

Research Article

Annealing Kinetic Model Using Fast and Slow Metastable Defects for Hydrogenated-Amorphous-Silicon-Based Solar Cells

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The two-component kinetic model employing “fast” and “slow” metastable defects for the annealing behaviors in pin-type hydrogenated-amorphous-silicon- (a-Si:H-) based solar cells is simulated using a normalized fill factor. Reported annealing data on pin-type a-Si:H-based solar cells are revisited and fitted using the model to confirm its validity. It is verified that the two-component model is suitable for fitting the various experimental phenomena. In addition, the activation energy for annealing of the solar cells depends on the definition of the recovery time. From the thermally activated and high electric field annealing behaviors, the plausible microscopic mechanism on the defect removal process is discussed.

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1. INTRODUCTION

In recent years, there has been an explosive, worldwide increase in solar module market due to the global warming and oil crisis. Because bulk crystalline silicon (c-Si) modules make up 90% of products, a severe shortage of c-Si wafers has caused an increase in the cost of the bulk c-Si solar modules. Thus, the increased cost of c-Si wafers currently threatens the photovoltaic business. Thin-film Si solar modules using hydrogenated-amorphous-silicon- (a-Si:H-) based absorbers, meanwhile, become a promising alternative to the bulk c-Si solar modules, because of their remarkably low consumption of raw Si material (<1% of consumption of bulk c-Si modules), large-scale deposition, and low-temperature production. Furthermore, thin-film Si photovoltaic technology profits from the wide experience base of the display industries [1].

However, the so-called “Staebler-Wronski effect (SWE)” in a-Si:H-based films remains as a major obstacle to the commercialization of thin-film Si solar modules. SWE is the light-induced degradation arising from the photocreation of dangling bonds (DBs) accomplished by the nonradiative recombination of photogenerated electron-hole pairs [2, 3]. Because it severely limits the conversion efficiency of a-Si:H-based films, many researchers have investigated SWE during

the past 30 years. Despite extensive investigations, there is no consensus on a microscopic mechanism that explains all the experimental phenomena. Similarly, the recovery mechanism for a-Si:H-based solar cells via thermal annealing [4, 5] or intense illumination at high temperature with a strong reverse bias [6] is still controversial. The recovery kinetics via thermal annealing is often interpreted by the stretched exponential (SE) kinetics [7, 8] wherein defect generation and annealing in a-Si:H-based materials is a dispersive process controlled by one type of metastable defect [9, 10]. However, several recent reports have provided experimental evidence that a-Si:H-based materials possess “fast” and “slow” metastable defects, and thus the total defect density cannot uniquely determine the state for the a-Si:H-based solar cells [11–14]. Yang and Chen suggested the existence of the fast and slow metastable defects in a-Si:H solar cells via two-step light soaking [11]; the cell subjected to an intense pre-soaking exhibits a thermal annealing behavior at the initial stage of 1-sun (AM 1.5, 100 mW/cm²) post-soaking, which could be linked to annealing of the fast metastable defects. Since these results are clearly contradictory to the conventional SE kinetic model, the author suggested a new model of recovery kinetics for pin-type a-Si:H-based solar cells employing the two-component (TC) metastable defect states [15]. We selected the fill factor (FF) as a measure of the state

for the a-Si:H-based solar cells due to the following reasons: (i) FF is sensitive to the quality of the intrinsic absorber (i-absorber); (ii) analytic equations relating FF to the collection length and to the defect density (or electron spin density) have been developed [16, 17]; and (iii) FF is the most degraded parameter against light-soaking in a-Si:H-based solar cells [18] and is less sensitive to the measurement temperature and illumination intensity compared to the open-circuit voltage and short-circuit current [4].

In this work, the TC model is simulated by varying its parameters. The model is also applied to experimental annealing data for the pin-type a-Si:H-based solar cells in order to verify the validity of the TC model. Finally, the microscopic mechanisms for the annealing behaviors of pin-type a-Si:H-based solar cells are discussed.

2. MODELING AND SIMULATION

If uniform E is assumed in i-absorber of pin-type a-Si:H-based solar cells, FF is governed by the carrier collection length (l_c):

$$l_c = \mu\tau E = \mu\tau \frac{(V_{bi} - V_a)}{d}, \quad (1)$$

where μ is the drift mobility, τ is the ambipolar carrier lifetime of photogenerated carriers, E is the internal electric field in the i-absorber, V_{bi} is the built-in potential in the solar cell, V_a is the biased voltage, and d is the thickness of the i-absorber. Faughnan and Crandall [16] reported the following empirical relation:

$$FF = C_0 + A \log \frac{l_c}{d}, \quad (2)$$

with $C_0 = 0.39$ and $A = 0.30$ [17].

Because τ is reciprocally proportional to the defect density in the i-absorber (N), if μ is assumed to be constant [17], then

$$FF \cong C_1 - A \log N = C_1 - K_1 \ln N, \quad (3)$$

where C_1 and K_1 ($= 0.30 \log e = 0.13$) are constants.

Accordingly, normalized FF (FF_n) can be expressed as

$$FF_n = \frac{FF(t) - FF_d}{FF_i - FF_d} = \frac{\ln(N/N_d)}{\ln(N_0/N_d)}, \quad (4)$$

where $FF(t)$ is FF as a function of time, FF_d is degraded FF via light soaking, FF_i is initial FF before light soaking, N_d is the defect density in the degraded i-absorber, and N_0 is the initial defect density in the i-absorber [8].

If it is assumed that the TC model has fast and slow metastable defect states (N_F and N_S), then $N = N_0 + N_F + N_S$ and $N_d = N_0 + N_{dF} + N_{dS}$. Furthermore, it is assumed that all the defects stem from a common pool of ground states, and there is otherwise no direct communication between the two

defect components under fixed annealing conditions. Thus, the system of rate equations for both components can be described as [11]

$$\begin{aligned} \frac{dN_F}{dt} &= G_F(N_T - N_F - N_S) - A_F N_F, \\ \frac{dN_S}{dt} &= G_S(N_T - N_F - N_S) - A_S N_S, \end{aligned} \quad (5)$$

where N_T is the total number of states which can be converted into defects, G_F and G_S are the respective constant defect generation coefficients for the fast and slow metastable defects, and A_F and A_S are the respective constant defect annealing coefficients for the fast and slow metastable defects. In the case of thermal annealing, we assume that $G_F = G_S = 0$ and $A_F \gg A_S$. Then, the rate equations for N_F and N_S can be expressed as the following simple first-order approximations:

$$\begin{aligned} \frac{dN_F}{dt} &= A_F N_F = -\frac{N_F}{\tau_F}, \\ \frac{dN_S}{dt} &= A_S N_S = -\frac{N_S}{\tau_S}, \end{aligned} \quad (6)$$

where τ_F and τ_S are time constants for the fast and slow metastable defects. Accordingly, N_F and N_S can be given by

$$\begin{aligned} N_F &= N_{dF} \exp\left(-\frac{t}{\tau_F}\right), \\ N_S &= N_{dS} \exp\left(-\frac{t}{\tau_S}\right). \end{aligned} \quad (7)$$

After replacement of N and N_d by (7), (4) becomes the following kinetic equation:

$$FF_n = \frac{\ln\{n[1 + \alpha \exp(-t/\tau_F) + \beta \exp(-t/\tau_S)]\}}{\ln n}, \quad (8)$$

where $n = N_0/(N_0 + N_{dF} + N_{dS}) = \exp[(FF_d - FF_i)/K_1]$ from (3), $\alpha (= N_{dF}/N_0)$ is the ratio of the photocreated fast metastable defect density to the initial defect density, and $\beta (= N_{dS}/N_0 = 1/n - 1 - \alpha)$ is the photocreated slow metastable defect density to the initial defect density. Therefore, annealing behaviors can be simulated by fitting three independent parameters, α , τ_F , and τ_S .

Figure 1 provides the simulated results as a function of t using (8). As shown in the figure, four different recovery rates are inspected with t : (i) initial fast rise when $t < \tau_F$, (ii) moderate increase when $\tau_S < t < \tau_S$, (iii) fast increase when $t > \tau_S$, and (iv) slow increase in the last tail. Figure 1(a) shows that the increase in τ increases with an increase in n . The rate of the increase decreases with the increase in n when $t < \tau_S$. An increase in the fraction of the fast metastable defect $[\alpha/(\alpha + \beta)]$ leads to a fast recovery, as shown in Figure 1(b). It is also found that τ_F is mainly responsible for the initial rise, whereas τ_S affects the last tail as well as the region when $t > \tau_S$ (see Figures 1(c) and 1(d)).

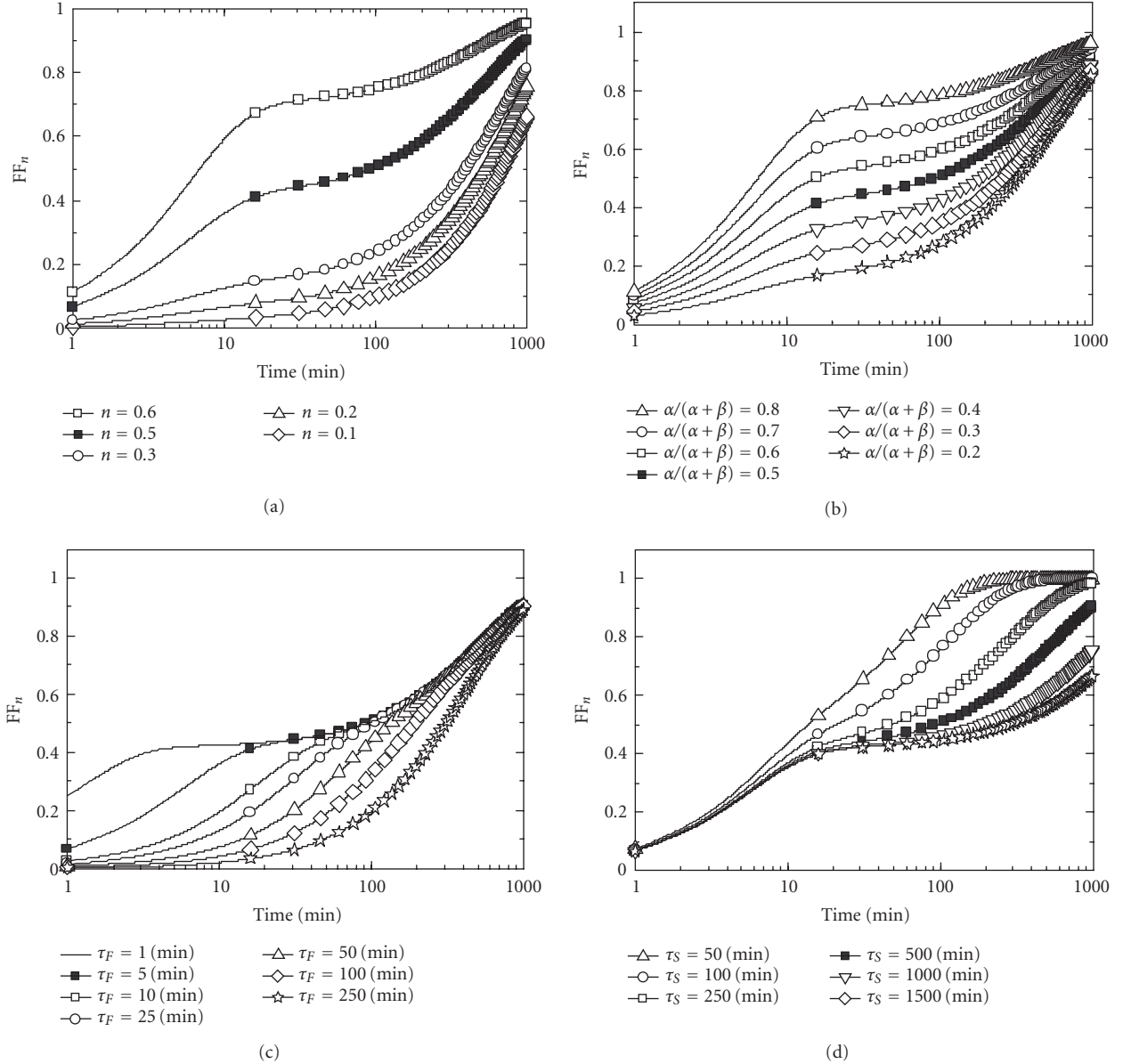


FIGURE 1: Simulation of annealing behaviors using the TC kinetic model; (a) with variable n , and fixed parameters of $\alpha/(\alpha + \beta) = 0.5$, $\tau_F = 5$ minutes, and $\tau_S = 500$ minutes, (b) with variable $\alpha/(\alpha + \beta)$, and fixed parameters of $n = 0.5$, $\tau_F = 5$ minutes, and $\tau_S = 500$ minutes, (c) with variable τ_F , and fixed parameters of $n = 0.5$, $\alpha/(\alpha + \beta) = 0.5$, and $\tau_S = 500$ minutes, (d) with variable τ_S , and fixed parameters of $n = 0.5$, $\alpha/(\alpha + \beta) = 0.5$, and $\tau_F = 5$ minutes. The symbols are modeled data included to distinguish each other.

3. RESULTS AND DISCUSSION

Figure 2 depicts the simulated results using the TC kinetic model for the experimental results with various thermal annealing temperatures (T_A) reported in [4]. The pin-type a-Si:H solar cell was fabricated via a dc glow discharge technique with a structure of glass/transparent conducting oxide (TCO)/hydrogenated p-type amorphous silicon-carbide (p-a-SiC:H)/undiluted i-a-Si:H (~ 520 nm)/n-type a-Si:H (n-a-Si:H)/Ti/Ag. FF_i and the initial efficiency are 0.70 and 9%, respectively. In all measurements, the cell was degraded under the 1-sun illumination at 40°C for 64 hours. Thus, FF_d

is always fixed at 0.55 ($n = 0.32$). With an increase in T_A , τ declines remarkably.

Figure 3 displays information on evaluated τ for the simulated data in Figure 2. Here, τ_i denotes the time for FF to recover $i\%$ of its total degradation, that is, $FF_i - FF_d$. As can be seen in Figure 3(a), τ_i is thermally activated, that is, $\tau_i = \nu_o^{-1} \exp(E_a/kT)$, where ν_o is the attempt frequency, E_a is the activation energy, k is Boltzmann's constant, and T is the absolute temperature. In [4], Bennett et al. demonstrated that the annealing behavior for their pin-type a-Si:H solar cells could be characterized a unique E_a of 1.2 eV. They selected τ_{50} as a measure of τ_i . From Figure 3(b), however, it

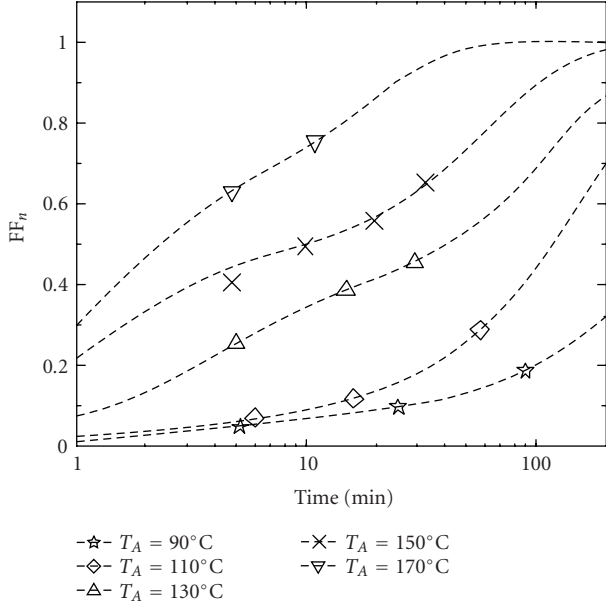


FIGURE 2: Recovery behaviors for the a-Si:H solar cell via thermal annealing as a function of T_A (from [4]). The symbols and dotted lines denote experimental data and simulated data using the TC kinetic model, respectively.

should be noted that E_a for pin-type a-Si:H-based solar cells depends on τ_i and the TC model present a gradual decrease in E_a and v_o with an increase in i . These phenomena are mainly caused by the existence of the fast and slow metastable defects in the TC model.

Figure 4 compares the simulated results using the TC model for the experimental results reported in [6]. The pin-type solar cell was fabricated at $\sim 200^\circ\text{C}$ via a plasma-enhanced chemical vapor deposition (PECVD) technique [5] with a structure of glass/SnO₂/p-a-SiC:H/H₂-diluted i-a-Si:H (~ 300 nm)/n-a-Si:H/ZnO/Al. At the open-circuit, the cell was degraded under the 50-sun illumination at 60°C for 30 minutes. Hence, n is always fixed at 0.43 by keeping FF_i and FF_d at 0.67 and 0.56, respectively. Then, the experimental recovery behaviors were inspected under 50-sun illumination with different V_a . It was found that FF for the pin-type H₂-diluted a-Si:H solar cell recovered more rapidly under intense irradiation at $T_A = 70^\circ\text{C}$ with an increase in reversed V_a . τ_F and τ_S for the TC model all gradually decrease with the increase in reversed V_a , thus indicating the high electric field induced fast recovery. The reduced τ is mainly due to the decrease in τ_S . It is clear that the TC model fits the experimental data well in the initial rise and last tail. Furthermore, the reported different recovery kinetics from identical FF_d , which depend on the illumination intensity [11] or temperature [4] during the light-induced degradation, prove that the photocreation of DBs, that is, $N_d - N_0$, is composed of different kinds of defects.

The author have developed hydrogenated protocrystalline Si (pc-Si:H) multilayer absorbers [18]. The pc-Si:H material is a highly H₂-diluted a-Si:H material existing just

TABLE 1: Fitting parameters used in Figure 5.

	n	$\alpha/(\alpha + \beta)$	τ_F (min)	τ_S (min)
undiluted a-Si:H	0.39	0.82	3.7	214.6
pc-Si:H multilayer	0.57	0.84	3.3	59.5

below the threshold of the a-Si:H-to- μc -Si:H transition. Using a photoassisted chemical vapor deposition (photo-CVD) technique, we prepared alternately H₂-diluted i-pc-Si:H multilayers by modulating the mass flow control of the hydrogen dilution ratio (H₂/SiH₄), and thereby i-pc-Si:H has the repeatedly layered structure of low H₂-diluted a-Si:H sublayers and highly H₂-diluted sublayers. The highly H₂-diluted sublayers possess isolated nanosized Si (nc-Si) grains embedded in a-Si:H matrix [19]. The i-pc-Si:H multilayers exhibit a fast light-induced metastability with a low degradation. Consequently, highly stabilized solar cells (stabilized efficiency = 9.0%) were achieved without using any back reflector [20, 21]. The pc-Si:H multilayer solar cells exhibit a very fast annealing behavior during 1-sun post-soaking, compared to a-Si:H solar cells [22], which is the clear evidence of the two-component metastable defects in the pc-Si:H multilayers. Figure 5 shows the fitted results using the TC model for the experimental recovery behaviors in an undiluted a-Si:H solar cell and a pc-Si:H multilayer solar cell in [8]. The pin-type solar cells were fabricated at 250°C via a photoassisted chemical vapor deposition (photo-CVD) technique with a structure of glass/SnO₂/p-a-SiC:H/p- μc -Si:H/i-absorber (~ 550 nm)/n- μc -Si:H/Al. The initial efficiency for the undiluted a-Si:H and pc-Si:H multilayer solar cells are 10.6 and 8.9%, and FF_i values are 0.73 and 0.69, respectively. The solar cells were degraded under the 1-sun illumination at 48°C for 20 hours. After light soaking, the pc-Si:H multilayer cell stabilized with $n = 0.57$, while the undiluted a-Si:H cell is still in degrading state with $n = 0.39$. The fitting parameters of the cells are listed in Table 1. Thermal annealing is performed in a vacuum chamber. The pc-Si:H multilayer cell exhibits the rapid recovery than the undiluted a-Si:H cell.

From the fast metastability and annealing behaviors of pc-Si:H multilayer solar cells, the vertically regular distribution of the isolated nc-Si grains [21] and the improved medium-range-order in the a-Si:H matrix [23] are considered to localize the photocreation near the grain boundary regions [19], and thereby suppress the photocreation of slow metastable defects in the pc-Si:H multilayers. From the visible photoluminescence (PL) peak measured at room temperature, the isolated nc-Si grains tend to act as radiative recombination centers of captured carriers, which may contribute to the good stability [14].

Because the pc-Si:H multilayer has a slightly wider optical band gap (effective band gap > 1.7 eV) than conventional a-Si:H layers (~ 1.7 eV), the pc-Si:H multilayer solar cell is promising as a top cell for a high-efficiency tandem cell [23]. The pc-Si:H multilayer/ μc -Si:H (~ 1.1 eV) double-junction tandem structure opened the possibility of a significantly

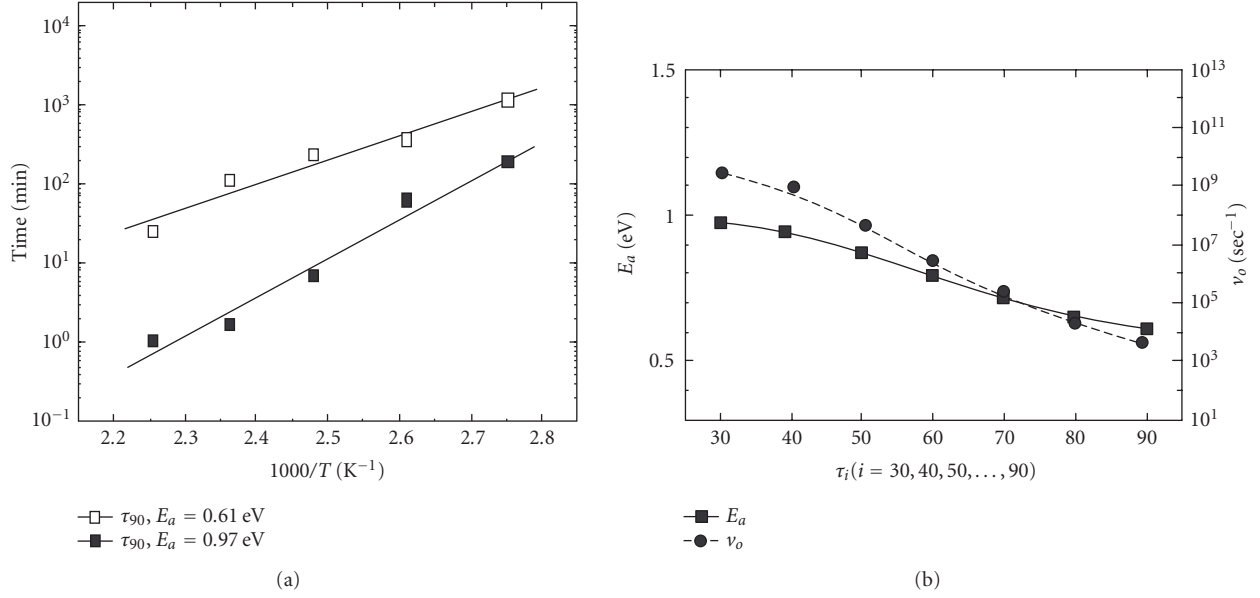


FIGURE 3: Information on evaluated τ for the simulated data in Figure 2; (a) Arrhenius plot for τ_{30} and τ_{90} . The calculated values of E_a for τ_{30} and τ_{90} are provided. (b) E_a and ν_0 versus τ_i .

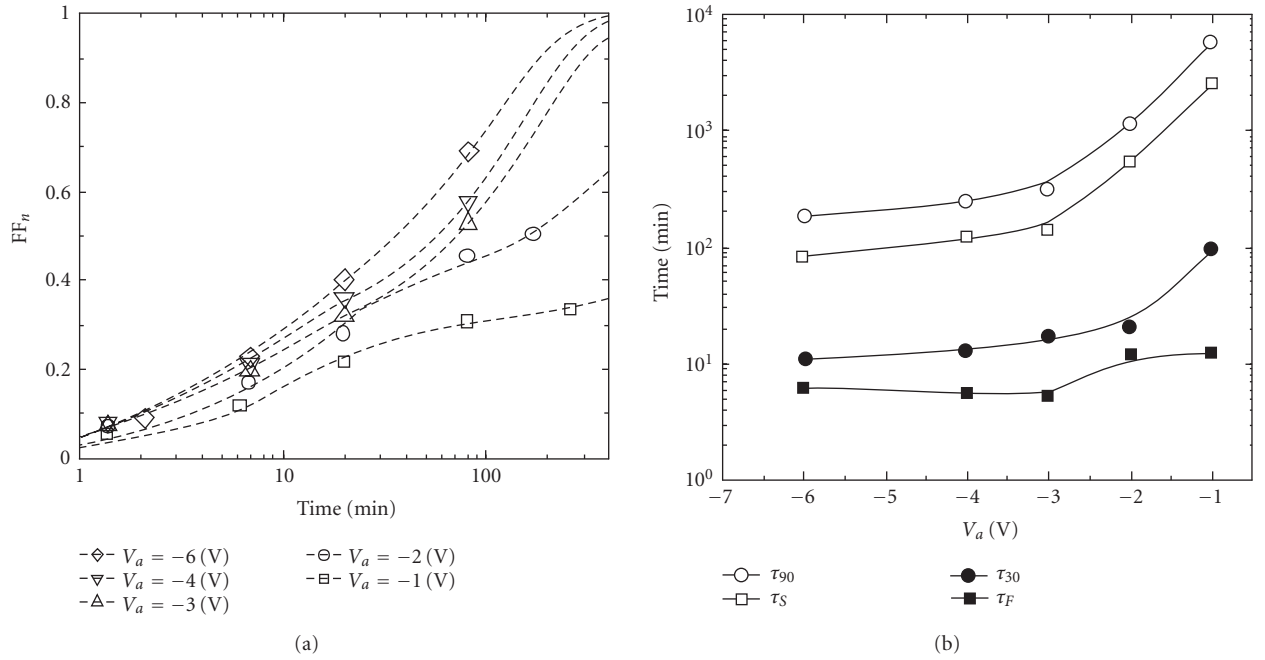


FIGURE 4: Annealing for the H₂-diluted a-Si:H solar cell under intense (50-sun) illumination at $T_A = 70^\circ\text{C}$ (from [6]); (a) recovery behaviors with different V_a and (b) τ_{30} and τ_{90} versus V_a . The symbols and dotted lines denote experimental data and simulated data using the TC kinetic model, respectively.

high-stabilized efficiency due to a low-degradation ratio [24].

The photocreation of DBs in an a-Si:H matrix can occur with a value of E_a [25] that is lower than E_a for light-induced long range H diffusion (0.9 eV) [26]. Hence, the breaking of strong Si-H bonds (~ 3 eV), proposed by Branz in the hy-

drogen collision model [27], cannot be easily accomplished by the nonradiative recombination of electron-hole pairs. Instead, the breaking of weak Si-Si bonds due to nonradiative recombination of electron-hole pairs is considered today as a plausible origin of SWE [28, 29]. Recently, Powell et al. [29] proposed the creation of two metastable HSiDB (complex of

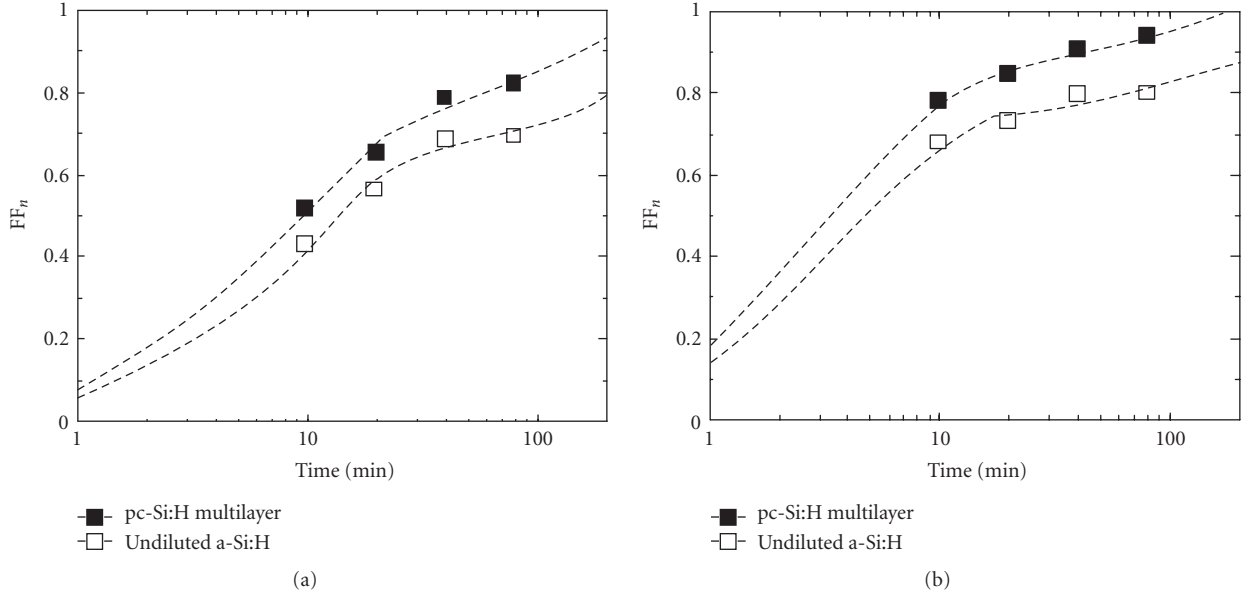


FIGURE 5: Comparison of the recovery behavior via thermal annealing between the undiluted a-Si:H and pc-Si:H multilayer solar cells (from [8]); (a) $T_A = 108^\circ\text{C}$ and (b) $T_A = 133^\circ\text{C}$. The symbols and dotted lines denote experimental data and simulated data using the TC kinetic model, respectively.

a DB and an Si–H bond, where H locates in a tetrahedral-like site (T_d), not a bond-centered site) defects; an H atom from a neighboring doubly hydrogenated weak Si–Si bond (HSiSiH) switches to a T_d site of the broken Si–Si bond and the other H from HSiSiH is also located in the energetically suitable T_d site. This model is reasonable because the spatial separation between H in the T_d site and DB is in agreement with the observed values of 4–5 Å by electron spin resonance (ESR) measurements [30]. On the other hand, it has been reported that annealing of DBs in a-Si:H films, which are photocreated at moderate temperature, has a similar value of E_a (1.1–1.2 eV) [31] to that for long range H diffusion (1.5 eV) [32]. This supports the supposition that long range H diffusion plays an important role in annealing of metastable defects. Thus, the hydrogen collision model is valid in the case of annealing and E_a can be interpreted as the energy for the thermal emission of H from a T_d site. In addition, ν_o corresponds with the phonon frequency of Si–H bonds [29] and with the thermal emission process. However, it is difficult to define unique E_a and ν_o for τ , because it varies with τ_i (see Figure 3(b)). Nevertheless, E in a solar cell is expected to lower the energy barrier for long range H diffusion [6]. From Figure 4, it can be concluded that the intense illumination at a high temperature also thermally emits mobile H by breaking Si–H bonds, and leads to high electric field induced annealing for the solar cells.

In the previous report [15], the author proposed the following mechanism for the recovery kinetics in pin-type a-Si:H-based solar cells based on the TC model: (i) mobile H is thermally emitted from a metastable HSiDB defect by breaking the Si–H bond. As reflected in Figure 2, the elevated T_A increases the thermal emission rate of mo-

bile H [32]; (ii) emission of bonded H is followed by weak Si–Si bond reconstruction. Meanwhile, mobile H migrates through the lattice [33, 34] and is subsequently captured at another weak Si–Si bond, which eventually forms an HSiDB defect. It should be noted that E in i-absorber assists the migration of mobile H to a shallower site, which is reflected in Figure 4; and (iii) thermal emission and recapturing processes of mobile H proceed until it is captured at the DB site of a metastable HSiDB defect, resulting in the annealing of two defects. Consequently, for the annealing process to reform the a-Si:H matrix, a considerable energy is required and many H atoms are involved. However, an internal field formed in solar cells can reduce the required energy for the annealing process. It should be noted that the classification between the fast and slow metastable defects is mainly determined by their activation energy and capture cross-section [14]. This classification is only relative and is dependent on the annealing conditions, that is, elevated T_A or V_a can increase α .

4. CONCLUSIONS

The author simulated the TC model in order to understand the effect of each parameter on the annealing kinetics in pin-type a-Si:H-based solar cells. This model displays that E_a depends on the definition of the recovery time. It is verified that the TC model fits the various experimental data. Thus, the TC model can be deemed useful for limiting the number of mechanisms responsible for annealing the photocreated defects in a-Si:H. From the thermally activated and high electric field induced nature of annealing behaviors, the plausible microscopic mechanism on the DB defect removal related to the

thermal emission of mobile H from T_d sites and long range H diffusion was discussed.

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