International Research Journal of Natural and Applied Sciences
ISSN: (2349-4077)
Impact Factor 7.032 Volume 9, Issue 11, November 2022
Website- www.aarf.asia, **Email** : editor@aarf.asia, editoraarf@gmail.com

Investigation by Tailoring of Mesoporous Co_3O_4(111) supported on Metal Wire Mesh for CO oxidation reaction

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Abstract. A new structured catalyst made up of mesoporous Co_3O_4 nanowires arrays supported on a stainless steel mesh is presented. Asignificant advantage of this material is that the formation and growth of cobalt oxide nanowires takes place on the surface of stainless steel wires. The obtained Co_3O_4 nanowires arrays supported on a stainless steel mesh with crystal plane (111) exhibit better catalytic activity toward CO oxidation, The light-off temperature (10% conversion) of CO oxidation on the catalyst was at 25°C and when the temperature reaches 120°C, the CO conversion ratio reaches 100%, suggesting that it might also be applicable in other fields such as lithium-ion batteries and supercapacitors.

1. Introduction

CO oxidation is one of the most widely investigated reactions in the field of heterogeneous catalysis because of its importance in equally environmental protection and fundamental studies [1-8].

Carbon monoxide (CO) is odourless, colorless and toxic in air, although oxidation of CO is an efficient manner to eliminate of the air pollutant. It is formed from the incomplete burning or partial oxidation of various fuels as coal, gasoline and natural gas. Some specific cases like CO2 lasers also generate CO. It is very essential to remove COfrom enclosed atmospheres and various residue gases. From the point of view of fundamental studies, CO is closely a frequently used probe molecule for studying the catalyst structure,

adsorption/desorption and reaction mechanism. Through the potential competitive adsorption and activation of oxygen, CO oxidation is an exceptional reaction for the evaluation of catalysts with easy operation and low cost. CO oxidation has been studied with various forms of Tricobalttetraoxide (Co_3O_4) catalysts and supported cobalt oxides [9-14]. The catalytic activity of Co_3O_4 depends on morphology, plane exposure and synthesis procedure.

Tricobalttetraoxide (Co_3O_4), a typical spinel-structure transition metal oxide, shows a strong morphology-dependence in the chemical reactions such as CO oxidation [15-20], CH_4 combustion [21], and selective reduction of NO with NH_3 [22]. The reaction rate is familiarly associated with the morphology of the oxide particles (i.e., atomic arrangement). For example Co_3O_4nanorods containing substantial amounts of exposed {110} planes exhibited superior catalytic activity for low-temperature CO oxidation to the spherical particles mainly enclosed by the {111} facets [15].Also, Co_3O_4 nanotubes [23], nanosheets [24], nanowires[25], and nanocubes [26] similarly showed distinct shape effect in CO oxidation. These results clearly confirm that controlling the morphology of nanostructured cobalt oxides is beneficialt expose more catalytically active sites. The relationship between the shape/crystal plane effect and the active sites is therefore a very important link between the structure and property of catalysts.

Diverse groups have prepared Co_3 O_4 nanostructures by different method such as hydrothermal [27-32], solvothermal [33,34], precipitation-oxidation [35], soft reactive grinding route [36], thermal decomposition [37-39].Among the different methods developed for synthesizing cobalt oxide, the ammonia-evaporation-induced method has been regarded as one of the most convenient and practical techniques [28, 29].

In the present work we have used this method to fabricate arrays of nanowires on a metal wire mesh that is used as catalytic.It has been tested in this work in the CO oxidation reaction at temperatures below 200°C.

2. Experimental Section

2.1. Synthesis of mesoporous Co_3O_4 supported on metal wire mesh

Mesoporous Co_3O_4 supported on metal wire mesh was synthesized by the typical procedures reported in the literature(42).Stainless steel wire meshes-supported Co_3O_4 catalyst was synthesized by the ammonia-evaporation-induced method. During standard preparation, 10 mL of 30 wt% ammonia solution 22.5 mL of H_2O and were successively poured without agitation into a Teflon vessel containing 10 mmol of $Co(NO_3)_2.6H_2O$ and 5 mmol of NH_4NO_3. The mixture was then magnetically stirred for half an hour in air. The magnet was then removed from the vessel and a rectangular piece (5 x 3 cm) of stainless steel wire mesh

[29] µm wire diameter and 45 µm screen opening] that had been previously washed in HNO_3 (4 M) at 60 °C for several hours was introduced into the reaction solution. The vessel with the solution and the mesh was covered by a watch glass and heated in an oven for 18 hours at 90 °C as a result that the nanorods could grow. Finally the sample was heated in air at 300°C for 2 hours.

2.2. Characterization techniques

The texture of the as-prepared samples was examined using a scanning electron microscope (Zeiss, DSM 942 model) attached with an energy dispersive X-ray analysis setup (EDAX) and High-resolution TEM (HRTEM) image was recorded using a JEM-2100 electron microscope operating at 200 kV.

2.3. Catalytic investigation

CO oxidation was tested in a flow reactor. Before reaction, Mesoporous Co_3O_4 supported on metal wire mesh was activated at 300°C for 1 h at 5%O_2/He. After the sample was cooled down to room temperature, a feed gas (1%CO/20%O_2/He) was passed over the catalyst with a flow rate of 30 mL/min. The catalyst was heated to the desired reaction temperature and then kept for 1 hour until the catalyst reaction reached a steady state.

The amounts of CO, CO_2 and O_2 in the inlet and outlet streams were analyzed by an online gas chromatograph. CO conversion was calculated from the measured CO concentration using the formula CO conversion = $[(Co_{in} - CO_{out})/CO_{in}]$, where CO_{in} and CO_{out} were the inlet and outlet CO concentration, respectively.

3. Results and discussion

3.1. Morphological study

Figure 1.shows SEM images of the supported Co_3O_4 particles on metal wire mesh. As observed, the spinel particles are arranged as flower-like nanowire arrays that are evenly spaced on the metal wire mesh (Figure1a and 1b). It can be observed that the Co_3O_4 sample is formed by nanorods 30 µm in length and 500 nm wide (Figure1b).

Figure 1c shows a typical HR-TEM image of the synthesized Co_3O_4. A layer separation of about 0.46 nm can be observed, matching to the separation between (111) planes of Co_3O_4 [43]. It indicates that the growing plane and the growing direction of the nanorods are (111) plane.

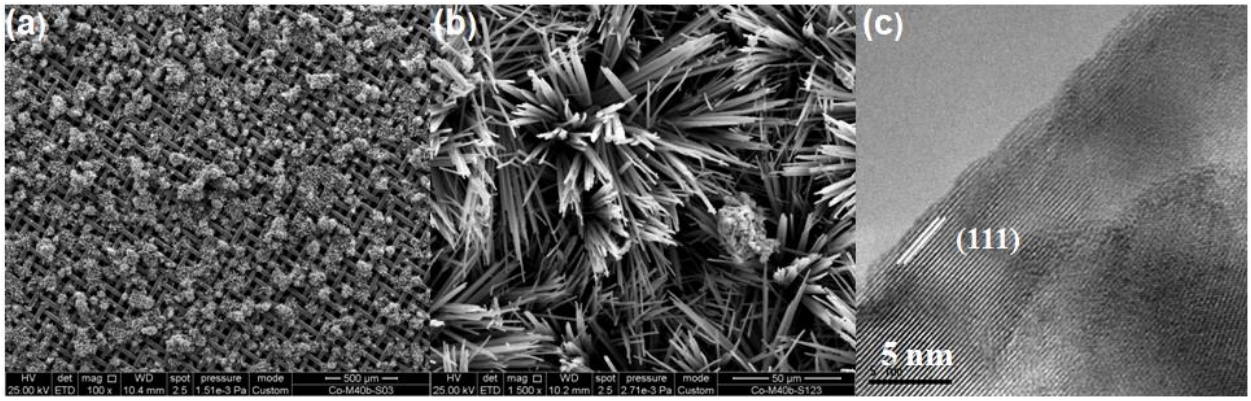

Figure 1. SEM (a, b) and HR-TEM (c) images of the mesoporous Co_3O_4 supported on metal wire mesh.

3.2. Catalytic properties of mesoporous Co_3O_4 supported on metal wire meshin CO oxidation

The CO catalytic performance of the obtained metal wire mesh supported mesoporous Co_3O_4toward CO oxidation is presented in Figure2. As shown in figure, the conversion of CO oxidation over the catalyst increases when the temperature increases.

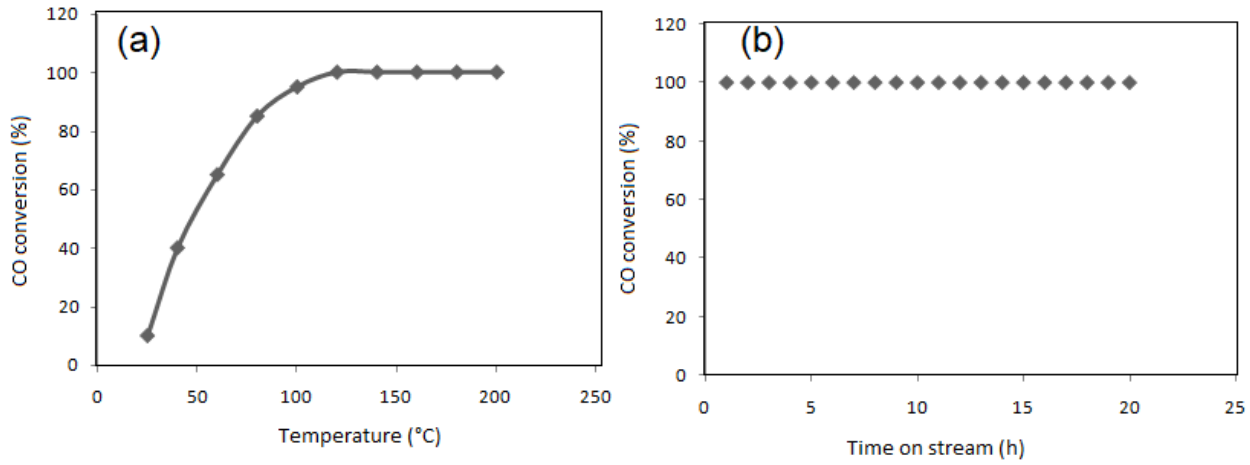

Figure 2.(a) Catalytic activity of mesoporous Co_3O_4 supported on metal wire mesh in the CO oxidation as the function of temperature, (b) durability tests of mesoporous Co_3O_4 supported on metal wire mesh. Reactions conditions: the gas flow was composed of 1%CO/20%O2/He (vol.) with a flow rate of 30 $mL.m^{-1}$.

The light-off temperature (10% conversion rate) at 25°C and when the temperature reaches 120°C, the CO conversion ratio reaches 100% which exhibits higher catalytic activity than that of the Co3O4 nanowires [44] and Co3O4 nanorods [45]. Microdiscs [46] and Co3O4 Urchin-Like [47].

In practical applications the long-term stability of the catalyst is important. We therefore tested the stability of the Co_3O_4 supported on metal wire mesh. The stability of the mesoporous Co_3O_4 supported on metal wire mesh is evaluated at a temperature of 120°C and the result is shown in Figure 2b. The conversion of CO is very stable over the 20 h test. No decreasing tendency is observed in the time-on-stream tests, and a longer reaction time can even be expected. This indicates that the mesoporous Co_3O_4 supported on metal wire mesh is a good catalyst for CO oxidation.

4. Conclusion

Mesoporous Co_3O_4 supported on metal wire mesh has been successfully synthesized via the ammonia-evaporation-induced method. The process is easily conducted and suitable for large- scale production. The obtained mesoporous Co_3O_4 supported on metal wire mesh exhibit better catalytic activity toward CO oxidation compared with those of other porous Co_3O_4 materials reported in the literature. Such obtained mesoporous Co_3O_4 supported on metal wire mesh materials may also be applicable to lithium-ion batteries and supercapacitors.

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