Hybrid Films of Polyimide/Ladder-Like Polysilsesquioxane: Preparation, Characterization, Structure and Properties

De-Zhen Wu*, Qiu-Shuang Gao, Sheng-Li Qi, Guo-Feng Tian, Zhan-Peng Wu
College of Materials Science and Engineering
Beijing University of Chemical Technology, Beijing, 100029, PR China
E-mail: wdz@mail.buct.edu.cn

Introduction

Polyimide (PI) is known as having excellent mechanical properties, good thermal stability and dielectric property and widely used in aerospace and microelectronic industries. In order to improve its erosion resistance of atomic oxygen, water absorption capacity and adhesive property, it is effective methods to introduce organic or inorganic materials containing Si-O bonds into PI. In this paper, Ladder-like polysilsesquioxane (LPSQ) with different functional groups were incorporated into PI to obtain their hybrid films of PI/LPSQ

Results and Discussion

A series of Ladder-like polysilsesquioxane (LPSQ) with phenyl (LPPSQ), nitrophenyl (SPNPSQ), and aminophenyl (SPAPSQ) groups have been synthesized and their chemical structures and XRD patterns are shown in Figure 1 and Figure 2 respectively. According to literature reports, the two broad Bragg peaks can be an indicator of an ordered ladder-like organization of siloxane bonds, which coincide with the simulated molecular width and thickness (or intermolecular spacing between the ladder) of the ladder chain. The prepared LPSQs can be dissolved in some common solvents. The LPSQs with different groups were dispersed into poly(amic acid)(PAA) solution to form a uniform and clear mixtures, and PI/LPSQ hybrid films have been prepared by casting the mixture solution onto glass substrates and further thermal treatments, as shown in Scheme 1.

The effects of functional groups of LPSQs on the structure and properties of PI/LPSQ hybrid films were investigated. SEM analysis showed that the LPNPSQ and SPAPSQ exhibited better compatibility with PI matrix, no phase separation can be observed, as shown in Figure 3, but less silicon contents on the film surface can be detected and the mechanical properties of hybrid films can be improved. UV-vis spectra of PI、PI/SPPSQ and PI/SPNPSQ hybrid films at different thermal treatment stages are shown in Figure 4, and of which the thickness is ~50 μm. For the three hybrid films, transparence decreased as heating temperature increasing, especially at 170℃. These are caused by the solvent volatilizing at ~170℃ and imidization at higher temperature. The transparence of PI/SPPSQ films changed greater than that of pure PI and PI/SPNPSQ. For the PI/LPPSQ hybrid films, the phase separation were mainly taken place at the stage of solvent volatilization, which can be confirmed by SEM imagines, as shown in Figure 5.

Conclusions

The LPSQs with nitrophenyl and aminophenyl groups exhibited better compatibility with PI matrix, no phase separation can be observed, and the mechanical properties of hybrid films can be improved. For the LPSQs with phenyl groups, the phase separation were mainly taken place at the stage of solvent volatilization,.

Reference

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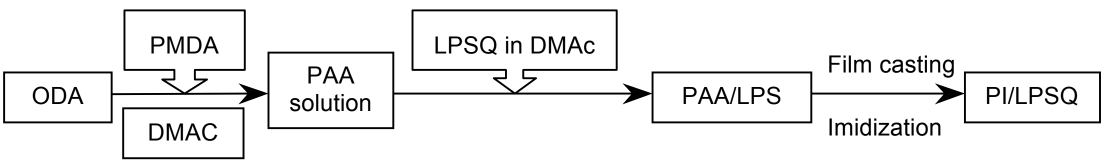

Scheme 1. Preparation flow chart of PI/LPSQ hybrid films

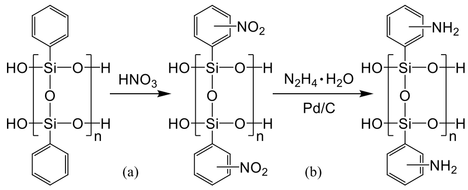

Figure 1. Reaction scheme and their structures of LPPSQ to LPNPSQ and LPAPSQ

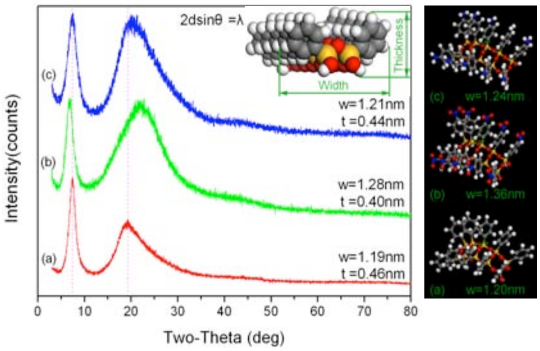

Figure 2. XRD patterns and molecular simulation images of (a) LPPSQ, (b) LPNPSQ and (c) LPAPSQ

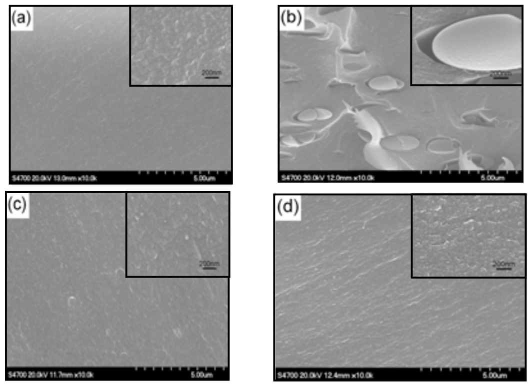

Figure 3. SEM micrographs of (a) pure PI, (b) PI/LPPSQ, (c) PI/LPNPSQ, and (d) PI/LPAPSQ hybrid films

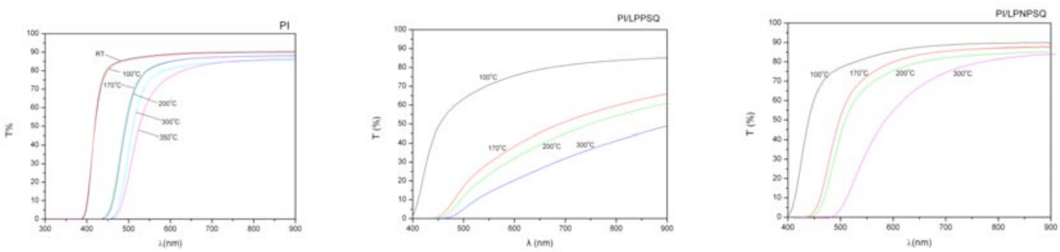

Figure 4. UV-vis spectra of pure PI, PI/SPPSQ(90/10) and PI/SPNPSQ(90/10) hybrid films at different temperature

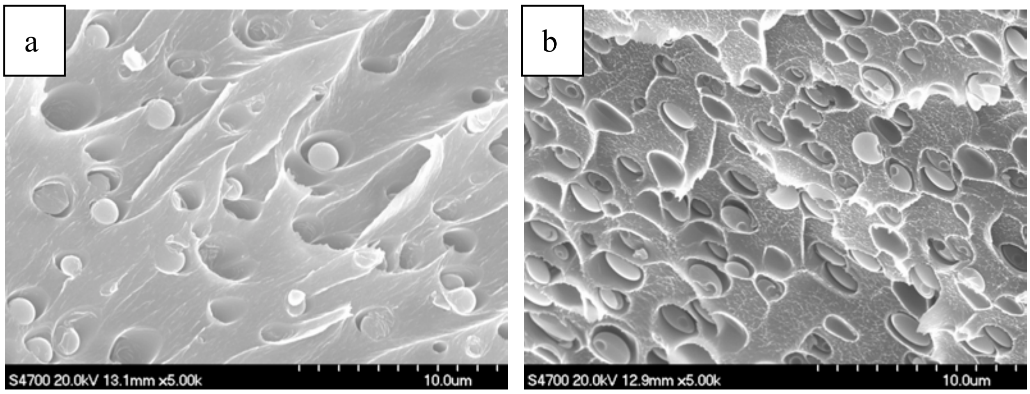

Figure 5. SEM micrograph for PI/SPPSQ(90/10) thermally treated at 100℃(a) and 200℃(b)