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Characterization of Chemical Vapor Deposited Tetraethyl Orthosilicate based SiO_2 Films for Photonic Devices

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Silicon has been the choice for photonics technology because of its cost, compatibility with mass production and availability. Silicon based photonic devices are very significant from commercial point of view and are much compatible with established technology. This paper deals with deposition and characterization of SiO_2 films prepared by indigenously developed chemical vapor deposition system. Ellipsometry study of prepared films showed an increase in refractive index and film thickness with the increment in deposition temperature. The deposition temperature has a significant role for stoichiometric SiO_2 films, FTIR measurement has shown the three characteristics peaks of Si-O-Si through three samples prepared at temperatures 700, 750 and 800 °C while Si-O-Si stretching peak positions were observed to be shifted to lower wavenumber in accordance to the temperature. FESEM analysis has confirmed the smooth surface without any crack or disorder while EDX analysis showed the corresponding peaks of compositional SiO_2 films.

Keywords: SiO_2 films, CVD, TEOS, refractive index.

1. INTRODUCTION

Silicon oxide (SiO_2) thin films have found wide applications such as gate spacers, etch stoppers, gate dielectrics, antireflection layers etc. [1, 2]. The basic building blocks required for optical interconnects are the source, modulator, waveguide and detector. The optical interconnects has the ability to transfer a large bandwidth signals over a long distances while minimizing power requirements outweighs the complications of integrating electro-optical technologies. The example of silicon photonic integrated circuits is the hybrid integration of active components and silica based planer lightwave circuits, which provides a means for photonic component integration within a chip [3]. In this integration, the passive components are realized by using silica waveguides while active components are hybridized within the silica. By using this approach, various photonic components have been integrated such as multiwavelength light source, optical wavelength selector, wavelength converters, all optical time division multiplexers etc. [4].

For thin deposition, chemical vapor deposition (CVD) is a well established method for several years. For the deposition, desired precursors after conversion into vapors are transported into the reactor onto the heated substrate where the active gas molecules are decomposed. Depending upon the choice of materials and process parameters the deposited films may be amorphous/polycrystalline/epitaxial in nature. Thin films made by CVD have got several applications such as coating, insulation, coating for wear resistant part etc. In-spite of other deposition methods, CVD method is a better one due to its high throughput, purity and inexpensive process. With optimization of process parameters during growth, the deposited film may be of good stoichiometric in nature. Organosilicon material such as tetraethyl orthosilicate (TEOS) has got a huge demand as a silicon source for CVD deposition which gives a better film quality and good step coverage as comparison to the other materials. Precursor TEOS is easy to handle, good chemically stable and safe [5, 6]. This precursor is available in liquid form and can be hydrolyzed into silicon dioxide in ambient moisture. To vaporize TEOS, one can use bubbler and heat it just above room temperature which enhances the partial pressure of TEOS. With the choice of TEOS precursor, it is important to note that silicon is already oxidized and therefore it is rearranged during deposition process whereas with silane it undergoes actual oxidation process. In this work, TEOS precursor has been used to deposit SiO_2 thin films at three different deposition temperatures and their optical and structural properties are presented.

2. EXPERIMENTAL DETAILS

A typical chemical vapor deposition system consists of following main components: quartz tube/reactor, single zone furnace, gas sources, mass flow controllers, bubbler to vaporize the precursor, gas feed lines, pressure gauge, rotary pump and temperature control unit. Fig. 1 shows a schematic of indigenously developed chemical vapor deposition system with Mass flow controllers MFC1, MFC2 and MFC3 are corresponding to the ammonia, oxygen and nitrogen gases respectively. The ON/OFF valves are named as V1 … V6 whereas NV1 and NV2 are needle valves. At the gas entrance of the quartz tube, a pressure gauge is connected which reads the pressure inside the tube sustained by vacuum pump connected to the outlet of quartz tube.

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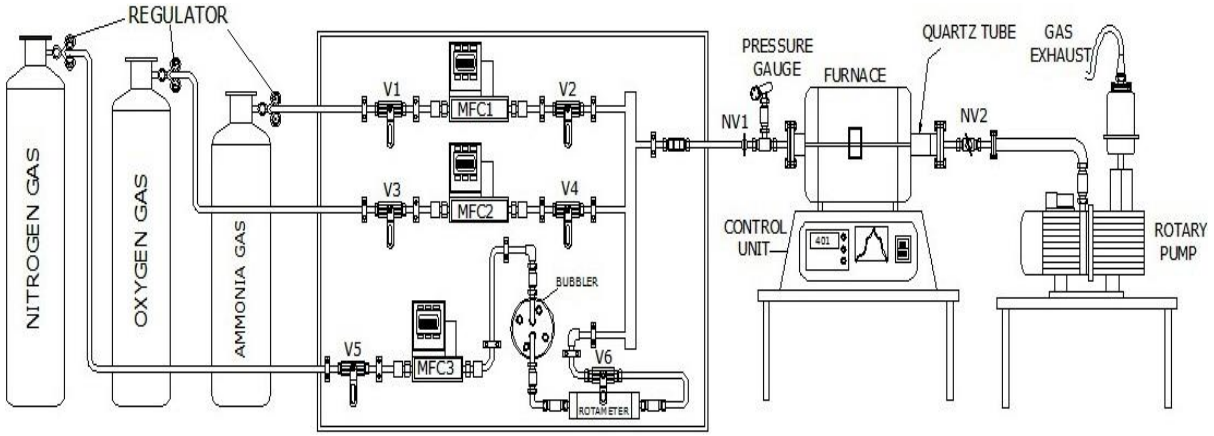


Fig. 1. Schematic of indigenously developed chemical vapor deposition system

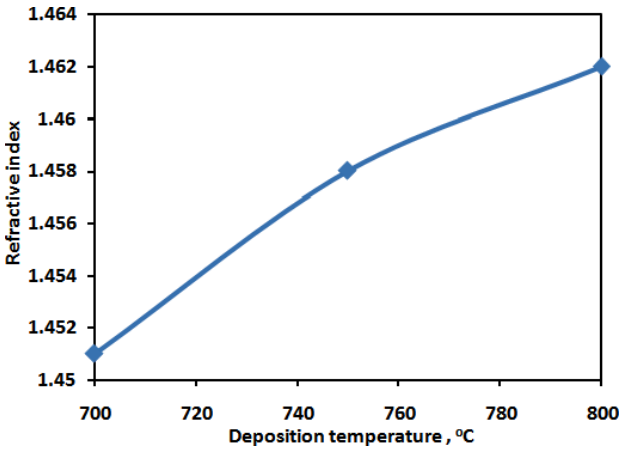
The length of quart tube is 450 mm and 43 mm is diameter. The maximum furnace temperature is 1200 °C which is programmable with 10 °C per minute ramping.

Before processing, the silicon substrates (P-type) were rinsed in de-ionized water after heating, separately in trichloroethylene, methanol and acetone for 10 min and dried in the presence of nitrogen. The reactor tube was purged with nitrogen gas to remove the contaminants. The reactant gases were introduced into the quartz tube through the common gas inlet made up of SS with OD ¼ inch. The mass flow controllers were fixed at 30 and 20 SSCM for oxygen and nitrogen gases respectively. The precursor tetraethyl orthosilicate was contained in stainless steel bubbler which was connected to the heater at 44 °C. Deposition of SiO_2 films was done in 12 min on three samples by varying deposition temperature from 700 – 800 °C while keeping other parameters constant. After deposition the films were characterized as deposited. The refractive index and film thickness were measured by Ellipsometer (PHILIPS-SD-1000) which used a laser of wavelength 632.8 nm as a source. To investigate the chemical components on the films, Fourier transform infrared measurement was done by FTIR (8400-Shimadzu). The surface morphology of deposited films was done by using field emission scanning electron microscope (FE-SEM: HITACHI S-4800) and films compositional analysis was done with EDAX (Bruker).

3. RESULTS AND DISCUSSION

Fig. 2 a shows the variation of refractive index of SiO_2 thin films as a function of deposition temperature. It is found that the refractive index increases as the deposition temperature increases from 700 – 800 °C. Actually, an increase in temperature causes the removal of excess elements from the film as a result increases the mass density which yields an increase in the refractive index of deposited films. Fig. 2 b shows the variation of SiO_2 film thickness in accordance with deposition temperature. It is clearly visible that the film thickness is increased as the temperature is increased. Deposition temperature is playing an important role and a small variation in gas phase reaction of different chemical species is responsible for the change in film thickness. To observe the chemical changes in the films deposited at different temperature, FTIR investigation was done and shown in Fig. 3.

FTIR spectra shows the three characteristic peaks of Si-O-Si of thermally grown films at various temperatures and found good matching with reported works [7 – 9]. With this we can say that the quantity of residual groups can be reduced by increasing deposition temperature [10].



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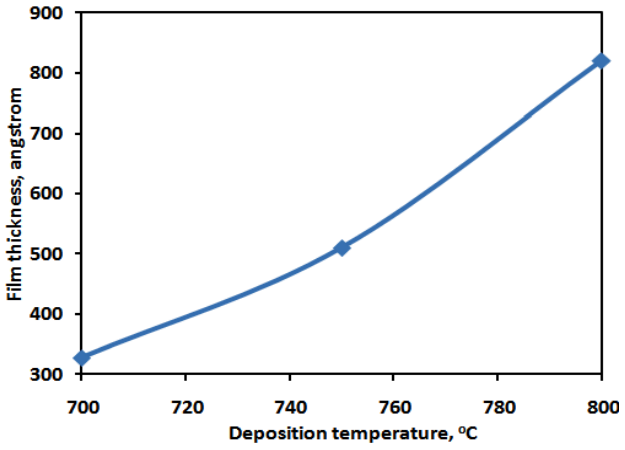


b

Fig. 2. a – effect of deposition temperature on refractive index; b – thickness of SiO_2 thin films prepared by APCVD

A low frequency peak observed at 461 cm^{-1} is due to the rocking mode which corresponds to the out of plane motion of oxygen atom.

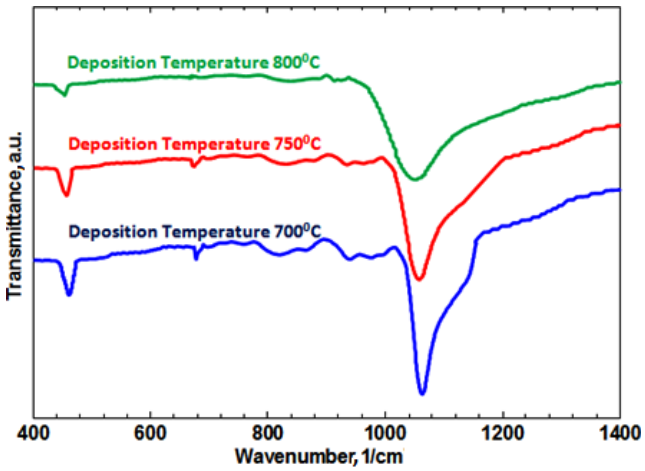
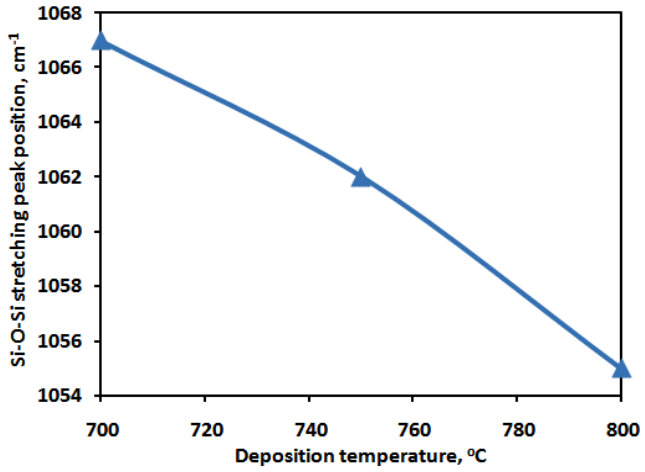


Fig. 3. FTIR transmittance spectra of SiO_2 thin films deposited at different temperature by APCVD



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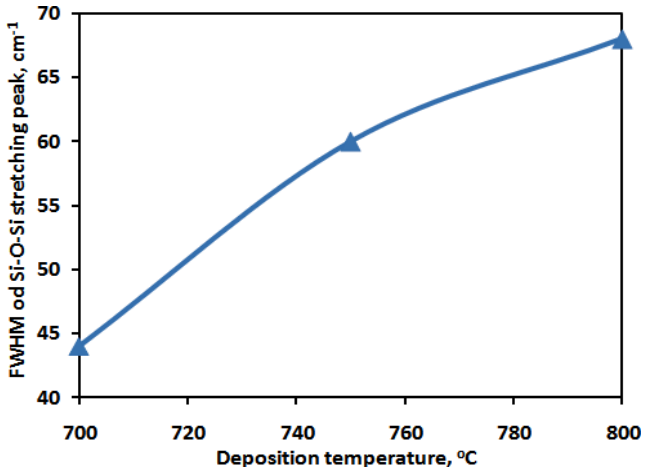

b

Fig. 4. a – variation in Si-O-Si stretching frequency; b – FWHM of Si-O-Si stretching frequency of SiO_2 thin films as a function of deposition temperature

However, weakest frequency peak at 812 cm^{-1} corresponds to the bonding vibration which involves the oxygen atom motion in the Si-O-Si plane and along the Si-O-Si angle bisector. In addition, a strongest vibration peak at 1067 cm^{-1} has been observed which is due to the asymmetric stretching vibration where oxygen atom motion influences in the Si-O-Si plane and parallel to a line joining the two silicon atoms. The stretching mode vibration peaks are found to be shifted at low wavenumber in accordance with an increase in deposition temperature from 700 – 800 °C. The position and shape of the main Si-O-Si vibrational bands shown in figure 3 indicates stoichiometric SiO_2 films.

To understand the shift in the Si-O-Si stretching peak as a function of deposition, we have plotted Fig. 4 a. It is found that as the deposition temperature increases the Si-O-Si stretching peak shifts to lower wavenumber from 1067 – 1055 cm^{-1}. This shifting in Si-O-Si stretching peaks have been assigned to the compositional changes in the films. We have also analyzed an important parameter i.e. full width at half maximum (FWHM) of Si-O-Si stretching peak which gives the information of structural order of prepared SiO_2 films. Fig. 4 b is plotted for FWHM of three silicon dioxide films deposited at temperature 700, 750 and 800 °C respectively. It can be seen clearly that FWHM of Si-O-Si stretching peaks are increased from 44 – 68 cm^{-1} in accordance with the variation in deposition temperature from 700 – 800 °C.

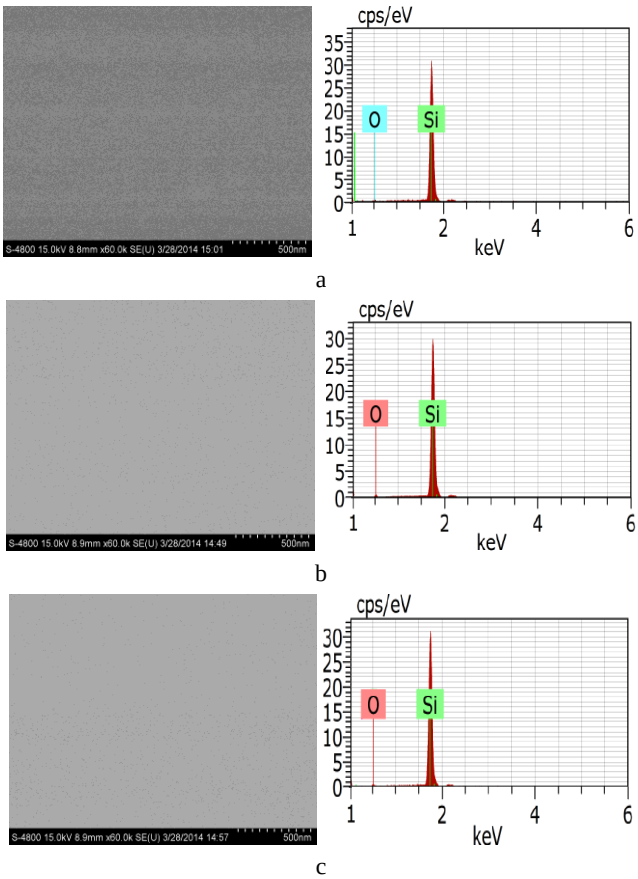

Fig. 5. FESEM and EDAX measurement of APCVD SiO_2 thin films deposited: a – at 700 °C; b – at 750 °C; c – at 800 °C

The surface morphology and chemical composition of deposited thin films were investigated by Field Emission Scanning electron microscopy (FESEM) and EDAX. From Fig. 5, left it is observed that the film deposited at 800 °C was smoother than deposited films at lower temperatures. All three samples of SiO_2 were uniform and found free from defect. EDAX images of SiO_2 films deposited at various temperatures are shown in Fig. 5, right. The elemental analysis showed the corresponding peaks of

oxygen and silicon while a small variation in oxygen and silicon count has been observed due to the variation in deposition temperature. The prepared thin films deposited by indigenously developed chemical vapor deposition system are found to be good in quality and has scope for photonic and optoelectronic applications such as for the fabrication of one-dimensional photonic crystals as low-dielectric constant material, as anti-reflection coating of solar cells, in waveguides etc.

4. CONCLUSIONS

An increase in refractive index and film thickness has been observed in accordance to the variation in deposition temperature. The higher deposition temperature causes the removal of excess elements from the film as a result increases the mass density which yields an increase in the refractive index. Similarly, a small variation in gas phase reaction of different chemical species is responsible for the change in film thickness. FTIR measurement has shown three characteristic peaks of Si-O-Si at 461, 812 and 1067 cm^{-1}. The position and shape of the main Si-O-Si vibrational bands indicates the stoichiometric SiO_2 films. Further, Si-O-Si stretching peak was found to be shifted to lower wavenumber with respect to an increase in deposition temperature which gives compositional variations in the deposited films. FWHM of Si-O-Si stretching peaks were observed to be increased from 44 – 68 cm^{-1} in accordance with the variation in deposition temperature from 700 – 800 °C. FESEM measurement has confirmed that the deposited films are free from defect and uniform while EDAX analysis has shown the elemental peaks of SiO_2 films. Deposited samples were found to be free from OH bond which confirms the suitability of these films for photonic devices. The prepared films can be used as low dielectric constant layer in the fabrication of one-dimensional photonic crystals with a combination of high dielectric constant layer.

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