ISSN 1070-3632, Russian Journal of General Chemistry, 2007, Vol. 77, No. 12, pp. 2117–2122. © Pleiades Publishing, Ltd., 2007.
Original Russian Text © M. Yildiz, S. Yilmaz, B. Dölger, 2007, published in Zhurnal Obshchei Khimii, 2007, Vol. 77, No. 12, pp. 1972–1977.

Synthesis, Spectral Properties, and Antimicrobial Activity of 2-Arilamino-2,4,4,6,6-Pentachloro-1,3,5,2λ^5,4λ^5,6λ^5-Triazatriphosphines and Poly[Bis(4-Fluorophenylamino)Phosphazene]

M. Yildiz[a], S. Yilmaz[a], and B. Dölger[b]

[a] *Department of Chemistry, Faculty of Arts and Sciences, Çanakkale Onsekiz Mart University, 17100 Çanakkale, Turkey*
fax: +(90286)218-05-33
e-mail: myildiz@comu.edu.tr

[b] *Department of Biology, Faculty of Arts and Sciences, Çanakkale Onsekiz Mart University, 17100 Çanakkale, Turkey*

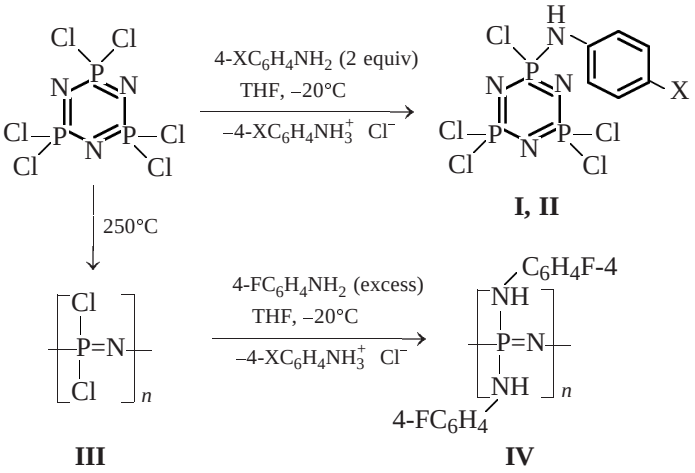
Received November 8, 2006

Abstract — 2-(4-Chloro and 4-fluorophenylamino)-2,4,4,6,6-pentachloro-1,3,5,2λ^5,4λ^5,6λ^5-triazatriphosphinines and poly[bis(4-fluorophenylamino)phosphazene] were synthesized by reactions of 4-fluoroaniline and 4-chloroaniline with 2,2,4,4,6,6-hexachloro-1,3,5,2λ^5,4λ^5,6λ^5-triazatriphosphinine and poly(dichlorophosphazene), respectively, in tetrahydrofuran under argon at –20°C, followed by heating under reflux. The products were isolated by column chromatography and were characterized by FTIR, NMR (^{1}H, ^{13}C, ^{31}P), and mass spectra, termogravimetry, and high-performance liquid chromatography. Antimicrobial activity of the monomeric compounds and polymer against 9 bacteria and 5 yeast cultures was evaluated by the disk diffusion method in dimethyl sulfoxide relative to a number of commercial antibiotics and antifungal agents. Aminophosphazene derivatives exhibited a broad spectrum of activity against both Gram-positive and Gram-negative bacterial with a magnitude comparable to reference antimicrobial agents. The polymeric product turned out to be more potent than the monomeric compounds.
DOI: 10.1134/S1070363207120079

During the past decade, reactions of hexachlorocyclotriphosphazene $N_3P_3Cl_6$ with primary and secondary amines, diamines, polyamines, hydroxylamine [1–6], phenols [7–9], alcohols, and oligo(ethylene glycols) [10] have been studied under different conditions with the goal of obtaining macrocyclic cyclophosphazene ethers [11]. Polyphosphazenes constitute a large class of inorganic polymers whose backbone incorporates alternating phosphorus and nitrogen atoms. Versatile reactivity of the precursor, poly(dichlorophosphazene) $(NPCl_2)_x$, makes it possible to introduce various side organic groups into the polymeric chain via nucleophilic replacement of chlorine on the phosphorus [12–14]. As a result, polymeric products possessing practically important properties could be obtained, e.g., polymeric electrolytes [14, 15], compounds with unusual thermal propeties [16, 17], biocompatible substances [18], and compounds having interesting optical properties [19, 20]; some of these derivatives attract interest from the commercial viewpoint. Therefore, phosphazenes have received increasing attention. On the other hand, polyanilines have been studied extensively for numerous potential applications due to their high conductivity, electroactivity, and other unique properties [21–23].

Here we report on the synthesis of 2,4,4,6,6-pentachloro-2-(4-fluorophenylamino)-1,3,5,2λ^5,4λ^5,6λ^5-triazatriphosphinine (**I**), 2,4,4,6,6-pentachloro-2-(4-chlorophenylamino)-1,3,5,2λ^5,4λ^5,6λ^5-triazatriphosphinine (**II**), and poly[bis(4-fluorophenylamino)phosphazene] (**IV**) and on their antimicrobial activity in vitro. The structure of the monomeric and polymeric products was determined by elemental analysis, FTIR spectroscopy, ^{1}H, ^{13}C, and ^{31}P NMR, thermogravimetric analysis, and high-performance liquid chromatography (HPLC).

The IR spectra of compounds **I**, **II**, and **IV** contained absorption bands belonging to vibrations of N–H bonds at 3180, 3161 and 3366 (m), C=C bonds at 1509, 1495, and 1511 (s), C–N bonds at 1377, 1376, and 1382 (m), and P=N bonds at 1196, 1189, and 1211 cm^{-1} (s), respectively (Fig. 1). Unlike polymeric



I, II **III** **IV**

I, X = F; **II**, X = Cl.

phosphazene **IV**, compounds **I** and **II** showed ν(P–Cl) bands at 592, 523 and 598, 526 cm^{-1}, respectively. The position of the P=N absorption band in the IR spectrum of initial hexachlorocyclotriphosphazene (1215 cm^{-1}) is similar to the position of the corresponding band in the spectra of octachlorocyclotetraphosphazene (1310 cm^{-1}), decachlorocyclopentaphosphazene (1355 cm^{-1}), dodecachlorocyclohexaphosphazene (1325 cm^{-1}), tetradecachlorocycloheptaphosphazene (1310 cm^{-1}), and higher cyclic homologs and

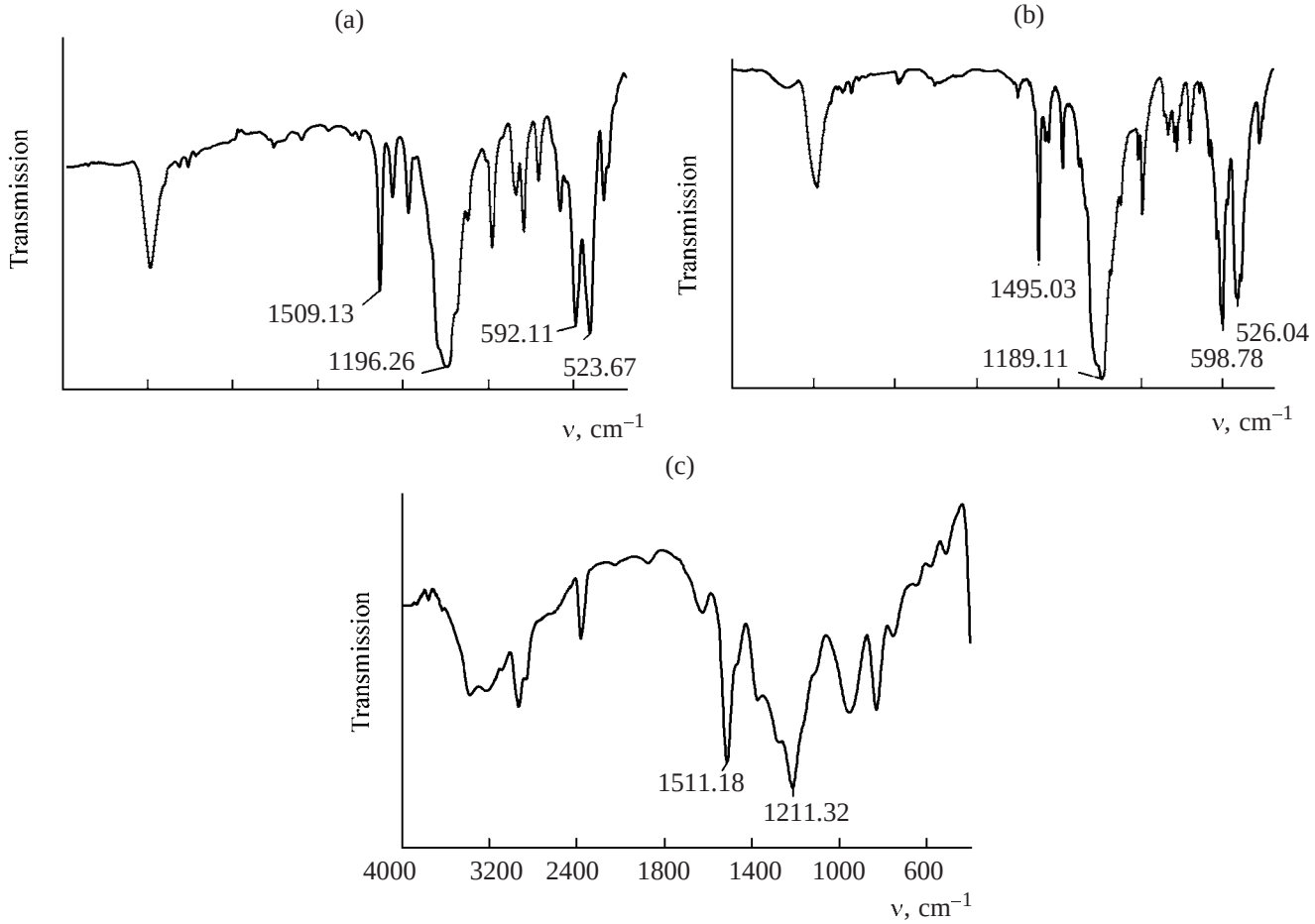


Fig. 1. IR spectra of compounds (a) **I**, (b) **II**, and (c) **IV**.

polymer (1305 cm^{-1}), as well as of acyclic monophosphazenes (1160–1230 cm^{-1}) [24]. The P=N stretching vibration frequencey in the IR spectrum of monomer **I** is lower by 15 cm^{-1} than that observed for polymer **IV**. The presence of chlorine atoms increases the intensity of the ν(P=N) band in the spectrum of poly(dichlorophosphazene) (**III**) as compared to poly[bis(4-fluorophenylamino)phosphazene] (**IV**) containing electron-withdrawing 4-fluorophenylamino groups.

The ^1H and ^{13}C NMR spectra of the obtained compounds are given in Experimental. The NH proton signal appears as a singlet at δ 5.52 ppm, doublet at δ 5.59 ppm ($^2J_{PH}$ = 9.0 Hz), or doublet at δ 5.40 ppm ($^2J_{PH}$ = 20 Hz) in the ^1H NMR spectra of **I**, **II**, and **IV**, respectively. Aromatic protons resonated at δ, ppm: 6.93, 7.09 m (**I**); 7.18, 7.38 d.d (**II**); 7.07, 7.19 m (**IV**). The ^{13}C NMR spectra of **I** and **IV** recorded with decoupling from protons contained six signals, whereas compound **II** displayed four signals. This means that cyclotriphosphazene **II**molecules) in solution have symmetrical structure and that molecules **I** and **IV** are not symmetric. The ^{13}C–^{31}P coupling constants through two, three, four, and five bonds in **IV** are much greater than the corresponding constants for compound **I**. The C^1 (*ipso*) and C^3 (*meta*) aromatic nuclei in molecule **I** interact with the phosphorus nucleus, while fluorine-containing polyphosphazene lacks such interactions.

The ^{31}P NMR spectra of **I**, **II**, **IV** may be interpreted as AB_2, AB_2, and A_n spin systems, respectively. According to the ^{31}P NMR data, no higher cyclophosphazene homologs are formed in the polymerization of hexachlorocyclotriphosphazene: the spectrum of polymer **IV** contained the only phosphorus signal at δ_P 6.31 ppm.

Study of the molecular weight distribution in polymeric products **III** and **IV** gave the following values: number-average molecular weight M_n 766 864 and 659 959 g mol^{-1}, weight-average molecular weight M_w 770 105 and 688 215 g mol^{-1}, and polydispersity index (PDI) 1.004 and 1.042, respectively.

The thermal stability of polyphosphazene **IV** was studied under atmospheric conditions in the temperature range from 20 to 1200°C. Figure 2 shows the TG and DTA curves for polymer **IV**. It is seen that decomposition starts at 136°C. Increase in the temperature from 136 to 381°C is accompanied by 50% weight loss, and it reaches 84% at 845°C. The DTA curve reveals two endothermic reactions at 300 and 510°C. Taking into account 50% weight loss, the observed peaks were attributed to structural rearrangements. Strong exothermic effects in the regions 400–450 and 650–700°C are likely to result from

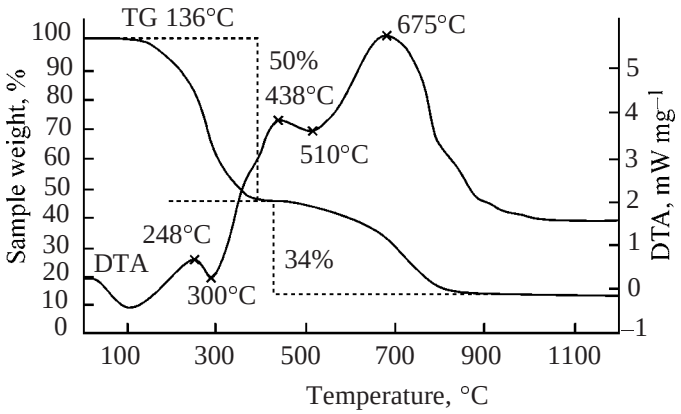


Fig. 2. Data of thermogravimetric and differential thermal analysis of polymer **IV**.

oxidation of the polymer. The corresponding weight loss amounts to 34% in the TG curve and represents liberation of CO_2, H_2O, and NH_3. The product formed after decomposition in the range 400–700°C is stable up to 1200°C. Presumably, thermal degradation of polymer **IV** is a complicated process involving both transformations of particular groups and oxidation.

The results of testing compounds **I**, **II**, and **IV** for antibacterial activity are summarized in the table; for comparison, the corresponding data for reference antibiotics are given. It is seen that compounds **I**, **II**, and **IV** are active against both Gram-positive and Gram-negative bacteria and yeast cultures, although the activity against Gram-positive bacteria is usually higher than against Gram-negative bacteria [25]. Polymer **IV** showed the strongest effect against all tested bacteria. Compounds **I**, **II**, and **IV** turned out to be more active with respect to *Staphylococcus aureus* than reference antibiotics except for Ofloxacin and Tetracyclin. Compounds **I** and **IV** were more active than Cefotaxime against acid-fast *Mycobacterium smegmatis* but less active than reference antibiotics against *Micrococcus luteus* and *Proteus vulgaris* (except for Penicillin in the latter case). A strong effect was also observed toward *Bacillus cereus* and *Escherichia coli*. Compounds **I**, **II**, and **IV** also showed more or less pronounced activity against other bacteria.

Polymer **IV** was more efficient than commercial antifungal agents (such as Nystatin, Ketoconazole, and Clotrimazole) in tests with the yeast cultures *Candida albicans* and *Kluyveromyces fragilis*. Compounds **I**, **II**, and **IV** showed different activities against other microorganisms. These differences may be rationalized in terms of different structures of cell membranes: cell membrane of Gram-positive bacteria consists of a single layer, *Gram-negative bacteria* have multilayered

Antibacterial activity of compounds **I, II**, and **IV** and some commercial antibiotics [a]

Microorganism	Inhibition zone diameter, mm											
	I	**II**	**IV**	P10	AM20	CTX30	VA30	OFX5	TE30	NY100	KET20	CLT10
Escherichia coli	15	14	19	18	12	10	22	30	28	–	–	–
Staphylococcus aureus	16	16	22	13	16	12	13	24	26	–	–	–
Klebsiella pneumoniae	13	–	17	18	14	13	22	28	30	–	–	–
Pseudomonas aeruginosa	14	11	18	8	10	54	10	44	34	–	–	–
Proteus vulgaris	11	14	13	10	16	18	20	28	26	–	–	–
Bacillus cereus	17	13	20	14	12	14	18	30	25	–	–	–
Mycobacterium smegmatis	12	–	16	15	21	11	20	32	24	–	–	–
Listeria monocytogenes	17	14	18	10	12	16	26	30	28	–	–	–
Micrococcus luteus	15	–	17	36	32	32	34	28	22	–	–	–
Candida albicans	18	14	22	–	–	–	–	–	–	20	21	15
Kluyveromyces fragilis	14	–	22	–	–	–	–	–	–	18	16	18
Rhodotorula rubra	15	–	20	–	–	–	–	–	–	18	22	16
H. guilliermondii	14	10	19	–	–	–	–	–	–	21	24	22
Debaryomyces hansenii	14	12	16	–	–	–	–	–	–	16	14	18

[a] P10 is Penicillin G, 10 units; AM20 is Ampicillin, 10 μg; CTX30 is Cefatoxime, 30 μg; V30 is Vancomycin, 30 μg; OFX5 is Ofloxacin, 5 μg; TE30 is Tetracyclin, 30 μg; N100 is Nystatin, 100 μg; KET20 is Ketoconazole, 20 μg; and CLT10 is Clotrimazole, 10 μg.

cell membranes, while yeast cell wall has even more complex structure [26].

Thus, the results of biological studies show that phosphazene derivatives are capable of generating novel metabolites displaying high affinity for most receptors. The strong anticandidal effect of these compounds makes them promising from the viewpoint of developing new anticandidal agents with a broad spectrum of activity. Compounds **I**, **II**, and **IV** may be selected for further pharmacological tests as potential drugs against many infectious diseases.

EXPERIMENTAL

Commercial hexachlorocyclotriphosphazene (2,2,4,4,6,6-hexachloro-1,3,5,2λ^5,4λ^5,6λ^5-triazatriphosphinine, Aldrich) was recrystallized from hexane and additionally purified by fractional vacuum sublimation at 55°C just before use. Benzene, tetrahydrofuran, and hexane (Merck) were distilled over metallic sodium in the presence of benzophenone and were stored over molecular sieves. 4-Fluoroaniline and 4-chloroaniline (Merck) were used without additional purification. Column chromatography was performed on silica gel (70–230 mesh, 60 Å; Aldrich); Kieselgel 60 F 254 plates were used for thin-layer chromatography. All operations were performed under argon using standard Schlenk glassware. The ^{1}H, ^{13}C, and ^{31}P NMR spectra were recorded on a Bruker DPX spectrometer at 400, 101.6, and 161.99 MHz, respectively. The ^{1}H and ^{13}C chemical shifts were measured relative to tetramethylsilane as internal reference; the ^{31}P chemical shifts were determined relative to 85% H_3PO_4 (external). The IR spectra were recorded on a Perkin–Elmer BX II instrument from samples prepared as KBr pellets. The elemental compositions were determined on a LECO CHNS-932 C,H,N analyzer. The melting points were measured on an Electrothermal IA 9100 apparatus in capillaries. Thermogravimetric analysis was performed on an NETZSCH Simulton Thermal Analyzer STA 409 EP instrument in the temperature range from 20 to 1200°C (rate 10 deg min^{-1}) in air.

The number-average (M_n) and weight-average (M_w) molecular weights and polydispersity indices (PDI) were determined by size-exclusion chromatography (SEC) on a Shimadzu instrument equipped with a refractive index detector (25°C) and a 3.3×300-mm column packed with SGX (grain size 100 Å, pore diameter 7 nm); dimethylformamide was used as eluent (flow rate 0.4 ml min^{-1}), and polystyrene was used as standard.

2,4,4,6,6-Pentachloro-2-(4-fluorophenylamino)-1,3,5,2λ^5,4λ^5,6λ^5-triazatriphosphinine (I). A solution of 3.19 g of 4-fluoroaniline in 50 ml of anhydrous THF was added dropwise over a period of 1 h to a solution of 5.0 g of hexachlorocyclotriphosphazene in 100 ml of anhydrous THF under stirring at –20°C in an argon atmosphere. The mixture was allowed to warm up to room temperature and was then heated under reflux for 4 h with protection from atmospheric moisture. The precipitate of 4-fluoroaniline hydrochloride was filtered off, the filtrate was evaporated on a rotary evaporator, and the residue was subjected to column chromatography on silica gel (120 g) using chloroform as eluent. The product was additionally purified by recrystallization from methylene chloride–*n*-hexane (3 : 1). Yield 3.76 g (62%), white crystals, mp 54°C. IR spectrum (KBr), ν, cm^{-1}: 3180 m (N–H); 1509 s (C=C); 1377 m (C–N); 1196 s (P=N); 592 s, 523 s (P–Cl). ^{31}P NMR spectrum ($CDCl_3$), δ_P, ppm: 13.62 t (1P, P^2, $^2J_{PP}$ = 48 Hz), 22.59 d (2P, P^4, P^6, $^2J_{PP}$ = 48 Hz). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 5.52 s (1H, NH), 7.07 m (2H, H_{arom}), 7.19 m (2H, H_{arom}). ^{13}C NMR spectrum ($CDCl_3$), δ_C, ppm: 116.78 d (1C, $^3J_{PC}$ = 22.8 Hz), 123.82 d (1C, $^4J_{PC}$ = 15.2 Hz), 123.9 s (1C), 132.69 s (1C), 159.35 d (1C, $^2J_{PC}$ = 2 Hz), 161.78 d (1C, $^5J_{PC}$ = 2 Hz). Mass spectrum, *m/z* for ^{35}Cl (I_{rel}, %): 425 (25) $[M + 3H]^+$, 422.5 (65) $[M]^+$, 421.7 (100) $[M - H]^+$, 386 (35) $[M - HCl]^+$, 312 (45) $[M - NHC_6H_4F]^+$. Found, %: C 17.03; H 1.16; N 13.23. $C_6H_5Cl_5FN_4P_3$. Calculated, %: C 17.06; H 1.19; N 13.27.

2,4,4,6,6-Pentachloro-2-(4-chlorophenylamino)-1,3,5,2λ^5,4λ^5,6λ^5-triazatriphosphinine (II) was synthesized in a similar way from 3.57 g of 4-chloroaniline. Yield 3.31 g (54%), white crystals, mp 75°C. IR spectrum (KBr), ν, cm^{-1}: 3161 m (N–H); 1495 s (C=C); 1376 m (C–N); 1189 s (P=N); 598 s, 526 s (P–Cl). ^{31}P NMR spectrum ($CDCl_3$), δ_P, ppm: 12.88 t (1P, P^2, $^2J_{PP}$ = 48 Hz), 22.54 d (2P, P^4, P^6, $^2J_{PP}$ = 49 Hz). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 5.59 d (1H, NH, $^2J_{PH}$ = 9.0 Hz), 7.18 m (2H, H_{arom}), 7.38 m (2H, H_{arom}). ^{13}C NMR spectrum ($CDCl_3$), δ_C, ppm: 122.35 d (1C, $^3J_{PC}$ = 7.6 Hz), 130.05 s (1C), 130.26 d (1C, $^5J_{PC}$ = 2.0 Hz), 135.72 s (1C). Mass spectrum, *m/z* for ^{35}Cl (I_{rel}, %): 441 (35) $[M + 2H]$, 439 (55) $[M]^+$, 437 (100) $[M - 2H]^+$, 402 (23) $[M - HCl]^+$, 312 (53) $[M - NHC_6H_4Cl]^+$. Found: C 16.30; H 1.13; N 12.76. $C_6H_5Cl_6N_4P_3$. Calculated, %: C 16.40; H 1.14; N 12.77.

Poly(dichlorophosphazene) (III) was synthesized and purified according to the procedures described in [12, 27]. IR spectrum (KBr), ν, cm^{-1}: 1255 s (P=N); 766 s, 578 s (P–Cl). M_n = 766 864 g mol^{-1}; M_w = 770 105 g mol^{-1}; PDI = 1.004.

Poly[bis(4-fluorophenylamino)phosphazene] (IV). A solution of 19.13 g of 4-fluoroaniline in 50 ml of anhydrous THF was added dropwise over a period of 1 h to a solution of 5.0 g of poly(dichlorophosphazene) (**III**) in 150 ml of anhydrous THF under stirring at –20°C in an argon atmosphere. The mixture was allowed to warm up to room temperature and was then heated under reflux for 48 h with protection from atmospheric moisture. The precipitate of 4-fluoroaniline hydrochloroide was filtered off, the filtrate was evaporated on a rotary evaporator, the residue was treated with water, and the product was extracted into chloroform. Yield 9.98 g (87%), brown solid, mp >300°C (decomp.). IR spectrum (KBr), ν, cm^{-1}: 3366 m (N–H), 1511 s (C=C), 1370 m (C–N), 1211 s (P=N). ^{31}P NMR spectrum ($CDCl_3$): δ_P 6.31 ppm, s. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 5.40 d (1H, NH, $^2J_{PH}$ = 20 Hz), 6.93 m (2H, H_{arom}), 7.09 m (2H, H_{arom}). ^{13}C NMR spectrum ($CDCl_3$), δ_C, ppm: 115.10 s (1C), 115.33 s (1C), 122.20 d (1C, $^3J_{PC}$ = 22.8 Hz), 131.77 d (1C, $^4J_{PC}$ = 2.8 Hz), 157.65 d (1C, $^2J_{PC}$ = 9.6 Hz), 160.78 d (1C, $^5J_{PC}$ = 10 Hz).

The following bacteria and yeast cultures were used in antibacterial tests: *Escherichia coli* ATCC 11230, *Staphylococcus aureus* ATCC 6538, *Klebsiella pneumoniae* UC57, *Micrococcus luteus* La 2971, *Proteus vulgaris* ATCC 8427, *Pseudomonas aeruginosa* ATCC 27853, *Mycobacterium smegmatis* CCM 2067, *Bacillus cereus* ATCC 7064, *Listeria monocytogenes* ATCC 15313, *Candida albicans* ATCC 10231, *Kluyveromyces fragilis* NRRL 2415, *Rhodotorula rubra* DSM 70403, *Debaryomyces hansenii* DSM 70238, and *Hanseniaspora guilliermondii* DSM 3432. The tests were carried out following the disk diffusion technique according to the procedure outlined by the National Committee for Clinical Laboratory Standards (NCCLS) [28]. Compounds **I**, **II**, and **IV** were dissolved in dimethyl sulfoxide (DMSO) to a concentration of 30 μg ml^{-1}. Empty sterilized paper disks were impregnated each with 20 μl of the above solution. The above-listed bacteria were incubated for 24 h at 30 ± 0.1°C by inoculation into Nutrient Broth (Difco), and yeasts were incubated

for 48 h on Malt Extract Broth (Difco). An inoculum containing about 10^6/ml bacteria cells or 10^8/ml yeast cells was spread over Mueller-Minton Agar (Oxoid) plates (1 ml per plate). The injected disks were placed on the inoculated agar by pressing slightly and were incubated for 24 h at 35°C in tests with bacteria or for 72 h at 25°C in tests with yeast cultures. Disks injected with the corresponding reference antibiotic were applied on each plate for comparison [28, 29].

ACKNOWLEDGMENTS

The authors are grateful to the Scientific and Technical Research Council of Turkey, TÜBİTAK for financial support of this work [grant no. TBAG-2159 (102T053)].

REFERENCES

1. Allen, C.W., *Chem. Rev.*, 1991, vol. 91, p. 119.
2. Contractor, S.R., Kilic, Z., and Shaw, R.A., *J. Chem. Soc. Dalton Trans.*, 1987, p. 2023.
3. Krishnamurthy, S.S., Sau, A.C., and Woods, M., *Advances in Inorganic Chemistry and Radiochemistry*, Emeléus, H.J., New York: Academic, 1978, vol. 21, p. 41.
4. Allcock, H.R., *Chem. Rev.*, 1972, vol. 72, p. 315.
5. Harris, P.J. and Williams, K.B., *Inorg. Chem.*, 1984, vol. 23, p. 1496.
6. Labarre, J.F., *Top. Curr. Chem.*, 1985, vol. 129, p. 173.
7. Kiliç, A., Begeç, S., Çetinkaya, B., Hökelek, T., Kiliç, Z., Gündüz, N., and Yildiz, M., *Heteroatom Chem.*, 1996, vol. 7, p. 249.
8. Allcock, H.R., Sunderland, N.J., Primrose, A.P., Rheingold, A.L., Guzei, I.A., and Parvez, M., *Chem. Mater.*, 1999, vol. 11, p. 2478.
9. Yildiz, M., Erdener, D., Ünver, H., and İskeleli, N.O., *J. Mol. Struct.*, 2005, vol. 753, p. 165.
10. Shaw, R.A. and Ture, S., *Phosphorus, Sulfur Silicon*, 1991, vol. 57, p. 103.
11. Yildiz, M., Kiliç, Z., and Hökelek, T., *J. Mol. Struct.*, 1999, vol. 510, p. 227.
12. Allcock, H.R., Kugel, R.L., and Valan, K.J., *Inorg. Chem.*, 1966, vol. 5, p. 1709.
13. Pemberton, L., Jaeger, R.D., and Gengembre, L., *Polymer*, 1998, vol. 39, p. 1299.
14. Selveraj, I.I., Chaklanobis, S., and Chandrasekhar, V., *Polym. Int.*, 1998, vol. 46, p. 111.
15. Chen-Yang, Y.N., Hwang, J.J., and Chang, F.H., *Macromolecules*, 1997, vol. 30, p. 3825.
16. Gomez, M.A., Marco, C., Fatou, J.G., Bowmer, T.N., Haddon, R.C., and Chichester-Hicks, S.V., *Macromolecules*, 1991, vol. 24, p. 3276.
17. Jaglowski, A.J., Singler, R.E., and Atkins, E.D.T., *Macromolecules*, 1995, vol. 28, p. 1668.
18. Allcock, H.R., *Biodegradable Polymers as Drug Delivery Systems*, New York: Marcel Dekker, 1990, p. 163.
19. Olshavsky, M.A. and Allcock, H.R., *Macromolecules*, 1995, vol. 28, p. 6188.
20. Allcock, H.R., Ravikiran, R., and Olshavsky, M.A., *Macromolecules*, 1998, vol. 31, p. 5206.
21. Roberts, S. and Ondrey, G., *Chem. Eng.*, 1996, vol. 6, p. 44.
22. Macdiarmid, A.G. and Epstein, A.J., *Faraday Discuss.*, 1989, vol. 88, p. 317.
23. Roncali, J., *Chem. Rev.*, 1992, vol. 92, p. 711.
24. Sulkowski, W., Sulkowska, A., Makarucha, B., and Kireev, V., *Eur. Polym. J.*, 2000, vol. 36, p. 1519.
25. McCutheon, A.R., Ellis, S.M., Hancock, R.E.W., Towers, G.H.N., *J. Ethnopharmacol.*, 1992, vol. 37, p. 213.
26. Mann, J. and Crabbe, M.J.C., *Bacteria and Antibacterial Agents*, Oxford (UK): Spectrum Academic, 1990, p. 74.
27. Hyounggee, B., Ok-Sang, J., Yong Kiel, S.D., and Youn, S.S., *Bull. Korean Chem. Soc.*, 1995, vol. 16, p. 1074.
28. *NCCLS, Performance Standards for Antimicrobial Disk Suspectibility Tests. Approved Standard NCCLS Publication M2-A5*, Villanova, PA (USA), 1993.
29. Collins, C.H., Lyre, P.M., and Grange, J.M., *Microbiological Methods*, London: Butterworths, 1989, 6th ed.