



## TUNABLE OPTICAL PROPERTIES OF SILICON DIOXIDE DISPERSED IN DIFFERENT SOLVENTS

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### ABSTRACT

The present study deals with the synthesis of silicon dioxide (SiO<sub>2</sub>) by sol gel method and also the investigation of its optical properties when dispersed in different coordinating and non-coordinating solvents. It was observed that absorption and emission properties of SiO<sub>2</sub> could be effectively tuned simply by altering the solvent environment. Slight variations in absorption coefficient indicated solvent interaction with the SiO<sub>2</sub> nanoparticles. Also, the band shapes varied in passing from coordinating to non-coordinating solvents which could be the result of change in solvent environment. The luminescence intensities of SiO<sub>2</sub> nanoparticles were found to be appreciably higher in case of water and acetone environments.

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### INTRODUCTION

Silicon dioxide (SiO<sub>2</sub>) is the second most abundant element exceeded by oxygen and weighs almost 25.7 % of the earth's crust (Leung *et al.*, 1993). It is inexpensive possessing excellent optical, mechanical and electrical properties but rarely found in pure crystal form in nature. As a stable insulator, it not only forms the heart of the metal-oxide-semiconductor (MOS) device but is also being utilized in therapeutics and diagnostic fields (Michalet *et al.*, 2005; Chan and Nie, 1998). The preferred sol gel method of synthesis of SiO<sub>2</sub> involves the evolution of a continuous inorganic network with homogeneous pore distribution profile. In the process, at first a colloidal suspension of SiO<sub>2</sub> is prepared which is then subjected to gelation. This results in a network characterized by a continuous liquid phase (gel). The elements react together to form Si-O-Si bonds and it is the arrangement of the atoms in the network that gives a specific shape to the SiO<sub>2</sub> particles. It is well known that the luminescence properties of SiO<sub>2</sub> nanoparticles are multicolored and size dependent. That is why; they are preferred for biological imaging (Bruchez Jr. *et al.*, 1998; Lacoste *et al.*, 2000; Weng *et al.*, 2005). Moreover, they are less toxic, highly stable in environment and also offer high quantum luminescence efficiency (Hayakawa and Nogami, 2005; Dabbousi *et al.*, 1995). Different researchers have investigated the synthesis of SiO<sub>2</sub> using different solvents (Naziruddin Khan, 2014; Aldwayyan *et al.*, 2013; Naziruddin Khan, M. and Al Dwayyan, 2008). However, few investigations have concentrated on the effect of dispersing solvents in modifying the optical absorption and emission properties of SiO<sub>2</sub> nanoparticles.

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In this study, the main aim is to investigate the optical properties of sol gel prepared SiO<sub>2</sub> nanoparticles dispersed in different solvents. We employed various solvents such as acetone, ethanol and water. Their varied surface functional groups enable them to form hydrogen bonds with SiO<sub>2</sub> and thereby alter their luminescence efficiencies.

### MATERIALS AND METHOD

Materials were of analytical grade and used without purification. Tetra ethyl orthosilicate (TEOS) was purchased from Sigma Aldrich, Germany and ethanol, acetic acid, sodium hydroxide (NaOH) and acetone from Merck, India. Doubly distilled water was used in all the experiments.

Tetra ethyl orthosilicate (TEOS) was mixed with water in the molar ratio 1:4. Acetic acid acted as the catalyst to promote the hydrolysis of TEOS. The mixture was agitated in a magnetic stirrer for 3 h. The resulting solution was centrifuged and then filtered. The filtrate was dried in an oven for 24 h at 100 °C. White crystals of SiO<sub>2</sub> were obtained.

#### Characterization techniques

The diffraction pattern of synthesized SiO<sub>2</sub> was recorded by an X'Pert PRO PANalytical X-ray diffractometer (XRD) using CuK $\alpha$  radiation (wavelength  $\lambda = 0.15418$  nm) at an accelerating voltage and applied current of 30 kV and 20 mA. The data was collected in the range of  $2\theta = 10^\circ - 80^\circ$  with a step of  $0.02^\circ$ . The chemical identity was ascertained by comparing with standards compiled by the Joint Committee on Powder Diffraction and Standards (JCPDS). Uv-vis absorption spectra were recorded in a UV 3600 spectrophotometer, with the sample contained in a 1 cm<sup>3</sup> stoppered quartz cell of 1 cm path length in the region 200-800 nm. Photoluminescence emission

spectrum was obtained by employing suitable excitation wavelength using PTI QM 40 spectrophotometer with a 150 W xenon lamp as the excitation source. The morphology as well as elemental composition of the samples was ascertained by Scanning Electron Micrograph (SEM) Evo LS10, Zeiss coupled with energy dispersive spectra (EDS).

## RESULTS AND DISCUSSION

### XRD analysis

The diffraction pattern of SiO<sub>2</sub> (Fig 1) showed a broad peak approximately at 2θ of 23° indicating the amorphous characteristic of the sample.

The particle size (D) was calculated using Scherer's equation (1) (Patterson, 1939) given by:

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

Where K is a constant called shape factor (0.9) depending on the shape of the nanoparticles, λ is the wavelength of X-ray (λ = 1.54 Å for Cu-K<sub>α</sub> radiation), θ is Bragg angle, β the full width half maxima. The estimated particle size was 1 nm.

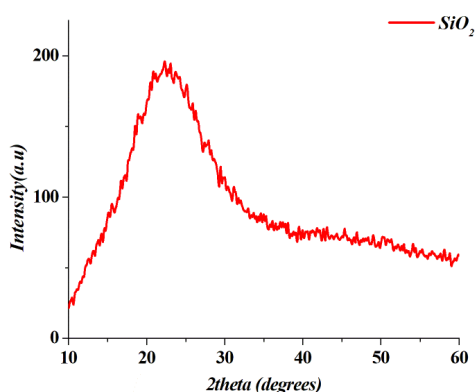


Figure 1 Diffraction pattern of nano SiO<sub>2</sub>.

### Composition

In EDS imaging (Fig 2), elements silicon (Si) and oxygen (O) were observed in the energy ranges 0-1 KeV and 1.6-1.73 KeV respectively (Jiao *et al.*, 2007). The additional peak of chlorine (Cl) could be due to the environment in which the sample was preserved.

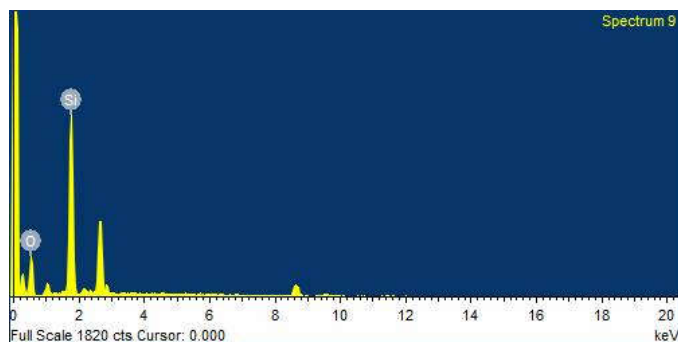


Figure 2 EDS showing elemental composition of SiO<sub>2</sub>.

### Absorption spectra

Optical absorption spectroscopy was employed to investigate the optical properties of synthesized SiO<sub>2</sub> nanoparticles in the

Uv-vis (200-600 nm) region in different solvents at room temperature. Fig 3(a)-3(c) displays the absorption spectra of as prepared SiO<sub>2</sub> in different solvents such as ethanol, acetone and water. The peak positions are similar but SiO<sub>2</sub> dispersed in acetone shows the highest absorbance.

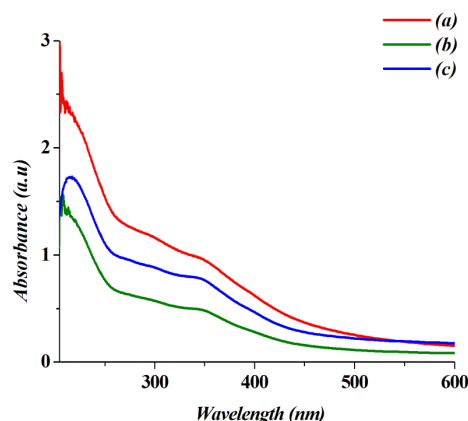


Figure 3 Absorption spectra of SiO<sub>2</sub> in (a) acetone (b) ethanol and (c) water.

### Optical band gap

The optical band gap (E<sub>g</sub>) was calculated using Tauc relation (2) (Tauc, 1970):

$$\alpha h\nu = C(h\nu - E_g)^n \quad (2)$$

Where α is absorption coefficient and n the type of transition. Fig 4 shows the Tauc plot from which the band gaps of SiO<sub>2</sub> have been estimated to be 4.21eV, 4.34 eV and 4.27 eV in acetone, ethanol and water respectively. These high band gap values can be attributed to the amorphous phase and predict sensitivity to the ultraviolet zone. It is interesting to observe that simply by changing the dispersing solvent, the band gap of SiO<sub>2</sub> could be tuned. Thus, the role of dispersing solvents cannot be totally ignored in recording the optical properties of a nanoparticle.

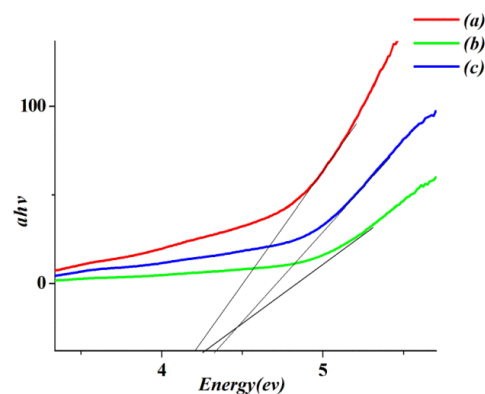


Figure 4 Band gap plot for SiO<sub>2</sub> in (a) acetone (b) ethanol and (c) water solvent.

### Photoluminescence (PL) spectra

It was interesting to note that when SiO<sub>2</sub> was dispersed in ethanol, luminescence band was observed in the ultraviolet zone which could be attributed to triplet-singlet transition (Paleari *et al.*, 2005). However, when it was dispersed in acetone and water, characteristic blue and blue-green emissions were observed. This could be due to the presence of self-trapped excitons (Paleari *et al.*, 2005; Weng *et al.*, 2005).

The PL spectra of SiO<sub>2</sub> showed appreciably high intensities in both acetone and water. This is particularly promising for optoelectronic applications. The most important point when SiO<sub>2</sub> is dispersed in water a broad emission band observed covering a large wavelength region which is associated with the formation of a band structure instead of the energy level. Not only that high intense emission spectra are observed because of a large number of electrons accumulated in the conduction band which is favorable for the application in optoelectronics devices. So, in presence of water molecules, SiO<sub>2</sub> particles disperse homogeneously as well as efficient for providing emissive nature covering all the visible range having the formation of a large number of excitons which may be capable to entrap energy efficiently.

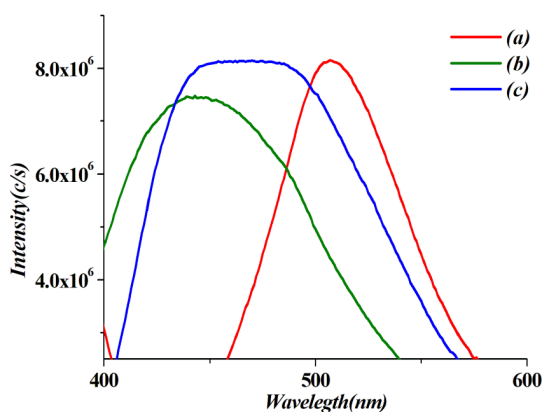


Figure 5 Photoluminescence (PL) emission spectra of SiO<sub>2</sub> in (a) acetone (b) ethanol and (c) water solvent.

### IR spectra

Fig 6 showed the transmittance spectra of SiO<sub>2</sub>. Peaks at 2925 and 3475 cm<sup>-1</sup> are due to O-H stretching vibrations (Brinker and Scherer, 1990). The first one at 2925 cm<sup>-1</sup> corresponds to intra-molecular rotation or scissoring movement and the second one at 3475 cm<sup>-1</sup> corresponds to elongation movement. The bands at 800 and 953 cm<sup>-1</sup> can be attributed to Si-OH stretching while those at 1045, 1080 and 1220 cm<sup>-1</sup> correspond to Si-O-Si bonding (Zarubin, 2001; Bai, 2017). Two types of vibration possible for Si-O-H, one corresponds to symmetric and another corresponds to asymmetric. Three types of stretching are possible for Si-O-Si bonding, the first one at 1045 cm<sup>-1</sup> corresponds to symmetric stretching, the second one at 1080 cm<sup>-1</sup> corresponds to asymmetric stretching and the third one at 1220 cm<sup>-1</sup> corresponds to bending or scissoring movement arose due to the movement of Si atom with respect to oxygen atom having highest degrees of freedom with the formation of agglomeration as depicted in SEM micrograph.

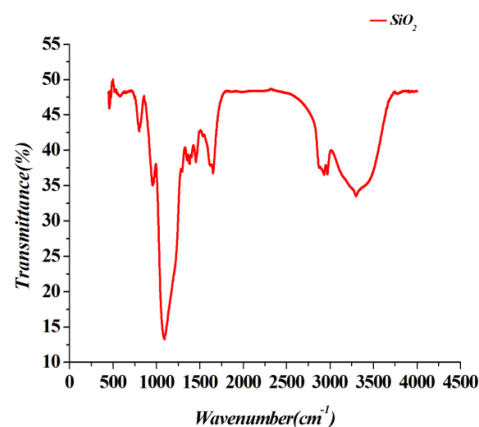


Figure 6 IR spectra of SiO<sub>2</sub> demonstrating functional bonds.

### Morphology

The SEM (Fig 7) images of SiO<sub>2</sub> were agglomerated structures with presence of moisture. It seemed as if small spheres have agglomerated to form irregular structures.

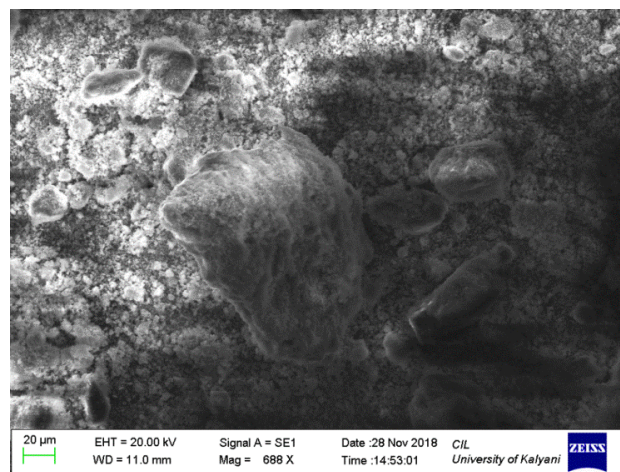


Figure 7 SEM image of SiO<sub>2</sub> indicating morphology.

### CONCLUSIONS

In the present study, silicon dioxide has been prepared by sol-gel route. The optical properties were recorded by dispersing SiO<sub>2</sub> in different solvents. It was observed that the solvent environment played a role in tuning the optical properties of the nanoparticles. This result can prove a breakthrough as regards optoelectronic applications. Also, the sample demonstrated high luminescence which makes it suitable for use in light detectors.

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