

# **Poly(4‑vinylpyridinum) trinitromethanide: a useful**  and efficient heterogeneous catalyst for the synthesis **of 2,3‑dihydroquinazolin‑4(1***H***)‑one derivatives under green conditions**

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## **Abstract**

In this project, poly(4-vinylpyridinum) trinitromethanide  $[P4VPH]C(NO<sub>2</sub>)$ <sub>3</sub> was synthesized as a new heterogeneous catalyst in an easy and short way. The prepared catalyst was fully characterized using fourier transform infrared spectroscopy, energy dispersive spectrometer, feld emission scanning electron microscopy, the brunauer–emmett–teller theory, thermo gravimetric analysis and X-ray difraction. Then, the catalytic activity of this reagent was investigated in the promotion of the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives. Short reaction times, high yields of the products, carrying out the reaction under solvent-free conditions and easy separation of the catalyst from the reaction mixture are the main advantages of this method. Also, this heterogeneous catalyst can be reused up to fve times without signifcant loss of its catalytic activity.

**Keywords** Heterogeneous catalyst · Solvent-free conditions ·  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$  · Multi-component reactions · 2,3-dihydroquinazolin-4(1*H*)-ones

## **Introduction**

Heterocyclic chemistry has an important place in organic chemistry research. Quinazolinones are one of the main groups of these compounds [\[1](#page-19-0), [2](#page-19-1)]. In this regard, 2,3-dihydroquinazolin-4(1*H*)-ones and their derivatives which contain bicyclic heterocycles can show a wide range of pharmacological and bio-logical activities such as antibacterial [\[3](#page-19-2)], antitumor [[4\]](#page-19-3), antimalarial [[5\]](#page-19-4), cellular phosphorylation inhibition [\[6](#page-19-5)], antifungal and antihypertensive [[7\]](#page-19-6) ones. Also

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2,3-dihydroquinazolin-4(1*H*)-ones are key intermediates in the synthesis of quinazolin-4(3*H*)-ones which are another member of this biologically important family [[8\]](#page-19-7). There are two common methods for the synthesis of 2,3-dihydroquinazolin-4(1*H*) one derivatives; a) two-component reaction of aldehydes with 2-aminobenzimide and b) three-component reaction of aldehydes, isatoic anhydride and ammonium acetate. Various catalysts including, sulfonated porous carbon (SPC) [\[9](#page-19-8)], *N*-propylsulfamic acid supported on magnetic  $Fe<sub>3</sub>O<sub>4</sub>$  nanoparticles (MNPs-PSA) [[10\]](#page-19-9), mesoporous CoAl<sub>2</sub>O<sub>4</sub> spinel nanocrystals (nano-CoAl<sub>2</sub>O<sub>4</sub>) [\[11](#page-19-10)], Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>SO<sub>3</sub>H (MNPs) [\[12](#page-19-11)], KAl(SO<sub>4</sub>)<sub>2</sub>.12H<sub>2</sub>O (Alum) [\[13](#page-19-12)], [Al(H<sub>2</sub>PO<sub>4</sub>)<sub>3</sub>] [[14\]](#page-19-13), *L*-proline nitrate [\[15](#page-19-14)], aluminum methane sulfonate [\[16](#page-19-15)] and CoFe<sub>2</sub>O<sub>4</sub>@Pr [\[17](#page-19-16)] have been used for the synthesis of this group of compounds. Each of these reported methods, despite their potential applications, are accompanied with disadvantages such as harsh conditions for the catalyst preparation, need to use of hazardous solvents, long reaction times, sensitivity of the catalyst and low efficiency. Therefore, the development of more efficient methods using easily preparable, applicable, reusable and efficient catalysts to make such heterocyclic compounds is still of interest.

In recent years, functional polymers have found wide applicability in the synthesis of organic compounds and related felds. One of the polymers that is frequently used as a support for the preparation of catalysts is poly(4-vinylpyridine) [[18–](#page-19-17)[22\]](#page-19-18). This is because of its distinguishing features such as non-toxicity, stability and nonhumidity, ease of separation, selectivity, recyclability and availability.

Based on these studies and following our previous reports regarding the use of trinitromethane, as well as considering the useful properties of poly(4-vinylpyridine), in this article we wish to introduce a new solid catalyst named as poly(4 vinylpyridinum) trinitromethanide and its use as an efficient and recyclable catalyst in the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives.

## **Experimental**

#### **General**

Chemicals were purchased from Fluka, Merck, and Aldrich Chemical Companies. Poly(4-vinylpyridine) was used to make the catalyst. Diferent aldehydes with purity percentage (97–99%), isatoic anhydride (98%) and anthranilamide (99%) have been used for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives. All the reactions are monitored by thin layer chromatography (TLC) with UV light as detecting agent and yields refer to the isolated products.

#### **Instrumentation**

The FT-IR spectra of the catalyst and products were recorded with a VERTEX 70 (Brucker, Germany) instrument using KBr pellets for the samples in the range of 4000–400 cm<sup>-1</sup>. The melting point of the obtained products were determined using an electrothermal IA9100 apparatus. The  ${}^{1}H$  NMR and  ${}^{13}C$  NMR were recorded



<span id="page-2-0"></span>**Scheme 1.** Preparation of  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ 

a 400 MHz Bruker Avance using TMS as an internal standard in  $DMSO-d<sub>6</sub>$  (Germany). X-ray difraction (XRD) measurements were performed at room temperature on difractometer Model XRD 6000, PHILIPS Xpert pro using Co-Kα radiation  $(K=1.54056 \text{ A}^{\circ})$  with voltage 40 kV and current 30 mA in a 2 h range of 10–80° with step size  $0.01^{\circ}$  and time step 1.0 s to assess the crystallinity of the catalyst (Netherlands). Thermogravimetric analyses of the catalyst (TGA) were performed on Polymer Laboratories PL-TGA thermal analysis instrument. Samples were heated from 25 to 600 °C at ramp 20 °C/ min under  $N_2$  atmosphere (America). Field emission scanning electron microscopy (FESEM) and energy dispersive spectrometer (EDS) were performed on a TESCAN model MIRA III and MIRA II respectively (Czech Republic). The adsorption–desorption isotherms and the specifc surface area ( $S<sub>BET</sub>$ ) were determined from nitrogen adsorption studies using Belsorp mini II system (Japan).

## **Synthesis of poly(4‑vinylpyridinum) trinitromethanide**

At frst, 1 mmol of trinitromethane under vacuum conditions and the obtained catalyst was washed with diethyl ether and dried in an oven to afford  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ as a yellow powder (Scheme [1\)](#page-2-0).

## **General procedure for synthesis of 2,3‑dihydroquinazolin‑4(1H)‑ones**

Method A: In order to synthesize 2,3-dihydroquinazolin-4(1*H*)-ones using a threecomponent reaction, in a 25 mL fask isatoic anhydride (1 mmol) was added to a mixture of aromatic aldehyde (1 mmol), ammonium acetate (1.2 mmol) and 20 mg of the catalyst  $[11]$  $[11]$ .

Method B: In another way, the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives were achieved through the reaction of two components of aldehydes with anthranilamide in the presence of 20 mg of the catalyst [\[15](#page-19-14)].

After addition of the reactants in both of the mentioned methods, the reaction mixture was stirred at 120 $\degree$ C under solvent-free conditions and the advancement of the reaction was checked by TLC [*n*-hexane: EtOAc (7:3)]. After the completion of the reaction and in order to separate the product and the catalyst, hot ethanol was added to the reaction container. Since the formed product is soluble in ethanol and



<span id="page-3-0"></span>**Fig. 1** FT-IR spectrum of P4VP and  $[P4VPH]C(NO<sub>2</sub>)$ <sub>3</sub>

the catalyst was insoluble in it, the catalyst was easily separated by fltration and the fnal product is obtained by recrystallization from ethanol.

## *The spectral (IR, 1 HNMR and 13CNMR) data of a new compound is as follow:*

2-(4-methylthiophenyl)-2,3-dihydroquinazolin-4(1*H*)-one; m.p.  $=$  209–210  $°C$ ; FT-IR (KBr): 3429, 3298, 3191, 2922, 1657, 1611, 1507, 1434, cm−1; 1 H NMR (DMSO-d6, 400 MHz): δ=2.47 (s, 3H, CH3), 5.72 (s, 1H, CH), 6.68 (t, *J*=7.6, 1H, Ar), 6.74 (d, *J*=7.6, 1H, Ar), 7.09 (s, 1H, NH), 7.22–7.29 (m, 3H, Ar), 7.43 (d, *J*=6.4, 2H, Ar), 7.61 (dd, *J<sub>1</sub>*=1.6, *J<sub>2</sub>*=2, 1H, Ar), 8.29 (s, 1H, NH) ppm.; <sup>13</sup>C NMR  $(DMSO-d<sub>6</sub>, 100 Mz)$ :  $\delta = 14.61, 66.03, 114.39, 114.94, 117.12, 125.63, 127.32,$ 127.42, 133.30, 138.15, 138.42, 147.79, 163.58.



<span id="page-4-0"></span>**Fig. 2** TGA curve of P4VP and  $[P4VPH]C(NO<sub>2</sub>)$ <sub>3</sub>



<span id="page-4-1"></span>**Fig. 3**  $XRD$  patterns of P4VP and  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ 

## **Results and discussion**

## **Characterization of the catalyst**

Several techniques such as FT-IR, TGA, XRD, FESEM, EDS and BET were used for the characterization of the catalyst.

## **FT‑IR analysis**

The FT-IR spectrum of poly (4-vinylpyridine) and of poly(4-vinylpyridinum) trinitromethanide are shown in Fig. [1.](#page-3-0) The broad band at around 3428  $cm^{-1}$  was observed in the spectrum of the polymer and the synthesized catalyst, which is related to the absorbed moisture [\[23](#page-19-19)]. In the FT-IR spectrum of poly (4-vinylpyridine), the peak in the region of  $1410 \text{ cm}^{-1}$  corresponds to the stretching vibrations of C–N and the peak in the region of 1603 cm−1 is assigned to the carbon–carbon double bond vibrations in the aromatic ring [[24\]](#page-19-20). In addition, the stretching vibrations related to the aliphatic and aromatic C-H have appeared in the region of 2927 and 3027 cm−1, respectively [[24\]](#page-19-20). In the spectrum of the synthesized catalyst, the peaks related to the symmetric and asymmetric tensile vibrations of the  $NO<sub>2</sub>$  group have been appeared in the region of 1381 cm<sup>-1</sup> and 1633 cm<sup>-1</sup>, respectively [[25\]](#page-19-21).



<span id="page-5-0"></span>**Fig.** 4 FESEM images of P4VP  $(a, b)$  and  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub> (c, d)$ 



<span id="page-6-0"></span>**Fig. 5** The EDS profiles of  $[P4VPH]C(NO<sub>2</sub>)$ <sub>3</sub>

Also, the absorption peak at  $3432 \text{ cm}^{-1}$  corresponds to the stretching vibrations of N–H [[24\]](#page-19-20). It should be mentioned that reduction in the number of peaks in the FT-IR spectra of the prepared catalyst can be related to the formation of the ionic complex which reduces its vibrational modes. The set of the spectral information expressed can be a confrmation of the formation of the desired catalyst.

#### **Thermal analysis**

In the next step, the thermogravimetric analysis (TGA) is used to determine the thermal stability of poly (4-vinylpyridine) and the synthesized catalyst (Fig. [2](#page-4-0)). As can be seen in the fgure of the P4VP, the frst weight loss is occured at temperature



<span id="page-6-1"></span>**Fig.** 6 The N<sub>2</sub> adsorption–desorption isotherm (**a**), and pore size distribution (**b**) of  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ 

of about 100  $\degree$ C, which can be attributed to the loss of the adsorbed water. In the second weight loss, which occurs at temperature around 330–425 °C, the polymer is completely demolished. Analysis of the TGA curve of the synthesized catalyst shows about 6% weight loss at about 30–100  $\degree$ C, which can be attributed to the removal of the adsorbed moisture. The second weight loss occurs at 150–230 °C which can be due to the removal of the trinitromethanide group. Finally, the third weight which occurs at temperature of  $250-400$  °C can be a result of the complete decomposition of the catalyst.

Entry	Catalyst (mg)	Solvent	Temperature $(^{\circ}C)$	Time (min.)	Yield $(\%)^b$
1		No solvent	r.t	120	Trace
$\overline{2}$		No solvent	100	120	Trace
3	40	No solvent	80	17	92
$\overline{4}$	40	No solvent	100	10	95
5	40	No solvent	120	8	97
6	20	No solvent	120	8	97
7	20	CH <sub>3</sub> CN	r.t	120	Trace
8	20	CH <sub>3</sub> CN	Reflux	120	30
9	20	EtOH	r.t	120	Trace
10	20	EtOH	Reflux	120	70
11	20	H <sub>2</sub> O	r.t	120	Trace
12	20	H <sub>2</sub> O	Reflux	120	50
13	20	H <sub>2</sub> O/EtOH	70	120	60

<span id="page-7-0"></span>**Table 1** Optimization of the reaction conditions in the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one catalyzed by  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ <sup>a</sup>

<sup>a</sup>Reaction conditions: 4-chlorobenzaldehyde (1 mmol), isatoic anhydride (1 mmol) and ammonium acetate (1.2 mmol)

b Isolated yields



<span id="page-7-1"></span>**Scheme 2.** Optimized conditions for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones using three component method

Entry	Substrate	Product	Time (min.)	Yield $(\%)$ a	Melting Point	
					Found	Rreported
$\,1\,$	<b>CHO</b>	ဂူ ŅH H	$\,$ 8 $\,$	97	198-200	199-201 [11]
$\overline{c}$	<b>CHO</b>	NH Ħ	$\,8\,$	95	$178 - 180$	185-186 [26]
$\overline{\mathbf{3}}$	<b>CHO</b>	о NH C	$\sqrt{6}$	96	193-194	199-201[27]
$\overline{\mathbf{4}}$	<b>CHO</b>	NH	15	92	$170 - 171$	$167 - 169$ [17]
$\sqrt{5}$	<b>CHO</b>	NН	$\sqrt{ }$	95	190-191	195-197 [28]
$\epsilon$	<b>CHO</b> Br	NH	$\,$ 8 $\,$	96	$175 - 178$	174-176 [17]
$\boldsymbol{7}$	<b>CHO</b>	NH	25	92	188-190	199-200 [29]
$\,$ 8 $\,$	<b>CHO</b>	Ĥ OН	$15\,$	95	$>300$	$>$ 300 [30]

<span id="page-8-0"></span>**Table 2** Synthesis of various 2,3-dihydroquinazolin-4(1*H*)-ones catalyzed by [P4VPH]C(NO<sub>2</sub>)<sub>3</sub>



## **Table 2** (continued)



#### **Table 2** (continued)

a Isolated yields



<span id="page-10-0"></span>**Scheme 3.** Optimized conditions for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones using the reaction between aldehydes and anthranilamide

## **Powder X‑ray difraction analysis**

The XRD patterns of the polymer and the synthesized catalyst are compared in Fig. [3.](#page-4-1) As shown in this fgure, the attachment of the nitro group causes some changes in the difraction patterns of the polymer. In the diagram related to the polymer, a strong peak at 2*θ* around 20° and a weak peak at 2*θ* around 40° have appeared. After the reaction of the polymer with trinitromethane, the location of the frst peak has changed and its intensity has also decreased (2*θ* around 23°). Also, the week peak at 2*θ* around 40° in the XRD of the catalyst is relatively disappeared.

	Entry Substrate	Product	Time (min.)	ັ່ Yield(%) $\rm{a}$	$\mathbf{r}$ د ر <b>Melting Point</b>	
					$\operatorname{Found}$	Rreported
$1\,$	<b>CHO</b>	ō ŃН	$\overline{4}$	98		202-204 203-206 [36]
$\sqrt{2}$	<b>ÇHO</b>		$\overline{4}$	95		177-179 185-186 [26]
$\sqrt{3}$	CHO	VН	$\sqrt{5}$	95		205-207 205-206 [36]
$\overline{\mathbf{4}}$	<b>ÇHO</b>		$10\,$	93		162-164 167-169 [17]
$\mathfrak s$	<b>CHO</b>		$\sqrt{6}$	95		190-192 195-197 [28]
$\sqrt{6}$	<b>ÇHO</b>	IH	$\boldsymbol{9}$	94		175-176 174-176 [17]
$\boldsymbol{7}$	<b>CHO</b>		$\overline{\mathbf{4}}$	96	$>300$	$>$ 300 [30]
$\,$ 8 $\,$	<b>ÇHO</b>	OН	$\boldsymbol{7}$	95		200-202 206-207 [31]
9	<b>ÇHO</b> OН	VН <b>OH</b>	$\sqrt{5}$	92		288-290 280-281 [15]

<span id="page-11-0"></span>**Table 3** Synthesis of various 2,3-dihydroquinazolin-4(1*H*)-ones catalyzed by  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ 

#### **Table 3** (continued)





#### **Table 3** (continued)

a Isolated yields

#### **Field emission scanning electron microscopy analysis**

To investigate the surface morphology and size distribution, feld emission scanning electron microscopy analysis with diferent magnifcations was used. According to the displayed FESEM images, when the polymer reacts with trinitromethane, the size of the synthesized catalyst particles has increased compared to the initial polymer. This increase in size can be due to dipole–dipole attraction between particles (Fig. [4](#page-5-0)).

## **Energy dispersive spectrometer analysis**

Figure [5](#page-6-0) shows the elemental analysis of poly(4-vinylpyridinum) trinitromethanide. The EDS data of the synthesized catalyst confrms the attendance of the expected elements in the structure of the catalyst including nitrogen, carbon and oxygen with an average weight percentage of 29.48, 43.94 and 26.58%, respectively.



<span id="page-14-0"></span>**Scheme 4.** The proposed mechanism of the studied reactions



<span id="page-14-1"></span>**Fig. 7** Reusability of the catalyst



<span id="page-15-0"></span>**Fig. 8** The EDS profiles of recovered  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ 

#### **Brunauer–emmett–teller theory analysis**

BET surface area, pore volume and pore size distributions were characterized by nitrogen adsorption/desorption experiment. The  $N<sub>2</sub>$  adsorption/ desorption isotherms related to the catalyst are shown in Fig. [6a](#page-6-1). The Brunauer‐Emmet‐Teller (BET) surface areas, total pore volume and Barret‐Joyne‐Halendu (BJH) pore diameter were 4.46 m<sup>2</sup> g<sup>-1</sup>, 0.005 cm<sup>3</sup> g<sup>-1</sup> and 4.44 nm, respectively. Also, the curve of pore size distribution by utilizing the BJH model is shown in Fig. [6](#page-6-1)b.

## **Catalytic activity**

After preparation and identifcation of the poly(4-vinylpyridinum) trinitromethanide catalyst, its catalytic activity was investigated in the preparation of 2,3-dihydroquinazolin-4 $(1H)$ -one derivatives. For this purpose, the reaction between 4-chlorobenzaldehyde, isatoic anhydride and ammonium acetate as a model one was studied at different temperatures (80–120  $^{\circ}$ C) in the presence of different amounts of the catalyst. The intended reaction was performed well at 120 °C in the presence of 20 mg of the catalyst. To check the efect of the solvent and also solvent-free conditions, the model reaction was also checked in acetonitrile, ethanol, water and water/ ethanol and in the absence of solvent in the presence of 20 mg of the catalyst. According to the results collected in Table [1,](#page-7-0) this reaction is performed with high efficiency in the absence of solvent (Table [1,](#page-7-0) entry  $6$ ) (Scheme [2](#page-7-1)).

After the optimization studies, a wide range of aromatic aldehydes, including electron-withdrawing and electron-donating groups, were used for the synthesis of their corresponding 2,3-dihydroquinazolin-4(1*H*)-one derivatives. The results of this



<span id="page-16-0"></span>**Fig. 9** FT-IR spectra of fresh and recovered  $[P4VPH]C(NO<sub>2</sub>)$ <sub>3</sub>

study are shown in Table [2.](#page-8-0) As it is clear, using this method, all the products were obtained with 90–98% yields during short times.

In the next step and after the good results obtained from the three-component condensation of diferent aldehydes, ammonium acetate and isatoic anhydride in the presence of  $[P4VPH]C(NO<sub>2</sub>)<sub>3</sub>$ , we have decided to obtain 2,3-dihydroquinazolin-4(1*H*)-one derivatives via two-component reaction between aldehydes and anthranilamide under the same optimal conditions. This method was also resulted in the desired products during very short period of times with high efficiency (Scheme [3](#page-10-0), Table [3\)](#page-11-0).

The possible mechanism for the synthesis of 2,3-dihydroquinazolin-4(1*H*) one derivatives under the catalytic efect of poly(4-vinylpyridinum) trinitromethanide for both of the synthetic methods is shown in Scheme [4.](#page-14-0) In method A, the catalyst activates the carbonyl group of isatoic anhydride. In continue this molecule can be attacked by ammonium acetate and after the decarboxylation reaction, the intermediate (I) is produced. From this point onwards, the progress



<span id="page-17-0"></span><sup>a</sup>Isolated yields

of the reaction is the same in both of the synthetic methods, so that, the aldehyde activated by the catalyst is attacked by the produced intermediate (I). After removal of a molecule of water, followed by tautomerization the intermediate (II) produces the intermediate (III) which can be converted to the fnal product during an intramolecular cyclization.

As it clear, the ability to recover and reuse is one of the most important features of a heterogeneous catalyst. In the present method to investigate this important feature for poly(4-vinylpyridinum) trinitromethanide catalyst, the synthesis of 2-(4-chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (Table [3,](#page-11-0) entry1) was selected as a model one. After the completion of the reaction, hot ethanol was added to the reaction mixture. The product was dissolved in hot ethanol and the catalyst remained insoluble. After fltration, the recovered catalyst was washed with diethyl ether and dried in an oven to be able to use in the next steps. This catalyst was able to enhance the model reaction for at least 5 runs with excellent efficiency in short period of times (Fig.  $7$ ). Next, in order to check the stability of the recycled catalyst, EDS and FTIR analyses were performed on it (Figs. [8](#page-15-0) and [9](#page-16-0)). The results well prove the good reusability of the catalyst.

The efectiveness of the poly(4-vinylpyridinum) trinitromethanide catalyst and other catalysts used in the synthesis of 2-(4-chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one (Table [2](#page-8-0), entry1) is compared in Table [4](#page-17-0). This comparison shows that the catalyst is in a good position compared to the previous methods in terms of time and yield. Also, the use of cheap catalyst and solvent-free conditions are the other features of the method used in this research compared to the other mentioned methods.

## **Conclusions**

This research focused on design and synthesis of poly(4-vinylpyridinum) trinitromethanide as a recyclable and efficient heterogeneous catalyst. The formation of the synthesized catalyst was confrmed by several techniques including FT-IR, TGA, XRD, FESEM, EDS and BET analysis. Poly(4-vinylpyridinum) trinitromethanide was used as an environmentally friendly catalyst for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives under solvent-free conditions. There are several advantages for this method, including high yields of the reaction products, short reaction times, use of non-toxic and inexpensive reagents, easy work-up produce and reusability and ease of handling of the catalyst.

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**Authors' contributions** All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by LNN, FS and OGJ. The frst draft of the manuscript was written by LNN and all authors commented on previous versions of the manuscript. All authors read and approved the fnal manuscript.

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**Data availability** All data generated or analyzed during this study are included in this published article [and its supplementary information fles].

#### **Declarations**

**Competing interests** The authors declare that they have no competing interests.

**Ethics approval and consent to participate** Not applicable.

**Consent for publication** Not applicable.

#### **Reference:s**

- <span id="page-19-0"></span>1. J. Safari, S. Gandomi-Ravandi, J. Mol. Catal A: Chem. **371**, 135 (2013)
- <span id="page-19-1"></span>2. I. Khan, A. Ibrar, N. Abbas, A. Saeed, Eur. J. Med. Chem. **76**, 193 (2014)
- <span id="page-19-2"></span>3. R.J. Alaimo, H.E. Russell, J. Med. Chem. **15**, 335 (1972)
- <span id="page-19-3"></span>4. M.J. Hour, L.J. Huang, S.C. Kuo, Y. Xia, K. Bastow, Y. Nakanishi, E. Hamel, K.H. Lee, J. Med. Chem. **43**, 4479 (2000)
- <span id="page-19-4"></span>5. P. Verhaeghe, N. Azas, M. Gasquet, S. Hutter, C. Ducros, M. Laget, S. Rault, P. Rathelot, P. Vanelle, Bioorg. Med. Chem. Lett. **18**, 396 (2008)
- <span id="page-19-5"></span>6. D.W. Fry, A.J. Kraker, A. McMichael, L.A. Ambroso, J.M. Nelson, W.R. Leopold, R.W. Connors, A.J. Bridges, Science **265**, 1093 (1994)
- <span id="page-19-6"></span>7. X. Cheng, S. Vellalath, R. Goddard, B. List, J. Am. Chem. Soc. **130**, 15786 (2008)
- <span id="page-19-7"></span>8. S.E. López, M.E. Rosales, N. Urdaneta, M.V. Godoy, J.E. Charris, J. Chem. Res. **2000**, 258 (2000)
- <span id="page-19-8"></span>9. A. Shokrolahi, A. Zali, M.A. Zarei, K. Esmaeilpour, Iran. J. Catal. **2**, 91 (2012)
- <span id="page-19-9"></span>10. A. Rostami, B. Tahmasbi, H. Gholami, H. Taymorian, Chin. Chem. Lett. **24**, 211 (2013)
- <span id="page-19-10"></span>11. J. Safaei-Ghomi, R. Teymuri, Iran. J. Catal. **11**, 113 (2021)
- <span id="page-19-11"></span>12. M.A. Bodaghifard, S. Safari, J. Coord. Chem. **74**, 1613 (2021)
- <span id="page-19-12"></span>13. M. Dabiri, P. Salehi, S. Otokesh, M. Baghbanzadeh, G. Kozehgary, A.A. Mohammadi, Tetrahedron Lett. **46**, 6123 (2005)
- <span id="page-19-13"></span>14. H.R. Shaterian, A.R. Oveisi, M. Honarmand, Synth. Commun. **40**, 1231 (2010)
- <span id="page-19-14"></span>15. S.P. Bahekar, N.D. Dahake, P.B. Sarode, H.S. Chandak, Synlett **26**, 2575 (2015)
- <span id="page-19-15"></span>16. Z. Song, L. Liu, Y. Wang, X. Sun, Res. Chem. Intermed. **38**, 1091 (2012)
- <span id="page-19-16"></span>17. T. Tamoradi, S.M. Mousavi, M. Mohammadi, New J. Chem. **44**, 3012 (2020)
- <span id="page-19-17"></span>18. B. Tamami, B.K. Parvanak, Iran. Polym. J. **18**, 191 (2009)
- 19. J. Albadi, A. Mansournezhad, M. Darvishi-Paduk, Chin. Chem. Lett. **24**, 208 (2013)
- 20. L.N. Nasirmahale, F. Shirini, O. Goli-Jolodar, H. Tajik, Polycyclic Aromat. Compd. **40**, 1059 (2018)
- 21. F. Shirini, N.G. Khaligh, O.G. Jolodar, Dyes Pigm. **98**, 290 (2013)
- <span id="page-19-18"></span>22. F. Shirini, M. Abedini, R. Pourhasan, Dyes Pigment. **99**, 250 (2013)
- <span id="page-19-19"></span>23. R.J. Kalbasi, N. Mosaddegh, J. Porous Mater. **19**, 557 (2012)
- <span id="page-19-20"></span>24. N. Sahiner, A.O. Yasar, N. Aktas, Int. J. Hydrog, Energy **41**, 20562 (2016)
- <span id="page-19-21"></span>25. J. Afsar, M.A. Zolfgol, A. Khazaei, D.A. Alonso, A. Khoshnood, Y. Bayat, A. Asgari, Res. Chem. Intermed. **44**, 7595 (2018)
- <span id="page-19-22"></span>26. A. Sahu, S. Mishra, P. Sahu, A. Gajbhiye, R.K. Agrawal, Curr. Organocatalysis **5**, 137 (2018)
- <span id="page-19-23"></span>27. F. Tamaddon, M. Kazemivarnamkhasti, Synlett **27**, 2510 (2016)
- <span id="page-19-24"></span>28. J. Safari, S. Gandomi-Ravandi, J. Mol. Catal. A: Chem. **390**, 1 (2014)
- <span id="page-19-25"></span>29. J. Chen, D. Wu, F. He, M. Liu, H. Wu, J. Ding, W. Su, Tetrahedron Lett. **49**, 3814 (2008)
- <span id="page-19-26"></span>30. V.B. Labade, P.V. Shinde, M.S. Shingare, Tetrahedron Lett. **54**, 5778 (2013)
- <span id="page-19-27"></span>31. S.G. Zhang, Z.B. Xie, L.S. Liu, M. Liang, Z.G. Le, Chin. Chem. Lett. **28**, 101 (2017)
- <span id="page-19-28"></span>32. H.R. Memarian, S. Ghahremani, Z. für Naturforsch, B. J. Chem. Sci. **72**, 403 (2017)
- <span id="page-19-29"></span>33. M. Nikpassand, L. ZareFekri, K.F. Sina, S.Z. Abed, O. Marvi, Russ. J. Gen. Chem. **85**, 1959 (2015)
- <span id="page-19-30"></span>34. S.D. Dindulkar, J. Oh, V.M. Arole, Y.T. Jeong, C. R. Chim. **17**, 971 (2014)
- <span id="page-20-0"></span>35. B.B.F. Mirjalili, Z. Zaghaghi, A. Monfared, J. Chin. Chem. Soc. **67**, 197 (2020)
- <span id="page-20-1"></span>36. Z. Tajik, H. Ghafuri, N. Ghanbari, P. Hanifehnejad, Res. Square **2021**, 1 (2021)
- <span id="page-20-2"></span>37. F. Russo, M. Chelardoni, Ann. Chim. (Rom) **56**, 839 (1966)
- <span id="page-20-3"></span>38. C.S. Rajput, S. Srivastava, A. Kumar, A. Pathak, Tetrahedron Lett. **65**, 152791 (2021)
- <span id="page-20-4"></span>39. M.T. Maghsoodlou, N. Hazeri, E. Fereidooni, S. Salahi, N. Mahmoudabadi, N. Khorshidi, J. Aboonajmi, M. Lashakri, Iran. J. Catal. **5**, 245 (2015)
- <span id="page-20-5"></span>40. A. Dutta, P. Trivedi, A. Kulshrestha, A. Kumar, V. Chaturvedi, D. Sarma, Appl. Organomet. Chem. **35**, 6116 (2021)
- <span id="page-20-6"></span>41. S. Karhale, D. Survase, R. Bhat, P. Ubale, V. Helavi, Res. Chem. Intermed. **43**, 3915 (2017)
- <span id="page-20-7"></span>42. M.A. Bodaghi Fard, A. Mobinikhaledi, M. Hamidinasab, Synth. React. Inorg. Met. **571**, 567 (2014)

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