Egypt. J. Chem. **59**, No.3, pp. 285 –303 (2016)

Synthesis, Biological Evaluation of 1,3,4-Oxadiazole, Triazole and Uracil Derivatives from Poly (ethylene terephthalate) Waste

Asmaa M. Fahim, El-Sayed M.A. Yakout[#], Eman A. Ragab[*] , Ahmad M. Farag [*] and Galal A. M. Nawwar

Green Chemistry Department, National Research Center, 33 El Bohouth St. (former El Tahrir st.) , Dokki, P.O. 12622 and [*]Chemistry Department , Faculty of Science, Cairo University, Giza, P.O. 12613, Egypt

POLY(ethylene terephthalate) (PET) was hydrolyzed in aqueous NaOH using solar energy. PET is used as versatile reagent for the synthesis of a variety of heterocyclic compounds. Uracil **(6a, b)**, 1,3,4-oxadiazole (**9, 12a, b)**, 1,3,4-triazole **(10**, **14)**, thiadiazole **(11a, b** and **15)** derivatives were synthesized from PET. The antimicrobial and antioxidant activities of the synthesized compounds were evaluated and showed significant activities.

Keywords: PET, Oxadiazole, Triazole, Thiadiazol, Uracil, Antimicrobial, Antioxidant.

Uracils, 1,3,4-oxadiazoles, 1,3,4-triazoles, and thiadiazoles were associated with diverse pharmacological activities as anti-mycobacterial[(1)] , antidiabetic agents[(2)], kinase and phosphodiestrase inhibitors[(3)], and for their valuable antiangiogenic[(4)], fungicidal, cytotoxic antitubercular[(5)], antimicrobial[(6)] and anthelmintic activities[(7)]. In this context, we report on the synthesis of uracil,1,3,4-oxadiazole, 1,3,4-triazole, and thiadiazole derivatives starting from di-butylterephthalate **(2a)** and 4-(butoxycarbonyl) benzoic acid **(2b)** which are obtained from degradation of PET bottle waste. We found that PET can be readily hydrolyzed by refluxing in sodium hydroxide solution to give terephthalic acid monomer[(8)] in high yield using sun energy. Fisher esterification of terephthalic acid in butanol afforded a mixture of di-butylterephthalate **(2a)** and 4-(butoxycarbonyl) benzoic acid **(2b)** [(9,10)] .

Results and Discussion

Chemistry

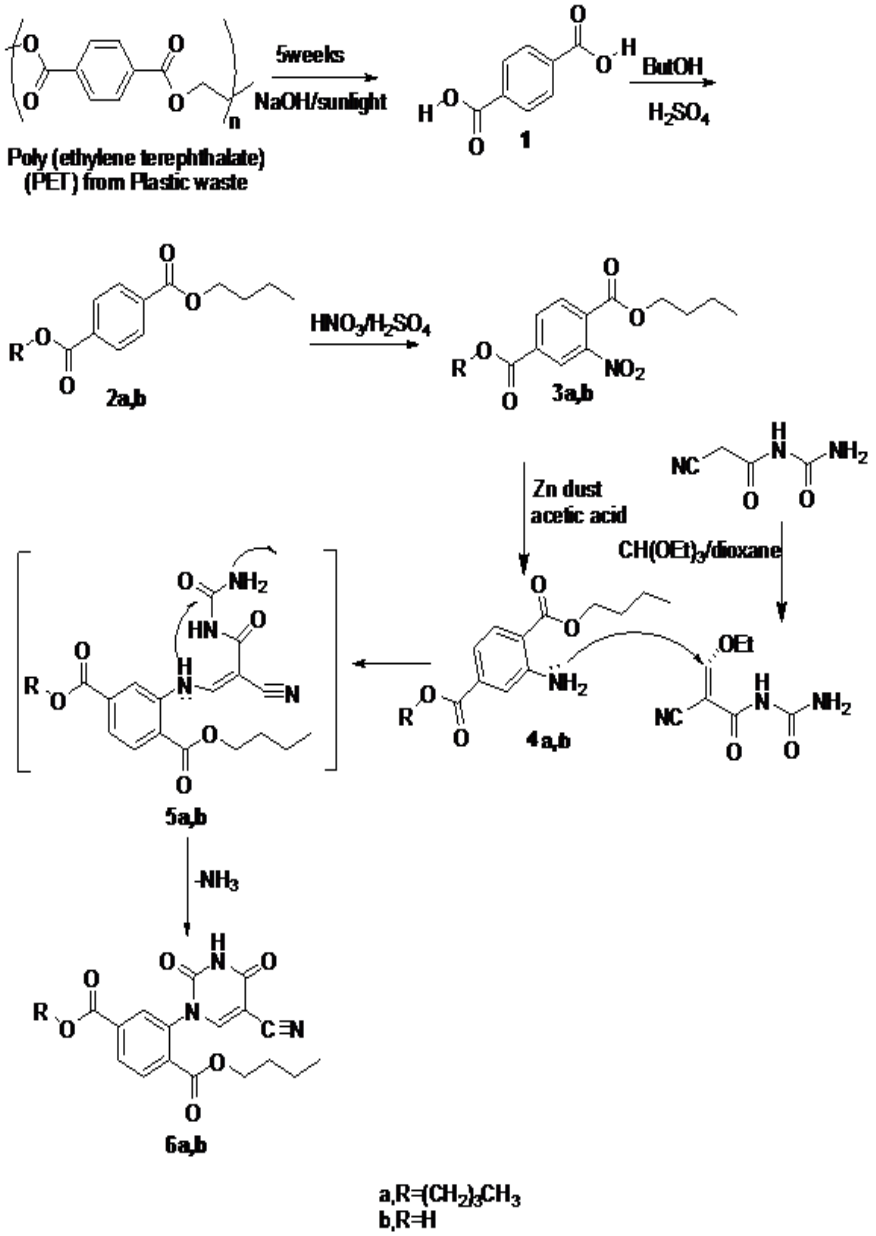
Sun degradation of PET plastic waste using NaOH solution for 5 weeks afforded terephthalic acid (**1**) in a 90% yield, this was followed by esterification with dry butanol containing H_2SO_4[(9-11)] to afford a mixture of di-butyl terephthalate **2a** (80%) and 4-(butoxycarbonyl) benzoic acid **2b** (10%)[(12)] (Scheme 1). Nitration of **2a** and **2b** provided a route to electrophilic substitution to give the corresponding mononitro- derivatives **3a, b** (Scheme 1). The ^{1}H NMR

[#] Corresponding author e-mail: sm_yakout@hotmail.com

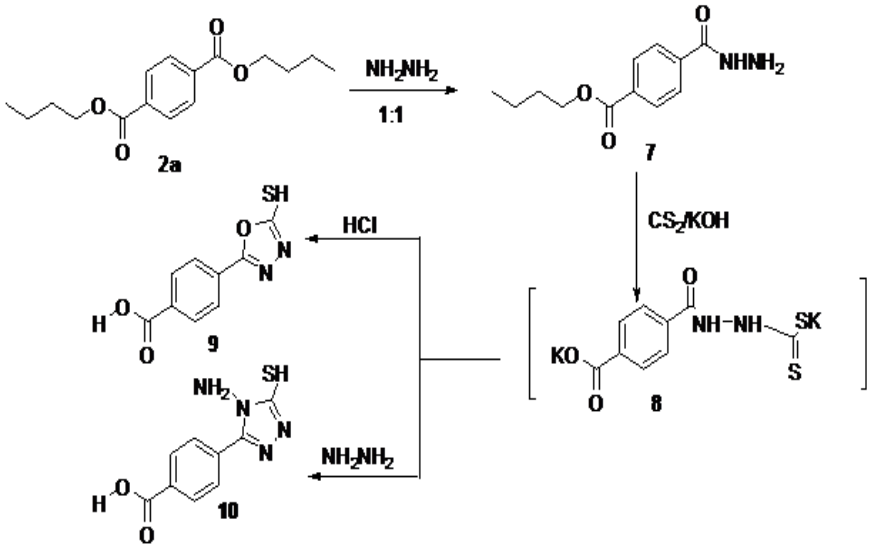
spectrum of the product **3a** showed three aromatic protons in different positions in the region of 8-8.4 ppm. Reduction of compounds **3a, b** using Zn-dust in boiling acetic acid afforded the corresponding mono-amino derivatives **4a, b.** The latter products were characterized by IR and ^{1}H NMR spectra. For example compound **4b** showed strong absorption bands at 3423 and 3380 cm^{-1} in its IR spectrum due to NH_2 group. Its ^{1}H NMR showed D_2O exchangeable signal at 6.64 ppm corresponding to NH_2 group. The mono amino derivatives **4a,b** were allowed to react with equimolar amount of cyanoacetyl urea and triethylorthoformate in refluxing dioxane for ~ 8 hr to produce the corresponding uracil derivatives **6a, b** in good yields (Scheme 1). The active methylene group in cyanoacetyl urea underwent condensation with triethylorthoformate to form the ethoxylidene intermediates (**5a,b**) which were attacked by nitrogen nucleophile through loss of ethanol, followed by intramolecular cyclization with loss of ammonia to afford the uracil derivatives **6a,b** (yield ≈62-75%)[13-15] (Scheme1). The structures of compounds **6a, b** were established on the basis of their spectral data [*cf. Experimental part*].

4-Butyrylbenzohydrazide **(7)** was prepared by the reaction of equimolar amounts of dibutylterephthalte (**2a**) and hydrazine hydrate in ethanol (Scheme 2). 4-Butylrylbenzohydrazide (**7**) was allowed to react with carbon disulfide in ethanol in presence of KOH to give the corresponding thiocarbazinate salt **8.** The latter salt underwent ring closure upon treatment with HCl to give 4-(5-mercapto-4-amino-1,3,4-oxadiazole)benzoic acid (**9**). The IR spectrum of the isolated product exhibited a strong bands at 1671(C=O) cm^{-1}, 3259 (OH) cm^{-1}. Its ^{1}H NMR spectrum revealed signals at δ 4.20, 11.84 ppm due to SH and OH protons; its mass spectrum revealed a peak at *m/z* 222 corresponding to its molecular ion.

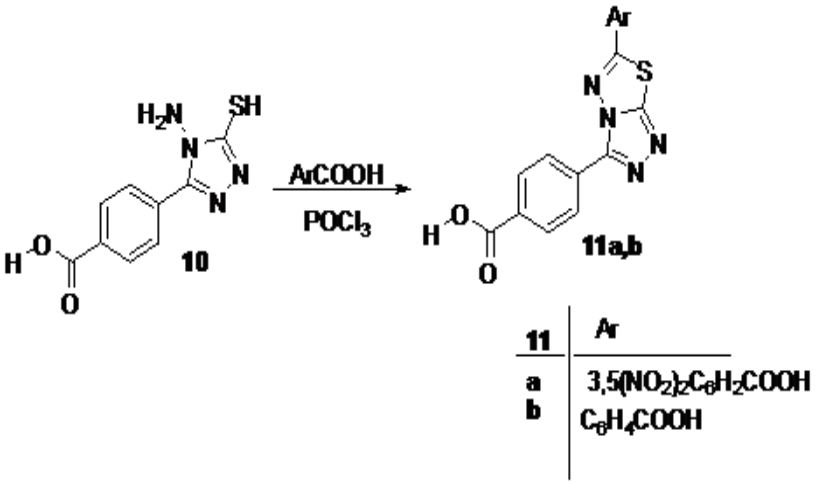
When the thiocarbazinate salt **8** was treated by hydrazine hydrate, it gave the corresponding 4-(5-mercapto-4-amino-1,3,4-triazole)benzoicacid (**10**) (Scheme 2). The structure of the latter product was established on the basis of its elemental analysis and spectral data which are compatible with the assigned structure. For example, its mass spectrum revealed a molecular ion peak at *m/z*236 [*cf. Experimental part*]. Treatment of the mercaptotriazole **(10)** with aromatic carboxylic acids, in the presence of $POCl_3$ in a one pot reaction, afforded [1,2,4]triazolo[3,4-*b*][1,3,4]thiadiazol-3-yl)benzoic acid derivatives (**11a, b)** yield ≈65-77% (Scheme **3**). The structures of the latter products were assigned on the basis of their analytical and spectral data. Thus, the IR spectra of the reaction products showed, in each case, OH absorption bands in the region 3580-3431 cm^{-1}. Their mass spectra revealed molecular ion peaks at the appropriate *m/z* value [*cf. Experimental part*].


5weeks
NaOH/sunlight
Poly (ethylene terephthalate)
(PET) from Plastic waste
1
BuOH
H2SO4
HNO3/H2SO4
2a,b
3a,b
Zn dust
acetic acid
CH(OEt)3/dioxane
4a,b
5a,b
-NH3
6a,b
a,R=(CH2)3CH3
b,R=H

Scheme 1.



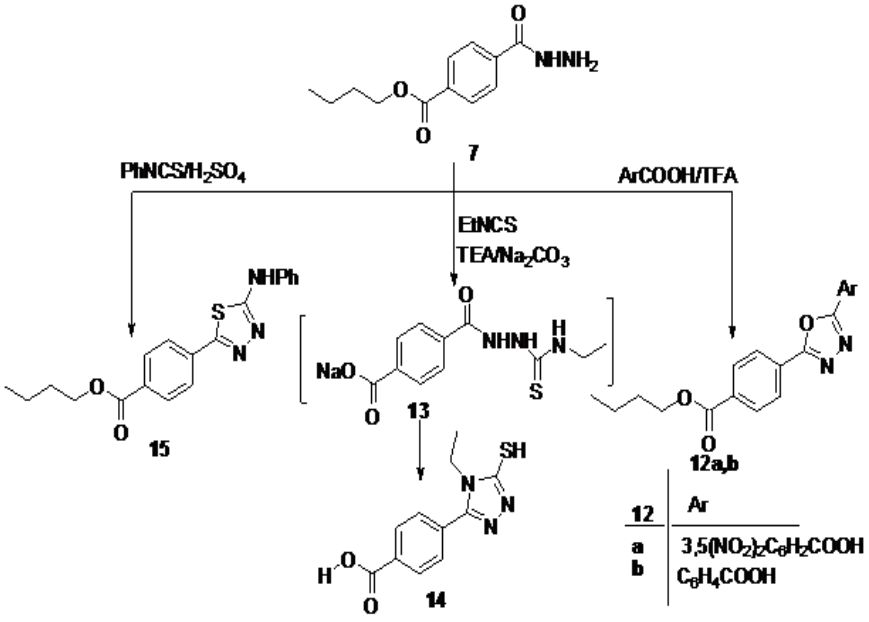
Scheme 2.



Scheme 3.

Treatment of 4-butyrylbenzohydrazide (**7**) with aromatic carboxylic acids; in the presence of trifluoroacetic acid, afforded 1*H*-benzo[1,2,3]triazol-1-yl(1,3,4-oxadiazol-2-yl) benzoate derivatives (**12 a,b)** in yield ≈71-77% (Scheme 4). The structures of the latter products were assigned on the basis of their analytical and

spectral data. Thus, the IR spectra of the reaction products showed, in each case, carbonyl absorption band around the region 1720 cm^{-1}. Their mass spectra revealed molecular ion peaks at the correct *m/z* values [*cf. Experimental part*].



Scheme 4.

4-Butyrylbenzohydrazide (**7**) reacted with ethyl isothiocyanate in dioxan, in the presence of catalytic amount of triethylamine to produce the corresponding mercaptotriazole derivative (**14)** (Scheme 4). The IR spectrum of the isolated product showed a strong carbonyl band at 1689 and OH band at 3428 cm^{-1}. The ^1H NMR spectrum of **14** revealed D_2O-exchangable signals at δ 11.9 and 4.3 ppm due to OH and HS protons, in addition to an aromatic multiplet in the region δ 7.69-8.19. Its mass spectrum showed molecular ion peak *m/z* 249 (100%). On the other hand, when the reaction was carried out in presence of H_2SO_4, the thiadiazole derivative **15** was obtained (Scheme 4). The elemental analyses and spectral data of the obtained product are in complete agreement with the assigned structure.

Biological Evaluation

Anti-microbial activity

Antibacterial and antifungal activities were performed at the Regional Center for Mycology and Biotechnology (RCMB), Cairo University, Cairo, Egypt. Initially, target compounds were evaluated *in vitro* for their antibacterial and

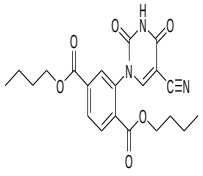
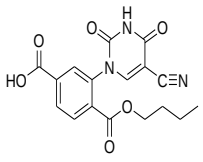
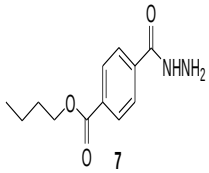
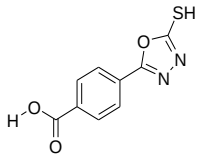
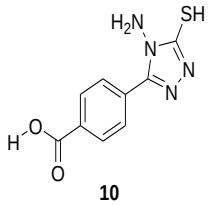
antifungal activity, by inhibition zone technique using two fungi: *A. fumigatus* (RCMB 02568, Af), *Candida albicans* (RCMB 05036,Ca), two Gram positive bacteria: *S. pneumonia* (RCMB 010010,Sp) and *B. subtilis* (RCMB 010069,Bs), two Gram-negative bacteria: *E. coli* (RCMB 010052,Ec) *and Neisseria gonrrhoeae* (NCCP11945, Ng)[19-21]. The results were depicted in Table 1. For the uracil-containing compounds **6a** and **6b**, it was observed that the free carboxylic acid derivative **6b** displayed better antibacterial activities than the ester analog **6a** against the two tested Gram positive and *E. coli* Gram negative bacteria, while both derivatives did not show any significant antifungal activity. Hinting that the presence of free carboxylic acid functionality is more favorable for the antibacterial activity for this scaffold.

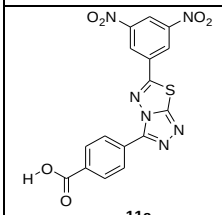
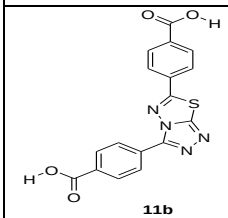
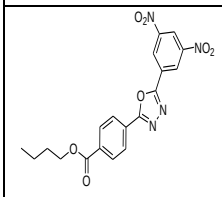
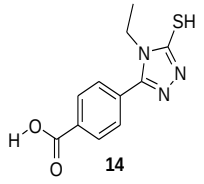
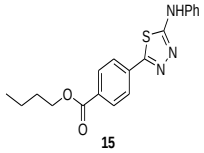
We then investigated the effect of attaching different non fused five-membered rings to benzoic acid (derivatives **8**, **10** and **14**) and 4-ethyl benzoate cores (derivatives **12a** and **15**).Generally, acid derivatives **8**, **10** and **14** showed better antibacterial activities than the esterified members **12a** and **15**, where ester members displayed better antifungal activity specially against *A. flavus.* Also, incorporation of substituted 1,3,4-triazole moiety in compound **9** resulted in a good antibacterial activity against Gram negative bacteria. Moreover, compound **9** bearing substituted 1,3,4-oxadiazole moiety emerged as the most active member against Gram positive *S. peneumoniae* among its acid analogous in this study. On the other hand, incorporation of fused system [1,2,4] triazolo[3,4-*b*][1,3,4]thiadiazol and [1,2,4] triazolo[3,4-*b*][1,3,4]thiadiazole with the benzoic acid core led to compounds **11a** and **11b**, respectively. Compound **11a** (18±0.043 *mg/ml* $^{-1}$) was more potent than **11b** against all tested Gram positive and Gram negative bacteria, whereas **11b** showed superior antifungal activity against *C. albicans* than **11a**.

Antioxidant activity

Organic acids and esters compounds[22,23] and *P*-heterocycles in particular [24] have been recognized for their antioxidant activity. Furthermore, their mechanism of action and the structure-activity relationships (SAR) were extensively studied. The antioxidant activity of the synthesized heterocyclic compounds was evaluated using DPPH radical scavenging assays [25]. This widely used method determines antioxidant activity by measuring the hydrogen donating ability of the compound being studied. IC50 values are displayed in Tables 2 & 3. Vitamin C (ascorbic acid) was used as a positive standard for the antioxidant activity in all experiments. Antioxidant assay was performed by ABST method % Inhibition = {[*A* sample – *A*test] / *A* control} × 100. The data presented in Table 2 showed that all new synthesized compounds showed good to moderate antioxidative activity. Nevertheless, N,S,O-heterocycles **2a**, **7**, and **9** exhibited good radical scavenging ability (56.42 µM ±8.18) as compared to the standard ascorbic acid (34.41%), whereas compounds **6a** and **6b** displayed moderate radical scavenging activity. However, compound **12a** showed minimum activity. Furthermore, [1,2,4]triazolo[3,4-*b*][1,3,4]thiadiazol-3-yl)benzoic acid (**11a) (**54.6 µM ±6.14**)** and 1,3,4-oxadiazolebenzoic acid **9 (**56.42 µM ±8.18) have demonstrated high radical scavenging activity in the micromolar range, and also di-butyl terephthalate (**2a)** (47.1µM±10.34) exhibited high antioxidant activity.

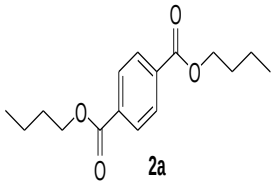
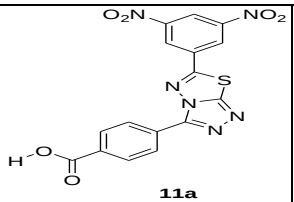
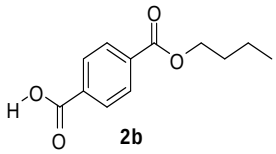
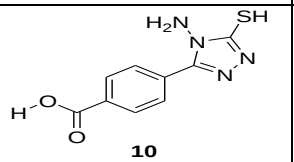
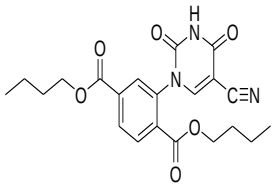
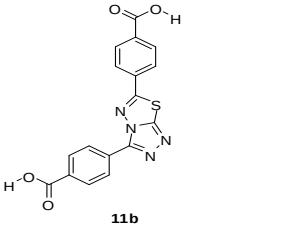
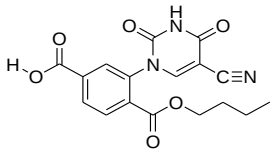
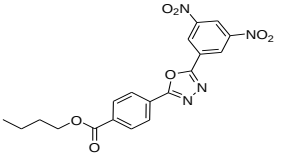
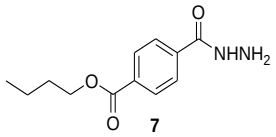
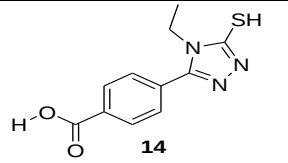
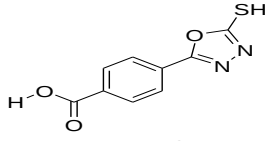
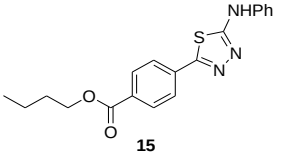
TABLE 1. The antimicrobial activity screening of the prepared compounds at concentration 2mg/disc compared with tetracycline and Amphotericin B as a reference drug.

Sample ID		**Antibacterial,activity (in mm/conc1mg/ml $^{-1}$)**				***Anti-fungial activity***
	Bacillus subtitles (G+)Bs	*Strepto-coccus peneumoniae (G+)Sp*	*Escheri-chia coli (G-)Ec*	*Neisseria gonrrhoeae (G-) Ng*	*Aspergillusf lavus (Fungus)Af*	*Candida albicans (Fungus) Ca*
 **6a**	13±0.031	14±0.033	15±0.036	13±0.031	NA*	NA
 **6b**	15±0.043	18±0.052	21±0.061	13±0.038	NA	NA
 7	16±0.067	15±0.063	16±0.067	18±0.076	21±0.088	15±0.063
 **9**	17±0.076	22±0.09	15±0.067	16±0.076	NA	NA
 **10**	17±0.07	16±0.06	20±0.08	18±0.076	NA	NA

 **11a**	18±0.043	20±0.048	16±0.038	18±0.043	NA	NA
 **11b**	15±0.040	15±0.040	14±0.038	15±0.040	0	12±0.032
 **12a**	NA	NA	NA	NA	13±0.031	12±0.029
 **14**	12±0.048	13±0.052	13±0.052	12±0.048	0	12±0.048
 **15**	13±0.036	14±0.039	13±0.036	13±0.036	0	10±0.028
Tetracycline	30±0.067	30±0.067	32±0.072	30±0.067	0	0
Amphotericin B	-	-	-	-	18±0.019	20±0.021

Mean zone of inhibition in mm ± Standard deviation beyond well diameter (6 mm) produced on a range of clinically pathogenic microorganisms.NA*: No Activity; The screening organisms, Mould: Gram positive bacteria: *B. subtilis* (RCMB 010069,Bs) and *S. pneumonia* (RCMB 010010,Sp), two Gram-negative bacteria: *E. coli* (RCMB 010052,Ec) and *Neisseria gonrrhoeae* (NCCP11945, Ng), two fungi *A. fumigatus* (RCMB 02568,Af), *Candida albicans* (RCMB 05036,Ca). Inhibition zone : High activity >12(mm), Moderate activity 9-11(mm),Slight activity 7-8(mm) and Non sensitive 0-6(mm).

TABLE 2. DPPH radical scavenging of new synthesized compounds.

Sample ID	DPPH IC_{50} (µM)[a]	Sample ID	DPPH IC_{50} (µM)[a]
Vitamin C 34±6.17[b]	34±6.17 [b]	Vitamin C	
 2a	47.1±10.34	 11a	54.6±6.14
 2b	68.34±10	 10	66.06±0.525
 6a	62.14±10.01	 11b	65.41±6.08
 6b	60.384±9.82	 12a	73.88±1.72
 7	58±16.25	 14	71.72±8.21
 9	56.42±8.18	 15	67.32±2.3

IC_{50} values represent as mean±SD of three determinations. [b]Reported IC_{50} =38.0 µM .

Experimental

Chemical reagents and instruments

Melting points were determined with an open capillary tube on an Electrothermal (variable heater) melting point apparatus. Later on, the used thermometer was calibrated by using standard compounds of known mps and the melting points of the new compounds were corrected exclusively. IR spectra were recorded on a JASCO FT-IR 6100 using KBr the bromide discs. NMR spectra were measured using JEOL E.C.A-500 MHz (^{1}H: 500.7 MHz, ^{13}C: 125.4 MHz) spectrometer. The mass spectra were performed at 70 eV on an MS-50 Kratos (A.E.I.) spectrometer provided with a data system. Elemental analysis of the products was carried out at the Microanalysis Laboratory, Cairo University, Cairo, Egypt, using Elemental C, H, N analyzer Vario EL II I Germany. The purity of all new samples was verified by microchemical analysis (C/H/N/ S) and spectroscopy. TLC: Merck 0.2 mm silica gel 60 F254 analytic aluminum plates. All international principles and local regulations concerning the care and use of laboratory animals were considered during the pharmacological screening.

General procedure

PET hydrolysis

Plastic bottles (22 g) were cut into small strips, mixed with NaOH (12 g) and exposed to sunlight for 5 weeks to obtain the sodium salt of terephthalate. The resulting materials were dissolved in water, followed by acidification using H_2SO_4 (5 mol/L) to afford a white precipitate of terephthalic acid (**1**), Yield 90 %; m.p. above 300 $^{\circ}$ C. Terephthalic acid (10g, 1.66 mol) was refluxed in absolute butanol (30 ml) and H_2SO_4 (5 ml) for 6 hr to give a mixture of dibutylterephthalate (**2a**) and 4-(butoxycarbonyl) benzoic acid (**2b**). Compound **2a** was obtained as yellow oil, 80% yield. IR (KBr) ν_{max}/cm^{-1}: 1723 (C=O); ^{1}H NMR (DMSO-d_6): 0.853 (t, 6H, 2C H_3), 1.003 (m, 4H, 2C H_2), 1.87 (dd, 4H, 2C H_2), 4.19 (t, 4H, 2OC H_2), 7.75 (dd, J_{HH} = 8.5Hz, 2H, *H*-Ar), 8.10 (dd, J_{HH}=8, 2H, *H*-Ar); *m/z* (%) 278 (M^{+}, 100.0%), 216 (27.5%); Anal calcd for $C_{16}H_{22}O_4$ (278.15): C, 69.04% ; H, 7.97 %; Found C, 69.00%; H, 7.90%.

4-(Butoxycarbonyl) benzoic acid (**2b**) was separated by washing with 10% Na_2CO_3 and recrystallized from EtOH as white solid in 20% yield; m.p 116^{o} C; IR(KBr) ν_{max}/cm^{-1}: 3466 (OH), 1722, 1693 (C=O), 2961, 2872 (CH); ^{1}H NMR (DMSO-d_6**)**: 0.89 (t, 3H, C H_3), 1.3 (m, 2H, CH_2) , 1.6 (m, 2H, C H_2), 4.02 (s, 1H, O*H*, D_2O exchangeable), 4.13(t, 2H, OC H_2) , 7.95 (dd, J_{HH} = 8.5Hz, 2H, *H*-Ar), 8.01 (dd, J_{HH} =8.5Hz, 2H, *H*-Ar); *m/z* (%) 206 (M^{+}, 100.0%), 191 (27.5%); Anal calcd for $C_{12}H_{14}O_4$ (222.09): C, 64.85 %; H, 6.35 %; Found C, 64.80 %; H, 6.83%.

*Synthesis of nitro derivatives (**3a, b**)*

A 500 ml three-necked round-bottomed flask, was equipped with a magnetic stirrer, thermometer, decanter and condenser, was charged with 20 g of compounds **2a** or **2b**. A mixture of conc. H_2SO_4 (25 ml) and fuming HNO_3 (75

ml) was added dropwise that caused that temperature to raise to 80 ^{o}C. After the addition was completed, the reaction was continued at 100^{o}C for 3 hr, and then poured onto water / ice mixture. The precipitate was filtered off, dissolved in hot water and recrystallized from EtOH / DMF (2:1) to afford nitro derivatives **3a, b**.

Dibutyl 2-nitroterephthalate (**3a**) was obtained as yellow powder in 75% yield; mp 119 ^{o}C; IR(KBr) ν_{max}/cm^{-1}: 1625 (C=O), 1466 (CH_2), 1313 (N-O, asymm.), 1120 (N-O, symmetric); ^{1}H NMR (DMSO-d_6)**:** 0.89 (t, 6H, 2C H_3), 1.34 (m, 4H, 2 H_2), 1.61 (m, 4H, 2C H_2), 4.26 (t, 4H, 2OCH_2), 7.94 (s, 1H, *H*-Ar), 8.3 (d, 1H, J_{HH} = 9 Hz, *H*-Ar), 8.43 (d, J_{HH} = 9 Hz, 1H, *H*-Ar); *m/z*(%) 323 (M^{+}, 100.0%); Anal calcd for $C_{16}H_{21}NO_6$ (323.14): C, 59.43 %; H, 6.55 %; N, 4.33%; Found: C, 59.40 %; H, 6.60%; N, 4.2%.

4-(Butoxycarbonyl)-3-nitrobenzoic acid (**3b**) was obtained as white substance in a 70% yield; mp 123^{o}C; IR(KBr) ν_{max}/cm^{-1}: 3025 (OH), 1675, 1653 (C=O), 1313(N-O, asymmetric), 1120 (N-O, symmetric); ^{1}H NMR (DMSO-d_6)**:** 0.89 (t, 3H, CH_3), 1.59 (m, 4H, 2 C H_2), 4.25 (t, 2H, OCH_2), 7.91 (s, 1H, *H*-Ar), 8.25 (d, J_{HH} = 9 Hz, 1H, *H*-Ar), 8.39 (d, J_{HH} = 9 Hz, 1H, *H*-Ar)), 4.05 (s, 1H, O*H*, D_2O exchangeable), 4.23 (t, 2H, OCH_2), ; *m/z*(%) 267 (M^{+}, 100.0%); Anal calcd for $C_{12}H_{13}NO_6$ (267.23): C, 53.93%; H, 4.90%; N, 5.24%; Found: C, 53.90%; H, 5.00%; N, 5.00%.

Preparation of amino derivatives (4a, b)

A 250 ml three- necked round-bottomed flask, fitted with a mechanical stirrer containing 9 g of Zn dust, 15 ml of acetic acid, 15 ml of water and 0.5 ml fuming HCl, was heated under reflux for 10 min. 3.2 g of the nitro derivatives **3a** or **3b** were added portion wise for 20 min. The reaction was stirred for another 10 min under reflux. The reaction was cooled down at room temperature followed by addition of 0.3 g of $NaHCO_3$. After the reaction was stirred for 3hr, the mixture was filtered off, concentrated and dissolved in EtOH (20ml) and 10 ml HCl. The mixture was heated for 1hr. The precipitated material was filtered off and crystallized from EtOH to afford the amino derivatives **4a,b.**

Butyl-2-amino-4-pentanoylbenzoate **(4a)** was obtained as yellow brown substance in a 65% yield; mp 129 ^{o}CIR(KBr) ν_{max}/cm^{-1}: 3423 (NH_2), 1709(C=O), ^{1}H NMR (DMSO-d_6)**:** 1.02 (t, 6H, 2C H_3), 1.38 (m, 4H, 2CH_2), 1.64 (m, 4H, 2C H_2) 4.24 (t, 4H, 2OCH_2), 7.65 (s, 1H, *H*-Ar), 8.03 (d, J_{HH} = 9.2 Hz, 1H, *H*-Ar), 8.6 (d, J_{HH} = 9.2 Hz, 1H, *H*-Ar), 10.45 (s, 2H, NH_2); *m/z* (%) 293 (M^{+}, 100%), 291 (M-2H, 21%); Anal calcd for $C_{16}H_{23}NO_4$ (293.36): C, 65.51%; H, 7.90%; N, 4.77%; Found: C, 65.54%; H, 7.92%; N, 4.73%.

3-Amino-4-(butoxycarbonyl)benzoic acid **(4b**) was obtained as yellow in 60% yield; mp 198; IR(KBr) ν_{max}/cm^{-1}: 3423 (NH_2), 1702 (C=O), 1623 (C=O); ^{1}H NMR (DMSO-d_6)**:** 1.02(t, 3H, CH_3), 1.37 (m, 2H, CH_2), 1.73 (m, 2H, CH_2), 4.05 (s, 1H, O*H*, D_2O exchangeable), 4.23 (t, 2H, OCH_2), 6.64 (s, 2H, NH_2, D_2O exchangeable), 7.0 (s,1H, *H*-Ar), 7.33 (dd, J_{HH} = 8.4 Hz, 1H, *H*-Ar), 7.67 (dd, J_{HH}

= 8.4 Hz, 1H, *H*-Ar); *m/z*(%) 237 (M^+, 100.0%), 219 (22%); Anal calcd for $C_{12}H_{15}NO_4$ (237.10): C, 60.75; H%, 6.37 %; N, 5.90 %; Found: C, 60.70%; H, 6.45%; N, 5.88%.

*Preparation of uracil derivatives (**6a, b**)*

Compounds **4a, b** (0.01 mol), triethylorthoformate (0.03 mol, 4.4 ml) and cyanoacetyl urea (0.127 g, 0.01mol) were refluxed in dioxane for 8 hr (until the evolution of NH_3). The solid product was formed on hot, filtered off and crystalized from the proper solvent to give **6a,b**.

Dibutyl 2-(5-cyano-2-hydroxy-4-oxopyrimidin-1(4*H*)-yl)benzene-1,4-dioate (**6a**) was obtained as brown substance (from EtOH/DMF, 2:1), in 62% yield; mp 198 ^oC ; IR (KBr) ν_{max}/cm^{-1} : 3348 (OH), 2225 (CN), 1702, 1612 (C=O); ^1H NMR (DMSO-d_6): δ 1.02 (t, 6H, 2CH_3), 1.3- 1.7 (m, 8H, 4CH_2), 4.23 (t, 4H, 2OCH_2)**,** 7.38-7.99(m, 3H, *3H*-Ar), 9.95(s, 1H, *H*-pyrimidin), 12.5(s, 1H, N*H*, D_2O exchangeable); ^{13}C NMR (DMSO *d6*), δ 13.8(*C*H_3), 18.9(*C*H_2), 31.1(*C*H_2), 64.5(*C*H_2), 114.1 (*C*H), 115.8 (*C*N), 119.8(*C*H), 125.9(*C*H), 130.6 (*C*H), 135.2(*C*H), 142.6(*C*H), 163(*C*H), 165.4 (*C*=O), 168.4 (*C*=O); *m/z* (%) 413 (M^+, 100.0%),223 (6.28%); Anal calcd $C_{21}H_{23}N_3O_6$ (413.42); C, 61.01%; H, 5.61%; N, 10.16;% Found C, 61.05%; H,5.62%; N,10.62%.

4-(Butoxycarbonyl) -2- (5-cyano-2- hydroxy-4- oxopyrimidin-1 (4*H*)-yl) benzoicacid (**6b**) was obtained as green substance (from EtOH**)** in 75% yield; mp 240^oC**;** IR (KBr) ν_{max}/cm^{-1}**:** 3245(OH), 2243 (CN), 1658(C=O) , 1602 (C=O); ^1H NMR (DMSOd_6): δ 1.02 (t, 3H, CH_3), 1.3 (m, 2H, CH_2), 1.7 (m, 2H, CH_2), 2.05 (s, 1H, O*H*), 4.05 (s, 1H**,** O*H*, D_2O exchangeable), 4.23 (t, 2H, OCH_2)**,** 7.47- 7.85 (m, 3H, *H*-Ar), 8.45 (s, 1H, *H*-pyrimidin); ^{13}C NMR (DMSO d_6): δ 13.8(*C*H_3), 18.9 (*C*H_2), 31.1(*C*H_2),64.5 (*C*H_2), 114.1 (*C*H), 115.8 (*C*N), 119.8 (*C*H), 127.8(*C*H), 131.1 (*C*H), 136.1(*C*H), 143 (*C*H), 163 (*C*H), 165.4 (*C*=O), 166(*C*=O); *m/z* (%) 341 (M^+, 100.0%),320 (25%); Anal calcd $C_{17}H_{15}N_3O_6$ (357.32); C, 57.14%; H, 4.23;%; N, 11.76;% Found C, 57.12%; H,4.25%;N,11.70%.

*Preparation of 4-butyrylbenzohydrazide (**7**)*

Dibutylterephthalate (**2a**) (10 ml) and hydrazine hydrate (1.4 ml) were refluxed in absolute ethanol (30 ml) for 8 hr. The reaction mixture was filtered off, washed with ether, dried then recrystallized from ethanol to afford 4-butyrylbenzohydrazide (**7**) as white solid in an 87% yield; m.p 300^o C; IR(KBr) ν_{max}/cm^{-1}: 3321(NH),1659 (C=O), 1612 (C=C), cm^{-1}; ^1H NMR (DMSOd_6**)**: δ 1.009 (t, 3H, C H_3), 1.379 (m, 2H, C H_2) , 1.660(m, 2H, CH_2), 3.73 (t, 2H, CH_2), 4.24 (br, 2H, NH_2, D_2O exchangeable), 7.81(dd, J_{HH} =7.5 Hz, 2H, *H*-Ar), 7.961(dd, J_{HH} = 7.5 Hz, 2H, *H*-Ar), 9.83 (br, 1H, N*H*, D_2O exchangeable); ^{13}C NMR (DMSO *d6*):δ 13.8 (*C*H_3), 18.9(*C*H_2), 31.1(*C*H_2), 64.5(O*C*H_2), 93.5 (*C*-CN), 114.1(*C*H), 115.8(*C*N), 119.8 (*C*H), 125.9 (*C*H), 130.6 (*C*H), 135.5(*C*H), 142.6 (*C*H), 163 (*C*=O), 167.9(*C*=O); *m/z* (%) 236 (M^+, 100.0%), 191 (27.5%); Anal calcd $C_{12}H_{16}N_2O_3$ (236.27) C, 61.00%; H, 6.83%, N, 11.86%; Found C, 61.23%; H, 6.88%, N, 11.80%.

*Synthesis of potassium thiocarbazinate (**8**)*

Potassium hydroxide (3 mmol) was dissolved in absolute ethanol (25 ml). The solution was cooled in ice bath. 4-Butyrylbenzohydrazide **(7)** (1 mmol) was added with stirring. Carbon disulfide (5 mmol) was added portionwise with constant stirring. The reaction mixture was agitated continuously for 12 hr at room temperature. The precipitated potassium thiocarbazinate was collected by filtration, washed with cold ethanol (50 ml) and dried in vacuum. The potassium salt, thus obtained, was used in the next step without further purification. The latter was treated with water and then filtered. the filtrate was cooled and neutralized to pH= 6 using diluted HCl, washed with water, dried and crystalized from ethanol to afford 4-(5-mercapto-1,3,4-oxadiazol-2-yl)benzoic acid **(9)** as yellow crystals in 80% yield; m.p>300 ^{o}C; IR (KBr) ν_{max}/cm^{-1}: 3259 (OH), 1671(C=O) ;^{1}H NMR (DMSOd_6**)**: 4.23(s, 1H, S*H*, D_2O exchangeable), 7.69(dd, J_{HH} = 8.5Hz, 2H, *H*-Ar), 8.19(dd, J_{HH} = 8.5Hz, 2H, *H*-Ar), 11.84 (s, 1H, O*H*) ; ^{13}C NMR (DMSO *d6*): 127.5(*C*H), 130.2(*C*H), 131.2(*C*H), 164.5(*C*-O), 169.3(*C*=O); *m/z* (%)222 (M^+, 100.0%), 205 (7.11%), 180 (41.49%); Anal calcd $C_9H_6N_2O_3S$(222.22) C, 48.64%; H, 2.72%, N, 12.61%,S, 14.43%; Found C, 48.68%; H, 2.69%;N, 12.63%; S,14.40%.

A mixture of potassium thiocarbazinate (1mmol) in water (5 ml) and hydrazine hydrate (99%, 3mmol) was heated for 18h at 100^{o} C with occasional shaking. The color of the reaction mixture was changed to green with evolution of hydrogen sulfide gas. A homogenous reaction mixture was obtained during reaction process. The reaction mixture was cooled to room temperature and diluted with cold water (20 ml). On acidification with HCl, 4-(5-mercapto-4-amino-1,3,4-triazole)benzoic acid **(10)** was precipitated, collected and recrystallized from DMF/H_2O (1:2), 80% yield, m.p 235^{o} C; IR (KBr) ν_{max}/cm^{-1}: 3855(OH), 3434.6(NH_2), 1623(C=O); ^{1}H NMR (DMSOd_6**)**: δ 4.2(s, 1H, S*H*, D_2O exchangeable), 7.99(dd, J_{HH} = 7.5Hz, 2H, *H*-Ar), 8.11(dd, J_{HH} = 7.5Hz, 2H, *H*-Ar), 11.84(s, 1H, O*H*), 14.03 (s, 2H, NH_2, D_2O exchangeable) ; ^{13}C NMR (DMSO *d6*): δ127(*C*H), 128(*C*H), 128.4(*C*H), 129.7(*C*H), 142(*C*-SH), 155(*C*H-triazole), 167 (*C*=O); *m/z* (%)236 (M^+, 100.0%), 180 (41.49%); Anal calcd $C_9H_8N_4O_2S$ (236.25) C, 45.75%; H, 3.41%, N, 23.72%, S, 13.57%; Found C, 45.77%; H, 3.42%, N, 23.65%, S,13.59%.

*Synthesis of 1,4 (6-substituted-[1,2,4]triazole[3,4-b][1,3,4]thiadiazoles **11 a,b.***

An equimolar mixture of compound **10** and the appropriate aromatic carboxylic acid (1 mmol) in phosphorous oxychloride (5ml) was refluxed for 5hr. The reaction mixture was cooled to room temperature and then gradually poured onto crushed ice with stirring. The mixture was neutralized with $NaHCO_3$ solution and allowed to stand overnight. The solid that separated out was filtered and washed thoroughly with cold water to give 4-(6-(3,5-dinitrophenyl)-[1,2,4]triazolo[3,4-b][1,3,4]thiadiazol-3-yl)benzoic acid (**11a**) in a 65% yield, mp 260^{o} C ; IR (KBr) ν_{max}/cm^{-1} 3431(OH), 1638(C=O),symmetric1153(N-O); ^{1}H NMR (DMSOd_6**)**: δ 2.05 (s, 1H, O*H*, D_2O exchangeable), 7.60 - 8.80 (m, 6H, *H*-Ar), 9.08(s, 1H, O*H*, D_2O exchangeable); ^{13}C NMR (DMSO *d6*): δ 118.1(*C*H),

127.4(*C*H), 130.2(*C*H), 135.3 (*C*H), 143.3(*C*H), 148(*C*H), 149.3(*C*-N),167.6(N-*C*-S), 169.3(*C*=O); *m/z*(%) 412 (M^+, 100.0%), 180 (41.49%); Anal calcd $C_{16}H_8N_6O_6S$ (412.02) C, 46.61%; H, 1.96%, N, 20.38%, S, 7.78%; Found C, 46.63%; H, 1.97%, N, 20.38%, S,7.80% ; 4-(6-*p*-Carboxyl-[1,2,4] triazolo [3,4-*b*] [1,3,4]thiadiazol-3-yl) benzoic acid (**11b**) 77% yield, m.p 280 ^{o} C ; IR (KBr) ν_{max}/cm^{-1}3580 (OH), 1641(C=O); ^{1}H NMR (DMSO d_6): δ 7.69 - 8.19 (m, 8H, *H*-Ar), 11.40, 13.27(2bs, 2H, O*H*, D_2O exchangeable), ^{13}C NMR (DMSO d_6): δ 127.5 (*C*H), 130.2(*C*-C=O), 135.8(*C*H|), 138.7 (*C*H), 143.3 (*C*H), 149 (*C*H), 167.6 (*C*H), 170 (*C*=O); *m/z*(%)366 (M^+, 100.0%), 180 (41.49%); Anal calcd $C_{17}H_{10}N_4O_4S$ (366.35) C, 55.73%; H, 2.75%, N, 15.29%, S, 8.75%; Found C, 55.74%; H, 2.75%, N, 15.30%, S,8.72%.

*General procedure for the preparation of butyl (5-aryl-1,3,4-oxaadiazole-2-yl)benzoate (**12a,b**)*

A mixture of compound **7** (1 mmol) and the appropriate aromatic carboxylic acid (1 mmol) in trifluoroacetic acid (10 ml) was refluxed for 4-6hr. The reaction mixture was slowly poured onto crushed ice and kept overnight. The solid thus separated out, was neutralized with $NaHCO_3$, filtered, washed with water and recrystallized from ethanol.

Butyl 4-(5-(3,5-dinitrophenyl)-1,3,4-oxadiazol-2-yl)benzoate (**12a**) was obtained as white powder; yield:77% , m.p220^o C ; IR (KBr) ν_{max}/cm^{-1} 3088(CH), 1719(C=O), 1473 asymmetric (NO), 1348 asymmetric (NO); ^{1}H NMR (DMSO d_6): δ 0.897(t, 3H, CH_3), 1.38 (m, 2H, C H_2), 1.66 (m, 2H, C H_2), 4.266(t, 2H, OCH_2), 7.9-8.85(m, 6H, *H*-Ar), 10.99(s, 1H, O*H*, D_2O exchangeable) ; ^{13}C NMR (DMSO *d6*): δ 13.8(*C*H_3), 18.9(*C*H_2), 31.1(*C*H_2), 64.5 (O*C*H_2), 118.1 (*C*H), 127.4 (*C*H), 128.9 (*C*H), 130.4(*C*H), 147.4 (*C*H), 149.3(*C*H), 164.5 (*C*-O), 165.9 (*C*=O), *m/z* (%)412 (M^+, 100.0%), 395 (73.37%); Anal calcd $C_{19}H_{16}N_4O_7$ (412.35) C, 55.34%; H, 3.91%, N, 13.59%; Found C, 55.32%; H, 3.92%, N, 13.59%.

4-{5-[4-(Butoxycarbonyl) phenyl]-1,3,4-oxadiazol-2-yl} benzoic acid (**12b**) was obtained as white powder; yield :71%; m.p223 ^{o} C IR (KBr) ν_{max}/cm^{-1} 3248(OH), 1719 (C=O), 1666 (C=O); ^{1}H NMR (DMSO d_6): δ 0.96(t, 3H, C H_3), 1.33 (m, 2H, CH_2), 1.75(m, 2H, CH_2), 2.05(s, 1H, O*H*, D_2O exchangeable), 4.25(t, 2H, OCH_2), 7.59 - 8.19 (m, 8H, *H*-Ar); ^{13}C NMR (DMSO d_6): δ13.8 (*C*H_3), 18.9 (*C*H_2), 31.1(*C*H_2), 64.5(O*C*H_2), 127-130.5(*C*H), 147.4(*C*H), 164.5(*C*H), 165.9(*C*=O), 170(*C*=O); *m/z* (%) 367 (M^+, 100.0%), 349(18.01%), 310(10.18%); Anal calcd $C_{20}H_{18}N_2O_5$ (366.37) C, 65.57%; H, 4.95%, N, 7.65%; Found C, 65.58%; H, 4.96%, N, 7.66%.

*Reactions of hydrazide **7** with isothiocyanate derivatives*

Compound **7** (0.01 mol) was refluxed with an equimolar amount of ethyl isothiocynate in dry dioxane (30 ml) in presence of NaOH for 3hr. After the solution was cooled, the solid formed was filtered off and recrystallized from EtOH.

4-(4-Ethyl-5-mercapto-4H-1,2,4-triazol-3-yl)benzoic acid (**14**) was obtained as green crystals in a 75% ; mp 198°C ; IR(KBr) ν_{max}/cm^{-1} **:** 3428 (OH), 1689(C=O);^{1}H NMR (DMSOd_6): δ 1.02(t, 3H, CH_3), 3.2(q, 2H, CH_2), 4.3 (s, 1H, S*H*), 7.69(dd, J_{HH} = 8.2 Hz ,2H, *H*-Ar), 8.19(dd, J_{HH} = 8.2 Hz, 2H, *H*-Ar), 11.9(s, 1H, OH); ^{13}C NMR (DMSO d_6): δ13.9 (*C*H_3), 19.11(*C*H_2), 126.7(*C*H), 129.8(*C*H), 130.6(*C*H), 134.1(*C*H), 165.6(*C*H), 167(*C*H), 178(*C*=O); *m/z* (%) 146 (M^+, 100.0%), 219 (25%); Anal calcd $C_{11}H_{11}N_3O_2S$ (249.29); C, 53.00%; H, 4.45%; N, 16.86% ; S,12.86% Found C, 53.00%; H,4.43%; N,16.87%; S,12.83%.

Moreover, compound **7** (0.01 mol) was heated with phenyl isothiocynate in dioxane and conc H_2SO_4 (3 ml) at 100°C for 1hr. The solution was then cooled, and water was added dropwise till precipitation is ended. The solid, thus formed, was filtered off, washed and crystallized from ethanol to obtain butyl 4-(5-(phenylamino)-1,3,4-thiadiazol-2-yl)benzoate **(15)** was obtained as green crystals in a 71%; m.p. 225°C ; IR(KBr) ν_{max}/cm^{-1} **:** 3406 (OH), 1688(C=O);^{1}H NMR (DMSOd_6): δ 0.91(t, 3H, CH_3), 1.40 (m, 2H, CH_2), 1.68(m, 2H, CH_2), 4.05(s, 1H, N*H*, D_2O exchangeable) 4.28(t, 2H, OCH_2), 6.46-7.01(m, 5H, *H*-Ar), 7.59 (dd, J_{HH} = 8.2 Hz, 2H, *H*-Ar) , 8.03 (dd, J_{HH} = 8.2 Hz, 2H, *H*-Ar) ;^{13}C NMR (DMSOd_6): δ 13.9 (*C*H_3), 18.9 (*C*H_2), 31.1 (*C*H), 64.5 (O*C*H_2), 117.8 (*C*H), 122.4 (*C*H), 127.4 (*C*H), 129.5(*C*H), 137.8(*C*H), 147.4(*C*H), 152.7 (*C*H), 165.9(*C*=O); *m/z* (%) 55 (M^+, 100.0%), 77(82.22%), 242(34%); Anal calcd $C_{19}H_{19}N_3O_2S$(353.44) C, 64.57%; H, 5.42%, N, 11.89%, S, 9.07%;Found C, 64.77%; H, 5.40%, N, 11.86%, S, 9.06%.

Conclusion

In this investigation, we have developed an efficient and simple method for the synthesis of a series of new 1,3,4-oxadiazol, triazoles , thiadiazoles and uracils in good yields. The starting materials were obtained from plastic bottle waste using the renewable source of energy. This technique was successful to produce potent broad-spectrum antimicrobial and antioxidant agents. The structure-activity relations study displayed in Tables 1 & 2 and Schemes 1-4 showed that the antibacterial activity of benzoic acid hydrazide **7** resembles that of the known parapene preservatives which are useful in industrial applications. In this study, novel heterocycles were successfully synthesized and twelve of them were evaluated for their antimicrobial and antioxidant activities. In addition, the 1,3,4-oxadiazol **9** showed higher activity against all types of strains. Also, compounds of thiadiazoles **11a** and 1,3,4-oxadiazole **9** exhibited high antioxidant activity due to presence of the OH group. It can be concluded that this class of compounds certainly holds great promise towards pursuit to discover novel class of antimicrobial and antioxidant agents. Further studies are being conducted to acquire more information about quantitative structure-activity relations (QSAR).

Acknowledgement : The authors thank the Micro-Analytical Center, Cairo University, Egypt, for carrying out the pharmacological screening.

References

1. **Abu-Zaied, M. A. E M. El Telbanie, G. H. Elgemeie, G. A. M. Nawwar**, Synthesis and in vitro anti-tumor activity of new oxadiazole thioglycosides *Eur. J.Med. Chem*., **46**, 229-235 (2011).

2. **S**. **Palekar, A. J. Damle**, **S. R. Shukla**, Synthesis and antibacterial activity of some novel bis-1,2,4-triazolo[3,4-b]-1,3,4-thiadiazoles and bis-4-thiazolidinone derivatives from terephthalic dihydrazide", *Eur. J.Med. Chem*., **44,** 5112–5116 (2009).

3. **M. R. Shiradkar, K.K. Murahari, H.R Gangadasu; T. Suresh; C.C.Kalyan,; D.Panchal; R. Kaur , P. Burange, J. Ghogare; V. Mokale, M. Raut,** Synthesis of new S-derivatives of clubbed triazolyl thiazole as anti-Mycobacterium tuberculosis agents *Bioorg. Med. Chem*, **15**, 3997-4008 (2007).

4. **A**. **Gürsoy**, **A**. **Karali**, "Synthesis and primary cytotoxicity evaluation of 3-[[(3-phenyl-4(3H)-quinazolinone-2-yl)mercaptoacetyl]hydrazono]-1H-2-indolinones *Eur. J. Med. Chem*., **38,** 633-643 (2003).

5. **P.Vicini; M. Incerti, I. Doytchinova; P.La Colla, B. Busonera, R. Loddo**, Synthesis and antiproliferative activity of benzo[d]isothiazole hydrazones; *Eur. J.Med. Chem.*, **41**, 624-632 (2006).

6. **Asmaa mahmoud Fahim, El-Sayed Mohamed Yakout and Galal A.M. Nawwar**, "Facile Synthesis of in-Vivo Insecticidal and Antimicrobial Evaluation of Bis Heterocyclic Moiety from PET Waste", Journal of biological sciences, **14** (3),196-208 (2014).

7. **R. Venkat Ragavan, V. Vijayakumar; N. Suchetha Kumari;** "Synthesis and antimicrobial activities of novel 1,5-diaryl pyrazoles", *Eur. J. Med. Chem*., **45**, 1173–1180 (2010).

8. **S. Miroslav, J. Hlaváč, H. Pavel, I. Fryšová, M. Hajdúch, V. Bertolasi, M. Maloň;** "Synthesis and cytotoxic activity of substituted 2-phenyl-3-hydroxy-4(1H)-quinolinones-7-carboxylic acids and their phenacyl esters *Eur. J. Med. Chem*. **41**, 467–474 (2006).

9. **D. Paszun, T. Spycha**; Chemical Recycling of Poly(ethylene terephthalate) *J. Ind. Eng. Chem. Res.*, **36**, 1373 (1997).

10. **A.Oku, L.-C.Hu, E. Yamada**, Alkali decomposition of poly(ethylene terephthalate) with sodium hydroxide in nonaqueous ethylene glycol: A study on recycling of terephthalic acid and ethylene glycol. A study on recycling of terephthalic acid and ethylene glycol", J. *Appl. Polym. Sci*. , **63**, 595 (1997).

11. **S.M. Al-Salem; P. Lettieri, J. Baeyens**; "Recycling and recovery routes of plastic solid waste (PSW): A review", Waste Management , **29,** 2625–2643 (2009).

12. **E. Denise Chenot, D. Bernadi, A. Comel;** "Preparation of Monoalkyl Terephthalates: An Overview", Synth. Commun., **37**, 483–490 (2007).

13. **C. Kawamura, K. Ito, R. Nishida, I. Yoshihara, N. Numa**; "Coating resins synthesized from recycled PET", Progress in Organic Coatings, **45,** 185–191 (2002).

14. **" K. Wojciechowski; A.Wyrebak; J. Gumulak** ; Direct dyes derived from iso- and terephthalic acids", Dyes and Pigments, 2003, **56**, 99–109.

15. **B. Zhuang, G. J. Hsu and K. Sung;** "A multi-component reaction to 5-cyanouracils: Synthesis and mechanistic study. *Bioorg. Med. Chem*., **14** , 3399-3404 (2006).

16. **K. M. Dawood; A. M. Farag; H. A. Abdel-Aziz**; Synthesis and antimicrobial evaluation of some 1,2,4-triazole, 1,3,4-oxa(thia)diazole, and 1,2,4-triazolo[3,4-b]-1,3,4-thiadiazine derivatives", *Heteroatom Chemistry*, **16** (7), 621-627 (2005).

17. **M. G. Soni, S.L.Taylor, N.A.Greenberg, G.A.Burdock;** "Evaluation of the health aspects of methyl paraben: a review of the published literature *Food Chemical Toxicology*, **40**, 1335-1373 (2002).

18. **S. M. A. Yakout , Y. A. Allam, G.A. M. Nawwar;** "Simple syntheses of benzothiazol-2-ylazoles. *Heteroatom Chemistry*, **10** (3), 177-182 (1999).

19. **A. Husain , M. Arif Naseer;** "Studies on fused heterocyclic 3,6-disubstituted-1,2,4-triazolo-1,3,4-thiadiazoles: synthesis and biological evaluation", *Med. Chem. Res.*, **20** (1), 47–54 (2011).

20. "Antifungal Activity of Essential Oils from Indian Medicinal Plants Against Human Pathogenic Aspergillus fumigatus and A. niger", S. Bansod, and M. Rai; W.J.M.S., **3** (2), 81-88 (2008).

21. Determination of minimum inhibitory concentrations", J. M. Andrews; *J. Antimicrob. Chemother*., **48**, 5-16 (2001).

22. "Synthesis and Evaluation of New Antioxidants for Styrene Butadiene Rubber" , Galal Nawwar, Sayed Yakout, M.S.A. El-Sadiek and Salwa El-Sabbgh; *Pigment & Resin Technology*, **40** (6), 399-409 (2011).

23. "Antioxidants in Organophosphorus Compounds Poisoning", S. M. Nurulain, P. Szegi, K.Tekes, S. N. Naqvi; *Arh. Hig. Rada Toksikol*, **64,** 169–177 (2013).

24. "Organophosphorus antioxidants—VIII. Kinetics and mechanism of the reaction of organic phosphites with peroxyl radicals", K. Schwetlick, J. Pionteck, T. W. D. Habicher; *Eur. Polym., J.* **23,** 383–388 (1987).

25. "Mechanisms of antioxidant action of organic phosphorus compounds", K.Schwetlick; *Pure and Appl. Chem.,* **55** (10), 1629-1636 (1983).

(*Received* 31/12/2015;
accepted 12/ 6/2016)

التشييد والتقييم البيولوجى لمشتقات 1,3,4- أوكساديازول ، ترايازول واليوراسيل بإستخدام نفايات البلاستيك (بولى إيثيلين تير فيثالات)

أسماء محمود فهيـم ، السيد محمد على الدين ياقـوت ، إيمان رجب* ، أحمد محمد فرج* و جلال عبد المعين نوار

قسم الكيمياء الخضراء – المركز القومى للبحوث – 33 ش البحوث بالدقى و *قسم الكيمياء – كلية العلوم – جامعة القاهرة – القاهرة – مصر .

إنّ لإعادة تدوير البلاستيك أهمية بالغة ودور مهم في الحد من نفاذ المصادر وتحقيق التنمية المستدامة وذلك بتأمين المواد الأولية من استغلال المخلفات بدلا من المواد الخام كما أن له دور مهم من الناحية البيئية وذلك بحماية الهواء و الماء من الملوثات حيث تجميعها وإعادة استعمالها بدلا من الحرق. ومن هذا المنطلق تم فى هذا البحث التعامل مع النفايات البلاستيكية ، وتحديدا زجاجات المياه البلاستيك ، وذلك عن طريق التحلل المائى لها بواسطة محلول هيدروكسيد الصوديوم وذلك للحصول على المادة الأولية للبوليمر (بولى إيثيلين تير فيثالات) وهى حمض التيرفيثاليك والتى تم الإستفادة منه فى تحضير العديد من المركبات العضوية الغير متجانسة الحلقة.

(a,b و 2و4- أوكساديازول (9 و 112 , (a,b وعلى هذا فقد تم تشييد مشتقات كل من اليوراسيل (6 و (15 بإستخدام مخلف البولى إيثيلين تير فيثالات (a, b و 3و4 – ترايازول (10 و 14) و ثياديازول 11 1 .

بالإضافة إلى ما سبق فإنه قد تم إجراء التقييم البيولوجي للمركبات المشيدة الجديدة كمضادات للميكروبات وكمضادات للأكسدة حيث أبدت بعض المركبات نشاطا ملحوظا يقترب من فاعلية المركبات القياسية. كما أثبتت النتائج وجود علاقة بين التركيب الكيميائى للمركبات الجديدة وفاعليتها البيولوجية.