/MS analysis. To explore the source of the coated constituents, extracts of fresh tea leaves, granules, infusions, and vapor of infusion were also analyzed and compared.

2. Methods

2.1. Chemicals and materials

All chemicals were purchased from E. Merck Co. (Darmstadt, Germany), unless stated otherwise. Boron trifluoride, toluidine blue O, and *tert*-butanol were purchased from Sigma-Aldrich Corporation (St Louis, MO, USA). Benzene was purchased from ALPS Chemical Co. (Taipei City, Taiwan). Water was purified using the Millipore Direct-Q clear water purification system (Millipore Corporation, Billerica, MA, USA). Fresh tea leaves (i.e., young green shoots) and tea granules were obtained from the same tea plant cultivar: *Camellia sinensis* L. Chin-Shin oolong was grown in Zhushan and Lugu, Nantou County, Taiwan. *Zisha* and *zhuni* teapots, which were manufactured in Yixing, Jiangsu Province, China, were used for the study of their surface coating.

2.2. Teapot maintenance

To analyze the chemical constituents of the waterproof film coated on the surface of the teapots by a scientific approach, three nearly identical used *zisha* or *zhuni* teapots were cleaned thoroughly with detergents, and were used for the following comparisons: teapot maintenance with water, oolong tea infusion, or old oolong tea infusion. In every tea preparation, 2 g of oolong tea granules or old oolong tea granules were set in a *zisha* or *zhuni* teapot, and 100 mL of boiling water was added to prepare the infusion. Boiling water without tea granules was concomitantly placed in a *zisha* or *zhuni* teapot as a background control. After 3 hours, the temperature of the tea infusion had cooled to room temperature. Thus, the infusions and expanded tea leaves in the teapots were decanted, and the teapots were cleaned by rinsing with running water. The aforementioned process of teapot maintenance was repeated 10 times per week. After 6 months (approximately 250 times), the waterproof films coated on the surface of teapots were subjected to GC/MS analysis.

2.3. GC/MS analysis and mass spectral identification

The constituents coated on the surface of teapots were dissolved in acetone, and the solvent was removed by a