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Making Glasses Conduct: Electrochemical Doping of Redox-Active Polymer Thin Films

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Optoelectronically-active macromolecules have been established as promising materials in myriad organic electronic applications (e.g., organic field-effect transistors (OFETs) and organic photovoltaic (OPV) devices). To date, however, the majority of the work surrounding these materials has focused on materials with a great deal of conjugation along their macromolecular backbones and with varying degrees of crystalline structure. Here, we describe an emerging class of macromolecular charge conductors, radical polymers, that: (1) do not contain conjugation and (2) are completely amorphous glasses. Radical polymers contain non-conjugated macromolecular backbones and stable radical sites along the side chains of the electronically-active materials. In contrast to conjugated polymer systems, these materials conduct charge in the solid state through oxidation-reduction (redox) reactions along these pendant groups. Specifically, we demonstrate that controlling the chemical functionality of the pendant groups and the molecular mobility of the macromolecular backbones significantly impacts the charge transport ability of the pristine (i.e., not doped) radical polymers species. Through proper control of these crucial parameters, we show that radical polymers can have electrical conductivity and charge mobility values on par with commonly-used conjugated polymers. Importantly, we also highlight the ability to dope radical polymers with redox-active small molecule species. This doping, in turn, increases the electrical conductivity of the glassy radical polymer thin films in a manner akin to what is observed in traditional conjugated polymer systems. In this way, we establish a means by which to fabricate optically-transparent and colorless thin film glasses capable of conducting charge in a rather rapid manner. We anticipate that these fundamental insights will prove crucial in developing new transparent conducting layers for future electronic applications.