*Russian Journal of Organic Chemistry, Vol. 38, No. 4, 2002, pp. 463–474. Translated from Zhurnal Organicheskoi Khimii, Vol. 38, No. 4, 2002, pp. 487–498.*


# Condensation of Hydroxypyrimidines with Carbonyl Compounds: I. Barbituric Acids*

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Received July 13, 2001

**Abstract**—Condensation reactions of barbituric acids with aldehydes, ketones, and their analogs, also with participation of C- and N-nucleophiles as third components, resulting in versatile pyrimidine-containing heterocyclic systems are considered.

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## I. INTRODUCTION

The ability of hydroxypyrimidines (here the term "hydroxypyrimidine" is used also with respect to their possible tautomers) to react with carbonyl compounds was amply demonstrated up till now. The interest to these reactions has been generated mostly by the search of new biologically active substances since the presence in an organic molecule of a pyrimidine moiety that actually is a pharmacophore frequently provides the molecule with some kind of biological effect. Yet the basic structures that were subjected to the studies of such transformations in some detail are few in number. Nonetheless the progress achieved in this particular field of pyrimidine derivatives chemistry is very significant. Therefore it seems appropriate to summarize the available data especially because the attempts done in this direction concerned only narrow range of substrates and of chemical reactions thereof. We should note at once that the present review deals firstly with hydroxypyrimidine reactions not with any class of carbonyl compounds but with carbonyl compounds in traditional sense, i.e. with aldehydes, ketones, quinones and their derivatives and heteroanalogs (e.g., with nitroso compounds); secondly, we consider in the review the condensations with the reaction center on the $C^5$ of the pyrimidine ring.

In the simplest instances where the molecule of the carbonyl compound brought into reaction with hydroxypyrimidine lacks any groups alongside the carbonyl that would be capable to participate in transformations two alternatives of condensations can be distinguished. In the first case the reagents react in 1 : 1 ratio to furnish 5-α-hydroxyalkyl(aralkyl) hydroxypyrimidine derivatives that as a rule undergo intramolecular dehydration resulting in a 5-ylidene compound with an activated C=C bond. In the second case the condensation provides 5,5'-ylidenebishydroxypyrimidines (here condensation occurs at the ratio pyrimidine–carbonyl compound 2 : 1). Many among 5,5'-ylidenebishydroxypyrimidines are able to undergo intramolecular cyclization to afford polycyclic structures.

A large group of hydroxypyrimidines transformations involves a third component, C- or N-nucleophile. These reactions provide additional possibilities of creating pyrimidine-containing heterocyclic systems.

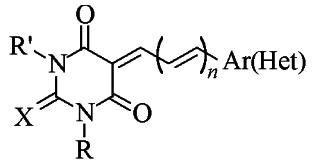
## II. 5-YLIDENE DERIVATIVES OF BARBITURIC ACIDS

Barbituric acids, their 2-thio- and 2-selenoanalogs, both substituted and unsubstituted at nitrogens, were most often studied as C-nucleophiles of pyrimidine character. Their reaction with carbonyl compounds, with aromatic or heteroaromatic aldehydes gives rise to 5-arylmethylene(hetarylmethylene)barbituric acids (**1**, $n = 0$), commonly in a high yield [1–49]. In this

* The study was carried out under financial support of investigations in the field of natural sciences from the Russian Ministry of Education, grant no. 168 of 2001.

way reaction of ferrocenecarboxaldehyde with the barbituric acid occurs [30]. In reaction between terephthalic aldehyde and barbituric acid take part both carbonyls of the former [31]. These transformations are treated in part or comprehensively in some reviews [32–34]. The character of solvent if it was not a pronounced base in the most cases did not affect the direction of reaction. Compounds (**1**) were obtained in water [2, 35], aqueous dioxane [35], aqueous DMF [36], in alcohols [7, 11, 13, 25, 26. 38–43], acetic acid [10, 12, 43–45], acetic anhydride [21], or in a mixture of acetic acid and acetic anhydride [3]. With benzaldehydes possessing electron-withdrawing groups the use of dehydration media (mixtures of acetic acid and acetic anhydride) are yet recommended [20]. The condensation is facilitated by some Lewis acids (e.g., anhydrous $ZnCl_2$) [46, 47]. A condensation of barbituric acid with several benzaldehydes was described to occur in DMF in the presence of trimethylchlorosilane [48].

Also some examples of acids (**1**) syntheses were described that were carried out with no solvent at microwave irradiation in the presence of KSF [50, 51].

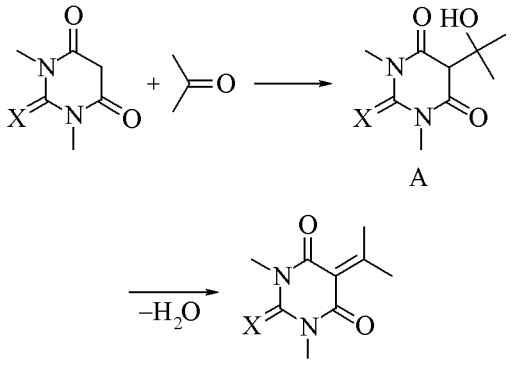


**1**

X = O, S, Se; *n* = 0–2.

These transformations are represented in a general scheme; the intermediate α-hydroxymethyl derivatives **A** are sometimes isolated (Scheme 1).

**Scheme 1.**

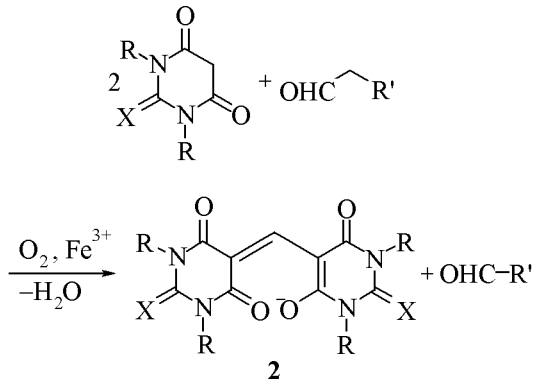


The substituted cinnamaldehydes, 5-aryl-2,4-pentadienals, and also their heterocyclic analogs readily undergo condensation with barbituric acids providing compounds (**1**, *n* = 1–3) [5, 15, 21, 39, 40, 52–54].

In these reactions instead of aromatic aldehydes the corresponding azomethines can be used [55].

At the same time the saturated aliphatic aldehydes with barbituric acids afford colored substances (chromogens) (**2**). The latter arise due to oxidative degradation of the intermediate 5-alkylidenebarbituric acids under the aerobic conditions [56, 57]. By an example of reaction between 1,3-diethylbarbituric acid and hexanal in the presence of air oxygen and $Fe^{3+}$ ions Scheme 2 was demonstrated to be valid [57].

**Scheme 2.**

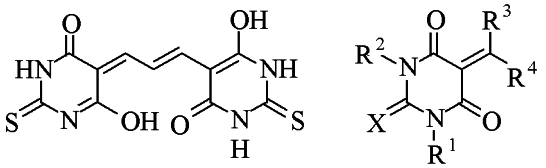


**2**

X = O, S; R = H, Alk; R' = Alk.

In the course of the process crotonic condensation and formation of pyrano[2,3-*d*]pyrimidine derivatives as precursors of the colored substances were observed [56]. Alkanals, 2-alkenals that may arise also *in situ* as products of the crotonic condensation, and 2,4-alkadienals react with 2-thiobarbituric acids to furnish yellow, orange, or red pigments [58, 59] depending on the heating time under aerobic conditions. The kinetics of these pigments formation were studied in [59]. We believe the information on isolation of individual 5-alkylidenebarbituric acids in reaction between aliphatic aldehydes and barbituric acid based only on the data of elemental analysis to be erroneous [60].

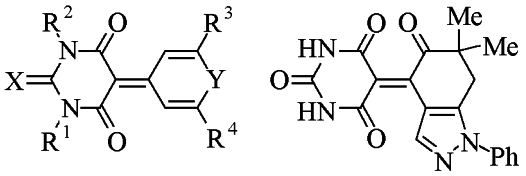
Malonic aldehyde, same as α,β-unsaturated aldehydes affords with 2-thiobarbituric acid a pigment that was assigned structure (**3**) [61]. This pigment formation is used in quantitative and qualitative analysis on malonic aldehyde in various, in particular biological, liquids by spectrophotometric [62] and fluorscence [63] methods. It was shown that the sensitivity of the method was reduced in neutral and alkaline media [62].



**3** **4**

A reaction between barbituric acid and polysaccharidepolyaldehydes was described [64]. Depending on the condensation conditions and reagents ratio the products contain in their molecules from one to three barbituric acid molecules per monomer unit.

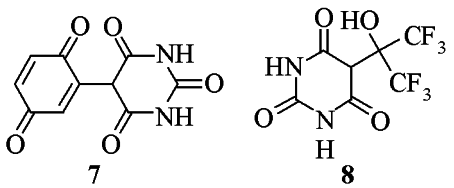
After heating barbituric acids with simple ketones (acetone, butanone) the corresponding 5-alkylidene derivatives (**4**) were isolated [65]. 5-Ylidene derivatives (**4**) arise also in reactions of barbituric acid with 4-cyano-3,4,5-triphenyl-2,5-cyclohexadiene-4-one in the presence of $TiCl_4$ [66]. 2-Monosubstituted and 2,6-disubstituted γ-pyrones, 4-pyridones with 1,3-(un)substituted barbituric and 2-thiobarbituric acids also afford 5-ylidenebarbituric acids (**5**) at heating in a mixture of acetic acid and acetic anhydride [67–70]. If the pyran ring contained carboxy groups in *2* and *6* positions, the condensation is accompanied by decarboxylation. The attempts to prepare by this procedure 5-(4-pyranylidene)barbituric acid from γ-pyrone or 4-oxo-4*H*-pyran-2,6-dicarboxylic acid were unsuccessful [67].



**5** **6**

**5**, X = O, S; Y = O, NH, C(CN)Ph.

In reaction with barbituric acid giving ylidene derivative (**6**) only one of keto groups from 6,6-dimethyl-1-phenyl-4,5,6,7-tetrahydroindazol-4,5-dione, is involved that in *4* position [71].



**7** **8**

However in reaction with *p*-quinone or 1,4-naphthoquinone the barbituric acid first adds to the C=C bond, and then the adduct is oxidized into 2-(5-pyrimidinyl)-*p*-quinone (**7**) [72]. This reaction was advanced as a method of quantitative analysis of *p*-quinone and 1,4-naphthoquinone in solution. Kinetics of reaction between barbituric acid and *p*-quinone were also studied [72].

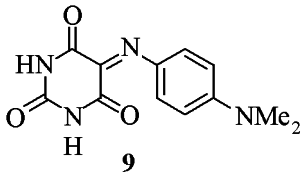
The hexafluoroacetone with barbituric acid at 20°C in the presence of tertiary amines instead of 5-ylidene derivative affords in a high yield 5-(1-hydroxy-1-trifluoromethyl-2,2,2-trifluoroethyl)barbituric acid (**8**) [73]. The substances similar to compound (**8**) apparently may be regarded as intermediates in formation of derivatives of (**1**–**6**) type. It is presumable that the dehydration of compound (**8**) is an equilibrium process, and the equilibrium is virtually completely shifted to compound (**8**) because of high electrophilicity of the C=C bond in the corresponding 5-ylidene derivative.

Kinetics of reactions of barbituric and 2-thiobarbituric acids with *p*-dimethylaminobenzaldehyde were studied [41, 74], benzaldehyde, anisaldehyde, salicylaldehyde, 4-hydroxybenzaldehyde [74], of 1,3-dimethylbarbituric acid with 2-nitro-, 4-nitro-, 2,4-dinitrobenzaldehydes [75]. The formation of 5-arylmethylene derivatives from barbituric and 2-thiobarbituric acids (**1**) and benzaldehyde in methanol, 2-methyl-1-propanol, acetonitrile [74] follows the kinetic law of a second order process, with first order with respect to each reagent. The limiting stage of the reaction is dehydration of the intermediate product, 5-(α-hydroxy-4-dimethylaminobenzyl)barbituric acid A (Scheme 1) [41, 74]. The reactivity of 2-thiobarbituric acid turned out to be higher than that of barbituric acid. Also was revealed that with decreasing polarity of the alcohol the reactivity of the barbituric acids is reduced. This is apparently due to the decrease in the solvation energy of barbituric acid anion and aldehyde that on the one hand reduces the activation enthalpy of the reaction and on the other hand increases the role of the entropy factor in the course of condensation and thus makes its contribution more important than that of the activation enthalpy.

The reaction between *p*-quinone and barbituric acid in aqueous methanol affording quinone (**7**) is also described with the second order kinetic equation, with the first order in each component [76].

Nitroso compounds as azaanalogs of aldehydes behave very similarly to the latter with respect to barbituric acids. For instance, *N*,*N*-dimethyl-4-nitrosoaniline afforded in reaction with barbituric acid the corresponding 5-imino derivative (**9**). Yet the

nitroso compound turned out to be less reactive than the aldehyde, but the kinetic relations in the reaction were the same [41].

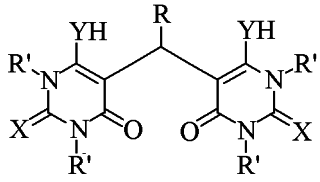


**9**

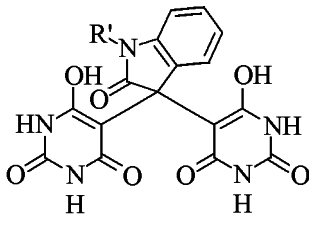
## III. 5,5'-YLIDENEBISBARBITURIC ACIDS

The known examples of formation of 5,5'-ylidenebisbarbituric acids (**19**), condensation products of composition aldehyde–pyrimidine 1 : 2, are relatively rare. For instance, barbituric and 2-thiobarbituric acids with benzaldehydes in pyridine yielded dipyridinium salts of 5,5'-benzylidenebis[(2-thio)barbituric] acids [77, 78], and from 2-thiobarbituric acid and benzaldehyde in aniline was obtained a dianilinium salt [79]. Note that unlike barbituric acid which furnishes these salts only with aldehydes containing electron-withdrawing groups [77], the 2-thiobarbituric acid is capable of reacting in this way also with less electrophilic aldehydes [78]. At acidifying these salts the tricyclic structure decomposed into 5-arylmethylene(2-thio)barbituric (**1**) and (2-thio)barbituric acids [77, 78]. Some bisderivatives of barbituric acids are stable not only as salts. For instance, with isatin and 1-methylisatin the barbituric acid gives the corresponding bisderivatives (**11**) at the $C^3$=O group of isatin [80], and 2-thiobarbituric (but not barbituric) acid with the most electrophilic aromatic aldehydes in alcohol provides the corresponding bis-2-thiobarbituric acids [78].

1,3-Dimethyl-6-mercaptouracil with formaldehyde in methanol at 20°C also forms 5,5'-methylenebis derivative (**10**) [81].



**10**

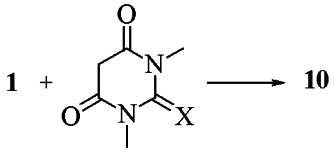


**11**

**10**, **11**, X, Y = O, S; R = H, Ar; R' = H, Me.

It is presumable that compounds (**10**) and (**11**) arise from addition of a 2-thiobarbituric acid molecule to the 5-ylidenebarbituric acids (**1**) that are intermediates in these reactions. This assumption was confirmed by special experiments [77] (Scheme 3).

**Scheme 3.**

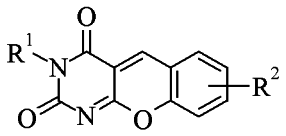


However kinetics of 5,5'-ylidenebarbituric acids formation were not studied up till now. As the most probable formation mechanism of compounds (**10**) and (**11**) may be regarded addition of a barbituric acid anion to the C=C bond in the corresponding 5-arylmethylenebarbituric acid (**1**).

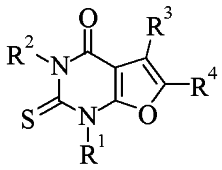
## IV. INTRAMOLECYLAR CYCLIZATION

The condensation products from barbituric acids and carbonyl compounds containing other reactive groups can undergo transformations providing versatile pyrimidine-containing heterocyclic systems.

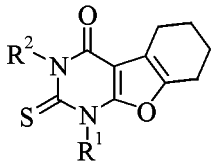
5-(*o*-Halobenzylidene)barbituric acids (**1**) when heated to 220–260°C suffer dehydrohalogenation affording 5-deaza-10-oxaflavins (**12**) in 85–92% yield [82].



**12**



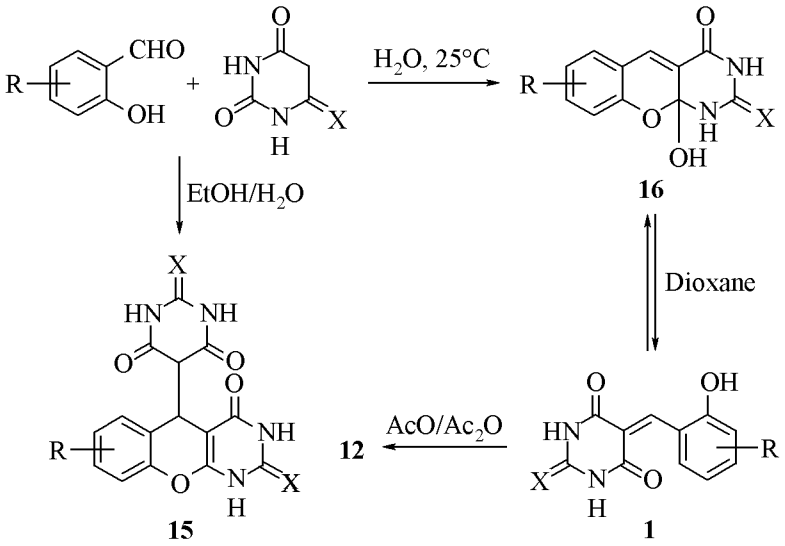
**13**



**14**

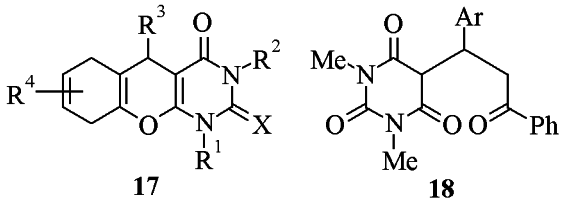
In contrast to *o*-halobenzaldehydes the aliphatic halocarbonyl compounds apparently alkylate the $C^5$ atom of the barbituric acids, and their carbonyl group takes part in formation of an ether bond with a carbonyl at $C^6$ of the pyrimidine ring. For instance, 1,3-diaryl-2-thiobarbituric acids with chloroacetone afford 1,3-diaryl-6-methyl-4-oxo-2-thioxo-1,2,3,4-tetrahydrofuro[2,3-*d*]pyrimidines (**13**, $R^3$ = H, $R^4$ = Me) [83], and with 2-bromocyclohexanone tetrahydrobenzofuro[2,3-*d*]pyrimidines (**14**) [84].

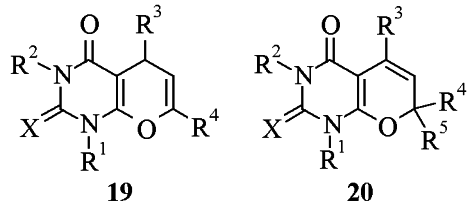
**Scheme 4.**




X = O, S.

Chromenopyrimidine derivatives (**12**) arise also at heating 5-salicylidenebarbituric acids obtained from barbituric acids and salicylaldehydes [82, 85]. Besides these compounds barbituric acids with salicylaldehydes afford 4 more types of substances: (1) 5-salicylidene derivatives (**1**) same as with the other benzaldehydes [86]; (2) 5,5'-salicylidenebisbarbituric acids (**10**) [82]; (3) 1,5-dihydro-5-[2-oxo(thioxo)-4,6-dioxohexahydropyrimidin-5-yl]-2*H*-chromeno[2,3-*d*]-pyrimidine-2,4(3*H*)-diones (**15**) in 83–85% yield [29, 86, 87], and (4) 10*a*-hydroxy-2,3,4,10*a*-tetrahydro-1*H*-chromeno[2,3-*d*]pyrimidine-2,4-diones (**16**) [86] (Scheme 4). Apparently one type of the above compounds was obtained in [6].




Compounds (**12**) are also obtained when 5-salicylidenebarbituric acids (**1**) prepared from barbituric acids and salicylaldehyde are subjected to intramolecular cyclization effected by acetic anhydride [35]. Hydrogenation of chromenopyrimidines (**12**) with sodium borohydride provides compounds (**17**, $R^3$ = H) that also arise at cyclization of 5-(*o*-hydroxybenzyl)barbituric acids or directly at boiling barbituric acid and salicylaldehydes in alcoholic solution containing methanesulfonic or *p*-toluenesulfonic acid [35].

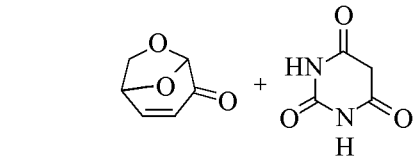
The condensation product of barbituric acid and 4,6-dihydroxypyrimidine-5-carboxaldehyde enters a complicated chain of transformations apparently due to nucleophilic attack on the $C^6$ atom in the dihydroxypyrimidine fragment by the carbonyl from the barbituric acid. As a result forms 5-aminomethylenebarbituric acid or derivatives thereof [88].

Unsubstituted and 1,3-disubstituted 2-thiobarbituric acids with benzoin on boiling in benzene in the presence of *p*-toluenesulfonic acid give rise to furo-[2,3-*d*]pyrimidines (**13**, $R^3$ = $R^4$ = Ph) in 55–60% yield [89].

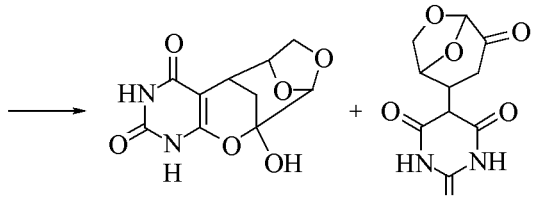
Yet with various monosaccharides the barbituric acids do not yield fused heterocyclic systems but only 5-glucopyranosyl derivatives [90].

Another approach to pyrimidines fused with oxygen-containing rings consists in reacting barbituric acids with α,β-unsaturated ketones. Therewith the barbituric acids first add to the activated C=C bond, and in reaction with chalcones the addition products (**18**) have been isolated in 67–76% yield [29]; then at the expense of the carbonyl groups forms a pyran ring to yield finally pyrano[2,3-*d*]pyrimidines (**19**). As examples of these reactions may be cited those of

barbituric acids with chalcones [91], 4-aryl-3-butene-2-ones [92, 93] in acetic acid in the presence of $P_2O_5$. However at boiling in methanol in the presence of triethylamine the thiobarbituric acids only add to the activated C=C bond of the chalcone. Similarly were prepared 1,3-diaryl-5,7,7-trimethyl-4-oxo-2-thioxo-1,2,3,4-tetrahydro- 7*H*-pyrano[2,3-*d*]pyrimidines (**20**) from 4-methyl-3-penten-2-one [94]. Compounds (**20**) were also obtained at heating 2-thiobarbituric acid with excess acetone or methyl ethyl ketone in the presence of triethylamine [62]. The reaction between barbituric acid and levoglucosenone (**21**) gives rise to a mixture of two substances: a product of barbituric acid addition across the C=C bond of levoglucosenone (**23**), and a product of cyclization of the latter into semiketal (**22**) [95].



**21**



**22** **23**

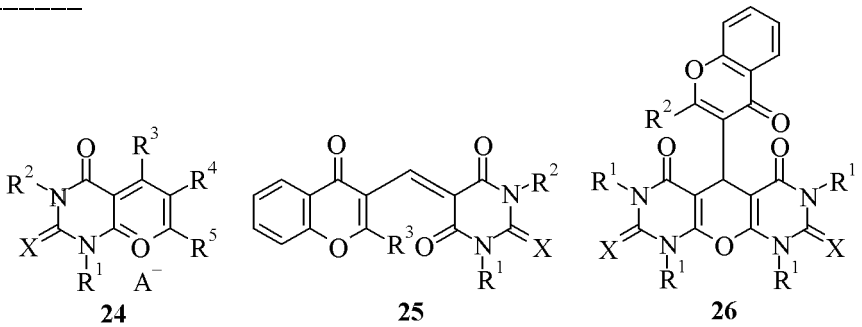
The presence of a hydroxy group in the *ortho*-position of the benzene ring in chalcone also provides a possibility of utilizing the group in preparation of pyrano[2,3-*d*]pyrimidine derivatives. For instance, the reaction of 1,3-diaryl-2-thiobarbituric acids with 4-(2-hydroxyphenyl)-3-buten-2-one in pyridine affords compounds (**17**, $R^3$ = $ch_2Ac$, $R^4$ = H) in 65–75% yield [96].

The reaction of 1,3-disubstituted barbituric acids with α,β-unsaturated β-chloroketones or β-diketones in the presence of perchloric or phosphoric acids yielded pyrano[2,3-*d*]pyrimidine derivatives (**24**) [97].

For a number of γ-pyrones and 3-acylchromones reactions with barbituric acids (X = O, S, R = H, Me, Et, Ph) were described carried out at heating first in a mixture of pyridine with triethylamine, and then in acetic acid containing sulfuric acid where occurred not only the condensation of the reagents but also a rearrangement with opening of the pyran ring followed by its closure [98–101]. From these transformations resulted in particular compounds (**25**, **26**) [98, 99].

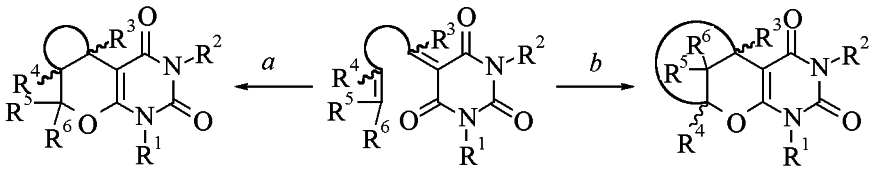
5-Arylmethylene- or 5-alkylidenebarbituric acids possessing a conjugated heterodiene fragment enter into intramolecular Diels–Alder reactions when the ylidene part of their molecules contains double bonds. The barbituric acid and its 1,3-substituted derivatives reacting with aldehydes of terpene or steroid nature [102], e.g., with (*R*)- and (*S*)-citronellal [103, 104], 6-alkenals [105, 106], and *o*-allyloxy-, *o*-allylamino- or *o*-(3-butenyl)benzaldehydes [107–110] afford the corresponding unsaturated 5-ylidene compounds that further undergo intramolecular hetero-Diels–Alder reactions with participation of the carbonyl oxygen from position *4*(*6*) of the pyrimidine ring and of the double bonds at $C^{\alpha}$ atom or at more removed atoms of the ylidene fragment. Therewith depending on the structure of the olefin fragment occurs either version *a* or *b* of its coordination with the heterodiene system, or both. As a result of these conversions form in good yields tri- or polycyclic pyrimidine-containing compounds (**27, 28**) (Scheme 5). At the use of chiral catalysts the reaction occurs enantioselectively [111].

Apart from the reaction with malonic aldehyde described above some other cases of barbituric acids reactions with β-dicarbonyl compounds are known. The condensation of 1,3-diaryl-2-thiobarbituric acid with ethyl acetoacetate gives rise to pyrano[2,3-*d*]-pyrimidines (**29**) [112]. By treating pyranopyrimidines (**29**) with the Grignard reagent the carbonyl group in position *7* was substituted by two alkyls, and



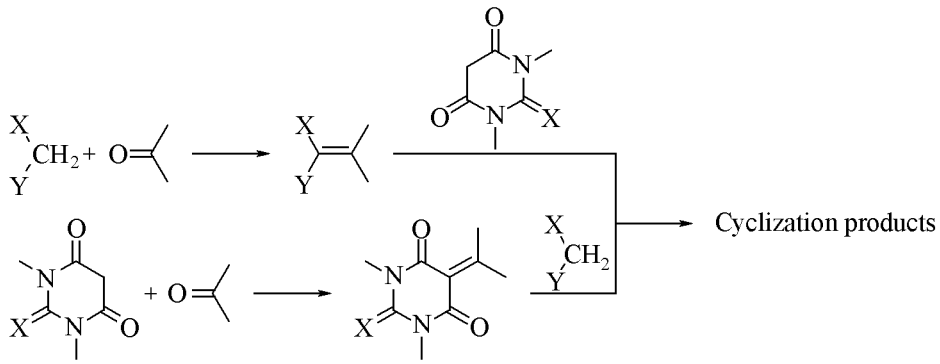
**24** **25** **26**

**Scheme 5.**

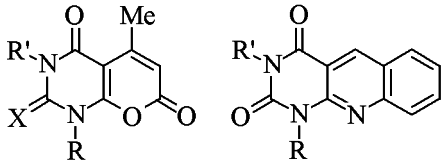


**27** **1** **28**

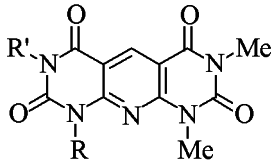
**Scheme 6.**




the resulting substances (**20**) possessed antibacterial and fungicidal properties.

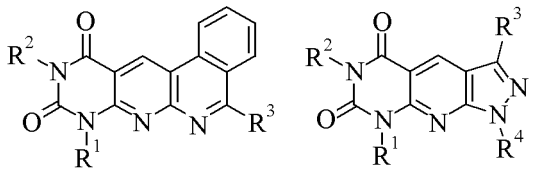


**29** **30**



**31**

By condensation of barbituric acids with *o*-triphenylphosphoranylideneaminobenzaldehyde or with 6-amino-1,3-dimethyluracil-5-carboxaldehyde in pyridine derivatives of pyrimido[4,5-*b*]quinoline (**30**, R = R' = H) and pyrido[2,3-*d*:6,5- *d*']dipyrimidine (**31**) respectively were obtained (yields 45–67%) [113]. Reaction of 3-amino-1-(*p*-tolyloxy)isoquinoline-4-carboxaldehyde with barbituric acid yielded 2,4-dioxo-10-(*p*-tolyloxy)-1,2,3,4-tetrahydrobenzo[*f*]-pyrimido[4,5-*b*]naphthyridine (**32**) [114]. The condensation of 1,3-diaryl-2-thiobarbituric acids with 3-R-5-amino-1-phenyl-4-pyrazolecarboxaldehydes gives rise to derivatives of 1*H*-pyrazolo[4',3':5,6]-pyrido[2,3-*d*]pyrimidine (**33**) [115].



**32** **33**

The amino group in a carbonyl compound can be produced *in situ*. Thus pyrimido[4,5-*b*]quinoline derivatives (**30**, R = R' = H) were obtained by reduction of 5-*o*-nitrobenzylidenebarbituric acids (**1**) [116].

## V. THREE-COMPONENT CONDENSATIONS

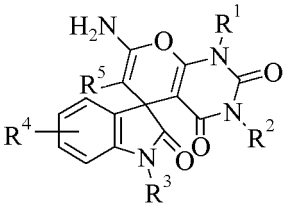
**Condensations involving C-nucleophiles.** The reactions of barbituric acids, carbonyl compounds, and C-nucleophiles can be described by a scheme including two pathways leading from the reagents to final products (Scheme 6). Examples for both paths were described.

Relatively seldom are events of barbituric acids addition to the activated multiple bonds of molecules from the other compounds classes arising from condensation of C-nucleophiles and aldehydes. As example the addition of barbituric acid to 2-benzylidene-1,3-indandione [117] and to arylidenemalononitriles may be cited [118].

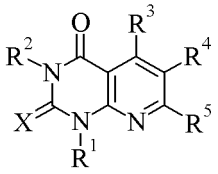
The final products of barbituric acid addition to 3-cyanomethylene-2-indolinones are spiro{oxindol-

3,5'-(5*H*-pyrano[2,3-*d*]pyrimidines)} (**34**) [119–121].

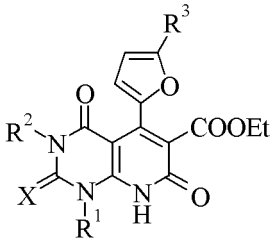
The addition of cyclohexanone and malononitrile to 5-arylmethylenebarbituric acids (**1**) was described [122, 123]. The probability of addition of the second molecule of CH-acid to the exocyclic C=C bond according to quantum-chemical calculations depends on the electron density on the bond [124]. Some cases of organomagnesium compounds addition to the C=C bond in the 5-arylmethylenebarbituric acids (**1**) are known [125].



**34**



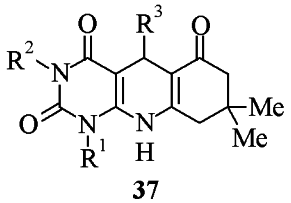
**35**



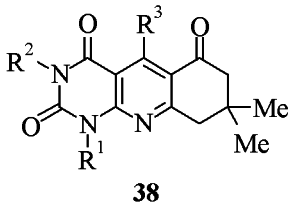
**36**

In reaction of 5-furfurylidenebarbituric acid with ethyl cyanoacetate or diethyl malonate in the presence of ammonium acetate were obtained respectively ethyl 7-amino-2,4-dioxo-5-(2-furyl)-1,2,3,4-tetrahydropyrido[2,3-*d*]pyrimidine-6-carboxylates (**35**, X = O, $R^3$ = 5-furyl, $R^4$ = COOEt, $R^5$ = $NH_2$) and ethyl 2,4,7-trioxo-5-(2-furyl)-1,2,3,4,7.8-hexahydropyrido-[2,3-*d*]pyrimidine-6-carboxylates (**36**) [126]. Compounds (**35**, X = O, S, $R^3$ = Ph, 2-furyl, 2-thienyl, 4-pyridyl, $R^4$ = COOEt, $R^5$ = Ph, $NH_2$) were prepared by reaction between aryl(hetaryl)methylenemalononitriles, aryl(hetaryl)cyanoacetates, and aryl-(hetaryl)benzoylacetates with 2-thiobarbituric acids [127].

5-Arylmethylenebarbituric acids (**1**) undergo condensation with 3-amino-5,5-dimethyl-2-cyclohexenone to furnish 5-aryl-8,8-dimethyl-2,4,6-trioxo-1,2,3,4,5,6,7,8,9,10-decahydropyrimido[4,5-*b*]quinolines (**37**) that are readily oxidized even with hydroxylamine into 5-aryl-8,8-dimethyl-2,4,6-trioxo-1,2,3,4,6,7,8,9-octahydropyrimido[4,5-*b*]quinolines (**38**) [128]. The ketone carbonyl group is not involved into these transformations.



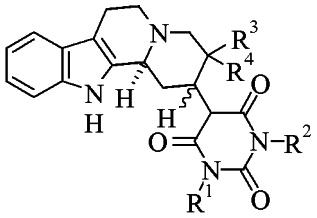
**37**



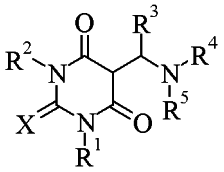
**38**

The role of the CH-acid component can be played by phenacylpyridinium bromide, and thus in acetic acid in the presence of ammonium acetate succeeds preparation of pyrido[2,3-*d*]pyrimidines (**35**, $R^3$ = Ar, $R^4$ = H, $R^5$ = Ph) in 40–82% yields [13, 38].

The reaction between 1,3-dimethylbarbituric acid, pyridoindoloacetaldehyde derivative, and 1-benzyloxy-1-butene provides analogs of alkaloid strychtozidine (**39**) [129]. This multistage process includes as a stage a hetero Diels–Alder reaction.



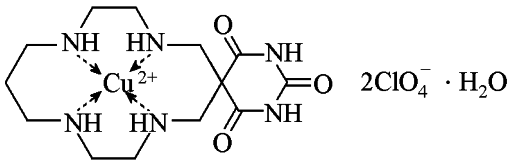
**39**



**40**

**Condensation involving N-nucleophiles.** The 2-thiobarbituric acids enter into Mannich reaction with formaldehyde, piperidine or morpholine [10], with anilines [16, 17] giving rise to the corresponding 5-aminomethyl derivatives (**40**). Mannich bases (**40**) form also in reaction of 5-arylidenebarbituric acids (**1**) with piperidine or morpholine thus suggesting that the arylidene derivatives (**1**) may be regarded as organic Lewis acids [130].

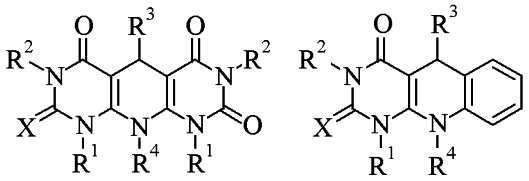
A similar reaction of barbituric acid with formaldehyde and $Cu(danda)^{2+}\,ClO_4^-$ (danda = 3,7-diazanonane-1,9-diamine) in water–methanol mixture results in copper(II) complex with a macroazacyclane ligand that contains barbituric acid as a spirosubstituent in the macroring (**41**) [131].



**41**

N-Nucleophile may act as a unit connecting two pyrimidine moieties into a pyridine ring. Thus a three-component condensation of barbituric or 1,3-diaryl-2-thiobarbituric acids with aromatic

aldehydes and ammonia or anilines provides derivatives of pyrido[2,3-*d*:6,5-*d*']dipyrimidine (**42**) [132, 133].



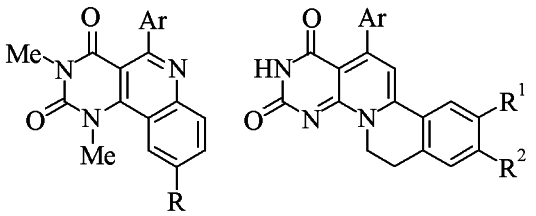
**42** **43**

5-(1,3-Diaryl-3-oxopropyl)-1,3-dimethylbarbituric acids (**18**) obtained from 1,3-dimethylbarbituric acid and appropriate chalcones on heating with ammonium carbonate in acetic acid afford in 60–68% yield pyrido[2,3-*d*]pyrimidines (**35**, $R^1$ = $R^2$ = Me, $R^3$ = Ar, $R^4$ = H, $R^5$ = Ph) [29].

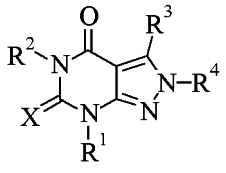
1,3-Diaryl-2-thiobarbituric acids react with *N*-methylaniline and *p*-tolualdehyde or *o*-nitrobenzaldehyde yielding pyrimido[4,5-*b*]quinoline derivatives (**43**) [134].

On the other hand 5-arylmethylene-1,3-dimethylbarbituric (**1**) or 5-(1,3-diaryl-3-oxopropyl)-1,3-dimethylbarbituric (**18**) acids at heating with aniline to 150°C are converted in 48–60% yield into pyrimido-[5,4-*c*]quinoline-2,4(1*H*,3*H*)-diones (**44**) [29].

Barbituric acid, benzaldehydes, and substituted or unsubstituted 1-methyl-3,4-dihydroisoquinolines condense providing derivatives of 8,15,17-triaza-D-homogonane (**45**) [135]. The same compounds arise in reactions of 1-methyl-3,4-dihydroisoquinoline with 5-arylmethylenebarbituric acids (**1**) [136, 137].



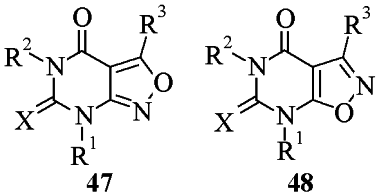
**44** **45**



**46**

The reaction of 5-arylmethylenebarbituric acids (**1**) with hydrazines provides pyrazolo[3,4-*d*]pyrimidine derivatives (**46**) [11. 138, 139]. Therewith the sulfur atom in *2*9 position of pyrimidine ring can be replaced by a hydrazono group (**46**, X = $NNH_2$) [139].

With hydroxylamine the 5-arylmethylenebarbituric acids (**1**) afford isoxazolo[3,4-*d*]pyrimidine derivatives (**47**) [11] or isoxazolo[5,4-*d*]pyrimidine derivatives (**48**) [140].



**47** **48**

Thus the reactions of barbituric acids with carbonyl compounds are promising for preparation of a great number of fundamental structures containing the pyrimidine ring, in particular, fused with other heterocycles.

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