

Solvent Free Efficient One Pot Synthesis of Acridinedione by Using $\text{SiO}_2\text{:MgZrO}_3$ Recyclable Metal Oxide Catalyst

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A highly effective and fast processing method was reported for the preparation of several acridinedione derivatives using a catalytic amount of silica-containing MgZrO_3 ($\text{SiO}_2\text{:MgZrO}_3$). The synthesized metal oxide material was characterized using FTIR, XRD, SEM-EDAX, TEM and BET surface area analysis. FTIR analysis confirmed the characteristic Mg–O vibrational band in the range of 600–400 cm^{-1} , along with bands at 550 and 405 cm^{-1} attributed to Si–O and Zr–O stretching vibrations, respectively. XRD analysis revealed a well-defined crystalline structure consistent with JCPDS card No. 19-2936. SEM imaging demonstrated a uniform cubic surface morphology, while EDAX analysis confirmed the presence of Zr, Mg, Si and O, supporting the successful formation of the desired material. TEM images further revealed the morphological features of the material and the corresponding SAED pattern was consistent with the crystalline phases identified by XRD. The material exhibited a BET surface area of 103.25 cm^2/g , which indicate a relatively high accessible surface area that may support its potential application in surface-mediated processes. Low catalytic loading of silica-containing MgZrO_3 was found to be an economical and reusable catalyst for organic transformation. The obtained percentage yield of the product was good, with a cost-effective experimental setup and the final products were pure and facile. The catalyst can be reused several times without loss of product yield or activity.

Keywords: Acridinedione, SiO_2 , MgZrO_3 , Nanoparticle, Recyclable, Metal oxide.

INTRODUCTION

Last decades an efficient synthetic method has been used for the synthesis of numerous organic and pharmacological materials by using multi-component reactions (MCRs) [1]. This method has capability to prepare the complex drug molecules such as heterocyclic compounds those are present in chemical structure of natural dyes, pharmaceutical drug molecules, alkaloids, agrochemicals, biological molecules such as DNA and RNA [2]. MCRs have several benefits such as have excellent structural diversity and complexity, cost effective, high yield with simple procedure to enhance atom economy, quick synthesis with selectivity, suitable for rapid and eco-friendly discovery of novels and complex drug molecules [3-5].

Heterocyclic compounds have cyclic rings those have at least one non-carbon atoms commonly like nitrogen, sulphur and oxygen. Acridine derivatives as an important class of nitrogen heterocyclic compounds with 1,4-dihydropyridine (DHP) ring are the core structure of a number of biologically interesting compounds due to their wide range of biological and photophysical properties [6,7]. Among these acridine-1-8-dione derivatives represent a distinctive subclass characterized by a rigid tricyclic framework and presence of two carbonyl functional groups [8]. This unique structural arrangement imparts remarkable chemical stability and functional versatility, making acridinedione valuable scaffolds in medicinal chemistry and material science [9,10].

Acridinediones and their derivatives have attracted significant interest due to their diverse biological activities and applications in various fields [11,12]. Furthermore, acridinedione derivatives have a wide range of biological activities such as antimicrobial [13,14], antifungal [15], antibacterial [16], antitumor [17], anticancer [18,19], antimalarial [20], DNA binding properties [21] and inhibitors of carbonic anhydrase [22]. Some derivatives have been investigated for their potential as β -channel openers in cardiovascular disease. 1,4-DHPs are also utilized as electroluminescence materials and can be used in laser dyes due to their strong fluorescent activities [23]. Acridinediones can be used as selective chemosensors for the detection of specific ions, particularly fluoride ions. This is achieved through a mechanism involving the abstraction of a proton facilitated by the fluoride ion, leading to observable spectroscopic changes [24]. Acridinediones can be used as photosensitizers and in organic dye laser applications [25]. Acridinedione derivatives have been explored for their potential applications in cosmetics, particularly in addressing pigmentation disorders and skin-aging-related concerns [26].

The multicomponent Hantzsch synthesis is an efficient one-pot approach for preparing acridinediones through the condensation of aldehydes, β -diketones and nitrogen-containing compounds such as urea, ammonium acetate, aniline or primary amines [27,28]. Various homogeneous and heterogeneous catalysts have been reported [29-38]; however, many methods require expensive or hazardous reagents/solvents, prolonged reaction times, harsh conditions, and tedious work up procedures [39-41]. Therefore, developing a green, economical and practical solvent-free approach is highly desirable.

EXPERIMENTAL

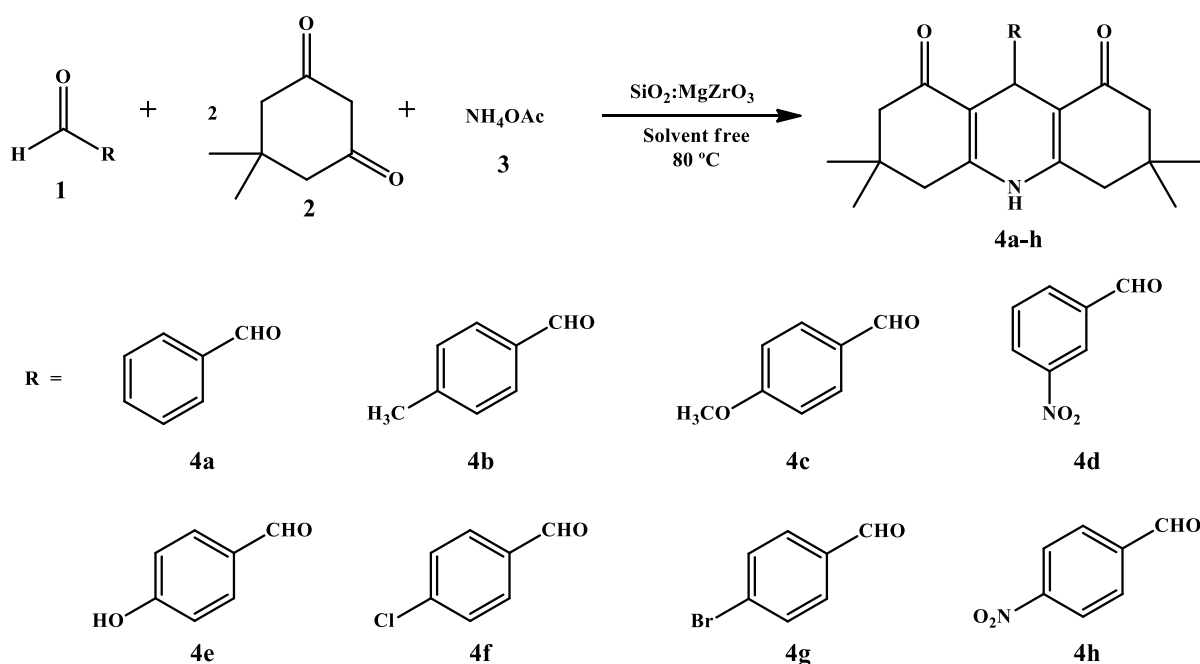
The starting materials, MgCl₂, ZrOCl₂ (Merck 99%), SiO₂ (Merck, >99%) dodecylbenzene sulphonic acid (Loba, 99.5%),

were used as reactants for the preparation of the catalyst. Double deionized water was used throughout the experiment.

Synthesis of SiO₂:MgZrO₃ catalyst: In this method, a green chemistry approach was adopted for the synthesis of heterogeneous metal oxide catalysts. An equimolar amount (1:1 ratio) of AR grade SiO₂ and MgZrO₃ mixed thoroughly and calcined at 600 °C for 3 h. The mixture was ground with mortar and pestle to acquire fine powder. The obtained powder was further calcined at 800 °C and grind by mortar and pestle after each interval of 3 h of time. Afterward, at the end mixture was heated up to the terminal temperature. The furnace was programmed as 10 °C/min from one temperature to subsequent higher temperature. The product SiO₂:MgZrO₃ was obtained (**Scheme-I**).

Characterization: The synthesized SiO₂:MgZrO₃ catalyst was characterized using various analytical techniques. FTIR spectra were recorded using an 8400S-Shimadzu spectrometer in the range of 4000–400 cm⁻¹. Optical properties were analyzed using a Perkin-Elmer Lambda-950 UV-visible spectrophotometer over 200–800 nm. X-ray diffraction analysis was performed using a Rigaku D/MAX-2500 diffractometer with CuK α radiation ($\lambda = 1.5406$ Å). Surface morphology and elemental composition were examined using a JEOL JED-2300LA scanning electron microscope coupled with an energy dispersive spectrometer. TEM images were obtained using a Philips CM-200 transmission electron microscope. The specific surface area (S_{BET}), pore volume (V_p) and pore diameter (D_p) were determined using a Quantachrome Autosorb automated gas sorption system (Autosorb-1 NOVA-1200) and a Mercury Porosimeter (Autosorb-IC).

General procedure for the synthesis of acridinedione using SiO₂:MgZrO₃ catalyst: In a 25 mL round bottom flask, mixture of an appropriate quantity of aromatic aldehyde (1 mmol), ammonium acetate (1.2 mmol), dimedone (2.2 mmol) and SiO₂:MgZrO₃ (0.6 mmol) were mixed. The react-



Scheme-I: Synthesis of acridinedione using SiO₂:MgZrO₃ catalyst

ion mixture was heated at 80 °C under solvent free condition for 35 min. After completion of the reaction, the reaction mixture was cooled to room temperature and then poured over crushed ice, stirred for 10-15 min, precipitated product was filtered, washed with water and dried. The product was placed in beaker and dissolving it in ethanol, after that the solution was filtered, the catalyst gets separated on filter paper and reaction product in the form of filtrate. This filtrate was evaporated; the crude product was obtained. This crude product was recrystallized by using ethanol again, the recrystallized product gives the isolated yield for various substituted acridinedione derivatives.

3,3,6,6-Tetramethyl-1,8-dioxo-9-(phenyl)decahydroacridine (4a): Yield: 93%; m.p.: 292-294 °C, Time: 35 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3348 (-N-H), 2877 (=C-H), 1608 (-C=O), 1508 (-C=C-), 1018 (C-O), 405 (Ar-CH); ^1H NMR (DMSO, 300 MHz, δ ppm): 9.10 (brs, 1H, -NH), 7.20 (m, 5H, Ar-H), 3.40 (s, 1H, -CH), 3.19 (s, 1H, -CH), 2.84 (s, 1H, -CH), 2.78 (m, 2H, -CH₂), 2.25 (m, 2H, -CH₂), 1.70 (m, 2H, -CH₂), 1.45 (m, 2H, -CH₂), 1.13 (s, 6H, -CH₃), 1.14 (s, 6H, -CH₃); ^{13}C NMR (CDCl₃, 75 MHz, δ ppm): 192.50 (-C=O), 151.82 (-C=C-), 149.75 (Ar-C-), 131.50 (-C=C-), 124.13 (Ar-CH), 50.35 (-CH₂-), 49.01 (-CH₂-), 43.96 (-CH-), 29.10 (-C-), 26.52 (-CH₃); m.f.: C₂₃H₂₉NO₂, calcd. MS: 350.20, EI-MS m/z : 351.25 (M+1).

3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-methylphenyl)-decahydroacridine (4b): Yield: 83%; m.p.: 286-288 °C, Time: 45 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3318 (-N-H), 2870 (=C-H), 1670 (-C=O), 1567 (C=C), 1248 (-C-N-); ^1H NMR (DMSO, 300 MHz, δ ppm): 9.15 (brs, 1H, -NH), 7.10 (m, 5H, Ar-H), 3.30 (s, 1H, -CH), 3.17 (s, 1H, -CH), 2.74 (s, 1H, -CH), 2.68 (m, 2H, -CH₂), 2.36 (s, 3H, -CH₃), 2.20 (m, 2H, -CH₂), 1.65 (m, 2H, -CH₂), 1.42 (m, 2H, -CH₂), 1.15 (s, 6H, -CH₃), 1.13 (s, 6H, -CH₃); ^{13}C NMR (DMSO, 75 MHz, δ ppm): 197.21 (-C=O), 141.21 (-C=C-), 137.74 (-C=C-), 127.35 (Ar-CH), 48.47 (-CH₂-), 45.11 (-CH₂-), 42.05 (-CH-), 28.15 (-C-), 24.55 (-CH₃), 26.72 (-CH₃); m.f.: C₂₄H₃₁NO₂, calcd. MS: 364.22, EI-MS m/z : 365.20 (M+1).

3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-methoxyphenyl)-decahydroacridine (4c): Yield: 90%; m.p.: 289-291 °C, Time: 30 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3509 (-N-H), 2965 (=C-H), 1680 (-C=O), 1566 (C=C-), 1260 (-C-O); ^1H NMR (DMSO, 300 MHz, δ ppm): 9.12 (brs, 1H, -NH), 7.21 (m, 5H, Ar-H), 3.75 (s, 3H, CH₃), 3.40 (s, 1H, -CH), 3.20 (s, 1H, -CH), 2.83 (s, 1H, -CH), 2.77 (m, 2H, -CH₂), 2.26 (m, 2H, -CH₂), 1.72 (m, 2H, -CH₂), 1.44 (m, 2H, -CH₂), 1.12 (s, 6H, -CH₃), 1.10 (s, 6H, -CH₃); ^{13}C NMR (DMSO, 75 MHz, δ ppm): 198.12 (-C=O), 158.10 (-C=C), 142.33 (Ar-C-), 135.36 (-C=C), 128.15 (Ar-CH), 126.12 (Ar-CH), 54.60 (-OCH₃), 52.25 (-CH₂-), 45.82 (-CH₂-), 48.65 (-CH), 28.26 (-C-), 26.18 (-CH₃); m.f.: C₂₄H₃₁NO₃, calcd. MS: 381.21, EI-MS m/z : 382.12 (M+1).

3,3,6,6-Tetramethyl-1,8-dioxo-9-(3-nitrophenyl)decahydroacridine (4d): Yield: 78%; m.p.: 302-304 °C, Time: 35 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3220 (-N-H), 3050 (=C-H), 1650 (-C=O), 1535 (C=C), 1340 (-NO₂); ^1H NMR (CDCl₃, 300 MHz, δ ppm): 9.18 (brs, 1H, -NH), 7.45-8.25 (m, 4H, Ar-H), 3.69 (s, 3H, CH₃), 3.45 (s, 1H, -CH), 3.25 (s, 1H, -CH), 2.80 (s, 1H, -CH), 2.75 (m, 2H, -CH₂), 2.29 (m, 2H, -CH₂), 1.76

(m, 2H, -CH₂), 1.48 (m, 2H, -CH₂), 1.25 (s, 6H, -CH₃), 1.12 (s, 6H, -CH₃); ^{13}C NMR (DMSO, 75 MHz, δ ppm): 199.50 (-C=O), 148.32 (Ar-C-), 146.25 (Ar-CH), 141.50 (-C=C-), 139.36 (Ar-CH), 125.12 (Ar-CH), 120.10 (Ar-CH), 140.26 (-C=C), 48.36 (-CH₂-), 44.15 (-CH₂-), 30.28 (-CH-), 29.45 (-C-), 26.14 (-CH₃); m.f.: C₂₃H₂₈N₂O₄, calcd. MS: 396.19, EI-MS m/z : 397.19 (M+1).

3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-hydroxyphenyl)-decahydroacridine (4e): Yield: 79%; m.p.: 303-305 °C, Time: 45 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3428 (-OH), 3252 (-N-H), 2965 (=C-H), 1640 (-C=O-), 1560 (-C=C-); ^1H NMR (CDCl₃, 300 MHz, δ ppm): 9.15 (brs, 1H, -NH), 7.22 (m, 5H, Ar-H), 5.10 (s, Ar-OH), 3.42 (s, 1H, -CH), 3.17 (s, 1H, -CH), 2.79 (s, 1H, -CH), 2.69 (m, 2H, -CH₂), 2.20 (m, 2H, -CH₂), 1.73 (m, 2H, -CH₂), 1.41 (m, 2H, -CH₂), 1.12 (s, 6H, -CH₃), 1.13 (s, 6H, -CH₃); ^{13}C NMR (DMSO, 75 MHz, δ ppm): 198.42 (-C=O), 157.27 (C=C), 132.21 (Ar-C-), 126.86 (Ar-CH), 127.64 (Ar-CH), 115.30 (Ar-CH), 139.28 (-C=C-), 50.58 (-CH₂-), 43.38 (-CH₂-), 31.69 (-CH), 29.21 (-C-), 25.30 (-CH₃); m.f.: C₂₃H₂₉NO₃, calcd. MS: 367.20, EI-MS: 368.24 (M+1).

3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-chorophenyl)decahydroacridine (4f): Yield: 81%; m.p.: 294-296 °C, Time: 30 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3138 (-N-H), 3043 (=C-H), 1680 (-C=O-), 1578 (-C=C-), 1243 (C-N-); ^1H NMR (CDCl₃, 300 MHz, δ ppm): 9.09 (brs, 1H, -NH), 7.22-7.24 (m, 4H, Ar-H), 3.48 (s, 1H, -CH), 3.35 (s, 1H, -CH), 2.90 (s, 1H, -CH), 2.81 (m, 2H, -CH₂), 2.35 (m, 2H, -CH₂), 1.77 (m, 2H, -CH₂), 1.51 (m, 2H, -CH₂), 1.23 (s, 6H, -CH₃), 1.10 (s, 6H, -CH₃); ^{13}C NMR (DMSO, 75 MHz, δ ppm): 200.42 (-C=O), 137.29 (C=C), 133.31 (Ar-C-), 128.86 (Ar-CH), 128.64 (Ar-CH), 127.30 (Ar-CH), 139.28 (-C=C-), 52.55 (-CH₂-), 44.48 (-CH₂-), 33.59 (-CH), 30.21 (-C-), 26.30 (-CH₃); m.f.: C₂₃H₂₈ClNO₂, calcd. MS: 385.17, EI-MS: 387.14 (M+2).

3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-bromophenyl)decahydroacridine (4g): Yield: 72%; m.p.: 272-274 °C, Time: 55 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3218 (-NH), 3045 (=C-H), 1645 (-C=O), 1560 (-C=C-), 1234 (-C-N); ^1H NMR (DMSO, 300 MHz, δ ppm): 9.11 (brs, 1H, -NH), 7.20-7.32 (m, 4H, Ar-H), 3.45 (s, 1H, -CH), 3.40 (s, 1H, -CH), 2.92 (s, 1H, -CH), 2.78 (m, 2H, -CH₂), 2.45 (m, 2H, -CH₂), 1.80 (m, 2H, -CH₂), 1.55 (m, 2H, -CH₂), 1.22 (s, 6H, -CH₃), 1.12 (s, 6H, -CH₃); ^{13}C NMR (DMSO, 75 MHz, δ ppm): 199.22 (-C=O), 142.19 (C=C), 133.31 (Ar-C-), 138.56 (Ar-CH), 128.50 (Ar-CH), 122.31 (Ar-CH), 139.28 (-C=C-), 52.55 (-CH₂-), 44.48 (-CH₂-), 33.59 (-CH), 30.21 (-C-), 26.30 (-CH₃); m.f.: C₂₃H₂₈BrNO₂, calcd. MS: 429.11, EI-MS: 431.14 (M+2).

3,3,6,6-Tetramethyl-1,8-dioxo-9-(4-nitrophenyl)decahydroacridine (4h): Yield: 76%; m.p.: 282-284 °C, Time: 55 min; FT-IR (KBr, $\nu_{\max}$, cm^{-1}): 3418 (-NH), 3056 (=C-H), 1665 (-C=O-), 1545 (-C=C-), 1360 (-NO₂), 1254 (-C-N-); ^1H NMR (DMSO, 300 MHz, δ ppm): 9.28 (brs, 1H, -NH), 8.05 (d, J = 8.2 Hz, 2H, Ar-H), 7.08 (d, J = 8.2 Hz, 2H, Ar-H), 3.60 (s, 1H, -CH), 3.50 (s, 1H, -CH), 2.98 (s, 1H, -CH), 2.81 (m, 2H, -CH₂), 2.52 (m, 2H, -CH₂), 1.89 (m, 2H, -CH₂), 1.51 (m, 2H, -CH₂), 1.13 (s, 6H, -CH₃), 1.13 (s, 6H, -CH₃); ^{13}C NMR (DMSO, 75 MHz, δ ppm): 199.50 (-C=O), 148.32 (Ar-C-), 146.25 (Ar-CH), 141.50 (-C=C-), 139.36 (Ar-CH), 125.12 (Ar-CH), 121.10 (Ar-CH), 141.16 (-C=C), 47.32 (-

CH₂), 43.14 (-CH₂), 29.18 (-CH-), 26.45 (-C-), 25.24 (-CH₃)
m.f.: C₂₃H₂₈N₂O₄, calcd. MS: 396.19, EI-MS: 397.14 (M+1).

RESULTS AND DISCUSSION

Characterization of SiO₂:MgZrO₃ catalyst

FTIR analysis: The FTIR spectrum of the modified SiO₂:MgZrO₃ catalyst is shown in Fig. 1. The broad vibrational region between 600 and 400 cm⁻¹ is associated with Mg–O vibrations, while the absorption bands at 550 and 405 cm⁻¹ can be assigned to Si–O and Zr–O stretching vibrations, respectively. The bands observed at 3348 and 1608 cm⁻¹ correspond to the O–H stretching and deformation vibrations, respectively, which can be attributed to the surface hydroxyl groups associated with the water-based synthesis [42]. These characteristic bands confirm the incorporation of the constituent metal oxides in the modified catalyst.

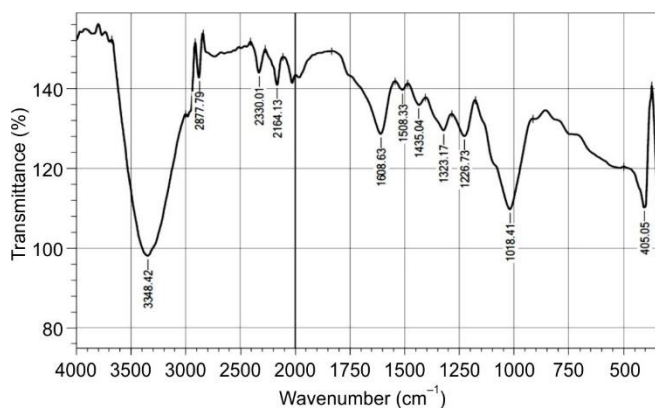


Fig. 1. FT-IR spectrum SiO₂:MgZrO₃ catalyst

XRD analysis: The crystalline structure of SiO₂:MgZrO₃ was investigated by XRD, and the corresponding pattern is shown in Fig. 2. The diffraction peaks at $2\theta = 31.61^\circ$, 34.43° , 42.87° , 45.33° , 50.43° and 62.65° were indexed to the (100), (010), (110), (101), (200), and (210) crystal planes, respectively. The observed reflections are in good agreement with JCPDS card No. 19-2936, confirming the crystalline nature

of the material. The reflections at 34.43° and 50.43° may further indicate the presence of a tetragonal ZrO₂/SiO₂-related phase. The well-defined diffraction peaks support the formation of a crystalline SiO₂:MgZrO₃ metal-oxide composite and are consistent with the morphological observations obtained from SEM and TEM analyses.

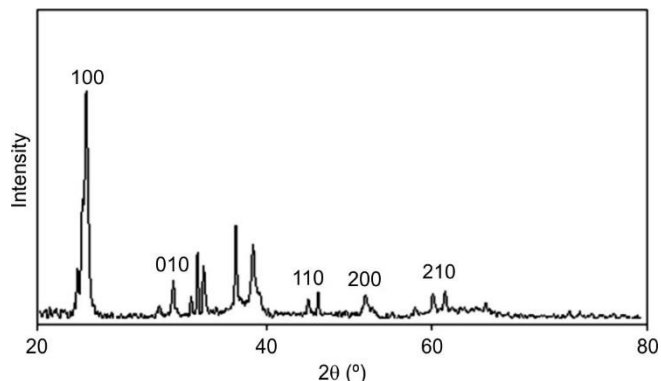


Fig. 2. XRD spectrum of SiO₂:MgZrO₃ metal oxide

Morphological studies: The surface morphology and elemental composition of the catalyst were examined by SEM-EDAX (Fig. 3). The SEM micrograph reveals relatively uniform, cubic-shaped crystalline nanoparticles. Incorporation of SiO₂ into MgZrO₃ followed by calcination at 800 °C resulted in a change in surface morphology, without a significant alteration in particle size. The aggregated particles were distributed within approximately 10-200 nm. EDAX analysis confirmed the presence of Mg, Zr, Si and O, with the elemental composition consistent with the constituent materials used for catalyst preparation. These observations support the successful formation of the SiO₂:MgZrO₃ composite.

The TEM image and corresponding SAED pattern are shown in Fig. 4. The TEM analysis further confirms the predominantly cubic morphology of the nanoparticles. The observed crystalline features and SAED reflections are consistent with the diffraction pattern obtained from XRD, providing further evidence for the crystalline nature of the synthesized SiO₂:MgZrO₃ catalyst.

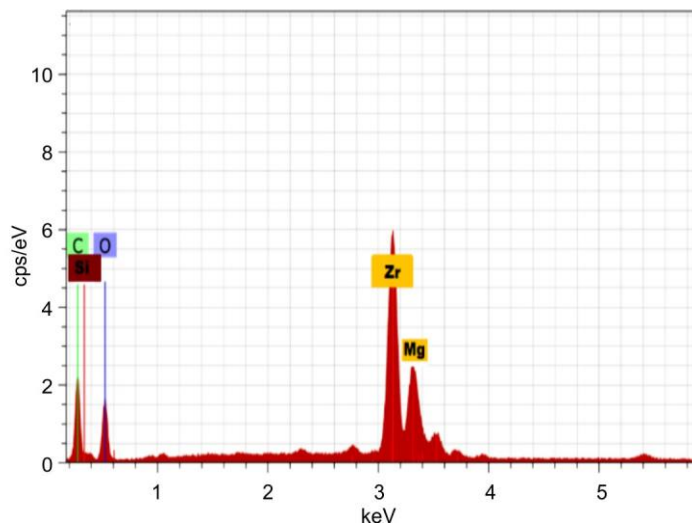
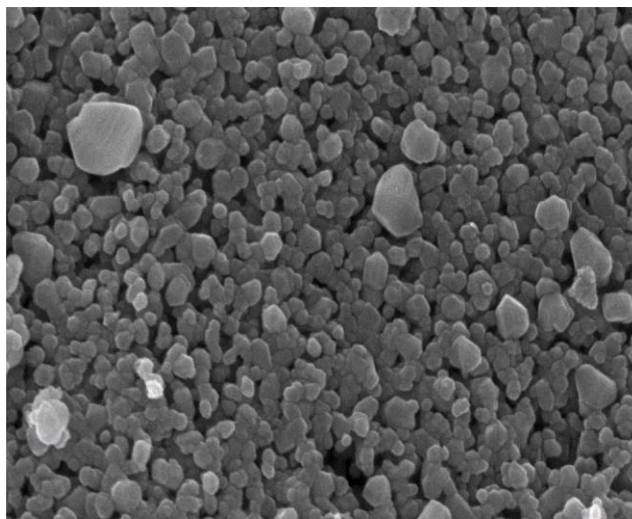


Fig. 3. SEM image and EDAX spectrum of SiO₂:MgZrO₃ nanoparticles

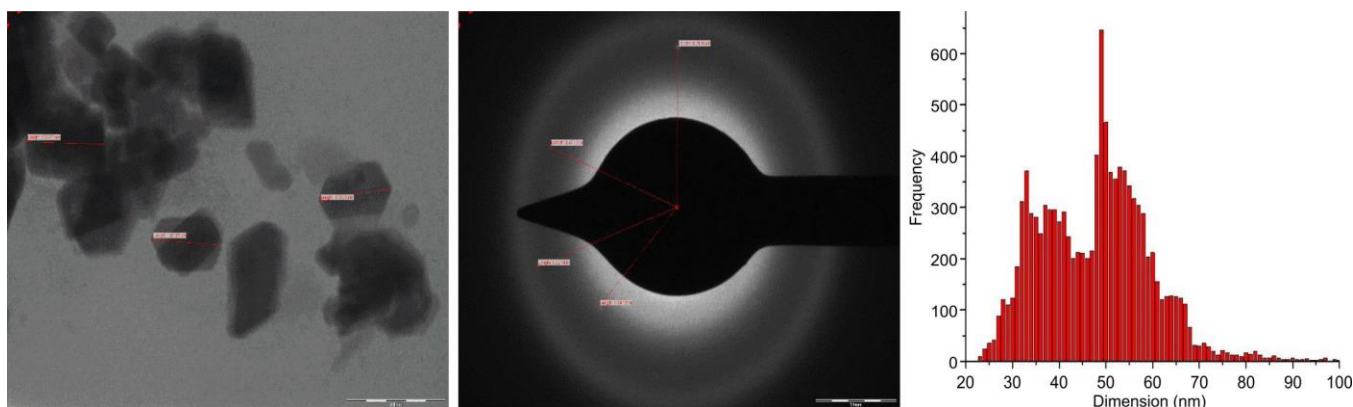


Fig. 4. TEM image, SAED pattern and particle size distribution of the $\text{SiO}_2\text{:MgZrO}_3$ catalyst

BET analysis: The textural properties of the catalyst were investigated by N_2 adsorption–desorption analysis, and the corresponding isotherm and BJH pore-size distribution are shown in Fig. 5. The catalyst exhibited a type-III adsorption–desorption isotherm with an H1 hysteresis loop, indicating the presence of mesoporous characteristics with predominantly cylindrical pores. The BJH analysis showed a relatively narrow pore-size distribution. The BET surface area, pore volume, and average pore diameter were determined to be $103.25 \text{ m}^2 \text{ g}^{-1}$, $0.85 \text{ cm}^3 \text{ g}^{-1}$ and $15.76 \text{ \AA}$, respectively. The relatively high surface area and mesoporous characteristics provide accessible surface sites for interaction with the reactants and may contribute to the observed catalytic efficiency.

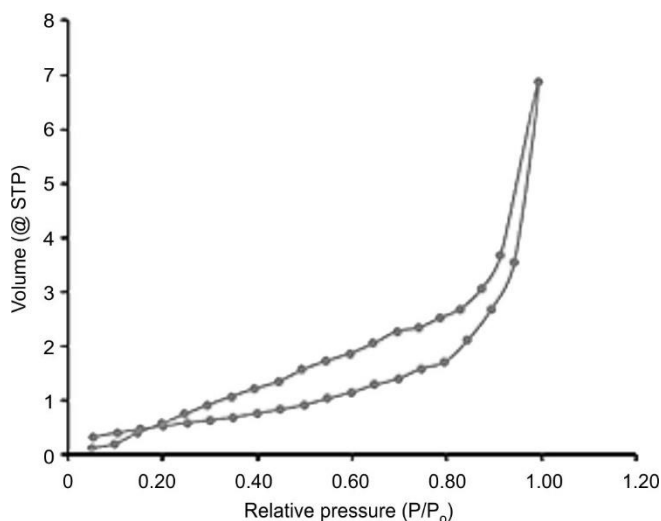


Fig. 5. BET adsorption-desorption isotherm of $\text{SiO}_2\text{:MgZrO}_3$ catalyst

Catalytic synthesis of acridinediones: The catalytic activity of $\text{SiO}_2\text{:MgZrO}_3$ was evaluated for the one-pot synthesis of acridinediones through the cyclocondensation of dimedone, substituted aromatic aldehydes and ammonium acetate under solvent-free conditions (**Scheme-I**). The reaction afforded the corresponding acridinediones **4a-h** in good to excellent yields, demonstrating the effectiveness of the modified metal-oxide catalyst for this multicomponent transformation.

In the absence of a catalyst, the model reaction did not proceed effectively (Table-1, entry 1). SiO_2 and MgZrO_3

individually promoted the reaction but afforded lower yields than the modified $\text{SiO}_2\text{:MgZrO}_3$ catalyst. In contrast, $\text{SiO}_2\text{:MgZrO}_3$ provided a substantially improved yield within a shorter reaction time (Table-1, entry 4). This enhanced performance can be associated with the combined surface characteristics of the metal-oxide components, including the accessible surface area and active surface sites of the composite.

TABLE-1
EFFECT OF DIFFERENT CATALYSTS
ON REACTION TIME AND YIELD

Entry	Catalyst	Time (min)	Isolated yield (%)
1	Without catalyst	60	Incomplete reaction
2	SiO_2	60	32
3	MgZrO_3	60	47
4	$\text{SiO}_2\text{:MgZrO}_3$	35	93

The catalyst loading was subsequently optimized using the model reaction between dimedone, benzaldehyde and ammonium acetate. Increasing the catalyst amount improved the product yield up to an optimum loading, with 93% yield of **4a** obtained under the optimized loading conditions (Table-2, entry 2). A catalyst loading of only 1.0 mol% was sufficient to promote the reaction within 35 min, whereas further increases in catalyst loading did not result in a significant improvement in yield. This result demonstrates the efficiency of $\text{SiO}_2\text{:MgZrO}_3$ at a relatively low catalyst loading.

TABLE-2
EFFECT OF VARIOUS MOLE PERCENTAGE
OF $\text{SiO}_2\text{:MgZrO}_3$ CATALYST

Entry	Weight % of $\text{SiO}_2\text{:MgZrO}_3$	Time (min)	Isolated yield (%)
1	0.5	35	63
2	1.0	35	93
3	1.5	35	93
4	2.0	35	73

The influence of the reaction medium was examined using solvent-free conditions, water, DMF, methanol and ethanol (Table-3, entries 1-5). The reaction proceeded slowly in water, whereas the solvent-free protocol provided the best result, affording 93% product yield. The solvent-free conditions were

therefore selected for subsequent reactions owing to the higher yield, reduced solvent requirement, and simplified work-up procedure. Under the optimized conditions, aromatic aldehydes **1**, dimedone **2** and ammonium acetate **3** were employed in a molar ratio of 1:2:1.5 and heated at 80 °C for 30-40 min. Both electron-rich and electron-deficient aromatic aldehydes underwent the multicomponent reaction efficiently, furnishing the corresponding substituted acridinediones **4a-h** in 72-93% yields (Table-3).

TABLE-3
EFFECT OF DIFFERENT SOLVENTS FOR
ACRIDINEDIONES USING SiO₂:MgZrO₃

Entry	Solvent	Time (min)	Isolated yield (%)
1	Solvent free	35	93
2	Water	35	34
3	DMF	35	43
4	CH ₃ OH	35	78
5	CH ₂ Cl ₂	35	61

The electronic nature of the aldehyde substituent also influenced the reaction outcome. Aldehydes bearing electron withdrawing groups generally afforded higher yields than those containing electron-donating groups. This behaviour can be rationalized by the increased electrophilicity of the aldehydic carbon in electron-deficient substrates, which facilitates the initial condensation step involved in acridinedione formation.

Reusability of the catalyst: The recyclability of SiO₂:MgZrO₃ was investigated using the model reaction under the optimized conditions. After completion of the reaction, the catalyst was separated, washed with methanol, and dried at 150 °C for 4 h before reuse. The catalyst retained good to excellent catalytic activity over five successive cycles, with no significant decrease in product yield (Table-4). No appreciable catalyst leaching was detected during the washing process, as confirmed by carrying out the reaction with the filtrate. The combination of low catalyst loading, solvent-free operation, good product yields, facile work-up and recyclability demonstrates the practical utility of SiO₂:MgZrO₃ as an efficient heterogeneous catalyst for the synthesis of acridinediones.

TABLE-4
RESULTS OF REACTIONS CONDUCTED
WITH THE RECYCLED CATALYST

Entry	Reaction run	Isolated yield (%)
1	1	93
2	2	90
3	3	83
4	4	80
5	5	80

Comparative studies: A comparative evaluation of different catalysts was performed to establish the catalytic role of the SiO₂:MgZrO₃ composite (Table-5).

Conclusion

A cost-effective efficient method is developed for eco-compatible synthesis of substituted acridinediones via one-pot three-component condensation of aromatic aldehyde, dimedone and ammonium acetate in solvent free medium

TABLE-5
CATALYTIC PERFORMANCE OF VARIOUS
CATALYSTS FOR ACRIDINEDIONE SYNTHESIS

Entry	Catalyst	Time (min)	Isolated yield (%)	Ref.
1	Silica sulfuric acid	1.5 h	95	[43]
2	In ₂ O ₃ -SiO ₂	100	92	[44]
3	Cellulose sulfuric acid	300	80	[45]
4	KH ₂ PO ₄	300	94	[46]

with SiO₂:MgZrO₃ as an efficient catalyst. The mildness of the conversion, the experimental simplicity, short reaction time, use of cheap, non-toxic, environmentally benign nature makes it an attractive process and the employment of a cost-effective catalyst, compatibility with various functional groups, excellent product yields and the easy work-up procedure make this approach attractive for synthesizing a variety of such derivatives.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

DECLARATION OF AI-ASSISTED TECHNOLOGIES

During the preparation of this manuscript, the authors used an AI-assisted tool(s) to improve the language and no data, results or scientific conclusions were generated by AI. All analyses, interpretations and conclusions are the authors' own. The authors reviewed and edited the content and take full responsibility for the published work.

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