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Steam gasification of char from wood chips fast pyrolysis: Development of a semi-empirical model for a fluidized bed reactor application

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ABSTRACT

This study, performed in the context of GAYA project, focuses on the development of a simple predictive model about steam gasification of char from woodchips fast pyrolysis. A semi-empirical model was developed through experiments in a macro thermogravimetric analyzer which owns the peculiar ability of fast heating, as well as to deal with macro-size particles and higher mass loads compared to conventional TGA. The experimental results show that gasification is controlled by chemical kinetics and internal transfer phenomena. During gasification, char particles can be considered as isothermal in a given range of temperatures and particle sizes, more likely for low values. The gasification model was based on the effectiveness factor, which involves the chemical kinetics and diffusion rate. The chemical kinetics were expressed by a classical Arrhenius law, whereas empirical expressions from mathematical fitting of the experimental data were established for the diffusion coefficient and surface function. The diffusion coefficient from this work is suspected to probably include supplementary rate limiting phenomena, apart from steam porous diffusion, such as H_2 inhibition and/or the decrease of temperature within char particles because of the endothermic character of gasification. The model globally predicts with accuracy the gasification rate in typical operating conditions of a fluidized bed reactor. For its simplicity and reliability, this approach can be used for the modelling of char gasification in the conditions of interest.

1. Introduction

Biomass gasification is a process with a high potential for energetic transition, as it represents a sustainable alternative for fossil fuels with low greenhouse gas emission. Through this process, biomass is decomposed into a gas mixture, mainly composed of H_2 and CO, due to the action of heat and chemical reagents such as H_2O, CO_2 or O_2 in stoichiometric default. The gas produced by gasification has many energetic applications, as the synthesis of biomethane that can be used as fuel for transportation and power generation.

The GAYA Project aims to demonstrate the industrial, technical and economic feasibility of biomethane production

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Nomenclature

C_{H2O}	steam concentration, $mol \cdot m^{-3}$
D	diffusion coefficient, $m^2 \cdot s^{-1}$
D_k	Knusden diffusion coefficient, $m^2 \cdot s^{-1}$
D_m	molecular diffusion coefficient, $m^2 \cdot s^{-1}$
E_a	activation energy $J \cdot mol^{-1}$
$f(X)$	surface function
k_0	pre-exponential factor $Pa^{-n} \cdot s^{-1}$
k_{int}	chemical kinetics constant, s^{-1}
M_{char}	char molecular weight, $kg \cdot mol^{-1}$
M_{H2O}	water molecular weight, $kg \cdot mol^{-1}$
M_{N2}	nitrogen molecular weight, $kg \cdot mol^{-1}$
L_c	characteristic length, m
$m(t)$	sample mass, g
m_0	initial mass sample, g
n	order of the reaction
P_t	total pressure, Pa
P_{H2O}	steam partial pressure, Pa
R	ideal gas constant, $J^{-1} \cdot mol^{-1} \cdot K^{-1}$
R_{ref}	reference reactivity, s^{-1}
$R(t)$ or $R(X)$	reactivity, s^{-1}
r_{app}	apparent reaction rate, $mol \cdot m^{-3} \cdot s^{-1}$
r_D	diffusion rate, $mol \cdot m^{-3} \cdot s^{-1}$
r_{int}	intrinsic reaction rate, $mol \cdot m^{-3} \cdot s^{-1}$
r_p	mean pore radius, m
S_p	specific surface area, $m^2 \cdot kg^{-1}$
T	temperature, K
X or $X(t)$	conversion
x	thickness, m

Greek nomenclature

ε	porosity
η	efficiency
Φ	Thiele modulus
ρ_{app}	apparent density, $kg \cdot m^{-3}$
ρ_{bulk}	bulk density, $kg \cdot m^{-3}$
$(\Sigma\Upsilon)_{H20}$	steam diffusion volume
$(\Sigma\Upsilon)_{N2}$	nitrogen diffusion volume
τ	tortuosity

Abbreviations

CO	carbon monoxide
CO_2	carbon dioxide
FBR	fluidized bed reactor
H_2	hydrogen
H_2O	steam
TGA	thermogravimetric analyzer

by biomass gasification in France. The technology selected, a fast internally circulating fluidized bed reactor, couples a bubbling fluidized bed reactor (FBR) for gasification and a transported bed reactor for combustion. In the gasifier, the biomass is pyrolyzed in an atmosphere composed of steam, resulting in the production of gas (~70% mass fraction), tar (~15% mass fraction) and char (~15% mass fraction). The pyrolysis products subsequently react among them and/or with the steam present in the atmosphere. The char is partially gasified during its residence in the gasifier. In the combustor, the oxidation of the remaining char provides the required heat for biomass thermochemical conversion in the gasifier which is globally endothermic.

The char residence time in the gasifier needs to be optimised in order to simultaneously maximize the syngas production through char steam gasification and leave enough residual solid for burning in the combustor to keep autothermicity. The control of char residence time in the gasifier is then a key parameter of the process. For this purpose, the understanding of char gasification kinetics on the conditions of interest and the development of a predictive model is essential. The gasification model should then be integrated into a reactor model which describes biomass decomposition in a FBR. Noubli et al. [1] describes the modelling approach selected for the reactor model.

Major part of the studies about char gasification in literature focuses on chemical kinetics, neglecting heat and mass transfer phenomena. Chemical kinetics is typically determined with experiments in a thermogravimetric analyzer (TGA), which enables to work on controlled conditions. Nevertheless, in industrial FBR, biomass feedstock usually consists of large particles: char gasification may then be limited by heat and mass transfer phenomena. Literature about macrosized char particles gasification, which is much less abundant in comparison to intrinsic kinetic studies, puts into evidence the limitation by transfer phenomena. Globally, a decrease of gasification rate is observed by increasing the char particle size. The critical char size from which transfers become limiting depends on the chemical and physical characteristics of the fuel, as well as on the reactor operating conditions. In the case of spherical wood char, several works agree that this critical size is located in the scale of the millimeter [2–4] in the temperature range of 800–1000 °C. A critical size in the same order of magnitude has been found for char from woodchips by Van de Steene et al. [5].

In order to describe the gasification of large char particles, several models considering chemical kinetics and transfer phenomena have been developed, either structural or volumetric [5–10]. These models describe the local changes in internal structure with conversion, notably pore development, but require too high computing capacities to be integrated in a reactor model. On the other hand, several studies have used analytical and semi-analytical approaches in order to simplify char gasification modelling. These approaches attempt to capture the main chemical and physical phenomena in a way as simple as possible [11–15]. Other authors have employed simple empirical correlations, as Standish et al. [16] who have included the initial particle size, raised to a power of 0.81, in an n^{th} order apparent kinetic expression. Teixeira et al. [17] developed an empirical correlation of conversion rate as a function of the operating conditions, namely temperature and reagent partial pressure, and char particle characteristics, namely size, porosity and pre-exponential factor.

More information about large char particles gasification models is provided in an extensive review by Gomez-Barea [18].

The objective of the present work, performed in the context of GAYA project, is to characterize and model steam gasification of large char particles. The char gasification model