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Bandgap Imaging in Cu(In,Ga)Se₂ Photovoltaic Modules by Electroluminescence

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ABSTRACT

A unique non-destructive characterization method for apparent bandgap imaging in photovoltaic (PV) devices based on acquisition of two electroluminescence (EL) images in different spectral ranges is presented. A method consists of a calibration procedure and a bandgap imaging procedure. Calibration has to be performed once per module type and EL imaging setup, and must provide a relation between the bandgap and the ratio between two spectrally independent EL images. After calibration, bandgap imaging only requires acquisition of two spectrally independent EL images followed by image processing, making the method very fast and suitable for in-line PV module characterization with regard to spatial (in)homogeneity and production process stability. The method is demonstrated on a commercial state-of-the-art Cu(In,Ga)Se₂ PV module where apparent bandgap fluctuations between 1.07 and 1.15 eV are detected.

KEYWORDS

characterization tools, bandgaps, electroluminescence, photovoltaic devices, solar cells

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1 INTRODUCTION

Cu(In,Ga)Se₂ (CIGS) is amongst the most efficient thin-film photovoltaic (PV) technologies [1,2]. Along with other beneficial material properties, the ability to tune the bandgap by controlling the indium to gallium ratio also contributes to the achieved efficiency, both in general bandgap optimization to best utilize the AM1.5 spectrum [3] as well as in bandgap gradients across the absorber for maximizing the photogenerated carrier extraction [4–8]. Because of polycrystallinity the CIGS is inherently inhomogeneous resulting in electrostatic variations [9] and bandgap fluctuations which were detected by various luminescence techniques on micrometer level [10–12]. Their negative impact on conversion efficiency was evaluated to a few percent absolute depending on the magnitude of variations [13–15]. On the module level constant stoichiometry is even more difficult to ensure over a large area, resulting in In:Ga ratio fluctuations and bandgap inhomogeneities [16,17], which were estimated to hamper the performance in the order of few percent absolute [18,19]. To cope with the issue of lateral bandgap fluctuations a fast, reliable and non-destructive characterization method for bandgap mapping is required. In this paper we present such a method based on acquisition of two electroluminescence images in different spectral ranges. A method with similar basis was used by Würfel to calculate diffusion lengths, while different aspects of using spectral information of electroluminescence images were reviewed by Kirchartz [20,21]. The method presented herein focuses on absorber homogeneity studies, in particular characterization of CIGS PV modules.

2 THEORETICAL BACKGROUND

Electroluminescence (EL) is a phenomenon present in semiconductor materials where electrically generated electron – hole pairs recombine radiatively and emit light. The spectrum of emitted light corresponds to the convolution of free charge carrier concentrations p and n (for holes and electrons, respectively). The profiles of p and n versus energy are defined by a product of available density of delocalized states and their occupation functions. The states are available for electrons in the conduction band, and for holes in the valence band. Bandgap is the energy difference between the conduction and the valence band and consequently also influences where the EL spectrum reaches its peak photon energy ($\hat{E}_\gamma$) caused by band-to-band radiative recombination.

In CIGS solar cells the EL emission at ambient temperature is found to dominantly originate from band-to-tail transitions [22,23]. The tail states are available energy states in the bandgap close to either conduction or valence band. In CIGS they are relatively shallow and their exponential decay is faster than the exponential decay of the Fermi-Dirac functions at room temperature, meaning that the normalized shape of the spectrum and $\hat{E}_\gamma$ are injection independent. The temperature dependence of $\hat{E}_\gamma$ is also reported to be minute in a narrow temperature range around 300 K [23]. The well-behaved properties of CIGS make the correlation between $\hat{E}_\gamma$ and apparent bandgap ($\tilde{E}_G$) possible.

We will refer to the term apparent bandgap because of two reasons. First, CIGS is a polycrystalline material with bandgap variations between grains and their boundaries [10–12] that cause EL radiation variations on micrometer level. Such small variations cannot be resolved on module level EL imaging due to resolution limitations. And second, due to bandgap engineering [4] the EL spectrum and intensity change along the depth profile through the absorber, thus the EL radiation emitted from the surface is the integral of EL radiation contributions originating from within the absorber.

During the EL image acquisition each pixel of the camera acquires the signal which is essentially the integral of the incident EL radiation spectrum emitted from the corresponding pixel's area on the sample multiplied by the spectral response of the camera. The detected pixel value is thus EL spectrum and camera spectral response dependent. While the latter is location independent, the EL spectrum depends on the injection level and defines the $\hat{E}_\gamma$, which are both location dependent. The acquired EL image is therefore a result of injection level and $\hat{E}_\gamma$ variations.

If two EL images are acquired under the same injection level, but in different spectral ranges in order to retain the $\hat{E}_\gamma$ dependence, they may be divided and produce an injection level independent ratio image ($\Phi_{\text{EL ratio}}(x,y)$). When the spectral ranges are appropriately different, the ratio image can be correlated with $\hat{E}_\gamma$, from which $\tilde{E}_G$ can be calculated.

3 EXPERIMENT

3.1 Sample

The sample used in the experiment is a commercially available full-size ($126 \times 98 \text{ cm}^2$) CIGS PV module independently purchased on the market. The module is a state-of-the-art product with efficiency under standard test conditions close to 14 %. Prior to testing the module was exposed outdoors to 500 kWh/m^2 of solar irradiation and afterwards stored indoors in the dark for a few months with some occasional performance and EL measurements.

3.2 Setups

The whole procedure requires three measurement setups: external quantum efficiency measurement, EL spectrum measurement, and EL image acquisition. All three setups are required for calibration while only the EL image acquisition setup is required for bandgap determination. The EL image acquisition setup must provide at least two EL images in appropriately different spectral ranges, i.e. the spectral ranges need to (at least partially) cover the spectral range of emitted EL, which may be achieved by using either different cameras or one camera and one or more filters.

3.2.1 External quantum efficiency measurement

External quantum efficiency was measured at TÜV Rheinland with a setup that measures local external quantum efficiency on full-size PV modules. During the measurements the $4.5 \times 4.5 \text{ cm}^2$ area in question is illuminated with bias light and added modulated monochrome probing light. Cells parallel to the area are shaded, while other parts of the module are homogeneously illuminated and a voltage is applied to the module such that the cell voltage across the measuring area is zero. The result of the measurement is the quantum efficiency averaged over the measuring area at position x, y ($Q_e(\lambda, x, y)$).

3.2.2 EL spectrum measurement

EL spectrum was measured with an integrating sphere connected with optical fiber to two spectrometers with cooled imaging detectors, a silicon CCD and an InGaAs array. The opening of the integrating sphere was a circle 1.6 cm in diameter. The result of the measurement is the electroluminescence spectrum averaged over the measuring area at position x, y ($\varphi_{\text{EL}}(\lambda, x, y)$).

3.2.3 EL image acquisition

Spectral imaging conditions are illustrated in Figure 1 together with EL emission spectrum of the module ($\phi_{EL}(\lambda)$ – dash-dot black line). EL images were acquired by two different cameras: an air cooled ($-80\text{ }^{\circ}\text{C}$) 640×512 pixels InGaAs camera with relatively constant quantum efficiency at 85 % between 1000 and 1600 nm (QE_{InGaAs} – solid bright red line), and an air cooled ($0\text{ }^{\circ}\text{C}$) 9 megapixel silicon camera with quantum efficiency up to 1100 nm (QE_{Si} – solid blue line). The InGaAs camera acquires the whole spectrum of CIGS EL radiation (dashed bright red line), while the silicon camera only acquires the high energy part of the spectrum (dashed blue line). The camera quantum efficiencies thus overlap in the wavelength range from 900 to 1100 nm, and the partial spectrum acquisition of silicon camera makes the difference in EL image ratios significant when the EL spectrum of CIGS PV module shifts. The results of the measurements are two EL images: $\Phi_{EL}(\lambda_{\text{InGaAs}}, x, y)$ and $\Phi_{EL}(\lambda_{\text{Si}}, x, y)$ obtained with InGaAs and silicon camera in their respective spectral ranges: λ_{InGaAs} and λ_{Si} .

Image processing of acquired EL images includes background subtraction, removal of bright outliers, and cropping the images to display the whole module area. The image acquired by the InGaAs camera was up scaled and registered to match the size and orientation of the image acquired by the silicon camera. We have used an ImageJ [24] based software package FIJI [25] to prepare the images. The EL images shown in Figure 2 are displayed in false-coloring using Fire LUT and linear scale to clearly display the differences between the images.

3.2.4 Spatial resolution

The spatial resolution of the method is defined by the lower resolution of the two cameras and by the accuracy of the registration (image alignment) procedure. In our case, the camera with the lowest resolution is the InGaAs camera, where the module's active area ($925 \times 1222\text{ mm}^2$) was imaged by 408×539 pixels. This resolution is further reduced as we scaled down the final bandgap image to 226×299 pixels in order to blur the effect of different geometrical distortions and consequent mis-registration. This yields a resolution of $4 \times 4\text{ mm}^2$.

4 RESULTS

4.1 Method prerequisite

The method is applicable and will produce reliable results when the EL spectrum shape of PV modules in question fulfils the following conditions: EL spectrum shape injection independence, EL spectrum shape temperature independence, and in case of meta-stable PV modules, which is the case for CIGS [26], also independence of EL spectrum shape from meta-stable effects. The EL spectrum shape for the selected module satisfies the above conditions which is demonstrated by overlapping normalized EL spectrum curves for: injection independence (Figure S1a), temperature independence in the anticipated range (Figure S1b), and short term meta-stability independence (Figure S1b, EL spectrum measured after 90 min of module inactivity).

Despite the EL spectrum shape independence the EL intensity is injection, temperature and meta-stability dependent. This requires that both EL images are acquired simultaneously or at the same conditions (including injection levels, temperature, and equal inactivity time before the measurement) to cancel out the influence of intensity dependencies.

4.2 Calibration procedure

Before imaging the bandgap a calibration procedure that links the image ratio to the measure of the bandgap must be performed. Herein we present a two-step calibration, consisting of first identifying the $\tilde{E}_G$ to $\hat{E}_\gamma$ correlation, and second identifying the $\hat{E}_\gamma$ to $\Phi_{EL\ ratio}$ correlation. The choice of the calibration procedure depends on the goal of bandgap imaging, e.g. for stoichiometry imaging the $\hat{E}_\gamma$ to $\Phi_{EL\ ratio}$ correlation may be combined with Ga/(In+Ga) ratio to $\hat{E}_\gamma$ correlation [27]. The calibration procedure may as well be reduced to one single step correlating the $\Phi_{EL\ ratio}$ to the desired quantity, either $\hat{E}_\gamma$, $\tilde{E}_G$, or Ga/(In+Ga) ratio. It suffices to perform the calibration at a few points in the module, that we will call calibration locations. The exact number of calibration points depends on the quantum efficiency of the cameras and luminescence spectrum. In our case minimum of four points were required to select the function that best fits selected points and to select the constants in eq. (2); however, additional points would improve the accuracy of the method.

Prior to calibration, calibration locations have to be chosen on the module. The locations are best chosen such that the full range of EL intensity ratios is covered. Simultaneously, the area around the selected locations must be as homogenous as possible such that the spatial average of ratio image on the area of EQE measurement and EL spectrum measurement is the same. Such a selection allows the following triplet to be accurately defined: the apparent bandgap (from EQE), the peak EL position (from EL spectrum) and the average image ratio (from ratio image), and will thus not limit the resolution of the whole method. The two EL images acquired by (a) the InGaAs camera ($\Phi_{\text{EL}}(\lambda_{\text{InGaAs}}, x, y)$), and (b) by the silicon camera ($\Phi_{\text{EL}}(\lambda_{\text{Si}}, x, y)$), together with selected calibration locations are shown in Figure 2. Note the significantly different intensity distributions between the images, and the selection of calibration locations.

4.2.1 Apparent Bandgap to Peak EL Calibration

For $\tilde{E}_{\text{G}}(\hat{E}_{\gamma})$ calibration we chose locations a, d, f , and g , which ensure significant EL intensity ratio variations. Q_{e} has been measured at those locations and the $\tilde{E}_{\text{G}}$ was extracted by linear extrapolation of the low energy edge of the normalized Q_{e} as stated in [28] and shown in Figure 3a. EL spectra were measured at 7 locations on the module. Normalized EL spectra are shown in Figure 3b, where the values of $\hat{E}_{\gamma}$ were extracted from the peaks of EL spectra.

Determined $\tilde{E}_{\text{G}}$ at different locations (a, d, f , and g) are plotted versus $\hat{E}_{\gamma}$ in Figure 4. Linear approximation (black – dashed line) to $\tilde{E}_{\text{G}} - \hat{E}_{\gamma}$ pairs is:

$$\tilde{E}_{\text{G}} = 1.122 \cdot \hat{E}_{\gamma} - 0.123 \text{ eV} \quad (1)$$

In the case, when the density of states for both bands are step functions, one would expect the eq. (1) to behave as an identity $\tilde{E}_{\text{G}} = \hat{E}_{\gamma}$ (grey – dash-dot line in Figure 4), or even $\hat{E}_{\gamma} > \tilde{E}_{\text{G}}$ in a case of an ideal semiconductor [28]. The odd behavior of the eq. (1) may be attributed to the bandgap grading, which may cause reabsorption of the photons produced by EL within the junction, such that only photons with lower energies leave the device and are detected as EL signal. The measured $\hat{E}_{\gamma}$ is thus shifted (grey arrows in Figure 4) towards lower energies, and the fact that the shift is larger at higher energies supports this explanation (the higher the energy, the larger the shift). However, other factors such as the $n \cdot p$ product usually being the

highest at the point of the lowest bandgap, and the influence of stoichiometry changes on the bandgap grading may also influence the observed slope.

4.2.2 Peak EL to EL Ratio Calibration

The $\hat{E}_\gamma(\Phi_{\text{EL ratio}})$ calibration is performed at all 7 calibration locations on the module. The $\hat{E}_\gamma - \Phi_{\text{EL ratio}}$ pairs are shown in Figure 5. The correlation is approximated with a logarithmic dependency (black – dashed line):

$$\hat{E}_\gamma = -0.0333 \text{ eV} \cdot \ln(\Phi_{\text{ELratio}}(x, y)) + 1.29 \text{ eV} \quad (2)$$

where $\Phi_{\text{EL ratio}}(x, y)$ is defined as a per pixel ratio between corrected and registered EL images taken with InGaAs and Si camera:

$$\Phi_{\text{ELratio}}(x, y) = \frac{\Phi_{\text{EL}}(\lambda_{\text{InGaAs}}, x, y)}{\Phi_{\text{EL}}(\lambda_{\text{Si}}, x, y)} \quad (3)$$

The correlation in eq. 2 depends on the shapes of emitted EL spectra and camera quantum efficiencies and is selected empirically with no physical background. For different means of acquisition of spectrally different EL images, i.e. filter use or different cameras, the correlation may appear different.

4.3 Bandgap imaging

The first step in bandgap imaging is acquisition of EL images of the PV module in different spectral ranges. Here we use the same two EL images for the calibration procedure and for the demonstration of the bandgap imaging. The first image was taken with the InGaAs camera and the second image was taken with the silicon camera. Both images were corrected as described in section 3.2.3. The image taken with the InGaAs was up scaled and registered to match the resolution and the orientation of the image taken with the silicon camera. The final images are shown in Figure 2: a) the one taken with the InGaAs camera ($\Phi_{\text{EL}}(\lambda_{\text{InGaAs}}, x, y)$), and b) the one taken with the silicon camera ($\Phi_{\text{EL}}(\lambda_{\text{Si}}, x, y)$).

The prepared images are divided pixel by pixel to create the ratio image ($\Phi_{\text{EL ratio}}(x, y)$) as described by eq. 3. The $\Phi_{\text{EL ratio}}(x, y)$ is then passed pixel by pixel through eq. 2 followed by eq. 1 to calculate the image of apparent bandgap, which is the final result of the method and is shown in Figure 6.

5 DISCUSSION

The EL image acquired by the InGaAs camera (Figure 2a) is much more inhomogeneous than the image acquired by the silicon camera (Figure 2b). The InGaAs camera EL image gives a better representation of the overall EL intensity than the Si camera due to its flat QE in the spectral range of the EL emission [29]. The EL image acquired by the silicon camera may be misleading because the silicon camera acquires only the high energy part of the EL spectrum, therefore the image tends to show higher EL intensity where the bandgap is higher, and not necessarily where the overall EL signal is higher. This fact should be taken into consideration when using silicon cameras for acquisition of EL images of the modules where the EL spectrum varies across the active area and where the camera spectral sensitivity does not cover the whole emitted EL spectrum.

The $\tilde{E}_G$ fluctuations across the module are significant and range from 1.07 to 1.15 eV. The average apparent bandgap across the module is 1.11 eV with standard deviation of 14.6 meV. Although we have not studied the impact of bandgap fluctuations on module's performance in this paper, based on quadratic fit to simulation results in [14], such fluctuations of the bandgap would result in absolute 0.22% decrease in efficiency at Shockley Queisser limit. In real devices the decrease of performance would be even a bit lower than that. Because the module's efficiency decreases gradually at small standard deviations and becomes ever steeper with increasing standard deviations of the bandgap, the resolution (bandgap-wise) of the method is fine enough to detect band gap inhomogeneities with impact on efficiency below 0.1% absolute on module scale.

Comparison of $\tilde{E}_G$ image (Figure 6) to the EL image taken with InGaAs camera (Figure 2a) shows that in general the areas with lower $\tilde{E}_G$ correspond to the areas with higher Φ_{EL} . Such behavior is expected, because: (a) areas with lower $\tilde{E}_G$ require lower voltage for the same current and consequently the same EL emission intensity, and (b) areas with higher $\tilde{E}_G$ require higher voltage for the same current and consequently the same EL emission intensity. Presence of the areas with different $\tilde{E}_G$ and different voltage requirements on the same PV module stimulates lateral currents over the module that flow from areas with higher $\tilde{E}_G$ to areas with lower $\tilde{E}_G$, as illustrated in [18]. Hence the distribution of EL emission from such

module is a result of the interplay between the bandgap and series resistance of the front and the back contacts.

6 CONCLUSION

A new method for non-destructive mapping of apparent absorber bandgap is presented and demonstrated for the CIGS PV module. It is based on the calibration procedure followed by the acquisition of two EL images in different spectral ranges. Once the calibration procedure is done and relationships (eq. 1 and 2) established the bandgap mapping is very fast and can be used as an integrated in-line characterization method in PV module production process for real time quality monitoring. The method requires single calibration to determine the EL images ratio to bandgap correlation for a given PV module type and selected EL measurement setup. Although the procedure herein is presented on the ratio of two EL images, acquiring more images in appropriate spectral ranges or even using hyperspectral EL imaging would further improve the accuracy.

The method revealed significant lateral apparent bandgap fluctuations in the state-of-the art commercial CIGS PV modules. The average bandgap of 1.11 eV is probably the designed bandgap value, which shows that otherwise high-quality production process renders minor inhomogeneities across larger areas. Further improvement of production process would improve the module homogeneity and increase module performance.

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FIGURE CAPTIONS

Figure 1.

Spectral imaging conditions. Quantum efficiencies of the cameras (QE_{Si} , QE_{InGaAs}), typical EL emission spectrum of CIGS (φ_{EL}), and EL signal as detected by different cameras. Note that signal detected with Si camera is multiplied by 100 in order to be visible in the figure.

Figure 2.

EL images of the CIGS PV module acquired by

- a) the InGaAs camera ($\Phi_{EL}(\lambda_{InGaAs}, x, y)$),
- b) the silicon camera ($\Phi_{EL}(\lambda_{Si}, x, y)$).

Figure 3.

- a) Apparent bandgap ($\tilde{E}_G$) determination by linear extrapolation of external quantum efficiency ($Q_e(E)$) to 0 (overlaid bright lines).
- b) Normalized EL spectra ($|\varphi_{EL}(E)|$) at different locations on the module.

Figure 4.

Apparent bandgap ($\tilde{E}_G$) relationship to the peak of EL spectrum ($\hat{E}_\gamma$) with a linear regression line (black – dashed), identity line (grey – dash-dot), and illustrated shifts (grey arrows).

Figure 5.

Peaks of EL spectra ($\hat{E}_\gamma$) vs. ratio of two EL images ($\Phi_{EL\ ratio}(x, y)$) at calibration locations (x, y) with logarithmic approximation of the relationship.

Figure 6.

Apparent bandgap mapping across the whole CIGS PV module based on two EL images and calibrated relationships.

FIGURES

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Figure 2. EL images of the CIGS PV module acquired by

a) the InGaAs camera ($\Phi_{EL}(\lambda_{InGaAs}, x, y)$),

b) the silicon camera ($\Phi_{EL}(\lambda_{Si}, x, y)$).

Figure 3.

a) Apparent bandgap ($\tilde{E}_G$) determination by linear extrapolation of external quantum efficiency ($Q_e(E)$) to 0 (overlaid bright lines).

b) Normalized EL spectra ($|\phi_{EL}(E)|$) at different locations on the module.

Figure 4. Apparent bandgap ($\tilde{E}_G$) relationship to the peak of EL spectrum ($\hat{E}_\gamma$) with a linear regression line (black – dashed), identity line (grey – dash-dot), and illustrated shifts (grey arrows).

Figure 5. Peaks of EL spectra ($\hat{E}_\gamma$) vs. ratio of two EL images ($\Phi_{EL \text{ ratio}}(x,y)$) at calibration locations (x, y) with logarithmic approximation of the relationship.

Figure 6. Apparent bandgap mapping across the whole CIGS PV module based on two EL images and calibrated relationships.