

Research Article

Electronic Impacts on the Solvent-Free Mechanochemical Synthesis of Salophen Ligands in Ball Mill

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To realize the scope of the solid-solid reaction, the functional solid diamines reacted with solid 3-ethoxysalicylaldehyde under high-speed ball milling in a solvent-free environment. The findings showed that a wide range of Salophen ligands could be produced in good to excellent yields. In comparison to the similar Salophen synthesis to date, the current study provided solvent-free, fast reaction, high yield, and easy work-up.

1. Introduction

Azomethine is a type of organic compound that can be used in a variety of chemical processes. The imine functionality allows it to be a major ligand for a variety of chelated compounds in chemical synthesis [1–3]. Salen and Salophen ligands are particularly appealing among these ligands. Metal-Salen and Salophen complexes have been intensively explored in medicinal [4–6] as well as material [7–9] chemistry in addition to their well-known catalytic characteristics in organic synthesis [10, 11] and chemo sensing applications [12–15]. The ability of tetradentate Salophen ONNO ligands to coordinate a wide range of transition metals in various oxidation states explains their appeal.

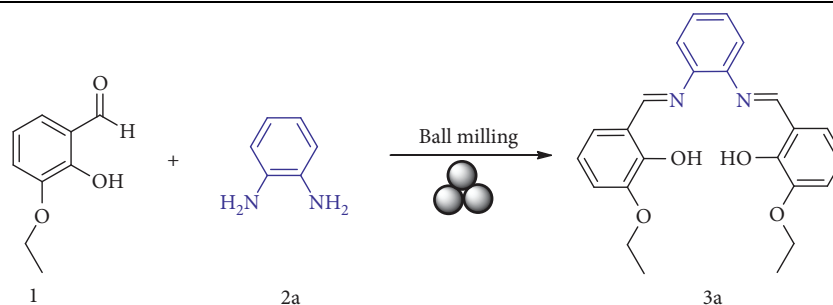
The conversion of carbonyl compounds, particularly aldehydes and amines, into the corresponding azomethine has been carried out in a variety of ways, with encouraging results. Previously, we made simple imine molecules from solid aldehyde and solid primary aromatic amine in a 1:1 ratio in mortar and pestle in solvent-free conditions [16, 17]. The manual grinding procedure, however, has limitations in terms of consistency and large-scale output. Furthermore, reports in the synthesis techniques employed in the Salophen ONNO ligand revealed that the synthesis was done in either solution media [18–22] or solvent-free [7, 23] with at least one of the reactants in the liquid state. However, the

former requires catalyzed, time-consuming, expensive, and environmentally harmful synthetic effort, whereas the latter reactions are aided by liquid reactants and so do not reveal the entire extent of the solid-solid reaction. A systematic examination of Salophen ligand synthesis by solvent-free mechanochemical ball mill reaction in various electronic environments remains unexplored as well.

Recently, the use of microwaves [24], ultrasound [25], ionic liquids [26], and deep eutectic solvents [27] has proven successful in the synthesis of azomethine. Nevertheless, the synthetic community has been interested in mechanochemical transformation employing ball milling in recent years [1]. Mechanochemical ball milling has emerged as a viable choice in chemical synthesis because of its several advantages, including solvent-free synthesis, increased pace, scalability, the ability to adjust reaction parameters, unusual selectivity, and so on [28–31].

As a result, in this paper, a broad range of high-speed ball mill parameter optimization was covered. A comprehensive examination of the electronic influence on solvent-free mechanochemical Salophen synthesis utilizing solid reactants exclusively with different functionality was presented. Finally, the comparative outlook was discussed to show the state of the art in Salophen synthesis.

The optimization process was tracked by looking at how reactant consumption and product creation changed as the

TABLE 1: Optimization of the mechanochemical ball milling conditions for Salophen synthesis^a.

Entry	Time (min)	Frequency (Hz)	Ball type	Ball size (mm)	Ball number	Jar type (10 ml)	Yield (%) ^b
1	20	30	SS	10	1	Teflon	64
2	40	30	SS	10	1	Teflon	72
3	60	30	SS	10	1	Teflon	90 [86 ^c]
4	80	30	SS	10	1	Teflon	74
5	100	30	SS	10	1	Teflon	80
6	120	30	SS	10	1	Teflon	74
7	60	25	SS	10	1	Teflon	63
8	60	20	SS	10	1	Teflon	64
9	60	15	SS	10	1	Teflon	62
10	60	10	SS	10	1	Teflon	45
11	60	30	ZrO ₂	10	1	Teflon	78
12	60	30	WC	10	1	Teflon	63
13	60	30	SS	Blank	1	Teflon	27
14	60	30	SS	5	1	Teflon	67
15	60	30	SS	7	1	Teflon	67
16	60	30	SS	12	1	Teflon	75
17	60	30	SS	5	2	Teflon	69
18	60	30	SS	7	2	Teflon	64
19	60	30	SS	7	3	Teflon	64
20	60	30	SS	10	2	Teflon	61
21	60	30	SS	10	1	SS	61

^aReaction conditions: 1 mmol (1) and 1 mmol (2a) were milled in a mixer mill; ^byield based on ¹H NMR analysis of the crude reaction mixture taken directly from the milling jar; ^cisolated yield; SS: stainless steel.

vibratory milling parameters changed. The production of azomethine proton as well as the decay of aldehyde proton in the ¹H NMR spectra of the product mixture was used to monitor the rate of azomethine creation.

The reaction progress was measured every 20 minutes for 2 hours to determine the influence of reaction time, and the yield increased till 60 minutes, then decreased at 80 minutes, and then began to climb at 100 minutes (Figure S1). This is most likely the imine that resulted from the reaction's hydrolysis in the container after 60 minutes. It is vital to remember that imine formation is an equilibrium reaction that moves in the opposite way when there is a lot of water around. This could be due to the water by-product not being able to evaporate and so remaining in the vessel, favoring the reverse reaction [32]. As a result, the ideal time for the reaction is 60 minutes.

The experiment to look into the effect of ball size on yield under the given reaction circumstances found that a 10 mm ball is the best size for excellent yield. The yield grows as the ball size increases until it reaches 10 mm, at which point it decreases (Figure S2). Similarly, when one 5 mm ball was compared to two 5 mm balls in the

container to demonstrate the number of ball effect under the given reaction circumstances, there was no significant effect on the yield. Even when using a variable number of 7 mm balls, comparable yields were obtained. Under similar reaction conditions, two and three 7 mm balls yielded nearly the same yield; however, one 7 mm ball yielded slightly more than two and three balls of the same size. One 10 mm ball, on the other hand, yielded significantly more than two 10 mm balls. The latter is most likely owing to a lack of ball movement due to unbalanced filling ratio in a Teflon jar with a capacity of 10 mL.

According to the experiment in the specified reaction setting, the reaction with stainless steel ball generated a superior yield than the matching zirconium and tungsten ball of the same size (Figure S3). Similarly, Teflon and stainless steel milling jars were compared to study the influence of milling jar. Under similar reaction circumstances, the Teflon container produced a higher yield than the stainless steel container.

The results of several vibration frequency studies revealed that 30 Hz is the best frequency for high yield. Under the given reaction circumstances, the yield achieved

TABLE 2: Substrate scope and versatility of the reaction in different electronic environments^a.

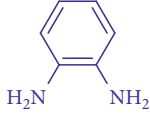
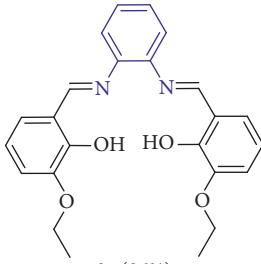
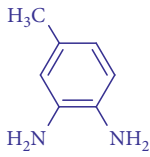
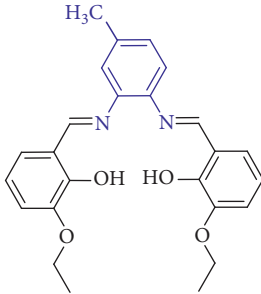

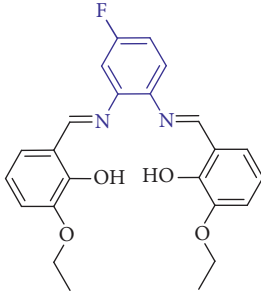
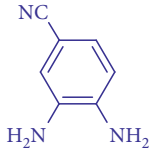
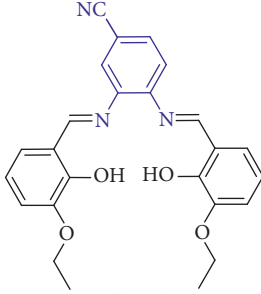
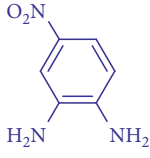
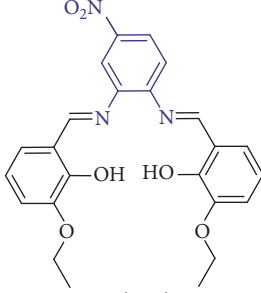
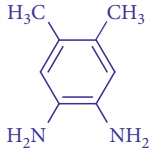
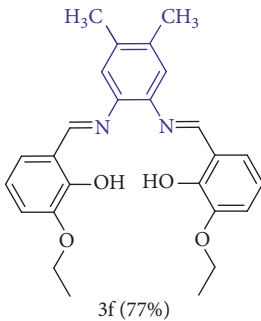

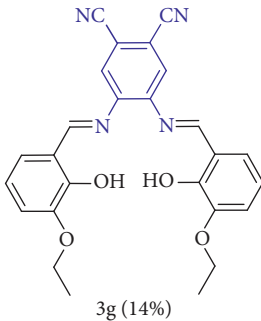
Entry	Diamine	Product (yield) ^b
1		 3a (86%)
2		 3b (69%)
3		 3c (44%)
4		 3d (33%)
5		 3e (10%)

TABLE 2: Continued.

Entry	Diamine	Product (yield) ^b
6		 3f (77%)
7		 3g (14%)

^aReaction conditions: 2mmol (1) and 1mmol (2a–g) were milled in a mixer mill. ^bIsolated yield.

for 20 and 25 Hz was nearly identical, but the yield obtained for lower frequencies decreased (Figure S4).

The optimal reaction conditions for the production of Salophen derivatives were discovered to be 30 Hz, 10 mm stainless steel ball, Teflon jar, and 1 h reaction period after multiple experimental optimization studies of the high-speed ball mill parameters (Table 1, entry 3).

2. Results and Discussion

The electronic effect on mono and di-substituted aromatic diamine and 3-ethoxysalicylaldehyde under ball mill conditions was evaluated in a high-speed vibratory ball mill, MM400 (Retsch). Table 2 lists the 7 target azomethines, abbreviated as **3a–3g**, along with their yields, with details provided in the Supporting Information (available here). The synthesized azomethines are colorful solids that are insoluble in water but soluble in acetone and methanol.

Using **3d** (entry 4 in Table 2) as a typical example target, the solid reactant 3-ethoxysalicylaldehyde (0.3324 g, 2.0 mmol) and the solid 3,4-diaminobenzonitrile (0.1332 g, 1.0 mmol) were milled in Teflon jar and stainless steel ball at the maximum frequency of MM400, 30 Hz, for 1 h without any solvent. Hexane was used to wash the solid product from the jar, then it was decanted and dried in a vacuo at RT. The product distribution was monitored by ¹H NMR (Figure 1) and FT-IR spectroscopy (Figure 2).

The identity of the synthesized azomethine was established by the ¹H NMR spectra. The azomethine proton was detected as a singlet at 8.56 ppm in the ¹H NMR spectrum of the produced azomethine. The aromatic ring hydrogens

were found in the range of 6.67–7.24 ppm. At 12.70 ppm, phenolic hydrogen was detected (Figure 1). The latter's downfield shift is related to intramolecular O–H···N H-bonding, which is a frequent property of o-hydroxysalicylidene systems [16].

Figure 2 shows the FT-IR spectra of the synthesized azomethine and the starting ingredients. The FT-IR spectrum shows a strong band attributed to C=N stretching vibrations at 1601 cm⁻¹, indicating that the azomethine structure was successfully formed. The CH aromatic stretching vibrations showed a weak absorption band at 2977–2889 cm⁻¹. The sharp single band at 2210 cm⁻¹ is attributed to the aromatic C≡N stretching vibrations. The aromatic rings' C=C stretching vibrations were assigned to the distinct absorption bands in the 1500–1460 cm⁻¹ range.

2.1. Substituent Effect. Substitution patterns on aromatic diamine may influence the electronic environment and distribution at the terminal NH₂ functionality, thus modifying its reactivity. To gain an understanding of the reactivity of the various diamines, the reaction conditions of the high-speed ball mill parameters were carefully optimized, and the substrate scope was investigated (Scheme 1).

The investigations began with a set of aromatic diamines containing substituents ranging from activating to strongly deactivating groups, which were then subjected to the given reaction conditions in order to analyze the electronic effect. The general reactivity trends were similar, indicating that the activation method did not have a significant impact on the reaction pathway. Diamines with different electronic

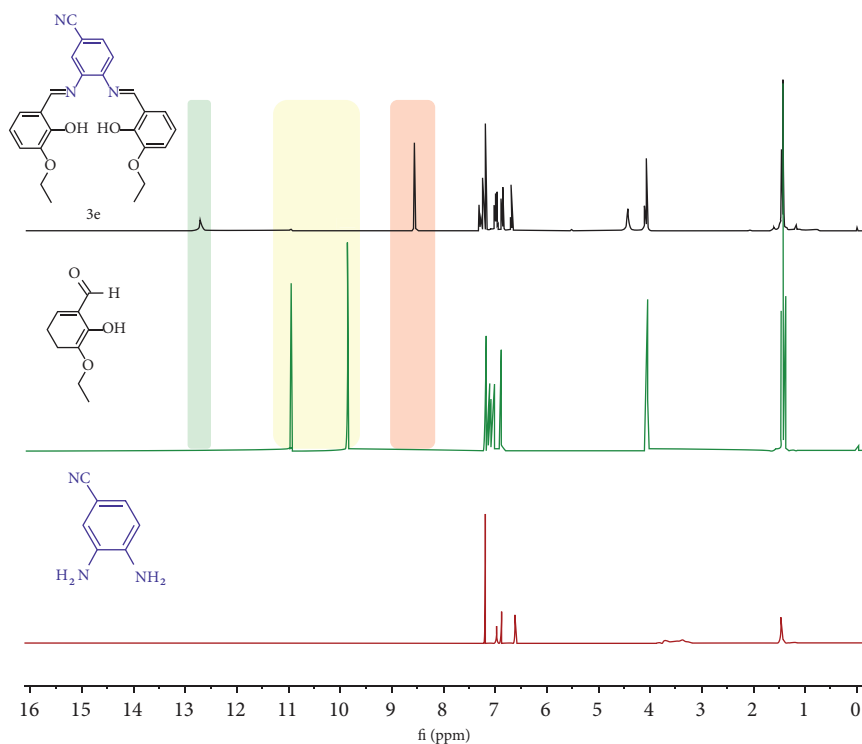
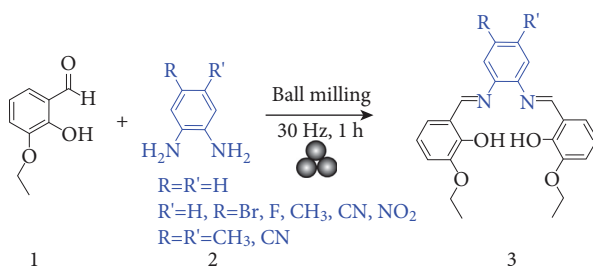


FIGURE 1: ^1H NMR Spectra of diamine, aldehyde, and Salophen ligand.



SCHEME 1: Solvent-free mechanochemical synthesis of Salophen ligands.

environment reacted with 3-ethoxysalicylaldehyde (**1**). In ball milling, electron-donating diamines yielded higher yields than diamines with electron-withdrawing groups, as expected. The diamine with a strong electron-withdrawing group had a lower yield (Table 2, entries 4 and 5). As a result, there was a significant yield difference between electron-donating methyl/ $-\text{CH}_3/$ and withdrawing nitro/ $-\text{NO}_2/$ substitutions (Table 2, entries 2 and 5), respectively, while both diamines yielded lower yields than the unsubstituted 1,2-phenylenediamine. Similarly, the di-substituted diamine (Table 2, entries 6 and 7) had a stronger effect than the corresponding mono-substituted diamine (Table 2, entries 2 and 4), respectively. The additional $-\text{CN}$ group had a negative effect on azomethine yield, whereas the $-\text{CH}_3$ group had a positive effect.

It was observed that even in the absence of a ball, such azomethine synthesis reactions occurred to some extent in vibratory milling (Figure S2). Furthermore, after the reaction, when the reaction products have sat in the container for

a while, the reaction will most likely continue to increase the yield. For these types of processes, the NMR solvent has also considerable impact. For example, when **3a** is synthesized without a ball and stored in an NMR tube with *d*-chloroform for 24 hours, the yield increases from 27% to 48%. However, staying in the NMR tube for additional 24 hours had no influence on the yield (Figure S5). As a result, it is preferable to characterize the reaction progress and product immediately in solid state if possible; otherwise, such situations must be noted in order to obtain reliable results.

The solid aromatic 1,2-phenylenediamine **2** was compared to ethylenediamine **4** and (1*S*, 2*S*)-(+)-1,2-diaminocyclohexane **5** in a comparative study to demonstrate the physical state as well as aromaticity effect of diamines, respectively. The results revealed that the liquid diamine **4** yielded better than both, whereas solid diamines **2** and **5** yielded similarly (Scheme 2).

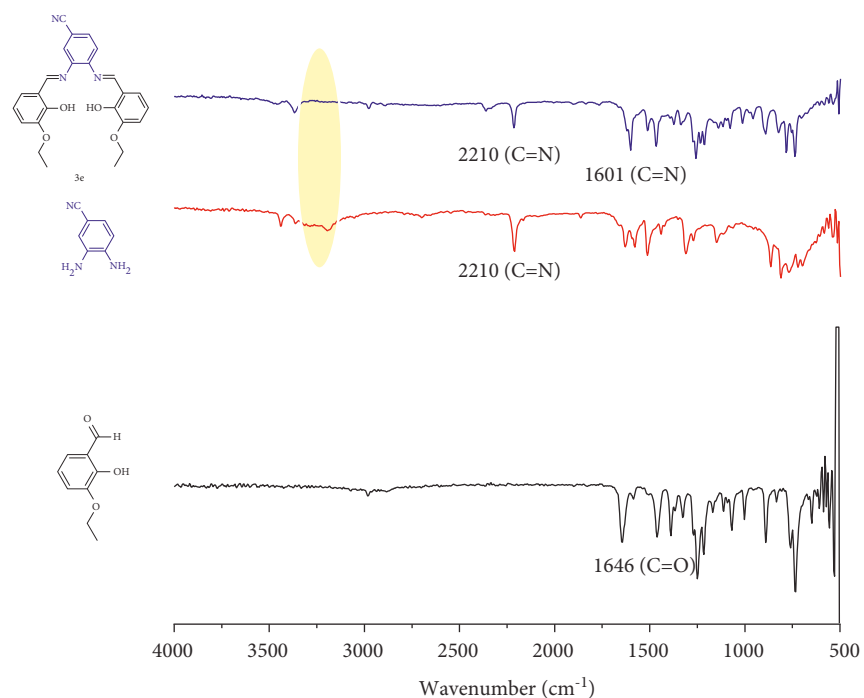
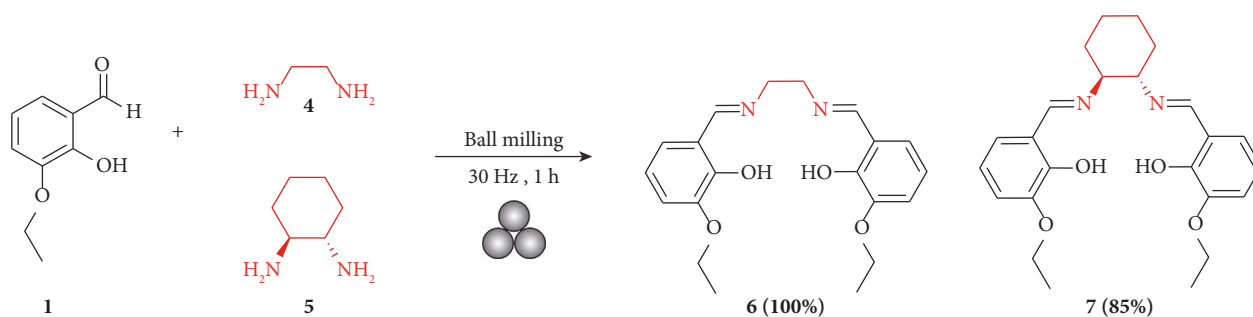


FIGURE 2: FT-IR spectra of aldehyde, diamine, and Salphen ligand.

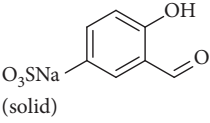
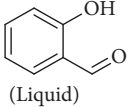
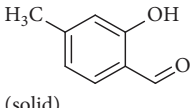
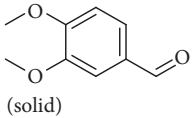
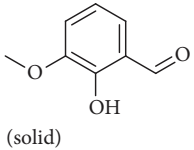
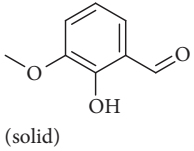
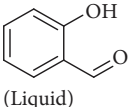
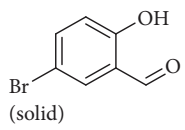


SCHEME 2: Solvent-free mechanochemical synthesis of Salen ligands.

TABLE 3: Comparison of current azomethine synthesis to different reported methods.

Entry	Diamine (1 mol)	Aldehyde (2 mol)	Solvent	Technique	Time	% yield
1	 R = Cl, Br	 (Liquid)	Ethanol	Stir [18]	4 h	67.96 (Cl) 63.52 (Br)
2	 (solid)	 R = Cl, Br (solid)	Ethanol	Reflux [33]	3–4 h	—
3		 (Liquid)	Ethanol	Reflux [13]	2 h	95
			Ethanol/H ₂ O	Reflux [13]	2 h	85

TABLE 3: Continued.

Entry	Diamine (1 mol)	Aldehyde (2 mol)	Solvent	Technique	Time	% yield
4		 (solid)	Ethanol	Reflux [20]	5 h	—
5		 (Liquid)	Ethanol	Reflux [39]	30 min	63.63
6		 (solid)	Ethanol	Reflux [19]	2 h	76
7		 (solid)	Ethanol	Reflux [5]	1 h	89.8
8		 (solid)	MeOH	Ball mill (LAG) [23]	1 h	60
9		 (solid)	Neat	Ball mill [32]	1 h	89.9
10		 (Liquid)	Neat	Ball mill [1]	2 h	98
11		 (solid)	Ethanol	Stir [34]	9 h	60
12			Ethanol	Reflux [35]	2 h	68
13			Ethanol	Reflux [36]	2 h	68
14			Ethanol	Reflux [37]	2 h	79.7
15			Hot ethanol	Stir [38]	2 h	90
16			Neat	Ball mill (in this work)	1 h	86

2.2. Comparative Outlook. In various reaction circumstances, azomethines can indeed be made from 3-ethoxysalicylaldehyde, o-vanillin, and other solid and liquid aldehydes. However, previously reported Salophen ligand syntheses used either traditional solution synthesis under reflux in protic solvents (e.g., methanol, ethanol, and so on) for a longer reaction time [13, 18, 20, 33] or LAG [23], or at least one of the reactants is liquid (i.e., reactant functioning as LAG) [32]. Except for Garca *et al.*, who synthesized a Salophen ligand containing bromine from 5-bromosalicylaldehyde and a diamine [1], there are no reports on the neat

mechanochemical synthesis of Salophen ligands in solid-solid type syntheses to our knowledge. As a result, the current study adds to our understanding of solid-solid Salophen synthesis in diverse electronic environments employing solvent-free mechanochemistry. In the synthesis of **3a**, mechanochemical ball milling was shown to be more effective than most of corresponding solution reactions reported in the literature (about 26% yield) [34–38]. In comparison to the similar Salophen synthesis to date, the current study provided solvent-free, fast reaction, high yield, and easy work-up (Table 3).

3. Conclusion

In this work, the electronic effect on Salophen production in a ball mill was effectively proved. The findings not only introduce a solvent-free mechanochemical synthesis of Salophen ligands from solid reactants but also it introduces a simple work-up procedure. A solvent-free mechanochemical synthesis of metal-Salophen complexes for medical applications is currently underway as a follow-up to this work.

Data Availability

Spectral data used to support the findings of this study have been submitted with the manuscript as supplementary material.

Conflicts of Interest

The author declares that there are no conflicts of interest.

Acknowledgments

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Supplementary Materials

General remarks, experimental section, and structure characterization data: ^1H NMR, ^{13}C NMR, FT-IR, and high-resolution mass of compounds, are available in supporting files. (*Supplementary Materials*)

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