Low Temperature Processed Atomically Thin Perovskite Oxide as Electron Transporting Layer in Perovskite Solar Cells

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Abstract

Perovskite solar cells with high performances typically use anatase TiO₂ as electron transporting layers (ETLs); however, they require high temperature sintering up to 500°C. Here, we demonstrate the atomically thin perovskite oxide Ca₂Nb₃O₁₀ (CNO) as an alternative ETL. The CNO layers can be deposited by low-temperature solution process and the resulting cell performances can be as high as 11.56%, which makes CNO a highly promising material as ETLs.

1. Introduction

Organic-inorganic lead halide (CH₃NH₃PbX₃, X=Cl, Br, or I) perovskite solar cells (PVSCs) have attracted huge attention with its remarkably increasing in power conversion efficiency to 22.1% within few years. The organic-inorganic lead halide perovskite has superior properties such as low temperature processability, high absorption coefficient, long carrier diffusion lengths, and low binding energies of electron-hole pairs. The conventional PVSCs usually consist of electron transporting layers and hole transporting layers (HTLs) to extract electrons and holes to the electrodes. Typically, compact TiO₂ is used in the planar heterojunction PVSCs as ETLs. However, high temperature sintering (~500°C) is required to attain an anatase phase TiO₂ for better charge transferring ability and this may limit the usages of flexible substrates. [1-4]

In this work, we demonstrate a two-dimensional atomically thin perovskite oxide $Ca_2Nb_3O_{10}$ as ETL in PVSCs. The CNO atomic sheets suspension were prepared by chemical exfoliation of layered perovskite oxide (KCa₂Nb₃O₁₀) under room temperature and the thickness of the sheets is only ~1.5nm (Fig. 1a).



Fig.1 (a) Structure of $Ca_2Nb_3O_{10}$ atomic sheets. (b) Device architecture of the FTO/CNO/perovskite/Spiro-OMeTAD/Au solar cells tested in this study.

The CNO atomic sheets monolayer were deposited onto the FTO glass layered-by-layered with Langmuir-Blodgett (LB) deposition method. Then the perovskite and Spiro-OMeTAD HTL were spin coated onto the CNO respectively and finally the gold top electrode was deposited by thermal evaporation (Fig. 1b). The whole process could be manipulated under 120°C.

2. Results and discussion

The photovoltaic characteristics of the devices were measured under simulated A.M. 1.5 illumination with intensity of 100mW/cm^2 . The current density-voltage curves and the corresponding values of open-circuit voltage (V_{oc}), short-circuit current density (J_{sc}), fill factor (FF), and power conversion efficiency (PCE) are shown in Fig. 2 and Table I.



Fig. 2 Current density-voltage curves of devices with different electron transporting layers. 1L, 3L, 5L, and 10L CNO indicates the repeating LB process for deposited CNO 1, 3, 5, and 10 times.

For devices with 1L and 3L CNO, the PCE were relatively low with 9.36% and 10.29%, resulting from the incomplete coverage of CNO layers (Fig. 3a and b). Devices with 5L CNO had superior PCE of 11.56% for the fully coverage of CNO film, which is shown in Fig. 3c. However, despite the complete coverage in 10L CNO (Fig. 3d), the slightly decreased in PCE of 10.68% was observed in these devices resulting from the lower conductivity with increasing ETL thickness.

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ETL	V _{oc} (V)	J _{sc} (mA/cm²)	FF (%)	PCE _{best} (%)	PCE _{avg} (%)
No ETL	0.92	14.72	35.71	4.84	3.62±0.99
TiO ₂	0.95	19.27	68.28	12.44	12.18±0.24
1L CNO	0.85	18.97	58.19	9.36	8.83±0.80
3L CNO	0.87	19.23	61.3	10.29	9.37±0.60
5L CNO	0.97	20.9	56.95	11.56	11.08±0.44
10L CNO	0.96	20.08	55.59	10.68	9.58±0.58

Table I Characteristics of perovskite solar cells with different electron transporting layers

In addition, we also compared the CNO devices with devices without ETL and devices with conventional compact TiO₂ ETLs. Devices without ETL had low PCE of 4.84% due to the inefficient electron transporting ability and strong carrier recombination. With the assist of 5L CNO, the resulting PCE was almost compatible to the typical TiO₂ devices of 12.44%. The results showed that our atomically thin CNO layers are very promising as electron transporting layers to achieve low temperature processed PVSCs.



Fig. 3 SEM top view of (a) 1L, (b) 3L, (c) 5L, and (d) 10L CNO film on FTO glass.

3. Conclusions

In this work, we demonstrated a two dimensional atomically thin layered perovskite oxide CNO as a new electron transporting layer in perovskite solar cells. The CNO film can be deposited with room temperature LB process and achieve complete coverage onto the FTO substrates. The resulting device performances can be greatly improved and is compatible to the performances for devices with conventional TiO_2 electron transporting layer.

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References

- Best Research-Cell Efficiencies. NREL 2017; Available from: <u>https://www.nrel.gov/pv/assets/images/efficiency-chart.png</u>.
- [2] G.C. Papavassiliou: Molecular Crystals and Liquid Crystals. 286 [1](1996)231.
- [3] G. Xing, N. Mathews, S. Sun, S.S. Lim, Y.M. Lam, M. Grätzel, S. Mhaisalkar and T.C. Sum: Science. **342** [6156](2013)344.
- [4] V. D'Innocenzo, G. Grancini, M.J. Alcocer, A.R. Kandada, S.D. Stranks, M.M. Lee, G. Lanzani, H.J. Snaith and A. Petrozza: Nat Commun. 5 (2014)3586.