



Graphene oxide films for field effect surface passivation of silicon for solar cells



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ABSTRACT

In recent years it has been shown that graphene oxide (GO) can be used to passivate silicon surfaces resulting in increased photocurrents in metal-insulator-semiconductor (MIS) tunneling diodes, and in improved efficiencies in Schottky-barrier solar cells with either metal or graphene barriers, however, the source of this passivation is still unclear. The suggested mechanisms responsible for the enhanced device performance include the dangling bond saturation at the surface by the diverse functional groups decorating the GO sheets which reduce the recombination sites, or field effect passivation produced by intrinsic negative surface charge of GO. In this work through a series of measurements of minority carrier lifetime with the microwave photo-conductance decay (μ PCD) technique, infrared absorption spectra, and surface potential with Kelvin probe force microscopy (KPFM) we show that there is no evidence of significant chemical passivation coming from the GO films but rather negative field effect passivation. We also discuss the stability of GO's passivation and the flexibility of this material for its application as temporary passivation layer for bulk lifetime measurements, or as a potential cheap alternative to current passivation materials used in solar cell fabrication.

1. Introduction

It is well known that one of the most effective approaches to improve the efficiency of silicon-based solar cells, whilst maintaining low cost, is to increase the lifetime of photo-generated carriers by reducing recombination at the surface and in the bulk of low cost materials [1]. Therefore the study of new materials which may suppress surface recombination in such materials is important both because they may promote a minority carrier lifetime improvement in commercial solar cells but also, in the laboratory, they may allow more accurate measurements of silicon's bulk lifetime. For these reasons such new materials remain a topic of widespread interest [2–4].

Generally, for surface passivation materials we need to consider three material property aspects: the characteristics of the material to be passivated (doping type and resistivity); the physical properties of the passivating materials (optical, chemical and electrical), to determine the type of passivation that the material will provide including bond saturation, the field effect control of carriers, refractive index, and stability; and processing requirements like surface cleaning and synthesis methods. With this in mind, in this work we have studied the passivation capabilities of graphene oxide which we know fulfils some

key requirements for surface passivation including: high transmittance [5], fixed surface negative charge [6,7] and high refractive index [8,9]. This is all combined with the fact that GO is water dispersible making its deposition and removal extremely simple. Most common GO deposition techniques include: dip coating, spin coating, and spray coating, techniques which can be easily incorporated to a production line. In addition, it has been recently shown that for a GO derivative dispersion with an optimal dilution, it is possible to obtain uniform coverage even on textured silicon surfaces for solar cells [10].

While passivation effects in structures incorporating GO interlayers, such as MOS and graphene/GO/Si devices, have already been reported in literature [11–16], these do not include the study of graphene oxide as a passivation material for solar cells in a scalable to manufacture system. Furthermore, to our knowledge there is no definitive answer as to what is causing the passivation on the surface from the available literature. Thus in this work we aim to answer such a question by providing strong evidence of the influence of the negative surface charge of GO in the passivation of silicon. Additionally, we demonstrate the passivation effect of GO in different silicon materials and the inclusion of GO's passivation capabilities compared to that of the industry standard material, silicon nitride (SiN_x) used for solar cell passivation

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[17] and the widespread temporary passivation attained from Iodine-Ethanol (I-E) solution immersion for bulk lifetime measurements [3].

2. Materials and methods

In this work a range of float zone (FZ) and Czochralski (Cz) grown, boron-doped silicon wafers with $\langle 100 \rangle$ crystal orientation, double (DSP) and single (SSP) side polished, with resistivities between 1 and $1000 \Omega\text{-cm}$, and thicknesses from 200 to $625 \mu\text{m}$ have been used to study the passivation effect of GO. Most of the measurements were carried out in cut samples of $2 \times 2 \text{ cm}^2$ which were subjected to the standard cleaning ‘‘RCA 1’’ procedure as described in Ref. [18]. However, the usually applied subsequent HF dip of the samples was not performed since it was found that such step resulted in a diminished passivation of the silicon surface due to the lack of hydrophilicity on the surface provided by either the native oxide or the oxide left by the H_2O_2 . It must be noted as well that the samples were cleaned and left in the desiccator for at least 3 days before the GO deposition, and their lifetime measured before and after the deposition to avoid the inclusion of the RCA 1 passivation effect into consideration, it was determined that this process takes place within that period before stabilisation.

GO was prepared by a modified Hummers method [19] whereby natural flake graphite (30 mesh, 96% C) was oxidised and exfoliated. Sulphuric acid (H_2SO_4 , 98%), hydrogen peroxide (H_2O_2 , 30%), ammonium hydroxide (NH_4OH , 35%) (Sigma Aldrich); sodium nitrate and potassium permanganate (KMnO_4 , Alfa Aesar) were all used as supplied and without any further purification. The oxidised graphite was repeatedly washed and exfoliated using a 3% wt. H_2SO_4 / 0.5% wt. H_2O_2 mixture and then washed further with deionised water until a pH of 7 was reached for the supernatant to ensure the removal of the H_2SO_4 . The GO pellet had a pH of 3.6. To adjust the pH of GO, $100 \mu\text{l}$ of ammonium hydroxide was added to under stir to give a pH of 9.9 ± 0.01 measured with a Mettler Toledo F20 pH meter.

For the silicon nitride deposition we used the Plasma-Enhanced Chemical Vapour Deposition (PECVD) technique for 2 min, to achieve a film thickness of $90 \text{ nm} \pm 3 \text{ nm}$ according to spectroscopic ellipsometry. This was done on a PlasmaPro 100 PECVD system from Oxford Instruments with: 20:20 sccm SiH_4 : NH_3 gas flow, 300 mTorr chamber pressure, $400 \text{ }^\circ\text{C}$ table temperature, 13.56 MHz plasma generated frequency and 50 W of power as deposition parameters.

Kelvin Probe force microscopy on the PeakForce tapping frequency-modulated mode (FM-KPFM) using SCM-PIT-V2 tips was used to map the surface potential of single GO flakes on a Bruker Dimension FastScan. Samples for this measurements were prepared by spin coating 0.5 mg/ml of GO for 60 s at a speed of 1000 rpm and left to dry on a desiccator overnight. Infrared absorption spectroscopy on $200 \text{ nm} \pm 20 \text{ nm}$ thick GO coated silicon samples was carried out with a Hyperion 3000 FT-IR microscope with a resolution of 4 cm^{-1} in transmission mode.

A Semilab WT-2000 PVN was used for minority carrier lifetime mapping by the microwave photoconductivity decay (μPCD) technique, with a 905 nm LASER excitation and a microwave source operating at $\sim 10 \text{ GHz}$. Hence the presented effective lifetime maps show the averaged lifetime down to a few tens of microns into the sample. The level of passivation achieved by GO layers was tested in several samples by the measurement of their effective lifetime (τ_{eff}) by μPCD mapping, with the assumption that both sides of each sample were equally passivated. Next, what is considered the upper limit of the surface recombination velocity (S_{UL}) was calculated by assuming the bulks lifetime (τ_{bulk}) to be infinite, thus giving $S_{\text{UL}} = W/2(\tau_{\text{eff}})$, with W representing the sample thickness.

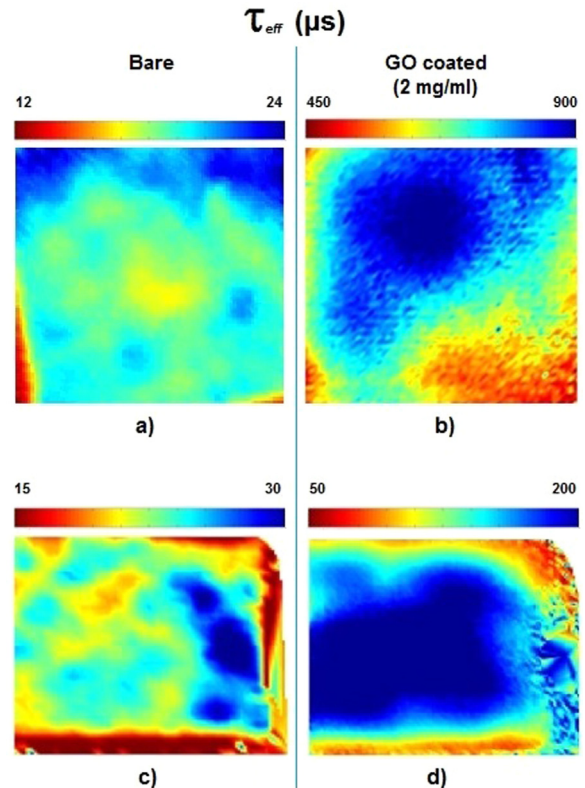


Fig. 1. Effective lifetime maps of a DSP FZ p-type ($> 1000 \Omega\text{-cm}$) silicon sample (a) bare and (b) GO coated, and a SSP Cz p-type ($2.8 \Omega\text{-cm}$) silicon sample (c) bare and GO coated.

3. Results and discussion

3.1. GO effect on minority carrier lifetime

We have carried out transient photoconductance mapping to compare the level of surface passivation achieved by GO in various sets of samples, here we present the comparison of GO's passivation on an electronic grade FZ material to that of a solar grade Cz material. Fig. 1 show the lifetime maps of the FZ and the Cz boron doped samples before (a, c), and after (b, d) GO passivation, respectively. For the FZ sample it can be observed that the τ_{eff} approaches 1 ms in some areas. Nevertheless, considering the maximum effective lifetime in the sample to be $\tau_{\text{max}} = 900 \mu\text{s}$ and $W = 500 \mu\text{m}$ we obtain a $S_{\text{UL}} \approx 27 \text{ cm s}^{-1}$, whereas from the $\tau_{\text{max}} = 820 \mu\text{s}$ and $W = 200 \mu\text{m}$ of the Cz sample the calculation results in a $S_{\text{UL}} \approx 30 \text{ cm s}^{-1}$. These values are comparable to the surface recombination velocities of some existent surface passivation materials [3,4]. Moreover, we have compared the surface passivation of GO to that of PECVD deposited SiN_x and I-E solution immersion. The passivation was carried out on samples cut from a solar grade SSP Cz p-type wafer ($25 \pm 7.5 \Omega\text{-cm}$), the lifetime comparison is presented in Fig. 2. The calculated upper limit surface recombination velocities from each sample are: 363 cm s^{-1} , 322 cm s^{-1} and 202 cm s^{-1} for GO (Fig. 2a), I-E (Fig. 2b) and SiN_x (Fig. 2c), respectively. These results indicate that in some silicon materials it is possible to attain very similar levels of surface passivation with GO coatings to those obtained by the widely used SiN_x and I-E techniques.

With respect to the surface passivation stability we have found that the GO passivation effect can last for several days, however there appears to be some unpredictable behaviour in GO's passivation after storage. We have observed there is an increase in averaged lifetime after a few days of air ambient storage of the samples, which we suggest to be attributed to the interlayer water molecule release which

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