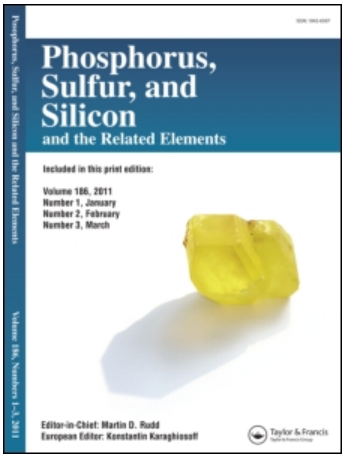

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Synthesis and Characterization of Cyclotriphosphazenes Bearing Chalcones Derivatives

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SYNTHESIS AND CHARACTERIZATION OF CYCLOTRIPHOSPHAZENES BEARING CHALCONES DERIVATIVES

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A series of new cyclotriphosphazenes bearing chalcones derivatives, $N_3P_3Cl_5[OC_6H_4CH{=}CHC(O)C_6H_4OC_nH_{2n+1}]$ and $N_3P_3[OC_6H_4CH{=}CHC(O){-}C_6H_4OC_nH_{2n+1}]_6$, has been synthesized. A convenient synthetic method was performed from the reaction of hexachlorocyclotriphosphazenes with one and six equivalents of (E)-3-(4-(alkyloxy)phenyl)-1-(4-hydroxyphenyl)prop-2-en-1-one (2a–c). The compounds differ in the length of alkyl groups, C_nH_{2n+1}, where n = 10, 12, and 14, respectively. All the products were obtained in high yields. The structures of the synthesized compounds were defined by elemental analysis, IR, ^1H, ^{13}C, and ^{31}P NMR.

Keywords Alkyloxy; chalcones; hexachlorocyclotriphosphazenes

INTRODUCTION

Phosphazenes are compounds that contain a framework of alternating phosphorus and nitrogen atoms, either in cyclic or linear form.[1] Studies on linear, cyclo-, and polyphosphazenes have been widely investigated. These compounds are reported to possess interesting biomedical properties[2] and have promising applications such as effective flame retardants for fiber materials.[3] Nucleophilic substitutions of hexachlorocyclotriphosphazenes have been widely reported. The reaction involves the substitution of chlorines by various nucleophiles such as phenols,[4,5] amine,[6] and azo compounds.[7]

Synthesis of cyclotriphosphazenes bearing cinnamates[8] and hydroxychalcones[9] as side groups had been studied for photosensitive phosphazenes that could undergo photocross-linking reaction under UV irradiation. In photochemistry, chalcone derivatives have been reported to possess outstanding nonlinear optic properties for optical communications and optical electronics,[10] liquid crystal displays,[11,12] and alignment film.[13] Chalcones have also been reported to promote excellent blue light transmittance and good crystallability,[14,15] high photosensitivity, and thermal stability for various crystalline electro-optical devices.

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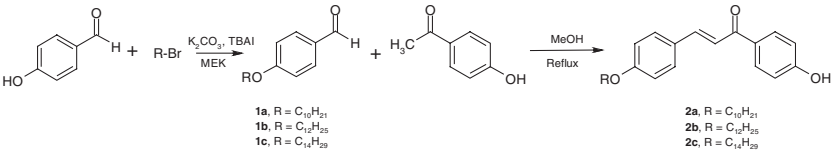
In this article, we report the new synthesis of cyclotriphosphazenes bearing chalcone derivatives (E)-3-(4-(alkyloxy)phenyl)-1-(4-hydroxyphenyl)prop-2-en-1-one (**2a–c**), which could be used as a reaction models for various crystalline electro-optical devices.

RESULTS AND DISCUSSION

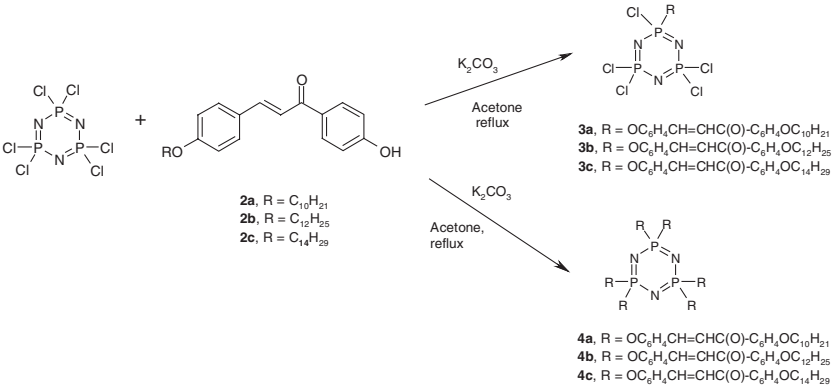
The series of chalcone derivatives (E)-3-(4-(alkyloxy)phenyl)-1-(4-hydroxyphenyl) prop-2-en-1-one (**2a–c**)[16] were first prepared via Claisen–Schmidt condensation of **1a–c**[17] and 4-hydroxyacetophenone by the route depicted in Scheme 1.



Scheme 1 Synthesis of chalcone derivatives **2a–c**.

The IR spectra of chalcone derivatives **2a–c** showed the presence of bands at 2849–2922 cm^{-1}, which were attributed to the introduction of the long alkyl chain via etherification of 4-hydroxybenzaldehyde. The presence of a new C=O stretching frequency at 1636–1648 cm^{-1} also substantiated the formation of the title compound. The structures of compounds **2a–c** were further confirmed by ^1H and ^{13}C NMR spectra. The ^1H NMR spectra of **2a–c** show the new peaks attributed to *trans* vinylic proton at 7.39–7.41 and 7.67–7.77, respectively, with a coupling constant, J_{ab} 15 Hz. The ^{13}C NMR spectra show the chemical shift of C=O within the chemical shift range of 188.73–190.14 ppm, and C=C peaks appeared at 127.48–130.18 ppm and 144.25–144.44 ppm, respectively. The elemental analysis is comparable to the theoretical value.

The synthetic route for the preparation of $N_3P_3Cl_5[OC_6H_4CH{=}CHC(O)C_6H_4OC_nH_{2n+1}]$ **3a–c** and $N_3P_3[OC_6H_4CH{=}CHC(O){-}C_6H_4OC_nH_{2n+1}]_6$ **4a–c** is illustrated in Scheme 2. Monosubsubstituted cyclotriphosphazenes **3a–c** were obtained from the reaction of hexachlorocyclotriphosphazenes with one equivalent of chalcone derivatives **2a–c**



Scheme 2 Synthesis of **3a–c** and **4a–c**.

in the presence of K_2CO_3 in acetone. The higher polarity of the acetone[18] was believed to increase the rate of reaction compared to THF[9] and dioxane.[19] The structures of the compounds were characterized by elemental analysis, IR, ^1H, ^{13}C, and ^{31}P NMR spectroscopy. The IR spectra showed P=N stretching vibrations at 1162 cm^{-1}, which are characteristic of cyclotriphosphazenes.[20,21] The absorption bands observed at 951 cm^{-1} were attributed to the presence of the P—O—C bond.[22] The characteristic peak at 1658 cm^{-1} corresponded to C=O. The ^{31}P NMR spectra showed two resonances as a triplet and doublet at 12.53 ppm and 23.00 ppm, respectively, with a coupling constant, J 60 Hz, which implied the replacement of one chlorine from the cyclotriphosphazene ring for the monosubstituted phosphazenes.[23] The ^1H and ^{13}C NMR data also confirmed the substitution of **2a–c**, with the chemicals shift moved slightly downfield.

The reaction of hexachlorocyclotriphosphazenes with six equivalents of **2a–c** under the same reaction conditions afforded **4a–c** in high yield. The IR spectra showed the characteristic absorption bands at 1162 cm^{-1}, which were attributed to P=N stretching vibrations. The presence of new bands at 951 cm^{-1} in **4a–c** is a strong indication for the formation of P—O—C bonds. ^{31}P NMR showed a single resonance at δ 9.03 ppm, which implied complete chlorine replacement.[23] The data obtained from elemental analysis, ^1H, and ^{13}C NMR showed good agreement to the corresponded structures.

CONCLUSION

A series of monosubstituted cyclotriphosphazenes was obtained from the reaction of hexachlorocyclotriphosphazenes with one equivalent of chalcone derivatives **2a–c**. Hexasubstituted cyclotriphosphazenes were obtained from the reaction of hexachlorocyclotriphosphazenes with six equivalents of chalcone derivatives **2a–c** via replacement of all the chlorine atoms.

EXPERIMENTAL

Materials

4-Hydroxybenzaldehyde, 4-hydroxyacetophenone, and 1-bromoalkanes were obtained from Merck Company and used without further purification. Hexachlorocyclotriphosphazene was provided by Aldrich and was recrystallized from hexane. Acetone was distilled from calcium hydride under nitrogen before use. All other reagents and solvent were used as received. The reactions were performed under dry nitrogen.

Measurements

The melting points of the synthetic products were determined on a melting point measurement device and uncorrected. Infrared spectra were recorded on a (FT-IR) 1605 Shimadzu Spectrometer as neat liquid films technique between sodium chloride plate or potassium bromide pellet (KBr) for solid materials. ^1H NMR spectra were recorded at 500 MHz on a Jeol Delta 2-NMR. ^{13}C NMR was recorded at 125.77 MHz. The chemical shifts for ^{31}P NMR are relative to the internal standard of 85% phosphoric acid.

4-Decyloxybenzaldehyde (1a)[17]

A mixture of 4-hydroxybenzaldehyde (6.11 g, 50 mmol), K_2CO_3 (8.29 g, 60 mmol), bromodecane (12.40 mL, 60 mmol), and TBAI (1.85 g, 5 mmol) in MEK (150 mL) was heated at reflux for 12 h. The mixture was filtered and cooled at room temperature. Water (30 mL) was added to the filtrate, and the layers were separated. The aqueous layer was extracted with dichloromethane (2 × 30 mL). The combined layers were washed with water (2 × 20 mL), dried ($MgSO_4$), filtered, and concentrated in vacuo. The crude product was purified by column chromatography (eluting with 1:20 ethyl acetate:petroleum ether) to afford **1a** (8.69 g, 66%) as a viscous brown oil. The FTIR and NMR data were consistent with the reported literature.[16] The same general procedure gave compounds **1b–c**, with the scale (mL, mmol, [bromoalkane]) and yields given below.

4-Dodecyloxybenzaldehyde (1b). Bromododecane (14.38 mL, 60 mmol). Yield: 13.03 g, 90%. The FTIR and NMR data were consistent with the reported literature.[16]

4-Tetradecyloxybenzaldehyde (1c). Bromotetradecane (16.31 mL, 60 mmol). Yield: 11.78 g, 74%. The FTIR and NMR data were consistent with the reported literature.[16]

(E)-3-(4-Decyloxyphenyl)-1-(4-hydroxyphenyl)prop-2-en-1-one (2a)[16]

A mixture of 4-hydroxyacetophenone (2.72 g, 20 mmol) and **1a** (5.25 mL, 20 mmol) in 60 mL of methanol was added under stirring to a solution of KOH (4.04 g, 72 mmol) in methanol (10 mL). The mixture was heated at reflux for 10 h. The reaction was cooled to room temperature and acidified with cold diluted HCl (2N). The resulting precipitate was filtered, washed, and dried. The crude product was recrystallized from hexane:ethanol (7:1) to give **2a** (6.83 g, 54%) as yellow crystals. The FTIR and NMR data were consistent with the reported literature.[16]. The same general procedure gave compounds **2b–c**, with the scale (mmol, mL [**1b–c**]) and yields given below.

(E)-3-(4-Dodecyloxyphenyl)-1-(4-hydroxyphenyl)prop-2-en-1-one (2b). **1b** (5.81 mL, 20 mmol). Yield: 8.32 g, 52%. The FTIR and NMR data were consistent with the reported literature.[16]

(E)-3-(4-Tetradecyloxyphenyl)-1-(4-hydroxyphenyl)prop-2-en-1-one (2c). **1c** (6.37 g, 20 mmol). Yield: 5.51 g, 54%. The FTIR and NMR data were consistent with the reported literature.[16]

Preparation of $N_3P_3Cl_5[OC_6H_4CH{=}CHC(O)C_6H_4OC_{10}H_{21}]$ (3a)

A mixture of hexachlorocyclotriphosphazenes (0.5 g, 1.44 mmol), **2a** (0.55 g, 1.44 mmol), and K_2CO_3 (2.87 g) in acetone (40 mL) was heated at reflux temperature for 1 h. The mixture was allowed to cool to room temperature and filtered. The filtrate was dried, filtered, and concentrated in vacuo. The crude solid was recrystallized from acetone to afford **3a** (0.18 g, 38%) as yellow solid. Mp 86–87°C; (Found: C, 43.25; H, 4.42; N, 6.07. $C_{25}H_{31}Cl_5N_3O_3P_3$ Requires C, 43.41; H, 4.52; N, 6.07%); ν_{max} (thin films/cm^{-1}) 1211, 1159, 1185 (P=N), 871 (P—O—C); δ_H (500 MHz, $CDCl_3$) 0.86 (3H, t, 1 × CH_3), 1.26–1.78 (16H, m, 8 × CH_2), 3.98 (2H, t, 1 × CH_2), 6.90 (2H, d, *J* 8.05, Ar—H), 7.34 (1H, d, *J* 15.45, 1 × olefinic H), 7.37 (2H, d, *J* 8.60, Ar—H), 7.57 (2H, d, *J* 8.05, Ar—H), 7.76 (1H, d, *J* 15.45, 1 × olefinic H), 8.04 (2H, d, *J* 8.05, Ar—H); δ_C (125.77 MHz, $CDCl_3$) 14.09, 22.64, 25.96, 29.10, 29.33, 29.43, 29.52, 29.52, 31.86, 68.20, 114.94, 118.84, 121.46, 127.07,

130.35, 130.44, 136.89, 145.50, 152.15, 152.23, 161.53, 188.88; δ_P (200 MHz, $CDCl_3$) 12.53 (t), 23.00 (d) *J* 60 Hz. The same general procedure gave compounds (**3b–c**) and (**4a–c**), with the scale (gram, mmol [**2b–c**]), reaction times, yields, and spectroscopy data given below.

Preparation of $N_3P_3Cl_5[OC_6H_4CH{=}CHC(O)C_6H_4OC_{12}H_{25}]$ (3b). **2b** (0.59 g, 1.44 mmol), 1 h. Yield: 0.28 g, 49%. Mp 77–78°C; (Found: C, 45.04; H, 4.21; N, 5.44. $C_{27}H_{35}Cl_5N_3O_3P_3$ Requires C, 45.06; H, 4.90; N, 5.84%); ν_{max} (thin films/cm^{-1}) 1212, 1159, 1185 (P=N), 872 (P—O—C); δ_H (500 MHz, $CDCl_3$) 0.86 (3H, t, 1 × CH_3), 1.25–1.79 (20H, m, 10CH_2), 3.99 (2H, t, 1 × CH_2), 6.09 (2H, d, *J* 8.60, Ar—H), 7.05 (2H, d, *J* 8.60, Ar—H), 7.34 (1H, d, *J* 15.45, 1 × olefinic H), 7.37 (2H, d, *J* 8.60, Ar—H), 7.56 (2H, d, *J* 8.55, Ar—H), 7.77 (1H, d, *J* 15.45, 1 × olefinic H); δ_C (125.77 MHz, $CDCl_3$) 13.90, 14.11, 22.67, 26.00, 29.12, 29.34, 29.52, 29.56, 29.56, 29.62, 29.62, 31.89, 114.79, 118.84, 121.45, 127.08, 130.36, 136.90, 145.51, 152.16, 152.42, 161.54, 188.90; δ_P (200 MHz, $CDCl_3$) 12.54 (t), 23.31 (d) *J* 60 Hz.

Preparation of $N_3P_3Cl_5[OC_6H_4CH{=}CHC(O)C_6H_4OC_{14}H_{29}]$ (3c). **2c** (0.59 g, 1.44 mmol), 1 h. Yield: 0.28 g, 50%. Mp 75–76°C; (Found: C, 45.96; H, 5.22; N, 5.55. $C_{29}H_{39}Cl_5N_3O_3P_3$ Requires C, 46.58; H, 5.26; N, 5.62%); ν_{max} (thin films/cm^{-1}) 1212, 1186, 1160 (P=N), 871 (P—O—C); δ_H (500 MHz, $CDCl_3$) 0.80 (3H, t, 1 × CH_3), 1.18–1.72 (24H, m, 12 × CH_2), 3.92 (2H, t, 1 × CH_2), 6.82 (2H, d, *J* 8.55, Ar—H), 7.27 (1H, d, *J* 15.45, 1 × olefinic H), 7.29 (2H, d, *J* 8.55, Ar—H), 7.49 (2H, d, *J* 8.60, Ar—H), 7.70 (1H, d, *J* 15.45, 1 × olefinic H), 8.00 (2H, d, *J* 8.05, Ar—H); δ_C (125.77 MHz, $CDCl_3$) 14.10, 22.66, 25.96, 29.11, 29.11, 29.34, 29.56, 29.56, 29.62, 29.62, 29.62, 30.95, 31.89, 68.20, 114.93, 118.83, 121.43, 121.47, 127.07, 130.35, 130.44, 136.89, 145.49, 152.14, 152.23, 161.53, 188.87; δ_P (200 MHz, $CDCl_3$) 12.53 (t), 23.30 (d) *J* 60 Hz.

Preparation of $N_3P_3[OC_6H_4CH{=}CHC(O)\text{-}C_6H_4OC_{10}H_{21}]_6$ (4a). **2a** (1.64 g, 4.31 mmol), 2 h. Yield: 1.53 g, 88%. Mp 126–127°C; (Found: C, 74.30; H, 7.58; N, 1.05. $C_{150}H_{186}N_3O_{18}P_3$ Requires C, 74.69; H, 7.76; N, 1.74%); ν_{max} (thin films/cm^{-1}) 3062 (C-H in aromatic), 1162 (P=N), 893 (P—O—C); δ_H (500 MHz, $CDCl_3$) 0.89 (3H, t, 1 × CH_3), 1.28–1.79 (16H, m, 8 × CH_2), 3.98 (2H, t, 1 × CH_2), 6.87 (2H, d, *J* 8.0, Ar—H), 7.05 (2H, d, *J* 8.0, Ar—H), 7.30 (1H, d, *J* 15.45, 1 × olefinic H), 7.49 (2H, d, *J* 8.60, Ar—H), 7.64 (1H, d, *J* 15.45, 1 × olefinic H), 7.86 (2H, d, *J* 8.55, Ar—H); δ_C (125.77 MHz, $CDCl_3$) 14.09, 22.65, 26.00, 29.16, 29.30, 29.38, 29.55, 29.55, 31.86, 68.15, 76.75, 114.75, 118.46, 120.98, 127.16, 130.19, 130.52, 135.82, 145.30, 153.21, 161.39; δ_P (200 MHz, $CDCl_3$) 9.03 (s, 3P, N_3P_3 ring).

Preparation of $N_3P_3[OC_6H_4CH{=}CHC(O)\text{-}C_6H_4OC_{12}H_{25}]_6$ (4b). **2b** (1.76 g, 4.31 mmol), 2 h. Yield: 1.31 g, 75%. Mp 122–123°C; (Found: C, 75.18; H, 8.23; N, 1.67. $C_{162}H_{210}N_3O_{18}P_3$ Requires C, 75.41; H, 8.20; N, 1.63%); ν_{max} (thin films/cm^{-1}) 3068 (C-H in aromatic), 1162 (P=N), 893 (P—O—C); δ_H (500 MHz, $CDCl_3$) 0.86 (3H, t, 1 × CH_3), 1.25–1.78 (20H, m, 10H_2), 3.95 (2H, t, 1 × CH_2), 6.85 (2H, d, *J* 8.60, Ar—H), 7.03 (2H, d, *J* 8.0, Ar—H), 7.29 (1H, d, *J* 15.45, 1 × olefinic H), 7.47 (2H, d, *J* 8.55, Ar—H), 7.62 (1H, d, *J* 15.45, 1 × olefinic H), 7.85 (2H, d, *J* 8.0, Ar—H); δ_C (125.77 MHz, $CDCl_3$) 14.07, 22.64, 25.98, 29.13, 29.30, 29.56, 29.59, 29.59, 30.84, 31.86, 67.94, 68.11, 114.72, 118.41, 120.97, 127.13, 130.16, 130.50, 135.79, 145.25, 153.18, 161.35, 188.67; δ_P (200 MHz, $CDCl_3$) 9.03 (s, 3P, N_3P_3 ring).

Preparation of $N_3P_3[OC_6H_4CH{=}CHC(O)\text{-}C_6H_4OC_{14}H_{29}]_6$ (4c). **2c** (1.88 g, 4.31 mmol), 2 h. Yield: 1.48 g, 78%. Mp 128–129°C; (Found: C, 74.53; H, 7.77; N, 1.63. $C_{162}H_{210}N_3O_{18}P_3$ Requires C, 75.41; H, 8.20; N, 1.63%); ν_{max} (thin films/cm^{-1}) 3068 (C-H in aromatic), 1162 (P=N), 887 (P—O—C); δ_H (500 MHz, $CDCl_3$) 0.80 (3H, t,

1 × CH_3), 1.19–1.72 (24H, m, 12 × CH_2), 3.90 (2H, t, 1 × CH_2), 6.79 (2H, d, *J* 7.4, Ar—H), 6.98 (2H, d, *J* 8.05, Ar—H), 7.22 (1H, d, *J* 15.45, 1 × olefinic H), 7.41 (2H, d, *J* 7.40, Ar—H), 7.56 (1H, d, *J* 15.45, 1 × olefinic H), 7.79 (2H, d, *J* 7.45, Ar—H); δ_C (125.77 MHz, $CDCl_3$) 14.09, 22.66, 26.01, 29.17, 29.34, 29.38, 29.59, 29.63, 29.65, 29.65, 29.65, 31.89, 68.15, 114.75, 118.47,120.98, 127.16, 130.52, 135.82, 145.29, 153.21, 161.39, 188.74; δ_P (200 MHz, $CDCl_3$) 9.03 (s, 3P, N_3P_3 ring).

REFERENCES

1. H. R. Allcock, *Phosphorus-Nitrogen Compounds* (Academic Press, New York, 1972).
2. G. Giavaresi, M. Tschon, V. Borsari, J. H. Daly, J. J. Liggat, M. Fini, V. Bonazzi, A. Nicolini, A. Carpi, M. Morra, C. Cassinelli, and R. Giardino, *Biomedicine and Pharmacotherapy*, **58,** 411 (2004).
3. C. W. Allen, *J. Fire Sci.*, **11,** 320 (1993).
4. G. A. Carriedo, F. J. Alonso, S. L. Vizcaíno, C. D. Valenzuela, and N. Yutronic, *Phosphorus, Sulfur, and Silicon*, **178,** 1549 (2003).
5. S. Sengupta, *S. Polyhedron*, **22,** 1237 (2003).
6. W. B. Stewart, S. J. Coles, D. B. Davies, M.B. Hursthouse, H. Ibisoglu, A. Kilic, R. A. Shaw, and I. Un, *Acta Crystallogr. B*, **62,** 321 (2006).
7. H. R. Allcock and C. Kim, *Macromolecules*, **22,** 2596 (1989).
8. H. R. Allcock and C. G. Cameron, *Macromolecules*, **27,** 3125 (1994).
9. H. R. Allcock and C. G. Cameron, *Macromolecules*, **27,** 3131 (1994).
10. J. Indira, P. P. Karat, and B. K. Sarojini, *J. Cryst. Growth*, **242,** 209 (2002).
11. B. T. Thaker, D. M. Patel, P. K. Tandel, M. S. Jesani, C. J. Vyas, and A. D. Vansadia, *Phase Transitions*, **78,** 521 (2005).
12. K. H. Jung, S. Y. Hyun, D. M. Song, and D. M. Shin, *Opt. Mater.*, **21,** 663 (2002).
13. T. Mihara, M. Tsutsumi, and N. Koide, *Mol. Cryst. Liq. Cryst.*, **412,** 247 (2004).
14. B. Zhou, W. Q Lu, Z. H. Zhou, and Y. Wu, *J. Mater. Chem.*, **10,** 1513 (2000).
15. D. Fichou, T. Watanabe, T. Takeda, S. Miyata, Y. Goto, and M. Nakayama, *Jap. J. Appl. Phys.*, **27,** 429 (1988).
16. N. K. Chudgar and S. N. Shah, *Liq. Cryst.*, **4,** 661 (1989).
17. A. N. Cammidge, S. Downing, and Z.Ngaini, *Tetrahedron Lett.*, **44,** 6633 (2003).
18. G. A. Carriedo, L. F. Catuxo, F. J. G. Alonso, P. G. Elipe, P. A. Gonzalez, and G. Sanchez, *J. Appl. Polym. Sci.*, **59,** 1879 (1996).
19. M. Kajiwara and H. Saito, *Angew. Makromol. Chem.*, **132,** 197 (1985).
20. K. Moriya, T. Masuda, T. Suzuki, S. Yano, and M. Kajiwara, *Mol. Cryst. Liq. Cryst.*, **318,** 267 (1998).
21. E. Cil, M. Arslan, and A. O. Gorgulu, *Polyhedron*, **25,** 3526 (2006).
22. A. Tarassoli and M. R. Shushizadeh, *Phosphorus, Sulfur, and Silicon*, **178,** 803 (2003).
23. K. H. Lee and D. C. Lee, *Polym. Bull.*, **42,** 543 (1999).