International Journal for Research in Engineering Application & Management (IJREAM)
ISSN : 2454-9150 Vol-05, Issue-02, May 2019

Study of SiO_x insulating layer for integration with Si technology

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Abstract: Laser ablation procedure was utilized to deposit Silicon suboxide (SiO_x) thin films on c-Si and glass using a silicon monoxide (SiO) target of 99.99% purity. The ablation was carried out with the principal line of 1064 nm wavelength of a pulsed Q-switched Nd:YAG laser. The pulse energy was 10 mJ/cm^2 and was incident upon the target with a frequency of 10 Hz. The substrate temperature was varied from 300 K to 873 K. This yields transparent thin films. The optical, microstructural and electrical characteristics of the as deposited films were studied. X-ray induced Photo-electron Spectroscopy (XPS) was used to determine the composition of the thin films. The film deposited at 300 K had the maximum oxygen content. The oxygen content decreased with the increase in substrate temperature. The optical constants of the SiOx films were obtained from modified Kramer-Kronig model. An Al/SiO_x/p-Si metal-insulator-semiconductor (MIS) structure was fabricated and its capacitance-voltage characteristics was studied.

Keywords* — *c-v, electrical, laser ablation, MIS, optical, SiO_x

I. INTRODUCTION

Sub-stoichiometric silicon oxide, SiO_x ($x \leq 1.6$) thin film has in the recent years garnered much interest in the research community due to its immense application as passivation layer which helps stave the high recombination rate in c-Si solar cells, to reduce drastically the optical reflectance and use in battery technology [1-7]. It also commands repute for use as active layer in Si based microelectronics, especially in memory devices. SiOx based resistive switching memory devices has also come into fore due its simple fabrication process and excellent performance. Mehonic et al. [8] in a recent communication, demonstrated that the conductance of Si rich silica (SiOx) resistive switches is intrinsically quantized in half integer multiple of G_0, the fundamental unit of conductance. It has been further demonstrated that unlike other resistive switching systems, this quantization is not due to drift metallic ions. In CMOS processing, this diffusion of metallic ions is not desirable. This is what makes the intrinsic switching of SiO_x so much more appealing [9]. The commercial introduction of resistive switching memory elements, notably memristors could potentially solve the issue of charge control as the memory devices are scaled to lower physical dimensions. It could in future replace RAMs, flash drives and hard disks as the universal non-volatile memory. In this context we present here the microstructural, optical, compositional and electrical characteristics of SiO_x thin films deposited at various substrate temperatures using laser ablation technique to better understand the material characteristics for microelectronic device applications.

II. PROCEDURE

SiO_x thin films were deposited onto glass substrates and crystalline p-Si substrates with thickness of 200 nm by laser ablation process using a SiO target (Sigma Aldrich, 99.99% purity). The 1064 nm fundamental line of a Q-switched Nd:YAG laser with pulse duration of 10 ns and 10 Hz repetition rate (10 mJ/cm^2 pulse energy) was incident directly onto the SiO target, through a quartz window. It ablated the target material. When the laser pulse of Nd:YAG is incident upon the SiO target, it melts and evaporates the target material. The target material also ionizes and travels within the luminous plasma plume formed inside the chamber, which expands away from the target towards the substrate. Upon reaching the substrate, the deposition material contained within the plume condenses and forms a thin layer on top of the substrate. In principle, laser ablation is a very versatile technique for deposition of thin films with large degree of control over the film stoichiometry, thickness and uniformity. In our case, the target to substrate distance was 3 cm. The deposition was carried out for 15 minutes duration. Before the deposition, the stainless steel laser ablation chamber was evacuated to 10^{-6} mbar pressure. During the deposition, the background oxygen pressure was kept fixed. The substrate temperature was varied at 373K, 473K and 673K, to obtain films with different oxygen content.

DOI : 10.35291/2454-9150.2019.0016

The microstructure of the as deposited films was studied using scanning electron microscopy (SEM) and x-ray diffraction (XRD). The film composition was determined by x-ray photoelectron spectroscopy (XPS). The optical studies were performed by measuring the transmittance using an ultra-violet-visible-near-infrared spectrophotometer (UV-VIS-NIR) in the UV and visible spectrum range. The capacitance-voltage (C-V) measurement was performed at room temperature, by fabricating an Al/SiO_x/p-Si MIS structure. The Al top contacts (with an area of 1 mm^2 each) were deposited by evaporation technique using suitable masking. The capacitance value was measured using a capacitor divider circuit. The voltage signal from the known capacitor and the unknown capacitor (attached in series to the known capacitor) were measured using a dual channel lockin amplifier. The lockin amplifier was interfaced for automatic data acquisition.

III. RESULT

Figure 1 shows a representative SEM micrograph of a SiO_x film deposited at 373 K. It indicated good grain growth with compact surface. The films deposited at lower temperature were rougher. This could be due to lower coalescence of the crystallite at lower temperatures. The film deposited at 473 K showed a smoother surface, but, for films deposited above 473K, the films surface was rougher. This was due to desorption of oxygen from the film surface. For all the films, the larger grains could be secondary overgrowths or larger chunks of deposits sputtered from the ablation target. XRD measurement indicated that the films were completely amorphous in nature. The thickness of the SiO_x thin films were around 200 nm for all films, as determined using a profilometer.

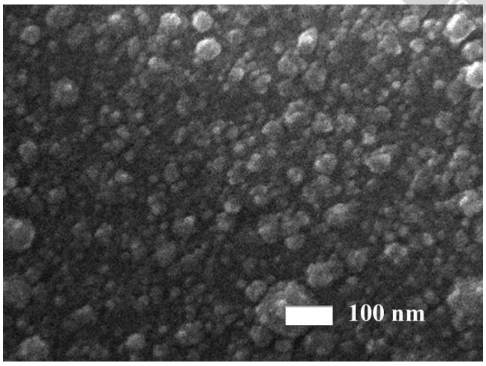

Fig. 1 Representative SEM micrograph of film deposited at 300 K

From the optical transmittance vs. wavelength plot (not shown here) for the SiO_x thin films it was observed that the transmittance was greater than 80 % through the whole visible range of the electromagnetic spectrum (400-800 nm). However, the transmittance decreased for the films deposited at higher temperature. By calculating the optical absorption coefficient (α), a plot of $(\alpha h\nu)^2$ vs. hν, extrapolated to α=0, gave the value of the band gap. The band gap of the films varied between 4.80 eV and 5.20 eV, with the band gap decreasing with the increase in substrate temperature. Apart from the band gap, the refractive index (n) and extinction coefficient (k) for the films were obtained using a modified Kramers-Kronig model [10]. From the computation, it was evident that the refractive index increased with the increase in deposition temperature, which could be due to the excess Si in the films deposited at higher temperature and due to the variation of defect concentrations within the films induced by the loss of oxygen content at higher temperatures. The extinction coefficient, k also increased with the increase in substrate temperature. The XPS spectra of representative films deposited at 373 K, indicated peaks for Si 2p at 102.7 eV and for O 1s shell at 532.4 eV. Although XPS peak of pure Si (x=0) occur at 99 eV, however, with the addition of O atoms, there is energy shift of the Si 2p shell, which rests at 103 eV for SiO_2(x=2) [11]. For any values of binding energy in between, the Si atom is resting at various oxidation states or their combination. In our case, the x value calculated was x=1.60 for the film at 373 K. With the increase of substrate temperature, x value was found to decrease. It was x= 1.52 for the films deposited at 473K and 1.36 for the films deposited at 673K. Our result was found to be consistent with those reported by Brodkorb *et al.* [12]. They also found that the films deposited at higher substrate temperature had higher Si content.

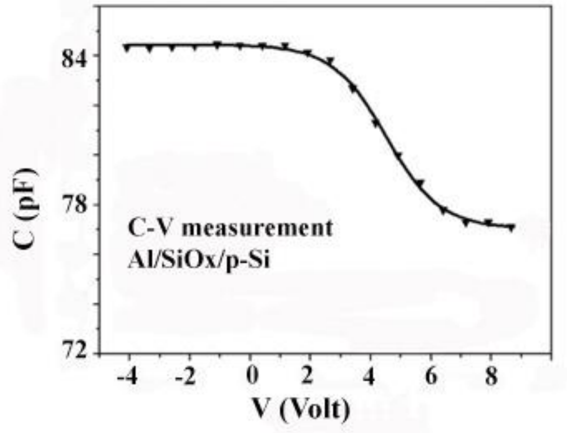

Fig. 2 C-V measurement for Al/SiO_x/p-Si structure

The capacitance-voltage measurement is an excellent tool to probe the applicability of an insulating layer for transistor fabrication. Fig. 2 shows the C-V characteristics of the Al/SiO_x/p-Si MIS with x= 1.6. From the C-V plot, the oxide capacitance was determined to be 84.4 pF. The maximum depletion width was calculated to be 87 nm. The computed flatband voltage was 3.72 V. From the flatband voltage, the effective oxide charge at the Si-$SiO_{1.6}$ interface was calculated to be 3.3 x 10^{11} cm^{-2}. The values obtained are quite promising but further refining of the material quality of the SiO_x film with lesser density of defects is required for applicability of the ablated silicon suboxide film for microelectronic transistor fabrication and possible integration with existing Si technology.

IV. CONCLUSION

Thin SiO_x films were deposited onto quartz and p-Si substrates by using laser ablation process using a pulsed Nd:YAG laser source operating at 1064 nm principle line, with 10 ns pulse duration, 10 mJ pulse energy and a repetition rate of 10 Hz, with an effective background pressure of 10^{-6} mbar. The deposition temperature was 373-673K. It was found that with the increase in substrate temperature during deposition, the oxygen content in the films decreased and the films became more sub-stoichiometric. For the film deposited at 373K, the x value was calculated to be, x=1.60 from XPS data. For the substrate temperature of 473K, x= 1.52 was obtained, and for the film deposited at 673K, the value of x was 1.36. From the optical measurements, the band gap of the SiO_x was found to vary between 4.80 eV to 5.20 eV, with band gap increasing with the decrease in substrate temperature. With the decrease in oxygen content in the films, they were more optically dense, with higher refractive index and extinction coefficient. C-V measurement on MIS structure made with the $SiO_{1.6}$ film deposited on p-Si substrate showed typical behaviour for p-type semiconductor with strong accumulation region followed by weak depletion and full depletion as one went from negative to positive d.c. bias. The oxide capacitance was 84.4 pF. The maximum depletion layer width was determined to be 87 nm. The flatband voltage was 3.72V. From the flatband voltage, the effective interface oxide charge was computed to be ~3.3 x 10^{11} cm^{-2}. The initial study indicates that after further refinement of the layer characteristics, especially the density of defects, these SiO_x layers could be possibly used for device application.

ACNOWLEDGEMENT

S.R.B. acknowledges with thanks the support provided by R.S. and R.A. at IST, UTL in carrying out this work.

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