High-dose MeV electron irradiation of Si-SiO₂ structures implanted with high doses Si⁺

S Kaschieva¹, Ch Angelov² and S N Dmitriev³

¹G. Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences,
72 Tzarigradsko Chaussee, 1784 Sofia, Bulgaria
²Institute for Nuclear Research and Nuclear Energy, Bulgarian Academy of Sciences,
72 Tzarigradsko Chaussee, 1784 Sofia, Bulgaria
³Flerov Laboratory of Nuclear Reactions, Joint Institute of Nuclear Research,
141980 Dubna, Moscow Region, Russia

E-mail: kaschiev@issp.bas.bg

Abstract. The influence was studied of 22-MeV electron irradiation on Si-SiO₂ structures implanted with high-fluence Si⁺ ions. Our earlier works demonstrated that Si redistribution is observed in Si⁺-ion-implanted Si-SiO₂ structures (after MeV electron irradiation) only in the case when ion implantation is carried out with a higher fluence \(10^{16} \text{ cm}^{-2}\). We focused our attention on the interaction of high-dose MeV electron irradiation \(6.0 \times 10^{16} \text{ cm}^{-2}\) with n-Si-SiO₂ structures implanted with Si⁺ ions (fluence \(5.4 \times 10^{16} \text{ cm}^{-2}\) of the same order magnitude). The redistribution of both oxygen and silicon atoms in the implanted Si-SiO₂ samples after MeV electron irradiation was studied by Rutherford back-scattering (RBS) spectroscopy in combination with a channeling technique (RBS/C). Our results demonstrated that the redistribution of oxygen and silicon atoms in the implanted samples reaches saturation after these high doses of MeV electron irradiation. The transformation of amorphous SiO₂ surface into crystalline Si nanostructures (after MeV electron irradiation) was evidenced by atomic force microscopy (AFM). Silicon nanocrystals are formed on the SiO₂ surface after MeV electron irradiation. The shape and number of the Si nanocrystals on the SiO₂ surface depend on the MeV electron irradiation, while their size increases with the dose. The mean Si nanocrystals height is 16-20 nm after irradiation with MeV electrons at the dose of \(6.0 \times 10^{16} \text{ cm}^{-2}\).

1. Introduction

The influence of high-energy electron irradiation on high-fluence ion-implanted Si-SiO₂ structures was studied by Klimenkov et al. The authors showed that high-energy electron irradiation can cause moving of clusters or atoms on the surface [1], and that, as an alternative to the heat treatment, the cluster formation can be stimulated by high-energy electron irradiation [2]. The behavior of a silicon solar cell before and after 1 MeV electron irradiation with \(10^{14} \text{ cm}^2\), \(10^{15} \text{ cm}^2\) and \(10^{16} \text{ cm}^2\) has also been studied. As the dose was raised, a slight increase was observed in the short circuit current, accompanied by a decrease in the maximum output power [3]. Previously, we compared the interface states generated by 20 MeV electrons in n- and p-type Si-SiO₂ structures (with an equal amount of

---

² To whom any correspondence should be addressed.
oxide). We demonstrated that most of the radiation defects created could be related to the main impurities in the Si substrate – phosphorus or boron vacancy complexes – for the structures based on n- and p-type Si wafers, respectively [4]. In another study, we also showed that the total concentration of electron irradiation-induced defects at the Si-SiO\textsubscript{2} interface of the structures is larger in the case of an n-type Si wafer [5]. Our study on MeV electron irradiation of the implanted Si-SiO\textsubscript{2} structures (with different fluence of \textsuperscript{Si} ions) demonstrated redistribution of silicon (after MeV electron irradiation) only in the case when the ion implantation is carried out with a higher (10\textsuperscript{16} cm\textsuperscript{-2}) fluence [6]. The aim of the present work was to study the influence of high-dose MeV electron irradiation (6.0×10\textsuperscript{16} cm\textsuperscript{-2}) on n-Si-SiO\textsubscript{2} structures implanted with Si ions with fluence of the same order of magnitude (5.4×10\textsuperscript{16} cm\textsuperscript{-2}). The redistribution of both oxygen and silicon atoms in \textsuperscript{Si} ion implanted Si-SiO\textsubscript{2} samples after 22-MeV electron irradiation was studied by Rutherford back-scattering spectroscopy. A transformation was observed of the amorphous SiO\textsubscript{2} surface of the implanted samples into crystalline Si nanostructures as a result of the MeV electron irradiation.

2. Experimental details
Si-SiO\textsubscript{2} structures were prepared on n-type Si <100> wafers with a resistivity of 6.2 Ω cm. A SiO\textsubscript{2} layer of 22-nm thickness was grown in dry ambient of O\textsubscript{2} + 6% HCl at 950 °C. The samples were then cooled in the same ambient at a rate of 1°C s\textsuperscript{-1}. The oxide thickness was determined by ellipsometry. Silicon ions with an energy of 12 keV and a dose of 5.4×10\textsuperscript{16} cm\textsuperscript{-2} were implanted through the SiO\textsubscript{2} layer of the samples. The values chosen of the oxide thickness and silicon-ion energy ensured that the maximum impact of the implantation damage should take place in the Si bulk wafer very close to the Si-SiO\textsubscript{2} interface.

The non-implanted and Si\textsuperscript{+}-implanted Si-SiO\textsubscript{2} samples were irradiated by 22-MeV electrons at a dose of 6.0×10\textsuperscript{16} cm\textsuperscript{-2}. The irradiation was carried out on the MT-25 Microtron in the Flerov Laboratory of Nuclear Reactions of the Joint Institute for Nuclear Research, (FLNR, JINR) Dubna, Russia. The distance between the Microtron’s window and the samples was 150 mm. The irradiation was performed under a pressure of about 1×10\textsuperscript{2} Pa. The sample temperature was controlled during the entire process of electron irradiation and kept close to room temperature.

3. Results and discussion
Figure 1 and figure 2, respectively, present the results of the RBS and AFM measurements of Si-SiO\textsubscript{2} structures implanted with 12-keV \textsuperscript{Si} ions (fluence 5.4×10\textsuperscript{16} cm\textsuperscript{-2}) before and after 22-MeV electron irradiation (6.0×10\textsuperscript{16} cm\textsuperscript{-2}). The RBS was used to determine accurately the impurity distribution and elemental areal density in the thin SiO\textsubscript{2} film. The RBS/C spectra demonstrated an additional widening and increasing of the main silicon peak and a decrease of the oxygen peak height (figure 1 a) after MeV electron irradiation. This means that the MeV electron irradiation increases the silicon concentration and decreases the oxygen concentration at the Si-SiO\textsubscript{2} sample surface. We assume that Si atoms in the wafers are displaced from their normal position in the
semiconductor lattice during MeV electron irradiation, so that the formation takes place of vacancies and interstitial atoms. The implanted Si+ ions and the displaced oxygen and silicon atoms are able to migrate; we observed their redistribution resulting from the MeV electron irradiation. In addition, the radiation defects already created by the Si+ ion implantation interact with those subsequently generated by MeV electrons, thus creating suitable conditions for stimulation of oxygen radiation-enhanced diffusion in the samples. As the free oxygen is mobile, its migration through the oxide is observed. The oxygen concentration decreases, but the silicon concentration increases in the electron irradiated structure, as can be seen in figure 1 a.

The random RBS spectrum demonstrates higher silicon concentration in the amorphous part of the sample (SiO$_2$). The free oxygen (generated by MeV electrons) penetrates through the oxide and an additional amorphous silicon layer is created on the SiO$_2$ surface. Part of the Si in the amorphous SiO$_2$ surface of the implanted samples is transformed into crystalline Si nanostructures as a result of the MeV electron irradiation (figure 1 b).

The formation of nanostructures in the SiO$_2$ was studied by atomic force microscopy. The microscopic morphology of the SiO$_2$ surface structure implanted with Si$^+$ ions with a dose of $5.4 \times 10^{16}$ cm$^{-2}$ is presented in figure 2 a. The changes resulting from irradiating the Si-SiO$_2$ structure with 22-MeV electrons with a dose of $6.0 \times 10^{16}$ cm$^{-2}$ are illustrated in figure 2 b. The AFM images were taken over areas of size $1 \times 1$ μm (figure 2 a) and $4.6 \times 4.5$ μm (figure 2 b) on the surface of the SiO$_2$-Si structures in air.

The Si$^+$ ion implantation ($5.4 \times 10^{16}$ cm$^{-2}$) generates amorphous Si precipitations.

Figure 1 b. Random (red curve) and channelling spectra (blue curve) of the Si-SiO$_2$ structures implanted with 12-keV Si$^+$ ions (dose of $5.4 \times 10^{16}$ cm$^{-2}$) and then irradiated by 22-MeV electrons with a dose of $6.0 \times 10^{16}$ cm$^{-2}$.

Figure 2. AFM surface morphology of Si-SiO$_2$ sample implanted with 12-keV Si$^+$ ions (dose of $5.4 \times 10^{16}$ cm$^{-2}$); before (a) and after (b) 22-MeV electron irradiation (dose of $6.0 \times 10^{16}$ cm$^{-2}$).
in the SiO$_2$ matrix, as can be seen in figure 2 a. It can be assumed that it also rearranges the Si-O-Si bonds and breaks many of the Si-O bonds. In addition, the radiation defects generated during the Si-ion implantation (whose maximum is located in the Si wafer close to the Si-SiO$_2$ interface) create a thin layer of amorphous Si at the silicon surface. The following 22-MeV electron irradiation (6.0×10$^{16}$ cm$^{-2}$) of the Si-implanted samples regroups these Si precipitations, so that new silicon nanostructures appear in the SiO$_2$ (figure 2 b). We assume that these new nanostructures are Si nanocrystals [7]. The number (per cm$^2$) of the nanocrystals generated is small; their mean height is determined at 16-20 nm.

The crystal height depends also on the MeV electron dose irradiation [8]. The silicon nanocrystals size increases with the MeV electrons dose. The Si nanocrystals’ size reaches their maximum increase after MeV electron irradiation with a dose of 6.0×10$^{16}$ cm$^{-2}$. This has to do with the limited number of Si ions and atoms able to migrate through the radiation defects generated by the MeV-electron irradiation.

4. Conclusions
We explored the effect of 22-MeV electron irradiation (dose of 6.0×10$^{16}$ cm$^{-2}$) on n-Si-SiO$_2$ structures implanted by Si$^+$ ions (dose of 5.4×10$^{16}$ cm$^{-2}$) and followed the oxygen and silicon atoms’ redistribution in the implanted Si-SiO$_2$ samples after the MeV-electron irradiation. The amount of Si in the structures remained the same, while O (being mobile) diffused through the radiation defects generated in the structures by the MeV electrons (in particular in the oxide). We further analyzed the redistribution of both the oxygen and silicon atoms in the implanted Si-SiO$_2$ samples after MeV-electron irradiation and observed a partial transformation of the amorphous SiO$_2$ surface of the implanted samples into crystalline Si nanostructures. The mean height of the Si nanocrystal was determined to be 16-20 nm.

Acknowledgement
This study was supported by Grant No. 22 - 15.01.2016, p. 16 of JINR, Dubna, Russia. The authors appreciate the assistance of Dr. A.G. Belov of FLNR, JINR Dubna, Russia, for the MeV electron irradiation of the samples.

References
[1] Nepijko S, Klimenkov M, Kuhlenbeck H and Freund H 1999 *Vac. Sci. Technol. A* 17 577
[2] Klimenkov M, Matz M and Borany J 2000 *Nucl. Instr. & Meth.* B 168 367
[3] Babae S and Ghozati S B 2017 *Radiation Phys. and Chem.* 141 98-102
[4] Stefanov K, Kaschieva S and Karpusov D 1998 *Vacuum* 51/2 235
[5] Kaschieva S, Halova E, Vlaikova E, Alexandrova S, Valcheva E and Dmitriev S N 2006 *Plasma Process. & Polym.* 3 237-40
[6] Kaschieva S, Angelov Ch, Dmitriev S N and Tsutsumanova G 2006 *Plasma Process. & Polym.* 3 233-6
[7] Kaschieva S, Gushterov A, Apostolova T and Dmitriev S N 2010 *Proc. 6th Int. Conf. Beam Technol. and Laser Appl.* p 75
[8] Kaschieva S, Gushterov A, Gushterova P and Dmitriev S N 2010 *J. Phys.: Conf. Ser.* 223 012031