Solution processed SiNxCyOz thin films thermally transformed from silicon oxide/melamine hybrid system

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A B S T R A C T

In this study, a solution-processable precursor of melamine and silicon oxide, was prepared and thermally converted into inorganic thin films of SiNxCyOz. Using tetra-ethoxysilane and hydroxyl-methyl-melamine, a transparent coating with a high loading content of silica of up to 50% was achieved through the hydrolysis/condensation reactions, which provided a transmittance of 85.1% (thickness of 1.5 ± 0.2 μm) and hard coating grade of 4H pencil test. When the silica/melamine coating was further heat-treated up to 900 °C in an inert environment, the organic melamine was converted into an inorganic compound composed of Si, N, C, and O atoms in the form of SiNxCyOz. The relative compositions of films varied with the heat-treatment temperature, e.g., providing SiN0.03C0.59O1.87 for the thin film heat-treated at 700 °C. The resulting inorganic thin films were mechanically strong and optically shiny with a low root mean square of roughness (<1.0 nm) giving dielectric constants varying from 2.75 to 1.82 with heat treatment temperature that could be used as low-k materials in commercialized optoelectronic devices.

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1. Introduction

Compared to conventional vacuum-based and vapor-phase depositions, the solution-based process has attracted a great deal of attention for the fabrication of large-area optoelectronic devices at low cost, for example, using the spin coating or ink-jet printing techniques [1–4]. Recently, a silane-based liquid precursor has been developed to fabricate a high purity silicon film for specific applications in silicon thin-film transistors, where a hydrogenated polysilane was synthesized in the liquid phase by the ring-opening polymerization of cyclic silane precursors (SiH6) via ultraviolet and heat treatment, and subsequently converted to a three-dimensional a-Si network [3]. More recently, an organic silane precursor, decapenyl-cyclopenta-silane, was thermally converted from an organic compound into an inorganic SiC3O3 carbosilane film [4]. However, it should be addressed that it is usually difficult to synthesize such stable organic silane compounds and that the candidate materials are limited.

When attempting to broaden the range of candidate precursors, sol–gel materials are considered desirable, because various solution-processable inorganic precursors, e.g., Si, Ti, Al, Zr, B, etc., can be used and they may be hybridized with various organic polymers and chemicals [5–8]. Sol–gel precursors have been hybridized with organic compounds to give organically-modified inorganic compounds through solution processes. Several polymer systems such as poly(methyl methacrylate) [9,10], poly(vinyl acetate) [11,12], epoxy [13], and poly(vinylidene chloride-co-acrylonitrile) [14] have been incorporated into silica structures to give hybridized sol–gel materials. Since the resulting hybrid coating is composed of inorganic–organic molecular networks formed through hydrolysis-condensation reactions, it is considered to be good candidate materials for subsequent conversion into a useful inorganic thin coating simply through thermal treatment [4].

Upon the thermal treatment of polymers, the organic carbon groups are often transformed into inorganic carbons via the hybridization of the carbon orbitals from sp3 to sp2 [15,16]. At temperature of around 400–600 °C, organic compounds generally decompose and emit gasses such as CO and CH4 remaining carbon and hydrogen...