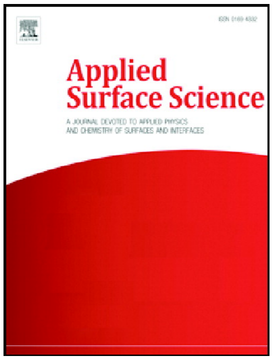

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Sol-gel synthesized plasmonic nanoparticles and their integration into dye sensitized solar cells

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ABSTRACT

In the present study, we synthesized silver nanoparticles in titanosilicate matrix through low coat non-hydrolytic sol-gel method using titanium isopropoxide (TIP), tetraethyleorthosilicate (TEOS, ($Si(OC_2H_5)_4$) and ethanol (C_2H_5OH) as solvents. These were investigated for the plasmonic effect of nanoparticles in dye sensitized solar cells (DSSC). The fabricated dye-sensitized solar cells with Ag: SiO_2-TiO_2 films coated in fluorine dopedtin oxide (FTO) glass plate with dye of amaranthus red shows an improved output voltage. The samples were optically and structurally well studied by absorption spectroscopy, fourier transform infrared spectroscopy(FTIR), X-Ray diffraction (XRD) and transmission electron microscopy(TEM). The XRD studies confirmed the crystalline nature of TiO_2 and Ag.

1. Introduction

Noble metal nanoparticles research has attracted a lot of attention due to a wide range of potential applications in plasmonic solar cells, surface enhanced Raman scattering, surface enhanced fluorescence and metamaterial absorbers to count a few [1-13]. Plasmonics is one of the most important fields that make use of the nanoscale properties of noble metals. It extracts the surface plasmon resonance properties of nano particles. In plasmonic solar cells, plasmonic structure excites more electrons due to increased light absorption and scattering and the plasmon-polaritons trapped more incident light within the structure. These factors favor the enhancement of efficiency of plasmonic DSSCs compared to normal DSSCs. [14-17]. Due to increasing energy consumption, there is a great need for new inexpensive sources of renewable energy. Sunlight is plentiful, inexhaustible and eco-friendly source of energy and therefore trapping of solar energy is established as one of the interesting research areas [18-20]. The energy harvesting may be improved by plasmonic nanoparticles (NPs) in two ways (i) by adopting light trapping schemes and (ii) exploiting spectral modification processes to shift frequencies of the solar spectrum. NPs can give a significant boost to both these aspects, by scattering and concentrating the electromagnetic field into the active region of the device. Tuning of specific spectral regions by optimizing the size, shape, distribution of the plasmonic NPs, and by choosing the right surrounding medium, one can also enhance the aforesaid aspects [21-24]. Further broad band optical absorption has tremendous potential for solar energy harvesting and it is a necessary condition for improving the efficiency of solar cells[25-27]. One of the factors that determines the performance of solar cells is the efficiency of the light absorption process that generates electron-hole pairs, as well as the subsequent extraction of these charge carriers. DSSCs with incorporation of plasmonic nanoparticles are promising alternatives to traditional solar cells [18, 28-30]. Recently solar cells such as DSSCs and organic solar cells (OSC) have received promising role for renewable energy exploration due to low cost, simple fabrication techniques, flexibility, and low toxicity. One of the main disadvantages is the lower energy-conversion efficiency compared to other solar cells [31-33].

Researchers have reported many methods for the synthesis of NPs like sol–gel techniques, melt-quenching, ion exchange, ion implantation etc. Sol-gel method is one of the important techniques used for nano scale synthesis and it has several advantages over other methods such as ultra homogeneity, high purity, low processing temperatures and possibility of making new glass/ceramic compositions[34-35]. Researchers are now looking for efficient photon energy harvesting solar cells. Preferences are given to develop solar cells using simple fabrication techniques which are more efficient and cost effective. The plasmonic DSSC solar cell fabrication has the advantage of incorporation of materials which are having superior optical properties with an enhanced and broadened absorption bandwidth. We present here the plasmoic effect of sivler nanoparticles in titanosilicate matrix for DSSC applications.

2. Experimental

In the present study highly dispersed silver NPs were successfully prepared by a simple and conventional non hydrolytic sol gel technique. For this we used raw materials such as titanium isopropoxide (TIP), tetraethyleorthosilicate (TEOS, ($Si(OC_2H_5)_4$) , ethanol (C_2H_5OH) and silver nitrate ($AgNO_3$). Figure 1 shows the scheme of the experimental procedure and the prepared samples are listed in Table 1. For the synthesis of silver NPs via sol-gel process we used a stoichiometric solution of titanium isopropoxide(TIP), TEOS, ethanol, nitric acid and most importantly silver nitrate. Homogeneity of the mixture of the samples was ensured by magnetic stirring of the solution. As shown in the scheme two solutions were prepared initially. First solution is a mixture of TEOS and ethanol. Second one is a mixture of TIP and ethanol. These solutions were separately prepared and latter was added to a third beaker and stirred well using magnetic stirrer. During stirring a few drops of HNO_3 was added which acts as a catalyst for the reaction. Weighed amount of silver nitrate was added to the above mixture. The stirring process was continued until a clear solution was obtained which indicates a homogeneous solution of the mixture. The procedure is repeated to prepare two more sets of solutions with different compositions. The four different homogenous solutions were poured into small polypropylene containers and tightly sealed using parafilms. These sealed samples were then placed in a dry place without disturbing the sample for a period of one month and allowed them for the gel formation[36-39]. A part of the solution of the samples is used for preparing films in FTO glass plate. The densified samples are characterized by using FTIR, XRD, TEM and optical absorption measurements.

3 Results and Discussion

3.1 Optical Extinction studies

Figure 2 shows the optical extinction spectra of the sol-gel samples synthesized via non hydrolytic method. The broad absorption is due to the plasmonic effect of silver particles in SiO_2 –TiO_2 sample. The heated samples at 800 ºC show a broad band around 300-400nm which is due to the surface plasmon resonance of the silver nano particles [34, 38-39]. The surface plasmon peak of silver NPs around 350-400 nm is due to the collective oscillation of all the free electrons in the silver nano particle resulting from the interaction with the electromagnetic radiation. The oscillation wavelength depends on size and shape as well as the nature of the surrounding medium in which the particle is embedded. Comparing all three silver doped samples in SiO_2 -TiO_2 matrices, sample annealed at 800ºC with silver concentration of 3 wt% shows maximum optical extinction.

3.2 Fourier Transform Infrared Spectroscopy

Figure 3 shows the FTIR spectrum of silver nano particles doped titanosilicate sample at room temperature and annealed at 800ºC . The room temperature sample shows many peaks in the range 400 to 4000 cm^{-1} which is due to the presence of water molecules and precursors as residues in the sample. When the sample is heat treated most of the peaks disappeared and proper linkage of Si-O-Ti is visible(920 cm^{-1}).

It is reported that the FTIR bands within the range of 900–1000 cm^{-1} are having composite features of Si–OH, Ti–OH and Si–O–Ti. The bands observed in the 860-990 cm^{-1} range are typical for TiO_2-SiO_2 glass. The peak observed at 1069cm^{-1} is assigned to Si-O-Si asymmetric stretching mode . The peak observed at 462 cm^{-1} corresponds to the Si-O-Si symmetric stretching [2,36-39].

3.3 X-Ray Diffraction(XRD)

The crystalline nature is studied by using X-ray powder diffraction method. The XRD spectrum confirmed the crystalline nature of silver particles. The broad peak in the diffraction pattern centered at $2\theta \sim 23^0$ is due to the amorphous nature of SiO_2 . The narrow peaks centerd at values of 25^0, 48^0, 54^0 and 62^0 correspond to (1 0 1), (2 0 0), (2 1 1) and (2 0 4) planes of TiO_2. The values at 38^0 , 44^0, 64^0, 78^0 correspond to (1 1 1), (2 0 0) , (2 2 0) and (3 1 1) planes of silver (ICDD file 04-0783).

Using the well-known Debye-Scherrer formula

$$\text{Particle size} = \frac{0.9\,\lambda}{\beta \mathrm{Cos}\theta}$$

the particle size was estimated from the XRD spectrum. where β is the measured FWHM, 2θ is the Bragg angle of diffraction peak and λ is the X-Ray wavelength. The major peak identified for silver particles at $2\theta \sim 38^0$. The value of FWHM from the XRD pattern is 0.50. The calculated size of silver nanoparticle using Scherrer formula is 17.5 nm [37-40].

3.4 Transmission Electron Microscopy (TEM)

The actual size, size distribution and shape of particles are determined using the TEM images. Figure 5 shows the TEM image of silver nanoparticles at different magnifications, which reveals the high resolution morphology of the nanostructured particles. The histogram of particle size is shown in figure 6. The average size of silver nanoparticles calculated from TEM images is found to be 16 nm[39].

3.4 Fabrication of DSSC

Sol-gel synthesized samples of SiO_2-TiO_2 and SiO_2-TiO_2+Ag3% were used for fabrication of DSSC. Each of these samples was coated on the conducting side of the Fluorine Tin Oxide (FTO) using a glass rod. The coated film plates were placed in an oven for 30 minutes at 75ºC and then the plates were allowed to cool to room temperature. Then a few drops of natural dye extracted from amaranthus red is incorporated into the film. After incorporating the dye the films were annealed in an oven at 450 ºC. Graphite coated on the conducting surface of the FTO glass plate was used as a counter electrode. These two plates were sandwiched together and secured with binder clips. The output voltage of the DSSC was measured using a digital multimeter. The measured output is shown in figure 7 and the values are tabulated in table 2. The measurement shows appreciable improvement for the output voltage due to the presence of the silver NPs in the SiO_2-TiO_2 matrix. This result is in conformity with earlier reports[41-42].

3.5 Conclusion

The SiO_2-TiO_2 doped silver nano particles were successfully synthesised using non-hydrolytic sol-gel method. The absorption spectrum of silver doped silica-titania reveals that there is a broad peak in the 380-410 nm range which is due to the surface plasmon resonance effect of silver nano particles. The sample with 3wt% doped silver and annealed at 800ºC show the highest plasmonic band. The nanoparticles formed under thermal treatment at 800ºC were characterized by using FTIR, XRD and TEM. The FTIR spectrum confirmed the Si-O-Ti linkage formation. The crystalline nature of the silver NPs is analysed through XRD and size is evaluated as 17.5 nm. The TEM measurements confirmed the crystal planes of silver and average size is found to be 16nm. We also successfully fabricated the plasmonic dye sensitized solar cell. The effect of NPs on DSSC is experimentally demonstrated and found that plasmonic effect can be used for improving the efficiency of solar cells.

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FIGURES CAPTIONS

Figure 1 Chart diagram of sol-gel SiO_2-TiO_2: Ag Sample Preparation

Figure 2 Absorbance of Ag doped sample

Figure 3 FTIR spectrum of Ag doped sample

Figure 4 XRD Spectrum of Ag doped SiO_2-TiO_2Sample

Figure 5 TEM image of silver nanoparticles

Figure 6 Histogram plot of Ag Nanoparticles

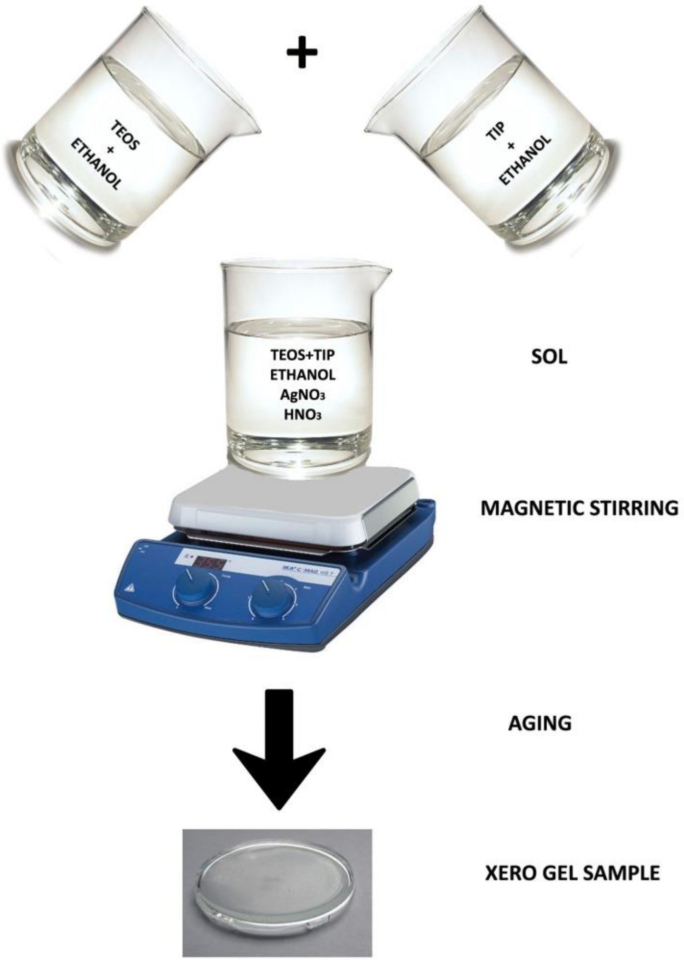
Figure 7 Measured output of fabricated DSSC with (a) SiO_2-TiO_2 Film (b) SiO_2-TiO_2 + Ag 3% film

TABLE CAPTIONS

Table 1 Sol-gel prepared SiO_2-TiO_2: Ag Sample

Table 2 Measured DSSC Outputs

FIGURES AND TABLES



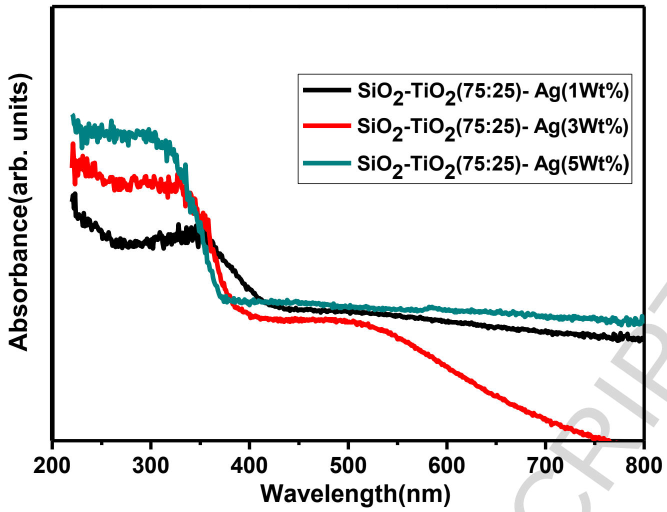


Figure 2 Absorbance of Ag doped sample

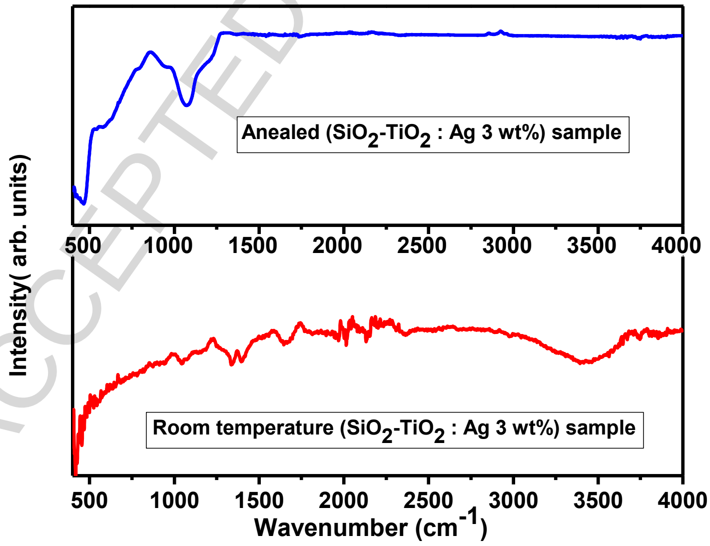


Figure3 FTIR Spectrum of Ag doped sample

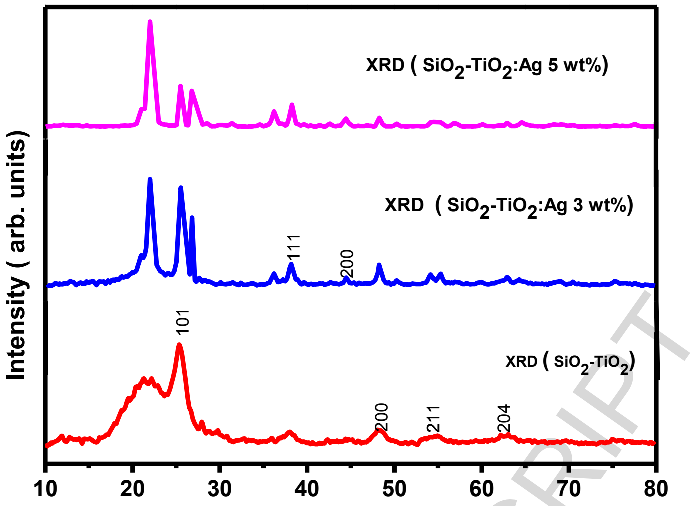


Figure 4 XRD spectrum of Ag doped SiO_2-TiO_2

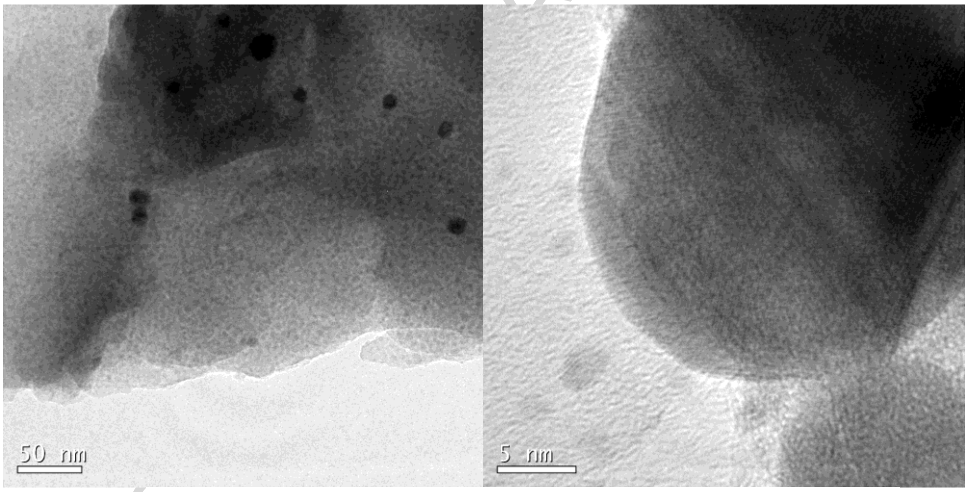


FIGURE 5 TEM IMAGE OF SILVER NANOPARTICLES

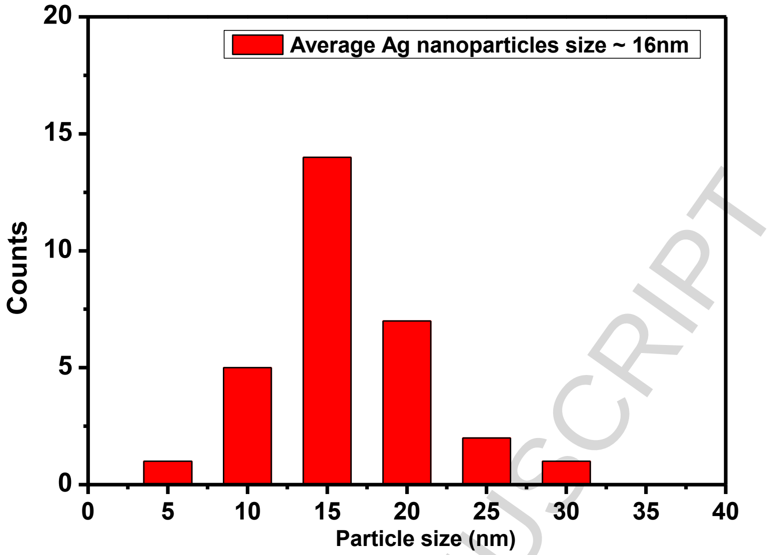


Figure 6 Histogram plot of Ag nanoparticles

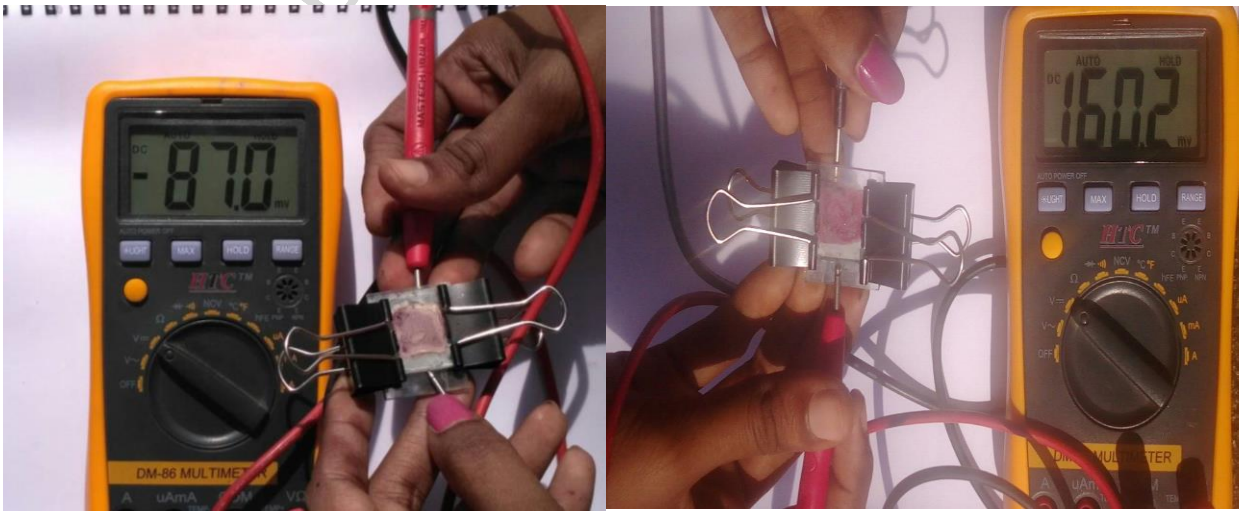


FIGURE 7 MEASURED OUTPUT OF FABRICATED DSSC WITH (A) SiO_2-TiO_2 Film (b) SiO_2-TiO_2 + Ag 2% fil

Table 1	
Sample	**Details of sample**
Sample 1	**SiO_2-TiO_2(75:25)**
Sample 2	**SiO_2-TiO_2(75:25)- Ag(1Wt%)**
Sample 3	**SiO_2-TiO_2(75:25)- Ag(3Wt%)**
Sample 4	**SiO_2-TiO_2(75:25)- Ag (5Wt%)**

Table 2 Measured DSSC Output

No	**Film coated on FTO glass**	**Measured DSSC voltage (mV)**
1	Dye (amaranthus red) doped SiO_2-TiO_2 Film	87
2	Dye (amaranthus red) doped SiO_2-TiO_2+Ag3% Film	160

HIGHLIGHTS

- Synthesis and structural characterization of plasmonic nanoparticles obtained through non hydrolytic sol-gel method.
- Integration of plasmonic nanoparticles into DSSC solar cells.
- Fine tuning the efficiency of DSSC using an optimum amount of Organic dye and Plasmonic nano particles