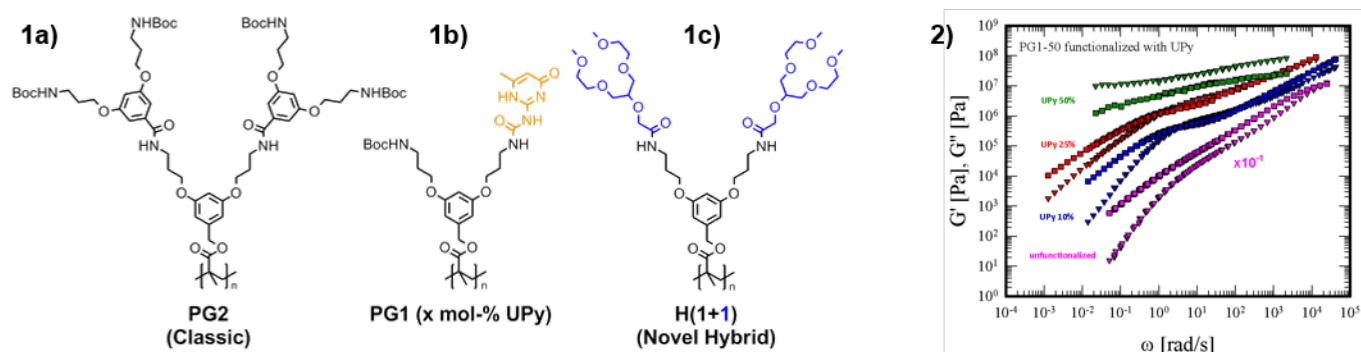


## Effect of supramolecular interactions in dendronized polymers on their thermal- and viscoelastic properties

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Dendronized polymers (DPs) represent an intriguing class of macromolecules combining the concepts of dendrimers and polymers [1]. By tailoring their thickness and persistence length through the generation number ( $g$ ) and backbone degree of polymerization ( $P_n$ ), respectively, DPs can be endowed with a wide range of conformations, spanning from flexible polymers to shape-anisotropic colloidal objects.



In order to approach the intrinsic properties of these interesting materials, we performed modifications to our prototype “classic” DPs [2] (Fig. 1a) aimed at enhancing or suppressing their intermolecular hydrogen-bonding interactions through incorporation of strongly hydrogen-bonding ureidopyrimidinone (UPy) moieties (Fig. 1b) or hybridization with oligo(ethylene glycol)-based dendrons [3] (Fig. 1c), respectively. We present the effects of  $g = 1-3$ , various  $P_n$ s and chemical modifications on the thermomechanical properties of these novel DPs, which have been systematically investigated by DSC and rheology. The studied DPs undergo very slow ageing owing to the reduced global mobility of these bulky molecules. Even so, segmental mobility is increased in the hybrid DPs and constrained in the UPy DPs, which translates into vastly different equilibration times and glass transition temperatures. The very large entanglement molecular weight of the classic DPs causes a frequency dependent rheological response, which is typically characterized by low values of the entanglement plateau modulus in the low frequency regime, similarly to bottlebrush polymers but tuneable through both  $g$  and  $P_n$ . At it, UPy-functionalization broadens the scope of rheological responses even further (Fig. 2). Our results demonstrate remarkable effects of the chemical structure on the viscoelastic properties of such super soft elastomers with ultra-high molar mass and pave the way into exciting applications.

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