主　論　文　の　要　旨

論文題目　　Multiscale numerical investigation on
　　　　　　step dynamics during solution growth of
　　　　　　4H-SiC
　　　　　　(4H-SiC の溶液成長のステップダイナミク
　　　　　　スに関するマルチスケール数値解析研究)

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論　文　内　容　の　要　旨

4H-SiC (silicon carbide) is a promising material for next-generation power devices due to its excellent physical properties. The top-seeded solution growth (TSSG) method is suitable for obtaining high-quality, large SiC bulk crystals. By applying the dislocation-conversion phenomenon in the TSSG method, SiC crystal with few dislocations can be obtained. High and steep macrosteps have been proven to be critical for converting dislocations. However, over-developed macrosteps also induce macroscopic defects. Step bunching is an essential process in macrostep formation. A systematic understanding of the mechanisms of step bunching is significant for controlling macrosteps structure. However, such an understanding is difficult to be obtained only through experimental studies due to the complex factors coupling mass transport and surface kinetics varying over different scales and the extreme ambiance required for SiC crystal to grow. Therefore, this thesis intends to utilize a multiscale numerical investigation to discuss factors affecting step bunching during the solution growth of SiC crystal.

First, a growth experiment was carried out on a 2-inch 4H-SiC crystal was grown on the C face of a 1º off-axis seed crystal using the TSSG method. Step-flow growth towards the [11-20] direction occurred over the entire growth surface. Inhomogeneity in the surface

morphology was observed. The step height increased monotonically along the step-flow direction. The non-uniform step height resulted in the spatial distribution of threading screw dislocation (TSD) conversion. No conversion of TSD took place in the upstream area of step flow due to small step height. In contrast, TSD conversion frequently happened in the center of the crystal and the downstream area, with a conversion ratio as high as 80% in these areas. A macroscale numerical simulation was carried out over the bulk solution to investigate the reason for the step-bunching distribution. An outward solution flow occurs under the growth surface. Therefore, the solution forms an anti-parallel and a parallel flow against the step flow in the upstream and downstream areas. Thereby the opposite affects the step bunching behaviors, respectively. This study revealed a step bunching phenomenon that occurs only in large-sized crystals' growth and pointed out that the solution flow direction is a critical controlling parameter for uniform growth.

In order to study the development of step bunching in quantity, a numerical model coupling mass transport and step kinetics in the mesoscale was constructed.
The macroscale numerical study results were utilized as boundary conditions in the mesoscale model. Surface roughness due to step bunching was studied under different solution flow velocities. The step bunching development with solution flow present is consistent with the experimental results. However, there was disagreement between the simulation and experimental results without solution flow. Since solute's incorporation into steps and transport together determines step bunching progress.
Next, numerical investigations around the development of step bunching induced by solvent physical properties representing mass transport and step kinetics were carried out. It is found that step bunching occurs due to the depletion of solute in the region with high step density, caused by a high step kinetic coefficient. On the other hand, by promoting the transport of the solute in the solution, the step speed becomes uniform, thereby the step bunching can be prevented. Furthermore, we proposed a non-dimensional Damköhler number for crystal growth in step-flow mode. It correlates incorporation rates with bulk diffusion rates and can build a phase map of growth rates and step bunching stability. Several solvents are located according to reported experimental results in the phase map, demonstrating the possible usage of the phase map as a pointer for solvent designing.

In order to evaluate a solvent for its tendency towards step bunching, knowledge of both diffusion coefficient and step kinetic coefficient is required. The latter, however, is

difficult to be measured through general experiments. The Kinetic Monte Carlo method was applied to conduct a microscale numerical investigation on step kinetics at crystal-solution interfaces. Step kinetic coefficients are calculated from kink density and crystallization rates at step edges of steps on vicinal {0001} facets. The vicinal interfaces contain periodic arrays of bilayer steps. Kinetic coefficients for both <1-100> and <11-20> oriented steps are determined under different temperatures, supersaturations, and the presence of impurity species. There are three types of steps on each polarity {0001} face, and the values of step kinetic coefficients are shown to be highly anisotropic. The kinetic coefficient of S_N steps increases exponentially with temperature. In contrast, the values of S_D and S_M steps remain constant until the temperature rises over 2000 K, under which a thermal roughening occurs on the {0001} facets. At low temperatures, the kinetic coefficient of the S_N step is about 3.8×10^5 µm/s, two orders smaller than the steps of SD and SM types, which is 5.9×10^7 and 4.6×10^7 µm/s, respectively. At low supersaturations, the movement of S_N steps is observed through one-dimensional nucleation on the step edges. At the initial stages of step-flow growth, the S_D steps bunch into S_N steps due to the different growth rates, while the poor mobility of S_N steps is considered as a factor that keeps crystal growth away from further step bunching. An aluminum-like impurity species is introduced in the simulation, and its influence on step kinetic coefficient and step bunching was investigated. The Al-like impurity behaves on the two polarity faces differently. On the Si face, incorporation occurs both on terraces and at step edges. The Langmuir-style adsorption on the terrace is considered as an origin of an impurity-induced step bunching. On the C face, step incorporation is realized. The suppression of step mobility due to impurity adsorption on step edges is considered a reason that suppresses step bunching on the C face.