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Cycloaddition of nitrile oxides to multiple bonds containing a heteroatom

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Abstract. The review presents a compilation and a systematic account of the reactions involving the cycloaddition of nitrile oxides to multiple bonds containing noncarbon atoms. An attempt is made to trace the rules governing these processes on the basis of the available experimental data. The bibliography includes 219 references.

I. Introduction

The oxides of carboxylic acid nitriles occupy and important place among the 1,3-dipoles used widely in organic synthesis. These readily available compounds interact fairly easily with various dipolarophiles. They are therefore frequently used to obtain heterocyclic products and also in the synthesis of biologically active compounds with definite stereochemical parameters and of analogues of natural products. Nitrile oxides have recently found applications as modifying additives to polymeric materials. Their use as model compounds in theoretical studies on 1,3-cycloaddition reactions is no less important.

A monograph[1] and a series of reviews[2–5] on the chemistry of nitrile oxides are available at the present time. They cover the literature up to 1989. Attention is concentrated in these publications on the most thoroughly investigated reactions involving the cycloaddition of the dipoles to C=C and C≡C bonds. On the other hand, studies on the reactions of nitrile oxides with multiple bonds containing noncarbon atoms are adequately dealt with in the above monograph[1] but only up to 1970. In the subsequent reviews, only slight attention was devoted to such interactions. Furthermore, the reactions of nitrile oxides with certain multiple bonds (for example, C=As, C=P^{V}, C≡P, and Ge=S) began to be investigated comparatively recently and these are not mentioned in the above reviews.

The present review presents a systematic account of the literature (up to 1992 inclusive) on the reactions involving the addition of nitrile oxides to multiple bonds and an attempt is also made to trace certain rules governing these processes on the basis of the above experimental data.

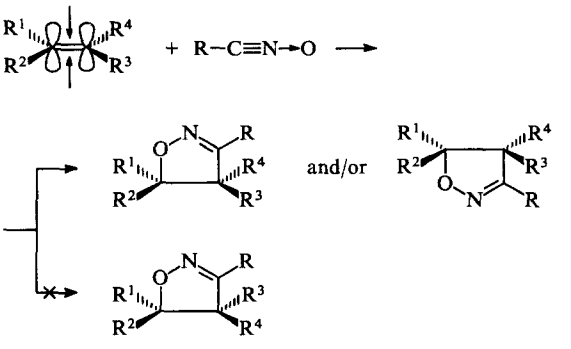
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II. General characteristics of the cycloaddition reactions of nitrile oxides

Before proceeding to the consideration of the reactions of nitrile oxides with particular multiple bonds, one should note certain features characteristic of all the cycloaddition reactions of nitrile oxides associated with their general mechanism.[2,6–9]

1. The high stereospecificity of the cycloaddition. The dipole adds in the *cis*-position relative to the multiple bond. The configuration, for example that of *cis*- and *trans*-olefins, is not then affected.



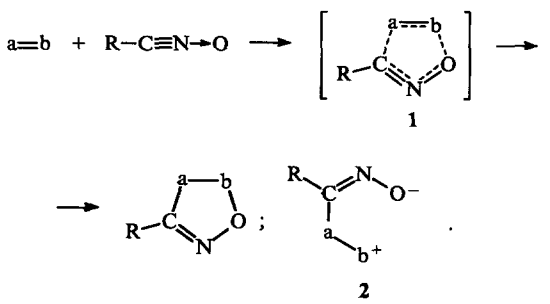
This property has found extensive application in the synthesis of analogues of natural products and biologically active compounds with specific stereochemical parameters.

2. Moderate enthalpies of activation (E_a = 9 – 16 kcal mol^{-1}) and high negative entropies of activation ($\Delta S_a^{\neq}$ = from –18 to –30 cal mol^{-1} K^{-1}), which are characteristic of a highly ordered transition state (TS).

3. A weak dependence of the rate of reaction on solvent polarity, indicating an insignificant charge separation in the transition state. This finding is also confirmed by the weak dependence of the rate of reaction on the nature of the substituents in the dipole and the dipolarophile. The values of the parameter ρ in the Hammett equation are in the range from 0.3 to 1.3.

Such experimental data indicate a concerted asynchronous mechanism of the cycloaddition reactions of nitrile oxides via the four-centre cyclic transition state **1** and rule out a stepwise mechanism via the formation of the highly charged zwitter-ions **2**.

The concerted mechanism of the cycloaddition presupposes the simultaneous dissociation of two π-bonds in the initial compounds and the formation of two new C–a and O–b σ-bonds. However, the new σ-bonds are formed at different rates and this is the reason why the cycloaddition reactions of nitrile oxides are somewhat asynchronous, which leads to a slight separation of the charges on the atoms of the transition state.



The structure of this transition state and the magnitude and signs of the charges depend both on the nature of the atoms a and b and on the nature of the substituents R in the dipole and the dipolarophile. The rate of reaction and the selectivity of the addition depend in their turn on the extent to which the formation of the transition state is favoured by energy factors.

In the description of the reactions under consideration, quantum-mechanical methods have been used recently to estimate the direction and relative rate of the addition. It is also possible to explain the influence of the nature of the substituents in the initial reactants on the course of the process.[2,9-13]

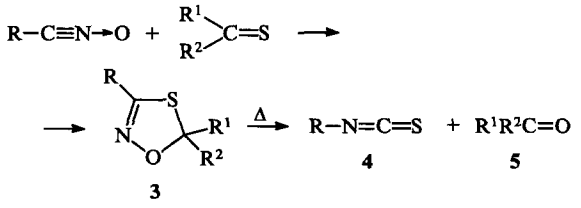
The lack of sufficient experimental data precludes at the present time a quantitative estimation of the reactivities of multiple bonds with different heteroatoms in the reactions with nitrile oxides. In purely qualitative terms, these bonds can be arranged in the following sequence based on decreasing reactivities: C=S, N=N, P^{V}=C > C=P^{III}, C=As (As=C), C=C, C=N, C=Se, B=N > C≡P, C≡C, Ge=S > P^{V}=N, C≡N > C=O. The atoms in each bond are arranged in an order such that the first adds to the oxygen atom at the end of the nitrile oxide group C≡N → O and the second adds to carbon atoms.

Next we shall consider the reactions of nitrile oxides with compounds containing all the bonds listed above (with the exception of C=C and C≡C which have been described in detail in other reviews[2-5]) in order of their decreasing reactivities.

III. Reactions involving the addition of nitrile oxides to multiple bonds

1. Addition to the C=S bond

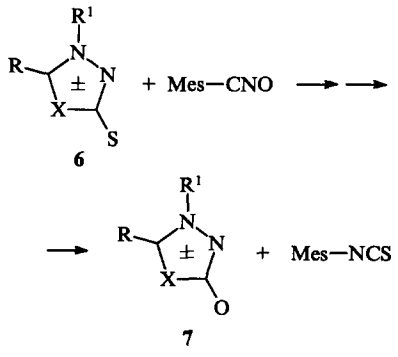
The C=S bond is the most reactive multiple bond in the series considered. At room temperature, the reactions involving the addition of nitrile oxides to C=S bonds take place over a period lasting from several minutes to several hours and lead to the formation of 3,5-disubstituted 1,4,2-oxathiazolines **3** in a satisfactory yield.[1,14-17]



Even in the case of sterically hindered aromatic nitrile oxides, the reaction with diphenyl thioketone at room temperature terminates after 30 min and leads to the cyclic product **3** in 80% yield.[18] In those cases where the dipolarophile molecule contains, apart from the C=S bond, also other reactive bonds, for example C=C [19,20] or C=N,[21,22] the nitrile oxides undergo cycloaddition primarily to the C=S bond.

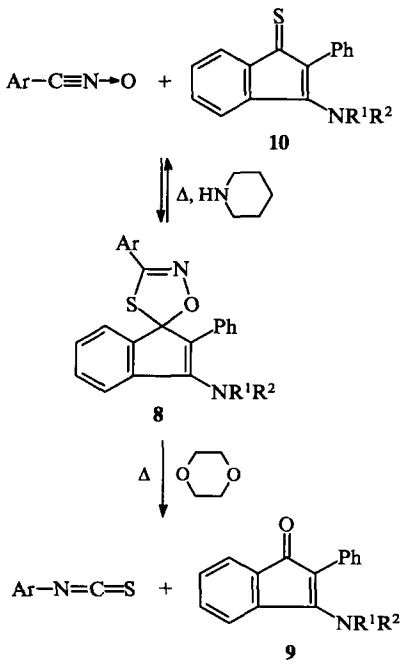
In most cases, compounds **3** are stable crystalline substances. On heating to 100-180 °C, they are cleaved to form the isothiocyanates **4** and the corresponding carbonyl compounds **5**.[14,23,2] Oxathiazoles containing an amino-group (NMe_2, NHR^3)[16,23,25,26] or the *p*-$Me_2NC_6H_4$ group[7] as the substituent R^1 (or R^2) are cleaved already at the instant of the reaction in the absence of heating. The influence of such substituents on the stability of the heterocycle is transmitted even through the C=C bond.[19,22]

In certain cases, the reactions of nitrile oxides with thiocarbonyl compounds are used to replace the sulfur atom by oxygen.[22,27,28] This method is particularly convenient for the synthesis of cyclic ketones from the more readily available thioketones.[21,28-32] A method for the conversion of the *meso*-ion of azolium thiolate **6** into the *meso*-ion of azolium olate **7** with the aid of the cycloaddition of mesityl cyanide has also been described.[33]

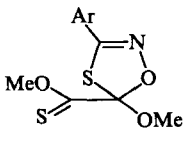


The fact that the cycloaddition reactions may be reversible under certain conditions is of interest. It has been shown[20] that, when the oxathiazole **8** is heated in dioxane, benzene, and dimethylformamide (DMF), its decomposition takes place with formation of the carbonyl compound **9**. The thermolysis of this compound in piperidine leads to the initial thioketone **10** (Scheme 1).

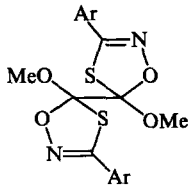
Scheme 1



O,O-Dimethyl dithiooxalate forms stable products of the cycloaddition of one (compound **11**) or two (compound **12**) molecules of a nitrile oxide.[34] Compound **12** exists in two diastereoisomeric forms, which have been separated. Dimethyl tetrathiooxalate reacts similarly with nitrile oxides, but the adducts proved to be unstable in this case.[34]



11

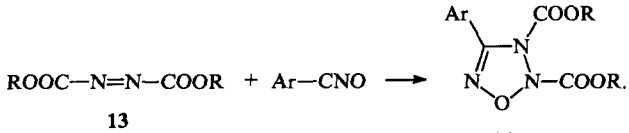


12

Kinetic studies on the reactions of nitrile oxides with 4,4'-disubstituted diphenyl thioketones have been carried out.[7] The regioselectivity of the addition of benzonitrile oxide to 2- thionoadamantane has been discussed[11] from the standpoint of the molecular orbital theory.

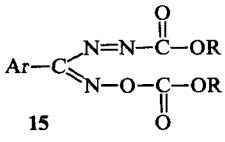
2. Addition to the N=N bond

Nitrile oxides readily react with the azodicarboxylates **13**.[35]

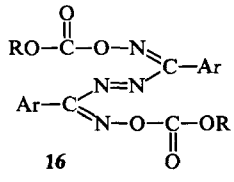


13 14

The process proceeds so vigorously that it is carried out at temperatures from −20 °C to −15 °C in order to prevent the formation of side products.[36, 37] The cycloaddition products—1,2,3,5-oxatriazolines **14**—were isolated only for the methyl esters of azodicarboxylic acid **13** (R=Me). At room temperature, they isomerise in solution to the linear products **15**. The latter can combine with yet another molecule of the nitrile oxide at the N=N bond, affording compound **16**.



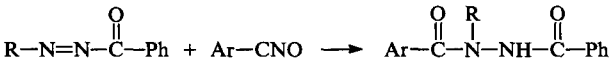
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16

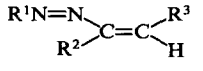
Only the isomerisation products **15** and **16**, the ratio of which depends on the nature of the group Ar, were isolated in the case of the ethyl esters **13** (R=Et). It has been suggested that the azo-compounds **15** and **16** obtained as a result of this reaction be used in the manufacture of dyes.[38]

Only the linear product **18** was isolated in the reactions of nitrile oxides with *N*-substituted *N*-benzoyl compounds **17**. The mechanism of its formation has not been established.[37]

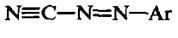


17 18

Despite the high reactivity of the N=N bond, the cyclo-addition of nitrile oxides to the conjugated azoolefins **19** and azonitriles **20** takes place regioselectively at the C=C [39] or C≡N [40] bond.



19



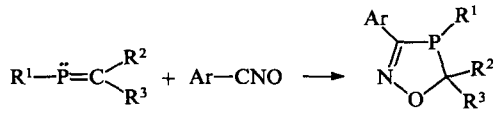
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3. Addition to the C=P bond

In this group of dipolarophiles, it is necessary to distinguish compounds with trivalent and pentavalent phosphorus atoms. Their behaviour in reactions with nitrile oxides differs appreciably.

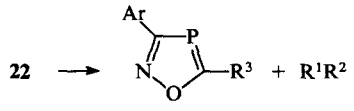
a. Addition to the C=P^{III} bond

Compounds of trivalent phosphorus are known to reduce nitrile oxides to nitriles.[1] However, in those cases where the reactivity of the phosphorus in the phosphaalkenes **21** is lowered due to steric factors, nitrile oxides undergo cycloaddition to the multiple bond with formation of 1,2,4-oxazaphospholines **22**.[41–45]



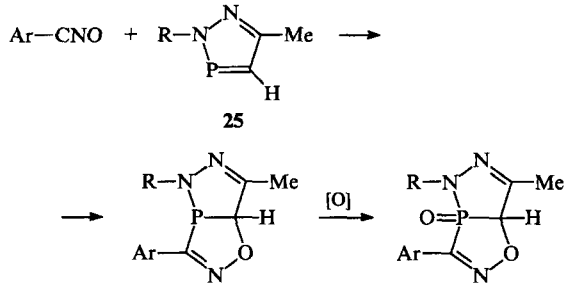
21 22

Compounds **22** are unstable in most cases. They either split off the R^1R^2 molecule and are converted into 1,2,4-oxazaphospholes **23** or are oxidised to compounds **24** with pentavalent phosphorus.[13]



23

$R^1 = Me_3Si$; $R^2 = Me_3SiO$ [39,40];
$R^1 = Cl$; $R^2 = Me_3Si$ [40,44,45].

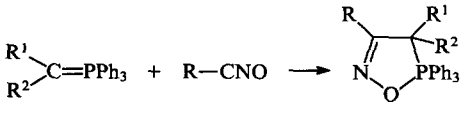


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It has been shown[13] that the selectivity of the cycloaddition of nitrile oxides to compound **25** agrees well with quantum-mechanical calculations.

b. Addition to the C=P^{V} bond

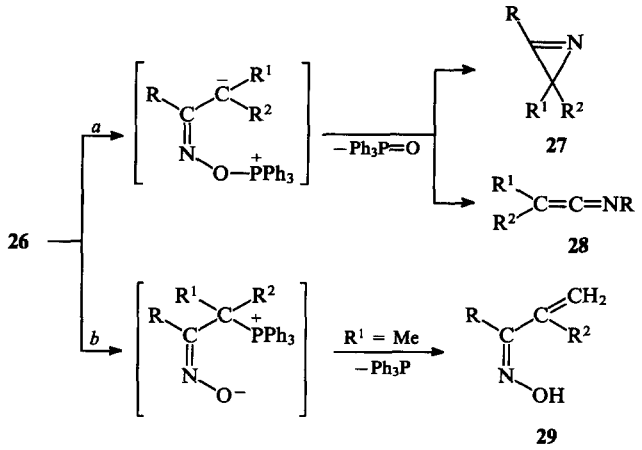
In the case of phosphinomethylenes, the cycloaddition of nitrile oxides proceeds with the opposite orientation of the atoms and leads to the formation of the P–O bond.[48–52]



26

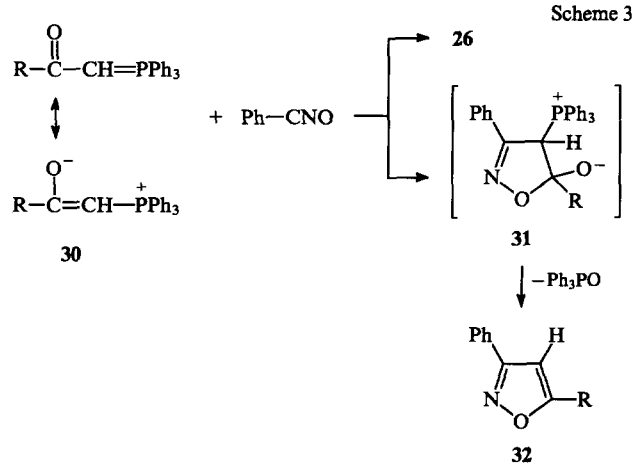
The resulting 1,2,5-oxazaphospholines **26** are unstable and in most cases are cleaved spontaneously. It has been noted in the literature that the cleavage usually begins with the dissociation of the C–P bond in the heterocycle (pathway *a*) and leads to the substituted azirines **27** or ketene imines **28**.[48, 50, 53–55] However, there are instances of the cleavage of the P–O bond (pathway *b*) with formation of the corresponding ketoximes **29**.[49] The mode of decomposition and the ratio of the final products depend on the nature of the substituents R, R^1, and R^2 (Scheme 2).

Scheme 2



The cycloaddition of nitrile oxides to phosphorus ketoylides **30** takes place at both C=P and C=C bonds. In the second case, the unstable cyclic adducts **31** are formed. They split off triphenylphosphine oxide and are converted into the oxazoles **32** (Scheme 3).[51,53]

Scheme 3



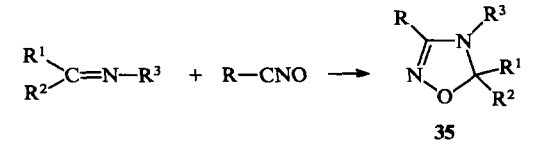
4. Addition to the C=As bond

The cycloaddition of nitrile oxides to the C=As bond began to be investigated comparatively recently, the first communication having appeared in 1981.[44] So far, this reaction is the only example where, depending on the conditions, one obtains either the product of the 'direct' (compound **33**—an As–C bond is formed) or 'reverse' (compound **34**—an As–O bond is formed) mode of addition (Scheme 4).[13,44,56]

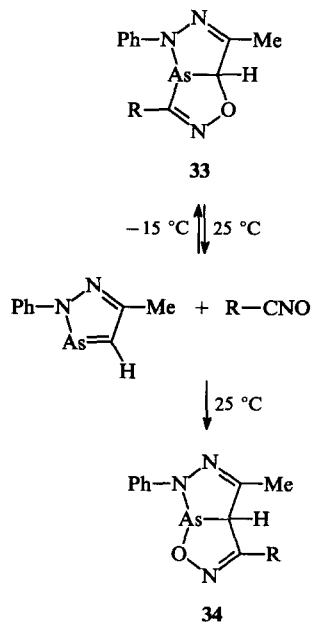
Compound **33** is obtained at a reduced temperature (−15 °C). At room temperature, it is spontaneously converted after 2 h into the isomer **34**. It has been shown that the change in the mode of addition takes place with cleavage of the oxazarsenoline ring of compound **33** to give the initial compounds. It is believed[44] that compounds **33** and **34** are formed via kinetic and thermodynamic mechanisms respectively.

5. Addition to the C=N bond

Among the reactions involving cycloaddition to multiple carbon–heteroatom bonds, those involving compounds containing the C=N bond have been studied most widely.

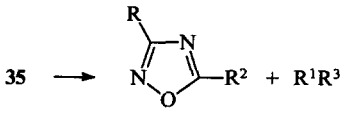


Scheme 4



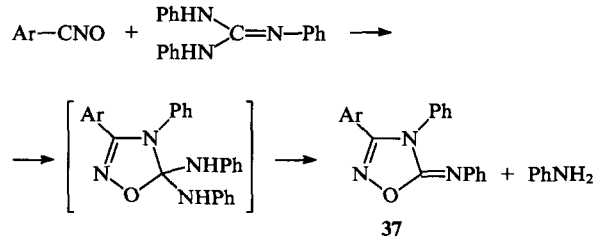
Nitrile oxides add fairly readily to this bond, forming substituted 1,2,4-oxadiazolines **35**.[1,57]

Compounds **35** are in most cases stable and can be readily isolated. At the same time, for a certain combination of the substituents R^1 and R^3, the R^1R^3 molecule is split off spontaneously and 1,2,4-oxadiazoles **36** are formed.



$R^1 = NH_2$; $R^3 = H$;[58,61]
$R^1 = OAlk$; $R^3 = H$;[58,59,62–64]
$R^1 = H$; $R^3 = OH$;[65–71]
$R^1 = CH_2C{\equiv}N$; $R^3 = H$;[72,73]
$R^1 = SMe$; $R^3 = H$.[58]

The product of the reaction of a nitrile oxide with *N,N',N''*-triphenylguanidine is stabilised somewhat differently. In this case, the aniline molecule is split off and 1,2,4-oxadiazoline **37** with an exocyclic C=N bond is formed.[74]



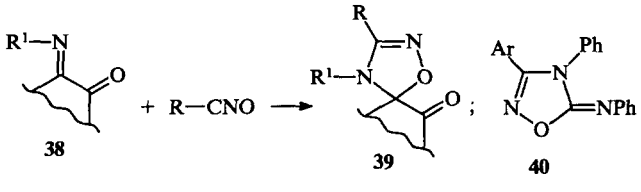
Studies have now been carried out on the interaction of nitrile oxides with virtually all classes of compounds containing the C=N bond: imines,[8,26,66,72,73,75–91] oximes,[56,65–71,92–94] derivatives of hydroxamic acid,[95] iminoesters,[58,59,62,64,96] iminothioesters,[58] amidines,[58,59,61] guanidine derivatives,[60,74,97] hydrazones,[98–101] azines,[99] and *O*(*S*)-alkyl-substituted ureas (thioureas).[58] However, despite extensive experimental data and the constantly growing interest in reactions involving the cycloaddition of nitrile oxides to the C=N bond, any kind of quantitative studies on this interaction are so far lacking. It is therefore at present impossible to determine unambiguously the dependence of the reactivity on the nature of the reactants and also to compare

the reactivity of the C=N bond with the reactivities of other multiple bonds in this reaction.

Analysis of the experimental data permits the hypothesis that electronic factors exert an insignificant influence on the reactivity of the C=N bond. As regards nitrile oxides, electron-accepting substituents in the *para*-position of the aromatic ring facilitate the reaction, shortening the time and increasing the yield of the cyclic product.[58,76]

The steric characteristics of the reactants have a much greater influence on the rate of cycloaddition. Benzonitrile oxide and its derivatives with substituents in the *meta*- or *para*-position react readily (frequently over a period of several minutes or hours at room temperature) with the C=N bond of the dipolarophile. On the other hand, in the case of sterically hindered nitrile oxides containing substituents in the *ortho*-position, the reaction does not take place at all[81] or leads to the formation of cyclic products in a low yield. In order to increase the yield, it is necessary to increase the reaction time,[102] to use more severe conditions, or to employ a catalyst.[71,94] With increase in the number and bulk of the substituents, the reactivity of the imino-group also diminishes somewhat. However, even in the case of trisubstituted imines the yield of the target product remains comparatively high: from 30% [77] to 60%–70% (provided that at least two substituents are methyl groups)[66] and up to 90% in the reaction with the reactive *p*-nitrobenzonitrile oxide.[76]

The above rules are obeyed also by the reactions involving the cycloaddition of nitrile oxides to exocyclic C=N bonds. Interaction with compounds of type **38** takes place at room temperature over a period of several hours and does not require the use of catalysts. The corresponding spirans **39** are then formed.[92,103,104]



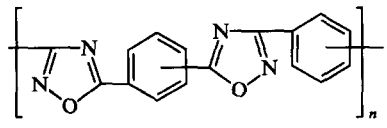
38 **39** **40**

On the other hand, more severe conditions are essential for the reactions of nitrile oxides involving the C=N bond of compound **40**; the reaction mixture is heated for several days in DMF at 75 °C with $BF_3 \cdot Et_2O$ as the catalyst.[74] The yield of the spiro-products is between 50% and 90% in all cases.

As in the case of linear azomethines, endocyclic C=N bonds in the ring readily combine with nitrile oxides, forming the corresponding bicyclic products. However, these reactions have certain unusual features. Together with the influence of steric characteristics of the substituents on the reactivity of the endocyclic C=N bond, in cyclic dipolarophiles there is an increased importance of electronic factors and a new important factor appears—that of ring strain. With increase in ring strain, there is an increase in reactivity of the imino-group, but the stability of the bicyclic compounds formed diminishes. It is noteworthy that it is stabilised in all cases with retention of the oxadiazoline ring formed.[96,105,106]

Reactions involving the cycloaddition of nitrile oxides to the C=N bonds in three-membered,[107] four-membered,[108,109] five-membered,[13,44,96,105,106,110–121] six-membered,[37,59,66,110,119,121–129] and seven-membered[102,123,130,131] rings have been investigated.

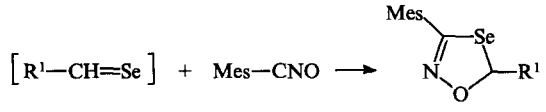
In conclusion of this Section, one should note that the reactions of nitrile oxides with imines can be used to obtain polymers or to modify them. Unfortunately, hitherto there has been only one study on this subject.[70] It was reported that the polycondensation of the bisoxides of the nitriles of terephthalic and isophthalic acids with the bisoximes of *para*- and *meta*-substituted benzaldehydes leads to polymers having the general structure **41**.



41

6. Addition to the C=Se bond

It has been reported recently that the oxide of mesityl cyanide adds even at 25 °C to the C=Se bond of selenoaldehydes, generated *in situ*, with formation of 1,4,2-oxaselenazolines **42**.[132]

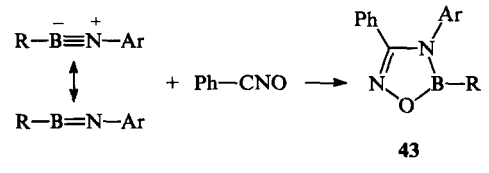


42

R^1 = H, Ph.

7. Addition to the N=B bond

There have been only two studies in which the reactions of nitrile oxides with borimides were described.[133,134] Cycloaddition products were obtained in both cases—2-bora-1,3,5-oxadiazolines **43**. It was noted that the reaction does not occur for R = C_6F_5 and Ar = mesityl.[134]

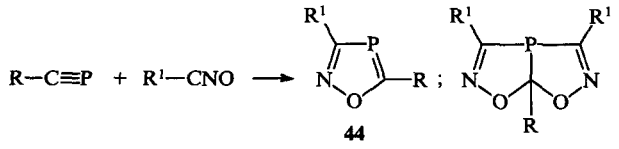


43

Ar = Ph; R = Ph;
Ar = Ph; R = 4-$MeOC_6H_4$.

8. Addition to the C≡P bond

As in the case of phosphaalkenes, the cycloaddition of nitrile oxides to phosphaalkynes is preferred to the oxidation-reduction process. The interaction even with a sterically hindered nitrile oxide terminates at room temperature in the course of 24 h and leads to the formation of stable 1,2,4-oxazaphospholes **44**. Compounds **44** combine fairly readily with a second nitrile oxide molecule via the endocyclic C=P bond, forming the bicyclic product **45** (R^1 = Ph, 4-ClC_6H_4).[45,136,137]

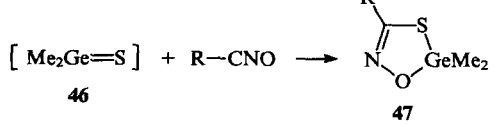


44 **45**

R = 1-Ad,[41] Bu^t.[135,136]

9. Addition to the Ge=S bond

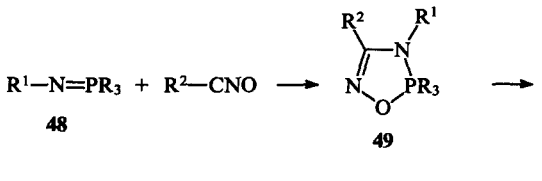
It has been reported[138] that the reaction of dimethyl germathione **46**, generated *in situ*, and nitrile oxides leads to the cyclic products **47**—2-germa-1,3,5-oxathiazolines—in 70% – 80%99 yield. Triethylamine is used as the catalyst.



46 **47**

10. Addition to the N=P^V bond

Nitrile oxides react readily with iminophosphoranes **48**. As in the case of phosphinomethylenes, cycloaddition leads to the formation of products with the 'reverse' orientation — 1,2,4,5-oxadiazophospholines **49**.[139-141] The rings formed are as a rule unstable and are cleaved during the reaction to the carbodiimides **50**.[140]

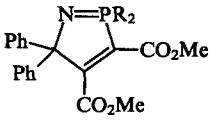


48 **49**

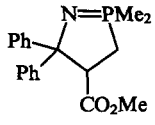
$\longrightarrow$ $R^2-N=C=N-R^1 + R_3P=O$

50

A product of type **49** can be isolated only in those cases where the N=P^{V}group is part of the composition of the ring (for example, in the azaphosphole **51** and the azaphospholine **52**) [141] or contains sufficiently powerful electron-accepting substituents ($R^1 = ArSO_2$, R = Cl).[142]



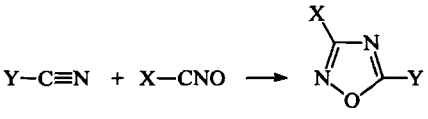
51



52

11. Addition to the C≡N bond

The interaction of nitrile oxides with nitriles has been investigated fairly widely. In all cases, the products are stable 3,5-disubstituted 1,2,4-oxadiazoles **53**.[1]



53

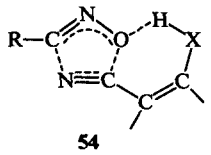
In the study of this reaction, use was made of nitriles containing a wide variety of substituents Y: Ar,[6,64,65,69,89,143-151] RC(O),[66,145,151] C_nH_m,[68,145,147,152-156] $C_nH_mHal_k$,[66,144,147,151,157-159] $R_2C(C\equiv N)$,[66,151,161-163] $R_2C=CH$,[164-173] $R_2C=C(C\equiv N)$,[162,172,174,175] $RN=N$,[40,66] $R(NR^1_2)C=CH$,[9,72,73,176] $RN=CR^1$,[177]), $R_nE^+-N^-$ (where E = S, P, N),[179] $R_2N-C(NH)-NH$,[97,139,180-182] ArO,[183,184] ArS,[185] $ArSO_2$,[186] $(AlkO)_2P(O)$ and $(AlkO)_2P(S)$,[187] $(NO_2)_3C$,[188] R_2N,[178] and Het.[66,134,136,139-141,151,189-192]

Kinetic studies have been carried out on the reaction under consideration.[6,138,154] The influence of the substituents in the reactants on the rate of cycloaddition can be determined unambiguously from the results of these studies and also from other experimental data. It was found that the reactivity of nitriles increases on passing from aliphatic to aromatic substituents and also following the introduction of electron-accepting groups and diminishes after the introduction of electron-donating groups.[66,72,150,160,190] As regards nitrile oxides, electron acceptors and donors increase somewhat the reactivity of benzonitrile oxides[72,154,187] but do not affect the reactivity of the oxides of benzoyl cyanides [$X-C_6H_4-C(O)-CNO$].[161]

Steric factors exert a much greater influence on the reactivity of the reactants. In the case of aromatic nitriles[148,150] and nitrile oxides,[187,189] it falls sharply after the introduction of substituents in the *ortho*-position. In many instances, such nitrile oxides fail altogether to enter into the reaction considered.[189]

The reactivity of unreactive nitriles and also the yield of cycloaddition products can be increased appreciably by employing catalysts (for example, $BF_3 \cdot Et_2O$).[137,152,163]

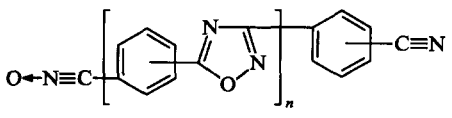
Despite the low reactivity of the C≡N bond in reactions with nitrile oxides, the cycloaddition of the dipole to unsaturated nitriles can be achieved in many instances exclusively or predominantly at the nitrile group.[9,40,66,72,97,162,164,171,174-176,182,189,191] Such increase in the reactivity of the C≡N bond has been explained[72,150,177,191] by the stabilisation of the transition state **54** and of the final product as a result of the formation of the O···H hydrogen bond.



54

X = NR^1, O.

On the other hand, in other cases the lower reactivity of the C=C bond has been explained by the influence of electronic or steric factors.[9,162]

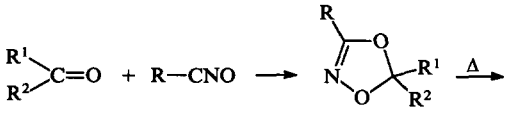


55

The possibility of using the above reaction for the synthesis of polymers has been investigated. The copolymers of the bisoxides of the nitriles of terephthalic and isophthalic acids with the bisnitriles of these acids. **41**,[70,193,194] as well as the polymer **55**, formed from *p*-cyanobenzonitrile oxide,[194,195] have been obtained. It was noted that these substances exhibit an increased stability. Their decomposition begins only at 400 °C.

12. Addition to the C=O bond

The cycloaddition of nitrile oxides to the C=O bond has been little investigated despite the fact that the first publication on this topic appeared as early as 1961.[196] In all cases, the occurrence of this interaction leads to the formation of stable 1,4,2-dioxazolines **56**.[1]



56

$\longrightarrow$ $R^1R^2C=O + R-N=C=O$

57

On heating above 150 °C, the dioxazoline ring **56** is cleaved with formation of the initial carbonyl compound and the corresponding isocyanate **57**.[57,197,198]

The interaction of compounds containing the C=O group, such as aliphatic[152,199] and aromatic[26,133,143,196,197,200] aldehydes, ketones,[75,87,196,201] vinyl ketones,[202,203] amides,[204] acid chlorides,[92,169,205] and trichlorocarbonyl isocyanate,[206] with nitrile oxides has been investigated. A series of studies have been devoted to the cycloaddition to exocyclic C=O bonds, which leads to the formation of the corresponding spiro-compounds.[91,103,129,152,199,207-210]

Merely the fact that the reactivity of the C=O bond decreases appreciably after the introduction of electron-accepting groups in the α-position relative to C=O has been firmly established for the reactions of nitrile oxides with carbonyl compounds. Thus carbonyl compounds which do not contain such substituents can interact with nitrile oxides only in the presence of catalysts

($BF_3 \cdot Et_2O$,[57,145,152,204,211] NEt_3 [203]). On the other hand, the reactions with aldehyde and ketones in which there are electron-accepting groups [CCl_3,[196,197,206] CF_3,[87,200,201] C(O)R, C(O)OR [196,197]] proceed without catalysts over a period of several hours and the yield of the cyclic adduct is 50%–90%. Aromatic aldehydes also exhibit an increased reactivity.[89,145,197,200] The behaviour of aromatic ketones in reactions with nitrile oxides has not been investigated.

The reactions of nitrile oxides with quinones have been investigated in fair detail. It was shown that *ortho*-quinones (*o*-benzoquinone, *o*-naphthoquinone, phenanthrenenequinone, and chrysenequinone) are active dipolarophiles and readily combine with2.9 nitrile oxides via the C=O (one or two) or C=C bonds depending on the nature of the substituents and the solvent.[92,103,210]

In contrast to *ortho*-quinones, the reactivity of *para*-quinones is very low. For example, the yield of the products of the cycloaddition of nitrile oxides to *p*-benzoquinone does not exceed 15%,[70,212–214] but increases appreciably when the $BF_3 \cdot Et_2O$ catalyst is used.[145,211]The reactions of nitrile oxides with substituted *p*-benzoquinones have been studied systematically since 1977.[12,198,215–218]

The dependence of the rate and mode (via the C=C or C=O bonds) of the addition on the number and nature of the substituents in the quinone has been demonstrated. The experimental data have been explained by the influence of steric and electronic factors[215,218] and have been compared with quantum-mechanical calculations.[12,217]

* * *

It is seen from the data described above that the cycloaddition reactions of nitrile oxides can be used successfully for the construction of five-membered heterocyclic structures. The latter can be both the final and intermediate products in the synthesis of various cyclic or acyclic compounds.

We hope that this review and also the recently published review of the reactions of nitrile oxides with heterocumulenes[219] will expand the range of applications of these readily available and fairly stable 1,3-dipoles.

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