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ORIGINAL ARTICLE

Flash-evaporation printing methodology for perovskite thin films

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Printing solutions have attracted wide attention from various research fields, but thin films made by printing technologies still cannot match the quality of the films made by conventional thin-film deposition methods. In this study, a flash-evaporation printing (FEP) technology, which employed a freestanding carbon nanotube (CNT) flash evaporator, was developed to address this issue and to achieve a compact physical vapor deposition geometry. The target material precoated on the CNT flash evaporator was printed onto a substrate by gas-phase transportation. The FEP methodology presented a printable solution for hybrid perovskite thin films. The as-fabricated photovoltaic devices showed power conversion efficiencies of $\sim 10.3\%$, and the thin films were also utilized as potential photodetectors. In addition, this technology can also be used to print patterns and a wide variety of materials on large panels. The environmentally friendly and cost-effective FEP technology will be of significant benefit to printed electronics, organic electronics and future flexible electronics.

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INTRODUCTION

For centuries, printing has been improving social civilization, from Gutenberg's printing press to today's state-of-the-art printing industry. Printing methodologies evolved into printed electronics when microelectronics were developed, making integrated circuits printable, flexible and cheaper. Printed electronics use printable paste and ink made from functional materials with complex recipes. I-11 Thus, the quality of printed films usually cannot be compared with thin films made by conventional physical vapor deposition (PVD) methods because of the high purity of the vapor source generated from the target in vacuum in PVD processes. This suggests that a great improvement of printed electronics is possible by integrating vacuum PVD procedures into printing processes.

Here, we propose a flash-evaporation printing (FEP) methodology that successfully combines a compact PVD process with printed electronics. The success is attributed to the flash evaporator made by cross-stacked superaligned carbon nanotube (CNT) films resulting in a fast high-temperature response. $^{12-14}$ The evaporator is $\sim 1.5~\mu m$ in thickness, and the heat capacity per unit area (HCPUA) is only $\sim 0.1~J~m^{-2}~K^{-1}$, over 3 orders of magnitude lower than metal heaters used in conventional thermal evaporation. A flash-evaporation transfer ribbon was developed by coating a target material on the evaporator.

The printing of the target material was conducted by the gas-phase transportation from the hot CNT evaporator stimulated by laser irradiation to the cold substrate below. The CNT evaporator, the facile transfer ribbon and the compact geometry promote the invariable flash evaporation that was implemented in a large thermal-evaporation facility to a different and new printing methodology. ^{15–17}

Organic-inorganic hybrid perovskites, such as CH₃NH₃PbI₃ (MAPbI₃), have recently captured intense research interest for their promising optoelectronic applications with low cost and high performance. 18-29 A two-step deposition 30 is the common route to synthesize perovskite thin films. In this process, lead iodide (PbI₂) is first introduced using solution or vapor deposition methods and is subsequently transformed into the perovskite by reacting with CH₃NH₃I (methylammonium iodide (MAI)). Typically, the as-formed compact PbI₂ layer blocks MAI penetration and diffusion into the deeper PbI2 layer, resulting in the incomplete transformation of PbI2 to the perovskite.³¹ Several strategies have been developed to solve this problem, such as incorporating a small amount of MAI³² or additive³³ into the precursor solution, producing a porous PbI2 nanostructured layer by solvent engineering, 31,34 using PbI₂/MAI multilayer stacks³⁵ and others.³⁶ However, fabricating perovskite thin films with a printing methodology is also an emerging trend.^{37,38} Here, the new

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FEP methodology was used to fabricate MAPbI₃ perovskite thin films. Nanoporous PbI₂ thin films were directly printed on substrates as precursors by FEP, and then high-quality perovskite thin films were produced by an *in situ* reaction with an MAI solution. The properties and morphology of the as-prepared films were shown to be satisfactory by X-ray diffraction (XRD), ultraviolet–visible (UV–Vis) absorption spectroscopy and scanning electron microscopy (SEM). To further reveal the potential of the FEP technique, perovskite thin films were integrated into photovoltaic devices that exhibited power conversion efficiencies (PCEs) of ~10.3%, and these films were also utilized to make photodetectors. Furthermore, this distinctive printing method is competent for patterning and is environmentally friendly and cost effective. The FEP technique will prove itself applicable to a wide variety of materials and will be a great impetus to printable electronics and flexible electronics.

MATERIALS AND METHODS

Materials

Unless specified otherwise, all materials were purchased from either Alfa Aesar (Ward Hill, MA, USA) or Sigma-Aldrich (St Louis, MO, USA). MAI was purchased from Dyesol (Queanbeyan, NSW, Australia).

Fabrication of the flash-evaporation transfer ribbon

CNT films, used as building blocks for the flash evaporator, were dry spun from superaligned CNT arrays synthesized on silicon wafers by low-pressure chemical vapor deposition. 39,40 A cross-stacked CNT sheet with 15 single layers was coated onto a 25 × 25 mm² metal frame. After stacking, the CNT sheet was dipped in ethanol and dried in air at room temperature to make the CNTs compact. A PbI₂ solution made of 1.3 M PbI₂ precursor in anhydrous dimethylformamide and dimethylsulfoxide (17:3, v/v) was then deposited onto the CNT sheet by spin-coating. The transfer ribbon was made by dropping 100 μl of the PbI₂ solution onto the CNT sheet mounted on a sample holder and then spinning at 2000 r.p.m. for 30 s. During the formation of the PbI2 film precursor, the transfer ribbon was heated to 100 °C for 10 min to remove the solvents. Notably, the transfer ribbon can be scaled up using larger CNT evaporators, as the mass production of a superaligned CNT array on an 8 inch wafer has been realized, and ~200 m-long CNT thin films can be spun from each wafer. 40 After the fabrication, a flash-evaporation transfer ribbon was placed on top of a substrate with a gap of 1 mm, as shown in the schematic diagram of the FEP (Figure 1a). A sample holder was employed to set the substrate and the transfer ribbon that can be seen in the compact FEP chamber (Supplementary Figure S1).

Temperature measurement of the hot CNT flash evaporator

The spectra of the hot CNT flash evaporator were recorded by a spectrometer (Konica Minolta CS-1000, Tokyo, Japan), and the temperature was derived by fitting the spectra with the blackbody radiation law, as we previously reported. 41,42 The variation of the laser-power-dependent heating temperature is shown in Supplementary Figure S2.

Photovoltaic device fabrication

Devices were fabricated on fluorine-doped tin oxide (FTO)-coated glass (Nippon Sheet Glass Co., Ltd, Tokyo, Japan). Initially, FTO was removed from the region under the anode contact area by etching the FTO with 2 M HCl and zinc powder. The substrates were then cleaned sequentially in soap, deionized water, ethanol, acetone, isopropanol and oxygen plasma. A compact layer of titanium dioxide (TiO₂) was subsequently deposited by spin-coating a mildly acidic solution of titanium isopropoxide in ethanol at 2000 r.p.m. for 60 s and annealing at 500 °C for 30 min. Then, 80 µl of a PCBM ([6,6]-phenyl-C61-butyric acid methyl ester) solution (10 mg ml⁻¹) was spin-coated on the top of TiO₂ at 4000 r.p.m. for 40 s in a glovebox. The PbI₂ film was printed on the PCBM/TiO₂ substrate via the FEP process. After PbI₂ deposition, the samples were transferred into a glovebox for further processing. The perovskite layer was obtained by dipping the PbI₂ precursor into an MAI solution (10 mg MAI in per ml isopropyl alcohol) for 10 min and drying at 100 °C for

30 min. The hole-transport layer was deposited by spin-coating 2,2',7,7'-tetrakis $(N,N-\operatorname{di}(4-\operatorname{methoxyphenyl})\operatorname{amino})-9,9'$ -spirobifluorene (Spiro-OMeTAD) in chlorobenzene solution with added tert-butylpyridine and lithium bis(trifluoromethanesulfonyl)imide. The manufacturing process of the device was completed by thermally evaporating Au as the back electrode.

Characterization

The quality and morphology of the thin films were characterized by XRD (Rigaku D/max-2500/PC, Tokyo, Japan), UV-Vis absorption spectroscopy (Perkin Elmer Lambda 950, Waltham, MA, USA) and SEM (FEI Sirion 200, operated at 10 kV, Hillsboro, OR, USA). The current-voltage characteristics of the solar cells were recorded under AM 1.5G 100 mW cm⁻² simulated sunlight (Newport Oriel 92192, Irvine, CA, USA) with a Keithley 2400 sourcemeter (Solon, OH, USA), previously calibrated with a calibrated Si solar cell. The solar cells were masked with a metal aperture defining the active area (0.06 cm²) of the devices. The incident photon-to-current efficiency (IPCE) spectra were characterized using a QEX10 solar cell quantum efficiency measurement system (QEX10, PV Measurements, Boulder, CO, USA). The photocurrent signal of the photodetector was measured using an Agilent (Santa Clara, CA, USA) B2902A sourcemeter by illuminating the device through a 0.2 cm² aperture with a xenon lamp (AuLight, CEL-HXF300, Beijing, China) filtered by a bandpass filter of 530 nm. The power was measured with a calibrated Thorlabs (Newton, NJ, USA) PM100D power meter. The response speed was surveyed by illuminating the device with a supercontinuum white-light source (NKT Photonics, Birkerød, Denmark, SuperK Compact), and the photocurrent was recorded with a Tektronix (Beaverton, OR, USA) oscilloscope (TDS2012C).

RESULTS AND DISCUSSION

The FEP technology is illustrated in Figure 1a, and a photograph of the experimental setup is shown in Supplementary Figure S1. A flash-evaporation transfer ribbon with PbI2 coated on CNT film-based evaporator was set on top of a substrate with a 1 mm gap. A picture of the 25 × 25 mm² freestanding CNT sheet is shown in Figure 1b, and Figure 1c provides an optical micrograph. The HCPUA and the thickness of the CNT evaporator were $\sim 0.1 \, \text{J m}^{-2} \, \text{K}^{-1}$ and 1.5 µm, respectively, that were calculated from the data of a singlelayer CNT film (0.0077 J m⁻² K⁻¹ and 100 nm per layer). ¹² The HCPUA and the thickness are much smaller than those of a conventional metal heater in a thermal evaporator; for example, the tantalum heater used in the study of Longo et al. 17 was 117 J m⁻² K⁻¹ and 100 µm in thickness. The ultrasmall HCPUA and thickness are endowed by the unique CNT material and can hardly be achieved by conventional materials, ensuring the feasibility of the FEP. The flashevaporation transfer ribbon is shown in Figure 1d (optical micrograph) and Figure 1e (SEM image). The base pressure of the process chamber was $\sim 10^{-3}$ mbar. Because the CNT evaporator is black, as shown in Figure 1b, the yttrium aluminum garnet laser with a wavelength of 1.06 µm was employed as a stimulus. The laser beam scanned the transfer ribbon with a speed of 1 m s⁻¹ through a quartz window. The laser spot was ~ 1 mm in diameter, and the pitch of the scanning lines was 100 $\mu m.$ The temperature of the laser-heated flash evaporator can be modulated by the laser power, as shown in Supplementary Figure S2, and the optimized power was 12 W for printing the PbI2 precursor film. The CNT evaporator can be heated to ~1100 °C instantaneously because of its ultrasmall heat capacity. Then, flash evaporation can start immediately, and gas-phase PbI₂ can be transported from the transfer ribbon to the substrate. It takes only ~ 4 s to print a $20 \times 20 \text{ mm}^2 \text{ PbI}_2$ film. More experiments showed that the thicknesses of the printed PbI2 films could be well controlled by the amount of PbI₂ spin-coated on the CNT films (Supplementary Figure S3). Figure 1f shows the morphology of the transfer ribbon after flash evaporation, revealing a very clean surface without PbI₂ residue, suggesting that the flash-evaporation process is efficient.

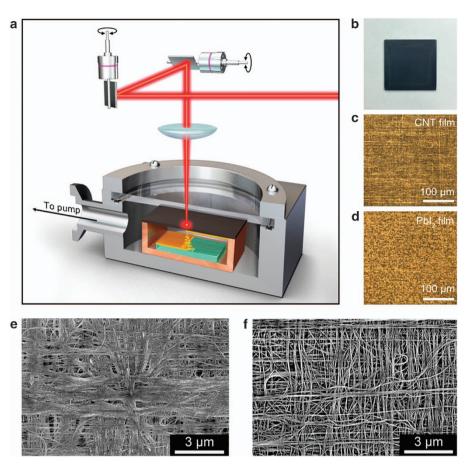


Figure 1 Illustration of the flash-evaporation printing (FEP) process and morphology of the transfer ribbon. (a) Schematic depiction of the experimental setup of the FEP. (b) Photograph of the carbon nanotube (CNT) sheet as the flash evaporator. (c, d) Optical micrographs of the CNT evaporator and the transfer ribbon. (e) Scanning electron microscopy (SEM) image of Pbl₂ on the CNT films. (f) Clean CNT films without Pbl₂ residue after flash evaporation.

The as-printed thin film was investigated by XRD and SEM. The XRD result is shown in Figure 2a. An intense peak at 12.6° corresponds to the (001) diffraction peak of PbI₂, validating the success of the FEP technology in printing the PbI₂ film. The surface morphology of the PbI₂ film was also investigated, and the SEM images at different magnifications are given in Figures 2c and d. The morphology of the PbI₂ film is uniform and abundant of nanoscaled grains that is obviously distinct from a solution-processed PbI₂ film (Supplementary Figure S4), and this highly porous nanostructure is favorable for subsequent MAI penetration and perovskite synthesis.

The well-prepared PbI₂ precursor allows for the fabrication of the final MAPbI₃ perovskite thin film. The PbI₂ film was dipped into an MAI solution (10 mg ml⁻¹ in isopropyl alcohol). The yellow PbI₂ film immediately turned dark red once the film was immersed into the MAI bath, indicating that the PbI₂ film reacted with MAI efficiently. The dipping process took 10 min, and then the *in situ* synthesized film was studied by SEM, XRD and UV–Vis absorption spectroscopy. The SEM images in Figures 2e and f show a uniform surface and a notable change of morphology induced by the MAI bath. The crystalline structure was further studied by XRD, as depicted in Figure 2a. Intense diffraction peaks at 14.1°, 28.4°, and 31.8° of the red plot correspond to the (110), (220), and (310) planes of the tetragonal MAPbI₃ phase.^{21,30,43} Moreover, the XRD pattern exhibits two extra peaks at 19.9° and 24.4° that were observed in the simulated perovskite XRD pattern reported by Malinkiewicz *et al.*⁴⁴ It is noteworthy that there is

only a small signature of a peak at 12.6° (PbI₂), indicating that there is little PbI₂ left on the substrate. Hence, the perovskite film has a high phase purity. UV–Vis absorption spectroscopy was then used to study the characteristic absorption of the perovskite films, as given in Figure 2b. The two comparative absorption spectra show the prominent changes of the optical properties induced by the MAI bath. This figure also shows a band-to-band optical absorption edge at ~ 800 nm (~ 1.55 eV) for the MAPbI₃ perovskite, with an absorption plateau at wavelengths below 500 nm. 23,45 The strong absorption of light indicates that the perovskite film may be promising for photovoltaic applications.

The FEP technology and the perovskite film made by the two-step strategy can be integrated into a solar cell. The details of the fabrication and characterization of the devices can be found in the Materials and Methods section. Figure 3a provides the structural diagram of the planar perovskite solar cell: an FTO glass served as a transparent electrode; a PCBM-coated TiO₂ film served as an electron-transport layer; a perovskite absorber layer was derived from the printed PbI₂, Spiro-OMeTAD served as the hole-transport layer and Au served as a back electrode. The color-enhanced cross-sectional SEM image of the device architecture is shown in Figure 3c, where each layer can be clearly defined according to the color variation. The energy-level diagram is shown in Figure 3b. Note that PCBM is also important to achieve better electron extraction and transportation that can be examined in the comparative studies in the following experiments. Figures 3d and e display the representative

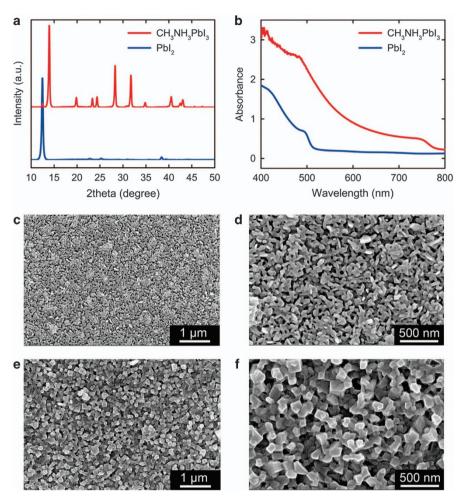


Figure 2 Characterization of the as-printed thin films. (a) X-ray diffraction (XRD) patterns and (b) ultraviolet–visible (UV–Vis) absorption spectra of the Pbl₂ and MAPbl₃ thin films deposited on FTO/TiO₂/PCBM substrates. (c, d) Scanning electron microscopy (SEM) images of the Pbl₂ film deposited on an FTO/TiO₂/PCBM substrate. (e, f) SEM images of the perovskite film derived from the Pbl₂ film dipped into methylammonium iodide (MAI) solution. FTO, fluorine-doped tin oxide; PCBM, [6,6]-phenyl-C61-butyric acid methyl ester; TiO₂, titanium dioxide.

current density versus voltage (*J–V*) characteristics and external quantum efficiency (EQE) or IPCE⁴⁶ spectra of the optimized planar devices.

Hysteresis phenomena were also revealed by the forward and reverse voltage scans. Table 1 summarizes the photovoltaic parameters of these devices. The optimized device with the PCBM layer showed an open-circuit voltage ($V_{\rm oc}$) of 0.94 V, a short-circuit current density ($J_{\rm sc}$) of 15.9 mA cm⁻², a fill factor of 68.9% and a PCE of 10.3% in the reverse scan. In the forward scan, a PCE of 8.9% was obtained with a $V_{\rm oc}$ of 0.94 V, a $J_{\rm sc}$ of 14.5 mA cm⁻² and a fill factor of 65.3%. Furthermore, the $J_{\rm sc}$ was confirmed by IPCE measurement (Figure 3e) with an integrated $J_{\rm sc}$ of 14.5 mA cm⁻². The device without the PCBM layer showed a much lower PCE with pronounced hysteresis, indicating the vital role of PCBM in modifying the TiO₂/perovskite interface. ^{21,47} Moreover, the nanostructured PbI₂ precursor made by FEP showed performance advantages compared with the solution-processed PbI₂ precursor synthesized in our lab.

The photovoltaic parameters of the best-performing device prepared starting from solution-processed PbI_2 are also listed in Table 1 for comparison, and the J-V characteristics of this batch of samples are provided in Supplementary Figure S5. The PCEs can be possibly improved by following the latest enhancement techniques, for

example, utilizing mixed-cation perovskites and mesoporous ${\rm TiO_2}$ or performing extra solution processing to smoothen the perovskite surface. In addition, an optimized FEP system with an ultrahigh vacuum, a load lock and a transfer chamber will be beneficial to the printed thin films, as the materials can thus avoid exposure to active residual and air.

The as-fabricated perovskite photovoltaic devices can also be used as photodetectors. The responsivity of the device can be calculated from the IPCE spectra (Figure 3e) by the following equation:^{49,50}

$$EQE = \frac{R_{\lambda}}{\lambda} \times \frac{hc}{e} \approx \frac{R_{\lambda}}{\lambda} \times (1240W \cdot nm/A)$$

where EQE is the external quantum efficiency, also known as IPCE, R_{λ} is the responsivity, λ is the light wavelength in nm, h is Planck's constant, c represents the velocity of light in a vacuum and e is the elementary charge. The R_{λ} versus λ curves are presented in Figure 4a. It shows that the devices with PCBM have a photoresponse from 350 to 780 nm, suggesting that the photodetector has a wide-range photoresponse from UV to visible light. The peak responsivity was $0.31\,\mathrm{A\,W^{-1}}$ at a wavelength of 740 nm, comparable to commercial large-area silicon photodiodes. Further experiments were performed to investigate the responses to varying degrees of input irradiance under the illumination of light with a wavelength of

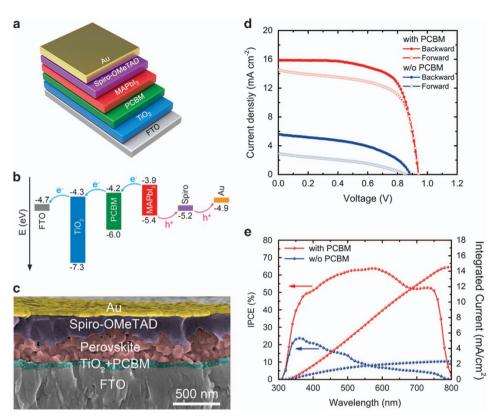


Figure 3 The structure and performances of the perovskite solar cells. (a) Structural diagram of the hybrid perovskite photovoltaic device. (b) Energy band alignment of the device stack. (c) False color cross-sectional-view scanning electron microscopy (SEM) image of a complete photovoltaic device. (d) *J–V* curves of the best-performing devices measured at a simulated AM1.5G solar irradiation of 100 mW cm⁻². (e) Incident photon-to-current efficiency (IPCE) spectra (the right-hand axis indicates the integrated photocurrent expected to be generated under AM1.5G irradiation).

Table 1 Photovoltaic parameters of the perovskite solar cells with and without PCBM interface layers

	Device	V _{oc} (V)	J _{sc} (mA cm ⁻²)	FF (%)	PCE (%)
TiO ₂ /PCBM	Reverse scan	0.94	15.9	68.9	10.3
	Forward scan	0.94	14.5	65.3	8.9
TiO ₂	Reverse scan	0.89	5.6	48.2	2.4
	Forward scan	0.86	2.9	36.1	0.9
Solution-processed	Reverse scan	0.93	16.4	61.3	9.3
	Forward scan	0.84	17.9	34.5	5.2

Abbreviations: FF, fill factor; J_{sc} , short-circuit current density; PCBM, [6,6]-phenyl-C61-butyric acid methyl ester: PCE, power conversion efficiency: TiO₂, titanium dioxide: V_{cc} , open-circuit voltage.

530 nm. Figure 4b shows the photocurrent versus incident light intensity plots for the devices, revealing linear responses. The linear dynamic range (LDR) can thus be calculated according to the equation of LDR=20 $\log(J_{\rm ph}^*/J_{\rm dark})$, where $J_{\rm ph}^*$ is the photocurrent measured at a light intensity of 1 mW cm^{-2,52} The LDR exceeded 80 dB for the photodetector with PCBM, and this is higher than those of InGaAs photodetectors (66 dB). The comparative curves in Figures 4a and b also indicate that the device with PCBM performs much better. Moreover, the device functioned well in the measurement of response speed with an oscilloscope. A supercontinuum white-light source with a pulse duration < 2 ns and a frequency of 500 Hz⁵³ was irradiated on the device. The transient photocurrent response of the device with PCBM is shown in Figure 4c. The rise time and decay time (defined as the time to reach 90% of the maximum photocurrent from the dark current and the time to recover to 1/e of

the maximum photocurrent, respectively⁵⁴) were ~10 and 200 μ s, respectively, agreeing well with the results in the literature.^{29,55} Therefore, it can be confirmed that the FEP methodology is well suited to fabricate perovskite films for solar cells and photodetectors.

The FEP technique can also be employed to print patterned thin films that is essential for printing technologies. Figure 5 shows a simple but efficient strategy to pattern perovskites with a shadow mask. The substrate was first covered by a mask with desired patterns, and then PbI2 was printed onto the mask-covered substrate with the FEP process, and finally the printed film was exposed to the MAI bath. The patterned perovskite film was thus fabricated on the substrate. The central panel illustrates a perovskite array on a flexible polyethylene terephthalate substrate. Another feasible strategy for printing patterns with FEP can be modeled from conventional printing technologies, such as ink-jet printers and impact printers. A conceptual diagram of a flash-evaporation printer is illustrated in Figure 6. The compact module integrates a transfer ribbon, shadow mask and laser. The designed pattern is point-by-point printed onto the substrate by scanning the surface with the FEP module. This printer provides a solution for the large-area fabrication of functional devices with the FEP methodology.

In addition to the printed solar cell and the efficient patterning, the FEP technology shows more advantages for the preparation of perovskite thin films. The distance between the transfer ribbon and the substrate is only ~ 1 mm, much smaller than that in conventional thermal evaporation. The close range can efficiently improve the PbI₂ utilization during evaporation and thus effectively reduce lead pollution to the environment. It has been shown that the CNT

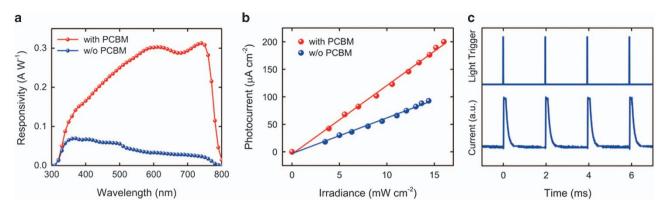


Figure 4 The properties of the perovskite photodetectors. (a) Spectral responsivity derived from the incident photon-to-current efficiency (IPCE) spectra. (b) Photocurrent linearity upon varying the 530 nm input irradiance. (c) Transient photocurrent response at a pulse frequency of 500 Hz with a device area of 0.2 cm².

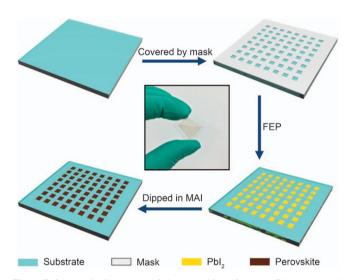


Figure 5 Schematic illustration of the deposition of regular ${\sf Pbl}_2$ arrays and the formation of patterned perovskites.

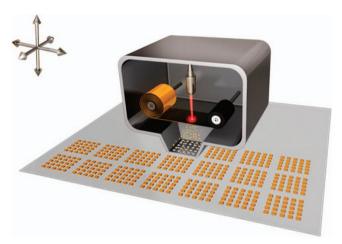


Figure 6 A conceptual diagram of a flash-evaporation printer. The addressable compact flash-evaporation printing (FEP) module integrates a transfer ribbon, shadow mask and laser.

evaporator maintains a good condition after flash evaporation (Figure 1f), indicating that the CNTs can be fully reused. Furthermore, the future applications of this technology are promising. For example, FEP can be developed into a roll-to-roll strategy for mass production with low cost that is facilitated by the flexibility of the CNTs (Figure 6). Additionally, the compact geometry of the FEP dramatically simplifies the process chamber (Supplementary Figure S1) and is more cost effective than conventional evaporation facilities. The FEP chamber also provides a clean and sealed vacuum environment for the procedures compared with solution methods that is beneficial to improve the thin film quality and interlayer interface. These merits indicate that the FEP is not only environmentally friendly but also cost effective in terms of the applications of the MAPbI₃ perovskite.

The FEP we developed is a distinctive technology. It allows thermalevaporation technology to evolve into a general printing solution. In addition to the PbI₂ films shown here, this technology can be applied to print most other evaporation materials. Supplementary Figures S6 and S7 show patterned Tris(8-hydroxyquinolinato)aluminum (Alq₃) and Au thin films, demonstrating the potential to print organic light-emitting diodes and interconnecting microdevices with direct metal printing. Research on the direct printing of perovskite

thin films using an MAPbI₃ transfer ribbon is also in progress, and it will be reported elsewhere in the future. It should be noted that the flash-evaporation transfer ribbons of different materials can be prepared through appropriate routes that is not limited to solution processing. Moreover, multiple stimuli can be employed by the FEP technology. In addition to lasers, the CNT evaporator can also be heated by Joule heating and hybrid heating methods. The CNT evaporator prevails over metal heaters because of its ultrasmall HCPUA and thickness. It can be heated to an elevated temperature within milliseconds. The cold end effect is also significantly reduced by the ultrathin structure, resulting in a more uniform temperature distribution (Supplementary Figure S8). The heating currents through the CNT evaporator are greatly reduced compared with metal evaporators, and this is meaningful to a compact FEP system with a Joule heating stimulus. Therefore, it is reasonable to expect the FEP to have prospective applications in a wide range of fields.

CONCLUSION

In summary, a convenient FEP technology was realized by incorporating a PVD technique and a printing method that provides an efficient approach for printing high-quality thin films. A flash-

evaporation transfer ribbon was crucial for the success of the FEP, and it was fabricated by coating target materials on a freestanding CNT sheet that has an ultrasmall heat capacity and fast thermal response. The printing was conducted by the gas-phase transportation from the transfer ribbon to a substrate. Using the FEP technique, a novel printable solution was developed to fabricate perovskite thin films, and detailed analysis revealed that the printed PbI₂ precursors and the corresponding MAPbI₃ thin films had ideal crystalline structures and high qualities. The potential of the FEP was verified by its ideal integration into the fabrication process of photovoltaic devices that showed PCEs of ~10.3%, and these films were also utilized as photodetectors. Moreover, patterned perovskite films can also be prepared by FEP using a shadow mask or point-by-point printing technique, and the compact geometry allows for convenient scale-up for large panel applications. The FEP is a promising methodology for printing functional materials, and this technology will be of great benefit to a variety of applications in printed electronics, organic electronics and future flexible electronics.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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Author contributions: YW and KJ conceived and supervised the research. YW and HW designed the experiments. HW performed most of the experiments, including the FEP experiments, characterization and data analysis. HW and XZ fabricated the photovoltaic devices and performed the tests of the solar cells. HW and HM performed the photodetector measurements. The paper was written by HW and YW with contributions from all the co-authors.

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