Si(100) ETCHING BY THERMAL-ENERGY HYDROGEN ATOMS

[1]Joo Hyun Kang, [1]Sam Keun Jo* and [2]John G. Ekerdt
[1]*Department of Chemistry, Kyung Won University, Sung-Nam, Kyung-Ki 461-701, S. Korea.*
[2]*Department of Chemical Engineering and Science and Technology Center, University of Texas, Austin, Texas 78712, USA.*
**samjo@plaza.snu.ac.kr*

ABSTRACT

Efficient Si(100) etching by thermal H atoms at low substrate temperatures has been achieved. Gas-phase etching product $SiH_4(g)$ upon H atom bombardment, resulting from direct abstraction of $SiH_3(a)$ by impinging H atoms, was detected with a quadrupole mass spectrometer over the substrate temperature range of 105 - 480 K. Facile depletion of all surface silyl (SiH_3) groups, the dissociative adsorption product of disilane (Si_2H_6) at 105 K, from Si(100)2x1 by D atoms and continuous regeneration and removal of $SiD_3(a)$ during atomic D exposure have been observed. At 480 K, however, etching stopped when adsorbed $SiH_3(a)$ were all consumed. These results provide direct evidence for efficient silicon surface etching by thermal hydrogen bombardment at cryogenic temperatures as low as 105 K. We attribute the high etching efficiency to the formation and stability of $SiH_3(a)$ on Si(100) at lowered surface temperatures, allowing the $SiH_3(a)$ abstraction reaction by additional H atom to produce $SiH_4(g)$.

Hydrogen, ubiquitous in the manufacturing processes of modern semiconductor electronic devices, plays a key role in hydride chemical vapor deposition (CVD) [1]. Hydrogen chemistry on silicon surfaces often dictates processing parameters [2]. Etching of semiconductor surfaces by energetic plasma beams has been widely employed [3]. The energetic particles in plasma, however, result in unwanted surface damages and roughening [4]. Potentially, dry etching by thermal-energy hydrogen atom beams offers a milder etching condition, giving damage-free, smoothly etched surfaces. Efforts have been made toward possibilities of hydrogen beam-induced silicon etching [5]. However, H beam etching efficiency observed at ambient and elevated temperatures has been reportedly very small.

In this Letter, we report our results that Si(100) etching efficiency can be greatly enhanced by lowering the substrate temperature. Glass, Jr., et al. [6] recently reported that the direct etching reaction, $SiH_3(a) + H(g) \rightarrow SiH_4(g)$, does not occur at T_s = 133 K and that SiH_4 desorbs only thermally at T_s = 153 - 213 K from a disproportionation reaction, $SiH_3(a) + SiH_x(a) \rightarrow SiH_4(g) + SiH_{x-1}(a)$, on a porous silicon surface [6]. In contrast, we detected real-time evolution of gas-phase SiH_4 from Si(100) upon atomic hydrogen exposure at temperatures as low as 105 K, while observing no thermal desorption of SiH_4 at $T_s \leq$ 500 K. Cryogenic surface temperatures were found to enhance the stability of $SiH_3(a)$ species, the formation of which is thermodynamically driven by the simultaneous Si-Si back-bond scission and Si-H bond formation. The enhanced stability, or higher surface concentration [7], of $SiH_3(a)$ led to the higher rate of $SiH_4(g)$ evolution, i.e., the etching rate.

Lightly B-doped (14~20 Ω?) p-type Si(100)-2x1 samples, 0.5 x 15 x 20 mm^3 in size, were coated with a 2000 Å of tungsten film sandwiched by 200 Å tantalum films to facilitate linear direct-current heating up to 1400K. The sample was then clamped with a pair of tantalum clips, which were attached to LN_2-cooled Cu blocks. The substrate temperature was monitored by a chromel-alumel thermocouple glued into a small hole

near the sample edge with Aremco (Utratemp™ 516) high-temperature ceramic glue. After a 5K/sec heating to 1300 K and 1K/sec cooling, Si(100)2x1 was clean and well ordered, as checked by Auger electron spectroscopy (AES) and low-energy electron diffraction (LEED). Disilane (Voltaix, 4% Si_2H_6 in He) was dosed using a crimped capillary-controlled 3/8"-diam. tubular doser positioned ca. 1 mm in front of the sample surface. Si_2H_6 exposures were controlled by the reservoir pressure, typically 1-5 torr, behind the pinhole and dosing time. Atomic H beam was generated by a hot (ca. 2000K) W spiral filament positioned in front of a 0.25"-diam. tubular H_2-gas doser facing the sample surface from a distance of 3 cm. Temperature-programmed desorption (TPD) and isothermal desorption (ITD) measurements were made with a shielded, differentially-pumped quadrupole mass spectrometer (QMS) through a 3 mm-diameter conical aperture positioned 1 mm (TPD) and 15 mm (ITD) in front of the sample. The ultra-high vacuum system with a base pressure of $1x10^{-10}$ torr was pumped by a combination of turbomolecular, Ti-sublimation, and ion pumps. Turbomolecular pumps were backed by diffusion and rotary pumps in series.

Si_2H_6 adsorbs dissociatively, with a small negative activation energy, as two SiH_3(a) groups at dangling bond sites on Si(100)2x1 at 105 K [7-14]. Dissociative adsorption probability of Si_2H_6 increased with decreasing surface temperature and was highest on well-ordered a Si(100)2x1 surface, while it quickly diminished with increasing surface roughness [7]. Physisorbed multilayer Si_2H_6 desorbed molecularly at 120 K. At low Si_2H_6 exposures, SiH_3(a) further decomposes into SiH(a) and SiH_2(a) when dangling bonds are available [9]. In subsequent TPD, a H_2 peak at 780 K (β_1) first and then another H_2 peak at 670 K (β_1) grew with increasing Si_2H_6 exposure. With increasing Si_2H_6 exposure, SiH_4 desorption at 610 K started at the onset of β_2-H_2 desorption and grew to its maximum beyond the β_2-H_2 peak saturation. The SiH_4 peak shifted to lower temperatures with increasing intensity, consistent with a second-order disproportionation reaction, $SiH_3(a) + SiH_x$ (x= 1-3) $\rightarrow$ $SiH_4(g) + SiH_{x-1}$ [7]. Since the surface lifetime of SiH_3(a) on Si(100) at low coverages is relatively long (several minutes) even at room temperature [11], continued high-flux Si_2H_6 exposure is expected to saturate all dangling bond sites with SiH_3(a). The kinetically-controlled decomposition of SiH_3(a) into SiH_2(a) and SiH(a) is likely to be hindered further by our cryogenic adsorption temperature (105 K) and high Si_2H_6 exposure, leading to a high SiH_3(a) coverage, in spite of the steric hindrance [9, 15]. We estimated SiH_3(a) coverage to be near unity for our Si_2H_6-saturated Si(100) surface on the basis of total H balance from SiH_4 and H_2 TPD [7]. Surface silyl species, SiH_3(a), saturated on Si(100) were found to be stable up to 600 K, where SiH_4(g) desorbed [9, 16]. The maximum 600-K peak intensity of SiH_4 from Si(100) exposed to Si_2H_6 at T_s = 105 K was about 2 orders-of-magnitude higher than that from Si(100) saturation-exposed to atomic H. No other desorbing species were detected between multilayer Si_2H_6 desorption at 120 K and SiH_4 desorption at 600 K in TPD. A trace amount of Si_2H_6 desorbed concomitantly with SiH_4 at 600 K. A fixed Si_2H_6 exposure to give a maximum SiH_4 desorption was used in the experiments here.

In Figure 1, we show thermal desorption spectra from Si(100) saturated with Si_2H_6 at T_s = 105 K without H atom bombardment. Upon adsorption Si_2H_6 adsorbs either molecularly or dissociatively at 105 K. Subsequent heating desorbs physisorbed Si_2H_6 at 120 K. At surface temperatures between 120 K and 500 K, the surface must be saturation-covered with SiH(a), SiH_2(a), and predominantly SiH_3(a) [1-2]. Upon further heating above 500 K, SiH_3(a) further decomposes and H_2 subsequently desorb at 670 K and 780 K. A small amount (a few % of total SiH_3(a)) of recombinative Si_2H_6 desorption at 600 K was also observed. For our H atom experiments, we used a substrate temperature window of 140 - 480 K, where the surface is saturated mostly with SiH_3(a) without any thermal desorption.

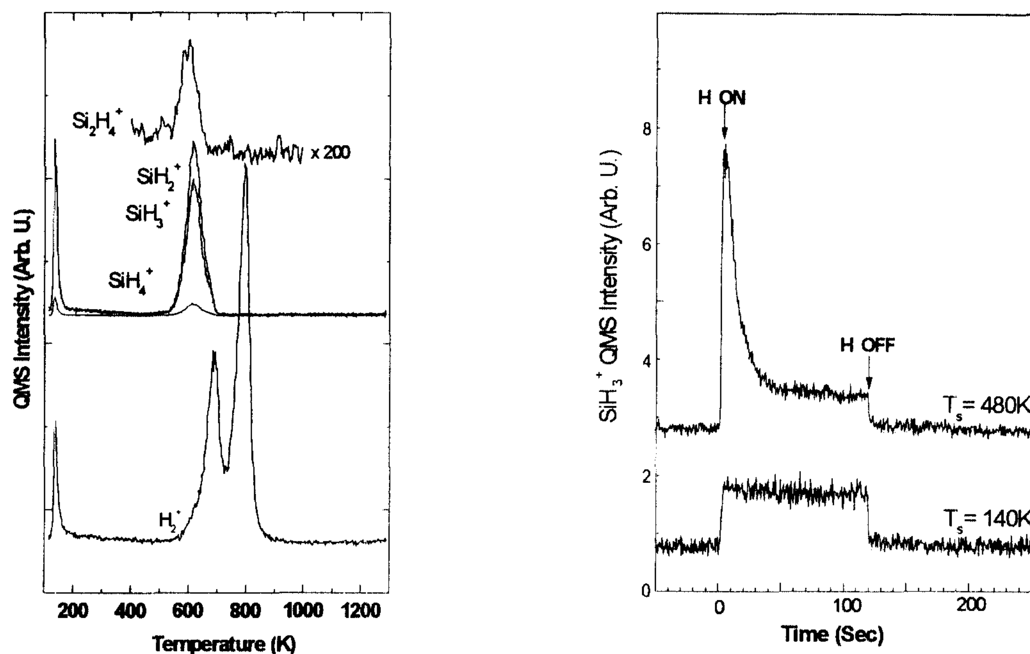

Fig.1. [LEFT PANEL] Thermal desorption specta of Si_2H_6 saturation-adsorbed on Si(100)2x1 at 105 K. Si_2H_6 (120 K), Si_2H_6 (600 K), SiH_4 (615 K), and H_2 (680 K & 790 K) desorbed during a linear temperature ramp of 4 K/sec.

Fig.2. [RIGHT PANEL] Real-time evolution of gas-phase SiH_4 from SiH_3-saturated Si(100) during 2-min. bombardment of thermal H atoms at substrate temperatures of T_S = 140K (Lower Panel) and T_s = 480K (Upper Panel). Si(100)2x1 surfaces were freshly saturated with Si_2H_6 at T_S = 105 K and then warmed up to the indicated temperatures prior to H beam exposures. H_2 pressure, measured by an uncorrected ionization gauge, during H atom dosing was $7x10^{-6}$ torr.

In Figure 2, we have shown the real-time evolution of gas-phase SiH_4 from SiH_3/Si(100)2x1 during exposure to thermal H atoms at two different substrate temperatures. A Si(100)2x1 surface was first dosed with Si_2H_6 at 105 K with an exposure to give a maximum SiH_4 desorption peak intensity at 600 K in TPD. Si_2H_6-dosed Si(100) was then raised to and held at either 140 or 480 K during H atom exposure. SiH_3^+ (m/e = 31) QMS signal was monitored for SiH_4 desorption. Evolution of SiH_4 was confirmed by fragment ions of the desorbing species with correct relative intensities for SiH_4 from m/e 28 to 32 (Si^+:SiH^+:SiH_2^+:SiH_3^+:SiH_4^+ = 60:37:100:80:7). The relative intensities of SiH_4 cracking fragments were determined *in situ* by back-filling the chamber with $5x10^{-7}$ torr SiH_4 at an ionization energy of 70 eV. Note the instantaneous rise of SiH_3^+ signals for both substrate temperatures and their fast (T_S = 480 K) and slow (T_S = 140 K) decay upon H atom bombardment. The SiH_3^+ signal did not decrease further below that at 2 min even with extended H atom exposure for T_S = 480 K, whereas it increased slightly further from the intensity at 2-min H exposure time for T_S = 140 K. It was confirmed that the residual SiH_3^+ signal intensity at 2-min H exposure time for T_S = 480 K came from sample

confirmed that the residual SiH_3^+ signal intensity at 2-min H exposure time for T_s = 480 K came from sample holder and other surfaces, by the observation that the residual signal intensity did not change even when the sample was turned away from H beam and that no SiH_4 and a less-than-saturation amount of H_2 desorbed in the post-H beam TPD (see Figure 2). Similar background signals were reported for CH_3(a) abstraction by atomic H on Cu(100) [21]. Therefore, SiH_3(a) must have been completely abstracted away by a 2-min H atom exposure at T_s = 480 K. On the contrary, a steady-state amount of SiH_3(a) must remain on the surface even with a long H-exposure time at T_s = 140 K. This conclusion is corroborated by our observation that the real-time SiH_4(g) evolution intensity with a 2-min H exposure is still significantly higher than the background level (Fig. 2) and that a significant amount of SiH_4 desorbs in subsequent TPD (Fig. 3).

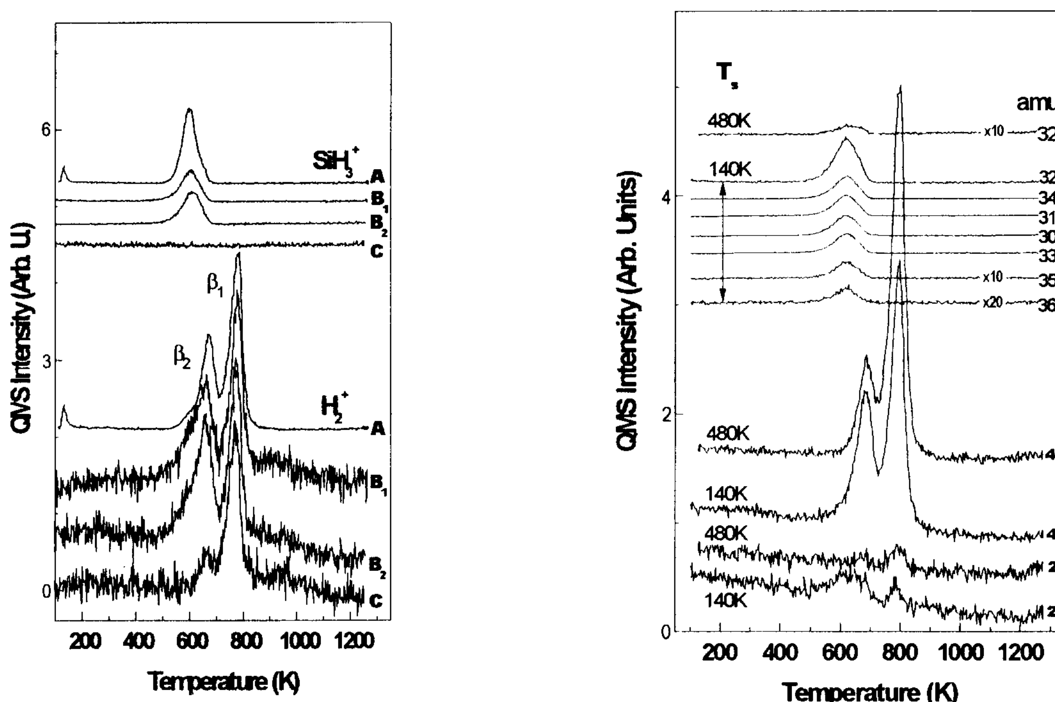

Fig. 3. [LEFT PANEL] Post-H beam TPD of H_2 and SiH_4 from Si(100) saturated with SiH_3(a) from Si_2H_6 exposure at T_s = 105 K. Spectra labeled A, B_1, and B_2 are with 0, 2, and 12-min H beam bombardment at a surface temperature of 140 K, respectively. Spectra labeled C are with 2-min H beam exposure at a surface temperature of 480 K. A 4K/sec ramp rate was used.

Fig. 4. [RIGHT PANEL] Post-D beam TPD of H_2, HD, D_2, and SiH_xD_{4-x} (x = 1-4) from Si(100) saturated with SiH_3(a) from Si_2H_6 exposure at T_s = 105 K. SiH_3/Si(100) was bombarded with D for 3 min. at T_s = 480 K and for 12 min. at T_s = 140 K, respectively. Substrate temperatures (T_s) during D beam exposure and desorbing mass fragments are indicated in the Figure. A 4K/sec ramp rate was used.

After 2-min H atom exposures, TPD was subsequently performed. The resultant TPD spectra are displayed in Figure 3. As shown in the spectra "A" for comparison, SiH_4 (600 K) and H_2 (660 K and 780 K)

desorption peaks were observed from the Si_2H_6-saturated surface without H atom exposure. A relatively higher β_2- to β_1-H_2 intensity ratio ($\beta_2 : \beta_1 = 1:2$) from Si(100)2x1 saturation-exposed to Si_2H_6 at T_s = 105 K, compared to that ($\beta_2 : \beta_1 = 1:3$) from Si(100)2x1 exposed to atomic H at T_s ~ 400 K, is attributed to the roughened surface due to Si adatoms deposited from disilane decomposition. A 2-min H atom beam exposure at T_s = 480 K quickly suppressed SiH_4 desorption completely. Moreover, the total amount of H_2 desorption (especially β_2-H_2) decreased, indicating an extensive and fast SiH_3 depletion by H atoms (Figure 3-B_2). The significantly decreased β_2-H_2 desorption suggests that the rate of SiH_3 abstraction by H atom beam, even at T_s as high as 480 K, is higher than that of H-atom addition to SiH(a) to form SiH_2(a) and/or of further decomposition of SiH_3 into neighboring dangling bond sites. In contrast, a 2-min H atom exposure at T_s = 140 K suppressed the SiH_4 desorption only by 50% with an increased β_2-H_2 peak intensity as shown in Figure 2-B_1. Even after a 12-min H atom exposure, both SiH_4 and β_2-H_2 TPD peak intensities are still higher than those of a 2-min exposure (Fig. 3-B_2).

Clearer insights can be obtained further from our isotope experiments, where SiH_3/Si(100) was exposed to deuterium (D) atoms at T_s = 140 K and 480 K. SiD_xH_{4-x} (x = 1-4), H_2, and D_2 desorption spectra following D-beam exposures are displayed in Figure 4. First, we discuss the results for T_s = 480 K. After a 3-min D atom exposure, ample β_1- and β_2-D_2 desorbed in subsequent TPD, while only trace amounts of H_2, HD (not shown) and SiD_xH_{4-x} (x = 1-4) desorbed. The only desorbing species during TPD following a 12-min D exposure at T_s = 480 K was D_2 with a β_2-to-β_1 desorption intensity ratio of 1:3, which is characteristic of H_2 TPD from an ideal 1.33-ML H/Si(100)3x1 surface (not shown). Neither proton-containing species (H_2, HD, SiH_xD_{4-x}) nor silane (SiD_4) was detected in TPD following extended D-atom exposures at T_s = 480 K. No SiD_4(g) desorption was detected, within our QMS detection limit, in post-D beam TPD until T_s was lowered to 300 K [7]. These results, together with the rapid decay of real-time SiH_4(g) evolution upon H atom exposure (Fig. 2), clearly suggest that thermal D atoms can readily break Si-Si dimer bonds to form SiD(a) and SiD_2(a), but not Si-Si backbonds to form SiD_3(a): $SiD_2(a) + D(g) \rightarrow SiD_3(a)$, at elevated substrate temperatures [2b]. While SiD_2(a) formation requires breakage of relatively weak (due to strain) dimer bonds, SiD_3(a) formation requires the stronger Si-Si backbond scission. Si-D bonds are stronger than Si-Si bonds [17]. In spite of the thermodynamically favored Si-D bond formation, an increased steric hindrance due to an additional Si-H bond is apparently too large for SiD_3(a) to be formed at elevated temperatures. The thermal stability of SiD_3(a) is very coverage-dependent and SiH_3(a) becomes unstable at T_s > 300 K when the SiD_3(a) coverage is low [7, 9, 16]. Boland [2] observed no etching from either Si(100)2x1:H or Si(100)3x1:H at T_s > 300 K. Only Si(100)1x1:H, a dihydride- and trihydride-covered structure realized by low-temperature (< 300 K) stabilization, was found to be susceptible to etching [2]. This is consistent with our results that the critical reaction step for etching, $SiD_2(a) + D(g) \rightarrow SiD_3(a)$, proceeds extensively only at temperatures below 300 K. SiD_3(a) formation must precede for the etching reaction to occur: $SiD_3(a) + D(g) \rightarrow SiD_4(g)$. Although adsorbed SiH_3(a) from Si_2H_6 is more efficiently abstracted by thermal H atoms with increasing temperature [7], SiH_3(a) generation via Si-Si backbond scission and Si-H bond formation becomes inefficient at T_s > 300 K. Thus, etching can not occur extensively at elevated temperatures.

We now turn to the interesting TPD features following D-atom exposures at T_s = 140 K. With a 3-min D atom exposure, H_2 desorption was mostly diminished, while strong (especially β_2) D_2 desorption, very similar to the H_2 TPD from SiH_3/Si(100) without H atom exposure (see Fig. 3), occurred. Strong silane desorption, mostly silanes with a high deuterium content, SiD_xH_{4-x}, including the fully deuterated SiD_4, was detected at 610

K just before the β_2-H_2 desorption at 670 K. Note the relative intensities of 32-amu TPD signal (approximately 1-to-50 ratio) for 3-min D exposure at T_s = 480 K and 12-min D exposure at T_s = 140 K. By referring to the relative cracking intensities determined *in-situ* by back-filling with SiH_4, it was estimated that D account for more than 95% of the total (H+D) balance on the surface. Similar results, with small further increases of β_2-H_2 peak and high-mass silane fragment ion signals, were obtained with extended D exposures (see Fig. 3). These TPD results, along with the continuous evolution of gas-phase SiD_4 (m/e = 36) under continuing D atom exposure at T_s = 140 K, clearly indicate that SiD_3(a) species are continuously generated on Si(100) from the concomitant Si-Si backbond scission and Si-D bond formation, while some newly formed SiD_3(a) are being abstracted away at the same time. Once SiD_3(a) is depleted, the surface will be quickly replenished with newly formed SiD_3(a). Thus, a steady-state surface coverage of SiD_3(a), determined by D flux and substrate temperature, is established. This steady-state amount of SiD_3(a) remaining on the surface will be reflected in SiD_4 and D_2 desorption in subsequent TPD. Evidently, silicon etching continues at T_s = 140 K, but not at T_s = 480 K, in this manner.

Olander et al. [18] reported a rapidly decreasing probability of volatile SiH_4(g) formation by atomic H from Si(111) with increasing T_s from 300 K to 1000 K. At T_s below room temperature, SiH_3(a) formation and thermal desorption as SiH_4 during TPD from a disproportionation reaction, $SiH_3(a) + SiH_x(a) \rightarrow SiH_4(g)$, at $T_s \sim$ 200 K were observed by Cheng et al. [16]. Direct abstraction reaction, $SiH_3(a) + H(g) \rightarrow SiH_4(g)$, from porous silicon surfaces at 133 K was dismissed recently by Glass, Jr., et al. [6] from their inability to decrease SiH and SiH_2 infrared absorption intensities under extensive D atom exposure. They concluded that the conversion of lower hydrides, SiH and SiH_2(a), to the trihydride SiH_3(a) was driven by thermal H atoms, but the reaction stopped once SiH_3(a) was formed at T_s = 133 K, SiH_3(a) desorbing as SiH_4(g) only by thermal heating up to 213-298 K as a result of the above-mentioned disproportionation reaction [6]. Watamori et al. [19] speculated formation and retention of SiH_3(a) and SiH_4(a) species on Si(100) by a thermal H atom beam at T_s = 100 K on the basis of the abnormally high total surface hydrogen coverage, $\theta_H \sim$ 4ML, estimated from their elastic recoil detection analysis. Either retention of SiH_4 on the surface at 100 K [19] or the lack of direct SiH_4(g) formation at 133 K [6] would inhibit continuous etching at these cryogenic temperatures. However, the surface retention of SiH_4 in a physisorbed state is not likely, considering the very low multilayer CH_4 desorption temperature of 40 K [20]. Therefore, our direct detection of gas-phase SiH_4(g) evolution during H atom exposure at substrate temperatures below room temperature, which is a direct evidence for H-atom induced Si etching, provides solid evidence for efficient silicon etching by atomic H at lowered substrate temperatures.

We now discuss briefly mechanistic aspects of the direct reaction of SiH_3(a) with thermal H atoms. Recently, a number of studies showed that direct abstraction of adsorbate species by gas-phase thermal H atoms can occur readily for H/Si(100) [22-23], H(D)/Cu(111) [24], H(D)/Ni(100) [25], CH_3I/Cu(100) [21], D/Ni(110) [26], and D-CH_3I/Ni(100) [27]. 'Hot-atom' mechanism, in which the impinging gas-phase H atoms are trapped in the surface potential but not fully relaxed prior to reaction, has been evoked to explain the experimental observations, in addition to Eley-Rideal (ER) mechanism, in which the reaction occurs without energy transfer to the surface prior to reaction. The strongly T_s-dependent rate of SiH_3 abstraction by atomic H from Si(100), as shown in Figure 1, is interesting in light of detailed reaction dynamics. The formation of SiH_3(a) by thermal H atoms is favored at tow temperatures, while its removal by thermal H atoms is facilitated at elevated temperatures. While our results suggest lowered temperatures (< 300 K) for efficient silicon etching, optimum conditions for efficient etching, i.e., SiH_3(a) abstraction efficiency, will be set by the steady-state coverage of SiH_3(a), which is determined by two opposing parameters, SiH_3(a) abstraction and SiH_3(a) stability, favoring high and low temperatures, respectively.

In summary, we have shown that the rate of Si(100) etching by thermal H atoms can be greatly enhanced by lowering substrate temperatures below room temperature. We present our direct detection of real-time gas-phase evolution of the etching product, $SiH_4(g)$, during H atom exposure by using a quadrupole mass spectrometer.

Acknowledgements:

This work was supported in part by the research grants from Kyung Won University (1997) and the Korea Research Foundation (KRF).

References:

[1] C. M. Greenlief and M. Armstrong, J. Vac. Sci. Tech. B 13 (1995) 1810.
[2] (a) J. J. Boland, Phys. Rev. B 44 (1991) 1383.
(b) J. J. Boland, Surf. Sci. 261 (1992) 17.
[3] M. Otobe, M. Kimura and S. Oda, Jpn. J. Appl. Phys. Pt. I. 33 (1994) 1442.
[4] K.-H. Hwang, E. Yoon, K.-W. Whang and J. Y. Lee, J. Electrochem. Soc. 144 (1997) 335.
[5] H. N. Wanka and M. B. Schubert, J. Phys. D. 30 (1997) L28.
[6] J. A. Glass, Jr., E. A. Wovchko and J. T. Yates, Jr., Surf. Sci. 348 (1996) 325.
[7] S. K. Jo, B. Gong and J. G. Ekerdt, to be published.
[8] R. Imbihl, J. E. Demuth, S. M. Gates and B. A. Scott, Phys. Rev. B 39 (1989) 5222.
[9] S. M. Gates, C. M. Greenlief, and D. B. Beach, J. Chem. Phys. 93 (1990) 7493.
[10] D.-S. Lin, E. S. Hirschorn, T.-C. Chiang, R. Tsu, D. Lubben and J. E. Greene, Phys. Rev. B 45 (1992) 3494.
[11] Y. Wang, M. J. Bronikowski, R. J. Hamers, Surf. Sci. 311 (1994) 64.
[12] C. M. Greenlief and M. Armstrong, J. Vac. Sci. Tech. B 13 (1995) 1810.
[13] F. Hirose and H Sakamoto, Appl. Surf. Sci. 107 (1996) 75.
[14] L. A. Okada, M. L. Wise and S. M. George, Appl. Surf. Sci. 82-83 (1994) 410.
[15] D. Lubben, R. Tsu, T. R. Bramblett and J. E. Greene, J. Vac. Sci. Tech. A9 (1991) 3003.
[16] C. C. Cheng and J. T. Yates, Jr., Phys. Rev. B 43 (1991) 4041.
[17] P. M. Agrawal, D. L. Thomson, and L. M. Raff, J. Chem. Phys. 92 (1990) 1069.
[18] (a) D. R. Olander, M. Balooch, J. Abrefah and W. J. Siekhaus, J. Vac. Sci. Tech. B (1987) 1404.
(b) J. Abrefah and D. R. Olander, Surf. Sci. 209 (1989) 291.
[19] M. Watamori, M. Naitoh, H. Morioka, Y. Maeda and K. Oura, Appl. Surf. Sci. 82 (1994) 417.
[20] J. Yoshinobu, H. Ogasawara and Maki Kawai, Surf. Sci. 363 (1994) 234.
[21] Y.-S. Park, J.-Y. Kim and J. Lee, Surf. Sci. 363 (1996) 62.
[22] D. D. Koleske, S. M. Gates and B. Jackson, J. Chem. Phys. 101 (1994) 3301.
[23] P. Kratzer, J. Chem. Phys. 106 (1997) 6752.
[24] C. T. Rettner and D. J. Auerbach, Surf. Sci. 357-358 (1996) 602.
[25] Th. Kammler, J. Lee and J. K?ers, J. Chem. Phys. 106 (1997) 7362.
[26] G. Eilmsteiner, W. Walkner and A. Winkler, Surf. Sci. 352-354 (1996) 263.
[27] Th. Kammler and J. K?ers, J. Chem. Phys. 107 (1997) 287.