

Syngas Production from CO₂ Reforming and CO₂-steam Reforming of Methane over Ni/Ce-SBA-15 Catalyst

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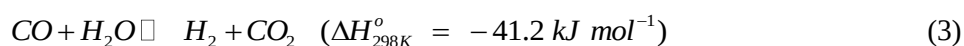
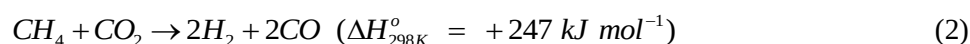
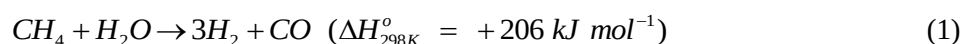
Abstract. This study compares the catalytic performance of mesoporous 10%Ni/Ce-SBA-15 catalyst for CO₂ reforming and CO₂-steam reforming of methane reactions in syngas production. The catalytic performance of 10%Ni/Ce-SBA-15 catalyst for CO₂ reforming and CO₂-steam reforming of methane was evaluated in a temperature-controlled tubular fixed-bed reactor at stoichiometric feed composition, 1023 K and atmospheric pressure for 12 h on-stream with gas hourly space velocity (GHSV) of 36 L g_{cat}⁻¹ h⁻¹. The 10%Ni/Ce-SBA-15 catalyst possessed a high specific BET surface area and average pore volume of 595.04 m² g⁻¹. The XRD measurement revealed the presence of NiO phase with crystallite dimension of about 13.60 nm whilst H₂-TPR result indicates that NiO phase was completely reduced to metallic Ni⁰ phase at temperature beyond 800 K and the reduction temperature relied on different degrees of metal-support interaction associated with the location and size of NiO particles. The catalytic reactivity was significantly enhanced with increasing H₂O/CO₂ feed ratio. Interestingly, the H₂/CO ratio for CO₂-steam reforming of methane varied between 1 and 3 indicated the occurrence of parallel reactions, i.e., CH₄ steam reforming giving a H₂/CO of 3 whilst reverse water-gas shift (RWGS) reaction consuming H₂ to produce CO gaseous product.

1. Introduction

Approximately 80% of the present global energy requirements are supplied by the hydrocarbon-based fossil fuels which eventually lead to depletion of limited fossil energy resources and global greenhouse effect problem [1]. According to Omae et al. [2], the amount of carbon dioxide has increased by 90 ppm (480 billion tons) in about 200 years since 1800. Therefore, it is necessary to seek for an alternative solution to remedy the environmental misdeed and substituting the hydrocarbon-based energy. Syngas is referring to a mixture of H₂ and CO which has been utilised as raw material for synthetic fuels production using Fisher-Tropsch synthesis for fossil fuels replacement [3]. Generally, the catalytic steam or CO₂ reforming of hydrocarbons reaction are employed to generate syngas.



However, severe catalyst deactivation due to carbon formation in CO₂ reforming reaction is always a setback for an industrial application whilst steam reforming reaction giving a stoichiometric molar ratio of H₂/CO around 3 is unsuitable for Fischer-Tropsch synthesis [4]. Therefore, the production of syngas from combined steam and CO₂ reforming (CO₂-steam reforming) of methane seems to be a promising method reasonably due to its remarkably feasible process to control the desired H₂/CO product ratio of 2 known as a practical feed composition for the Fischer-Tropsch (FT) process by manipulating feedstock ratio of CO₂ and H₂O [5]. As seen in (1)-(3), the CO₂-steam reforming of methane reaction involves the following reactions, i.e., steam reforming of methane, CO₂ reforming of methane and water-gas shift (WGS) or reverse WGS (RWGS) reaction.



A large number of scientific publications reported that noble metals such as Ru, Rh, Pt, Pd and Ir exhibit considerable catalytic performance, stability and great coking resistance in both steam and CO₂ reforming processes [6]. However, industrial application of noble metals is unsuitable considering their high cost and restricted availability and hence, researches on non-noble metal catalyst becoming more practical and indispensable. Ni-based catalysts are one of the outstanding catalyst for reforming processes but Ni sintering of the metallic and support phases and coke formation resulting rapid catalyst deactivation is an inevitable setback at such high reaction temperature [7]. Therefore, it is necessary to develop Ni-based catalysts concerning to introduce or modify the interaction between the support and Ni metallic particles, such as using mesoporous SBA-15 as support. The mesoporous SBA-15 having an ordered structure which allows and confines Ni metallic particles in the channels and consequently, the sintering of Ni metallic particles can be mitigated and improve the stability of catalysts [8].

Mesoporous SBA-15 has attracted a significant attention in catalysis due to its hexagonal ordered structure which can be employed as an excellent host to disperse and confine nanoparticles. Apart from that, mesoporous SBA-15 also possess large specific surface areas, long-range ordering of mesoporous channels and thick pore walls with high hydrothermal stability [9]. These properties are not only beneficial to the dispersion of loading metal for creating more active sites, but also ensure the transformation of reactants and products from preventing carbon deposition and restrain the mobility of species that lead to metal sintering [10]. Furthermore, the surface silanol groups over the mesostructure provide a unique platform for immobilizing metal precursors inside the channels of SBA-15 material [11].

Ce-containing mixed oxides can be considered to be very promising supports enhancing the catalytic performance via strong metal-support interaction and improving dispersion of the active metal on catalyst support [12]. It found out that the mobility of surface oxygen species over the catalyst surface has been promoted due to the strong redox and oxygen vacancy formation between Ce⁴⁺ and Ce³⁺ [13]. The presence of Ce on support surface provides an additional storage capacity where oxygen can repeatedly undergoes the reduction and oxidation cycles and continuously producing mobile surface oxygen during the reaction. Subsequently, the oxygen transfer process improve the coking resistance of active metals. Thus, ceria-containing catalysts have been widely studied in CO oxidation, water-gas shift reaction and reforming processes. Shi et al. [14] revealed the addition of Ce species to Cr/SBA-15 catalysts remarkably changed the redox properties and improved the catalytic performance of Cr-based catalysts in the dehydrogenation of ethane to ethylene reaction. Wang et al. [15] have investigated the catalytic performance of Ce-SBA-15 supported Ni catalysts for methane dry reforming. They reported the incorporated Ce into SBA-15 framework promoted the

nickel metallic particles formation with a smaller size compared to loaded CeO₂ on SBA-15 surface, which prevented the carbon formation and improved the catalytic activity.

It can be predicted that the combination of Ni and Ce-SBA-15 will lead to a good catalytic performance. This research intends to investigate the effect of steam addition on catalytic performance of 10%Ni/Ce-SBA-15 catalyst for CO₂ reforming of methane reaction and examine the influence of different feedstock ratios on activity of CO₂-steam reforming of methane reaction.

2. Experimental

2.1. Catalyst preparation

Ce-SBA-15 support was synthesized using triblock-poly(ethylene glycol)-block-poly(propylene glycol)-block-poly(ethylene glycol), referred as P-123 (EO₂₀PO₇₀EO₂₀) and tetraethyl orthosilicate (TEOS) as silica source and organic template agent, respectively. A calculated amount of EO₂₀PO₇₀EO₂₀ was dissolved in HCl solution with controlled pH of about 1 at 313 K and homogenized completely before the addition of TEOS and Cerium(III) nitrate hexahydrate (Ce(NO₃)₃·6H₂O) solutions. The mixture was further stirred at the same temperature for 24 h. Subsequently, it was transferred into a Teflon-lined autoclave and kept at 373 K for 24 h. The resulting white solid powder was filtered, washed rigorously with distilled water and dried in oven at 323 K overnight, followed by calcination in furnace at 823 K with a heating rate of 2 K min⁻¹ for 5 h in order to obtain 5%Ce-95%SBA-15 support.

The 10%Ni/Ce-SBA-15 catalyst was prepared by a conventional wetness impregnation method, using as-synthesized Ce-SBA-15 as support and Ni(NO₃)₂·6H₂O as metallic precursor. The measured amounts of Ni(NO₃)₂·6H₂O aqueous solution and Ce-SBA-15 support were mixed and magnetically stirred at room temperature for 3 h. The mixture was then dried in oven at 373 K overnight and air-calcined again in furnace at 1073 K with a ramping rate of 2 K min⁻¹ for 5 h.

2.2. Catalyst characterization

The measurement of Brunauer-Emmett-Teller (BET) surface area for support and catalyst was performed in a Micromeritics ASAP-2010 apparatus using N₂ physisorption isotherms data obtained at 77 K. The specimen was pretreated in N₂ flow at 573 K for 1 h to remove moisture and surface contamination before each measurement. X-ray diffraction (XRD) measurements of both Ce-SBA-15 support and 10%Ni/Ce-SBA-15 catalyst were also carried out in a Rigaku Miniflex II system using Cu monochromatic X-ray radiation (with wavelength, $\lambda = 1.5418 \text{ \AA}$) at 30 kV and 15 mA. All samples were scanned from 3° to 80° with relatively small scan speed and step size of 1° min⁻¹ and 0.02°, respectively to achieve high-resolution patterns.

The average crystallite size of NiO phase, $d(\text{NiO})$ was estimated from Scherrer's formula [16] whilst Ni⁰ metallic particle size, $d(\text{Ni}^0)$ obtained after NiO reduction was approximately calculated using the relative molar volumes of Ni⁰ metallic and NiO phases as shown in (4) [17].

$$d(\text{Ni}^0) = 0.84d(\text{NiO}) \quad (4)$$

By assuming spherical shape of Ni⁰ particles, the metallic dispersion, D was estimated using (5) [18].

$$D(\%) = \frac{90nm}{d(\text{Ni}^0)} \quad (5)$$

H₂ temperature-programmed reduction (H₂-TPR) was conducted on an AutoChem II-2920 system. Prior to reduction step, calcined 10%Ni/Ce-SBA-15 catalyst was pre-treated at 373 K for 30 min in He inert gas (50 ml min⁻¹). Specimen was subsequently reduced in 50 ml min⁻¹ of 10%H₂/Ar mixture from 373 to 1173 K with a heating rate of 10 K min⁻¹. Additionally, scanning electron microscopy coupled with energy-dispersive X-ray spectrometry (SEM-EDX) analyses were performed for spent catalysts in a Hitachi Tabletop Microscope TM3030Plus unit to investigate the external morphology of carbonaceous deposit on the surface of catalyst after reaction.

2.3. CO₂ or CO₂-steam reforming of methane

Both CO₂ reforming and CO₂-steam reforming of methane reactions were carried out under atmospheric pressure at 1073 K in a quartz tube fixed-bed continuous flow reactor (O.D. = 3/8 in. and length, *L* = 17 in.) for 12 h on-stream. Approximately, 0.1 g of catalyst was mounted by quartz wool in the middle of reactor. Additionally, the tubular reactor was placed vertically in a split temperature-controlled furnace and reaction temperature was accurately and continuously measured with time-on-stream (TOS). Gas hourly space velocity, GHSV = 36 L g_{cat}⁻¹ h⁻¹ was implemented for all runs to ensure the negligible internal and external transport resistances were acquired.

For CO₂ reforming of methane test, a precisely regulated amount of CH₄ and CO₂ gases was diluted in a mixer with N₂ gas employed as a tie component for material balance purpose and ensuring the total flow rate of 60 ml min⁻¹ for all runs before the gaseous mixture was directed to the inlet of tubular reactor. In the case of CO₂-steam reforming reaction, purified water was also fed to the top of reactor by a KellyMed KL-602 syringe pump and vaporized before being blended with other gaseous reactants. The total flow rate of reactant mixture diluted in N₂ was also maintained at 60 ml min⁻¹ for bi-reforming reaction. In addition, the composition of gaseous product stream from the bottom of fixed-bed reactor was analysed in a gas chromatograph (Agilent 6890 Series GC system) possessing TCD detector.

3. Results and discussion

3.1. Characterization results

3.1.1. Textural properties. In order to examine the mesoporous structure of 10%Ni/Ce-SBA-15 catalyst, N₂ adsorption-desorption isotherm curves of both support and catalyst are displayed in Figure 1. Based on the IUPAC classification, the isotherms of Ce-modified SBA-15 support had a type IV isotherm curve with an obvious H1-type hysteresis loop, typical for mesoporous materials possessing cylindrical channels [19]. Additionally, the sharp inflection observed in isotherm curves at *p/p*⁰ of 0.65 to 0.85 was reasonably due to capillary condensation of N₂ within ordered mesopores (Wang et al., 2012) and suggested the existence of large mesopores [20]. Interestingly, the isotherm profiles of the 10%Ni/Ce-SBA-15 catalyst were similar to that of Ce-SBA-15 support indicating that the mesoporous structure of support was largely maintained during NiO incorporation. As seen in Table 1, despite of unchanged mesoporous structure, the unavoidable drop in both surface area and total pore volume was evident for 10%Ni/Ce-SBA-15 catalyst rationally owing to the successful penetration of NiO nanoparticles into the mesoporous channels of Ce-SBA-15 support. Additionally, some pore walls may be altered during the NiO incorporation resulting in a decrease of average pore diameter from 66.63 to 63.81 Å as given in Table 1 [21].

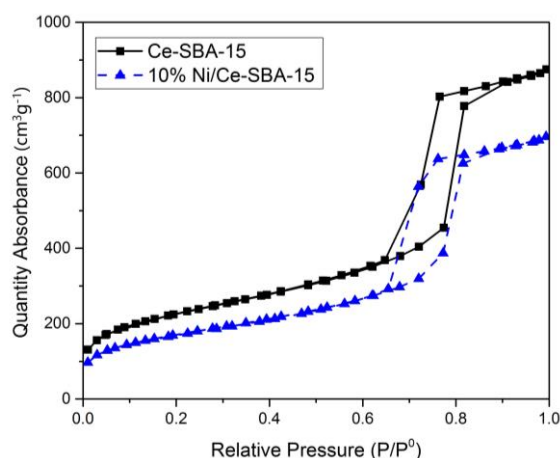


Figure 1. N₂ physisorption isotherms of Ce-SBA-15 support and 10%Ni/Ce-SBA-15 catalyst.

Table 1. Textural properties of support and catalyst.

Sample	S_{BET} (m ² g ⁻¹) ^a	V_p (cm ³ g ⁻¹) ^b	D_p (Å) ^c
Ce-SBA-15	760.47	1.21	66.63
10%Ni/Ce-SBA-15	595.04	1.08	63.81

^a BET specific surface area.

^b Total pore volume obtained at $p/p^0 = 0.99$.

^c Average pore diameter calculated by Barret-Joyner-Halenda (BJH) desorption method.

3.1.2. X-ray diffraction measurement. Figure 2 shows the XRD patterns of fresh mesoporous Ce-SBA-15 support and 10%Ni/Ce-SBA-15 catalyst. The X-ray diffractograms of both catalyst and support possessed a similar broad diffraction peak at 2θ angle of around 23° belonging to amorphous SiO₂ phase of SBA-15 framework [22]. The similarity in XRD patterns for SiO₂ phase between catalyst and support employed would further confirm the stability of mesoporous structure of Ce-SBA-15 support unaffected by embedded NiO particles. In addition, four sharp peaks with high intensity on the diffractogram of 10%Ni/Ce-SBA-15 catalyst located at $2\theta = 37.46^\circ$, 43.54° , 63.03° and 75.14° were assigned to the corresponding (1 1 1), (2 0 0), (2 2 0) and (3 1 1) crystal planes of NiO phase (JCPDS card No. 47-1049). However, there was no detection of CeO₂ phase suggesting that CeO₂ particles were well dispersed into the mesoporous channels of SBA-15 support or Ce atoms isomorphously substituted Si atoms in the silica framework. Additionally, the average crystallite size of NiO particles was about 13.60 nm, estimated from the most intense NiO line at 2θ of 43.54° by Scherrer's equation. Hence, the Ni⁰ metallic particle dimension and metal dispersion were computed as *ca.* 11.42 nm and 7.88% using (4) and (5), respectively.

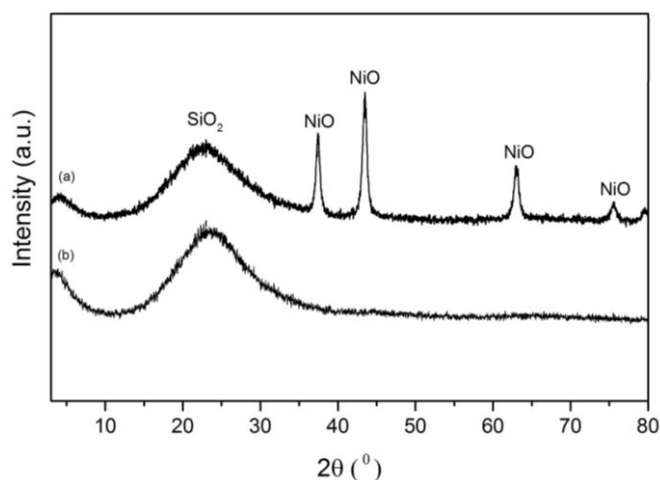


Figure 2. XRD patterns of (a) 10%Ni/Ce-SBA-15 catalyst and (b) Ce-SBA-15 support.

3.1.3. H_2 temperature-programmed reduction. Figure 3 shows the H_2 -TPR profile of 10%Ni/Ce-SBA-15 catalyst with 10% H_2 /Ar mixture. The reduction of NiO to Ni^0 metallic phase was widely reported as a single process [23, 24]. However, two broad peaks, P1 and P2 were observed at reduction temperature of about 650 and 728 K, correspondingly. He et al. [25] reported that nickel silicate was reduced at temperature above 973 K and nickel silicate was not detected on XRD pattern of 10%Ni/Ce-SBA-15 catalyst (see Figure 2). Thus, both P1 and P2 peaks belonged to the reduction of NiO phase to Ni^0 metallic phase. Additionally, the presence of multiple peaks in H_2 -TPR was attributed to the location of NiO particles on siliceous SBA-15 support and the extent of metal-support interaction depending on crystallite size. In fact, the low temperature peak (P1) was assigned to the reduction of surface NiO phase with weak metal-support interaction [26] while the high temperature peak (P2) corresponded to the reducibility of small NiO species resided in the mesoporous framework of SBA-15 support [27].

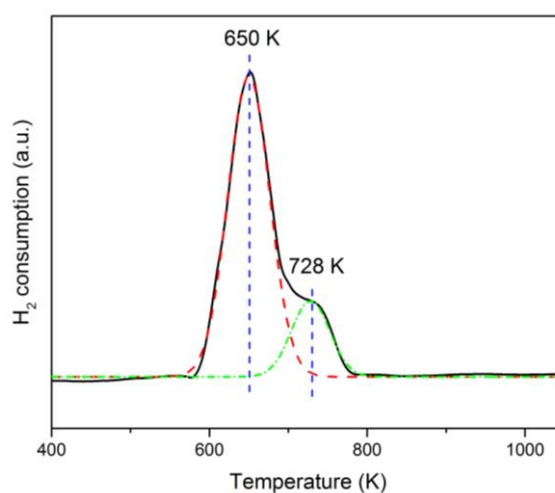


Figure 3. H_2 -TPR profile of 10%Ni/Ce-SBA-15 catalyst at a heating rate of 10 K min⁻¹.

3.1.4. SEM-EDX analysis. The spent 10%Ni/Ce-SBA-15 catalysts after CO_2 reforming (at 800 K and $P_{CH_4} = P_{CO_2} = 20$ kPa) and CO_2 -steam reforming (at 800 K, $P_{CH_4} = 45$ kPa, $P_{H_2O} = 30$ kPa and $P_{CO_2} = 15$ kPa) of methane reactions were examined using SEM-EDX to investigate the surface morphology of carbonaceous species, elemental composition and dispersion. Figure 4 and Figure 5 display the SEM image, elemental mapping and EDX spectrum of spent 10%Ni/Ce-SBA-15 catalysts for CO_2

reforming and CO₂-steam reforming of methane, respectively. As seen in Figure 4(a) and Figure 5(a), whisker-like and graphitic carbons were present on the surface of spent catalysts from both reactions. It was evident that the dispersion of metallic Ni particles was relatively fine on Ce-SBA-15 support surface (see Figure 4(b) and Figure 5(b)). Interestingly, the carbon content of spent catalyst from CO₂-steam reforming reaction (see the inset in Figure 5(c)) was about 12.1% lower than that of used catalyst from CO₂ reforming of methane (see Figure 4(c)). The reduction in carbon content for CO₂-steam reforming reaction was reasonably due to the steam gasification of deposited carbon [28].

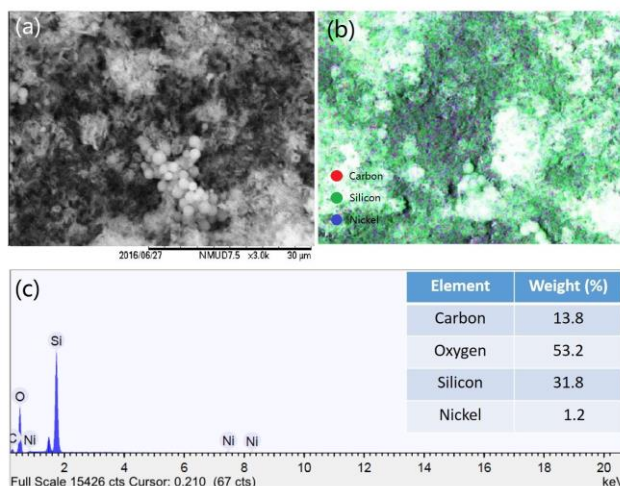


Figure 4. Spent 10%Ni/Ce-SBA-15 catalyst after CO₂ reforming of methane reaction: (a) SEM image, (b) EDX mapping image and (c) EDX spectrum.

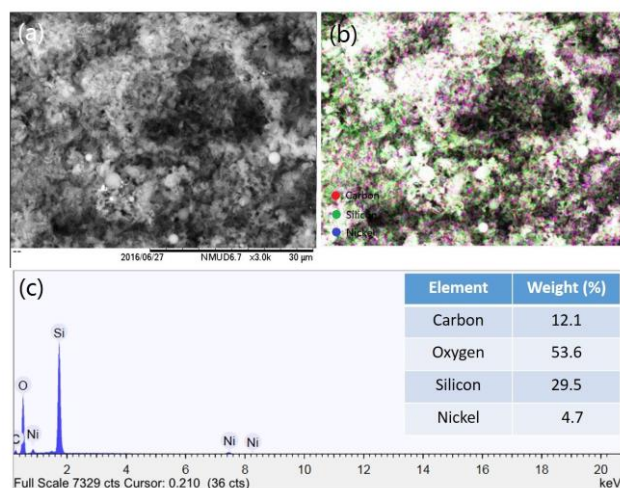


Figure 5. Spent 10%Ni/Ce-SBA-15 catalyst after CO₂-steam reforming of methane reaction: (a) SEM image, (b) EDX mapping image and (c) EDX spectrum.

3.2. Evaluation of CO₂ or CO₂-steam reforming reactions of methane

A time-on-stream test of 12 h was performed for 10%Ni/Ce-SBA-15 catalyst in order to examine the catalytic performance for both CO₂ dry reforming and CO₂-steam reforming of methane. Both reactions were carried out at stoichiometric feed composition and 1073 K. The transient profiles of CH₄ conversion and H₂/CO ratio for both reactions are shown in Figure 6 and Figure 7. It is apparent that catalyst performance was stable within 12 h on-stream for all reaction runs. Theoretically, the steam addition to CO₂ reforming of methane would increase the CH₄ conversion rationally due to the

parallel occurrence of methane steam reforming reaction. In fact, as seen in Figure (6), the CO_2 -steam reforming of methane exhibited a far lower CH_4 conversion compared to that of CO_2 reforming of methane. This observation was reasonably due to the H_2O , CO_2 and their decomposed products occupied the sites on the catalyst suppressing the CH_4 decomposition, as reported by Qin et al. [29]. However, the 10%Ni/Ce-SBA-15 catalyst in this study exhibited a significant CH_4 conversion of 75% for CO_2 -steam reforming of methane outperforming Danilova et al. [30] synthesized porous Ni-based catalysts. In their study, porous Ni-based catalysts were only capable of achieving a satisfactory CH_4 conversion of 60% and below overall for a CO_2 -steam reforming of methane with 20 h on-stream. Figure (7) shows H_2/CO ratio with time-on-stream for CO_2 reforming and CO_2 -steam reforming of methane at 1073 K. The H_2/CO ratio for CO_2 reforming of methane reaction was about 0.9 at 1073 K due to the concomitant reverse water-gas shift reaction whereas a higher value of 2.4 was observed for CO_2 -steam reforming of methane reaction which evident the simultaneous presence of CH_4 steam reforming during reaction runs.

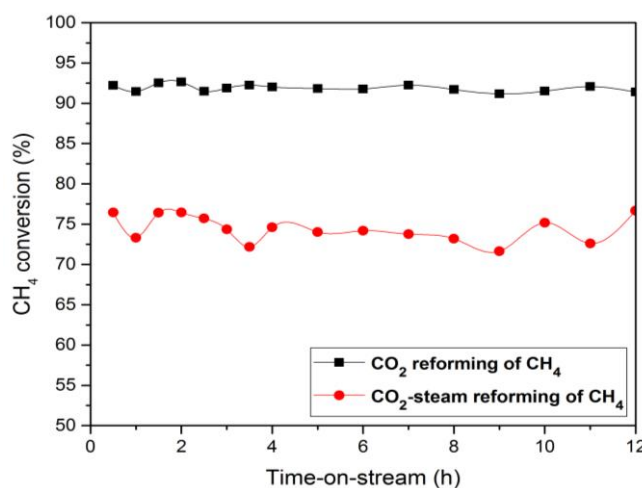


Figure 6. CH_4 conversion with time-on-stream for CO_2 reforming and CO_2 -steam reforming of methane at 1073 K.

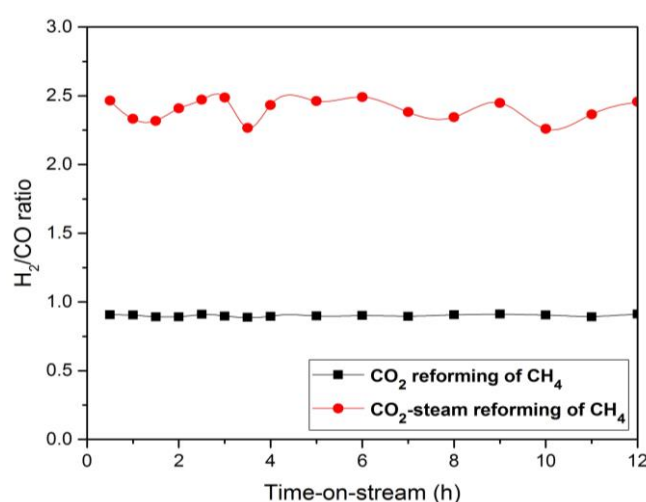


Figure 7. H_2/CO ratio with time-on-stream for CO_2 reforming and CO_2 -steam reforming of methane at 1073 K.

3.3. Effect of feed ratios on CO_2 -steam reforming of methane

A varying feed ratios of steam (P_{H_2O}) and CO_2 (P_{CO_2}) from 10 to 20 kPa were employed in CO_2 -steam reforming of methane with 10%Ni/Ce-SBA-15 catalyst to investigate different feedstock ratio effect on catalytic performance and H_2/CO product ratio for synthetic fuels application. As seen in Figure 8, growing P_{H_2O} was beneficial to the reaction and significantly increased the CH_4 conversion and H_2 yield by up to 11.44% and 26%, respectively. This observation was reasonably due to the presence of excess P_{H_2O} enhanced the CH_4 steam reforming reaction and producing a high H_2/CO ratio.

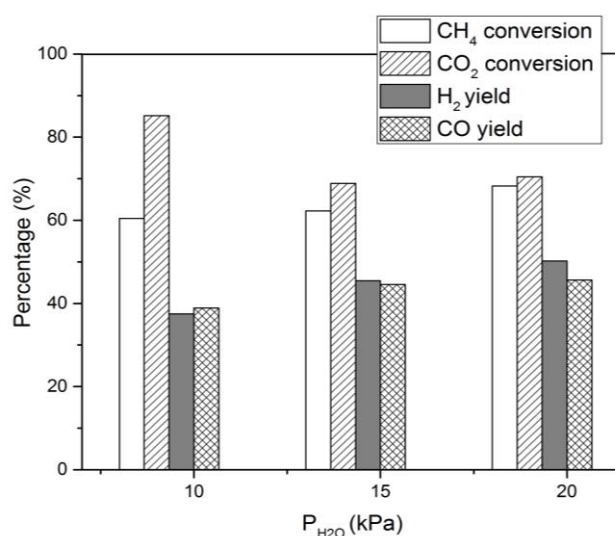


Figure 8. Effect of steam feed ratios on CH_4 conversion, CO_2 conversion, H_2 yield and CO yield for CO_2 -steam reforming of methane at $P_{CH_4} = 45$ kPa, $P_{CO_2} = 15$ kPa and 1073 K.

In fact, in the study of thermodynamics of combined CO_2 -steam reforming of methane, catalyst activity and H_2/CO ratio increased considerably with the addition of excess steam. A considerable decrease of CO_2 conversion (see Figure 8) with growing P_{H_2O} was expected and supported by the works of Özkara-Aydinoğlu [31]. In the paper, the CH_4 was reportedly reacted with H_2O instead of CO_2 whenever both H_2O and CO_2 are played as co-oxidant due to the high stability of CO_2 nature. The foregoing discussion was further confirmed since the CH_4 conversion increased while CO_2 conversion decreased with rising P_{H_2O} as seen in Figure 8.

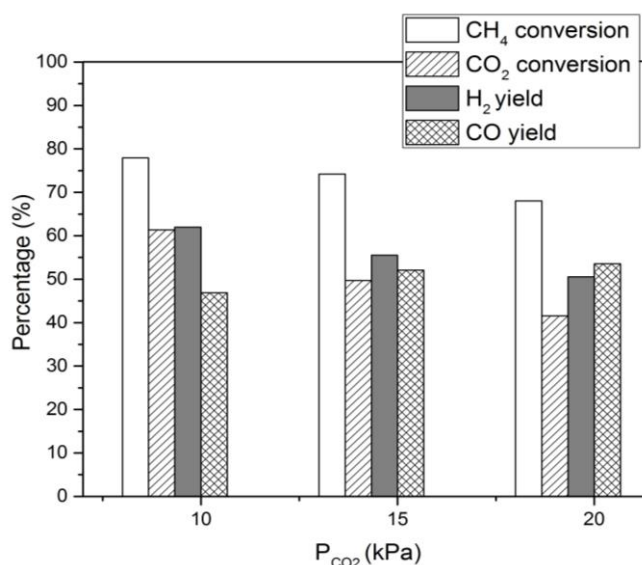


Figure 9. Effect of CO₂ feed ratios on CH₄ conversion, CO₂ conversion, H₂ yield and CO yield for CO₂-steam reforming of methane at P_{CH₄} = 45 kPa, P_{H₂O} = 30 kPa and 1073 K.

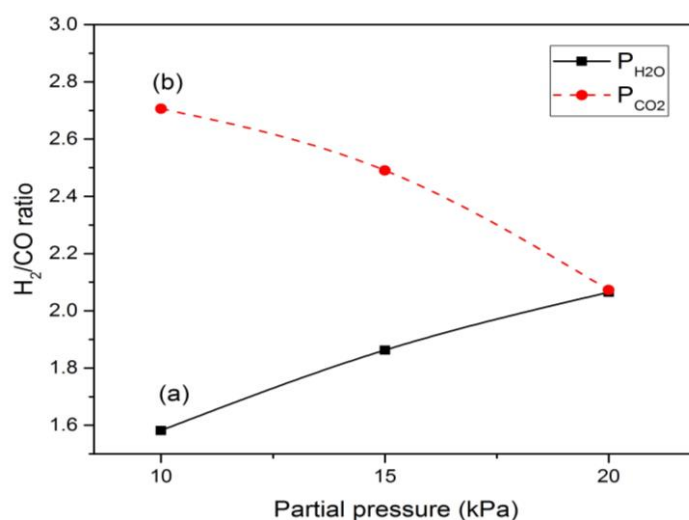


Figure 10. Effect of (a) varying steam feed ratios, P_{H₂O} on H₂/CO ratio at 1073 K; P_{CH₄} = 45 kPa and P_{CO₂} = 15 kPa and, (b) varying CO₂ feed ratios, P_{CO₂} on H₂/CO ratio at 1073 K; P_{CH₄} = 45 kPa and P_{H₂O} = 30 kPa.

Figure 9 shows the influence of varying CO₂ feed ratios (P_{CO₂}) on the catalyst activity at 1073 K. Besides the CO yield, all the catalyst activities decreased with growing P_{CO₂}, particularly H₂ yield dropped by 19.35%. As seen in Figure 9, both CH₄ and CO₂ conversion exhibited a significant decrease trend suggesting the excessive presence of CO₂ coming from CH₄ steam reforming could suppress the CH₄ adsorption and decomposition on catalyst surface and hence lessened the CO₂-steam reforming of methane reaction. Interestingly, the P_{H₂O} and P_{CO₂} exhibited a totally opposite trend in H₂/CO ratio as seen in Figure 10. This trend was similarly observed by Huang et al. [32] who studied on the effects of feed gas composition on the activity of 3%MgO-10%Ni/SBA-15 catalyst. The catalyst proposed in their study exhibited a high CH₄ conversion of 98.3% in CO₂-steam reforming of methane. However, in the case of production of long chain hydrocarbons a H₂/CO ratio of 2 is desirable instead. The catalyst used in this study managed to obtain a more appropriate H₂/CO ratio of 2.07 in comparison to that of Huang et al. [32]. Interestingly, the H₂/CO ratio of 2.07 could be obtained at about of 20 kPa for both P_{H₂O} and P_{CO₂}. In fact, the H₂/CO ratio for CO₂-steam reforming of methane varied between 1 and 3 indicated the occurrence of parallel reactions, i.e., CH₄ steam reforming giving a H₂/CO of 3 whilst reverse water-gas shift (RWGS) reaction consuming H₂ to produce CO gaseous product. In this study, the H₂/CO ratio of about 2.07 could be obtained with appropriate feed ratios of (i) CH₄:H₂O:CO₂ = 3:1.33:1 or (ii) CH₄:H₂O:CO₂ = 3:2:1.33 at 1073 K and 1 atm.

4. Conclusions

The 10%Ni/Ce-SBA-15 catalyst synthesized by wetness impregnation method had been successfully evaluated for CO₂ reforming and CO₂-steam reforming of methane reaction in a quartz fixed-bed reactor at different H₂O:CO₂ feed ratios, 1073 K and 1 atm. The 10%Ni/Ce-SBA-15 catalyst possessed high BET surface area of 595.04 m²g⁻¹. The XRD and EDX results were in accordance that the CeO₂ were effectively embedded into ordered mesoporous SBA-15 framework. The crystallite size of NiO was reduced to metallic Ni from 13.60 to 11.42 nm and they were finely dispersed on the catalyst surface whilst H₂-TPR analysis showed that the reduction of NiO was a single process. The SEM-EDX measurement showed that the addition of steam to CO₂ reforming of methane improved the coking

resistance of catalyst due to the activated oxygen from steam to the catalyst surface enhancing the gasification process of deposited carbon. Apart from that, the 10%Ni/Ce-SBA-15 catalyst experienced a gradual increase in CH₄ conversion and H₂ yield with rising P_{H₂O} whilst a decrease in CH₄ conversion and H₂ yield with growing P_{CO₂}. The P_{H₂O} and P_{CO₂} of 20 kPa seemed to be the optimal partial pressure for CO₂-steam reforming of methane in terms of conversion, H₂ yield and H₂/CO ratio of 2. Furthermore, the H₂/CO ratio of about 2.07 could be obtained with appropriate feed ratios of (i) CH₄:H₂O:CO₂ = 3:1.33:1 or (ii) CH₄:H₂O:CO₂ = 3:2:1.33 at 1073 K and 1 atm.

Acknowledgments

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