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ANALYSIS OF STRUCTURE AND ANTIMICROBIAL ACTIVITY OF CeO_2 AND Nd_2O_3 NANOPARTICLES

Hanif Yuliani[1,*], Rina Dewi Mayasari[1], Eryanti Kalembang[1], Yelvia Deni[1], Defi Rahma Santi[2], Putri Widya Pangestika[2], Setyo Purwanto[3], Bambang Sugeng[4], Suyanti[4], Moch. Setyadji[4]

[1]*Pusat Teknologi Material, Badan Pengkajian dan Penerapan Teknologi, Gedung 224, Puspiptek, Tangerang Selatan, Banten 15314, Indonesia*
[2]*Program Studi Fisika, Institut Teknologi Kalimantan, Jl. Soekarno-Hatta Km 15, Balikpapan, Kalimantan Timur 76127, Indonesia*
[3]*Pusat Sains dan Teknologi Bahan Maju, Badan Tenaga Nuklir Nasional, Gedung 42, Puspiptek, Tangerang Selatan, Banten 15314, Indonesia*
[4]*Pusat Sains dan Teknologi Akselerator, Badan Tenaga Nuklir Nasional, Jl. Babarsari, Yogyakarta 55281, Indonesia*

*Corresponding Author Email: hanif.yuliani@bppt.go.id

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ABSTRACT

Cerium oxide (CeO_2) and Neodymium oxide (Nd_2O_3) nanoparticles using local content have been synthesized by precipitation method. The CeO_2 and Nd_2O_3 nanoparticles were characterized by X-Ray Diffraction (XRD) and Fourier Transform Infrared (FTIR) to analyze the material phase and structure. The XRD spectrum shows that CeO_2 and Nd_2O_3 nanoparticles have face-centered cubic and hexagonal, and cubic, respectively. The anti-microbial activity of CeO_2 and Nd_2O_3 nanoparticles was analyzed by diffusion method using gram-negative bacteria (E. coli, S. aureus, P. aeruginosa), and gram-positive bacteria (S. entericatyphi, L. monocyogenes), and fungus (C. albicans). The result confirms that CeO_2 and Nd_2O_3 nanoparticles have the capability of microbial pathogen inhibition. The CeO_2 nanoparticles have the effective activities of inhibition for the microbial of S. aureus and S. entericatyphi, whereas Nd_2O_3 nanoparticles can inhibit the microbial of P. aeruginosa, S. entericatyphi, and L. monocyogenes.

Keywords: CeO_2, Nd_2O_3, face-centered cubic, hexagonal, anti-microbe

INTRODUCTION

Nowadays, nanotechnology, primarily nanomaterials research and development, has been widely used for various applications in daily life, such as medicine and food [1], sensors [2], energy [3], and others. In the medicine application, the nanomaterials developed for antibiotic or antifungal applications, mainly based on rare earth elements (REE) as the raw materials [4]. Indonesia has a lot of natural resources of cerium oxide (CeO_2) and neodymium oxide (Nd_2O_3) classified in REE [5]. The CeO_2 and Nd_2O_3 have great potential to produce new various antibiotics [6-8].

Several previous studies have reported anti-microbial activity on REE nanomaterials [6-7,9]. Parvathya and Venkatramanb have investigated the differences in the synthesis methods due to the antimicrobial activity, i.e., green synthesis (G-CeO_2) and chemical synthesis (C-CeO_2). The results showed that G-CeO_2 nanoparticles had higher activity than C-CeO_2 against Escherichia coli, Pseudomonas aeruginosa, Streptococcus pneumonia, and Proteus Vulgaris bacteria [7]. The CeO_2 nanoparticles also obtain a good antibacterial activity towards both gram-negative and positive bacteria because it has Ce^{3+} ions and rich surface oxygen vacancies [8].

In this paper, we reported the materials properties and anti-microbial activities of CeO_2 and Nd_2O_3 nanoparticles. Anti-microbial activities of the nanoparticles were tested with six types of microbes, i.e., Staphylococcus aureus, Escherichia coli, Pseudomonas aeruginosa, Salmonella entericatyphi, Listeria monocytogenes, and Candida albicans. The results of this research are the preliminary study of REE research for antibiotic applications which are expected to be new potential antibiotics.

METHOD

Synthesis of CeO_2 and Nd_2O_3 Nanoparticles

The CeO_2 and Nd_2O_3 nanoparticles have been synthesized in the Laboratory of Center of Technology for Material BPPT by precipitation method using carbonate ($NaHCO_3$) and hydroxide (NH_4OH) precursors. Synthesis of nano-CeO_2 was carried out using 0.03 M cerium nitrate hexahydrate ($Ce(NO_3)_3.6H_2O$), 0.02 M NH_4OH, and 0.03 M $NaHCO_3$. Those solutions were mixed at the temperature of 55°C for 15 minutes and followed by drying at the temperature of 220°C for 2 hours and calcination at the temperature of 600°C for 3 hours [10]. Synthesis of nano-Nd_2O_3 used the same synthesis process as nano-CeO_2 with neodymium nitrate hexahydrate ($Nd(NO_3)_3.6H_2O$) precursor.

Material Characterization and Anti-microbial Test

The material structures were characterized by *x-Ray diffraction* (XRD) Rigaku and *Fourier Transform Infra-red* (FTIR) Thermo Scientific Nicolet iS50. The anti-microbial activities of the samples were analyzed in the Laboratory of Microbiology, Center of Technology for Pharmaceutical and Medical BPPT, using well diffusion method against and six pathogen microbes (Candida albicans, Staphylococcus aureus, Listeria monocytogenes, Salmonella entericatyphi, Escherichia coli, Pseudomonas aeruginosa) from the collection of the Inter-University Research Center (PAU) ITB. Kloramfenicol antibiotic was used simultaneously for positive control.

RESULT AND DISCUSSION

Crystal Structure of CeO_2 and Nd_2O_3 Nanoparticles

FIGURE 1 shows the X-ray diffraction (XRD) pattern of the samples of CeO_2 and Nd_2O_3 nanoparticles. Analysis of the XRD profile for nano-CeO_2 (bottom) using Match and Rietveld program reveals that the samples formed 58.5% CeO_2 phase (ICDD 98-002-8753) with face center cubic structure and space group of F m -3 m (225). Besides, the formed minor phase is 41.5% thermonatrite compound ($Na_2CO_3 \cdot H_2O$) with an orthorhombic structure and space group of P c a 21 (29).

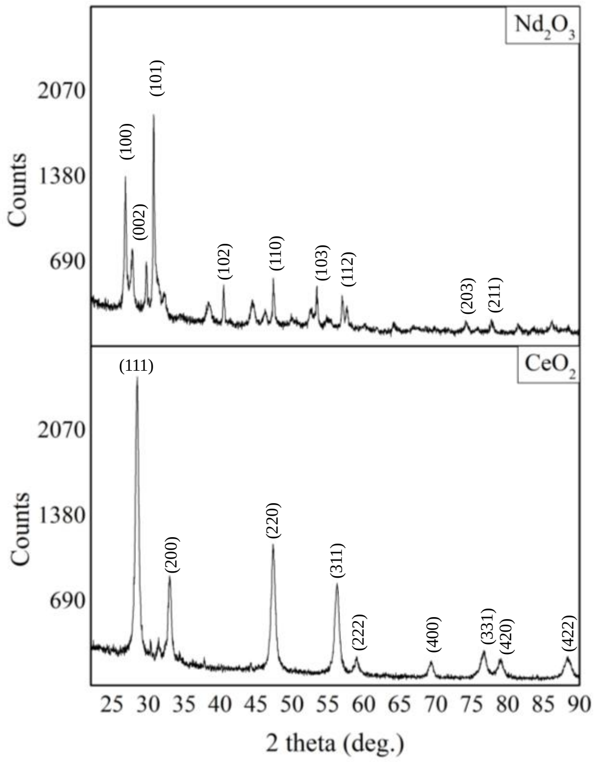

FIGURE 1. The XRD spectrum of CeO_2 and Nd_2O_3 Nanoparticles.

The data analysis of the XRD pattern for nano-Nd_2O_3 (top) indicates 2 (two) Nd_2O_3 phases, i.e., 39.6% Nd_2O_3 with cubic structure (ICDD 98-064-5664) and space group of I a -3 (206), and 25.4% Nd_2O_3 with hexagonal structure and space group of P 63/m m c (194) (ICDD 98-003-2514). The residues are the impurities of 11.9% *nitrate* ($NaNO_3$) phase with a space group of R -3 c (167) (ICDD 98-006-4868) and 23.1% *neodymium hydroxide* with a space group of P 63/m (176) (ICDD 98-000-0398).

Functional Group Analysis of CeO_2 and Nd_2O_3 Nanoparticles

FIGURE 2 shows the result of FTIR analysis for both CeO_2 and Nd_2O_3 nanoparticles. The representation of FTIR absorbance peaks is summarized in TABLE 1. The FTIR result of nano-CeO_2 confirmed the XRD result in which the contained impurity is thermonatrite ($Na_2CO_3 \cdot H_2O$) compound at a wavenumber of 1107.61 cm^{-1}, 1429.28 cm^{-1}, 2360.79 cm^{-1}, and 2978.71 cm^{-1} which represent the vibrational bond of C–O, C=O, and O–H, respectively. The CeO_2 compounds were detected at the wavenumber of 549.08 cm^{-1}, 616.97 cm^{-1}, and 864.20 cm^{-1}. The other FTIR spectrum of CeO_2 detected Ce-O stretching band at 475 cm^{-1}, 545 cm^{-1}, and 615 cm^{-1} [11].

The FTIR result of nano-Nd_2O_3 also validated the XRD result in which nano-Nd_2O_3 has the same impurities, i.e., Sodium Nitrate ($NaNO_3$) at the wavenumber of 1367.82 cm^{-1} and 1489.99 cm^{-1} which denote the vibrational bond of N–O. Furthermore, the wavenumber of 3606.11 cm^{-1} came from O–H bond of NdOH. Nd_2O_3 compounds were spotted at the wavenumber of 534.29 cm^{-1}, 667.53 cm^{-1}, and 856.77 cm^{-1}. The similar result of the FTIR spectrum in Nd_2O_3 have been reported by Yuvakkumar and Hong (2015) [8].

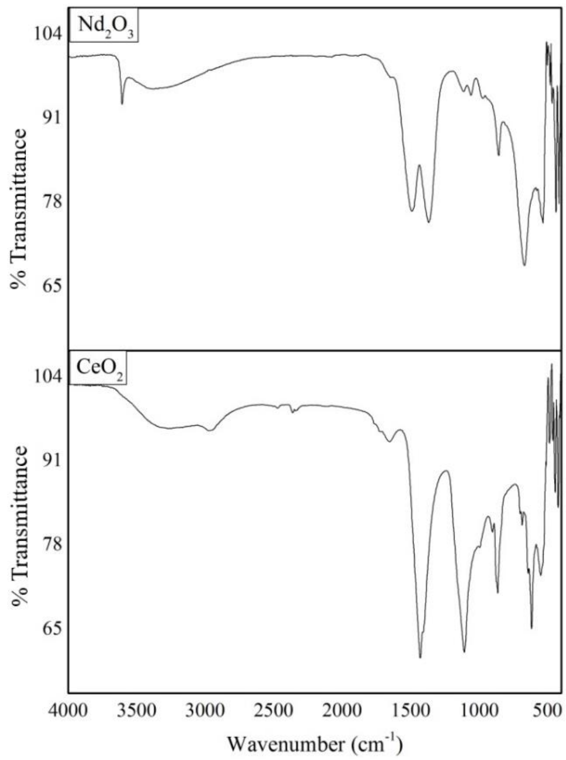

FIGURE 2. The FTIR spectrum of CeO_2 and Nd_2O_3 Nanoparticles.

TABLE 1. The representation of the FTIR spectrum in the Nd_2O_3 and CeO_2 nanoparticles.

Compound	Wavenumber (cm^{-1})	Absorbance	Representation of functional group
CeO_2	549.08	Ce – O	CeO_2
	616.97	Ce – O	CeO_2
	864.20	Ce – O	CeO_2
	1107.61	C – O	$Na_2CO_3 \cdot H_2O$
	1429.28	C = O	$Na_2CO_3 \cdot H_2O$
	2360.79	O – H	$Na_2CO_3 \cdot H_2O$
	2978.71	O – H	$Na_2CO_3 \cdot H_2O$
Nd_2O_3	534.9	Nd – O	Nd_2O_3
	667.53	Nd – O	Nd_2O_3
	856.77	Nd – O	Nd_2O_3
	1489.99	N – O	$NaNO_3$
	1367.82	N – O	$NaNO_3$
	3606.11	O – H	NdOH

Anti-Microbial Activities of CeO_2 and Nd_2O_3 Nanoparticles

The results of the anti-microbial activity test of nano-CeO_2, nano-Nd_2O_3, and positive control were summarized in TABLE 2. From the result analysis, nano-CeO_2 and nano-Nd_2O_3 have higher inhibition ability than Control + to against bacteria (Gram + and Gram -) and fungi.

TABLE 2. Anti-microbial activities of CeO_2 and Nd_2O_3 nanoparticles.

Types of Microbes	Inhibition Zone Diameter (mm)		
	Control (+)	**CeO_2**	**Nd_2O_3**
Candida albicans	0.34	3.00	1.33
Staphylococcus aureus	26.13	4.00	2.33
Listeria monocytogenes	9.99	3.00	3.67
Salmonella entericatyphi	4.72	4.00	5.67
Escherichia coli	14.54	0.67	3.00
Pseudomonas aeruginosa	3.58	4.28	3.67

Babenko et al. have reported inhibitory activity of nano-CeO2 towards Candida albicans, the interaction between nano-CeO_2 and fungi cell surface causes the irreversible change of cell structure and generate blocking capability for fungi enzymatic activity [11].

TABLE 3 shows the review of the inhibition test of nano-CeO_2 and nano-Nd_2O_3 against Gram + (*S. aureus*). The same result also has been researched by Reddy Yadaf et al. and Malleshappa et al. [13-14]. Meanwhile, TABLE 4 shows the inhibitory activity of nano-CeO_2 nano-Nd_2O_3

towards Gram – (*E. coli*). A similar result also has been obtained by Malleshappa et al. [14]. Moreover, the inhibitory activities of nano-CeO_2 and nano-Nd_2O_3 against Gram – (*P. aeruginosa*) have been reported by Ravishankar et al. [15], which have a comparable result of TABLE 5.

TABLE 3. Anti-microbial activities of CeO_2 and Nd_2O_3 nanoparticles towards gram-positive (*S. aureus*) bacteria.

***S aureus* strain**	**Inhibition Activity**		
	Concentration (mg/mL)	**Test Result**	**Referensi**
NCIM-5022	10	1.67	[12]
	10	3.33	
NCIM-5022	10	0.53	[13]
	10	1.47	
PAU ITB	10	4.00	Sample CeO_2
	10	1.33	Sample Nd_2O_3

TABLE 4. Anti-microbial activities of CeO_2 and Nd_2O_3 nanoparticles towards gram-negative (*E. coli*) bacteria.

***E. coli* strain**	**Inhibition Activity**		
	Concentration (mg/mL)	**Test Result**	**Reference**
NCIM-5022	10	2.67	[13]
	10	4.67	
PAU ITB	10	0.67	CeO_2
	10	3.00	Nd_2O_3

TABEL 5. Anti-microbial activities of CeO_2 and Nd_2O_3 nanoparticles towards gram-negative (*P. aeruginosa*) bacteria.

***P aeruginosa* strain**	**Inhibition Activity**		
	Concentration (mg/mL)	**Test Result**	**Reference**
NCIM-2242	10	3.33	[4]
	15	3.57	
	20	4.50	
PAU ITB	10	4.28	CeO_2
	10	3.67	Nd_2O_3

The anti-microbial mechanism of CeO_2 nanoparticles has been reported by Passos Farias et al. [4], microbial inhibition activity of CeO_2 nanoparticles is caused by the *oxidative stress* of microorganism cell membrane.

CONCLUSION

The CeO_2 and Nd_2O_3 nanoparticles have a high anti-microbial activity towards pathogen microbes, i.e., gram-positive bacteria (*Staphylococcus aureus*, *Listeria monocytogenes*), gram-negative bacteria (*Pseudomonas aeruginosa*, *Escherichia coli*, *Salmonella entericatyphi*), and fungi (*Candida albicans*).

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