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Aliphatic Aldehydes in Multicomponent Syntheses of 4-Alkyl-Substituted Partially Hydrogenated Quinolines, Fused 4*H*-Pyrans, and 2-Amino-4-ethyl-5-methylbenzene-1,3-dicarbonitrile

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Abstract—The Knoevenagel condensation of aliphatic aldehydes with CH acids, malonodinitrile, cyanothioacetamide, cyclohexane-1,3-dione, dimedone, 4-hydroxycoumarin, 3-aminophenol, and *N*-(cyclohex-1-enyl)morpholine leads to formation of 4-alkyl-substituted partially hydrogenated quinolines, fused 4*H*-pyrans, and 2-amino-4-ethyl-5-methylbenzene-1,3-dicarbonitrile. The structure of the latter was proved by the X-ray diffraction data.

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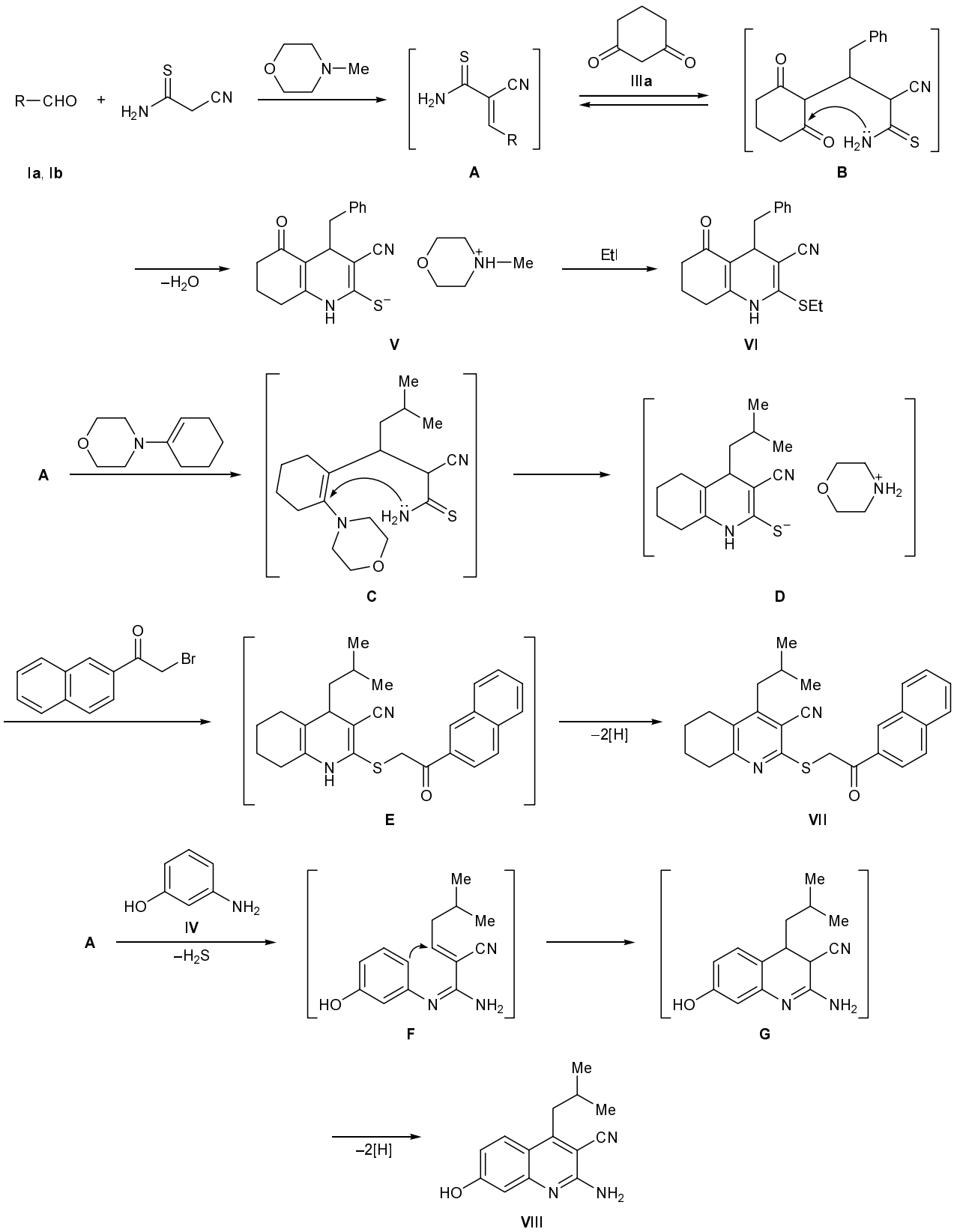
Multicomponent syntheses of organic compounds have now acquired increasing practical importance [1]. In particular, this methodology was successfully applied to the synthesis of aryl(or hetaryl)-substituted chalcogenopyridines [2]. On the other hand, analogous reactions involving aliphatic aldehydes have been studied poorly, presumably because of their high toxicity, high inflammability, and strong tendency to undergo isomerization [3].

In continuation of our studies on the synthesis of alkyl-substituted carbo- and heterocycles via multicomponent condensations [4], in the present work we examined Knoevenagel [5] reactions of aliphatic aldehydes **I** with the following CH acids: malonodinitrile (**II**), cyanothioacetamide, cyclohexane-1,3-dione (**IIIa**), dimedone (**IIIb**), 4-hydroxycoumarin, 3-aminophenol (**IV**), and *N*-(cyclohex-1-enyl)morpholine. Phenylacetaldehyde (**Ia**) reacted with cyanothioacetamide and cyclohexane-1,3-dione (**IIIa**) in ethanol at 20°C in the presence of an equimolar amount of *N*-methylmorpholine to give *N*-methylmorpholinium 4-benzyl-3-cyano-5-oxo-1,4,5,7,8-hexahydroquinoline-2-thiolate (**V**). Presumably, the reaction involves intermediate formation of cyano(2-phenylethylidene)thioacetamide **A** which takes up cyclohexane-1,3-dione (**IIIa**) according to Michael. Michael adduct **B** thus formed undergoes intramolecular cyclization to salt **V** with elimination of water (Scheme 1). The structure of salt **V** follows from its spectral parameters (see Experimental), as well as from chemical transformations. Thus the alkylation of **V** with ethyl iodide afforded the corresponding 2-ethylsulfanylhexahydroquinoline **VI**.

Four-component condensation of isovaleraldehyde (**Ib**), cyanothioacetamide, *N*-(cyclohex-1-enyl)morpholine, and 2-bromoacetylnaphthalene in the presence of *N*-methylmorpholine as catalyst led to formation of substituted 5,6,7,8-tetrahydroquinoline **VII**. The process is likely to include Stork reaction [6] of the Knoevenagel condensation product **A** with *N*-(cyclohexen-1-yl)morpholine, which gave adduct **C**. Intramolecular transamination of the latter afforded salt **D** whose alkylation with bromomethyl 2-naphthyl ketone *in situ* produced sulfide **D**. The latter was likely to be unstable and was oxidized (presumably with atmospheric oxygen) to final tetrahydroquinoline **VII** (Scheme 1).

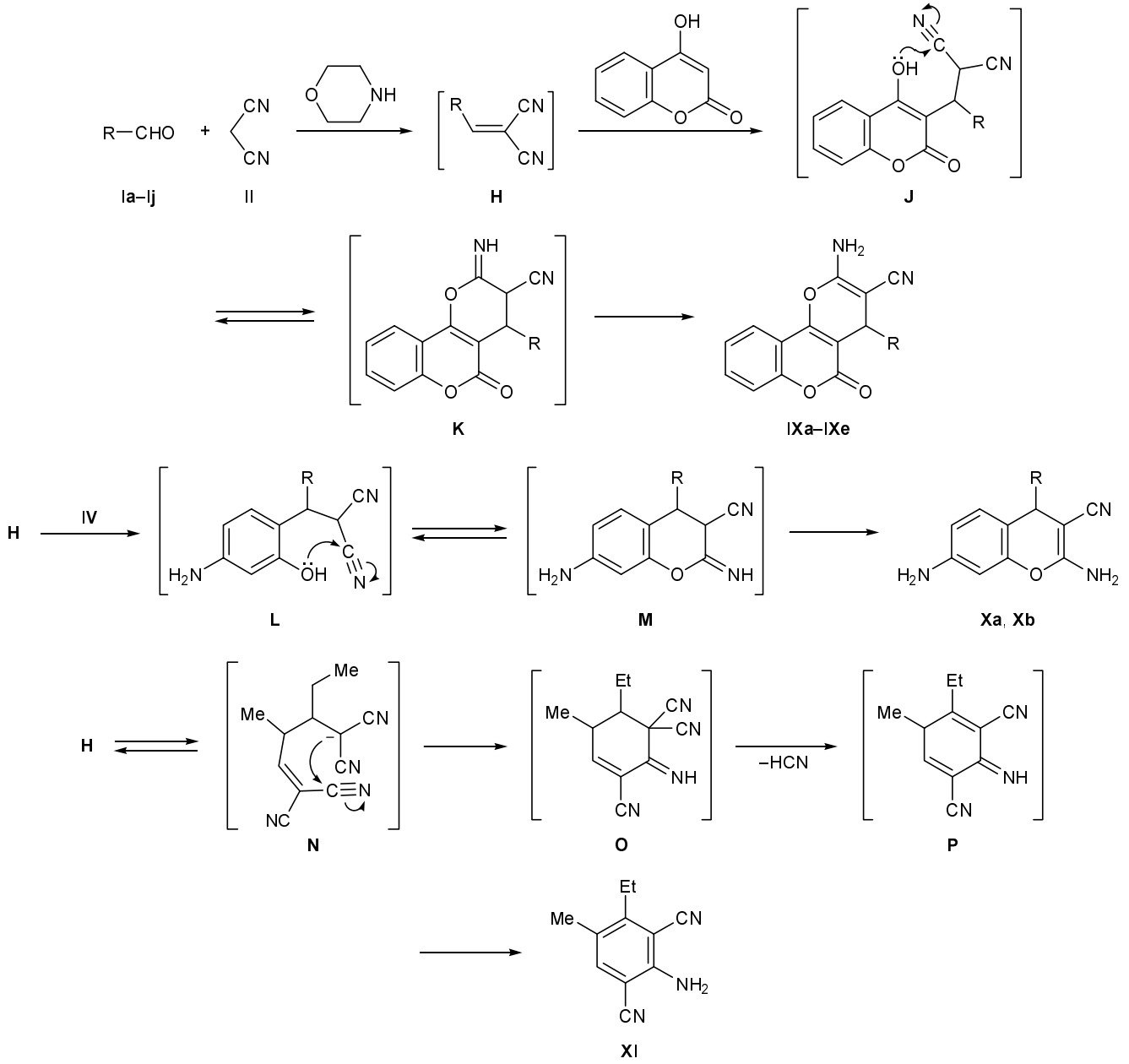
Unexpectedly, by condensation of isovaleraldehyde (**Ib**) with cyanothioacetamide and 3-aminophenol (**IV**) we obtained substituted quinoline **VIII**. It should be noted that arylmethylidenemalonodinitriles are known to react with 3-aminophenol to give 4-aryl-substituted 2-amino-3-cyano-4*H*-1-benzopyrans [7]. The formation of compound **VIII** in this reaction may be

Scheme 1.



I, R = $PhCH_2$ (**a**), Me_2CHCH_2 (**b**).

Scheme 2.



I, R = Et (**c**), Me (**d**), $Me(CH_2)_{10}$ (**e**), $Me(CH_2)_5$ (**f**), EtCH(Me) (**g**), $Me(CH_2)_8$ (**h**), PhCH(Me) (**i**), PrCH(Me) (**j**); **IX**, R = Me (**a**), $Me(CH_2)_{10}$ (**b**), $Me(CH_2)_5$ (**c**), EtCH(Me) (**d**), $Me(CH_2)_8$ (**e**); **X**, R = PrCH(Me) (**a**), PhCH(Me) (**b**).

rationalized as follows. The thioamide group in Knoevenagel condensation product **A** is more reactive than the cyano group. Therefore, initial attack by the amino group of 3-aminophenol is directed at the thioamide group of intermediate **A**, yielding hypothetical structure **F**. Next follows intramolecular Michael reaction leading to formation of 3,4-dihydroquinoline derivative **G** which is stabilized via aromatization to substituted quinoline **VIII** (Scheme 1).

Three-component condensation of aliphatic aldehydes **I** with malonodinitrile (**II**) and 4-hydroxycoumarin in ethanol at 20°C in the presence of morpholine gave fused 4-alkyl-4*H*-pyrans **IX** which are potential biologically active compounds. Their analogs are used in the treatment of cardiovascular diseases and CNS disorders [8], as well as for protection of crops from herbicide damage [9]. In the first step, condensation of aldehyde **I** with malonodinitrile gives

alkene **H** which then reacts with 4-hydroxycoumarin according to the Michael addition pattern. Michael adduct **J** readily undergoes intramolecular ring closure to 2-iminopyran **K**, and the latter is stabilized as the corresponding 2-amino-4-alkyl-4*H*-pyran **IX** (Scheme 2). The reaction direction does not change essentially when 3-aminophenol (**IV**) is used instead of 4-hydroxycoumarin. In this case, the products are fused 4*H*-pyrans **X**. It is reasonable to presume that the process includes intermediate formation of the corresponding Michael adduct **L** and 2-iminopyran **M**.

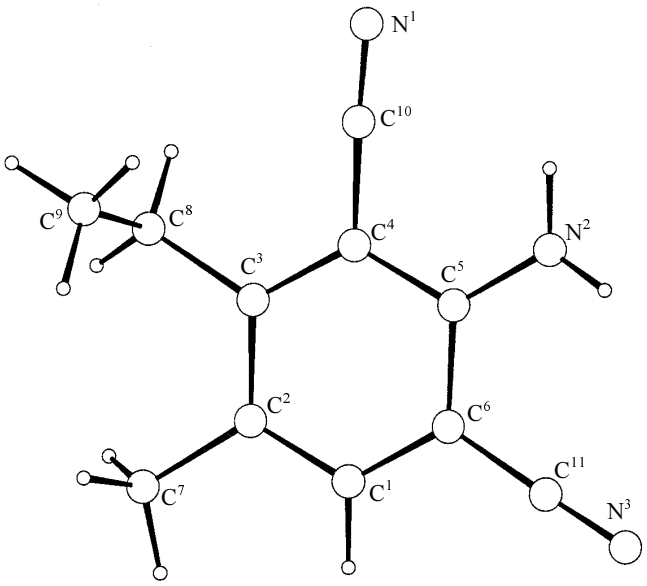


Fig. 1. Structure of the molecule of 2-amino-4-ethyl-5-methylbenzene-1,3-dicarbonitrile (**XI**) according to the X-ray diffraction data. Principal bond lengths (Å) and bond angles (deg): N^2–C^5 1.354(2), C^1–C^2 1.380(2), C^1–C^6 1.392(2), C^2–C^3 1.399(2), C^3–C^4 1.404(2), C^4–C^5 1.405(2), C^5–C^6 1.406(2); $C^2C^1C^6$ 122.75(14), $C^1C^2C^3$ 118.23(13), $C^2C^3C^4$ 118.99(13), $C^3C^4C^5$ 123.29(13), $C^4C^5C^6$ 116.11(13), $C^1C^6C^5$ 120.58(14).

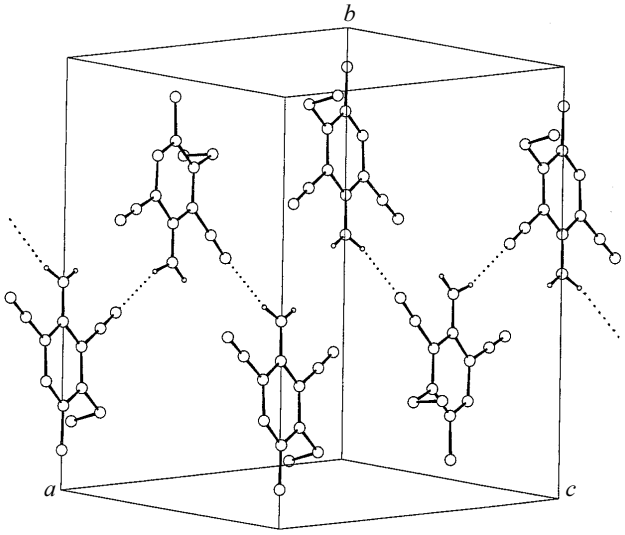


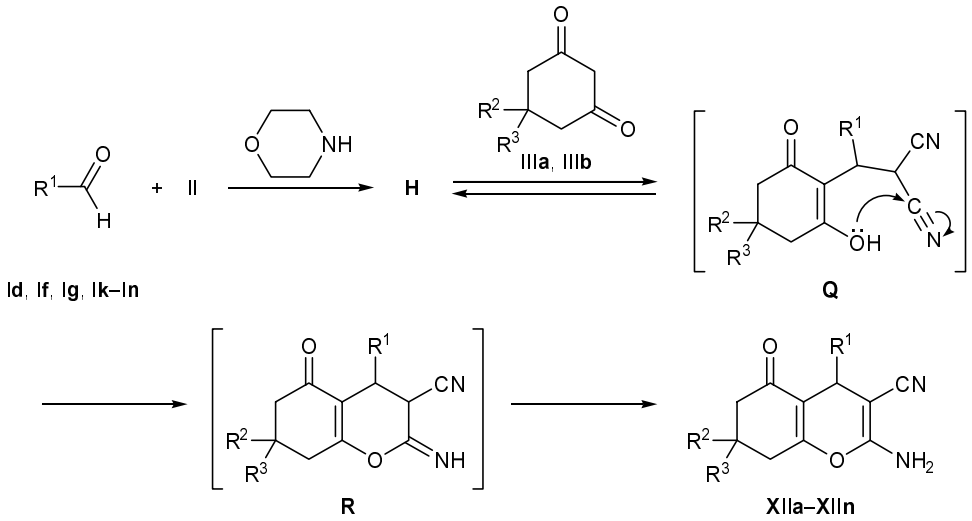
Fig. 2. Packing of 2-amino-4-ethyl-5-methylbenzene-1,3-dicarbonitrile (**XI**) molecules in crystal; dotted lines denote intermolecular hydrogen bonds.

In order to confirm the formation in the above reaction of Knoevenagel condensation products, alkenes **H**, we examined the two-component reaction of propanal (**Ic**) with malonodinitrile (**II**). The reaction conditions were the same as in the above three-component syntheses. However, instead of the expected propylidenemalonodinitrile **H** (R = Et) we isolated substituted aniline **XI**. Arylmethylidenemalonodinitriles are well known [10] and are widely used in organic synthesis [11]. Obviously, the formation of compound **XI** is the result of fast dimerization of alkene **H** according to Michael. Salt-like adduct **N** thus formed undergoes intramolecular cyclization to substituted cyclohexene **O**. Elimination of hydrogen cyanide from the latter yields cyclohexa-1,4-diene **P** which is stabilized as tautomeric 2-amino-4-ethyl-5-methylbenzene-1,3-dicarbonitrile (**XI**) (Scheme 2).

The structure of compound **XI** was determined by X-ray analysis (Fig. 1). The bond lengths and bond angles in molecule **XI** have their usual values [12]. The N^2 atom has planar–trigonal configuration within the experimental error [the sum of the bond angles is 357.4(3.8)°]. Effective conjugation between the lone electron pair on N^2 and π system of the aromatic ring leads to appreciable shortening of the N^1–C^1 bond {1.354(2) Å against the standard value 1.45 Å for purely single N(sp^2)–C(sp^2) bonds [13]} and reduction of the endocyclic $C^4C^5C^6$ bond angle to 116.1(1)° [14]. Molecules **XI** in crystal give rise to infinite chains via intermolecular hydrogen bonds N^2–H $\cdots$ N^1 (Fig. 2) with the following parameters: N^2–H 0.89(2), $N^1\cdots N^2$ 3.077(2), $N^1\cdots$H 2.22(2) Å; $\angle N^1HN^2$ 160.4(8)°.

The condensation of aliphatic aldehydes **Id**, **If**, **Ig**, and **Ik**–**In** with malonodinitrile (**II**) and cyclic 1,3-diketone **IIIa** or **IIIb** as CH acid gave 4-alkyl-substituted fused 2-amino-4*H*-pyrans **XII** which are structural analogs of 4-aryl derivatives described in [15]. As in the above reactions, initially formed Knoevenagel product **H** acts as Michael acceptor in the subsequent transformations. The respective Michael donor, cyclohexane-1,3-dione (**IIIa**) or dimedone (**IIIb**), reacts with alkene **H** to form adduct **Q**. The latter undergoes intramolecular ring closure to 2-iminopyran **R** which is stabilized as the corresponding amino tautomer, 2-amino-4*H*-pyran **XII** (Scheme 3).

Scheme 3.



I, R^1 = Et_2CH (**k**), $Ph(CH_2)_2$ (**l**), $Me(CH_2)_6$ (**m**), $MeCH(Ph)CH_2$ (**n**); **III**, $R^2 = R^3$ = H (**a**), Me (**b**); **XII**, R^1 = Me (**a**), Et_2CH (**b**, **n**), $Me(CH_2)_5$ (**c**, **l**), PrCH(Me) (**d**, **j**), $Ph(CH_2)_2$ (**e**, **i**), $MeCH(Ph)CH_2$ (**f**), PhCH(Me) (**g**), $Me(CH_2)_6$ (**h**, **k**), EtCH(Me) (**m**); $R^2 = R^3$ = H (**a**–**h**), Me (**i**–**n**).

The structure of compounds **XII** is confirmed by spectral data. The IR spectra of substituted 4*H*-pyrans **XII** contain absorption bands due to stretching vibrations of the carbonyl group and conjugated cyano group at 1700–1715 and 2190–2222 cm^{-1}, respectively, and absorption bands belonging to stretching and bending vibrations of the amino group appear at 3198–3451 and 1637–1652 cm^{-1}, respectively. Compounds **XII** showed in the ^1H NMR spectra signals from protons in the substituents and characteristic signals from protons of the amino group as a broadened singlet at δ 6.61–6.89 ppm [16].

Thus we have demonstrated that aliphatic aldehydes can successfully be used in multicomponent syntheses of 4-alkyl-substituted partially hydrogenated quinolines, fused 4*H*-pyran derivatives, and 2-amino-4-ethyl-5-methyl-benzene-1,3-dicarbonitrile.

EXPERIMENTAL

The IR spectra were recorded on an IKS-40 instrument from samples dispersed in mineral oil. The ^1H NMR spectra were measured on Bruker WP-100SY (100 MHz; compounds **IXe**, **XIId**, **XIIf**, **XIIh**, and **XIIk**), Bruker WM-250 (250.13 MHz; **VII**, **IXc**, **IXd**, **Xa**, **Xb**), Bruker AM-300 (300 MHz; **V**, **VI**, **XIIa**, **XIIg**), Varian Mercury-400 (400.397 MHz; **VIII**, **IXa**, **XIIc**, **XIIj**, **XIIl**–**XIIn**), and Bruker DRX-500 spectrometers (500.13 MHz; **IXb**, **XI**, **XIIe**, **XIIi**) from solutions in DMSO-d_6 using TMS as internal reference. The mass spectra were obtained on a Kratos MS-890 mass spectrometer (electron impact, 70 eV; direct sample admission into the ion source; compounds **VI**, **Xa**, and **XI**) and on a Chrommas GC/MS-Hewlett–Packard 5890/5972 system (electron impact, 70 eV; HP-5MS column; compound **VIII** was injected as a solution in CH_2Cl_2). The melting points were determined on a Kofler melting point apparatus. The progress of reactions and the purity of products were monitored by TLC on Silufol UV-254 plates using acetone–hexane (3:5) as eluent; spots were visualized by treatment with iodine vapor or under UV light.

X-Ray diffraction study of a single crystal of compound **XI** (0.25×0.31×0.44 mm) was performed at room temperature on an Enraf–Nonius CAD-4 automatic four-circle diffractometer (CuK_α irradiation, scan rate ratio $2\theta/\omega$ = 1.2, θ_{max} = 65°, spherical segment $0 \le h \le 14$, $0 \le k \le 17$, $-13 \le l \le 13$). Total of 1860 reflections were measured, 1699 of which were symmetry-independent (R_{int} = 0.014). Monoclinic crystals with the following unit cell parameters: a = 12.034(9), b = 14.793(9), c = 11.356(9) Å; β = 94.88(9)°; V = 2014.2 $Å^3$; M 185.23; Z = 8, d_{calc} = 1.22 g/cm^3; μ = 5.68 cm^{-1}; $F(000)$ = 785.99; space group $C2/c$. The structure was solved by the direct method and was refined by the least-squares procedure in full-matrix anisotropic approximation using CRYSTALS software package [17]; 1403 reflections with $I > 3(I)$ were used in the refinement procedure (171 refined parameters, 8.2 reflections per parameter). All hydrogen atoms

Coordinates of non-hydrogen atoms and their equivalent isotropic thermal parameters U_{eq} (Å^2) in the structure of 2-amino-4-ethyl-5-methylbenzene-1,3-dicarbonitrile (**XI**)

Atom	*x*	*y*	*z*	U_{eq}
N^1	0.09193(12)	0.41419(8)	0.37868(11)	0.0698
N^2	0.13625(11)	0.45421(8)	0.67809(11)	0.0587
N^3	0.18189(11)	0.38686(9)	0.97835(11)	0.0691
C^1	0.1487(1)	0.21529(9)	0.76734(13)	0.0521
C^2	0.1292(1)	0.17011(8)	0.66128(12)	0.0511
C^3	0.11344(9)	0.22131(8)	0.55760(11)	0.0455
C^4	0.11655(9)	0.31603(8)	0.5656(1)	0.0420
C^5	0.13314(9)	0.36281(8)	0.6733(1)	0.0429
C^6	0.14971(9)	0.30915(8)	0.7755(1)	0.0453
C^7	0.12416(16)	0.0681(1)	0.65936(19)	0.0719
C^8	0.08747(11)	0.1780(1)	0.43866(13)	0.0556
C^9	–0.03821(13)	0.16914(13)	0.40909(16)	0.0709
C^{10}	0.1017(1)	0.36904(8)	0.46028(11)	0.0488
C^{11}	0.1678(1)	0.35168(9)	0.88824(12)	0.0520

were visualized from the difference synthesis of electron density, and their positions were refined in isotropic approximation. Absorption by the crystal was taken into account by the azimuthal scanning technique [18]. Chebyshev's weight scheme [19] with five parameters (3.08, 1.90, 2.70, 0.34, and 0.67) was used in the refinement. The final divergence factors were $R = 0.034$, $R_W = 0.038$, GOF = 1.095; residual electron density from the Fourier difference series: 0.11 and –0.11 *e*/Å^3. The coordinates of non-hydrogen atoms are listed in table.

***N*-Methylmorpholinium 4-benzyl-3-cyano-5-oxo-1,4,5,6,7,8-hexahydroquinoline-2-thiolate (V).** Cyanothioacetamide, 1.0 g (10 mmol), and *N*-methylmorpholine, 1.1 ml (10 mmol), were added to a solution of 1.12 ml (10 mmol) of phenylacetaldehyde (**Ia**) in 15 ml of ethanol under stirring at 20°C. The mixture was stirred for 20 min and 1.12 ml (10 mmol) of cyclohexane-1,3-dione (**IIIa**) was added. The mixture was stirred for 5 min (until it became homogeneous) and was left to stand for 24 h. The precipitate was filtered off and washed with ethanol and acetone. Salt **V** thus obtained was used in further transformations without additional purification. Yield 2.94 g (74%), mp 156–158°C. IR spectrum, ν, cm^{-1}: 3314 (NH), 2196 (C≡N), 1713 (C=O). ^{1}H NMR spectrum, δ, ppm: 1.77 m (2H, 7-H), 2.14 m (4H, CH_2), 2.58 d (2H, C**H**$_2$Ph, J = 6.12 Hz), 2.69 s (3H, Me), 3.04 t (4H, CH_2NCH_2, J = 4.41 Hz), 3.58 t (1H, 4-H), 3.74 t (4H, CH_2OCH_2), 6.87–7.29 m (5H, H_{arom}), 7.96 br.s (1H, NH); signal from the $^+$NH proton was not observed, presumably because of fast H–D exchange. Found, %: C 60.30; H 6.75; N 10.38. $C_{22}H_{27}N_3O_2S$. Calculated, %: C 66.47; H 6.85; N 10.57.

4-Benzyl-2-ethylsulfanyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carbonitrile (VI). Salt **V**, 3.98 g (10 mmol), was dissolved in 10 ml of DMF, 0.8 ml (10 mmol) of ethyl iodide was added under stirring, and the mixture was stirred for 2 h and was left to stand for 48 h. The mixture was then slowly diluted with an equal volume of water under stirring, and a solid precipitated. The mixture was left to stand for 4 h, and the precipitate was filtered off and washed with water, ethanol, and hexane. Yield 2.56 g (79%), mp 190–192°C (from EtOH). IR spectrum, ν, cm^{-1}: 3313 (NH), 2204 (C≡N), 1718 (C=O). ^{1}H NMR spectrum, ppm: 1.19 t (3H, Me, J = 7.02 Hz), 1.92 m (2H, 7-H), 2.22 m (4H, CH_2), 2.63 d (2H, C**H**$_2$Ph, J = 6.04 Hz), 2.85 q (2H, SCH_2, J = 7.02 Hz), 3.83 t (1H, 4-H), 6.99 m (2H, H_{arom}), 7.21 m (3H, H_{arom}), 9.03 br.s (1H, NH). Mass spectrum, *m*/*z* (I_{rel}, %): $[M]^+$ absent, 233 (100) $[M - PhCH_2]^+$, 205 (49.8), 176 (8.1), 116 (12.4), 91 (35.7) $[PhCH_2]^+$, 77 (6.2) $[Ph]^+$, 65 (19.3), 51 (7.4). Found, %: C 70.15; H 6.02; N 8.51. $C_{19}H_{20}N_2OS$. Calculated, %: C 70.34; H 6.21; N 8.63.

4-Isobutyl-2-[2-(2-naphthyl)-2-oxoethylsulfanyl]-5,6,7,8-tetrahydroquinoline-3-carbonitrile (VII). Cyanothioacetamide, 1.0 g (10 mmol), and *N*-methylmorpholine, 1.1 ml (10 mmol), were added to a solution of 1.08 ml (10 mmol) of isovaleraldehyde (**Ib**) in 15 ml of ethanol under stirring at 20°C. The mixture was left to stand for 24 h, 2.49 g (10 mmol) of 2-bromo-1-(2-naphthyl)ethanone was added, and the mixture was stirred for 4 h and was slowly diluted with an equal volume of water. The precipitate was filtered off and washed with water, ethanol, and hexane. Yield 3.32 g (80%), mp 214–216°C (from AcOH). IR spectrum, ν, cm^{-1}: 2225 (C≡N), 1709 (C=O). ^{1}H NMR spectrum, ppm: 0.95 d (6H, Me, J = 5.13 Hz), 1.72 m (4H, CH_2), 1.95 m [1H, C**H**$(Me)_2$], 2.54 m (4H, CH_2), 2.66 d (2H, C**H**$_2$CH, J = 5.16 Hz), 4.87 s (2H, SCH_2), 7.56–7.77 m (3H, H_{arom}), 7.98 m (2H, H_{arom}), 8.11 d (1H, H_{arom}, J = 6.02 Hz), 8.76 s (1H, 1-H, naphthyl). Found, %: C 75.19; H 6.21; N 6.58. $C_{26}H_{26}N_2OS$. Calculated, %: C 75.33; H 6.32; N 6.76.

2-Amino-7-hydroxy-4-isobutylquinoline-3-carbonitrile (VIII) was synthesized as described above for salt **V** using 1.09 g (10 mmol) of 3-aminophenol instead of cyclohexane-1,3-dione (**IIIa**). The precip-

itate was washed with ethanol and hexane. Yield 1.47g (61%), mp 273–275°C (from *i*-PrOH). IR spectrum, ν, cm^{-1}: 3606 (OH); 3442, 3358, 3190 (NH_2); 2224 (C≡N); 1632 (δNH_2). ^1H NMR spectrum, δ, ppm: 1.01 d (6H, Me, J = 5.08 Hz), 2.04 m [1H, C**H**$(Me)_2$], 2.97 d (2H, CH_2, J = 5.04 Hz), 6.52 br.s (2H, NH_2), 6.78 s (1H, 8-H), 6.99 d (1H, 5-H, J = 7.94 Hz), 7.72 d (1H, 6-H), 10.12 br.s (1H, OH). Mass spectrum, *m*/*z* (I_{rel}, %): 242 (100) $[M + 1]^+$. Found, %: C 69.57; H 6.14; N 17.25. $C_{14}H_{15}N_3O$. Calculated, %: C 69.69; H 6.27; N 17.41.

4-Substituted 2-amino-5-oxo-4,5-dihydropyrano-[3,2-*c*]chromene-3-carbonitriles IXa–IXe (*general procedure*). Morpholine, 0.09 ml (10 mmol), was added to a mixture of 10 mmol of aldehyde **Ic**–**Ij** and 0.66 g (10 mmol) of malonodinitrile (**II**) in 15 ml of ethanol under stirring at 20°C. The mixture was stirred for 5 min, 1.62 g (10 mmol) of 4-hydroxycoumarin was added, and the mixture was stirred for 30 min and was left to stand for 24 h. The colorless crystalline product was filtered off, washed with ethanol and hexane, and recrystallized from ethanol.

2-Amino-4-methyl-5-oxo-4,5-dihydropyrano-[3,2-*c*]chromene-3-carbonitrile (IXa). Yield 2.34 g (92%), mp 218–220°C. IR spectrum, ν, cm^{-1}: 3410, 3344, 3205 (NH_2); 2201 (C≡N); 1718 (C=O); 1647 (δNH_2). ^1H NMR spectrum, δ, ppm: 1.38 d (3H, Me, J = 7.21 Hz), 3.39 q (1H, 4-H, J = 7.21 Hz), 7.05 br.s (2H, NH_2), 7.40 m (2H, H_{arom}), 7.66 t (1H, H_{arom}, J = 8.01 Hz), 7.84 d (1H, H_{arom}, J = 8.01 Hz). Found, %: C 65.98; H 4.15; N 10.84. $C_{14}H_{10}N_2O_3$. Calculated, %: C 66.14; H 3.96; N 11.02.

2-Amino-5-oxo-4-undecyl-4,5-dihydropyrano-[3,2-*c*]chromene-3-carbonitrile (IXb). Yield 3.55 g (90%), mp 128–130°C. IR spectrum, ν, cm^{-1}: 3407, 3302, 3188 (NH_2); 2197 (C≡N); 1715 (C=O); 1639 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.86 t (3H, Me, J = 7.12 Hz), 1.15–1.38 m (18H, CH_2), 1.58 m and 1.75 m (1H each, 4-CH_2), 3.44 t (1H, 4-H, J = 6.98 Hz), 7.16 br.s (2H, NH_2), 7.45 m (2H, H_{arom}), 7.70 t (1H, H_{arom}, J = 7.99 Hz), 7.83 d (1H, H_{arom}, J = 7.99 Hz). Found, %: C 72.87; H 7.49; N 6.95. $C_{24}H_{30}N_2O_3$. Calculated, %: C 73.07; H 7.66; N 7.10.

2-Amino-4-hexyl-5-oxo-4,5-dihydropyrano-[3,2-*c*]chromene-3-carbonitrile (IXc). Yield 2.89 g (89%), mp 170–171°C. IR spectrum, ν, cm^{-1}: 3396, 3318, 3202 (NH_2); 2200 (C≡N); 1717 (C=O); 1639 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.82 t (3H, Me, J = 7.08 Hz), 1.11–1.37 m (8H, CH_2), 1.54 m and 1.72 m (1H each, 4-CH_2), 3.42 t (1H, 4-H, J = 7.02 Hz), 7.18 br.s (2H, NH_2), 7.45 m (2H, H_{arom}), 7.70 t (1H, H_{arom}, J = 7.95 Hz), 7.81 d (1H, H, H_{arom}, J = 7.95 Hz). Found, %: C 70.17; H 6.04; N 8.50. $C_{19}H_{20}N_2O_3$. Calculated, %: C 70.35; H 6.21; N 8.64.

2-Amino-4-(2-methylpropyl)-5-oxo-4,5-dihydropyrano[3,2-*c*]chromene-3-carbonitrile (IXd). Yield 2.58 g (87%), mp 202–204°C. IR spectrum, ν, cm^{-1}: 3399, 3286, 3197 (NH_2); 2212 (C≡N); 1705 (C=O); 1640 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.70 d (3H, Me, J = 6.98 Hz), 0.92 t (3H, Me, J = 7.01 Hz), 1.25–1.59 m (2H, CH_2), 1.74 m (1H, CH), 3.39 d and 3.48 d (0.5H each, 4-H, J = 4.82 Hz; presumably, the signal is doubled due to the presence of two diastereoisomers), 7.23 br.s (2H, NH_2), 7.48 m (2H, H_{arom}), 7.70 t (1H, H_{arom}, J = 7.01 Hz), 7.73 d (1H, H_{arom}, J = 7.01 Hz). Found, %: C 68.78; H 5.25; N 9.31. $C_{17}H_{16}N_2O_3$. Calculated, %: C 68.91; H 5.44; N 9.45.

2-Amino-4-nonyl-5-oxo-4,5-dihydropyrano-[3,2-*c*]chromene-3-carbonitrile (IXe). Yield 3.23 g (88%), mp 127–128°C. IR spectrum, ν, cm^{-1}: 3400, 3281, 3195 (NH_2); 2207 (C≡N); 1710 (C=O); 1644 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.81 t (3H, Me, J = 7.12 Hz), 1.12–1.33 m (14H, CH_2), 1.63 m (2H, 4-CH_2), 3.43 t (1H, 4-H, J = 6.02 Hz), 7.27 br.s (2H, NH_2), 7.45 m (2H, H_{arom}), 7.69 d.d (1H, H_{arom}, J = 7.15, 2.03 Hz), 7.77 d.d (1H, H_{arom}, J = 7.14, 1.96 Hz). Found, %: C 71.89; H 6.96; N 9.32. $C_{22}H_{26}N_2O_3$. Calculated, %: C 72.11; H 7.15; N 7.64.

4-Substituted 2,7-diamino-4*H*-chromene-3-carbonitriles Xa and Xb were synthesized as described above for compounds **IX** from aldehydes **Ij** and **Ii**, respectively, and 1.09 g (10 mmol) of 3-aminophenol (**IV**) instead of 4-hydroxycoumarin. The products were recrystallized from ethanol.

2,7-Diamino-4-(1-methylbutyl)-4*H*-chromene-3-carbonitrile (Xa). Yield 1.98 g (77%), mp 151–153°C. IR spectrum, ν, cm^{-1}: 3211–3402 (NH_2), 2213 (C≡N), 1648 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.61 d (3H, Me, J = 6.84 Hz), 0.85 t (3H, Me, J = 7.05 Hz), 1.12 m (1H, CH), 1.35–1.66 m (4H, CH_2), 3.18 d (1H, 4-H, J = 3.38 Hz), 5.09 br.s (2H, 2-NH_2), 6.18 s (1H, 8-H), 6.32 d (1H, 5-H, J = 5.26 Hz), 6.51 br.s (2H, 7-NH_2), 6.80 d (1H, 6-H, J = 5.26 Hz). Mass spectrum, *m*/*z* (I_{rel}, %): 258 (7.1) $[M + 1]^+$, 257 (4.6) $[M]^+$, 187 (39.5), 186 (100) $[M - MeCHPr]^+$, 143 (18.3), 131 (19.7), 114 (12.5), 82 (10.9), 72 (5.7) $[MeCHPr + 1]^+$, 66 (4.4) $[CH_2(CN)_2]^+$. Found, %: C 69.88; H 7.27; N 16.18. $C_{15}H_{19}N_3O$. Calculated, %: C 70.01; H 7.44; N 16.33.

2,7-Diamino-4-(1-phenylethyl)-4*H*-chromene-3-carbonitrile (Xb). Yield 1.87 g (64%), mp 160–162°C.

IR spectrum, ν, cm^{-1}: 3198–3422 (NH_2), 2215 (C≡N), 1648 (δNH_2). ^1H NMR spectrum, δ, ppm: 1.21 d (3H, Me, J = 4.95 Hz), 2.87 m (1H, C**H**Me), 3.42 d (1H, 4-H, J = 4.72 Hz), 5.09 br.s (2H, NH_2), 6.08 s (1H, 8-H), 6.29 d (1H, 5-H, J = 5.09 Hz), 6.42 br.s (2H, 2-NH_2), 6.73 d (1H, 6-H, J = 5.09 Hz), 6.93 m (2H, H_{arom}), 7.19 m (3H, H_{arom}). Found, %: C 74.02; H 5.69; N 14.30. $C_{18}H_{17}N_3O$. Calculated, %: C 74.21; H 5.88; N 14.42.

2-Amino-4-ethyl-5-methylbenzene-1,3-dicarbonitrile (XI). Morpholine, 0.09 ml (1 mmol), was added to a mixture of 0.73 ml (10 mmol) of propionaldehyde (**Ic**) and 0.66 g (10 mmol) of malonodinitrile (**II**) in 15 ml of ethanol under stirring at 20°C. The mixture was stirred for 10 min and left to stand for 48 h, and the precipitate was filtered off and washed with ethanol and hexane. The product showed fluorescence under UV irradiation. Yield 1.24 g (67%), mp 122–123°C (from EtOH). IR spectrum, ν, cm^{-1}: 3451, 3360, 3275 (NH_2); 2222, 2210 (C≡N); 1643 (δNH_2). ^1H NMR spectrum, δ, ppm: 1.16 t (3H, **Me**CH_2, J = 7.62 Hz), 2.17 s (3H, Me), 2.73 q (2H, CH_2), 6.09 br.s (2H, NH_2), 7.38 s (1H, 6-H). Mass spectrum, m/z (I_{rel}, %): 186 (15) $[M + 1]^+$, 185 (100) $[M]^+$, 184 (23) $[M - 1]^+$, 170 (86), 156 (44), 143 (15), 129 (8), 116 (23), 102 (9), 89 (13), 77 (12), 63 (8), 51 (13), 39 (14). Found, %: C 71.18; H 6.08; N 22.74. $C_{11}H_{11}N_3$. Calculated, %: C 71.33; H 5.99; N 22.68.

4-Substituted 2-amino-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitriles XIIa–XIIn were synthesized as described above for compounds **IX** from the corresponding aldehydes **Id**, **If**, **Ig**, and **Ii**–**In** and 10 mmol of CH acid **IIIa** or **IIIb**. The products were recrystallized from ethanol.

2-Amino-4-methyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIa). Yield 1.63 g (80%), mp 201–202°C (sublimes at 150°C). IR spectrum, ν, cm^{-1}: 3409, 3335, 3203 (NH_2); 2214 (C≡N); 1700 (C=O); 1652 (δNH_2). ^1H NMR spectrum, δ, ppm: 1.10 d (3H, Me, J = 6.88 Hz), 1.82 m (2H, 7-H), 2.25 t (2H, 8-H, J = 6.94 Hz), 2.44 t (2H, 6-H, J = 7.05 Hz), 3.05 q (1H, 4-H, J = 5.48 Hz), 6.81 br.s (2H, NH_2). Found, %: C 64.55; H 6.12; N 13.58. $C_{11}H_{12}N_2O_2$. Calculated, %: C 64.69; H 5.92; N 13.72.

2-Amino-4-(1-ethylpropyl)-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIb). Yield 2.60 g (89%), mp 152–153°C. IR spectrum, ν, cm^{-1}: 3400, 3312, 3195 (NH_2); 2204 (C≡N); 1711 (C=O); 1640 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.84 t and 0.92 t (3H each, Me, J = 6.19 Hz), 1.08–1.49 m (5H, CH_2, CH), 1.92 m (2H, 7-H), 2.32 t (2H, 8-H, J = 6.88 Hz), 2.45 t (2H, 6-H, J = 7.14 Hz), 3.43 d (1H, 4-H, J = 5.51 Hz), 6.89 br.s (2H, NH_2). Found, %: C 69.14; H 7.57; N 10.62. $C_{15}H_{20}N_2O_2$. Calculated, %: C 69.21; H 7.74; N 10.76.

2-Amino-4-hexyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIc). Yield 2.19 g (80%), mp 111–112°C. IR spectrum, ν, cm^{-1}: 3425, 3314, 3180 (NH_2); 2199 (C≡N); 1712 (C=O); 1637 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.84 t (3H, Me, J = 7.21 Hz), 1.05–1.28 m (10H, CH_2), 1.43 m (2H, 7-H), 1.99 m (2H, 8-H), 2.34 t (2H, 6-H, J = 7.07 Hz), 3.18 t (1H, 4-H, J = 4.81 Hz), 6.87 br.s (2H, NH_2). Found, %: C 69.88; H 7.91; N 10.13. $C_{16}H_{22}N_2O_2$. Calculated, %: C 70.04; H 8.08; N 10.21.

2-Amino-4-(1-methylbutyl)-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIId). Yield 2.21 g (85%), mp 130–132°C. IR spectrum, ν, cm^{-1}: 3396, 3288, 3205 (NH_2); 2200 (C≡N); 1714 (C=O); 1642 (δNH_2). ^1H NMR spectrum, δ, ppm: 0.60 d (3H, Me, J = 7.12 Hz), 0.88 t (3H, Me, J = 6.85 Hz), 1.16–1.72 m (5H, CH_2, CH), 1.93 m (2H, 7-H), 2.29 t (2H, 8-H, J = 7.16 Hz), 2.41 t (2H, 6-H, J = 7.24 Hz), 3.11 d and 3.20 d (0.5H each, 4-H, J = 3.18 Hz; the signals is doubled due to the presence of two diastereoisomers), 6.89 br.s (2H, NH_2). Found, %: C 69.04; H 7.65; N 10.82. $C_{15}H_{20}N_2O_2$. Calculated, %: C 69.21; H 7.74; N 10.76.

2-Amino-5-oxo-4-(2-phenylethyl)-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIe). Yield 2.06 g (70%), mp 122–123°C. IR spectrum, ν, cm^{-1}: 3406, 3275, 3212 (NH_2); 2195 (C≡N); 1701 (C=O); 1646 (δNH_2). ^1H NMR spectrum, δ, ppm: 1.66–1.92 m (6H, CH_2, 7-H), 2.29 t (2H, 8-H, J = 7.07 Hz), 2.43 t (2H, 6-H, J = 7.18 Hz), 3.27 t (1H, 4-H, J = 3.05 Hz), 6.75 br.s (2H, NH_2), 7.12 m (3H, H_{arom}), 7.23 m (2H, H_{arom}). Found, %: C 73.28; H 6.04; N 9.33. $C_{18}H_{18}N_2O_2$. Calculated, %: C 73.45; H 6.16; N 9.52.

2-Amino-5-oxo-4-(2-phenylpropyl)-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIf). Yield 1.97 g (64%), mp 132–133°C. IR spectrum, ν, cm^{-1}: 3391, 3270, 3196 (NH_2); 2199 (C≡N); 1710 (C=O); 1644 (δNH_2). ^1H NMR spectrum, δ, ppm: 1.13 d (3H, Me, J = 6.84 Hz), 1.54 m (1H, C**H**Me), 1.95 m (4H, CH_2, 7-H), 2.34 t (2H, 8-H, J = 7.15 Hz), 2.48 t (2H, 6-H, J = 7.02 Hz), 2.92 t (1H, 4-H, J = 3.04 Hz), 6.85 br.s (2H, NH_2), 7.23–7.45 m (5H, H_{arom}). Found, %: C 73.85; H 6.36; N 8.87. $C_{19}H_{20}N_2O_2$. Calculated, %: C 74.00; H 6.54; N 9.08.

2-Amino-5-oxo-4-(1-phenylethyl)-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIg). Yield 2.59 g (88%), mp 149–150°C. IR spectrum, ν, cm^{-1}: 3404, 3262, 3185 (NH_2); 2206 (C≡N); 1713 (C=O); 1643 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 1.10 d (3H, Me, J = 7.20 Hz), 1.96 m (1H, C**H**Me), 2.38 m (4H, 7-H, 8-H), 2.86 t (2H, 6-H, J = 7.11 Hz), 3.41 d (2H, 4-H, J = 3.03 Hz), 6.88 br.s (2H, NH_2), 7.22 m (5H, H_{arom}). Found, %: C 73.22; H 6.02; N 9.35. $C_{18}H_{18}N_2O_2$. Calculated, %: C 73.45; H 6.16; N 9.52.

2-Amino-4-heptyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIh). Yield 2.22 g (77%), mp 116–118°C. IR spectrum, ν, cm^{-1}: 3400, 3285, 3198 (NH_2); 2210 (C≡N); 1715 (C=O); 1647 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 0.87 t (3H, Me, J = 6.87 Hz), 1.15–1.39 m (14H, 6CH_2, 7-H), 1.95 t (2H, 8-H, J = 6.90 Hz), 2.32 t (2H, 6-H, J = 6.95 Hz), 3.18 t (1H, 4-H, J = 3.19 Hz), 6.86 br.s (2H, NH_2). Found, %: C 70.69; H 8.25; N 9.54. $C_{17}H_{24}N_2O_2$. Calculated, %: C 70.80; H 8.39; N 9.71.

2-Amino-7,7-dimethyl-5-oxo-4-(2-phenylethyl)-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIi). Yield 2.55 g (79%), mp 199–200°C. IR spectrum, ν, cm^{-1}: 3411, 3296, 3200 (NH_2); 2212 (C≡N); 1714 (C=O); 1648 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 1.08 s (6H, 7-Me), 1.20 m and 1.35 m (1H each, C**H**$_2$Ph), 2.11 d and 2.23 d (1H each, 6-H, ^2J = 15.62 Hz), 2.31 d and 2.38 d (1H each, 8-H, ^2J = 17.62 Hz), 2.48 m (2H, CH_2), 3.26 t (1H, 4-H, J = 2.81 Hz), 6.79 br.s (2H, NH_2), 7.10 m (3H, H_{arom}), 7.23 m (2H, H_{arom}). Found, %: C 74.40; H 6.65; N 8.51. $C_{20}H_{22}N_2O_2$. Calculated, %: C 74.51; H 6.88; N 8.69.

2-Amino-7,7-dimethyl-4-(1-methylbutyl)-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIj). Yield 2.59 g (90%), mp 148–150°C. IR spectrum, ν, cm^{-1}: 3395, 3284, 3192 (NH_2); 2203 (C≡N); 1713 (C=O); 1649 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 0.66 d (3H, **Me**CH, J = 6.81 Hz), 0.93 t (3H, **Me**CH_2, J = 7.61 Hz), 1.05 s (3H, 7-Me), 1.09 (3H, 7-Me), 1.25 m (1H, C**H**Me), 1.31–1.79 m (4H, CH_2), 2.15 d and 2.24 d (1H each, 6-H, ^2J = 16.02 Hz), 2.35 d and 2.45 d (1H each, 8-H, ^2J = 18.02 Hz), 3.21 d (1H, 4-H, J = 2.81 Hz), 6.65 br.s (2H, NH_2). Found, %: C 70.71; H 8.22; N 9.64. $C_{17}H_{24}N_2O_2$. Calculated, %: C 70.80; H 8.39; N 9.71.

2-Amino-4-heptyl-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIk). Yield 2.82 g (89%), mp 173–174°C (sublimes at 140°C). IR spectrum, ν, cm^{-1}: 3412, 3299, 3205 (NH_2); 2190 (C≡N); 1711 (C=O); 1647 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 0.84 t (3H, Me, J = 7.17 Hz), 1.01 m (10H, CH_2), 1.20 s (6H, 7-Me), 1.42 m (2H, CH_2), 2.23 d (2H, 6-H, ^2J = 16.04 Hz), 2.39 d (2H, 8-H, ^2J = 17.96 Hz), 3.17 t (1H, 4-H, J 3.15 Hz), 6.83 br.s (2H, NH_2). Found, %: C 71.96; H 9.05; N 8.76. $C_{19}H_{28}N_2O_2$. Calculated, %: C 72.12; H 8.92; N 8.85.

2-Amino-4-hexyl-7,7-dimethyl-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIl). Yield 2.87 g (95%), mp 172–173°C. IR spectrum, ν, cm^{-1}: 3387, 3295, 3211 (NH_2); 2298 (C≡N); 1707 (C=O); 1645 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 0.88 t (3H, Me, J = 7.61 Hz), 1.05 s and 1.09 s (3H each, 7-Me), 1.20–1.32 m (8H, CH_2), 1.38 m and 1.47 m (1H each, 4-CH_2), 2.16 d and 2.24 d (1H each, 6-H, ^2J = 15.62 Hz), 2.31 d and 2.43 d (1H each, 8-H, ^2J = 17.62 Hz), 3.16 t (1H, 4-H, J = 4.00 Hz), 6.61 br.s (2H, NH_2). Found, %: C 71.33; H 8.54; N 9.17. $C_{18}H_{26}N_2O_2$. Calculated, %: C 71.49; H 8.67; N 9.26.

2-Amino-7,7-dimethyl-4-(2-methylpropyl)-5-oxo-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIm). Yield 2.58 g (94%), mp 168–170°C. IR spectrum, ν, cm^{-1}: 3380, 3291, 3214 (NH_2); 2195 (C≡N); 1702 (C=O); 1648 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 0.67 d (3H, **Me**CH, J = 6.41 Hz), 0.95 t (3H, **Me**CH_2, J = 7.21 Hz), 1.05 s and 1.10 s (3H each, 7-Me), 1.28 m (2H, CH_2), 1.47 m (1H, CH), 2.14 d and 2.25 d (1H each, 6-H, ^2J = 15.71 Hz), 2.35 d and 2.45 d (1H each, 8-H, ^2J = 18.42 Hz), 3.22 d (1H, 4-H, J = 2.91 Hz), 6.67 br.s (2H, NH_2). Found, %: C 69.87; H 7.88; N 10.06. $C_{16}H_{22}N_2O_2$. Calculated, %: C 70.04; H 8.08; N 10.21.

2-Amino-7,7-dimethyl-5-oxo-4-(1-ethylpropyl)-5,6,7,8-tetrahydro-4*H*-chromene-3-carbonitrile (XIIn). Yield 2.54 g (88%), mp 167–168°C. IR spectrum, ν, cm^{-1}: 3397, 3304, 3219 (NH_2); 2200 (C≡N); 1712 (C=O); 1647 (δNH_2). ^{1}H NMR spectrum, δ, ppm: 0.84 t (3H, Me, J = 7.61 Hz), 1.01 t (3H, Me, J = 7.64 Hz), 1.04 s and 1.09 s (3H each, 7-Me), 1.11 m (3H, $CHCH_2$), 1.41 m (2H, CH_2), 2.15 d and 2.24 d (1H each, 6-H, ^2J = 16.02 Hz), 2.34 d and 2.45 d (1H each, 8-H, ^2J = 18.02 Hz), 3.34 d (1H, 4-H, J = 2.81 Hz), 6.63 br.s (2H, NH_2). Found, %: C 70.64; H 8.21; N 9.55. $C_{17}H_{24}N_2O_2$. Calculated, %: C 70.80; H 8.39; N 9.71.

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