Investigation of Silicon Nano-structured Layers Suitable for Realization of Light Emitting Diodes on Glass Substrate

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Abstract-Si nanocrystal layers with luminescent behavior on glass substrate have been prepared using a time multiplexed rf-plasma hydrogenation and reactive ion etching at temperatures below 350^{o}C. Various techniques such as Raman scattering, Photo-luminescent, field emission scanning electron microscopy and atomic force microscopy were used to investigate the nanocrystalline nature, luminescence behavior and structural features of the layers. Effects of crystallization plasma power and temperature have been also studied by photoluminescence analysis. This approach has been also utilized to realize multilayered light emitting diodes on glass substrates and electroluminescent examination has been used to further study the fabricated diode structures.

I. Introduction

Silicon is a certain leading material in microelectronics technology however its indirect band gap commonly makes it a poor optical material in optoelectronics. Since the first observation of photoluminescence behavior of porous Si in 1990 [1], intense research focused on porous Si however there remains only limited hope for active light emitting porous Si-based devices. Several strategies have been developed to turn Si into an efficient light-emitting material. The traditional approach is to reduce the Si dimensionality to nanometer scales, where both quantum confinement and surface chemistry effects improve the efficiency of light generation [2, 3].

In this line of research great deal of research on nc-Si has proposed them as (CMOS)-compatible, promising materials for optoelectronic applications. These nc-layers are generally applied in the form of nc-Si/SiO2 multilayers (MLs). These ML structures are usually prepared by molecular beam epitaxy, magnetron sputtering, epitaxial layer transfer technology and plasma enhanced chemical vapor deposition (PECVD). PECVD is a simple and convenient method for the deposition of Si and is completely compatible with Si integrated circuit(IC) technology. Thermal crystallization or laser crystallization of the deposited amorphous Si (a-Si):SiO2 layered structures is mostly involved in the reported PECVD-assisted approaches [4-9]. These techniques clearly involve much cost and complication to the fabrication process. Also vast technological limitation in IC fabrication is another drawback of these methods.

In this paper, we report the material study of the prepared structures as well as successful fabrication of light-emitting diodes on standard soda-lime glass substrates. The reported technique seems very promising for large-area applications due to its low cost, simplicity and reproducibility. The evolution of the nano-porous structures on glass substrate was investigated by means of photoluminescence (PL), field emission scanning electron microscopy (FE-SEM), Raman scattering and atomic force microscopy (AFM).

II. Experimental Details

Figure 1 shows the fabrication process of the nc-layer schematically. As shown in this figure, the deposited amorphous layer (figure 1.a) converts into a nano-crystalline layer by means of energetic hydrogenation and annealing steps (figure 1.b). By applying proper etching using a reactive ion etcher (RIE), the remaining amorphous layer is removed, leaving behind the nano-crystalline silicon grains (figure 1.c) [10].

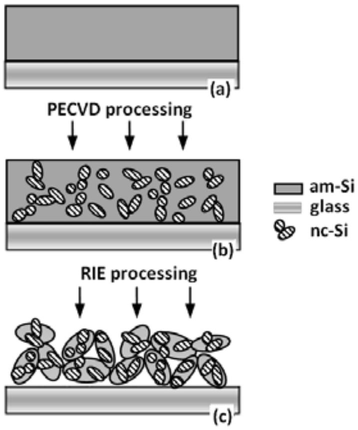

Figure 1: A flow diagram of the process for preparing a Si-nc layer with luminescent behavior. (a) A 100 nm thick am-Si layer is deposited on the cleaned soda-lime glass. (b) The hydrogenation/annealing treatment repeated four times to produce nano-crystalline grains embedded in am-Si. (c) Partial isolation of the grains by removing the amorphous regions between the nc-Si grains using an RIE process.

Then to intensify the luminescent intensity per unit area a vertical stack of nc-Si layers has been implemented as the intrinsic layer in a P-i-N structure which is shown schematically in figure 2. An ITO layer acts as the transparent bottom contact while nickel acts as the top contact in the fabricated LED's.

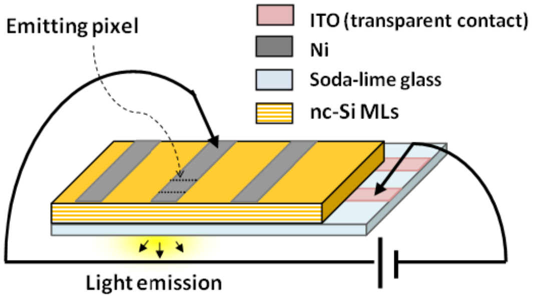

Figure 2: Schematic diagram of the multilayer nano-crystalline structure used to form a light-emitting diode. The diode was fabricated on a glass substrate. An ITO layer acts as transparent underlying contact and Nickel thin film as top electrical contact.

III. Result and Discussion

SEM images in figure 3.a to 3.c display the morphology of the nano-porous layers. Photoluminescence (PL) analysis indicates the light-emitting behavior of the processed layer on glass substrate, at a wavelength around 550 nm. These spectra show a direct dependency between the number of applied RIE sequences and the observed frequency shift (figure 3.d).

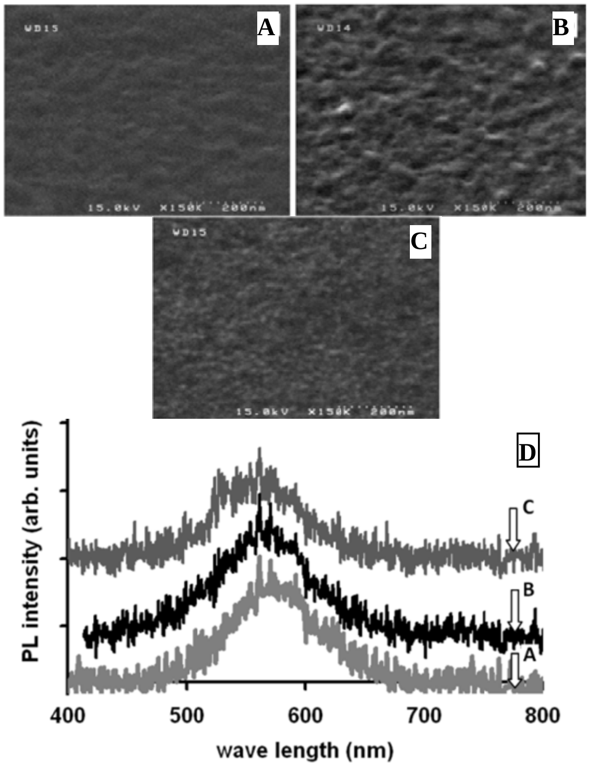

Figure 3: The evolution of isolated nc-Si grains by RIE process in samples (A) to (C) for 1, 2, and 3 RIE sequences respectively. In sample (C) grains well below 10 nm in diameter and fully isolated can be observed. (D) PL spectra from samples A, B and C corresponding to 1, 2 and 3 sequences of RIE process. As the number of etching sequences increased the peak in the spectra showed a blue shift. In addition, the grain size shows a reduction from A to C.

Figure 4 manifests the spectrum resulted from Raman scattering from the prepared nc-Si layer with a broad double peak at around 520 nm. The shape and energy of the achieved Raman spectrum is in accordance with the reported nano-porous Si layers [11, 12] where a small peak is expected at 520 cm^{-1}.

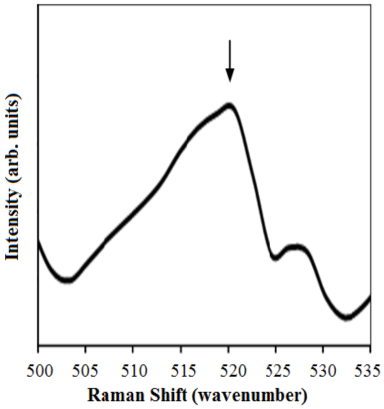

Figure 4: Raman scattering of the fabricated nano structured layer which shows a broad peak at the frequency of about 520 cm^{-1} which is the characteristic of the nc-silicon.

Top view AFM image of the surface of the sample prepared with 3 sequences of the RIE etching process is given in figure 5. Individual grains can be observed in the images where the average height of the grains was less than 6 nm from the histograms presented in part (b).

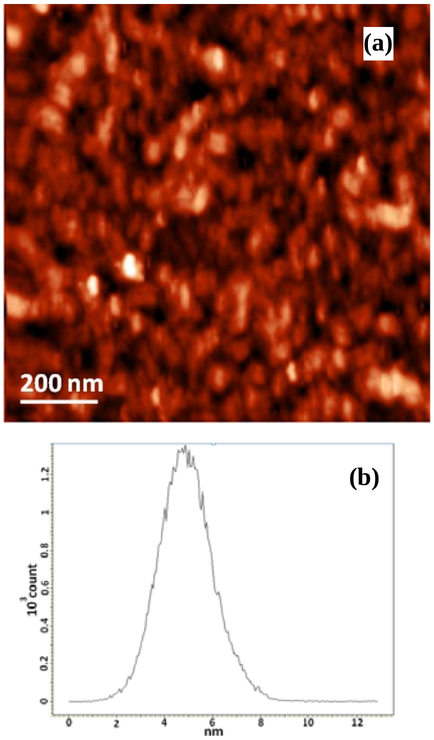

Figure 5: AFM images and analysis of a sample that has undergone 3 sequences of RIE etching. (a) Top view

AFM image showing isolated nano-clusters, each of which consists of multiple smaller pores. (b) A histogram of the surface roughness with mean Z value of 5 nm.

The effect of crystallization temperature on the Photoluminescent behavior and the morphology of the samples have been studied (figure 6.a) which showed no serious dependency on the processing temperature. This fact makes the application of this method to flexible substrates like PET-based LEDs feasible, which is being pursued.

A study on the plasma power effect of the crystallization step resulted in a shift in the Photoluminescent spectrum (figure 6.b). The higher applied power leads to weaker luminescent intensity with a red shift in the frequency which is believed to be related to the formation of larger crystalline grains in higher powers. Since the etch rate of crystalline grains are slower than the amorphous area, the grain size condition in the crystallization step is pursued after the RIE process. Porous nc-layer with larger diameter, exceeding the critical quantum size, leads to weaker luminescence behavior with lower energy spectrum.

Finally a multilayer structure based on the fabricated nc-layers has been applied to implement a light emitting device.

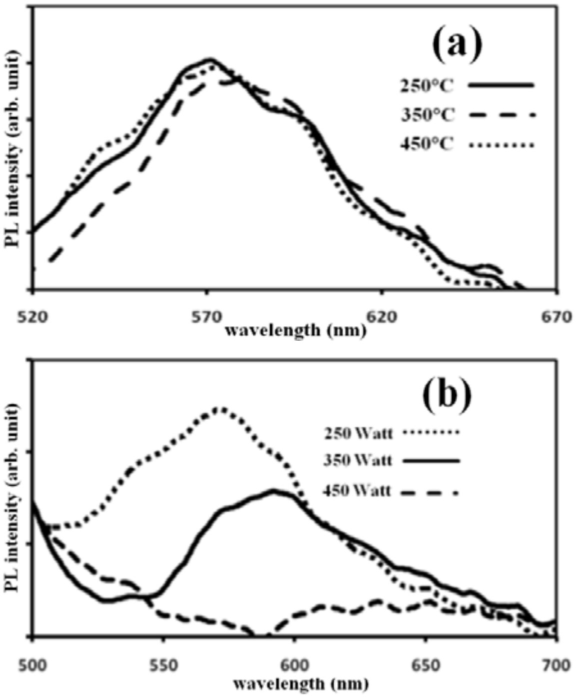

Figure 6: PL spectra from samples corresponding to different crystallization conditions: (a) crystallization temperatures of 250°C, 350°C and 450°C, (b) crystallization plasma power densities of 1.8, 2.5 and 3.2kW/m^2. As observed, temperature plays a less critical role in the luminescent performance whereas the plasma power is crucially important.

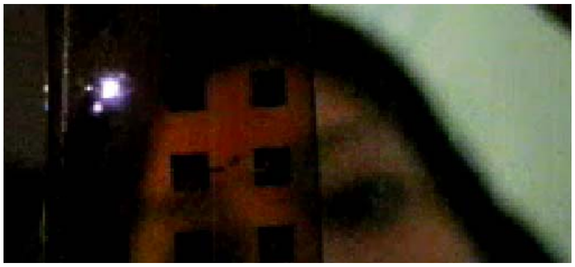

Figure 7: Optical image of light emitting pixels fabricated on soda-lime glass with one 3×3 mm^2 pixel (left), images from pixels recorded in a dark room (right).

The optical image in figure 7 displays the completely addressable glowing pixel with the dimension of 3×3mm^2 on soda lime glass. In addition, Electroluminescence spectrum of the device has been recorded which is presented in figure 8. It can be seen there is a broad peak at wavelengths around 550-600 nm in the EL spectrum which agrees with the data obtained from the PL measurements. This can imply on the same origin of the photoluminescence and the electroluminescence.

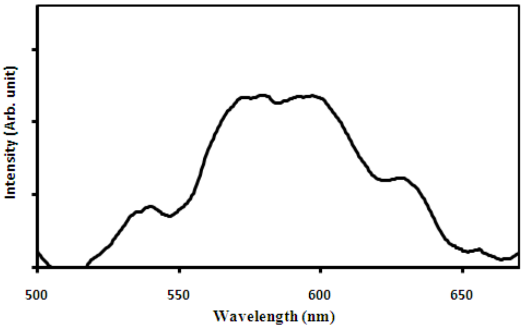

Figure 8: The EL spectrum of the emitting pixel, where a broad peak is observed around 550-600 nm, which is in accordance with previous PL measurements.

IV. Conclusion

A novel consecutive application of RIE and PECVD units led to the preparation of high quality photoluminescence multilayer structures directly on glass substrates. The process is suitable for the fabrication of Si-based light emitting devices on glass and can be extended to large area applications. In addition, this process can be implemented in a single machine, combining the plasma deposition and plasma etching, without any serious limitations on sample area. Low cost, simplicity and automated fabrication of multilayer structures are additional advantages making this process suitable industrial application.

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