



Full Length Article

Deactivation kinetics for lignite gasification in a fluidized bed reactor

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ABSTRACT

Lignite gasification may be characterized by the active site restriction result from deactivation. As was aimed here, a deactivation model (DM) can be developed by “pseudo-steady-state” mass balance, i.e. describing variation of available surface with time. For this purpose, bench-scale fluidized bed reactor (FBR) was operated for obtaining gasification product profiles. Gas samples were taken from the reactor effluent stream, and fed continuously to the gas analyzers for on-line simultaneous measurements of H₂, CO₂, CO, CH₄ and O₂. Data were used for extracting model parameters (k'' and k_D) from the linear form of DM. Experiments at temperature range of 973–1173 K were conducted. Fluidizing mixture (air and steam) entered the bed through a distributor of a 200-mesh stainless steel sieve and fluidized the single charge of lignite with a mean particle diameter approximately 250 μm . The agreement between the experimental and predicted carbon concentrations was confirmed for DM. The latter may be successfully used to design the fluidized bed combustors or gasifiers.

1. Introduction

Coal is generally used as an energy source by direct combustion. But combustion of coal with an unprocessed form causes various environmental problems. The amount of harmful emissions depends on the efficiency and utilization method of combustion process and the properties of the fuel source used. In this regard gasification seems to be an appropriate technique for coal utilization [1].

But no matter the process, some obstacles like agglomeration and sintering still appear as the common operation problems. Fluidized Beds have fuel flexibility concerning the particle size and moisture content but suffer from two major setbacks: a generic problem associated with increased tar content in the gas products that inhibits its efficient utilization and a fuel-specific problem, caused by the low melting temperatures of ashes that paves the way for particle sintering, agglomeration, leading to the inevitable defluidization of the bed [2].

Agglomeration problems are related to the transformations of mineral matter in solid fuels which depend on the type and composition of the fuel. The melting behavior of mineral matter in fuel is considered to be an important parameter. Where a sorbent is used for emissions control, such as the use of limestone in FB combustion, mineral species derived from the sorbent can also have a role. A model for prediction of agglomeration problems in specific operating conditions seems crucial for the utilization of solid fuels [3].

As for the sintering, present mining processes produce large quantities of coal fines, and also some beneficiation techniques require

grinding of the raw material to fine or ultra-fine particles. With increasing quantities of coal fines occurrence of sintering seems more likely as it was observed in some of our studies [4].

Agglomeration and sintering may cause unforeseen consequences in the operation. Silica sand is the most typical bed material in fluidized beds because of its good mechanical properties and abundant reserves [5–9]. However, agglomeration of silica sand bed material is widely reported [10]. Inorganic alkali material in the fuel, mainly potassium and sodium cause agglomeration by the formation of silicates with the silica from the sand which has low melting points. The content of these inorganic components can vary between fuels; especially in the case of some biomass types as well as various low-rank coals. As a result, sand particles get coated with an adhesive layer. With the collision of solid structure sticky sand particles create larger agglomerates [11].

Some precautions for reducing the agglomeration risk, is to use mineral based bed materials, such as Al₂O₃, olivine and dolomite which are resistant toward agglomeration or to add some materials like kaolin, calcium oxide, calcium carbonate and bauxite which have the ability to trap or react with components causing agglomeration. Minerals are considered to be more or less brittle and sensitive toward attrition when used as bed material in fluidized bed gasification [10]. Most of the previous reviews focused on the agglomeration characteristics of fluidized beds based on chemical properties of fuel. Physics based parameters like particle size, velocity and collision frequency also affect the agglomeration. It is important to conceive these effects along with the particle chemistry. For example, bed additives not only transform the

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Nomenclature			
k''	reaction rate constant, s^{-1}	D	reactor diameter, m
k_D	deactivation rate constant, s^{-1}	L	reactor height, m
C	concentration, $kmole.m^{-3}$	m	mass, kg
F	molar FLOW rate, $kmole.s^{-1}$	P	pressure, kPa
V	reactor volume, m^3	T	temperature, K
Q	volumetric flow rate, $m^3.s^{-1}$	d_p	particle size, m
y	mole fraction, $kmole.kmole^{-1}$	x_C	carbon conversion ratio, $kmole.kmole^{-1}$
r	reaction rate, $kmole.m^{-3}.s^{-1}$	<i>Greek Letters</i>	
S	particle surface area, m^2	α	stoichiometric coefficient of steam, (-)
t	Time, s	β	stoichiometric coefficient of air, (-)
k_s	surface reaction rate constant, $m.s^{-1}$	γ	total amount of converted carbon, $kmole.m^{-3}$
E_A	activation energy for surface reaction, $kJ.kmole^{-1}.K^{-1}$	ρ	density, $kg.m^{-3}$
E_D	activation energy for deactivation, $kJ.kmole^{-1}.K^{-1}$		

chemical structure of the bed materials but also change the mean particle size and density. This would affect the hydrodynamics in bed and physics-based parameters, which may affect agglomeration rate [12].

Namkung et al. investigated agglomeration tendency using different operating conditions consisting particle size, coal/sand ratio, temperature and fuel type. They used various materials like kaolin, alumina and additives to prevent bed agglomeration and found out that agglomeration tendency increased with smaller particle sizes and higher temperatures [13].

Lin et al. performed thermodynamic equilibrium calculations to identify the stable silica, potassium, chlorine and sulfur species. Their results showed that potassium silicates were the main form present in the bed. They also developed a simple model to describe the defluidization time including parameters like function temperature, fluidization velocity and particle size. The model was based on a competition between the breaking force induced by bubbles in the bed and the adhesive force caused by ash coating and sintering [14].

Tang et al. studied two mineralogically different lignite chars and reported that commonly known kinetic models like VM (Volumetric Model), SCM (Shrinking Core Model) and RPM (Random Pore Model) are unsatisfactory, also in some cases invalid to express the kinetics of char gasification. Therefore proposed a new active site/intermediate

model (ASIM), in terms of a simplified calcium-catalyzed mechanism of char gasification to characterize a conversion-dependent maximum in reaction rate [15].

Dahlin et al. evaluated different additives to assess the ability to prevent ash agglomeration during the high-sodium lignite gasification. They conducted some series of muffle furnace tests selected meta-kaolin for a following work with a pilot-scale coal gasifier. Agglomeration and deposition problems during gasification of high-sodium lignite successfully prevented at a maximum operating temperature of 1200 K and a meta-kaolin (mean size of 920 μm) feed rate with roughly equivalent to the ash content of the lignite (approximately 10 wt%) [16].

Khadilkar et al. conducted a research with particle classes of the composite fuels, based on differences in density and size, in order to understand the physics and chemistry at particle level. They determined slag-liquid formation tendencies under fluidized bed operating temperatures both computationally and experimentally. They used a thermodynamic simulation software and proposed an integrated ash agglomeration model that accounts for particle hydrodynamics as well as particle class level ash chemistry to predict agglomeration kinetics [17].

As a continuation of previous studies outlined above, the aim of this study is to examine deactivation kinetics for lignite gasification in a

Table 1
Lignite characterization tests.

Tests	Results										
Pore size analysis: AUTOPORE II 9220	Total pore volume: 0.116 mL/g; Mean pore radius: 0.003 μm ; Porosity: 17.417%										
Tests	Results	Volatile matter	Moisture	Fixed carbon	Ash	Calorific value (kJ/kg)					
Proximate analysis: (ASTM-D-7582)/(ASTM-D-5865)	Weight (%):	35.65	6.28	11.96	46.12	9 990					
Tests	Results	C	H	O	N	S	Ash				
Ultimate analysis: (ASTM-D-5373)/(ASTM-D-7582)/(ASTM-D-5016)/(ASTM-D-121)	Dry basis (%):	30.97	2.69	10.62	4.82	1.69	49.21				
	Ash-free dry basis (%):	60.98	5.29	20.90	9.50	3.33					
Tests	Results	MgO	Al ₂ O ₃	SiO ₂	CaO	Fe ₂ O ₃	SO ₃	K ₂ O	Na ₂ O	P ₂ O ₅	H. Loss
Complete chemical analysis: (ASTM-D-4326-04)	Ash (%):	2.49	13.27	43.00	21.05	7.93	6.56	1.08	1.32	0.48	1.66
	Slag (%):	2.11	14.69	46.40	11.95	6.97	2.12	1.18	0.61	0.35	12.47

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