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Ultrasound-promoted one-pot three component synthesis of tetrazoles catalyzed by zinc sulfide nanoparticles as a recyclable heterogeneous catalyst

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ABSTRACT

Ultrasound irradiation was applied for the appropriate and rapid synthesis of 1-substituted tetrazoles through cyclization reaction of various primary amines, sodium azide and triethyl orthoformate. This reaction was effectively catalyzed by ZnS nanoparticles as an efficient, recoverable and reusable catalyst. Compared with conventional methods, this method has the considerable advantages such as shorter reaction times, easier work-up, purer products with high yields and mild conditions. The ZnS nanoparticles catalyst is an excellent instance to replace Brønsted acids for the preparation of 1-substituted tetrazole derivatives in very short reaction times with excellent yields. The catalyst can be recovered and reused several times without significant loss of its catalytic activity.

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1. Introduction

Tetrazoles and their derivatives have gained considerable attention because they have a wide range of applications in information recording systems in material science [1], medicinal chemistry [2], photography [3] and agriculture [4]. Pharmaceutical applications of tetrazoles include antiallergic, antimicrobial, sedatives [5], anti-inflammatory [6], anti-HIV [7], hormonal [8], lipophilic spacers [9] and diuretics activity [10]. Furthermore, a range of proline derived tetrazoles have been reported as an enantioselective catalyst in asymmetric synthesis; additionally, they play a very important role as ligands in coordination chemistry [11]. Tetrazole derivatives can be prepared by different methods. Generally, the most convenient and versatile procedure for the preparation of tetrazoles is the cycloaddition between nitriles, cyanates, cyanamides and azides [4]. Several methods have been reported for the synthesis of 1-substituted tetrazoles involve acid-catalyzed cycloaddition between hydrazoic acid and isocyanides [12]. Acid-catalyzed cycloaddition between isocyanides and trimethyl azide [13], acetic acid or trifluoroacetic acid have catalyzed cyclization between primary amines or their salts, and an orthocarboxylic acid ester and sodium azide [14,15]. Acidic ionic liquid [16], ytterbium triflate [17], zeolites [18], indium triflate [19], silica sulfuric acid [4] have catalyzed cyclization of an amine, triethyl orthoformate and sodium azide to afford tetrazoles. Unfortunately, all of these methods have some drawbacks such as long reaction times, use of expensive and toxic reagents and high boiling solvents, harsh reaction conditions, low yield, tedious work-up, and even the need for excess amounts of highly toxic and explosive hydrazoic acid [2]. Therefore, in order to overcome these drawbacks, it is necessary to develop a simple, convenient and more efficient method for the synthesis of tetrazoles.

Recently, nanostructured materials have attracted much attention because of their unique properties that distinguish them from the bulk materials [20]. ZnS is one of the very important materials applied in organic synthesis with excellent catalytic activity [3]. In 2010, Lang et al. have been used mesoporous ZnS nanospheres as a new heterogeneous catalyst for the synthesis of 5-substituted 1H-tetrazole derivatives from various nitriles and sodium azide in DMF as a solvent [3].

In recent years, ultrasound has been extensively applied as a fantastic tool for different types of chemical reactions [21]. Acoustic cavitation is a physical phenomenon that promotes chemical reactions under ultrasound irradiation. This phenomenon includes the formation, growth, and implosive collapse of bubbles in a liquid during a very short period of the time. Precipitate implosion of these bubbles in the liquids generates localized hot spots with very short lifetimes. The hot spot has an equivalent temperature of 5000 °C and pressure of about 2000 atmospheres.

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These transient, localized hot spots run high energy chemical transformations [22,23].

In continuation of ongoing to our work on the application of ultrasound in the organic reactions [24–26], we decided to carry out the ultrasonication reaction of primary amines, sodium azide and triethyl orthoformate catalyzed by ZnS NPs as a recyclable catalyst.

2. Experimental

2.1. Materials

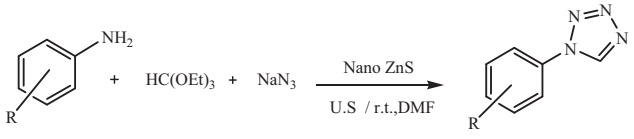
All commercially available reagents were used without further purification and purchased from the Merck Chemical Company in high purity. The used solvents were purified by standard procedure as followed; DMF: drying over $CaSo_4$ and distillation under reduced pressure, EtOAc: washing with aqueous 5% Na_2CO_3, then with saturated aqueous NaCl and drying with $MgSO_4$, MeOH: by fractional distillation, EtOH: drying over CaO and distillation under reduced pressure, and acetonitrile: drying over P_2O_5 and distillation.

2.2. Apparatus

IR spectra were recorded as KBr pellets on a Nicolet FT-IR spectrophotometer. ^1H and ^{13}C NMR spectra were recorded in $CDCl_3$ on a Bruker DRX-400 spectrometer with tetramethylsilane as internal reference. A BANDELIN ultrasonic HD 3200 with probe model KE 76, with the diameter of 6 mm, was used to produce ultrasonic irradiation and homogenizing the reaction mixture. Piezoelectric crystal of this kind of probe normally works in the range of 700 kHz, but by using some proper clamps, the output frequency of piezoelectric crystal have controlled and reduced to 20 kHz. Therefore, the induced frequency of probe to the reaction mixture is equal to 20 kHz. By changing the power of Tip the cavitations rate is displaced so that the Tip frequency under various amounts of power is constant. A thermal method was used for the calibration of ultrasonic power. XRD patterns were recorded by an X'PertPro (Philips) instrument with 1.54 Ångström wavelengths of X-ray beam and Cu anode material. Scanning electron microscope (SEM) of nanoparticles was performed on a FESEM Hitachi S4160. Transmission electron microscopy (TEM) was performed with a Jeol JEM 2100UHR, operated at 200 kV. Melting points obtained with a Yanagimoto micro melting point apparatus are uncorrected. The purity determination of the substrates and reaction monitoring were accomplished by TLC on silicagel polygram SILG/UV 254 plates (from Merck Company).

2.3. Synthesis of ZnS nanoparticles

ZnS nanoparticles were synthesized according to the previously reported method [27]. Zinc nitrate hexahydrate (0.01 mol) was dissolved in 20 ml of propylene glycol under intensive stirring at 90 °C. Then, 10 ml of thioacetamide solution (1 M) was previously prepared and added dropwise to the solution over 10 min. Afterward, the solution was exposed to irradiate at microwave oven followed a working cycle of 50 s. (on) and 100 s. (off) at 350 W. After completion of the reaction, the mixture was slowly cooled to room temperature. The precipitate was filtered and consecutively washed with deionized water (5 × 50 ml) and absolute ethanol (3 × 10 ml) and was dried at room temperature.



Scheme 1. Synthesis of 1-substituted 1*H*-1,2,3,4-tetrazoles by ultrasound irradiation and ZnS nanoparticles.

2.4. General procedure for synthesis of 1-substituted 1H-1,2,3,4-tetrazoles

A mixture of selected primary amine (1 mmol), triethyl orthoformate (1.2 mmol) and sodium azide (1 mmol) in the presence of 0.01 g ZnS nanoparticles was added to *N*,*N*-dimethylformamide as solvent and the reaction mixture was irradiated in ultrasonic apparatus with the power 50 W. The progress of the reaction was monitored by thin layer chromatography (TLC). After completion of the reaction, the mixture was diluted by 1:1 H_2O:ethylacetate (10 ml), stirred at ambient temperature (20 min) and centrifuged to separate the solid catalyst. The organic layer of the solution was separated, dried over sodium sulfate, and the organic solvent and other residues were stripped in a vacuum evaporator. The product was purified by recrystallization in a mixture of EtOAc:MeOH (3:1) to yield pure product. The obtained pure tetrazoles were characterized by spectroscopic data and melting points.

2.5. Spectral data for 1-substituted 1H-1,2,3,4-tetrazoles derivatives

2.5.1. 1-phenyl-1H-1,2,3,4-tetrazole

Yellow solid. m.p = 63–65 °C; IR (KBr)/ν (cm^{-1}): 3 051 (C–H, sp^2 stretch Ar), 1677 (C=N), 1588, 1488 (C=C); ^1H NMR (400 MHz, $CDCl_3$) δ ppm = 7.07–7.34 (m, 5H, *ArH*), 8.20 (s, 1H *tetrazole*).

2.5.2. 1-(4-Boromophenyl)-1H-1,2,3,4-tetrazole

White solid. m.p = 183–185 °C; IR (KBr)/ν (cm^{-1}): 3151 (C–H, sp^2 stretch, Ar), 1659 (C=N), 1576, 1481 (C=C); ^1H NMR (400 MHz, $CDCl_3$) δ ppm = 6.92–6.94 (d, 2H, *ArH*), 7.40–7.42 (d, 2H, *ArH*), 8.09 (s, 1H *tetrazole*); ^{13}C NMR (100 MHz, $CDCl_3$) δ ppm = 116.43, 120.76, 132.03, 143.99, 149.29.

2.5.3. 1-(4-Methylphenyl)-1H-1,2,3,4-tetrazole

Light yellow solid. m.p = 93–94 °C; IR (KBr)/ν (cm^{-1}): 3022 (C–H, sp^2 stretch, Ar), 2918 (C–H, sp^3 stretch), 1664 (C=N), 1607, 1506 (C=C); ^1H NMR (400 MHz, $CDCl_3$) δ ppm = 2.34 (s, 3H, *Me*), 6.94–6.96 (d, 2H, *ArH*), 7.11–7.13 (d, 2H, *ArH*), 8.17 (s, 1H *tetrazole*); ^{13}C NMR (100 MHz, $CDCl_3$) δ ppm = 20.79, 119.08, 129.63, 130.17, 142.95, 149.77.

2.5.4. 1-(3-Methylphenyl)-1H-1,2,3,4-tetrazole

White solid. m.p = 112–114 °C; IR (KBr)/ν (cm^{-1}): 3066 (C–H, sp^2 stretch, Ar), 2920 (C–H, sp^3 stretch), 1680 (C=N), 1598, 1483 (C=C); ^1H NMR (400 MHz, $CDCl_3$) δ ppm = 2.33 (s, 3H, *Me*), 6.86 (s, 1H, *ArH*), 6.89–6.91 (d, 2H, *ArH*), 7.18–7.22 (t, 1H, *ArH*), 8.21 (s, 1H *tetrazole*); ^{13}C NMR (100 MHz, $CDCl_3$) δ ppm = 21.43, 115.93, 119.97, 124.06, 129.19, 139.26, 145.28, 149.23.

2.5.5. 1-(2-Methylphenyl)-1H-1,2,3,4-tetrazole

White solid. m.p = 153–155 °C; IR (KBr)/ν (cm^{-1}): 3015 (C–H, sp^2 stretch, Ar), 2870 (C–H, sp^3 stretch), 1664 (C=N), 1488, 1590 (C=C); ^1H NMR (400 MHz, $CDCl_3$) δ ppm = 2.33 (s, 3H, *Me*), 7.02–7.03 (d, 1H, *ArH*), 7.05–7.07 (d, 1H, *ArH*), 7.18–7.22 (t, 2H, *ArH*), 8.08 (s, 1H *tetrazole*); ^{13}C NMR (100 MHz, $CDCl_3$) δ ppm = 17.94, 117.68, 123.43, 127, 128.71, 130.72, 144.10, 147.78.

2.5.6. 1-[4-(1H-tetrazol-1-yl)phenyl]ethanone

Yellow solid. m.p = 148–150 °C; IR (KBr)/ν (cm^{-1}): 3075 (C–H, sp^2 stretch, Ar), 2995 (C–H, sp^3 stretch), 1669 (C=N), 1499, 1585

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