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Pilot scale demonstration of integrated extraction–adsorption eco-process for selective recovery of antioxidants from berries wastes

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ABSTRACT

A new integrated extraction–adsorption process has recently been proposed for selective recovery of antioxidants from black chokeberry wastes. This green process consists in combining extraction of antioxidants and simultaneous enrichment of the extracts in a single operation. The objective of the present work was to demonstrate pilot scale feasibility of this new process for production of larger quantities of antioxidant rich extracts. To prepare better the pilot scale experiment, the process was firstly optimized at a laboratory scale. Best results were obtained at process duration of 8 h and 10 cycles of liquid phase. A scale-up factor of 50 between laboratory and pilot scale was applied for quantities of raw source, adsorbent (both from 4 to 200 g) and solvent (from 0.8 to 40 L). The results obtained at pilot scale were similar to those at laboratory scale, showing that the integrated extraction–adsorption process could be used for production of large quantities of extracts highly rich in antioxidant phenolics.

1. Introduction

Natural antioxidants are subject to growing interest as substitute to synthetic substances for applications in food, cosmetic and pharmaceutical fields. Hence, it is necessary to develop production of antioxidant rich extracts from natural raw sources or their by-products. Fruits are an important source of antioxidant phenolics, particularly red and dark blue fruits and berries, partly due to their high content in anthocyanins. Among the berries, *Aronia melanocarpa* berries (black chokeberries) are one of the richest plant sources of phenolic compounds, especially anthocyanins (mainly cyanidins and pelargonidins) (Galván D'Alessandro et al., 2012; Kulling and Rawel, 2008; Wu et al., 2004). *Aronia* extracts could be used as a food colorant (due to the intense color of anthocyanins) (Bridle and Timberlake, 1997; Kraemer-Schafhalter et al., 1998) or as a natural antioxidant in food and cosmetic industries, especially for limiting lipids degradation (Balasundram et al., 2006). Different bioactivities have also been reported for these berries and their extracts, such as prevention and treatment of cardiovascular diseases (Naruszewicz et al., 2007) and colon cancer (Malik et al., 2003; Zhao et al., 2004), antidiabetes and antimutagenic effects (Gasiorowski et al., 1997), which could be used for applications in healthcare field. *Aronia* berries are mainly used to produce juice (about 90%) (Sojka et al., 2013), which exhibits the highest antioxidant capacity among fruit juices (Kulling and Rawel, 2008). After juice extraction (by pressing), berry wastes contained up to 16% of the initial mass of berries (Baranowski et al., 2009) and still contain high amount of phenolics (up to 8% d.b. (dry basis content)) (Oszmianski and Wojdylo, 2005). These by-products of food industry could be valorized by selective extraction of high added-value antioxidant substances. Nowadays, many investigations are carried out for valorization of food industry by-products by recovering antioxidant phenolics applying classical and emerging extraction and purification processes (Azmir et al., 2013; Casazza et al., 2010; Galanakis, 2012; Karsheva et al., 2013). To enrich the obtained extracts in phenolic antioxidants, the adsorption process using polymeric resins seems to be the most suitable due to its high efficiency, simplicity, easy scale-up and low costs (Buran et al., 2014; Soto et al., 2011). Recently Wang et al. have proposed a strategy to obtain antioxidant rich extracts from pomegranate leaves optimizing firstly extraction step and then optimizing fractionation of the obtained extract by adsorption (Wang et al., 2013).

Extraction efficiency from vegetal sources could be improved by integrating the extraction to a purification step in a single process (Dimitrov et al., 2005; Zhang et al., 2011). Recently, a new integrated extraction–adsorption eco-process has been proposed to produce antioxidant rich extracts from *Aronia* berries wastes

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(Galván D'Alessandro et al., 2013). In this process, liquid phase is circulating through two contactors: an extractor and an adsorption column (Fig. 1). In the extractor, antioxidant substances are extracted from the raw source to the solvent phase, and in the adsorption column, they are discharged from the solvent by fixing on the adsorbent, which enables to separate them from the main part of the others co-extracted molecules and to maintain favorable for the extraction concentration gradient between the raw source and the solvent. This integrated process allowed to recover selectively antioxidant phenolics from black chokeberry by-products in a single eco-process (extraction and purification at room temperature, only food grade solvents used, reduced consumption of time, solvent and energy). Integration of extraction and adsorption steps allowed to enrich about 15 times the extracts in phenolic compounds, improving also up to 25% their extraction yields and preserving their antioxidant capacity in the final products (Galván D'Alessandro et al., 2013).

It is very difficult to obtain the same process efficiency in industrial scale as in laboratory scale. Therefore, scale-up is an important step to evaluate processes before the industrial scale. Usually, pilot scale is used as intermediate between laboratory and industrial scales. If the process is also efficient in pilot scale as in laboratory scale, the process could by industrialized easier.

The objective of the present work was to demonstrate pilot scale feasibility of this new integrated process, which would enable its application for the production of larger quantities of antioxidant rich extracts. To prepare better the pilot scale experiment, optimization of the integrated extraction–adsorption was firstly conducted at laboratory scale.

2. Materials and methods

2.1. Solvents, reagents and adsorbent

Ultrapure water (prepared using a Milli-Q system), ethanol (96%, provided by Flandre Chimie, France) and their mixtures were used as solvents. All reagents were supplied by Sigma–Aldrich (France), i.e. potassium chloride (>99%), sodium acetate (>99.9%), gallic acid (>98%), 6-hydroxy-2,5,7,8-tetramethylchroman-2-carboxylic acid (Trolox) (>97%), Folin–Ciocalteu phenol reagent (2 N), 2,2-diphenyl-1-picrylhydrazyl (DPPH), methanol (p.a. grade), sodium carbonate (>99%) and sodium azide (>98%).

All experiments were carried out with Amberlite XAD7HP polymeric adsorbent (Rohm and Haas, France). Before use, adsorbent was prepared according to the procedure described elsewhere (Galván D'Alessandro et al., 2013).

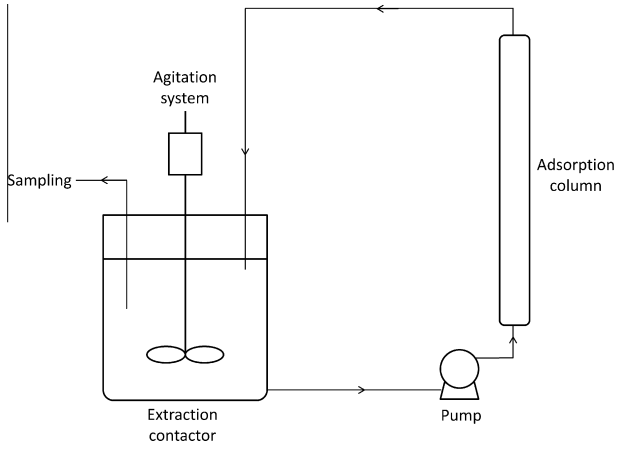

Fig. 1. Schematic representation of extraction–adsorption integrated process.

2.2. Raw source

Berries of *A. melanocarpa* from the region of Elena (Bulgaria) were used. They were passed in a small laboratory extruder to obtain *Aronia* juice. The generated wastes (pellets of about 0.3–0.4 cm in diameter and 0.5–1.0 cm in length) were additionally dried at 45 °C in an oven (Function Line Heraeus T6, IMLAB, Lille, France) for 2 days. The obtained *Aronia* pellets (94.7% d.b.) were stored in the dark at room temperature until use. The amount of extractible polyphenols in *Aronia* pellets using water as solvent at ambient temperature was estimated by carrying out 4 successive extractions (1 g of *Aronia* pellets contacted to 40 mL of ultrapure water for 2 h at moderate agitation (100 min^{-1}), then the solid residue extracted 3 more times with 40 mL of ultrapure water in each extraction at same conditions). The obtained 4 extracts were centrifuged for 10 min at 10,000 rpm (Eppendorf Centrifuge 5804R, Hamburg, Germany) and stored at −20 °C in the dark until analysis.

2.3. Experimental methods

2.3.1. Integrated extraction–adsorption experiments

Integrated extraction–adsorption process consists in combining an extraction contactor and an adsorption column, liquid phase circulating between them in a closed loop. Experimental set-up is schematically presented in Fig. 1. Equipment and materials at laboratory and pilot scale were chosen in respect to a scale-up factor of 50 (Table 1).

Experiments at laboratory scale were carried out with the experimental set-up described elsewhere (Galván D'Alessandro et al., 2013). The 1 L glass extractor was equipped with an agitator and a temperature regulation system (circulation of water in an external jacket connected to a thermostat). Liquid phase was circulated by a peristaltic pump (Masterflex L/S, Cole Parmer Instrument Company, Barrington, USA). Experiments were carried out at room temperature (22 °C) with an agitation speed in the extractor of 40 min^{-1}, 4 g of raw source in the extractor, 4 g of Amberlite XAD7HP adsorbent packed in the adsorption column and 800 mL of liquid (initially ultrapure water). Time and flow rate were fixed to 2, 5 or 8 h and 1, 3 or 5 L h^{-1}, respectively.

Pilot scale experiments were carried out in a 100 L glass extractor equipped with an agitator and combined with an adsorption column (60 cm high and 4 cm internal diameter). Liquid phase was circulated by a peristaltic pump (Rumo 100, Heidolph,

Table 1
Equipment dimensions, materials quantities and operating conditions for experiments at laboratory and pilot scale.

	Laboratory scale		Pilot scale
	Experiment n°2	Experiment n°4	
Integrated process extraction–adsorption			
Glass extractor volume (L)	1	1	100
Solvent quantity (water) (L)	0.8	0.8	40
Aronia wastes quantity (g)	4	4	200
Column internal diameter (cm)	1.1	1.1	4
Fixed bed height (cm)	12	12	50
Adsorbent quantity (g)	4	4	200
Duration of the process (h)	8	8	8
Liquid phase flow rate (L h^{-1})	1	5	50
Superficial velocity in the column (cm s^{-1})	0.56	2.80	2.80
Number of liquid phase recyclings	10	50	10
Column elution			
Volume of eluent (L)	0.1	0.1	5
Eluent flow rate (mL min^{-1})	2	2	100