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Note

New tandem reactions for the construction of heterocycles via dianion strategy

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The reaction of chiral and achiral salen ditopic ligands with sodium in dry isopropanol generates remote O,O-dianions. The dianions thus generated *in situ* are trapped by suitable phosphorus dielectrophiles to yield 2-oxo and 2-thioxo-1,3,7-dioxazaphosphadecines and 2-oxo and 2-thioxo-1,3,7,10-dioxadiazaphosphatridecines in moderate to good overall yield. The products are characterized by elemental and spectral (IR, ^{1}H, ^{13}C, ^{31}P NMR and Mass) studies. A plausible mechanistic logic and concept utilized in the synthesis have been discussed.

Keywords: N-(2-Hydroxymethylphenyl)salicylideneimine, (+/-)-N,N′-bis(salicylidene)-*trans*-1,2-diaminocyclohexane, dianion, dioxazaphosphadecines, dioxadiazaphosphatridecines.

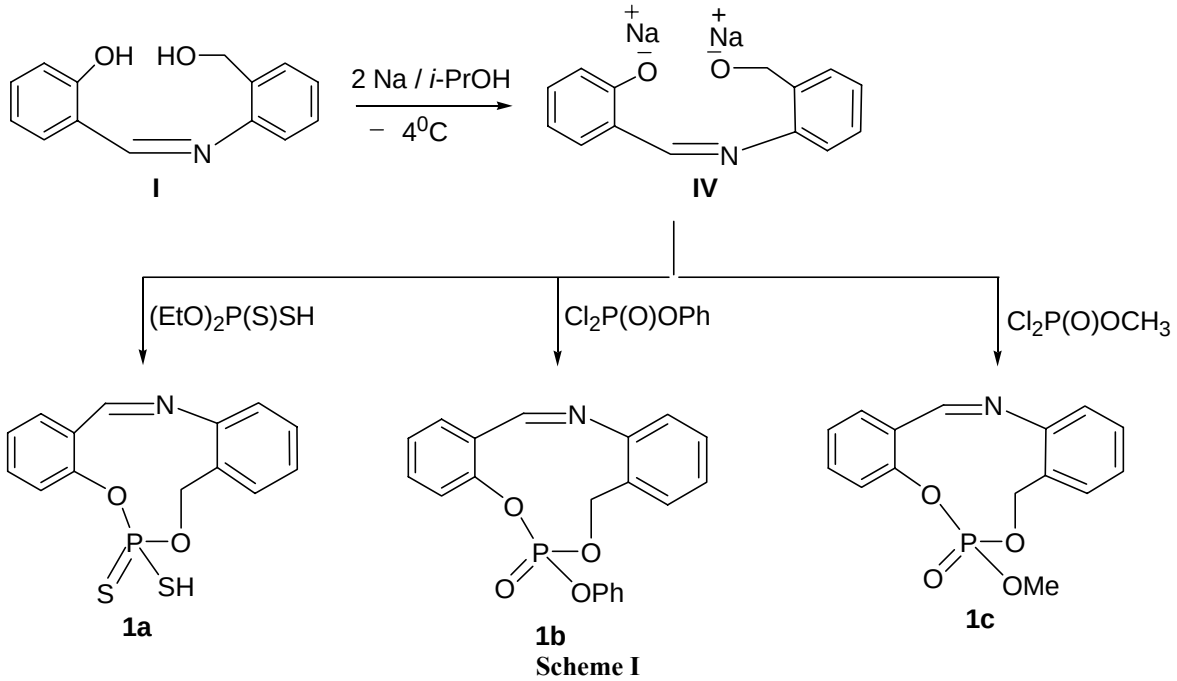
The formation of phosphorus-oxygen bond is one of the most fundamental and important processes for producing natural and unnatural useful compounds in organic synthesis. An increasing interest has been paid for several years to the chemistry of heterocyclic rings containing phosphorus due to their unique physical properties, specific chemical reactivity[1-3], and their remarkable potential biological activity[4-6]. Dioxaphosphepines have been synthesized and screened for their biological activity[7,8]. Dichlorophosphates and dichlorothiophosphates are of growing importance as targets for synthesis largely because of their ability to mimic biological activity. Numerous reactions employing $Cl_2P(X)OR$ (where, X = O, S) as a reagent have been reported[6] and reviewed[9-11]. Established methods for the synthesis of phosphorus derivatives commonly involve the use of phosphorus dielectrophiles, which are being attacked by the dianions formed. To date many reviews have covered the chemistry of dianions[12-14]. The development of chiral Schiff base ligands has received considerable interest since Jacobsen[15] and subsequently Katsuki[16] reported significant success in asymmetric epoxidation of unfunctionalized olefins by the chiral manganese(III) salen Schiff base catalysts. Salen ligands give complexes, which also hold promises in enantioselective cyclopropanation of styrenes, asymmetric aziridination of olefins, asymmetric Diels-Alder cycloaddition and enantioselective ring opening of epoxides[17]. With the aim to broaden further the range of N, P and O heterocycles and in continuation of our work[18-23] on phosphorus heterocycles, some new heterocycles, which are not reported in the literature, have been synthesized from ditopic ligands *via* dianion-mediated cyclizations.

Results and Discussion

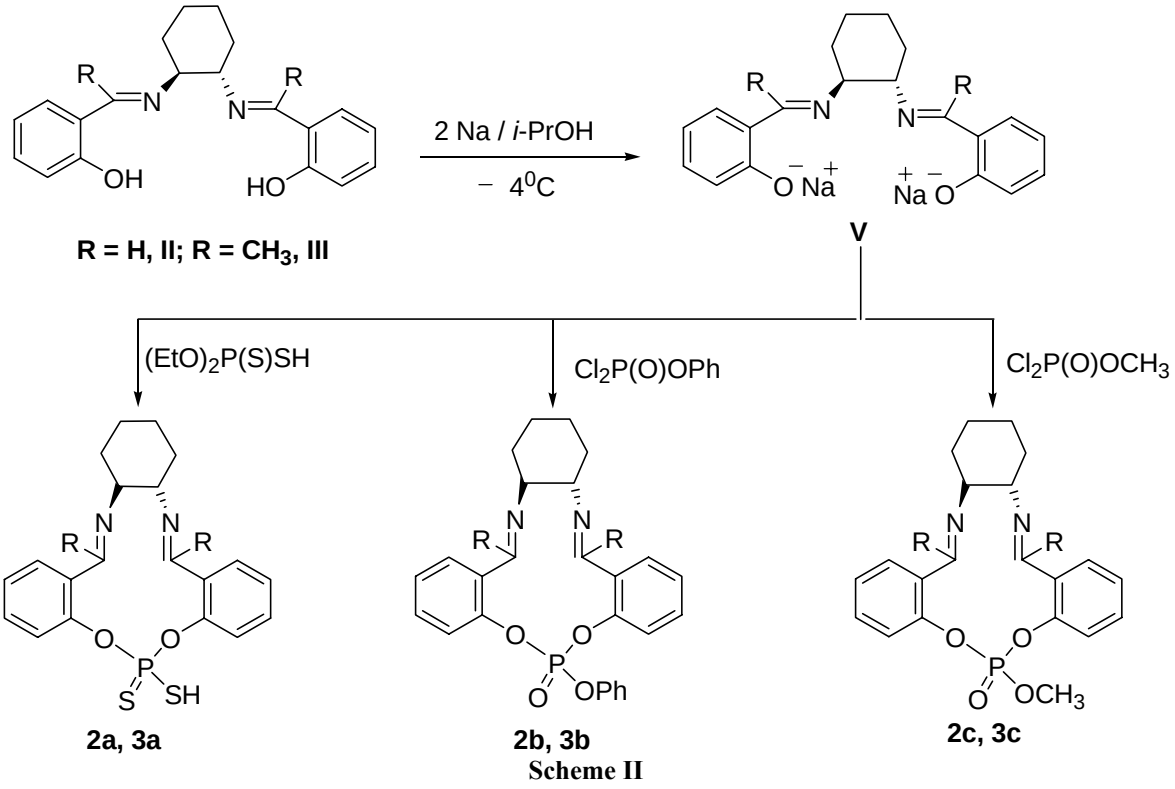
The synthesis of the remote dianions **IV** and **V** is quite straightforward. It relies on the double deprotonation of the neutral compounds **I**, **II** and **III**. Treatment of ditopic ligands **I**, **II** and **III** with sodium isopropoxide in 1:2 molar ratio followed by addition of diethyldithiophosphate or phenyl/methyldichlorophosphates gives 2-thioxo-2-thiol-2-oxo-2-phenoxy/-methoxydibenzo-dioxazaphosphadecines (**1a-c; Scheme I**) and dibenzodioxadiazaphosphatridecines (**2a-c, 3a-c; Scheme II**), respectively. To the best of our knowledge there are no reports available on the synthesis of ten/thirteen-member oxazaphospha-heterocycles. With the aim to broaden the range of useful phosphaheterocycles that may provide an easy access to synthetic intermediates and therapeutic agents, cyclization reactions of remote O,O-dianions *via* cyclophosphorylation have been carried out in one-pot. A remarkable feature of this reaction is the construction of oxygen-phosphorus bonds *via* a tandem process. The methodology opens an access to a broad variety of new chiral and achiral N, O and P containing heterocycles in good yields.

The disappearance of absorption band and signal corresponding to the -OH group in both IR and ^{1}H NMR spectra and the appearance of new bands in the region 1160-1030 cm^{-1} is consistent with the P-O-C stretching. There are also fairly strong bands in the region 1280-1215 and 825-750 cm^{-1} ascribable to ν(P=O) and ν(P=S) stretching vibrations[24] respectively, suggesting oxygen-phosphorus bond formation and cyclic structure of prepared compounds.

A sharp absorption band in the region 1600-1640 cm^{-1} exhibits ν(C=N). ^{1}H NMR spectra display a



Scheme I



Scheme II

singlet at δ 8.25 ppm for azomethine proton, a multiplet in the range δ 6.61-7.89 ppm for aromatic protons and a singlet around 4.75 ppm for benzylic CH_2 proton. A multiplet at δ 3.31 ppm for two aliphatic C-H protons and multiplet in the range δ 1.43-1.95 ppm displays for eight aliphatic CH_2 protons of cyclohexane ring. The signal for the methoxy protons appears in the expected region around δ 3.86 ppm. In addition the ^{13}C NMR spectra support the assigned structures. The ^{31}P NMR signals

of the phosphaheterocycles encompassed a range of 1.59 to −11.26 ppm in the pentacoordinate region, which is a common shift for phosphepines. In conclusion, we have developed a convenient one pot synthesis of oxazaphosphaheterocycles that can be run on moderate scale and is operationally scalable.

Experimental Section

Chemicals were obtained from Sigma-Aldrich, Merck, Fluka and Lancaster, and are used as such without further purification. All solvents (AR or extra pure grade) used for spectroscopic and other physical studies were further purified by literature methods[25]. All operations were performed under nitrogen atmosphere using standard glasswares. IR spectra were recorded as KBr discs and in nujol mull on JASCO FT/IR-5300 spectrophotometer. Melting points were determined using a calibrated thermometer by Remi Digital Melting Point apparatus and are uncorrected. Elemental analyses were performed by Central Drug Research Institute, Lucknow. NMR (^{1}H, ^{13}C and ^{31}P) spectra were recorded on a JEOL AL 300 FT NMR spectrometer. All chemical shifts were reported in parts per million, relative to TMS as an internal standard in $CDCl_3$. Mass spectra were recorded at 70 eV ionising voltage on a JEOL –D300 MS instrument.

Synthesis of N-(2-Hydroxymethylphenyl)salicylic-deneimine[26] I

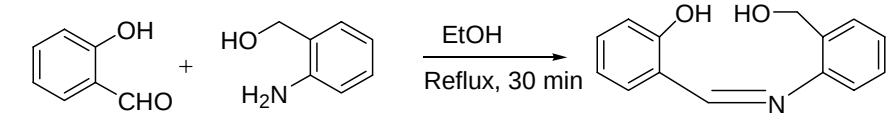
This ditopic ligand was first reported by Shyamal and Kale in 1980[26]. To a stirred solution of *o*-aminobenzylalcohol (46 mmoles, 5.66 g) in absolute ethanol (50 mL) added drop wise ethanolic (50 mL) solution of salicylaldehyde (46 mmoles, 4.82 mL) and refluxed for 30 min to get yellowish brown solution. Now concentrated the solution by rotavapour to get yellow solid and recrystallised the crude by dichloromethane to give **I**, yield 60 %, m.p. 121°C. Anal. Calcd. for $C_{14}H_{13}NO_2$: C, 73.99; H, 5.76; N, 6.16. Found: C, 73.64; H, 5.43; N, 6.37; IR (KBr): 3400 (OH), 2908 (CH), 1618 cm^{-1} (C=N); ^{1}H NMR (300 MHz, $CDCl_3$): δ 13.05 (s, 1H, OH), 8.61 (s, 1H, CH=N), 6.93-7.53 (m, 9H, ArH and OH, CH_2OH), 4.87 (s, 2H, CH_2); ^{13}C NMR (75 MHz, $CDCl_3$): δ 61.40, 116.86, 117.56, 118.57, 119.36, 126.65, 127.88, 128.40, 131.96, 132.99, 134.03, 146.19, 160.62, 162.74. FAB MS: *m/z* 228 $[M+H]^+$.

Synthesis of (+/-)-N,N′-bis(salicylidene)-*trans*-1,2-diaminocyclohexane II

To a solution of 1,2-diaminocyclohexane (1.23 mmole, 140 mg) in absolute ethanol (1 mL) was added salicylaldehyde (2.46 mmoles, 300 mg). The resulting mixture was then refluxed for 3 hr. After cooling to room temperature, water (5 mL) was added and the mixture stirred for 30 min. The yellow precipitate formed was filtered, washed with water and dried. Recrystallization from absolute ethanol afforded yellow needles of **II,** yield 70.5 %, m.p. 74°C. Anal. Calcd. for $C_{20}H_{22}N_2O_2$: C, 74.51; H, 6.88; N, 8.69; Found: C, 74.26; H, 6.49; N, 8.47. IR (Nujol): 3060 (OH), 2932 (CH), 1630 cm^{-1} (C=N); ^{1}H NMR (300 MHz, $CDCl_3$): δ 13.29 (s, 2H, OH), 8.25 (s, 2H, CH=N), 7.25-6.75 (m, 8H, ArH), 3.29 (m, 2H, CH), 1.88 (m, 4H, CH_2), 1.78 (m, 2H, CH_2), 1.43 (m, 2H, CH_2); ^{13}C NMR (75 MHz, $CDCl_3$): δ 164.70, 160.97, 132.14, 131.47, 118.67, 118.58, 116.77, 72.65, 33.11, 24.18. FAB MS: *m/z* 323 $[M+H]^+$.

Synthesis of (+/-)-N,N′-bis(2-hydroxyacetophenonylidene)-*trans*-1,2-diaminocyclohexane[27] III

To a solution of 1,2-diaminocyclohexane (1.23 mmole, 140 mg) in absolute ethanol (1 mL) was



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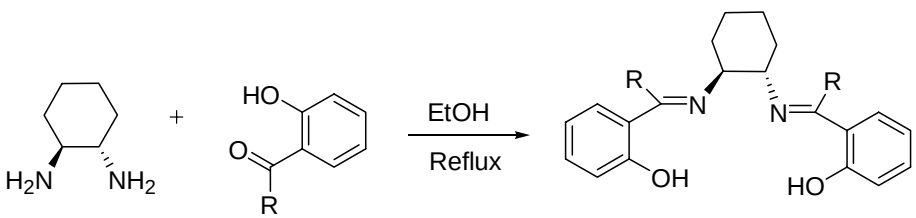

R = H, **II**; R = CH_3, **III**

added 2-hydroxyacetophenone (2.46 mmoles, 334 mg). The resulting mixture was then refluxed for 36 hr. After cooling to room temperature, water (5 mL) was added and the mixture stirred for 30 min. The yellow precipitate formed was filtered, washed with water and recrystallised from absolute ethanol to give yellow needles **3, y**ield 70.5 %, m.p. 144-145°C. Anal. Calcd. for $C_{22}H_{26}N_2O_2$: C, 75.40; H, 7.48; N, 7.99; Found: C, 75.25; H, 7.26; N, 7.85. IR (Nujol): 3060 (OH), 1618 cm^{-1} (C=N); ^1H NMR (300 MHz, $CDCl_3$): δ 16.40 (s, 2H, OH), 7.37-6.67 (m, 8H, ArH), 3.80 (m, 2H, CH), 2.20 (s, 6H, CH_3), 1.90 (m, 4H, CH_2), 1.84 (m, 2H, CH_2), 1.46 (m, 2H, CH_2); ^{13}C NMR (75 MHz, $CDCl_3$): δ 170.65, 163.48, 132.18, 128.22, 119.10, 118.30, 116.97, 62.82, 32.19, 24.02, 14.20. FAB MS: *m/z* 351 $[M+H]^+$.

Synthesis of 2-thioxo-2-thioldibenzodioxazaphosphadecine 1a

Typical procedure: To a stirred suspension of sodium (2 mmoles, 46 mg) in dry isopropanol (5 mL) was added alcoholic solution (55 mL) of ligand I (1 mmole, 227 mg) drop wise with constant stirring at –4°C for 6 hr in an inert atmosphere. Further (1 mmole, 186.23 mg) of diethyldithiophosphate in dry isopropanol (5 mL) was added drop wise with constant stirring to the yellowish solution of dianion generated *in situ*, and the reaction mixture was stirred at room temperature for an additional 6 hr. Completion of the reaction was checked by TLC. The reaction mixture was evaporated with the help of rotary evaporator and the residue was subjected to column chromatography (diethyl ether) to give 1a (196 mg, 61 %), m.p. 97°C. Anal. Calcd. for $C_{14}H_{12}NO_2S_2P$: C, 52.32; H, 3.76; N, 4.35. Found: C, 52.07; H, 3.48; N, 4.27. IR (Nujol): 1614, 1458, 1030, 908, 825, 758, 559 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 8.61 (s, 1H), 7.53-6.93 (m, 8H, ArH), 4.87 (s, 2H, OCH_2), 1.74 (s, 1H, SH); ^{13}C NMR (75 MHz, $CDCl_3$): δ 62.24, 117.69, 118.39, 119.57, 119.63, 127.47, 128.70, 129.22, 132.78, 133.81, 134.85, 147.03, 161.46, 163.57; ^{31}P NMR (121 MHz, $CDCl_3$): δ 1.59; FAB MS: *m/z* 321 (M^+).

1b: Same procedure as for **1a** with 227 mg (1 mmole) of, N-(2-hydroxymethylphenyl)-salicylideneimine, 46 mg (2 mmoles) of sodium, 210 mg (1 mmole) of phenyldichlorophosphate; yield: 211 mg (58 %), m.p. 90°C. Anal. Calcd. for $C_{20}H_{16}NO_4P$: C, 65.76; H, 4.41; N, 3.83. Found: C, 65.43; H, 4.23; N, 3.72; IR (Neat): 1643, 1489, 1385, 1207, 1097, 929, 825 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 8.49 (s, 1H), 6.88-7.41 (m, 13H, ArH), 4.75 (s, 2H, OCH_2); ^{13}C NMR (75 MHz, $CDCl_3$): δ 61.49, 117.12, 119.05, 120.26, 125.00, 126.93, 127.38, 128.06, 128.55, 129.27, 129.68, 131.02, 131.24, 132.29, 133.24, 134.50, 146.32, 160.93, 162.96; ^{31}P NMR (121 MHz, $CDCl_3$): δ –11.23; FAB MS: *m/z* 365 (M^+).

1c: Same procedure as for **1a** with 227 mg (1 mmole) of, N-(2-hydroxymethylphenyl)-salicylideneimine, 46 mg (2 mmoles) of sodium, 148.91 mg (1 mmole) of methyldichlorophosphate; yield: 166 mg (55 %), m.p. 75°C. Anal. Calcd. for $C_{15}H_{14}NO_4P$: C, 59.42; H, 4.65; N, 4.62. Found: C, 59.12; H, 4.37; N, 4.26; IR (Nujol): 1632, 1385, 1215, 1091, 929, 825 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 8.57 (s, 1H), 7.53-6.92 (m, 8H, ArH), 4.62 (s, 2H, OCH_2), 3.80 (s, 3H, OCH_3); ^{13}C NMR (75 MHz, $CDCl_3$): δ 21.58, 61.47, 117.13, 117.78, 119.04, 126.95, 128.09, 128.56, 132.28, 133.26, 134.41, 146.26, 160.91, 162.89; ^{31}P NMR (121 MHz, $CDCl_3$): δ –6.15; FAB MS: *m/z* 303 (M^+).

2a: Same procedure as for **1a** with 322 mg (1 mmole) of, N,N′-1,2-cyclohexylene bis-(salicylideneimine), 46 mg (2 mmoles) of sodium, 186.23 mg (1 mmole) of diethyldithiophosphate; yield: 254 mg, (61 %), m.p. 98°C. Anal. calcd. for: $C_{20}H_{21}N_2O_2S_2P$: C, 57.67; H, 5.08; N, 6.72. Found: C, 57.35; H, 4.86; N, 6.48; IR (Nujol): 3746, 2675, 1763, 1630, 1278, 1215, 1093, 929, 844, 825, 756 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 8.25 (s, 2H), 6.78-7.25 (m, 8H, ArH), 3.33 (m, 2H, CH), 1.51-1.92 (m, 8H, CH_2CH_2), 1.74 (s, 1H, SH); ^{13}C NMR (75 MHz, $CDCl_3$): δ 23.12, 32.04, 71.56, 115.72, 117.54, 117.60, 126.66, 130.42, 131.10, 133.31, 159.92, 163.66; ^{31}P NMR (121 MHz, $CDCl_3$): δ 1.59; FAB MS: *m/z* 416 (M^+).

2b: Same procedure as for **1a** with 322 mg (1 mmole) of, N,N′-1,2-cyclohexylene bis(salicylideneimine), 46 mg (2 mmoles) of sodium, 210 mg (1 mmole) of phenyldichlorophosphate; yield: 271 mg (59 %), m.p. 78°C. Anal. Calcd. for: $C_{26}H_{25}N_2O_4P$: C, 67.82; H, 5.47; N, 6.08. Found: C, 67.68; H, 5.23; N, 5.82; IR (Nujol): 1631, 1280, 1143, 1093, 922, 825 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 8.25 (s, 2H), 6.69-7.25 (m, 13H, ArH), 3.29 (m, 2H, CH), 1.25-1.95 (m, 8H, CH_2CH_2); ^{13}C NMR (75 MHz, $CDCl_3$): δ 24.09, 33.01, 72.50, 116.08, 116.71, 116.97, 118.53, 118.55, 123.97, 129.33, 129.71, 131.41, 132.12, 132.21, 149.67, 160.93, 164.65; ^{31}P NMR (121 MHz, $CDCl_3$): δ –7.09; FAB MS: *m/z* 460 (M^+).

2c: Same procedure as for **1a** with 322 mg (1 mmole) of, N,N′-1,2-cyclohexylene bis(salicylic-deneimine), 46 mg (2 mmoles) of sodium, 148.91 mg (1 mmole) of methyldichlorophosphate; yield: 207 mg (52 %), m.p. 67°C. Anal. Calcd. for: $C_{21}H_{23}N_2O_4P$: C, 63.32; H, 5.82; N, 7.03. Found: C, 63.12; H, 5.54; N, 6.78; IR (Neat): 1763, 1628, 1215, 1091, 929, 825 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 8.25 (s, 2H), 6.81-7.89 (m, 8H, ArH), 3.33 (m, 2H, CH), 1.26-1.92 (m, 8H, CH_2CH_2), 3.76 (s, 3H, OCH_3),); ^{13}C NMR (75 MHz, $CDCl_3$): δ 29.58, 23.12, 32.04, 71.56, 115.72, 117.54, 117.60, 126.66, 130.42, 131.10, 133.31, 159.92, 163.66; ^{31}P NMR (121 MHz, $CDCl_3$): δ –3.75; FAB MS: *m/z* 398 (M^+).

3a: Same procedure as for **1a** with 350 mg (1 mmole) of, N,N′-1,2-cyclohexylenebis(2-hydroxy-acetophenonylideneimine), 46 mg (2 mmoles) of sodium, 186.23 mg (1 mmole) of diethyldithio-phosphate; yield: 280 mg (63 %), m.p. 130°C. Anal. Calcd. for $C_{22}H_{25}N_2O_2S_2P$: C, 59.44; H, 5.67; N, 6.30. Found: C, 59.13; H, 5.23; N, 6.08; IR (KBr): 3491, 2856, 2675, 1763, 1626, 1383, 825, 723 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 6.61-7.32 (m, 8H, ArH), 3.80 (m, 2H, CH), 2.20 (s, 6H, CH_3), 1.19-1.90 (m, 8H, CH_2CH_2), 1.82 (s, 1H, SH); ^{13}C NMR (75 MHz, $CDCl_3$): δ 14.20, 24.02, 32.19, 62.82, 116.97, 118.30, 119.10, 128.22, 132.18, 163.48, 170.62; ^{31}P NMR (121 MHz, $CDCl_3$): 1.63; FAB MS: *m/z* 444 (M^+).

3b: Same procedure as for **1a** with 350 mg (1 mmole) of, N,N′-1,2-cyclohexylenebis(2-hydroxy-acetophenonylideneimine), 46 mg (2 mmoles) of sodium, 210 mg (1 mmole) of phenyldichloro-phosphate; yield: 273 mg (56 %), m.p. 80°C. Anal. Calcd. for $C_{28}H_{29}N_2O_4P$: C, 68.84; H, 5.98; N, 5.73. Found: C, 68.56; H, 5.72; N, 5.46; IR (Neat): 1641, 1286, 1192, 1093, 968, 825 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 6.63-7.31 (m, 13H, ArH), 3.81 (m, 2H, CH), 2.20 (s, 6H, CH_3), 1.18-1.85 (m, 8H, CH_2CH_2); ^{13}C NMR (75 MHz, $CDCl_3$): δ 14.34, 24.16, 32.33, 62.92, 116.01, 117.07, 118.49, 119.18, 120.04, 124.50, 128.34, 130.26, 130.63, 132.35, 163.70, 170.83; ^{31}P NMR (121 MHz, $CDCl_3$): δ –11.26; FAB MS: *m/z* 488 (M^+).

3c: Same procedure as for **1a** with 350 mg (1 mmole) of, N,N′-1,2-cyclohexylenebis(2-hydroxy-acetophenonylideneimine), 46 mg (2 mmoles) of sodium, 148.91 mg (1 mmole) of methyl-dichlorophosphate; yield: 213 mg (50 %), m.p. 81°C. Anal. Calcd. for $C_{23}H_{27}N_2O_4P$: C, 64.78; H, 6.38; N, 6.57. Found: C, 64.52; H, 6.13; N, 6.23; IR (Nujol): 1763, 1610, 1215, 1091, 929, 825 cm^{-1}; ^1H NMR (300 MHz, $CDCl_3$): δ 6.93-7.66 (m, 8H, ArH), 3.91 (m, 2H, CH),, 2.49 (s, 6H, CH_3), 1.18-1.97 (m, 8H, CH_2CH_2), 3.82 (s, 3H, OCH_3); ^{13}C NMR (75 MHz, $CDCl_3$): δ 26.07, 14.20, 24.02, 32.19, 62.82, 116.97, 118.30, 119.10, 128.22, 132.18, 163.48, 170.62; ^{31}P NMR (121 MHz, $CDCl_3$): δ – 0.94; FAB MS: *m/z* 426 (M^+).

Conclusion

In conclusion, a quite simple procedure, low consumption of solvent, fast reaction rates, mild reaction condition and good yield of the reaction make this protocol an attractive and useful contribution to the preparation of a rare class of nitrogen, oxygen and phosphorus macroheterocycles. Other current targets include novel heterocycles, macrocycles and chiral ligands for homogeneous catalysis.

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