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Research article

No time to waste organic waste: Nanosizing converts remains of food processing into refined materials

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ABSTRACT

Modern food processing results in considerable amounts of side-products, such as grape seeds, walnut shells, spent coffee grounds, and harvested tomato plants. These materials are still rich in valuable and biologically active substances and therefore of interest from the perspective of waste management and "up-cycling". In contrast to traditional, often time consuming and low-value uses, such as vermicomposting and anaerobic digestion, the complete conversion into nanosuspensions unlocks considerable potentials of and new applications for such already spent organic materials without the need of extraction and without producing any additional waste. In this study, nanosuspensions were produced using a sequence of milling and homogenization methods, including High Speed Stirring (HSS) and High Pressure Homogenization (HPH) which reduced the size of the particles to 200–400 nm. The resulting nanosuspensions demonstrated nematicidal and antimicrobial activity and their antioxidant activities exceeded the ones of the bulk materials. In the future, this simple nanosizing approach may fulfil several important objectives, such as reducing and turning readily available waste into new value and eventually closing a crucial cycle of agricultural products returning to their fields - with a resounding ecological impact in the fields of medicine, agriculture, cosmetics and fermentation. Moreover, up-cycling via nanosizing adds an economical promise of increased value to residue-free waste management.

1. Introduction

Modern food processing on an industrial scale inevitably results in considerable amounts of side-products, which often pose a significant ecological and economic burden. Millions of tons of seemingly "spent" materials, such as the seeds and skins of grapes in winemaking, the spent grain in breweries, the outer pericarp and shell of walnuts and the leftovers of brewed coffee, are produced each year in Germany alone (Beres et al., 2017; Brosowski et al., 2016; Fernandes et al., 2017; Kosseva, 2009; Mathias et al., 2017; Panusa et al., 2013). Some of these left-overs can be used in agriculture, for instance as animal feed (spent grain) or as cheap fertilizer at home (coffee) (Hardgrove and Livesley, 2016). Others are refined, for instance as grape seed oil or flour, or as walnut shell colourant. Yet many of these approaches result in modest applications and do not reflect the true value hidden in this apparent "waste".

From an ecological, environmental as well as economic perspective, this is rather unfortunate, as those materials are readily and cheaply available, initially food-grade and known to be rich in interesting substances, such as sparingly soluble polyphenols and tannins (Panusa et al., 2013). Previously, some of the applications of these waste materials have come forward, such as coffee shampoos for improved hair growth (Yesudian, 2012). Yet extraction of these compounds of interest is not straight-forward, as the waste materials at hand *per se* are still primarily waste - and not the kind of carefully harvested natural products designated for extraction, fractionation and isolation. Spent coffee or grain, for instance, readily starts to rot if not frozen or treated swiftly. At the

List of abbreviations: Spent coffee grounds, GC; Grape seeds, GS; High Pressure Homogenization, HPH; High Speed Stirring, HSS; Laser Diffraction, LD; Light Microscopy, LM; Photon Correlation Spectroscopy, PCS; Phosphate Buffer Saline, PBS; Standard Deviation, SD; Tomato Stem, TS; Walnut Shells, WS.

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same time, the presence of water complicates extractions with organic solvents, whilst prior drying consumes significant amounts of energy and offsets the potential benefits of harvesting from the spent instead of virgin material.

It is therefore hardly surprising that most of such leftovers are still more or less “disposed of” - often in the environment - rather than refined using traditional processes (Idris et al., 2004). Possible, more controlled alternatives, such as vermicomposting and anaerobic digestion are effective, yet also time consuming and costly, requiring at least four months to more than a year, respectively, and eventually may not reflect to true value of these left-over materials. (Manyuchi and Phiri, 2013; Zhang and Jahng, 2012).

Intriguingly, some of the drawbacks just mentioned, such as the risk of fouling and the presence of water, are not that dramatic when considered from the perspective of nanotechnology (Bystrzejewska-Piotrowska et al., 2009). Some of the waste may even be “in the perfect shape” for nanosizing. In the previous studies, high speed stirring (HSS) and high-pressure homogenization (HPH) was applied to equally “well shaped” crude plant materials, such as barks of *Pterocarpus erinaceus* Poir., fruits of *Solanum incanum* L. and even the entire dessert thumb *Cynomorium coccineum* L., to yield sterile, ready-to-use and biologically active nanosupensions in fairly good-quality - and with comparable ease (Griffin et al., 2016, 2017). Similar to the by-products mentioned, *Cynomorium coccineum* and *Solanum incanum*, itself a common weed, are readily available and amenable to nanosizing, resulting in - biologically active - particles in the diameter range of 300–600 nm and 2 μm, respectively. The objectives of this study have therefore been (i) to collect and convert common by-products into new and possibly valuable nanosized materials as part of a novel combined waste management and up-cycling strategy, (ii) to investigate the physical properties of the particles, (iii) to preliminarily evaluate their biological, *i.e.* antioxidant, antimicrobial and nematicidal activities for possible applications, and eventually (iv) to provide the foundation for future studies considering other forms of by-products and applications.

2. Materials and methods

2.1. Selection and acquisition of organic leftovers and waste materials

As part of this study, four common side-products of food production or consumption have been considered, which were selected explicitly as they are (a) widely, readily and cheaply available, often as part of food found in local stores, (b) constitute a major part of industrial or even household leftovers found across the Globe, (c) often end up in the environment, (d) promise to harbour at least some biologically active compounds and (e) appear to be amenable to processing by nanotechnological methods (Adi and Noor, 2009; Griffin et al., 2016; Idris et al., 2004; Vuong, 2017). These selection criteria have been chosen carefully to ensure both, a probable biological activity of materials based on prior knowledge, and large-scale availability, as small-scale by products, such as the odd used teabag, may not be economically feasible or ecologically relevant. The samples selected initially according to these criteria are depicted in Fig. 1 and include grape seeds (in form of commercially available grape seed flour available from a local health food store in Saarbruecken), the dried outer pericarp layer of walnuts (in form of commercially available powder sold as colourant from Holger Senger, Dransfeld, Germany, and referred to here as “walnut shell”, not to be confused with the harder inner pericarp which usually ends up in household waste), stems of harvested tomato plants (Vine tomatoes, self-cultivated by Claus Jacob (CJ) on his balcony) and spent coffee grounds (Lavazza

Fig. 1. The raw waste materials investigated as part of this study include (clockwise from top left): Coffee grounds after a normal brew of supermarket coffee, commercially available grape seed flour, walnut shell powder provided by a commercial supplier as colourant, harvested and dried stems of home-grown tomato plants. Photos provided by Sharoon Griffin.

Caffè Crema Classico, Lavazza, Turin, Italy, purchased from a local Aldi Süd supermarket in Sankt Ingbert, Germany) after brewing in a standard household filter coffee maker and a brisk quality check by Sharoon Griffin (SG) and CJ. As emphasized already, all of these materials are produced in larger quantities and available readily, usually at no extra costs apart from swift collection and transportation.

2.2. Production and subsequent characterization of nanosized materials

The product-specific waste was used as obtained (see above). Prior processing was only required in the case of harvested tomato stems, and involved air-drying, according to literature at room temperature (25–30 °C) in shade in a well-ventilated area for two weeks (Capecka et al., 2005). A coarse powder was obtained by grinding with mortar and pestle. Nanosizing procedures were performed as described by us previously with slight adjustments to account for the “waste character” of the material (Griffin et al., 2016). These techniques of nanosizing insoluble coarse materials are depicted in the flowchart of Fig. 2 and are currently used in research focussing on the development of cosmetics (Griffin et al., 2016; Keck and Muller, 2006; Scholz and Keck, 2015a, b).

Briefly, each sample was initially reduced in size by dry milling, apart from the spent coffee grounds, which were already “particulate”, moist and in many aspects therefore rather amenable to further processing. Bead milling was achieved using a FastPrep 24 Instrument (MP Biomedicals, Solon, OH, USA), with ceramic beads from Precellys Kits (Bertin Technologies, Montigny-le-Bretonneux, France). Subsequently, the samples were suspended in distilled water together with Plantacare® 2000 UP (Base, Ludwigshafen, Germany) as 1% w/w coarse suspension. Each sample was then subjected to HSS with a Polytron® PT2100 (Kinematica GmbH, Luzern, Switzerland) attached to a Polytron® PT-DA-2112/EC aggregate. Three 1 min cycles of HSS were performed at 11,000 rpm. Subsequently, HPH was employed as so-called “premilling stage” in an APV Gaulin LAB40 high pressure homogenizer (APV GmbH, Mainz, Germany) with three consecutive cycles at 200, 500 and 1000 bar pressure and a final homogenisation cycle at