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**Table S1.** Designation and formulation of the CO-BPU samples.

Samples	CO(g)	PCL(g)	ISO(g)	IPDI(g)	HT(g)	APTES(g)
CO-BPU	3	3	0	2.6	0	0

**Table S2.** Designation and formulation of the ISO-BPU samples.

Samples	CO(g)	PCL(g)	ISO(g)	IPDI(g)	HT(g)	APTES(g)
ISO-BPU-1	3	3	0.2	3	0.6	0
ISO-BPU-2	3	3	0.45	3.46	0.6	0
ISO-BPU-3	3	3	0.9	4.34	0.6	0
ISO-BPU-4	3	3	1.35	5.21	0.6	0

**Table S3.** Designation and formulation of the APTES-BPU samples.

Samples	CO(g)	PCL(g)	ISO(g)	IPDI(g)	HT(g)	APTES(g)
APTES-BPU-1	3	3	0.45	3.54	0.6	0.11

<b>APTES-BPU-2</b>	3	3	0.45	3.76	0.6	0.44
<b>APTES-BPU-3</b>	3	3	0.45	4.05	0.6	0.89
<b>APTES-BPU-4</b>	3	3	0.45	4.34	0.6	1.34

**Table S4.** Fitting quality parameters ( $R^2$ ) for the deconvolution of C=O stretching vibration region in FTIR spectra of all samples.

Quality Parameters	CO-BPU	ISO-BPU				APTES-BPU			
		1	2	3	4	1	2	3	4
$R^2$	0.979	0.997	0.989	0.990	0.985	0.999	0.987	0.984	0.999

**Table S5.** Summary of the Assignments of Deconvoluted Subpeaks for the C=O Stretching Vibration Absorption Band in the FTIR Spectrum of CO-BPU.

Assignment	Subpeak	Wavenumber( $\text{cm}^{-1}$ )	Area(%)	
$\nu(\text{C=O})$ urea amide	<b>H-bonded (Ordered)</b>	<b>I</b>	1648	24.0
	<b>Free</b>	<b>II</b>	1699	32.8
$\nu(\text{C=O})$ ester urethane amide	<b>Free</b>	<b>III</b>	1730	43.2
<b>Total degree of H-bonded</b>			24.0	

**Table S6.** Summary of the Assignments of Deconvoluted Subpeaks for the C=O Stretching Vibration Absorption Band in the FTIR Spectrum of ISO-BPU.

Assignment	Subpeak	Wavenumber(cm <sup>-1</sup> )				Area(%)				
		ISO				ISO				
		1	2	3	4	1	2	3	4	
ν(C=O) urea amide	H-bonded (Ordered)	I	1633	1633	1633	1631	8.6	9.0	6.2	7.5
	H-bonded (Disordered)	II	1663	1662	1667	1660	19.8	24.2	32.2	27.3
	Free	III	1698	1694	1699	1692	22.4	20.2	21.7	20.7
ν(C=O) ester urethane amide	H-bonded (Ordered)	IV	1723	1718	1720	1713	38.4	33.9	26.9	29.2
	Free	V	1736	1737	1732	1733	10.8	12.6	13.0	15.3
	Total degree of H-bonded						66.8	67.1	65.3	64.0

**Table S7.** Summary of the Assignments of Deconvoluted Subpeaks for the C=O Stretching Vibration Absorption Band in the FTIR Spectrum of APTES-BPU.

Assignment	Subpeak	Wavenumber(cm <sup>-1</sup> )				Area(%)				
		APTES				APTES				
		1	2	3	4	1	2	3	4	
ν(C=O) urea amide	H-bonded (Ordered)	I	1634	1639	1633	1631	6.1	11.9	13.6	9.1
	H-bonded (Disordered)	II	1668	1666	1669	1667	24.5	22.6	17.3	22.3
	Free	III	1694	1695	1693	1690	13.3	9.1	10.1	13.6
ν(C=O) ester urethane amide	H-bonded (Disordered)	IV	1705	1701	1706	1702	5.4	26.5	7.9	8.0
	H-bonded (Ordered)	V	1723	1725	1721	1721	44.5	23.1	35.7	32.8
	Free	VI	1739	1736	1735	1735	6.2	6.7	15.4	14.3
Total degree of H-bonded						80.5	84.1	74.5	72.2	

**Table S8.** FTIR Phase Separation Table of the Prepared Bio-based Polyurethane in the Wavenumber Range of 1760–1600  $\text{cm}^{-1}$ .

Symbol	$A_b(\%)$	$A_f(\%)$	R	DPS(%)	DPM(%)
CO-BPU	24.0	76.0	0.32	24.0	76.0
ISO-BPU-1	66.8	33.2	2.01	66.8	33.2
ISO-BPU-2	67.1	32.9	2.04	67.1	32.9
ISO-BPU-3	65.3	34.7	1.88	65.3	34.7
ISO-BPU-4	64.0	36.2	1.78	64.0	36.0
APTES-BPU-1	80.5	19.5	4.13	80.5	19.5
APTES-BPU-2	84.1	15.9	5.29	84.1	15.9
APTES-BPU-3	74.5	25.5	2.92	74.5	25.5
APTES-BPU-4	72.2	27.8	2.60	72.2	27.8

**Table S9.** Normalized Performance Comparison of the Samples Relative to CO-BPU

	CO-BPU	ISO-BPU-2	APTES-BPU-2
Thermal Stability ( $^{\circ}\text{C}$ )	1	1.01	1.02
Mechanical Strength (MPa)	1	3.11	7.02
Acidic Degradation (h)	1	2.74	2.87
Alkaline Degradation(h)	1	2.5	3.45