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Mechanism of Microwave Effect on the Extraction Process of Tea Polyphenols

Dan Li, Tao He, Boyu Li, Ziqin Wang, and Zhengming Tang*

School of Electronic and Information Engineering, China West Normal University, Nanchong 637009, China

ABSTRACT: Microwave-assisted extraction (MAE) is an effective method for extracting tea polyphenols. However, the research on MAE mainly focuses on experimental methods, which not only leads to a large amount of experimental work but also generates a lot of material waste. In addition, due to the lack of mechanism research, it is difficult to find a more effective method. In this study, based on electromagnetic field theory, the heat and mass transfer model of tea polyphenol extraction is established based on measuring the dielectric properties of the extract. The distribution of temperature, diffusion coefficient, and flow rate of microwave-assisted extraction of tea polyphenols are all analyzed in detail. The results show that the temperature distribution in the extraction system is uneven. The middle temperature of the extraction solution is high and the edge is low. Moreover, with the increase of microwave power and extraction temperature, the diffusion coefficient is gradually increased, and the flow rate increases, which is more conducive to the extraction process as time goes by. This study provides a theoretical basis for the microwave-assisted extraction of tea polyphenols, reducing experimental workload and material waste.

1. INTRODUCTION

Tea has a long history in China, and a large amount of tea and defective products are discarded during the production and processing of tea. However, these discarded tea leaves have plenty of tea polyphenols and other functional components. Tea polyphenols are a polyphenol compound found, which is a yellowish-green powder. It includes catechins, acetophenones, and phenolic acids [1]. Tea polyphenols have demonstrated antioxidant, anti-aging, antibacterial, antiviral, and other health-promoting bioactivities, with potential applications in cosmetology, radiation protection, diuretics, fatigue reduction, blood glucose regulation, lipid modulation, and medicine [1–3]. Thus, extracting tea polyphenols from defective products can transform these substances into value-added components.

Currently, tea polyphenol extraction methods mainly include solvent extraction, metal ion precipitation, and ultrasonic extraction [4]. However, most of these methods have some drawbacks, such as being time-consuming, yielding low results, leaving behind toxic metal ion residues, and having high production costs [5]. Microwave-assisted extraction (MAE) has been proven to be a promising alternative method [6]. However, existing studies on tea polyphenol extracts primarily rely on experimental methods and lack theoretical analysis. Therefore, the temperature, flow rate, and diffusion distribution of tea polyphenol extraction system for defective green tea extraction are analyzed from the view of heat and mass transfer. The coupling mechanism of physical fields such as temperature field, fluid parameter field, and concentration field during microwave extraction is fully considered in this paper, and the above parameters provide a theoretical basis for studying the extraction conditions of tea polyphenols.

2. HEAT AND MASS TRANSFER PROCESS OF MICROWAVE EXTRACTION OF TEA POLYPHENOLS

2.1. Measurement of Dielectric Properties of Green Tea Extract

Because dielectric property is an important parameter in MAE, the dielectric properties of the extraction fluid needed for the calculations are measured using the ridge waveguide method [7], and the experimental system is built as shown in Fig. 1.

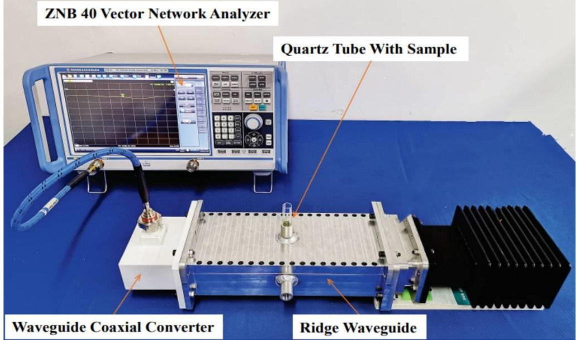

FIGURE 1. Experimental system for measuring dielectric constant.

The measured data are fitted by Matlab, and the relation equation is fitted by a cubic polynomial with the mediator real and imaginary parts, respectively: $R^2 = 0.9683$, $R^2 = 0.9889$, and if $R^2 > 0.9$, the fitting effect is considered to be good, which shows the significant relationship between dielectric

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* Corresponding author: Zhengming Tang (tangzm@cwnu.edu.cn).

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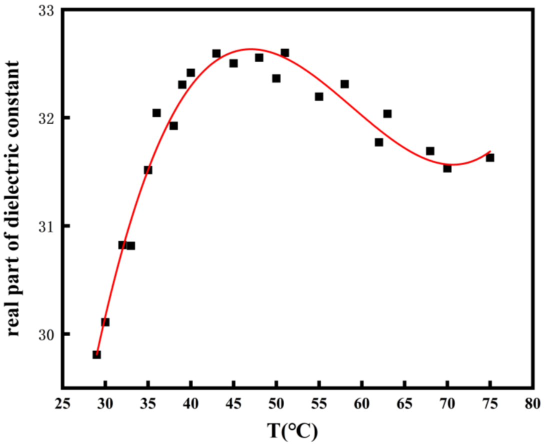

FIGURE 2. Relationship between the real part of dielectric constant and temperature.

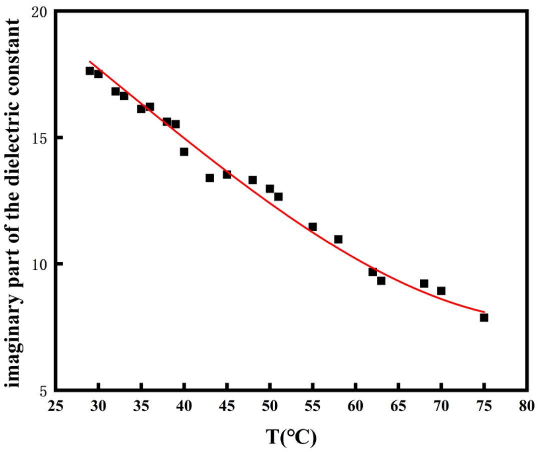

FIGURE 3. Relationship between the imaginary part of dielectric constant and temperature.

properties and temperature. Origin 2021 software is used to plot the data, and the relationships between the real and imaginary parts of the dielectric constant and temperature are obtained, as shown in Fig. 2 and Fig. 3, respectively.

By fitting the measured data with the origin, the law of changes of the real and imaginary parts of the dielectric constant with temperature is obtained, as shown in the following formula:

$$\begin{aligned} \varepsilon' &= 3.0207+1.6183*T-0.0287*T^2+0.0002*T^3 \\ \varepsilon'' &= 24.325-0.1902*T-0.0020*T^2+0.00002*T^3 \end{aligned} \tag{1}$$

As can be seen in Fig. 2, the real part of the dielectric constant increases at first and then decreases with the increase of temperature. The possible reasons are as follows: At the beginning of the extraction, the temperature of the solution rises; the cell wall and cell membrane break; the polar molecules in the solution increase, resulting in a gradual increase in dielectric constant. As the extraction proceeds, some components of the tea extract solution will undergo oxidation, decomposition, or complex reaction, and ethanol as a solvent will volatilize as time goes by, which will lead to a decrease in polar molecules in the solution. Hence, the real part of the dielectric constant decreases. Fig. 3 shows that the imaginary part of the dielectric constant gradually decreases with the increase of temperature. The possible reason is that at the beginning, 70% ethanol mainly exists in the solution. The imaginary part of the dielectric constant is relatively large, and as the temperature increases, ethanol will evaporate. Therefore, the imaginary part of the dielectric constant shows a gradually decreasing trend.

2.2. Model Assumption

To study the heat and mass transfer law of the extraction solution during the MAE process, the following assumptions are made for simplifying the modeling. The initial temperature of the extracting solution and the concentration of tea polyphenols are uniform, and the mass of the tea powder is negligible relative to the mass of the solvent ethanol so that the mass of the tea powder is ignored in the equation of conservation of mass. The volume of the ethanol solvent does not change during the whole process of the extraction; the extraction vessel does not absorb microwave energy; the thickness of the extraction vessel is ignored [8].

2.3. Modeling and Meshing

The heat and mass transfer model and grid division results of tea polyphenols for extracted by COMSOL are shown in Fig. 4. The model parameters are listed in Table 1. Among them, the dimensions of the microwave cavity refer to the real parameters of the microwave extractor (XH-300B). Normalized Power Absorption (NPA) is defined as the ratio of the average simulated dissipated power in the lossy medium to the effective input power, which can be used to determine the mesh size. The mesh division is determined in accordance with the mesh size when the NPA value hardly changes with finer mesh size [9]. Finally, the number of grid cells is 40091, and the average mesh quality is 0.66.

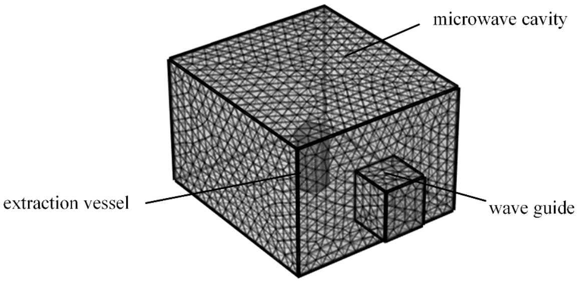

FIGURE 4. Mesh characterization of MAE coupled multiphysics field models.

2.4. Setting of Control Equations and Boundary Conditions

Maxwell's equations are used to solve the electric field distribution of microwaves within the extraction system [10, 11]:

$$\nabla \cdot \frac{1}{\mu_r}(\nabla \cdot E) + k_0^2 \frac{j\sigma}{\omega \varepsilon_0} \cdot E = k_0^2 \varepsilon_r \cdot E \tag{2}$$

TABLE 1. MAE multiphysics field coupling model parameters.

variable symbol	variable name	values and units
wo	Microwave Cavity Width	335 mm
do	Microwave Cavity Depth	375 mm
ho	Microwave Cavity Height	245 mm
wg	Waveguide Width	95 mm
dg	Waveguide Depth	81 mm
hg	Waveguide Height	80 mm
Rp	Extraction vessel radius	28.8 mm
hp	Extraction vessel height	100 mm
M_1	The molar mass of target molecule	458.375 g/mol
M_2	Molar mass of solvent	46 g/mol
m	Sample mass	2 g
ε	Relative dielectric constant	$\varepsilon = \varepsilon' - j \cdot \varepsilon''$

TABLE 2. Initial conditions, boundary conditions, and equations [15].

Physical Fields	Initial conditions	Boundary conditions and equations
Microwave Field	$t = 0, E = 0, H = 0$	(1) The boundary between the microwave cavity and the waveguide wall: $n \cdot H = 0, n \cdot E = 0$ (2) Boundaries of glass, solvent, and air: $n(H2 - H1) = 0, n(E2 - E1) = 0$ $\sqrt{\frac{\mu_0\mu_r}{\varepsilon_0\varepsilon_r - j\sigma/\omega}}\, n \cdot H + E - (n \cdot E)n = (n \cdot E_s)n - E_s$
Temperature field	$t = 0, T = T_0$	
Concentration field	$t = 0, c_0 = 0$	At the interface between the solvent and the glass: $\nabla_c = 0,\ n(k\nabla T) = h(T - T_\infty)$

where E is electric field strength, V/m; ε_0 is the vacuum dielectric constant (8.854×10^{-12} F/m); ε_r is the relative dielectric constant; σ is the electrical conductivity, S/m; μ_r is the relative dielectric constant; ω is the angular velocity, rad/s; k_0 is the wave number of free space.

The electromagnetic field heating component is a function of the electric field and is calculated by the following equation [12]:

$$Q = 2\pi f \varepsilon_0 \varepsilon'' \left|E\right|^2 \tag{3}$$

where f is the microwave frequency of 2450 MHz; Q is the electromagnetic power loss; ε'' is the imaginary part of the dielectric constant. The average power loss density Q calculated in Equation (4) is substituted into the Fourier energy conservation equation to obtain the temperature distribution [13, 14]:

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p u \cdot \nabla T = \nabla \cdot (k \nabla T) + Q \tag{4}$$

where C_p is the specific heat capacity, J/kg·K; k is the coefficient of thermal diffusion, W/m·K; u is the velocity vector; ρ is the material density, kg/m^3; and T is the temperature, °C. In the MAE process, the temperature in the extraction system generally does not exceed the boiling point of the solvent, so there is no phase transition, assuming that the density is a constant, $\rho C_p u \cdot \nabla T = 0$.

Based on the above solution results, the concentration field can be calculated from the law of conservation of mass by establishing a relationship through the Arrhenius equation:

$$\frac{\partial C_i}{\partial t} + \mu \cdot \nabla c_i = \nabla \cdot (D \nabla c_i) \tag{5}$$

where c_i is the concentration of the extraction target component; D is the diffusion coefficient of the extraction target component; μ is the flow rate of the extraction target component.

The boundary conditions and equations required to simulate the MAE heat and mass transfer process are listed in Table 2.

3. RESULTS AND ANALYSIS

3.1. Effect of Microwave Power on Temperature Distribution

With the same extraction time of 60 s, the internal temperature distribution of the extract is studied when the microwave power is 150 W, 250 W, 350 W, and 450 W. The temperature of the extract is increased by the high-frequency rotation of the polar molecules in the extraction system. The polar molecules in the extraction system under the action of microwave rotate at high frequency, causing the particles in the material to undergo intense friction, resulting in an increase in the temperature of the extraction solution.

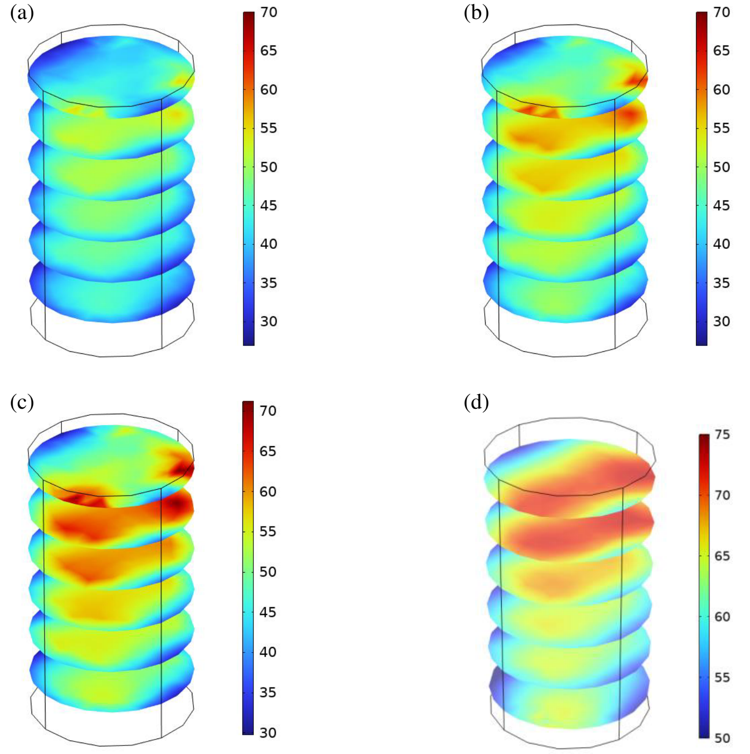

FIGURE 5. Temperature distribution of the extraction solution at different microwave powers. (a) 150 W; (b) 250 W; (c) 350 W; (d) 450 W. (°C).

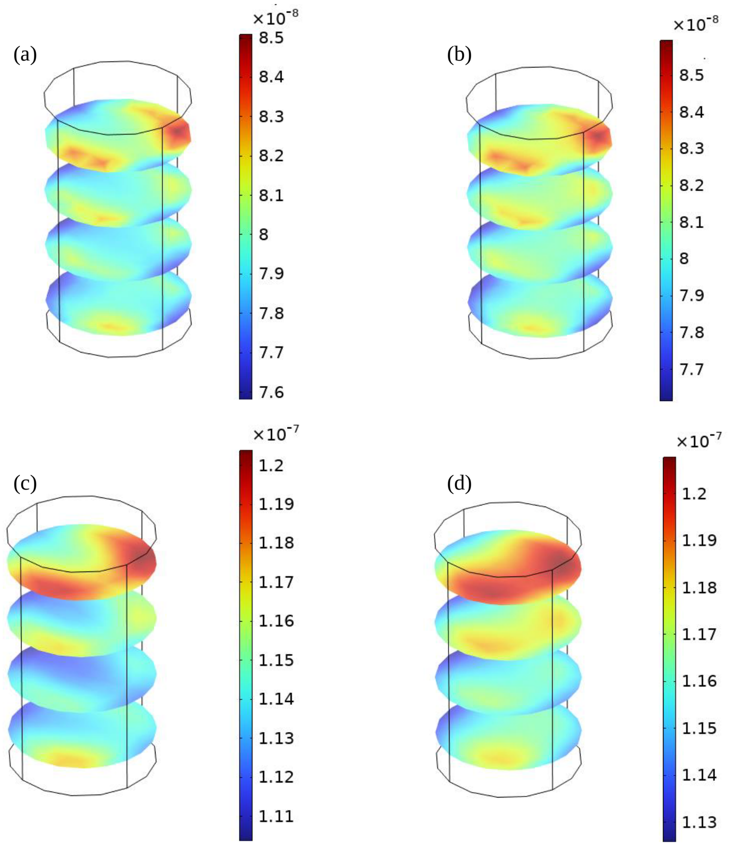

FIGURE 6. Distribution of diffusion coefficients of the extraction system at different microwave powers. (a) 150 W; (b) 250 W; (c) 350 W; (d) 450 W. (m^2/s).

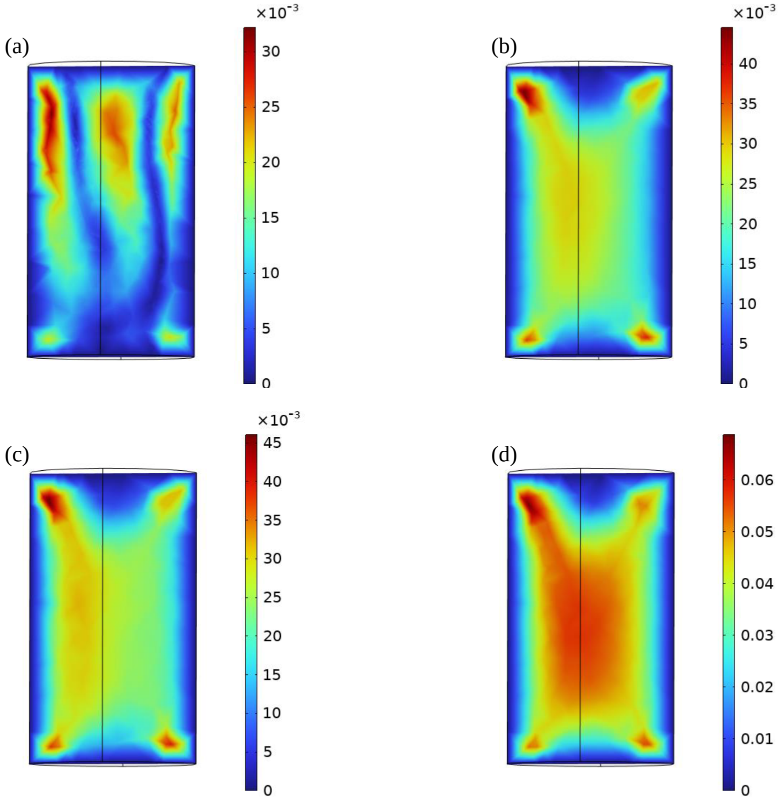

FIGURE 7. Flow rate distribution of the extraction system at different action times. (a) 20 s; (b) 40 s; (c) 60 s; (d) 80 s. (m/s).

It can be seen from Fig. 5 that the temperature of the extraction system is obviously uneven. The middle temperature of the extraction solution is high and the edge is low. Meanwhile, some hot spot appears beside the waveguide, which is due to the different microwave powers, and the higher the temperature of the waveguide port is, the closer it is. At the same time, the higher the microwave power is, the higher the temperature of the extraction system is, which can be attributed to the effect of the microwave power on the extraction liquid. The greater the electric field intensity is, the stronger the dipole rotation effect is; the more the microwaves absorbed by the extraction liquid; the faster the temperature rises. Microwave enhances the heat transfer effect, affects the extraction process, and reveals the extraction mechanism of tea polyphenols assisted by microwave from the perspective of heat transfer.

3.2. Effect of Microwave Power on the Distribution Coefficients of the Extraction Solution

The distribution of diffusion coefficients at the 60th second is shown in Fig. 6, when the microwave powers are 150 W, 250 W, 350 W, and 450 W, respectively.

As shown in Fig. 6, the distribution of the diffusion coefficient is crucial for the mass transfer within the extraction system. As can be seen from the figure, the distribution of the diffusion coefficient is basically consistent with its temperature change, and the diffusion coefficient gradually expands with the increase of microwave power at the same time, which in turn accelerates the diffusion of substances. According to MAE process analysis, the cell is overheated; the cell wall and cell membrane are damaged; the cell wall and cell membrane are broken, and the components in the cell are released; and tea polyphenols penetrate into the extraction solution through the cell wall. The target components of tea polyphenol molecules are quickly dissolved from the cell, revealing the mechanism of microwave-assisted extraction of tea polyphenols from the perspective of mass transfer.

3.3. Flow Rate Distribution of the Extraction Solution at Different Times

The flow rate distribution in the extraction system at different times is investigated when the microwave power is 250 W. The flow rate distributions of the yz-plane in the extraction solution at 20 s, 40 s, 60 s, and 80 s are shown in Fig. 7. Different colors indicate different diffusion speeds of tea polyphenols, and the diffusion distribution diagram presents the diffusion phenomenon of tea polyphenols under microwave action more intuitively, which helps to reveal the mass transfer mechanism of MAE.

Figure 7 shows the distribution of the flow rate of the extraction system which shows a tendency of diffusion from the center to the surroundings, and the flow rate in the extraction system increases with the progress of extraction. An increase in flow rate can accelerate the mixing of the extract, which in turn accelerates the mass transfer process.

4. CONCLUSION

Based on the measurement of the dielectric properties of tea extract, the heat and mass transfer model of tea polyphenols extracted by microwave is established by coupling the electromagnetic field, fluid field, temperature field, and concentration field. The temperature distribution, diffusion coefficient, and flow rate of the extraction systemare are analyzed from the view of heat and mass transfer. The simulation results show that the temperature distribution in the extraction system is not uniform. Specifically, in the center area of the extract, the temperature of the liquid is higher, while the temperature in edge area is lower. The higher the microwave power is, the higher the center temperature of the extraction system is, and the higher the diffusion coefficient is. Moreover, the flow rate increases as time goes by, and the diffusion of tea polyphenols is enhanced. The more obvious the diffusion is, the more favorable it is for the extraction. This study provides a theoretical basis for the study of the optimum technological conditions for extracting tea polyphenols by microwave.

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