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RESEARCH ARTICLE

INVESTIGATION OF THE ABSORPTION COEFFICIENT, REFRACTIVE INDEX, ENERGY BAND GAP, DISPERSION ENERGY AND URBACH ENERGY OF NANOCRYSTALLINE CERAMIC SUPERCONDUCTOR YSRBICUO

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ABSTRACT

In this era of High temperature superconductors (HTS), having a lot of applications like high –voltage generator, Ceramic superconductor yttrium strontium bismuth copper oxide (YSBCO) was synthesized thermo chemically by solid state method at different treating temperatures. A characteristic feature of all solid-state reactions is that they involve the formation of product phase(s) at the interfaces of the reactants. UV-VIS analysis of the sample was carried out. Tunable band gaps can be obtained by varying annealing temperatures. The optical constants of refractive index, normal-incidence reflectivity, extinction coefficient, and absorption coefficient showed systematic variation with temperature. The dispersion of refractive index was analyzed by the Wemple- Di Domenico single-oscillator model. Urbach energy was elucidated from the raw data.

INTRODUCTION

Optical experiments provide a good way of examining the properties, particulary measuring the absorption coefficient for various energies gives information about the band gaps of the material. Knowledge of these band gaps is extremely important for understanding the electrical properties and is therefore of great practical interest. Georg Bednorz *et al*,(1986) were exploring a new class of ceramics for superconductivity. Bednorz encountered a barium-doped compound of lanthanum and copper oxide whose resistance dropped down to zero at a temperature around 35 K (−238.2 °C). Their results were soon confirmed by many groups (Tanaka *et al*, 2012). Shortly after, Scientists (Anderson, 1887) came up with the first theoretical description of these materials, using the resonating valence bond theory, but a full understanding of these materials is still developing today. These superconductors are now known to possess a d-wave (Kotliar *et al*,1988) pair symmetry.

The optical properties of the superconductors have attracted a great deal of attention due to its potential applications in high speed optoelectronic devices. Especially, incorporating the photonic crystals concept into superconductor electronics has opened novel approaches for tuning the photonic crystal (PC) properties such as transmission wavelength (Raymond *et tal*,2000;Berman *et al*, 2006; Yan-bin Chen *et al*,2003; Wu,200 Kokabi, 2005;5; Takeda, 2003; Lyubchanskii *et al*, 2009; Rauh *et al*, 2008; Pei *et al*, 2007) [1-9] . In this regard, (Chen *et al*.,2003) have investigated the control of photonic band gap (PBG) of the superconductor-dielectric lattice by variation of superconductor operation temperature. The inertia of temperature variation is not favourable for the applications such as optical switches that require rapid permittivity change.

Optical gap is different from the quasi-particle gap (at least in the conventional BCS theory for bulk materials, the superconducting gap coincides with the quasi-particle gap arising from paring). In optical experiments (and Tauc plot is used for determining optical gap) one excites particles that remain to interact with the hole they leave behind; in contrast, in photo-emission (and inverse photo-emission) experiments, one truly removes (adds) particles from (to) systems. The optical gap is in general smaller than the quasi-particle gap, the difference being the binding energy of the bound states that electrons can form with holes (forming excitons). This binding energy can be small, whereby optical gap can be very close to the quasi-particle gap, but this is not in general the case. Incidentally, because bound states cannot be calculated by means of finite-order perturbation theory, calculation of the optical gap in general requires solving the relatively complicated Bethe-Salpeter equations.

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Due to chemical and physical properties arising from the large surface–volume ratios and also the quantum size effect, compared with those of bulk counterparts (Zhi Meng *et al*, 2013; Chang *et al*, 2007; Alivisatos, 1996; Hu *et al*, 1999; Brus *et al*, 1995; Heath, 1999) increasing attention has been paid to the synthesis and characterization of nanomaterials.

In this work the authors describes the optical behaviour of $PbSrBaTiO_6$, a nanocrystalline superconductor material. The energy band gap values of the sample were analyzed for different temperatures and they are fundamentally important to the design of practical devices (Nepal *et al*.). Measuring the band gap is an important factor determining the electrical conductivity in nano material industries. In solid state physics a band gap, is an energy range in an ideal solid where no electron states can exist. This is equivalent to the energy required to free an outer shell electron from its orbit about the nucleus to become a mobile charge carrier, able to move freely within the solid material (Reenu *et al*, 2014). The band gap energy of insulators is large (> 4eV), but lower for semiconductors (< 3eV).

Tauc plot derived from the band gap energy values, showed a direct relation with temperature. The Urbach energy of the sample was also studied. The optical constants of refractive index, extinction coefficient, and absorption coefficient showed a systematic variation with temperature. The dispersion of refractive index was analyzed by the Wemple-DiDomenico single-oscillator model and such optical behaviour is rarely reported. Tunable band gap can be possible to fabricate.

MATERIALS AND METHODS

The new nanocrystalline ceramic superconductor sample Yttrium strontium bismuth copper oxide (YSrBiCuO/YSBCO) was prepared by the solid state thermo chemical reaction technique using a high-energy ball milling process through mechanically assisted synthesis. High purity Yttrium oxide, strontium carbonate, bismuth oxide, cupric oxide powders were used as the raw materials and weighed according to their molecular formula. Mechanical mixing, ball milling and attrition milling were utilized to insure homogeneity. Then the material calcined at different treating temperatures, 30^0C, 500^0C, 800^0C and 950^0C. Control of temperature is often necessary to ensure that the desired crystalline phase is formed with optimum particle size (Vinila *et al*, 2014). Then UV-Vis spectrum of these materials was taken. The optical constants of refractive index, extinction coefficient, normal-incidence reflectivity, and absorption coefficient were calculated, showed systematic variation with temperature. The dispersion of refractive index was analyzed by the Wemple- Di Domenico single-oscillator model.

UV-VIS. Analysis

The UV-VIS optical absorption spectrum of the sample were taken and studied at room temperature. The sample obtained after calcination at different temperatures was subjected to UV-VIS-Near IR analysis (Fig.1) using Varian, Cary 5000 Spectrophotometer over a spectral range of 175-3300nm with an accuracy of ±0.1nm (UV-Vis.). This type of sample has high mechanical hardness, high thermal conductivity, large dielectric constant, and high resistance to harsh environment. UV-Visible spectrum give information about the excitonic and inter transition of nano materials (Choudhury *et al*, 2013).The optical absorption data were analyzed using the classical relation for near edge optical absorption of semiconductors (Dhannia *et al*,2009; Varghese *et al*, 2002).

The UV analysis can be thought as a good quality check for the optical behaviour of the ceramic materials. Figure.1 shows the UV-VIS behaviour of the sample YSrBiCuO at 950^0C.

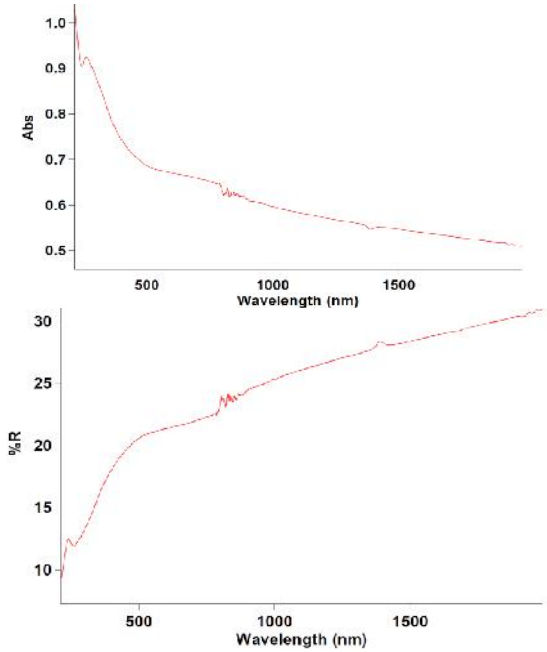

Figure1 UV-VIS spectrum of YSrBiCuO (a) Absorbance (b) reflectance

The diffuse reflectance spectra were translated into the absorption spectra by the Kubelka-Munk method. Kubelka-Munk's equation is described as follows:

$\alpha = (1-R)2/2R$ -(1), where α is the absorption coefficient and R the reflectivity at a particular wavelength(Keigo *et al*, 2005). According to the Tauc relation, the absorption coefficient α for a material is given by $\alpha = A(h\nu - Eg)^n$-- (2), Where Eg the band gap, constant A is different for different transitions, (hv) is energy of photon in eV and n denotes the nature of the sample transition (Tauc *et al*, 1972). The 'n' in the equation has values 1/2, 2, 3/2 and 3 for allowed direct, allowed indirect, forbidden direct and forbidden indirect transitions (Khan *et al*, 2010; Jeon *et al*, 2011; Kumar *et al*, 2011) respectively. The band gap energy can be determined using the Tauc relation. The TAUC plot of a sample defines the optical band gap as the region A in fig.2. The tauc plot of the sample is given in Fig 6. It is reported that optical gap energy of nano –sized crystal depends on its crystallite size, it increases with decreasing crystallite size in the nano size range (Lu *et al*, 1996; Golego *et al*, 1998).

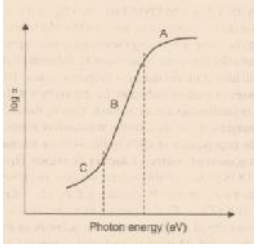

Fig.2optical band gap energy variation with absorption

The absorption coefficient at the photon energy below the optical gap (tail absorption) depends exponentially on the photon energy: $\alpha(\hbar v) \sim exp\ (\hbar v/E_u)$ --(4) where E_u is called Urbach energy.The region B in the fig.2 represents the Urbach energy. The absorption edge called the Urbach energy, depends on temperature, thermal vibrations in the lattice, induced disorder, static disorder, strong ionic bonds and on average photon energies (Sumi *et al*, 1971). The edge arises due to a radiative recombination between trapped electrons and trapped holes in tail and gap states as shown in Fig.2, and is dependent on the degree of structural and thermal disorder (Dennis *et al*, 2014).

Optical Properties

Different type of transitions are discussed as follows:
Direct *(perpendicular) transitions: dipole-allowed interband transitions. k = ki = kf*

Indirect transitions: "inclined" transitions within the 1st Brillouin zone: *the k-conservation can not be realized by a reciprocal lattice vector . Phonon supplies the missing momentum to the electron.*
Ef - Ei = hv + Eph (phonon absorption)
Ef - Ei = hv - Eph (phonon emission)
kf = ki + kph

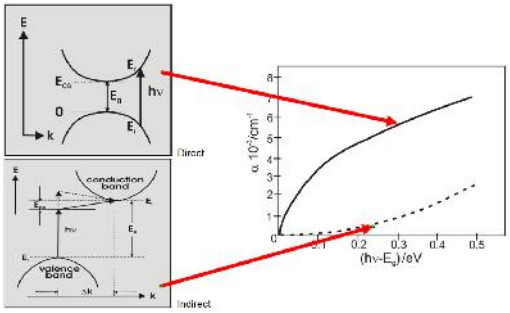

Fig.3 direct, indirect optical transition

By plotting hv against α (phonon absorption) of the sample prepared, YSBCO (figure 4), the following graph is obtained which implies that it belongs to indirect transition, so that the value of n can be taken as 2 for further calculations.

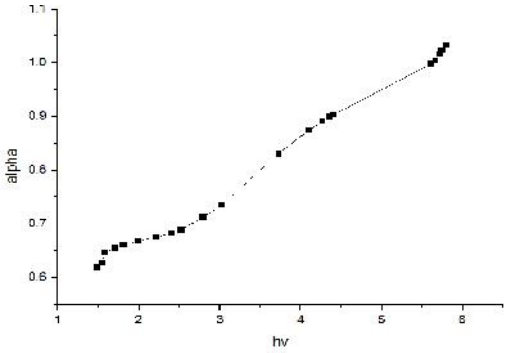

Fig.4. hv vs. α plot of YSrBiCuO

Penetration depth (x)–the inverse of the absorption coefficient (α^{-1}) – average distance at which traveled by a photon before it gets absorbed.

The light intensity vs. distance for a few typical examples of absorption behavior is shown in fig. 5.

The absorption coefficient depends on the material and also on the wavelength of light which is being absorbed. Knowing the absorption coefficients of materials aids engineers in determining which material to use in their designs.
The absorption coefficient, α, is related to the extinction coefficient, k, by the following formula:

$$\alpha = \frac{4\pi k}{\lambda}$$

where λ is the wavlength. If λ is in nm, multiply by 107 to get the absorption coefficient in the the units of cm^{-1}.

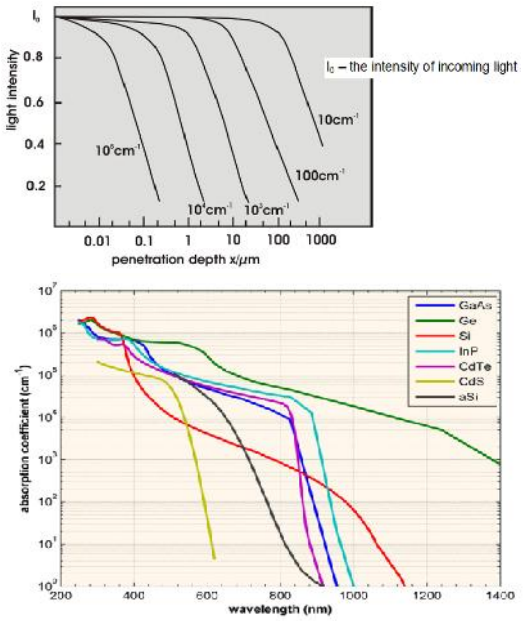

Fig.5 penetration depth plot related to absorption behaviour.

It is observed in manycases that optical absorption by defects also appears at energy lower than optical gap (region C of fig.2). This region is related to the structural properties of materials (Kugler, 2013).

The extinction coefficient and the absorption coefficient are related as $\alpha(E) = 4\pi/\lambda\ (k(E))$—(3).

Variation of band gap energy with samples annealed at different temperatures is shown in figure 7.The natural logarithm of the absorption coefficient, α(v), was plotted as a function of the photon energy, hv (Fig.8). The value of Eu was calculated by taking the reciprocal of the slopes of the linear portion in the lower photon energy region of curves. The measurement of temperature-dependent Urbach tails distinguishes a temperature-dependent tail and a temperature-independent part, which mainly are due to intrinsic defects. The temperature-dependent part of the Urbach tail, is purely of intrinsic reasons (Letz *et al*, 2010). The latter can be controlled by improving the crystal growth and the purity of the ingredients.

Refractive Index and Dispersion

From the data of YSBCO, Variation of refractive index with wavelength was also studied. The refractive index values show a linear decrease with the increase in wavelength, Fig.9 shows the variation of the dispersion curve with annealing temperatures. Refractive index value shows a slight increase with increasing annealing temperature and attains a fixed value after a particular wavelength. The refractive index values showed a linear decrease with the increase in wavelength when

plotted with refractive index along the Y-axis & wavelength along the X axis(figure 9).

The dispersion of refractive index below the interband absorption edge is analyzed using the Wemple-DiDomenico(W-D) model [(Wemple *et al*, 1971). In the W-D model, the refractive index n can be written as n2 -1= Ed E0/ (Ed2-E2) - - (5),

where E is the photon energy, Eo is the oscillator energy, and Ed is the dispersion energy. Wemple and DiDomenico reported that the dispersion energy may depend upon the charge distribution within each unit cell, and that it would be closely related to chemical bonding (Wemple *et al*, 1971). The oscillator energy Eo and dispersion energy Ed are obtained from the slope (EoEd)-1 and intercept Eo/Ed on the vertical axis of the straight line portion of (n2-1)-1 versus E 2plot. The static refractive index n(0) at zero photon energy is evaluated from Equation (5), i.e. n2(0)=1+Ed/Eo—(6) (Wug-Dong Park).

RESULTS AND DISCUSSION

The optical analysis of the ceramic material prepared by solid state reaction technique and treated at different temperatures is successfully done using UV-Vis Spectro photometer. Here the indirect allowed transitions are considered. UV-VIS analysis, clearly confirms that band gap energy of the nano ceramic superconductor YSrBiCuO increases as the annealing temperature of the sample is increased. The calculated values of the band gap energy of the sample at different values of temperature is given in table -1.

The Tauc plot is plotted with hv along the X-axis and (hvα)2 along the Y-axis. The band gap at a particular temperature is found by extrapolating the X axis. The Tauc plot of the sample at temperatures 950◦C is given in Fig.6. The band gap energy values of YSrBiCuO at different temperatures calculated are listed in the table.1.

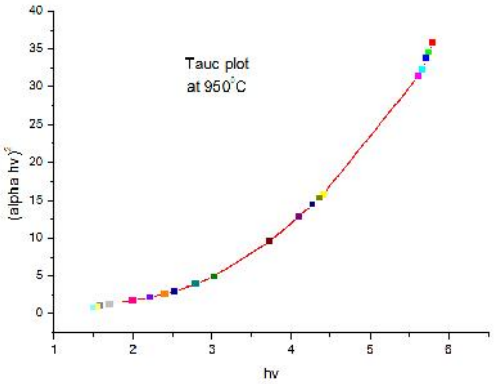

Fig.6 The Tauc plot of YSrBiCuO

Table1 Band gap energy values of YSrBiCuO at different temperatures

Temperature	Band gap energy in eV
30◦C	4.83
500◦C	5.16
800◦C	5.41
950◦C	5.79

The energy levels are dependent on the degree of structural order–disorder in the lattice. It is observed that band gap energy rises with increase in annealing temperature of the sample(fig.7). The band gap increases with the crystallite size but decreases as the perosvkite phase is formed which proves the quantum confinement also decreasing its dislocation density. The decrease in band gap energy shows that the sample tends to be more conducting as the temperature is increased.

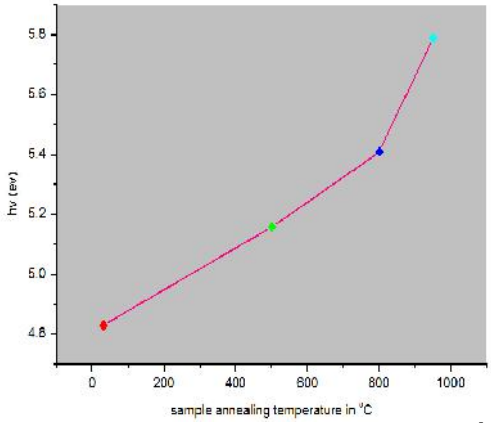

Fig.7 Band gap energy variation with the samples annealed at 30^{0}C, 500^{0}C, 800^{0}C & 950^{0}C *of* YSrBiCuO

Tauc plot data well confirms that the band gap energy of the sample increases slightly when the temperature is increased. As the temperature is increased the crystallite size also increases which shows an increase in band gap energy (Reenu, 2014). The energy levels are dependent on the degree of structural order–disorder in the lattice. Therefore, the increase of structural organization in nano ceramic leads to a reduction of the intermediatary energy levels and consequently increases the Eg values.

Urbach energy is calculated by plotting the natural logarithm of the absorption coefficient with the energy in eV (Figure 8). This value is found to be lower than the band gap energy and hence Sumi-Toyozawa (ST) model theory can be well applied to this material.

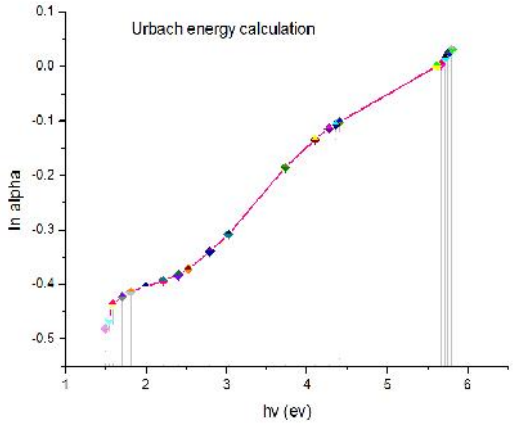

Fig.8 Absorption variation with photon energy of YSrBiCuO

Data of the sample at different annealing temperatures was used for the calculation of refractive index of the sample. Analysis clearly shows that refractive index of the sample decreases as the wavelength increases and attains a definite value at all temperatures. This linear variation of the refractive index with the wavelength is due to dispersion of light energy at the different interstitial layers. The refractive index also shows a linear relation with the photon energy (fig.9). The increase in refractive index is due to crystallization of the perovskite phase. The refractive index of perovskites is known to be proportional to their electronic polarization per unit volume which is inversely proportional to distance between

atomic planes. This result can also be explained by an increase in crystallite size.

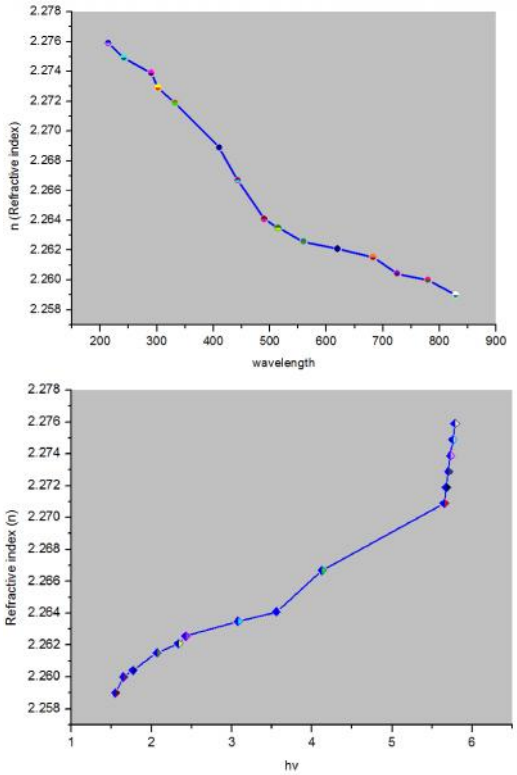

Fig.9 Variation of Refractive index (n) with wavelength and photon energy (YSrBiCuO).

Refractive index of the sample annealed at different temperatures can be calculated using Sellmeir dispersion formula (DiDomenico *et al*,1969,1971).

The dispersion energy of the sample is calculated using the Wemple-DiDomenico (WD) model. Results are plotted graphically in (Fig.10).

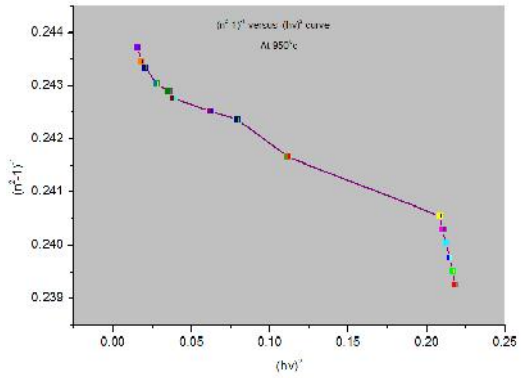

Fig.10 $(n^2-1)^{-1}$ versus$(\ hv)^2$ curve.(YSrBiCuO)

The data of the dispersion of the refractive index (n) were evaluated according to the single oscillator model proposed by wimple and DiDomenico as, $n^2 = 1 + (E_dE_0)/(E_0^2-hv^2)$ ---- (7). where E_o is the oscillator energy and E_d is the oscillator strength or dispersion energy. Plotting of $(n^2-1)^{-1}$ against $(\ h\ v\ )^2$ allows to determine, the oscillator parameters, by fitting a linear function to the smaller energy data, E_o and E_d can be determined from the intercept, (E_o/E_d) and the slope $(1/E_oE_d)$. E_o is considered as an average energy gap to, it varies in proportion to the Tauc gap $E_o \sim 2E_g$.

The oscillator model can be also written as $n^2-1=S_0\ \lambda_0^2\ /[1-(\ \lambda o\ /\ \lambda)^2\ ]$ --- (8) where λ is the wavelength of the incident radiation , S_o is the average oscillator strength and λ_o is an average oscillator wavelength.

The curves of $(n^2 - 1)^{-1}$ against $(1/\lambda^2)$ (Fig.11) are fitted into straight lines following the sell Meier's dispersion formula. The value of S_o and (λ_o) are estimated from the slope (1/So) and the infinite wavelength intercept $(1/S_0\ \lambda_o)^{\ 2}$. The optical parameters of the sample were calculated.

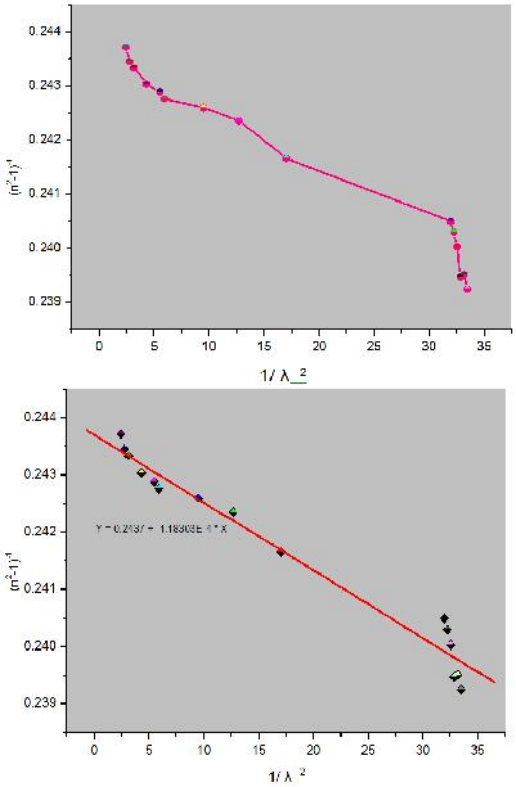

Fig.11 $(n^2-1)^{-1}$ versus $1/\ \lambda^{\ 2}$ curve (curve and linear fit)

The curves with straight line graphs confirms the sell Meier's dispersion formula. It is clear that as the temperature is increased band gap energy decreases or E_0 decreases respectively. The dispersion energy also shows a decline as the temperature rises and the sample attains its perosvkite phase. Further the mechano chemical process has an advantage due to low-costs and widely available materials, leading to a simplified process.

CONCLUSION

UV-VIS. data was analyzed to deduce the optical properties of the nano ceramic material YSrBiCuO which can be applied to UV_VIS shielding applications. The UV emission peak shifts significantly to higher wavelengths with increasing annealing temperatures. The increase in the band gap energy increases the dielectric properties of the material. It is confirmed that tunable band gaps are obtained by varying annealing temperatures. Optical measurements confirmed that absorbance and reflectance increases with temperature. According to Wemple- DiDomenico single-oscillator model the dispersion energy decreases as the sample attains its perosvkite phase. For new generation capacitors nano crystalline ceramics YSrBiCuO materials will prove as a future substitute.

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