Research Article

Solvent-Free Synthesis of New Coumarins

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A solvent-free synthesis of five series of coumarin derivatives using microwave assistant is presented herein. The synthesized compounds are fully characterized by UV-VIS, FT-IR, and NMR spectroscopy.

1. Introduction

Coumarin (2H-Lbenzopyran-2-one) and its derivatives possess a wide range of various biological and pharmaceutical activities. They have a wide range of applications as antitumor [1, 2], anti-HIV [3, 4], anticoagulant [5, 6], antimicrobial [7, 8], antioxidant [9, 10], and anti-inflammatory [11, 12] agents. The antitumor activities of coumarin compounds have been extensively examined [13–16]. Although most of the existing natural coumarins have been isolated from higher plants, some of them have been discovered in microorganisms, for example, aminocoumarin antibiotics: novobiocin, coumermycin A1, and chlorobiocin (produced by the actinomycete *Streptomyces niveus*) [17]. Synthetic coumarin derivatives have been obtained by chemical modification of the coumarin ring. Recently, density functional theory (DFT) has been accepted by the quantum chemistry community as a cost-effective approach for the computation of molecular structure, vibration frequencies, and energies of chemical reactions. Many studies have shown that the molecular structures and vibration frequencies calculated by DFT methods are more reliable than MP2 methods [18–26]. While there is sufficient evidence that DFT provides accurate description of the electronic and structural properties of solids, interfaces, and small molecules, relatively little is known about the symmetric performance of DFT applications to their molecular associates.

Structure activity relationships of coumarin derivatives have revealed that the presence of substituted amino derivatives is an essential feature of their pharmacological action. Based on these findings, we try to describe the synthesis of some compounds featuring different heterocyclic rings fused onto the coumarin moiety with the aim of obtaining more potent pharmacologically active compounds.

2. Experimental

2.1. General. The chemicals used for the synthesis were supplied by Sigma-Aldrich. Purity of the compounds was checked on thin layer chromatography (TLC) plates (Silica Gel G) using the solvent systems benzene-ethyl acetate-methanol (40:30:30, v/v/v) and toluene-acetone (75:25, v/v). The spots were located under UV light (254 and 365 nm). Melting points were determined on Gallenkamp (MFB-600) melting point apparatus and were uncorrected. The IR spectra of the compounds were recorded on a Shimadzu FT-IR-8300 spectrometer as KBr disk. The UV-VIS spectra were performed on Cintra-5-Ges scientific equipment. The ¹H-NMR and ¹³C-NMR spectra (solvent DMSO-d₆) were recorded on Bruker 400 MHz spectrophotometer using TMS as internal standard.

2.2. Synthesis of 1-aminoquinolin-2(1H)-one (1). 1-aminoquinolin-2(1H)-one (**1**) was synthesized according to [27],

and the structure of the compound was confirmed with elemental analyses and spectral analyses (IR, UV-VIS, ^1H -NMR, and ^{13}C -NMR).

2.3. Synthesis of (E)-1-((2-hydroxybenzylidene)amino)quinolin-2(1H)-one (2) and (E)-1-((1-(thiophen-2-yl)ethylidene)amino)quinolin-2(1H)-one (3). N-aminocoumarin (0.16 g, 0.001 mol) and 2-hydroxybenzaldehyde (or 1-(thiophen-2-yl)ethanone) (0.001 mol) were placed together in an open small test tube; after 6 minutes, the product was separated out and recrystallized.

2.4. Synthesis of 1,1'-((1Z,1'Z)-(1,4-phenylenebis(methanylylidene))bis(azanylylidene))bis(quinolin-2(1H)-one) (4). N-aminocoumarin (0.32 g, 0.002 mol) and terephthaldehyde (0.134 g, 0.001 mol) were placed together in an open small test tube; after 8 minutes, the product was separated out and recrystallized.

2.5. Synthesis of 2-(2-oxoquinolin-1(2H)-yl)isoindoline-1,3-dione (5) and 3-methylene-1-(2-oxoquinolin-1(2H)-yl)pyrrolidine-2,5-dione (6). N-aminocoumarin (0.16 g, 0.001 mol) and phthalic anhydride (or itaconic anhydride) (0.001 mol) were placed together in an open small test tube; after 5 minutes, the product was separated out and recrystallized.

2.6. Synthesis of 1-(naphthalen-1-yl)-3-(2-oxoquinolin-1(2H)-yl) urea (7). N-aminocoumarin (0.16 g, 0.001 mol) and 1-naphthylisocyanate (0.17 g, 0.001 mol) were placed together in an open small test tube; after 8 minutes, the product was separated out and recrystallized.

2.7. Synthesis of 1,1'-(hexane-1,6-diyl)bis(3-(2-oxoquinolin-1(2H)-yl)urea) (8) and 1,1'-(2-methyl-1,4-phenylene)bis(3-(2-oxoquinolin-1(2H)-yl)urea) (9). N-aminocoumarin (0.32 g, 0.002 mol) and hexamethylene diisocyanate (or toluene-2,5-diisocyanate) (0.001 mol) were placed together in an open small test tube; after 8 minutes, the product was separated out and recrystallized.

2.8. Synthesis of 2-((2-oxoquinolin-1(2H)-yl)amino)thiazol-5(4H)-one (10), 2-(carboxy(2-oxoquinolin-1(2H)-yl)amino)acetic acid (11) and 1-(pyridin-2-ylamino)quinolin-2(1H)-one. N-aminocoumarin (0.16 g, 0.001 mol) and rhodanine (or 2-mercaptopyridine or 2-mercaptosuccinic acid) (0.001 mol) were placed together in an open small test tube; after 8 minutes, the product was separated out and recrystallized.

2.9. Synthesis of 3H-[1,2,4,5]tetrazino[1,6-a]quinolin-2-amine (13). N-aminocoumarin (0.16 g, 0.001 mol) and thiosemicarbazide (0.1 g, 0.001 mol) were placed together in an open small test tube; after 10 minutes, the product was separated out and recrystallized.

2.10. The Calculation Method. Gaussian 03, Revision C.01 [28] was used for the calculation of ground-state geometry

SCHEME 1

which was optimized to a local minimum without any symmetry restrictions using basis set 3-21G [29, 30]. The Becke three-parameter hybrid (B3) [31, 32] exchange functional in combination with the Lee-Yang-Parr (LYP) [33] correlation functional (B3LYP) was used for all geometry optimizations, thermodynamic functions at conditions (temperature = 298.150 Kelvin and pressure = 1.0 Atm), high occupied molecular orbital (HOMO), and low unoccupied molecular orbital (LUMO) distribution, and some physical properties for compound 3.

3. Results and Discussion

3.1. Chemistry. For the synthesis of new coumarin derivatives, the reaction sequences outlined in Schemes 1, 2, 3, 4, 5, 6, 7, and 8 were followed. We started from coumarin (1) which is commercially available or, alternatively, readily accessible through a Pechmann and Perkin condensation [25]. Recrystallization solvent was chloroform. Yield 91%; M.P. 131–133°C. The structure of compound (1) was confirmed from its spectral data. UV-VIS in methanol, λ_{max} nm: 280 (0.93) and 227 (1.8). The IR spectrum showed two strong absorption bands at 3290 to 3300 cm^{-1} and strong band at 1645 cm^{-1} , corresponding to $-\text{NH}_2$ and $-\text{C}=\text{O}$, respectively; 1595 ($\text{C}=\text{C}$ aromatic), 3045 ($\text{C}-\text{H}$ aromatic), 1242 ($\text{C}-\text{N}$). ^1H -NMR: 4.1 (s, 2H, $-\text{NH}_2$), 6.7 (t, Ar-H), 7.4 (d, Ar-H) and 7.1 (d, Ar-H). ^{13}C -NMR: 126, 127, 127.8, 128, 123.3, 128.5, 129, 155 and 157.

3.1.1. Compound (2). Yellow solid; Yield 92; mp 86–88°C; IR (KBr) (cm^{-1}): 3076, 2945, 2920, 1681, 1653, 1583, 1520. ^1H NMR (300 MHz, CDCl_3): (ppm) 2.3 (s, 3H, $-\text{CH}_3$), 7.1–7.9 (9H, Ar-H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 15, 23, 126, 128, 128.2, 129, 129.8, 130, 131, 133, 135, 143, 144, 157.

3.1.2. Compound (3). Yellow solid; Yield 88; mp 215–217°C; IR (KBr) (cm^{-1}): 3200, 3046, 1681, 1621, 1573, 1487. ^1H NMR (300 MHz, CDCl_3): (ppm) 3.3 (s, 1H, $-\text{OH}$), 9.0 (s, 1H, $\text{N}=\text{CH}$), 6.6–7.7; ^{13}C NMR (125 MHz, CDCl_3): (ppm) 17.6, 118.7, 125.0, 125.5, 125.9, 127.4, 128.2, 136.5, 137.1, 139.0, 161.3, 164.6.

3.1.3. Compound (4). Yellow solid; Yield 78; mp 230–232°C; IR (KBr) (cm^{-1}): 3070, 1670, 1604, 1595, 1456. ^1H NMR (300 MHz, CDCl_3): (ppm) 6.1 (dd, 1H, $=\text{CH}$), 7.1–7.6 (m, 1H, Ar-H), 7.9–8.2 (m, 1H, Ar-H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 117.5, 126.0, 126.6, 128.3, 128.7, 131.2, 131.4, 131.9, 133.0, 137.5, 141.9, 159.3.

SCHEME 2

SCHEME 3

SCHEME 4

SCHEME 5

SCHEME 6

SCHEME 7

TABLE 1: Total energy, relative energies and heat of formation for **3**.

Compound	Potential energy	Total energy	Kinetic energy
3	-1445030.1135 Kcal/Mol	-724778.6338 Kcal/Mol	720251.4797 Kcal/Mol

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3.1.4. Compound (5). White solid; Yield 90; mp 340 dec. °C; IR (KBr) (cm^{-1}): 3016, 1661, 1601, 1557 ^1H NMR (300 MHz, CDCl_3): (ppm) 3.1 (s, 2H, $-\text{CH}_2$), 5.9 (s, 1H, $=\text{CH}$), 6.3 and 6.5 (dd, 1H, $=\text{CH}$), 6.9–7.3 (1H, s, C–H, Aromatic); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 17 39.7, 99.6, 117, 122.7, 127.3, 127.6, 127.9, 129.2, 130.9, 140.8, 155.6, 155.9, 169.3.

3.1.5. Compound (6). Yellow solid; Yield 89; mp 94–96 °C; IR (KBr) (cm^{-1}): 3001, 2925, 1681, 1604, 1562. ^1H NMR (300 MHz, CDCl_3): (ppm) 6.6 (dd, 1H, $=\text{CH}$), 6.8 (dd, 1H, $=\text{CH}$), 7.3–7.6 (1H, Ar–H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 110.8, 118.6, 119.3, 121.8, 124, 136, 138.6, 156.2, 156.9.

3.1.6. Compound (7). White solid; Yield 87; mp 216–218 °C; IR (KBr) (cm^{-1}): 3271 (N–H), 3049 (C–H Aromatic), 1660 and 1654 (C=O), 1608 and 1552 (C=C Aromatic). ^1H NMR (300 MHz, CDCl_3): (ppm) 5.9 (s, 1H, $-\text{CH}$), 6.2 (dd, 1H, $=\text{CH}$), 6.9–7.5 (m, $-\text{C}-\text{H}$ Aromatic); 7.6 (dd, $-\text{C}-\text{H}$ Aromatic); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 111.3, 112.2, 122.1, 122.3, 122.5, 122.6, 122.9, 127.1, 129, 129.7, 139.2, 140.1, 144.2, 144.6, 153.9, 154.2, 163.6.

3.1.7. Compound (8). White solid; Yield 91; mp 248–250 °C; IR (KBr) (cm^{-1}): 3300 (N–H), 3095 (C–H Aromatic), 2931, 2858 (C–H ali), 1664 (C=O), 1556, 1458 (C=C Aromatic). ^1H NMR (300 MHz, CDCl_3): (ppm) 1.3 (t, 2H, CH_2CH_2), 6.3 (s, 1H, $-\text{NHCO}$), 3.3 (s, 2H, NHCH_2), 6.9–7.5 (6H, Ar–H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 111.8, 114.0, 114.3, 117.5, 119.0, 121.1, 121.2, 121.6, 121.9, 124.1, 124.6, 128.2, 130.3, 131.2, 143.1, 144.2, 144.7, 144.9, 152.9, 161.0.

3.1.8. Compound (9). White solid; Yield 88; mp 210–212 °C; IR (KBr) (cm^{-1}): 3307 (N–H), 3020 (C–H Aromatic), 2926 and 2850 (C–H ali), 1670 and 1658 (C=O), 1633 and 1602 (C=C arom). ^1H NMR (300 MHz, CDCl_3): (ppm) 2.3 (m, 2H, $-\text{CH}_2$), 3.1 (m, 2H, $-\text{CH}_2$), 6.9–7.2 (m, 1H, Ar–H); 7.3–7.5 (1H, Ar–H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 28.2,

31.9, 37.0, 117.6, 122.4, 122.8, 124.1, 124.4, 125.2, 141.3, 143.0, 158.6.

3.1.9. Compound (10). Yellow Yield 93; oily; IR (KBr) (cm^{-1}): 3182 (N–H), 3072 (C–H Aromatic), 1668 (C=O), 1598 and 1456 (C=C arom), 1573 (C=N). ^1H NMR (300 MHz, CDCl_3): (ppm) 3.6 (s, 2H, $-\text{CH}_2$), 5.8 (d, 1H, $=\text{C}-\text{H}$), 6.4 (d, 1H, $=\text{C}-\text{H}$), 7.0–7.3 (1H, Ar–H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 41.0, 111.6, 119.1, 122.3, 122.6, 122.9, 125.3, 146.8, 151.1, 152.9, 156.1.

3.1.10. Compound (11). Yellow solid; Yield 90; mp 102–104 °C; IR (KBr) (cm^{-1}): 3182, 3070, 1708, 1570. ^1H NMR (300 MHz, CDCl_3): (ppm) 3.8 (s, 2H, CH_2), 5.9 (dd, 1H, $=\text{C}-\text{H}$), 6.6 (d, 1H, $=\text{C}-\text{H}$), 6.9–7.2 (1H, Ar–H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 42.7, 117.4, 120.1, 121.9, 122.1, 122.4, 122.8, 15.3, 127.8, 151.4, 158.5, 161.4, 168.8.

3.1.11. Compound (12). Yellow oily; Yield 90; IR (KBr) (cm^{-1}): 3217 (N–H), 3180–2563 (OH), 2945, 2920, 1714, 1651, 1568, 1462. ^1H NMR (300 MHz, CDCl_3): (ppm) 5.2 (s, 1H, $-\text{NH}$), 6.1 (d, 1H, $=\text{C}-\text{H}$), 6.4 (d, 1H, $=\text{C}-\text{H}$), 7.0–7.2 (m, 1H, Ar–H), 7.3–7.5 (m, 1H, Ar–H); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 117.9, 118.4, 120.4, 121.1, 121.4, 122.5, 137.9, 145.0, 152.1, 155.9, 171.0, 171.7.

3.1.12. Compound (13). Yellow solid; Yield 87; solid; mp 92–94 °C; IR (KBr) (cm^{-1}): 3254 and 3147 (N–H), 3003, 2676, 1622, 1577, 1556. ^1H NMR (300 MHz, CDCl_3): (ppm) 5.9 (1H, s, $=\text{C}-\text{H}$), 6.4 (1H, s, $=\text{C}-\text{H}$), 6.9 (1H, dd, $-\text{C}-\text{H}$ aromatic), 7.3 (1H, dd, $-\text{C}-\text{H}$ aromatic); ^{13}C NMR (125 MHz, CDCl_3): (ppm) 113.1, 117.0, 122.4, 127.6, 127.9, 133.1, 136.1, 140.5, 140.8, 157.2.

3.2. Computational Studies

3.2.1. Atomic Charges (Mulliken Charges). An earlier study [34] has shown that atomic charges were affected by the presence of the substituent of rings. For compound **3** the 3D geometrical structure is given in Figure 1. The data obtained show that highest atomic charge in compound **3** is at [N(7) –0.744890] followed by the next charge value at [O(11) –0.0. 500312]. These data show clearly that these atoms are the most reactive toward the addition, substitution reactions, and bonding with the metal. The determined bond angle and twist angle, stretch (1. 9443), bend (7. 6317), stretch-bend (0. 0909), and the 3D geometrical structure indicate that this molecule is a nonplanar molecular and the stereochemistry is [C(9): C(10): (Z); N(12)-C(13): (Z)].

FIGURE 1: Optimized 3D geometrical structure for compound 3.

FIGURE 2: The highest occupied molecular orbital (HOMO) of compound 3.

3.2.2. Density Function Theory (DFT). DFT calculations were performed for compound 3. Optimized molecular structure of the most stable form is shown in Figure 1, Table 1; the calculated energies and relative energies are presented in Table 1. Molecular orbital calculations provide a detailed description of orbitals including spatial characteristics, nodal patterns, and individual atom contributions. The contour plots of the frontier orbitals for the ground state of compound 3 are shown in Figures 2 and 3, including the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO). It is interesting to see that both orbitals are substantially distributed over the conjugation plane. It can be seen from Figure 2 that the HOMO orbitals are located on the substituted molecule while LUMO orbitals resemble those obtained for the unsubstituted molecule, and therefore the substitution has

an influence on the electron donation ability, but only a small impact on electron acceptance ability [35]. The orbital energy levels of HOMO and LUMO of compound 3 are listed in Table 2. It can be seen that the energy gaps between HOMO and LUMO are about -5.419 eV. The lower value in the HOMO and LUMO energy gap explains the eventual charge transfer interaction taking place within the molecules. The dipole moments of compounds 3 were also calculated and listed in Table 3.

4. Conclusions

In this study, the new coumarins have been successively synthesized and characterized by using various spectroscopic

FIGURE 3: The lowest unoccupied molecular orbital (LUMO) of compound **3**.TABLE 2: HOMO and LUMO energies of **3**.

Compound	HOMO	LUMO	ΔE
3	−8.992 eV	−3.573 eV	−4.390

TABLE 3: The dipole moments (Debye) of **3**.

Compound	x-component	y-component	z-component	Magnitude
3	−0.984845	3.875375	−1.511631	4.274749

methods. The synthesized compound **3** was studied theoretically, and the atomic charges, heat of formation, and stereochemistry were estimated, and it was found that compound **3** is not planar.

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