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Synthesis and Characterization of Oxime-bearing Hexakis(2-formylphenoxy)-phosphazene and its Derivatives

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ABSTRACT: *Hexakis(2-formylphenoxy)cyclotriphosphazene (**2**) was obtained from the reaction of hexachlorocylotriphosphazene (**1**) with 2-hydroxybenzaldehyde. Hexakis(2-[(hydroxyimino)methyl]-phenoxy)cyclotriphosphazene **(3)** was synthesized from the reaction of **2** with hydroxlaminehydrochloride in pyridine. Hexasubstituted compounds **4, 5, 6, 8, 9**, and **10** were obtained from the reactions of **3** with methyl iodide, ethyl bromide, allyl bromide, propanoyl chloride, benzoyl chloride, and 4-methoxybenzoyl chloride, respectively. Disubstituted product **7** was obtained from the reaction of **3** with chloroacetyl chloride. Pure and defined products could not be obtained from the reaction of **3** with acetyl chloride, benzyl chloride, and 2-chlorobenzoyl chloride. The compounds were characterized by elemental analysis and IR, ^{1}H, ^{13}C, and ^{31}P NMR spectroscopy.*
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INTRODUCTION

Phosphazenes, which are the best known and most intensively studied phosphorusnitrogen compounds [1–5], possess a number of characteristics such as biomedical properties and applications due to their strong antitumor activity [6–11]. Their antimicrobial and biological activities on bacterial and yeast cells have been studied [12–14]. Some applications include flame-retardant materials [15], model compounds for polyphosphazenes, starting materials for the preparation of cyclolinear and/or cyclomatrix phosphazene substrates, commercial polymers with carbon backbone-containing pendant cyclophosphazene groups, inorganic hydraulic fluids and lubricants, biologically important substrates such as anticancer agents, insect chemosterilants, pesticides and fertilizers, supports for catalysts, dyes, and crown ether phase transfer catalysts for nucleophilic substitution reactions, core substrates for dendrimers, thermal initiators for anionic polymerization reactions, and photosensitive materials [16].

Syntheses of different linear, cyclic, or polyphosphazenes have been reported [17–29]. There are also a large number of literature reports on reactions of the functional groups on phosphazene substituents [10,30]. Typical of these include coupling reactions of trimeric phosphazene azides with aryloxy, alkoxy, and dialkylamino cosubstituents [31], *N*-vinylic phosphazenes with azodicarboxylic and acetylenic esters [32], oxime-phosphazene derivatives with alkyl and acyl substituents [33,34], polymers from 4-formylphenoxy [35,36], maleic [37], and 3,4-methylenedioxyphenoxy substituents [38], and oxime-bearing hexakis(4-formylphenoxy)phosphazene and derivatives [33,34,39].

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In this paper, we report on the preparation of oxime-cyclophosphazene from hexakis(2-formylphenoxy)cyclotriphosphazene, and studies on its reactions with methyl iodide, ethyl bromide, allyl bromide, propanoyl chloride, benzoyl chloride, 4-methoxybenzoyl chloride, chloroacetyl chloride, acetyl chloride, benzyl chloride, and 2-chlorobenzoyl chloride. We hope that this original work is potentially a useful addition to the literature and can guide to some high-polymer works.

RESULTS AND DISCUSSION

The reaction of **1** with 6 equiv. of 2-hydroxybenzaldehyde in the presence of K_2CO_3 in THF gave hexakis(2-formylphenoxy)cyclotriphosphazene (**2**). Oxime compound hexakis(2-[(hydroxyimino)methyl]phenoxy)cyclotriphosphazene (**3**) was synthesized from the reaction of **2** with hydroxlaminehydrochloride in pyridine.

Hexasubstituted compounds were obtained from the reactions of **3** with methyl iodide, ethyl bromide, allyl bromide, propanoyl chloride, benzoyl chloride, and 4-methoxybenzoyl chloride in acetone in the presence of K_2CO_3 via replacement of all the oxime protons with alkyl and acyl groups. Disubstituted product was obtained from the reaction of **3** with chloroacetyl chloride. Pure and defined products could not be obtained from the reaction of **3** with acetyl chloride, benzyl chloride, and 2-chlorobenzoyl chloride.

The compounds were characterized by elemental analysis and IR, ^{1}H, ^{13}C, and ^{31}P NMR spectroscopy (structures **2–10** are shown in Scheme 1). Physical properties and analytical data for **2–10** are given in Table 1. Compounds **2–10** were synthesized in high yields except **5** and **9**. The solvents used for the purification of compounds **2, 4, 5, 6**, and **10** could not be removed completely. Thus, the presence of these trace amounts of solvent affect the elemental analysis, in particular the carbon value.

The characteristic stretching peaks in the IR spectra of the phosphazenes have been assigned as in Table 2. The P=N stretching vibrations, which are observed between 1170 and 1200 cm^{-1}, are characteristic of cyclophosphazenes. These peaks are shifted to longer wavelengths for **2–10** than in **1**, which appeared at 1218 cm^{-1}. The OH stretching vibrations in the IR spectra of **3** and **7** indicate the oxime compounds. While **3** is the initial oxime, all hydrogen atoms of the OH groups of **7** could not be replaced by the propanoyl substituent.

NMR data for **2–10** are given in Tables 3–5. The ^{31}P NMR shifts of **2–10** change between 7.77 and 8.63 ppm. Although there is only one peak in the ^{31}P NMR spectra of **1** and **2** at 21.12 and 8.21 ppm, respectively, two peaks, with very weak second signals, are observed at $\delta = 8.63$, 8.59, $\delta = 8.05$, 8.13, $\delta = 8.16$, 9.10, $\delta = 8.31$, 8.61, $\delta = 8.41$, 8.05, $\delta = 8.15$, 8.06, $\delta = 7.91$, 7.77 and $\delta = 8.01$, 7.92 ppm for **3, 4, 5, 6, 7, 8, 9**, and **10**, respectively. It is assumed that the weak peaks are due to the *cyn*- and *anti*-isomers of the —C=N— groups. The effects of the *cyn*- and *anti*-isomerism are also observed in the ^{13}C NMR spectra of **3–10** except **6.** These data demonstrate that compounds **3–10** consist of a mixture of two isomers. Although there are different phosphorus environments in the molecule of **7**, the main peak is observed as a singlet. It is understood that the phosphorus peaks are not affected by these changes because the substituted groups are far off the phosphorus atoms.

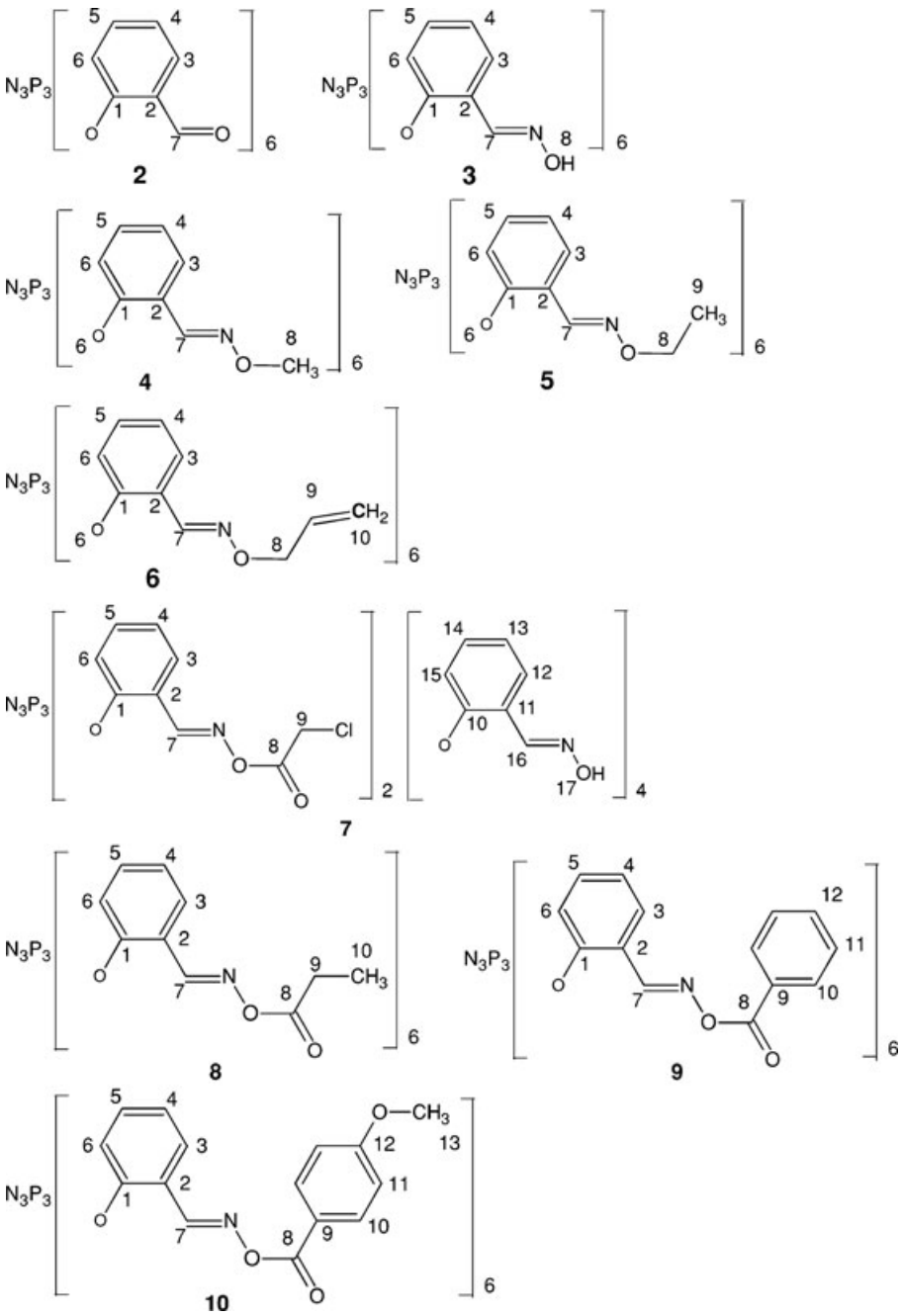
In addition, ^{1}H and ^{13}C NMR data confirm the structures of **2–10** (Scheme 1). In the ^{1}H NMR spectra (Table 4), the OH protons are observed at 11.51 (11.43 for isomer) and 11.50 ppm for **3** and **7**, respectively. It is understood from the integral intensities that there are six OH protons in **3**, which is the original oxime-phosphazene, and four OH protons in **7**. This observation for **7** indicates that propanoyl group has not replaced all OH protons in **3**. The aldehyde proton for **2** appears at 9.96 ppm. The azomethine protons for **3, 4, 5, 6, 7, 8, 9**, and **10** are observed at 8.10 (H^7), 8.45 (H^7), 8.50 (H^7), 8.21 (H^7), 8.20–8.08 (H^7, H^{16}), 8.39 (H^7), 8.57 (H^7), and 8.70 (H^7) ppm, respectively. The azomethine protons are shifted to the lower down field region for compounds **4–10** than for **3**. The aromatic protons for all the compounds appear between 6.80 and 8.30 ppm.

The detailed ^{13}C NMR spectral data are given in Table 5. Aldehyde carbon atom for **2** is observed at 192.21 ppm at the lowest down field region of the carbon atoms. The azomethine carbon atoms are shifted to the lower down field region for **5, 8–10** than for **3** and in the substituted moiety of the molecule for **7** except **4** and **6** in which the benzyl group releases an electron to the molecule.

EXPERIMENTAL

General Remarks

Solvents and other liquids used in the experimental works were dried by conventional methods. Hexachlorocyclotriphosphazene $[N_3P_3Cl_6]$ (**1**) was recrystallized from hexane. Other chemicals were used as purchased. Hexakis(2-formylphenoxy)cyclotriphosphazene (**2**) was prepared as described by Carriedo et al. [25]. The reactions of $[N_3P_3Cl_6]$ with phenols were carried out under dry nitrogen.



SCHEME 1 Structures of compounds **2–10**.

IR spectra were recorded on an ATI Unicam Mattson 1000 FTIR spectrometer. ^{1}H, ^{13}C, and ^{31}P NMR spectra were recorded using a Bruker DPX-300 spectrometer operating at 300.13, 75.46, and 121.49 MHz, respectively. ^{1}H and ^{13}C NMR chemical shifts were measured using $SiMe_4$ as an internal standard, whereas those for ^{31}P were measured using 85% H_3PO_4 as an external standard. Chemical shifts downfield from the standard were assigned positive δ values. Microanalysis was carried out by a LECO 932 CHNS-O apparatus.

Synthesis of Compound 2. A mixture of **1** (8.27 g, 23.79 mmol), 2-hydroxybenzaldehyde (17.64 g, 144.44 mmol), and K_2CO_3 (40.00 g, 289.40 mmol) was stirred in THF (250 mL) at 0°C

TABLE 1 Physical Properties and Analytical Data for **2–10**

Compound	*Yield (%)*	*Atom*	*Found (%)*	*Calculated (%)*
2	76	C	56.57	58.55
		H	3.77	3.51
		N	3.85	4.88
3	91	C	52.05	53.00
		H	4.23	3.81
		N	11.87	13.25
4	66	C	52.66	55.66
		H	5.01	4.67
		N	9.21	12.17
5	45	C	55.61	57.95
		H	5.26	5.40
		N	9.99	11.26
6	72	C	59.28	60.45
		H	5.16	5.07
		N	9.20	10.57
7	81	C	49.81	50.01
		H	3.77	3.47
		N	10.59	11.41
8	75	C	55.42	55.75
		H	4.81	4.70
		N	7.55	9.79
9	59	C	64.66	64.00
		H	4.14	3.84
		N	6.90	8.00
10	81	C	59.99	61.54
		H	4.36	4.13
		N	6.26	7.18

and then was reacted at ambient temperature for 72 h. The solvent was removed under vacuum. The residue was extracted with CH_2Cl_2 (3 × 75 mL). After the solvent was removed, a white solid (**2**) formed in 76% (15.60 g) yield.

Synthesis of Compound 3. A mixture of **2** (12.00 g, 13.92 mmol) and hydroxylaminehydrochloride (6.00 g, 84.34 mmol) was refluxed in pyridine (30 mL) for 3 h. After the reaction was complete, the mixture was slowly poured into cold water. The precipitate was filtered and washed twice with cold water. It was resolved in hot acetone and reprecipitated from cold water three times. The white solid (**3**) was washed with alcohol and dried at 60°C under vacuum. Yield: 91% (12.00 g).

TABLE 3 ^{31}P NMR Data for **1–10**

	Major Isomer		*Minor Isomer*	
Compound	*ppm*	*Relative Intensity (%)*	*ppm*	*Relative Intensity (%)*
1	21.12	–	–	–
2	8.21	–	–	–
3	8.63	88	8.59	12
4	8.05	56	8.13	44
5	8.16	75	9.10	25
6	8.31	83	8.61	17
7	8.41	79	8.05	21
8	8.15	81	8.06	19
9	7.91	88	7.77	12
10	8.01	75	7.92	25

Reaction of 3 with methyl iodide. A solution of methyl iodide (0.40 mL, 0.91 g, 6.42 mmol) in acetone (10 mL) was slowly added dropwise to a stirred and cooled (0°C) mixture of **3** (0.80 g, 0.84 mmol) and K_2CO_3 (2.00 g, 14.47 mmol) in acetone (30 mL). The reaction was carried out at room temperature for 12 h and then refluxed for 24 h. After the reaction was complete, the resultant precipitate was filtered off and the solvent was removed. An oily product was obtained. It was dissolved in a very little quantity of acetone alcohol and precipitated with hexane. Although the solvent was tried to remove under vacuum at 60°C for 48 h, it could not be removed completely. A very viscous product (**4**) was obtained. Yield: 66% (0.57 g).

Reaction of 3 with ethyl bromide. A solution of ethyl bromide (0.50 mL, 0.74 g, 6.79 mmol) in acetone (10 mL), **3** (0.80 g, 0.84 mmol), and K_2CO_3 (2.00 g, 14.47 mmol) in acetone (30 mL) was used

TABLE 2 Characteristic IR Vibrations (in cm^{-1}) for **2–10**

Compound	ν_{QH}	$\nu_{C-H\ ar.}$	$\nu_{C-H\ al.}$	$\nu_{C=O}$	$\nu_{P=N}$	ν_{N-O-C}	ν_{P-O-C}
2	–	3035, 3101	2760, 2885	1696	1182	–	961
3	3304	3005, 3077	2891, 2927	–	1176	–	955
4	–	3040, 3076	2897, 2939	–	1174	1097	960
5	–	3062, 3107	2883, 2979	–	1175	1059	958
6	–	3011, 3083	2909, 2939	–	1170	1038	949
7	3394	3035, 3077	2856, 2930	1702	1200	1056	984
8	–	2981, 3041	2862, 2903	1774	1170	1062	955
9	–	3041, 3071	2867, 2933	1702	1170	1038	943
10	–	3010, 3070	2843, 2927	1741	1174	1067	936

TABLE 4 ^{1}H NMR Data for **2–10**

Compound	*^{1}H NMR*
2	7.05–7.74 (m): H^{Ar}, 9.96 (s): H^{7}
3	11.51 (s): H^{8}, 11.43 (s): H^{8} (is), 8.10 (s): H^{7}, 8.35 (s): H^{7} (is), 7.79 (d): H^{3}, 7.00–7.50 (m): H^{5}, H^{4}, 6.90 (d): H^{6}
4	3.90 (s): H^{8}, 3.84 (s): H^{8} (is), 6.80–8.20 (m): H^{Ar}, 8.45 (s): H^{7}
5	8.50 (s): H^{7}, 8.75 (s): H^{7} (is), 6.80–8.30 (m): H^{Ar}, 3.99–4.30 (m): H^{8}, H^{8} (is), 1.15–1.48 (m): H^{9}, H^{9} (is)
6	8.21 (s): H^{7}, 7.72 (d): H^{3}, 7.22 (m): H^{4}, H^{5}, 6.97 (d): H^{6}, 6.00 (m): H^{9}, 5.28 (d): H^{10}_{trans}, 5.18 (d): H^{10}_{cis}, 4.77 (d): H^{8}
7	11.50 (s): H^{17}, 8.20 (s): H^{7}, 8.08 (s): H^{16}, 6.80–7.90: H^{3}, H^{4}, H^{5}, H^{6}, H^{12}, H^{13}, H^{14}, H^{15}, 2.1:H^{9}
8	8.39 (s): H^{7}, 8.05 (d): H^{3}, 6.80–7.40: H^{3}, H^{4}, H^{5}, H^{6}
9	8.57: H^{7}, 7.00–8.30: aromatic (H^{3}, H^{4}, H^{5}, H^{6}, H^{11}, H^{12}, H^{13}, H^{14})
10	8.70(s): H^{7}, 6.70–8.60: H^{Ar}, 3.84 (s): H^{13}, 3.90: H^{13} (is)

For numbering see Scheme 1. $CDCl_3$-*d* (for **4, 5**, and **10**) and DMSO-*d* (for **2, 3, 6–9**) were used as solvents in NMR analyses. Isomer (is), singlet (s), doublet (d), and multiple! (m).

TABLE 5 ^{13}C NMR Data for **2–10**

Compound	*^{13}C NMR*
2	192.21: C^{7}, 161.37: C^{1}, 117.00–137.00: C^{2}—C^{6}
3	156.49: C^{1}, 148.18: C^{7} (is), 147.80: C^{7}, 144.97: C^{3}, 144.00: C^{2} (is), 142.86: C^{2}, 130.95: C^{5}, 130.39: C^{5} (is), 126.48: C^{4}, 125.83: C^{4} (is), 121.19: C^{6} (is), 120.89: C^{6}
4	148.36: C^{1}, 145.55: C^{7}, 144.83: C^{3} (is), 144.52: C^{3}, 143.54: C^{2}, 143.50: C^{2} (is), 131.08: C^{5} (is), 130.78: C^{5}, 126.55: C^{4}, 125.43: C^{4} (is), 120.76: C^{6}
5	156.93: C^{1}, 148.40: C^{7}, 148.42: C^{7} (is), 144.63: C^{3}, 144.19: C^{3} (is), 143.43: C^{2} (is), 143.28: C^{2}, 130.92: C^{5} (is), 130.66: C^{5}, 126.52: C^{4}, 126.22: C^{4} (is), 120.80: C^{6} (is), 120.60: C^{6}, 63.93: C^{8} (is), 69.67: C^{8}, 14.64: C^{9} (is), 14.56: C^{9}
6	148.04: C^{1}, 143.42: C^{7}, 134.60: C^{9}, 131.59: C^{3}, 126.97: C^{2}, 126.43: C^{5}, 124.15: C^{4}, 121.27: C^{6}, 118.02: C^{10}, 74.91: C^{8}
7	172.49: C^{8} (is), 168.25: C^{8}, 150.73: C^{7}, 148.91: C^{1}, 147.66: C^{10}, 142.55: C^{16}, 135.28: C^{3}, 134.86: C^{12}, 133.68: C^{2}, 132.28: C^{11},127.88: C^{5}, 126.93: C^{14}, 125.26: C^{4}, 122,55: C^{13}, 121.41: C^{6}, 120.89: C^{15},19.59: C^{9}, 21.52: C^{9} (is)
8	171.50: C^{8}, 171.71: C^{8} (is), 150.64: C^{7}, 148.80: C^{1}, 136.40: C^{3} (is), 133.64: C^{3}, 128.04: C^{2}, 127.74: C^{2} (is), 126.53: C^{5}, 122.65: C^{4}, 121.97: C^{6} (is), 121.43: C^{6}, 26: C^{9} (is), 25.62: C^{9}, 9.29: C^{10} (is), 9.00: C^{10}
9	168.78 : C^{8}, 163.13: C^{7} (is), 163.05 : C^{7}, 152.50 : C^{1} (is), 152.19: C^{1}, 134.20 : C^{12}, 133.74 : C^{3}, 132.80: C^{3} (is), 129.60: C^{9}, 129.16: C^{10}, 128.17: C^{11}, 128.10: C^{2}, 127.26: C^{5}, 122.17: C^{4}, 121.57: C^{6}
10	164.48: C^{8} (is), 164.33: C^{8}, 163.82: C^{7}, 163.76: C^{7} (is), 163.61: C^{12}, 163.39: C^{12} (is), 151.79: C^{1}, 150.28: C^{1} (is), 132.90: C^{3} (is), 132.67: C^{3}, 131.89: C^{10}, 131.82: C^{10} (is), 128.25: C^{2} (is), 128.06: C^{2}, 126.33: C^{5}, 126.22: C^{5} (is), 123.21: C^{4}, 123.15: C^{4} (is), 120.87: C^{6}, 120.55: C^{6} (is), 114.11: C^{9}, 113.82: C^{11}, 113.48: C^{11} (is), 55.48: C^{13}

For numbering see Scheme 1. $CDCl_3$-*d* (for **4, 5**, and **10**) and DMSO-*d* (for **2, 3, 6–9**) were used as solvents in NMR analyses. Isomer (is), singlet (s), doublet (d), and multiplet (m).

for the preparation of **5**, as in **4**. The reaction was carried out at room temperature for 1 h and then refluxed for 24 h. Compound **5** was obtained as an oily product. It was dissolved in a very little quantity of acetone alcohol and precipitated with hexane. The brown color solid (**5**) was dried at 60°C under vacuum for 48 h. Yield: 45% (0.42 g).

Reaction of 3 with allyl bromide. A solution of allyl bromide (0.50 mL, 0.65 g, 5.77 mmol) in acetone (10 mL), **3** (0.80 g, 0.84 mmol), and K_2CO_3 (1.60 g, 11.57 mmol) in acetone (30 mL) was used for the preparation of **6**, as in **4**. The reaction was carried out at room temperature for 8 h and then refluxed for 48 h. Compound **6** was obtained as an oily product. It was dissolved in a very little quantity of alcohol and precipitated with hexane. The white solid (**6**) was dried at 60°C under vacuum for 48 h. Yield: 72% (0. 72 g).

Reaction of 3 with chloroacetyl chloride. A solution of chloroacetyl chloride (0.46 mL, 0.70 g, 5.77 mmol) in acetone (10 mL), **3** (0.80 g, 0.84 mmol), and K_2CO_3 (1.60 g, 11.57 mmol) in acetone (30 mL) was

used for the preparation of **7**, as in **4**. The reaction was carried out at room temperature for 8 h and then refluxed for 12 h. Compound **7** was obtained as an oily product. It was dissolved in a very little quantity of alcohol and precipitated with hexane. The white solid (**7**) was dried at 60°C under vacuum for 48 h. Yield: 81% (0.7 6 g).

Reaction of 3 with propanoyl chloride. A solution of propanoyl chloride (0.40 mL, 0.42 g, 4.58 mmol) in acetone (10 mL), **3** (0.60 g, 0.63 mmol), and K_2CO_3 (1.60 g, 11.57 mmol) in acetone (30 mL) was used for the preparation of **8** as for **4**. The reaction was carried out at room temperature for 8 h and then refluxed for 24 h. Compound **8** was obtained as an oily product. It was dissolved in a very little quantity of alcohol and precipitated with hexane. The brown color solid (**8**) was dried at 60°C under vacuum for 48 h. Yield: 75% (0.60 g).

Reaction of 3 with benzoyl chloride. A solution of benzoyl chloride (0.55 mL, 0.66 g, 4.73 mmol) in acetone (10 mL), **3** (0.60 g, 0.63 mmol) in acetone (30 mL), and K_2CO_3 (1.60 g, 11.57 mmol) was used for the preparation of **9** as for **4**. The reaction was carried out at room temperature for 12 h and then refluxed for 72 h. Compound **9** was obtained as an oily product. It was dissolved in a very little quantity of alcohol and precipitated with hexane. The brown solid (**9**) was dried at 60°C under vacuum for 48 h. Yield: 59% (0.55 g).

Reaction of 3 with 4-methoxybenzoyl chloride. A solution of 0.70 g (4.10 mmol) 4-methoxybenzoyl chloride in acetone (10 mL), **3** (0.60 g, 0.63 mmol), and K_2CO_3 (2.00 g, 14.47 mmol) in acetone (30 mL) was used for the preparation of **10** as for **4**. The reaction was carried out at room temperature for 24 h. After the reaction was complete, the mixture was slowly poured into water. The yellow solid (**10**) was washed with hot alcohol and acetone and dried at 60°C under vacuum for 48 h. Yield: 81% (0.90 g).

REFERENCES

[1] Allcock, H. R. Phosphorus-Nitrogen Compounds; Academic Press: New York, 1972.
[2] Diaz, C.; Izquierdo, I.; Mendizabal, F.; Yutronic, N. Inorg Chim Acta 1999, 294, 20.
[3] Kılıç, A.; Begeç, S.; Çetinkaya, B.; Kılıç, Z.; Gündüz, N.; Yıldız, M.; Hökelek, T. Heteroat Chem 1996, 7, 44, 249.
[4] Carriedo, G. A.; Alonso, F. J. G.; Elipe, P. G.; Brillas, E.; Julia, L. Org Lett 2001, 3, 11, 1625.
[5] Allcock, H. R.; Morrissey, C. T.; Way, W. K.; Winograd, N. Chem Mater 1996, 8, 2730.
[6] Allcock, H. R. In: Inorganic and Organometallic Polymers; Allcock, H. R.; Zeldin, M.; Wynne, K. J. (Eds.); ACS Symposium Series: Washington, DC, 1988; Vol. 360, Ch. 19.
[7] Laurencin, C. T.; Koh, H. J.; Neenan, T. X.; Allcock, H. R.; Langer, R. J Biomed Mater Res 1987, 21, 1231.
[8] Allcock, H. R.; Pucher, S. R.; Fitzpatrick, R. J.; Rashid, K. Biomaterials 1992, 13, 857.
[9] Giavaresi, G.; Tschon, M.; Borsari, V.; Daly, J. H.; Liggat, J. J.; Fini, M.; Bonazzi, V.; Nicolini, A.; Carpi, A.; Morra, M.; Cassinelli, C.; Giardino, R. Biomed Pharmacother 2004, 58, 411.
[10] Jun, Y. J.; Kim, J. I.; Jun, M. J.; Sohn, Y. S. J Inorg Biochem 2005, 99, 1593.
[11] Brandt, K.; Kruszynski, R.; Bartczak, T. J.; Czomperlik, I. P. Inorg Chim Acta 2001, 322, 138.
[12] Ozturk, A. I.; Yılmaz, O.; Kırbag, S.; Arslan, M. Cell Biochem Funct 2000, 18, 117.
[13] Konar, V.; Yılmaz, O.; Ozturk, A. I.; Kırbag, S.; Arslan, M. Bioorg Chem 2000, 28, 214.
[14] Yılmaz, O.; Aslan, F.; Ozturk, A. I.; Vanlı, N. S.; Kırbag, S.; Arslan, M. Bioorg Chem 2002, 30, 5, 303.
[15] Allen C. W. J Fire Sci 1993, 11, 320.
[16] Gleria, M.; Jaeger, R. D. J Inorg Organomet Polym 2001, 11, 1.
[17] Laguna, M. T. R.; Tarazona, M. P.; Carriedo G. A.; Alonso, F. J. G.; Fidalgo, J. I.; Saiz, E. Macromolecules 2002, 35, 7505.
[18] Cil, E.; Arslan, M.; Aslan, F.; Ozturk A. I. Phosphorus Sulphur Silicon 2003, 178, 5, 1037.
[19] Ozturk, A. I.; Aslan, F.; Cil, E.; Arslan, M., Kılıç, A. Phosphorus Sulphur Silicon 2003, 178, 10, 2097.
[20] Arslan, M.; Aslan, F.; Ozturk A. I. Heteroat Chem 2003, 14, 2, 138.
[21] Aslan, F.; Ozturk, A. I.; Arslan, M. Heteroat Chem 2003, 14, 5, 413.
[22] Diaz, C. V.; Izquierdo, I. G. Polyhedron 1999, 18, 1479.
[23] Bertani, R.; Facchin, G.; Gleria, M. Inorg Chim Acta 1989, 165, 73.
[24] Kılıç, A.; Begeç, S.; Kılıç, Z.; Hokelek, T. J Mol Struct 2000, 516, 255.
[25] Carriedo, G. A.; Catuxo, L. F.; Alonso, F. J. G.; Elipe, P. G.; Gonzalez, P. A.; Sanchez, G. J Appl Polym Sci 1996, 59, 1879.
[26] Allcock, H. R.; Turner, M. L. Macromolecules 1993, 26, 3.
[27] Carriedo, G. A.; Catuxo, L. F.; Alonso, F. J. G.; Elipe, P. G.; Gonzalez, P. A. Macromolacules 1996, 29, 16, 5320.
[28] Bilge, S.; Kılıç, Z.; Çaylak, N.; Hökelek, T. J Mol Struct 2004, 707, 139.
[29] Begeç, S.; Kılıç, A. Heteroat Chem 2005, 16, 308.
[30] Belluco, U.; Bertani, R.; Michelin, R. A.; Mozzan, M.; Zingales, F.; Gleria, M. Inorg Chim Acta, 1995, 229, 13.
[31] McIntosh, M. B.; Hartle, T. J.; Allcock, H. R. J Am Chem Soc 1999, 121, 884.
[32] Palacios, F.; Alonso, C.; Rubialesa, G. G.; Ezpeleta, J. M. Tetrahedron 2004, 60, 2469.
[33] Çil, E.; Arslan, M.; Görgülü, A. O. Heteroat Chem 2006, 17, 112.
[34] Çil, E.; Arslan, M.; Görgülü, A. O. Can J Chem 2005, 83, 2039.
[35] Yuan, W.; Zhu, L.; Huang, X.; Zheng, S.; Tang, X. Eur Polym J 2005, 41, 1867.

[36] Yuan, W.; Tang, X.; Huang, X.; Zheng, S. Polymer, 2005, 46, 1701.
[37] Fontana, G.; Minto, F.; Gleria, M.; Facchin, G.; Bertani, R.; Favero, G. Eur Polym J 1996, 32, 1273.
[38] Muraki, T.; Ueta, M.; Ihara, E.; Inoue, K. Polym Degrad Stabil 2004, 84, 87.
[39] Çil, E.; Arslan, M.; Görgülü, A. O. Polyhedron 2006, 25, 3526.