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Kinetics of change in colour and rosmarinic acid equivalents during convective drying of lemon balm (*Melissa officinalis* L.)

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ABSTRACT

The course of colour changes of lemon balm (*Melissa officinalis* L.) was examined during hot-air drying at temperatures ranging from 30 to 90 °C, constant specific humidity of 10 g kg^{-1} dry air and uniform air airflow of 0.2 m s^{-1}. CIELAB colour data was fitted to a first-order reaction kinetics model, in which the temperature dependence of the rate constant is modelled by the Arrhenius-type relationship. Total hydroxycinnamic acid derivatives were quantified by the photometric method and expressed as rosmarinic acid equivalent content. The a^* and b^* values were the most sensitive parameters to describe the colour changes of *M. officinalis* during drying. Redness was increased while yellowness gradually decreased as drying progressed towards the equilibrium moisture content. Colour degradation immediately started at the beginning of the drying process when moisture content was still high in the material. The rosmarinic acid equivalents were decreased with a progressive increase of temperature of drying air and it was assumed to be responsible for the browning of leaves due to enzymatic oxidation, especially at higher temperatures. Hue proved as best to express the optical impression of colour change. Low drying temperatures are recommended to avoid colour deterioration and to preserve high rosmarinic acid equivalent content.

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1. Introduction

The dried leaves of lemon balm (*Melissa officinalis* L.) are widely used either for culinary purposes or in the form of herbal tea infusion for their sedative and digestive properties. *M. officinalis* is also being included in pharmaceutical formulations due to the antioxidant potential of some of its constituents (Dastmalchi et al., 2008). Colour of medicinal and aromatic plants is considered as a primary quality criterion to the consumers, who prefer leaves with a natural appearance. Degradation of colour or browning can be indirectly related to quality deterioration due to enzymatic reactions caused by the activity of polyphenol oxidase (PPO) during postharvest applications. Enzymatic oxidation is a desirable and an essential processing step for the plant *Camellia sinensis* L. in order to produce black tea, however, browning reactions are undesirable in producing herbal tea. Although drying by convection is the most common preservation technique for medicinal and aromatic plants (Müller, 2007), the dried plant material often suffers from undesirable colour changes. During drying discolouration of leaves from bright green to pale green or to olive brown green mainly occurs due to degradation of chlorophyll pigments, which is sometimes accompanied by browning. Plant species belonging to Lamiaceae contain significant concentrations of phenolic compounds such as flavonoids and phenolic acids (Fecka and Turek, 2007), which can act as a substrate for PPO enzyme in the presence of molecular oxygen (Queiroz et al., 2008). In particular, the leaf of *M. officinalis* is rich in hydroxycinnamic acid derivatives mainly rosmarinic acid (Barros et al., 2013). More specifically, Bomme et al. (2008) reported that the content varied between 7.5 and 13.6% in a large collection of *M. officinalis* accessions. Drying intends to inactivate the enzymes

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present in fresh leaves by removing water from the product and reducing water activity. It also intends to preserve or even enhance the total phenolic content and the antioxidant activity of Lamiaceae herbs (Capeska et al., 2005). Yukizaki et al. (2008) found that the total polyphenols and rosmarinic acid content in water extracts of pre-dried *M. officinalis* were affected by the temperature of drying air. Consequently, due to the temperature sensitivity of the bioactive compounds found in plants of the Lamiaceae family, improper drying temperature may lead to irreversible colour alterations of dried leaves. Besides that the interaction among chemical substances and water vapour can induce changes in colour.

For the evaluation of colour, measurements based on the CIELAB system are commonly applied using lightness (L^*), redness (a^*) and yellowness (b^*) as parameters. According to Müller (1992) colour of *Salvia officinalis* L. was adequately preserved by drying at 50 °C, whereas an increase in temperature to 60 °C resulted in a distinct colour degradation of the dried herb. A work published by Rocha et al. (1993a) indicated that the L^* and a^* values were the most appropriate components to depict the impact of drying temperature on colour of *Ocimum basilicum* L. leaves. This deterioration of colour during drying of *O. basilicum* was attributed to chlorophyll degradation into pheophytin (Rocha et al., 1993b). Colour changes in terms of L^*, a^* and b^* values after convective drying of *Majorana hortensis* Moench. and *Rosmarinus officinalis* L. were documented in the literature (Singh et al., 1996). Apart from redness (a^*) and yellowness (b^*), chroma (C^*) and hue angle (h^*) can also be used to assess the colour quality of *Origanum majorana* L. (Pank et al., 1999). In a study on *Artemisia dracunculus* L., hue has been proven as the best parameter to depict the visual impression of browning on plant leaves effectively (Arabhosseini et al., 2011). Additionally, it was stated that a higher temperature or a longer drying process can cause significant colour changes in *A. dracunculus*. Furthermore, Martinov et al. (2007) found a positive correlation between the temperature of drying air and the degree of discolouration of *Hypericum perforatum* L. leaves in terms of hue value. The effect of conventional drying methods on colour characteristics of some herbs of Lamiaceae such as *Satureja thymbra* L. (Arslan and Özcan, 2012), *Mentha* × *piperita* L. (Arslan et al., 2010) and *R. officinalis* L. (Arslan and Özcan, 2008) has been investigated. Also, in a comparative study of six drying treatments, the extent of colour change between fresh and dried herbs of *Thymus daenensis* subsp. *daenensis* was assessed (Rahimmalek and Goli, 2013). However, none of the above studies on herbs of Lamiaceae investigated the course of colour changes during drying, except the work carried out for microwave drying of *O. basilicum* L. (Demirhan and Özbek, 2009). As far as *M. officinalis* is concerned, limited data on colour can be retrieved from the literature (Argyropoulos et al., 2009).

Therefore, the objectives of the current study were (i) to investigate the course of CIELAB colour changes during hot-air drying of *M. officinalis*, (ii) to model the behaviour of the most appropriate colour parameters at different temperatures within the range of 30 and 90 °C and (iii) to evaluate the effect of temperature of drying air on rosmarinic acid equivalents (RAE) of the leaves.

2. Materials and methods

2.1. Plant material

Herbs of lemon balm (*M. officinalis*) cultivar 'Citronella' were harvested in the middle of June (2010) before flowering from a two year old crop of an organic farm in Magstadt, 20 km west of Stuttgart (Germany). The harvest was always carried out in the morning after dew. The height of the collected plants was 70 cm in average by cutting them to a height of 10 cm above the ground. Since Ph. Eur. defines the leaf as the main source of the active ingredients, prior to drying experiments the leaves of the top 40 cm were picked manually from the stems. Herbal material was stored in the refrigerator at 12 °C for consecutive drying experiments.

2.2. Drying experiments

The hot-air drying experiments were carried out using the through-flow chamber of the high precision laboratory dryer (Argyropoulos et al., 2011a,b) designed at the Institute of Agricultural Engineering, Universität Hohenheim in Stuttgart (Germany) at air temperatures of 30, 45, 60, 75 and 90 °C and specific humidity of 10 g H_2O per kg of dry air, resulting in corresponding relative humidity of 36, 16, 7.7, 4 and 2.2%, respectively. A uniform air flow through the sample was maintained constant at 0.2 m s^{-1}. Loading density of fresh leaves was 2.2 kg m^{-2}, which develops an initial layer depth of about 30 mm (Argyropoulos and Müller, 2014). The leaves were dried to the equilibrium moisture content of each drying condition (Argyropoulos et al., 2012). During the drying process, samples were collected at regular intervals and analysed in terms of colour and moisture content. Drying was terminated when a constant mass was achieved for a certain time corresponding to the equilibrium moisture content of each drying condition. The experiments were repeated at least three times for each drying condition. The moisture content of leaves, expressed in wet basis (% w.b.) was determined by the oven method (103 ± 2 °C for 24 h).

2.3. Colour measurement

The colour of leaves was measured periodically during the drying processes by a reflectance Minolta colorimeter (model CR-400 Minolta Co, Ltd., Japan). The instrument was calibrated with a standard white plate at D65 illumination before taking measurements ($Y = 93.7$, $x = 0.3158$, $y = 0.3324$). The material was spread over a sampling plate and four measurements were performed at random locations by placing the colorimeter head directly above the sample. The colour parameters were expressed as L^* describing lightness ($L^* = 0$ for black, $L^* = 100$ for white), a^* describing intensity in green-red ($a^* < 0$ for green, $a^* > 0$ for red), b^* describing intensity in blue-yellow ($b^* < 0$ for blue, $b^* > 0$ for yellow). Moreover, chroma (C^*) and hue (h^*) were estimated by the a^* and b^* values. Chroma indicates colour saturation that is proportional to its intensity and is

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