SILICA GEL SUPPORTED γ-FERRITE AS A HETEROGENOUS CATALYST FOR HIGH YIELD SYNTHESIS OF β-LACTAMS (2-AZETIDINONES) UNDER ECO-FRIENDLY CONDITIONS VIA 1,2,4-TRIAZOLES

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A mild and efficient method has been reported for the preparation of Schiff base through the condensation reaction of various aromatic aldehydes with substituted aromatic amines in the presence of silica supported γ-Ferrite (ferric oxide) as a heterogeneous catalyst under solvent-free conditions. and this Schiff base used for Environmentally benign synthesis followed by the reaction with chloroacetylchloride in green conditions to yield the β-lactams (a-l) with excellent yields. The advantages of this eco-friendly, mild economic method are such as simplicity of the reaction procedure, moderate to high product yields, and simple working steps, very short reaction times.

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Introduction

A large number of 3-chloro monocyclic *β*-lactam possess powerful antibacterial, antimicrobial, anti-inflammatory, anticonvulsant, anti-tubercular and enzyme inhibition activity.[1-4] Synthesis of Schiff bases have been described in variant conditions using sulphuric acid[5]and glacial acetic acid.[6] 2-Azetidinone(*β*-lactam) ring system is the common structural feature of a number of broad spectrum *β*-lactam antibiotics, including penicillin, cephalosporin , carbapenems, nocardicins, monobactams, clavulanic acid, sub-lactams and azobactams, which have been widely used as chemotherapeutic agents to treat bacterial infections and microbial diseases.[7] The need for effective *β*-lactam has motivated synthetic organic and medicinal chemists to design new functionalized 2-azetidinones**.** The 1 H-1,2,4-triazole compounds possess important pharmacological activities such antifungal and antiviral activities. 1,2,4-triazole residues are bearing in such type of drugs for e.g fluconazole [8], the powerful azole antifungal agent as well as the potent antiviral N-nucleoside ribavirin[9] Furthermore, various 1,2,4-triazole derivatives have been reported as fungicidal[10], insecticidal[11], anti-microbial[12], and some showed antitumor activity[13], as well as anticonvulsants[14], antidepressants[15] and plant growth regulator anticoagulants[16]. Other laboratories reported the same biological activity of the triazole family[17-19].In connection with our work on 1,2,4-triazole having hydrazide and b-lactam moieties, we demonstrate here some potency of the reported hydrazide and b-lactams derivatives of 1,2,4 triazole. Green chemistry is the design of chemical products and processes that reduce or eliminate the way of use and generation of hazardous substances.[20] Advances in green chemistry address both obvious hazards and those associated with such global issues as climate change, energy production, availability of a safe and adequate water supply, food production, and the presence of toxic substances in the environment.[21] A number of classic organic reactions, traditionally run in organic solvents, can be carried out in water with the proper design of catalysts and reaction conditions.[22]

Microwave assisted heterocyclic synthesis is an efficient and Eco-friendly synthetic strategy for heterocyclic compounds and has now become a modern and powerful tool for green chemistry.[23-25] Microwave technology has recent application for the cyclization reactions of heterocyclic compounds. Nowadays such type of important reactions like nucleophilic substitution, ring formation reaction are done by these green technology, Microwave irradiation has been applied to organic reactions in the absence of solvent and/or in the presence of a solid support, such as clays, alumina and silica, resulting in shorter reaction times and better product yields than those obtained by using conventional heating.[26- 27] A solvent-free or solid state reaction may be carried out using the reactants alone or incorporating them in clays, zeolites , silica, alumina or other matrices [28] . In recent years, β-zeolite [29] , montomorillonite clays [30] , SiO2 supported γ-Fe2O3 were employed as catalysts for this purpose to obtain relatively better results.

SiO_2 is a particularly interesting oxide as it is widely used industrially as filler, adsorbent, drying agent, catalyst support and reagent. γ-Fe_2O_3 is the inorganic oxide most commonly utilized to carry out surface organic chemistry.[31] Although some of these methods represent a convenient procedure with good to high product yields, representation of a new, efficient and facile procedure for synthesis of these compounds can be very significant. In continuation of our research, because of these economical And environmental reasons, here we hope to report a procedure. Using SiO_2/γ-Fe_2O_3 as a catalyst for the synthesis of schiff bases From carbonyl compounds and different amines in the absence Of a solvent under mild reaction conditions.The structures of all the newly synthesized compounds were confirmed by elemental analysis, IR, ^{1}H NMR, ^{13}C NMR, and HR-MS.

Experimental

All melting points were determined in open capillaries. The IR spectra were recorder on shimadzu FT-IR 8300 Spectrophotometer. Samples were prepared by finely dispersing powder material on a KBr carrier. ^{1}H NMR and ^{13}C NMR spectra were measured on a Brucker DRX-300 were spectrometer in CDCl3 at 300 MHz using TMS as an internal standard. Chemical shifts reported on δ scales. The purity of the compounds was checked by thin layer chromatography (TLC) on silica gel (G-60 mesh) using chloroform: methanol as an eluent. Synthesized sample were purified by column chromatography using silica Gel (230-400 mesh). All other chemicals were purchased from Sigma-Aldrich and the reagent grade chemicals were purchased from commercial sources and further purified before use.

Reagents

All starting materials of commercial grade were purchased from Sigma–Aldrich and used without further purification. SiO_2 and Fe_2O_3 were purchased from Merck. The purity of all compounds was checked by TLC on glass plates coated with silica gel (E-Merck G254**).**

Preparation and determination of silica- supported γ-ferrite catalyst

The heterogenous silica supported catalyst was prepared in our laboratory by simple combination method. The mixture of silica and γ-ferrite in appropriate ratio was mixed together and this reagent was heated in an oven at 100 °C for 1 h and then the obtained homogeneous mixture, free flowing, white-red powder substance is sensitive towards moisture and thus should be stored in a sealed flask in a dedicator for later use**.** The exact loading of the γ-ferrite at the surface of silica was identified by IR spectra and prepared catalyst was then used for further catalytic study.

General procedure for the synthesis of Schiff base and β-lactams (2-Azetidinones) by using catalyst.

[a] Microwave method for the synthesis of (chloroacetyl)-3-mercapto-4-methyl-1,2,4-triazole (1)

An equimolar mixture of microwave method a solid supported mixture of compounds 3-mercapto-4-methyl-1,2,4 Triazole (0.01) and chloroacetylchloride (0.01) in ethanol was mixed thoroughly in open glass vessel and subjected to the microwave irradiation at low power setting (25 %, 200 W) for 2.40–4.15 min, then allowed to cool, The reaction progress was monitored by TLC using Chloroform: methanol solvents, when reaction was completed the reaction mixture was cooled and poured into crushed ice. The product obtained was filtered, dried, wash with water and recrystallized from ethanol to get compound **1.** The compound **1** was identified by spectrophotometric analysis. Molecular formula; $C_5H_6ClN_3OS$, Yield 80 %, m.p. 108-109 °C**,** brown crystals. The structure was established on the basis of spectral analysis, IR (KBr), υ_{max} 1688, 1553, 1468, 1338 (1,2,4-triazole nucleus), 2592 (Ar-SH), 2825, 1476, 1210 (N-CH_3), 718 (S-CO-S) 1615 (S-CO) cm^{-1}; ^{1}H NMR ($CDCl_3$) δppm : 8.42 (s, H, Ar-H of 1,2,4 triazole), 4.48 (s, 2H , CH_2), 3.60 (s, 3H, N-CH3). ^{13}C NMR ($CDCl_3$, 100 MHz). Four characteristic signals appeared for (N-CH_3), (aliphatic CH_2), (C of 1,2,4-triazole), (CO aliphatic) in the δ (ppm) ranges 27.1, 46.1-47.1, 160.4-162.5, 187.2-188.6 ppm, respectively. All these fact collectively suggest the successful synthesis of compound **1.**

[b] Microwave method for the synthesis of (hydrazinoacetyl)-3-mercapto-4-methyl-1,2,4-triazole (2)

A mixture of compound 1 (0.01 mol) and hydrazine hydrate (0.01 mole) in ethanol was performed in open glass vessel and subjected to the microwave irradiation at low power setting (25 %, 200 W) for 2-3 min, After cooling The product was filtered and purified over a column chromatography using chloroform: methanol (7:3 v/v). The purified product was recrystallized from ethanol at room temperature to yield compound **2.** The compound **2** was identified by spectrophotometric analysis. Molecular formula; $C_5H_9N_5OS$, Yield 81 %, m.p. 136-139 °C, pale brown crystals, The structure was established on the basis of spectral analysis, IR (KBr) υ_{max} 1688, 1553, 1468, 1338 (1,2,4-triazole nucleus), 2825,1476,1210 (N-CH_3), 1668 (CO-NH), 3351 (NH_2) cm^{-1}; ^{1}H NMR ($CDCl_3$) δ (ppm) : 8.42 (s, H, Ar-H of 1,2,4-triazole), 3.73(s, 2H, CH2), 3.62 (s, 3H, N-CH_3), 2.1 (s, 2H, NH_2),2.0 (s, 1H, NH). ^{13}C NMR ($CDCl_3$, 100 MHz) four characteristic signals appeared for (N-CH_3), (aliphatic CH_2), (C of 1,2,4 triazole), (CO aliphatic) in the δ (ppm) ranges 27.1, 66.1, 161.4-162.3, 193.2-196.6 ppm, respectively. All these fact collectively suggest the successful synthesis of compound **2.**

[c] Microwave method for the synthesis of [(2-(aryl)hydrazine-acetyl]-3-mercapto-4-methyl-1,2,4-triazoles (3)

A mixture of the compound **2** (0.01 mol) and arylaldehyde (0.01 mol) and SiO_2/γ-Fe_2O_3 in ethanol was performed in open glass vessel for 1 minute on microwave irradiation. The progress of reaction was monitored by TLC using hexane and ethyl acetate (9:1v/v) as eluent. was SiO_2/γ-Fe_2O_3 removed by filtration and the eluate was concentrated to give a product which was recrystallized from chloroform, to give compound **3a-3l** in excellent yield,. The compounds **3a-3l** was identified by spectrophotometric analysis.

[(2-(4-Chlorobenzylidene) hydrazino acetyl] -3-mercapto-4-methyl-1,2,4-triazole (3a)

Molecular formula; $C_{12}H_{12}ClN_5OS$, Yield 94 %, m.p. 152-55 °C, light yellow crystals, The structure was established on the basis of spectral analysis, IR (KBr) υ_{max} 1678, 1555, 1468, 1332 (1,2,4-triazole nucleus), 2822, 1470, 1215 (N-CH_3), ^{1}H NMR ($CDCl_3$) δ (ppm) : 8.44 (s, H, Ar-H of 1,2,4 triazole), 3.74 (s, 2H, CH_2), 3.59 (s, 3H, N-CH_3), 8.52 (s, 1H of azomethine), 2.0 (s, 1H, NH). ^{13}C NMR ($CDCl_3$, 100 MHz)/four characteristic signals appeared for (N-CH_3), (aliphatic CH_2), (C of 1,2,4-triazole), (CO aliphatic), (C of aromatic aldehyde) in the δ (ppm) ranges 27.1, 63.1, 143.4-162.3, 196.6, and 128.9-131.6 ppm respectively. All these fact collectively suggest the successful synthesis of compound **3a.**

[(2-(2,4-Dichlorobenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3b)

$C_{12}H_{11}Cl_2N_5OS$ Yield 91 %, m.p. 135-140 °C, light yellow crystals, IR(KBr) υ_{max} 1678, 1468, For 1,2,4-triazole nucleus, 1214 (N-CH_3), 755 (C-Cl), 1608 (N=CH); ^{1}H NMR ($CDCl_3$) δ (ppm): 8.44 (s, H, Ar-H of 1,2,4 triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine), 2.0 (s, 1H, NH), 7.40, 7.70, 7.98 for benzylidemine; ^{13}C NMR ($CDCl_3$) 27.1 (N-CH_3), 63.1 (aliphatic CH_2), 143.1-161.8 (C of 1,2,4-triazole), 196.5(CO aliphatic) 128.3-132.6 (C of aromatic aldehyde); Mass (HRMS): m/z : 344, M^+ **.**

[(2-(2-Bromobenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3c)

$C_{12}H_{12}BrN_5OS$; Yield: 88 %, m.p, 145-147 °C, light red powder, IR(KBr) υ_{max} 1677, 1452 for 1,2,4-triazole nucleus, 635 (C-Br), 1602 (N=CH), 1212 (N-CH_3); ^{1}H NMR ($CDCl_3$) δ (ppm): 8.42 (s, H, Ar-H of 1,2,4 triazole), 3.61 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine) 2.0 (s, 1H, NH) 7.41-7.72,7.58 for benzylidemine; ^{13}C NMR ($CDCl_3$): 27.1 (N-CH_3), 63.7 (aliphatic CH_2), 143.1-162.8 (C of 1,2,4-triazole), 196.5 (CO aliphatic) 121.1-132.8 (C of aromatic aldehyde), 143.3 (C of azomethine); Mass (HRMS): m/z : 354, M^+.

[(2-(3-Bromobenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3d)

$C_{12}H_{12}BrN_5OS$, Yield 90 %, m.p, 142-144 °C, red powder, IR(KBr) υ_{max} 1674, 1468, (1,2,4-triazole nucleus), 1215 (N-CH_3), 635 (C-Br), 1602 (N=CH); ^{1}H NMR ($CDCl_3$) δ (ppm) 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60(s, 3H, N-CH_3), 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine), 2.0 (s, 1H, NH) 7.41, 7.82, 7.58 for benzylidemine; ^{13}C NMR ($CDCl_3$) 27.1 (N-CH_3), 63.7(aliphatic CH_2), 143.1-162.5 (C of 1,2,4-triazole), 196.5 (CO aliphatic) 123.1-133.8 (C of aromatic aldehyde) 143.3 (C of azomethine); Mass (HRMS): m/z: 354, M^+.

[(2-(4-Bromobenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3e)

$C_{12}H_{12}BrN_5OS$. Yield 82 %, m.p, 145-146 °C, yellow powder, IR(KBr), υ_{max} 1674, 1468, (1,2,4-triazole nucleus), 1215 (N-CH_3) 638 (C-Br), 1609 (N=CH); ^{1}H NMR ($CDCl_3$) δ(ppm) 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3) 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine), 2.0 (s, 1H, NH) 7.70-7.58 for benzylidemine; ^{13}C NMR ($CDCl_3$) 27.1 (N-CH_3), 63.7(aliphatic CH_2),143.1-162.5 (C of 1,2, 4-triazole), 196.5 (CO aliphatic) 125.1-132.7 (C of aromatic aldehyde), 143.3 (C, of azomethine); Mass (HRMS): m/z: 354, M^+.

[(2-(2-Methoxybenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3f)

$C_{13}H_{15}N_5O_2S$. Yield 84 %, m.p, 150-152 °C, amorphous yellow powder, IR(KBr), υ_{max} 1672, 1462, (1,2,4-triazole nucleus), 1215 (N-CH3), 2865 and 1170 (OCH_3) 1590 (N=CH). ^{1}H NMR ($CDCl_3$) δ (ppm) 8.42 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine), 2.0 (s, 1H, NH), 7.72, 7.26 for benzylidemine, 3.82 for 3H of OCH_3. ^{13}C NMR ($CDCl_3$) 27.1 (N-CH_3), 63.8 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole), 196.5 (CO aliphatic), 111.2-157.7 (C of aromatic aldehyde), 143.3 (C, of azomethine), 55.6 (C of OCH_3). Mass (HRMS): m/z: 305, M^+.

[(2-(3-Methoxybenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3g)

$C_{13}H_{15}N_5O_2S$ Yield 94 %, m.p, 151-152 °C, yellow powder, IR(KBr), υ_{max} 1672, 1462, (1,2,4-triazole nucleus), 1215 (N-CH_3), 2860, 1165 (OCH_3), 1598 (N=CH). ^{1}H NMR ($CDCl_3$) δ(ppm) 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine), 2.0 (s, 1H, NH), 7.52, 7.06 for benzylidenemine, 3.83 for 3H of OCH_3. ^{13}C NMR ($CDCl_3$): 27.1 (N-CH_3), 63.8 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole), 196.5 (CO aliphatic), 111.2-160.7 (C of aromatic aldehyde), 143.3 (C, of azomethine), 55.8 (C of OCH_3). Mass (HRMS): m/z: 305, M^+

[(2-(4-Methoxybenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3h)

$C_{13}H_{15}N_5O_2S$. Yield 96 %, m.p, 154-156 °C, dark yellow powder, IR(KBr), υ_{max} 1672, 1462, (1,2,4-triazole nucleus), 1215 (N-CH_3), 2866, 1162(OCH_3) 1598 (N=CH). ^{1}H NMR ($CDCl_3$) δ (ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine), 2.0 (s, 1H, NH), 7.82, 7.06 for benzylidemine, 3.83 for 3H of OCH_3. ^{13}C NMR ($CDCl_3$): 27.1 (N-CH_3), 63.8 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole) 196.5 (CO aliphatic) 114.2-162.7 (C of aromatic aldehyde), 143.3 (C of azomethine), 55.8 (C of OCH_3). Mass (HRMS): m/z: 305, M^+.

[(2-(3,4,5-Trimethoxybenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3i)

$C_{15}H_{19}N_5O_4S$. Yield 88 %, m.p. 156-158 °C, yellow powder, IR(KBr), υ_{max}: 1672, 1460 (1,2,4-triazole nucleus), 1215 (N-CH_3), 2865 and 1170 (OCH_3), 1598 (N=CH). ^{1}H NMR ($CDCl_3$) δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2) 8.54 (s, 1H of azomethine), 2.0 (s, 1H, NH), 7.14 for benzylidemine, 3.83 for 6H of OCH_3. ^{13}C NMR ($CDCl_3$) 27.1 (N-CH_3), 63.8 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole), 196.5 (CO aliphatic), 104.2-152.7 (C of aromatic aldehyde), 143.3 (C of azomethine), 56.1, 60.4 (C of OCH_3). Mass (HRMS): m/z: 365, M^+.

[(2-(2-Methylbenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3j)

$C_{13}H_{15}N_5OS$. Yield 93 %, m.p, 158-160 °C, red powder, IR(KBr), υ_{max}: 1672, 1460, (1,2,4-triazole nucleus), 1215 (N-CH_3), 2927 (CH_3), 1549 (N=CH). ^{1}H NMR ($CDCl_3$) δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2), 8.52 (s, 1H of azomethine), 2.0 (s,

1H, NH), 7.24, 7.70 for benzylidemine, 2.48 for 3H of CH_3. ^{13}C NMR ($CDCl_3$): 27.1 (N-CH_3), 63.8 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole), 196.5 (CO aliphatic), 124.2-135.3 (C of aromatic aldehyde), 143.3 (C of azomethine), 16.8 (C of CH_3). Mass (HRMS), m/z: 289, M^+.

[(2-(3-Methylbenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3k)

$C_{13}H_{15}N_5OS$. Yield 88 %, m.p, 159-161 °C, light red powder, IR(KBr): υ_{max} 1672, 1460, (1,2,4-triazole nucleus), 1215 (N-CH_3), 2927 (CH_3), 1549 (N=CH); ^1H NMR ($CDCl_3$) δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2), 8.52 (s, 1H of azomethine), 2.0 (s, 1H, NH), 7.24, 7.70 for benzylidemine, 2.48 for 3H of CH_3. ^{13}C NMR ($CDCl_3$): 27.1 (N-CH_3), 63.8 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole), 196.5 (CO aliphatic), 124.2-135.3 (C of aromatic aldehyde), 143.3 (C, of azomethine), 16.8 (C of CH_3). HRMS: m/z: 289, M^+.

[(2-(4-Methylbenzylidene)hydrazinoacetyl]-3-mercapto-4-methyl-1,2,4-triazole (3l)

$C_{13}H_{15}N_5OS$. Yield 89 %, m.p, 160-161 °C, light yellow powder, IR(KBr): υ_{max} 1672, 1460, (1,2,4-triazole nucleus), 1215 (N-CH_3), 2928 (CH_3), 1547 (N=CH). ^1H NMR ($CDCl_3$) δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.60 (s, 3H, N-CH_3), 3.76 (s, 2H, CH_2), 8.52 (s, 1H of azomethine), 2.0 (s, 1H, NH), 7.27, 7.70 for benzylideneimine, 2.38 for 3H of CH_3. ^{13}C NMR ($CDCl_3$): 27.1 (N-CH_3), 63.8 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole), 196.5 (CO aliphatic), 126.2-140.3 (C of aromatic aldehyde), 143.3 (C of azomethine), 21.8 (C of CH_3). HRMS: m/z: 289, M^+.

General microwave method for the synthesis of [{4-[4-chlorophenyl]-3-chloro-2-oxoazetidineimino}-acetyl-3-mercapto-4-methyl-1,2,4-triazole (4a)

An equimolar mixture of compound **3a** (0.01) and triethylamine (0.01) in ethanol then added chloroacetyl chloride dropwise in ice bath then reaction mixture was performed in open glass vessel for about 2 min, the progress of reaction was monitored by TLC using hexane and ethyl acetate (9:1 v/v) as eluent, and the reaction mixture was concentrated to give a product which was recrystallized from chloroform, to give compounds **4a-4l** were synthesized in the similar manner using compound **3a-3l**. The synthesized compounds were identified by spectrophotometric analysis.

Synthesis of N-[{4-(4-chlorophenyl)-3-chloro-2-oxo-azetidine-imino}acetyl-3-mercapto-4-methyl-1,2,4-triazole, (4a)

Yield 87 %, m.p. 210-215 °C, light red crystals. The structure was established on the basis of spectral analysis, IR(KBr): υ_{max} 1668, 1545, 1332 (1,2,4-triazole nucleus), 2826, 1469, 1213 (N-CH_3). ^1H NMR ($CDCl_3$): δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.74 (s, 2H, CH_2), 3.58 (s, 3H, N-CH_3), 5.49 (s, 1H of CH-Cl), 2.0 (s, 1H, NH). ^{13}C NMR ($CDCl_3$) four characteristic signals appeared for (N-CH_3), (aliphatic CH_2), (C of 1,2,4-triazole), (CO aliphatic), (C of aromatic aldehyde), (CO of β-lactam ring), (C of β-lactam ring) in the δ(ppm) ranges 26.1, 62.1, 163.4-142.3, 196.6, 128.9-132.6, 197.2, 76.2 ppm. Mass (HRMS): m/z: 385, M^+. Anal.: Calcd for molecular formula $C_{14}H_{13}Cl_2N_5O_2S$: C 43.53; H 3.49; Cl 18.36; N 18.13; found C 43.50, H 3.45, Cl 18.30; N 18.10.

Synthesis of N-[{4-(2,4-dichlorophenyl]-3-chloro-2-oxo-azeti-dineimino}-acetyl-3-mercapto-4-methyl-1,2,4-triazole (4b)

Yield 85 %, m.p. 215-220 °C, light yellow crystals, IR(KBr): υ_{max} 1664, 1545, 1332 (1,2,4-triazole nucleus), 2826, 1466, 1214 (N-CH_3). ^1H NMR ($CDCl_3$) δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.72 (s, 2H, CH_2), 3.60 (s, 3H, N-CH_3), 5.42 (s, 1H of CH-Cl), 2.0 (s, 1H, NH). 5.08 (d, 1H, N–CH–Ar), 7.32-7.64 (m, 6H, Ar). ^{13}C NMR ($CDCl_3$) 27.1 (N-CH_3), 62.1 (aliphatic CH_2), 143.1-161.6 (C of 1,2,4-triazole), 196.5 (CO aliphatic), 125.3-142.0 (C of aromatic aldehyde), 163.2 (CO of β-lactam ring), 64.3 (C of β-lactam ring). Mass (HRMS): m/z: 420, M^+. Anal.: Calcd for $C_{14}H_{12}Cl_3N_5O_2S$: C 39.97; H 2.88; Cl 25.28; N 16.65; O 7.62; S 7.61; Found C 39.96; H 2.83; Cl 25.23; N 16.62; O 7.52; S 7.61.

Synthesis of N-[{4-(2-bromophenyl]-3-chloro-2-oxo-azetidine-imino}-acetyl-3-mercapto-4-methyl-1,2,4-triazole (4c)

Yield 88 %, m.p. 235-238 °C, pale yellow crystals, IR(KBr): υ_{max} 1664, 1542 (1,2,4- triazole nucleus), 635 (C-Br), 2826, 1214 (N-CH_3), 1762 (C=O), 755 (C-Cl). ^1H NMR ($CDCl_3$) δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.75 (s, 2H, CH_2), 3.60 (s, 3H, N-CH_3), 5.47 (s, 1H of CH-Cl), 2.0 (s, 1H, NH), 5.04 (d, 1H, N–CH–Ar), 7.16-7.54 (m, 8H, Ar). ^{13}C NMR ($CDCl_3$), 27.1 (N-CH_3), 62.0 (aliphatic CH_2), 143.1-163.6 (C of 1,2,4-triazole), 196.4 (CO aliphatic), 121.3-142.0 (C of aromatic aldehyde), 163.4 (CO of β-lactam ring), 63.6 (C of β-lactam ring). Mass (HRMS): m/z: 430, M^+. Anal.: Calcd for $C_{14}H_{13}BrClN_5O_2S$: C 39.07; H 3.08; Br 18.58; Cl 8.28; N 16.25; O 7.42; S 7.44. Found: C 39.06; H 3.07; Br 18.20; Cl 8.23; N 16.22; O 7.41; S 7.41.

Synthesis of N-[{4-(3-bromophenyl]-3-chloro-2-oxo-azetidi-neimino}-acetyl-3-mercapto-4-methyl-1,2,4 triazole (4d)

Yield 87 %, m.p.232-238 °C, dark yellow crystals. IR(KBr) υ_{max} 1662, 1542, (1,2,4-triazole nucleus), 637 (C-Br) 2823, 1214 (N-CH_3), 1760 (C=O), 763 (C-Cl). ^1H NMR ($CDCl_3$) δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.75 (s, 2H, CH_2), 3.60 (s, 3H, N-CH_3), 5.44 (s, 1H of CH-Cl), 2.0 (s, 1H, NH), 5.08 (d, 1H, N–CH–Ar), 7.26-7.44 (m, 8H, Ar). ^{13}C NMR ($CDCl_3$): 27.1 (N-CH_3), 62.0 (aliphatic CH_2), 143.1-162.6 (C of 1,2,4-triazole), 196.4 (CO aliphatic) 122.5-145.0 (C of aromatic aldehyde), 163.4(CO of β-lactam ring), 64.6 (C of β-lactam ring). Mass (HRMS): m/z: 430, M^+. Anal.: Calcd for $C_{14}H_{13}BrClN_5O_2S$: C 39.04; H 3.04; Br 18.55; Cl 8.23; N 16.26; O 7.43; S 7.42. Found C 39.01; H 3.02; Br 18.50; Cl 8.25; N 16.24; O 7.40; S 7.40.

Synthesis of N-[{4-(4-bromophenyl]-3-chloro-2-oxo-azetidine-imino}acetyl-3-mercapto-4-methyl-1,2,4-triazole (4e)

Yield 77 %, m.p. 234-239 °C, dark yellow crystals. IR(KBr): υ_{max} 1662, 1546 (1,2,4-triazole nucleus), 638 (C-Br), 2825, 1214 (N-CH_3), 1768 (C=O), 769 (C-Cl). ^1H NMR

($CDCl_3$): δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.72 (s, 2H, CH_2), 3.62 (s, 3H, N-CH_3), 5.40 (s, 1H of CH-Cl), 2.2 (s, 1H, NH), 5.02 (d, 1H, N–CH–Ar), 7.16-7.94 (m, 8H, Ar). ^{13}C NMR ($CDCl_3$): 27.0 (N-CH_3), 61.0 (aliphatic CH_2), 143.8-160.6 (C of 1,2,4-triazole), 196.4 (CO aliphatic) 122.4-145.9 (C of aromatic aldehyde), 163.2 (CO of β-lactam ring), 64.8 (C of β-lactam ring). Mass (HRMS): m/z: 430, M^+. Anal.: Calcd for $C_{14}H_{13}BrClN_5O_2S$: C 39.04; H 3.04; Br 18.55; Cl 8.23; N 16.26; O 7.43; S 7.42. Found: C 39.00, H 3.01; Br 18.59; Cl 8.20; N 16.27; O 7.41; S 7.45.

Synthesis of N-[{4-(2-methoxyphenyl]-3-chloro-2-oxo-azetidineimino}acetyl-3-mercapto-4-methyl-1,2,4-triazole (4f)

Yield 82 %, m.p. 224-229 °C, yellow crystals. IR(KBr): υ_{max} 1672, 1460 (1,2,4-triazole nucleus), 1215 (N-CH_3), 2868 and 1173 (OCH_3), 1763 (C=O), 766 (C-Cl). ^1H NMR ($CDCl_3$): δ(ppm): 8.44 (s, H, Ar-H of 1,2,4-triazole), 3.75 (s, 2H, CH_2), 3.62 (s, 3H, N-CH_3), 5.45 (s, 1H of CH-Cl), 2.4 (s, 1H, NH). 5.05 (d, 1H, N–CH–Ar), 6.86-7.14 (m, 8H, Ar), 3.84 (s, 3H, OCH_3). ^{13}C NMR ($CDCl_3$), 27.2 (N-CH_3), 63.0 (aliphatic CH_2), 142.8-161.6 (C of 1,2,4-triazole), 196.4 (CO aliphatic), 112.4-155.9 (C of aromatic ring), 163.5 (CO of β-lactam ring), 64.2 (C of β-lactam ring). Mass (HRMS): m/z: 381, M^+. Anal.: Calcd for $C_{15}H_{16}ClN_5O_3S$: C 47.18; H 4.22; Cl 9.27; N 18.36; O 12.53; S 8.42. Found C 49.10; H 4.21; Cl 9.20; N 18.37; O 11.41; S 8.48.

Synthesis of N-[{4-(3-methoxyphenyl]-3-chloro-2-oxoazetidineimino}acetyl-3-mercapto-4-methyl-1,2,4-triazole (4g)

Yield 85 %, m.p. 234-237°C, deep yellow powder. IR (KBr): υ_{max} 1672, 1458 (1,2,4-triazole nucleus), 1215 (N-CH_3), 2868 and 1175 (OCH_3), 1768 (C=O), 762 (C-Cl). ^1H NMR ($CDCl_3$): δ(ppm): 8.42 (s, H, Ar-H of 1,2,4-triazole), 3.73 (s, 2H, CH_2), 3.61 (s, 3H, N-CH_3), 5.42 (s, 1H of CH-Cl), 2.6 (s, 1H, NH), 5.07 (d, 1H, N–CH–Ar), 6.83-7.24 (m, 8H, Ar), 3.82 (s, 3H, OCH_3). ^{13}C NMR ($CDCl_3$,): 27.4 (N-CH_3), 62.2 (aliphatic CH_2), 162.8-141.4 (C of 1,2,4-triazole), 196.2 (CO aliphatic), 112.5-165.9 (C of aromatic ring), 162.5 (CO of β-lactam ring), 64.2 (C of β-lactam ring), 53.8 (C of OCH_3). Mass (HRMS):m/z: 381, M^+. Anal.: Calcd for $C_{15}H_{16}ClN_5O_3S$: C 47.18; H 4.22; Cl 9.27; N 18.36; O 12.56; S 8.40. Found: C 48.10; H 4.23; Cl 9.28; N 18.38; O 12.41; S 8.42.

Synthesis of N-[{4-(4-methoxyphenyl]-3-chloro-2-oxo-azetidineimino}acetyl-3-mercapto-4-methyl-1,2,4-triazole (4h)

Yield 87%, m.p. 237-239 °C, deep brown crystals. IR(KBr): υ_{max} 1672, 1468, (1,2,4-triazole nucleus), 1218 (N-CH_3), 2867 and 1165 (OCH_3), 1769 (C=O), 762 (C-Cl). ^1H NMR ($CDCl_3$): δ(ppm): 8.42 (s, H, Ar-H of 1,2,4-triazole), 3.72 (s, 2H, CH_2), 3.63 (s, 3H, N-CH_3), 5.43 (s, 1H of CH-Cl), 2.2 (s, 1H, NH), 5.10 (d, 1H, N–CH–Ar), 6.93-7.14 (m, 8H, Ar), 3.84 (s, 3H, OCH_3). ^{13}C NMR ($CDCl_3$): 27.2 (N-CH_3), 62.1 (aliphatic CH_2), 162.2-142.4 (C of 1,2,4-triazole), 196.1 (CO aliphatic), 114.5-155.9 (C of aromatic ring), 164.5 (CO of β-lactam ring), 64.1 (C of β-lactam ring), 54.8 (C of OCH_3). Mass (HRMS): m/z: 381, M^+. Anal.: Calcd for $C_{15}H_{16}ClN_5O_3S$: C 47.18; H 4.22; Cl 9.28; N 18.34; O 12.58; S 8.41. Found C 47.12; H 4.21; Cl 9.26; N 18.32; O 12.51; S 8.40.

Synthesis of N-[{4-(3,4,5-trimethoxyphenyl]-3-chloro-2-oxo-azetidineimino}acetyl-3-mercapto-4-methyl-1,2,4-triazole (4i)

Yield 75 %, m.p. 278-279 °C, shiny yellow powder. IR (KBr): υ_{max} 1672, 1461 (1,2,4-triazole nucleus), 1216 (N-CH_3), 2862 and 1175 (OCH_3), 1479 (C=C aromatic), 1765 (C=O), 764 (C-Cl). ^1H NMR ($CDCl_3$): δ(ppm): 8.43 (s, H, Ar-H), 2.0(s, 1H, NH), 5.07 (d, 1H, N–CH–Ar), 6.53-7.8 (m, 4H, Ar), 3.82 (s, 9H, OCH_3). ^{13}C NMR ($CDCl_3$), 27.0 (N-CH_3), 62.5 (aliphatic CH_2), 162.5-142.0 (C of 1,2,4 triazole), 196.7 (CO aliphatic), 104.5-152.9 (C of aromatic ring), 163.5 (CO of β-lactam ring), 64.2 (C of β lactam ring), 57.1 (C of OCH_3). Mass (HRMS): m/z: 441, M^+. Anal.: Calcd for $C_{17}H_{20}ClN_5O_5S$: C 46.21; H 4.54; Cl 8.08; N 15.84; O 18.10; S 7.25. Found: C 46.18; H 4.51; Cl 8.06; N 15.82; O 18.11; S 7.24.

Synthesis of N-[{4-(2-methyl-phenyl]-3-chloro-2-oxo-azetidineimino}acetyl-3-mercapto-4-methyl-1,2,4 triazole (4j)

Yield 92 %, m.p. 232-234 °C, yellow powder. IR(KBr): υ_{max} 1676, 1460 (1,2,4-triazole nucleus), 1214 (N-CH_3), 1472 (C=C aromatic), 1742 (C=O), 762 (C-Cl). ^1H NMR ($CDCl_3$) δ(ppm): 8.39 (s, H, Ar-H of 1,2,4-triazole), 3.68 (s, 2H, CH_2), 3.61 (s, 3H, N-CH_3), 5.45 (s, 1H of CH-Cl), 2.1 (s, 1H, NH), 5.12 (d, 1H, N–CH–Ar), 6.93-7.51 (m, 8H, Ar), 2.62 (s, 3H, OCH_3). ^{13}C NMR ($CDCl_3$): 26.8 (N-CH_3), 62.3 (aliphatic CH_2), 162.7-141.1 (C of 1,2,4 triazole), 195.7 (CO aliphatic), 124.5-142.9 (C of aromatic ring), 162.5 (CO of β-lactam ring), 64.8 (C of β-lactam ring), 17.1 (C of CH_3). Mass (HRMS): m/z: 365, M^+. Anal.: Calcd for $C_{15}H_{16}ClN_5O_2S$: C 49.24; H 4.44; Cl 9.68; N 19.24; O 8.70; S 8.75. Found: C 47.18; H 4.41; Cl 9.66; N 18.92; O 8.01; S 7.94.

Synthesis of N-[{4-(3-methylphenyl]-3-chloro-2-oxo-azetidineimino}acetyl-3-mercapto-4-methyl-1,2,4 triazole (4k)

Yield 92 %, m.p. 230-232 °C, light yellow powder. IR(KBr): υ_{max} 1672, 1460 (1,2,4-triazole nucleus), 1215 (N-CH_3), 2929 (CH_3), 1472 (C=C aromatic), 1740 (C=O), 760 (C-Cl). ^1H NMR ($CDCl_3$) δ(ppm): 8.49 (s, H, Ar-H of 1,2,4-triazole), 3.78 (s, 2H, CH_2), 3.62 (s, 3H, N-CH_3), 5.43 (s, 1H of CH-Cl), 2.3 (s, 1H, NH), 5.02 (d, 1H, N–CH–Ar), 7.13-7.52 (m, 8H, Ar), 2.31 (s, 3H, CH_3). ^{13}C NMR ($CDCl_3$): 27.8 (N-CH_3), 62.1 (aliphatic CH_2), 161.7-142.1 (C of 1,2,4-triazole), 197.7 (CO aliphatic), 122.5-144.9 (C of aromatic ring), 161.5 (CO of β-lactam ring), 64.7 (C of β-lactam ring), 20.1 (C of CH_3). Mass (HRMS): m/z: 365, M^+**.** Anal.: Calcd for $C_{15}H_{16}ClN_5O_2S$: C 49.25; H 4.41; Cl 9.69; N 19.14; O 8.74; S 8.76. Found: C 48.28; H 4.31; Cl 9.62; N 19.12; O 8.71; S 8.74.

Synthesis of N-[{4-(4-methyl-phenyl]-3-chloro-2-oxo-azetidineimino}acetyl-3-mercapto-4-methyl-1,2,4 triazole (4l)

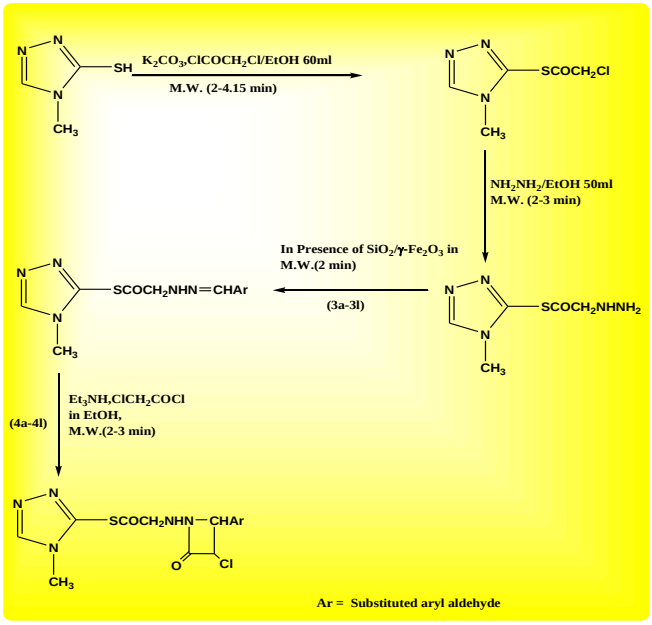
Yield 91 %, m.p. 235-235 °C, light red powder, IR (KBr): υ_{max} 1676, 1461 (1,2,4 triazole nucleus), 1217(N-CH_3), 2928 (CH_3), 1474 (C=C aromatic), 1742 (C=O), 765 (C-Cl). ^1H NMR ($CDCl_3$): δ(ppm): 8.36 (s, H, Ar-H of 1,2,4-triazole), 3.68 (s, 2H, CH_2), 3.51 (s, 3H, N-CH_3), 5.40 (s, 1H of CH-Cl), 2.0(s, 1H, NH), 5.02(d, 1H, N–CH–Ar), 7.13-7.32 (m, 8H, Ar), 2.30 (s, 3H, CH_3). ^{13}C NMR ($CDCl_3$): 27.0 (N-

CH_3), 62.3 (aliphatic CH_2), 162.7-141.1 (C of 1,2,4-triazole), 195.7 (CO aliphatic), 124.5-144.0 (C of aromatic ring), 164.5 (CO of β-lactam ring), 63.9 (C of β-lactam ring), 18.1 (C of CH_3). Mass (HRMS): m/z: 365, M^+**.** Anal.: Calcd for $C_{15}H_{16}ClN_5O_2S$: C 49.25; H 4.41; Cl 9.69; N 19.14; O 8.74; S 8.75. Found C 47.28; H 4.39; Cl 9.52; N 19.08; O 8.69; S 8.71.

Results and Discussion

Recently, the interest in the sulfur containing moieties has been significantly surged since a wide range of biological activities associated with the scaffold have been identified. The present two pot procedure is efficient, general and versatile. Further application of the methodology to synthesize heterocyclic is in progress in our laboratory.

3-Mercapto-4-methyl-1,2,4-triazole on reaction with chloroacetylcholoride yielded (chloroacetyl)-3-mercapto-4-methyl-1,2,4-triazole (**1**). The formation of compound **1** was confirmed by the appearance of a signal at 8.42 ppm (s, H, Ar-H of 1,2,4-triazole), 4.48 (s, 2H, CH_2) , and 3.60 (s, 3H, N-CH_3) in ^{1}H NMR spectra and IR spectral band due to 718 (S-CO-S) 1615 (S-CO) cm^{-1}, respectively, also confirmed the formation of compound **1,** and on amination with hydrazine hydrate, yielded of (hydrazineacetyl)-3-mercapto-4-methyl-1,2,4 triazole compound **2.** In the ^{1}H NMR spectra of **2** a singlet peak at 2.1 ppm was observed due to 2H of NH_2 and a singlet peak at 2.0 ppm was due to (s, 1H) NH. Furthermore, in the IR spectra, the bands at 1668 cm^{-1} (C=O of amide) and 3351 cm^{-1} (NH_2) also confirmed the formation of compound **2**. The compound **2** on condensation with various aromatic aldehydes yielded α-(arylidene-hydrazino)acetyl]-3-mercapto-4-methyl-1,2,4-triazoles (**3a-l**), the formation of (**3a-l**) was supported by the appearance of singlet peaks at 8.52 ppm due to (1H) azomethine group. The ^{1}H NMR spectra and IR spectra (the bands at 1635, 1647 cm^{-1} for N=CH, acyclic) also confirmed the formation of **3a-3l** compounds.



Scheme 1. Reaction routes

The compounds **(3a-l)** on reaction with triethylamine in presence of chloroacetylchloride underwent cyclization process afforded [{4-[4-aryl]-3-chloro-2-oxoazetidine-imino}acetyl-3-mercapto-4-methyl-1,2,4-triazoles **(4a-l)**. In ^{1}H NMR spectra peaks at 4.69 (s, 1H of C-CH-Cl) 2.52 for (s, 1H, N-CH-Ar) groups have confirmed the formation of 2-azetidinones. In the IR spectra of **(4a-l)** the bands at 1710, 1721 cm^{-1} for C=O (cyclic) of azitidinone ring also confirmed the formation of azitidinones **(Scheme 1).** All the Schiff base prepared in presence of heterogenous catalysts and all cyclization reactions proceeds under microwave irradiation were completed within 1-4 min.The impact of microwave irradiation in presence of catalyst for the synthesis of compounds **(3a-l)** and **(4a-l)** have been synthesized within effective high yield result shown in Table 1.

Table 1. Microwave synthesis for schiff base **3a-l** catalysed by heterogenous catalyst (SiO_2/γ-Fe_2O_3) and β-lactams **4a-l** .

Compound	Power, W	Microwave	
		Yield, %	Time, min
3a	800	94	1
3b	800	91	1.5
3c	800	88	1
3d	800	90	2
3e	800	82	2
3f	800	84	2
3g	800	94	2
3h	800	96	1
3i	800	88	3
3j	800	93	2
3k	800	88	1
3l	800	89	1
4a	800	87	2
4b	800	85	2.5
4c	800	88	2
4d	800	87	3.5
4e	800	77	3.5
4f	800	82	2
4g	800	85	1
4h	800	87	2.5
4i	800	75	2
4j	800	92	1.5
4k	800	92	2.5
4l	800	91	2

On microwave irradiation power.

Conclusions

In the chemical synthesis, MW methodology has opened up new route and opportunities for the synthetic chemists and researchers by providing novel methods not practical by conventional methods, this is a wide application in synthetic organic chemistry. MW assisted reactions are found to be rapid, efficient and safe. The approach is eco-friendly and this pollution preventive strategy is now an important part of modern combinatorial and green chemistry. Recent synthetic studies on various processes by utilizing MW irradiation have shown the high potential of MW irradiation in obtaining high yield products. The experimental procedures in microwave methods are simple

and economic. More research should have been performed to explore novel chemical compounds which can be synthesized under MW-assisted methodology.

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