Lanthanum Aluminate Silicon Based Photovoltaic Cell Dependency on Temperature Aye Aye Thant^{*}, ChawLoon Thu^{**}

Abstract

The materials used in this work were regent grade of lanthanum chloride (LaCl₃.7H₂O) and aluminium chloride (AlCl₃.6H₂O). The lanthanum aluninate (La_{1-x}Al_xO₃: x = 0.02 mol) clear solution was firstly performed by adding appropriate amount of (NH₄OH) into the mixture solution. Lanthanum aluminate sol-gel was refluxed with oil–bath at 110°C for 3hr. Lanthanum aluminate (LAO2) thin film was formed on defect-free p-Si (100) substrate by spin coating technique. To change the coating layer into oxide film, they were sintered at 400°C to 600°C in open atmosphere. X-ray Diffraction Analysis (XRD) and Scanning Electron Microscopy (SEM) measurement were made to examine the film morphology and microstructural properties. As a result of XRD investigation, the observed peaks were matched with those of standard library file. Film thickness was obtained by cross-sectional SEM images. $1/C^2$ -V characteristics were also observed for film qualification. I-V characteristics were measured under illumination by sodium lamp. The best power conversion efficiency of 10.65% photovoltaic cell with the LAO2 layer annealed at 500 °C showed a short-circuit current (I_{sc}) of 1.22 x 10⁻⁵ A, an open-circuit voltage (V_{oc}) of 3.19V and a fill factor (F_f) of 0.39 under illumination by sodium lamp.

Introduction

Photovoltaic (PV) cells are devices that convert sunlight to electricity. PV stands for photo (light) and voltaic (electricity), where photons free electrons from common silicon by sunlight. Photovoltaic (PV) cells are also known as solar cells. A photovoltaic cells is made of thin wafers of slightly different types of silicon. One type of silicon doped with quantities of Boron, is called p-type (p for positive) and it contains positively charged "hole", which are missing electrons. (Electrons are negatively charged particles that orbit the nuclei of atoms. The other type of silicon is doped with small amounts of phosphorus and is called n-type (n for negative).

It contains extra electron. Putting these two thin p and n materials together with electrons constitutes an electric current. A basis solar cell consisting of n-type and p-type semiconductor material forms a pn- junction. The crystalline silicon is often referred to as a junction which, when exposed to light, will produce a movement generation photovoltaic technology, while the second generation photovoltaics consists of thin film solar cell materials such as amorphous silicon (a-Si), cadmium telluride (CdTe), copper indium gallium diselenide (CIGS), and thin film crystalline silicon. The driving force for the development of thin film solar cells has been their potential for the reduction of manufacturing costs. While silicon solar panels are assembled from individual cells processed from about 100 cm² silicon wafers, thin film semiconductor materials can be deposited onto large surfaces, which is beneficial to volume production [1-6]. Lanthanum aluminate single crystal has a dielectric constant of 24–25, a large bandgap of 5eV, and good thermal stability up to 2100°C [7,8]. The low dielectric constant value of lanthanum aluminate thin film can be mainly attributed to the poor interfacial characteristics between lanthanum aluminate and Si substrate [9].

Experimental Details

The raw materials of lanthanum chloride and aluminium chloride were chosen as starting materials. The purity of materials were found to be 99.99%. Firstly, (1-x) LaCl₃.7H₂O and (x) AlCl₃.6H₂O were mixed according to the stoichometric composition of x=0.02 mol

^{*} Assistant Lecture, Department of Physics, Yadanabon University, Mandalay, Myanmar

^{**} Demostrator, Department of Physics, Yadanabon University, Mandalay, Myanmar

and stirred with glass rod for 1 hr, then homogeneous mixture was obtained. After that four drops of NH_4OH and 0.1 ml of distilled water were added to homogenous mixture and stirred with glass rod. Then, wet mixture was obtained. This mixture was left for 48 hr and heated with oil bath 110°C for 3 hr. After heating LAO2 (LaAlO₃) sol-gel was formed. The viscosity of the resulting sol-gel was measured by viscometer. After obtained sol-gel ultrasonification and centrifugal washing was repeated 5-6 times for each bath of precipitated sol-gel. After this sol-gel was dried in an oven at 100° C for 30 min. After cooling down at room temperature LAO2 precursor sol-gel was obtained. The viscosity of this LAO2 precursor sol-gel next time was measured.

The substrate used in this study was p-Si (100). The p-Si (100) wafer of dimension (1cm x 1cm) and thickness of $280 \sim 300 \mu m$ were used as substrate. Before film fabrication, p-Si (100) substrate was cleaned by using ultrasonic cleaning method. The Si cleaning sequences were as follow:

- 1. The silicon wafers were washed ultrasonically in distilled water.
- 2. They were washed in boiling acetone and then boiled in propanol for 5 minutes to remove greasy films.
- 3. They were immersed in nitric acid HNO_3 for 5 minutes in order to remove ionic contamination.
- 4. They were etched in buffered hydrofluoric acid (34.6% NH₄F: 6.8% HF: 58.6 % H₂O) for 5 minutes to remove oxide films.
- 5. Then the silicon wafers were cleaned in distilled water and dried on flat oven at 100°C in open air for a few minutes.
- 6. Finally, the cleaned Si-wafer was obtained.

The clear precursor sol-gel was poured onto cleaned p-Si (100) substrate which was placed on substrate holder of spinner. The spinning speed was 800 rpm. The substrate temperature was 120° C and the spinning time was 5 min. After fabrication, LAO2 films were annealed at 400° C, 500° C, 600° C respectively for 1h. Finally, LAO2 thin film was obtained.

To get the front exposed area (0.2 cm x 0.2cm) on LAO2 film, remaining regions were wrapped in a piece of tape. Back size of Si-substrate was also masked with a piece of tape and back exposed area (0.4cm x 0.4 cm) was appeared. The packed sample was immersed in Nickel solution and electroless Ni-plating was performed for 5 min and dried at room temperature. After removing the mask, front and back nickel conductive layers were observed. And then, Cu wire were soldered on the Nickel conductive layer Ni/LAO2/Ni with Cu electrode cell was obtained.

Characterization

Structural and microstructural properties were made on LAO2 thin films by XRD and SEM technique. X- ray diffraction was used for crystal phase identification. X- ray diffraction patterns were obtained with a Bruker D8 Advance diffractometer using Cu-K α radiation at 40 kV and 30 mA at a scan speed of 0.02s with an increment of 0.02° per step. The surface morphology and grain morphology of LAO2 thin films were investigated by using scanning electron microscope (JEOL, Model No. JSM- 5610 LV) with acceleration voltage 15 kV and magnification 5000. The grain sizes of LAO2 thin films were observed from the SEM images. The film thickness was also studied by using SEM. Film quality of LAO2 film was checked by $1/C^2$ -V characteristics. The capacitance values were measured by Quad Tech LCR meter model 1730. I-V characteristics of fabricated cells were examined. Cu was used as top to bottom electrodes. Measurements were performed at room temperature. Current-Voltage characteristics under illumination conditions were measured. Maximum photovoltage (V_m) and maximum photocurrent (I_m) of fabricated LAO2:Si cells were investigated, by using the DT-830 B DIGITAL MULTIMETER, sodium lamp LCH580-X (220V & 500W) as a

monochromatic light source and digital light meter model LX-108, to count the monochromatic light intensity. Short-circuit current (I_{sc}), open-circuit voltage (V_{oc}) (measurement at dark condition 0, series resistance (R_s), fill factor (FF), conversion efficiency (η_{con}) and quantum yield (Y) of fabricated cells were studied.

Results and Discussion

The LAO2:Si film was examined by XRD technique and shown in Figure 1 (a-c). There were 8 reflections on XRD Pattern of sample at 400°C. It was formed that six of all reflections were well consistent with the standard peak. Among the fabricated specimen at 500°C, only four peaks were matched with the standard. Amongs the sample at 600°C, six reflections were matched.



Figure 1. XRD profile of LAO2:Si thin film device at (a) 400° C (b) 500° C (c) 600° C

The resulting SEM image of LAO2:Si thin films were shown in Figure 2 (a-c). The surfaces were seemed to be crack-free and uniform grain distribution. The density of SEM image looked fairly low. The images were found to be rough and particles were oriented toward left-side. Some pores and grain-growth patterns were also observed on these images. The minimum grain size was 1.71 μ m at 400°C. Film thicknesses were measured by cross-sectional SEM micrographs.

(a)



(b)





Figure 2. SEM image of LAO2:Si thin film device at (a) 400° C (b) 500° C (c) 600° C

 $1/C^{2-}$ V characteristics of LAO2:Si thin films were examined by using Cu-electrode and Quad Tech LCR Digibridge meter model 1730. $1/C^{2-}$ V graphs measured at a high frequency of 1 kHz were studied and recorded as in Figure 3 (a-c). The C⁻² value increased linearly with the reverse bias voltages for all fabricated films. However, the slope and maximum value of C⁻² were different from all films. The built-in voltage (V_{bi}) was estimated to be 0.70 V, 0.82 V and 0.55 V for the film at 400°C, 500°C and 600°C respectively. The acceptor dopant concentration (N_a), donar dopant concentration (N_d) and effective dopant concentration (N_I) were determined. The depletion layer width (W) and barrier height (ϕ) were calculated. These film performance parameters were described in Table 1.

	Process Temperature			
Parameters	400°C	500°C	600°C	
Slope (F^2/V)	5.80×10^{21}	9.73×10^{21}	3.13×10^{21}	
$(C^{-2})_{max}(F^{-2})$	4.80×10^{22}	6.05×10^{22}	4.90×10^{22}	
$V_{bi}(V)$	0.70	0.82	0.55	
N_a (cm ⁻³)	2.05×10^{17}	1.22×10^{17}	3.80×10^{17}	
N_d (cm ⁻³)	7.15×10^{14}	9.86x10 ¹⁶	1.02×10^{12}	
N_{I} (cm ⁻³)	7.13×10^{14}	5.46×10^{16}	1.02×10^{12}	
W (cm)	2.29×10^{-5}	2.58×10^{-5}	2.32×10^{-5}	
φ(eV)	0.93	1.06	0.76	

Table 1. Slope, $(C^{-2})_{max}$, V_{bi} , N_a , N_d , N_I , W and ϕ for LAO2: Si thin films





Figure 3. $1/C^2$ -V characteristic curve of LAO2:Si thin film device at (a) 400°C (b) 500°C (c) 600°C

The current-voltage characteristics of LAO2:Si films under illumination were represented in Figure 4. According to the figure, it was found that the current starting from third quadrant increased gradually through the fourth quadrant and finally reached to first quadrant of the circle. According to this graph, it was known that the voltage changed from fourth quadrant to first quadrant was nearly to 3V for all films. As the detail analysis of short circuit current (I_{sc}) and open circuit voltage (V_{oc}) for LAO2 films processed at 400°C, 500°C and 600°C, it was clearly obvious that the voltages changed from fourth quadrant to first quadrant were 2.66V, 3.19 V and 2.66 V. According to the figure, maximum short-circuit current was found at 500°C.

The fill factor (FF) of LAO2:Si films were observed and they were 0.351, 0.389 and 0.355 at 400 °C, 500 °C and 600 °C respectively. The fill-factor, the variation of quantum yield (Y) and the photovoltaic parameters for LAO2: Si films with different process temperature were collected in Table 2.



Figure 4. I-V characteristics of LAO2:Si thin film devices at various process temperatures under illumination

Parameters	Process Temperature			
	400 °C	500 °C	600 °C	
$I_{m}(A)$	0.866 x 10 ⁻⁶	1.45 x 10 ⁻⁶	1.02 x 10 ⁻⁶	
$V_{m}(A)$	1.2754	1.8956	1.2684	
$P_{m}(W)$	1.1045 x 10 ⁻⁶	2.7490 x 10 ⁻⁶	1.2938 x 10 ⁻⁶	
I _{sc} (A)	1.35 x 10 ⁻⁶	1.22 x 10 ⁻⁵	1.62 x 10 ⁻⁶	
$V_{oc}(V)$	2.661	3.193	2.657	
η_{con} (%)	4.2903	10.6508	5.0144	
F _f	0.3508	0.3893	0.3546	
Y	6.00×10^3	11.20×10^3	6.95×10^3	
R _s	1978	8195	1710	

Table 2. The photovoltaic parameters for LAO2:Si films with different process temperatures

Conclusions

From the XRD analyses of lanthanum aluminate samples, there were 8 reflections on XRD Pattern of sample at 400°C. Six of all reflections were well consistent with the standard peak. As the fabricated specimen at 500°C, only four peaks were matched with that standard. The six reflections were matched as the sample at 600°C. The minimum crystallite size was 48.6nm with processing temperature at 400°C.

The SEM surface was seemed to be crack-free and of uniform grain distribution. The density of SEM image looked fairly low. The images were found to be rough and particles were oriented toward left-side. Some pores and grain-growth patterns were also observed on these images. All microstructures were examined to be temperature dependent. The film thicknesses were 25.3 μ m, 35.1 μ m and 36.0 μ m respectively. Minimum film thickness of LAO2:Si films appeared at 400°C.

The C⁻² value increased linearly with the reverse bias voltages for all fabricated films but the slope and maximum values were different. The built-in voltage was estimated to be 0.70V, 0.82V and 0.55V for the film at 400°C, 500°C and 600°C, respectively. From illumination, it was found that observed the voltage changed from fourth quadrant to first quadrant was nearly to 3V for all films. According to the detail analysis of short circuit current (I_{sc}) and open circuit voltage (Voc) for LAO2 films processed at 400°C, 500°C and 600°C, it was obvious that the voltages changed from fourth quadrant to first quadrant were 2.66V, 3.19 V and 2.66V. It was found the laboratory-fabricated LAO sol solution was actually formed and can be applied as a promising candidate for PV cell application.

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