

## Supporting Information

### **Influence of interfacial energy in growth of SiC single crystals from high temperature solutions**

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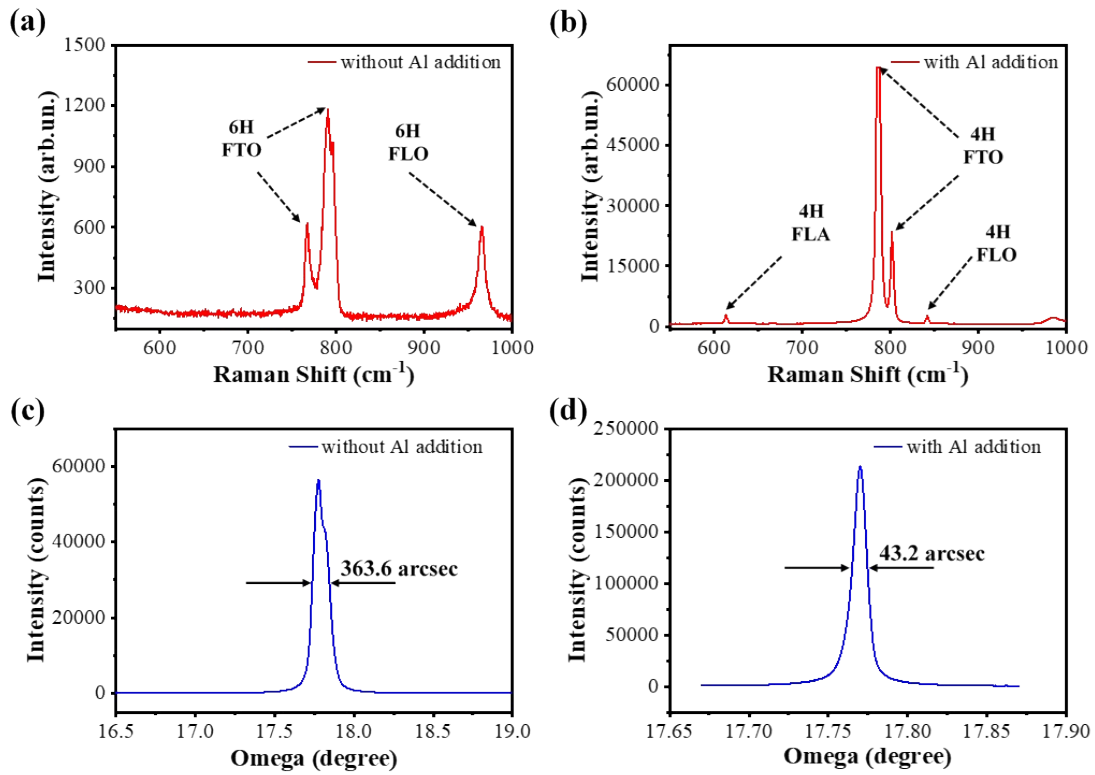
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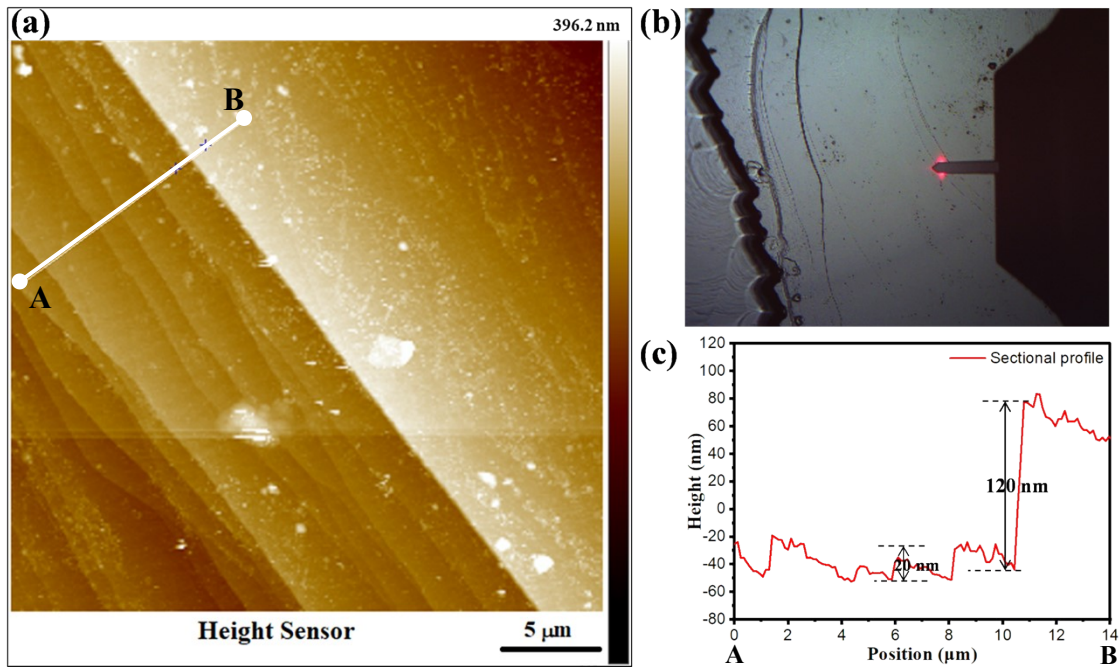
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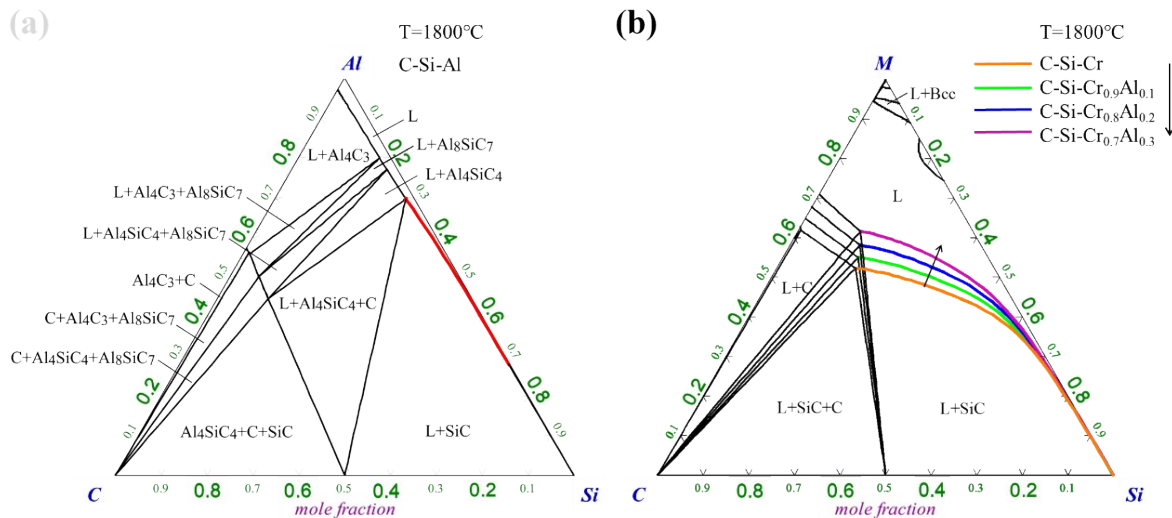
**Figure S1.** Raman spectra of SiC single crystals grown via TSSG (a) without and (b) with Al addition to the solution. X-ray rocking curves of SiC single crystals grown from solution (c) without Al and (d) with Al addition.

The height of smaller step flows (see in Figure 3d) was measured with AFM. The result shows that one of the smaller steps has a height of  $\sim 120$  nm.



**Figure S2.** (a) AFM height image of smaller steps in terrace of SiC grown from the solution with adding Al. (b) The photo image for the region measured by AFM. (c) Sectional profile corresponding to the region marked by the white line AB in the AFM height image (a).

Ternary phase diagram of C-Si-Al and pseudo quaternary phase diagram of C-Si-M ( $M = \text{Cr}, \text{Cr}_{0.9}\text{Al}_{0.1}, \text{Cr}_{0.8}\text{Al}_{0.2}, \text{and } \text{Cr}_{0.7}\text{Al}_{0.3}$ ) at 1800 °C were calculated with the thermodynamic calculation software, FactSage (6.3). Extremely small region of liquid phase in the isothermal section of C-Si-Al ternary phase diagram indicates that the solubility of carbon in Al-Si solvent is very low, see [Figure S3\(a\)](#). The liquidus shifts gradually towards orientation of low carbon concentration with the increase of Al content as shown in [Figure S3\(b\)](#), indicating the solubility of carbon in the C-Si-Cr solvent decreases instead of increases by adding Al. Typically, the carbon solubility decreases by less than 1 at% with 10 at% Al addition.



**Figure S3.** Isothermal section of ternary phase diagram of (a) C-Si-Al and quasi-ternary phase diagram of (b) C-Si-M ( $M = \text{Cr}, \text{Cr}_{0.9}\text{Al}_{0.1}, \text{Cr}_{0.8}\text{Al}_{0.2}, \text{and } \text{Cr}_{0.7}\text{Al}_{0.3}$ ) at 1800 °C.

Figure S4 shows the photo images of the surfaces of solutions with and without Al addition after cooling at room temperature. Numerous small black particles are seen on the solution surface with Al addition, see Figure S4(b). Some of the black particles present regular hexagonal shape, which are SiC particles characterized by the EDS (Figure S4(c)). In contrast, nearly no SiC particle is observed on the whole surface of solution free of Al after the growth. The element composition of solutions with and without Al are measured by EDS. The solution free of Al is composed of C, Si, Cr, and Ce with an atomic ratio of 24.45 %, 28.25 %, 31.03 %, and 16.29 %. The solution with Al is composed of C, Si, Cr, Ce, and Al with an atomic ratio of 23.64%, 29.77%, 28.65%, 15.75%, and 2.18%.

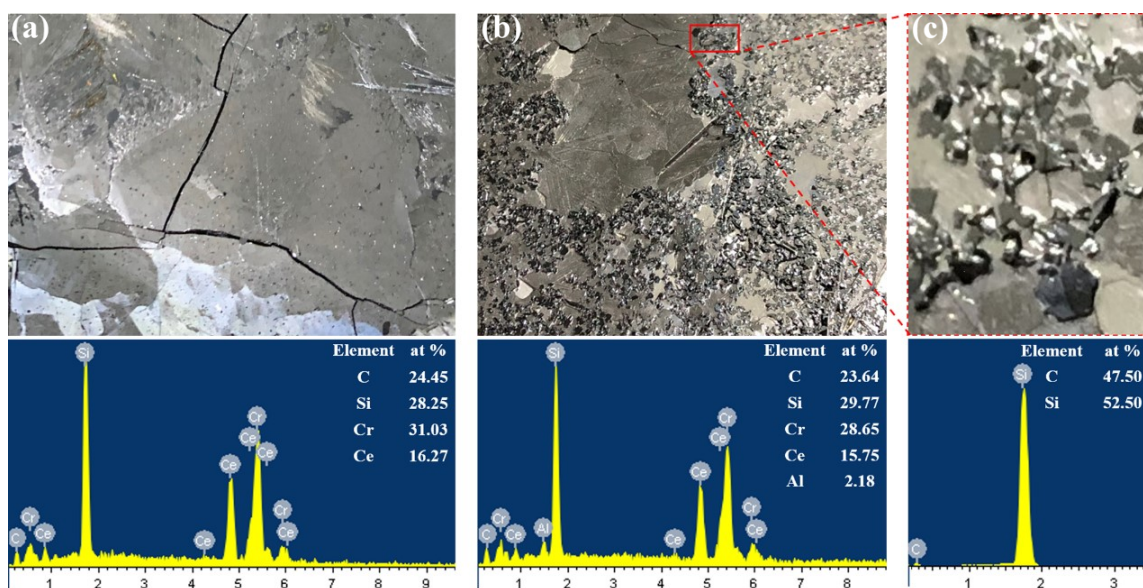


Figure S4. (a) Photo image and EDS result of the solution surface without Al addition after crystal growth at room temperature. (b) Photo image and EDS result of the solution surface with Al addition after crystal growth at room temperature. (c) Magnified photo image of the area dotted by red rectangle shown in Figure S4b. EDS result of the black particles on the solution surface shows these particles are SiC particles.

The effect of Al on the solution surface morphology (Figure S4) can be explained from the view point of the interfacial energy. The change of Gibbs free energy ( $\Delta G$ ) in the process of SiC homogeneous nucleation from the high temperature solution is expressed as,

$$\Delta G = V \cdot \Delta\mu + S \cdot \gamma_{SiC - solution} \quad (S1)$$

where  $V$  and  $S$  is the volume and surface area of SiC micro-particle from homogeneous nucleation, respectively,  $\Delta\mu$  the change of chemical potential for per volume SiC molecule precipitating from the solution under a certain supersaturation,  $\gamma_{SiC - solution}$  the interfacial energy between SiC particle and solution. For the homo-nucleation of SiC from supersaturation solution, the  $\Delta\mu$  is negative, acting as the driving force. The  $\gamma_{SiC - solution}$  is positive, which is the barrier for homo-nucleation. Decreased  $\gamma_{SiC - solution}$  by adding Al leads to lower barrier for the homo-nucleation of SiC. Thereby, many SiC particles from the homo-nucleation are seen on the surface of the solution with Al addition after growth. However, for the solution free of Al, heterogeneous nucleation of SiC on the crucible wall occurs in the cooling process after crystal growth due to the larger  $\gamma_{SiC - solution}$ .