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Extraction of polyphenols from black tea – Conventional and ultrasound assisted extraction

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ABSTRACT

Products from plant raw materials gain increasing importance in food-, cosmetics and pharmaceutical industry. By way of contrast, due to lack of detailed physico-chemical fundamentals, existing production processes are economically not optimal designed. This leads to a need for deeper understanding of the processes and furthermore a systematic process and equipment design for the potentially applicable extraction techniques.

Using the example of polyphenol extraction from black tea (Kenya), the conventional and ultrasound assisted extractions are investigated. Here, the state of the art as well as a comparison between the two techniques is in focus. Especially, resulting quasi-equilibria and mass transport kinetics serves as a criteria. The physico-chemical background is discussed taking particle size distributions and scanning electron microscope (SEM) measurements into account. Conclusively, process alternatives are projected and discussed. Hence, the present study makes influences of ultrasound technique on physico-chemical characteristics during extraction a subject of discussion.

1. Introduction

Plant-based products gain increasing importance in fields of nutrition, cosmetics and pharmaceutical industries. In recent years, the FAO (Food and Agriculture Organization of the United Nations) predicted annual growth rates of 6–8% for plant-based medical foods and phyto pharmaceuticals. Actually, in the USA, growth rates for these products lie about 15% and hence are higher than initially predicted [1–3]. Besides plant-based pharmaceuticals, for cosmetics and wellness products as well as plant-based flavors and perfumes double-digit annual growth rates are predicted [4–9]. Here, not only the primal extract can be named as a product, but as well the purified substances out of these extracts.

Pretreatment of the raw material and the extraction of valuable components are first steps in processing. Actually, due to a lack of physico-chemical fundamentals these unit operations are economically not optimal designed. First methodical approaches to design optimal processes are published for the extraction [2,10–12] as well as for further purifications of plant-based extracts [13]. For design and optimization of solid–liquid extraction processes, currently statistical experiment planning and physicochemical modeling are mostly investigated. First and foremost, equilibria as well as mass transport kinetics for target and side components are in focus. Considering technical and economical constraints, these parameters have to be enhanced.

Extraction characteristics and hence the equipment are depending on the processed raw material and the solvent system [14]. For example, the location of target and side components as well as the structure of the raw material or the moisture content can be named as crucial factors on equilibrium and mass transport kinetics. Process intensifications and hence enhanced mass transport kinetics and quasi equilibrium behavior can be achieved by using ultrasound techniques [15]. Using the example of polyphenolextraction from black tea, the influence of ultrasound on mass transport and equilibrium are analyzed and discussed. Differences between conventional and ultrasound assisted maceration are investigated considering resulting particle size distributions and scanning electron microscope (SEM) analyzes.

2. Material and methods

For the analysis and discussion of different process concepts, especially the equilibrium and mass transport kinetic is considered. The fundamentals of enhancements are discussed employing particle size distribution and SEM measurements.

The laboratory experiments are carried out with black tea from Kenya. As extracting solvent pure ethanol as well as a mixture consisting of ethanol/water 90/10 (m/m) are used. The total content of polyphenols is quantified by five stage maceration (each 24 h), fol-

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lowed by 96 h control percolation with 0.1 BV/h to ensure total extraction. This procedure is state-of-the-art [11]. For the total amount of polyphenols and hence a yield of Yi = 100% a loading of $c_{polyphenols} = 211.4\ g/kg_{Dry\ substance\ tea}$ is determined. The dry substance of the extract is defined as a further side component.

Ultrasound-assisted extractions (UAE) were performed in an ultrasonic extraction reactor PEX1 (R.E.U.S., Contes, France) with 14 × 10 cm internal dimensions and maximal capacity of 1 L, equipped with a transducer at the base of jug operating at a frequency of 25 kHz with maximum input power (output power of the generator) of 150 W. The double-layered mantle (with water circulation) allowed the control of extraction temperature by cooling/heating systems. Considering the actual input power from the device is converted to heat which is dissipated in the medium, calorimetric measurements were performed to assess actual ultrasound power, calculated as shown in the Eq. (1) below.

$$P = m \cdot c_P \cdot \frac{dT}{dt} \qquad (1)$$

where c_P is the heat capacity of the solvent at constant pressure (J/g/°C), m is the mass of solvent (g) and dT/dt is temperature rise per second.

Fig. 1 depicts the used equipment for the comparison of mass transport kinetics as well as for the determination of the equilibrium line. Mass transport kinetics and phase equilibria are determined at a constant temperature of 40 °C and a solid/liquid ratio of 1/3 (m/m).

The solid–liquid equilibrium line is determined in multi stage maceration experiments. Each maceration step has to reach equilibrium for this. Due to additional total extractions and closing mass balances for each component in each phase, the equilibrium concentrations in solid and liquid phase can be measured.

To close all mass balances, in addition to polyphenol concentration, the amount of water, dry substance and further side components as well as for the solvent has to be measured in both phases. Table 1 shows the necessary methods.

The total amount of polyphenols is determined using the Folin–Ciocalteu test [16] from Seppal. Here, the adsorption Abs_i is detected at a specific wave length of 620 nm. By using reference solutions with a defined concentration of $c_{Ref} = 3\ g/L$, the concentrations of polyphenols $c_{PP,i}$ in each fraction can be determined via Eq. (2).

$$c_{PP,i} = \frac{Abs_i}{Abs_{Ref}} \cdot c_{Ref} \qquad (2)$$

Fig. 1. R.E.U.S. ultrasound equipment.

Table 1
Materials and methods.

	In raw material	In liquid phase
Total polyphenol	Multi stage maceration	Folin–Ciocalteu test
Dry substance	Dry balance	Dry balance
Water	Dry balance, toluene distillation	Karl–Fischer titration
Solvent mass	Dry balance	Calculation

The dry substance as well as the residual humidity in the raw material is determined using the moisture analyzer Sartorius MA 150. SEM measurements are necessary for analyzing the raw material matrix and hence the quantification of the ultrasonic influence. The measurements are carried out using FEI/Philips XL30 FEG ESEM. Each sample is compared with a reference sample of fresh tea.

3. Therory

3.1. Physico-chemical properties

One way to consider the physico-chemical fundamentals of extraction is to build up models, describing the different phenomena. For model-based process design the determination of associated model parameters is necessary. Models considering equilibrium behavior [17] can be named as well as models considering equilibrium, mass transport kinetic and fluid dynamic [18]. With an increasing amount of physico-chemical data, the experimental effort increases as well as the prediction accuracy of these models. Further models include the named physico-chemical properties and, in addition, the structure of the raw material as well as particle size and loading distributions of the ingredients [11].

The parameters named here are used to identify and discuss the differences between the two types of extraction: Conventional maceration versus ultrasound assisted extraction. Equilibrium and mass transport kinetics are in focus. Nevertheless, the basics of ultrasound extraction are not discussed in detail. In literature the fundamentals as well as fields of application are given for laboratory and industrial scale [15].

3.2. Equipment concepts

The optimal equipment for the extraction of valuable components from plant material depends on the raw material as well as on the processed volume rates significantly [19].

In general, a differentiation between raw material characteristics, physico-chemical parameters and economic factors is made. As raw material characteristics

- Accessibility of target and side components.
- Structure of the raw material matrix.
- Moisture content and swelling behavior of the processed raw material, respectively can be named [14]. Physico-chemical parameters depending on the characteristics are [19].
- Equilibrium.
- Mass transport kinetic.
- Fluid dynamic (depending on the equipment).
- The design of an economically optimized process requires to take further economic factors into account [13].
- Apparative efforts and invetiment cost, respectively.
- Operating cost.

The solvent and energy consumption influences the operating conditions significantly. These costs have to be considered regarding the process design. An optimum between solvent consumption