Extraction of Essential Oils from Zingiberaceace Famili by using Solvent-free Microwave Extraction (SFME), Microwave-assisted Extraction (MAE) and Hydrodistillation (HD)

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ABSTRACT— ***Solvent-free microwave extraction (SFME) and microwave-assisted extraction (MAE) is a combination of dry and wet distillation with microwave heating. Both methods were compared with hydrodistillation (HD) for the extraction of essential oil from three types of rhizomes in Zingibereaceace family: Mango ginger (Curcuma amada), Java turmeric (Curcuma xanthorrhiza Roxb), and Black turmeric (Curcuma aeruginosa). Better results were obtained with SFME and MAE in terms of rapidity with 1 h extraction time using SFME and MAE vs. 4 h of extraction time using HD. SFME and MAE gives highest yield of essential oils at every second during the extraction process compared with HD.***

Keywords— SFME, MAE, HD

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1. INTRODUCTION

The Numerous techniques has been developed to produce high yield of essential oils, shorten extraction time, reduced solvent consumption, and increased pollution prevention. One of these techniques used microwave energy and it can be done by using two methods known as Solvent-free Microwave Extraction (SFME) and Microwave-assisted Extraction (MAE). These two methods use a microwave reactor where the samples were with or without added solvent or water. The internal heating of the in situ water within the material distends it and makes the glands and oleiferous receptacles burst. The vapor consisting of essential oils from the extraction will pass through a condenser outside the microwave cavity. The distillate is collected continuously in the collecting flask. SFME is based on the combination of microwave heating and dry distillation whereas MAE is based on the combination of microwave heating and wet distillation which used non-polar or polar solvent. Both methods were done under atmospheric pressure. The SFME and MAE apparatus is illustrated in Figure 1.

Essential oils is a complex mixture of volatile and non-volatile substances produced by different parts of the natural materials (e.g. vegetable, spice, fruit, etc). For volatile substance, it contains terpenes, sesquiterpenes and oxygenated derivatives whereas for non-volatile, it consists of fatty acid, waves, prosalens and carotenoids (David & Charles, 1999). The essential oil have various functions. It can be used for treatment of various diseases, flavour in food or as a preservative and also in cosmetic industries, especially for production of shower bath, lotion, shampoo and cologne (Lis-Balchin and Deans 1997).

Curcuma amada is commonly known as Mango ginger. It is the origin of Indo-Malayan and has been distributed widely from Asia to Africa and Australia. In food industry, Mango ginger has been used in the manufacture of pickles, culinary preparations and salads for flavour, candy and sauce (Kullu et al. 2013). In medicine industry, it can be used to treat a range of mood. It is also used in traditional medicine and Ayurvedic medicine. Mango ginger has many bioactive molecules where it demonstrates antibacterial, antifungal, anti-inflammatory, anti-hypercholesterolemic, insecticidal, antipyretic and antioxidant properties (Singh et al. 2010).

Curcuma xanthorrhiza Roxb is known as Java turmeric. It is a native Indonesian plant and grown in Thailand, Philippines, Sri Lanka and Malaysia. It has many applications and can be used for food and drink, medicine, textile and cosmetic industry. Curcuma xanthorrhiza has been reported to be useful to treat hepatitis, liver complaint, cancer, diabetes, rheumatism, hypertension and heart disorder. It has also shown anti-bacterial, anti-spasmodic, anti-inflammatory, anti-oxidant and antifungal effects (Devaraj et al. 2014).

Curcuma aeruginosa, is popularly known as Black turmeric in Malaysia, wild arrowroot or East Indian arrowroot in India and Waan-Ma-Haa-Mek in Thailand. It is found in Malaysia, Burma, Indonesia, and in South India (Ranjini and Vijayan 2005). In medicine, the rhizome of this plant is used to treat asthma, cough, scurvy, and mental derangements. It also helps women in confinement to accelerate the lochia and decrease pain and inflammation of the uterus. This rhizome can also be used for externally as poultice for inching (Reanmongkol et al. 2006). The compound identified from the essential oil of the rhizomes and leaves of Curcuma aeruginosa displayed antinociceptive and anti-inflammatory effects in experimental animals (Santos, Rao, and Others 2000).

To investigate the potentials of SFME and MAE, comparisons in terms of extraction time, yield and major composition.were made with hydro distillation (HD) for the extraction of essential oils from these three types of rhizome in Zingiberacea family: Mango ginger (Curcuma amada); Java turmeric (Curcuma xanthorrhiza Roxb); and Black turmeric (Curcuma aeruginosa) which are commonly used in the pharmaceutical and cosmetic industries..

2. METHOD

2.1 Moisture Content

The samples were cleaned with skins removed and ground before placing on the dish. Each dish contains about 10±0.001g of sample. The electric drying oven was set at different temperatures (30-90 ^{0}C) to find the optimum moisture content. The drying time was fixed at 24 h. After 24 h of drying, the sample was placed in a desiccator for about 5 to 10 minutes before weighing using an electronic balance (± 0.001g).

2.2 SFME and MAE Techniques

SFME and MAE extractions were performed in a Milestone ETHOS, which is a multimode microwave reactor. It can deliver maximum power up to 1000 W. The dimension of the PTFE-coated cavity is 35 x 35 x 35 cm. The parameters of the extraction process such as time, temperature, pressure and power were controlled using the 'EasyWAVE' 3.5 software program which is installed in the terminal controller-personal computer.

In SFME method, 500 g of fresh rhizomes sample (Curcuma amada, Curcuma xanthorrhiza, Curcuma aeruginosa) were cut into pieces before placing in the reactor without any added solvent or water, while for MAE method, 1000 ml of water was added and the mixture was blended. The essential oil was collected and stored at 0°C.

2.3 HD Method

Fresh rhizome samples (500 g) were mixed with 1000 ml of water and extracted for 4 hours (until no more essential oil was obtained) in an all-glass Clevenger apparatus. The setup of the apparatus is the same as SFME and MAE, but a heating mantle was as the heating source.

2.4 Yield

The total yield of essential oil was calculated immediately after collecting by using

$$EO(\%)=W\_EO/W\_fs \times 100 \quad (1)$$

where EO is essential oil, W_EO is weight of EO and W_fs is weight of fresh rhizome samples. The yield of essential oil droplet at every second during the extraction was calculated by using

$$EO⁄s=(EO\ (\%))/(T\_f\text{-}T\_d\ ) \quad (2)$$

where T_f is the duration of the extraction process (sec) and T_d is the time for the first droplet of essential oil (sec).

2.5 GC-MS Analysis

The essential oils were analysed by GC-MS (QP2010 Plus SHIMADZU). It was obtained using the following conditions: column oven temperature 40^{0}C, equilibration time 3 min, injection temperature 250^{0}C, injection mode split, the flow control mode is linear velocity 30.2 cm/sec, pressure 24.9 kPa, total flow 8.4ml/min, purge flow 6.0 ml/min and hold time 5.50 min; column: 30 m x 0.25 mm i.d., film thickness 0.25 µm. Identification of the components was achieved from the comparison of their mass spectral fragmentation patterns with those stored in the data bank (Wiley/NBS library).

3. RESULTS AND DISCUSSION

The moisture content (MC) of sample, the extraction time, yield (%) and yield per second (%) and major composition of essential oils from three samples of rhizomes are tabulated in Table 1. One of the advantages of the SFME and MAE is rapidity. The increase in extraction temperature is different from each technique due to the heating source. HD gives the highest temperature which is equal to the temperature of boiling water (100^{0}C) whereas for SFME and MAE, the temperature does not reach 100^{0}C. This is caused by the position of the temperature sensor which is located inside the microwave reactor not in the sample. Figure 2 represents the heating profiles during the extraction using SFME, MAE and HD of the three rhizomes' essential oils. To reach the time to obtain the first droplet of essential oil, it is necessary to heat only 10 – 20 minutes with SFME and MAE against 25 – 30 minutes of HD. There are slightly different for the times to obtain the first droplet as the reactor used for SFME and MAE is for a capacity of 4 litres whereas for HD is only 2 litres. The quantity of essential oil obtained by SFME and MAE after 1 h was sufficient for analytical determinations. The SFME and MAE methods also allowed a substantial saving of energy. HD required 4 h of extraction times (until no more essential oils was obtained) compared to SFME and MAE, which required only 1 h and the total yield of essential oil is not much different especially for mango ginger and java turmeric. For black turmeric, it showed the highest yield of essential oils using SFME and MAE compared with HD.

The yield of essential oils obtained by SFME were 0.05% for Mango ginger, 0.37% for Java turmeric and Black turmeric and those obtained by MAE were 0.23% for Mango ginger, 0.59% for Java turmeric and 0.33% for Black turmeric. For HD, the yield of essential was 0.26% for Mango ginger, 1.69% for Java turmeric and 0.13% for Black turmeric. Regarding the yield ratios between SFME and MAE (1 h) with HD (4 h), HD gives only 0.06% for Mango ginger, 0.42% for Java turmeric and 0.03% for Black turmeric, respectively, of the essential oil oil present.

The composition of essential oils isolated by SFME, MAE and HD shows a minor difference in terms of their oxygenated compound profiles. The Mango ginger essential oil isolated by SFME is dominated by beta-myrcene (15.22%) which is non-oxygenated compound. However, for both MAE and HD, it is dominated by caryophyllene oxide (an oxygenated compound), 23.43% and 12.55%, respectively. Vice versa from Mango ginger, the essential oil from Java turmeric isolated by SFME is dominated by oxygenated compound known as camphor, 23.34% and for MAE and HD, both essential oils are dominated by non-oxygenated compound known as cedr-8-ene, 47.72% and 24.1%, respectively. The Black turmeric essential oil is dominated by an oxygenated compound. By using SFME, it is dominated by boldenone (23.01%), whereas for MAE, it is dominated by trans-benzofuran, 6-ethenyl-4,5,6,7-tetrahydro-3,6-dimethyl-5-isopropenyl (35.82%). For HD, it is dominated by eucalyptol (29.13%), respectively.

Figure 3 represents the total oxygenated compound in three essential oils using SFME, MAE and HD. It can be seen that Mango ginger and Black turmeric essential oils are dominated by the oxygenated fraction which is more than 60% whereas for Java turmeric, it is dominated by the non-oxygenated fraction as the oxygenated fraction is less than 40%. The essential oil from Black turmeric gives the higher amount of oxygenated compound and lower amount of non-oxygenated compound by using SFME and for both Mango ginger and Java turmeric, HD produced higher amount of oxygenated compound and lower amount of non-oxygenated compound. That implies that the properties of the oils is dependent on the extraction method. (Okoh, Sadimenko, and Afolayan 2010). Substances with higher oxygenated compound are most valuable as it is highly odoriferous. Monoterpenes is the non-oxygenated compound and it is less valuable than oxygenated compounds as it only contributes to the fragrance of the essential oil to a minor extent.

There are different total number of compounds found in the essential oil extracted by SFME, MAE and HD. MAE has fewer compounds followed by SFME and HD. The presence and loss of some compounds in the essential oil is not an uncommon situation of the extraction. When the extraction time was reduced together with the amount of water used in the extraction, it will reduce the deterioration of the principal compounds by thermal and hydrolytic reaction and the generation of degradation products. Water can accelerates many reaction especially reaction via carbocation as intermediates as it is a polar solvent (Lucchesi et al. 2004).

4. CONCLUSION

The SFME and MAE methods are a combination of microwave heating and distillation (dry and wet). It provides more yield of essential oil, reduces the extraction time and allows a substantial saving of energy. Although it cannot provide valuable essential oil for every sample, it is still one of a good alternative for the extraction of essential oil as it only takes about 1 h of extraction process compared with HD which requires 4 h of extraction process to collect sufficient essential oil for analytical determination.

5. ACKNOWLEDGEMENT

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6. REFERENCES

List and number all bibliographical references in 10-point Times New Roman, single-spaced, at the end of your paper. For example, [1] is for a journal paper, [2] is for a book and [3] is for a conference (symposium) paper.

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