Graphite/Carbon Black Counter Electrode Deposition Methods to Improve the Efficiency and Stability of Hole-Transport-Layer-Free Perovskite Solar Cells

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ABSTRACT: The interfacial compatibility between the graphite/carbon black composite counter electrode (Gr/CB CE) and the perovskite layer is a crucial determinant of the performance of the hole-transport-layer-free carbon-based perovskite solar cells, and judicious selection of the Gr/CB CE application method is essential for achieving an optimum contact. In this work, three different types of Gr/CB CEs application methods are investigated: (1) deposition of Gr/CB on the fluorine-doped tin oxide (FTO) substrate, followed by clamping to the perovskite layer, (2) direct deposition of Gr/CB onto the perovskite layer, and (3) deposition of Gr/CB onto the PbI₂ precursor layer, followed by immersion in methylammonium iodide solution for the in situ conversion of PbI₂ to perovskite. The results revealed that Method 3 produced superior Gr/CB−perovskite contacts, resulting in up to 8.81% power conversion efficiency. The devices prepared using Method 3 also exhibited the best stability in the air, retaining 71.1% of their original efficiency after 1600 h of continuous testing. These results demonstrate that Gr/CB CEs can be considered excellent alternatives to the costly noble metals often employed in perovskite solar cells (PSCs) when deposited using a suitable technique.

1. INTRODUCTION

Organic−inorganic hybrid perovskite solar cells (PSCs) have received substantial attention in the photovoltaic research community owing to the remarkable optical and electrical properties of metal halide perovskites, such as high absorption coefficient, tunable band gap, low exciton binding energy, long carrier diffusion length, and high charge carrier mobility.¹−³ Due to these exceptional optoelectronic characteristics, the power conversion efficiency (PCE) of PSCs has exceeded 25.7%, surpassing the excellent PCE of 23.4% for copper indium gallium selenium solar cells and nearing the highest PCE of 27.6% for crystalline silicon solar cells.⁴

Despite remarkable progress in this emerging photovoltaic technology, some issues still hinder the commercialization of PSCs, such as the instability of perovskite contacts toward temperature, oxygen, and moisture in both fabrication and operation processes, high costs of hole-transport layer (HTL) material (i.e., spiro-OMeTAD, P3HT, and PTAA) and noble metal (i.e., Au and Ag) counter electrodes (CEs), and the high-energy-consumption device fabrication processes that are expensive and not favorable for large-scale production.⁶−¹⁰

To address these issues, alternative materials such as Al, Cu, ITO, PEDOT:PSS, and carbon can be used as CEs in PSCs.¹¹,¹² Carbon-based materials exhibit numerous merits because of their hydrophobic properties, chemical inertness, good electrical and thermal conductivity, water resistance, and excellent electrochemical stability.¹³ Carbon also has a work function of 5.0 eV, which is comparable to those of Au (5.1 eV) and Ag (4.6 eV).¹⁴ The most common carbon materials used in the fabrication of PSCs are carbon black,¹⁵ carbon nanotubes,¹⁶ carbon ink,¹⁷ commercial carbon paste,¹⁸,¹⁹ spongy carbon,²⁰ and graphite.²¹

Although carbon-based PSCs (C-PSCs) have made great progress, their exceptional efficiency of up to 16% is significantly lower than that of traditional PSCs.²²,²³ This is primarily due to insufficient contact between the perovskite and the carbon interface caused by the postdeposition process of the carbon CE. Pinholes are unavoidable at the perovskite/carbon contact because the carbon paste is made up of graphite and carbon sphere. The lamella structure of the graphite layer tends to produce interspace between the graphite flakes and perovskites, resulting in poor contact at the perovskite/carbon interface.¹² This causes serious photocarrier recombination and reduces the fill factor significantly.

Received: April 24, 2022
Accepted: June 3, 2022
Published: June 21, 2022
In this work, we compare the performance and durability of C-PSCs prepared using three different approaches to deposit the graphite/carbon black (Gr/CB) CE: (1) deposition of Gr/CB on a fluorine-doped tin oxide substrate first and then clamping on the top surface of perovskite layer; (2) deposition of Gr/CB directly on the perovskite layer, and (3) deposition of Gr/CB onto the PbI₂ precursor layer, followed by soaking in a methylammonium iodide solution for in situ PbI₂ transformation to perovskite. These experiments aimed to better understand and optimize the contact between Gr/CB CE and the perovskite layer, which is crucial for efficient hole extraction and proper electron blocking. The mass fraction of graphite flakes in the Gr/CB composite was also optimized to attain high conductivity, low sheet resistance, crack-free, and highly adhesive CEs, which significantly influences the connection between the Gr/CB CE and the perovskite layer. The photovoltaic parameters and interfacial charge-transfer kinetics were investigated by current density−voltage measurements and electrochemical impedance spectroscopy, respectively, to find the optimum method for Gr/CB deposition and obtain highly efficient and stable C-PSCs.

2. EXPERIMENTAL SECTION

2.1. Materials. Titanium dioxide (TiO₂, 90-NRT, Dyesol), absolute ethanol (>99%, Sigma-Aldrich), α-terpineol (90% technical grade, Sigma-Aldrich), graphite powder (<20 μm, synthetic, Sigma-Aldrich), acetylene carbon black (100% compressed, Strem chemicals), lead iodide (PbI₂, 99%, Dyesol), dimethyl sulfoxide (DMSO, 99.7%), N,N-dimethyl formamide (DMF, 99.7%), titanium(IV) isopropoxide (TTIP, 99%, Sigma-Aldrich), glacial acetic acid (Sigma-Aldrich), and methylammonium iodide (MAI, 99%, Sigma-Aldrich) were used without further purification. Fluorine-doped tin oxide coated glass (FTO; sheet resistance 12 Ω/sq, Solaronix) substrates were used for all cells.

2.2. Preparation of Counter Electrodes. The carbon paste was first prepared by mixing five different mass fractions of graphite flakes (60, 70, 80, 90, and 100 wt %) in a carbon black powder using mortar and pestle. Then, 10 mL of α-terpineol was added and ground for 20 min. Finally, 1 mL of TTIP and 100 μL of glacial acetic acid were added to the mixture and then ball-milled for 10 h. The CEs prepared from Gr/CB composites with graphite mass fractions of 60, 70, 80, 90, and 100 wt % were referred to as G60, G70, G80, G90, and G100, respectively.

2.3. Device Fabrication. FTO substrates were washed with detergent solution, water, acetone, and ethanol in sequence for 20 min each in an ultrasonic cleaner. The compact TiO₂ layer was fabricated on the FTO substrate by spin-coating a mildly acidic solution of TTIP and HCl diluted in ethanol at 3000 rpm for 30 s and annealed at 500 °C for 30 min. Subsequently, the mesoporous TiO₂ layer was then deposited onto the compact TiO₂ layer by spin-coating a solution of the TiO₂ paste diluted in ethanol (1:3.5 mass ratio) at 5000 rpm for 30 s, followed by heating at 130 °C for 10 min and 500 °C for 30 min. Next, 1 M PbI₂ precursor solution (462 mg of PbI₂ dissolved in 800 μL of DMF and 200 μL of DMSO) was spin-coated on the mesoporous TiO₂ layer at 3000 rpm for 30 s and then dried at 70 °C for 30 min. After completing the above steps, the perovskite coating and the Gr/CB CE deposition were fabricated in three different approaches, as illustrated in Figure 1.

For Method 1, a PbI₂ film was immersed in the MAI solution (10 mg/mL in IPA) for 30 min. The film immediately changed from yellow to brown, indicating the perovskite formation. The film was then rinsed with IPA to remove
ammonium salt residues in the perovskite film before annealing at 70 °C for 30 min on a preheated hot plate under the ambient atmosphere. A prefabricated Gr/CB CE on FTO was then directly clamped onto the above perovskite film to complete device fabrication.

For Method 2, the perovskite film was formed according to the same procedure given in Method 1. Once the perovskite film cooled to room temperature, a Gr/CB CE was deposited onto the film by doctor-blading, followed by drying at 70 °C for 30 min.

For Method 3, the Gr/CB CE was directly doctor-bladed onto the PbI2 layer, dried at 70 °C for 30 min, and then cooled to room temperature. The film was then immersed in a solution of the MAI solution for 30 min to convert PbI2 to perovskite before being rinsed with IPA and dried in air at 70 °C.

2.4. Characterization and Measurements. A surface profiler (KLA-tencor) was used to determine the thickness of Gr/CB CE films. The square resistances of the Gr/CB CE were measured using a four-point probe resistivity measurement device (4PP, model DFP-02). The structural and top-view surface morphologies of Gr/CB CEs were characterized by scanning electron microscopy (FE-SEM, model JOEL JSM-7610F). The crystal structure of the prepared films was examined by X-ray diffraction (XRD; MAXima-X 7000, Cu Kα radiation). To measure the PSC performance, the photocurrent–voltage (J–V) was measured using a source meter (Keithley 2400) under simulated AM1.5G, 100 mW/cm² illumination with an active area of 0.3 cm². Electrochemical impedance spectroscopy (EIS) measurements of the devices in the dark were performed using an electrochemical workstation (CompactStat, Ivium) over the frequency range of 0.1 Hz to 1 MHz with a 10 mV modulation amplitude. The in-plane I–V measurements of symmetric carbon/perovskite/carbon devices were performed using the same electrochemical workstation in a two-electrode configuration. X-ray photoemission spectroscopy (XPS) was performed at BL3.2Ua in the Synchrotron Light Research Institute (SLRI), Thailand. The energy of electrons emitted from the sample was analyzed by the concentric hemispherical analyzer (CLAM2, Thermo VG Scientific), 20° from the surface normal. The binding energy was calibrated to the C 1s peak at 284.6 eV. A contact angle measurement was used to determine the contact angles of water on the Gr/CB CE surface (Image).

3. RESULTS AND DISCUSSION

3.1. Structure of Carbon-Based Perovskite Solar Cells. Figure 2a shows the schematic of the C-PSC structure. The devices comprised an FTO substrate, compact TiO2 layer, mesoporous TiO2 layer, perovskite light absorber layer, and carbon layer as the CE. The energy-level diagram of a C-PSC device is shown in Figure 2b. Under illumination, the perovskite absorber layer becomes excited, and the generated excitons rapidly dissociate into free conduction band (CB) electrons and valence band (VB) holes. The free electrons are transferred from the CB of the perovskite (−3.9 eV) to the CB of TiO2 (−4.2 eV) before being finally collected at the FTO substrate. Meanwhile, the holes are transferred from the VB of...
the perovskite (−5.4 eV) to the carbon CE (−5.0 eV).24,25 Here, the perovskite material is used as a light harvester and an electron/hole-transport layer.

3.2. Four-Point Probe Measurements. The sheet resistance, resistivity, and conductivity of the fabricated Gr/CB composite CEs were investigated using four-point probe measurements. The electrode sheet resistance was calculated using

\[ R_s = \left( \frac{\pi}{\ln 2} \right) \frac{V}{I} \]  

(1)

where \( V \) is the change in voltage across the probes and \( I \) is the current delivered.

The graphite to carbon black mass fractions was varied to optimize the conductivity and achieve low sheet resistance. The thickness of the tested samples is ∼60 μm. As shown in Supporting Figure S1, pure graphite (G100) exhibits the highest resistivity among graphite-containing samples, which may be due to the large particle size of graphite flake, resulting in poor particle connection in the film. With the addition of carbon black particles, the sheet resistance and resistivity decrease dramatically. The resistivity of carbon CE G60, G70, G80, G90, and G100 are 149, 157, 220, 229, and 479 Ω-μm, respectively. According to our previous study,26,27 graphite flakes are the primary pathway for charge transport, while carbon black acts as a significant conductive interparticle conduit for graphite. Carbon black partly fills the interparticle spaces between the large graphite flakes, facilitating good electrical contact and lowering the resistance of Gr/CB CEs.

3.3. Scanning Electron Microscopy (SEM). Representative scanning electron microscopy (SEM) images of the perovskite and Gr/CB CE are shown in Figure 3. Figure 3a shows the top-view SEM image of the perovskite film obtained after dipping PbI₂ film into the MAI/IPA solution for 30 min. The surface of the perovskite film exhibits distinct tetragonal morphology with excellent uniformity size crystals and smooth surface coverage as revealed under high magnification (inset of Figure 3a), and the average crystallite size is around 500–600 nm. In the production of Gr/CB CEs, the Gr/CB connectivity and compactness are critical for improving hole extraction and lowering contact resistance in solar cells. As shown in Figure 3b, large graphite flakes are bound to each other and well surrounded with carbon black particles to form the Gr/CB CE with TiO₂ as binders. Graphite flakes provide good electronic conductivity for the Gr/CB CEs, while the carbon black nanoparticles act as conductive fillers and bridge the gaps among the graphite flakes.28 The increase in the conductivity of the Gr/CB CEs is caused by the excellent interaction between graphite and carbon black. As a result of the well-distributed carbon black particles, the interfacial contact between perovskite layers and Gr/CB CEs improves. The surface morphologies of G60, G70, G80, G90, and G100 are shown in Supporting Figure S2a–e, respectively.

3.4. X-ray Photoelectron Spectroscopy (XPS). X-ray photoelectron spectroscopy (XPS) was performed to analyze
the elemental composition and chemical states in the prepared Gr/CB composite. The high-resolution C 1s, O 1s, and Ti 2p XPS spectra of Gr/CB are presented in Figure 4. The results depicted in Figure 4a show the recorded C 1s spectra from 278 to 294 eV, over which five peaks were detected. The two dominant peaks centered at 284.3 eV and 285.4 eV derived from the sp²-C and sp³-C hybridized carbon atoms, accompanied by three minor peaks at 286.5, 288, and 290.6 eV, which correspond to hydroxyl or epoxy groups, carbonyl, and shake-up π→π* satellite peaks, respectively. In the O 1s XPS spectra shown in Figure 4b, two peaks can be identified: one at 531.5 eV and the other at 533.2 eV. The first peak is attributed to the C–OH groups and the second to H2O. The Ti 2p XPS spectrum in Figure 4c shows two prominent peaks at binding energies around 459.5 eV for Ti 2p3/2 and 465.2 eV for Ti 2p1/2 respectively, which are approximately close to the values of the Ti4+ valence state of stoichiometric TiO2. The chemical analysis of C 1s and O 1s regions of the Gr/CB composite can be found in Supporting Table S1.

3.5. X-ray Diffraction (XRD). X-ray diffraction (XRD) technique was carried out to characterize the crystal structures of components of each layer in the device. The XRD patterns of FTO/TiO2/perovskite/carbon, FTO/TiO2/perovskite, and FTO/TiO2 films are shown in Figure 5. Diffraction peaks of perovskite were observed at 2θ values of 14.14, 19.92, 23.54, 24.52, 28.48, 31.92, 34.62, 40.68, and 43.16°, which corresponded to the (110), (112), (211), (202), (220), (310), (224), and (314) planes, respectively. The presence of these peaks indicated the formation of a tetragonal phase in the perovskite crystal structure in the samples. A very sharp peak could be observed at around 26.64°, corresponding to graphite’s basal (002) diffraction peak. Compared to FTO/TiO2/perovskite, the FTO/TiO2 film only showed a small peak of TiO2 at 25.5°, which represented the tetragonal anatase (101) structure of the mesoporous. The diffraction peaks (marked with asterisks) at 26.54, 33.74, 37.8, 51.78, 62.08, and 65.56° were attributed to the FTO substrate.25

3.6. Photovoltaic Density of C-PSCs. Figure 6 shows the photocurrent-voltage (J–V) characteristics of the C-PSC devices prepared using three Gr/CB CE assembly methods (labeled M1, M2, and M3 for methods 1, 2, and 3, respectively), and the derived photovoltaic parameters are given in Table 1. The results revealed that different Gr/CB layer deposition methods have a significant effect on the overall PCE.

![Figure 5. X-ray diffraction patterns of FTO/TiO2/perovskite/carbon, FTO/TiO2/perovskite, and FTO/TiO2.](https://doi.org/10.1021/acsomega.2c02555)

![Figure 6. J–V curves of C-PSCs prepared using different Gr/CB CE assembly methods under simulated AM1.5G, 1 Sun illumination.](https://doi.org/10.1021/acsomega.2c02555)

Table 1. Photovoltaic Performance Parameters of the Fabricated Devices

| device | η (%) | FF | Voc (V) | Jsc (mA/cm²) |
|--------|-------|----|---------|--------------|
| M1     | 1.47  | 0.34 | 0.73    | 6.02         |
| M2     | 8.15  | 0.45 | 0.77    | 23.2         |
| M3     | 8.81  | 0.47 | 0.78    | 24.1         |

The M1 device achieves the lowest photovoltaic performance with an open-circuit voltage (Voc) of 0.73 V, a current density (Jsc) of 6.02 mA/cm², a fill factor (FF) of 0.34, and a PCE of 1.47%. This low performance is due to the significant internal resistance across the interface between the perovskite and the Gr/CB CE. In contrast, M2 exhibited highly enhanced performance, with Voc of 0.78 V, Jsc increasing to 23.2 mA/cm², and FF of 0.45, respectively, yielding an enhanced PCE of 8.15%. The direct deposition of Gr/CB onto the perovskite facilitates better contact between them, minimizes parasitic electrical resistance, and enhances device performance. The J–V results reveal that M3 performs even better than M2, with a Voc of 0.78 V, Jsc of 24.1 mA/cm², and FF of 0.47, yielding an excellent PCE of 8.81%. The M3 device exhibited a higher Jsc due to the light scattering by large-sized perovskite cuboids and also due to the enhanced charge carrier mobility as a result of the reduced electron–hole pair recombination. The Gr/CB CE plays an important role in facilitating effective charge extraction/injection at the Gr/CB–perovskite interface, improving both the efficiency and stability of the fabricated devices. By depositing the Gr/CB CE on the PbI2 layer
first, followed by immersion in the MAI solution for the in situ conversion of PbI₂ to perovskite, the Gr/CB CE becomes chemically embedded into the perovskite at the interface. This improves the interfacial contact between the perovskite and the Gr/CB CE, which results in efficient hole extraction.

Some preliminary photovoltaic experiments on Gr/CB devices were conducted to evaluate the performance of Gr/CB CEs comprising various mass fractions of graphite to carbon black. The J−V characteristics for devices G60, G70, G80, G90, and G100, respectively, are shown in Table S2. Pure graphite had the lowest PCE, indicating poor photovoltaic performance due to the poor interfacial contacts between the perovskite layer and the graphite flakes. The device performance improved significantly when carbon black particles were introduced. The higher performance of the G70 device could be attributed to the more contact sites in the Gr/CB composite CEs than in the pure graphite-based CE, which was important for efficient PSCs. However, increasing the carbon black content resulted in a slightly decreased device performance because of the higher surface area and greater recombination caused by the carbon black particles.

3.7. In-Plane J−V Measurements of Symmetric Carbon/Perovskite/Carbon Devices. To investigate charge transfer at the Gr/CB−perovskite interface in isolation from the FTO/TiO₂ and TiO₂/perovskite interfaces, we measured the dark J−V characteristics of symmetric carbon/perovskite/carbon in-plane devices prepared using Methods 2 and 3. We also performed the exact measurements on asymmetric carbon/FTO/carbon devices for comparison. As shown in Supporting Figure S3, all devices exhibit linear J−V characteristics, indicating ohmic contacts, and the Method 3 device demonstrates a higher current compared to Method 2. The reduced resistance implies better charge transfer across the Gr/CB−perovskite interface, which is consistent with the superior photovoltaic parameters of Method 3.

3.8. Electrochemical Impedance Spectroscopy (EIS). Electrochemical impedance spectroscopy (EIS) was performed to investigate the dynamic response of the fabricated devices, especially the interfacial charge-transfer characteristics occurring across the interface between the Gr/CB CE and the perovskite film. Spectra were recorded in the dark at a bias voltage of −0.8 V (chosen because it is similar to Vₜₒₜ of the devices), and the results are shown in Figure 7a.

Two semicircles can be identified in each Nyquist plot: a large arc at low frequency and a small arc at high frequency. The data are well fitted by the equivalent circuit shown in Figure 7a, which contains a series resistor (Rₛ), two parallel RC elements comprising resistors R₁ and R₂, and capacitors C₁ and C₂. The values of the fitted parameters are given in Table 2.

The reduction in R₁ in the order M1 > M2 > M3, indicating that the quality of the Gr/CB−perovskite contact increases on moving from Method 1 to Method 3, which is consistent with the observed improvement in fill factor. Assigning the other circuit elements to particular interfaces or physical processes is not straightforward. There is still some disagreement in the literature about the correct equivalent circuit model for perovskite solar cells.

For low perovskite loading, devices tend to behave like sensitized solar cells. The high-frequency arc is due to electronic charge accumulation and transfer at the external contacts (e.g., the counter electrode). The low-frequency arc arises from recombination at the TiO₂−perovskite interface, in which case R₁ can be identified as contact resistance and R₂ as recombination resistance. Following this interpretation, the decrease in R₁ in the order M1 > M2 > M3 can reduce the Gr/CB−perovskite contact resistance due to an improved interface quality, consistent with the fill factor trend. The increase in Rₛ can then be interpreted as an increasing recombination resistance, which could be explained by reduced surface recombination at the Gr/CB contact due to a less defective interface, and is consistent with the increase in Vₑₒₑ.

On the other hand, for higher perovskite loading, the high-frequency arc is thought to arise from a resistance associated with transport and recombination in the bulk perovskite layer.
in parallel with the geometric capacitance of the device, and the low-frequency arc is due to ionic and electronic charge accumulation at the contacts.\textsuperscript{38} It may not even be possible to assign a unique physical process to each arc in intermediate cases.\textsuperscript{35} We, therefore, performed additional EIS measurements on devices with two coplanar CEs (CCE) (Figure 7b), so we could focus on the Gr/CB perovskite interface without needing to consider the FTO–TiO\textsubscript{2} or TiO\textsubscript{2}–perovskite interfaces in our analysis. These EIS measurements were performed for the CEs fabricated using Methods 2 and 3, and the corresponding devices were labeled as CCE-M2 and CCE-M3. Only one semicircle is observed for these devices, which can be well fitted by a series resistor and a single RC element. Results of these fits are also given in Table 2. The magnitude of \( R_1 \) is smaller than observed for regular devices, which we tentatively attribute to the different electrode geometry and a barrier caused by one of the two Gr/CB–perovskite interfaces being reverse biased, which does not occur in the regular device. We also note that similar total resistances have been observed in coplanar Ag–perovskite–Ag devices.\textsuperscript{39} Despite the smaller resistance values, there is a clear trend of decreasing \( R_1 \) and \( R_t \) in the order CCE-M2 > CCE-M3, as found for the regular devices. Therefore, we conclude that \( R_1 \) can be associated with the Gr/CB–perovskite contact in traditional and coplanar devices. Therefore, Method 3 results in a superior interfacial contact compared with Methods 1 and 2.

### 3.9. Stability Analysis

Device stability under ambient conditions is an essential consideration during the development of PSCs, with moisture being one of the major issues causing device degradation. Gr/CB CEs can act as a protective layer to prevent water in the air from penetrating the perovskite layer. We tested the stability of devices fabricated using M2 and M3 stored under ambient conditions without any encapsulation. The devices were tested periodically over a total duration of 1600 h. The detailed photovoltaic parameters are plotted in Figure 8.

The devices fabricated using Method 3 exhibit a much slower degradation than those fabricated using Method 2. The \( V_{oc} \) and FF of M2 and M3 remain almost stable over the entire testing period, whereas \( J_{sc} \) started to decay after a specific time, causing the efficiency to decrease. The \( J_{sc} \) of M3 remained virtually constant during the first 1000 h, whereas M2 was nearly unchanged during the first 800 h. However, once they reached a critical point in time, the efficiency and \( J_{sc} \) started to decline due to the decomposition of the perovskite. The decay of \( J_{sc} \) for M3 is much slower than that of M2, and similar behavior is observed for the PCE. From the initial point until the end of the test, the PCE of M3 decreased by 28.9%, while that of M2 decreased by a much larger 47.6%.

This is mostly due to the chemically stable and highly hydrophobic nature of carbon (the contact angle of water directly dropped on the Gr/CB CE is approximately 114.8°, as shown in Figure S4), which protects the perovskite from humidity degradation.\textsuperscript{5,40,41} However, the effectiveness of the carbon layer in preventing moisture ingress depends on the method of application, with M3 being superior. This study indicates the underlying potential of Gr/CB CEs and demonstrates that the application method is crucial to obtaining stable, high-efficiency PSCs.
4. CONCLUSIONS

In summary, we have compared various Gr/CB CE deposition strategies for assembling carbon-based perovskite solar cells with improved Gr/CB—perovskite interfacial contact. Deposition of the Gr/CB CE onto the PbI$_2$ precursor layer, followed by immersion in the MAI solution for the in situ conversion from PbI$_2$ to perovskite (Method 3), was the most promising method. It allows chemical embedding of the Gr/CB layer into the perovskite layer. This substantially improves the contact between the perovskite and the Gr/CB CE, as evidenced by decreases in both series resistance and interfacial charge-transfer resistance. An optimized PCE of 8.81% and good long-term stability (71.1% retention of original efficiency after 1600 h) under ambient conditions were obtained for the assembled C-PSCs. These results demonstrate the great potential of using Gr/CB CEs to eliminate expensive noble metals, hole-transport layer, and costly high-vacuum deposition methods in the assembly of PSCs. Although a high current density and an acceptable open voltage were achieved, the low FF limited the PCE of the devices. Further attempts are necessary to reduce the series resistance of the devices by improving the carbon/perovskite contact and the transport properties of the absorber layer.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c02555.

Supporting Information includes a four-point probe, SEM, XPS analyses, photovoltaic parameters, J–V curves, in-plane devices, and contact angle (PDF).

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Author Contributions

The manuscript was written through the contributions of all authors. All authors have approved the final version of the manuscript.

**Notes**

The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

Universiti Brunei Darussalam Research Grant (UBD/RSCF/1.4/FICBF(b)/2022/040) is acknowledged for the financial support.

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