

Supplemental Information for

Unlocking Enhanced Thermal Conductivity in Polymer Blends through

Active Learning

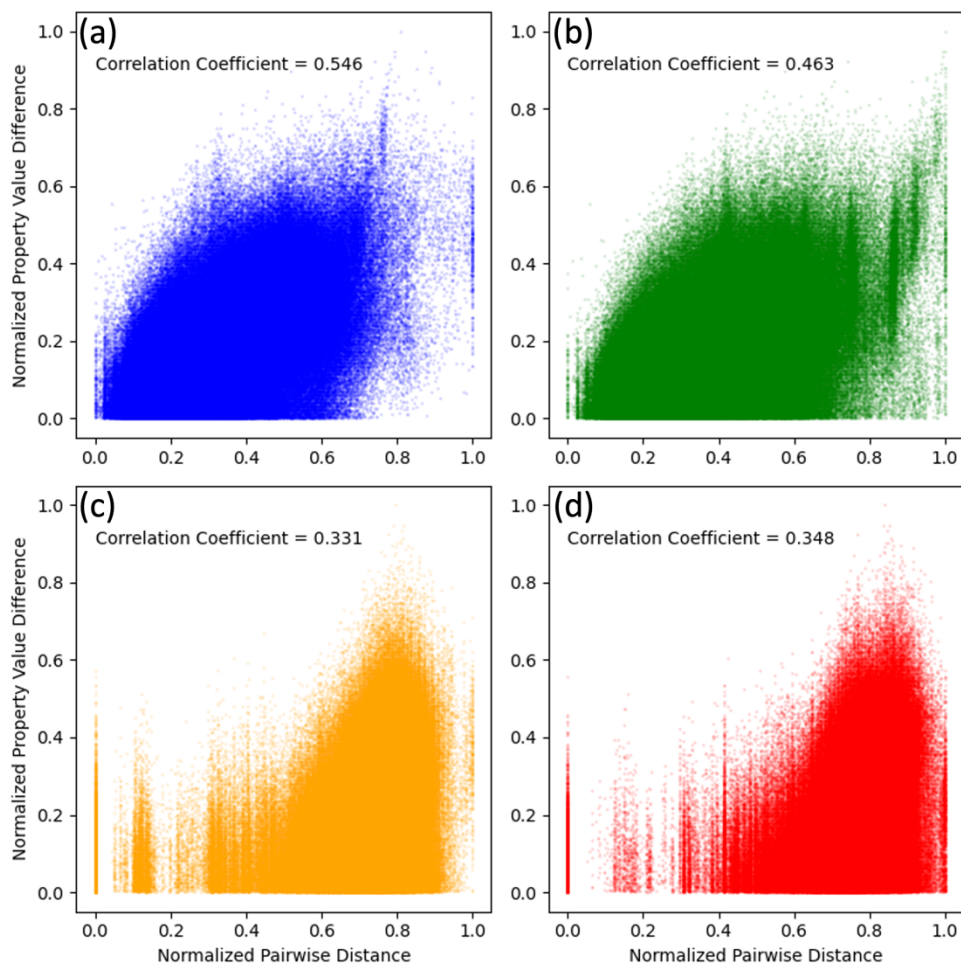
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Affiliations:

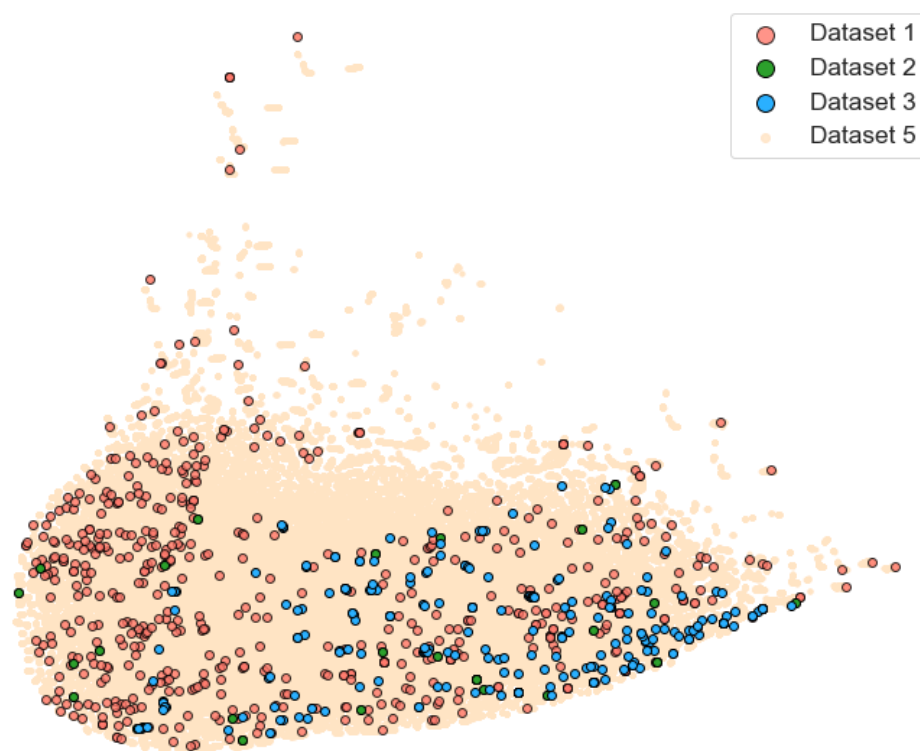
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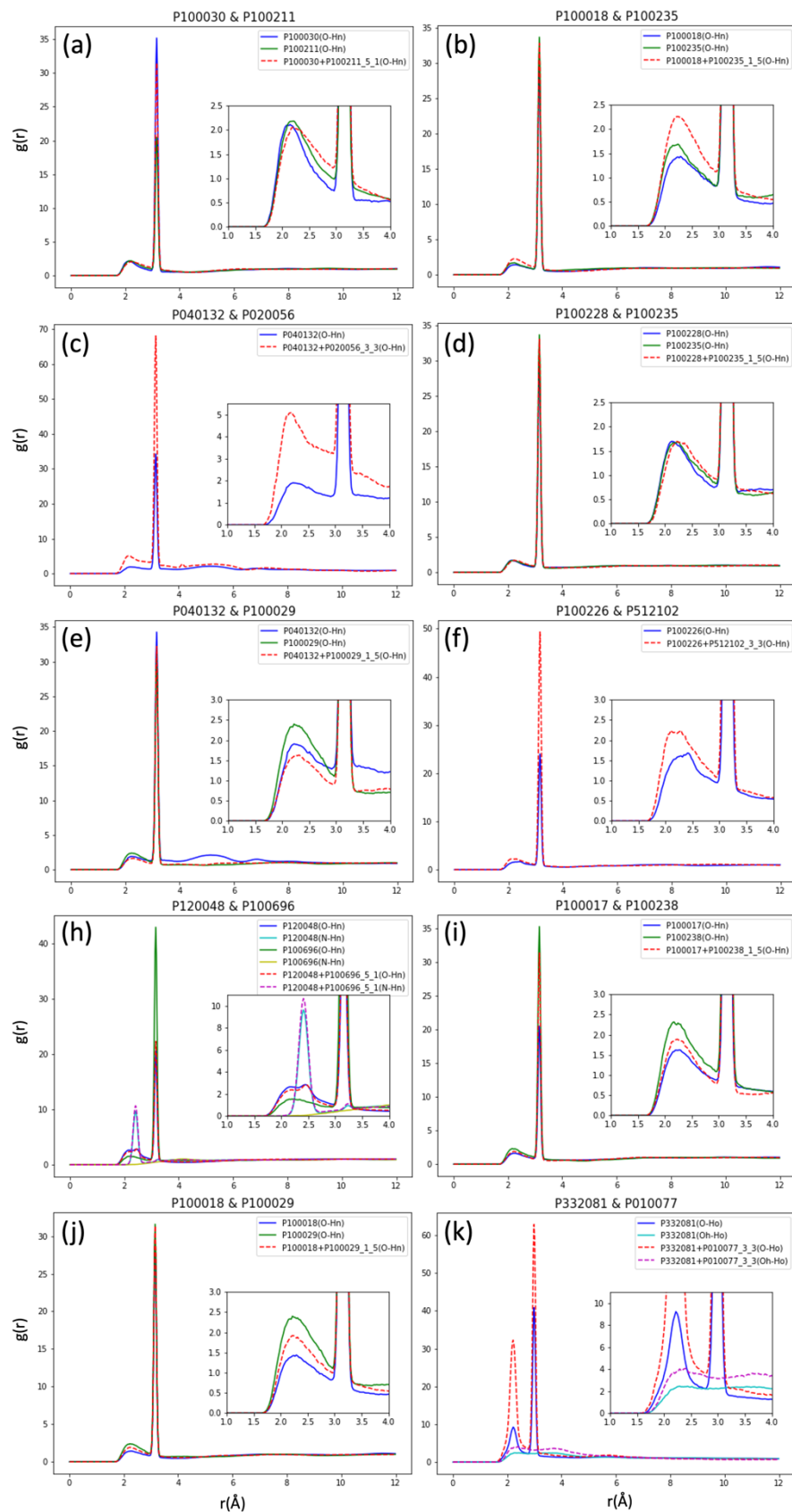
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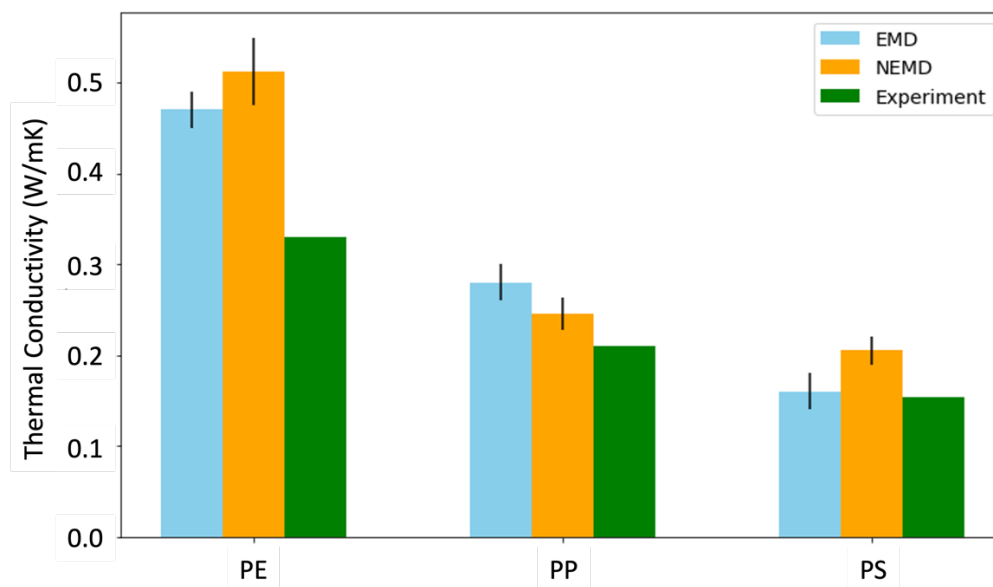
Supplementary Figure 1. Relationship between the pairwise absolute differences in the TC property values of polymers and the corresponding pairwise Euclidean distances as represented by each representation method: (a) PE-WS, (b) PE-WC, (c) MF-WS, and (d) MF-WC. Both the pairwise distance and property value differences are normalized using min-max normalization. Spearman's rank correlation coefficient is calculated and shown in each subplot.



Supplementary Figure 2. 2D t-SNE visualization of chemical space of Dataset 1, 2, 3, and 5 using PE-WS representation.

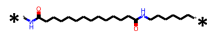
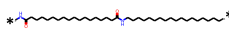
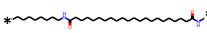
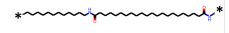
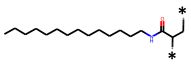
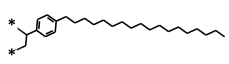
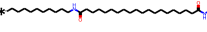
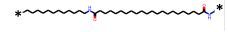
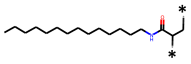
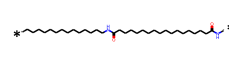
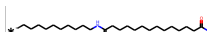
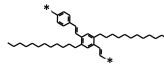
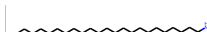
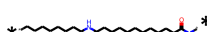
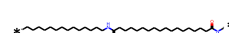
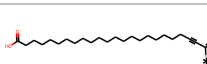
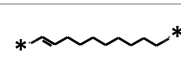


Supplementary Figure 3. Radial pair distribution function (RDF) of atoms involved in hydrogen bonds. Panels (a) through (k) represent the top-10 predicted polymer blend candidates (dashed lines) along with their constituent single-component polymers (solid lines), indexed from #1 to #10. The atoms contributing to the RDF calculation are indicated in the respective legends as “X-Y,” where “X” represents the reference atom (proton acceptor), and “Y” represents the hydrogen atom (proton donor). In the legends, “O” indicates a double-bond oxygen atom, “Oh” indicates an oxygen atom within the hydroxyl group, “N” indicates a nitrogen atom, “Hn” indicates a hydrogen atom bonded to a nitrogen atom, and “Ho” indicates a hydrogen atom within the hydroxy group. See Table S3 for details of the atom symbols.



Supplementary Figure 4. Comparative TC of polyethylene (PE), polypropylene (PP), and polystyrene (PS) from EMD, NEMD, and experiments. The NEMD results are averaged over four independent runs with the standard deviation as the error bar, which is the mean $\pm$ standard deviation. The EMD results are from Ref. ¹. The experimental values are the median of experimental TCs of the three polymers sourced from the PoLyInfo database ², accessed on February 20, 2024.

Supplementary Table 1. Blending information of the top-10 predicted polymer blend candidates and their constituent single-component polymers. The two constituent single-component polymers are denoted by subscripts “1” and “2”, respectively. PID is from PoLyInfo ² and polymerization points of the monomer are denoted by “*”.

Index	PID ₁	PID ₂	Blending Ratio	Structure ₁	Structure ₂
#1	P100211	P100030	1:5		
#2	P100018	P100235	1:5		
#3	P040132	P020056	3:3		
#4	P100228	P100235	1:5		
#5	P040132	P100029	1:5		
#6	P100226	P512102	3:3		
#7	P100696	P120048	1:5		
#8	P100017	P100238	1:5		
#9	P100018	P100029	1:5		
#10	P332081	P010077	3:3		

Supplementary Table 2. TC and R_g validation results for top-10 predicted polymer blend candidates and their constituent single-component polymers. The two constituent single-component polymers are denoted by subscripts “1” and “2”, respectively, and polymer blends are denoted by subscript “blend”. All TC and R_g values are in units of W/mK and Å, respectively. TC of polymer blends higher than that of both constituent single-component polymers are in bolded.

Index	TC ₁	TC ₂	TC _{blend}	$R_{g\ 1}$	$R_{g\ 2}$	$R_{g\ blend}$
#1	0.355	0.453	0.396	22.410	22.372	20.256
#2	0.411	0.373	0.464	23.852	21.919	21.075
#3	0.375	0.368	0.378	11.883	13.523	13.198
#4	0.387	0.373	0.467	24.796	21.919	23.537
#5	0.375	0.461	0.490	11.883	21.332	19.459
#6	0.342	0.315	0.355	19.014	16.220	17.649
#7	0.412	0.373	0.437	21.925	20.665	20.925
#8	0.346	0.345	0.416	22.084	21.847	18.625
#9	0.411	0.461	0.401	23.852	21.332	20.575
#10	0.420	0.338	0.442	15.048	17.818	16.651

Supplementary Table 3. Proton donors and acceptors in the hydrogen bond considered in this work. The proton acceptor and donor atoms are highlighted in red. “R” represents a non-hydrogen atom.

Proton acceptor	Structure	Proton donor	Structure
N	$\begin{array}{c} \text{H} - \text{N} - \text{R} \\ \\ \text{R} \end{array}$	Ho	$\text{H} - \text{O} - \text{R}$
N3	$\begin{array}{c} \text{R} - \text{N} - \text{R} \\ \\ \text{R} \end{array}$	Hn	$\begin{array}{c} \text{H} - \text{N} - \text{R} \\ \\ \text{R} \end{array}$
O	$\text{O} = \text{R}$		
Os	$\text{R} - \text{O} - \text{R}$		
Oh	$\text{H} - \text{O} - \text{R}$		
Ss	$\text{R} - \text{S} - \text{R}$		
Cl	$\text{Cl} - \text{R}$		
F	$\text{F} - \text{R}$		

Supplementary Table 4. A summary of all possible models for the TC (A), R_g (B), and H-bond (C) data, with complete independence (most restrictive) at the top the saturated model at the bottom.

model	df (residual)	deviance	log likelihood	p-value
(A, B, C)	4	6.940	-23.672	0.139
(A, BC)	3	6.676	-23.540	0.083
(B, AC)	3	5.687	-23.045	0.128
(C, AB)	3	2.655	-21.529	0.448
(AC, BC)	2	5.423	-22.914	0.066
(AB, BC)	2	2.391	-21.398	0.302
(AB, AC)	2	1.402	-20.903	0.496
(AB, AC, BC)	1	1.009	-20.707	0.315
(ABC)	0	0	\	\

Supplementary Table 5. Hierarchical model selection for all the reduced models, with conditional independence at the top and complete independence (most restrictive) at the bottom.

Independence	H₀	H_a	p-value
Conditional	(AB, AC)	(AB, AC, BC)	0.036
	(AB, BC)	(AB, AC, BC)	0.240
	(AB, AC)	(AB, AC, BC)	0.531
Joint	(C, AB)	(AB, BC)	0.608
	(A, BC)	(AB, BC)	0.038
	(B, AC)	(AB, AC)	0.038
	(C, AB)	(AB, AC)	0.263
Mutual	(A, B, C)	(C, AB)	0.038

Calculation of marginal and conditional odds ratio

Each odds ratio is also calculated with a 95% confidence interval (CI). If it does not include 1, it indicates that the odds are significant. From the results below, Supplementary Equations (1) to (6), only the conditional odds ratio between A and B, for the fixed level C=1, is significant, as shown in Supplementary Equation (4).

Marginal odds ratio:

$$\hat{\theta}_{AB} = \frac{\mu_{00+}\mu_{11+}}{\mu_{01+}\mu_{10+}} = \frac{(30 + 69) * (10 + 32)}{(84 + 154) * (3 + 5)} = 2.18,$$
$$95\% \text{ CI} = [0.9895, 4.8195]$$
(1)

$$\hat{\theta}_{AC} = \frac{\mu_{0+0}\mu_{1+1}}{\mu_{0+1}\mu_{1+0}} = \frac{(30 + 84) * (5 + 32)}{(69 + 154) * (3 + 10)} = 1.45,$$
$$95\% \text{ CI} = [0.7438, 2.8463]$$
(2)

Conditional odds ratio:

$$\hat{\theta}_{AB(C=0)} = \frac{\mu_{000}\mu_{110}}{\mu_{010}\mu_{100}} = \frac{30 * 10}{84 * 3} = 1.19$$
$$95\% \text{ CI} = [0.3068, 4.6194]$$
(3)

$$\hat{\theta}_{AB(C=1)} = \frac{\mu_{001}\mu_{111}}{\mu_{011}\mu_{101}} = \frac{69 * 32}{154 * 5} = 2.87$$
$$95\% \text{ CI} = [1.0715, 7.6739]$$
(4)

$$\hat{\theta}_{AC(B=0)} = \frac{\mu_{000}\mu_{101}}{\mu_{001}\mu_{100}} = \frac{30 * 5}{69 * 3} = 0.72$$

$$95\% CI = [0.1626, 3.2288]$$

(5)

$$\hat{\theta}_{AC(B=1)} = \frac{\mu_{010}\mu_{111}}{\mu_{011}\mu_{110}} = \frac{84 * 32}{154 * 10} = 1.75$$

$$95\% CI = [0.8177, 3.7256]$$

(6)

SI REFERENCES

1. Feng, T. *et al.* Size Effects in the Thermal Conductivity of Amorphous Polymers. *Phys. Rev. Appl.* **14**, 044023 (2020).
2. Polymer Database(PoLyInfo) - DICE :: National Institute for Materials Science.
<https://polymer.nims.go.jp/en/>.