Bulk heterojunction (BHJ) organic solar cells (OSCs) have attracted intensive research attention over the past decades owing to their unique advantages including mechanical flexibility, lightweight, and low-cost fabrications. To date, OSC devices have achieved power conversion efficiencies (PCEs) exceeding 12%. Much of the progress was enabled by the development of high-performance donor polymers with favourable morphological, electronic, and optical properties. A key problem in morphology control of OSCs is the trade-off between achieving small domain size and high polymer crystallinity. For example, the thickness of OSC blends containing state-of-the-art PTB7 family donor polymers are restricted to ~100 nm due to their tendency to form disordered and impure domains with low charge mobility. To further improve the device performance and promote commercialization of OSCs, there is a strong demand for the design of new donor polymers that can achieve an optimal blend morphology containing highly crystalline yet reasonably small domains.

In this work, we report recent progress on a new family of conjugated polymers with strong temperature-dependent aggregation (TDA) properties. These polymers are mostly disaggregated and easily dissolve in solution at high temperatures, yet they can strongly aggregate when the solution is cooled to room temperature. This unique TDA property enables us to control the disorder-order transition during solution processing. By preheating the solution to high temperature, the disaggregated polymer chains have sufficient time to aggregate during spin coating and form crystalline domains that are not excessively large, and the overall blend morphology can be optimized by various processing conditions (e.g., temperature, spin rates, concentration, etc.). This well-controlled and near-optimal BHJ morphology produced over a dozen cases of efficient OSCs with an active layer nearly 300 nm thick that can still achieve high FFs (70-77%) and efficiencies (10-11.7%). By studying the structure-property relationship of the donor polymers, we show that the second position branched alkyl chains and the fluorination on the polymer backbone are two key structural features that enable the TDA property. Moreover, our comparative studies show that the TDA polymer family can be used to match with non-fullerene acceptors yielding OSCs with low voltage losses. The key difference between the empirical matching rules for fullerene and non-fullerene OSCs is that TDA polymers with slightly reduced crystallinity appear to match better with small molecular acceptors and yield higher OSC performances.

Figure 1: The schematic illustration of the spin-coating and morphology forming process. Polymers change from “spaghetti” to “flat noodles” during this cooling process.