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Extraction of polydatin and resveratrol from *Polygonum cuspidatum* root: Kinetics and modeling

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ABSTRACT

The biochemical activities of polydatin and resveratrol have been proven to own considerable medicinal potential. However, the extraction processes are yet to be fully characterized. This study explores particularly the optimization and kinetics of polydatin and resveratrol extraction from Rhizoma Et Radix Polygoni Cuspidati (the root of *Polygonum cuspidatum*) in agitation extractions by ethanol and water mixtures. The effects of the parameters on several extraction processes have been fully investigated. The optimized condition for polydatin was found at 0.4 mole fraction of ethanol as the extraction solvents, temperature 323 K, agitation speed 500 rpm and solid to solvent ratio of 1:90. However, the optimized condition for resveratrol was 0.4 mole fraction of ethanol, temperature 323 K, agitation speed 800 rpm, solid to solvent ratio of 1:120. Furthermore, the extraction kinetics behavior of polydatin and resveratrol follows a first order kinetics with diffusion coefficient ranging from 3.59×10^{-11} to 7.53×10^{-11} $m^2 s^{-1}$ and 3.90×10^{-11} to 10.10×10^{-11} $m^2 s^{-1}$ for polydatin and resveratrol respectively. The values of activation energy for the extraction were found to be 3.96 kJ mol^{-1} for polydatin and 17.83 kJ mol^{-1} for resveratrol.

Keywords: Polydatin; Resveratrol; *Polygonum cuspidatum*; Batch extraction; Agitation; Kinetic model

1. Introduction

Rhizoma Et Radix Polygoni Cuspidati (the root of *Polygonum cuspidatum*) is a traditional herbal medicine belonging to the Polygonaceae family and widely grows in China and other Asian countries (Chen et al., 2012). Since stone age, people explores the medical usefulness of the root of *Polygonum cuspidatum* (PC) on amenorrhea, hepatitis, arthralgia, chronic bronchitis, jaundice, hypertension, and hyper-cholesterolemia (Park et al., 2004; Grimsby and Kesseli, 2010; Bralley et al., 2008). Modern chemistry shows that polydatin (POLY) and resveratrol (RES) are the main bioactive constituents in PC (Gu et al., 2006). As an important stilbenoid (Xing et al., 2009), POLY has neuroprotective effect on cerebral injury caused by ischemia, promotes losing weight, provoking appetite in low-income mothers with young kids, and enhancing heart function and microcirculation (Zhao et al., 2003; Jordan et al., 2008; Cheng et al., 2006). Meanwhile RES, a common type of polyphenol, exists extensively in food and plants. RES has been identified to exert a broad spectrum of biochemical and physiological activities, including estrogenic effects, antiplatelet and anti-inflammatory properties,

Abbreviations: PC, *Polygonum cuspidatum*; POLY, polydatin; RES, resveratrol; $C_{l,exp,i}$, experimental concentration of the component in the liquid phase at time t; $C_{l,pre,i}$, predicted concentration of the component in the liquid phase at time *t*; *N*, number of observations; PE(%), relative percent error; *R*, the universal gas constant (kJ mol^{-1} K^{-1}); RMSE, root mean square error; D, diffusion coefficient; *r*, the radius of the particle; V_s, volume of solids in the system; V_l, volume of the liquid phase; γ, partition coefficient; C_l, concentration of the component in the liquid phase at time *t*; $C_{l\infty}$, concentration in the liquid phase at equilibrium; k_{obs}, observed first order rate constant; *t*, extraction time.

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antioxidant capacity, cardioprotection and anticancer activity (Karacabey and Mazza, 2008; Lanzilli et al., 2012; Zhang et al., 2009). Interestingly, a recent study even reported that RES may improve health and extend maximum lifespan of diverse species (Baur et al., 2006).

Solvent, temperature and mechanical action (such as pressure and shaking) are the important factors when choosing extraction method (Leal et al., 2011). Organic solvent extraction is one of the most classical techniques to obtain herbal extracts. There are several organic solvent extraction methods such as agitation, centrifugation, percolation, Soxhlet extraction, etc. (Wang and Weller, 2006). Several extraction experiments on POLY and RES from PC root have been carried out, like ultra high pressure extraction (Yu et al., 2006; Liao et al., 2010). However, the kinetics and modeling data of POLY and RES by the agitation extraction are yet to be investigated.

The kinetics of solute extraction from natural sources relates to releasing solute from porous matrices into a solvent phase by means of mass transfer (Campos et al., 2005). Analysis of the experimental kinetic data curves was carried out by a steady state model equation derived by Spiro and Siddique (Stapley, 2002). This research is set to demonstrate the kinetic model for this extraction process in order to predict extraction rate constant, initial extraction rate and the equilibrium concentration. Meanwhile, understanding mass transfer at the solid–liquid interface plays a key role in scaling-up of the process at industrial level, unlike the micro productions at laboratory scale (Goto et al., 1998). In attempt to address these issues, this research focus on the influence of different extraction process parameters such as solvent, solid to solvent ratio, the speed of agitation, temperature on extraction yields. The optimized conditions acquired from the research could be further applied in the designation of commercial productions, and more significantly, the solvent used in our study (ethanol and water) is environmental friendly and biological sustainable.

2. Materials and methods

PC root was supplied by Tongrentang Ltd., branch of Xi'an. The PC powder size used was 0.85–2.00 mm which is brown yellow in color, moisture content NMT 7.5%, bulk density 0.4072 g/ml. Methanol and water were of HPLC grade. Methanol was purchased from Kermel Chemical Reagent Ltd., Tianjin, China. Ethanol was purchased from Tianli Chemical Reagent Ltd., Tianjin, China and the redistilled deionized water was offered by the lab. Reference standards polydatin and resveratrol were obtained from the National Institute for Control of Pharmaceutical and Biological Products, China.

The extraction solvents prepared were composed of ethanol and water and the mole fractions of ethanol in the extraction solvents were 0.2, 0.4, 0.6 and 0.8, which means that the volume concentrations of ethanol were about 44.7%, 68.3%, 82.9% and 92.8%, respectively.

2.1. *Batch extraction*

Batch extraction was conducted in a 3-neck flask (sealed) of 250 ml capacity equipped with one bladed (pitched blade) plastic muddler for agitation. Definite amount of the PC root powder was put in the flask and 150 ml ethanol and water mixtures at different concentrations as the extraction solvents were added to it. The mixtures were agitated for 120 min. 0.5 ml samples were withdrawn at 5, 10, 15, 30, 45, 60, 90 and 120 min respectively and centrifuged (12,000 rpm, 5 min) to get supernatant and then diluted for HPLC analysis. It is noteworthy that an equal amount of solvents must be added to the mixture after taking out the sample. A series of parameters impacting the extraction process (such as solvent, solid to solvent ratio, agitation speed and extraction temperature) were optimized and the final extraction at the optimized conditions was investigated to get the maximum yield. All the experiments were carried out in three times and the average data has been presented in the paper. In addition, the uncertainty of the experimental values is less than 2%.

2.2. *Analytical method*

Analysis of POLY and RES was carried out by an HPLC system (SHIMADZU LC-2010A; Japan) with an Xterra C_{18} column (150 mm × 4.6 mm). A mixture of water and methanol was used for gradient elution, i.e. methanol from 40% to 80% for the first 5 min, methanol from 80% to 100% between 5 and 7 min, methanol 100% between 7 and 12 min, and 40% for the last 3 min of the elution. The flow rate was set at 1.0 ml/min, and the UV detector was set at the wavelength of 303 nm.

3. Analysis of kinetic data

According to the current study, Spiro and Siddique have developed the kinetic expression which predicts first order behavior and analyzes the concentration data to apply to the infusion of tea and coffee (Stapley, 2002). Taking the analogy of the process of solid–liquid extraction and infusion, the attempt has been made to use the Spiro and Siddique model which fitted an equation of the form:

$$\ln\left(\frac{C_{l\infty}}{C_{l\infty}-C_l}\right)=k_{\text{obs}}t \qquad (1)$$

where C_l stands for the concentration of the component in the liquid phase at time t, $C_{l\infty}$ represents its concentration in the liquid phase at equilibrium, k_{obs} is an observed first order rate constant and t is the extraction time.

The basis for derivation of the above kinetic model lies in the fact that, the rate of mass transfer is proportional to the difference between the average concentrations in the solid and in the liquid phase (corrected by a partition coefficient). This kinetic study is analyzed by a lumped parameter model (Spiro and Jago, 1982). Spiro and Siddique model used to represent the extraction process is equilibrium-dependent solid-liquid extraction model and it takes into consideration the diffusion-dependent solid–liquid extraction process at the same time. Diffusion coefficient of the solute from the solid to liquid can be determined by the observed rate constant. Assuming spherical geometry of the particle of the PC powder, gives:

$$k_{\text{obs}}=\frac{12D}{r^2}\left(1+\frac{V_s}{\gamma V_l}\right) \qquad (2)$$

where D represents the diffusion coefficient (mean value in the extraction period), r is the radius of the particle, V_s stands for the volume of solids in the system, V_l is the volume of the liquid phase and γ is the partition coefficient, defined as:

$$\gamma=\left(\frac{C_l}{C}\right)\quad \text{equilibrium} \qquad (3)$$