

Interphase Engineering for Ambient Energy Harvesting: From Coordination Polymers to High-Efficiency Photocapacitors and Hybrid Photovoltaics

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Abstract

The transition toward autonomous Internet of Things (IoT) devices demands sustainable power sources capable of efficient energy harvesting and storage under ambient illumination. Achieving high performance requires precise control over interphase properties at multiple device interfaces, a challenge addressed through systematic molecular and material engineering across photovoltaic, charge transport, and energy storage components.[1,2]

At the perovskite/electron transport layer interface, the molecular nature of conventional buckminsterfullerene (C_{60}) layers has been identified as a critical limitation, leading to weak interfaces, loose packing, and susceptibility to degradation. A C_{60} -based ionic salt, CPMaC, was synthesized to address these constraints. The $CH_2-NH_3^+$ head group enables ionic bonding with the perovskite surface through formamidinium vacancy occupation, producing a threefold increase in interfacial toughness (1.43 J m^{-2} vs 0.50 J m^{-2} for C_{60}). Inverted perovskite solar cells incorporating CPMaC achieve power conversion efficiencies of approximately 26% with only ~2% degradation after 2100 hours of continuous operation at 65°C under 1-sun illumination. Minimodules demonstrate 23% efficiency with less degradation. Further advances in interphase chemistry have been realised through fluorinated self-polymerizable additives, which form hydrophobic dipole layers at the perovskite interface, enhancing charge extraction and stability while yielding champion efficiencies of 26.01% with 90% retention after 840 hours at 85°C . [3,4,5]

The importance of interphase engineering extends to hole transport materials, where mixed-valence copper coordination polymers have been investigated as sustainable alternatives to heavily doped organic semiconductors. One-dimensional polymer chains linked through $(Cu^I I_2 X_2)$ copper halide rhombi exhibit band-like transport at room temperature, with iodide-bridged coordination polymers achieving conductivities of 1 mS cm^{-1} and hole mobilities of $5.8 \times 10^{-4} \text{ cm}^2 (\text{V s})^{-1}$. Density functional theory calculations reveal an effective hole mass of $6 m_e$, with charge transport governed by copper-halide conjugation along polymer chains. Integration into carbon-based perovskite solar cells yields high and stable power conversion efficiency with enhanced thermal stability.[6]

These interphase engineering principles have been applied to develop three-terminal photocapacitors integrating high-efficiency dye-sensitized solar cells with polyviologen-based asymmetric supercapacitors for ambient energy harvesting and storage . Under 1000 lux indoor illumination, photocharging voltages of 920 mV are achieved with power conversion efficiencies exceeding 30% and overall photocharging efficiencies up to 18% . Polyviologen electrodes exhibit low reorganization energies , enabling efficient charge transfer and exceptional cycling stability with full capacitance retention after 3000 charge-discharge cycles . The integrated photocapacitor system successfully powers multilayer IoT networks at 500 lux for 72 hours, demonstrating 3.5-fold superior inference throughput compared to commercial amorphous-silicon modules while achieving 93% accuracy on CIFAR-10 classification tasks with an energy requirement of only 0.81 mJ per inference. [7]

The convergence of coordination polymer chemistry, ionic interface engineering, and device architecture optimization establishes a comprehensive framework for developing self-sustaining energy systems.

1. Flores-Diaz, N.; De Rossi, F.; Keller, T.; Morritt, H.; Perez Bassart, Z.; Lopez-Rubio, A.; Fabra, M. J.; Freitag, R.; Gagliardi, A.; Fasulo, F.; Muñoz-García, A. B.; Pavone, M.; Javanbakht Lomeri, H.; Sanchez Alonso, S.; Grätzel, M.; Brunetti, F.; Freitag, M. Unlocking High-Performance Photocapacitors for Edge Computing in Low-Light Environments. *Energy Environ. Sci.* **2025**, *18*, 4704-4716.
2. Hoyer, R. L. Z.; Koutsourakis, G.; Freitag, M.; Jehl Li-Kao, Z.; Österberg, T.; Aliwell, S.; Bellanger, M.; Brown, T. M.; Brunetti, F.; Carnie, M. J.; et al. Reaching a Consensus on Indoor Photovoltaics Testing. *Joule* **2025**, *9*, 102127.
3. Shi, N.; Tian, H.; Yang, G.; Wang, P.; Lei, Y.; Li, W.; Jiang, J.; Jiang, X.; Shen, Z.; Freitag, M.; Guo, X.; Pang, S. Multifunctional Fluorinated Self-Polymerizable Additive Improves the Performance of Perovskite Photovoltaics. *ACS Energy Lett.* **2026**, DOI: 10.1021/acsenerylett.5c03901.
4. Bradford, D.; Benesperi, I.; Jinno, H.; Bhati, N.; Avilés-Betanzos, R.; Maréchal, F.; Oskam, G.; Shih, C.-J.; Grätzel, M.; Sánchez, S.; Sivula, K.; Freitag, M. Flash-Infrared-Annealing-Enabled High-Temperature Sintering of Photoanodes on Flexible Polymer Foils for Ultralight Photovoltaics. *ACS Energy Lett.* **2026**, *11*, 699-706.
5. You, S.; Zhu, H.; Shen, Z.; Wang, X.; Shao, B.; Wang, Q.; Lu, J.; Yuan, Y.; Dou, B. D.; Sanehira, E. M.; Russell, T.; Lorenz, A.; Dong, Y.; Chen, L.; Casareto, M.; Rolston, N.; Beard, M. C.; Berry, J. J.; Freitag, M.; Yan, Y.; Bakr, O. M.; Zhu, K. C₆₀-Based Ionic Salt Electron Shuttle for High-Performance Inverted Perovskite Solar Modules. *Science* **2025**, *388*, 964-968.
6. Michaels, H.; Golomb, M. J.; Kim, B. J.; Edvinsson, T.; Cucinotta, F.; Waddell, P. G.; Probert, M. R.; Konezny, S. J.; Boschloo, G.; Walsh, A.; Freitag, M. Copper Coordination Polymers with Selective Hole Conductivity. *J. Mater. Chem. A* **2022**, *10*, 9582-9591.
7. Flores-Diaz, N.; De Rossi, F.; Das, A.; Deepa, M.; Brunetti, F.; Freitag, M. Progress of Photocapacitors. *Chem. Rev.* **2023**, *123*, 9327-9355.
8. Zhu, H.; Shao, B.; Shen, Z.; You, S.; Yin, J.; Wehbe, N.; Wang, L.; Song, X.; Abulikemu, M.; Basaheeh, A.; Jamal, A.; Gereige, I.; Freitag, M.; Mohammed, O. F.; Zhu, K.; Bakr, O. M. In Situ Energetics Modulation Enables High-Efficiency and Stable Inverted Perovskite Solar Cells. *Nat. Photonics* **2025**, *19*, 28-35.