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PHOTOINDUCED LIVING POLYMERIZATION FOR ADVANCED POLYMER SYNTHESIS

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The ability of plants to convert solar energy into chemical energy via photoredox processes (natural photosynthesis) has inspired generations of chemists to try to reproduce such systems. Lately, the use of visible light photoredox catalysis in organic chemistry has enabled the synthesis of known chemical compounds through novel synthetic routes, eliminating side reactions and complex purification procedures. Recently, we have successfully implemented photoredox catalysis in polymer chemistry leading to the development of novel light-induced polymerizations, namely PET-RAFT technology, which involves the reversible deactivation of thiocarbonylthio compounds (RAFT agents) by photoredox catalysts via a photoinduced electron or energy transfer (PET) process. In this technology, ppm amount of photoredox catalyst is employed to catalyze RAFT agent and generate radicals for subsequent polymerization, instead of external radical initiator in the traditional RAFT formulation. The RAFT agent plays the role of initiator, chain transfer agent and termination agent. Although this technology was developed from traditional RAFT polymerization with minor changes in formulation, it presented different reaction mechanisms and offered a number of significantly "green" attributes to living radical polymerizations, including: (1) low energy consumption and mild reaction conditions, (2) spatial and temporal control on radical polymerization, (3) high oxygen tolerance, (4) versatile photocatalysts and (5) selective polymerization activation. In this talk, these benefits from PET-RAFT technology will be summarized and demonstrated by our recent results. This technology is not only useful in contributing to the development of green chemistry and sustainable polymer manufacturing, but also in providing opportunities for the innovation of new methods of organic and polymer synthesis.

Recent Publications

1. J. M. R. Narayanam and C. R. J. Stephenson (2011), Visible light photoredox catalysis: applications in organic synthesis, *Chem. Soc. Rev.*, 40 (1), 102–113; (b) C. K. Prier, D. A. Rankic and D. W. C. MacMillan (2013), Visible Light Photoredox Catalysis with Transition Metal Complexes: Applications in Organic Synthesis, Chem. Rev., 113 (7), 5322–5363. (c) N. Corrigan, S. Shanmugam, J. Xu, C. Boyer (2016), Photocatalysis in Organic and Polymer Synthesis, *Chem. Soc. Rev.* 45(22), 6165-6212.
2. J. Xu, K. Jung, A. Atme, S. Shanmugam and C. Boyer (2014), A robust and versatile photoinduced living polymerization of conjugated and unconjugated monomers and its oxygen tolerance, *J. Am. Chem. Soc.*, 136 (14), 5508–5519; (b) J. Xu, K. Jung and C. Boyer (2014), Oxygen Tolerance Study of Photoinduced Electron Transfer–Reversible Addition–Fragmentation Chain Transfer (PET-RAFT) Polymerization Mediated by Ru $(bpy)_3$, *Macromolecules,* 47 (13), 4217–4229; (c) J. Xu, K. Jung, N. A. Corrigan and C. Boyer (2014), Aqueous photoinduced living/controlled polymerization: tailoring for bioconjugation, Chem. Sci., 5 (9), 3568–3575; (d) J. Xu, S. Shanmugam, H. T. Duong and C. Boyer (2015), Organo-photocatalysts for photoinduced electron transfer-reversible addition–fragmentation chain transfer (PET-RAFT) polymerization, *Polym. Chem.,* 6 (31), 5615-5624. (e) S. Shanmugam, J. Xu and C. Boyer (2015), Exploiting Metalloporphyrins for Selective Living Radical Polymerization Tunable over Visible Wavelengths, J. Am. Chem. Soc. 137 (28), 9174-9185. (f) J. Xu, S. Shanmugam, C. Fu, K. Aguey-Zinsou and C. Boyer (2016), Selective Photoactivation: From a Single Unit Monomer Insertion Reaction to Controlled Polymer Architectures, J. Am. Chem. Soc. 138 (9), 3094-3106. (g) J. Xu, C. Fu, S. Shanmugam, C. J. Hawker, G. Moad and C. Boyer (2017), Synthesis of Discrete Oligomers by Sequential PET-RAFT Single-Unit Monomer Insertion, *Angew. Chem. Int. Ed.,* 56 (29), 8376-8383.

Biography

Jiangtao Jason Xu is an Australian Research Council (ARC) Future Fellow and Lecturer at School of Chemical Engineering, UNSW Sydney. He received his BS (2001) and PhD (2007) Degrees in Polymer Chemistry from Fudan University (China), where he focused on RAFT polymerization and advanced polymer synthesis. Following Post-doctoral research experience in UNSW and University of Melbourne, he joined UNSW again to develop visible light-induced living polymerization and precision polymer synthesis. He was awarded the prestigious ARC Future Fellowship in 2016. He has more than 70 peer-reviewed publications in high-impact journals, attracting more than 2700 citations and an H-index of 30. His areas of research interests are green chemistry and sustainable polymer synthesis, precision polymer synthesis mimicking natural perfection, photoredox catalysis for living polymerization, advanced polymer materials for nanomedicine and bioengineering applications.

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