

Supporting Information

Multi-responsive γ -Methylene- γ -Butyrolactone/*N*-Vinyl Caprolactam Copolymers Involving pH-depend Reversible Lactonization

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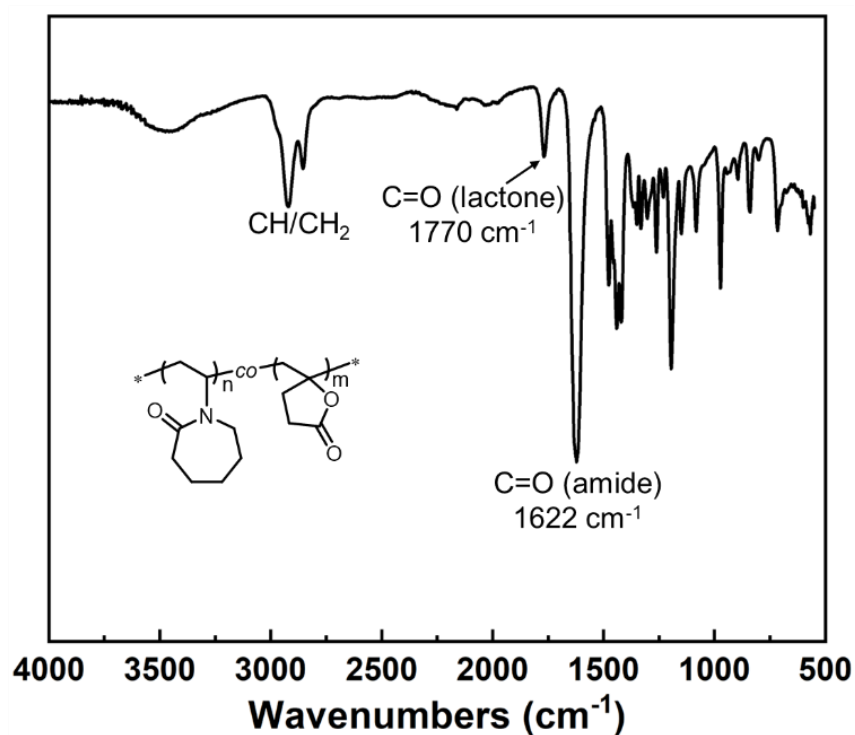


Figure S1. FT-IR spectrum of P(NVCL-co- γ M γ BL) ($M_n = 20000$ g/mol, $F_{\gamma M \gamma BL} = 0.05$) prepared by conventional radical polymerization (Table 1, entry 1).

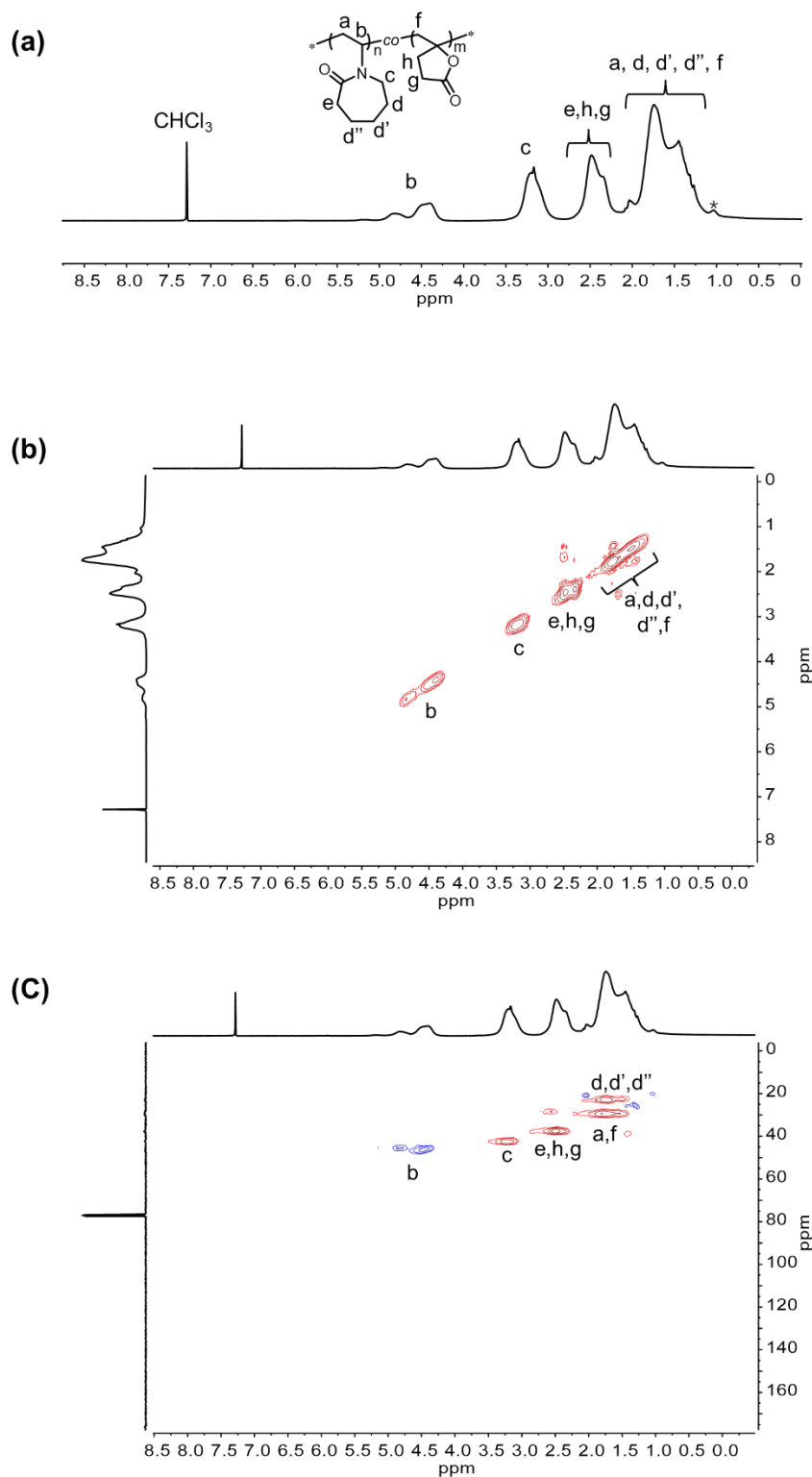


Figure S2.(a) ^1H NMR, (b) COSY and (c) HSQC of P(NVCL-*co*- $\gamma\text{M}\gamma\text{BL}$) ($M_n = 11500$ g/mol, $F_{\gamma\text{M}\gamma\text{BL}} = 0.1$) prepared by OMRP (Table 2, entry 4). * ^1H signal of the methyl groups of the initiating fragment R (R = $-\text{C}(\text{CN})(\text{CH}_3)-\text{CH}_2-\text{C}(\text{CH}_3)_2\text{OCH}_3$).

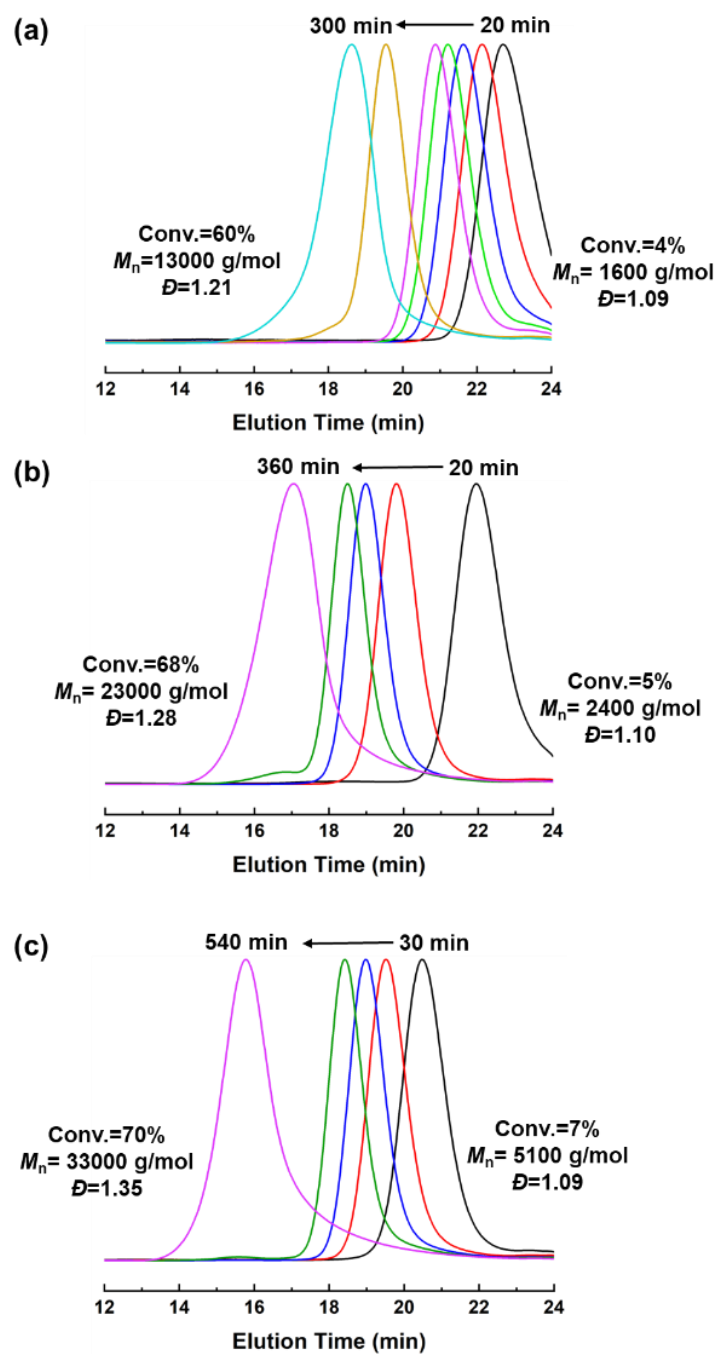


Figure S3. Overlay of SEC traces for the OMRP of NVCl and $\gamma M \gamma BL$ with different monomers/RCo molar ratios: (a) $[\text{monomers}]_0/[\text{R-Co(acac)}_2]_0=250/1$, (b) $[\text{monomers}]_0/[\text{R-Co(acac)}_2]_0=500/1$, (c) $[\text{monomers}]_0/[\text{R-Co(acac)}_2]_0=750/1$. ($f^\circ_{\gamma M \gamma BL} = 0.1$, Table 2 entries 1-3).

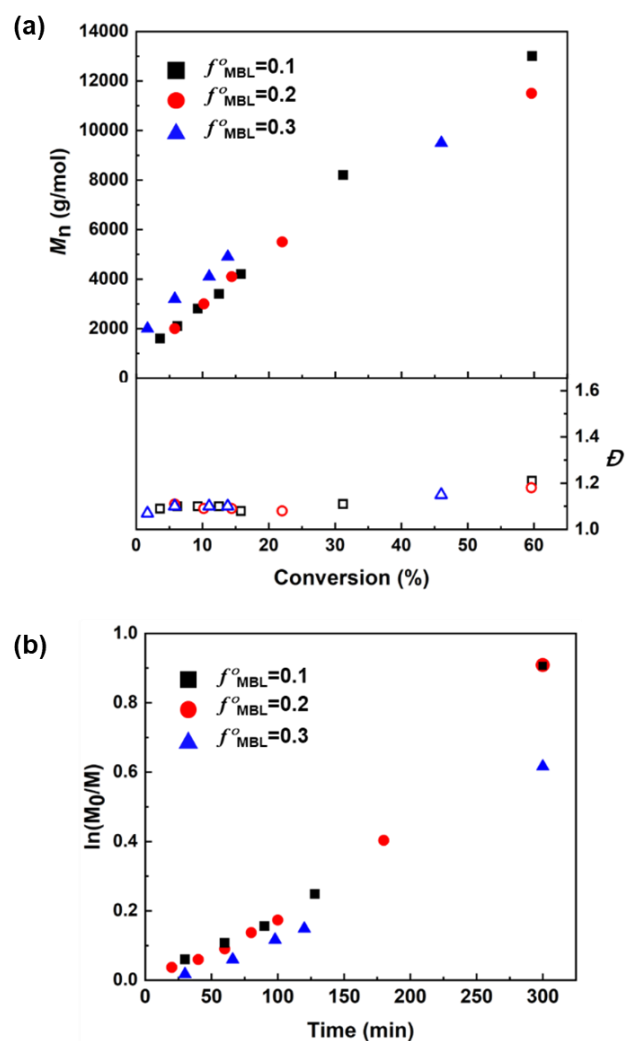


Figure S4. (a) Dependence of M_n (full symbols) and \mathcal{D} (hollow symbols) on the total monomers conversion and (b) time dependence of $\ln(M_0/M)$ for the OMRP of NVCL/γMγBL with different feed ratio: \blacksquare $[\text{NVCL}]_0/[\gamma\text{M}\gamma\text{BL}]_0=0.9/0.1$, \bullet $[\text{NVCL}]_0/[\gamma\text{M}\gamma\text{BL}]_0=0.8/0.2$, \blacktriangle $[\text{NVCL}]_0/[\gamma\text{M}\gamma\text{BL}]_0=0.7/0.3$ ($[\text{comonomers}]_0/[\text{R-Co}(\text{acac})_2]_0=250/1$, Table 2 entries 1, 4 and 5).

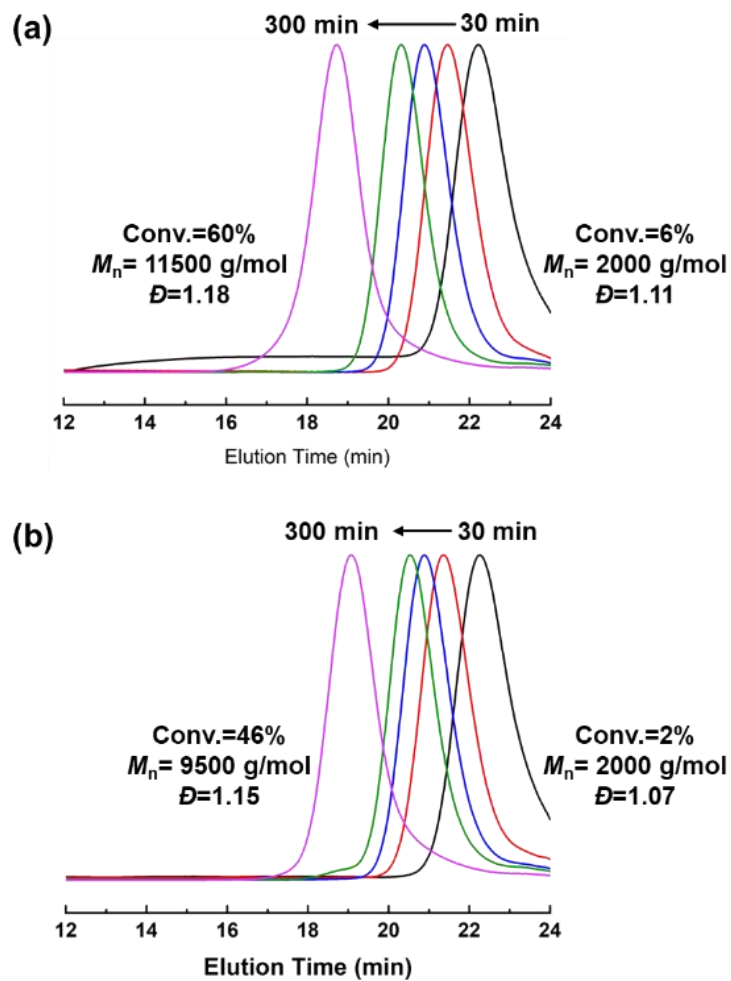


Figure S5. Overlay of SEC traces for the OMRP of NVCl and $\gamma M \gamma BL$ with different feed compositions: a) $[NVCl]_0/[\gamma M \gamma BL]_0 = 0.8/0.2$, b) $[NVCl]_0/[\gamma M \gamma BL]_0/[R-Co(acac)_2]_0 = 0.7/0.3$. ($[comonomers]_0/[R-Co(acac)_2]_0 = 250/1$, Table 2 entries 4 and 5).

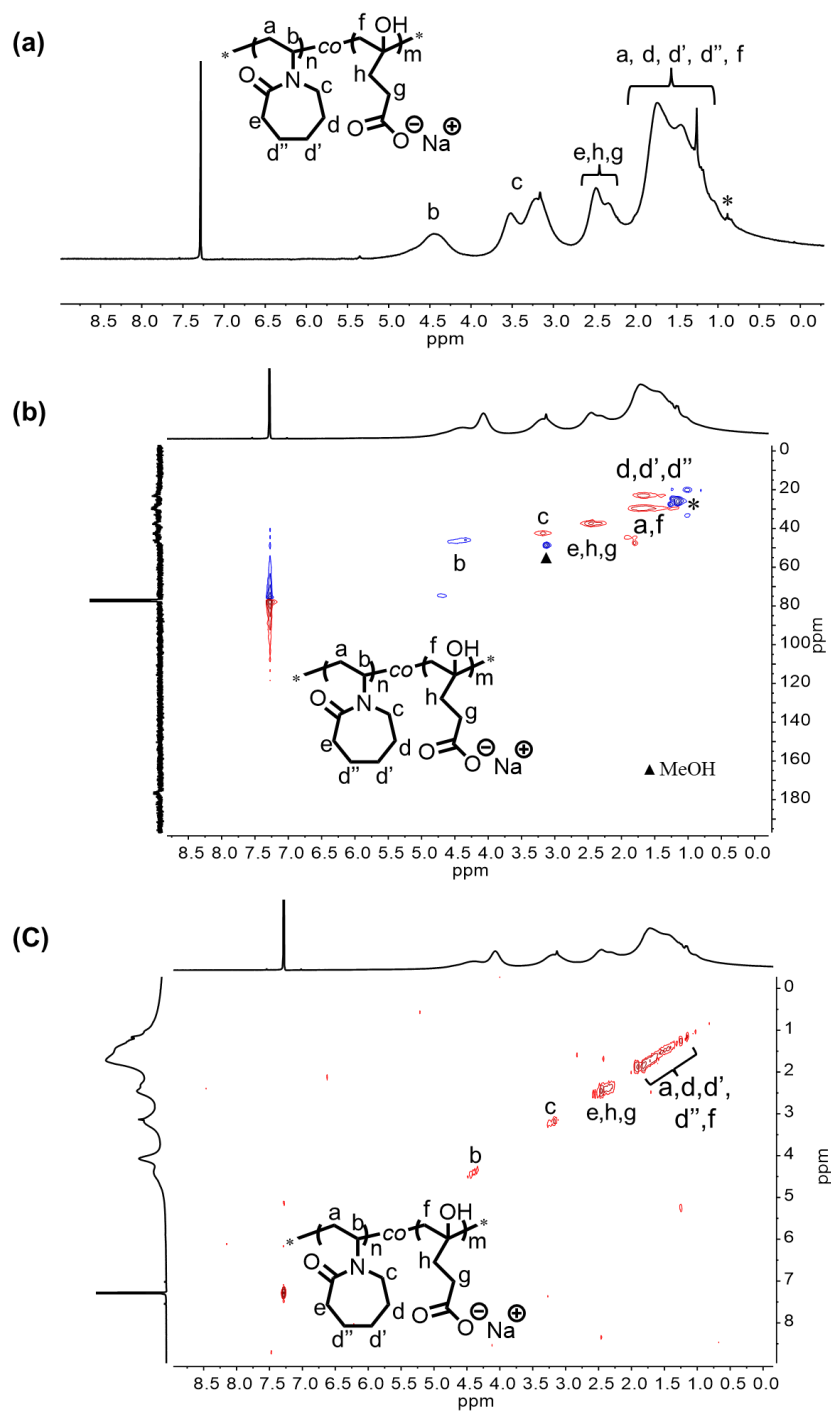


Figure S6. (a) ^1H NMR, (b) HSQC and (c) COSY of P(NVCL-co-HPEA) (Precursor: P(NVCL-co- γ M β BL) ($M_n = 11500$ g/mol, $F_{\gamma\text{M}\beta\text{BL}} = 0.1$) prepared by OMRP (Table 2, entry 4). $^*^1\text{H}$ signal of the methyl groups of the R initiating fragment (R= $-\text{C}(\text{CN})(\text{CH}_3)-\text{CH}_2-\text{C}(\text{CH}_3)_2\text{OCH}_3$).

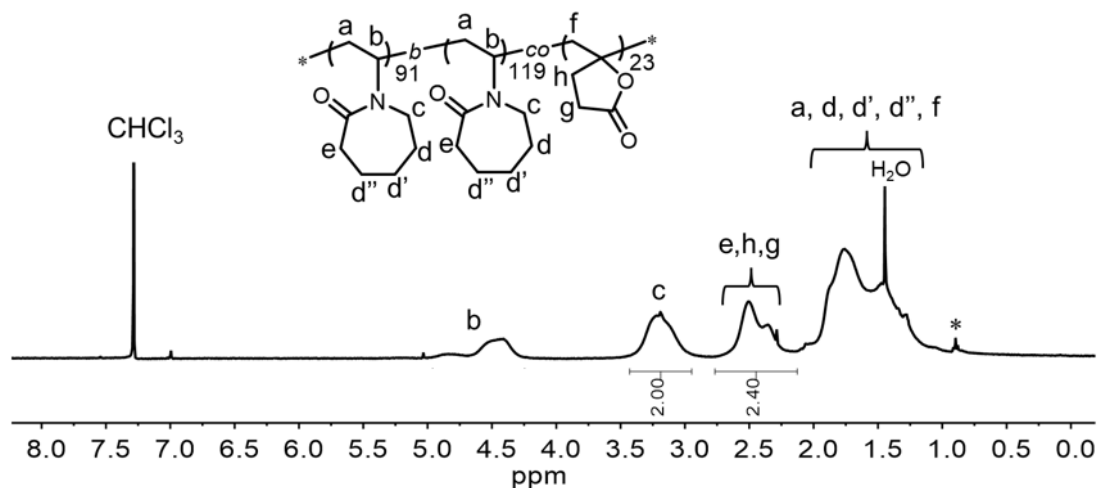


Figure S7. ¹H NMR spectrum of P(NVCL)₉₁-b-P(NVCL)₁₁₉-co-γMγBL₂₃. *¹H signal of the methyl groups of the R initiating fragment (R= -C(CN)(CH₃)-CH₂-C(CH₃)₂OCH₃).

Table S1. Surface zeta potential of P(NVCL)₉₁-b-(NVCL)₁₁₉-co-HPEA₂₃ particles in aqueous solution (5mg/mL) at different temperatures.

Temperature (°C)	65	70	80
Zeta potential (mV)	-41.04	-51.85	-55.25