PREPARATION OF HIGH QUALITY POLYCRYSTALLINE SILICON THIN FILMS BY ALUMINUM INDUCED CRYSTALLIZATION

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ABSTRACT: *In this paper, high-quality polycrystalline silicon (poly-Si) thin films on glass substrates are formed by Aluminum-induced crystallization (AIC). In AIC processes, bi-layer structures of amorphous silicon (a-Si) / Al are transformed into ones of (Al+ residual Si)/ poly-Si after simply annealing at 500°C in vacuum furnace. After Al chemical etchings, it isobserved that the obtained structures are poly-Si thinfilms on glasses with some amount of residual Si as" islands"scattered on theirsurfaces. The number of these "Si islands" remarkedly reduced by choosing an appropriate thickness ratio of pre-annealled Al and Si layers that prepared by magnetron dc sputtering. In this study, at initial Al/a-Si thickness ratio of 110/230 nm, the high-quality poly-Si thin films are formed with very few"Si islands" on the surfaces after AIC processes. Theobtained smooth surfaces are not appearing "dendritic" in optical transmission microscopy (OTM) images, have large grain size of tens of nanometers in SEM images and have average surface roughness of about 2.8 nm in AFM images. In addition, XRD Θ -2Θ measurements show a strong Si (111) peak at the 2Θ angle of 28.5°, presenting good crystalline phases. The films also reveal good p-type electrical conductivityin that their high carrier concentration and mobility in Hall effect measurements are* 10^{18} cm^{-3} *and 48* cm^2/Vs*, respectively.*

Keywords: *Aluminum-induced crystallization, polycrystalline silicon thin film.*

1.INTRODUCTION

Polycrystalline silicon thin films on low-cost substrates prepared by aluminum-induced crystallization (AIC) technique are of great interest for electronic devices, such as solar cells and thin-film transistors. Crystallized Si films can be formed on foreign substrates using AIC at temperatures below the eutectic temperature in Si –Al phase diagram. It is based on the overall layer exchange between adjacent Si and Al films during annealing process, resulting in the transformation from amorphous topolycrystalline Si phases. The advantages of the AIC technique are: a low-temperature process (< 577°C, the eutectic temperature), large and homogenous silicon grains and p+ type doping (Al) of the resulting crystalline silicon layer. However, the obtained poly-Si thin films by AIC often contain "Si islands" on the surfaces [1]. These "Si islands" are attributed to have a negative effect on optical and electrical properties of films. Therefore,the preparation ofhigh-quality poly-

Si thin films without Si islands is needed. Many reports conducted the investigationson the morphology andthe structure of residual "Si islands", but no ones had clearindication on their formation mechanism as well asthe control of the amount of these remaining Si on surface of poly-Si thin films.

In this paper, the best poly-Si films, with very little amount of residual Si on the surfaces, are obtained by choosing proper thickness ratio of pre-annealled Al and Si layers in AIC process. After annealing and chemical etching Al by appropriate acid solution, the samples are evaluated by X-ray diffraction (XRD) measurements, scanning electron microscopy (SEM), optical transmission microscopy (OTM), atomic force microscopy (AFM), energy dispersive X-ray spectroscopy (EDX) analyses and Hall measurements.

2. EXPERIMENTAL DETAILS

Corning 7059 glassesare used as substrates for depositions. Both initial Al and Si layers are deposited at room temperature with operatingAr pressure of about $3.5x10^{-3}$ torr by magnetron dc sputtering using Leybold Univex 450 system. At first, Al layers with various thicknesses such as 110 nm (A), 100 nm (B) and 90 nm (C) are deposited on the glass substrates at a fixed deposition rate of 1.19 nm/s using Al (4N) target. All Al-coated glass substrates are exposed to air for 5 min to form a thin Al oxide layer on their surfaces prior to Si deposition. Then, a-Si layers with the same230 nm thicknesses are deposited onto these Al oxide layers at fixed 0.56 nm/s rate using p-type silicon (4N) target. When the a-Si depositions finish, the (a-Si/Al/glass) structures are annealed at 500°C for 5h in vacuum furnace.The layer exchange process occurs to form Al layers on the top of the poly-Si layers. At last, top Al layers was etched off in standard Al etching solution (80% phosphoric acid, 5% nitric acid, 5% acetic acid, 10% DI water) for 4h after the annealing process.

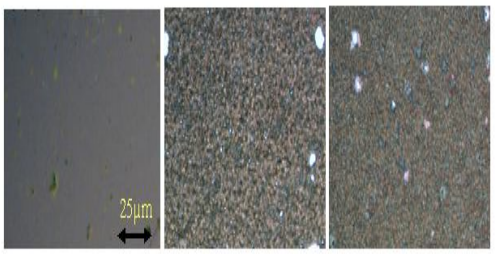
The samples characterizationsis performed using a variety of analytic techniques. The OTM, SEM (JEOL JSM-7401F), AFM (5500 AFM SYSTEM- AGILENT) are used to investigate the morphology of poly-Si films. The XRD (D8 ADVANCE – BRUKER) is used to evaluatethe degree of crystallizationand preferential orientation of obtained poly-Si thin films. EDX (JEOL JSM-7401F) is used to identify the contents of Al, Si, O elements in samples. The electrical properties of the samples are carried out by Hall effect measurement (Ecopia HMS-3000).

3.RESULTS

3.1. Surface morphology

After annealing and etching off residual Al by standard acid solution, samples are observed by optical transmission microscopy (Fig. 1). The sample A shows a surface that completely different from the others. There are very few"Si-islands" or "dendrites"observed on its surface. The image indicates that the film is continuous and smooth. This remark is

confirmed in SEM (Fig. 2) and AFM (Fig. 3) images. In contract, sample B or C is less smooth than sample A. There are a lot of "Si-islands", presenting residual Si on their surfaces [1,2].



Sample A Sample B Sample C

Figure 1. Optical transmission microscopy images of three samples (A, B, C).

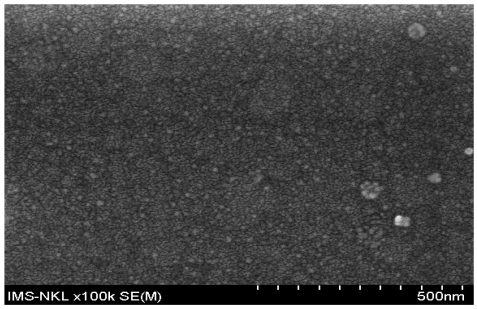


Figure 2. SEM image of sample A after etching.

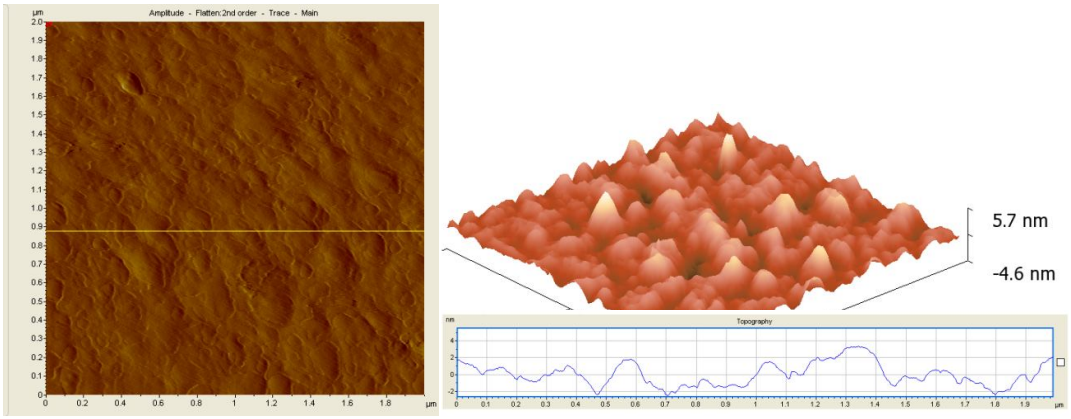


Figure 3. AFM image of the sample A

Fig. 2 shows SEM image of the sample A with uniform grain sizes of about 20-30 nanometers. This image is different from the ones of the samples containing "Si islands" on the surface reported by other authors [3,4]. This reveals that sample A represent a continuous poly-Si thin film without above residual Si.

In addition,the AFM image in Fig. 3 shows the surface morphology with average surface roughness of about 2.8 nm in scanned2 μm^2 area. The surfaceis quite smoother than the one reported by G. J. Qi et al. [5] (Rα ~ 5nm for 160nm thickness and Rα ~ 16 nm for 80 nm

thickness). This result indicates that asmooth poly-Si thin film has been obtained.

3.2. Crystallinity and electrical conductivity

The crystallinity of the Si layer after AIC process are investigated by XRD measurement.

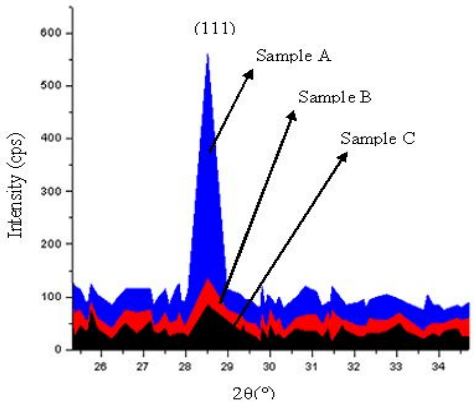


Figure 4. XRD profiles of three samples showed strong (111) orientation.

Fig 4 shows XRD profiles of A, B, C samples. In that, sample A reveals a strong Si (111) peak at 2 theta angle of 28.5°. Samples B and C also showSi (111) peaks but the crystallization is less than sample A. It is possible to infer that samples with residual Si on their surface have a low quality of crystallographic properties. For this reason, their electron mobilities showed in Table 1 are very different.

Carrier concentrations of three samples havethe same values in the range of 10^{18} cm^{-3}. These values do not change much for poly-Si thin film prepared by AIC [6]. The mobility of sample A is three times greater than one of sample B and two times greater than one of sample C. It is possible to conclude that electrical conductivity of sample A, which does not have residual islands on its surface, is better than ones of the others. The resistivities of samples B and C areabout one order ofmagnitude larger than one of sample A.

In order to estimate Al content within the poly-Si thin film, EDX analysis is used. The result in Fig. 5 reveals a small amount of aluminum embedded in the final crystallized sample A.The 1.98% percent of Al atoms is not very high if Aluminum is considered as an acceptor dopant in Si material.Because Aluminum is a shallow acceptor dopant, it leads the samples to p-type conduction.The result also shows that amount of oxygen are also incorporated in the film. This oxygen content is attributed to the formation of a thin native oxide on the surface caused by annealing sample at high temperature or by using mixture of acidsto remove Al on the surface of the sample.

Table 1. Results of Hall effect measurements of A, B, C samples

Sample	Carrier concentration (cm^{-3})	Mobility (cm^2/Vs)	Resistivity (Ω.cm)
A	1.7×10^{18}	48	$7,8 \times 10^{-2}$
B	$2,4 \times 10^{18}$	16	$1,6 \times 10^{-1}$
C	$1,5 \times 10^{18}$	23	$1,8 \times 10^{-1}$

Thin Film Standardless Standard Quantitative Analysis						
Fitting Coefficient : 0.4325						
Element	(keV)	Mass%	Counts	Error%	Atom%	K
O K	0.525	12.21	1538.08	0.02	19.61	0.8522
Al K	1.486	2.33	278.05	0.19	2.22	0.8986
Si K (Ref.)	1.739	85.46	9170.23	0.01	78.17	1.0000
Total		100.00			100.00	

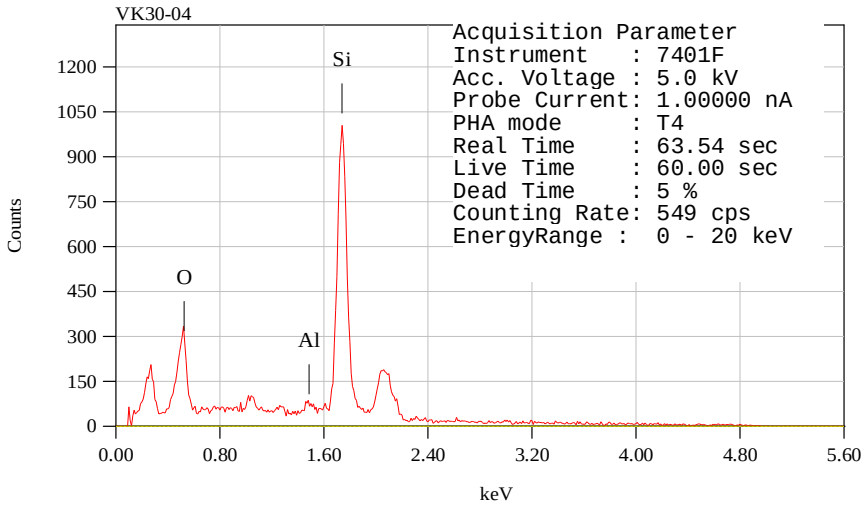


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Figure 5. EDX spectrocopy of sampleA.

4. CONCLUSIONS

Bychoosing an appropriate thickness ratio of initial Al and Si layers, we obtain the best sample with little residual Si on the surface. The crystalline structure, surface morphology, and electrical conductivity analyses show a strong influence of thickness ratio of initial bi-layer on the formation of high-quality polycrystalline silicon thin film by AIC.

SỰ HÌNH THÀNH MÀNG SILICON ĐA TINH THỂ BẰNG PHƯƠNG PHÁP NHÔM THÚC ĐẨY TINH THỂ HÓA

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TÓM TẮT: *Màng silic đa tinh thể kết tinh tốt, dẫn điện loại p được chúng tôi chế tạo bằng phương pháp nhôm thúc đẩy tinh thể hóa. Trong phương pháp này, cấu trúc màng đa lớp gồm: đế thủy tinh / Al / silic vô định hình (a-Si) sẽ chuyển đổi thành cấu trúc: đế thủy tinh / silic đa tinh thể (poly-Si) / Al (+ silic dư) chỉ bằng cách xử lý mẫu ở 500°C sau 5 giờ trong lò nung chân không. Kết thúc quá trình, màng silic đa tinh thể được hình thành trên đế thủy tinh sau khi lớp nhôm dư được loại bỏ bằng cách xử lý mẫu bằng phương pháp hóa học thông thường. Tuy nhiên, trên bề mặt màng silic đa tinh thể thu được thông thường vẫn còn rất nhiều các "ốc đảo silic" dư sót lại sau quá trình loại bỏ nhôm. Trong nghiên cứu này, chúng tôi đưa ra cách thức đơn giản, có khả năng hạn chế các silic dư còn lại trên bề mặt của màng silic đa tinh thể thu được bằng cách thay đổi tỷ lệ bề dày của lớp kim loại Al và silic ban đầu. Kết quả cho thấy với tỷ lệ bề dày của lớp Al/a-Si ban đầu là 110/230 nm, màng silic đa tinh thể thu được hầu như đã loại bỏ được hết các silic dư trên bề mặt. Các phân tích như OTM, SEM, AFM, XRD, EDS và đo tính chất điện bằng phương pháp Hall cũng đã chứng minh tính chất tốt của một màng silic đa tinh thể thu được ở tỷ lệ bề dày trên bằng phương pháp nhôm thúc đẩy tinh thể hóa.*

Từ khóa:*màng silic đa tinh thể,phương pháp nhôm thúc đẩy tinh thể hóa.*

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