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Effect of vibrating-type ultrafine grinding on the physicochemical and antioxidant properties of Turkish galls in Uyghur medicine

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ABSTRACT

We evaluated the influence of a vibrating-type ultrafine grinding treatment on the physical and chemical properties and antioxidant activity of Turkish gall powder (TGP) with particle sizes of >450, 400–250, 250–100, 100–50, and < 50 μm. The physical properties were analyzed and we found that the small particle size of the TGP increased the bulk (from 0.68 to 0.32 g/cm^3) and tapped densities (from 0.83 to 0.70 g/cm^3) and increased the repose (from 35.43° to 56.28°), slide (from 35.54° to 59.23°), and crash angles (from 35.68° to 42.91°). The total ash content (from 2.34% to 1.75%) decreased, but the moisture index and water solubility index were improved. The extract of TGP with a particle size of <50 μm indicated that highest gallic acid content (9.47 mg/g), methyl gallate content (34.78 mg/g) and ellagic acid contents (0.79 mg/g) accompanied with the best DPPH, hydroxyl radical, and superoxide radical scavenging activities ($p < .05$). Moreover, the FTIR analysis suggested that the ultrafine grinding did not damage the main structure of the components as the particle sizes decreased. The SEM images demonstrated the shape of the TGP with different particle sizes and surface morphology. The microscopic identification exhibited the vessels, oil cells, starch grains, and cluster crystals, which provided referential information in evaluating the Turkish gall quality.

1. Introduction

Traditional Chinese medicine Turkish galls (galls of *Quercus infectoria*) have been used medicinally by Uyghur natives and are commonly known as "Moza" in Uyghur literature [1]. *Quercus infectoria* Olivier (Fagaceae) is an oak shrub species that is mainly present in Turkey, Greece, Syria, and Iran [2], which is a popular Uyghur medicinal plant. Female gall wasps use their ovipositor to stab the young branches of this shrub and spawn. The shrub is stimulated to grow tissue around the larvae. A gall forms when starch grains disappear around the larvae [3]. The main components of Turkish galls are tannin and a small amount of free gallic acid, methyl gallate, and ellagic acid [4]. The galls are used as a constituent of toothpowder and in the treatment of gingivitis and toothache in Uyghur traditional medicine [5]. Additionally, Turkish galls exhibit various biological properties, including antioxidative [2], anti-inflammatory [6], and antimicrobial [7]. Turkish galls should be further considered and investigated for the treatment of colon cancer [1], and their therapeutic potential should be studied.

The active ingredients of drugs mainly exist between the cytoplasm and nucleus. Their efficacy is hardly manifested when the cells are intact [8]. This phenomenon is one of the reasons that the curative effect of traditional Chinese medicine is not evident at high dosages [9]. Ultrafine grinding technology has been broadly applied in traditional Chinese medicine processing [10]. Herbal medicine is ground to dissolve the active components of the cells; hence, the drug can work rapidly and effectively [11]. After ultrafine grinding, only a few intact cells are observed using scanning electron microscopy [10]. The vibrating-type ultrafine grinder is an effective tool for preparing herbal medicines. The herbal material is mixed with the grinding media by using a rod in the grinding chamber where the material is strongly crushed, compressed, and sheared in a short period of time. This grinding technique breaks the cell walls of the plants or animal medicinal materials to improve their bioavailability [12]. The vibrating-type ultrafine grinder is efficient and energy saving. Meanwhile, it can alter the physicochemical properties of the materials, including moisture content, water solubility, and flowability [9]. Moreover, the advancing technology is expanding the application of the vibrating-type ultrafine grinder to other fields, such as in agriculture [13], electronic materials [14], energy-saving technology and other industries [15].

To the best of our knowledge, the effects of the vibrating-type ultrafine grinder on Turkish galls have not been reported. Therefore, this study aimed to comparatively study the five stages of Turkish gall powders (TGPs) and their physicochemical properties and antioxidant

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activities. This study explored the application of the vibrating-type ultrafine grinding technique in Turkish galls. The second objective of this study was to provide a reference in improving the product quality of Turkish galls as medicine.

2. Materials and methods

2.1. Materials

Dried Turkish galls were provided by Xinjiang Ciconhabo Uyghur Medicine Co. Ltd. (Urumqi, Xinjiang province, China). DPPH (1, 1-diphenyl-2-picrylhydrazyl), salicylic acid, tris-hydroxymethyl aminomethane, pyrogallic acid were purchased from Sigma Chemical Co. (St. Louis, MO, USA). Gallic acid standard, methyl gallate standard and ellagic acid reference substances (purity≥99%) were bought from the Chinese Institute for the Control of Pharmaceutical and Biological Products. (Beijing, China). Phosphoric acid and methanol (Fisher, Fair Lawn, New Jersey, USA) were chromatographic grade purity. KBr was FTIR grade purity. All other chemicals and reagents were of analytical grade or better.

2.2. Sample preparation and particle size measurement

The dried Turkish galls were coarsely ground by the disc-mill, and powders were screened through a sieve with 20 mm bore diameter. Then, the powders were ground in 25 °C with TY-L8 vibrating type ultrafine grinder (Jinan tianyu equipment Ltd., China) and were screened by the sieves of different diameter. The grinding conditions were as follows: feeding 1 kg, cryogenic temperature 25 °C, crushing for 8 min. Particle size distribution of TGP was determined by a Microtrac S3500 Particle Size Analyzer (Microtrac Inc., USA) which is a laser diffraction instrument. Eventually, five types of TGP with particle sizes of >450 μm, 400–250 μm, 250–100 μm, 100–50 μm, <50 μm were obtained by the dry method. At least three measurements were recorded for each powder sample.

2.3. Basic physical property analysis

2.3.1. Test moisture of TGP

The detection method of moisture was following the Pharmacopoeia of the People's Republic of China (Chinese Pharmacopoeia Commission, 2015, third section 0832). Each sample was measured in triplicate.

2.3.2. Test total ashes content of TGP

The inspection method of total ashes content was following the Pharmacopoeia of the People's Republic of China (Chinese Pharmacopoeia Commission, 2015, fourth section 2302). Each sample was measured in triplicate.

2.3.3. Test water solubility of TGP

Powder samples (B) were dispersed in distilled water (1:50, *w*/*v*) and mixed in a water bath for 45 min at different temperatures of 40, 55, 70, 85, and 100 °C. The mixtures were centrifuged at 4000 rpm for 20 min [16]. The supernatant solution was dried and weighed (A). The water solubility index (WSI) was calculated as follows:

$$\mathrm{WSI}\ (\%) = \frac{\mathrm{A}}{\mathrm{B}} \times 100 \tag{1}$$

where A was the weight of the supernatant after drying (g) and B was the weight of the powder (g).

2.3.4. Test other physical property of TGP

The repose, slide, and crash angles and the bulk and tapped densities of the TGP were measured using an intelligent powder integrative characteristic tester (BT-1001, Dandong, China). At least three determinations were recorded for each test.

2.4. Chemical components analysis

Chromatography was conducted using an Agilent 1220 Infinity LC-DAD series system (Agilent Technologies, Hewlett-Packard, Germany) under the following conditions: WondaSil C18 column (250 mm × 4.6 mm, 5 μm), 25 °C column temperature, 258 nm wavelength, 2 μL injection volume, and 1.0 mL/min flow rate [17]. The mobile phase composition was 0.2% aqueous phosphoric acid (*v*/v) (A) and methanol (B). The gradient elution was as follows: 0–10 min, 10%–30% B; 10–28 min, 30%–66% B; 28–32 min, 66%–66% B; and 32–40 min, 66%–90% B.

The TGP with different particle sizes was subjected to a reflux extraction with 50% methanol for 1 h at 1:50 (*w*/*v*) solid–liquid ratio. The residue was extracted twice with 50% methanol for 40 min. The supernatant was directly evaporated and dried after high-speed centrifugation. The dry samples were re-dissolved with methanol in a 100 mL volumetric flask. The mixture was filtered through a 0.22 μm nylon membrane filter and degassed by sonication. The samples were injected in triplicate into the chromatographic system. The peak area and sample concentration were calculated using the calibration curve. The gallic acid reference substance stock solution was diluted to 52.60, 63.12, 73.64, 84.16, 94.68, 105.20, and 115.72 μg/mL. The methyl gallate reference substance stock solution was diluted to 204.10, 238.12, 272.13, 306.15, 340.17, 374.18, and 408.20 μg/mL. The ellagic acid reference substance stock solution was diluted to 3.61, 4.81, 6.01, 7.21, 8.42, 9.62, and 10.82 μg/mL. Each concentration was determined in triplicate, and the peak area was plotted in the calibration curve.

2.5. Antioxidant capacity evaluation

2.5.1. DPPH radical scavenging assay

The antioxidant activity of the TGP extracts was assessed by evaluating their DPPH radical scavenging activity in accordance with the method of Zhu et al. [18] with slight modifications. A total of 0.1 mM DPPH was dissolved in 100% ethanol, and 100 μL of this sample in 100% ethanol (0.12–1.65 mg/ml) was mixed well with 100 μL of freshly prepared DPPH. The mixture was incubated for 30 min in the dark, and the reading was obtained at 517 nm by using a microplate reader. The capability to scavenge the DPPH radical was calculated as follows:

$$\mathrm{DPPH}\ (\%) = \left[A_i - \frac{A - A_j}{A_i}\right] \times 100 \tag{2}$$

where A_i was the absorbance of the DPPH solution without the sample, A was the absorbance of the test sample mixed with the DPPH solution, and A_j was the absorbance of the sample without the DPPH solution. IC50 indicated the concentration of the sample at 50% of the DPPH radical scavenging activity.

2.5.2. Hydroxyl radical scavenging assay

The hydroxyl radical scavenging assay was performed in accordance with the method of Zhang et al. with slight modifications [19]. The mixture that contained 10 mM FeSO4 (1 mL), 10 mM salicylic acid ethanol solution (1 mL), 1 mL TGP extract solution (0.31–2.83 mg/mL), and 30% H_2O_2 (200 μL) was shaken in a test tube and then incubated in a water bath at 37 °C for 45 min. The reactant was cooled, and the absorbance of the solution was measured at 510 nm by spectrophotometry. The hydroxyl radical scavenging activity (HRSA) was calculated using the following formula:

$$\mathrm{HRSA}\ (\%) = \left(\frac{A_0 - A_1}{A_0}\right) \times 100 \tag{3}$$

where A_0 was the absorbance of the control and A_1 was the absorbance