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Optical Analysis, Urbach and Bandgap Energy of $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15-Manganese - Zinc Ferrite system doped with Titanium

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Abstract: ***The authors prepared a crystalline ceramic material $Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15 by conventional solid state reaction technique which involves selection of raw materials, mixing, milling and calcination at high temperature. Characterization studies are done using U-V analysis and Wemple- Di Domenico single-oscillator model. Absorption spectra and reflectance spectra of the sample are analyzed. Optical constants such as refractive index, extinction coefficient, normal-incidence reflectivity, and absorption coefficient are calculated and their dependence on temperature is studied. Band gap energy is determined using Tauc method. Urbach energy and Dispersion of refractive index is also analyzed.***

Keywords: **$Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15**, U-V analysis, Wemple- DiDomenico single-oscillator model, Absorption spectra, Reflectance spectra, Optical constants, Band gap energy, Tauc method.

1. Introduction

Ceramics are a group of solids that can be defined as nonmetallic, inorganic, polycrystalline, brittle, multi-elemental, most of them are insulators of current and heat. They acquire strength by firing or sintering. They have a wide range of applications. Ceramics are made up of very minute crystallites. These crystallites have different structure and composition.

Recent discoveries revealed enormous potential of ceramics based on unique dielectric, ferroelectric, ferromagnetic, magnetoresistive and optical properties. The last few decades have seen the development of ceramic at high temperature. This has changed an interesting curiosity to a useable technology, with particular applications in the medical field. Here, the authors designed a unique crystalline ceramic Manganese-Zinc Ferrite system doped with Titanium - **$Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15.** Conventional solid state reaction technique which involves selection of raw materials, mixing, milling and calcinations at high temperature is used in its preparation. Characterization studies are done using U-V analysis and Wemple- Di Domenico single-oscillator model. Absorption spectra and reflectance spectra of the sample are analyzed. Different types of optical constants such as refractive index, extinction coefficient, normal-incidence reflectivity, and absorption coefficient are calculated and their dependence on temperature is studied. Band gap energy is determined using Tauc method. Dispersion of refractive index is also analyzed.

The term ―band gap" refers to the energy difference between the top of the valence band to the bottom of the conduction band; electrons are able to jump from one band to another. In order for an electron to jump from a valence band to a conduction band, it requires a specific minimum amount of energy for the transition, the band gap energy [1]. Insulators have large band gap energy (>4eV), but semiconductors have lower values (<3eV). The measurement of band gap energies is very important in semiconductor, micro/nano material and solar industries. Band gap energy of any material can be directly measured from the UV absorption spectrum obtained after UV analysis. Tauc relation is used to obtain band gap energy values from UV absorption spectrum. It is found that band gap energy values are directly related with temperature. Another important aspect is the calculation of Urbach energy. The values of optical constants of refractive index, extinction coefficient, and absorption coefficient changes with temperature. The relation of refractive index with wavelength is analyzed by Wemple-DiDomenico single-oscillator model.

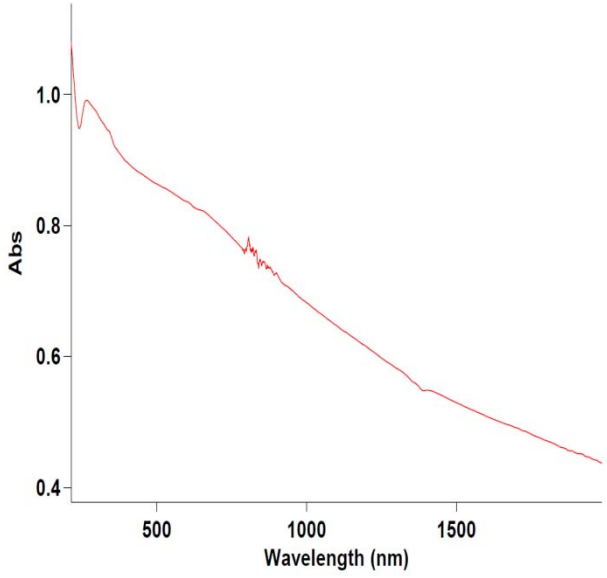
2. Experimental

The solid-state reaction route is the most widely used method for the preparation of polycrystalline solids from a mixture of solid starting materials. Solids do not react together at room temperature over normal time scales and it is necessary to heat them to much higher temperatures, in order for the reaction to occur at an appreciable rate. The factors on which the feasibility and rate of a solid state reaction depend include, reaction conditions, structural properties of the reactants, surface area of the solids, their reactivity and the thermodynamic free energy change associated with the reaction [2, 3]. Conventional solid state reaction technique which involves selection of raw materials, mixing, milling and calcination at high temperature is used in the preparation of **$Mn_{0.8+x}Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15**. Raw chemical powders of fine quality and purity were accurately weighed out in required amounts according to their stoichiometric formula. This is followed by mechanical mixing. Ball mill facility is utilized for this process which is of several hours long. After ball milling the milled powders are calcined at 30^0C, 500^0C, 800^0C and 950^0C for several hours. The powders are cooled, reground and again calcined. This process is repeated again and again until a homogeneous

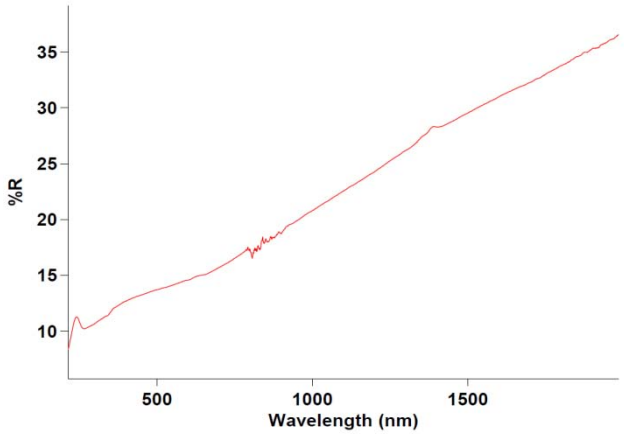
powder is obtained. The powder is pelletized. These pellets are sintered. The sintering environment including temperature, time and cooling rate play a vital role in obtaining high quality crystalline material. Control of temperature is often necessary to ensure that the desired crystalline phase is formed with optimum particle size [4]. Characterization studies are done using U-V analysis and Wemple- Di Domenico single-oscillator model. Absorption spectra and reflectance spectra of the sample are analyzed. Different types of optical constants such as refractive index, extinction coefficient, normal-incidence reflectivity, and absorption coefficient are calculated and their dependence on temperature is studied. Band gap energy is determined using Tauc method. Dispersion of refractive index is also analyzed

2.1 UV-VIS. Analysis

Absorption spectra and reflectance spectra of **$Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15** are analyzed by Ultraviolet (UV) absorption spectroscopy. It provides rapid real-time analysis. These spectra are studied at room temperature. The optical absorption data were analyzed using the classical relation for near edge optical absorption of semiconductors [5,6]. The prepared sample was subjected to UV-VIS-Near IR analysis (Fig.1) by using Varian, Cary 5000 Spectrophotometer over a spectral range of 175-3300nm with an accuracy of ±0.1nm (UV-Vis.). This type of sample which has undergone good heat treatment is mechanically so hard and has so much resistance to harsh environment. The prepared material also shows high thermal conductivity and large dielectric constant. The UV- VIS spectrum gives information about the excitonic and inter transition of nano materials [7]. Figure 1 shows Absorbance and reflectance spectra of the sample at 950^0C.



(a)



(b)

Figure 1: UV-VIS spectrum of **$Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15** (a) Absorbance (b) reflectance

Absorption spectrum is translated into diffuse reflectance spectra using Kubelka-Munk method. Kubelka-Munk's equation is described as follows: α =(1-R)2/2R --(1), α where is the absorption coefficient and R the reflectivity at a particular wavelength [8]. Kubelka and Munk proposed a system of two differential equations based on the model of light propagation. These two differential equations have two flux terms I and J.I is the flux of light in down direction and J is the flux of light in up direction. If the value of $I=I_t$ at the bottom of the prepared sample, then transmittance is given by the equation $T=I_t/I_0$--(2) and reflectance is given by the equation $R=J_r/I_0$ --(3)

The optical band gap can be determined using the Tauc relation. Here the optical band gap is determined by plotting the square root of the product of the absorption coefficient and photon energy versus photon energy. So obtained curve must contain a section of straight line. Then this small section of straight line is extended to the x-axis, and the x-intercept of this line gives the band gap. According to the Tauc relation, the absorption coefficient α for a material is given by $\alpha = A(hv- Eg)^n$-- (4), 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[9]. 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 [10,11,12] respectively. The TAUC plot of the prepared sample defines the optical band gap as the region A in fig.2. The tauc plot of the prepared sample is given in Fig 3. It is reported that optical gap energy of micro/nano–sized crystal depends on its crystallite size, it increases with decreasing crystallite size in the nano size range [13,14].

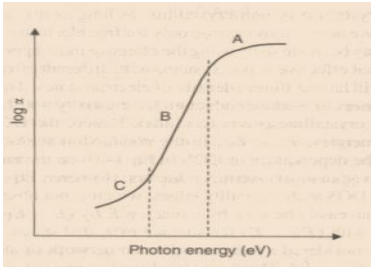


Figure 2: Optical band gap energy variation with absorption

The absorption coefficient at the x axis (photon energy)below (tail absorption) varies exponentially with the photon energy: $\alpha(\hbar \nu) \sim exp\ (\hbar \nu/E_u)$ --(5) where E_u is a term called Urbach energy.In this figure 2 the region B the 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 [15]. 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 [16].

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[17]. The extinction coefficient and the absorption coefficient are related as $\alpha(E) = 4\pi/\lambda\ (k(E))$—(6).

The change in the values of band gap energy with samples prepared at different temperatures is shown in figure4. The natural logarithm of the absorption coefficient, α(ν), was plotted as a function of the photon energy, hν (Fig.5). 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 latter can be controlled by improving the crystal growth and the purity of the ingredients. The temperature-dependent part of the Urbach tail, is purely of intrinsic reasons [18].

2.2 Refractive Index and Dispersion

Refractive index of the sample changes with wavelength. This variation of refractive index is what we call dispersion of refractive index. This dispersion can be described by a Cauchy relation. Cauchy‘s equation is a purely empirical relationship between Refractive index and wavelength. The most general form of Cauchy‘s equation is $n(\lambda) = B + C\ \lambda 2 + D\ \lambda 4 + \cdots$ --(7) where n is the refractive index, λ is the wavelength, B, C, D, etc., are coefficients that can be determined for a material by fitting the equation to measured refractive indices at known wavelengths. The coefficients are usually quoted for λ as the vacuum wavelength in micrometers. Usually, it is sufficient to use a two-term form of the equation: $n(\lambda) = B + C\ \lambda 2$ –(8) where the coefficients B and C are determined specifically for this form of the equation[19]. Refractive index is inversely proportional to wavelength (figure 6). Refractive index depends on temperature also. Refractive index is directly proportional to temperature. But it remains at a unique value after a particular wavelength.

The dispersion of refractive index below the interband absorption edge is analyzed using the Wemple-DiDomenico(W-D) model [20]. In the W-D model, the refractive index n can be written as $\mathbf{n^2 - 1 = E_d E_0 / (E_d^2 - E^2)}$ --(9),where E is the photon energy, E_o is the oscillator energy, and Ed is the dispersion energy. The oscillator energy E_o and dispersion energy E_d are obtained from the slope $(E_o E_d)^{-1}$ and intercept Eo/Ed on the vertical axis of the straight line portion of $(n^2-1)^{-1}$ versus E^2plot. The static refractive index $n(0)$ at zero photon energy is evaluated from Equation (9), i.e. $n^2(0) = 1 + E_d/E_o$—(10) [21]. Fig.7 shows the proportionality of dispersion of refractive index with temperature.

3. Results and Discussion

The Tauc plot relation is used for determining optical band gap. The equation $(h\nu\alpha)^{(1/n)} = A(h\nu - Eg)$—(11), is used for this purpose. Here h is Planck's constant, ν is frequency of vibration, α is absorption coefficient, Eg is band gap and A is proportional constant. The value of n denotes the nature of the sample transition. For direct allowed transition n = ½. For direct forbidden transition n = 3/2. For indirect allowed transition n =2. For indirect forbidden transition n = 3. Absorption spectrum is translated into diffuse reflectance spectra using Kubelka-Munk method. Thus, the vertical axis is converted to quantity F(R∞), which is proportional to the absorption coefficient. The α in the Tauc equation is substituted with F(R∞). Using the Kubelka-Munk function, the (hνF(R∞))2) was plotted against the hν. The curve that plots the value of (hν - (hνF(R∞))2) on the horizontal axis hν and vertical axis (hνF(R∞))2 is drawn. Here, the unit for hν is eV (electron volts), and its relationship to the wavelength λ (nm) becomes $h\nu = 1239.7/\lambda$ .A line is drawn tangent to the point of inflection on the curve and the hν value at the point of intersection of the tangent line and the horizontal axis is the band gap Eg[22]. The Tauc plot of the sample at temperatures 950˚C is given in Fig.3. It is found that band gap energy of sample $\mathbf{Mn_{0.8+x}\ Zn_{0.2}Ti_xFe_{2-2x}O_4}$ **with x=0.15** increases with temperature. The values of band gap energy of the sample at different temperature values is given in table -1.

Absorption spectra and reflectance spectra of $\mathbf{Mn_{0.8+x}\ Zn_{0.2}Ti_xFe_{2-2x}O_4}$ **with x=0.15** are studied by Ultraviolet (UV) absorption spectroscopy. It provides rapid real-time analysis. These spectra are studied at 30˚C (room temperature). The optical absorption data were analyzed using the classical relation for near edge optical absorption of semiconductors. Dispersion of refractive index is also observed. Refractive index of the sample changes with wavelength. This variation of refractive index is what we call dispersion of refractive index. This dispersion can be described by a Cauchy equation.The change in the value of refractive index is analyzed using the Wemple-DiDomenico(W-D) model.

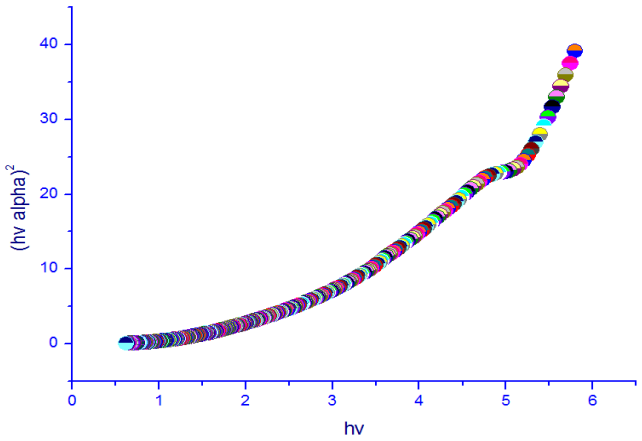


Figure 3: The Tauc plot of $Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ **with x=0.15**

Table 1: Band gap energy values of $Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ **with x=0.15** at different temperatures

Temperature	Band gap energy in eV
30◦C	4.22
500◦C	4.30
850◦C	4.34
950◦C	4.40

It is crystal clear that band gap energy varies with temperature (fig.4).The energy levels of the prepared sample depends on structural imperfection of the lattice. The band gap of the sample increases with size of the crystal.

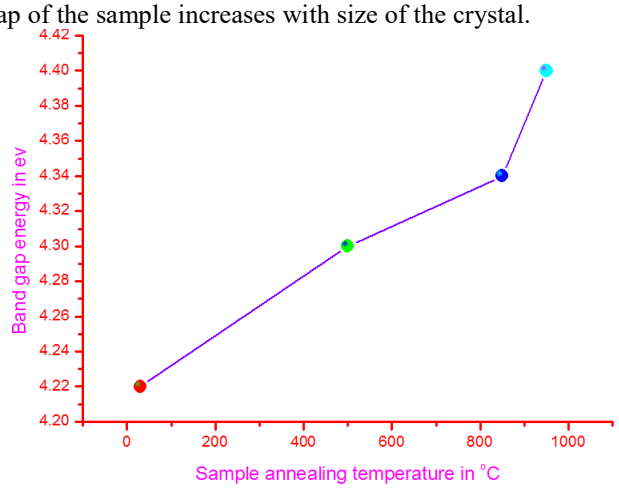


Figure 4: Band gap energy variation with the samples at varied annealing temperature of $Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15

As temperature increases the size of the crystal also increases and hence there will be an increase in band gap energy. It is implied from Tauc plot that the band gap energy is directly proportional to temperature. Since energy levels depend on extent of imperfection in the crystal lattice, the increase of orderly arrangement in ceramics leads to a reduction in energy and hence Eg values increases.

Urbach energy of our prepared sample can also be calculated. Among optical transitions, there is a case of extended to localized and localized to extended state transitions. For these types of transitions there is an exponential decay of density of states of the localized states into the optical band gap one can find an exponential relation between absorption coefficient and frequency:

$$\alpha(\omega) \sim \exp\left(\frac{\hbar\omega}{E_u}\right) \quad \text{--(12)}$$

where Eu is called 'Urbach energy'. According to Urbach rule, $\alpha=\alpha_o \exp[(hv - E_o)/\Delta E]$ _(8), where α_o and E_o are material-dependent constants ΔE, which is called the Urbach width, is also a material-dependent constant. Here Urbach energy is calculated from the graph by plotting the natural logarithm of the absorption coefficient with energy in units of eV (Figure 5). The calculated energy value is lower than the band gap energy. Hence Sumi-Toyozawa (ST) model is also applicable to this material.

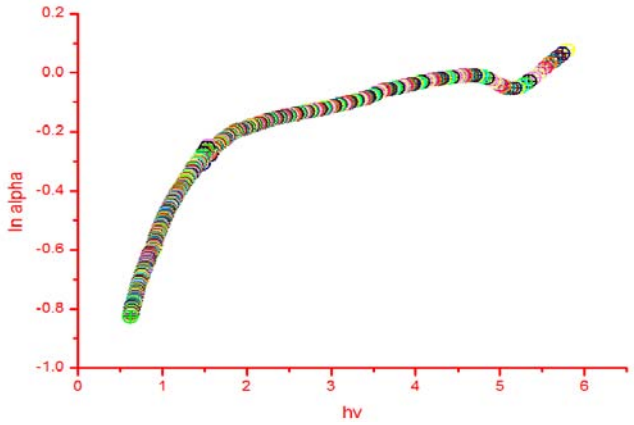


Figure 5: Absorption variation with photon energy of $Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ **with x=0.15**

Refractive index of ceramics varies with wavelength. This variation of refractive index is what we call dispersion of refractive index. This dispersion can be described by a Cauchy equation. Refractive index varies inversely with wavelength. Refractive index is also temperature dependent. Refractive index increases as temperature increases. But it remains at a unique value after a particular wavelength. Light energy gets dispersed at the different interstitial layers of crystal lattice. This dispersion is responsible for this indirect proportionality of refractive index with wavelength. Refractive index is also directly proportional to photon energy (fig.6). This phenomenon is caused by the crystallization in the single phase cubic spinel structure phase. The refractive index of these materials is proportional to their electronic polarization per unit volume. This electronic polarization per unit volume is in turn inversely proportional to distance between atomic planes. This is the reason for the increase in the size if the crystallite.

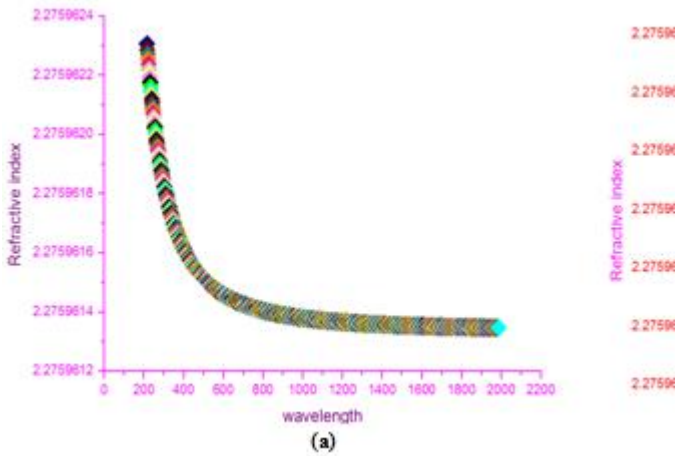


Figure 6: Variation **of** Refractive index **(n)** with (a) wavelength and (b) photon energy of **$Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15**

The variation of refractive index below the interband absorption edge is analyzed using the Wemple-Di Domenico(W-D) model. Graphical representation of results is given in (Fig.7).

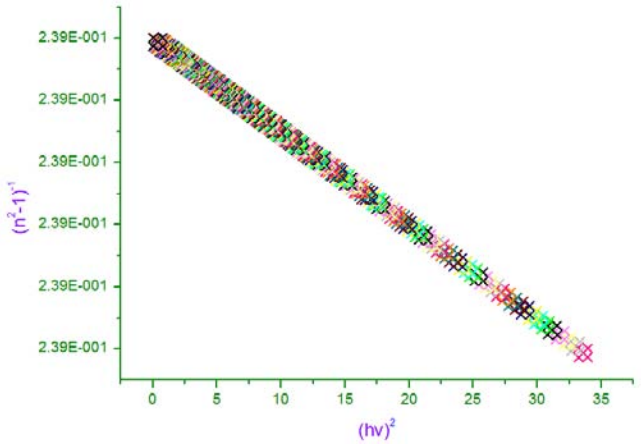


Figure 7: $(n^2-1)^{-1}$ versus$(\ hv)^2$ curve.

$(n^2-1)^{-1}$ is plotted against $(\ hv\ )^2$.This helps in the determination of the oscillator parameters. It can be done by the fitting of a linear function to the smaller energy data. E_o and E_d are measurable from the intercept (E_o/E_d) and the slope $(1/E_oE_d)$. E_o can be considered as an average energy gap. It is proportional to the Tauc gap $E_o \sim 2E_g$.

Another mentionable relation from Wemple-DiDomenico(W-D) model is $n^2\text{-}1=S_0\ \lambda_0^2\ /[1-(\ \lambda o\ /\ \lambda)^2\ ]$ --- (13), where λ, the wavelength of the incident radiation , S_o, the average oscillator strength and λ_o, the average oscillator wavelength. The graph obtained by plotting $(n^2-1)^{-1}$ against $(1/\lambda^2)$ (Fig.8) are linearly fitted obeying sell Meier's dispersion formula. S_o and λ_o are measurable from the slope (1/So) and the infinite wavelength intercept $(1/S_0\ \lambda_o)^2$. The optical parameters of the sample are calculated. These values are listed in table.2

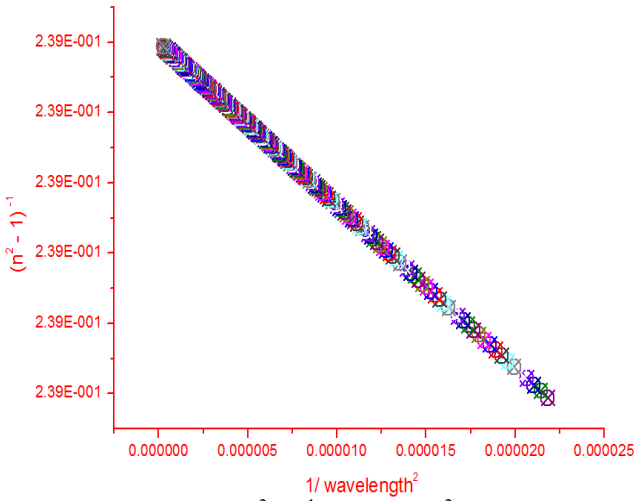


Figure 8: $(n^2\text{-}1)^{-1}$ versus $1/\lambda^2$ curve

Table 2.The calculated values of optical parameters of **$Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15**

sample	E_g (eV)	E_O(eV)
30◦C	4.22	8.44
500◦C	4.30	8.60
850◦C	4.34	8.68
950◦C	4.40	8.80

It is very clear that band gap energy is directly proportional to temperature where as dispersion energy is inversely proportional to temperature. The straight line graph confirms the sell Meier's dispersion formula.

4. Conclusion

As temperature increases there will be a significant shift in UV emission peak. From optical measurements it is crystal clear that absorbance and reflectance increases with temperature. Band gap energy shows a direct proportionality to temperature. High energy values of optical band gap plays a vital role in increasing the dielectric properties of the nanocrystalline ceramics. Tunable band gaps can be reached by varying annealing temperatures. Very high band gap energy and favourable optical properties of **$Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15** makes it a high potential candidate for UV-VIS shielding applications. Analyzing Wemple- DiDomenico single-oscillator model it is observed that dispersion energy lowers as the sample attains crystalline phase. At high temperatures nanocrystalline ceramic attains high energy values of optical band gap. Hence we can proudly say that, nanocrystalline ceramic material **$Mn_{0.8+x}$ $Zn_{0.2}Ti_xFe_{2-2x}O_4$ with x=0.15** becomes a very good dielectric at high temperature.

5. Acknowledgement

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