

# Entrapped Styrene Butadiene Polymer Chains by Sol-Gel Derived Silica Nanoparticles with Hierarchical Raspberry Structures

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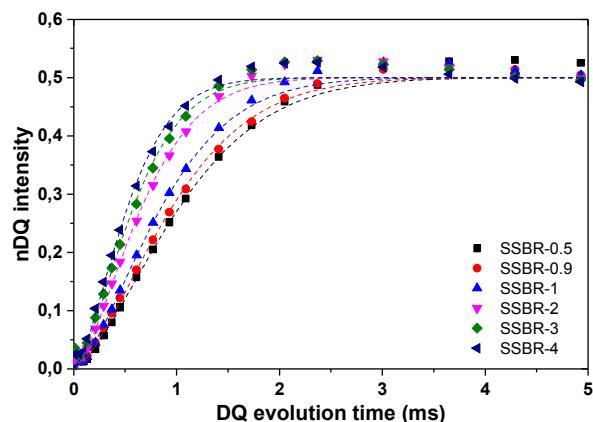
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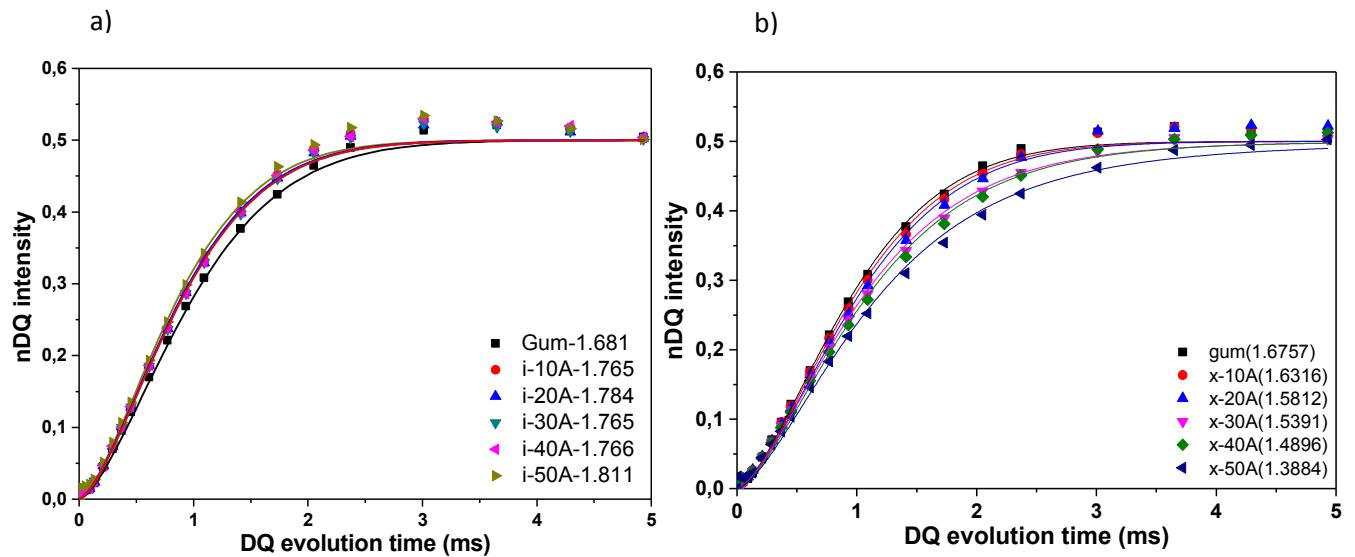
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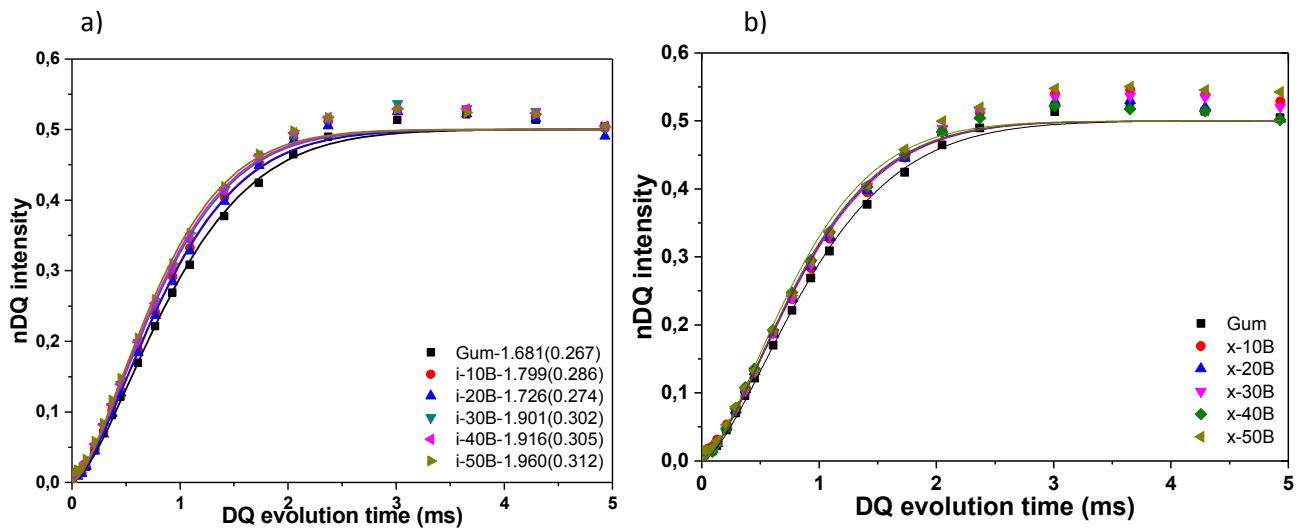
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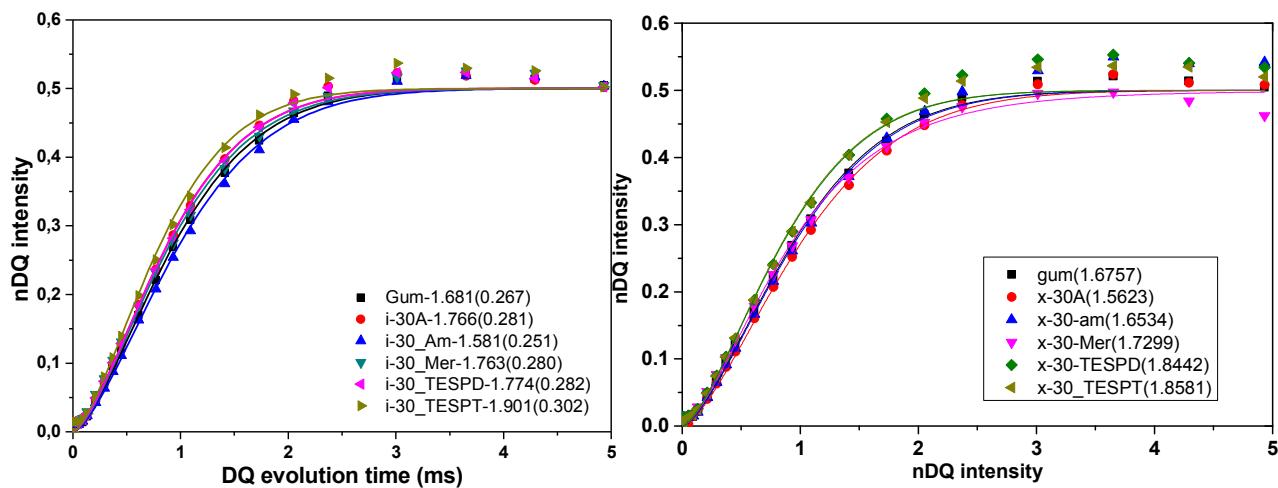
**Fig. S1: Double-quantum buildup curves of different rubber system along with fits:- Effect of sulfur loading on crosslink density of rubber**



**Fig. S2: a) Effect of alkoxy-based (abSiO<sub>2</sub>) and b) precipitated silica (pSiO<sub>2</sub>) loading on crosslink density of rubber**



**Fig. S3: a) Effect of alkoxy (abSiO<sub>2</sub>) and b) precipitated silica (pSiO<sub>2</sub>) loading with presence of TESPT coupling agent on crosslink density of rubber**



**Fig. S4: Effect of different silane coupling agent on cross link density of rubber as reflected in DQ NMR results.**

**Table S1: Crosslink densities of different silica filled SSBR composites as estimated by NMR and swelling**

Silica (in phr)	$D_{res}/2\pi$ (kHz)				$V_{FR}$ (mol/kg)			
	i	i-TESPT	x	x-TESPT	i	i-TESPT	x	x-TESPT
<b>gum</b>	0.268				0.146			
<b>10</b>	0.281	0.286	0.259	0.291	0.154	0.155	0.149	0.158
<b>20</b>	0.284	0.274	0.252	0.294	0.174	0.206	0.153	0.188
<b>30</b>	0.281	0.302	0.245	0.296	0.213	0.272	0.157	0.220
<b>40</b>	0.281	0.305	0.237	0.298	0.221	0.302	0.159	0.258
<b>50</b>	0.288	0.312	0.221	0.307	0.289	0.397	0.161	0.273

**Table S2: Crosslink densities of various silane-modified silica-filled SSBR composites as estimated by NMR and swelling**

30 phr of silica	$D_{res}/2\pi$ (kHz)		VFR(mol/kg)	
	abSiO <sub>2</sub>	pSiO <sub>2</sub>	abSiO <sub>2</sub>	pSiO <sub>2</sub>
<b>Gum</b>	0.268			0.146
<b>Pristine</b>	0.281	0.245	0.212	0.157
<b>DMS</b>	0.277	0.258	0.209	0.144
<b>APTES</b>	0.252	0.263	0.200	0.155
<b>ODTES</b>	0.255	0.253	0.207	0.185
<b>MPTES</b>	0.280	0.275	0.252	0.206
<b>NXT</b>	0.262	0.259	0.251	0.207
<b>TESPD</b>	0.282	0.293	0.264	0.203
<b>TESPT</b>	0.303	0.296	0.272	0.220