A Discussion about Defects Passivation and Etching Mechanisms in Polycrystalline Silicon Hydrogenated by MW-ECR Plasma

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Abstract- In this work we have investigated the microwave plasma discharge assisted by electron cyclotron resonance for hydrogenation of thin film polycrystalline n^+pp^+ silicon solar cells in terms of defects passivation and surface etching. The polycrystalline silicon films were formed by high temperature chemical vapor deposition. Influence of various process parameters such as microwave plasma power (P_{MW}), hydrogenation time (t_H) and substrate temperature (T_H) on the sheet resistance of the n^+ emitter region and on the open-circuit voltage (V_{oc}) of the n^+pp^+ structure were investigated. The n^+ emitter region was obtained by phosphorus diffusion using a spin-on dopant P507 solution from filmtronics. After 75 min of hydrogenation, V_{oc} is greatly improved by a factor of 2.5 and reaches values up to 440 mV at 500°C and microwave power of 650W. This enhancement in V_{oc} after plasma hydrogenation is originating from a reducing recombination velocity at the grain boundaries due to the passivation of dangling bonds by hydrogen atoms, leading to an important increasing of the diffusion length. Above our hydrogenation parameters values, degradation is observed, especially an etching of the emitter region. The origin of such behaviour were thoroughly explained in detail.

Index Terms- Defects passivation, grain boundary, plasma hydrogenation, polycrystalline silicon, solar cells.

I. INTRODUCTION

The thin polycrystalline silicon (pc-Si) solar cells are considered to be ones of the most promising cells capable of achieving both high efficiency and low cost. On the other hand polycrystalline silicon substrates contain many kinds of defects and impurities compared to the single crystal silicon substrates, so the efficiency of the polysilicon solar cells have been inferior to that of single crystal cells. The impurities can be removed by gettering [1, 2] and the defects can be passivated by hydrogen [3-5] what increase the performance of polycrystalline silicon solar cells. A great number of papers have recently appeared, where the results of hydrogen passivation of n^+pp^+ cells manufacturing on the basis of thin polycrystalline silicon deposited by thermal CVD are published. A particular attention was paid to the methods used to introduce hydrogen in the n^+pp^+ cells. The most widely hydrogenation techniques employ the immersion of n^+pp^+ cells in a dense hydrogen plasmas and/or the deposition of a hydrogenated a-SiN$_x$ layer by plasma enhanced chemical vapor deposition (PECVD) to provide simultaneously an antireflection coating, surface and bulk passivation [6, 7]. A large improvement of the electronic properties of n^+pp^+ cells was achieved by the plasma hydrogenation whereas the deposition of a hydrogenated a-SiN$_x$ layer provides a moderate benefit [6]. Nevertheless, plasma hydrogenation induces an etching of the emitter region (n^+) as well [6, 8].

The microwave discharge assisted by electron cyclotron resonance (MW-ECR) plasmas are of interest because of the large quantity of passivating species, the low ion energy (low ion-beam damage) and the low pressures afforded these plasmas. As a result, we showed in our previous papers [9, 10] that hydrogen plasma generated in MW-ECR plasma system provide an important improvement in open circuit voltage (V_{oc}) measured on n^+pp^+ polycrystalline silicon cells with low damage of the emitter region (n^+). But up today, there is no sufficiently full analysis of the physical processes running the diffusion of hydrogen from the plasma to a p-type region and the optimal conditions leading to high values of V_{oc}.

In this connection, the goal of this work consists to the study of hydrogenation process of thin film polycrystalline n^+pp^+ silicon cells using MW-ECR plasma system. The influence of the microwave plasma power (P_{MW}), the substrate temperature (T_H) and the process time (t_H) on the efficient of the defect passivation and emitter surface etching are investigated. The passivation effectiveness is witnessed through the open-circuit voltage of mesa structures on n^+pp^+ poly-Si films, while the etching process is monitored via optical interferometric microscope and sheet resistance (R_{sq}) of the n^+ emitter region.

II. EXPERIMENTAL

The polysilicon films were formed on thermally oxidized silicon wafers by rapid thermal chemical vapor deposition (RTCVD) at 1080°C, under atmospheric pressure of the trichlorosilane and diborane as precursor and doping gas, respectively. This results in polysilicon films with columnar structure, mainly <220> oriented grains [6, 11]. The thickness of stack layers of p^+ and p was fixed at 0.5 µm and 4 µm with a doping concentration of $5x10^{19}cm^{-3}$ and $10^{17}cm^{-3}$, respectively. The n^+ emitter region was formed by phosphorus diffusion at 900°C for 50 min from a spin-on dopant oxide source (P507 solution Filmtronics Inc.). It results in an ercf function distribution with a surface concentration of $5x10^{20}cm^{-3}$ and a junction depth of about 0.5µm. After the diffusion process, the samples were subjected to a 10%HF solution treatment to

remove the residual dopant oxide on the surface. An average sheet resistance of the n^+ emitter region of 30 Ω/□ was measured by the four-point probe technique. As for the solar cell structure, we used the side-contacted mesa. Access to the p^+ back surface field region was made by reactive ion etching (RIE), where SF_6 gas etches the n^+p with a rate of 1.6μm/min, thus forming a mesa cell. However, the mesa contact structure used here resulted in high series resistances, and therefore small short circuit currents were measured.

Hydrogenation experiments were carried out using MW-ECR plasma system (Roth & Rau) at ICUBE (formely InESS) laboratory, Strasbourg University, France. A schematic of such system has been shown elsewhere [6]. Hydrogen gas was excited by 2.45GHz microwave in a resonant chamber, where a magnetic field is applied to maintain the ECR condition. The gas pressure of plasma was 0.6 Pa at a hydrogen flux (H_2) of 30 sccm and the hydrogenation time was changed in the range of 15 to 90 min. The microwave (MW) plasma power and hydrogenation (substrate) temperature were varied from 100 W to 650 W and 250°C to 500°C, respectively. In all cases, the input MW power and the reflection were monitored, there was little reflection (<5%). In order to avoid out-diffusion of hydrogen during the cool-down phase, the plasma was maintained around 10 to 20 min until the substrate temperature reached to 280°C.

III. RESULTS AND DISCUSSIONS

The electrical properties of polycrystalline silicon are dominated by grain boundary defects. So, the measured Voc of the as-deposited silicon film was 180 ± 10 (mV). In our study, the hydrogenation experiments of mesa cells through the n+ emitter have been carried out in MW-ECR plasma system (Roth & Rau). Such system allows the user to manipulate various operating parameters so as to optimize the hydrogenation process.

A. Effect of microwave plasma power

Initially, the influence of MW plasma power, P_{MW}, which was varied from 100 to 650W, on V_{oc} was analyzed. The plasma process was carried out at 400°C for 1 hour. Fig.1 plots the evolution of V_{oc} as a function of the MW plasma power. At P_{MW} of 100W, the hydrogen plasma could not be initiated. Therefore, the measured V_{oc} is almost unchanged compared to that of the non-hydrogenated pc-Si n^+pp^+ silicon solar cells. With further increase of P_{MW}, a significant and continuous increase of V_{oc} is observed. For a P_{MW} of 650W, V_{oc} reached to 370 mV. This improvement in V_{oc} is due to the passivation of defects states at the grain boundaries and suppression of the band bending, which act as barriers for majority carriers and recombination sites for minority carriers. Our results are in good concordance with those reported elsewhere [12-14]. Although we did not examine the plasma content in our plasma system, it is expected that electrons act as the agents that absorb the microwave energy at these low discharge pressures [15, 16]. In addition, the ionization of hydrogen species will occur by a collision with the energetic electrons, resulting in the formation of H^+ ions. With increase in MW plasma power, more and more H_2 molecules could be excited, dissociated and ionized, leading to an increase in both H^+ and electron densities. The incident H^+ impinging the sample surface in contact with plasma is usually caused by the divergent

magnetic field and the sheath potential originating from the differences in ion and electron velocities. The increased of microwave plasma power give rise to an increase in H^+ reaching the n^+ surface. The question is how hydrogen diffuses from the n^+ surface to p-type region. According to Johnson [17], the charge state of hydrogen (H^+, H^- and H^0) in silicon depends on the Fermi level position in the band gap. It is also assume that near and above room temperature charge change, $H^+↔H^-$, occurs through the state configuration of H^0 [17, 18]. Accordingly, it could be assumed that H^0 species have an extremely high diffusion coefficient compared to capture an electron or another H^0 to give respectively H^- or H_2. Consequently, H^+ absorb an electron at the n^+ surface to become H^0 ($H^+ + e^- → H^0$) and the H^0 diffuse away toward the p-type region without a Coulomb barrier to overcome [17]. After traversing the depletion layer, a neutral hydrogen atom would be ionized by capturing a free hole ($H^0 + h^+ → H^+$) and continue to diffuse as a proton into the p-type substrate. Then, defects passivation would progress with increasing MW plasma power which gives rise to an increasing availability of atomic hydrogen reaching the p-type region. Based on hydrogen diffusion coefficient data [19], the penetration depth of hydrogen H^- in the n-silicon layer and H^+ and H^0 in the p-silicon layer can be estimated to be 68.4 μm, 74.6 and 130 μm, respectively. This means that the solubility of hydrogen in silicon is reached under the selected conditions of MW-ECR plasma hydrogenation. However, because of the type of the charges, n^+-type region in the structure used for the V_{oc} measurements could hinder the diffusion of the hydrogen atoms through the whole sample and therefore could reduce the passivation effectiveness of defects. Another road that may be influence the hydrogen diffusion in polysilicon is the presence of oxygen. It has been often suggested that the high electrical activity of grain boundaries in polysilicon deposited at high temperature is linked to decoration by oxygen atoms [11, 20]. The origin of the oxygen is probably from the bulk and/or from the surface or other defects [20]. In this case hydrogen will diffuse slowly because the presence of oxygen lowers the propensity for generation of vacancies [21, 22].
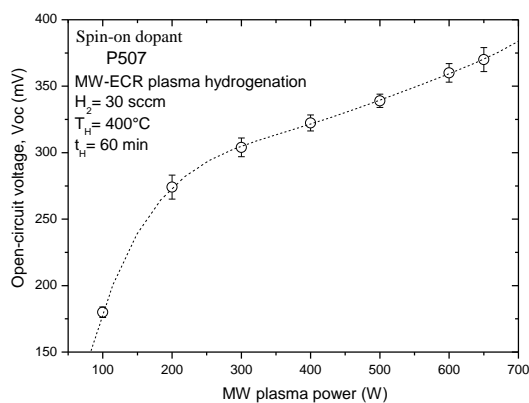

Figure 1: Influence of microwave plasma power on open-circuit voltage measured on the pc-Si n^+pp^+ mesa cells. The hydrogenation conditions are indicated.

In addition to the passivation of defects, the MW-ECR plasma produces etching of n^+ region and thereby increases the emitter sheet resistance [9, 10]. Measurements of the emitter sheet resistance (R_{sq}) of the n^+pp^+ structures by the four-point probe technique showed an increase by a factor of 4 to 8 after MW-ECR plasma hydrogenation. In figure 2, we present the etched layer thickness and the difference in emitter sheet resistance before and after hydrogenation as a function of the microwave plasma power. A slight layer thickness etching of n^+ region was measured by optical interferometry at 650W accompanied with slow increase in R_{sq}. When MW plasma power decreases, etching and R_{sq} simultaneously increase to reach a maximum near 300W. However, at 200W the total thickness of the emitter was almost completely etched and the four-point probe technique was not valid to give a correct measure of the emitter sheet resistance. Etching of the silicon surface in the presence of hydrogen plasma is a competition between electron-enhanced etching of the silicon surface and redistribution of silicon from electron induced decomposition of the etch product [23]. This usually involves the presence of H_2 on the silicon surface, reaction between hydrogen and the silicon atoms to form an adsorbed product molecule, and finally desorption of the product molecule into the gas phase [24]. The consequence of the above processes are largely determined by both electrons and ions energies. An increase of MW plasma power enhances the ionization of hydrogen (low density of H_2 molecules on silicon surface) and hence the plasma density due to high frequency of inelastic collisions between hydrogen and electrons, which results in the decrease of electron energy [16]. Consequently, density of molecular hydrogen in the plasma and on silicon surface could be less. In such a case, electron induced etching via hydrogen molecules on silicon surface might be diminished. Therefore in the present investigation, a decreasing trend of etching level with the increase of MW plasma power was observed. At a MW power of 200W, the average electron energy could be lower than the threshold energy of 15-30 eV required for inelastic collision [25] to create hydrogen ions, consequently silicon surface is mostly covered with molecular hydrogen. Therefore, most of the electrons remain in the higher energy. In such case, electron induced etching prevails, which is obvious at a MW plasma power of 200W in the present study as shown in Fig. 2.
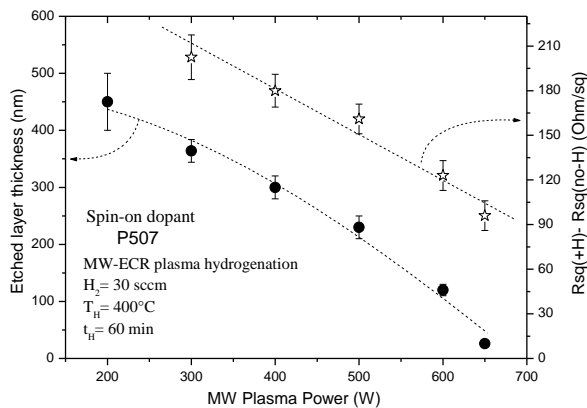

Figure 2: Etched layer thickness and the difference in emitter sheet resistance without and with hydrogenation versus microwave plasma power.

B. Effect of substrate temperature

Fig. 3 plots the open-circuit voltage of hydrogenated n^+pp^+ mesa cells versus samples temperature in the MW-ECR plasma system for a fixed MW plasma power, hydrogen gas flux and hydrogenation time. Clearly, the influence of the temperature is very important. The V_{oc} increases drastically with the substrate temperature from 180 mV before hydrogenation to 425 mV after 60 min of hydrogenation at 500°C. This can be explained by the diffusion constant of hydrogen atoms in silicon leading to higher diffusion lengths for higher temperatures. Also, we can see in fig. 3 that V_{oc} begin to saturate at temperature higher than 450°C resulting probably from a competition between in-diffusion and out diffusion of hydrogen into silicon [6].
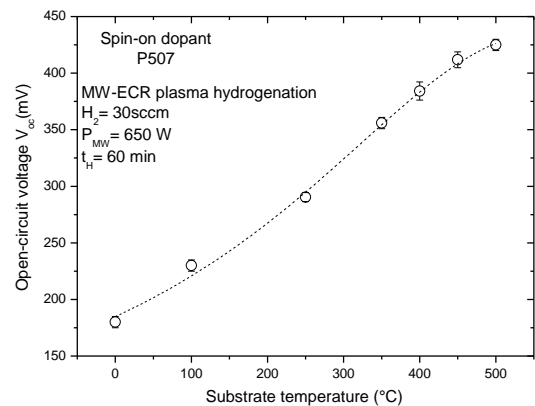

Figure 3: Open-circuit voltage versus hydrogenation temperature measured on pc-Si n^+pp^+ mesa cells. The hydrogenation time is 60 min, the MW power is 650W and H_2 flow is 30 sccm

The substrate temperature has also a significant effect on the emitter sheet resistance of the n^+pp^+ structures. Figure 4 plots the emitter sheet resistance of hydrogenated n^+pp^+ mesa cells versus substrate temperature in the MW-ECR plasma system. For MW
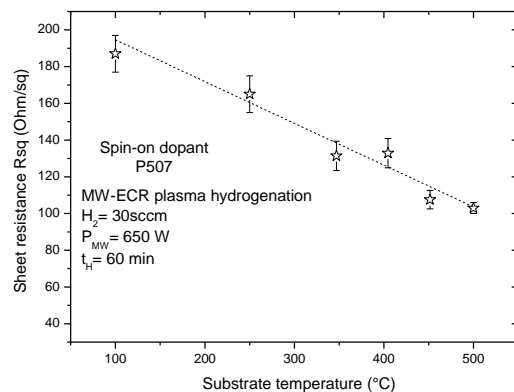

Figure 4: Measured sheet resistance versus hydrogenation temperature of n^+pp^+-type made on CVD pc- Si layers. The hydrogenation time is 60 min, the MW plasma power is 650W and H_2 flow is 30 sccm.

plasma power of 650W and after 60 min of hydrogenation, the sheet resistance R_{sq} showed an increase by a factor of 3–6 depending on substrate temperature. Indeed, the slight increase in R_{sq} is observed at 500°C. When the substrate temperature decreases, R_{sq} increases to reach a maximum at 100°C. These observations can be correlated to the etching of the emitter region during hydrogenation. Indeed, it can be explained by the following statements: (1) high MW plasma power increases the number of ionized species in the plasma, which increases the probability for reactive species to reach the sample surface; (2) high density of hydrogen species on the silicon surface induces a very high recombination probability, and (3) at low temperature, the interactions of the reactive species with the silicon surface are more important that for high temperature, increasing the each rate through longer average time spent on the surface for the reactive species. The fact that hydrogen etching becomes more efficient at low temperature has also been predicted theoretically by Van de Walle [26].

C. Effect of hydrogenation time

The hydrogenation time and the concentration of hydrogen introduced into the samples are important parameters for an effective passivation of defects. In figure 5, the measured open-circuit voltage, V_{oc}, is presented as a function of hydrogenation time. It can be seen that the electronic properties of the polycrystalline silicon films dramatically improved with time compared to the ones of as-prepared pc-Si. Improvement of V_{oc} saturates and even decreases for prolonged hydrogenation times. Indeed, values of V_{oc} approaching 440 mV and 425 mV were measured for 75 min and 90 min, respectively.
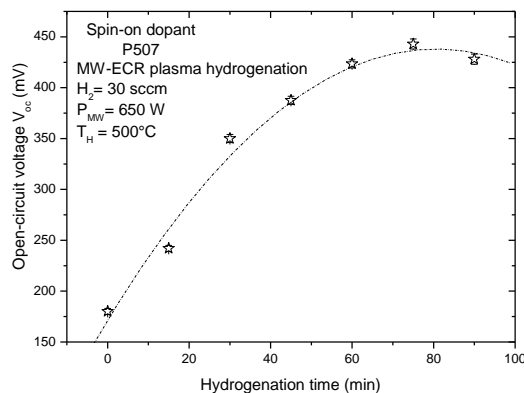

Figure 5: Measured open-circuit voltage versus hydrogenation time of polysilicon based n^+pp^+ solar cells. The hydrogenation temperature is 500°C and H_2 flow is 30 sccm.

The hydrogenation time dependence of the passivation efficiency arises from the concentration of hydrogen introduced into the n^+pp^+ silicon solar cells. The enhancement of V_{oc} is due to the passivation of defect states at grain-boundaries. The increase of V_{oc} with hydrogenation time means a higher concentration of hydrogen penetrates deeply in the polysilicon layer and therefore passivates more defects like dangling bonds. However, the decrease of the V_{oc} for long plasma hydrogenation time can be attributed to the formation of defects. In fact, many

researchers reported that hydrogenation could influence silicon in two ways: it passivates the defects by hydrogen bonding and also generates new defects [27]. When hydrogenation is completed, the hydrogen concentration might exceed the concentration of broken bonds by about two orders of magnitude [28]. The excess of hydrogen can be bonded to weak Si-Si bonds thus leading to the formation of two Si-H complexes: a hydrogen atom involved in a Si-Si bond produces dangling bonds. Other possible defects induced by excessive hydrogenation are Si-H_2 inside the grains rather than at grain boundaries [29] and a formation of subsurface defects that result from the accumulation of diatomic hydrogen (H^*_2) which are usually located at a depth of 100 nm from the surface [30]. The formation of vacancy-H complexes can decrease the open circuit voltage as well.

It can be noticed that the conditions yielding high V_{oc} values (fig. 5) correspond to those that cause substantial etching of the pc-Si surface. Indeed, figure 6 plots the measured sheet resistance of of n^+pp^+-type made on CVD pc-Si layers versus hydrogenation time. Clearly, the influence of the time is very important. The sheet resistance R_{sq} increases gradually with the time from 30 $\Omega/\square$ before hydrogenation to 196 $\Omega/\square$ after 90 min of hydrogenation at 500°C. This can be explained by the etching rate constant of emitter region by hydrogen plasma treatment leading to higher etching rate for long times of hydrogenation.
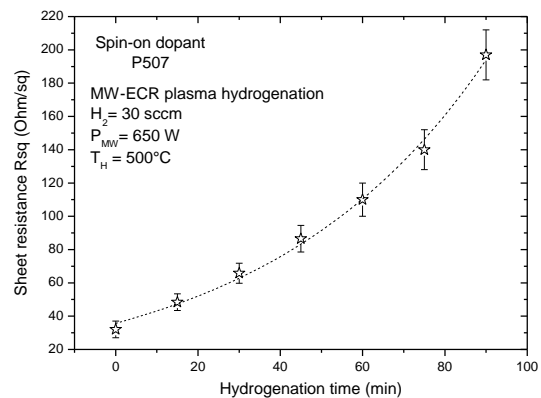

Figure 6: Evolution of sheet resistance versus hydrogenation time of n^+pp^+ polysilicon based cells. The hydrogenation was carried out at 500°C for 30sccm of H_2 flow and MW plasma power of 650W.

Finally, another mechanism most likely influence the open-circuit voltage values of our polysilicon solar cells is the damage of emitter region induced by the MW-ECR plasma hydrogenation. Such damage actually has an advantage as it increases the concentration of trapped hydrogen near the surface and allows more hydrogen to diffuse into the material [22]. On the other hand it reduces the phosphorus concentration in the n^+ region used for the V_{oc} measurements. This phenomenon leads to an increased in leakage current and consequently to a decrease in the photo-generation rate of the material [31].

IV. CONCLUSION

In this work we have investigated the effect of microwave plasma discharge assisted by electron cyclotron resonance on the

electronic properties of n^+pp^+ mesa structure solar cells made on fine-grained polycrystalline silicon films. We observed a large improvement of the open-circuit voltage by using microwave plasma power at 650W. Thus, the recombination strength of defects located at the grain boundaries is strongly reduced and the V_{oc} increased from 180 mV before plasma hydrogenation to 370 mV after 60 min of hydrogenation at 400°C and 30 sccm of hydrogen flow. However, MW-ECR plasma hydrogenation induces an etching of the emitter as well. The optimization of MW-ECR plasma hydrogenation of pc-Si n^+pp^+ solar cells as well as the physical processes running the interplay between the silicon surface and the hydrogenation conditions is evoked. Finally, we find that conditions yielding high V_{oc} values up to 440 mV correspond to those that cause substantial etching of the poly-Si surface.

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