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Modeling the kinetics of extracting oleoresin from dried turmeric (*Curcuma longa* L.) rhizome using acetone as solvent

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ABSTRACT

Kinetics of extracting/leaching (solid–liquid extraction) oleoresin from dried turmeric (*Curcuma longa* L.) powder was studied to determine the time taken to reach the equilibrium state. Acetone was used as solvent and the extraction was carried out at temperatures ranging from 35 °C to 50 °C using solid-to-solvent ratios between 1/5 and 1/30 (g/g). Equilibrium time was affected by temperature but not by the solid-to-solvent ratio. Results suggest that the increase in oleoresin yield was insignificant after 60 min of extraction time. A highest oleoresin yield of 6.49% (db) was obtained at 50 °C with solid-to-solvent ratio (1/30). A non-linear regression model was fitted to represent the kinetics of oleoresin yield. Effect of process variables (temperature and solvent-to-solid ratio) on model parameters, k (maximum possible extraction, percent dry basis) and a (initial extraction rate as a fraction of k – under specified conditions) were also studied. Equilibrium time is used to reduce lengthy extraction time.

1. Introduction

Turmeric oleoresin is the organic extract of turmeric (*Curcuma longa* L.) comprising mainly of curcuminoids and minor amounts of oils and resins. The curcuminoids represent the predominant coloring pigment of turmeric. The curcuminoids include curcumin, demethoxycurcumin and bisdemethoxycurcumin (Ammon and Wahl, 1991). Dried turmeric powder forms a key spice in Asian and Eastern cuisine as it imparts color and flavor to food and also used in cosmetics and Ayurvedic medicine (Tilak et al., 2004). As a natural bioactive component, curcuminoids have become very popular because they possess many health promoting effects. Some of the activities attributed to curcuminoids include: antioxidant (Jayaprakasha et al., 2006), anticancer (Goel et al., 2001), anti-viral and anti-infection (Mazumder et al., 1995).

Comparing supercritical carbon-dioxide extraction with the solvent extraction (Soxhlet), Braga et al. (2003) indicated that a higher curcuminoid yield was obtained by the latter method. Three phase partitioning (a technique used for protein separation) was evaluated for extraction of turmeric oleoresin by Kurmudle et al. (2011). The process consists of the simultaneous addition of t-butanol and ammonium sulfate to the aqueous slurry of turmeric powder. But, according to Directive 2008/128/EC and JECFA (2006), t-butanol is not listed in the solvents to be used in the extraction of turmeric oleoresin (EFSA Journal, 2010). Selection of the process to obtain turmeric oleoresin is dependent on the intended usage of the turmeric extracts (Braga et al., 2003). Solvent extraction is the most common technique for oleoresin extraction because of its simplicity and cost-effectiveness. Commercially oleoresin is obtained from dried turmeric rhizome (plant source) by the leaching method (solid–liquid extraction) using mainly acetone as solvent (FDA, 2013). Leaching is the preferential separation of one or more constituents of a solid mixture by contact with a liquid solvent; leaching and solvent extraction are sometimes synonymously used. The process of solvent extraction/leaching of component(s) involves, the contact of solvent with the solid feed containing the components, penetration of solvent into the cellular matrix of the solids, dissolution of the components into the solvent, diffusion of components out of the solid and subsequent diffusion into the bulk. This transfer operation continues till a state is reached when the component concentration of the miscella (solution of components and solvent) increases no further thereby attaining a constant value and, correspondingly, the residual concentration of components in the marc (leached solid) decreases no further; at this stage, equilibrium is said to be reached between the concentration of component in the marc and that in the miscella (Treybal, 1981); the correlation between the respective concentration of component in marc and in the miscella is known as equilibrium relationship. In a batch process (namely, Soxhlet extractor), the time to reach the state of equilibrium is known as equilibrium time – which is studied through the kinetics of

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extraction. Equilibrium relationship and extraction kinetics constitute basic information input while designing an extractor.

Several factors can affect the efficiency of the leaching process such as type of solvent used, extraction temperature, solvent-to-solid ratio and the number of extraction stages. Different solvents like hexane, alcohol, acetone, ethylene dichloride etc. are used in the extraction of oleoresins from spices (Revathy et al., 2011). Joint FAO/WHO Expert Committee on Food Additives, (JECFA, 2006) and Directive 2008/128/EC have permitted the use of six food grade solvents for the extraction of curcuminoids from turmeric: acetone, ethanol, methanol, isopropanol, hexane and ethyl acetate (EFSA Journal, 2010). They also specified limits for residual acetone (Table 1). Surveys indicate that the acetone extract yielded maximum curcuminoid (Govindarjan, 1980; Verma and Jain, 2011). The many advantages of using acetone as a solvent include: maximum reproducible extraction, avoids problems with pectins, and permits a much lower temperature for sample concentration (Viguera et al., 1998).

Modeling of the extraction kinetics of the solid–liquid extraction of oleoresin from turmeric is scarce in the literature. The aim of this work is to study the kinetics of extracting oleoresin yield at varying extraction time at various operating temperatures and solid-to-solvent mass ratios in a modified Soxhlet apparatus (batch process). A regression model was then fitted to experimental data of oleoresin yield at various extraction times, whence the equilibrium time was determined. Also, in order to ensure that acetone was within safety limits in the oleoresin extract, quantification of residual acetone in oleoresin was done using GC/MS.

2. Materials and method

2.1. Raw materials

Dried turmeric fingers of Alleypey variety were procured from Synthite Industries Ltd., Kolenchery, Kerala. The turmeric fingers were triturated by manual grinding and were sieved through a sieve shaker with sieves of Tyler series (20–200 mesh). Particles collected between the sieves 60 and 70 (average size: 231 μm) were used for extraction. This fraction constituted 80% of the ground mass of the raw material. The moisture content was determined by keeping the sample in hot air oven (Relco-DTC96SI, Kolkata, West Bengal) at 105 °C until a constant weight is reached (gravimetric method). The ground powder in all cases had an initial moisture content of 14.48% (dry mass basis).

2.2. Solvents

The solvent used for extraction was acetone of analytical grade (E-Merck, Mumbai). HPLC grade acetonitrile was used for GC/MS analysis.

2.3. Batch extraction of turmeric oleoresin (solid–liquid leaching/extraction)

The extraction was carried out in a modified Soxhlet apparatus called the Soxtherm (Gerhardt SE-TR, Germany) in which thimbles were not used. Extraction temperatures ranged from 35 °C to 50 °C, extraction time from 10 min to 180 min and solid-to-solvent ratio in g/g from 1/5 to 1/30. Five grams of turmeric powder was taken in each vessel with the addition of a measured amount of solvent and placed in the extractor and maintained at the specific extraction temperature. At specific time intervals, the vessels were withdrawn from the soxhlet extractor, centrifuged ((REMI/R, Kolkata-24, West Bengal) at 3000 RPM to separate the solid from the miscella and the solvent was recovered from the miscella. The extract was kept in a vacuum oven (SPAC-N-SERVICE, Kolkata-42, West Bengal) to remove traces of solvent and stored in desiccators under vacuum until further analysis. The oleoresin yield obtained was reported in grams per gram of dried turmeric powder (dry mass basis) and multiplied by 100. All experiments were done in triplicates. Precautionary measures were taken to vent out the leaked vapors of acetone if any, to the atmosphere.

Table 1
Specified limits (ppm) of residual solvents in JECFA and Commision Directive 2008/128/EC.

Solvent	JECFA (2006)	Commision Directive 2008/128/EC
Acetone	30 ppm	50 ppm

2.4. Mathematical model

A first order kinetic model formerly used for the assessment of extraction progress of bioactive compounds from plant sources (Karacabey et al., 2013; Dandekar and Gaikar, 2002) was employed to describe the extraction kinetics of oleoresin from turmeric powder. The experimental data were fitted to the following non-linear regression model that establishes a relationship between oleoresin yield and extraction time:

$$Y = k(1 - \exp^{-at}) \quad (1)$$

where Y = oleoresin yield as percentage of the mass of dry solid, t = extraction time (min), k = the maximum possible extraction achieved (as the time duration for extraction tend to infinity) under specified conditions, a = the initial (at time instant, $t = 0$) extraction rate as fraction of k – under specified conditions.

Iterative estimation of values of parameters (k and a) was done by Minitab 16 software. The figures were generated using Origin Proversion 9.

2.5. Data analysis and model evaluation

The effect of temperature and solid-to-solvent ratio on oleoresin yield at varying extraction time (duration) was determined by analysis of variance. In order to determine the difference among means, Duncan's multiple range test at 0.05% level of significance was conducted and letter groupings were generated. The SPSS statistical program version 15.0 was used for the analyses. Correlation coefficient (R^2) and the root mean square error (RMSE) were calculated so as to evaluate the goodness of fit of the non-linear regression model to the experimental data. Lower the RMSE values and higher the R^2 values, the better are the goodness of fit. Variability of the model parameters (k and a) with temperature and solvent-to-solid ratio were determined individually by developing a quadratic response surface model.

2.6. GC/MS quantification of residual acetone

The TRACE 1300 GC equipped with TSQ 8000 Triple Quadrupole Mass Spectrometer (Thermo Fisher Scientific, USA) was used to quantify residual acetone in the oleoresin samples. The analytical capillary column used was TR-FAME (70% cyanopropyl equiv polysilphenylene siloxane) of dimensions 30 m × 0.25 mm × 0.25 μm. The carrier gas was helium (constant flow, 1.5 ml/min), 1 μl of sample was injected and the split ratio was 1:40. The injector temperature was set at 200 °C. The GC ramp rate was set as follows: 50 °C for 8 min followed by increase in temperature at the rate of 25 °C/min to 250 °C and maintained for 2 min. The mass spectrometer was set in SCAN and SIM mode scanning the mass range from m/z 40–100 and m/z 43, 58, respectively. Acetonitrile was used as