

# Development, Processing, and Characterization of Flexible Hydrophobic Polyimide Aerogel Thin Films

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Flexible polyimide aerogels are versatile multifunctional materials with potential use in aviation, automotive, construction, next-generation consumer electronics, astronaut suits, and outdoor apparel thanks to their combination of high strength and stiffness properties at low densities, retention of thermally insulative properties under compression, high-temperature stability, and nonflammability. Polyimide aerogels, originally developed for monolithic panels and 3D forms, have now been adapted to dust-free monolithic thin films. Manufacturing polyimide aerogel thin films requires matching formulation with correct processing parameters to achieve a positive material result. In this work, we evaluated several polyimide formulations towards production of mechanically durable, halogen-free hydrophobic all-polyimide aerogel thin films. Polyimide gels were fabricated by creating oligomers comprising combinations of diamines such as 4,4'-oxydianiline (ODA), 4,4'-(1,3-phenylenediisopropylidene)bisaniline (BisM), polypropylene glycol (PPG), and 2,2'-dimethylbenzidine (DMBZ) with 3,3',4,4'-biphenyltetracarboxylic dianhydride (BPDA) followed by crosslinking with a polyfunctional crosslinker (e.g., a triamine or a triisocyanate) to form a high-strength three-dimensional structure. All formulations were cast as both monoliths and thin films in thicknesses ranging from <0.1 mm to 1 mm using an adjustable doctor blade. Gel density was varied by adjusting polymer weight percent of the sol. Processing parameters including carrier film material and relative casting viscosity were also optimized for best results. After gelation and aging, the gels were purified through solvent exchange and dried from near-critical carbon dioxide. After drying, fluoroalkyl acrylate copolymer coatings were optionally applied to increase hydrophobicity. Bulk density, specific surface area, pore size distribution, moisture uptake, optical transparency, and mechanical properties of the resulting aerogels were characterized and compared to their respective monolithic forms. Composition-property relationships and composition-processing relationships will be discussed, as well as transition from lab to pilot-scale processing.