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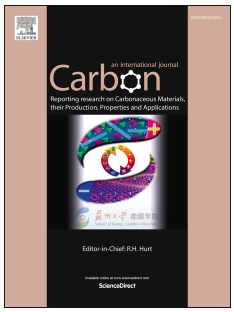

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ACCEPTED MANUSCRIPT

Solid State Magnetic Resonance Investigation of the Thermally-Induced Structural Evolution of Silicon Oxide-Doped Hydrogenated Amorphous Carbon

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Abstract

Due to their increased stability in extreme environments, relative to amorphous hydrogenated carbons (a-C:H), amorphous thin film silicon oxide-doped hydrogenated amorphous carbons (a-C:H:Si:O) are being commercially developed as solid lubricants and protective coatings. Although various properties of a-C:H:Si:O have been investigated, no definitive structure of a-C:H:Si:O has ever been proposed, nor has its thermally-induced structural evolution been thoroughly studied. The aim of this work is to better understand the structure of a-C:H:Si:O through solid-state nuclear magnetic resonance (NMR) and electron paramagnetic resonance (EPR) spectroscopies. Deeper insights into the thermally-driven structural evolution are obtained by annealing a-C:H:Si:O between 50^{o}C and 300^{o}C under anaerobic conditions and taking NMR/EPR measurements after each step. EPR results show that the number of paramagnetic defects decreases by 70% with annealing at 300^{o}C. ^{1}H NMR shows the hydrogen concentration decreases with annealing temperature from 2×10^{22} g^{-1}, and then levels off at approximately 0.7×10^{22} g^{-1} for anneals between 200^{o}C and 300^{o}C. The carbon-silicon-oxygen network exhibits some structural reorganization, seen directly as a slight increase in the sp^2/sp^3 ratio in the ^{13}C NMR with annealing. These results combined with relaxation data are interpreted according to a two-component structure largely defined by differences in hydrogen and defect contents.

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