

Modeling Of Residence Time Distribution in A Catalytic Distillation Column With Bale Packing For Tertiary Butyl Alcohol Production

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Abstract-The computational fluid dynamics (CFD) technique was used to investigate the aerodynamics of catalytic Bale packing (used for the synthesis of tertiary butyl alcohol (TBA) in a pilot scale catalytic distillation column) using commercial CFD software, Fluent 6.1. The global porosity of catalytic bales was considered instead of geometrical features for 2D simplification. The residence time distribution (RTD) simulations were performed and the results compared with the experimental studies. A user defined scalar technique for RTD was found successful in simulating the exit age at different Reynold numbers. The comparison with experimental results indicated a satisfactory agreement. The CFD simulation is an efficient diagnostic tool for determination of RTD.

Keywords: CFD, User defined Scalars, Bales packing, and Residence time distribution modeling

I. INTRODUCTION

In catalytic distillation columns, flow dynamics and mass transfer properties of catalyst packing plays a crucial role to achieve better separation and product yield [1]. Computational fluid dynamics CFD is a valuable aid to understand flow dynamics and mass transfer properties of catalyst. Some of the limitations encountered experimentally can be overcome by CFD modelling techniques. The work of Higler et al [2], Gulik [3] and Calis et al [4], dealt with the hydrodynamics and mass transfer analysis in different structural catalytic packing using CFD technique. This work discusses a CFD technique that involves the aerodynamic analysis of Bales type catalytic packing, which is used for pilot scale studies in the synthesis of Tertiary Butyl Alcohol (TBA). The TBA is one of the important octane enhancers in gasoline pool. Commercially it is produced by the acid-catalyzed hydration of Isobutylene. The reaction occurs in the liquid phase in the presence of 50-60% H₂SO₄ at mild temperature, the yield is approximately 95% [5]. The catalytic distillation is an integration of reaction and separation in a single vessel. Some chemicals are presently investigated on bench scale by using catalytic distillation technique [1, 6, and 7]. One of the chemical TBA has recently been explored by Zhang et al [8] on counter current fixed bed reactor (CCFBR) configuration, which is loaded with Bales type packing. This paper study the aerodynamics of catalytic bales type packing using CFD for residence time distribution simulation and wall wiper effects. The novelty of this work is the use of ANSYS Fluent software for CFD residence time distribution simulation, study of internal effects of TBA reactor, and comparison of CFD results with Calis et al [4] experimental data of TBA pilot plant.

II. ANALYSIS OF FLOW DYNAMICS OF CATALYTIC PACKING BY CFD

The CFD simulations show a good output and understanding for the flow dynamics and mass transfer of catalytic packings used for catalytic distillation. Van Baten et al [9] obtained detailed information of liquid velocity distributions, hold-up distributions, and dispersion which is important for logical design of catalytic distillation columns. Axial and radial dispersion for KATAPAK –S structure was studied by Van Baten et al [10] using air-water system and a column with a rectangular cross-section of 30mm x 20mm. The authors compared experimental results with those obtained using CFD simulations, but the agreement was not completely satisfactory. The classic plug-dispersion flow model (PD model) was used to derive the dispersion coefficients from RTD curves.

2.1 Catalytic Bales type packing

The catalytic bales (CB) are extensively used as industrial packing mainly for heterogeneous processes [1, 11]. The characteristics of CB packing highly depend on the flow configuration, catalyst amount loaded in the bales, column diameter and particle size [1, 12, and 13]. The packing consists of steel mesh and glass fiber cloth,

which is wrapped in the form of bales. These bales are sewn shut after catalyst beads are loaded. The resulting belt is rolled with alternating layers of steel mesh to form a cylinder of catalyst bales as shown in figure 1.



Fig.1 Bales Type Catalytic Packing

III. COMPUTATIONAL FLUID DYNAMICS (CFD)

The CFD is the numerical tool of predicting fluid flow, heat transfer, mass transfer, chemical reactions, and related problems by solving the sets of mathematical equations. CFD gives the detailed internal information, which is not possible, experimentally [14]. Most recently CFD been employed for the investigation of reactive distillation hardware [14, 15]. The following conservation equations are used to solve CFD simulation for pressure drop and residence time distribution. The details of equations that CFD solve is given in section IV. In this paper mapping of 2D model similar to experimental reactive distillation column is presented.

The continuity equation is a declaration of conservation of mass [16]. For 2D axisymmetric geometries, the continuity equation is given by

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x}(\rho v_x) + \frac{\partial}{\partial r}(\rho v_r) + \frac{\rho v_r}{r} = S_m \quad (1)$$

Where x is the axial coordinate, r is the radial coordinate, v_x is the axial velocity, and v_r is the radial velocity. The source S_m is the mass added to the continuous phase from the dispersed second phase (e.g due to vaporization of liquid droplets) and any user defined source. The v_x , and v_r are the axial compact way to write equation by use of Einstein notation [15].

$$\frac{\rho \partial U_i}{\partial x_i} = 0 \quad (2)$$

The momentum equation is the declaration of conservation of momentum in each of the three component directions. The three momentum equations are collectively called the Navier-stokes equation [16].

$$\frac{\partial}{\partial t}(\rho U) + \frac{\partial}{\partial x_j}(\rho U_i U_j) = -\frac{\partial p}{\partial x} i + \frac{\partial}{\partial x_j} \left[\mu \left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} - \frac{2 \partial U_k}{3 \partial x_k} S_{ij} \right) \right] + \rho g_i + F_i \quad (3)$$

The term the left hand is convection, and on the right hand side is the pressure gradient, the source terms, the divergence of the stress tensor (which is responsible for the diffusion momentum) and the gravitational force and another generalized term respectively. The most commonly used turbulence model for practical flow problems is the $k-\epsilon$ model [17], which is also called two-equation model. The feature of this model is that it uses one differential equation for the turbulent velocity scale and another for the turbulent length scale. The variables modeled are the turbulent kinetic energy, k , and the rate of dissipation of turbulent kinetic energy ϵ . The equation for the dissipation rate is derived by taking a moment of the Navier-Stokes equation using the fluctuating vorticity. The resulting equation is a relatively complicated equation. The Standard $k-\epsilon$ model is described below [15, 17].

$$\rho U_j \frac{\partial k}{\partial x_j} = \tau_{ij} \frac{\partial U_i}{\partial x_j} - \rho \epsilon + \frac{\partial}{\partial x_j} \left[(\mu + \mu_T / \sigma_k) \frac{\partial k}{\partial x_j} \right] \text{Turbulence Kinetic Energy.} \quad (4)$$

$$\rho U_j \frac{\partial \epsilon}{\partial x_j} = C_{\epsilon 1} \epsilon / k \tau_{ij} \frac{\partial U_i}{\partial x_j} - C_{\epsilon 2} \rho \epsilon^2 / k + \frac{\partial}{\partial x_j} \left[(\mu + \mu_T / \sigma_\epsilon) \frac{\partial \epsilon}{\partial x_j} \right] \text{Dissipation Rate} \quad (5)$$

Where μT is the eddy viscosity: $\mu T = C \mu \rho k^2 / \epsilon$, and $C\epsilon_1=1.44$, $C\epsilon_2=1.92$, $C\mu=0.09$, $\sigma_k=1$, $\sigma_\epsilon=1.3$ are the empirical closure coefficients, which will vary somewhat, depending on the application and what investigators have found. However, the coefficients should agree with the experimental results. Therefore, the coefficients shown above are not universally optimal, and any CFD results obtained using the turbulence k- ϵ model should be closely scrutinized [17].

3.1 Residence Time Distribution

Residence time distribution (RTD) is a valuable aid to give the mixing characteristics and analyzes the performance of the reactor or any process equipment. The information usually consists of flow profile, dispersion, non-ideality, mixing length and channeling inside the equipment. On the basis of this information, design, flow conditions and process parameters are reviewed or optimized. Similarly the performance and flow dynamics characteristics of catalytic packing can also be determined by RTD, but the limitation arises when the insitu information is required [18]. CFD is a non-invasive analysis technique, which give the *insitu* information in detail. This can be done via modeling the computational geometry similar to the real geometry and import it into a CFD software, which offers RTD determination techniques (details are in section IV under methods and procedure).

3.2 - Residence Time Distribution Theory

The time spent by the tracer or a particle in the vessel is called residence time of the tracer in the vessel. The most useful function for continuous-flow processes is the exit age distribution, $E(t)$ as shows in figure 2, which represents the proportion of the fluid that leaves the relevant section in time t .

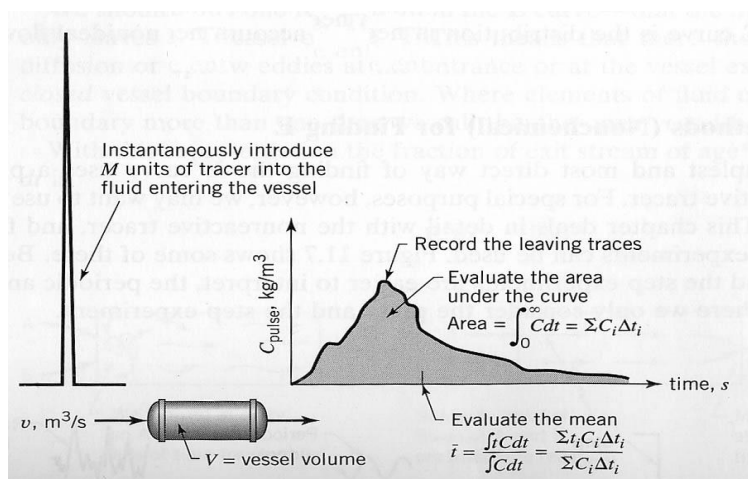


Fig. 2 Exit age distribution curve from pulse tracer experiment [19].

The residence time is measured by injecting a pulse of tracer at the inlet of the section of interest and measuring the concentration at the outlet, $C(t)$, as a function of time (t). The graph of tracer concentration, $C(t)$, versus time (t) is called the C curve and the area under the graph is the amount of tracer injected. The curve can now be normalized; that is, the area under it is equal to 1.0. This is achieved by dividing each $C(t)$ value by the area under the original curve and the values obtained are now equal to the $E(t)$ values mentioned above [19,20]. This can be expressed mathematically, as follows.

$$\text{Area under curve} = \int_0^{\infty} C(t) dt = \text{amount of tracer injected } (M_t) \quad (6)$$

The other function that expresses residence time distribution is the F curve. The $F(t)$ curve is a fraction of the liquid that has a residence time less than time (t) and can be obtained by making a sudden step change in the tracer concentration in the inlet stream. The concentration of tracer in the outlet stream, when normalized, is the $F(t)$ curve. The $F(t)$ related with $E(t)$ as follows.

$$E(t) = dF(t) / dt \quad (7)$$

The mean residence time can be determined from the $E(t)$ or $F(t)$ curves. For the latter, by definition, t_m is given as the time when $F(t)=0.5$. From $E(t)$ curves, t_m is given by the first moment of the distribution [19].

$$t_m = \int_0^{\infty} tE(t)dt \quad (8)$$

The second moment of the distribution is the variance, σ^2 , which describes the measure of the spread of the curve about the mean value and is defined as

$$\sigma^2 = \int_0^{\infty} (t - t_m)^2 E(t) dt \quad (9)$$

In addition to the mean and variance of the curves, the third and fourth moments of the distribution can also be calculated to describe the distribution accurately. These moments are the skewness and kurtosis of the distribution, respectively, which describe the degree of departure from a symmetrical curve and the type of peak of the distribution. The residence time distribution can also be normalized with respect to time by dividing the time values by the mean residence time t_m to give dimensionless time (Θ). The resulting distribution $E(\Theta)$ allows comparison of distributions with different mean residence times [20].

Discrete time intervals to $E(t)$ values may be achieved using the following equations.

$$t_m = \frac{\sum_{i=0}^{\infty} t_i C_i(t)}{\sum_{i=0}^{\infty} C_i(t)} \quad (10)$$

$$\sigma^2 = \frac{\sum_{i=0}^{\infty} t_i^2 C_i(t) - t_m^2}{\sum_{i=0}^{\infty} C_i(t)} \quad (11)$$

Where C_i is the tracer concentration at time t_i .

By using the value of mean residence time and variance, the dispersion number (D/uL where D is the axial or longitudinal dispersion coefficients and u is velocity and L is the length of reactor bed) can be calculated, which is the inverse of mass Peclet number (Pe). The method for calculating the (D/uL) value from the measured residence time distribution is given by Levenspiel [19] and depends on the actual value of (D/uL) and the type of vessel for which the distribution has been measured. The reactor Peclet number is defined as:

$$Pe_r = uL/D \quad (12)$$

For open system (defined as one in which, the fluid flows into and out without disturbance, such as the mid-section of a pipe of constant diameter) the Peclet number can be calculated from following equation [18, 20].

$$\sigma^2 / t_m^2 = 2/Pe_r + 8/Pe_r^2 \quad (13)$$

Equation 12 is used where two-point analysis is required; the equation can be rearranged and solved for Pe_r by iteration using t_m and σ^2 obtained from method of moments.

3.3 Predicting RTD by User Defined Scalar Techniques

Commercial CFD software Fluent® offers several approaches for the determination of residence time distribution. One of them is User defined scalar (UDS) in which passive tracer is introduced. The tracer fluid treated as a continuum is solved by scalar- transport equations for tracer species. The general time dependent equation is written as:

$$d/dt (\text{amount}) + \text{net flux} = \text{source}$$

Where, amount = $\rho V \Phi$ = quantity in cell; Flux is the rate of transport through a cell face. The UDS transport equation solved by Fluent ® is:

$$\partial C/\partial t + \partial(u_x C)/\partial x + \partial(u_y C)/\partial y = D[\partial^2 C/\partial x^2 + \partial^2 C/\partial y^2] + S(C) \quad (14)$$

The calculations are basically the numerical equivalent of the tracer tests undertaken on actual process equipment, in which a chemical or particulate tracer is added to the inlet stream of a reactor and measurements of the exit concentration are then recorded at regular time intervals [21]. The tracer created in Fluent should have identical properties of fluid, so that it does not affect the reactor flow dynamics. The time required by Fluent to solve the equations is reduced by switching off the hydrodynamics equations, which is unnecessary when the solution is independent of time for the fluid and the particle flow fields [21]. The tracer concentration is recorded on the exit of the vessel which gives the normalized concentration against time called exit age distribution or E curve.

IV. METHODS AND PROCEDURE

A commercial CFD software package FLUENT 6.2 [24] was employed to predict Bales type packing aerodynamics in pilot scale reactor. FLUENT is the most widely used CFD program for modeling engineering fluid flows due to its robustness, accuracy and user friendliness. For modeling with FLUENT the three steps followed were: Pre-processing, Solver execution, and Post processing. The details on modelling process, assumptions, description, feed distributor, and solver execution can be seen in Wasi and Imad [23]. The computational domain consists of fifteen wall wipers, which are across 500 mm porous zone. The height of the vessel is 640 mm as shown in figure 3. To avoid higher computational efforts and memory limitation 2D domain are modeled and meshed with highly refined structural meshing scheme. In order to check grid independence of solution, simulation was run on different grid sizes and finally 2.5 grid cell is selected. Similarly a single CB is assumed instead of five bales with same number of wall wipers which are similar in actual model [11]. The reason for gas outlet was to avoid back flow of gas. The back flow pressure extends the convergence time of simulation [22].

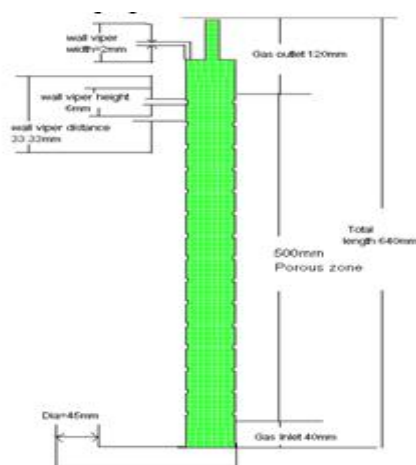


Fig. 3 Computational Domain and Grid.

The following boundary type was assigned (table 1) in gambit before domain export as a meshed file.

Table 1. Boundary types assigned in Gambit

Gambit	Real	Type
Inflow	Gas inlet	
Outflow	Gas outlet	OUTFLOW
Fluid	Porous medium	Continuum

V. RESIDENCE TIME DISTRIBUTION SIMULATION

Post processing was used for the analysis of Residence Time Distribution RTD of Bales packing in dry condition. To compare experimental results with CFD, simulation run on similar experimental Reynolds numbers given in Table 2.

Table 2. Reynold numbers used in RTD simulation

Reynold numbers	Velocity (m/sec)
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20	7e-3
56	0.02
92	0.033
129	0.045

For RTD analysis, only power law porous media model was employed, the values set for power law coefficients are $C_0 = 64000$ and $C_1 = 1.7077$. In order to compute mean and local values, standard k- ϵ turbulent model was chosen because fluctuations of mean quantities are only calculated by using turbulent models. The k- ϵ turbulent model parameters are computed from Table 2 Reynolds numbers, and are shown in Table 3.

Table 3. k- ϵ Turbulent model variables for RTD simulations

Turbulence Intensity	Turbulence Kinetic energy m^2/s^2	Turbulence Dissipation m^2/s^3
0.11	8.89e-7	1.91e-9
0.096	5.6e-7	3.0e-8
0.90	1.35e-5	1.13e-7
0.085	2.2e-5	2.4e-7

After the solution was converged, the UDS set to 1 from UDS panel. The other parameters were specified as follows.

- Mass flow rate was chosen in flux function panel.
- Unsteady function was specified.
- In materials define panels diffusion coefficients was set to 1×10^{-10} kg/m-s for nitrogen.
- Nitrogen was selected instead of Air in boundary condition panel.
- Specified value was selected in User define scalar boundary condition in gas inlet, gas outlet and walls.
- The region was adapted just on velocity Inlet, the adapted values (-1,-0.038) and (1, -0.0388).
- The tracer monitor surface was created using line option.
- Define surface monitor panel was turned on and the Area-Weighted Average under type was chosen.
- The solution was turned off except the UDS value.
- The iteration run on different time steps for better accuracy.
- The saved file was opened in spreadsheet software *Microsoft Excel*® 2001 for further RTD calculations.

VI. RESULTS AND DISCUSSION

The flowing tracer was taken at six different times of about 90 sec as shows in Figure 4. User defined scalar technique was employed for RTD determination. The mean residence times predicted by this method are compared in Figure 5. The curve shows that predicted mean residence time was higher than the experimental obtained residence times. It can also be noticed that at higher gas Reynolds numbers, the relationship between the CFD and the experimental curves is linear but this difference increases as Reynolds number decreases. The prediction of the RTD depends on modelling of both convective and diffusive processes as per dissipation rate calculated by equation 5, therefore the solution is sensitive to the choice of diffusion coefficient. However, in the present work, the diffusion coefficient was estimated roughly, and the prediction of mean residence time could therefore be improved by a more accurate value of the diffusion coefficient. In addition, the convective term in the scalar transport equation depends on the accurate estimate of the velocity field, which is a function of domain geometry. Since this was a two-dimension simulation and macro scale geometry of Bales packing was not taken into account, so in this respect further improvement may also be necessary.

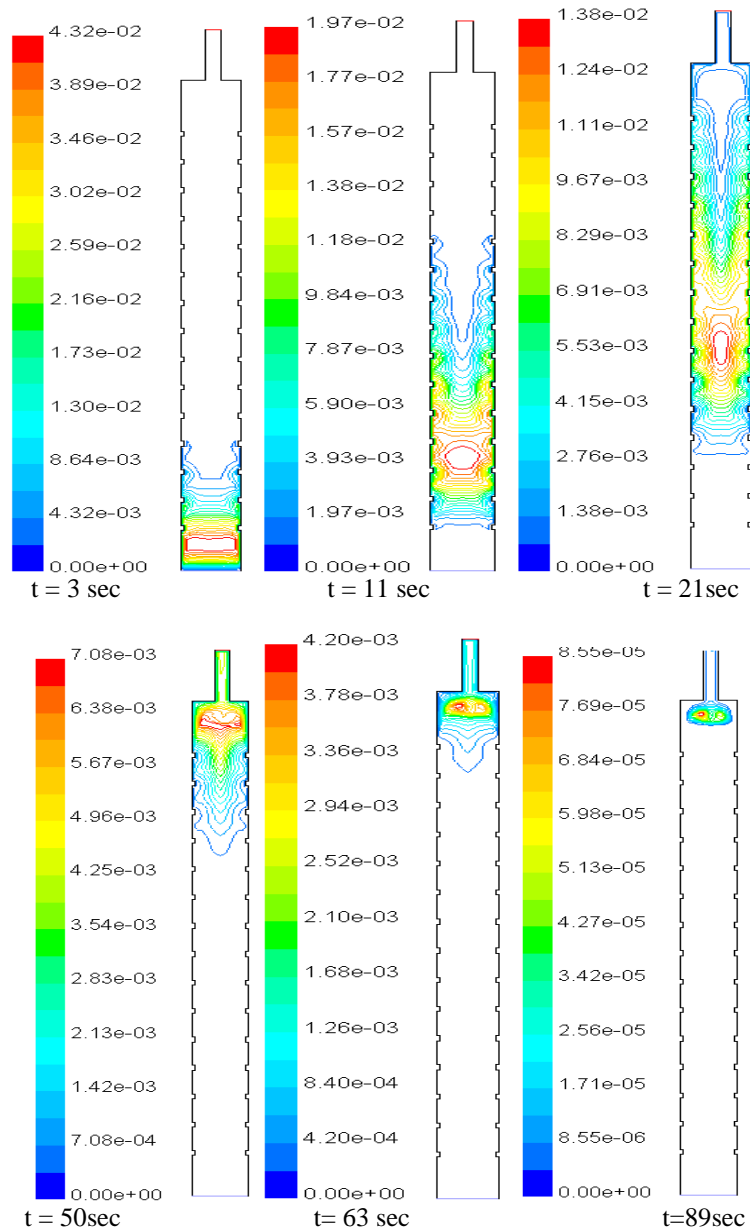


Fig. 4 Gas tracer distribution contours at $Re_g = 129$

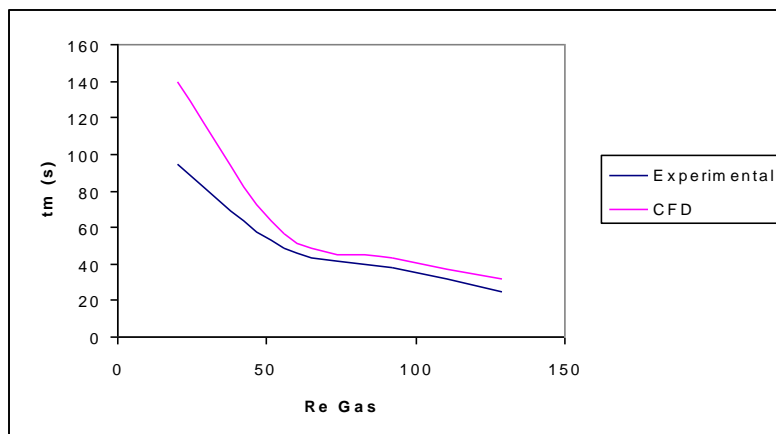


Fig. 5 Mean Residence time comparison of experimental and Predicted CFD data.

The figure 6 shows the exit age distribution curves at different Reynolds numbers. Qualitatively, CFD gives a similar experimental curve [11] with sharp peaks, which indicate the plug flow behavior. As it can be seen from the curves, the peaks shift towards the shorter times, which is a common and expected response.

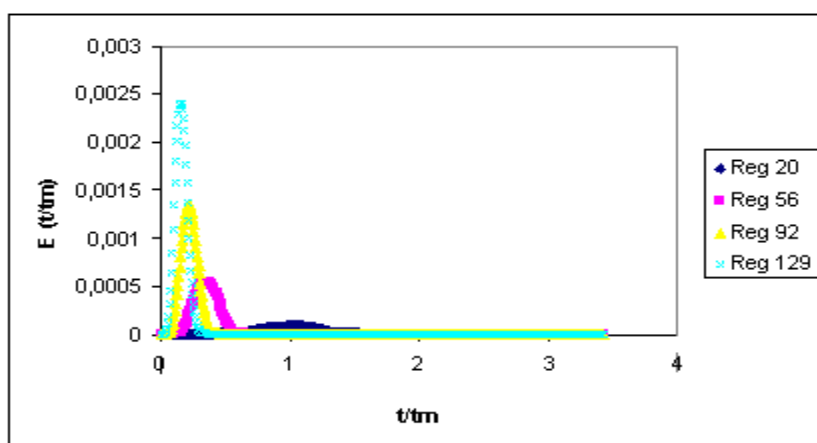


Fig. 6 Exit age distributions as functions of different gas Reynolds numbers

Moreover, the exit age distribution also shows that the peak decrease as Reynolds number increases which testify the short circuit inside reactor. The Reactor Peclet Number calculated from equation 13 using CFD RTD data is shown in figure 7.

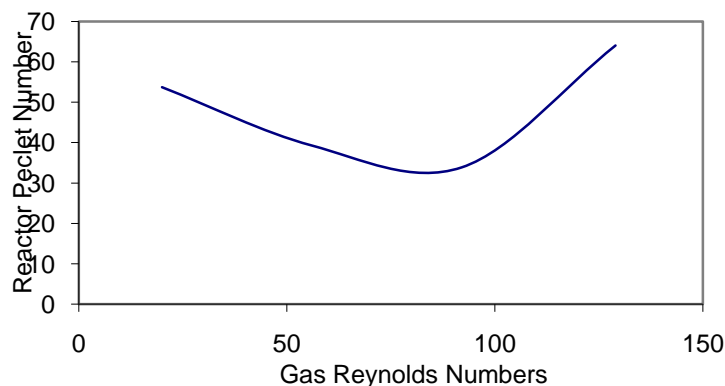


Fig. 7 The gas Reynolds numbers Vs Peclet Numbers.

VII. CONCLUSION

This study shows that a commercial CFD (FLUENT) code can be used to predict the aerodynamics of a catalytic Bale packing in terms of residence time distribution parameters and internal (wall wiper effects) velocity profiles. A user defined Scalar technique for RTD determination was presented to take into account the porosity of the catalytic bales. The technique was successful in simulating the exit age or E-curves at different Reynolds numbers. The data predicted by E-curves was used in the determination of mean residence times, and which was compared with the experimental results. This comparison with the experimental results indicates a satisfactory agreement and the values obtained were relatively close, despite a two-dimensional geometry used in the CFD. It is anticipated that an improved and better agreement will result by using actual geometry (3-D) and obtaining an experimental data for the diffusivity. The CFD simulation results presents detailed aerodynamics investigation of bench scale reactor plugged with catalytic Bales. The CFD is an efficient diagnostic tool for such type of studies. However, due to the limited availability of the literature on flow dynamics CFD studies dealing with catalytic Bale packing, this conclusion requires further verification.

Nomenclature

C	Concentration of tracer (-)
C_0	Empirical coefficient (pa/m)
C_1	Empirical coefficient (-)
C_2	Inertial resistance coefficients, (1/m)

D	Diffusivity (m ² /s)
E	Exit gas distribution
G	Gravity acceleration, m/s ²
E	Turbulence dissipation rate
I	Turbulence intensity
k	Turbulent kinetic energy, m ² /s ²
L	Length of bales, m
P	Pressure, Pascal
Pe _r	Peclet number
Re _g	Gas Reynolds number, (UL/Da)
t _m	Mean residence time, s
U	Gas superficial velocity, m/s

Greek- letter

α	Permeability, m ²
ε	Porosity, (-)
μ	Viscosity, kg/ms
θ	Time (dimensionless)
σ^2	Second moment variance

Abbreviations

CAD	Computer Aided Design
CB	Catalytic Bales
CCFBR	Counter Current Fixed Bed Reactor
CD	Catalytic distillation
CFD	Computational Fluid Dynamics
FLUENT	A modeling software
RTD	Residence time distribution
TBA	Tertiary Butyl Alcohol
UDS	User defined scalar

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