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# Effect of band gap on power conversion efficiency of single-junction semiconductor photovoltaic cells under white light phosphor-based LED illumination

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#### ABSTRACT

On the basis of the detailed balance principle, curves of efficiency limit of single-junction photovoltaic cells at warm and cool white light phosphor-based LED bulbs with luminous efficacy exceeding 100 lm/W have been simulated. The effect of energy band gap and illuminance on the efficiencies at warm and cool light is discussed. The simulations carried out show that maximum power conversion efficiency at 1000 lx reaches 52.0% for cool light and 53.6% for warm one, while the optimal energy band gap is 1.80 eV and 1.88 eV, respectively. The simulated limits are also referenced to experimental data presented in literature to show that there is still a lot of room for improving indoor photovoltaic cells.

## 1. Introduction

The progress in artificial lighting led to declassification of incandescent lamps by white light LED bulbs. The luminous efficacy of tungsten filament lamps is about 15 lm/W, while halogen lamps with IR filter reach no more than 30 lm/W. On the other hand, the luminous efficacy of white light LED bulbs for indoor lighting has already got  $100 \, \text{lm/W}$ . So the LED technology significantly reduces energy consumed for lighting and we can say that LED bulbs are the most environment friendly among all artificial light sources. White light LED lamps have already replaced incandescent lamps and are replacing fluorescent and gas-discharge lamps step-by-step. Main property of LED lighting is no infrared emission, so indoor photovoltaic cells working under such radiation does not harvest any infrared radiation.

Photovoltaic cells for indoor application recently attract a great deal of attention. Such cells can generate electric power using artificial light and the power can be consumed to recharge batteries of mobile devices or to power wireless sensors. There are many works dealing with the conversion of radiation emitted by artificial light sources (incandescent bulbs [1,2], fluorescent or other gas-discharge lamps [1,3–7] and recently also white light LED bulbs [1,5,6,8]) into electric power by photovoltaic systems. Much of the research was focused on silicon cells and simultaneously the results got on non-silicon cells such as GaAs, CIGS, AlGaAs, GaInP, dye-sensitized, organic or perovskite solar cells

were often compared just with silicon cells [1, 4, 6, 8–11]. However, the comparison between the band gap of monocrystalline silicon (1.12 eV) and the spectra of artificial sources of light shows that silicon does not fit the spectra so much. This thesis has been confirmed in several works, theoretically or experimentally. For instance Minnaert and Veelaert estimated theoretically power conversion efficiency of CdTe, CIGS and c-Si cells under various artificial light [1]. Their calculations were based on the quantum efficiencies of the solar cells and spectra of typical artificial light sources. They came to the conclusion that monocrystalline silicon cells could have the best power conversion efficiency only under incandescent source of light but under fluorescent and white light LED lamps the power conversion efficiency estimated for CdTe cells overwhelms the other cells. In a subsequent work, Minnaert and Veelaert, applying the same attitude, noticed also that the cell made of GaAs could be even better then CdTe at white light LED bulb [12]. Overwhelming of CdTe and GaAs cells over Si cells has been also noticed by V. Bahrami-Yekta and T. Tiedje calculating efficiencies at artificial lighting by means of real parameters of cells [10]. What is more there are several experimental works confirming these theoretical estimations, for instance: (i) the work of De Rossi et al. where explicitly higher efficiency of a-Si cell than poly-Si cell under compact fluorescent lamp (CFL) and under light-emitting diode illumination is reported [5], (ii) the work of Foti et al. where a-Si cell achieved 9.1% under fluorescent lamp in spite of illuminance of the cell was lower than 100 lx [3], (iii) the works of

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Mathews et al. [13] and of Freunek et al. [6], where the GaInP cell exhibited higher efficiency than a-Si one at low indoor illuminance as well as (iv) several works reporting much higher efficiencies of dye-sensitized, organic and perovskite solar cells than silicon cells at artificial lighting [4,9,11,14–17].

However, we can still raise a question about an energy band gap of semiconductor optimal for indoor photovoltaics. Obviously there is no explicit answer to this question, because the optimal semiconductor band gap depends on the spectrum of the incident radiation [18–20]. Therefore, due to the fact that the white light LED bulbs are already commonly applied for lighting, we can raise a question about the optimal band gap under such kind of lighting. This work gives the answer to it.

Our analysis is based on the detailed balance principle between thermal radiation of surroundings and an ideal single-junction semiconductor cell. Then it is the same attitude as it was applied by Shockley and Queisser to the photovoltaic cell irradiated by a black body of 6000 K [18] and later advanced to terrestrial solar radiation by Henry [21]. Exactly tabled efficiency values of ideal cell under AM 1.5G defined in the document ASTM G173-03 [22] were presented in 2016 by Rühle [23]. On the other hand, to the best of our knowledge the power conversion efficiency for ideal cell under warm and cool white light LED bulbs of light efficacy reaching 100 lm/W has not been presented yet. So the effect of illuminance and correlated color temperature on curves of power conversion efficiency has not been discussed as well. Exact calculations of efficiency limit for ideal cell enable us to define theoretical maxima of short-circuit current, open-circuit voltage and power generated under specific circumstances of illuminance.

The aim of the work is presentation of thermodynamic limit curves of power conversion efficiency for an ideal semiconductor cell under white light phosphor-based LED bulbs with luminous efficacy exceeding 100 lm/W. We take into consideration real spectra of two types of phosphor-based LED lamps namely of warm light with coordinated color temperature (CCT) equal to 3000 K and luminous efficacy equal to 113 lm/W and of cool light with 6500 K and 103 lm/W, respectively. The effect of illuminance on the curves is also presented. The simulated curves are referred to the exact curve of power conversion efficiency limit for AM 1.5G. We also compare our curves with theoretical efficiency estimated earlier for phosphor-based LED lamps and presented in Refs. [6,24]. Finally, we refer the simulated limits to the experimental data presented in literature to show how much can be achieved by optimization of indoor photovoltaic systems.

### 2. Theoretical model

As we mentioned in the Introduction the efficiency limit estimated by Shockley and Queisser for ideal single-junction photovoltaic cell was based on the detailed balance principle. This principle requires that a cell being in thermal equilibrium with surroundings must absorb and emit the same amount of photons in terms of spectral distribution as well as equality between solid angles of absorption and emission. Moreover Shockley and Queisser took an ideal semiconductor into consideration. It means that absorption of a photon always leads to generation of electron-hole pair and emission of photon is always a result of electron-hole recombination. So non-radiative recombination does not occur in the ideal semiconductor. Furthermore, it is assumed that voltage between the cell electrodes is equal to the splitting Fermi levels of electrons and holes in the bulk of semiconductor. Finally, it is accepted that all photons falling on the cell are absorbed if their energy is greater than or equal to band gap and the cell is transparent for all photons with less energy.

In thermal equilibrium with surroundings of the temperature Tc the spectrum of power density of radiation absorbed and emitted by the cell can be calculated easily. It is the best to considerate the cell enclosure in a cavity with walls maintained at  $T_c$  temperature. Due to the detailed balance principle the rate of radiative recombination assigned to an unit

area of the cell of  $T_c$  temperature takes the form:

$$I_{R0} = \frac{1}{2\pi^2 \, \hbar^3 c^2} \int_0^{+\infty} A * \frac{(\hbar \omega)^2}{exp\left(\frac{\hbar \omega}{kT_C}\right) - 1} d(\hbar \omega) = \frac{1}{2\pi^2 \, \hbar^3 c^2} \int_{E_g}^{+\infty} \frac{(\hbar \omega)^2}{exp\left(\frac{\hbar \omega}{kT_C}\right) - 1} d(\hbar \omega), \tag{1}$$

where  $\hbar$  is reduced constant Planck, c in speed of light in vacuum, k is Boltzmann constant,  $\hbar\omega$  is energy of photon,  $E_g$  is energy band gap of semiconductor, while A is its absorptivity. For ideal semiconductor it is accepted that A equals 1 for photons with  $\hbar\omega \geq E_g$  and 0 in the case of  $\hbar\omega < E_g$ . In Eq. (1) the emission by both flat surfaces of the cell is taken into consideration, while the emission from the sides is negligible [18]. It is worth highlighting here that Eq. (1) accounts only these acts of recombination which result in emission of photons by the cell surfaces. On the other hand it does not include the radiative recombination which leads to emission of photons in the bulk of semiconductor but do not take part in the emission from the surfaces of the cell due to earlier absorption in the semiconductor.

If non-equilibrium radiation falls on the cell with the absorptivity *A* then the resultant rate of electron-hole generation per unit area of the cell equals:

$$I_G = \int_0^{+\infty} A * J_{ph,\hbar\omega} d(\hbar\omega) = \int_{E_g}^{+\infty} J_{ph,\hbar\omega} d(\hbar\omega), \tag{2}$$

where  $J_{ph,\hbar\omega}$  the energy spectrum of photon flux falling on the unit area of the cell

Both in the dark and under illumination the current-voltage characteristics of ideal cell is determined by the rate of generation and recombination of electron-hole pairs. In the case when the cell is kept at the temperature  $T_c$  and simultaneously is illuminated by the radiation with spectrum  $J_{ph,h\omega}$  the current-voltage characteristics of the cell takes the following form:

$$J = e I_{R0} \left[ \exp\left(\frac{eV}{kT_C}\right) - 1 \right] - e I_G, \tag{3}$$

where e is elementary charge and V is a voltage between electrodes of the cell. To be precise we have to add, following Würfel [19], that Eq. (3) is based on the Boltzmann approximation of the Fermi distribution. For this approximation the Fermi energies have to be a few  $kT_c$  away from the band edges. For the operation of solar cell where Fermi levels splitting is several  $kT_c$  smaller than band gap Eq. (3) is a good approximation.

The maximum of power conversion efficiency can be calculated according to the following:

$$\eta = \frac{MAKS(-J \times V)}{P_{in}} \tag{4}$$

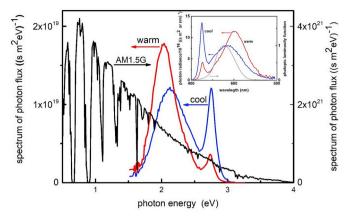
with 
$$P_{in} = \int_0^{+\infty} \hbar \omega^* J_{ph,\hbar\omega} d(\hbar\omega)$$
. (5)

where  $P_{in}$  is surface density of incident radiation power. The function MAKS returns a maximum value of the product of -J and V. The function  $\eta$  depends on temperature, on band gap of cell (i.e.  $T_c$ ,  $E_g$ ) and it is also determined by spectrum of photon flux falling on unit area of cell (i. e.  $J_{ph,h\omega}$ ).

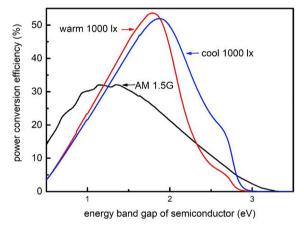
## 2.1. Spectra of photon flux

Artificial light irradiating photovoltaic cells inside buildings is different from solar radiation. The indoor radiation compared to outside solar radiation is (i) of much narrower spectrum of photons, (ii) of much





**Fig. 1.** Spectra of photon flux for AM 1.5G (black line), cool (blue line) and warm (red line) white light phosphor-based LED bulbs with CCT and luminous efficacy equal to 6500 K,  $103\,\mathrm{lm/W}$  and  $3000\,\mathrm{K}$  and  $113\,\mathrm{lm/W}$ , respectively. The inset presents spectra of photon radiance of the lamps originally obtained by Burattini et al. [25] and photopic luminosity function (CIE 1931  $V(\lambda)$  function). The spectra were transformed into photon flux presented in the main body of the figure under the assumption that the illuminance of the surface equals  $1000\,\mathrm{lx}$ .



**Fig. 2.** Maximum of power conversion efficiency against energy band gap for AM 1.5G (black line), at cool (blue line) and warm (red line) white phosphorbased LED bulbs with spectra presented in Fig.1.

lower density of photon flux and (iii) of diffusion characteristics, which means that it falls upon the cell from wide solid angle [8,15].

A high luminous efficacy of white light LED bulbs results mainly from the fact that the emitted radiation is limited to visible range. Fig. 1 presents spectra of photon flux for the solar radiation AM 1.5G defined in the document ASTM G173-03 [22] and of two white light phosphor-based LED bulbs namely emitting warm white light and cool white light. The correlated color temperature of the former was 3000 K and its luminous efficacy was 113 lm/W while for the latter these parameters were 6500 K and 103 lm/W, respectively. The spectral photon radiances of the bulbs ( $I_{ph,\Omega,\lambda}$ ) are presented in inset of Fig. 1. To get

spectral photon flux density falling on a specific surface  $(J_{ph,\lambda})$  we have to estimate the solid angle subtended by the bulb  $(\Omega_b)$  obviously under assumption that we have the Lambertian emitter:

$$J_{ph,\lambda} = I_{ph,\Omega,\lambda} \quad \Omega_b \tag{6}$$

To determine the illuminance of the surface we have to transform quantities of radiometry to quantities of photometry. The latter deals with the measurement of light in terms of its perceived brightness to human eye. To make this transformation we have to use the standardized model of the eyes response to light known as photopic luminosity function (CIE 1931  $V(\lambda)$  function) [26]. Knowing spectral photon flux density falling on the specific surface we can get its illuminance  $(J_{\nu})$  by calculation the following integral:

$$J_{\nu} = 683 \frac{lm}{W} \int_{380 \text{ nm}}^{780 \text{ nm}} J_{ph,\lambda} * \frac{h c}{\lambda} * V(\lambda) d\lambda, \tag{7}$$

where h is Planck constant. Typical indoor illuminance starts from ca. 100 lx (corridors) and ends at ca. 1000 lx (office desk). In the case of surgery lighting the illuminance can reach even 10 000 lx and it is still 10 times less than direct sunlight [26]. The main body of Fig. 1 shows the spectra of photon flux density per energy of photons falling on surface at illuminance equal to 1000 lx transformed from warm and cool white light LED radiations presented in inset of this figure.

#### 2.2. Limit of power conversion efficiency

Fig. 2 shows curves of efficiency limit of ideal single-junction cell versus semiconductor energy band gap under radiation with spectra presented in Fig. 1. The curves were calculated according to Eqs. (1)–(5). The temperature of the cell was taken as equal to 25 °C. As it is seen the limit of power conversion efficiency for phosphor-based LED bulbs overcomes 50% and the best value of band gap is higher than for solar radiation. It is also worth noticing here that the efficiency curves for LED bulbs are not as flattened curves as the one for solar radiation. The range of band gap when efficiency is of more than 90% of its maximum yields 0.36 eV for warm light and 0.44 eV for cool light while for AM 1.5G the same range of efficiency covers 0.67 eV.

As it is known the Shockley-Queisser limit for solar radiation restricts the maximum experimental efficiencies which could be at least hypothetically achieved by single-junction semiconductor cells under unconcentrated light [19,20,27]. It is also worth presenting what hypothetical limits of single-junction semiconductor cell could be achieved by indoor phosphor-based LED bulbs with luminous efficacy exceeding  $100\,\mathrm{lm/W}$ . Table 1 presents maximum values of power conversion efficiency (Eq. (4)), optimal band gap, short-circuit current (Eq. (3) at  $V\!=\!0$ ), open-circuit voltage (Eq. (3) at  $J\!=\!0$ ) and maximum power generated by photovoltaic cells. The results have been calculated for real spectra of photon flux presented in Fig. 1, however at various illuminance of the cell surface.

As it is seen the increase in illuminance from 100 lx to 1000 lx results in the increase of power conversion efficiency from 49.7% to 52.0% for cool light and from 51.1% to 53.6% for warm light. However power generated by cool lamp is a little bit higher than by warm lamp because this bulb emits more blue light than the warm one. Interesting is also the fact that the optimal band gap is explicitly different for warm and cool

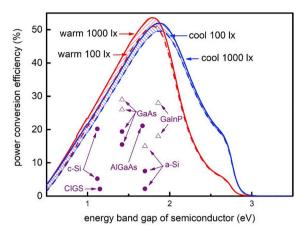
**Table 1**Parameters of two ideal semiconductor cells optimal for warm and cool white light phosphor-based LED lamps.

| $J_{\nu}$ | efficienc | efficiency (%) at optimal $E_g$ |      | optimal $E_g$ (eV) |       | $J_{sc}$ (A/m <sup>2</sup> ) at optimal $E_g$ |      | $V_{oc}$ (V) at optimal $E_g$ |       | maximum generated power ( $W/m^2$ ) at optimal $E_g$ |  |  |
|-----------|-----------|---------------------------------|------|--------------------|-------|---|------|-------------------------------|-------|--|--|--|
|           | warm      | cool                            | warm | cool               | warm  | cool  | warm | cool                          | warm  | cool   |  |  |
| 100 lx    | 51.1      | 49.7                            | 1.80 | 1.88               | 0.134 | 0.136   | 1.30 | 1.38                          | 0.158 | 0.171  |  |  |
| 200 lx    | 51.9      | 50.4                            |      |                    | 0.268 | 0.272   | 1.32 | 1.40                          | 0.321 | 0.346  |  |  |
| 500 lx    | 52.9      | 51.3                            |      |                    | 0.670 | 0.680   | 1.34 | 1.42                          | 0.817 | 0.881  |  |  |
| 1000 lx   | 53.6      | 52.0                            |      |                    | 1.341 | 1.360   | 1.36 | 1.44                          | 1.657 | 1.786  |  |  |



**Table 2**Maxima of power conversion efficiency for ideal semiconductor cells of selected band gap at illuminance equal to 1000 lx.

| $E_g$ (eV) |      | 0.90 | 1.00 | 1.10 | 1.20 | 1.30 | 1.40 | 1.50 | 1.60 | 1.70 | 1.80 | 1.90 | 2.00 | 2.20 | 2.40 |
|------------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| η(%)       | warm | 19.0 | 23.3 | 27.6 | 32.0 | 36.4 | 40.8 | 45.3 | 49.0 | 52.1 | 53.6 | 51.0 | 43.2 | 22.5 | 11.5 |
|            | cool | 18.0 | 21.5 | 25.4 | 29.5 | 33.5 | 37.6 | 41.8 | 45.6 | 45.6 | 51.3 | 51.9 | 49.6 | 38.8 | 27.3 |



**Fig. 3.** Power conversion efficiency limit under illumination by warm and cool white light phosphor-based LED bulb at 100 lx and 1000 lx. Points present experimental results taken from Refs. [5,11,13,17,24] and the energy band gaps for semiconductors from Refs. [6,24,28,29].

LED bulbs. They are equal to 1.80 eV for warm light bulb and 1.88 eV for cool light bulb and they are higher than the optimal band gap for AM1.5G by 0.46 eV and 0.54 eV, respectively.

We must also mention about earlier considerations on efficiency limit of photovoltaic cells under white light phosphor-based LED bulbs. This issue has been discussed by Freunek et al. in Ref. [6] and by Teran et al. in Ref. [24]. Both groups labelled higher optimal band gap, namely 1.90-2.00 eV. Simultaneously the maximum of power conversion efficiency was estimated at 47.70% by the first group and at 60% by the second group. These results are only roughly in agreement with our calculations. However, in the case of the work [6] we find neither parameters nor spectrum of the white light phosphor-based LED bulb used in the performed calculations. So we cannot refer in details to the results presented in Ref. [6] but we can say in general that the spectral exitance of the LED bulb used in Ref. [6] had to be different from the spectral exitance of the warm or cool white light LED bulbs presented in Fig. 1. In the second case the spectral characteristics of the white phosphor LED bulb used for calculation is presented in Ref. [24] along with the luminosity function. We can compare this characteristics with our spectra also presented along with the luminosity function in inset of Fig. 1. The band of phosphor emission is much narrower in Ref. [24] than in our cases. In general in the case of narrow irradiation spectrum we expect higher power conversion efficiency and simultaneously if relatively more radiation is emitted in blue range higher value for optimal band gap is predicted. Nevertheless, it is worth highlighting here that human eyes work more comfortably when source of lighting emits radiation that covers the whole visible range. So the white phosphor LED used in Ref. [24] is not as suitable for human eyes as the warm and cool LED bulbs presented here.

## 3. Discussion

It is obvious that power conversion efficiency of real cells is restricted by the value of limit function at the same energy band gap. To get closer to this limit various projects used to be taken. For instance a lot of work is undertaken (i) to form an appropriate junction (by adequate semiconductor doping), (ii) to reduce the reflection of light from the cell (e. g., by texture) and (iii) to get complete absorption of incident light in the layer of thickness that does not exceed the diffusion length of minority charge carriers (by optimization of cells). However, due to the fact that peak of power conversion efficiency in the case of white light phosphorbased LED is narrower than for AM 1.5G, the hypothetical power conversion efficiency at such lighting is strongly determined by energy band gap of semiconductor. Table 2 presents maxima of power conversion efficiency for ideal semiconductor cells of selected band gap at illuminance equal to 1000 lx. It is seen that the limit of power conversion efficiency at the band gap of 1.8 eV is two times higher than at 1.1 eV both for warm and cool light. So it makes clear why crystalline Si with its band gap equal 1.12 eV is not an adequate material for indoor photovoltaics.

Fig. 3 presents efficiencies at 100 lx and 1000 lx for warm and cool light. The shadowed areas correspond to hypothetical limit of power conversion efficiency at illuminance higher than 100 lx and lower than 1000 lx. Points in Fig. 3 present experimental results obtained under white LED bulbs taken from literature. Presented points, starting from the lowest band gap semiconductor and in the case of selected material from the highest efficiency, deal with: (i) crystalline Si with 20.19% at incident power equal to  $20.5 \text{ mW/cm}^2$  [11] and 5.2% at 580 lx [24], (ii) CIGS with 2.01% at incident power equal to 20.5 mW/cm<sup>2</sup> [11], (iii) GaAs with 29% at 1000 lx [13], with 26% at 200 lx [11], with 19.4% at 580 lx for thin GaAs and 15.5% at 580 lx for thick GaAs [24], (iv) Al<sub>0.2</sub>Ga<sub>0.8</sub>As with 21.1% at 580 lx [24], (v) amorphous Si with 15% at 200 lx and the same value at 1000 lx [13], with 7.5% at 200 lx [5], with 2.18% at incident power equal to 20.5 mW/cm<sup>2</sup> [11], and (vi) GaInP with 28% at 200 lx and 18% at 1000 lx [13]. Solid circles in Fig. 3 present exact values of  $\eta$  reported in the references while the open triangles were estimated on the basis of power generated by the cells and illuminance given in Ref. [13]. To get the values of power conversion efficiency we assumed that incident radiation power was like for our warm white light LED bulb at the same illuminance. As it is seen in Fig. 3 there is still a lot of room between the limit curves and experimental results. The gap between points and curves shows how much could be achieved by optimization. The only exception is crystalline Si, which has already achieved power conversion efficiency equal to 20.19% [11].

As we mention above the optimal energy band gap for single-junction cells is 1.8 eV at warm LED light and 1.88 eV at cool LED light. So from the cells of c-Si, CIGS, GaAs, AlGaAs, a-Si and GaInP the best adjusted gap for LED lighting is met in the case of AlGaAs, a-Si and GaInP because for these materials the limit of efficiency overcome 45%. Another material that can be consider for indoor applications is selenium because it is a high band gap material (Eg = 1.83–2 eV, depending on fabrication conditions). Moreover, Se devices are air-stable nontoxic, and extremely simple to fabricate [30]. For GaAs which on other hand gives the best cell for AM 1.5G [31] the limit of efficiency equals 37.6% at cool light and 40.8% at warm light. It is worth noticing that experimental efficiency reported for this material has already yielded 29% so according to the efficiency limit it can be improved by no more than 30%.

Obviously, in addition to semiconductor cells, we can also consider organic, dye-sensitized or hybrid perovskite cells for indoor photovoltaics. Such cells are based on a junction of different materials and therefore their PCE limit cannot simply be described by S-Q model [27]. However, due to their specific properties they can also be attractive for indoor applications. At present, the PCE under LED lighting has already reached 19.5% for the best dye-sensitized solar cells at 350 lx [32], 16.8% for organic ternary bulk-heterojunctions at 300 lx [33], and in the



case of perovskite photovoltaic cells the highest maximum power density is  $19.9 \,\mu\text{W/cm}^2$  at  $200 \,\text{lx}$  and  $115.6 \,\mu\text{W/cm}^2$  at  $1000 \,\text{lx}$  [34].

#### 4. Conclusions

On the basis of the detailed balance principle, curves of efficiency limit of single-junction photovoltaic cells at warm and cool white light phosphor-based LED bulbs with luminous efficacy exceeding  $100\,\mathrm{lm/W}$  have been simulated. The calculations show that maximum power conversion efficiency at  $100\,\mathrm{lx-}1000\,\mathrm{lx}$  reaches 52.1%-53.6% for warm light and 49.7%-52.0% for cool one, while the optimal energy band gap is  $1.80\,\mathrm{eV}$  and  $1.88\,\mathrm{eV}$ , respectively.

The simulated limits are also referenced to experimental data presented in literature. From the cells of c-Si, CIGS, GaAs, AlGaAs, a-Si and GaInP the best adjusted gap for LED lighting is met in the case of AlGaAs, a-Si and GaInP because for these materials the limit of power conversion efficiency overcomes 45%. According to literature, these three kinds of cells are currently achieving 21.1% [24], 15% [13] and 28% [13], respectively. So we can say there is still a lot of room for improving indoor photovoltaic cells.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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