

## Supporting Information

### <sup>1</sup>H NMR Analysis of Polymers

A representative analysis is provided for amine polymer PA1 and guanidine polymer PG1. Peak A of SI1 and SI2 accounts for two protons from the R group of the RAFT agent. Comparing the integration of this peak to those from pendant methylenes gives the degree of polymerization (DP) for each polymer.

**For amine polymer PA1:** Peak B of SI1 constitutes two protons of the amine monomers. This equates to an average 13.553 amine pendants per polymer chain ( $=27.106/2$ ). Peak C constitutes 3 protons from the methyl monomers, giving an average of 7.190 per chain ( $=21.571/3$ ). This gives an overall DP of 20 ( $13.553+7.190$ , rounded to the nearest digit) and represents a 35% methyl content overall ( $=7.190/(13.553+7.190)$ ).

The final estimate of Mn is hence:  $MW_{RAFT} + DP_{AEMA} \times MW_{AEMA} + DP_{MMA} \times MW_{MMA}$

$403.67 + (13.553 \times 165.62) + (7.190 \times 100.12) = 3370 \text{ g mol}^{-1}$  rounded to 3 significant figures.

**For guanidine polymer PG1:** Peak B of SI2 constitutes two protons of the guanidine monomers. This equates to an average 11.056 guanidine pendants per polymer chain ( $=22.112/2$ ). Similarly, the peak seen at 3.60 ppm is the sum of integrations for a methylene group of the guanidine (C, 2 protons) and the methyl ester D (3 protons). Therefore, the average number of methyl groups per polymer chain equates to  $(43.395 - 22.112)/3 = 7.0943$ . This gives an overall DP of 18 ( $11.056 + 7.0943$ , rounded to the nearest digit) and represents a 39% methyl content overall ( $=7.0943/(11.056 + 7.0943)$ ).

The final estimate of Mn is hence:  $MW_{RAFT} + DP_{GEMA} \times MW_{GEMA} + DP_{MMA} \times MW_{MMA}$

$403.67 + (11.056 \times 171.20) + (7.0943 \times 100.12) = 3360 \text{ g mol}^{-1}$  rounded to 3 significant figures.

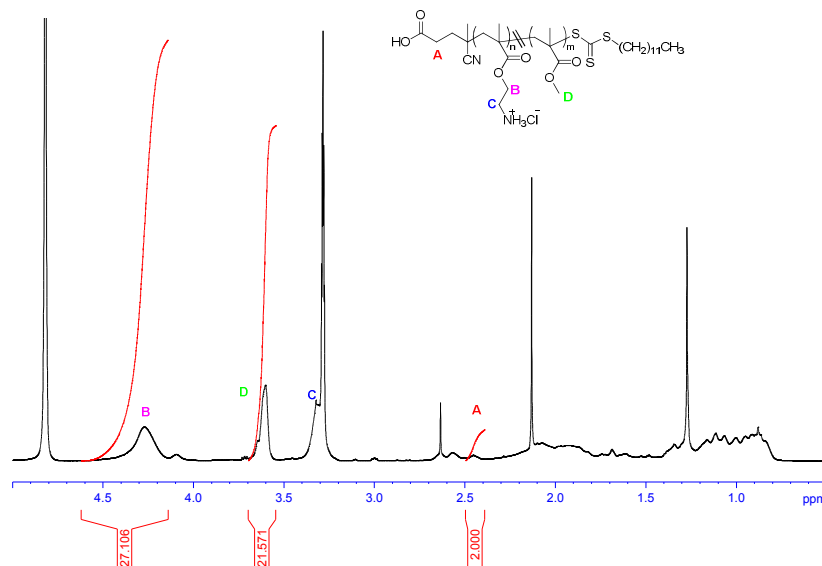


Figure SI1. <sup>1</sup>H NMR spectrum of amine polymer PA1.

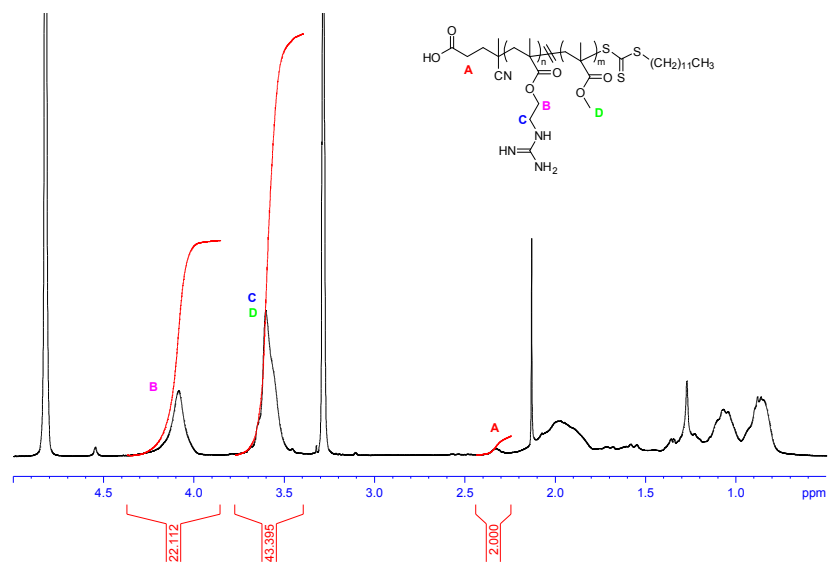


Figure S12.  $^1\text{H}$  NMR spectrum of guanidine polymer PG1.

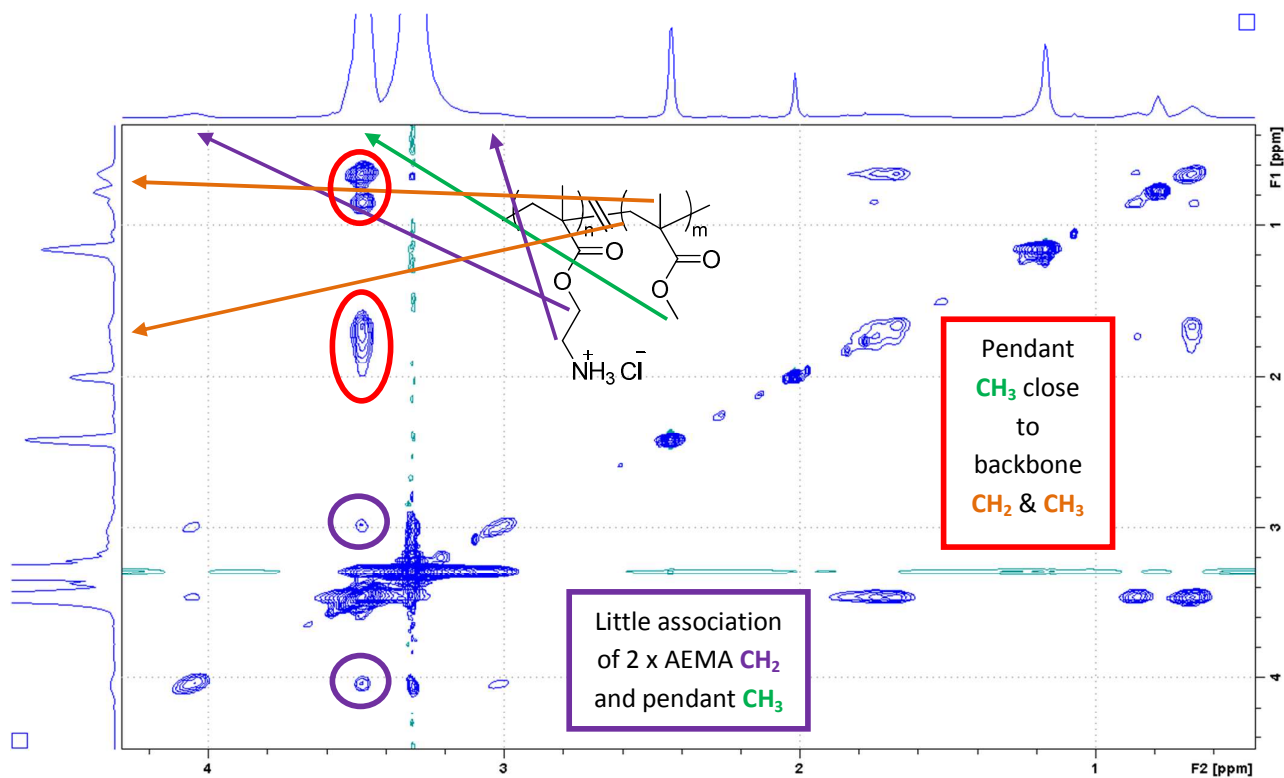
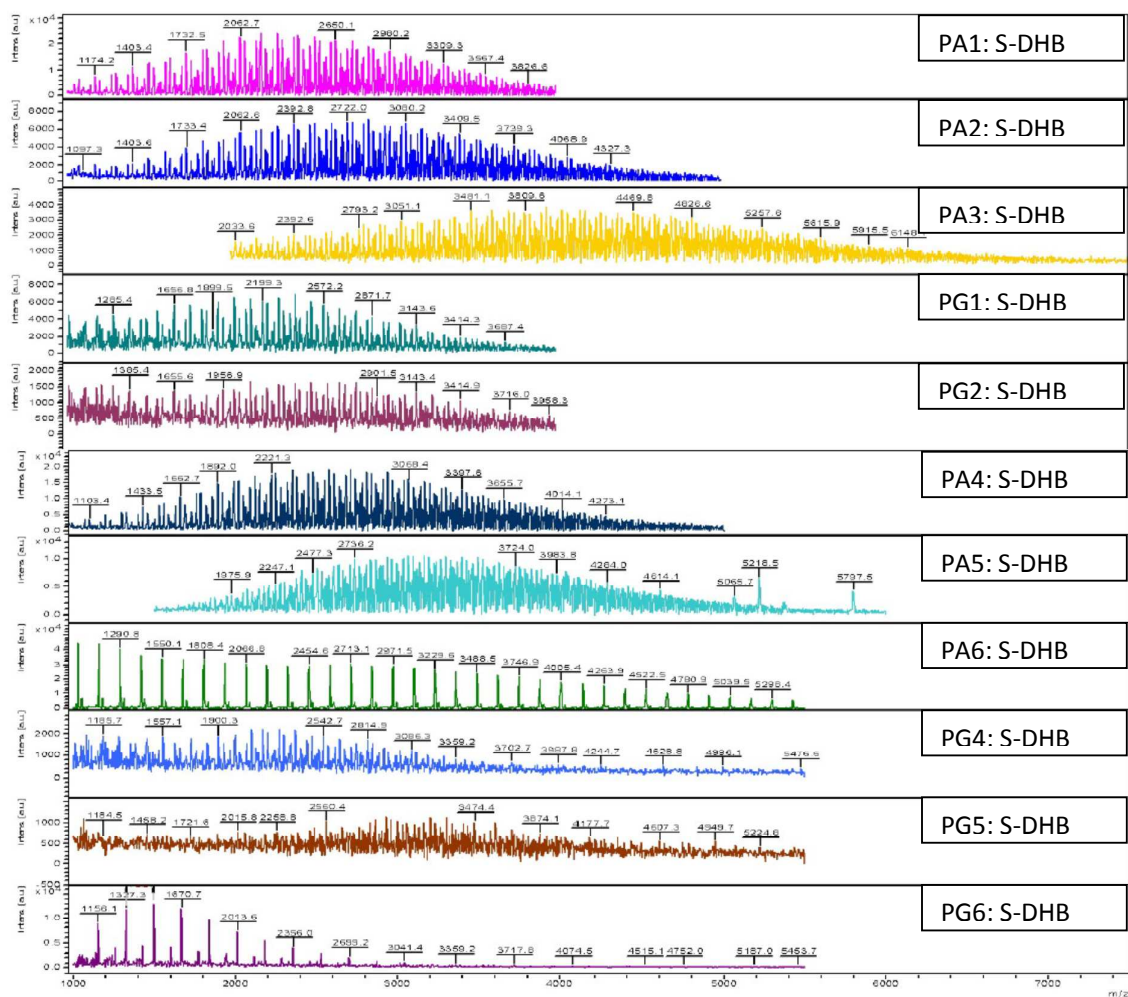


Figure S13. NOESY spectrum of amine polymer PA5



**Figure S14.** Representative MALDI-ToF spectra for the amine and guanidine series generated using S-DHB at the cationic matrix