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DIELS-ALDER CYCLOADDITION : SYNTHESIS AND REACTIONS OF N-ARYL-1, 2-(DICARPOXY IMIDE)-3-BENZOYL-6-PHENYL-4-CYCLOHEXENES

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ABSTRACT

The N-aryl-1,2-(dicarpoxy imide)-3-benzoyl-6-phenyl-4-cyclohexenes 3a-f were synthesized by Diels-Alder cycloaddition of 1,5-diphenyl-2,4-pentadiene-1-one 1 with N-arylmaleimides 2a-f. Some of the synthesized compounds were subjected to reduction, hydrolysis and dehydrogenation to give compounds 4a-c, 5a-c and 6a–c respectively. The structures of the new synthesized compounds were established from their elemental analyses and spectral data.

Keywords: *Synthesis; Diels-Alder reaction, N-Arylmaleimides and N-Arylphthalimides.*

INTRODUCTION

Diels-Alder (DA) reaction is a [4 + 2]-cycloaddition (Corral, et. al., 1997 ; Kumar and Balasubrahmanyam, 1997; Padwa, et. al. 1997 ; Thiemann, et. al., 1997 and Zhang and Trudell, 1996) i.e. an addition that involves a diene (with 4 centers involved in the reaction) and a second species with a double bond (2 centers involved in the reaction), (Sauer et. al. 1980) usually referred to as 'dienophile'. This results in strongly negative entropy of activation, due to the highly ordered activated complex, and in a stereospecific course of the reaction.

The DA reaction is characterized by large negative volumes of reaction and activation, due to the simultaneous formation of two new σ-bonds. It is therefore not surprising that increasing the pressure can speed up the reaction considerably. (Klärner and Breitkopf, 1999 and Jenner, 1999). Sometimes, the volume of activation is even more negative than the reaction volume. (Kiselev, et. al., 2001 and Kiselev, et. al., 2002).

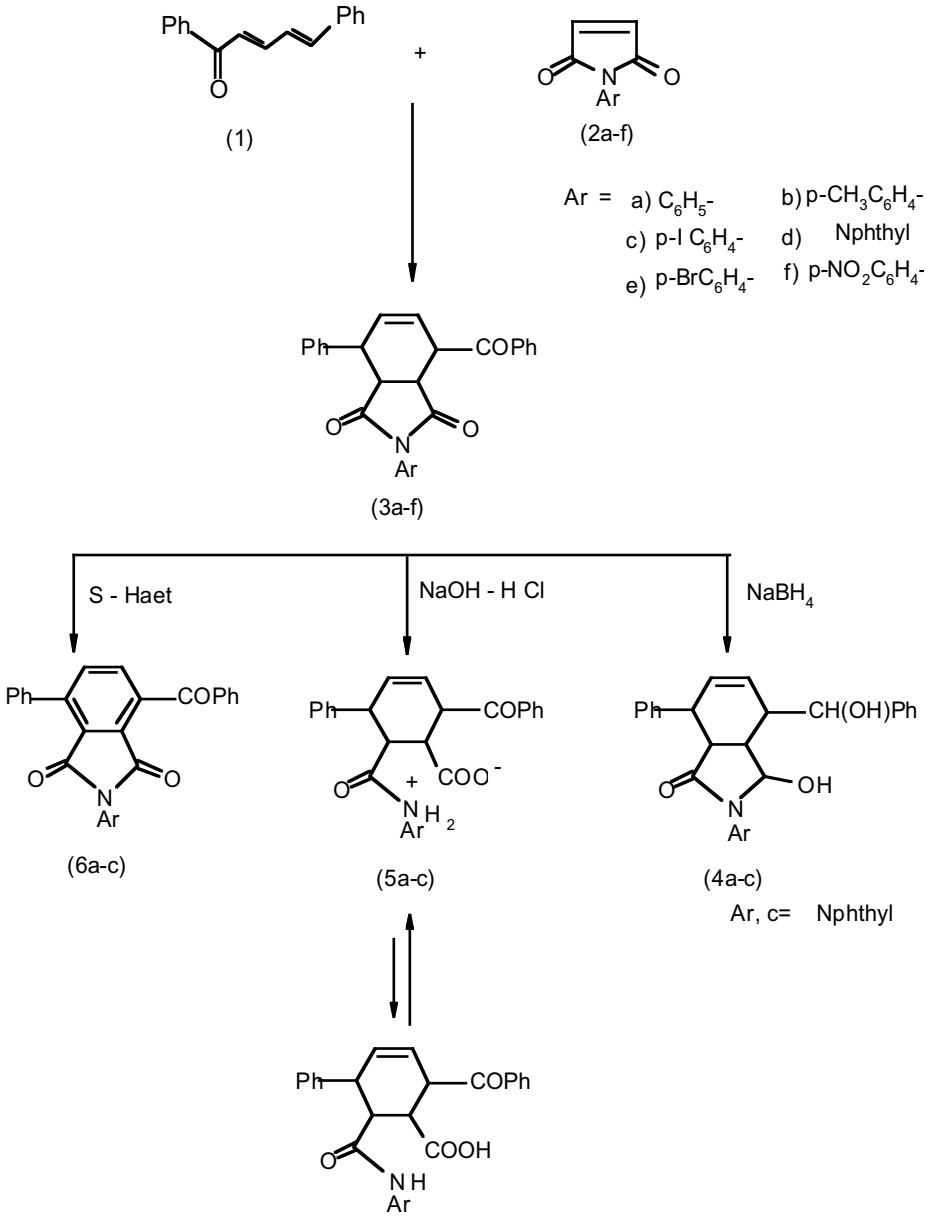
The maleimide moiety can be used as a platform in synthesis as dienophile due to its ability toward DA cycloaddition reactions with dienes. (Al-Shara'ey and Khalifa 2007 ; Baldwin et. al. 1991 ; Ghabrial and Gaber 2003 and Grigg, et. al., 1988).

The present work, focused on the synthesis and reactions of some new heterocyclic compounds from the reaction of 1,5-diphenyl-2,4-pentadiene-1-one 1 as a diene with N-arylmaleimide 2a-f as dienophile by DA cycloaddition reaction.

RESULTS AND DISCUSSION

The starting material 1,5-diphenyl-2,4-

pentadiene-1-one 1 was prepared by condensation reaction between cinnamaldehyde and acetophenone as described in the literature (Cava et. al., 1973 and Kiselev et. al., 2004). The trans, trans configuration of the diene 1 must be stereospecific, for the DA cycloaddition reaction. Thus, the N-aryl-1,2-(dicarboxy imide) - 3 - benzoyl - 6 - phenyl - 4 - cyclohexenes 3a-f were synthesized from reaction of 1 with each of N-arylmaleimides 2a-f (Ar = C_6H_5-, *p*-$CH_3C_6H_4$-, *p*-I C_6H_4-, *p*-$NO_2C_6H_4$-, *p*-BrC_6H_4- and α-naphthyl). The preparation compounds 3a-f constitutes a new and facile procedure for synthesis of the latter compounds (cf. Scheme 1).



The result of elemental analyses for 3a-f was in agreement with their molecular formulas corresponding to the addition of one molecule of 1 to one molecule of each of 2a-f, respectively.

The IR spectra of 3a-f showed the presence of stretching bands for unsaturated and aromatic (=C-H), saturated (C-H), carbonyl groups (C=O), aliphatic and aromatic (C=C) and bending bands for saturated (C-H). Its 1H NMR spectra revealed the presence of signals for the aliphatic and aromatic protons in the proper positions and the mass spectra of compounds 3a and c showed molecular ion peaks at m/z 407 and 533 which are in agreement their formulas (cf. Experimental).

Some of the synthesized compounds were subjected to reduction, hydrolysis and dehydrogenation to give compounds 4a-c, 5a-c and 6a-c, respectively.

The reduction of 3a-c with sodium borohydrid in diethyl ether (Mase et al., 2002) gave 3-hydroxy benzyl-6-phenyl-7-oxo-8-aza-9-hydroxy bicycle [3.4.0]non-4-enes 4a-c. The I.R spectra of 4a-c showed the presence of stretching bands characteristic for hydroxyl groups, aromatic (=C-H), aliphatic (C-H), carbonyl group, aliphatic and aromatic (C=C), and bending bands for aliphatic (C-H) and the ^{1}H NMR spectra displayed signals for aliphatic, hydroxyl and aromatic protons in the proper positions. The mass spectrum of 6a as a typical example for this series showed a peaks at m/z 346 for $[C_{26}H_{20}N]^+$ and 93 for $[C_6H_7N]^+$.

The hydrolysis of 3a-c in basic solution (Knight, 1979) followed by neutralized with aqueous acid gave N-aryl carboxy amide-2-carboxy-3-benzoyl-6-phenyl - 4 - cyclohexenes 5a-c which exists in an equilibrium form with its zwitterionic species. The I.R spectra of 5a-c showed the presence of stretching broad band for zwitterionic species, stretching bands for aliphatic (C-H), carbonyl groups, aliphatic and aromatic (C=C), bonds and bending bands for aliphatic (C-H) and 1H NMR spectra showed appearance of signals for aliphatic, aromatic and amide protons. The mass spectrum of 5a as a typical example of this series showed a peaks at m/z 364 for $[C_{26}H_{22}NO]^+$ and 105 for $[C_7H_5O]^+$.

Dehydrogenation of 3a-c was obtained via fusion reaction with sulfur to produce phthalimides 6a-c. The I.R spectra of 6a-c showed stretching bands for aromatic, (=C-H), carbonyl groups and aromatic (C=C) with the disappearance of the stretching and bending bands for aliphatic (C-H) in 6a, c and 1H NMR spectra showed the appearance of signals for aromatic protons with disappearance of the signals for the aliphatic protons in 6a, c. The mass spectrum of 6a as a typical example of this series, showed a peaks at m/z 256 for $[C_{19}H_{12}O]^+$ and 192 for $[C_{14}H_7O]^+$.

EXPERIMENTAL :

All melting points are uncorrected. The IR spectra in mill (nujol) were recorded on BUCK Model 500 IR. The ^{1}H NMR spectra were recorded on Observe 200 MHz, Gemini-200 and Bruker Wp-80 spectrometers using $CDCl_3$ as a solvent. Chemical shifts (δ) are in ppm relative to internal tetramethylsilane (TMS) as internal standard. Mass spectra were recorded on a Hewlett-Packard-GC-MS type 2988 series using DIP technique at 70 EV. Microanal-

yses were performed at the Micro analytical Center of Cairo University using a Perkin-Elmer 2400. C, H, N elemental analyzer. Thin layer chromatography (TLC) was preformed on aluminum plates coated with 0.25 mm layer of silica gel f_{245} (fluka).

1) General procedure for the synthesis of N-aryl-1,2-(dicarpoxy imide)-3benzoyl-6-phenyl-4-cyclohexene 3a-f.

A mixture of 1,5-diphenyl-2,4-pentadiene-1-one 1 (1.17 g, 0.005 mol) and each of N-arylmaleimides 2a-f (0.005 mol) was heated in an oil bath at 140°C. TLC showed that the reaction was completed after 3 hours. The solid obtained after cooling were recrystallized from ethanol to give 5a-f.

3a: Ar = Phenyl

White crystals, Yield: 73.89%, m.p, 140°C. IR : ν_{max} (cm^{-1}) 3040 (aliphatic and aromatic, =C-H); 2950, 1900 (aliphatic C-H); 1720-1710 (C=O groups); 1620, 1520 (aliphatic and aromatic, C=C) and 1420, 1300(aliphatic C-H). ^1H NMR: 2.9 (1H, d, H6); 3.7-4.8(3H, d, m, aliphatic protons, H_1, H_2, H_3); 6.5-6.8(2H, dd, H_4, H_5); 7.0-8.1(15H, m, aromatic protons). MS: m/z (%): 407(55) [$M^{\cdot+}$] in agreement with $[C_{27}H_{21}NO_3]^{\cdot+}$, 105(75) $[C_7H_5O]^+$. Anal. Calcd. For $C_{27}H_{21}NO_3$: C, 79.60; H, 5.15; N, 3.43 %. Found: C, 79.90; H, 5.28; N, 3.33 %.

3b: Ar = *p*-Methyl phenyl

White crystals, Yield: 78.09 %, m.p 198°C. IR : ν_{max} (cm^{-1}) 3030(aliphatic and aromatic, =C-H); 2920(C-H); 1700 (C=O groups); 1630, 1480(aliphatic and aromatic, C=C) and 1400, 1350(bend. C-H, aliphatic). ^1H NMR: 2.5(3H, s, $-CH_3$); 2.7 (1H, d, H_6); 3.5-4.9(3H, d, m, aliphatic protons, H_1, H_2, H_3); 6.5-6.8(2H, dd, H_4, H_5), 7.0-8.2(14H, m, aromatic protons). Anal. Calcd. For $C_{28}H_{23}NO_3$: C, 79.80; H, 5.46; N, 3.32 %. Found: C, 79.40; H, 5.60; N, 3.91 %.

3c: Ar = *p*-Iodo phenyl

White crystals, Yield: 62.03 %, m.p, 159°C. IR : ν_{max} (cm^{-1}) 3040(aliphatic and aromatic, =C-H); 2900(aliphatic, C-H); 1700(C=O groups); 1600, 1500(aliphatic and aromatic, C=C) and 1400, 1350(bend. C-H, aliphatic). ^1H NMR: 2.6(1H, d, H_6), 3.8-4.8(3H, d, m, aliphatic protons, H_1, H_2, H_3); 6.2-6.6(2H, dd, H_4, H_5); 7.0-8.2(14H, m, aromatic protons). MS: m/z (%): 533[$M^{\cdot+}$] (53), 105(80). Anal. Calcd. For $C_{27}H_{20}INO_3$: C, 60.78; H, 3.75; N, 2.62; I, 23.82 %. Found: C, 60.41; H, 3.33; N, 2.31; I, 23.60 %.

3d: Ar = *p*-Nitro phenyl

White crystals, Yield: 69.91 %, m.p 190°C. IR : ν_{max} (cm^{-1}) 3010(aliphatic and aromatic, =C-H); 2980, 2900(aliphatic, C-H); 1720(C=O groups); 1610, 1500 (aliphatic and aromatic, C=C and NO_2) and 1430, 1350(bend. C-H, aliphatic). 1H NMR: 2.6(1H, d, H_6), 3.8-4.8(3H, d, m, aliphatic protons, H_1, H_2, H_3); 6.2-6.6 (2H, dd, H_4, H_5), 7.0-8.2(14H, m, aromatic protons). Anal. Calcd. For $C_{27}H_{21}N2O_5$: C, 71.68; H, 4.42; N, 6.19 %. Found: C, 71.28; H, 3.96; N, 6.08 %.

3e: Ar = *p*-Bromo phenyl

Yellow crystals, Yield: 81.89 %, m.p 160°C. IR : ν_{max} (cm^{-1}) 3020(aliphatic and aromatic, =C-H); 2970, 2900(aliphatic, C-H); 1700(C=O groups); 1600, 1500(aliphatic and aromatic, C=C) and 1400, 1300(bend. C-H, aliphatic). ^1H NMR: 2.6(1H, d, H6), 3.8-4.8(3H, d, m, aliphatic protons, H_1, H_2, H_3); 6.2-7.0(2H, dd, H_4, H_5), 7.0-8.2(14H, m, aromatic protons). Anal. Calcd. For $C_{27}H_{20}BrNO_3$: C, 66.66; H,

4.11; N, 2.88; Br, 16.44 %. Found: C, 66.73; H, 4.40; N, 2.73 %.

3f: Ar =α-Naphthyl

White crystals, Yield: 83.77 %, m.p 180°C. IR : ν_{max} (cm^{-1}) 3040 (aliphatic and aromatic, =C-H); 2900(aliphatic, C-H); 1690(C=O groups); 1620, 1500(aliphatic and aromatic, C=C) and 1450, 1350(bend. C-H, aliphatic). ^1H NMR: 2.6(1H, d, H_6), 3.8-4.8(3H, d, m, aliphatic protons, H_1, H_2, H_3); 6.4-6.8(2H, dd, H_4, H_5), 7.0-8.2(17H, m, aromatic protons). Anal. Calcd. For $C_{31}H_{23}NO_3$: C, 81.40; H, 5.03; N, 3.06 %. Found: C, 81.02; H, 5.21; N, 3.42 %.

2) General procedure for the synthesis of 3-hydroxybenzyl-6-phenyl-7-oxo-8-aza-8-aryl-9-hydroxy bicyclo [3.4.0] non-4-enes 4a-c.

A mixture of each of 3a-c (0.01 mole) and sodium borohydride (0.024 mole) in dry diethyl ether (50 ml) was stirred. TLC showed that the reaction was completed after 3 hours. The resulting mixtures were decomposed by a dropwise addition of ice water and dilute actice acid then extracted with diethyl ether. The diethyl ether was evaporated and the residue was recrystallized from ethanol as crystals.

4a: Ar = Phenyl

Colorless solid, Yield: 68.35 %, m.p 210°C. IR : ν_{max} (cm^{-1}) 3400(-OH groups); 3020 (aliphatic and aromatic, =C-H); 2950, 2900 (aliphatic, C-H); 1710-1700(C=O group); 1500(aliphatic and aromatic, C=C); 1400, 1320 (bend. C-H aliphatic) and 1000 (for, C-O). ^1H NMR: 2.4(1H, d, H6); 3.6-4.8(7H, s, m, H_1, H_1', H_2, H_3, H_9, aliphatic and hydroxyl protons); 6.4-6.8(2H, dd, H_4, H_5); 7.0-8.0(15H, m, aromatic protons). MS: m/z (%): 346[M^+] (70), 175(70) and 93(76). Anal. Calcd. For $C_{27}H_{25}NO_3$: C, 78.83; H, 6.08; N, 3.40 %. Found: C, 78.67; H, 6.11; N, 3.52 %.

4b: Ar = *p*-Methyl phenyl

Yellow crystals, Yield: 71.49%, m.p 140°C. IR : ν_{max} (cm^{-1}) 3400(–OH groups); 3020(aliphatic and aromatic, =C-H); 2950, 2900(aliphatic, C-H); 1710-1700(C=O group); 1490(aliphatic and aromatic, C=C); 1400, 1300(bend. aliphatic, C-H) and 1000(for, C-O). 1H NMR: 2.2(3H, s, -CH3); 2.5(1H, d, H6); 3.5-4.6(7H, s, m, H_1, H_1', H_2, H_3, H_9, aliphatic and hydroxyl protons); 6.4-6.8(2H, dd, H_4, H_5); 7.0-8.2(14H, m, aromatic protons). Anal. Calcd. For $C_{28}H_{27}NO_3$: Calcd C, 79.05; H, 6.35; N, 3.29 %. Found: C, 79.00; H, 6.41; N, 3.03 %.

4c: Ar = α-Nphthyl

White crystals; yield 81.46 %, m.p 285°C; IR : ν_{max} (cm^{-1}) 3400(-OH groups); 3050 (aliphatic and aromatic, =C-H); 2950(aliphatic, C-H); 1710-1700(C=O group); 1480(for aliphatic and aromatic, C=C); 1400, 1300 (bend. aliphatic, C-H) and 1000 (for, C-O). ^1H NMR: 2.4(1H, d, H_6); 3.6-4.8(7H, s, m, H_1, H_1', H_2, H_3, H_9, aliphatic and hydroxyl protons); 6.4-6.8(2H, dd, H_4, H_5); 7.0-8.0(17H, m, aromatic protons). Anal. Calcd. For $C_{31}H_{27}NO_3$: Calcd C, 80.69; H, 5.85; N, 3.03 %; Found: C, 80.55; H, 5.75; N, 2.95 %.

3) General procedure for the synthesis of 1-(aryl carboxy amide)-2-carboxy-3-benzoyl-6-phenyl-4-cyclohexenes 5a-c.

Solutions of each of 3a-c (0.01 mole) in aqueous sodium hydroxide (30 ml, 0.1 N) were stirred. TLC showed that the reaction was completed after 2.5 hours. The resulting mixtures were neutralized with hydrochloric

acid (0.1) and then extracted with chloroform. The extract was evaporated and the residue was recrystallized from ethanol.

5a: Ar = Phenyl.

White crystals, Yield: 59.57 %, m.p 250°C. IR : ν_{max} (cm^{-1}) 3500 - 2900 (zwitterionic species); 3010(aliphatic and aromatic, =C-H); 2900(aliphatic, C-H); 1720-1710 (C=O groups); 1600, 1480(aliphatic and aromatic, C=C); 1400, 1350 (bend. aliphatic, C-H) and 1000 (for, C-O). 1H NMR : 2.7(1H, d, H_6); 3.6-4.7(3H, d, H_3, and m, H_1, H_2, aliphatic protons); 6.2-6.8 (2H, dd, H_4, H_5); 7.0-7.8 (15H, m, aromatic and amide protons). MS: m/z (%): 364[M^+] (70), 105(80). Anal. Calcd. For $C_{27}H_{23}NO_4$: Calcd C, 76.23; H, 5.41; N, 3.29%. Found : C, 76.33; H, 5.13; N, 3.37%.

5b: Ar = *p*-Methyl phenyl :

Brown crystals, Yield: 64.47 %, m.p 225°C. IR : ν_{max} (cm^{-1}) 3500-2900(zwitterionic species); 3000(aliphatic and aromatic, =C-H); 2900 (aliphatic, C-H); 1720-1710 (C=O groups); 1630, 1490(aliphatic and aromatic, C=C); 1400, 1330(bend. aliphatic, C-H) and 1000(for, C-O). ^{1}H NMR: 2.4(3H, s, -CH_3); 2.7 (1H, d, H_6); 3.6-4.7(3H, d, H_3, and m, H_1, H_2, aliphatic protons); 6.2-6.8(2H, dd, H_4, H_5); 7.0-7.8(14H, m, aromatic and amide protons) Anal. Calcd. For $C_{28}H_{25}NO_4$: Calcd C, 76.53; H, 5.69; N, 3.18%. Found: C, 76.40; H, 5.62; N, 3.28%.

5c: Ar = *p*-Iodo phenyl :

Yellow crystals; Yield: 70.00 %; m.p 220°C. IR : ν_{max} (cm^{-1}) 3500-2900(zwitterionic species); 2920(aliphatic, C-H); 1720-1710(C=O groups); 1620, 1490(aliphatic and aromatic, C=C); 1400, 1350(bend. aliphatic, C-H) and 1000(C-O). ^{1}H NMR: 2.7(1H, d, H6); 3.6-4.6 (3H, d, H_3, and m, H_1, H_2, aliphatic protons); 6.2-6.8(2H, dd, H_4, H_5); 7.0-7.8(14H, m, aromatic and amide protons). Anal. Calcd. For $C_{27}H_{22}INO_4$: Calcd C, 58.80; H, 3.99; N, 2.54; I, 23.04 %. Found: C, 58.80; H, 3.63; N, 2.12; 23.07 %.

4) General procedure for the synthesis of phthalimides 6a-c.

A mixture of each of 3a-c (0.01mole) and sulfur (0.7 g, 0.02 mole) was heated in an oil bath at 180°C till the evolution of H_2S ceased. The solid obtained was recrystallized from 96% ethanol to give 6a-c as white crystals.

6a: Ar = Phenyl :

Yield : 59.57 %, m.p 250°C. IR : ν_{max} (cm^{-1}) 3010(aromatic, C-H); 1710-1700(C=O groups); 1480(aromatic, C=C). 1H NMR: 7.0-8.2(17H, m, aromatic protons). MS: *m/z* (%): 256[M^+] (40), 192(15). Anal. Calcd. For $C_{27}H_{17}NO_3$: Calcd C, 80.39; H, 4.21; N, 3.47 %. Found C, 80.39; H, 4.06; N, 3.11 %.

6b: Ar = *p*-Methyl phenyl :

Yield : 69.28 %, m.p 200°C. IR : ν_{max} (cm^{-1}) 3000-2950(for aliphatic and aromatic, C-H); 1710-1700(C=O groups); 1480(aromatic, C=C) and 1400, 1300(bend. aliphatic, C-H). Anal. Calcd. For $C_{28}H_{19}NO_3$: Calcd C, 80.57; H, 4.55; N, 3.35 %. Found: C, 80.17; H, 4.69; N, 3.35 %

6c: Ar = p-Iodo phenyl :

Yield : 83.45 %; m.p 180°C. IR : ν_{max} (cm^{-1}) 3050(for aromatic, =C-H); 1710-1700 (C=O groups); 1480(aromatic, C=C). ^{1}H NMR: 7.0-8.2(16H, m, aromatic protons). Anal. Calcd. For $C_{27}H_{16}INO_3$: Calcd C, 61.24; H, 3.02; N, 2.64; I, 24.03 %. Found. C, 61.31; H, 3.36; N, 2.45; I, 24.40 %.

Estimation of biological activity :

The synthesized compounds were screened in vitro for their antimicrobial activities against three strains of bacteria (*Staphylococcus aureus, Streptococcus* and *Escherichia coli*) and two fungal species (*fungi niger* and *Aspergillus flavus*) using the filter paper disc method (Küçükbay et. al., 2003 and Pandeya et. al., 2000). The table below exhibits the distinct results which indicated that no to strong activities of the tested compounds toward the bacterial and fungal species which were observed.

Table 1 : The antibacterial activity of some new synthesized compounds.

Com.	Staphaureus aureus	Strep tococcus	Escherichia coli	Fungi niger	Aspergillus flavus
3c	-	-	-	-	-
3e	-	-	-	-	-
3f	-	-	-	-	-
4a	++	+	-	+	++
5a	-	+	-	+	-
5b	++	+	-	++	++

(-) No activity, (+) moderate and (++) strong activity.

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الملخص العربى

إضافـة (ديلـز – الـدر) الحلقية : تخليق وتفاعـلات N – اريل – ١، ٢- (ثنائى كربوكسى اميد) – ٣ – بنزويـل – ٦ – فينيل – ٤ – هكسين حلقى

عبدالله عبدالكريم الشراعى1 فتحى أنور محمد خليفه2 اقبال محمد عبده دعقان1

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فى هذه الدراسة تم تخليق لبعض المركبات الجديدة من التفاعلات الحلقية لمركبات N- اريل – ١، ٢-(ثنائى كربوكسى أميد) – ٣ – بنزويل – ٦ – فينيل – ٤ – هكسين حلقى (3a-f) وذلك عن طريق تفاعـل (ديلـز – الدر) للإضافة الحلقية بين ١، ٥ – ثنائى فينايل – ٢، ٤ – بنتاداين – ١ – اون (١) مع N – اريل مليمايدات (2a-f). ى بعض هذه المركبات المخلقة أخضعت لتفاعلات الاختزال، التحلل المائى وإزالة ذرات ذرات الهيدروچين فأعطت المركبات (4a-c)، (5a-c) و (6a-c) على التوالى.

البناء التركيبى لهذه المركبات المخلقة الجديدة تم تحديده على أساس نتائج التحاليل العنصرى والتحليل الطيفى التى أجريت لها بدقة.

كلمات مفتاحية : تفاعل ديلز – الدر، N- اريل مليميدات و N- اريل فثلميدات.

JOESE 5

DIELS-ALDER CYCLOADDITION : SYNTHESIS AND REACTIONS OF N-ARYL-1, 2-(DICARPOXY IMIDE)-3-BENZOYL-6-PHENYL-4-CYCLOHEXENES

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