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Research Article

Thermal oxidation of carbon monoxide in air using various self-prepared catalysts

Nguyen Thi Bich Thao, Nguyen Nhat Huy*

ABSTRACT

Carbon monoxide (CO) is a very toxic pollutant emitted from wood fired boiler, which is widely used in small and medium enterprises in Vietnam. The treatment of CO containing flue gas faces many difficulties due to the inert property of CO that cannot be removed by traditional adsorption and absorption methods and one of the effective CO treatments is catalytic oxidation. Therefore, we aimed to prepare various catalysts on different carriers for treatment of CO in flue gas, including γ-Al_2O_3-based metal oxides (Co_3O_4/Al_2O_3, Cr_2O_3/Al_2O_3, and CuO/Al_2O_3), CuO–MnOx/OMS-2, and CuO-MnOx/zeolite. The CO removal tests were conducted in a continuous fixed bed reactor in laboratory scale with temperature range of 50 – 550 °C. The characteristics of catalytic materials were then determined by various methods such as Brunauer-Emmett-Teller measurement, X-ray diffraction, energy-dispersive X-ray spectroscopy, Fourier transform infrared spectroscopy, scanning electron microscopy, and thermogravimetric analysis. Results showed that CuO-MnOx/OMS-2 was the best catalyst with high removal efficiency of 98.41% at reactor temperature of 250 °C while gas outlet temperature of < 50 °C, proving the suitability of this material for practical treatment of CO in flue gas. The reaction follows Mars-Van-Krevelen mechanism with the presence of Cu^{2+}-O^{2-}-$Mn^{4+} \leftrightarrow Cu^{+}$-$\square$-$Mn^{3+} + O_2$ redox in the structure of the material. Moreover, the effect of environmental factors such as flow rate, inlet CO concentration, and catalysts amount on the CO removal efficiency were investigated and noted for designing and operation purposes. Concentration of outlet CO met well QCVN 19: 2009/BTNMT - National technical regulation on industrial emissions for dust and inorganic substances. Therefore, CuO-MnOx/OMS-2 catalyst material could be a potential catalyst for treatment of CO in flue gas of boiler.

Key words: carbon monoxide, oxidation, catalyst, air pollution control

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INTRODUCTION

Every year, millions of tons of carbon monoxide (CO) emitted into the environment cause serious consequences for human health[1]. In addition to natural emission sources, carbon monoxide (CO) is also generated by incomplete combustion of carbon-containing substances in wood-fired boilers, waste incinerators and other processes. In Vietnam, many small and medium enterprises are using wood boilers as main heat energy source for their production process. Due to the limited budget and low technology, most of the wood boilers are cheap and inefficient and the incomplete combustion results in high concentration of CO in the flue gas. In order to meet the more and more strictly environmental regulations for emissions, several methods have been studied and some have been commercialized[2]. One of the most effective ways to treat CO is oxidation in the presence of a catalyst[3].

Noble metal containing catalysts such as Au/TiO_2, Au/ZrO_2 and Pt/SnO_2 have been used for low temperature CO oxidation. However, due to the high cost and limited availability of noble and precious metals, the research group paid more attention to the goal of preparation and testing using popular metal catalysts for this application. In particular, Cu is widely mentioned because of its high activity for CO conversion at low temperatures[4]. Studies show that the catalytic activity of CuO depends on the oxidation state of Cu and especially on the nature of the carrier. A number of common carriers have been studied including CeO_2, Al_2O_3, zeolite, and OMS-2. The increase in catalytic activity of Cu when carried on reducing oxides like CeO_2 is explained by the synergistic effect due to the good dispersion of CuO on CeO_2 lead to the possibility of reduction at lower temperatures than CuO[5]. Among new catalysts, octahedral molecular sieves (OMS) on the basis of manganese oxide which has a wide range of reactions, especially for oxidation reactions. Currently, there have not been many studies focusing on the treatment of CO in the waste biomass boiler (which has a temperature of about 250 °C) using catalysts that do not contain nobles metals.

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Therefore, in this study, the research group focused on the preparation of different catalytic materials and investigated the activity of these catalysts to apply for oxidation of CO in air at different temperatures. The effect of environmental factor such as inlet CO concentration, gas flowrate, and catalyst amount were also investigated.

MATERIALS AND METHODS

Material synthesis and characterization

All chemical used are analytical-grade from China and Vietnam. Figures of all materials are displayed in **Figure 1**. Manganese oxide octahedral molecular sieves (OMS-2) were synthesized by a hydrothermal method[6] as follows: 11.33 g $MnSO_4.H_2O$ dissolved in 120 mL of deionized water was added to a solution of 7.57 g $KMnO_4$ in 38 mL of deionized water and 4 mL of concentrated HNO_3. The obtained mixture was transferred into a 165 mL Teflon–lined stainless steel autoclave and heated at 160 °C for 24 h. The product was then filtered, washed with deionized water and dried at 105 °C for 12 h. The material was obtained after calcination at 400 °C for 4 h and denoted as OMS-2.

Catalyst of CuO-MnO_x/OMS-2 with 15 wt.% of CuO-MnOx (Cu:Mn molar ratio of 6:4) was prepared by impregnation method. Calculated amounts of $Cu(NO_3)_2.3H_2O$ and $MnSO_4.H_2O$ were dissolved in deionized water and impregnated with OMS–2. The mixture was mixing and sonicated for 30 min, followed by stirring and heated at 80 °C for 15 min. After that, the material was dried at 105 °C for 12 h and finally ground and calcined at 400 °C for 4 h.

CuO/Al_2O_3 catalysts were prepared by impregnation method with copper nitrate aqueous solution of the desired concentrations[7]. At first, 2.82 g of $Zn(NO_3)_2.6H_2O$ and 11.29 g of $Al(OH)_3$ were dissolved in distilled water. The mixture was then dried at 105 °C and calcination in air at 500 °C for 6 h. After that, CuO was impregnated by adding of 5.584 g $Cu(NO_3)_2$ into the mixture, followed by drying at 105 °C and calcination in air at 600 °C for 6 h.

Cr_2O_3/Al_2O_3 sample containing 15.7 wt.% of Cr_2O_3 were prepared by mixing a known mass of finely powdered $Al(OH)_3$ with a calculated amount of CrO_3 solid, followed by drying at 120 °C and calcination in air at 800 °C for 6 h[8].

The materials were characterized by Brunauer–Emmett–Teller (BET, Porous Materials, BET-202A), scanning electron microscopy (SEM) with energy-dispersive X-ray spectroscopy (EDS) (JEOL JSM-IT200), X-ray diffraction (XRD, D2 Phaser, Bruker), Fourier transform infrared spectroscopy (FTIR, Bruker-FTIR), and thermogravimetric analysis (TGA, TGA Q500 machine).

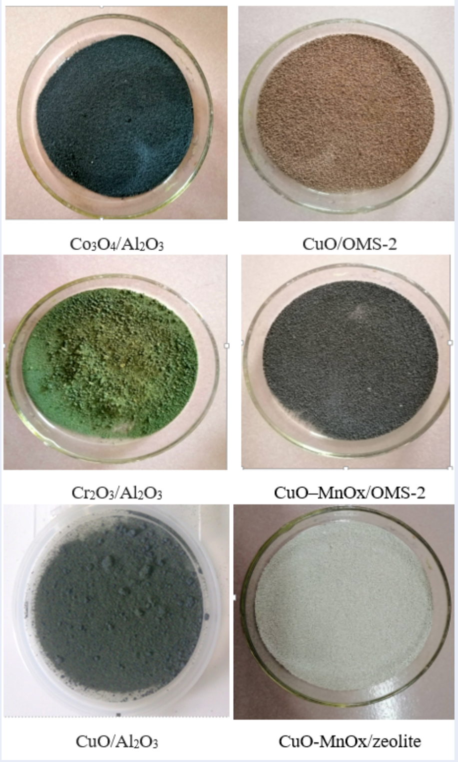
Co_3O_4/Al_2O_3 CuO/OMS-2
Cr_2O_3/Al_2O_3 CuO–MnOx/OMS-2
CuO/Al_2O_3 CuO-MnOx/zeolite

Figure 1: **Pictures of catalysts used in the study**.

Catalytic activity test

The micro-flow reaction system (**Figure 2**) consists of 3 parts: gas supply system, reaction system, and analysis system. Gas flow is supplied from clean gas pump and gas cylinder containing 5% CO balanced in N_2. The CO gas is then diluted with clean air for reaching desire concentration of around 2000 ppm before feeding into the reaction. All gas flowrates were controlled by mass flow controller (MFC) with typical total flowrate was kept stably at around 1 L/min. Catalysts with amount of 0.4 or 1 g and particle size through a 20 - 40 mesh sieve was placed inside the reactor to form a packed column inside the reaction tube. The catalyst was pre-activated in air stream at 250 °C for 30 min every first use of a new catalyst. All the experiments were conducted three times and the average values are presented in the manuscript.

The concentration of CO in the inlet and outlet were continuously monitored by using a portable emissions analyzer (Testo 350 XL, Germany). The efficiency of CO treatment was then calculated as **Equa-**

tion (1) and used as a criteria for evaluation the ability of the catalysts at different reaction temperatures.

$$H = \frac{C_{in} - C_{out}}{C_{in}} \times 100\% \qquad (1)$$

Where H is the removal efficiency (%). C_{in} and C_{out} are the inlet and outlet CO concentration (ppm), respectively.

In this study, different types of catalyst (i.e., Co_3O_4, CuO, Cr_2O_3, and CuO-MnOx) and carrier (i.e., Al_2O_3, OMS-2, and zeolite) was tested in order to find the best material for CO oxidation. Moreover, environmental factors such as temperature (50 - 500 °C), flow rate (0.52 - 1.3 L/min), CO concentration (500 - 4500 ppm), and catalysts amount (0.25 - 1.25 g) on the CO removal efficiency were investigated.

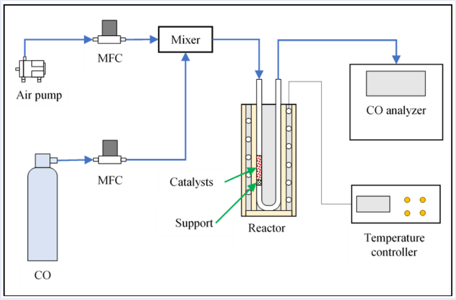

Figure 2: **Experimental set-up for catalytic oxidation of CO.**

RESULTS AND DISCUSSION

Material characterization

The BET surface area of catalysts are summarized in **Table** 1. It can be seen that surface area of OMS-2 support is 62.5 0 m^2/g and that of CuO-MnOx/OMS-2 is 52.76 m^2/g, which was slightly lower than that of OMS-2 support. The decrease in surface area of OMS-2 material after doping with copper oxide-manganese oxide mixture somehow demonstrates the dispersion of metal oxides on the surface of the support. SEM images of OMS-2 materials are presented in **Figure** 3. Both pure and doped OMS-2 materials exhibits nanowires morphology with diameters of about 13 nm and lengths of several μm. **Figure** 4 shows the XRD patterns of OMS-2 catalysts. The diffraction peaks at 2θ of 12.6°, 17.9°, 28.7°, 37.5°, 41.9°, 49.9°, and 60.1° are attributed to the crystalline phase of cryptomelane (KMn_8O_{16}), indicating that the nanowires OMS-2 materials has a cryptomelane-type structure[9].

Table 1: **BET surface are of catalysts**

Catalyst	BET surface area (m^2/g)
OMS-2	62.50
CuO-MnO_x/OMS-2	52.76

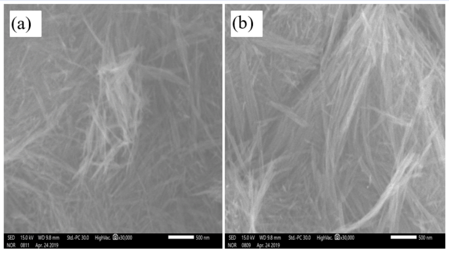

Figure 3: **SEM images of (a) OMS-2, (b) CuO-MnOx/OMS-2.**

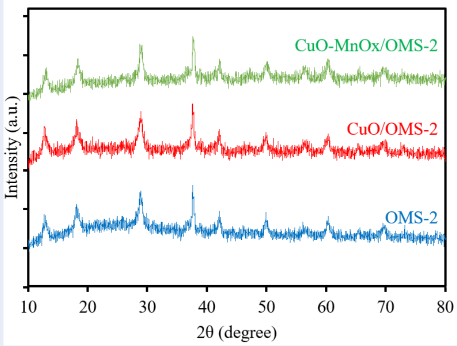

Figure 4: **XRD patterns of OMS-2, CuO/OMS-2, and CuO-MnO_x/OMS-2 catalysts.**

EDS result of CuO-MnOx/OMS-2 is presented in **Figure** 5 while those of all OMS-2 materials are summarized in **Table** 2. The major components of the OMS-2 materials were oxygen and manganese while copper was detected in doped samples. Surface chemical property of the material has strong effect on the catalytic activity of the material[10]. FTIR results in **Figure** 6 present an octahedral structure of OMS-2 with wave number in range of 800 - 400 cm^{-1}[11]. The peak at 470 cm^{-1} is attributed to Mn^{4+} in octahedral structure while peak at 475 cm^{-1} is assigned to the oxygen transfer[12]. The oscillation of Cu-O were observed at peaks of 430, 439, and 461 cm^{-1} for CuO/OMS-2 sample[13,14]. These peaks were also found in FTIR spectra of CuO-MnOx/OMS-2 but with lower intensity, indicating lower amount of copper oxide on the OMS-2 surface. In addition, the present of water was also observed at peaks of 3430 and 1626 cm^{-1}[15].

Thermogravimetric analysis of OMS-2 materials were also done, and the results of CuO CuO-MnOx/OMS-2 is presented in **Figure** 7. The weight loss of around 2% with temperature < 400 °C could be due to the water release while the loss at temperature range of 400 - 700 °C could be the structural decomposition of the materials at high temperature.

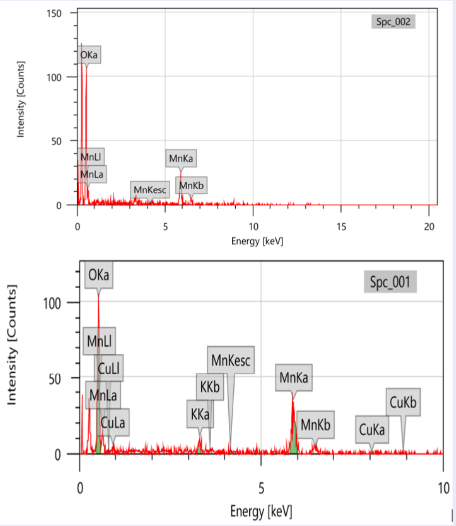

Figure 5: **EDS results of OMS-2 and CuO-MnO_x/OMS-2.**

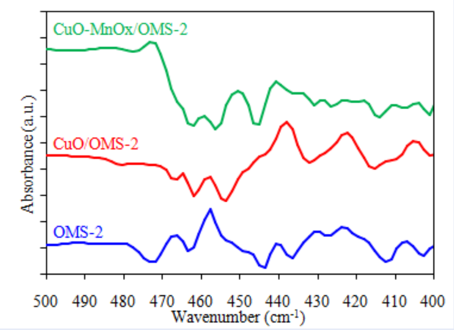

Figure 6: **FTIR of OMS-2, CuO/OMS-2 and CuO-MnOx/OMS-2 catalysts.**

Activity of catalysts based on Al_2O_3 carrier

Experimental results with Co_3O_4/Al_2O_3, CuO/Al_2O_3, and Cr_2O_3/Al_2O_3 catalysts in **Figure** 8 show that CO conversion increases with reaction temperature. For Co_3O_4/Al_2O_3 and CuO/Al_2O_3, the highest conversion efficiency was achieved at 500 °C, reached 99.60% for Co_3O_4/Al_2O_3 and 93.94 % for CuO/Al_2O_3. In t he temperature range of 50 - 250 °C, CO gas is inert and not converted to CO_2. CO conversion started from 300 °C and increased at higher temperatures. For Cr_2O_3/Al_2O_3 material as catalyst, CO conversion did not occur or was very limited even when increasing the reaction temperature. The highest performance at 400 °C temperature was only 11.62%. This proves that the catalytic activity of Cr_2O_3/Al_2O_3 is very low and this material is not suitable for CO conversion in the exhaust gas.

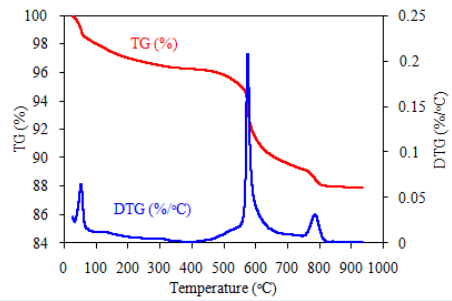

Figure 7: **TGA result of CuO-MnOx/OMS-2 catalysts.**

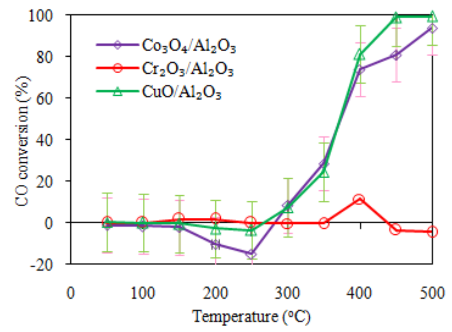

Figure 8: **CO conversion efficiency of Co_3O_4, Cr_2O_3, and CuO catalysts based on Al_2O_3 carrier at different temperatures.**

Activity of catalysts based on OMS-2 and zeolite carrier

The ability of CuO/OMS-2, CuO-MnOx/OMS-2, and CuO-MnOx/zeolite catalysts for CO conversion are shown in **Figure** 9. Similar to Al_2O_3 -based catalysts, CO conversion efficiencies by these catalysts was ineffective at 50 °C and increased with the reaction temperature. For CuO/OMS-2, the highest removal efficiency at 500 °C and reached 73.15%. At the temperature range of 50 - 250 °C, low CO conversion of

Table 2: Atomic percentage of element

Materials	O	Mn	Cu
OMS-2	40.11	59.89	-
CuO/OMS-2	60.40±3.18	36.24±2.97	3.36±1.50
CuO-MnO$_x$/OMS-2	64.45±2.66	30.97±2.14	1.13±0.70

6.44% was observed. And the conversion starts to be effective when the temperature was higher than 250 °C.

For CuO-MnOx/OMS-2 material, CO conversion increases with both the reaction temperature and the amount of catalyst. When more catalyst are used, CO conversion efficiency will be higher and more stable over time. With 0.4 g of material, the highest efficiency was 98.66% (at 600 °C). With 1 g of catalyst, the highest conversion efficiency was 99.96% only at 500 °C. Moreover, it is also observed from **Figure 9** that, the conversion efficiency increases very fast at low temperatures from 50 - 250 °C but the efficiency starts to slow down when temperature over 250 °C and become more stable. The results also proved that CuO-MnOx/OMS-2 materials was effective for CO removal and stable over time. On the octahedral molecular catalytic surface (OMS-2), the Mn-O bonds are relative weak, so that flexible surface oxygen atoms are able to participate in the reaction and restore to its original state when oxygen supply from gas flow is available. Compared to some other catalysts (e.g., Pt-catalytic conversion system reaches 50% at 290 °C[16]), CuO-MnOx/OMS-2 has higher activity due to the combination of Mn and Cu can produce Hopcalite $CuMn_2O_4$ with high oxidizing activity for CO removal.

For CuO-MnOx/zeolite catalyst, the highest efficiency was at a temperature of 500 °C and reached 96.92%. In the temperature range of 50 - 350 °C, CO conversion did not occurred. The conversion of CO starts to be effective at 400 °C but from this temperature or above, the ability of CO conversion increases slowly and stably over time. In the contrary, the treatment efficiency increased very rapidly from 0 to 94.63% in temperature range of 350 - 450 °C. It is obvious that CuO-MnOx/OMS-2 had the highest removal efficiency of 98.41% at low temperature of 250 °C. Therefore, it was chosen as catalyst for further experiments.

In order to clarify the effect of CuO and MnOx on the OMS-2 support, the comparison experiments were conducted using OMS-2, CuO/OMS-2, and CuO-MnO$_x$/OMS-2 materials. As seen in **Figure 10**, CO removal increases with the increase of temperature.

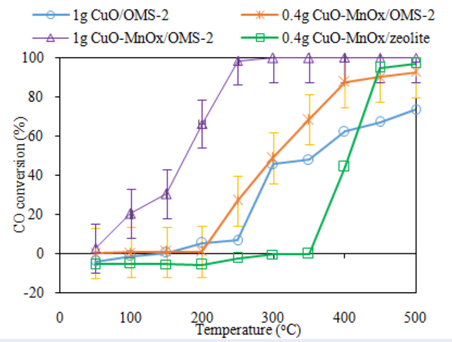

Figure 9: CO conversion efficiency of OMS-2 and zeolite based catalysts at different temperatures.

For OMS-2, CO gas was relative inert at low temperature of 50 - 100 °C while it started to be oxidized to CO_2 at temperature of 150 °C. This can be explained by the low activity of OMS-2 support without the presence of copper, which only reached the removal efficiency of 31.89% at 200 °C. Compared to OMS-2, CuO/OMS-2 and CuO-MnOx/OMS-2 had higher catalytic activity due to the doping of copper with Cu^{2+} -O^{2-} -Mn^{4+} bonds on the support surface. At catalyst surface, CO combines with oxygen in Cu^{2+} -O^{2-} -Mn^{4+} to form CO_2 and leaves an oxygen vacancy (Cu^{+} - o -Mn^{3+}). These oxygen vacancies were then instantly occupied by free oxygen in the air thus act as active sites for reaction of CO[17]. Accordingly, the reaction of CO is continuously occurred on the surface of the catalyst due to the presence of Cu^{2+} -O^{2-} -$Mn^{4+} \leftrightarrow Cu^{+}$ - o -Mn^{3+} + O_2 redox. The oxidation of CO to form CO_2 follows Mars-Van-Krevelen mechanism[6].

Figure 11 demonstrates the effect of calcination temperature on the activity of CuO-MnO$_x$/OMS-2 material for CO oxidation. One can see that the catalytic activity gradually decreases with the increase of calcination temperature from 400 to 600 °C but significantly decreases with further increase of temperature to 700 and 800 °C. Thus, the activity of CuO-MnO$_x$/OMS-2 depends on calcination temperature

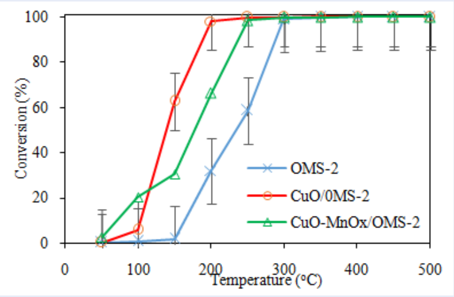

Figure 10: **CO conversion efficiency of OMS-2, CuO/OMS-2 and CuO-MnOx/OMS-2 catalysts at different temperatures.**

and the temperature of 400 °C is suitable for post-treatment of the material in terms of material structure, removal efficiency, and energy consumption.

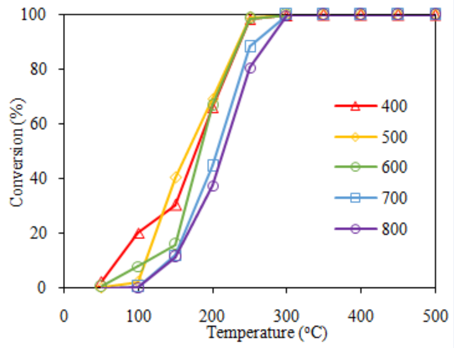

Figure 11: **Effect of calcination tem perature on the a ctivity of CuO-MnO$_x$/OMS-2 catalysts for CO oxidation.**

Effect of other environmental factors

Figures 12, 13, 14 **and** 15 illustrates the effect of operational condition on the CO removal using CuO-MnOx/OMS-2 material. In **Figure** 12, the CO removal efficiency increases with the increase of CO concentration from 526 to 2300 ppm, where it reaches maximum efficiency of 98.41%. This could be explained by the gas bulk mass transfer of CO increase in this low inlet concentration range. However, further increase of CO concentration slightly decreases its removal efficiency, possibly due to the limitation of catalyst surface active sites for CO adsorption and reaction. This should be noted when designing a catalytic system for practical application where CO concentration in flue gas fluctuates from 200 to 5000 ppm. The lower inlet concentration of CO could result in lower removal efficiency while the outlet concentration is required to meet emission standard (QCVN 19: 2009/BTNMT).

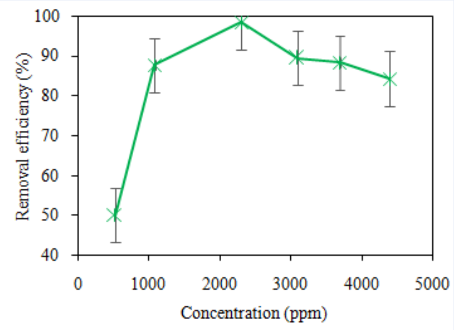

Figure 12: **Effect of inlet CO concentration (1 g CuO-MnOx/OMS-2, 250 °C, 1 L/min, n = 3).**

In actual small wood boilers, the flowrate of flue gas usually fluctuates due to the variation in steam amount need of the production process. **Figure** 13 presents the effect of gas flowrate on the removal efficiency of CO. It is obvious that the removal efficiency continuously decreases with the increase of flowrate. The reason is mostly based on the gas retention time, where higher flowrate means shorter retention time for CO reaction on the catalyst surface. This result also indicates that if a designed catalyst system works well with the highest flue gas flowrate, it could even work better under lower flowrate condition.

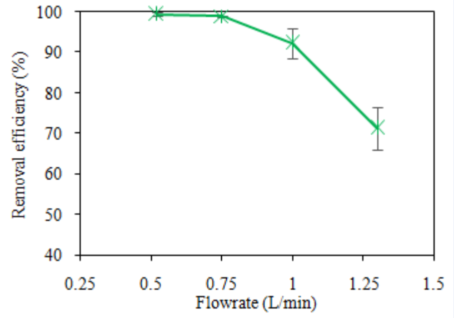

Figure 13: **Effect of inlet gas flow rate (1 g CuO-MnOx/OMS-2, ~2300 ppm CO, 250 °C, n = 3).**

In order to clarify the effect of both flowrate and concentration on the removal ability of CO, the constant amount of CO was maintained while both flowrate and concentration was changed. The result is displayed in **Figure** 14 for flowrate range of 0.5 to 1.0 L/min and CO range of 4132 to 2045 ppm. It can be observed that the removal efficiency is more dependent on the flowrate than on the concentration in the investigated range. This implies that the gas retention time should be a more important designing parameter than the operational concentration of CO concentration in the practical application.

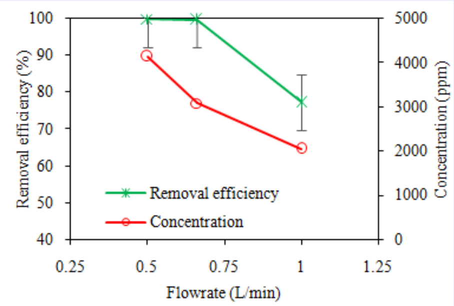

Figure 14: **Effect of both inlet gas flow rate and concentration under constant mass flowrate of CO (1 g CuO-MnOx/OMS-2, 250 °C, n = 3).**

The amount of catalyst used in a practical system is important design criteria. Too much catalyst would take more cost and high pressure drop while too less catalyst result in low efficiency. The effect of catalyst amount on CO removal is demonstrated in **Figure** 15. It is obvious that the efficiency increases from 64.6 to 99.7% with the catalyst amount from 0.25 to 1.25 g due to the more active surface available for CO adsorption and reaction. However, the linear increase of efficiency was not observed for whole investigated range. The amount of 1 g could be the most suitable with the efficiency of 98.4% while 1.25 g is an excess amount with inefficient use by CO concentration of 2300 ppm and gas flowrate of 1 L/min.

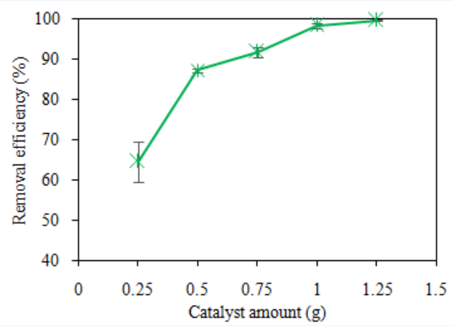

Figure 15: **Effect of catalyst amount (CuO-MnOx/OMS-2, ~23 00 ppm CO, 1 L/min, 250 °C, n = 3).**

CONCLUSION

Catalysts of Co_3O_4/Al_2O_3, Cr_2O_3/Al_2O_3, CuO/Al_2O_3, CuO-MnOx/OMS-2, and CuO-MnOx/zeolite were synthesized by impregnation and hydrothermal methods and applied for oxidation of carbon monoxide gas in air. The results show that most catalysts increased activity with the increase of reaction temperature. Among them, CuO–MnOx/OMS-2 catalyst material had the highest efficiency at 250 °C and stable activity during reaction time. The most suitable operational condition was found to be inlet CO concentration of 2300 ppm CO, gas flowrate of 1 L/min, and catalyst amount of 1 g. These result shows a potential use of CuO–MnOx/OMS-2 material as an effective catalyst for treatment of CO-containing flue gas in practical application.

ACKNOWLEDGEMENT

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ABBREVIATION

CO:	Carbon monoxide	Cácbon monoxít
OMS:	Octahedral Molecular Sieves	Rây phân tử bát diện
OMS-2:	2 × 2 type OMS	Rây phân tử bát diện có cấu trúc đường ống 2 x 2
BET:	Brunauer - Emmett - Teller	Phương pháp đo diện tích bề mặt riêng
SEM:	Scanning Electron Microscope	Kính hiển vi điện tử quét
XRD:	X-Ray Diffraction	Nhiễu xạ tia X
FTIR:	Fourier Transform Infrared	Quang phổ hồng ngoại
TGA:	Thermogravimetric Analysis	Phân tích nhiệt trọng trường
QCVN:	National technical regulation	Quy chuẩn Việt Nam
BTNMT:	Ministry of Natural Resources and Environment	Bộ Tài nguyên và Môi trường

CONFLICT OF INTEREST

There is no conflict of interest regarding this manuscript.

AUTHOR CONTRIBUTION

Nguyen Thi Bich Thao do the experiment, collect and process data, and prepare the manuscript

Nguyen Nhat Huy outline the research, plan the experiment, check and complete the manuscript

REFERENCES

1. Sykes OT, Waker E. The neurotoxicology of carbon monoxide - Historical perspective and review. Cortex. 2016;74:440–448.
2. Li P, Miser DE, Rabiei S, et al. The removal of carbon monoxide by iron oxide nanoparticles. Appl Catal B: Environmental. 2003;43:151–162.
3. Thoảng HS, Lộc LC. Chuyển hóa hidrocacbon và cacbon oxit trên các hệ xúc tác kim loại và oxit kim loại. Hà Nội: Nxb. Khoa học tự nhiên và Công nghệ; 2007.
4. Pillai UR, Deevi S. Room temperature oxidation of carbon monoxide over copper oxide catalyst. Appl Catal B. 2006;64:146–153.
5. Mrabet D, Abassi A, Cherizol R, Do TO. One-pot solvothermal synthesis of mixed Cu-Ce-Ox nanocatalysts and their catalytic activity for low temperature CO oxidation. Appl Catal A: General. 2012;p. 447–448. 60-66.
6. Liu XS, et al. Highly active CuO/OMS-2 catalysts for low-temperature CO oxidation. Chemical Engineering Journal. 2010;162(1):151–157.
7. El-Shobaky HG, Mokhtar M, El-Shobaky GA. Physicochemical surface and catalytic properties of CuO-ZnO/Al2O3 system. Applied Catalysis A: General. 1999;180(1-2):335–344.
8. El-Shobaky HG, Ghozza AM, El-Shobaky GA, Mohamed GM. Physicochemical surface and catalystic properties of Cr2O3/Al2O3 system. Colloids and Surfaces A: Physicochemical and Engineering Aspects. 1999;152(3):315–326.
9. Sithambaram S, Nyutu EK, Suib SL. OMS-2 catalyzed oxidation of tetralin: A comparative study of microwave and conventional heating under open vessel conditions. Applied Catalysis A: General. 2008;348:214–220.
10. Comotti M, Li WC, Spliethoff B, Schth F. Support effect in high activity gold catalysts for CO oxidation. Journal of the American Chemical Society. 2006;128:917–924.
11. Garces L, Hincapie B, Makwana V, Laubernds K, Sacco A, Suib S. Effect of using polyvinyl alcohol and polyvinyl pyrrolidone in the synthesis of octahedral molecular sieves. Microporous and mesoporous materials. 2003;63:11–20.
12. Abulizi A, Yang GH, Okitsu K, Zhu JJ. Synthesis of MnO 2 nanoparticles from sonochemical reduction of MnO 4 in water under different pH conditions. Ultrasonics sonochemistry. 2014;21:1629–1634.
13. Chang YN. Fourier Transform Infrared (FTIR) Analysis of Copper Oxide Thin Films Prepared by Metal Organic Chemical Vapor Deposition (MOCVD). In: MRS Proceedings; 1992. p. 443.
14. Rahman A, Ismail A, Jumbianti D, Magdalena S, Sudrajat H. Synthesis of copper oxide nano particles by using Phormidium cyanobacterium. Indonesian Journal of Chemistry. 2010;9:355–360.
15. Varughese G, Rini V, Suraj S, Usha K. Characterisation and optical studies of copper oxide nanostructures doped with lanthanum ions. Advances in Materials Science. 2014;14:49–60.
16. Luo J, Zhang Q, Huang A, Suib SL. Microporous Mesoporous Materials; 2002. p. 35–36.
17. Pu ZY, Liu XS, Jia AP, Xie YL, Lu JQ, Luo MF. Enhanced Activity for CO Oxidation over Pr- and Cu-Doped CeO2 Catalysts: Effect of Oxygen Vacancies. The Journal of Physical Chemistry C. 2008;112:15045–15051. 2008/09/25.

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Bài nghiên cứu

Nghiên cứu xử lý khí thải chứa CO sử dụng xúc tác tự chế tạo

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TÓM TẮT

Carbon monoxide (CO) là chất gây ô nhiễm không khí độc hại sinh ra từ lò hơi đốt củi vốn được sử dụng rộng rãi trong các doanh nghiệp vừa và nhỏ tại Việt Nam. Việc xử lý khí thải CO đang phải đối mặt với nhiều khó khăn do tính chất trơ của CO và không thể được xử lý bằng các phương pháp hấp phụ và hấp thụ truyền thống và một trong những phương phap xử lý CO có hiệu quả là phương pháp oxy hóa có sử dụng xúc tác. Do đó, chúng tôi đã hướng đến việc điều chế các chất xúc tác khác nhau trên các chất mang khác nhau để xử lý CO trong khí thải, bao gồm γ-Al_2O_3 (Co_3O_4/Al_2O_3, Cr_2O_3/Al_2O_3, và CuO/Al_2O_3), CuO–MnOx/OMS-2, và CuO–MnOx/zeolit. Quá trình thử nghiệm xử lý CO được thực hiện trong một thiết bị phản ứng cố định liên tục ở quy mô phòng thí nghiệm với phạm vi nhiệt độ từ 50 - 550 ^{o}C. Các đặc trưng của vật liệu xúc tác được xác định bằng các phương pháp như đo diện tích bề mặt riêng, nhiễu xạ tia X, phổ tán xạ năng lượng tia X, phổ dao động hồng ngoại, hiển vi điện tử quét và phân tích nhiệt trọng trường. Kết quả cho thấy CuO-MnOx/OMS-2 là chất xúc tác tốt nhất với hiệu suất xử lý cao đến 98,41% ở nhiệt độ thiết bị phản ứng là 250 ^{o}C nhưng nhiệt độ khí đầu ra < 50^{o}C, chứng minh sự phù hợp của vật liệu cho quá trình xử lý CO trong khí thải. Điều này được giải thích tuân theo cơ chế Mars-Van-Krevelen với sự tồn tại của cặp oxy hóa - khử $Cu^{2+}\text{-}O^{2-}\text{-}Mn^{4+} \leftrightarrow Cu^{+}\text{-}\square\text{-}Mn^{3+} + O_2$ trong cấu trúc của vật liệu. Bên cạnh đó, ảnh hưởng của các điều kiện vận hành khác như nồng độ CO đầu vào, lưu lượng dòng khí thải, và khối lượng xúc tác sử dụng lên hiệu quả xử lý CO cũng được khảo sát để đưa ra những đề xuất hợp lý cho việc thiết kế và vận hành hệ thống xử lý CO bằng xúc tác. Nồng độ CO sau quá trình xử lý đạt quy chuẩn QCVN 19: 2009/BTNMT - Quy chuẩn kĩ thuật quốc gia về khí thải công nghiệp đối với bụi và các chất vô cơ. Như vậy, vật liệu xúc tác CuO-MnO_x/OMS-2 là một chất xúc tác tiềm năng trong việc ứng dụng để xử lý CO trong khí thải lò hơi đốt biomass.

Từ khoá: cácbon monoxít, oxy hóa, xúc tác, kiểm soát ô nhiễm không khí

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