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Recovery of polyphenols from the by-products of plant food processing and application as valuable food ingredients

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ABSTRACT

Residues from plant food processing are valuable sources for the recovery of polyphenols, pectins, and proteins. These compounds may be used as natural antioxidants and functional food ingredients. The present review exemplifies innovative strategies for the valorization of by-products originating from apple, grape and sunflower processing. Apple pomace is an important starting material for pectin extraction. The color of apple pomace and of the pectins recovered therefrom is caused by oxidative browning of phenolic compounds. This limits the use of apple pectins as food gelling agents in very light-colored products. Consequently, a patented process for the simultaneous recovery of pectin and phenolic compounds from apple pomace has been developed. Phlorizin, the most abundant phenolic compound in apple pomace extracts, is the basic structure of a new class of oral antidiabetic drugs. Type 2 diabetes mellitus may be treated by the inhibition of sodium-glucose co-transporter-2 (SGLT 2). In a recently patented process, dihydrochalcones are enriched and purified from undesired *ortho*-dihydroxy phenol compounds being prone to oxidation and covalent binding to proteins.

While pigments from apple pomace are obtained by enzymatic oxidation of phlorizin using fungal polyphenoloxidases, anthocyanin-based pigments may be extracted from grape skins without using sulfite applying a novel enzyme-assisted process. Consequently, anthocyanins and phlorizin oxidation products are valuable alternatives for the replacement of synthetic azo dyes, some of which have been associated with health risks.

De-oiled sunflower press cake is a promising source of food protein as an alternative to soy and egg protein being devoid of toxic substances and low in antinutrients. Conventional alkaline protein extraction yields dark-colored products having reduced nutritional and functional quality. Therefore, a novel process for the production of light-colored sunflower protein isolates has been developed, combining mild-acidic protein extraction with subsequent adsorptive removal of phenolic compounds.

1. Introduction

In contrast to tropical and subtropical fruits, those from the temperate zone are characterized by a large edible portion, while the proportion of waste accruing from fruit processing is relatively low. Nevertheless, total waste arising from apple juice processing and winemaking accumulates to huge amounts, thus being a valuable source of waste valorization. Solely in Germany, 200,000–250,000 tons of wet apple pomace is available per year. According to literature reports, total mass of grape pomace resulting from European wine production was estimated to vary between 5 and 14.5 million tons (Meyer, Jepsen, & Sørensen, 1998; Schieber, Stintzing, & Carle, 2001b; Torres & Bobet, 2001; Torres et al., 2002). Similarly, sunflower expeller resulting from oil extraction is abundant, since sunflower is one of the four most important annual crops grown for edible oil production. Its annual world production amounted to 37.4 million tons in 2012 (The Statistics Division of the Food and Agriculture Organization of the United Nations; FAOSTAT). Sunflower meal obtained from the press cake is so far mostly used as animal feed. It contains up to 4% of polyphenols being considered as anti-nutritive in animal nutrition. In addition, due to rapid polyphenol oxidation and their concomitant interactions with proteins under alkaline condition, the conventional extraction of sunflower proteins from the press cake is still challenging (Weisz, Carle, & Kammerer, 2013; Weisz, Schneider, Schweiggert, Kammerer, & Carle, 2010).

Polyphenols are unevenly distributed in plant tissues, and processing may result in the separation of those parts of the fruit and grain being particularly rich in polyphenols. This holds true for apple, grape, cereals and oilseeds, where UV absorbing flavonoids and phenolic acids, attractant anthocyanins and deterrent tannins are mainly deposited in the outer layers of the skin, aleurone cells, seed coats and hulls, respectively. On the other hand, the apple core and the seeds of apple

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and grape are rich in astringent polyphenols. Consequently, apple and grape pomace as well as sunflower hulls containing the core, skin and pips, and pigmented seed hulls represent rich sources of polyphenols worth being exploited for the recovery of such components as natural food ingredients.

Polyphenols are characterized by high structural diversity, and may be subdivided into two major groups, the flavonoids and non-flavonoid compounds. Flavonoids share a common C6–C3–C6 carbon skeleton comprising flavanones, flavan-3-ols, flavan-3,4-diols, flavones, flavonols, dihydroflavonols as well as chalcones, dihydrochalcones, and aurones. The non-flavonoids are represented by phenolic acids, such as gallic acid, protocatechuic acid, and cinnamic acid derivatives, stilbenes and lignans. Polyphenols are present in plant tissues either in non-glycosylated form or as glycosides and/or associated with various organic acids and/or complex polymerized molecules with high molecular weight such as tannins.

Beginning with the first reports of the so-called “French paradox” (Renaud & De Lorgeril, 1992), interest in polyphenols has distinctly been boosted because of their putative health benefits (Schieber et al., 2001b). During the last two decades, polyphenols have been associated with a multitude of health beneficial effects possibly preventing damages and diseases caused by oxidative stress (Havsteen, 2002; Kammerer, Saleh, Carle, & Stanley, 2007).

Due to their widespread occurrence in fruits, vegetables, nuts, cereals, and oilseeds, phenolic compounds are an integral part of human nutrition. However, their dietary intake considerably varies depending on age, gender and nutritional habits (German National Nutrition Survey II, 2008). According to Scalbert and Williamson (2000) the daily per capita polyphenol intake amounts to an estimate of 1 g.

Since the consumers' interest in healthy food has significantly increased within the last few years, there is a steady trend toward the production of functional foods and functional food ingredients. Among other secondary plant metabolites, polyphenols are believed to contribute to the health protective effect of many food commodities. Therefore, they have even been named “vitamins of the 21st century” (Stich, 2000). Consequently, supplementation of foods with polyphenols recovered from waste materials accruing from fruit, vegetable, cereals and oilseed processing may be a valuable strategy to increase the dietary ingestion of such compounds.

For an economic recovery of polyphenols sufficient availability of raw material is mandatory. As earlier stated, in the case of pomace from apple juice extraction and winemaking such by-products are abundant. The same is true for sunflower press cake arising from oil extraction. Therefore, these by-products have been the focus of our research aiming at sustainable food production. In the following, the recent developments in the field of recovery and application of polyphenols from wastes accruing from food processing are exemplified by our attempts of apple pomace, grape pomace and de-oiled sunflower meal valorization.

2. Exploitation of apple pomace as a valuable source of polyphenols and pectin

The global apple (*Malus domesticus* Borkh.) production is steadily growing with an annual world production amounting to 76.1 million tons in 2011. Within the last 50 years (1961–2011), apple production area has almost been tripled reaching its maximum extension (6.3 million ha) in 1995; however, subsequently being reduced to 4.7 million ha corresponding to the latest records (FAOSTAT). According to recent estimates, 70%–75% of the apple is freshly consumed, while only about 25%–30% of the annual production is converted into value added products of which 65% are processed into juice, and the remaining quantity is sold as apple cider, purées, jams, as well as dried and ready-to-use apple products (Bhushan, Kalia, Sharma, Singh, & Ahuja, 2008; Joshi, Sandhu, Attri, & Walia, 1991; Shalini & Gupta, 2010).

Although most fruits from the temperate zone such as the apple are characterized by a high proportion (about 75%) of edible tissue, seeds, stems, and in some cases even apple peels accrue from apple processing into juices and other derived products. According to Bhushan et al. (2008), the left-over of juice processing, the so-called pomace, consists mainly of skin and flesh (95%), seeds (2%–4%) and stems (1%). Global apple juice production was estimated at 1.4 million tons in 2004–2005 with China steadily capturing the top position in world apple and apple juice production. China contributes around 600,000 tons of juice, and a further raise of Chinese production going along with the growing local consumption is to be expected. In Germany, about 700,000 tons of apples are annually processed into juice (VdF, 2013). Therefore, roughly 200,000 to 250,000 tons of apple pomace arise from German apple juice processing (Endress, 2000), while pomace generation from Japanese, Iranian, US-American, Spanish and New Zealand apple processing was reported to amount to 160, 97, 27 and 20 thousand tons for each of the latter countries, respectively (Bhushan et al., 2008; Gullón, Falqué, Alonso, & Parajó, 2007; Lu & Foo, 1998; Roberts, Gentry, & Bates, 2004; Takahashi & Mori, 2006). However, data concerning apple pomace availability are rather conflicting. According to other sources, taking into account the Brazilian, Chinese, and Indian production amounting to 0.8, 1 and 1 million tons of pomace (Shalini & Gupta, 2010; Vendruscolo, Albuquerque, Streit, Esposito, & Ninow, 2008; Wang, Wang, Fang, Wang, & Bu, 2010), the overall global availability of apple pomace may exceed 3,600,000 tons.

The numerous ways of apple pomace utilization have recently been reviewed by various authors (Bhushan et al., 2008; Kennedy et al., 1999; Shalini & Gupta, 2010). Hitherto, apple pomace has mostly been disposed and used as a ruminant feed. This is mainly due to the high water content (>70%) and thus, the high susceptibility of the wet waste material toward microbial degradation. Unless immediately dried, apple pomace suffers from brown discoloration due to retained polyphenol oxidase (PPO) activities, and microbial spoilage. Unlike pectin from industrial mango peels, rapid depolymerization and deesterification of apple pectins catalyzed by genuine and microbial pectinolytic enzymes is inevitable (Sirisakulwat, Sruamsiri, Carle, & Neidhart, 2010). Consequently, to enable pomace stabilization for its further valorization, fast dehydration of apple pomace is mandatory, thus requiring an industrial drying facility which is associated with high investments and energy demand.

As previously demonstrated by Schieber et al. (2003), harsh heat treatment in three-stage drum dryers used for the industrial production of apple pomace did not affect the stability of apple polyphenols being mainly retained in the pomace. Although requiring expensive drying, pectin extraction is still considered to be the most efficient way of apple pomace valorization (Endress, 2000; Fox, Asmussen, Fischer, & Endreß, 1991). To improve apple pectin color by removing the polyphenols from the product stream, an innovative process for the recovery of pectin and phenolic compounds has been developed (Schieber et al., 2003). The latter may serve as a natural counterpart to synthetic antioxidants and as a source of phlorizin (Bhushan et al., 2008; Kammerer, Boschet, Kammerer, & Carle, 2011; Kammerer, Kammerer, Jensen, & Carle, 2010). The de-pectinized residues from pectin extraction are still a source of fibers, and may be further used for food and non-food applications (Endress, 1991). Apart from pectin and low molecular polyphenols, pomace still contains a multitude of valuable compounds such as malic acid, saccharides (fructose, glucose, and sorbitol), cuticular waxes, and the seeds. The latter are rich in highly unsaturated fatty oil, carotenoids and tocopherols, and high molecular weight condensed polyphenolics, and yellow pigments (Fromm, Bayha, Carle, & Kammerer, 2012a, 2012b; Fromm, Loos, Bayha, Carle, & Kammerer, 2013).

In the following, the most recent developments considering the simultaneous recovery of pectin and polyphenols including yellow-colored pigments, seed oil and saccharides suitable for their use as natural sweeteners are summarized aiming at the complete exploitation of apple pomace.