

ELECTROPHILIC HETEROCYCLIZATION OF UNSATURATED AMINO COMPOUNDS
IN THE SYNTHESIS OF NITROGEN-CONTAINING HETEROCYCLES (REVIEW)

Yu. I. Gevaza and V. I. Staninets

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The literature data on the electrophilic heterocyclization reactions of unsaturated amines, enamines, amidines, amides, and urethanes are correlated. The factors that affect the reactivities of unsaturated amino compounds in electrophilic intramolecular-cyclization reactions are discussed. The problems involved in the regioselectivity of the addition of electrophiles and the stereochemistry of the resulting products are examined, and kinetic data are presented.

The electrophilic heterocyclization of unsaturated polyfunctional compounds as a general method for the synthesis of various heterocycles [1, 2] has also been successfully used for the preparation of nitrogen-containing heterocycles. Special examples of the formation of these classes of heterocycles have been presented in a review [2]. The syntheses of five- and six-membered nitrogen heterocycles based on the addition of mercury salts to unsaturated amines have been discussed in a previous review [3], and the preparation of nitrogen-containing heterocycles by electrophilic heterocyclization of unconjugated dienes was examined in [4].

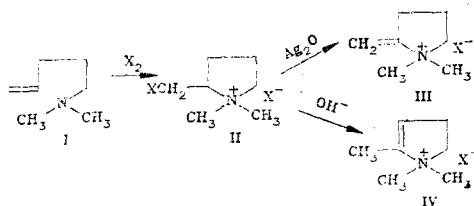
In the present review we examined the electrophilic heterocyclization of unsaturated amines, enamines, amidines, amides, and urethanes, which proceeds under the influence of electrophiles of various types and leads to the formation of, in addition to heterocycles that contain only one nitrogen atom, compounds with two nitrogen atoms, a nitrogen atom and an oxygen atom, a nitrogen atom and a sulfur atom, etc. The factors that affect the reactivities of the unsaturated compounds in this reaction are discussed, and data on the stereochemistry of the addition of electrophiles are presented, primarily with respect to reactions involving the iodocyclization of unsaturated amino compounds.

In the present review we have not included research on the synthesis of nitrogen-containing heterocycles based on the intramolecular cyclization of unsaturated N-chloramines [5].

1. Electrophilic Heterocyclization of Unsaturated Amines

Research on the addition of bromine and hydrogen bromide [6-8] to unsaturated amines was published at the end of the 19th century. However, at that time, it was impossible to unambiguously establish the structures of the resulting products. Wilstätter [9] was the first researcher to demonstrate that the action of iodine on N,N-dimethylamino-4-pentene (I) leads to the formation of quaternary salts II. Splitting out of hydrogen halides from the latter by means of Ag_2O or NaOH [10] gives, respectively, salts III or IV.

A systematic study of the iodo- and mercuricyclization of unsaturated amines in the sixties and seventies was begun by two groups of researchers [2, 3]. Some general principles involved in the occurrence of these reactions were found.



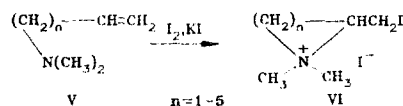
Institute of Organic Chemistry, Academy of Sciences of the Ukrainian SSR, Kiev 252660.
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1. The structures of the heterocyclization products depend on the length of the chain of the unsaturated amine, its conformation and configuration, the nature of the substituents attached to the double bond, and other structural factors.

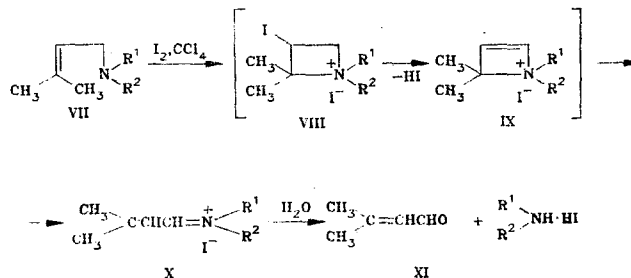
2. The heterocyclization of unsaturated amines proceeds in good yields for electrophiles such as iodine and the mercury salt. The formation of adducts with acyclic structures is also observed in the case of other nucleophiles.

3. The regioselectivity of the addition of electrophilic agents to unsaturated amines is primarily determined by the Markovnikov (Markownikoff) rule.

4. A study of the rates of iodocyclization of unsaturated amines V with different remotenesses of the reaction centers showed [11] that pyrrolinium and piperidinium rings (VI, n = 3,4) are formed more readily than others.

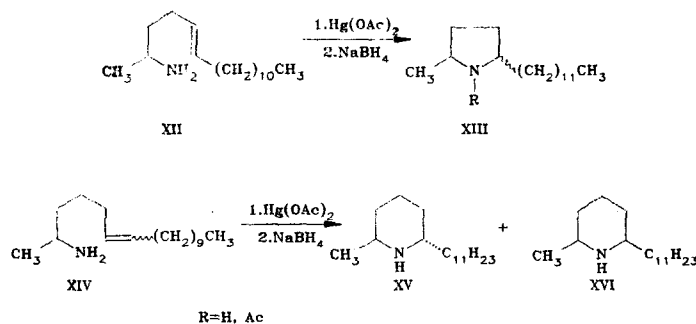


N,N-Dimethylallylamine (V, n = 1) and N,N-dimethylbuten-3-ylamine (V, n = 2) undergo virtually no iodination. However, in the case of the iodination of N,N-dialkylamino-3-methyl-2-butenes (VII) [12] it is assumed that the production of aldehyde XI is proceeded by the formation of azetidinium iodide VIII, which, through splitting out of HI, is converted to unstable azet-2-inium iodide IX. The latter readily undergoes decomposition to give the thermodynamically more favorable iminium salt structure X.

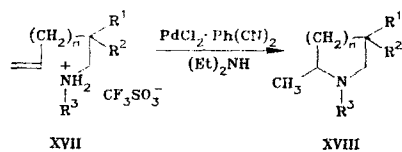


The rate constants in the iodocyclization of N,N-dialkylamino-2-butenes [12] are approximately three orders of magnitude lower than the rate constants in the iodination of N,N-dialkylamino-4-pentenenes [2, 11]. The thermodynamic characteristics of the iodocyclization of the former indicate the considerable structural requirements in the formation of a four-membered cyclic state. It is interesting that the rates of iodocyclization of N,N-dimethylpenten-4-ylamine and its analog with a triple bond are approximately the same [13].

Derivatives of pyrrolidine (XII) and alkaloids, viz., solenopsine A (XV) and isosolenopsine A (XVI) [14], respectively, in the acetoxymercuration of 2-aminohepta-5-decenes (XII, XIV) [6].

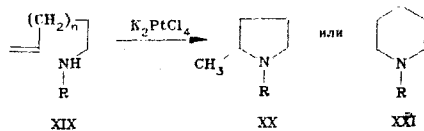


Aminoalkene trifluoromethanesulfonates XVII (n = 1, 2) in the presence of a Pd complex, depending on the length of the unsaturated chain, give either N-substituted 2-methylpyrrolidines XVIIIa (n = 1) or 2-methylpiperidines XVIIIa (n = 2) [15].



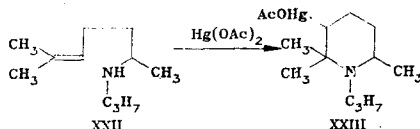
XVII, XVIII a $R^1=R^2=R^3=H$; b $R^1=R^3=H, R^2=CH_3$; c $R^1=R^2=CH_3, R^3=H$; d $R^1=R^2=H, R^3=i-C_3H_7$

Similar results were obtained in the cyclization of unsaturated amines XIX by platinum salts [16, 17]. 2-Methylpyrrolidines XXA-d or piperidines XXIa-d were isolated when $n = 1$, whereas only 2-methylpiperidine was obtained in the case of XIXa ($n = 2$).

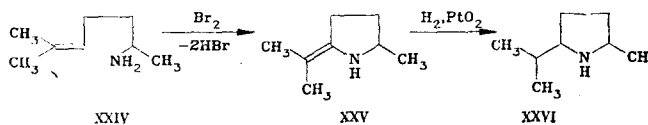


XIX—XXI a $R=H$; b $R=CH_3$; c $R=C_3H_7$; d $R=i-C_3H_7$

The nature and number of the substituents attached to the double bond have a great effect on the size of the resulting ring when iodine and mercury salts are used as the electrophilic agents. Thus only piperidine XXIII was obtained in the mercuricyclization of amine XXII, whereas the presence of only one methyl substituent attached to the double bond leads to a mixture of substituted pyrrolidines and piperidines [3].

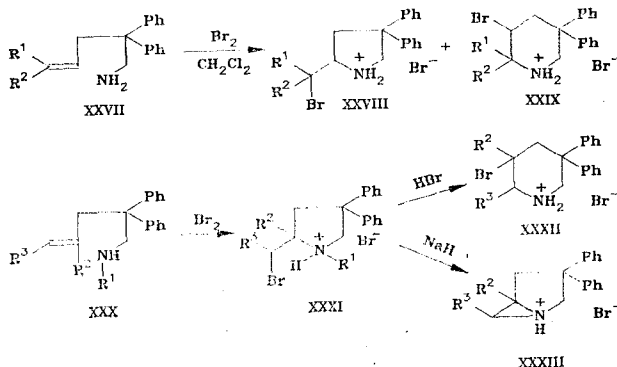


The bromination of 2-amino-6-methyl-5-heptene (XXIV) gave pyrrolidine XXV [18, 19], which, upon hydrogenation on platinum, gave 2-methyl-5-isopropylpyrrolidine (XXVI) [19].



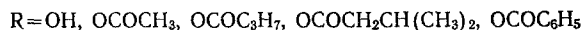
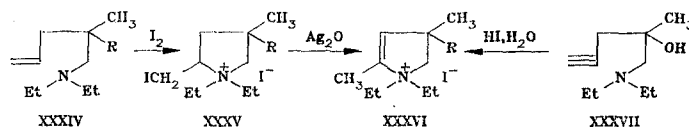
Upon treatment with HBr, 4-heptenylamine and 5-hexenylamine are converted to, respectively, 2-methylpyrrolidine and 2-methylpiperidine [20].

Upon bromination, 2,2,5-triphenyl-4-pentenylamine (XXVIIa) and 2,2-diphenyl-5-methyl-4-pentenylamine (XXVIIb) form mixtures of pyrrolidine (XXVIIIa,b) and piperidine (XXIXa,b) derivatives [21], whereas the bromination of pentenylamines XXXa-c leads only to pyrrolidine derivatives XXXI [22]. The latter, upon treatment with HBr, undergo isomerization [23-25] to piperidines XXXII, whereas they give XXXIII upon reduction with sodium hydride.

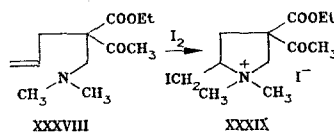


XXVII—XXIX a $R^1=H, R^2=Ph$; b $R^1=R^2=CH_3$; XXX—XXXIII a $R^1=R^2=R^3=H$; b $R^1=R^2=H, R^3=CH_3$; c $R^1=R^3=H, R^2=CH_3$

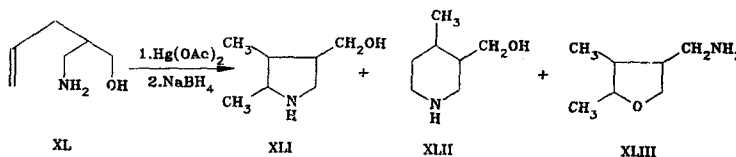
Substituents in the middle of the chain of an unsaturated amine do not affect the size of the resulting ring. Thus only quaternary pyrrolidinium salts XXXV are formed in the iodination of diethylamino-2-methyl-4-penten-1-ol and its esters XXXIV [26-28]. Splitting out of hydrogen iodide from salt XXXVa by means of Ag_2O gives pyrrolidinium iodide XXXVI [29], which was also obtained by cyclization of amino alcohol XXXVII [30]. The rate of iodocyclization of XXXIV depends only slightly on the effective volume of substituents R [27].



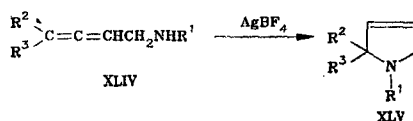
In the case of amine XXXVIII, which contains a carbethoxy group and a keto group in the β position relative to the nitrogen atom, substituted pyrrolidine XXXIX was obtained as a result of iodocyclization [31]. Of the three functional groups (carbethoxy, keto, and amino), the most nucleophilic amino group participates in ring formation.



Both nitrogen heterocycles XLI and XLII and amine XLIII were obtained in the mercuri-cyclization of unsaturated amine XL, in which participation of both the amino and hydroxy groups in ring formation is possible [3]. The preferred formation of XLI and XLII constitutes evidence for the primary participation of the more nucleophilic amino group in the reaction.

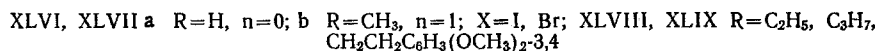
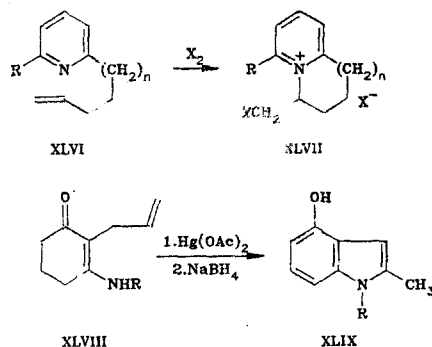


The reaction of amine XLIV with AgBF_4 gave 3-pyrroline XLV [32].



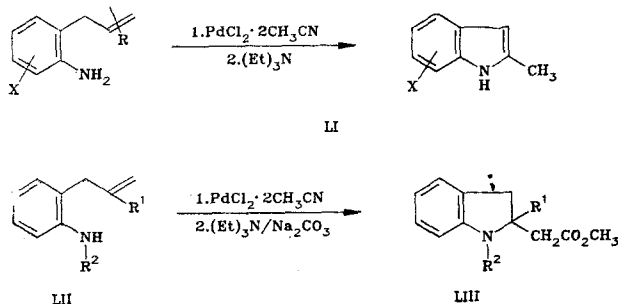
A number of examples of the formation of two-ring and polycyclic structures in the heterocyclization of unsaturated amino compounds are known.

As a result of iodination [33], 2-(1-butenyl)pyridine (XLVIa) is converted to salt XLVIIb [34].



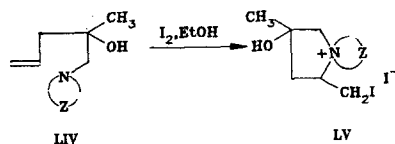
The mercuration-dehydrogenation [35] of 2-allyl-3-aminocyclohexen-2-ones (XLVIII) leads to 4-hydroxyindoles XLIX.

The cyclization of 2-allylanilines L by palladium salts gives indoles LI [36]. In the case of substituted 2-allylanilines LII under similar conditions 2,3-dihydroindoles LIII were obtained [37]. 2-Phenylselenomethylidihydroindole is also formed in the cyclization of 2-allylaniline by means of PhSeCl [38].



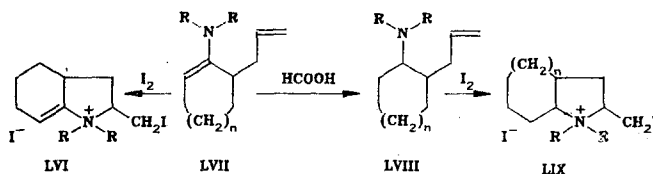
L, LI X=H, 4-Me, 4-COOEt, 4-OMe; R=H, 2(3)-Me; LII, LIII a R¹=H, R²=Me; b R¹=H, R²=Ac; c R¹=R²=CH₃

Spirobicyclic compounds LV were obtained in the iodination [26, 28] of N,N-dialkylamino-2-methyl-4-penten-2-ols (LIV).



Z=-(CH₂)_n-, -(CH₂)₂O(CH₂)₂-; n=2, 5, 6

Hexahydroindole derivatives LVI are formed in the iodination of enamines LVII in methanol at room temperature [39, 40], whereas the iodination of N,N-dialkylamino-2-allylcycloalkanes LVIII leads to the formation of the corresponding octahydroindole derivatives LIX (n = 2) or cyclopenta[b]pyrrolidine derivatives LIX (n = 1) in quantitative yields [41].

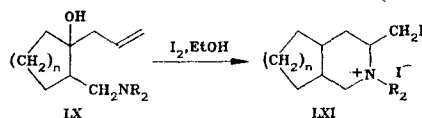


R-R=-(CH₂)₂O(CH₂)₂-, -(CH₂)₅(^o); n=1, 2

The differences in the rates of iodination of the amines of the 2-allylcyclohexane and 2-allylcyclopentane series are evidently associated with the conformational differences in the cyclohexane and cyclopentane rings [41].

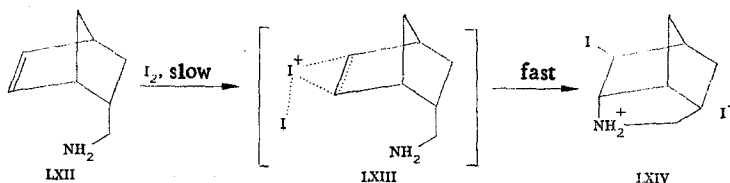
The heterocyclization of N,N-dialkylamino-2-hydroxy-2-allylcyclohexanes gives substituted octahydroindoles [42].

The iodination of cis- and trans-2-dialkylaminomethyl-1-allylcycloalkanols LX leads to the isomeric N,N-dialkyl-2-iodomethylcycloalkano[c]piperidinium iodides (LXI) [43]. In the case of amino alcohols LX (n = 1) primarily the isomers in which the allyl and aminomethyl groups are cis-oriented undergo the reaction.



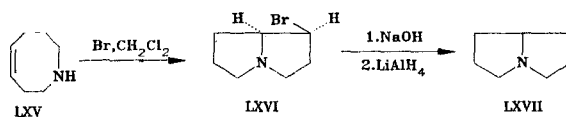
R=CH₃, -(CH₂)₅-; n=1, 2

The iodocyclization of 5-endo-aminomethyl-2-norbornene (LXII) leads to the formation of 9-exo-iodo-5-azatricyclo[4.2.1.0^{3,7}]nonane iodides (LXIV) [44, 45]. It has been noted that in the iodination of norbornene compounds one does not observe a significant difference in the reaction rates as a function of the nucleophilicity of the functional group, as one does observe in the case of 4-pentenyl-substituted compounds, where the reaction proceeds through a slightly polarized π complex of iodine with the double bond [2]. The relatively slight effect of the nucleophilicity of the functional groups on the rate of iodination of norbornenes is explained by the fact that the overall rate-determining step of the reaction is the step involving the formation of iodonium cation LXIII, whereas the participation of the functional group in the formation of the heteroring is realized in a subsequent fast step [45].

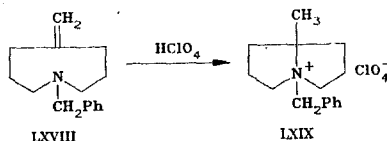


Of general interest is the heterocyclization of eight-membered and other macrocyclic unsaturated amines, since it often leads to analogs of natural alkaloids.

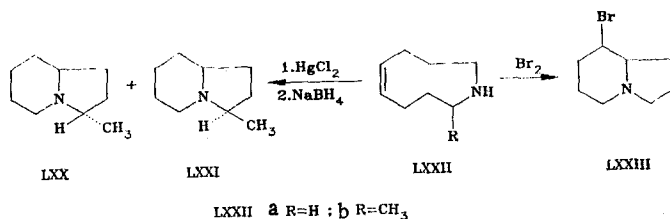
It has been shown [46] that the bromination of 1-azacyclo-4-octene (LXV) leads to bromopyrrolizidine LXVI, which, upon treatment with alkali and subsequent reduction with LiAlH_4 , gives pyrrolizidine LXVII. Iodine (I_2), HgCl_2 , PhSeBr , and PhSBr have been used as electrophiles in the reaction [47]. In all cases trans addition of these electrophiles has been demonstrated.



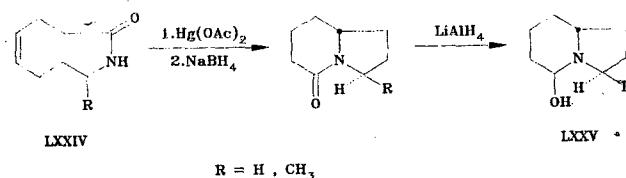
Pyrrolizidine perchlorate LXIX was obtained by the action of HClO_4 on amine LXVIII [48].



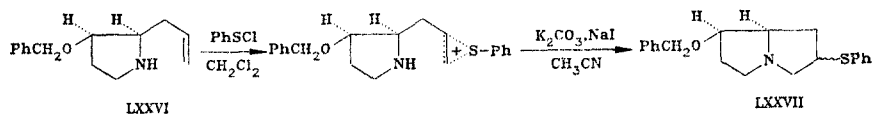
Upon bromination and mercuration, 2,3,4,7,8,9-hexahydro-1H-azonines give, respectively, bromooctahydroindolizidine LXXIIa and cis- and trans-octahydro-3-methylindolizidines LXX and LXXI [49].



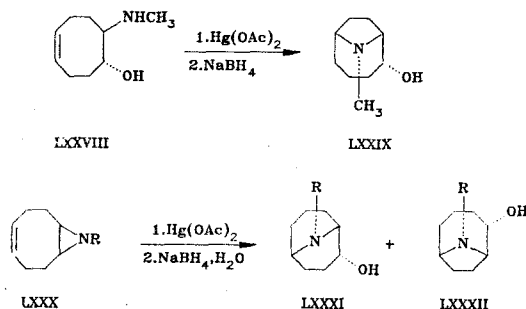
Octahydroindolizidine derivatives LXXV have also been obtained as a result of the mercuric cyclization and subsequent reduction of hexahydroazonin-2-ones LXXIV [50].



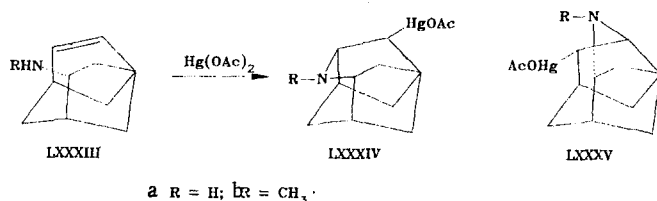
A new stereoselective synthesis of cis-1H,8H-1-benzoyloxypyrrolizidine LXXVII was accomplished in [51] starting from 2-allyl-3-benzoyloxypyrrolidine (LXXVI) and PhSCl.



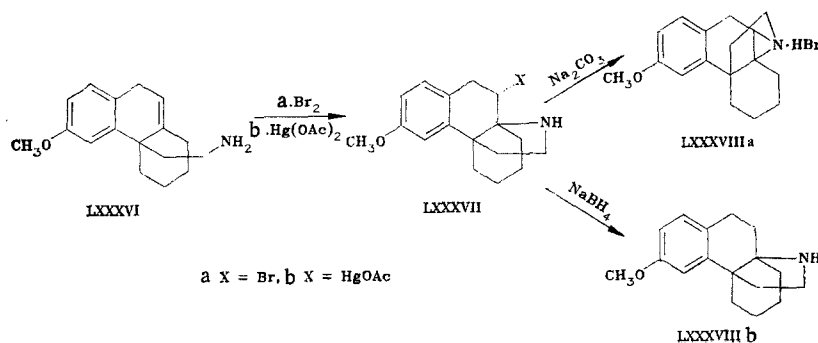
9-Azabicyclo[4.2.1]nonane (LXXIX) is primarily formed in the mercuricyclization of 5-hydroxy-6-methylaminocyclooctene (LXXVIII) [52], whereas aminocyclooctene LXXX, under similar conditions, is converted to a mixture of isomeric azabicyclononanes LXXXI and LXXXII [53, 54].



Polycyclic carcass systems LXXXIV and LXXXV were obtained as a result of the acetoxymercuration of endo-6-aminotricyclo[5.3.1.0^{4,9}]undec-2-enes (LXXXIII). Only 3-azawurzitane LXXXIVa was isolated in the case of primary amine LXXXIIIa, whereas secondary amine LXXXIIIb gives a mixture of LXXXIVa and LXXXVb in a ratio of 3:1. Only the endo compounds undergo heterocyclization [55, 56].



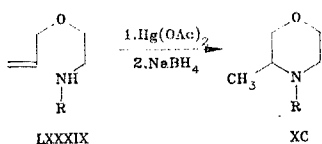
Three-ring amine LXXXVI, as a result of acetoxymercuration [57] and bromination [58, 59], is converted to 3,14-dihydroxymorphinane LXXXVIIa,b, which is subsequently converted to LXXXVIIIa,b.



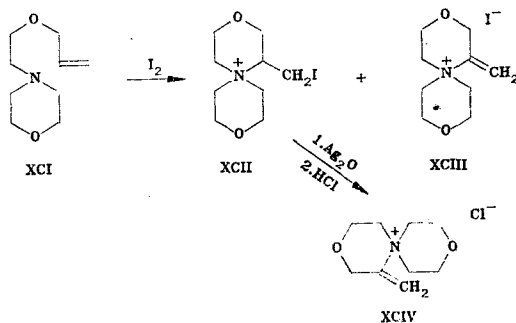
2. Heterocyclization of Unsaturated Oxy Amines, Amidines, and Other Compounds.

The corresponding substituted oxazolines and imidazolines were obtained in the bromination of α -allyloxy- and α -allylaminopyridines [2], whereas vinyloxypyridine and 2-pyridyl vinyl sulfide give oxazolo- and thiazolo[3,2-a]pyridinium salts [60, 61].

Morpholine derivatives XC were obtained in the mercuricyclization of allyl esters LXXXIX [3].

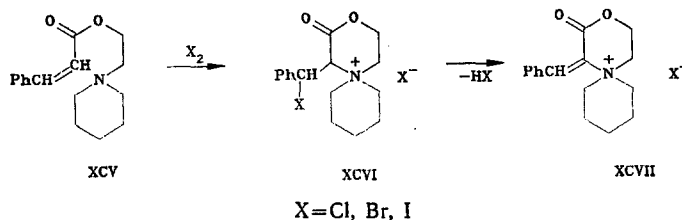


Two salts, viz., XCII and XCIII, were isolated in the iodination of allyl β -aminoethyl esters of the XCI type. Salt XCIII is probably formed as a result of splitting out of HI from salt XCII. A similar salt (XCIV) was isolated in the treatment of iodide XCII with moist silver oxide [29].



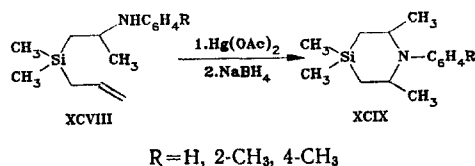
Depending on the nature of the electrophilic agent, the halogenation of *N,N*-dialkylamino-alkyl cinnamates XCV leads to salts XCVI or XCVII or to a mixture of them [62].

The chlorination of ethers XCV with chlorine in CHCl_3 gives salt XCVI. Only salt XCVII ($X = \text{Br}, \text{Cl}$) is formed from them in the case of iodination in methanol and bromination in CHCl_3 . The chlorination and bromination of ethers XCV with *N,N*-dichloro(dibromo)-5,5-dimethylhydantoin in methanol leads to a mixture of salts XCVI and XCVII.



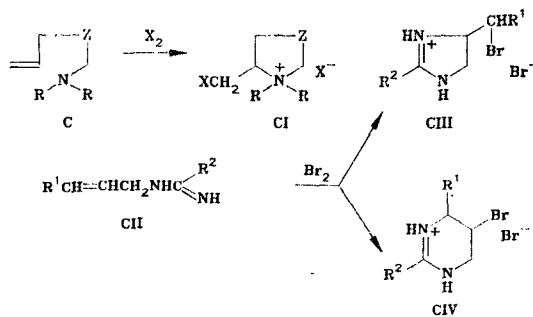
The facile splitting out of HX from salt XCVI and its conversion to XCVII are explained by the inductive effect of the adjacent ammonium and carbonyl groups on the lability of the hydrogen atom in the 2 position.

Stereoisomeric 1-aza-4-silacyclohexanes XCIX were obtained in the acetoxymercuration of silicon-containing amine XCVIII [63].



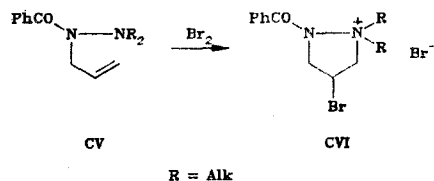
Imidazolidine derivatives CI ($Z = \text{NH}$) and oxazolidine derivatives (Z = O) were obtained as a result of the halogenation of aminomethyl ethers C ($Z = \text{O}$) and diamines ($Z = \text{NH}$) [64, 65].

Depending on their structures, both imidazolines CIIa,b and 1,4,5,6-tetrahydropyrimidines CIVc, d were obtained in the chlorination and bromination of amidines CII [66, 67].

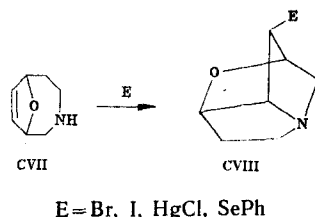


C, CI X=Br, I; Z=O, NH, S; CII—CIV a R¹=H, R²=Ph; b R¹=H, R²=CCl₃; c R¹=Ph, R²=CCl₃; d R¹=Ph, R²=CH₂F

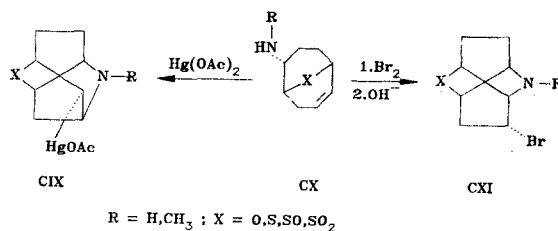
Upon bromination, N-allyl-N,N'-dialkylhydrazines CV are converted to pyrazolidinium bromides CVI [68, 69].



3-Aza-9-oxabrendane derivatives CVIII were obtained in good yields as a result of the electrophilic heterocyclization of 3-aza-9-oxabicyclo[4.2.1]non-7-ene (CVII) [70, 71].



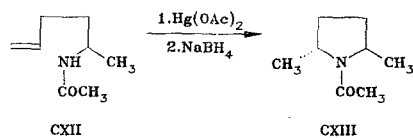
2-Aza-7-oxatwistane (CXI, X = O) and 2-aza-7-oxaisotwistane (CIX, X = O) are formed, respectively, in the bromination and acetoxymercuration of 2-methylamino-9-oxabicyclo[3.3.1]non-6-ene (CX, X = O) [7]. Similarly, 2-aza-7-thiatwistane (CXI, X = S) was obtained in the bromination of CX (X = S) [73], whereas the corresponding 2-aza-7-thiaisotwistanes (CIX, X = S, SO, and SO₂) were synthesized in the mercuriation of compounds of the CX type (X = S, SO, and SO₂) [74].



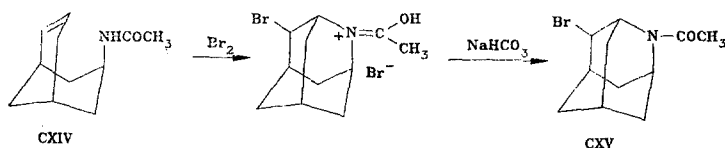
3. Heterocyclization of Unsaturated Amides and Thioamides, Ureas and Thioureas, and Urethanes

Depending on the pH of the medium, amides of unsaturated carboxylic acids, upon iodination in water, undergo cyclization to give lactims or lactams [75, 76]. Butenylamides of carboxylic acids give substituted oxazolidines upon bromination [77].

Pentenylamide CXII, like pentenylamines, is converted by the action of mercuric acetate to 2,5-dimethylpyrrolidine CXIII [78]; however, in contrast to pentenylamines, only trans-dimethylpyrrolidines CXIII are formed as a result of the reaction.

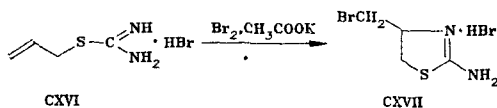


N-Acetyl-4-bromo-2-azaadamantane (CXV) was isolated in the bromination of amide CXIV [79].

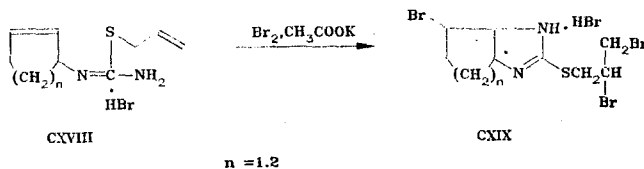


As a result of halogenation, thioamides of allylacetic acid give quaternary salts of γ -lactams [80], whereas monothioallylamides of malonic acid give substituted thiazolines [81, 82].

Upon bromination in the presence of potassium acetate, isothiuronium salts CXVI are converted to 2-amino-2-thiazolines CXVII [83].

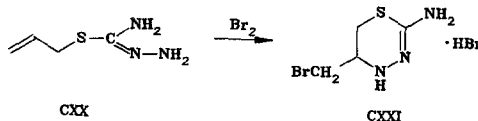


Similarly, substituted cyclopentano- (CXIX, $n = 1$) and cyclohexano[3,2-d]imidazolium bromides (CXIX, $n = 2$) were obtained from N-(3-cyclopentenyl)- and N-(3-cyclohexenyl)-S-allyl-isothiouras (CXVIII).

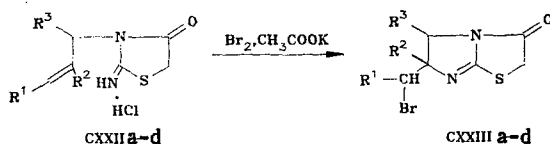


A thiazolidine derivative is formed in the bromination of N-(3-cyclopentenyl)-N'-(b-naphthyl)-S-allylthioisourea [83].

The bromination of S-allylthiosemicarbazide (CX) gives a 2,5-disubstituted 1,3,4-thiazine (CXXI) [84].

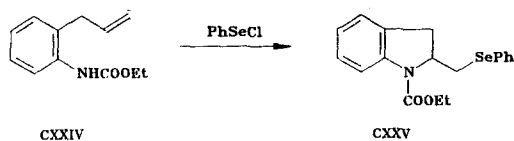


The bromination of N-allylpseudohydantoin CXXII leads to tetrahydroimidazo[2,1-b]thiazoles CXXIII [85].

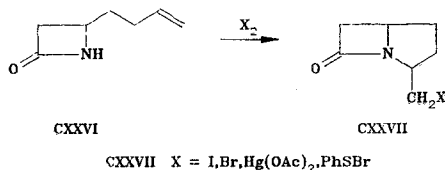


CXXII, CXXIII a $R^1=R^2=R^3=H$; b $R^1=R^3=H$, $R^2=CH_3$; c $R^1-R^3=-(CH_2)_2-$, $R^2=H$;
 d $R^1-R^3=-(CH_2)_3-$, $R^2=H$

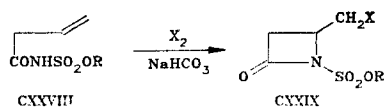
The reaction of PhSeCl with unsaturated urethanes gives derivatives of N-carbethoxy-2-phenylselenomethylpyrrolidine, N-carbethoxyindole (CXXV), N-carbethoxy-3-phenylselenomethyl-1,2,3,4-tetrahydroquinoline and -tetrahydroisoquinoline, N-carbethoxy-6-phenylselenooctahydrocyclopenta[b]pyrrole, etc. [38, 86]. The heterocyclization of urethanes with the participation of phenylselenophthalimide and mercury and palladium salts as electrophiles proceeds similarly [87-89].



The iodination and mercuration of 4-(3-butenyl)azetidin-2-one (CXXVI) gave two-ring β -lactams CXXVII, which have a skeleton of the antibiotic thienamycin [90]. The reaction of azetidinone CXXVI with bromine leads only to the dibromide, whereas a mixture of products of cyclization and addition of PhSBr is obtained with PhSBr.

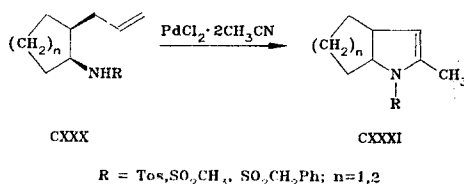


Unsaturated sulfonylamides CXXVIII react with bromine and iodine to give β -lactams CXXIX [91].



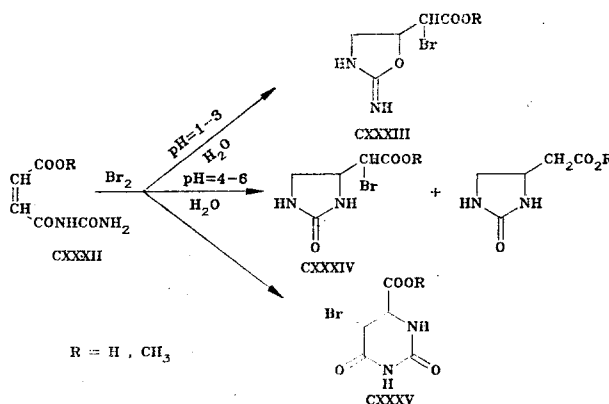
X = Br, I; CXXVIII, CXXIX R = CH₃, C₂H₅, CH₂CCl₃, C₆H₅

The two-ring systems 2-azabicyclo[3.3.0]oct-3-ene (CXXXI, n = 1) and 2-azabicyclo[4.3.0]-non-3-ene (CXXXI, n = 2) were obtained by cyclization of the corresponding N-tosyl-2-allyl-cyclopentenyl(hexenyl)amines (CXXX) in the presence of palladium salts [92].



3-Methylisocarbostyryl and 1-hydroxyisoquinoline, respectively, are formed in the reaction of palladium salts with 2-allylbenzamide [93] and o-styrylbenzamide [94].

One method for the synthesis of orotic acid (CXXXV) is bromination of maleic acid ureide (CXXXII) [95]. Depending on the reaction conditions, 2-iminooxazolidines CXXXIII and hydantoin CXXXIV can be obtained as a result of the bromination of ureide CXXXII [96, 97].



Thus the electrophilic heterocyclization of unsaturated compounds, which proceeds with the participation of the amino groups of these compounds, is of considerable interest for the synthesis of diverse classes of nitrogen-containing heterocycles, including those of natural origin.

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