Incredible Role of Glycerol in Multicomponent Synthesis of 2,3-Dihydroquinazoline-4(1*H*)-ones and 1-Amidoalkyl-2-naphthols

Maryam Hajjami,* Fariba Bakhti, and Elahe Ghiasbeygi

Department of Chemistry, Faculty of Science, Ilam University, P.O. Box 69315516, Ilam, Iran

Abstract. An efficient and green synthesis of 1-amidoalkyl-2-naphthols *via* one-pot three-component condensation of aromatic aldehydes, acetamide and 2-naphthol in the presence of catalytic amounts of glycerosulfonic acid in glycerol as a green solvent was elaborated. Also a simple method for the one-pot three-component synthesis of 2,3-dihydroquinazoline-4(1*H*)-ones using of isatoic anhydride, aldehydes and ammonium acetate in the presence of glycerol as a green catalyst and solvent was described. In this light we introduced the brilliant and valuable role of glycerol in the synthesis of 2,3-dihydroquinazoline-4(1*H*)-ones and 1-amidoalkyl-2-naphthols.
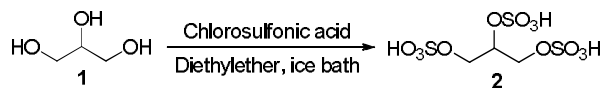
Keywords: 1-amidoalkyl-2-naphthol, glycerol, glycerosulfonic acid, multicomponent reaction, 2,3-dihydroquinazoline-4(1*H*)-ones, one-pot, catalyst

INTRODUCTION

Multicomponent reactions (MCRs), have elicited increasing interest since they have been performed without the need to isolate any intermediate during their processes, this reduces time and saves both energy and raw materials.[1] This reactions play as an important tool for building diverse and complex organic molecules in a tandem manner. They are also suitable for library synthesis to meet the demands for high-throughput screening in the pharmaceutention from both academic and industrial researchers.[2] Two of these MCRs are the synthesis of 1-amidoalkyl-2-naphthols and 2,3-dihydroquinazoline-4(1*H*)-ones. Amidoalkyl naphthols are an important group of compounds because they have been found to possess useful biological traits.[3] 1-amidoalkyl-2-naphthol derivatives are of significant medical relevance since they can be converted into hypertensive and bradycardia active 1-aminoalkyl-2-naphthols by amid hydrolysis reaction.[4] 1-amidoalkyl-2-naphthols can also be converted to 1,3-oxazine derivatives with different biological activities.[5] Also 2,3-Dihydroquinazolinone derivatives are an important class of fused heterocycles that display a wide range of biological, pharmacological, and medicinal properties involving antitumor, antibiotic, antipyretic, analgesic, antihypertonic, diuretic, antihistamine, antidepressant, and vasodilating activities.[6] The preparation of 1-amidoalkyl-2-naphthols can be carried out by the condensation of aryl aldehydes,

2-naphthol and acetonitrile or amide in the presence of Lewis or Brønsted acid catalysts[7] such as K-10 clay,[8] $Ce(SO_4)_2$,[9] iodine,[10] $K_5CoW_{12}O_{40} \cdot 3H_2O$,[11] p-TSA,[12] sulfamic acid,[13] cation-exchanged resins,[14] silica-sulfuric acid,[15] silica supported dual acidic ionic liquid,[16] $MoO_3–ZrO_2$[17] and trityl chloride.[18] Also, 2,3-dihydroquinazoline 4(1*H*)-ones prepared *via* one-pot three-component reaction of aldehydes, isatoic anhydride and source of ammonia. Therefore various procedures have been developed for preparing this important class of compounds.[19] Other methods including synthesis of 2,3-dihydroquinazoline 4(1*H*)-ones *via* one-pot three-component reaction catalyzed by L-pyrrolidine-2-carboxylic acid-4-hydrogen sulfate (supported on silica gel),[20] Cu-CNTs,[21] copper-catalyzed,[22] $Ce(CH_3SO_3)_3 \cdot 2H_2O$,[23] montmorillonite K-10,[24] MCM-41-SO$_3$H,[25] gallium triflate,[26] were also reported. However, most of the synthetic protocols reported so far suffer from drawbacks such as low yields, long reaction times, the use of expensive or toxic metal salts as catalysts, the use of toxic solvents and tedious work-up procedures. Therefore, introducing clean, versatile and eco-friendly processes have been under permanent attention.

It is well known that most organic reactions occur in a liquid phase.[27] Glycerol is a simple polyol compound. It is a colorless, odorless, viscous liquid that is widely used in pharmaceutical formulations. Glycerol has three hydroxyl groups that are responsible for its solubility in water and its hygroscopic nature. The glycerol

---

* Author to whom correspondence should be addressed. (E-mail: mhajjami@yahoo.com)

backbone is central to all lipids known as triglycerides. From the standpoint of green chemistry, glycerol is a valuable solvent for organic synthesis. Like polar solvents such as water, DMSO and DMF, glycerol is able to facilitate the dissolution of inorganic salts, acids, bases, enzymes and many metal transition complexes. Furthermore, it also dissolves organic compounds that are poorly miscible in water, many hydrophobic solvents, such as ethers and hydrocarbons, are immiscible in glycerol.[8] In this context, glycerol has a clear advantage compared with most organic solvents. Indeed, glycerol is a nontoxic, biodegradable and nonflammable solvent for which no special handling precautions or storage is required. It is also a by-product of the production of biodiesel *via* trans-esterification and co-product of the vegetable oil industry.[28] In this light we introduced the important role of glycerol in the synthesis of 2,3-dihydroquinazoline-4(1*H*)-ones and 1-amidoalkyl-2-naphthols.

EXPERIMENTAL

All chemicals were purchased from Aldrich and Merck and used without further purification. The known products were characterized by the comparison of their spectral (^{1}H NMR, and ^{13}C NMR, Bruker NMR spectrometer FX 400Q; MS, mass spectrometer 70 eV) and physical data with those of authentic samples.

General Procedure for the Synthesis of 2,3-Dihydroquinazoline-4(1*H*)-ones

A mixture of aldehyde **3** (1 mmol), isatoic anhydride **4** (1 mmol) and ammonium acetate **6** (3 mmol) in glycerol **1** (3 ml) was stirred at 80 °C in a boil bath for the appropriate time. After the completion of the reaction, as indicated by TLC, the reaction mixture was cooled, water was added and the mixture was stirred. That glycerol was dissolved in H_2O and separated from the mixture. The remained was dissolved in boiling EtOH and stirred. Solvent was evaporated. Finally solid product was recrystallized from ethanol. Some of the synthesized compounds were characterized by ^{13}CNMR and ^{1}HNMR analyses, and values were compared with the literature data of known products.

Synthesis of Glycerosulfonic Acid

In order to prepare glycerosulfonic, to glycerol **1** (1 mmol) in diethyl ether, chlorosulfonic acid (3 mmol) was added slowly in ice bath. The mixture was stirred for 1 h. After evaporation of the solvent the product is designated as glycerosulfonic acid (**2**, Scheme 1). The viscous liquid was then supported on the silica gel (1.424 g).

Scheme 1. Synthesis of glycerosulfonic acid

General Procedure for the Synthesis of 1-Amidoalkyl-2-naphthols

In a typical reaction, to a solution of 2-naphthol **7** (1 mmol), aldehyde **3** (1 mmol) and acetamide **8** (1.5 mmol) in glycerol **1** (0.5 ml), glycerosulfonic acid **2** (0.664 g) was added. Then the reaction mixture was stirred at 80 °C in a boil bath for the appropriate time (see Table 5). Reaction progress was monitored by TLC. After the completion of the reaction, the mixture was cooled to room temperature, water was added and the mixture was stirred for 10 minutes. That glycerol was dissolved in H_2O and separated from the mixture. The residue was dissolved in boiling EtOH and stirred for 10 minutes. Catalyst was filtered and solvent was evaporated under reduced pressure. Finally solid product was recrystallized from ethanol (Scheme 1). Synthesized compounds were characterized by ^{13}CNMR and ^{1}HNMR analyses, and values were compared with the literature data of known products.

Characterization Data of Selected 2,3-Dihydroquinazoline-4(1*H*)-ones and 1-Amidoalkyl-2-naphthols

*2-(4-Chlorophenyl)-2,3-dihydoquinazolin-4(1*H*)-one (**6b**)*
Mp: 196−197 °C. ^{1}H NMR (250 MHz, DMSO-d_6): δ/ppm = 8.29 (s, 1H), 7.61−7.43 (m, 5H), 7.26−7.20 (t, 1H), 7.12 (s, 1H), 6.75−6.63 (m, 2H), 5.75 (s, 1H). ^{13}C NMR (62 MHz, DMSO-d_6): δ/ppm = 163.9, 148.1, 141.1, 133.8, 133.4, 129.2, 128.7, 127.8, 117.7, 115.4, 114.9, 66.2.

*2-(4-Ethoxyphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (**6d**)*
Mp: 174−176 °C; ^{1}H NMR (400 MHz, CDCl₃): δ/ppm = 7.98 (s, 1H), 7.6 (d, *J* = 6 Hz, 2H), 7.34 (s, 1H), 7.26 (d, *J* = 2 Hz, 1H), 6.87−7.03 (m, 3H), 6.65 (d, *J* = 5.6 Hz, 1H), 5.87 (s, 1H), 5.75 (s, 1H), 4.12 (t, *J* = 4 Hz, 2H), 1.45 (q, *J* = 6.4 Hz, 3H). ^{13}C NMR (100 MHz, CDCl₃): δ/ppm = 164.8, 160.3, 147.4, 133.9, 130.3, 128.8, 128.7, 119.6, 115.7, 114.9, 114.5, 68.7, 63.6, 31.8, 30.9, 14.8.

*2-(4-Methylphenyl)-2,3-dihydroquinazolin-4(1*H*)-one (**6g**)*
Mp: 225−226 °C; ^{1}H NMR (250 MHz, DMSO-*d*6): δ/ppm = 8.21 (s, 1H), 7.62−7.59 (d, 1H), 7.38−7.35 (d, 2H), 7.26−7.16 (m, 3H), 7.03 (s, 1H), 6.75−6.63 (m, 2H), 5.71 (s, 1H), 2.49−2.42 (s, 3H). ^{13}C NMR (62 MHz, DMSO-*d*6): δ/ppm =164.1, 148.4, 139.1, 138.2, 133.7, 129.3, 127.8, 127.2, 117.5, 115.4, 114.9, 66.8, 21.2.

N-*[(4-Chloro-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide (9b)*
Mp: 237–239 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ / ppm = 10.1 (s, 1H), 8.51 (d, *J* = 8, 1H), 7.81 (m, 3H), 7.71 (d, *J* = 8.8, 1H),7.4 (t, *J* = 7.2, 2H), 7.29 (m, 2H), 7.19 (d, *J* = 8.4, 2H), 7.14 (d, *J* = 8, 1H), 2.02 (s, 3H). ^{13}C NMR (100 MHz, DMSO): δ / ppm = 169.9, 153.7, 142.3, 132.7, 131.1, 129.9, 129.1, 128.9, 128.4, 126.9, 123.6, 122.9, 118.9, 118.8, 56.6, 47.9, 23.1, 19.1.

N-*[(4-Hydroxy-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide (9f)*
Mp: 234–236 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ / ppm = 9.99 (s, 1H), 9.24 (s, 1H), 8.42 (d, *J* = 8.4, 1H), 7.88 (s, 1H), 7.78 (m, 2H), 7.37 (s, 1H), 7.25 (t, *J* = 6.4, 2H), 7.06 (d, *J* = 8.4, 1H), 7.02 (d, *J* = 8, 2H), 6.67 (d, *J* = 8.4, 2H), 1.98 (s, 3H). ^{13}C NMR (100 MHz, DMSO): δ / ppm = 169.5, 156.2, 153.5, 133.0, 132.8, 129.5, 129.0, 127.8, 126.6, 123.8, 122.8, 119.6, 119.0, 115.3, 48.0, 23.2.

N-*[(3-nitro-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide (9h)*
Mp: 206–209 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ / ppm = 10.17 (s, 1H), 8.65 (d, *J* = 8, 1H), 8.06 (m, 1H), 8.03 (s, 1H), 7.86 (m, 3H), 7.57 (m, 2H), 7.43 (t, *J* = 7.2, 1H), 7.27 (m, 3H), 2.04 (s, 3H). ^{13}C NMR (100 MHz, DMSO): δ / ppm = 170.2, 153.8, 148.2, 145.9, 133.3, 132.6, 130.4, 130.1, 129.2, 128.9, 127.3, 123.3, 123.1, 121.7, 120.9, 118.9, 118.3, 48.1, 23.0.

RESULTS AND DISCUSSION

Here in we investigated glycerol as green media with dual role for the one-pot three-component reaction of isatoic anhydride, aromatic aldehydes, and ammonium acetate for the synthesis of 2,3-dihydroquinazoline derivatives. Also following our current study in the synthesis of 1-amidoalkyl-2-naphthols[29] we would like to report that silica supported glycerosulfonic acid as a solid acid catalyst which has been used in the synthesis of 1-amidoalkyl-2-naphthol derivatives in glycerol as solvent (Scheme 2).

Synthesis of 2,3-Dihydroquinazolines

To find the optimized experimental conditions, the reaction of 4-chlorobenzaldehyde with isatoic anhydride and ammonium acetate in the presence of glycerol as solvent and catalyst, was selected as a model reaction (Table 1). A simple comparison between the entries 4 and 5
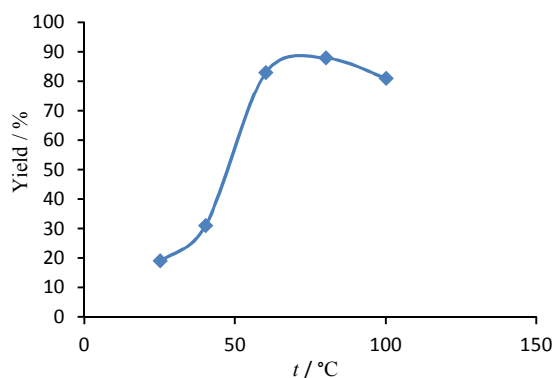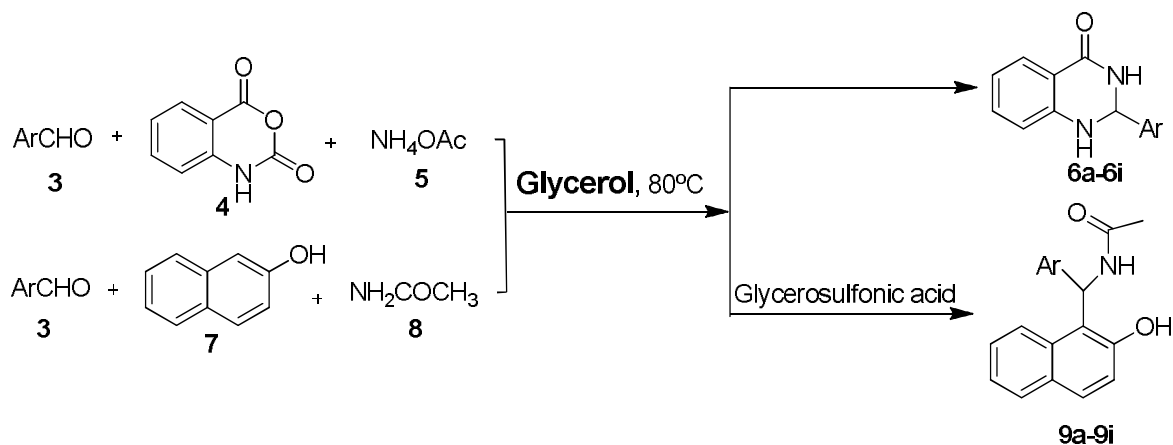

Figure 1. The effect of temperature on the reaction of 4-chlorobenzaldehyde, isatoic anhydride and ammonium acetate

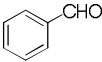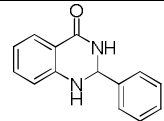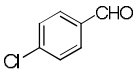**Table 1.** Optimization for synthesis of 2-(4-chlorophenyl)-2,3-dihydroquinazolin-4(1*H*)-one
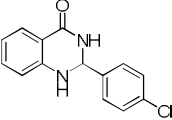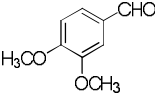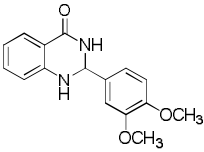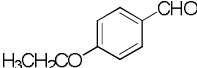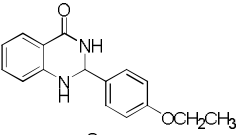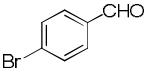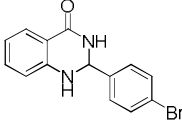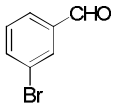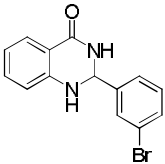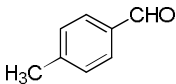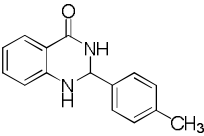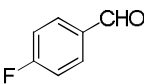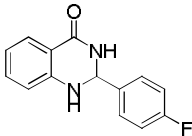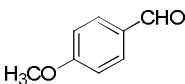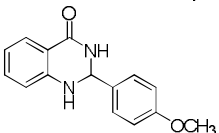
Entry	*n*(isatoic anhydride) / mmol	*n*(ammonium acetate) / mmol	*v*(glycerol) / mL	Yield / %[a]
1	1.2	1.5	3	–
2	1.2	3	3	37
3	1	3	1	58
4	1	3	2	64
5	1	3	3	97

[a] Crude yields.

Scheme 2. One-pot Synthesis of 2,3-dihydroquinazolines and 1-amidoalkyl-2-naphthols

Table 2. One-pot synthesis of 2,3-dihydoquinazolines in glycerol at 80 °C[a]

Entry	Substrate	Product	Time / h	Yield / %[b]	Mp / °C (Lit.)
1	(benzaldehyde, CHO)	(2-phenyl-2,3-dihydroquinazolin-4(1H)-one)	1	88	222–224 (219–222)[30]
2	(4-chlorobenzaldehyde, CHO, Cl)	(2-(4-chlorophenyl) product)	6	97	196–197 (198–200)[31]
3	(3,4-dimethoxybenzaldehyde, CHO, H₃CO, OCH₃)	(2-(3,4-dimethoxyphenyl) product, OCH₃, OCH₃)	3.35	86	204–207 (209–210)[30]
4	(4-ethoxybenzaldehyde, CHO, H₃CH₂CO)	(2-(4-ethoxyphenyl) product, OCH₂CH₃)	3	83	174–176 (167–168)[32]
5	(4-bromobenzaldehyde, CHO, Br)	(2-(4-bromophenyl) product, Br)	5	83	190–192 (195–197)[33]
6	(3-bromobenzaldehyde, CHO, Br)	(2-(3-bromophenyl) product, Br)	3	87	174–178 (184–185)[32]
7	(4-methylbenzaldehyde, CHO, H₃C)	(2-(4-methylphenyl) product, CH₃)	3	80	225–226 (233–234)[34]
8	(4-fluorobenzaldehyde, CHO, F)	(2-(4-fluorophenyl) product, F)	2	96	194–196 (192–193)[32]
9	(4-methoxybenzaldehyde, CHO, H₃CO)	(2-(4-methoxyphenyl) product, OCH₃)	2	95	181–184 (180–181)[34]

[a] Reaction conditions: Aldehydes (1 mmol), isatoic anhydride (1 mmol), ammonium acetate (3 mmol), glycerol (3 mL), $\vartheta = 80$ °C.
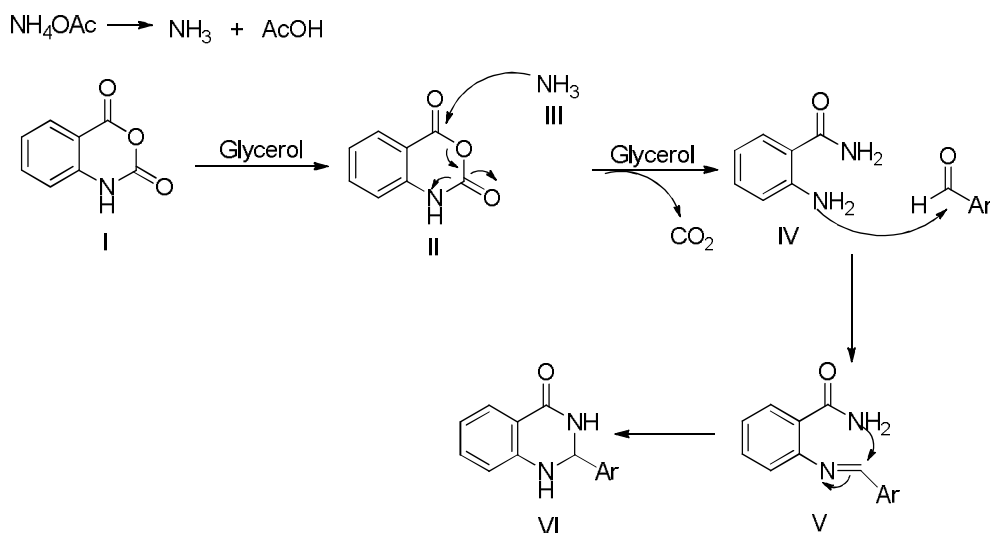[b] Crude yields.

reveals that apart from isatoic anhydride and ammonium acetate, the amount of glycerol effect on the yield of product. Therefore, we employed the optimized conditions (1 mmol isatoic anhydride, 3 mmol ammonium acetate and 3 mL glycerol) for the synthesis of 2,3-dihydroquinazolines (Entry 5, Table 1).

Furthermore, the model reaction was studied at different temperatures. As can be seen rising temperatures to 80 °C increased the yield of product. With further increase in temperature, the yield was reduced. Therefore when the reaction was performed at 80 °C, a higher yield was observed (Figure1).

To further establish the scope of optimized reaction conditions, variety of electronically divergent aromatic aldehydes were treated with isatoic anhydride and ammonium acetate in glycerol. The presence of electron-withdrawing and electron-releasing groups on the aromatic rings does not affect the yield of the product. The results of Table 2, clearly indicate the feasibility of three-component reaction. The products were synthesized in excellent yield with simple workup procedure. After completion the reaction, the glycerol was dissolved in water and separated from product. All results are compiled in Table 2.

An acceptable proposed mechanism for the synthesis of 2,3-dihydroquinazoline-4(1*H*)-ones is illustrated in Scheme 3. Ammonium acetate is dissociated to ammonia and acetic acid. In the presence of glycerol as catalyst, the carbonyl group of isatoic anhydride (I) is activated, to form an intermediate (II), then ammonia (III) attacks on the carbonyl unit of an intermediate (II) to produce an intermediate (IV), which in turn affords an intermediate (IV) through decarboxylation reaction and proton transfer. Subsequently, the reaction of aldehyde activated by glycerol with (IV) proceeds to resulting in the formation of the imine intermediate (V), which is followed by cyclization to yield the final product (VI). Generally, glycerol through hydrogen bonds with carbonyl group increased the electrophilicity of both isatoic anhydride and aldehyde carbonyl groups. So they react well with nucleophiles and the desired products achieved.

Table 3. The optimization of solvent for synthesis of *N*-[(4-chloro-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide in the duration of 7 h

Entry	Solvent	Yield / %[b]
1	Solvent-Free	-
2	EtOH	25
3	H_2O	29
4	H_2O + Glycerol	36
5	Glycerol	95

[a] Reaction conditions: aldehydes : 2-naphthol : acetamide : glycerosulfonic acid = 1 mmol : 1 mmol : 1.5 mmol : 0.664 g, $\vartheta = 80\,°C$.
[b] Crude yields.

To select an appropriate solvent, the reaction of 2-naphthol (1 mmol), 4-chlorobenzaldehyde (1 mmol) and acetamide (1.5 mmol) was examined as a model reaction (table 3). As mentioned in Table 3 and entry1 nothing

Scheme 3. Proposed mechanism for the synthesis of 2,3-dihydroquinazolines

Synthesis of 1-Amidoalkyl-2-naphthols

In the synthesis of 1-amidoalkyl-2-naphthols, in order to increase the acidic character of glycerol, it was reacted with chlorosulfonic acid and glycerosulfamic acid was synthesized. The structure of synthesized catalyst was examined by Fourier transform infrared spectroscopy (FTIR) analyses were carried on FTIR spectrophotometer (Bruker, Germany) Vertex 70 in the range of 400–4000 cm^{-1}. FTIR spectroscopy was used to detect the presence of binding groups in glycerol and glycerosulfonic acid (Figure 2). The vibration signals around 3000 cm^{-1} are typical O–H bands. And signals around 1000–1200 cm^{-1} are typical SO_2 bands.

Mass spectroscopy has been used to identify the mass of glycerosulfonic acid (Figure 3). The peak around 331.0 *m/z* is typical glycerosulfonic acid.

happens after 7 h under solvent-free condition. Also in ethanol or H_2O the small amount of product produced. Mixing water with glycerol led to more output, but it is not acceptable. It was observed that reaction proceeds more effectively in glycerol as the reaction medium (Table 3, Entry 5).

To further optimize the conditions, we examined the amount of glycerosulfonic acid as catalyst (Table 4). In the absence of any kinds of catalysts, a desirable product was obtained in very low yield (Table 4, Entry 1). Subsequently, the yield of *N*-[(4-chloro-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide gradually increased with increasing the amount of glycerosulfonic acid (Table 4, Entry 2–4). So the best yield was achieved in the presence of 0.664 g of catalyst.

Furthermore, In order to show the effect of temperature on 1-amidoalkyl-2-naphthols synthesis the
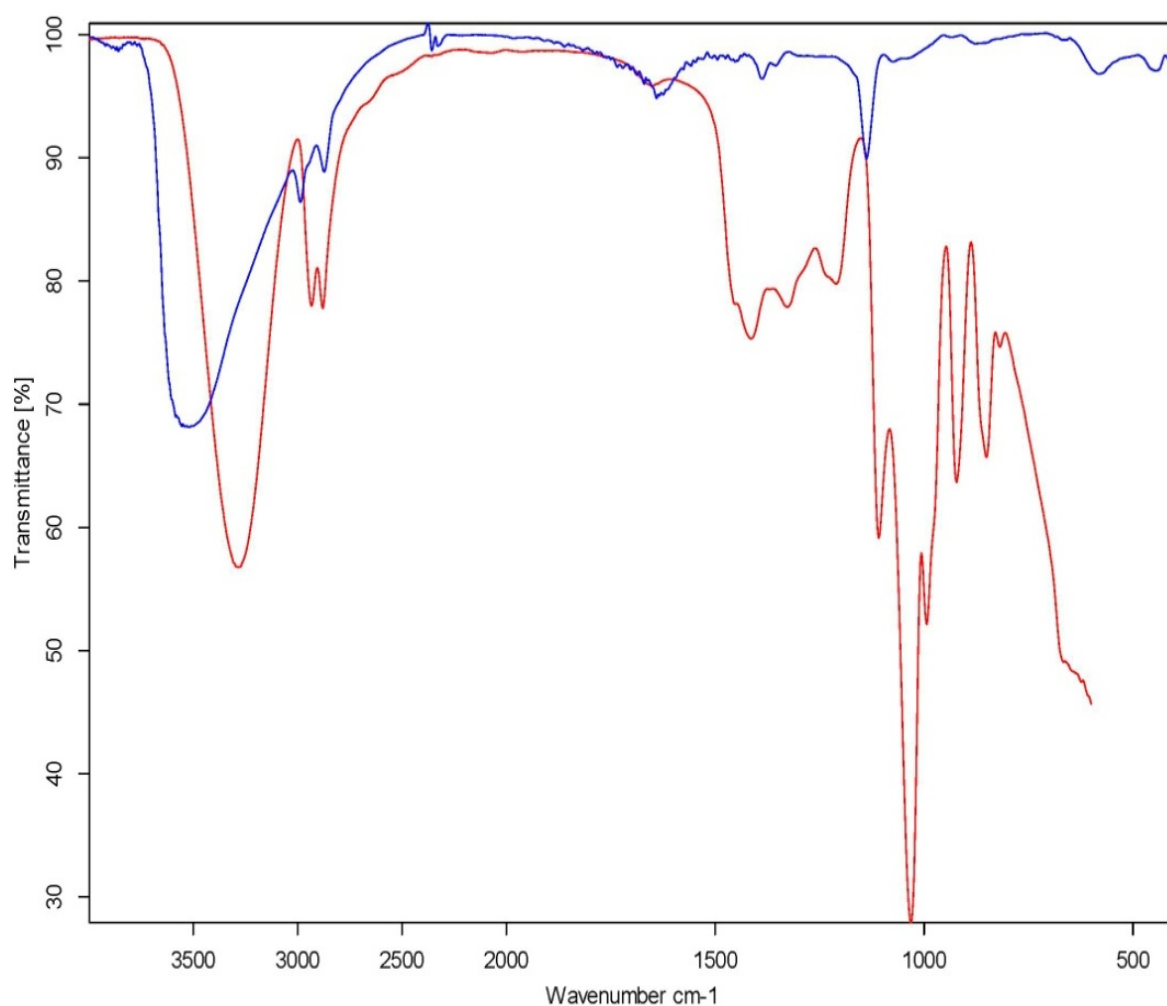
Figure 2. FT-IR spectra of glycerol and glycerosulfonic acid.

model reaction was studied at different temperatures. As shown in Figure 4, 80 °C is the best temperature for the reaction and at higher and lower temperatures did not achieve satisfactory results.

To explore the range of suitable substrates for this new catalytic system, the one-pot three component reac-

Table 4. The optimization of glycerosulfonic acid as catalyst for synthesis of *N*-[(4-chloro-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide at 80 °C[a]

Entry	m(Catalyst) / g	Time / h	Yield / %[b]
1	–	7	10
2	0.166	8	71
3	0.385	6	79
4	0.664	4.5	92
5	0.873	7.5	68
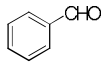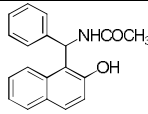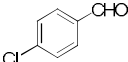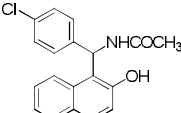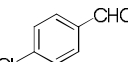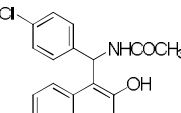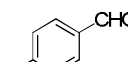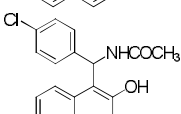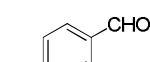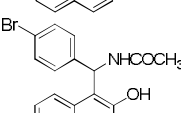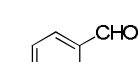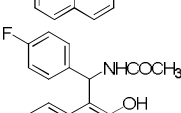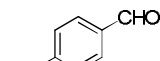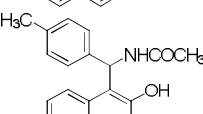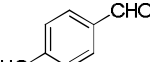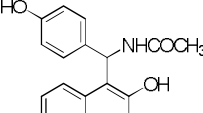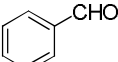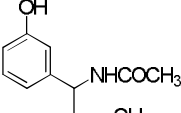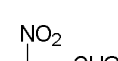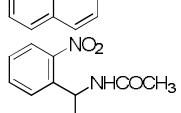
[a] Reaction conditions: aldehydes : 2-naphthol : acetamide = 1 mmol : 1 mmol : 1.5 mmol, in glycerol (0.5 mL), $\vartheta = 80$ °C.
[b] Crude yields.

tion of various aromatic aldehydes, 2-naphthol and acetamide catalyzed glycerosulfonic acid was investigated under the optimized conditions, and the results are given in Table 5. In all the reactions, excellent yields were obtained at appropriate reaction times. Clean and complete conversions leading to the corresponding 1-amidoalkyl-2-naphthols were observed. Aromatic aldehydes carrying either electron-withdrawing or electron-donating groups were all suitable for the reactions. The synthesis of *N*-[(4-chloro-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide was examined in the absence of glycerol or glycerosulfonic acid. As shown in entry 3, the reaction didn't occur in the absence of glycerol. Also without any glycerosulfonic acid just a trace amount of catalyst was achieved (Table 5, entry 4).

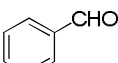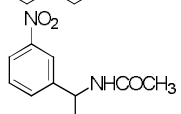On the basis of the previously reported mechanism for the synthesis of 1-amidoalkyl-2-naphthol, the acid catalysis condensation reaction *via* in situ generation of ortho-quinone methides (o-QMs) is presented. The o-QMs have been reacted with acetamide *via* conjugate addition to form 1-amidoalkyl-2-naphthols as final products.

Table 5. Glycerosulfonic acid catalyzed synthesis of 1-amidoalkyl-2-naphthols[a]

Entry	Substrate	Product	Time / h	Yield / %[b]	Mp / °C (Lit.)
1			5	98	238–240 (242–244)[35]
2			4.5	92	237–239 (229–230)[36]
3			7	0[c]	–
4			7	10[d]	237–239 (229–230)[36]
5			3.45	88	233–235 (228–230)[36]
6			5.5	89	244–246 (230–232)[37]
7			6.45	90	215–219 (221–223)[35]
8			6	92	234–236 (233–235)[29]
9			5.5	95	227–229 (227–228)[29]
10			5	93	206–209 (236–237)[35]
11			4	92	216–218 (218–219)[38]

[a] Reaction conditions: aldehydes (1 mmol), 2-naphthol (1 mmol), acetamide (1.5 mmol), glycerosulfonic acid (0.664 g), $\vartheta = 80$ °C.
[b] Crude yields.
[c] Reaction performed in the absence of glycerol.
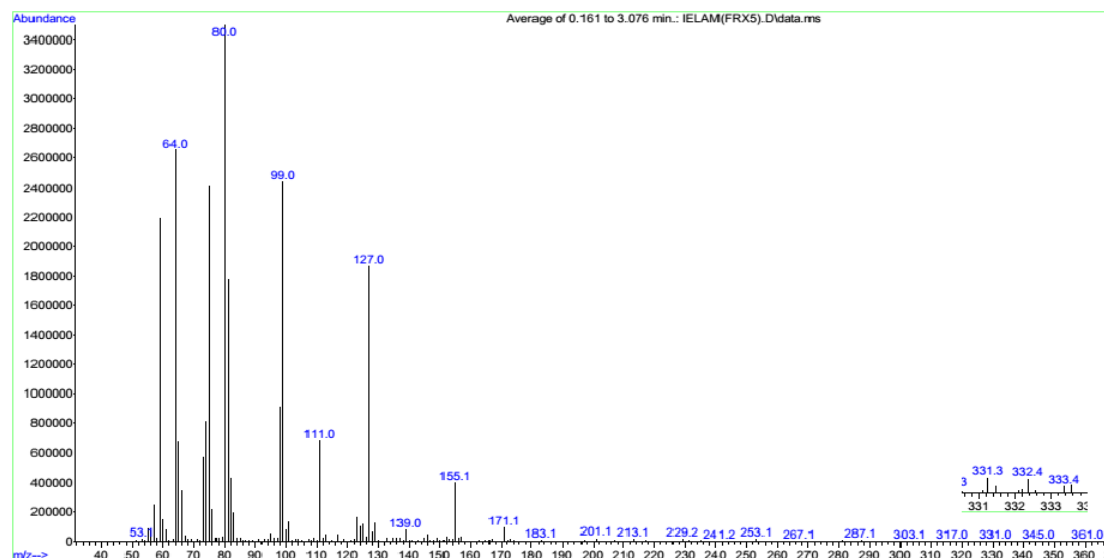[d] Reaction performed in the absence of glycerosulfonic acid.

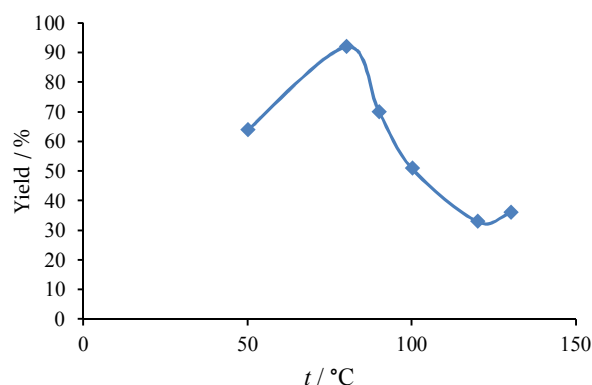**Figure 3.** Mass spectra of glycerosulfonic acid.

Figure 4. The effect of temperature on the synthesis of *N*-[(4-chloro-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide.

CONCLUSIONS

In conclusion, we have developed a green, versatile and easy manner for the synthesis of 2,3-dihydroquinazoline-4(1*H*)-one and 1-amidoalkyl-2-naphthol derivatives *via* one-pot three-component condensation reactions in glycerol as a green media at 80 °C. We have reported for the first time the use of glycerosulfonic acid as an efficient and heterogeneous catalyst for the one-pot synthesis of a variety of amidoalkylnaphthols in the presence of glycerol as a green solvent. The present protocol enjoys high yields of products, a wide range of substrates, simple workup, use of glycerol as a green solvent and use of glycerosulfonic acid as a new non-metallic catalyst.

REFERENCES

1. H. Khabazzadeh, K. Saidi, and N. J. Seyedi, *Chem. Sci.* **121** (2009) 429–433.
2. W. Q. Jiang, L. T. An, and J. P. Zou, *Chin. J. Chem.* **26** (2008) 1697–1701.
3. R. Ghorbani-Vaghei and S. M. Malaekehpour, *Cent. Eur. J. Chem.* **8** (2010) 1086-1089.
4. A. Shahrisa, S. Esmati, and M. J. Gholamhossini Nazari, *Chem. Sci.* **124** (2012) 927–931.
5. A. Zare, A. Hasaninejad, M. Merajodin, E. Kamali, M. Akbari, Z. Parsaee, A. Salimi Beni, and A. R. Moosavi-Zare, *Sci. Iran.* **18** (2011) 433–438.
6. M. Narasimhulu and Y. R. Lee, *Tetrahedron* **67** (2011) 9627–9634.
7. H. R. Shaterian, and H. Yarahmadi, *Tetrahedron Lett.* **49** (2008) 1297–1300.
8. H. R. Safaei, M. Shekouhy, S. Rahmanpure, and A. Shirinfeshan, *Green Chem.* **14** (2012) 1696–1704.
9. S. Kantevari, S. V. N. Vuppalapati, and L. Nagarapu, *Catal. Commun.* **8** (2008) 1857–1862.
10. N. P. Selvam, and P. T. Perumal, *Tetrahedron Lett.* **47** (2006) 7481–7483.
11. B. Das, K. Laxminarayana, B. Ravikanth, and B. R. J. Rao, *J. Mol. Catal. A: Chem.* **161** (2007) 180–183.
12. L. Nagarapu, M. Baseeruddin, S. Apuri, and S. Kantevari, *Catal. Commun.* **8** (2007) 1729–1734.
13. M. M. Khodaei, A. R. Khosropur, and H. Moghanian, *Synlett* (2006) 916–920.
14. S. B. Patil, M. P. Surpur, and S. D. Samant, *Ultrason. Sonochem.* **14** (2007) 515–518.
15. S. B. Patil, M. P. Surpur and S. D. Samant, *Synth. Commun.* **37** (2007) 1659–1664.
16. Q. Zhang, J. Luo, and Y. Wei, *Green Chem.* **12** (2010) 2246–2254.
17. S. Samantaray, G. Hota, and B. G. Mishra, *Catal. Commun.* **12** (2011) 1255–1259.
18. A. Khazaei, M. A. Zolfigol, A. R. Moosavi-zare, A. Zare, A. Parhami, and A. Khalafi-Nezhad, *Appl. Catal. A: Gen.* **386** (2010) 179–187.
19. M. Wang, T. T. Zhang, Y. Liang, and J. J. Gao, *Monatsh. Chem.* **143** (2012) 835–839.
20. A. Ghorbani-Choghamarani, and P. Zamani, *J. Iran. Chem. Soc.* **9** (2012) 607–613.

21. J. Safari, and S. Gandomi-Ravandi, *J. Mol. Catal. A: Chem.* **341** (2013) 135–140.
22. F. Wang, P. Zhao, and C. H. Xi, *Tetrahedron Lett.* **52** (2011) 231–235.
23. M. Wang, Z. G. Song, and T. T. Zhang, *Monatsh. Chem.* **141** (2010) 993–996.
24. P. Salehi, M. Dabiri, M. Baghbanzadeh, and M. Bahramnejad, *Synth. Commun.* **36** (2006) 2287–2292.
25. S. Rostamizadeh, A. Amani, G. H. Mahdavinia, H. Sepehrian, and S. Ebrahimi, *Synthesis* (2010) 1356–1360.
26. J. Chen, D. Wu, F. He, M. Liu, H. Wu, J. Ding, and W. Su, *Tetrahedron Lett.* **49** (2008) 3814–3818.
27. G. Srihari, M. Nagaraju, and M. M. Murthy, *Helv. Chim. Acta* **90** (2007) 1497–1504.
28. Y. Gu and F. Jerome, *Green Chem.* **12** (2010) 1127–1138.
29. M. Hajjami, F. Ghorbani, and F. Bakhti, *Appl. Catal. A: Gen.* **470** (2014) 303–310.
30. P. Salehi, M. Dabiri, M. A. Zolfigol, and M. Baghbanzadeh, *Synlett* (2005) 1155–1157.
31. A. Davoodnia, S. Allameh, A. R. Fakhari, and N. Tavakoli-Hoseini, *Chin. Chem. Lett.* **21** (2010) 550–553.
32. A. Ghorbani-Choghamarani, and M. Norouzi, *J. Mol. Catal. A: Chem.* **395** (2014) 172–179.
33. A. Saffar-Teluri and S. Bolouk, *Monatsh. Chem.* **141** (2010) 1113–1115.
34. M. Baghbanzadeh, P. Salehi, M. Dabiri, and G. Kozehgary, *Synthesis* (2006) 344–348.
35. J. Luo and Q. Zhang, *Monatsh. Chem.* **142** (2011) 923–930.
36. A. R. Hajipour, Y. Ghayeb, N. Sheikhan, and A. E. Ruoho, *Tetrahedron lett.* **50** (2009) 5649–5651.
37. H. R. Shaterian, H. Yarahmadi, and M. Ghashang, *Tetrahedron* **64** (2008) 1263–1269.
38. G. C. Nandi, S. Samai, R. Kumar, and M. Singh, *Tetrahedron Lett.* **50** (2009) 7220–7222.
39. K. C. Ashalu and J. N. Rao, *J. Chem. Pharm. Res.* **5** (2013) 44–47.

Supporting Information
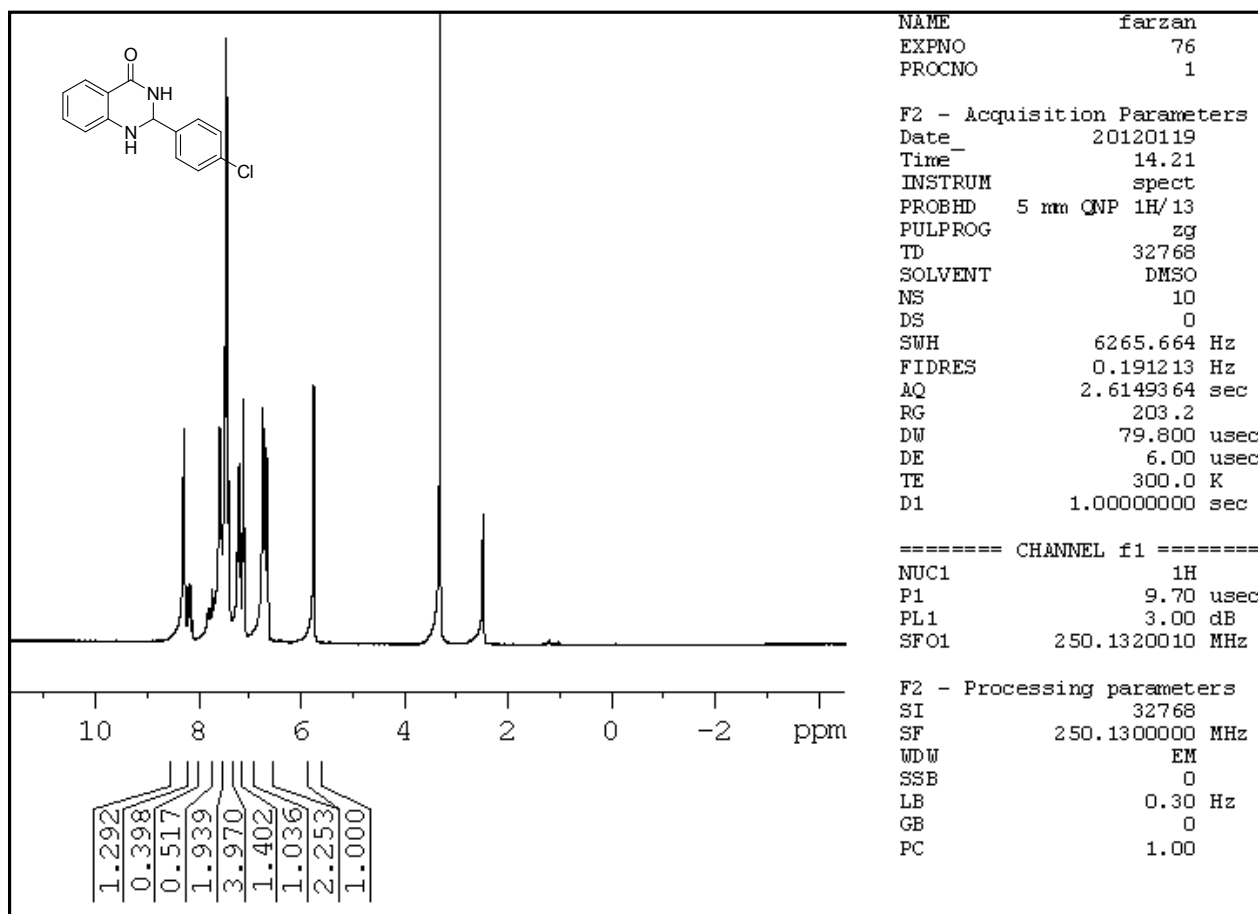
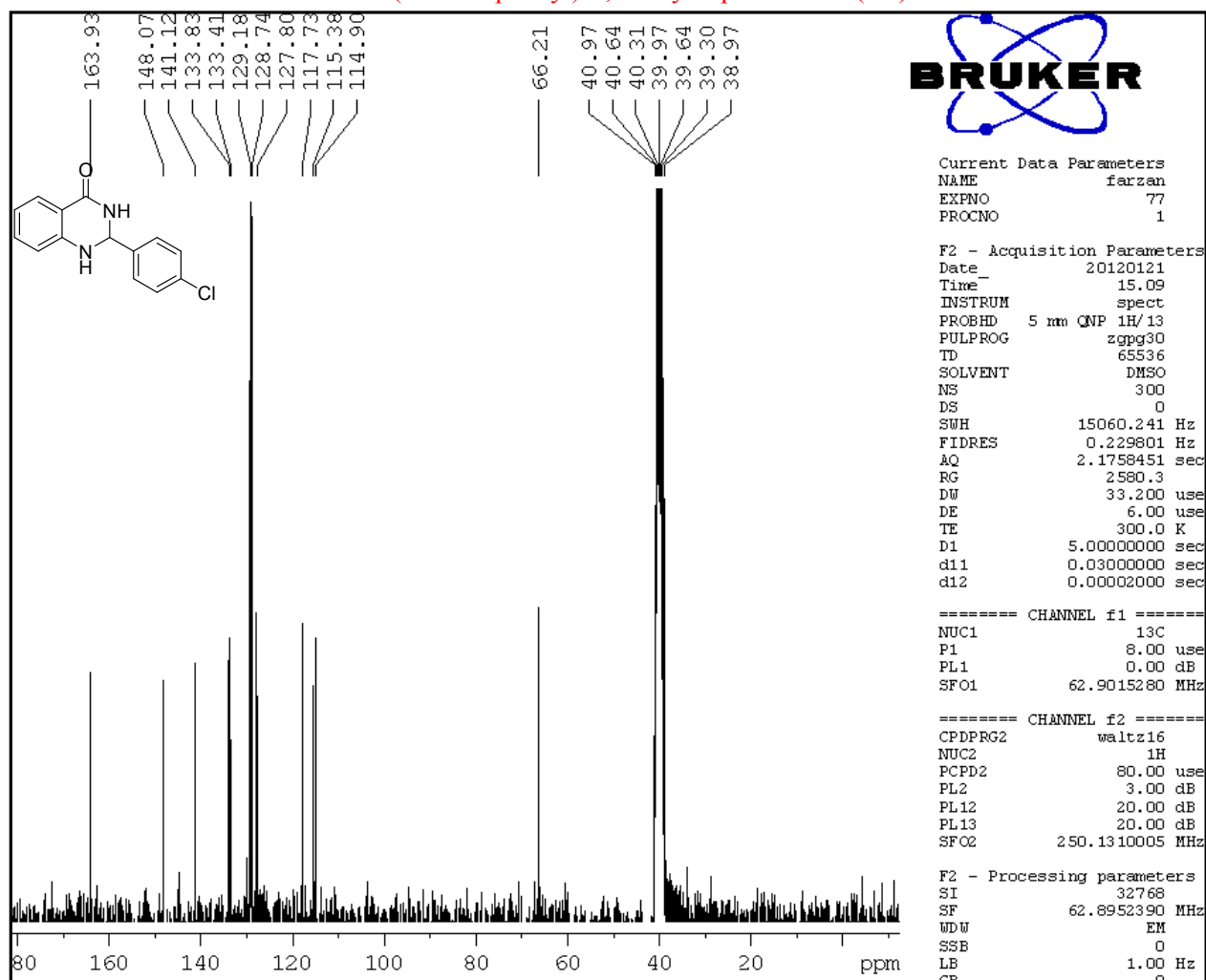# Incredible role of glycerol in multicomponent synthesis of 2,3-dihydroquinazoline-4(1H)-ones and 1-amidoalkyl-2-naphtols

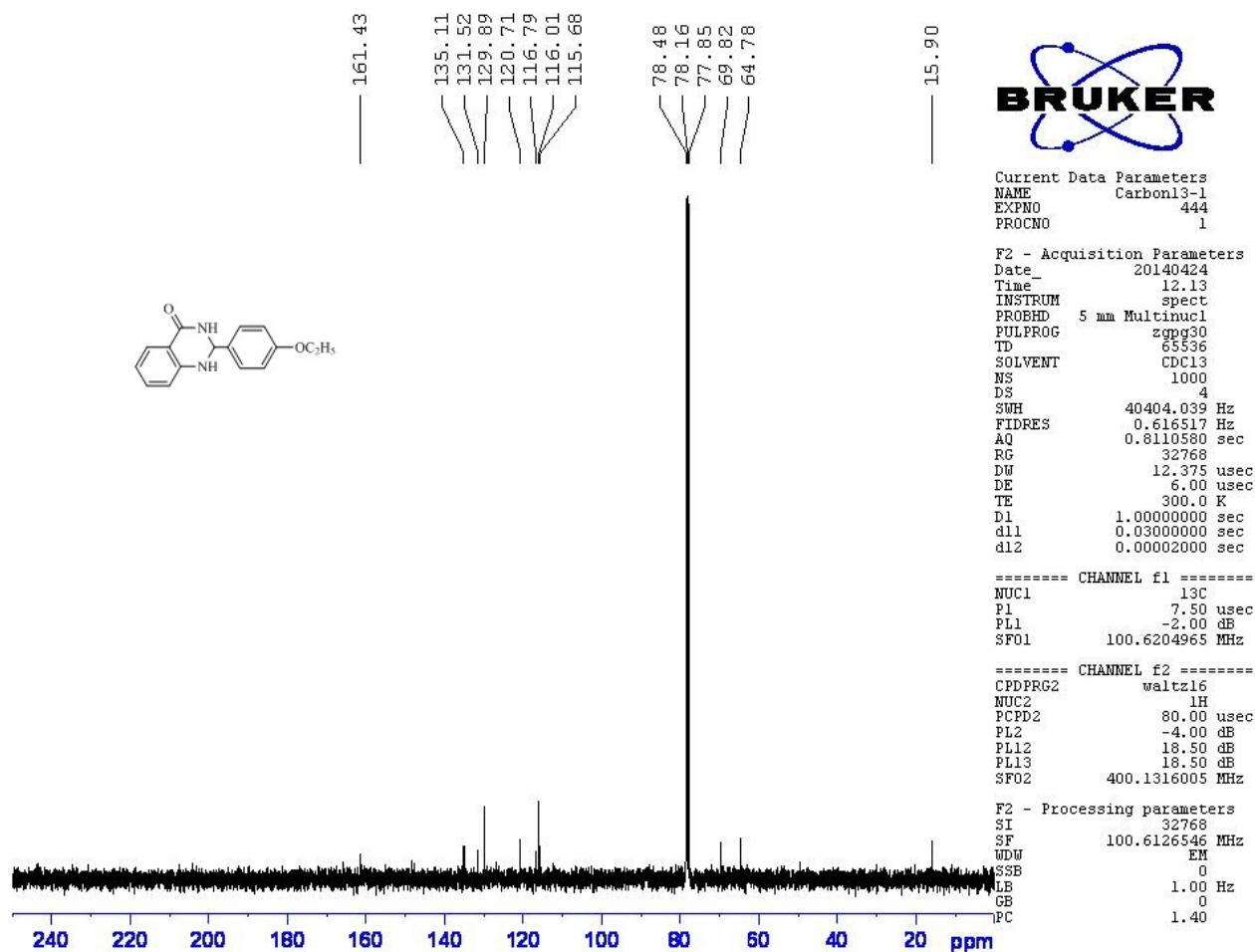Maryam Hajjami, ∗ Fariba Bakhti and Elahe Ghiasbeygi

Department of Chemistry, Faculty of Science, Ilam University, Ilam, Iran E-mail address: mhajjami@yahoo.com & m.hajjami@ilam.ac.ir
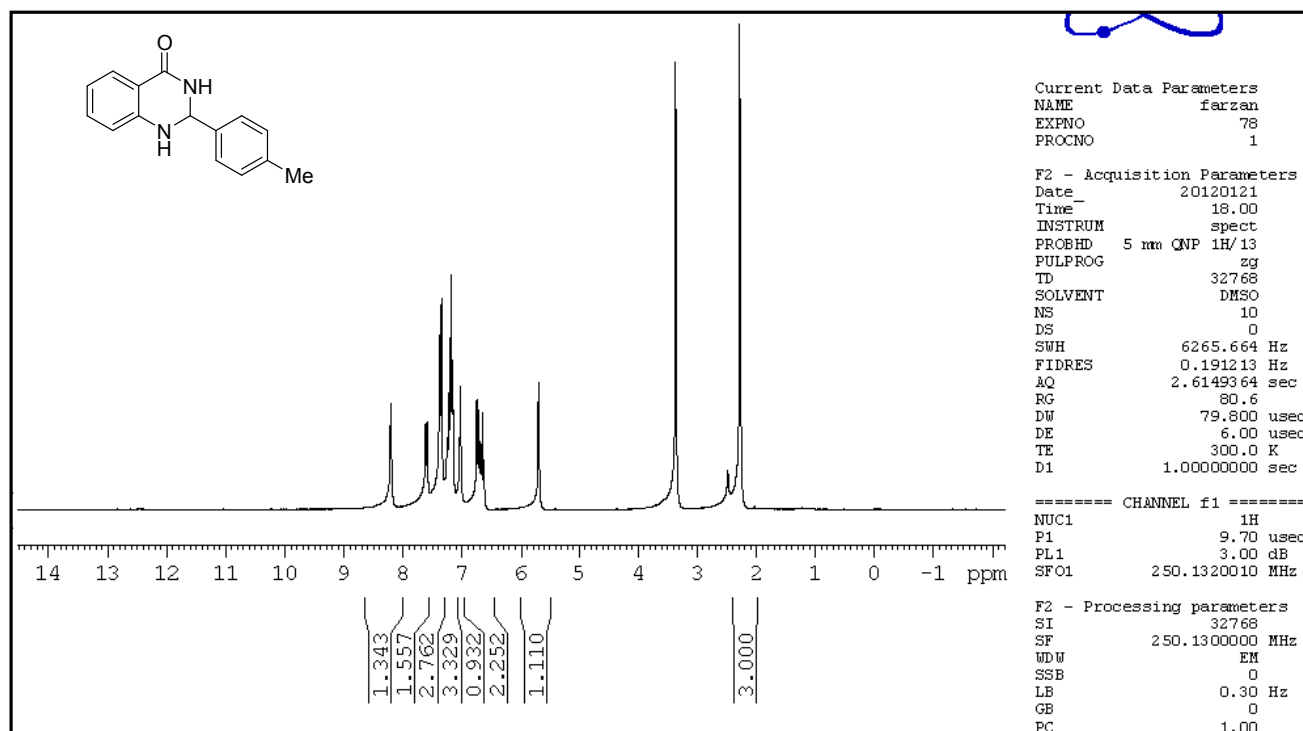
∗ Correspondence: Tel/Fax: +98 8412227022. E-mail addresses: mhajjami@yahoo.com

^{1}H NMR of 2-(4-chlorophenyl)-2,3-dihydoquinazolin-4(1H)-one


```
NAME                    farzan
EXPNO                       76
PROCNO                       1

F2 - Acquisition Parameters
Date_                 20120119
Time_                    14.21
INSTRUM                  spect
PROBHD     5 mm QNP  1H/13
PULPROG                     zg
TD                       32768
SOLVENT                   DMSO
NS                          10
DS                           0
SWH               6265.664 Hz
FIDRES           0.191213 Hz
AQ               2.6149364 sec
RG                       203.2
DW                 79.800 usec
DE                   6.00 usec
TE                   300.0 K
D1              1.00000000 sec

======== CHANNEL f1 ========
NUC1                        1H
P1                   9.70 usec
PL1                  3.00 dB
SFO1          250.1320010 MHz

F2 - Processing parameters
SI                       32768
SF            250.1300000 MHz
WDW                         EM
SSB                          0
LB                   0.30 Hz
GB                           0
PC                        1.00
```

Integration values: 1.292, 0.398, 0.517, 1.939, 3.970, 1.402, 1.036, 2.253, 1.000
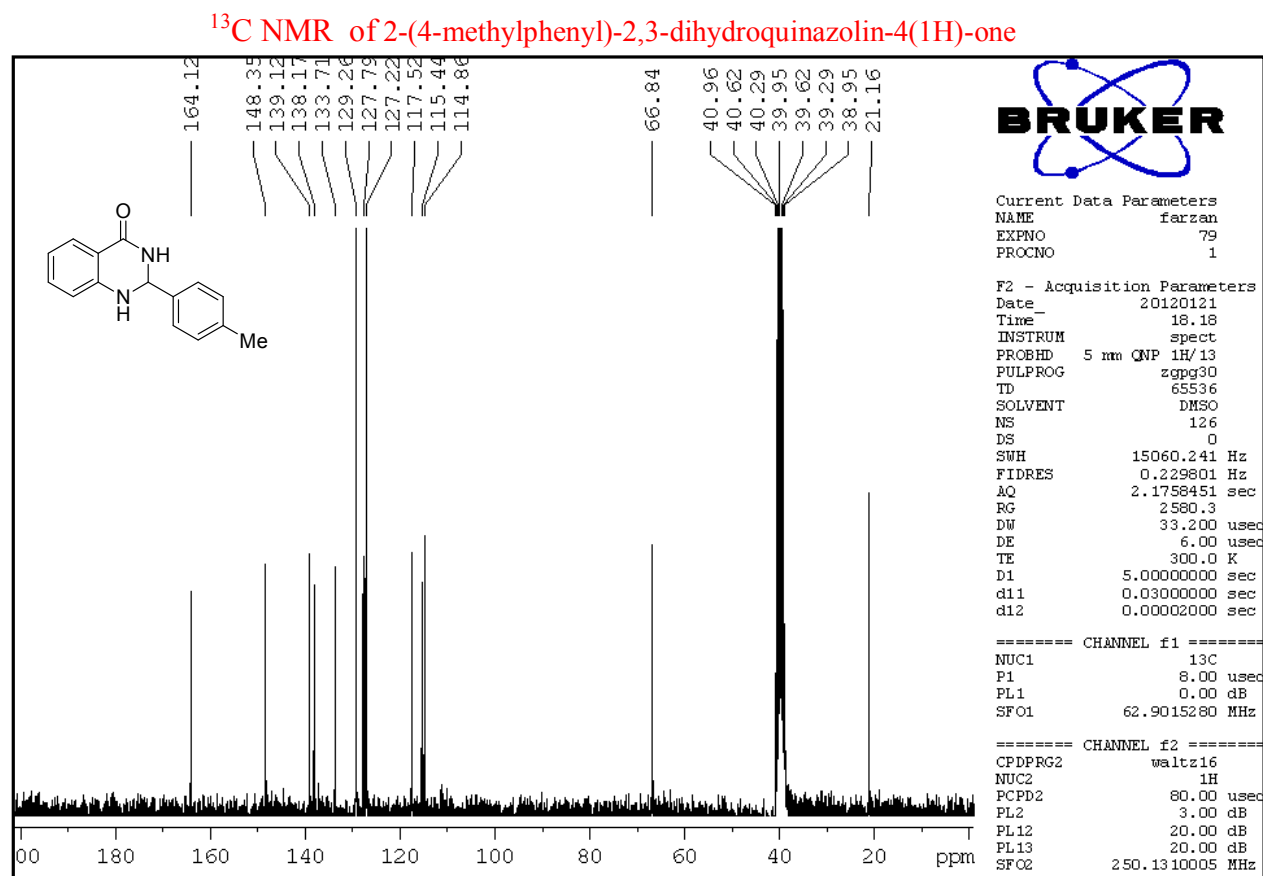
^{13}C NMR of 2-(4-chlorophenyl)-2,3-dihydoquinazolin-4(1H)-one

¹H NMR of 2-(4-ethoxyphenyl)-2,3-dihydroquinazolin-4(1H)-one
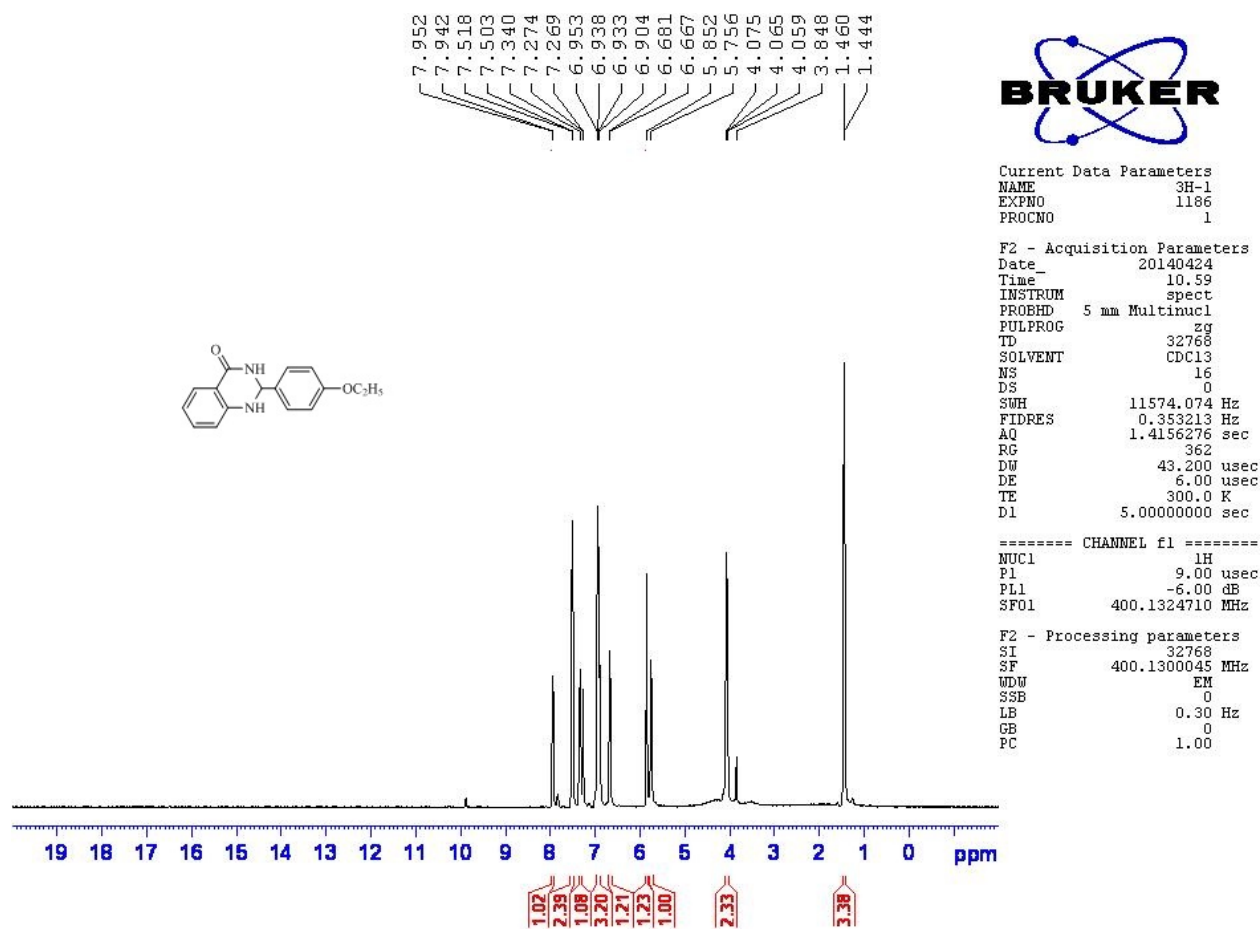
^{13}C NMR of 2-(4-ethoxyphenyl)-2,3-dihydroquinazolin-4(1H)-one

^{1}H NMR of 2-(4-methylphenyl)-2,3-dihydroquinazolin-4(1H)-one


```
Current Data Parameters
NAME                farzan
EXPNO                   78
PROCNO                   1

F2 - Acquisition Parameters
Date_             20120121
Time                 18.00
INSTRUM              spect
PROBHD    5 mm QNP 1H/13
PULPROG                 zg
TD                   32768
SOLVENT               DMSO
NS                      10
DS                       0
SWH             6265.664 Hz
FIDRES         0.191213 Hz
AQ            2.6149364 sec
RG                    80.6
DW             79.800 usec
DE               6.00 usec
TE                300.0 K
D1          1.00000000 sec

======== CHANNEL f1 ========
NUC1                    1H
P1               9.70 usec
PL1              3.00 dB
SFO1       250.1320010 MHz

F2 - Processing parameters
SI                   32768
SF         250.1300000 MHz
WDW                     EM
SSB                      0
LB               0.30 Hz
GB                       0
PC                    1.00
```

^{13}C NMR of 2-(4-methylphenyl)-2,3-dihydroquinazolin-4(1H)-one

^{1}H NMR of *N*-[(4-chloro-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide
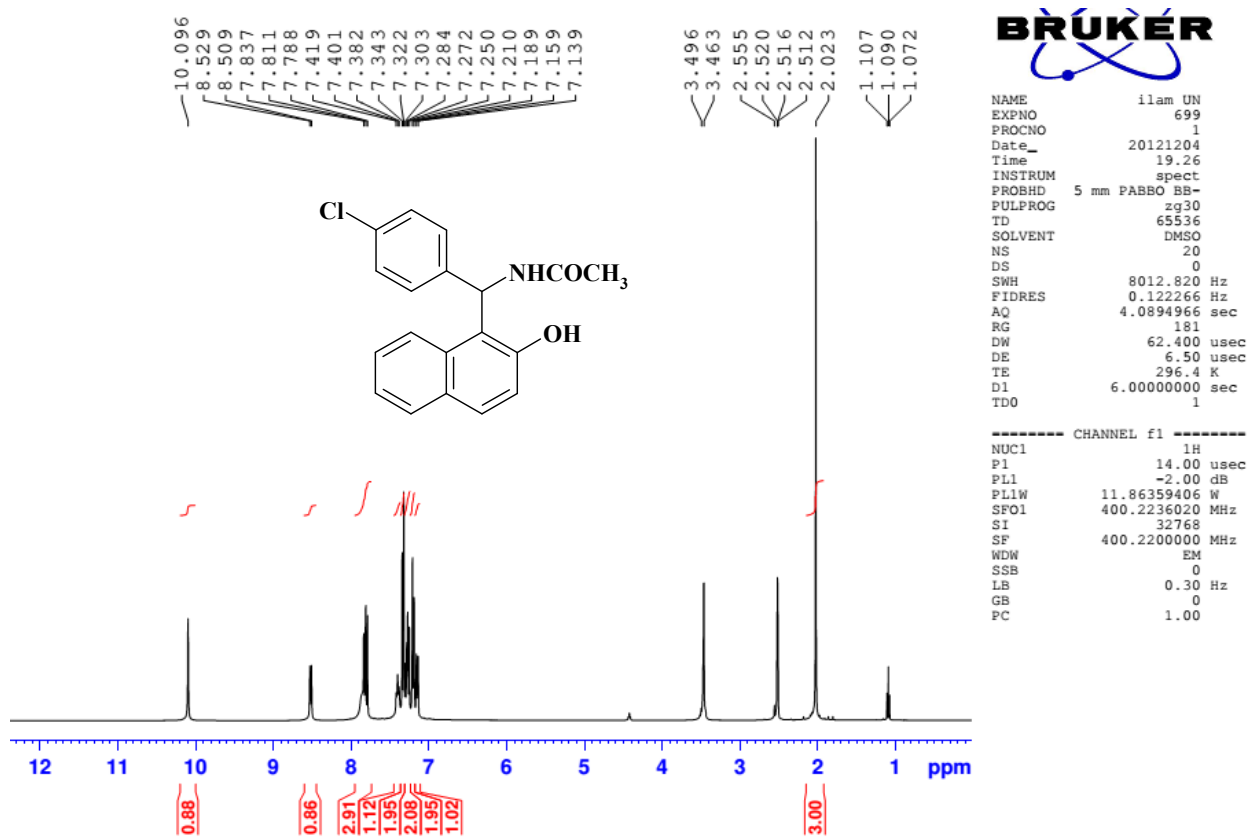
^{13}C NMR of *N*-[(4-chloro-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide
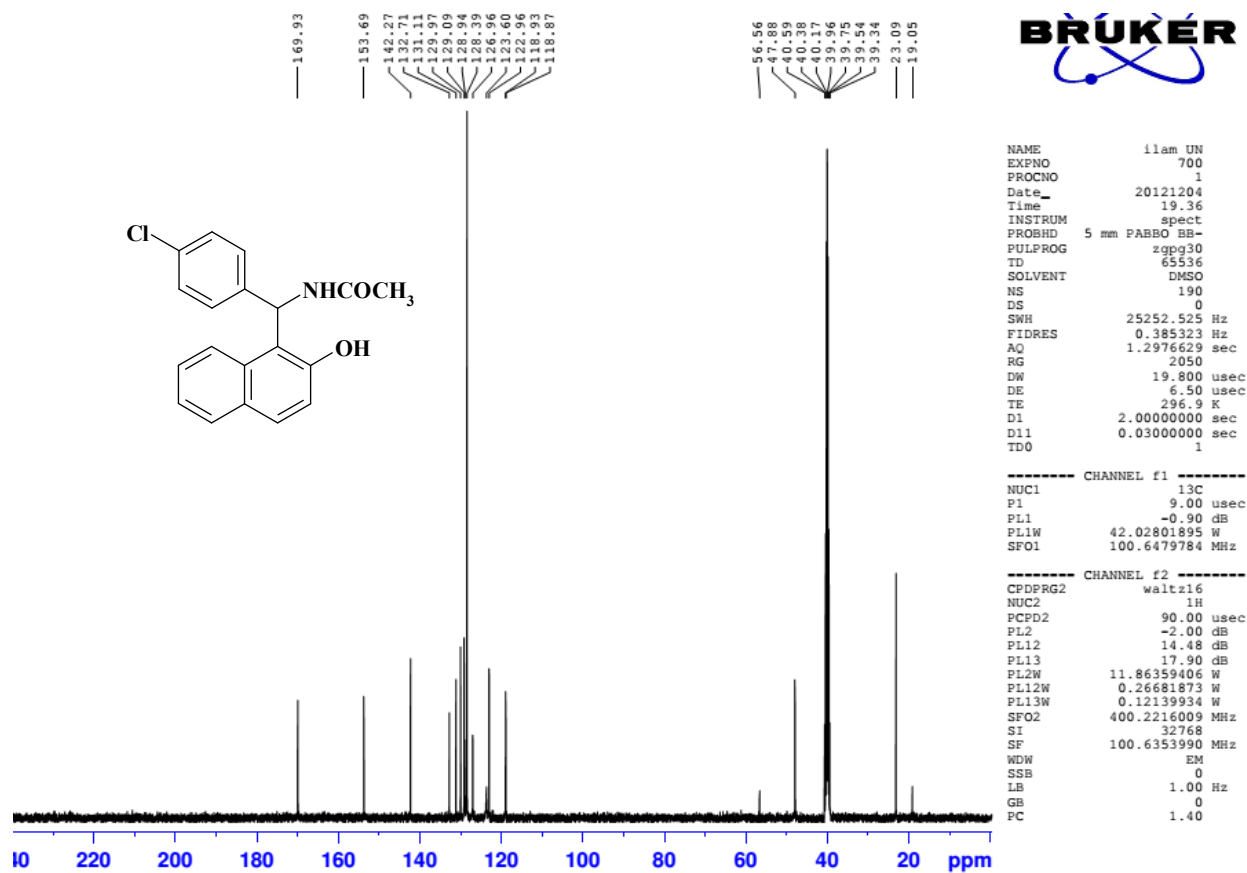
^{1}H NMR of *N*-[(4-hydroxy-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide
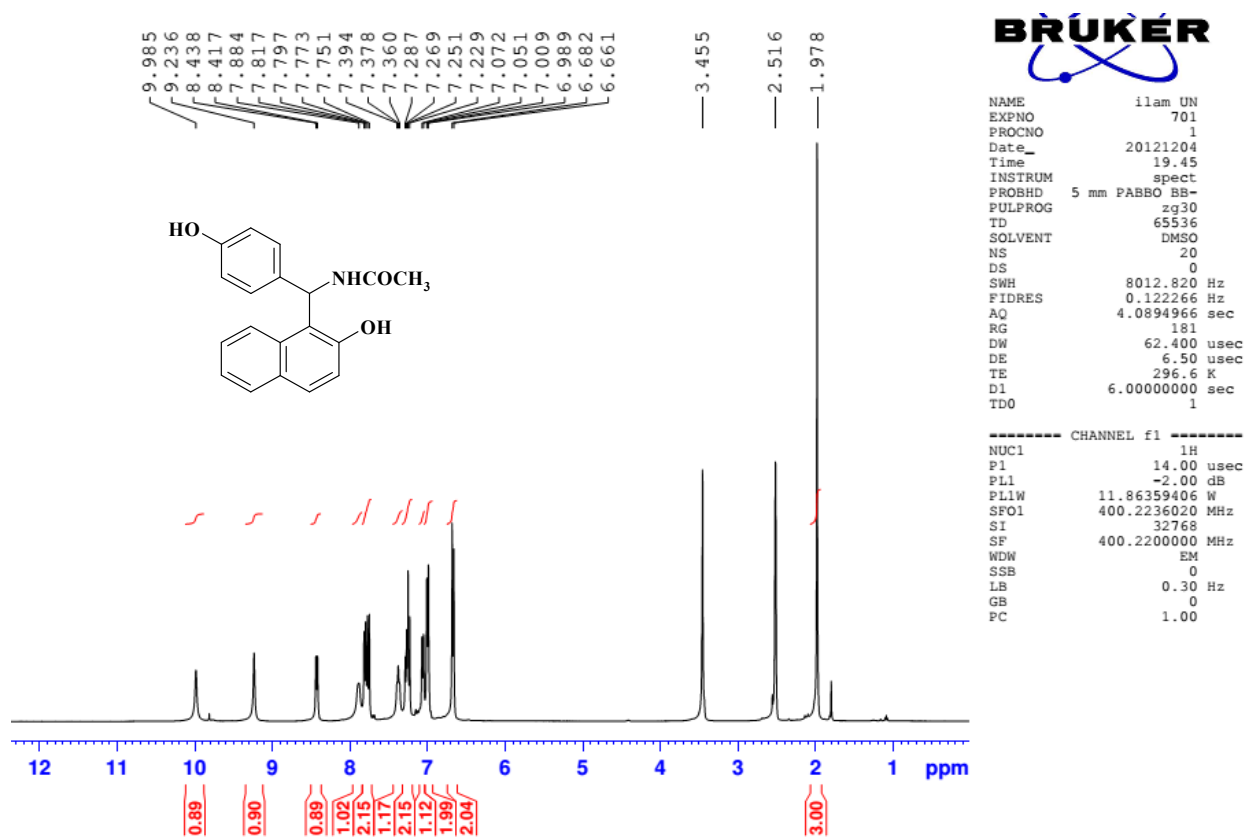
^{13}C NMR of *N*-[(4-hydroxy-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide
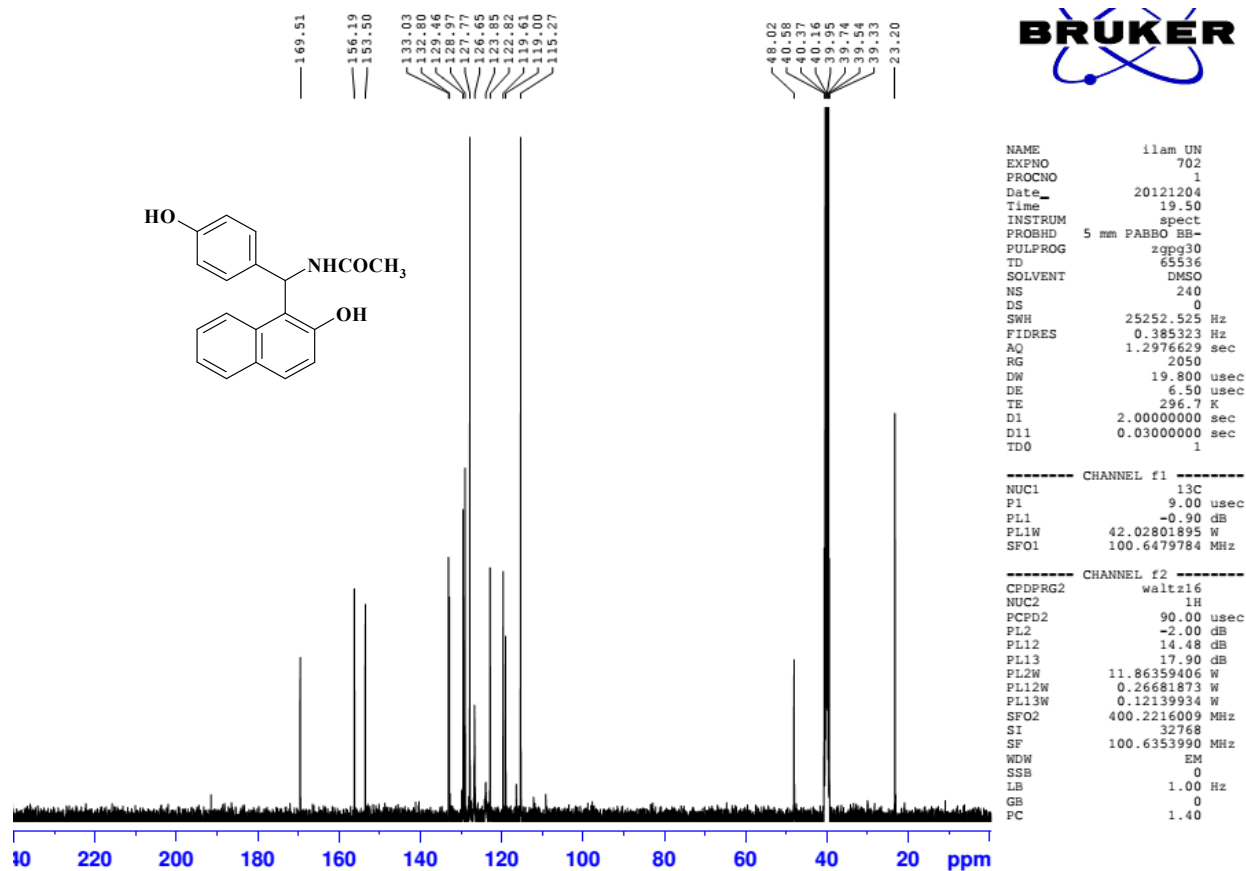
^{1}H NMR of *N*-[(3-nitro-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide
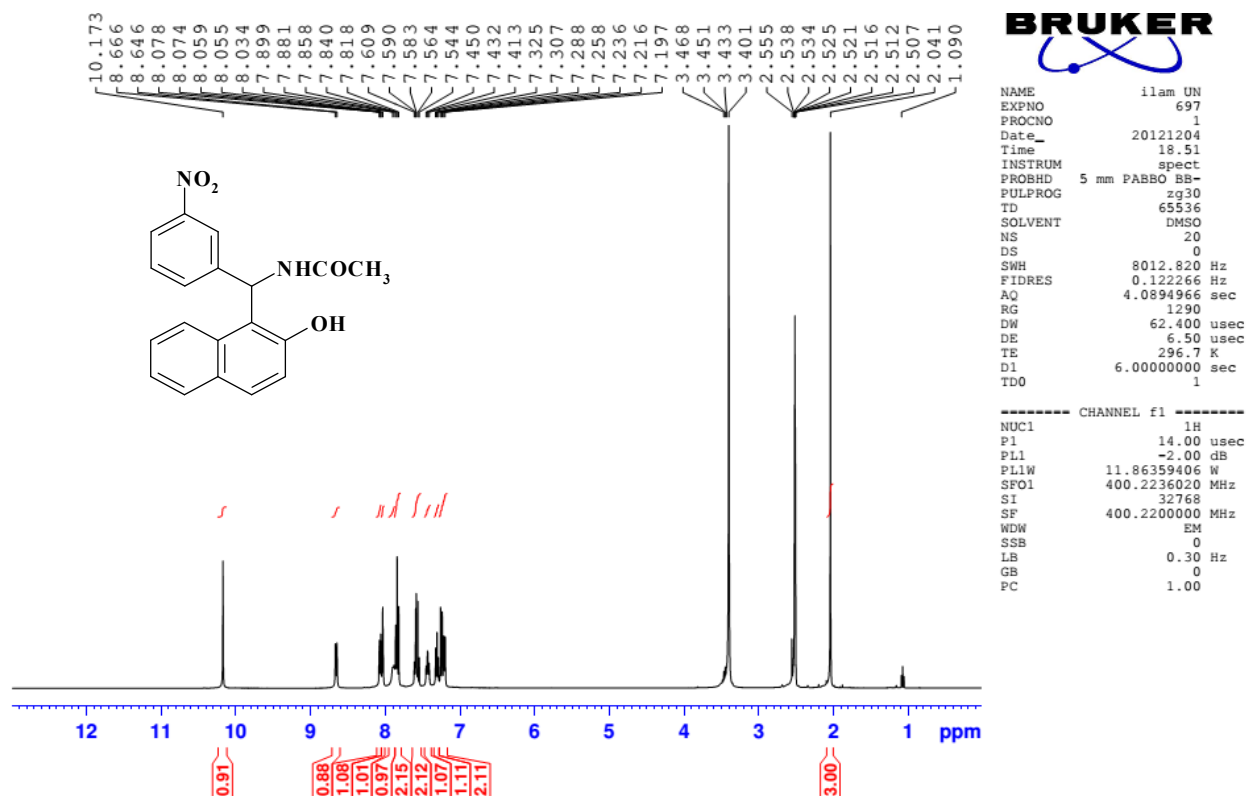
¹³C NMR of *N*-[(3-nitro-phenyl)-(2-hydroxy-naphtalen-1-yl)-methyl]-acetamide
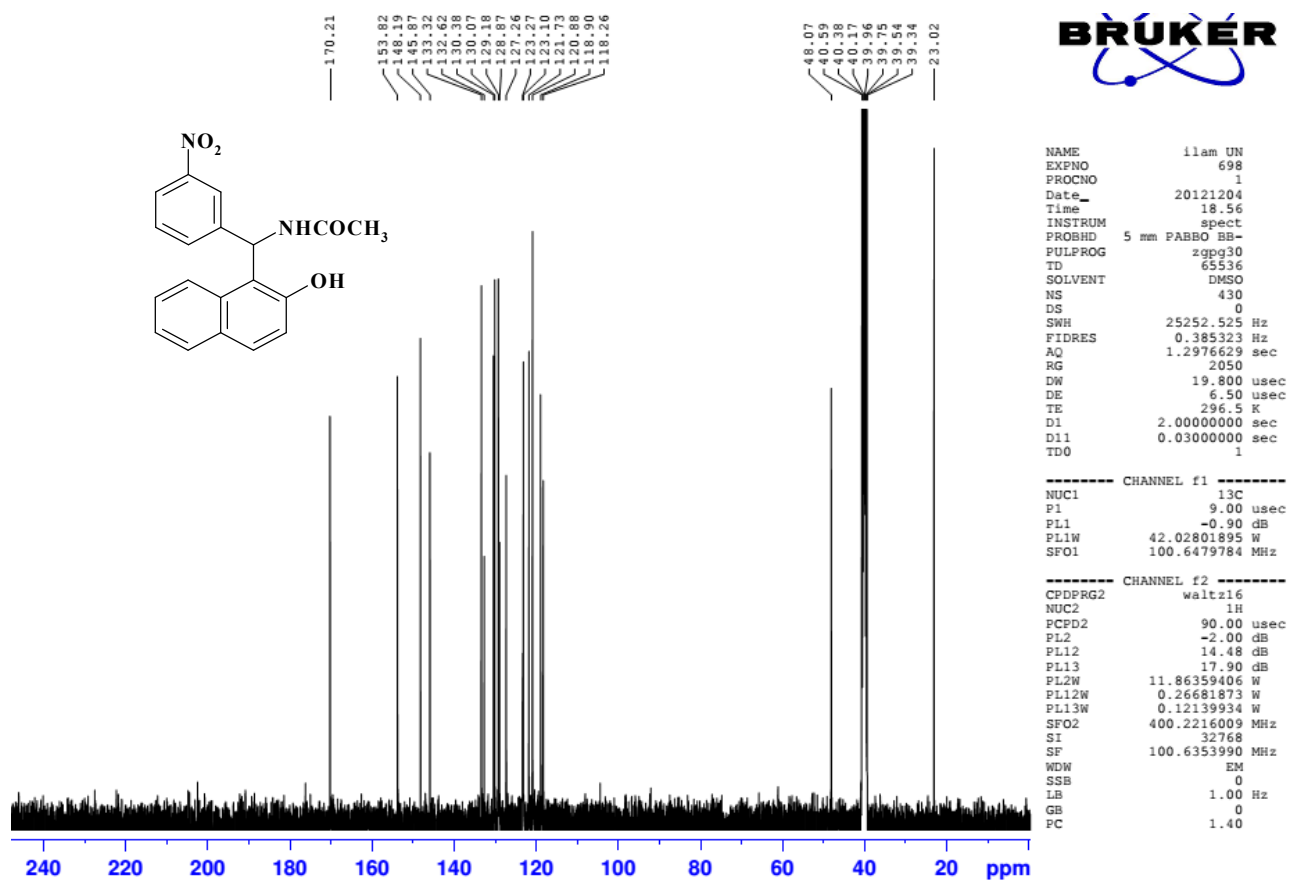
Mass spectroscopy of glycerosulfonic acid
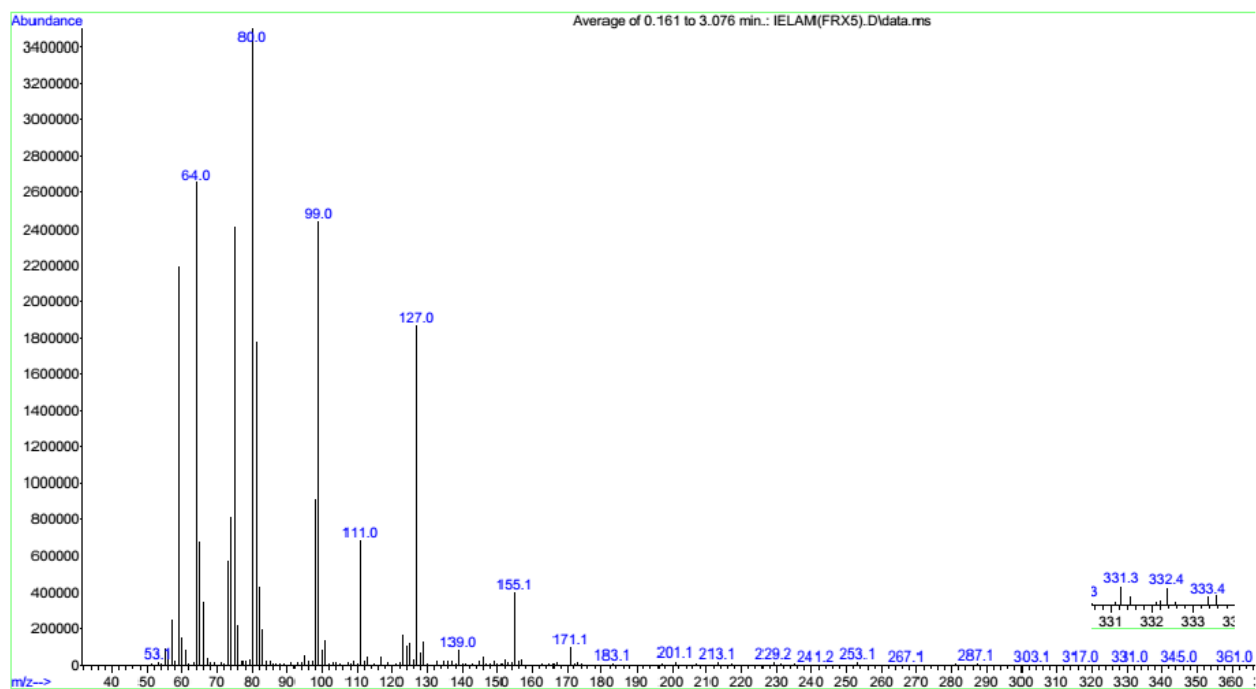
FTIR spectroscopy of glycerol (blue line) and glycerosulfamic acid (red line)