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Deposition and characterization of RF-sputtered-Ta_2O_5 thin films for O_2 reduction reaction in polymer electrolyte membrane fuel cells (PEMFC)

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ABSTRACT

Ta_2O_5 thin films, proposed as the replacement of the precious Pt-based electro-catalyst in fuel cells, were sputtered on laser textured silicon substrate at 200 °C and then air annealed at 350 °C, 400 °C and 450 °C with the aim to improve crystallinity and uniformity. Characterization such as FESEM, XRD, Hall effect and Cyclic Voltammetry were employed to investigate the morphological, structural, electrical and electrochemical properties of the as-sputtered as well as annealed samples. Initial results showed that the films obtained by increasing the substrate temperature during sputtering are smoother and have better adhesion to the etched silicon substrates. It has also been observed that the rate of deposition increases resulting in thicker films for longer deposition time at higher temperature. Upon annealing, Ta_2O_5 films achieved better crystallinity consisting of orthorhombic phases. The average thicknesses of the films are in the range of 400 nm–700 nm. The proposed catalyst also shows better enhancement for oxygen reduction reaction (ORR) in prolonged time of continuous potentiostatic electrolysis as to be used in fuel cells.

1. Introduction

Direct Methanol Fuel Cell (DMFC) and Proton Exchange Membrane Fuel Cell (PEMFC) are among the many types of fuel cell that possess impending potential application as portable power generation and electric drive system. Consequently, the general cost of a fuel cell and its performance must be optimized. However, the cost of Platinum (Pt) hampers the possibility of low cost fuel cell. Therefore, a non-precious metal catalyst material is desired. One of the common problems in polymer electrolyte membrane fuel cells (PEMFC) is the catalyst loading at the cathode due to the slow-moving oxygen reduction reaction (ORR). This reaction time is almost double compared to that of the anode. To increase the rate of ORR, PEMFC requires the use of highly active catalysts to promote both the fuel oxidation at the anode and ORR at the cathode [1]. The cathode demands a huge amount of potential (> 0.8 V) to hold a long period of operation. This is challenging because of the poor stability of Pt nanoparticles supported carbon (Pt/C) at the ORR catalyst

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[2]. Tantalum oxide (Ta_2O_5) based material is one of the favourable material to be used in a fuel cell because of its ability to sustain in acidic environments and act as an active electrocatalyst for ORR [3]. The ionization potential of Ta_2O_5, which tantalum with the highest oxidation state, is 7.8 eV [3]. The performance of Ta_2O_5 as a catalyst is comparable to platinum (Pt) even though Ta_2O_5 showed poor electrochemical activity due to limited electrical conductivity. The main focus of the catalyst structures is the pore size and its distribution because these two factors affect the interaction between the ionomer and the catalyst particles [4]. Therefore, a membrane-electrode assembly was developed from a structure consisting of high surface area, good electrical conductivity and porosity to improve the performance of a PEMFC. Another challenge here is to fabricate a non-precious metal nanocatalyst with a reliable method. To further improve the operating stability and performance, fuel cell electrodes with high surface area, good electrical conductivity, and suitable porosity must be developed. A study suggests to fabricate the electrode using macro porous silicon technology [5].

In this study, porous silicon structure is proposed as an alternative to the conventional carbon paper that is being used as catalyst support material [6]. The prospects of novel porous silicon based nanostructures being used as a catalysts support material for PEMFC synthesized by chemical etching methods will also be investigated [7]. Pichonat T. reported that by replacing carbon paper with porous silicon structure, the advantage of both Nafion® and silicon will promote proton conduction and mass production of a fuel cell. The physical parameter of interest is the catalyst support porosity. Porous support material such as porous silicon wafer was also reported to be utilized as a gas diffusion layer and as well as active area for electrocatalyst placement [8]. S. Aravamudhan et al. stated that fuel cell uses novel porous silicon electrodes that are fabricated by wet etching through macro-porous silicon technology. As the pores in porous silicon act both as structures and reservoir, it also helps reducing the size of the fuel cell. This will create a capillary action that will pump the fuel towards the cell's reaction sites. The porous silicon electrode thus eliminates the need for an active external fuel pump [9].

The reactive magnetron sputtering represents a good candidate to deposit optical and electrical structures on sensitive target material and substrates. By using RF sputtering, the prospect of controlling several deposition parameters such as pressure, gas flows, target material, and power is possible. It allows a wide range of film stoichiometry, thereby it permits to deposit layers with gradient of composition [9]. Bash E. et al. reported to have done a complete annealing study of Ta films after sputter deposition process. Their XRD results shows that samples that were annealed at 200 °C showed the strongest (002) peaks and disappears after annealing at 400 °C, but several oxides peaks appear at higher temperatures [10]. The synthesized new materials of nanostructured electrocatalyst will be evaluated in terms of cyclic voltammetry (CV). CV is the most useful technique in electrochemistry. It can quickly provide qualitative information about catalysts and electrochemical reactions. This can help to further improve the electrocatalyst layer or material structure. The development of electrodes for fuel cell can be clearly grouped into two, which are the catalyst support material and non-Pt electrocatalyst nanomaterials.

Therefore, in this work, a membrane-electrode assembly was developed from a structure consisting of high surface area, good electrical conductivity and porosity to improve the performance of the catalyst. This study investigates the influence of different substrate temperatures during RF Sputtering growth of Ta_2O_5 thin films. Ta_2O_5 was sputtered onto silicon wafer via reactive sputtering and directly proceed to annealing process to achieve crystallinity of Ta_2O_5. The physical and electrical characterization of the samples such as X-ray diffraction, scanning electron microscopy and Hall measurement as well as the measurement of the ORR potential were performed. Lastly, the correlation between the physical and electrical properties of the samples with the resulting catalytic activity for the ORR was discussed.

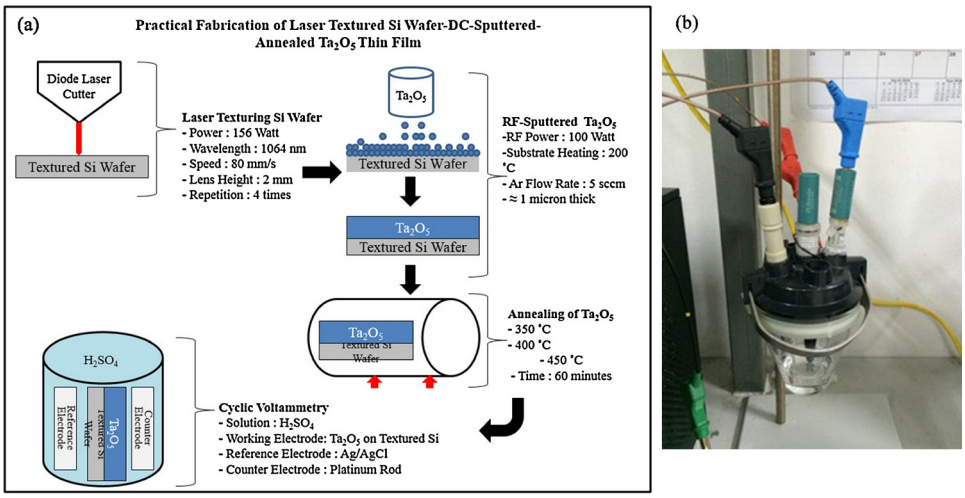

Fig. 1. (a) Schematic diagram of practical fabrication of laser textured Si wafer-DC-sputtered-annealed Ta_2O_5 thin film and (b) final CV measurement setup.