

**LAYER-BY-LAYER PLASMA ENHANCED CHEMICAL
VAPOUR DEPOSITION OF NANOCRYSTALLINE SILICON
THIN FILMS**

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**FACULTY OF SCIENCE
UNIVERSITY OF MALAYA
KUALA LUMPUR**

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GOH BOON TONG

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ABSTRACT

This work is focused on the study of hydrogenated silicon (Si:H) thin films and nanostructures grown by layer-by-layer (LBL) deposition technique using a home-built radio-frequency (rf) plasma enhanced chemical vapour deposition (PECVD) system. The initial phase of this work involved preparation and characterization of hydrogenated silicon (Si:H) thin films by continuous (CD) and LBL deposition techniques on crystal silicon (c-Si) and glass substrates at different rf powers, substrate temperatures and hydrogen to silane flow-rate ratios. The effects of the deposition conditions on the optical and structural properties of the films are studied by optical transmission spectroscopy, Fourier transform infrared (FTIR) spectroscopy and X-Ray diffraction (XRD). The influence of substrates on these properties is also investigated. The second phase is focused on the study of the morphology, crystallinity, crystallite size, silicon-oxygen bonding and photoluminescence (PL) properties of the Si:H films grown on c-Si substrates by LBL deposition technique at the same deposition conditions in the first phase. These properties of the films are characterized by Micro-Raman scattering spectroscopy, field emission scanning electron microscopy (FESEM), high resolution transmission electron microscopy (HRTEM) and Micro-photoluminescence spectroscopy including further analysis done on the characterization results obtained from XRD and FTIR measurements done in the first phase in this work.

The results of this work demonstrated that rf power and substrate temperature produced significant changes to the optical and structural properties of the LBL films compared to the CD films. Increase in rf power increased the deposition rate of the LBL and vice-versa for the CD films. Also, increase in rf power increased the disorder of the CD films however suppressed the disorder in the LBL films. The preferred crystalline orientation was also changed from Si (311) to Si (111) plane with increase in rf power. Increase in substrate temperature increased the deposition rate, refractive index and structural order in the LBL films. The substrate temperature showed significant effects on optical band gap and hydrogen content in the LBL films. The LBL films deposited at substrate temperatures of 100 and 200°C showed large optical energy gaps suggesting that broadening of the band gap was due to quantum confinement effects. The LBL films deposited on c-Si substrates showed highly crystalline structure as compare to the other deposited films. The periodic hydrogen plasma treatment on the growth surface of the film during the LBL deposition processes showed effectively enhances the electro-optical properties of these LBL films.

The LBL deposition produced silicon nanostructures with Si nano-crystallites embedded in either amorphous silicon (a-Si) or mixed phases of a-Si and amorphous silicon oxide (a-SiO) matrix for the films deposited on c-Si substrates. These nanostructures of nanocrystalline silicon (nc-Si) grains produced high intensity of PL emission due to enhancement of quantum confinement effects by the presence of high crystalline volume fraction ($X_C \sim 41-54\%$) of Si nano-crystallites (~ 2 nm) in the matrix. The intensity of the PL emissions was strongly dependent on crystalline volume fraction, crystallite size and oxygen content in the a-SiO matrix. These parameters were significantly controlled by the rf power and substrate temperature. Based on these results, the growth kinetics and structural configuration of the LBL grown nc-Si grains were proposed. It was shown that high intensity of PL emission was emitted by these clusters of nc-Si grains.

ABSTRAK

Kerja penyelidikan ini menumpu kepada pengajian terhadap filem nipis silikon berhidrogen (Si:H) dan berstruktur nano yang dihasilkan dengan menggunakan teknik pemendapan lapisan-demi-lapisan (LBL) oleh sistem pemendapan wap kimia secara peningkatan plasma (PECVD) yang bina sendiri. Fasa awal penyelidikan ini melibatkan penyediaan dan pencirian filem nipis silikon berhidrogen yang dihasilkan menggunakan teknik pemendapan berterusan dan LBL di atas substrak hablur silikon dan kaca. Keadaan pemendapan yang digunakan untuk menghasilkan filem nipis ini adalah pembolehubah kuasa rf, suhu substrak dan nisbah kadar aliran hidrogen kepada silane. Kesan daripada pembolehubah keadaan pemendapan terhadap sifat-sifat optik dan struktur filem nipis ini dikaji melalui spektroskopi pemancaran optik, spektroskopi transformasi Fourier inframerah (FTIR) dan belauan sinar-X (XRD). Pengaruh substrak terhadap sifat-sifat tersebut juga dikaji. Fasa kedua menumpu kepada pengajian terhadap sifat-sifat morfologi, kehabluran, saiz hablur, pengikatan silikon-hidrogen dan luminasi foto (PL) untuk filem nipis silikon berhidrogen yang dihasilkan di atas substrak hablur silikon oleh teknik pemendapan LBL seperti fasa awal. Sifat-sifat tersebut dikaji oleh spektroskopi penyebaran mikro-Raman, mikroskopik pengimbasan elektron yang bermedan pancaran (FESEM), mikroskopik pemancaran elektron yang beresolusi tinggi (HRTEM) dan spektroskopi luminasi foto termasuk kajian terperinci atas keputusan yang diperolehi daripada XRD and FTIR dalam fasa awal.

Keputusan penyelidikan ini menunjukkan bahawa kuasa rf dan suhu substrak menghasilkan perubahan yang nyata keatas sifat-sifat optik dan struktur filem nipis LBL berbanding dengan filem nipis CD. Penambahan kuasa rf meningkatkan kadar pemendapan untuk filem nipis LBL dan sebaliknya untuk filem nipis CD. Penambahan kuasa rf juga meningkatkan struktur tak tertib dalam filem nipis CD namun ia menghalang struktur tak tertib dalam filem nipis LBL. Keutamaan orientasi untuk struktur hablur juga diubah daripada satah Si (311) kepada Si (111) apabila kuasa rf ditingkatkan. Penambahan suhu substrak meningkatkan kadar pemendapan, indeks biasan dan struktur tertib di dalam filem nipis LBL. Suhu substrak menunjukkan kesan nyata ke atas jurang tenaga optik dan kandungan hidrogen di dalam filem nipis LBL. Filem nipis LBL yang dimendapkan pada suhu substrak 100 dan 200°C mempunyai jurang tenaga optik yang tinggi. Kelebaran jurang tenaga optik ini adalah disebabkan oleh kesan pengurangan kuantum. Filem nipis LBL yang dimendapkan di atas substrak hablur silikon menunjukkan kehabluran yang tinggi berbanding dengan filem nipis yang lain. Rawatan hidrogen plasma secara berkala keatas permukaan pertumbuhan filem dalam masa process pemendapan LBL menunjukkan keberkesanan dalam mempertingkatkan sifat-sifat elektro-optik bagi filem nipis LBL.

Pemendapan LBL menghasilkan silikon berstruktur nano yang mengandungi silikon kristalit nano terbenam sama ada dalam amorfus silikon (a-Si) atau fasa bercampur di antara a-Si dan amorfus silikon oxide (a-SiO) matriks untuk filem nipis yang dimendapkan di atas substrak hablur silikon. Bijian silikon berhablur nano (nc-Si) ini menghasilkan pancaran PL dengan keamatan yang tinggi disebabkan peningkatan kesan pengurangan kuantum oleh sebab kehadiran kristalit nano (~ 2 nm) yang mempunyai pecahan isipadu hablur yang tinggi ($X_C \sim 41-54\%$) dalam matriks berkenaan. Keamatan untuk pancaran PL tersebut sangat bergantung kepada pecahan isipadu berhablur, saiz kristalit dan kandungan oxygen di dalam matriks a-SiO. Parameters ini dapat dikawal secara nyata oleh kuasa rf dan suhu substrak. Berdasarkan keputusan ini, kinetik pertumbuhan dan konfigurasi struktur untuk bijian nc-Si sudah dikemukakan. Ia menunjukkan bahawa pancaran PL yang tinggi adalah dihasilkan oleh bijian nc-Si.

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