INITIAL POLYMER DEGRADATION AS A PROCESS IN THE MANUFACTURE OF CARBON-CARBON COMPOSITES

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(Received 26 April 1991; accepted in revised form 26 November 1991)

Abstract—The fundamental pyrolysis step in carbon-carbon composite processing utilizing a 2-D phenolic resin-carbon fiber composite was investigated both experimentally and analytically. The gas permeabilities of a degrading composite in the in-plane and the through-thickness directions were measured and analyzed as a function of porosity by the Kozeny-Carman eqn. Degradation kinetics of the polymer matrix was determined by the composite degradation methodology capable of describing different stages of gas evolution. Subsequently, a mathematical model was developed combining heat, mass, and momentum-transport phenomena with the degradation kinetics of the matrix. Integrating kinetic and thermodynamic variables and experimentally determined material correlations, the model could provide pressure, temperature, and degree of degradation as a function of processing time and through-thickness position in the composite. The effect of composite size and heating rate was investigated by the model, verifying practical experiences in the carbonization step during carbon-carbon manufacturing processes.

Key Words—Carbon-carbon composites, carbonization modeling, gas evolution, permeability.

1. INTRODUCTION

When a polymer matrix is exposed to high temperatures or heated at high heating rates, the energy is transferred from the surrounding area to the polymer matrix, resulting in degradation reactions. Polymer degradation results in volatile generation and weight loss of the solid material. In these pyrolysis or carbonization processes, which are essential to carbon-carbon composite manufacture, the decomposed gases are trapped within the matrix network structure, causing pressure gradients to form. This pressure build-up may be responsible for composite delamination or general damage to the matrix, resulting in cracks and voids.

As a result of matrix decomposition, permeability and porosity are continually being developed within the composite. This pressure gradient across the thickness, combined with increasing permeability and porosity, enables the decomposition gases to flow through a network of developed pores. At the same time, the gas flow takes energy out of the composite through heat convection, further causing a temperature gradient within the degrading matrix. This temperature gradient must be kept to a minimum if uniform composite parts are to be produced after carbonization. The pressure build-up and temperature gradients, as well as the developed pore structure, are considered important in subsequent processing steps. Thus, the initial carbonization process is considered as the rate-determining step in the manufacture of carbon-carbon composites, affecting the final performance of carbon-carbon composites[1–3].

One of the most important parameters in carbon-carbon processing is the material permeability, which may be defined by Darcy's law in terms of flow resistance to porous medium. The permeability of the changing matrix structure must be related to pressure build-up, char damage, composite anisotropy, resin infiltration rate, etc. In this study, the permeability of a degrading composite was measured at room temperature for the in-plane and through-thickness directions in a 2-D phenolic resin-carbon fiber composite laminate in order to investigate the effect of anisotropy.

As schematically shown in Fig. 1, degradation kinetics of the matrix was determined first because it was considered an intrinsic property of degrading composites. The weight-loss kinetics was then experimentally correlated with the minimum number of composite parameters: matrix density and porosity, which were utilized to estimate other material properties of composite during degradation.

The operating conditions in the carbonization process can be determined by understanding coupled phenomena of heat, mass, and momentum transport during degradation of the matrix material. However, the phenomena are coupled in such a way that a numerical analysis must be utilized. Therefore, based on the heat, mass, and momentum-transport eqns, as well as degradation kinetics of the matrix, a mathematical model was developed. Integrating all the intrinsic material and processing parameters into the mathematical model, a one-dimensional model provided a description of the carbonization process in terms of pressure build-up, degree of degradation, and temperature distributions, as a function of time and position.

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