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Optical investigation of soda lime glass with buried silver nanoparticles synthesised by ion implantation

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A B S T R A C T

In the present research work silver nanoparticles embedded soda lime glass nanocomposites were fabricated by Ag^+–Na^+ ion-exchange technique followed by 200 keV Ar^+ ion implantation at normal incidence with fluence of 1×10^{15}, 1×10^{16}, 5×10^{16} and 1×10^{17} ions cm^{-2}. Modifications induced in sample morphology were investigated using field-emission electron microscopy (FE-SEM) while energy dispersive analysis of X-rays (EDAX) spectroscopy was used for compositional analysis. UV–visible spectroscopy was utilized to carry out a comprehensive optical analysis of these nanocomposites so as to estimate some of the useful optical parameters like optical energy gap (E_g), Urbach's energy (E_u), refractive index (n), real (ε_1) and imaginary (ε_2) parts of dielectric function, skin depth and optical conductivity. Appearance of a narrow band in the transmission spectra around 342 nm in UV region for the nanocomposites synthesised with the doses of 1×10^{16}, 5×10^{16} and 1×10^{17} Ar^+ ions cm^{-2} can be of significant importance as UV based narrow band pass filters. The optical energy gap of virgin soda lime glass decreased from the value of 3.65 eV to 3.30 eV for silver nanoparticles embedded soda lime glass nanocomposite with a dose of 1×10^{17} Ar^+ ions cm^{-2} whereas Urbach's energy showed an increase from 0.19 eV for virgin soda lime glass to 1.09 eV for the same sample. Similarly, refractive index increased from the value of 1.51 for virgin glass to the value of 1.83 for the sample with 1×10^{17} Ar^+ ions cm^{-2} at 500 nm. In addition to these optical parameters, dispersion behavior of refractive index of thus formed nanocomposites has also been studied on the basis of single-effective-oscillator model put forward by Wemple and DiDomenico.

1. Introduction

A composite in general is a combination of two or more materials such that the resulting material is a blend of improved unusual properties. When at least one of the materials in a composite is in nanoscopic regime then it gives rise to a new novel class of materials called nanocomposites [1]. For past few years research community is trying hard to generate different types of nanocomposites as they are assumed to be high performance materials and offer potential for numerous applications involving mechanically reinforced light weight components, nonlinear optics, sensors and many other systems extending from biomedical techniques to packaging [2]. Among different classes of nanocomposites, the one with noble metallic nanoparticles (Au, Ag, Cu) are attracting enormous attention because of their exceptionally unique optical features owing to surface plasmon resonance (SPR) which led to the emergence of new multidisciplinary fields like nanophotonics, nanoplasmonics, nanobiotechnology, etc. [3]. Surface plasmon resonance is a phenomenon in which free electrons are excited coherently and give rise to in phase oscillations at the periphery of metal-dielectric contact, for noble metal nanoparticles the frequency of these resonance oscillations lie in the visible region and are observable when size of the metal nanoparticles is smaller than the wavelength of the incident electromagnetic radiation [4]. Shape, size and distribution of metal nanoparticles govern the frequency and intensity of SPR peak, moreover the SPR peak is also very sensitive to the nature of the surrounding media as they provide possibility to tailor the chemico-physical behavior of these metallic nanoentities [5].

There are numerous reports in literature which reveal that the transparent dielectric media with buried metallic nanoparticles exhibit fascinating linear and non-linear optical properties that have paved way to new developments in optical industry [6,7]. Among various transparent dielectric media soda lime glass holds a special place as an encapsulating host for the noble metallic nanoparticles as it is known to possess high optical transparency, good insulating properties, high mechanical strength, easy fabrication in preferred shapes and sizes, capability to endure highly intense radiations, averting air oxidation of

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metal nanoparticles and also its easy availability at commercial level. Thus glass based nanocomposites are excellent candidates for designing optical switches, optical sensors, memory devices, solid-state lasers, solar cells, optical waveguides, etc. [8].

Inspite of the innumerable advantages, applications and opportunities offered by these novel class of materials, still challenges like agglomeration of nanoparticles under the influence of weak van der Waal's forces, control over their structure and chemistry need to be tackled so as to optimize their unique optical features [9]. The various advanced applications in optical industry not only demand appropriate choice of the host matrix but also a suitable fabrication mechanism as precise control over shape and size and uniform distribution of nanoparticles is must to harness their capabilities to extremum [10]. There are different pathways through which metal glass nanocomposites can be synthesised. Some of them are ion implantation, two-step method such as ion exchange followed by appropriate treatments such as thermal annealing in air or any suitable atmosphere or laser or ion irradiation, sol-gel method, co-sputtering, etc. [11,12]. In this experiment silver nanoparticles embedded soda lime glass nanocomposites have been fabricated by employing ion-exchange technique followed by Argon ion implantation. Ion-exchange method is one of the widely used methods of introducing desired metal ions into the glass networks and is well known for offering opportunities like freedom to select the ions to be exchanged that may be monovalent or polyvalent and flexibility to choose different glass compositions [13]. On the other hand ion implantation is also a well-established route of generating nanocomposites with uniformly distributed nanoparticles. The increasing popularity of this technique is due to the fact that the overall near-surface properties of a material get modified. Moreover, nanocomposites formed by this method are stable and long-lasting, since the nanoentities synthesised by this methodology are protected from the adjoining environment as they are formed few layers beneath the surface and become an integral part of the host material [14].

Silicon-oxygen tetrahedron is the basic building block of any type of silicate glass. The oxygens that are shared among two tetrahedral are called bridging oxygens (BOs) and the one which are not shared are termed as non-bridging oxygens (NBOs). In case of alkali glasses like soda lime glass, the alkali ions (Na^+, K^+) are added as network modifiers and they get linked to the NBOs [15,16]. When soda lime glass is subjected to ion-exchange, some of the sodium ions (Na^+) present in glass are replaced by silver ions (Ag^+). When such silver exchanged glasses are exposed to laser irradiation, thermal annealing then silver ions present in the glass are converted into neutral silver (Ag^0) which in due course migrate and aggregate to form silver nanoparticles [8,11,17,18].

Over the last two-three decades a lot of work had been done on metal glass nanocomposites synthesised by ion-exchange technique followed by ion implantation, however this work is mainly confined to their structural analysis and mere optical characterization like F. Caccavale et al. [18] studied the formation of silver nanoparticles in ion-exchanged soda-lime glass by irradiation using Secondary Ion Mass Spectrometry (SIMS), Rutherford Backscattering, Nuclear Reactions (NRA) and UV–visible absorption spectroscopy, D. Manikandan et al. [19] performed photoluminescence studies along with UV–visible absorption spectroscopy and Glancing Incidence X-Ray Diffraction of embedded copper nanoclustors in soda lime glass by ion-exchange method followed by irradiation with Helium ions, A K Sbouh et al. [21] studied radiation-enhanced diffusion of silver induced by Argon implantation into silver doped glass using Rutherford Backscattering Spectroscopy (RBS), but in-depth studies on their optical behavior has seldom been done. So the main objective of this paper is to present a comprehensive optical analysis of silver nanoparticles embedded soda lime glass nanocomposites synthesised by the dual-step methodology mentioned above as systematic and detailed studies based on the estimation of various optical parameters of the nanocomposites formed by this method still demand a thorough investigation of optical parameters like optical energy gap (E_g), Urbach's energy (E_u), refractive index (n), real (ε_1) and imaginary (ε_2) parts of dielectric function, skin depth, optical conductivity as these are of significant importance while designing devices for a variety of optical applications such as ultrafast optical fibre communication, waveguides, electromagnetic interference (EMI) shields, etc.

2. Experimental

2.1. Materials

In the present work, analytical reagent grade chemicals were used. Commercially available soda lime glass slides of dimensions (75 mm × 25 mm × 1.3 mm) with composition (wt%) Si (35.87%), O (47.50%), Na (8.79%), Ca (4.64%), Mg (2.50%), Al (0.70%) (using energy dispersive analysis of X-rays (EDAX) spectroscopy) provided by Polar Industrial Corporation, Mumbai, India were used as substrates. The $AgNO_3$ (Mol. wt = 169.87 g mol^{-1}, 99.8% pure) and $NaNO_3$ (Mol. wt = 84.99 g mol^{-1}, 99% pure) were purchased from Rankem (New Delhi, India) and used as received.

2.2. Fabrication of nanocomposites

First of all, the glass slides were washed thoroughly with deionized water and cleaned with extra-pure acetone (99.5% purity) in order to remove surface impurities. Silver was introduced into the glass slides by ion-exchange technique in which the preheated glass slides were immersed for 30 s in a molten salt bath of $AgNO_3$ and $NaNO_3$ in a ratio of 1:4 (wt%) at 350 °C so that some of the Na^+ ions are replaced by the Ag^+ ions. Thus obtained Ag^+–Na^+ ion exchanged glass samples were further rinsed with distilled water to remove any excess $AgNO_3$ sticking to the surface. After ion exchange no change in color of the glass slide was observed (Fig. 1). In order to promote aggregation of silver to nanometeric dimension, the ion exchanged samples were implanted with 200 keV Ar^+ ions at normal incidence using fluence of 1×10^{15}, 1×10^{16}, 5×10^{16} and 1×10^{17} ions cm^{-2} at room temperature.

The Ag^+–Na^+ ion exchanged glass slide acquired a very light yellowish color when implanted with a dose of 1×10^{15} Ar^+ ions cm^{-2} that further changed to light brown for 5×10^{16} Ar^+ ions cm^{-2} and finally to dark brown with mirror formation when implanted with a dose of 1×10^{17} Ar^+ ions cm^{-2} (Fig. 1). Beam current was kept at 2.0 μA cm^{-2} for all the four doses. The projected range for 200 keV Ar^+ ions in soda lime glass is around 209.8 nm using SRIM calculations [22]. A schematic diagram of entire experimental procedure is shown in Fig. 1.

Microstructural information related to surface morphology, size and distribution of silver nanoparticles in soda lime glass matrix was obtained using Quanta 200F field-emission electron microscopy (FE-SEM) operated at an accelerating voltage of 20 kV along with energy dispersive analysis of X-rays (EDAX) spectroscopy for compositional information of thus formed silver glass nanocomposites. For FE-SEM measurements the samples were coated with an extremely thin layer of gold by using sputtering technique. While optical analysis of these nanocomposites was carried out with the help of UV–visible spectroscopy in all the three modes namely absorption, transmission and reflection using Shimadzu Double Beam Double Monochromator Spectrophotometer (UV-2550) equipped with an Integrating Sphere Assembly ISR-240A in the wavelength range of 190 nm to 900 nm with a resolution of 5 nm. The absorption and transmission spectra were recorded using air as the reference whereas for diffuse reflection spectra $BaSO_4$ powder was used as the reference material.

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