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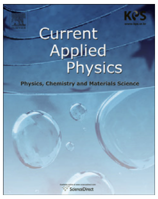

Erosion dynamics of faceted pyramidal surfaces

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ABSTRACT

Anisotropic etching using aqueous KOH was carried out on n-type Si (100) surfaces. Atomic force microscopy (AFM) was employed to study the etched surfaces for different times. The AFM data was investigated employing dynamic scaling theory formalism. The rms roughness was found to increase with etching time except for 2 min. Roughness exponent calculations revealed that the surface became locally rougher for etching times upto 4 min after which the local roughness decreases and the system ceases to obey Family-Vicsek scaling condition. A high value (~1.882) of the growth exponent β indicates a rapid out-of-plane growth as well as faceting of the pyramids on the surface. The surface roughness evolution is found to follow an eroding system characterized by quenched random fluctuations. Finally, a vacuum annealing study done up to 1000 °C reveals that atoms having lower binding energies tend to get dislodged and diffuse from about 900 °C.

1. Introduction

Anisotropic silicon surface texturing is a major processing step in the photovoltaic (PV) industry which helps in reducing the front reflection [1]. This was earlier achieved by the method of photolithography followed by anisotropic etching to obtain upright or inverted pyramidal structures [1–3]. The down side for such a process was the turn around time and the process cost. This can considerably be reduced by texturing silicon surfaces with alkaline etchants like potassium hydroxide (KOH), sodium hydroxide (NaOH), tetra-methyl ammonium hydroxide (TMAH) etc. thereby producing random pyramidal structures on surfaces [4–8]. These random pyramid arrays exhibit reduced reflectivities which is a major criterion for c-Si PV cell manufacture [9]. In addition to their use in the PV industry, these surfaces are recently finding their applicability as surface-enhanced Raman scattering (SERS) substrates. These low cost manufactured substrates have suitable microstructure for acquiring stable, sensitive and reliable SERS signals [10]. A recent report has also used these silicon pyramidal substrates as low-cost broadband silicon plasmonic Schottky detector with high responsivity and improved signal to noise ratio operating in the sub-bandgap regime [11]. Besides these, this kind of texturing has a wide range of applications in micro-electromechanical systems (MEMS) including pressure, acceleration, angular rate, gas-flow sensors etc., to mention only a few [12]. As a result, this route of silicon surface texturing has found considerable importance in the industry.

In addition to the low cost involved, another advantage of this method is that it provides smooth defect-free surfaces with no physical damage to the bulk. However, with the reduction of feature sizes of modern day devices, the roughness of the etched surface has begun to play an important role. Hence, a meticulous understanding of the production conditions is required which will considerably improve our knowledge of the mechanisms that lead to the specific morphological structures on the etched surfaces. Efforts have been made both by experimental and theoretical groups to characterise and understand the surface morphology under wet etch conditions [12]. The silicon etching rate has been found to depend on the crystallographic plane, etchant concentration, temperature, sonication [12–14] etc.

Anisotropic etching of silicon using aqueous KOH solution has been reported by a number of researchers [12]. Several authors have tried to explain various mechanisms owing to the formation of hillocks on these surfaces. These include pseudomasking of hydrogen bubbles [15–17], reaction products [15,18,19], SiO_2 precipitates [20–22], impurity micromasks [23] etc. Although it is now agreed that the hillocks are nucleated by some sort of a stabilizing mechanism, however, the nature of the mechanism still remains to be understood [12]. Quantitative studies in this direction have been done by employing scaling theory concepts which analyses the entire surface in terms of surface fluctuations [24–26]. This has

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helped researchers in understanding surface physical or chemical growth or erosion mechanisms governing topography [24,27–30]. Under this formalism, a rough surface can be described by a surface height profile $h(x,t)$, where h denotes the surface height at a position x on the surface at time t. A standard measure of the surface roughness (also referred to as the interface width) is the root-mean-square (rms) value w, expressing the variation of the height function $h(i,t)$ over a two dimensional substrate with linear size L [31],

$$w(L,t) = \sqrt{\frac{1}{L}\sum_{i=1}^{L}\left[h(i,t) - \bar{h}(t)\right]^2} \tag{1}$$

where h(i,t) is the height of coloumn at time t and the mean height is given by

$$\bar{h} = \frac{1}{L}\sum_{i=1}^{L} h(i,t) \tag{2}$$

To monitor the roughening process quantitatively we measure the interface width as a function of time. A typical plot of the time evolution of interface width has two regions separated by a cross over time t_x:

(i) The width initially increases as a power law of time [32] where the growth exponent β characterizes the time dependent dynamics of the roughening process.

$$w(L,t) \sim t^{\beta} \tag{3}$$

(ii) This is followed by a saturation regime during which the width ceases to increase any further and reaches a saturation value w_{sat}. As L increases, the saturation width w_{sat} increases as well and the dependence follows a power law,

$$w_{sat}(L) \sim L^{\alpha}[t \gg t_x] \tag{4}$$

The exponent α, called the roughness exponent is a second critical exponent that characterizes the roughness of the saturated interface. A smaller value of α implies a rougher local surface where α lies between 0 and 1. A local surface generally refers to a length scale smaller than the correlation length of the surface under consideration. An estimate of the correlation length can be obtained from the knee position of a w versus L curve.

Some research groups have tried to understand the mechanism of alkaline etching using scaling concepts. Self-affine behaviour of NaOH etched crystalline Si surfaces under various conditions have been studied by Kleinke and co-workers [24]. In this study, a small drop of KOH was used as an etchant accounting for an increasing concentration gradient. This in turn generated a surface interfacial turbulence. Their results are indicative of the fact that such surfaces under saturated ambient conditions can be explained using Kessler-Levine-Tu (KLT) percolation model [33]. KOH etching of silicon was studied experimentally and modelled using Monte Carlo methods by Aldao and his group [25]. They had demonstrated that the hillock formation mechanism was a consequence of site-dependent detachment probablity on surface morphology. Their results showed both conventional and anomalous scaling depending on the stability of the hillock apices. However, the evolution of surface topography remained to be investigated in all the above studies.

In this paper, we study the dynamics of erosion for KOH etched n-type Si(100) surface using dynamic scaling theoretical concepts. Etching of the Si surface was performed with 7% aqueous KOH solution at 80 °C without sonication. In the current study, a low KOH concentration solution was taken in order to achieve a high pyramid denisty [20,34]. It is found that the onset of pyramid formation takes place from about 1 min of etching. The evolution of morphology clearly shows that the Family-Vicsek [35] scaling concept is not valid for the present scenario. Moreover, employing percolation concepts we show that the etching phenomenon follows a surface percolation model. The effect of annealing on the pyramidal facets was also studied by varying temperature in the range 600 °C to 1000 °C.

2. Experimental

Si samples (10 × 10 mm) were obtained from an n-type Si(100) single crystal wafer. The samples were cleaned and rinsed using deionized water ($\rho \sim 18.2 M\Omega$) from a Milli-Q Gradient water purification system (Millipore). All the samples were precleaned by sonicating with isopropyl alcohol for 15 min each before etching. SiO_2 layer on the n-Si(100) wafers was removed by dipping it in a solution of HF and deionized water in ratio 1:10 (by volume) for 30s along with sonication. After SiO_2 removal, the samples were rinsed thoroughly with DI water and dried thereafter. The samples were thereafter etched in 7% aqueous KOH at 80 °C without sonication for 1, 4, 8, 15, 30, 45 and 60 min. In the current study, a low KOH concentration solution was taken in order to achieve a high pyramid denisty [20,34]. Atomic force microscopy (AFM) imaging of the samples were done in the tapping mode at a slow scan rate of 0.3–0.5 Hz. All the images acquired using AFM had a resolution of 512 × 512 lines. Scanning electron microscopy (SEM) using a JEOL JSM 6610V instrument was performed for samples of higher etching times since heights of the large pyramids formed were beyond the z-range (~5 μm) of the AFM scanner. A JEOL JFC-1600 gold coating unit was used for making the samples conducting. SEM imaging was done at 15 keV. To study the effect of annealing on the pyramidal facets vacuum annealing experiments were done on the prepared samples at 600 °C, 700 °C, 800 °C, 900 °C and 1000 °C for 3 min using a Nabertherm vacuum annealing set up at a pressure of approximately 10^{-5} mbar.

3. Results and discussions

Fig. 1 shows AFM images for the evolution of morphology of the KOH etched n-Si surfaces. The pristine Si surface prior to etching is shown in Fig. 1a. After etching for about 1 min, dense mound-like structures are observed on the eroded surface (Fig. 1b). At 2 min, distinct but relatively sparse pyramids appear. After this, the pyramids increase both in size and density till 15 min as seen from Fig. 1. After 15 min of etching the height of the pyramids go beyond the z-range of the AFM scanner. Hence, AFM imaging of the samples could not be performed for samples having etch times more than 15 min. It is noteworthy here that for the 15 min etched surface apices of few pyramids appeared cut off due to scanner limitation (Fig. 2). The topographic fate of these surfaces were however evident from the SEM images (Fig. 3). It is observed from these images that high index facets [20] form on the (111) planes of the Si pyramids as the etching time increases. It is further observed that what appeared to be small mounds on the 1 min etched Si surface from the AFM image are actually small pyramids as seen from a magnified SEM image. At this time, the facets on the pyramids start forming on the Si surfaces although the process does not get completed. Well formed and dense pyramids appear only after 15 min of etching.

Dynamical scaling approach utilizing AFM data was employed to study the quantitative evolution of the roughening phenomenon. The variation of the interface width (w) or surface roughness was