



High quality syngas production via steam-oxygen blown bubbling fluidised bed gasifier



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ABSTRACT

This paper presents experimental and modelling results of fuel gas obtained in a steam-oxygen blown fluidised bed pilot scale (500 kW_{th}) coal gasifier that is part of the ZECOMIX (Zero Emission of CarbOn with MIXed Technologies) research infrastructure run by ENEA. As there is poor information about start up and steady state operation of fluidised bed gasifiers at such a scale, in this work we investigated the influence of some important parameters such as coal ignition temperature, feeding rate of particulate bed material (olivine sand) and transition from oxidant to reducing environment. The experimental runs confirmed the crucial importance of S/O (steam/oxygen) ratio on the product gas composition. High quality syngas (low content of methane, below 1.0 v/v %, and high content of H₂ and CO: CO + H₂ 61.0 v/v %) was obtained with S/O = 1.1.

A shortcut model of the gasifier was also formulated, considering instantaneous coal pyrolysis, kinetics of coke steam reforming reactions in the fluidised bed, and achievement of thermodynamic equilibrium for the water gas shift reaction. The model takes into consideration the average residence time of coke particles into the gasifier: since this is a characteristic parameter of the reactor, the performance of different fluidised bed gasifiers can be simulated.

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1. Introduction

Worldwide efforts to reduce carbon dioxide emissions has stimulated power generation with lower environmental impact. The European Union, indeed, is bound to reduce greenhouse gases by 20%, compared to 1990, by 2020. On the other hand, the utilization of fossil fuels will grow, mainly because of increasing energy needs in developing countries. As a direct consequence, the emissions of carbon dioxide will increase unless measures for their limitation are considered. The challenge in the scientific community is to combine fossil fuel use with carbon dioxide capture and sequestration, without affecting economic development and promoting sustainability. In this context, clean coal technologies are seen as a promising way for the production of low-carbon energy [1] because of coal abundance and price stability. A number of coal fuelled power plants is presently in operation; in these cases, a sensible route to keep on producing energy from coal is reducing

CO₂ emissions to the atmosphere via flue gases (post combustion CO₂ capture). On the other hand, research efforts should be focused on new technology developments able to promote energy systems providing low-carbon gaseous fuels, among these hydrogen, as innovative energy carriers. The hydrogen production from coal, indeed, is the core of important international R&D initiatives such as HYPOGEN [2,3] sponsored by European Union, or large CTL (coal-to-liquid) plants in China [4], whereas the Japanese Government promoted the EAGLE project [5].

Gasification of solid fuels is a versatile technology which can be fitted with chemical looping combustion [6] and/or CO₂ capture processes. Gasification of solid fuels as coal or biomass with O₂ and H₂O yields a synthetic gas (mainly composed of CO, CO₂, H₂O, H₂) that is more amenable to de-carbonisation (than the corresponding post-combustion flue gas) because of lower flow rate. It is processed through a fluidised bed made of a CaO-based solid sorbent for the separation of CO₂ [7–11]. By scrubbing the synthetic fuel gas prior to carbon separation, sulphur compounds (hydrogen sulphide and carbonyl sulphide) are largely abated and the CO₂ acceptor could extend its operating life.

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Symbols and abbreviations	
A(θ)	age distribution curve [–]
a	hydrogen/carbon atomic ratio in the empirical formula of daf coal [–]
a _i	coefficients in Eq. (16) [–]
b	oxygen/carbon atomic ratio in the empirical formula of daf coal [–]
C	molar concentration [kmol/m ³]
CGE	cold gas efficiency [–]
c _p	specific heat capacity [kJ kmol ⁻¹]
CRF	char reactivity factor [–]
daf	dry and ash free [–]
ER	equivalence ratio [–]
f _c	fraction of unreacted carbonaceous particle in the fluidised bed [–]
H	enthalpy convective flow [kJ h ⁻¹]
h	specific enthalpy [kJ kmol ⁻¹]
HHV	higher heating value [kJ kmol ⁻¹]
K	equilibrium constant [–]
k	kinetic constant [s ⁻¹]
LHV	lower heating value [kJ kmol ⁻¹]
MW	molecular weight [kg kmol ⁻¹]
n	molar flow rate [kmol h ⁻¹]
p	partial pressure [Pa]
P	pressure [Pa]
Q	heat flow [kJ h ⁻¹]
R	ideal gas constant [J mol ⁻¹ K ⁻¹]
SC	steam to coal weight ratio [–]
S/O	steam to oxygen weight ratio [–]
T	temperature [°C]
u	velocity [m s ⁻¹]
w	water content of coal [kg/kg]
w _i	weight fraction of i-th component in the daf coal [–]
WGS	water gas shift reaction [–]
X	extent of chemical reaction [–]
\bar{X}	average extension of chemical reaction [–]
y	stoichiometric coefficient in pyro-combustion reaction [–]
<i>Greek letters</i>	
θ	residence time of coke in the gasifier [h]
$\bar{\theta}_r$	average residence time of coke in the gasifier [h]
<i>Subscripts</i>	
0	inlet, reference
eq	equilibrium
f	formation
g	gasification
i	i-th chemical component
mf	minimum fluidisation
<i>Superscripts</i>	
0	inlet, reference

In this work, experimental and simulation results of coal gasification in a BFB (bubbling fluidised bed) oxygen-blown coal gasifier will be presented. A number of modelling and experimental works on coal at laboratory test rig scale [12–19] have been reported in the scientific literature, however there is a lack of detailed information about the start up procedure and steady state operation of a BFB oxygen-blown coal gasifier at pilot scale (500 kW_{th}).

In the model approach proposed in this work, the production of syngas has been broken down into three stages: in the first stage, the coal feedstock undergoes instantaneous pyrolysis and combustion processes in a single step: coal is rapidly converted into coke, ash and volatile matter. In the second stage, solid and heavy hydrocarbon primary products react with steam to produce a syngas, according to an endothermic chemical reaction dominated by kinetic effects. Finally the third step is governed by the water gas shift thermodynamic equilibrium.

Alternative approaches are proposed in the literature: some authors [20,21] considered and optimised a fitting parameter, the CRF (char reactivity factor), in order to fit experimental data collected during gasification of carbonaceous solid fuels. In other studies, the modelling approach [22] neglects the combustion of solid carbon in the pyro-combustion process.

2. The ZECOMIX pilot scale research infrastructure

The ZECOMIX (Zero Emission of CarbOn with MIXed Technologies) Research Infrastructure for H₂ and electricity production [23,24] is utilized for the experimental tests and to validate the gasification model. The main aim of this research infrastructure is to demonstrate, via a series of modelling and experimental activities, the feasibility of innovative low emission processes for the production of electricity and hydrogen from coal. Several processes such as coal gasification, steam methane reforming, clean-up of syngas, CO₂ capture by means of calcium looping cycle and

combustion of hydrogen in a gas micro-turbine are being investigated at ENEA with the ZECOMIX system. ZECOMIX has been conceived as an experimental platform in the sense that a number of experimental configurations can be tested:

- Gasification tests: coal is gasified with O₂ and H₂O in order to obtain a synthetic, nitrogen-free fuel gas: the product gas (raw syngas) is driven directly to a scrubber and then to the flare;
- CO₂ capture tests: the fuel gas to be decarbonised is obtained by mixing pure gases from bottles. This kind of tests allows to investigate the decarbonising process by changing the composition of the gas to be decarbonised. The inlet temperature of the syngas could be set at up to 600 °C by means of a suitable electric heater. Post and pre combustion CO₂ capture can be tested in this reactor;
- Micro-turbine tests: the gas turbine is fuelled with the syngas obtained from the mixing unit;
- Base-case tests: such tests are carried out with the syngas produced by the gasifier and diverted to the carbonator. Having completed the decarbonising process, the syngas undergoes a scrubbing process and then is fed to the micro-turbine. The syngas is previously compressed up to 600,000 Pa by means of a compressor and mixed with steam, as required by the micro-turbine combustion chamber;
- Regeneration tests: the carbonator is operated in regeneration mode by firing a number of suitable burners placed at the bottom of the reaction chamber. Auxiliary methane burners are located in the wind-box to heat the previously exhausted sorbent up to the calcination temperature in the start-up phase.

The coal gasification unit (500 kW_{th}) is mainly composed of a steam/oxygen-blown, BFB (bubbling fluidised bed) gasifier and an over-bed coal feeding system (see Fig. 1). The gasifier has a variable cross section area: the smallest, rectangular (0.38 × 0.36 m) surface

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