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Introducing the concept of sono-chemical potential: A phenomenological model for ultrasound assisted extraction



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ABSTRACT

The current study presents a mathematical formulation that describes the role of ultrasonic irradiation in ultrasonic assisted extraction with thermodynamic terms. This model described the influence of ultrasound irradiation on the chemical potential of extracted compounds and predicted the increase of the yield of extraction using ultrasound irradiation. The term sono-chemical potential was described for the first time in an analogy to the piezo- or the electrochemical potential. Subsequently, the derived formula was applied on the extraction of polyphenols from spearmint, where the model showed the increase factor in the extracted amounts during ultrasound assisted extraction; the predicted value for a five minute extraction was 2.89 and the experimental values were 2.99, 2.92 and 2.32 for the hydroxycinnamic acid derivatives content, the antioxidant activity and the total phenolics respectively. The model could be further verified and used to explain the enhanced recovery of various compounds from diverse matrixes.

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1. Introduction

Natural antioxidants have attracted attention due to their health-promoting properties (Navarrete et al., 2011). The traditional extraction technique for polyphenols from plant materials or food industry wastes is solid-liquid extraction (maceration) (Azmir et al., 2013; Virot et al., 2010), but it has been linked with various disadvantages, such as (i) time-consuming extraction procedures, (ii) the thermal degradation of phenolic compounds, (iii) the use of less eco-friendly organic solvents, and (iv) low yield of extraction (Grigonis et al., 2005). In the last two decades, innovative extraction techniques have been proposed that use added energy such as ultrasound and microwave irradiation in order to tackle the aforementioned drawbacks. The application of added energy usually achieves higher yield of polyphenols in shorter extraction time in comparison with solid-liquid extraction. Characteristically, the recovery of phenolic compounds from marjoram using ultrasound assisted extraction was 96% higher than solid-liquid extraction (Hossain et al., 2012). Ahmad-Qasem and co-workers (2012) also reported that ultrasound irradiation reduced the extraction time from 24 h to 15 min without changes in polyphenolic composition of olive leaf extracts.

Ultrasound-assisted extraction has been used successfully to recover polyphenols and other bioactive compounds from various plants (Adjé et al., 2010; Jerman Klen and Mozetič Vodopivec, 2011; Rodríguez-Rojo et al.2012). As a possible mechanism for ultrasound assisted extraction was proposed the ultrasonic irradiation enhancement involves the shear force created by implosion of cavitation bubbles upon the propagation of the acoustic waves in the kHz range. Collapse of bubbles can produce physical, chemical and mechanical effects which result in the disruption of biological membranes to facilitate the release of extractable compounds and enhance penetration of solvent into cellular materials and improve mass transfer (Dai and Mumper, 2010).

It is well-known that the extraction is affected by the diffusion coefficient and the dissolution rate of compounds until they reach the equilibrium concentration inside the solvent. The ability of each polyphenol to be solubilized, transferred or diffused into a given solvent is governed by thermodynamics (Galanakis et al., 2013). One of the primary thermodynamic factors describing the extraction procedure is the chemical potential. The effect of ultrasound irradiation on chemical potential has not been described mathematically yet. Previous attempts on mathematical modeling of ultrasound assisted extraction have mainly focused on kinetic parameters of the extraction (Pan et al., 2012; Yue et al., 2012), the fitting of kinetic models by linear regression (Cheung et al., 2012) or second-order polynomial equations (Prakash Maran et al., 2013) with terms that lack physical meaning. A phenomenological kinetic model also demonstrated that ultrasound-assisted extraction is a two-step process with ultrasound irradiation influencing only the first step (Milić et al., 2013).

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Nomenclature molar volume of component i (cm³ mol⁻¹) Е sound energy density (J cm⁻³/N m⁻²) V_{i} G Gibbs free energy (I mol⁻¹) activity of component i flux of component j (mol cm⁻² s⁻¹) sonochemical potential of component i ($I \text{ mol}^{-1}$) Ji number of moles of component i activity coefficient of component i n_{i} γ_i pressure (N m⁻²) Р chemical potential (I mol⁻¹) μ R universal gas constant (8.312 | K⁻¹ mol⁻¹) mole fraction of component i χ_{i} S entropy ($J K^{-1}$) T temperature (K) volume (cm³)

The main objective of the present work was to develop a mathematical model to describe the ultrasound assisted extraction of compounds from matrix as food- to a solvent- phase in terms of thermodynamics. In this attempt, the concept of sono-chemical potential is introduced for the first time; an analogous term is the electrochemical potential, which describes the enhancement of the mass transfer due to the application of pulsed electric fields (Toepfl et al., 2007). Furthermore, the developed phenomenological model was applied to study the ultrasound assisted extraction-of phenolic compounds from an aromatic plant (*Mentha viridis*) compared to the conventional solid-liquid extraction.

2. Model approach

2.1. Theory

The various kinds of mathematical modeling in food engineering have been classified and overviewed by Gekas (1992). Among them of a special interest are the so-called phenomenological or thermodynamic models. The development of this category of models was based on the efforts and the work of the pioneers of the Irreversible Thermodynamics (Haase, 1993; Onsager, 1931; Prigogine, 1960) Starting either from the Entropy (S) ensemble or from the Gibbs free energy (G) ensemble, the obtained thermodynamic identities (Schroeder, 1999) could lead to the development of two main lines of thermodynamic modeling, which are considered below:

- (a) The equations of Irreversible Thermodynamics (Onsager approach, Stefan-Maxwell approach) ideally suited for the modeling of systems where more than one driving forces act; an example is given in the modeling of the Reverse Osmosis unit operation by Kedem and Katchalsky (1958).
- (b) The modeling which considers the chemical potential (μ) as the universal driving force, after its extension to cover the effect from other phenomena than diffusion, for example hydrostatic pressure or electrical or other energy fields (Strathmann, 2000, 2004).

In both lines of modelling, the coupling phenomena in multidriven systems are explicitly taken into account. Furthermore, food and biological systems are multi-phased and multi-component so that additional coupling phenomena occur between the fluxes and the chemical potentials of the components involved (Haase, 1993). In this work the following simplified scheme of our system is considered (Fig. 1a and b). In these simplified diagrams, ultrasound energy is supplied to the solvent molecules, thereby increasing their susceptibility to enhance the chemical potential driven transport of the component 1 from phase I to phase II. In the spirit of the thermodynamic approach of the extended chemical potential, the concept of the sono-chemical potential has been

coined by the authors, in an analogy to the piezo- or the electro-chemical potential previously defined by Strathmann (2004).

2.2. Formulation

Following the Gibbs free energy ensemble approach, to the thermodynamic parameters $\{T, P, n_i\}$ where T: temperature, P: pressure and n_i : the number of moles of a component i, the sound energy density E is added in the presence of sound fields (Fig. 1b).

Then the thermodynamic identity for *G* is the following: Case Fig. 1a:

$$dG = -SdT + VdP + n_i d\mu,$$

(1)

leading to the following thermodynamic identity of the chemical potential, being a first generation property for a given component i (Pérez and Romulus, 1993) under isothermal conditions and without hydrostatic pressure terms between two liquid phases

$$d\mu_{i} = RTd \ln a_{i} \tag{2}$$

and

$$d\mu_{i} = RTd \ln a_{i} + V_{i}dP \tag{3}$$

Case Fig. 1b:

$$dG = -SdT + VdP + n_i d\mu_i + VdE (4)$$

In this case the external field is the sound energy supplied to the solvent and neglecting hydrostatic pressure effects and assuming isothermal conditions, the thermodynamic identity of the sonochemical potential, β_i , of the component i is the following:

$$d\beta_i = RTd \ln a_i + V_i dE \tag{5}$$

 β_i is the, so defined by us, sono-chemical potential and E the sound energy density in pressure units (J/cm³ or N/m²). It can be seen from the unit agreement that the sound energy is only effective in the liquid solvent and not the plant matrix. We consider the driving force for the mass transfer of phenol from the plant matrix (phase I) to the solvent (phase II) during conventional extraction (Fig. 1a) as the difference in chemical potential, which can be formed as follows:

$$\mu_i^{II} = \mu_i^0 + RT \ln a_i^{II} \tag{6}$$

and

$$\mu_i^I = \mu_i^0 + RT \ln a_i^I \tag{7}$$

It is also valid that $\alpha_i = \gamma_i x_i$.

The activity coefficient as well as the activity of ethanol in phase II is equal to 1 (pure ethanol), whereas in phase I, we accept the convention by Wesselingh and Krishna (2006) that the natural logarithm of a component not present in the food matrix equals to 2 therefore:

$$\mu_i^{II} = \mu_i^0 + RT \ln 1 \tag{8}$$

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