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Three-dimensional morphological instabilities in chemical vapor deposition films

Chi-Chuan Hwang [a,*], Hwei-Yen Yang [b], Jin-Yuan Hsieh [b], Yuan-Mon Dai [a]

[a] *Department of Mechanical Engineering, Chung Yuan University, Chung-Li, 32023, Taiwan*
[b] *Department of Mechanical Engineering, Nan Ya Junior College, Chung-Li, 32034, Taiwan*

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Abstract

In this article, an analysis of nonlinear three-dimensional (3-D) morphological instabilities in chemical vapor deposition (CVD) is presented. We establish a set of mathematically governing equation and boundary condition for the system of CVD and derive a weakly nonlinear evolution equation by considering a diffusion-limited growth condition to study the morphological instabilities in the CVD process. This evolution equation not only can predict the behaviors of interfacial growth of films, but can also be a basis of weakly nonlinear analysis. The analysis from critical condition is adopted to investigate the two-dimensional (2-D) band-like cells and the 3-D hexagonal structures. © 1997 Elsevier Science S.A.

Keywords: Chemical vapor deposition (CVD); Diffusion; Surface diffusion; Surface morphology

1. Introduction

In producing modern electronic devices, chemical vapor deposition (CVD) is one of the main processes for thin-film growth. A CVD process is designed to supply reactive gases to the surface of substrate under conditions that encourage surface reaction and discourage reaction elsewhere. CVD processes exist for a vast variety of films, but they are particularly useful for forming refractory, nonmetallic films that are not easily evaporated. Nowadays, due to higher demands on the property of the electronic device and the corresponding increasing cost for equipping, theoretical modeling or numerical simulation of the film, morphology has become more important in the research of CVD processes.

Normal CVD process is undertaken under high pressure circumstance (typically 0.1 to 1 atm) since the mean free path of the gaseous particle is much shorter than the typical length of morphological features and the value of the Knudsen number [1] is very small ($Kn \ll 1$). Therefore, transport of the reactants to the surface is diffusion-driven and a continuum model is appropriate. van den Brekel and Jansen [2,3] studied the stability of the planar gas-solid interface during CVD processes. They considered a stagnant boundary layer of the gas and assumed the surface reaction to be proportional to the supersaturation. In the model, a reversible reaction is taken into account and the Gibbs-Thompson relation is used to relate the equilibrium concentration on the surface to the curvature of the interface; however, the important effect of surface diffusion is neglected. Therefore, the only stabilizing factor in their model is capillarity. They showed that vapor growth is essentially an unstable process. The study was then extended to nonisothermal conditions, and they found that the results remained qualitatively the same. What is most important, their study showed that, as the process is diffusion-limited, the effect of nonisothermal conditions was most dominant. Bales et al. [4] undertook numerical simulation to the van den Brekel-Jansen model and considered both the diffusion-limited and the reaction-limited processes. They found the morphology to be fingerlike and very similar to what has been found in the Hele-Shaw problem [5] for diffusion-controlled process. The morphology was also similar to the results predicted by Kardar et al. [6] for reaction-controlled growth.

Palmer and Gordon [7,8], neglecting temperature gradient, included the effects of surface diffusion, gas diffusion and reevaporation in the growing film. By assuming that the reaction took place in the gas phase and only adsorption and reevaporation took place on the surface, they found that gas diffusion is the destabilizing factor, while both surface diffusion

* Corresponding author. Fax: 886-3-4563160. E-mail: chichuan@cchp01.cc.cycu.edu.tw.

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