Synthesis, Characterization, and Applications of Novel Additives of Polysiloxane Containing *N*,*N*′-Bis(diphenylsilyl)tetraphenylcyclodisilazane

Yangping Zhu,[1,2] Li Guo,[1,2] Zhijie Zhang,[1] Zemin Xie,[1] Caihong Xu[1]

[1]*Center for Molecular Science, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100080, China*
[2]*Graduate School of the Chinese Academy of Sciences, Beijing 100039, China*

Received 28 April 2006; accepted 28 December 2006
DOI 10.1002/app.26100

ABSTRACT: Three *N*,*N*′-bis(diphenylsilyl)tetraphenylcyclodisilazane-based derivatives, *N*,*N*′-bis(3,3,3-trimethyl-1,1-diphenyl-disiloxanyl)tetraphenylcyclodisilazane, *N*-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl)-*N*′-(3,3,3-trimethyl-1,1-diphenyl-disiloxanyl)-tetraphenylcyclodisilazane, and *N*,*N*′-bis-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl) tetraphenylcyclodisilazane, were synthesized. These compounds were synthesized in an easy and effective route. X-ray single-crystal diffraction analyses showed that the four-member rings were planar rings, and the structures are different with the different substitution. The compound *N*,*N*′-bis-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl) tetraphenylcyclodisilazane was added to the silicone rubber as additive to enhance the thermal stability greatly increased the thermal stability of the silicone rubber, without altering the glass transition temperature. The weight loss at 350°C in nitrogen atmosphere for 24 h reduced from 55.8% for 0 wt % to 9.8% for 10 wt % addition *N*,*N*′-Bis-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl)tetraphenylcyclodisilazane. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 105: 749–756, 2007

Key words: thermal properties; additives; polysiloxanes; high temperature materials; synthesis

INTRODUCTION

Silicone rubbers have many excellent properties such as good resistance to heat and to cold, good hydrophobicity, and good electroinsulating properties; and have been widely used in many devices to withstand very crucial conditions or environments.[1,2] The increasing development of the aviation, aerospace, and so on is bringing the demands for polysiloxanes resisting higher temperatures. There are mainly two methods to improve the thermal properties of the silicone rubber. One method is change the main-chain structure. Polyarylene-siloxanes,[3–5] poly-*m*-carboranesiloxanes,[6–9] and polysiloxanes containing *N*,*N*′-bis(diphenylsilyl)tetramethylcyclodisilazane[10–13] were of this type. They had very good thermal stability via introduce the bulky group and reactive group respectively. However, the applications of these materials were restricted by the synthesis of the monomers, especially poly-*m*-carboranesiloxanes and polysiloxanes containing *N*,*N*′-bis(diphenylsilyl)tetramethylcyclodisilazane. The other method is adding a thermal stabilizer. Hexphenylcyclotrisilazane was used as additive to improve the thermal stability of the silicone rubber[14] and increase the resistance to high temperature, whereas its thermal resistance a declined in few months later because of the bad compatibility. As one of the methods of overcoming these problems, the facile synthesized *N*,*N*′-bis(diphenylsilyl)tetraphenylcyclodisilazane was employed as additives of silicone rubber instead of *N*,*N*′-bis(diphenylsilyl)tetramethylcyclodisilazane.[15,16] To avoid the probably problems like hexphenylcyclotrisilazane, vinyl group was used with these compounds to crosslink with silicone rubber. These compounds were prepared in an easy route and were well characterized. One compound was added to silicone rubber as an additive, which greatly increased the thermal stability of silicone rubber.

EXPERIMENTAL

General comments

Diphenyldicholorosilane, hexmethyldisilazane, and trimethylchlorosilane were purchased from Beijing Chemical Works and fractional distilled before use. Vinyldimethylchlorosilane was purchased from Aldrich and was used without any further purification. Solvents were purified according to reported methods before use. *N*-Butyllithium (2.5 mol/L in *n*-hexane)

Correspondence to: Z. Zhang (zhangzj@iccas.ac.cn).

Journal of Applied Polymer Science, Vol. 105, 749–756 (2007)

was purchased from Acros Company and was used as received. Phenyl silicone rubber used, containing 16% diphenylsilicone and 0.425% vinyl, has a M_n of 510,000.

^{1}H NMR and ^{29}Si NMR were preformed on Bruker DMX300 spectrometer at 300 MHz using tetramethylsilane as an external reference. Melting points were measured with WRS-1A Digital Melting Point Apparatus. Elemental analyses were performed by Center for Physiochemical Analysis and Measurement, Institute of Chemistry, Chinese Academy of Science, Beijing. High-resolution mass spectrometry was performed with an APEX II FT-ICRMS spectrometer. DSC was carried out in a Mettler Tol DSC8200^e in nitrogen atmosphere at heating rate of 10°C/min. TG was carried out in a PerkinElmer Pyris 1 Thermogravimetric Analyzer in nitrogen atmosphere at a flow rate of 60 mL/min. The weight of sample used was 5–10 mg.

Preparation of 1,3-dichloro-1,1,3,3-tetraphenyldisilazane (1)

A mixture of Ph_2SiCl_2 (506 g, 2 mol) and hexmethyldisilazane (MM^N, 161 g, 1 mol) was stirred at 120°C to distill trimethylcholrosilane for over 24 h. After trimethylcholrosilane was totally distilled out, the mixture was cooled to room temperature. The crude product was recrystallized from the mixed solvent of *n*-hexane and toluene to give 348 g (yield 77.24%) of pure **1** as white crystals. Melting point: 110.3–111.1°C.

^{1}H NMR ($CDCl_3$), δ (ppm): 2.5 (s, 1H, NH), 7.2–7.6 [m, 20H, $(C_6H_5)_4$]. ^{29}Si NMR($CDCl_3$), δ (ppm): −8.25 [s, $NSi(C_6H_5)_2Cl$]. Anal. Calcd for $C_{24}H_{21}Si_2NCl_2$ (%): C, 63.99; H, 4.70; N, 3.11; Cl, 15.74. Found (%): C, 63.70; H, 4.56; N, 2.91; Cl, 15.49.

Synthesis of *N,N′*-bis-(chloro-diphenyl-silanyl) tetraphenylcyclodisilazane (2)

Compound **1** (135 g, 0.3 mol) was dissolved in 600 mL benzene. To this was added *n*-butyllithium (2.5*M*/L in *n*-hexane, 120 mL) in ice-water bath. The solution was warmed to room temperature and stirred for 4 h, after which *n*-hexane was removed. The mixture was refluxed under stirring for 12 h. The reaction mixture was filtered quickly and benzene was removed under reduced pressure. The crude product was recrystallized from the mixed solvent of toluene and *n*-hexane to give 176 g pure colorless crystal (yield 71%). Melting point: 256.5–257.4°C.

^{1}H NMR ($CDCl_3$), δ (ppm): 6.8–7.6 [m, $(C_6H_5)_8$]. ^{29}Si NMR ($CDCl_3$), δ (ppm): −12.36 [s, $NSi(C_6H_5)_2Cl$], −13.60 [s, $NSi(C_6H_5)_2N$]. Anal. Calcd for $C_{48}H_{40}Si_4N_2Cl_2$ (%): C, 69.62; H, 4.87; N, 3.38; Cl, 8.56. Found (%): C, 69.61; H, 4.95; N, 3.46; Cl, 8.67.

Synthesis of *N,N′*-bis-(hydroxy-diphenyl-silanyl) tetraphenylcyclodisilazane (3)

A suspension of compound **2** (124 g, 0.15 mol) in 200 mL ethyl ether was added dropwisely to a mixture of ethyl ether (200 mL), distilled water (30 mL), and ammonia water (30 mL) in ice-water bath. The mixture was then stirred at room temperature for 1 h. The precipitated crystals were filtered out and washed with *n*-hexane. The crude product was recrystallized from the mixed solvent of THF and *n*-hexane to give 89.32 g (yield 75.3%) of pure white crystals of cyclodisilazane **3**. Melting point: 222.7–223.6°C.

^{1}H NMR ($CDCl_3$), δ (ppm): 2.05 (s, 2H, OH), 6.8–7.5 [m, 40H, $(C_6H_5)_8$]. ^{29}Si NMR($CDCl_3$), δ (ppm): −32.84 [s, $NSi(C_6H_5)_2OH$], −15.89 [s, $NSi(C_6H_5)_2N$]. Anal. Calcd for $C_{48}H_{42}Si_4N_2O_2$ (%): C, 72.87; H, 5.35; N, 3.54. Found (%): C, 72.89; H, 5.44; N, 3.29.

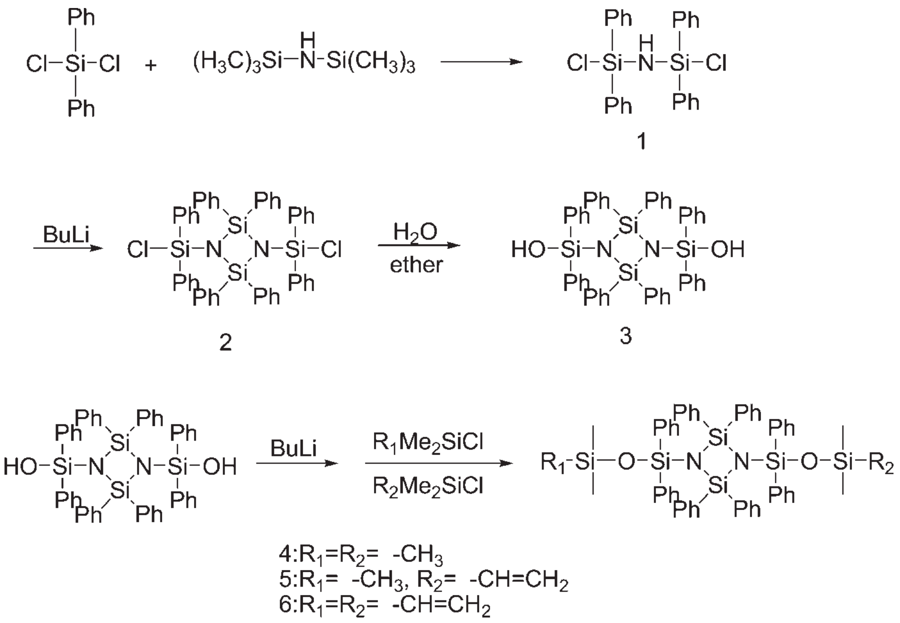
Synthesis of *N,N′*-bis(3,3,3-trimethyl-1,1-diphenyldisiloxanyl)tetraphenylcyclodisilazane (4)

Compound **3** (2.37 g, 3 mmol) was dissolved in 50 mL THF and was added to *n*-butyllithium (6 mmol in *n*-hexane) at −78°C. After the solution was warmed to room temperature for 4 h, Me_3SiCl (20 mmol) was added to this solution. The solution was warmed to reflux for 4 h; THF and *n*-hexane were then removed. About 150 mL toluene was added to the residue, and the solution was stirred for 4 h. Subsequently the precipitate was filtered out, and the solvent toluene was removed. The crude product was recrystallized from the mixed solvent of THF and *n*-hexane to give 2.04 g pure colorless crystal (yield 72.70%).

^{1}H NMR ($CDCl_3$), δ (ppm): −0.30 (s, 18H, $SiCH_3$), 6.9–7.5 [m, 40H, $(C_6H_5)_8$]. ^{29}Si NMR($CDCl_3$), δ (ppm): 8.53[s, $Si(CH_3)_3O$], −14.20[s, $NSi(C_6H_5)_2N$], −36.39[s, $NSi(C_6H_5)_2O$]. HRMS (SIMS) *m/z* $[M + H]^+$ for $[C_{54}H_{58}N_2O_2Si_6 + H]^+$ Calcd: 935.3186, Found: 935.3181.

Synthesis of *N*-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl)-*N′*-(3,3,3-trimethyl-1,1-diphenyldisiloxanyl)-tetraphenylcyclodisilazane (5)

Compound **3** (2.37 g, 3 mmol) was dissolved in 50 mL THF and was added to *n*-butyllithium (6 mmol in *n*-hexane) at −78°C. After the solution was warmed to room temperature for 4 h, a mixture of $Me_2ViSiCl$ (10 mmol) and Me_3SiCl (10 mmol) in 20 mL THF was added to this solution in 4 h. The solution was refluxed for 4 h; THF and *n*-hexane were then removed. About 150 mL toluene was added to the residue and the solution was stirred for 4 h. The precipitate was filtered out, and the solvent toluene was removed. The crude product was recrystallized from the mixed solvent of THF and *n*-hexane to give 2.16 g pure colorless crystal (yield 76%).



Scheme 1

^{1}H NMR ($CDCl_3$), δ (ppm): −0.3355 [s, 6H, ($CH{=}CH_2$)Si(CH^*_3)$_2$], −0.2794 [s, 9H, Si(CH^*_3)$_3$], 6.86–7.48 [m, 40H, (C_6H_5)$_8$], 5.37 (m, 1H, SiCH=C), 5.72(t, 2H, $C{=}CH_2$). ^{29}Si NMR($CDCl_3$), δ (ppm): 7.7 [s, Si(CH_3)$_3$O], −3.6 [s, ViSi(CH_3)$_2$O], −15.0[s, NSi(C_6H_5)$_2$N], −36.9 [s, NSi(C_6H_5)$_2$O]. HRMS (SIMS) *m/z* $[M + H]^+$ for $[C_{55}H_{58}N_2O_2Si_6 + H]^+$ Calcd: 947.3186, Found: 947.3163.

TABLE I
The X-ray Crystallography Data of Compounds 4, 5, and 6

	Compound 4	Compound 5	Compound 6
Molecular formula	$C_{54}H_{58}N_2O_2Si_6$	$C_{55}H_{58}N_2O_2Si_6$	$C_{56}H_{58}N_2O_2Si_6$
Formula weight	935.56	947.57	959.58
Crystal system	Monoclinic	Triclinic	Triclinic
Space group	$C2/c$	$P1$	$P\bar{1}$
a (Å)	13.601 (3)	10.583 (2)	10.579 (1)
b (Å)	16.848 (3)	11.316 (2)	11.329 (1)
c (Å)	22.675 (5)	13.099 (3)	13.049 (1)
α (°)	90	65.76 (3)	66.01 (1)
β (°)	91.37 (3)	69.94 (3)	70.05 (1)
γ (°)	90	78.41 (3)	78.32 (1)
V (Å^3)	5194.8 (18)	1340.2 (5)	1339.5 (2)
Z	4	1	1
Crystal description	Platelet, colorless	Block, colorless	Platelet, colorless
Crystal size (mm)	0.79 × 0.41 × 0.28	0.73 × 0.63 × 0.55	0.77 × 0.76 × 0.46
Temperature (°C)		23	
Radiation		Mo-Kα (λ = 0.71073 Å) graphite monochromated	
μ (Mo-Kα)	0.202	0.196	0.197
θ_{max} (°)	27.48	27.35	27.48
	$-17 \leq h \leq 17$	$-13 \leq h \leq 13$	$-13 \leq h \leq 13$
	$-21 \leq k \leq 21$	$-14 \leq k \leq 14$	$-14 \leq k \leq 14$
	$-29 \leq l \leq 29$	$-15 \leq l \leq 16$	$-15 \leq l \leq 16$
Total data collected	5875	12,046	12,506
No. of indep. Reflns.	5875	5947	6058
R_{int}	0.0500	0.0367	0.0337
Absorp. corr.		Empirical	
Transmission: t_{min}/t_{max}	0.9464/0.8567	0.9003/0.8695	0.9143/0.8633
No. of data/restr/params	5875/0/289	5947/3/586	6058/0/298
$R[F^2>2\sigma(F^2)]$	0.0455	0.0426	0.0462
$wR(F^2)$	0.1332	0.1165	0.1330
Goodness-of-fit	1.033	1.042	1.045

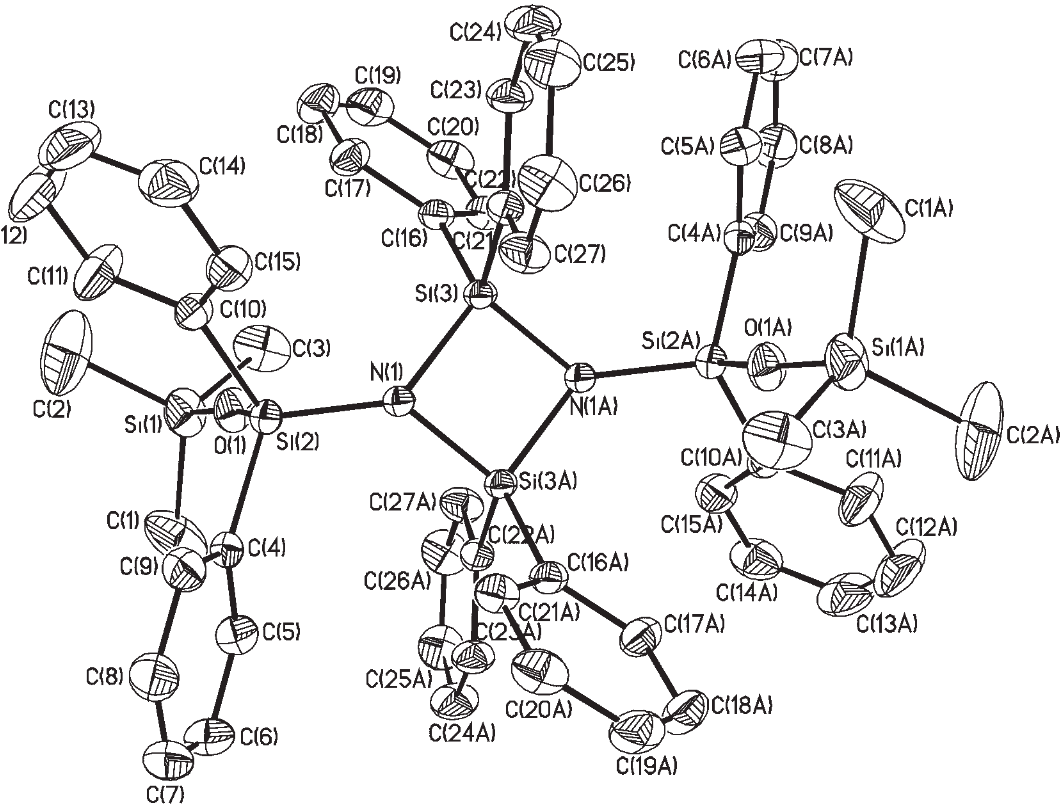


Figure 1 A view of the molecular structure of compound **4**, with displacement ellipsoids showing at the 30% probability level. H atoms of phenyl and methyl group are not shown for clarity.

Synthesis of *N*,*N*′-bis-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl)tetraphenylcyclodisilazane (6)

Compound **3** (2.37 g, 3 mmol) was dissolved in 50 mL THF and was added to *n*-butyllithium (6 mmol in *n*-hexane) at −78°C. After the solution was warmed to room temperature for 4 h, $Me_2ViSiCl$ (20 mmol) was added to this solution. The solution was refluxed for 4 h, after which THF and *n*-hexane was removed. About 150 mL toluene was added to the residue and the solution was stirred for 4 h. The precipitate was then filtered out, and solvent toluene was removed. The crude product was recrystallized from the mixed solvent of THF and *n*-hexane to give 2.34 g pure colorless crystal (yield 83.39%).

^{1}H NMR ($CDCl_3$), δ (ppm): −0.24 (s, 12H, $SiCH_3$), 6.9–7.5 [m, 40H, $(C_6H_5)_8$], 5.38 (m, 2H, SiCH=C), 5.72(t, 4H, C=CH_2). ^{29}Si NMR($CDCl_3$), δ (ppm): −2.69 [s, ViSi $(CH_3)_2O$], −14.09 [s, $NSi(C_6H_5)_2N$], −36.05 [s, NSi $(C_6H_5)_2O$]. HRMS (SIMS) *m/z* $[M + H]^+$ for $[C_{56}H_{58}N_2O_2Si_6 + H]^+$ Calcd: 959.3186, Found: 959.3173.

X-ray crystallography

Single crystals of compound **4**, **5**, and **6** suitable for X-ray crystallographic analyses were grown by slow evaporation of their respective solution in THF/*n*-hexane at room temperature. The crystals were chosen and mounted on a glass fiber using epoxy resin.

The diffraction experiments were carried out at 276 K on a Rigaku RAXIS-RAPID diffractometer with graphite-monochromated Mo Kα radiation (λ = 0.71073Å). Data were processed using PROCESS-AUTO[17] program package, and absorption corrections were applied by the empirical method.[18]

The structures were solved by direct method using SIR92,[19] and refined on F^2 (with all independent reflections) by the full-matrix least-squares technique using SHELXL97 program.[20] All nonhydrogen atoms were refined anisotropically, and hydrogen atoms were introduced at the positions calculated theoretically and treated with riding models.

Vulcanization of the silicone rubber

Compounds **6** and **4** were added to phenyl silicone rubber as additives, and the mixtures were heated at 200°C for 24 h. Then the vulcanization was processed via dicumyl peroxide (DCP) at temperature of 150°C and pressure of 9–10 MPa for about 20 min.

RESULTS AND DISCUSSION

Synthesis and characterization

It is well known that the impurities in the polysiloxanes greatly affect the thermal stability of the polysiloxane. Our focus was on the synthesis of the deriva-

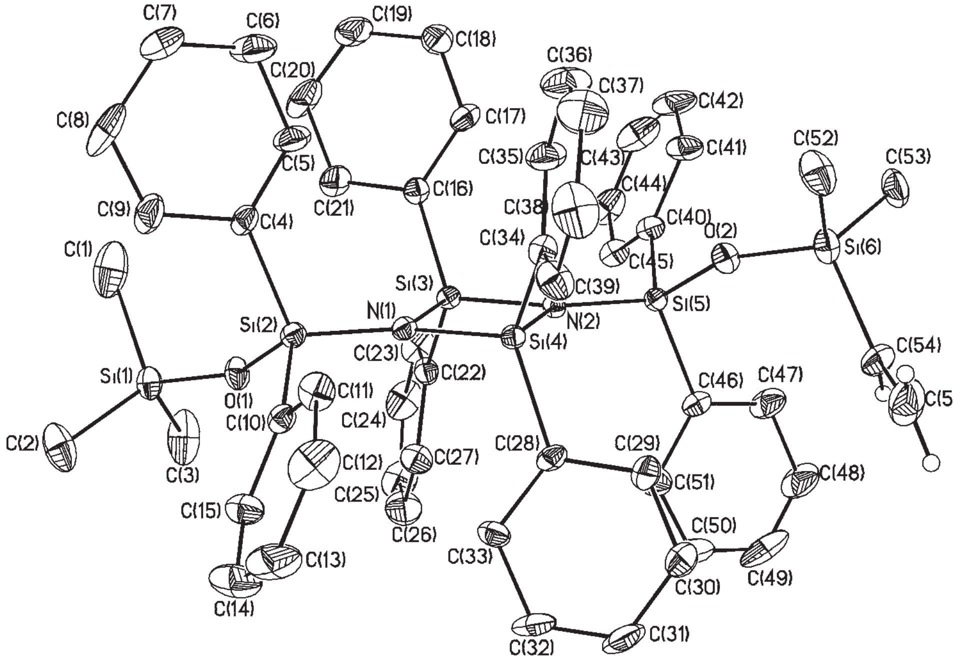


Figure 2 A view of the molecular structure of compound **5**, with displacement ellipsoids showing at the 30% probability level. H atoms of phenyl and methyl group are not shown for clarity.

tive of the *N*,*N*′-bis(diphenylsilyl)tetraphenylcyclodisilazane with the expectation that these small molecules would be easier to be synthesized, purified, and characterized than the corresponding polymers. The compounds were synthesized in the Scheme 1.

In this study, hexmethyldisilazane was used as a starting material to synthesize 1,3-dichloro-1,1,3,3-tetraphenyldisilazane (DCTPS). The reaction was processed in a moderate temperature (about 120°C), and the yield of DCTPS is about 78%. The method we

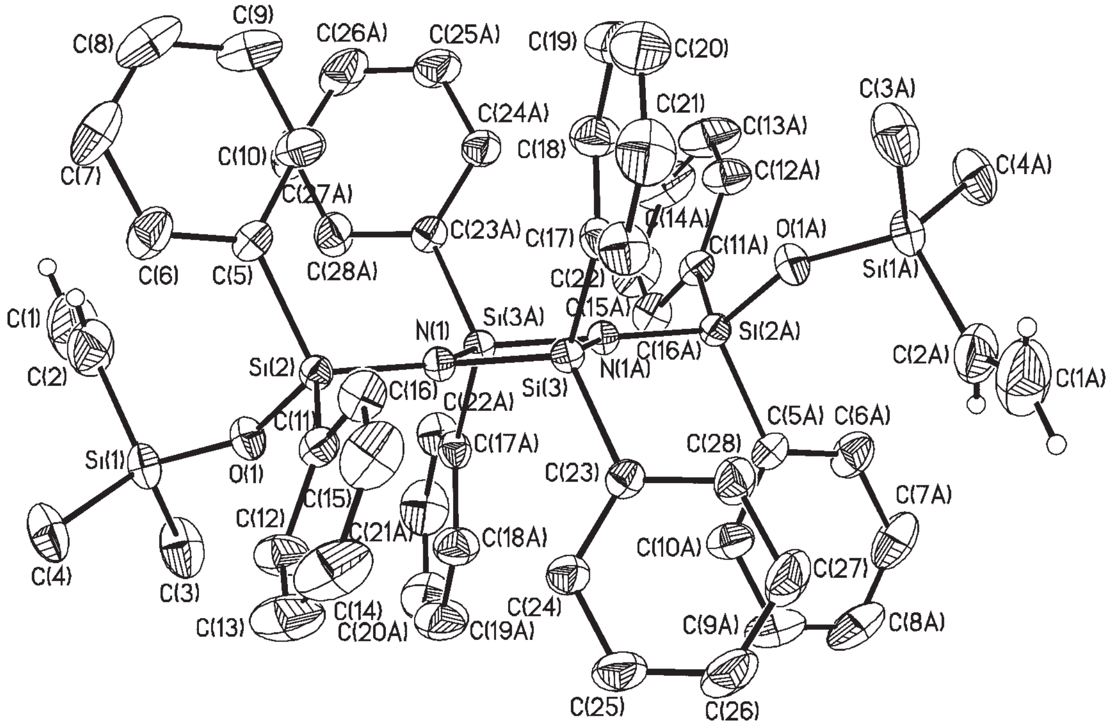


Figure 3 A view of the molecular structure of compound **6**, with displacement ellipsoids showing at the 30% probability level. H atoms of phenyl and methyl group are not shown for clarity.

TABLE II
DSC Data of the Silicone Rubber Blends With Different Weight Percent of Compound 6

Weight percent of compound 6	Tg (°C)
0	−87
5	−85.61
10	−85.72

used is much simpler and more effective than the reported. In the literature, the DCTPS was synthesized by the reaction of octamethylcyclotetrasilazane (D_4^N) or hexmethylcyclotrisilazane (D_3^N) with excess diphenyldichlorosilane and was carried out at a high temperature, at least 180°C, and the yield was no more than 70%.[16,21]

N,*N*′-Bis-(chloro-diphenyl-silanyl)tetraphenylcyclodisilazane (BCPTPC) was synthesized by a ring-closure reaction of DCTPS, illustrated in Scheme 1, as described by Breed and Wiley.[22] In the literature, the solvent is xylene, and the react temperature is about 140°C. In our study, benzene was chosen as the solvent for the reaction, and the temperature of the ring-closure reaction was changed to 80°C, meanwhile the yield of BCPTPC did not change very much. The product was characterized using ^{29}Si NMR, ^{1}H NMR, and element analysis. In the ^{1}H NMR spectra, the multiple peaks at 6.8–7.6ppm correspond to the hydrogen in —C_6H_5. In the ^{29}Si NMR spectra, the single peak at −12.36ppm corresponds to the exocyclic silicon atoms, and the single peak at −13.60ppm corresponds to the endocyclic silicon atoms.

By using a suspension of BCPTPC in ethyl ether dropped to a mixture of ammonia water and ethyl ether, *N*,*N*′-bis-(hydroxy-diphenyl-silanyl)tetraphenylcyclodisilazane (BHPTPC), the hydrolysis product of BCPTPC, was obtained. The chemical shifts of the exocyclic and endocyclic silicon atoms changed to −32.84 and −15.89ppm, respectively.

By the lithiation of the BHPTPC, three derivatives of the *N*,*N*′-bis(diphenylsilyl)tetraphenylcyclodisilazane (**4**, **5**, and **6**) were synthesized. All these three compounds were characterized using NMR, high-resolutional mass spectrometry, and single crystal diffraction methods. The chemical shift of the exocyclic silicon atoms attached to the nitrogen atoms changed to −36.39, −36.90, and −36.0ppm. The endocyclic silicon atoms also have about 1 ppm changes in the ^{29}Si NMR spectrums. All the three compounds are colorless crystal. The melting points of the compound **4**, **5**, and **6** were high; therefore, DSC method was used to measure the melting point of these compounds with a heating rate of 10°C min^{-1}. The melting points of compound **4**, **5**, and **6** are 338.62, 323.82, and 338.62°C.

Crystal structures

The structures of the compounds **4–6** were determined by X-ray crystallography in this study. Their crystallography data are listed in Table I, and their crystal drawings are shown in Figure 1–3. Like most other cyclodisilazanes,[23,24] all these compounds have a planar four-member ring. However, the differences of the exocyclic silicon atoms attached to the nitrogen atoms are caused by the different substitute. The distance between the silicon atoms and the four-member ring in compound **4** is 0.497 Å, whereas the values of which are 0.140 and 0.129 Å for compound **5** and 0.132 Å for compound **6**, respectively. The structure of compound **5** indicated that the different-substituted derivative could be obtained via the slow addition of the mixed chlorosilane. This provided a method to synthesize different functional compounds.

Thermal analysis

As an additive of the silicone rubber to improve the thermal stability, it is necessary to evaluate the thermal properties of the silicone rubber containing the additive. In this study, compound **6**, as the additive, was added to the silicone rubber in different weight percent.

To evaluate the influence of the additives to the glass transition temperature, the samples were analyzed by DSC method. The result was shown in Table II. The result suggested that the addition of the compound **6** caused slightly rise of the glass transition temperature, but did not reduce the temperature range of use in the cold temperature.

TGA and IGA methods were used to characterize thermal stability of the silicone rubber. The TGA and DTG curves were shown in Figures 4 and 5. The addi-

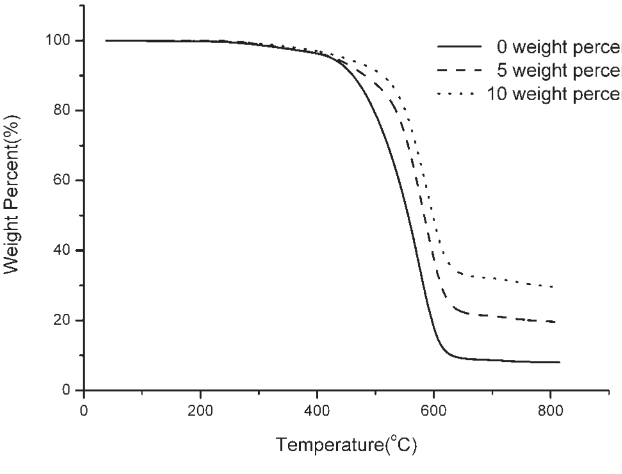


Figure 4 TG curves of the silicone rubber blends with different weight percent of compound **6**.

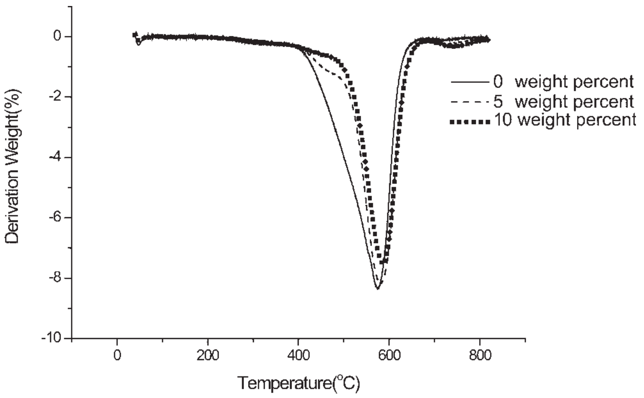


Figure 5 DTG curves of the silicone rubber blends with different weight percent of compound **6**.

tion of the additive caused the rise of the decomposition onset temperature and residue weight in TGA curves. At the heating rate of 10°C/min, the decomposition onset temperatures were 466.06°C, 508.76°C, and 518.90°C, and the residue weight was 8.0, 19.6, and 29.4 wt % respectively. The residue weight was increased by the addition of the additive. This indicated that the additive react with the polysiloxane in the degradation of the polysiloxane. In the DTG curves, the addition of the additive led the rise of the maximum decomposition temperature, 575.26°C, 580.29°C, and 586.21°C, respectively.

IGA method is an important method to characterize thermal stability of polysiloxane. At a given temperature, the lesser the weight loss, the better thermal stability the material has. The IGA experiments were carried out at 350°C in the nitrogen atmosphere at a flow rate of 60 mL/min. The result was shown in the Figure 6. After 24h, the weight loss of the blank sample was 55.8 wt %, whereas that of the added 5 and 10 wt % compound **6** were 27.1 and 9.8 wt %, respectively. The addition of the compound **6** greatly increased the thermal stability of the silicone rubber.

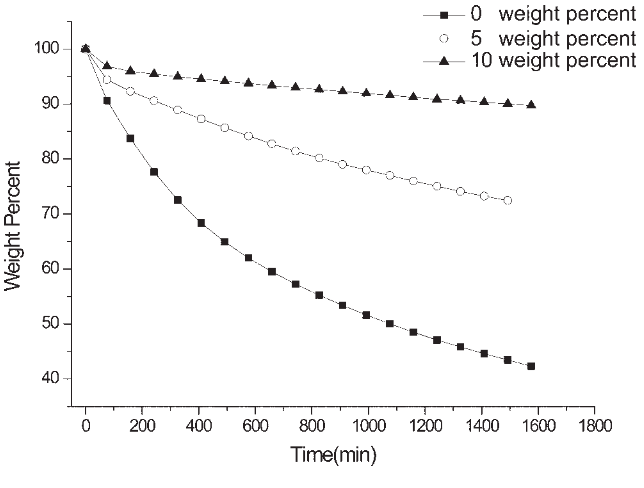


Figure 6 The IGA curve of the silicone rubber blends with different weight percent of compound **6** at 350°C.

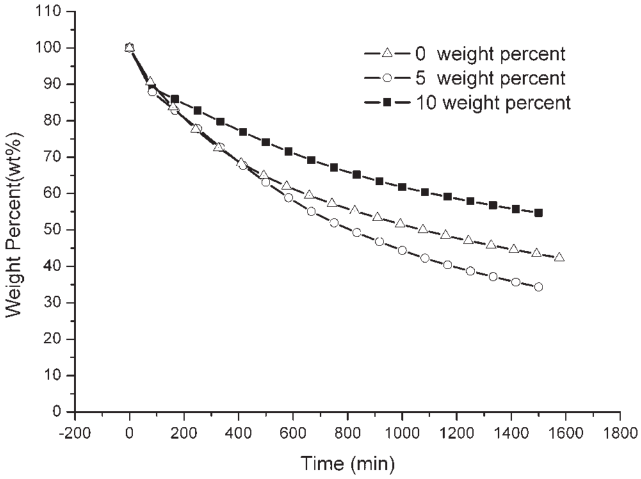
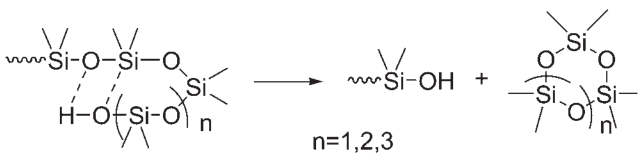


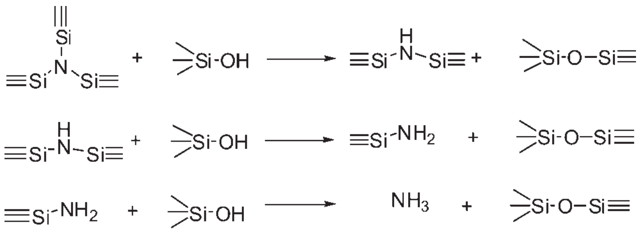
Figure 7 The IGA curve of the silicone rubber blends with different weight percent of compound **4** at 350°C.

The introduced vinyl group played an important role in the degradation at high temperature. As a comparison, the compound **4** was also added to silicone rubber as additive. The IGA method was carried out at 350°C in nitrogen atmosphere. The result was shown in Figure 7. The result was quite different from that of compound **6**. The addition of compound **4** did not increase the thermal stability of the silicone rubber. After 24 h, the weight loss adding 5% compound **4** is even greater than without adding compound **4** as shown in Figure 7.

Xie et al.[12] have provided strong supporting evidence that the most important reason for weight loss of silicone rubber at high temperature in nitrogen atmosphere is the main-chain degradation by forming small cyclosiloxanes, caused by residue SiOH or the adsorptive water in polysiloxane (see Scheme 2). Silicon–nitrogen compounds can react with SiOH and water to form the Si—O—Si bonds (see Scheme 3). Through eliminating the SiOH and water in the silicone rubber, the main-chain degradation is decreased, and then the thermal stability is improved as shown in Figure 6. However, in the open atmosphere at high temperature, the uncrosslinked small molecular silicon-nitrogen compounds are trend to volatilize to the atmosphere, and the thermal stability of the polysilox-



Scheme 2



Scheme 3

anes was improved only at high addition of the additives, as shown in Figure 7.

CONCLUSIONS

Three *N*,*N*′-bis(diphenylsilyl)tetraphenylcyclodisilazane-based derivatives were synthesized. These compounds were synthesized in an easy and effective route. X-ray single-crystal diffraction analyses showed that the four-member rings were planar rings, and the structures are different with the different substitution. The compound *N*,*N*′-bis-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl)tetraphenylcyclodisilazane added to the silicone rubber as additive to improve thermal stability greatly increased the thermal stability of the silicone rubber, without altering the the glass transition temperature. The weight loss at 350°C in nitrogen for 24 h was reduced from 55.8% for 0 wt % addition to 9.8% for 10 wt % addition *N*,*N*′-Bis-(3,3-dimethyl-1,1-diphenyl-3-vinyl-disiloxanyl)tetraphenylcyclodisilazane. The introduced vinyl group also played an important role in the degradation at high temperature. The addition of the nonvinyl-containing compound did not increase the thermal stability of silicone rubber at high temperature.

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