

Functionally Substituted 1,2,4-Triazoloquinoline, Tetrahydro-quinolinone and Benzotriazolotriazine Derivatives from 5,5-Dimethylcyclohexan-1,3-Dione and Arylmethylenemalonic Acid Derived Michael Acceptors

A. A. Elagamey, H. H. Nawar and F. M. A. El-Taweel*

Department of Chemistry, Faculty of Science, Damietta University, New Damietta, Egypt.

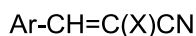
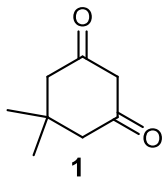
SEVERAL new functionally [1,2,4]triazolo[1,5-*a*]quinoline-4-carbonitriles (8a-c) and [1,2,4] triazolo[1,5-*a*]quinoline-4-carbox-amides 12a,b were prepared from *N'*-(5,5-dimethyl-3-oxocyclohexen-1-yl)benzohydrazide (5) and arylmethylene nitriles 2a-c and 2d,e, respectively. Reacting 2-oxo-3*H*-indol-3-ylidene malonic acid derivatives 3a,b with 5 afforded dioxospiro [3*H*-indole-3,4'-quinoline]-3'-carbonitrile (15) and 2-oxo-[1*H*] indol-3-ylcyanoacetate (17), respectively. Treating 5 with the enaminoester 18 gave hexahydro-7,7-dimethyl-2,5-dioxoquinolin-3-yl]benzamide (20). Coupling of 5 with the aryl diazonium salts and heteroaryl diazonium salts gave the hydrazones (21) and tetrahydro-[1,2,4]triazolo[5,1-*c*] [1,2,4] benzotriazine-6-ylidene)benzohydrazide (26).

5,5-Dimethylcyclohexan-1,3-dione (1) is a versatile reagent that has been extensively used for preparation of a variety of aromatic and heteroaromatic compounds⁽¹⁻⁷⁾. These compounds are interesting as potential biodegradable agrochemicals⁽⁸⁾, pharmaceuticals⁽⁹⁾, as nonpeptide human deficiency virus (HIV) protease inhibitors, blood anticoagulants⁽¹⁰⁾ and as plant growth regulating agents⁽¹¹⁾. Other derivatives such as hexahydroquinoline derivatives bearing an acyl group on C-3 have been exhaustively studied as interesting calcium antagonist modulators⁽¹²⁾.

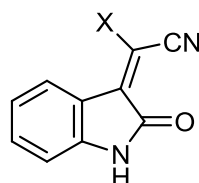
In the past few years, we have been involved in a program aimed at developing new accesses to the synthesis of functionally substituted heterocycles using readily available starting components.

In continuation of this effort, we describe here new procedures for the synthesis of different heterocycles. The present work resulted in the formation of functionally substituted quinoline, 1,2,4-triazoloquinoline and benzotriazolotriazine derivatives of potential biological importance utilizing 5,5-dimethyl-1,3-cyclohexanedione (1) and activated nitriles 2a-e and 3a,b as starting materials.

*E-mail: fathyeltaweel@yahoo.com



- 2a, Ar = Ph ; X = CN
 b, Ar = C₆H₄OCH₃(*p*) ; X = CN
 c, Ar = 2-thienyl ; X = CN
 d, Ar = C₆H₄OH(*p*) ; X = CONH₂
 e Ar = C₆H₄Br(*p*) - ; X = CONH₂



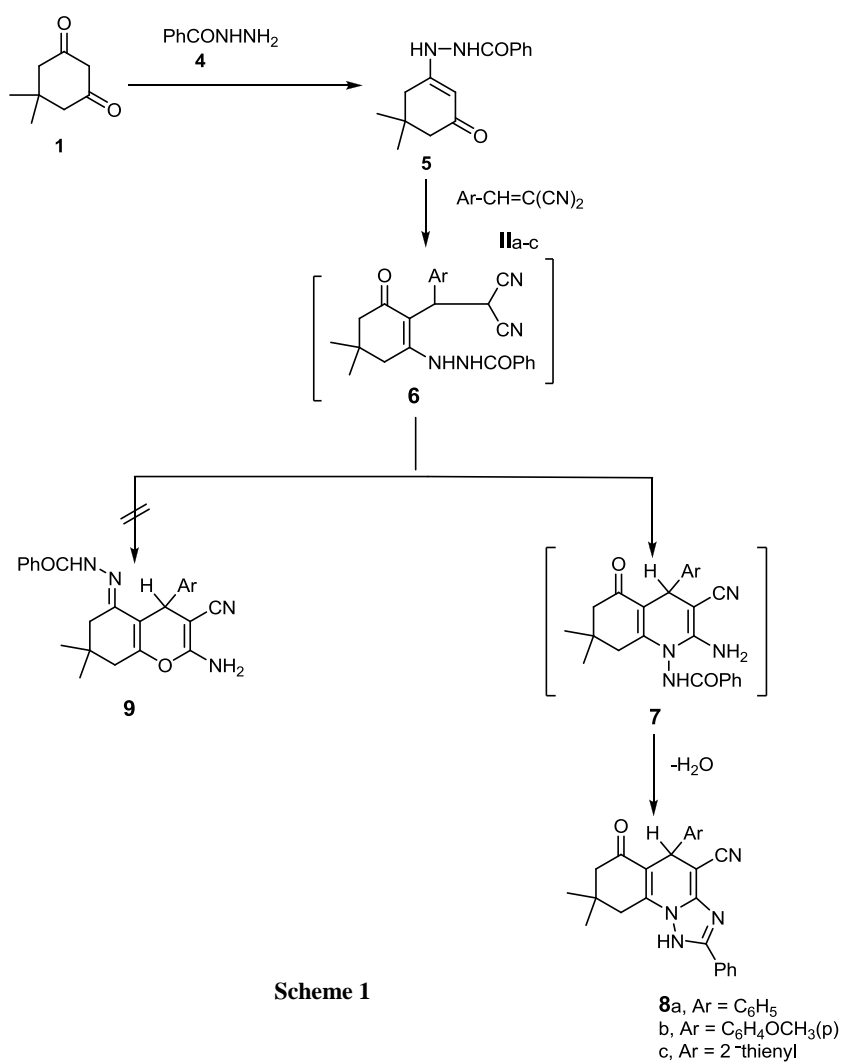
- 3a**, X = CN
 b, X = CO₂C₂H₅

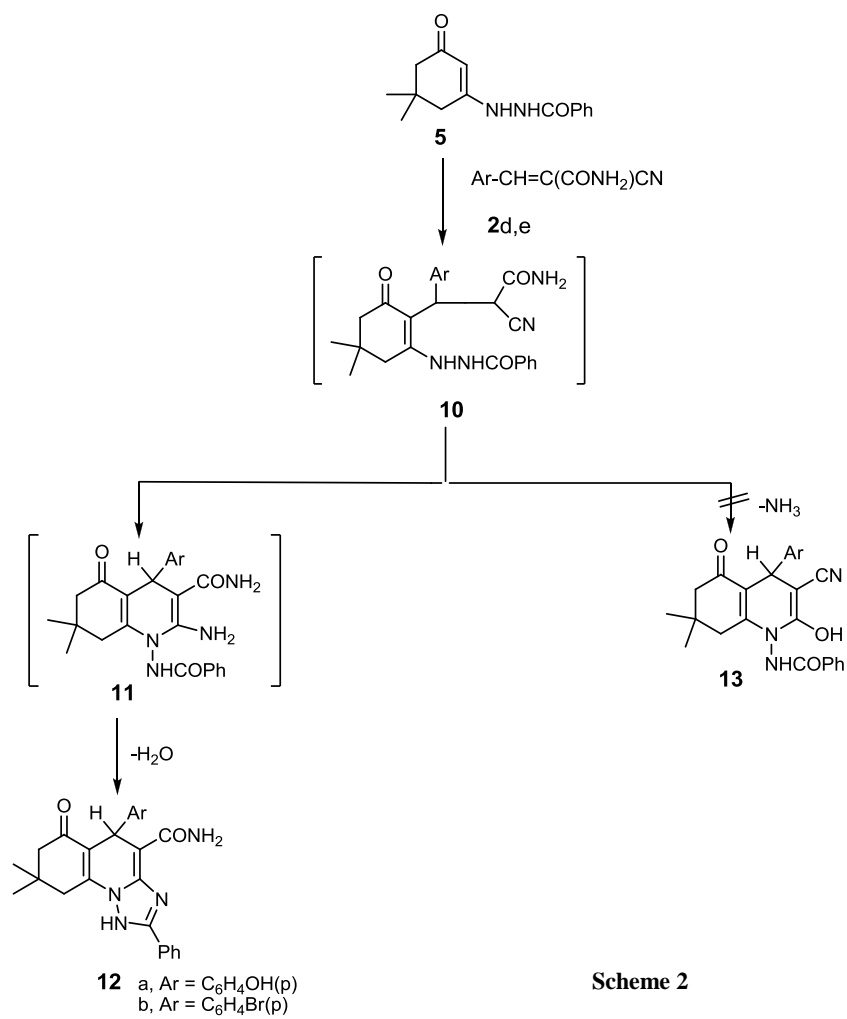
N'-(5,5-Dimethyl-3-oxocyclohexen-1-yl)benzohydrazide (5)¹⁵, prepared *via* condensing 5,5-dimethylcyclohexane-1,3-dione (1) with benzoylhydrazine (4), readily reacted with the arylmethylenemalononitriles (2a-c) in ethanol containing piperidine as catalyst to afford 5-aryl-1,5,6,7,8,9-hexahydro-8,8-dimethyl-6-oxo-2-phenyl[1,2,4]triazolo[1,5-*a*]quinoline-4-carbonitriles (8a-c) rather than *N'*-(2-amino-4-aryl-3-cyano-5,6,7,8-tetrahydro-7,7-dimethyl-4*H*-chromen-5-ylidene)benzohydrazide (9, Ar = phenyl). The triazoloquinoline structures 8 were preferred over the possible alternative chromene derivatives 9 based on elemental analyses and spectral data. The IR spectra revealed the presence of signals corresponding to carbonyl functions at $\tilde{\nu} = 1690 - 1694 \text{ cm}^{-1}$. Signals at similar positions described for the formation of analogous systems have been previously reported⁽¹⁰⁾.

Also, the ¹H-NMR spectrum of 8c showed the presence of a signal at $\delta = 3.54 \text{ ppm}$ for one proton linked to sp³ carbon. Compounds 8 were proposed to be formed *via* Michael type addition of the active methylene carbon in 5 across the activated double bond in the arylmethylenemalononitriles 2 to give Michael adducts 6, which cyclized to 7. The latter readily eliminate water to afford the final isolable products 8a-c (*c.f.* Scheme 1).

Similarly, benzohydrazide 5 reacted with the arylmethylenecyanoacetamides 2d,e in refluxing ethanol and in presence of piperidine to yield 5-aryl-1,5,6,7,8,9-hexahydro-8,8-dimethyl-6-oxo-2-phenyl-[1,2,4]triazolo[1,5-*a*]quinoline-4-carbox-amides (12a,b) rather than 4-aryl-1-(benzoylamino)-1,4,5,6,7,8-hexahydro-2-hydroxy-7,7-dimethyl-5-oxoquinoline-3-carbonitriles (13). Structures 12a,b were established for the reaction products based on their IR spectra which

clearly indicate the absence of any signals corresponding to cyano groups. Compounds 12a,b were formed probably *via* the sequence demonstrated in Scheme 2.





Compound 5 also reacted with (2-oxoindolin-3-ylidene)malononitrile (3a) in ethanol and in presence of piperidine as catalyst to give 2'-amino-1'-benzoylamino - 1,2,5',6', 7', 8' - hexahydro - 7',7'- dimethyl -2,5'-dioxospiro[3*H*-indole - 3, 4'- quinoline] -3' - carbonitrile (15) rather than *N'* -{2-amino-3-cyano-1', 2', 5, 6, 7, 8 – hexahydro - 7, 7-dimethyl-2'-oxospiro[4*H*-chromene-4,3'-[3*H*]-indole]-5-ylidene}benzohydrazide (16) as elucidated by its IR spectrum which showed the presence of a carbonyl group for a cyclic ketone at 1694 cm⁻¹.

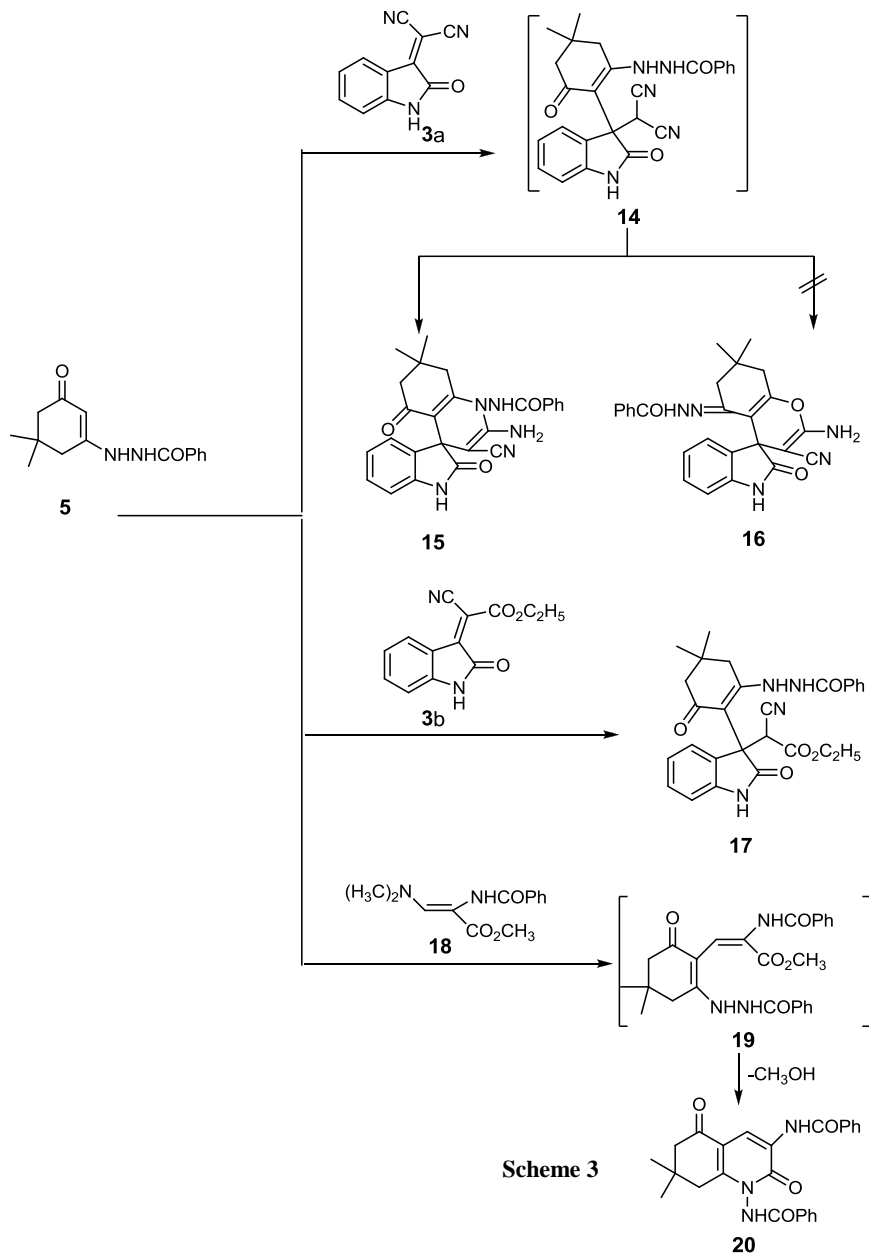
Reacting *N*-(5,5-dimethyl-3-oxocyclohex-1-enyl)benzohydrazide (5) with ethyl (2-oxoindolin-3-ylidene)cianoacetate (3b) in ethanol catalysed by piperidine resulted in the formation of ethyl {3-[1-(benzoylhydrazino)-2,3-dihydro- 5, 5- dimethyl- 3- oxo-cyclohex - 2- enyl] - 2-oxo-[1*H*]indol-3yl}cianoacetate (17).

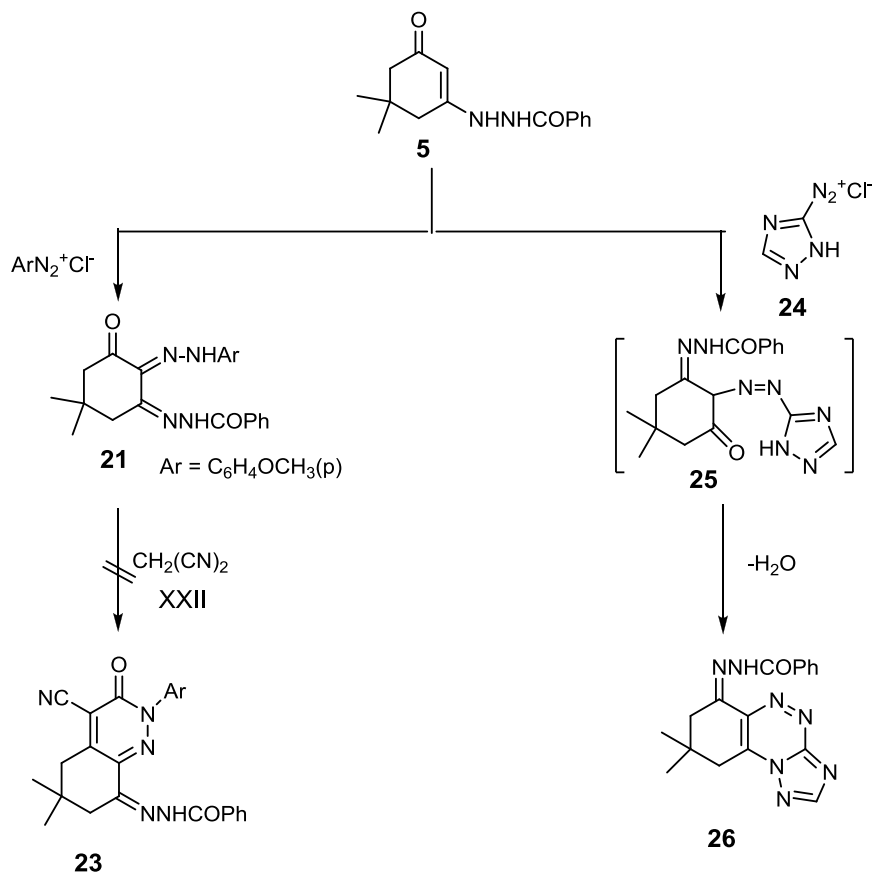
The reactivity of 5 towards enaminoesters such as methyl 2-benzoylamino-3-dimethylaminopropenoate (18) was also studied. Thus, compound 5 reacted with 18 in refluxing acetic acid to afford *N*-[(1-benzoylamino)-1,2,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxoquinolin-3-yl]benzamide (20). Compound 20 was supported by elemental analysis and spectral data (*c.f.* Experimental).

Compound 20 was proposed to be formed by initial condensation of 5 at C-2 with 18 to give the intermediate 19 *via* elimination of one molecule of dimethylamine, followed by cyclization through methanol elimination (*c.f.* Scheme 3).

The reactivity of 5 towards aryl and heteroaryl diazonium salts was also investigated. Thus, compound 5 was coupled with arene diazonium chloride to give *N'*- [2- (4- methoxyphenyl)hydrazono-5,5-dimethyl-3-oxocyclohexylidene]-benzohydrazide (21). Trials to prepare *N'*-[4-cyano-2,3,5,6,7,8-hexahydro-2-(4-methoxyphenyl)-6,6-dimethyl-3-oxo-cinnolin-8-ylidene]benzohydrazide(23), *via* condensation of 21 with malononitrile, failed.

Further, compound 5 was coupled with the diazonium salt 24 to afford *N'*-(8, 8-dimethyl -6, 7, 8, 9- tetrahydro - [1,2,4] triazolo [5, 1- c] [1,2,4] benzotriazine-6-ylidene)benzohydrazide (26). Compound 26 was supposed to be obtained through intermediate 25, which readily underwent cyclization utilizing the highly nucleophilic triazole NH⁽¹³⁾ (*c.f.* Scheme 4).





Scheme 4

Experimental

Melting points have been determined on a Griffin & George (London) MBF 010T apparatus and have not been corrected. Recorded yields correspond to the pure products. IR (KBr) spectra were recorded on a Perkin Elmer SP-880 spectrophotometer, at Faculty of Science, Damietta University. $^1\text{H-NMR}$ spectra were measured on a Varian 300 MHz instrument at Cairo University and a 500 MHz spectrometer at Minnesota (U.S.A) University in DMSO-d_6 as solvent using TMS as an internal standard (Chemical shifts are reported in δ/ppm units). Mass spectra were measured on a Finnigan MAT GS/MS INCOL XL. Microanalyses were performed on a LECOCHN-932 instrument in the Microanalytical Data Unit, Mansoura University.

N'-(5,5-Dimethyl-3-oxo-cyclohexen-1-yl)benzohydrazide (5)

A solution of 5,5-dimethylcyclohexan-1,3-dione (1, 0.01 mol) and benzoylhydrazine (4, 0.01 mol) in ethanol (50 ml) containing piperidine (0.1 ml) was refluxed for 1hr then left to cool. The precipitate formed was collected by filtration and recrystallized from ethanol to give 5 as colorless crystals (85%), m.p. 230-232 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3196 (NH), 1674 (CO). ^1H NMR (DMSO- d_6): δ/ppm = 1.01 (s, 6H, 2CH₃), 2.02 (s, 2H, CH₂), 2.49 (s, 2H, CH₂), 3.36 (s, 2H, CH₂), 4.94 (s, 1H, CH), 7.49-7.88 (m, 5H, aromatic protons), 10.52 (s, 1H, NH).- C₁₅H₁₈N₂O₂ (258.32): Calcd. C, 69.74; H, 7.02; N, 10.84%. Found C, 69.81; H, 6.67; N, 10.69%. Mol.mass 258 (MS).

General procedure for preparation of 5-aryl-1,5,6,7,8,9-hexahydro-8,8-dimethyl-6-oxo-2-phenyl-[1,2,4] triazolo [1,5-a]quinoline-4-carbonitriles (8a-c)

A solution of compound 5 (0.01 mol) in ethanol (50 ml) was refluxed for 3 hr and then left to cool at room temperature. The solids formed were collected by filtration, recrystallized from the proper solvents and then identified as 8a-c. ^1H NMR spectra of 8a,b could not be obtained due to the insolubility of the samples in DMSO.

1,5,6,7,8,9-Hexahydro-8,8-dimethyl-6-oxo-2,5-diphenyl-[1,2,4]triazolo[1,5-a]quinoline-4-carbonitrile (8a)

Colorless crystals from ethanol /1,4-dioxane, yield(70%), m.p. 270-272 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3446 (NH), 2230 (conjugated CN), 1694 (CO), 1606 (C=N).- C₂₅H₂₂N₄O (394.18): Calcd. C, 76.12; H, 5.62; N, 14.20%. Found C, 75.92; H, 5.13; N, 14.33%. Mol. mass 394 (MS).

1,5,6,7,8,9-Hexahydro-5-(4-methoxyphenyl)-8,8-dimethyl-6-oxo-2-phenyl-[1,2,4] triazolo[1,5-a]quinoline-4-carbonitrile (8b)

Colorless crystals from ethanol, yield (75%), m.p. 266-268 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3367(NH), 2228 (conjugated CN), 1691(CO), 1655(C=N).- C₂₆H₂₄N₄O₂ (424.49): Calcd. C, 73.56; H, 5.70; N, 13.20%. Found C, 73.41; H, 5.54; N, 13.41%. Mol. mass 424 (MS).

1,5,6,7,8,9-Hexahydro-8,8-dimethyl-6-oxo-2-phenyl-5-(thiophen-2-yl)-[1,2,4] triazolo[1,5-a]quinoline-4-carbonitrile (8c)

Yellow crystals from ethanol/1,4-dioxane, yield (80%), m.p. 275-277 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3366 (NH), 2230 (conjugated CN), 1690 (CO), 1603 (C=N). ^1H NMR (DMSO- d_6): δ/ppm = 1.15 (s, 6H, 2CH₃), 2.49 (s, 2H, CH₂), 2.59 (s, 2H, CH₂), 3.54 (s, 1H, 5-H), 7.17-8.30 (m, 9H, 8 aromatic protons and 1H, NH).- C₂₃H₂₀N₄SO (400.14): Calcd C, 68.98; H, 5.03; N, 13.99%. Found C, 68.83; H, 5.21; N, 14.05%. Mol. mass 400 (MS).

5-Aryl-1,5,6,7,8,9-hexahydro-8,8-dimethyl-6-oxo-2-phenyl-[1,2,4]triazolo[1,5-a]quinoline-4-carboxamides (12a,b)

A sample of compound 5 (0.01 mol) in ethanol (50 ml) containing piperidine (0.1 ml) was heated with 0.01 mol each of arylmethylene cyanoacetamides 2d,e under reflux for 3 hr. The reaction mixture was left to cool at room temperature.

The precipitates were collected by filtration, recrystallized from ethanol and identified as 12a,b. ^1H NMR spectra could not be recorded due to solubility reasons.

1,5,6,7,8,9-Hexahydro-5-(4-hydroxyphenyl)-8,8-dimethyl-6-oxo-2-phenyl-[1,2,4] triazolo[1,5-a]quinoline-4-carboxamide (12a)

Orange crystals from ethanol, yield (60%), m.p. 285-287 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3372, 3302, 3177 (NH₂, NH), 1696 (CO), 1639 (CO amide). - C₂₅H₂₄N₄O₃ (428.18) Calcd. C 70.08, H 5.65, N 13.08%. Found C 70.13, H 5.70, N 13.23%. Mol. mass 428 (MS).

5-(4-Bromophenyl)-1,5,6,7,8,9-hexahydro-8,8-dimethyl-6-oxo-2-phenyl [1,2,4] triazolo [1,5-a] quinoline-4-carboxamide (12b)

Yellow crystals from ethanol, yield (65%), m.p. 290-292 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3443, 3409, 3346 (NH₂, NH), 1689 (CO), 1663 (CO amide).- C₂₅H₂₃BrN₄O₂ (490.10): Calcd. C, 61.11; H, 4.72; N, 11.40%. Found C, 61.25; H, 4.63; N, 11.51%. Mol. mass 400 (MS).

General procedure for preparation of 2'-amino-1'-(benzoylamino)-1,2,5',6',7,8-hexahydro-7',7'-dimethyl-2,5'-dioxospiro [3H-indole-3,4'-quinoline]-3'-carbonitrile (15) and ethyl {3-[1-(benzoylhydrazino)-5,5-dimethyl-3-oxocyclohex-2-enyl]-2-oxo-1,2-dihydro-3H-indol-3-yl}cyanoacetate (17)

A suspension of compound 5 (0.01 mol) and (0.01 mol) of (2-oxoindolin-3-ylidene)cyanoacetate (3b) in ethanol (50 ml) containing piperidine (0.1 ml) was refluxed for 1hr. The solids deposited upon heating were collected by filtration, recrystallized and then identified as 15 and 17, respectively.

2'-Amino-1'-(benzoylamino)-1,2,5',6',7,8'-hexahydro-7',7'-dimethyl-2,5'-dioxospiro [3H-indole-3,4'-quinoline]-3'-carbonitrile (15)

Colorless crystals from ethanol / dimethylformamide, yield (70%), m.p. >300 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3478, 3333 (NH₂, NH), 2187 (conjugated CN), 1716 (CO), 1694 (CO), 1642 (NH₂). ^1H NMR (DMSO-d₆): δ/ppm = 0.83(s, 3H, CH₃), 0.90 (s, 3H, CH₃) 2.04 (s, 2H, CH₂), 2.85 (s, 2H, CH₂), 6.45 (s, 2H, NH₂), 6.76-8.02 (m, 9H, aromatic protons), 10.24 (s, 1H, NH), 11.3 (br, s, 1H, NH).- C₂₆H₂₃N₅O₃ (453.49): Calcd. C, 68.86; H, 5.11; N, 15.44%. Found C, 68.75; H, 5.05; N, 15.36%. - Mol. mass 453 (MS).

Ethyl {3-[1-(benzoylhydrazino)-5,5-dimethyl-3-oxocyclohex-2-enyl]-2-oxo-1,2-dihydro-3H-indol-3-yl}cyanoacetate (17)

Yellow crystals from ethanol, yield (60%), m. p. 240 - 242 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3480, 3381, 3277 (NH), 2162 (CN), 1697 (CO), 1638 (CO). ^1H NMR (DMSO-d₆): δ/ppm = 0.82 (s, 3H, CH₃), 0.86 (s, 3H, CH₃), 0.94 (t, 3H, CH₃), 1.57 (s, 2H, CH₂), 2.53 (s, 2H, CH₂), 3.69 (m, 2H, CH₂), 3.82(s, 1H, CH), 6.63-8.0 (m, 11H, 9 aromatic protons and 2H, 2NH), 9.97 (s, 1H, NH). -C₂₈H₂₈N₄O₅ (500.55) Calcd C 67.19, H 5.64, N 11.19%. Found C 67.22, H 5.66, N 11.21%. Mol. mass 500 (MS).

N-[1-(benzoylamino)-1,2,5,6,7,8-hexahydro-7,7-dimethyl-2,5-dioxoquinolin-3-yl]benzamide (20)

A solution of compound (5,0.01 mol) and (0.01 mol) of methyl 2-benzoylamino-3-dimethylaminopropenoate (18) in glacial acetic acid (15ml) was refluxed for 6 hr. The solvent was evaporated under reduced pressure and the residue was triturated with ethanol. The precipitate was collected by filtration and recrystallized from ethanol/dimethylformamide to give compound 20 as colorless crystals, yield (70 %), m.p. 264-266 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3312, 3191 (NH), 1685 (CO), 1651 (CO). ^1H NMR (DMSO- d_6): δ/ppm = 1.07 (s, 6H, 2CH₃), 2.47 (d, J = 16 Hz, 2H, CH₂), 2.71 (d, J = 16 Hz, 2H, CH₂), 7.54-8.05 (m, 10H, aromatic protons), 8.7 (s, 1H, 4-H), 9.53, 11.8 (2s, 2H, 2NH). - C₂₅H₂₃N₃O₄ (429.47): Calcd C, 69.92; H, 5.40; N, 9.78%. Found C, 69.85; H, 5.22; N, 9.80%. Mol. mass 429 (MS).

N'-[2-(4-methoxyphenyl)hydrazono-5,5-dimethyl-3-oxocyclohexylidene]benzohydrazide (21)

A solution of (0.01 mol) of compound 5 in ethanol / piperidine (50 ml) (1:1) was treated with (0.01 mol) of *p*-methoxyphenyl diazonium chloride (prepared from (0.01 mol) of sodium nitrite and the appropriate amount of *p*-methoxyaniline hydrochloride with stirring). The reaction mixture was left in the refrigerator over night. The precipitate formed was collected by filtration, recrystallized from ethanol to give red crystals of 21, m.p. 200-202 °C, yield (80 %). IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3446, 3206 (NH), 1739 (CO), 1656 (CO). ^1H NMR (DMSO- d_6): δ/ppm = 1.15 (s, 6H, 2CH₃), 2.49 (s, 2H, CH₂), 2.59 (s, 2H, CH₂), 3.81 (s, 3H, OCH₃), 7.17-8.30 (m, 11H, 9 aromatic protons and 2H, 2NH). - C₂₂H₂₄N₄O₃ (392.45): Calcd C, 67.33; H, 6.16; N, 14.28%. Found C, 68.83; H, 5.21; N, 14.05%. Mol. mass 392 (MS).

N'-(6,7,8,9-Tetrahydro-8,8-dimethyl-[1,2,4]triazolo[5,1-*c*][1,2,4]benzotriazin-6-ylidene)benzohydrazide (26)

An aqueous solution of sodium nitrite (0.7 gm, in 5 ml of water) was added to a cold (0°C) solution of 5-amino-1,2,4-triazole (0.01mol) in concentrated hydrochloric acid (5 ml). The resulting diazonium salt solution was then added to a cold solution of compound 5 (0.01 mol) in ethanol (50 ml) containing 2 gm of sodium acetate. The mixture was stirred at room temperature for 1 hr and the solid product was collected by filtration and recrystallized from ethanol to give orange crystals, yield (75%), m.p. 182-184 °C. IR: $\tilde{\nu}/\text{cm}^{-1}$ = 3197 (NH), 1673 (CO). ^1H -NMR (DMSO- d_6): δ/ppm = 1.01 (s, 6H, 2CH₃), 2.01 (s, 2H, CH₂), 2.49 (s, 2H, CH₂), 5.69 (s, 1H, 2-H), 7.50-7.88 (m, 5H, aromatic protons), 10.51 (s, 1H, NH). - C₁₇H₁₇N₇O (335.36): Calcd C, 60.88; H, 5.11; N, 29.24%. Found C, 60.76; H, 5.32; N, 29.11%. Mol. mass 335 (MS).

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طرق تحضير مشتقات جديدة من الترايازولوكينولين رباعي هيدرو
كينولين والبنزوترايازولوترايازين من 5,5-ثنائي ميثيل سيكلوهكسان-
1,3-دايون ومشتقات أريليدين حمض المالونيك

عبدالغني علي العجمي ، هاجر حسين نوار و فتحي محمد الطويل
قسم الكيمياء- كلية العلوم بدمياط – جامعة دمياط – مصر

حيث أن المركبات الحلقية الأروماتية غير متجانسة الحلقة ذات تأثير بيولوجي لأن مشتقاتها تستخدم مثلا كمبيدات زراعية فقد إتجهنا في هذا البحث إلي تحضير عدة مشتقات جديدة من الترايازولوكينولين ، الكينولون و الترايازولوبنزوترايازين من 5,5-ثنائي ميثيل سيكلوهكسان-1,3-دايون و مشتقات أريليدين حمض المالونيك.

تم إثبات التركيب البنائي للمركبات الجديدة باستخدام طرق التحليل العنصري و الطيفي مثل الأشعة تحت الحمراء، الأشعة فوق البنفسجية ، الرنين النووي المغناطيسي و طيف الكتلة.