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Recent Progresses in Perovskite Solar Cells

Serafettin Demic, Ahmet Nuri Ozcivan,
Mustafa Can, Cebrail Ozbek and Merve Karakaya

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Abstract

Perovskite solar cell (PSC) can be regarded as a continuation of dye sensitized solar cell (DSSC) in terms of the sensitization phenomena that occurred in the functioning molecules. In 2012, a breakthrough propose has been made for the sensitization of PSCs, in which a solid-state structure is offered as an equivalent sensitizer used in DSSC. The power conversion efficiency (PCE) of those solid-state cells reached about twofold of its initial value during the past several years. Immediately after, the researchers followed this propose worldwide. They have introduced an improved efficiency of as much as 20%, which was originally started from its initial value of 4%, just in 4 years. Thus, the new concept, solid perovskite molecules, has eliminated the need for the liquid electrolyte in DSSC while still carrying the advantages of organic solar cells (OSCs). Therefore, the distinctive material of PSC—the organometallic halide molecules (also known as OMH or organic-inorganic trihalides)—inclined an unexpected reputation for solar cell (SC) researches. Hence, it seems that we will witness a new age for solar conversion devices depending on the recent hopeful progresses on PSCs. The high rate of photovoltaic (PV) conversion capacity in PSC is generally expressed by the basic properties possessed by the organic-inorganic perovskite crystal, such as better optical properties and well diffused charges along huge distances during the charge transport. In addition, a low temperature processing is applicable during its production. Moreover, the perovskite layer provides a tunable band gap. Therefore, depending on better developments on designed molecules, PSC may gain extreme performances compared to the other competitors, such as OSC or DSSC devices. This chapter starts with a general discussion on the need for an affordable clean energy conversion device that is urgent for the future of humanity, due to publicly well-known global warming issue. In Section 2, basic properties of PSC are mentioned together with their structure and working principles. Section 3 continues with an overview on organometallic perovskite molecules after a brief introductory history is presented. The absorption and band gap properties are also discussed. Since most perovskite materials need a hole transporting material (HTMs) within the PSC, the kinds of HTMs that are designed for PSCs are described in Section 3. The rendering of long-term stabilization has special importance

for PSCs since the instability issue remained idle in spite of those recent increased efficiency values attained by various research groups. Therefore, the stability issues are discussed in a separate part in Section 4. We finally close the chapter discussing the challenges and opportunities relying on the chapter content. We note that the recent investigations on PSCs have special importance for its large-scale realization in order to make them ready for the photovoltaic industry of the future. Hence, there are various announced meetings focusing on its mass production due to the unexpected sharp rise of the perovskite efficiency in the last 6 years. Hence, all the new cutting-edge scientific findings are also dealt with commercialization issues now, in order to attain the desired low cost fabrication, including the yield of high purity and the formation of smooth films during the continual manufacture of perovskite layers.

Keywords: perovskite solar cells, hole transport materials, spiro-OMeTAD

1. Introduction: the need for new and affordable power converters

The novelties in solar cell (SC) technology may be attracting the widest human attention in the world due to its significance on its effectiveness on electricity production from free and abundant sunlight [1–3]. Namely, petroleum is the main source of energy in the world by a percentage of 40, where most of the petroleum is produced just by a few oil exporting countries. The other countries are the consumers. They exist with increasing petroleum dependency, which means that the most of the countries rely on exporters. Unfortunately, the petroleum consumption accelerates quickly because of energy needs due to expanding industrial zones. It is expected to grow further unless an affordable novel clean energy technology becomes available.

To make a continuous delivery of the energy by means of an environmentally friendly manner, it is mandatory for governments to make immediate precautions against fuel consumption, particularly by strictly improving their alternative energy themes. Among a variety of energy sources, electricity is known as “clean energy”. The best quality of electricity is its ease of transformation in other energy forms; thereby, it is crucial for the human society. Accordingly, half of the energy consumption is made by electricity using machinery, for that, conversion of energy into electric energy from other resources is related to fuel saving. This property directed countries to invest on renewable energy transformers (e.g., Germany has already announced that much of the country’s consumption is fulfilled through renewables). Hence, many researchers from various institutions worldwide developed new types of alternative energy conversion devices in order to condense their capability more and more. Among them, SCs are the most promising devices since sun energy is accepted to be almost infinite for human needs.

Many new types of SCs in photovoltaic (PV) panels are used in new existing investments for meeting the residential and commercial energy needs. Maximizing efficiency is a must to satisfy those huge consumption needs. Hence, a majority of the investments obliged to use silicon-based PV panels, which have high conversion rates. Whereas for their production

processes, we need to use huge amount of water and energy and to release lots of pollutants to the environment as well. At this point, organic solar cell (OSC) technology attracts attention especially for the utilization of environmentally friendly production procedures. By means of this favorable property, in the last two decades, many new usages of organic materials in electric energy producing devices are proposed and applied. These novel devices are basically expected to satisfy low cost criteria and high efficiency; thereby, recent SC researches are especially shaped around these related topics.

Among SCs, many efficient types of organic materials and devices are present today. For example, commercial dye sensitized solar cells (DSSCs) are in the marketplace for a decade. However, this type of SCs is weak for its possible leaking in electrolyte liquid and imperfect encapsulation. The consistent and cutting-edge studies on this challenge give the fruit of a DSSC like but a novel SC: named, Perovskite Solar Cell (PSC). Relatedly, this chapter mainly deals with the main issues of PSC, i.e., its ideal phenomena, device structure, materials, and manufacturing challenges [4–17].

2. Operating principle and structural layout of PSC

As for the operating principle of PSCs, the present information obtained on “how they operate” is rather insufficient for now. Moreover, the whole working principle is not even well explained [18, 19]. There have already been many different approaches to point out a well-defined PSC operation mechanism. Nevertheless, the proficient working of methylammonium lead iodide (corresponding chemical formula is $\text{CH}_3\text{NH}_3\text{PbI}_3$ and the methylammonium inside the formulas are also introduced as MA or CH_3NH_3^+ in the related literature)-based PSC has not yet been completely understood. Accordingly, this confusion raises the necessity for deriving novel clues on (1) light absorption, (2) charge separation, (3) charge transport, and (4) charge collection. This is because these are general SC parameters that are used to express the principal working process during conversion of sunlight into electricity.

Selection of light harvesters is the first step for the determination of the physical layout of a SC. Hence, investigation of perovskite's optoelectronic properties has priority during the design process. This is needed in order to theoretically construct the fundamental energy conversion process, and therefore, to decide the respective SC layout [1]. In this term, we know that organometallic perovskite —like $\text{CH}_3\text{NH}_3\text{PbI}_3$ — may exhibit both electron and hole transport features, together. Hence, PSCs can be decided to be layered either using well-known *p-n* junction layout or *p-i-n* junction layout. The decision between the two layouts is made as follows: If the light harvester, perovskite, is an intrinsic semiconductor, a *p-i-n* junction is needed. Whereas a *p-n* junction is required if the light harvester, i.e. perovskite, has an *n*-type or *p*-type property. This is because both types are able to carry electrons or holes to the light harvester [1].

PSCs are fabricated depending on two typical structures of perovskites. These structures are called mesoporous and planar structures. These structures are unique to the perovskite crystal. **Figure 1** illustrates the consequent scheme for the device layout for both mesoporous- and

planar-type PSC. The former contains a mesoporous-type metal oxide layer (i.e., TiO_2 or Al_2O_3) coated with perovskite sensitizer. On the other hand, the latter includes a perovskite film sandwiched between electron and hole transporting layers. Here, in case of perovskite dots are stick to the surface of TiO_2 layer, charge is separated through electron injection. Namely, the charge certainly follows away from perovskite to the metal oxide (also known as transparent conductive oxide layer, TCO), such as TiO_2 [20]. Up to here, the operation principle is considered to be analogous to DSSCs. The remaining operation mechanism, however, is different from DSSCs for both mesoporous- and planar-type PSCs due to the charge accumulation and charge transport characteristics that occur in the PSCs [18, 19, 21]. Comparing these two architectures, it is evident that the charge transport rate is virtually the same for both of them, while mesoporous-type cell performs higher recombination rates [22]. The planar-type PSCs are found applicable to flexible solar cells since the typical high temperature rates are not mandatory in order to fabricate them.

Figure 1. (a) The model structure of mesoporous halide PSC and (b) the model structure of planar hetero-junction structured halide PSC.

Besides these two structures, hole conductor-free (HTM-free) PSCs have also been investigated by many scientists. The first report on HTM-free $\text{CH}_3\text{NH}_3\text{PbI}_3/\text{TiO}_2$ hetero-junction SC was released by Etgar et al. [23]. In this study, the mesoporous TiO_2 was coated with a thick perovskite layer having large crystal functioning as both light harvester and hole transport material, simultaneously. In order to obtain highly efficient PSCs and to avoid building shunt pathways, a thick perovskite film having a smooth surface is required. The width of the depletion layer at the $\text{CH}_3\text{NH}_3\text{PbI}_3/\text{TiO}_2$ junction plays an important role on the performance of the HTM-free PSC. For instance, it is possible to enlarge the width by increasing the depleted part of TiO_2 according to this study. Such arrangement has given an increased power conversion efficiency (PCE) of almost 11% [24]. Another HTM-free PSC is the one that has tripled layers that is implemented by Han et al. The respective multilayer arrangement consists of simple layout from mesoporous $\text{TiO}_2/\text{ZrO}_2/\text{C}$ layers [25]. Here, the ZrO_2 film is especially used as blocking layer so that the photo-generated electrons could not flow back to the contact. Accordingly, electron hole recombination process occurring inside the device is delayed. With the help of such HTM-free PSC, researchers have already achieved a PCE of around 13% [25].

The charge transfer processes and the latest progresses in PSC are summarized in **Figure 2** and **Table 1** [26]. The explained photonic interaction matches well for a mesoporous-type PSC. Here, the energy level of the perovskite is designed in order to function in the following junction architecture: $\text{TiO}_2/\text{CH}_3\text{NH}_3\text{PbI}_3/\text{spiro-OMeTAD}$. The charge transfer process for analogous mesoporous architecture requires the steps specified in **Table 1** [26–28]. Here, the

general PCE of mesoporous-type PSC is controlled through the processes given in the table. Where the rate of charge generation as well as charge transport occurred from (1) to (3) should be much faster than the rate of undesired recombination occurred from (4) to (8) given in **Figure 2**. In this way, a high power conversion efficiency can be obtained.

Step no	Event	Occurrence
1	Photoexcitation of the Perovskite	$\text{Perovskite} + h\nu \rightarrow (e^- \dots h^+)_{\text{Perovskite}}$
2	Electron injection	$(e^- \dots h^+)_{\text{Perovskite}} \rightarrow e^-_{\text{cb}(\text{TiO}_2)} + h^+_{(\text{Perovskite})} \rightarrow h^+_{(\text{HTM})}$
3	Hole injection	$(e^- \dots h^+)_{\text{Perovskite}} \rightarrow h^+_{(\text{HTM})} + e^-_{\text{Perovskite}}, e^-_{(\text{Perovskite})} \rightarrow e^-_{\text{cb}(\text{TiO}_2)}$
4	Photoluminescence	$(e^- \dots h^+)_{\text{Perovskite}} \rightarrow h\nu$
5	Nonradiative recombination	$(e^- \dots h^+)_{\text{Perovskite}} \rightarrow \nabla$
6	Back electron transfer at $\text{TiO}_2/\text{Perovskite}$ interface	$e^-_{\text{cb}(\text{TiO}_2)} + h^+_{(\text{Perovskite})} \rightarrow \nabla$
7	Back charge transfer at Perovskite/HTM interface	$h^+_{(\text{HTM})} + e^-_{(\text{Perovskite})} \rightarrow \nabla$
8	Charge recombination at TiO_2/HTM interface	$e^-_{\text{cb}(\text{TiO}_2)} + h^+_{(\text{HTM})} \rightarrow \nabla$

Table 1. The required charge transfer process for analogous mesoporous architecture [26–28].

Figure 2. Electron and hole transport process for mesoporous PSC.

It is known that for a single-junction SC the ideal band gap should start from 1.1 and extend to 1.4 eV [27]. One of the most important characteristics related to perovskite material processes is that it is possible to vary this band gap by altering its composition. Fortunately, in PSC, the band gap may be arranged by changing from 1.2 and extend to 2.3 eV. This band gap control can be made by substituting halide composition of $\text{CH}_3\text{NH}_3\text{Pb}(\text{I}_{1-x}\text{Br}_x)_3$ as

described by Noh et al. [23]. Another way to alter the respective band gap is to increase the cation size, nevertheless it may also result in a decrease in the band gap, unfortunately [28].

The solid-state HTM was first implemented to DSSCs to substitute the undesired liquid electrolyte since it causes fast degradation of the cell if it is used with perovskite crystals. This advancement in structural layout has showed an increase in the open circuit voltage (V_{oc}) of the cell due to the high redox potential of the liquid. Within those cells, HTM layer gradually becomes a solid-state layer instead of a liquid one. Accordingly, the stability of PSC improves. Since after the development of solid-state DSSCs, the most common HTM utilized in PSCs is spiro-OMeTAD (2,2',7,7'-tetrakis (*N,N*-di-*p*-methoxyphenylamine)-9,9'-spirobifluorene). This is a kind of small molecule which has improved the PCE of PSC up to 19% [29]. It was shown that an HTM with high conductivity decreases the series resistance and gives rise to improvement in the fill factor (FF) of the device [30]. For example, Seok et al. improved the V_{oc} and FF, just by substituting spiro-OMeTAD with polytriarylamine-type HTM [31]. The FF in turn can also be increased through improving the film quality by achieving enlarged grains and reduced grain boundaries [32].

In addition to replacing organic HTMs with other organic HTMs for the purpose of increasing PCE of the PSCs, there are also some other studies on substitution of organic HTMs with inorganics in order to increase the stability of the PSCs. It is well known that perovskite used as light harvester are rather susceptible to moisture and air resulting in degradation of the cell because of perovskite's low energy of formation [14]. One of the most important reasons why HTM usage is necessary is to protect the photoactive perovskite layer from moisture and air exposure to achieve highly stable PSCs. It is also known that metal oxides have much higher mobility as well as good stability than the generally used organic HTMs mentioned above [33, 34]. In an attempt to increase the stability, therefore, You et al. replaced organic HTM with inorganic charge transport layers [14]. In this study, *p*-type (nickel oxide, NiO_x) and *n*-type (zinc oxide, ZnO) metal oxides were used as electron and hole transport layers, respectively. The device architecture was composed of glass/indium tin oxide/ NiO_x / $CH_3NH_3PbI_3$ /ZnO/Al, where NiO_x was used to provide a perovskite layer with better crystallinity and ZnO acted as a coverage layer for perovskite layer, thus preventing degradation. According to the results of this study, the metal oxide-based PSCs demonstrated PCE of around 16% and higher stability compared to those fabricated with other common organic HTMs.

3. PSC materials

3.1. Perovskite as a solar device material: structure

Perovskite is a crystal with a special structure constituting ABX_3 formulation. It contains three same kind cationic ions and two different anionic ions. Here, A, B, and X represent large-dimension earth metal cation, rather smaller-dimension metal cation compared to A, and, either oxygen or halogen, respectively. The perovskite structure is symmetric. Within this structure, A is always larger than X (see **Figure 3**). Here, A-cations hold the octahedral corner coordinates while B-cations hold the octahedral holes; in 3-D structure. Within the composi-

tion, X can be substituted with numerous elements of O, Cl, Br, I, and S. The placements of atoms in 3-D structure are mainly regarded to be related with chemical charge neutralization process that is required for stability [8]. The crucial thing for the perovskite minerals is that, although the fundamental structure appears simple, they perform quite different characteristics under special circumstances, i.e., against structural distortion.

Figure 3. Crystal structure belonging to perovskite structure.

Gustav Rose, a German mineralogist, first recognized perovskite. Nevertheless, its entitlement is given after another mineralogist, Lew Perovskii, from Russia [35–37]. Perovskite is therefore originally a name for the mineral calcium TiO_2 (CaTiO_3) discovered by Perovskii, where it is basically used to refer a crystal structure that is the same as CaTiO_3 . Starting from its discovery, perovskite structure has widely been used in diverse fields of research, such as superconductors, ferroelectricity, thermoelectric, dielectric, magnetoresistive, piezoelectric, conducting, electrooptic, semiconducting, [5, 8, 9, 38–47], etc. The wide range of their application carried out in scientific research is mostly due to their tunable behaviors within the crystal structure observed since its discovery [4]. In particular, researchers have focused on organometallic perovskites which must include anionic halogens (I, Cl, F, and Br) and cationic metals of carbon family like Ge, Pb, and Sn. The attention on them is basically due to their unique structure and their promising response to photovoltaic applications [5–7].

The well-known perovskites are crystals of MgSiO_3 , SrFeO_3 , SrZrO_3 , BaTiO_3 , LiNbO_3 , and KMgO_3 . They originally give clear diffraction peaks at 110, 112, 220, 310, 224, and 314. In spite of its applicability to the mentioned diverse areas, the attention on the original molecules remained rather idle. Mostly, its actual fame has begun with the discovery of organometallic halide perovskite (OMH perovskite) related to the studies made for searching the interactions between the organic-inorganic materials. Namely, a research result claimed that a perovskite material shifts from the semiconducting state to the conduction region along with increased dimensionality [8, 9, 38]. In the mentioned and other related researches, A is exchanged by a cationic organic molecule, B is exchanged by an inorganic transition metal, and X is exchanged by an anionic halide. Hence, the new structure becomes inherently referred to as “OMH perovskite”. Among many transition metal alternatives, Sn^{+2} and Pb^{+2} metals substituted for X

are the ones which have collected the scientific interest on the following formulas: “ $\text{CH}_3\text{NH}_3\text{SnI}_3$ ” and “ $\text{CH}_3\text{NH}_3\text{PbI}_3$.” This attention increases day by day since the respective new molecules inherently carry significant optoelectrical properties and they are able to be processed in rather low temperatures compared to other OMH molecules with rather different formulas [47–49]. Even more, Pb^{+2} and Sn^{+2} cations are nonreactive, stand stable at room temperature, and their supply channels are wide due to being abundant, which makes them easily accessible cheap materials [50].

As for PV applications, Miyasaka et al. are the first group who used perovskite in SC panel production in 2009 [10]. They fabricated a SC with a liquid phase electrolyte based on mesoporous TiO_2 sensitized with $\text{CH}_3\text{NH}_3\text{X}_3$. Here, X is able to be substituted by I, Cl, or Br, depending on the application purpose. Similar to this first study on PSCs, the most prevalent organic compound used to fabricate PSCs are (1) CH_3NH_3^+ (methyl ammonium), (2) $\text{CH}_3\text{CH}_2\text{NH}_3^+$, (ethyl ammonium), and, (3) $\text{NH}_2\text{CH}=\text{NH}_2^+$ (form amidinium) [20, 51, 52]. Though, among all, the most common perovskite material implemented to the recent SC applications perhaps have been methylammonium lead iodide ($\text{CH}_3\text{NH}_3\text{PbI}_3$) with other lead halide structures ($\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Br}_x$ or $\text{CH}_3\text{NH}_3\text{PbI}_{3-x}\text{Cl}_x$) [37, 53].

3.2. Perovskite's absorption properties

It is beneficial to remind the absorbance of DSSC device here. Namely, the previous form of PSC, DSSCs, contains an *n*-type TiO_2 that has mesoporous structure. The dyes are stick to TiO_2 where the light is absorbed by dye molecules impregnated in a redox electrolyte sea, meantime. Here, the electrolyte regenerates the stable dye again and again by supplying electrons after having each excitation followed by charge transfer. The TiO_2 plays the role of helping the enlargement of the electrode surface as much as possible on behalf of the electrode's job. In this term, the soaring of incident photons become eased. In practice, an approximate 10 μm film thickness of TiO_2 is generally enough in DSSC to make a complete absorption of the visible light [9, 54, 55].

On the other hand, such broad thickness is not appropriate for the solid-state SCs. Because there are various factors that restrict the increase in film thickness. Due to these restrictions, the thickness is limited below 2 μm [9, 30]. In order to relieve these limiting effects, different active semiconducting layers are jointly coated with different thin film types in many research groups. Another approach to resolve this issue is the use of nano-sized quantum dots (QDs). These additional cares are taken in order to increase the sensitivity against the photons. As a result, the absorption band becomes carried to around near-infrared region (NIR) in several researches [30, 56–60].

Under the light of these facts expressed about improved absorption, more sensitization of DSSCs to the incident light worked in 2006 where $\text{CH}_3\text{NH}_3\text{PbI}_3$ and $\text{CH}_3\text{NH}_3\text{BrI}_3$ were used in the redox couple [9, 61, 62]. In some studies, on the other hand, they are applied as solid-state hole conductors. Nevertheless, those applications could only give efficiency as low as 0.2%. Whereas when they were applied to liquid-based DSSCs, it has reached as high as 10 times of this value escalating to around 2%. Therefore, the solid-state applications remained rather idle.

devices, the degradation process should be clearly understood where there are few reports about the effects of O_2 on perovskite films. According to the study about the degradation mechanism of the perovskite under water [109], it is shown that the hydrolysis reaction of $CH_3NH_3PbI_3$ arises under humid condition.

Photo-stability: The mesoporous TiO_2 layer in the device is preferred in order to easily transport photo-generated electrons. On the other hand, TiO_2 is inherently sensitive to ultraviolet light, which may cause degradation in PSCs. The instability of encapsulated and nonencapsulated perovskite devices was investigated through considering the device efficiency by Snaith et al. [110], and it is showed that the degradation of the first-type device occurs faster than that of the second type. Nevertheless, the first device has given a more stable condition in the lack of UV light. In order to overcome this photo-instability due to the TiO_2 layer, the respective authors suggest some methods such as pacifying the trap states, replacing the TiO_2 layer with other materials, and avoiding UV light from the TiO_2 layer. For example, stability at over 1000 hours at $40^\circ C$ is accomplished when mesoporous Al_2O_3 is used instead of TiO_2 layer in the PSC device. Nonetheless, the PCE was decreased to around by half of its first value after the first 200 hours. A continuing reduction is also observed in both V_{oc} and FF values.

Thermal stability: Thermal stability is an issue regarding both perovskite material and in HTM layer. The intrinsic thermal instability of a perovskite material was reported in the literature [111], and it is showed that even though the film was maintained in an inert condition, the degradation of perovskite is seen at $85^\circ C$. It means that the SCs may not be used properly in cases where the SC temperature exceeds this temperature level.

まとめ

Perovskite has been the most initiative and promising material in terms of energy harvesting among organic, inorganic, and organic-inorganic SCs [8]. In case the difficulties, which are mainly about stability, are overcome, the humanity would face to a new and affordable solar energy harvester for future use. Therefore, the novelties in PSC technology attract a large amount of attention in our recent world due to its significance on its effectiveness on electricity production from free and abundant sunlight.

The main prospects for the future cover the strategies in terms of reducing the band gap, enhancing the light absorption capacity, and improving the conversion efficiency. In addition, exposing the charge transport properties and improving interfacial engineering methods in device fabrication are important. Especially, the chemistry work on chemical composition has great value. Hence, the lion's share for future work should be explaining the photo-physical mechanism of solid-state SCs with respect to different chemicals.

Unfortunately, the present information obtained on "how PSCs operate?" is rather insufficient at least for now. Because the complete working principle is not well explained [18, 19]. Therefore, deriving novel clues on (1) light absorption, (2) charge separation, (3) charge transport, and (4) charge collection are needed during research since these four key issues are general SC parameters to identify the principal working of solar conversion process.

The PSC is mostly accompanied with a HTM layer. In view of the available HTMs, the inorganic ones are rather less progressed reaching an applied efficiency of 11%. On the other hand, these applications are promising in view of their improved stability against ambient conditions. With respect to these, polymeric HTMs offer tunable oxidization potential and better surface morphology. In addition, the researchers also attained advanced HOMO levels [78, 91–93]. The efficiency of polymer HTMs-based SCs is reported to reach more than 16% [31, 94, 95]. The third kind of HTMs is small molecules. Among these, spiro-OMeTAD has been the most used HTM in PSCs till now since it has already been evaluated in solid-state DSSCs and OLED devices [67, 83, 112] (that is, spiro-OMeTAD has already been a standard HTM for researchers). The use of hybrid lead halide with spiro-OMeTAD-based HTM is reported to give an efficiency of around 15% PCE [82, 84]; however, the cost of spiro-OMeTAD remains high due to its difficult synthesis process. Therefore, novel small molecules should be designed and synthesized in order to develop cost-effective PSC devices.

We note that PSCs evolved rapidly and the semiconducting phenomena occurred within the cell remained shaded until now. Hence, the semiconducting behavior is continuously investigated benefitting from the current-voltage characteristics of SCs in some research groups [113]. Deriving an increased efficiency level in PSCs is also related to the way how deposition is carried out. The perfection within the selected coating method is also important [114, 115]. Besides all discussion presented here, we note that the use of completely wet processing method during the production of SCs is especially important for an easy fabrication. Therefore, studies made on “all-solution production process” are highly valuable since vacuum production is expensive and highly energy dependent [116].

Author details

Serafettin Demic^{1*}, Ahmet Nuri Ozcivan², Mustafa Can³, Cebirail Ozbek^{1,2} and Merve Karakaya⁴

*Address all correspondence to: serafettin.demic@gmail.com

1 Department of Materials Science and Engineering, Izmir Katip Celebi University, Izmir, Turkey

2 Department of Electrical and Electronics Engineering, Izmir Katip Celebi University, Izmir, Turkey

3 Department of Engineering Sciences, Izmir Katip Celebi University, Izmir, Turkey

4 Department of Materials Science and Engineering, Izmir Institute of Technology, Izmir, Turkey

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