

CFD Simulations of coal gasification in a fluidized bed

REYES URRUTIA Andrés^{a*}, SORIA José^a, MORA BASAURE Claudia^a, ZAMBON Mariana^a and MAZZA Germán^a

^aInstituto Multidisciplinario de Investigación y Desarrollo de la Patagonia Norte (IDEPA, CONICET-UNCo) y Facultad de Ingeniería, Universidad Nacional del Comahue, Buenos Aires 1400, 8300 Neuquén – Argentina.

Abstract

In the world there is a trend towards new forms of energy, generated mainly by the progressively increment in demand for non-renewable resources, which are becoming more expensive and scarce. This has motivated countries and companies to seek new alternatives and technologies related to the exploitation and use of other energy sources. An alternative is coal, whose global reserves constitute about 65% of fossil fuel reserves in the world (Shafiee and Topal, 2009).

In this work, ANSYS-Fluent 14.0 is used for simulating coal gasification in a pilot-scale fluidized bed whose characteristics are detailed in the experimental work published by Ocampo et al. (2002).

Heterogeneous reactions representing gasification and combustion have been programmed in C ++ and coupled to the software resolution algorithm. A molecular formula has been derived for the volatile material present in the coal particles. It has also been proposed an appropriate stoichiometry for the reaction that represents the release of pyrolysis gas. In addition, homogeneous reactions are also considered in the simulations performed.

The bed reactor used by Ocampo et al. (2002) has a side feeding of coal. The height of the unit is 2 m. The bed has initially a height of 1m and is composed by sand and limestone. The simulations were performed on a 2D system with a structured mesh of 4000 cells. Multiphase Euler-Euler approach was used in order to solve the unsteady system. The particles of coal and limestone are spherical and uniform in size. Gidaspow drag model was adopted. Additionally, Gunn correlation was selected for modeling heat transfer between the gas phase and granular phases, as suggested in ANSYS-Fluent (2011) for granular systems.

In this paper we present the results of the gas composition at the reactor outlet for different operating conditions and we compare them with experimental results reported by Ocampo et al. (2002). Furthermore the results are compared with those obtained by Armstrong et al. (2011) which have also been obtained by CFD simulation.

A stationary bed temperature has also been reported throughout this process, in concordance with the experimental procedure.

The results are in good agreement with experimental data and a significant improvement over previous simulations (which use the same technique) has been achieved. It constitutes an important validation of the results obtained in this work.

Keywords: CFD; fluidized bed; coal gasification

1. Introduction

Gasification is a chemical process which consists in converting carbonaceous materials into gaseous products. Fluidized bed reactors represent an interesting alternative to carry out gasification since they allow a great control of emissions and they operate more efficiently than other units. Additionally, its versatility allows not only to vary or mix all kind of fuels (biomass, coal, sludge, household waste, etc.), but also modify the operating conditions so as to achieve an optimal operating state. In this framework,

* Authors to whom the correspondence should be sent : andres.reyes@idepa.gov.ar

the CFD simulation provides important information and a better understanding of the response of variables of interest when changing any of the conditions that define their operation.

Gasification in fixed beds as well as in fluidized beds has been extensively studied. In most of them, the theoretical molecular formula of the volatile species present in the coal arises from knowing species released during volatilization and their stoichiometric coefficients. Their values are obtained from empirical correlations, published in literature (for example see Loison and Chauvin, 1964), based on the volatile fraction present in the particles. In this work, unlike previous ones, the molecular formula of the volatile species has a direct relationship with the ultimate analysis of the carbon particles, presented in Table 1.

The object of this work is to simulate and reproduce the steady-state operating conditions of a pilot-scale bubbling fluidized bed presented by Ocampo et al. (2002). This study was carried out for two extreme operating temperatures.

Also, results from simulations are compared with those reported by Armstrong et al. (2011) obtained by means of CFD application. The authors considered that the yield of the volatile products released by pyrolysis is determined by using the correlations derived by Loison and Chauvin (1964). The authors do not consider the resistance of gas film surrounding the char particles in the effective kinetic expression ($1/h_{M,i}$ in equation 1) while in our work there have been taken into account.

2. Computational model

Among the alternatives available in ANSYS-Fluent 14.0, for the numerical solution of multiphase system, the Euler-Euler approach has been selected. Thus, the different phases are treated mathematically as interpenetrating and continuous. The approach introduces the concept of volume fraction to define phases in each cell in the computational domain.

The system consists of three phases. The primary phase is a gaseous one and contains a mixture of O₂, H₂, N₂, CH₄, H₂O, CO, CO₂ and Tar. According to Petersen and Werther (2005) and Yu et al. (2007), Tar is generally considered as condensed aromatic nuclei. They suggest that the chemical formula for Tar can be expressed as C₆H₆. The remaining two phases (limestone and coal), are defined as spherical solids. This two secondary phases are represented by an Eulerian approach. Solids in secondary phases are assumed to remain with constant size. Besides, Limestone particles (considered as inert) have an effective density of 2700 kg/m³ and have a mean particle diameter of 0.6 mm. The main features of coal are specified in Table 1.

The software solves the conservation equations for each of the phases along with the kinetic fluctuations using the kinetic theory of granular flows. The $k-\varepsilon$ model is adopted for treating turbulence.

Homogeneous reactions are resolved by the software, by using a model that considers both the kinetics and the rate of mixing (Finite-Rate/Eddy-Dissipation Model).

Second order discretization scheme for solving the convective term has been set.

The time step used is 0.001 seconds with a maximum of 100 iterations per time step. This ensures proper behavior of the residual.

A two-dimensional mesh was built using 4000 quadrilateral cells (20x200). This domain discretization arises from a mesh independence study. Average mole fractions of the reactor outlet were evaluated from simulations carried out with three different grids: 10x100, 20x200 and 40x400 cells. The 10x100 mesh results were not acceptable in comparison with 40x400 mesh, whereas, 20x200 mesh simulation results showed an error less than 1% in comparison with 40x400 mesh results, and a significant reduction in simulation times.

Unfortunately, Ocampo et al.'s (2002) do not report any results related to the hydrodynamic behavior of the bed. However, in the literature there are many other works where ANSYS-Fluent fluidized bed simulations, under Eulerian-Eulerian approach, have been successfully validated. Reyes et al. (2011), simulated an air-sand fluidized bed including heat transfer evaluation. In this work, CFD evaluations of pressure drop and minimum fluidization velocity were compared with corresponding experimental values. A very good agreement was reported for comparisons. Taghipour et al. (2005) also studied the hydrodynamics of a two-dimensional gas-solid fluidized bed reactor from both experimental and theoretical (CFD) point of view. Their CFD results were compared with experiments conducted in a

fluidized bed containing spherical glass beads with average diameters between 250 and 300 μm . Calculated values of pressure drop and bed expansion but also qualitative gas-solid flow pattern indicated reasonable agreement with experiments.

Recently, Mostafazadeh et al. (2013) investigated the hydrodynamic of a three phase gas-solid fluidized bed consisting in solid particles with diameters of 1 and 2 mm and particle densities of 2400 and 2500 kg/m^3 . The authors inform that the CFD results of pressure drop are in agreement with the measured data.

3. Experimental setup by Ocampo et al. (2002)

Figure 1 outlines the experimental device used by Ocampo et al. (2002). The reactor has an internal diameter of 0.22 m and a height of 2 m with walls constructed in stainless steel AISI 310. The coal feed is 0.3 m above the gas distributor. Initial height of the bed (fixed bed condition) is 1 m.

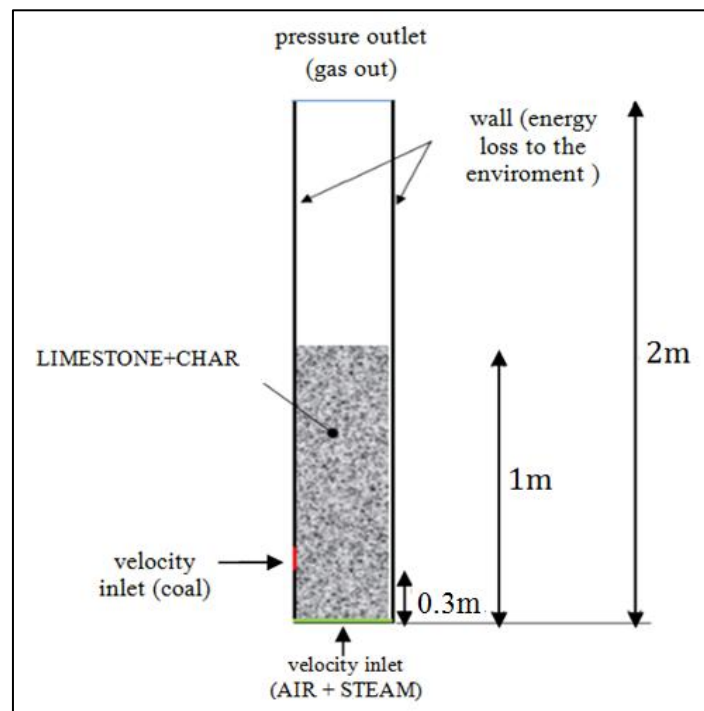


Figure 1. Schematics of the experimental set-up used in the work of Ocampo et al. (2002) along with the boundary conditions used in simulations

4. Initial and boundary conditions

The initial condition of the bed has been referred to in section 3. An appropriate set of ratios of char to limestone particles has been used in calculations on the basis of maintaining the stationary thermal condition in the bed. The initial temperature of this mixture was assumed to be the same as that of the operation of the bed. The initial porosity of the bed was set at 0.4, typical value for fixed bed condition. Only nitrogen is considered in the initial condition.

The boundary conditions used in the simulations and the volume (area in 2D approach) corresponding to the mixture of limestone and char at the beginning of the simulation are shown in Figure 1. Experimental energy losses are computed by means of a heat transfer coefficient (Gerber et al. 2010). The properties and characteristics of the solids used in this work are shown in Table 1 and the operating conditions are summarized in Table 2.

Table 1. Features and properties of solids (Ocampo et al. 2002)

Coal	
Proximate analysis	
Moisture	2.6%
Volatile matter	41.8%
Fixed carbon	54.1%
Ash	1.5%
Ultimate analysis	
Carbon	75.3%
Hydrogen	5.4%
Nitrogen	1.8%
Oxygen	15.6%
Sulphur	0.4%
Ash	1.5%
Others	
High heating value (wet basis)	29695 kJ/kg
Mean particle size	0.62 mm
Apparent density	1250 kg/m ³
Free swelling index	1
Char	
Mean particle size	0.62mm
Apparent density	450

Table 2. Operating Conditions (Ocampo et al. 2002)

	Case 1	Case 2
air supply (kg/h)	21.9	17
steam supply (kg/h)	4.7	4.7
coal feed (kg/h)	8	8
temperature at entrance (K) of air and steam	693.15	686.15
temperature of reactor (K)	1128.15	1085.15

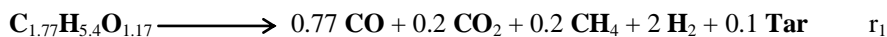
5. Moisture and volatile losses and chemical reactions involved

5.1 Moisture release

The moisture present in the coal particles is negligible. Then, its release towards the gas phase has not been considered in the simulations.

5.2 Volatile release

Frequently, combustion systems include fuels that are not easily described as pure species. However, if the heating value and the ultimate analysis (elemental composition) of the fuel are available and known, it can be defined an equivalent fuel specie and an equivalent heat of formation for this fuel (ANSYS–Fluent, 2011). This method was used to define the chemical formula of the volatile specie present in carbon particles as $C_{1.77}H_{5.4}O_{1.17}$. The release kinetic expression for this species along with its proposed stoichiometry is represented by r_1 .



The single rate model (Badzioch et al. 1970) was applied to simulate this stage. It supposes a linear function for the volatiles release rate. The rate constant of the kinetic expression was calculated by using the Arrhenius law. Values of kinetic parameters were $A = 4.92 \times 10^5 \text{ s}^{-1}$ and $E_a = 7.4 \times 10^7 \text{ J/kgmol}$.

5.3 Heterogeneous reactions

As the ash content in coal particles is low, it can be supposed that no diffusion limitations exist caused by an interface ash layer (Nagpal et al. 2003). Also it has been assumed that the reaction inside the pores is negligible. In this way a simple expression for the evaluation of the net effective rate r_j is given by

equation 1. This takes into account both the diffusion in the film and the surface chemical reaction for each heterogeneous reaction in Table 3.

$$r_j = \frac{Ac \cdot C_{i,bulk}}{1/h_{M,i} + 1/k_j} \quad (1)$$

The extrinsic parameter Ac is the specific surface area of carbon particles per unit reactor volume. $C_{i,bulk}$ is the concentration of the reactant species within the gaseous phase.

The mass transfer coefficient $h_{M,i}$ film was calculated using the correlation of Ranz and Marshall (1952) (equation 2).

The rate constant k_j depends on the temperature according to the Arrhenius law (equation 4).

The values of parameters $k_{j,0}$, b and E_a have been reported by Nagpal et al. (2003) and are shown in table 4.

Table 3. Heterogeneous Reactions

Combustion	$C + O_2 \longrightarrow CO_2$	r_2
Boudouard	$C + CO_2 \longrightarrow 2CO$	r_3
Water-gas	$C + H_2O \longrightarrow CO + H_2$	r_4
Methanation	$C + 2 H_2 \longrightarrow CH_4$	r_5

$$Sh = \frac{h_{M,i} d_p}{D_{m,i}} = 2 + 0.6 Sc^{1/3} Re_p^{1/2} \quad (2)$$

where

$$Re_p = \frac{\rho_{gas} |v_{gas} - v_{char}| d_p}{\mu_{gas}} \quad (3)$$

$$k_j = k_{j,0} T_p^b e^{-E_a/R T} \quad (4)$$

Table 4. Combustion and Gasification Kinetic Parameters (Nagpal et al. 2003)

Reaction	E_a (J/Kmol)	$k_{j,0}$ (m/(s.K ^b))	b
r_2	85.63×10^6	1.22	1
r_3	129.7×10^6	3.42	1
r_4	240×10^6	208	1
r_5	12.56×10^6	2777.7	0

5.4 Homogeneous reactions

Homogeneous reactions and the corresponding kinetic expressions were taken from the work of De Souza-Santos (1989) and are detailed in Table 5.

Table 5. Homogeneous Reactions and Kinetic Rate (de Souza-Santos, 1989)

Reaction	r_i (Kmol/m ³ .s)	
$CO + 0.5 O_2 \longrightarrow CO_2$	$r_6 = 1.0 \times 10^{15} e^{-16000/T_g} C_{CO} C_{O_2}^{0.5} \rho_g^{1.5}$	r_6
$H_2 + 0.5 O_2 \longrightarrow H_2O$	$r_7 = 5.159 \times 10^{15} e^{-3430/T_g} T^{-1.5} C_{H_2}^{1.5} C_{O_2} \rho_g^{2.5}$	r_7
$CH_4 + O_2 \longrightarrow 2H_2O + CO_2$	$r_8 = 3.552 \times 10^{14} e^{-15700/T_g} T^{-1} C_{CH_4} C_{O_2} \rho_g^2$	r_8
$CO + H_2O \rightleftharpoons H_2 + CO_2$	$r_9 = 2780 e^{-1510/T_g} [C_{CO} C_{H_2O} - \frac{C_{H_2} C_{CO_2}}{0.0265 e^{3968/T_g}}]$	r_9

6. Results and discussion

Figure 2 shows the mole fractions values obtained in the effluent stream of the bed, for the two operating conditions. CFD simulations show a short transient of approximately 4 seconds. After that period, the system reaches the steady state. Average mole fractions values were calculated under this condition.

Figure 3 shows the temperature of the mixture ($T_{mixture}$) at a height of 0.5 m for both cases. An initial proportion of char and limestone of 66% and 34% respectively was used. This proportion allows a stable thermal condition similar to that reported by Ocampo et al. (2002). In both cases it is possible to observe that, during the simulation period, temperature values fluctuate around the experimental operation value.

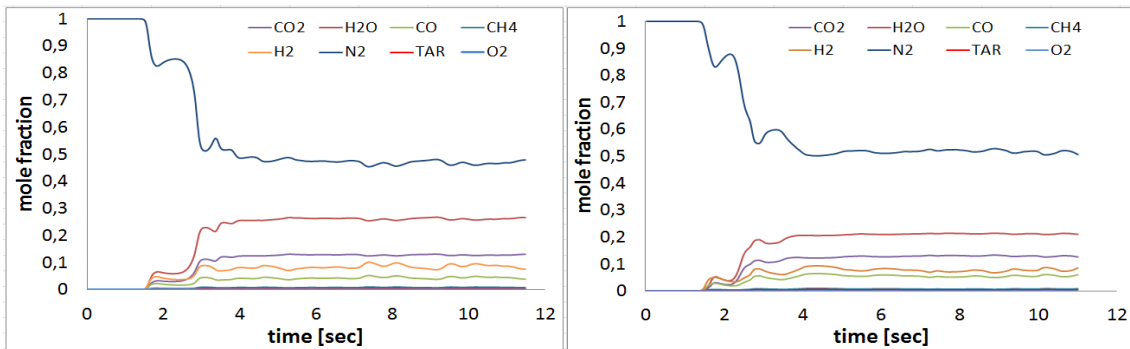


Figure 2. Mole fractions in the reactor outlet. The curves on the left figure correspond to Case 1, and that on the right plot are the results for Case 2

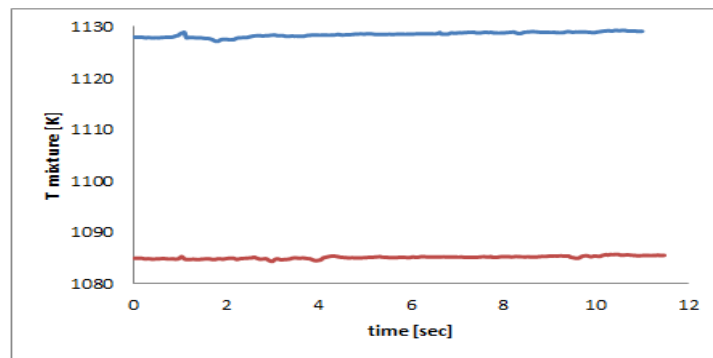


Figure 3. Temperature of the mixture at a height of 0.5 m in both simulations

Table 6 shows the average molar percentages values obtained by CFD simulations (called CFD_1) for the two operating conditions together with the experimental results reported by Ocampo et al. (2002) and those calculated by Armstrong et al. (2011) by means of CFD (denoted as CFD_2). It is important to point out here that Ocampo et al's results are established on a dry basis (table 7). Under this condition the comparison is not valid. To compare our CFD predictions with these values, the steam fraction was previously eliminated from our results (figures 4 and 5).

Table 6. Results obtained in this study vs. experimental results obtained by Ocampo et al. (2002) and Armstrong et al. (2011) also obtained by CFD

Species	T(1085.15 K)			T(1128.15)		
	CFD_1	Experimental	CFD_2	CFD_1	Experimental	CFD_2
CO ₂	12.75	18.38	10.906	12.89	19.31	11.02
CO	4.34	10.59	6.748	5.59	10.94	8.50
CH ₄	0.79	1.07	0.004	0.73	0.84	0.006
H ₂	8.45	8.84	3.898	7.78	8.53	4.26
N ₂	47.17	61.10	53.538	51.52	60.37	56.55
H ₂ O	26.09	not given	24.90	21.11	not given	19.65
Tar	0.41	not given	0.007	0.38	not given	0.009

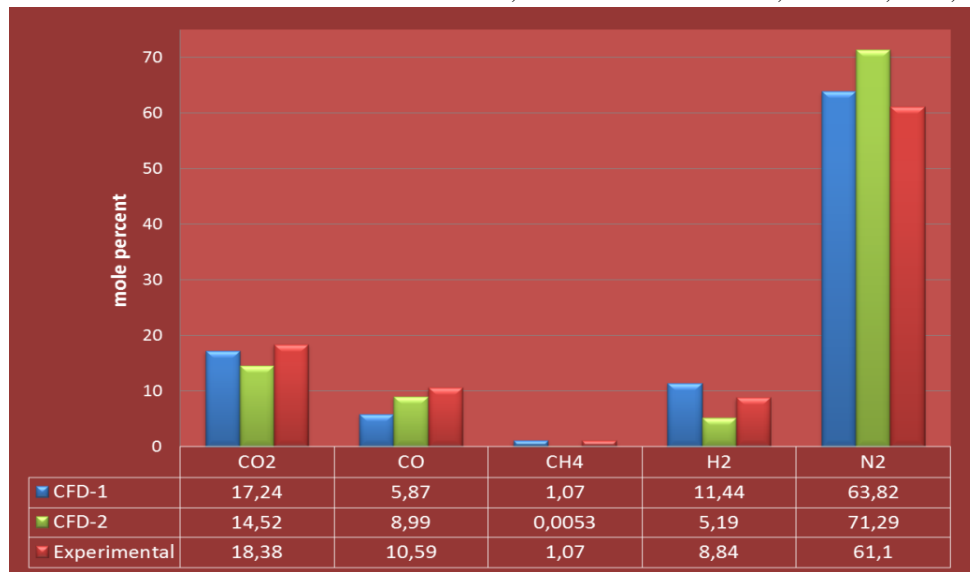


Figure 4. Comparison between dry-basis mole percent at the reactor outlet for Case 1

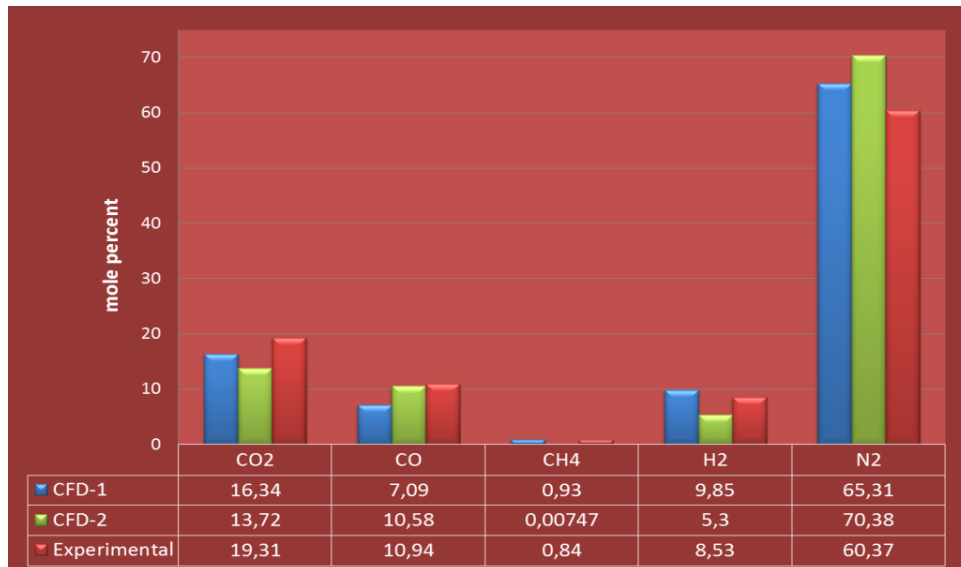


Figure 5. Comparison between dry-basis mole percent at the reactor outlet for Case 2

From the values listed above, it can be concluded that the results of the CFD study obtained in this work show a good agreement with experimental measurements for CH₄ mole fraction percent. For the other species, with an exception for CO, there is an improvement in comparison with the results reported by Armstrong et al. (2011). The discrepancies observed in CO predictions, for the two temperatures simulated, might be caused by the hypothesis of a complete combustion adopted in the simulations instead of an appropriate CO/CO₂ ratio as a function of temperature (e.g. Arthur's law (Mazza et al. 2009)).

7. Conclusion

Numerical simulations applying the Eulerian-Eulerian model for a coal gasification process in a pilot-scale bubbling fluidized bed were performed. Kinetic expressions corresponding to heterogeneous reactions of gasification and combustion have been programmed in C++ as specific *user defined functions (UDF's)* and coupled with the software resolution algorithm. A molecular formula has been derived for the volatile material present in the coal particles. It has also been proposed an appropriate

stoichiometry for the kinetic rate of the volatile material release reaction. We have also considered the reactions occurring in the homogeneous phase.

The results concerning the composition of outlet gas stream are in good agreement with the experimental results reported by Ocampo et al. (2002) used to validate the simulations. Furthermore, it can also be concluded that the methodology applied in this work is an improvement over the methodology used by Armstrong et al. (2011) in their CFD study.

The higher error of simulations (with respect to experimental values) is observed for CO concentration. To achieve a better agreement between simulations and experiments, simulations should be performed by using a suitable CO/CO₂ ratio as a function of the temperature to account for the coal incomplete combustion reaction. This improvement will be a more realistic approach to coal gasification process.

Acknowledgements

Authors express their gratitude for the financial support by the *Consejo de Investigaciones Científicas y Técnicas (CONICET)*, the *Agencia Nacional de Promoción Científica y Tecnológica (ANPCyT)* and the *Universidad Nacional del Comahue (UNCo)*.

References

- ANSYS, Inc., ANSYS-Fluent 12, Theory Guide, 2011.
- Armstrong, L.M., S. Gu, and K.H. Luo, 2011, Parametric Study of Gasification Processes in a BFB Coal Gasifier. *Industrial & Engineering Chemistry Research*, 50, 5959-5974.
- Badzioch, S. and P. G. W. Hawksley, 1970, Kinetics of thermal decomposition of pulverized coal particles. *Ind. Eng. Chem. Process Des. Dev.*, 9, 521-530.
- Gerber, S.; F. Behrendt and M. Oevermann, 2010, An Eulerian modeling approach of wood gasification in a bubbling fluidized bed reactor using char as bed material. *Fuel*, 89 (10), 2903-2917.
- De Souza-Santos ML., 1989, Comprehensive modeling and simulation of fluidized bed boilers and gasifiers. *Fuel*, 68 (12), 1507-1521.
- Loison R. and R. Chauvin, 1964, Pyrolyse rapide du charbon, *Chimie et Industrie*, 91 (3), 269-275.
- Mazza, G., Q. Falcoz, D. Gauthier and G. Flamant, 2009, A particulate model of solid waste incineration in a fluidized bed combining combustion and heavy metal vaporization. *Combustion and Flame*, 156, 2084-2092.
- Mostafazadeh M., Rahimzadeh H. and M. Hamzei, 2013, Numerical analysis of the mixing process in a gas–solid fluidized bed reactor, *Powder Technology*, 239, 422-433.
- Nagpal, S., T.K. Sarkar and P.K. Sen, 2005, Simulation of petcoke gasification in slagging moving bed reactors. *Fuel Processing Technology*, 86, 617-640.
- Ocampo, A., E. Arenas, F. Chejne, J. Espinel, C. Londoño, J. Aguirre and J.D. Perez, 2003, An experimental study on gasification of Colombian coal in fluidised bed. *Fuel*, 82, 161-164.
- Petersen, I. and J. Werther, 2005, Experimental investigation and modeling of gasification of sewage sludge in the circulating fluidized bed. *Chem. Eng. Prog.*, 44, 717-736.
- Ranz W.E. and W.R. Marshall, 1952, Jr. Evaporation from Drops, *Chem. Eng. Prog.*, 48(3), 141-146.
- Reyes A., Galeazzi F., Zambon M., Flamant G., Gauthier D and G. Mazza, 2011, Estudio de la pérdida de carga y de la transferencia de calor en un lecho fluidizado burbujeante mediante la utilización de Fluidodinámica computacional (CFD), *Mecánica Computacional*, 30,1669-1687.
- Shafiee S. and E. Topal, 2009, When will fossil fuel reserves be diminished? *Energy Policy*, 37, 181–189.
- Taghipour F., Ellis N. and C. Wong, 2005, Experimental and computational study of gas–solid fluidized bed hydrodynamics, *Chemical Engineering Science*, 60, 6857 - 6867.
- Yu L., J. Lu, X. Zhang and S. Zhang, 2007, Numerical Simulation of the bubbling fluidized bed coal gasification by kinetic theory of granular flow (KTGF). *Fuel*, 86, 722-734.

Simulation CFD de la gazéification du charbon en lit fluidisé

REYES URRUTIA Andrés^{a*}, SORIA José^a, MORA BASSAURE Claudia^a, ZAMBON Mariana^a et MAZZA Germán^a

^a Instituto Multidisciplinario de Investigación y Desarrollo de la Patagonia Norte (IDEPA, CONICET-UNCo) y Facultad de Ingeniería, Universidad Nacional del Comahue, Buenos Aires 1400, 8300 Neuquén – Argentina.

Résumé

Dans le monde il y a une tendance vers de nouvelles formes d'énergie, générée principalement par l'augmentation progressive de la demande de ressources non renouvelables, qui sont de plus en plus rares et chères. Cela a provoqué de l'intérêt des différents pays ainsi que des entreprises à rechercher de nouvelles alternatives et les technologies liées à l'exploitation et l'utilisation des autres sources d'énergie. Une alternative peut être le charbon, dont les réserves mondiales représentent environ 65% des réserves de combustibles fossiles dans le monde (Shafiee et Topal, 2009).

Dans ce travail, le logiciel ANSYS Fluent-14.0 est utilisé pour simuler la gazéification du charbon dans un lit fluidisé à l'échelle pilote dont les caractéristiques sont détaillées dans le travail expérimental publié par Ocampo et al. (2002).

Les réactions hétérogènes représentant la gazéification et la combustion ont été programmées en C++ et couplées à l'algorithme de résolution du logiciel. Une formule moléculaire a été obtenue à partir de la matière volatile présente dans les particules de charbon. Il est également proposé une stoechiométrie appropriée pour la réaction de dégagement de gaz de pyrolyse. Les réactions homogènes sont également prises en compte dans les simulations.

Le réacteur à lit fluidisé utilisé par Ocampo et al. (2002), présente d'un côté un dispositif qui permet l'entrée du charbon. La hauteur de l'unité est de 2 m. Dans le moment initial le lit fixe a une hauteur de 1 m et se compose de sable et de calcaire. Les simulations ont été réalisées sur un approche 2D avec un maillage structuré conformé par 4000 cellules. L'alternative de modélisation Euler-Euler multiphasique a été utilisée pour résoudre le système transitoire. Les particules de solide (charbon et calcaire) sont toujours supposées en géométrie sphérique et de taille uniforme. Le modèle de résistance Gidaspow a été adopté. En outre, la corrélation Gunn a été sélectionnée pour le transfert de chaleur entre les phases gazeuse et granulaire, comme il a été suggéré par ANSYS Fluent (2011) pour ce type de système.

Cet article présente les résultats de la composition du gaz à la sortie du réacteur pour de différentes conditions de fonctionnement et sa comparaison avec les résultats expérimentaux rapportés par Ocampo et al. (2002). Les résultats ont été aussi comparés avec ceux obtenus par Armstrong et al. (2011) par la voie de la simulation CFD. La température du lit fluidisé au fil du temps a également été observée puis affichée et le résultat est coïncidant dans son comportement stationnaire avec la valeur expérimentale.

Les résultats de concentration montrent aussi un bon accord avec les données expérimentales et une amélioration significative au cours des simulations précédentes (en utilisant la même technique). Sur cette base, ils valident le processus proposé dans ce travail.

Mots-clés: CFD; lit fluidisé; gazéification du charbon