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SELF-ORGANIZED ZRO_2 DENDRITES BY SOL-GEL PROCESS

Rongsheng Zhou[*,**], Qiangmin Wei[**], Kai Sun[**], Lumin Wang[**]
[*]School of Mechanical Engineering, Shanghai Jiao Tong University, Shanghai 200030, China
[**]Department of Materials Science and Engineering, University of Michigan, Ann Arbor, MI 48109, USA

Dentritic crystal growth is one of the most extensively investigated theoretical topics in the general area of nonequilibrium pattern formation. The dentritic growth, explained by the diffusion-limited aggregation (DLA) model[1], is controlled by shape instability, in which predominented factor is diffusion-either the diffusion of latent heat from the growing solidification front or the diffusion of chemical constituents toward and away from that front. This instability leads to the small bumps grow out into fingers because they concentrate the diffusive fluxes ahead of them and therefore grow more rapidly than a flat surface. Compared with theoretical investigation, experimental study of evolution in dendrite crystal growth is relatively limited. Here we report the growth of ZrO_2 dendrites using Sol-Gel processes. We suggest that the undercooling generated by the crystallization of ZrO_2 in the solution and the concentration of ZrO_2 are the two dominant contributors to this phenomenon.

Powders of ZrO_2 were first prepared by a sol-gel process using $ZrOCl_2 \cdot 8H_2O$, Urea, HMTA, hydroxypropyl cellulose (HPC) and isopropyl alcohol. During the process, the solution of $ZrOCl_2 \cdot 8H_2O$ with HPC was added to ice-cooled deionized water. Urea and HMTA were then added slowly to the aqueous $ZrOCl_2$ solution. Finally the aqueous solution was added to isopropyl alcohol to form a milky suspension with a volume ratio of alcohol to aqueous solution 5. The milky suspension was heated to 200ºC. After the suspension was dry, the ZrO_2 particles were obtained. The ZrO_2 particles were so fine that they tend to aggregate together, but they can be dispersed in the ethanol with an ultrasonic dispenser. After dropping the solution of ZrO_2 to a glass slide or a silicon substrate and drying for a few minutes, the dentrites formed. Fig. 1 shows the Scanning Electron Microscopy images of ZrO_2 crystal dendrites on the surface of a silicon wafer. It can be seen that an interlocking network of highly developed snowflakes is generated. Fig. 1B illustrates the details of a single, isolated dendrite. The morphology consists of a growing needle with a parabolic tip. Figs.1 C and 2D are the ZrO_2 dendrites on various parts of surface showing the slightly different structures which can be attributed to the effects of boundary conditions or the surface properties.

Another experiment was made on a glass surface with a higher undercooling for dendrite formation. The velocity of ZrO_2 atoms on the glass surface is slower than that on the silicon surface, resulting in the unique morphology of ZrO_2 dendrites (Fig. 2A) which is not as sharp as those on the silicon surface. The concentration of ZrO_2 in the solution is another factor controlling the dendrite formation. Until now we found only one concentration can form this pattern and others produce spherical ZrO_2 particles as shown in Fig. 2B. This sensible solution can be explained by the solvability theory[2] in which the surface tension and crystalline anisotropy determine the dentritic structure.

[1] Witten, T.A., SandercL.M., Phys. Rev. Lett. 1981, 47 1400
[2] Bisang, U., and Bilgram J.H., 1996, Phys. Rev. E 54 5309

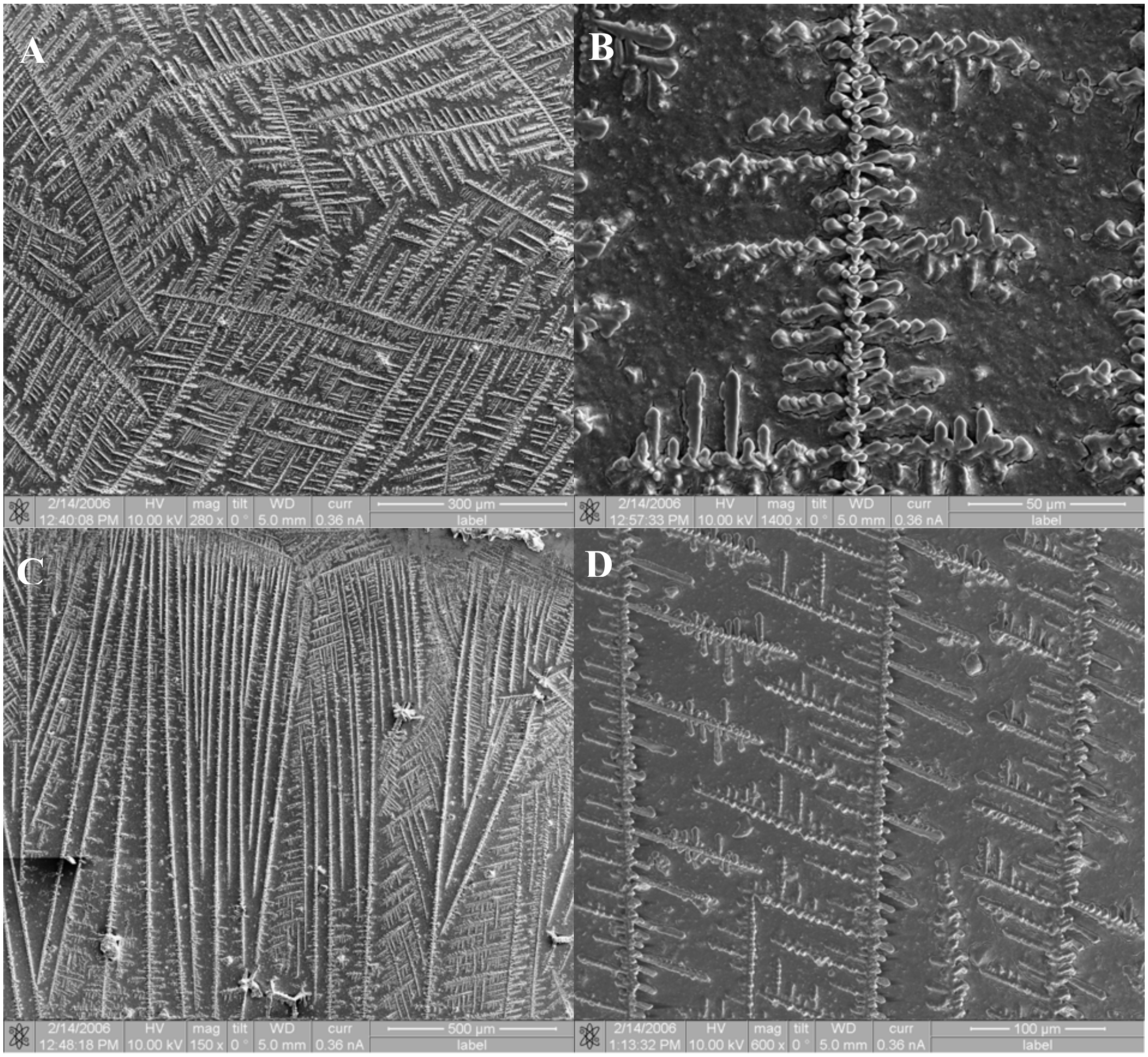

Fig. 1 SEM images of ZrO_2 dendrites formed on silicon surface. A, B, C and D are from different parts of the surface showing the effects of different boundary conditions.

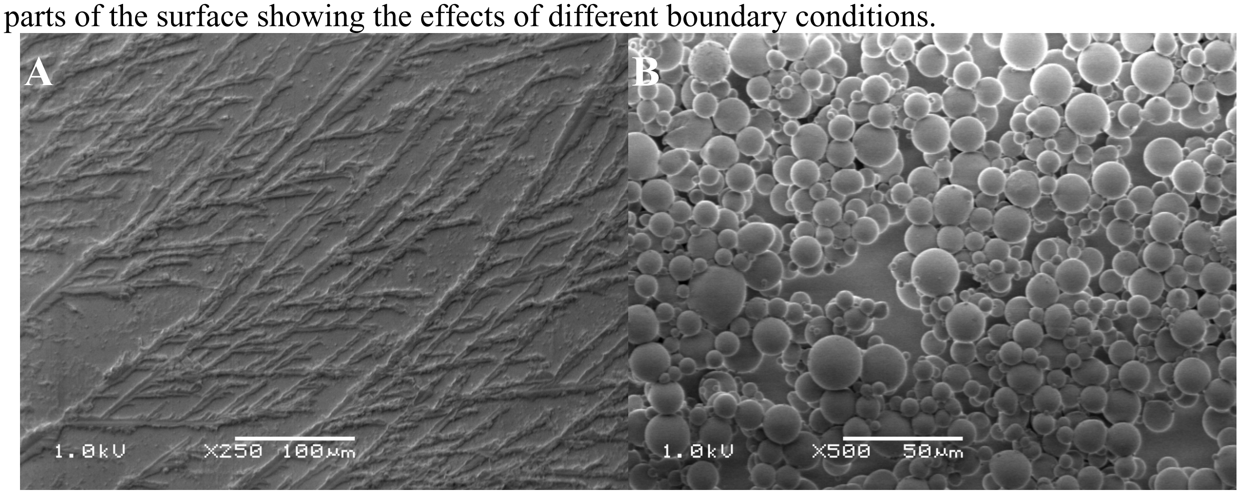

Fig. 2 SEM images of a ZrO_2 dendrites and particles formed on a glass surface. A and B are generated by using different concentrations of ZrO_2 in the solution.

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