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Efficient synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones using nano-sized protonated ZSM–5 as a biodegradable and reusable solid acid catalyst

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ABSTRACT

Nano-sized protonated ZSM–5 (H-ZSM-5nanozeolite) was successfully synthesized by the hydrothermal method using the bagasse ash (BGA) as a new silica source. Cultivated BGA in the south of the Caspian Sea (Mazandaran province, Iran) was used for extracting silica powder. H-ZSM-5 was characterized by Fourier transform infrared (FT-IR), X-ray diffraction (XRD), scanning electronic microscopy (SEM) and Brunauer–Emmett–Teller (BET), Barrett-Joyner-Halenda (BJH) techniques. The catalytic activity of H-ZSM-5 nanozeolite was evaluated for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones via one-pot three-component reaction of aromatic aldehydes, 2-aminobenzothiazole or 2-aminobenzimidazole and isatoic anhydride under solvent-free conditions. Short reaction time, high yields, a simple experimental procedure in the absence of any toxic solvents and recovery of catalyst are the advantages of this protocol.

Keywords: H-ZSM-5 nanozeolite, 2,3-Dihydroquinazolin-4(1H)-ones, Hydrothermal, Bagasse, Catalyst.

1. Introduction

Solid acids are a widely used class of catalysts for petrochemical processes. Considering the concepts of the green chemistry of avoiding waste and toxic by-products, solid acidic catalysts become increasingly important for the production of fine, specialty and pharmaceutical chemicals [1–3]. The catalytic activity of these catalysts is related to acidic Lewis (LS) and/or Brønsted (BS) sites on their surfaces [4]. Zeolites, which have extensive industrial use as catalysts, adsorbents, and ion exchangers, are crystalline aluminosilicates with periodic arrangements of cages and channels. The most important applications are found in the field of fluid catalytic cracking (FCC), hydrocracking, isomerization, alkylation and reforming reactions [5-7]. ZSM-5 was first reported by Mobil in 1972 [8]. Many different researches on H-ZSM-5 zeolite have been conducted due to its particular structural and physical-chemical performance, shape selectivity, stability and the flexibility in tailor-making of catalyst for various reactions [9,10].

The H-ZSM-5 zeolite can be used in its H-form [11,12] or after ion exchange with different metal ions such as Nickel [13] or Zinc [14]. Pt- and Ir-exchanged H-ZSM-5 zeolites are used in catalytic hydroisomerization of n-alkanes [15]. In the catalysis community, there has been a great deal of interest in nanocrystalline zeolites due to potential improvements in catalytic activity resulting from increased surface areas and decreased diffusion path lengths [16,17]. Several industrial applications of zeolites are based upon technology adapted from the acid silica/alumina catalysts originally developed for the catalytic cracking reaction. This means that the activity requested is based on the formation of Brønsted acid sites arising from the creation of "bridging hydroxyl groups" within the pore structure of the zeolites. These "bridging hydroxyl groups" are usually formed either by ammonium or polyvalent cation exchange followed by a calcination step. The "bridging hydroxyl groups", which are protons associated with negatively charged framework oxygen's linked into alumina tetrahedral, are the Brønsted acid sites, as demonstrated in scheme 1 [18].

The H-ZSM-5 nanozeolite was synthesized from bagasse as a new silica source by the hydrothermal

Scheme 1. The structure of H-ZSM-5 nanozeolite.

method and characterized by using FT-IR, XRD, BET, BJH, SEM techniques.

Developing new multicomponent reactions (MCRs) is an area of considerable interest due to the fact that the products are produced in a single step and also the variety could be formed by changing the reaction components [19-21]. Some of the important classes of pharmaceutical compounds have been prepared through the synthesis of multi-component reactions. Among these, 2,3-dihydroquinazolin-4(1H)-one derivatives with a wide range of biological activities, play important roles in biochemical processes. They can easily be oxidized to their quinazolin-4(3H)-one analogs, which are found in some natural products [22]. Also, they are well known to diuretics, antitumor agents and herbicidal agents [23]

In this study, we disclose that H-ZSM-5 nanozeolite can be used as a novel nanocatalyst for the synthesis of 2,3-dihydroquinazolin-4(1H)-ones via the one-pot three-component reaction of aromatic aldehydes, 2-aminobenzothiazole or 2-aminobenzimidazole and isatoic anhydride (Scheme 2).

2. Experimental

2.1. Materials and apparatus

SiO₂ powder was extracted from stem cane using Kalapathy et al. method with some modifications [24]. Hydrochloric acid was purchased from Sigma. Sodium hydroxide was also purchased from Merck. Phase and crystallinity of the silica extracted from bagasse (BGA) and H-ZSM-5 nanozeolites were recorded by powder

XRD Patterns (Bruker D$_8$ diffractometer) with K$_\alpha$= Cu radiation at 40 kV and 30 mA at 2 = 5 to 50°. FT-IR spectrum was recorded at room temperature using FT-IR spectrometer (Vector 22- Bruker) in the range of 500-3500 cm^{-1} for H-ZSM-5 and 2,3-dihydro-2-aryl-4(1H)-quinazolinones, respectively. X-ray fluorescence (XRF, 8410 Rh 60 kV) was used to analyze the chemical composition of extracted silica powder in the forms of oxides. In order to indicate the morphology of nanozeolites, SEM images were done using a SEM, KYKY-EM-3200. Also, the linear portion of the Brumauere Emmette Teller (BET, model BELSORP mini 100 instruments) plots was applied to determine the surface areas of the H-ZSM-5 nanozeolite. The pore size distribution was calculated from nitrogen adsorption data using the conventional Barrette Joynere Halenda (BJH) method. The NMR spectra were recorded on a Bruker Avance DPX 300 MHz and 400 MHz instrument. The spectra were measured in DMSO-d$_6$ relative to TMS (0.00 ppm). IR spectra were recorded on a JASCO FT-IR 460 plus spectrophotometer. TLC was performed on silicagel Poly Gram SIL G/UV 254 plates.

2.2. Preparation of pure silica powder from BGA ash

In this study, for extraction of silica from BGA ash, at first BGA was washed with distilled water to remove dust and stacked particles. Subsequently, it was dried in an oven at 70°C and burned in air to obtain black ash. Then, BGA black ash is calcined in an electrical furnace at 550°C for 8 h to take out all of the organic materials to achieve white ash. The silica containing ash powder was dissolved in 2 M NaOH under vigorous stirring at 100°C for 4 h and was cooled to room temperature. Afterward, the formed sodium silicate solution was centrifuged and titrated with 1M HCl to neutralize the formation of silica gel. Silica gel was aged for 24 h at 25°C under constant environmental conditions. Xerogel was filtered and washed with distilled water several times to neutralize gel. Then, the product was dried at 80°C for 12 h. Finally, obtained xerogel was ground with mortar and pestle to obtain white amorphous silica powder. For every 1000 gr of BGA, 32 gr of silica are extracted (3.2 %).

Scheme 2. Synthesis of 2,3-dihydro-2-aryl-4(1H)-quinazolinones.

2.3. Synthesis of H-ZSM-5 nanozeolite

H-ZSM-5 nanozeolite was synthesized using silica powder extracted from BGA ash and aluminum isopropoxide as silica and aluminum sources, respectively. Also, TMAOH was used as template (structure directing agent). To prepare aluminosilicate gel, silicate and aluminate solutions were mixed in a molar ratio of Al_2O_3: 60.25 SiO_2: 0.30 Na_2O: 15.06 TMAOH: 1156.62 H_2O. At first, 0.12 g aluminum isopropoxide was dissolved in 8.5 mL solution of TMAOH (1M) with vigorous and constant stirring. Then, 11.4 mL deionized water and 0.0035 g NaOH were added to the above solution. Then, 2.3 g silica extracted from BGA was added to the obtained solution under vigorous stirring that was continued for 12 h at ambient temperature. After that, ultrasound pretreatment was done for 30 min. Afterward, the aluminosilicate gel was transferred into Teflon-lined stainless steel autoclave and hydrothermally heated at 100°C for 18 h in an oven. After this time, the mixture was filtered and product was washed with double distilled water until pH reached about 8. Finally, it was dried at 105°C overnight. After that, the dried sample was converted into ammonium form by the three-time ion exchange with 1 M NH_4Cl at 70°C for 5 h. Subsequently, the obtained sample was again filtrated, washed, and dried overnight. Finally, the dried sample was transformed into the hydrogen form through calcination at 550°C for 10 h [25].

2.4. General procedure for the synthesis of 2,3-dihydriquinazolin-4(1H) derivatives

A stirred mixture of isatoic anhydride (1 mmol) amines containing 2-aminobenzothiazole or 2-amino benzimidazole (1.1 mmol), aromatic aldehydes (1 mmol) and 0.045 g of H-ZSM-5 nanozeolite was reacted in an oil bath at 100°C for the appropriate times. Completion of the reaction was indicated by TLC. After completion of the reaction, the mixture was cooled to room temperature. The solid residue was dissolved in hot ethanol and the catalyst was filtered off. The filtrate solution was concentrated and the solid product was recrystallized in aqueous EtOH (70%) to give pure products.

Selected spectral data

*3-(2'- benzothiazolo)-2,3- dihydro- 2- (2,6- dichloro phenyl)- quinazolin-4(1H)-ones (**Table 3 Entry 10**):*

m.p.= 229-239°C. IR (KBr): $\bar{v}$= 3357, 1638, 1528, 1436, 1246, 745 cm^{-1}. ^{1}HNMR (300 MHz, DMSO-d$_6$): δ = 6.79-6.83 (2H, m), 6.94-6.96 (2H, d, *J = 7.6 Hz*), 7.31-7.53 (6H, m), 7.72-7.80 (2H, m), 8.05 (1H, d, *J = 7.6 Hz*) ppm. ^{13}CNMR (75 MHz, DMSO-d$_6$): δ = 161.9, 158.1, 148.1, 147.1, 139.0, 136.1, 133.4, 133.0, 129.1, 128.9, 128.1, 126.8, 126.8, 124.6, 122.2, 121.4, 119.1, 116.3, 114.0, 67.9 ppm.

*3- (2'- benzimidazolo)-2,3- dihydro-2- (2,4-dimethoxy phenyl)-quinazolin-4(1H)-one (**Table 3 Entry 11**):*

m.p.= 209-214°C. IR (KBr): $\bar{v}$= 3403, 3354, 1661, 1593, 1690, 1161, 740 cm^{-1}. ^{1}HNMR (300 MHz, DMSO-d$_6$): δ = 3.63 (3H, s), 3.90 (3H, s), 6.29 (1H, dd, J_1= 2.3 Hz , J_2= 8.4 Hz), 6.59 (1H, d, *J = 2.1 Hz*), 6.72-6.86 (3H, m), 7.05-7.141 (2H, m), 7.27- 7.37 (1H, m), 7.38-7.43 (2H, m), 7.51-7.58 (2H, m), 7.87(1H, d, *J= 6 Hz*) 12.56 (1H, s) ppm. ^{13}CNMR (75 MHz, DMSO-d$_6$): δ = 162.7, 160.6, 157. 5, 146.7, 146.6, 139.8, 134.8, 133.1, 128, 125.8, 121.4, 121.4, 119.6, 117.4, 117.3, 115.4, 113.6, 111.8, 103.9, 99.1, 64.6, 55.7, 55.1 ppm.

*3- (2'-benzimidazolo)-2,3- dihydro-2- (2,6-dichloro phenyl)-quinazolin-4(1H)-one (**Table 3 entry 12**):*

m.p.= 214-218°C. IR (KBr): $\bar{v}$= 3346, 1662, 1616, 1525, 1440, 1333 cm^{-1}. ^{1}HNMR (300 MHz, DMSO-d$_6$): δ = 6.80 (1H, d, *J = 7.2* Hz), 6.94(1H, t, *J = 8 Hz*), 7.29-7.55 (8H, m), 7.80 (1H, t, *J = 8 Hz*), 8.04 (1H,d, *J = 7.2 Hz*), 12.53 (1H, s) ppm. ^{13}CNMR (75 MHz, DMSO d$_6$): δ = 161.9, 158.1, 148.1, 147.1, 139.0, 136.1, 133.4, 133.1, 129.1, 128.9, 128.1, 126.8, 124.6, 122.2, 121.4, 119.1, 116.3, 114.0, 67.9 ppm.

3. Results and Discussion

3.1. Characterization of silica extracted from BGA

The chemical composition of BGA ash in the form of stable oxides was analyzed with X-ray fluorescence (XRF) (Table1). The sample consisted of SiO_2 with purity approximately 86.498 % (wt.). There were small amounts of other inorganic oxides including P_2O_5, Al_2O_3, K_2O, CaO, Fe_2O_3, Na_2O and MgO along with SiO_2. This exhibits that BGA ash component has sufficient SiO_2 to use as a silica source in the synthesis of H-ZSM-5 zeolite.

SEM images of BGA ash and the extracted SiO_2 are displayed in Fig. 1.

Table 1. Chemical compositions of BGA ash (wt %) by XRF.

| Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	K_2O	CaO	Fe_2O_3
0.220	1.202	3.380	86.498	6.075	2.019	0.348	0.240
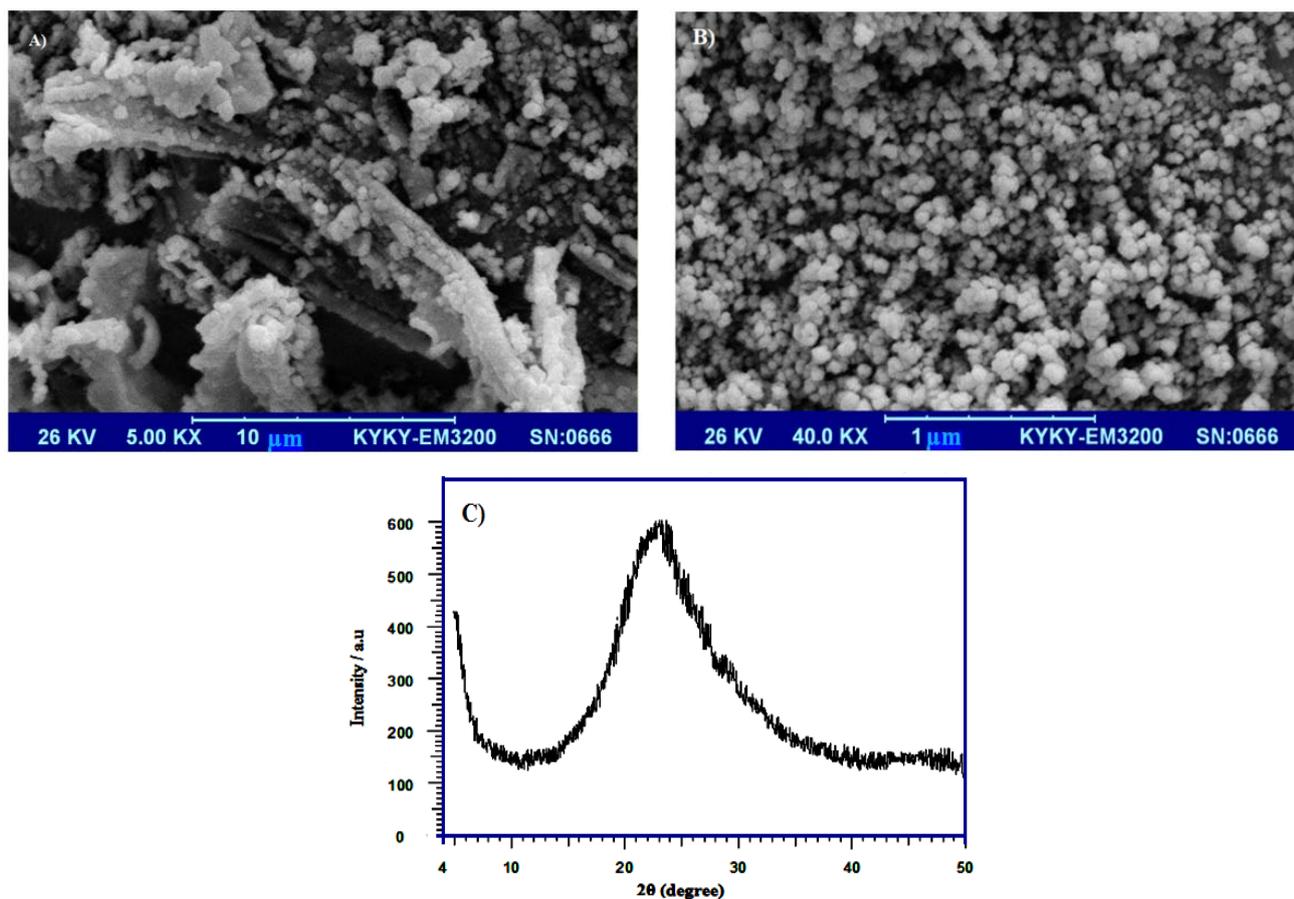
Fig. 1. A) SEM images of BGA ash and B) SiO$_2$ extracted from BGA ash C) the XRD pattern of SiO$_2$.

As it is illustrated, the SEM image of SiO$_2$ from BGA ash displays the presence of the spherical particles with an average diameter of 35 nm. Also, Fig. 1C shows the XRD pattern of silica extracted from BG ash. A broad peak with 22.4° displayed the formation of amorphous silica without impurity which is suitable for the synthesis of zeolite through the dissolution of NaOH and formation of sodium silicate.

3.2. Characterization of H-ZSM-5 nanozeolite

Fig. 2A show FT-IR spectrum of the H-ZSM-5 nanozeolite. The presence of absorption bands around 542 and 450 cm^{-1} is characteristic of the ZSM-5 crystalline structure. The absorption bands at 1220 and 542 cm^{-1} provide information on the difference between ZSM-5 zeolite and other type of zeolites. The external asymmetric stretching vibration near 1220 cm^{-1} is due to the presence of structures containing four chains of five-member rings of ZSM-5 structure. The band near 790 cm^{-1} is assigned to the symmetric stretching of external linkages and the one near 542 cm^{-1} is attributed to a structure-sensitive vibration caused by the double five-member rings of the external linkages. The absorption band near 450 cm^{-1} is due to the T-O bending

vibrations of SiO$_4$ and AlO$_4$ internal tetrahedral. The bands around 790, 1080-1200 cm^{-1} are characteristics of SiO$_4$ tetrahedron units. XRD powder pattern of the H-ZSM-5 nanozeolite is exhibited in Fig. 2B with comparison of the main peaks at 2θ = 7.9, 8.9, 23.2 and 24.5° with the reference sample [26]. The XRD pattern of the calcined synthesized zeolite is exhibited in Fig. 2B. The characteristic peaks of H-ZSM-5 zeolite, which are in accordance with the standard pattern, are observed and no peaks related to the presence of an impurity in the sample are observed in XRD. Fig. 2C demonstrates SEM of nanoparticles of H-ZSM-5 zeolite obtained at 100°C for 48 h. It consists of almost spherical particles with sizes in the range of nanometer.

Fig. 3A displays the N$_2$ adsorption/desorption isotherms of H-ZSM-5. There is a type 1 isotherm with the H$_4$ hysteresis loop in which micro-pore zeolites were shown. From BET equation and t-plot method, surface area (352.975 m^2 g^{-1}) and total pore volume (0.189 cc g^{-1}) are acquired. As can be seen in Fig. 3B, the synthesized H-ZSM-5 nanozeolite has a nearly narrow pore size distribution (BJH). As can be seen, the pore size distribution is centered at about 1.69 nm which describes the microporous nature of H-ZSM-5.
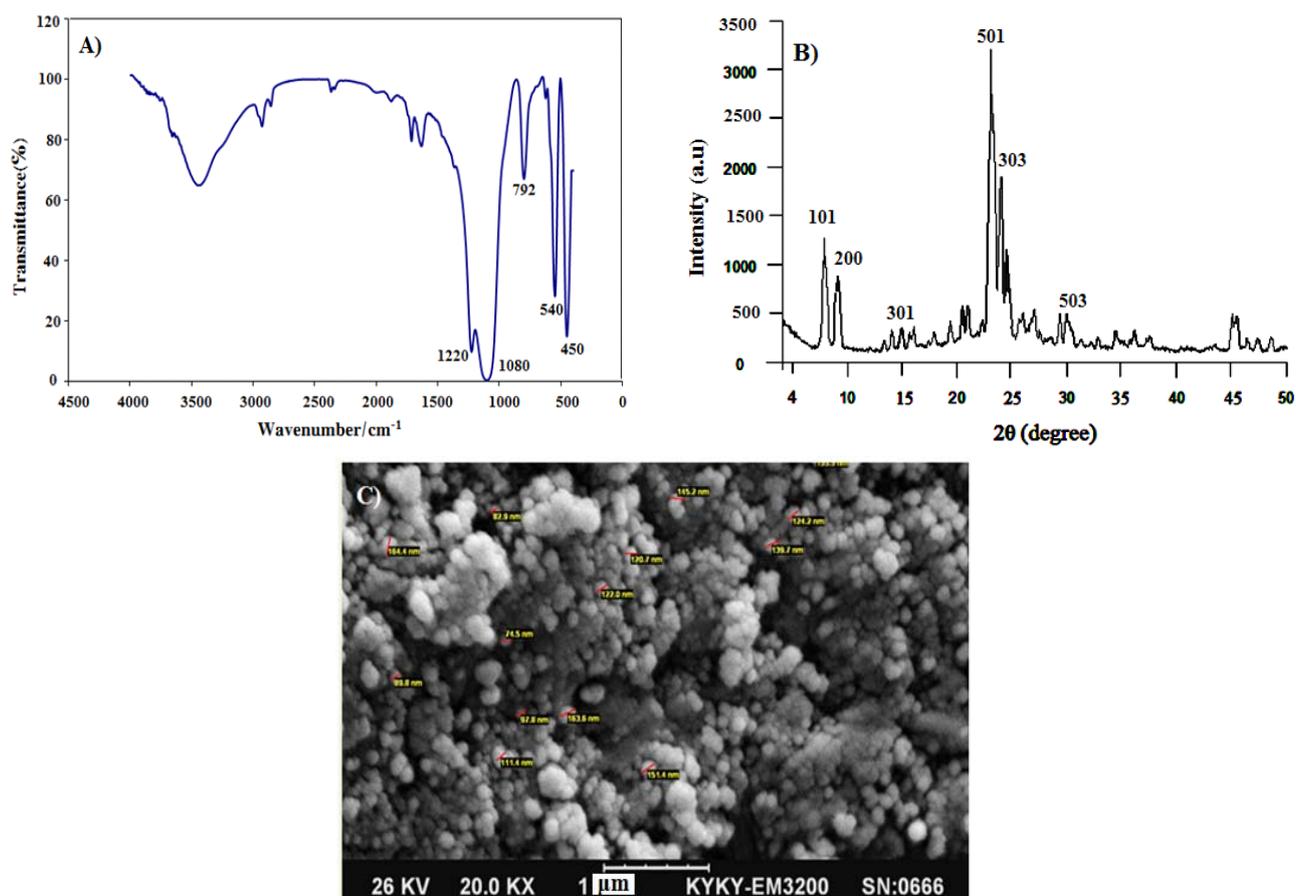
**Fig. 2.** A) FT-IR spectrum B) XRD powder pattern C) SEM spectrum of the H-ZSM-5 nanozeolite.
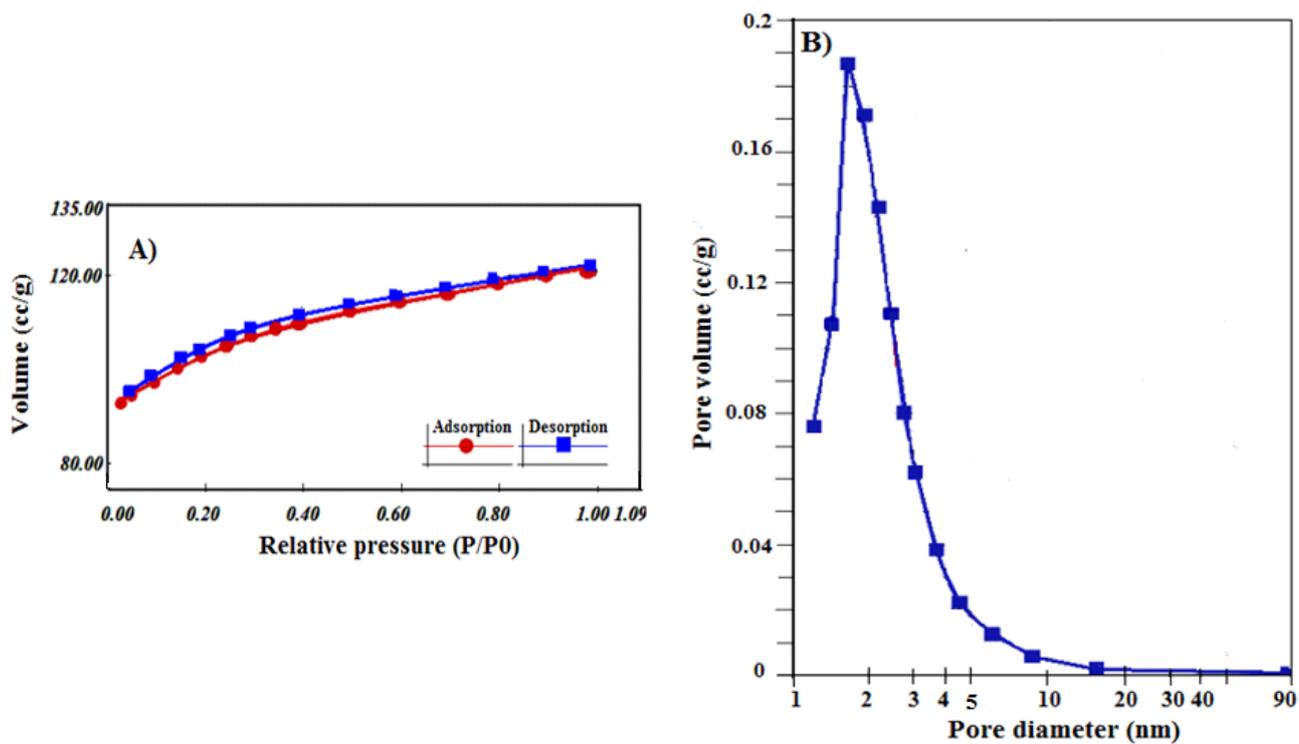
Fig. 3. A) The N_2 adsorption/desorption isotherms B) Corresponding BJH pore size distribution of H-ZSM-5 nanozeolites.

3.3. Catalytic activity of H-ZSM-5 nanozeolite

At first, to choose optimum conditions, we tried to obtain 2,3-dihydriquinazolin-4(1H)-ones from the reaction of 4-chlorobenzaldehyde (1 mmol), isatoic anhydride (1 mmol), and 2-aminobenzothiazole (1.1 mmol) as a model, under solvent-free conditions in different temperatures in the presence of a catalyst (0.02 g). Next, the model was examined at different amounts of catalyst at 100°C (Table 2). As shown in Table 2 the best result was obtained at 100°C and 0.045 g of the catalyst.

To generalize the optimum conditions, different derivatives of 3-(2′- benzothiazole)-2,3-dihydro quinazolin-4(1H)-ones were prepared by the one-pot reaction of an aromatic aldehyde, isatoic anhydride, and 2-aminobenzothiazole under solvent-free conditions (Table 3, Entry 1-9). Also, 2-aminobenzimidazole was applied instead of 2-aminobenzothiazole to obtain 3-(2′-benzimidazolo)-2,3-dihydro-2-quinazolin- 4(1H)-one derivatives in high yields (Table 3, Entry 10-12).

As shown in Table 3, aromatic aldehydes carrying both electron-donating and electron-withdrawing groups were used and in all cases the corresponding product was obtained in high yield and short reaction times.

According to the literature survey [27], the suggested mechanism for the formation of the products is shown in Scheme 3. First, the intermediate (1) was formed from the reaction of isatoic anhydride and N-nucleophilic primary amine in presence of H-ZSM-5 nanozeolite as catalyst. Then, the intermediate (2) was formed through decarboxylation reaction. The proton transfer of the intermediate (2) affords to form 2-amino-N-substituted-amide (3). Subsequently; the reaction of activated

aldehyde with (3) proceeds to prepare the imine intermediate (4). The part of amide functional group in intermediate (4) could be tautomerized in the presence of the catalyst. Thus, intermediate (5) could be prepared by an intramolecular nucleophilic attack of the amide nitrogen on the activated imine carbon, followed by a 1,5-proton transfer to yield, the 2,3-dihydroquinazolin-4(1H)-ones as products (Scheme 3).

This research suggests that an acid-base interaction of H-ZSM-5 nanozeolite as a catalyst and isatoic anhydride produces a reactive intermediate (1). The N-nucleophilic primary amine attacks on the carbonyl group of (1) to produce a reactive intermediate (2), which in turn affords (3) through decarboxylation reaction. The proton transfer of (3) could form 2-amino-N-substituted-amide (4). Subsequently, the reaction of activated aldehyde with (4) proceeds to prepare the imine intermediate (5). The part of amide functional group in intermediate (4) could be tautomerized in the presence of the catalyst. Thus, intermediate (6) could be prepared by an intramolecular nucleophilic attack of the amide nitrogen on activated imine carbon, followed by a 1,5-proton transfer to yield the 2,3-dihydroquinazolin-4(1H)-ones as products (Scheme 3).

To show the merit of the present work in comparison with other reported results in the literature, we compared results of $Zr(HSO_4)_4$ with Agar [28], 1-butyl-3-methylimidazolium bromide [bmim]Br as an ionic liquid [29], catalyst free [30], SBA-IL [31], Al $(H_2PO_4)_3$ [32] and $Zr(HSO_4)_4$ [33] as catalysts for the synthesis of 3- (2′- benzimidazolo)- 2,3- dihydro-2- (4- methyl)-quinazolin-4(1H)-one. As shown in Table 4, H-ZSM-5 nanozeolite can act as a high efficiency catalyst in short time and produce excellent yields of products.

Table 2. Optimization of the reaction conditions for the synthesis of 2,3-dihydroquinazolin-4(1H)-one derivatives in presence of H-ZSM-5 nanozeolite as catalyst.[a]

Entry	Amount of the catalyst (g)	Temperature (°C)	Time(min)	Yield (%)
1	0.02	90	20	87
2	0.02	100	19	90
3	0.02	110	18	90
4	0.02	120	16	90
5	0.03	100	18	88
6	0.045	100	15	92
7	0.050	100	14	92
8	0.055	100	13	92

[a]Reaction of 4-chlorobenzaldehyde (1 mmol), isatoic anhydride (1 mmol) and 2-aminobenzothiazole(1.1mmol) as a model reaction in presence of H-ZSM-5 nanozeolite as catalyst under thermal and solvent-free conditions

Table 3. Preparation of varieties of 2,3-dihydroquinazolin-4(1H)-one derivatives.[a]

Entry	Aldehyde	X	Product	Time (min)	Yield (%)[b]	m.p. (°C) Found	m.p. (°C) reported[c]
1		S		15	92	192-194	190-193
2		S		19	89	185-188	184-186
3		S		17	92	230-232	231-234
4		S		10	89	201-203	198-200
5		NH		11	92	255-258	253-255
6		S		15	88	237-239	233-236
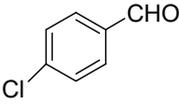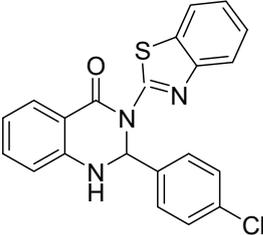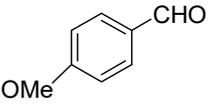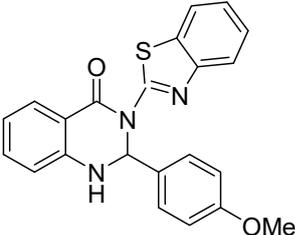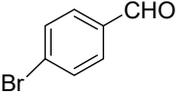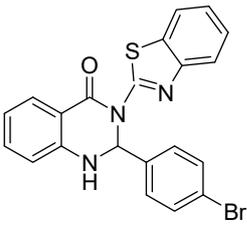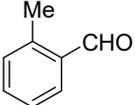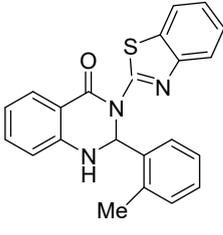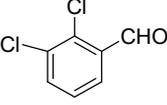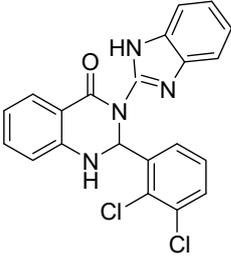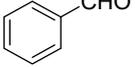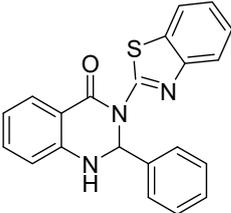
Table 3. (*Continued*).
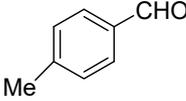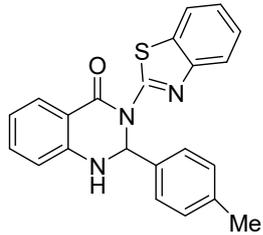
7		S		19	93	197-200	198-199
8		S		23	85	244-246	245-246
9		S		10	91	228-232	225-230
10		S		14	90	229-239	-
11		NH		15	90	209-214	-
12		NH		10	92	214-218	-
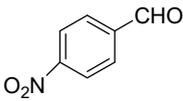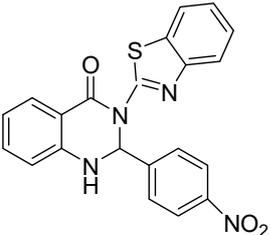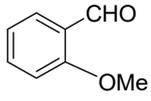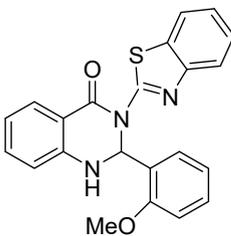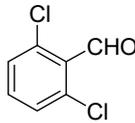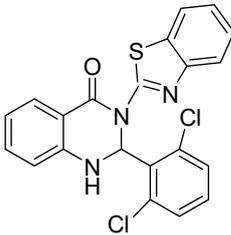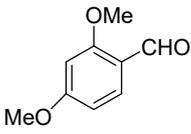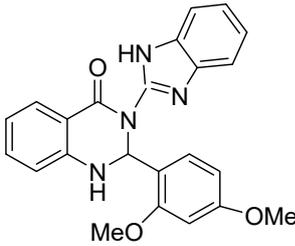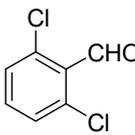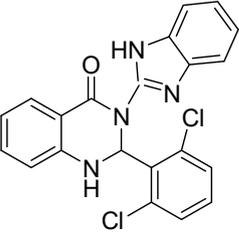
[a]Reaction of aromatic aldehyde (1 mmol), isatoic anhydride (1 mmol) and 2-aminobenzothiazole (1.1mmol) in the presence of H-ZSM-5 nanozeolite as catalyst under solvent-free conditions at 100°C.
[b]Yield refer to the isolated pure products.
[c]Melting point of products was compared with the literature [28].
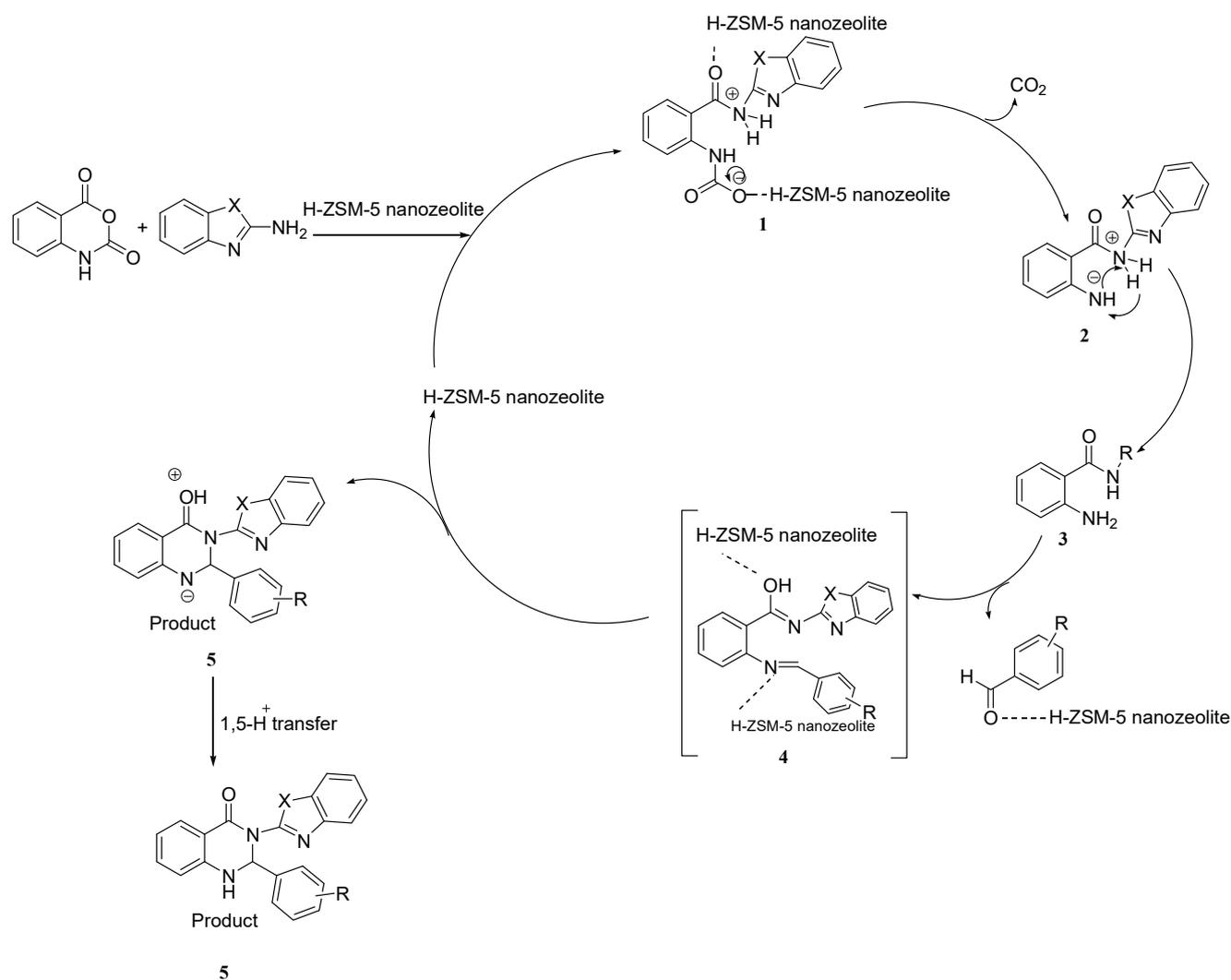
Scheme 3. Plausible mechanism for preparation of 2,3-dihydroquinazolin-4(1H)-ones using H-ZSM-5nanozeolite as catalyst.

Table 4. Comparison of our method with other methods reported in literatures, for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives.

Entry	Catalyst	Conditions	Time (min)	Yield (%)	Ref.
1	Agar	H_2O:EtOH (3:1), Reflux	35	89	[28]
2	[bmim]Br	Solvent-free, 130°C	30	91	[29]
3	Catalyst free	Solvent-free, 130°C	-	61	[30]
4	SBA-IL	Solvent-free, 130°C	20	86	[31]
5	Al $(H_2PO_4)_3$	Solvent-free, 100°C	20	83	[32]
6	Zr(HSO$_4$)$_4$	Solvent-free, 110°C	30	92	[33]
7	H-ZSM-5 nanozeolite	Solvent-free, 100°C	19	93	This work

We also investigated the recycling of the catalyst under solvent-free conditions using a model reaction between isatoic anhydride, 4-chlorobenzaldehyde, and 2-aminobenzothiazole. After completion of the reaction, the reaction was cooled to room temperature and the crude solid product was dissolved in hot ethanol. The solution was filtered for separation of the catalyst. The catalyst was washed twice (2×10 mL) using hot ethanol. Then it was dried at 100°C for 1 h. The recovered catalyst was reused four times with low decrease in activity (The yields were 92, 89, 88 and 85%, respectively).

4. Conclusions

H-ZSM-5 nanozeolite was synthesized by a hydrothermal method which is confirmed with FT-IR, XRD, SEM, BET and BJH techniques. This catalyst was successfully applied for the synthesis of the one-pot three-component reaction of isatoic anhydride, aromatic aldehydes and 2-amino benzothiazole or 2-amino benzimidazole to afford corresponding 2,3-dihydroquinazolin-4(1H)-ones under solvent-free conditions. Short reaction times, high yields, a simple experimental procedure in the absence of any toxic solvents and recovery of catalyst are the advantages of our protocol.

Acknowledgments

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