Nonlinear stability analysis of the growth surface during diamond chemical vapor deposition

Pushpa Mahalingam and David S. Dandy
Department of Chemical Engineering, Colorado State University, Fort Collins, Colorado 80523-1370
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The morphological stability of the solid–gas interface under conditions of diffusive transport of reactant species towards the surface during the chemical vapor deposition process is analyzed using linear and nonlinear perturbation theories. The Landau coefficient, which represents the nonlinear growth rate, is calculated using the direct method of undetermined coefficients. A dispersive relation is derived which relates the effects of species diffusive transport towards the growing interface, surface diffusion, and geometrical factors with the stability of perturbations on the interface. The resulting relation is applied to the diamond chemical vapor deposition process. Linear and nonlinear instability of the interface is obtained for diamond chemical vapor deposition conditions. Linear instability increases but the Landau coefficient becomes larger, indicating greater nonlinear stability as the reactor temperature increases, although both linear and nonlinear analyses suggest more stability as the reactor temperature increases. However, during typical diamond growth conditions, it is predicted that the diamond–gas interface is unstable to both infinitesimal and finite amplitude disturbances. © 1998 American Institute of Physics.

I. INTRODUCTION

The onset of morphological instability in the chemical vapor deposition (CVD) process and the critical properties that cause an initially planar interface to break down have been predicted by means of linear perturbation analyses.1–9 In general, CVD constitutes a free-boundary problem for which the interface shape and position are unknown, and therefore the temperature and concentration fields at the gas–solid interface are also unspecified. If the amplitude of the interface shape perturbation can be assumed to be infinitesimal, linear perturbation analysis may be applied to study the onset of interface instability. This approach has been adopted by a number of investigators in the study of CVD processes1–9 and the analogous solidification problem.10–16

Overviews of morphological linear stability analysis applied to the solidification problem are available.17–19

Linear stability analysis predicts critical values of the governing parameters for the onset of instability.20 The amplitude of the perturbation imposed on the interface is infinitesimal in this case. However, a linear analysis is limited in scope to predicting only the onset of instability; it cannot be used to analyze the possible evolution of the unstable planar interface to a nonplanar cellular interface. The exponential temporal growth rate of the perturbation predicted by linear theory is not suitable for describing the actual growth of the interface due to the fact that higher-order terms in the growth rate expression are neglected—terms that are not negligible once the interface shape deviates sufficiently far from planarity. In order to study the perturbation and growth behavior of an interface more accurately over a relatively longer period of time, it is necessary to retain higher-order terms in the growth rate, requiring a nonlinear analysis.

A number of investigations have been carried out to extend beyond the linear growth regime when examining the stability of a solidification growth front,21–30 but the study of the nonlinear stability of a solid–gas interface during a CVD process has been reported in only one instance.31 Three types of theoretical treatment of the nonlinear problem for the directional solidification of a binary alloy can be identified in the literature. One approach, using expansion methods, provides nonplanar interface solutions where the amplitude of perturbation is finite but very small. This is a weakly nonlinear method and was adopted by Wollkind and Segal21 using a Stuart–Watson approach22,23 to study interface stability during the solidification of a dilute binary alloy. The Landau coefficient in the amplitude equation was calculated by means of an adjoint eigenvalue approach. Wollkind et al.24 also studied the interface stability of a binary alloy using the direct method to calculate the Landau coefficient. They showed that, for the moving boundary problems under consideration, the direct method was superior to the adjoint operator method, particularly when the solution itself is desired in addition to the solvability condition.

A second approach has been to replace the full set of equations governing the system with a set of equations that are much easier to solve. These are the so-called geometrical25 or boundary-layer models.26 The essential feature of these models is that they approximate the temperature gradients at the interface, thereby eliminating the need to solve the equation governing energy transport. The problem is then reduced to solving a first-order nonlinear equation in one variable. The drawback to this approximation is that it ignores all global properties associated with the transport equation and at long times these models produce unphysical phenomena such as boundaries that cross. Nevertheless, this approach provides important insights into solidification processes.