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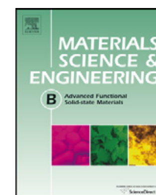

The influence of thermal treatments under hydrostatic pressure prior to irradiation on the annealing characteristics of the VO defect in Si

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ABSTRACT

Cz-Si samples, initially subjected to thermal treatments under high hydrostatic pressure, were subsequently irradiated by fast neutrons. This paper describes a series of infrared spectroscopy measurements that enabled us to determine the effect of the pre-treatments on the annealing characteristics of the VO defect in Si. We found that the activation energies of the two main annealing reactions: $VO + O_i \rightarrow VO_2$ and $VO + Si_I \rightarrow O_i$ that the defect participates, are comparatively smaller than those of initially untreated samples, correspondingly. We argue that the pre-treatments reduce the potential barrier for the migration of the VO defect ($VO + O_i \rightarrow VO_2$) and also reduces the binding energy of the Si_I's, bound at large defect clusters ($VO + Si_I \rightarrow O_i$).

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1. Introduction

A-center, that is the vacancy-oxygen (VO) pair, is among the dominant defects produced in Cz-grown Si material subjected to any kind of irradiation at room temperature. The main reasons are that the monovacancies produced by irradiation (i) are too mobile to survive at such temperatures and (ii) that they are selectively trapped by oxygen atoms being at large concentrations in Cz-Si. The structure and the properties of the center have been meticulously studied by various experimental techniques such as electron paramagnetic resonance (EPR), deep level transient spectroscopy (DLTS), infrared spectroscopy (IR), etc. In the negative charge state the defect gives rise [1] to an EPR spectrum labeled Si-A. The suggested model [2] for its structure is a nearly substitutional oxygen atom, and more specifically an oxygen atom located in an off-center substitutional site in the ⟨1 0 0⟩ direction. DLTS measurements have associated [3] with the A-center an acceptor level in the band gap at E_c-0.18 eV. IR spectroscopy which has been proven a powerful technique in investigating oxygen-vacancy related defects in Si, has associated [4,5] two localized vibrational mode bands at 830 and 877 cm^{-1} related to the neutral and the negative charge states of the VO defect, respectively.

Upon heating the samples, the VO defect participates in numerous reaction channels [6]. It may be associated with an oxygen atom to form the VO_2 defect, with a lattice vacancy to form the V_2O defect or even with another VO defect to form the V_2O_2 complex. It may also dissociate or being destructed by self-interstitial atoms. Obviously, the contribution of all these reactions in the VO annealing is not the same. In neutron irradiated Cz-Si, the two reactions $VO + O_i \rightarrow VO_2$ and $VO + Si_I \rightarrow O_i$ are considered [6] as the most important reactions involved in the annealing process of the VO defect. An important parameter of each reaction process that a defect participates is the activation energy. Its knowledge is significant not only for the basic physics of defect studies but also as a crucial piece of information for defect control and defect engineering processes. On the other hand, pressure is an important external parameter besides temperature which affects the behavior and properties of defects. The present work is focused on the study of the influence of thermal treatments under hydrostatic pressure on the activation energies of the processes that govern the annealing of the VO defect and the growth of the VO_2 defect. It is well known [7,8] that thermal treatments at 450 °C introduce thermal donors, while thermal treatments at 650 °C introduce new donors which are associated to some kind of oxygen precipitates. In addition, the application of high hydrostatic pressure can induce [9] changes in the structure of defects. Thermal treatments under high hydrostatic pressure reduce [10] significantly the concentration of oxygen, thus enhancing the formation of thermal donors at 450 °C and promoting oxygen precipitation at 650 °C. We note that oxygen dimer, which is the first stage of oxygen precipitation process, is not expected [11] to survive at the above temperatures. Thus, the formation of the VO_2 defect as a result of the reaction $V + O_2 \rightarrow VO_2$ is not expected to occur in irradiated Si initially subjected to thermal treatment under high hydrostatic pressure. Only the VO defect is

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formed, which is expected to convert to the VO_2 defect at ∼300 °C. The particular aim of this work is to study the evolution of the VO defect and its conversion to the VO_2 defect in a crystal environment which is different from that of an initially untreated material, due to the pre-treatments, and calculate the activation energies of the involved processes. Their values are expected, in general, to be different than those of the initially untreated material.

2. Experimental details

We used three groups of samples Cz-Si A_0, A_3, A_7 with initial oxygen content 1.4×10^{18} cm^{-3}. The samples labeled A_0 are initially untreated. The samples A_3 and A_7 were subjected to thermal treatments with the application of hydrostatic pressure: A_3 (450 °C, 10.65 kbar, 10 h), A_7 (650 °C, 11 kbar, 10 h). After the above treatments all the samples were irradiated by fast neutrons with a fluence of 8×10^{16} n/cm^2 at Tirr ≈ 50 °C. Afterwards, 30 min isochronal anneals were carried out in steps of ∼10 °C, in the range from 50 to 550 °C. After each annealing step the IR spectra were taken at room temperature by employing a Jasco IR spectrometer of dispersive kind.

3. Experimental results and discussion

Fig. 1 shows fragments of the IR spectra of the A_0, A_3 and A_7 samples after the pre-treatments, after the irradiations and after the anneal at 350 °C, in the course of the isochronal annealing sequence starting from 50 °C. After the irradiation only the VO defect (828 cm^{-1}) is produced, as expected.

Fig. 2 shows the evolution with temperature of the amplitude of the O_i (1104 cm^{-1}), VO (828 cm^{-1}) and VO_2 (885 cm^{-1}) defects in the temperature range of 50–550 °C, during the isochronal anneal sequence, of the samples A_0, A_3 and A_7. As it is seen from the evolution curves there is a gradual increase in the amplitude of the VO band between 200 and ∼300 °C. This inverse annealing stage, occurring prior to the onset of the decay of the VO defect and its conversion to the VO_2 defect, is apparently due to the additional formation of VO defects. In fact, in the course of the annealing sequence, vacancies are liberated in this temperature range which are captured by oxygen atoms to form the VO defects. Sources of these vacancies are large clusters of defects [11,12], divacancies [13] and disordered regions [14]. The decrease of the O_i signal in the same temperature range, supports the interpretation for the additional VO formation.

The annealing of the VO defect is a rather complicated process. At least two main mechanisms should be considered [6]. At first, we observe that, the decrease of the amplitude of the 830 cm^{-1} band of the VO defect is accompanied by the emergence and growth in the spectra of the 885 cm^{-1} band of the VO_2 defect. The reaction that describes the phenomenon [15] is the $VO + O_i \rightarrow VO_2$, which is a first order process considering that $[O_i]$ is more than one order of magnitude larger than that of the [VO]. The corresponding differential

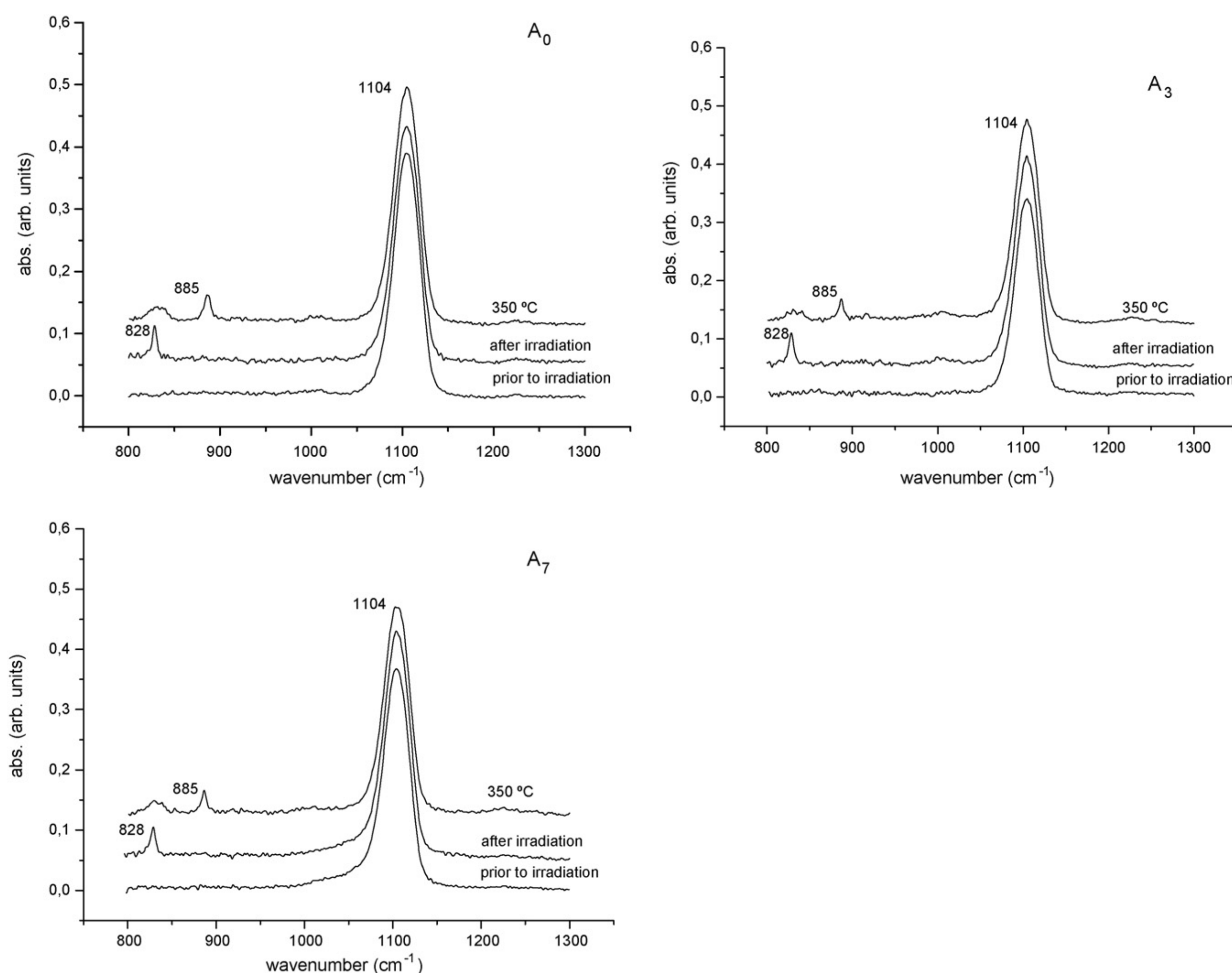

Fig. 1. Fragments of the IR spectra of the samples A_0, A_3 and A_7 after the pre-treatments, after the irradiation and after the isochronal anneal at 350 °C, correspondingly.

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