

## Synthesis of bis-thiazolidinones catalyzed by nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> under microwave irradiation

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### ABSTRACT

A rapid and efficient method for the synthesis of bis-thiazolidinones has been achieved by pseudo five-component reaction of aldehydes, ethylenediamine and thioglycolic acid in the presence of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> as catalyst under microwave irradiation. Nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> has been characterized by powder X-ray diffraction, scanning electronic microscopy (SEM), energy dispersive spectroscopy (EDS), vibrating sample magnetometer (VSM), dynamic light scattering (DLS), thermogravimetric analysis (TGA) and FT-IR spectroscopy. In this research, microwave irradiation is used as a green and complementary technique for preparation of bis-thiazolidinones. Use of simple and readily available starting materials, excellent yields, short reaction times, reusability of the catalyst, low amount of catalyst and using of microwave as clean method are some advantages of this protocol.

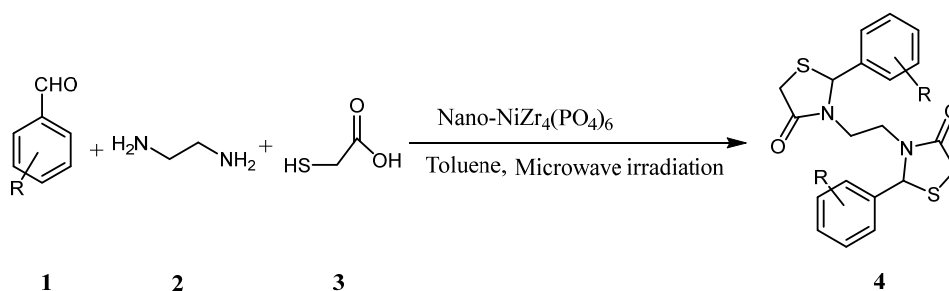
**Keywords:** Bis-thiazolidinones, Nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>, Five-component, Microwave irradiation, Nanocatalyst.

### 1. Introduction

Thiazolidinones are one of the biologically significant scaffolds because of their wide range of biological and pharmacological activities such as anticancer [1], analgesic [2], antibacterial [3], antitubercular [4], antihistaminic [5], and anti-HIV [6,7]. Therefore, the improvement of efficient methods for the synthesis of thiazolidinones is of great interest. A few methods have been reported for the synthesis of bis-thiazolidinones in the presence of the catalysts such as HClO<sub>4</sub>-SiO<sub>2</sub> [8] Fe<sub>2</sub>O<sub>3</sub> [9], ChCl (Choline Chloride) / urea based ionic liquid [10], and ZnCl<sub>2</sub> [11]. However, some of the reported methods endure drawbacks including long reaction times, harsh reaction conditions and use of toxic and non-reusable catalyst. Therefore, to avoid these limitations, the exploration of an efficient, easily available catalyst with high catalytic activity and short reaction times for the preparation of bis-thiazolidinones is still favored. The feasibility of achieving one-pot multi-component synthesis under microwave irradiation with a heterogeneous catalyst

could improve the reactions rates and shorten the reactions times [12-14]. Microwave irradiation (MWI) is utilized for a diversity of organic syntheses due to short reaction times, easy workup and good yields [15,16]. The reaction mixture is heated from the inside when the microwave energy is transferred directly to the molecules (catalysts, solvent and reagents). The heterogeneous catalysts absorb microwave irradiation; hence they can provide an internal heat source for the reactions [17-19]. The heterogeneous catalysts could be recovered by easy operations and reused many times without significant loss of its catalytic activity. In addition, the high surface-to-volume ratio of nanoparticles provides a larger number of active sites per unit area compared to their heterogeneous counterparts. NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> ceramics as heterogeneous catalysts have been interested due to their unique properties and potential applications in diverse fields [20,21]. Herein we wish to report the synthesis of bis-thiazolidinones by one-pot pseudo-five-component condensation of araldehydes, ethylenediamine and thioglycolic acid with nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> as a reusable and robust heterogeneous catalyst under microwave irradiation in toluene (Scheme 1).

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**Scheme 1.** Synthesis of bis-thiazolidinones using nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

## 2. Experimental

### 2.1. Chemicals and apparatus

Reagent grade chemicals and solvents were purchased from Sigma-Aldrich or Merck and were used without further purification. NMR spectra were obtained on a Bruker Avance-400 MHz spectrometer (<sup>1</sup>HNMR at 400 Hz, <sup>13</sup>CNMR at 100 Hz) in DMSO-*d*<sub>6</sub> or CDCl<sub>3</sub> using TMS as an internal standard. Chemical shifts (δ) are given in ppm and coupling constants (J) are given in Hz. We used the Milestone microwave (Microwave Labstation, MLS GmbH- ATC-FO 300) for synthesis. The IR spectra were recorded on FT-IR Magna 550 apparatus using with KBr plates. Melting points were determined on Electro thermal 9200, and are not corrected. The elemental analyses (C, H, N) were obtained from a Carlo ERBA Model EA 1108 analyzer. Powder X-ray diffraction (XRD) was carried out on a Philips diffractometer of X'pert Company with monochromatized Zr Kα radiation (λ = 1.5406 Å). In order to study the particle size and morphology of the synthesis nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>, FE-SEM images of the products visualized by a HITACHI S4160 Field Emission Scanning Electron Microscope. The magnetic measurement of samples were carried out in a vibrating sample magnetometer (VSM) (Meghnatis Daghigh Kavir Co.; Kashan Kavir; Iran) at room temperature in an applied magnetic field sweeping between ±10,000 Oe.

### 2.2. Preparation of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>

ZrOCl<sub>2</sub> was used as zirconium source. Firstly 1 mmol of ZrOCl<sub>2</sub>.8H<sub>2</sub>O and 1 mmol of Ni(NO<sub>3</sub>)<sub>2</sub>.2H<sub>2</sub>O were added in 15 mL of HO(CH<sub>2</sub>)<sub>2</sub>OH and sonicated at 30 W power to completely dissolution. Afterward 0.8 ml H<sub>3</sub>PO<sub>4</sub> (85%), 4 mmol of NH<sub>4</sub>Cl and 1.4 mL of CH<sub>3</sub>NH<sub>2</sub> water solution (25.0-30.0%) were added consecutively and sonicated for 30 min. Then, the reaction mixture was transferred into a Teflon-lined autoclave under autogenous pressure at 200 °C for 5 days. When the reaction was completed, dispersed precipitate was obtained. The solid was filtered and

washed with distilled water and ethanol several times. Subsequently product was dried at 50 °C for 5 h and calcinated at 700 °C for 2 h. Afterward the solid was added in 20 mL of DMF and sonicated at 95 W power for 2 h. Finally the resulting product was filtered, washed with distilled water and absolute ethanol and dried at 150 °C for 2 h in vacuum to afford pure nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> ceramics.

### 2.3. General procedure for the synthesis of bis-thiazolidinones

A mixture of aldehydes (2 mmol), ethylenediamine (1 mmol), thioglycolic acid (2 mmol) and 2 mol % of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> in toluene (5 mL) was subjected to microwave irradiation (400 W) for the specific time. The reaction was monitored by TLC. After completion of the reaction, CHCl<sub>3</sub> was added to dilute the reaction mixture after terminating the reaction. The catalyst was insoluble in the solvent and was separated by centrifuging. The CHCl<sub>3</sub> was evaporated and the crude mixture was separated by silica gel column chromatography (diethyl ether/petroleum ether) to get pure product.

## 3. Results and Discussion

XRD patterns of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> were shown in Fig. 1. The pattern agrees well with the reported pattern for nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> nanoparticles (JCPDS No. 45-0013). The morphology and particle size of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> were investigated by scanning electron microscopy (SEM) as shown in Fig. 2. The SEM images show particles with diameters in the range of nanometers.

The elemental compositions of the nanocatalyst were analyzed by Energy Dispersive Spectroscopy (EDS). EDS confirmed the presence of Ni, Zr, P and O in the compound (Fig. 3).

The magnetic properties of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> was revealed with the help of a vibrating sample magnetometer (VSM) (Fig. 4). The amount of saturation-magnetization for nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> is ~ 0.03 emug<sup>-1</sup>.

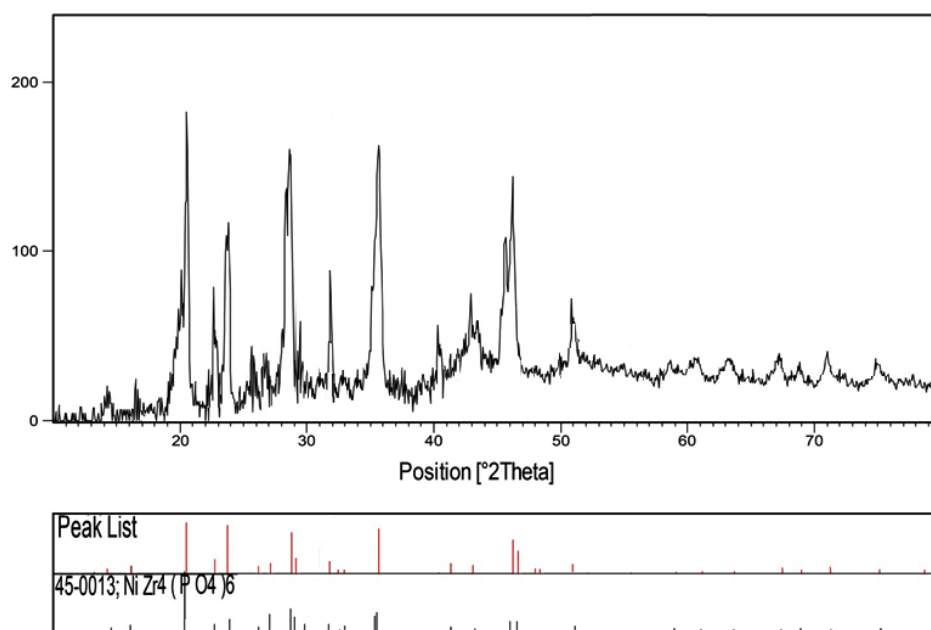


Fig.1. XRD pattern of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

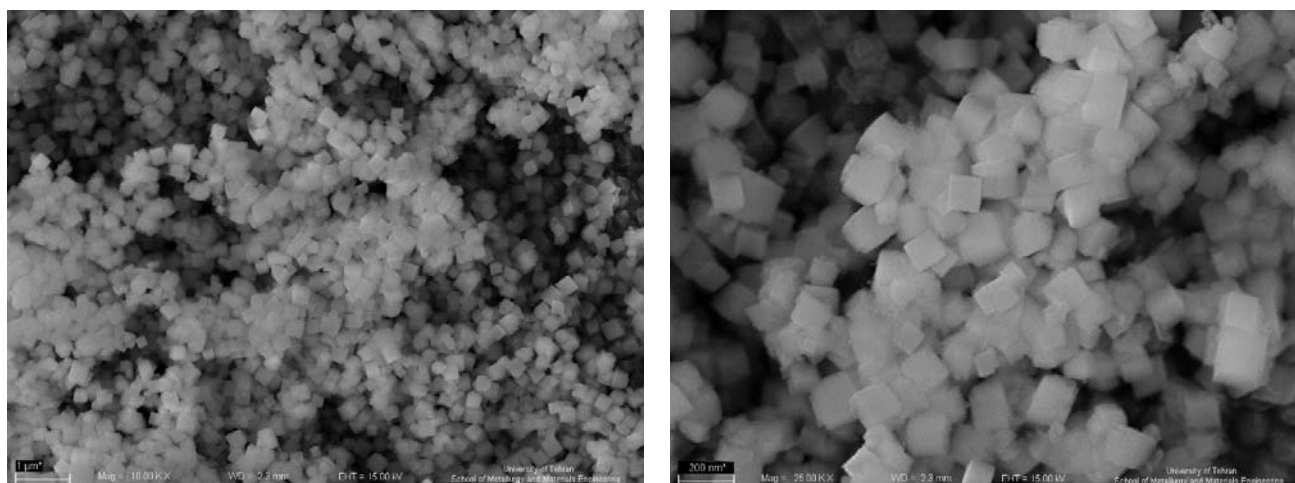


Fig. 2. FE-SEM images of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

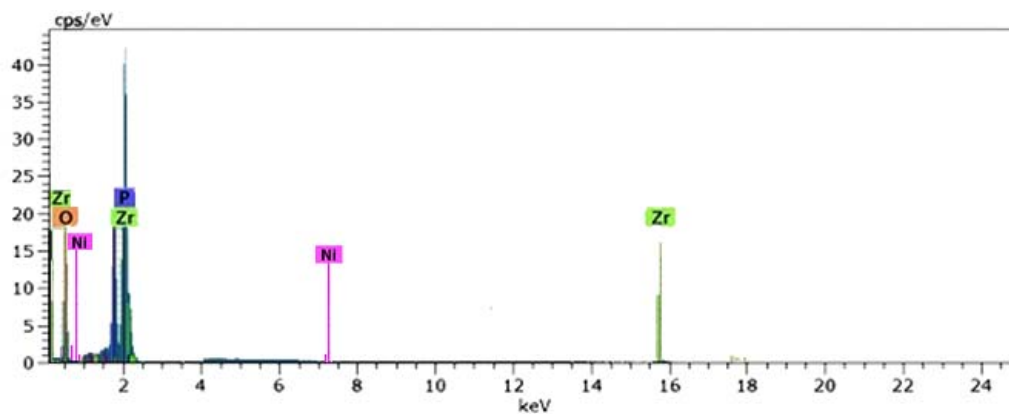


Fig. 3. EDS of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

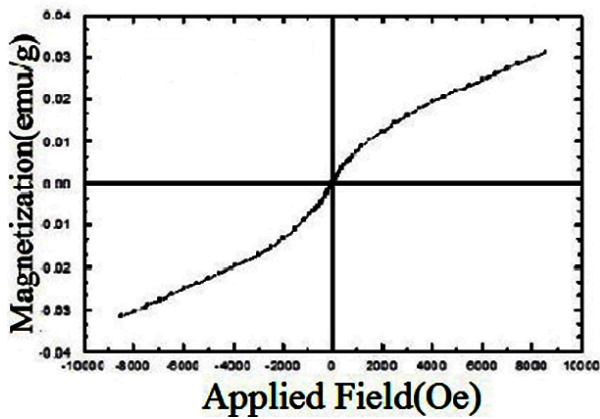


Fig. 4. The VSM curve of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

In order to investigate the size distribution of nanoparticles, DLS (dynamic light scattering) graph of

the nanoparticles were presented in Fig. 5. This size distribution is centered at a value of 138.3 nm.

Thermogravimetric analysis (TGA) evaluates the thermal stability of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>. A 1.9% decrease in weight between 50 and 300 °C is due to losing absorbed solvent on the external surface and molecules trapped among nanoparticles (Fig. 6).

Fig. 7 shows a FT-IR spectrum of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>. The bands at 1012, 1096, and 1165 cm<sup>-1</sup> correspond to asymmetric stretching P–O vibrations in the PO<sub>4</sub> tetrahedron (ν<sub>3</sub>); the band at 965 cm<sup>-1</sup> is due to symmetric stretching vibrations (ν<sub>1</sub>). Three bands of asymmetric bending vibrations (ν<sub>4</sub>) occur at 562, 589, and 625 cm<sup>-1</sup>. The symmetric bending vibration (ν<sub>2</sub>) is responsible for the absorption band at ~445 cm<sup>-1</sup> [22,23].

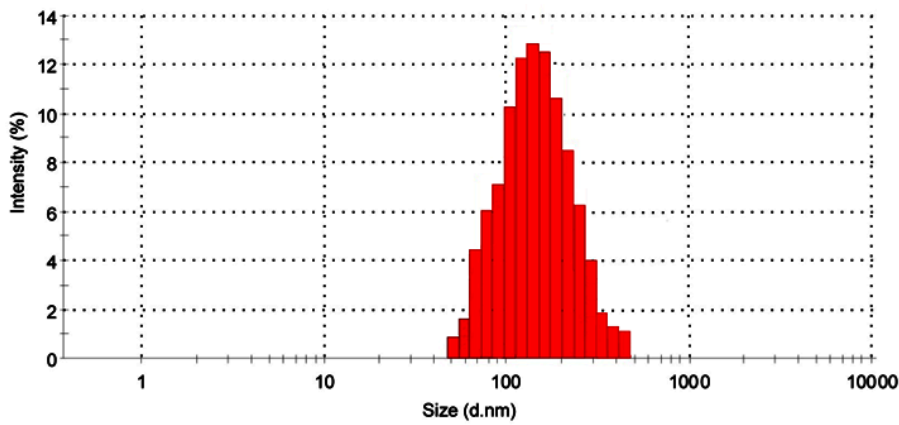


Fig. 5. DLS of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

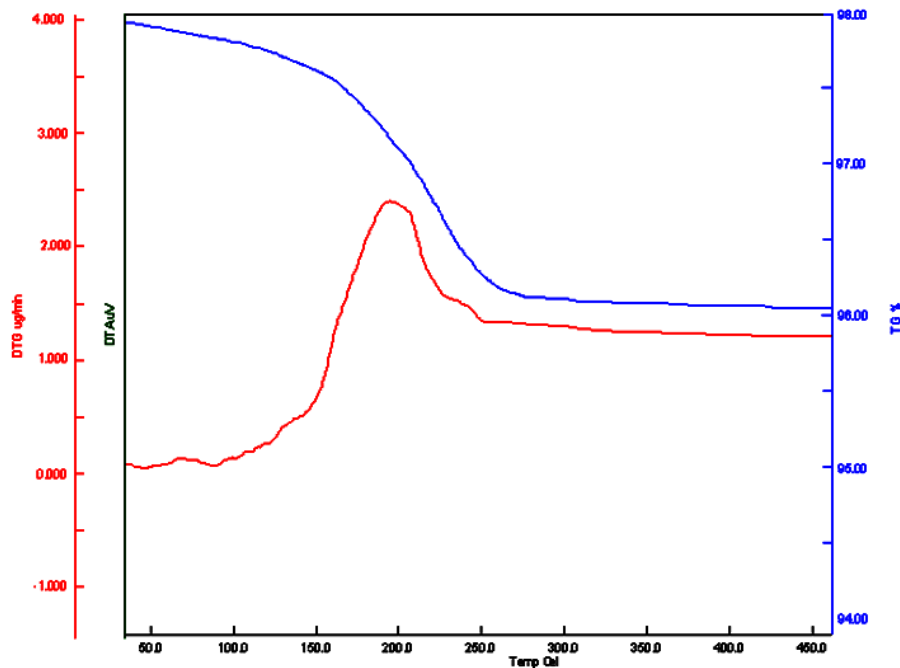


Fig. 6. DTG/TGA curve of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

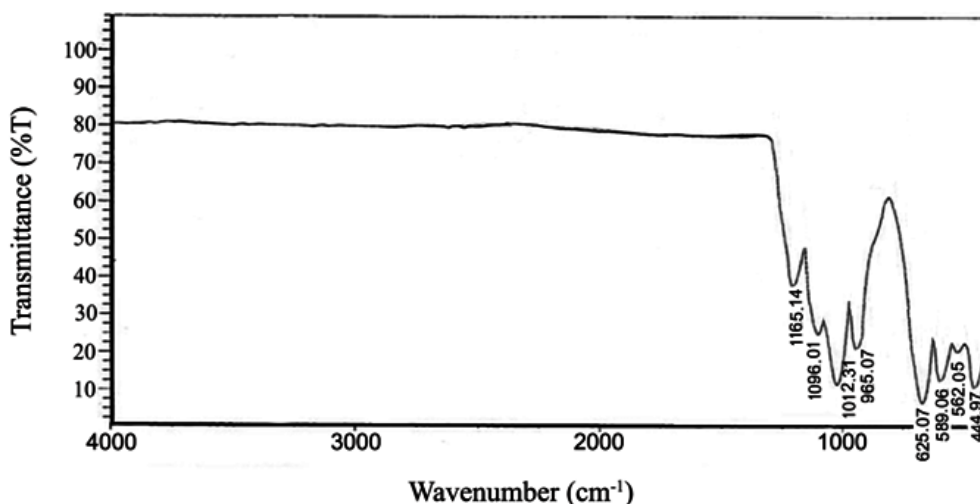


Fig. 7. FT-IR spectrum of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.

We initially attempted a pseudo five-component condensation of 4-chlorobenzaldehyde (2 mmol), ethylenediamine (1 mmol) and thioglycolic acid (2 mmol) as a model reaction in different conditions. Yields were determined in the presence of nano-SnO NPs, nano-Fe<sub>3</sub>O<sub>4</sub>, nano-CuI, nano-ZrO<sub>2</sub> NPs, nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> and the results are shown in Table 1. In this research, microwave irradiation is used as a green and complementary technique for preparation of bis-thiazolidinones. In order to investigate the effect of intensity of microwave power on reaction, the reaction

was also performed at different powers of the microwave irradiation (Table 2). The best results were obtained under microwave irradiation (400 W) in toluene and the reaction gave satisfying results in the presence of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> as catalyst. When 1, 2 and 3 mol% of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> were used; the yields were 85%, 92% and 92%, respectively. Consequently, 2 mol% of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> were expedient and excessive amount of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> did not change the yields, significantly.

Table 1. Synthesis of 3,3'-(ethane-1,2-diyl)bis(2-(4-chlorophenyl)thiazolidin-4-one) under different conditions.<sup>a</sup>

Entry	Solvent	Catalyst (mol%)	Time (min)	Yield (%) <sup>b</sup>
1	toluene (reflux)	No catalyst	400	<10
2	DMF (reflux)	nano-SnO (5)	200	42
3	toluene (reflux)	nano-Fe <sub>3</sub> O <sub>4</sub> (3)	240	30
4	toluene (reflux)	nano-CuI (4)	200	53
5	toluene (reflux)	nano-ZrO <sub>2</sub> (2)	150	65
6	CH <sub>3</sub> CN (reflux)	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2)	130	50
7	DMF (reflux)	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2)	130	55
8	toluene (reflux)	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2)	100	84
9	EtOH (400 W) <sup>c</sup>	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2)	15	62
10	CH <sub>3</sub> CN (400 W) <sup>c</sup>	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2)	15	65
11	DMF (400 W) <sup>c</sup>	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2)	10	71
12	toluene (400 W) <sup>c</sup>	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (1)	10	85
13	toluene (400 W) <sup>c</sup>	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2)	10	92
14	toluene (400 W) <sup>c</sup>	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (3)	10	92

<sup>a</sup>Reaction conditions: A mixture of 4-chlorobenzaldehyde (2 mmol), ethylenediamine (1 mmol), thioglycolic acid (2 mmol) and catalyst for various times.

<sup>b</sup>Isolated yield.

<sup>c</sup>Under microwave irradiation.

**Table 2.** Different powers of microwave irradiation for the synthesis of **4a** in toluene using nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> (2 mol%).

Entry	Power	Time (min)	Yield <sup>a</sup> (%)
1	200	10	74
2	300	10	82
3	400	10	92
4	500	10	90

<sup>a</sup>Isolated yield.

With the optimal conditions in hand, we turned to discover the efficacy of the catalyst using different aromatic aldehydes and the results are summarized in Table 3. We found that our method is compatible with diverse aromatic aldehydes such as 4-chlorobenzaldehyde, 4-methylbenzaldehyde, 4-nitrobenzaldehyde, picolinaldehyde and 4-isopropylbenzaldehyde. A variety of aromatic aldehydes, including ortho, meta and para-substituted aryl aldehydes were isolated in good to excellent yields under microwave irradiation. It has been observed that better yields are achieved with substrates having electron-withdrawing groups.

Owing to the presence of 2 and 2' equivalent stereogenic centers, bithiazolidinones can be obtained as *rac.* 2R,2'R/2S,2'S and 2R,2'S-*meso* isomers. These bithiazolidinones have been obtained, as previously

reported, by the reaction of mercaptoacetic acid with N,N'-dibenzylideneethylenediamines [24,25].

After workup, the crude mixture of isomers was separated by silica gel column chromatography (diethyl ether/petroleum ether in variable ratio mixtures). In general *meso* isomers eluted more slowly than corresponding racemates. The *racemate* isomer **4f** was obtained in higher yields than *meso* isomer **4f** (86 % for the *rac.* isomer **4f** and 14 % for the *meso* isomer **4f** that the crude mixture of isomers was separated by silica gel column chromatography). The ratio of isomers was determined from the <sup>1</sup>H NMR spectrum of the crude product. The <sup>1</sup>H NMR spectra of the compounds **4a-4m** (*rac.* isomer) displayed a doublet of doublets at δ 3.80-3.95 ppm due to the methylene proton HA at C-5 (-CO-CHAHB-S) because of its interaction with the geminal proton HB at C-5 (-CO-CHAHB-S) and the proton at the chiral C-2 (S-CHAR-N); doublet of doublets at δ 3.50-3.75 ppm due to the methylene proton HB at C-5 (-CO-CHAHB-S) because of its interaction with the geminal proton HA (-CO-CHAHB-S) and a diastereotopic proton Ha (-N-CHaHb-CHaCHb-N-) of the ethylene fragment. This last proton Ha (-N-CHaHb-CHaCHb-N-) displayed a doublet of doublets or a multiplet at δ 2.50-2.85 ppm because of its interaction with the geminal proton Hb (-N-CHaHb-CHaCHb-N-) and the proton HB (-CO-CHAHB-S) at the C-5.

**Table 3.** Synthesis of bis-thiazolidinones (racemate) using nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>.<sup>a</sup>

Entry	aldehyde	product	Time (min)	Yield <sup>b</sup> (%)	m.p. (°C)		Ref.
					Found	Reported	
1	4-Cl-C <sub>6</sub> H <sub>4</sub>	<b>4a</b>	10	92	150-152	285-288	[24]
2	4-Me-C <sub>6</sub> H <sub>4</sub>	<b>4b</b>	15	85	158-160	—	
3	C <sub>6</sub> H <sub>5</sub>	<b>4c</b>	10	88	155-157	152-155	[8]
4	4-NO <sub>2</sub> -C <sub>6</sub> H <sub>4</sub>	<b>4d</b>	10	92	164-166	—	
5	3-NO <sub>2</sub> -C <sub>6</sub> H <sub>4</sub>	<b>4e</b>	10	90	222-224	—	
6	pyridin-2-yl	<b>4f</b>	10	86	170-172	167-169	[25]
7	pyridin-3-yl	<b>4g</b>	15	85	191-193	198-200	[25]
8	pyridin-4-yl	<b>4h</b>	10	87	221-223	224-225	[25]
9	2-Cl-C <sub>6</sub> H <sub>4</sub>	<b>4i</b>	10	90	143-145	210-211	[24]
10	4-isopropyl-C <sub>6</sub> H <sub>4</sub>	<b>4j</b>	15	83	163-165	—	
11	3-Cl-C <sub>6</sub> H <sub>4</sub>	<b>4k</b>	10	89	198-200	195-197	[24]
12	4-F-C <sub>6</sub> H <sub>4</sub>	<b>4l</b>	10	92	228-230	227-229	[24]
13	3-Me-C <sub>6</sub> H <sub>4</sub>	<b>4m</b>	15	86	185-187	—	

<sup>a</sup>Reaction conditions: A mixture of aldehydes (2 mmol), ethylenediamine (1 mmol), thioglycolic acid (2 mmol) in the presence of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> in toluene as catalyst under microwave irradiation (400 W).

<sup>b</sup>Isolated yield.

The Hb proton (-N-CHa**Hb**-CHaCHb-N-) at the aliphatic chain suffered the anisotropic effect from the near amide group or aryl substituents and it went to down field at  $\delta$  3.35-4.0 ppm appearing overlapped with HB or HA (-CO-CH**AHB**-S) as a multiplet. These germinal protons of each methylene group reside in magnetic non-equivalent environments [24,25].

The reusability of the nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> catalyst was examined for the synthesis of product **4a** and it was found that product yields decreased to a small extent on each reuse (run 1, 92%; run 2, 92%; run 3, 92%; run 4, 91%; run 5, 90%). In the recycling procedure of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub>, CHCl<sub>3</sub> was added to dilute the reaction mixture after terminating the reaction. The catalyst was insoluble in the solvent and was separated by centrifuging. The deposited catalyst was washed with ethanol 4–5 times to confirm the complete removal of any organic residuals; the catalyst was reused for further catalytic reaction cycles.

To compare the efficiency of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> with the reported catalysts for the synthesis of bis-thiazolidinones, we have tabulated the results in Table 4. As Table 4 indicates, nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> is superior with respect to the reported catalysts in terms of reaction time, yield and conditions. In addition, our catalyst was recyclable for five times.

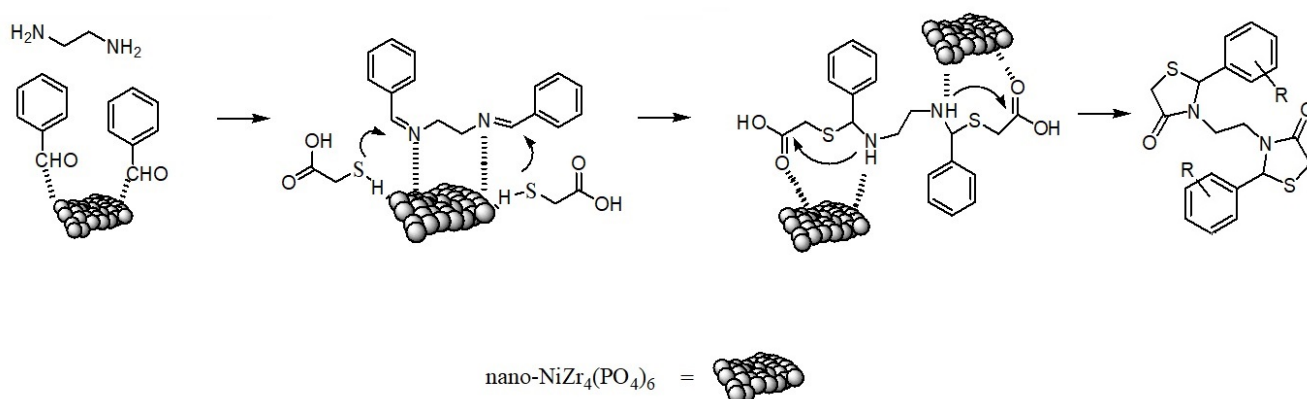
The reaction mixture is heated from the inside since the microwave energy is transferred directly to the reagents. The solid catalysts absorb microwave irradiation, thus they can serve as an internal heat source for the reactions. High catalytic activity and ease of recovery from the reaction mixture through filtration or centrifugation methods, and several reuse times without significant losses in performance are additional eco-friendly attributes of this catalytic system. As expected, the increased surface area due to small particle size increased reactivity of catalyst. This factor is responsible for the accessibility of the substrate molecules on the catalyst surface.

A probable mechanism for the synthesis of bis-thiazolidine derivatives using nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> nanoparticles, is shown in Scheme 2. A proposed mechanism is outlined via primary imine intermediate formation followed by attack of the sulfur atoms of the thioglycolic acids on the activated imine groups followed by intramolecular cyclization with the elimination of H<sub>2</sub>O giving rise to the cyclized product bis-thiazolidines. In this mechanism, the surface atoms of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> activate the C=O, C=N, S-H groups for better reaction with nucleophiles (Scheme 2).

**Table 4.** Comparison of catalytic activity of nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> with other reported catalysts.

Entry	Catalyst (condition)	Time (min)	Yield (%) <sup>a</sup>	Ref.
1	HClO <sub>4</sub> -SiO <sub>2</sub> (2.5 mol%, toluene, 100 °C)	250	52	[8]
2	Fe <sub>2</sub> O <sub>3</sub> (benzene, reflux)	300	76	[9]
2	Choline Chloride/ urea based ionic liquid (80 °C)	60	74	[10]
3	ZnCl <sub>2</sub> (dioxane, reflux)	300	65	[11]
4	nano-NiZr <sub>4</sub> (PO <sub>4</sub> ) <sub>6</sub> (2 mol%, toluene, microwave irradiation)	10	94	This work

<sup>a</sup>Isolated yield.



**Scheme 2.** Proposed reaction pathway for the synthesis of bis-thiazolidinones.



#### 4. Conclusion

In conclusion, we have developed a simple and highly efficient protocol for the synthesis of bis-thiazolidinones by one-pot pseudo-five-component condensation of araldehydes, ethylenediamine and thioglycolic acid with nano-NiZr<sub>4</sub>(PO<sub>4</sub>)<sub>6</sub> as a retrievable and robust heterogeneous catalyst under microwave irradiation in toluene. High yields, short reaction times, atom economy, reusability of the catalyst, low catalyst loading and using of microwave as clean method are some of the remarkable advantages of the present protocol.

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