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# Spray Coating Luminescence Layers on Glass for Si Solar Cells Efficiency Enhancement

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**Abstract:** The article presents experimental research focused on the improvement of solar cells efficiency using the photoluminescence down-shifting effect. In the paper, the authors present the results of solar cells performance enhanced with the proposed solution. As light energy converted active materials, rare earth elements of europium and dysprosium were implemented. In the experiments, luminescent layers were deposited on top of photovoltaic protective glass in order to absorb the highest possible amount of ultraviolet light from the incident solar spectrum. Spray coating deposition technology with various types of ink compositions was used for process optimization. It was observed that there are optimal concentrations and solvent types for the best conversion effect and consequently, the improvement of solar cell external quantum efficiency (EQE) together with the PV cell absolute efficiency enhancement.

**Keywords:** down-shifting; photoluminescence; external quantum efficiency; rare earth elements; spray-coating methods

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## 1. Introduction

The maximum efficiency of photoconversion in solar cells is limited by fundamental physical laws, as well as technical problems. The main aspects of solar cell's efficiency limits are non-absorption of low-energetic photons (about 15% loss), thermalization losses as a result of absorption of the high-energetic photons (about 33% loss), and recombination losses (about 15% loss) [1,2]. In 1961 Shockey and Queisser [3] calculated the maximum generated power as a function of the energy bandgap of solar cell material. According to that, the maximum efficiency of silicon solar cells with an energy bandgap 1.1 eV is 31% [3,4]. Within decades, a lot of concepts appeared to overcome the efficiency limit of solar cells, such as multijunction structure [5], intermediate band [6], split spectrum system [7], hot carrier acquisition system [8], carrier multiplication [9], and spectrum modification [1,4,10,11].

The spectral modification method is divided into three types of processes: up-conversion (UC), where two low-energy photons are absorbed and then re-emitted as one higher-energy photon; down-conversion (DC), where one high-energy photon is absorbed and two lower-energy photons are re-emitted; and down-shifting (DS), which is similar to DC, but there is only one lower-energy re-emitted photon [1,4,10–12].

The down-shifting process was proposed for photovoltaic applications at first in 1979 by Hovel et al. [13]. However, the concept of DS (the enhancement of photovoltaic device based on using high-energy photons from UV range) appeared a little earlier in so-called luminescent solar concentrators [14–16]. During the decades, researchers estimated that DS could enhance solar cell efficiency at the level of about 10% [17–19]. The idea of the process is presented in Figure 1.

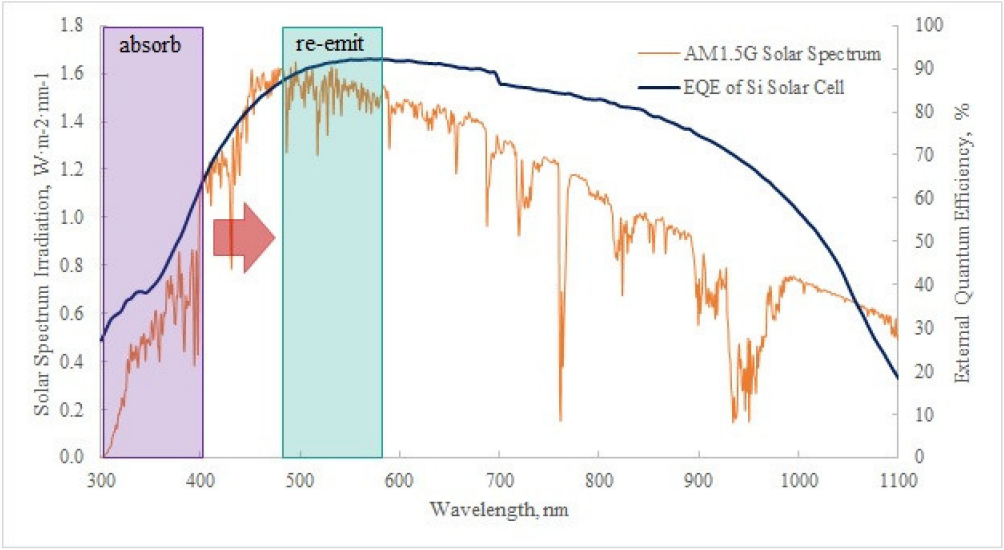



**Figure 1.** The idea of DS process in solar cell. The UV part of solar spectrum radiation is not used by silicon solar cell efficiently; hence, the DS layer is shifting radiation to the range effectively matched to solar cell response.

Although the simulations look very promising, the practical realization of the DS layers remains a big challenge, and there is still a lot to be developed in this field. An efficiency increase of few percentage points is a big achievement [20]. Recent advances comprise, e.g., a 0.51% efficiency increase in a large area crystalline silicon solar cell when coated with EVA copolymer film containing $Eu^{3+}$ [21]. Another recent successful implementation of the rare-earth ions has been reported in [22], where the incorporation of the europium-activated nanophosphors resulted in over 3 percentage points increase in the perovskite solar cell efficiency. Rare-earth elements have been extensively tested for this application, but there is also a wide variety of other luminescent materials that are being considered for this role: chalcogenides, metal oxides and nanoparticles and quantum dots of various kinds [23], e.g., in [24], the authors report a 0.5% efficiency increase in a perovskite solar cell due to the graphene quantum dots.

Nowadays, most popular solutions focused on the light spectrum modification for higher efficiency of solar cells and modules photoconversion are based on LSC (luminescent solar concentrator), sometimes called edge-concentrator. It was initially proposed by Weber [14] and later by Goetzberger [15]. Presently, there are many variants of this construction, but the general idea of the light-trapping flat area with the thin stripes of the solar cells fixed on the concentrator edge is preserved. Even though in this construction light absorbing plate is relatively large and the total cell costs are limited, there are several drawbacks of this solution. The most important are very limited photocurrent due to tiny actual cell area, significant light scattering within the light trapping layer, and untypical construction of the concentrated module.

Alternatively, converting layers for concentrator modules may be placed around the cells arranged within the module with some limited spacings between each other [25,26]. The main advantage of this structure is the utilization of the full-scale standard solar cells and typical module construction but still some spacings between the cells limit active module area, which causes lower values of the photocurrent. Taking this into consideration, the authors proposed an alternative solution aiming at the highest module performance.

The glass, which protects solar cells in photovoltaic modules, unfortunately, absorbs sun radiation, especially in the UV range. Herein, authors propose to cover it by down-shifting luminescent layers in order to obtain the effect, which is schematically presented in Figure 1. In the experiments, the glass was covered by a down-shifting layer; hence, the UV radiation was shifted to visible range and then transferred to solar cell surface. The optical transmittance of glass in the visible range was much higher than for UV, which gave the

efficiency enhancement and what is more, that range was more efficiently converted by solar cells.

Herein, the authors focused on the enhancement process of solar cell response in UV range using DS layers based on compounds with rare earth elements. The paper defines layer fabrication and deposition method. Additionally, optical transmittance and morphology of obtained layers are presented and discussed. Finally, the I-V characteristics and the external quantum efficiency (EQE) of polycrystalline silicon solar cell without and with DS layers is presented and compared.

## 2. Materials and Methods

The luminescent materials chosen for the experiments were two pigments from Nemoto Lumi-materials CO., LTD: $Sr_4Al1_4O_{25}$: Eu, Dy (BGL) and $SrAl_2O_4$: Eu, Dy (G). Firstly, the photoluminescent properties of raw materials were measured. Then, the actual layers were prepared according to three technology variants:

- The pigments were sited in polymer matrix of poly (methyl methacrylate) (PMMA) compound with an average molecular weight of 350,000 by GPC, purchased from Sigma Aldrich Company (label only-PMMA variant);
- The pigments were sited in ethanol and after deposition on glass covered with polymer matrix of PMMA (label ethanol variant);
- The pigments were sited in isopropanol and after deposition on glass covered with polymer matrix of PMMA (label isopropanol variant);

In two last variants, the layers were deposited by spray-coating method using hand airbrush. In all options, the PMMA concentration in the base mixture was 2%. In the first variant, the luminescent material, PMMA and solvent (chlorobenzene) were weighted. The layers were prepared in seven various concentrations of pigments: 1%, 2% 5%, 10%, 15%, 20%, and 50%. Subsequently, the mixtures were stirred with a magnetic stirrer for 24 h under hermetic cover. In two other options, ethanol and isopropanol variants, only the PMMA (base) was prepared in the same way. Pigments were added in four quantity variants: 0.1 g, 0.2 g, 0.5 g, and 1 g and then flooded with 30 mL of ethanol or isopropanol. After that, alcohol (ethanol or isopropanol) with pigments was put in an ultrasonic cleaner for 30 min in order to disperse the pigment evenly in the alcohol volume. Subsequently, the mixtures were deposited by spray-coating method on glass substrate (for optical transmittance measurements and external quantum efficiency (EQE) measurements) as well as silicon substrate (for scanning electron microscopy (SEM) and optical microscopy measurements). In the first option, where pigments were incorporated in PMMA matrix, the layers were deposited and dried at room conditions for 24 h. In the second and third variant, the layers were deposited and left for an hour to let the alcohol evaporate (ethanol or isopropanol, respectively). Afterwards, the pigments were covered by PMMA matrixes as protection layers. Thereafter, the layers were drying in room conditions for 24 h. Finally, the layers and silicon solar cells without and with layers were characterized. The schematic of the processes is presented in Figure 2.

The transmittances of the layers deposited on the glass substrate have been evaluated using Filmetrics aRTie-UV thin-film analyzer with Deuterium-Tungsten source (model LSDT2). The external quantum efficiencies of the polycrystalline silicon solar cell with and without the luminescent layers have been acquired with PVE300 Bentham EQE set-up. The I-V curves have been measured with A-class AM 1.5 Solar Simulation System by Quantum Design GmbH, meeting IEC 60904-9 standard. For SEM analysis, Carl Zeiss EVO MA10 SEM scanning electron microscope was used.

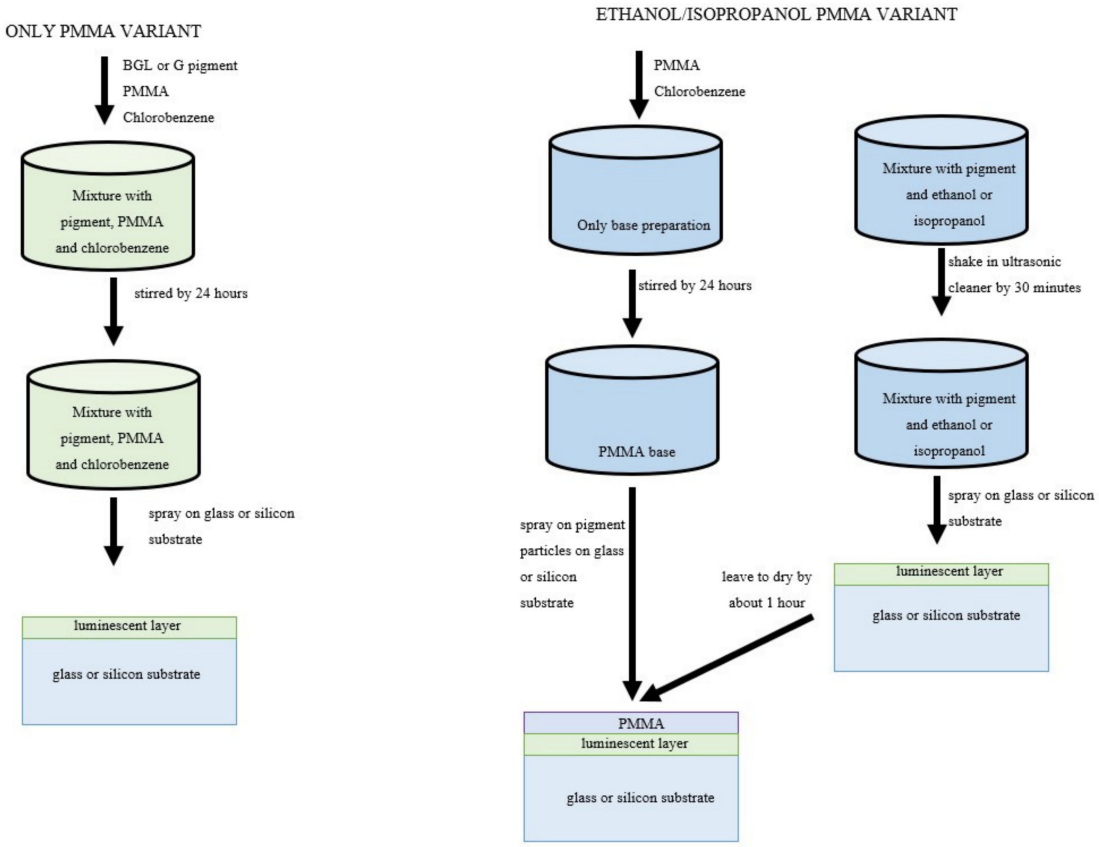



**Figure 2.** Schematic of the experimental process.

## 3. Results and Discussion

The optical transmittance of deposited layers is presented in Figure 3 for pigment BGL and in Figure 4 for pigment G. In both cases, a slight decrease in transmittance value was observed near 375 nm. Combining these results with the photoluminescence analysis of BGL and G compounds, it can be concluded that this value is close to the excitation wavelength where the maximum emission can be achieved (example in Figure 5).

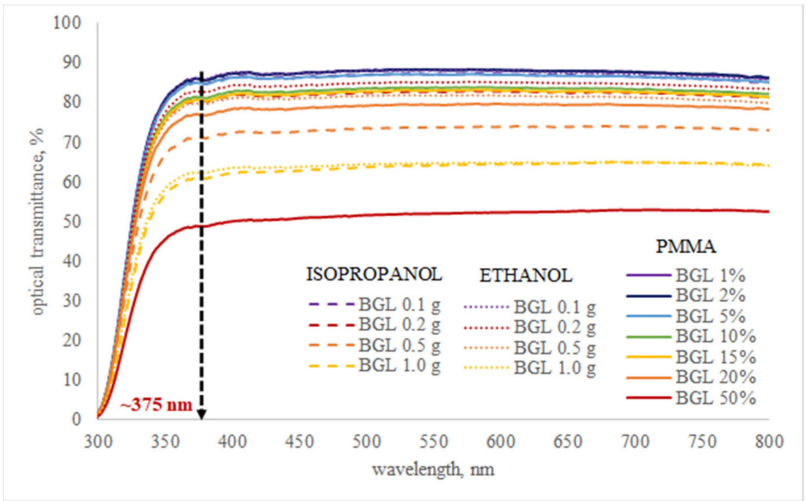



**Figure 3.** The optical transmittance characteristic of layers prepared with spray-coating method based on BGL pigment in all three variants.

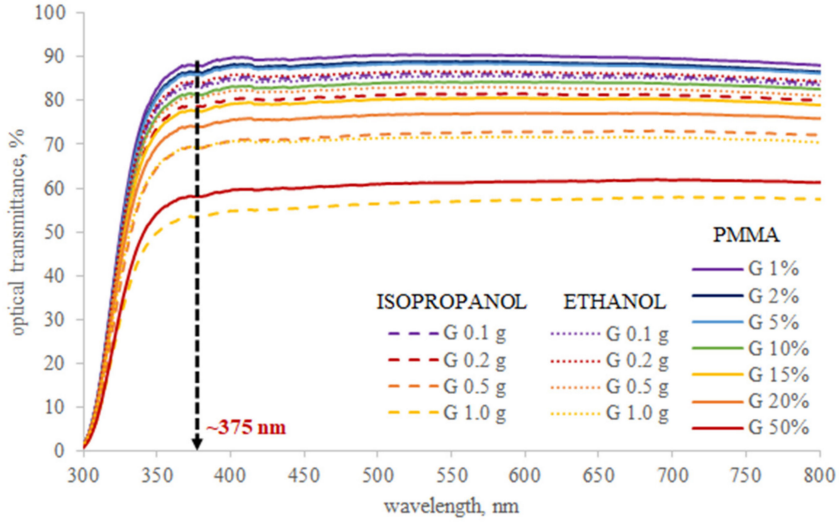



**Figure 4.** The optical transmittance characteristic of layers prepared with spray-coating method based on G pigment in all three variants.

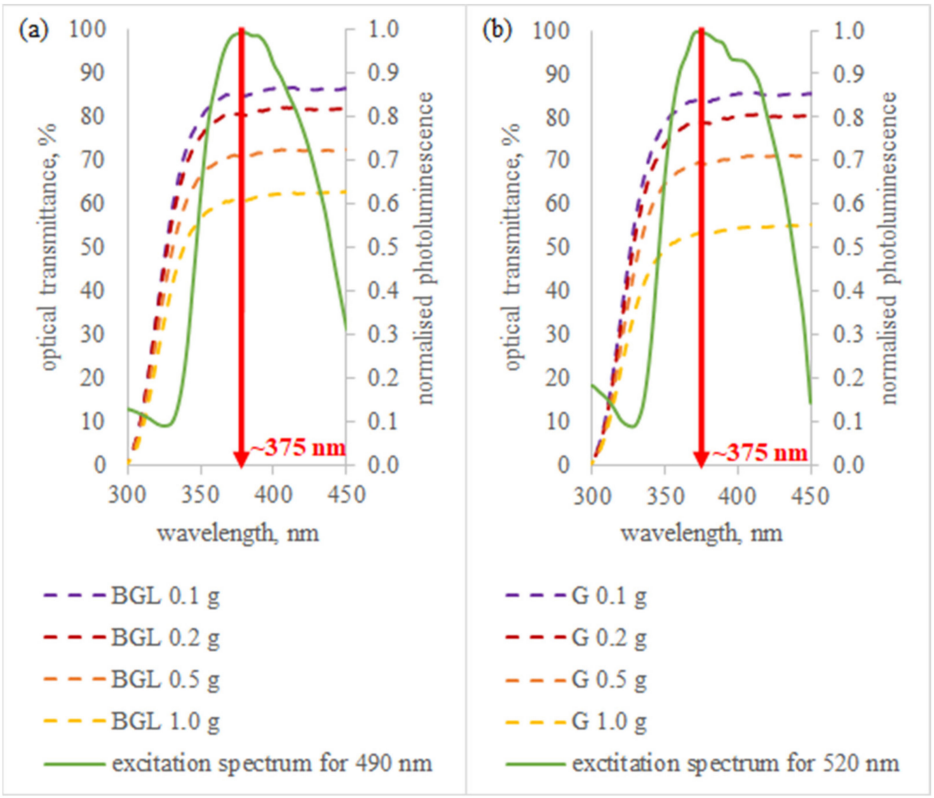



**Figure 5.** The optical transmittance characteristic of layers prepared with spray-coating method based on BGL (**a**) and G (**b**) pigments in isopropanol variant and the excitation spectrum of BGL (**a**) and G (**b**).

For the further evaluation of manufactured layers, the emission parameters should be investigated. The proposed solution is focused on two main elements:

- To improve the spectral response in the UV radiation range;
- To improve the efficiency of photoconversion.

Thus, to confirm these assumptions, at first, the external quantum efficiency (EQE) of polycrystalline silicon solar cells without and with DS layers were characterized. The

results for BGL and G pigments are gathered in Tables 1 and 2, respectively, where for a given wavelength, a difference of EQE for a polycrystalline silicon solar cell with and without the DS layer is shown. The results show that the layers in the ethanol variant have not increased the EQE characteristic for UV range; however, the layers deposited in isopropanol or PMMA variants are promising in some variants. The value of EQE increased relatively by almost 3.2% for the 0.2 g isopropanol variant and more than 4.5% for only the PMMA variant of BGL pigment. The G pigments fared even better in this comparison. The value of EQE was increased relatively more than 8.7 % for the 0.5 g isopropanol variant and more than 16.7% for only the PMMA variant. In Figures 6 and 7, the best improvements of EQE for BGL and G pigment, respectively, in the UV range are highlighted.

**Table 1.** The most important points for EQE characteristic for two variants of deposition for pigments BGL.

| Method | | Absolute Difference in EQE | | Relative Difference in EQE | |
|---|---|---|---|---|---|
| | | λ, nm | Percentage Points, % | λ, nm | Relative Difference, % |
| Isopropanol variant | 0.1 g | - | - | - | - |
| | 0.2 g | 445 | 1.72 | 355 | 3.19 |
| | 0.5 g | 350 | 0.66 | 350 | 1.68 |
| | 1.0 g | - | - | - | - |
| Only-PMMA variant | 1% | 430 | 2.01 | 350 | 4.06 |
| | 2% | 450 | 2.43 | 355 | 4.56 |
| | 5% | - | - | - | - |
| | 10% | 365 | 0.08 | 365 | 0.17 |
| | 15% | 365 | 0.26 | 365 | 0.57 |
| | 20% | 365 | 0.64 | 365 | 1.43 |
| | 50% | - | - | - | - |

**Table 2.** The most important points for EQE characteristic for two variants of deposition for pigments G.

| Method | | Absolute Difference in EQE | | Relative Difference in EQE | |
|---|---|---|---|---|---|
| | | λ, nm | Percentage Points, % | λ, nm | Relative Difference, % |
| Isopropanol variant | 0.1 g | 445 | 3.13 | 360 | 5.83 |
| | 0.2 g | 445 | 5.32 | 350 | 5.91 |
| | 0.5 g | 445 | 5.33 | 360 | 8.73 |
| | 1.0 g | 445 | 3.85 | 360 | 6.92 |
| Only-PMMA variant | 1% | 445 | 7.84 | 355 | 13.13 |
| | 2% | 445 | 10.43 | 355 | 16.74 |
| | 5% | 445 | 6.84 | 355 | 11.54 |
| | 10% | 350 | 0.87 | 350 | 2.20 |
| | 15% | 410 | 1.34 | 375 | 2.42 |
| | 20% | 430 | 3.12 | 350 | 5.29 |
| | 50% | 425 | 3.21 | 355 | 5.44 |

The next step of analysis is research on possible improvement of solar cell efficiency. Results of I-V characteristics measurements in standard test conditions (STC) show that there is a small growth of efficiency in some cases (marked in red in Tables 3 and 4). The main parameters of these measurements for pigment BGL and G are presented in Tables 3 and 4, respectively. Evidently, for BGL pigment, there is a slight improvement of efficiency, which increased from 10.98% to 11.00% for layers containing 0.5 g of powder in the isopropanol variant, as well as in the PMMA variant for 2% concentration from 10.98% to 10.99%. Pigment G benefits in this comparison, where for the isopropanol variant, an efficiency of 11.01% for 0.2 g and 0.5 g is achieved. The best results were obtained for layers based on G pigment in only the PMMA solution method, where the concentration of pigment was 2%. The efficiency growth up to 11.05% was observed in this case. In

Figures 8 and 9, the comparison of efficiencies for BGL and G pigments, respectively, are summarised.

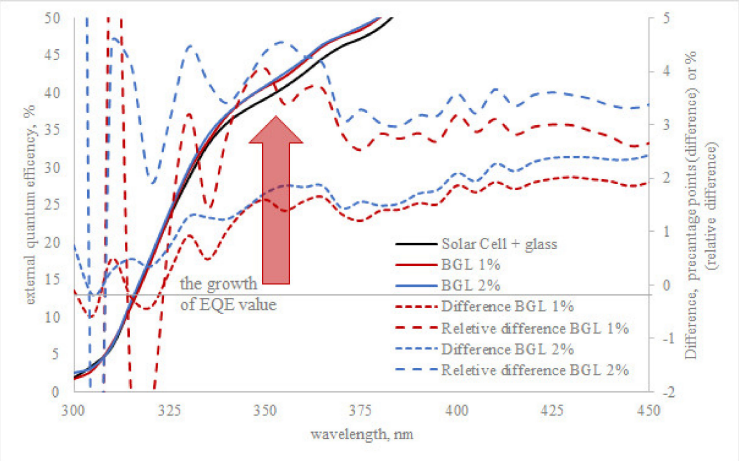



**Figure 6.** The best improvement of EQE for BGL pigment in only-PMMA variant.

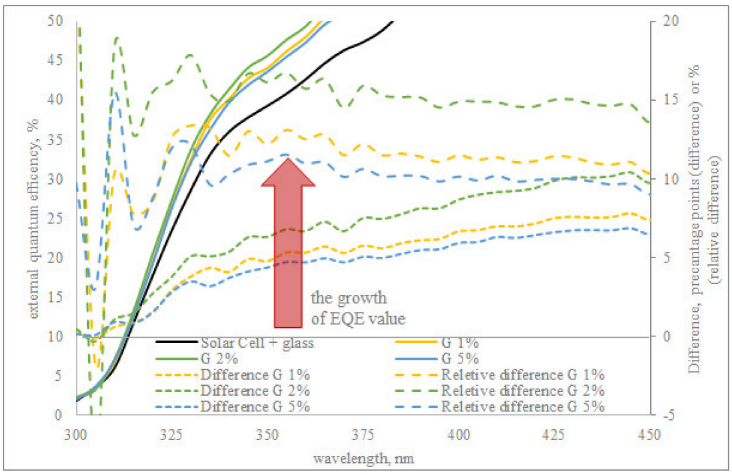



**Figure 7.** The best improvement of EQE for G pigment in only-PMMA variant.

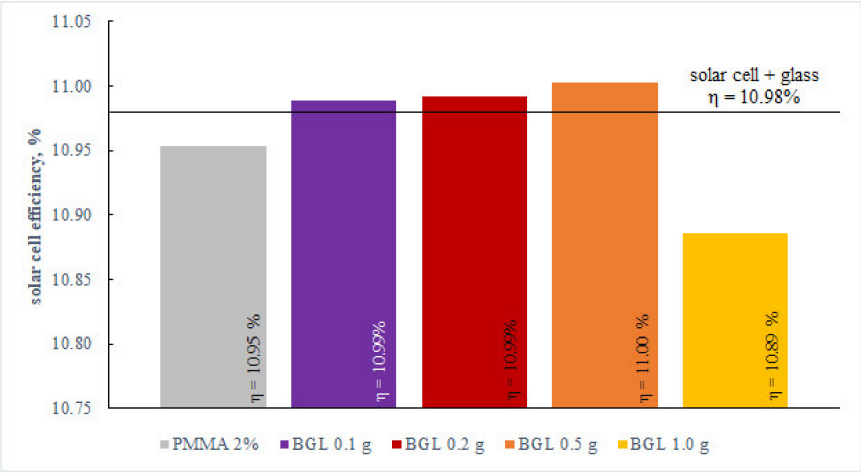



**Figure 8.** The efficiency comparison of BGL pigment in isopropanol variant.

**Table 3.** The characteristic value of the model of photovoltaic panel without and with layers based on BGL by spray-coating deposition method in two variants.

| Substrate/ BGL Pigment Variants | | $J_{SC}$, mA·cm$^{-2}$ | $V_{OC}$, mV | $P_{MMP}$, mW·cm$^{-2}$ | FF, % | η, % |
|---|---|---|---|---|---|---|
| **Pure glass** | | 32.9 | 481 | 10.98 | 69.4 | 10.98 |
| **Base of PMMA 2% on glass** | | 32.9 | 481 | 10.95 | 69.3 | 10.95 |
| **Isopropanol variant on glass** | 0.1 g | 33.29 | 479.1 | 10.99 | 68.9 | 10.99 |
| | 0.2 g | 33.24 | 479.1 | 10.99 | 69.0 | 10.99 |
| | 0.5 g | 33.27 | 479.1 | 11.00 | 69.0 | 11.00 |
| | 1.0 g | 33.01 | 478.6 | 10.89 | 68.9 | 10.89 |
| **Only-PMMA variant on glass** | 1% | 32.99 | 479.1 | 10.97 | 69.3 | 10.97 |
| | 2% | 33.11 | 479.1 | 10.99 | 69.3 | 10.99 |
| | 5% | 33.10 | 479.1 | 10.97 | 69.3 | 10.97 |
| | 10% | 32.79 | 479.0 | 10.89 | 69.3 | 10.89 |
| | 15% | 32.76 | 479.0 | 10.87 | 69.3 | 10.87 |
| | 20% | 32.50 | 478.7 | 10.79 | 69.3 | 10.79 |
| | 50% | 32.01 | 478.1 | 10.59 | 69.2 | 10.59 |

**Table 4.** The characteristic value of the model of photovoltaic panel without and with layers based on G by spray-coating deposition method in two variants.

| Substrate/ G Pigment Variants | | $J_{SC}$, mA·cm$^{-2}$ | $V_{OC}$, mV | $P_{MMP}$, mW·cm$^{-2}$ | FF, % | η, % |
|---|---|---|---|---|---|---|
| **Pure glass** | | 32.9 | 481 | 10.98 | 69.4 | 10.98 |
| **Base of PMMA 2% on glass** | | 32.9 | 481 | 10.95 | 69.3 | 10.95 |
| **Isopropanol variant on glass** | 0.1 g | 33.29 | 479.1 | 10.99 | 68.9 | 10.99 |
| | 0.2 g | 33.24 | 479.1 | 10.99 | 69.0 | 10.99 |
| | 0.5 g | 33.27 | 479.1 | 11.00 | 69.0 | 11.00 |
| | 1.0 g | 33.01 | 478.6 | 10.89 | 68.9 | 10.89 |
| **Only-PMMA variant on glass** | 1% | 32.99 | 479.1 | 10.97 | 69.3 | 10.97 |
| | 2% | 33.11 | 479.1 | 10.99 | 69.3 | 10.99 |
| | 5% | 33.10 | 479.1 | 10.97 | 69.3 | 10.97 |
| | 10% | 32.79 | 479.0 | 10.89 | 69.3 | 10.89 |
| | 15% | 32.76 | 479.0 | 10.87 | 69.3 | 10.87 |
| | 20% | 32.50 | 478.7 | 10.79 | 69.3 | 10.79 |
| | 50% | 32.01 | 478.1 | 10.59 | 69.2 | 10.59 |

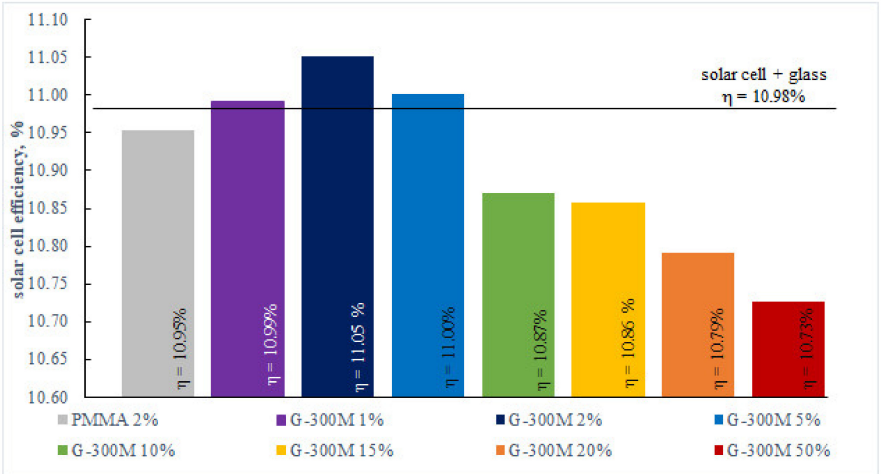



**Figure 9.** The efficiency comparison of G pigment in only PMMA variant.

Presented results show that it is possible to improve the EQE value for UV radiation range and thus, really increase the solar module efficiency using cheap and easy to deposit methods; however, specific analysis of both material composition and deposition technique is needed. It was clearly observed that promising candidates for efficient luminescent material might be selected from both groups of tested inorganic materials (BGL and G), but the PMMA matrix should be further optimized or even exchanged for preserving the efficient optical effect. Another issue is the proper mechanical condition of the deposited matrix for the long-life stability of the proposed construction in real-life harsh environmental conditions. All of the mentioned problems will be the topics of further investigation.

The layers have also been analyzed using SEM imaging. For SEM analysis, Carl Zeiss EVO MA10 SEM scanning electron microscope was used. In order to obtain a broader picture of the experiment, two detectors were used: SE for analyzing secondary electron images and BSD for analyzing backscattered electron images.

In Figure 10, there is an example of SEM pictures of the layer based on pigment G in only-PMMA variant for different concentrations. As can be observed, the layers are not homogeneous. The lower concentration has plenty of empty spaces where the pigment particles have not been located. This may cause the measurement results to have large discrepancies. Similar results can be observed for other layers too. A more detailed characterization of this issue is presented elsewhere [27].

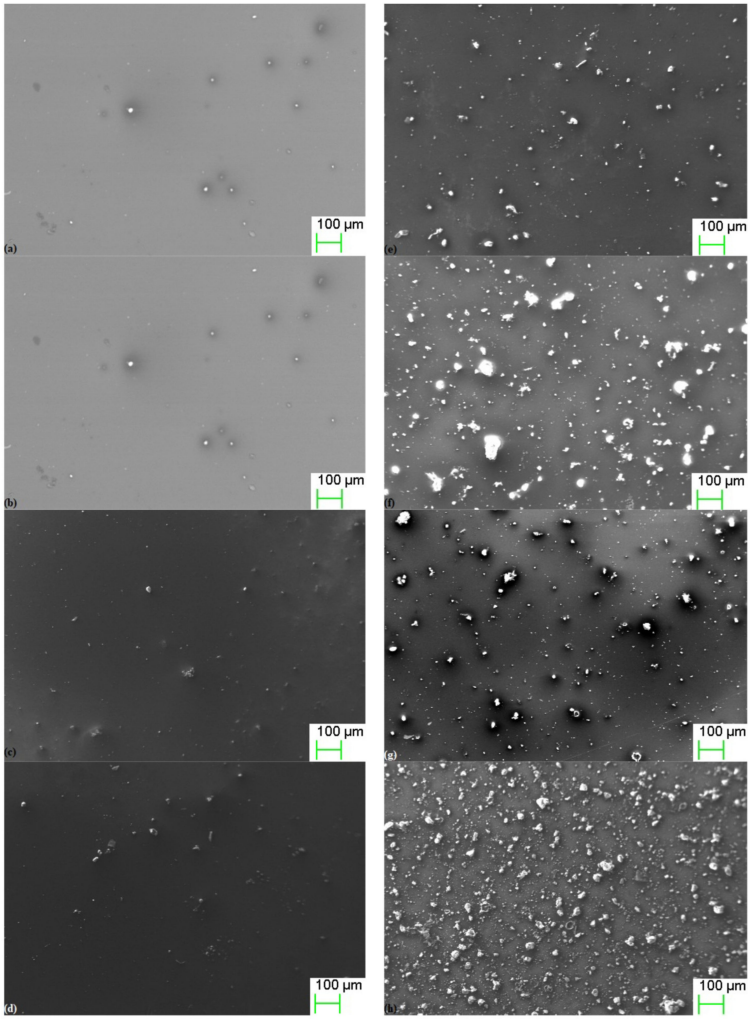



**Figure 10.** The SEM pictures of the layers deposited by the spray method in the version only with PMMA for the G pigment at a concentration of 1% for the SE (**a**) detector and BDS (**b**) detector, and with the SE detector for the concentration of 2% (**c**), 5% (**d**), 10% (**e**), 15% (**f**), 20% (**g**), and 50% (**h**).

## 4. Conclusions

The article presents results of technological experiments leading to successful employment of solar light conversion towards improvement on Si-based PV modules. The spray-coating method has been implemented as an effective, simple, and scalable deposition technique for converting layers of novel, innovative compositions. As a result, a series of converting layers with the desirable properties was obtained. The analysis of external quantum efficiency for each variant of these layers showed that using BGL or G pigment can improve the value of Si-based PV module EQE in the UV radiation range. The highest growth was achieved for G pigment, where the EQE value increased from 40.80% to 47.63% for the wavelength 355 nm. This is an improvement of 6.83 percentage points and consequently leads to a relative improvement of 16.74%.

The enhancement of solar cell efficiency is not so spectacular, although there is a huge potential for an increase in that parameter. As in the case of EQE improvement, also here, the greatest increase in photoconversion efficiency was observed for the pigment G-based layer in the variant with only PMMA for 2% concentration of the pigment. The growth is from 10.98% up to 11.05%.

The results show that elaborated composition and deposition solutions have a huge potential for future applications. There is a possibility to use simple, inexpensive, and scalable techniques to manufacture the down-shifting conversion layers not only to improve the EQE characteristic in the UV range but also to enhance the solar cell efficiency.

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