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Reactive sputtering deposition of SiO_2 thin films

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Abstract: SiO_2 layers were deposited in a UHV chamber by 1 keV Ar^+ ion sputtering from a high purity silicon target, using different values of the oxygen partial pressure (5×10^{-6}–2×10^{-4} mbar) and of the ion beam current on the target (1.67–6.85 mA). The argon partial pressure during operation of the ion gun was 1×10^{-3} mbar. The substrate temperature was held at 550 °C and the films were deposited to a thickness of 12.5–150 nm, at a rate from 0.0018–0.035 nm s^{-1}. Structural characterization of the deposited thin films was performed by Rutherford backscattering spectrometry (RBS analysis). Reactive sputtering was proved to be efficient for the deposition of silica at 550 °C, an oxygen partial pressure of 2×10^{-4} mbar (ion beam current on the target of 5 mA) or, at a lower deposition rate, ion beam current of 1.67 mA and an oxygen partial pressure of 6×10^{-5} mbar. One aspect of these investigations was to study the consumption of oxygen from the gas cylinder, which was found to be lower for higher deposition rates.

Keywords: SiO_2; thin films; reactive sputtering; RBS analysis.

INTRODUCTION

Silicon dioxide, in its various crystalline and amorphous phases, is a widely employed material in many technological domains. Two important aspects are currently related to SiO_2. It is well known that SiO_2 is the most important material in the field of microelectronics. On the other hand, SiO_2 is the basic material for the understanding of the behaviour of oxide glasses, used for storage of nuclear waste. Diffusion of components is a critical point in both cases. It was observed, for example, that silicon diffuses in an oxide film during thermal growth of SiO_2 on silicon substrates.[1–8] Concerning nuclear glasses, they are exposed to irradiation, which can influence many physical processes, one of the most important being self-diffusion.[9] To develop a new generation of materials (nano-electronics, nuclear glasses), a detailed understanding of the diffusion processes is essential.

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In order to study diffusion mechanisms and determine a general model for the ageing of nuclear glasses, stoichiometric SiO_2 thin films are required. Roma *et al.*[10] reported the first theoretical results for oxygen self-diffusion in SiO_2. They considered two distinct modes – the *closed mode*, when no exchange is possible with the atmosphere, and the *open mode*, when exchange with the atmosphere is allowed. To verify their results, special techniques are required to prepare the SiO_2 samples. Quantitative measurements of oxygen self-diffusion in SiO_2 is feasible in $Si(^{18}O)_2/Si(^{16}O)_2$ bilayer structures using secondary ion mass spectrometry (SIMS), which can clearly distinguish between these two isotopes. In this context, reactive ion beam sputtering offers advantages, such as precise control of the oxygen partial pressure during the deposition process.

The aim of this study was to determine the experimental conditions for SiO_2 deposition. The reactive ion beam sputtering technique from a pure Si target was applied for two obvious reasons: (*i*) it provides good control of the deposition parameters and of the layer thickness; and (*ii*) the rf sputtering technique would not be possible for the deposition of $Si(^{18}O)_2$ films, as such targets are not available. All experiments in this study were performed with $(^{16}O)_2$, because the use of $(^{18}O)_2$ would be too costly for these purposes. It was found that stoichiometric SiO_2 thin films could be produced by suitable adjustment of the deposition parameters.

EXPERIMENTAL

The SiO_2 layers were deposited by reactive ion beam sputtering in a UHV chamber. A schematic presentation of the installation is shown in Fig. 1. The base pressure in the deposition chamber was 4×10^{-9} mbar. A high purity silicon target (diameter 76 mm), which was bombarded with 1 keV Ar^+ ions, at a 45° incidence angle was used. The ion source was an ERIS type ion gun (Electrostatic Reflex Ion Source), irradiating an area of about 1 cm^2 of the target. The ion beam current on the target, I_t, was varied from 1.67 to 6.85 mA, using a constant argon (purity 99.996 %) partial pressure of 1×10^{-3} mbar. Different values of the oxygen $(^{16}O)_2$ (purity 99.995 %) partial pressure P_O, from 5×10^{-6} to 2×10^{-4} mbar, were applied for reactive deposition. Substrates of quasi-monocrystalline graphite (10 mm×10 mm) and silicon (15 mm×8 mm) were employed. Before deposition, the substrates were cleaned by standard chemical procedures. The substrates were placed parallel to the target, at a distance of 150 mm. During the deposition, they were maintained at 550 °C. The SiO_x films were deposited at a rate from 0.0018–0.035 nm s^{-1} to a thickness of 12.5–150 nm.

The thickness of the films was determined by a surface profilometer on the optically flat silicon substrates. Structural characterization of the films was performed by Rutherford backscattering spectrometry (RBS analysis) on quasi-monocrystalline graphite substrates, using a 1 MeV He^+ ion beam at normal incidence with the detector at 165° backscattering angle. The experimental spectra were analyzed with the program PERM, version 2003.02.[11,12]

RESULTS AND DISCUSSION

In a previous work,[13] reactive ion beam sputtering was shown to be efficient for the deposition of silica at 550 °C, using 1 keV Ar^+ ions with an oxygen partial pressure P_O of 2×10^{-4} mbar and an ion beam current on the target I_t of 5 mA.

The calculated layer thickness, using the bulk density of SiO_2, agreed with a value of 125 nm to that measured with a profilometer. This suggests a reasonable purity of the deposited layer.

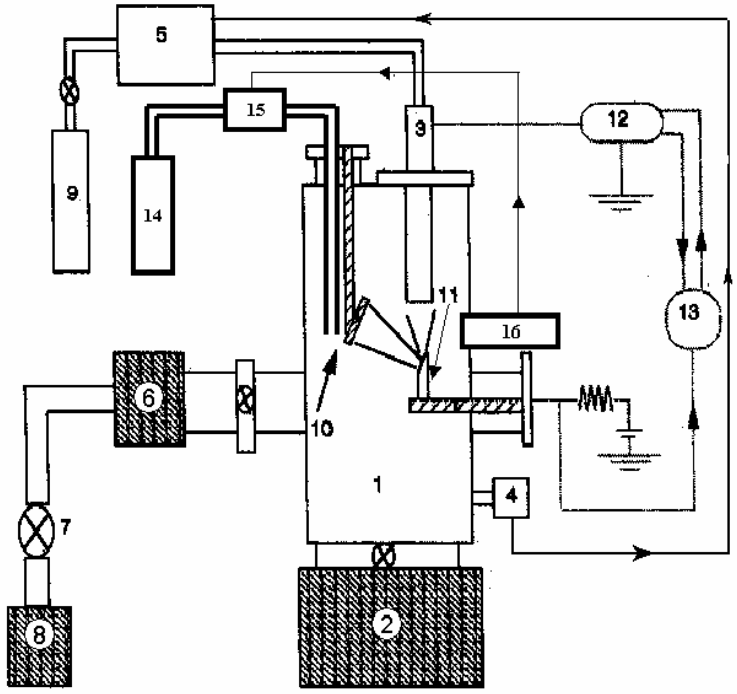

Fig. 1. Schematic presentation of the installation for reactive ion beam sputtering. 1– UHV chamber; 2– ionic pump; 3– ion guns; 4– vacuum gauge; 5– gauge and thermoregulated valve; 6– turbo pump; 7– auxiliary valve; 8– primary pump; 9– argon bottle; 10– sample-holder; 11– target-holder; 12– gun-supply; 13– computer; 14– oxygen bottle; 15– regulation valve; 16– quadrupole gas analyser.

The oxygen consumption during the deposition depended on two parameters for a given thin film thickness: the oxygen partial pressure and the ion beam current on the target. Lowering P_O, at a given I_t, reduced the gas consumption. Increasing I_t, and hence the deposition rate, at a given P_O, also reduced the consumption.

One aspect of these investigations was the precise control of the oxygen consumption from the cylinder. In order to reduce the oxygen consumption, a constant I_t of 5 mA at a reduced P_O of 5×10^{-6}, 2×10^{-5}, 5×10^{-5} and 1×10^{-4} mbar was applied. However, RBS analysis gave an under-stoichiometry of oxygen: $SiO_{0.25}$, $SiO_{0.35}$, $SiO_{0.6}$ and $SiO_{1.25}$, respectively.

Still attempting to reduce the oxygen consumption, an attempt was made to reach the stoichiometry using a higher I_t of 6.85 mA, keeping P_O at 2×10^{-4} mbar, but again an under-stoichiometry resulting in $SiO_{1.85}$ was observed.

The stoichiometry can be attained, under given circumstances, in two ways: increasing P_O and I_t (unrealizable in the present case because at high ion beam currents, the behaviour of the ion gun becomes unstable) and lowering P_O and I_t.

The SiO_2 stoichiometry of the layers was attained using a P_O of 6×10^{-5} mbar and an I_t of 1.67 mA. RBS Analysis of a sample produced using these parameters is presented in Fig. 2. In Fig. 2a, the experimental spectrum and the fit obtained by introducing Si, O and C in the PERM code can be seen. The extracted depth profiles, Fig. 2b, indicate a very uniform SiO_2 stoichiometry of the deposited layer. However, as the applied Ar^+ ion beam current on the target was lower, the deposition rate was much lower and, consequently, a layer of small thickness (12.5 nm) was obtained.

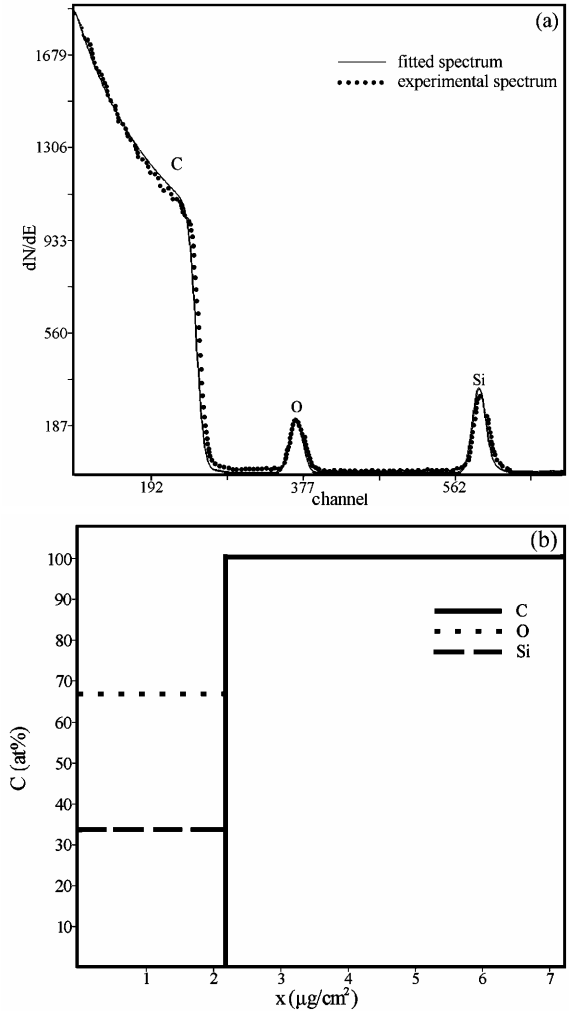

Fig. 2. RBS Analysis of a SiO_x/graphite sample deposited at 550 °C, $P_O = 6\times10^{-5}$ mbar and $I_t = 1.67$ mA: (a) experimental and fitted spectra and (b) extracted concentration profiles.

Therefore, stoichiometric SiO_2 thin films can be obtained under the conditions: a P_O of 2×10^{-4} mbar with an I_t of 5 mA (deposition rate ≈ 0.029 nm s^{-1}), or a P_O of 6×10^{-5} mbar with an I_t of 1.67 mA (deposition rate 0.0018 nm s^{-1}). The advantage deposition under the former conditions is a much higher deposition rate, while the stability of the ion beam current is better during deposition under the latter conditions.

To calculate the oxygen consumption in the cases of the high and low deposition rates, the pressure loss in the employed $(^{16}O)_2$ cylinder after one hour of deposition was measured and the maximal thickness of the layers that could be obtained using the total available amount of oxygen was calculated. The results show that, with the available quantity of oxygen, 2100 nm of an SiO_2 thin film can be obtained under the conditions: $P_O = 2\times10^{-4}$ mbar and $I_t = 5$ mA, while for $P_O = 6\times10^{-5}$ mbar and $I_t = 1.67$ mA, only 400 nm of SiO_2 would be obtain. This infers that working with a higher I_t and a higher P_O results in lower oxygen consumption.

CONCLUSIONS

It has been shown that the reactive ion beam sputtering technique is efficient for the deposition of silica thin films at 550 °C, using an oxygen partial pressure of 2×10^{-4} mbar and an ion beam current on the target of 5 mA, or at a lower deposition rate, using ion beam current of 1.67 mA and oxygen partial pressure of 6×10^{-5} mbar. The oxygen consumption was calculated to be smaller under a 2×10^{-4} mbar partial pressure. The experimental method enables the oxygen consumption to be controlled precisely during the deposition process. RBS Analysis showed that the obtained SiO_2 thin films were stoichiometric.

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И З В О Д

ДЕПОНОВАЊЕ ТАНКИХ СЛОЈЕВА SiO_2 МЕТОДОМ РЕАКТИВНОГ ЈОНСКОГ РАСПРАШИВАЊА

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Танки слојеви SiO_2 депоновани су методом реактивног јонског распрашивања, при различитим вредностима парцијалног притиска кисеоника (5×10^{-6}–2×10^{-4} mbar) и при различитим вредностима струје на мети (1,67–6,85 mA). За распрашивање мете од силицијума коришћен је јонски сноп аргона, енергије 1 keV. Парцијални притисак аргона, при свим депоновањима, износио је 1×10^{-3} mbar. Температура подлоге, при свим депоновањима, износила је 550 °C. Дебљина депонованих слојева кретала се у интервалу од 12,5 до 150 nm, а брзина депоновања у интервалу од 0,0018 до 0,035 nm s^{-1}. Структурна карактеризација слојева извршена је спектрометријом Радерфордовог повратног расејања (RBS анализа). Показано је да

се танки слојеви SiO_2 могу добити методом реактивног јонског распрашивања при вредности парцијалног притиска кисеоника 2×10^{-4} mbar (струја на мети 5 mA) и 6×10^{-5} mbar (струја на мети 1,67 mA). Један од циљева истраживања био је испитивање потрошње кисеоника при депоновању и нађено је да је она мања при већим брзинама депоновања.

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