Supplementary Information

Assessing the Impact on the Glass Transition Temperature of Bituminous Binder from Ultra-thin Diamond Nanothread

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S1. Model information for the DNT modified bituminous binder

Details of the atomistic models established in this work. Here DNT with random dispersion and uniform alignment are named as BB-DNT^{R} and BB-DNT^{A}, respectively. The incorporation of functional groups in a 5.05wt% system leads to a variation in the weight percentage since the calculation of DNT mass involves the inclusion of functional groups. Moreover, the addition of the same number of DNTs may also result in fluctuation of wt% due to the difference in the number of bitumen molecules in different systems.

Table S1. Detailed composites of DNT modified bitumen models.

Model	Numbers of molecules					Total atom numbers	DNT wt%
	Asphaltene	Naphthene	Polar aromatics	Saturate	DNT		
Neat	99	149	170	467	0	51421	0.00
BB-5.06wt%DNT^{AI}	96	143	164	451	12	52052	5.07
BB-5.06wt%DNT^{AII}	96	144	164	451	12	52076	5.06
BB-5.06wt%DNT^{AIII}	96	144	164	451	12	52076	5.06
BB-5.06wt%DNT^{AIV}	96	143	164	451	12	52052	5.07
BB-5.06wt%DNT^{AV}	96	144	164	452	12	52144	5.06
BB-1.29wt%DNT^{R}	98	147	168	463	3	51524	1.29
BB-2.56wt%DNT^{R}	97	146	167	459	6	51681	2.56
BB-3.81wt%DNT^{R}	97	145	165	455	9	51919	3.81
BB-5.05wt%DNT^{R}	96	144	165	453	12	52242	5.05
BB-6.30wt%DNT^{R}	95	142	163	447	15	52209	6.30
BB-8.37wt%DNT^{R}	93	140	160	440	20	52243	8.37
BB-5.81wt%DNT^{R}-$CH_3^{15\%}$	95	142	162	447	12	52053	5.81
BB-6.56wt%DNT^{R}-$C_2H_5^{15\%}$	95	140	160	441	12	51894	6.56
BB-9.09wt%DNT^{R}-$C_6H_5^{15\%}$	93	139	159	437	12	52081	8.96
BB-7.31wt%DNT^{R}-$COOH^{15\%}$	94	142	162	445	12	51806	7.31
BB-6.31wt%DNT^{R}-$C_6H_5^{5\%}$	94	141	162	444	12	51726	6.31
BB-7.78wt%DNT^{R}-$C_6H_5^{10\%}$	94	140	160	441	12	52038	7.78
BB-8.96wt%DNT^{R}-$C_6H_5^{15\%}$	93	139	159	437	12	52081	8.96
BB-10.47wt%DNT^{R}-$C_6H_5^{20\%}$	91	137	157	431	12	51943	10.47
BB-5.79wt%DNT^{R}-$COOH^{5\%}$	95	142	162	447	12	51729	5.79
BB-6.61wt%DNT^{R}-$COOH^{10\%}$	95	142	163	447	12	51939	6.61
BB-7.31wt%DNT^{R}-$COOH^{15\%}$	94	142	162	445	12	51806	7.31
BB-8.15wt%DNT^{R}-$COOH^{20\%}$	94	141	161	443	12	51796	8.15

S2. Calculation of elastic modulus performed by tensile simulation.

The elastic modulus is calculated from E=σ/ε, where σ refers to the stress and ε refers to the strain. We simulated the system under tensile stress in the x-direction at a temperature of T=300K and pressure of p=1atm. The total strain during the tensile process was 20%. Based on the stress-strain curve, the elastic range is selected as the strain changes from 0 to 0.06. Taking the stress-strain within this range, the calculated elastic modulus is about 1153 MPa.

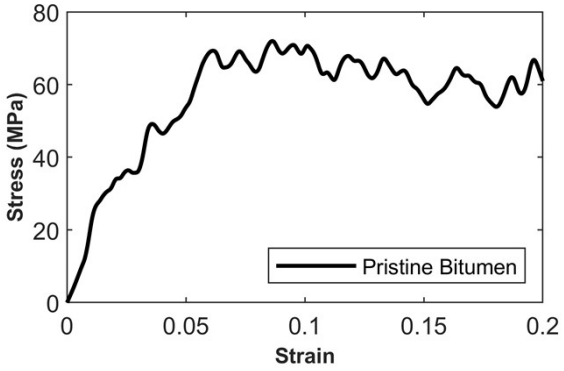

Figure S1. Stress-strain curve of pristine bitumen in the x-direction.

S3. Distance between the axes of two adjacent DNTs

When $-C_2H_5$ functionalization reaches 5%, T_g appears to be significantly reduced. To exclude potential numerical errors, additional calculations are conducted for three different models with 5% -COOH (by varying the dispersion of the DNTs), from which a uniformly decreased T_g is observed. According to the atomic configurations and distance calculation between adjacent DNTs, the reduction in T_g is most pronounced when the DNTs are under the most dispersed distribution and the asphaltene has the lowest degree of aggregation.

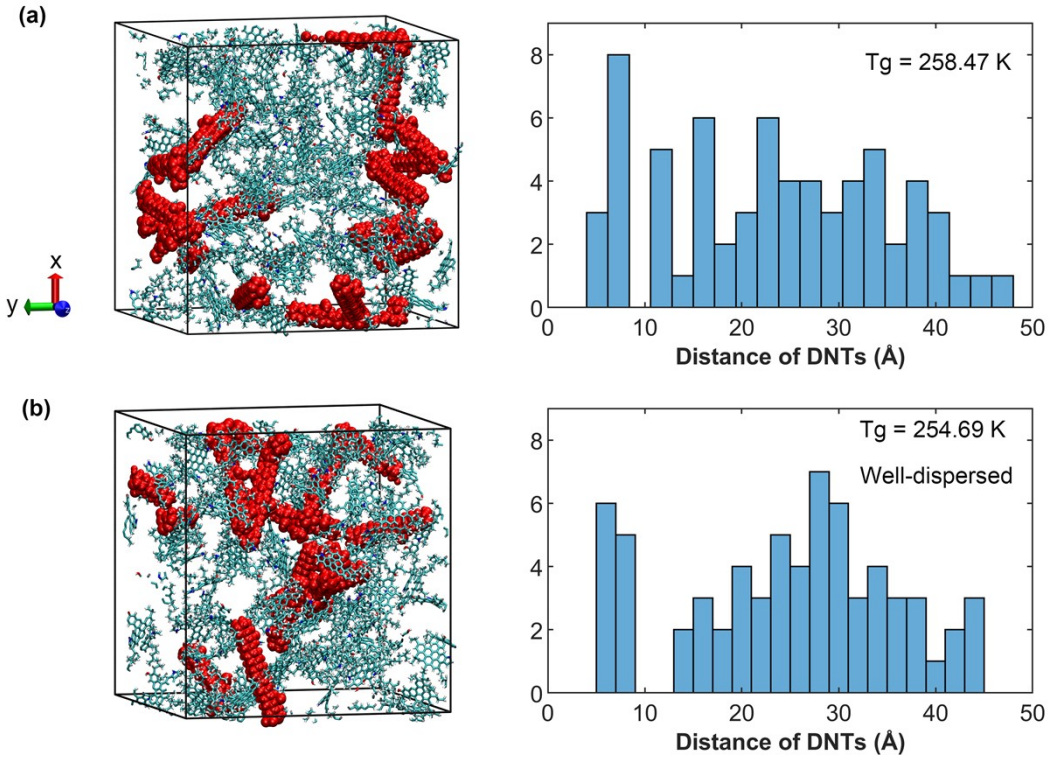

Figure S2. (a) The atomistic model (left panel) and the distribution probability of the distance between adjacent DNTs in a random distribution fashion (right panel). (b) The atomistic model (left panel) and the distribution probability of the distance between adjacent DNTs in a random distribution fashion (right panel), where DNTs effectively disaggregate the asphaltene molecules.

S4. Pearson correlation coefficient (PCC) for different molecules

By calculating the Pearson correlation coefficient, the molecules with the highest correlation coefficient can be considered as one of the key factors affecting the glass transition temperature.

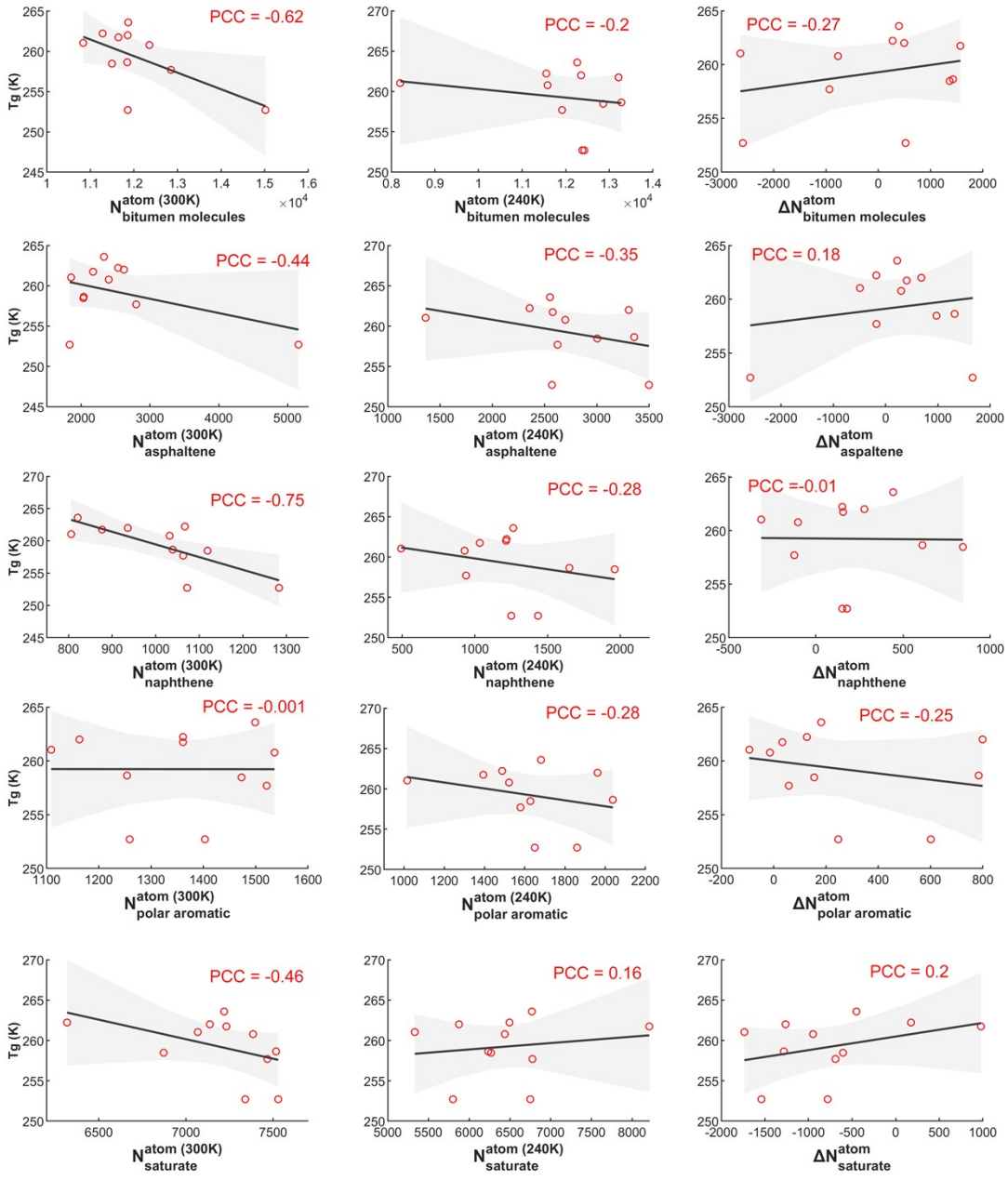

Figure S3. Pearson correlation coefficient (PCC) between the atom number and number loss around DNTs and T_g at radius of 15 Å under different temperatures. Here $\Delta N^{atom} = N^{atom\,(240K)} - N^{atom\,(300K)}$.