ABSTRACT

Radical polymers hold great potential as solid-state conducting materials due to their distinctive charge transport mechanism and intriguing optical properties resulting from their singly occupied molecular orbital energy levels. Furthermore, the paramagnetic nature of their open-shell structures broadens their applicability, allowing them to be magnetic field-active while also offering promising spin transport properties. These molecular design features position radical polymers as interesting materials for next-generation quantum information systems as well.

The thesis contains an overview of recent advances of conductive polymers in solid state devices, especially in optoelectronics and spintronic applications. In turn, by synthesizing and understanding the underlying charge transport mechanisms of 2,2,6,6-tetramethylpiperidin-1-oxyl (TEMPO) derivatives (dendrimers, liquid crystals, etc.), the discussion then shifts to the progress in remarkable electromagnetic responses in solid-state devices. Then, the discussion moves on to enabling the synthesis of a series of stereoregular polymers for advanced applications such as giant magnetoresistance (GMR) and inverse spin Hall effect (ISHE) in spintronic materials. We leveraged stereoselective cationic polymerization to design a polymer with a stable persistent radical in each repeat unit that enables the long-range order necessary for spin transport. This approach overcomes conventional requirements for doping in organic spin-pumping devices while showcasing high conductivity, long spin-diffusion lengths, and processability.

In conclusion, the needs to extend research of open-shell macromolecules are urgent, with the aspiration that this effort offers essential contexts and references to stimulate advancements in this field. This approach seeks to unleash the full potential of radical polymers (and organic radicals in a wider scope), in terms of pioneering scientific contributions and societal influence.