Process Practice and its Effects on Surface Defects on Flexible PV Barrier Films

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ABSTRACT

To the present day, there is still an increasing interest in the development of Cu (In, Ga) Se_2 (CIGS) thin film solar cells on flexible polyimide substrates. These cells offer advantages of low cost, light weight and excellent radiation hardness as well as building-integrated-photovoltaic (BIPV) applications. These solar cells typically consist of six different layers of thin film including; Ag / Transparent Conductive Oxide (TCO) of ZnO: Al /CdS/CIGS/Mo/Al_2O_3 [1]. The Al_2O_3 insulating layer is additionally introduced into the structure of the solar cell in order to prevent diffusion of water vapour and oxygen ingress. Hence, optimize the photovoltaic (PV) module performance and lifespan, a study to understand the nature of surface topography defects for the Al_2O_3 barrier layer which allow the water vapour and oxygen ingress to the active layer (CIGS) is required. Surface metrology techniques including; Optical Microscopy, White Light Scanning Interferometry (WLSI), Atomic Force Microscopy (AFM) and Scanning Electron Microscopy (SEM) were employed for collecting and transmitting enormous amount of data for PV barrier layers defects down to nm scale. Feature segmentation analysis method proved to be an effective tool for discrimination between insignificant and the most significant features which are directly responsible for PV module degradation.

Keywords: Photovoltaics, Defects, Surface topography, ALD and WVTR.

1. INTRODUCTION

Thin film solar cells with a Cu (In, Ga) Se_2 (CIGS) absorber layer are a promising candidate for high efficiency and low cost photovoltaics. However, they are highly susceptible to long term environmental degradation; hence the encapsulation of these cells is required to protect them against moisture and oxygen from the ambient. Traditionally, glass is used as encapsulating material for many rigid-based solar cells because of its negligible water vapour transmission rate (WVTR). However, robust, transparent and flexible materials are required in the case of flexible CIGS cells. Replacing glass with flexible plastic material would reduce the weight, improve the durability, increase the flexibility, and help enable low-cost, continuous manufacturing for flexible PV modules. Studies regarding the encapsulation materials for CIGS PV modules was carried out by Carcia *et al* (2010), the authors demonstrated that an encapsulation layer of 55 nm Al_2O_3 deposited by atomic layer deposition (ALD) on top of a CIGS cell as shown in Fig (1) provides excellent moisture permeation protection [2]. The atomic layer deposition (ALD) is a unique process that produces dense, highly conformal, nearly pinhole-free thin films that are ideal for gas diffusion barriers. The capability to deposit virtually pinhole-free inorganic films at low temperature renders ALD a good candidate technique for this application.

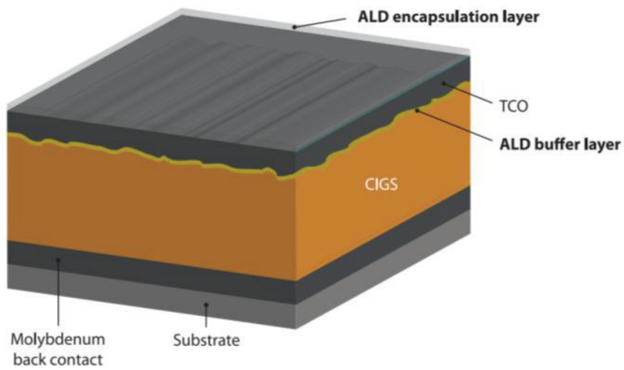

Fig (1) ALD encapsulation layer for CIGS PV module [3]

1. ENCAPSULATION PERFORMANCE NEEDS

For many years, one of the long-term goals of Photovoltaic researchers is to promote the development of cost-effective photovoltaic modules with more than 20 years useful lifetimes. Significant progress has been made, but additional research is still required and manufacturers will have to establish rigorous quality control standards in order to achieve the long-term goal. Lifetime improvement and prediction of the PV modules requires detailed information on the degradation mechanisms in the field and in accelerated aging tests. For applications in which PV modules are used in harsh environment, it is required that the Al_2O_3 barrier film withstand high humidity and temperature or aqueous environment. According to international standard (IEC 61646), A WVTR of ~10^{-1} g/m^2/day is sufficient for most packaging applications, but ≤10^{-6} g/m^2/day is required for encapsulation of long-life flexible PV modules as shown in fig (2). To achieve a lifetime of about 10.000 h, the estimated WVTR should be around 10^{-6} g/m^2/day and the OTR should be around 10^{-3}cm^3/m^2/day at room temperature [4]. In this paper, new method for studying and predicting the stability of thin-film PV modules based on feature segmentation analysis have been developed.

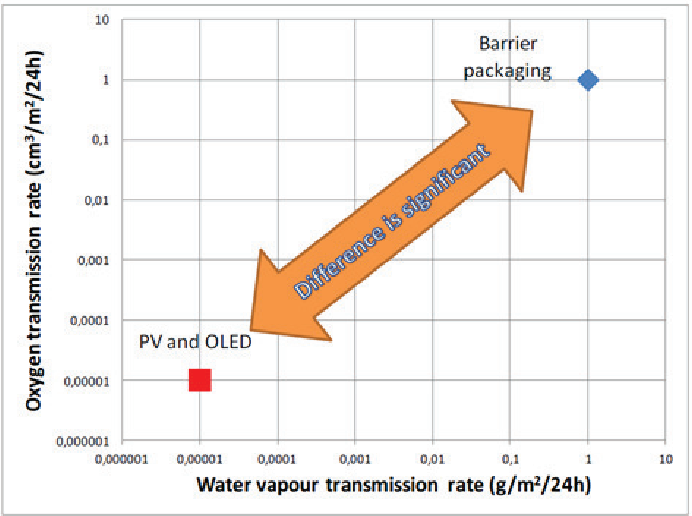

Fig (2) OTR versus WVTR requirements for different applications [4]

3. EXPERIMENTAL SECTION

Harsh environment can cause high diffusion rates of water vapour or moisture across the barrier if the film has a high effective diffusion coefficient and high defects density. Reduction in the diffusion coefficient can be obtained through barrier films with low defect concentrations, and this could be achieved through an optimal vacuum deposition processes. In this study, all Al_2O_3 ALD samples were prepared in the following manner; the substrate material used was Polyethylene naphthalate (PEN), and the thickness of this material is specified to be 125 microns. This material has a good transparency and relatively low cost but it has a high density of “pits” from fillers and belt marks and other defects from dust and surface scratching. These features are believed to be a source of shunt defects in the dielectric [5]. As a result, the surface quality of the substrate is essential to reduce pixel defects to an acceptable level. Therefore, another PEN thin-layer of approximately 3-4 microns thick was applied on top of the PEN substrate to planarise the pits and spikes features. Following to that a barrier film of 40nm thick of Al_2O_3 has been deposited by thermal and plasma-assisted ALD employing Tri-methyl aluminium Al $(CH_3)_3$ precursor dosing together with H_2O oxidant source. The ALD is a unique process that produces dense, highly conformal, nearly pinhole-free thin films that are ideal for gas diffusion barriers. The ALD can also effectively coat the substrate with conformal film, control the film thickness precisely, and tune the film composition flexibly [6].

In this study which follows, six representative Al_2O_3 ALD samples coded as 12k1001, 12k1002, 12k0901, 12k0902, 12K0803 and 12K0804, were assessed. These samples have 80mm diameter, and are coated with 40nm ALD Al_2O_3, where each pair of samples was prepared in a clean room under conditions shown in table 1. Prior to the surface measurements and after the completion of the ALD process, these samples are measured for water vapour transmission rate (WVTR) using Isostatic standard test instrumentation (MOCON®) at specified conditions (38C° and 90% RH respectively) with a stabilisation time of 5 days.

Table 1. Shows the samples pre-coating condition

Sample No	WVTR (g/m²/24 hrs.)	Practice No	Conditions
12k1001	$5x10^{-4}$	Practice 1	Polymer surface unprotected before loading for ALD.
12k1002	$<5x10^{-5}$		
12k0902	$1x10^{-3}$	Practice 2	Polymer surface protected to the last moment before loading into ALD. However, some visible scratches were reported on sample 12k0902.
12k0901	$<5x 10^{-4}$		
12k0803	$6x10^{-4}$	Practice 3	Contact cleaning of the polymer before ALD.
12k0804	$<5x10^{-4}$		

Blunt *et al* (2013) correlated defects size-scale with WVTR values for a representative set of 40nm thick Al_2O_3 ALD samples, where the latest segmentation feature parameter analysis (ISO 25178-pt2., 2012) was used. The authors" outcomes would appear to suggest that small numbers of large defects are the dominant factor in determining WVTR for these barrier layers.

4. CHARACTERISATION TECHNIQUES

With increasing competition in the photovoltaic industry, the development of quality control tools to maximise efficiency and lifespan is a critical issue. Certain Areal surface texture parameters have been shown to be an effective tool to predict the PV module performance and lifespan [9]. In this study, surface metrology techniques along with SEM were employed to acquire both two-dimensional (2D) and three-dimensional (3D) surface topography information over a relatively large field of view in seconds without contacting or otherwise damaging the samples. Using the optical microscopy, direct imaging with no need of sample pre-treatment can be achieved. However, the technique has a low resolution of only sub-micron or a few hundredths of a nanometre, mainly due to the light diffraction limit. The WLSI technique (CCI-6000) has nanometre-level accuracy and repeatability, making the collected data ideal for production monitoring. Using this instrument, a depth resolution of 0.1nm is theoretically plausible, and using a ×20 objective lens a lateral spatial resolution of 0.88μm is achievable while providing a large field of view of $1mm^2$. The high depth resolution means that surface steps, long range surface roughness and discontinuities in a sample can be studied. In addition, the SEM and the AFM are complementary techniques that provide a more complete representation of a surface when used together than if each were the only technique available. According to Stedman amplitude-wavelength plots which was developed in 1987 [10], these two techniques overlap in their capabilities to provide nanometre scale lateral information. However, they deviate in the fact that the AFM can provide measurements in all three dimensions, including height information with a vertical resolution of <0.5Å, whereas the SEM has the ability to image very rough samples due to its large depth of field and large lateral field of view. This paper seeks to give an overview and catalogue most of the Al_2O_3 barrier film surface defects structures, sizes and shape which have a negative effect on the PV module performance, where some "hints" to reduce and avoid these significant defects will be suggested.

5. RESULTS AND DISCUSSION

Counting all the defects (significant and non-significant) based on the visual assessment with no specific criteria does not give a robust method of identifying which defects are responsible for the high WVTR value. Thus, to evaluate functionally significant attributes, such as the defects density, size and distribution of the micro and nano-scale features caused solely by the ALD process, the feature parameters were used to effectively discriminate between the most significant and non-significant defects, which are postulated to be directly responsible for higher WVTR. During the literature of this study, it was found that there is no such a universal definitions of defects structures existed in Al_2O_3 ALD barrier films. Therefore, a defect table was developed to define critical (significant) defects structure based on the "Wolf pruning" Method. The study has classified numerous significant defect types, listed in table [1]. In this case the term „significant" is distinct from „critical" and refers to the threshold for detection. Sq values of approximately 0.8 nm where found for defect free areas of the planarised Polyethylene Naphthalate (PEN) with 40 nm ALD Al_2O_3 used in the study (under x20 magnification).

Table 2. Types of defects and their size scale

Type of defect	Feature Size	
	Height/depth	**Width**
Spikes (non-significant)	≥ 2.4 nm height	1 pixel
Cracks	≥50nm depth	≥300μm length
Scratches	≤2.5μm height	≥300μm
Ghost defects (unmeasurable feature)	N/A	30-50 μm lateral dimension
Pinholes	≥2.5nm depth	≤15 μm lateral dimension
Peaks/particles	≥2.5nm height	≥15 nm width
Holes	≥2.5nm depth	≥50 μm lateral dimension

The results in fig (3) indicated that those samples (12k1001 and 12k1002) which were deliberately left exposed in a clean room environment over night show lower numbers of large defects and low WVTRs indicating that the films have collected particles, but the particles are not a significant impact on the WVTR values. Sample 12k0901 has less large defects recorded than the other samples. This is believed to be attributed to the nature of sample handling conditions. The procedure for sample handling and purging/cleaning of the ALD coating equipment was optimised for this sample. This ensured few or even no particles were present on the surface prior to the ALD process. Therefore, the WVTR value was very low. In contrast to this, sample 12k0902 prepared with the same conditions as sample 12k0901demonstrated a higher large defect count than the other samples and still had the highest WVTR value ≈ 1×10^{-3} $g/m^2/day$. This sample has larger defects than the other investigated samples, which may have had a negative effect on the barrier properties thus giving an increase in the WVTR. Lastly, sample (12k0803 and 12k0804) show evidence of more particles and scratches, the MOCON tests show a low WVTR, indicating that the web-roller used to clean the substrates before the ALD process, may increase the WVTR by causing scratches.

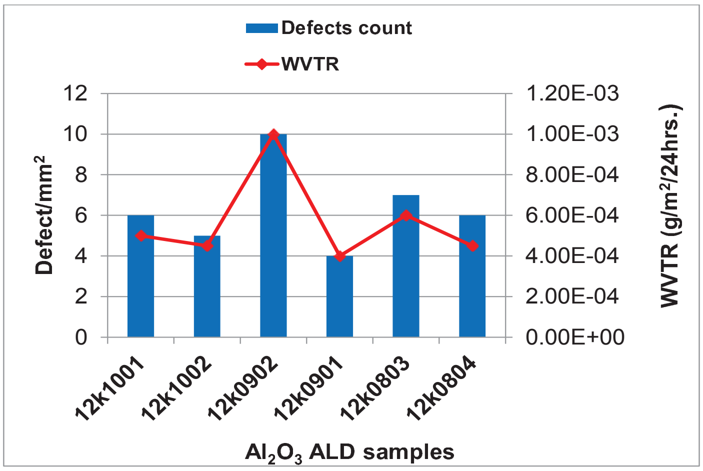

Fig (3) WVTR versus defects size scale.

5.1 CATALOGUE OF AL_2O_3 ALD DEFECTS

A number of different defects were observed in the topography of the samples, these could be classified as shown in the following images. Figure (4) and Figure (5) show two types of defects, debris type defect which was captured by the Scanning Electron Microscopy technique(SEM), and pit type defect which was observed by an optical microscopy (Keyence VHX-600 digital HD CCD).

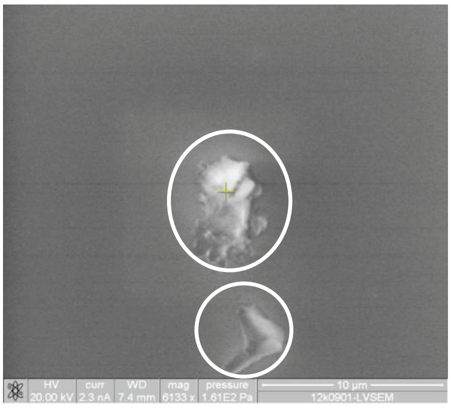

Fig (4) SEM image "debris type defect"

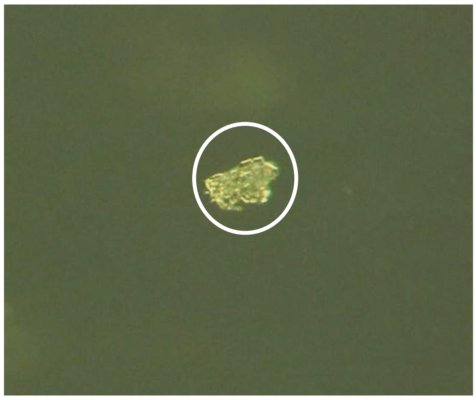

Fig (5) Optical Microscopy image "pit type defect"

The following figure (6) shows very large (hole) type defect. This hole has a size of approximately 75μm width and ≤1μm depth. This hole has a very significant effect on the barrier performance.

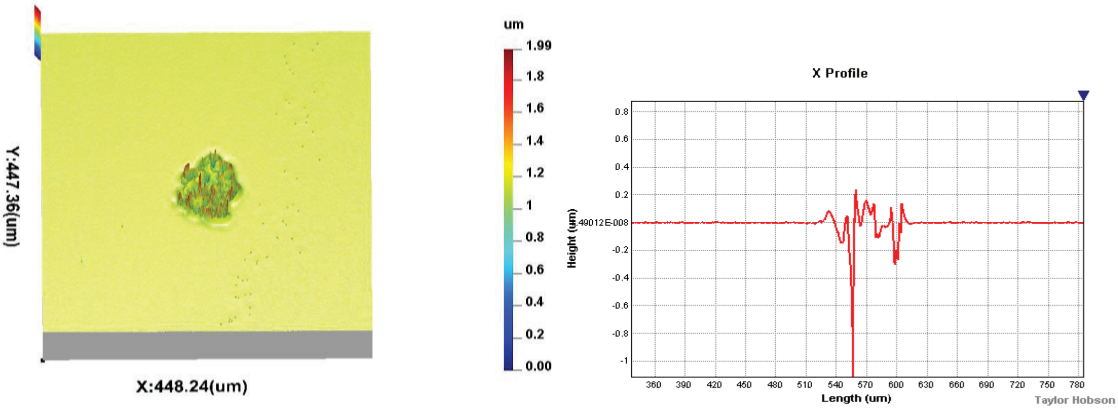

Fig (6) CCI image "Hole type defect"

Atomic Force Microscopy (AFM) was also used in contact mode, the technique has been seen to have the potential to investigate defects within nm size. Figure (7) illustrates spike type defect with 200nm height and ≤2nm width.

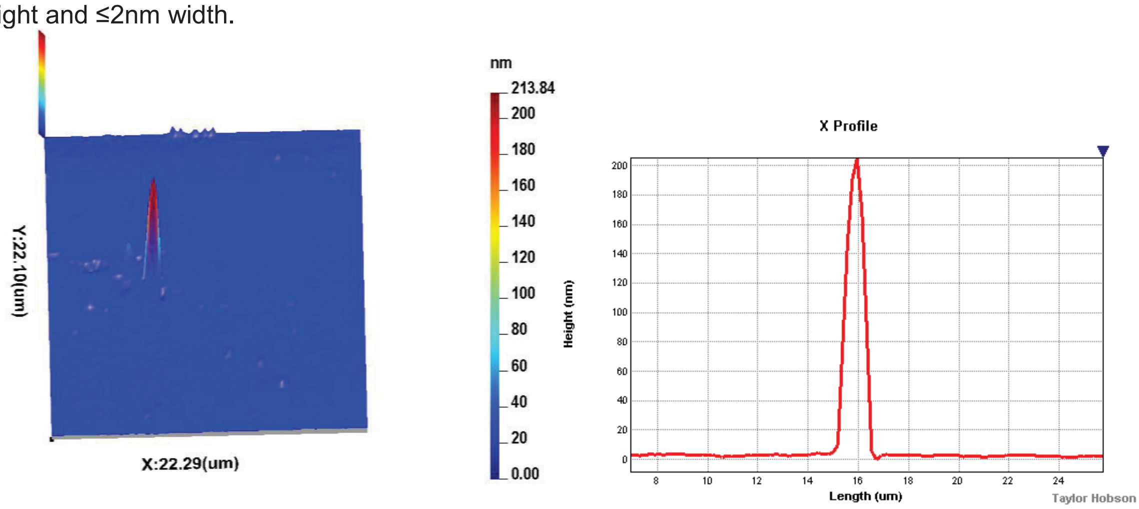

Fig (7) AFM image-3D and 2D profile "spike type defect"

Finally, referring back to table (2), Figure (8) shows an example of peak type defect. This type of defect was investigated by (CCI-6000) instrument.

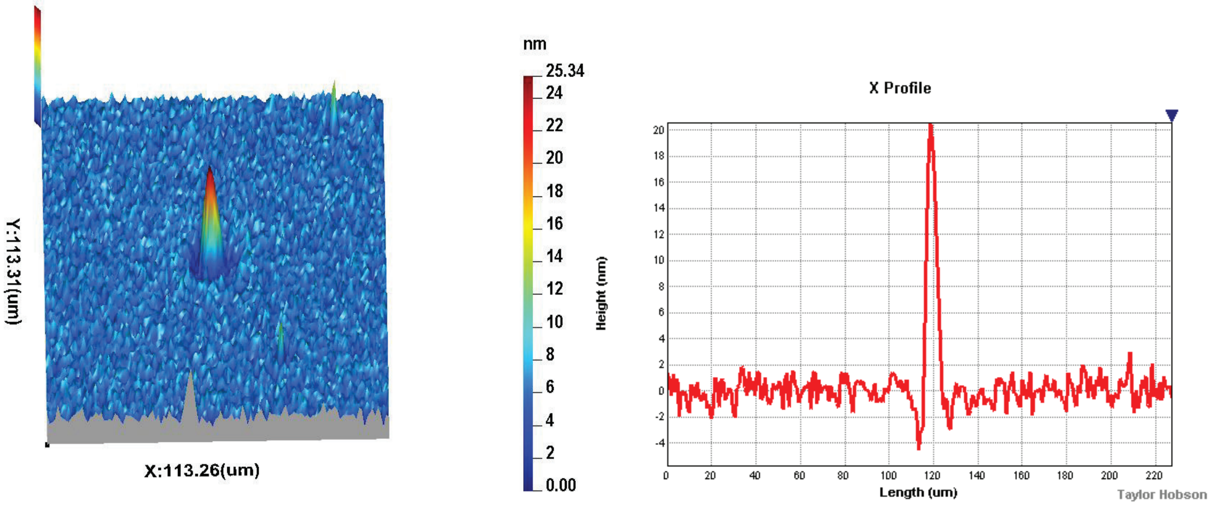

Fig (8) CCI image- 3D and 2D profile “peak type defect”

6. CONCLUSION AND RECOMMENDATIONS

To summarise, this study gave a good insight into the best practice to be used when pre-preparing the samples for Al_2O_3 ALD coating process. The type, size and shape of the defects which have a negative effect on the WVTR value were defined. Hence, to achieve low WVTR value, longer lifespan and best efficiency the following criteria are recommended to be followed when preparing the polymer layer for ALD coating.

- The study emphasises the importance of reducing contamination during ALD process.
- contact roller cleaning has little or even marginally worse effects on the defect count as compared to un-cleaned/exposed substrates
- Limiting atmospheric exposure ensures best WVTR results (Practice 1 recommended).

In conclusion, the findings of this study would appear to suggest that small numbers of large defects are the dominant factor in determining the quantity of water vapour permeation through the PV barrier film.

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