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ScienceDirect
Energy Procedia 75 (2015) 246 – 251
Energy Procedia

The 7th International Conference on Applied Energy – ICAE2015

Development of a novel reformer for tar-free syngas production

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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 sec 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 has a strong influence on the tar reforming efficiency.

Peer-review under responsibility of Applied Energy Innovation Institute
Keywords: Tar reformer; regeneration; steam gasification

1 Introduction

Gasification of solid feedstock for syngas production or heat and power generation increasingly attracts interest all over the world [1, 2]. Steam gasification in a fluidized bed gasifier 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 [3] over the years. TIGAR can produce syngas of high calorific value using steam gasification in the circulating fluidized bed. However, the presence of tar from the process of low temperature gasification can cause pipe blockage and also reduces the overall cold gas efficiency of the process. It is, therefore, necessary to realize a high conversion of the tar to gases. One of the basic methods for tar conversion is partial oxidation reforming with high temperature combustion. 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 [4]. Thus, a novel tar reformer using the technology of highly efficient heat regeneration is developed in this study.

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1876-6102
Peer-review under responsibility of Applied Energy Innovation Institute
doi:10.1016/j.egypro.2015.07.319

The combination of regenerative burners [5] 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 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 methods and results

Before starting with the detailed engineering, basic design parameters of the regenerative tar reformer have been determined based on numerical results. For this purpose two commercial software packages, CHEMKIN-Pro and FLUENT 15.0, have been used (conditions of simulation model are shown in Table 1). In a first step, the relationship between residence time and reactor temperature is investigated by perfectly stirred reactor (PSR) calculations using detailed chemistry including tar [6]. The results reveal the necessary temperature which has to be provided for effective tar reforming. Then, the amount of oxygen to achieve this temperature is estimated from 3D-calculations with simplified gas combustion chemistry [7]. All hydrocarbons with aromatic rings are regarded as tar components. In the 3D-calculations, radiation is modeled by the Discrete Ordinate method; heat loss to the boundaries is evaluated by solving 1D-heat conduction through the walls. Syngas is supplied with the preheating temperature of 1170 K; composition and amount of tar are set with reference to experiments performed in the TIGAR pilot gasification facility using woody biomass. The amount of oxygen addition is expressed by the volumetric ratio R_{O2} of oxygen to syngas (Equation 1). Figure 1 shows the relation between residence time in the reactor and tar reforming efficiency (Equation 2) with reactor temperature as parameter. It is seen that the residence time necessary to accomplish almost complete tar reforming becomes shorter, when the reactor temperature is increased. Thus, a smaller reactor can be realized with higher reactor temperature. However, a high temperature must be obtained by oxidizing syngas, which diminishes the energy efficiency of the gasification process.

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

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

Table 1. Conditions of simulation model

Reaction condition		Software	CHEMKIN-Pro	FLUENT 15.0
		Flow field	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	9.58	
		N_2	6.34	
		Tar	2.08	
	Temperature, ℃		1170	
Temperature condition			Constant	1D heat conduction
Reaction model			Detailed chemistry [6]	Reduced chemistry [7]
Radiation model			-	Discrete Ordinate
Emissivity			-	0.5
Thermal conductivity, W/mK			-	2.18

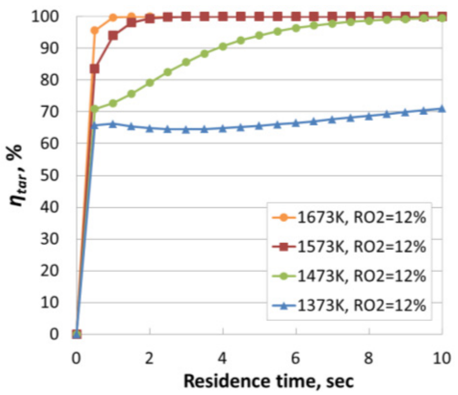


Fig. 1. Tar reforming efficiency vs. residence time.

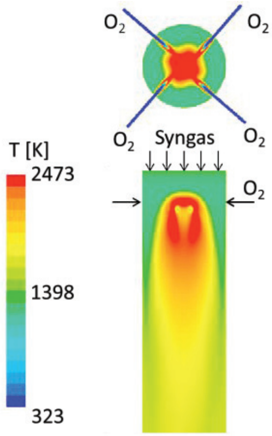


Fig.2. Temperature distribution in the mixing zone for reactor with tangential oxygen supply.

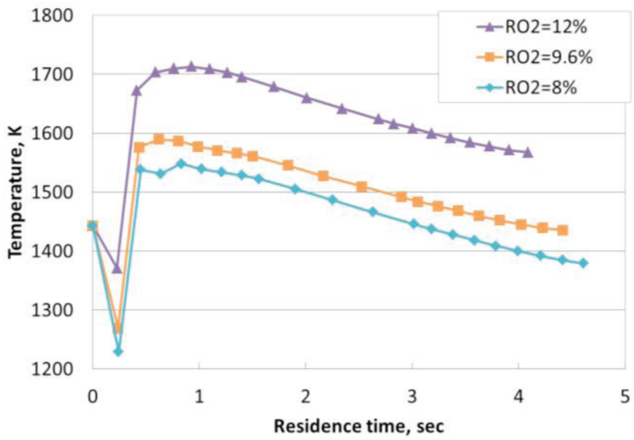


Fig.3. Cross-sectional mean gas temperature vs. residence time with amount of oxygen supply as parameter.

Therefore, the reactor temperature cannot be raised arbitrarily and is set to 1573 K here. In this case, complete tar reforming can be achieved, when the residence time is longer than 4 sec (Fig.1).

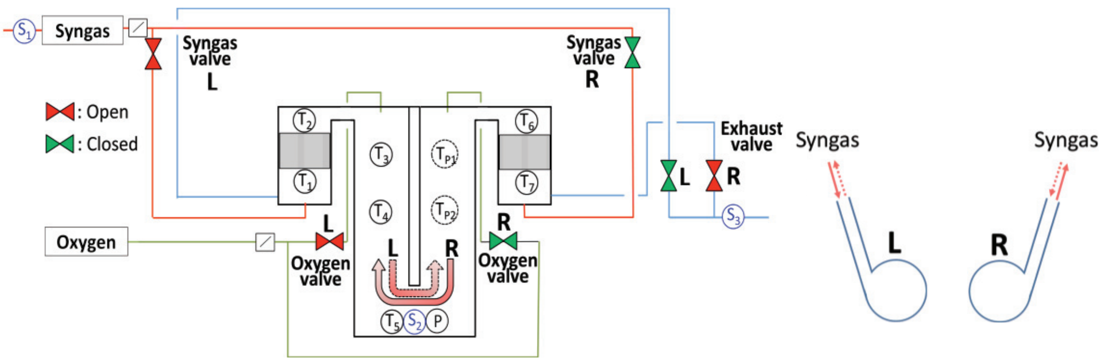
Steady 3D-calculations are performed to confirm the temperature distribution in the reactor. Note, that in practice, the temperature profile may be rather unsteady due to periodically changing the flow direction as explained in section 3. However, simply steady calculations are done here for two reasons. The unsteady behaviour 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. 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 due to the partial oxidation reaction. Oxygen is injected to the reactor tangentially from four nozzles as indicated at the top of Fig. 2. It is seen, that a circular (cylindrical) temperature field is formed due to the slightly swirling flow induced by the oxygen nozzles, which is supposed to enhance the mixing of oxygen with syngas. Figure 3 shows the mean gas temperature versus residence time, where residence time has been evaluated as distance of cross-section from the top divided by the mean axial flow velocity. Note, that mean gas temperature and mean flow velocity are calculated as mass-weighted averages over the cross-section. The results show that the temperature first abruptly decreases due to low-temperature oxygen injection, but then quickly recovers because of exothermic reaction. After a local maximum slightly behind the oxygen injection, the temperature gradually decreases with time (distance), presumably due to heat loss to the wall. It is found that the reactor temperature exceeds 1573K over the entire length in the case of R_{O2}=12%.

3 Experimental methods and results

The numerical results (Fig. 2) revealed that high temperatures are achieved around the oxygen streams. It is also confirmed (but not shown in the paper) that oxygen is completely consumed downstream. However, a relatively large area of low temperature exists near the top and upper side walls due to inadequate mixing. Therefore, in order to improve mixing the regenerative tar reformer has been designed with opposite supply streams, i.e. the swirl flow has been changed from oxygen to syngas as illustrated in Fig. 4. After being preheated by heat storage units, the strong swirling flow of syngas, which has an approximately ten times larger volume than oxygen, enters the reformer where it then mixes with the choked oxygen flow injected from 4 holes on the top of the reformer. Note, that the reactor is U-shaped in the present reformer to prevent a shortcut from inlet to outlet.

Reforming performance is checked by separating a portion of the syngas produced in a pilot gasification plant to the new reformer. The pilot plant has a feeding capacity of 6 ton/day. 30 Nm^3/h of syngas is reformed here. The heat storage units are honeycomb structures, which have been selected from 4 different types by evaluating the effect of heat transfer area on heating performance using basic heat transfer theory, and by doing preliminary tests with the actual reformer. Four rotary valves are used to switch the flow direction of syngas in intervals of 30 or 60 sec. Soot blowers operated with nitrogen are mounted above and below the honeycombs. Gas is sampled at three locations named S_1-S_3. In the present regenerative tar reformer, a little amount of gas is slipping, meaning that entering gas immediately goes out of the reactor through the same valve when the flow direction is switched. Therefore, the result of η_{tar} at station S_3 cannot always be 100%, even if the efficiency is 100% at S_2. This problem can be solved by using a three-tower-type reactor, as is already confirmed in practical use [8]. In this study, the purpose is to confirm the tar reforming efficiency for various amounts of oxygen; thus, only S_1 and S_2 are used for the evaluation of tar reforming performance. The total residence time of the reactor is 8 sec in case of reaction temperature of 1573K and syngas flow rate of $30Nm^3/h$; thus, the residence time up to S_2 is about 4 sec.

Figure 5 shows trend data of the regenerative tar reformer. The switching cycle and R_{O2} are 60 sec and 11.3%, respectively. Gas temperature on right and left sides of the reactor periodically changes due to the flow switching. Temperature T_5 at the central position in the reactor is almost constant.



(a) (b)

Fig. 4. Schematic of regenerative tar reformer. (a) Diagram of the reformer. (b) Structure of syngas inlet/outlet.

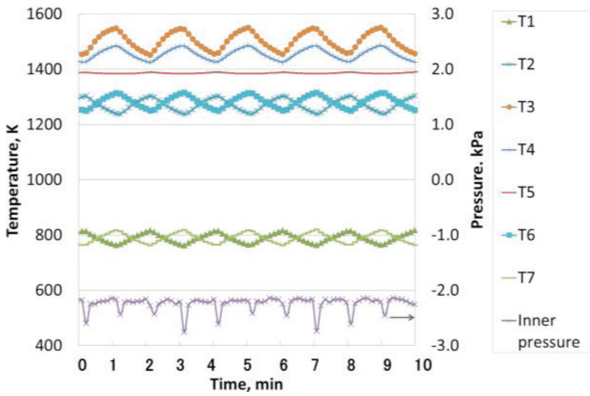


Fig. 5. Trend data of regenerative tar reformer.

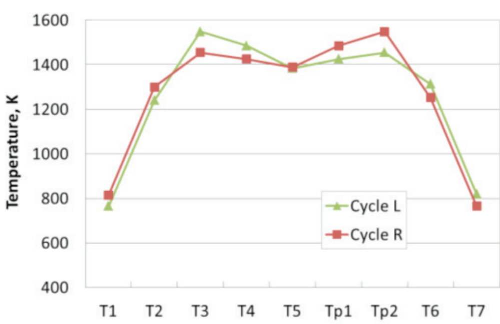


Fig. 6. Temperature distribution in flow direction.

The pressure in the reactor is stable in spite of periodic movement of switching valves, oxygen injection and soot blowing. This is the result of a well-considered control sequence, which takes the delay of opening-closing timing of the electric valves into account. The temperature distributions in flow direction for cycles L and R are shown in Fig.6. Temperature T_{P1} and T_{P2} are not measured but assumed to be the same as T_3 and T_4 of the reverse cycle (Fig.4). Although the inlet temperature of about 800 K is slightly lower than expected due to heat losses of the separated line between pilot plant and regenerative tar reformer, temperature augmentation of syngas by the heat storage units worked as predicted. Also the heat loss of the reformer itself is predictable. A noteworthy difference from the calculated temperature distribution (Fig. 3) is the shape of the profile, which is M-shaped in Fig.6, presumably due to the wall temperature staying high from the previous cycle. It can be said that this heat recirculation effect by the wall is one of the merits of the regenerative reactor.

Figure 7 shows the relationship between R_{O2} and η_{tar}. The measurement of heavy and light tars is separated by the condensation temperature. Tar which condensed above 150 degree C is measured by weight and regarded as heavy tar, while that condensing below 150 degree C is regarded as light tar and detected by GCMS. The change of the sum of both tars and reforming efficiency with R_{O2} are shown in Fig.7. It is readily seen that η_{tar} increases with R_{O2} and more than 99% of light and heavy tars are reformed to gas in the case of R_{O2}=11.3%. This result indicates that a 30% reduction of oxygen supply compared to the results of a conventional oxidation reformer obtained by the authors can be achieved by the regenerative tar reformer. However, the temperature distribution (Fig.6) to accomplish the tar reforming efficiency of more than 99% is lower than the numerical prediction for the case of R_{O2}=12% (Fig.3). The main reason for this discrepancy is supposed to be caused by the different supply methods. In addition, there are a few experimental conditions which differ from the numerical conditions, e.g. the addition of Argon by 15vol% (dry) to the syngas for gas analysis reasons.

Therefore, another calculation is performed using the experimental conditions as input data. Figure 8 reveals that the inversion of oxygen and syngas supplies results in a wider high temperature zone compared to Fig. 2, owing to improvement of mixing between syngas and oxygen. The simulated temperature shows good agreement with the experimental data (Fig.9). The maximal temperature exceeds 1650 K. In addition, the efficiency as calculated for PSR with detailed chemistry using the temperature distribution shown in Fig. 9 yields 82%, although η_{tar} is only 65% in case of reaction temperature less than 1400K (Fig.2). Therefore, it can be concluded that the formation of a high temperature zone is important for good performance of tar reforming, even though the residence time in this zone may be short.

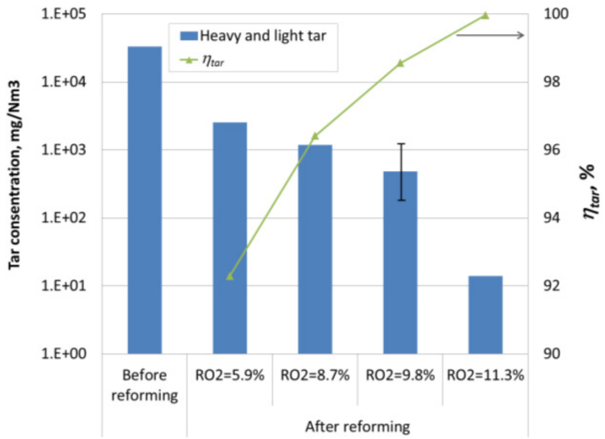


Fig.7. Tar reforming efficiency as experimentally verified in regenerative reformer.

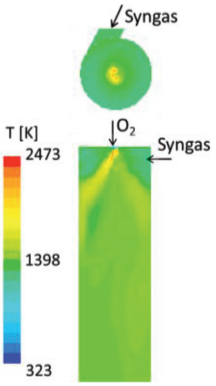


Fig.8. Temperature distribution in the mixing zone for reactor with tangential syngas supply.

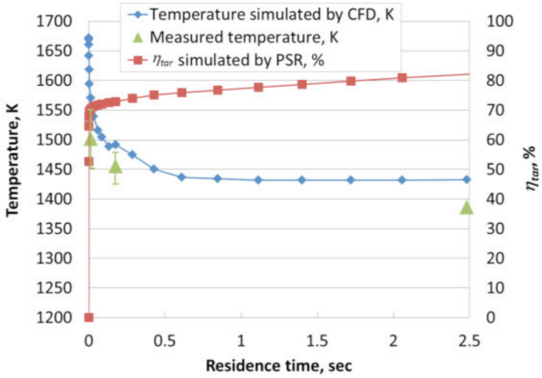


Fig.9. Temperature distribution and tar reforming efficiency. (comparison of experiment with simulation)

4 Conclusions

Regenerative reforming of light and heavy tars contained in syngas by steam gasification was investigated. In a first step, basic design parameters of the reformer such as appropriate size of the reactor and amount of oxidant were determined by using numerical predictions. The results showed that the appropriate average temperature in the reactor, necessary minimal residence time and oxygen flow rate are 1573 K, 4 sec and 12% of the syngas volume, respectively. Experiments with the new regenerative tar reformer proved stable operation and reforming efficiency exceeding 99% at an oxygen flow rate of 11.3%. This result also proved that the concept of regenerative reformer yields higher system efficiency, because the same high reforming efficiency as obtained with a conventional reformer could be achieved with 30% less oxygen consumption. It was found that the formation of a high temperature zone has a strong effect on high reforming efficiency.

Acknowledgements

This study has been performed as part of the project “SNG Production from Woody Biomass Using Gasification Process”, sponsored by the New Energy and Industrial Technology Development Organization (NEDO).

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