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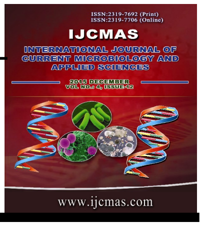

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Original Research Article

Preparation of Dioxadiazole from Aryl Acid Hydrazide with Adipoyl Chloride

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ABSTRACT

Keywords

Dioxadiazole, Aryl Acid, Hydrazide, Adipoyl Chloride, triethylamine

The present work involved preparation of diaromatic heterocyclic compounds The first step involved preparation of 2-substitution butyl benzoate (1-4), second step preparation of 2-substitution benz acid hydrazide (5-8),then preparation of dioxadiazole (9-12) from the reaction of two molecules of 2-substitution benz acid hydrazide with adipoyl chloride in the present triethylamine as catalyst. All the prepared compounds in this work were characterized by melting points with other physical properties, infrared (FTIR) and 1H-NMR, 13C-NMR spectra Screening microbial activity of the preparation dioxadiazole compounds were evolution against two types of (Gram positive).

Introduction

Oxadiazole is a five-number heterocyclic aromatic chemical compound having two carbons, two nitrogen, and one oxygen atoms and two double bonds having general formula C2H2N2O. There are four possible isomers of oxadiazole (1, 2, 3, 4) depending on the position of nitrogen atom in the ring. The1,3,4-oxadiazole undergoes number of reactions including electrophillic substitution, nucleophilic substitution, thermal and photochemical(1-11). Guin and co-workers (12) reported a direct route to both symmetrical and unsymmetrical 2,5-disubstituted-1,3,4- oxadiazoles by means of an imine C-H functionalization of *N*-arylidenearoylhydrazide using Cu()2 as catalyst. Pouliot and co-workers (13) reported the use of diethylamino difluoro sulfinium tetrafluoroborate ([Et2NSF2] BF4), XtalFluor-E, as a new cyclo dehydration agent for the preparation of 1,3,4-oxadiazoles from 1,2-diacyl hydrazine. Oxadiazole rings have been introduced into drug discovery oxadiazoles are having a large impact on multiple drug discovery programs across a variety of disease areas, including diabetes, obesity(14), inflammation (15), cancer (16), and infection (17).

1,3,4-oxadiazole used in medicine and agriculture, in the production of polymers, laser dyes, photographic materials(18).

Experimental

The melting points were determined on a Kofler Block apparatus and are uncorrected.

Infrared spectra were recorded in 400 - 4000 cm-1 region by a Specord FT-IR on SHIMADZU FTIR-8400.

spectrometer using KBr tablet. 1H-NMR,13C-NMR Spectra were measured on ambient Broker DT-400 MHz,Iran.

spectrometer in deuterated DMSO,] The magnetic stirrer and the other necessary laboratory equipment used. All fine chemicals and, reagents were purchased froM,Aldrich chemical Co. U.S.A. and microbial activity were done in the plant biology department laboratories.

General Procedure for Synthesis of Esters (19)(1-4)

In beaker dissolve the different aromatic carboxylic acids with benzene. In a 100 ml round bottomed flask place (0.025mol) of aromatic carboxylic acid and (1.5ml) of butanol and added (3-4 drops) of concentrated sulphuric acid. Fit the flask with a reflux condenser for (2hrs). Cool the mixture at room temperature, physical properties shown in table (1).

General Procedure for Synthesis of Benz Acid Hydrazide (19) (5-8)

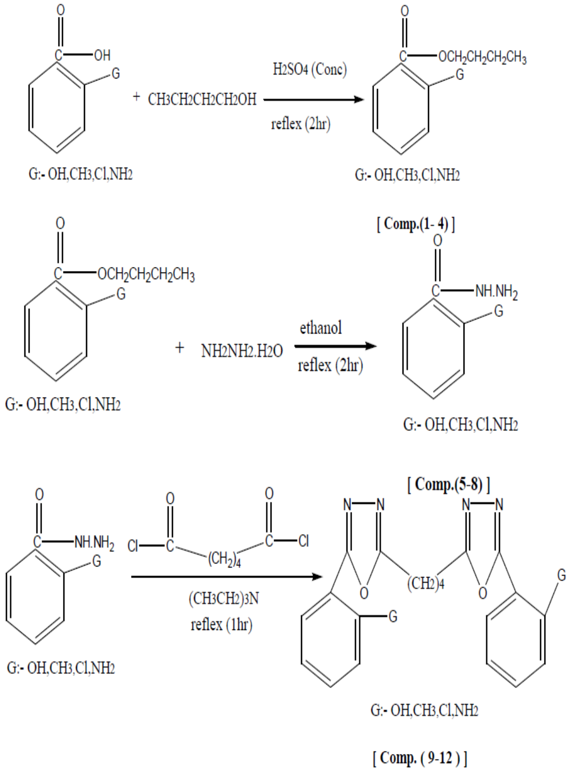
In a 100ml round bottomed flask place (0.012mol) of esters and (0.012mol) of hydrazine hydrate and added (10ml) of ethanol.fit the mixture refluxed for (2hrs). Cool the mixture at room temperature, the color precipitate was filtered, dried and recrystallization with (O-xylene), physical properties shown in table (2).

General Procedure for Synthesis for Dioxadiazole (20) (9-12)

In a 100 ml round bottomed flask dissolve (0.00105mol) of aromatic acid hydrazide (9-12) in benzene. Place (1ml) of trimethyl amine and(0.2ml) of adipoyl chloride to round bottom flask,the mixture was refluxed for(1hrs).cool the mixture at room temperature, physical properties shown in table (3).

Results and Discussion

A four oxadiazole products (9,10,11,12) were obtained by series reactions starting with preparation of ester then preparation of benz acid hydrazaid ended with di oxadiazoles in the chosen solvent, in normal conditions. All products are stable oily compounds, rather insoluble in common solvents, with high melting points. (Table 3).



[Comp.(1- 4)]

[Comp.(5-8)]

[Comp. (9-12)]

Table.1 Physical Properties of Prepared Ester Compounds

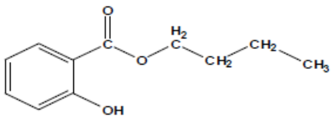
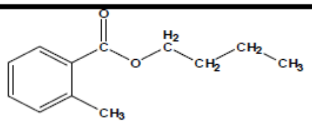
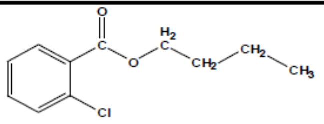
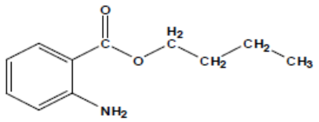
Comp. No.	Compound structure	Color	Melting Points °C	Yield %
1	 butyl 2-hydroxybenzoate	White	145-160	57
2	 butyl 2-methylbenzoate	Off white	100-102	66
3	 butyl 2-chlorobenzoate	Yellow	120-123	66
4	 butyl 2-aminobenzoate	Bijie	137-139	70

Table.2 Physical Properties of Prepared 2-Substituted Benz Acid Hydrazide Compounds

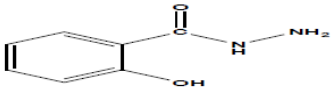
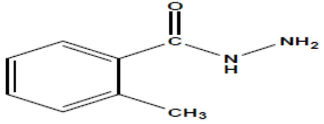
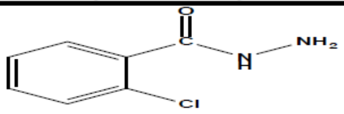
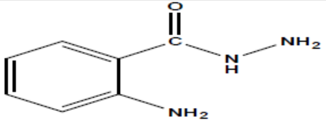
Comp. No.	Compound structure	Color	Melting Points °C	Yield %	Recrystallization Solvent
5	 2-hydroxy benz acid hydrazide	White	150-155	73	O-xylene
6	 2-methyl benz acid hydrazide	Off white	102-104	40	O-xylene
7	 2-chloro benz acid hydrazide	Off white	136-140	53	O-xylene
8	 2-amino benz acid hydrazide	Deep yellow	144-147	83	O-xylene

Table.3 Physical Properties of Prepared Dioxadiazole Compounds

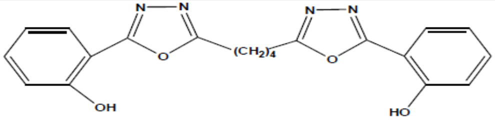
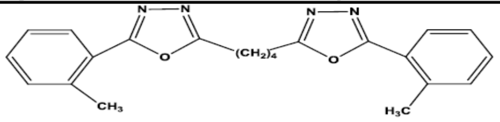
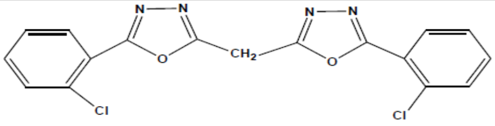
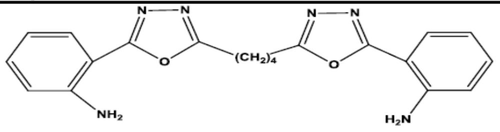
Comp. No.	Compound structure	Color	Melting Points °C	Yield %
9	 [(5,5̀-(butane-1,4-dyl)-2,2̀-(2-hdroxy benzene)]-bis1,3,4-oxadiazole	White	oily	38
10	 [(5,5̀-(butane-1,4-dyl)-2,2̀-(2-methyl benzene)]-bis1,3,4-oxadiazole	Off white	oily	25
11	 [(5,5̀-(butane-1,4-dyl)-2,2̀-(2-chloro benzene)]-bis1,3,4-oxadiazole	Off white	oily	31
12	 [(5,5̀-(butane-1,4-dyl)-2,2̀-(2-amino benzene)]-bis1,3,4-oxadiazole	Deep yellow	oily	18

Table.4 FTIR Spectrum of Dioxadiazole Compound

Comp.	cm^{-1} C—H aromatic	cm^{-1} C—H aliphatic	cm^{-1} C═N	cm^{-1} C—O	cm^{-1} N—N	cm^{-1} C═C aromatic	ν cm^{-1} C—N	ν cm^{-1} C—O—C	ν(other bands) cm^{-1}
9	3010	2848	1627	1139	1485	1589 1573	1380	1255	
10	3062	2923 2860	1685	1159	1456	1575 1558	1382	1274	
11	3056	2983	1682	1172	1434	1591 1558 1473	1386	1244	(Cl)754
12	3028	2921	1680	1141	1460	1583 1490 1465	1371	1232	*(NH2) 3432,3346*

Table.5.Antimicrobial Activity of Oxadiazole Compounds

Samples	E.coil(-ve)	Pseudomonas (-ve)	Bacillus(+ve)	Staphylococcus (+ve)
benzene	-	-	-	-
Oxadia-9	2.6mm	2mm	2mm	2.4mm
Oxadia-10	2.5mm	2.2mm	2,4mm	1.7mm
Oxadia-11	2.5mm	2.4mm	2.4mm	2.5mm
Oxadia-12	1.7mm	10mm	2,1mm	2.2mm

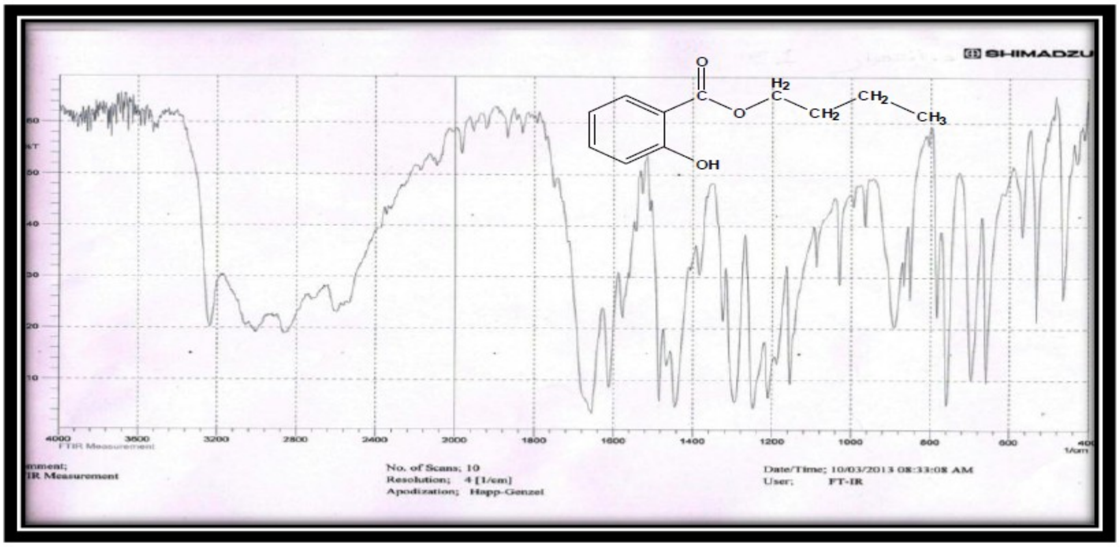


Fig.-1 FTIR Spectrum of compound(**1**)

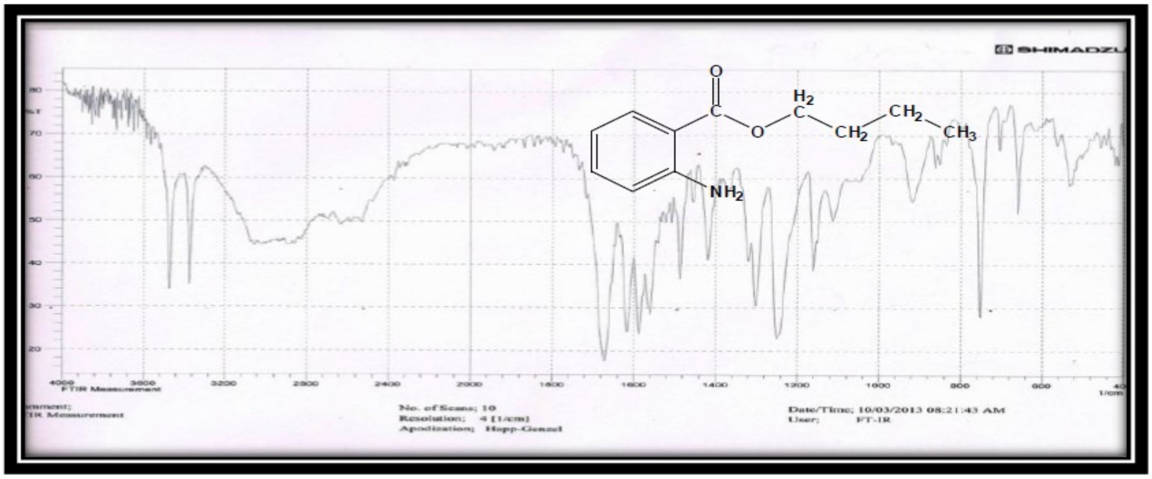


Fig.-2 FTIR Spectrum of compound(**4**)

Fig.3 FTIR Spectrum of Compound(**6**)

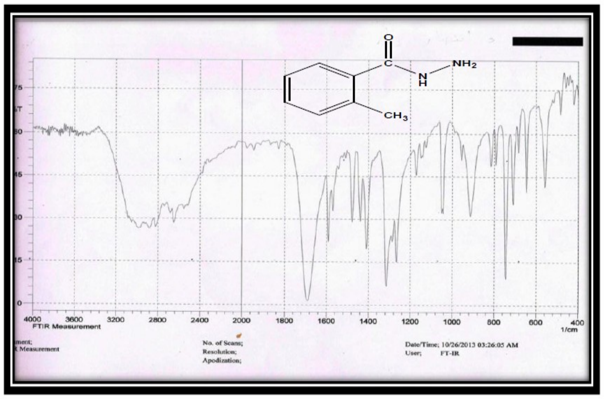


Fig.4 FTIR Spectrum of Compound(**7**)

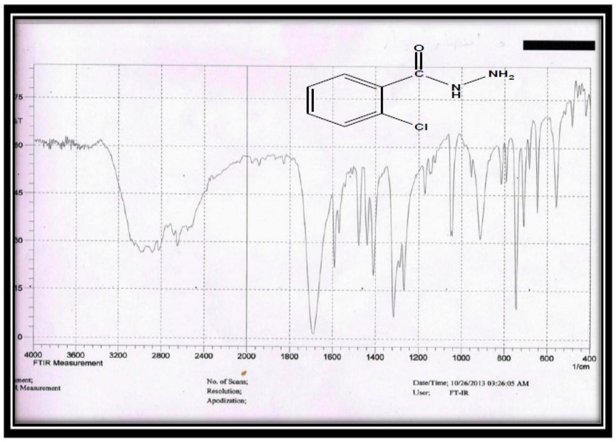


Fig.5 FTIR Spectrum of Compound(**9**)

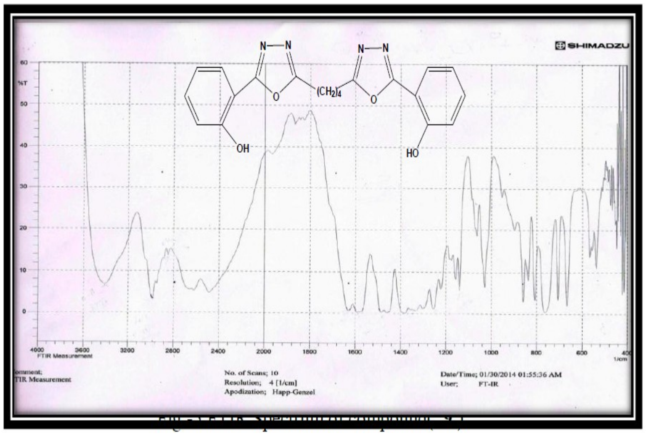


Fig.6 FTIR Spectrum of Compound (**10**)

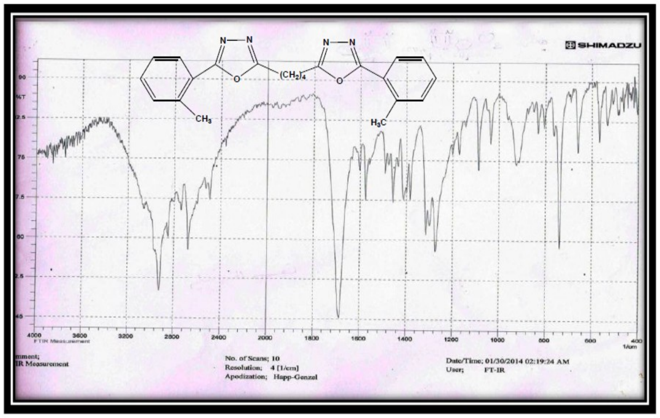


Fig.7 1H -NMR Spectrum of Compound (6)

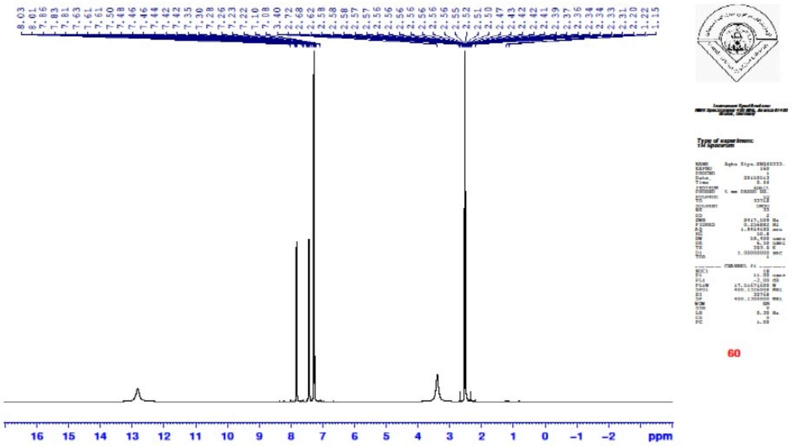


Fig.8 13C-NMR Spectrum of Compound (6)

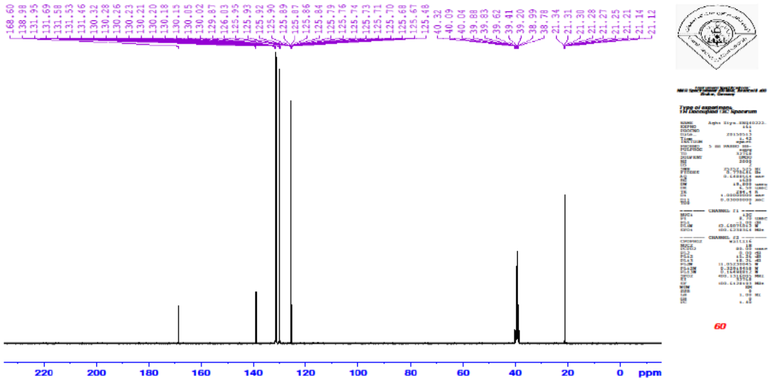


Fig.9 1H -NMR Spectrum of Compound (11)

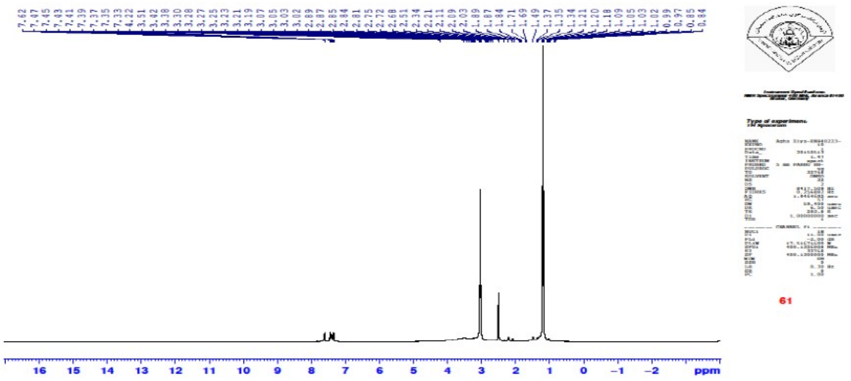
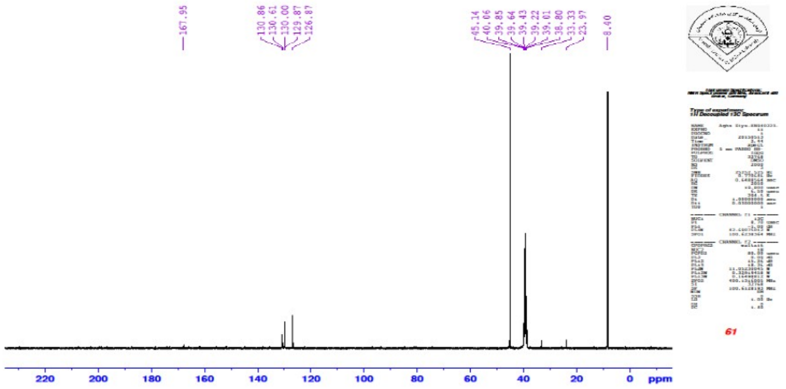


Fig.10 13C-NMR Spectrum of Compound (11)



FTIR Spectra for the compounds ester (1-4) were show appearance absorption bands of (C=O)of ester at(1734,1743,1750,1742 cm-1), Fig(1,2), While FTIR spectra of acid hydrazaid compounds (5-8) showed disappearance of the absorption bands(C=O) ester with appearance absorption bands(C=O) of amide at (1654- 1689)cm-1 as well as distinguish band at the region (3100-3400) cm -1 of (NH) and (NH2), hydrazaide Fig (3,4).While FTIR of compounds (9 – 12) showed disappearance of the absorption bands(C=O) and absorption bands of (NH2) and (NH), hydrazaide with appearance of absorption bands of(C-N),(C=N) and (CH2),Fig (5,6).

1H-NMR spectra of oxa diazoles comp. (9-12), indicate disappearance of proton signals for the(amino group hydrazide) of all compounds)at **δ** (9,5) ppm, and appearance of a protons of methylene (CH2) at (3.4) ppm and protons of (aromatic ring) at(7-8) ppm for all prepared compounds. Fig(7,9).

Antibacterial Activity by Well Diffusion Method

Pertiplates containing (20 ml) muller Hiuton medium were seeded with (24hr) culture of bacterial strains, well were cut and 20micromill liter of the plant extracts (namely aqueous, methanol and chloroform extracts) were added, the plates were then incubated at 370C for (24hr),the antibacterial activity was assayed by measuring the diameter of the inhibition zone formed around the well. chloramphenicol dis was used as appositive control

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