Supporting Information

Table S1 recipes for different samples of SBS-xFu

Sample names	The weight of SBS(g)	The weight of furfuryl mercaptan(g)	The volume of toluene(mL)
SBS-2Fu	2	0.0595	20
SBS-5Fu	2	0.1480	20
SBS-10Fu	2	0.2960	20
SBS-15Fu	2	0.4440	20
SBS-20Fu	2	0.5920	20

Table S2 data of the elemental analysis for sample SBS-15Fu

Sample name	theoretical value			experimental value		
	C%	S%	H%	C%	S%	H%
SBS-15Fu	83.15	5.09	9.21	82.63	5.02	9.08

Table S2 recipes for different samples of SBS-xFu-M series

Sample name	The weight of SBS(g)	The addition of furfuryl mercaptan(g)	The addition of bismaleimide (g)	The volume of toluene(mL)
SBS-2Fu-M	2	0.0595	0.0929	20
SBS-5Fu-M	2	0.1480	0.2323	20
SBS-10Fu-M	2	0.2960	0.4645	20
SBS-15Fu-M	2	0.4440	0.6968	20
SBS-20Fu-M	2	0.5920	0.9290	20

Table S3 recipes for different samples of SBS-15Fu-yM series

Sample name	The weight of PB(g)	The addition of furfuryl mercaptan(g)	The addition of bismaleimide (g)	The volume of toluene(mL)
SBS-15F-20M	2	0.4440	0.1394	20
SBS-15F-40M	2	0.4440	0.2787	20
SBS-15F-60M	2	0.4440	0.4184	20
SBS-15F-80M	2	0.4440	0.5574	20
SBS-15F-100M	2	0.4440	0.6968	20

Table S4 date of gel fraction and swelling ratio

Sample name	SBS-2Fu-M	SBS-5Fu-M	SBS-10Fu-M	SBS-15Fu-M (SBS-15F-100M)	SBS-20Fu-M	SBS-15F-20M	SBS-15F-40M	SBS-15F-60M	SBS-15F-80M
gel fraction	65.3	69.3	72.2	78.5	81.9	55.3	69.1	73.8	74.8

swelling ratio	21.9	18.2	15.8	13.6	13.4	22.5	17.6	15.6	14.7

Table S5 data of tensile-strain of SBS-xFu series

Sample	SBS	SBS-2Fu	SBS-5Fu	SBS-10Fu	SBS-15Fu	SBS-20Fu
tensile strength(MPa)	1.86	12.86	16.62	15.39	7.14	3.26
Ultimate strain (%)	932.35	1079.66	1095.18	991.96	1127.56	1064.91
Tensile stress at elongation of 300%(MPa)	0.79	2.14	1.99	2.37	1.64	1.21
Tensile stress at elongation of 500%(MPa)	0.95	3.17	3.71	4.28	2.07	1.53
Tensile stress at elongation of 700%(MPa)	1.42	5.53	6.95	7.67	3.53	2.40

Table S6 data of tensile-strain of SBS-xFu-M series

Sample	SBS-2Fu-M	SBS-5Fu-M	SBS-10Fu-M	SBS-15Fu-M	SBS-20Fu-M
tensile strength(MPa)	12.82	14.47	8.59	9.45	5.97
Ultimate strain (%)	777.52	706.35	679.74	751.99	344.17
Tensile stress at elongation of 300%(MPa)	4.30	6.00	6.65	6.66	6.05
Tensile stress at elongation of 500%(MPa)	6.04	8.33	7.39	7.04	------
Tensile stress at elongation of 700%(MPa)	9.91	14.14	------	8.45	------

Table S7 data of tensile-strain of SBS-15Fu-yM series

Sample	SBS-15Fu	SBS-15Fu-20M	SBS-15Fu-40M	SBS-15Fu-60M	SBS-15Fu-80M	SBS-15Fu-100M
tensile strength(MPa)	7.14	10.07	12.02	11.54	14.96	9.45
Ultimate strain (%)	1127.56	914.54	922.54	933.89	843.21	751.99
Tensile stress at elongation of 300%(MPa)	1.64	2.76	3.67	4.60	6.68	6.66
Tensile stress at elongation of 500%(MPa)	2.07	4.25	5.43	6.37	8.67	7.04
Tensile stress at elongation of 700%(MPa)	3.53	6.49	7.70	8.46	11.08	8.45

Table S8 data tensile-strain of 3 generations of the remolded sample (SBS-15Fu-80M)

The generation of the sample	0	1	2	3
tensile strength(MPa)	14.96	14.10	12.71	12.03
Ultimate strain (%)	843.21	875.19	892.66	885.71
Tensile stress at elongation of 300%(MPa)	6.68	5.98	5.36	4.73
Tensile stress at elongation of 500%(MPa)	8.67	8.15	7.49	6.82

Tensile stress at elongation of 700%(MPa)	11.08	10.48	9.68	9.08

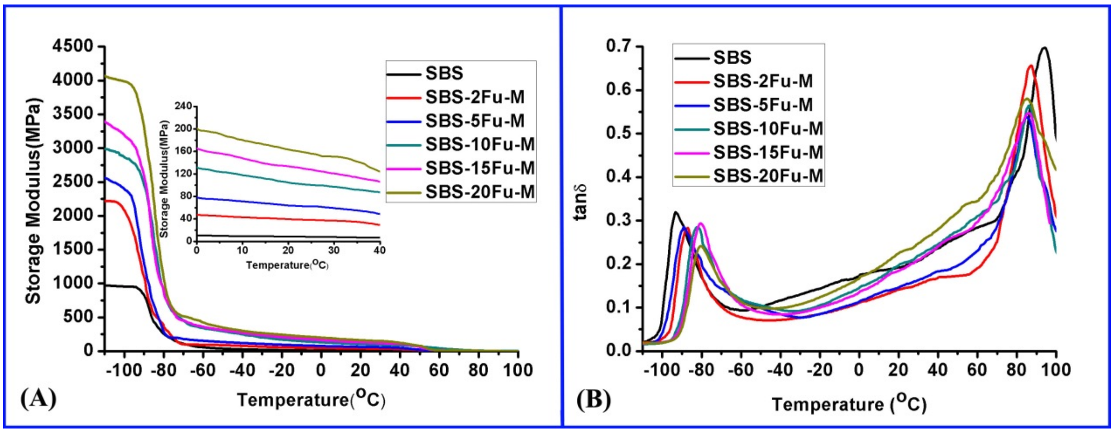

Figure S1 (A) storage modulus curves and (B) tanδ curves of the SBS-xFu-M series materials

Table S9 DMA data of the composites

Sample name	Tg1(℃)	peak tanδ1	Tg2(℃)	peak tanδ2	Storage modulus at room temperature(MPa)
SBS	-93.4	0.32	94.2	0.69	8.1
SBS-2Fu-M	-87.2	0.28	87.1	0.66	38.2
SBS-5Fu-M	-88.7	0.28	85.4	0.54	62.3
SBS-10Fu-M	-82. 2	0.28	85.9	0.56	100.3
SBS-15Fu-M	-80.4	0.29	86.1	0.55	128.0
SBS-20Fu-M	-80.4	0.24	85.4	0.58	156.1

Dynamic Thermomechanical properties for the crosslinked SBS based on DA reaction

SBS -xFu-M series was selected as typical example for investigation. Shown in Figure S1 were the DMA (Dynamic Thermomechanical analysis) spectra for pure SBS and SBS-xFu-M series at a scanning rate of 5℃/min. Figure S1A plots the relationship between the storage modulus and temperature and Figure S1B shows tan δ curve for the materials with different branch ratio of furan groups with fixed RFM at 1:1. As shown in Figure S1B, the glassy region of SBS series material was found at temperature below about -8℃, in which the chain movement was frozen and chains formed rigid network and the materials possessed high storage modulus. As the temperature increased, the storage modulus reduces gradually due to the liberation of chain's movement. As shown in Figure S1A, the storage modulus of SBS was improved substantially by the crosslinking caused by DA reaction between the furan groups on the SBS chains and bismaleimide in the range of scanned temperature. By comparison of the storage modulus at room temperature (Table S9), it was found that the storage modulus at room temperature was increased by increasing RF. For example, the storage modulus at room temperature could be improved from 8.1 MPa for the pure SBS to 156.1 MPa for the sample of SBS-20F-u-M, almost 20 times increased. The plot of tanδ was presented on Figure S1B and Table S9 summarized the data of Tg and its peak value. As shown in Figure S1B, it was illustrated that SBS and SBS-xFu-M series materials possessed two characteristic relaxation processes. One at low temperature (about -90~-80℃) belonged to the glass-transition temperature (Tg) of PB block(Tg1) in the SBS. The other at bout 90℃ was related to Tg of PS(Tg2). With the introduction of RF, the position of Tg1 moved toward higher temperature, while the position of Tg2 was shifted toward lower temperature. The formation of covalent crosslinking via D-A reaction between bismaleimide and furan modified PB block of SBS resulted in restriction of the chains movement for PB domains. Therefore, Tg1 moved towards high temperature. For the PS domains, the movement of Tg2 could be explained with that the PS was plasticized by the PB phase. Therefore, Tg2 was reduced by the introduction of the crosslinking caused by D-A reaction. The crosslinking caused by D-A reaction made the interaction between soft and hard domains strengthen. As shown in the AFM phases, for the SBS-xFu-M series material, the phases of the material changed with the branching ratio of furan groups. SBS-2Fu-M showed the lamellar structure with bright and dark phases interwinding together. Then, homogeneous structure was observed for SBS-5Fu-M. With further increasing RF up to 10 %, microphase separated structure appeared again and the PB domains had changed into dispersed phase. The AFM phases showed inner structure of the materials and the inner structure could explained that the PS phase was plasticized by the PB phase and the effect enhanced with the increase of RF. Hence, the position of Tg2 is shifted toward lower temperature and the more furan groups were branched, the more the position moved.