STRUCTURAL AND OPTICAL PROPERTIES OF NANOCRYSTALLINE SILICON THIN FILMS GROWN BY 150MHz VHF-PECVD

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To my beloved husband, my parents and sister, for their love and support

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ABSTRACT

Nanocrystalline silicon thin film is a promising material potentially used in the optoelectronic field due to its improved and unique properties. In this work, nanocrystalline silicon thin films were grown by using a 150MHz VHF-PECVD to study the effect of deposition times, substrate temperatures and RF powers on their structural and optical properties. The thicknesses of the films were found to be in the range of 100 nm to 300 nm. Surface analysis from FESEM and AFM showed the existence of grain-like morphology which was later determined by EDX as silicon grains. The average grain diameter given by AFM analysis was around 50 nm. Surface roughness was found to be strongly dependent on the grain diameter where larger grain sizes showed a rougher surface. In average, surface rms roughness was 1.00 nm. Analysis from Raman showed that the films comprised of two phases, namely amorphous and nanocrystalline as revealed by a peak at 510 cm^{-1} with pronounced shoulder on lower frequency. The presence of nanocrystalline silicon was evident from the red-shift of peak frequency from those of pure crystalline silicon at 520 cm^{-1}. The average grain size as obtained from Raman was around 3 nm. Optical energy band gap, E_g^{opt} deduced from Tauc's plot and energy band gap, E_g obtained from PL were found to be higher than 1.12 eV within the range of 1.66 – 2.51 eV. All analysis showed that the properties of nc-Si were size dependent and followed the quantum confinement effect.

ABSTRAK

Filem tipis silikon nanokristal merupakan satu bahan yang menjanjikan potensi untuk digunakan dalam bidang optoelektronik disebabkan penambahbaikan dan keunikan sifatnya. Dalam kajian ini, filem tipis silikon nanokristal telah ditumbuhkan dengan menggunakan 150MHz VHF-PECVD untuk melihat kesan masa pertumbuhan, suhu substrat dan kuasa RF ke atas sifat struktur dan optik mereka. Ketebalan filem adalah dalam julat 100 nm hingga 300 nm. Analisis permukaan dari FESEM dan AFM menunjukkan kehadiran struktur seperti butiran yang kemudian ditentukan oleh EDX sebagai butiran silikon. Purata diameter butiran yang diberi oleh analisis AFM ialah sekitar 50 nm. Kekasaran permukaan didapati adalah sangat bergantung kepada diameter butiran di mana butiran yang lebih besar memberikan permukaan yang lebih kasar. Secara purata, kekasaran permukaaan ialah 1.00 nm. Analisis dari Raman menunjukkan filem terdiri daripada dua fasa, iaitu amorfus dan nanokristal seperti ditunjukkan melalui puncak pada 510 cm^{-1} dengan bahu pada frekuensi yang lebih rendah. Kehadiran silikon nanokristal adalah terbukti melalui peralihan-merah frekuensi puncak daripada puncak silikon kristal tulen pada 520 cm^{-1}. Purata saiz butiran yang diperolehi daripada Raman ialah di sekitar 3 nm. Tenaga jurang optik, E_g^{opt} seperti disimpulkan oleh plot *Tauc* dan tenaga jurang, E_g yang diperolehi daripada PL adalah lebih tinggi daripada 1.12 eV di dalam julat 1.66 – 2.51 eV. Semua analisis menunjukkan sifat silikon nanokristal adalah bergantung kepada saiz dan memenuhi kesan pengurungan kuantum.

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LIST OF ABBREVIATIONS / SYMBOLS

nc-Si	-	Nanocrystalline silicon
Si	-	Silicon
Ge	-	Germanium
SE	-	Spectroscopic ellipsometer
PL	-	Photoluminescence
EDX	-	Energy Dispersive X-ray Spectroscopy
AFM	-	Atomic Force Microscope
XRD	-	X-ray Diffraction
PECVD	-	Plasma Enhanced Chemical Vapour Deposition
FESEM	-	Field Emission Scanning Electron Microscopy
FTIR	-	Fourier Transform Infrared Spectroscopy
E_g	-	Energy gap
X_c	-	Crystalline volume fraction
I_a	-	Integrated intensity for amorphous phase
I_c	-	Integrated intensity for crystalline phase
I_m	-	Integrated intensity for grain boundary phase
δ	-	Average grain size
$\Delta\omega$	-	Peak shift for nanocrystalline silicon compared to the crystalline silicon
RF	-	Radio frequency
π	-	Pi = 3.1415926
SiH_4	-	Silane
Ar	-	Argon
RMS	-	Root mean square
at. %	-	Atomic percentage

Al_2O_3	-	Sapphire
O	-	Oxygen
H	-	Hydrogen
sym	-	symmetric
λ	-	Wavelength
eV	-	Electron volt
E_g^{opt}	-	Optical energy bandgap
α	-	Absorption coefficient
T	-	Transmittance
$h\nu$	-	Photon energy
uv	-	Ultraviolet
d	-	Spacing between atomic planes in crystalline phase
e-h	-	electron-hole
VB	-	valence band
CB	-	conduction band
$E_g(\infty)$	-	bulk bandgap
m_e, m_h	-	effective masses of electron and hole
ε	-	bulk optical dielectric constant or relative permittivity
α_B	-	Bohr exciton radius
ε_0, ε	-	permittivity of vacuum and relative permittivity of the semiconductor
μ	-	reduced mass of the electron and hole
e	-	electron charge
E_u	-	Urbach energy
k	-	wavenumber
d	-	thickness of film
n	-	index of refraction
k_e	-	extinction coefficient
c, v	-	velocity of light in vacuum and medium

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CHAPTER 1

INTRODUCTION

1.1 Background of Research

In 1959, Richard Phillips Feynman highlights a topic that has change the world as we know it today. His talk, “There’s Plenty of Room at the Bottom” has become the starting point on the manipulation and control of things at a small scale. Many decades after his inspiring talk, we are now able to manipulate and control materials so small, that we cannot even see it with naked eyes.

Nanomaterials, ranging from a few to a couple hundred nanometres in size, have offered a whole new perspective on the properties of material at atomic scale. It is of interests that as size of materials are comparable to that of exciton Bohr radius, its properties are different from their bulk due to the domination of quantum confinement effect.

Nanostructured materials can be produced in two ways; top-down and bottom-up approach. As the name implies, in top-down approach, large scale materials are made smaller and smaller until it reaches nanoscale while in bottom-up approach, nanostructure materials are build from the bottom: atom-by-atom, molecule-by-molecule, or cluster-by-cluster. Top-down approach gives a control of the manufacturing of smaller and more complex object. However, it has a disadvantage of possible imperfection of the surface structure that would have a significant impact on its properties. Meanwhile, the nature of self-assembly in bottom-up approach gives advantage of a more homogenous chemical composition with fewer defects to the surface morphology. This approach also made the formation and structure of material much easier (Cao, 2004). Techniques that have been developed for bottom-up approach includes sputtering, plasma-enhanced chemical vapor deposition (PECVD) and ion implantation.

There are three forms of nanostructure, quantum well, quantum wire and quantum dots (also referred to as nanocrystals or nanoparticles). The classification of the nanostructures is based on the dimensions that are reduced. If there is only one dimension being reduced to nanoscale, it is called quantum well. If two dimensions are on nanoscale, it is called quantum wire while quantum dot is referred to materials that are made nanoscale in all three dimensions.

Silicon (Si) is the principal component in most semiconductor devices. Its unique properties allow Si to remain a semiconductor at a higher temperature compared to germanium. Si also has the ability to form a native oxide layer to create a better semiconductor/dielectric interface than any other material. However, Si is also known as a poor light emitter, a result of its indirect energy gap. When a report is made on visible photoluminescence (PL) from porous silicon in 1990, it has triggered the hope of developing Si as a component in the optoelectronic devices (Jutzi and Schubert, 2003; Kumar, 2008). Reports on the shifting of photoluminescence peak to a higher energy as silicon is reduced to nanoscale has further increase the interest in nanostructure silicon.

Nanocrystalline silicon (nc-Si) has been a widely studied structure because of the discoveries that its properties can be tuned just by controlling the size of the crystals. There are also the advantages of enhanced carrier mobility, very low light-induced degradation and light absorption in infrared region of solar spectrum which further increase the possibility of using it as a leading material in optoelectronic devices (Chowdhury *et al.*, 2008).

With all these new, improved and unique properties reported as being possibly owned by nanostructure Si, more and more studies are need to be conducted in order to better understand the wonder of this material. The understandings of its structural and optical properties are important for future research activities.

1.2 Problem Statement

Recently, nanoscale crystalline materials have been the most talked about material especially in the field of optoelectronics. It triggered such interest because of the emergence of new, unique an interesting properties compared to those of its bulk material. nc-Si in different sizes, shape and quality are produced through different physical vapour deposition (PVD) and chemical vapour deposition (CVD) growth techniques. Among the techniques, plasma enhanced CVD (PECVD) has been the one widely used due to its ability to grow high quality nc-Si at high deposition rate and at lower cost. A conventional PECVD operates at plasma excitation frequency of 13.56 MHz. Over the decades, a variety of plasma excitation frequencies have been developed to increase the quality of nc-Si films grown by PECVD. However, most of them are in the medium high frequency range (MHF) such as 50MHz, 54.24MHz and 60MHz. The idea of using a very high frequency (VHF) PECVD to grow nc-Si films has not captured the attention of many researchers so far despite the piling reports on better quality of the films obtained by increasing the plasma excitation frequency. Therefore, this research seek to gain an insight on the growth of nc-Si films by PECVD at the highest plasma excitation frequency ever reported, which is 150MHz. The growth process is then controlled by varying three specified growth parameters which are deposition time, substrate temperature and RF power to see their effect on its structural and optical properties.

1.3 Objectives of the Research

(i) To grow nanocrystalline silicon (nc-Si) using 150MHz Plasma Enhanced Chemical Vapor Deposition (PECVD)

(ii) To characterize the structural properties (surface morphology, crystallinity, grain size) of nc-Si under different growth parameters (deposition time, substrate temperature, RF power)

(iii) To characterize the optical properties (absorption coefficient, energy band gap) of nc-Si under different growth paramaters (deposition time, substrate temperature, RF power)

1.4 Scope of the Research

This research focused on the characterization of nc-Si grown by 150 MHz PECVD technique developed at Ibnu Sina Institute, Universiti Teknologi Malaysia, Skudai. Substrate temperature, plasma power and deposition time are being varied to see the effect it has on the structural and optical properties of nc-Si. The following setting parameters has been identified: Substrate temperatures vary from 100°C to 400°C, deposition times between 5 to 20 minutes and RF power ranging from 12-24W. Samples of nc-Si thin films were fabricated on three different substrates for specific characterization measurements; corning glass 7059 for structural studies, quartz for optical studies and, crystalline Si wafer for Fourier Transform Infrared (FTIR) studies.

Film thickness is obtained by using spectroscopic ellipsometer (SE). Field Emission Scanning Electron Microscopy (FESEM) and Atomic Force Microscopy (AFM) were then used to probe the surface morphology of nc-Si thin films. The elemental composition is characterized by Energy Dispersive X-ray (EDX) and Fourier Transform Infrared spectroscopy (FTIR). Photoluminescence spectroscopy (PL) and UV/Vis spectrophotometer were used to obtain the information on absorption coefficient and energy gap of the nc-Si thin films.

1.5 Significance of the Research

Before 1990, less attention were given to Si as a promising material in the field of optoelectronic. Only after the Canham report on visible photoluminescence of porous-Si did scientist starts to have interest in this particular material. With the emerging of nanoscience, there have been more reports on Si at nanoscale as having interesting properties which starts to make it the centre of attention. Among many Si nanostructures, intensive studies on nc-Si has lead to promising potential in optoelectronic applications such as in thin film solar cells, thin film transistors and single electron transistors (Tamir and Berger, 2000; Shen *et al*., 2003; Ali, 2006; Chowdhury *et al*., 2008; Dalal *et al*., 2008). The properties of nc-Si thin films are strongly affected by its growth techniques and growth parameters. Research on the structural and optical properties can enhance the understanding of the relation between growth parameters and properties of nc-Si thin films. Furthermore, the application of an improved PECVD method in nc-Si thin films growth is expected to give interesting results on the surface morphology, grain size distribution and energy band gap as compared to previous PECVD method. Results analyzed from this study can also be used in further research to determine the optimum parameters needed to grow high quality nc-Si thin films.

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