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CVD Infiltration Strengthening of a Molded Graphite

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FILLING THE PORES of a brittle material can result in a substantial improvement in physical and mechanical properties. This has been demonstrated for cement,[1] concrete,[2] ceramic tile bodies[3] and carbon[4] with pore filling accomplished by a vacuum/pressure cycle applied to force a polymer solution or molten coal tar pitch into the pores. Although this process is quite successful, it is somewhat inhibited in its accessibility to the fine pores. Infiltration by a gaseous means, such as chemical vapor deposition (CVD), does not, in principle, have this restriction. This note reports the CVD strengthening of a molded graphite.

Bokros[5] has reviewed the deposition, structure and properties of pyrolytic carbon resulting from the cracking of hydrocarbons under a variety of temperature and pressure conditions. Kotlensky[6] specifically treats the case of CVD infiltration of carbon artifacts, primarily carbon felts and filament wound shapes. He reports some data on the mechanical properties of pyrolytic carbon infiltrated graphite. For a base graphite of a 1.71 g/cm^3 and a 4200 psi flexural strength, CVD treatment increased the density to 1.85 g/cm^3 and the strength to 8000 psi.[7] Treatment in benzene at 750°C to give a 7% weight gain is reported to have increased another graphite's strength from 2500 to 8410 psi.[8]

In this research, the effect of CVD infiltration on flexural strength over a wide range of carbon additions to a molded graphite* has been studied. The molded graphite had an initial density of 1.64 g/cm^3 and an open porosity, measured by helium displacement, of $\approx$21%, with the macropores between filler particles having an average diameter of $\approx 3\ \mu$m. The uninfiltrated molded graphite had flexural strengths of 3870 ± 110 psi for fracture across the grain and 3770 ± 200 psi for fracture with the grain (5 specimens each).†

Iiltration of specimens of $^1/_4$-in. square cross section was accomplished by the chemical vapor deposition of pyrolytic carbon from a propylene (C_3H_6) source.[9] It was accomplished at 800°C in a gas mixture of 50% helium-50% propylene (1 atm total pressure) in a 3-in. diameter tube furnace at a total gas flow rate of 0.8 ft^3/h. Various deposition times up to 42 h were applied to obtain different weight gains of pyrolytic carbon up to 14%. Strengths were measured in three point bending on a commercial testing machine.‡

The increase of flexural strength with increasing carbon infiltration is plotted in Fig. 1. The relation between strength and per cent weight gain for the two orientations of fracture across, and with, the grain can each be described by separate straight lines. About a 5% increase in strength occurs for each 1% increase in weight, with the infiltration slightly more efficient at increasing the strength of specimens fractured in the transverse or across-grain orientation. The infiltration, pyrolytic carbon deposition, also decreases the average size of the macropores in the graphite, for after a weight gain of 10% (30 h infiltration), the average macropore size decreased from 3 μm to <0.2 μm in diameter.

Scanning electron microscopy of the fracture surface of the base graphite broken across the grain exposes many sharp angular pores and carbon particles, while the infiltrated specimen (to a 14% weight gain) exhibits rounded pore surfaces and particles. On the basis of these results it is postulated that, in addition to just filling the pores and cracks that might initiate fracture, deposition of the pyrolytic carbon is also increasing the radii of curvature of these defects.

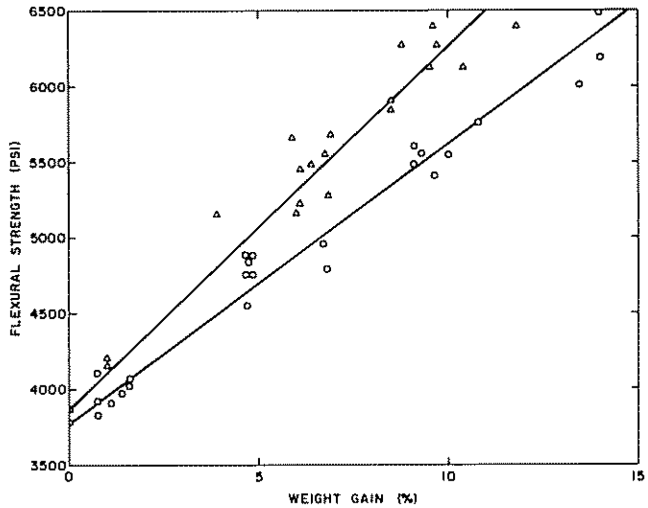

Fig. 1. Increase in flexural strength with increasing weight gain during CVD pyrolytic carbon infiltration of molded graphite. (Δ) Across grain; (O) with grain.

*Grade 3499 from Airco Speer Carbon-Graphite Div. of Airco, Inc., St. Marys, Pa.

†According to the graphite manufacturer, across the grain means ırallel to the molding direction of the specimen.

‡Instron Corp., Canton, Mass.

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