# Reviews

## Cyclization of 2-halobenzoyl chlorides with dinucleophiles as a versatile approach to fused heterocycles ✾

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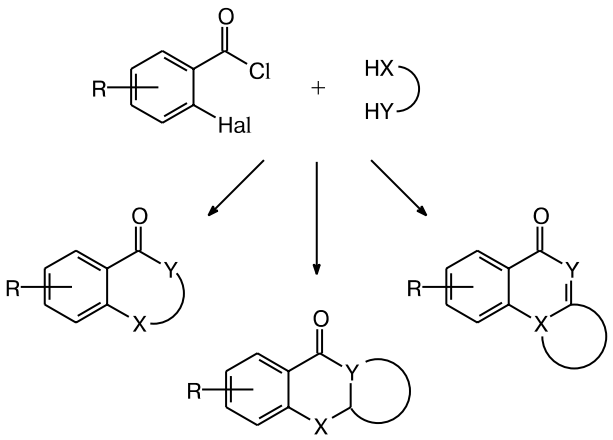
The review surveys methods for the construction of bi- and polycyclic nitrogen-, oxygen-, and sulfur-containing heterocycles based on reactions of 2-halobenzoyl chlorides with dinucleophiles.

**Key words:** 2-halobenzoyl chlorides, dinucleophiles, [*a*]-annelated quinolones, [*a*]- and [*b*]-annelated quinazolinones, imidazo[2,1-*b*]benzothiazinones, chromones, macrocycles, 2-chloronicotinoyl chlorides, benzodiazepines, benzoannelated oxa- and thiazepines, azaheterylacetonitriles, aminoazoles, aminoazines.

The synthesis of fused nitrogen-, sulfur-, and oxygen-containing heterocycles has attracted increasing interest in recent years.[1—9] Such heterocycles are either pharmacophoric fragments of known drugs or natural biologically active organic compounds. The most important of them are heterocyclic systems based on pyridine or its analogs. As an example we refer to a well known class of fluoroquinolones, which are pharmaceuticals exhibiting unique antibacterial activity.[10—36]

Approaches to the synthesis of fused heterocycles are generally based on the use of bifunctional building blocks, among which are 2-halobenzoic acid derivatives. In the present review, we survey studies on the synthesis of heterocycles based on $S_N$—$S_N$ tandem reactions of 2-halobenzoyl chlorides derivatives with bifunctional nucleophiles (Scheme 1).

**Scheme 1**




### 1. *C*,*N*-Dinucleophiles

Among 1,3-*C*,*N*-dinucleophiles, α-azaheterylacetonitriles are of paramount importance. In particular, their

* Hereinafter, the papers marked with an asterisk ✾ are dedicated to Academician Oleg Nikolaevich Chupakhin on the occasion of his 70th birthday.

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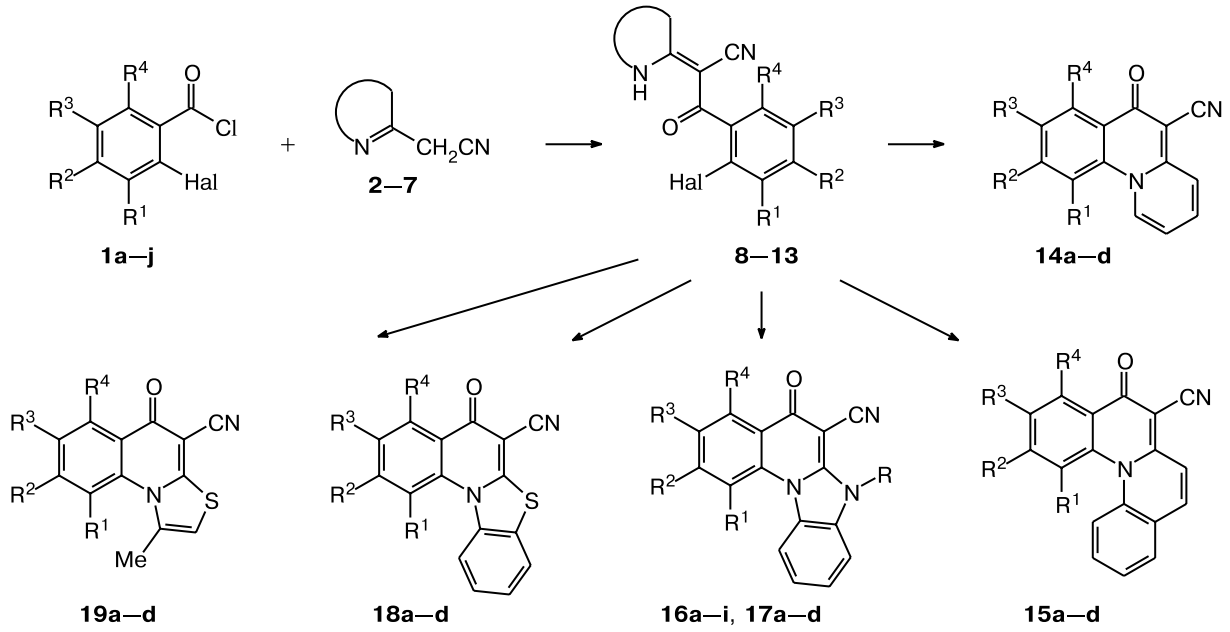
**Scheme 2**




— pyridin-2-yl (**2**, **8**), quinolin-2-yl (**3**, **9**), 1*H*-benzimidazol-2-yl (**4**, **10**), 1-methylbenzimidazol-2-yl (**5**, **11**), benzothiazol-2-yl (**6**, **12**), 4-methylthiazol-2-yl (**7**, **13**).

**1, 8—13:** $R^1 = R^2 = R^3 = R^4 =$ H, Hal = F (**a**), Cl (**b**), Br (**c**), I (**d**); $R^1 = R^2 = R^4 =$ H, $R^3 = NO_2$, Hal = F (**e**), Cl (**f**), Br (**g**), I (**h**); $R^1 = R^2 = R^3 = R^4 =$ Hal = F (**i**); $R^1 = R^2 = R^3 =$ Hal = F, $R^4 =$ H (**j**).

**14a—d—19a—d:** $R^1 = R^2 = R^3 = R^4 =$ H (**a**); $R^1 = R^2 = R^4 =$ H, $R^3 = NO_2$ (**b**); $R^1 = R^2 = R^3 = R^4 =$ F (**c**); $R^1 = R^2 = R^3 =$ F, $R^4 =$ H (**d**).

**16e—i:** $R^1 = R^3 =$ F, $R^4 =$ H, $R^2$ = pyrrolidin-1-yl (**e**), 4-methylpiperidin-1-yl (**f**), 3-methylpiperidin-1-yl (**g**), morpholin-4-yl (**h**); $R^2 = R^4$ = pyrrolidin-1-yl (**i**).

R = H (**16**), Me (**17**).

reactions with derivatives of 2-halobenzoyl chlorides afford [*a*]-annelated derivatives of tri- and tetracyclic fluoroquinolones (Scheme 2).

Acylation of α-azaheterylacetonitriles **2—7** with 2-halobenzoyl chlorides **1a-j** in the presence of triethylamine gives *C*-acyl derivatives **8—13**. The latter were demonstrated[37] to exist as enamino ketones. Depending on the structure of the azaheterocyclic fragment, compounds **8—13** were transformed into pyridoquinolones **14a-d**, quinolinoquinolones **15a-d**, benzimidazoquinolones **16a-j** and **17a-d**, benzothiazoloquinolones **18a-d**, and thiazoloquinolones **19a-d**.[8,37—44]

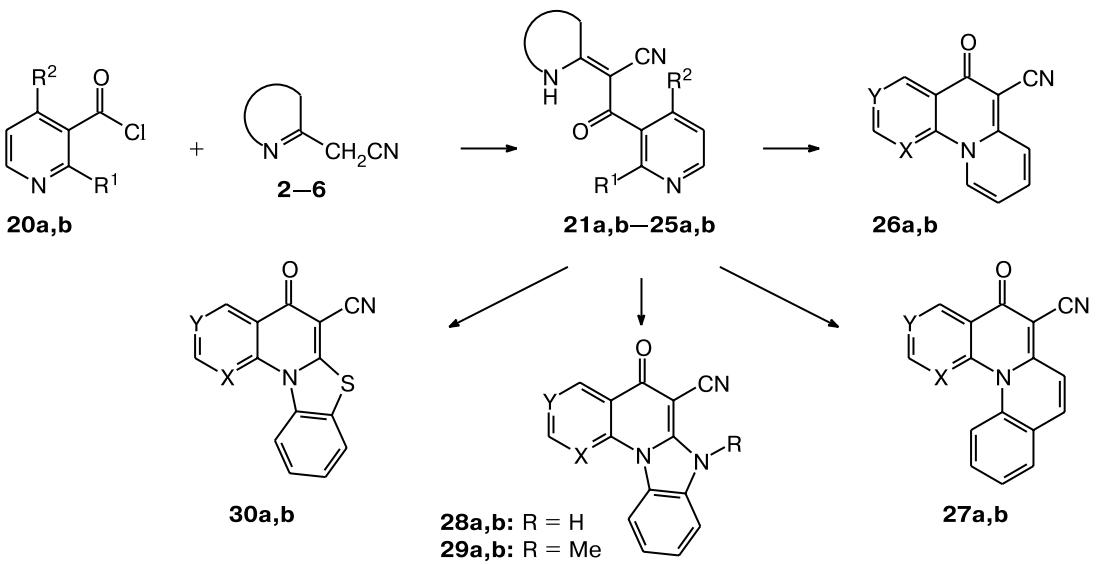
In the series of 2-halobenzoyl derivatives **8—13a-d** devoid of other substituents in the benzene ring, compound **8a** most readily undergoes intramolecular cyclization (refluxing in DMF for 2 h). 2-Chloro-5-nitrobenzoyl derivatives **8f—13f** are transformed into fused quinolones upon refluxing in DMF for 2—3 h, whereas nonactivated 2-halobenzoyl derivatives **8b-d—13b-d** undergo transformation only upon prolonged refluxing in *N*-methylpyrrolidone or 4-bromophenyl ether.[38] Therefore, the presence of an electron-withdrawing substituent in the *para* position with respect to the halogen atom facilitates cyclization of halobenzoyl derivatives **8e-g—13e-g**. Pentafluorobenzoylacetonitriles **8i—13i** are more readily subjected to cyclization than derivatives **8a-g—13a-g** (refluxing in DMF for 1 h or in *N*-methylpyrrolidone for 30 min).[38]

The reaction of 2-cyanomethylbenzimidazole **4** with tetra(penta)fluorobenzoyl chlorides **1i,j** in toluene produced 2-(1,3-dihydrobenzimidazol-2-ylidene)-3-oxo-3-polyfluorophenylpropionitriles **10i,j**, whose heating in acetonitrile in the presence of diazabicycloundec-7-ene (DBU) or in DMF in the presence of amines gave fluorine-containing benzimidazo[1,2-*a*]quinolone derivatives **16c—i**.[44]

Heating of benzimidazoquinolones **16a,e,f** in 85% sulfuric acid for 4 h is accompanied by decyanation. Evidently, this reaction involves hydrolysis of the nitrile group, but the resulting acid is unstable and is readily decarboxylated.[39,44]

The reactions of azaheterylacetonitriles **2—7** with pyridine-3(4)-carboxylic acid chlorides containing the halo-

**Scheme 3**



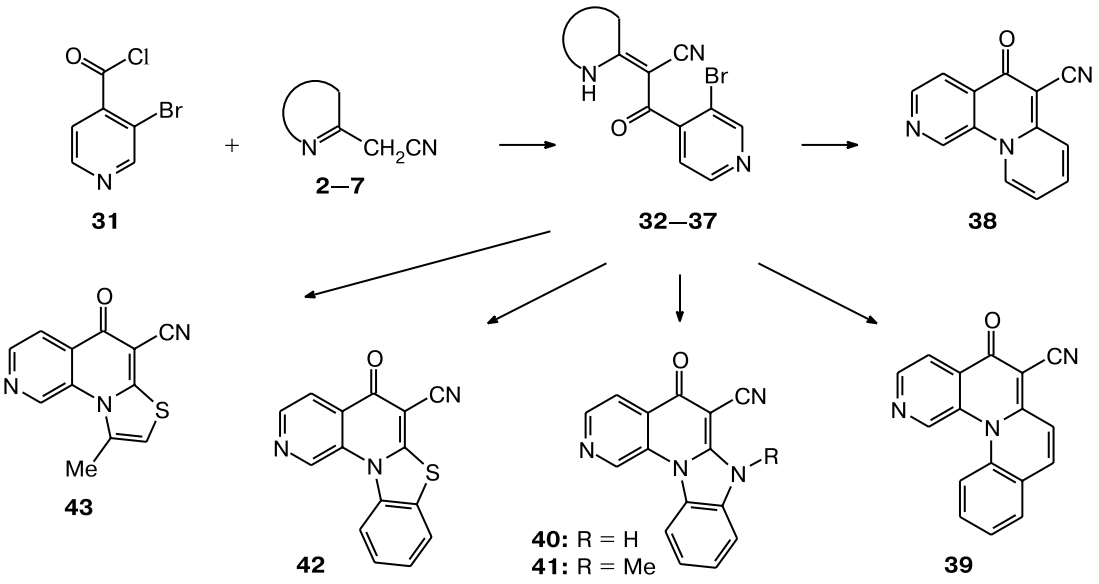

— pyridin-2-yl (**2**, **21**), quinolin-2-yl (**3**, **22**), 1*H*-benzimidazol-2-yl (**4**, **23**), 1-methylbenzimidazol-2-yl (**5**, **24**), benzothiazol-2-yl (**6**, **25**).

**20—25:** $R^1$ = Cl, $R^2$ = H (**a**); $R^1$ = H, $R^2$ = Cl (**b**).

**26—30:** X = N, Y = CH (**a**); X = CH, Y = N (**b**).

gen atom in the *ortho* position relative to the carbonyl group give fused systems containing two or three heterocycles.[41,42] Acylation of 2-cyanomethyl derivatives of azaheterocycles **2—6** with 4- and 2-chloronicotinoyl chlorides **20a,b** giving rise to cyanoketones **21—25** followed by intramolecular nucleophilic substitution of the activated halogen atom in the latter compounds provides the basis for the synthesis of pyrido-, quinolino-, benzimidazo-, and benzothiazolonaphthyridines **26—30** (Scheme 3). It was demonstrated[41] that 2-chloronicotinoyl derivatives

**Scheme 4**



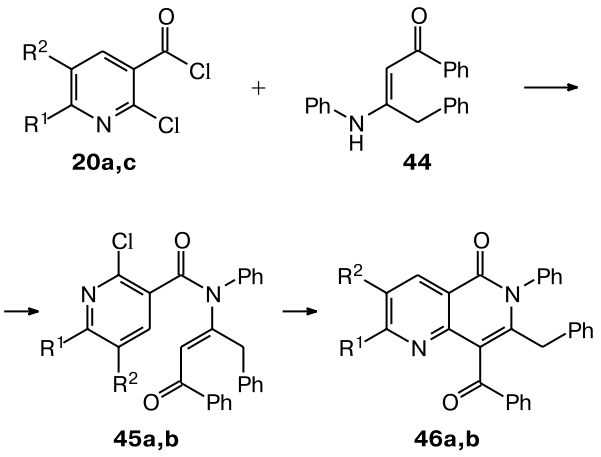

— pyridin-2-yl (**2**, **32**), quinolin-2-yl (**3**, **33**), 1*H*-benzimidazol-2-yl (**4**, **34**), 1-methylbenzimidazol-2-yl (**5**, **35**), benzothiazol-2-yl (**6**, **36**), 4-methylthiazol-2-yl (**7**, **37**).

**21a—25a** are much less reactive than the corresponding 4-chloronicotinoyl derivatives of 2-cyanomethylene-substituted heterocycles **21b—25b**. The latter were not isolated in pure form because of higher mobility of the halogen atom at position 4 of the pyridine ring compared to the halogen atom at position 2 and, all the more, at position 3.[41]

Compounds **2—7** react with 3-bromoisonicotinoyl chloride **31** to give C-acyl derivatives **32—37**, respectively (Scheme 4), which are transformed into fused 1,7-naphthyridines **38—43** by refluxing in *N*-methylpyrrolidine for 2 h. Acyl derivative **37** undergoes cyclization only in the presence of *N*,*N*-dimethylbenzylamine.[41]

Enaminones can also serve as 1,3-*C*,*N*-dinucleophiles. The reaction of 1,4-diphenyl-3-phenylaminobut-2-en-1-one (**44**) with 2-chloronicotinoyl chlorides **20a,c** yields *N*-acylated products **45a,b**, which undergo cyclization to 8-benzoyl-7-benzyl-1,6-naphthyridin-5(6*H*)-ones **46a,b** in the presence of sodium hydride (Scheme 5).[45]

**Scheme 5**

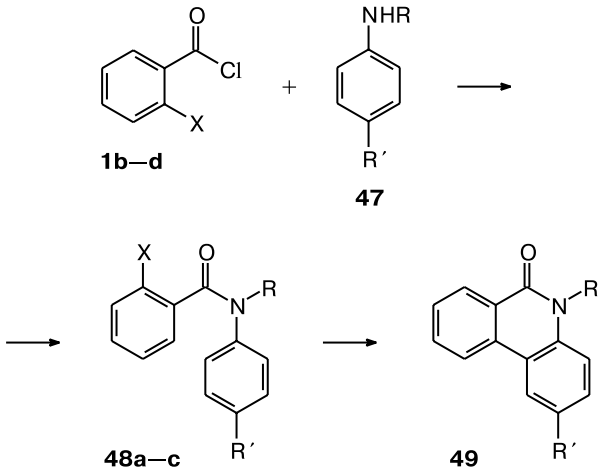


**20:** $R^1 = R^2 = H$ (**a**), $R^1 = Cl$, $R^2 = F$ (**c**);
**45, 46:** $R^1 = R^2 = H$ (**a**), $R^1 = Cl$, $R^2 = F$ (**b**).

Aniline derivatives or diphenylamines **47** can be used as 1,3-*C*,*N*-dinucleophiles. Intramolecular photocyclization of 2-halobenzoanilides **48a—c** (cyclohexane as the solvent, under nitrogen, irradiation at λ 254 nm) affords phenanthridinone derivatives **49** (Scheme 6).[46—60] 2-Bromo- and 2-iodoanilides **48b,c** undergo photocyclization to give products in lower yields than those obtained in the reactions with 2-chloro derivatives **48a**.[50—53] The substituents R′ influence the cyclization path by changing the π-donor properties of the aniline fragment and the barrier to rotation about the amide bond.[46]

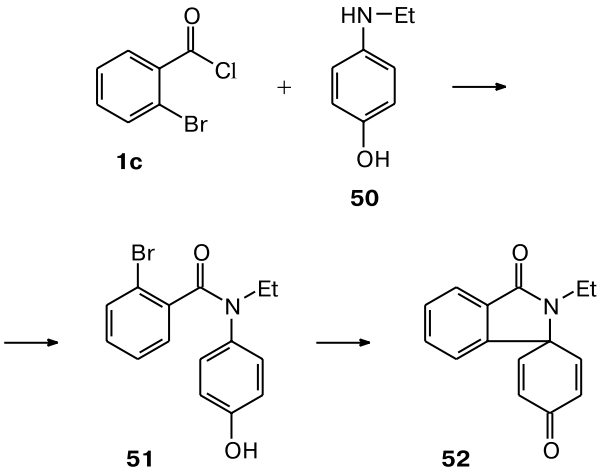
4-Ethylaminophenol **50** can act as a 1,2-*C*,*N*-dinucleophile. Photolysis of 2-bromo-*N*-ethyl-4′-hydroxybenzanilide **51** in an aqueous solution of sodium hydroxide gives rise to spirodienone **52** (Scheme 7)[61].

**Scheme 6**



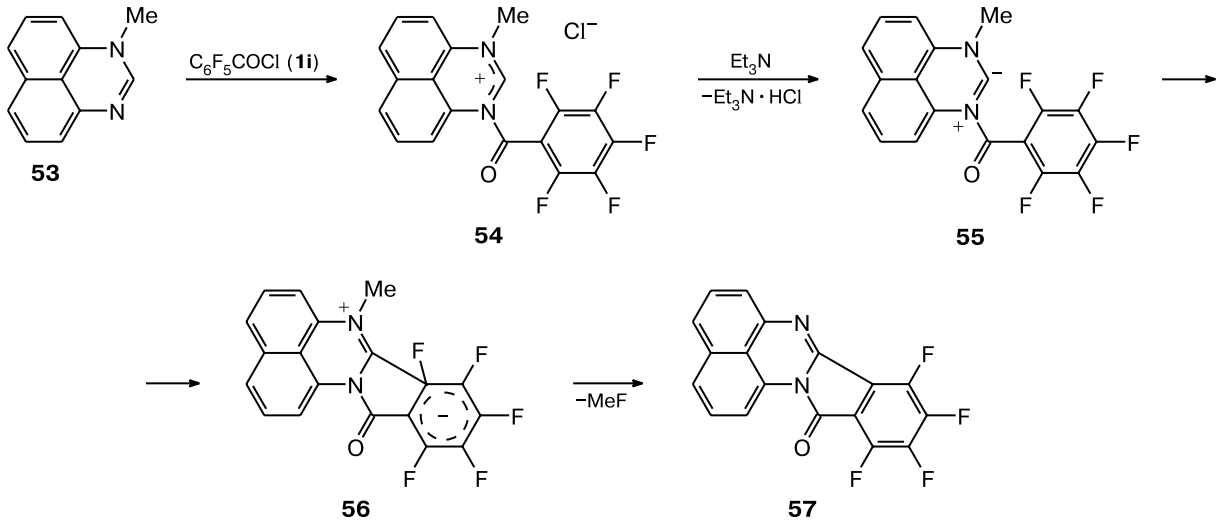
R = H, R′ = H, Me, Cl, $CF_3$, $CO_2Et$, $CO_2Pr$;
R = 4-R′-$C_6H_4$, R′ = H, Me;
**48:** X = Cl (**a**), Br (**b**), I (**c**).

**Scheme 7**

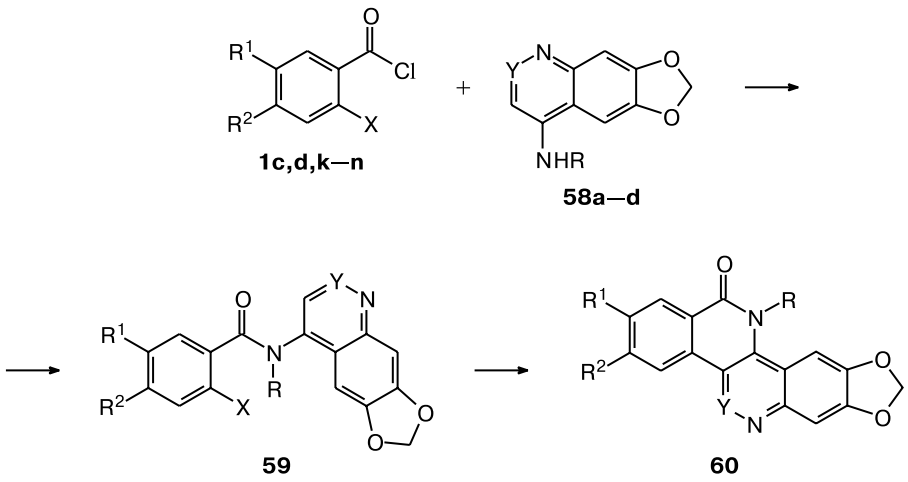


1-Methylperimidine **53** also exhibits properties of 1,2-*C*,*N*-dinucleophiles in the reaction with pentafluorobenzoyl chloride **1i** in an anhydrous medium in the presence of two equivalents of triethylamine (Scheme 8). Under the action of triethylamine, the initially formed *N*-perfluorobenzoylperimidinium salt **54** loses the H(2) proton to form ylide **55**. Then the carbanionic center intramolecularly attacks the *o*-carbon atom of the perfluorobenzene ring to give σ-complex **56**, which is transformed into final product **57** with elimination of the $CH_3F$ molecule.[62]

**Scheme 8**



**Scheme 9**



**1:** X = Br, $R^1 = R^2$ = H (**c**), OMe (**k**), $R^1 = NO_2$, $R^2 = NH_2$ (**l**); X = I, $R^1 = R^2$ = H (**d**), OMe (**m**), $R^1 = NO_2$, $R^2 = NH_2$ (**n**);
**58:** Y = N, R = $Bu^n$ (**a**), $CH_2CH_2NMe_2$ (**b**); Y = CH, R = $Bu^n$ (**c**), $CH_2CH_2NMe_2$ (**d**).

4-Amino derivatives of 6,7-methylenedioxycinnoline and quinoline **58a—d** were also used as *C*,*N*-dinucleophiles. Compounds **59**, which were prepared by aroylation of **58a—d** with 2-halobenzoyl chlorides **1c,d,k—n**, were subjected to cyclization in DMF in the presence of palladium acetate, tri(*o*-tolyl)phosphine, and silver carbonate to form polycyclic products **60** (Scheme 9).[63—65]

2-Allylpyrrolidines **61a,b** serve as examples of 1,4-*C*,*N*-dinucleophiles. Their *o*-iodobenzoyl derivatives **62a,b** were subjected to palladium-catalyzed cyclization to seven-membered heterocycles **63a,b** containing the exocyclic alkenyl group (Scheme 10).[66]

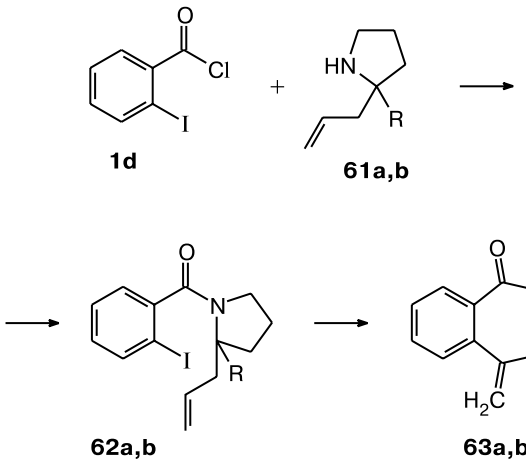
Finally, 4-allyloxybutylamine **64** provides an example of 1,9-*C*,*N*-dinucleophiles. Cyclization of *N*-[4-(allyloxy)butyl]-2-iodobenzamide **65** in the presence of tri-*n*-butyltin hydride produced lactam **66** (Scheme 11).[67]

## 2. *N*,*N*-Dinucleophiles

2-Aminoazoles, 2-aminobenzoazoles, 2-aminoazines, substituted guanidines, *etc.* can be used as 1,3-*N*,*N*-dinucleophiles.

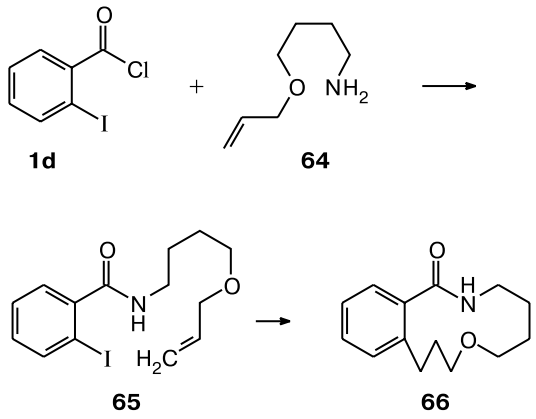
Cyclocondensation of 2-aminoazoheterocycles **67—71** with 2-halobenzoyl chlorides **1a,e,i,j,o,p** in dichloromethane in the presence of diisopropylethylamine at room temperature produced [*b*]-annelated 4(3*H*)-quinazolinone derivatives **72—76** (Scheme 12).[9,68,69]

**Scheme 10**



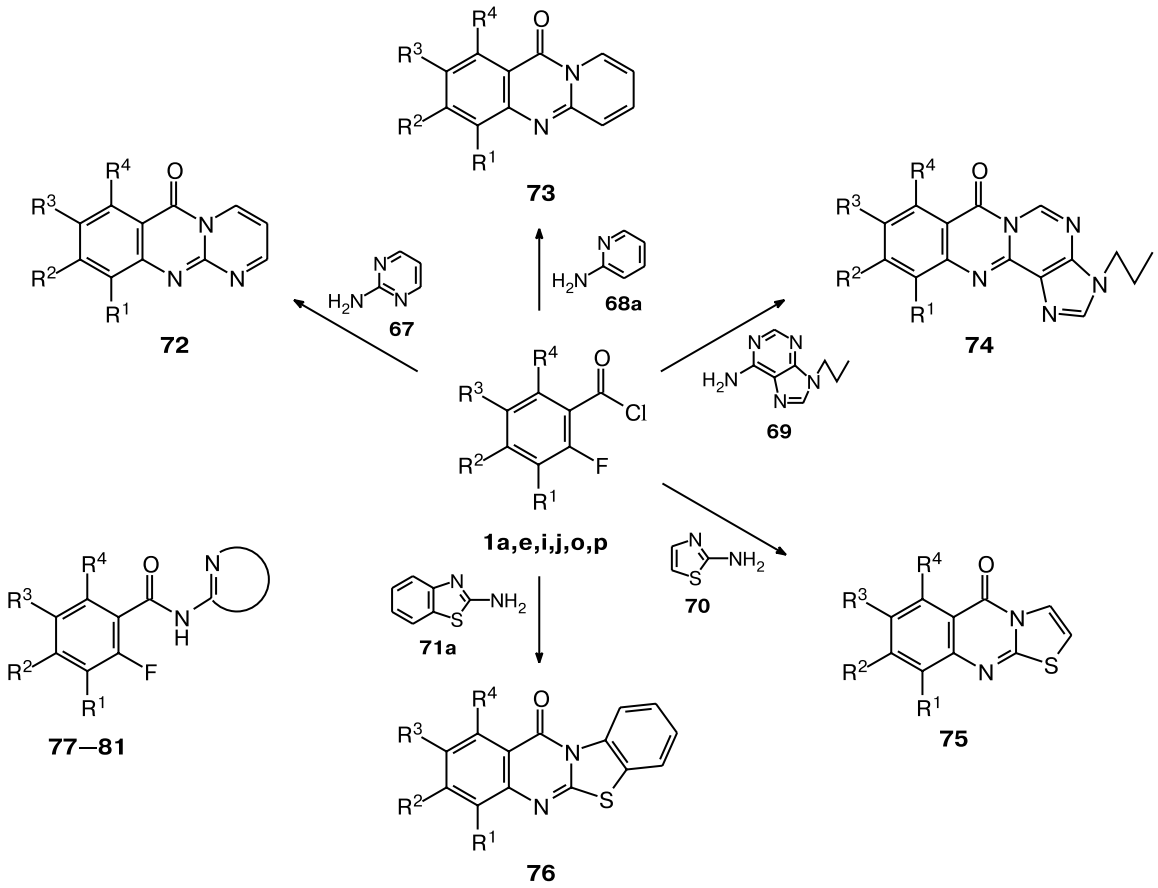

**61—63:** R = H (**a**), $CO_2Me$ (**b**).

**Scheme 11**




The linear annelation of the azolo and azine fragments to quinazolinone in products **72—76** was confirmed

**Scheme 12**



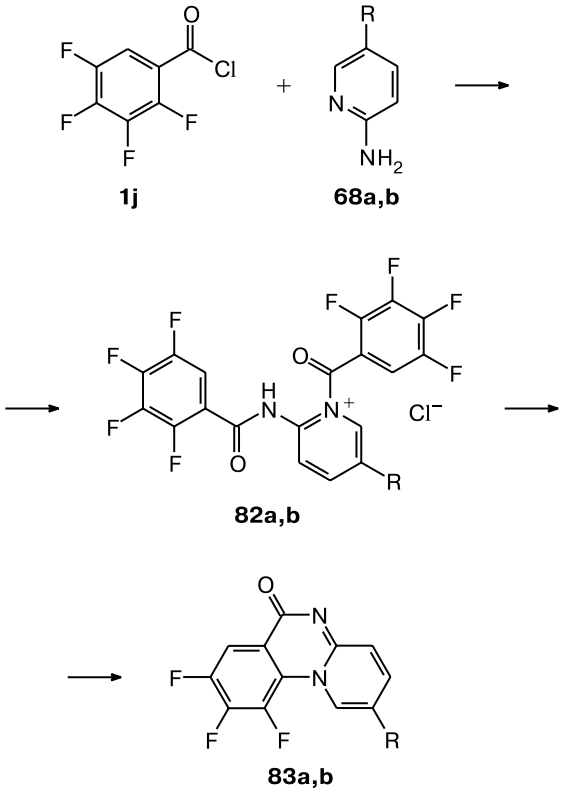

**1, 72—76:** $R^1 = R^2 = R^3 = R^4 = H$ (**a**); $R^1 = R^2 = R^4 = H$, $R^3 = NO_2$ (**e**); $R^1 = R^2 = R^3 = R^4 = F$ (**i**); $R^1 = R^2 = R^3 = F$, $R^4 = H$ (**j**); $R^1 = R^4 = H$, $R^2 = R^3 = F$ (**o**); $R^1 = R^2 = R^4 = H$, $R^3 = F$ (**p**).

 — pyrimidin-2-yl (**77**), pyridin-2-yl (**78**), 9-propyl-*9H*-purin-6-yl (**79**), thiazol-2-yl (**80**), benzothiazol-2-yl (**81**).

by $^{13}C$ NMR spectroscopy and X-ray diffraction analysis. Amides **77—81** were obtained as by-products in the reactions of 2-halobenzoyl chlorides **1a,e,i,j,o,p** with 2-aminoazoheterocycles **67—71**.[9]

The reactions of 2-aminopyridine **68a** and 2-amino-5-methylpyridine **68b** with tetrafluorobenzoyl chloride **1j** produced salts **82a,b**, respectively, which were transformed into 6*H*-pyrido-[1,2-*a*]quinazolin-6-ones **83a,b** by refluxing in toluene in the presence of triethylamine (Scheme 13). The angular structure of tricyclic derivatives **83a,b** was confirmed by $^{19}F$ and $^{13}C$ NMR spectroscopy and two-dimensional heteronuclear HetCOR and HMBC experiments.[70]

**Scheme 13**

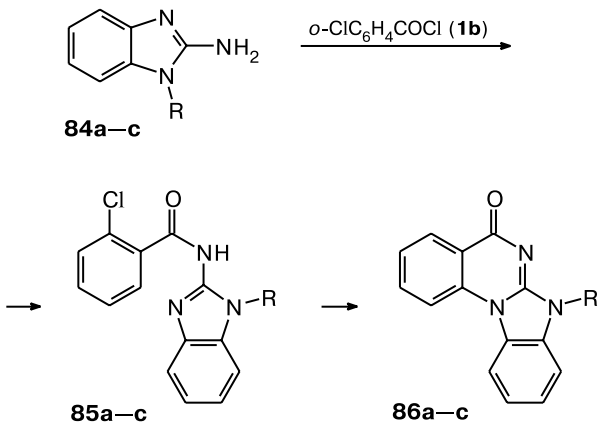



**68, 82, 83:** R = H (**a**), Me (**b**).

Intramolecular thermal condensation (200—210 °C) of *o*-chlorobenzamides **85a—c** derived from 2-aminobenzimidazoles **84a—c** afforded benzimidazo[2,1-*c*]quinazolin-5(7*H*)-one derivatives **86a—c** (Scheme 14).[71]

Thio and oxa analogs of compounds **86**, *viz.*, 5*H*-benzothia(oxa)zolo[3,2-*a*]quinazolin-5-ones **87a—d** and **88a—d**, were prepared from *N*-[2-benzothia(oxa)zolyl]-2-fluorobenzamides **89a—d** and **90a—d** by thermal cyclization (in a melt for 5 min). Amides **89a—d** and **90a—d** were synthesized by heating 2-fluorobenzoyl chloride **1a** and 2-aminobenzothiazole **71a—d** or 2-aminobenzoxazole

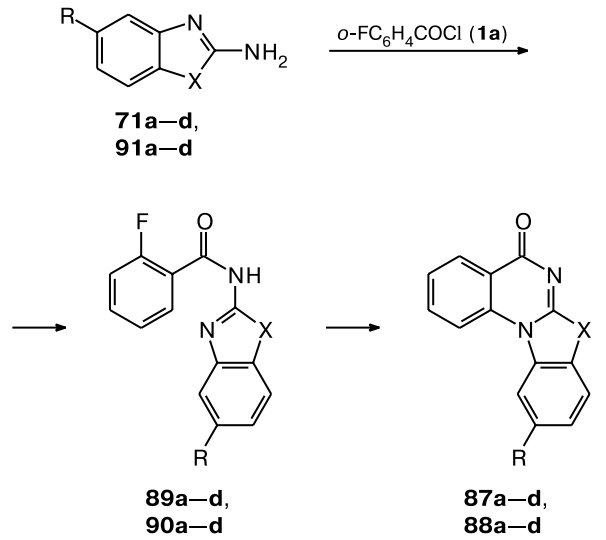
**Scheme 14**




**84—86:** R = H (**a**),Me (**b**), Et (**c**).

**91a—d** in benzene in the presence of triethylamine (Scheme 15).[72,73]

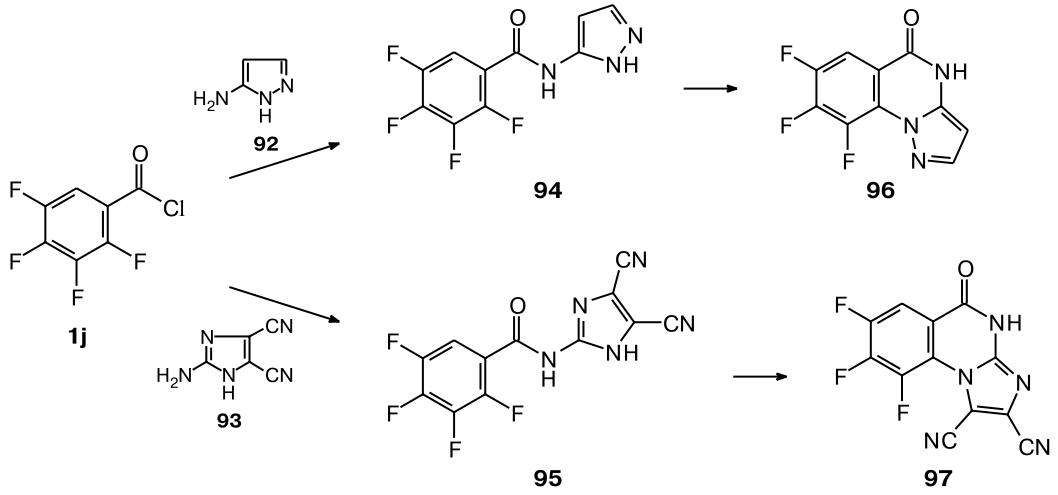
**Scheme 15**




**71, 87, 89:** X = S; **88, 90, 91:** X = O;
**71, 87—91:** R = H (**a**), Cl (**b**), OMe (**c**), OEt (**d**).

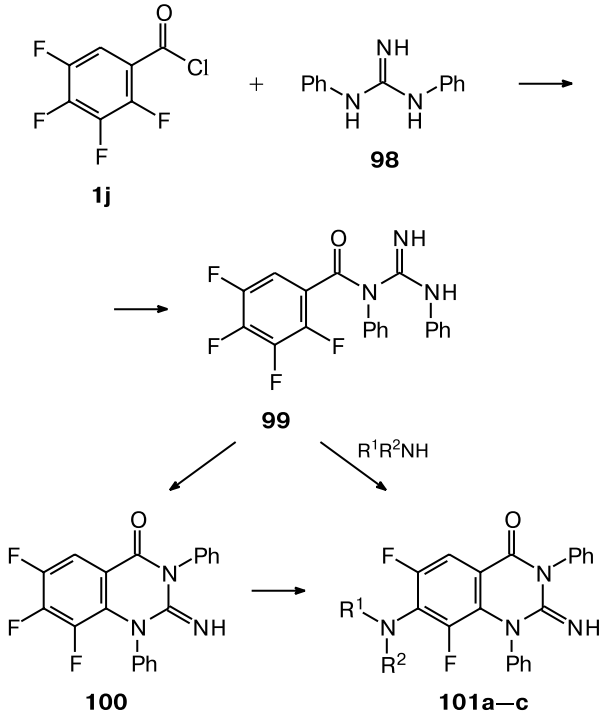
We used 3-aminopyrazole (**92**) and 2-aminoimidazole-4,5-dicarbonitrile (**93**) as *N*,*N*-dinucleophiles.[74] The reactions of tetrafluorobenzoyl chloride **1j** with aminoazoles **92** and **93** in dichloromethane in the presence of triethylamine at room temperature for one day produce monoaroyl derivatives **94** and **95**, respectively, whose heating in acetonitrile in the presence of DBU for 3—5 h affords trifluoro-containing pyrazolo[2,3-*a*]quinazolin-5-one **96** and imidazo[1,2-*a*]quinazolin-5-one **97** (Scheme 16).

**Scheme 16**



*N*,*N*′-Diphenylguanidine **98** can also be used as a 1,3-*N*,*N*-dinucleophile. We demonstrated[74] that its acylation with tetrafluorobenzoyl chloride **1j** gave rise to intermediate **99**, which was transformed into 2-imino-1,3-diphenyl-2,3-dihydro-1*H*-quinazolin-4-one derivatives **100** or **101a—c** on heating in DMF in the presence or absence of cycloalkylamines, respectively (Scheme 17).

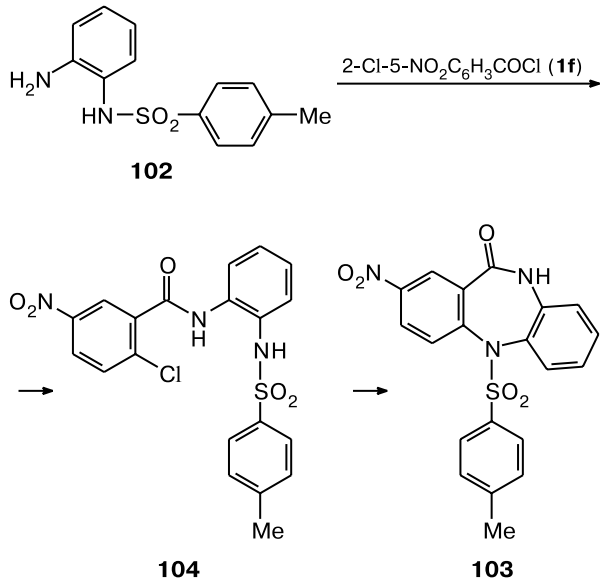
**Scheme 17**



**101:** $NR^1R^2$ is pyrrolidin-1-yl (**a**), morpholin-4-yl (**b**), 4-methylpiperidin-1-yl (**c**).

The tosyl derivative of *o*-phenylenediamine **102** also can act as a 1,4-*N*,*N*-dinucleophile, which made it possible to construct the seven-membered heterocycle, *viz.*, dibenzo[*b*,*e*][1,4]diazepine **103**, by cyclization of amide **104** (Scheme 18).[75]

**Scheme 18**



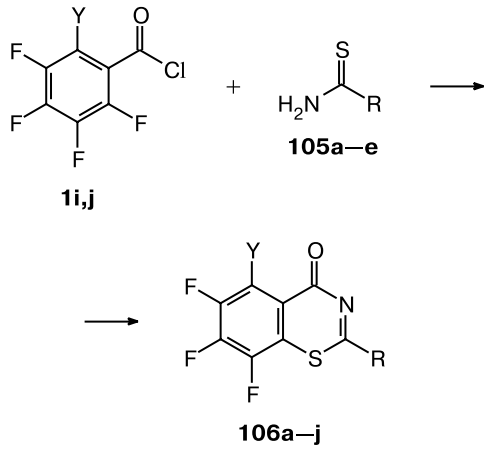
### 3. *S*,*N*-Dinucleophiles

Thioamides, phenylthiourea, *o*-aminothiophenol, and imidazole-2-thione derivatives, can be used as 1,3-*S*,*N*-dinucleophiles.

Refluxing of polyfluorobenzoyl chlorides **1i,j** with thiobenzamide **105a**, thio-2(4)-toluylamides **105b,c**, or

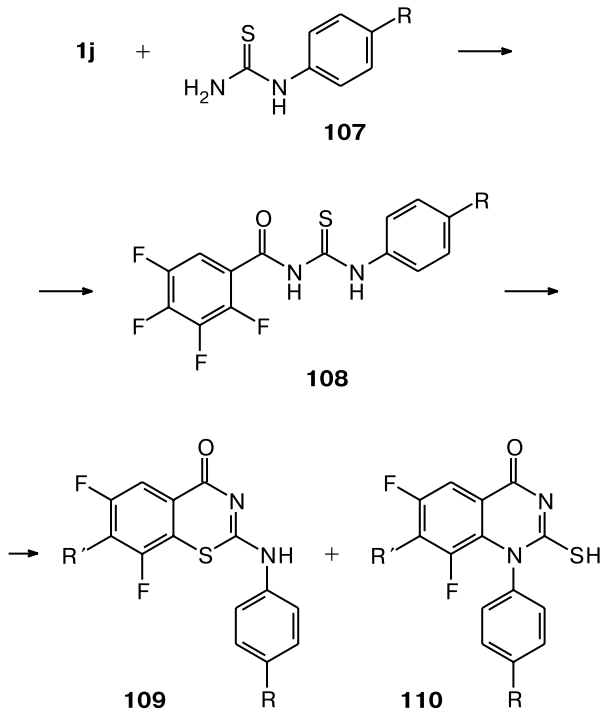
thio-2(4)-pyridinecarboxylamides **105d,e** in toluene afforded fluorine-containing [1,3]-benzothiazin-4-one derivatives **106** (Scheme 19).[76]

**Scheme 19**




**1:** Y = F(**i**), H(**j**);
**105:** R = Ph (**a**), 2-$MeC_6H_4$ (**b**), 4-$MeC_6H_4$ (**c**), pyridin-2-yl (**d**), pyridin-4-yl (**e**);
**106:** R = Ph, Y = F (**a**), H (**b**); R = 2-$MeC_6H_4$, Y = F (**c**), H (**d**); R = 4-$MeC_6H_4$, Y = F (**e**), H (**f**); R = pyridin-2-yl, Y = F (**g**), H (**h**); R = pyridin-4-yl, Y = F (**i**), H (**j**).

**Scheme 20**

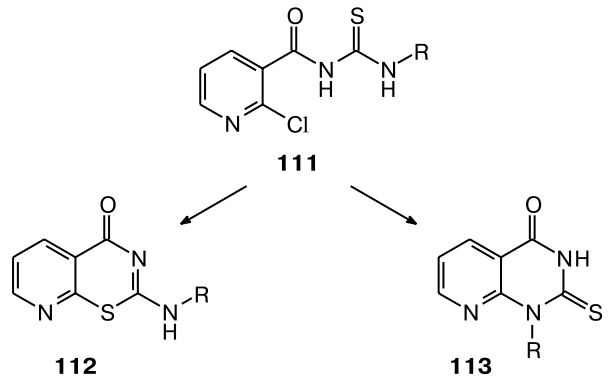



R = H, Me; R′ is pyrrolidin-1-yl, 4-methylpiperazin-1-yl.

Refluxing of acid chloride **1j** with phenylthioureas **107**, which can serve both as 1,3-*S*,*N*- and 1,3-*N*,*N*-dinucleophiles, in toluene for 1—2 h affords *N*-acylated products **108**. Compounds **108** undergo two possible cyclizations to form the thiazine or pyrimidine ring giving rise to derivatives **109** and **110**, respectively (Scheme 20).[76]

2-Chloronicotinoyl derivatives of phenylthiourea **111** were subjected to cyclization by refluxing in toluene or ethanol to form pyridothiazines **112**. Under alkaline conditions (triethylamine, 1 *M* NaOH, lithium hydride in DMF or a 1 : 1 methanol—ammonia mixture), substituted thioureas **111** give the corresponding pyridothiouracils **113** (Scheme 21).[77]

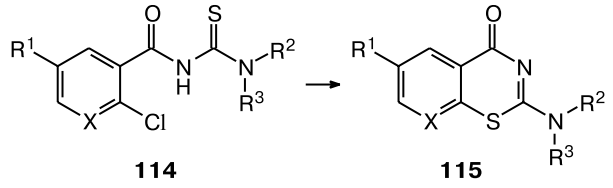
**Scheme 21**




R = Me, Et, $EtCH_2$, Ph, *p*-$MeC_6H_4$, *o*-$MeC_6H_4$, *p*-$OMeC_6H_4$, *p*-$BrC_6H_4$, *p*-$NO_2C_6H_4$, *m*-$NO_2C_6H_4$, *p*-$NMe_2C_6H_4$, α-naphthyl.

Cyclization of phenylthiourea derivatives **114** in DMF in the presence of LiOH or in benzene in the presence of triethylamine affords pyrido- and benzothiazinones **115** (Scheme 22).[77,78]

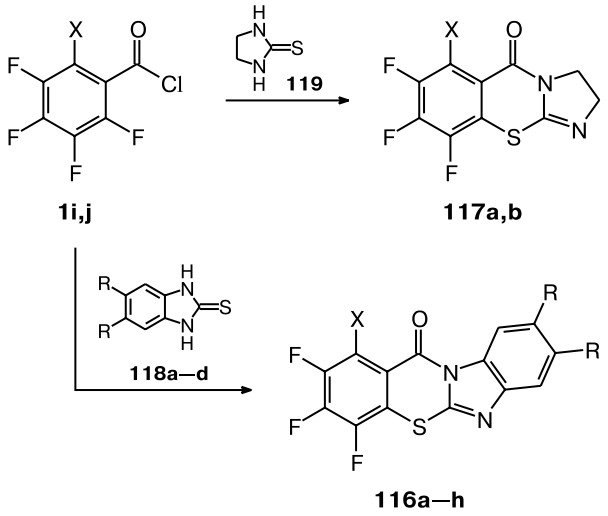
**Scheme 22**




X = N, CH; $R^1$ = H, $NO_2$; $NR^2R^3$ = $NEt_2$, $NPh_2$, $NMeC_6H_5$, pyrrolidin-1-yl, piperidin-1-yl, morpholin-4-yl.

We developed procedures for the synthesis of fluorinated imidazo[2,1-*b*][1,3]benzothiazinones **116a—h** and **117b** (Scheme 23) by the reactions of polyfluorobenzoyl chlorides **1i,j** with benzimidazole-2-thiones **118a—d** and imidazolidine-2-thione **119**, which act as 1,3-*S*,*N*-dinucleophiles, in toluene or pyridine.[79] Compounds **116a**

**Scheme 23**

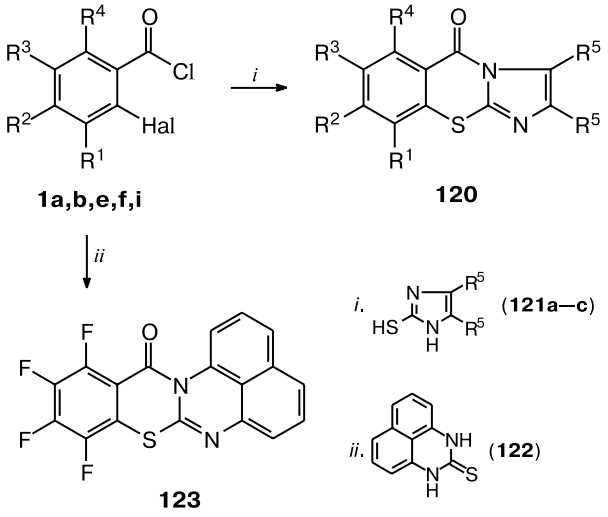


**1:** X = F (**i**), H (**j**);
**116:** R = H, X = F (**a**), H (**b**); R = Cl, X = F (**c**), H (**d**); R = F, X = F (**e**), H (**f**); R = Me, X = F (**j**), H (**h**);
**117:** X = F (**a**), H (**b**);
**118:** R = H (**a**), Cl (**b**), F (**c**), Me (**d**).

and **117a** were also synthesized[80] through bis-benzoyl intermediates under conditions of basic catalysis.

Imidazo[2,1-*b*][1,3]benzothiazin-4-ones **120** were synthesized by heating 4,5-disubstituted 2-mercaptoimidazoles **121a—c** with *o*-halobenzoyl chlorides **1a,b,e,f,i** in pyridine (Scheme 24).[81] The reaction of pentafluorobenzoyl chloride **1i** with the dianion generated from

**Scheme 24**

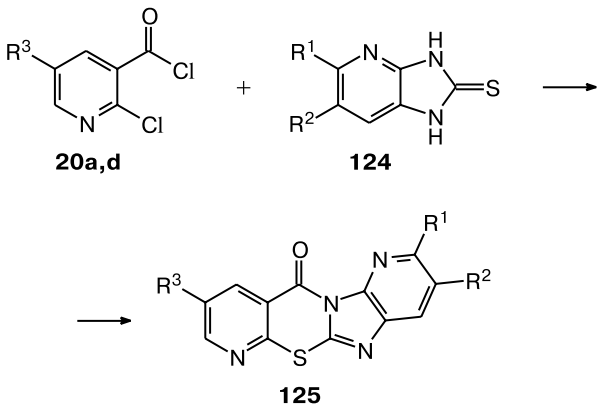


**1:** $R^1 = R^2 = R^3 = R^4$ = H, Hal = F (**a**), Cl (**b**);
$R^1 = R^2 = R^4$ = H, $R^3 = NO_2$, Hal = F (**e**), Cl (**f**);
$R^1 = R^2 = R^3 = R^4$ = Hal = F (**i**);
**121:** $R^5$ = H (**a**), Me (**b**), Ph (**c**).

perimidine-2-thione **122** under the action of *n*-butyllithium gave fused benzothiazinoperimidine system **123** (Scheme 24).[80]

The reaction of 2-chloronicotinoyl chlorides **20a,d** with thiones **124** in benzene in the presence of triethylamine yielded 11*H*-pyrido[3,2-*e*]pyrido[3′,2′:4,5]imidazo[2,1-*b*][1,3]thiazin-11-ones **125** (Scheme 25).[82]

**Scheme 25**



**20:** $R^3$ = H (**a**), $NO_2$ (**d**); **124, 125:** $R^1$, $R^2$ = H, Me.

The following scheme of the synthesis exemplifies the tandem reaction of 2-halobenzoyl chloride derivatives with dinucleophiles and mononucleophiles. Condensation of 1-(*o*-fluorobenzoyl)-2-methylthio-2-imidazolidine **126a** or 2-chloronicotinoyl derivative **126b** with *N*-aminomorpholine or *N*-aminopiperidine gives imidazo[2,1-*b*]quinazolin-5(3*H*)-ones **127a,b** and imidazo[1,2-*a*]pyrido[2,3-*d*]pyrimidines **127c,d** (Scheme 26).[83,84] The use of 2-methylthio-3,4,5,6-tetrahydropyrimidine **128** in an analogous reaction leads to the formation of pyrimido[2,1-*b*]quinazolin-6-ones **129**.[83]

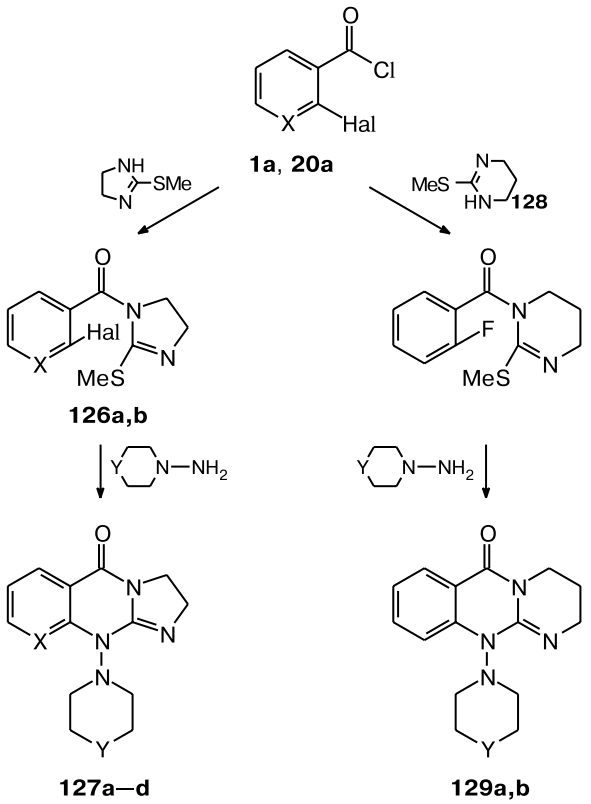
The tandem reaction of 2-chloronicotinoyl chloride **20a** with methylaminoacetonitrile and carbon disulfide[85] giving rise to 6,8-dimethyl-5-oxo-5,6-dihydro-9-thia-1,6-diazabenzocycloheptene-7-carbonitrile (**130**) is shown in Scheme 27.

Annelation of the 1,4-thiazepine ring can be performed also with the use of 2-aminothiophenol **131**. Cyclization of 2-chloro- and 2,5-dichlorobenzoyl derivatives of *o*-aminothiophenol **132** in the presence of alkali gave dibenzo[*b*,*f*][1,4]thiazepines **133** (Scheme 28).[75]

### 4. *C*,*O*-Dinucleophiles

Aceto-, benzoyl-, and pentafluorobenzoylacetic esters were used as 1,3-*C*,*O*-dinucleophiles in the reactions with 2-halobenzoyl chlorides.[86—90] The reactions of polyfluorobenzoyl chlorides **1i,r** with acylacetic esters in

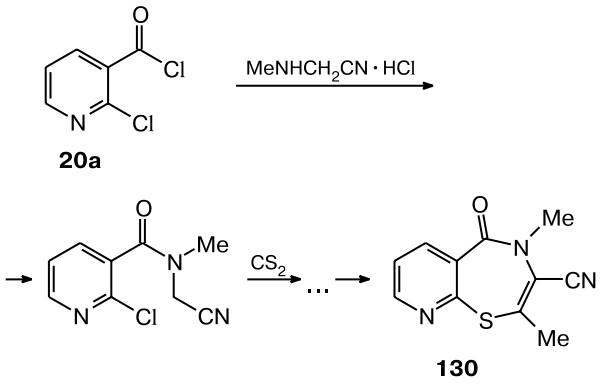
**Scheme 26**



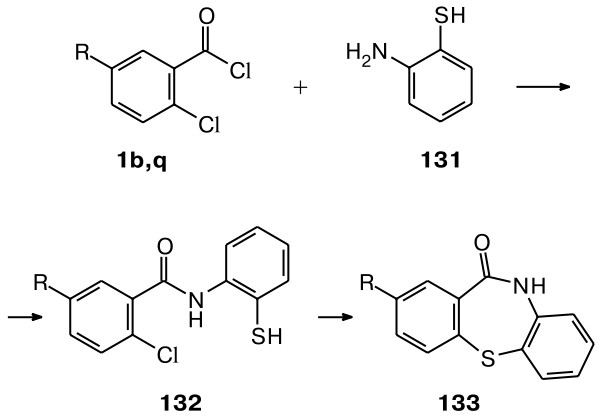
**126:** X = CH, Hal = F (**a**); X = N, Hal = Cl (**b**);
**127:** X = CH, Y = O (**a**), $CH_2$ (**b**); X = N, Y = O (**c**), $CH_2$ (**d**);
**129:** Y = O (**a**), $CH_2$ (**b**).

benzene were carried out in the presence of magnesium ethoxide. The reactions produced fluorine-containing

**Scheme 27**

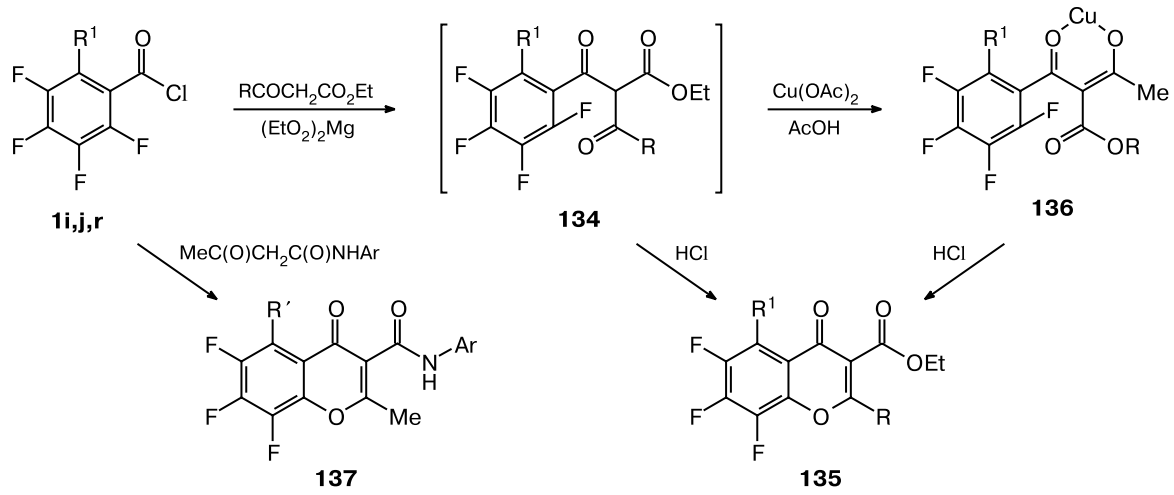


**Scheme 28**



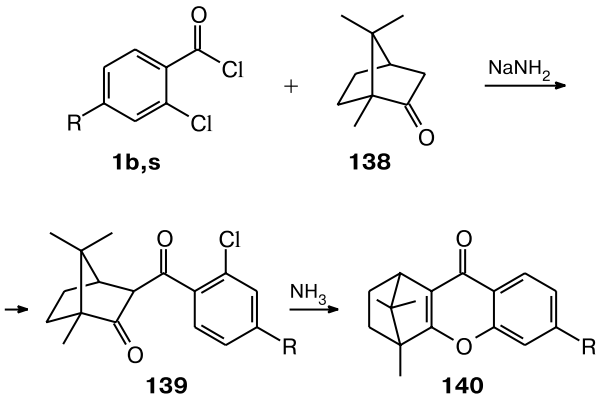
**1:** R = H (**b**), Cl (**q**); **132, 133:** R = H (**a**), Cl (**b**).

**Scheme 29**



**1:** $R^1$ = F (**i**), H (**j**), OMe (**r**);
R = Me, Ph, $C_6F_5$; Ar = Ph, 2-$MeC_6H_4$, 4-$MeC_6H_4$, 4-$OMeC_6H_4$.

**Scheme 30**




**1:** R = H (**b**), Cl (**s**); **139, 140:** R = H (**a**), Cl (**b**).

3-carbethoxy-2-methylchromones **135** instead of the expected β,β´-diketo esters **134** (Scheme 29). Chromones **135** can be prepared by heating the corresponding copper chelates **136** in an acidic medium.[90]

We prepared fluorine-containing chromone-3-carboxamides **137** by the reactions of tetra(penta)fluorobenzoyl chlorides **1i,j** with acetoacetamides, which act as 1,3-*C*,*O*-dinucleophiles, in dichloromethane in the presence of triethylamine (see Scheme 29).[91]

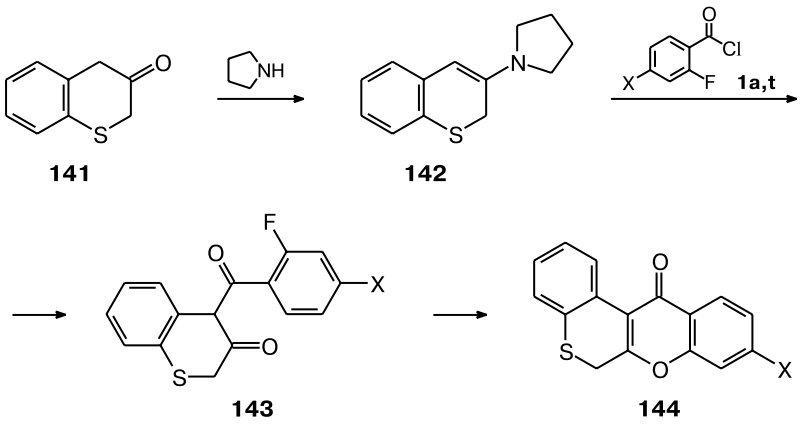
Camphor **138** was also used as a 1,3-*C*,*O*-dinucleophile. Acylation of **138** in the presence of sodium amide affords aroyl derivatives **139**. Cyclocondensation of compounds **139** in the presence of ammonia gave dimethylmethanotetrahydroxanthones **140** (Scheme 30).[92]

The reaction with the use of thiochroman-3-one **141** as a *C*,*O*-dinucleophile[93] (Scheme 31) yielded enamine **142**, which was subjected to acylation with 2-fluorobenzoyl chlorides **1a,t** and was transformed into diketone **143** due to hydrolysis. Heating of diketone **143** in toluene in the presence of $K_2CO_3$ resulted in its cyclization to give 6*H*,12*H*-[1]benzothiopyrano[3,4-*b*][1]benzopyran-12-one derivative **144**.

We prepared 5,6,7-trifluoro-2,3-dihydro-1*H*-cyclopenta[*b*]chromen-9-one (**149**) by heating tetrafluorobenzoyl chloride **1j** with enamine **145** in dichloromethane in the presence of triethylamine followed by hydrolysis and cyclization (Scheme 32).

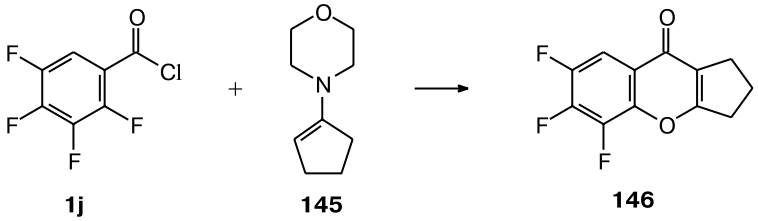
3-Methoxy-2(1*H*)-pyridinone (**147**) provides yet another example of a 1,3-*C*,*O*-binucleophilic reagent. Esters **148** were prepared by *O*-acylation of pyridinone **147** with 2-bromobenzoyl chlorides **1c,k** in the presence of $K_2CO_3$ and tetrabutylammonium bromide. Azacoumarin derivatives **149** were synthesized by the transformation of compounds **148** in their reactions with tris(trimethylsilyl)silane in benzene at 80 °C (Scheme 33).[94]

**Scheme 31**

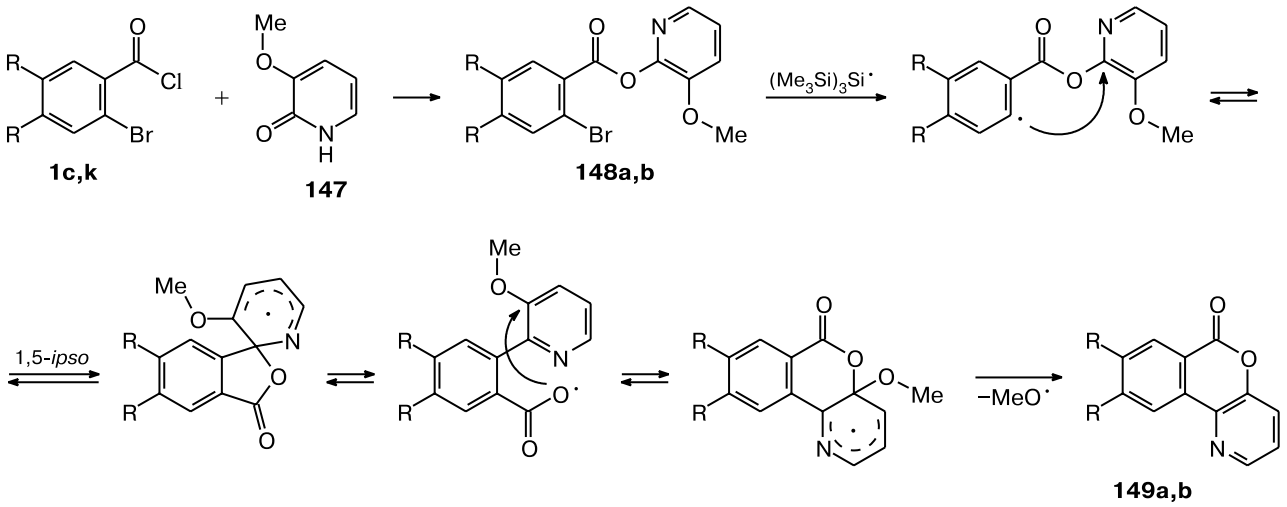



**1:** X = H (**a**), F (**t**); **143, 144:** X = H (**a**), F (**b**).

**Scheme 32**

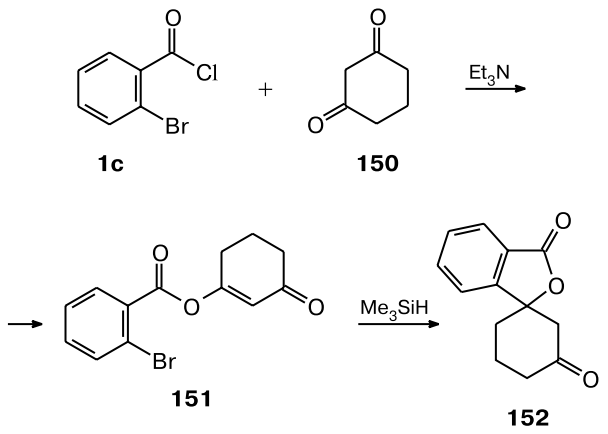


**Scheme 33**



**1:** R = H (**c**), OMe (**k**); **148, 149:** R = H (**a**), OMe (**b**).

**Scheme 34**

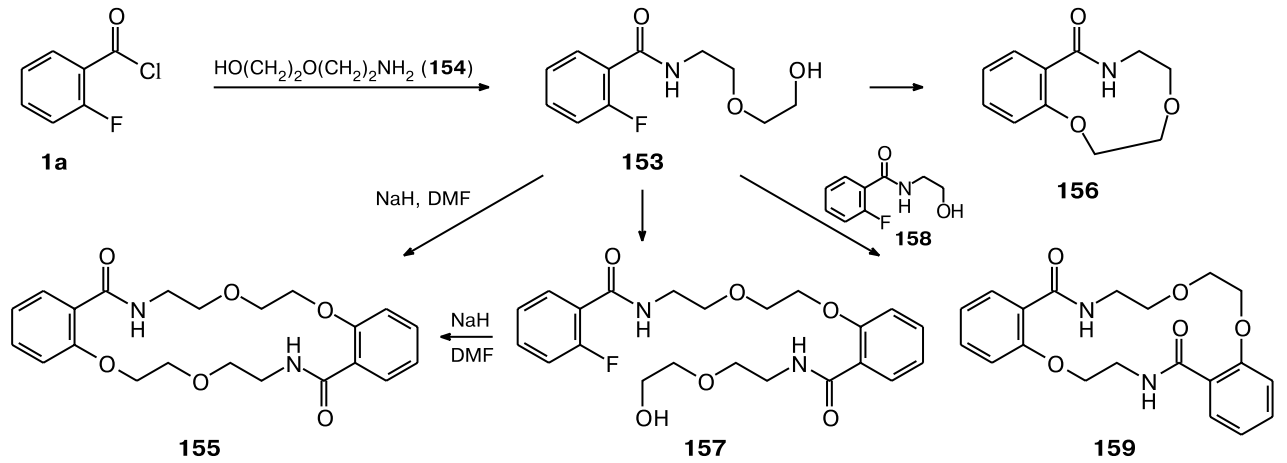


1,3-Cyclohexanedione **150** was also used as a 1,2-*C*,*O*-dinucleophile. The reaction of diketone **150** with 2-bromobenzoyl chloride **1c** gave enol ester **151**, whose free-radical intramolecular cyclization afforded ketospirolactone **152** (Scheme 34).[95]

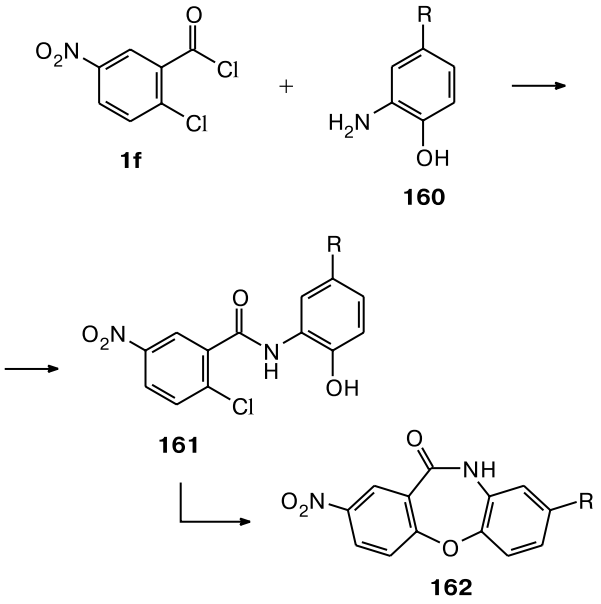
### 5. *N*,*O*-Dinucleophiles

Interesting compounds, including macrocyclic diamides, were prepared based on 2-fluorobenzoyl chloride **1a** and amino alcohols, which serve as 1,4- and 1,7-*N*,*O*-dinucleophiles (Scheme 35).[96,97] 2-Fluoro-*N*-(hydroxyalkyl)benzamide **153** was synthesized from **1a** and 2-(2-aminoethoxy)ethanol (**154**) in dichloromethane in the presence of aqueous sodium hydroxide. Treatment of a solution of amide **153** in DMF (2.0 mol $L^{-1}$) with

**Scheme 35**



**Scheme 36**

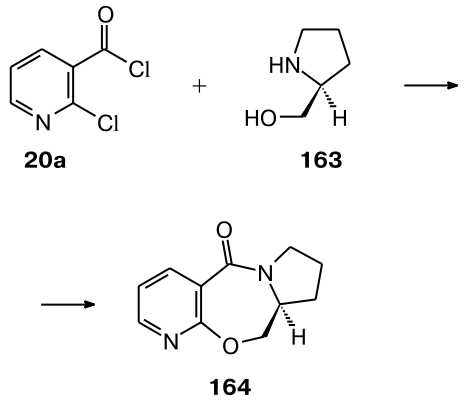



**160—162:** R = H (**a**), Me (**b**), Cl (**c**), OMe (**d**).

sodium hydride (4 equiv.) at 55 °C for 22 h afforded 9,10:19,20-dibenzo-1,4,11,14-tetraoxa-7,17-diazacycloeicosane-8,18-dione (**155**). In the case of a low concentration of reagent **153** (0.01 mol $L^{-1}$), the reaction at 55 °C produced (after 4 days) a mixture of compound **155** and 9,10-benzo-1,4-dioxa-7-azepin-8-one (**156**). Alcohol **157** was isolated under milder conditions (room temperature, 20 h). Condensation of an equimolar mixture of amide **153** and aroyl derivative **158**, which was derived from **1a** and 2-aminoethanol, in DMF in the presence of sodium hydride at room temperature afforded (after 4 days) 9,10:16,17-dibenzo-1,4,11-trioxa-7,14-diazacycloheptadecane-8,15-dione (**159**).[96]

*o*-Aminophenol derivatives **160** are typical 1,4-*N*,*O*-dinucleophiles. Cyclization of 2-chloro-5-nitrobenzamides of *o*-aminophenols **161** under alkaline conditions gave dibenzo[*b*,*f*][1,4]oxazepin-11(10*H*)-ones **162** (Scheme 36).[75]

**Scheme 37**




Pyrido[2,3-*f*]oxazepin-5-one **164** was synthesized starting from 2-chloronicotinoyl chloride **20a** and *S*-(+)-2-pyrrolidinemethanol **163**, which was also used as a 1,4-*N*,*O*-dinucleophile (Scheme 37).[98]

* * *

The above-considered examples of cyclization of 2-halogen derivatives of benzoyl chlorides with dinucleophiles provide evidence for a high synthetic potential of these reactions and demonstrate that these reactions can be used for the synthesis of various heterosystems. In particular, this approach is a good supplement to the known procedure for the construction of fluoroquinolones based on cyclization of polyfluorobenzoyl acrylates.[99—103]

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## References

1. G. A. Mokrushina, E. V. Nosova, G. N. Lipunova, and V. N. Charushin, *Zh. Org. Khim.*, 1999, **35**, 1447 [*Russ. J. Org. Chem.*, 1999, **35** (Engl. Transl.)].
2. G. N. Lipunova, E. V. Nosova, G. A. Mokrushina, L. P. Sidorova, and V. N. Charushin, *Khim.-farm. Zh.*, 2000, **34**, No. 1, 20 [*Pharm. Chem. J.*, 2000, **34**, No. 1 (Engl. Transl.)].
3. E. V. Nosova, G. N. Lipunova, L. P. Sidorova, and V. N. Charushin, *Khim.-farm. Zh.*, 2001, **35**, No. 11, 15 [*Pharm. Chem. J.*, 2001, **35**, No. 11 (Engl. Transl.)].
4. G. N. Lipunova, E. V. Nosova, M. A. Kravchenko, N. N. Mochul´skaya, L. P. Sidorova, E. V. Tsoi, G. A. Mokrushina, O. M. Chasovskikh, and V. N. Charushin, *Khim.-farm. Zh.*, 2004, **38**, No. 10—11 [*Pharm. Chem. J.*, 2004, **38**, No. 10—11 (Engl. Transl.)].
5. G. N. Lipunova, E. V. Nosova, L. P. Sidorova, and V. N. Charushin, *Khim.-farm. Zh.*, 2004—2005, in press [*Pharm. Chem. J.*, 2004—2005, in press (Engl. Transl.)].
6. D. T. W. Chu, P. B. Fernandes, and A. G. Pernet, *J. Med. Chem.*, 1986, **29**, 1531.
7. *Organofluorine Compounds in Medicinal Chemistry and Biomedical Applications*, Eds R. Filler, Y. Kobayashi, and L. M. Yagupolskii, Amsterdam, 1993, 480 pp.
8. F. S. Babichev, Yu. M. Volovenko, and A. G. Patratii, *Khim.-farm. Zh.*, 1989, **23**, No. 6, 695 [*Pharm. Chem. J.*, 1989, **23**, No. 6 (Engl. Transl.)].
9. M. J. Deetz, J. P. Malerich, A. M. Beatty, and B. D. Smith, *Tetrahedron Lett.*, 2001, 1851.
10. M. Taguchi, H. Kondo, Y. Inoue, Y. Kawahata, Y. Jinbo, F. Sakamoto, and G. Tsukamoto, *J. Med. Chem.*, 1992, **35**, 94.

11. S. Atarashi, H. Tsurumi, Y. Fujiwara, and I. Hayakawa, *J. Heterocycl. Chem*., 1991, **28**, 329.
12. S. Atarashi, S. Yokohama, K. Yamazaki, K. Sakano, M. Imamuro, and I. Hayakawa, *Chem. Pharm. Bull*., 1987, **35**, 1896.
13. Pat. 563734 Eur., 1993; *Chem. Abstr*., 1994, **120**, 134497x.
14. Pat. 563732 Eur., 1993; *Chem. Abstr*., 1994, **120**, 164227b.
15. Pat. 69339 Japan, 1992; *Chem. Abstr*., 1992, **117**, 137670x.
16. Pat. 424802 Eur., 1989; *Chem. Abstr*., 1991, **115**, 136090e.
17. Pat. 223289 Japan, 1991; *Chem. Abstr*., 1992, **116**, 39403a.
18. D. T. W. Chu, R. Hallas, J. J. Clement, J. J. Alder, E. McDonald, and J. J. Platner, *Drugs Expl. Clin. Res*., 1992, **18**, 275.
19. H. Kondo, M. Taguchi, Y. Inoue, F. Sakamoto, and G. Tsukamoto, *J. Med. Chem*., 1990, **33**, 2012.
20. Pat. 02802 South Africa, 1985; *Chem. Abstr*., 1987, **106**, 213930x.
21. Pat. 117388 Japan, 1990; *Chem. Abstr*., 1992, **117**, 111597d.
22. Y. Jinbo, M. Taguchi, H. Kondo, Y. Inoue, H. Tsujishita, Y. Kotera, F. Sakamoto, and G. Tsukamoto, *J. Med. Chem*., 1993, **36**, 2621.
23. D. T. W. Chu, P. B. Fernandes, A. K. Claiborne, L. Shen, and A. G. Pernet, *Drugs Expl. Clin. Res*., 1988, **14**, 379.
24. M. P. Wentland, G. Y. Lesher, M. Reuman, M. D. Gruett, B. Singh, S. C. Aldous, P. H. Dorff, J. B. Rake, and S. A. Coughlin, *J. Med. Chem*., 1993, **36**, 2801.
25. J. Nishimura, A. Minanida, and J. Matsumoto, *J. Heterocycl. Chem*., 1988, **25**, 479.
26. Pat. 126288 Japan, 1992; *Chem. Abstr*., 1992, **117**, 191858c.
27. Pat. 221383 Japan, 1992; *Chem. Abstr*., 1993, **118**, 169118k.
28. Pat. 208286 Japan, 1992; *Chem. Abstr*., 1993, **118**, 147546w.
29. Pat. 5318965 USA, 1994; *Chem. Abstr*., 1995, **122**, 160653f.
30. V. Ceccetti, A. Fravolini, R. Fringuelli, G. Mascellani, P. Pagella, M. Palmioli., G. Segre, and P. Terni, *J. Med. Chem*., 1993, **36**, 3449.
31. D. T. W. Chu and R. E. Maleszka, *J. Heterocycl. Chem*., 1987, **24**, 453.
32. D. T. W. Chu, R. Hallas, S. K. Tanaka, J. Alder, D. Balli, and J. Plattner, *Drugs Expl. Clin. Res*., 1994, **20**, 177.
33. D. Barret, H. Sasaki, H. Tsutsumi, M. Murato, T. Terasawa, and K. Sakane, *J. Org. Chem*., 1995, **60**, 3928.
34. Pat. 470578 Eur., 1992; *Chem. Abstr*., 1992, **117**, 7943c.
35. Pat. 228138 Japan, 1994; *Chem. Abstr*., 1995, **123**, 9454q.
36. Y. Jinbo, H. Kondo, M. Taguchi, Y. Inoue, F. Sakamoto, and G. Tsukamoto, *J. Med. Chem*., 1994, **37**, 2791.
37. Yu. M. Volovenko, A. G. Nemazanyi, V. A. Shevchenko, and F. S. Babichev, *Dokl. Akad. Nauk Ukr. SSR* [*Reports Ukr. Acad. Sci.*], 1983, No. 9, 27 (in Russian).
38. Yu. M. Volovenko, A. G. Nemazanyi, I. G. Ryabokon´, and F. S. Babichev, *Ukr. Khim. Zz*. [*Ukr. Chem. J*.], 1988, **54**, 295 (in Russian).
39. F. S. Babichev, V. K. Patratii, Yu. M. Volovenko, N. G. Prodanchuk, V. G. Sinchenko, O. G. Nemazani, and T. A. Silaeva, *Ukr. Pharm. Zh*., 1988, 65; *Chem. Abstr*., 1989, **110**, 170097u.
40. USSR Pat. 1414849, 1988; *Chem. Abstr*., 1989, **110**, 8224m.
41. F. S. Babichev, Yu. M. Volovenko, A. G. Nemazanyi, and S. I. Tyukhtenko, *Ukr. Khim. Zz*. [*Ukr. Chem. J*.], 1986, **52**, 506 (in Russian).
42. F. S. Babichev, Yu. M. Volovenko, and A. G. Nemazanyi, *Dokl. Akad. Nauk Ukr. SSR* [*Reports Ukr. Acad. Sci.*], 1984, No. 5, 17 (in Russian).
43. Yu. M. Volovenko, A. G. Nemazanyi, G. L. Vesel´skaya, S. I. Tyukhtenko, and F. S. Babichev, *Dokl. Akad. Nauk Ukr. SSR* [*Reports Ukr. Acad. Sci.*], 1986, No. 12, 30 (in Russian).
44. G. N. Lipunova, E. V. Nosova, P. V. Vasil´eva, and V. N. Charushin, *Izv. Akad. Nauk, Ser. Khim*., 2003, 436 [*Russ. Chem. Bull., Int. Ed*., 2003, **52**, 457].
45. M. Vales, V. Lokshin, G. Pepe, R. Guglielmetti, and A. Samat, *Tetrahedron*, 2002, **58**, 8543.
46. J. Grimshaw and A. P. DeSilva, *J. Chem. Soc., Perkin Trans. 2*, 1982, 857.
47. J. Grimshaw and D. Mannus, *J. Chem. Soc., Perkin Trans. 1*, 1977, 2456.
48. T. Harayama, T. Akiyama, and Y. Nakano, *Chem. Pharm. Bull*., 1997, **45**, 1723.
49. T. Harayama, H. Toko, H. Nishioka, H. Abe, and Y. Takeuchi, *Heterocycles*, 2003, **59**, 541.
50. B. S. Thyagarajan, N. Kharasch, H. B. Lewis, and W. Wolf, *Chem. Commun*., 1967, 614.
51. A. Mondon and K. Krohn, *Chem. Ber*., 1972, **105**, 3726.
52. S. Lalitha, S. Rajewsari, B. R. Pai, and H. Sugura, *Indian J. Chem*., 1977, **15B**, 186.
53. Pat. 7877067 Japan, 1978; *Chem. Abstr*., 1978, **89**, 163430b.
54. J. Grimshaw and A. P. DeSilva, *J. Chem. Soc., Chem. Commun*., 1980, 302.
55. H. Hara, O. Hoshino, and B. Umezawa, *Tetrahedron Lett*., 1972, 5031.
56. W. Carruthers and N. Evans, *J. Chem. Soc., Perkin Trans. 1*, 1974, 1523.
57. R. K. Y. Zee-Chung, S. J. Yan, and C. C. Cheng, *J. Med. Chem*., 1978, **21**, 199.
58. G. R. Lenz, *Tetrahedron Lett*., 1973, 1963.
59. Y. Kanaoka and K. Itoh, *J. Chem. Soc., Chem. Commun*., 1973, 647.
60. G. R. Lenz, *J. Org. Chem*., 1974, **39**, 2839.
61. Z. Horii, Y. Nakashita, K. Kunisawa, and C. Iwata, *Chem. Pharm. Bull*., 1973, **21**, 2879.
62. A. F. Pozharskii and G. G. Yurchuk, *Khim. Geterotsikl. Soedin*., 1981, 838 [*Chem. Heterocycl. Compd.,* 1981 (Engl. Transl.)].
63. Int. Patent 0341653, 2003; *Chem. Abstr*., 2003, **138**, 401770e.
64. Int. Patent 0341660, 2003; *Chem. Abstr*., 2003, **138**, 401771f.
65. S. K. Singh, A. L. Ruchelman, T. K. Li, A. Liu, L. F. Liu, and E. J. Lavoie, *J. Med. Chem*., 2003, **46**, 2254.
66. M. Ikeda, S. Akamatsu, Y. Kugo, and T. Sato, *Heterocycles*, 1996, **42**, 155.
67. A. A. G. Faraco, M. A. F. Prado, R. J. Alves, F. Souza, D. Jose, R. B. Alves, and R. F. P. Faraco, *Synt. Commun*., 2003, **33**, 463.
68. G. Timari, G. Hajos, and A. Messmer, *J. Heterocycl. Chem*., 1990, **27**, 2005.
69. L. W. Deady, M. F. Mackay, and D. M. Werden, *J. Heterocycl. Chem*., 1989, **26**, 161.
70. E. V. Nosova, G. N. Lipunova, M. I. Kodess, P. V. Vasil´eva, and V. N. Charushin, *Izv. Akad. Nauk, Ser. Khim*., 2004, No. 9 [*Russ. Chem. Bull., Int. Ed*., 2004, **53**, No. 9].

71. I. I. Popov, S. L. Boroshko, B. A. Tertov, and E. V. Tyukavina, *Khim. Geterotsikl. Soedin.*, 1989, 272 [*Chem. Heterocycl. Compd.,* 1989 (Engl. Transl.)].
72. D. H. Kim, *J. Heterocycl. Chem.*, 1981, **18**, 801.
73. D. H. Kim, *J. Heterocycl. Chem.*, 1981, **18**, 287.
74. G. N. Lipunova, E. V. Nosova, A. A. Laeva, M. I. Kodess, and V. N. Charushin, *Zh. Org. Khim.*, 2004, **40**, No. 10 [*Russ. J. Org. Chem.*, 2004, No. 10, **40** (Engl. Transl.)].
75. K. Nagarajan, A. Venkateswarlu, C. L. Kulkarni, and R. K. Shah, *Indian J. Chem.*, 1974, **12**, 227.
76. E. V. Nosova, G. N. Lipunova, M. A. Kravchenko, and V. N. Charushin, *Kislorod- i serusoderzhashchie geterotsikly* [*Oxygen- and Sulfur-Containing Heterocycles*], Ed. V. G. Kartsev, IBS PRESS, Moscow, 2003, **2**, 163 (in Russian).
77. D. Koscik, P. Kristian, and J. Gonda, *Collect. Czech. Chem. Commun.*, 1983, **48**, 3320.
78. D. Koscik, P. Kutschy, and M. Dzurilla, *Chem. Pap.*, 1992, **46**, 405.
79. G. N. Lipunova, E. V. Nosova, G. A. Mokrushina, E. G. Ogloblina, G. G. Aleksandrov, and V. N. Charushin, *Zh. Org. Khim.*, 2003, **39**, 270 [*Russ. J. Org. Chem.*, 2003, **39** (Engl. Transl.)].
80. W. R. Dolbier, C. Burkholder, K. A. Abboud, and D. Loehle, *J. Org. Chem.*, 1994, **59**, 7688.
81. Yu. M. Volovenko, F. S. Babichev, S. V. Litoinenko, and A. V. Turov, *Otkrytiya, Izobreteniya* [*Discoveries, Inventions*], 1988, 108 (in Russian).
82. Y. V. Nageswar, M. S. Murty, and T. H. Ramalingam, *Indian J. Chem.*, 1988, **27B**, 1142.
83. N. P. Peet, J. Malecha, M. E. LeTourneau, and S. Sunder, *J. Heterocycl. Chem.*, 1989, **26**, 257.
84. Pat. 4871732 USA, 1989; *Chem. Abstr.*, 1990, **112**, 179036x.
85. W. Doeling, M. Biedermann, and H. Hartung, *Eur. J. Org. Chem.*, 1998, 1237.
86. H. H. Vorozhtsov, V. A. Barkhash, A. T. Prudchenko, and T. I. Khomenko, *Dokl. Akad. Nauk SSSR,* 1965, **164**, 1046 [*Dokl. Chem.*, 1965 (Engl. Transl.)].
87. N. N. Vorozhtsov, V. A. Barkhash, A. T. Prudchenko, and T. I. Khomenko, *Zh. Obshch. Khim.*, 1964, **34**, 1501 [*J. Gen. Chem. USSR*, 1964, **34** (Engl. Transl.)].
88. V. I. Saloutin, Ya. V. Burgart, and O. N. Chupakhin, *Ftorsoderzhashchie trikarbonil´nye soedineniya* [*Fluorine-Containing Tricarbonyl Compounds*], Izd-vo UrO RAN, Ekaterinburg, 2002, 242 pp. (in Russian).
89. G. M. Coppola and R. W. Dodsworth, *Synthesis*, 1981, 523.
90. S. P. Kisil, Ya. V. Burgart, V. I. Saloutin, and O. N. Chupakhin, *J. Fluorine Chem.*, 2001, **108**, 125.
91. G. N. Lipunova, E. V. Nosova, M. I. Kodess, and V. N. Charushin, *Zh. Org. Khim.*, 2004, **40**, No. 10 [*Russ. J. Org. Chem.*, 2004, **40** (Engl. Transl.)].
92. J. Sotiropoulos, N. E. Batuoti, and A. N. Lamazoure, *J. Heterocycl. Chem.*, 1987, **24**, 907.
93. C. D. Gabbutt, J. D. Hepworth, and B. M. Heron, *J. Chem. Soc., Perkin Trans. 1*, 1992, 3015.
94. W. Zhang and G. Pugh, *Tetrahedron Lett.*, 2001, **42**, 5613.
95. W. Zhang and G. Pugh, *Tetrahedron Lett.*, 1999, **40**, 7595.
96. A. G. Schultz, M. S. Smyth, and F. S. Tham, *J. Org. Chem.*, 1991, **56**, 6952.
97. A. G. Schultz, D. J. Pinto, M. Welch, and R. K. Kullnig, *J. Org. Chem.*, 1988, **53**, 1372.
98. Int. Patent 02102808, 2002; *Chem. Abstr.*, 2003, **138**, 39304v.
99. G. N. Lipunova, E. V. Nosova, N. N. Mochul´skaya, A. A. Andreiko, O. M. Chasovskikh, and V. N. Charushin, *Izv. Akad. Nauk, Ser. Khim.*, 2002, 613 [*Russ. Chem. Bull., Int. Ed.*, 2002, **51**, 663].
100. V. N. Charushin, G. N. Lipunova, E. V. Nosova, L. P. Sidorova, and O. M. Chasovskikh, *Mendeleev Commun.*, 1998, 131.
101. G. N. Lipunova, G. A. Mokrushina, E. V. Nosova, L. I. Rusinova, and V. N. Charushin, *Mendeleev Commun.*, 1997, 109.
102. G. N. Lipunova, L. P. Sidorova, E. V. Nosova, N. M. Perova, V. N. Charushin, and G. G. Aleksandrov, *Zh. Org. Khim.*, 1999, **35**, 1729 [*Russ. J. Org. Chem.*, 1999, **35** (Engl. Transl.)].
103. E. V. Nosova, G. N. Lipunova, G. A. Mokrushina, O. M. Chasovskikh, L. I. Rusinova, V. N. Charushin, and G. G. Aleksandrov, *Zh. Org. Khim.*, 1998, **34**, 436 [*Russ. J. Org. Chem.*, 1998, **34** (Engl. Transl.)].

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