

# Supporting Information

## Monocomponent Non-Isocyanate Polyurethane Adhesives Based on a Sol-Gel Process

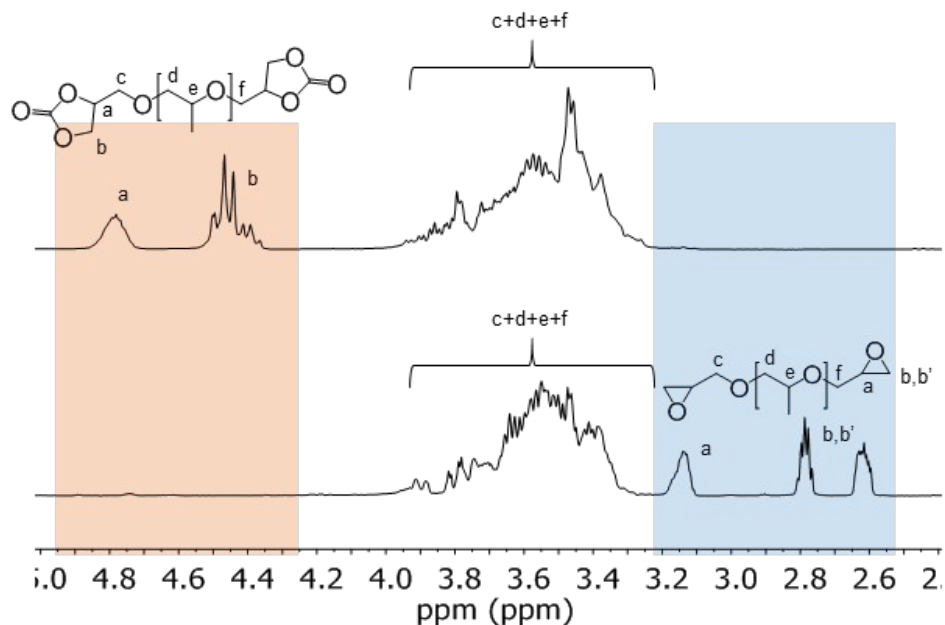
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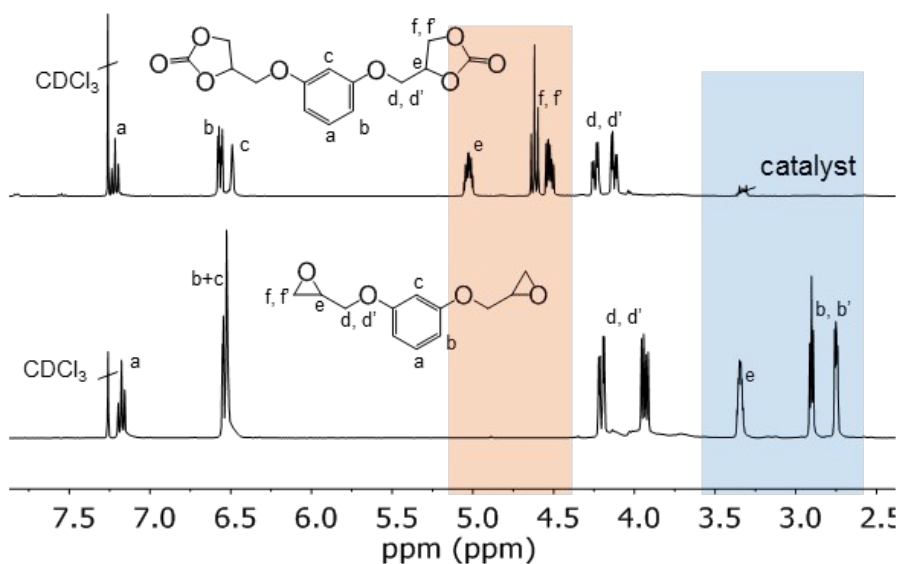
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**Figure S1.** NMR comparison of the diepoxide (below) and dicarbonate (above) PPG. Marked in blue, disappearance of the epoxy signals. Marked in salmon, appearance of cyclic carbonate signals.



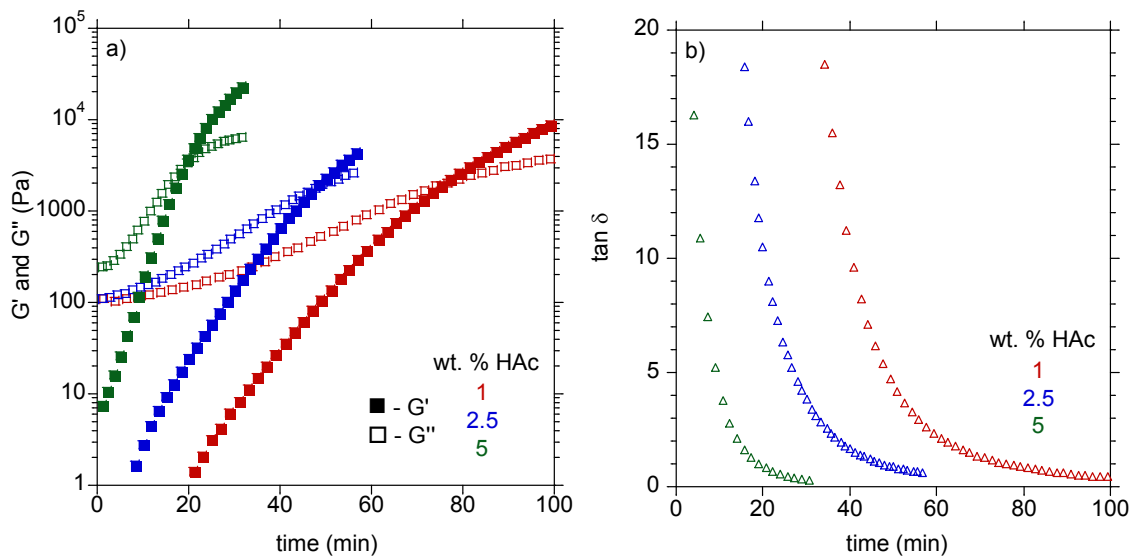
**Figure S2.** NMR comparison of the epoxide (below) and dicarbonate (above) resorcinol. Marked in blue, disappearance of epoxy signals. Marked in salmon, appearance of cyclic carbonate signals.

**Table S1.** AHEW and CEW values of raw materials. <sup>a</sup> Value provide by CRODA.

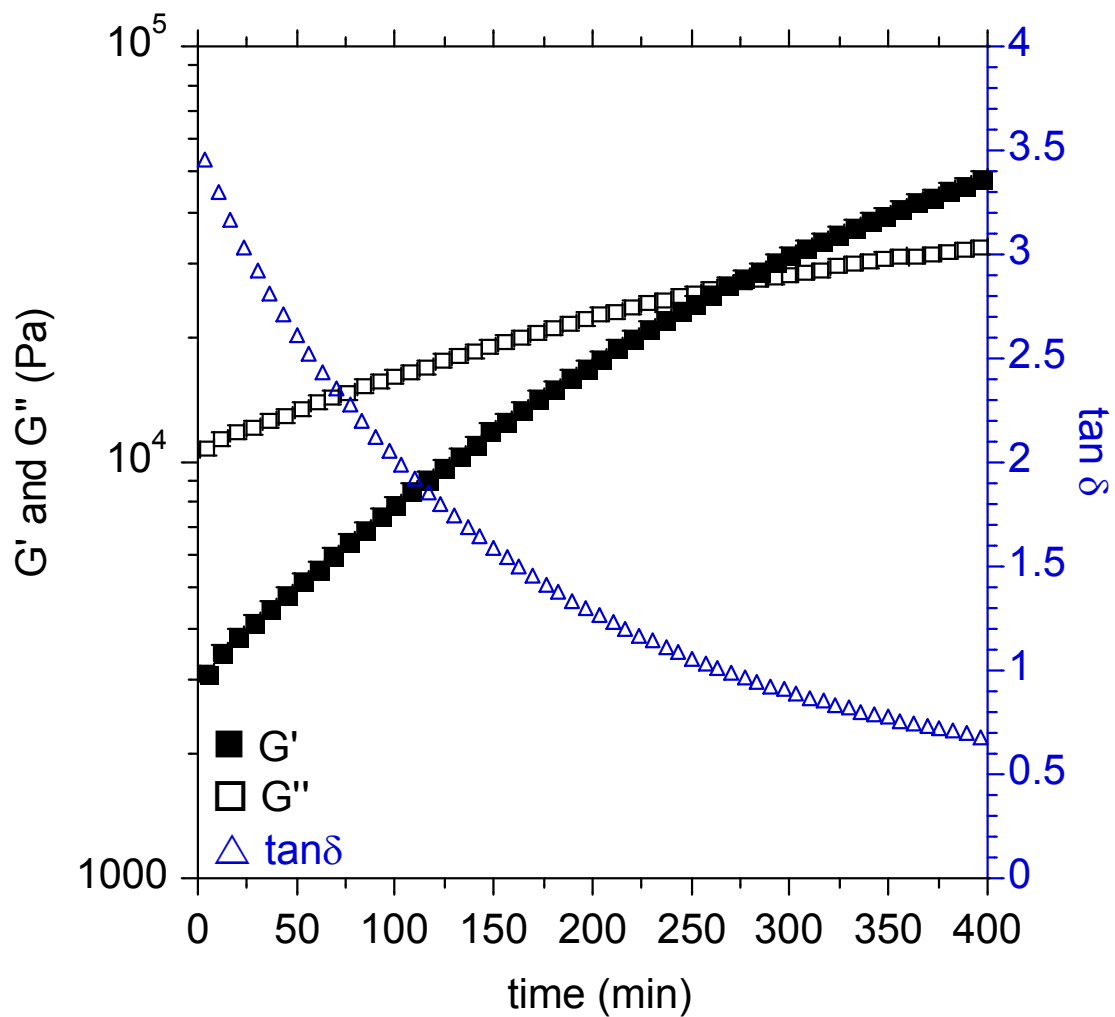
Reactant	CEW (g/eq.)	AHEW (g/eq)
PPGdiCC	333	-
RdiCC	190	-
Priamine 1074 <sup>TM</sup>	-	270 <sup>a</sup>

**Table S2.** Molecular weights of the different compositions of the synthesized PHU prepolymers.

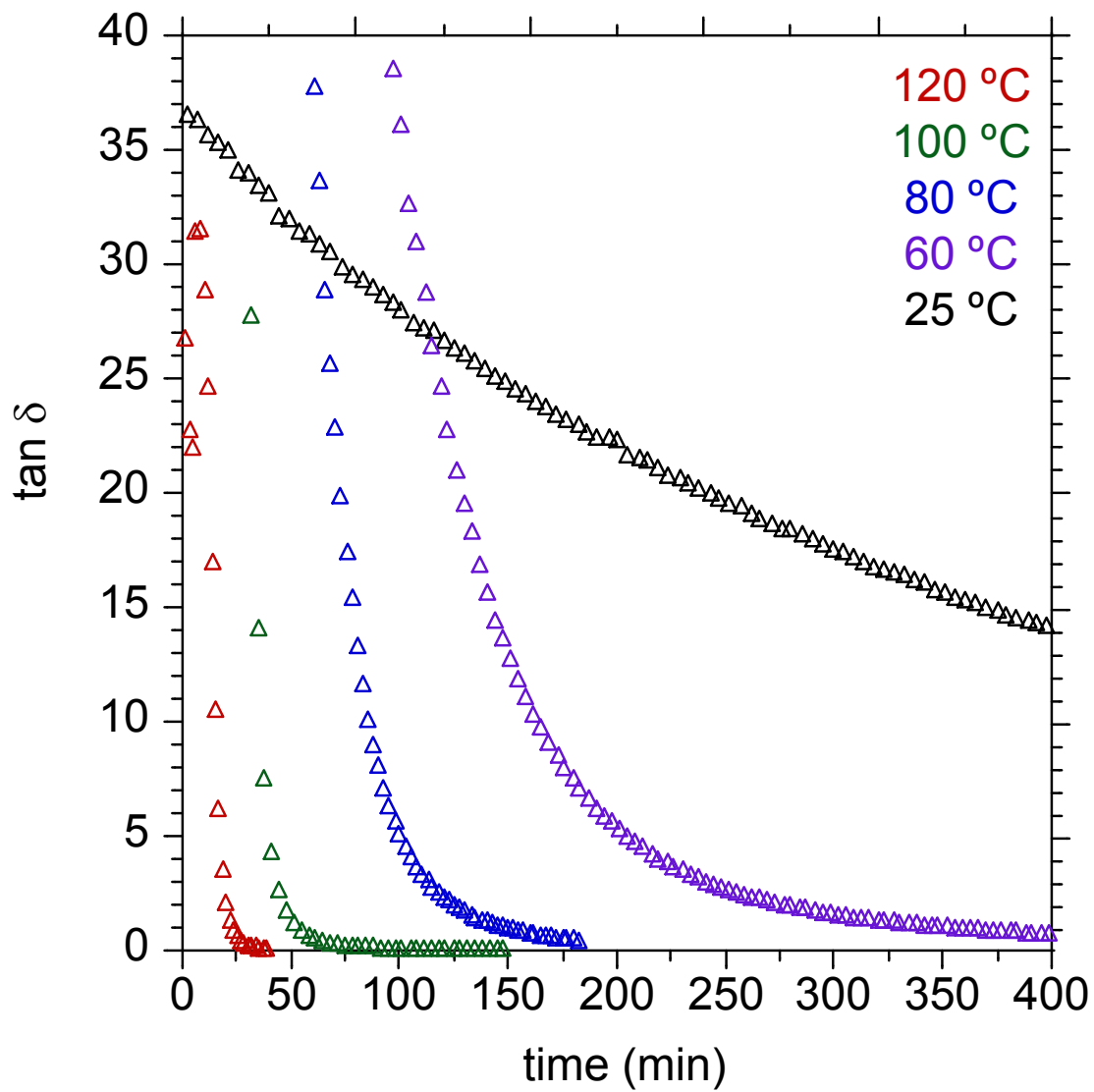
Composition (PPGdiCC/RdiCC)	M <sub>n</sub> (kg/mol)	M <sub>w</sub> (kg/mol)	PDI
100/0	4.4 ± 0.4	10.7 ± 1.2	2.5 ± 0.4
95/5	3.8 ± 0.4	10.4 ± 1.1	2.7 ± 0.6
90/10	4.5 ± 0.3	10.9 ± 0.5	2.4 ± 0.3
80/20	4.4 ± 1.3	11.3 ± 0.7	2.7 ± 0.7
60/40	6.5 ± 1.2	18.6 ± 5.1	2.8 ± 0.3



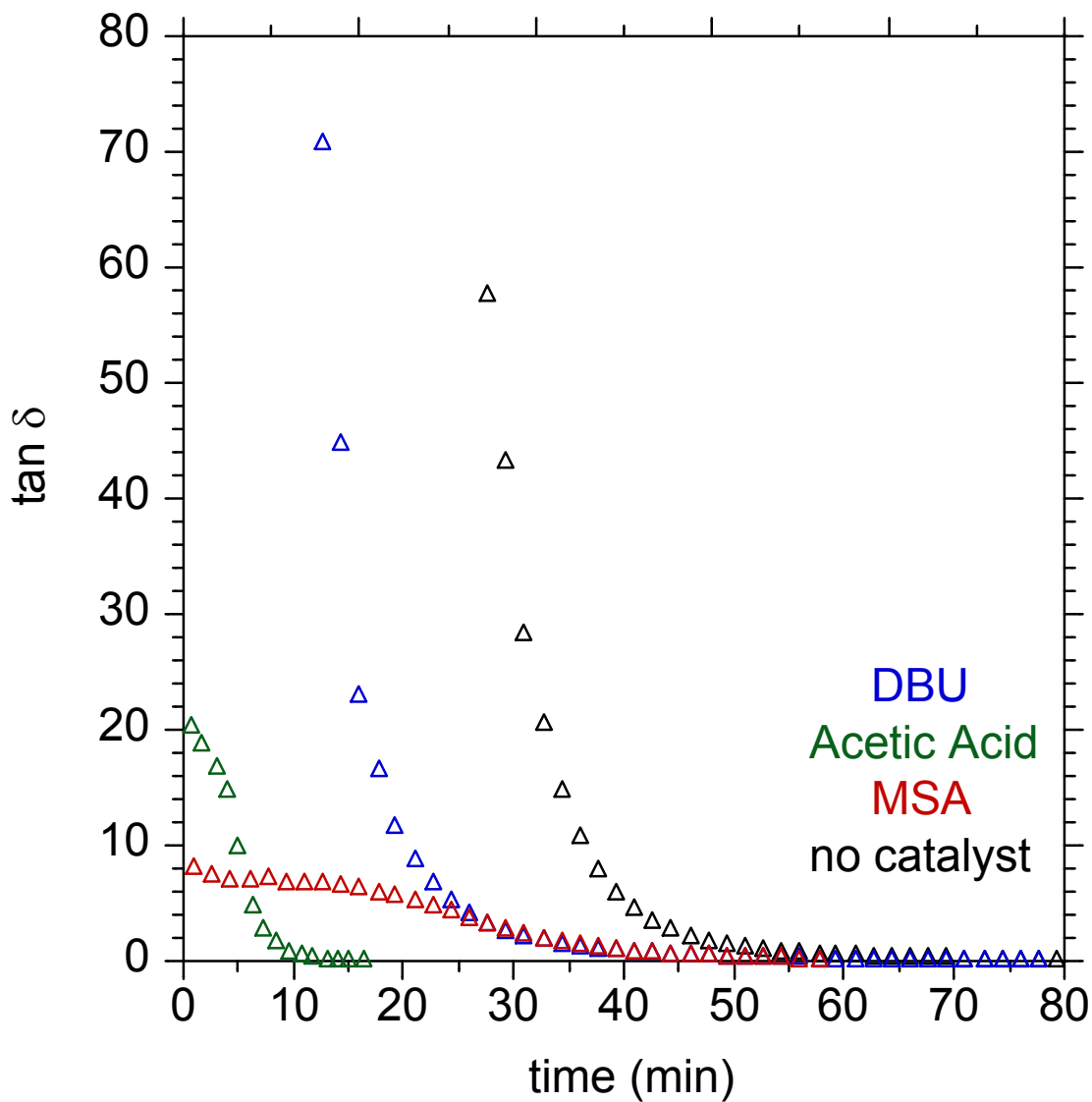
**Figure S3.** a)  $G'$  and  $G''$  and b)  $\tan \delta$  values for rheological measurements employing 1, 2.5 and 5 wt. % of acetic acid at 60 °C.



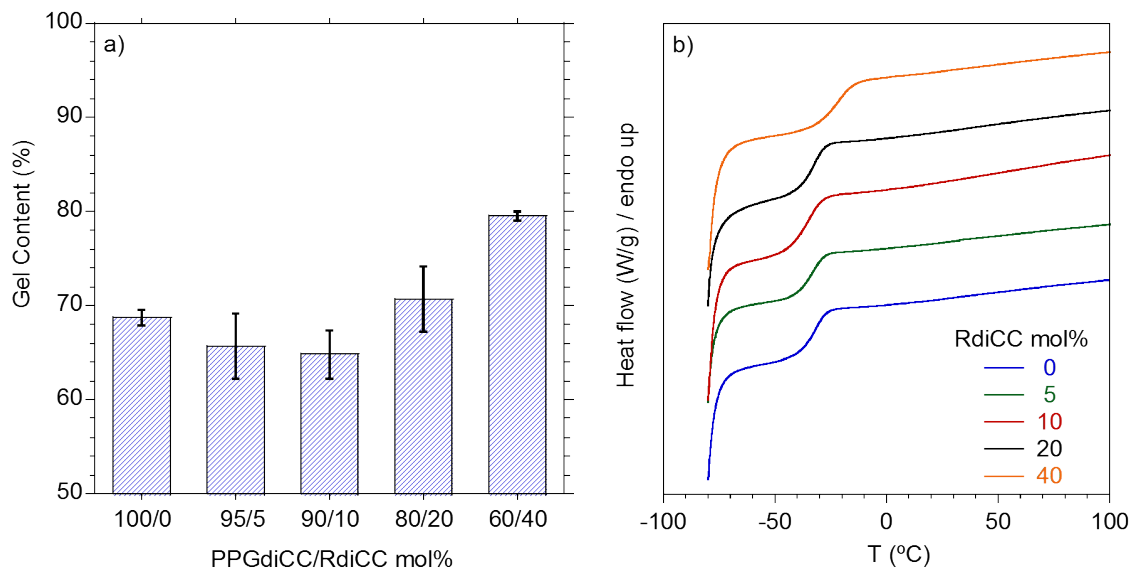
**Figure S4.**  $G'$ ,  $G''$  and  $\tan\delta$  values for rheological measurements employing 2.5 wt. % of acetic acid at 25 °C.



**Figure S5.** Tan $\delta$  values of the rheological measurements of the system depending on temperature. (Related to Figure 3a of the manuscript).



**Figure S6.** Tan $\delta$  values for rheological measurements carried out at 100 °C in the presence of the different catalysts. (Values related to Figure 4b in the manuscript).



**Figure S7.** a) Gel content and b) DSC temperature sweeps for the different ratios of PPGdiCC/RdiCC explored.

**Table S3.** Values of shear strength for the different formulations and the type of failure.

Entry	Composition (PPGdiCC/RdiCC)	Lap-Shear strength (MPa)	Nature of adhesive failure
1	100/0	1.1 ± 0.2	Cohesive
2	95/5	1.5 ± 0.2	Cohesive
3	90/10	1.9 ± 0.2	Cohesive
4	80/20	1.6 ± 0.1	Cohesive
5	60/40	2.9 ± 0.6	Cohesive
6	60/40 <sup>a</sup>	2.8 ± 0.8	Cohesive

**Table S4.** Summary of results for the formulations prepared at 25 °C. <sup>a</sup> Equilibrium water content measured after 96 h immerse in water; <sup>b</sup> gel content after soxhlet extraction in boiling THF for 24h; <sup>c</sup> glass transition temperature; <sup>d</sup> temperature at which sample lost 5 weight%. \*No gel content, all sample was dissolved in THF.

Entry	Composition (PPGdiCC/RdiCC)	Catalyst	EWC <sup>a</sup> (%)	GC <sup>b</sup> (%)	T <sub>g</sub> <sup>c</sup> (°C)	Td5% <sup>d</sup> (°C)
1		HAc (2.5 wt%)	28.8 ± 0.4	40.9 ± 1.1	-30	215
2	60/40	HAc (5 wt%)	30.0 ± 3.3	18.6 ± 3.6	-26	222
3		HAc (7.5 wt%)	27.2 ± 2.3	*	-29	202