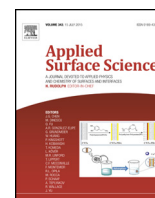




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Plasmonic silicon solar cell based on silver nanoparticles using ultra-thin anodic aluminum oxide template

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ABSTRACT

This study fabricated a plasmonic silicon solar cell covered with silver (Ag) nanoparticles (NPs) using an ultra-thin anodic aluminum oxide (AAO) template as a deposition mask. An ultra-thin AAO template of approximately 200 nm was produced using a single-step anodization process in which an Al substrate was etched with phosphoric acid (H₃PO₄) for 3 min. We then used scanning electron microscopy (SEM) to examine the thickness and density of the AAO as a function of anodization duration, the results of which were confirmed by optical transmission measurement. The photovoltaic performance of the resulting silicon solar cell with Ag NPs was characterized according to photovoltaic current-voltage and external quantum efficiency. The inclusion of Ag NPs resulted in a 32.92% increase in conversion efficiency, compared with reference solar cells produced without Ag NPs.

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1. Introduction

Nanomaterials and broadband antireflective layers present the greatest potential for the further development of solar cells. These treatments include plasmonic metallic nanostructures, silicon nanowires, and active nanomaterials [1–3] metallic nanostructures for light trapping [4], and surface texturing [5]. The reflection coefficient of solar cells can be reduced through the application of antireflective coatings (ARCs). Photovoltaic performance can be further enhanced by taking advantage of the plasmonic absorption and optical properties [6,7] of metallic nanoparticles. The optical properties of cells with plasmonic nanostructures depend largely on electromagnetic near-field enhancement and plasmonic far-field scattering at resonance wavelengths [8–10].

Surface plasmon resonance (SPR) depends largely on the nanoscale properties of the metals themselves, including the size and shape of the particles as well as the surrounding material [11–14]. This fact underlines the importance of controlling these parameters when seeking to optimize the performance of solar cells. Several techniques have been developed to attain precise control over these parameters. Two particularly efficient methods are ion-beam lithography (IBL) and electron-beam lithography (EBL); however, the implementation of these methods is extremely expensive and time consuming. This paper proposes

a reproducible and inexpensive alternative based on AAO lithography. AAO templates have attracted considerable attention for their nano-honeycomb structure with a high aspect ratio and highly ordered hexagonal pore structure [15,16,20]. These devices have appeared in biosensors, nano-magnetics, and electrochemical devices [17–19]. Researchers have previously reported the use of these techniques in the fabrication of elaborate arrays comprising metallic nanodots of hexagonal configuration uniformly distributed across the surface of a substrate [20,21]. AAO masks fabricated in this study are thinner (total thickness of only 200 nm) than existing AAO membranes [22,23]. The proposed fabrication process involves the use of AAO lithography for the deposition of Ag NPs on solar cells with a TiO₂ spacer layer. The optical and electrical properties of the resulting cells were then investigated in detail.

2. Experiment

This study employed p-type (boron doped) Si wafers, polished on one side to a thickness of 525- μ m with a (100) orientation and resistivity of 10 Ω -cm. The samples used for processing were 1 cm \times 1 cm. Following standard wafer-cleaning processes, the samples were coated with a phosphorus liquid source using spin-on film technology. The samples underwent spinning at a speed of 6000 rpm for 20 s, followed by prebaking at 200 °C for 5 min and then 400 °C for 10 min in an RTA-chamber. The samples were then capped with a 200-nm-thick SiO₂ layer and heated in the RTA-chamber under ambient N₂ at 900 °C for 2 min to initiate the diffusion of the phosphorus. The samples were then soaked

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in a hydrofluoric acid (HF) solution to remove the layers of SiO₂ and phosphorus-oxide. The samples underwent isolation etching using a photolithographic process with a solution of HNO₃:HF:de-ionized (DI) water at a ratio of 1:1:2 to produce individual cells 4 cm × 4 mm. Ohmic contact electrodes were produced by depositing an Al film with a thickness of 200 nm on the back side as well as a Ti/Al film (20-nm-Ti/200-nm-Al) on the front side using e-beam evaporation. The final step in the fabrication of bare solar cells involved annealing under ambient N₂ at 450 °C for 15 min. TiO₂ spacer layers of various thicknesses were then deposited on the top surface of the bare solar cells using e-beam evaporation. The proposed ultra-thin nanoporous AAO template was then used as a mask for the deposition of a silver (Ag) film, which resulted in the formation of Ag nanoparticles (NPs) following the removal of the AAO mask.

Aluminum foil (0.25 mm-thick) was used as the starting material in the fabrication of the AAO film. The aluminum foil was ultrasonically cleaned in acetone for 10 min followed by isopropyl alcohol for 5 min, before being rinsed in deionized water for 10 min. The foil samples were then annealed under ambient atmospheric conditions at 400 °C for 4 h. The resulting aluminum foil samples exhibited surface roughness on the micrometer scale, and therefore had to be polished in order to obtain a smooth, scratch-free, and stress-free surface prior to anodization. Chemical polishing was performed using an electrochemical solution of H₃PO₄:H₂SO₄:H₂O (2:2:1) at 50 °C for 30 min to obtain a mirror finish. The polished aluminum foil then underwent a single-step anodization process in a solution of 0.3 M oxalic acid (C₂H₂O₄) at an applied voltage of 40 V, at 10 °C for 2–4 min. The AAO thin-film formed on the aluminum template with self-ordering pores. The thickness of the AAO depends on the duration of the anodization process. One key point is ensuring that the AAO mask layer is sufficiently thin to avoid the effects of shadowing during metallic evaporation [24]. Following anodization, photoresist (AZ5214E) was coated on the AAO film and hard-baked at 125 °C for 30 min to increase the mechanical strength. The samples were then immersed for 30–60 min in a solution of copper chloride (CuCl₂), hydrochloric acid (HCl), and DI water at a ratio of 1:1:20 by weight, in order to remove the aluminum substrate. The back barrier layer of the thin AAO film was removed using chemical wet etching in a 6 wt% H₃PO₄ solution at 25 °C for various durations in order to obtain fully-opened pores with a diameter of approximately 100 nm at the bottom of the AAO. Finally, the AAO was degreased by being soaked in acetone for 10 min and isopropyl alcohol for 5 min, before being rinsed in deionized water for 10 min. This resulted in a free-standing ultra-thin AAO film floating on water in a beaker. To compare the oxide barrier layers obtained in the various etching processes, we conducted a series of experiments in which membranes were pore-etched in the conventional manner under the same experimental conditions. We then characterized the morphology and pore structures of the AAO membranes using a field emission scanning electron microscope (FE-SEM, LEO 1530). We measured the optical transmission of the AAO to examine how much of the back barrier layer was removed by etching for various durations. The performance of the resulting solar cell devices with and without Ag NPs on the TiO₂ spacer layer was characterized by measuring the dark current–voltage (*I*–*V*), external quantum efficiency (EQE) and photovoltaic *J*–*V* under AM 1.5G illumination.

3. Results and discussion

Fig. 1 presents a bottom-view SEM image of the proposed AAO template. The diameter of the resulting pores is approximately 80–100 nm with a coverage density of 38.2%. Fig. 2 presents SEM images showing cross-sections of AAO templates after undergoing

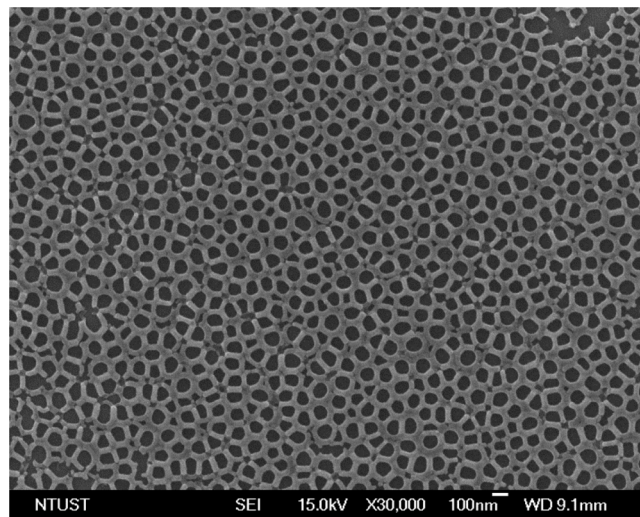


Fig. 1. Bottom-view SEM image showing self-ordering pores of proposed AAO template.

anodization for (a) 2 min, (b) 3 min, and (c) 4 min. The resulting AAO pore structure has a circular shape and high aspect ratio when anodization time exceeds 3 min. Fig. 3 illustrates the variations in AAO thickness as a function of anodization time under 0.3 M H₂C₂O₄, 40 V, 10 °C. Figs. 2 and 3 clearly illustrate how an increase in anodization time resulted in the thickening of the AAO template [25]. Fig. 4 presents the thickness of the AAO templates after etching the barrier layer for 15 and 30 min. Extending the duration of chemical etching resulted in a decrease in the thickness of the AAO template, as follows: 15 min etching time (310 nm) and 30 min (190 nm). Fig. 5 presents an SEM image showing the reverse side of an AAO template in which the back barrier layer of the AAO film was removed by etching for 30 min. The image clearly shows that the nano-holes opened from the top-surface to the bottom-surface. Generally, the use of a thick AAO template with a high aspect ratio produces shadow effects during metallic evaporation. In contrast, Lei and Chim reported that a thin AAO template is susceptible to splitting [24]. In this study, etching the barrier layer for 30 min did not cause the thin film to break. Fig. 6 presents the optical transmission performance of AAO templates without etching and after etching the barrier layer for 15 or 30 min. The optical transmittance of the AAO template increased with etching time, such that after etching for 30 min, optical transmittance of >80% was achieved at wavelengths between 450 and 750 nm. The increase in optical transmittance can be attributed to a reduction in the absorption by the walls of the nano-holes in the thinner AAO film as well as an expansion in the free space (volume of air) resulting from an increase in the number of nano-holes that opened from the top-surface to the bottom-surface of the AAO template.

Fig. 7 presents the Ag NPs produced through the deposition of Ag followed by the removal of the AAO layer. For the sake of comparison, a portion of AAO mask layer (marked “after Ag film deposition”) was left on the test sample. The profile of the Ag NPs is similar to that of the patterns seen in the AAO mask with regard to lattice and dimensions. The resulting Ag NPs have a diameter of 70–100 nm with distribution coverage of approximately 32.42%, as determined using image-J software. As shown in Fig. 7, inconsistencies in the periodicity of Ag nanoparticles can be attributed to the use of a single-step anodization process to obtain an ultra-thin AAO template, which resulted in the removal of a small number of Ag nanoparticles due to poor adhesion when the AAO template was removed after Ag film deposition.

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