

EVALUATION OF ACOUSTIC PARAMETERS OF CeO₂ - ETHYLENE GLYCOL NANOFLUIDS FOR DIFFERENT CONCENTRATIONS

V. S. SAI KUMAR^a, K. V. RAO^{b*}, CH.PRASAD^b

^a*Department of Physics, Guru Nanak Institute of Technology, Ibrahimpatnam, 501506, Telangana, India*

^b*Centre for Nano Science and Technology, Institute of Science and Technology, Jawaharlal Nehru Technological University Hyderabad, Hyderabad 500085, Telangana, India*

The present work investigates the synthesis of Cerium Oxide nano particles synthesized by a simple solution combustion synthesis using Cerium nitrate as a precursor material and Urea as a fuel. The prepared CeO₂ nanoparticles were characterized by X-ray diffraction, scanning electron microscopy, EDX analysis. Additionally as prepared CeO₂ nanoparticles were suspended in ethylene glycol, in order to obtain CeO₂-Ethylene glycol nanofluid. The nanofluids were prepared by the dispersion of CeO₂ nanoparticles in Ethylene glycol solution using an ultrasonicator. CeO₂-Ethylene glycol nanofluid additionally characterized with nanofluid interferometer for the evaluation of ultrasonic parameters like adiabatic compressibility (β), acoustic impedance (Z) and thermal conductivity (k).

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1. Introduction

Nanofluid is the name presented by Argonne National Laboratory to illustrate the suspension of nanoparticles in a base fluid. Base fluids such as water, ethylene glycol and engine oil have very low thermal conductivities. The thermal conductivity of nanometer sized particles is typically in the order of magnitude higher than those of the base fluids. The addition of nanoparticles to the base fluid even at low volume concentrations, results in significant increases in thermal performance.[1-6] Recently, an increase in the thermal conductivity coefficient of nanofluids which contain a little amount of metallic particles like Cu or nonmetallic particles like CeO₂, Al₂O₃, and SiC had also been reported. The natural potential of nanomaterials which are dispersed in base fluids for heat transfer also has an important effect on the rate of increase in thermal conductivity coefficient of nanofluids. For example, carbon nanotubes, due to their high thermal conductivity coefficient, are known as an ideal material for making nanofluids.[7-8]

Ceria (CeO₂) is an oxide with important and vast applications in areas of catalysis, electrochemistry, photochemistry, and materials science [9-13]. Cerium oxide has outstanding physical and chemical properties therefore it is used as LPG sensor as well as electrolyte materials for solid fuel cells [14-16]. Also it is highly efficient ultraviolet (UV) absorber to protect light-sensitive materials, as a coating material for protection of corrosion of metals, as an oxidation catalyst and as a counter electrode for electrochemical devices [17-19].

In this paper a simple synthesis method is proposed to produce CeO₂ nanoparticles. We report the solution combustion synthesis method using cerium nitrate as an oxidizer and urea as a fuel. The nano particles were characterized by X-ray diffraction (XRD) analysis, Scanning electron microscopy (SEM) and Nanofluid interferometer.

*Corresponding author: seshusai7@gmail.com

2. Experimental

2.1 Preparation of CeO₂ nanoparticles

Solution combustion synthesis has been recognized as the cost effective and easy synthesis method for nanoparticles with high purity and homogeneity. Cerium oxide nanoparticles were successfully prepared by Solution combustion synthesis. As per the propellant chemistry, in combustion reaction the ratio of fuel and oxidizer is fixed in such a way that the net reducing valence of the fuel equals to the net oxidizing valence of the oxidizer. Cerium nitrate hexahydrate (Ce (NO₃)₃.6H₂O) salt with appropriate amount of Urea is dissolved in distilled water at fuel to oxidizer ratio $\phi = 1$. The solution further mixed by magnetic stirring is put on the hot plate. As the temperature reached 100⁰C, water started to boil and evaporate from the solution, which increased solution viscosity significantly, during which the mixture trapped with carbonaceous fumes and gases evolved with precipitate remained as a deposit. Then the powder is calcined for two hours at 600⁰C. Calcined sample resulted in the formation of light yellowish white powder of CeO₂ nanoparticles. Further, CeO₂ – Ethylene glycol nanofluids are prepared by the dispersion of CeO₂ nanoparticles of different concentrations in Ethylene glycol solvent using ultrasonicator.

2.2. Characterization

The crystal structures of the calcined samples were characterized by powder X-ray diffraction (XRD) using an (Bruker D8, Advance, Germany) X-ray diffractometer . XRD was performed within the range of $20^0 \leq 2\theta \leq 80^0$ by using Cu K α as radiation (1.5406 Å) in configuration. The particle size and morphology of the calcined powders were characterized by Scanning electron microscopy (SEM). For SEM/EDX analysis, Carls Zeiss (Merlin compact 60-27) field emission scanning electron microscope was used to examine the size, shape and morphology of the annealed nanoparticles. Nano fluids were subjected to ultrasonic studies at room temperature. The velocity values of ultrasonic wave propagation through nano fluid samples were measured using single frequency continuous wave ultrasonic interferometer (Model NF-10X, Mittal Enterprises, India) with an accuracy of ± 0.05 % at frequency of 2 MHz. Nanofluids are suspensions of nano particles in fluids that show significant enhancement of their properties at modest nanoparticle concentrations. Nano fluids are considered to offer important advantages over conventional heat transfer fluids. Nano fluids contain suspended metallic and metal oxide nanoparticles, which increases the thermal conductivity of the base fluid by a substantial amount [7].

3. Results and discussion

3.1. XRD analysis

The XRD patterns of the as-prepared sample and calcined CeO₂ nanoparticles at 600⁰C in the range of $2\theta = 20^0-80^0$ are as shown in Fig. 1. The CeO₂ was characterized by the typical cubic fluorite like crystal structure with predominant XRD peaks at $2\theta = 28, 33, 47, 56, 59, 69, 76$ and 79^0 which corresponded to the (111), (200), (220), (311), (222), (400), (331) and (420) planes respectively. All peaks are in good agreement with the JCPDS file no:75-0076 for CeO₂ which exhibits cubic fluorite phase.

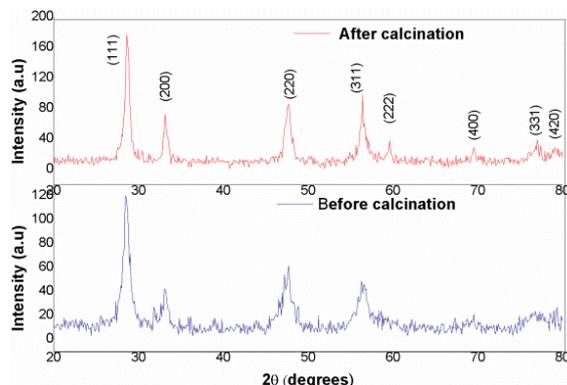


Fig. 1. XRD pattern of the as prepared and calcined CeO₂ nanoparticles

The crystallite size of the CeO₂ nanoparticles was determined by the X-ray line broadening method using the Scherer equation: $D = \frac{k\lambda}{\beta_D \cos\theta}$, where D is the crystallite size in nanometers, λ is the wavelength of the radiation (1.54056 Å for CuK_α radiation), k is a constant equal to 0.94, β_D is the peak width at half-maximum intensity, and θ is the peak position.

Applying the Scherer formula [21] and the full width at half maximum (FWHM) to the all planes, average values of crystallite sizes have been calculated for the as-prepared CeO₂ as 10.48 nm and calcined CeO₂ nanoparticles as 13.33 nm respectively.

The interplanar spacing and length of a unit cell of CeO₂ values are obtained by the following equations $2d\sin\theta = n\lambda$ and $\sqrt{3}d_{111} = a$, as 3.0879 Å⁰ and 5.3483 Å⁰ respectively

XRD theoretical density of CeO₂ nanoparticles is obtained by the following equation $\rho = \frac{nM}{Na^3}$, where M is molar mass, n is number of atoms, N is Avagadro's number and a is length of a unit cell respectively. XRD theoretical density obtained as 7.4716 gm/c.c for CeO₂ nanoparticles respectively [21-23].

3. 2 Scanning Electron Microscopy

A characteristic SEM micrograph of the CeO₂ nanoparticles obtained by solution combustion synthesis method is shown in Fig. 3. The powder consists of particles in the nano-range in both the images at higher magnifications. Particles obtained have agglomeration and porosity due to the excess fuel causing evolution of more amount gases corroborating to the porosity

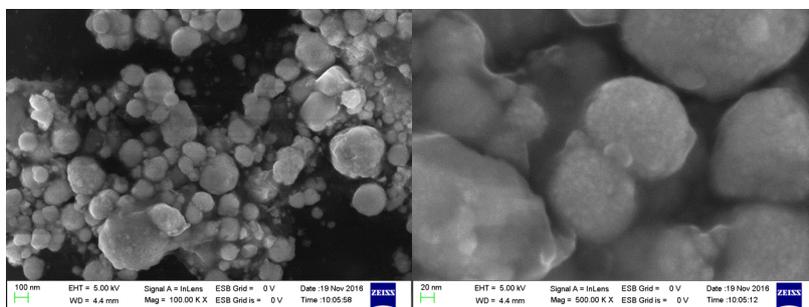


Fig.3. SEM analysis of pure CeO₂ nanoparticles

3. 3 EDX analysis

The EDX analysis of the CeO₂ nanoparticles obtained by solution combustion synthesis method is shown in Fig. 4. The powder consists of particles with porosity have elements Carbon

(C), Oxygen (O) and Cerium (Ce) in the EDX spectrum. Carbon impurity is obtained due to fuel in the preparation of CeO₂ nanoparticles.

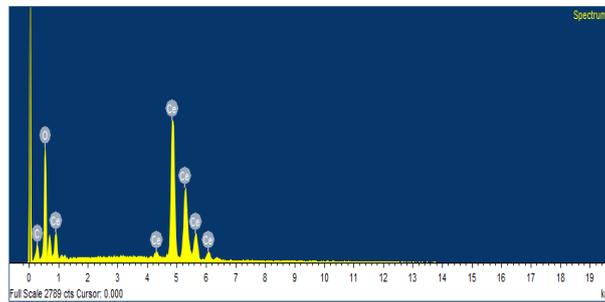


Fig.3. SEM analysis of pure CeO₂ nanoparticles

3.4 Ultrasonic studies

The ultrasonic parameters like adiabatic compressibility (β), acoustic impedance (Z) and thermal conductivity (k), are calculated using ultrasonic velocity of CeO₂-Ethylene glycol nanofluid for different concentrations obtained by ultrasonic interferometer.

The ultrasonic velocity is determined by following expression [24].

$$v = \lambda f \quad (1)$$

where $f = 1.9925$ MHz, λ is wavelength determined from Nanofluid interferometer, v is Ultrasonic velocity.

The adiabatic compressibility of the CeO₂- ethylene glycol nanofluid is determined by the Newton-Laplace's relation [25].

$$\beta = \frac{1}{\rho v^2} \quad (2)$$

Where ρ is density of nanofluid and v ultrasonic velocity.

The acoustic impedance of CeO₂- ethylene glycol is calculated for all concentrations using the relation [26].

$$Z = \rho v \quad (3)$$

Where ρ is density of nanofluid and v ultrasonic velocity.

The thermal conductivity for CeO₂- ethylene glycol nanfluid is determined by using modified version of Bridgman's equation [27,28].

$$k = 2.8 \left(\frac{N}{V} \right)^{2/3} K v \quad (4)$$

where N is avagadro number , V is molar volume of the nanofluid , K is Boltzmann's constant and v ultrasonic velocity.

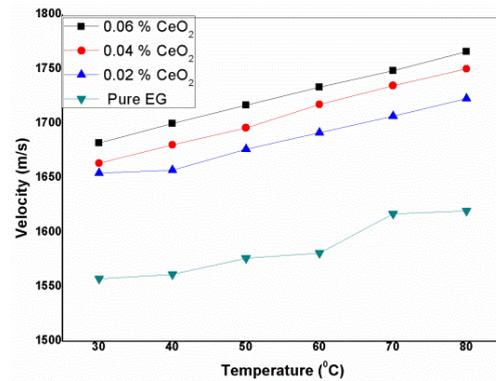


Fig.4. Ultrasonic velocity of base fluid and CeO₂-Ethylene glycol nanofluid at low concentrations(0.02%,0.04% and 0.06%)

Fig.4. indicates ultrasonic velocity of base fluid and CeO₂-Ethylene glycol nanofluid at different concentrations (0.02,0.04 and 0.06). From Fig.4, it is found that ultrasonic velocity increases with increase in concentrations of nanofluids. With the dispersion of ultrasonic vibrations through the nanofluids, the Brownian motion in the fluid resulting in increase in velocity and the random movements of nanoparticles are increased with increase in concentration. Size, ultrasonic velocity is quite sensitive to the size, morphology and dispersion of the particles. Ultrasonic velocity increases with the concentration of nanofluid in respect with particle fraction but at very higher concentrations ultrasonic velocity increases with the concentration of nanofluid due to agglomeration of nanoparticles.

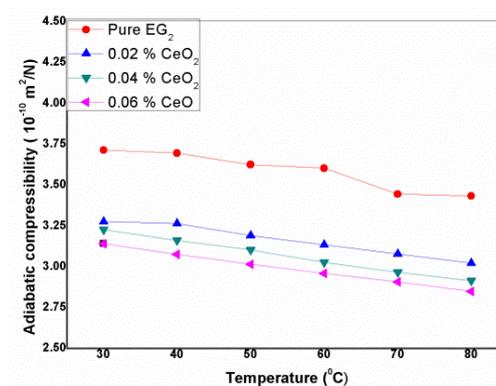


Fig.5. Adiabatic compressibility of base fluid and CeO₂-Ethylene glycol nanofluid at low concentrations(0.02%,0.04% and 0.06%)

Fig.5. indicates adiabatic compressibility of base fluid and CeO₂-Ethylene glycol nanofluid at different concentrations (0, 0.02, 0.04 and 0.06). From Fig.5, it is observed that adiabatic compressibility decreases with increase in concentrations of nanofluids. The decrease in adiabatic compressibility shows the weaker force of interaction between particles and base fluid molecules. Compressibility decreases due to the fact that metal ions form a core compact structure with the solvent molecules through hydrogen bonding. Weak forces operating between molecules results in variation in the values of adiabatic compressibility.

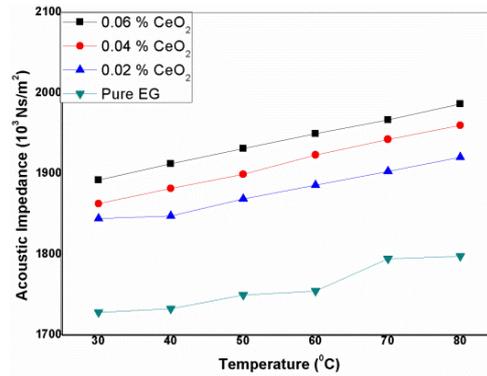


Fig.6. Acoustic impedance of base fluid and CeO₂-Ethylene glycol nanofluid at low concentrations(0.02%,0.04% and 0.06%)

Fig.6. indicates acoustic impedance of base fluid and CeO₂-Ethylene glycol nanofluid at different concentrations (0, 0.02, 0.04 and 0.06). From Fig.6, it is observed that acoustic impedance increases with increase in concentrations of nanofluids.

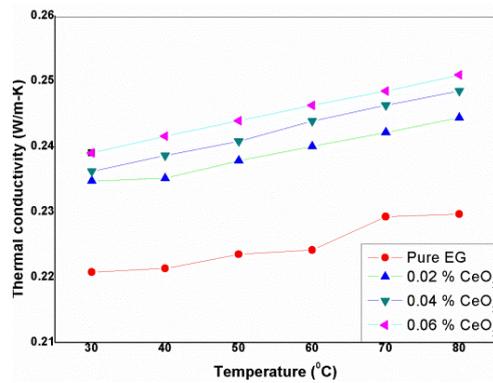


Fig.7. Thermal conductivity of base fluid and CeO₂-Ethylene glycol nanofluid at low concentrations(0.02%,0.04% and 0.06%)

Fig.7. indicates thermal conductivity of base fluid and CeO₂-Ethylene glycol nanofluid at different concentrations (0, 0.02,0.04 and 0.06). From Fig.7, it is observed that thermal conductivity increases with increase in concentrations of nanofluids. Comparing the thermal conductivities of base fluid and CeO₂-Ethylene glycol nanofluid,CeO₂-Ethylene glycol nanofluid has better thermal conductivity. Nanofluids have high thermal conductivities at very low nanoparticles concentrations, the exact mechanism of which is not known. Brownian motion of suspended nanoparticles is attributed as one of the key factors of the greatly enhanced thermal conductivity.

Table1, 2, 3 and 4 indicates the acoustic parameters like ultrasonic velocity, adiabatic compressibility, acoustic impedance and thermal conductivity values for the base fluid and CeO₂-Ethylene Glycol nanofluid at different concentrations.

Table 1. Acoustic parameters of base fluid (Ethylene Glycol)

S.No	Temperature (°C)	Velocity (m/s)	Adiabatic Compressibility ($10^{-10} \text{ m}^2/\text{N}$)	Acoustic Impedance (10^3 Ns/m^3)	Thermal Conductivity (W/m-K)
1	30	1557	3.7155	1728	0.2207
2	40	1561	3.6965	1732	0.2213
3	50	1576	3.6259	1749	0.2234
4	60	1580	3.6044	1754	0.2241
5	70	1616	3.4459	1794	0.2292
6	80	1619	3.4340	1797	0.2296

Table 2. Acoustic parameters of 0.02% CeO₂-Ethylene Glycol nanofluid

S.No	Temperature (°C)	Velocity (m/s)	Adiabatic Compressibility ($10^{-10} \text{ m}^2/\text{N}$)	Acoustic Impedance (10^3 Ns/m^3)	Thermal Conductivity (W/m-K)
1	30	1654	3.2774	1844	0.2347
2	40	1657	3.266	1847	0.2351
3	50	1676	3.1927	1868	0.2378
4	60	1691	3.1347	1885	0.2399
5	70	1706	3.0791	1902	0.2422

Table 3. Acoustic parameters of 0.04% CeO₂-Ethylene Glycol nanofluid

S.No	Temperature (°C)	Velocity (m/s)	Adiabatic Compressibility ($10^{-10} \text{ m}^2/\text{N}$)	Acoustic Impedance (10^3 Ns/m^3)	Thermal Conductivity (W/m-K)
1	30	1663	3.2270	1862	0.2362
2	40	1680	3.1622	1881	0.2386
3	50	1695	3.1045	1899	0.2408
4	60	1717	3.0275	1923	0.2438
5	70	1734	2.9674	1942	0.2463
6	80	1750	2.9153	1960	0.2485

Table 4. Acoustic parameters of 0.06% CeO₂-Ethylene Glycol nanofluid

S.No	Temperature (°C)	Velocity (m/s)	Adiabatic Compressibility ($10^{-10} \text{ m}^2/\text{N}$)	Acoustic Impedance (10^3 Ns/m^3)	Thermal Conductivity (W/m-K)
1	30	1681	3.1420	1892	0.2390
2	40	1699	3.0762	1912	0.2415
3	50	1716	3.0165	1931	0.2439
4	60	1733	2.9595	1949	0.2462
5	70	1748	2.9077	1966	0.2484
6	80	1765	2.8507	1986	0.2509

4. Conclusions

CeO₂ nanoparticles have been effectively synthesized by a simple solution combustion synthesis method. The XRD results indicated that the synthesized CeO₂ nanoparticles had the pure cubic fluorite structure. The crystallite sizes increases with calcination and the crystallite size before and after calcinations areas 10.48 nm and 13.33 nm for CeO₂ nanoparticles respectively. From SEM analysis it is observed that the obtained pure CeO₂ nanoparticles that the grain sizes of CeO₂ are in nanosize and agglomeration of nanoparticles is observed due to combustion of fuel. Ultrasonic interferometer is employed for achieving ultrasonic parameters like ultrasonic velocity (v), adiabatic compressibility (β), acoustic impedance (Z) and thermal conductivity (k). From acoustic parameters it is observed that ultrasonic velocity increases with increase in concentrations of nanofluids, adiabatic compressibility decreases with increase in concentrations of nanofluids, acoustic impedance increases with increase in concentrations of nanofluids and thermal conductivity increases with increase in concentrations of nanofluids. Improvement in thermal conductivity of CeO₂-Ethylene glycol nanofluids is experiential.

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