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# A Short Review on Bimetallic Co-based Catalysts for Carbon Dioxide Reforming of Methane

Mahadi B. Bahari<sup>1</sup>, H.D. Setiabudi<sup>1, 2, \*</sup>, Nurul Ainirazali<sup>1</sup>, Dai-Viet N. Vo<sup>3</sup>

<sup>1</sup>Faculty of Chemical and Process Engineering Technology, College of Engineering Technology, Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia.

<sup>2</sup>Centre of Excellence for Advanced Research in Fluid Flow (CARIFF),

Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia.

<sup>3</sup>Center of Excellence for Green Energy and Environmental Nanomaterials (CE@GrEEN), Nguyen Tat Thanh University, 300A Nguyen Tat Thanh, District 4, Ho Chi Minh City 755414, Vietnam

\*Corresponding author.

E-mail address: [herma@ump.edu.my](mailto:herma@ump.edu.my) (H.D. Setiabudi)

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## Abstract

The development of catalysts that afford excellent catalytic performance along with the ability to withstand coking challenge is fundamental in carbon dioxide reforming of methane since the undesired coke was inevitably generated during reaction. Apart from high resources and low cost as Ni, a slight focus of attention has been given towards the Co-based catalyst since this metal capable in improving catalytic stability and lowering the coke formation. However, the lower catalytic activities of Co compared to Ni-based catalyst in reforming works are the real challenges that need to be solved. In this study, a short review on various approaches that have been implemented by researchers in their works for improving the catalytic performance such as employment of different types of support, optimization of the cobalt content in the catalyst, incorporation of second metal as promoter or addition of second metal to form bimetallic catalysts. In addition, the outlook of related of bimetallic Co-based has been proposed to provide more critical information.

*Keywords: Syngas, Bimetallic Co-based; Monometallic Co-based, Carbon dioxide reforming of methane;*

## 1. Introduction

The reliance on fossil fuels to meet energy demand has created environmental problems by the significant emission of greenhouse gas, CO<sub>2</sub> and CH<sub>4</sub>. Besides, the depletion in the reserves of fossil fuels also is one of the concerning issues which the world is currently faced. Thus, syngas has been acclaimed as a potential option to replace fossil fuels since it holds many valuable uses included as energy to power engines, fuel for solid oxide fuel cells (SOFCs) and feedstock for Fischer-Tropsch synthesis (FTS) [1]. The well-established technologies for syngas generation such as steam reforming have their own drawbacks relate with anthropogenic CO<sub>2</sub> emission. Therefore, there has been increasing interests in carbon dioxide reforming of methane since this route converting the two kinds of greenhouse gases (CH<sub>4</sub> and CO<sub>2</sub>) into syngas [2-4]. Moreover, the reforming process produces syngas with a H<sub>2</sub>:CO molar ratio close to unity, making it an ideal for FTS [5, 6].

The employment of noble metals as catalysts has been widely explored for carbon dioxide reforming of methane due to their excellent catalytic performance along with strong coke resistance. Hou et al. (2006) [7] investigated the catalytic performance of several transition metals (Ru, Rh, Pt, Pd and Ir) over Al<sub>2</sub>O<sub>3</sub> support during carbon dioxide reforming of methane at 1073 K under CH<sub>4</sub>/CO<sub>2</sub> feed ratio of 1.0. They reported that all those transition metals exhibited stable catalytic performance with no coke formation during 240 min on stream, except Pd which contributed

to coke formation at about 4.9 mg coke  $\text{g}_{\text{cat}}^{-1} \text{h}^{-1}$  [7]. This observation strongly reflects the excellent coke-resistant capability of those transition metals in carbon dioxide reforming of methane since coke deposited could not be inevitably prevented due to  $\text{CH}_4$  cracking and Boudouard. Although those metals exhibited superior performance and resisted carbon formation, these types of metals are infrequently used for industrial purpose owing to their expensive cost and limited availability [8]. Thus, non-noble metals have gained significant attention as catalyst for carbon dioxide reforming of methane.

Non-noble metals such as Ni and Fe were preferred for carbon dioxide reforming of methane since they are relatively cheap, excellent capacity of C-H bond scission and possess high catalytic activity [9, 10]. Guo et al. (2004) [11] reported carbon dioxide reforming of methane to synthesis gas over Ni-based catalysts exhibited higher activity and better stability using a stoichiometric feed ratio (1:1) at 1023 K. However, coke formation is the main problem in the utilization of non-noble catalyst for carbon dioxide reforming of methane resulted from  $\text{CH}_4$  decomposition and CO disproportionation [12]. Then, cobalt was appointed potential catalyst owing to its higher stability and lower carbon deposition, aside from high resources and low cost [13, 14]. Park et al. (2018) [14] attained stable catalytic activity and low coke accumulation by employing Co supported on  $\text{Al}_2\text{O}_3$  in carbon dioxide reforming of methane.

In this work, a short review focusing on the previous development of Co-based catalyst for carbon dioxide reforming of methane including the catalytic performance and capability in suppressing the carbon deposition will be covered.

## 2. Dry reforming of methane over monometallic Co-based catalysts

Although Co-based catalyst showed excellent carbon resistance and good stability, their lower catalytic performances is the main reason for less application compared Ni-based catalysts [15]. Therefore, lot of research recently were focused on improving the metal dispersion, reducing the crystallite size of metal particles on catalyst surface and increasing the basic properties of catalysts. A few approached that had been reported in the literature review are through the employment of basic supports, addition of second metal as promoter or using different catalyst preparation technique. A summary of the previous findings related to the employment of Co-based catalysts in carbon dioxide reforming of methane attained from literature is presented in Table 1.

Table 1. List of cobalt based catalysts which have been examined in carbon dioxide reforming of methane.

Catalyst	Co (wt.%)	Method	T (K)	Surface area ( $\text{m}^2 \text{g}^{-1}$ )	Pore diameter (nm)	GHSV ( $\text{L g}_{\text{cat}}^{-1} \text{h}^{-1}$ )	Conversion (%)		$D_a$ (%)	$\text{H}_2$ yield (%)	$C_d$ (%)	Ref .
							$\text{CH}_4$	$\text{CO}_2$				
Co/MA	10	Incipient wetness impregnation	973-1073	141.9	6.28	36	47.7-76.2	58.3-82.2	n.a.	28.5-64.8	n.a.	[15]
La-Co/MA		Sequence incipient wetness impregnation		136.4	6.30		78.8-93.7	82.4-93.2		61.6-84.2		
Co/MA	10	Incipient wetness impregnation	1023	203.9	n.a.	36	70.8	71.2	n.a.	n.a.	n.a.	[16]
Y-Co/MA		Sequence incipient wetness impregnation		155.8			85.8	90.5				
CoAl/ $\text{Al}_2\text{O}_3$ CoAl/ $\text{CeO}_2$ CoAl/ $\text{MgAl}_2\text{O}_4$ CoAl/ $\text{SiO}_2$ CoAl/ $\text{ZrO}_2$	5	Wet impregnation	1123	105.7 8.9 94.9 264.0 13.2	9.8 >40.0 12.0 13.7 >40.0	60	79.3 49.3 87.6 5.3 67.6	89.0 67.5 93.7 12.8 84.8	6.9 11.6 0.1 36.1 5.8	73.2 35.5 84.2 8.3 71.8	4.2 1.4 3.4 1.5 26.2	[17]
Co/SBA-15 $\text{Sm}_2\text{O}_3$ /Co/SBa-15	10	Two-solvents impregnation	973	628.0 564.0	1.3 0.9	n.a.	24.0 11.4	38.0 29.1	11.7 2.1	n.a.	n.a. 56.9	[18]
Co/SBA-15 Co/ $\text{SiO}_2$ Rh-Co/SBA-15	12	Two-solvents impregnation	823	639.0 171.0 448.0	3.7 n.a. 3.7	67	16.9 4.97 48.8	n.a.	0.3 1.8 0.1	n.a.	n.a.	[19]
Co/ $\text{CeO}_2$	20	Wet impregnation	1023	39.4	1.2	30000 <sup>b</sup>	87.6	79.5	n.a.	37.6	n.a.	[20]
Co/ $\text{La}_2\text{O}_3$				16.5	1.2	n.a.	50.0	60.0		45.0		[21]

Table 1 (continued)

Catalyst	Co (wt.%)	Method	T (K)	Surface area (m <sup>2</sup> g <sup>-1</sup> )	Pore diameter (nm)	GHSV (L g <sub>cat</sub> <sup>-1</sup> h <sup>-1</sup> )	Conversion (%)		D <sub>a</sub> (%)	H <sub>2</sub> yield (%)	C <sub>d</sub> (%)	Ref.
							CH <sub>4</sub>	CO <sub>2</sub>				
Co/CeO <sub>2</sub> Ni-Co/CeO <sub>2</sub>	8 4	Incipient wetness impregnation	973	18.9 18.3	n.a.	n.a.	0.6 64.2	0.8 71.0	96.0 16.2	0.01 39.5	0 17.0	[22]
Co/ZrO <sub>2</sub> Co/CeO <sub>2</sub>	5	Wet impregnation	973	3.6 7.4	27.7 18.9	3.8	n.a.			75.8 59.4	28.2 11.0	[23]
Co-MgO	5 10 15 20 30	Co-precipitation	973	80.7 97.5 100.1 88.3 78.2	14.8 13.8 11.2 14.1 12.9	12	25.8 66.8 22.1 26.5 24.2	33.5 74.5 31.4 29.0 36.6	26.5 1.0 60.1 43.7 52.7	23.0 60.7 33.7 31.5 43.7	n.a.	[24]
Co/ZSM5 1Ni1Co/ZSM5 1Ni2Co/ZSM5 2Ni1Co/ZSM5	n.a.	Wet impregnation	973	247.0 292.0 237.0 284.0	n.a.	60	52.8 55.4 59.9 61.8	64.1 65.4 69.6 69.6	n.a. n.a. 6.1 19.3	n.a.	20.6 n.a. n.a. n.a.	[25]
Co/Al <sub>2</sub> O <sub>3</sub>	2	Controlled adsorption Dry impregnation	973	88.0 84.0	14.7 16.0	22 000 <sup>b</sup>	52.0 31.6	53.2 30.7	21.9 37.4	n.a.	n.a.	[26]
Co/SBA-15 Ru-Co/SBA-15	12	Two-solvents impregnation	1063	589.0 629.0	40.0 39.0	12.1	44.0 82.0	n.a. 71.0	n.a.	n.a.	n.a.	[27]
Co/CeO <sub>2</sub> Co-Ni/CeO <sub>2</sub>	7.5 3.75	Surfactant assisted co-precipitation	1023	67 105	n.a.	30	86.0 83.5	n.a.	2.5 0.8	n.a.	n.a.	[28]
Co/ZrO <sub>2</sub> Co-La/ZrO <sub>2</sub> Co-K/ZrO <sub>2</sub> Co-Mn/ZrO <sub>2</sub> Co-Ce/ZrO <sub>2</sub> Co-Mg/ZrO <sub>2</sub>	5	Impregnation Sequential impregnation	923	22.0 27.0 n.a. n.a. n.a. n.a.	n.a.	60	36.5 27.2 6.8 22.3 48.9 0	47.5 43.7 13.4 33.1 61.1 0	39.9 0.0 53.1 19.2 3.7 100	30.8 20.8 1.1 14.0 46.0 0	4.0 0.2 2.0 0.8 2.6 2.0	[29]

n.a. = not available, D<sub>a</sub> = deactivation rate, C<sub>d</sub> = Amount of carbon deposited.

<sup>a</sup>Gas hourly space velocity (GHSV) in (h<sup>-1</sup>)

The selection of basic supports for Co-based catalyst are important in carbon dioxide reforming of methane since it could provide thermal stability to Co active metals as well as suppress the deposition of carbon. Park et al. (2018) [17] examine the influence of different supports (Al<sub>2</sub>O<sub>3</sub>, CeO<sub>2</sub>, MgAl<sub>2</sub>O<sub>4</sub>, SiO<sub>2</sub>, and ZrO<sub>2</sub>) on the catalytic performance of CoAl catalyst in carbon dioxide reforming at 1123 K and stoichiometric condition. The authors stated that the superior performance exhibited by CoAl supported on MgAl<sub>2</sub>O<sub>4</sub> compared to Al<sub>2</sub>O<sub>3</sub>, ZrO<sub>2</sub>, CeO<sub>2</sub> and SiO<sub>2</sub> (cf. Fig. 1) was attributed to the balance between the rate of carbon deposition and surface oxidation, and does not closely related to quality of Co dispersion.

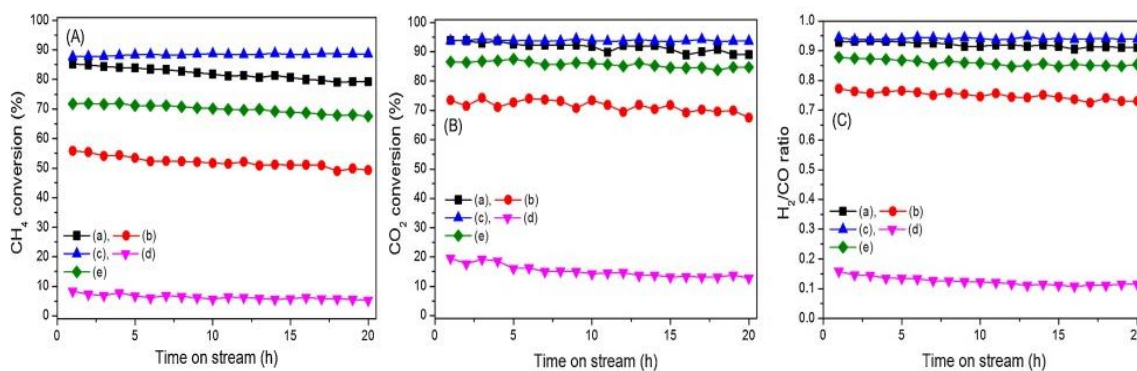


Fig. 1 The catalytic performance of CoAl catalyst supported on different metal oxides; (a) Al<sub>2</sub>O<sub>3</sub>, (b) CeO<sub>2</sub>, (c) MgAl<sub>2</sub>O<sub>4</sub>, (d) SiO<sub>2</sub>, and (e) ZrO<sub>2</sub> [17].

The employment of mesoporous materials such as SBA-15 and mesoporous alumina also capable in resolving Co low performance issue through improving the dispersion of active metal particles accredited to their high surface area. Additionally, this material could confine the active nanoparticles inside the pore channels and prevent the metal particles from sintering that leads to catalyst deactivation [30, 31]. Recently, our works [16] and Tran et al. (2020) [15] investigate the effect of MA employment as support for Co-based catalysts in carbon dioxide reforming of methane and observed that the reactant conversions attained through reforming was good at about > 65.0% at 1023 K. However, using SBA-15 as support, Jabbour et al. (2014) [26] noticed that the conversions only achieved about 44.0% with H<sub>2</sub>: CO ratio of approximately 0.6, which led to the conclusion that Co/SBA-15 was not selective toward the carbon dioxide reforming of methane.

The introduction of second active metal as catalyst promoter are also very promising approach in solving Co low performance since it probably able to significantly increase the oxygen mobility, enhance metal dispersion and reduce the acidity of catalyst [32]. In fact, the catalyst deactivation could be prevented owing to the improvement in catalyst carbon resistance [32]. Rare earth metal oxides such as CeO<sub>2</sub>, La<sub>2</sub>O<sub>3</sub>, Y<sub>2</sub>O<sub>3</sub> and Sm<sub>2</sub>O<sub>3</sub> gained considerable attention in catalytic reforming due to their basic attributes and high oxygen storage capability. In recent studies, the addition of La<sub>2</sub>O<sub>3</sub> as promoter effectively increased CH<sub>4</sub> conversion up to 93.7% at 1073 K accredited to the small crystallite size and the basic character of La<sub>2</sub>O<sub>3</sub> [15]. Our previous work also experienced the improvement in catalytic performance of Co-based catalyst after the incorporation of Y<sub>2</sub>O<sub>3</sub> as promoter [16]. Apart from that, the great oxygen storage capacity owned by La<sub>2</sub>O<sub>3</sub> and Y<sub>2</sub>O<sub>3</sub> help facilitating the carbon gasification process thus resulted in low carbon deposited as reported in those studies. Yet, Taherian et al. (2017) [18] noticed a contrary result of reforming activity after introduced Sm<sub>2</sub>O<sub>3</sub> as promoter on Co/SBA-15 with CH<sub>4</sub> conversion attained lowered than 25%. The authors clearly clarified that this negative effect was not resulted from promoter addition, but because of the oxidation of Co to inactive phase and sintering of Co particles at high temperatures.

### 3. Dry reforming of methane over bimetallic Co-based catalysts

Aside from all aforementioned efforts, it is also had been reported that the combination of Co with other metals to form bimetallic catalyst could alter the catalyst attributes, thus capable in enhancing the catalytic performance. According to Takanabe et al. (2005) [33], combination between Co-Ni to form bimetallic catalyst capable in improving reforming activity, especially CH activation on the metallic surface, and also avoiding the undesirable metal oxidation. In literature, the combination of cobalt and nickel supported on various types of support was extensively carried out for the carbon dioxide reforming of methane attributed to the formation of Co-Ni alloy. Indeed, there are some studies reported that bimetallic Co-Ni catalyst managed to exhibit superior reforming activity in carbon dioxide reforming of methane compared to the monometallic Co-based catalysts.

Luisetto et al. (2012) [28] evaluated the catalytic performance of bimetallic Co-Ni over CeO<sub>2</sub> support synthesized via surfactant assisted co-precipitation method. The authors found that the combination of this bimetallic catalyst resulted in larger crystallites compared to monometallic catalyst which consequently lowering the metal dispersion. However, the bimetallic catalyst attained superior catalytic activity compared to monometallic (cf. Fig. 2) which caused the author concluded that the improvement was accredited to the basic attributed of the Co-Ni alloy and not associated with the dispersion quality of active metal.

In another studies, Özkara-Aydınoğlu and Aksoylu (2010) [29] examined the effect of addition of metal additives, namely La, Ce, Mn, K and Mg as bimetallic for Co supported on ZrO<sub>2</sub> in carbon dioxide reforming of methane (at T = 923 K and CH<sub>4</sub>/CO<sub>2</sub> = 1). In the report, the bimetallic Co-Ce/ZrO<sub>2</sub> exhibited superior catalytic performance compared to other bimetallic and monometallic catalysts. The author also explained that Ce addition assisted in increasing the dispersion of Co on the whole ZrO<sub>2</sub> surface and enhancing the gasification of coke deposited. However, for other bimetallic catalysts, only the catalytic stability was improved within 6 h reforming while the reforming activity recorded were lowered than monometallic catalysts especially for Co-K and Co-Mn bimetallic catalysts (cf. Fig. 3). The lower performance by both bimetallic was resulted for both K-promoted and Mn-promoted Co/ZrO<sub>2</sub> catalysts attributed to the large particle size and low metal dispersion compared to monometallic.

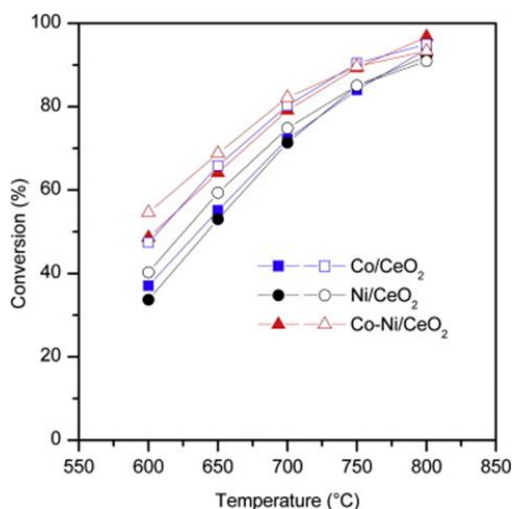


Fig. 2. The impact of bimetallic combination between Co-Ni toward the reforming activity [27].

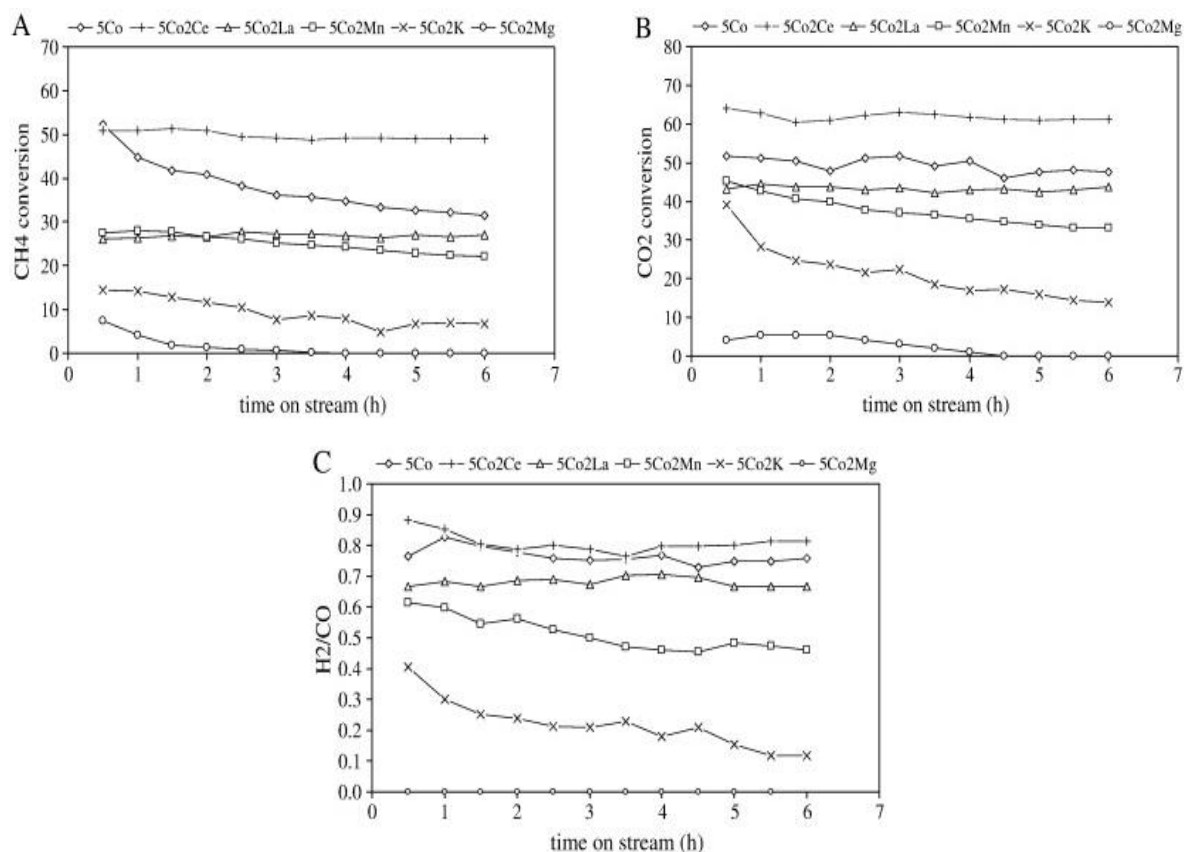


Fig. 3 The effect of addition of La, Ce, Mn, K and Mg as bimetallic for Co supported on ZrO<sub>2</sub> toward catalytic performances in carbon dioxide reforming of methane [28].

In bimetallic catalyst design, the ratio between Co and Ni is really crucial aspect since it could strongly affect the reforming performance and stability. Estephane et al. (2015) [24] studies the influence of different Ni:Co ratios in bimetallic Co-Ni supported by ZSM5 toward the catalytic performance in carbon dioxide reforming of methane. The author observed that all the bimetallic catalysts attained higher reforming activities compared to Co/ZSM5, regardless of Ni:Co ratios. Additionally, Ni-Co bimetallic with high cobalt loading had the superior catalytic performance in

term of activity and stability while possessed lowest coke deposition after 12 h reforming (cf. Fig. 4). Therefore, the author concluded that role of Co metal in bimetallic are crucial since the presence of this metal is efficient in inhibiting carbon deposition.

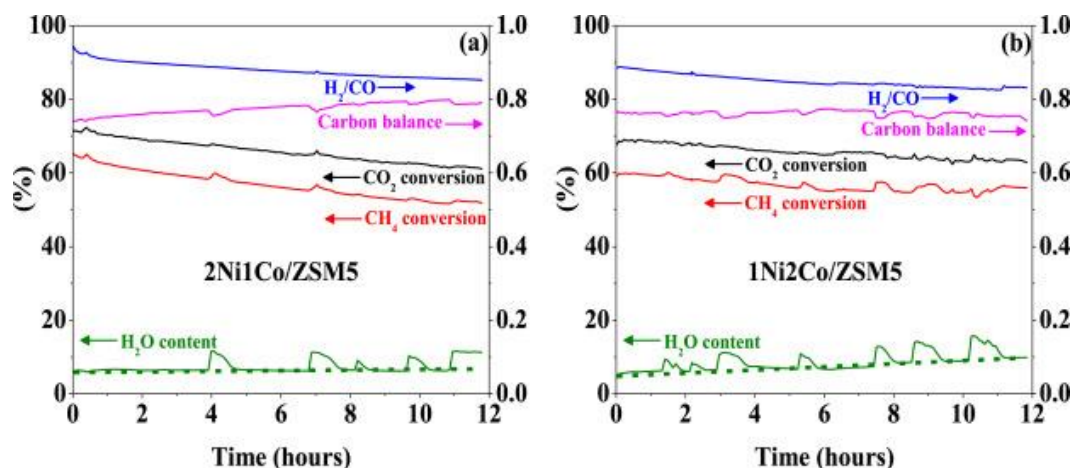


Fig. 4 The influence of Ni:Co ratios in bimetallic Co-Ni supported by ZSM5 toward the catalytic performance in carbon dioxide reforming of methane [24].

#### 4. Concluding remarks and future prospects

As explained in this work, the monometallic Co only capable in improving catalytic stability and carbon resistance, but not catalytic performance. Although there are many efforts that have been conducted by researchers as discussed in this paper, the combination of bimetallic Co-based catalyst could be the crucial techniques to tackle the issue of lower catalytic performance by monometallic Co. However, through this study, there is still limited knowledge regarding the formation of bimetallic Co-based catalysts and its performance in carbon dioxide reforming of methane. For future works, it is recommended to further investigate the synergistic effect of Co with different metals in bimetallic combination in order to form the best combination bimetallic of Co-based catalyst. Furthermore, there are many aspects that should be covered during the evaluation of bimetallic Co such as metal ratio, support or technique of preparation which could maximize the catalytic performance while suppress carbon accumulation.

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