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Low-Temperature Sintering of Indium Tin Oxide Thin Film Using Split Gliding Arc Plasma

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The feasibility of sintering on indium tin oxide (ITO) thin film by a mesoplasma under open atmosphere was investigated. An ITO film of 300 nm thickness was prepared on a glass substrate by spin coating an ITO nanoparticle suspension. A mesoplasma of a gliding arc was irradiated on the ITO film. The plasma generated a temperature of 160 °C at the irradiation position. By utilizing nitrogen (N₂) gas as the plasma working fluid, the sheet resistance was effectively reduced, compared with the use of air. A small amount of hydrogen (H₂) added was more effective. A conventional gliding arc (GA) caused film damage owing to the appearance of an arc spot on the treated film, although a split gliding arc (S-GA) was effectively treated without arc spot appearance and damage. A silica binder was added to the ITO nanoparticle suspension in order to increase film adhesion and conductivity by increasing the film density. A sheet resistance of $1.9 \text{ k}\Omega/\text{sq}$ was obtained at an optimum H₂ concentration (1.0 vol %) in N₂ and a silica binder concentration (12.5 vol %), which was similar to the resistance obtained by sintering the film on a hot plate at 300 °C. [DOI: 10.1143/JJAP.47.6956]

KEYWORDS: nanoparticle ITO film, low-temperature sintering, atmospheric pressure mesoplasma, split gliding arc, resistance

1. Introduction

Indium tin oxide (ITO) is a transparent conducting oxide (TCO), and an ITO film is widely used as an electrode in liquid crystal displays (LCDs),¹⁾ solar cells,^{2,3)} and touch panels.⁴⁾ ITO films are prepared on glass and plastic substrates by various dry processes under vacuum (vacuum deposition,⁵⁾ electron beam deposition,⁶⁾ and sputtering⁷⁻⁹⁾) as well as wet coatings using metal chloride or hydrate as a precursor.^{10,11)} To decrease the initial apparatus cost, increase the productivity, and make large-area coating possible, a dielectric barrier discharge (DBD) using an atmospheric pressure plasma has recently been attempted.^{12,13)} However, the DBD method has the disadvantage of high running cost, since helium gas is required as the working gas to generate a uniform plasma. Also, the DBD method is not applicable to complex-shape substrates owing to a narrow discharge gap. Another breakthrough preparation method for on ITO film, which is the sintering of the film prepared with a nanoparticle suspension,¹⁴⁾ is considered to be a highly cost-effective method. However, the sintering temperature is typically more than 300 °C: thus, this method is difficult to apply to materials with a low thermal stability, such as polymer films. Thus, we sought to determine whether an atmospheric pressure mesoplasma, which has a temperature midway between a cold plasma and a thermal plasma, can sinter ITO at a low temperature owing to its combination advantage based on its thermal and chemical activated species.

We have also developed a plasma-energized jet (PEN-Jet),¹⁵⁾ a technique for generating plural plasmas using a branched electrical circuit,¹⁶⁾ and a split gliding arc (S-GA) for the treatment of conductive materials.¹⁷⁾ PEN-Jet and S-GA generated with a pulse power supply are typical mesoplasmas. In the present study, the feasibility of low-

temperature sintering of a nanoparticle ITO film was investigated using a conventional gliding arc (GA) and S-GA.

2. Experimental Methods

2.1 Preparation of ITO film

The ITO nanoparticle suspension was prepared by mixing aggregated and crystallized ITO nanoparticles (primary size, approximately 20 nm) with a liquid mixture of methanol and isopropyl alcohol (IPA). The solid content in the ITO nanoparticle suspension was 10 wt %. A silica binder in liquid form was added in the ITO suspension to improve adhesion to the substrate. The silica content was 20 wt % in the silica binder solution. The concentration of silica binder solution mixed with the ITO nanoparticle suspension was considered an experimental parameter ranging from 0 to 20 vol %.

The nanoparticle ITO film was prepared with the ITO nanoparticle suspension mixed with the silica binder using a spin coater on a borosilicate glass substrate $(25 \times 20 \text{ mm}^2, 1 \text{ mm} \text{ thick})$. The concentration of silica binder solution (silica content, 20 wt %) in the ITO nanoparticle suspension ranging form 0 to 25 vol %, was considered another experimental parameter. The film was eventually coated in two steps as follows: first, $100 \,\mu$ l drop and 2,000 rpm for 20 s then $100 \,\mu$ l drop again, and 4,000 rpm for 20 s. The coated film was dried for approximately 10 min prior to sintering. The target thickness of the film was 300 nm.

The sheet resistance of the film was measured with a fourpoint tester (NPS Σ -5). The film was observed with a scanning electron microscope (SEM; Hitachi S-4500II). To evaluate the ability of mesoplasma sintering, the ITO film was also sintered on a hot plate at 150 and 300 °C for 5 min under air or nitrogen (N₂) atmosphere (1 atm).

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2.2 Gliding arcs and conditions

A gliding arc (GA) is a discharge between the electrodes