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Integration of gasoline prereforming into autothermal reforming for hydrogen production

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Abstract

An integrated process for hydrogen production which couples gasoline prereforming and autothermal reforming (ATR) over nickel-based catalysts was investigated using stainless steel fixed-bed reactors. Meanwhile the integrated process was compared with the gasoline ATR process without prereforming. The results indicate that in the gasoline ATR process without prereforming, the nickel-based ATR catalyst deposited with coke after short reaction time-on-stream under the following working conditions ($T = 770$ °C, $P = 5.0$ bar, steam-to-carbon feed ratio (S/C, mol/mol) of 2.7, oxygen-to-carbon feed ratio (O_2/C) of 0.5 and gas hourly space velocity (GHSV) of 28,000 ml g^{-1} h^{-1}). Quite the contrary, in the integrated process, almost 100% gasoline conversion and 99.4% selectivity to hydrogen were obtained and maintained well under similar working conditions during 100 h reaction time-on-stream. Actually, almost coke-free operation of the process was achieved, which was confirmed by scanning electron microscopy (SEM) and O_2-TPSR characterizations of the used ATR catalyst. Reformate that contains no light alkenes or other higher hydrocarbons could be obtained from the ATR reactor. In addition, the nickel-based prereforming catalyst prepared by a coprecipitation method had high catalytic activity and promising stability.

Keywords: Gasoline; Prereforming; Autothermal reforming; Coke deposition; Nickel-based catalyst

1. Introduction

Hydrogen is forecast to become one of the two principal energy carriers (with electricity) in the 21st century [1]. The fuel cell is favorable for its high fuel-to-electricity conversion efficiency (40–60%) and being environmentally benign [2]. H_2 is the ideal fuel for fuel cells, the lack of infrastructure for H_2 distribution and delivery makes gasoline an ideal fuel source for H_2 production onboard or onsite, due to its worldwide availability of distribution infrastructure and high energy density [3]. Options for converting hydrocarbons into H_2 include steam reforming (SR), catalytic partial oxidation (CPO) and ATR (also called "oxidative steam reforming" sometimes), which combines the endothermic SR reactions and exothermic CPO reactions. ATR offers advantages such as less complicated reactor design, lower reactor weight and wider available construction materials over endothermic SR or exothermic CPO [4], therefore, H_2 production from the ATR of hydrocarbons has been investigated widely. Moon et al. [5] investigated the ATR of isooctane with direct mixing of feedstock at around 700 °C and found that higher reaction GHSV resulted in lower selectivity to H_2 and CO_2. Ayabe et al. [6] investigated the ATR of propane at working conditions of $T = 800$ °C, $C_3H_8/O_2 = 2.0$ and S/C = 1.5 over nickel-based catalysts. When the reaction was performed at GHSV of 57,000 h^{-1}, ethylene was found in products. Even using micro-channel reactor to perform the ATR of propane, considerable content of alkenes were still detected in products under high reaction GHSV conditions [7]. Alkenes in reformate from the ATR reactor are unacceptable, for their slip into the downstream processing units such as water gas shift (WGS) and CO preferential oxidation (PROX) would result in catalyst

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deactivation in those units [8]. Thus, when complex fuels like gasoline or diesel are used as feedstock, reaction temperatures higher than 850 °C are necessary to achieve complete fuel conversion (into C_1 species) over nickel-based reforming catalysts. However, little attention has been paid to the non-catalytic gas phase reactions during preheating the ATR reaction feedstock and their influences on the over all gasoline ATR process using nickel-based catalysts and stainless steel reactors. Actually, reactions like cool flame, pyrolysis and steam cracking take place before the catalytic ATR reactions during preheating the feedstock mixture [9]. Thus, the fuel actually reformed in the ATR catalyst bed are a spectrum of lighter hydrocarbons generated from those homogeneous gas phase reactions. Alkenes are the thermodynamically favorable products of those reactions and they have much higher coke deposition risk on the nickel-based ATR catalysts. Thus, the main obstacles to H_2 production from the gasoline ATR over nickel-based reforming catalysts are catalyst deactivation by coke deposition and the slip of higher hydrocarbons into the down-stream processing units.

Prereforming has been successfully integrated in the process of naphtha or natural gas steam reforming for syngas or H_2 production, during which higher hydrocarbons are converted into C_1 components at 400–550 °C irreversibly by the following reactions [10].

$$C_nH_m + nH_2O = nCO + (n + (m/2))H_2 \quad (1)$$

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O \quad \Delta H^\circ_{298} = -206\,\text{kJ/mol} \quad (2)$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \quad \Delta H^\circ_{298} = -41\,\text{kJ/mol} \quad (3)$$

However, its integration into the process of H_2 production from gasoline ATR has not been studied yet, so this work aims to study the influences of prereforming introduction on the overall gasoline ATR process, particularly on the coke deposition aspects. The results indicate that by using such an integrated process, gasoline could be efficiently converted into H_2 over nickel-based reforming catalyst. The products from prereformer could be heated to temperatures up to 800 °C without coke deposition risk. Higher hydrocarbons in the reformate from the ATR reactor could be eliminated in the integrated process. Those combined characteristics of the integrated process make it potential application to small-scale onsite H_2 production unit using the worldwide gasoline infrastructure.

2. Experimental

2.1. Catalyst preparation and characterization

The detailed procedures for preparing the prereforming catalyst by a coprecipitation method and characterization were described in a previous report [11]. The composition of the prereforming catalyst was 50.0 wt.% NiO, 18.0 wt.% La_2O_3 and balance alumina. The ATR catalyst was prepared by incipient-to-wetness impregnation. The nickel loading was 8.0 wt.% and lanthana 2.0 wt.%. Commercial 23.0 wt.% MgO modified α-Al_2O_3 support (Jiangsu Yixing Catalyst Support Plant) with a BET surface area of 0.5 $m^2\,g^{-1}$ were impregnated with catalyst precursors $Ni(NO_3)_2{\cdot}6H_2O$ and $La(NO_3)_3{\cdot}6H_2O$, dried at 110 °C for 12 h and calcined at 850 °C for 4 h.

The morphologies of the used ATR catalysts were characterized using JSM-6360 LA scanning electronic microscope (SEM). Powdered specimens were spread on the SEM slabs and were sputtered with gold. To detect if there were other kinds of carbon other than filamentous one deposited on the ATR catalyst in the integrated process, oxygen temperature programmed surface reactions (O_2-TPSR) of the used ATR catalysts were carried out in a quartz micro-reactor using Micrometrics Autochem 2910 connected to a mass spectrometer Omnistar^{TM}. About 100 mg catalyst was packed in the reactor installed in a furnace with a temperature programmed controller, the sample was first exposed to Ar gas with a flux of 30 $cm^3\,min^{-1}$ and heated from room temperature (RT) to 400 °C at 10 °C min^{-1}, then it was cooled down to RT. Then Ar was switched to a mixture of 2% O_2–He gas stream (30 $cm^3\,min^{-1}$). After stabilization, the sample was heated at a rate of 10 °C min^{-1} up to 900 °C. The burnt-off products were analyzed by the online mass spectrometer.

2.2. Catalytic performance measurement

The experiments were performed in tubular fixed-bed reactors on a home-made setup, as shown in Fig. 1. The reactors (ID = 16 mm and OD = 22 mm) were made of 316 stainless steel (Shandong Jinxiang Stainless steel Plant). For the prereforming reaction, 1.0 g catalyst was used as particles with the size of 420–630 μm without dilution. The catalyst was pre-reduced in a gas stream of 30% H_2–Ar (50 $cm^3\,min^{-1}$) at 600 °C for 2 h. The thermocouple used to control the reaction temperature was placed at the center of the catalyst bed. The reaction temperature was fixed at 480 °C and the pressure at 5.0 bar with varying space velocity from 40,000 to 168,000 ml $g^{-1}\,h^{-1}$. The simulated gasoline has a composition of 74.0 wt.% isooctane, 20.0 wt.% xylene, 5.0 wt.% cyclo-hexane and 1.0 wt.% 1-octene (Shanghai Medical and Reagent Incorp.), the purity of chemicals are higher than 99.0%. The average molecular formula was $C_{7.86}H_{15.58}$ and the average molecular weight 109.9 g mol^{-1}. Deionized water and simulated gasoline were metered and delivered to the evaporator by HPLC pumps. The pressure of reaction was maintained by a backpressure regulator connected with a precise pressure gauge to read the pressures. The products in reformate were monitored by an on-line Varian-CP 3800 gas chromatograph equipped with FID (flame ionization detector) and TCD (thermal conductivity detector).

In the integrated process, the reactions were carried out in such a way that reformate gas from the prereformer was fed directly to the ATR reactor, while all the air feed was introduced directly to the ATR reactor, as shown in Fig. 1. The prereformer was packed with 4.0 g catalyst and the ATR reactor with 8.5 g catalyst. The activation of the prereforming and ATR catalysts was performed at 600 and 800 °C in a 100 $cm^3\,min^{-1}$ 30% H_2–Ar gas flux for 3 h, respectively. In the stability test of the process, reaction GHSV for prereforming catalyst was

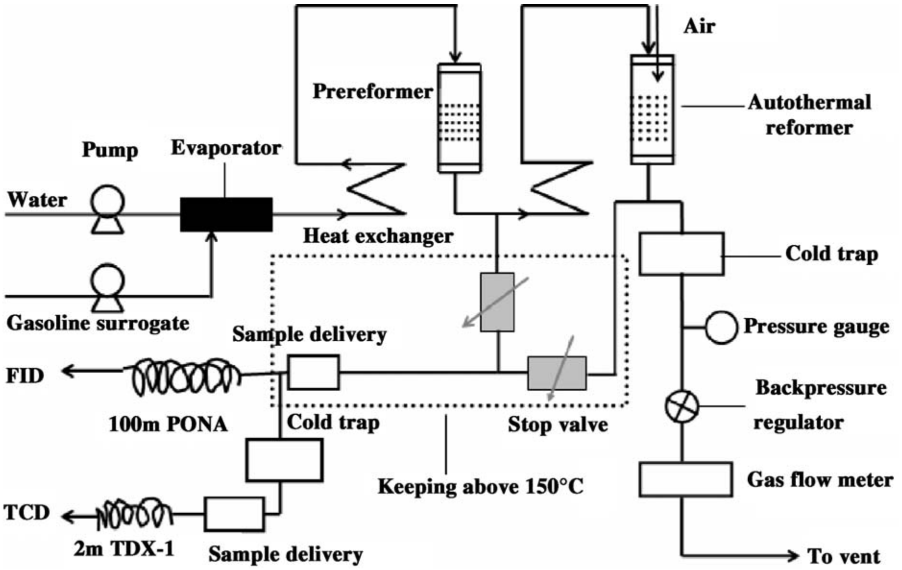

Fig. 1. Schematic diagram of setup for reactions and analysis.

40,000 ml g^{-1} h^{-1}, and for the ATR catalyst 28,000 ml g^{-1} h^{-1}. In the gasoline ATR process without prereforming, the feed rate, catalyst amount and the working conditions were kept similar. The analysis of reformate from the prereformer, or those from the ATR reactor was also performed by the online Varian-CP 3800.

In this work, the conversion of gasoline is defined as the part (carbon based) of the fuel converted into C_1 species, possible higher hydrocarbons in reformate were supposed to be ‘‘unconverted’’. Thus, the conversion of gasoline could be calculated using the concentrations of species in reformate according to:

$$\text{Conversion}\,(\%) = \frac{[\text{CO}] + [\text{CO}_2] + [\text{CH}_4]}{[\text{CO}] + [\text{CO}_2] + [\text{CH}_4] + n\sum \text{C}_n\text{H}_m} \times 100 \tag{4}$$

The selectivity to H_2 (S_{H_2}) is defined as the part of hydrogen atomic numbers in all the products except steam divided by hydrogen atomic numbers in H_2 and calculated according to:

$$S_{H_2}(\%) = \frac{[\text{H}_2] \times 2}{[\text{H}_2] \times 2 + 4 \times [\text{CH}_4] + m\sum \text{C}_n\text{H}_m} \times 100 \tag{5}$$

where C_nH_m are the possible hydrocarbons other than methane in reformate. In the process of data reduction, carbon, oxygen and hydrogen balance was calculated using the inert nitrogen as an internal standard; data with balances over 98% are adopted.

3. Results and discussion

3.1. Gas phase reactions in the ATR feedstock during preheating

Higher hydrocarbons tend to react with air (oxygen) more readily than methane does in the mixture of air (oxygen) and hydrocarbon fuels. In the process of H_2 production from the ATR or CPO of liquid hydrocarbons, cool flame reactions, pyrolysis and/or steam cracking may take place during preheating. For instance, Ciajalo and D’Anna [12], Ranzi et al. [9] investigated the influence of reaction temperature on the products selectivities during the low-temperature oxidation of stoichiometric *n*-heptane in a jet-stirred-flow reactor. Reactions under working conditions of $T = 347$ °C, $P = 10.0$ bar and residence time of 0.1 s resulted in 54.5% conversion of *n*-heptane and dominant (up to 80%) ketohydroperoxide product. The low temperature mechanism is a complex process involving propagation and chain branching, differing from high temperature oxidation mechanism significantly. At higher reaction temperatures, β-decomposition reactions of alkyl radicals prevail over the reactions with oxygen insertion into the hydrocarbon skeleton. Reactions under working conditions of $T = 547$ °C, $P = 10.0$ bar and residence time of 0.1 s brought on 77.6% of *n*-heptane conversion and predominant β-decomposition products such as conjugate alkenes. Our previous work on the partial oxidation of *n*-heptane also found that reactions under the working conditions of $T = 400$ °C, $O_2/C = 0.65$ and residence time about 0.1 s resulted in 25.9% *n*-heptane conversion into C_1 species [13].

The influence of steam on those reactions were investigated in this work using isooctane as a model compound under the following working conditions ($T = 500$–630 °C, $P = 1.0$ bar, S/C = 2.0, $O_2/C = 0$ or 0.3 and residence time about 0.9 or 0.6 s), and the results are shown in Fig. 2. Even with about 90% (mol) steam in feedstock, alkenes could be generated at about 500 °C. In the mixture of steam and isooctane, reactions at 500 °C resulted in less than 1.0% isooctane conversion, while air introduction promoted the conversion of isooctane to 8.5% but slightly decreased selectivity to alkenes due to the formation of CO_x. Higher reaction temperatures also boosted the fuel conversion and selectivity to alkenes, for instance, reactions under working conditions of $T = 630$ °C brought on 58.0%

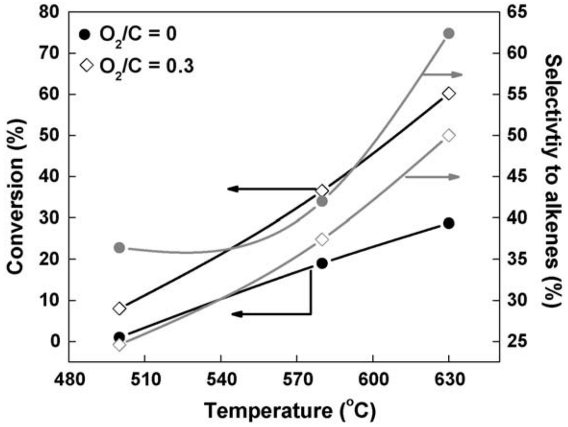

Fig. 2. Conversion of isooctane and selectivity to alkenes in the blank reactions of isooctane, air and steam at $P = 1.0$ bar and $S/C = 2.0$ during preheating (residence time $t = 0.9$ s at $O_2/C = 0$ or $t = 0.6$ s at $O_2/C = 0.3$).

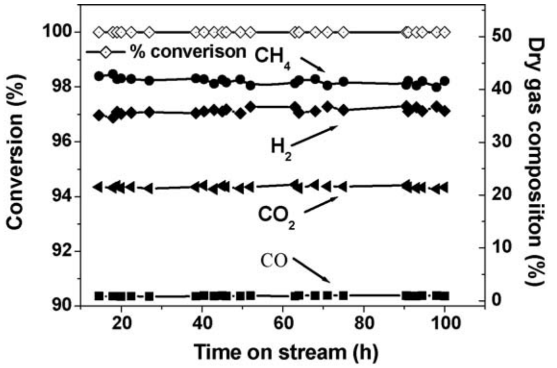

Fig. 4. Stability testing of the prereforming catalyst (working conditions: $T = 480$ °C, $P = 5.0$ bar, GHSV = 40,000 ml g^{-1} h^{-1}, $S/C = 2.7$ and $H_2/C = 0.1$).

isooctane conversion and about 50.0% selectivity to alkenes. Actually, preheating the mixture of air, isooctane and steam with the above mentioned composition at temperatures above 650 °C leads to over 90.0% oxygen conversion in the gas phase. So, in the gasoline ATR process without prereforming, gas phase reactions consume nearly all the oxygen feed, generating alkenes with high coke deposition risk. Thus, nickel-based catalysts were supposed unsuitable for application to the ATR of liquid fuels due to their higher coke deposition tendency than precious-metal based ones.

3.2. Catalytic performance of the gasoline prereforming catalyst

The performance of the prereforming catalyst was tested at $T = 480$ °C, $P = 5.0$ bar, $S/C = 2.7$ and GHSV = 40,000–168,000 ml g^{-1} h^{-1}, and the results are indicated in Fig. 3. The gasoline conversion maintained almost 100.0% within GHSV range of 40,000–144,000 ml g^{-1} h^{-1}. Even under working conditions of GHSV = 168,000 ml g^{-1} h^{-1}, 95.6% conversion of gasoline was obtained. The products selectivities showed substantially no change, suggesting that the reactions performed over the wide GHSV range are thermodynamically controlled and the steam reforming of hydrocarbons can proceed at a fast intrinsic reaction rate. The high activity of the catalyst may be related with the fine nickel oxide particle formed and lanthana incorporation into the takovite structure during the coprecipitation process [14]. Fig. 4 shows the results of stability test of the prereforming catalyst under working conditions of $T = 480$ °C, $P = 5.0$ bar, GHSV = 40,000 ml g^{-1} h^{-1}, $S/C = 2.7$ and $H_2/C = 0.1$. During the reaction time-on-stream of 100 h, the composition of products were stable and the conversion of gasoline maintained almost 100.0%. This suggests that the catalyst has good coke deposition resistance and promising stability. Prereforming converted the complex fuels into a much simpler methane rich gas stream irrespective of the hydrocarbons in the feedstock. Within prereforming reaction temperatures, reactions such as pyrolysis and steam cracking of higher hydrocarbons could be neglected; alkenes do not polymerize or interact with inert aromatics to form poly-aromatic hydrocarbons. This has several beneficial effects: (1) steam is a coke inhibitor, the catalyst makes the hydrocarbons convert into H_2 and C_1 species instead of coke; (2) the H_2 formed further inhibits the coke formation; (3) because heavy hydrocarbons react rapidly to give CH_4, H_2, and CO_x, the reformate from prereformer contains almost no higher hydrocarbons. This reformate stream can be fed to the high temperature (800 °C) reformer without the risk of forming coke, thus a higher throughput of the fuel processor could be predicted. However, prereforming is performed at much lower reaction temperatures than ATR reactions do, therefore, stringent requirement of sulfur content in fuels is necessary to reduce the deactivation of nickel-based catalysts due to sulfur poisoning.

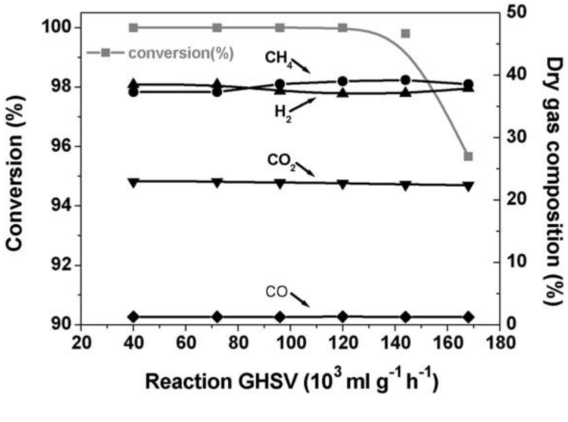

Fig. 3. The performance of prereforming catalyst at different reaction GHSV (working conditions: $T = 480$ °C, $P = 5.0$ bar, $S/C = 2.7$ and $H_2/C = 0.1$).

3.3. Gasoline autothermal reforming

CPO or ATR promises higher H_2 production capacity because of the reduced heat transfer limitation in contrast with SR. However, the pioneering work on the ATR of gasoline or diesel fuel surrogate for H_2 production carried out at Argonne National Laboratory indicated that obvious higher hydrocarbons slip took place even over precious-metal based monolithic catalyst under high reaction GHSV conditions [15]. The influence of reaction GHSV on the higher hydrocarbons slip in the ATR process without prereforming were also investigated in this work using isooctane as a model compound under working conditions of $T = 720$ °C, $P = 1.0$ bar, $O_2/C = 0.50$ and $S/C = 2.0$. It turned out that reactions at GHSV = 30,000 ml g^{-1} h^{-1} led to higher hydrocarbons concentration in reformate (dry-basis, *i.e.* steam was excluded in the calculation) of 0.6 g m^{-3}, which changed into 2.7 g m^{-3} at reaction GHSV of 60,000 ml g^{-1} h^{-1} and further increased to 40.0 g m^{-3} at GHSV of 120,000 ml g^{-1} h^{-1}. Higher hydrocarbons, particularly alkenes slip into the WGS and PROX units could bring on coke deposition risk [8], therefore, if the reformate from ATR reactor is to directly deliver to the WGS and PROX units without condensing and to make the downstream processing of CO by WGS and PROX proceed smoothly, the reformate should be free of higher hydrocarbons. Thus, on one hand, to make a fuel processor more compact, higher inlet temperature in ATR reactor is essential; on the other hand, higher preheating temperature promoted the gas phase reactions and higher coke and soot formation risk could be expected. The gasoline ATR process without prereforming over nickel-based catalysts could not solve this contradiction. However, prereforming introduction before the ATR reactor could make the gas stream fed to the ATR reactor almost free of higher hydrocarbons and a preheating temperature as high as 800 °C could be applied without risk of coke deposition.

In the gasoline ATR process without prereforming, the reactions was performed under working conditions of $T = 770$ °C, $P = 5.0$ bar, $S/C = 2.7$, $O_2/C = 0.5$ and GHSV = 28,000 ml g^{-1} h^{-1} for reaction time-on-stream of about 8 h, and the reformate composition change versus reaction time-on-stream is indicated in Fig. 5(a). At the beginning of the reactions, no higher hydrocarbons was detected in products. But conversion of gasoline decreased to about 99.5% after about reaction time-on-stream of 5 h, suggesting that the catalyst could not maintain its activity for long-term operation. The ATR catalyst performance versus reaction time-on-stream of 100 h in the integrated process is shown in Fig. 5(b). During the test period, no higher hydrocarbons were detected (detection limit of the FID is about 1 ppm) in reformate and almost 100.0% conversion of gasoline and 99.4% selectivity to H_2 were achieved and maintained well. However, the reformate compositions from the ATR reactor in the two processes, along with the thermodynamically controlled product composition are very close, as shown in Table 1. The results indicate that the introduction of prereforming does not change the equilibria of the reactions, but it does alter the concrete reaction routes.

Table 1
The composition of reformate experimentally determined and thermodynamic equilibrium composition (working conditions: $T_{outlet} = 770$ °C, $P = 5.0$ bar, GHSV = 28,000 ml g^{-1} h^{-1}, $S/C = 2.7$ and $O_2/C = 0.5$)

	Composition (%)				
	H_2	CO	N_2	CH_4	CO_2
Theoretical	35.8	8.8	41.9	0.1	13.4
Experimental					
ATR with prereforming	36.2	7.7	41.7	0.1	14.3
ATR without prereforming	35.5	8.1	42.3	0.1	14.0

The morphologies of used ATR catalysts characterized by SEM are shown in Fig. 6. Coke deposition on the ATR catalyst can be clearly seen after reaction time-on-stream of 8 h in the ATR process without prereforming, which is demonstrated in Fig. 6(a). Part of the active metallic surface was covered by the filamentous carbon. In this process, gas phase reactions during preheating tend to consume all the oxygen in the feedstock, bringing on a complex gas stream with high content of alkenes. Thus, the actual reactions in the ATR catalyst bed are the steam reforming of those products from gas phase reactions. While filamentous carbon formation rate is closely related to the hydrocarbon structures in the process of steam reforming [16]; it decreases in the order of acetylenes, alkenes and paraffins. Coke deposition was found not only on the ATR catalyst but also on the stainless steel reactor wall. The mechanism of coke deposition on the stainless steel reactor wall involves two steps. Firstly, there is a catalytic phase in which the reactor tube may play an important role; once the metal surface is covered with coke, a second heterogeneous, but non-catalytic mechanism dominates [17]. In this mechanism, coke precursors from the gas phase react with the coke surface via radical reactions, leading to further growth of the coke layer. It was believed that

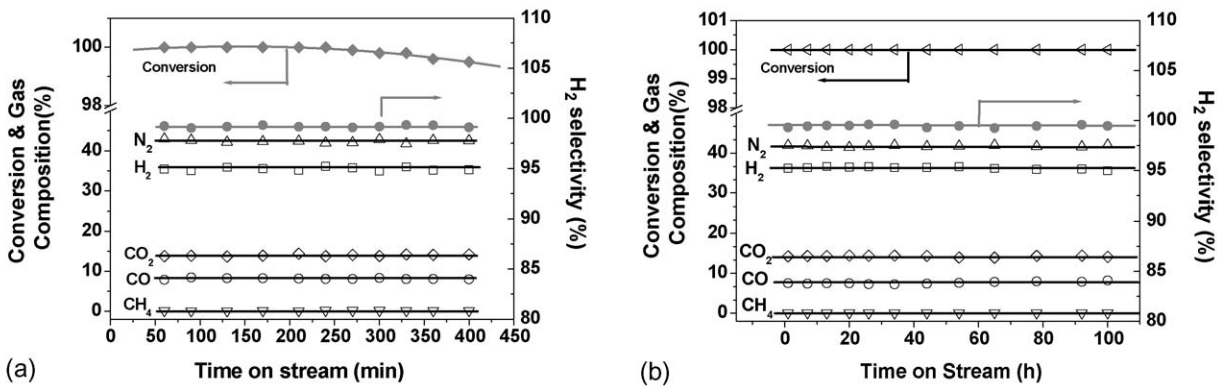

Fig. 5. The catalytic performance of ATR catalyst versus time on stream: (a) in the gasoline ATR process without prereforming and (b) in the integrated process ($T_{outlet} = 770$ °C, $P = 5.0$ bar, GHSV = 28,000 ml g^{-1} h^{-1}, $S/C = 2.7$ and $O_2/C = 0.5$).

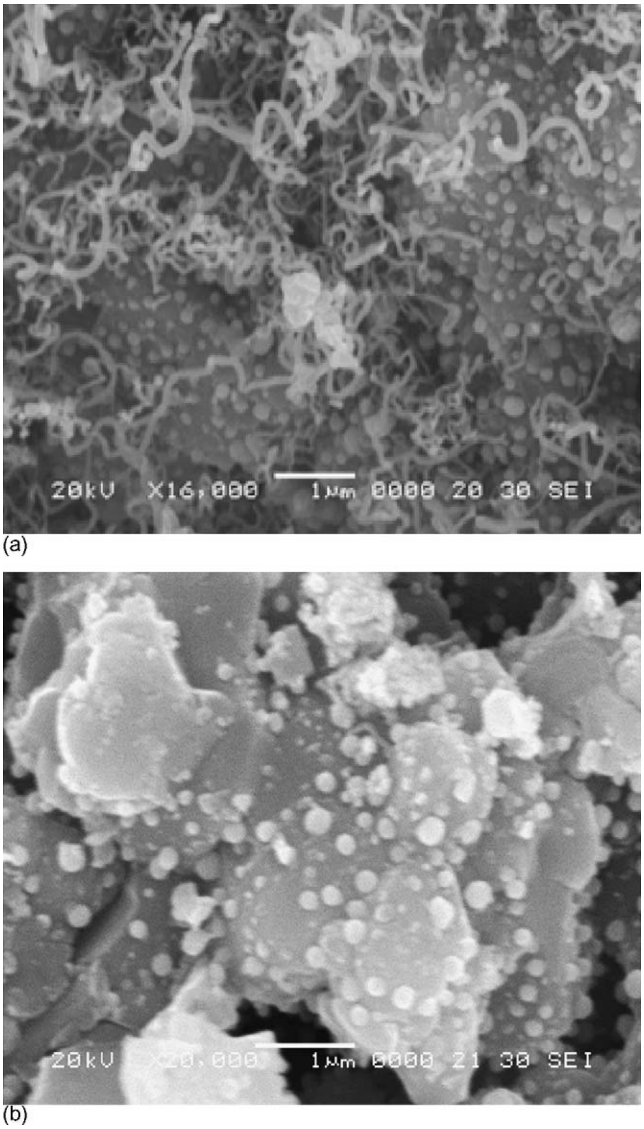

Fig. 6. SEM morphology of the ATR catalysts: (a) in the gasoline ATR process without prereforming after reaction time-on-stream of 8 h and (b) in the integrated process after reaction time-on-stream of 100 h.

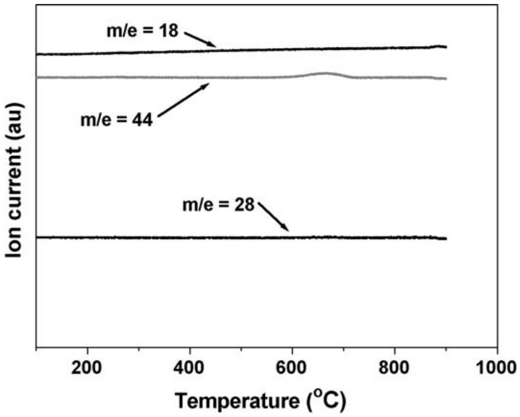

Fig. 7. O_2-TPSR diagram of the ATR catalyst after reaction for 100 h in the integrated process.

coke generated from non-catalytic gas phase reactions tend to initiate the coke deposition on the ATR catalyst [18]. In comparison with the process without prereforming, the morphology of the used ATR catalyst in the integrated process is illustrated in Fig. 6(b), which confirmed that there is no filamentous coke deposition. To detect if there was other kinds of carbon deposited on the ATR catalyst in the integrated process, O_2-TPSR experiments of the used ATR catalysts were also performed and the results are shown in Fig. 7, which confirmed that the ATR catalyst used in the integrated process was almost coke-free. Thus, in the integrated process, undesired gas phase reactions leading to the formation of alkenes were successfully avoided and the ATR catalyst stability in the integrated process was significantly enhanced. Simultaneously, air was introduced after the prereforming process; which served as a good coke gasification reagent and further suppressed the coke deposition. Moreover, the composition of reformate from the ATR reactor thermodynamically favors the reverse Boudart's reaction, which also contribute to the coke-free operation of the integrated process.

4. Conclusions

In the process of H_2 production from gasoline ATR over nickel-based reforming catalysts using direct feedstock mixing, the higher risk of coke deposition on the ATR catalyst and stainless steel reactor wall are associated with the gas phase reactions such as cool flame reaction, pyrolysis and steam cracking. It has been demonstrated clearly that using an integrated process, e.g., introduction of prereforming before ATR benefits the stability of process significantly. In the integrated process, gasoline first convert into methane rich gas during prereforming, therefore, undesired gas phase reactions leading to coke deposition are efficiently suppressed and catalyst deactivation caused by coke deposition minimized. In fact, coke-free operation of the process using nickel-based reforming catalysts was achieved. Elimination of higher hydrocarbons, especially light alkenes in reformate from the ATR reactor reduced the risk of coke deposition on the downstream WGS and/or PROX catalysts. Prereforming introduction does not alter the thermodynamic equilibria and products selectivities under much higher GHSV working conditions than those in steam reforming; meanwhile it allows a higher inlet temperature in the ATR reactor without coke deposition risk. Those combined characteristics of the integrated process make it potential application to the fuel cell distributed power generation. In addition, highly active gasoline prereforming catalyst with promising stability was developed and worth further investigation. Future work should focus much on the influence of prereforming introduction on the compactness of a fuel processor or H_2 production unit based on the gasoline ATR techniques, therefore, further investigation on the stability and catalytic performance of prereforming catalyst under much higher reaction GHSV conditions is of great significance.

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