

# Synthesis and Properties of Ethylene-Substituted Styrene Copolymers

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Received 27 December 2007; accepted 20 May 2008

DOI 10.1002/app.28854

Published online 18 September 2008 in Wiley InterScience (www.interscience.wiley.com).

**ABSTRACT:** The copolymerization of ethylene and substituted styrenes [RSt's; *p*-methylstyrene (MSt), *p*-*tert*-butylstyrene (BSt), 2-vinylnaphthalene (VN), and *p*-(*tert*-butyldimethylsilyloxy)styrene (BMSiOSt)] were investigated with dimethylsilylene(tetramethylcyclopentadienyl)(*N*-*tert*-butyl) titanium dichloride to yield the corresponding ethylene-RSt copolymers. The substituent on the styrene (St) monomers did not affect the monomer reactivity ratio. The effect of the substituent structure of RSt on the thermal and mechanical properties was studied with differential scanning calorimetry, dynamic mechanical thermal spectroscopy, and elongation testing. The glass-transition temperature ( $T_g$ ) of the copolymers increased with increasing RSt content, and the order of  $T_g$  was as follows: BSt > VN > MSt = St. A copolymer with

*p*-hydroxystyrene (HOSt) was successively synthesized by means of deprotection of the copolymer with BMSiOSt. The copolymer showed a much higher  $T_g$  than the other copolymers because of the hydrogen connection of its OH groups. The mechanical properties of the copolymer in the glass state, at a lower temperature than  $T_g$ , were almost independent of the nature of the RSt. The substituent of the St monomers affected the pattern of the stress-strain curve in the elongation testing in the amorphous state. An improvement in the shape memory effect was observed in poly(ethylene-*co*-BSt). © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 110: 3770–3777, 2008

**Key words:** copolymerization; mechanical properties; polyolefins; thermal properties; thermodynamics

## INTRODUCTION

The development of transition-metal catalysts for olefin polymerization has enabled us to synthesize new types of polyolefins with narrow molecular weights and composition distributions.<sup>1</sup> The catalysts show high activity and the efficient incorporation of bulky monomers in copolymerizations with common olefins. The copolymer of ethylene with higher  $\alpha$ -olefin monomers is one of the most useful polyolefins; it is synthesized with various metallocene catalysts. Half-sandwiched titanocene catalysts, including bridged cyclopentadienyl and amido ligands, the so-called constrained-geometry catalysts (CGCs), are some of the most effective catalysts for ethylene/ $\alpha$ -olefin copolymerization. CGCs also successfully achieve the copolymerization of ethylene and styrene (St; Scheme 1).<sup>2–6</sup> The copolymer with St shows the characteristics of semicrystalline rubbery, amorphous rubbery, and amorphous glasstomer features with increasing St content.<sup>7</sup> Copolymers with

some aromatic vinyl monomers have been synthesized with CGCs.<sup>8–12</sup> One of the authors investigated the copolymerization of ethylene and 2-vinylnaphthalene (VN) with a CGC and yielded the corresponding copolymer with a high VN content.<sup>13</sup> The copolymer with VN showed a higher glass-transition temperature ( $T_g$ ) than that with St. The structural modification of substituted styrene (RSt) monomers in ethylene-based copolymers should be an effective method for the development of properties of the copolymers. In this study, we examined the copolymerization of ethylene and RSt [*p*-methylstyrene (MSt), *p*-*tert*-butylstyrene (BSt), VN, and *p*-(*tert*-butyldimethylsilyloxy)styrene (BMSiOSt)], as shown in Schemes 1 and 2, with a CGC. We also studied the effect of the substituents on the thermal and mechanical properties of the resulting copolymers.

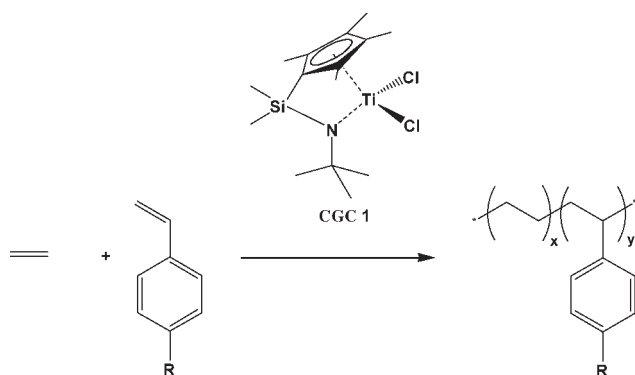
## EXPERIMENTAL

### Materials

A CGC, dimethylsilylene(tetramethylcyclopentadienyl)(*N*-*tert*-butyl)titanium dichloride (**1**), was commercially obtained from Boulder Scientific Co., Ltd. (Mead, CO), and was used as received. Methylaluminoxane (MAO) and methyl isobutylaluminoxane

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**Scheme 1** Copolymerization of ethylene and RSt with CGC.

(MMAO) were commercially obtained from Albemarle Co., Ltd. (Baton Rouge, LA), and Tosoh Finechem Co. (Tamaguchi, Japan), respectively, as toluene solutions and were used as solids after the evaporation of toluene. St, MSt, and BSt were commercially obtained and purified with distillation under reduced pressure after they were dried with  $\text{CaH}_2$ . BMSiOSt was commercially obtained and purified by  $\text{CaH}_2$  after filtration and dried under a nitrogen atmosphere. Ethylene was commercially obtained from Sumitomo Seika Chemicals Co., Ltd. (Osaka, Japan), and was used without further purification. Toluene was commercially obtained and dried with  $\text{CaH}_2$ .

### Copolymerization

The copolymerization was conducted in a 100-mL glass reactor equipped with a magnetic stirrer. Measured amounts of MAO or MMAO and toluene were added to the reactor under a nitrogen atmosphere. Ethylene was introduced into the reactor at 40°C under 1 atm of pressure until the solvent was saturated with ethylene; then, a measured amount of RSt was added. The copolymerization was started by the introduction of a toluene solution of catalyst **1** into the reactor. The copolymerization was terminated by the addition of a small amount of methanol. The resulting copolymer was precipitated in a large excess of methanol and recovered by filtration. The copolymer obtained was extracted with boiling *o*-dichlorobenzene to remove the catalyst residues. The solution of extracted polymer was concentrated by an evaporator and precipitated in a large excess of methanol. The resulting copolymer was filtered and dried *in vacuo* at 60°C for 6 h.

### Analytical procedures

The copolymers for NMR spectroscopy were dissolved in 1,1,2,2-tetrachloroethane- $d_2$  or  $\text{CDCl}_3$ .  $^1\text{H}$ -

NMR spectra were recorded at 110 or 60°C with a Jeol JMN-LA300 NMR spectrometer (Tokyo, Japan) in the pulse Fourier transfer mode. The molecular weights and molecular weight distributions of the copolymers were measured at 140°C by means of gel permeation chromatography (GPC; PL-GPC-220) (Polymer Laboratories, Amherst, MA) with trichlorobenzene as the solvent and calibrated with standard polystyrene (poly-St) samples. The thermal properties of the copolymers were investigated with a Rigaku DSC 8230 instrument (Tokyo, Japan) at a heating rate of 10°C/min after previous heating to 200°C and cooling to -50°C at 10°C/min. Samples for elongation testing and dynamic mechanical thermal spectroscopy were melted at 140°C and pressed under 10 MPa of pressure in a mold 0.1 mm thick; they were then rapidly cooled in water at room temperature. Specimens were stamped out with a dumbbell cutter (Japan Industry Standards no. 7). The mechanical properties of the test films ( $20 \times 6 \times 0.1 \text{ mm}^3$ ) were acquired with a Tensilon RTE-1210 (Orientech, Tokyo, Japan) at an elongation rate of 1 mm/min. Dynamic mechanical thermal spectroscopy was investigated with a VIB-DDV-II-EP (TOYO Baldwin, Tokyo, Japan) at 110 Hz at a heating rate of 2°C/min from -50 to 100°C.

## RESULTS AND DISCUSSION

### Copolymerization

The copolymerization of ethylene and RSt (MSt, BSt, and BMSiOSt) was investigated with CGC **1** in toluene at 40°C. Copolymerization with St was also conducted as a reference. The results are summarized in Table I. The corresponding copolymers were obtained in good yield. The copolymerization diagrams and the relationship between the comonomer content in the feed and in the copolymer are shown in Figure 1. The copolymerization results with VN, which were previously reported,<sup>13</sup> are also plotted in the figure. All of the copolymerizations traced almost the same curve. The results indicate that the substituent of the St monomer did not affect the monomer reactivity ratio in the copolymerizations. A clear relationship between the comonomer content and the polymer yield or molecular weight of the copolymers was not detected. The results indicate that the insertion of the copolymers did not hinder the insertion of the monomers and did not induce frequent chain-transfer reactions.

### Thermal properties

The thermal properties of the ethylene (E)-RSt copolymers were investigated with differential scanning calorimetry (DSC; DSC curves of the

TABLE I  
Copolymerization of Ethylene and RSt with Catalyst 1<sup>a</sup>

Run	RSt	RSt in the feed (mol/L)	Time (min)	Polymer yield (kg mol of Ti <sup>-1</sup> h <sup>-1</sup> )	RSt in the copolymer (mol %) <sup>b</sup>	$M_n$ (10 <sup>-4</sup> ) <sup>c</sup>	$M_w/M_n$ <sup>c</sup>
1	St	0.5	45	275	15.1	5.5	1.7
2	St	1.0	60	319	29.6	6.4	1.5
3	St	2.0	90	288	44.1	3.1	2.7
4	MSt	0.5	90	134	16.6	5.0	1.4
5	MSt	0.8	120	120	22.5	4.5	1.3
6	MSt	1.0	120	111	29.6	5.0	1.3
7	BSt	0.5	90	181	15.6	5.1	1.6
8	BSt	1.0	105	200	24.2	2.2	1.6
9	BSt	2.0	90	317	43.1	5.6	1.9
10	BMSiOSt	0.5	120	54	17.5	1.3	4.8

<sup>a</sup> Polymerization conditions: CGC 1 [Ti] = 0.4 mmol/L; MMAO [Al]/[Ti] = 1000 in the copolymerization with St; MAO [Al]/[Ti] = 3000 in the copolymerization with MSt, BSt, and BMSiOSt; ethylene = 1.0 atm; polymerization temperature = 40°C.

<sup>b</sup> Comonomer content determined by <sup>1</sup>H-NMR spectroscopy.

<sup>c</sup> Number-average molecular weight ( $M_n$ ) and molecular weight distribution ( $M_w/M_n$ ) determined by GPC with poly-St standard samples.

copolymers are available in the Supporting Information in the online version of this article), and the results are summarized in Table II. The relationship between the comonomer content and  $T_g$  is illustrated in Figure 2. The data of the E-VN copolymer, which was synthesized in a previous study, are also plotted in the figure. The  $T_g$  values of all of the copolymers increased with increasing RSt content. The  $T_g$  values of the copolymers decreased in the following order: E-BSt > E-VN > E-MSt = E-St. The  $T_g$  values of the E-RSt copolymers were estimated by some equa-

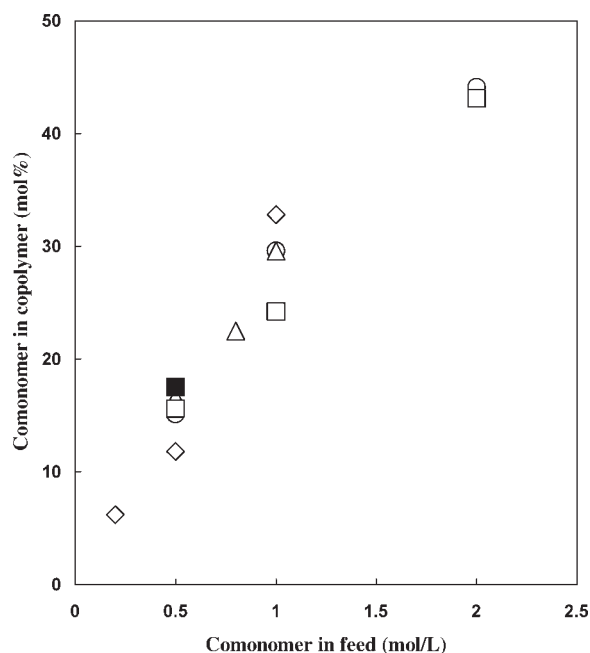


Figure 1 Copolymerization diagram of ethylene and RSt: (○) St, (△) MSt, (□) BSt, (■) BMSiOSt, and (◇) VN.

tions,<sup>14</sup> for example, the Fox equation.<sup>15</sup> [ $T_g$  of the E-RSt copolymer was estimated with the Fox equation:  $T_g^{-1} = w_1 T_{g1}^{-1} + w_2 T_{g2}^{-1}$ , where  $w_1$ ,  $w_2$ ,  $T_{g1}$ , and  $T_{g2}$  are the weight percentage of ethylene units, the weight percentage of RSt units, the glass-transition temperature of polyethylene, and the glass-transition temperature of the homopolymer of RSt, respectively.] The  $T_g$  of the copolymer corresponded to the content and the  $T_g$  of the homopolymer of RSt and increased with increasing RSt content and  $T_g$  of the RSt homopolymer. The  $T_g$ 's of the RSt homopolymer were as follows: poly(2-vinylnaphthalene) (poly-VN)  $T_g = 151^\circ\text{C}$  (424 K),<sup>16</sup> poly(*p*-tert-butylstyrene) (poly-BSt)  $T_g = 126^\circ\text{C}$  (399 K),<sup>17</sup> poly(*p*-methylstyrene)  $T_g = 106^\circ\text{C}$  (379 K),<sup>18-23</sup> and poly-St  $T_g = 100^\circ\text{C}$  (373 K). According to the Fox equation, the copolymer with VN should show the highest  $T_g$  among these copolymers in theory. However, the copolymer with BSt almost showed a higher  $T_g$  than the copolymer with VN. One explanation for the unexpected results may be the differences in the interactions between the side chains of the (co)polymers, as shown in Scheme 2. In the case of poly-VN and the E-VN copolymer,  $\pi$ -stacking between the side groups of naphthalene should have been predominant. On the other hand, both the  $\pi$ -stacking of phenyl groups and the entanglement of butyl groups would have occurred in the poly-BSt and E-BSt copolymer. In the homopolymers, the  $\pi$ -stacking of the linked phenyl region in the side groups would be effective, as shown in Scheme 3(1). Poly-VN, which had two phenyl groups per monomer unit, showed a higher  $T_g$  than poly-BSt, which had one phenyl group per monomer unit. On the other hand, entanglement between the substituents of the side groups (or main chain and side groups) should have been

TABLE II  
Structure and Properties of the E-RSt Copolymers

Sample	RSt	mol %	wt %	$M_n$ ( $10^{-4}$ ) <sup>a</sup>	$M_w/M_n$ <sup>a</sup>	$T_m$ (°C) <sup>b</sup>	$T_{g1}$ (°C) <sup>c</sup>	$T_{g2}$ (°C) <sup>d</sup>	$\Delta T_g$ (K) <sup>e</sup>	Young's modulus (MPa) <sup>f</sup>	Rupture stress (MPa) <sup>f</sup>	Rupture strain (%) <sup>f</sup>	Yield stress (MPa) <sup>f</sup>	Yield strain (%) <sup>f</sup>
1	St	15.1	39.8	5.5	1.7	58.0	-15.5			2.4	2.2	741		
2	St	29.6	61.0	6.4	1.5		1.2	20.7	19.5	1.0			0.2	63.3
3	St	44.1	74.5	3.1	2.7		20.7			1.5	1.9	487	0.5	16.6
4	MSt	16.6	45.6	5.0	1.4		-10.4	14.7	25.1	2.3	0.14	1079		
5	MSt	22.5	55.0	4.5	1.3		1.3	26.2	24.9	1.8			0.5	78.8
6	MSt	29.6	63.9	5.0	1.3		9.3			1.8			0.7	97.8
7	BSt	15.6	51.4	5.1	1.6		13.3	42.7	29.4	3.7	15.6	849		
8	BSt	24.4	64.8	2.2	1.6		35.1			66.3	2.7	341		
9	BSt	43.1	81.2	5.6	1.9		61.0			1965	47.8	3.0		
10	BMSiOSt	17.5	64.0	1.3	4.8		-6.5							
11	HOSiSt <sup>g</sup>	17.5	47.6				37.5							
12	VN	6.5	27.7	3.1	1.6	97.7		30.7		36.0	16.1	443		
13	VN	13.9	47.0	3.7	1.4	73.9	5.1	38.7	33.6	5.8	11.5	550		
14	VN	32.8	72.9	0.3	3.9		32.2							

<sup>a</sup> Number-average molecular weight ( $M_n$ ) and molecular weight distribution ( $M_w/M_n$ ) determined by GPC with poly-St standard samples.

<sup>b</sup> Melting temperature determined by DSC.

<sup>c</sup>  $T_g$  determined by DSC.

<sup>d</sup>  $T_g$  determined by dynamic mechanical thermal spectroscopy.

<sup>e</sup>  $\Delta T_g = T_{g2} - T_{g1}$ .

<sup>f</sup> Determined with elongation testing.

<sup>g</sup> Obtained with the deprotection of sample 10.

more effective than the  $\pi$ -stacking of phenyl groups in the ethylene-based copolymers with isolated RSt comonomers, as shown in Scheme 3(2). The copolymer with BSt, which had a branched substituent, should have shown a higher  $T_g$  than the copolymer

with VN. Another explanation is the difference in the free volume of these copolymers. The E-VN copolymer would have had a larger free volume than that of the E-BSt copolymer because of its large side of naphthalene. The effective increase in the free

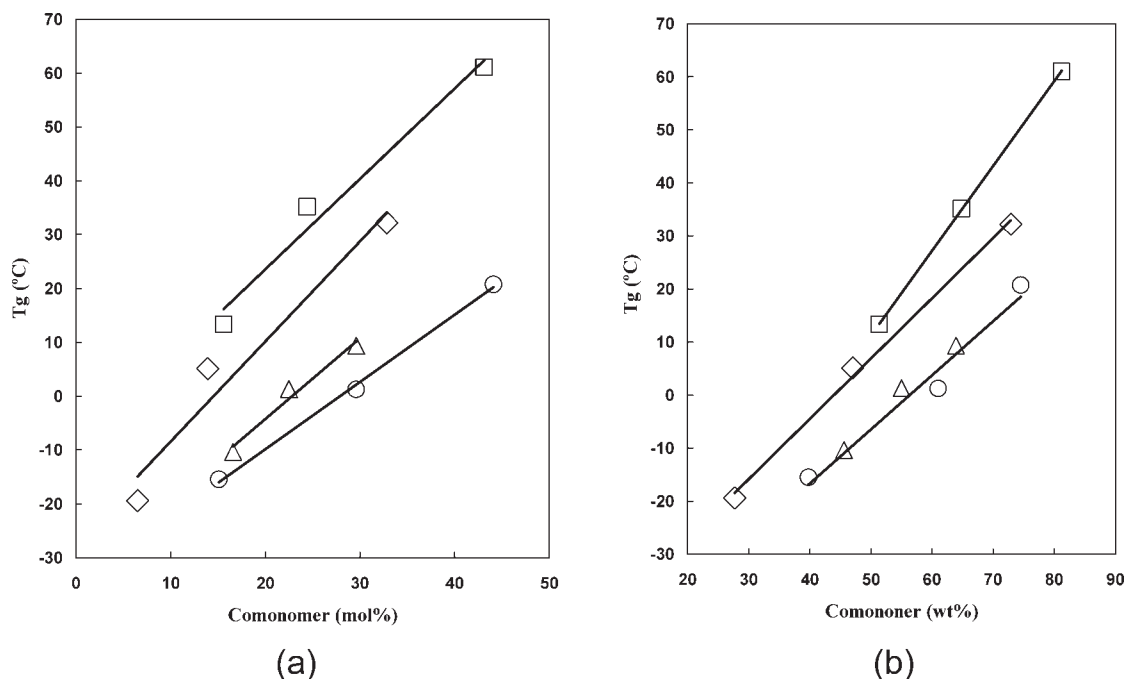
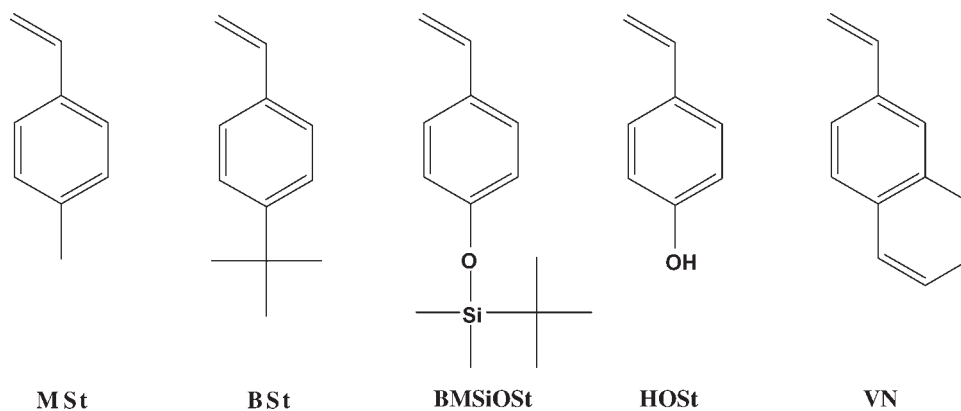


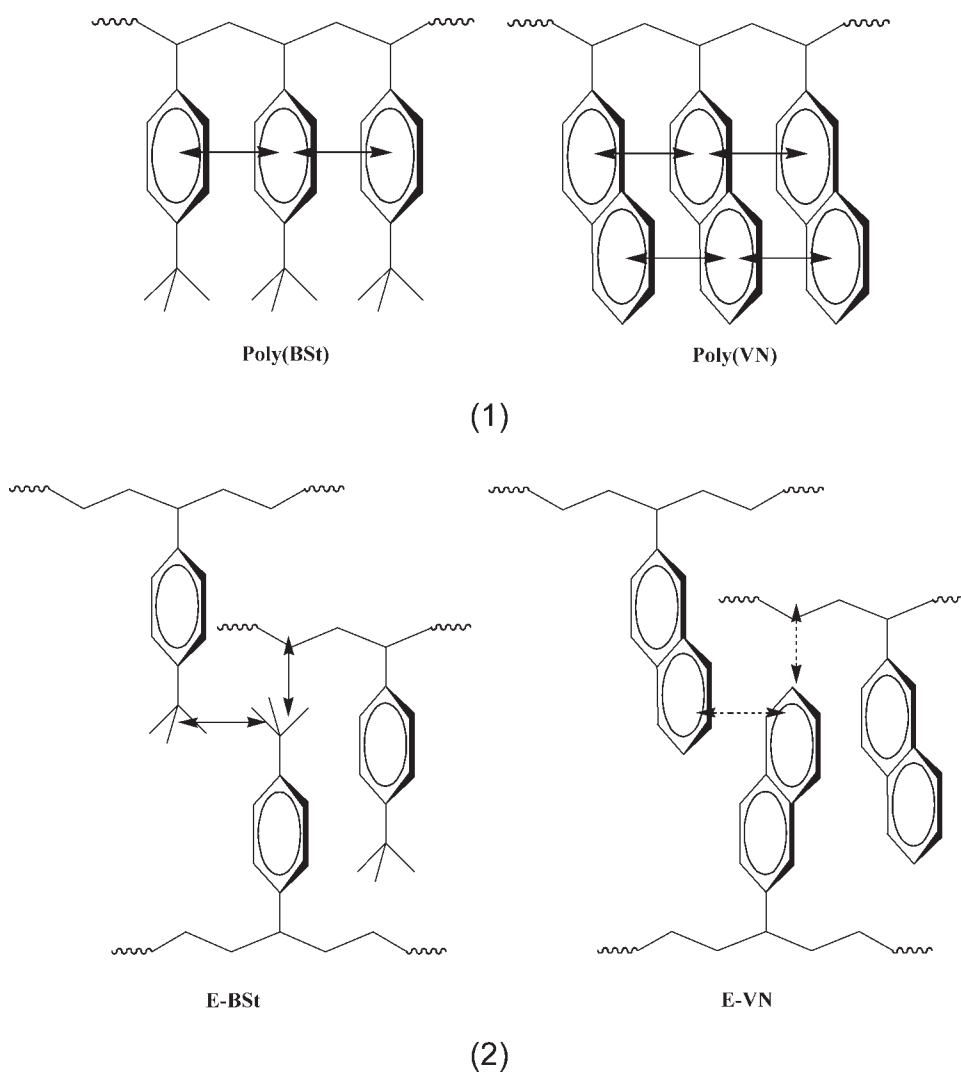
Figure 2 Relationship between the comonomer content [(a) mol % and (b) wt %] and  $T_g$  of the copolymer with (○) St, (△) MSt, (□) BSt, and (◇) VN.



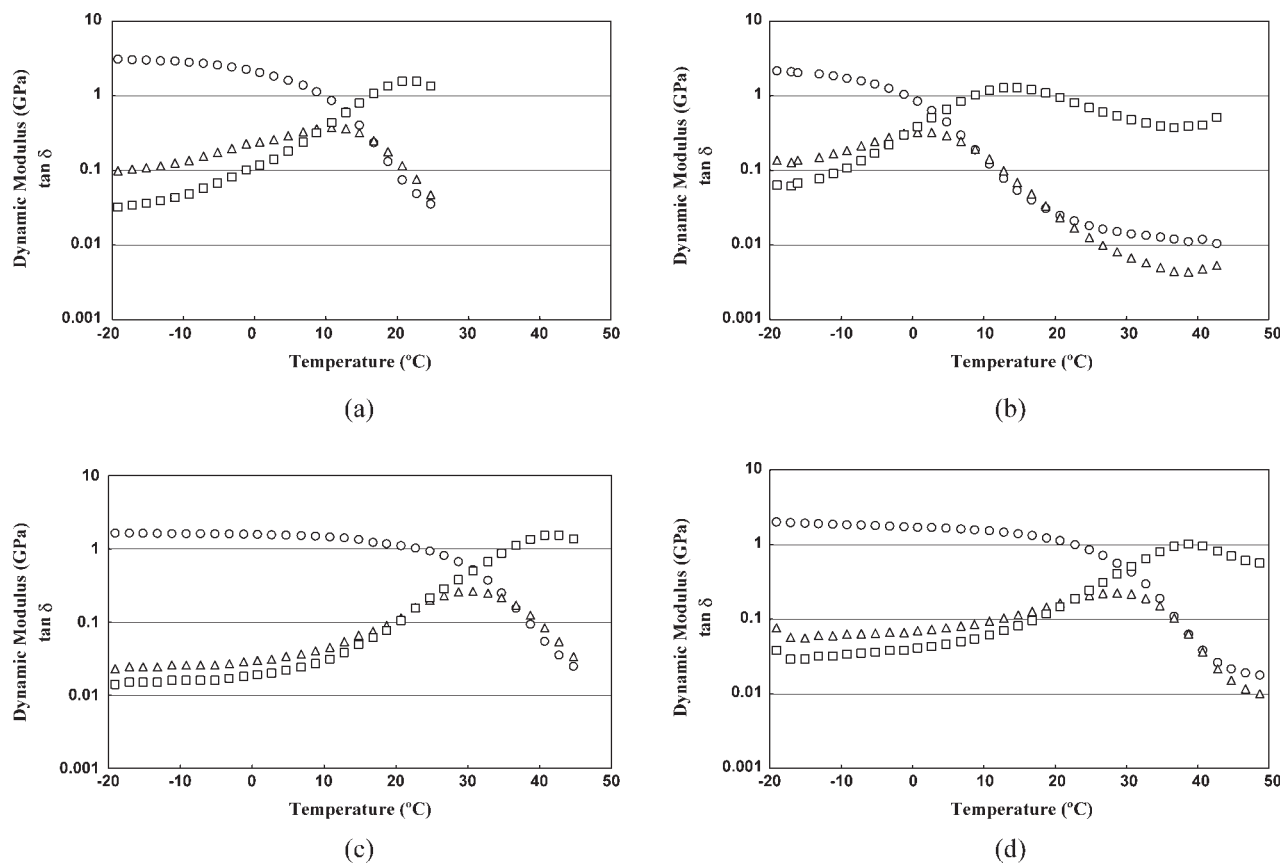
**Scheme 2** Structure of the RSt monomers used in this study.

volume in the E-VN copolymer would have caused a lower  $T_g$  than that in the E-BSt copolymer, as reported in ethylene/ $\alpha$ -olefin copolymers with various numbers of *n*-alkyl branches.<sup>24</sup>

The E-*p*-hydroxystyrene (HOSt) copolymer was successively synthesized by means of the deprotection of the protected group in poly[ethylene-*co-p*-(*tert*-butyldimethylsilyloxy)styrene] with HCl.<sup>25</sup> The



**Scheme 3** Interaction model of RSt in the (1) homopolymer and (2) copolymer with ethylene.



**Figure 3** Dynamic mechanical thermal spectroscopy of (a) sample 2, E-St; (b) sample 4, E-MSt; (c) sample 7, E-BSt; and (d) sample 13, E-VN: (○) storage modulus (GPa), (△)  $E''$  (GPa), and (□)  $\tan \delta$ .

copolymer showed a much higher  $T_g$  (37.5°C) than the other copolymer (−6.5°C) because of the hydrogen connection between its OH groups. The same observation was reported in poly(ethylene-*co*-cycloolefin)s.<sup>26</sup>

### Dynamic mechanical thermal spectroscopy

Dynamic mechanical thermal spectroscopy of the copolymers was used to study the properties in the glass state and the transition behavior from the glass state to the amorphous state. Typical spectra of the copolymers are shown in Figure 3. The storage modulus of the copolymers in the glass state, at a lower temperature than  $T_g$ , were almost independent of the nature of the copolymers and ranged from 1.5 to 3 GPa. Although the substituent structure of RSt somewhat affected the loss modulus ( $E''$ ) of the copolymers, the  $E''$  values of E-BSt and E-VN were lower than those of E-St and E-MSt. The  $T_g$ 's determined from dynamic mechanical thermal spectroscopy, with temperatures at a maximum of  $\tan \delta$ , were higher than those determined from DSC analysis. The difference in these temperatures ( $\Delta T_g$ ) were interrelated with structure of the substituent and

increased with increasing bulkiness of the substituent in the following order: VN > BSt > MSt > St. Although the reason for these results is not yet clear, theoretical study of the glass transitions of the copolymers is underway.

### Elongation testing

Elongation testing of E-RSt copolymers was conducted at room temperature. Samples that had close molecular weights were used in the testing to exclude the effect of the molecular weight of the copolymers. The results are summarized in Table II. Some stress-strain curves of the copolymers, whose  $T_g$ 's were lower than room temperature without the crystal portion, are shown in Figure 4. The copolymer with St (61.0 wt %, sample 2) or MSt (63.4 wt %, sample 6) showed a yield point in the stress-strain curves, which indicated soft and weak properties. On the other hand, the stress increased with increasing strain, which indicated soft and ductile features, in the stress-strain curves of the copolymer with BSt (51.4 wt %, sample 7) or VN (47.0 wt %, sample 13). The difference was derived from the bulkiness of the comonomer substituent, and

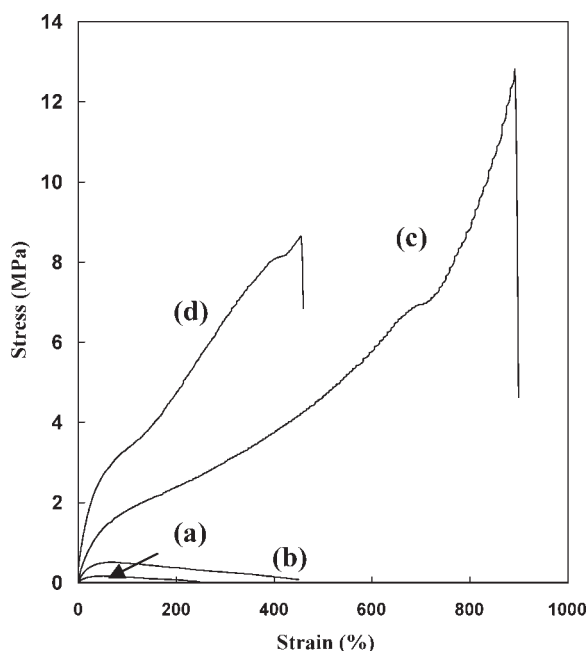
the structure may have affected the properties of the copolymers in the amorphous phase. The copolymer with high BSt, which showed a higher  $T_g$  than room temperature (sample 9), showed hard and brittle features. The properties of the copolymers with BSt were controllable from soft to hard materials by means of the control of the comonomer content.

### Shape memory effect

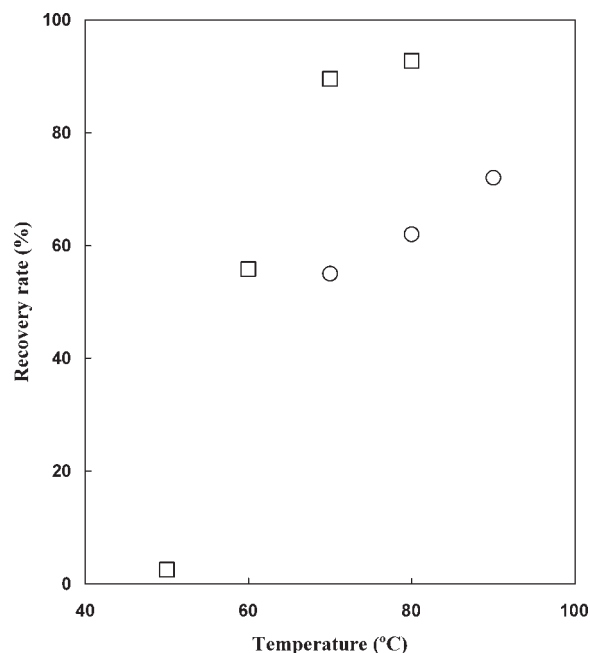
The shape memory effects of E-St (sample 3) and E-BSt (sample 9) were investigated under the conditions described in the Experimental section. The results are summarized in Figure 5. The recovery rate increased with increasing test temperature. The recovery rate of the copolymer with BSt was higher than those of the copolymer with St. The improvement in the shape memory effect in E-BSt was derived from not only the increase in  $T_g$  but also from the entanglement of the substituent *tert*-butyl groups, as previously discussed with regard to the  $T_g$  values of the copolymers.

## CONCLUSIONS

The copolymerization of ethylene and RSt was carried out with a CGC, and the thermal and mechanical properties of the copolymers were investigated. The substituent of St did not affect the monomer reactivity ratio in the copolymerization with ethylene, and the corresponding copolymers with high RSt contents were obtained in good yield. The



**Figure 4** Stress-strain curves of ethylene-substituted St copolymers: (a) sample 2, E-St; (b) sample 5, E-MSt; (c) sample 7, E-BSt; and (d) sample 13, E-VN.



**Figure 5** Relationship between the temperature and recovery rate of E-St: (○) sample 3 and (□) sample 9, E-BSt.

copolymer with BSt showed the highest  $T_g$  among the copolymers. The mechanical properties at lower temperatures than the  $T_g$  were almost independent of the substituent structure, whereas those in the amorphous state were affected by the substituent structure. In comparison with the copolymers containing about 50–60 wt % RSt, E-St and E-MSt showed soft and weak properties; on the other hand, E-BSt and E-VN showed soft and ductile properties in the amorphous state. An improvement in the shape memory effect was observed in E-BSt.

The copolymer with BSt showed a higher  $T_g$  than the copolymer with VN. The result was opposite of the theoretical consideration based on the  $T_g$  of the BSt and VN homopolymers. This unexpected result was derived from the difference in interactions between the RSt units in the homopolymer and copolymer. The branched substituent of the *tert*-butyl group enhanced the entanglement of the copolymer and caused the high  $T_g$  of the E-BSt copolymer.

The substituent structure of the RSt strongly affected not only the thermal properties but also the mechanical properties of the E-RSt copolymers. Theoretical study of the substituent effect in the E-RSt copolymers should be useful not only for clarifying the unexpected results in these experiments but also for designing the copolymer structure for certain desired properties and will be reported elsewhere.

The authors gratefully acknowledge H. Hagihara (National Institute of Advanced Industrial Science and Technology) for GPC measurements.

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