Supporting Information

Artemisinin-Passivated Mixed-Cation Perovskite Films for Durable Flexible Perovskite Solar Cells with Over 21% Efficiency

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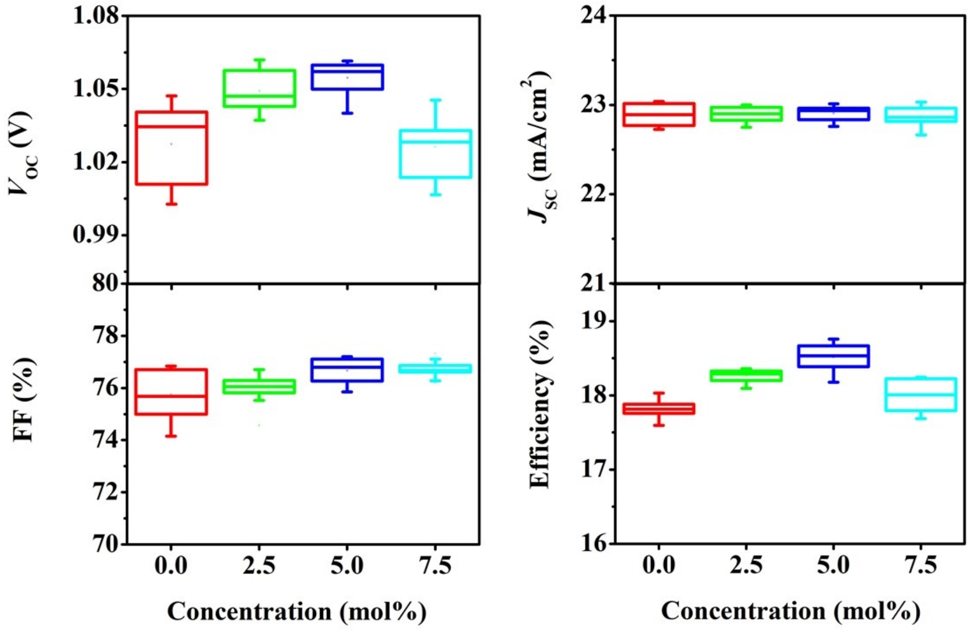


Figure S1. Photovoltaic parameters of flexible PSCs with different HCl concentrations under AM 1.5G illumination. The configuration of the cells was PEN/ITO/HfO_x/SnO_2 (control or modified)/perovskite/Spiro-OMeTAD/MoO_x/Ag.

Table S1. Photovoltaic parameters of the flexible perovskite solar cells (PSCs) with a structure of PEN/ITO/HfO_x/SnO_2 (control or modified)/perovskite/Spiro-OMeTAD/MoO_x/Ag under AM 1.5G illumination. Different concentrations of HCl were added into the SnO_2 solutions.

Perovskite	V_{OC} **(V)**	J_{SC} **(mA/cm^2)**	**FF (%)**	**PCE (%)**
Control (Average)	1.027±0.016	22.89±0.11	75.75±0.85	17.81±0.12
Champion	1.043	23.02	75.07	18.03
2.5 mol% (Average)	1.049±0.008	22.90±0.08	76.01±0.48	18.26±0.83
Champion	1.061	22.86	75.72	18.37
5.0 mol% (Average)	1.055±0.006	22.90±0.08	76.66±0.46	18.52±0.16
Champion	1.057	23.01	77.11	18.75
7.5mol% (Average)	1.033±0.012	22.89±0.11	76.63±0.29	18.12±0.21
Champion	1.033	23.01	76.78	18.25

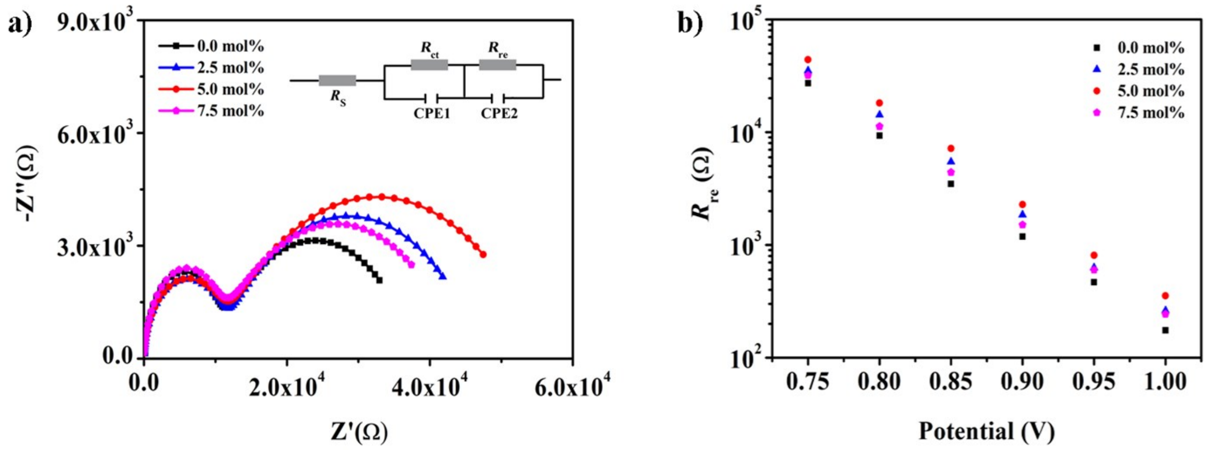


Figure S2. (a). Nyquist plots of the flexible PSCs based on SnO_2 ETLs with different concentrations of HCl at a bias of 0.75 V. (b) Dependence of fitted recombination resistance (R_{re}) on the bias voltage for all the flexible PSCs. The configuration of the cells was PEN/ITO/HfO_x/SnO_2(modified)/perovskite/Spiro-OMeTAD/MoO_x/Ag. The inset shows the equal electrical circuit for fitting.

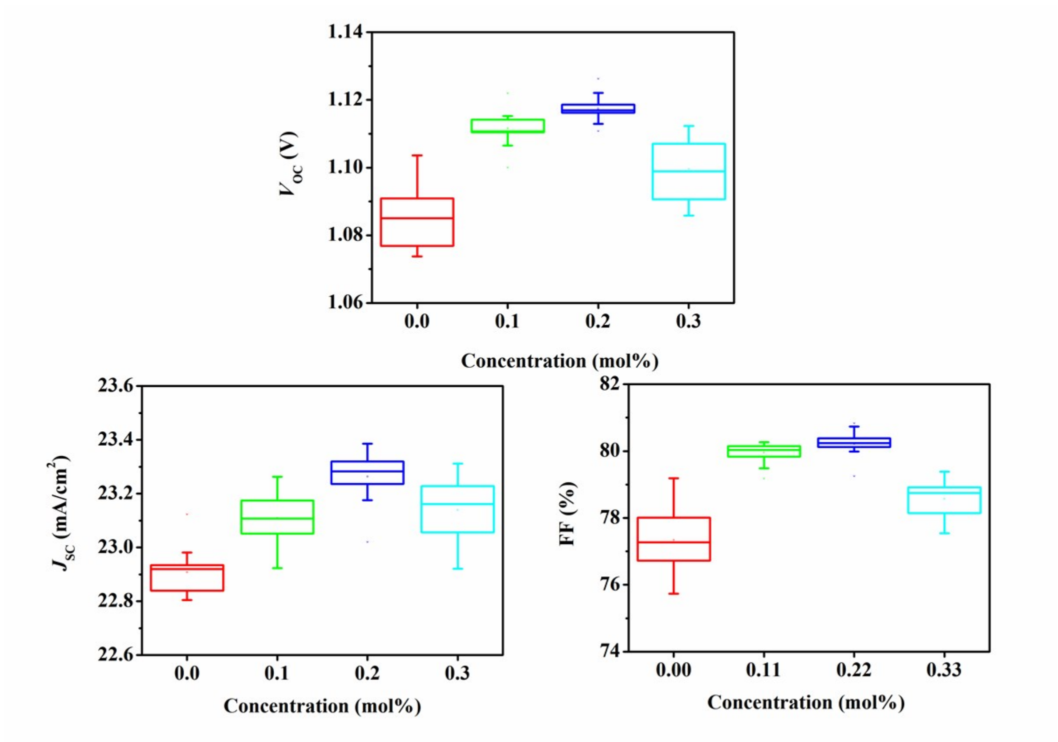


Figure S3. Photovoltaic parameters of flexible PSCs with the varied concentration of incorporated artemisinin under AM 1.5G illumination. The configuration of the cells was PEN/ITO/HfO_x/SnO_2(modified)/perovskite/PFAI/Spiro-OMeTAD/MoO_x/Ag.

Table S2. Photovoltaic parameters of the flexible perovskite solar cells (PSCs) with a structure of PEN/ITO/HfO_x/SnO_2(modified)/perovskite/ PFAI/Spiro-OMeTAD/ MoO_x/Ag under AM 1.5G illumination. Different concentrations of artemisinin were added into the perovskite solutions.

Perovskite	**V_{OC} (V)**	**J_{SC} (mA/cm^2)**	**FF (%)**	**PCE (%)**
Control (Average)	1.085±0.009	22.91±0.09	77.34±0.93	19.22±0.30
Champion	1.097	22.98	78.69	19.84
0.11 mol% (Average)	1.112±0.005	23.11±0.10	79.96±0.29	20.55±0.16
Champion	1.122	23.26	79.99	20.88
0.22 mol% (Average)	1.117±0.003	23.26±0.09	80.20±0.37	20.84±0.13
Champion	1.126	23.32	80.35	21.10
0.33 mol% (Average)	1.099±0.009	23.14±0.10	78.57±0.55	19.98±0.32
Champion	1.105	23.31	79.14	20.39

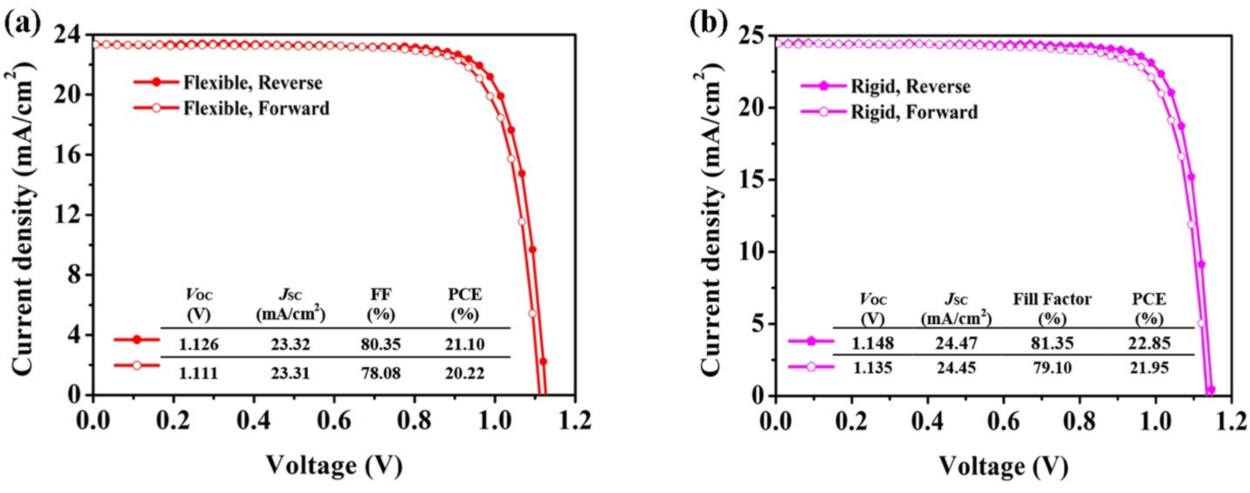
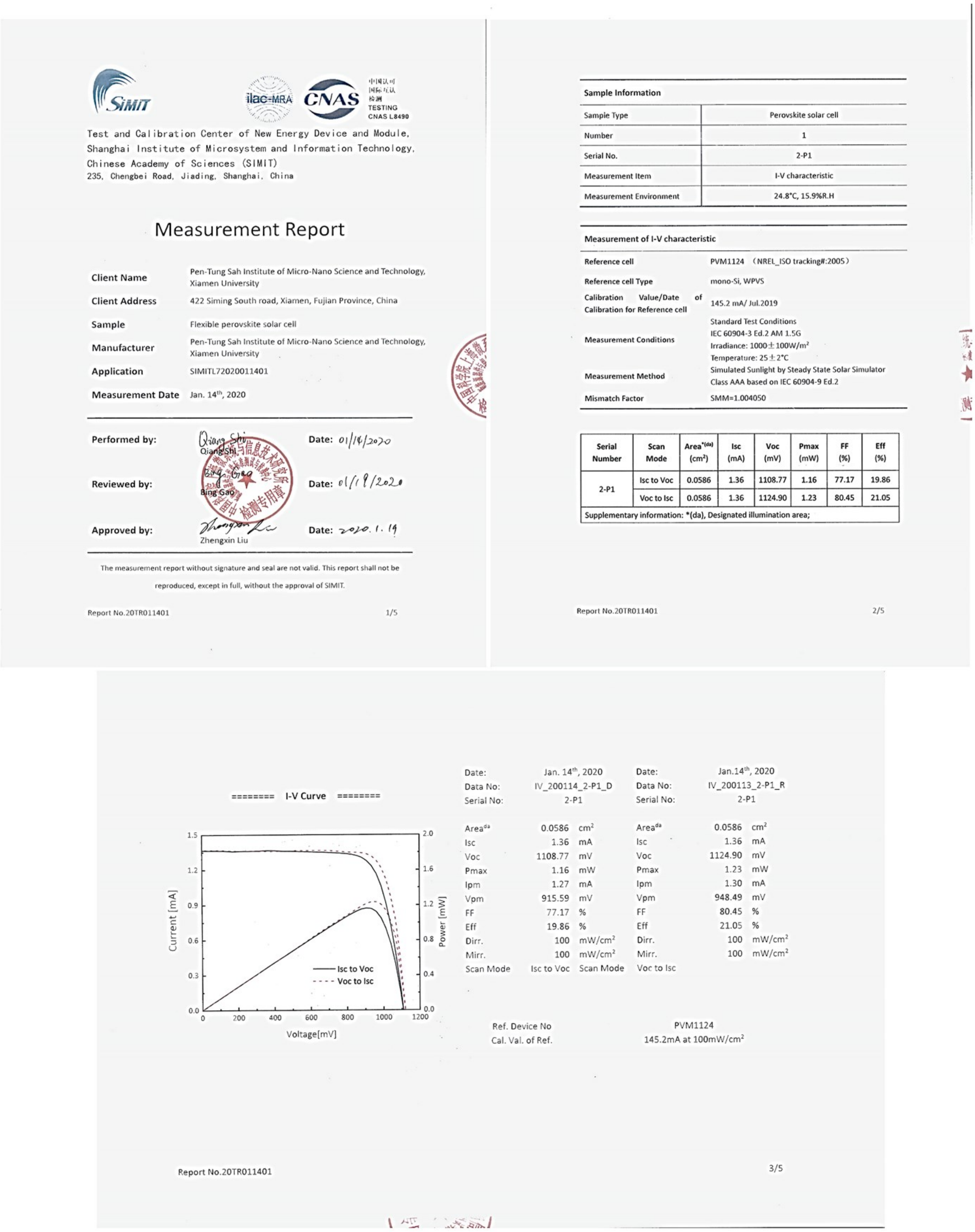


Figure S4. *J*-*V* curves of (a) the best flexible PSC and (b) the best rigid PSC under backward scan and forward scan directions under AM 1.5G illumination.



Test and Calibration Center of New Energy Device and Module,
Shanghai Institute of Microsystem and Information Technology,
Chinese Academy of Sciences (SIMIT)
235, Chengbei Road, Jiading, Shanghai, China

Measurement Report

Client Name	Pen-Tung Sah Institute of Micro-Nano Science and Technology, Xiamen University
Client Address	422 Siming South road, Xiamen, Fujian Province, China
Sample	Flexible perovskite solar cell
Manufacturer	Pen-Tung Sah Institute of Micro-Nano Science and Technology, Xiamen University
Application	SIMITL72020011401
Measurement Date	Jan. 14th, 2020

Performed by: Qiang Shi Date: 01/14/2020

Reviewed by: Bing Gao Date: 01/19/2020

Approved by: Zhengxin Liu Date: 2020.1.19

The measurement report without signature and seal are not valid. This report shall not be reproduced, except in full, without the approval of SIMIT.

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Sample Information

Sample Type	Perovskite solar cell
Number	1
Serial No.	2-P1
Measurement Item	I-V characteristic
Measurement Environment	24.8°C, 15.9%R.H

Measurement of I-V characteristic

Reference cell	PVM1124 (NREL_ISO tracking#:2005)
Reference cell Type	mono-Si, WPVS
Calibration Value/Date of Calibration for Reference cell	145.2 mA/ Jul.2019
Measurement Conditions	Standard Test Conditions IEC 60904-3 Ed.2 AM 1.5G Irradiance: 1000±100W/m² Temperature: 25±2°C
Measurement Method	Simulated Sunlight by Steady State Solar Simulator Class AAA based on IEC 60904-9 Ed.2
Mismatch Factor	SMM=1.004050

Serial Number	Scan Mode	Area*(da) (cm²)	Isc (mA)	Voc (mV)	Pmax (mW)	FF (%)	Eff (%)
2-P1	Isc to Voc	0.0586	1.36	1108.77	1.16	77.17	19.86
	Voc to Isc	0.0586	1.36	1124.90	1.23	80.45	21.05

Supplementary information: *(da), Designated illumination area;

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Date:	Jan. 14th, 2020		Date:	Jan.14th, 2020	
Data No:	IV_200114_2-P1_D		Data No:	IV_200113_2-P1_R	
Serial No:	2-P1		Serial No:	2-P1	
Area(da)	0.0586	cm²	Area(da)	0.0586	cm²
Isc	1.36	mA	Isc	1.36	mA
Voc	1108.77	mV	Voc	1124.90	mV
Pmax	1.16	mW	Pmax	1.23	mW
Ipm	1.27	mA	Ipm	1.30	mA
Vpm	915.59	mV	Vpm	948.49	mV
FF	77.17	%	FF	80.45	%
Eff	19.86	%	Eff	21.05	%
Dirr.	100	mW/cm²	Dirr.	100	mW/cm²
Mirr.	100	mW/cm²	Mirr.	100	mW/cm²
Scan Mode	Isc to Voc	Scan Mode	Voc to Isc		

Ref. Device No PVM1124
Cal. Val. of Ref. 145.2mA at 100mW/cm²

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Figure S5. The photovoltaic performance of a unencapsulated flexible PSC based on the perovskite film incorporating 0.22 mol% artemisinin certified by Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences (SIMIT, China). Reverse scan: V_{OC} = 1.125 V, I_{SC} = 1.36 mA, FF = 80.45%, and PCE

= 21.05%; Forward scan: V_{OC} = 1.109 V, I_{SC} = 1.36 mA, FF = 77.17%, and PCE = 19.86%. The certified active area of the mask is 0.0586 cm^2.

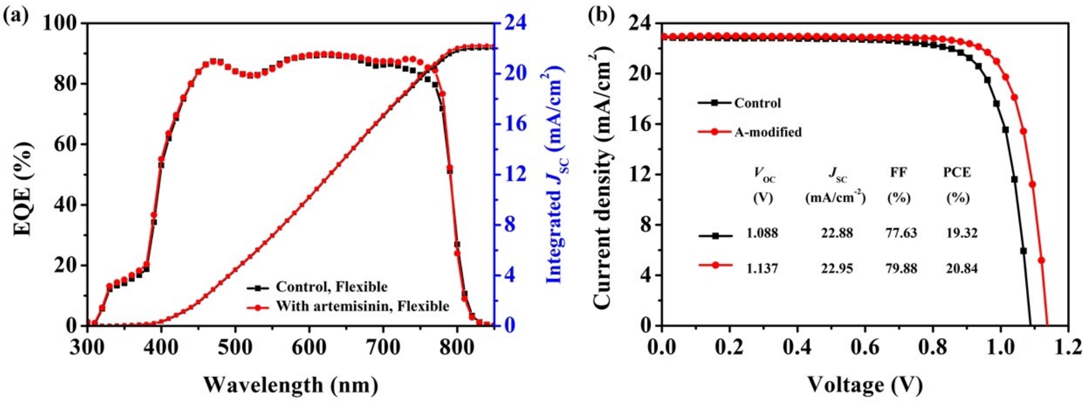

Figure S6. (a) EQE curves and (b) the corresponding *J-V* curves of a control flexible PSC and a artemisinin-doping (0.22 mol%) flexible PSC.

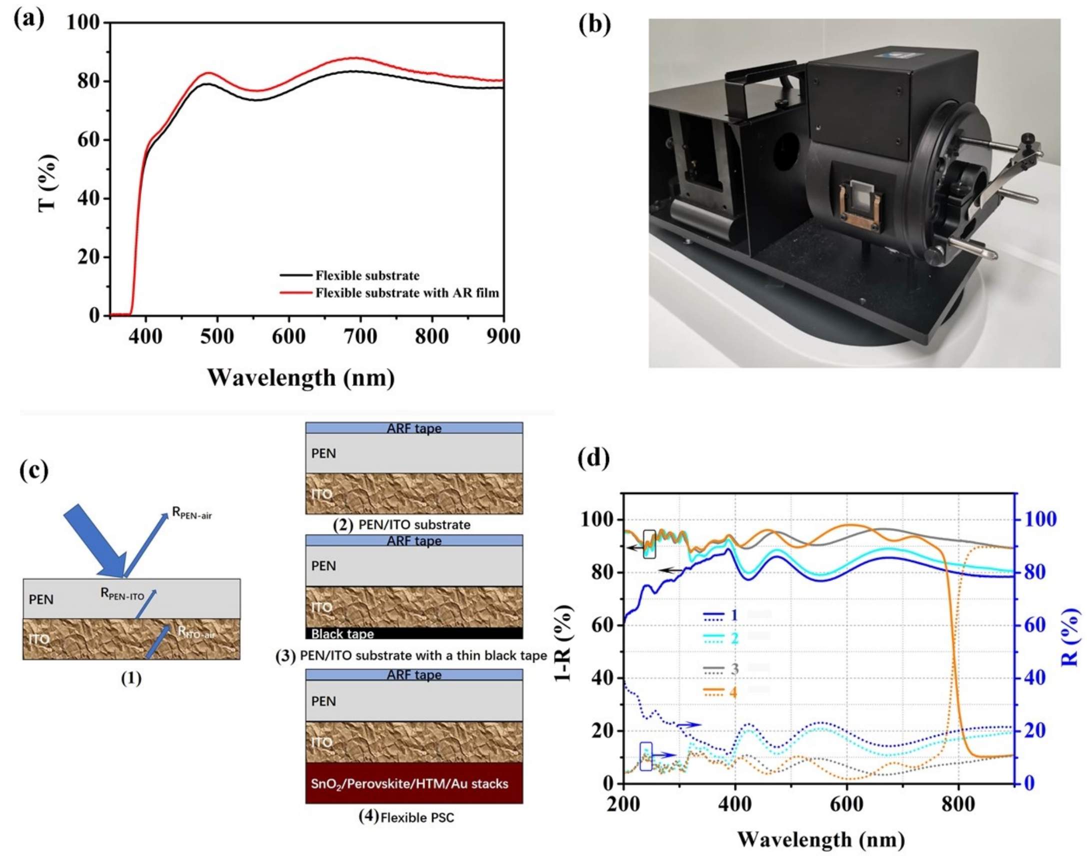

Figure S7. (a) The transmittance of the flexible substrate and flexible substrate with an ARF film; (b) The photograph of the integrating sphere; (c) Illustration of the four flexible structures for the reflection measurements; (d) 1-R (λ) spectra of the four flexible structures.

Table S3. Optical parameters of the different flexible structures at different wavelengths and corresponding EQE values. (T-Transmittance, R-Reflection; 1/2/3/4 are corresponding to the sample numbers in Figures S6 c and S6 d).

Optical properties and EQE	**λ=500 nm**				**λ=550 nm**				**λ=600 nm**				**λ=650 nm**			
Samples	1	2	3	4	1	2	3	4	1	2	3	4	1	2	3	4
T(%)	78.2	82.0	-	-	73.6	76.8	-	-	76.7	80.2	-	-	81.8	85.9	-	-
R (%)	17.1	14.6	6.7	9.9	23.2	20.9	9.6	6.6	19.8	17.0	6.7	2.0	15.0	11.7	3.7	4.5
1-R (%)	82.9	85.4	93.3	90.1	76.8	79.1	90.4	93.4	80.2	83.0	93.3	**98.0**	85.0	88.3	96.3	92.7
EQE (%)	84.1				84.8				**89.4**				89.1			

From Figure S6 a, it can be seen that the light transmittance at around 600 nm of the bare PEN/ITO substrate is below 80%. As a previous study pointed out, the absorption of PEN material is around 390 nm. [1] We need to determine what causes such a low transmittance? We thought that the reflection should be responsible for that, which could happen at the air/PEN interface, PEN/ITO interface, and ITO/air interface, according to the following equation:

$$T(\lambda) = 1 - R(\lambda) - A(\lambda) \quad (1)$$

where A(λ) could be ignored at the wavelength of 600 nm. Hence, the equation (1) can be simplified as:

$$T(\lambda) = 1 - R(\lambda) \quad (2)$$

Hence, we measured the reflection of the flexible substrates using a Cary 5000 with an integrating sphere shown in Figure S6 b. There are three windows in the integrating sphere for the light source, the sample, and the detector. The reflecting light of the flexible substrates can be collected and then measured. The reflecting light of the flexible substrates can be collected within the integrating sphere and then measured. There are several interfaces in the flexible substrates, including air/PEN, PEN/ITO, and ITO/air, which can cause light reflection due to the different **reflective indexes**.

PEN/ITO is hard to be determined by the integrating sphere, which is inside the PEN/ITO substrates. Hence, we mainly focused on the air/PEN and ITO/air interfaces of the flexible PEN/ITO substrates. We tried four combinations of flexible structures, as shown in Figure R1c, as the bare PEN/ITO, anti-reflection film (ARF)/PEN/ITO (air-PEN interface reflection), ARF/PEN/ITO/black tape (ITO/air interface reflection), and ARF/PEN/ITO/Sn-O_2/perovskite/HTM/Au structures. The results are given in Figure S6 d and Table S3.

It can be concluded from Figures S6 a and S6 d that:

(1) **Reflection is mainly responsible for the low light transmittance at around 600 nm. If reducing most of the reflection light, the incident light can be fully utilized.**
(2) Reflection happening at the air-PEN interface is relatively low as ~3.5%.
(3) Reflection happing at the ITO/air interface is strong owing to the largest difference of the reflective indexes between ITO and air.
(4) The stacks of the functional layers of the flexible PSC can effectively reduce the ITO/air interfacial refection.

Based on the parameters are given in Table S3, it can be seen that the light can be fully utilized (for example, at the wavelength of 600 nm, the light-utilization can be as high as **98%** when fabricating perovskite solar cells on PEN/ITO).

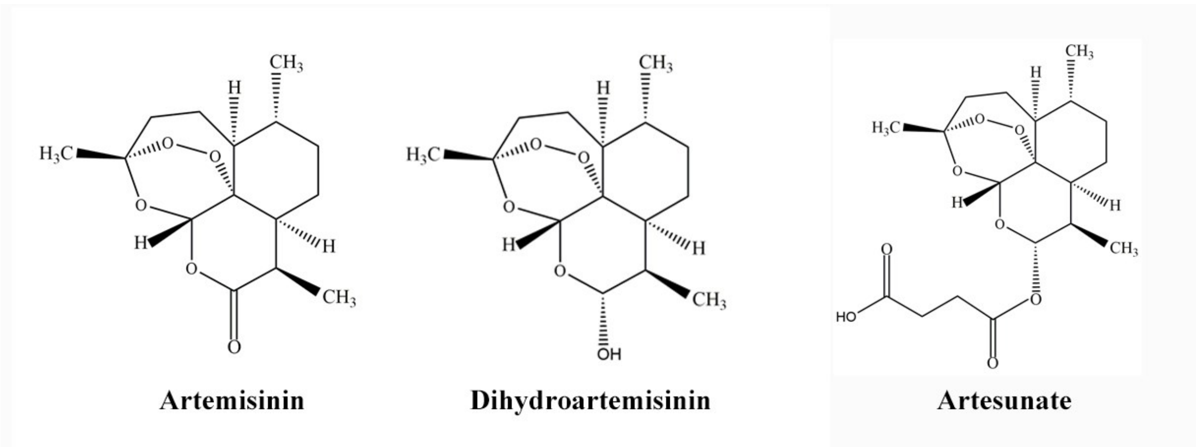

Figure S8. The molecular structures of three artemisinin derivatives: artemisinin, dihydroartemisinin, and artesunate.

Table S4. Photovoltaic parameters of the flexible PSCs with a structure of PEN/ITO/HfO_x/SnO_2(modified)/perovskite(doped)/ PFAI/Spiro-OMeTAD/ MoO_x/Ag under AM 1.5G illumination. Different artemisinin derivatives were added into the perovskite solutions and the concentrations are fixed as 0.22 mol%.

Perovskite	V_{OC} **(V)**	J_{SC} **(mA/cm²)**	**FF (%)**	**PCE (%)**
Artemisinin (Average)	1.117±0.003	23.26±0.09	80.20±0.37	20.84±0.13
Champion	1.126	23.32	80.35	21.10
Artesunate (Average)	1.126±0.005	23.24±0.24	79.36±0.41	20.76±0.24
Champion	1.130	23.39	79.48	21.01
Dihydroartemisinin (Average)	1.084±0.007	22.87±0.09	76.67±1.03	19.01±0.28
Champion	1.095	22.96	77.35	19.47

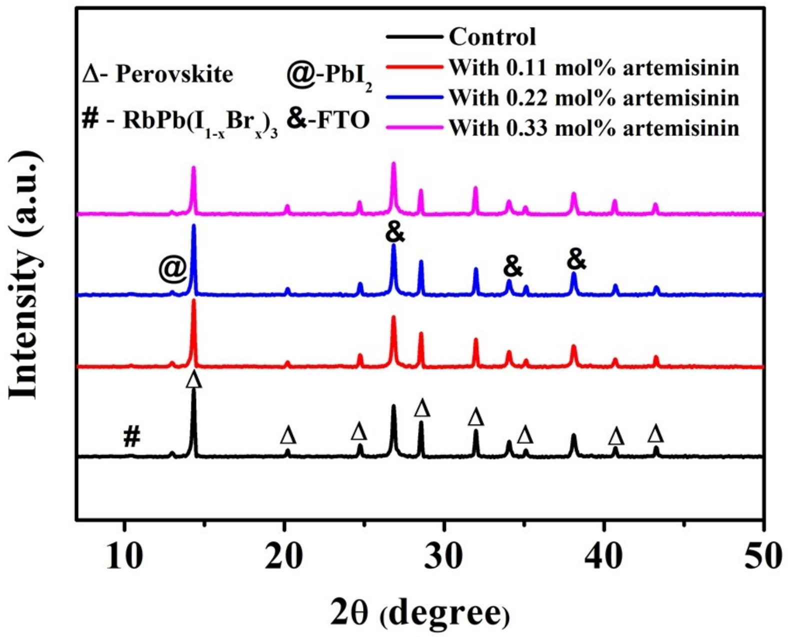

Figure S9. XRD patterns of neat perovskite film and perovskite films consisting of varied concentrations of artemisinin.

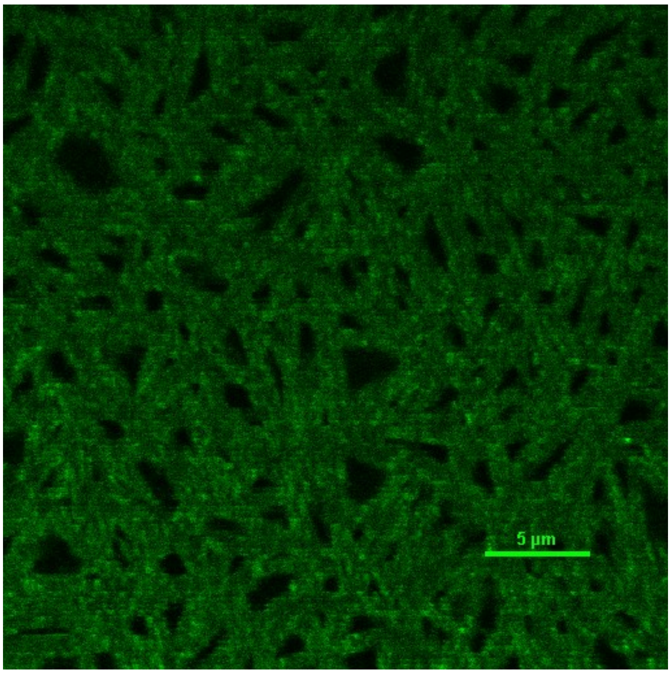

Figure S10. Confocal PL image of a pure PbI_2 film.

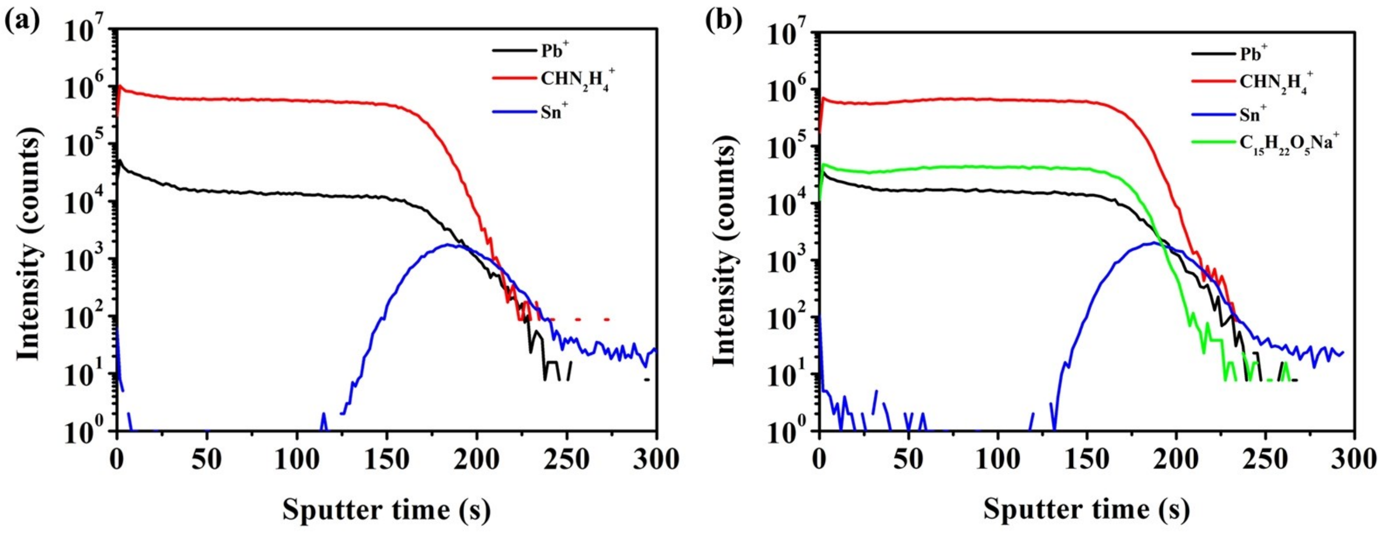

Figure S11. TOF-SIMS depth profile of (a) a control perovskite film and (b) a artemisinin perovskite film. The experiments were conducted on a TOF-SIMS 5-100 machine (ION-TOF GmbH, Germany).

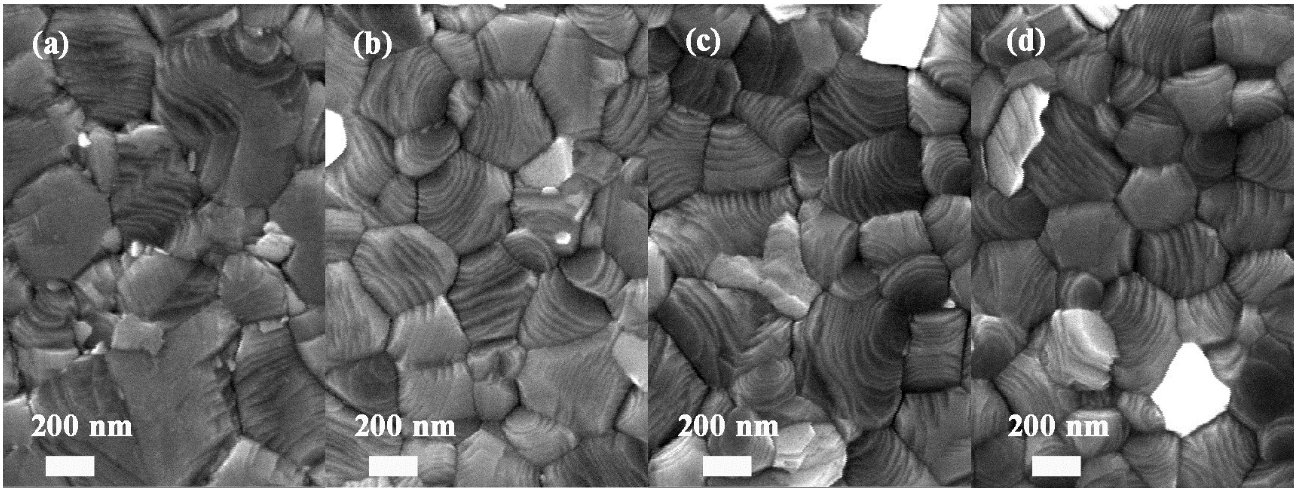

Figure S12. Top-view SEM images of (a) the control perovskite film and (b-d) the perovskite films incorporating different concentrations of artemisinin.

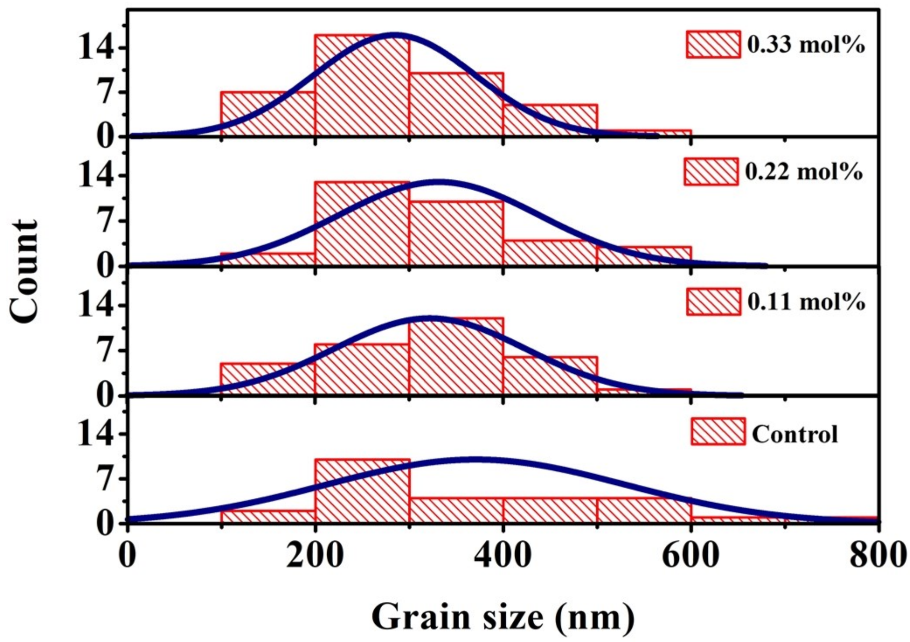

Figure S13. Histograms of the grain-size distributions for the control perovskite film and the perovskite films incorporating different concentrations of artemisinin.

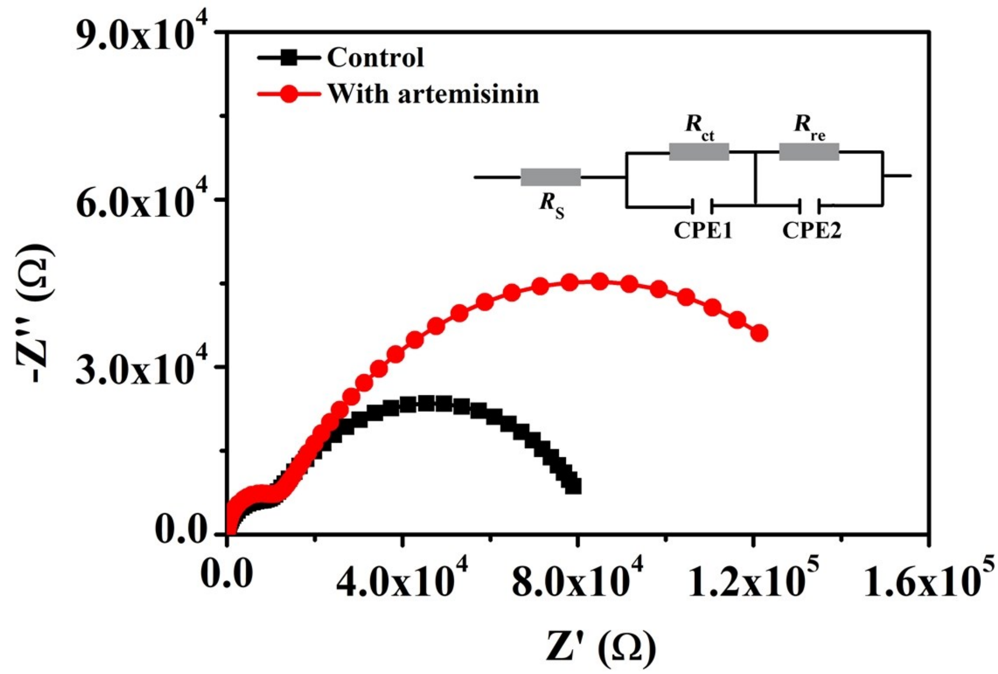

Figure S14. Typical Nyquist plots for a control flexible PSC and artemisinin-incorporated flexible PSC at a bias voltage (0.75 V) in the dark in the frequency range of 0.1 MHz to 1 Hz.

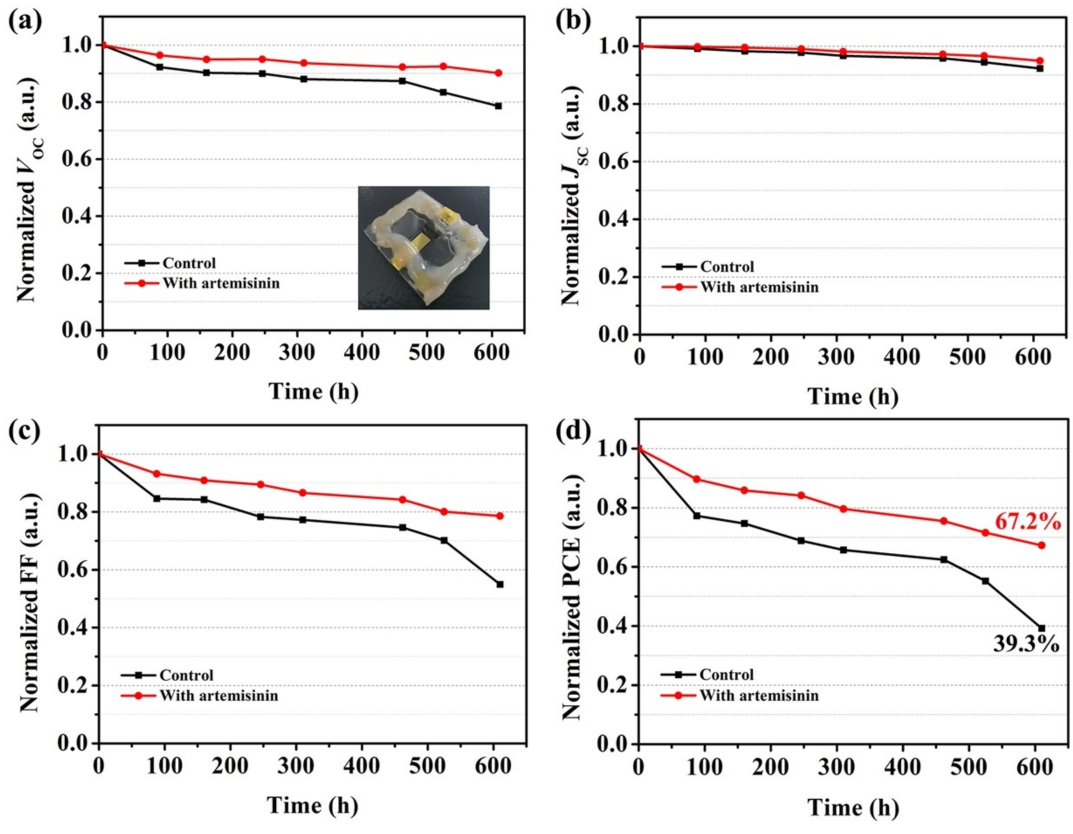

Figure S15. Dependence of the normalized average photovoltaic performance of flexible PSCs with and w/o artemisinin on aging time during damp heat testing at 85 °C/75% RH. The flexible devices were encapsulated based on a modified sealing process, according to a previous report. [2] The devices were first encapsulated with two ultra-thin glasses with a UV-glue, Norland optical adhesive NO68 (USA), and then their edges were sealed with another UV-glue, MORESCO MOISTURECUT WB90US (Japan), again. The devices were kept at 85 °C and 75% RH in a thermos-hygrostat in the dark. The inset in a) shows the photograph of one of the encapsulated devices. Five cells of control devices and artemisinin-doped devices were measured each time. PTAA with decreased additives was used as HTM. [2] The average initial photovoltaic parameters of the control devices are: V_{OC}-1.077 V, J_{SC}-21.61 mA/cm^2, FF-75.53%, and PCE-17.58%; those of the artemisinin-doped ones are: V_{OC}-1.094 V, J_{SC}-21.92 mA/cm^2, FF-77.67%, and PCE-18.63%.

References:

1. E. Sowade, H. Kang, K. Y. Mitra, O. J. Weiß, J. Weber and R. R. Baumann. *J. Mater. Chem. C*, 2015, **3**, 11815-11826.
2. T. Matsui,* T. Yamamoto, T. Nishihara, R. Morisawa, T. Yokoyama, T. Sekiguchi, and T. Negami. *Adv. Mater.*, 2019, **31**, 1806823.