Abstract

Dye-sensitized solar cells (DSSCs) have potential to replace conventional solar cells due to ease of fabrication and cost effectiveness. Till date, significant work has been done on DSSC, yet power conversion efficiency is far away from its theoretical value. Therefore, systematic and detailed investigation is required for the efficiency enhancement in DSSC. Out of the various components , photoanode plays the role of heart in DSSC, therefore in this thesis, our prime focus is to fabricate efficient photoanodes for the application in DSSC. Light harvesting and recombination losses are the primary processes which are responsible for controlling the efficiency of photoanodes in DSSC. To improve these processes, mesoporous silica and different anisotropic shaped single crystalline silver nanoparticles have been investigated in the present thesis work.

Mesoporous silica has been explored in DSSCs to improve the light harvesting as well as suppression of back recombination without affecting the amount of dye loading on TiO_2. Mesoporous silica was synthesized using tetraethyl orthosilicate and polyethylene block polymer as precursors. Synthesized silica was characterized by X-ray photoelectron spectroscopy, X-ray diffraction, Brunauer Emett and Teller measurement, Scanning electron microscopy and Transmission electron microscopy. The effect of morphology of photoanodes for the incorporation of mesoporous silica, on the power conversion efficiency of DSSCs was investigated by fabricating photoelectrodes using single layer approach (where mesoporous silica was introduced by mixing with TiO_2 paste) and double layer approach (where first layer is of TiO_2 paste and second layer contains mesoporous silica mixed in the TiO_2 paste). Finally these photoelectrodes were used for the fabrication of DSSCs and were studied for their photovoltaic performance. It was observed that mesoporous silica had optimum effect when used in single layer approach. The lower efficiency in double layer approach (2%) in comparison to single layer approach (2.6%) is due to the poor contact between first layer and second layer which in turn leads to the loss of charge carriers at the interface and is further supported by decrease in fill factor, decrease in dark current onset potential as well as decrease in recombination resistance. In addition to this, DSSCs were fabricated by incorporating different weight% of mesoporous silica in TiO_2 paste. An improvement of 50% was observed for devices fabricated using 0.75 wt% of mesoporous silica. Possible mechanism behind the improvement was investigated using Electrochemical Impedance Spectroscopy and UV-Vis spectroscopy.

Further, the effect of crystallinity of silver (Ag) nanoparticles on their dephas-

ing time of Surface plasmon resonance (SPR) was demonstrated. Our theoretical formulation indicates that the dephasing time is higher for single crystalline Ag nanoparticles as compared to that of polycrystalline nanoparticles, which is attributed to the presence of scattering centers in the latter. This suggests that single crystalline Ag nanoparticles are interesting candidates for the enhancement of effective absorption cross-section of dyes. In order to validate our theoretical formulation, synthesis of single crystalline and polycrystalline Ag nanoparticles was done using chemical reduction of silver nitrate with sodium borohydride and their effect on absorption cross-section of di-tetrabutylammonium cis bis(isothiocyanato)bis(2,2-8bipyridyl-4,4dicarboxylato)ruthenium(II) (N719) dye was studied. It was observed that dye incorporated with single crystalline Ag nanoparticles showed a significant enhancement as compared to polycrystalline Ag nanoparticles (24.42% in solution, 21.01% in thin film form in single crystalline Ag nanoparticles while 8.52% in solution, 7.97% in thin film form in polycrystalline Ag nanoparticles respectively). Moreover, synthesis of spherical as well as different anisotropic shaped Ag nanoparticles was done using chemical reduction of silver nitrate with sodium borohydride by varying the concentration of cetyl trimethylammonium bromide (CTAB). The X-Ray diffraction of all synthesized Ag nanoparticles revealed fcc structure. Capping of CTAB on the surface of Ag nanoparticle was confirmed by Fourier transform infrared spectroscopy. The morphological analysis using Transmission electron microscopy showed formation of different anisotropic shapes such as oval, rod, cubical, pentagonal and prismatic when CTAB concentration is kept at 4 mM. The single crystalline nature of all different shaped Ag nanoparticles was confirmed by Nano beam diffraction patterns. The presence of multiple SPR peaks for different anisotropic shapes of Ag nanoparticles was revealed by UV-vis spectroscopy and was further utilized to obtain broadband enhancement in commonly used N719 dye in DSSCs. It was observed that the incorporation of different anisotropic shaped Ag nanoparticles into the dye resulted in a broadband enhancement of its absorption ($\sim$65%), which is much more as compared to that incorporating spherical shaped Ag nanoparticles ($\sim$21%). This enhancement was attributed to the improvement in the effective cross-section of the dye upon incorporation of different anisotropic shaped Ag nanoparticle shapes.

The investigations show that mesoporous silica and different anisotropic shaped single crystalline silver nanoparticles are the promising candidates for the efficient photoanodes in DSSC.