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Improvement of the thermoelectric properties of PEDOT:PSS films via DMSO addition and DMSO/salt post-treatment resolved from a fundamental view

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Organic semiconductors have attracted intense attention due to their potential use in mechanically flexible, lightweight, and inexpensive electronic devices. Especially, poly(3,4-ethylene dioxythiophene):poly(styrenesulfonate)(PEDOT:PSS) is the most studied conducting polymer system due to its intrinsically high electrical conductivity, low thermal conductivity, and high mechanical flexibility. The combination of DMSO-solvent doping and physical-chemical DMSO/salt de-doping in a sequence has been used to improve the thermoelectric PEDOT:PSS films. A high power factor of ca.105.2 μW m-1 K-2 has been achieved for the PEDOT:PSS film after post-treatment with 10 % Na2SO3 in the DMSO/salt mixture (v/v), outperforming NaHCO3. The initial DMSO-doping treatment induces a distinct phase separation by facilitating the aggregation of the PEDOT molecules. At the same time, the subsequent DMSO/salt de-doping post-treatment strengthens the selective removal of the surplus non-conductive PSS chains. Substantial alterations in the oxidation level, chain conformations, PEDOT crystallites and their preferential orientation are observed upon treatment on the molecular level. At the mesoscale level, the purification and densification of PEDOT-rich domains enable the realization of inter-grain coupling by the formation of the electronically well-percolated network. Thereby, both electrical conductivity and Seebeck coefficient are optimized.

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