4H-SiC 表面における窒素の取り込みおよび脱離の反応の動力学に関する SiC 消費速度を因子とした理解

Understandings of the kinetics of N-incorporation and N-removal reactions for the 4H-SiC surface using the SiC consumption rate as an essential factor

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[Motivation] To maximize the SiC surface N density which still has plenty of room to improve, it is inevitable to understand the SiC surface nitridation kinetics. We have proposed a kinetic model shown by Eq. (1), by simplifying the N-incorporation rate (N_r) and rate constant of N-removal (k) to be independent of the nitridation time (t) [1],

$$A_N(t) = \frac{N_r}{k}(1 - e^{-kt}) \qquad (1)$$

where $A_N(t)$ is the surface N density. The saturated $A_N(t)$ would be determined by N_r/k. In this study, we will investigate the essential factors to determine N_r/k by decomposing it into k and N_r for 4H-SiC/SiO₂ structure.

[Experimental] 4°-off 4H-SiC(0001) substrates with epitaxial layers ($N_D \sim 1 \times 10^{16}$ cm⁻³) were oxidized in O₂ to grow SiO₂ with the thickness (T_{OX}) varying from 6 to 30 nm. Nitridation was performed by annealing in N₂, N₂/O₂ or N₂/H₂ ambient with various O₂ partial pressures (P_O). Change of SiO₂ thickness (ΔT_{OX}) during the annealing was determined by grazing incident x-ray reflectivity. After removing the SiO₂ layer in diluted HF, the relative ratio of N (*N/Si*) was estimated with the peak area ratio of N1s to Si2p core-level XPS as the indicator for $A_N(t)$.

[Results and discussions] Fig. 1 shows a typical t dependence of *N/Si*. The initial nitridation rate (N_r^{ini}, see Fig. 1) was defined experimentally as the indicator for N_r [1], and k was obtained by fitting the curve calculated using Eq. (1) with N_r^{ini} to the experimental data, where $A_N(t)$ was replaced by *N/Si*. Verification of Eq. (1) was reported in the previous work [1] for the case where only the passive oxidation rate was taken into account as the factor to determine k. In this study we expand the cases to the active/passive mixed oxidations where the SiC consumption rate (R_{SiC}) should be considered to include both active and passive oxidations. Note that R_{SiC} could be expressed by two experimental parameters: passive oxidation rate and the ratio between active and passive oxidation rates (active-to-passive ratio). Fig. 2 shows the relationships between k and the linear SiO₂ growth rate (R^{lin}), which was obtained from the linear ΔT_{OX}-t relationship (data not shown) as the indicator for the passive oxidation rate. Linear relationships between k and R^{lin} were found irrespective of T and T_{OX}, while the slope strongly depends on P_O. This could be explained that the active-to-passive ratio is sensitively affected by P_O regardless of a small variation of T, considering the reported P_O-dependence of the passive/active transition [2]. Thus, k is suggested to be proportional to R_{SiC}. Regarding N_r, three processes are proposed, which are the removal of topmost C atoms to generate vacant sites, the adsorption of oxidants and nitridation species to the vacant sites. By considering R_{SiC} as the essential factor, it was proposed that N_r should increase with R_{SiC}, and saturate when R_{SiC} is sufficiently large [3]. Here we further proposed that when R_{SiC} is sufficiently small, N_r should be small and show little dependence on R_{SiC}, due to the limited reaction sites and the competition for those sites between nitrogen and oxygen. Fig. 3 summarizes the relationship between N_r^{ini} and k which was assumed as the indicator for R_{SiC}. It was found that N_r^{ini} could be approximated as constant when k is sufficiently small. Except for these cases, the increase of N_r^{ini} with k is consistent with the finding that N_r^{ini} increases with R_{SiC} [3]. From this data, it is suggested that both k and N_r could be described as a function of R_{SiC}, which means that R_{SiC} should be an essential factor to determine the values of both k and N_r.

[Conclusions] We proposed that both k and N_r could be described by using R_{SiC} as the essential factor. The integrity of the model was discussed experimentally for various conditions. **[Acknowledgement]** This work was partially supported by JSPS KAKENHI. **[References]** [1] T. Yang and K. Kita, 69th JSAP Spring Meeting (2022). [2] N. S. Jacobson et al., Oxid. Met. **75**, 1 (2011). [3] T. Yang and K. Kita, JJAP **61**, SC1077 (2022).
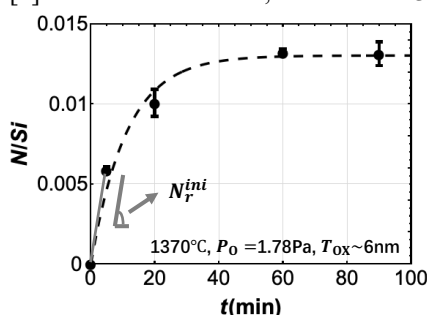

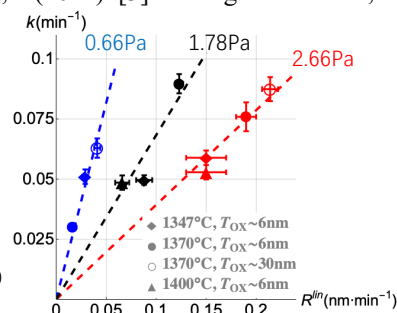**Fig.1.** The t dependence of *N/Si* for a typical nitridation condition.

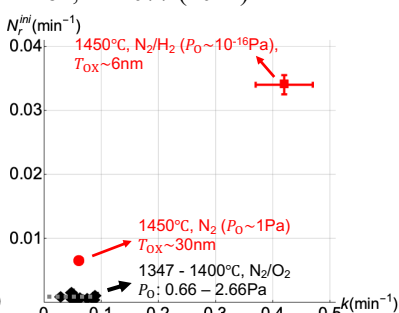**Fig.2.** The k-R^{lin} relationships for the conditions shown in the legend.

Fig.3. The N_r^{ini}-k relationships for the conditions shown in the legend.