

# **Simple and Dopant-free Hole-Transporting Material based on (2-ethylhexyl)-9H-Carbazole for Efficient Planar Perovskite Solar Cells**

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# 1 Materials

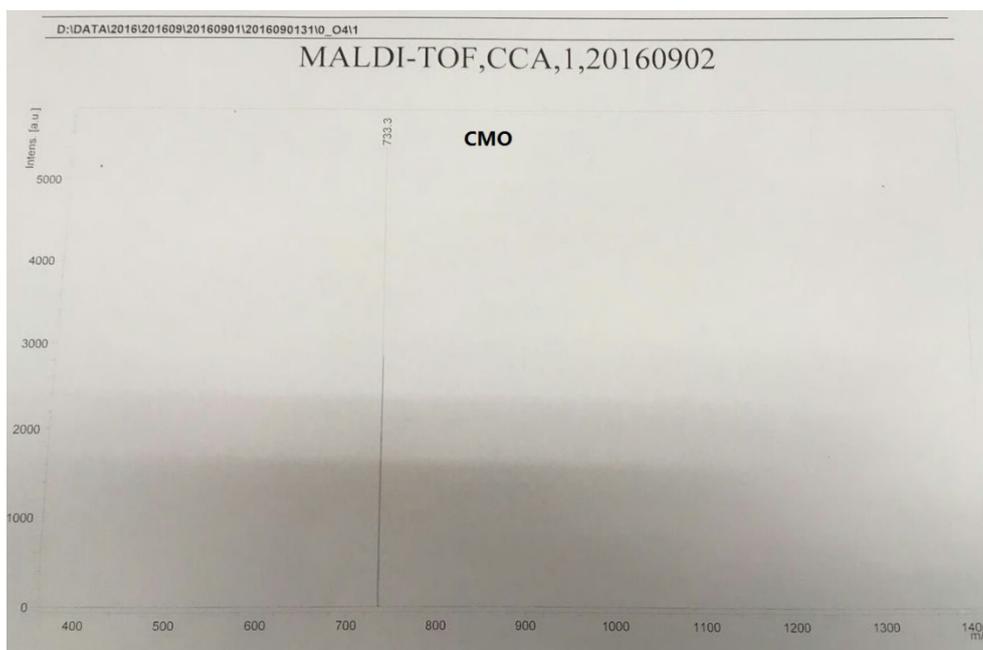


Fig. S1 MALDI-TOF spectrum of compound CMO.

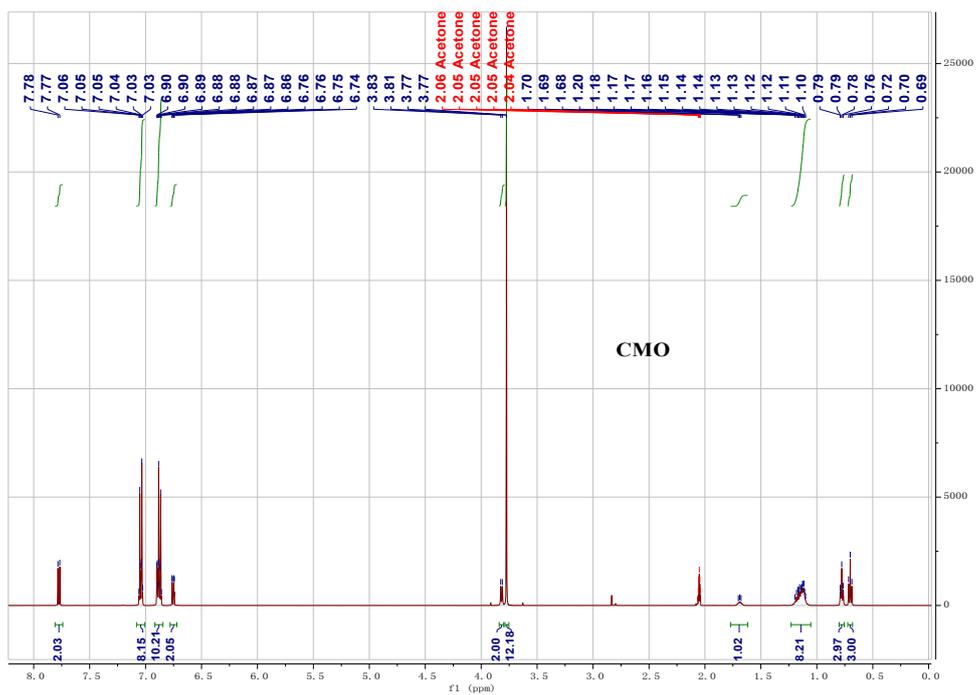
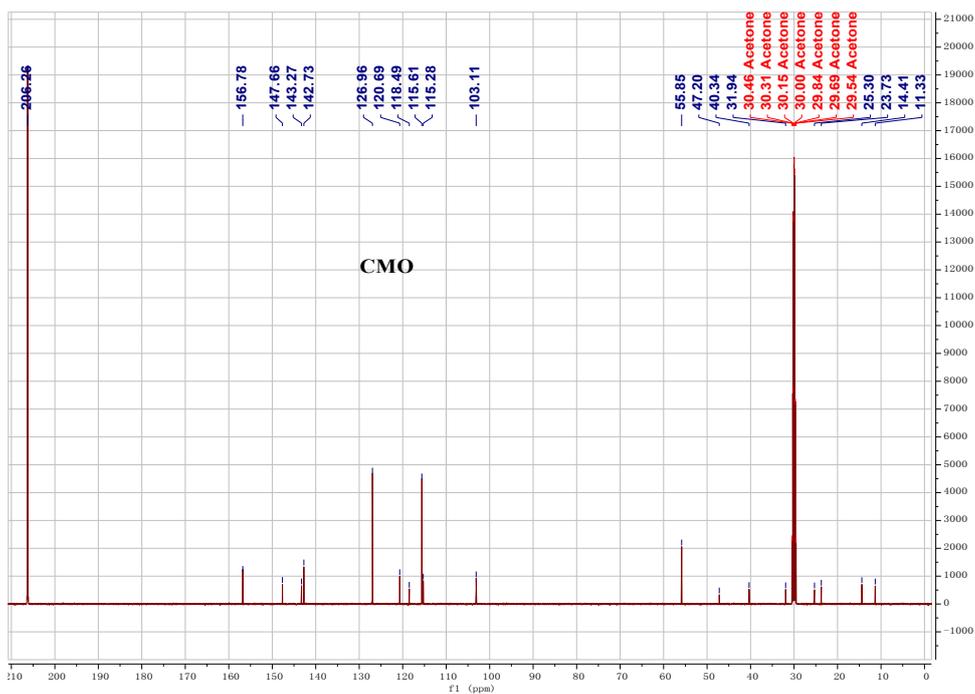


Fig. S2  $^1\text{H}$  NMR spectrum of compound CMO.



**Fig. S3**  $^{13}\text{C}$ NMR spectrum of compound CMO.

## 2 Methods

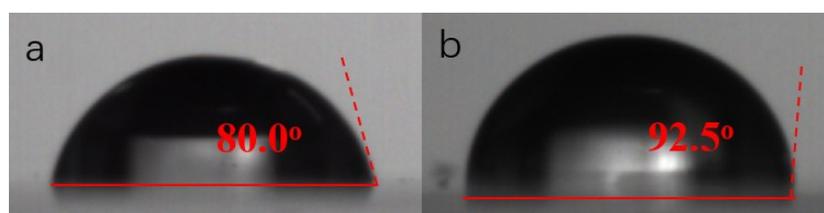
MALDI-TOF spectra, nuclear magnetic resonance (NMR) spectra, absorption spectra and the electrochemical cyclic voltammogram were recorded on Bruker BIFLEX III, Bruker DMX-500, Hitachi U-3010 UV-vis spectrophotometer and Zahner IM6e electrochemical workstation, respectively. Atomic force microscopy (AFM) images were captured on Multimode 8 micro-scope (Bruker, Santa Barbara, CA). Scanning electron microscopy (SEM) images were taken on S-4700, Hitachi. The photovoltaic characteristics are measured under  $100 \text{ mW cm}^{-2}$  (AM 1.5G) illumination using a Keithley Model 2400 multisource meter (Cleveland, OH, USA). A solar simulator (500 W Xe lamp) (ORIEL Solar 3A94023A, America) is employed as the light source. Steady-state photo-luminescence (PL) measurements were conducted on FLS980 (Edinburgh Instrument, UK). Time-resolved PL spectra were acquired on Life-spec

(Edinburgh Instrument, UK) by monitoring the signal at 785 nm excited with a 477 nm laser (2 MHz).

### 3 Fabrication of the pero-SCs devices

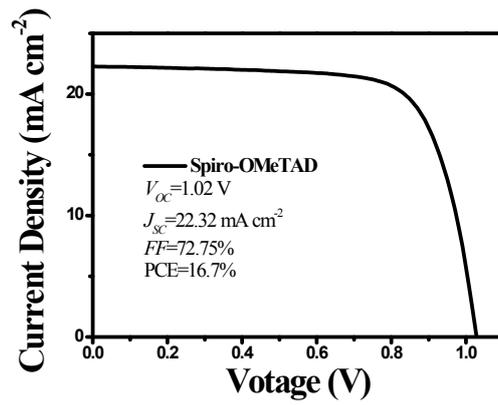
The fabrication of device was according to the previously reported procedure with some modification. Patterned FTO/glass substrates were ultrasonically cleaned with deionized water, acetone and ethanol respectively, and then followed by oxygen plasma cleaning for 10 min before use. FTO substrates were immersed into 200 mM  $\text{TiCl}_4$  aqueous solution for 60 min at 70°C and then washed with deionized water and ethanol. The spin-coating precursor solution was prepared by dissolving 0.191 g of MAI and 0.553 g of  $\text{PbI}_2$  in 1 mL of anhydrous DMF and DMSO (with a volume ratio of 4: 6). After the clean FTO substrates with compacted  $\text{TiO}_2$  were transferred into glove box, hole transport material CMO in CB was spin coated onto perovskite films at 5000 rpm for 30 s in glove box. Finally, 80 nm thick gold electrodes were deposited on the top of HTM by thermal evaporation at  $1.0 \times 10^{-4}$  Pa. For each solar cell, the active area was determined to be 0.07 cm<sup>2</sup>.

### 4 Water contact angles



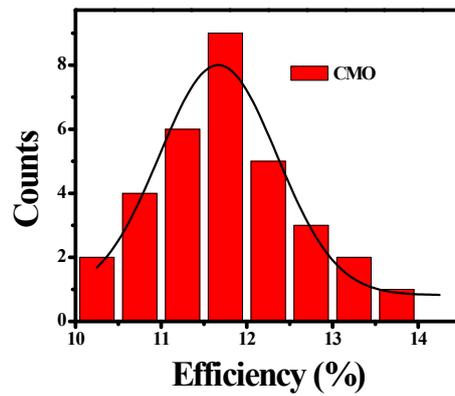
**Fig. S4** Water contact angles on (a) as-cast Spiro-OMeTAD, and (b) CMO films.

### 5 J–V curve of photovoltaic device for Spiro-OMeTAD



**Fig. S5** J–V curves of perov-SCs device based on Spiro-OMeTAD as HTM (72.3 mg mL<sup>-1</sup>) doping 4-tert-butylpyridine (28.8  $\mu$ L) and Li-TFSI/acetonitrile (17.5  $\mu$ L, 520 mg mL<sup>-1</sup>).

### 6 Histograms of 30 device PCEs



**Fig. S6** Histograms of 30 device PCEs based on CMO as HTM.