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The effect of surface conditions on the glass-transition dynamics of poly(phenylmethylsiloxane) confined in cylindrical nanopores

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In this contribution, we demonstrate the results of the study on the effect of surface conditions on the polymer glass-transition dynamics in nanopore confinement. The tested material is poly(phenyl methyl siloxane) (PMPS) embedded in alumina nanopores with modified surface properties. The main experimental techniques used are dielectric spectroscopy, differential scanning calorimetry, and contact angle measurements. Two different strategies were used to modify the character of the inner surface of the pores: (i) functionalization with the use of highly polar phosphoric acid units separated from each other by non-polar triethoxysilane groups (N = number of spacers N changes from 0 to 24) and via (ii) atomic layer deposition technique. Changes in the surface conditions dramatically affect interactions between the polymer and the confining surface as reflected in the values of the interfacial free energy changing from 0.5 mN/m up to 18.7 mN/m. Surprisingly we found that the characteristic “two glass transitions event” seen in nanopore confinement does not vanish, even in the absence of strong interfacial interactions. In this way, changes in the glass-transition behavior of the tested polymer in confined geometry cannot be rationalized in terms of the differences in polymer/substrate interfacial energy. Eliminating strongly adhered surfaces does not eliminate the puzzling two-T_{gs} effect seen in cylindrical nanopores.

References: *Macromolecules* 2021, 54, 23, 10951–10968, *Macromolecules* 2022, 55, 8, 3208–3220

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