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## Stability study of thermal cycling on organic solar cells

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### Abstract

We present a side-by-side comparison of the stability of three different types of benchmark solution processed organic solar cells (OSC), namely polymer:fullerene, small molecule: fullerene and polymer:non-fullerene solar cells, subject to thermal cycling stress conditions. Upon 5 complete thermal cycles between -100 °C and 80 °C, we find that all the device types investigated exhibit superior stability, albeit with a distinct temperature dependence of device efficiency. The good thermal cycling stability of the devices studied is consistent with the highly flexible nature of the organic photoactive materials used in OSC. Our results suggest that OSC, especially those based on **plastic** substrates, could be a promising candidate for applications with large variations and rapid change in operating temperature such as outer space applications. Also, there is substantial difference in the efficiency drops from high to low temperature for different OSC blend systems. The distinct temperature dependence of device performance suggests that maintaining optimum performance with minimal variations with operating temperature is a key challenge to address for such PV applications.

Keywords: organic solar cells, stability, thermal cycling, space.

### Introduction

Solution processed organic solar cells (OSC) are attracting great interest as a low cost and versatile alternative to traditional inorganic based such as silicon and III-V semiconductor photovoltaic (PV) technologies, with their power conversion efficiencies (*PCE*) now achieving the threshold for commercial viability (>10%). A key driving force for this improvement has been the recent advances in material design, leading to a range of high performance photoactive materials. For example, the recent development of a range of polymer and small molecule electron donor/acceptor materials has led to the design of various types of novel high efficiency OSC.<sup>1,2</sup> OSC based on small molecule donors have already achieved comparable *PCE* to their polymer-based counterparts.<sup>3,4</sup> Another breakthrough has been the establishment of a class of non-fullerene small molecule electron acceptor materials, leading to a record *PCE* of over 13% for organic solar cells, already exceeding that of conventional OSC typically based on fullerenes.<sup>5,6</sup>

Nevertheless, commercialisation of solution processed OSC has been limited by their relatively low environmental stability.<sup>7,8</sup> Under standard operating environments, unencapsulated OSC typically undergo rapid device degradation, losing their performance within days to months.<sup>9</sup> This limitation has imposed further challenges in device encapsulation, which results in significant process limitations, higher costs and only partially effective process solutions. While numerous environmental parameters (e.g. light, heat, oxygen and humidity) have been identified as stress factors for the degradation of OSC, the detailed degradation mechanisms, especially their links to materials and device design, still remain widely unclear. Understanding the root cause of degradation is therefore a main research focus of the OSC community, attracting wide research interests with established stability study procedures.

The efficiency of OSC is strongly dependent on the donor:accepter blend morphology, typically on a nanoscale, in order to simultaneously achieve optimised charge generation and collection. Morphology stability, particularly under thermal stress, is therefore a key requirement for the outdoor application of OSC, since devices operating under standard conditions can reach a temperature of ~65°C. Recently the crystallisation of [6,6]-Phenyl C<sub>61</sub> butyric acid methyl ester (PCBM) throughout the blend film has been proposed as a primary cause for thermally-induced degradation of fullerene-based OSC. Under thermal stress conditions, fullerene can diffuse within the polymer matrix, and nucleate/crystallise to form nano-to micron-scale crystallites.<sup>10,11</sup> We and others have correlated the formation of such crystallites to a loss in device performance during thermal stress, and developed a number of approaches (e.g. use of materials with a high glass transition temperature, chemical crosslinking and light induced fullerene dimerisation) to improve the thermal stability of OSC.<sup>12-14</sup> Recently, a number of studies have reported superior thermal stability of OSC based on a number of non-fullerene acceptor materials.<sup>15,16</sup>

While the majority of thermal stability studies have focused on the degradation of the blend morphology due to thermal stress, relatively less research efforts have been dedicated to the degradation behaviour of OSC subjected to thermal cycling stress conditions in which the temperature changes in a short period of time and possibly causes physical damage to the devices. Such information is not only relevant to the real-world outdoor applications (accelerated day/night alternations), but also outer space applications. While a limited number of studies have investigated the potential of OSC for outer space applications by studying their performance under AMO illumination,<sup>17</sup> the stability of OSC under rapidly-changing, wide temperature-range thermal cycling environments, another critical consideration for out space applications, is still lacking.

In this work, we report the initial investigation of the potential use of solution processed OSC in environment of large temperature variations and/or rapid change in operating temperature. In particular, we perform a direct, side-by-side comparison of the stability of three established types of OSC systems, namely polymer:fullerene, small molecule donor:fullerene and polymer:non-fullerene, subject to 5 thermal cycles between -100 °C and 80 °C. We found that all the devices investigated exhibit great durability under thermal cycling stress even when rigid glass substrates and metal electrodes were used, which is likely linked to the highly flexible nature of the photoactive layers in the device. Our findings

support suggestions that solution processed OSC could be suitable for outer space applications.

## Experimental

### Materials

ITO glass ( $15 \Omega/\square$ ) substrates were purchased from Luminescence Technology. Poly[N-9'-heptadecanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)] (PCDTBT), benzodithiophene terthiophene rhodamine (BTR), Poly[(2,6-(4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)-benzo[1,2-b:4,5-b']dithiophene))-alt-(5,5-(1',3'-di-2-thienyl-5',7'-bis(2-ethylhexyl)benzo[1',2'-c:4',5'-c']dithiophene-4,8-dione))] (PBDB-T), and 3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)-indanone))-5,5,11,11-tetrakis(4-hexylphenyl)-dithieno[2,3-d:2',3'-d']-s-indaceno[1,2-b:5,6-b']dithiophene (ITIC) were purchased from 1-Material. [6,6]-phenyl-C<sub>71</sub>-butyric acid methyl ester (PC<sub>71</sub>BM) was purchased from Solenne BV. Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) (CLEVIOS P VP.AI 4083) was purchased from Heraeus. Polyethylenimine, 80% ethoxylated solution (PEIE) was purchased from Sigma Aldrich. All the materials are used as received.

### Fabrication of OSC

All devices are fabricated in conventional structure: Indium tin oxide (ITO)/PEDOT:PSS (30 nm)/PCDTBT:PC<sub>71</sub>BM (70 nm) or BTR:PC<sub>71</sub>BM (220 nm) or PBDB-T:ITIC (100 nm)/PEIE/Ag. ITO glass substrates were sequentially cleaned with detergent, deionized water, acetone, and 2-propanol in ultrasonic bath. PEDOT:PSS was spin-coated onto dried substrates treated with oxygen plasma for 150 s. All active layer solutions were dissolved and stirred on a hot plate in a nitrogen filled glovebox for at least 12 hours before the fabrication. PCDTBT:PC<sub>71</sub>BM (18 mg/ml with 1:2 weight ratio in chlorobenzene), BTR:PC<sub>71</sub>BM (40 mg/ml with 1:1 weight ratio in chloroform), and PBDB-T:ITIC (20 mg/ml with 1:1 weight ratio in chlorobenzene) were spin-coated onto the PEDOT:PSS coated substrates. After spin-coating, a solvent vapour annealing treatment using tetrahydrofuran was performed on BTR:PC<sub>71</sub>BM samples for 2 minutes; thermal annealing on a 100 °C hotplate was performed on PBDB-T:ITIC samples for 10 minutes. Diluted PEIE solution (9.45 µl PEIE diluted with 3.87 ml ethanol with presence of MgSO<sub>4</sub> powder for at least 12 hours) was first filtered with 0.45 µm PVDF syringe filter and then spin-coated onto the active layer at 2000 rpm for 20 s. The samples were finally transferred to a separated evaporator for silver deposition at a base pressure of  $2 \times 10^{-5}$  Torr. All the devices have active area of 0.15 cm<sup>2</sup> and are not encapsulated for the measurement.

### J-V characterization and thermal cycling

The devices were loaded in a temperature controlled probe stage (Linkam LTSE420-P) under a constant nitrogen flow. J-V characteristics of the devices were measured by a Keithley 2400 sourcemeter under simulated AM1.5G solar illumination (Newport 92193A-1000) through a quartz window on the stage. The temperature was precisely controlled between -100 °C and 80 °C with a rate of 50 °C/min. Once reaching the target temperature, we allowed 1 minute for thermal equilibrium between the device and the stage. scanned the J-V of the devices were then scanned right after the equilibrium at 0, 2.5, and 5 minutes.

## Results and discussion

We selected three established benchmark OSC systems, namely PCDTBT:PC<sub>71</sub>BM, BTR:PC<sub>71</sub>BM, and PBDB-T:ITIC blends respectively, for detailed study. PCDTBT:PC<sub>71</sub>BM is a widely studied benchmark polymer:fullerene system with over 7 years of operating lifetime reported.<sup>18</sup> BTR:PC<sub>71</sub>BM represents a new generation of small molecule donor-based OPV system with comparable efficiency to polymer-based solar cells.<sup>3</sup> PBDB-T:ITIC is a newly established, high performance polymer:non-fullerene blend system with comparable efficiency to their fullerene-based counterpart.<sup>15</sup> These three types of OSC represent the state-of-the-art material and device designs of solution processed OSC. Solution processed PEDOT:PSS and PEIE were used as hole transport layers and electron transport layers respectively. All devices were fabricated with the same device architecture for ease of comparison and the chemical structures of the materials used for device fabrication is illustrated in Fig. 1.

Fig. 2 shows the evolution of device parameters subject to 5 thermal cycles between -100 °C and 80 °C in nitrogen atmosphere. It is obvious that all the devices investigated show a distinct change in all device parameters upon switching between low and high temperatures, resulting in a significant change in overall device performance. This large temperature dependence is consistent with a strong temperature dependence of the charge carrier kinetics, especially the charge collection efficiency, consistent with previous reports.<sup>19,20</sup> Interestingly, the PBDB-T:ITIC system has the strongest temperature dependence of device efficiency, with a drop of over 80% in PCE from 80 °C to -100 °C, albeit with a smallest change in open circuit voltage ( $V_{oc}$ ). In contrast, the BTR:PC<sub>71</sub>BM system exhibits the least drop of ca. 55% at a low operating temperature (Fig. 2a). The origin of the different temperature dependencies in different blend systems requires further studies, but is likely linked to the fundamental differences in charge carrier kinetics in different OPV blend systems. Nevertheless, a good overall stability upon temperature switching is seen for all the devices investigated, evidenced by a minimal change in device performance upon a complete low-to-high temperature cycle. A more pronounced change in device performance is seen in the PBDB-T:ITIC system upon constant thermal stress at 80 °C, an indication of post thermal annealing effect and thermal degradation.

Fig. 3 shows the comparison of the J-V characteristics of the OSC blend systems measured at different temperatures and before/after thermal cycling. Devices using PCDTBT:PC<sub>71</sub>BM and BTR:PC<sub>71</sub>BM show the best device stability upon thermal cycling, with minimal change in device performance (Fig. 3a and 3b). This is due to superior device stability both under constant thermal stress and upon thermal cycling. The PBDB-T:ITIC system shows more pronounced degradation, mainly attributed to poorer device stability under constant thermal stress at 80 °C (Fig. 3c). It is obvious that the BTR:PC<sub>71</sub>BM system exhibits less temperature dependence of both  $J_{sc}$  and  $FF$  than the other systems investigated, leading to the smallest variation in PCE during the thermal cycling conditions. This is in agreement with our previous report that the BTR:PC<sub>71</sub>BM system possesses a low level of trap density, hence significantly reducing the non-geminate charge recombination losses.<sup>21</sup> All the device parameters are summarized in Table 1.

BTR:PC<sub>71</sub>BM is selected for a more intense thermal cycling study as apparently it is the most stable system upon thermal cycling as shown above. The device was tested for more thermal cycles of up to 50 cycles with the same temperature range from -100 °C to 80 °C. The device performance was measured under one sun at room temperature right after every 5 thermal cycles. As shown in Fig. 2e, there is only a little drop after the first 5 thermal cycles which is consistent with the data above. However, after that, all the device parameters behave very similarly even it has experienced up to 50 thermal cycles and all the device parameters retain more than 98 % of the original values. The corresponding J-V data after 0, 5, 25 and 50 thermal cycles are selected from Fig. 2e and plotted in Fig. 3d showing only minimal changes on their J-V characteristics.

Environmental stability has been widely recognized as a critical barrier for the commercialization of solution processed OSC. As a key consideration for outdoor applications, the degradation of device performance of OSC due to thermal stress has been widely investigated in terms of changes in blend morphology, and various approaches have been developed to address this issue. A separate challenge that has received less investigation is the durability of the device to alternated temperature cycles. This is particularly relevant to the outdoor operation of devices, since the day/night shifts typically cause a step change in operating temperatures that varies with locations and target applications, and a thermally stable device may not necessarily have good stability under such stress conditions. It should be noted that in order to achieve superior device stability under such applications, a good device thermal stability under constant elevated and reduced environmental temperatures, in conjunction with good device durability to thermal cycling stress conditions, is necessary. Therefore, the work presented herein represents new insights into the device stability under thermal cycling stress conditions. For devices whose degradation is controlled by thermal cycling stress rather than continuous thermal stress, thermal cycling measurements can be used as an accelerated method to evaluate their thermal stability.

It should be noted the device stability upon thermal cycling is most likely a measure of the robustness of device mechanical properties. Our work shows that OSC generally have very good stability upon thermal cycling, even for glass-based, lab-scale devices. For devices based on plastic substrates, it is expected that the device stability upon thermal cycling will be further improved due to the fact that the thermal expansion coefficient of polymers (for both the materials for substrates and photoactive layers) are similar ranged between  $\sim 20 \times 10^{-6} \text{ K}^{-1}$  and  $\sim 100 \times 10^{-6} \text{ K}^{-1}$ , [ref 1, 2] whereas the thermal expansion coefficient of glass ranges between  $1 \times 10^{-6} \text{ K}^{-1}$  and  $10 \times 10^{-6} \text{ K}^{-1}$  a similar thermal expansion coefficient to that of polymers<sup>22</sup>, [ref 3], whereas it is known that the different thermal expansion coefficients of glass and polymers can cause lesser difference of the coefficient between materials could reduce the chance of causing cracks or delamination which is the real challenge in the OPV operation of OSCs upon thermal cycling.<sup>23</sup> [ref 4] This Our results indicate that OSCs can have superior thermal cycling stability, in excellent agreement with the findings of OSCAR project, a recent real-life stratospheric study for both organic and perovskite solar cells led by Manca et al.<sup>24</sup> Combining the key advantages of the solution processed solar cells such as superior power to weight ratio and lower costs, our findings suggest that solution processed solar cells based on flexible substrates, such as solution processed organic, perovskite and

quantum dots solar cells, may be a suitable candidate for applications with large variations and rapid change in operating temperature such as outer space applications.

While we find that OSC typically exhibit good durability under thermal cycling conditions, it should be noted that the large variations in device performance for all device types studied under low and high temperatures may represent a challenge for more extreme outer space applications such as distant planet exploration, with, for example, a good overall device efficiency across a broad temperature range being desirable for such applications. While our previous work has suggested that the molecular design of a trap-free PV system may be the key in minimizing the variation in device performance at different temperatures, more work is needed to better understand the impacts of cell temperatures on their performance, especially on a charge carrier kinetics level. Undoubtedly, a systematic study of the thermal cycling stability of different class of solution processed solar cells as a function of their material and device design, as well as a mechanistic study to understand their degradation mechanisms under thermal cycling stress, is urgently needed. Nevertheless, our work suggests that OSC could be a promising candidate for outer space applications, thereby paving the way for more detailed investigations in this direction.

## Conclusion

We present a side-by-side comparison of the device stability of three established types of OSC subject to thermal cycling stress conditions. We found that all the devices investigated exhibit very good stability under thermal cycling stress between  $-100^{\circ}\text{C}$  and  $80^{\circ}\text{C}$ , and attribute this to the highly flexible nature of OSC. Our findings suggest that solution processed OSC can be a promising candidate for outer space applications.

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## Figure and table captions

Fig. 1 (a) OSC device structure and (b) Chemical structures of the active materials investigated in this study.

Fig. 2 Normalized device parameters, (a)  $J_{SC}$ , (b)  $V_{OC}$ , (c)  $FF$ , and (d)  $PCE$ , of PCDTBT:PC<sub>71</sub>BM, BTR:PC<sub>71</sub>BM, and PBDB-T:ITIC as a function of time during thermal cycling between  $-100^{\circ}\text{C}$  and  $80^{\circ}\text{C}$  for 5 cycles. (e) Device parameters of BTR:PC<sub>71</sub>BM devices as a function of time measured right after every 5 thermal cycles at  $25^{\circ}\text{C}$  for 50 thermal cycles in total. The temperature changing rate for all measurements is  $50^{\circ}\text{C}/\text{min}$ .

Fig. 3  $J$ - $V$  characteristics of (a) PCDTBT:PC<sub>71</sub>BM, (b) BTR:PC<sub>71</sub>BM, and (c) PBDB-T:ITIC at room temperature (RT),  $-100^{\circ}\text{C}$ ,  $80^{\circ}\text{C}$ , and RT after 5 cycles. (d)  $J$ - $V$  characteristics of BTR:PC<sub>71</sub>BM at RT after different number of thermal cycles (corresponding to Fig. 2e).

Table 1 Summary of device parameters of PCDTBT:PC<sub>71</sub>BM, BTR:PC<sub>71</sub>BM, and PBDB-T:ITIC at room temperature (RT), -100 °C, 80 °C, and RT after 5 cycles.

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