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Electrical and Optical Properties of Pyrochlore-Type $Y_2Ti_2O_7$ Nanoparticles by Combustion Technique

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ABSTRACT

Pyrochlore-type Yttrium Titanate ($Y_2Ti_2O_7$) ceramics has been successfully synthesized by citric acid based single step auto-igniting combustion technique. The successive phase formation of the prepared combustion powder has been investigated using X-Ray diffraction analysis (XRD) and Fourier Transform Infrared Spectroscopy (FT-IR) analysis. X-Ray diffraction analysis (XRD) showed the formation of $Y_2Ti_2O_7$ nanoparticles with cubic structure in the space group Fd3m. High Resolution Transmission Electron Microscopy (HR-TEM) have been used to determine the particle size and to confirm the nanoparticle size. The average particle size can be estimated from TEM analysis is ~ 42 nm. From the absorption spectrum the optical band gap of the material is determined, which is found to be 4.05 eV. The dielectric properties of the sintered pellet were carried out in the frequency range 1Hz-10MHz at room temperature. The value of dielectric constant ($\mathcal{E}'$) and loss tangent (tan δ) were ~ 36 and 6×10^{-3} at 1MHz,respectively.

Keywords: Yttrium Titanate, combustion technique, ceramics, dielectric constant.

I. INTRODUCTION

Recently, rare-earth titanates with the pyrochlore type structure ($A_2B_2O_7$) has attracted much consideration due to their structural and conducting properties [1]. Rare earth titanates with the general formula $RE_2Ti_2O_7$, the sites of larger A-cations are occupied by rare earth ions and the sites of smaller B-cations are occupied by titanic ions. pyrochlore-structured materials display some interesting properties such as good thermal stability, mixed ionic–electronic conductivity, optical nonlinearity, low phonon cutoff energy, and luminescence [2–4]. Because

of the above-mentioned properties, these materials can be used as ion conductors in fuel cells, gas sensors,electrolytes for solid-oxide fuel cells, dielectric materials [5]. In the Yttrium Titanate ($Y_2Ti_2O_7$) pyrochlore structure, the Y^{3+} occupy the 16c site and are coordinated with 8 oxygen ions, while Ti^{4+} occupy the 16d site and are located at the center of the distorted octahedra of oxygen and the two O^{2-} sites in the unit cell 48f and 8a.Yttrium Titanate($Y_2Ti_2O_7$)compounds, recently has received considerable attention as a possible candidate for application as host materials for efficient Er^{3+} luminescence, hydrogen storage material, oxygen–ion conductor, photocatalyst for water-splitting to produce hydrogen and ceramic pigment [6-7].In this present work $Y_2Ti_2O_7$nanoparticles are synthesized by modified combustion method and the structural, optical and dielectric properties are studied. Aim of this method is shorter duration and low reaction temperature, the prepared nanopowders are extremely homogeneous and high phase pure as compared with the conventional methods.

II. EXPERIMENTAL TECHNIQUES

Stoichiometric amounts of Yttrium oxide Y_2O_3 (99.99% Purity, Sigma Aldrich) and Titanium dioxide TiO_2 (99.99% Purity, Sigma Aldrich) were used as the starting materials for the auto-ignition combustion synthesis. $Y_2O_3$and TiO_2 were dissolved in nitric acid and the two solutions are mixed with de-ionized water to form an aqueous solution. Appropriate amount of citric acid was added into the solution containing the metal ions, keeping the citric acid to cation ratio unity. The mixed solution was stirred well at room temperature, and then liquor ammonia was added to the solution to adjust the oxidant/fuel ratio unity. The system containing the complex precursor mixture at neutral pH, was heated on a hot plate to about 250^0 C. The solution boiled on heating and undergoes dehydration followed by decomposition leading to a smooth deflation producing a form. Then, the form ignites by itself on persistent heating giving fluffy and voluminous combustion product.

The as prepared powders were characterized by powder X-Ray diffraction analysis using X-ray powder diffractometer (Bruker AXS D8 Advance) with CuKα radiation (1.5406 Å). FT-IR spectrum

of the sample has been recorded with a Thermo-Nicolet Avatar 370, in the range 400–4000 cm^{-1}. The high resolution TEM images were taken on a Transmission Electron Microscope, Joel-JEM 2100. The optical measurements of $Y_2Ti_2O_7$ nanopowder were measured using UV-Vis (Varian, Cary 5000) spectrophotometer in the range 200-800 nm.The powders calcinated at 700^0C have been mixed with PVA and pressed to form cylindrical compacts at a pressure about 350 MPa using a hydraulic press, which then sintered at 1400^0C for 4 hrs. The dielectric properties were performed with Network analyzer in the frequency range 1Hz to 10MHz.

III. RESULTS AND DISCUSSION

3.1. X-RAY Diffraction Analysis

The X-Ray diffraction pattern of the as prepared combustion powder is calcinated at $700^0$is shown in Fig.1. All of the diffraction peaks were indexed for a phase pure cubic structure of space group Fd3m. Lattice parameter of $Y_2Ti_2O_7$ nanopowder was calculated from the XRD pattern is a = 10.053 Å, which agrees very well with the standard JCPDS card no.42-0413. The average crystallite size estimated from the XRD peaks using the Debye-scherrer formula was ~31 nm. The calcination temperature (700^0) is low as compared with the solid-state reaction process [8]. No other peaks related to impurities are detected in the XRD pattern, which confirms the phase pure $Y_2Ti_2O_7$ pyrochlore oxide.

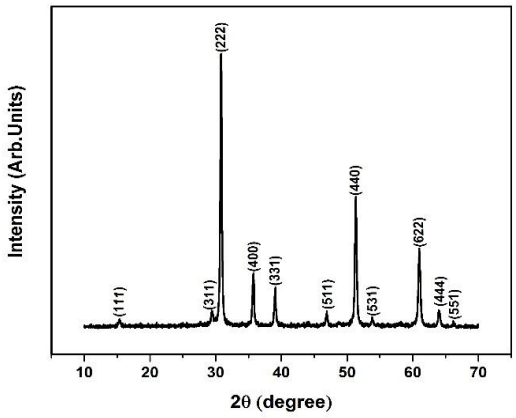


Fig.1 XRD pattern of combustion

3.2.FT-IR ANALYSIS

FT-IR spectrum demonstrate the formation of pyrochlore phase. Fig.2 shows the FT-IR spectra of the synthesized combustion powder. Broad absorption band at 3729.95 cm^{-1}can be attributed to the stretching vibration of hydroxyl group (O-H), representing the water observed from moisture. The sharp peak appearing at 471.94 cm^{-1} attributed to Y-O stretching vibrations. The strong band at 573.08 cm^{-1} assigned to the Ti-O-Ti stretching vibrations. The presence of metal oxygen bond in the FT-IR spectra further confirmed the formation of crystalline pyrochlore phase. The band at 1381·19 is ascribed tothe COO– stretching vibration for Ti^{4+}– carboxylic acid complex suggesting that the re-existed strong co-ordination interaction between Ti^{4+} and citric acid [9].

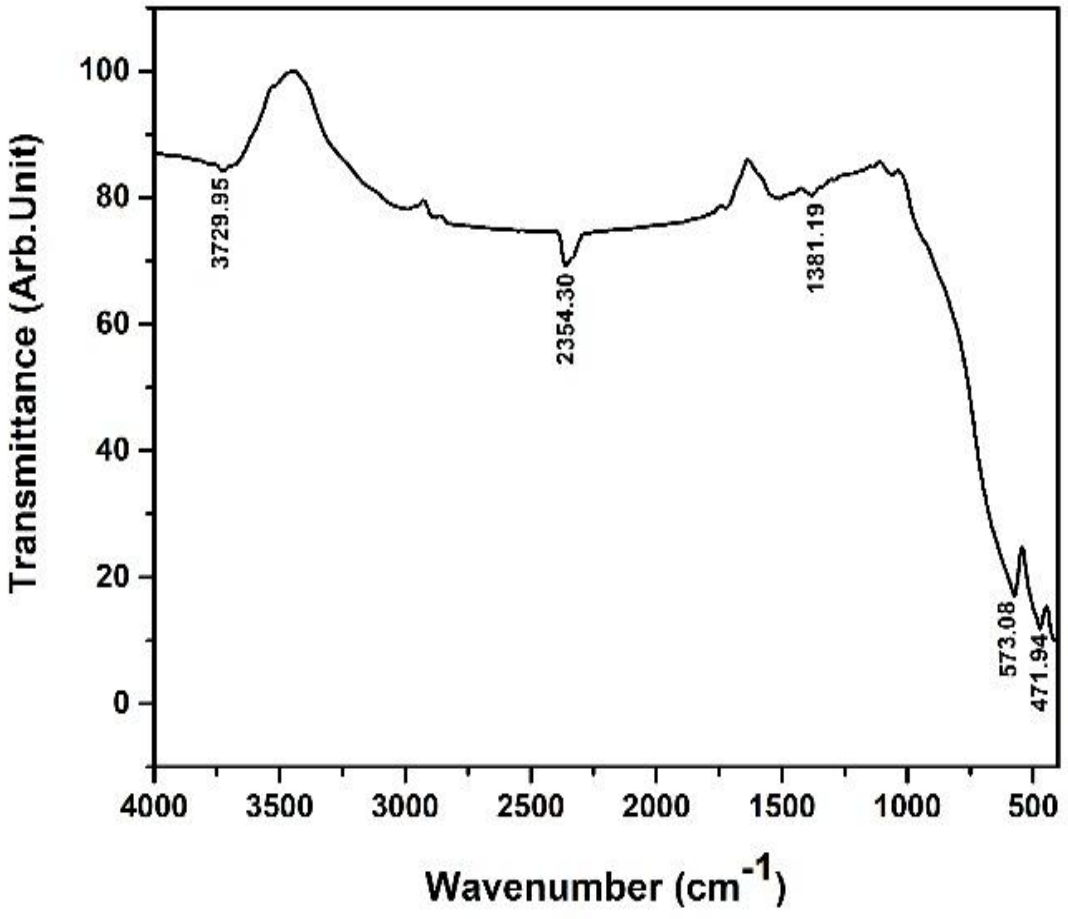


Fig. 2 FT-IR spectra of $Y_2Ti_2O_7$ sample

3.3. Tem Analysis

The microstructure and average particle size of the sample were directly attained from the Transmission Electron Microscopy (TEM).The

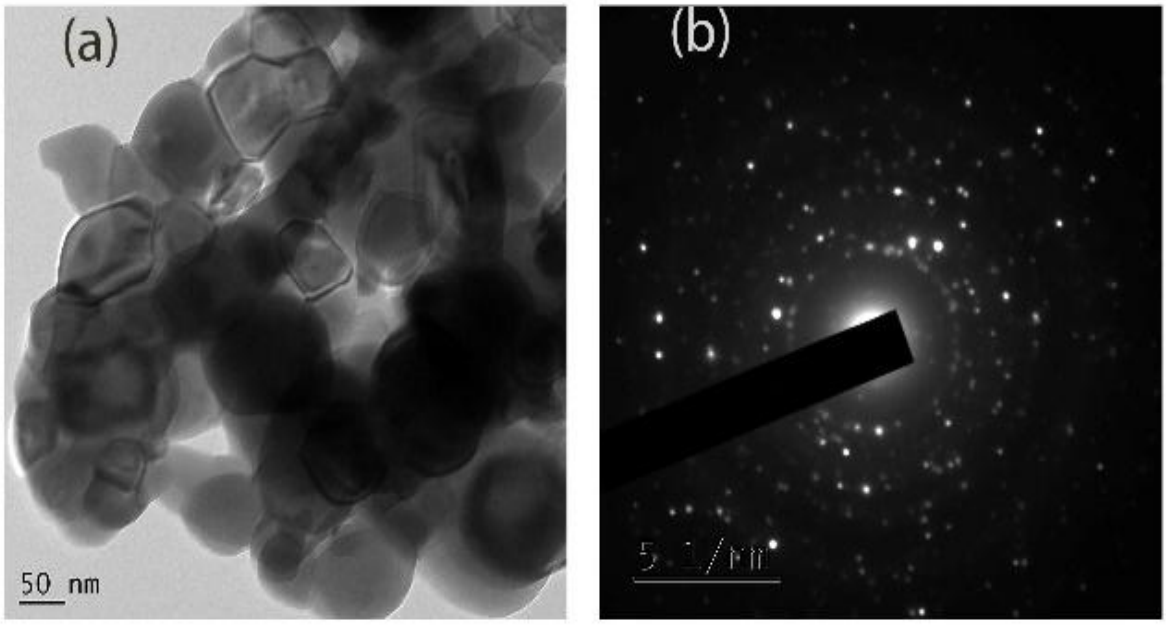


Fig. 3(a) TEM image and (b) SAED pattern of $Y_2Ti_2O_7$nanopowder

High Resolution TEM images for the prepared $Y_2Ti_2O_7$ nanocrystals is shown in Fig.3(a), it can be found that quasi-spherical $Y_2Ti_2O_7$nanoparticles have good dispersibility, and the average particle size was evaluated from the TEM micrograph is 48 nm. Due to the slight agglomeration of crystallites, the particle size gained from XRD analysis and TEM analysis are different. Selected Area Electron Diffraction (SAED) pattern is shown in Fig.3(b). The ring nature of the SAED pattern is an indicator of the polycrystalline nature of the crystallites.

3.4. UV-VIS Spectra Analysis

Fig.4(a) shows the ultraviolet- visible reflection spectrum of the $Y_2Ti_2O_7$ nanopowder. The absorption edge was obtained from the spectrograph, which is found to take place at ~ 293 nm. The band

gap energy was calculated using the tauc relation, $\alpha(h\upsilon) \approx B(h\upsilon - E_g)^{1/n}$, where B is the absorption constant, $h\upsilon$ is the photon energy, E_g is the band gap energy, $\alpha$is the absorption coefficient, which is determined from the Kubelka- Munk theory. The exponent n depends on the type of transition. Tauc plot with n=1/2 for a direct allowed transition (plotted as $(\alpha h\upsilon)^2$ versus $h\upsilon$) of $Y_2Ti_2O_7$ nanoparticles is shown in the Fig.4(b). The value of band gap energy (E_g) was obtained by extrapolating a straight line to $h\upsilon$ axis, which is found to be 4.05 eV.

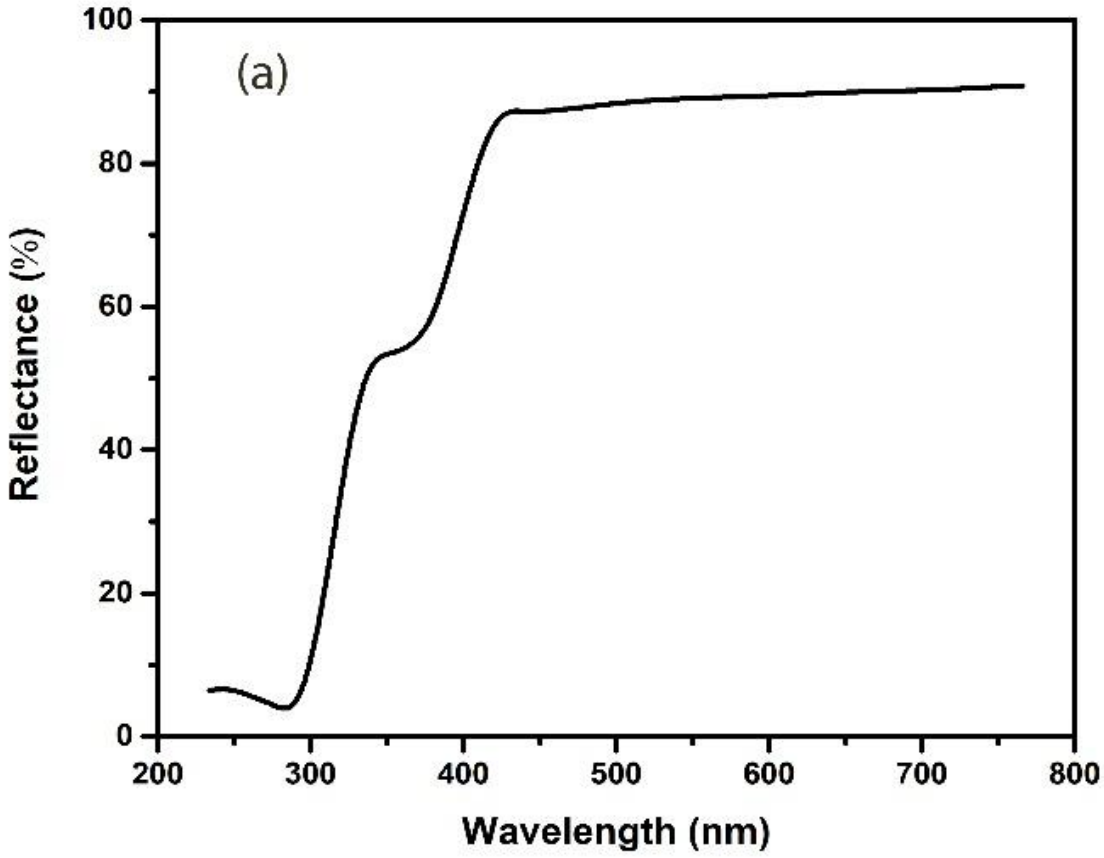
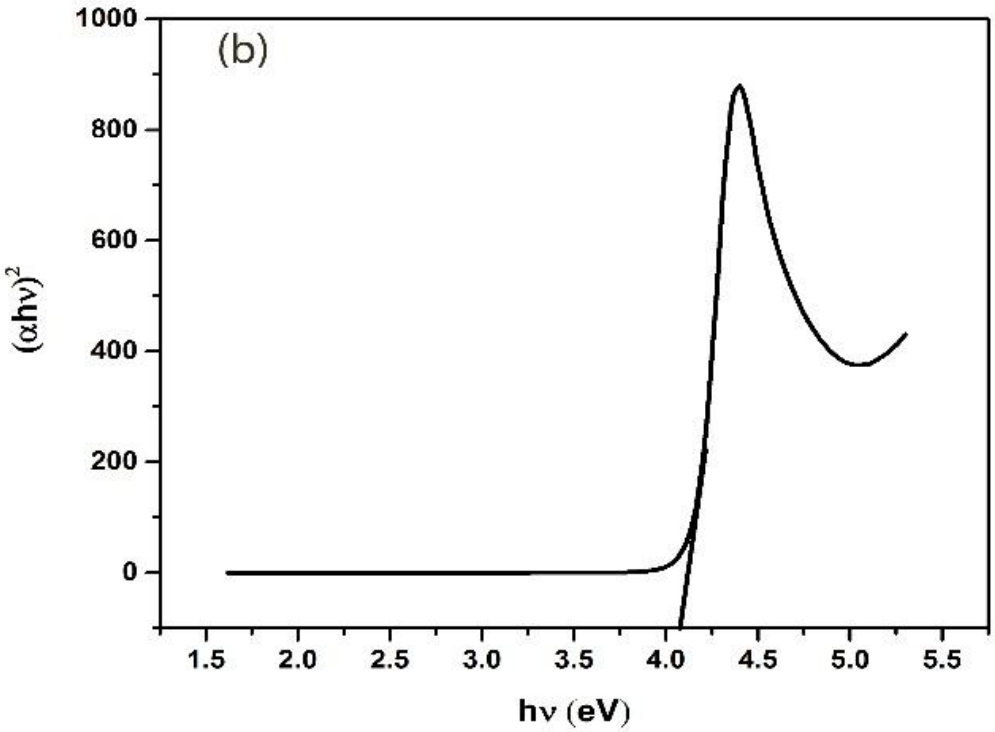


Fig. 4 (a) UV-Visible reflection spectrum and



(b)optical band gap of $Y_2Ti_2O_7$

3.5. DIELECTRIC Properties

The dielectric characteristics of $Y_2Ti_2O_7$ sintered pellets as a function of frequency is shown in Fig.5. The variation in the real part of dielectric permittivity (Ɛ') and loss tangent (tan δ) were measured in the frequency range 1Hz-10MHz at room temperature. The dielectric constant (Ɛ') and loss tangent (tan δ) were found to be ~36 and ~6×10^{-3} respectively, at a frequency of 1MHz.

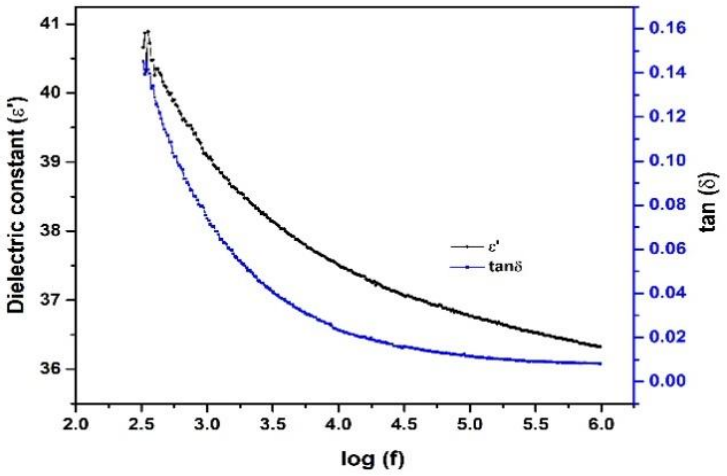


Fig. 5 Variation of dielectric constant (Ɛ') and loss tangent (tan δ) for $Y_2Ti_2O_7$with frequency

Dielectric constant and loss tangent were decrease with increase in frequency and attain a constant value at higher frequency corresponds to the interfacial polarization and ion polarization, respectively. Dielectric loss is the electrical energy lost as heat in the polarization process in the presence of an applied ac field. The total polarization of the dielectric material can be agreed as the sum of the four types of polarization such as electronic, ionic, orientationaland interfacial polarization [10].

VI. CONCLUSIONS

$Y_2Ti_2O_7$ nanoparticles were successfully synthesized by an auto-ignition combustion process. The successive phase formation of $Y_2Ti_2O_7$ nanopowder has been confirmed by the XRD and FT-IR analysis. The high resolution TEM micrograph shows the microstructure and the particle size, which is in the range 30-50 nm. The ring nature of the diffraction (SAED) pattern is an indicative of polycrystalline nature of the crystallites. The band gap energy of the material is determined from the UV-Vis spectra was found to be 4.05 eV. The dielectric properties have been studied as the function of frequency, the dielectric constant and dielectric loss decrease with increase in frequency.

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