



# Stabilizing Dendron-Modified Talc-Based Electrolyte for Quasi-Solid Dye-Sensitized Solar Cell



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## ABSTRACT

Organic-inorganic layered materials, such as organotals, are a promising alternative as gelling agent for liquid electrolytes in dye-sensitized solar cells. Tals could provide an abundant, low cost and environmentally friendly option for solidifying the electrolyte. This work focuses on generation 5 polyamideamino dendron-modified talc with emphasis is on how it affects the performance and stability of the DSSC. The talc was shown to improve the initial photocurrent by up to 39% by acting as a light scatterer and/or a recombination barrier compared to reference solar cells with liquid electrolyte. Non-destructive analysis based on photographic image technique revealed that during the aging the additive absorbed charge carriers, tri-iodide, from the electrolyte reducing the performance of the solar cells. The degradation could, however, be prevented by intercalating polyiodides into interlamellar space of the talc as the resulting dendron chains did not absorb tri-iodide charge carriers from the electrolyte. These quasi-solid solar cells maintained 95% of their initial efficiency under light-soaking at 1 Sun for about 1000 h. The cells with a quasi-solid electrolyte showed up to 5% higher efficiency than those with liquid electrolyte.

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## 1. INTRODUCTION

Recently the use of clays as additives in dye sensitized solar cell (DSSC) electrolytes has been investigated. These materials act as gelling agents with liquid electrolytes and they are a potential alternative to solve technical problems of conventional DSSCs based on liquid electrolytes such as electrolyte leakage and solvent vaporization [1,2], which limit both the stability and the practical use [3]. In the literature, clays used as gelling agents in the electrolytes of DSSCs have mainly been phyllosilicates such as dioctahedral mica [4], and smectite: laponite [5], bentonite [6], montmorillonite [7,8] and saponite [9].

Gel electrolytes present some issues such as diffusion into the solid matrix, low penetration of the electrolyte into the photo-electrode of nanoparticles of TiO<sub>2</sub>, and an increase in the charge transfer resistance in the photoelectrode/electrolyte interface [10].

However, they have many advantages: they provide longer lifetime of cells over a large temperature range, reduced fluidity compared to liquid electrolyte and light-scattering through clay nanoparticles that can also increase the photocurrent of the cells [11–13]. Moreover, gel electrolytes could possibly be printed which is important in fabrication of large area DSSC modules in an industrial scale [14].

Synthetic clays have distinct characteristics compared with spherical-type inorganic nanoparticles such as SiO<sub>2</sub> [15] or TiO<sub>2</sub>; due to their thin lamellar structure they present a large aspect ratio, which results from the delamination process. In addition, they can show good compatibility with organic electrolytes because their surface is modified with organic molecules [5]. Contrary to frequently referenced clay electrolytes [5,9,16], we use talc (Mg<sub>3</sub>Si<sub>4</sub>O<sub>10</sub>(OH)<sub>2</sub>), which is a 2:1 phyllosilicate without layer charge composed of an octahedral sheet of magnesium oxide-hydroxide sandwiched by two sheets of silica [17] and functionalized with a polyamidoamine dendron, PAMAM [18]. Dendrons are a radial portion of a dendrimer, which are a class of macromolecules with regular and highly branched and symmetric

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three-dimensional structure [19,20]. PAMAM is the most widely studied dendrimer, because of their early discovery and easy synthesis compared to other dendrimers [21]. PAMAMs are hydrophilic macromolecular compounds that consist of amine core, amidoamine as the repeating unit, and primary amine as the terminal groups [22]. The synthesis and characterization of the PAMAM-modified talcs were reported in our previous work [18].

Recently, we studied a sequence of generations of PAMAM-talc (G1 up to G7) as gelators for electrolyte for DSSCs [23]. Polyiodides were intercalated into the organo-modified talc space by adsorption of iodine vapor, producing charge-transfer complexes, PAMAM/I<sub>2</sub>. Our widespread optimization regarding the concentration of the talc, the generation and the effect of iodine absorption showed that in the generation 5 PAMAM-talc with iodine addition resulted in the highest photocurrent and efficiency [23].

Here, the effects of generation 5 PAMAM-talc as an additive in a composite gel electrolyte for DSSCs are investigated in more detail with specific emphasis on the stability, which has not been investigated before. PAMAM-talc materials without intercalated polyiodides appear to adsorb iodide species from the electrolyte solution significantly decreasing the performance [23]. The charge transport dynamics of electrolytes gelled by a sequence of PAMAM dendron generations-modified talc was already published in our previous work [23], where the mechanism of re-reduction of Ru-complex dye was demonstrated, using transient absorption spectroscopy, to be similar to that presented when using gel polymer electrolytes, where dye cation re-reduction is based on the formation of an oxidized dye-iodine complex, which produces the dye and the di-iodine radical [24]. Here, one important aspect is to study how the clay reacts with the electrolyte, in particular with the iodine, upon aging. Changes in the charge carrier

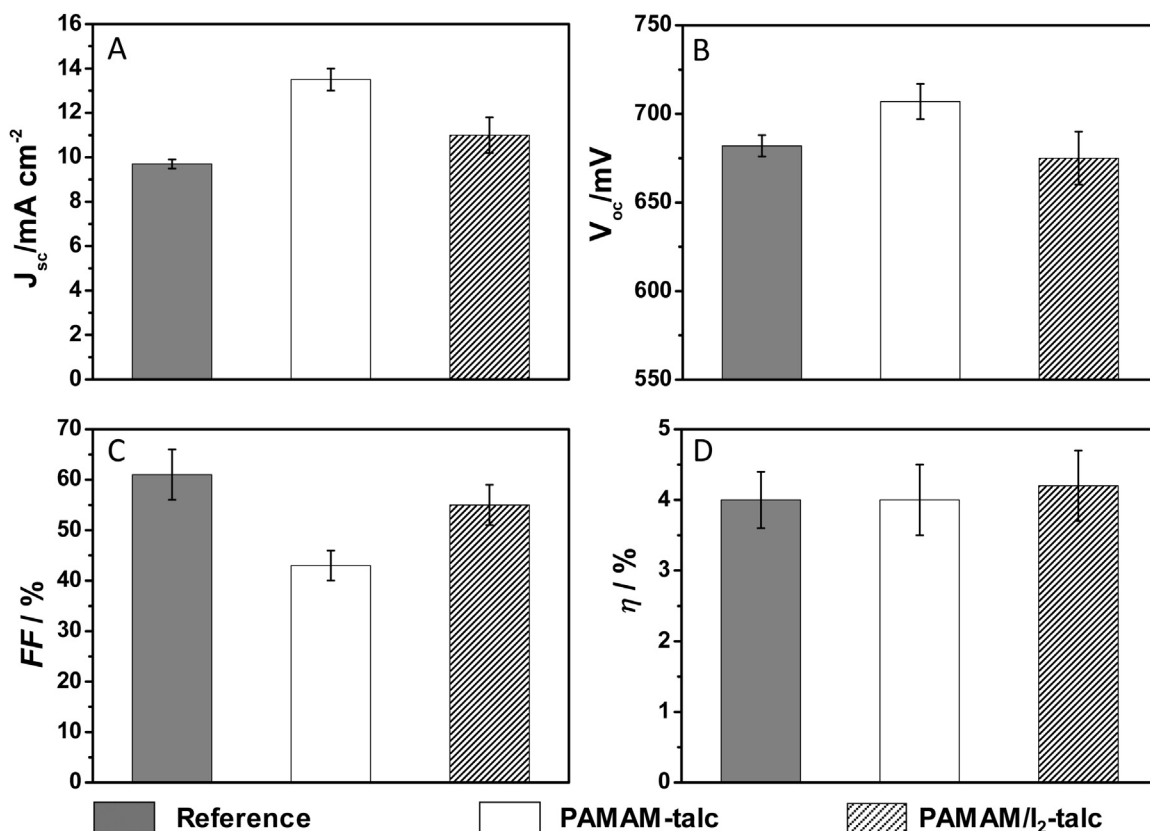
concentration can result in visual changes in the color of the electrolyte. These kind of visual changes can be monitored quantitatively in situ by using image-processing tools as described by Asghar et al. [25,26]. Besides the image analysis, the quasi-solid DSSCs were characterized before and after the ageing through various electrochemical and optical analysis techniques. The purpose is to evaluate the advantages of using a gel electrolyte and the effect of the intercalation of the polyiodides into interlamellar space of the clays on the stability of the devices.

## 2. MATERIALS AND METHODS

### 2.1. Gelation of the Electrolyte and Cell Assembly

Gel electrolytes were prepared by dispersing the organotalcs functionalized with a generation 5 polyamidoamine dendron (PAMAM-talc) and with this material previously intercalated with polyiodides (PAMAM/I<sub>2</sub>-talc) in a liquid electrolyte composed by 0.6 mol L<sup>-1</sup> of 1-propyl-3-imidazolium iodide (PMII, io-li-tec), 0.1 mol L<sup>-1</sup> of iodide (I<sub>2</sub>, Aldrich) and 0.5 mol L<sup>-1</sup> of n-methylbenzimidazole (NMB, Alfa Aesar) in 3-methoxypropionitrile (3-MPN, Sigma Aldrich) [18]. The mixtures were stirred for 16 h at 45 °C and after that; they were homogenized using a probe ultrasound (90 min, 100 W, 2 min pulse on and 1 min pulse off) [23].

Fluorine-doped tin oxide (FTO) glass (TEC-15, Pilkington) was used as substrate for cells. First of all, to prepare photoelectrodes, substrates were treated with a 30 mmol L<sup>-1</sup> aqueous solution of tetrachlorobis(tetrahydrofuran)titanium(IV) (TiCl<sub>4</sub>·2THF, Sigma-Aldrich) at 70 °C for 30 min. After that, two layers of DyeSol DSL 18NR-T TiO<sub>2</sub> paste and one layer of DyeSol 18NR-AO were sequentially screen-printed on top of a glass substrate and sintered



**Fig 1.** The average initial A) short-circuit photocurrent ( $J_{sc}$ ), B) open-circuit voltage ( $V_{oc}$ ), C) fill factor (FF), and D) energy conversion efficiency ( $\eta$ ), values of dye-sensitized solar cells prepared with reference, PAMAM-talc and PAMAM/I<sub>2</sub>-talc electrolytes under 1 Sun. The error bars show the standard deviation of five solar cells for each group.

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