



Enhanced surface hardness of flexible polycarbonate substrates using plasma-polymerized organosilicon oxynitride films by air plasma jet under atmospheric pressure

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ARTICLE INFO

Article history:

Received 19 August 2010

Accepted in revised form 29 January 2011

Available online 3 February 2011

Keywords:

Hardness

Atmospheric pressure plasma

Plasma polymerization

PECVD

XPS

ABSTRACT

An investigation is conducted on the enhanced surface hardness of flexible polycarbonate (PC) substrates using high-rate deposition (~19.1–19.9 nm/s) of plasma-polymerized organosilicon oxynitride (SiO_xC_yN_z) films with an atmospheric pressure plasma jet (APPJ) at various substrate distances. It is found that the transparent, hard and flexible SiO_xC_yN_z films can be deposited onto PC substrates at room temperature (23 °C) by injection of precursor tetramethyldisiloxan (TMDSO) into air plasma jet at atmospheric pressure. Pencil hardness measurements (ASTM D-3363) demonstrate that the surface hardness of the PC substrate is greatly enhanced from a soft surface of 3B for un-coated PC substrate to a hard surface of 7H for SiO_xC_yN_z film-coated PC substrate. This study indicates that the performance of surface hardness on PC substrates is highly dependent on the surface characteristics of the PC substrates. The surface morphology of the PC substrate is observed by atomic force microscopy (AFM) and field emission scanning electron microscopy (FESEM). The atomic compositions and chemical bonds of APPJ-synthesized SiO_xC_yN_z films are analyzed using X-ray photoelectron spectroscopy (XPS) and Fourier transformed infrared spectroscopy (FTIR).

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1. Introduction

Transparent polymer materials (e.g., polycarbonate, PC) have been widely used because of their advantages of low specific weight, low cost and ease of design in a variety of applications, e.g., display panels for hand-held devices, compact disks, plastic vessels, corrective lenses and safety windows [1]. However, their use is limited due to their soft surfaces. These limits can be overcome by coating the polymers with hard transparent protective coatings, such as SiO_x [1], SiO_xN_y [2] and Al₂O₃ [3]. Hard transparent protective coatings on polymers are mostly obtained by physical vapor deposition (evaporation or sputtering) [4] or by the sol-gel technique [5]. Plasma-enhanced chemical vapor deposition (PECVD) provides a beneficial effect on the adhesion of hard coatings to polymer substrate due to an interfacial region (~50 nm thick) which is produced between the film and the substrate [2]. PECVD also provides high deposition rates, substantially higher than those of the sputtering technique [6]. PECVD has thus become a promising technique for providing hard transparent protective coatings on transparent polymers, especially at atmospheric pressure, due to its economical (low cost, high processing

speeds and simple system which does not use vacuum equipment) and ecological advantages [7]. Plasma polymerization, a special PECVD process with precursors such as tetraethoxysilane (TEOS) [8–14], hexamethyldisilazane (HMDSO) [15–20] and hexamethyldisilazane (HMDSN) [21,22], has been proven to synthesize organosilicon thin films onto hard silicon or glass substrates at atmospheric pressure. However, only a few studies have reported the deposition of organosilicon thin films onto soft polymer substrates, such as PC [7,23,24], polyethylene terephthalate (PET) [25–27] and polyethylene-2,6 naphthalate (PEN) [27]. Plasma polymerization at atmospheric pressure using precursors such as TEOS [23] or HMDSO [7,24–27] for the restricted low process temperature (<70 °C) avoids substrate dimension changes during the film formation. Hence, the high deposition rate for deposition of organosilicon thin films onto polymeric substrates without substrate dimensional changes, which occur during film formation, plays a key role in the plasma polymerization of thin films onto polymeric substrates at atmospheric pressure. The boiling temperatures of TEOS and HMDSO liquids are 168 °C and 125 °C, respectively. In general, the lower the boiling temperature of the precursor liquid, the higher the vapor pressure of the precursor liquid. The higher the vapor pressure of the precursor liquid, the higher the concentrations of precursor vapors fed into plasmas and the higher the deposition rates of films produced onto substrates. In this study, a high-rate deposition of plasma-

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polymerized organosilicon oxynitride ($\text{SiO}_x\text{C}_y\text{N}_z$) films onto flexible PC substrates is attempted using the precursor TMDSO liquid (with lower boiling temperature of $\sim 70^\circ\text{C}$) to allow higher concentrations of TMDSO vapors injected into air plasma jet at atmospheric pressure and room temperature.

Recently, flexible polymeric substrates have been used for the applications of flexible displays [28], flexible electronics [29] and flexible solar cells [30]. Hence, the growth of inorganic thin films on the flexible polymeric substrates has been of considerable interest during the last few years. However, cracks in the inorganic thin films, which occur during flex tests, applied to coated-flexible polymeric substrates are due to poor adhesion with the flexible polymeric substrates and the film's own fragile nature. Morent et al. [31]

demonstrated that the adhesion between thin layers used for flexible and stretchable electronics can be highly enhanced by air plasmas. Hence, this study proposed a method which combines a pretreatment of flexible PC substrates using an air plasma jet, with a deposition of flexible organic–inorganic hybrid composite films of organosilicon oxynitride ($\text{SiO}_x\text{C}_y\text{N}_z$) onto flexible PC substrate by an injection of precursor TMDSO into an air plasma jet, which produces hard flexible coatings on the flexible PC substrates. The effects of the process parameter, namely the substrate distances, on the enhanced surface hardness against pencils, and the transmittances at wavelengths of 350–1000 nm for flexible PC substrates by APPJ, are discussed in this study. AFM, FESEM, XPS and FTIR are used to investigate how the film structure and the surface characteristics of APPJ-synthesized

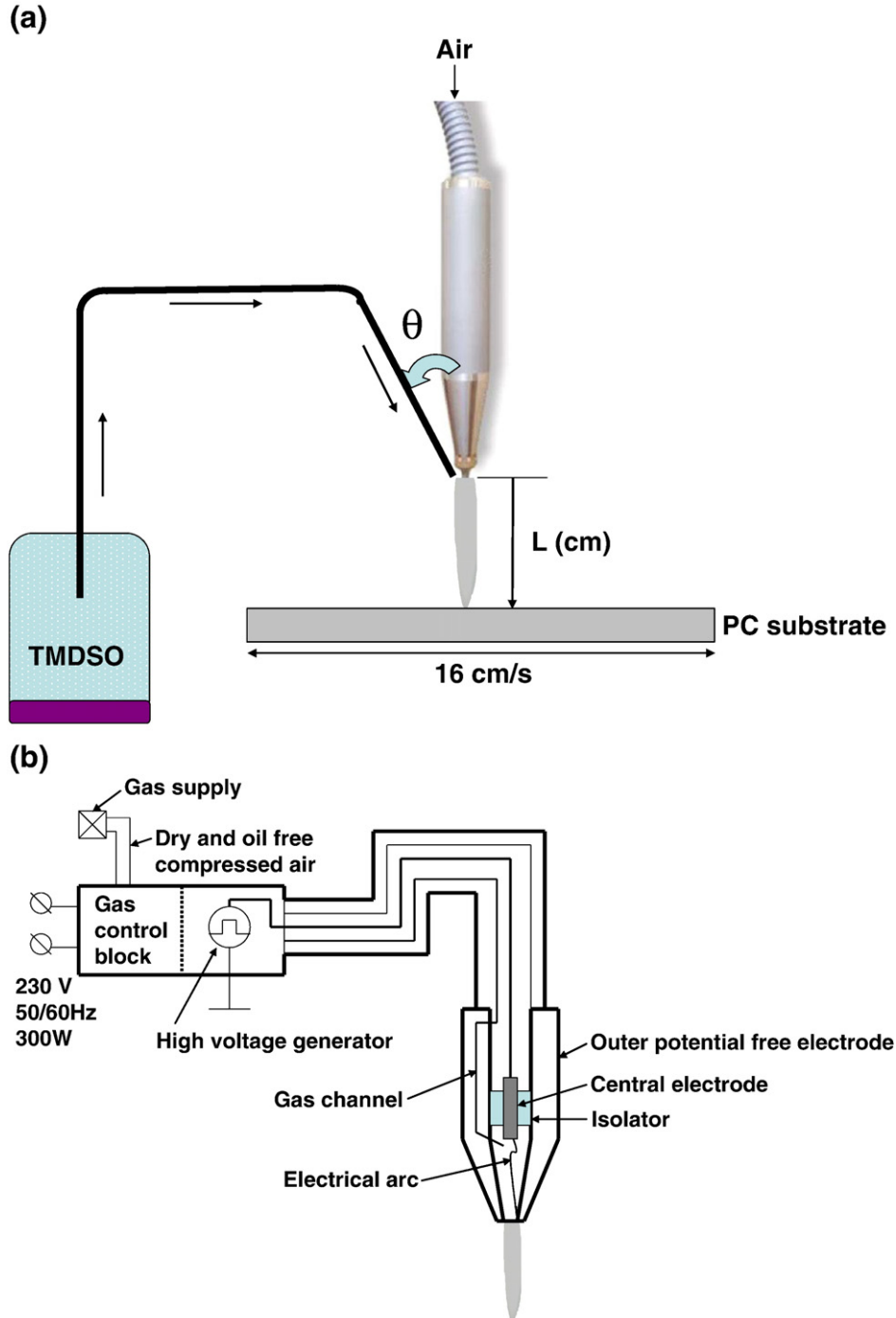


Fig. 1. Schematic diagram of (a) the APPJ set-up and (b) the plasma unit for deposition of $\text{SiO}_x\text{C}_y\text{N}_z$ films onto PC substrates.

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