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Adhesion properties of Cu/Cr films on polyimide substrate treated by dielectric barrier discharge plasma

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Abstract

In order to interconnect device elements or layers on the flexible substrate, the width and conductivity of metallization lines are very important for minimizing the size of flexible electronic devices. Especially, plasma surface treatments to improve the adhesion of metals(Cu/Cr)/PI are very critical in realizing required flexibility of metallization lines on the flexible substrates. In this study, we investigated adhesion properties of Cu/Cr/PI systems by varying surface treatment time, power, and the content of O_2 gas using atmospheric dielectric barrier discharge (DBD) plasma treatment technique. The results of contact angle measurements and atomic force microscopy (AFM) showed an increase in surface roughness resulting in the decrease of contact angle. Analysis of the elemental ratios by X-ray photoelectron spectroscopy (XPS) showed an increase in the concentration of oxygen, resulting in a decrease in the concentration of carbon and the formation of new carbon–oxygen structures. As a result, improved adhesion strengths between PI and Cu/Cr films were observed. T-peel test showed the highest peel strength of 72.6 gf/mm.

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1. Introduction

Technologies for flexible electronics have been developed to make electronic or microelectromechanical devices on inexpensive and flexible organic substrates. For the interconnection of device elements or layers on flexible substrates, metallization on the flexible substrate such as polyimide (PI) is very important. Polyimide has the desirable properties of high-temperature resistance, good mechanical strength, good dimensional stability and low dielectric constants [1–3]. In this case, the width and conductivity of metallization lines are very important parameters for minimizing the size of flexible electronic devices. Especially, plasma surface treatments to improve the adhesion of metals(Cu/Cr)/PI are very critical in

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realizing required flexibility of metallization lines on the flexible substrates.

In order to fabricate flexible electronic devices, the PI surface has to be metallized by high-conductivity metal lines. Here the poor adhesion of metals to PI, which is a consequence of the low specific surface energy, has to be overcome to achieve the flexibility of fabricated devices [4]. Adhesion of metals is strongly influenced by the chemical nature of the surface layer, because the surface layer bonds to a metal layer mainly by physi- or chemisorption [5]. A well-established method to increase adhesion physio-chemically is to expose the surface to a corona discharge in air at atmospheric pressure. Using atmospheric dielectric barrier discharges (DBD) is obviously attractive to the industrial users, because high engineering costs usually associated with vacuum-based plasma can be avoided [6]. DBD in air (or other gases or vapors) that runs at or near atmospheric pressure is very convenient for the activation or modifica-

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Fig. 1. Chemical structure of PMDA-ODA polyimide film.

tion of polymer surface. Such discharges provide chemically mild and mechanically non-destructive means of altering surface properties targeting improved surface characteristics. In this way, the surface energies of many polymer surfaces can be increased considerably as shown by a drastic decrease of the water contact angles [7]. Chemical change in the surface layer may also be induced by exposure to lowpressure gas discharges. Structural changes of the surface topography like roughening as well as chemical functionalization are also observed in low-pressure plasma treatment experiments [8]. In addition to chemical effects, the physical surface roughening may also play an important role in adhesion improvement [9].

In this study, we investigated adhesion properties of Cu/ Cr/PI systems by varying surface treatment time, power, and the flow rate of O_2 gas using dielectric barrier discharge (DBD) plasma treatment technique. The surface roughness, contact angle, and chemical bonding states of the PI film surfaces treated by O_2 atmospheric plasma were investigated by atomic force microscopy (AFM), contact angle measurements, and X-ray photoelectron spectroscopy (XPS). Also, the adhesion strengths of metals films on flexible substrates (Cu/Cr/PI systems) were measured using T-peel test [10]. The measurements showed that the obtained adhesion improvement is primarily attributed to chemical effects as well as increase in surface roughness.

2. Experiment

After all experiments were performed on PI films (Du Pont; Kapton[®]) with the thickness of 125 µm. Fig. 1 shows the chemical structure of PMDA-ODA polyimide that consists of 22 carbon atoms, 5 oxygen atoms, 2 nitrogen atoms, and 12 hydrogen atoms for each repeat unit. The DBD plasma experiments for surface treatment of PI films were carried out using a atmospheric DBD plasma system by ATMOS® and performed in Ar/O2 (20 slm/30 sccm) flow rate at the atmospheric pressure with the electrode arrangement shown schematically in Fig. 2. The lower electrode is a rectangular copper plate. The PI samples themselves are served as a dielectric barrier for the discharge. The samples under treatment were put on the top of the lower grounded electrode. The vertical position of the lower electrode can be varied in order to adjust the discharge gap length. The two electrodes were connected to a high frequency generator and the discharge was driven at the frequency of 13.56 MHz. In the experiments, the gap width of 4 mm was used. The treatment time was maintained for 35 s. The load power was varied from 50 to 150 W.

After O_2 plasma treatments of PI substrates, the in-situ deposition of Cu/Cr was performed on the O_2 plasmatreated PI substrates. The 50-nm-thick Cr adhesion layer was first deposited by d.c. magnetron sputtering of Cr targets. Also, the 100-nm-thick Cu seed layer was deposited by r.f. magnetron sputtering of Cu targets.

For the evaluation of adhesion properties of the Cr layer on the plasma-treated PI surfaces, Cu electroplating with the



Fig. 2. Schematic view of the experimental set-up used for DBD plasma treatment at atmospheric pressure.

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