



Nickel-catalyzed One-pot, Three-component Synthesis of 3,4-disubstituted Isoxazole-5(4*H*)-ones in Aqueous Medium

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ABSTRACT

An efficient and simple cyclocondensation approach for the one-pot, three-component synthesis of 3,4-disubstituted isoxazole-5(4*H*)-ones has been described. This three-component reaction was completed easily and cleanly to give corresponding isoxazoles in the presence of Ni(OAc)₂·H₂O as the catalyst. The reaction was implemented in aqueous media at room temperature and the corresponding heterocyclic products were obtained in good to high yields. The present work offers notable advantages, including easy work-up, simple procedure, mild condition, and no use of organic solvents.

Keywords: 3,4-disubstituted isoxazole-5(4*H*)-ones, Ni(OAc)₂·H₂O, water, three-component synthesis, β-oxoesters

1. INTRODUCTION

Multi-component reactions (MCRs) expedite the synthesis of target molecules by joining three or more substrates in one vessel reaction. These types of reactions are generally mild, and in many cases can also be useful for producing a variety of pharmaceuticals, complex organic molecules are cost-effective approach. Another important feature of MCR is that it tolerates a diversity of precursors, decreasing the steps need towards to products and yielding the products with high yields, minimal work-up and purification [1, 2].

The isoxazole moiety is a key five-membered cyclic oxime ester, bearing one oxygen and one nitrogen atoms at adjacent positions, to be attractive target in the bioorganic, synthetic

organic chemistry and the pharmaceutical industry [3, 4]. Organic compounds containing isoxazole was also applied as merocyanine dyes [5]. In addition, molecules carrying the isoxazole moiety are widely found to be associated with diverse biological activities [6-8] such as antimicrobial, fungicidal, anticonvulsant, HDAC inhibitory, protein-tyrosine phosphatase 1B (PTP1B) inhibitory, analgesic, antioxidant, anti-apoptotic, COX-2 inhibitory, nematicidal, anti-inflammatory, antiviral, and anti-tubercular. Moreover, the isoxazole core is also structural component of many drugs, e.g., inhibitor of a tumor necrosis factor-alpha (TNF-α) [9], sulfisoxazoles [10], antibiotics [11] and were used as anti-androgens agent [12]. Isoxazole-

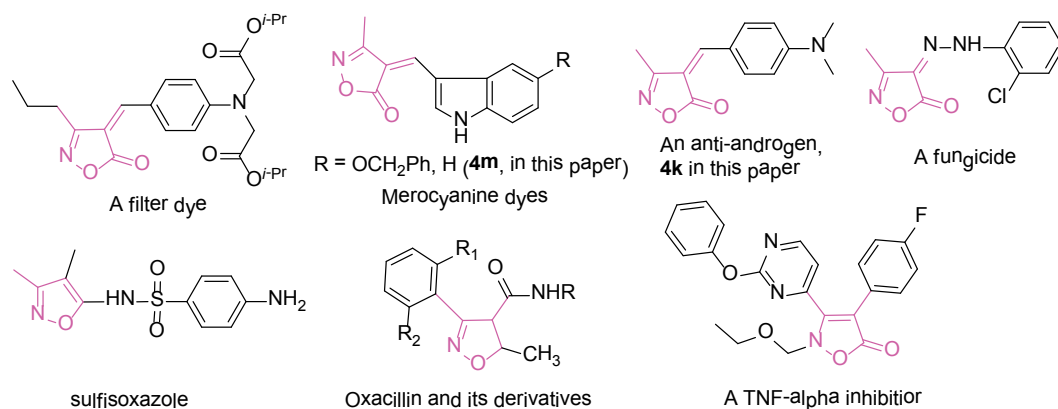


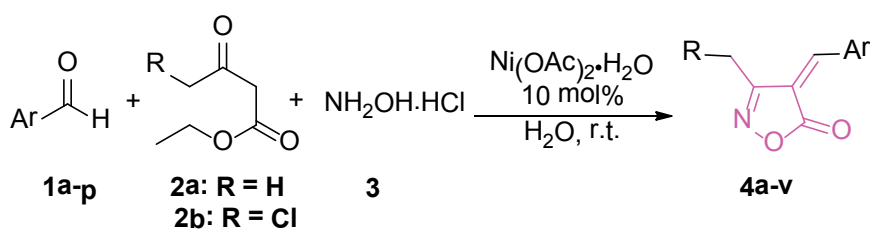
Figure 1. Interesting isoxazole-containing compounds.

5(4*H*)-ones are powerful proaromatic acceptors and could also be used for the development of optical storage, nonlinear optical research [5], and filter dye in photographic films [13]. A number of interesting compounds with isoxazole nucleus are shown in Figure 1.

On account of the wide range of applications of isoxazoles, a great deal of attention has been focused on to the development of efficient methodologies for the synthesis of this class of heterocyclic compounds. During the recent years, 3,4-disubstituted isoxazole-5(4*H*)-ones were prepared by using catalytic amounts of sodium benzoate [14], Na_2S [15], sodium silicate [16], and DABCO [17]. The three-component reaction of β -oxoesters, hydroxylamine hydrochloride, and aromatic aldehydes using different conditions such as NaOAc and visible light [18], pyridine under ultrasonic irradiation [19, 20], pyridine under reflux [21], and catalyst-free/grinding or

heating [22] also provides isoxazole-5(4*H*)-ones. Furthermore, we synthesized arylmethylene isoxazole-5(4*H*)-ones by using some of catalysts including sodium citrate [23], sodium saccharin [24], sodium tetraborate [25], NaN_3 [26], boric acid [27], as well as potassium phthalimide (PPI) [28]. Although 4*H*-isoxazole-5-ones have so far been synthesized, to the best of our knowledge, no reports that include the use of $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ for cyclocondensation of aldehydes (**1a-p**), β -oxoesters (**2a-b**), and $\text{NH}_2\text{OH} \cdot \text{HCl}$ (**3**) have been reported. With regard to the above mentioned information in the mind, it was decided to $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ is used as a new catalyst in the synthesis of 3,4-disubstituted isoxazole-5(4*H*)-ones in aqueous media (Scheme 1).

Water has a special place as the most attractive solvent in the synthetic chemistry. Also, the use of water as the solvent, not only



Scheme 1. The one-pot, three-component reaction of aryl/heteroaryl aldehydes (**1a-p**), β -oxoesters (**2a-b**), and $\text{NH}_2\text{OH} \cdot \text{HCl}$ (**3**) in the presence of $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$.

decreases the risk of the organic solvents, but also increases the rate of chemical reactions. Also, MCR in water can be visualized as a well-designed synthetic method to attain a wide range of diverse molecular frameworks [29, 30].

2. MATERIALS AND METHODS

2.1. Instruments and Characterization

All the reagents were obtained from commercial sources and used without further purification. Melting points were measured on a Buchi 510 melting point apparatus and are uncorrected. ^1H NMR and ^{13}C NMR spectra were recorded at ambient temperature on a BRUKER AVANCE DRX-400 MHz spectrophotometer using CDCl_3 as the solvent. The purity of synthesized compounds as well as the development of the reactions was monitored by thin layer chromatography (TLC) analysis on Merck pre-coated silica gel 60 F_{254} aluminum sheets, visualized by UV light. All of the targeted products are reported in the literature and are characterized by comparison of their spectral and physical data on the basis of literature descriptions.

2.2. General Procedure for the Synthesis of 3,4-Disubstituted Isoxazole-5(4H)-ones (4a-v)

A mixture of β -oxoester (1 mmol), $\text{NH}_2\text{OH}\cdot\text{HCl}$ (0.0695 g, 1 mmol) and $\text{Ni}(\text{OAc})_2\cdot\text{H}_2\text{O}$ (10 mol %) in 5 mL of distilled water was stirred at room temperature for 10 min, then aldehyde (1 mmol) was added to the mixture. The reaction mixture was stirred at ambient temperature until the reaction was completed (monitored by TLC analysis). After completion of reaction, the precipitates were separated by gravity filtration, washed with cold distilled water and dried in open air. Crude products were recrystallized in EtOH (95%) to afford the title compounds. The products are known and their physical properties were confirmed by those reported in the literature [20-28].

Selected Spectral Data

3-Methyl-4-(4-methylbenzylidene)isoxazol-5(4H)-one (4c):

^1H NMR (400 MHz, CDCl_3): δ 2.33 (s, 3H), 2.48 (s, 3H), 7.36 (d, $J = 8.0$ Hz, 2H), 7.42 (s, 1H), 8.32 (d, $J = 8.4$ Hz, 2H); ^{13}C NMR (100 MHz, CDCl_3): δ 11.6, 22.1, 118.2, 129.8, 129.9, 134.2, 145.8, 150.2, 161.4, 168.3.

4-(4-(Dimethylamino)benzylidene)-3-methylisoxazol-5(4H)-one (4k):

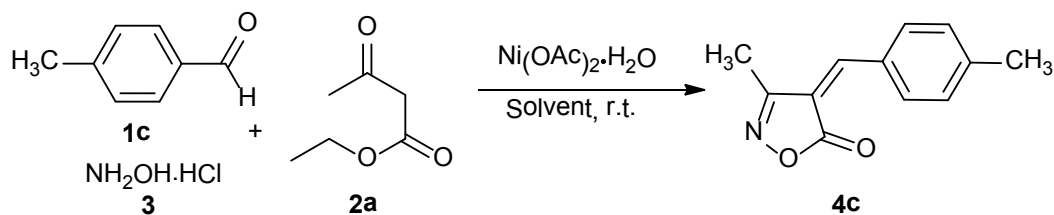
^1H NMR (400 MHz, CDCl_3): δ 2.27 (s, 3H), 3.19 (s, 6H), 6.75 (dd, $J = 1.2, 8.4$ Hz, 2H), 7.24 (s, 1H), 8.43 (d, $J = 8.4$ Hz, 2H); ^{13}C NMR (100 MHz, CDCl_3): δ 11.7, 40.1, 110.9, 111.5, 121.5, 137.7, 149.3, 154.2, 161.7, 170.2.

3. RESULTS AND DISCUSSION

At first, the cyclocondensation reaction of *p*-methylbenzaldehyde (**1c**), ethyl acetoacetate (**2a**), and $\text{NH}_2\text{OH}\cdot\text{HCl}$ (**3**) was selected as the model. The synthesis of 3-methyl-4-(4-methylbenzylidene)-4*H*-isoxazole-5-one (**4c**) in the presence of different catalytic amounts of $\text{Ni}(\text{OAc})_2\cdot\text{H}_2\text{O}$ in various solvents was explored, and the results are summarized in Table 1.

It was found that by increasing the amounts of the catalyst from 2.5 mol% to 10 mol%, the yield of **4c** improved from 60% to 96% (Table 1, entries 1-4). Increasing the amount of catalyst beyond 10 mol%, did not lead to a significant improvement the yield of the product (Table 1, entries 5-7). It seems that 10 mol% of $\text{Ni}(\text{OAc})_2\cdot\text{H}_2\text{O}$ is the most optimal loading of the catalyst. In order to explore the most favorable solvent, the same reaction was carried out in several solvents such as EtOH, acetone, 1,4-dioxane, *n*-hexane, and a mixture of H_2O -EtOH (1:1, V:V), H_2O -acetone (1:1, V:V), and H_2O -1,4-dioxane (1:1, V:V) (Table 1, entries 8-14) at room temperature using the same catalyst yielded 64%, 20%, 32%, 38%, 70%, 25%, and 45%, respectively. The polarity of solvent and presence of catalyst

Table 1. Effects of different solvents and amounts of catalyst on the cyclocondensation of *p*-methylbenzaldehyde (**1c**), ethyl acetoacetate (**2a**), and $\text{NH}_2\text{OH}\cdot\text{HCl}$ (**3**).^a

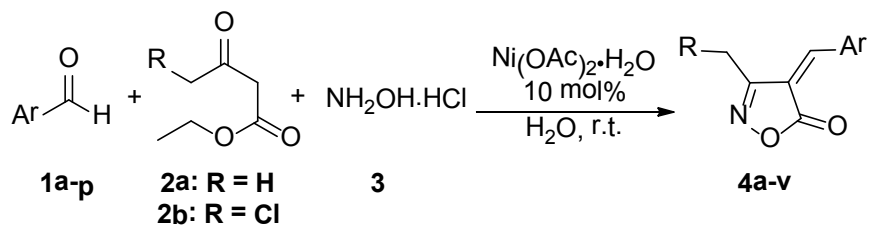


Entry	Solvent	Amounts of catalyst (mol%)	Time (min)	Yield (%) ^b
1	H_2O	2.5	90	60
2	H_2O	5	80	85
3	H_2O	7.5	80	90
4	H_2O	10	80	96
5	H_2O	15	80	95
6	H_2O	18	80	96
7	H_2O	20	80	95
8	$\text{C}_2\text{H}_5\text{OH}$	15	80	64
9	$\text{CH}_3\text{C}(\text{O})\text{CH}_3$	15	80	20
10	1,4-Dioxane	10	120	32
11	<i>n</i> -Hexane	10	120	38
12	$\text{H}_2\text{O}:\text{C}_2\text{H}_5\text{OH}$ (1:1)	10	100	70
13	$\text{H}_2\text{O}:\text{CH}_3\text{C}(\text{O})\text{CH}_3$ (1:1)	10	120	25
14	$\text{H}_2\text{O}:\text{1,4-dioxane}$ (1:1)	10	120	45
15	Solvent-free	10	120	40

^a Reaction conditions: *p*-methylbenzaldehyde **1c** (1 mmol), ethyl acetoacetate **2a** (1 mmol), $\text{NH}_2\text{OH}\cdot\text{HCl}$, **3** (1 mmol), solvent (5 mL), room temperature. ^b Isolated yield of product.

play an important role for the success of the reaction. In the organic solvents, the yield of **4c** was lower and longer reaction time was required, whereas in aqueous media the product is obtained in good yield (Table 1, entries 1-7). To optimize the yield on the basis of the results, water instead of organic solvents was chosen as media of reaction. The reaction performed without solvent at room temperature for 120 min (Table 1, entry 15) gave **4c** in only

40% yield. The model reaction has also been carried out at gram-scale quantity (20 mmol of the each of the reactants) and provides the corresponding product in 90% isolated yield. With these optimized reaction conditions in hand, the scope of this reaction was examined with various aryl/heteroaryl aldehydes, and ethyl 4-chloroacetoacetate for synthesis of a series of 3,4-disubstituted isoxazole-5(4*H*)-ones. The results are shown in Table 2.

Table 2. Synthesis of 3,4-disubstituted isoxazole-5(4*H*)-ones (**4a-v**).^a

Entry	Ar-CHO	R	Product	Time (min)	Isolated Yields (%)	Mp (°C)	
						Found	Lit. ^b
1		H	4a	115	90	141-143	141-143
	1a						
2		H	4b	75	95	171-173	175-177
	1b						
3		H	4c	80	96	135-136	135-136
	1c						
4		H	4d	60	98	215-216	211-214
	1d						
5		H	4e	120	88	240-242	238-241
	1e						
6		H	4f	75	92	146-147	143-145
	1f						
7		H	4g	80	90	141-142	142-144
	1g						

Table 2. (Continued).

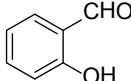
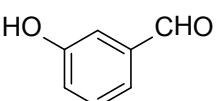
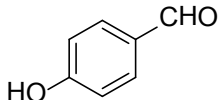
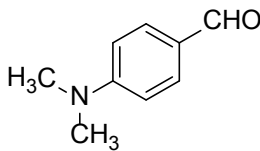
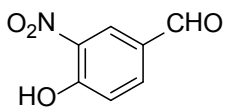
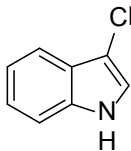
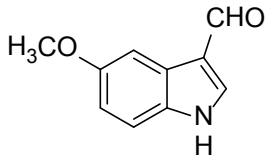
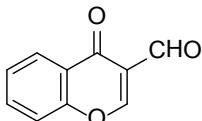
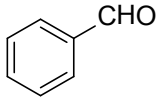
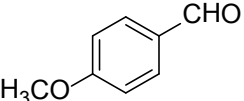
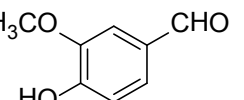
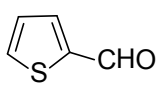
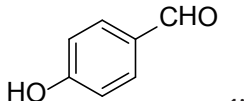
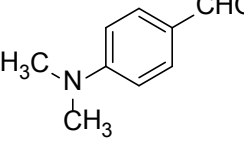
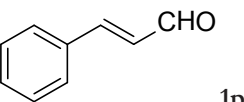
Entry	Ar-CHO	R	Product	Time (min)	Isolated Yields (%)	Mp (°C)	
						Found	Lit. ^b
8		H	4h	130	78	198-200	198-201
9	1h 	H	4i	100	92	202-203	199-201
10	1i 	H	4j	60	94	211-213	214-216
11	 1j	H	4k	80	92	227-228	220-221
12	 1l	H	4l	40	77	267-268	266-267
13	 1m	H	4m	120	84	240-241	239-241
14	 1n	H	4n	110	80	234-235	235-237
15	 1o	H	4o	120	91	243-244	251, 242-244

Table 2. (Continued).

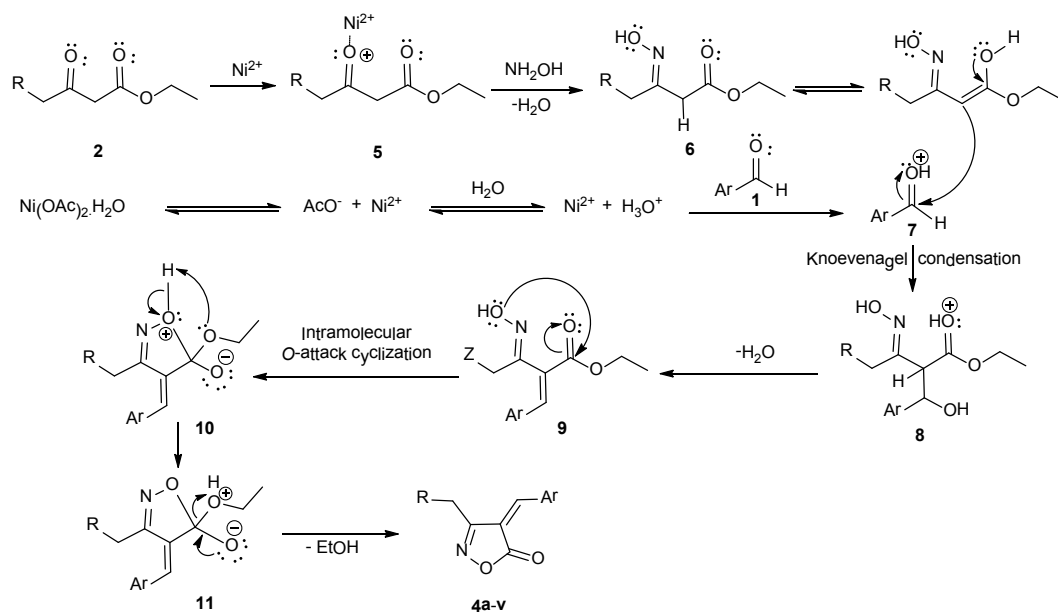
Entry	Ar-CHO	R	Product	Time (min)	Isolated Yields (%)	Mp (°C)	
						Found	Lit. ^b
16		Cl	4p	70	86	182-183	183-186
	1a						
17		Cl	4q	65	95	174-175	174-176
	1b						
18		Cl	4r	60	95	143-145	142-145
	1d						
19		Cl	4s	70	87	146-147	146-148
	1f						
20		Cl	4t	70	92	183-185	183-186
	1j						
21		Cl	4u	75	91	179-181	179-180
	1k						
22		H	4v	80	90	175-176	173-178
	1p						

^a Reagents and conditions: aldehyde **1** (1 mmol), β -oxoester **2** (1 mmol), hydroxylamine hydrochloride **3** (1 mmol), water (5 mL), room temperature. ^b Melting points are listed in Refs. 20-28.

As shown in Table 2, aromatic aldehydes with electron-donating substituents afford the corresponding products in good to excellent yields (Table 2, entries 2-4, 8-12, 17-18 and 20-21) in short reaction times. Furthermore, It was found that the yield was satisfactory when the reaction was carried out with 4-hydroxy-3-nitrobenzaldehyde (**1l**) (Table 2, entry 12), whereas the yield of the product was unsatisfactory when the reaction was performed with 4-nitrobenzaldehyde even for 24 h, which possibly due to electronic effects. Trace amount of products were formed when the aromatic aldehydes having electron-withdrawing substituents such as chlorine and nitro was used. In such cases, the starting materials were recovered intact and only very slight amounts of starting materials were consumed. The reaction using α,β -unsaturated aldehydes such as cinnamaldehyde (**1p**) leads to the corresponding product (**4v**) in high yield (Table 2, entry 22). When salicylaldehyde (**1h**) were utilized, the corresponding product

(**4h**) was produced in relatively lower yield (Table 2, entry 8), which may be due to the steric hindrance of the hydroxyl group at the *ortho*-position. Fascinatingly, when heteroaryl aldehydes including furan-2-carbaldehyde (**1e**), thiophene-2-carbaldehyde (**1f**), thiophene-3-carbaldehyde (**1g**), indol-3-carbaldehyde (**1m**), 5-methoxyindol-3-carbaldehyde (**1n**), and chromene-3-carbaldehyde (**1o**) were used (Table 2, entries 5-7, 13-15 and 19), the reaction proceeded efficiently and afford the corresponding products in high yields.

The products were identified by spectral data and mixed melting points with authentic sample. For example, the ^1H NMR spectrum of **4c** in CDCl_3 showed two sharp singlets for methyl protons of the *p*-position at the phenyl ring ($\delta = 2.33$ ppm) and methyl protons of the isoxazole ring ($\delta = 2.48$ ppm). The signal for the olefinic proton observed as a singlet at $\delta = 7.42$ ppm. The signals of the *ortho*- and *meta*-protons relative to the *p*-methyl group appeared as two doublets at $\delta = 7.36$ and 8.32



Scheme 2. A plausible mechanism for the three-component synthesis of 3,4-disubstituted isoxazole-5(4H)-ones (**4a-v**).

ppm, respectively. In the ^{13}C NMR spectrum, the signals corresponding to ester $\text{C}=\text{O}$ and imine $\text{C}=\text{N}$ groups of **4c** were observed at $\delta = 168.3$ and 161.4 ppm, respectively. Signals appeared at $\delta = 11.6$ and 22.1 ppm indicated the presence of methyl carbons at isoxazole ring and *para*-position of phenyl ring, respectively. The $\text{CH}=\text{C}$ resonance was visible at 150.2 ppm. The resonances of the other carbons were visible at $\delta = 118.2$ - 145.8 ppm. The NMR spectra of the other compounds **4a-v** are similar to those of **4c** except for the chloromethyl or aryl groups in C-3 position of isoxazole moiety, which show, in each case, characteristic signal in appropriate regions of the spectra.

So far, the mechanism for the formation of 3,4-disubstituted isoxazole-5(4*H*)-ones (**4a-v**) catalyzed by $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ has not been clear. On the basis of the literature evidences [20-28], a plausible reaction pathway for this condensation is proposed in Scheme 2. As showed in the pathway sketched in Scheme 2, it is reasonable to assume that the oxime derivatives **6** were formed by the condensation reaction of $\text{NH}_2\text{OH} \cdot \text{HCl}$ with Ni-coordinated ethyl acetoacetate **5**. Then Knoevenagel adducts **9** were formed through the condensation of **6** and protonated aryl aldehydes **7**. The next step may involve intramolecular *O*-attack cyclization of **9** to cyclic derivatives **10**, which undergo a proton exchange, followed by **11** is deethanolized to target products **4a-v**.

The reusability of the $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ in the model reaction was evaluated for four repeated times (Table 3). The catalyst present

in the aqueous filtrate medium. TLC showed that there was no starting materials or product in the filtered solution. After completion of reaction, the catalyst was easily recovered through evaporation of the solvent from aqueous filtrate solution and was used for the subsequent cycle. The results indicate that the catalyst revealed good catalytic activity even after four times of the recovery.

In order to efficiency of $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ for the reaction of *p*-methylbenzaldehyde, ethyl acetoacetate, and $\text{NH}_2\text{OH} \cdot \text{HCl}$, a comparison with the pervious reported catalysts was also made (Table 4). As indicated in Table 4, $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ is comparable to the previously reported methods in terms of reaction times and yields.

4. CONCLUSIONS

In conclusion, $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ was used as a catalyst for a facile synthesis of 3,4-disubstituted isoxazole-5(4*H*)-ones with high yields *via* one-pot cyclocondensation of aryl aldehydes or heterocyclic aldehydes with β -oxoester and $\text{NH}_2\text{OH} \cdot \text{HCl}$ in aqueous media. The merits of described process are easy work-up, mild conditions, the relatively shorter reaction times, high yields, simplicity and the using of water as the friendliness ecologically solvent, which makes it an interesting way towards the synthesis of isoxazole-5(4*H*)-ones.

ACKNOWLEDGEMENT

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Table 3. Results obtained using recycled of $\text{Ni}(\text{OAc})_2 \cdot \text{H}_2\text{O}$ in the model reaction.

No. of cycles	1	2	3	4	5
Time (min)	80	80	85	90	90
Isolated yields (%)	96	94	92	89	85

Table 4. Comparison of the results using Ni(OAc)₂·H₂O with those obtained by the other reported catalysts for the synthesis of **4b**.^a

Entry	Catalyst /conditions	Catalyst amount (mol%)	Time (min)	Yield (%)	Reference
1	Ni(OAc) ₂ ·H ₂ O/r.t.	10	75	95	Current work
2	Sodium benzoate/H ₂ O/r.t.	10	90	87	[14]
3	Na ₂ S/EtOH/r.t.	5	90	88	[15]
4	Sodium silicate/H ₂ O/r.t.	5	90	91	[16]
5	Pyridine/H ₂ O/ultrasound	100	60	82	[19]
6	Catalyst free/grinding	-	48	61	[22]
7	Catalyst free/105-110 °C	-	15	66.3	[22]
8	Sodium citrate/H ₂ O/r.t.	10	60	91	[23]
10	Sodium saccharin/H ₂ O/r.t.	10	50	91	[24]
11	Sodium tetraborate/H ₂ O/r.t.	10	50	95	[25]
12	Sodium azide/H ₂ O/r.t.	5	240	85	[26]
13	Boric acid/H ₂ O/r.t.	10	50	92	[27]
14	PPI/H ₂ O/r.t.	10	70	96	[28]

^a 4-Methoxybenzaldehyde (**1b**), ethyl acetoacetate (**2a**), and NH₂OH.HCl (**3**) in the presence of catalyst.

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