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A statistics modeling approach for the optimization of thin film photovoltaic devices

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ABSTRACT

The growing interest in exploring thin film technologies to produce low cost devices such as *n-i-p* silicon solar cells, with outstanding performances and capability to address the highly relevant energy market, turns the optimization of their fabrication process a key area of development. The usual one-dimensional analysis of the involved parameters makes it difficult and time consuming to find the optimal set of conditions. To overcome these difficulties, the combination of experimental design and statistical analysis provides the tools to explore in a multidimensional fashion the interactions between fabrication parameters and expected experimental outputs.

Design of Experiment and Multivariate Analysis are demonstrated here for the optimization of: (1) the low temperature deposition (150 °C) of high quality intrinsic amorphous silicon (*i*-a-Si:H); and (2) the matching of the *n*-, *i*-, and *p*-silicon layers thickness to maximize the efficiency of thin film solar cells. The multiple regression method applied, validated through analysis of variance and evaluated against exact numerical simulations, is shown to predict the overall intrinsic layer properties and the devices performance.

The results confirm that experimental design and statistical data analysis are effective approaches to improve, within a minimum time frame and high certainty, the properties of silicon thin films, and subsequently the layer structure of solar cells.

1. Introduction

It is well known how complex the optimization of silicon thin films can be. When the optimization process relies on one-dimensional analysis it can, not only be misleading, but also difficult and time consuming. One dimensional analysis, such as the OVAT system (One Variable At a Time) (Injac et al., 2008), finds the optimal set of conditions by randomly varying factors (fabrication conditions) in a sequence. However, there is a major drawback in this method as it assumes that there is no interaction between the factors, which is far from reality particularly in the photovoltaics field. As more variables are in play, the more difficult it becomes to grasp all the interactions between factors, hence there is a higher probability to miss the optimum combination which can only be reached at the expense of more experiments (thus, time) and resources.

On the other hand, with simulation guidance, namely Design of Experiment (DoE) and Multivariate Analysis (MVA), one has the tools to explore in a multidimensional fashion the interactions between deposition parameters (input variables, or factors) and expected outputs (film or device properties).

Computational simulation over the past 30 years evolved to the point where it helps researchers to validate proposed physical structures and predict experimental results. One of the earliest and simplest methods of simulation guidance, applied to thin film modeling, are the least squares estimation for curve fitting (Appelbaum et al., 1992) and a statistical and simulation method (Marvin, 1988) proven to be in agreement with experimental data considering the associated uncertainty.

Other modeling tools were developed over the years, hand in hand with the evolution of computer processing and calculation power (Lee et al., 2011; Geisler et al., 2000; Brenot et al., 2000). These soft computing methods offer an invaluable advantage over analytical studies, since the modulation does not require extensive information and understanding of the system (Bae et al., 2010), as

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they account for experimental uncertainty, systematic errors and uncontrollable variables in the calculations.

Though in fields like organic and bio-chemistry (Guo et al., 2007), pharmacy (Ye et al., 2000) and food technology (Fekete et al., 2012), DoE and computer modeling techniques are common practices; in the photovoltaic (PV) field they have so far been less frequently applied (Bae et al., 2010; Satake et al., 2002). Nonetheless, the application of DoE to study thin films has grown recently. For example, applications of DoE are found in the study of electrochromic films (Wojcik et al., 2013), transparent conductive oxides (Kao et al., 2012; Kumar et al., 2013; Chang et al., 2000), indium sulfide (In_2S_3) (Wang et al., 1879), aluminum nitride (AlN) (Akiyama et al., 1998) and silicon nitride films (Dergez et al., 2013; Sharma et al., 2011); despite the fact that the latter is often object of study with neural networks (Lee et al., 2011; Seung-Soo et al., 1996; Liau et al., 2002; Kim et al., 2003).

Given the importance of photovoltaics to address the massive future energy demands, a line of research based on DoE and MVA is essential for further development of low cost but high efficient solar cell (SC) manufacturing processes. This is of particular importance due to the complex interplay between the different physics (optics, solid state physics, electronics, etc.) involved in the mechanism of sunlight-to-electricity conversion and the fabrication techniques of such devices (*e.g.* by plasma enhanced chemical vapor deposition - PECVD) which combine both chemical and physical processes that require careful tuning to provide optimally-engineered structures. Such volume of parameters and constrains makes data reproduction from literature often impossible, given the fact that each manufacturing system is unique and, therefore, the device production should be tailored to its specificities.

To this end, a novel method is proposed in the present work to optimize the fabrication processes *via* the statistical reinterpretation of previously obtained experimental data. As a proof-of-concept, this method was applied here to improve the fabrication of intrinsic thin film silicon (*i*-a-Si:H) layers and a non-conventional solar cell structure with single *n*-*i*-*p* layers, grown by PECVD at low temperature (150 °C). The low temperature deposition and *n*-*i*-*p* configuration enables the potential application of such devices to a broad range of non-conventional and opaque substrates (e.g. polymeric, paper-based (Vicente et al., 2015)), that can degrade under the usual fabrication high temperatures (200–300 °C) applied in PECVD, and is another step forward for truly low-cost and disposable electronics.

Fig 1 summarizes the structure of the method and the terminology used throughout this article. The optimization method starts with the delineation of the regime of interest and experimental conditions to be tested (DoE) in the corresponding study (*i*-layer or SC layers thickness combination). In the input stage, the data for the MVA is gathered, namely the fabrication parameters (factors $X_i (i = 1, 2, 3)$) and the corresponding film/device properties (responses $A_n (n = 1, 2, \ldots, p)$), after fabrication and characterization. The next stage (Mathematical fitting) deals with the actual modeling and fitting. The model output is the multidimensional combination of the optimum predicted characteristics which give rise to the best combination of fabrication conditions to produce high quality films/SCs. The experimental data, arriving from the fabrication and characterization of films/SCs according to the model predictions, can also re-feed the model to refine it.

Two different statistical modeling studies were performed (Fig. 1b) to evaluate the capabilities of the method: (1) First, the optimization of the *i*-layer optoelectronic and morphological properties, through the variation of the deposition parameters, within the relevant experimental space; (2) second, the optimization of the thickness combination between the *n*-, *i*-, *p*-layers of the solar cell structure; the optimum *i*-layer obtained in (1), according to the model prediction, was used in the second study as the SC active layer. For each study, the model receives an initial set of inputs (the factors which correspond to the fabrication parameters, and the responses as the measured film/device properties - optical, electrical and morphological) to predict the optimum combination of parameters and achieve high quality thin films/SCs. The table 1 summarizes all the analyzed quantities.

2. Statistical system description

The first step consists in sorting out the relevant combination between the input parameters by applying a stepwise regression to the initial experimental data pool, which is presented in detail in Section 3.4. The stepwise regression is an approach that facilitates searching and selecting the appropriate regression model, particularly when there is little theory to guide the selection of terms for a model, as in the current case. Moreover, it is assumed that the errors are independent and identically distributed with a zero mean and common variances.

The mathematical fittings, calculated by the stepwise regression, comprise three-way full factorial and polynomial quadratic functions. Those calculations are performed individually for each response, film/SC property (A_n), to decrease the fitting error associated to the limited experimental data. The information regarding the model validation and significance of the factors and interactions that make up the fitting function can be found in supplementary information (see Fig. S1). The obtained fitting function, described by equation 1, predicts how each film/SC property $A_n (n = 1, 2, \ldots, p)$ varies within the experimental space; where $p = 6$, in the case of intrinsic thin film optimization (first optimization step), and $p = 4$ for SC thickness combination (second optimization step). The experimental space (S) is defined by the factors (fabrication parameters $X_i (i = 1, 2, 3)$), i.e. P_W, P_{gas}, D_H, in the first study, and *n*-, *i*-, *p*-layer thickness, in the second study. β_i are the coefficients of interaction and ε represents the random error of the model.

$$A_n = \sum_{i=1}^{3}\beta_i X_i + \sum_{i=1}^{2}\sum_{j=i+1}^{3}\beta_{ij}X_iX_j + \sum_{i=1}^{1}\sum_{j=i+1}^{2}\sum_{k=j+1}^{3}\beta_{ijk}X_iX_jX_k + \sum_{i=1}^{3}\beta_{ii}X_i^2 + \varepsilon \quad (1)$$

To conduct a MVA, the individual responses were then combined in a single experimental space. This way, one can identify the optimum regime and determine which fabrication parameters should be then tested to obtain improved *i*-a-Si:H/SCs.

The calculation of the optimum set of fabrication conditions follows the inclusion-exclusion principle (Koshy, 2002), where A_n are finite sets within the experimental space ($A_n \subset S$). To determine the optimum set, one can exclude the union of all the sets whose combination of fabrication parameters give rise to poor performance characteristics within the universe S, $Y_n < A_n(X_iX_jX_k) < Z_n$ (Y and Z are continuous numerical values obtained from solving the response equation). The optimum combination of deposition parameters is therefore the complementary of the sets (A_n') in the union, as given by Eq. (2).

$$\left|\bigcap_{n=1}^{p} A_n'\right| = \left|S - \bigcup_{n=1}^{p} A_n\right| = |S| - \sum_{n=1}^{p}(-1)^{n+1} \sum_{1\leqslant q_1<\ldots<q_n\leqslant p} \left|A_{q1}\cap\ldots\cap A_{qn}\right| \quad (2)$$

Finally, the predicted optimum fabrication parameters were tested through validation experiments. Further improvements to the model reliability can be made by introducing the new experimental data in the model to perform a refined fitting iteration.