CFD FLOW AND MASS TRANSFER SIMULATION IN A PACKED BED WITH SUPERCRITICAL SOLVENT: PRELIMINARY RESULTS

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The CFD simulation methodology is applied to the study of the natural convection flow in equipment used in SCF technology that involves a packed bed of particles. The ranges of heat and mass transfer are defined where those free convective effects show themselves. This happens at low particle Reynolds number and large Rayleigh (either GrPr or GrSc) numbers, for both heat and mass transport. Rayleigh number is large in SCF because of liquid-like densities and gas-like viscosities.

Free convection effects of heat and mass transfer are of prime importance when modelling the behaviour of supercritical fluids in fixed bed reaction equipment and in solid extractors. Density gradients, caused by temperature or composition variations, tend to control the volume forces and the overall flow pattern of the fluid, making the buoyancy terms of the Navier – Stokes equations an important term to model when doing a CFD simulation of a fixed bed under the described conditions. Buoyancy and sinking forces interact with forced flow in extraction with SC fluids, making them sensitive to the gravity field [1-3]

In order to simulate a supercritical extraction, a Toluene – CO_2 system is analysed. Mesh sensitivity analysis is done and results of a preliminary simulation are shown. Carbon dioxide in the supercritical conditions has been selected as a flowing fluid. Their transport properties at high pressure are incorporated within a CFD commercial code in order to estimate them online within the simulation process. Particle-to-fluid mass transfer is analysed [4], and flow and composition contour fields are presented, and flow pattern is analysed according to density profiles obtained. Downflow operation regime is modelled.

INTRODUCTION

During the last decades, supercritical fluid extraction (SCFE) has been successfully applied as an alternative to conventional separation methods in the chemical, food, pharmaceutical, biochemical and environmental industries. SC fluids often prove to be efficient extractants with better transport properties (diffusivity, mass transfer coefficient, penetration ability) than most commonly employed liquid solvents. When using CO_2 as a fluid (T_c = 304.1 K; P_c = 7.38 MPa), further advantages are the energy savings and the reduction of degradation of temperature-sensitive substances, as well as the advantages derived from its nonflammability, nontoxicity, easy regeneration, and commercial availability. CFD has proven to be a reliable tool when modeling heat and mass transfer in fixed beds. Its capabilities to solve three-dimensional Navier-Stokes equations together with mass and energy balances make it a good choice when modeling transport phenomena in a fixed bed.

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1. GEOMETRICAL MODEL

An 11-sphere arrangement, with 9 particle-to-wall contact points and 14 particle-to-particle contact points was built. A 4-layer arrangement with a 60^{o} rotation around the reactor axis within each layer and with a diameters ratio of 3.923 was chosen to be the geometrical model (Fig. 1). Modeled geometry has been constructed following the bottom-up technique (generating surfaces and volumes from nodes and edges) in order to control mesh size around critical points (i.e. particle-to-particle and particle-to-wall contact points), necessary to avoid grid elements skewness, and also to gain computational resources by reducing the number of elements in zones of low interest (i.e. geometrical zones away from contact points).

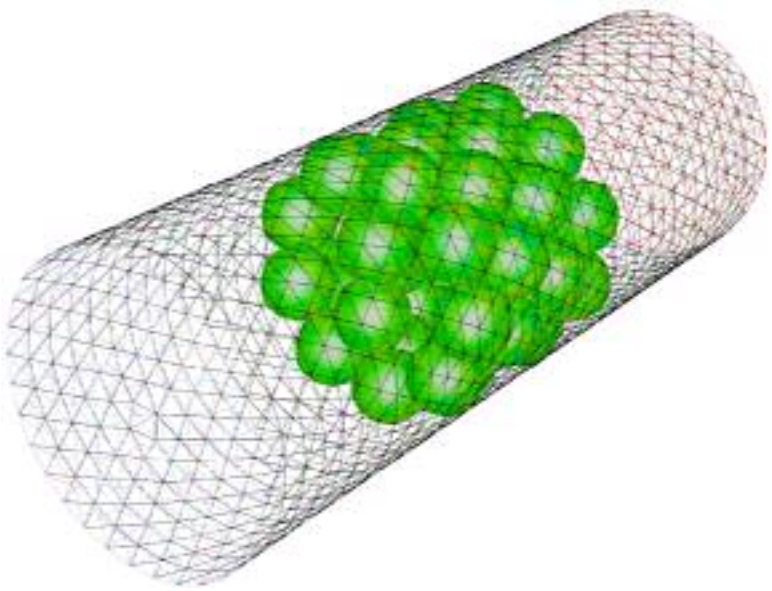

Figure 1. Fixed bed geometrical model

2. MESH SENSITIVITY ANALYSIS

In order to evaluate the mesh response to different flow regimes, a forced convection situation was analyzed for the binary system Toluene – CO_2. Four different mesh densities were tested at four flow velocities, analyzing laminar, transition and turbulent flow regions.

In the CFD model, CO_2 (298 K, 1 ATM) was circulated through a fixed bed of spheres covered with liquid toluene in equilibrium with the circulating gas. A First-Order discretization scheme was adopted, and under-relaxation parameters for pressure, momentum, body forces, turbulence, C_7H_8 and energy were set at 0.2, 0.4, 0.4, 0.5, 0.8 and 0.5 respectively.

For each simulation, velocity, composition and density profiles along the bed were recorded and analyzed. With the collected data, the mass transfer coefficient was obtained from the mass balance equation. From the obtained values of k, Sh was computed and compared with the correlations proposed by Steinberger & Treybal [5] and Wakao & Kaguei [6], and with experimental measurements extracted from Resnick & White [7]. Results are shown in Figure 2. It can be noticed that in the laminar flow zone (Re < 100), the results don't show

dependence on the mesh density. For higher values of *Re* there is a divergence between the results obtained for both meshes, probably due to the fact that at higher *Re*, turbulent transport term in the transport equation becomes more important. In RANS modeling, the equations possess a smooth exact solution, and the numerical solution approaches that solution as we refine the grid. The aim of grid refinement is numerical accuracy. Therefore, for our specific study case, an accurate turbulence modeling requires a denser mesh around the particles surface in order to capture in a more suitable way the involved turbulence phenomena.

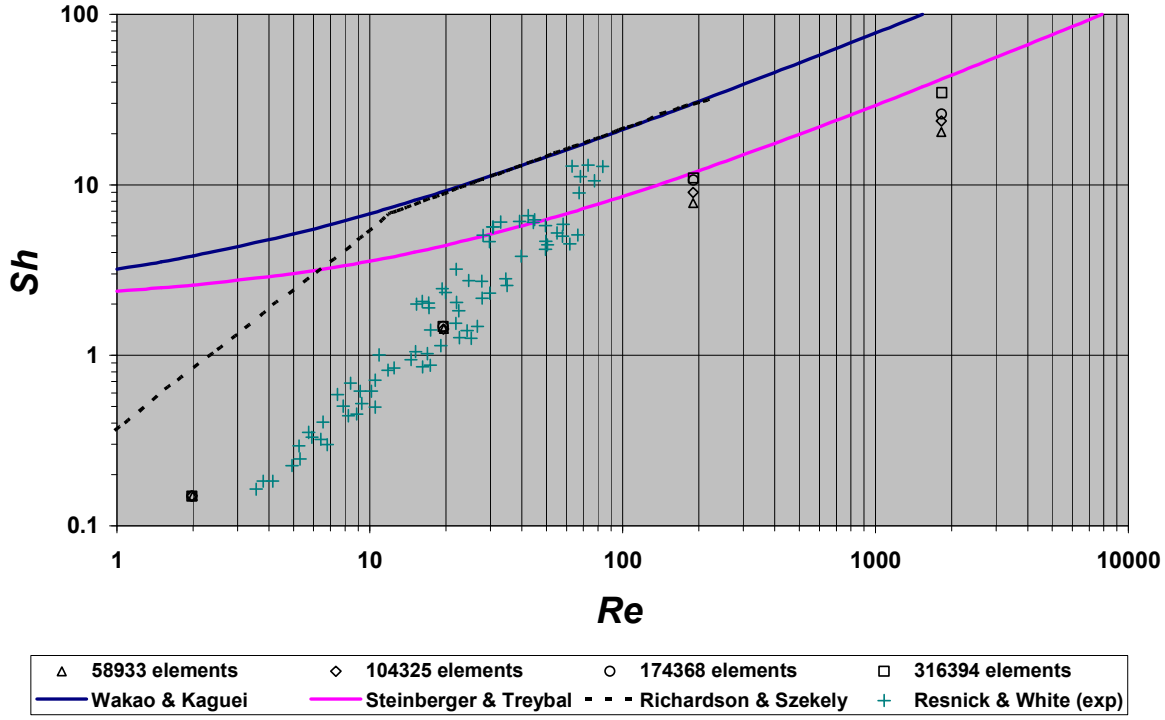

Figure 2. Mesh sensitivity analysis.

The results obtained with the finer mesh fit better the prediction of Steinberger & Treybal [5] in the transition and turbulent flow zone (Re > 100), probably due to a better capture of the vorticity energetic scales associated effects. For lower values of *Re* it can be seen that the obtained results present a considerable deviation from the widely accepted correlations, but a good agreement is shown when comparing numerical results with the experimental data obtained by Resnick & White [7].

3. HIGH PRESSURE SUPERCRITICAL EXTRACTION: PRELIMINARY MODELING

In order to evaluate the computational time, a preliminary simulation was set, imposing CO_2 at 354K and 1950 kPa as circulating fluid (u = 1 x 10^{-5} m/s). Equilibrium conditions were imposed to particle surface. A coupled solver with second order discretization scheme was chosen as the numerical solver. Simulation was run under unsteady conditions, and flow rate, density and molar concentration at the outlet were recorded as convergence monitors. Velocity and concentration fields can be seen in Figures 3 and 4.

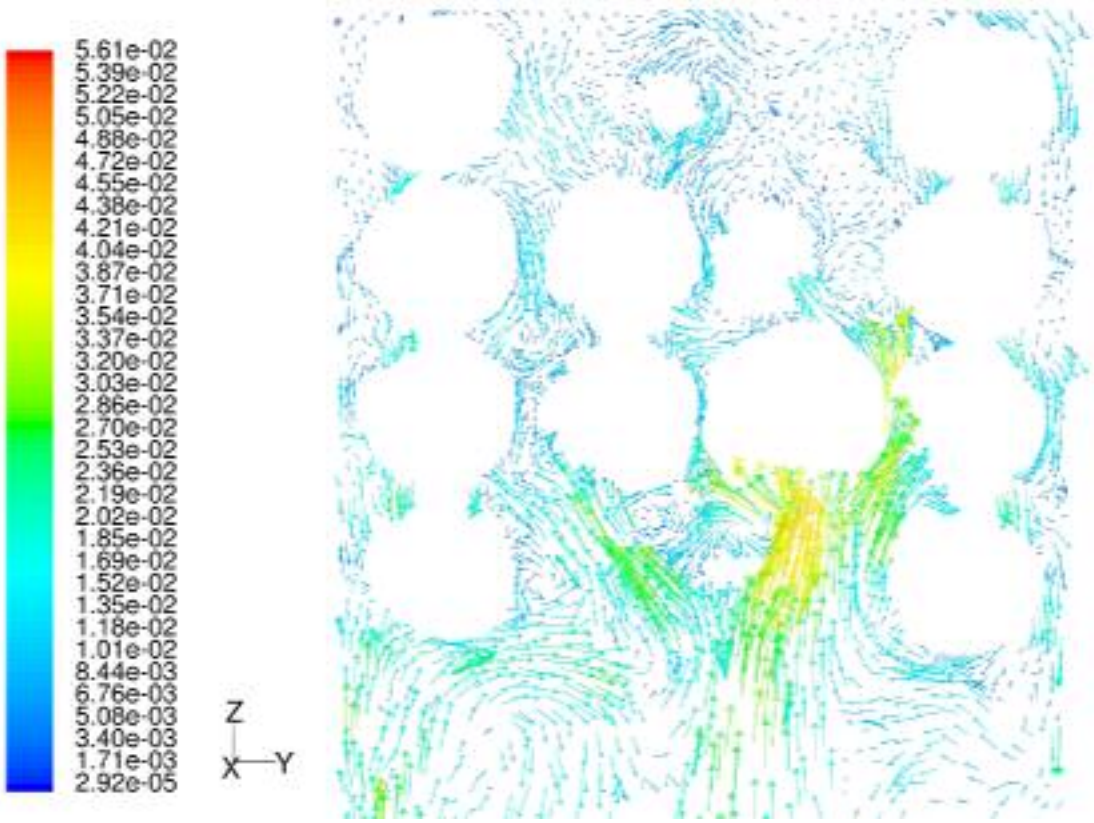

Figure 3. Velocity field for the preliminary simulation

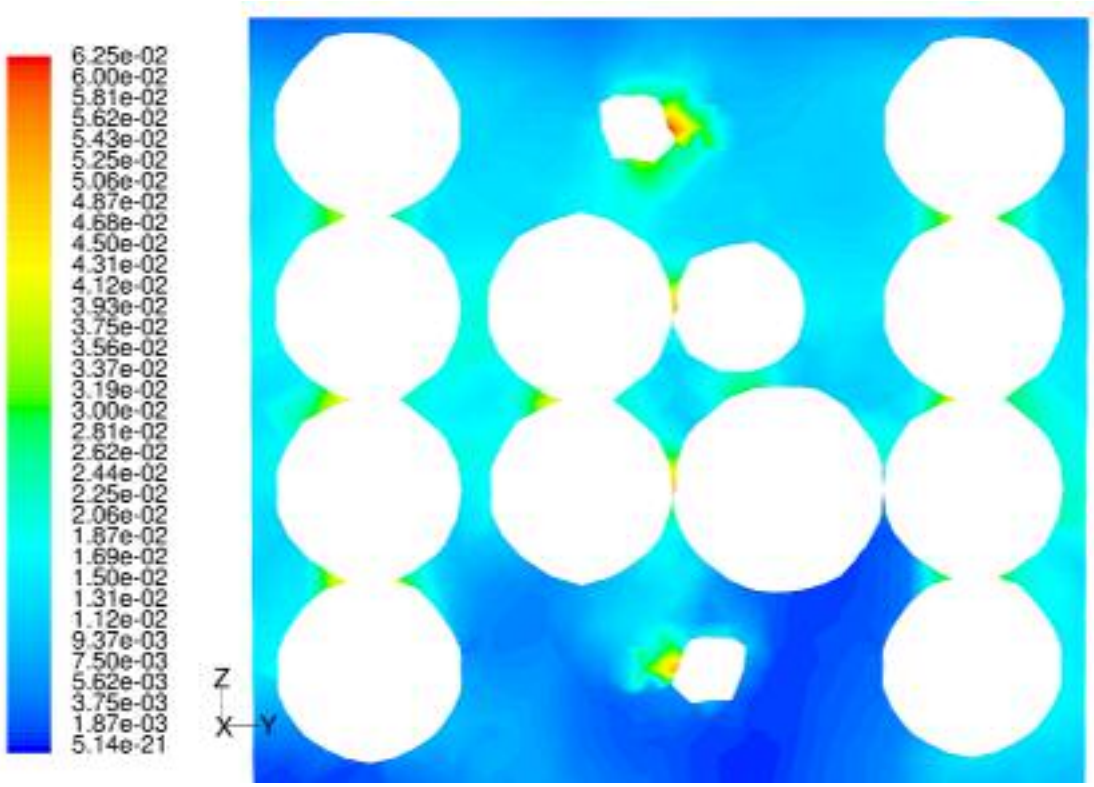

Figure 4. C_7H_8 molar concentration field (in kmol/m^3) for the preliminary simulation

CONCLUSIONS

CFD proves to be a reliable tool when modeling mass transfer in packed beds. The solution of 3D Navier-Stokes equations together with mass and energy balances can be a suitable way to estimate mass transfer coefficients in fixed beds without experimentation.

At low velocities (laminar flow), the numerical results obtained are independent of the mesh density. As supercritical extractions are carried in laminar flow regime, it can be expected to have good numerical results with a coarse mesh, saving computational time in simulations.

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