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Modeling and optimization of ultrasound-assisted extraction of polyphenolic compounds from *Aronia melanocarpa* by-products from filter-tea factory

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ABSTRACT

Aronia melanocarpa by-product from filter-tea factory was used for the preparation of extracts with high content of bioactive compounds. Extraction process was accelerated using sonication. Three level, three variable face-centered cubic experimental design (FCD) with response surface methodology (RSM) was used for optimization of extraction in terms of maximized yields for total phenolics (TP), flavonoids (TF), anthocyanins (MA) and proanthocyanidins (TPA) contents. Ultrasonic power (X_1: 72–216 W), temperature (X_2: 30–70 °C) and extraction time (X_3: 30–90 min) were investigated as independent variables. Experimental results were fitted to a second-order polynomial model where multiple regression analysis and analysis of variance were used to determine fitness of the model and optimal conditions for investigated responses. Three-dimensional surface plots were generated from the mathematical models. The optimal conditions for ultrasound-assisted extraction of TP, TF, MA and TPA were: X_1 = 206.64 W, X_2 = 70 °C, X_3 = 80.1 min; X_1 = 210.24 W, X_2 = 70 °C, X_3 = 75 min; X_1 = 216 W, X_2 = 70 °C, X_3 = 45.6 min and X_1 = 199.44 W, X_2 = 70 °C, X_3 = 89.7 min, respectively. Generated model predicted values of the TP, TF, MA and TPA to be 15.41 mg GAE/ml, 9.86 mg CE/ml, 2.26 mg C3G/ml and 20.67 mg CE/ml, respectively. Experimental validation was performed and close agreement between experimental and predicted values was found (within 95% confidence interval).

1. Introduction

Aronia melanocarpa (black chokeberry) belongs to *Rosaceae* family and originates from North America. Nowadays it is also cultivated around Europe [1]. It was used in traditional medicine, but in recent years the interest for this herb increased also due to its potential use as a food colorant [2] and as a source for valued phytonutrients [3]. It was found that *A. melanocarpa* is one of the richest herbal sources of phenolic compounds and that content of proanthocyanidins, anthocyanins and phenolic acids in these herb is high [4,5]. *A. melanocarpa* berries are one of the richest plant sources of anthocyanins (class of flavonoids): cyanidin-3-O-galactoside, cyanidin-3-O-arabinoside, cyanidin-3-O-xyloside and cyanidin-3-O-glucoside which are responsible for dark red, blue, and purple color of berries [6,7]. 25% of the total polyphenols in chokeberry fruits are anthocyanins [8]. Of aromatic acids, the most dominant are chlorogenic and neochlorogenic [3]. (−) Epicatechin oligomers are dominant proanthocyanidins which represent 66% of chokeberry fruit polyphenols [8]. Due to their strong antioxidant capacity and possible positive effect on human health, proanthocyanidins have a great potential in nutrition and medicine [9]. Compared to other fruit, antioxidant activity of chokeberry is significantly higher [10,11]. Positive effect of *A. melanocarpa*, on human health was subject of numerous studies, one of which showed favorable effect of *A. melanocarpa* in control and prevention of diabetes mellitus type II and diabetes-associated complications [12,13]. Protective effect on colon cancer was confirmed *in vivo* and in *in vivo* studies [4]. In another study, hepatoprotective effect of *A. melanocarpa* was confirmed in rats treated acutely with carbon tetrachloride [14]. Chokeberries could have positive effect on the prevention and treatment of cardiovascular diseases or on risk factors for such diseases [4].

The health benefit constituents from medicinal plants are usually used in the form of extracts, liquid or dry. For the production of quality extracts, investigation on extraction process parameters influence for optimal process set-up is necessary. The conventional solvent extraction techniques are used in order to obtain organic compounds, as constituents of liquid extract from the plant

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materials. These techniques usually have low efficiency which could be significantly improved by using some of the novel extraction techniques, such as ultrasound-assisted extraction (UAE), supercritical fluid extraction (SFE) and microwave-assisted extraction. UAE could be particularly suitable because it is rather cheap and has the lowest instrumental requirements [15]. The enhancement in extraction obtained by using ultrasound is mainly attributed to the effects of acoustic cavitations produced in the solvent by the passage of an ultrasonic wave [16]. Cavitation comes as a result of creation, growth and implosion of gas bubbles under the ultrasonic treatment. These bubbles collapse on the surface of the solid plant material and release high pressure and temperature, which generate shock waves towards the solid surface. Ultrasound also exerts a mechanical effect, allowing greater penetration of solvent into the sample matrix, increasing the contact surface area between solid and liquid phase [15,16]. Increased mass transfer and significant disruption of cell walls come as a result of these combined effects. Ultrasonic waves may also cause some chemical effects which are rather undesirable due to the changes in chemical composition, possible degradation of targeted compounds and production of free radicals within the gas bubbles [17]. Therefore, extraction conditions such as time, temperature, ultrasonic power and frequency must be accurately determined.

The most common and often used approach on the process optimization uses one-factor-at-a-time, where influence of independent variables on responses are investigated one by one, while all other factors are kept under constant values. Behavior of investigated response under different values of independent variables is later represented in set of charts. This approach could be time-consuming and expensive for certain experiments. In addition, possible interaction effects between variables may not be evaluated and misleading conclusions may be drawn [18]. In order to overcome these disadvantages, response surface methodology (RSM) could be applied. RSM is a collection of statistical and mathematical techniques useful for developing, improving and optimizing processes in which a response of interest is influenced by several variables and the objective is to optimize this response [19]. Analyzing the effects of the independent variables, this experimental methodology generates a mathematical model which describes the chemical or biochemical processes [20].

The objective of this study was to valorize a by-product of food industry by extracting valuable phenolic compounds from such source, and by that to produce antioxidant rich extracts for further applications. In this research material used for valorization was by-product of aronia obtained after production of fruit filter tea.

For the production of fruit filter tea dry aronia cake (left after production of aronia juice in juice factory) was used. Such cake in filter tea factory was milled, grind and fractionated. After processing certain amount of material, around 20%, is of particle size lower than the particle size of pores of filter tea bag. It is called "fruit dust". As such it cannot be used for the production of fruit filter tea, therefore its represent by-product of this industrial branch.

For quality utilization of this by-product investigation on process parameters influence (ultrasonic power, temperature and extraction time) and development of mathematical models, which could describe ultrasound-assisted extraction of different groups of polyphenolics from *A. melanocarpa*, will be provided. Obtained models should provide determination of optimal extraction conditions with maximized yield of each investigated response.

2. Materials and methods

2.1. Samples and reagents

In the filter-tea industry, during the production of fine cuts" (herbal material of particle size from 0.315 to 2.0 mm, which is further use for the production of final product, filter-tea) operation like cutting, grinding and milling, sifting and fractionating are applied [21]. After fractionation, certain amount of herbal material of particle size lower than 0.315 mm is produced. Herbal material of particle size lower than 0.315 mm may not be used in the further production of filter-tea, as the particle size of such material are lower than particle size of pores of the filter bag material. Thus, this kind of material (usually called herbal dust "or herbal powder") is discharged from the production as by-product (Fig. 1).

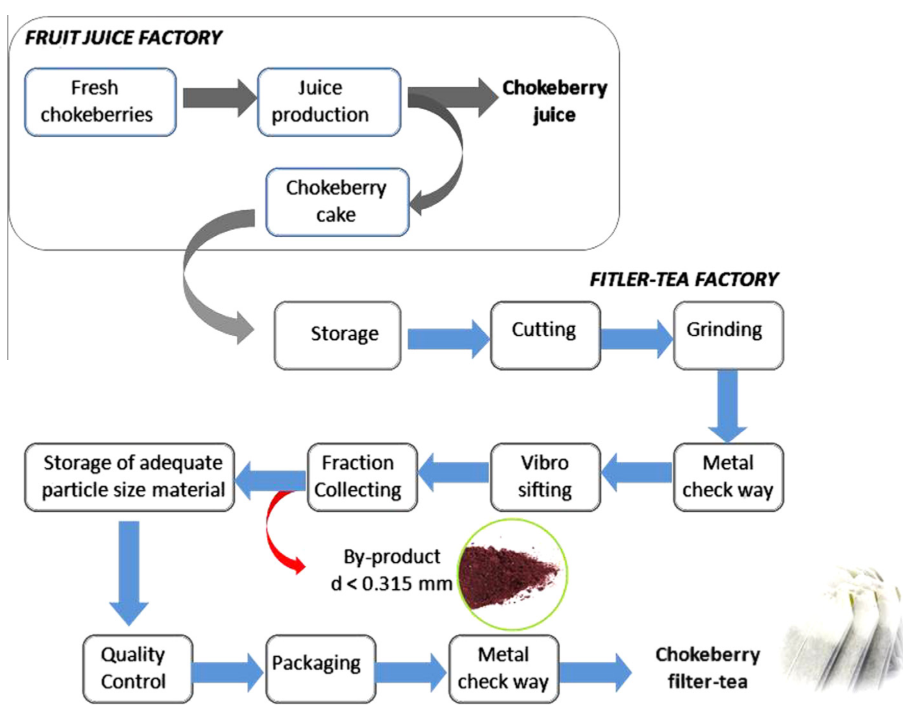

Fig. 1. Production of *A. melanocarpa* by-products.

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