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ONE-POT SYNTHESIS OF 2-ACYLAMINOBENZIMIDAZOLES FROM THE REACTION BETWEEN TRICHLOROACETYL ISOCYANATE AND 1,2-PHENYLENEDIAMINE DERIVATIVES AND THEORETICAL STUDY OF STRUCTURE AND PROPERTIES OF SYNTHESIZED 2-ACYLAMINOBENZIMIDAZOLES

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ABSTRACT

The two-component reaction of 1,2-phenylenediamine derivatives and trichloroacetyl isocyanate proceeded smoothly and cleanly at room temperature and *N*-(1*H*-benzimidazol-2-yl) acetamide derivatives afforded in excellent yields and no side reactions were observed. The structures of the products were confirmed by IR, ^{1}H NMR, ^{13}C NMR, and elemental analysis.

Also, in this investigation, the structural, electronic properties, IR, ^{13}C and ^{1}H NMR parameters of synthesized molecules were computed in gas in the M062X/6-311G(d,p) level of theory. ^{1}H and ^{13}C NMR chemical shifts were evaluated by employing of the gauge-invariant atomic orbital (GIAO) method. NBO analysis was exploited to examining of the atomic charges and their second order stabilization energy within these molecules.

Keywords: two-component reaction; 1,2-phenylenediamine; trichloroacetyl isocyanate; heterocycle; benzimidazol; GIAO method; NBO analysis.

INTRODUCTION

Benzimidazoles are extensively used structural motifs in medicinal chemistry and can be found in a number of biologically active molecules [1-6]. They are important chemical classes as a result of their significant biological activities against several viruses such as HIV, influenza, herpes (HSV-1), and Epstein-Barr [7-9]. Several compounds from this class have been used as antitumor [1], antibacterial [2], antiviral [10, 11] and antiproliferative agents [12, 13]. Therefore, the synthesis of benzimidazole derivatives has attracted considerable interest. Several synthetic methodologies have been reported in the literature for the synthesis of these compounds. Most involve formation of thioureas using isothiocyanates followed by cyclodesulfurization using desulfurizing agents such as mercury(II) oxide [14], mercury(II) chloride [15], copper(I) chloride [16], methyl iodide [17], tosyl chloride [18], dicyclohexylcarbodiimide (DCC) and PS-carbodiimide [19]. Most of the above reagents are either expensive or highly toxic in nature and commonly require cumbersome work-up and purification procedures. Other methods are cyclization of aminothioureas that often gives poor yield during the reduction of nitro group, S_NAr reaction of chlorobenzimidazole that requires elevated temperature and usually yields selfarylation products [20], and an one-pot procedure by using isoselenocyanates or isothiocyanates with various substituted diamines [21-25].

We were particularly interested in the synthesis of benzimidazoles via a method suitable for large scale preparations as well as not requiring toxic starting materials and reagent. In connection with our interest in the synthesis of heterocycles [26-35], the synthesis of *N*-(1*H*-benzimidazol-2-yl) acetamide derivatives *v*ia a two-component reaction of 1,2-phenylenediamine derivatives and trichloroacetyl isocyanate, in high yields and fairly mild reaction condition, is reported herein (Scheme 1). Also, in this work, we perform a systematic computational study of electronic structure, and properties synthesized molecules.

EXPERIMENTAL

The starting materials and solvents were obtained from Merck (Germany) and Fluka (Switzerland) and were used without further purification. The melting points were measured on an electrothermal 9100 apparatus and are uncorrected. The IR spectra were recorded on a Jasco FT-IR 6300 spectrometer. The ^{1}H- and ^{13}C-NMR spectra were measured ($CDCl_3$ and DMSO-d_6 solution) with a Bruker DRX-250 Avance spectrometer at 250.0 and 62.5 MHz, respectively. The elemental analyses were realized using a Heraeus CHN-O-rapid analyzer.

General procedure

To a magnetically stirred solution of trichloroacetyl isocyanate (**2**, 1 mmol) in CH_2Cl_2 (5mL) was added 1,2-phenylenediamine derivatives (**1**, 1mmol) at -10ºC. The mixture was stirred at room temperature for 20 minutes until 2 hours. Then the mixture was filtrated and washed with CH_2Cl_2, and the products **3a-c and**, **4b-c** were obtained.

***N*-(1*H*-benzimidazol-2-yl)-2,2,2-trichloroacetamide (3a):** White crystals, yield: 100%; m.p. 219.0 - 220.7 ºC (dec); Anal. Calcd. for $C_9H_6Cl_3N_3O$: C, 38.81; H, 2.17; N, 15.09. Found: C, 38.89; H, 2.13; N, 15.14; IR (KBr, cm^{-1}): 3421 (NH), 3154 (ArH), 1712 (C=O), 1605 (C=N), 759 (C-Cl); ^{1}H-NMR (250 MHz, DMSO-d_6, δ / ppm): 7.26 (2H, *dd*, J_1 = 5.7, J_2 = 3.5, aromatic CH), 7.65 (2H, *dd*, J_1 = 5.7, J_2 = 3.5, aromatic CH), 9.71 (1H, *s*, NHCOCCl$_3$), 11.66 (1H, *s*, NH of benzimidazole ring); ^{13}C- NMR (62.5 MHz, DMSO-d_6, δ / ppm): 92.43 (CCl_3), 125.41, 126.50 and 130.85 (aromatic carbons), 150.52 (C=N), 161.65 (C=O).

2,2,2-trichloro-*N*-(5-chloro-1*H*-benzimidazol-2-yl) acetamide (3b) and 2,2,2-trichloro-*N*-(6-chloro-1*H*-benzimidazol-2-yl) acetamide (4b): Brown crystals, yield: 85%; m.p. 207.1- 208.8 ºC; Anal. Calcd. for $C_9H_5Cl_4N_3O$: C, 34.54; H, 1.61; N, 13.43. Found: C, 34.61; H, 1.56; N, 13.50; IR (KBr, cm^{-1}): 3423 (NH), 3155 (ArH), 1707 (C=O), 1601 (C=N), 766 (C-Cl); ^{1}H-NMR (250 MHz, DMSO-d_6, δ / ppm): 7.28 (1H, *d*, $^3J_{HH}$ = 8.5 Hz, aromatic CH), 7.61 (1H, *d*, $^3J_{HH}$ = 8.5 Hz, aromatic CH), 7.81 (1H, *s*, aromatic CH), 9.70 and 9.85 (2H, 2*s*, 2 NHCOCCl$_3$), 11.54 (1H, *bs*, NH of benzimidazole ring); ^{13}C- NMR (62.5 MHz, DMSO-d_6, δ / ppm): 92.54 and 92.60 (2CCl_3), 124.16, 125.79, 127.33, 129.20, 130.32 and 132.84 (aromatic carbons), 150.69 and 150.81 (2C=N), 161.61 and 161.84 (2C=O).

2,2,2-trichloro-*N*-(5-methyl-1*H*-benzimidazol-2-yl) acetamide (3c) and 2,2,2-trichloro-*N*-(6-methyl-1*H*-benzimidazol-2-yl) acetamide (4c): White crystals, yield: 88%; m.p. 209.5 - 211 ºC (dec); Anal. Calcd. for $C_{10}H_8Cl_3N_3O$: C, 41.06; H, 2.76; N, 14.36. Found: C, 40.99; H, 2.78; N, 14.42; IR (KBr, cm^{-1}): 3417 (NH), 3150 (ArH), 1709 (C=O), 1601 (C=N), 759 (C-Cl); ^{1}H-NMR (250 MHz, DMSO-d_6, δ / ppm): 2.29 (3H, *s*, CH_3), 7.03 (1H, *d*, $^3J_{HH}$ = 8.4 Hz, aromatic CH), 7.46 (1H, *d*, $^3J_{HH}$ = 8.4 Hz, aromatic CH), 7.48 (1H, *s*, aromatic CH), 9.59 and 9.69 (2H, 2*s*, 2NHCOCCl$_3$), 11.57 (1H, *bs*, NH of benzimidazole ring); ^{13}C- NMR (62.5 MHz, DMSO-d_6, δ / ppm): 21.09 (CH_3), 92.77 and 92.81 (2CCl_3), 125.31, 125.53, 126.75, 127.95, 131.05 and 136.01 (aromatic carbons), 150.95 and 151.03 (2C=N), 161.75 and 161.93 (2C=O).

Computational Methods

The Gaussian 09 suite of program applied to carried out all calculations [36]. The standard 6-311G(d,p) basis set [37] and the hybrid functional of Truhlar and Zhao (M062X) [38] are used for optimization of molecule.

A vibrational analysis is carried out after optimization of at each stationary point found. The result of this analysis, confirm its identity as an energy minimum.

The population analysis is performed by the natural bond orbital method [39] using NBO program [40] under Gaussian 2009 program package in the M062X/6-311G(d,p) level of theory.

NMR calculations are calculated using the Gauge independent atomic orbital (GIAO) [41] method at the M062X /6-311G(d,p) level of theory.

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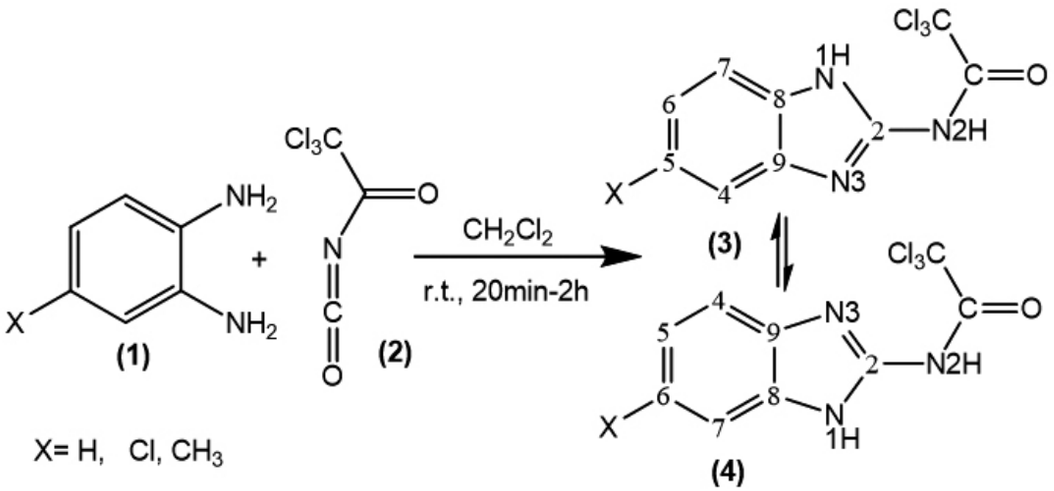
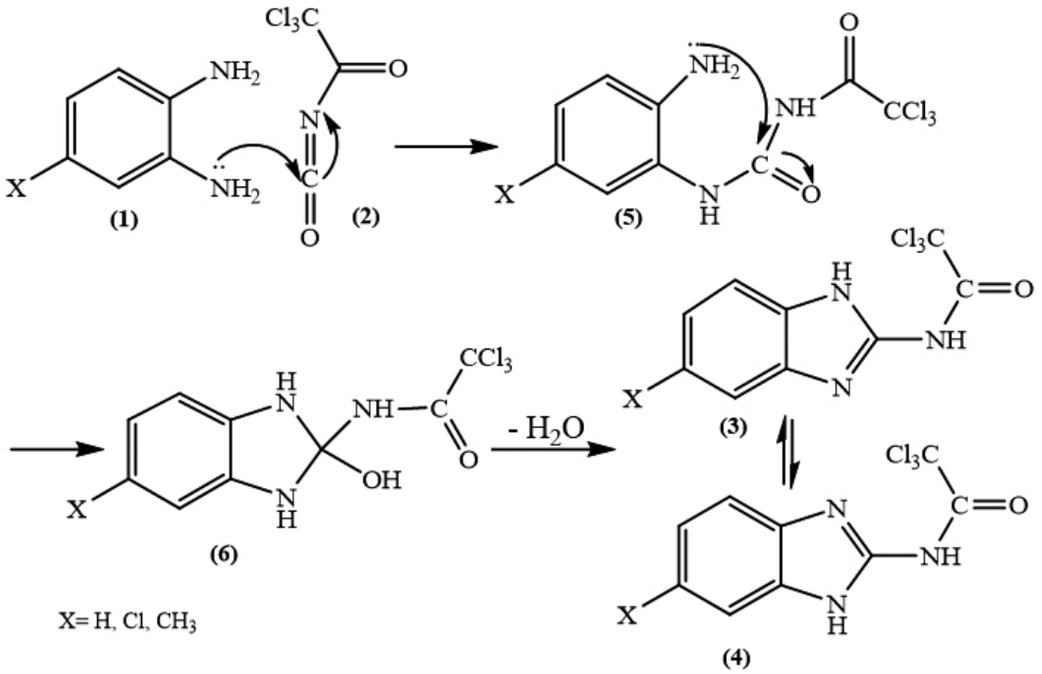
RESULTS AND DISCUSSION

The two-component reactions of 1,2-phenylenediamine derivatives **(1)** and trichloroacetyl isocyanate **(2)** led to *N*-(1*H*-benzimidazol-2-yl) acetamide derivatives **(3)** and **(4)** in high yields, and fairly mild reaction conditions (Scheme 1). A mechanistic rationalization for this reaction was provided in Scheme 2. The structures of the products were deduced from their ^{1}H-NMR, ^{13}C-NMR, IR spectra, and elemental analysis.



Scheme 1. Two component reaction of trichloroacetyl isocyanate and 1,2- phenylenediamine derivatives.



Scheme 2. A proposed mechanism for the formation of (3) and (4) molecules.

Energetic aspects.

The absolute energy and relative energy values of **(3)** and **(4)** isomers are tabulated in Table 1. Theoretical studies show that stability of (**3**)-isomer is more than (**4**)-isomer (Table 1). On the other hand, relative energies values show that **(3)** and **(4)** isomers are isoenergetic. The energy difference between **(3)** and **(4)** isomers in the presence of X=Cl is more than X=Me. Resonance structure of **(3)** and **(4)** are possible in the presence of X=Cl (Figure 1). Therefore, the minor difference between isomers results from contribution of II-form.

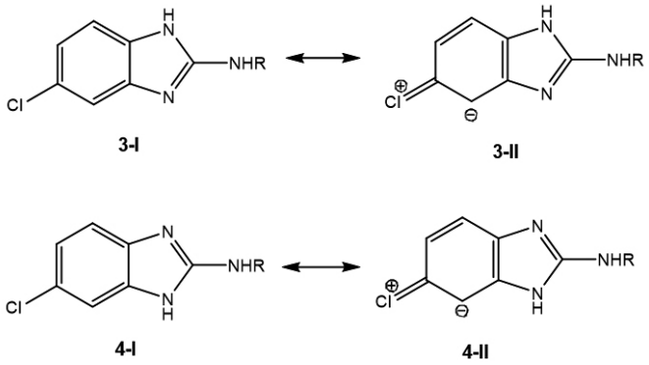


Figure 1. Possible resonance structure of **(3)** and **(4)** molecules in the presence of X=Cl.

Polarizabilities

Polarizabilities describe the response of a system in an applied electric field. They determine not only the strength of molecular interactions (such as the long range intermolecular induction, dispersion forces, etc.) as well as the cross sections of different scattering and collision processes, but also the nonlinear optical properties of the system [42].

The isotropic polarizability $<\alpha>$ is calculated as the mean value as given in the following equation:

$$<\alpha>=\frac{(\alpha_{xx}+\alpha_{yy}+\alpha_{zz})}{3}$$

And the polarizability anisotropy invariant is:

$$\Delta\alpha=\left[\frac{(\alpha_{XX}-\alpha_{YY})^2+(\alpha_{YY}-\alpha_{ZZ})^2+(\alpha_{ZZ}-\alpha_{XX})^2}{2}\right]^{\frac{1}{2}}$$

The isotropic and anisotropic polarizability values of **(3)** and **(4)** isomers are calculated (Table 1). These values show that polarizability of **(3)** isomer is more than **(4)** isomer.

Table 1. Absolute energies (Hartree), relative energies (kcal/mol), isotropic and anisotropic polarizability (Bohr^3) of (3) and (4) molecules in the M062x/6-311G(d,p) level of theory.

	E	ΔE	α_{iso}	α_{aniso}
X=H				
3	-1966.5607	0.00	147.66	96.77
4	-1966.5590	1.09	145.86	87.37
X=Cl				
3	-2426.1588	0.00	161.23	125.69
4	-2426.1572	1.02	160.05	118.69
X=Me				
3	-2005.8669	0.00	161.42	110.69
4	-2005.8656	0.86	160.41	105.94

Molecular orbital analysis

The energies of the frontier orbitals (HOMO and LUMO) along with the corresponding HOMO–LUMO energy gaps for **(3)** and **(4)** isomers are given in Table 2.

As seen shown in Table 2, HOMO energy of **(4)** isomer is lower than **(3)** isomer. But, LUMO energy of **(3)** isomer is lower than **(4)** isomer. These values show that stabilities of HOMO and LUMO increase in the presence of X=Cl, and decrease in the presence X=Me for two isomers. Also, HOMO-LUMO gap and hardness of **(4)** isomer is more than **(3)** isomer.

Figure 2 presents the plots of HOMO and LUMO **(3)** and **(4)** isomers. Figure 2 displays the HOMO are distributed on six and five membered rings. On the other hand, LUMO has less distribution on these rings. HOMO and LUMO of two isomers are π-bonding.

The values of chemical potential and electrophilicity in **(3)** and **(4)** isomers are listed in Table 2. According to the calculation results, the chemical potential values decrease in the presence of X=Cl, and increase in X=Me. On the other hand, **(4)** isomer has more chemical potential rather than (3) isomer.

Electrophilicity values increase in the presence of X=Cl, and decrease in X=Me. On the other hand, **(3)** isomer has more electrophilicity rather than **(4)** isomer.

Table 2. Frontier orbitals energies (a.u), HOMO-LUMO gap (eV)m hardness (eV), chemical potential (eV) and electrophilicity of (3) and (4) molecules in the M062x/6-311G(d,p) level of theory.

	E(HOMO)	E(LUMO)	ΔE	η	μ	ω
X=H						
3	-0.28950	-0.02389	7.23	3.61	-4.26	2.52
4	-0.29470	-0.01502	7.61	3.81	-4.21	2.33
X=Cl						
3	-0.29387	-0.02952	7.19	3.60	-4.40	2.69
4	-0.29525	-0.02325	7.40	3.70	-4.33	2.54
X=Me						
3	-0.28363	-0.02093	7.15	3.57	-4.14	2.40
4	-0.28525	-0.01190	7.44	3.72	-4.04	2.20

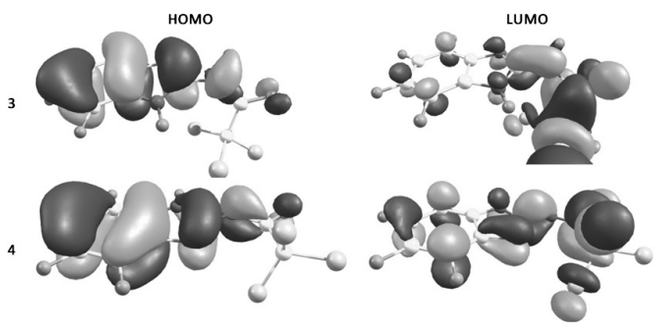


Figure 2. Molecular orbital surfaces of (3) and (4) molecules.

Thermodynamic parameters

A mechanistic rationalization for this reaction is provided in Scheme 2 and thermochemical analysis is studied for this reaction. The values of ΔH, ΔG and ΔS are reported in Table 3 in which the individual terms are referred to a temperature of 298 K. The reactions can be considered as:

(1) + (2) → (3) + H_2O reaction 1
(1) + (2) → (4) + H_2O reaction 2

Table 3. Thermochemical parameters of (a) (3) , (4) molecules and (b) (1) and (2) reactions in the M062x/6-311G(d,p) level of theory.

(a)

	G(Hartree)	H(Hartree)	S(kcal/mol.K)
X=H			
1	-342.7939	-342.7543	83.32
2	-1700.0585	-1700.0120	97.95
3	-1966.4574	-1966.4000	120.81
4	-1966.4566	-1966.3983	122.67
X=Cl			
1	-802.4093	-802.3675	87.97
3	-2426.0677	-2426.0067	128.37
4	-2426.0662	-2426.0047	129.41
X=Me			
1	-382.0740	-382.0307	91.14
3	-2005.7377	-2005.6767	128.36
4	-2005.7381	-2005.6755	131.85

(b)

	ΔG(kcal/mol)	ΔH(kcal/mol)	ΔS(kcal/mol.K)
X=H			
Reaction 1	-8.36407	-12.9512	-0.01539
Reaction 2	-7.88277	-11.9139	-0.01352
X=Cl			
Reaction 1	-5.14872	-8.86671	-0.01247
Reaction 2	-4.22188	-7.63114	-0.01817
X=Me			
Reaction 1	-8.49899	-13.1658	-0.01565
Reaction 2	-8.74434	-12.3701	-0.01216

These values illustrate ΔG and ΔH values are negative. Therefore, free energy values of the **(1)** and **(2)** reactions indicate theses reactions are favorable thermodynamically. It can be seen, reaction (1) is more favorable thermodynamically. Since in this reaction two particles form two, ΔS should be zero value, approximately.

Vibrational analysis

The synthesized molecules possess C1 point group symmetry. Vibrational spectral assignments of the title compound is performed by ab initio M062x method using 6-311G(d,p) basis. Selected vibration wavenumbers of (3) and (4) isomers are gathered in Table 4. These calculations show that:

CH_3

The stretching vibrations of CH_3 are expected in the range 2900–3050 cm^{-1} [47, 48]. The bands at 3061.2 (**3Me**) and 3057.1375 (**4Me**) cm^{-1} result from symmetric stretching in which three of the C-H bonds expand and contract in-phase.

C=O

The band at 1872.2 (**3H**), 1873.3 (**3Me**), 1878.0 (**3Cl**), 1874.7 (**4H**), 1872.2 (**4Me**), and 1870.4 (**4Cl**) cm^{-1} attribute to stretching of C=O bond. The experimental value of this stretch places at 1712 (X=H), 1709 (X=Me), and 1707 (X=Cl) cm^{-1}.

N-H

The bands at 3688.2 (**3H**), 3695.8 (**3Me**), 3689.5 (**3Cl**), 3693.4 (**4H**), 3195.5 (**4Me**), and 3688.6 (**4Cl**) cm^{-1} are attributed to stretching of N1-H1 bond. The bands at 3594.3 (**3H**), 3584.6 (**3Me**), 3580.4 (**3Cl**), 3578.4 (**4H**), 3191.3 (**4Me**), and 3598.5 (**4Cl**) cm^{-1} are resulted from stretching of N2-H2 bond.

Table 4. Selected vibration wavenumbers (cm^{-1}) of (3) and (4) molecules in the M062x/6-311G(d,p) level of theory.

molecule	υ(NH)	υ(ArH)	υ(C=O)	υ(C=N)
3H	N2H2: 3594.30 N1H1:3688.1753	3182.7530 3207.9523 3210.3673 3223.4445	1872.1957	1631.8526
3Cl	N2H2: 3580.4284 N1H1: 3689.5169	3194.5073 3207.8545 3216.7806	1878.0478	1629.4555
3Me	N2H2: 3584.5844 N1H1: 3695.7840	3150.5328 3167.8398 3202.0035 3206.1387	1873.3016	1630.2663
4H	N2H2: 3578.4417 N1H1: 3693.4412	3188.2176 3198.7879 3206.0878 3216.5720	1874.7319	1633.6889
4Cl	N2H2: 3598.4793 N1H1: 3688.5654	3226.7728 3234.6812 3239.5881	1872.1699	1634.4802
4Me	N2H2: 3191.2977 N1H1: 3195.5407	3145.2875 3191.2977 3195.5407 3222.3126	1870.4125	1635.1629

Experimental and theoretical ^{1}H-NMR, ^{13}C-NMR

The theoretical and experimental ^{1}H and ^{13}C NMR chemical shifts of the synthesized compounds are gathered in Table 5. Relative chemical shifts are estimated by using the corresponding TMS shielding calculated at the same theoretical level as the reference.

The experimental ^{1}H-NMR spectrum of **3a** exhibits distinct signals at δ_H 7.26 and 7.65 ppm (4H, 2dd) arising from the aromatic CH groups and at δ_H 9.71 and 11.66 ppm (2H, 2s) of the 2NH groups. These values show the most chemical shift value for H1 atom. Therefore, there is lowest the electronic charge densities around H1 atom. The chemical shifts of H4 and H7 atoms are higher than H5 and H6 atoms. It means that the electronic charge densities around the H4 and H7 are lower than H5 and H6.

The ^{13}C-NMR spectrum of **3a** reveals six distinct resonances arising from the CCl_3 group (δc 92.43 ppm), aromatic carbons (δc 125.41, 126.50 and 130.85 ppm), C=N (δc 150.52 ppm), and C=O (δc 161.65 ppm). The **3b**, **4b** and **3c**, **4c** compounds are tautomers, thus there are two NH signals for $NHCOCCl_3$ in the ^{1}H-NMR spectra of **3a, 4b** and **3c, 4c** compounds. In addition, there are two C=O, two CCl_3, and two C=N signals in the ^{13}C-NMR spectra **3a, 4b** and **3c, 4c** compounds.

NBO analysis

The most important interaction between ‘‘filled” (donor) Lewis type NBOs and ‘‘empty” (acceptor) non-Lewis NBOs is reported in Table 6. The results of NBO analysis collected in Table 6 show that the LP(N1) participates as donor and the BD(2)*(C2-N3) antibond as acceptor [LP (1) N1 →BD*(2) C2 - N3] in **(3)** isomer. In the **(4)** isomer, LP(N7) participates as donor and the BD(2)*(C-O) antibond as acceptor [LP (1) N7 → BD*(2) C – O].

The natural population analysis (NPA) is evaluated in terms of natural atomic orbital occupancies. Table 7 presents the molecular charge distribution on the skeletal atoms for studied molecules. Usually, it is noted that the strong negative and positive partial charges are placed on N2 and $C_{carbonyl}$ skeletal atoms in **(3)** and **(4)** molecules.

Table 5. ^{1}H and ^{13}C NMR chemical shifts (ppm) of (3) and (4) molecules in the M062x/6-311G(d,p) level of theory respect to TMS.

^{1}H NMR

		H1	H4	H5	H6	H7
3						
X=H	exp	11.66	7.65	7.26	7.26	7.65
	theo	8.56	8.36	7.84	7.91	7.86
X=Cl	exp	9.70	7.81	-	7.28	7.61
	theo	8.47	8.88	-	7.71	7.69
X=Me	exp	9.59	7.48	-	7.03	7.46
	theo	8.37	8.13	-	7.75	7.76
4						
X=H	exp	11.66	7.65	7.26	7.26	7.65
	theo	7.00	8.36	7.84	7.76	7.63
X=Cl	exp	9.85	7.61	7.28	-	7.81
	theo	7.73	8.29	7.69	-	7.67
X=Me	exp	11.57	7.46	7.03	-	7.48
	theo	7.69	8.35	7.76	-	7.57

^{13} C NMR

		C2	C4	C5	C6	C7	C8	C9
3								
X=H	exp	150.52	125.41	126.50	126.50	125.41	130.85	130.85
	theo	150.62	137.02	137.96	138.93	122.76	144.72	155.28
X=Cl	exp	150.69	124.16	129.20	127.33	125.79	130.32	132.84
	theo	152.09	136.50	150.32	139.53	123.13	142.76	155.74
X=Me	exp	151.03	125.53	127.95	126.75	125.31	131.05	136.01
	theo	150.29	136.71	148.25	140.76	122.64	142.43	155.43
4								
X=H	exp	150.52	125.41	126.50	126.50	125.41	130.85	130.85
	theo	148.99	136.01	137.68	137.85	120.78	143.38	156.00
X=Cl	exp	150.81	125.79	127.33	129.20	124.16	132.84	130.32
	theo	150.72	138.31	138.56	152.60	122.35	145.26	153.63
X=Me	exp	150.95	125.31	126.75	127.95	125.53	136.01	131.05
	theo	148.87	137.74	139.13	151.55	122.31	145.19	153.39

Table 6. Second order delocalization energies (E(2), kcal/mol) for (3) and (4) molecules in the M062x/6-311G(d,p) level of theory.

molecules	Donor →acceptor	E(2)	E(j)-E(i), a.u	F(i,j), a.u
3H	LP (1) N 1 →BD*(2) C2 - N3	66.00	0.37	0.139
3Cl	LP (1) N1 →BD*(2) C2 - N3	66.49	0.36	0.140
3Me	LP (1) N1 → BD*(2) C2 - N3	66.54	0.36	0.140
4H	LP (1) N 7 → BD*(2) C - O	63.44	0.39	0.142
4Cl	LP (1) N 7 → BD*(2) C - O	67.56	0.39	0.146
4Me	LP (1) N7 → BD*(2) C - O	68.24	0.39	0.147

Table 7. NBO atomic charge for (3) and (4) molecules in the M062x/6-311G(d,p) level of theory.

	C2	C4	C5	C6	C7	C8	C9	N1	N2	N3	$C_{carbonyl}$	O
3												
X=H	0.5827	-0.1960	-0.2190	-0.1984	-0.2401	0.1306	0.1145	-0.5849	-0.6824	-0.5055	0.6839	-0.5521
X=Cl	0.5878	-0.2263	-0.0368	-0.2268	-0.2206	0.1259	0.1304	-0.5815	-0.6853	-0.5031	0.6851	-0.5481
X=Me	0.5791	-0.2069	-0.0325	-0.2019	-0.2294	0.1232	0.1237	-0.5820	-0.6847	-0.5046	0.6854	-0.5534
4												
X=H	0.6145	-0.1996	-0.2186	-0.2022	-0.2453	0.1309	0.1256	-0.5960	-0.6567	-0.5372	0.6833	-0.5576
X=Cl	0.5800	-0.1721	-0.2474	-0.0175	-0.2712	0.1474	0.1085	-0.5766	-0.6943	-0.4855	0.6897	-0.5588
X=Me	0.5709	-0.1806	-0.2251	-0.0081	-0.2529	0.1431	0.1046	-0.5774	-0.6927	-0.4877	0.6898	-0.5623

CONCLUSION

The reported method offers a mild, simple, and efficient route for the preparation of *N*-(1*H*-benzimidazol-2-yl) acetamide derivatives *via* a two-component reaction of 1,2-phenylenediamine derivatives and trichloroacetyl isocyanate, in high yields and fairly mild reaction conditions. Also, theoretical studies in the M062X/6-311G(d,p) level of theory show that (3) and (4) isomers are isoenergetic. The experimental values of the vibrational frequencies indicate a good correlation with calculated results. Molecular orbital analysis indicated frontier orbitasl are π-bonding. NBO analysis reveals maximum stabilization energy for the interaction LP (1) N1 →BD*(2) C2 - N3 in (3) isomer and LP (1) N7 → BD*(2) C – O in the (4) isomer.

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