Synthesis of a New Series of 4*H*-benzo[*h*]chromenes by a Multicomponent Reaction under Solvent-Free Microwave Conditions

Poursattar Marjani, Ahmad*+; Khalafy, Jabbar; Eslamipour, Paria
Department of Organic Chemistry, Faculty of Chemistry, Urmia University, Urmia, I.R. IRAN

Ahmadi Sabegh, Mehdi
Department of Chemistry, Ahar Branch, Islamic Azad University, Ahar, I.R. IRAN

ABSTRACT: *Multicomponent condensation of 1-naphthol, malononitrile, and arylglyoxals in the presence of Mg-Al hydrotalcite under solvent-free MicroWave (MW) conditions gave a new series of 2-amino-4-aroyl-4H-benzo[h]chromene-3-carbonitriles in high yields (70-89%). The structure of all products was elucidated by their FT-IR, ^{1}H-NMR, ^{13}C-NMR spectral data and microanalysis.*

KEYWORDS: *Arylglyoxals; 1-Naphthol; Malononitrile; Mg-Al hydrotalcite; 4H-benzo[h]chromenes; Microwaves; Multicomponent reaction.*

INTRODUCTION

The synthesis of 2-amino-4*H*-chromene derivatives has gained great interest in recent years as they are the main components of many naturally occurring products and due to their wide range of biological and medicinal applications such as anti-angiogenic [1] anti-bacterial [2], anti-cancer [3], anti-coagulant [4], anti-flammatory [5], anti-fungal [6], anti-HIV [7], anti-genotoxic [8], anti-oxidant [9], anti-tumor [10] and anti-viral [11] activities.

The synthesis of 2-amino-4*H*-chromenes by one-pot, three-component reaction using aromatic aldehydes, 1-naphthol, and malononitrile or dimedone under different catalytic conditions have been reported [12,13].

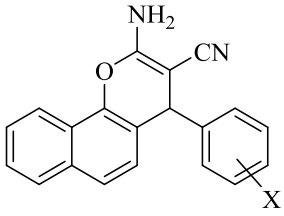
2-Amino-4*H*-benzo[*h*]chromene moiety has been considered as a promising and attractive substituent in the development of potential antitumor agents, for example, Ly290181 (Fig. 1, X= 3-NO_2) is a powerful antiproliferative agent [14]. In addition, unsubstituted, 6- and 7-substituted derivatives of 2-amino-4*H*-benzo[*h*]chromene nucleus have antitumor and cytotoxic activities [15]. (Fig. 1, X= 4-Cl, 2- or 4-NO_2).

In continuation of our interest in the synthesis of new heterocyclic compounds by one-pot, multicomponent reactions [16-24], herein, we report a novel multicomponent strategy for the synthesis of a new series of 2-amino-4-aroyl-4*H*-benzo[*h*]chromene-3-carbonitrile derivatives *via* a one-pot, the three-component reaction of 1-naphthol, malononitrile and arylglyoxals in the presence of Mg-Al hydrotalcite under solvent-free microwave (MW) conditions.

EXPERIMENTAL SECTION

Infrared spectra were recorded on a Thermo Nicolet Nexus 670 FT-IR instrument using KBr discs. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker Avance AQS

** To whom correspondence should be addressed.*
+ E-mail: a.pursattar@gmail.com ; a.poursattar@urmia.ac.ir
1021-9986/2018/4/51-57 7/$/5.07



X= 3- NO_2, 4-Cl, 2- or 4-NO_2

Fig. 1: Structure of 2-amino-4H-benzo[h]chromene derivatives with pharmaceutical activities.

300 MHz spectrometer at 300 and 75.5 MHz, respectively. Chemical shifts were measured in DMSO-d_6 as solvent relative to TMS as the internal standard. Melting points were measured on a Philip Harris C4954718 apparatus and are uncorrected. Elemental analyses were performed by using a Leco Analyzer 932. Microwave irradiation reactions were carried out in Yusch Heating Microwave oven (1000 W).

General procedure for the synthesis of 2-amino-4-aroyl-4H-benzo[h]chromene-3-carbonitriles 4a-g

A mixture of arylglyoxal (1 mmol), 1-naphtol (1 mmol) and malononitrile (1 mmol) in presence of Mg-Al hydrotalcite (72 mg) in a mortar was irradiated at 200 W, for an appropriate time (Table 1) under microwave condition. After completion of the reaction, as indicated by TLC, methanol (5 mL) was added to the reaction mixture. The reaction mixture was filtered to separate the catalyst. The filtrate was concentrated under vacuum and the solid residue was recrystallized from ethanol to give the desired products in high yields (70-89%).

2-Amino-4-benzoyl-4H-benzo[h]chromene-3-carbonitrile (4a)

White solid, 73%, mp 215-217 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ (ppm): 8.36 (d, J = 8.4 Hz, 1H, Ar), 8.08 (d, J = 8.4 Hz, 1H, Ar), 7.92-7.87 (m, 4H, Ar), 7.79 (bs, exchanged by D_2O addition, 1H, NH), 7.73 (bs, exchanged by D_2O addition, 1H, NH), 7.71 (t, J = 7.8 Hz, 1H, Ar), 7.64-7.54 (m, 4H, Ar), 5.74 (s, 1H, exchanged by D_2O addition, CH). ^{13}C-NMR (75.5 MHz, DMSO-d_6) δ (ppm): 165.4, 153.1, 149.3, 131.7, 130.1, 129.6, 127.9, 127.7, 126.5, 124.5, 123.6, 120.7, 119.9, 118.5, 117.2, 108.1, 79.2, 56.5, 18.6. FT-IR (KBr) ν_{max} (cm^{-1}): 3372, 3165, 3064, 2925, 2354, 2251, 2169, 1696, 1635, 1450, 1370, 1267, 1186, 1074, 802, 761, 663. Anal. Calcd for $C_{21}H_{14}N_2O_2$: C, 77.29; H, 4.32; N, 8.58; Found: C, 77.44; H, 4.20; N, 8.31%.

2-Amino-4-(4-bromobenzoyl)-4H-benzo[h]chromene-3-carbonitrile (4b)

Light yellow-white solid, 78%, mp 238-240 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ (ppm): 8.35 (d, J = 9 Hz, 1H, Ar), 8.08 (d, J = 8.1 Hz, 1H, Ar), 7.88 (t, J = 9 Hz, 2H, Ar), 7.83 (s, exchanged by D_2O addition, 2H, NH_2), 7.64 (t, J = 6 Hz, 1H, Ar), 7.68-7.61 (m, 4H, Ar), 7.73 (d, J = 6 Hz, 1H, Ar), 5.76 (s, 1H, CH). FT-IR (KBr) ν_{max} (cm^{-1}): 3438, 3319, 3268, 3198, 3063, 2934, 2425, 2378, 2252, 2082, 2024, 1912, 1680, 1482, 1378, 1262, 1223, 1136, 1081, 1008, 821, 742, 682, 570. Anal. Calcd for $C_{21}H_{13}BrN_2O_2$: C, 62.24; H, 3.23; N, 6.91; Found: C, 62.31; H, 3.09; N, 7.18%.

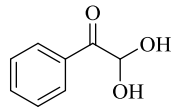
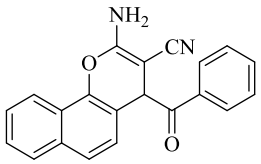
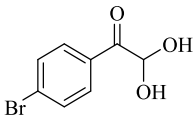
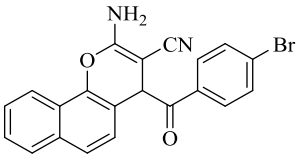
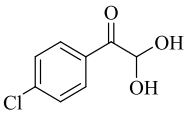
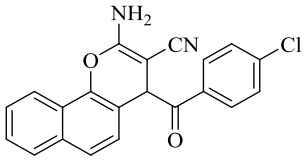
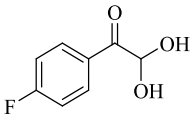
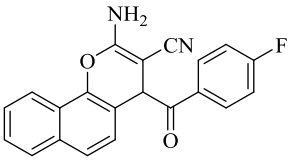
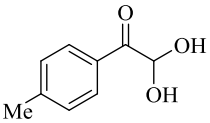
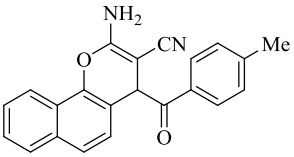
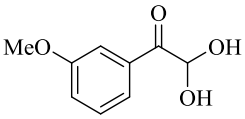
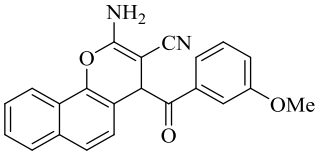
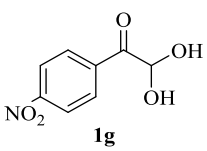
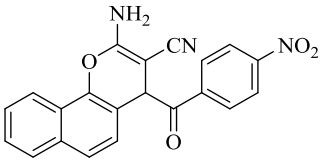
2-Amino-4-(4-chlorobenzoyl)-4H-benzo[h]chromene-3-carbonitrile (4c)

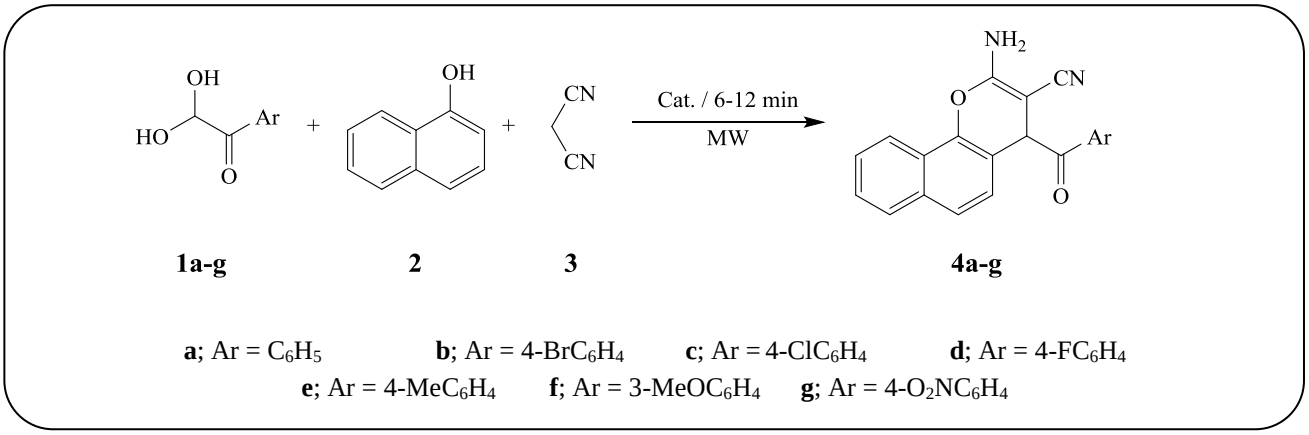
Light yellow-white solid, 81%, mp 235-237 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ (ppm): 8.36 (d, J = 8.1 Hz, 1H, Ar), 8.09 (d, J = 8.1 Hz, 1H, Ar), 7.85 (s, exchanged by D_2O addition, 2H, NH_2), 7.77 (d, J = 8.7 Hz, 2H, Ar), 7.87-7.59 (m, 6H, Ar), 5.76 (s, 1H, CH). ^{13}C-NMR (75.5 MHz, DMSO-d_6) δ (ppm): 165.2, 152.3, 134.7, 131.8, 130.1, 129.9, 129.8, 129.6, 129.0, 127.7, 126.6, 124.6, 123.7, 120,7, 120.1, 118.6, 117.2, 108.9, 35.7. FT-IR (KBr) ν_{max} (cm^{-1}): 3436, 3266, 3198, 3061, 2948, 2719, 2596, 2365, 2252, 2211, 1915, 1677, 1484, 1377, 1262, 1225, 1138, 1090, 1012, 822, 736, 681, 640, 567, 498, 425; Anal. Calcd for $C_{21}H_{13}ClN_2O_2$: C, 69.91; H, 3.63; N, 7.76; Found: C, 69.70; H, 3.47; N, 7.60%.

2-Amino-4-(4-fluorobenzoyl)-4H-benzo[h]chromene-3-carbonitrile (4d)

Light yellow solid, 89%, mp 210-213 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ (ppm): 8.36 (d, J = 8.4 Hz, 1H, Ar), 8.09 (d, J = 8.4 Hz, 1H, Ar), 7.98-7.90 (m, 2H, Ar), 7.88 (s, exchanged by D_2O addition, 2H, NH_2), 7.85-7.66 (m, 3H, Ar), 7.62 (d, J = 6.9 Hz, 1H, Ar), 7.48 (t, J = 8.7 Hz, 2H, Ar), 5.74 (s, 1H, CH). ^{13}C-NMR (75.5 MHz, DMSO-d_6) δ (ppm): 165.3, 152.3, 149.2, 131.7,

Table 1: The melting points and yields of products 4a-g.

Entry	Substrate	Products	Time (Min)	M.p (ºC)	Yield (%)
1	 **1a**	 **4a**	6	215-217	73
2	 **1b**	 **4b**	12	238-240	78
3	 **1c**	 **4c**	12	235-237	81
4	 **1d**	 **4d**	11	210-213	89
5	 **1e**	 **4e**	8	229-231	70
6	 **1f**	 **4f**	7	218-220	72
7	 **1g**	 **4g**	10	230-232	79



Scheme 1: Synthesis of 2-amino-4-aroyl-4H-benzo[h]chromene-3-carbonitriles 4a-g.

130.5, 129.0, 127.6, 126.4, 126.0, 124.5, 123.7, 120.7, 120.0, 118.7, 117.3, 116.7, 116.5, 108.4, 35.6. FT-IR (KBr) ν_{max} (cm^{-1}): 3452, 3327, 3065, 2914, 2467, 2201, 1685, 1595, 1510, 1379, 1234, 1159, 1086, 1036, 802; Anal. Calcd for $C_{21}H_{13}FN_2O_2$: C, 73.25; H, 3.81; N, 8.14; Found: C, 73.08; H, 3.95; N, 8.07%.

2-Amino-4-(4-methylbenzoyl)-4H-benzo[h]chromene-3-carbonitrile (4e)

Light yellow-white solid, 70%, mp 229-231 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ (ppm): 8.44 (d, *J* = 7.5 Hz, 1H, Ar), 8.08 (d, *J* = 7.5 Hz, 1H, Ar), 7.86 (s, exchanged by D_2O addition, 2H, NH_2), 7.79-7.60 (m, 6H, Ar), 7.42 (d, *J* = 6.9 Hz, 2H, Ar), 5.71 (s, 1H, CH), 2.42 (s, 3H, Me). ^{13}C-NMR (75.5 MHz, DMSO-d_6) δ (ppm): 165.4, 153.3, 149.1, 139.8, 131.6, 130.1, 129.0, 127.9, 127,6, 126.5, 126.3, 124.4, 123.9, 120.7, 119.9, 118.6, 117.4, 107.8, 35.7, 21.4. FT-IR ν_{max} (cm^{-1}): 3383, 3146, 2927, 2860, 2251, 1698, 1626, 1508, 1371, 1267, 1186, 1093, 1023, 959, 875, 803, 740, 659, 600; Anal. Calcd for $C_{22}H_{16}N_2O_2$: C, 77.63; H, 4.74; N, 8.23; Found: C, 77.52; H, 4.88; N, 8.09%.

2-Amino-4-(3-methoxybenzoyl)-4H-benzo[h]chromene-3-carbonitrile (4f)

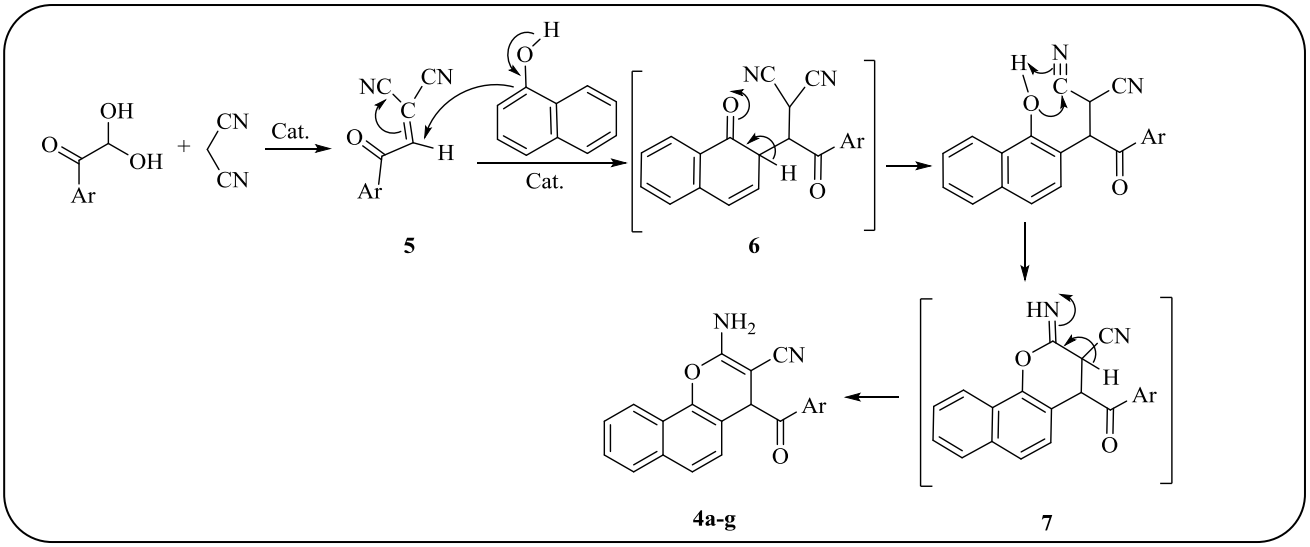
Yellow solid, 72%, mp 218-220 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ (ppm): 8.37 (d, *J* = 7.8 Hz, 1H, Ar), 8.09 (d, *J* = 7.8 Hz, 1H, Ar), 7.88 (s, exchanged by D_2O addition, 2H, NH_2), 7.79-7.68 (m, 3H, Ar), 7.42-7.63 (m, 4H, Ar), 7.12 (d, *J* = 8.1 Hz, 1H, Ar), 5.76 (s, 1H, CH), 3.89 (s, 3H, OMe). ^{13}C-NMR (75.5 MHz, DMSO-d_6) δ (ppm): 165.4, 160.0, 131.7, 130.7, 130.6, 127.6, 124.5, 123.9, 120.8, 120.3, 120.1, 117.4, 115.9, 113.3, 108.7, 55.7, 35.7. FT-IR ν_{max} (cm^{-1}): 3378, 3229, 3088, 2931, 2200, 1678, 1519, 1370, 1244, 1140, 1099, 1041, 801; Anal. Calcd for $C_{22}H_{16}N_2O_3$: C, 74.15; H, 4.53; N, 7.86; Found: C, 74.34; H, 4.45; N, 7.75%.

2-Amino-4-(4-nitrobenzoyl)-4H-benzo[h]chromene-3-carbonitrile (4g)

Yellow solid, 79%, mp 230-232 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ (ppm): 8.44 (d, *J* = 8.7 Hz, 2H, Ar), 8.41 (d, *J* = 6.6 Hz, 1H, Ar), 8.20 (d, *J* = 8.7 Hz, 2H, Ar), 8.11 (d, *J* = 9 Hz, 1H, Ar), 7.91 (s, exchanged by D_2O addition, 2H, NH_2), 7.90-7.85 (m, 2H, Ar), 7.72 (d, *J* = 6.6 Hz, 1H, Ar), 7.68 (t, *J* = 8.1 Hz, 1H, Ar), 5.90 (s, 1H, CH). ^{13}C-NMR (75.5 MHz, DMSO-d_6) δ (ppm): 165.0, 150.6, 147.7, 135.3, 132.2, 129.8, 129.1, 128.9, 127.9, 127.1, 125.0, 124.7, 123.7, 120.7, 120.4, 118.6, 117.1, 111.3, 35.8. FT-IR (KBr) ν_{max} (cm^{-1}): 3362, 2938, 2256, 1693, 1602, 1516, 1341, 1195, 1096, 853, 810, 752, 670. Anal. Calcd for $C_{21}H_{13}N_3O_4$: C, 67.92; H, 3.53; N, 11.32; Found: C, 67.82; H, 3.69; N, 11.17%.

RESULTS AND DISCUSSION

After some preliminary experimentation, we found that the reaction of arylglyoxal monohydrates **1a-g** with 1-naphthol (**2**) and malononitrile (**3**) in the presence of Mg-Al hydrotalcite as a heterogeneous catalyst under solvent-free microwave conditions afforded the desired 2-amino-4-aroyl-4*H*-benzo[*h*]chromene-3-carbonitrile derivatives **4a-g** as shown in Scheme 1. It should be mentioned that the reaction failed to give the desired product in the absence of the catalyst.



Scheme 2: Proposed mechanism of reaction for the synthesis of products 4a-g.

The proposed mechanism for this reaction is shown in Scheme 2. Initially, the arylglyoxal monohydrates **1a-g** react with malononitrile (**3**) to provide the corresponding 2-(2-oxo-2-arylethylidene)malononitriles **5** by a *Knoevenagel* condensation in the presence of a catalyst. The Mg-Al hydrotalcite accelerates the dehydration process by activation of the C=O group. The ortho C-alkylation of α-naphthol takes place in the second step, which can also be promoted by the catalyst to afford the intermediate **6**. The latter cyclization through nucleophilic attack of OH substituent on CN scaffold provides intermediate **7**, which followed by subsequent tautomerization to give the desired products **4a-g**.

The structures of substituted 4*H*-benzo[*h*]chromene-3-carbonitriles **4a-g** were characterized using FT-IR, ^{1}H-NMR, ^{13}C-NMR spectral data, and microanalysis. The characteristic singlets at δ = 5.71-5.90 ppm and δ = 7.73-7.91 ppm were ascribed to the CH and NH_2 groups respectively, which were present in all products. In the ^{13}C-NMR spectra of products, signals located at δ = 165-165.4 ppm was attributed to the carbonyl group. In the FT-IR spectra, the characteristic absorption bands at 1677-1698 and 2200-2256 cm^{-1} could be assigned to the vibrations of carbonyl and nitrile groups respectively.

CONCLUSIONS

In conclusion, we have described a facile procedure for the synthesis of a new series of 2-amino-4-aroyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitriles by one-pot, the three-component reaction of 1-naphtol, arylglyoxals and malononitrile in the presence highly efficient catalyst under solvent-free microwave conditions. The shorter reaction times, mild reaction condition, easy workup, good to excellent yields, availability of starting materials and its applicability in the synthesis of other heterocyclic compounds are the merits of this method. These products may have great potential for pharmacological and biological applications.

Acknowledgments

The authors gratefully acknowledge the financial assistance from Urmia University.

Received : Mar. 7, 2018 ; Accepted : Jun. 18, 2018

REFERENCES

[1] Abdelhamid A.O., Elghandour A.H., Ahmed S.A., Zaki Y.H., Reaction of Hydrazonoyl Halides 49 1: Synthesis and Antimicrobial Activity of Some New Pyrimido[1,2-*b*][1,2,4,5]tetrazin-6-one, Tetrazino[3,2-*b*]quinazolin-5-one, Pyrimidino[1,2-*b*]1,2,4,5-tetrazin-5-one and Triazolo[4,3-*a*]pyrimidine Derivatives, *J. Sulfur Chem.*, **26**: 405-410 (2005).

[2] (a) Kidwai M., Saxena S., Rahman Khan M.K., Thukral S.S., Aqua Mediated Synthesis of Substituted 2-Amino-4*H*-chromenes and in Vitro Study as Antibacterial Agents, *Bioorg. Med. Chem. Lett.*, **15**(19): 4295-4298 (2005);

(b) Cieplik J., Raginia M., Pluta J., Gubrynowicz O., Bryndal I., Lis T., Synthesis and Antibacterial Properties of 1,2,3-Aryl-1,2,3,4-tetrahydropyrimido[4,5-*d*]pyrimidine Derivatives. *Acta Pol. Pharm. Drug Res.*, **65**: 427-434 (2008).

[3] Ranganatha V.L., Zameer F., Meghashri S., Rekha N.D., Girish V., Gurupadaswamy H. D., Khanum S., Design, Synthesis, and Anticancer Properties of Novel Benzophenone-Conjugated Coumarin Analogs, *Arch. Pharm. Chem.*, **346**: 901-911 (2013).

[4] Bonsignore L., Loy G., Secci D., Calignano A., Synthesis and Pharmacological Activity of 2-Oxo-(2*H*)1-benzopyran-3-carboxamide Derivatives, *Eur. J. Med. Chem.*, **28**: 517-520 (1993).

[5] a) Al-Thebeiti M.S., Synthesis of Some New Derivatives of Thiazolo-[3,2-*a*]pyrimidine-3,5,7(2*H*)-trione of Potential Biological Activity, *Boll. Chim. Farm.*, **140**: 221-223 (2001);
(b) Shigeta S., Mori S., Watanabe F., Takahashi K., Nagata T., Koike N., Wakayama T., Saneyoshi M., Synthesis and Antiherpesvirus Activities of 5-Alkyl-2-thiopyrimidine Nucleoside Analogues, *Antivir. Chem. Chemother*., **13**: 67-82 (2002).

[6] a) Jeso V., Nicolaou K.C., Total Synthesis of Tovophyllin B, *Tetrahedron Lett.*, **50**: 1161-1163 (2009);
(b) Thareja S., Verma A., Kalra A., Gosain S., Rewatkar P.V., Kokil G.R., Novel Chromeneimidazole Derivatives as Antifungal Compounds: Synthesis and In Vitro Evaluation, *Acta Pol. Pharm.*, **67**: 423-427 (2010).

[7] Karia D.C., Pandya H.K., Godvani N.K., Synthesis, Characterization and Anti-HIV Activity of 4-Hydroxy-3-(5-methylisoxazol-3-yl)pyrano(3,2-c)chromene-2,5-dione, *Asian J. Biochem. Pharm. Res.*, **2**(2): 126-130 (2012).

[8] Chabchoub F., Messaâd M., Mansour H.B., Chekir-Ghedira L., Salem M., Synthesis and Antigenotoxic Activity of some Naphtho[2,1-*b*]pyrano[3,2-*e*][1,2,4]triazolo[1,5-c]pyrimidine Derivatives, *Eur. J. Med. Chem.*, **42**: 715-718 (2007).

[9] Kwak J.-H., Kang H.E., Jung J.K., Kim H., Cho J., Lee H., Synthesis of 7-Hydroxy-4-oxo-4*H*-chromene-and 7-hydroxychroman-2-carboxylic Acid *N*-alkyl Amides and Their Antioxidant Activities, *Arch. Pharm. Res.*, **29**: 728-734 (2006).

[10] a) Akbarzadeh T., Rafinejad A., Mollaghasem J.M., Safavi M., Fallah-Tafti A., Pordeli K., Ardestani M.S., Shafiee A., Foroumadi A., 2-Amino-3-cyano-4-(5-arylisoxazol-3-yl)-4*H*-chromenes: Synthesis and in Vitro Cytotoxic Activity, *Arch. Pharm. Chem.*, **345**: 386-392 (2012);
(b) El-Agrody A.M., Fouda A.M., Al-Dies A.M., Studies on the Synthesis, in Vitro Antitumor Activity of 4*H*-benzo[*h*]chromene, 7*H*-benzo[*h*]chromene[2,3-*d*]pyrimidine Derivatives and Structure-activity Relationships of the 2-, 3- and 2,3-positions, *Med. Chem. Res.*, **23**: 3187-3199 (2014);
(c) El-Agrody A.M., Khattab E.S.A.E.H., Fouda, A.M., Synthesis, Structure-activity Relationship (SAR) Studies on some 4-Aryl-4*H*-chromenes and Relationship between Lipophilicity and Antitumor Activity, *Lett. Drug Des. Discov.*, **11**: 1167-1176 (2014).

[11] Martínez-Grau A., Marco J., Friedlander Reaction on 2-Amino-3-cyano-4*H*-pyrans: Synthesis of Derivatives of 4*H*-pyran[2,3-*b*]quinoline, New Tacrine Analogues, *Bioorg. Med. Chem. Lett*., **7**(24): 3165-3170 (1997).

[12] Zhang J., Hu X., Zhou Z., Efficient and Eco-Friendly Procedure for the Synthesis of 2-Amino-4*H*-Chromenes Catalyzed by Diammonium Hydrogen Phosphate, *Iran. J. Chem. Chem. Eng. (IJCCE).*, **34**(4): 47-51 (2015).

[13] Mobinikhaledia A., Foroughifar N., Mosleh T., Hamta A., Synthesis of Some Novel Chromenopyrimidine Derivatives and Evaluation of Their Biological Activities, *Iranian J. Pharm. Res.*, **13**(3): 873-879 (2014).

[14] a) Panda D., Singh J.P., Wilson L., Suppression of Microtubule Dynamics by LY290181 A. Potential Mechanism for its Antiproliferative Action, *J. Biol. Chem.*, **272**: 7681-7687 (1997);
(b) Wood D.L., Panda D., Wiernicki T.R., Wilson L., Jordan M.A., Singh J.P., Inhibition of Mitosis and Microtubule Function Through Direct Tubulin Binding by a Novel Antiproliferative Naphthopyran LY290181, *Mol. Pharmacol.*, **52**: 437-444 (1997).

[15] El-Agrody A.M., El-Mawgoud H.K.A., Fouda A.M., Khattab E.S.A.E.H., Synthesis, In-vitro Cytotoxicity of 4*H*-benzo[*h*]chromene Derivatives and Structure–Activity Relationships of 4-Aryl Group and 3-, 7-positions, *Chem. Pap.*, **70**: 1279-1292 (2016).

[16] Khalafy J., Etivand N., Dilmaghani S., Ezzati M.; Poursattar Marjani A., A Convenient and Mild Synthesis of New 2-Aryl-3-hydroxy-6,7-dihydro-1*H*-indol-4(5*H*)-ones via a One-pot, Three-component Reaction in Water, *Tetrahedron Lett.*, **55**: 3781-3783 (2014).

[17] Khalafy J., Ezzati M., Rimaz M., Poursattar Marjani A., Yaghoobnejad Asl H., An Efficient and Facile Regioselective Synthesis of New Substituted (*E*)-1-(3-aryl-7,8-dihydrocinnoline-5(6*H*)-ylidene)hydrazines and (1*E*,2*E*)-1,2-bis(3-aryl-7,8-dihydrocinnoline-5(6*H*)-ylidene)hydrazines, *J. Iran. Chem. Soc.*, **11**(4): 1067-1074 (2014).

[18] Poursattar Marjani A., Khalafy J., Mahmoodi S., A Simple One-pot Synthesis of New 9-Aroyl-3,4,6,7,9,10-hexahydro-1,8(2*H*,5*H*)-acridinediones, *ARKIVOC.*, **iii**: 262-270 (2016).

[19] Khalafy J., Badparvar F., Poursattar Marjani A., Synthesis of 1-Aryl-6,6-dimethyl-2-phenyl-6,7-dihydro-1*H*-indol-4(5*H*)-ones by Two Steps, in a Three-component Reaction, *J. Chil. Chem. Soc.*, **61**(3): 3112-3115 (2016).

[20] Poursattar Marjani A., Khalafy J., Chitan M., Mahmoodi S., Microwave-assisted Synthesis of Acridine-1,8(2*H*,5*H*)-diones via a One-pot, Three Component Reaction, *Iran J. Chem. Chem. Eng. (IJCCE).*, **36**(2): 1-6 (2017).

[21] Poursattar Marjani A., Khalafy J., Haghi A., An Efficient Route for the Synthesis of 11-Aroyldiindeno[1,2-*b*:2',1'-*e*]pyridine-10,12-diones, *J. Heterocycl. Chem.*, **54**(6): 3294-3298 (2017).

[22] Ezzati M., Khalafy J., Poursattar Marjani A., Prager R.H., The Catalyst-free Syntheses of Pyrazolo[3,4-*b*]quinolin-5-one and Pyrazolo[4′,3′:5,6]pyrido[2,3-*d*]pyrimidin-5,7-dione Derivatives by One-pot, Three-component Reactions, *Tetrahedron.*, **73**(47): 6587-6596 (2017).

[23] Poursattar Marjani A., Taghartapeh M.R., Soltani A.R., Khalafy J., Kanani Y. Molecular Structures, Hirshfeld Surface Analysis, and Spectroscopic Properties of 6,8-Dimethyl-3-(4-chlorophenyl)-7-oxo-7,8-dihydropyrimido[4,5-*c*]pyridazin-5(6*H*)-one and 6,8-Dimethyl-3-(4-chlorophenyl)-7-thioxo-7,8-dihydropyrimido[4,5-*c*]pyridazin-5(6*H*)-one, *J. Struct. Chem.*, **58**(7): 1332-1340 (2017).

[24] Poursattar Marjani A., Ebrahimi Saatluo B., Nouri F., An Efficient Synthesis of 4*H*-chromene Derivatives by a One-pot, Three-component Reaction, *Iran. J. Chem. Chem. Eng. (IJCCE).*, **37**(1): 149-157 (2018).