CrossMark

Modelling water vapour permeability through atomic layer deposition coated photovoltaic barrier defects

Mohamed Elrawemi [a,*], Liam Blunt [a], Leigh Fleming [a], David Bird [b], David Robbins [b], Francis Sweeney [a]

[a] EPSRC Centre for Innovative Manufacturing in Advanced Metrology, School of Computing and Engineering, University of Huddersfield, Huddersfield, UK
[b] Centre for Process Innovation Limited, Sedgefield, County Durham, UK

ABSTRACT

Transparent barrier films such as Al_2O_3 used for prevention of oxygen and/or water vapour permeation are the subject of increasing research interest when used for the encapsulation of flexible photovoltaic modules. However, the existence of micro-scale defects in the barrier surface topography has been shown to have the potential to facilitate water vapour ingress, thereby reducing cell efficiency and causing internal electrical shorts. Previous work has shown that small defects (≤ 3 μm lateral dimension) were less significant in determining water vapour ingress. In contrast, larger defects (≥ 3 μm lateral dimension) seem to be more detrimental to the barrier functionality. Experimental results based on surface topography segmentation analysis and a model presented in this paper will be used to test the hypothesis that the major contributing defects to water vapour transmission rate are small numbers of large defects. The model highlighted in this study has the potential to be used for gaining a better understanding of photovoltaic module efficiency and performance.

© 2014 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

1. Introduction

In today's industry, the most common type of solar photovoltaic (PV) cell is fabricated from either rigid crystalline silicon or thin-film materials [1]. The rigid construction of Si solar cells hampers their economic integration into residential and commercial buildings; however, thin film solar cell technologies may prove to be most appropriate with respect to cost, ease of manufacture and installation. These thin film cells are based on the material $CuIn_{1-x}GaxSe_2$ (CIGS) as the absorber layer (p-type) and they at present have efficiency levels at or beyond that of Si based rigid solar modules [2]. The key weakness of these cells however is their moisture sensitivity. This is a critical problem if this technology is expected to meet the requirements of international standard IEC61646 [3] which requires that all PV modules survive 1000 h in an environment of 85 °C and 85% relative humidity (RH) [3]. At the present time, no cost effective, flexible transparent encapsulation can fulfil the requirements of the water vapour transmission rate (WVTR) for flexible PV modules [4]. The WVTR of current barriers is in the range of 10^{-1} g/m²/day, while it should not be higher than 10^{-4} g/m²/day to assure life-times of 20 years and more [4,5].

Therefore, a robust, transparent flexible encapsulation method for flexible PV modules is needed.

Thin layers of aluminium-oxide (Al_2O_3) of the order of a few tens of nanometres deposited via the atomic layer deposition (ALD) technique have been introduced to allow PV module transparency and flexibility and to provide an effective barrier layer. These barrier films ideally have WVTR of less than 10^{-4} g/m²/day [6]. The term 'barrier' here refers to the ability of Al_2O_3 to resist the diffusion of water vapour into and through itself. Nevertheless, the barrier properties are often influenced by a wide range of variables, making conclusions regarding film properties sometimes difficult. It is known that barrier film permeability can be affected by the chemical and physical structures of the barrier, concentration of the permeant, temperature and humidity [7,8] as well as surface defects on the barrier coating that may be induced during the deposition processes [9,10]. Da Silva Sobrinho et al. [11] stated that the source of defect-driven permeation has been primarily attributed to pinhole defects [12,13] though more recent studies have shown that in the absence of pinhole defects permeation rates are still reduced by three orders of magnitude over the substrate material [14]. The remaining permeation is shown to be the result of defects in the sub-micromere to several micrometre range, produced by the surface microstructure [15] and/or low density of the films [14–16]. More detailed reviews of permeation mechanisms and the performance of various permeation barriers have been given elsewhere [12,17]. In this paper a theoretical model is presented to allow the prediction of the amount of water vapour permeation through PV barrier film defects. The results of the model are then

* Corresponding author at: EPSRC Centre for Innovative Manufacturing in Advanced Metrology, School of Computing and Engineering, University of Huddersfield, Huddersfield, HD1 3DH, UK.
E-mail addresses: Mohamed.elrawemi@hud.ac.uk, elrawemi@yahoo.com (M. Elrawemi), David.Bird@uk-cpi.com (D. Bird).

compared to experimental results where defects measured using surface metrology techniques are correlated with WVTR.

2. Theoretical model

Ashely [18] developed an equation to calculate the permeability co-efficient of the water vapour through a polymer barrier film. The equation was based on Henry's law of solubility [19], Fick's laws of diffusion [20], Stefan and Exner's findings [21] and Von Wroblewski hypothesis [22]. Ashely [23] indicated that the permeability coefficient P_r depends on the solubility coefficient, S, as well as the diffusion coefficient, D. Eq. (1) expresses the permeability in terms of solubility and diffusivity, D, and it can be defined as the volume of vapour passing through a unit area of the barrier layer per unit time, with a unit pressure difference across the sample [23].

$$P_r = \frac{(\text{quantity of permeant}) \times (\text{film thickness})}{(\text{area}) \times (\text{time}) \times (\text{pressure drop across the film})}$$

$$P_r = DS = \frac{qL}{At\Delta P} \left(\frac{cm^3 cm}{cm^2 \cdot s \cdot Pa} \right) \qquad (1)$$

where q is the amount of permeant passing through a film of thickness L and over area A during time t driven by a partial pressure differential Δp across the film [24]. In a typical water vapour permeation measurement, for example, a "MOCON" test, ΔP in Eq. (1) corresponds to the partial pressure difference between nitrogen containing water vapour at 90% RH on one side, and ultra-pure nitrogen on the other side. In this type of permeation test there is no pressure gradient across the sample so it is then reasonable to use the absolute value of the permeant's partial pressure P, instead of ΔP [11] Thus, Eq. (2) can be presented as the following;

$$P_r = DS = \frac{qL}{AtP} \left(\frac{cm^3 cm}{cm^2 \cdot s \cdot Pa} \right). \qquad (2)$$

Da Silva Sobrinho et al. [11] developed an equation to determine the amount of permeant per unit of time (Q) through the polymer; this equation was based on Henry's law of solubility [19]. This equation is mathematically expressed as;

$$Q = \frac{q}{t} = \frac{ADSP}{L} = \frac{AD\varnothing}{L} \qquad (3)$$

where the validity of Henry's law is assumed, and Ø represents the water vapour concentration in the film surface and it has been estimated to be 1 g/cm³ [11]. For the case of water vapour which has a little [25] or even no interaction with the barrier film [11,26], the water vapour transmission is completely governed by defect geometries and densities [11]. In the present paper a model of water vapour permeation through the barrier defects is presented to study the effect of the defects on water vapour permeation.

2.1. Single defect case

The basic assumption is that the combined film of thickness L is made up of a transparent flexible barrier coating of (Al_2O_3) with a single circular hole (defect) of radius (R_0), and that it is exposed to permeant water vapour from the lower side as shown in Fig. 1.

Considering only steady-state permeation, where temperature and partial pressure of the water vapour are constant, and the total pressure is the same on both sides of the barrier layer. The next step is to determine the amount of the water vapour q_H, leaving the barrier film, see Fig. 1. In steady state, this amount is clearly determined by the water passing through the defect in the barrier. However, in the case of the
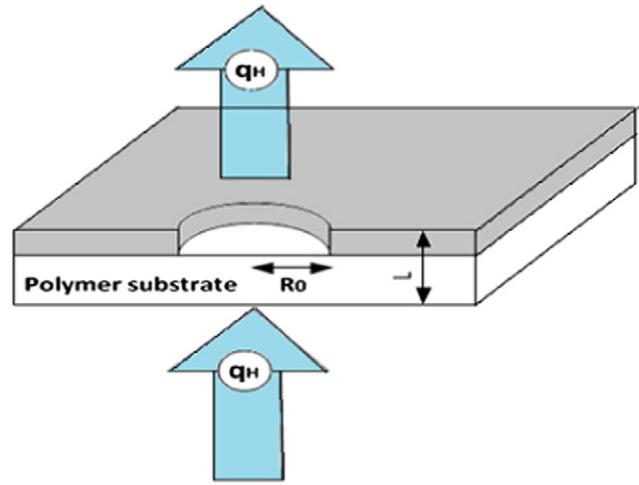

Fig. 1. A schematic representation of a hole type defect in a coated barrier film.

hole in the barrier film as shown in Fig. 1, the amount of permeant traversing the polymer and through the hole per unit time can be provided by modifying Eq. (3) and introducing the barrier film as having a circular "hole" area (πR_0^2).

$$Q = \frac{q_H}{t} = \frac{\pi R_0^2 D \varnothing}{L} \qquad (4)$$

where R_0 is the hole radius, D is the diffusion coefficient of the barrier film (cm²/s), $\varnothing$ is the water vapour concentration (g/cm³) and L is the combined film thickness. However, to determine the rate of the water vapour that penetrates hole over the substrate area (g/m²/day), Eq. (4) can be expressed as the following [24];

$$WVTR = \frac{Q}{A} \quad (g/m^2/day) \qquad (5)$$

where Q is the amount of the water vapour passing through a film of thickness L and area A during time t driven by a partial pressure differential P across the film [23].

2.2. Case of many defects

Independent holes assume that the presence of one does not affect water vapour permeation through the other, so that their respective quantities of water vapour permeation are additive [27]. So far, a theoretical model to determine the amount of the water vapour per unit of time, traversing a single hole in a barrier coating has been assumed and in order to discuss water vapour permeation through a barrier coating containing numerous defects (holes), Eq. (5) can be modified for (N) holes as follows:

$$WVTR = \sum_{0}^{N} \left(\frac{Q}{A} \right) N \qquad (6)$$

N, is the number of defects (holes) in the sample area.

3. Experimental details

The experimental study was based on a set of two 80 mm diameter samples. These two samples were supplied by the Centre for Process Innovation (CPI), and were coded as 2705 and 2706. These two samples are coated with 40 nm of Al_2O_3 using ALD technique [28]. The ALD depositions were made using Oxford Instruments FlexAL tool, where trimethyl aluminium was used as the metal precursor [29]. The reactor temperature used to deposit the aluminium oxide was 120 °C and the