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CFD SIMULATION OF BCL PROCESS: EFFECT OF TEMPERATURE ON THE YIELD DISTRIBUTION OF PRODUCT

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ABSTRACT

Indonesian has 61.274 milliar tons of coal which 70% of this are brown coal (lignite). The huge number of coal can be used to cover oil consumptions that increase every year. One of the processes to utilize of these coals is The Brown Coal Liquefaction (BCL) Technology.

Brown Coal Liquefaction (BCL) Technology is the process to convert brown coal (lignite) with high-molecular-weight hydrocarbons to valuable liquid products such as light-oil, middle-oil, and heavy-oil. It is carried out experimentally in one liter stirrer autoclave under high operating conditions (Temperature: 430 – 450^{o}C; Initial hydrogen pressure: 8 – 12 MPa).

Initially, coal slurry, solvent, limonite catalyst, and gas hydrogen were injected to the reactor. After appropriate operating conditions, the complex cracking and hydrogenation reactions were took place during 60 minutes of residence time.

For the description of multiphase and reaction occurring throughout the BLC process, a commercial computational fluid dynamics (CFD) code Fluent 6.3 modeling technique was used. The Eulerian-Eulerian modeling approach was applied to predict fluid-solid flow behavior. The results of this simulation showed a good prediction of the yield distillate profile under different temperature. Therefore, the optimized operating condition was obtained.

Keywords: BCL Technology, CFD, cracking, hydrogenation.

1. INTRODUCTION

Petroleum oil requirements have increased yearly for industries, transportations, and households. This caused the decreasing of crude oil reserves. The estimated life of 20 years of world oil reserves and the reality of increased oil consumptions have rendered coal liquefaction as an alternative process to substitute petroleum oil.

Coal is the most abundant fossil fuel known in the world, and also a vital global energy source. Indonesia has 61.274 milliar tons of coal which 70% of these are brown coal (lignite) that are not valuable. To increase the coal's value, a process that convert coal into liquid was obtained, called Brown Coal Liquefaction (BCL) Technology.

BCL is a catalytic cracking and hydrogenating processes for the conversion of coal with high molecular weight compounds into more valuable lower boiling liquid products, such as light oil, middle oil, and heavy oil, in hydrogen-rich atmosphere at elevated temperature and pressure. The cracking reactions are slightly endothermic while the hydrogenating reactions are highly exothermic, making the overall hydrocracking process highly exothermic in nature.

Even though considerable amount of research has been reported on BCL Technology along experimental batch reactor (Hartiniati, 2000; Komatsu, 2000; Narita et.al., 2000; Okuyama et.al, 2000; Silalahi, L.H., and Yustiniati, 2000), but very few computational fluid dynamics (CFD)

models on the optimization of BCL process are found in open literature. Meanwhile, BCL involves a complex combination of physical and chemical process. Therefore, mathematical modelling to describe fluid-solid flow behaviour is needed to understand the process, especially for optimization and reactor design.

Weller et.al. (1950) were developed a coal conversion model through formation of asphaltene. Until now, this model can be accepted. The other researchers, Li et.al. (2008) were developed kinetic of coal liquefaction during heating-up and isothermal stages. From their description, rate of reaction preasphaltene-asphaltene into oil and gas is a controller in coal liquefaction process.

Detailed knowledge of hydrodynamics in multiphase BCL reactors is presently devoted to the application of computational fluid dynamics (CFD). CFD is a powerful modelling and simulation tools for analyzing fluid-solid flow behaviour and reaction engineering with complex geometries. Information about distribution concentration, velocity, pressure and temperature can be obtained by using CFD. Besides that, it also can be used to solve single-phase, multiphase and complex reaction problems.

In this work, we have used FLUENT 6.3 (CFD packages) to simulate multiphase fluid-solid flow behaviours in autoclave's BCL based on Eulerian-Eulerian approach. This simulation showed a good prediction of yield distillate profile under different temperature. Therefore, optimized operating conditions were obtained.

2. THEORY

Considering a two-dimensional axis-symmetric domain, set of model equations below was solved using commercial software FLUENT 6.3 along with meshing software GAMBIT 2.0. The time average continuity and momentum equations for each phase i (fluid, solid) are reported below:

Mass continuity phase *i* (*i* = fluid, solid):

$$\frac{\partial}{\partial t}\left(\rho_i \varepsilon_i\right) + \nabla.\left(\rho_i \varepsilon_i \mathbf{U}_i\right) = r_i \tag{1}$$

where: $\varepsilon_f + \varepsilon_S = 1$

Energy continuity phase *i* (*i* = fluid, solid):

$$\frac{\partial}{\partial t}\left(\varepsilon_i \rho_i H_i\right) + \nabla.\left(\varepsilon_i \rho_i \mathbf{U}_i H_i\right) = -\varepsilon_i \frac{\partial P_i}{\partial t} + \boldsymbol{\tau}_i : \nabla \mathbf{U}_i - \nabla.\mathbf{q}_i + S_i \tag{2}$$

where: $H_i = \sum_i \varepsilon_i \int_{Tref}^{T} Cp_i dT$ (3)

Conservation of momentum phase *i* (*i* = fluid, *k* = solid, *k* ≠ *i*):

$$\frac{\partial}{\partial t}\left(\rho_i \varepsilon_i \mathbf{U}_i\right) + \nabla.\left(\rho_i \varepsilon_i \mathbf{U}_i \mathbf{U}_i\right) = -\varepsilon_i \nabla P + \nabla.\boldsymbol{\tau}_i + \rho_i \varepsilon_i \mathbf{g} - \beta(\mathbf{U}_i - \mathbf{U}_k) \tag{4}$$

Turbulence has been modelled by k-$\in$ Turbulence Models, which turbulence viscosity has defined by:

$$\mu_{t,i}^{(t)} = \rho_i \varepsilon_i C_\mu \frac{k_i^2}{\in_i} \tag{5}$$

Where k indicates turbulence kinetic energy and $\in$ indicates rate of dissipation, these are calculated with equations below:

$$\frac{\partial}{\partial t}(\rho_i \varepsilon_i k_i) + \nabla.(\rho_i \varepsilon_i k_i U_i) = \nabla.\left(\varepsilon_i \frac{\mu_t}{\sigma_k} \nabla k_i \right) + (\varepsilon_i G_k - \varepsilon_i \rho_i \in_i) \quad (6)$$

$$\frac{\partial}{\partial t}(\varepsilon_i \rho_i \in_i) + \nabla.(\rho_i \varepsilon_i \in_i U_i) = \nabla.\left(\varepsilon_i \frac{\mu_t}{\sigma_k} \nabla \in_i \right) + \frac{\in_i}{k}(C_{1\in} \varepsilon_i G_k - C_{2\in} \varepsilon_i \rho_i \in_i) \quad (7)$$

In this work, kinetic model of coal liquefaction is obtained based on Li et.al. (2008) research, where rate of reaction is controlled by reaction preasphaltene-asphaltene (PAA) into oil and gas (O+G). The scheme is in the figure below:

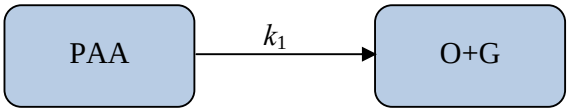

Figure 1. Scheme of kinetic model

This kinetic reaction equation is entered to hydrodynamics model with component rate reaction is:

$$\frac{dA}{dt} = -K_1 A \phi \quad (8)$$

3. EXPERIMENTAL SET-UP AND PROCEDURE OF CFD

The elegibility of Musi Banyuasin coal as a feedstock for brown coal liquefaction process was conducted by using reactor Autoclave 1 litre at the center of research of Coal Liquefaction Laboratory, BPPT Serpong. This Laboratory own a set equipments to test the liquefaction with the autoclave reactors and also the examination equipments of coal liquefaction product such as gas product with the gas chromatography, product liquid by vacuum distilation, and the residue product by solvent extraction. This laboratory is also equipped with the equipments of catalyst crushing which can attenuate the catalyst size of below 1 micronmetre.

Initially, the coal entered into the reactor of one litre together with solvent and catalyst at amount which have been determined. Then the reactor closed and filled with the hydrogen gas and arrested during two hours to know the leakage (leak test). After the leak test, it has been heated until the appropriate operating conditions (Temperature = 430 – 450 ^{o}K, and initial hydrogen pressure = 8 – 12 MPa). The complex cracking and hydrogenating reactions were took place during 60 minutes of residence time. The volume fraction of coal in the slurry entered to the reactor of 30 %, with the average diameter of 75 μm.

Gas products were taken and analysed by GC-FID and GC TCD to know its composition. Meanwhile product which is in the form of slurry were collected and sent to distillation column to separate the light oil fraction (LO), middle oil (MO) and heavy oil (HO).

Figure 2 shows the reactor geometry by using GAMBIT 2.0 as pre-processor in FLUENT 6.3, where green zone indicates mix of coal slurry, limonite catalyst, and heavy oil solvent; gray zone indicates hydrogen gas; and blue zone indicates the stirrer. The numerical calculations were carried out on a two-dimensional mesh of 1545 cells. The reactor had a diameter of 0.076 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. 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.

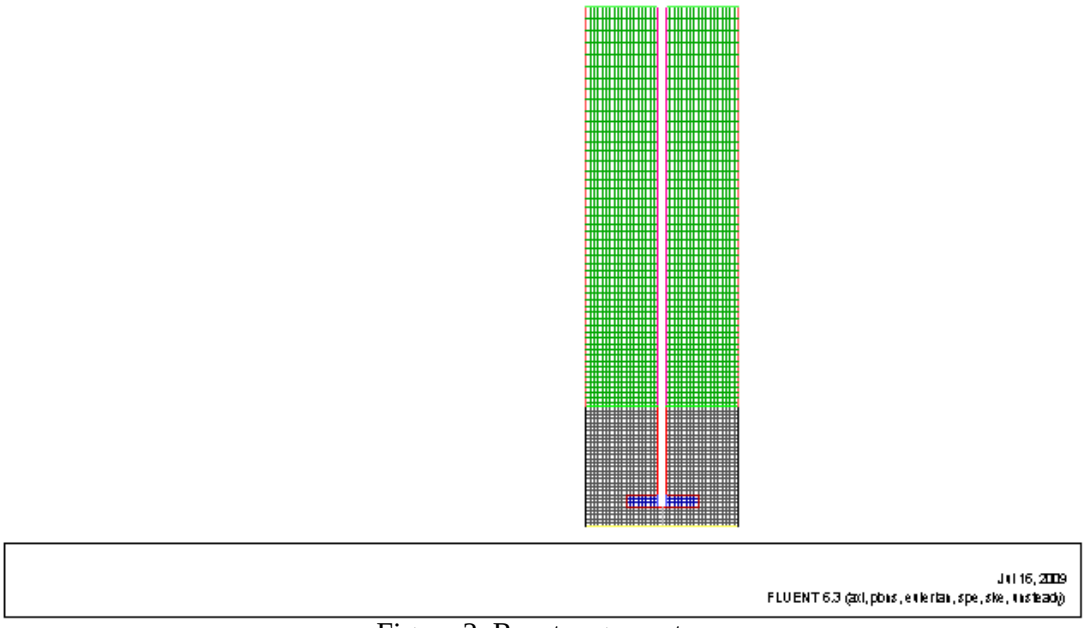

Figure 2. Reactor geometry

In order to solve the governing equations, the assumptions used in this model are:

- Coal was entered to the reactor in the slurry form.
- First order reaction and irreversible.
- Calculation was started at isothermal operating conditions.

Comparison between simulation result and experimental can be used to validate the data, therefore we can get the optimized operating conditions. The experimental data are reported in the table 1.

Table 1. Experimental data

Component	Temperature		
	430°C	450°C	470°C
DISTILATE	22.08	40.53	40.23
H2O	22.54	13.42	12.36
CLB	51.86	38.57	37.8
CO+CO2	3.23	6.7	4.24
C1-C4	4.41	7.94	9.03
H2 cons.	-4.11	-7.16	-3.67

4. RESULTS AND DISCUSSIONS

Yield of Product of Distilat at Various Operating Temperature

The effect of various operating temperature on the yield of distilate product is presented in figure 3. The experimental data indicated that the amount of distilate increase as the operating temperature increase, the largest of increasing product at the temperature of 450^{o}C. However the result of simulation did not show the significant increasing of distilate product for different operating temperature. Because the model did not consider the heating stage of BCL process. At the temperature of 450 and 470^{o}C, the amount of distilate yield from experimental data is almost similar to the simulation results which is about of 37.2 % to 40.5 %. The optimum yield of distilate was obtained at the operating temperature of 450^{o}C.

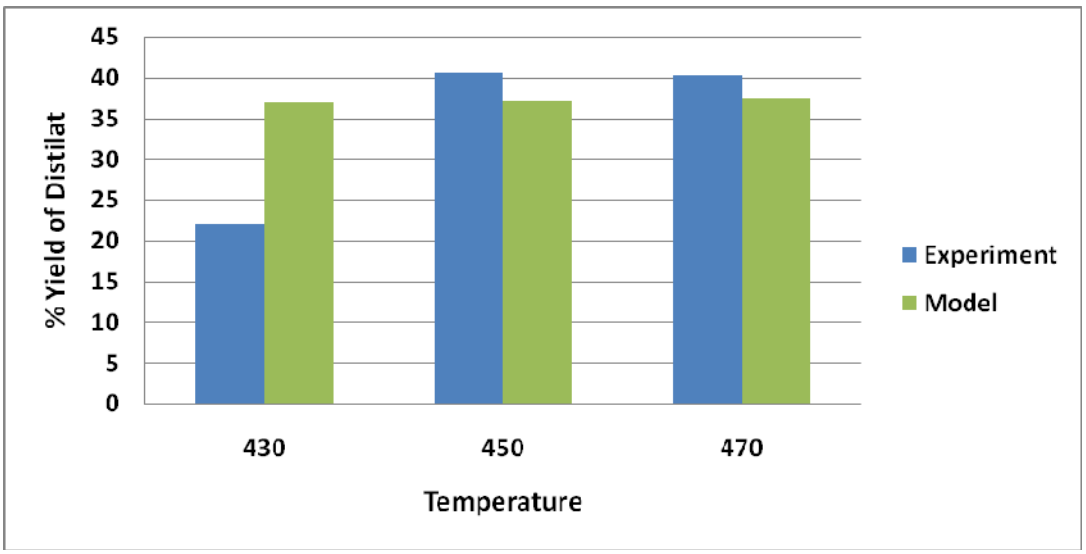

Figure 3. The Yield of Distilate Product at Various Operating Temperature

Contours of Coal Slurry at Various Operating Temperature

Figure 4 and 5 illustrated the Flow pattern of coal slurry temperatur at 450^{o}C and 470^{o}C. The red area represents the highest temperature of coal slurry and the blue area represents the lowest one. We can see from this figure that the flow pattern of temperature at the bottom region of the reactor was lower than the upper region . This is due to the effect of the geometry of impeller caused the temperature at the bottom part of reactor was not homogen.

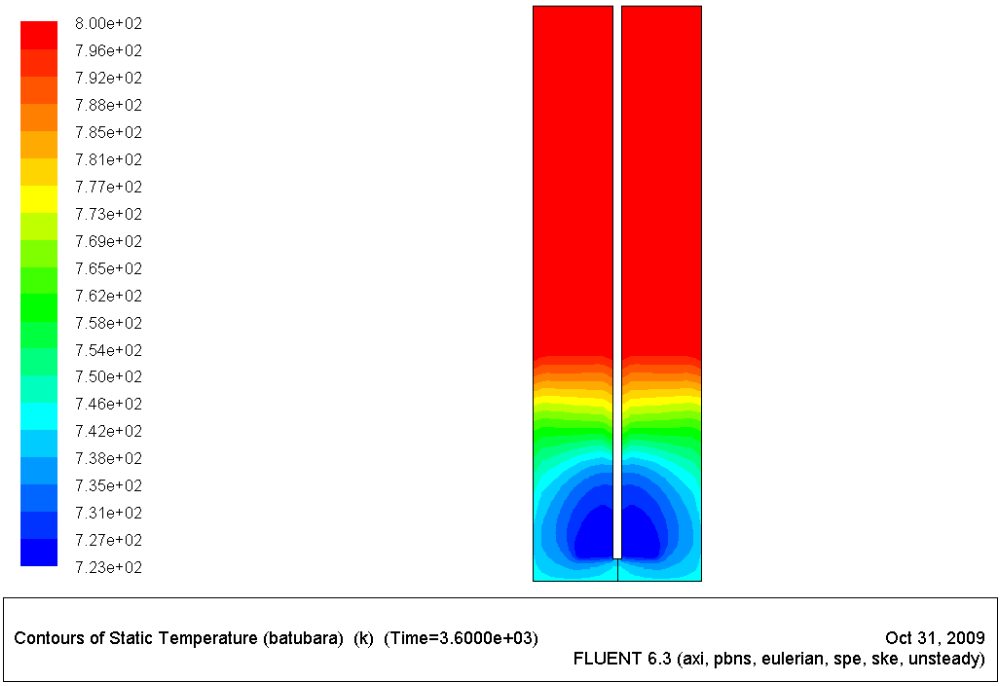

Figure 4. Contours of coal slurry temperature at 450^{o}C

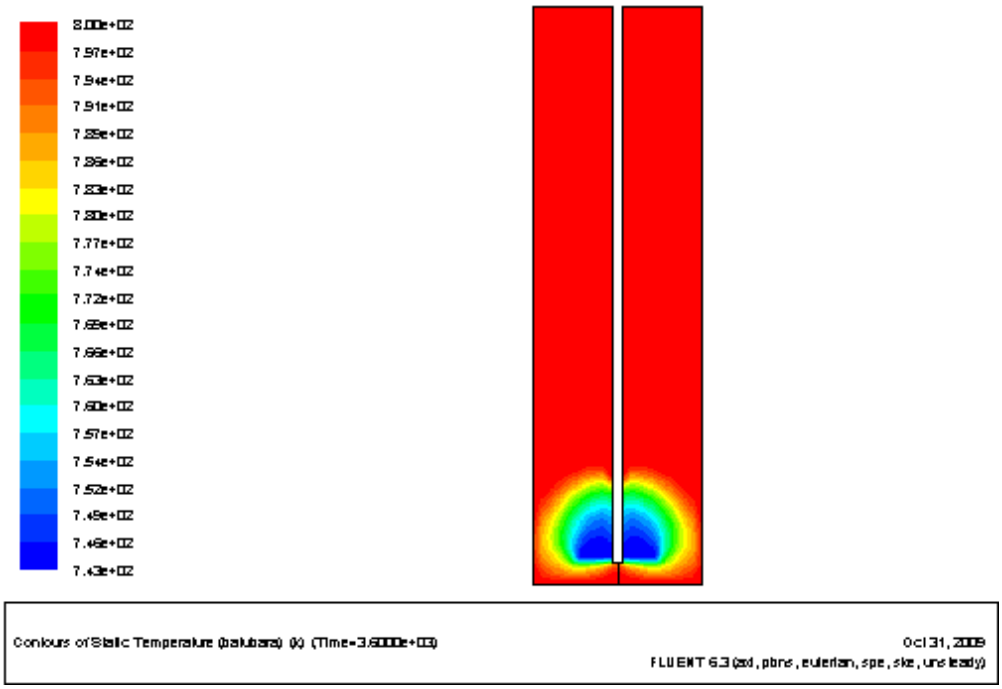

Figure 5. Contours of coal slurry temperature at 470^{o}C

The Profile of Radial Coal Slurry Temperature at Different Height of Reactor

Figure 6 describes the temperature of coal slurry at the radial position for various height of reactor. It can be seen that for the height of 0.02 metre to 0.06 metre the temperature of coal slurry at the reactor's wall is higher than the centre. Above 0.06 metre of reactor height, the temperature of coal slurry tends constant. The figure indicates the temperature of coal slurry increase from the centre of reactor to the wall.

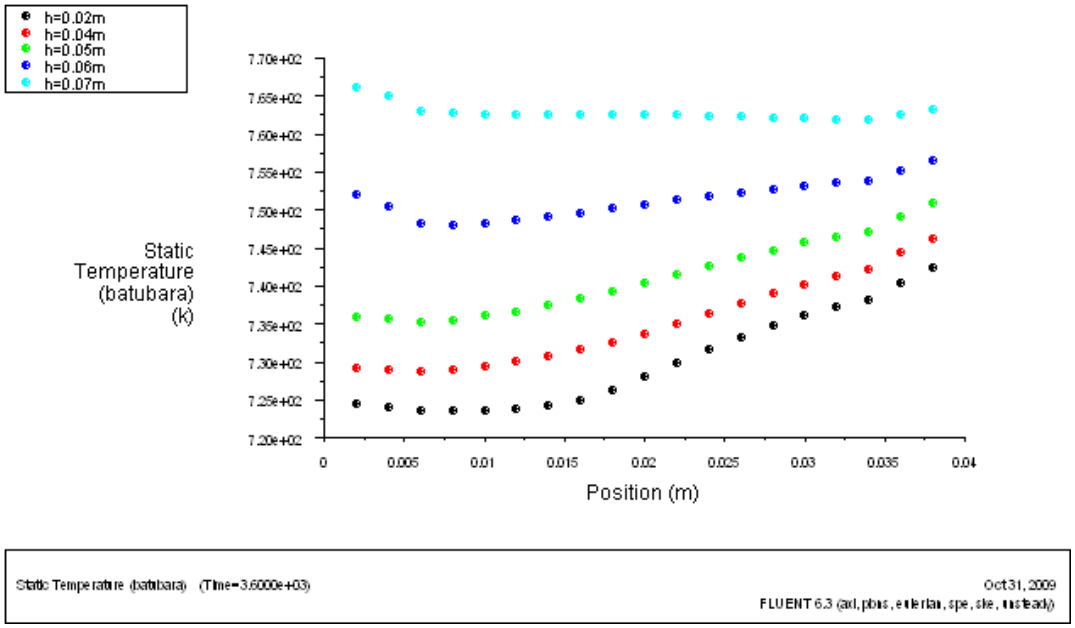

Figure 6. The Radial Coal Slurry Temperature Profile at Different Height of Reactor

5. CONCLUSION

The commercial code of Fluent 6.3 can be used to simulate two-dimensional transient multiphase and kinetics model of BCL reactor. The result of CFD simulations shows reasonable agreement with experiment. The yield of distilate product from CFD simulation is almost similar to experimental data. Optimized operating temperature obtained at 450^{o}C.

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