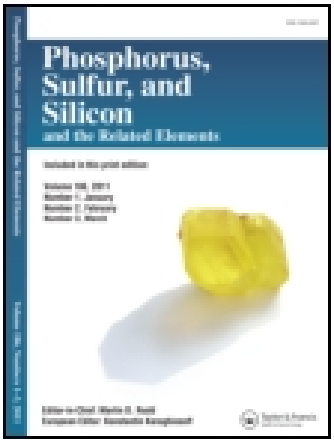

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Tween-20: An Efficient Catalyst for One-Pot Synthesis of α-Aminophosphonates in Aqueous Media

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TWEEN-20: AN EFFICIENT CATALYST FOR ONE-POT SYNTHESIS OF α-AMINOPHOSPHONATES IN AQUEOUS MEDIA

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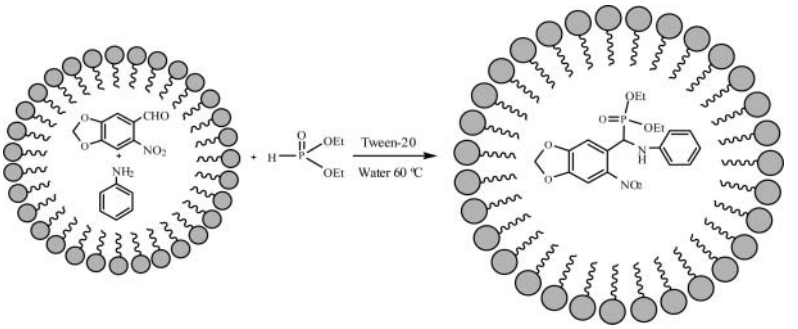
GRAPHICAL ABSTRACT



Abstract *A green one-pot three-component synthesis has been developed for α-aminophosphonates by condensation of aldehydes, amines, and diethylphosphite by using nonionic surfactant Tween-20 as catalyst in aqueous media. The results showed that this synthetic route for α-aminophosphonates takes just 25–60 min for completion at 60 °C and affords 64%–91% yields depending on the nature of the amine substrates. The major advantages of this novel method are green reaction conditions with water as solvent, simple workup, less reaction times, and high to moderate yields.*

Keywords α-Aminophosphonates; Aqueous media; Three-component one-step reaction; Tween-20

INTRODUCTION

Multicomponent, one-pot synthesis has received considerable attention because of its wide range of applications in pharmaceutical chemistry for the creation of structural

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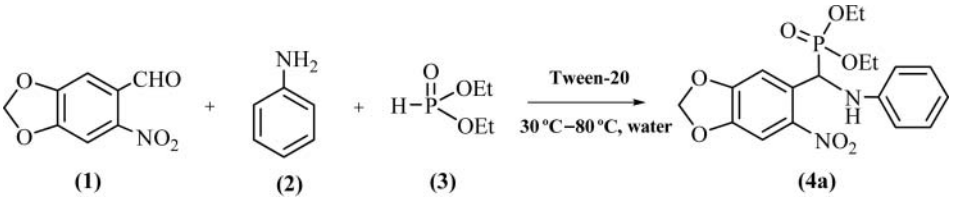
diversity and combinatorial libraries of drug discovery.[1] Multicomponent reactions are extremely convergent, producing remarkably high increase of molecular complexity in a single-step operation.[2] Multicomponent synthesis of α-aminophosphonates has gained much interest in recent times, due to their structural similarity to the corresponding α-aminophosphonic acids and transition state mimics of peptide hydrolysis.[3,4] Many natural and synthetic α-aminophosphonates and their derivatives have greater prominence as anti-HIV, cancer, biotic, bacterial, tumor, and antiviral agents.[5–10] Furthermore, α-aminophosphonates are used in agricultural industry as fungicidal, herbicidal agents, and plant growth regulators.[11–13] Consequently, different methods have been developed for their synthesis. Among them, nucleophilic addition of phosphites to imines is an established most useful method in which catalysts like $InCl_3$, $SnCl_4$, amberlite-IR 120, sulfamic acid, TiO_2, $In(OTf)_3$, Silica sulphuric acid, $Al(H_2PO_4)_3$, $Mg(ClO_4)_2$, $TaCl_5$-SiO_2, bismuth nitrate penta hydrate, and lithium perchlorate[14–25] are conveniently used. However, many of these catalysts involved in their synthesis required tedious procedures and complex workups. Also, their high preparation cost is of great concern especially with low product yields. Therefore, development of an efficient, inexpensive, and eco-friendly synthesis for them is imperative.

In recent times, nonionic surfactant Tween-20 is used as a catalyst, emulsifier, and complexing agent in both aqueous and nonaqueous media.[26] It is one of the most commonly used detergents in industry as solubilizer with a wide range of applications to biological systems.[27] Tween-20 solubilizes the nucleophilic reagent, as well as enhances the nucleophilicity of the counterions. Besides, the surfactant properties of Tween-20 improve the reaction kinetics by increasing interfacial area. In green chemistry, use of water as solvent has attracted much attention in organic synthesis because of its unique reactivity and selectivity that is not possible in organic solvents.[28] Water is readily available, safest, and the most nontoxic solvent in the chemical world. It is also considered as an ideal solvent due to economical and environmental concerns.[29] However, the possibility of using water as solvent is limited because the majority of reactants are sparingly soluble in water. This solubility problem can be overcome by the use of a surfactant.[30] Surfactants, which are usually organic compounds that are amphiphilic and contain both hydrophobic and hydrophilic groups, have been recently employed to facilitate the reactions ''in water" (http://en.wikipedia.org/wiki/Surfactant). As a part of our continued interest in the development of synthetic methods for the preparation of phosphonate derivatives,[31–33] we found that Tween-20 can be readily used as efficient catalyst for the preparation of α-aminophosphonates in aqueous media.

RESULTS AND DISCUSSION

In order to delineate the standard operating conditions the reaction among 6-nitrobenzo[*d*][1,3]dioxole-5-carbaldehyde, diethylphosphite and aniline was selected as a model (Scheme 1).

At room temperature, there is no sufficient quantity formation of the corresponding α-aminophosphonates in water in the presence of Tween-20 catalyst even after 5 h of reaction time (Table 1, Entry 1). Increase of the reaction temperature from 30 °C to 60 °C led to formation of α-aminophosphonates up to 84% yields (Figure 1). Further increase of temperature did not show any improvement in the yields. This reaction progress is sluggish and did not go to completion even after 12 h of the reaction time in the absence of Tween-20 (Table 1, Entry 7). This shows that Tween-20 is essential and remarkably catalyzes these reactions.



Scheme 1

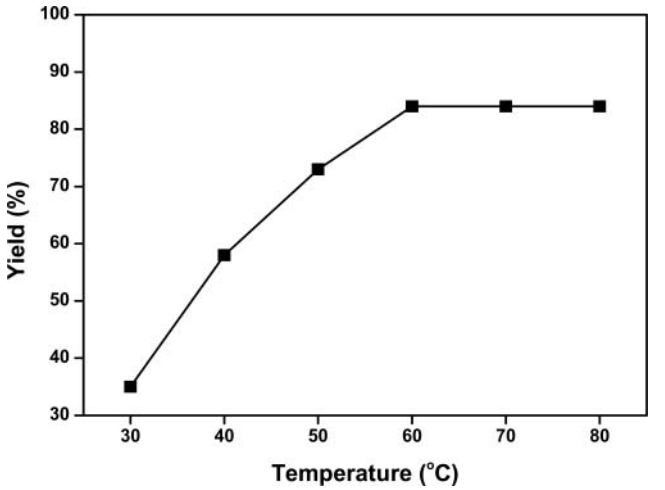


Figure 1 The influence of temperature on the preparation of α-aminophosphonate **4a** by using tween-20 in aqueous media.

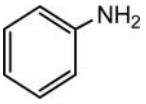
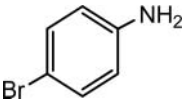
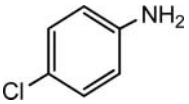
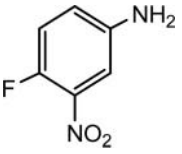
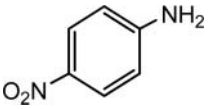
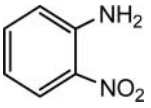
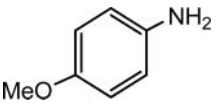
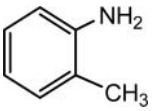
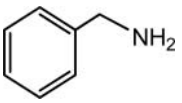
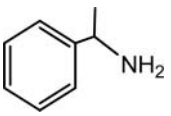
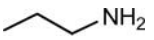
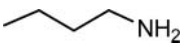
After optimization of experimental conditions with aniline, we extended this three-component coupling reaction with several other amines keeping 6-nitrobenzo[*d*][1,3]dioxole-5-carbaldehyde and diethylphosphite as constant substrates and Tween-20 as catalyst. In all the cases, the reactions were completed within 25–60 min and gave the corresponding α-aminophosphonates in 69%–91% isolated yields. In these reactions, initial formation of imine intermediate is the key step for which further addition of diethylphosphite occurs to form α-aminophosphonates. Observation of the synthetic data (Table 2) reveals that the reaction goes smoothly with aromatic amines (**4a–h**) and affords high yields. Nature and position of the substituent in the aromatic ring of the amines

Table 1 Optimization of reaction conditions for synthesis of **4a**

Entry	Temperature (°C)	Tween-20 (mol%)	Time (min/h)	Yield (%)[a]
1	30/RT	5	5 h	35
2	40	5	30 min	58
3	50	5	30 min	71
4	60	5	30 min	84
5	70	5	30 min	84
6	80	5	30 min	84
7	60	0	12 h	—

[a]Isolated yield.

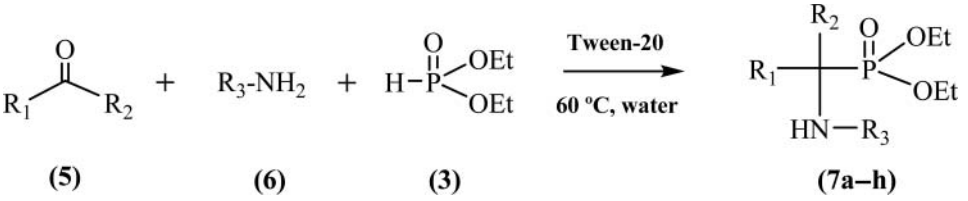
Table 2 Synthesis of α-aminophosphonates (**4a-l**)

Entry	Amine	Product (**4**)[a]	Time (min)	Yield (%)[b]
1		**4a**	30	84
2		**4b**	30	89
3		**4c**	30	87
4		**4d**	30	84
5		**4e**	30	85
6		**4f**	30	82
7		**4g**	25	91
8		**4h**	30	91
9		**4i**	60	74
10		**4j**	60	77
11		**4k**	60	69
12		**4l**	60	71

[a]The products were characterized by NMR, IR, mass and elemental analysis.
[b]Isolated yields.

exert only marginal effect on the reaction rate and product yields. However, in the case of aliphatic amine substrates (**4j–l**), overall reaction times are longer and product yields are significantly low. Perhaps this anomaly maybe attributed to the varied stability of the imine intermediate formed during the course of the reaction. Aromatic imines are resonance stabilized to a larger extent when compared to their aliphatic analogues due to the presence of aromatic ring in conjugation with the imine double bond.

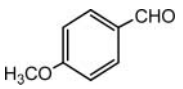
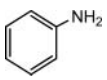
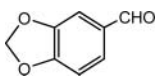
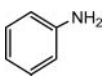
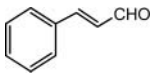
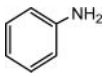
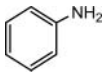
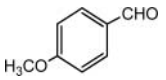
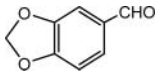
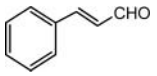
To explore the scope and limitations of this reaction, we extended our protocol to other aromatic and aliphatic carbonyl compounds and aromatic and aliphatic amines under optimized conditions (Scheme 2). The reactions proceeded efficiently with aromatic



Scheme 2

amines when compared with those aliphatic amines as indicated by the data in Table 3. However, the condensation of ketone with both aromatic and aliphatic amines gave lower yields (Table 3, Entries 4, 8). This may perhaps be due to less electrophilic character with

Table 3 Synthesis of α-aminophosphonates from various carbonyl compounds

Entry	Carbonyl Compound (**5**)	Amine (**6**)	Product (**7**)	Time (min)	Yield (%)[a]
1			**7a**	30	85
2			**7b**	30	88
3			**7c**	30	74
4			**7d**	30	62
5			**7e**	60	70
6			**7f**	60	72
7			**7g**	60	64
8			**7h**	60	51

[a]Isolated yields.

carbonyl carbon atom. All the products were characterized by ^{1}H NMR, ^{13}C NMR, IR, and Mass spectral data.

CONCLUSIONS

A simple and environmental friendly protocol for the synthesis of α-aminophosphonates using Tween-20 as catalyst in aqueous media is reported. The major advantages of this procedure are operational simplicity, wide substrate scope, high yields, and the nonuse of hazardous reagents and organic solvents. In many cases, relatively pure products are obtained after workup, which gave analytical samples only on one-column chromatographic run. This method being a more practical one serves as an alternative to the existing synthetic methods for α-aminophosphonates.

EXPERIMENTAL

IR spectra were recorded in KBr pellets on a PerkinElmer 683 spectrophotometer. ^{1}H, ^{13}C, and ^{31}P NMR spectra were taken on Bruker avance 500 MHz spectrometer in $CDCl_3$ using TMS as internal standard. ^{31}P NMR (202.44 MHz) was taken in $CDCl_3$ using 85% H_3PO_4 as external standard with broadband ^{1}H decoupling. EI-Mass spectra were obtained on JEOL GCMATE II GC-MS spectrometer at SAIF, IIT-Madras, Chennai. All chemicals were purchased from Sigma Aldrich and used with out further purification. Double distilled water was used as solvent.

General Procedure for α-Aminophosphonates (4a–l) and (7a–h).

A mixture of aldehyde (1 mmol), amine (1 mmol), diethylphosphite (1 mmol), and Tween-20 (5 mol%) in water (3 mL) was stirred at 60 ∞C. When the reaction was completed (monitored by TLC), the reaction mixture was extracted with CH_2Cl_2. The extract was dried over anhydrous Na_2SO_4 and concentrated under reduced pressure. The crude product obtained was subsequently purified by silica gel column chromatography using EtOAc:hexane (1:4) as eluent.

Physical, Analytical, and Spectral data for the Compounds 4a-j.

Diethyl (6-nitrobenzo[d][1,3]dioxol-5-yl)(phenylamino)methylphosphonate (4a). Brown Solid, Yield: 84%, mp: 175 °C–176 °C. IR (KBr) (ν_{max} cm^{-1}): 3301 (NH), 1525 (N=O), 1262 (P=O), 749 (P–$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.16 (3H, t, J = 7.0 Hz, P–$OCH_2C\underline{H}_3$), 1.29 (3H, t, J = 7.5 Hz, P−$OCH_2C\underline{H}_3$), 3.71–3.79 (1H, m, P−$OC\underline{H}_2CH_3$), 3.94–4.01 (1H, m, P−$OC\underline{H}_2CH_3$), 4.08–4.16 (2H, m, P−$OC\underline{H}_2CH_3$), 4.60 (1H, d, J = 24.0 Hz, P−CH), 5.93 (1H, s, $OC\underline{H}_2O$), 6.46–7.19 (7H, m, Ar−H), 6.77 (1H, s, NH). ^{13}C NMR (125.7 MHz, $CDCl_3$) δ: 16.0 (d, J = 5.9 Hz, P−$OCH_2\underline{C}H_3$), 16.3 (d, J = 5.6 Hz, P−$OCH_2\underline{C}H_3$), 49.6 (d, J = 152.0 Hz, P−CH), 63.5 (d, J = 7.2 Hz, P−$O\underline{C}H_2CH_3$), 63.9 (d, J = 7.0 Hz, P−$O\underline{C}H_2CH_3$), 103.1 (C-6), 107.5 (C-9), 114.6 (C-2^{1} and C-6^{1}), 123.5 (C-4^{1}), 128.9 (C-3), 129.3 (C-3^{1} and C-5^{1}), 131.0 (C-1), 143.4 (C-8), 144.0 (C-4), 147.6 (C-1^{1}), 152.3 (C-2). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 20.60. EI-MS (*m*/*z*,%): 409 (M+1, 20), 408 ($M^{+\bullet}$, 100), 380 (30), 270 (50), 137 (34). Anal. Calcd for $C_{18}H_{21}N_2O_7P$: C, 52.94; H, 5.18; N, 6.86. Found: C, 52.90; H, 5.12; N, 6.76.

Diethyl(4-bromophenylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl)methylphosphonate (4b)

Yellow Solid, Yield: 89%, mp: 181 °C–183 °C. IR (KBr) (ν_{max} cm^{-1}): 3299 (NH), 1525 (N=O), 1260 (P=O), 746 (P–$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.14 (3H, t, J = 7.2 Hz, P–OCH_2CH_3), 1.24 (3H, t, J = 7.6 Hz, P–OCH_2CH_3), 3.76–3.82 (1H, m, P–OCH_2CH_3), 3.89–3.96 (1H, m, P–OCH_2CH_3), 4.04–4.19 (2H, m, P–OCH_2CH_3), 4.84 (1H, d, J = 26.0 Hz, P–CH), 5.94 (2H, s, OCH_2O), 6.50–7.05 (6H, m, Ar–H), 6.93 (1H, s, NH). ^{13}C NMR (125.7 MHz, $CDCl_3$) δ: 16.0 (d, J = 5.9 Hz, P–OCH_2CH_3), 16.3 (d, J = 5.6 Hz, P–OCH_2CH_3), 50.2 (d, J = 152.2 Hz, P–CH), 63.5 (d, J = 7.2 Hz, P–OCH_2CH_3), 63.9 (d, J = 7.0 Hz, P–OCH_2CH_3), 103.1 (C-6), 107.5 (C-9), 110.7 (C-4^1), 115.1 (C-2^1 and C-6^1), 128.9 (C-3), 132.1 (C-3^1 and C-5^1), 143.3 (C-1), 144.4 (C-4), 144.5 (C-8), 147.6 (C-1^1), 152.3 (C-2). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 20.54. EI-MS (*m/z*,%): 488 (M+1, 99), 486 ($M^{+\bullet}$, 100), 459 (30), 457 (30), 350 (60), 348 (55). Anal. Calcd for $C_{18}H_{20}BrN_2O_7P$: C, 44.37; H, 4.14; N, 5.75. Found: C, 44.30; H, 4.09; N, 5.64.

Diethyl(4-chlorophenylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl)methylphosphonate (4c)

Yellow Solid, Yield: 87%, mp: 168 °C–169 °C. IR (KBr) (ν_{max} cm^{-1}): 3292 (NH), 1593 (N=O), 1234 (P=O), 752 (P–$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.05 (3H, t, J = 7.5 Hz, P–OCH_2CH_3), 1.19 (3H, t, J = 6.9 Hz, P–OCH_2CH_3), 3.72–3.94 (2H, m, P–OCH_2CH_3), 4.00–4.12 (2H, m, P–OCH_2CH_3), 5.12 (1H, d, J = 26.4 Hz, P–CH), 6.01 (2H, s, OCH_2O), 6.19 (1H, s, NH), 6.23–7.68 (6H, m, Ar–H). ^{13}C NMR (125.7 MHz, $CDCl_3$) δ: 16.2 (d, J = 5.5 Hz, P–OCH_2CH_3), 16.4 (d, J = 5.9 Hz, P–OCH_2CH_3), 55.7 (d, J = 152.4 Hz, P–CH), 63.3 (d, J = 6.6 Hz, P–OCH_2CH_3), 63.4 (d, J = 7.0 Hz, P–OCH_2CH_3), 103.4 (C-6), 114.6 (C-2^1 and C-6^1), 123.5 (C-9), 128.9 (C-1), 129.3 (C-3^1 and C-5^1), 131.0 (C-3), 139.4 (C-1^1), 143.4 (C-8), 144.0 (C-4), 147.6 (C-4^1), 152.3 (C-2). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 22.02. EI-MS (*m/z*,%): 444 (M+1, 20), 442 ($M^{+\bullet}$, 100), 306 (35), 304 (80), 119 (22), 117 (22). Anal. Calcd for $C_{18}H_{20}ClN_2O_7P$: C, 48.83; H, 4.55; N, 6.33. Found: C, 48.72; H, 4.43; N, 6.22.

Diethyl(4-fluoro-3-nitrophenylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl)methylphosphonate (4d)

White Solid, Yield: 84%, mp: 168 °C–169 °C. IR (KBr) (ν_{max} cm^{-1}): 3310 (NH), 1546 (N=O), 1230 (P=O), 747 (P–$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.12 (3H, t, J = 7.6 Hz, P–OCH_2CH_3), 1.31 (3H, t, J = 7.6 Hz, P–OCH_2CH_3), 3.74–3.82 (1H, m, P–OCH_2CH_3), 3.88–4.12 (1H, m, P–OCH_2CH_3), 4.15–4.30 (2H, m, P–OCH_2CH_3), 4.82 (1H, d, J = 26.0 Hz, P–CH), 5.98 (2H, s, OCH_2O), 6.14–7.65 (5H, m, Ar–H), 6.41 (1H, s, NH). ^{13}C NMR (125.7 MHz, $CDCl_3$) δ: 16.1 (d, J = 5.8 Hz, P–OCH_2CH_3), 16.3 (d, J = 6.2 Hz, P–OCH_2CH_3), 51.7 (d, J = 150.4 Hz, P–CH), 62.4 (d, J = 6.8 Hz, P–OCH_2CH_3), 62.8 (d, J = 7.2 Hz, P–OCH_2CH_3), 101.2 (C-6), 110.4 (C-2^1), 112.5 (C-9), 118.2 (C-5^1), 124.6 (C-6^1), 129.6 (C-1), 130.0 (C-3), 136.4 (C-3^1), 140.2 (C-1^1), 142.4 (C-4^1), 146.6 (C-2), 148.2 (C-4),152.4 (C-8). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 22.10. EI-MS (*m/z*,%): 472 (M+1, 18), 471 ($M^{+\bullet}$, 100), 333 (45). Anal. Calcd for $C_{18}H_{19}FN_3O_9P$: C, 45.87; H, 4.06; N, 8.92. Found: C, 45.79; H, 4.01; N, 8.84.

Diethyl(4-nitrophenylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl)methylphosphonate (4e)

Red Solid, Yield: 85%, mp: 191 °C–192 °C. IR (KBr) (ν_{max} cm^{-1}): 3296 (NH), 1598 (N=O), 1235 (P=O), 751 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.05 (3H, t, J = 7.2 Hz, P−$OCH_2C\underline{H}_3$), 1.19 (3H, t, J = 7.8 Hz, P−$OCH_2C\underline{H}_3$), 3.72–3.92 (2H, m, P−$OC\underline{H}_2CH_3$), 4.02–4.12 (2H, m, P−$OC\underline{H}_2CH_3$), 5.02 (1H, d, J = 26.4 Hz, P−CH), 5.51 (s, 2H, $OC\underline{H}_2O$), 6.17 (1H, s, NH), 6.27–7.65 (6H, m, Ar−H). ^{13}C NMR (125.7 MHz, $CDCl_3$) δ: 16.2 (d, J = 5.7 Hz, P−$OCH_2\underline{C}H_3$), 16.4 (d, J = 5.6 Hz, P−$OCH_2\underline{C}H_3$), 55.8 (d, J = 152.4 Hz, P–CH), 63.2 (d, J = 7.0 Hz, P−$O\underline{C}H_2CH_3$), 63.4 (d, J = 7.0 Hz, P−$O\underline{C}H_2CH_3$), 101.2 (C-6), 108.0 (C-9), 115.5 (C-2^1 and C-6^1), 121.3 (C-4^1), 123.1 (C-3), 129.0 (C-3^1 and C-5^1), 129.2 (C-1), 144.9 (C-4), 147.4 (C-8), 147.6 (C-1^1), 148.0 (C-2). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 21.12. EI-MS (m/z,%): 454 (M+1, 18), 453 ($M^{+\bullet}$, 100), 315 (45). Anal. Calcd for $C_{18}H_{20}N_3O_9P$: C, 47.69; H, 4.45; N, 9.27. Found: C, 47.58; H, 4.38; N, 9.20.

Diethyl(2-nitrophenylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl)methylphosphonate (4f)

Brown Solid, Yield: 82%, mp: 125 °C–127 °C. IR (KBr) (ν_{max} cm^{-1}): 3333 (NH), 1520 (N=O), 1239 (P=O), 762 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.10 (3H, t, J = 8.2 Hz, P−$OCH_2C\underline{H}_3$), 1.22 (3H, t, J = 8.5 Hz, P−$OCH_2C\underline{H}_3$), 3.82–4.09 (2H, m, P−$OC\underline{H}_2CH_3$), 4.14–4.26 (2H, m, P−$OC\underline{H}_2CH_3$), 5.32 (1H, d, J = 25.2 Hz, P−CH), 5.90 (2H, s, $OC\underline{H}_2O$), 6.16 (1H, s, NH), 6.24–8.12 (6H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 24.08. Anal. Calcd for $C_{18}H_{20}N_3O_9P$: C, 47.69; H, 4.45; N, 9.27. Found: C, 47.61; H, 4.40; N, 9.24.

Diethyl(4-methoxyphenylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl)methylphosphonate (4g)

Yellow Solid, Yield: 91%, mp: 181 °C–183 °C. IR (KBr) (ν_{max} cm^{-1}): 3296 (NH), 1555 (N=O), 1235 (P=O), 751 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.16 (3H, t, J = 7.4 Hz, P−$OCH_2C\underline{H}_3$), 1.30 (3H, t, J = 7.8 Hz, P−$OCH_2C\underline{H}_3$), 3.79 (3H, s, Ar−OCH_3), 3.85–3.99 (2H, m, P−$OC\underline{H}_2CH_3$), 4.08–4.17 (2H, m, P−$OC\underline{H}_2CH_3$), 4.92 (1H, d, J = 24.6 Hz, P−CH), 5.94 (2H, s, $OC\underline{H}_2O$), 5.62 (1H, s, NH), 6.75–7.55 (6H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 22.20. Anal. Calcd for $C_{19}H_{23}N_2O_8P$: C, 52.06; H, 5.29; N, 6.39. Found: C, 52.01; H, 5.22; N, 6.32.

Diethyl (6-nitrobenzo[d][1,3]dioxol-5-yl) (o-tolylamino)methylphosphonate (4h)

Bright yellow Solid, Yield: 91%, mp: 157 °C–159 °C. IR (KBr) (ν_{max} cm^{-1}): 3348 (NH), 1547 (N=O), 1242 (P=O), 762 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.02 (3H, t, J = 7.6 Hz, P−$OCH_2C\underline{H}_3$), 1.15 (3H, t, J = 7.6 Hz, P−$OCH_2C\underline{H}_3$), 3.76–3.92 (2H, m, P−$OC\underline{H}_2CH_3$), 3.96–4.14 (2H, m, P−$OC\underline{H}_2CH_3$), 4.80 (1H, d, J = 25.0 Hz, P−CH), 5.04 (2H, s, $OC\underline{H}_2O$), 6.28 (1H s, NH), 6.65–7.58 (6H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 22.18. Anal. Calcd for $C_{19}H_{23}N_2O_7P$: C, 54.03; H, 5.49; N, 6.63. Found: C, 53.98; H, 5.41; N, 6.59.

Diethyl (benzylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl) methylphosphonate (4i)

White Solid, Yield: 74%, mp: 131 °C–133 °C. IR (KBr) (ν_{max} cm^{-1}): 3344 (NH), 1524 (N=O), 1240 (P=O), 760 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.03 (3H, t, J = 8.5 Hz, P−$OCH_2C\underline{H}_3$), 1.19 (3H, t, J = 7.8 Hz, P−$OCH_2C\underline{H}_3$), 3.78–3.92 (2H, m, NCH_2), 3.96–4.10 (2H, m, P−$OC\underline{H}_2CH_3$), 4.17–4.28 (2H, m, P−$OC\underline{H}_2CH_3$), 4.88 (1H, d, J = 24.6 Hz, P−CH), 5.86 (2H, s, $OC\underline{H}_2O$), 6.25 (1H, s, NH), 6.45–7.52 (7H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 20.12. Anal. Calcd for $C_{19}H_{23}N_2O_7P$: C, 54.03; H, 5.49; N, 6.63. Found: C, 54.01; H, 5.44; N, 6.57.

Diethyl(6-nitrobenzo[d][1,3]dioxol-5-yl)(1-phenylethylamino) methylphosphonate (4j)

Brick red Solid, Yield: 77%, mp: 125 °C–127 °C. IR (KBr) (ν_{max} cm^{-1}): 3265 (NH), 1530 (N=O), 1266 (P=O), 752 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.17 (3H, t, J = 8.4 Hz, P−$OCH_2C\underline{H}_3$), 1.31 (3H, t, J = 8.6 Hz, P−$OCH_2C\underline{H}_3$), 1.42 (3H, d, J = 15.8 Hz, $CHC\underline{H}_3$), 3.72–3.85 (2H, m, P−$OC\underline{H}_2CH_3$), 3.92–4.08 (2H, m, P−$OC\underline{H}_2CH_3$), 4.18–4.31 (1H, m, NCH), 4.82 (1H, d, J = 24.2 Hz, P−CH), 5.98 (2H, s, $OC\underline{H}_2O$), 6.46 (1H, s, NH), 6.57–7.82 (7H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 21.92. Anal. Calcd for $C_{20}H_{25}N_2O_7P$: C, 55.05; H, 5.77; N, 6.42. Found: C, 55.01; H, 5.72; N, 6.37.

Diethyl (6-nitrobenzo[d][1,3]dioxol-5-yl)(propylamino) methylphosphonate (4k)

White Solid, Yield: 69%, mp: 144 °C–146 °C. IR (KBr) (ν_{max} cm^{-1}): 3255 (NH), 1565 (N=O), 1254 (P=O), 746 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 0.94 (3H, t, J = 8.6 Hz, $CH_2C\underline{H}_3$), 1.04 (3H, t, J = 6.8 Hz, P−$OCH_2C\underline{H}_3$), 1.18 (3H, t, J = 7.2 Hz, P−$OCH_2C\underline{H}_3$), 1.40-1.56 (2H, m, $CH_2C\underline{H}_2CH_3$), 2.42–2.56 (2H, m, $NHC\underline{H}_2CH_2$), 3.80–3.99 (2H, m, P−$OC\underline{H}_2CH_3$), 4.14–4.26 (2H, m, P−$OC\underline{H}_2CH_3$), 4.86 (1H, d, J = 24.4 Hz, P−CH), 5.99 (2H, s, $OC\underline{H}_2O$), 6.32 (1H, s, NH), 6.82–7.02 (2H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 20.81. Anal. Calcd for $C_{15}H_{23}N_2O_7P$: C, 48.13; H, 6.19; N, 7.48. Found: C, 48.09; H, 6.11; N, 7.40.

Diethyl (butylamino)(6-nitrobenzo[d][1,3]dioxol-5-yl) methylphosphonate (4l)

Brown Solid, Yield: 71%, mp: 187 °C–189 °C. IR (KBr) (ν_{max} cm^{-1}): 3310 (NH), 1525 (N=O), 1230 (P=O), 747 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 0.92 (3H, t, J = 8.8 Hz, $CH_2C\underline{H}_3$), 1.12 (3H, t, J = 7.6 Hz, P−$OCH_2C\underline{H}_3$), 1.22 (3H, t, J = 6.8 Hz, P−$OCH_2C\underline{H}_3$), 1.35–1.40 (2H, m, $CH_2C\underline{H}_2CH_3$), 1.44–1.49 (2H, m, $CH_2C\underline{H}_2CH_2$), 2.55–2.63 (2H, m, $NHC\underline{H}_2CH2$), 3.88–4.12 (2H, m, P−$OC\underline{H}_2CH_3$), 4.15–4.30 (2H, m, P−$OC\underline{H}_2CH_3$), 5.02 (1H, d, J = 25.0 Hz, P−CH), 5.94 (2H, s, $OC\underline{H}_2O$), 6.14 (1H, s, NH), 6.92–7.24 (2H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 21.02. Anal. Calcd for $C_{16}H_{25}N_2O_7P$: C, 49.48; H, 6.49; N, 7.21. Found: C, 49.41; H, 6.35; N, 7.12.

Diethyl (4-methoxyphenyl)(phenylamino)methylphosphonate (7a)

Viscous Liquid, Yield: 85%. IR (KBr) (ν_{max} cm^{-1}): 3312 (NH), 1232 (P=O), 749 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.05 (3H, t, J = 7.2 Hz, P−$OCH_2C\underline{H}_3$), 1.20 (3H, t, J = 7.4 Hz, P−$OCH_2C\underline{H}_3$), 3.52 (3H, s, Ar−OCH_3), 3.72–4.04 (2H, m, P−$OC\underline{H}_2CH_3$), 4.08–4.24 (2H, m, P−$OC\underline{H}_2CH_3$), 4.63 (1H, d, J = 25.2 Hz, P−CH), 5.45 (1H, s, NH), 6.50–7.34 (9H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 20.04. Anal. Calcd for $C_{18}H_{24}NO_4P$: C, 61.88; H, 6.92; N, 4.01. Found: C, 61.76; H, 6.84; N, 3.94.

Diethyl benzo[d][1,3]dioxol-5-yl(phenylamino)methylphosphonate (7b)

Yellow Solid, Yield: 88%, mp: 162 °C–164 °C. IR (KBr) (ν_{max} cm^{-1}): 3308 (NH), 1264 (P=O), 755 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.12 (3H, t, J = 7.8 Hz, P−$OCH_2C\underline{H}_3$), 1.18 (3H, t, J = 7.4 Hz, P−$OCH_2C\underline{H}_3$), 3.92–4.10 (2H, m, P−$OC\underline{H}_2CH_3$), 4.14–4.20 (2H, m, P−$OC\underline{H}_2CH_3$), 4.52 (1H, d, J = 24.8 Hz, P−CH), 5.45 (2H, s, $OC\underline{H}_2O$), 6.77 (1H, s, NH), 6.94–8.45 (8H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 21.15. Anal. Calcd for $C_{18}H_{22}NO_5P$: C, 59.50; H, 6.10; N, 3.85. Found: C, 59.45; H, 6.01; N, 3.74.

(E)-Diethyl 3-phenyl-1-(phenylamino)allylphosphonate (7c)

White Solid, Yield: 74%, mp: 103 °C–105 °C. IR (KBr) (ν_{max} cm^{-1}): 3328 (NH), 1258 (P=O), 759 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.09 (3H, t, J = 6.8 Hz, P−$OCH_2C\underline{H}_3$), 1.21 (3H, t, J = 7.2 Hz, P−$OCH_2C\underline{H}_3$), 3.54–3.78 (2H, m, P−$OC\underline{H}_2CH_3$), 3.90-4.12 (2H, m, P−$OC\underline{H}_2CH_3$), 5.02 (1H, d, J = 24.8 Hz, P−CH), 5.45 (1H, d, J = 7.8 Hz, Ar−CH = C$\underline{H}$), 5.68 (1H, d, J = 9.2 Hz, Ar−C$\underline{H}$ = CH), 6.12 (1H, s, NH), 7.14–8.28 (10H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 20.64. Anal. Calcd for $C_{19}H_{24}NO_3P$: C, 66.07; H, 7.00; N, 4.06. Found: C, 66.00; H, 6.91; N, 3.96.

Diethyl 1-(phenylamino)cyclohexylphosphonate (7d)

White Solid, Yield: 62%, mp: 94 °C–96 °C. IR (KBr) (ν_{max} cm^{-1}): 3324 (NH), 1257 (P=O), 758 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.09 (3H, t, J = 7.0 Hz, P−$OCH_2C\underline{H}_3$), 1.18 (3H, t, J = 7.4 Hz, P−$OCH_2C\underline{H}_3$), 1.57–1.64 (2H, m, $CH_2C\underline{H}_2CH_2$), 1.66–174 (2H, m, $C\underline{H}_2CH_2CH_2$), 1.78–1.87 (2H, m, $CH_2CH_2C\underline{H}_2$), 1.90 (2H, t, J = 9.2 Hz, CH_2CO), 1.96 (2H, t, J = 9.0 Hz, CH_2CO), 3.71–3.89 (2H, m, P−$OC\underline{H}_2CH_3$), 4.01–4.16 (2H, m, P−$OC\underline{H}_2CH_3$), 5.42 (1H, d, J = 24.4 Hz, P−CH), 6.41 (1H, s, NH), 6.52–7.52 (5H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 21.42. Anal. Calcd for $C_{16}H_{26}NO_3P$: C, 61.72; H, 8.42; N, 4.50. Found: C, 61.64; H, 8.36; N, 4.42.

Diethyl (4-methoxyphenyl)(propylamino)methylphosphonate (7e)

Viscous Liquid, Yield: 70%, IR (KBr) (ν_{max} cm^{-1}): 3345 (NH), 1254 (P=O), 751 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 0.91 (3H, t, J = 8.4 Hz, $CH_2C\underline{H}_3$), 1.08 (3H, t, J = 7.4 Hz, P−$OCH_2C\underline{H}_3$), 1.18 (3H, t, J = 7.2 Hz, P−$OCH_2C\underline{H}_3$), 1.45–1.59 (2H, m, $CH_2C\underline{H}_2CH_3$), 2.52–2.64 (2H, m, $NHC\underline{H}_2CH_2$), 3.65 (3H, s, Ar−OCH_3), 3.82–3.96 (2H, m, P−$OC\underline{H}_2CH_3$), 4.04–4.19 (2H, m, P−$OC\underline{H}_2CH_3$), 5.07 (1H, d, J = 24.2 Hz, P−CH), 6.45 (1H, s, NH), 6.99–7.68 (4H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 21.12. Anal. Calcd for $C_{15}H_{26}NO_4P$: C, 57.13; H, 8.31; N, 4.44. Found: C, 57.07; H, 8.24; N, 4.32.

Diethyl benzo[d][1,3]dioxol-5-yl(propylamino)methylphosphonate (7f)

White Solid, Yield: 72%, mp: 132 °C–134 °C. IR (KBr) (ν_{max} cm^{-1}): 3255 (NH), 1254 (P=O), 746 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 0.98 (3H, t, J = 8.8 Hz, $CH_2C\underline{H}_3$), 1.02 (3H, t, J = 7.4 Hz, P−$OCH_2C\underline{H}_3$), 1.12 (3H, t, J = 7.8 Hz, P−$OCH_2C\underline{H}_3$), 1.48–1.60 (2H, m, $CH_2C\underline{H}_2CH_3$), 2.52–2.68 (2H, m, $NHC\underline{H}_2CH_2$), 3.74–3.92 (2H, m, P−$OC\underline{H}_2CH_3$), 4.04–4.20 (2H, m, P−$OC\underline{H}_2CH_3$), 5.12 (1H, d, J = 22.8 Hz, P−CH), 6.07 (2H, s, $OC\underline{H}_2O$), 6.45 (1H, s, NH), 6.84–7.14 (3H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 22.08. Anal. Calcd for $C_{15}H_{24}NO_5P$: C, 54.71; H, 7.35; N, 4.25. Found: C, 54.64; H, 7.28; N, 4.17.

(E)-Diethyl 3-phenyl-1-(propylamino)allylphosphonate (7g)

Yellow Solid, Yield: 64%, mp: 103 °C–105 °C. IR (KBr) (ν_{max} cm^{-1}): 3328 (NH), 1258 (P=O), 759 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.02 (3H, t, J = 8.4 Hz, $CH_2C\underline{H}_3$), 1.12 (3H, t, J = 7.0 Hz, P−$OCH_2C\underline{H}_3$), 1.22 (3H, t, J = 7.2 Hz, P−$OCH_2C\underline{H}_3$), 1.44–1.54 (2H, m, $CH_2C\underline{H}_2CH_3$), 2.47–2.56 (2H, m, $NHC\underline{H}_2CH_2$), 3.85–3.99 (2H, m, P−$OC\underline{H}_2CH_3$), 4.07–4.22 (2H, m, P−$OC\underline{H}_2CH_3$), 4.52 (1H, d, J = 24.8 Hz, P−CH), 5.57 (1H, d, J = 7.6 Hz, Ar−CH = C$\underline{H}$), 5.99 (1H, d, J = 10.4 Hz, Ar−C$\underline{H}$ = CH), 6.44 (1H, s, NH), 7.17–8.47 (5H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 21.14. Anal. Calcd for $C_{16}H_{26}NO_3P$: C, 61.72; H, 8.42; N, 4.50. Found: C, 61.65; H, 8.37; N, 4.44.

Diethyl 1-(propylamino)cyclohexylphosphonate (7h)

White Solid, Yield: 51%, mp: 129 °C–131 °C. IR (KBr) (ν_{max} cm^{-1}): 3339 (NH), 1229 (P=O), 745 (P−$C_{aliphatic}$). ^{1}H NMR (500 MHz, $CDCl_3$) δ: 1.02 (3H, t, J = 8.0 Hz, $CH_2C\underline{H}_3$), 1.09 (3H, t, J = 7.4 Hz, P−$OCH_2C\underline{H}_3$), 1.18 (3H, t, J = 7.6 Hz, P−$OCH_2C\underline{H}_3$), 1.40–1.64 (2H, m, $CH_2C\underline{H}_2CH_3$), 1.68–1.74 (2H, m, $CH_2C\underline{H}_2CH_2$), 1.78–1.84 (2H, m, $C\underline{H}_2CH_2CH_2$), 1.85–1.90 (2H, m, $CH_2CH_2C\underline{H}_2$), 1.94 (2H, t, J = 9.2 Hz, CH_2CO), 1.99 (2H, t, J = 9.8 Hz, CH_2CO), 2.51–2.68 (2H, m, $NHC\underline{H}_2CH_2$), 3.88–4.12 (2H, m, P−$OC\underline{H}_2CH_3$), 4.18–4.42 (2H, m, P−$OC\underline{H}_2CH_3$), 5.24 (1H, d, J = 24.0 Hz, P−CH), 6.32 (1H, s, NH), 6.43–8.12 (5H, m, Ar−H). ^{31}P NMR (202.4 MHz, $CDCl_3$) δ: 20.32. Anal. Calcd for $C_{13}H_{28}NO_3P$: C, 56.30; H, 10.18; N, 5.05. Found: C, 56.24; H, 10.11; N, 4.97.

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