In-situ investigation of perovskite solar cells efficiency and stability in mimic stratospheric environment for high-altitude pseudo satellites

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ABSTRACT

Perovskite solar cells with high power-per-weight have great potential to be used for aerospace applications such as satellites or high-altitude pseudo-satellites. These latter are unmanned aircrafts exclusively powered by solar energy, typically flying in the stratosphere where conditions of pressure, temperature and illumination are critically different than on earth surface. In this work, we evaluate the performance and stability of high efficiency perovskite solar cells under mimic

stratospheric environment. In-situ measurements at controlled conditions of pressure, temperature and illumination were developed. We show that the cells can operate efficiently in a large range of temperatures from -50°C to +20°C, with a maximum power conversion efficiency at -20°C, which is ideal for use in the stratosphere. Besides, performances are maintained after a number of temperature cycles down to -85°C, representative of temperature variations due to diurnal cycles. An efficient encapsulation is developed, which could be critical to avoid the accelerated degradation of the cells under vacuum. Finally, a promising stability for 25 days of day-night cycles was demonstrated, which suggests that perovskite solar cells could be used to power high altitude pseudo-satellites.

Introduction

Perovskite solar cells (PSCs) represent a major breakthrough in the field of photovoltaics (PV) due to their rapid progress in power conversion efficiency (PCE) and potentially low manufacturing costs[1]. One key advantage of PSCs is that they can be fully printed at low temperature on light-weight flexible substrates[2][3][4]. Recently, PSCs with power-per-weight as high as 23 $W.g^{-1}$ has been demonstrated by using ultra-thin 1.4-µm-thick PET foils as a substrate[5]. This high specific power (power-per-weight) makes perovskite PV very attractive to be used in future aerospace applications such as High-Altitude Pseudo-Satellite (HAPS). HAPS are unmanned airship, plane or balloon flying in the stratosphere, operating like satellites but closer to the earth. Target applications include search and rescue missions, disaster relief, environmental monitoring and agriculture[6]. HAPS are powered exclusively by solar energy and are able to continuously fly for months. Solar panels integrated onto the wings and body directly power the aircraft engines and instrumentation, while secondary batteries charged in daylight power overnight flight.

It is well known that perovskite solar cells have intrinsic material instability at high temperature[7] and are vulnerable to environmental conditions such as UV light[8], humidity[9] and oxygen[10]. Perovskite degradation mechanisms on earth surface have already been extensively investigated, which resulted in significant improvements in stability of perovskite solar devices in the last few years[11]. More recently, these systems have also demonstrated excellent radiation hardness to high-energy particles making them promising for space applications[12][13].

Here, we specifically address the stability of high efficiency triple cation perovskite solar cells for HAPS. HAPS such as Airbus Zephyr typically operate in the stratosphere, above clouds and conventional air traffic. In this environment, moisture degradation can be mitigated as the stratosphere is extremely dry. However, other environmental conditions specifically found at these altitudes could be detrimental to PSCs and reduce their lifetime. The cells temperature during the day can vary between -20°C to +10°C on average, while the temperature at night can be as low as -85°C. Besides, the pressure in the stratosphere varies between 1 and 250 mbar, much lower than the average sea-level pressure of 1013.25 mbar. However, the impact of vacuum on PSCs performance and stability has been mostly unexplored. Then, the solar spectrum at high altitude is analogous to AM0 spectrum found in outer space, approximated by the 5800 K blackbody (1349 $W.m^{-2}$ at an altitude of 32 km vs. 1366.1 $W.m^{-2}$ for AM0)[14]. AM0 solar spectrum contains a

significantly higher level of UV radiations, potentially harmful to the cell's performance. In 2018, stratospheric mission OSCAR studied organic-based solar cell operation in extra-terrestrial conditions by launching a weather balloon with organic and perovskite solar cells in the stratosphere for 3 h[14][15]. The perovskite cells underwent quite a severe drop from 14.1% to 9.3% PCE after the 3 h flight. However, the cells didn't use state-of-the-art materials (MAPbI$_3$ was used as an active layer and TiO$_2$ as an electron transport layer). Besides, the flight duration was rather short and didn't allow for day/night cycles measurement. The authors stressed that prolonged operation could pose significant problems to the devices, however the scarcity of data and intrinsic limitations of this type of real time monitoring do not provide this information.

In this work, by using in-situ measurements in controlled environment, we were able to determine the stability and suitability of high-efficiency PSCs in stratospheric conditions. PSCs were first characterized under AM0 spectrum, i.e. without parasitic absorption from the earth's atmosphere. Then, the performance of PSCs submitted to temperature variations typically found at high altitude under illumination and in the dark were measured. The effect of vacuum was also investigated, and a simple and efficient encapsulation strategy was demonstrated. Finally, the dynamics of degradation with day-night cycles over the course of 25 days was evaluated. We show that PSCs could power HAPS for days without major degradation in cells performance.

Materials and methods

1.1 Perovskite solar cell fabrication and testing

15 Ω.□$^{-1}$ ITO/glass substrates (Lumtec) were sequentially cleaned with detergent in DI water, acetone, and isopropyl alcohol in an ultrasonic bath. Subsequently, they were dried with N$_2$ and exposed to O$_2$ plasma for 10 min. A planar layer of SnO$_2$ (15% colloidal dispersion in H$_2$O, Alfa Aesar, diluted in deionized water at 1:6.5 volume ratio) with thickness of ~25 nm was subsequently deposited via spin coating at a spin speed of 3000 rpm and an acceleration 3000 rpm for 30 s. This was followed by sintering the substrates at 150°C for 30 min. A triple cation perovskite films were deposited in a N$_2$ atmosphere using single-step deposition method from the precursor solution containing FAI (172 mg) (Dyesol), PbI$_2$ (507 mg) (TCI), MABr (22 mg) (Dyesol) and PbBr$_2$ (73 mg) (TCI) in anhydrous N,N-Dimethylformamide (99.8%, Sigma-Aldrich)/dimethylsulphoxide (99.7%, Sigma-Aldrich) (8:2 (v:v)). Thereafter, 53 µL of CsI (99.999% trace metal, Sigma-Aldrich), (390 mg, 1 ml DMSO) was added to the precursor solution. The precursor solution was spin-coated onto the planar SnO$_2$ films in a two-step program at 1000 and 6000 rpm. for 10 and 20 s, respectively. During the second step, 300 µl of chlorobenzene (99.9%) was dropped on the spinning substrate 5 s prior to the end of the program. This was followed by annealing the films at 100°C for 1 hour. To complete the fabrication of devices, 2,2',7,7'-tetrakis(*N*,*N*-di-*p*-methoxyphenylamine)-9,9-spirobifluorene (Spiro-OMeTAD, 90 mg in chlorobenzene) as a hole-transporting material (HTM) was deposited by spin coating 100 µL of the prepared solution at 4000 rpm for 20 s. The Spiro-OMeTAD (Sigma-Aldrich) was doped with

Bis(trifluoromethane)sulfonimide lithium salt (99.95%, Sigma-Aldrich) dissolved in acetonitrile (520 mg/ml), Tris(2-(1H-pyrazol-1-yl)-4-tert-butylpyridine)-cobalt(III) Tris(bis(trifluoromethylsulfonyl)imide) (FK 209, from Dyenamo) and 4-tert-Butylpyridine (96%, Sigma-Aldrich) with concentrations of 34 μL, 10 μL and 19 μL respectively. Finally, device fabrication was completed by thermally evaporating gold wire (99.9% 1mm, Kurt J.Lesker) to form a ~70 nm gold layer as a back contact.

Perovskite solar cells were characterized under mimic HAPS environment, as described in figure S1. The cells were kept in a measurement chamber (Linkam Scientific) which allowed to control the temperature (from -196°C to 350°C) and pressure (down to 10^{-3} mbar). Typically, the cells were measured at 10 mbar and 0°C. The chamber was positioned below a solar simulator using a 450 W xenon lamp (Sol3A Class AAA Solar Simulator, Oriel, USA). The current-voltage (J-V) characteristics of the perovskite devices were recorded with a digital source meter (Keithley model 2400, USA). Both forward and reverse bias scans were taken from 1.2 to -0.1 V with a sweep interval of 0.015 V, resulting in 81 data points respectively. The Air Mass Zero J-V characteristics follow the same experiment set-up and measurement parameters as AM1.5G. However, the AM1.5G filter was replaced with an AM0 filter. Alongside this replacement, the lamp was calibrated by integrating the measured EQE J_{sc} and matching the pixel J_{sc} under the AM0 spectrum with this value. EQE spectra were obtained by QEX10 Quantum Efficiency Measurement System.

For long-term measurements (>10 h) with day-night cycles, PSCs were positioned below a LEDs matrix. The intensity of the LEDs was adjusted to one-sun equivalent so that the initial J_{sc} of the devices measured in the chamber were matched with the J_{sc} measured under one-sun illumination. The LEDs were programmed to stay on for 16 h and off for 8 h. For these measurements, the cells were exposed to air at 1 bar and kept at ☐20°C. A source-meter unit was used to measure forward and reverse J-V curves every 15 min. The cells were kept at open circuit between measurements.

Results and Discussion

1. Performance of perovskite solar cells under AM1.5G and AM0 illumination

In this work, PSCs with active areas of 0.15 cm^2 and structure glass/ITO/SnO$_2$/Cs$_{0.05}$(MA$_{0.17}$FA$_{0.83}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$/Spiro-OMeTAD/Au were entirely fabricated at low temperature (<150°C). SnO$_2$ nanoparticles were used as an electron transport layer (ETL) instead of TiO$_2$ as they can be printed and dried at low temperature, and were shown to be more transparent and resistant to UV radiations as compared to TiO$_2$ ETL[8]. Besides, a triple-cation perovskite based on composition Cs$_{0.05}$(MA$_{0.17}$FA$_{0.83}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$ was used for its better thermal stability and higher overall PCE as compared to MAPbI$_3$[16]. The external quantum efficiency (EQE) for this cell is shown in figure 1(a), along with the AM1.5G and AM0 solar spectra. The response in UV-blue region is excellent with a sharp EQE edge at 300 nm. The transmittance spectrum of glass/ITO substrates is also shown. The transmittance edge overlaps

almost perfectly the EQE edge, which indicates that the EQE in the UV region is only limited by ITO absorption, without any parasitic absorption in the SnO_2 ETL. This is particularly important for PSCs operating under AM0 illumination as spectral irradiance in the UV range is significantly higher than under AM1.5G illumination. This allows to harness more sunlight and increase overall efficiency as compared to less transparent TiO_2 ETL[17].

From the EQE spectrum, the J_{sc} of the cell under AM1.5G and AM0 illumination was calculated by integrating the area under the curve. Our solar simulator was then fitted with an AM0 filter and the lamp intensity was calibrated with the EQE-calculated AM0 J_{sc} (as described in [18]). The AM1.5G and AM0 current-voltage curves measured at room temperature are shown in figure 1(b). The champion cell yielded an AM1.5G PCE of 18.4% and AM0 PCE of 16.9%. The efficiency difference under AM1.5G and AM0 illumination is expected for this perovskite composition. Indeed, for a single junction cell with a bandgap of □1.6 eV, the AM1.5G efficiency theoretical limit is □30.5%, while the AM0 limit is □28%[19]. The theoretical ratio AM1.5G PCE/AM0 PCE = 1.09 is in perfect agreement with our results. Despite the lower AM0 PCE, the higher irradiance of AM0 spectrum in the UV and visible range allow for an increased output power, with a maximum power density of 18.4 mW.cm^{-2} and 22.8 mW.cm^{-2} under AM1.5G and AM0 illumination, respectively.
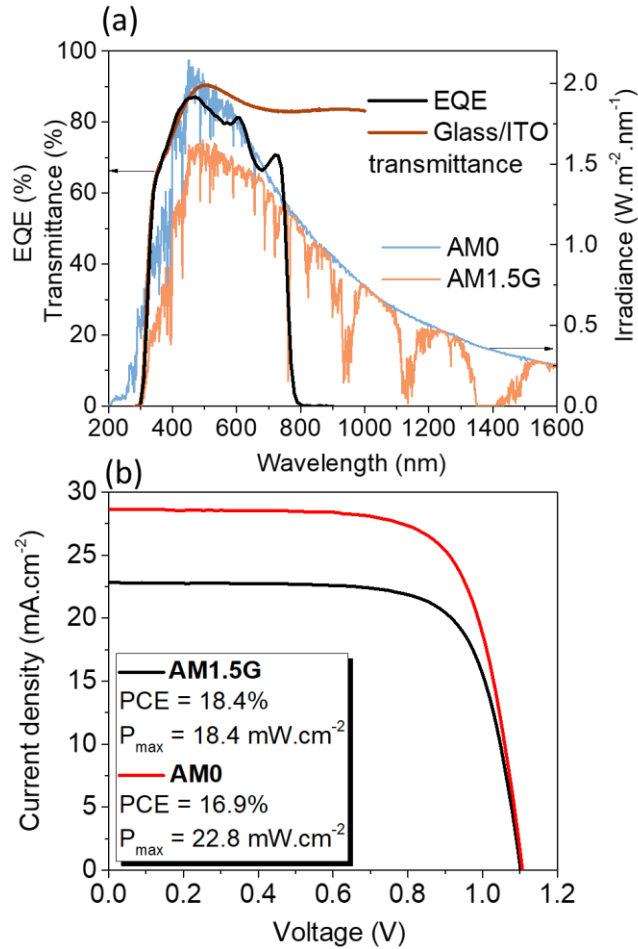
Figure 1. (a) External quantum efficiency of perovskite solar cell based on structure glass/ITO/SnO$_2$/Cs$_{0.05}$(MA$_{0.17}$FA$_{0.83}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$/Spiro-OMeTAD/Au and transmittance of glass/ITO substrates. Air mass 1.5 global (AM1.5G) and air mass zero (AM0) solar spectral irradiance are shown in red and blue lines, respectively. (b) Current-voltage curves for perovskite solar cells measured under AM1.5G and AM0 illumination.

2. Temperature variations

Usually, PSCs performance and behavior are routinely investigated at room temperature or at high temperature to mimic operating conditions in harsh environments such as desert regions[7]. However, for HAPS the temperature range is critically different. Once HAPS have reached the stratosphere and are flying steadily, they are exposed to temperature variations as illustrated in figure S2. At night, the cells temperature can drop down to -86°C, but during the day the cells temperature can reach 30°C in the extreme case. On average, the cells temperature during the day is between -20°C and +10°C. Besides, during their ascent they must cross the tropopause at □10 km, the second coldest local minimum in Earth's atmosphere with an air temperature of -50°C. It is known that temperature can have significant effects on the crystal structure, phase transition and degradation of perovskite, as well as electronic properties and stability of interlayers. Several authors have studied the effect of temperature variations on the performance of PSCs using

methylammonium lead iodide (MAPbI$_3$) as an active layer[20][21][22]. They have come to a consensus that the cells perform best at room temperature, while the performance drop at higher or lower temperatures, which was explained by limitations in the transport and extraction of charge carriers. Triple cation perovskite compounds such as Cs$_{0.05}$(MA$_{0.17}$FA$_{0.83}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$ have shown dramatically different optical and structural properties than MAPbI$_3$ at low and high temperatures, such as the absence of phase transition from orthorombic to tetragonal phase at low temperature. However, to our knowledge there is no report of the effect of temperature on the performance of full PSCs based on triple cation perovskite.

The PV parameters for PSCs with Cs$_{0.05}$(MA$_{0.17}$FA$_{0.83}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$ active layer measured between +20°C and -70°C under AM0 illumination and vacuum (10 mbar) are shown in figure 2 (The PV parameters and the statistical data under AM1.5G are available in Figure S3 and Figure S4). These conditions of temperature, light and pressure are representative of stratospheric environment. The fill factor (FF) stays relatively constant from 20°C to -40°C, but then rapidly decreases down to -70°C. By looking into the changes in the series resistance (R$_S$) and the shunt resistance (R$_{Sh}$) at different temperature (see Figure S5), we found that there is a considerable increase in the R$_S$ when the temperature goes below c.a. -30°C which correlates well with the rapid drop in the FF. The change in FF is probably due to changes of charge carrier dynamic at lower operating temperature. In particular, it could be decreases in mobility of the charge extraction layers. The V$_{oc}$ increases with decreasing temperatures, from 1.13 V at 20°C to almost 1.17 V at -40°C, which is in good agreement with other works and can be explained by the reduction of traps density in the perovskite bulk or at interfaces at low temperatures[23]. Then, the V$_{oc}$ slightly decreases below -40°C. This is consistent with the variations in perovskite bandgap, which is rather constant from +20°C to -40°C but then continuously decreases at temperatures below -40°C, as measured by Hetterich *et al.* for triple cation perovskite[24]. However, we note that this change is minor and the V$_{oc}$ remains higher at -70°C (1.15 V) than at room temperature (1.13 V). Finally, the J$_{sc}$ is rather constant over the whole temperature range, implying that the perovskite does not undergo a phase transition down to -70°C[24]. Overall, the PCE slightly increases at temperatures below 20°C, reaching a local maximum of 18.2% AM0 PCE at -20°C, before decreasing because of the drop in FF at lower temperature. Hence, differently to MAPbI$_3$, the triple cation perovskites with composition Cs$_{0.05}$(MA$_{0.17}$FA$_{0.83}$)$_{0.95}$Pb(I$_{0.83}$Br$_{0.17}$)$_3$ perform better at temperatures between 0°C and -40°C than at room temperature. This is mainly because the fill factor stays constant in this range of temperature, whereas in the case of MAPbI$_3$ the FF starts to fall as soon as the temperature is decreased below 20°C[20][21][22].

At -50°C the cells still have a high PCE of 17.2%, which indicates that PSCs can operate very efficiently in the tropopause. Besides, at -70°C and -60°C, PSCs are still operating with relatively high PCEs of 13.2% and 15.7%, respectively. This is particularly important as the stratospheric air temperature at dawn and dusk can be much lower than at noon, as shown in figure S2. It means that PSCs can harvest solar energy during the entire day, from dawn to dusk, which is particularly important to mitigate the use of HAPS batteries. These measurements reveal that perovskite solar

cells with a triple cation perovskite active layer can have very high PCE in the stratosphere, which range of temperature is ideal for maximizing PSCs performance. It also suggests that record PCEs achieved in the literature with triple cation perovskites, usually measured at 25°C, could be even higher at low temperatures. However, in the case of optimized devices with passivated interfaces and lower trap density, the improvement in PCE at low temperature could be reduced.
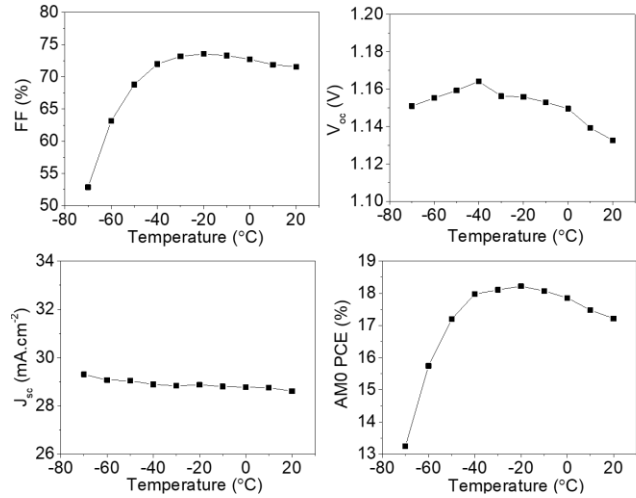

Figure 2. PV parameters (fill factor, V_{oc}, J_{sc} and PCE) of perovskite solar cells measured between -70°C and +20°C under constant AM0 illumination and vacuum (10 mbar).

Then, PSCs were submitted to thermal cycles between 20°C and -85°C in the dark to simulate the temperature variations during day and night, and evaluate the effect on the mechanical and physical properties of the cells. A perovskite solar cell stack has multiple interfaces that could lead to mechanical failure during temperature cycling because of the different thermal expansion coefficients of the embedded layers[25]. J-V curves were measured at 20°C under AM1.5G illumination before and after 1, 6, 11 and 16 temperature cycles. To rule out the effect of light, the cells were kept in the dark during the temperature ramps (20°C.min^{-1}). In figure 3, it is observed that PSCs retained 100% of their initial performance after 16 temperature cycles. We note that these cells were measured under vacuum at 10 mbar and with an encapsulation composed of polyimide/epoxy/glass (as described later). This means that the encapsulation materials used here also enable mechanical stability against these large temperature variations. McGehee *et al.* demonstrated that an encapsulant with a low elastic modulus enables to dissipate strain more efficiently and results in much higher mechanical stability[25]. Similarly, it is thought that the polyimide foil used here between the perovskite cell and the epoxy is able to release strain during these temperature variations and enables high operational stability of the cell. Hence, polyimide could be used as a simple technique to avoid fractures and delamination at the interface between the cell and the encapsulant.
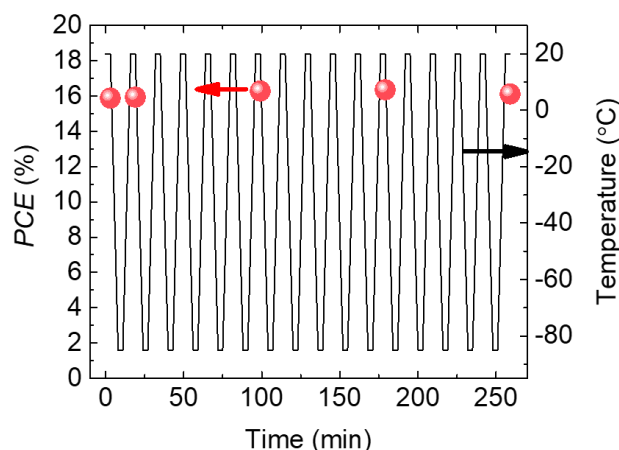
Figure 3. AM1.5G Power conversion efficiency (PCE) of perovskite solar cell after 1, 6, 11 and 16 thermal cycles between 20°C and -85°C in the dark at 10 mbar. The red symbols indicate the PCE measured at 20°C and the black line indicates the temperature.

3. Effect of low pressure

Next, we investigate the effect of vacuum on the performance and stability of the cells. In 2018, Q. Bao *et al.* have shown that triple cation PSCs undergo severe degradation under white light in ultrahigh vacuum (UHV) environment[26]. It was suggested that the MA cation decomposes into gaseous NH_3 and HX (X = halogen, I, Br), with the formation of the highly volatile component CH_3NH_3X as the intermediate step of the degradation, which are pumped away in UHV conditions, eventually causing permanent degradation of the cell. These measurements were only performed on perovskite films directly exposed to vacuum, without interlayers, metallic electrode, nor encapsulation, at base pressure of 1 x 10^{-8} torr ($\Box$1.3 x 10^{-8} mbar). Hence, gaseous products could easily and quickly be lost outside the film. However, these degradation processes may be kinematically limited in a full device stack or in a well-sealed device when the volatile components in the reaction are not allowed to escape as easily as in vacuum.

To address this issue, we developed a simple encapsulation method based on a polyimide/epoxy/glass stack. A drawing of the cell's stack and encapsulation is shown in figure 4(a). It was noticed that the active materials undergo a chemical reaction with the epoxy, wherever the polyimide foil was not applied and the perovskite became transparent after a few days in dark/N_2, as seen in figure 4(b). However, the area covered with polyimide remained dark, suggesting that the perovskite was preserved. It demonstrates that polyimide can be used as a simple barrier layer to avoid any chemical reaction with the epoxy glue. Using this encapsulation method, we were able to store PSCs in dark air ($\Box$20°C, $\Box$30% relative humidity) for > 6 months without any loss of performance.

To evaluate the photostability of cells under vacuum, encapsulated and non-encapsulated cells were exposed to 1 sun of AM0 illumination at mild vacuum of 10 mbar (specific of HAPS environment) and controlled temperature of 0°C. (The AM1.5G version is available in the Figure S6) In figure 4(c), the AM0 PCE of the encapsulated cell drops from 18.3% to 15% after 5 h of illumination. However, when the lamp is turned off the cell quickly recovers to ☐95% of its initial PCE after ☐1 h in the dark, before degrading again under illumination, with similar kinetics as the first illumination cycle. Quite surprisingly, the non-encapsulated cell shows a similar behavior, with similar degradation rate under AM0 illumination and recovery during the dark period. The only difference is that the V_{oc} seems to randomly jump from ☐1.13 V to ☐1.16 V (see Figure S2) for the non-encapsulated sample, which is not clearly understood but cannot be directly attributed to the absence of encapsulation. We think that the gold electrode itself (and potentially the Spiro layer) can act as an encapsulation, to some extent, and prevent the loss of volatile degradation products from the cell stack during the first hours of illumination. Once the light is turned off, these degradation products can react back to re-form the active perovskite phase, which explains the recovery of the cell in the dark. This "self-healing" mechanism is only possible because no compound is lost outside the device stack. However, for longer stability studies, the vacuum could accelerate the permanent degradation of non-encapsulated PSCs as the gold contact cannot provide an efficient sealing, which makes the use of an efficient encapsulation like the one described here necessary. At this stage, it was not possible to perform long-term measurements (> 10 h) under vacuum and controlled temperature to directly address the effect of encapsulation on the long-term stability of the cells under vacuum, which will be the scope of future work.
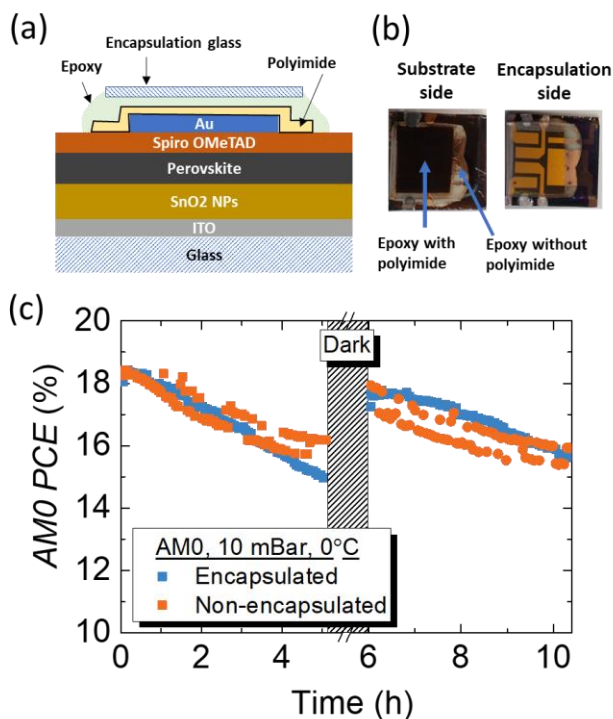
Figure 4. (a) Schematic of perovskite solar cell architecture with polyimide/epoxy/glass encapsulation. (b) Pictures of perovskite cell viewed from substrate and encapsulation side after storage in dark/N_2. (c) Stability of encapsulated and non-encapsulated devices at 10 mbar and 25°C under AM1.5G illumination.

4. Day/night cycles and long-term performance

In stratosphere, HAPS experience natural sunlight's diurnal cycle similar to earth surface, with periods of 24 h (periods can be much shorter for satellites). Only a few works have studied the long-term stability of perovskite solar cells over day/night phases. During the day, ie. under illumination, ionic defects migration induced by electric field is known to deteriorate the device performances after only several hours[27]. However, during the night, full recovery of efficiency can be observed for cells at early degradation stages (< 20% loss in PCE) [28]. This behavior can be explained by a cyclic ion movement mechanism : under illumination, ionic species migrate toward the electrode interfaces where they accumulate. These reactive species have been found to rupture the crystal structure of perovskite materials and adjacent functional layers, potentially resulting in serious degradation of device performance [29]. However, the crystal structure can be fully or partially recovered in the dark when ionic species move back to the perovskite bulk and fill the vacancies created during the day. For more advanced degradation stage under illumination (>20% loss), the initial PCE could not be fully recovered in the dark and in this case, a slow and permanent degradation is observed after several day-night cycles [28]. This is explained by non-reversible degradation processes which also occur because of ion migration (such as electrode degradation when iodide ions react with Ag), and potentially other degradation factors.

To measure the long-term stability of PSCs in conditions as close to real HAPS operation in stratosphere, we subjected PSCs to day/night cycles, where the cells were continuously soaked with 1 sun of LEDs lighting for 16 h and then kept in the dark for 8 h. The performances were monitored over the course of 25 days by acquiring J-V scans every hour as shown in figure 5. A typical PCE dynamics of "degradation during the day – recovery during the night", referred as "type I" diurnal behavior by Katz *et al.* can be observed[28]. The cells PCE drops relatively quickly under illumination for the first 5 h, before stabilizing at around 80% of the initial PCE. On the contrary to Katz *et al.* who showed that the decrease of the PSC parameters during the day was only partially compensated for by nighttime recovery, eventually leading to a PCE reduced by more than 50% after 8 days, our PSCs show a different behavior and retain close to 80% of their initial efficiency even after 25 days. This could be ascribed to the faster recovery process in the dark (60 to 90 min). Hence, the dark duration (8 h) is sufficient for the cell to recover entirely before the PCE drops again under illumination. If we consider that the initial decay is due to reversible mechanisms such as ion migration and might not account for the long-term degradation of the device, we can introduce the "stabilized" PCE defined as the PCE after 12 h of illumination (see inset of figure 5(a)). In this case, the device retains 95% of its "stabilized" efficiency after 25 days. It demonstrates that PSCs could be used in stratospheric environment for days, without major loss in performance due to sunlight diurnal cycles. It is however unknown how UV light would affect the materials properties since PSCs are measured under UV-free LED lighting during these

day-night cycles. However, recent measurements have suggested that UV-A (360-380 nm band) don't induce any performance degradation when using SnO_2 as an ETL[8]. Besides, any potential UV-induced degradation could be addressed relatively easily by using a UV filter or down-conversion materials.
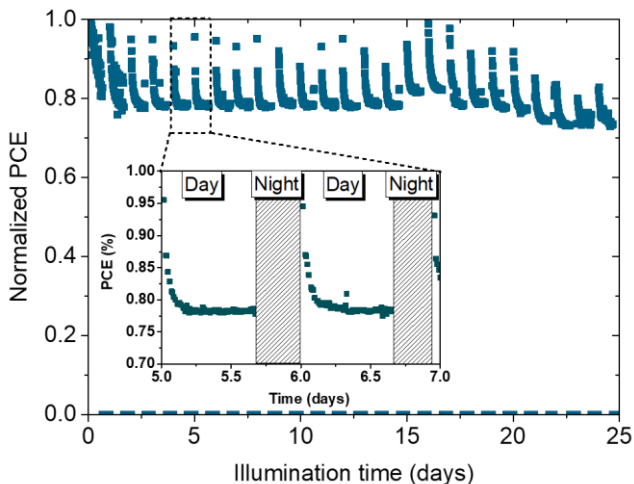

Figure 5. Normalized power conversion efficiency obtained during days/nights cycles (16 h illumination/8 h dark) of perovskite solar cells under 1 sun of LEDs illumination (atm. pressure, ☐20°C) for 25 days.

Conclusions

In summary, the performance and stability of perovskite solar cells used in stratospheric environment to power HAPS or unmanned aerial vehicles have been evaluated by in-situ studies in a mimic stratospheric environment. This work shows that the PSCs based on a triple cation perovskite can operate in a large range of temperature from -70°C to +20°C, with a maximum PCE at -20°C, which is ideal to operate in HAPS environment. Besides, the performances are maintained after temperature cycles down to -85°C, which can be reached during nighttime. A simple encapsulation strategy based on polyimide/epoxy/glass stack has been developed to mitigate the photodegradation of the cells under vacuum and to allow for the recovery of efficiency in the dark. The kinetics of degradation during day-night cycles has been monitored and indicates that the cells retain ☐80% of their initial efficiency after 25 days. The present results demonstrate that PSCs have great potential to be used for high-altitude pseudo-satellites. The next step toward this goal will be to achieve light-weight PSCs with high specific power and high stability in the harsh stratospheric environment. Besides, these results demonstrate that PSCs could perform remarkably well in polar regions or during wintertime in temperate regions of the earth, where the temperature can drop well below zero degree Celsius.

Conflicts of interest

There are no conflicts to declare.

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Supporting Information
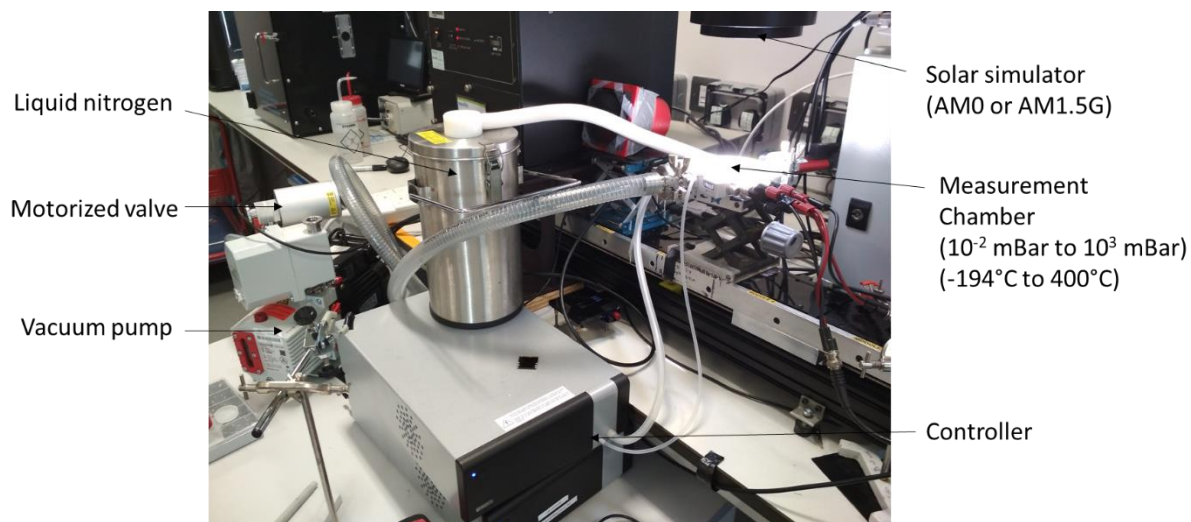
Figure S1. Set-up used for current-voltage measurements of perovskite solar cells under mimic HAPS environment.
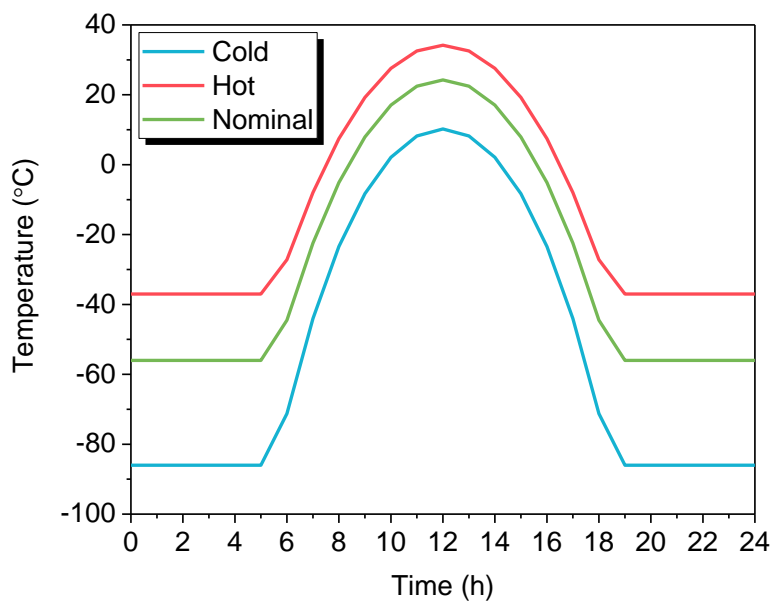

Figure S2. Solar cells temperature throughout the day in stratosphere calculated at summer solstice, -15° latitude in southern hemisphere. Values of -86°C (cold), -56°C (nominal) and -37°C (hot) were taken as assumptions for the outside air temperature at night. The cold and hot temperatures represent the extreme cases found in stratosphere.
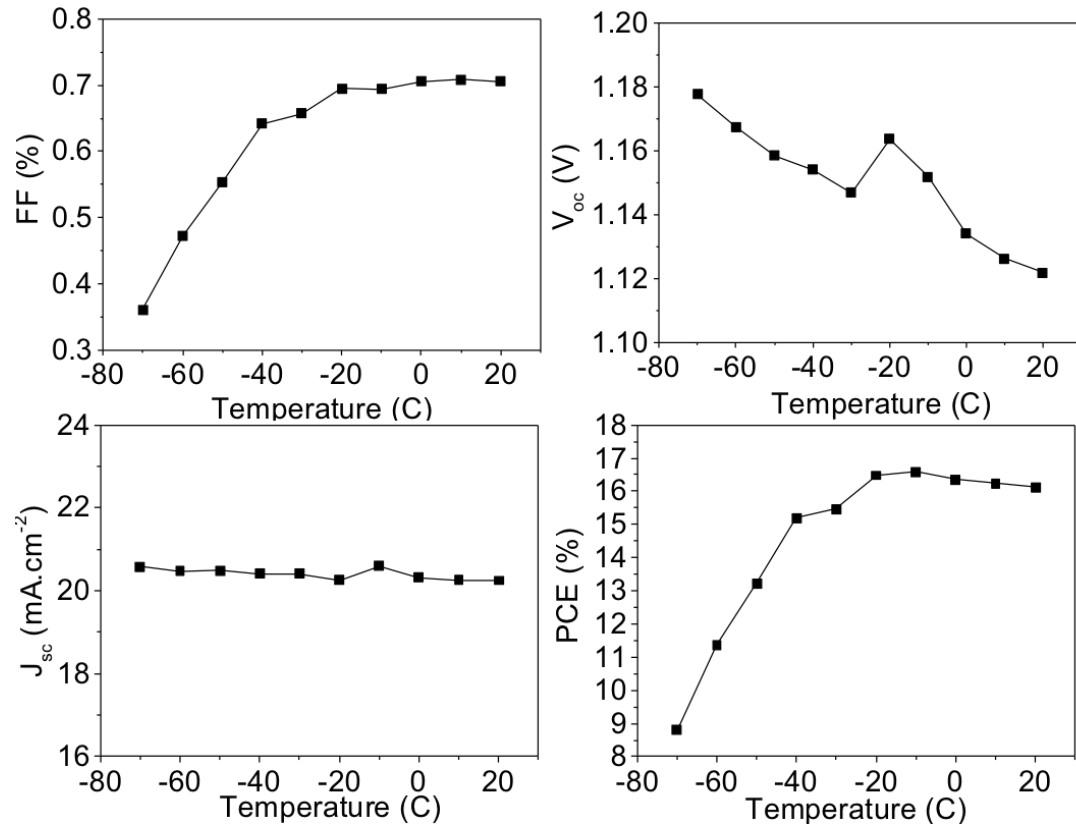
Figure S3. PV parameters (fill factor, V_{oc}, J_{sc} and PCE) of perovskite solar cells measured between -70°C and +20°C under constant AM1.5G illumination and vacuum (10 mbar).
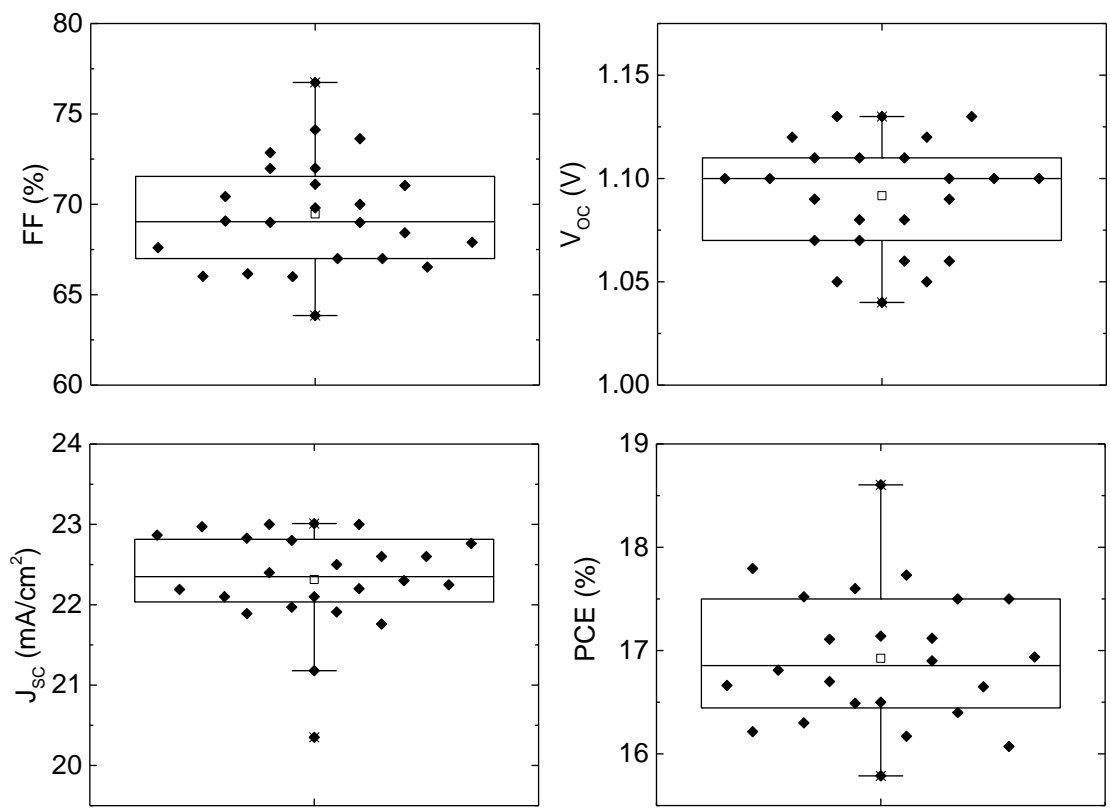
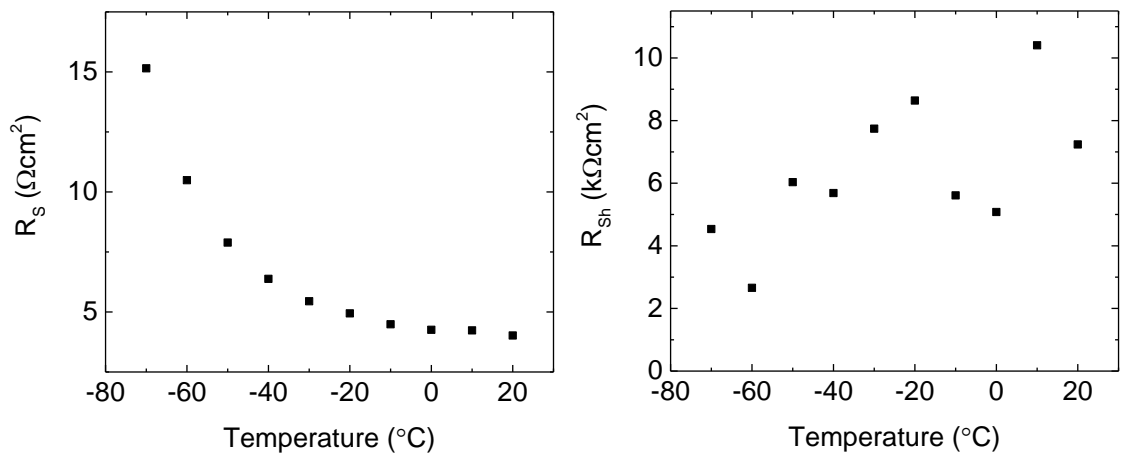**Figure S4.** Statistical result of the device parameters under AM1.5G.

Figure S5. Series resistance (R_S) and shunt resistance (R_{Sh}) at different temperatures extracted from the inverses of the slopes at V_{OC} and J_{SC}, respectively.
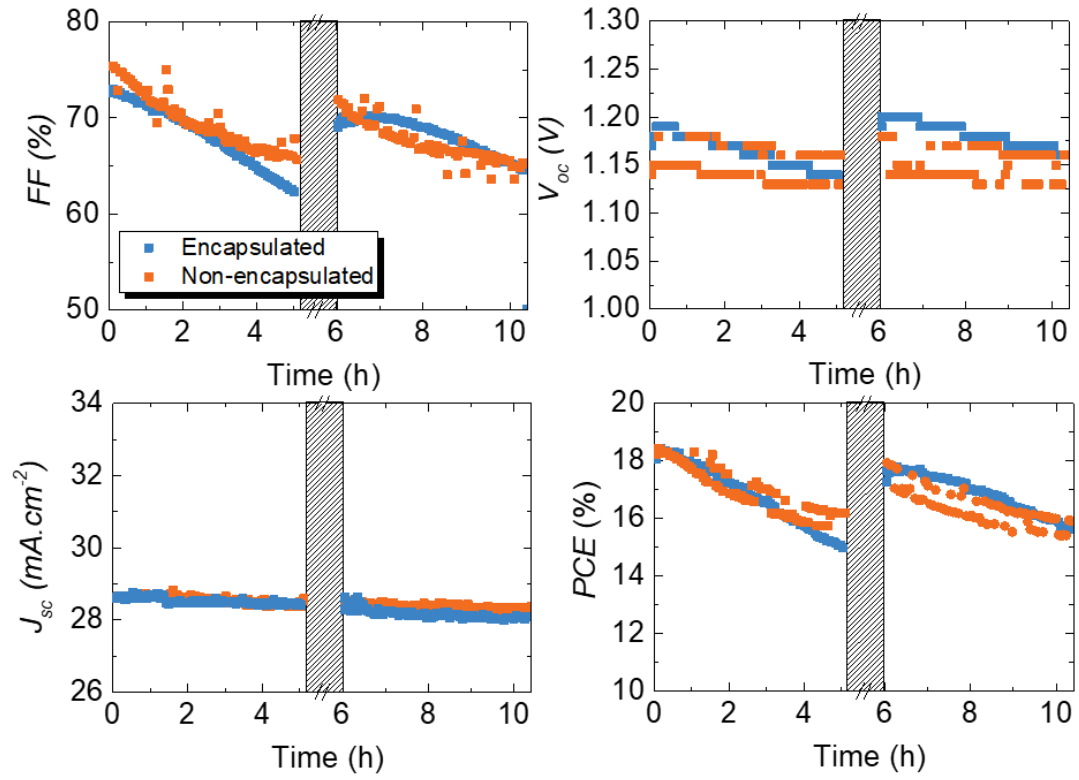
Figure S6. Variations in PV parameters of encapsulated and non-encapsulated devices measured at 10 mbar and 25°C under AM1.5G illumination.