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Analysis of the Complex Dielectric Permittivity Behavior of Composites based on $\text{Al}_2\text{O}_3/\text{AlN}$ and Precursor-Derived-SiC in the 1 MHz – 18GHz Frequency Range*

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The current research involves the synthesis of microwave-absorbing ceramic-based composites which exhibit a controllable variation in complex dielectric permittivity with respect to frequency. In particular, the ability to achieve a controllable non-Debye frequency response which conforms to an inverse power law over several frequency octaves (i.e. the real and imaginary components of the dielectric permittivity vary as $f^{-\beta}$, where f is the frequency and β is a controllable coefficient between 0 and 1) is of value, as is the ability to vary the complex permittivity at a given frequency of interest by adjusting the material composition during synthesis. Ultimately, these properties may enable the development of suitable materials for vacuum electronic amplifiers with previously unattainable bandwidth, due to closer synchronism between electromagnetic fields and the electron beam, and

enhanced power production capabilities, due to improved suppression of spurious oscillations.

This study focuses on the synthesis and dielectric analysis of both porous and fully dense Al_2O_3 -precursor-derived-SiC, as well as AlN-precursor-derived-SiC composites, obtained by applying various types of high temperature treatments to as-pyrolyzed porous composites. Synthesis of the porous Al_2O_3 -SiC composites was conducted by polymer infusion of a commercial one-step polymer precursor to SiC into partially sintered alumina discs, which were then pyrolyzed in argon at 1000°C. A set of samples was produced for single, double, triple, four times, and five times repeated infusion and pyrolysis steps, and a second set of samples was produced with an air-fire step at 600°C after each pyrolysis step. The complex dielectric permittivities of the as-pyrolyzed and pyrolyzed/ air-fired composites, as well as of an untreated porous alumina sample, were measured as functions of frequency over the range of 0.001-18GHz.

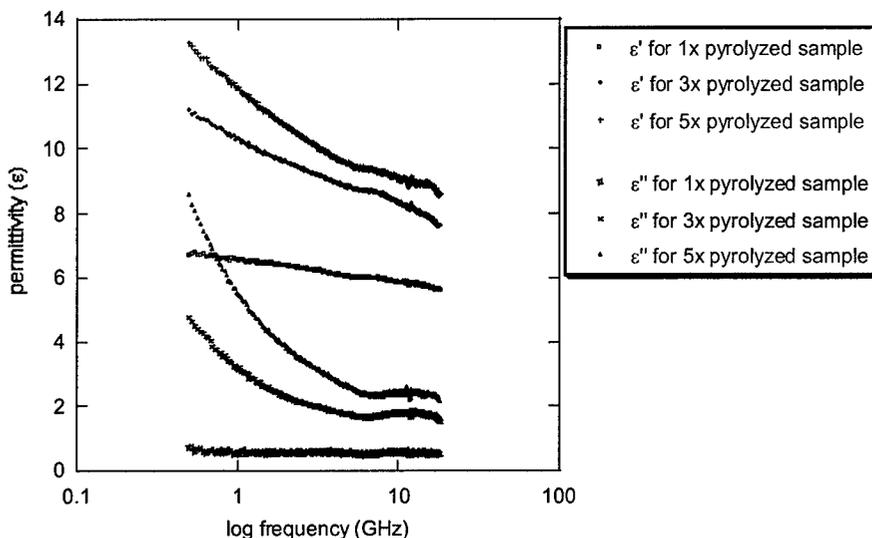


Figure 1: Measured real (ϵ') and imaginary (ϵ'') components of dielectric permittivity vs. log frequency over the 0.5-18 GHz frequency range for as-pyrolyzed Al_2O_3 -SiC samples subjected to 1, 3, and 5 repeat pyrolysis treatments in Ar at 1000°C

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The real (ϵ') and imaginary (ϵ'') components of dielectric permittivity for the as-pyrolyzed (non-air-fired) Al_2O_3 -SiC composites subjected to 1, 3, and 5 repeat pyrolysis treatments are graphed as functions of frequency in the microwave frequency range in Figure 1. The as-pyrolyzed samples demonstrate an inverse power law behavior in permittivity with respect to frequency, with an increasing number of repeat polymer infusion and pyrolysis steps leading to a systematically larger amplitude in ϵ' and ϵ'' as a function of frequency. Such a relationship is not

observed in the untreated and air-fired samples, where there is little or no dispersion in the frequency response. The as-pyrolyzed samples demonstrate dielectric relaxations, possibly caused by an interfacial polarization mechanism, as well as desirable non-Debye behavior. Relaxation behavior becomes increasingly Debye with successive infusion and pyrolysis steps. A porous microstructure with a hierarchical vesicular morphology is linked to the non-Debye behavior.

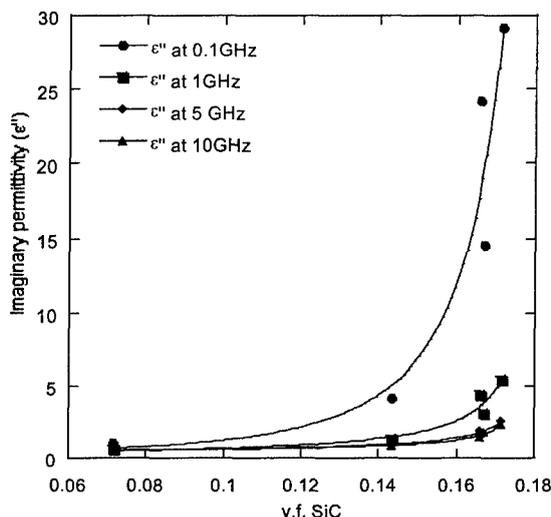


Figure 2: Measured imaginary component of dielectric permittivity at 0.1, 1, 5, and 10 GHz as a function of SiC volume fraction in the as-pyrolyzed samples

The network of conductive SiC throughout the as-pyrolyzed composites is believed to enhance percolation, which increases the contribution of dc conductivity to the overall dielectric response (particularly ϵ''). The dc conductivity is seen to increase with the number of infusion and pyrolysis repetitions. The graph in Figure 2 shows that the addition of SiC causes ϵ'' to increase most dramatically at the lowest frequency measured. This implies that the presence of SiC surrounding the Al_2O_3 particles in the as-pyrolyzed composites encourages percolation of charge and greatly enhances dc conductivity. The poor dielectric response in the air-fired samples may be caused by the transformation of SiC to highly insulating silicon oxides or oxycarbides, which limits percolation and therefore the dc conductivity contribution to dielectric response.

At high frequencies, the sub-linear universal dielectric response (UDR) dominates the overall dielectric response in the as-pyrolyzed samples, since the

disordered structure of pyrolyzed SiC is closely associated with UDR. Untreated and air-fired samples exhibit a weak super-linear power law at the highest frequencies, possibly caused by surface effects from the formation of silicon oxides during air firing, or it may be the low-frequency tail of a high frequency relaxation with a peak beyond the frequency range of the present measurements. At low frequencies, electrode polarization and interfacial polarization are thought to dominate the ϵ' response.

Of the samples studied, the 5-times-pyrolyzed sample appears to possess the most useful dielectric response in the frequency range investigated for the materials requirements outlined. Both ϵ' and ϵ'' exhibit an inverse power law frequency response of relatively high amplitude. This sample also exhibits non-Debye behavior, a low frequency dielectric relaxation, a strong UDR contribution at higher frequencies, and a large dc conductivity contribution to ϵ'' at low frequencies.