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The catalytic activity of copper oxide nanoparticles towards carbon monoxide oxidation catalysis: microwave – assisted synthesis approach

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ABSTRACT

In this research, we report a simple, versatile, and reproducible method for the synthesis of copper oxide nanoparticles via microwave assisted synthesis approach. The important advantage of this catalyst is due to its important role not only in the low temperature oxidation of CO but also in potential applications in pharmaceutical and fine chemical synthesis. The results reveal that the copper oxide catalyst has particularly a remarkable high activity for CO oxidation catalysis as it was found that copper oxide (CuO) catalyst has 100% conversion of carbon monoxide into carbon dioxide at 175 ^{o}C. This also could be attributed to the high degree of dispersion of the copper oxide nanoparticles with a particle size of 25-35 nm. Those nanoparticles were characterized by various spectroscopic techniques including; X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), and transmission electron microscopy (TEM).

Keywords: *Carbon monoxide oxidation; copper oxide; nanoparticles; microwave synthesis; catalysis.*

1. INTRODUCTION

During the past few decades, metal oxide nanoparticles have been considered as a highly important class of nanostructured materials not only for its attractive, unique and superior physical and chemical properties but also due to its wide applications in catalysis field particularly when combined with other metal nanoparticles[1-4]. The research efforts are focused on those nanostructured materials as they are potential candidates for tremendous applications in catalysis industry, advanced technological and medical applications, and material science [5-9]. It is well established that heterogeneous catalysis based on metal oxides has attracted tremendous research efforts as of its vital role not only in chemicals, refining, and pharmaceuticals industry but also in other critical and strategic applications like removal of harmful gases and volatile organic compounds which are mainly considered as hot topics in controlling the environmental pollution[10-14]. The oxidation catalysis of carbon monoxide into carbon dioxide by oxygen is one of the keys to solve one of the main issues that have a harmful impact on thw environment as it is well known that even small exposure (ppm) to this invisible and odorless gas leads to severe side effects, lethal symptoms and even death. Hence; many scientific research groups are working hard to design new catalytic systems that are basically tailored to be used efficiently in low temperature CO oxidation and then contribute to the efforts exerted in order to control the environmental pollution[5, 7, 15-18].

The use of transition metals at the nanoscale has been reported by several research groups working in the field of air quality and environmental protection. In those reported data, Pd, Pt and Rd nanoparticles were used as the main active catalysts that are capable of converting carbon monoxide into carbon dioxide and hence, reducing or eliminating the potential risk of CO inhalation[1, 19-22]. Among several transition metals, palladium-based nanoparticles and copper-based catalysts have been attractive to several research groups due to their versatile synthetic and wide catalytic applications[23-27]. Palladium metal has been extensively used to catalyze several chemical reaction transformations under both homogeneous and heterogeneous reaction media[28, 29].

Also, the design of bimetallic Pd-based nanoparticles by combination of palladium with other transition metals like Ni, Au, Pt, Ag, Co, and Cu have shown to have diverse applications in organic synthesis and in the three-way catalytic treatment of the exhaust gas from automobiles as well[30-33]. Among several bimetallic nanoparticles, copper oxide-supported transition metal catalysts showed good catalytic activities. Copper is considered as promising transition metal due to many advantages such as abundant reserve, low cost, versatility, less harmful to the environment, and wide use in different applications. In addition, using copper oxide as a solid support for palladium catalysts is highly advantageous by preventing the potential agglomeration of palladium nanoparticles and hence, enhancing the catalytic activity[34-38].

In this scientific research, we used a facile one-step microwave-assisted synthesis approach to prepare copper oxide nanoparticles with an excellent catalytic activity towards carbon monoxide oxidation catalysis. This adopted synthetic approach is a fast, mild, reliable, reproducible, and efficient method to prepare highly active copper oxide nanoparticles using microwave heating.

2. EXPERIMENTAL

The chemicals were used as received. Absolute ethanol (99.9%), deionized water (D.I. H_2O) was used for all experiments. Copper (II) nitrate hemipentahydrate and hydrazine hydrate (80 %) were purchased from Sigma Aldrich.

2.1. Synthesis of Copper Oxide Nanoparticles. A solution containing 150 mg of copper (II) nitrate hemipentahydrate Cu $(NO_3)_2.2.5H_2O$ in 50 mL deionized water was sonicated for 1.5 hr stirred for another 1.5 hr at room temperature. Then, 100 µl of hydrazine hydrate was added to the entire prepared mixture then heated using microwave irradiation for 60 s. The mixture was then filtered, washed with hot deionized water and ethanol and finally, the resulting solid product was dried in the oven at 80˚C untill constant catalyst weight.

2.2. Characterization of Catalyst. The X-ray photoelectron spectroscopy (XPS) analysis was performed on a Thermo Fisher Scientific ESCALAB. While the X-ray diffraction patterns were measured using an X'Pert X-ray diffraction unit. A JEOL JEM-1230 electron microscope was used to capture TEM images.

2.3. Catalysis Measurements. The catalytic performance of the prepared catalyst was evaluated for CO catalytic oxidation; the catalyst sample was placed inside a programmable tube furnace flow reactor as described in previous publications[5]. The temperature of the tested sample was measured by a thermocouple placed near the sample. In a typical experimental procedure, a gas mixture consisting of 3.5% wt. CO and 20% wt. O_2 in helium was passed over the sample while the temperature was ramped. The gas mixture was set to flow over the sample at a rate of 100 cc/min controlled via digital flow meters. The conversion of CO to CO_2 was monitored using an infrared gas analyzer. All the catalytic activities were measured (using 50 mg sample) after a heat treatment of the catalyst at 110 ^{0}C in the reactant gas mixture for 15 minutes in order to remove moisture and any remaining adsorbed impurities.

3. RESULTS AND DISCUSSION

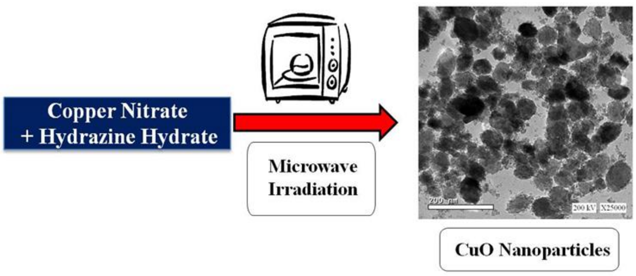

Figure 1. Graphical Abstract of Microwave-assisted synthesis of CuO nanoparticles. (nu este mentionata nicaieri)

The nice dispersion and distribution of CuO nanoparticles (25-35 nm) have a great influence on the catalytic performance of the prepared catalyst as shown in figure 2.

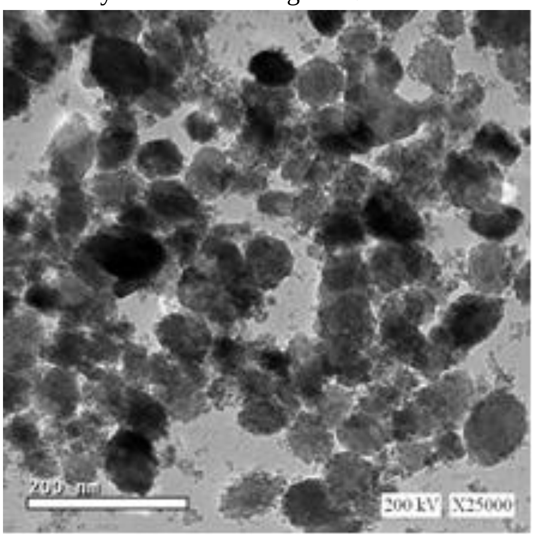

Figure 2. TEM – images of CuO nanoparticles.

Figure 3 displays the XRD pattern of copper oxide nanoparticles (CuO) that was prepared by microwave irradiation method. It was also easily noticed that the XRD reflections of CuO match that of JCPDS no. 48-1548 corresponding to monoclinic structure. The diffraction peaks are ascribed to the (110), (111), (112), (202), (112), and (113) planed of copper oxide NPs as shown in figure 2 which is in good agreement with other reported data [5, 7, 17, 23, 26, 29].

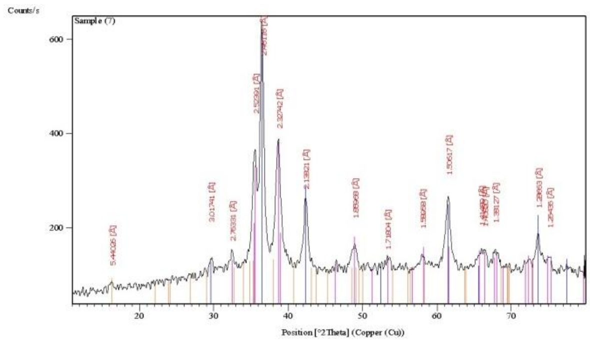

Figure 3. XRD Pattern of CuO nanoparticles.

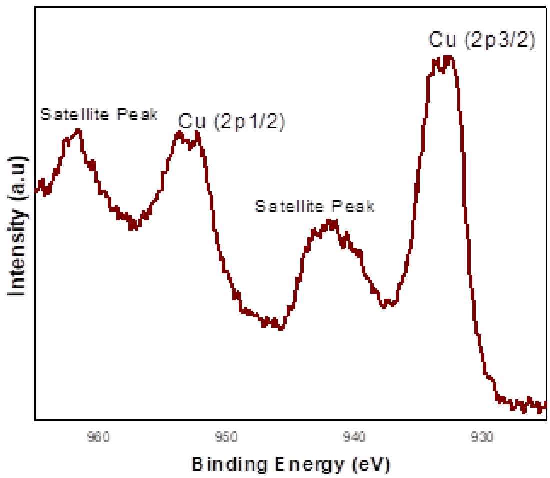

Figure 4. XPS (Cu2p) of CuO nanoparticles.

The XPS technique is widely used as a more sensitive technique for the analysis of surface oxides than XRD. To characterize the surface composition of the supported nanocatalysts, XPS measurements were carried out as shown in Figure 4 for the CuO catalyst. Figure 3 reveals the existence of copper oxide. The XPS show that the binding energy of Cu $2P^{3/2}$ was located at 933.1 eV and the binding energy of Cu $2P^{1/2}$ was located at 953.1 eV, indicating that Copper was present as Cu^{2+}. There is also shake-up satellite peaks located at eV 941.9, 961.7 eV [5, 7, 17, 23, 26, 29].

Figure 5 compares the catalytic oxidation of CO over copper oxide (CuO). The catalytic activity of pure copper oxide (CuO) nanoparticles was measured using tube furnace.

The data shows that copper oxide (CuO) catalyst has 100% conversion of carbon monoxide into carbon dioxide at 175 ^{o}C indicating a high catalytic activity due to the nice dispersion of the copper oxide nanoparticles and hence its decreasing tendency for aggregation and sintering. Therefore, it appears that copper oxide (CuO) catalyst provides reasonable optimization between the adsorption of the CO and O_2 molecules on the copper oxide nanoparticles interfaces in order to allow efficient oxidation of CO.

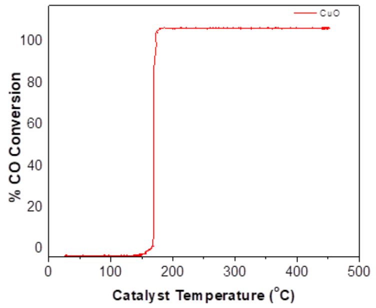

Figure 5. CO catalytic oxidation of copper oxide (CuO) nanoparticles.

4. CONCLUSIONS

In conclusion, a facile one-step approach has been developed for the synthesis of copper oxide (CuO) nanoparticles via microwave-assisted synthesis. The important advantage of this method over other methods is the reproducibility of the final product in order to avoid other issues raised by other techniques. Furthermore, the same method can be used to synthesize bimetallic nanoalloys supported on metal oxide nanoparticles as nanocatalysts for carbon monoxide oxidation. The current research results reveal that the copper oxide (CuO) catalyst has particularly high activity for CO oxidation. Optimizations of the particle size, chemical composition, and shape of those catalysts could help to design a new family of efficient nanocatalysts for the low temperature oxidation of CO. We are currently exploring other effects of microwave heating duration; different supports including carbon based structures as well as the effect of particle size, and shape of the supported metal nanoparticle catalysts.

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