

# A novel deposition method of graphene-based bl-TiO<sub>2</sub> for large-area Perovskite Solar Devices

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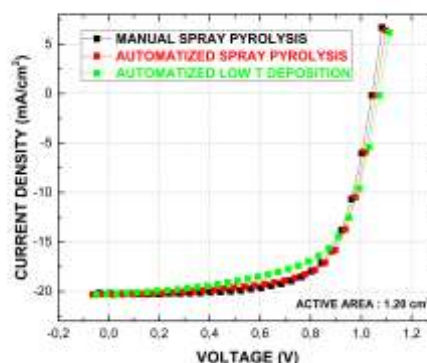
During the last two decades, hybrid organic/inorganic solar cells have been developing in several forms. Among these, perovskite solar cell (PSC) technology is one of the most promising in terms of power conversion efficiency (PCE). Recently, graphene-based materials has started to play a role in solar cells, fuel cells and thermoelectrics [1]. In PSCs, for example, graphene-based materials were employed as an additive for mesoporous TiO<sub>2</sub> (m-TiO<sub>2</sub>) [2]. In the mesoscopic architecture (FTO/bl-TiO<sub>2</sub>/m-TiO<sub>2</sub>/perovskite/spiro-OMeTAD/Au), TiO<sub>2</sub> blocking layer (bl-TiO<sub>2</sub>) can be optimized by adding graphene nanoflakes within the bl-TiO<sub>2</sub> precursor solution as already shown for small area devices, by means of spin coating [3] or spray pyrolysis [4] deposition techniques. The latter represents the usual effective bl-TiO<sub>2</sub> deposition technique, performed by manual airbrush deposition [5]. In order to make PSC technology suitable for industrial production, a repeatable scaled-up process is mandatory. This is achievable only by employing an automatized deposition process: we deposited the graphene-based bl-TiO<sub>2</sub> layer by an automatized spray coating machine onto large area substrates (up to 15x15 cm<sup>2</sup>). By means of automatized spray-pyrolysis (at 460°C) of graphene-based bl-TiO<sub>2</sub> we were able to realize devices (20 samples, each of 1.20 cm<sup>2</sup> active area) with electrical performances equal in terms of PCE = (15.1 ± 0.4) %, to those achieved by manual deposition, demonstrating a very high reproducibility. In order to improve the

scalability of the process in terms of productivity and energy consumption, a low temperature (90°C) automatized deposition process for graphene-based bl-TiO<sub>2</sub> was developed by removing the ACAC (Acetylacetone) from the bl-TiO<sub>2</sub> sprayed solution. In this way, we modified the electron transport layer (ETL) fabrication process by subsequently depositing bl-TiO<sub>2</sub> and m-TiO<sub>2</sub> and then sintering together at 480°C. Even in this case, we obtained performances comparable (PCE exceeding 14.5%) with those of manual and automatized spray pyrolysis. This work, based on a low temperature automated process is, according to our knowledge, a novelty in graphene-based large-area device literature and paves the way for a novel ETL deposition process. Moreover, the low energy consumption process reduces the carbon foot print related to the Life Cycle Assessment (LCA).

## References

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## Figures



**Figure 1:** J-V curves of large-area cells with graphene-based bl-TiO<sub>2</sub> by adopting the three different deposition processes