



**DIELECTRICITY, ELECTRET, PIEZOELECTRET AND PIEZORESISTIVITY
DISCOVERED IN EXFOLIATED-GRAPHITE-BASED FLEXIBLE GRAPHITE, WITH
COMPARISON WITH ISOTROPIC GRAPHITE**

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Introduction

This paper reports the dielectric, electret, piezoelectret and piezoresistive behavior of flexible graphite (FG), which is a sheet obtained by the compaction of exfoliated graphite in the absence of a binder, as made possible during compression by the mechanical interlocking among the units of exfoliated graphite, which exhibits a cellular structure¹⁻⁷. Each unit of exfoliated graphite (known as a worm) is derived from a graphite flake. Due to the high degree of compaction, the resulting sheet exhibits strong preferred orientation of the carbon layers in the plane of the sheet.

Comparison is made with isotropic graphite (IG), which is graphite fabricated by isostatic pressing, typically involving coke and coal tar pitch as the carbon precursors. Due to the isotropic pressing, the grain orientation is essentially random in IG⁸⁻¹⁵.

The electret differs from all prior electrets (polymers and ceramics) in its electrical conductivity, which enables the electret to function as a DC power source. A nonconductive electret functions as a capacitor, whereas a conductive electret functions as a resistor. Thus, a nonconductive electret cannot function as a DC power source.

The effect of stress on the electret enables stress sensing that is based on measurement of either the electric field or the capacitance. This means that the graphite senses itself in the absence of any attached or embedded sensor. This is in contrast to piezoresistivity, which enables sensing that is based on the measurement of the electrical resistance.

Materials and Methods

The FG (Polycarbon Inc., U.S.A.) exhibits strong in-plane preferred orientation, with density = 0.98 g/cm³. Comparison is made with IG (Toyo Tanso, Japan) with density = 1.77 g/cm³.

The dielectric behavior is studied at 2 kHz. The electret, piezoelectret and piezoresistive behavior is studied under DC condition. No poling is involved. All testing is performed in-plane in the elastic regime.

Results and Discussion

The relative permittivity κ (2 kHz) is 1170, compared to 530 for isotropic graphite (IG,). The DC resistivity ρ is $7.5 \times 10^{-6} \Omega \cdot m$, compared to $1.2 \times 10^{-5} \Omega \cdot m$ for IG. The inherent DC electric field E

of the electret for an inter-electrode distance $l = 40$ mm is 2.5×10^{-5} V/m, compared to 1.2×10^{-4} V/m for IG. The E increases linearly with l . The linearity is explained in terms of an analytical model, which considers that the amount of participating carriers is proportional to l , while the fraction of carriers that participate is decreased when l is increased.

The dielectric behaviour is also supported by the polarization-induced increase in the apparent resistance and the abrupt decrease in this resistance upon polarity reversal. The presence of an electret is also supported by the asymmetry in the apparent resistance change upon polarity reversal.

The electric power density (given by E^2/ρ) is 0.33 W/m³, compared to 7.8 W/m³ for IG, for $l = 1200$ m. Discharge involves short-circuiting the electret; charge involves open-circuiting the electret.

The E , ρ , κ and capacitance increase monotonically with increasing stress for both carbons. The E and ρ increase smoothly with increasing stress; for FG, the increases are totally reversible for stress ≤ 1.85 MPa and slightly irreversible for stress ≤ 3.18 MPa, in contrast to the slight irreversibility at ≥ 2.7 MPa for IG. The reversible increase of ρ and κ upon tension is probably due to a reversible stress-induced microstructural change, which may be associated with interfacial loosening that is reversible, but with slight irreversibility. For FG, a possible interface is that between the merged parts of the cell walls in the cellular structure of exfoliated graphite. For IG, a possible interface is the grain boundaries. The fractional increases in E and κ at ≤ 3 MPa are $\leq 110\%$ and $\leq 73\%$, respectively, compared to $\leq 6\%$ and $\leq 32\%$, respectively, for IG.

For FG, the κ -stress curve abruptly increases in slope at 2.1 MPa (absent for IG), which is accompanied by the onset of some κ increase irreversibility. This 2.1-MPa slope increase is attributed to a microstructural change that does not affect the strain reversibility or ρ , but increases κ . The change possibly involves loosening an interface in FG, with the loosening promoting polarization without affecting conduction. For IG, some irreversibility of the κ increase occurs at all stresses.

The piezoelectret coupling coefficient d_{33} is 3.7×10^{-8} pC/N, compared to 1.6×10^{-7} pC/N for IG. For FG, d_{33} is contributed mainly by the increase in stored charge (i.e., the strengthening of the electric dipole of the electret) upon tensile stress application. For IG, d_{33} is approximately equally contributed by the increase in stored charge and the increase in κ . The gage factor is 50, compared to 3510 for IG.

In spite of the low κ , IG exhibits high E , high d_{33} and high power density compared to FG. This suggests that the low ρ of isotropic graphite contributes to the formation of a relatively strong electric dipole. This is consistent with the notion that the dipole formation involves the movement of a fraction of the charge carriers. The combination of low ρ and high E results in high power density for IG.

Similar behavior has also been observed in PAN-based carbon fiber (Teijin, Japan). In spite of its high κ (1.25×10^4), the carbon fiber exhibits low E (1.28×10^{-5} V/m), thus resulting in low power

density (1.14 W/m³).

Conclusions

This paper reports the in-plane dielectric, electret, piezoelectret and piezoresistive behavior of exfoliated-graphite-based flexible graphite, with demonstrated feasibility of electret-based electric powering and piezoelectret-based/piezoresistivity-based mechanical stress sensing under elastic tension. The powering is enabled by the inherent DC electric field E and DC electrical conductivity. Compared to IG, FG exhibits higher κ , lower ρ , lower E , lower power density and lower d_{33} .

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