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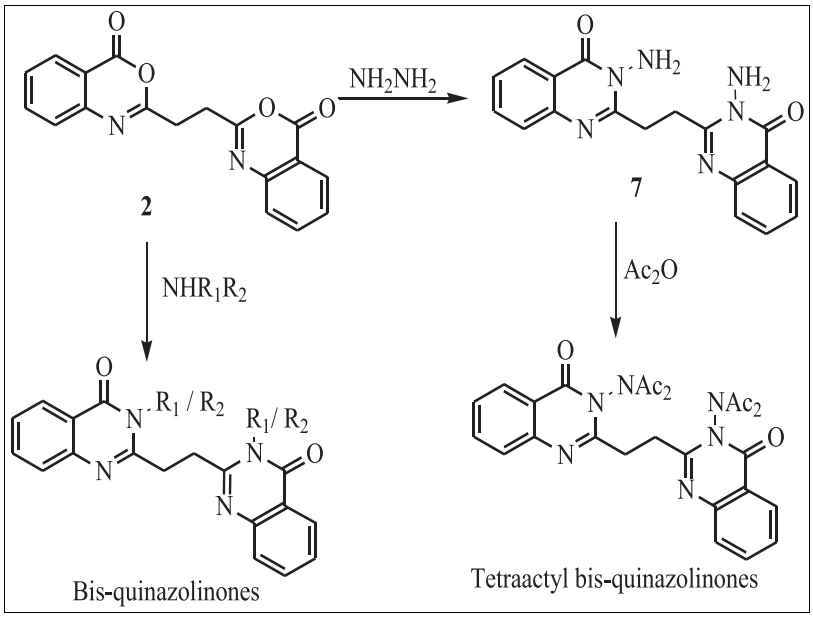

A novel bis benzoxazin-4-one derivative has been synthesized and utilized to construct a new series of bis quinazolin-4(3*H*)-one derivatives *via* the reactions with different nitrogen nucleophiles namely, primary amines (ammonia, ethanolamine, and 4-aminoantipyrine), secondary amines (morpholine and piperidine), diamine (o-phenylenediamine), hydrazine hydrate, and hydroxylamine. The insecticidal efficacy of newly synthesized compounds was also studied. The structural features of the synthesized compounds were assigned by spectral analysis.

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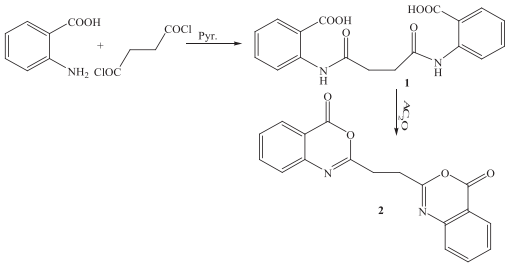
INTRODUCTION

Benzoxazinones (BAs) are natural products and represent part of the Gramineae defense system against insects, bacteria, and fungi [1–3]. Quinazolines exhibit a broad spectrum of biological activity including anti-inflammatory and antiplatelet aggregation [4], cytotoxicity [5], and cardiovascular activity [5,6]. These compounds also act as antihistaminic [7,8], antifungal, and antimalarial agents [9,10], and they exhibit potential psychomotor activity [11]. Pronounced pharmacological and biological activities of both benzoxazinones and quinazolines stimulated us to continue our previous work [12–16], for synthesis of novel heterocycles.

RESULTS AND DISCUSSION

The cotton leaf worm *Spodoptera littoralis* (Boisd) is a serious pest causing enormous losses to many economically important cultivated crops such as cotton, soybean, groundnut, tobacco, and vegetables [17]. It has been found that it causes 26–100% yield loss in the field [18]. The present study aimed to evaluate the newly synthesized bis quinazolin-4(3*H*)-one derivatives against cotton leaf worm. The bis-benzoxazin-4-one derivative **2** as a key starting material has been obtained in good yield by cyclization of 2({4-[(2-carboxyphenyl) amino]-4-oxobutanoyl} amino) benzoic acid **1**, which was obtained *via* the reaction of anthranilic acid and succinyl chloride in pyridine (Scheme 1). The structure of compound **2** was deduced from correct analytical and spectroscopic data. IR spectrum displays an absorption band for carbonyl group at 1756 cm^{-1}, and the ^{1}H-NMR (DMSO-d_6) revealed signals at δ (ppm) at: 7.90–7.30 (m, 8H, ArH), 1.90 (s, 4H, CH_2CH_2). Furthermore, the IE-MS of compound **2** shows the correct molecular ion peak at *m/z* = 320 (5%). (cf. the

Scheme 1



Experimental Section and Fig. 1). Aiming to synthesize novel quinazolinone derivatives which expected to have significant insecticidal efficiency, the benzoxazinone derivative **2** was subjected to react with different nitrogen nucleophiles. The new quinazolinones were obtained *via* ring opening of the benzoxazinone followed by ring closure under the influence of the reaction conditions.

Aminolysis of bis-benzoxazinone **2** with secondary amines namely, morpholine and piperidine, in refluxing dioxane provided bis-succinamide derivatives **3** and **4,**

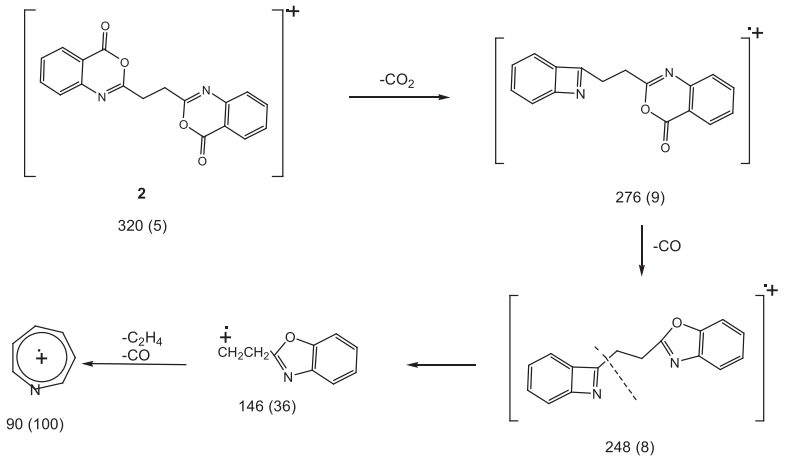
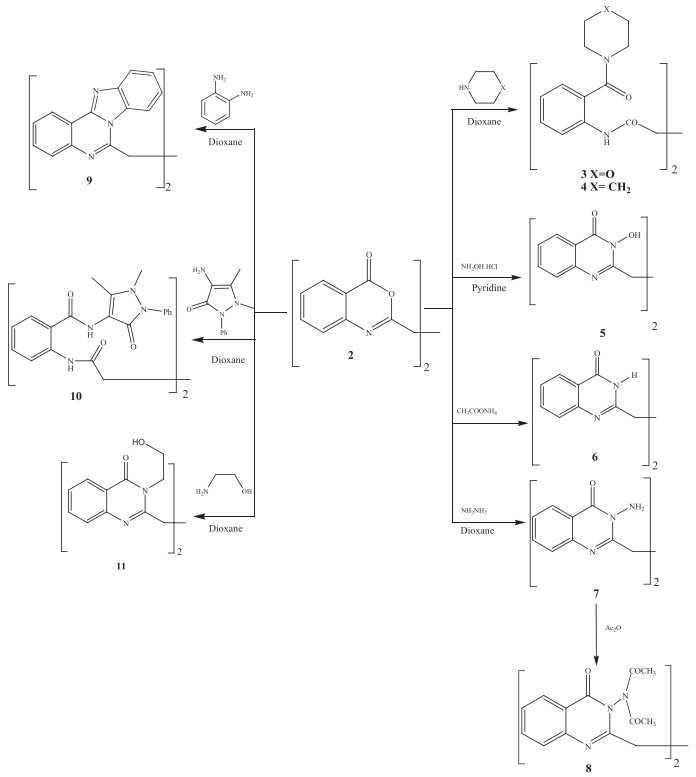


Figure 1. EI-MS fragmentation of compound 2.

Scheme 2



respectively. Upon treatment of bis-benzoxazinone **2** with hydroxylamine hydrochloride in boiling pyridine, the quinazolinone derivative **5** was produced (Scheme 2). Furthermore, fusion of **2** with ammonium acetate in sand bath at 230°C gave the quinazolinone derivative **6.** Hydrazinolysis of **2** by stirring with hydrazine hydrate (80%) in dioxane at room temperature for 1 h afforded the quinazolinone derivative **7**. The structural features of quinazolinone **7** were chemically substantiated beside the correct microanalytical and spectroscopic data from its acetylating with freshly distilled acetic anhydride to give the corresponding acetylating product **8**. Moreover, 1, 2-dibenzimidazol [1, 2-c] quinazolinoethan **9** was obtained *via* reaction of bis-benzoxazinone **2** with *o*-phenylenediamine in boiling dioxane (Scheme 2). However, bis-benzoxazinone **2** reacted with 4-aminoantipyrine in refluxing dioxane to give the amide derivative **10**. It is known that reaction of primary aromatic amines with benzoxazinones gives the corresponding quinazolinones *via* oxazinone ring cleavage by the nucleophilic attack of amino group on the carbonyl functionality followed by ring closure through attack of nitrogen-lactam form —CONH— on the double bond of the lactim form —N=C—OH with the elimination of water molecule. In case of 4-aminoantipyrine, the cyclization process seems difficult because of the bulk of antipyrine moiety, and hence, the amide **10** was formed instead of the corresponding quinazolinone. On the other hand, treatment of **2** with ethanolamine in refluxing dioxane afforded quinazolinone derivative **11** (Scheme 2).

CONCLUSION

The quinazolinone derivatives under investigation might be used in a proper formulation form to invent new potent pesticides to control cotton leaf worm *S. littoralis* in order to decrease the loss in the global cotton yield.

EXPERIMENTAL

Chemistry. All melting points were measured on Griffin and Geory melting point apparatus and are uncorrected. IR spectra were recorded on Pye Unicam SP 1200 spectrophotometer using KBr Wafer technique. ^{1}H-NMR spectra were determined on a Varian Gemini 200 MHz using TMS as internal standard (chemical shifts in δ-scale). EI-MS were measured on a Schimadzu-GC-MS, QP 1000 EX instrument operating at 70 eV. Elemental analyses were carried out at the microanalytical unit, Faculty of Science, Ain Shams University by using Perkin-Elmer 2400 CHN elemental analyzer, and satisfactory analytical data (±0.4) were obtained for all compounds. The homogeneity of the synthesized compounds was controlled by TLC [Using TLC aluminum sheets silica gel F_{254} (Merck)].

2({4-[(2-Carboxyphenyl)amino]-4-oxobutanoyl}amino)benzoic acid 1. A solution of succinyl chloride (1.55 g, 0.01 mol) in dry ether (10 mL) was added dropwise to a solution of anthranilic acid (2.74 g, 0.02 mol) in pyridine (20 mL) with stirring. The reaction mixture was stirred for further 30 min at room temperature then poured into ice-cold water and acidified with concentrated hydrochloric acid (35%). The obtained precipitate was collected by filtration and washed several times with water, dried, and crystallized from benzene to give **1** as buff crystals, m.p: 234–236°C, Yield: 80%, IR (KBr, ν): 3332 cm^{-1} (NH, OH) and 1672 cm^{-1} (C=O). ^{1}H-NMR (DMSO) δ: 11.39 (br. s, 2H, 2COOH, exchangeable with D_2O), 8.12–7.34 (m, 8H, ArH), 4.98 (br. s, 2H, 2NH, exchangeable), 2.10 (s, 4H, CH_2CH_2). MS [*m/z*, (%)]: 356 (missed), 276 (4)[M-($2H_2O$, CO_2)], 247 (3.8), 146 (30), 129 (5.5), 119 (100), and 90 (53). Anal. Calcd. for $C_{18}H_{16}N_2O_6$ (356): C, 60.67; H, 4.49; N, 7.87. Found: C, 60.64; H, 4.30; N, 7.90.

2-{2-(4-Oxo-4H-benzo[d][1,3]oxazin-2-yl)ethyl}-4H-benzo[d][1,3]oxazin-4-one 2. A mixture of **1** (3.56 g, 0.01 mol) and freshly distilled acetic anhydride (10 mL) was heated for 4 h on water bath. The reaction mixture was allowed to cool, and the obtained solid was separated by filtration, washed with petroleum ether 40–60°C, dried, and crystallized from toluene to give **2** as green crystals, m.p: 224–226°C, Yield: 85%, IR (KBr, ν): 1756 cm^{-1} (C=O) and 1641 cm^{-1} (C=N). ^{1}H-NMR (DMSO) δ: 7.90–7.30 (m, 8H, ArH), 1.90 (s, 4H, CH_2CH_2). MS [*m/z*, (%)]: 320 (5), 276 (9), 248 (8), 247 (15), 116 (4), 131 (2), and 90 (100). Anal. Calcd. for $C_{18}H_{12}N_2O_4$ (320): C, 67.50; H, 3.75; N, 8.75. Found: C, 67.64; H, 3.55; N, 8.90.

N^1,N^4-bis[2-(morpholine-4-carbonyl)phenyl]succinamide 3. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and morpholine (0.54 g, 0.006 mol) in dioxane (20 mL) was heated under reflux for 3 h. The solid formed after cooling was filtered off, dried, and then crystallized from dioxane to give **3** as white crystals, m.p: 159–160°C, Yield: 70%, IR (KBr, ν): 3226, 3182 cm^{-1} (NH) and 1682 cm^{-1} (C=O). ^{1}H-NMR (DMSO) δ: 9.67 (br. s, 2H, 2NH, exchangeable with D_2O), 7.46–7.23 (m, 8H, ArH), 3.43–3.38 (t, 8H, 2 CH_2OCH_2), 2.46 (t, 8H, 2 CH_2NCH_2), 2.04 (s, 4H, CH_2CH_2). MS [*m/z*, (%)]: 494 (2), 408 (6), 321 (80), 247 (14), 202 (85), 55 (100). Anal. Calcd. for $C_{26}H_{30}N_4O_6$ (494): C, 63.16; H, 6.07; N, 11.33. Found: C, 63.24; H, 6.15; N, 11.40.

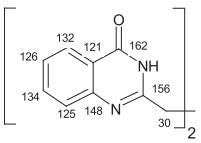
N^1,N^4-bis[2-(piperidine-4-carbonyl)phenyl]succinamide 4. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and piperidine (0.54 g, 0.006 mol) in dioxane (20 mL) was heated under reflux for 4 h. The solid formed after cooling was filtered off, dried, and then crystallized from dioxane to give **4** as white crystals, m.p: 160–162°C, Yield: 70%, IR (KBr, ν): 3200 cm^{-1} (NH) and 1691 cm^{-1} (C=O). ^{1}H-NMR (DMSO) δ: 8.91 (br. s, 2H, 2NH, exchangeable with D_2O), 7.64–7.31 (m, 8H, ArH), 3.43–3.38 (m, 12H, 2

$CH_2CH_2CH_2$), 2.82 (t, 8H, 2 CH_2NCH_2), 1.94 (s, 4H, CH_2CH_2). MS [*m/z*, (%)]: 490 (2), 406 (3), 321 (17), 247 (3), 202 (26), 162 (5), 84 (100). Anal. Calcd. for $C_{28}H_{34}N_4O_4$ (490): C, 68.57; H, 6.94; N, 11.43. Found: C, 68.49; H, 6.95; N, 11.40.

3-Hydroxy-2-[2-(3-hydroxy-4-oxo-3,4-dihydroquinazolin-2-yl) ethyl]quinazolin-4-(3H)-one 5. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and hydroxyl amine hydro chloride (0.43 g, 0.006 mol) in pyridine (20 mL) was heated under reflux for 8 h. The reaction mixture was concentrated and the solid formed after cooling was washed with water, filtered off, dried, and then crystallized from benzene to give **5** as yellow crystals, m.p: 260–262°C, Yield: 75%, IR (KBr, ν): 3115 cm^{-1} (OH) and 1649 cm^{-1} (C=O). ^{1}H-NMR (DMSO) δ: 8.12–7.19 (m, 8H, ArH), 2.12 (s, 2H, 2OH), 1.91 (s, 4H, CH_2CH_2). MS [*m/z*, (%)]: 350 (24), 333 (100), 317 (27), 160 (18), 145 (25). Anal. Calcd. for $C_{18}H_{14}N_4O_4$ (350): C, 61.71; H, 4.03; N, 15.99. Found: C, 61.68; H, 3.99; N, 15.89.

2-[2-(4-Oxo-3,4-dihydroquinazolin-2-yl)ethyl]quinazolin-4-(3H)-one 6. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and ammonium acetate (1.5 g) was fused on sand bath at 230°C for 2 h. The reaction mixture was stirred with hot water for 15 min, and the solid formed was filtered off, dried, and then crystallized from ethanol to give **6** as pale yellow crystals, m.p: 290–292°C, Yield: 75%, IR (KBr, ν): 3175 cm^{-1} (NH) ,1675 cm^{-1} (C=O), 1610 (C=N). ^{1}H-NMR (DMSO) δ: 11.77 (s, 2H, 2NH, exchangeable with D_2O), 8.43–7.11 (m, 8H, ArH), 1.92 (s, 4H, CH_2CH_2). Anal. Calcd. for $C_{18}H_{14}N_4O_2$ (318): C, 67.92; H, 4.40; N, 17.61. Found: C, 67.88; H, 4.33; N, 17.65.

^{13}C NMR for compound **6**



MS [*m/z*, (%)]: 318 (5), 173 (4), 145 (4), 103 (8), 91 (7), 57 (100).

3-Amino-2-[2-(3-amino-4-oxo-3,4-dihydroquinazolin-2-yl)ethyl] quinazolin-4-(3H)-one 7. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and hydrazine hydrate (0.31 g, 0.006) in dioxane (20 mL) was stirred for 1 h. The solid formed was filtered off, dried, and then crystallized from dioxane to give **7** as white crystals, m.p: over 300°C, Yield: 90%, IR (KBr, ν): 3316, 3204 cm^{-1} (NH_2), 1672 cm^{-1} (C=O), 1628 (C=N). MS [*m/z*, (%)]: 348 (45), 332 (56), 317 (16), 158 (10), 144 (24), 173 (14), 198 (15), 186 (100). ^{1}H-NMR (DMSO) δ: 8.23–7.34 (m, 8H, ArH), 2.12 (s, 4H, CH_2CH_2), (2H, NH_2, hided under DMSO protons signals). Anal. Calcd. for $C_{18}H_{16}N_6O_2$ (348): C, 62.07; H, 4.59; N, 24.14. Found: C, 62.16; H, 4.53; N, 24.22.

3-Diacetylamino-2-[2-(3-diacetylamino-4-oxo-3,4-dihydro quinazolin-2-yl)ethyl] quinazolin-4-(3H)-one 8. A mixture of quinazoline **7** (1.05 g, 0.003 mol) and freshly distilled acetic anhydride (10 mL) was heated under reflux for 4 h. The solid formed after evaporate the solvent was filtered off, washed with petroleum ether 40–60, dried, and then crystallized from ethanol to give **8** as blue crystals, m.p: 236–238°C, Yield: 80%, IR (KBr, ν, cm^{-1}): 1737, 1702 cm^{-1} (C=O), 1606 (C=N). MS [*m/z*, (%)]: 516 (30), 416 (25), 374 (78), 332 (23), 274 (11), 186 (63), 56 (100). ^{1}H-NMR (DMSO) δ: 8.11–7.46 (m, 8H, ArH), 2.49 (s, 6H, $2CH_3CO$), 2.04 (s, 4H, CH_2CH_2). Anal. Calcd. for $C_{26}H_{24}N_6O_6$ (516): C, 60.46; H, 4.65; N, 16.27. Found: C, 60.50; H, 4.59; N, 16.30.

1, 2-Dibenzimidazol[1,2-c]quinazolinoethane 9. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and o-phenylenediamine (0.675 g, 0.006 mol) in dioxane (20 mL) was heated under reflux for 8 h. The solvent was removed under reduced pressure, and the solid formed was filtered off, dried, and then crystallized from dioxane to give **9** as white crystals, m.p: over 340°C, Yield: 80%, IR (KBr, ν): 3086 cm^{-1} (CH—Ar), 1618 cm^{-1} (C=N). MS [*m/z*, (%)]: 464 (8), 229 (33), 172 (20), 77 (20), 68 (33), 64 (45), 56 (100). ^{1}H-NMR (DMSO) δ: 7.42–7.07 (m, 16H, ArH), 2.05 (s, 4H, CH_2CH_2). Anal. Calcd. for $C_{30}H_{20}N_6$ (464): C, 77.59; H, 4.31; N, 18,10. Found: C, 77.49; H, 4.49; N, 17.98.

N^1,N^4-bis[2-(4-aminoantipyrine-4-carbonyl)phenyl]succinamide 10. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and 4-aminoantipyrine (1.22 g, 0.006 mol) in dioxane (20 mL) was heated under reflux for 8 h. The solvent was removed under reduced pressure, and the solid formed was filtered off, dried, and then crystallized from dioxane to give **10** as yellow crystals, m.p: 240–242°C, Yield: 78%, IR (KBr, ν): 3253 cm^{-1} (NH), 1670 cm^{-1} (C=O). ^{1}H-NMR (DMSO) δ: 9.22 (s, 2H. NH exchangeable with D_2O), 8.95 (s, 2H. NH exchangeable with D_2O), 8.23–8.04 (m, 8H, ArH), 7.92–7.34 (m, 10H, ArH), 2.2 (s, 12H, $4CH_3$) 1.95 (s, 4H, CH_2CH_2). MS [*m/z*, (%)]: 728[M^{+2}](35), 295 (35), 109 (36), 71 (26), 64 (100). Anal. Calcd. for $C_{40}H_{38}N_8O_6$ (726): C, 66.11; H, 5.23; N, 15.43. Found: C, 65.88; H, 5.51; N, 15.42.

3-(2-Hydroxyethyl)-2-{2-[3-(2-hydroxyethyl)-4-oxo-3,4-dihydroquinazolin-2-yl]ethyl}qunazolin-4(4H)-one 11. A mixture of benzoxazinone **2** (1.0 g, 0.003 mol) and ethanol amine (0.49 g, 0.006 mol) in dioxane (20 mL) was heated under reflux for 5 h. The solvent was removed under reduced pressure, and the solid formed was filtered off, dried, and then crystallized from ethanol to give **11** as white crystals, m.p: 178–180°C, Yield: 78%, IR (KBr, ν): 3503 cm^{-1} (OH), 1674 cm^{-1} (C=O). ^{1}H-NMR (DMSO) δ: 8.12–7.72 (m, 8H ArH), 4.12–3.82 (m, 8H, $2NCH_2$—CH_2O) 3.64 (s, 2H, OH), 1.89 (s, 4H CH_2—CH_2) MS [*m/z*, (%)]: 406 (9), 363 (47), 244 (17), 201 (35), 173 (44), 120 (100). Anal. Calcd. for $C_{22}H_{22}N_4O_4$ (406): C, 65.01; H, 5.42; N, 13.79. Found: C, 64.99; H, 5.32; N, 13.72.

METHOD OF BIOASSAY TECHNIQUE

The leaf dipping technique was adopted on the 4th instar larvae to simulate the actual treatments under field conditions [19]. A stock solution of each chemical was freshly prepared. Subsequent water dilution was made to achieve concentrations of 100, 200, 400, and 800 ppm. Discs of 5-cm diameter were made from cotton leaves collected from unsprayed fields. The leaves were washed, dried, immersed in a test solution for 10 s and allowed to dry on corrugated kitchen foil at ambient temperature for 1–1.5 h. Leaf discs immersed in distilled water as control treatment. On drying, the leaf discs were placed in individual petri dishes (9-cm diameter). Each treatment (concentration) was replicated 3 times, including water solvent control. Ten of 4th instar larvae were placed on each leaf disc (replication), and thus the total number of tested larvae per concentration was 30. The bioassay was kept at temperature 25 ± 2°C and 65 ± 5% relative humidity to determine the different biological criteria. Mortality was assessed after 72-h exposure to tested compounds; the LC_{50} and toxicity index were also estimated.

MATERIALS AND METHODS

The cotton leaf worm strain in the present study was taken from field colony and reared in central agricultural pesticide laboratory. This strain was obtained from Sharkia governorate. The strain was kept under laboratory conditions at 25 ± 2°C and 65 ± 5% relative humidity away from any chemical pressure.

CHEMICALS USED

The newly synthesized quinazolinone derivatives.

Table 1 and Figures 2 and 3 showed the insecticidal activity data of some synthesized quinazolinone derivatives. These data revealed that as the concentration of tested compound increases the insecticidal activity increases. From this table we detected that the acetylating quinazolinone derivative **(8)** was the more effective compound where it recorded LC_{50} at 147.895 ppm followed by compounds **(9)**, **(7)**, **(11)**, **(5)**, **(4)**, and **(10)** with LC_{50} 160.687, 178.317, 283.418, 351.822, 353.589, and 917.032 ppm, respectively. On the other hand compound

Table 1

The insecticidal activity of some synthesized quinazolinone derivatives.

		Concentration ppm						
		100	200	400	800	LC_{50}	Slope	Toxicity index
% mortality	4	6.67	26.67	50.0	86.67	353.589	2.784	41.83
	5	16.67	33.33	50.0	76.67	351.822	1.829	40.04
	6	0.0	16.67	30.0	43.33	1048.957	1.318	14.09
	7	33.33	53.33	70.0	93.33	178.317	1.964	82.94
	8	40.0	56.67	76.67	100.0	147.895	1.648	100
	9	36.67	56.67	73.33	90.0	160.687	1.74	92.04
	10	23.33	43.33	60.0	73.33	917.032	1.481	16.13
	11	3.33	16.67	30.0	43.33	283.418	1.623	52.18

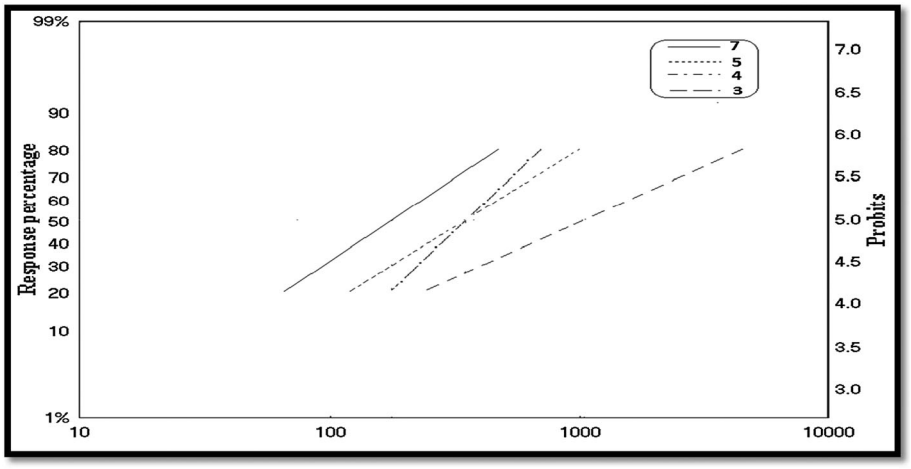


Figure 2. Ldp line of synthesized (4–7) quinazolinone derivatives.

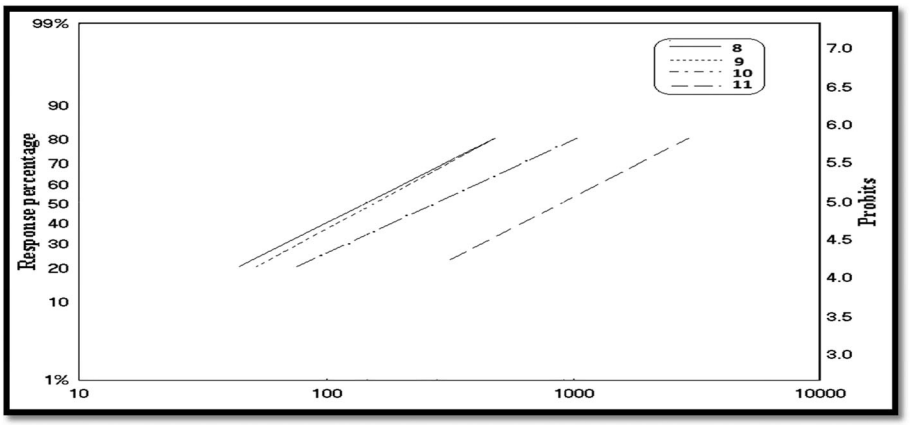


Figure 3. Ldp line of synthesized **8–11** quinazolinone derivatives.

(6) was the least efficient compound among the other tested quinazolinone derivatives against cotton leaf worm *S. littoralis* insect where its LC_{50} is 1048.957 ppm.

The tested quinazolinone derivatives are a new anthranilic diamide insecticide, which effectively controls pest insects, and has been shown to be effective against insects that have developed resistance to older classes of chemistry.

Anthranilic diamides selectively bind to ryanodine receptors in insect muscles resulting in an uncontrolled release of calcium from internal stores in the sarcoplasmic reticulum [20,21] causing impaired regulation of muscle contraction leading to feeding cessation, lethargy, paralysis, and death of target organisms.

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