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Development of a regenerative reformer for tar-free syngas production in a steam gasification process

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HIGHLIGHTS

- Regenerative tar reforming is investigated numerically and experimentally.
- Over 99% of light and heavy tars are reformed to gas.
- 30% O_2 reduction compared to conventional oxidation reformer can be achieved.

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ABSTRACT

A novel reformer using highly efficient heat regeneration for tar-free syngas production is developed and its performance demonstrated in a pilot-scale plant using steam gasification. Basic design parameters of the regenerative tar reformer, namely residence time and amount of oxidant are determined based on numerical results. It has been predicted that good performance could be achieved at an operation temperature about 1573 K, the residence time exceeding 4 s and an oxidant addition of 12% of the syngas flow rate. The regenerative tar reformer so designed shows stable operation. Over 99% of light and heavy tars are reformed to gas in the case of 11.3% oxygen addition to syngas. Further it is seen that a reduction of oxygen consumption more than 30% compared to a conventional oxidation reformer can be achieved. The formation of a high temperature zone and good mixing of syngas and oxygen has a strong influence on the tar reforming efficiency.

1. Introduction

Gasification of solid feedstock for syngas production or heat and power generation increasingly attracts interest all over the world [1–3]. In order to advance environment-friendly technology, many techniques of coal and/or biomass gasification have been investigated for the best usage of the fuel [4,5]. Steam gasification in a fluidized bed gasifier [6–9] generally yields a higher cold gas efficiency compared to gasifiers working under high temperature and pressure. Twin IHI gasifier called TIGAR has been developed by the authors [10] over the years. TIGAR can produce syngas of high calorific value using steam gasification in the circulating fluidized bed. Basically, disadvantage of low temperature gasification is a presence of tar in syngas, which causes some troubles in the latter process. In addition, the overall cold gas efficiency of the process will decrease if the tar cannot be reformed appropriately. It is, therefore, necessary to realize a high conversion of the tar to gases. One method to crack the tar is the use of catalysts [11,12]. In this system, the tar is generally reformed at temperatures of about 900 °C or less; therefore, the cold gas efficiency after the reformer which is basically decreased by the reforming is about the same or higher compared to the efficiency before the reformer. However, guarantee of long-term durability of the catalyst sometimes becomes an issue that needs to be solved due to sulfur poisoning and time degradation. Another basic method for tar conversion is partial oxidation reforming with high temperature combustion. This method can crack the tar stably and continuously within a residence time of a few seconds (from 1.6 to 6 s) [13–15]. On the other hand, there is the problem of reducing the total cold gas efficiency due to consumption of syngas, when large amounts of oxidant are used [16]. Thus, a novel tar reformer using the technology of highly efficient heat regeneration is developed in this study.

The combination of regenerative burners [17,18] forms a heating system which allows self-heat recovery of exhaust gas in the combustion process. Cyclic regenerative combustion operates on the principle of short-term heat storage using ceramic heat exchangers. The flow direction is periodically changed by switching valves. Owing to reduction of fuel consumption and NO_x emission, this system has been specifically adopted and popularized in

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high temperature furnaces. However, to the authors' knowledge, the regenerative technique has not been applied to tar reforming yet. The main objective of this study is to design and show the performance of a regenerative tar reformer, which allows a decrease of oxygen supply and thereby an increase in cold gas efficiency.

2. Numerical simulations

2.1. Simulation methods

Basic design parameters of the regenerative tar reformer have been determined based on numerical results before starting with the detailed engineering. For this purpose two commercial software packages, CHEMKIN-Pro [19] and FLUENT 15.0 [20], have been used (conditions of simulation model are shown in Table 1). In a first step, the relationship between residence time in the reactor and tar reforming efficiency with reactor temperature as parameter is investigated by perfectly stirred reactor (PSR) calculations using detailed chemistry including tar reforming reactions [21]. The results reveal the necessary temperature which has to be provided for effective tar reforming. Then, the amount of oxygen to achieve the temperature obtained by PSR is estimated from 3D-calculations with simplified gas combustion chemistry without tar reaction [22].

2.1.1. PSR calculations

Syngas is supplied with the preheating temperature of 1443 K by heat regeneration as shown in Table 1. Contrary to general applications of regenerative burners, not oxygen but syngas is preheated here, since the volume of syngas is much larger than the volume of oxygen needed for partial oxidation. Syngas composition and amount of tar are set with reference to experiments performed in the TIGAR pilot facility using woody biomass. Operation of the TIGAR pilot plant was very stable and changes in syngas composition were less than 3% during three days continuous operation. Therefore, the average concentration is used in the calculations presented here. The tar modeling [21] does not include all kinds of actual tar components measured in the experiment; a number of heavier tars is neglected. Hydrocarbons with aromatic ring numbers from 1 to 4 are considered. Therefore, light tars measured in the experiment (6 major kinds of tar with aromatic ring number from 1 to 3) and heavy tar (approximated by pyrene in the amount such that the weight is the same as the total weight of all heavy tars in the experiment) are defined as input data in the PSR simulation. In the outlet of PSR, all hydrocarbons with aromatic rings are regarded as unreformed tar components. The amount of oxygen addition is expressed by the volumetric ratio R_{O2} of oxygen to syngas (Eq. (1)). In addition, tar reforming efficiency is expressed by Eq. (2).

$$R_{O2} = \text{volume of oxygen/volume of syngas (wet)} * 100\% \quad (1)$$

$$\eta_{tar} = (1 - \text{total mass of tar at outlet/total mass of tar at inlet}) * 100\% \quad (2)$$

2.1.2. Steady 3D-calculations

Steady 3D-calculations are performed to confirm the temperature distribution in the reactor. Note, that in practice, the temperature profile may be rather unsteady due to periodically changing the flow direction as explained in Section 3. However, simply steady calculations are done here for two reasons. The unsteady behavior is likely to continue for only a short time compared to the switching period. Secondly, the purpose of the calculation is to find the amount of oxygen (R_{O2}) necessary to obtain a mean temperature, which is close to the reactor temperature required from the PSR studies. Syngas inlet temperature and syngas composition are almost the same values as in the PSR studies. However, to reduce the computation time of the 3D-calculations, other hydrocarbons except for tar such as CH_4 and C_2H_6 are expressed as C_nH_m where n and m are estimated from the change of standard enthalpy of formation for the actual gas composition. Although tar is included in the initial syngas composition, tar cracking reactions are not implemented in this 3D-calculation. Radiation is modeled by the discrete ordinate method; heat loss to the boundaries is evaluated by solving 1D-heat conduction through the walls. Minimal mesh size in the 3D-calculations is smaller than one third of oxygen nozzle diameter and total amount of cells is more than 2 million in order to simulate the actual mixing and reacting processes.

2.2. Numerical results

2.2.1. PSR results

Fig. 1 shows the relation between residence time in the reactor and η_{tar} with reactor temperature as parameter. In the lowest

Table 1
Conditions of simulation model.

Reaction condition	Software	CHEMKIN-Pro	FLUENT 15.0
	Flow	Unsteady	Steady state
Inlet gas condition			
Gas composition (wt%)			
	H_2	1.78	
	H_2O	32.59	
	CO	29.63	
	CO_2	18.00	
	C_mH_n	CH_4: 5.92. C_2H_4: 3.39, C_3H_8: 0.27	9.58
	N_2	6.34	
	Tar	2.08 (with reaction)	2.08 (without reaction)
Temperature (K)		1443	
Temperature condition		Constant	1D heat conduction
Reaction model		Detailed chemistry including tar reforming reactions (1804 reactions with 158 species)	Reduced chemistry (4 reactions with 7 species) $H_2 + 1/2O_2 \Leftrightarrow H_2O$ $CO + H_2O \Leftrightarrow CO_2 + H_2$ $C_mH_n + mH_2O \rightarrow mCO + (n/2 + m)H_2$ $C_mH_n + (m/2)O_2 \rightarrow mCO + (n/2)H_2$ $m = 1.276$, $n = 4.010$
Radiation model		–	Discrete ordinate
Emissivity		–	0.5
Thermal conductivity (W/mK)		–	2.18
Reactor surface temperature (°C)		–	50

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