J. of Supercritical Fluids 96 (2015) 200–210

Contents lists available at ScienceDirect

The Journal of Supercritical Fluids

journal homepage: www.elsevier.com/locate/supflu

Acoustic wave generation in near-critical supercritical fluids: Effects on mass transfer and extraction

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ARTICLE INFO

Article history:
Received 25 April 2014
Received in revised form 17 August 2014
Accepted 17 August 2014
Available online 28 September 2014

Keywords:
Supercritical fluid extraction
Acoustic waves
Mass transfer enhancement
Computational fluid dynamics

ABSTRACT

The supercritical fluid extraction (SFE) process has attracted increasing interest over the recent decades. This is particularly motivated by concerns regarding the environmental effects of using traditional organic (potentially toxic) solvents. However, the conventional process of extraction using supercritical solvents has a slow dynamics even when solute free solvent is re-circulated and therefore improvements in the extraction process are required. The use of acoustic waves represents a potential efficient way of enhancing mass transfer processes in the supercritical region. Mass transfer enhancement using high amplitude resonant acoustic waves is investigated in this study. To carry out the investigation, three different cases are simulated. Acoustically augmented flow and transport in supercritical fluid (CO_2) generated by resonant standing wave in a cylindrical shaped resonator (case 1) is simulated first. Based on the knowledge gathered from the simulations of the first case, simulations of acoustically augmented flow and transport in supercritical extraction systems, namely – in a fixed bed extractor (case 2) and membrane contactor (case 3) are also carried out. The oscillatory flow fields in these cases are created by the vibration of one of the end walls of the systems. A real-fluid model for representing the thermo-physical and transport properties of the supercritical fluid is considered. The fully compressible form of the Navier–Stokes equations is used to model the flow fields and an implicit time-marching scheme is used to solve the equations. Due to diverging thermo-physical properties of supercritical fluid near the critical point, large scale oscillations are generated even for small sound field intensity. The effects of near-critical property variations on the formation process of the streaming structures are also investigated. Predicted results from these simulations confirm that acoustic waves significantly accelerate the kinetics of the supercritical extraction process, especially in the near-critical region of operation and improve the final extraction yield. These improvements are attributed to an increase in the overall convective mass transfer coefficient.

1. Introduction

Supercritical fluids are commonly used in industries now-a-days as 'green' solvents and efficient heat transport media [1]. Due to the increasing number of applications of supercritical fluids in chemical and thermal process industries, convective transport in near-critical and supercritical fluids has drawn a lot attention to the researchers in the recent decade [2–5]. Although, supercritical fluid is widely used in process industries and in power generation, the transport dynamics is relatively slow near the critical point and therefore improvements in convective transport (both thermal and mass) are required [6]. Near the critical point of the solvent, the supercritical fluid extraction process has a reduced yield due to the lower solubility of the solvent. Hence, the operating pressure of the extraction process is set relatively higher than the critical pressure (i.e. far from the critical pressure). However, large scale oscillations can be produced in the near-critical regimes of operation due to the high compressibility and density of the solvent at those (near-critical) states. The use of acoustic excitation represents a potential efficient way of enhancing transport processes in the near critical region [7,8]. This is due to the effects produced by compressions and decompressions, as well as by radiation pressure, streaming, etc. It is well-known that sound sources may generate flow fields in which the particle velocities are not simply sinusoidal, and a pattern of time-independent vertical flows or steady circulations is often observed in the body of compressible media [9,10]. These second-order steady flow patterns are known as '*acoustic streaming*'. The steady flow velocities increase as the intensity of the sound sources increases, however the secondary streaming velocity magnitude is much lower than the primary oscillatory particle velocity magnitude, even at high sound source intensity levels. Acoustic streaming may be effective in accelerating

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http://dx.doi.org/10.1016/j.supflu.2014.08.032
0896-8446/

certain kinds of rate processes, especially the cooling process of electronic systems under micro-gravity conditions, where natural convections are greatly reduced or completely eliminated. Such secondary streaming flows can also be employed to enhance mixing processes and to augment heat and mass transfer through resonator walls. Studies on acoustic wave induced convective transport inside an enclosure have been conducted by several researchers [10–15]. Riera et al. [16,17] proposed power ultrasound assisted supercritical fluid extraction to enhance the mass transport in almond oil extraction. Although results from both of these studies show that power ultrasound significantly accelerates the kinetics of the process and improves the final extraction yield, a clear physical description of the transport processes were not specified in either one of the papers. Recently, we developed a numerical model to simulate the supercritical fluid extraction of caffeine in a fixed bed (coffee beans) extractor [18]. The effects of acoustically augmented flows on the extraction process were studied with the help of this model. However, formation of any second-order vortical flow structures was not observed in the fixed bed extractor.

In the present study, the generation and propagation of mechanically driven acoustic waves in sub- and supercritical carbon dioxide are investigated via a high-order numerical scheme. Supercritical fluid extraction of caffeine from a solid matrix of coffee beans in a fixed bed extractor is considered following our previous work [18]. Ethanol recovery from an aqueous feed using supercritical carbon dioxide in a membrane contactor is also simulated. Numerical models to simulate the SFE process in both these systems are developed. A cylindrical shaped resonator filled with carbon dioxide is considered for this case. The NIST database 23 [19] is used to obtain the ρ–p–T relations for supercritical carbon dioxide as well as the different thermo-physical and transport properties of the fluid. The effects of system pressure and near-critical property variations on the formation process of the streaming structures are investigated. Novel applications of the acoustically augmented supercritical fluid (CO_2) extraction in fixed bed extractor (caffeine extraction) and membrane contactor (ethanol recovery) are demonstrated numerically. Due to the selective and non-toxic extraction properties, supercritical fluids (supercritical CO_2 in this case) are a good candidate for recovering chemicals from aqueous feed using the membrane contactors. The low binary diffusivity and solubility of the solvent makes the dynamics of the process slow. Hence, acoustically excited waves can be used in these systems to enhance the transport dynamics.

2. Physical background

For pure substances, there is an inflection point in the critical isotherm on a p–v diagram. This means that at the critical point, $(\partial p/\partial v)_T = (\partial^2 p/\partial v^2)_T = 0$ near the critical point, the thermo-physical properties of fluids exhibit unusual behaviors; the specific heat (c_p) and the isothermal compressibility, $(1/\rho)(\partial\rho/\partial p)_T$ show strong divergence, causing a vanishing thermal diffusivity (α). The critical pressure (p_c) for carbon dioxide is 7.3773 MPa, the critical temperature (T_c) is 304.1282 K and the critical density (ρ_c) is 467.6 kg/m^3 [20]. If both pressure and temperature are beyond each critical value, carbon dioxide is in a supercritical state. Fig. 1 shows the density and thermal diffusivity vs. temperature diagram for carbon dioxide with changing pressure. The density at atmospheric pressure is almost constant, while the density in the supercritical-pressure condition widely varies across the phase interface from the liquid or gas phase to the supercritical fluid phase. Hence, even around 10 K above the critical temperature (304.1282 K), carbon dioxide is found to be compressible but the density remains high. The thermal diffusivity (also plotted in Fig. 1) is zero at the critical point and shows a strong divergence around the pseudo-critical states. Due to these divergences of thermo-physicals properties near the critical point, thermal diffusion at 10 K above the critical point (at the critical isochors) is about 170 times slower than that at atmospheric conditions (and about 3000 times slower at $T_c + 0.1$ K, $\rho = \rho_c$). The binary diffusion coefficient of ethanol (C_2H_5OH) in supercritical carbon dioxide (as obtained from Catchpole and King [21] correlation), D_{12} is shown in Fig. 2. It is observed that the binary diffusion coefficient of ethanol in supercritical carbon dioxide is a strong function of pressure and temperature and it closely follows the density variation of supercritical carbon dioxide in the near critical region with small coefficient of diffusivity near the critical point.

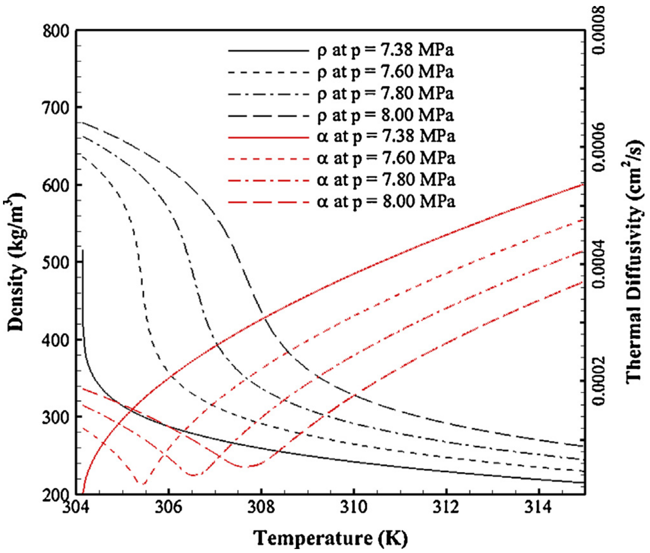

Fig. 1. Variation of density, ρ and thermal diffusivity, α as functions of pressure and temperature for near-critical carbon dioxide [19].

3. Problem description

To numerically investigate the flow and transport induced by mechanically driven acoustic waves in sub- and supercritical carbon dioxide, three different cases are studied. The first case

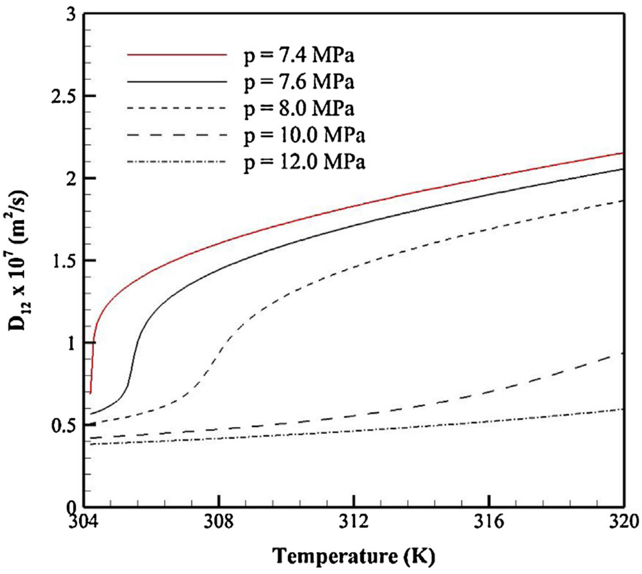

Fig. 2. Variation of binary diffusion coefficient (D_{12}) of ethanol in supercritical carbon dioxide as functions of pressure and temperature [21].