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<https://doi.org/10.15017/1906403>

出版情報 : Proceedings of International Exchange and Innovation Conference on Engineering & Sciences (IEICES). 3, pp.127-129, 2017-10-19. Interdisciplinary Graduate School of Engineering Sciences, Kyushu University

バージョン :

権利関係 :

Hetero-interface of CeO₂ Nanoparticles Single Crystal On MgO Nanowires Surface

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Abstract: *The interface has a synergetic effect in catalytic reactions. Metal oxides such as MgO and CeO₂ are active catalysts in CO oxidation; however, there are limitations in their activity. We assume the combined interface of MgO and CeO₂ can be integrated to produce a better active catalyst. Although there is a 28% lattice mismatch between MgO-CeO₂, we have successfully deposited CeO₂ nanoparticles on MgO nanowires by the VLS method using the PLD technique. Here for the first time we show the images of the deposited nanoparticles obtained by SEM and EDS accompanied with their equivalent Photo-luminescence graphs. The paper discusses in depth how the interface between decorated MgO-nanowires with CeO₂-nanoparticles started nuclei and how temperature and oxygen partial pressure affected their sizes. This work will serve as a base for future studies on other bimetal oxides interfaces*

Keywords: Nanowires; Nanoparticles; Catalyst, CO Oxidation

1. INTRODUCTION

Bimetal oxide active catalyst is essential for safely remove harm emissions such like CO as part of green chemistry. Over the last years, there are many studies for several catalysts which active for CO oxidation [1,2]. Bulk metal oxide are inactive in low temperature and moisture while metal catalyst is more resistive to moisture [3]. Nanoparticles catalyst shows better activity due to its surface area, some metal addition to them shows higher activity according to synergetic effect of two materials [3]. Nano-rods of some material have ability to be redox are good catalyst for Oxidation reaction such like Co₃O₄, Fe₂O₃ and CeO₂ [1,4,5]. It is reported that molecules are easily dissociated on Nanowires surfaces [7] however, they become still inactive at lower temperature due to desorption process. Nanowires core and shells of different material like NiO/MgO[6,12], MgO/Fe₃O₄[8], Ge/Si[9], MgO/TiO [10], and ZnO/ MgO[11] show different unique properties. Interface of NWs and NPs have been fabricated from different Metal oxides such as MgO and CeO₂ which are active catalysts for CO oxidation[6,13,17]. MgO Nanowires surface could be controlled by using MgO (100) Substrate as long as surface activity can be changed from plane to another due to its oxygen defects [14,15,16]. CeO₂ has ability to be redox and hence good catalyst for oxidation [2]. Although CeO₂ crystal ($A = 5.411 \text{ \AA}$) is 28% mismatch from MgO crystal ($A = 4.212 \text{ \AA}$) [18], we successfully fabricate MgO nanowires which have been decorated with CeO₂ Nanoparticles as the first step to study interface catalytic activity. VLS method has been used for NWs growth and NPs decoration. Here we reported Nanoparticles sizes behaviors versus various deposition temperatures and oxygen partial pressure with their photo-luminescence graphs. This study is important to well control NPs morphology and size for better understanding bi-metal oxides interface.

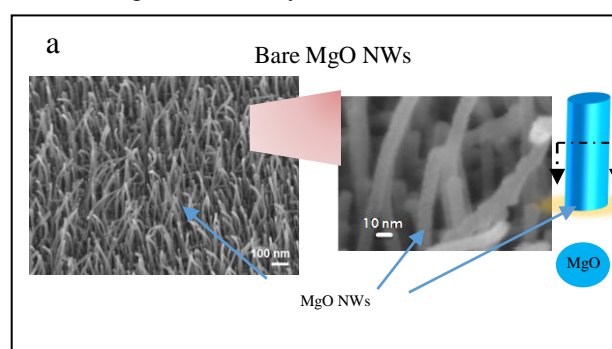
2. EXPERIMENTAL

2.1 Methodology

MgO Nanowires well defined surface fabricates and grows by using gold catalyst droplets (1nm thickness) on single crystal of MgO(100) substrate [15,16,20]. Vapor-liquid- Solid (VLS) method was used for both Nanowires and Nanoparticles deposition by using pulsed ArF excimer laser $\lambda = 193 \text{ nm}$ with energy intensity 40 MJ and frequency 10 Hz which enough to energize Mg-metal target. A fixed total pressure is (10Pa) during the growth after creating high vacuum (10⁻⁵ Pa) with Oxygen partial pressure (10⁻³) in Ar gas [21]. Pre-annealing was performed to 500 °C for 10 minutes then to 750°C. Nanowires length could be controlled according to the growth time ($\sim 5 \mu\text{m} = 5 \text{ hours}$) [19]. In our study CeO₂ Nanoparticles were been fabricated in same conditions of pulsed laser to energize CeO₂ target in high deposition rate (100nm/min) for 1 minute. Temperature and oxygen partial pressure were changed to characterized the particle size by field emission scanning electron microscopy (FESEM) JEOL JSM-7610 while energy dispersive spectroscopy (EDS) was used to evaluate the particles distribution along the wires. Photo-luminescence characterized the defects states of different morphology of them.

2.2 Results and Discussion

Figure 1a, 1b and 1c shows schematic illustration of bare MgO Nanowires versus MgO-NWs/CeO₂-NPs with related FESEM images. Cerium atom from CeO₂ seems to start nucleation by bonding to oxygen atom of MgO on surface, then the crystal of CeO₂ would formulate with 10% strain due to 28% mismatch between MgO & CeO₂ crystal [18].



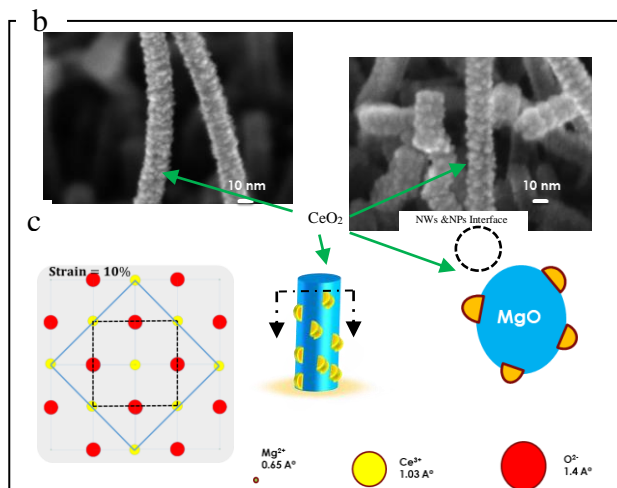


image of Bare MgO nanowires.
(b) SEM Image of Interface MgO NWs decorated with CeO₂ NPs. (C) shows schematic illustration for interface nucleation.

It is very important to put into our consideration that both surface of MgO and CeO₂ are exposed to surrounding. CeO₂ NPs appears successfully distributed along MgO NWs. STEM Image with energy dispersive spectroscopy (EDS) where interface is clear are shown in Figure 2a, 2b.

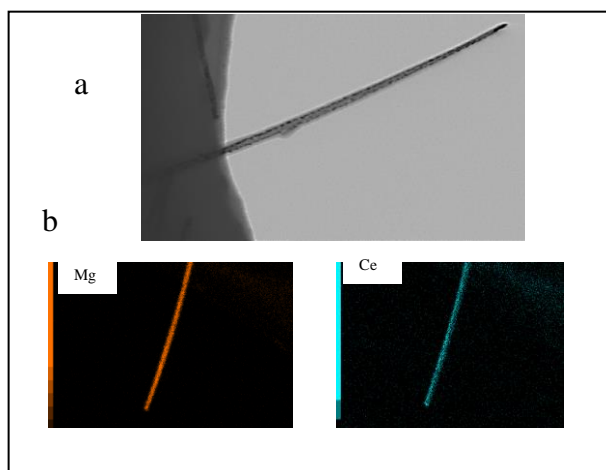


Fig 2. (a) CeO₂ NPs clearly appear on MgO NWs
(b) successful distribution of CeO₂ along MgO NWs

Particle size changes according to different Temperature and different Po₂ as shown in Figure 3. NPs tend to be smaller and smoother in higher temperature according to lower contact angle between particles and surface. MgO and CeO₂ have almost the same surface energy (1.2 J/m²) [22,23]. According to surface thermodynamics higher temperature will lead to more negative gibbs surface free energy which would make particle smoother [24]. By increasing oxygen pressure it seems to be harder on surface to start nucleation process as a result of lack of Cerium atoms which is essential to grow CeO₂ crystal where Ce:O are 12:8 respectively.

Photo-luminescence of Bare MgO NWs Vs MgO/ CeO₂ at oxygen defects at Po₂ =10⁻³ versus different temperatures of (RT to 800 °C), and at 800 °C versus different oxygen partial temperatures in figure 4a, 4b. All graphs have same peaks with different intensity.

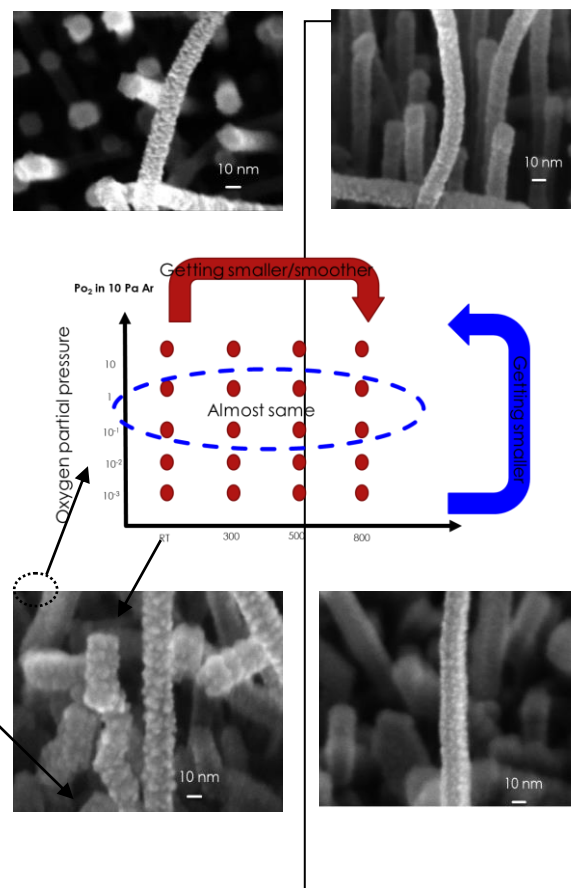


Fig 3. shows Different CeO₂ NPs sizes verses temperature deposition & Po₂.

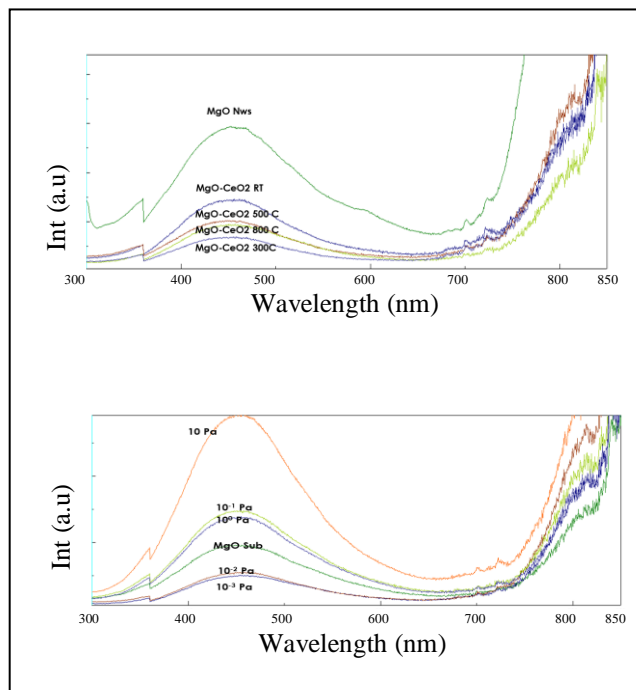


Fig 4. (a) PL MgO-CeO₂ NWs VS Temp at Po₂ =10⁻³.
(b) PL MgO-CeO₂ NWs VS Po₂ at 800 °C.

3. CONCLUSION

MgO Nanowires have been successfully decorated with CeO₂ Nanoparticles for first time in spite of big mismatch 28% between their crystals. NPs size and distributions have been investigated as first stage of study catalytic effect of Bi-metal oxide interface to be a base to study catalytic effect of interface for CO

oxidation. That would require to well understanding NPs fabrication to well control the interface in well-defined surface. MgO shows good controllability for Nanowires fabrications while CeO₂ has ability to be redox which make them good candidates to study their interface for CO oxidation.

Deposition temperature and oxygen partial pressure effect on NPs size. High oxygen partial pressure makes the nucleation process harder and decrease CeO₂ crystal growth due to CeO₂ crystal Ce:O 12:8 respectively which would lead to smaller particles. High Temperature increases the negativity of Gibbs surface free energy which would lead to smoother surface. Photo-luminescence defects of Bare MgO NWs Vs MgO/ CeO₂ shows same peaks but different intensities. This study will be base to fabricate more NPs different metal oxide with unique properties such higher catalytic activity.

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