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A greener approach towards isolating clove oil from buds of *Syzygium aromaticum* using microwave radiation

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ABSTRACT

The selection of suitable technique and apposite operating conditions are the key attributes for efficient extraction of phytochemicals from plant materials. Microwave assisted extraction (MAE) was incorporated to isolate essential oil from clove buds and the parameters were optimized using the grey based Taguchi method to maximize the yield of clove oil (Y, % w/w), eugenol content (y, % w/w) and bacterial inhibition. The results of single response analysis and multi-response analysis were compared. It was possible to achieve the best results (Y: 13.11%, w/w; y: 11.93%, w/w; zone of inhibition (ZOI): 24.80 mm) using the optimized conditions of multi-response study viz., amount of plant material − 30 g; volume of water − 200 mL; power − 600 W and extraction time − 30 min. The wastes generated during extraction were explored for their utilization as a by-product or raw material for other processes. Moreover, the performance of MAE was compared with that of hydrodistillation in terms of processing conditions, responses, energy and environmental impact. It was possible to reduce water usage, extraction time, energy requirement and CO_2 emission while achieving the improved quantity and quality of essential oil along-with a higher ZOI using the green MAE technique.

1. Introduction

The concepts of sustainability and green have created a positive scenario all over the world not only because of stricter government norms but also because of green consumerism. Green consumerism focuses on environmentally responsible consumption, i.e.; neutral or positive impact on environment (Chander and Muthukrishnan, 2015; Sachdeva et al., 2015), which has led to the emergence of green product and eco innovation. A product can be considered as a green product when it contributes towards zero or minimum carbon footprint, for examples organic product, herbal product, etc. Green products can have attributes like elimination or minimization of toxic substances, reduction in hazards, diminution in harmful side effects, environmentally benign nature, etc (Chander and Muthukrishnan, 2015; Joshi and Rahman, 2015; Maniatis, 2016; Sachdeva et al., 2015). In U.S., consumers expend about $ 25 billion per year for purchasing green products (Ferrel and Hartline, 2011). In eco innovation, environment sustainability norms are required to be utilized at various stages of product formation (Veleva and Ellenbecker, 2001). Hence, in the present situation, a green product should be produced by incorporating concept of sustainable development (Anastas and Warner, 2000; Anastas and Zimmerman, 2003; Manahan, 2006), principles of green chemistry (Anastas and Warner, 2000) and principles of green engineering (Anastas and Zimmerman, 2003).

Essential oil, mainly used in perfume, cosmetic, sanitary, medicine, flavoring and food industries (Bakkali et al., 2008), can be considered a green product because of its benevolent nature. Clove oil falls under the category of essential oil and is considered as “generally recognized as safe (GRAS)” (www.fda.gov). *Syzygium aromaticum* (Myrtaceae), commonly named as clove, is widely cultivated in Indonesia, India, Malaysia, Srilanka, West Indies, Madagascar and Tanzania (Kamatou et al., 2012; Srivastava et al., 2005). Clove oil can be extracted through leaves and buds, however, with varying composition (Srivastava et al., 2005). Eugenol, eugenyl acetate, β-caryophellene and α-humulene are the principal components of clove oil. Eugenol (4-allyl-2-methoxy phenol) is the prime constituent of the oil isolated from clove buds (Chaieb et al., 2007; Srivastava et al., 2005). The essential oil possesses larvicidal properties which can inhibit the growth of dengue responsible *Aedes aegypti* (Diptera: Culicidae) (Cortes-Rojas et al., 2014). It is possible to prevent vector borne disease using clove oil insect repellent (Shapiro, 2012). In recent literature, strong genotoxic effect (DNA damaging) was reported for eugenol which can be contributed in treatment of cancer (Slamenova et al., 2009). The clove oil, containing higher amount of eugenol, possesses a strong antimicrobial activity against multi-resistant *S. epidermidis*, Herpes simplex (HSV-I) and hepatitis C viruses (Chaieb et al., 2007). Bioactivity assessment has shown

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that both, the oil and eugenol, can inhibit animal and plant microorganisms, food poisoning bacteria, spoilage bacteria, and Extended-Spectrum-Beta-Lactamase (ESBL) enzymes of pathogenic bacteria (Dhara and Tripathi, 2013; Dorman and Deans, 2000). A wide spectrum of biological activities of clove oil leads to its extensive use in insect repellent, dental freshener, mouthwash, toothpaste and dental formulations (Srivastava et al., 2005). Dental materials having eugenol are commonly utilized in clinical dentistry (Markowitz et al., 1992). LDPE film coated with clove bud powder can be utilized in food packaging for safeguarding the food (Abdali and Ajji, 2015). Because of varied activities and applications, clove oil and eugenol have been included in various review articles focusing in these areas (Bakkali et al., 2008; Chaieb et al., 2007; Cortes-Rojas et al., 2014; Kamatou et al., 2012; Lubbe and Verpoorte, 2011; Prakash et al., 2015; Raut and Karuppayil, 2014).

Driven by the opportunities of the clove oil in numerous areas, it had been extracted by both conventional as well as novel techniques. A well established distillation method, either steam distillation or hydrodistillation, was used for extracting and characterizing clove oil from clove bud or leaf (Alma et al., 2007; Bhuiyan et al., 2010; Srivastava et al., 2005). In another study, a liquid extract after percolation through clove bud particles by dichloromethane was subjected to medium pressure liquid chromatography to isolate eugenol for preparing sulfonic derivative (Sudarma et al., 2013). These conventional techniques do not fall under the concepts of sustainable and green development because of various drawbacks like exhaustive solvent inventory, prolonged extraction time, labor intensiveness, severe energy consumption and environmental problems. As an environmentally benign method, supercritical carbon dioxide extraction (SFE) was appraised for different studies. Reverchon and Marrone (1997) isolated essential oil from clove buds using carbon dioxide as supercritical fluid and proposed a mathematical model. Results of clove oil extracted under supercritical conditions were compared with those of steam distillation, hydrodistillation and solvent extraction in terms of yield, eugenol content, extraction time, color and texture. SFE was found to be the optimum process for extracting good quality oil (Wenqiang et al., 2007). Another study was focused on optimization of process parameters of SFE of eugenol (Chatterjee and Bhattacharjee, 2013). This study also proposed an understanding of fixed bed clove bud matrix behavior and mass transfer coefficient. A systematic assessment of effect of bed geometry on extraction of clove oil using supercritical fluid was, also, performed (Zabot et al., 2014). Scopel et al. (2014) have studied phase equilibria, mathematical model and assessed antimicrobial activity of clove bud extract obtained using supercritical fluid. Though SFE is considered to be one of the best green technologies, it has constraints like elevated operating pressure, high capital as well as operating expenditure, safety concerns and lack of mechanical stirring leading to reduced extraction kinetics (Wei et al., 2016a, 2016b). Another novel technique which was employed in isolating the oil from clove buds was ultrasound assisted extraction (UAE). Alexandru et al. (2013) have compared the efficacy of batch- and flow-ultrasound reactors for extraction of clove buds. Total phenolic content and radical scavenging activity were selected as the responses. A flow reactor at higher flow rate (1350 mL/min) gave the best results. In another study employing UAE for extracting essential oil, optimization of process parameters was performed and antimicrobial activity was evaluated (Tekin et al., 2015). However, incorporation of organic solvent is required while utilizing ultrasound assisted extraction, thereby, separation and purification of the oil may become a laborious and energy intensive task. Further, UAE and SFE were also combined to improve the mass transfer characteristics. A comparison with conventional technique and SFE showed that the combined UAE-SFE was able to provide a better extraction yield (Wei et al., 2016a, 2016b).

Chemat et al. (2012) have defined green extraction and its principles. The principles are focused on renewable plant resources, water as a solvent, innovative technologies, co-products generation instead of waste, safe and robust process and improved quality of the product. In the recent trend of eco innovation, microwave assisted extraction (MAE) has provided numerous benefits mainly reduction in energy consumption, reduced volume of solvents, limited usage of toxic solvents and little waste products generation (Chemat et al., 2012; Desai et al., 2010; Leonelli and Mason, 2010). Further, this technique has a potential of commercial scale operation because of good rate of return and low break-even point (Filly et al., 2014; Leonelli and Mason, 2010; Petigny et al., 2014). These benefits make MAE a green and sustainable alternative to the established techniques. MAE has been used extensively in essential oil extraction from various plant matrices (Ferhat et al., 2006; Fidalgo et al., 2016; Filly et al., 2014; Petigny et al., 2014;). Authors have, also, effectively utilized this promising technique in isolating the volatile oil from various plant materials (Desai and Parikh, 2012; Desai and Parikh, 2015; Thakker et al., 2016). However, a wide spectrum of plant materials and varied behavior of different natural products have made it mandatory to conduct the parametric study or optimization study for the selected plant material and targeted compound. It was, also, found that the optimization focusing on more than one response was given a less preference in natural product extraction though the optimum conditions based on yield of the product, quality of the product and the application may offer faster acceptance of the process and find industrial relevance. And it is of great importance to evaluate the suitability of the results of multiple responses study since same numbers of experiments are used leading to the possibility of compromising either quality or quantity or the both of the responses.

The focal point of our previous study was the inclusion of multiple responses for arriving at a single set of operating conditions (Thakker et al., 2016) while the current work was carried out to optimally isolate essential oil from clove buds using MAE with a focus on developing a green extraction process. For achieving this goal, optimization of process parameters influencing yield of clove bud oil as well as eugenol and antimicrobial activity as responses was one of the objectives of this study. For better understating of the operation, one factor at a time study was executed, which provided the information regarding the effect of different parameters and formed the basis for the selection of their levels. Optimization of process parameters was performed using the grey based Taguchi method. Analysis of variance (ANOVA) was utilized in qualitative as well as quantitative manner to examine the impact of each parameter on responses. A comparison in terms of processing conditions, results, energy utilization and CO_2 release was made between single response study and multi-response study. Outcome of the study was compared with traditional hydrodistillation for the feasibility study with respect to energy consumption and CO_2 mitigation apart from the responses under consideration. Waste generated from the study was evaluated for their possible utilization in other applications. The results obtained from the present work would be beneficial for the industrial applicability.

2. Materials and methods

2.1. Materials

Dried clove buds were purchased from local market of Surat, Gujarat, India and stored at a room temperature in the moisture free environment. Clove buds were ground to reduce the size to 565 μm before conducting experiments. Eugenol (98% pure; Spectrochem Pvt. Ltd., Mumbai, India) and methanol (HPLC grade; Lombart Fine Chem. Ltd., Surat, India) were procured for analysis purpose.

2.2. Methods

2.2.1. Microwave assisted extraction

The Dean and Stark apparatus (1 L capacity) was kept in the modified microwave oven (ME83HD-B, 850 W (output power), 2450 MHz, Samsung, India) for extracting oil from clove buds. Rehydration of raw