

Green and Cost-Effective Synthesis of Sulfamidophosphonates Using ZnO Nanoparticles as Catalyst [†]

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Abstract: A simple and efficient protocol for the one-pot three-component synthesis of structurally diverse sulfamidophosphonates from the condensation of sulfanilamide, aldehydes, and triethylphosphite in ethanol using ZnO nanoparticles as catalysts under microwave irradiation has been developed. The structures of all compounds have been identified by appropriate spectroscopic methods such as FTIR, ¹H, ¹³C, ³¹P NMR and ESI-MS.

Keywords: sulfamidophosphonate; microwave; ZnO nanoparticles; catalyst

1. Introduction

Significant factors that are intimately linked with the advancement of ecologically sustainable methods include the concept of atom economy, which pertains to the maximization of the utilization of all reactants employed in a process, along with a focus on enhancing overall efficiency. Additionally, there is a need to eliminate toxic intermediates/products from such processes while simultaneously minimizing the production of waste to the greatest extent possible [1–3]. Multicomponent reactions (MCRs) have arisen as a compelling technique in this regard, enabling the facile synthesis of elaborate molecules through a one-pot approach, devoid of the need for intermediate isolation and purification. This affords a reduction in expenses, as well as time and energy consumption, making it a fascinating tool for organic synthesis [4–6].

Furthermore, the use of environmentally sustainable energy sources for the promotion of chemical reactions has a prominent role [7]. In this regard, the use of microwave irradiation in synthesis generally leads to decreased reaction time, enhanced yield and selectivity, as well as the facilitation of organic transformations that would otherwise demand severe temperature and pressure conditions [8–10].

However, the use of heterogeneous catalysts in organic synthesis has attained a notable degree of significance. This is due to the fact that not only do they facilitate environmentally sustainable syntheses, but they also yield a high percentage of products with exceptional selectivity. Various accounts have substantiated the outstanding performance of nanoparticles as heterogeneous catalysts in multicomponent reactions with regard to their selectivity, reactivity, and augmented product yields [11].

Among various metal nanostructures, nanoparticles (ZnO-NPs) have received significant attention due to their remarkable properties and potential applications in diverse fields [12]. ZnO-NPs, in particular, exhibit high availability and can produce high product yields in short reaction times with only mild reaction conditions, as compared to conventional catalysts. Moreover, they can be readily recycled [13]. ZnO-NPs have been utilized as an active catalyst in several reactions, such as the synthesis of coumarins through Knoevenagel condensation [14] and the synthesis of functionalized benzenes [15].



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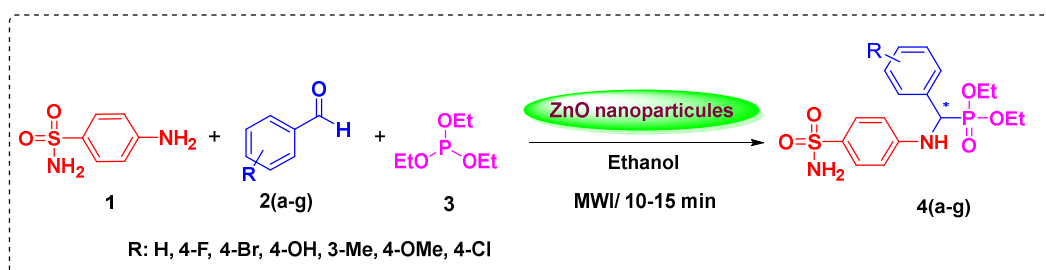
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In the current study, we described the efficient use of ZnO-NPs for the synthesis of sulfamidophosphonate derivatives with optimal duration and yields.

2. Results and Discussion

To facilitate the advancement of our investigations in the field of the synthesis of new compounds containing sulfonamide and phosphonate moieties, we are keen to explore the synthesis of novel derivatives of α -sulfamidophosphonate through a one-pot Kabachnik–Field’s reaction. Our approach involves the use of a green, clean eco-friendly method using microwave irradiation in the presence of zinc oxide nanoparticles [16–18] as a reusable and heterogeneous catalyst.

A series of new α -sulfamidophosphonate derivatives has been successfully synthesized with remarkable efficiency via a one-pot, three-component Kabachnik–Field’s reaction. The reaction involved the use of sulfanilamide (**1**) with various aromatic aldehydes (**2a–g**) and trialkyl phosphite (**3**) under microwave irradiation catalyzed by catalytic quantities of (ZnO-NPs) in ethanol. The reaction was completed with excellent yields (Scheme 1).



Scheme 1. Synthesis of α -sulfamidophosphonate derivatives.

The first stage of this work involved the refinement of the reaction parameters, encompassing the identification of suitable solvents, catalysts, and temperatures. The results of optimization are shown in Table 1. In this work, the effects of some solvents were also investigated for the production of **4(a–g)**.

Table 1. Optimization for the synthesis of α -sulfamidophosphonates with/without ZnO NPs.

Entry	Microwave				Microwave with ZnO NPs			
	Solvent	Time/min	Temp/°C	Yields %	Time/min	Temp/°C	Yields %	Catalyst mol %
1	No solvent	30	100	-	30	100	20	10
2	CH ₂ Cl ₂	20	100	40	20	100	55	10
3	MeOH	20	100	50	20	100	60	10
4	Acetone	20	100	40	20	100	53	10
5	EtOH	15	100	60	15	100	93	10

Microwave irradiation was employed to investigate the reaction efficiency of various solvents at a temperature of 100 °C. It was observed that the reaction yields were significantly low in solvents such as CH₂Cl₂, MeOH, acetone, and in the absence of solvent. In contrast, the employment of EtOH as the solvent resulted in a substantially higher yield of 60%, as demonstrated in Table 1.

On the other hand, under the same conditions, the reaction was carried out in the presence of 10 mol% of ZnO-NPs as a green catalyst and the product was obtained in 93% yield after 15 min.

The results show that in the EtOH, the yields are higher than the other solvents.

According to the results of the optimization reported in Table 1, we observed that catalytic use of ZnO-NP (10% molars), and EtOH as solvent under microwave irradiations were estimated as the optimal reaction conditions.

The reaction between sulfanilamide **1**, benzaldehyde **2a**, and triethylphosphite **3** was selected as a model to evaluate the feasibility of α -sulfamidophosphonates and to optimize the reaction conditions.

The structures of the synthesized compounds are confirmed by elemental analysis as well as by IR and ^1H , ^{13}C , and ^{31}P NMR spectral data.

The ^{31}P NMR spectrum of compound **4a** demonstrated a single peak at a chemical shift $\delta = 22.19$ ppm.

In the ^1H NMR spectrum, a deshielded doublet of doublets at $\delta = [5.0\text{--}5.30]$ ppm was consistently observed, which corresponds to $\text{NH}^*\text{CH}(\text{R})\text{PO}(\text{OEt})_2$. The two CH_2 groups of the mustard moiety detected at $\delta = [4.14\text{--}3.87]$ and $[3.94\text{--}3.63]$.

The FT-IR spectrum displayed a distinctive absorption band around $[3351.92\text{--}3286.65] \text{ cm}^{-1}$, which corresponds to the NH group, while the sulfamide group exhibited signals at $[1153.18\text{--}1147.64] \text{ cm}^{-1}$ and $[1327.16\text{--}1310.95] \text{ cm}^{-1}$. Additionally, the phosphonate group appeared around $[1229.42\text{--}1205.92] \text{ cm}^{-1}$.

The ^{13}C NMR spectrum presented characteristic doublets related to the presence of phosphorus ($\text{J}_{\text{C-P}}$ couplings), while the two ethoxy groups of phosphonate moiety were identified at $[16.37\text{--}15.94]$ ppm ($\text{J}_{\text{C-P}} \sim 5.1\text{--}5.8$ Hz) and $[62.95\text{--}61.17]$ ppm ($\text{J}_{\text{C-P}} \sim 6.6\text{--}7$ Hz). The asymmetric carbon $\text{NHCH}(\text{R}) \text{PO}(\text{OEt})_2$ was observed at $[50.51\text{--}54.26]$ ppm, exhibiting a doublet with a large coupling constant of $\text{J}_{\text{C-P}} \sim 150.6\text{--}155$ Hz.

3. General Procedure for the Synthesis of Sulfamidophosphonate Derivatives

A mixture of sulfanilamide (**1**) (1 mmol) different substituted aromatic aldehydes (**2a–g**) (1 mmol) and triethyl phosphite (**3**) (1 mmol) in the presence of ZnO nanoparticles catalyst (10 mol %) and EtOH as solvent were laid in a flat-bottom flask and irradiated with MWI. Movement of the reaction was observed by TLC experiment dichloromethane-methanol (99/1) for every 2mins. After completion of the reaction, the reaction mixture was filtered, and the catalyst was washed with ethyl acetate. The solvent was evaporated from the mixture and the residue was purified by Et_2O to afford the pure α -sulfamidophosphonates in excellent yields. All other compounds were prepared by the same procedure.

4. Conclusions

In summary, facile and greener synthetic routes were developed for the synthesis of novel α -sulfamidophosphonates using ZnO-NPs as a catalyst. A synthetic approach based on a one-pot, three-component Kabachnik–Field's reaction was devised, which used commercially available starting materials (Appendix A).

Supplementary Materials: The presentation materials can be downloaded at: <https://www.mdpi.com/article/10.3390/ECP2023-14733/s1>.

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Conflicts of Interest: The authors declare that there is no conflict of interest.

Appendix A

diethyl (phenyl((4-sulfamoylphenyl)amino)methyl)phosphonate (4a)

White powder, 93% yield, m.p. 198–200 °C, Rf = 0.22 (CH₂Cl₂/MeOH: 96/4). **IR** (KBr): 3342.05, 1149.70–1319.94, 1227.04 cm^{−1}. **¹H NMR** (400 MHz, DMSO) δ 1.05 (t, *J* = 7 Hz, 3H, CH₃), 1.18 (t, *J* = 7 Hz, 3H, CH₃), 3.76–3.72 (m, 1H, CH₂), 3.92–3.86 (m, 1H, CH₂), 4.08–4.00 (m, 1H, CH₂), 5.15 (dd, 1H, *CH), 6.89 (d, *J* = 8.8 Hz, 4H, H_{Ar}), 7.09–7.05 (m, 1H, H_{Ar}), 7.26–7.23 (m, 1H, NH), 7.33 (t, *J* = 12 Hz, 2H, H_{Ar}), 7.44 (d, *J* = 8.8 Hz, 2H, NH₂), 7.54–7.52 (m, 2H, H_{Ar}) ppm. **³¹P NMR** (100 MHz, DMSO) δ 22.19 ppm. **¹³C NMR** (101 MHz, DMSO) δ 150.09, 136.26, 131.51, 128.24, 128.01, 127.49, 126.91, 112.36, 62.49, 62.30, 53.44, 16.25, 16.00, ppm. Anal. Calcd for C₁₇H₂₃N₂O₅PS C, 51.25; H, 5.82; N, 7.03. Found: C, 51.30; H, 5.70; N, 7.10.

diethyl ((4-fluorophenyl)((4-sulfamoylphenyl)amino)methyl)phosphonate (4b)

Yellow powder, 91% yield, m.p. 168–170 °C, Rf = 0.22 (CH₂Cl₂/MeOH: 96/4). **IR** (KBr): 3332.54, 1149.16–1318.92, 1225.03 cm^{−1}. **¹H NMR** (400 MHz, DMSO) δ 1.06 (t, *J* = 7.1 Hz, 3H, CH₃), 1.17 (t, *J* = 7.0 Hz, 3H, CH₃), 3.82–3.73 (m, 1H, CH₂), 3.95–3.87 (m, 1H, CH₂), 4.04 (dq, *J* = 11.2, 7.0, 2.8 Hz, 2H, CH₂), 5.20 (dd, 1H, *CH), 6.93–6.85 (m, 4H, H_{Ar}), 7.20–7.04 (m, 3H, H_{Ar}), 7.45 (d, *J* = 7.1 Hz, 2H, NH₂), 7.56 (ddd, *J* = 7.6, 5.4, 2.2 Hz, 2H, H_{Ar}) ppm. **¹³C NMR** (101 MHz, DMSO) δ 162.95, 160.53, 150.09, 132.58, 131.74, 130.38–130.25, 127.11, 115.17–114.93, 62.81, 62.57, 52.75, 16.35, 16.15 ppm. Anal. Calcd for C₁₇H₂₂FN₂O₅PS: C, 49.04; H, 5.33; N, 6.73. Found: C, 49.15; H, 5.40; N, 6.80.

diethyl ((4-bromophenyl)((4-sulfamoylphenyl)amino)methyl)phosphonate (4c)

White powder, 88% yield, m.p. 172–174 °C, Rf = 0.24 (CH₂Cl₂/MeOH: 96/4). **IR** (KBr): 3351.92, 1150.03–1322.47, 1225.53 cm^{−1}. **¹H NMR** (400 MHz, DMSO) δ 1.08 (t, *J* = 7.1 Hz, 3H, CH₃), 1.18 (t, *J* = 7.0 Hz, 3H, CH₃), 3.87–3.74 (m, 1H, CH₂), 3.98–3.87 (m, 1H, CH₂), 4.12–3.99 (m, 2H, CH₂), 5.20 (dd, 1H, *CH), 6.95–6.81 (m, 4H, H_{Ar}), 7.07 (dd, *J* = 9.7, 6.6 Hz, 1H, NH), 7.40 (d, *J* = 8.4 Hz, 2H, H_{Ar}), 7.46 (d, *J* = 8.9 Hz, 2H, NH₂), 7.54 (dd, *J* = 8.6, 2.2 Hz, 2H, H_{Ar}) ppm. **¹³C NMR** (101 MHz, DMSO) δ 150.02, 135.54, 132.34, 131.80, 128.19, 127.09, 62.85, 62.59, 52.87, 16.35, 16.15 ppm. Anal. Calcd for C₁₇H₂₂BrN₂O₅PS C, 42.78; H, 4.65; N, 5.87. Found: C, 42.85; H, 4.57; N, 5.77.

diethyl ((4-methoxyphenyl)((4-sulfamoylphenyl)amino)methyl)phosphonate (4d)

White powder, 89% yield, m.p. 158–160 °C, Rf = 0.22 (CH₂Cl₂/MeOH: 96/4). **IR** (KBr): 3317.19, 1153.18–1310.95, 1223.14 cm^{−1}. **¹H NMR** (400 MHz, DMSO) δ 1.07 (t, *J* = 7.0 Hz, 3H, CH₃), 1.18 (t, *J* = 7.0 Hz, 3H, CH₃), 3.72 (s, 4H, CH₂ + OCH₃), 3.94–3.82 (m, 1H, CH₂), 4.14–3.94 (m, 2H, CH₂), 5.15 (dd, 1H, *CH), 6.95–6.77 (m, 6H, H_{Ar}), 7.02 (dd, *J* = 9.8, 6.4 Hz, 1H, NH), 7.44 (dd, *J* = 8.9, 2.0 Hz, 4H, NH₂ + H_{Ar}) ppm. **¹³C NMR** (101 MHz, DMSO) δ 158.68, 150.15, 131.42, 129.43, 127.949, 126.902, 113.51, 112.40, 62.42, 62.27, 54.26, 16.30, 16.09 ppm. Anal. Calcd for C₁₈H₂₅N₂O₆PS C, 50.46; H, 5.88; N, 6.54. Found: C, 50.51; H, 5.80; N, 6.62.

diethyl ((4-hydroxyphenyl)((4-sulfamoylphenyl)amino)methyl)phosphonate (4e)

White powder, 91% yield, m.p. 90–92 °C, Rf = 0.11 (CH₂Cl₂/MeOH: 96/4). **IR** (KBr): 3343.79, 1147.64–1323.66, 1219.42 cm^{−1}. **¹H NMR** (400 MHz, DMSO) δ 1.06 (t, *J* = 7.0 Hz, 3H, CH₃), 1.18 (t, *J* = 7.0 Hz, 3H, CH₃), 3.76–3.67 (m, 1H, CH₂), 3.88 (dt, *J* = 10.3, 7.2 Hz, 1H, CH₂), 4.06–4.00 (m, 2H, CH₂), 5.0 (dd, 1H, *CH), 6.71 (d, *J* = 8.6 Hz, 2H), 6.92–6.84 (m, 4H, H_{Ar}), 6.97 (dd, *J* = 9.8, 6.2 Hz, 1H, NH), 7.32 (dd, *J* = 8.7, 2.2 Hz, 2H, H_{Ar}), 7.45 (d, *J* = 8.9 Hz, 2H, NH₂), 9.38 (s, 1H, OH) ppm. **¹³C NMR** (101 MHz, DMSO) δ 156.54, 150.21, 131.32, 129.42, 126.91, 126.10, 114.89, 112.36, 62.30, 61.17, 52.86, 16.31, 16.09 ppm. Anal. Calcd for C₁₇H₂₃N₂O₆PS: C, 49.27; H, 5.59; N, 6.76. Found: C, 49.35; H, 5.67; N, 6.84.

diethyl (((4-sulfamoylphenyl)amino)(m-tolyl)methyl)phosphonate (4f)

White powder, 90% yield, m.p. 194–196 °C, Rf = 0.21 (CH₂Cl₂/MeOH: 96/4). **IR** (KBr): 3336.3, 1149.46–1315.53, 1208.35 cm^{−1}. **¹H NMR** (400 MHz, DMSO) δ 1.06 (t, *J* = 7.0 Hz,

3H, CH₃), 1.18 (t, *J* = 7.0 Hz, 3H, CH₃), 2.28 (s, 3H, CH₃), 3.78–3.64 (m, 1H, CH₂), 3.94–3.82 (m, 1H, CH₂), 4.12–3.98 (m, 2H, CH₂), 5.10 (dd, 1H, *CH), 6.94–6.83 (m, 4H, H_{Ar}), 7.13–7.01 (m, 2H, H_{Ar}), 7.21 (t, *J* = 7.6 Hz, 1H, NH), 7.38–7.30 (m, 2H, H_{Ar}), 7.48–7.42 (m, 2H, NH₂) ppm. ¹³C NMR (101 MHz, DMSO) δ 150.17, 137.09, 136.21, 131.47, 128.81, 128.24, 127.94, 127.41, 126.96, 125.43, 62.51, 62.36, 53.38, 16.30, 16.03 ppm. Anal. Calcd for C₁₈H₂₅N₂O₅PS: C, 52.42; H, 6.11; N, 6.79. Found: C, 52.54; H, 6.21; N, 6.87.

diethyl ((4-chlorophenyl)((4-sulfamoylphenyl)amino)methyl)phosphonate (**4g**)

White powder, 89% yield, m.p.178–180 °C, R_f = 0.20 (CH₂Cl₂/MeOH: 96/4). IR (KBr): 3286.65, 1148.98–1327.16, 1205.50 cm^{−1}. ¹H NMR (400 MHz, DMSO) δ 1.02 (t, *J* = 7.0 Hz, 3H, CH₃), 1.22 (t, *J* = 7.0 Hz, 3H, CH₃), 3.78–3.63 (m, 1H, CH₂), 3.88 (dt, *J* = 10.5, 7.5 Hz, 1H, CH₂), 4.11 (p, *J* = 7.1 Hz, 2H, CH₂), 5.30 (dd, 1H, *CH), 6.76 (d, *J* = 8.9 Hz, 2H, H_{Ar}), 6.92 (s, 2H, H_{Ar}), 7.38–7.22 (m, 3H, H_{Ar}), 7.53–7.42 (m, 3H, NH₂ + H_{Ar}), 7.66 (d, *J* = 7.5 Hz, 1H, NH) ppm. ¹³C NMR (101 MHz, DMSO) δ 149.61, 133.93, 133.38, 132.17, 129.60, 129.16, 127.36, 112.11, 62.94, 62.67, 50.51, 16.30, 15.94 ppm Anal. Calcd for C₁₇H₂₂ClN₂O₅PS: C, 47.17; H, 5.12; N, 6.47. Found: C, 47.25; H, 5.08; N, 6.55.

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