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OXIMES OF SIX-MEMBERED HETEROCYCLIC COMPOUNDS WITH TWO OR THREE HETERO-ATOMS: I. SYNTHESIS AND STRUCTURE (REVIEW)

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Data on methods for the production of and on the structure of pyridazine, pyrimidine, pyrazine, triazine, oxazine, thiazine, oxadiazine, and thiadiazine aldoximes, ketoximes, and amidoximes, and their derivatives are reviewed.

Keywords: oxadiazine, oxazine, oxime, pyrazine, pyridazine, pyrimidine, thiadiazine, thiazine, triazine.

The oximes of heterocyclic compounds with two or three heteroatoms are widely used as intermediates in fine organic synthesis. In this review the basic methods for the synthesis of pyridazine, pyrimidine, pyrazine, triazine, oxazine, thiazine, oxadiazine, and thiadiazine aldoximes, ketoximes, and amidoximes and their derivatives are summarized. The basic methods for investigation of the structure of the oximes of six-membered heterocyclic compounds with two or three heteroatoms are discussed in relation to isomerism. The reactions and biological activity of the oximes of heterocyclic compounds with two or three heteroatoms will be examined in the second part of the review.

1. SYNTHESIS

1.1. Synthesis of Pyridazine Aldoximes, Ketoximes, and Amidoximes

The classical method for the synthesis of pyridazine aldoximes and ketoximes is based on the reaction of the aldehyde or ketone with hydroxylamine hydrochloride in ethanol [1] or in an aqueous solution of K_2CO_3 [2]. Pyridazine aldoximes were obtained as minor products during the nitrosation of the alkyl chain of the alkyl derivatives in the $NaNO_2$/HCl system [3].

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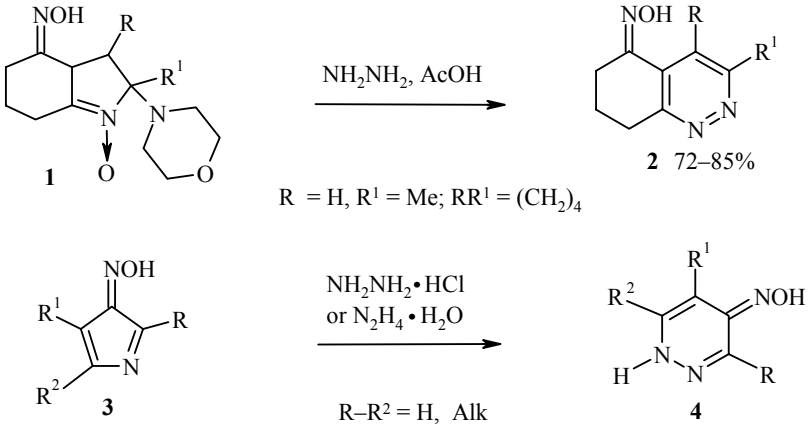
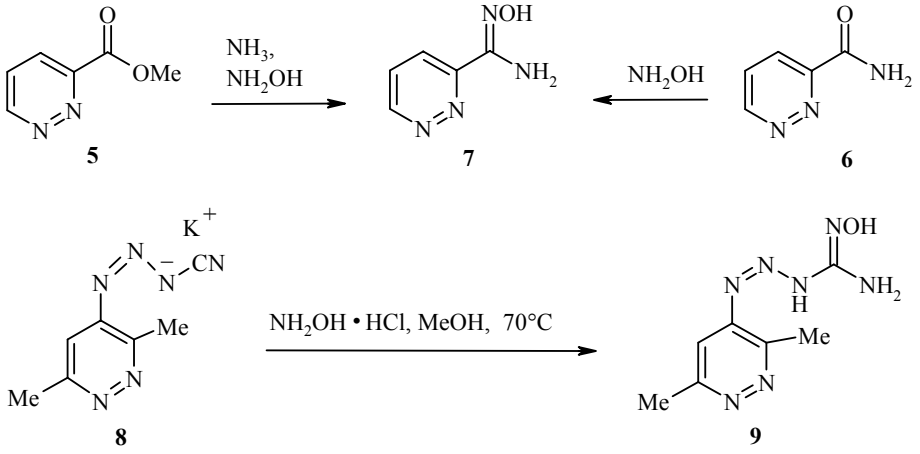
Pyrroline N-oxides **1**, which react with hydrazine in boiling acetic acid as equivalents of 1,4-dicarbonyl compounds, form the oximes of hydrogenated cinnolines **2** [4]. In reaction with hydrazine the pyrroline 3-oximes **3** give pyridazine oximes **4** [5].

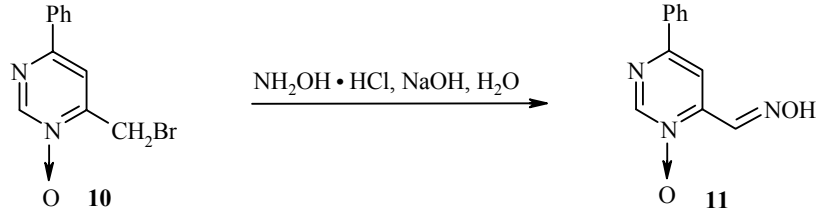


A few papers have been devoted to the synthesis of pyridazine amidoximes. Thus, pyridazine-3-amidoxime [6, 7], cinnoline-4-amidoxime [8], and 4-benzopyridazine amidoxime [9] are formed readily from the corresponding nitriles and hydroxylamine. Pyridazine-3-amidoxime **7** was obtained from the corresponding ester **5** in the NH_3/NH_2OH system or in the reaction of the amide **6** with hydroxylamine [6, 7]. It is interesting that the nitrile derivative of pyridazine **8** gives the amidoxime **9** with a yield of 72% during reaction with hydroxylamine hydrochloride [10].

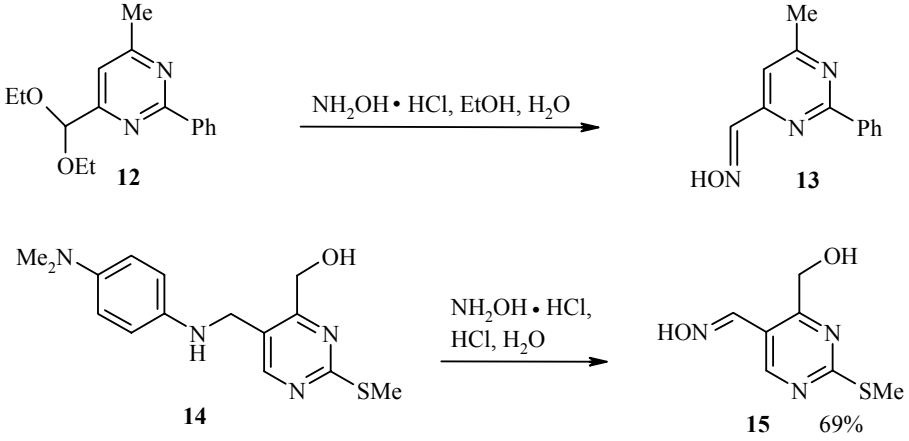


1.2. Synthesis of Pyrimidine Aldoximes, Ketoximes, and Amidoximes

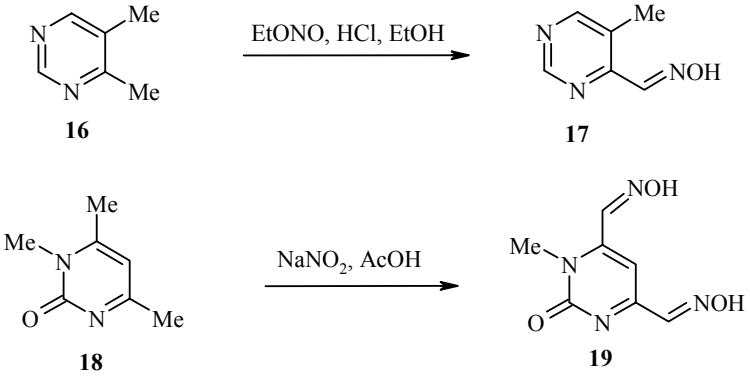
Pyrimidine aldoximes and ketoximes are obtained by the reaction of the aldehyde or ketone with hydroxylamine hydrochloride in ethanol [11-13], methanol [14], water [15], DMSO [16], $Na_2CO_3/EtOH/H_2O$ [17], or NaOAc/EtOH [18]. 6-Bromomethyl-4-phenylpyrimidine 1-oxide **10** reacts readily with hydroxylamine hydrochloride in NaOH solution at room temperature with the formation of the oxime of 6-formyl-4-phenylpyrimidine 1-oxide **11** with a yield of 80%. 6-Dibromomethyl-4-phenylpyrimidine 1-oxide reacts similarly [19].



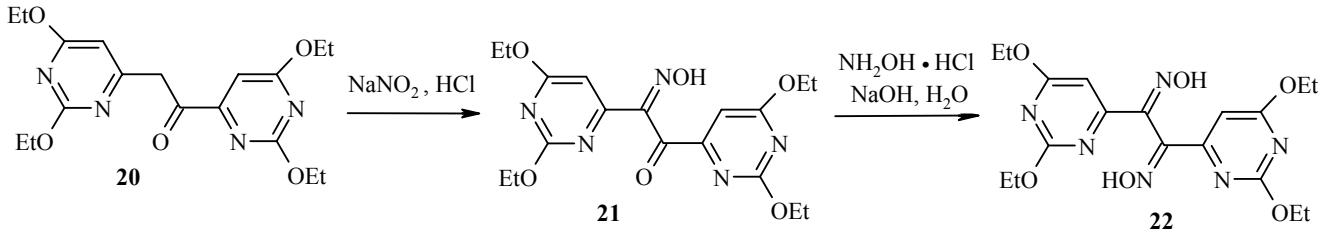
The aldoxime **13** can be obtained directly from the acetal **12** [20]; during the action of hydroxylamine on N-(4-hydroxymethyl-2-methylthiopyrimidin-5-ylmethyl)-N',N'-dimethyl-4-phenylenediamine **14** the dimethylaminophenylamino group is removed, and the oxime **15** is formed [21].



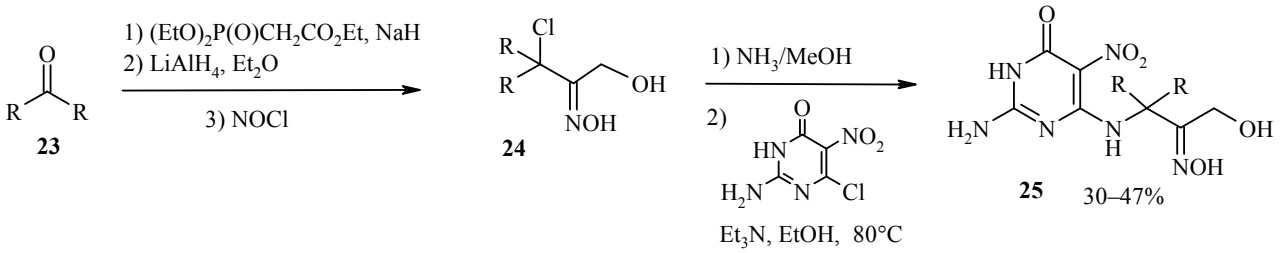
Pyrimidine oximes were obtained as a result of nitrosation of the alkyl chain in the alkyl derivatives. The employed nitrosation systems were EtONO/KNH_2/NH_3 [22], EtONO/HCl/EtOH [23], AmONO/HCl/EtOH [24], and *i*-AmONO/NH_3 [25]. It is interesting that the nitrosation of 4-methylpyrimidine with isoamyl nitrite takes place both in acidic and in basic media. Thus, the reaction of 4-methylpyrimidine in the *i*-AmONO/*t*-BuOK system gives pyrimidine-4-aldoxime with a 40% yield. If the reaction with isoamyl nitrite is conducted in the presence of HCl the yield amounts to 80% [26]. In the reaction of 4,5-dimethylpyrimidine **16** with EtONO only the activated methyl group at position 4 is nitrosated, and this leads to the formation of 5-methylpyrimidine-4-aldoxime **17** as the only product [27]. However, the reaction of 1,4,6-trimethylpyrimidin-2-one **18** with an excess of sodium nitrite gives 4,6-bis(hydroxyiminomethyl)-1-methylpyrimidine **19** with a yield of 75% [28].



The reaction of the ketone **20** with sodium nitrite in a water–dioxane solution of HCl gives the α-keto oxime **21** with an 86% yield. Further reaction of compound **21** with hydroxylamine hydrochloride in an aqueous solution of alkali leads to the formation of the dioxime **22** [29].

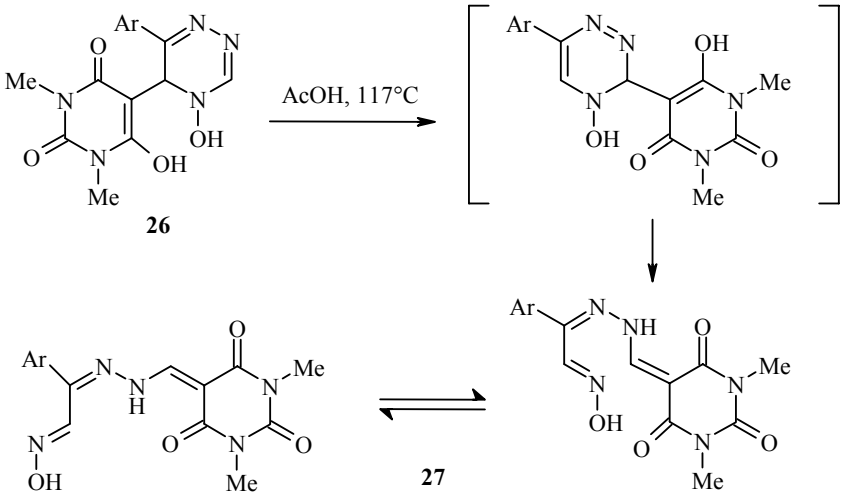


The synthesis of the oxime derivatives of 2-amino-5-nitro-3H-pyrimidin-4-one was set out in detail in the two patents [30, 31]. Reaction of the ketones **23** with the Wittig reagent, reduction, and nitrosation-chlorination led to the oxime intermediate **24**, and further reaction of compound **24** with NH_3/MeOH and 2-amino-6-chloro-5-nitropyrimidin-4-one led to the oxime **25** [31].

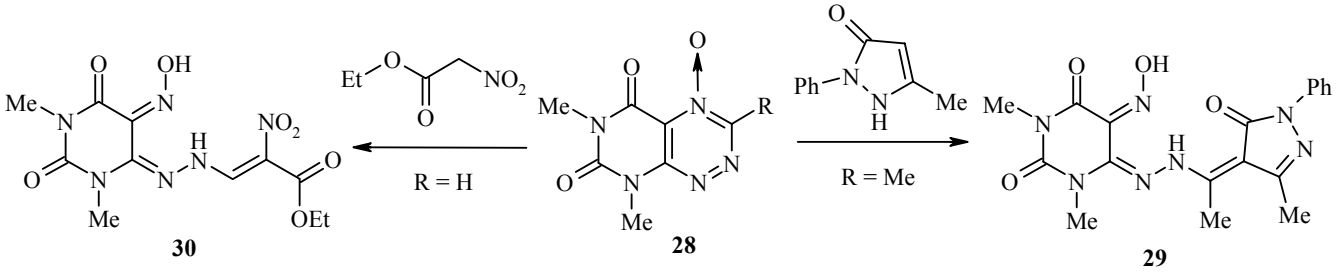


R = Et; RR = $-(CH_2)_6-$

When the derivatives of 2,4,6-pyrimidinetrione **26** are boiled in acetic acid the 1,3,4-triazine ring is opened, and 3-aryl-6-(1,3-dimethyl-2,4,6(1H,3H,5H)-trioxo-5-pyrimidylidene)-1-hydroxy-1,4,5-triazahexa-1,3-dienes **27** are formed with yields of 80-85% [32].

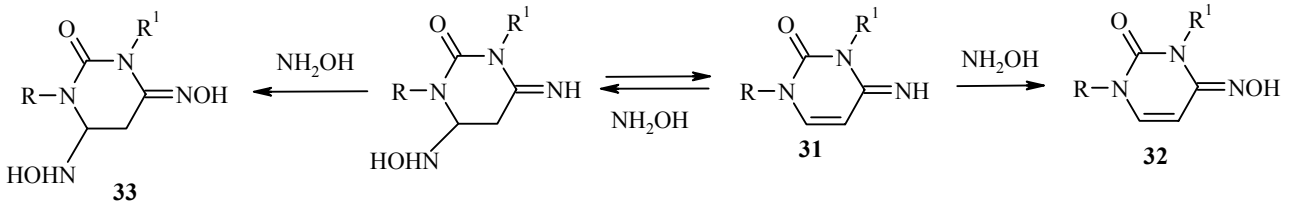


The 1,2,4-triazine ring is also opened in the reaction of 3-substituted 6,8-dimethyl-8H-pyrimidino-[5,4-*e*][1,2,4]triazine-5,7-diones **28** with C-nucleophiles, such as 5-methyl-2-phenylpyrazol-3-one and ethyl nitroacetate, leading to the products **29** and **30** containing a hydroxyimino group at position 5 of the pyrimidine ring [33].



30 **28** **29**

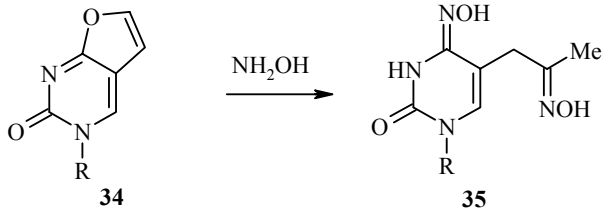
Cytosine, 1-methylcytosine, and 1,3-dimethylcytosine **31** react with hydroxylamine to form a mixture of two products **32** and **33** [34, 35]. However, cytosine and dry hydroxylamine at 37°C form only the product **32** (R = R^1 = H) with a good yield.



33 **31** **32**

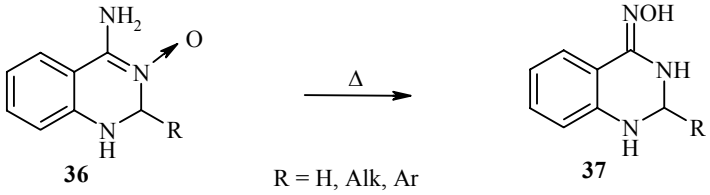
R, R^1 = H, Me

Furopyrimidones **34** react with hydroxylamine with opening of the furan ring and the formation of the 4-oximes of 1-alkyl-5-(2-hydroxyiminopropyl)-1H-pyrimidine-2,4-dione **35** [36].



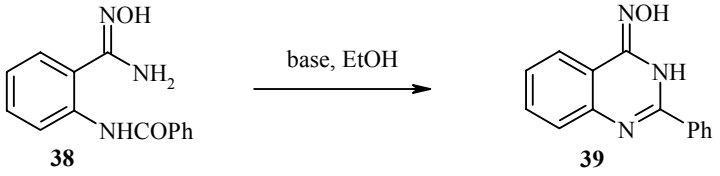
34 **35**

Thermolysis of 6-amino-1,2-dihydro-3H-quinazolines **36** gives the quinazoline oximes **37** as the only products [37].

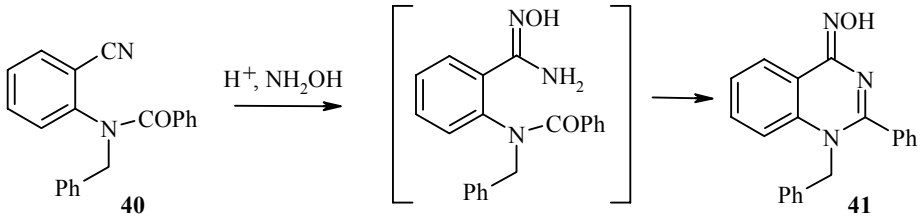


36 **37**

In an acidic or basic medium the N-benzoyl derivatives of the oximes of 2-aminobenzamides undergo cyclization to quinazolone oximes [38, 39]. For example, the amidoxime **38** is easily transformed into the corresponding quinazolone 4-oxime **39** with a yield of 67%. The reaction of the nitrile **40** with hydroxylamine also leads to the formation of a substituted quinazolone 4-oxime **41** [39].

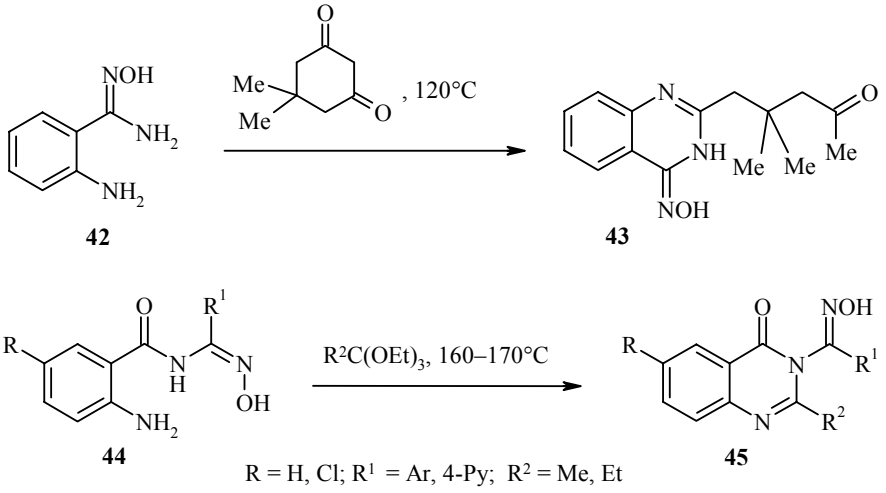


38 **39**



40 41

In the reaction of the 2-aminobenzamide oxime **42** and dimedone the quinazolone 4-oxime is formed with a yield of 23% [40]. The reactions of N-hydroxyiminobenzyl-2-aminobenzamides **44** with triethyl orthoacetate or triethyl orthopropionate at 160-170°C give 2-alkyl-3-hydroxyiminoaralkyl-4-quinazolones **45** with yields of 31-98% [41].

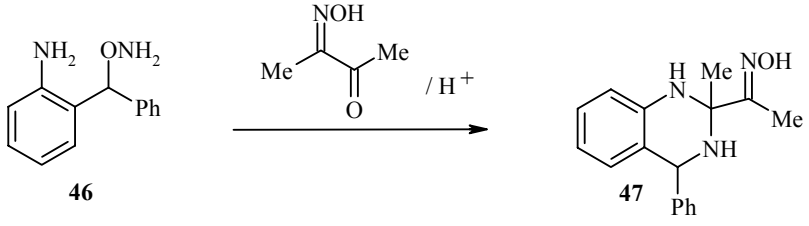


42 43

44 45

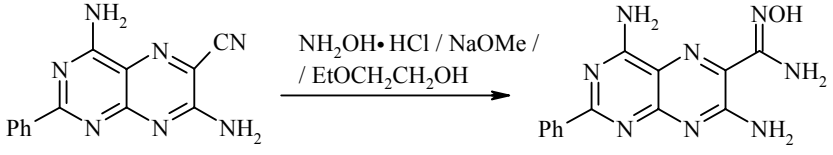
R = H, Cl; R^1 = Ar, 4-Py; R^2 = Me, Et

2-[1-(Hydroxyimino)ethyl]-2-methyl-4-phenyl-1,2,3,4-tetrahydroquinazoline **47** was obtained with a yield of up to 70% by the cyclization of the hydroxylamine **46** and 2,3-butanedione monoxime in the presence of *p*-toluenesulfonic acid or other acid catalysts [42].



46 47

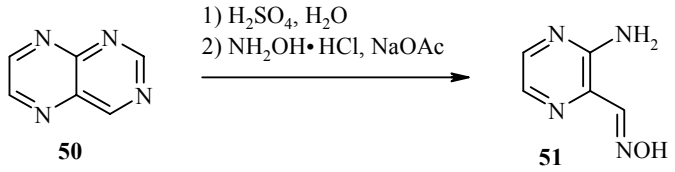
The classical method for the synthesis of pyrimidine amidoximes is based on the reaction of the nitrile with hydroxylamine hydrochloride in an aqueous solution of K_2CO_3 [43], Na_2CO_3/H_2O [44], ethanol [45], Et_3N/MeOH [46], or *i*-Pr_2NEt/DMF [47]. In the $NH_2OH{\cdot}HCl$/NaOMe/$EtOCH_2CH_2OH$ system 4,7-diamino-6-cyano-2-phenylpteridine **48** forms the oxime **49** [48].



48 49

1.3. Synthesis of Pyrazine Aldoximes, Ketoximes, and Amidoximes

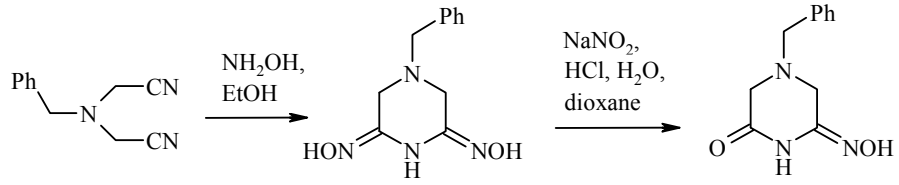
Pyrazine aldoximes and ketoximes are produced by the reaction of the aldehyde or ketone with hydroxylamine hydrochloride in the pyridine/EtOH [49] or $Et_3N/CHCl_3$ [50] systems. Boiling of pteridine **50** in sulfuric acid followed by treatment of the reaction mixture with hydroxylamine hydrochloride gives the oxime of 2-amino-3-formylpyrazine **51** with a yield of 85% [51].



50 **51**

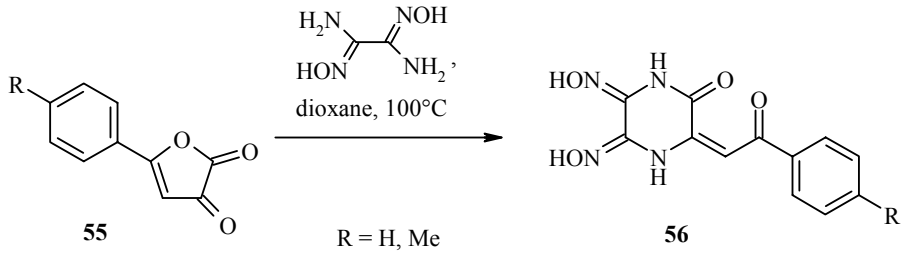
In addition, pyrazine-2-carbaldehyde can be obtained from 2-methylpyrazine in the $BuONO/NaNH_2/NH_3$ system [52].

The reaction of the dinitrile **52** with hydroxylamine in ethanol leads to the dioxime **53** with a 50% yield [53]; in the $NaNO_2/H_2O/HCl$ system this compound is converted into the product **54** from partial hydrolysis of one oxime group with a yield of 77% [53].



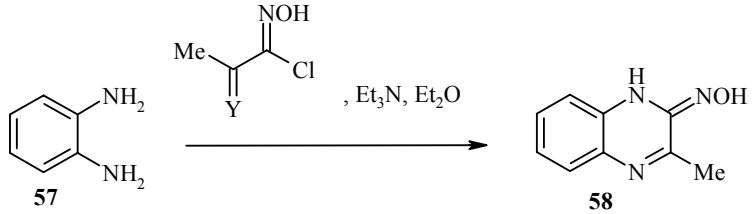
52 **53** **54**

The reaction of 5-aryl-2,3-dihydro-2,3-furandiones **55** with diaminoglyoxime in dry dioxane gives the 5,6-dioximes of 3-oxopiperazines **56** [54].



55 R = H, Me **56**

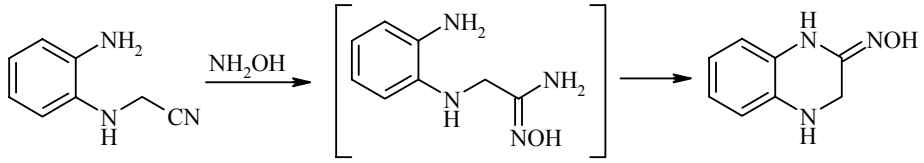
2-Hydroxyimino-1,2-dihydroquinoxalines were produced successfully by the cyclization of *o*-diaminobenzenes with halo oximes [55, 56]. For example, the reaction of diaminobenzene **57** and oxime derivatives of the MeC(=Y)C(=NOH)Cl type in ether in the presence of triethylamine gives the oximes **58** with yields of 52-90% [56].



57 **58**

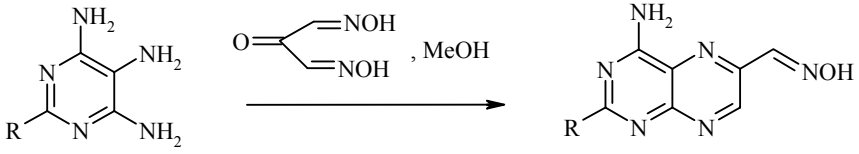
Y = O 90%; Y = NOH 90%; Y = NNHPh 52%

In a water–alcohol solution of sodium carbonate N-cyanomethyl-*o*-phenylenediamine **59** and hydroxylamine hydrochloride give 2-hydroxyimino-1,2,3,4-tetrahydroquinoxaline **60** with a yield of 78% [57].



59 **60**

The reactions of 4,5,6-triaminopyrimidine **61** with $O=C(CH=NOH)_2$ in methanol give pteridine oximes **62** with yields of 83-99% [58].



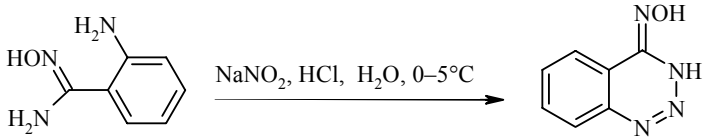
61 **62**

R = H, SMe, Ph

The classical method for the synthesis of pyrazine amidoximes is based on the reaction of the nitrile with hydroxylamine hydrochloride in the Na_2CO_3/EtOH [59], *i*-Pr_2NEt/DMF [47], and Et_3N/MeOH [46] systems. 2-Pyrazine amidoxime can also be obtained from pyrazine-2-carbothioamide and hydroxylamine [60].

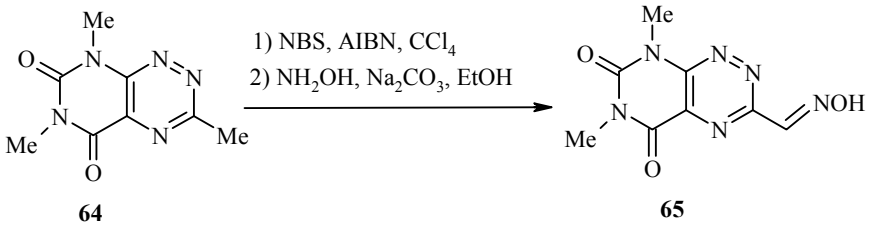
1.4. Synthesis of 1,2,3-, 1,2,4-, and 1,3,5-Triazine Aldoximes, Ketoximes, and Amidoximes

1,2,3-Triazine oxime **63** was obtained from *o*-aminobenzamide oxime **42** and $NaNO_2$/HCl/H_2O at 0-5°C [61].



42 **63**

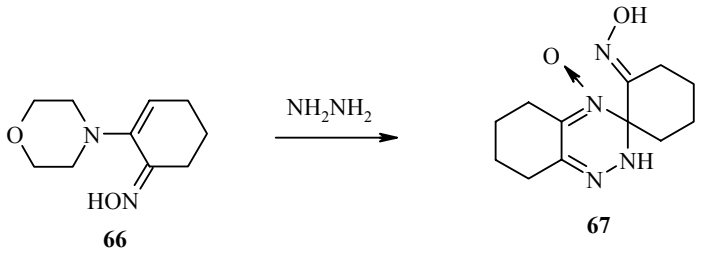
1,2,4-Triazine oximes can be obtained by several methods [62]: 1) Pyrimido[5,4-*e*][1,2,4]triazine aldoximes are produced successfully from the corresponding dibromomethyl derivatives in the $NH_2OH{\cdot}HCl/Na_2CO_3$/ethanol system [62]. 2) A two-stage reaction – bromination of 3-methylfervenulin **64** with NBS/AIBN/CCl_4 followed by treatment of the reaction mixture with hydroxylamine – leads to the formation of 6,8-dimethyl-5,7-dioxo-5,6,7,8-tetrahydropyrimido[5,4-*e*]triazine-3-carbaldehyde **65** with a yield of 62% [62].



64 **65**

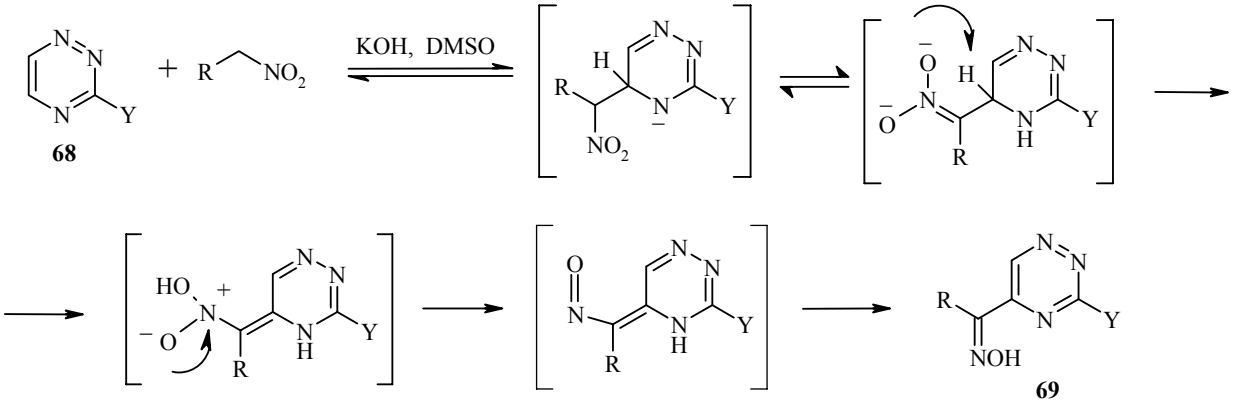
AIBN – azobisisobutyronitrile

If the enamine **66** is boiled in the presence of a large excess of hydrazine 2'-hydroxyiminospirodihydro-1,2,4-triazino[3,1]cyclohexane 4-oxide **67** is formed with a yield of 73% [63].



66 **67**

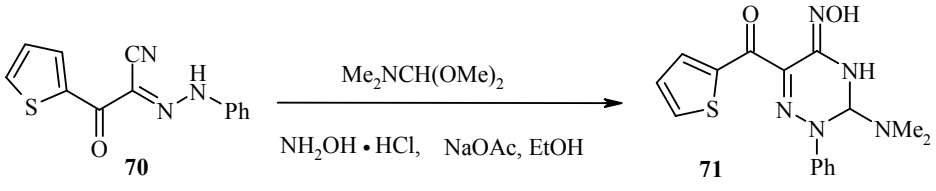
The 3-substituted 1,2,4-triazines **68** react readily with nitronate anions generated from nitroalkanes and KOH in DMSO and form aldoximes **69** with yields of 57-95% [64, 65].



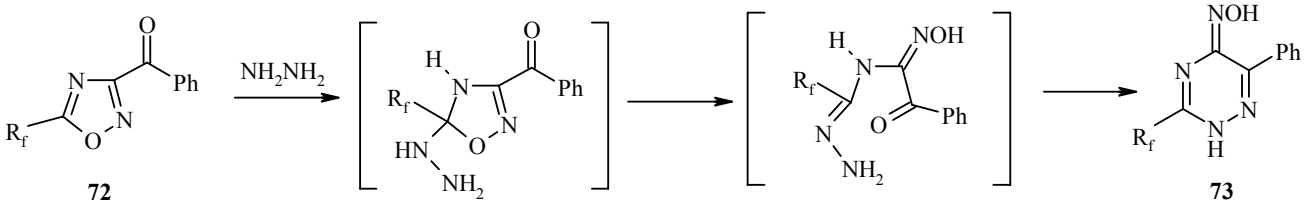
68 **69**

R = H, Me; Y = H, Me, Ph, SMe, OMe

With the dimethyl acetal of DMF in the $NH_2OH \cdot HCl/NaOAc/EtOH$ system the phenylhydrazononitrile **70** is readily converted into the triazine oxime **71** [66].



70 **71**

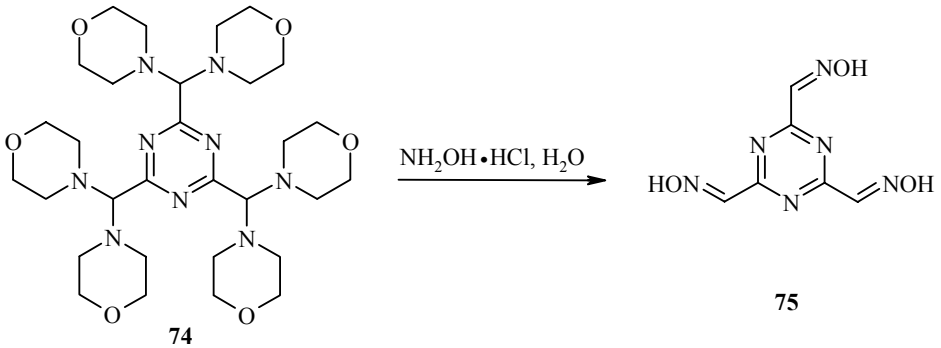


72 **73**

$R_f = CF_3, C_3F_7, C_7F_{15}$

Enlargement of the oxadiazole ring is observed during the reaction of 3-benzoyl-5-perfluoroalkyl-1,2,4-oxadiazoles **72** and hydrazine in dry DMF. Addition of the hydrazine at the C=N double bond occurs at the first stage, and opening of the oxadiazole ring and recyclization at the second. The triazole oximes **73** were isolated with yields of 65-92% [67].

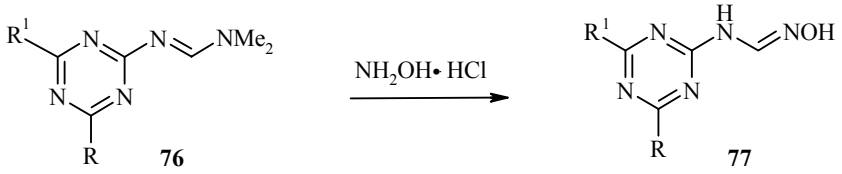
Several papers have been devoted to the synthesis of 1,3,5-triazine oximes. Thus, boiling of tris-dimorpholinomethyl-1,3,5-triazine **74** with hydroxylamine hydrochloride in water gives 1,3,5-triazine-2,4,6-tricarbaldoxime **75** with a yield of 79% [68]. 4,6-Diamino-1,3,5-triazine-2-carbaldoxime was obtained successfully from 6-bromomethyl-1,3,5-triazine-2,4-diamine and hydroxylamine [69].



74 **75**

1,3,5-Triazine aldoximes were also obtained as a result of nitrosation of the alkyl chain in the corresponding methyl derivatives in the EtONO/NaOMe/Et_2O/MeOH [70], *i*-AmONO/*t*-BuOK/*t*-BuOH [70], or EtONO/HCl [27] systems.

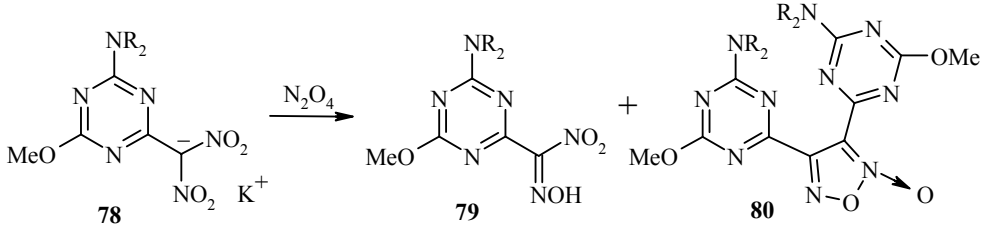
1,3,5-Triazine amidoximes were synthesized from the corresponding nitriles and hydroxylamine hydrochloride in a water–alcohol solution of sodium bicarbonate [71]. The amidoximes **77** were obtained from the imines **76** and hydroxylamine hydrochloride [72].



76 **77**

R, R^1 = H, OH, Me, NEt_2, SMe, morpholino

The reaction of the potassium salts of 6-dinitromethyl-1,3,5-triazines **78** with N_2O_4 leads to the formation of two products – the nitrolic acids **79** (yields 56-67%) and the 1,2,5-oxadiazoles **80** (24-32%) [73].

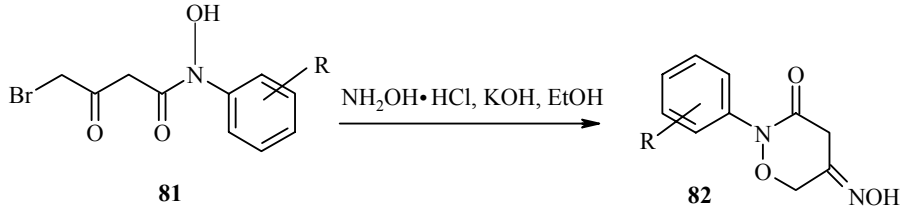


78 **79** **80**

R = Me; $R_2 = (CH_2CH_2)_2O$

1.5. Synthesis of 1,2-, 1,3-, and 1,4-Oxazine Aldoximes and Ketoximes

N-Aryl-N-hydroxy-4-bromo-3-oxobutyramides **81** in the $NH_2OH·HCl/KOH/EtOH$ system at room temperature gives the 1,2-oxazine oximes **82** with yields of 37-65% [74].

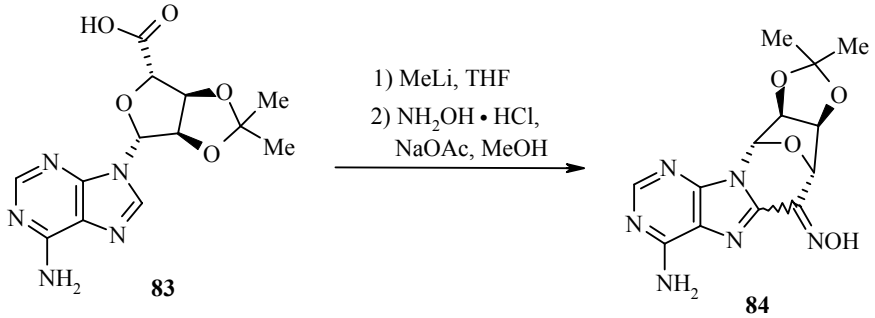


81 **82**

R = H, Hal, Alk

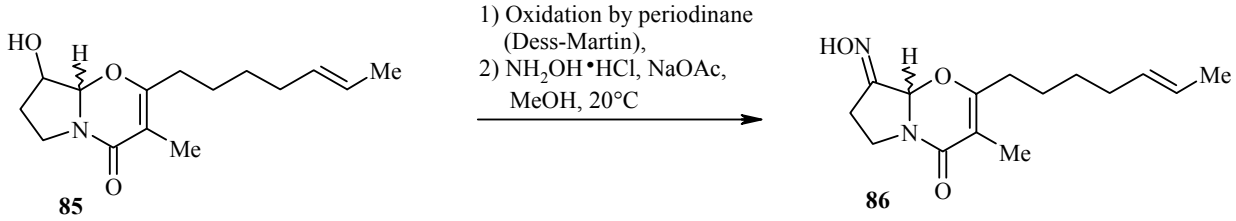
The classical method for the synthesis of 1,3-oxazine ketoximes is based on the reaction of the ketones in the $NH_2OH·HCl/NaHCO_3/MeOH/H_2O$ system [75].

A two-stage method for the synthesis of the oximes **84** is based on the cyclization of the ester **83** in the presence of MeLi followed by treatment of the reaction mixture with hydroxylamine hydrochloride in methanol [76].



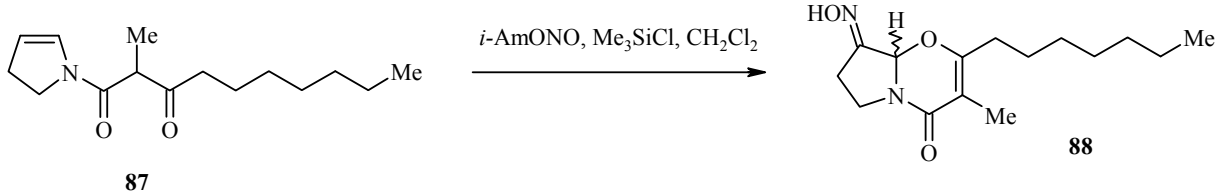
83 **84**

Brevioxime **86** [77], isolated in 1977 from *Penicillium brevicompactum*, was recently synthesized both in the racemic form [78-81] and in the form of pure optical isomers [82]. During the oxidation of the alcohol **85**, obtained in eleven stages from 1,5-pentanediol, followed by oximation ($NH_2OH·HCl/NaOAc/MeOH$) *rac*-brevioxime is formed with a 62% yield [78].



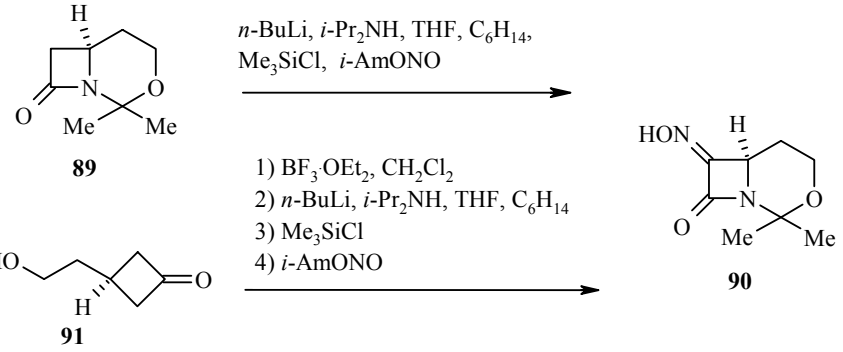
85 **86**

Brevioxime and its analogs (e.g., compound **88**) were synthesized by the cyclization of derivatives of 1-(2,3-dihydropyrrol-1-yl)-2-methyldecane-1,3-dione (e.g., compound **87**) in the presence of nitrosyl chloride, produced in the *i*-AmONO/Me_3SiCl/CH_2Cl_2 system [80].



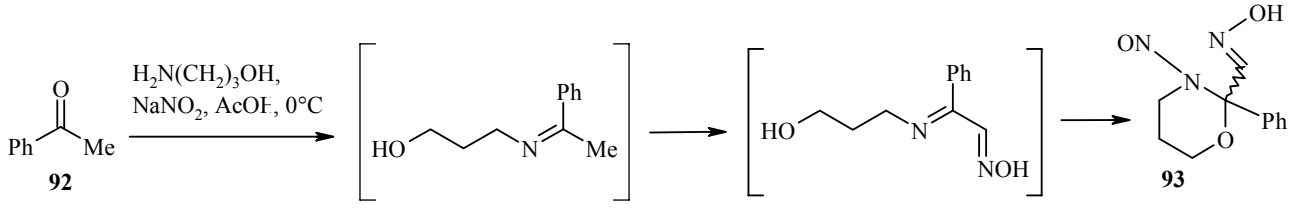
87 **88**

Several methods have been developed for the synthesis of the 7-oxime of 2,2-dimethyl-3-oxa-1-aza-bicyclo[4.2.0]octane-7,8-dione [83, 84]. Thus, the reaction of the amide **89** in the *n*-BuLi/*i*-Pr_2NH/*i*-AmONO/-Me_3SiCl/hexane/THF system gives the oxime **90** as the only product. The second method is based on the cyclization of the azetidinone **91** in a four-stage reaction. Thus, the reaction of compound **91** with boron trifluoride etherate, lithium diisopropylamide, and Me_3SiCl and then with isoamyl nitrite gives compound **90** [83].



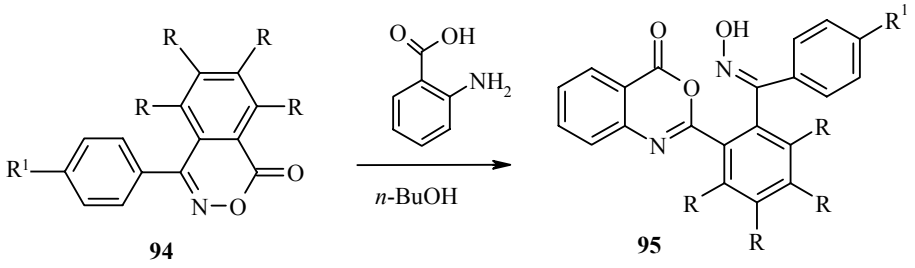
89 **90** **91**

The nitrosation of acetophenone **92** in dioxane in the presence of 3-hydroxypropylamine leads to the formation of 3-nitroso-2-phenyl-1,3-oxazine-2-carbaldehyde **93**. The product **93** is formed through the nitrosation of imine intermediates [84].



92 **93**

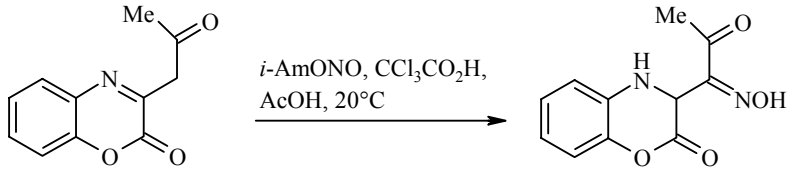
1,3-Oxazine oximes of the **95** type (yields up to 80%) were obtained successfully by thermal recyclization of substituted 4-arylbenzo[*d*][1,2]oxazin-1-ones **94** in the presence of anthranilic acid in *n*-butanol [85-87].



94 **95**

R = H, Br; R^1 = Br, Me

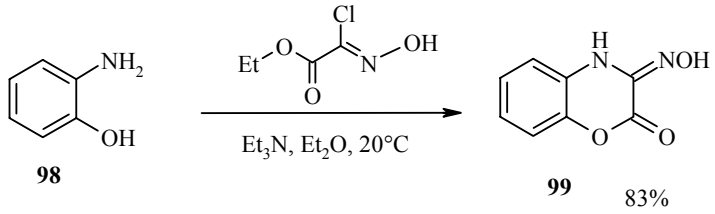
1,4-Oxazine aldoximes and ketoximes were obtained by the reaction of the aldehyde or ketone with hydroxylamine hydrochloride in ethanol [88] or in pyridine [89].



96 **97**

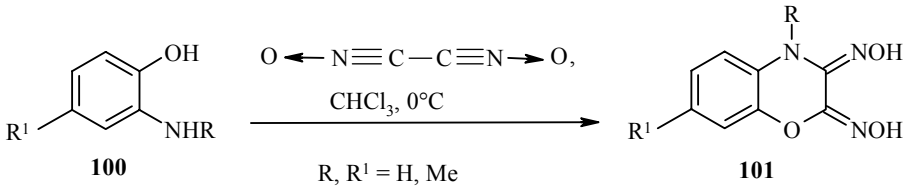
The 1-oxime of 1-(2-oxo-3,4-dihydro-2H-benzo[1,4]oxazin-3-yl)propane-1,2-dione **97** was obtained by nitrosation of the propanone **96** in the *i*-AmONO/CCl_3CO_2H/AcOH system [90].

The 3-oxime of 2H-[1,4]benzoxazine-2,3(4H)-dione **99** was obtained by the cyclization of *o*-aminophenol **98** in the presence of the *Z*-isomer of ethyl chloro(hydroxyimino)acetate and triethylamine in diethyl ether [91].



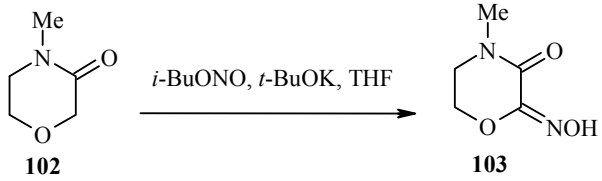
98 **99** 83%

The derivatives of *o*-aminophenol **100** and dicyanogen di-N-oxide in chloroform at 0°C give 1,4-benzoxazine-2,3-dioximes **101** [92].



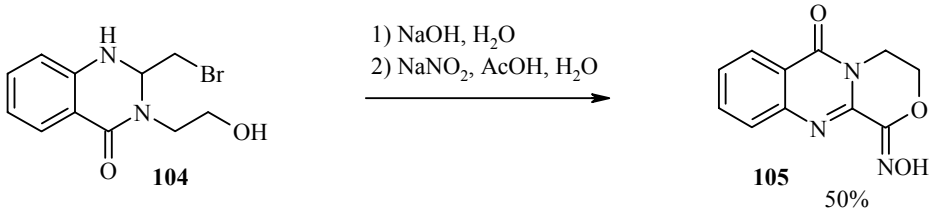
100 **101**

In the *i*-BuONO/*t*-BuOK/THF system 4-methyltetrahydro-1,4-oxazin-3-one **102** is nitrosated selectively at position 2 and forms the oxime **103** [93].



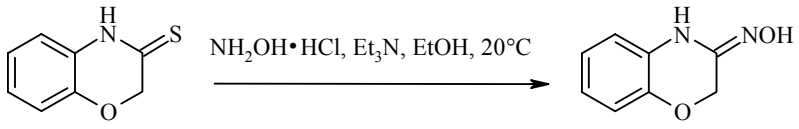
102 **103**

3,4-Dihydro[1,4]oxazino[3,4-*b*]quinazolones are readily nitrosated at position 1 in the $NaNO_2$/AcOH/H_2O system [94]. In addition a new two-stage method was developed for the synthesis of 1-hydroxyimino-3,4-dihydro-1H,6H-[1,4]oxazino[3,4-*b*]quinazolin-4-one **105** from 2-bromomethyl-3-(2-hydroxyethyl)quinazolin-4-one **104** in the presence of aqueous alkali followed by oximation in the $NaNO_2$/AcOH/H_2O system [94].



104 **105** 50%

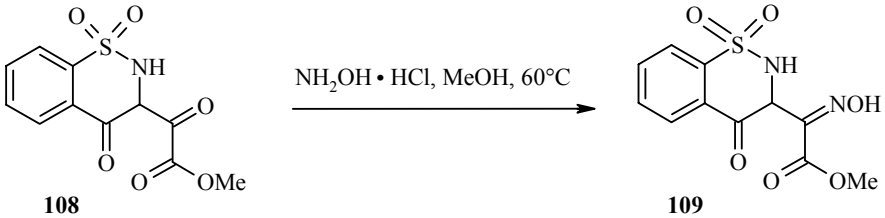
The classical method for the synthesis of 1,4-oxazine amidoximes is based on the reaction of oxazine nitriles in the NH_2OH/MeOH [95], $NH_2OH{\cdot}HCl/Et_3N$/EtOH [96], or $NH_2OH{\cdot}HCl/NaHCO_3$/EtOH/H_2O [97] systems. In the $NH_2OH{\cdot}HCl/Et_3N$/EtOH system at room temperature 4H-benzo[1,4]oxazine-3-thione **106** gives the oxime **107** with a yield of 72% [98].



106 **107**

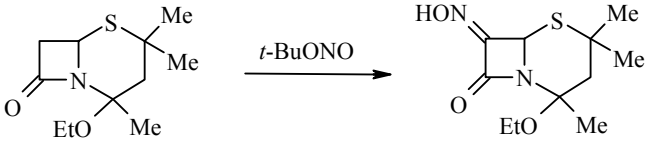
1.6. Synthesis of 1,2-, 1,3-, and 1,4-Thiazine Aldoximes, Ketoximes, and Amidoximes

The basic method for the synthesis of 1,2-thiazine ketoximes is based on the reaction of the ketone with hydroxylamine hydrochloride in the presence of sodium acetate in methanol [99-101]. It is interesting that in the presence of an equimolar amount of hydroxylamine hydrochloride only the carbonyl group of the side chain in the cyclic thiazine **108** is oximated. The reaction product **109** was obtained with a yield of 81% [101].



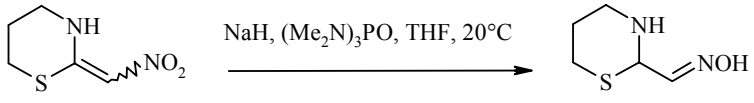
108 **109**

1,3-Thiazine oximes are usually obtained from the thiocarbonyl derivatives of 1,3-thiazines in the $NH_2OH{\cdot}HCl/Et_3N/EtOH/H_2O$ [102], $NH_2OH/MeOH$ [103], and $NH_2OH/EtOH$ systems [104]. Trimethylcephem **110** is readily nitrosated by *tert*-butyl nitrite with the formation of the oxime **111** [105]. In addition, N_2O_3 in methylene chloride has also been used in the selective nitrosation of cephems to oximes [106].



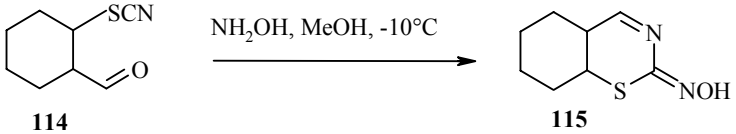
110 **111**

The nitrovinyl derivative of 1,3-thiazine **112** is easily transformed into 5,6-dihydro-4H-1,3-thiazine-2-carbaldehyde oxime **113** in the $NaH/(Me_2N)_3PO/THF$ system [107].



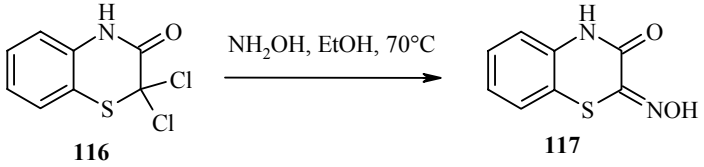
112 **113**

The cyclization of 2-thiocyanato-1-cyclohexanecarbaldehyde **114** in the presence of a solution of hydroxylamine in methanol at -10°C gives a derivative of 2-hydroxyimino-1,3-thiazine **115** [108].

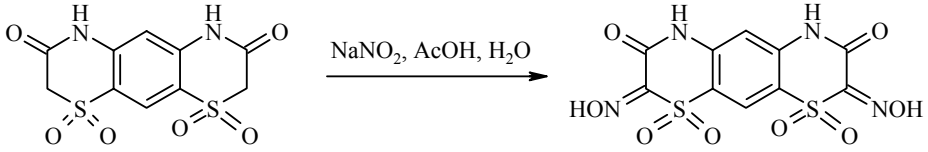


114 **115**

2-Hydroxyimino-1,4-benzothiazin-3(H)-one **117** was obtained from the corresponding dichloro derivative **116** and hydroxylamine in ethanol [109]. Nitrosation was also used successfully for the production of 1,4-benzothiazine S-dioxides. For example, the reaction of compound **118** with an excess of sodium nitrite in aqueous acetic acid gives the dioxime **119** as the only product [110].

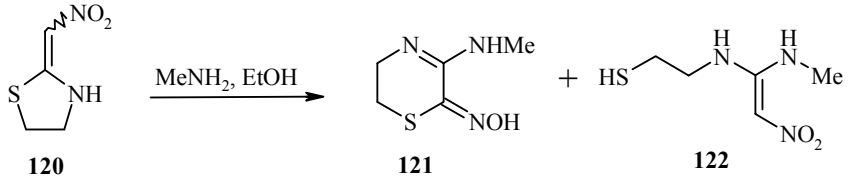


116 **117**



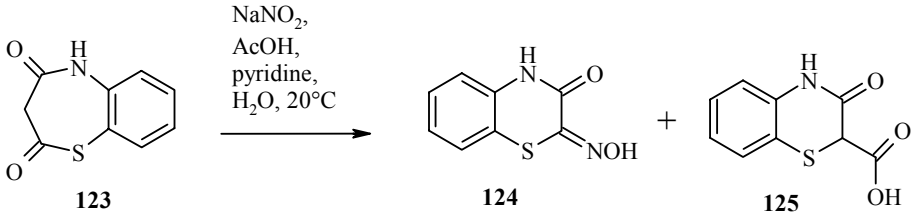
118 **119**

Ring enlargement in 2-(nitromethylene)thiazolidine **120** by the action of methylamine leads to the formation of the oxime **121** and 2-{[1-(methylamino)-2-nitroethenyl]amino}ethanethiol **122**. Increase of the amount of methylamine in the reaction mixture increases the yield of compound **122** [111].



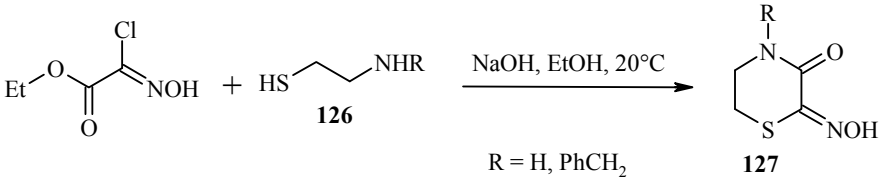
120 **121** **122**

Under the condition of nitrosation ($NaNO_2$/AcOH/pyridine/H_2O) 1,5-benzothiazepine-2,4-dione **123** is converted into 2-hydroxyimino-1,4-benzothiazin-3(4H)-one **124** (yield 17%) and 2H-1,4-benzothiazine-3,4-dihydro-3-oxo-2-carboxylic acid **125** (45%) [112].



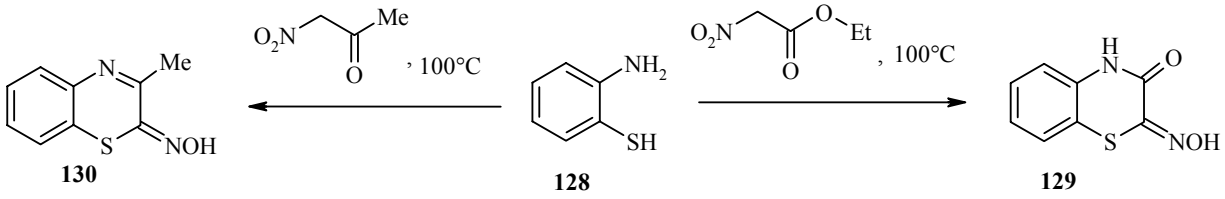
123 **124** **125**

The synthesis of 1,4-thiazine oximes was realized by the cyclization of derivatives of 2-aminoethanethiols or 2-aminothiophenol with ethyl 2-chloro-2-(hydroxyimino)acetate [113-115], ethyl nitroacetate [116], or nitroacetone [117]. Thus, 2-aminoethanethiol **126** and ethyl chloro(hydroxyimino) acetate in an alkaline medium give 2-hydroxyiminotetrahydro-1,4-thiazin-3-ones **127** with yields of up to 83%. Heating of



126 **127**

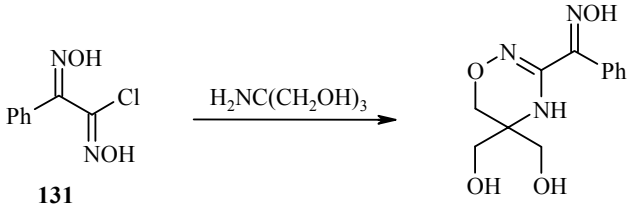
2-aminothiophenol **128** with ethyl nitroacetate or nitroacetone at 100°C without a solvent leads to the products **129** and **130** respectively with yields of up to 91%.



130 **128** **129**

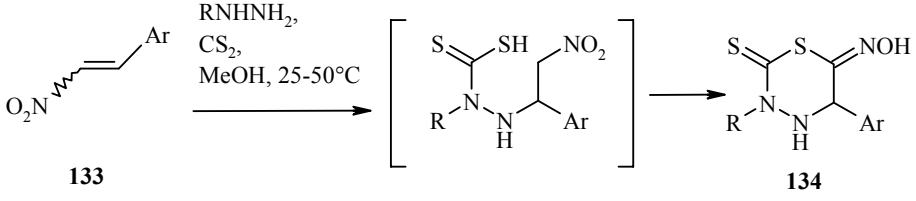
1.7. Synthesis of Oxadiazine and Thiadiazine Oximes

The reaction of the dioxime **131** with tris(hydroxymethyl)aminomethane gives the 1,2,4-oxadiazine oxime **132** as the only product [118].



131 **132**

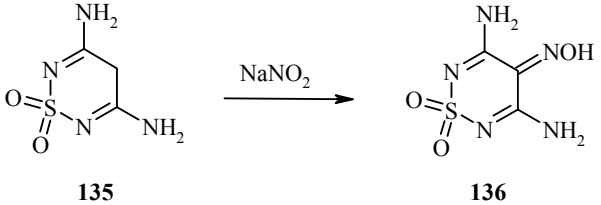
6-Hydroxyiminotetrahydro-1,3,4-thiadiazine-2-thiones **134** were obtained from aliphatic hydrazines, carbon disulfide, and ω-nitrostyrenes **133** [119, 120].



133 **134**

R = Alk, CH_2CH_2OH, CH_2CH_2CN

The respective oximes were also obtained by the nitrosation of 3,5-disubstituted 1,2,6-thiadiazine 1,1-dioxides [121-123]. For example, the nitrosation of 3,5-diamino-1,2,6-thiadiazine 1,1-dioxide **135** with sodium nitrite in acetic acid leads to the oxime **136** [121].



135 **136**

2. STRUCTURE

One of the most reliable methods for the determination of the structure of the isomeric oximes of six-membered heterocyclic compounds with two and three heteroatoms is NMR spectroscopy. The ^{1}H NMR spectra of the oximes of pyridazine [124-126], pyrimidine [11, 126-129], pyrazine [124], quinoxaline [130], 1,3,4-triazine [68], and 1,3,4-thiadiazine [120] have been investigated in greatest detail.

The structure of the *Z*- [131] and *E*- [132] isomers of the oximes of 4-pyrimidinecarbaldehyde, 1-benzyl- 2-phenyl-1H-quinazolone [39], and the solvate of 3-(1-hydroxyiminoethyl)-1-phenyl-4-hydro-1,2,4-triazine-5,6-dione with methanol [133], the *E*-isomer of 1-(3-methylsulfanyl-1,2,4-triazin-5-yl)ethanone O-acryloyl-oxime [134], and the palladium complex of 3-acetyl-5-benzyl-1-phenyl-4,5-dihydro-1,2,4-triazin-6-one oxime [135, 136] was confirmed by the data from X-ray crystallographic analysis.

IR spectroscopy was also used to study the structure of pyridazine [124], pyrimidine [11], quinazoline [127], quinoxaline [130], and 1,2,4-triazole [67] oximes. In the IR spectrum of 2-pyrimidine aldoxime there is a band for the free OH group at 3600 cm^{-1}. In the spectrum of 3,5-dimethylpyrimidine-2-aldoxime there is also a strong band for the nonbonded hydroxyl group at 3600 cm^{-1}, and there is in addition a band at 3300 cm^{-1} due to the strong intermolecular hydrogen bond, since the band completely only disappears in a very dilute solution ($4\cdot10^{-5}$ M). The above-mentioned oximes were therefore isolated as *Z*-isomers. During irradiation of the *Z*-isomer of 2,5-dimethylpyrimidine-2-aldoxime with ultraviolet light its *E*-isomer was obtained [11].

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