

Preparation and properties of a novel bio-based and non-crystalline engineering elastomer with high low-temperature and oil resistance

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ABSTRACT: A series of poly(succinic acid/sebacic acid/itaconic acid/butanediol/propanediol) bio-based and non-crystalline engineering elastomers (BEE) were obtained by changing the molar ratio of succinic acid (SA) to sebacic acid (SeA) from 5:5 (BEE-5) to 8:2 (BEE-8). We prepared bio-based engineering elastomer composites (BEE/CB) by mixing BEE with carbon black N330. The low-temperature and oil resistance properties of the BEE/CB composites were investigated in terms of low-temperature brittleness, coefficient of cold resistance under compression, oil resistance test at different temperatures, and tensile properties. The results showed that the low-temperature brittleness temperature of the BEE/CB composites ranged from -50 to -60°C and the coefficient of cold resistance under compression was 0.18 high at -60°C for BEE-7/CB and 0.23 high at -40°C for BEE-8/CB. The oil resistance properties of BEE-7/CB were higher than those of nitrile-butadiene rubber N240S (NBR N240S), and the oil resistance properties of BEE-8/CB were even as high as those of nitrile-butadiene rubber N220S (NBR N220S). © 2015 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2016**, *133*, 42855.

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INTRODUCTION

With the extensive interest in sustainable development, both academia and the chemical industry are making attempts to replace petrochemical-based monomers with natural ones.^{1,2} However, most of the synthetic elastomers are derived from petroleum-based monomers. It is strategically important to construct new elastomers or replace existing elastomers by ones based on renewable resources. Bio-based elastomers have received extensive interest in recent years.^{3,4} Most of the bio-based elastomers currently produced are used as medical materials and are thermosets. Thus, it is important to prepare bio-based elastomers, especially those targeted for engineering applications. Various bio-based elastomers have been synthesized for engineering applications. Wang *et al.*^{5,6} synthesized novel soybean-oil-based elastomers that had good machinability and tunable properties. Wang *et al.*^{7,8} synthesized a novel crosslinkable bio-based poly(diisooamyl itaconate-co-isoprene) (PDII) elastomer by emulsion polymerization. The synthesized elastomer had a high molecular weight and low T_g and could be readily processed and crosslinked by existing rubber processing facilities and methods. Wei *et al.*⁹ synthesized a novel bio-based

engineering polyester elastomer (BEE) by polymerizing commercial bio-based monomers—sebacic acid, itaconic acid, succinic acid, 1,3-propanediol, and 1,4-butanediol. The BEE had a low glass-transition temperature (T_g), high mechanical, and environmental properties.

The properties of elastomers are easily influenced by environmental factors such as temperature, oil, and light. In many fields such as the aerospace industry, automobile industry, and machinery industry, elastomers are mainly used in an oil environment for oil-resistance hoses, rollers, and seals. Elastomers are also used at low temperatures especially in the aerospace industry. The investigation of the low temperature and oil resistance properties of elastomers has very important significance. Many studies have been carried out on the low temperature and oil resistance properties of elastomers, showing that the properties of low temperature resistance and oil resistance are conflicting. The molecular chain structural features such as high molecular chain polarity can provide oil resistance in most cases, and the low glass transition temperature and crystallization, which affect flexibility of molecular chains, are the keys to low temperature resistance. However, high polarity reduces the

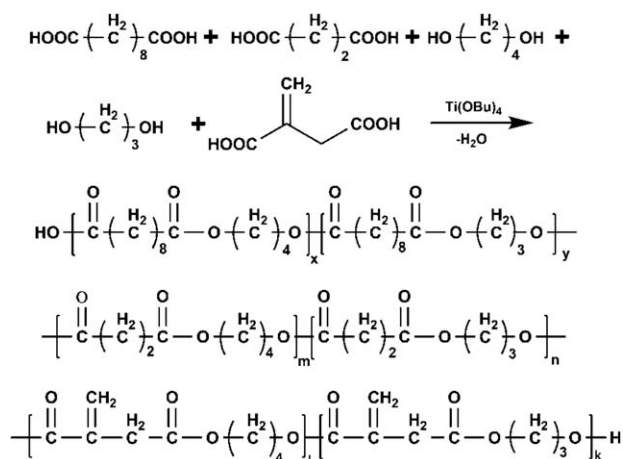


Figure 1. Synthesis of BEE.

flexibility of molecular chains. Thus, it is a challenge to optimize the low temperature and oil resistance properties of an elastomer.^{10–15}

In this study, we synthesized a novel bio-based, non-crystalline engineering elastomer with high molecular chain polarity by the polymerization of succinic acid, sebacic acid, itaconic acid, 1,4-butanediol, and 1,3-propanediol. In our previous work,⁹ we synthesized a bio-based polyester elastomer with the molar ratio of 1,3-propanediol (PDO) to 1,4-butanediol (BDO) of 1:1, crystallinity was the lowest among the elastomer we obtained. If the content of itaconic acid (IA) increased, the mechanical properties of the BEE would decrease. In this work, we changed only the molar ratio of succinic acid (SA) to sebacic acid (SeA) to adjust the flexibility of the macromolecular chains for low crystallinity and outstanding low temperature resistance of the BEE. Meanwhile, the BEE, whose repeat units are based on saturated ester groups, has potentially good oil resistance.¹⁶ We prepared carbon black BEE/CB composites by mixing the BEEs with Carbon Black N330. The low temperature resistance of the BEE/CB composite was investigated by the low-temperature brittleness test, the measurement of the coefficient of cold resistance under compression, and differential scanning calorimeter (DSC). The oil resistance of the BEE/CB composite in ASTM 1# oil and ASTM 3# oil at different temperatures was compared with those of nitrile-butadiene rubber N220S (with 41% acrylonitrile) and nitrile-butadiene rubber N240S (with 26% acrylonitrile), which were both widely used in an oil environment. The comparison was proposed based on the results of tensile testing and oil resistance test.

EXPERIMENTAL

Materials

The 1,4-butanediol (BDO) and 1,3-propanediol (PDO) were purchased from Alfa Aesar. Itaconic acid (IA), succinic acid (SA), and sebacic acid (SeA) were obtained from Guangfu Fine Chemical Institute of Tianjin. Phosphorous acid was supplied by Beijing Yili Fine Chemical. Tebrabutyl titanate (TBT) and dicumyl peroxide (DCP) were purchased from Alfa Aesar. Carbon black N330 was supplied by Qingdao Degussa. Nitrile-butadiene rubber N220S (with 41% acrylonitrile) and nitrile-

Table I. Molecular Weights of Various BEEs

Composites	$M_n (\times 10^4)$	$M_w (\times 10^5)$	$M_z (\times 10^4)$	PDI
BEE-5	3.8	1.3	4.0	3.4
BEE-6	3.8	1.2	3.9	3.3
BEE-7	4.4	1.4	4.0	3.2
BEE-8	4.0	1.6	5.2	4.1

Table II. Results of Low-temperature Brittleness of Various BEE/CB Composites

Composites	Temperature (°C)	Result
BEE-5/CB	-59	Intact
	-60	Broken
BEE-6/CB	-57	Intact
	-58	Broken
BEE-7/CB	-55	Intact
	-56	Broken
BEE-8/CB	-50	Intact
	-51	Broken

butadiene rubber N240S (with 26% acrylonitrile) were bought from Japan JSR. Standard Oil 1# and Standard Oil 3# for ASTM oil resistance performance test for rubber were supplied by Japan Sun Oil.

Synthesis

Synthesis of BEE. The BEE was synthesized according to the procedure given in our previous work.⁹ The synthesis of BEE-5 is described here as an example. The comonomers PDO

Table III. Results of Coefficient of Cold Resistance under Compression of Various BEE/CB Composites

Composites	Temperature (°C)	Coefficient
BEE-5/CB	-4	0.53
	-5	0.05
BEE-6/CB	-5	0.52
	-10	0.06
	-20	0.58
BEE-7/CB	-10	0.62
	-20	0.58
	-30	0.43
	-40	0.36
BEE-8/CB	-50	0.21
	-60	0.18
	-10	0.79
	-20	0.66
	-30	0.59
	-40	0.23
	-50	0.04

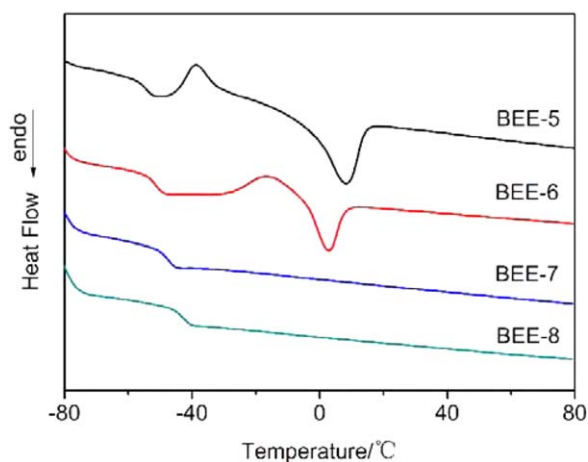


Figure 2. DSC thermograms of various BEE/CB composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

(12.56 g, 0.165 mol), BDO (14.87 g, 0.165 mol), IA (5.85 g, 0.045 mol), SA (15.06 g, 0.1275 mol), and SeA (25.79 g, 0.1275 mol) and the inhibitor phosphorous acid (0.0074 g, 0.01 wt % of total comonomers) were charged into a well-dried 100-mL, three-necked flask equipped with a condenser, a stirrer, and a gas inlet. The flask was immersed into a silicone oil bath under a nitrogen atmosphere. The temperature was slowly increased to 180°C and let stay there for about 2 h until the end of esterification. Then, heating was stopped to allow the temperature to come down to 100°C, and the catalyst $\text{Ti}(\text{O}i\text{Bu})_4$ (0.0371 g, 0.05 wt % of total comonomers) was transferred into the reactor. Then, the temperature was slowly raised to 220°C and let stay there under low pressure (< 300 Pa) for 3–4 h until the Weissenberg effect was observed. Finally, the resulting copolymer was taken out of the flask at a temperature below 100°C. The polymerization equation is shown in Figure 1.

Preparation of BEE/CB Samples. The BEE/CB composites were prepared by mixing the BEE with 40 phr of carbon black N330 and 0.16 phr of DCP in a Haake mixer (Rheomix 600p, Thermal Electron) at 50°C and a rotational speed of 40 rpm. All the

composites were finally vulcanized under 15 MPa at 160°C for 15 min to produce 2-mm-thick sheets.

Preparation of NBR/CB Samples. The NBR/CB composites were prepared by mixing BEE with 40 phr of carbon black N330 and 2 phr of DCP in a Haake mixer (Rheomix 600p, Thermal Electron) at 50°C and a rotational speed of 40 rpm. All the composites were finally vulcanized under 15 MPa at 160°C for 20 min to produce 2-mm-thick sheets.

Characterization

Gel Permeation Chromatography. The molecular weights of the BEE were determined by gel permeation chromatography (GPC) measurements on a Waters Breeze instrument equipped with three water columns (Styragel HT3_HT5_HT6E) using tetrahydrofuran as the eluent (1 mL min^{-1}) and a Waters 2410 refractive index detector. A polystyrene standard was used for calibration.

Differential Scanning Calorimetry. The differential scanning calorimetry (DSC) thermograms of the BEE were recorded in the temperature range -80°C to 150°C on a Mettler-Toledo using a heat rate of $10^\circ\text{C min}^{-1}$ under nitrogen.

Measurements of Coefficient of Cold Resistance under Compression. The coefficients of cold resistance under compression of the BEE were measured according to standard HG/T 3866-2008 (This standard is available on the authority website please check). All the samples were prepared according to Chinese standard GB/T 2941. The specimens were cylinders with a diameter of 10 mm and a height of 10 mm. The specimens were tested on a compression coefficient of cold resistance tester (RH-7040, Ming Zhu Company, China). At least three specimens of each composite were tested for an average value.

Low-temperature Brittleness Test. The low temperature brittleness tests of the BEE were conducted according to Chinese standard GB/T 1682-94. All the specimens were rectangular-shaped with a gauge length of 25 mm, a width of 6 mm, and a thickness of 2 mm. All the specimens were tested on a rubber and plastic brittle low temperature testing instrument (MZ-4068, Ming Zhu Company, China).

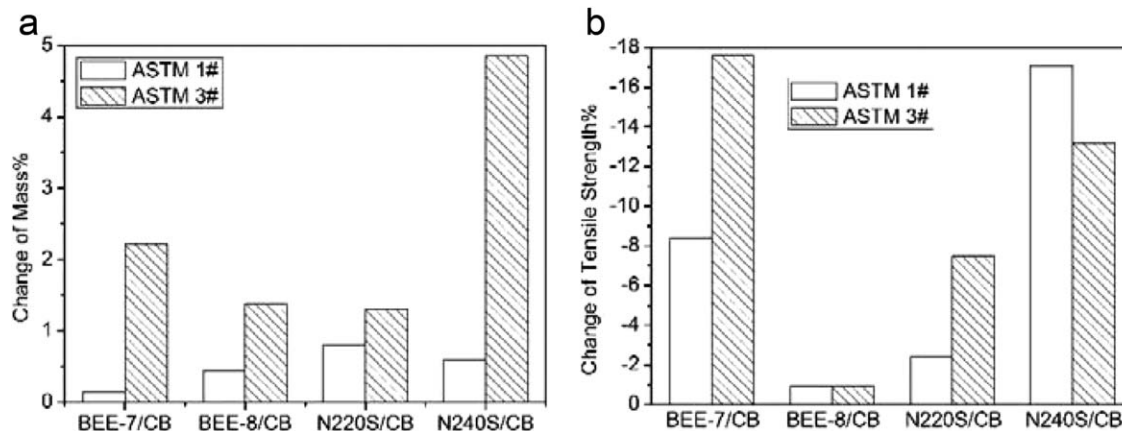


Figure 3. Change rates of (a) mass and (b) tensile strength of various BEE/CB composites and NBR/CB composites after soaking in ASTM 1# oil and ASTM 3# oil for 24 h at room temperature.

Table IV. Thermal Properties of Various BEE/CB Composites

Composites	T_g (°C)	T_m (°C)	T_c (°C)	ΔH (J g ⁻¹)
BEE-5/CB	-54.90	8.2	-38.6	-28.34
BEE-6/CB	-54.71	2.94	-16.7	-16.08
BEE-7/CB	-52.35	-	-	-
BEE-8/CB	-44.39	-	-	-

Tensile Test. The tensile tests were conducted according to ASTM D412. All the specimens were dumbbell-shaped with a gauge length of 25 mm, a width of 6 mm, and a thickness of 2 mm. The specimens were tested on a CMT4104 electronic tensile tester (SANS, China). At least five specimens of each composite were tested for an average value.

Oil Resistance Test. The effects of the oil tests of the BEE were evaluated according to Chinese standard GB/T 1690-2006.

RESULTS AND DISCUSSION

Dependence of Composition on Molecular Weight and Distribution of the Copolyester

The number average molecular weights (M_n) and polydispersity indices (PDI) of the BEEs with various ratios of SA to SeA are summarized in Table I. The M_n is about 4.0×10^4 because the Weisenberg effect occurred by the end of the reaction, the resulting copolymer wrapped up stirrer and the reaction can't

Table V. Mechanical Properties of Various BEE/CB Composites and NBR/CB Composites after Soaking in Different Oils for 24 h at Room Temperature

Composites and oil	Tensile strength (MPa)	Elongation at break (%)	Shore A hardness	Permanent set (%)
BEE-7/CB				
Before soaking	11.9	538	63	3
After soaking				
ASTM 1#	10.9	514	60	3
ASTM 3#	9.8	446	58	2
BEE-8/CB				
Before soaking	10.6	480	75	4
After soaking				
ASTM 1#	10.5	477	77	4
ASTM 3#	10.5	434	74	3
N220S/CB				
Before soaking	25.2	358	61	2
After soaking				
ASTM 1#	24.6	340	59	2
ASMT 3#	23.3	342	58	3
N240S/CB				
Before soaking	15.2	247	67	2
After soaking				
ASTM 1#	12.6	168	64	1
ASMT 3#	13.2	173	62	3

Table VI. Property Change Rates of Various BEE/CB Composites and NBR/CB Composites after Soaking in Different Oils for 24 h at Room Temperature

Composites and oil	Change of tensile strength (%)	Change of elongation at break (%)	Change of shore A hardness (%)	Change of mass (%)
ASTM 1#				
BEE-7/CB	-8.4	-4.5	-3	0.15
BEE-8/CB	-0.9	-0.6	2	0.44
N220S/CB	-2.4	-5.0	-2	0.80
N240S/CB	-17.1	-32.0	-3	0.57
ASTM 3#				
BEE-7/CB	-17.6	-17.1	-5	2.22
BEE-8/CB	-0.9	-9.6	-1	1.38
N220S/CB	-7.5	-4.5	-3	1.3
N240S/CB	-13.2	-30.0	-5	4.86

Table VII. Mechanical Properties of Various BEE/CB Composites and NBR/CB Composites after Soaking in ASTM 1# Oil at Different Temperatures for 24 h

Temperature (°C)	Tensile strength (MPa)	Elongation at break (%)	Shore A hardness	Change of mass (%)
BEE-7/CB				
Before soaking	11.9	538	63	-
25	10.9	514	60	0.15
80	10.2	342	52	0.30
100	9.1	378	45	0.20
125	7.9	296	43	1.10
150	4.9	284	40	1.90
BEE-8/CB				
Before soaking	10.6	480	75	-
25	10.5	477	77	0.44
80	10.2	453	70	0.08
100	8.7	429	66	0.17
125	7.2	387	58	0.41
150	5.8	260	54	0.70
N220S/CB				
Before soaking	25.2	358	61	-
25	24.6	340	59	0.80
100	23.6	258	60	0.60
150	13.9	252	57	4.60
N240S/CB				
Before soaking	15.2	247	67	-
25	12.6	168	64	1.38
100	10.8	146	62	8.11
150	4.3	106	66	8.22

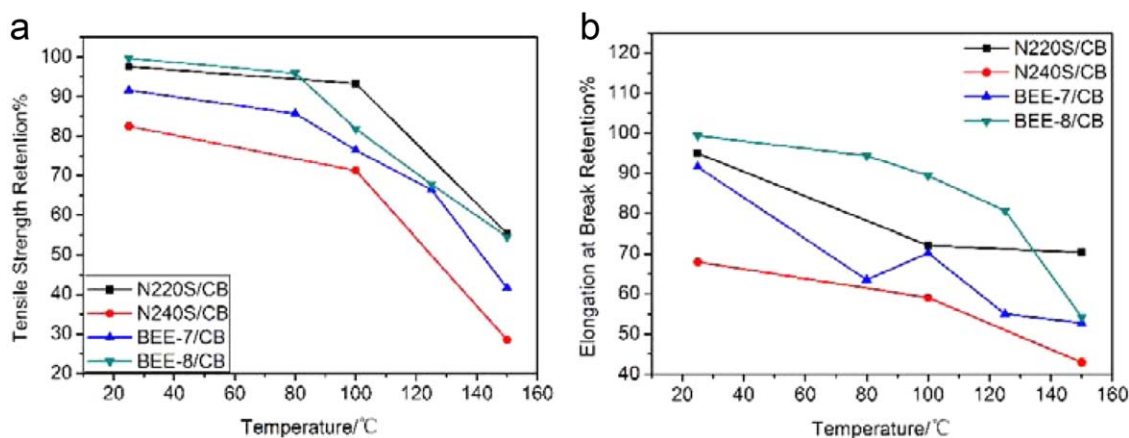


Figure 4. Retentions of (a) tensile strength and (b) elongation at break of BEE/CB composites and NBR/CB composites after being soaked in ASTM 1# oil at different temperatures for 24 h. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

continue. The PDI change little as the ratio of SA to SeA changes. But the PDI is a little high because the reaction was polycondensation, which is random copolymerization. The viscosity of copolymer will increase as the reaction goes on. The movement ability of molecular chain will also decrease.

Low-temperature Resistance Properties of BEE/CB Composites

The results of low-temperature brittleness and coefficient of cold resistance under compression are shown in Tables II and III, respectively.

It can be seen that the brittleness temperatures of the various BEE/CB composites range from -50 to -60°C , an indication that the BEE/CB composites still have high strength and impact resistance at low temperatures. The coefficient of cold resistance under compression of BEE-5/CB is 0.05 at -5°C and that of BEE-6/CB is 0.07 at -10°C , indicating that the BEE-5/CB and BEE-6/CB composites lose elasticity at relatively high temperatures and the low-temperature resistance properties are poor. However, the coefficient of cold resistance under compression of BEE-7/CB is still 0.18 at -60°C and that of BEE-8/CB is still 0.23 at -40°C , indicating that the BEE-7/CB and BEE-8/CB composites have excellent low-temperature resistance.

The results show that the coefficient of cold resistance under compression depends on the ratio of SA to SeA. An explanation for this dependence was obtained by a study of thermal properties by differential scanning calorimetry (DSC).

The DSC heating thermograms and thermal properties for various BEE/CB composites are shown in Figure 2 and Table IV, respectively. It can be seen that glass transition temperatures (T_g) range from -55 to -45°C . The crystallization melting peak disappears at molar ratios of SA to SeA of 7 or higher the hybridization and disorder of molecular chains increase as the SA content increases. Therefore, the low-temperature resistance properties of BEE/CB increase as the molar ratio of SA to SeA increases. The low-temperature resistance properties of BEE-7/CB and BEE-8/CB composites are excellent.

Oil Resistance

Oil Resistance Properties of BEE/CB Composites. The oil resistance properties of BEE-7/CB and BEE-8/CB are compared with those of nitrile-butadiene rubber N220S (NBR N220S) and nitrile-butadiene rubber N240S (NBR N240S), which are widely used in an oil environment.

Table VIII. Mechanical Properties of VARIOUS BEE/CB Composites and NBR/CB Composites after Being Soaked in ASTM 3# Oil at Different Temperatures for 24 h

Temperature ($^{\circ}\text{C}$)	Tensile strength (MPa)	Elongation at break (%)	Shore A hardness	Change of mass (%)
BEE-7/CB				
Before soaking	11.9	538	63	-
25	9.8	446	58	2.22
80	7.7	436	40	5.10
100	7.0	375	46	7.83
125	4.2	309	35	8.80
150	3.1	349	28	10.76
BEE-8/CB				
Before soaking	10.6	480	75	-
25	10.5	434	74	1.38
80	7.9	414	65	7.24
100	7.7	354	54	8.11
125	4.8	255	50	7.25
150	4.7	235	49	8.22
N220S/CB				
Before soaking	25.2	358	61	-
25	23.3	342	58	1.29
100	17.3	260	57	9.70
150	10.4	176	54	13.83
N240S/CB				
Before soaking	15.2	247	67	-
25	13.2	173	62	4.86
100	8.2	123	55	35.69
150	6.4	99	54	40.37

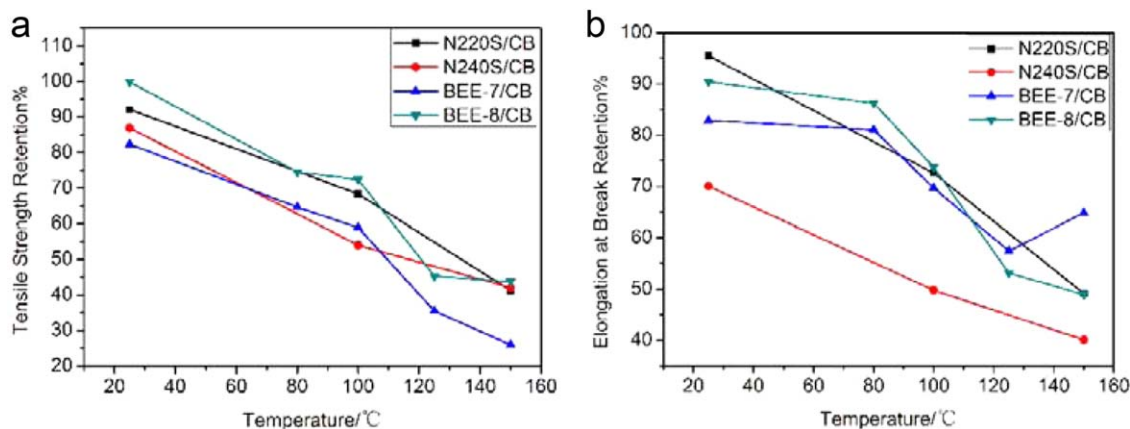


Figure 5. Retentions of (a) tensile strength and (b) elongation at break of BEE/CB composites and NBR/CB composites after being soaked in ASTM 3# oil at different temperatures for 24 h. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

It can be seen from Table V that after the composites are soaked in ASTM 1# oil and ASTM 3# oil for 24 h at normal temperature, the tensile strength, elongation at break, and hardness decrease. Table VI shows the rates of change of various properties of the BEE/CB composites and NBR/CB composites after being soaked in ASTM 1# oil and ASTM 3# oil for 24 h at room temperature. The oil resistance properties of BEE-8/CB are better than those of BEE-7/CB because of the increase in density of ester groups as the SA content increases. The oil resistance properties of N220S/CB are better than those of N240S/CB because the content of acrylonitrile is higher in N220S than in N240S.

It also can be seen that the rates of change of tensile strength, elongation at break, and mass of BEE-7/CB are lower than those of N240S/CB, but higher than those of N220S/CB. The rates of change of tensile strength, elongation at break, and mass of BEE-8/CB are higher than those of N220S/CB and N240S/CB in ASTM 1#. The rate of change of tensile strength of BEE-8/CB is lower than those of N220S/CB and N240S/CB, but the rates of the elongation at break and mass of BEE-8/CB are little higher than those of N220S/CB and lower than N240S/CB in ASTM 3#.

High Temperature Oil Resistance Properties of BEE/CB Composites. Table VII shows that the elongation at break and hardness of the BEE/CB and NBR/CB composites after being soaked in ASTM #1 oil decrease as the temperature increases. The mechanical properties decrease sharply as the temperature exceeds 100 °C because as the temperature increases, the mobility of the molecules increases, more oil molecules will permeate into the rubber, and the aging of the molecular chains leads to their destruction.¹⁷ The tensile strength of BEE-7/CB and BEE-8/CB composite are still 4.9 and 5.8 MPa high and the elongation at break remains at 284 and 260% high at 150 °C.

As shown in Figure 4, the retentions of tensile strength and elongation at break of the BEE-7/CB and BEE-8/CB composites are higher than those of NBR N240S/CB, but lower than those of NBR N220S/CB. BEE-8/CB has a lower tensile strength retention, but a higher elongation at break retention than NBR N220S/CB. The oil resistance of BEE-8/CB in ASTM 1# oil is about the same as that of NBR N220S/CB.

Table VIII shows that the tensile strength, elongation at break, and hardness of the BEE/CB and NBR/CB composites also decrease as the temperature increases after the composites are soaked in ASTM 3# oil for 24 h. As the temperature exceeds 100 °C, the mechanical properties decrease sharply. The tensile strengths of the BEE-7/CB composite and BEE-8/CB composite are still 3.1 and 4.7 MPa high and the elongations at break remain at 349 and 235% high at 150 °C, indicating that the BEE-7/CB and BEE-8/CB composites can maintain high elasticity at high temperatures in ASTM 3# oil.

It can be seen in Figure 5 that the tensile strength retention of the BEE-7/CB composite is higher than that of NBR N240S/CB, but lower than that of NBR N220S/CB at temperatures. The elongation at break retention of BEE-7/CB is higher than that of NBR N240S/CB, but about the same as that of NBR N220S/CB. The tensile strength retention and elongation at break retention of BEE-8/CB composite are about the same as those of NBR N220S/CB, but are higher than those of NBR N240S/CB.

CONCLUSIONS

A series of noncrystallized bio-based engineering polyester elastomers (BEEs) with high low-temperature and oil resistance properties were obtained successfully by changing the molar ratio of SA to SeA from 5:5 to 8:2. The low-temperature resistance properties of the BEE-7/CB composite were excellent. This composite even had high strength and impact resistance at -55 °C, and the coefficient of cold resistance was still 0.18 at -60 °C and the elastomer still had elasticity. The BEE-8/CB composite also had high strength and impact resistance at -50 °C, and the coefficient of cold resistance was 0.23 at -40 °C. The oil resistance properties of the composites were found to increase as the molar ratio of SA to SeA increased.

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