Improved alignment of PEDOT:PSS induced by *in-situ* crystallization of "green" dimethylsulfone molecules to enhance the polymer thermoelectric performance

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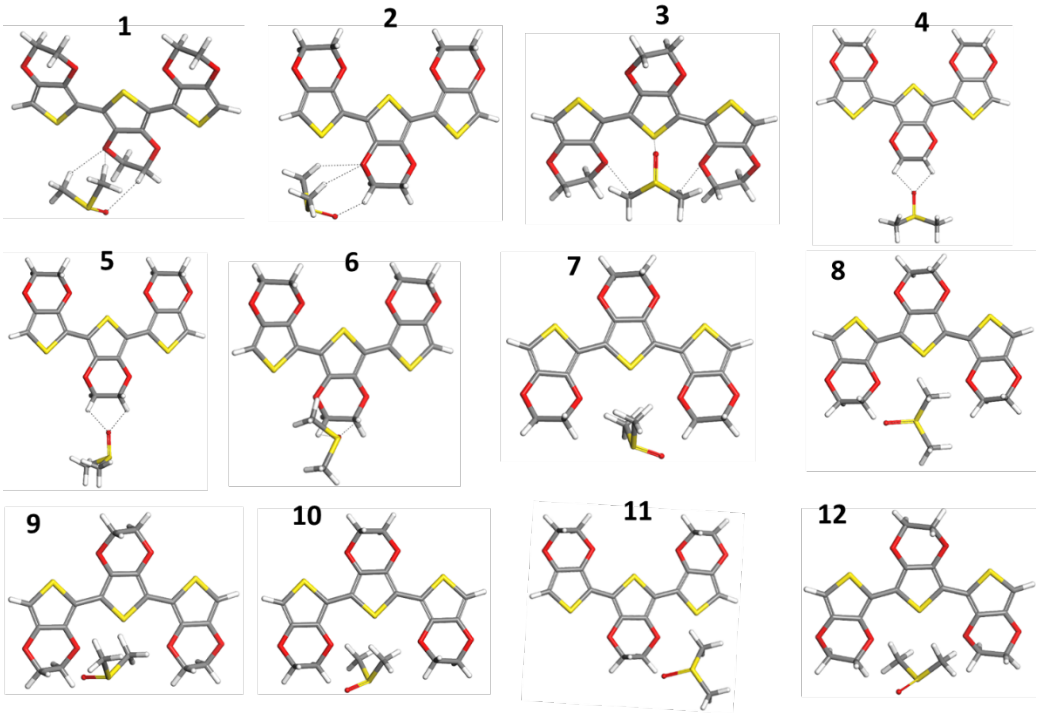

Fig. S1 PEDOT-DMSO initial structures for DFT calculations.

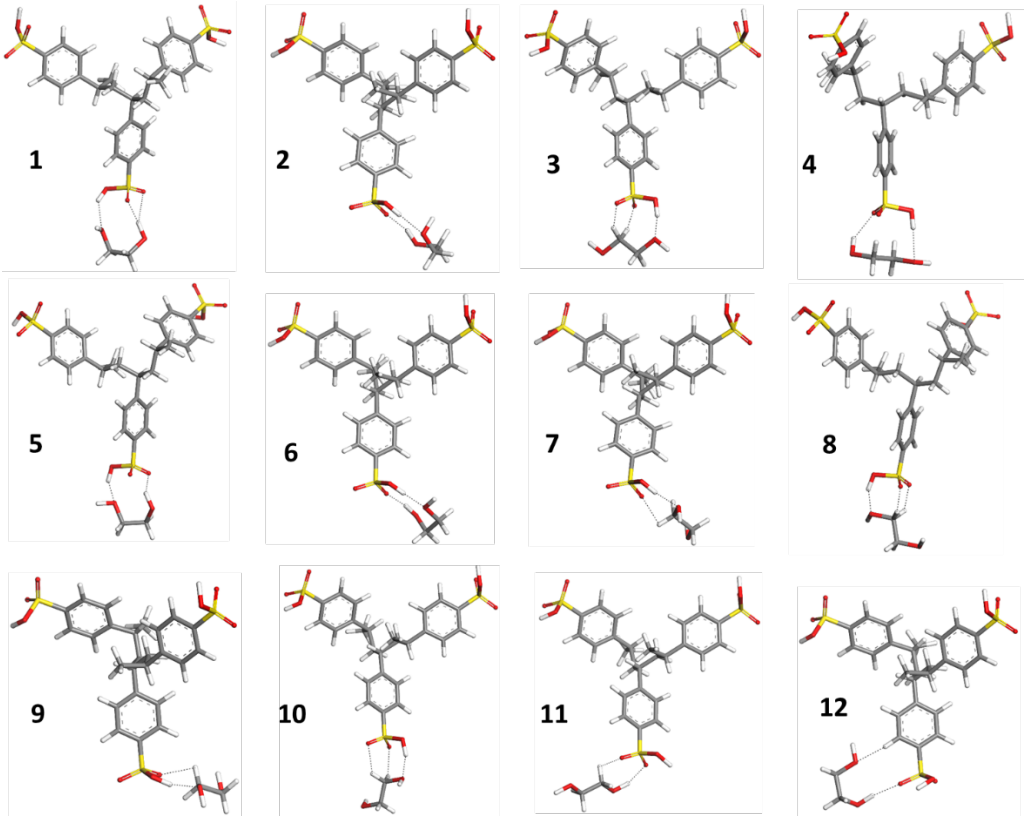

Fig. S2 PSSH-EG initial structures for DFT calculations.

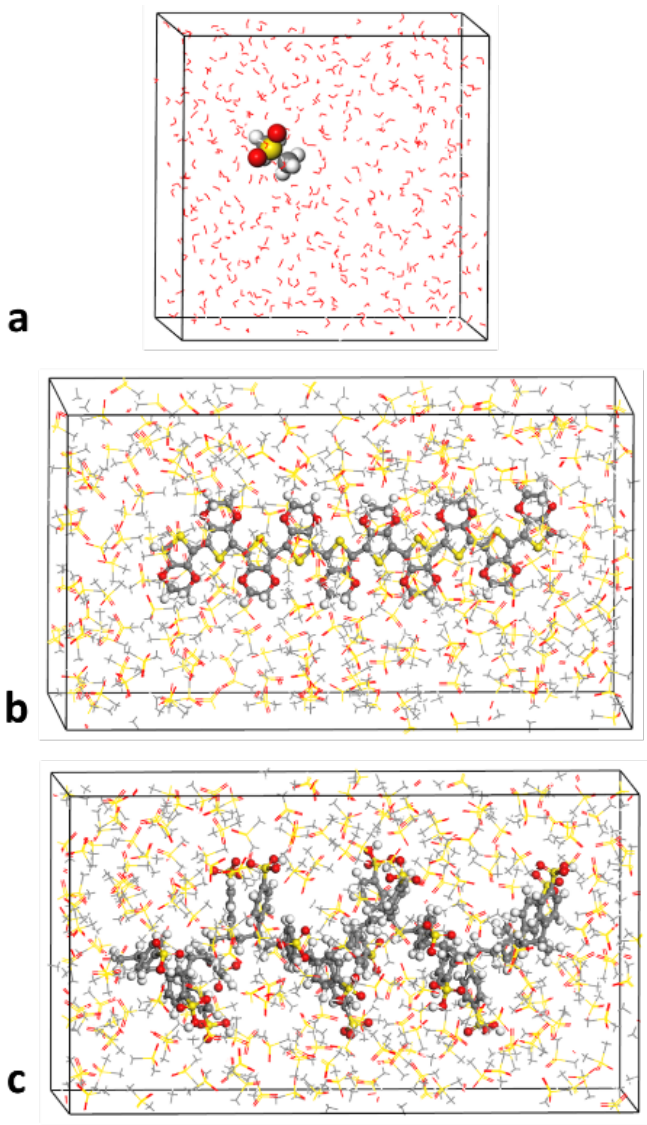

Fig. S3 Three periodic cells constructed to calculate free energy of solvation. A) $DMSO_2$ in water, b) $EDOT_9$ in $DMSO_2$ and c) SS_{18} in $DMSO_2$.

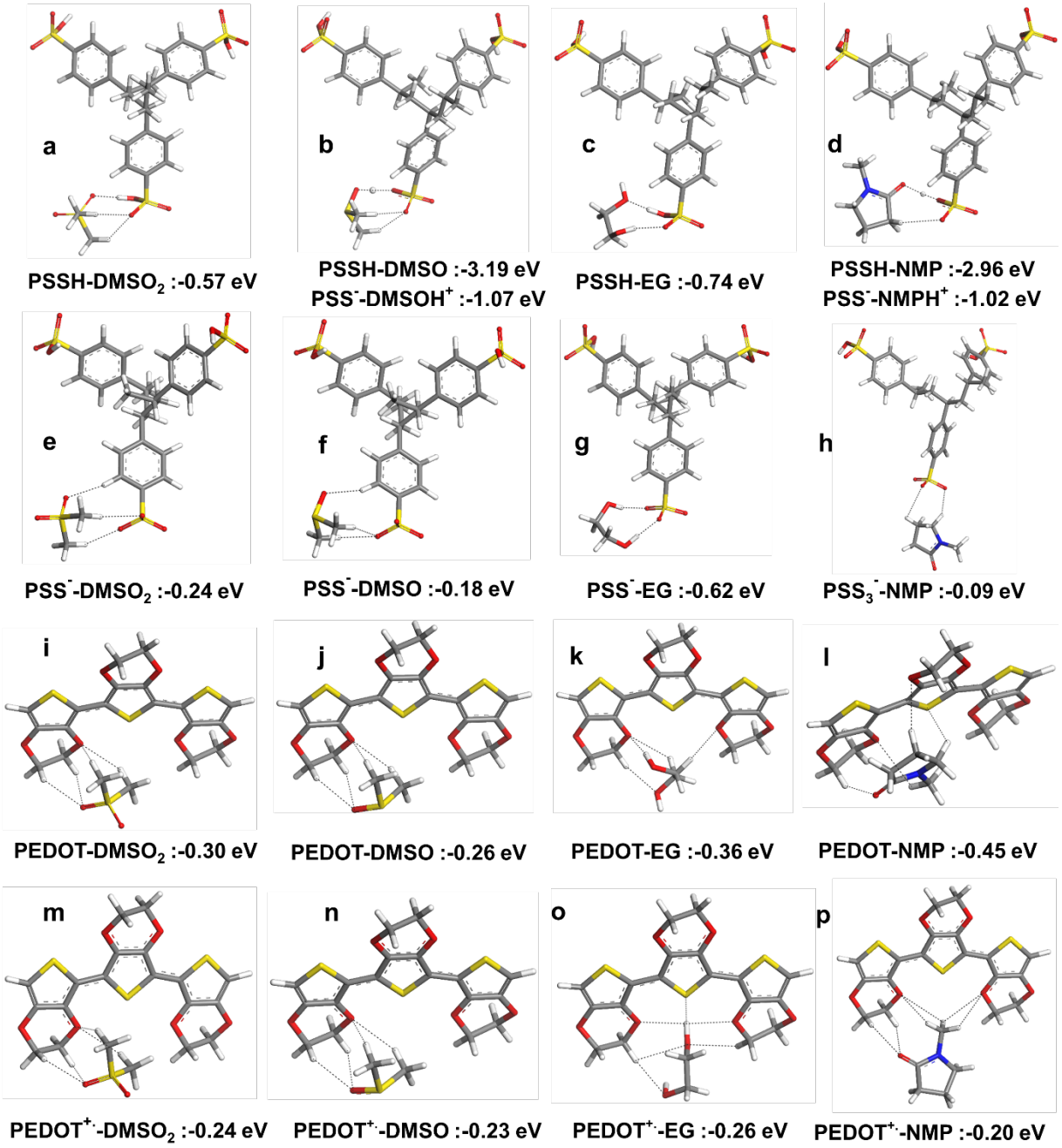

Fig. S4 Optimized lowest energy structures and interaction energies *in water solvation* for a-d) PSSH trimer with $DMSO_2$, DMSO, EG and NMP e-h) PSS^- trimer with $DMSO_2$, DMSO, EG and NMP, i-l) PEDOT trimer with $DMSO_2$, DMSO, EG and NMP, m-p) $PEDOT^{+\cdot}$ trimer with DMSO, $DMSO_2$, EG and NMP.

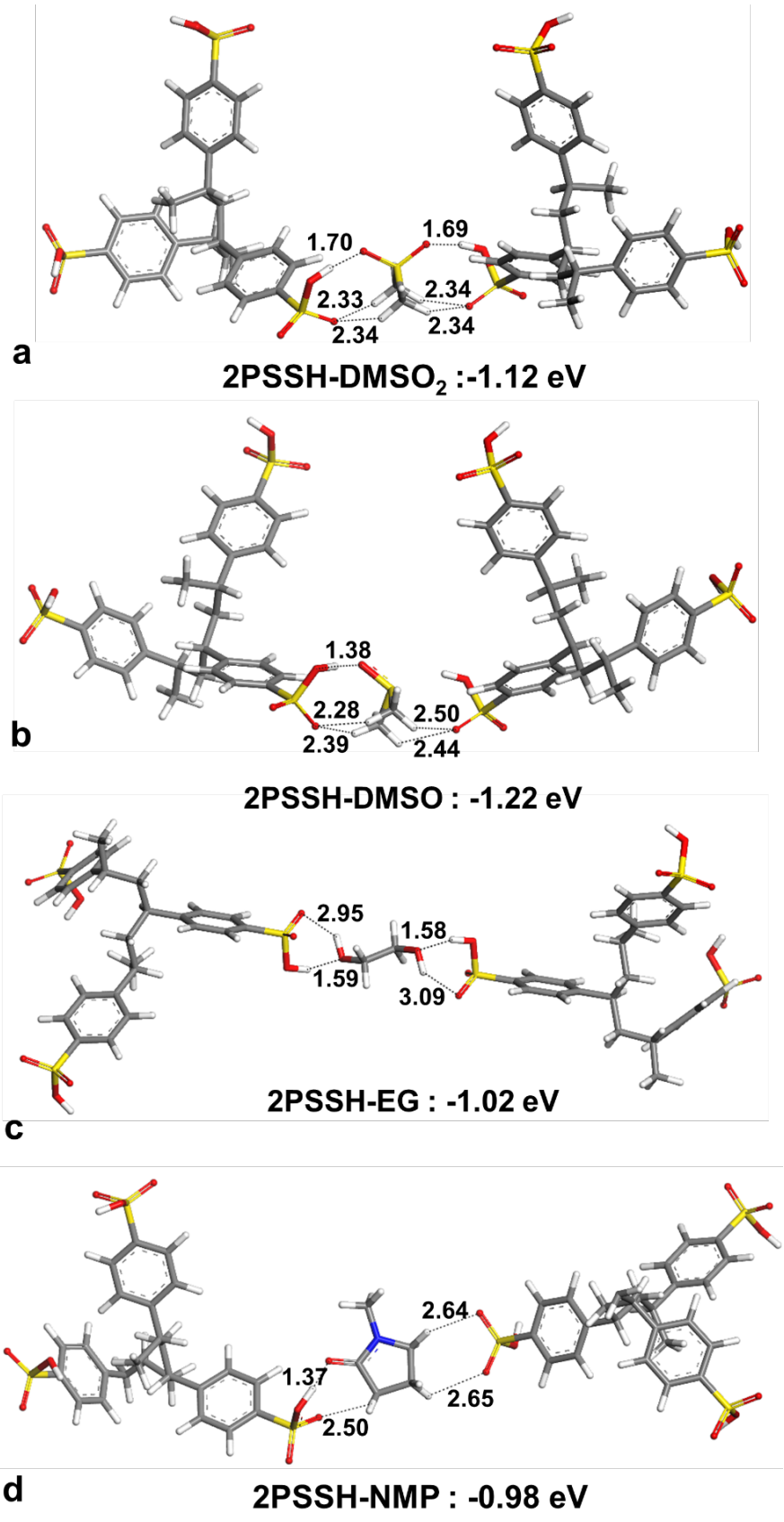

Fig. S5 Optimized lowest energy structures and interaction energies *in water solvation effect* for a) two PSSH trimers with DMSO2, b) two PSSH trimers with DMSO, b) two PSSH trimers with EG, b) two PSSH trimers with NMP. Interaction energies calculated with water solvation effect are given in parenthesis.

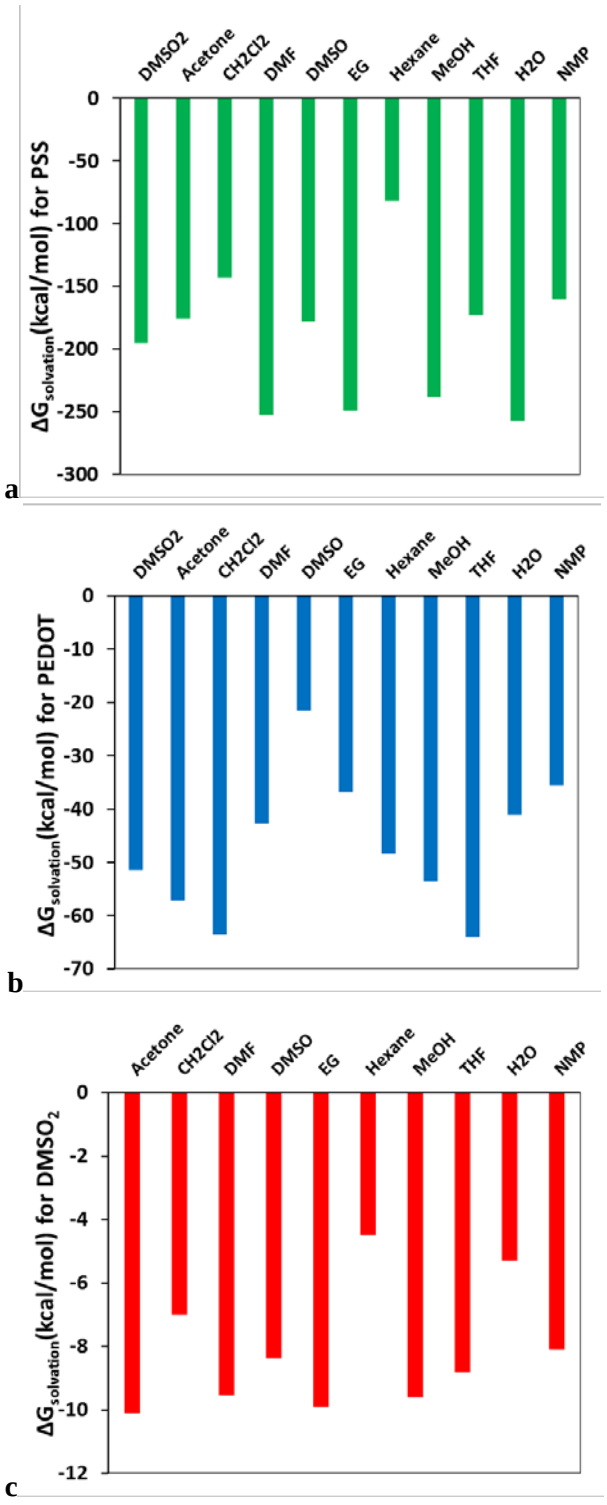

Fig. S6 Free energy of solvation (ΔG_{sol}) for a) SS_{18}^{-3} b) $EDOT_9^{+3}$ c) $DMSO_2$ in different solvents.

Table S1. Comparison of this work with some reported thermoelectric properties of PEDOT films treated with different chemicals.

Reference	Process	σ (S/cm)	S (μV/ K)	PF (μW/mK^2)	κ (W/mK)	ZT
This work	PEDOT:PSS treated with $DMSO_2$	1080	17.1	32	0.54	0. 02
1	PEDOT:PSS treated with DMSO, then DMSO and hydrazine	677	41	115	0.17	0.2
2	PEDOT:PSS treated with a mixture of DMSO and hydrazine	578	67	112	-	
3	PEDOT:PSS treated with p-toluenesulfonic acid monohydrate, then hydrazine/DMSO solution	~1300	~50	318	0.30	0.31
4	Addition of DMSO in to PEDOT:PSS and then treated with poly(ethylene oxide)	1061	38.4	157	-	-
5	PEDOT:PSS treated with DMSO/$NaBH_4$	~580	~40	98	0.45	0.06

References

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