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Acylation of Hydrazine and Its Substituted Derivatives with Derivatives of αβ-unsaturated Monobasic Acids

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The literature on the formation of hydrazides of αβ-unsaturated acids and also five-membered heterocyclic compounds in the interaction of hydrazine and its mono- and di-alkyl-substituted hydrazines with derivatives of saturated monobasic acids is reviewed. The properties and applications of these compounds are described.
The bibliography includes 97 references.

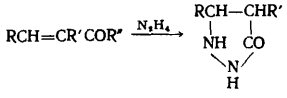
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1. ACYLATION OF HYDRAZINE AND MONOSUBSTITUTED HYDRAZINES WITH αβ-UNSATURATED ACIDS AND THEIR DERIVATIVES

Alkylhydrazines $RNHNH_2$ are known to be acylated at the NH_2 or NH group depending on the reagent used and the reaction conditions[1]. As a rule, when mild acylating agents (for example esters) are used and the system is cooled, monoacyl derivatives substituted at the NH_2 groups are formed[2-6]. Under more severe conditions, products of acylation at the NH group are obtained[3]. When monoalkylhydrazines are acylated with acid anhydrides[7,8], the products are acyl derivatives substituted at the NH group, while reaction with acid chlorides or heating with an excess of the anhydride yield *NN'*-diacylalkylhydrazines[9-11].

On the other hand, the interaction of hydrazine with derivatives of αβ-unsaturated acids usually leads to pyrazolidinones (I):[12]

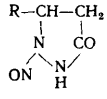


For this reason, the monomeric hydrazides of the majority of αβ-unsaturated acids have not been obtained until quite recently[13,14] and there are numerous contradictory literature data concerning the formation in this reaction of pyrazolidinones or hydrazides. For example, it has been stated[15] that phenylhydrazides of α-methyl and α-butylacrylic acids have been synthesised from their chlorides and phenylhydrazine. On the other hand, when the acids themselves were heated with phenylhydrazine, the products were 4-alkyl(ethyl or butyl)-2-phenyl-3-pyrazolidinones.

In the attempt to synthesise the hydrazides of other αβ-unsaturated acids pyrazolidinone derivatives were obtained†. Thus in the reaction of hydrazine hydrate with acrylic acid unsubstituted 3-pyrazolidinone is formed[15,17] while in the reaction of phenylhydrazine the product is 2-phenyl-3-pyrazolidinone[15,17,19]. Phenylhydrazine reacts with crotonic acid to form 5-methyl-2-phenyl-3-pyrazolidinone[18,20], while the reaction with ββ-dimethylacrylic acid gives 5,5-dimethyl-2-phenyl-3-pyrazolidinone[21,22].

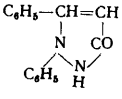
In 1963 Shvaika and Makarenko[23] described the preparation of the hydrazide of methacrylic acid by heating methyl methacrylate with hydrazine hydrate. However, it was found that in reality this results in the formation of 4-methyl-3-pyrazolidinone[24-28]. The infrared spectrum of this compound does not contain an absorption band corresponding to C=C stretching vibrations. Under the action of nitrous acid, the compound is converted[24,26] into 4-methyl-1-nitroso-3-pyrazolidinone and not into the azide of methacrylic acid. The formation of a coloured complex by the acyl derivative of this compound and a trivalent iron ion, recommended as a test for the hydrazide of methacrylic acid[23,29], is also a qualitative test for 3-pyrazolidinones[17,30]. The preparation of the hydrazide of acrylic acid by the reaction of methyl acrylate with anhydrous hydrazine has been reported[31]. However, on treatment with hydrochloric acid this substance too is converted into the hydrochloride of 3-pyrazolidinone, which suggests that it is not a hydrazide but a cyclic isomer.

Muckermann[32] described the preparation of hydrazides by heating crotonic and cinnamic acids with hydrazine hydrate. The products formed hydrochlorides and their reaction with benzaldehyde gave crystalline compounds to which the author[32] attributed to the structure of hydrazones. However, the reaction with nitrous acid gave *N*-nitrosopyrazolidinones (II) instead of the expected azides, which was accounted for by the cyclisation of the unsaturated hydrazides on treatment with nitrous acid[29,32-35]:



(II)

1,5-Diphenyl-Δ^4-3-pyrazolone (III) was obtained by heating cinnamic acid with phenylhydrazine[36]. The author suggested that the phenylhydrazide is formed initially and then cyclises and is oxidised by atmospheric oxygen:

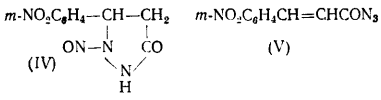


(III)

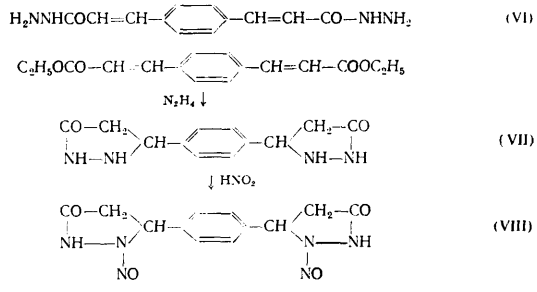
Two compounds were obtained on treating ethyl *m*-nitrocinnamite with hydrazine hydrate[37,38]. Both substances interacted with aldehydes and ketones and on this basis the author regarded them as different forms of hydrazides. However, one of these compounds, formed as the main product, is converted under the action of nitrous acid[38] into *N*-nitrosopyrazolidinone (IV) and the other (the side

†In the presence of oxygen, hydrazine can reduce the double bond of unsaturated acids[16].

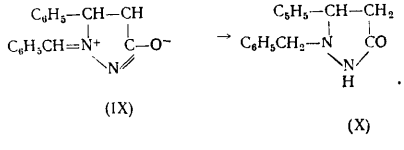
product) is converted into the azide (V), i.e. it is the acid hydrazide:



By allowing diethyl *p*-phenylenediacrylate to react with hydrazine hydrate in alcohol, Ruggli and Theilheimer [39] obtained a substance which was erroneously regarded as the dihydrazide (VI), although the structure of the bis-pyrazolidinone derivative (VII) was also considered for it. On treatment with nitrous acid, the substance is converted into the *N*-nitroso-derivative (VIII):

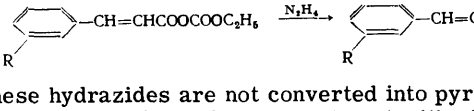


The formation of nitroso-derivatives under the action of nitrous acid demonstrates that the products of the reaction of $\alpha\beta$-unsaturated acids with hydrazine hydrate have the structure of pyrazolidinones [25-27,31]. However, a serious objection against this structure is the capacity of these compounds to condense with aldehydes and ketones. This contradiction was resolved by Godfredsen and Vangedal [40], who proposed the betaine structure (IX) for the product of the condensation of 5-phenyl-3-pyrazolidinone with benzaldehyde. On catalytic hydrogenation of this compound, one mole of hydrogen is absorbed and 1-benzyl-5-phenyl-3-pyrazolidinone (X) is obtained [40], which confirms the structure of compound (IX):



Later products of the interaction of 5-phenyl-3-pyrazolidinone with other aldehydes were also obtained [28,41].

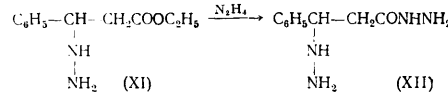
The hydrazides of cinnamic, *m*-nitrocinnamic, and *p*-nitrocinnamic acids have been obtained by the action of hydrazine hydrate on the mixed anhydrides of the corresponding acids with a carbonic ester:



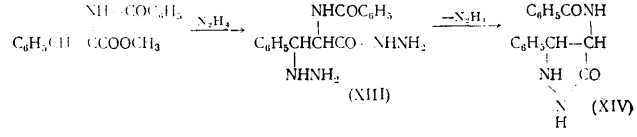
These hydrazides are not converted into pyrazolidinones on heating and yield azides on treatment with nitrous acid. It has been suggested [40] that the first stage in the formation of pyrazolidinone is the addition of hydrazine to the double carbon–carbon bond, which is followed by cyclisation to 3-pyrazolidinone.

The addition of monosubstituted hydrazine involves either the free or the alkylated amino-group of the hydrazine depending on the reaction conditions which results in the formation of 3-pyrazolidinone with substituents at the N_1 [24,31,40-43] or N_2 [15,18-22] nitrogen atoms.

The cyclisation is fairly slow and the hydrazine-ester formed initially, for example (XI), yields the hydrazide (XII) in the presence of an excess of hydrazine:



In the attempt to synthesise the hydrazides of α-benzoylaminocinnamic acid by the reaction of the methyl ester of this acid with an excess of hydrazine hydrate, Stodola [44] obtained the hydrazide (XIII) which on heating *in vacuo* was converted into 4-benzamido-5-phenyl-3-pyrazolidinone (XIV) with elimination of hydrazine. The benzamido-group was unaffected:



Hydrazides of unsaturated β-hydrazino-acids have also been obtained by treating esters of cinnamic and *m*- and *p*-nitrocinnamic acids with an excess of hydrazine hydrate [11,40,45].

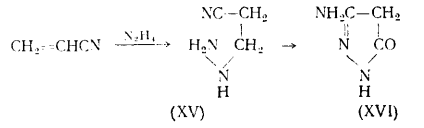
In a series of studies Deles and Polackowa [46-49] investigated the effect of the substituent on the rate of reaction and the yield of the product of the interaction of hydrazine with esters of ring-substituted cinnamic acids. When esters of cinnamic, *m*- and *p*-bromocinnamic, *m*- and *p*-nitrocinnamic, and *p*-methoxycinnamic acids were treated with hydrazine hydrate, a mixture of the β-hydrazino-ester, the hydrazides of the unsaturated and β-hydrazinodihydrocinnamic acids, and the corresponding pyrazolidinone was obtained. The authors established that the pyrazolidinones are formed by the cyclisation of the hydrazino-esters, but in the presence of an excess of hydrazine hydrate they are in turn capable of being converted with ring opening into the hydrazides of the β-hydrazino-acids.

Thus the first step in the formation of pyrazolidinone is the addition of hydrazine to the double bond of the $\alpha\beta$-unsaturated acid. Probably pyrazolone derivatives are formed similarly from esters of $\alpha\beta$-acetylenic acids [29,50,51]. This mechanism appears to be very probable [52], since the double bond in derivatives of $\alpha\beta$-unsaturated acids readily combines with nucleophilic reagents, including hydrazines [7,15-20,42,43,50-64].

The main chemical evidence in the determination of the structure of the products of these acylation reactions comes from their interaction with nitrous acid. Pyrazolidinones then yield *N*-nitroso-derivatives and hydrazides give the azides. In the hydrogenation of hydrazides over metallic catalysts one mole of hydrogen is absorbed with formation of saturated acids, while pyrazolidinones are not hydrogenated.

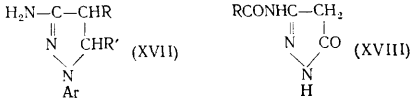
The most thoroughly investigated addition to an activated double bond is the interaction of nucleophilic agents (including hydrazines) with acrylonitrile. Several reviews have been published on the cyanoethylation reaction [52,54-57]. Hydrazine readily reacts with acrylonitrile yielding mono-, di-, tri-, and tetra-cyanoethylation products depending on the reaction conditions [7,57]. In the

presence of basic catalysts the β-cyanoethylhydrazine (XV) obtained cyclises to 3-aminopyrazoline (XVI):[7]



(XV) (XVI)

Substituted aminopyrazolines (XVII) were obtained by the cyanoethylation of phenylhydrazine in an alkaline medium and also by the reaction of phenylhydrazine with the nitriles of methacrylic, crotonic, or cinnamic acids[60,61]:



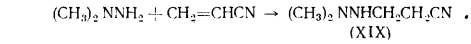
(XVII) (XVIII)

Acylhydrazines are also cyanoethylated at the amino-group both without catalysts and in the presence of acids[62,63]. In an alkaline medium the imino-group is probably cyanoethylated with subsequent cyclisation to the acylaminopyrazoline (XVIII). Monoalkylhydrazines react with the nitriles of αβ-unsaturated acids, forming derivatives substituted at the NH_2 or NH group[7,63] depending on the reaction conditions.

The interaction of derivatives of αβ-unsaturated acids with hydrazine and substituted hydrazines is nowadays widely used for the synthesis of pyrazolidinones[26-28,43,64-69]. It is also used for the identification of the products obtained in studies on other reactions.

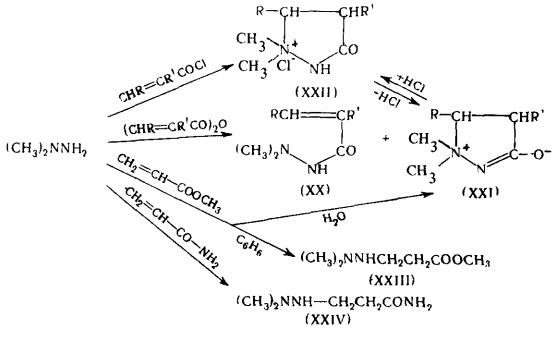
2. INTERACTION OF αβ-UNSATURATED ACID DERIVATIVES WITH ASYMMETRICALLY DISUBSTITUTED HYDRAZINES

Derivatives of αβ-unsaturated acids react with asymmetrically disubstituted hydrazines in different ways depending on the reagent and the reaction conditions. For example, dimethylhydrazine adds to the double bond of acrylonitrile[59] with formation of a monocyanoethylation product (XIX):



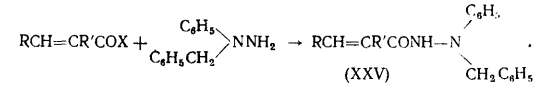
(XIX)

The action of the chlorides, anhydrides, and esters of αβ-unsaturated acids on asymmetric dimethyl- and benzylphenyl-hydrazines was investigated by Sokolova and co-workers[25,26,70-76]. The reactions of dimethylhydrazine with derivatives of αβ-unsaturated acids are represented by the following general scheme[26]:



where (a) R = R′ = H, (b) R = H and R′ = CH_3, and (c) R = CH_3 and R′ = H.

The products of the acylation of dimethylhydrazine with the anhydrides of acrylic, methacrylic, and crotonic acids are the dimethylhydrazides of these acids (XX) together with isomeric compounds (XXI) which the authors call 1,1-dimethylpyrazolinium 3-oxides[25,26,70,71]. When the chlorides of αβ-unsaturated acids are used, the salts (XXII), i.e. the hydrochlorides (XXII) of compounds (XXI), are formed[26,71]. The interaction of aqueous solutions of the salts (XXII) with anion-exchange resins yields the bases (XXI). The mode of the reactions of the esters of the same acids with dimethylhydrazine and the yields depend on the structure of the ester and the nature of the solvent. In polar solvents (water, alcohol) 1,1-dimethylpyrazolinium 3-oxide (XXI, a) is formed in almost quantitative yield from methyl acrylate and dimethylhydrazine, while the reaction with methyl methacrylate under the same conditions results in the formation of compound (XXI, b) in a much lower yield, which agrees with the lower activity of the C=C bond in the methacrylic acid ester[26,73]. In a non-polar medium or without a solvent dimethylhydrazine reacts with methyl acrylate, the product of addition to the double bond (XXIII) being formed. When an even weaker acylating agent (acrylamide) is used, the reaction with dimethylhydrazine yields only the product of addition to the double carbon–carbon bond (XXIV) regardless of the polarity of the medium[26]. On the other hand, in the reaction of the chlorides and anhydrides of αβ-unsaturated acids with asymmetric benzylphenylhydrazine cyclic compounds were not obtained. High yields of the benzylphenylhydrazides of the corresponding acids (XXV) were produced[26,72]:

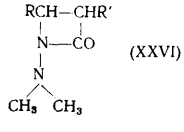


(XXV)

It is suggested[26,71,72] that cyclic compounds are not formed due to the screening effect of the bulky substituents at the nitrogen atom and the lower basicity of benzylphenylhydrazine compared with dimethylhydrazine, which hinders the nucleophilic addition of hydrazine to the double carbon–carbon bond[26].

The Structure and Properties of Hydrazine Alkylation Products

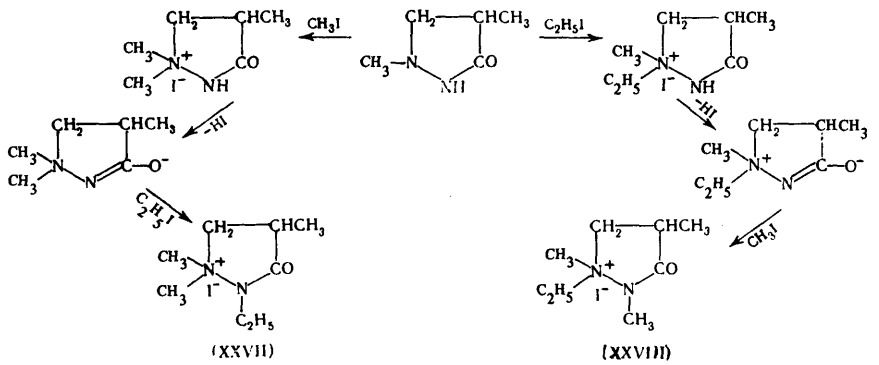
The interaction of dimethylhydrazine with derivatives of αβ-unsaturated acids is a method for the synthesis of the asymmetrically disubstituted hydrazides of these acids and also of a new class of heterocyclic compounds with a dipolar structure. On the basis of PMR spectra and studies of the hydrogenation products, the structure of *N*-dimethylamino-2-azetidinones (XXVI) was attributed initially to the cyclic isomers of the dimethylhydrazides[74,76]:



(XXVI)

However, it was subsequently shown by synthesis of pyrazolidinones of known structure[28] and also by the synthesis of various alkiodides and the study of their PMR spectra[26,74,75] that these compounds have the structure of compound (XXI).

Isomeric alkiodides (XXVII) and (XXVIII), which differ in the disposition of the methyl and ethyl groups at the nitrogen atom, were obtained by altering the order in which the initial 1,4-dimethyl-3-pyrazolidinone was methylated and ethylated [25,26]:



(XXVII) (XXVIII)

The crystalline alkiodides (XXVII) and (XXVIII), which have the same composition, exhibit different properties. With the β-lactam structure (XXVI) the same substance having all three alkyl groups at one nitrogen atom should be obtained regardless of the order of methylation and ethylation. However, the study of the PMR spectra of the alkiodides obtained showed [75] that two alkyl groups are located at one nitrogen atom while the third is at the second nitrogen atom.

The infrared spectra of compounds (XXI) show intense absorption bands in the region of 1580 cm^{-1}, which the authors assigned to absorption by the oxide group $-N=C-O^-$. [25,26,71] Such bands are also observed in the spectra of dipolar compounds with the linear structure (XXIX)‡ containing the group $N=C-O^-$: [77-80]

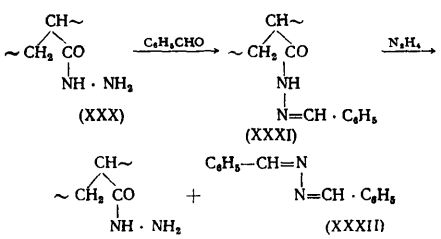
$$RCO\bar{N}-\overset{+}{N}(CH_3)_3 \leftrightarrow R\overset{O^-}{\overset{|}{C}}=N-\overset{+}{N}(CH_3)_3 \quad (XXIX)$$

In 1966 Wadsworth [65] synthesised several derivatives of pyrazolinium 3-oxides and called them, by analogy with Hinmann's compounds [77], cyclic amine-imides [65]. The pyrolysis of these cyclic compounds gave good yields of the dimethylhydrazides of methacrylic and crotonic acids [65]. Sokolova et al. [80] obtained by this procedure from 1,1-dimethylpyrazolinium 3-oxides the dimethylhydrazide of acrylic acid (XX, a), which is difficult to synthesise. Dimethylhydrazides of αβ-unsaturated acids are bases, like pyrazolinium 3-oxides, capable of combining with hydrogen halides and alkyl iodides [25,26,71]. The interaction of the methiodides of the dimethylhydrazides with bases (anion exchangers) results in the formation of the trimethylamine-imides of the corresponding unsaturated acids (XXIX). [78,80] The dimethylhydrazides of acrylic and methacrylic acids, their hydrochlorides, methiodides, and the trimethylamine-imides obtained from them are capable of polymerising in the presence of free-radical initiators with formation of high-molecular polyelectrolytes [81].

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The interaction of arylhydrazines with derivatives of αβ-unsaturated acids [42,43,62,66-69,82-86] is one of the methods of synthesising "phenidones", i.e. 1-aryl-3-pyrazolidinones, which are important in photographic and cinematographic practice. They exhibit the properties of "superadditivity", increasing sharply the activity of hydroquinones in developing solutions [87-94]. 1-Phenyl-3-pyrazolidinone is used as a reducing agent in the determination of phosphorus in proteins [95].

Since the acylation of hydrazine is accompanied by cyclisation to pyrazolidinones, the preparation of unsaturated acid hydrazides is difficult and therefore Kern and coworkers [13,14] employed polymer-analogue reactions to prepare the polyhydrazides of acrylic and methacrylic acids. For this purpose, polyacrylic and polymethacrylic acid esters were treated with hydrazine hydrate. The polymeric hydrazides obtained in this way (XXX) are converted by treatment with aldehydes into polymeric hydrazones (XXXI) and can be used as "aldehyde-exchange" resins [14]. After the decomposition of the polymeric hydrazone (XXXI) with dilute sulphuric acid, the aldehyde may be re-isolated by steam distillation and the polymeric hydrazide is at the same time hydrolysed to polyacrylic acid. The polymeric hydrazone may be decomposed with hydrazine hydrate. Then the polymer is re-isolated as the hydrazide (XXX) and the aldehyde as the aldazine (XXXII): [14]



Polymeric hydrazones obtained by the interaction of polymeric acrylic hydrazide with aromatic hydroxyaldehydes may couple with diazo-compounds, forming polymeric azo-dyes with colours ranging from yellow to brown and brownish-black [14].

Polyhydrazides obtained by treating with hydrazine the copolymer of styrene and methyl acrylate have been proposed for the investigation of chain reactions and for use as polymeric soaps [29,96].

Polymeric hydrazides of methacrylic acid have been proposed as metal extractants. They form chelate complexes with nickel, cadmium, zinc, copper, and cobalt ions [97].

‡Hinmann [77] called such compounds trimethylamine-imides. This name cannot be regarded as sufficiently exact. Possibly these substances could be called *NNN*-trimethylhydrazinium *N'*-alkylidene-α-oxides.

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