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## Pouch-sealing as an effective way to fabricate flexible dye-sensitized solar cells and their integration with supercapacitors

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### HIGHLIGHTS

- Vacuum sealing was used for the first time to encapsulate dye-sensitized solar cells.
- Pouch-sealing enabled a flexible integrated energy harvesting and storage system.
- A DSSC and a EDLC were integrated with a simple and scalable encapsulation technique.

### GRAPHICAL ABSTRACT



### ARTICLE INFO

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### ABSTRACT

The scientific interest in integrated energy harvesting and storage (HS) devices has increased exponentially in the last decade since they represent an optimal solution to power portable electronic devices and low consuming Internet of Thing (IoT) sensor nodes. The integration of energy storage devices with photovoltaics can allow to avoid problems such as continuous battery replacement and periodic maintenance, reducing overall costs. In this context, dye sensitized solar cells (DSSCs) integrated with a supercapacitor represent the best choice in terms of lifetime, charge-discharge efficiency, and simplicity of connection avoiding electrical signal conditioning between the two devices. DSSCs have many similarities with supercapacitors, with the only aspect that remains uncovered being the sealing of the device. Herein we propose a common vacuum sealing technology for the integration of a supercapacitor and a DSSC made with shared current collectors, to maximize the integration between the two technologies. The HS device showed a maximum overall photon to electrical conversion and storage efficiency (OPECSE) of 6.10% under only 0.1 SUN illumination, thanks to the high photoconversion efficiency showed by the pouch sealed DSSC, equal to 6.62%. The HS device showed a high stability under bending condition and repeated photo-charge/discharge cycling.

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## 1. Introduction

The most critical issue regarding renewable power generation is related to the uncertainty of source availability. The likely lack of perfect synchronism between electrical energy production and power utilization could render renewable energy plants less appealing with respect to thermoelectric power plants. To address this issue and utilize photo-generated power even when the electromagnetic radiation is unavailable, an energy storage media is necessary [1].

During the last years the number of scientific publications dedicated to combined energy harvesting and storage devices/installations radically increased [2–5]. This growth testifies the interest of the research community in actively promoting this fascinating energy sector of self-sustainable energy harvesting and storage (HS) systems.

Among the different photovoltaic technologies, dye sensitized solar cells (DSSCs) represent a highly appreciable compromise regarding cost, production easiness, efficiency and improved performance under real environment conditions or cloudy circumstances [6,7]. They are still considered the photovoltaic (PV) technology with the lowest pay-back-time [8]. Moreover, even if the record efficiencies obtained for DSSCs are effectively lower than that of other photovoltaic technologies when characterized under standard AM 1.5G illumination condition at 1 SUN intensity, their performances significantly improves when the incoming electromagnetic radiation intensity is lowered, characteristic profusely testified by literature reports [9,10]. Under reduced or dim light conditions DSSCs can achieve an outstanding efficiency of about 35% [11]. These qualities render DSSCs one of the most interesting solutions for implementation in HS devices in particular if wearable and portable application are considered. In this field, HS devices could have a huge impact on the energy market, since nowadays it seems surely a useful item to recharge portable electronic devices or smartphones, but that could have find also applications in other field such as the Internet of Things and smart agriculture [12,13]. In this perspective, an efficient technology available at an accessible price could serve as an appealing incentive for rapid dissemination in the technological energy landscape [14].

A HS device is composed of two sections, and the integration between harvester and storage cells can be performed in different strategies, with the most common approach based on the possibility of sharing a common electrode between the solar cell and the energy storage device [15].

Regarding the energy storage section of HS devices, the two categories that have gathered more interest are batteries and supercapacitors (SCs) [16].

Even though batteries represent a more mature, widely utilized technology and possess a much higher energy density compared to SCs, most scientific integration reports focus on SCs, especially electrical double layer capacitors (EDLCs). This is because EDLCs can withstand an extraordinary long cycle life of approximately  $10^6$  cycles [17–19].

This feature assumes a predominant significance since the number of expected HS charge-discharge cycles could be of several dozens in a single day. With intense activity, battery-like storage section would lead to only one month of uninterrupted operation or less. In contrast, an HS device with an EDLC as energy storage section would last more than 20 years with the same daily number of cycles.

Furthermore, EDLCs can withstand much higher power densities with respect to usual batteries [20]. Since a PV cell or module produces a variable current output depending on the radiation intensity, the incoming power into the storage section is not fixed and is a function of environmental conditions and consequently of time. Because of this, EDLCs are certainly more appropriate for working under these circumstances, since they can accept power under different rate conditions with a marginal difference in the energy stored.

Another important feature that makes EDLCs suitable for integration with a PV harvester is the lower technical specifications required with respect to batteries in terms of voltage. While batteries require being

charged to a specific voltage to let the electrochemical reactions occur, EDLCs do not need a specific voltage since the insertion of cations and anions at the electrode's surfaces does not require a specific voltage value to occur.

The last advantage in using EDLCs instead of batteries in HS integrated devices is that they are intrinsically safer. EDLCs do not involve redox reactions that lead to the formation of dangerous compounds. For example, over-charging a lithium-ion battery may cause the growth of Li metal dendrites or  $O_2$  evolution [21].

When dealing with electrochemical devices like DSSCs and EDLCs, one of the main drawbacks is the evaporation of the liquid electrolyte over time which limits the lifetime of the device. From this perspective, the encapsulation technique becomes of utmost importance and should ensure thermal and chemical stability, be compatible with large scale production, durable against water vapor and oxygen penetration and, in the case of flexible devices, possess mechanical flexibility and reduced weight [22,23]. For energy storage devices such as batteries and EDLCs, a very common encapsulation technique is vacuum sealing in pouch bags, also known as coffee-bags. The pouch usually consists of a laminated material where multiple layers serve to isolate the device from the atmosphere, providing flexibility, mechanical stability, and allowing thermal sealing. Despite being long established for energy storage devices, this technique was never proposed for the encapsulation of DSSC even though it would offer many advantages in terms of stability, ease of fabrication, and integration with energy storage devices. The vacuum sealing would serve the dual function of confining the liquid electrolyte and closely packing the DSSC electrodes, reducing as much as possible the distance ions must travel for dye regeneration. Moreover, when combined with the selection of thermally stable electrolyte such as ionic liquids, vacuum sealing technique was previously demonstrated as suitable encapsulation technique for supercapacitors showing high stability at temperatures as high as  $100\text{ }^\circ\text{C}$  [24]. Therefore, extending this encapsulation technique for DSSC could enable to obtain high thermally stable flexible solar cells. Indeed, it was reported that DSSC can achieve a very good thermal stability at up to  $85\text{ }^\circ\text{C}$  with an accurate choice of building materials, putting special attention on the electrolyte selection, and choosing thermally stable ionic liquids rather than other organic solvents. In this case, it was observed that humidity contamination inside the device represented a more crucial cause of degradation rather than exposure to high temperatures [25,26].

Herein the sealing of a flexible DSSC into a pouch is reported for the first time, and at the same time, the possible direct integration with a supercapacitor, exploiting the same sealing process, is demonstrated. The flexible pouch-DSSC shows remarkable performance both in terms of bending stability and photovoltaic efficiency in low illumination conditions. The integrated HS devices obtained by coupling the pouch-DSSC with a pouch-SC results in the highest combined energy harvesting and storage efficiency ever obtained with these two classes of devices in flexible configuration. The evolution of all the figure of merit of the HS device under bending and low illumination condition have been deeply investigated in order to verify the possible application of this new device for portable and wearable portable power pack.

## 2. Experimental

### 2.1. DSSC fabrication

For the photoanode, a titanium grid (Dexmet Corporation) was coated with a diffusing  $TiO_2$  nanoparticle paste (Solaronix) by means of a doctor blade technique as reported elsewhere [27]. Then the grid was annealed at  $450\text{ }^\circ\text{C}$  for 30 min and then soaked into a dye solution (N719, 0.3 mM in ethanol) for 15 h. Subsequently, the substrate was rinsed with ethanol to remove excess dye not adsorbed on the semiconductor nanostructure. Titanium (Ti) was chosen due to its chemical stability with respect to the  $I^-/I_3^-$  based DSSC electrolyte and because it resulted in better photovoltaic performances compared to other

materials when coupled with TiO<sub>2</sub> as the active material for the DSSC photoanode fabrication [28]. The Ti grids had an open area of 40%, resulting in a lower actual photo-active area of the photoanodes with respect to their total geometric area. The counter electrode was obtained by depositing a thin layer of platinum onto a titanium foil by sputtering technique (Q150T ES, Quorum Technologies Ltd.). The layer thickness was 5 nm, and the deposition time was 10 s. An iodide/triiodide-based electrolyte was prepared with the following composition: 0.45 M NaI, 0.056 M I<sub>2</sub> and 0.55 M 4 tert-butylpyridine dissolved in methoxypropionitrile (MPN). MPN was chosen instead of the commonly used acetonitrile as a low volatile solvent to prevent electrolyte evaporation during the vacuum sealing procedure. A Whatman® glass microfiber filters (grade GF-C) were used as separators between photoanode and counter electrode, and they were soaked with the electrolyte right before sealing the device.

## 2.2. HS device fabrication

The flexible HS device was fabricated integrating a DSSC and SC in the same pouch cell. The layout and a picture of the device are shown in Fig. 1. The device was sealed using standard pouch cell material made of laminated aluminium with a thickness of 120 µm. On the front side, a transparent polyethylene terephthalate (PET) window was created to allow light exposure to the DSSC photoanode. The two materials were sealed using a vacuum sealing tape. A two-electrode device was fabricated by the sharing of the current collectors between the DSSC and the SC.

The SC was fabricated as reported elsewhere [29]. In brief, the electrodes were prepared by depositing a water-based slurry, with composition 85 wt% active carbon (AC), 10 wt% carbon black (CB) and 5% sodium carboxymethyl cellulose (CMC), on an aluminium foil. The electrodes were vacuum dried in a Büchi glass oven for 12 h at 120 °C, cut and stacked with the same fibreglass separator used for the DSSC. Three of the four side of the pouch cell were thermally sealed together. An additional sealing strip was performed transversally between the DSSC and the SC to avoid electrolyte mixing. Finally, the device was vacuum sealed after injecting 100 µL cm<sup>-2</sup> of [BMIM][BF4] for the SC and the same amount for the DSSC electrolyte, as defined before.

## 2.3. Electrical and electrochemical characterization

The DSSC were tested under standard AM1.5G solar spectrum, simulated with a 91195 Newport solar simulator. To examine the behaviour of the DSSC in low-light condition, neutral density filters were used to reduce the incoming power density. A 2440 Keithley source measurement unit (SMU) was used to perform the measurements. Current-voltage measurements were performed at different light intensities ranging from 0.1 to 1 SUN, and also while bending the DSSC

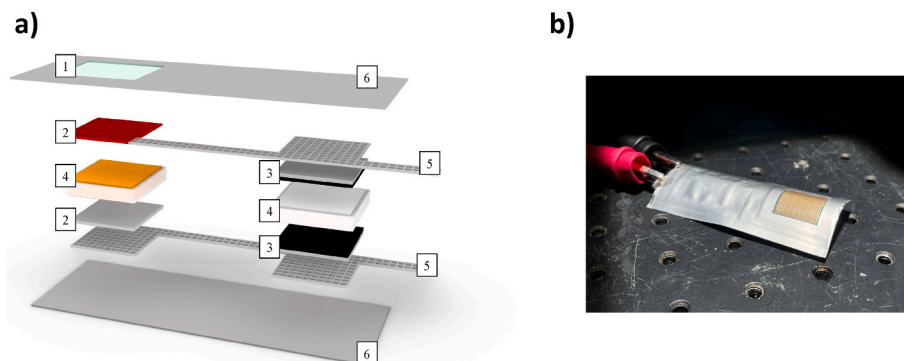
with a curvature radius of 2.5 cm. The performance of the DSSC was tested also at smaller curvature radius, ranging from 0.7 cm to 1.4 cm, under 1 SUN illumination. The performances of the HS device were measured by conducting photo-charge/constant-current discharge tests, from which the overall photon-to-electron conversion and storage efficiency (OPECSE) was calculated using the formula:  $OPECSE = (1/2C \times \Delta V^2) / G \times t \times S$ , where  $C$  is the capacitance of the SC,  $\Delta V$  is the maximum voltage achieved during photo-charge,  $G$  is the electromagnetic power density impinging onto DSSC,  $t$  is the photo-charging time, and  $S$  is active area of the DSSC.

## 3. Results and discussion

The focus of the present manuscript was to report, for the first time, the exploitation of a pouch sealing approach to DSSCs. While this sealing technique is quite common in the field of energy storage devices, there is no evidence of its translation to third-generation photovoltaics. Furthermore, this approach enabled the fabrication of a flexible HS device capable of harvesting and directly storing solar energy, even at low illumination levels and under bending condition.

In Fig. 2a, the photovoltaic response of the pouch sealed flexible DSSC at different illumination levels, in flat and bent condition, is reported. For the test at different illumination values, the bent condition was investigated at a bending radius of 2.5 cm. As can be seen, reducing the light intensity from 1 SUN to 0.1 SUN, the short circuit current density of the solar cells scales down proportionally, as expected. This behaviour is further confirmed by the variation of the ratio between the current density ( $J_{sc}$ ) and the power density of the incident electromagnetic radiation ( $G$ ), reported in Fig. 2c. For the flat condition, the ratio  $J_{sc}/G$  slightly increases at lower illumination levels, confirming the advantage of DSSC with respect to other photovoltaic technology in low-light conditions. This behaviour is mainly due to the lower probability of recombination between photogenerated electrons and holes upon reducing the number of electrons in the conduction band of the TiO<sub>2</sub>, which happens reducing the intensity of the radiation impinging on the solar cell [30]. For the bent condition, this behaviour seems to disappear, and the  $J_{sc}/G$  ratio stays mostly constant.

Here, the angular variation of the incident light with respect to the exposed area of the DSSC, induced by the curvature, likely results in a non-homogeneous exposure, which becomes more evident at lower illumination levels. Nevertheless, a good stability is observed in bent conditions. The same trend is confirmed by the variation of the energy conversion efficiency, as shown in Fig. 2b. For the flat cell, the efficiency rises from 3.35% under 1 SUN (100 mW/cm<sup>2</sup>) and reaches 6.62% at only 0.1 SUN (10 mW/cm<sup>2</sup>) of impinging sunlight. These results are consistent with our previous work on flexible HS, where the need to avoid electrolyte mixing was achieved by confining it in polymeric membranes [31]. The pouch sealing techniques simplify the fabrication of the HS



**Fig. 1.** a) 3D rendering of the device assembly: 1) transparent PET window, 2) DSSC electrodes, 3) SC electrodes, 4) fibreglass separator filled with electrolyte, 5) shared grid current collector, 6) shared pouch sealing foil. b) the HS device under illumination in bent conditions.

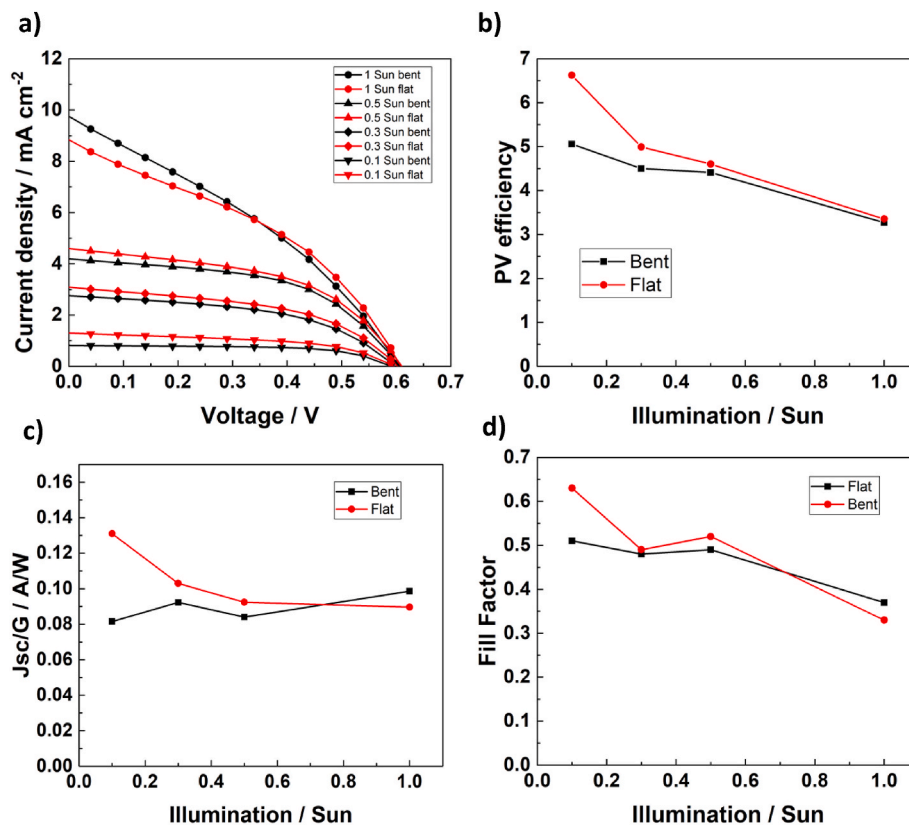


Fig. 2. Flat vs bent characterization results for the flexible DSSC at different illumination conditions. In a) the J-V characteristic. In b) the PV efficiency. In c) the  $J_{sc}/G$  ratio while in d) the cell FF.

device and offers the advantage of avoiding electrolyte evaporation, which is one of the weak points of DSSC [32].

Since the open circuit voltage remains almost constant at different illumination levels, this improvement is mainly related both to the slight increase in the  $J_{sc}/G$  ratio mentioned before and to the increase of the fill factor observed when the incident radiation becomes weaker (Fig. 2d). The same behaviour is observed in bending conditions, in which the efficiency of the DSSC rise from 3.27% to 5.05% when the incident power goes from 100 mW/cm<sup>2</sup> to 10 mW/cm<sup>2</sup>, showing a noteworthy resilience to mechanical deformation with only a slight reduction of the main photovoltaic parameters. Nevertheless, to further investigate the behaviour of the proposed pouch sealed flexible DSSC, additional tests at smaller bending radii (from 1.4 cm to 0.7 cm) were performed under 1 SUN illumination and the relative variation of the photovoltaic performances is reported in Fig. S1. It can be seen that the effect of the non-homogeneous illumination due to the curvature of the DSSC observed before, becomes a crucial factor strongly affecting the DSSC performance. Indeed, at very low bending radius, the DSSC surface it is not uniformly lightened by the simulated solar light that impinges on the device along the normal direction. Therefore, the lateral part of the solar cell will receive a much lower amount of light with respect to the flat configuration, reducing the photogenerated current. Nevertheless, the overall behaviour of the DSSC remains stable, as demonstrated by the almost absent variation of the FF and of the  $V_{oc}$  of the cell. Indeed, the relative reduction in conversion efficiency is only dependent on the lower photogenerated current density, which in turn depend on the non-homogeneous illumination, as commented.

Once the possibility of employing the pouch sealing technique for DSSCs was demonstrated, the same method was used to fabricate the integrated HS device, and its performance was evaluated. Harvesting-storage integration was investigated during the photo-charging and subsequent constant current discharge phases. The terminals of the

source measure unit were connected to the electrodes of the HS device to record the photo-charging potential. During photo-charge, the HS device was placed in the solar simulator under 1 SUN illumination condition. The charge step was stopped at 0.6 V since an obvious limiting plateau due to the DSSC  $V_{oc}$  was reached. For the discharge phase, the sun simulator was turned off, and a negative current was applied to the device. The process was repeated at five different discharge current values, between 1 mA and 20 mA. The results are reported in Fig. 3a-b. Under 1 SUN illumination, the HS device was charged up to the  $V_{oc}$  of the DSSM in 35 s and reached a maximum OPECSE of 3.33% at a photo-charging potential of 0.43 V. Notably, a remarkable 95% capacity retention was observed when the discharging current rise from 1 mA to 20 mA, with capacity values of 0.058mAh and 0.055mAh, respectively (Fig. 3b). To test the cyclability of the HS, repeated photo-charge/discharge measurements were performed using the same procedure mentioned before. During photo-charge, the device was irradiated with 1 SUN illumination while, during discharge a constant current of 10 mA was applied. The results are reported in Fig. 3c-d. It is clear how no evidence of performance drop is observed, showing an outstanding cycling stability after 50 cycles.

Similar to the single DSSC unit, the HS flexible device was also tested under different illumination conditions. Fig. 4a shows the photo-charge/discharge profile obtained with the same impinging power densities previously investigated for DSSC alone. It is evident that at lower illumination levels, the charging time increases as expected, since the photogenerated current decreases. Moreover, reducing the light intensity to 0.1 SUN, a plateau in the charging potential appears, and the goal voltage of 0.6 V cannot be achieved. This was expected since at 0.1 SUN, the  $V_{oc}$  of the DSSC reduces to values slightly lower than 0.6 V, and the photo-charging phase was stopped at 0.598 V. The photo-charging phase took 10 min but 91% of the maximum charging voltage was reached after only 2 min (Fig. 4a). During the rest of the

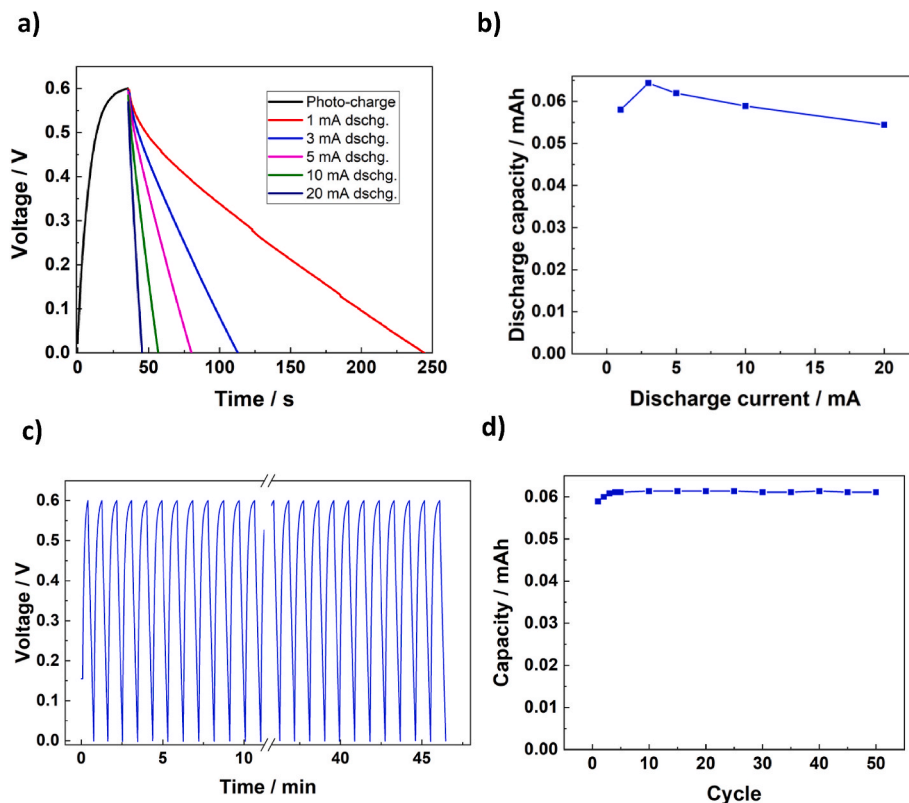


Fig. 3. a) The photo-charge and discharge profiles of the HS module. In b), the discharge capacities at different discharge currents. c) 1 SUN –10 mA photo-charge/discharge repeated for 50 cycles. d) Discharge capacity with respect to discharge cycle.

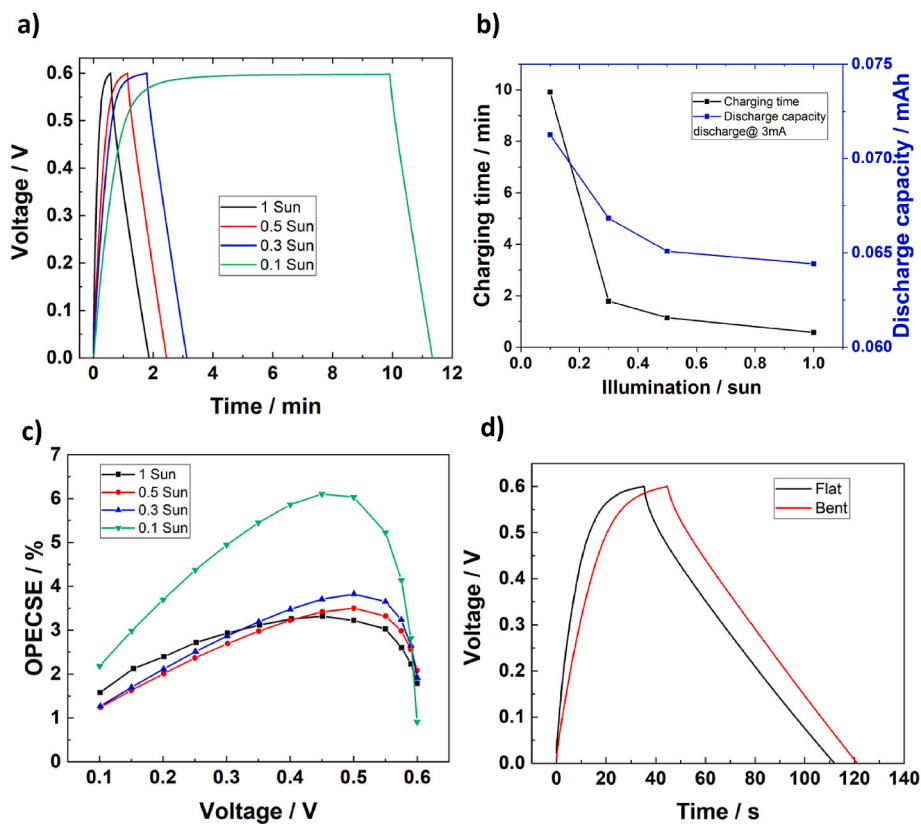


Fig. 4. a) Photo-charge/discharge measurements performed at different illumination levels. Discharge curves were measured at 3 mA. b) Variation of photo-charging time and discharge capacity at different illumination levels. c) Overall photon-to-electron conversion and storage efficiency versus charging voltage at different illumination levels. d) Comparison between photo-charge/discharge measurements performed in flat and bent conditions.

photo-charging process, despite the small current generated by the DSSC the HS voltage slightly increase. This is ascribed to a quasi-equilibrium establishment between the PV and storage devices. Such a steady state can be ascribed to the equilibration of the PV current near the OCV of the PV itself and the leakage current of the SC side. From a practical point of view, in this steady state the PV contributes to a better consolidation of the stored charge. Indeed, in this direct configuration the PV module is behaving as a constant current and constant voltage power source. That is, before the 95% of the goal voltage, the PV generates an almost constant current and the SC charges hence the HS module voltage varies linearly. During the rest 5% goal voltage variation, the PV current drops according to J-V characteristic (Fig. 2a) and reaches those values comparable with the leakage currents of the SC at constant voltage floating periods. Hence, at the different illumination conditions the charging power is tuned as in a galvanostatic rate capability test. It is known that during such experiments, SC experience capacitance fading due to dissipations. Because of these reasons, related to charges distribution mainly at the porous electrodes interface, with the increasing charging power it less likely to reach the steady state current densities at constant voltages. In other words, ions diffusion within pores represents the limiting step with this point of view if the SC works in his safe voltage window. This behaviour can be confirmed by the results reported in Fig. 4b, where the extracted discharge capacity at different illumination condition is reported. Reducing the illumination intensity, the photo-charging time increases. Slower charging rate allows a more uniform electrodes charging hence the discharge capacity increases at fixed goal voltage. A 10% increase in discharge capacity is observed reducing the illumination level from 1 SUN to 0.1 SUN.

The OPECSE plotted as a function of the voltage achieved by the SC at the end of the charge step (Fig. 4c) allow to evaluate the trade-off between the final potential and overall efficiency of the integrated device, which is fundamental for practical application of integrated HS devices. The profile of the OPECSE remains consistent for all the illumination conditions, and from this profile, the optimum voltage at which the photo-charging process should be stopped can be determined. However, as discussed previously, the excess solar energy can be exploited to maintain the SC state of charge. In Fig. 4d, the maximum OPECSE and the corresponding charging voltage, at different illumination conditions, are reported. The OPECSE increases from 3.33% to 6.10% when reducing the light intensity from 1 SUN to 0.1 SUN, respectively. It can be observed, that for all the illumination conditions, the maximum overall efficiency value is reached at more than 0.45 V which is 25% less than the open circuit volage of the DSSC. The goal for a HS device should be to increase this value as much as possible. Nevertheless, for the 0.1 SUN case, it is remarkable that the OPECSE still maintains an appreciable value higher than 5% for a voltage of 0.55 V, which is only 8% less than the plateau voltage reached at this illumination level.

Finally, in Fig. 4d, the comparison between the photo-charge-discharge process performed in flat and bent conditions, at bending radius of 2.5 cm, is reported, showing only a slight increase of the charging time when all the HS device has been subjected to bending, mainly due to the lower current photogenerated by the DSSC in this condition, as observed before. As performed previously for DSSC, also in this case smaller bending radius were evaluated. The relative reduction in the photo-charge/discharge performances is reported in Fig. S2. In line with the different currents photogenerated by the solar cells at different bending radii, the photo-charging time increase at lower bending radii, affecting the value of the overall efficiency of the integrated device (OPECSE). Nevertheless, it is clear how this variation is only dependent on the difference and non-uniformity of the illumination on the solar cell surfaces, rather than device instabilities caused by the flexible configuration. Indeed, the discharge capacity remains constant demonstrating how it is possible to consistently extract the solar energy that was stored in the supercapacitor, even in bent conditions.

## 4. Conclusions

In conclusion, an integrated energy harvesting and storage device coupling a DSSC and a supercapacitor sharing the current collectors and sealed with a single packaging strategy is proposed. The pouch sealing commonly exploited for batteries and supercapacitors is here transferred to the photoelectrochemical solar cell, allowing a higher level of the integration concerning the fabrication process of integrated devices. The fabricated pouch sealed DSSC show a conversion efficiency of 3.35% under 1 SUN illumination condition and it increases its performance while the intensity of the impinging light is reduced, reaching a maximum efficiency of 6.62% under 0.1 SUN illumination, showing the suitability of this photovoltaic technology for real life application where diffused and low light illumination condition are frequent. The integrated HS device was able to reach a maximum OPECSE of 3.33% at 1 SUN illumination and showed to retain the compatibility of the DSSC with low light illumination, reaching a maximum OPECSE of 6.10% when the light intensity was reduced to 0.1 SUN. At all illumination condition, it was possible to photo-charge and discharge the HS device up to the open circuit voltage of the DSSC and, despite the charging time increased at lower illumination condition, it was observed that it may lead to a more uniform electrode charging in the supercapacitor and a consequent higher discharge capacity. Moreover, the high stability of the pouch sealing method was showed both for the DSSC and for the integrated device. Both devices were tested under flat and bent condition, showing only a slight decrease in photovoltaic parameters, for the DSSC, and in charging time for the HS device. Finally, no performance drop was observed performing cyclic photo-charge/discharge test of the integrated HS device.

The new approach to seal together the harvesting and storage section can be extended to other third generation photovoltaics and other energy storage devices such as rechargeable batteries opening the way to new generation of portable self-rechargeable power pack.

## CRedit authorship contribution statement

**Roberto Speranza:** Methodology, Investigation, Writing – original draft, Writing – review & editing, Visualization. **Pietro Zaccagnini:** Methodology, Investigation, Writing – review & editing. **Alberto Scalia:** Methodology, Investigation. **Elena Tresso:** Conceptualization, Supervision, Project administration, Funding acquisition. **Andrea Lamberti:** Conceptualization, Validation, Writing – review & editing, Supervision, Project administration, Funding acquisition.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jpowsour.2023.233581>.

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