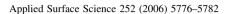
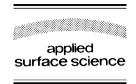


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## X-ray photoelectron spectroscopy characterization of oxidated Si particles formed by pulsed ion-beam ablation

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> Received 2 April 2005; received in revised form 13 July 2005; accepted 27 July 2005 Available online 26 September 2005

#### Abstract

X-ray photoelectron spectroscopy (XPS) is used to probe oxidation states of Si species in particles deposited using a pulsed ion-beam evaporation method. The effects of He ambient gas, ion beam intensity and post-treatments on the oxides composition and oxygen content have been studied. It is found that presence of He ambient gas led to a profound oxidation of Si species as compared to that prepared in vacuum at the same ion-beam ablation energy, i.e. both increase of  $SiO_2$  component and oxygen concentration in the oxides coverage. The deposition in He also resulted in an increase of oxygen concentration even under lower ablation intensity, but a higher Si suboxides concentration. It is revealed that the reaction between Si and O was controlled by the ion beam intensity (temperature of Si plasma) and the gas ambient (collision probability of Si and O species). The difference in structure of oxide layers for samples obtained under various conditions is discussed based on the results of XPS analyses.  $\bigcirc$  2005 Elsevier B.V. All rights reserved.

Keywords: Silicon oxides; Pulsed ion-beam evaporation; Ablation plasma; X-ray photoelectron spectroscopy

#### 1. Introduction

Si-based materials including Si oxides and crystalline Si are of great interests in many industrial applications such as in optoelectronics, microelectronics and biomedical areas, due to their unique

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electronic and optical properties [1–4]. A pulsed ionbeam deposition method, i.e. intense pulsed ion-beam evaporation (IBE), has been recently applied to study this category of materials, e.g. polycrystalline Si thin films and photoluminescent Si thin films/nanoparticles [5–7]. Among material syntheses by ablation plasma generated from a target material, this pulsed ion-beam approach is similar to pulsed laser deposition (PLD). Note that the pulsed ion-beam technique possesses several noticeable advantages over its laser

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counterpart, i.e. high efficiencies of beam generation and beam-target interaction, and large beam size that ensure high-rate, large-area deposition as well as surface modifications [8]. These characteristics are very attractive for practical applications. For instance, polycrystalline Si thin films and nanoparticles can be readily prepared in vacuum at room temperature [5–7] that is usually unavailable with PLD method and thus introduction of ambient gases, heating of substrates and/or high-temperature post-annealing were elaborated to improve the poor crystallinity of as-deposited Si species by PLD [9–11]. In our previous study, it was found that Si oxides played a major role in photoluminescence properties of IBE-prepared Si samples and the PL has been ascribed to defect bands in the Si oxide layers [7]. However, detailed dynamics of Si deposition by IBE is still not clear. In this paper, we present the study of oxidation phenomena of Si samples produced by the IBE process, using X-ray photoelectron spectroscopy (XPS) to probe the oxidation state of Si in samples prepared under different conditions, i.e. deposition in a vacuum, He gas ambient or changing the ion-beam ablation energy, and post-treatments in air (aged) and oxidation in O2 gas at an elevated temperature (post-oxidized), respectively. Oxides composition and oxygen concentration have been compared for the different samples and main factors influencing the oxidation process is subsequently discussed.

#### 2. Experimental

The deposition of Si samples was carried out in an intense pulsed ion-beam apparatus, ETIGO-II. The experimental scheme has been described in details previously [7]. A pulsed ion beam ( $\sim$ 70 ns, 70 or 40 J/cm²) was used to ablate a sintered polycrystalline silicon target, and subsequently the ablated Si species were deposited onto a lightly p-doped (1 0 0) Si substrate (10 mm  $\times$  10 mm, washed in a HF solution). The deposition was accomplished by a single shot of ion-beam ablation in a vacuum at  $2 \times 10^{-2}$  Pa or with a He ambient gas at 532 Pa, respectively. Prior to each one-shot deposition, the target was mechanically polished with 220# SiC papers to obtain a fresh surface and then loaded into the deposition chamber immediately to minimize their natural oxidation in air.

All the Si samples were located at a target–substrate distance ( $d_{TS}$ ) of 60 mm and in the center position of ablation plasma, where a deposited layer thickness is typically in the range of 100–200 nm depended on the varied parameters.

Measurements of XPS were performed on a JEOL JPS-100SX equipment using an unmonochromatized Mg K $\alpha$  ( $h\nu = 1253.6 \text{ eV}$ ) X-ray source with a 0.8- $\mu$ m Al foil filter. The X-ray source was run at 200 W (10 kV and 20 mA). The X-ray beam with a circular cross-section area ( $\sim 1 \text{ cm}^2$ ) was irradiated onto sample surface at an incident angle of 22.5° with respect to the surface plane and the photoelectrons were detected at 90° take off angle. Photoelectrons were analyzed with a concentric hemispherical analyzer at a pass energy of 30 eV. Under these conditions, the full widths at half-maximum (FWHM) of the Si 2p peaks of clean Si and SiO<sub>2</sub> (quartz) reference samples were about 1.4 and 2.0 eV, respectively. Furthermore, Ar<sup>+</sup> sputtering (800 V, 19 mA,  $10^{-2} \text{ Pa}$ ) was applied to etch the samples for obtaining compositional information from the underlayers. The charging effects on the spectra have been corrected according to Au 4f<sub>7/2</sub> peak at 84.0 eV.

#### 3. Results and discussion

All the XPS curves were deconvoluted using Lorentzian–Gaussian functions after Shirley background subtraction. The peak fitting parameters are summarized in Table 1. A fitting process was conducted and optimized as follows. It was assumed that the intermediate oxidation states consist of  $\mathrm{Si}^{1+}$ ,  $\mathrm{Si}^{2+}$  and  $\mathrm{Si}^{3+}$  [12], i.e. Si suboxides  $\mathrm{SiO}_x$  (0 < x < 2) and the binding energy of  $\mathrm{Si}^{4+}$  is fixed to that of  $\mathrm{SiO}_2$  reference sample (103.4 eV). The spin-orbit splitting

Table 1
Summary of XPS spectra fitting results for Si samples prepared by IBF

Components	Peak position (eV)	FWHM (eV)
Si	$99.1 \pm 0.1 \ (99.3 \pm 0.05)^{a}$	1.38
Si <sub>2</sub> O	$100.1 \pm 0.05$	1.58
SiO	$101.2 \pm 0.05$	1.60
$Si_2O_3$	$102.35 \pm 0.05$	1.78
$SiO_2$	103.4	2.0

<sup>&</sup>lt;sup>a</sup> Values of samples after Ar<sup>+</sup> sputtering.

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