

CFD MODELING OF BROWN COAL LIQUEFACTION REACTOR

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ABSTRACT

Direct coal liquefaction is the direct conversion process of a solid coal into a liquid under high pressure and temperature. Under this condition, hydrogen and catalyst must be added to obtain the fuel and to improve the efficiency of the process. Experimentally, direct liquefaction of brown coal was carried out on 1 liter autoclave with limonite catalyst and heavy oil as a solvent. The operating condition was 450°C and 12 MPa, residence time of 0-60 minutes. Detailed investigation of the flow pattern in the reactor is very important for the process optimization. This study presents two-dimensional transient hydrodynamic and reaction modeling of Brown Coal Liquefaction (BCL) reactor using CFD. The model demonstrated the capability of commercial CFD code fluent 6.3 to describe the flow pattern in the reactor. The model predicted the distribution of yield, temperature, and velocity vector in the reactor.

Keywords: BCL, CFD, Multiphase Flow, Reaction

INTRODUCTION

The requirement of worldwide oil has increased every year. It was estimated that the crude oil reserves will tighten. One of the solutions to overcome this problem is to look for the other alternative of petroleum substitution. The source of alternative energy of oil substitution can be found in coal. One of the countries which have fairly large quantities of coal reserves is Indonesia. Coal reserve of Indonesia is about 36.3 million ton and 85 % of the reserve in the form of low rank coal (lignite). The utilization of coal reserve can be conducted by using the technology of coal liquefaction.

The Brown Coal Liquefaction (BCL) technology was developed to suit the characteristics of the Indonesian low rank coal. BCL is the process to convert the brown coal to the synthetic oil in order to replace the natural sources of petroleum. It is the advantage approach for the clean and effective utilization of coal. The BCL technology has been studied experimentally by numerous research groups (Hartiniati, 2000, Komatsu, 2000, Narita et al., 2000, Okuyama et al., 2000; Silalahi, L. H. and Yusnitati,

2000) using batch reactor. However, there is not much research conducted in modeling of BCL technology. Meanwhile, a mathematical model describing the underlying physics helps in understanding the behavior of the BCL process especially for optimizing the reactor design and process conditions. Fluent 6.3 CFD code can be used to model the BCL reactor. Fluent provides some information about the flow field of concentration, temperature, pressure and velocity in the region where the experimental is not able to be done. At present, Fluent 6.3 can perform the hydrodynamics and reaction kinetics model to the higher level of reliability.

Computational fluid dynamics (CFD) is a powerful design tool for engineers and scientists. Nowadays, CFD has been applied by several researchers (Benyahnia, Arastoopour et al. 2002, Gao, Xu et al. 1999, Van Wachem, Schoutten et al. (1999)) to solve solid-fluid system. CFD can be used confidently to solve a single phase flow problems. On the other hand, CFD modeling of multiphase flow is still complicated due to many interactions involved. However, a considerable research effort still has to be needed in order to provide the valuable information especially in the Brown Coal Liquefaction process. Computational Fluid dynamics (CFD) presents the solving of numeric equations of mass conservation, momentum, energy and additional equations such as turbulence and reactions. These equations cannot be solved analytically. The equations can be solved by finite volume method numerically.

Two most common approaches to model multiphase flows are the Euler-Euler approach and the Euler-Lagrange approach. In the Euler-Euler approach, both the continuous and dispersed phases are considered in an Euler representation, which creates mass and momentum equations for each phase. The presence of each phase is described by a volume fraction. In the alternative Euler-Lagrange approach, the mixture is treated as a pseudo-continuous phase with variable density. However, the Lagrange approach restricts the number of particles. The Euler-Lagrange approach is more computationally intensive than the Euler-Euler approach, because it allows the tracking of particles (bubbles) in the continuous phase. Recently, the Euler approach has been most often used and is preferable when considering fluid-solids flow systems.

Kinetics of coal liquefaction is very complex due to the formation of many compounds. Weller et al. (1950) developed a model for the conversion of coal to oil passes through the formation of asphaltenes, which is still accepted today. Five reaction mechanisms based on a series of irreversible and pseudo first-order reactions through the liquefaction investigation of six Turkish coals with microwave heating were modeled by the other researcher (Simsek et al., 2001). Their result showed that the best fit between experimental and model value was the model which suggested the oils, asphaltenes, and preasphaltenes were formed directly from coal. Li et al (2008) proposed a model for heating-up and isothermal stages of the coal liquefaction kinetics to estimate the rate constants of both stages. Their result showed that the rate-controlled process of coal liquefaction is the reaction of preasphaltene plus asphaltene to oil plus gas.

Detailed hydrodynamic and reaction kinetics is very important to provide an advanced understanding of the phenomena occurring in BCL reactors. This research has used CFD to simulate two-dimensional transient hydrodynamics and kinetics model of BCL reactor. The model demonstrated the capability of commercial CFD code Fluent 6.3 to

describe the flow pattern in the reactor. The model predicted the distribution of yield, temperature and velocity inside the reactor.

CFD MODELING

The governing conservation equations of fluid flow represent the statement of the conservation laws of mass, momentum and energy. For reacting flows, the chemical species transport and mixing can be estimated by using species-transport equations.

Hydrodynamics Model

The Conservation Equations

The mass conservation of phase i ($i = \text{fluid, solid}$) (Fluent, 2006):

$$\frac{\partial}{\partial t}(\rho_i \varepsilon_i) + \nabla \cdot (\rho_i \varepsilon_i \mathbf{U}_i) = r_i \quad (1)$$

With definition: $\varepsilon_f + \varepsilon_s = 1$

The conservation of momentum of phase i ($i = \text{fluid, } k = \text{solid, } k \neq i$) can be written as (Fluent, 2006):

$$\frac{\partial}{\partial t}(\rho_i \varepsilon_i \mathbf{U}_i) + \nabla \cdot (\rho_i \varepsilon_i \mathbf{U}_i \mathbf{U}_i) = -\varepsilon_i \nabla P + \nabla \cdot \boldsymbol{\tau}_i + \rho_i \varepsilon_i \mathbf{g} - \beta(\mathbf{U}_i - \mathbf{U}_k) \quad (2)$$

The conservation of energy for phase i yield (Fluent, 2006):

$$\frac{\partial}{\partial t}(\varepsilon_i \rho_i H_i) + \nabla \cdot (\varepsilon_i \rho_i \mathbf{U}_i H_i) = -\varepsilon_i \frac{\partial P_i}{\partial t} + \boldsymbol{\tau}_i : \nabla \mathbf{U}_i - \nabla \cdot \mathbf{q}_i + S_i \quad (3)$$

$$\text{Where: } H_i = \sum_i \varepsilon_i \int_{T_{ref}}^T C_{p_i} dT \quad (4)$$

Additional Equations

Interphase Exchange Coefficients:

From the Syamlal-O'Brian model for the drag force formulation (Fluent, 2006):

$$\beta = \frac{3}{4} C_D \frac{\varepsilon_s \varepsilon_g}{v_{r,s}^2} \frac{\rho_g}{d_s} \left(\frac{\text{Re}_s}{v_{r,s}} \right) |\mathbf{U}_s - \mathbf{U}_g| \quad (5)$$

The drag coefficient, C_D is given by:

$$C_D = \left(0.63 + \frac{4.8}{\sqrt{\text{Re}_s / v_{r,s}}} \right)^2 \quad (6)$$

$$\text{Re}_s = \frac{\rho_g d_s |\mathbf{U}_s - \mathbf{U}_g|}{\mu_g} \quad (7)$$

where $v_{r,s}$ is the terminal velocity correlation for the solid phase:

$$v_{r,s} = 0.5A - 0.03\text{Re}_s + 0.5\sqrt{(0.06\text{Re}_s)^2 + 0.12\text{Re}_s(2B - A) + A^2} \quad (8)$$

where:

$$A = \varepsilon_g^{4.14} \quad (9)$$

$$B = 0.8 \varepsilon_g^{1.28} \text{ for } \varepsilon_g \leq 0.85 \quad (10)$$

$$B = 0.8 \varepsilon_g^{2.65} \text{ for } \varepsilon_g > 0.85 \quad (11)$$

Solids Pressure

The solids phase pressure (P_s) consists of a kinetic term and the particle collisions term. It is calculated from the equation of state which is the same as the van der Waals equation of state for gases (Chapman and Cowling, 1970):

$$\begin{aligned} P_s &= (1 + 2(1 + e_s)\varepsilon_s g_o)\varepsilon_s \rho_s \Theta_s \\ &= \rho_s \varepsilon_s \Theta_s + 2g_o \rho_s \varepsilon_s^2 \Theta_s (1 + e_s) \end{aligned} \quad (12)$$

Where Θ_s is the granular temperature. e_s is the coefficient of restitution for particle collisions. g_o , the radial distribution function (Sinclair and Jackson, 1989) is given by:

$$g_o = \left[1 - \left(\frac{\varepsilon_s}{\varepsilon_{s,\max}} \right)^{\frac{1}{3}} \right]^{-1} \quad (13)$$

The value of maximum solid packing, $\varepsilon_{s,\max}$ for this simulation is 0.6.

Solids Shear Stress

The solids stress tensor contains bulk and shear viscosities. The solid phase bulk viscosity can be expressed as:

$$\mu_b = \frac{4}{3} \varepsilon_s \rho_s d_s g_o (1 + e_s) \left(\frac{\Theta_s}{\pi} \right)^{1/2} \quad (14)$$

The solids phase shear viscosity is given by:

$$\begin{aligned} \mu_s &= \frac{2\mu_{s,dil}}{(1+e)g_o} \left[1 + \frac{4}{5}(1+e_s)g_o\varepsilon_s \right]^2 \\ &+ \frac{4}{5} \varepsilon_s \rho_s d_s g_o (1 + e_s) \left(\frac{\Theta_s}{\pi} \right)^{1/2} \end{aligned} \quad (15)$$

The solid phase dilute viscosity is:

$$\mu_{s,dil} = \frac{5}{16} \rho_s \varepsilon_s l_s \sqrt{2\pi\Theta_s} \quad (16)$$

$$\text{Where is } l_s = \frac{\sqrt{2} d_s}{12 \varepsilon_s} \quad (17)$$

Granular Temperature

The granular temperature, Θ_s , is calculated by solving the turbulent kinetic energy equation for solid phase:

$$\frac{3}{2} \frac{\partial}{\partial t} (\rho_s \varepsilon_s \Theta_s) + \nabla \cdot (\rho_s \varepsilon_s \mathbf{U}_s \Theta_s) = T_s : \nabla \mathbf{U}_s + \nabla \cdot (k_{\Theta_s} \nabla \Theta_s) - \gamma_s \quad (18)$$

The diffusion coefficient for granular energy k_{Θ_s} is represented by:

$$k_{\Theta_s} = \frac{2k_{\Theta_s,dil}}{(1+e_s)g_o} \left(1 + \frac{6}{5}(1+e_s)g_o \varepsilon_s \right)^2 + 2\varepsilon_s^2 \rho_s d_s g_o (1+e_s) \left(\frac{\Theta_s}{\pi} \right)^{1/2} \quad (19)$$

where:

$$k_{\Theta_s,dil} = \frac{75}{64} \rho_s \varepsilon_s l_s \sqrt{2\pi\Theta_s} \quad (20)$$

The collisional energy dissipation, γ_s , is given by:

$$\gamma_s = 3 \left(1 - e_s^2 \right) \varepsilon_s^2 \rho_s g_o \Theta_s \left[\frac{4}{d_s} \left(\frac{\Theta_s}{\pi} \right)^{1/2} - \nabla \mathbf{U}_s \right] \quad (21)$$

k-ε Turbulence Models

Generally, the FCC riser reactor is under turbulent flow conditions. Therefore, it is important to use an appropriate turbulence model to describe the effect of turbulent fluctuations of velocities and scalar variables for the basic conservation equations. A $k-\varepsilon$ model was used to describe the turbulent motions in both phases. In the $k-\varepsilon$ model, the turbulent viscosity is defined as:

$$\mu_{t,i}^{(t)} = \rho_i \varepsilon_i C_\mu \frac{k_i^2}{\varepsilon_i} \quad (22)$$

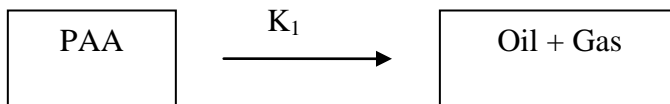
The turbulence kinetic energy, k , and its rate of dissipation, ε , can be calculated from the following transport equations:

$$\frac{\partial}{\partial t} (\rho_i \varepsilon_i k_i) + \nabla \cdot (\rho_i \varepsilon_i k_i \mathbf{U}_i) = \nabla \cdot \left(\varepsilon_i \frac{\mu_t}{\sigma_k} \nabla k_i \right) + (\varepsilon_i G_k - \varepsilon_i \rho_i \varepsilon_i) \quad (23)$$

$$\frac{\partial}{\partial t} (\varepsilon_i \rho_i \varepsilon_i) + \nabla \cdot (\rho_i \varepsilon_i \varepsilon_i \mathbf{U}_i) = \nabla \cdot \left(\varepsilon_i \frac{\mu_t}{\sigma_k} \nabla \varepsilon_i \right) + \frac{\varepsilon_i}{k} (C_{1\varepsilon} \varepsilon_i G_k - C_{2\varepsilon} \varepsilon_i \rho_i \varepsilon_i) \quad (24)$$

Reaction Kinetics Model

Based on the model purposed by Li et al (2008) that the rate-controlled process of coal liquefaction is the reaction of preasphaltene plus asphaltene to oil plus gas, the scheme of the reaction in this model is assumed as below:



Where: PAA is representative of preasphaltene plus asphaltene. It is assumed the irreversible and pseudo first-order reactions. The reaction kinetics is enclosed to the hydrodynamic model by solving the species equation of the components in the form of the reaction rates as following:

$$\frac{dA}{dt} = -K_1 A \phi \quad (25)$$

Table 1: Experimental Data of Yield Distribution at temperature of 450°C, pressure of 10 Mpa.

Temperature	450 C
DISTILATE	40.53
H2O	13.42
CLB	38.57
CO+CO2	6.7
C1-C4	7.94
H2 cons.	-7.16

Numerical Procedure

The geometry of the reactor is described in Figure 1. It was constructed by GAMBIT which is the pre-processor of FLUENT 6.3. The numerical calculations were carried out on a two-dimensional mesh of 1545 cells. The reactor had a diameter of 0.075 metre and the height of 0.26 metre. In order to reduce the computational effort, and because the reactor is symmetric, the reactor was split to become a half part.

Initially, the coal slurry, catalyst and heavy-oil solvent were settled at the bottom of the reactor to a level above the impeller, then hydrogen was injected to the rest of the reactor. The volume fraction of coal in the slurry entered to the reactor of 30 %, with the average diameter of 75 μm . The operating conditions of reactor were the temperature of 450°C and the pressure of 12 MPa. A schematic of the reactor and the initial position of slurry solution are shown in figure 1. Within the domain, there are three fluid zones, representing the impeller zone (blue colours), the region where the slurry is initially located (grey colours) and the rest of tank where hydrogen is filled (green colours). The calculations were started using the initial assumptions similar to the specified volume fractions. The hydrogenising and cracking process was simulated over 60 minutes. A constant time step of 0.01 was used. The simplification assumptions are summarized below:

- Each phase has been described as a continuum
- Coal entered the reactor in the form of coal slurry
- The reaction is the irreversible and pseudo first-order.
- The calculation starts at the reactor condition of isothermal achieved.

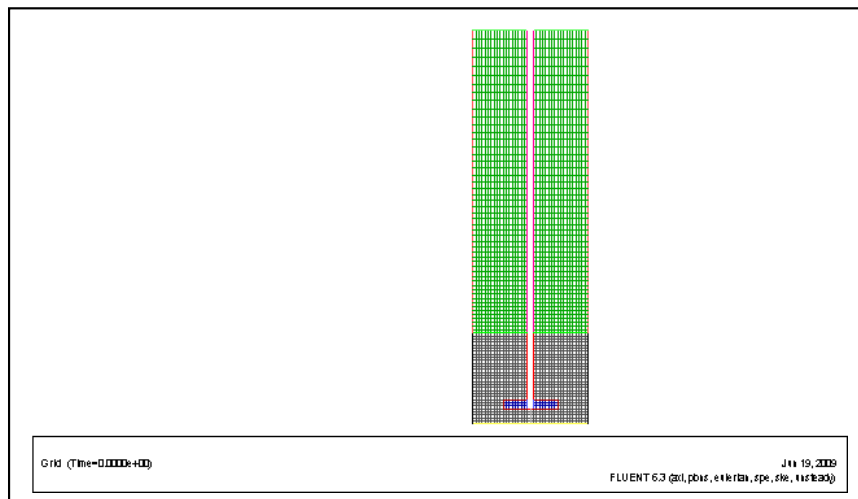


Fig. 1: Two-dimensional geometry of BCL reactor.

RESULTS AND DISCUSSION

Velocity Vector of Coal

The velocity vector of coal is described in Figure 2. We can see from this figure that the flow in the reactor shows an axial-radial flow pattern. It is shown that the highest velocity of coal can be found near the impeller which is about 0.2 m/s. The coal flows to the impeller toward the wall and divides into two flows under the influence of the reactor wall. Several coals flow downward along the wall to the bottom of the reactor, and then back to the impeller zone. The other coals rise upward along the centre of the reactor to the top of the reactor and back to the impeller region forming the symmetric double loop. The swirls can not reach the top of the reactor because the position of impeller and the initial coal slurry were settled just at the lower part of the reactor.

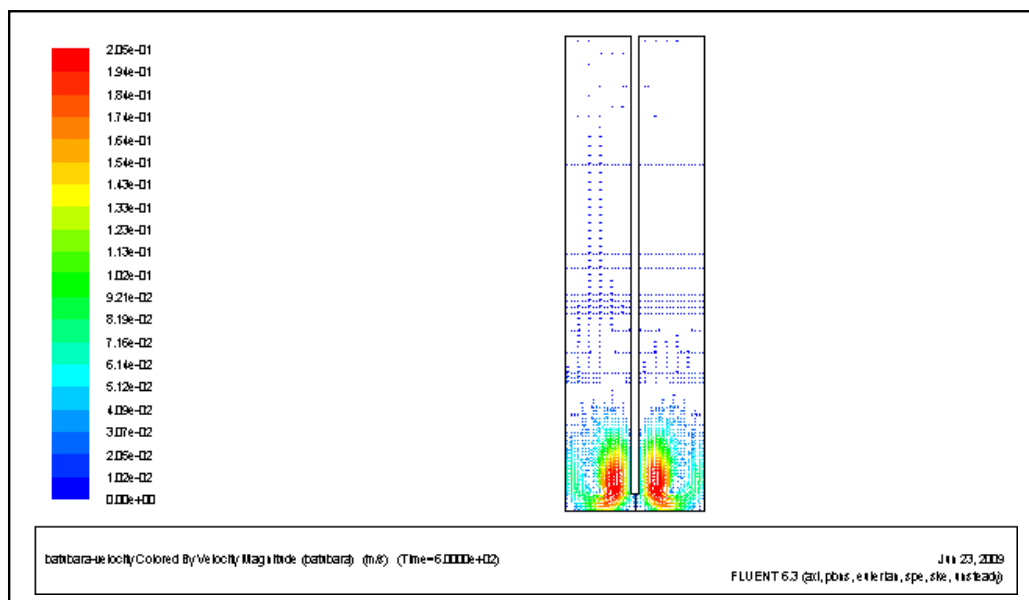


Fig. 2: Velocity vector of Coal Phase at Simulation Times of 600 seconds (10 minutes).

Yield Distribution of Distillate and CLB

Figures 3 and 4 illustrate the yield distribution of distillate and Crude Liquid Bottom (CLB) products along the reactor height for different reaction times. It is shown, initially the yield of products increased along the reactor. However it is constant at the times of four minutes. We also can see the yield decreased with the reaction times. This is due to the distillate and CLB products have not yet cracked to the lower molecular-weight hydrocarbon. Over 1 hour reaction times, the yield of distillate product is constant and the maximum yield of 37.9 % obtained. It is lower compared to the experimental result in table 1, which is about 40.53%. The maximum yield of CLB product is about 48.6 %, while the experimental CLB yield is 38.57 %. It is higher than experimental data. These results show a reasonable agreement with experiments.

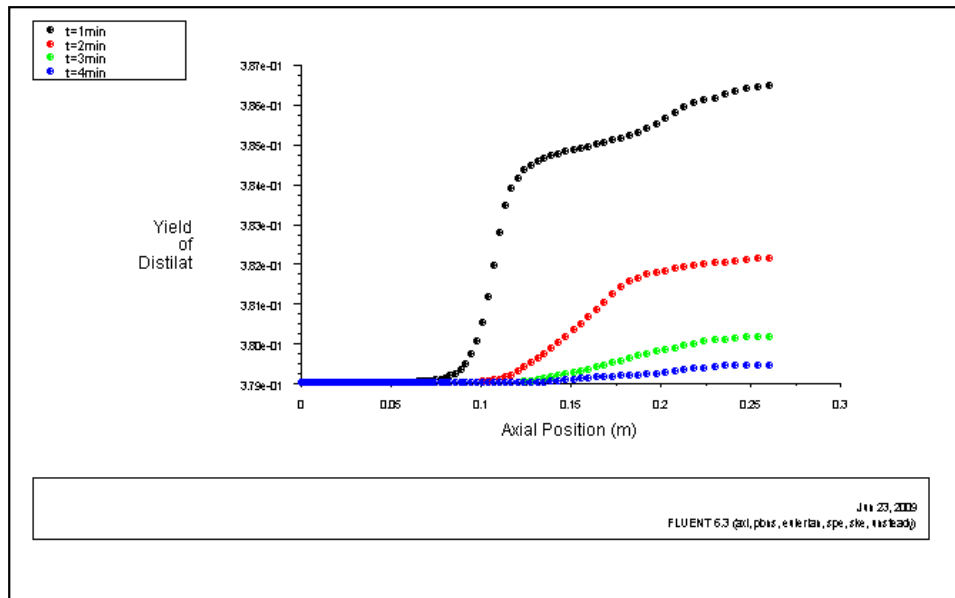


Fig. 3: Yield Distribution of Distillate at Different Reaction Times.

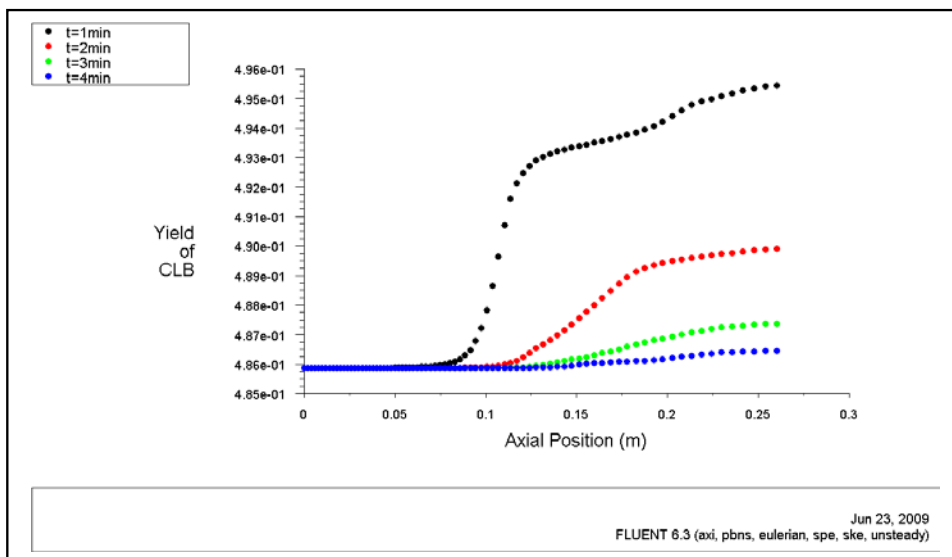


Fig. 4: Yield Distribution of Crude Liquid Bottom (CLB) at Different Reaction Times.

Temperature Profile of Hydrogen

In Figure 4 shows the temperature profiles of hydrogen phase at the simulation time of 10 minute. Below the reactor height of 0.20 metre, the temperature of hydrogen was constant of about 450°C (723 K). This is due to the perfect mixing occurring at the reactor bottom because the effect of impeller. Therefore, the assumption of isothermal reactor is nearly accurate. However beyond 0.2 metre of the reactor height, the temperature increased significantly. This is because of the increasing amount of gasses at the reactor top due to hydrogenising and cracking reaction. It can be seen that the maximum temperature occurred at the top of the reactor.

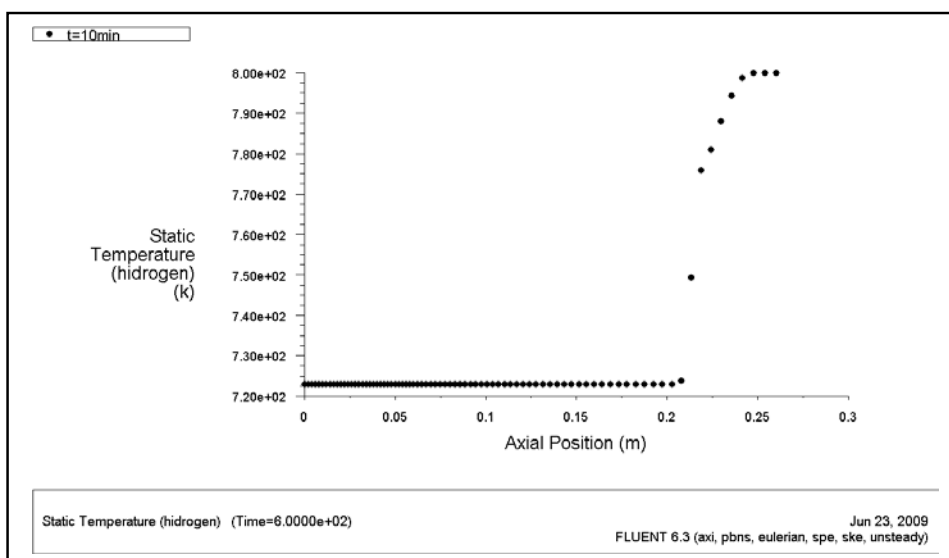


Fig. 4: Axial Temperature Profile of Hydrogen Phase at Simulation Times of 600 s (10 minutes).

CONCLUSION

The commercial code of Fluent 6.3 can be used to simulate two-dimensional transient hydrodynamics and kinetics model of BCL reactor. The result of CFD simulations shows reasonable agreement with experiment. As expected, that the flow in the reactor is an axial-radial flow pattern. The apparent symmetric double loop flow pattern is found in the reactor. The perfect mixing occurred at the reactor bottom because of the effect of impeller. The assumption of isothermal reactor is nearly accurate.

ACKNOWLEDGEMENT

The author wishes to thank for the financial support from Government of Province of South Sumatera and Ministry of Research and Technology of Republic of Indonesia in RUSNAS PEBT Project, the experimental data provided by BPPT.

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- Refinery Technology
- Catalytic Cracking, Hydrocracking
- Brown Coal Liquefaction Technology