Searching of new, cheap, air- and thermally stable hole transporting materials for perovskite solar cells

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In this work, two thermal- and air-stable, hole transporting materials (HTM) in perovskite solar cells are analyzed. Those obtained and investigated materials were two polyazomethines: the first one with three thiophene rings and 3,3′-dimethoxybenzidine moieties (S9) and the second one with three thiophene rings and fluorene moieties (S7). Furthermore, presented polyazomethines were characterized by Fourier transform infrared spectroscopy (FTIR), UV–vis spectroscopy, atomic force microscopy (AFM) and thermogravimetric analysis (TGA) experiments. Both polyazomethines (S7 and S9) possessed good thermal stability with a 5% weight loss at 406 and 377 °C, respectively. The conductivity of S7 was two orders of magnitude higher than for S9 polymer (2.7 × 10−8 S/cm, and 2.6 × 10−10 S/cm, respectively). Moreover, polyazomethine S9 exhibited 31 nm bathochromic shift of the absorption band maximum compared to S7.

Obtained polyazomethines were investigated by UV–vis and XRD. Electrical parameters of perovskite solar cells (PSC) were investigated at Standard Test Conditions (STC). It was found that both polyazomethines protect perovskite which is confirmed by ageing test where Voc did not decrease significantly for solar cells with HTM in contrast to solar cell without hole conductor, where Voc decrease was substantial. The best photoconversion efficiency (PCE ~ 6.9%), among two investigated in this work polyazomethines, was obtained for device with the following architectures FTO/TiO2/TiO2 + perovskite/S7/At. Stability test proved the procreative effects of polyazomethines on perovskite absorber.

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1. Introduction

Recently, perovskites solar cells have been an extremely hot topic in a photovoltaic world due to their excellent optical and electrical properties. General formula of hybrid perovskite is ABX3, where B is a bivalent cation such as lead (Pb2+) or tin (Sn2+), X is an anion, usually halide ion, for example chlorine (Cl−), iodide (I−) or bromide (Br−) while A is an organic cation as methylammonium (CH3NH3+), or formamidinium (NH(CH2)2) [1]. The crystal structure of perovskite consist of a corner sharing (BX6) octahedron where cation B is located in the centre while anion X in the corners. The biggest cation A is situated between the octahedrons and its main issue is to balance a perovskite structure [2]. The archetype of perovskite solar cells are Dye Sensitized Solar Cells (DSSC) which contain mesoporous TiO2 covered by organic, dye molecules, for which operating principle is given by Anders Hagfeldt and Michael Grätzel [3]. The parameters which make perovskite attractive to photovoltaic purposes are optimal band-gap energy which is equal to 1.5 eV–1.8 eV, low exciton binding energy, relatively high carrier lifetime (even 100 ns) compared with organic solar cells (10 ns) and high diffusion length of 100–1000 nm, depending on the compound composition [4]. Moreover, perovskites have low non-radiative recombination rates losses compared to other thin-film polycrystalline semiconductors [5].

Main disadvantages to be mentioned are, low ambient stability and UV light, oxygen and moisture sensitivity [6]. Furthermore, manufacturing of perovskite solar devices requires very expensive components. While, perovskite material itself is rather cheap, the most common, p-type hole transporter (Spiro-OMeTAD) is even several times more expensive than gold or platinum [7]. Perfect HTM should possess high hole mobility, energy levels aligned to
the other structure components: HOMO and LUMO levels of HTM should be accordingly higher and lower than valence and conduction band of perovskite, and do not damage the perovskite itself [8]. Moreover, it shall be hydrophobic and therefore, remains stable protection and encapsulation barrier for the perovskite layer [9,10]. Basically, organic and inorganic hole transporting materials can be distinguished.

The most common organic hole transporting material is 2,2′,7,7′-tetrakis-(N,N-di-p-methoxyphenylamine)-9,9′-spirobifluorene (Spiro-OMeTAD) and it is highly successful because of its large hole mobility and high solubility [9]. Nevertheless, Spiro-OMeTAD contains LiTFSi (bis(trifluoromethane)sulfonimide lithium salt) dopant which is sensitive to humidity and leads to poor stability of perovskite solar cells [8]. Besides that, Spiro-OMeTAD is very expensive due to complicated synthesis and high purity requirement [11]. This is the main reason why many research groups try to obtain cheaper HTM by modifying this compound. As a result, such examples as spiro-cyclopenta[2,1-b:3,4-b]dithiophene (spiro-CPTD), tetra[4-
\[N,N-(4,4-dimethoxydiphenylamino)]phenyl]ethane(TAE-1) [9] or Trux-OMeTAD which consists of a C3h Truxene-core with arylamine terminals and hexyl side-chains, can be found [10].

On the other hand poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) is cheap polymer with high conductivity. Typically, PEDOT:PSS applied as an HTM layer in inverted structure and it is usually located on the top of devices. Nonetheless, sometimes it can provide a part of a scaffold structure but in such a case it is very important to be aware of destructive influence of water on perovskite. Therefore, the PEDOT:PSS in toluene solution has to be used instead of water dilution. Liu et al. have shown that PEDOT:PSS has similar properties as Spiro-OMeTAD [8]. Moreover, it is possible to control conductivity of PEDOT by dopant changing [12]. Small amount of PSS improves conductivity simultaneously increasing the charge carrier mobility and therefore, better electrical parameters of device are obtained [12]. Besides, it affects the delayed degradation of perovskite [8]. Other common HTMs are poly(arylamine), poly(3-hexylthiophene) [13] or carbazole [14]. Interesting group of organic materials that can be found as hole transporting materials are azomethines. They are compounds with H=NC= groups known also as Schiff bases. Due to their donor properties it is possible to apply them in organic photovoltaics [15].

Among inorganic HTMs, Cu based inorganic semiconductors, quantum dots and Kesterites can be distinguished [11,16–18].

Despite all, the high performance and cheap HTM, and effective perovskite protective obstacle are still required. For this reason, the main goal of this work was to test two polyazomethines as HTMs and as a protective barrier in perovskite solar cells. Polyazomethines (PAZ) have been investigated over the past 30 years mainly as thermostable, biological, and liquid crystalline (LC) compounds [19,20]. However, in the last 16 years PAZs are also investigated as a donor component of an active layer in polymer solar cells [21–37]. The first article about polymer solar cells with PAZ was published by Sharma et al. in 1996 [23]. The authors studied the photovoltaic properties of poly(phenyl azomethine furane) (PPAF) in a monolayer device with a Ag/PPAF/ITO architecture and received a photoconversion efficiency (PCE) value of approximately 0.019%.

The highest value of PCE, i.e., 0.52%, was obtained by our group for the polymer solar cell with the active layer based on PC71BM and polyazomethine with three thiophene rings per unit [25] and for polyazomethine with triphenylamine and p-terphenyl moieties (PCE = 0.56%) [27].

In this work two various HTM layers in perovskite solar cells are analyzed varying the chemical structure of polymer in perovskite devices. Taking into account the thermal and air stability of polyazomethines, we proposed two PAZs: one with three thiophene rings and 3,3’-dimethoxybenzidine moieties (S9) and second one with three thiophene rings and fluorene moieties (S7) investigated as HTMs in perovskite solar cells. Independently, due to the high stability, PAZs were examined as a protection barrier to avoid degradation. Therefore, the aim of this work was to PAZs bifunctionality presentation showing possibility for application in
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