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Self-assembled monolayer enabling improved buried interfaces in blade-coated perovskite solar cells for high efficiency and stability

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ABSTRACT

Despite the rapidly increased power conversion efficiency (PCE) of perovskite solar cells (PVSCs), it is still quite challenging to bring such promising photovoltaic technology to commercialization. One of the challenges is the upscaling from small-sized lab devices to large-scale modules or panels for production. Currently, most of the efficient inverted PVSCs are fabricated on top of poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA), which is a commonly used hole-transporting material, using spin-coating method to be incompatible with large-scale film deposition. Therefore, it is important to develop proper coating methods such as blade-coating or slot-die coating that can be compatible for producing large-area, high-quality perovskite thin films. It is found that due to the poor wettability of PTAA, the blade-coated perovskite films on PTAA surface are often inhomogeneous with large number of voids at the buried interface of the perovskite layer. To solve this problem, self-assembled monolayer (SAM)-based hole-extraction layer (HEL) with tunable headgroups on top of the SAM can be modified to provide better wettability and facilitate better interactions with the perovskite coated on top to passivate the interfacial defects. The more hydrophilic SAM surface can also facilitate the nucleation and growth of perovskite films fabricated by blade-coating methods, forming a compact and uniform buried interface. In addition, the SAM molecules can also be modified so their highest occupied molecular orbital (HOMO) levels can have a better energy alignment with the valence band maxima (VBM) of perovskite. Benefitted by the high-quality buried interface of perovskite on SAM-based substrate, the champion device shows a PCE of 18.47% and 14.64% for the devices with active areas of 0.105 cm² and 1.008 cm², respectively. In addition, the SAM-based device exhibits decent stability, which can maintain 90% of its initial efficiency after continuous operation for over 500 h at 40 °C in inert atmosphere. Moreover, the SAM-based perovskite mini-module exhibits a PCE of 14.13% with an aperture area of 18.0 cm². This work demonstrates the great potential of using SAMs as efficient HELs for upscaling PVSCs and producing high-quality buried interface for large-area perovskite films.

KEYWORDS

self-assembled monolayer, buried interface, perovskite solar cells, upscaling, blade-coating

1 Introduction

The emerging organic–inorganic hybrid perovskite solar cells (PVSCs) have reached a world-record power conversion efficiency (PCE) of 25.7% over the past decade, proving their great potential as a promising candidate for photovoltaic (PV) applications [1]. Since the efficiency of small-size PVSCs is approaching their theoretical limit [2], the research focus is gradually shifting to upscaling perovskite PV technology with continuous deposition methods such as blade-coating and slot-die coating, where the intractable problem is controlling the crystallization and growth of large-size perovskite films [3–7]. Different strategies such as solvent engineering and additive engineering have been employed to control perovskite crystallization and growth during scalable coating processes for achieving improved device efficiency and reproducibility [8–15]. Besides the perovskite layer improvement, the surface properties of substrates are also extremely important for upscaling perovskite films on top. Normally, for small-size inverted PVSCs, a spin-coated poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine] (PTAA) layer is used as the hole transport layer to achieve high efficiency [16–18]. However, the spin-coated PTAA layer is no longer suitable for large-area PVSCs due to its poor wettability and non-uniformity. Furthermore, it is still challenging
to ensure a compact adhesion between PTAA and large-area perovskite films. For example, Huang et al. [19] have found that large number of interfacial voids were formed at the buried interface of blade-coated perovskite films (perovskite/PTAA interface) due to the evaporation of dimethyl sulfoxide (DMSO) solvent. They then employed a solid-state lead-coordinating additive with high boiling point to partially replace DMSO and decrease the number of voids at the buried interface of the perovskite film for higher device efficiency and stability. Therefore, it is necessary to develop new interlayer materials for growing large-area perovskite films with better buried interface.

Recently, self-assembled monolayer (SAM) has been introduced as hole-extraction layer (HEL) for small-area PVSCs with high photovoltaic performance [20–23]. SAMs are ordered arrays of organic molecules which contain an anchoring group bonding to the substrate surface, a linkage determining the packing, and a functional headgroup tuning its surface properties. For example, the acid radical groups of SAM molecules can chemically bond with the hydroxyl groups on the surface of transparent conductive oxide (TCO) electrode, allowing the formation of a dense monolayer on the surface of TCO [24, 25]. The headgroup of SAM molecules can be modified to provide better wettability and facilitate better interactions with the perovskite on the top. Therefore, the SAM HEL has obvious advantages for upsampling perovskite films on the top, for example: (i) It can be prepared by both lab-scale and large-scale processes with good uniformity; (ii) it can be processed with green solvents such as ethanol in ambient air, which is suitable for large-scale production with less environmental impact; (iii) the functional headgroups of SAM molecules can be easily tuned to modify the interfacial properties for the upper perovskite films. Inspired by the above points, it is straightforward to consider the SAM rather than the PTAA for upsampling perovskite PV technology. However, as far as we know, there is little exploration in this area.

Therefore, in this work, we aim to explore the potential of the application of SAM in upsampling PVSCs. Firstly, we investigated the effect of substrates on the growth of blade-coated perovskite films in ambient air with high relative humidity (RH). By investigating the buried interface of perovskite films grown on PTAA and SAM layers, we find that the SAM surface facilitates the nucleation and growth of perovskite film after blade-coating process, forming a dense perovskite layer with strong adhesion to the substrate [26]. Owing to the high-quality buried interface of perovskite layer on SAM-based substrate, the corresponding device shows a champion efficiency of 18.47% and 14.64% for the devices with active areas of 0.105 cm² and 1.008 cm², respectively. Furthermore, the SAM-based device shows decent long-term operational stability under continuous light illumination at 40 °C in an inert glovebox. Finally, we have demonstrated the potential of SAM in perovskite mini-module by achieving a PCE of 14.13% with an aperture area of 18.0 cm².

2 Experimental

Materials: Prepatterned indium tin oxide (ITO) glasses with sheet resistance of 15 Ω/sq were purchased from Liaoning Advanced Electron Technology Co., Ltd. (China). Poly(bis(4-phenyl)(2,4,6-trimethylphenyl)amine (PTAA), methylammonium iodide (MAI), fullerene (C₆₀), and bathocuproine (BCP) were bought from Xi’an Polymer Light Technology Corp. (China). Lead iodide (PbI₂, 99.9985%) and [2-(3,6-dimethoxy-9H-carbazol-9-yl)ethyl]phosphonic acid (MeO-2PACz) were purchased from Tokyo Chemical Industry Co., Ltd. (TCI, Japan). All solvents including isoopropanol (IPA, 99.5%), DMSO (99.5%), 2-methoxyethanol (2-ME, 99.5%), and chlorobenzene (CB, 99.5%) were purchased from J&K (China). Silver was purchased from commercial sources with high purity. All the materials were used as received without further purification.

Fabrication of PVSCs: Prepatterned ITO glasses were sequentially cleaned with detergent, deionized water, acetone, and ethanol for 30 min, respectively. Then they were dried in an oven at 75 °C overnight. Before use, ITO glasses were treated by ultraviolet-ozone for 20 min. The PTAA (2 mg/mL in chlorobenzene) layer was deposited on ITO glass by spin-coating (4,000 rpm for 30 s), followed by thermal annealing at 100 °C for 10 min. For the SAM formation, the MeO-2PACz solution (0.3 mg/mL in IPA) was spin-coated at 3,000 rpm for 30 s and then annealed at 100 °C for 10 min. A washing spin-coating step (with IPA) was conducted at 3,000 rpm for 20 s to remove the excess MeO-2PACz molecules. Subsequently, the perovskite film was blade-coated with air knife blowing at room temperature in ambient air with a relative humidity of ~50%. The 1 M methylammonium lead iodide (MAPbI₃) precursor solution was prepared by mixing equimolar MAI and PbI₂ in 2-ME with addition of DMSO (1.5%, v/v). Then, the precursor solution was blade-coated with a gap of ~100 μm at a speed of 15 mm/s. The blowing pressure of the air knife was 2 bar. The as-coated perovskite film was annealed at 100 °C for 3 min in ambient air. Finally, the C₆₀ (25 nm), BCP (6 nm), and Ag (100 nm) layers were thermally evaporated on the perovskite film through a shadow mask with an active area of 0.105 cm² or 1.008 cm². The mini-modules were fabricated on the pre-patterned ITO glass substrates (5.0 cm × 5.0 cm) with P1 (200 μm width) following the same procedure as the small-area solar cells. The fabricated modules have 5 sub-cells, and each sub-cell has a width of 9.0 mm. For the P2 and P3 process, the laser scribing was performed with a SCIPRIOS Laser system (PE-S2S-300, 532 nm). The widths of P2 and P3 were measured to be ~100 nm and ~110 μm, respectively. The distances between P1 and P2, P2 and P3 are about 200 and 400 μm, respectively. The total scribing line width was 1,000 μm, giving a geometrical fill factor (GFF) of 89%.

Characterizations: The buried interface morphologies of perovskite films were characterized by scanning electron microscopy (SEM, TESCAN MIRA3) at 5 kV accelerating voltage. The X-ray diffraction (XRD) patterns of perovskite films were measured using a Bruker eco D8 with 30 kV voltage and 15 mA current. The ultraviolet photoelectron spectroscopy (UPS) characterizations were conducted on an AXIS SUPERA X (KRATOS Scientific) at an ultrahigh vacuum of 1.0 × 10⁻¹⁰ Torr. Current density–voltage (J–V) curves of PVSCs were measured by a Keithley 2400 source meter under illumination of a solar simulator (Enlitech, SS-F5). The illumination intensity was calibrated using a silicon calibrated detector (Newport). External quantum efficiency (EQE) spectra of the devices were obtained by an EQE measurement system (Enlitech). The electroluminescence (EL) images of the PVSCs were captured using a MV-CA050-10GM camera (Hikrobot Technology).

3 Results and discussion

3.1 Buried interface of blade-coated perovskite films

We first sought to study the crystallization process of blade-
coated perovskite films in ambient air. To exclude other effects from perovskite composition or substrates, the prototypical MAPbI$_3$ was blade-coated on ITO glass substrates directly. Considering that a fast solvent evaporation rate is required to achieve rapid nucleation of perovskite during blade-coating, the more volatile 2-ME was used as the solvent instead of the less volatile dimethylformamide (DMF) for perovskite precursors. We found that the addition of an appropriate amount of DMSO in perovskite precursor solution was critical to the quality of perovskite films, especially the morphology of perovskite layer at the buried interface (Fig. 1(a)). As shown in the SEM image in Fig. 1(b), there are large numbers of voids at the buried interface of the perovskite film and the perovskite crystals exhibit irregular shapes when the solvent is pristine 2-ME. These results can be ascribed to the fast nucleation and growth of perovskite at the initial stage of the blade-coating process, where the fast evaporation of 2-ME solvent induces a dry environment to suppress the sustained growth of perovskite crystals into a continuous network. It is found that the poor perovskite morphology is alleviated when DMSO is added into the perovskite precursor solution with a volume ratio of 1% (Fig. 1(c)), indicating that DMSO is still not sufficient to retard the fast crystallization of perovskite during blade-coating. Further increasing the DMSO ratio to 1.5% allows the formation of a relatively dense and uniform perovskite film as shown in Fig. 1(d). However, voids are formed when the DMSO additive is increased to 2% (Fig. 1(e)). It can be ascribed to the slow volatilization of the excess DMSO during thermal annealing, leaving interfacial voids in the perovskite films.

To evaluate the effect of substrate surface on the growth of perovskite films, we then further investigated the buried interface of perovskite layers blade-coated on PTAA- and SAM-based ITO glass substrates, respectively. MeO-2PACz was used as the SAM in this work. As shown in Figs. 2(a) and 2(b), both the blade-coated perovskite films on PTAA- and SAM-based substrates exhibit smooth and shiny surfaces. The XRD results further confirm that both perovskite films exhibit good crystallinity with the identical preferential (110) direction at 14.2° (Fig. S1 in the Electronic Supplementary Material (ESM)). However, significant differences are found on the buried interfaces of the perovskite films. As shown in Fig. 2(c), the perovskite grain boundary on the PTAA-based substrate is clear, and the grain has stereoscopic sense and depth. The bright and dark area of the buried interface indicates that the perovskite layer blade-coated on PTAA is not flat and compact. On the contrary, for the buried interface of the perovskite film on SAM, the grain boundary is relatively unclear and the whole plane is relatively flat with a relatively uniform brightness (Fig. 2(d)). To further evaluate the adhesion of the perovskite layer on PTAA and SAM, we conducted cross-sectional SEM on the samples. As shown in Figs. 2(e) and 2(f), it is obvious that there is a gap at the junction between the perovskite film and PTAA-based substrate. The buried interface of perovskite shows a fluctuating edge rather than a smooth straight line when it is blade-coated on PTAA, while the perovskite film is tightly attached to the substrate without observable gaps when it is blade-coated on the SAM. Based on the above results, it can be concluded that the adhesion between the blade-coated perovskite and SAM-based substrate is more compact. It is believed that such difference in the buried interface of perovskite films will affect the electronic properties of the device especially the charge extraction from the perovskite films, which will be discussed in the following section.

To understand the substrate-dependent buried interfacial properties of blade-coated perovskite, we measured the surface wettability of PTAA- and SAM-based substrates, respectively. As shown in Figs. 3(a) and 3(b), the contact angle of the SAM-based substrate is 52.5°, much smaller than that of PTAA-based substrate (76.7°). Therefore, we speculate that the different buried interface morphologies of perovskite films can be ascribed to the wettability of the substrates which influences the nucleation and growth of the upper perovskites. As shown in Fig. 3(c), at the initial nucleation stage after the blade-coating, the formation of perovskite nuclei is retarded by the hydrophobic PTAA surface, resulting in discrete perovskite nuclei with a large distance. Then at the growth stage during the following thermal annealing process, the intermediate phase can convert into dense perovskite crystals through vertical direction, while the perovskite growth in horizontal direction may not be sufficient to connect the crystals due to the large distance between the initial nuclei. On the other hand, the relatively hydrophilic SAM surface can...
facilitate the perovskite nucleation at the initial stage of the blade-coating, forming dense perovskite nuclei on its surface. Then at the perovskite growth stage, the intermediate phase can form dense crystals in both vertical and horizontal directions, ensuring the adhesion of perovskite films on SAM-based substrates as illustrated in Fig. 3(d). In addition to the above-mentioned mechanism, we speculate that the methoxy headgroups of the MeO-2PACz SAM molecules pointing to the perovskite layer may form coordination bonds with lead ions (Fig. S2 in the ESM), which further enhances the adhesion at the buried interface of perovskite films [27, 28].

We further investigated the energy level alignment at the buried interface of perovskite films by UPS measurement. As shown in Fig. 4(a), the SAM-based substrate shows lower cut-off binding energy than that of PTAA-based substrate, indicating that the SAM surface has a deeper Fermi level ($E_F$). In addition, the two substrates have an identical energy gap of 0.42 eV between the highest occupied molecular orbital (HOMO) level and $E_F$. Based on the UPS results, the energy level alignment at the buried perovskite interface is illustrated in Fig. 4(b), which shows HOMO levels and $E_F$. Based on the UPS results, the energy level alignment at the buried perovskite interface is illustrated in Fig. 4(b), which shows that the HOMO level of MeO-2PACz SAM has a better energy alignment with the valence band maxima (VBM) of perovskite films.

3.2 Photovoltaic performance of PTAA- and SAM-based PVSCs

We further evaluated the photovoltaic performance of PTAA- and SAM-based PVSCs fabricated by blade-coating method. The PVSC devices have a structure of ITO glass/PTAA or SAM/MAPbI$_3$/C$_{60}$/BCP/Ag. As reported in previous work that the humidity of ambient air has strong impact on the photovoltaic performance of PVSCs [29], we have found a narrow humidity window (around 45%–55% RH) for blade-coating PVSCs with optimal performance (Fig. S3 in the ESM). We then blade-coated perovskite films on both PTAA- and SAM-based substrates under around 50% RH for PVSCs. The current $J$–$V$ characteristics of the champion devices are illustrated in Fig. 5(a), and the detailed photovoltaic parameters are summarized in Table 1. For the small-area PVSCs with 0.105 cm$^2$, the SAM-based PVSC exhibits a PCE of 18.47%, slightly higher than the best PTAA-based device with an efficiency of 17.33%. It can be found that all the devices show a negligible hysteresis during forward and reverse scans. Compared to the PTAA-based device, the SAM-based device shows improved open-circuit voltage ($V_{OC}$), short-circuit
current ($J_{SC}$) and fill factor (FF). The enhanced $V_{OC}$ can be ascribed to the deeper HOMO level of the SAM surface as indicated in the UPS results. The $J_{SC}$ improvement was evaluated by EQE measurement. From the EQE spectra shown in Fig. 5(b), it is obvious that the SAM-based device has a higher EQE than the PTAA counterpart in the wavelength ranges of 350–400 nm and 650–750 nm, resulting in a higher $J_{SC}$. Such improvement can be ascribed to the enhanced light absorption in SAM-based devices because the monolayer nature of SAM ensures the lowest parasitic absorption in devices.

As described in the above section that the contact between PTAA and perovskite layer at the buried interface is much poorer, which may be detrimental to the device photovoltaic performance. Such detrimental effect of buried interface will become significant with increasing the device area. As shown in Fig. 5(c), with the device area increasing to 1.008 cm$^2$, the SAM-based PVSC shows 14.64% efficiency while the PTAA-based device only has an efficiency of 11.92%. Electroluminescence (EL) imaging has been widely used to evaluate the quality of the electrical contacts of solar cells [30]. Therefore, we performed EL imaging on both PTAA- and SAM-based PVSCs as shown in Fig. 5(d). With the same voltage bias of 1.8 V, the brightness of PTAA-based devices is much lower than that of SAM-based devices, while the current density of the PTAA-based device is much higher than that of the SAM-based device (Fig. S4 in the ESM), indicating more energy loss through non-radiative recombination in the PTAA-based device. In addition, it is obvious that the SAM-based PVSC

<table>
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<tr>
<th>Substrate</th>
<th>Area (cm$^2$)</th>
<th>$V_{OC}$ (V)</th>
<th>$J_{SC}$ (mA/cm$^2$)</th>
<th>FF (%)</th>
<th>PCE (%)</th>
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<tbody>
<tr>
<td>PTAA</td>
<td>0.105</td>
<td>1.056</td>
<td>21.21</td>
<td>77.7</td>
<td>17.33</td>
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<tr>
<td>PTAA</td>
<td>0.105</td>
<td>1.054</td>
<td>21.20</td>
<td>76.1</td>
<td>16.93</td>
</tr>
<tr>
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<td>1.032</td>
<td>17.77</td>
<td>65.3</td>
<td>11.92</td>
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<tr>
<td>PTAA</td>
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<td>1.030</td>
<td>17.48</td>
<td>62.6</td>
<td>11.22</td>
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<tr>
<td>SAM</td>
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<td>1.081</td>
<td>21.57</td>
<td>78.9</td>
<td>18.39</td>
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<tr>
<td>SAM</td>
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<td>1.083</td>
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<td>1.065</td>
<td>19.90</td>
<td>67.0</td>
<td>14.20</td>
</tr>
<tr>
<td>SAM</td>
<td>1.008</td>
<td>1.073</td>
<td>19.83</td>
<td>68.8</td>
<td>14.64</td>
</tr>
</tbody>
</table>

Figure 5  (a) $J$–$V$ characteristics of PTAA-based PVSCs fabricated by blade-coating with an active area of 0.105 cm$^2$. (b) The EQE and integrated $J_{SC}$ of PTAA- and SAM-based PVSCs. (c) $J$–$V$ characteristics of SAM-based PVSCs fabricated by blade-coating with an active area of 1.008 cm$^2$. (d) EL imaging of PTAA- and SAM-based PVSCs. (e) MPPT of PTAA- and SAM-based PVSCs under AM 1.5G illumination in a N2 filled glovebox.
SAM-based PVSCs show decent stability under both MPPT light-soaking effect with ever-increasing performance, while the PCE of PTAA-based devices decreases directly. The efficiency of PTAA-based devices decreases to 50% after about 180 h, while SAM-based devices can maintain more than 90% efficiency after over 500 h. In addition, we further conducted the thermal stability of the devices according to the ISOS-D-2I protocol that the devices were stored under dark condition with elevated temperature [31]. Both the PTAA- and SAM-based devices were stored in an inert glovebox at 65 °C under dark condition and measured the J–V characteristics at set intervals. As shown in Fig. S6 in the ESM, the SAM-based device exhibits better thermal stability than the PTAA-based device, showing no degradation after nearly 200 h of aging, whereas the PTAA-based device loses around 12% of its initial efficiency after nearly 200 h. We believe that the fast degradation of the PTAA-based device is due to the poor buried interface of its perovskite film. The voids at the buried interface triggers decomposition of perovskite under continuous illumination, inducing the inferior stability performance of PTAA-based devices.

Moving forward, we blade-coated large-area perovskite films on the SAM-based substrates and fabricated perovskite mini-modules (5 cm × 5 cm) as shown in Fig. 6(a). The perovskite mini-module deposited on SAM-based substrate exhibits a Voc of 5.36 V, a Jsc of 4.17 mA/cm² and an FF of 63.2%, yielding a module efficiency of 14.13% with an aperture area of 18.0 cm² as shown in Fig. 6(b). Table S1 in the ESM shows the comparison of this work to previous reported PVSCs fabricated by blade-coating method. This is the first time to use SAM as HEL for perovskite modules and the SAM-based PVSCs show decent stability under both MPPT condition and elevated temperature aging condition. Although the current SAM-based perovskite mini-module only shows a PCE of 14.13%, we believe that by further molecular design of the SAM for both charge transport and perovskite defects passivation, it will promote the development of perovskite modules in both efficiency and stability.

4 Conclusions

In summary, efficient and stable MAPbI₃-based PVSCs have been fabricated by blade-coating on MeO-2PACz SAM-based substrates in ambient air with high humidity (~ 50% RH). Compared to the commonly used PTAA, the better wettability of SAM surface facilitates the perovskite nucleation and growth, resulting in a superior buried interface of perovskite films with stronger adhesion and uniformity. Owing to the compact contact between perovskite and SAM-modified substrates and the deep HOMO level of the MeO-2PACz SAM surface, the champion SAM-based PVSC fabricated by blade-coating exhibits a high efficiency of 18.47% for 0.105 cm² devices and 14.64% for 1.008 cm² devices. Furthermore, the SAM-device retains 90% of its initial efficiency after over 500 h under simulated 1 sun illumination at MPPT in a N₂ filled glovebox. Finally, an efficiency of 14.13% is achieved when we upscale lab-scale cells to 5 cm × 5 cm mini-modules with an aperture area of 18.0 cm² on SAM-based substrate, showing the great potential of SAMs in the application perovskite modules.

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Electronic Supplementary Material: Supplementary material (further details of XRD measurements, illustration of SAM-modified buried interface, J–V curves, LED spectrum of stability tests and thermal stability results) is available in the online version of this article at https://doi.org/10.26599/NRE.2022.9120004.

![Figure 6](image_url) (a) Photography of SAM-based perovskite mini-module (5 cm × 5 cm) by blade-coating method. (b) J–V characteristics of SAM-based perovskite mini-module fabricated by blade-coating with an aperture area of 18.0 cm².
Declaration of conflicting interests

The authors declare no conflicting interests regarding the content of this article.

References


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