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Synthesis of (4-Arylpyrrolidin-2-ylidene) Derivatives of Cyclic β-Dicarbonyl Compounds from Cinnamoyl Precursors

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Abstract—1,4-Conjugate Michael addition of nitromethane to the enone fragment of cinnamoyl derivatives of carbo- and heterocyclic β-dicarbonyl compounds provided the corresponding nitromethyl derivatives. The chemoselective reduction of the nitro group in the latter afforded 4-arylpyrrolidin-2-ylidene derivatives of β-dicarbonyl compounds in high yield. The reaction products with the heterocyclic β-dicarbonyl fragment are formed as equilibrium mixture of *E*– and *Z*-rotamers in which the *E*-rotamer predominates.

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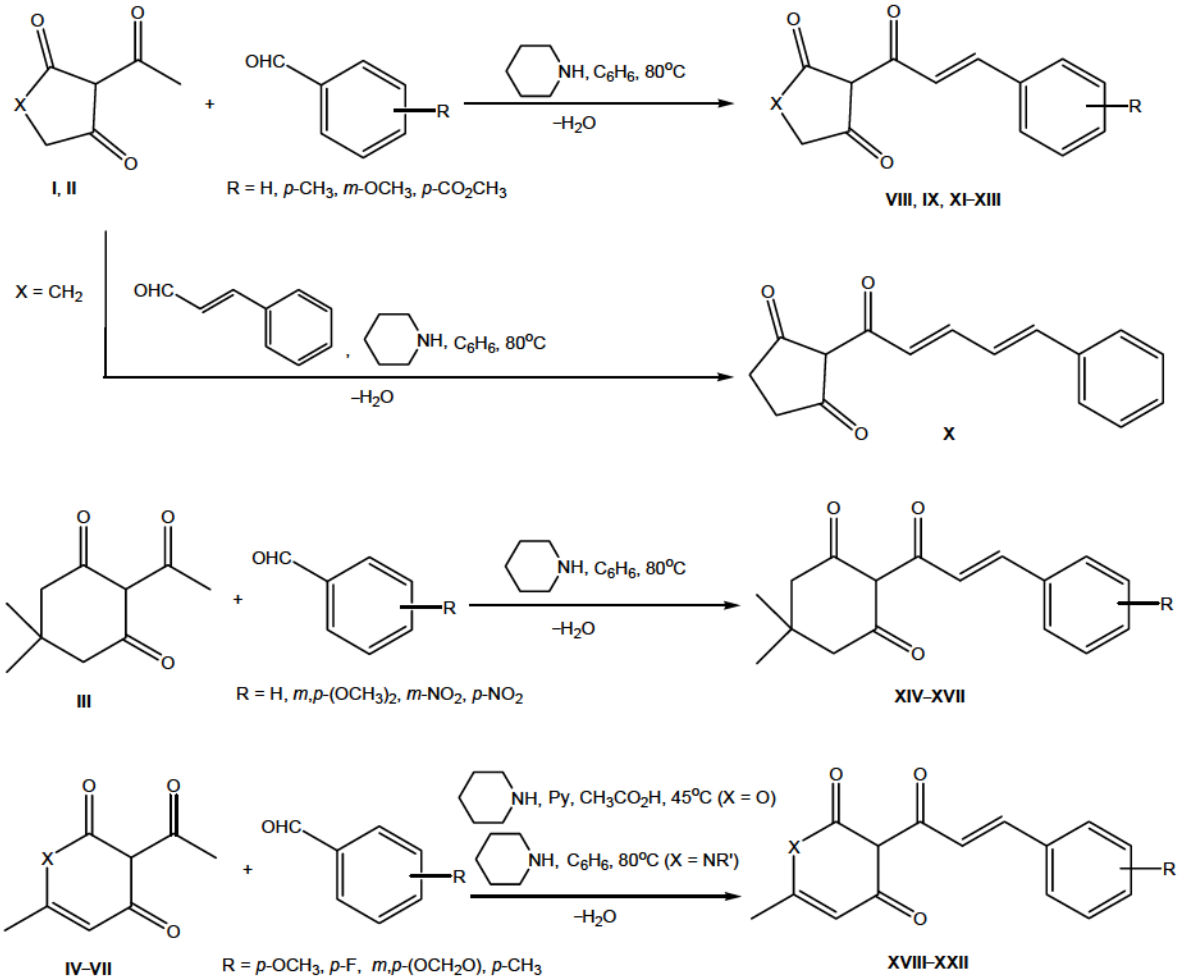
Cyclic β-di- and β-tricarbonyl compounds belong to the most efficient polyfunctional precursors in fine organic synthesis of complex naturally occurring and biologically active substances of versatile classes [1]. Among them cinnamoyl derivatives of carbo– and heterocyclic β-dicarbonyl compounds are of special interest since the fragment of α,β-unsaturated ketone in their side chain provides additional possibilities for selective chemical reactions at the enone system of these substances. Thus, methods for selective reduction of the enone moiety in cinnamoyl derivatives of cyclopentane-1,3-dione and tetronic acids developed by us [2] were applied in the synthesis of interphenylene prostanoids and their precursors [3]. The reaction of binucleophilic reagents with the enone system of cinnamoyl derivatives of 6-methyl-3*H*-pyran-2,4-dione [4], 4-hydroxycoumarin and 4-hydroxyquinol-2-one furnished a number of heterocyclic (2-pyrazoline [5–7], pyrazole [6, 8, 9], pyridinyl [10], pyrimidine [11], 1,4-diazepine [12], 1,5-benzodiazepine [13], 1,5-benzothiazepine, and 1,4-benzothiazine [14]) compound possessing various useful pharmacological and optical properties.

Conjugate addition of mononucleophilic nitroalkanes to Michael acceptors is widely used for the formation of new carbon–carbon bonds with the simultaneously introduction of a nitro group [15]. The ability of the nitro derivatives thus formed to convert into organic compounds of different classes promoted their application as key intermediates for dyes, medicines, and agrochemicals [16]. Although the conjugate addition of nitroalkanes to the enone system of chalcones, substituted cinnamic aldehydes and cinnamic acid esters has been extensively studied, a literature survey revealed that nothing was khown about reactions of cinnamoyl derivatives of cyclic β-dicarbonyl compounds with nitroalkanes.

We report here the results of our investigation that demonstrate for the first time the possibility of 1,4-conjugate Michael addition of nitromethane to the enone moiety of cinnamoyl derivatives of five- and six-membered carbo- and heterocyclic β-dicarbonyl compounds. The application of the Michael adducts thus formed in the synthesis of (4-arylpyrrolidin-2-ylidene) derivatives of cyclic β-dicarbonyl compounds as also demostrated.

Cinnamoyl derivatives of five-membered β-dicarbonyl compounds [cyclopentane-1,3-dione (**VIII**–**X**), tetronic acid (**XI**–**XIII**)] and of six-membered analogs [dimedone (**XIV**–**XVII**), 6-methyl-3*H*-pyran-2,4-dione (**XVIII** and **XIX**), 6-methyl-1-alkyl-(aryl)-1,3-tetrahydropyridine-2,4-dione (**XX**–**XXII**)] were obtained in high yields by Claisen–Schmidt conden-

Scheme 1.



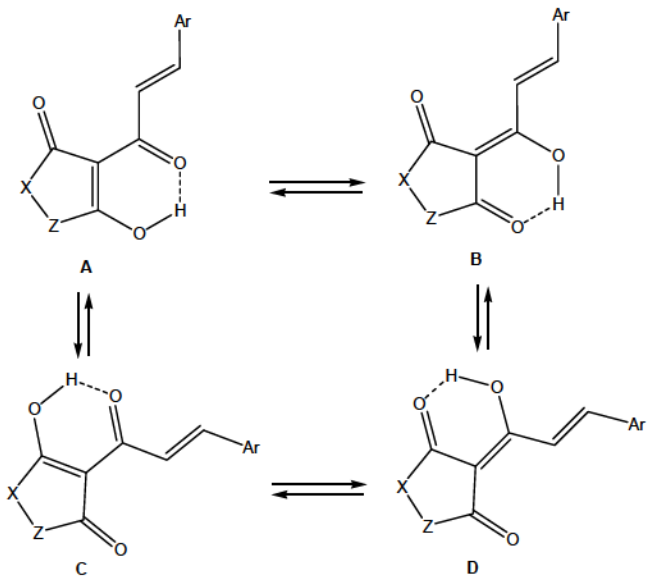
X = CH_2 (**I**), O (**II**, **IV**), N–Bn (**V**), N–CH_3 (**VI**), N–$C_6H_4OCH_3$-*p* (**VII**); X = CH_2, R = *p*-CH_3 (**VIII**), *p*-CO_2CH_3 (**IX**); X = O, R = H (**XI**), *m*-OCH_3 (**XII**), *p*-CO_2CH_3 (**XIII**); R = H (**XIV**), *m*,*p*-$(OCH_3)_2$ (**XV**), *m*-NO_2 (**XVI**), *p*-NO_2 (**XVII**); X = O, R = *p*-OCH_3 (**XVIII**), *p*-F (**XIX**); X = N–Bn, R = *p*-OCH_3 (**XX**); X = N–CH_3, R = *m*,*p*-(OCH_2O) (**XXI**); X = N–$C_6H_4OCH_3$-*p*, R = *p*-CH_3 (**XXII**).

sation of the readily available 2(3)-acetyl precursors **I**–**VII** with aromatic aldehydes [3, 17, 18] (Scheme 1).

In the study of Michael reaction involving the cyclic β-tricarbonyl compounds it should be taken into consideration that in contrast to chalcones and cinnamic acid esters the latter are vinylogous acids that may exist as an equilibrium mixture of endo- (**A** and **C**) and exocyclic (**B** and **D**) enol forms. The structure of the predominant tautomer is dependent to a great extent on the character of the cyclic β-dicarbonyl fragment (the ring size, the presence or absence of a heteroatom), on the aggregate state of these substances, on the nature of the solvent used, etc. [19]. The conversion of *endo*-cyclic tautomers (**A** and **C**) into the *exo*-cyclic ones (**B** and **D**) transfors the enone system of the side chain into the diene moiety that makes the Michael reaction at this fragment problematic (Scheme 2).

Reaction of cinnamoyl derivatives of five-membered (**VIII**–**XIII**) and six-membered (**XIV**–**XXII**) β-dicarbonyl compounds with nitromethane was carried out at room temperature. In the reaction nitromethane served both as reagent and solvent. Since, as already mentioned, the cinnamoyl derivatives **VIII**–**XXII** are vinylogous acids, more than 1 equiv. basic catalyst is necessary. 1,1,3,3-Tetramethylguanidine (TMG) (1.5 equiv.) was applied as the base because its vinylogous salts were readily soluble in nitromethane, thus the reaction took place as a homogeneous process (Scheme 3).

Scheme 2.

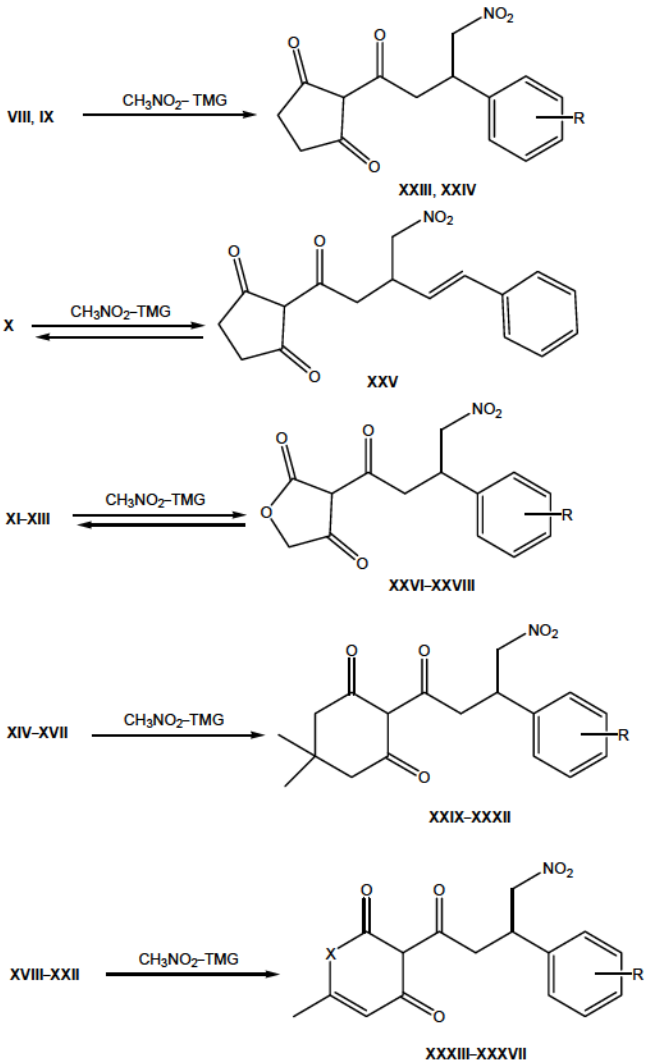


X = CH_2, O, S, N–R; Z = $(CH_2)_n$, CHAlk, $CH(Alk)_2$, $CH_2(CHAlk)CH_2$, $CH_2C(Alk)_2CH_2$, AlkC=CH, etc.

Under the described conditions nitromethane reacts with all cinnamoyl derivatives according to scheme of 1,4-conjugate addition to the enone fragment of cinnamoyl moiety to give nitromethyl derivatives **XXIII**–**XXXVII** in 42–70% yield. The compounds **XXIII**–**XXIV** and **XXIX**–**XXXVII** are stable while the derivatives of cyclopentane-1,3-dione **XXV** and tetronic acids **XXVI**–**XXVIII** decompose during chromatographic purification (retro-Michael reaction occurs).

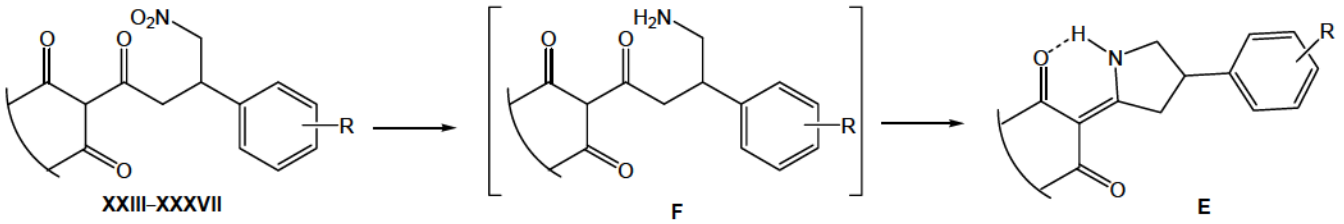
The NMR spectra of compounds **XXIII**–**XXXVII** lack signals of a double bond conjugated with a carbonyl group of the side chain [vinyl protons in the range δ 7.70–8.53 ppm (^{1}H) and sp^2-hybridized carbon atoms at δ 116.40–127.21 and 141.86–148.73 ppm (^{13}C)]. The signal of the intramolecularly chelated enol proton of the six-membered β-tricarbonyl compounds in the ^{1}H NMR spectra commonly appears as a narrow singlet (δ 15–19 ppm) and serves as a reference in the interpretation of the signals of enolized cyclic β-tricarbonyl system. Its presence in the spectra of compounds **XXIX**–**XXXVI** shows that the cyclic β-tricarbonyl fragment survived. This signal is shifted upfield by Δδ 0.9–1.9 ppm as compared with the analogous signal in the spectra of precursors **XIV**–**XXII** due to decreased conjugation of the exocyclic carbonyl group in the nitromethyl derivatives. In the ^{1}H NMR spectra of five-membered β-tricarbonyl compounds this signal was not detected except for compound **XXIV** (δ 11.77 ppm, broad peak). ^{1}H NMR spectra of nitromethyl derivatives **XXIII**–**XXXVII** exhibited two doublets of doublets of the protons of the methylene group adjacent to the carbonyl group of the acyl chain in the range 3.13–3.73 ppm (^2J 15.5–18.5 Hz), and two doublets of doublets are observed originating from the protons of nitromethyl substituent in the region 4.54–4.92 ppm (^2J 12.0–13.0 Hz). The signals due to the protons of the CH group in the branched aliphatic chain resonate as a quintet in the

Scheme 3.



R = *p*-CH_3 (**VIII**, **XXIII**), *p*-CO_2CH_3 (**IX**, **XIII**, **XXIV**, **XXVIII**), H (**XI**, **XIV**, **XXVI**, **XXIX**), *m*-OCH_3 (**XII**, **XXVII**), *m*, *p*-$(OCH_3)_2$ (**XV**, **XXX**), *m*-NO_2 (**XVI**, **XXXI**), *p*-NO_2 (**XVII**, **XXXII**); X = O, R = *p*-OCH_3 (**XVIII**, **XXXIII**), *p*-F (**XIX**, **XXXIV**); X = N–Bn, R = *p*-OCH_3 (**XX**, **XXXV**); X = N–CH_3, R = *m*, *p*-(OCH_2O) (**XXI**, **XXXVI**); X = N–$C_6H_4OCH_3$-*p*, R = *p*-CH_3 (**XXII**, **XXXVII**).

Scheme 4.



region 3.91–4.27 ppm (^3J 7.0–7.5 Hz) or as a multiplet. ^{13}C NMR spectra of these compounds exhibit signals of the carbon atoms of CH_2 groups in the region 41.68–45.92 ppm [C(O)CH2] and 78.94–80.23 ppm (CH_2NO_2) as well as the signal of the aliphatic CH group at 38.31–39.90 ppm. Relatively wide range of the chemical shifts of the secondary carbon atoms at the carbonyl group of the acyl chain in the spectra of compounds **XXIII**–**XXXVII** originates from a notable influence of the nature of the cyclic β-tricarbonyl fragment. The downfield shift of this signal occurs on going from the five-membered β-tricarbonyl compounds **XXIII**–**XXVIII** to six-membered **XXIX**–**XXXVII** and from six-membered carbocyclic **XXIX**–**XXXII** to six-membered heterocyclic β-tricarbonyl compounds **XXXIII**–**XXXVII**. IR spectra of compounds **XXIII**–**XXXVII** show in appropriate region strong absorption bands of carbonyl groups of the cyclic β-tricarbonyl system and also the characteristic bands of the stretching vibrations of the nitro group in the region 1379–1387 and 1551–1556 cm^{-1}.

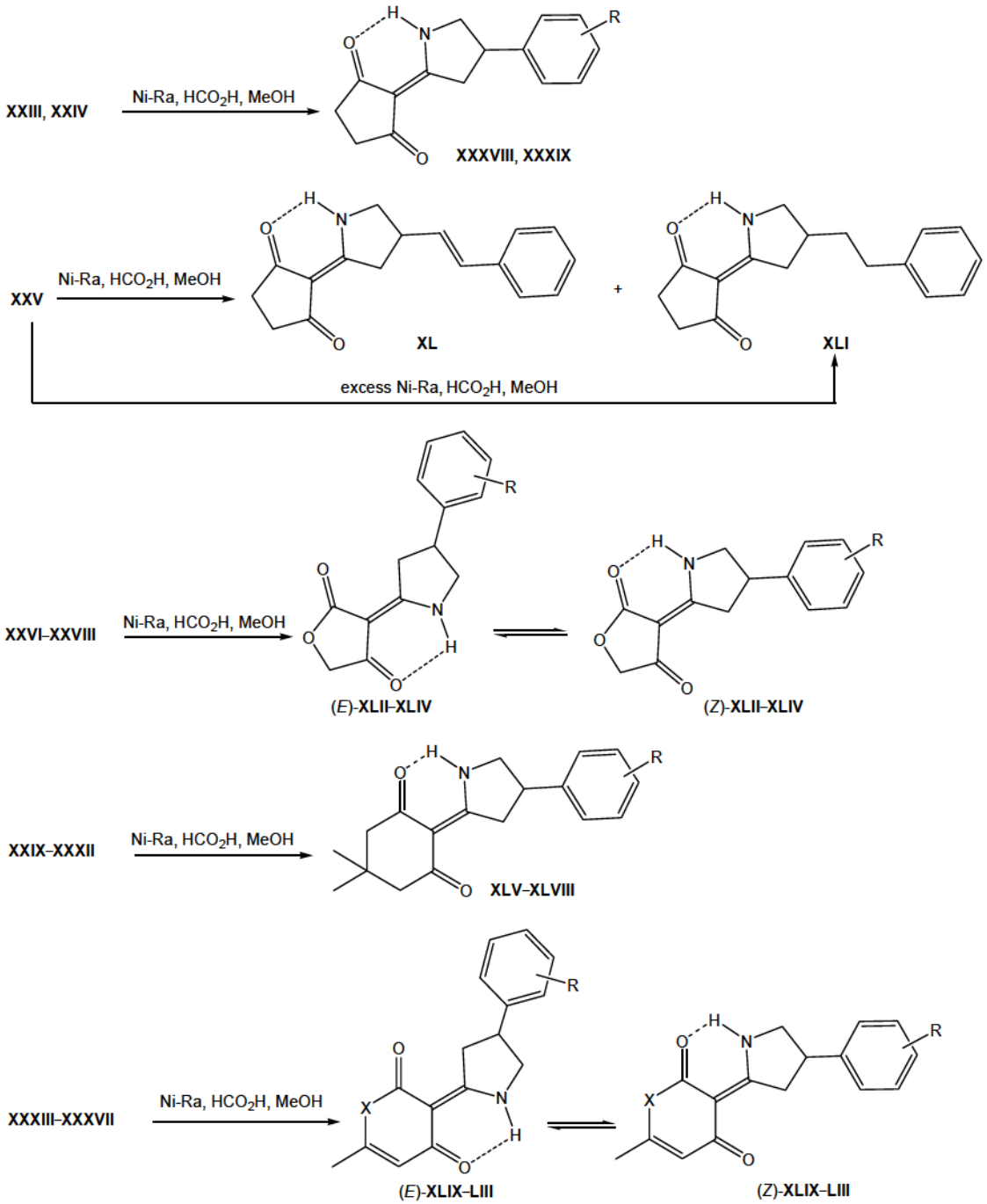
Thus, the described method is a general procedure for obtaining 2(3)-(4-nitro-3-arylbuta-noyl)-substituted carbo– and heterocyclic β-dicarbonyl compounds that are multifunctional precursors of new synthetically and biologically important substances. Compounds **XXIII**–**XXXVII** are convenient precursors of previously unknown (4-aryl-pyrrolidin-2-ylidene) derivatives of cyclic β-dicarbonyl compounds **E**. Synthesis of the latter can be achieved by selective reduction of the nitro group in the nitromethyl substituent of compounds **XXIII**–**XXXVII** into amino-function which is prone to intramolecular reaction with the carbonyl group of the acyl chain in the intermediates **F** (Scheme 4).

Earlier similar approach was employed for the formation of pyrrolidin-2-one fragment in synthetic schemes to obtain antidepressant (*S*)-rolipram [20], alkaloid (+)-9a-*epi*-stemoamide [21] as well as (*R*)- and (*S*)-stereoisomers of antispasmodic agent baclofen used in the treatment of multiple sclerosis, apoplexy, cranial traumas, and meningitis [22].

The selective reduction of the nitro group of polyfunctional compounds **XXIII**–**XXXVII** we carried out with the use of Raney nickel in the mixture of methanol and 90% formic acid at room temperature [23], since this system is found to be compatible with many functionalities including ketones. The selective reduction of the nitro group in compounds **XXIII**, **XXIV**, **XXIX**, **XXX**, and **XXXIII**–**XXXVII** by this system led to the formation of (4-arylpyrrolidin-2-ylidene) derivatives of β-dicarbonyl compounds **XXXVIII**, **XXXIX**, **XLV**, **XLVI**, and **XLIX**–**LIII** in 47–93% yield. Low yield (4–15%) of enaminodicarbonyl compounds **XLII**–**XLIV** under these conditions can be attributed to fast decomposition of unstable nitromethyl derivatives **XXVI**–**XXVIII** in acidic medium. Compounds **XXXI** and **XXXII** containing the nitro group in the aromatic ring furnished a complex mixture of reaction products due to the simultaneous reduction of both nitro groups and intermolecular reactions of the formed amines. The reduction of stable styryl derivative **XXV** also occurred nonselectively giving a difficultly separable mixture of enamino derivatives **XL** and **XLI**. However the application of excess Raney nickel led to the formation of phenethyl derivative **XLI** as a single product (Scheme 5).

Enaminodiones **XXXVIII**–**LIII** and related compounds are of great synthetic potential because they combine the ambidente nucleophilicity of enamines with ambident electrophilicity of enamines with the ambident electrophilicity of enones. For this reason these compounds can serve as versatile multipurpose precursors for the synthesis of new heterocycles, pharmaceuticals, and other useful substances. On the other hand, enaminocarbonyl moiety is known as the therapeutically valuable pharmacophores [25, 27]. Pyrrolidin- and piperidin-2-ylidene moieties are structural components that are present in numerous natural and synthetic substances exhibiting a broad spectrum of biologic activity [24–26]. Therefore, enaminodicarbonyl compounds **XXXVIII**–**LIII** are of

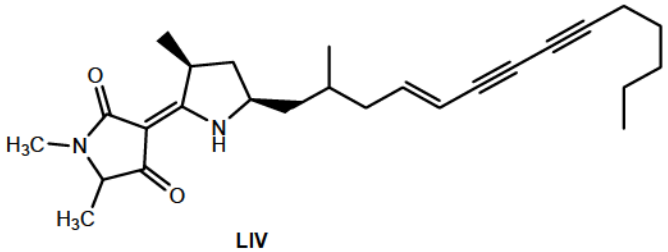
Scheme 5.



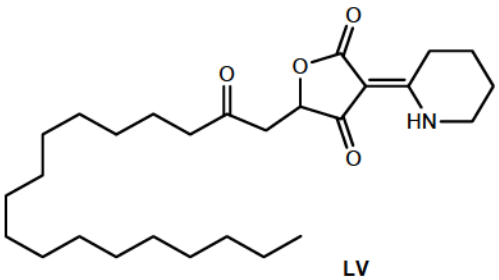
R = *p*-CH_3 (**XXIII**, **XXXVIII**), *p*-CO_2CH_3 (**XXIV**, **XXVIII**, **XXXIX**, **XLIV**), H (**XXVI**, **XXIX**, **XLII**, **XLV**), *m*-OCH_3 (**XXVII**, **XLIII**), *m*, *p*-$(OCH_3)_2$ (**XXX**, **XLVI**), *m*-NO_2 (**XXXI**, **XLVII**), *p*-NO_2 (**XXXII**, **XLVIII**); X = O, R = *p*-OCH_3 (**XXXIII**, **XLIX**), *p*-F (**XXXIV**, **L**); X = N–Bn, R = *p*-OCH_3 (**XXXV**, **LI**); X = N–CH_3, R = *m*, *p*-(OCH_2O) (**XXXVI**, **LII**); X = $NC_6H_4OCH_3$-*p*, R = *p*-CH_3 (**XXXVII**, **LIII**).

interest as new therapeutically useful molecules with potent hystaminergic, anticonvulsant, antiphlogistic and other activity. Among the naturally occurring substances structurally related to the enaminodicarbonyl compounds **XXXVIII**–**LIII** one can mention the allelochemical agent fischerellin **A** (**LIV**) exhibiting fungicidal and herbicidal activity [28] and alkaloid plakoridine C (**LV**) (Scheme 6) [29].

Scheme 6.



LIV



LV

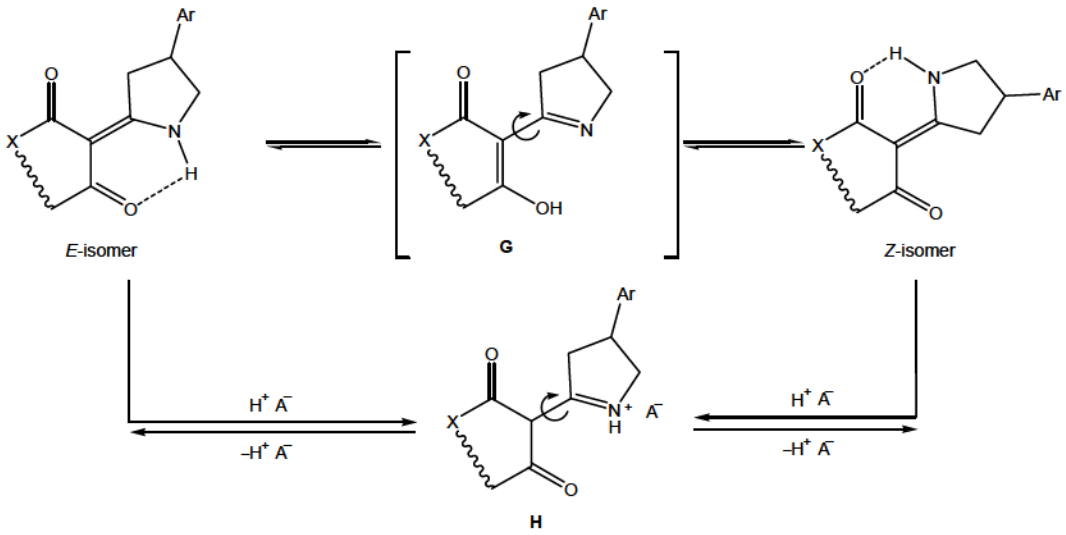
The structure of obtained enaminodicarbonyl compounds was proved by physicochemical methods. Their ^{1}H NMR spectra lack the proton signals of the nitromethyl substituent at δ 4.54–4.92 ppm. More upfield signals of magnetically nonequivalent protons of the methylene group at the nitrogen atom of the pyrrolidine ring are observed as two multiplets or doublets of doublets (^2J 10.5–12.5 Hz). One of them resonates at 4.03–4.25 ppm, and the other in most cases overlaps with the proton signal of the nodal CH group in the region 3.59–3.88 ppm. The chemical shifts of the multiplet (doublets of doublets, ^2J 18.0–20.0 Hz) signals of the second methylene group of the pyrrolidine ring are not essentially differ from those of the moiety $C(O)CH_2$ in the initial nitromethyl derivatives and are present in the region 3.59–4.09 and 3.15–3.56 ppm.

In the ^{13}C NMR spectra of these compounds the signal of the secondary carbon atom at the NH group of pyrrolidine ring is shifted upfield by ~23–24 ppm as compared to the carbon signal of the nitromethyl substituent of their synthetic precursors and is present at 54.63–56.84 ppm.

In the ^{1}H NMR ($CDCl_3$) spectra of derivatives of carbocyclic β-dicarbonyl compounds **XXXVIII**, **XXXIX**, **XLV**, and **XLVI** the signal of the proton of NH group involved in an intramolecular hydrogen bonding resonates as a broadened singlet in the region 10.46–11.88 ppm. In the spectra of derivatives of heterocyclic β-dicarbonyl compounds **XLIV**, **XLIX**–**LI**, and **LIII** two broadened singlets are observed in the region 9.89–13.20 ppm, the downfield one being more intensive. In the ^{1}H and ^{13}C NMR spectra of compounds **XLIV**, **XLIX**, **L**, and **LIII** most signals are doubled indicating that in solution the (4-arylpyrrolidin-2-ylidene) derivatives of the cyclic β-dicarbonyl compounds are present as an equilibrium mixture of enaminodicarbonyl rotamers interconverting via the formation of intermediate **G** [28]. The presence of heteroatom in the cyclic β-dicarbonyl fragment causes the formation of a mixture of *E*– and *Z*-isomers differing by the character of the intramolecular hydrogen bonding. Our results are consistent with the published data on tautomerism of the above mentioned fischerellin A and plakoridine C whose spectra exhibit a similar pattern (Scheme 7).

Taking into account the fact that in the ^{1}H NMR spectra of exocyclic enaminodicarbonyl compounds

Scheme 7.



the signals of the enamine proton shows the most pronounced downfield shift when this proton is chelated with the more electron-withdrawing carbonyl group of the ketone rather than that of ester or amide [30] we presume that the predominant rotamer of derivatives **XLII–XLIV** and **XLIX–LIII** has the *E*-configuration. Its content in the equilibrium mixture of isomers is ~55% for compound **XLIV** and 70–90% for compounds **XLIX–LIII**.

In the ^{1}H and ^{13}C NMR spectra of enaminodicarbonyl compounds **XLIV** and **XLIX–LIII** registered in a solution of deuteroacetic acid the doubling of signals is absent due to the formation of immonium salts **H** resulting from the *C*-protonation of the enaminodicarbonyl fragment of these substances in acidic medium [31].

IR spectra of the obtained enaminodiones show the absorption bands in the region 1612–1659 and 1564–1596 cm^{-1} attributable to the enaminodicarbonyl system.

EXPERIMENTAL

Melting points of the compounds obtained were determined on a Boëtius melting point apparatus. IR spectra were recorded on a Bomem Michelson 100 spectrometer; samples were examined as film (liquid) or KBr pellets (solid). ^{1}H and ^{13}C NMR spectra were registered on a Bruker Avance-500 spectrometer (500 and 125 MHz, respectively), using TMS as internal reference. The mass spectra were measured using an HPLC-MS/MS system consisting of an Accela chromatograph and LCQ-Fleet mass-selective detector with positive or negative ion detection (APCI and ESI). Progress of the reactions was monitored, and the purity of compounds obtained was checked by TLC on Silufol UV-254 or Alufol UV-254 plates (Merck). Kieselgel 60 HF254 TLC-standard (Merck) and Kieselgel 60 (Fluka) were used for column chromatography.

Cinnamoyl derivatives of cyclopentane-1,3-dione **VIII–X** and of tetronic acids **XI–XIII** were prepared according to the procedure [3]. Derivatives of dimedone **XIV–XVII** and 1-alkyl(aryl)-6-methyl-1,3-tetrahydropyridine-2,4-diones **XX–XXII** were synthesized by method [17], and of 6-methyl-3*H*-pyran-2,4-dione **XVIII** and **XIX**, by method [18].

Physico-chemical characteristics of cinnamoyl derivatives of cyclopentane-1,3-dione **VIII** and **IX** were reported in [32], of tetronic acids **XI–XIII**, in [3, 33], of dimedone **XIV** and **XV**, in [17], of 6-methyl-3*H*-pyran-2,4-dione **XVIII** and **XIX**, in [12, 34].

2-[(2*E*,4*E*)-5-Phenylpenta-2,4-dienoyl]cyclopentane-1,3-dione (X). Yield 61%, mp 148–150°C (from acetone). IR spectrum, ν, cm^{-1}: 1690, 1609 max, 1591, 1537, 1404. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.57 br (2H, CH_2), 2.69 br (2H, CH_2), 7.03–7.11 m (2H, 2CH), 7.33–7.40 m (3H_{Ar}, $H^{3,4,5}$), 7.43 d (1H, CH, $^3J_{trans}$ 15.5 Hz), 7.50 d.d (2H_{Ar}, $H^{2,6}$, ^3J 8.0, ^4J 1.0 Hz), 7.78 d.d.d (1H, CH, $^3J_1$ 15.5, $^3J_2$ 9.0, ^4J 1.5 Hz), 15.14 br (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 31.22 (CH_2), 34.39 (CH_2), 111.11, 122.19 (CH), 127.14 (CH_{Ar}), 127.69 (2CH_{Ar}), 128.89 (2CH_{Ar}), 129.85 (CH), 135.67, 144.25 (CH), 147.58 (CH), 180.61, 200.42, 209.28 (here and hereinafter the signals of substituted C_{sp2} atoms are not assigned). Mass spectrum (APCI): *m/z* 255 $[M]^+$. Found, %: C 75.61; H 5.50. $C_{16}H_{14}O_3$. Calculated, %: C 75.57; H 5.55.

(*E*)-5,5-Dimethyl-2-[3-(3-nitrophenyl)acryloyl]cyclohexane-1,3-dione (XVI). Yield 72%, mp 112–114°C. IR spectrum, ν, cm^{-1}: 1661, 1630, 1529 max, 1437 br, 1350. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.13 s (6H, 2CH_3), 2.45 s (2H, CH_2), 2.61 s (2H, CH_2), 7.60 t (1H, H^5_{Ar}, ^3J 8.0 Hz), 7.91 d [1H, C(O)CH=, $^3J_{trans}$ 16.0 Hz], 7.98 br.d (1H, H^6_{Ar}, ^3J 7.5 Hz), 8.24 d.d (1H, H^4_{Ar}, ^3J 8.0, ^4J 1.5 Hz), 8.36 d (1H, =CH, $^3J_{trans}$ 16.0 Hz), 8.44 br.s (1H, H^2_{Ar}), 18.40 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.21 (2CH_3), 30.52 (C^5), 48.30 (CH_2), 52.96 (CH_2), 111.34, 123.50 (CH), 124.72 (CH), 126.04 (CH), 129.79 (CH), 133.62 (CH), 136.77, 142.11 (CH), 148.73, 187.26, 195.56, 201.35, Mass spectrum (APCI): *m/z* 314 $[M - H]^-$; 316 $[MH]^+$. Found, %: C 64.69; H 5.44; N 4.40. $C_{17}H_{17}NO_5$. Calculated, %: C 64.75; H 5.43; N 4.44.

(*E*)-5,5-Dimethyl-2-[3-(4-nitrophenyl)acryloyl]cyclohexane-1,3-dione (XVII). Yield 68%, mp 156–159°C. IR spectrum, ν, cm^{-1}: 1661, 1630, 1599, 1520 max, 1433 br, 1344 max. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.13 s (6H, 2CH_3), 2.45 s (2H, CH_2), 2.61 s (2H, CH_2), 7.79 d (2H_{Ar}, ^3J 8.5 Hz), 7.89 d [1H, C(O)CH=, $^3J_{trans}$ 16.0 Hz], 8.26 d (2H_{Ar}, ^3J 8.5 Hz), 8.38 d (1H, =CH, $^3J_{trans}$ 16.0 Hz), 18.39 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.21 (2CH_3), 30.53 (C^5), 48.29 (CH_2), 52.96 (CH_2), 111.43, 124.09 (2CH_{Ar}), 127.21 (CH), 129.22 (2CH_{Ar}), 140.99, 141.86 (CH), 148.60, 187.14, 195.62, 201.45. Mass spectrum (APCI): *m/z* (I_{rel}, %) 314 (40.5) $[M - H]^-$, 629 (100) $[2M - H]^-$; 316 (24) $[MH]^+$, 631 (100) $[2M + H]^+$. Found, %: C

64.73; H 5.45; N 4.43. $C_{17}H_{17}NO_5$. Calculated, %: C 64.75; H 5.43; N 4.44.

(*E*)-1-Benzyl-3-[3-(4-methoxyphenyl)acryloyl]-6-methylpyridine-2,4(1*H*,3*H*)-dione (XX). Yield 70%, mp 155–156°C. IR spectrum, ν, cm^{-1}: 1649, 1624, 1603, 1524, 1510 max, 1423, 410. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 2.28 s (3H, CH_3), 3.83 s (3H, OCH_3), 5.31 br.s (2H, NCH_2Ph), 5.90 s (1H, CH heterocycle), 6.89 d (2H_{Ar}, ^3J 8.5 Hz), 7.15 br.d (2H_{Ar}, ^3J 8.0 Hz), 7.25–7.28 m (1H_{Ar}), 7.32–7.35 m (2H_{Ar}), 7.64 d (2H_{Ar}, ^3J 8.5 Hz), 7.91 d [1H, C(O)CH=, $^3J_{trans}$ 16.0 Hz], 8.51 d (1H, =CH, $^3J_{trans}$ 16.0 Hz), 16.76 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 21.39 (CH_3), 46.77 (CH_2), 55.38 (OCH_3), 102.11 (CH), 105.32, 114.24 (2CH_{Ar}), 123.09 (CH), 125.97 (2CH_{Ar}), 127.45 (CH_{Ar}), 128.13, 128.97 (2CH_{Ar}), 130.82 (2CH_{Ar}), 136.28, 144.52 (CH), 153.66, 161.68, 163.27, 176.92, 194.18. Mass spectrum (APCI): *m/z* 374 $[M - H]^-$; 376 $[MH]^+$. Found, %: C 73.54; H 5.61; N 3.75. $C_{23}H_{21}NO_4$. Calculated, %: C 73.58; H 5.64; N 3.73.

(*E*)-3-[3-(Benzo[*d*][1,3]dioxol-5-yl)acryloyl]-1,6-dimethylpyridine-2,4(1*H*,3*H*)-dione (XXI). Yield 82%, mp 233–237°C. IR spectrum, ν, cm^{-1}: 1620–1657 br, 1601 max, 1535, 1529, 1502, 1487, 1448, 1393, 1252 max. ^1H NMR spectrum (DMSO-d_6 + $CDCl_3$), δ, ppm: 2.40 s (3H, CH_3), 3.42 s (3H, NCH_3), 5.95 s (1H, CH heterocycle), 6.08 s (2H, OCH_2O), 6.92 d (1H_{Ar}, ^3J 8.0 Hz), 7.19–7.20 m (2H_{Ar}), 7.73 d [1H, C(O)C<u>H</u>=, $^3J_{trans}$ 15.5 Hz], 8.38 d (1H, =CH, $^3J_{trans}$ 15.5 Hz), 16.29 s (1H, OH enol). ^1H NMR spectrum (CD_3CO_2D), δ, ppm: 2.43 s (3H, CH_3), 3.53 s (3H, NCH_3), 6.04 s (2H, OCH_2O), 6.07 s (1H, CH in heterocycle), 6.88 d (1H_{Ar}, ^3J 8.0 Hz), 7.22 br.d (1H_{Ar}, ^3J 8.0 Hz), 7.30 br.s (1H_{Ar}), 7.83 d [1H, C(O)C<u>H</u>=, $^3J_{trans}$ 16.0 Hz], 8.36 d (1H, =CH, $^3J_{trans}$ 16.0 Hz). ^{13}C NMR spectrum (CD_3CO_2D), δ, ppm: 21.78 (CH_3), 32.07 (CH_3), 102.87 (CH_2), 102.97, 107.92 (CH_{Ar}), 109.45 (CH_{Ar}), 124.53 (CH), 126.86 (CH_{Ar}), 131.01, 145.42 (CH), 151.22, 155.91, 164.33, 172.20, 176.66, 178.21 (CH), 199.25. Mass spectrum (APCI): *m/z,* 314 $[MH]^+$. Found, %: C 65.12; H 4.82; N 4.50. $C_{17}H_{15}NO_5$. Calculated, %: C 65.17; H 4.83; N 4.47.

(*E*)-6-Methyl-1-(4-methoxyphenyl)-3-[3-(4-methyl-phenyl)acryloyl]pyridine-2,4(1*H*,3*H*)-dione (XXII). Yield 83%, mp 185–189°C. IR spectrum, ν, cm^{-1}: 1659 (max), 1626 max, 1612 sh, 1531, 1512 max, 1468, 1398, 1248. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 1.98 s (3H, CH_3), 2.34 s (3H, CH_3), 3.86 s (3H, OCH_3), 5.96 s (1H, CH heterocycle), 7.04 d (2H_{Ar}, ^3J 9.0 Hz), 7.10 d (2H_{Ar}, ^3J 9.0 Hz), 7.12 d (2H_{Ar}, ^3J 9.0 Hz), 7.54 d (2H_{Ar}, ^3J 9.0 Hz), 7.88 d [1H, C(O)C<u>H</u>=, $^3J_{trans}$ 16.0 Hz], 8.53 d (1H, =CH, $^3J_{trans}$ 16.0 Hz), 16.92 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 21.52 (CH_3), 22.40 (CH_3), 55.53 (OCH_3), 101.58 (CH), 105.45, 115.20 (2CH_{Ar}), 124.37 (CH), 129.08 (2CH_{Ar}), 129.16 (2CH_{Ar}), 129.43 (2CH_{Ar}), 130.71, 132.56, 140.96, 144.77 (CH), 154.16, 159.77, 163.91, 177.85, 194.14. Mass spectrum (APCI): *m/z* 374 $[M - H]^-$; 376 $[MH]^+$. Found, %: C 73.56; H 5.61; N 3.75. $C_{23}H_{21}NO_4$. Calculated, %: C 73.58; H 5.64; N 3.73.

Conjugate addition of nitromethane to the enone system of cinnamoyl derivatives of β-dicarbonyl compounds. General procedure. To a solution or suspension of 1 mmol of cinnamoyl derivative of β-dicarbonyl compound **VIII–XXII** in 10 mL of anhydrous nitromethane 1,1,3,3-tetramethylguanidine (1.5 mmol, 0.19 mL) was added dropwise upon stirring. The homogeneous reaction mixture was stirred at room temperature for 24–72 h (TLC monitoring). Nitromethane was evaporated in a vacuum as an azeotropic mixture with benzene. The residue was acidified with cold 1 N HCl. Solid products were washed successively with cold 1 N HCl and water, then were dried in air and recrystallized. Oily products were dissolved in chloroform, the solution was dried over anhydrous sodium sulfate. After evaporation of the solvent in a vacuum the residue was purified by column chromatography on silica gel.

2-[3-(4-Methylphenyl)-4-butanoyl]cyclopentane-1,3-dione (XXIII). Yield 42%, oily substance. IR spectrum, ν, cm^{-1}: 1703, 1693, 1639, 1632, 1620, 1582 br, 1553 max, 1441, 1435 sh, 1379. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 2.30 s (3H, CH_3), 2.49 m (2H, CH_2 of ring), 2.73 m (2H, CH_2 of ring), 3.23 d.d [1H, C(O)CH^A of chain, ^2J 17.5, ^3J 6.5 Hz], 3.55 d.d [1H, C(O)CH^B of chain, ^2J 17.5, ^3J 8.0 Hz], 4.11 quintet (1H, CH, ^3J 7.5 Hz), 4.60 d.d (1H, CH^ANO_2, ^2J 12.5, ^3J 8.5 Hz), 4.67 d.d (1H, CH^BNO_2, ^2J 12.5, ^3J 7.5 Hz), 7.13 *AB*-quartet (4H_{Ar}). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 21.07 (CH_3), 27.83 (CH_2), 33.44 (CH_2), 38.50 (CH), 42.01 (CH_2), 79.82 (CH_2), 114.67, 127.36 (2CH_{Ar}), 129.68 (2CH_{Ar}), 135.44, 137.62, 198.66, 199.66, 203.08. Mass spectrum (APCI): *m/z* 304 $[MH]^+$. Found, %: C 63.30; H 5.59; N 4.65. $C_{16}H_{17}NO_5$. Calculated, %: C 63.36; H 5.65; N 4.62.

Methyl 4-[4-(2,5-dioxocyclopentyl)-1-nitro-4-oxobutan-2-yl]benzoate (XXIV). Yield 91%, mp 120–122°C. IR spectrum, ν, cm^{-1}: 1711 max, 1703,

1637, 1612, 1574, 1551, 1433, 1381 w, 1290. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.49 br. signal (2H, CH_2 of ring), 2.73 br. signal (2H, CH_2 of ring), 3.27 d.d [1H, C(O)CH^A of chain, ^2J 17.5, ^3J 6.5 Hz], 3.56 d.d [1H, C(O)CH^B of chain, ^2J 17.5, ^3J 8.0 Hz], 3.89 s (3H, CO_2CH_3), 4.21 quintet (1H, CH, ^3J 7.0 Hz), 4.67 d.d (1H, CH^ANO_2, ^2J 12.5, ^3J 8.5 Hz), 4.75 d.d (1H, CH^BNO_2, ^2J 12.5, ^3J 6.5 Hz), 7.37 d ($2H_{Ar}$, ^3J 8.0 Hz), 7.98 d ($2H_{Ar}$, ^3J 8.0 Hz), 11.77 br (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 27.58 (CH_2), 33.23 (CH_2), 38.50 (CH), 41.68 (CH_2), 52.00 ($CO_2\underline{C}H_3$), 78.94 (CH_2), 114.47, 127.55 ($2CH_{Ar}$), 129.57, 130.04 ($2CH_{Ar}$), 143.66, 166.37, 198.02, 199.62, 202.90. Mass spectrum (APCI): *m/z* 348 $[MH]^+$. Found, %: C 58.75; H 4.96; N 4.07. $C_{17}H_{17}NO_7$. Calculated, %: C 58.79; H 4.93; N 4.03.

(*E*)-2-[3-(Nitromethyl)-5-phenylpent-4-enoyl]-cyclopentane-1,3-dione (XXV). Unstable compound, decomposes during purification. ^{1}H NMR spectrum of unpurified sample ($CDCl_3$), δ, ppm: 2.50 br (2H, CH_2 of ring), 2.75 br (2H, CH_2 of ring), 3.10 d.d [1H, C(O)CH^A of chain, ^2J 17.0, ^3J 6.0 Hz], 3.31 d.d [1H, C(O)CH^B of chain, ^2J 17.0, ^3J 7.5 Hz], 3.67 sextet (1H, CH, ^3J 7.0 Hz), 4.52 d.d (1H, CH^ANO_2, ^2J 12.0, ^3J 8.5 Hz), 4.59 d.d (1H, CH^BNO_2, ^2J 12.0, ^3J 6.0 Hz), 6.07 d.d (1H, C$\underline{H}$=CHPh, ^2J 16.0, ^3J 8.5 Hz), 6.53 d (1H, CH=C$\underline{H}$Ph, ^2J 16.0 Hz), 7.22–7.32 m ($5H_{Ar}$).

Methyl 4-[4-(2,4-dioxotetrahydrofuran-3-yl)-1-nitro-4-oxobutan-2-yl]benzoate (XXVIII). Yield 47%, oily substance. IR spectrum, ν, cm^{-1}: 1715, 1607–1682 br, 1568, 1556, 1435–1485 br, 1379, 1285. ^{1}H NMR spectrum of unpurified sample ($CDCl_3$), δ, ppm: 3.13 d.d [1H, C(O)CH^A of chain, ^2J 15.5, ^3J 9.0 Hz], 3.29 d.d [1H, C(O)CH^B of chain, ^2J 15.5, ^3J 5.5 Hz], 3.87 s (3H, CO_2CH_3), 4.06–4.12 m (1H, CH), 4.20 br.s (2H, CH_2 heterocycle), 4.62 d.d (1H, CH^ANO_2, ^2J 13.0, ^3J 10.0 Hz), 4.79 d.d (1H, CH^BNO_2, ^2J 13.0, ^3J 5.5 Hz), 7.34 d ($2H_{Ar}$, ^3J 8.5 Hz), 7.90 d ($2H_{Ar}$, ^3J 8.5 Hz). ^{13}C NMR spectrum of unpurified sample ($CDCl_3$), δ, ppm: 39.90 (CH), 42.58 (CH_2), 52.14 ($CO_2\underline{C}H_3$), 70.47 (CH_2), 79.40 (CH_2), 97.00 (C^3), 127.78 ($2CH_{Ar}$), 129.10, 129.94 ($2CH_{Ar}$), 145.77, 166.86, 177.06, 192.10, 195.35.

5,5-Dimethyl-2-(4-nitro-3-phenylbutanoyl)cyclohexane-1,3-dione (XXIX). Yield 89%, mp 80–81°C. IR spectrum, ν, cm^{-1}: 1664, 1601, 1562 sh, 1555 max, 1431, 1381. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.04 s (3H, CH_3), 1.07 s (3H, CH_3), 2.34 s (2H, CH_2 of ring), 2.52 s (2H, CH_2 of ring), 3.41 d.d [1H, C(O)CH^A of chain, ^2J 17.0, ^3J 6.5 Hz], 3.65 d.d [1H, C(O)CH^B of chain, ^2J 17.0, ^3J 7.5 Hz], 4.14 quintet (1H, CH, ^3J 7.5 Hz), 4.63 d.d (1H, CH^ANO_2, ^2J 12.5, ^3J 8.5 Hz), 4.70 d.d (1H, CH^BNO_2, ^2J 12.5, ^3J 6.5 Hz), 7.26–7.33 m ($5H_{Ar}$), 17.59 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.09 (CH_3), 28.15 (CH_3), 30.73 (C^5), 39.48 (CH), 43.77 (CH_2), 46.20 (CH_2), 52.40 (CH_2), 79.82 (CH_2), 112.30, 127.58 ($2CH_{Ar}$), 127.79 (CH_{Ar}), 128.94 ($2CH_{Ar}$), 138.99, 195.26, 196.78, 201.99. Mass spectrum (APCI): *m/z* 332 $[MH]^+$. Found, %: C 65.23; H 6.40; N 4.27. $C_{18}H_{21}NO_5$. Calculated, %: C 65.24; H 6.39; N 4.23.

2-[3-(3,4-Dimethoxyphenyl)-4-nitrobutanoyl]-5,5-dimethylcyclohexane-1,3-dione (XXX). Yield 57%, mp 79–80°C. IR spectrum, ν, cm^{-1}: 1661, 1591, 1562 sh., 1553 max, 1520, 1464, 1441, 1379, 1263, 1240. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.03 s (3H, CH_3), 1.07 s (3H, CH_3), 2.34 s (2H, CH_2 of ring), 2.52 s (2H, CH_2 of ring), 3.35 d.d [1H, C(O)CH^A of chain, ^2J 17.0, ^3J 6.5 Hz], 3.69 d.d [1H, C(O)CH^B of chain, ^2J 17.0, ^3J 7.5 Hz], 3.84 s (3H, OCH_3), 3.87 s (3H, OCH_3), 4.09 quintet (1H, CH, ^3J 7.5 Hz), 4.60 d.d (1H, CH^ANO_2, ^2J 12.5, ^3J 8.5 Hz), 4.68 d.d (1H, CH^BNO_2, ^2J 12.5, ^3J 7.0 Hz), 6.77 s ($1H_{Ar}$), 6.80 s ($2H_{Ar}$), 17.67 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.08 ($2CH_3$), 30.70 (C^5), 39.32 (CH), 43.52 (CH_2), 46.21 (CH_2), 52.41 (CH_2), 55.82 (OCH_3), 55.90 (OCH_3), 80.04 (CH_2), 110.73 (CH_{Ar}), 111.30 (CH_{Ar}), 112.33, 119.56 (CH_{Ar}), 131.28, 148.44, 149.05, 195.32, 196.94, 202.03. Mass spectrum (APCI): *m/z* 392 $[MH]^+$. Found, %: C 61.32; H 6.46; N 3.60. $C_{20}H_{25}NO_7$. Calculated, %: C 61.37; H 6.44; N 3.58.

5,5-Dimethyl-2-[4-nitro-3-(3-nitrophenyl)butanoyl]cyclohexane-1,3-dione (XXXI). Yield 72%, mp 94–97°C. IR spectrum, ν, cm^{-1}: 1657, 1562 sh., 1553 max, 1529, 1435, 1379, 1352. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.06 s (3H, CH_3), 1.08 s (3H, CH_3), 2.36 s (2H, CH_2 of ring), 2.55 s (2H, CH_2 of ring), 3.46 d.d [1H, C(O)CH^A of chain, ^2J 18.0, ^3J 7.0 Hz], 3.65 d.d [1H, C(O)CH^B of chain, ^2J 18.0, ^3J 7.5 Hz], 4.27 m (1H, CH), 4.69 d.d (1H, CH^ANO_2, ^2J 13.0, ^3J 9.5 Hz), 4.78 d.d (1H, CH^BNO_2, ^2J 13.0, ^3J 6.0 Hz), 7.53 t (1H, H^5_{Ar}, ^3J 8.0 Hz), 7.65 br.d (1H, H^6_{Ar}, ^3J 8.0 Hz), 8.13–8.15 m (1H, H^4_{Ar}), 8.17 narrow t (1H, H^2_{Ar}, ^4J 1.5 Hz), 17.35 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.06 (CH_3), 28.16 (CH_3), 30.81 (C^5), 39.00 (CH), 43.81 (CH_2), 45.98 (CH_2), 52.30 (CH_2), 79.05 (CH_2), 112.20, 122.60 (CH_{Ar}), 122.93 (CH_{Ar}), 129.97 (CH_{Ar}), 134.11 (CH_{Ar}), 141.32, 148.53, 195.37, 196.68, 201.20. Mass spectrum (APCI): *m/z* 377 $[MH]^+$.

Found, %: C 57.38; H 5.33; N 7.49. $C_{18}H_{20}N_2O_7$. Calculated, %: C 57.44; H 5.36; N 7.44.

5,5-Dimethyl-2-[4-nitro-3-(4-nitrophenyl)butanoyl]cyclohexane-1,3-dione (XXXII). Yield 68%, mp 67–70°C. IR spectrum, ν, cm^{-1}: 1664, 1603, 1562, 1553 max, 1520, 1429, 1379, 1348. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 1.05 s (3H, CH_3), 1.08 s (3H, CH_3), 2.35 s (2H, CH_2 of ring), 2.54 s (2H, CH_2 of ring), 3.43 d.d [1H, C(O)CH^A of chain, ^2J 17.5, ^3J 6.5 Hz], 3.66 d.d [1H, C(O)CH^B of chain, ^2J 17.5, ^3J 7.5 Hz], 4.27 m (1H, CH), 4.67 d.d (1H, CH^ANO_2, ^2J 13.0, ^3J 9.0 Hz), 4.76 d.d (1H, CH^BNO_2, ^2J 13.0, ^3J 6.0 Hz), 7.48 d ($2H_{Ar}$, ^3J 8.5 Hz), 8.19 d ($2H_{Ar}$, ^3J 8.5 Hz), 17.34 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.07 (CH_3), 28.15 (CH_3), 30.81 (C^5), 39.12 (CH), 43.69 (CH_2), 46.00 (CH_2), 52.32 (CH_2), 78.97 (CH_2), 112.20, 124.16 ($2CH_{Ar}$), 128.75 ($2CH_{Ar}$), 146.55, 147.51, 195.41, 196.72, 201.17. Mass spectrum (APCI): *m/z* 377 $[MH]^+$. Found, %: C 57.40; H 5.36; N 7.47. $C_{18}H_{20}N_2O_7$. Calculated, %: C 57.44; H 5.36; N 7.44.

3-[3-(4-Methoxyphenyl)-4-nitrobutanoyl]-6-methyl-3*H*-pyran-2,4-dione (XXXIII). Yield 91%, mp 115–116°C. IR spectrum, ν, cm^{-1}: 1728, 1639, 1614, 1562, 1553 max, 1516, 1456, 1420, 1379 w., 1252. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 2.28 s (3H, CH_3), 3.41 d.d [1H, C(O)CH^A of chain, ^2J 18.0, ^3J 6.5 Hz], 3.66 d.d [1H, C(O)CH^B of chain, ^2J 18.0, ^3J 7.5 Hz], 3.78 s (3H, OCH_3), 4.13 quintet (1H, CH, ^3J 7.0 Hz), 4.59 d.d (1H, CH^ANO_2, ^2J 12.0, ^3J 8.0 Hz), 4.69 d.d (1H, CH^BNO_2, ^2J 12.0, ^3J 6.5 Hz), 5.94 s (1H, CH in heterocycle), 6.85 d ($2H_{Ar}$, ^3J 8.5 Hz), 7.20 d ($2H_{Ar}$, ^3J 8.5 Hz), 16.04 br (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 20.74 (CH_3), 38.32 (CH), 44.77 (CH_2), 55.25 (OCH_3), 80.01 (CH_2), 99.71 (C^3), 101.49 (CH), 114.34 ($2CH_{Ar}$), 128.65 ($2CH_{Ar}$), 130.81, 159.05, 161.08, 169.50, 180.92, 203.66. Mass spectrum (APCI): *m/z* 348 $[MH]^+$. Found, %: C 58.77; H 4.89; N 4.01. $C_{17}H_{17}NO_7$. Calculated, %: C 58.79; H 4.93; N 4.03.

6-Methyl-3-[4-nitro-3-(4-fluorophenyl)butanoyl]-3*H*-pyran-2,4-dione (XXXIV). Yield 62%, mp 125–126°C. IR spectrum, ν, cm^{-1}: 1724, 1639, 1603, 1562 max, 1556 sh., 1512, 1441, 1387, 1225. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 2.28 s (3H, CH_3), 3.42 d.d [1H, C(O)CH^A of chain, ^2J 18.0, ^3J 6.5 Hz], 3.65 d.d [1H, C(O)CH^B of chain, ^2J 18.0, ^3J 7.5 Hz], 4.18 quintet (1H, CH, ^3J 7.5 Hz), 4.61 d.d (1H, CH^ANO_2, ^2J 12.5, ^3J 8.5 Hz), 4.72 d.d (1H, CH^BNO_2, ^2J 12.5, ^3J 6.0 Hz), 5.95 s (1H, CH in heterocycle), 7.02 d.d ($2H_{Ar}$, $^3J_{H,H} = {}^3J_{H,F}$ = 8.5 Hz), 7.27 d.d ($2H_{Ar}$, $^3J_{H,H}$ 8.5, $^4J_{H,F}$ 5.5 Hz), 15.98 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 20.75 (CH_3), 38.31 (CH), 44.70 (CH_2), 79.71 (CH_2), 99.65 (C^3), 101.41 (CH), 115.92 d ($C^{3,5}{}_{Ar}$, $^2J_{CF}$ 21.3 Hz), 129.27 d ($C^{2,6}{}_{Ar}$, $^3J_{CF}$ 7.5 Hz), 134.65, 161.05, 162.19 d (C–F, $^1J_{CF}$ 245 Hz), 169.70, 180.92, 203.35. Mass spectrum (APCI): *m/z* 336 $[MH]^+$. Found, %: C 57.28; H 4.20; N 4.15. $C_{16}H_{14}FNO_6$. Calculated, %: C 57.32; H 4.21; N 4.18.

1-Benzyl-6-methyl-3-[3-(4-methoxyphenyl)-4-nitrobutanoyl]pyridine-2,4(1*H*,3*H*)-dione (XXXV). Yield 70%, mp 175–176°C. IR spectrum, ν, cm^{-1}: 1649, 1609 max, 1551, 1516, 1429, 1381 w, 1252. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 2.28 s (3H, CH_3), 3.57 d.d [1H, C(O)CH^A of chain, ^2J 18.0, ^3J 7.0 Hz], 3.73 d.d [1H, C(O)CH^B of chain, ^2J 18.0, ^3J 7.0 Hz], 3.77 s (3H, OCH_3), 4.16 m (1H, CH), 4.58 d.d (1H, CH^ANO_2, ^2J 12.0, ^3J 9.0 Hz), 4.73 d.d (1H, CH^BNO_2, ^2J 12.0, ^3J 6.0 Hz), 5.26 br.s (2H, NCH_2Ph), 5.86 s (1H, CH in heterocycle), 6.83 d ($2H_{Ar}$, ^3J 8.5 Hz), 7.10 br.d ($2H_{Ar}$, $H^{2,6}{}_{benzyl}$, ^3J 7.0 Hz), 7.21 d ($2H_{Ar}$, ^3J 8.5 Hz), 7.27 br.t ($1H_{Ar}$, $H^4{}_{benzyl}$, ^3J 7.0 Hz), 7.34 t ($2H_{Ar}$, $H^{3,5}{}_{benzyl}$, ^3J 7.0 Hz), 15.18 s (1H, OH enol). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 21.43 (CH_3), 38.53 (CH), 45.92 (CH_2), 46.80 (CH_2), 55.23 (OCH_3), 80.23 (CH_2), 101.48 (CH), 105.38 (C^3), 114.23 ($2CH_{Ar}$), 125.94 ($2CH_{Ar}$), 127.61 (CH_{Ar}), 128.72 ($2CH_{Ar}$), 129.03 ($2CH_{Ar}$), 131.48, 135.95, 154.49, 158.89, 162.89, 175.40, 204.21. Mass spectrum (APCI, ESI): *m/z* 437 $[MH]^+$. Found, %: C 65.99; H 5.52; N 6.46. $C_{24}H_{24}N_2O_6$. Calculated, %: C 66.04; H 5.54; N 6.42.

3-[3-(Benzo[*d*][1,3]dioxol-5-yl)-4-nitrobutanoyl]-1,6-dimethylpyridine-2,4-(1*H*,3*H*)-dione (XXXVI). Yield 82%, mp 184°C. IR spectrum, ν, cm^{-1}: 1649 br, 1607 max, 1566, 1551, 1504, 1489, 1435, 1381 w, 1248. ^1H NMR spectrum (DMSO-d_6 + $CDCl_3$), δ, ppm: 2.38 s (3H, CH_3), 3.38 s (3H, NCH_3), 3.41 d.d [1H, C(O)CH^A of chain, ^2J 17.5, ^3J 6.5 Hz], 3.55 d.d [1H, C(O)CH^B of chain, ^2J 17.5, ^3J 7.0 Hz], 3.91–3.97 m (1H, CH), 4.85 d.d (1H, CH^ANO_2, ^2J 13.0, ^3J 10.0 Hz), 4.92 d.d (1H, CH^BNO_2, ^2J 13.0, ^3J 6.0 Hz), 5.98 s (1H, CH in heterocycle), 5.99 d (2H, OCH_2O, ^2J 11.0 Hz), 6.76 d.d ($1H_{Ar}$, ^3J 8.0, ^4J 1.0 Hz), 6.80 d ($1H_{Ar}$, ^3J 8.0 Hz), 6.99 br.s ($1H_{Ar}$), 14.99 s (1H, OH enol). ^{13}C NMR spectrum (DMSO-d_6 + $CDCl_3$), δ, ppm: 21.10 (CH_3), 30.34 (CH_3), 38.74 (CH), 45.54 (CH_2), 79.53 (CH_2), 99.37 (CH), 100.82 (CH_2), 104.22 (C^3), 107.85 (CH_{Ar}), 107.99 (CH_{Ar}), 121.01 (CH_{Ar}), 133.85, 146.14, 147.20, 156.66, 161.82, 173.96, 203.86.

Mass spectrum (APCI): *m/z* 375 $[MH]^+$. Found, %: C 57.72; H 4.81; N 7.51. $C_{18}H_{18}N_2O_7$. Calculated, %: C 57.75; H 4.85; N 7.48.

6-Methyl-3-[3-(4-methylphenyl)-4-nitrobutanoyl]-1-(4-methoxyphenyl)pyridine-2,4(1*H*,3*H*)-dione (XXXVII). Yield 92%, mp 63–66°C. IR spectrum, ν, cm^{-1}: 1659, 1614 max, 1551, 1512, 1458, 1410, 1379, 1250. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 1.99 s (3H, CH_3), 2.28 s (3H, CH_3), 3.54 d.d [1H, C(O)CH^A of chain, ^2J 18.5, ^3J 7.0 Hz], 3.68 d.d [1H, C(O)CH^B of chain, ^2J 18.5, ^3J 6.5 Hz], 3.85 s (3H, OCH_3), 4.13–4.19 m (1H, CH), 4.54 d.d (1H, CH^ANO_2, ^2J 12.5, ^3J 9.0 Hz), 4.69 d.d (1H, CH^BNO_2, ^2J 12.5, ^3J 6.5 Hz), 5.92 s (1H, CH in heterocycle), 7.01–7.09 m ($6H_{Ar}$), 7.14 d ($2H_{Ar}$, ^3J 8.0 Hz). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 21.04 (CH_3), 22.38 (CH_3), 38.59 (CH), 45.73 (CH_2), 55.57 (OCH_3), 80.13 (CH_2), 100.95 (CH), 105.49 (C^3), 115.23 ($2CH_{Ar}$), 127.53 ($2CH_{Ar}$), 128.92 (CH_{Ar}), 129.04 (CH_{Ar}), 129.44 ($2CH_{Ar}$), 130.30, 136.47, 137.14, 154.71, 159.87, 163.58, 176.08, 203.93. Mass spectrum (APCI): *m/z* 437 $[MH]^+$. Found, %: C 66.04; H 5.51; N 6.46. $C_{24}H_{24}N_2O_6$. Calculated, %: C 66.04; H 5.54; N 6.42.

Reduction of nitromethyl derivatives XXIII–XXXVII [23]. A solution or a suspension of nitromethyl derivative (5 mmol) and Raney nickel of the grade T-1 (0.2–0.3 g) in methanol (3 mL) was stirred with 90% formic acid (2.5 mL) at room temperature. After completion of the reaction (TLC monitoring) Raney nickel was filtered off. The organic solution was evaporated in a vacuum. The residue was dissolved in chloroform, the resulting solution was washed with water and dried over anhydrous sodium sulfate. After evaporation of the solvent the residue was purified by column chromatography on silica gel.

2-[4-(4-Methylphenyl)pyrrolidin-2-ylidene]cyclopentane-1,3-dione (XXXVIII). Yield 47%, mp 211–212°C. IR spectrum, ν, cm^{-1}: 1682 w., 1620 max, 1582, 1574 sh, 1481, 1234. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 2.33 s (3H, CH_3), 2.50–2.52 m (2H, CH_2 in dicarbonyl fragment), 2.55–2.57 m (2H, CH_2 in dicarbonyl fragment), 3.32 d.d (1H, $C\underline{H}^AH^B$, ^2J 19.0, ^3J 7.0 Hz), 3.66–3.73 m (2H), 3.79 d.d (1H, $CH^A\underline{H}^B$, ^2J 19.0, ^3J 9.5 Hz), 4.06–4.13 m (1H), 7.09 d ($2H_{Ar}$, ^3J 8.0 Hz), 7.14 d ($2H_{Ar}$, ^3J 8.0 Hz), 10.46 br.s (1H, NH). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 20.91 (CH_3), 33.42 (CH_2), 34.31 (CH_2), 39.61 (CH_2), 39.82 (CH), 55.06 (CH_2), 104.97, 126.55 ($2CH_{Ar}$), 129.62 ($2CH_{Ar}$), 137.08, 137.95, 171.71, 202.20, 205.55. Mass spectrum (APCI): *m/z* 256 $[MH]^+$. Found, %: C 75.24; H 6.73; N 5.49. $C_{16}H_{17}NO_2$. Calculated, %: C 75.27; H 6.71; N 5.49.

Methyl 4-[5-(2,5-dioxocyclopentylidene)pyrrolidin-3-yl]benzoate (XXXIX). Yield 55%, mp 188–191°C. IR spectrum, ν, cm^{-1}: 1720, 1626 max, 1574, 1485, 1285. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 2.52–2.54 m (2H, CH_2 in dicarbonyl fragment), 2.57–2.59 m (2H, CH_2 in dicarbonyl fragment), 3.33–3.41 m (1H), 3.76–3.86 m (3H), 3.92 s (3H, CO_2CH_3), 4.14–4.22 m (1H), 7.28 d ($2H_{Ar}$, ^3J 8.5 Hz), 8.01 d ($2H_{Ar}$, ^3J 8.5 Hz), 10.48 br.s (1H, NH). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 33.47 (CH_2), 34.34 (CH_2), 39.53 (CH_2), 40.04 (CH), 52.22 ($CO_2\underline{C}H_3$), 54.84 (CH_2), 105.07, 126.85 ($2CH_{Ar}$), 129.43, 130.37 ($2CH_{Ar}$), 146.24, 166.57, 171.21, 202.50, 205.70. Mass spectrum (APCI): *m/z* 300 $[MH]^+$. Found, %: C 68.20; H 5.69; N 4.69. $C_{17}H_{17}NO_4$. Calculated, %: C 68.21; H 5.72; N 4.68.

(*E*)-2-(4-Styrylpyrrolidin-2-ylidene)cyclopentane-1,3-dione (XL). ^1H NMR spectrum ($CDCl_3$), δ, ppm [in a mixture with compound **XLI**]: 2.47–2.58 m (4H, $2CH_2$ in carbocycle), 3.14 d.d (1H, $C\underline{H}^AH^B$, ^2J 19.0, ^3J 7.0 Hz), 3.30–3.37 m (1H, CH in pyrrolidine), 3.57 d.d (1H, $NHC\underline{H}^AH^B$, ^2J 12.0, ^3J 7.5 Hz), 3.63 d.d (1H, $CH^A\underline{H}^B$, ^2J 19.0, ^3J 8.5 Hz), 3.93 d.d (1H, $NHCH^A\underline{H}^B$, ^2J 12.0, ^3J 8.5 Hz), 6.15 d.d (1H, $C\underline{H}$=CHPh, $^3J_{1\,trans}$ 16.0, $^3J_2$ 8.0 Hz), 6.52 d (1H, CH=$C\underline{H}$Ph, $^3J_{trans}$ 16.0 Hz), 7.25 t.t ($1H_{Ar}$, H^4, ^3J 7.0, ^4J 1.5 Hz), 7.28–7.35 m ($4H_{Ar}$), 10.40 br.s (1H, NH). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm (in a mixture with compound **XLI**): 33.35 (CH_2), 34.27 (CH_2), 38.18 (CH_2), 38.35 (CH), 53.22 (CH_2), 105.06, 126.18 ($2CH_{Ar}$), 127.80 (CH=), 128.26 (CH_{Ar}), 128.58 ($2CH_{Ar}$), 131.20 (CH=), 136.18, 171.58, 202.23, 205.46. Mass spectrum [in a mixture with compound **XLI**]: *m/z* 268 $[MH]^+$.

2-(4-phenethylpyrrolidin-2-ylidene)cyclopentane-1,3-dione (XLI). Yield 14%, mp 112–114°C. IR spectrum, ν, cm^{-1}: 1686, 1637 sh, 1612–1632 br. max, 1583 max, 1479, 1234. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 1.82 q (2H, $CHC\underline{H}_2CH_2Ph$, ^3J 7.5 Hz), 2.46–2.53 m (5H, $2CH_2$ carbocycle + CH pyrrolidine), 2.63 d.t (1H, $C\underline{H}^AH^BPh$, ^2J 14.0, ^3J 7.5 Hz), 2.72 d.t (1H, $CH^A\underline{H}^BPh$, ^2J 14.0, ^3J 7.5 Hz), 2.90 d.d (1H, $CH^A\underline{H}^B$, ^2J 19.5, ^3J 7.0 Hz), 3.34 d.d (1H, $NHC\underline{H}^AH^B$, ^2J 12.0, ^3J 7.0 Hz), 3.51 d.d (1H, $CH^A\underline{H}^B$, ^2J 19.5, ^3J 9.0 Hz), 3.79 d.d (1H, $NHCH^A\underline{H}^B$, ^2J 12.0, ^3J 8.5 Hz), 7.16 br.d ($2H_{Ar}$, $H^{2,6}$, ^3J 7.5 Hz), 7.20 t.t ($1H_{Ar}$, H^4, ^3J 7.5, ^4J 1.0 Hz), 7.29 t ($2H_{Ar}$, $H^{3,5}$, ^3J 7.5 Hz), 10.34 br.s (1H, NH). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 33.34 (CH_2),

33.86 (CH_2), 34.26 (CH), 34.27 (CH_2), 35.65 (CH_2), 37.88 (CH_2), 53.29 (CH_2), 105.01, 126.19 (CH_{Ar}), 128.24 ($2CH_{Ar}$), 128.53 ($2CH_{Ar}$), 140.80, 172.19, 202.35, 205.50. Mass spectrum (APCI): *m/z* 270 $[M\mathrm{H}]^+$.

(*E,Z*)-3-(4-Phenylpyrrolidin-2-ylidene)tetrahydrofuran-2,4-dione (XLII). Yield 4%. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 3.15 d.d (1H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, ^2J 18.0, ^3J 7.0 Hz), 3.59–3.66 m (1H), 3.62 d.d (1H, NHC$\underline{\mathrm{H}}^A\mathrm{H}^B$, ^2J 12.0, ^3J 7.0 Hz), 3.72 quintet (1H, CH, ^3J 7.5 Hz), 4.06 d.d (1H, NHCH$^A\underline{\mathrm{H}}^B$, ^2J 12.0, ^3J 8.5 Hz), 4.41 s (2H, CH_2 in dicarbonyl fragment), 7.24–7.29 m ($3H_{Ar}$), 7.34 t ($2H_{Ar}$, ^3J 7.5 Hz), 10.11 br (1H, NH). Mass spectrum (APCI): *m/z* 244 $[M\mathrm{H}]^+$.

(*E,Z*)-3-[4-(3-Methoxyphenyl)pyrrolidin-2-ylidene]tetrahydrofuran-2,4-dione (XLIII). Yield 8%. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 3.15 d.d (1H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, ^2J 18.0, ^3J 7.0 Hz), 3.59–3.64 m (2H), 3.69 quintet (1H, CH, ^3J 7.5 Hz), 3.74 s (3H, OCH_3), 4.03 d.d (1H, NHC$\underline{\mathrm{H}}^A\mathrm{H}^B$, ^2J 11.5, ^3J 8.0 Hz), 4.41 s (2H, CH_2 in dicarbonyl fragment), 6.81–6.85 m ($2H_{Ar}$, $H^{4,6}$), 6.87 narrow t ($1H_{Ar}$, H^2, ^4J 2.0 Hz), 7.25 t ($1H_{Ar}$, H^5, ^3J 7.5 Hz), 10.10 br. signal (1H, NH). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 39.23 (CH), 39.64 (CH_2), 54.96 (OCH_3), 55.27 (CH_2), 70.94 (CH_2), 87.60, 112.26 (CH_{Ar}), 112.71 (CH_{Ar}), 118.95 (CH_{Ar}), 129.72 (CH_{Ar}), 143.13, 159.41, 169.86 (2C), 193.89. Mass spectrum, *m/z*: (APCI) 272 $[M - \mathrm{H}]^-$, 275 $[M + 2]^+$; (ESI) 275 $[M + 2]^+$.

Methyl 4-[5-(2,4-dioxodihydrofuran-3(2*H*)-ylidene)pyrrolidin-3-yl]benzoate (XLIV). Mixture of *E*– and *Z*-isomers, 1.2 : 1. Yield 15%, mp 251–252°C. IR spectrum, ν, cm^{-1}: 1736 sh, 1722, 1670 max, 1585, 1493, 1285, 1259, 1244. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 3.15 d.d (0.55H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, *E*-isomer, ^2J 19.0, ^3J 8.0 Hz), 3.18 d.d (0.45H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, *Z*-isomer, ^2J 19.0, ^3J 7.5 Hz), 3.61–3.75 m (2H), 3.79–3.83 m (1H), 3.84 s (3H, CO_2CH_3), 4.06–4.12 m (1H), 4.39 s (0.9H, CH_2 in tetronate ring of *Z*-isomer), 4.43 s (1.1H, CH_2 in tetronate ring of *E*-isomer), 7.45 br.d ($2H_{Ar}$, ^3J 8.5 Hz), 7.93 d ($2H_{Ar}$, ^3J 8.5 Hz), 9.89 br.s (0.45H, NH, *Z*-isomer), 10.37 br.s (0.55H, NH, *E*-isomer). ^{1}H NMR spectrum (CD_3CO_2D), δ, ppm: 3.29–3.40 m (1H), 3.82–3.96 m (3H), 3.91 s (3H, CO_2CH_3), 4.25–4.34 m (1H), 4.57 s (2H, CH_2 in tetronate of ring), 7.43 d ($2H_{Ar}$, ^3J 8.0 Hz), 8.04 d ($2H_{Ar}$, ^3J 8.0 Hz). ^{13}C NMR spectrum (DMSO-d_6), δ, ppm: 38.93 (CH), 39.26 (CH_2), 39.60 (CH_2), 52.04 ($CO_2\underline{C}H_3$), 54.63 (CH_2), 55.26 (CH_2), 70.49 (CH_2), 71.61 (CH_2), 87.14, 88.09, 127.43 ($2CH_{Ar}$), 128.21, 129.53 ($2CH_{Ar}$), 147.13, 165.95, 169.62, 169.68, 172.03, 173.29, 193.04, 194.74. Mass spectrum (APCI): *m/z* 300 $[M - \mathrm{H}]^-$, 302 $[M\mathrm{H}]^+$.

5,5-Dimethyl-2-(4-phenylpyrrolidin-2-ylidene)-cyclohexane-1,3-dione (XLV). Yield 93%, mp 132–135°C. IR spectrum, ν, cm^{-1}: 1643, 1583 max, 1556, 1439. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.06 s (6H, $2CH_3$), 2.34 s (2H, CH_2), 2.42 s (2H, CH_2), 3.45 d.d (1H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, ^2J 19.5, ^3J 6.5 Hz), 3.63–3.73 m (2H, CH + NHC$\underline{\mathrm{H}}^A\mathrm{H}^B$), 3.93 d.d (1H, CH$^A\underline{\mathrm{H}}^B$, ^2J 19.5, ^3J 9.0 Hz), 4.09 d.d (1H, NHCH$^A\underline{\mathrm{H}}^B$, ^2J 11.5, ^3J 8.5 Hz), 7.21 br.d ($2H_{Ar}$, $H^{2,6}$, ^3J 7.0 Hz), 7.26 t ($1H_{Ar}$, H^4, ^3J 7.5 Hz), 7.33 t ($2H_{Ar}$, $H^{3,5}$, ^3J 7.5 Hz), 11.88 br.s (1H, NH). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.32 (CH_3), 28.51 (CH_3), 30.52 (C^5), 40.30 (CH), 43.30 (CH_2), 51.79 (CH_2), 52.54 (CH_2), 54.87 (CH_2), 105.63, 126.86 ($2CH_{Ar}$), 127.20 (CH_{Ar}), 128.93 ($2CH_{Ar}$), 141.89, 175.20, 196.37, 199.05. Mass spectrum (APCI): *m/z* 284 $[M\mathrm{H}]^+$. Found, %: C 76.26; H 7.49; N 4.93. $C_{18}H_{21}NO_2$. Calculated, %: C 76.29; H 7.47; N 4.94.

5,5-Dimethyl-2-[4-(3,4-dimethoxyphenyl)pyrrolidin-2-ylidene]cyclohexane-1,3-dione (XLVI). Yield 67%, mp 165–168°C. IR spectrum, ν, cm^{-1}: 1641, 1582 max, 1553, 1520, 1439, 1265, 1240. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.06 s (6H, $2CH_3$), 2.35 s (2H, CH_2), 2.42 s (2H, CH_2), 3.41 d.d (1H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, ^2J 19.5, ^3J 7.5 Hz), 3.59–3.72 m (2H, CH + NHC$\underline{\mathrm{H}}^A\mathrm{H}^B$), 3.87 s (6H, $2OCH_3$), 3.93 d.d (1H, CH$^A\underline{\mathrm{H}}^B$, ^2J 19.5, ^3J 9.0 Hz), 4.06 d.d (1H, NHCH$^A\underline{\mathrm{H}}^B$, ^2J 11.0, ^3J 9.0 Hz), 6.72 s ($1H_{Ar}$, H^2), 6.76 d ($1H_{Ar}$, ^3J 8.0 Hz), 6.82 d ($1H_{Ar}$, ^3J 8.0 Hz), 11.86 br.s (1H, NH). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 28.39 ($2CH_3$), 30.52 (C^5), 40.13 (CH), 43.30 (CH_2), 51.78 (CH_2), 52.52 (CH_2), 54.89 (CH_2), 55.93 ($2OCH_3$), 105.63, 110.06 (CH_{Ar}), 111.40 (CH_{Ar}), 118.85 (CH_{Ar}), 134.14, 148.15, 149.21, 175.23, 196.42, 199.05. Mass spectrum (APCI): *m/z* 344 $[M\mathrm{H}]^+$. Found, %: C 69.93; H 7.31; N 4.05. $C_{20}H_{25}NO_4$. Calculated, %: C 69.95; H 7.34; N 4.08.

6-Methyl-3-[4-(4-methoxyphenyl)pyrrolidin-2-ylidene]-3*H*-pyran-2,4-dione (XLIX). Mixture of *E*– and *Z*-isomers, 7.3 : 1. Yield 72%, mp 144–146°C. IR spectrum, ν, cm^{-1}: 1709, 1659, 1593 max, 1566 sh, 1516, 1462, 1248. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.15 s (3H, CH_3), 3.44 d.d (0.88H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, *E*-isomer ^2J 19.5, ^3J 7.0 Hz), 3.55 d.d (0.12H, C$\underline{\mathrm{H}}^A\mathrm{H}^B$, *Z*-isomer ^2J 19.5, ^3J 7.0 Hz), 3.66–3.78 m (2.12H, CH, *E*– and *Z*-isomers + NHC$\underline{\mathrm{H}}^A\mathrm{H}^B$, *Z*-isomer), 3.80 s (3H, OCH_3), 3.92 d.d (0.88H, CH$^A\underline{\mathrm{H}}^B$, *E*-isomer ^2J 19.5, ^3J 9.0 Hz), 4.06 d.d (0.12H, CH$^A\underline{\mathrm{H}}^B$, *Z*-isomer ^2J 19.5, ^3J 9.5 Hz), 4.16 d.d (0.88H, NHCH$^A\underline{\mathrm{H}}^B$, *E*-isomer ^2J 11.5, ^3J 8.5 Hz),

5.72 s + 5.74 s (1H, CH of pyrandione in *Z*– and *E*-isomers), 6.87 d (2H_{Ar}, ^3J 8.5 Hz), 7.14 d (2H_{Ar}, ^3J 8.5 Hz), 10.63 s (0.12H, NH, *Z*-isomer), 12.38 s (0.88H, NH, *E*-isomer). ^{1}H NMR spectrum (CD_3CO_2D), δ, ppm: 2.15 s (3H, CH_3), 3.42 br.d.d (1H, C$\underline{H}^A$H^B, ^2J 19.0, ^3J 6.0 Hz), 3.74 quintet (1H, CH, ^3J 8.0 Hz), 3.77 s (3H, OCH_3), 3.82 d.d (1H, NHC$\underline{H}^A$H^B, ^2J 12.0, ^3J 7.5 Hz), 3.91–4.04 br (1H, CH$^A\underline{H}^B$), 4.23 d.d (1H, NHCH$^A\underline{H}^B$, ^2J 12.0, ^3J 8.5 Hz), 5.95 s (1H, CH of pyrandione), 6.90 d (2H_{Ar}, ^3J 8.5 Hz), 7.21 d (2H_{Ar}, ^3J 8.5 Hz). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 19.78 (CH_3, *Z*-isomer), 20.09 (CH_3, *E*-isomer), 39.29 (CH, *E*-isomer), 40.03 (CH, *Z*-isomer), 43.17 (CH_2, *Z*-isomer), 43.56 (CH_2, *E*-isomer), 55.14 (CH_2, *Z*-isomer), 55.33 (OCH_3, *E*- + *Z*-isomers), 56.10 (CH_2, *E*-isomer), 94.50 (*E*- + *Z*-isomers), 107.39 (CH, *E*-isomer), 109.38 (CH, *Z*-isomer), 114.36 (2CH_{Ar}, *E*- + *Z*-isomers), 127.83 (2CH_{Ar}, *E*- + *Z*-isomers), 133.35 (*E*- + *Z*-isomers), 158.80 (*E*- + *Z*-isomers), 163.63 (*E*- + *Z*-isomers), 163.78 (*E*- + *Z*-isomers), 177.25 (*Z*-isomer), 177.82 (*E*-isomer), 184.22 (*E*-isomer), 185.22 (*Z*-isomer). ^{13}C NMR spectrum (CD_3CO_2D), δ, ppm: 19.87 (CH_3), 40.17 (CH), 44.97 (CH_2), 55.64 (OCH_3), 57.16 (CH_2), 95.03, 107.74 (CH), 115.32 (2CH_{Ar}), 128.96 (2CH_{Ar}), 134.56, 159.87, 165.98 (2C), 179.06, 185.22. Mass spectrum (APCI): *m/z* 300 [*M*H]$^+$. Found, %: C 68.22; H 5.74; N 4.67. $C_{17}H_{17}NO_4$. Calculated, %: C 68.21; H 5.72; N 4.68.

6-Methyl-3-[4-(4-fluorophenyl)pyrrolidin-2-ylidene]-3*H*-pyran-2,4-dione (L). Mixture of *E*– and *Z*-isomers, 4 : 1. Yield 52%, mp 129–133°C. IR spectrum, ν, cm^{-1}: 1709, 1659, 1591 max, 1564 sh, 1514, 1475, 1462, 1275, 1232, 1225. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.15 s + 2.17 s (3H, CH_3), 3.45 d.d (0.8H, C$\underline{H}^A$H^B, *E*-isomer, ^2J 19.5, ^3J 6.5 Hz), 3.56 d.d (0.2H, C$\underline{H}^A$H^B, *Z*-isomer, ^2J 19.0, ^3J 6.5 Hz), 3.70–3.80 m (2H, CH + NHC$\underline{H}^A$H^B of *E*– and *Z*-isomers), 3.93 d.d (1H, CH$^A\underline{H}^B$, *E*– and *Z*-isomers, ^2J 19.5, ^3J 9.0 Hz), 4.08 d.d (0.2H, CH$^A\underline{H}^B$, *Z*-isomer, ^2J 20.0, ^3J 8.5 Hz), 4.19 d.d (0.8H, NHCH$^A\underline{H}^B$, *E*-isomer, ^2J 11.5, ^3J 8.5 Hz), 5.72 s + 5.74 s (1H, CH of pyrandione in *E*– and *Z*-isomers), 7.03 d.d (2H_{Ar}, $^3J_{H-H} = {}^3J_{H-F}$ = 8.5 Hz), 7.19 d.d (2H_{Ar}, $^3J_{H-H}$ 8.5, $^4J_{H-F}$ 5.5 Hz), 10.63 br.s (0.2H, NH, *Z*-isomer), 12.39 br.s (0.8H, NH, *E*-isomer). ^{1}H NMR spectrum (CD_3CO_2D), δ, ppm: 2.15 s (3H, CH_3), 3.44 br.d.d (1H, C$\underline{H}^A$H^B, ^2J 20.0, ^3J 6.0 Hz), 3.78–3.87 m (2H, CH + NHC$\underline{H}^A$H^B), 3.95–4.08 br. signal (1H, CH$^A\underline{H}^B$), 4.28 d.d (1H, NHCH$^A\underline{H}^B$, ^2J 11.5, ^3J 8.0 Hz), 5.95 s (1H, CH of pyrandione), 7.08 d.d (2H_{Ar}, $^3J_{H,H} = {}^3J_{H,F}$ = 8.5 Hz), 7.32 d.d (2H_{Ar}, $^3J_{H,H}$ 8.5, $^4J_{H,F}$ 5.5 Hz). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 19.79 (CH_3, *Z*-isomer), 20.09 (CH_3, *E*-isomer), 39.29 (CH, *E*-isomer), 40.01 (CH, *Z*-isomer), 43.15 (CH_2, *Z*-isomer), 43.53 (CH_2, *E*-isomer), 55.04 (CH_2, *Z*-isomer), 56.02 (CH_2, *E*-isomer), 94.55 (*E*- + *Z*-isomers), 107.38 (CH, *E*-isomer), 109.35 (CH, *Z*-isomer), 115.92 d ($C^{3,5}{}_{Ar}$, $^2J_{CF}$ 21.3 Hz, *E*- + *Z*-isomers), 128.36 d ($C^{2,6}{}_{Ar}$, $^3J_{CF}$ 7.5 Hz, *E*- + *Z*-isomers), 137.18 (*E*- + *Z*-isomers), 161.97 d (C–F, $^1J_{CF}$ 245 Hz, *E*- + *Z*-isomers), 163.75 (2C, *E*- + *Z*-isomers), 176.94 (*Z*-isomer), 177.53 (*E*-isomer), 181.31 (*Z*-isomer), 184.22 (*E*-isomer). ^{13}C NMR spectrum (CD_3CO_2D), δ, ppm: 19.87 (CH_3), 40.20 (CH), 44.93 (CH_2), 57.06 (CH_2), 95.07, 107.82 (CH), 116.65 d ($C^{3,5}{}_{Ar}$, $^2J_{CF}$ 21.3 Hz), 129.74 d ($C^{2,6}{}_{Ar}$, $^3J_{CF}$ 7.5 Hz), 138.64, 163.07 d (C–F, $^1J_{CF}$ 242.5 Hz), 166.02 (2C), 178.88, 185.36. Mass spectrum (APCI): *m/z* 288 [*M*H]$^+$. Found, %: C 66.86; H 4.87; N 4.85. $C_{16}H_{14}FNO_3$. Calculated, %: C 66.89; H 4.91; N 4.88.

1-Benzyl-6-methyl-3-[4-(4-methoxyphenyl)pyrrolidin-2-ylidene]pyridine-2,4-(1*H*,3*H*)-dione (LI). Mixture of *E*– and *Z*-isomers, 2.3 : 1. Yield 75%, oily substance. IR spectrum, ν, cm^{-1}: 1612–1649 br. max, 1572 br, 1514, 1479, 1470, 1452, 1429, 1346, 1298, 1273, 1250. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.17 s (3H, CH_3), 3.52 d.d (1H, C$\underline{H}^A$H^B *E*– and *Z*-isomers, ^2J 19.5, ^3J 7.0 Hz), 3.64–3.70 m (1H, CH *E*– and *Z*-isomers), 3.75 d.d (1H, NHC$\underline{H}^A$H^B, ^2J 12.0, ^3J 7.0 Hz), 3.79 s (3H, OCH_3), 4.03 d.d (1H, CH$^A\underline{H}^B$, ^2J 19.5, ^3J 9.5 Hz), 4.14 d.d (1H, NHCH$^A\underline{H}^B$, ^2J 12.0, ^3J 9.0 Hz), 5.18 br.s (2H, NCH_2Ph), 5.79 br.s (1H, =CH of pyridinedione ring), 6.85 d (2H_{Ar}, ^3J 8.0 Hz), 7.12–7.15 narrow.m (4H_{Ar}), 7.23 t (1H_{Ar}, H$^4{}_{benzyl}$, ^3J 7.0 Hz), 7.30 t (2H_{Ar}, H$^{3,5}{}_{benzyl}$, ^3J 7.0 Hz), 11.89 br (0.3H, NH *Z*-isomer), 13.17 br (0.7H, NH *E*-isomer). ^{1}H NMR spectrum (CD_3CO_2D), δ, ppm: 2.25 s (3H, CH_3), 3.55 d.d (1H, C$\underline{H}^A$H^B, ^2J 19.5, ^3J 7.5 Hz), 3.74–3.80 m (1H, CH), 3.77 s (3H, OCH_3), 3.85 d.d (1H, NHC$\underline{H}^A$H^B, ^2J 12.5, ^3J 7.5 Hz), 4.12 d.d (1H, CH$^A\underline{H}^B$, ^2J 19.5, ^3J 8.5 Hz), 4.26 d.d (1H, NHCH$^A\underline{H}^B$, ^2J 12.5, ^3J 9.0 Hz), 5.26 br.s (2H, NCH_2Ph), 6.08 br.s (1H, =CH of pyridinedione ring), 6.90 d (2H_{Ar}, ^3J 8.5 Hz), 7.13 d (2H_{Ar}, ^3J 7.5 Hz), 7.22 d (2H_{Ar}, ^3J 8.5 Hz), 7.24 t (1H_{Ar}, ^3J 7.5 Hz), 7.32 t (2H_{Ar}, ^3J 7.5 Hz). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm: 20.92 (CH_3), 39.52 (CH), 43.84 (CH_2), 45.95 (CH_2), 55.31 (OCH_3), 55.67 (CH_2), 100.57, 108.34 (CH), 114.27 (2CH_{Ar}), 125.85 (2CH_{Ar}), 127.12 (CH_{Ar}), 127.87 (2CH_{Ar}), 128.81 (2CH_{Ar}), 133.74, 137.58, 150.20, 158.67, 165.00, 178.12, 182.46. ^{13}C NMR spectrum (CD_3CO_2D), δ, ppm: 21.07 (CH_3), 40.37 (CH), 45.60 (CH_2), 47.44 (CH_2), 55.64 (OCH_3),

56.93 (CH_2), 101.05, 108.78 (CH), 115.33 ($2CH_{Ar}$), 126.88 ($2CH_{Ar}$), 128.24 (CH_{Ar}), 129.02 ($2CH_{Ar}$), 129.79 ($2CH_{Ar}$), 134.80, 138.12, 153.32, 159.85, 166.66, 172.13, 181.71. Mass spectrum (APCI): *m/z* 389 $[MH]^+$. Found, %: C 74.17; H 6.26; N 7.25. $C_{24}H_{24}N_2O_3$. Calculated, %: C 74.21; H 6.23; N 7.21.

(*E,Z*)-3-[4-(Benzo[*d*][1,3]dioxol-5-yl)pyrrolidin-2-ylidene]-1,6-dimethylpyridine-2,4(1*H*,3*H*)-dione (LII). Yield 58%, mp 248–250°C. IR spectrum, ν, cm^{-1}: 1612–1639 br. max, 1564, 1504, 1477, 1433, 1273, 1258, 1244. ^1H NMR spectrum (CD_3CO_2D + $CDCl_3$), δ, ppm: 2.31 s (3H, CH_3), 3.39 s (3H, NCH_3), 3.52 d.d (1H, $C\underline{H}^AH^B$, ^2J 19.5, ^3J 7.5 Hz), 3.72 quintet (1H, CH, ^3J 8.0 Hz), 3.82 d.d (1H, $NHC\underline{H}^AH^B$, ^2J 12.5, ^3J 7.5 Hz), 4.08 d.d (1H, $CH^A\underline{H}^B$, ^2J 19.5, ^3J 9.0 Hz), 4.23 d.d (1H, $NHCH^A\underline{H}^B$, ^2J 12.5, ^3J 9.5 Hz), 5.92 s (2H, OCH_2O), 6.01 br.s (1H, =CH of pyridinedione ring), 6.76 s ($2H_{Ar}$), 6.80 s ($1H_{Ar}$), 11.52 br.s (1H, NH). ^{13}C NMR spectrum (CD_3CO_2D + $CDCl_3$), δ, ppm: 21.37 (CH_3), 31.15 (CH_3), 40.81 (CH), 45.45 (CH_2), 56.79 (CH_2), 101.05, 102.19 (CH_2), 108.17 (CH_{Ar}), 108.31 (CH), 109.33 (CH_{Ar}), 121.07 (CH_{Ar}), 136.67, 147.76, 149.20, 153.22, 166.66, 179.45, 181.60. Mass spectrum (APCI): *m/z* 327 $[MH]^+$. Found, %: C 66.20; H 5.55; N 8.60. $C_{18}H_{18}N_2O_4$. Calculated, %: C 66.25; H 5.56; N 8.58.

6-Methyl-1-(4-methoxyphenyl)-3-(4-*p*-tolylpyrrolidin-2-ylidene)pyridine-2,4(1*H*,3*H*)-dione (LIII). Mixture of *E*– and *Z*-isomers, 4.3 : 1. Yield 65%, mp 189–190°C. IR spectrum, ν, cm^{-1}: 1649, 1622 max, 1574, 1512, 1466, 1294, 1248. ^1H NMR spectrum ($CDCl_3$), δ, ppm: 1.85 s + 1.87 s (3H, CH_3, *Z*– and *E*-isomers), 2.31 s + 2.33 s (3H, CH_3, *E*– and *Z*-isomers), 3.49 d.d (0.81H, $C\underline{H}^AH^B$, *E*-isomer ^2J 19.5, ^3J 7.0 Hz), 3.61–3.71 m (1.38H, CH, *E*– and *Z*-isomers + $C\underline{H}^A\underline{H}^B$, *Z*-isomer), 3.75 d.d (0.81H, $NHC\underline{H}^AH^B$, *E*-isomer, ^2J 12.0, ^3J 7.0 Hz), 3.83 s + 3.85 s (3H, OCH_3, *E*– and *Z*-isomers), 3.96 d.d (0.81H, $CH^A\underline{H}^B$, *E*-isomer, ^2J 19.5, ^3J 9.5 Hz), 4.04 d.d (0.19H, $NHC\underline{H}^AH^B$, *Z*-isomer, ^2J 10.5, ^3J 8.5 Hz), 4.11–5.15 m (0.19H, $NHCH^A\underline{H}^B$, Z-isomer), 4.15 d.d (0.81H, $NHCH^A\underline{H}^B$, *E*-isomer, ^2J 12.0, ^3J 9.0 Hz), 5.77 s + 5.79 s (1H, CH of pyridinedione in *Z*– and *E*-isomers), 6.96–7.02 m ($2H_{Ar}$), 7.07–7.14 m ($6H_{Ar}$), 11.59 s (0.19H, NH, *Z*-isomer), 13.20 s (0.81H, NH, *E*-isomer). ^1H NMR spectrum (CD_3CO_2D), δ, ppm: 1.91 s (3H, CH_3), 2.29 s (3H, CH_3), 3.53 d.d (1H, $C\underline{H}^AH^B$, ^2J 19.5, ^3J 7.0 Hz), 3.75 quintet (1H, CH, ^3J 8.0 Hz), 3.82–3.88 m (1H, $C\underline{H}^AH^B$), 3.85 s (3H, OCH_3), 4.09 d.d (1H, $CH^A\underline{H}^B$, ^2J 19.5, ^3J 8.5 Hz), 4.25 d.d (1H, $NHCH^A\underline{H}^B$, ^2J 12.0, ^3J 9.0 Hz), 6.11 s (1H, CH of pyrandione), 7.03 d ($2H_{Ar}$, ^3J 8.5 Hz), 7.13–7.18 m ($6H_{Ar}$), 11.38 s (1H, NH). ^{13}C NMR spectrum ($CDCl_3$), δ, ppm, signals corresponding to (*E*)-isomer: 20.98 (CH_3), 21.98 (CH_3), 39.75 (CH), 43.24 (CH_2), 55.48 (CH_2), 55.51 (OCH_3), 100.57, 107.92 (CH), 114.76 ($2CH_{Ar}$), 126.63 ($2CH_{Ar}$), 129.51 ($2CH_{Ar}$), 129.79 ($2CH_{Ar}$), 131.45, 136.69, 138.83, 149.89, 159.30, 165.64, 178.00, 183.11; signals corresponding to (*Z*)-isomer: 20.98 (CH_3), 21.61 (CH_3), 40.31 (CH), 43.30 (CH_2), 54.80 (CH_2), 55.51 (OCH_3), 100.57, 110.66 (CH), 114.76 ($2CH_{Ar}$), 126.74 ($2CH_{Ar}$), 129.51 ($2CH_{Ar}$), 129.74 ($2CH_{Ar}$), 131.45, 136.79, 138.83, 148.31, 159.46, 168.10, 177.19, 181.15. ^{13}C NMR spectrum (CD_3CO_2D), δ, ppm: 21.08 (CH_3), 22.06 (CH_3), 40.66 (CH), 45.50 (CH_2), 55.96 (OCH_3), 56.84 (CH_2), 101.07, 108.68 (CH), 115.66 ($2CH_{Ar}$), 127.85 ($2CH_{Ar}$), 130.54 ($2CH_{Ar}$), 130.84 ($2CH_{Ar}$), 131.64, 137.78, 139.93, 153.03, 160.98, 167.33, 172.12, 179.58. Mass spectrum (APCI): *m/z* 389 $[MH]^+$. Found, %: C 74.18; H 6.21; N 7.24. $C_{24}H_{24}N_2O_3$. Calculated, %: C 74.21; H 6.23; N 7.21.

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