Supporting Information

Molecular Dynamics Simulations of Hydrated Poly(amidoamine) Dendrimer /Graphene-Oxide Nanocomposite Membranes

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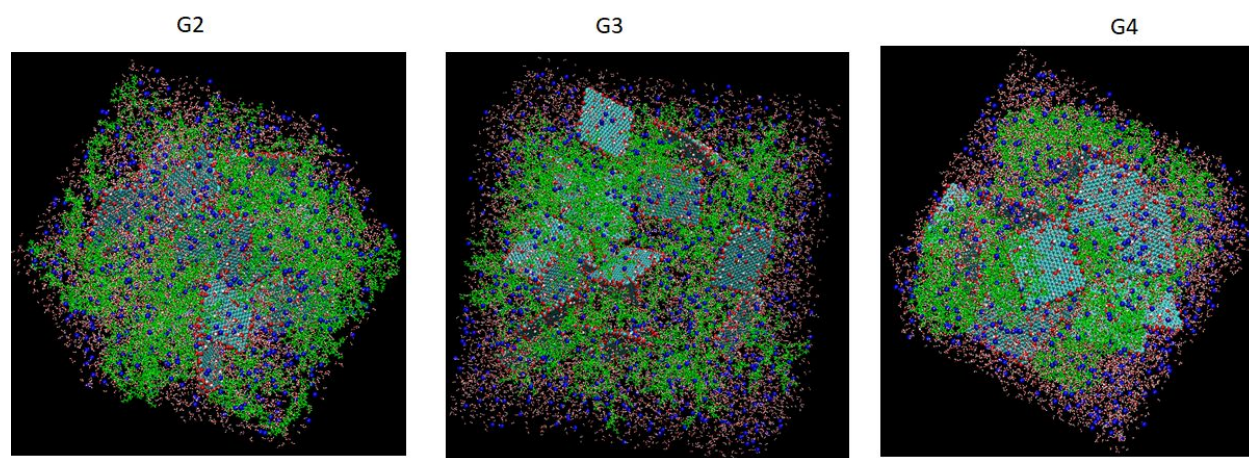
Figure S1: Initial structures of the GOPAMAM2 (G2), GOPAMAM3(G3) and GOPAMAM4(G4) models. Dendrimer molecules are shown in green, GO flakes as dark cyan (with red and white beads corresponding to the oxygen and the hydrogen atoms, respectively) and counterions in blue. Atoms of water molecules are shown as red (oxygen) and white (hydrogens) dots.
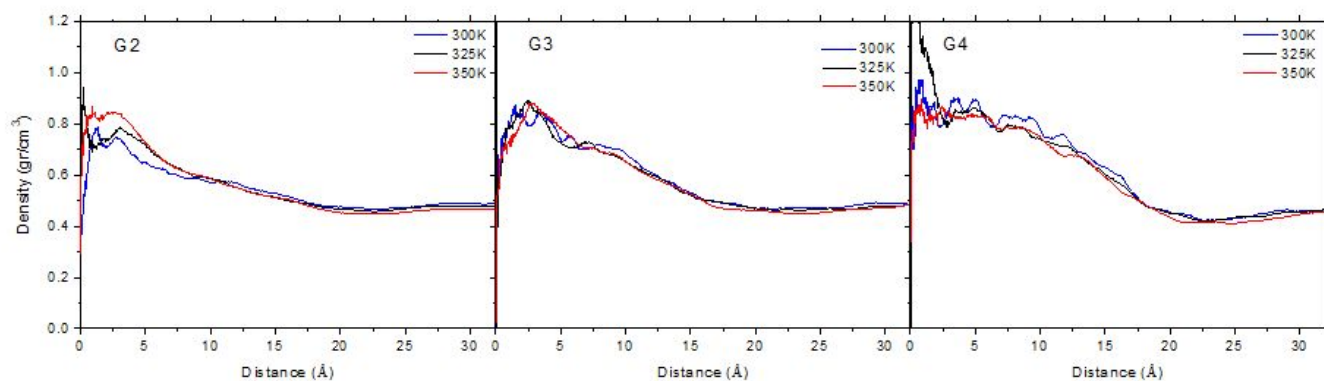
Figure S2: Density distributions of the dendrimer molecules as a function of the distance from the center of mass of the dendritic structure, for the GOPAMAM2 (G2), the GOPAMAM3 (G3) and the GOPAMAM4 (G4) models.
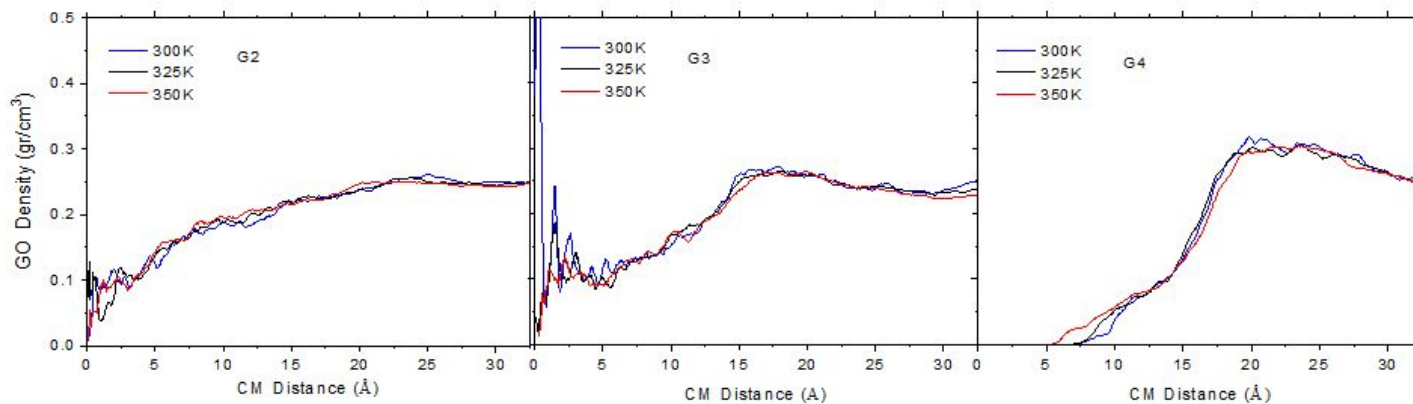

Figure S3: Density profiles of GO with respect to the center of mass of the dendritic molecules for the GOPAMAM2 (G2), the GOPAMAM3 (G3) and the GOPAMAM4 (G4) models, at all the examined temperatures.
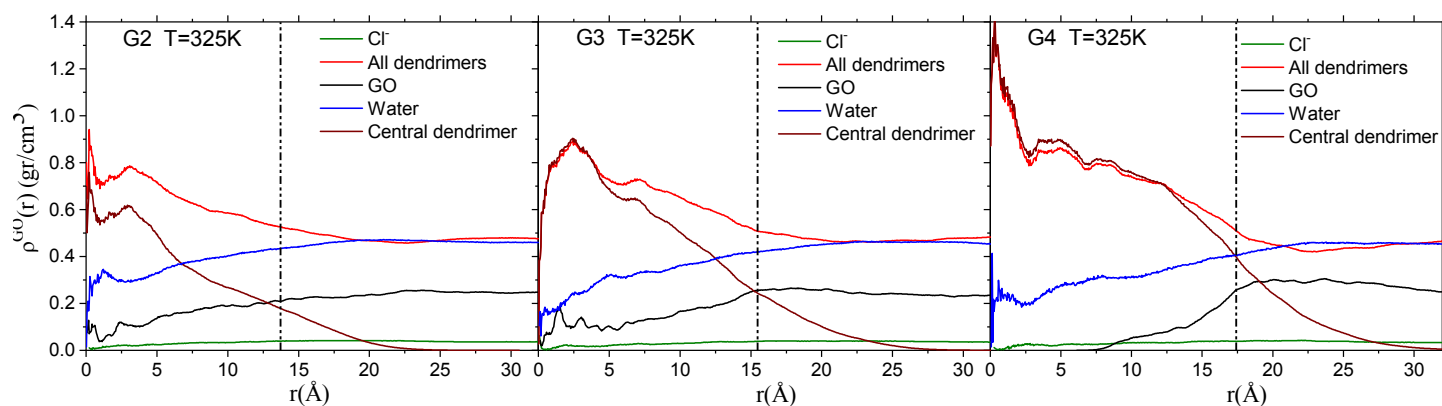
S2

Figure S4: Density profiles of all the components with respect to the center of mass of the dendritic molecules for the GOPAMAM2 (G2), the GOPAMAM3 (G3) and the GOPAMAM4 (G4) models at T=325K. The dash-dot vertical lines denote the location of the radius of gyration of the dendrimer at each generation. The density profiles at T=300K and T=350K are qualitatively and quantitively very similar to those shown for T=325K.
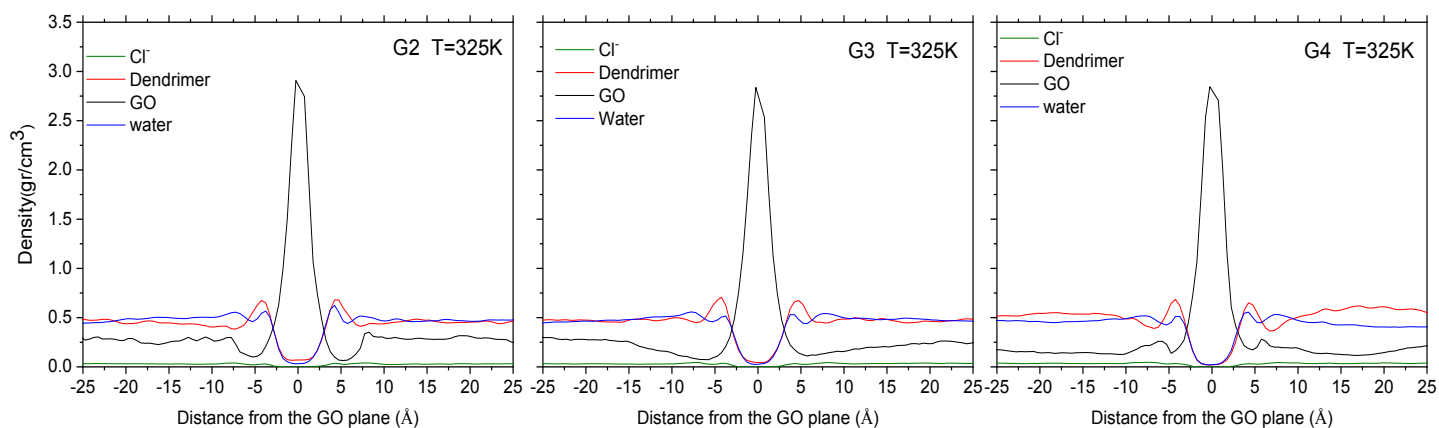

Figure S5: Density profiles of the different constituents of the composites, in a direction normal the GO plane. Position 0 denotes the location of the reference GO plane. The profiles at different temperatures is similar.
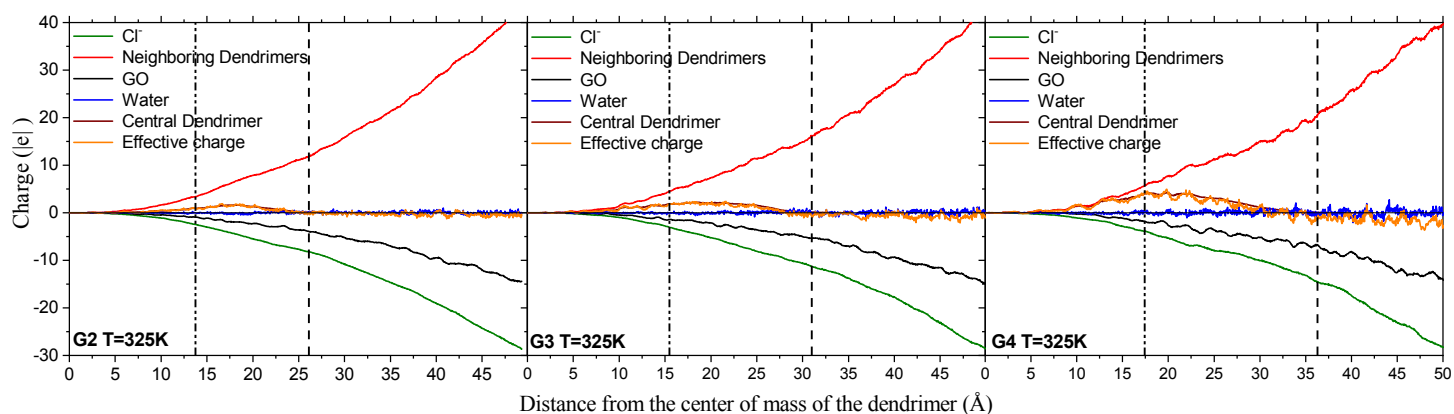
Figure S6: Radial charge distributions arising from the different moieties, with respect to the center of mass of a dendrimer, at T=325K. The behavior at the other temperatures is similar. Dashed-dot lines denote the location of the radius of gyration of the dendrimer molecules and dashed lines their boundary.
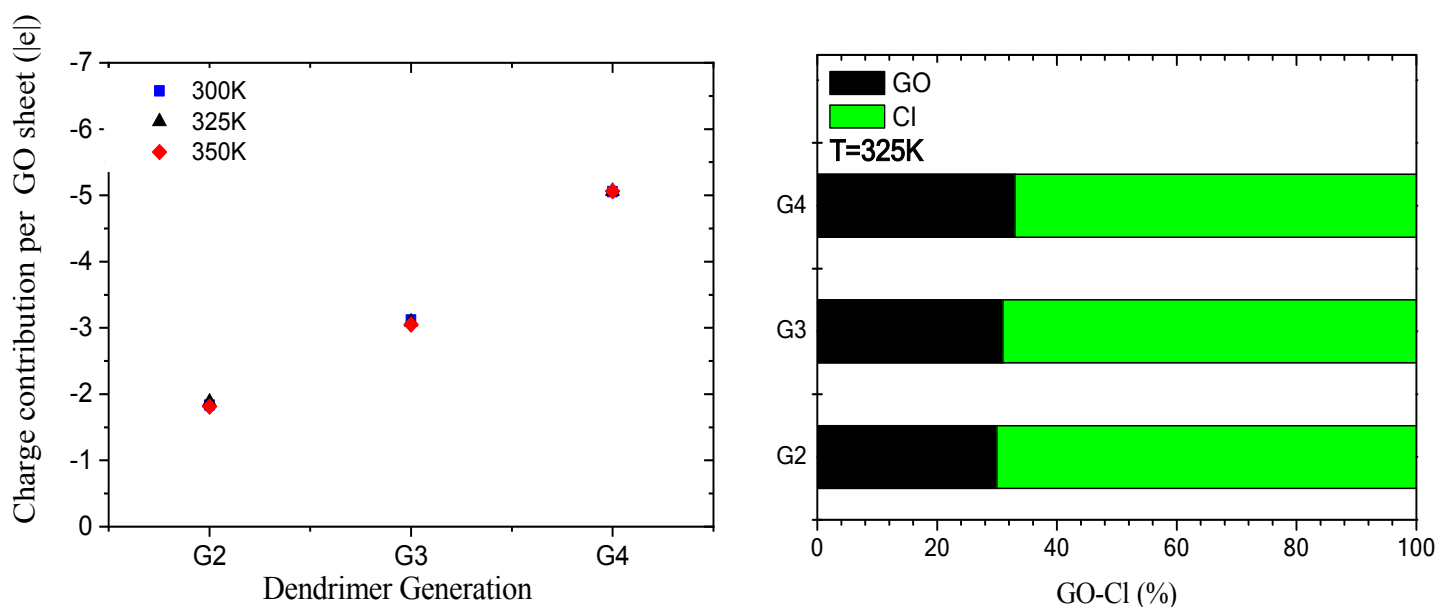

Figure S7: (Left): charge per GO sheet contributing to the neutralization of a dendrimer's positive charge. (Right) : relative contribution of the negative charges arising from the GO flakes and the

Cl⁻ counterions, to the neutralization of the charge of the PAMAM molecules at T=325K. The behavior at the other temperatures is similar.
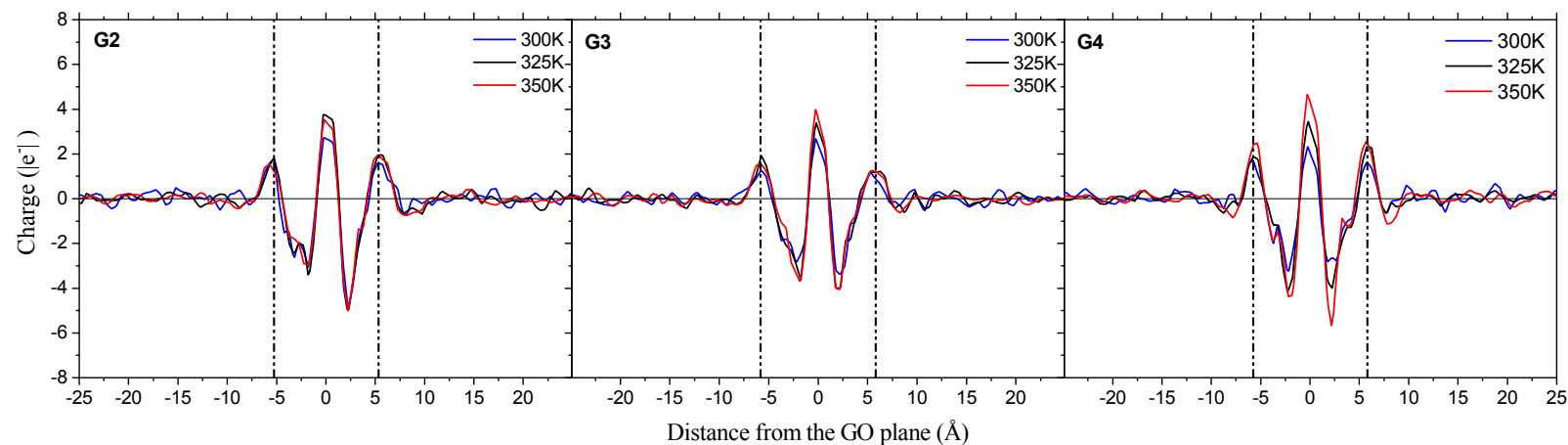

Figure S8: Charge distributions along a direction normal to the GO plane arising from the charged moieties. The 0 value denotes the location of the GO sheet. The vertical dashed lines delimit the boundaries of a GO sheet, as follows from the location of the sharp peaks appearing in the corresponding density profiles appearing in figure 7 of the main text.
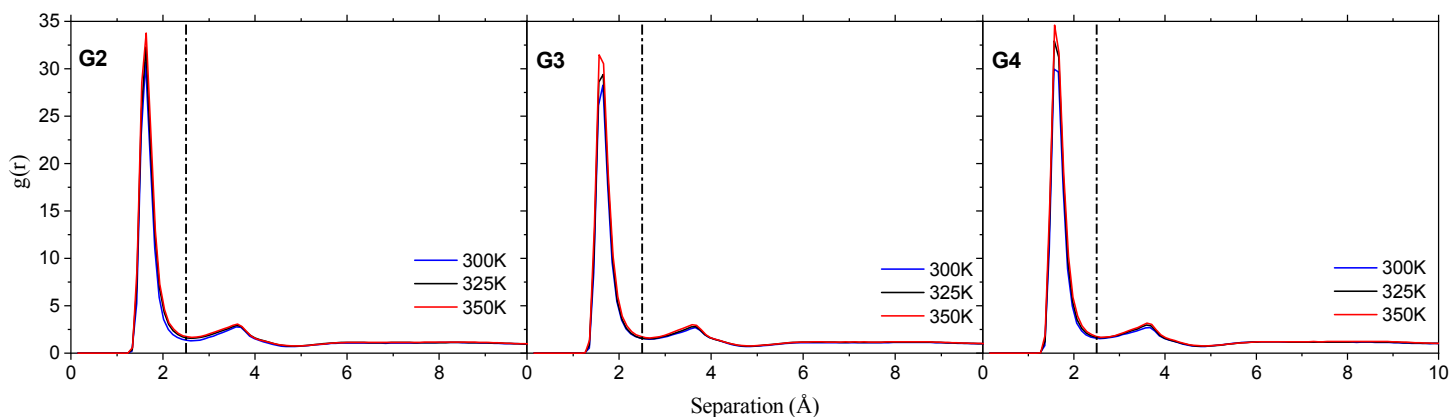

Figure S9: Pair distribution functions of hydrogens belonging to the protonated primary amines of the dendrimers and oxygens belonging to the negatively charged hydroxyl groups of the GO. Only pairs for which the acceptor-hydrogen-donor angle was higher than 120º were considered. The dashed-dotted line denotes the first minimum of the distribution functions.
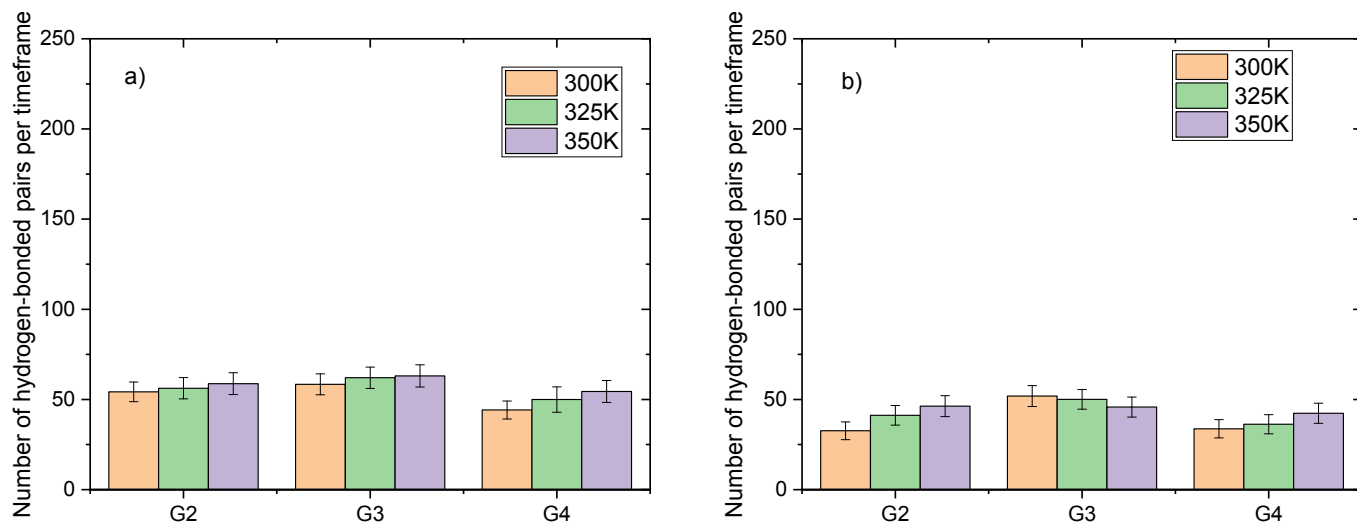
Figure S10: Average number of hydrogen bonds per saved timeframe, formed between the hydrogens of the protonated amine groups of the dendrimers and a) the hydroxyl oxygens of GO , b) the epoxide oxygens of GO.
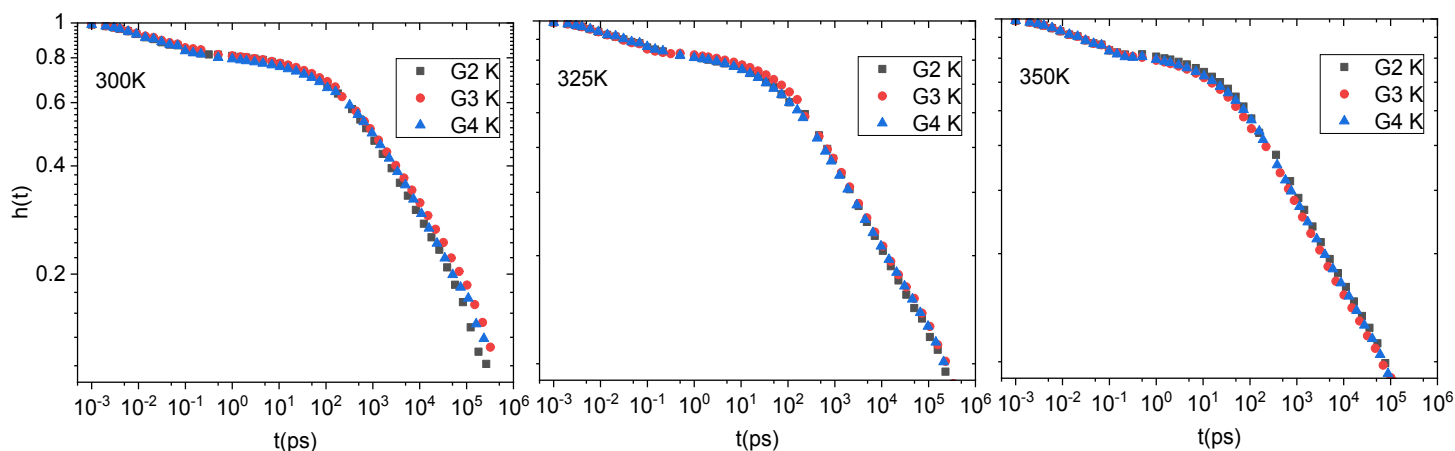

Figure S11: Hydrogen bonding correlations functions (eq. 1 in the main text) in log-log representation, between the hydrogens of the protonated amine groups of the dendrimers and the carboxyl oxygens of GO, for the GOPAMAM2 (G2), GOPAMAM3 (G3) and the GOPAMAM4 (G4) models, at T=300K, T=325K and T=350K.
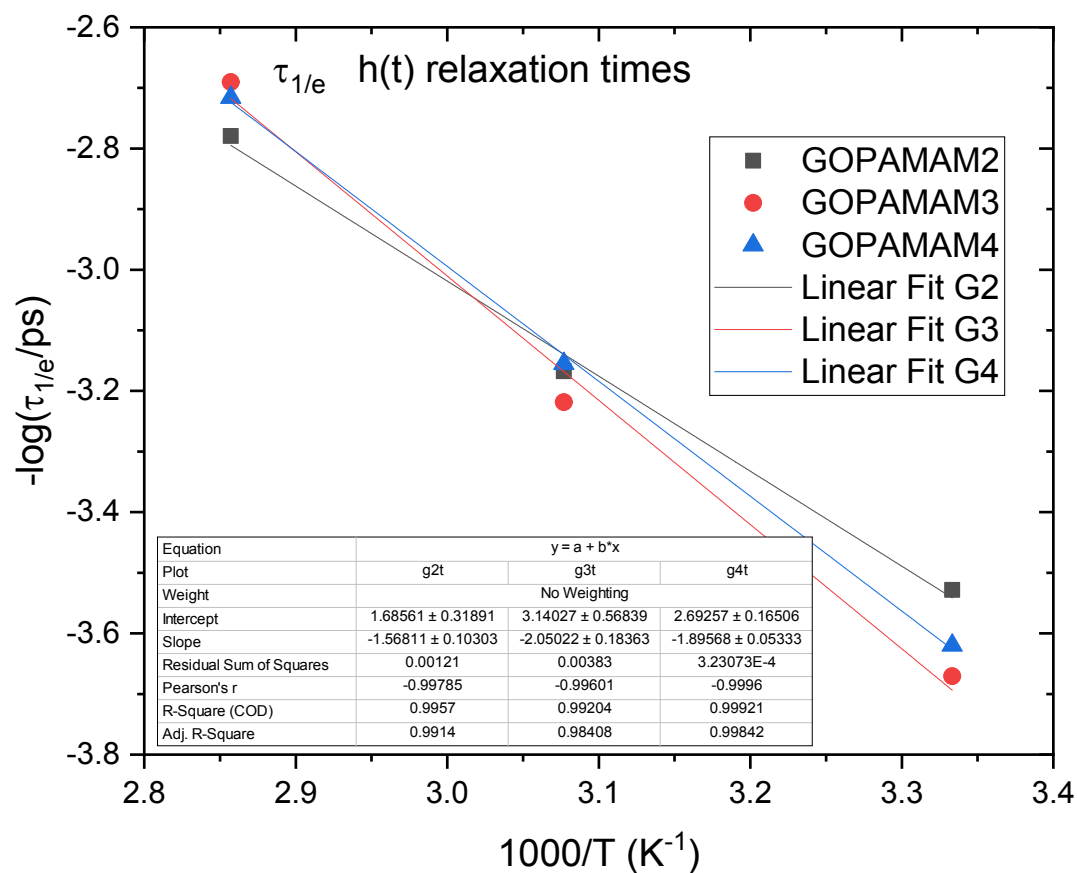
Figure S12: Linear fits through the data points referring to the systems with the 3 different dendrimer generations. Fit parameters are shown in the inset table.

The inset table within the figure reads:

Equation	y = a + b*x		
Plot	g2t	g3t	g4t
Weight	No Weighting		
Intercept	1.68561 ± 0.31891	3.14027 ± 0.56839	2.69257 ± 0.16506
Slope	-1.56811 ± 0.10303	-2.05022 ± 0.18363	-1.89568 ± 0.05333
Residual Sum of Squares	0.00121	0.00383	3.23073E-4
Pearson's r	-0.99785	-0.99601	-0.9996
R-Square (COD)	0.9957	0.99204	0.99921
Adj. R-Square	0.9914	0.98408	0.99842