SYNTHETIC ROUTES TO P(O)-SUBSTITUTED FIVE-MEMBERED AROMATIC HETEROCYCLES (2010-2021)

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Abstract. *Phosphoryl-substituted heterocycles include a large variety of advanced materials, useful reactive intermediates for organic synthesis, natural compounds, and biologically active pharmaceuticals. In the past decade, significant advances have been made in the synthesis of phosphoryl-substituted five-membered heterocycles. This review covers the synthetic strategies towards monocyclic N,O,S-aromatic heterocycles, such as pyrroles, pyrazoles, imidazoles, 1,2,3-triazoles, isoxazoles, oxazoles, and thiazoles, bearing a phosphoryl substituent, reported from 2010 to 2021.*

Contents

1. Introduction

Phosphorylated heterocycles represent structural motifs found in a great number of biologically active synthetic and natural compounds, including agrochemicals and drugs.[1] There are numerous reports on the use of phosphorylated heterocycles as ligands and Lewis acids in metal-catalyzed organic reactions.[2-4]

Moreover, they are widely used in materials chemistry for the synthesis of polymeric materials and dyes with outstanding properties.[5]

Some phosphoryl-substituted monocyclic five-membered heterocycles exhibit efficient anti-inflammatory, neuroprotective, and antitumor activity. In particular, phosphoryl-substituted pyrroles were used to design analgesic,[6] thyromimetic,[7] and anti-inflammatory[8] agents. Phosphoryl-modified pyrazoles are promising candidates as pesticides,[9-10] haptens,[11-13] phosphatidylinositol phosphate kinase inhibitors,[14-15] and anticancer drugs.[16] Phosphoryl-substituted imidazoles serve as 1,6-bisphosphatase inhibitors[17] and show antiproliferative activity.[18] Phosphoryl-substituted oxazoles are used as 1,6-bisphosphatase inhibitors.[19-20] Phosphoryl-substituted 1,2,3-triazoles are of considerable interest as stable phosphohistidine analogues,[21-24] phosphoribosyltransferase inhibitors,[25] and ligands for lanthanide metals.[26]

A variety of highly efficient methodologies for the synthesis of phosphorylated aromatic heterocycles and their derivatives have been reported in the past; meanwhile, the development of novel methodologies is in continuous demand. Of particular interest are methods that offer a broad substrate scope and give products with high atom economy, molecular complexity, and great efficiency under mild conditions.[27-30] In general, the known methods for the synthesis of phosphoryl-substituted aromatic heterocycle are classified into two synthetic strategies: 1) the construction of the heterocyclic core using phosphoryl-containing reagents and 2) phosphorylation of the pre-synthesized heterocyclic core.

Several great reviews dealing with the synthesis of P(O)-substituted heterocyclic compounds have been published in the literature in recent years. Many of them highlighted the phosphorylation as the most common approach[31-32] and specifically the radical phosphorylation,[33] metal-catalyzed/promoted C-H bond phosphorylation.[34] Another array of reviews described the use of a specific kind of transformations, including photoredox catalyzed reactions,[35] electrocatalytic[36] and radical[37] C-P bond formation, multicomponent reactions,[38] and functionalization of phosphorus-centered radicals.[39] Some reviews are focused on the type of P-functional group [40] or on the type of heterocycles, see *e.g.* phosphorylindoles,[41-42] pyrazoles,[43-44] and pyrrolidines.[45] However, systematic reviews of the published data on different methods for the synthesis of most of phosphorylated monocyclic five-membered *N,O,S*-heterocycles are lacking.

The main focus of this review is monocyclic five-membered aromatic phosphoryl-substituted *N,O,S*-heterocycles, including pyrroles, pyrazoles, imidazoles, 1,2,3-triazoles, isoxazoles, oxazoles, and thiazoles. The review is an attempt to systematically consider and classify methods for the synthesis of these compounds. It is organized in a classical manner and includes the classification according to the type of *N*-heterocycles using the following criteria: a) the number of heteroatoms, b) the nature of synthetic precursors, and c) the type of chemical transformations involving these heterocycles. In many cases, the reaction mechanisms are briefly discussed in order to more completely describe the synthetic approaches to phosphoryl-substituted *N*-heterocycles. The review focuses on the scientific advances made over the past decade, in the period from 2010 up to the end of 2021. However, some earlier, pioneering studies are cited if necessary.

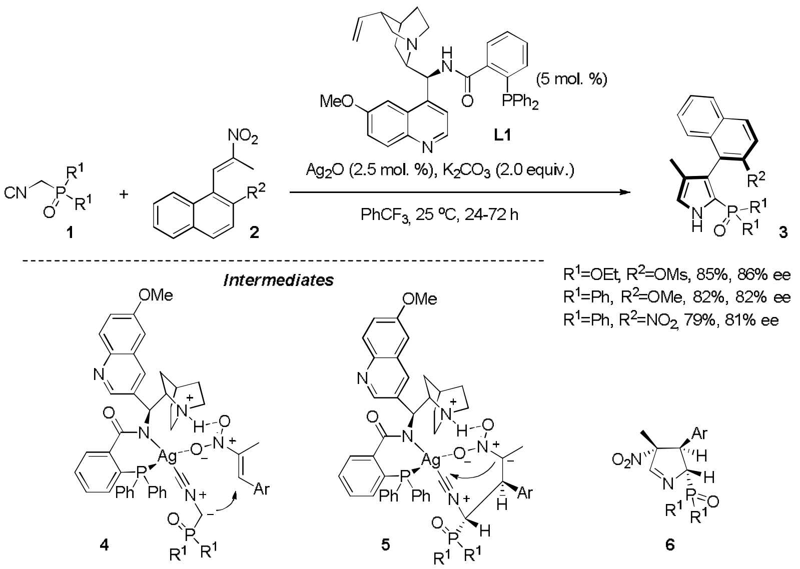
2. Pyrroles

The synthesis of phosphoryl-substituted pyrroles was first reported in 1932 by Mingoia, who used the reaction of pyrrolyl-1-magnesium bromide with $POCl_3$.[46] More recently, the reaction of metalated pyrroles with compounds containing a phosphorus−halogen bond has gained popularity as a method for the preparation of these compounds.[47] Other historical approaches are based on the reaction of 2-chloro-2-phosphorylethylidene hydrazines with 1,3-dicarbonyl compounds in the presence of bases[48-49] and the reaction between (isocyanomethyl)phosphonates or β-ketophosphonates and Michael acceptors.[50-53] It is worth noting that examples of the application of such classical methods as the Knorr and Hantzsch reactions for the synthesis of phosphorylated pyrroles are unknown. This is apparently because of the difficulties in the preparation of phosphorylated reagents required for the synthesis within the framework of these approaches.

Recent examples of the synthesis of phosphorylated pyrroles, which include the formal [3+2]- and [4+1]-cycloaddition, intramolecular cyclization, and oxidative phosphorylation, are summarized below.

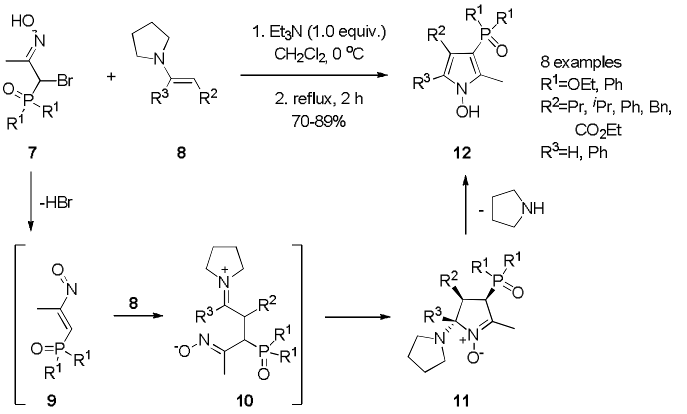
2.1. Formal [3+2]-cycloaddition

He *et al.*[54] demonstrated that 3-(naphthalen-1-yl)pyrrolyl-3-phosphonates can be synthesized by the Barton-Zard reaction (Scheme 1). Nitroolefins **2** bearing a β-*ortho*-substituted naphthyl group reacted with phosphorylated α-isocyano substrates **1** under the catalysis of Ag_2O. This approach allows the preparation of rotationally stable molecules with axial chirality. The use of the chiral catalyst **L1** provided the synthesis of pyrroles **3** with a good level of asymmetric induction (up to 81% ee). The authors suggested that the reaction proceeds within complexes **4** and **5** between the silver atom, the catalyst, and both reagents and involves two successive nucleophilic attacks, resulting in the pyrrole ring closure. Non-aromatic intermediate **6** eliminates nitric acid giving the target product.



Scheme 1. Synthesis of pyrrole-containing compounds with axial chirality by the Barton-Zard reaction.

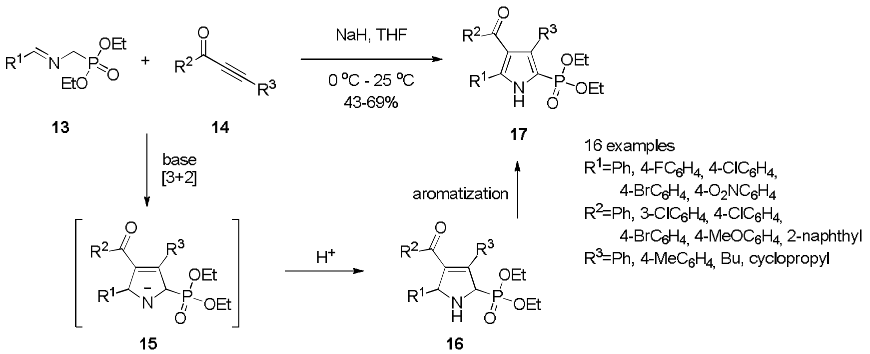
Palacios and co-workers[55] reported the synthesis of *N*-hydroxypyrroles from phosphorylated 1,2-oxazabuta-1,3-dienes and enamines (Scheme 2).



Scheme 2. Synthesis of *N*-hydroxypyrroles.

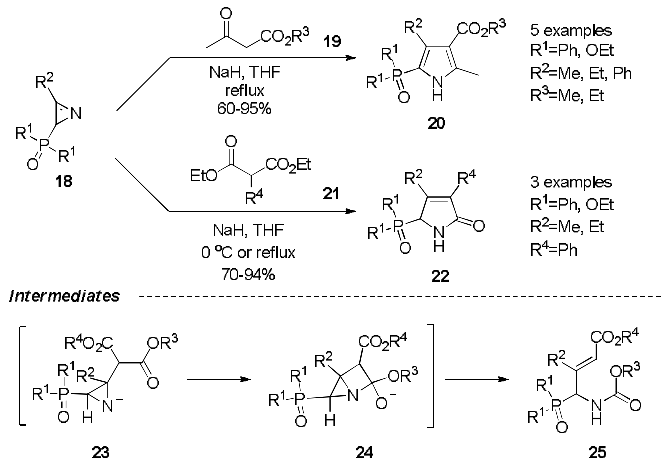
The authors suggested that the reaction occurs as the conjugate addition of enamine **8** to the diene system of nitrosoalkene **9** generated *in situ* from bromide **7** followed by the intermolecular ring closure of nitrosoimine **10** into cyclic *N*-oxide **11** and the elimination of pyrrolidine from intermediate **11** to form pyrrolyl-3-phosphonates **12**. The evaluation of the reaction scope revealed that various alkyl- and aryl-substituted enamines **9** were tolerated. The yields of products **12** varied from 70% to 89% (Scheme 2).

Meng *et al.*[56] synthesized pyrrolyl-2-phosphonates by the formal [3+2]-cycloaddition of phosphonate azomethine ylides with alkynyl ketones (Scheme 3). Initially, the reaction of substituted [(methylideneamino)methyl]phosphonate **13** with a base affords azomethine ylide followed by the [3+2]-cycloaddition of the latter to alkynyl ketone **14**. The resulting dihydropyrrole anion **15** is protonated giving intermediate **16**, which undergoes oxidative aromatization to form pyrrole **17**. A variety of electron-withdrawing and electron-donating aryl-substituted alkynyl ketones **14** successfully participated in the reaction giving the corresponding pyrroles **17** in good yields. This reaction can be accomplished also in a three-component fashion involving aromatic aldehyde, aminomethylphosphonate, and ynone with a slight decrease in the yield of the target heterocycle.



Scheme 3. [3+2]-Cycloaddition of phosphonate azomethine ylides to alkynyl ketones.

Palacios and co-workers[57-58] synthesized phosphorylated pyrroles starting from *2H*-azirines **18** and enolates of β-dicarbonyl compounds (Scheme 4). It was shown that this approach allows the preparation of 2-phosphorylpyrroles **20** and 5-phosphoryl-1,5-dihydro-*2H*-pyrrol-2-ones **22** depending on the starting dicarbonyl compounds, β-keto esters **19**, or malonic acid esters **21**, respectively.

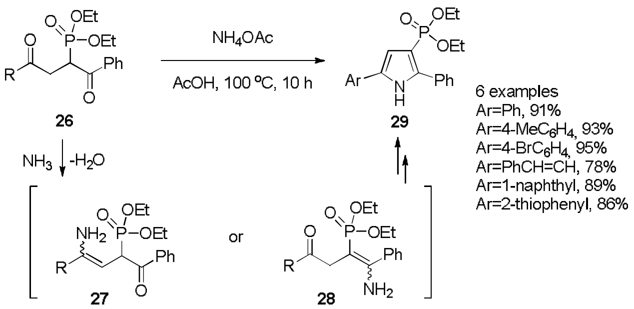


Scheme 4. Synthesis of phosphorylpyrroles from *2H*-azirines.

The proposed mechanism of the reaction with acetoacetic esters involves the attack of the dicarbonyl anion on 2*H*-azirine **18** followed by the intramolecular cyclization of intermediate **23** to substituted 1-azabicyclo[2.1.0]pentane **24**. This intermediate undergoes a bond cleavage cascade giving unsaturated phosphorylated amide **25** and its subsequent base-mediated cyclization affords pyrrole **20** or pyrrolinone **22**. For other dicarbonyl compounds, the reaction mechanism is similar and differs only by the carbonyl group susceptible to intramolecular nucleophilic attack. Notably, this reaction with cyclic ethyl 2-oxocyclopentanecarboxylate[58] can be used to prepare 2-phosphoryl-1,4,5,6-tetrahydrocyclopenta[*b*]pyrroles (Scheme 4).

2.2. Formal [4+1]-cycloaddition

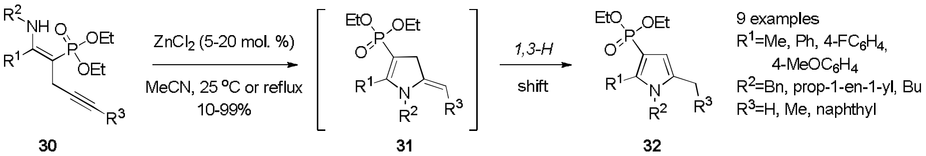
Recently, Tiwari *et al.*[59] elaborated an efficient synthesis of *α*-phosphorylated 1,4-diketones **26** and demonstrated that the latter compounds can react with ammonium acetate to form 3-phosphorylated pyrroles (Scheme 5). A plausible mechanism for this transformation begins with the formation of the enaminoketones **27**/**28**, which undergo cyclization to dihydropyrroles followed by the isomerization of the latter to pyrroles. A variety of groups in 1,4-diketone **26**, including aryl, vinylaryl, and hetaryl, were tolerated leading to products **29** in good to excellent yields.



Scheme 5. Synthesis of 3-phosphorylated pyrroles from α-phosphorylated 1,4-diketones.

2.3. Cyclization reactions

Stevens and co-workers[60] demonstrated that pyrrolyl-3-phosphonates **32** can be synthesized by the Zn(II)-catalyzed intramolecular hydroamination of *β*-phosphonato-*β*-propargyl enamines **30** (Scheme 6). The proposed mechanism involves the regioselective *5-exo-dig* cyclization of compound **30** generating 2-methylene-2,3-dihydropyrrole **31**, which undergoes 1,3-*H* shift to pyrrole **32**. Notably, nonterminal alkynes participated in the reaction in lower yields.

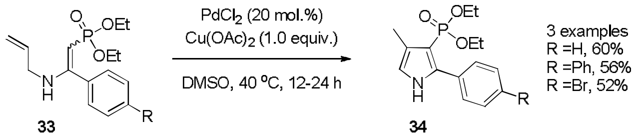


Scheme 6. Intramolecular hydroamination in the synthesis of pyrroles.

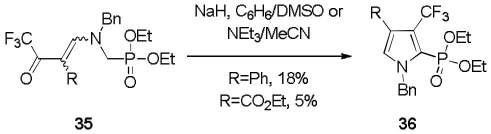
Mishra and Deb disclosed the Pd(II)-catalyzed oxidative cyclization approach to pyrrolyl-3-phosphonates **34** starting from *β*-ketophosphonates **33** bearing 4-phenylaryl and 4-bromoaryl substituents (Scheme 7).[61] The heterocyclizations were performed using 20 mol% of $PdCl_2$ and a stoichiometric amount of $Cu(OAc)_2$. The reaction is believed to occur as the oxidative addition of the enamine C–H bond to Pd(II) followed by palladation of the second C–H bond and reductive elimination of the resulting pyrrole.

The nucleophilic 5-*exo-trig* cyclization of *N*-methylphosphonatoenaminoketones **35** can also be used to form the phosphoryl-substituted pyrrole ring (Scheme 8).[62] This approach allows the synthesis of

3-trifluoromethylpyrrolyl-2-phosphonates **36** in reasonable yields. The heterocyclizations were accomplished using sodium hydride and triethyamine to give products in reasonable yields.



Scheme 7. Cyclization of *N*-allylenaminophosphonates.

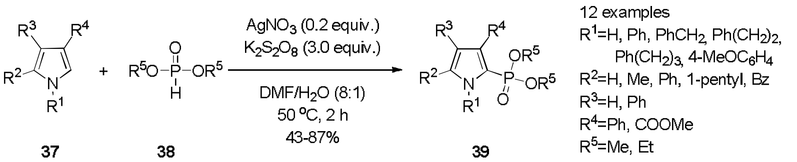


Scheme 8. Synthesis of 3-trifluoromethylpyrrolyl-2-phosphonates.

2.4. Phosphorylation of pyrroles

Recently, Kim and co-workers[63] reported the synthesis of pyrrolyl-2-phosphonates **39** *via* oxidative phosphorylation of pyrroles in the presence of the $AgNO_3$-$K_2S_2O_8$ system (Scheme 9). 1,3-Diphenylpyrroles **37** reacted smoothly with phosphonates **38** to form products **39** in good yields, while the yields were low in the case of 1-alkyl-substituted pyrroles.

Besides, examples are known[64-66] when such systems as I_2-$K_2S_2O_8$, as well as $AgNO_3$, $Mn(OAc)_3$, or (*tert*-BuO)$_2$ (in the case of *N*-sulfonyl-substituted pyrroles), are used.



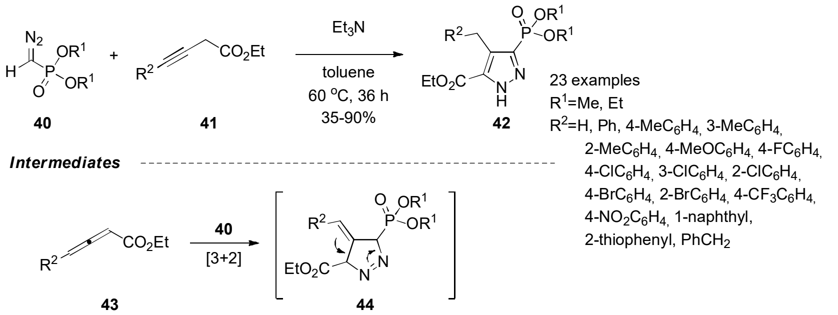
Scheme 9. Oxidative phosphorylation of pyrroles in the presence of $AgNO_3$ and $K_2S_2O_8$.

3. Pyrazoles

The synthesis of phosphorylated pyrazoles was first reported in 1899 by Michaelis and Pasternachk,[67] who used the reaction of 2,4-dihydro-3*H*-pyrazol-3-ones with $POCl_3$. Currently, the main synthetic approaches to 3(5)-phosphoryl-substituted pyrazoles are based on the electrophilic and transition metal-catalyzed phosphorylation of pyrazoles, the [3+2]-cycloaddition of diazo compounds to phosphorylated dienophiles, and the [3+2]-cycloaddition of the Ohira-Bestmann reagent to different dienophiles. These results are considered in the review by Beletskaya and co-workers published in 2016.[68] Recent examples of the synthesis of phosphorylated pyrazoles, which are not included in the mentioned review, are given below.

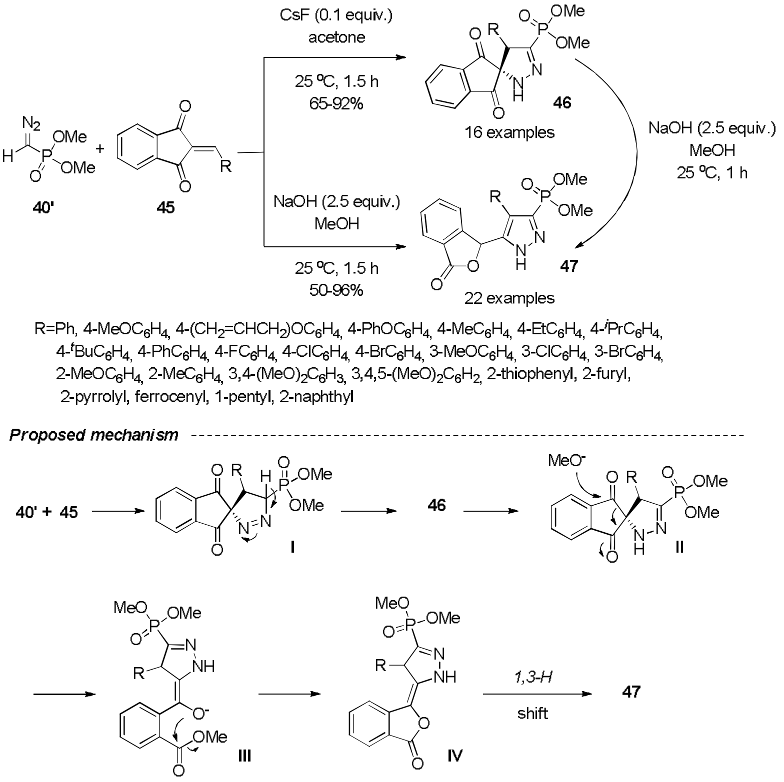
3.1. Formal [3+2]-cycloaddition

Recently, Tian *et al.*[69] reported that Seyferth-Gilbert reagents **40** reacted with 3-alkynoates **41** on warming in the presence of triethylamine to furnish 2-phosphorylated pyrazoles **42** (Scheme 10). The reaction has a broad scope since various 3-alkynoates can be involved in this reaction. The methodology was also extended to allenoates **43**. The proposed mechanism involves the Et_3N-promoted tautomerization of 3-alkynoates **41** to the corresponding allenoates **43** followed by the [3+2]-cycloaddition of the latter to Seyferth-Gilbert reagent **40** to form cycloadduct **44**, which undergoes isomerization to the final pyrazole product **42** *via* a 1,3-hydrogen shift.



Scheme 10. Formal [3+2] -cycloaddition of Seyferth-Gilbert reagents to 3-alkynoates.

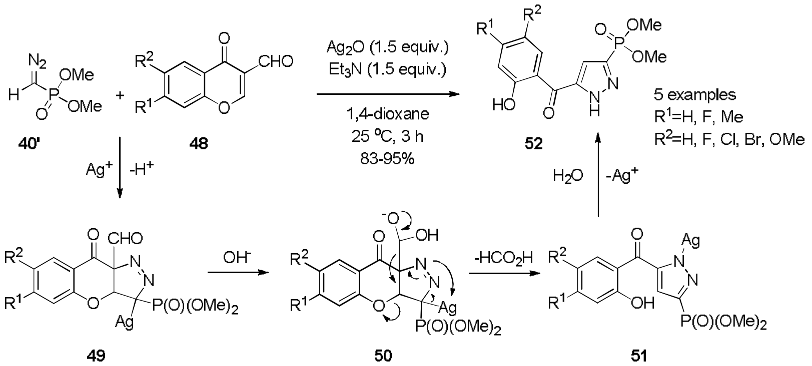
Mohanan and co-workers[70] studied the reaction of 2-arylideneindane-1,3-diones **45** with Seyferth-Gilbert reagent **40'** and found that the reaction pathway depends on the reaction conditions (Scheme 11). The reaction using CsF as the base in acetone affords phosphoryl-substituted spiropyrazoline **46**. However, the reaction with NaOH as the base in methanol gives phosphorylated 3-pyrazolylphthalides **47** as the major products. It was demonstrated that spiropyrazolines **46** are rearranged into phthalides **47** in the presence of a methanol solution of NaOH. A variety of electron-withdrawing and electron-donating aryl-substituted indane-1,3-diones successfully participated in the reaction. Interestingly, 2-thiophenyl-, 2-furyl-, 2-pyrrolyl-, and ferrocenyl-substituted indane-1,3-diones were also tolerated in the reaction.



Scheme 11. Reaction of 2-arylideneindane-1,3-diones with the Seyferth-Gilbert reagent.

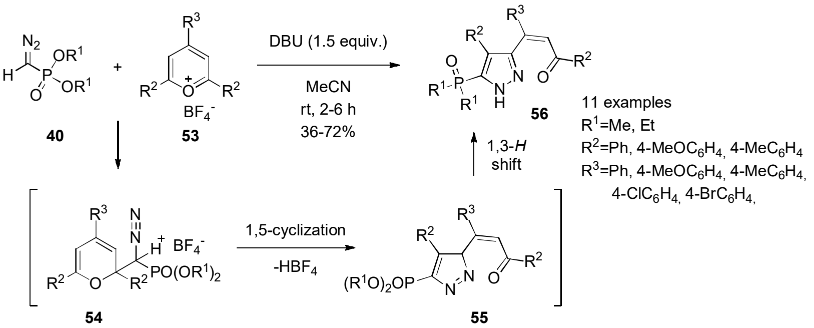
The proposed mechanism of heterocyclization involves the [3+2]-cycloaddition of Seyferth-Gilbert reagent **40'** to alkene **45** to form intermediate **I**, which generates spiropyrazoline **46** through the tautomerization. The subsequent transformation of compound **46** into pyrazole was believed to occur as a sequence of the five-membered ring opening *via* the nucleophilic attack of the methoxide anion on the carbonyl group of indanedione **II** giving intermediate **III**, the ring reclosure of the latter into **IV**, and its isomerization to product **47** (Scheme 11).

Lan, Yang, and co-workers[71] synthesized pyrazolyl-3-phosphonates **52** by the reaction between Seyferth-Gilbert reagent **40'** and 3-formylchromones **48** (Scheme 12). The reaction is characterized by high yields. The proposed mechanism of the formation of pyrazoles **52** involves the [3+2]-cycloaddition of the silver(I) complex with the Seyferth-Gilbert reagent to 3-formylchromone **49** giving tricyclic intermediate **50**, which undergoes the rearrangement accompanied by the C–O and C–Ag bond cleavage and elimination of formic acid to form compound **51**. In the final step, the resulting silver derivative of phenyl pyrazolyl ketone **51** is hydrolyzed to final product **52**.



Scheme 12. Reaction of the Seyferth-Gilbert reagent with 3-formylchromones.

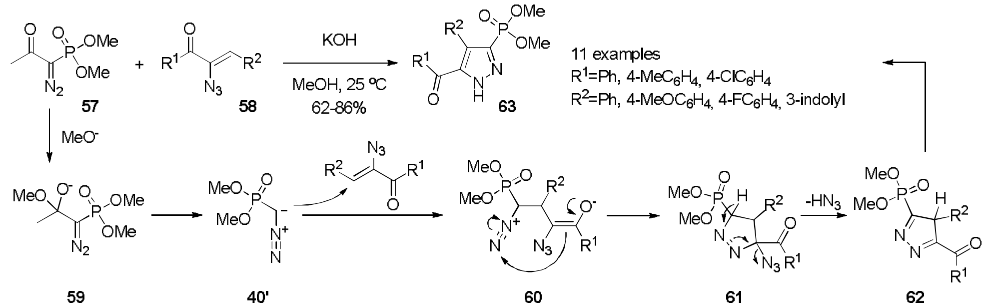
Recently, Shukla, Rastogi, and co-workers[72] showed that the reaction of Seyferth-Gilbert reagent **40** with 2,4,6-triarylpyrylium tetrafluoroborates **53** in the presence of a strong base, resulting in the formation of phosphorylated pyrazole chalcones **56** (Scheme 13). The heterocyclization is believed to occur through the initial insertion of the diazo compound at the α-position of the pyrylium ring to form intermediate **54**, which undergoes the cascade ring opening/ring closure giving pyrazole intermediate **55**.



Scheme 13. Reaction of the Seyferth-Gilbert reagent with pyrylium salts.

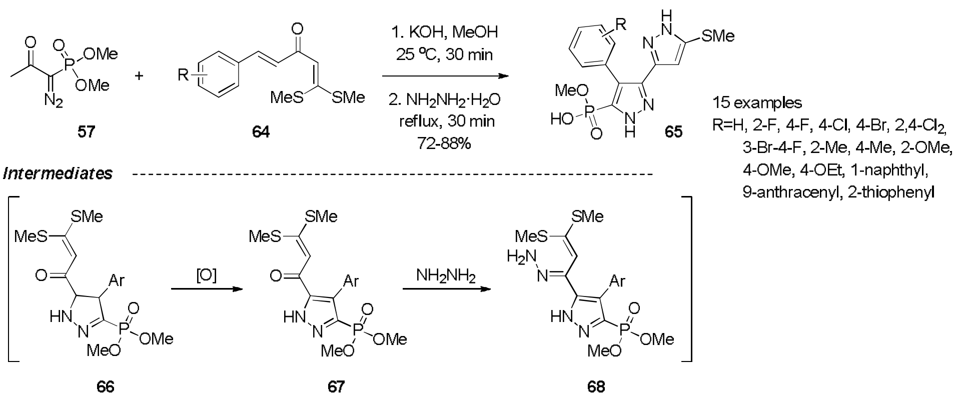
Madugula *et al.*[73] used vinyl azides **58** as alkenes in the reaction with Ohira-Bestmann reagent **57** giving 5-acylpyrazolyl-3-phosphonates **63** (Scheme 14). In this case, the azide anion acts as a leaving group in the aromatization of dihydropyrazole. The proposed reaction mechanism involves the activation of

Ohira-Bestmann reagent **57** in the presence of the methoxide anion to form intermediate **59**. The resulting Seyferth-Gilbert reagent **40'** undergoes the Michael addition to vinyl azide **58** to give zwitterionic intermediate **60**, which is cyclized to dihydropyrazole **61** followed by the elimination of HN_3 to form pyrazole **62** and isomerization of the latter to give the final product **63**. The reaction is general with respect to aryl-substituted keto vinyl azides **58** giving products in yields from good to high.



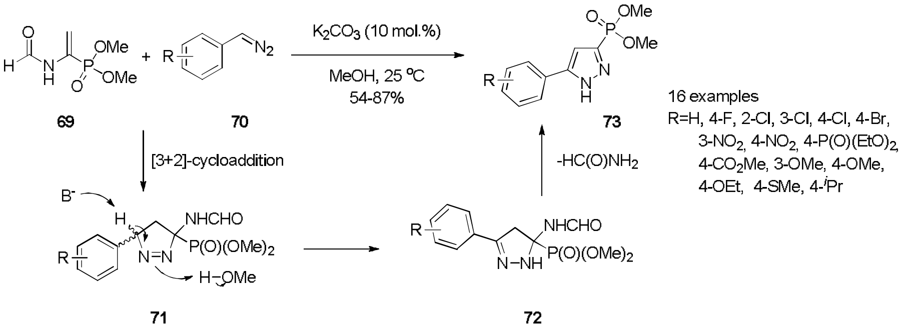
Scheme 14. Reaction of the Ohira-Bestmann reagent with vinyl azides.

Shanmugam and co-workers[74] described an interesting approach to the synthesis of 3-(pyrazol-3-yl)pyrazolyl-5-phosphonates **65** based on the coupling of α-aroylidineketene dithioacetals **64** with Ohira-Bestmann reagent **57** and hydrazine (Scheme 15). A variety of electron-withdrawing and electron-donating aryl-substituted bis-vinyl ketones **64** successfully participated in the reaction providing the corresponding pyrazoles **65** in good yields. In this reaction, the phosphonate moiety undergoes selective monodemethylation. The reaction mechanism involves the transformation of Ohira-Bestmann reagent **57** into Seyferth-Gilbert reagent **40'** as the initial step followed by [3+2]-cycloaddition with alkene **64** to form cyclic adduct **66**. The latter undergoes oxidation giving pyrazole **67**. The subsequent formation of hydrazone **68**, its intramolecular cyclization, and the methyl group transfer from the phosphonate moiety to the hydrazine afford the final bispyrazole product **65**.



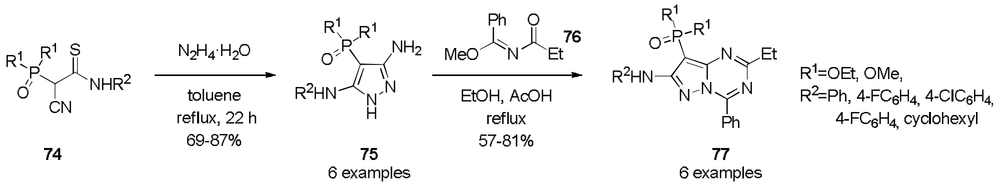
Scheme 15. Synthesis of 3-(pyrazol-3-yl)pyrazolyl-5-phosphonates.

Beletskaya and co-workers[75] reported the base-catalyzed synthesis of phosphorylated pyrazoles **73** from vinylphosphonates **69** and aryldiazomethanes **70** (Scheme 16). This reaction is applicable to diversely functionalized aryldiazomethanes; however, the reaction using ethyl diazoacetate for the aromatization leads to the loss of the phosphonate moiety. In the first step of the reaction, the [3+2]-cycloaddition of aryldiazomethane **70** to vinylphosphonate **69** gives dihydropyrazole **71**, which undergoes isomerization to intermediate **72** followed by the elimination of formamide to get final product **73**.



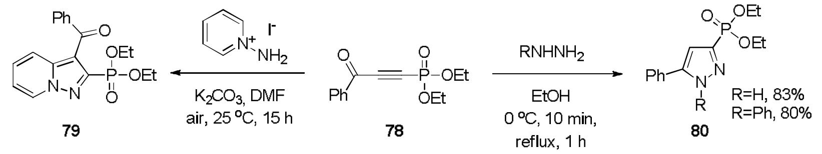
Scheme 16. Cycloaddition of aryldiazomethanes to vinylphosphonate.

Jalloul *et al.*[76] synthesized 3,5-diaminopyrazolyl-4-phosphonates **75** by the formal [3+2]-cycloaddition of 2-cyano-2-phosphoryl thioamides **74** with hydrazine hydrate (Scheme 17). The synthesized pyrazoles can be used in the design of more complex heterocyclic systems. Thus, the treatment of compounds **75** with methyl (*E*)-*N*-propionylbenzamidate **76** affords (7-amino-2-ethyl-4-phenylpyrazolo[1,5-*a*][1,3,5]triazin-8-yl)phosphonates **77**.



Scheme 17. Synthesis of 3,5-diaminopyrazolyl-4-phosphonates.

Zhao and co-workers[77] developed an approach to the synthesis of pyrazolyl-3-phosphonates **80** based on the [3+2]-cycloaddition of (3-phenyl-3-oxoprop-1-yn-1-yl)phosphonate **78** to organic hydrazines (Scheme 18). It is worth noting that the reaction of alkyne **78** with *N*-aminopyridinium iodide gives diethyl (3-benzoylpyrazolo[1,5-a]pyridin-2-yl)phosphonate **79**.

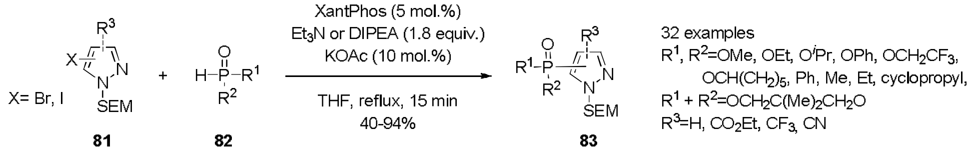


Scheme 18. Synthesis of pyrazol-3-ylphosphonates.

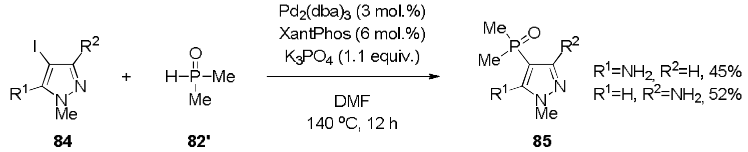
3.2. Phosphorylation of pyrazoles

Currently, the phosphorylation of pyrazoles is poorly described. Thus, Cossy and co-workers[78-79] performed the palladium-catalyzed reaction of iodo- and bromopyrazoles **81** with different dialkyl and aryl phosphites **82** (Scheme 19). The combination of a palladium(II) catalyst, Pd(OAc)$_2$, the commonly used ligand, XantPhos, and a base additive, Et$_3$N/DIPEA with KOAc, provided an efficient phosphorylation furnishing 3,4,5-substituted pyrazoles **83** in yields of up to quantitative. This reaction can be accomplished using both halopyrazoles without additional substituents and those containing electron-withdrawing groups, such as ester, cyano, or trifluoromethyl moieties.

Expanding this reaction, Mykhailiuk *et al.*[80] recently reported a palladium(II)-catalyzed synthesis of amino-substituted pyrazoles **85** *via* the phosphorylation of amino-substituted 4-iodopyrazoles **84** with dimethylphosphine oxide **82'** (Scheme 20). The reaction proved to be most effective when conducted in DMF with $Pd_2(dba)_3$ as the catalyst of choice.

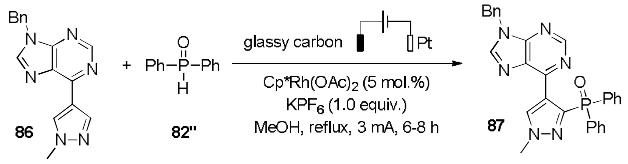


Scheme 19. Phosphorylation of iodo- and bromopyrazoles.



Scheme 20. Phosphorylation of amino-substituted 4-iodopyrazoles.

Wu *et al.*[81] described the only example of the rhodium-catalyzed phosphorylation of substituted pyrazole **86** enabled by anodic oxidation (Scheme 21). It was shown that the treatment of pyrazole-containing polyheterocyclic system **86** with diphenylphosphine oxide **82''** in the presence of $Cp^*Rh(OAc)_2$ and KPF_6 using a glassy carbon electrode affords product **87**.



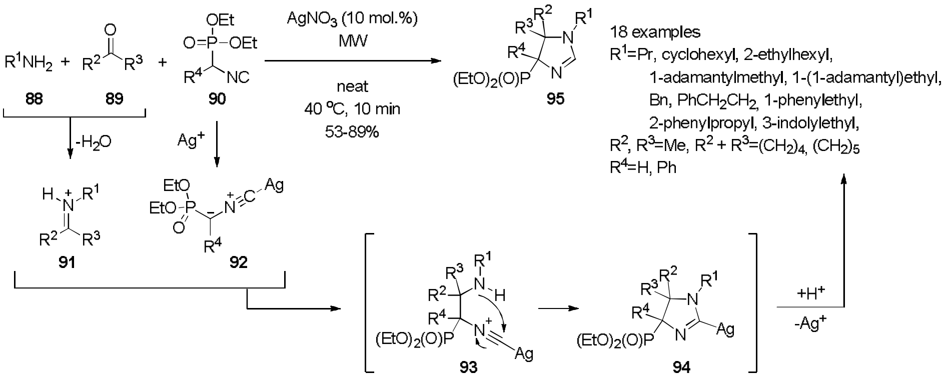
Scheme 21. Phosphorylation of pyrazole enabled by anodic oxidation.

4. Imidazoles

Phosphoryl-substituted imidazoles were first mentioned in 1973 by Ratcliffe *et al.*[82] and in 1978 by Bartlett *et al.*[83] However, currently there are a few available synthetic approaches to these compounds. In particular, noteworthy are the palladium-catalyzed phosphorylation of bromoimidazoles,[84] the reaction of imidazoles with pentavalent phosphorus compounds,[85-86] the intramolecular cyclization of [(1-isocyano-2,2-dichloro)-ethenyl]phosphonate derivatives,[87] and the formal [3+2]-cycloaddition of isocyanomethylphosphonates to imines.[88] The recently developed methods for the synthesis of phosphoryl-substituted pyrazoles, based on the cyclization of new linear phosphorus-containing precursors and the phosphorylation, are discussed below.

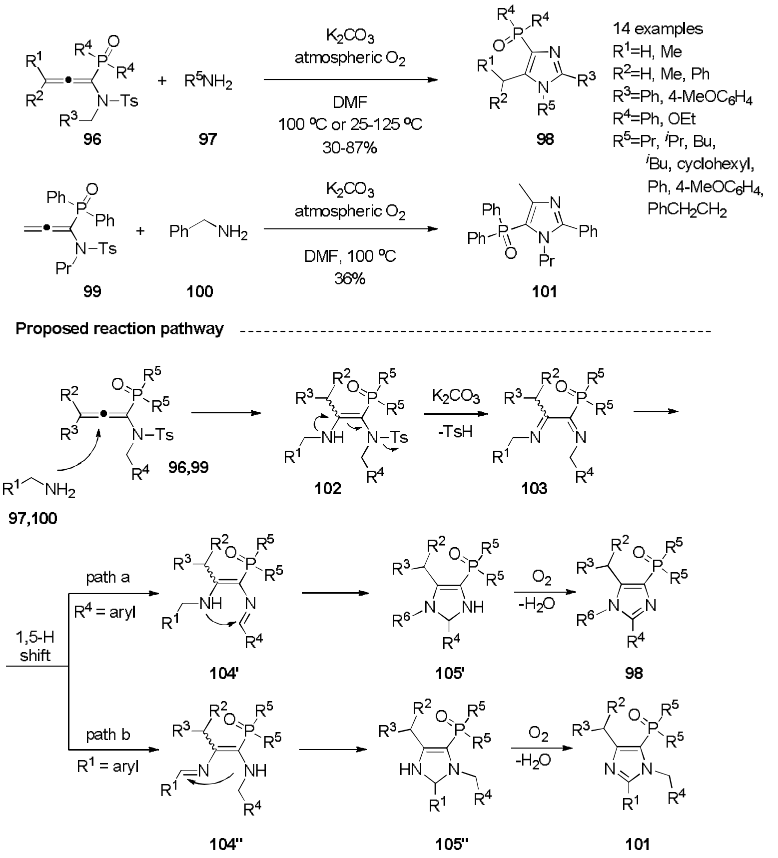
4.1. Synthesis of imidazoles from imines

Escolano and co-workers[89] reported the synthesis of 4-phosphoryl-substituted imidazolines **95** by the $AgNO_3$-catalyzed microwave-assisted three-component coupling of amines **88**, ketones **89**, and α-isocyanophosphonates **90** (Scheme 22). A variety of groups at amines, including aryl, cycloalkyl, and alkyl, were tolerated. The proposed reaction mechanism involves the complexation and deprotonation of the phosphonate reagent to form intermediate **92**. The nucleophilic addition of the resulting ylide to the iminium cation **91**, generated from ketone **89** and amine **88**, is followed by the intramolecular ring closure of intermediate **93** through the attack of the amine nitrogen atom on the isonitrile carbon atom to give imidazole **94**. The final step is the protodemetalation of the latter giving product **95**.



Scheme 22. Reaction between amines, ketones, and α-isocyanophosphonates.

Cao and co-workers[90] synthesized phosphorylimidazoles **98** and **101** by the reaction of allenyl sulfonamides with amines (Scheme 23). It was found that the reaction of allenyl sulfonamides **96** containing the benzyl substituent at the nitrogen atom with alkyl- and arylamines **97** affords 4-phosphorylimidazoles **98**.

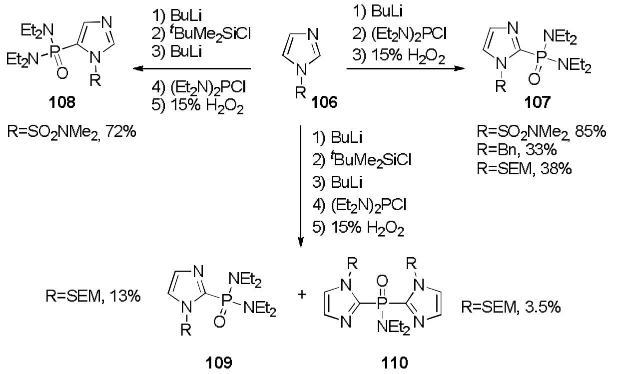


Scheme 23. Reaction of allenyl sulfonamides with amines.

Meanwhile, the reaction of *N*-propyl sulfonamide **99** with benzylamine **100** gives 5-phosphorylimidazole **101** as the major product. The proposed reaction mechanism involves the attack of the amine on the allenyl moiety to form diamine **102** followed by the base-mediated elimination of the tosyl group to give intermediate **103**. In the subsequent steps, the 1,5-proton shift leads to the formation of the C=N double bond conjugated with the aryl moiety (intermediates **104**) and the intramolecular attack of the amino group on this bond resulting in the five-membered ring closure to form **105**. The final step is the oxidative aromatization of the latter giving the reaction products (Scheme 23).

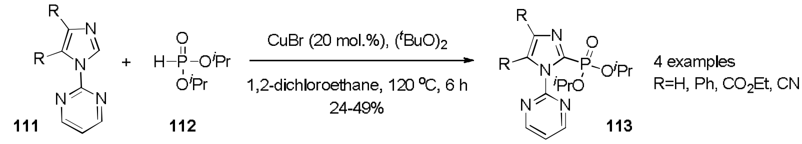
4.2. Phosphorylation of imidazoles

Pavlenko *et al.*[91] were the first to perform the phosphorylation of the imidazole ring through the formal replacement of the hydrogen atom by the phosphoryl moiety (Scheme 24). This approach involves the lithiation of imidazole **106** followed by the treatment with bis(diethylamino)chlorophosphine or 1-chloro-*N,N,N',N'*-tetramethylphosphadiamide. The final oxidation with hydrogen peroxide affords phosphorylated imidazolines **107** and **108-110**, respectively. Notably, bis-imidazole **110** was obtained as a side product in the reaction with *N*-SEM-substituted imidazole **106**.



Scheme 24. Phosphorylation of imidazoles.

Singh and co-workers[92] performed the CuBr-catalyzed oxidative phosphorylation of *N*-(2-pyrimidinyl)imidazoles **111** with diisopropyl phosphonate **112** in the presence of di-*tert*-butyl peroxide (Scheme 25). Product **113** modified with phenyl, CO_2Et, or CN substituents were obtained in reasonable yields. In this case, the 2-pyridinyl moiety serves as a directing group, which increases the selectivity of the replacement at the 2 position.



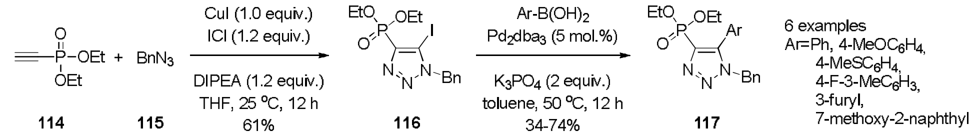
Scheme 25. Oxidative phosphorylation of *N*-(2- pyrimidinyl)imidazoles.

5. 1,2,3-Triazoles

Phosphorylated 1,2,3-triazoles were first described in 1973 by Tanaka *et al.*,[93] Heep *et al.*,[94] and Purovik *et al.*[95] The main routes to this class of compounds are based on the [3+2] azide-alkyne cycloaddition and the [3+2]-cycloaddition of diazomethylphosphonates to imines.[96-97] Besides, phosphoryl-substituted 1,2,3-triazoles can be synthesized *via* rearrangement of linear precursors.[98-100] Recent studies were focused on the development of the first two of the three methods aimed at increasing the regioselectivity and extending the range of possible substrates.

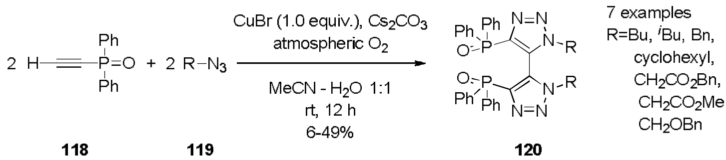
5.1. Azide-alkyne cycloaddition

Thiery *et al.*[101] synthesized 5-iodo-1,2,3-triazolyl-4-phosphonate **116** by the CuCl-catalyzed [3+2]-cycloaddition of alkynylphosphonate **114** to benzyl azide **115** *via in situ* iodination with ICl (Scheme 26). The reaction was further extended to the synthesis of substituted 5-aryl-substituted 4-pyrazolylphosphonates **117** by the Suzuki reaction of compound **116** with arylboronic acids.



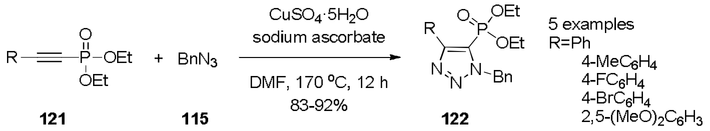
Scheme 26. Synthesis of 5-substituted 4-pyrazolylphosphonates.

Virieux and co-workers[102] reported the synthesis of bis-1,2,3-triazoles **120** through cascade [3+2]-cycloaddition reactions of diphenylphosphorylacetylene **118** with organic azides **119** accompanied by the oxidative dimerization in the presence of CuBr as the catalyst and atmospheric oxygen as the oxidant (Scheme 27). The reaction is general with respect to functionalized, linear, and cyclic aliphatic organic azides. To note, these compounds were used for the preparation of catalytically active Ir(I) complexes.



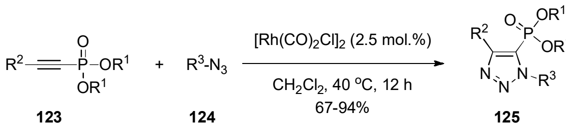
Scheme 27. Synthesis of bis(4-diphenylphosphoryl-1,2,3-triazoles).

Huang *et al.*[103] demonstrated that the [3+2]-cycloaddition of internal alkynylphosphonates **121** to benzyl azide **115** can be catalyzed by copper(II) in the presence of a reducing agent (Scheme 28). In the presence of $CuSO_4 \cdot 5H_2O$ and sodium ascorbate, diethyl phosphonates and benzyl azides underwent the "click reaction". This approach allows the selective synthesis of 4-aryl-substituted triazolyl-5-phosphonates **122** in high yields, but it requires a high temperature for the reaction to occur.



Scheme 28. $CuSO_4$-catalyzed [3+2]-cycloaddition of phosphorylalkynes to azides.

Song *et al.*[104] demonstrated that the same reaction can be catalyzed by rhodium complexes (Scheme 29).

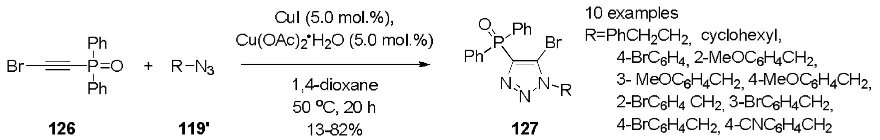


18 examples
R^1=Me, Et, ^{i}Pr, Bu
R^2=Ph, 4-$O_2NC_6H_4$, 4-ClC_6H_4, 4-BrC_6H_4, 4-MeC_6H_4, 4-$MeOC_6H_4$, 2-(6-methoxynaphthyl), 2-furyl
R^3=Bn, 4-$MeC_6H_4CH_2$, 4-$ClC_6H_4CH_2$, Ph, 4-$MeOC_6H_4$, 4-$MeO_2CC_6H_4$, $PhCH_2CH_2$, 4-(*N*-phthalimidyl)butyl

Scheme 29. Rhodium-catalyzed azide-alkyne cycloaddition in the synthesis of triazoles.

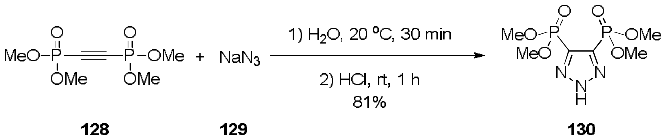
The highest yields were obtained using benzyl azide derivatives **124** and the rhodium(II) catalyst, $[Rh(CO)_2Cl_2]$. This approach allows the selective synthesis of substituted triazolyl-5-phosphonates **125** and does not require high temperatures. However, the reaction is sensitive to the bulkiness of the substituent in acetylene **123** and does not occur with the *tert*-butyl-containing derivative (Scheme 29).

Recently, Orita and co-workers[105] showed that 5-bromo-4-phosphoryltriazoles **127** can be synthesized in a regioselective manner by the copper-catalyzed azide-alkyne cycloaddition of bromo(phosphoryl)ethyne **126** with organic azides **119'** using the dual copper system $CuI/Cu(OAc)_2$ (Scheme 30). Remarkably, the reaction of alkyne **126** in the presence of only one catalyst [CuI or $Cu(OAc)_2$] furnished products **127** in poor yields. Aryl- and alkyl-substituted organic azides successfully participated in the reaction.



Scheme 30. $CuI/Cu(OAc)_2$-catayzed synthesis of 5-bromo-4-phosphorytriazoles.

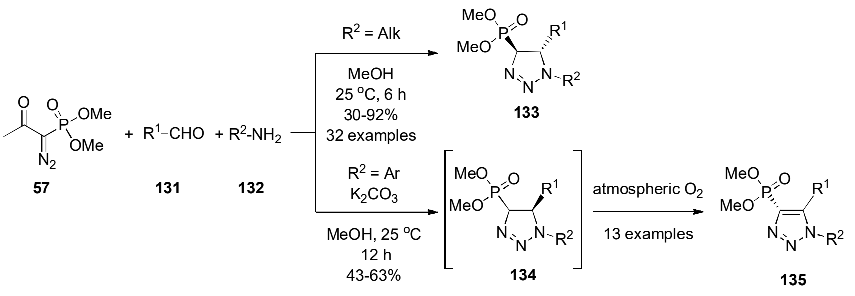
Artyushin *et al.*[106] used the metal-free [3+2]-cycloaddition of sodium azide **129** with bis(phosphoryl)acetylene **128** to prepare 2*H*-triazolediyl-4,5-bisphosphonate **130** (Scheme 31). Water was used as a promoting medium for the 1,3-dipolar cycloaddition providing the formation of the product in high yield.



Scheme 31. Synthesis of bis(phosphoryl)triazole.

5.2. Synthesis of 1,2,3-triazoles using the Ohira-Bestmann reagent

Mohanan and co-workers[107] studied the three-component reaction between aldehydes **131**, amines **132**, and Ohira-Bestmann reagent **57** (Scheme 32).



R^1=Ph, 3-$MeOC_6H_4$, 4-$MeOC_6H_4$, 3,4,5-$(MeO)_3C_6H_2$, 2-HOC_6H_4, 3,4-$(HO)_2C_6H_3$, 4-HO-3-$EtOC_6H_3$, 4-$Me_2NC_6H_4$, 4-$(HO)_2BC_6H_4$, 4-BrC_6H_4, 3-$O_2NC_6H_4$, 4-$HC(O)C_6H_4$, ferrocenyl, 2-thiophenyl, 3-thiophenyl, 2-furyl, phenylvinyl, penten-1-yl

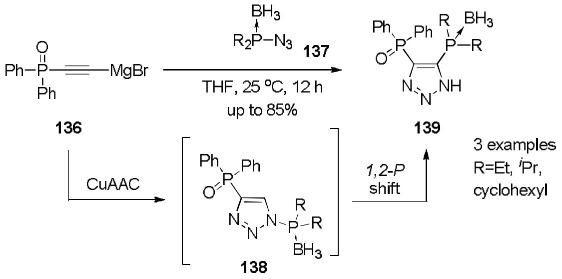
R^2=Pr, ^{i}Pr, Bu, ^{t}Bu, 1-octyl, 1-nonyl, Bn, $PhCH_2CH_2$, cyclohexyl, 1-adamantyl, 3-hydroxy-1-propyl, $CH_2CH_2NEt_2$, 3-morpholino-1-propyl, Ph, 3-$MeOC_6H_4$, 4-$MeOC_6H_4$, 3,4-$(MeO)_2C_6H_3$, 3,4-$Me_2C_6H_3$, 4-FC_6H_4, 4-ClC_6H_4, 4-BrC_6H_4, 4-IC_6H_4

Scheme 32. Three-component reaction between aldehydes, amines, and the Ohira-Bestmann reagent.

The reaction proceeds through a domino condensation/1,3-dipolar cycloaddition sequence and affords triazoline derivatives with excellent diastereoselectivity. The authors showed that if at least one substituent in the aldehyde and amine is not aromatic, the reaction affords triazoline **133** as the final product. If both reagents contain aromatic substituents, the reaction is accompanied by the further oxidation of intermediate **134** with atmospheric oxygen to give triazole **135**.This insertion reaction tolerated both electron-withdrawing and electron-donating aryl groups in the aldehyde and amine, as well as different alkenyl substrates (Scheme 32).

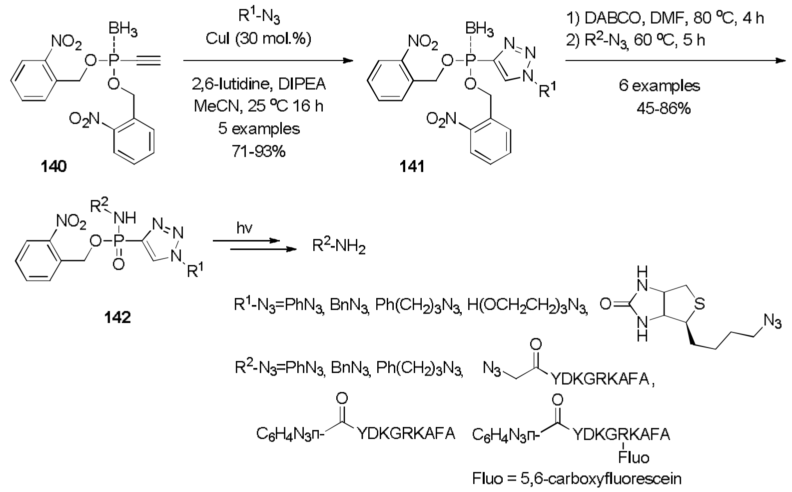
5.3. Synthesis of polydentate borophosphonate complexes

Gandelman and co-workers[108] synthesized BH_3-substituted phosphinophosphoryltriazoles **139** from magnesium phosphorylacetylides **136** and appropriate phosphine azides **137** (Scheme 33). The *N-C* migration of the $R_2P(BH_3)$ group in intermediate **138** is the key step of the transformation. The reaction is general with respect to azides bearing the BH_3-coordinated phosphine group with linear and cyclic alkyl substituents.



Scheme 33. Synthesis of phosphinophosphoryltriazoles.

Siebertz and Hackenberger[109] developed photocleavable conjugates based on phosphoryltriazoles **142** (Scheme 34). The synthesis of these conjugates involves the [3+2]-cycloaddition of BH_3-functionalized acetylenyl phosphite **140** with organic azides, resulting in the formation of triazole scaffold **141**, and the subsequent Staudinger reaction giving target conjugates **142**.

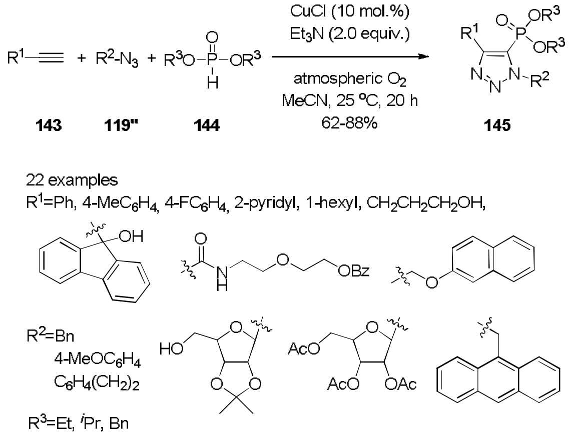


Scheme 34. Photocleavable conjugates based on phosphoryltriazoles.

The proposed approach allows the selective binding of azide-containing molecules in the form of triazole-containing borophosphonate complexes and the subsequent UV-assisted release of the corresponding amine. The authors examined the applicability of this concept using small molecules such as benzyl azide, 3-phenylpropyl azide, *etc*. Moreover, it was demonstrated that this reaction can be performed with small peptides bearing the terminal azido glycine or azidobenzoic acid moiety. Besides, the authors showed that the synthesized conjugates can be immobilized (Scheme 34).

5.4. Phosphorylation of 1,2,3-triazoles

Li *et al.*[110] employed the CuCl-catalyzed three-component reaction between organic azides **119''**, alkynes **143**, and phosphites **144** to prepare triazolyl-5-phosphonates **145** (Scheme 35). The proposed reaction mechanism involves the [3+2] azide-alkyne cycloaddition and the phosphorylation of the copper(I)-containing intermediate. Aryl- and alkyl-substituted alkynes, as well organic azides, were found to be efficient in the reaction providing products in good to high yields.



Scheme 35. Reaction between alkynes, azides, and phosphites.

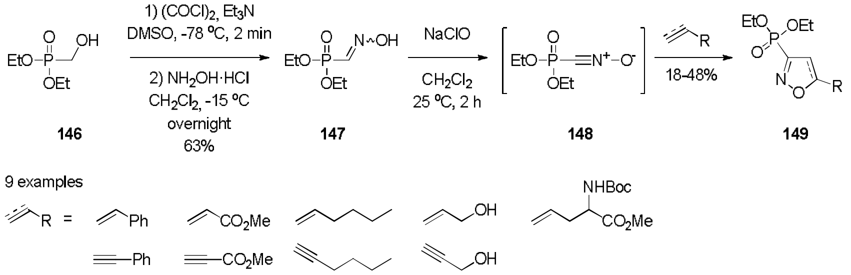
6. Isoxazoles

Phosphoryl-substituted isoxazoles were first described in 1973 by Heep.[94] Noteworthy are such synthetic approaches to phosphorylated isoxazoles based on the reaction of nitrile oxides with C-nucleophiles,[94] [3+2]-cycloaddition reactions involving nitrile oxides[111-114] and silyl nitronates,[115] and the cyclization of linear bifunctional phosphorus-containing precursors with hydroxylamine.[116-117] In recent years, a method was developed based on the formal [3+2]-cycloaddition of nitrile oxides to dipolarophiles and methods were proposed for the synthesis of phosphoryl-substituted isoxazoles based on the cyclization of new linear phosphorus-containing precursors. These methods are discussed below.

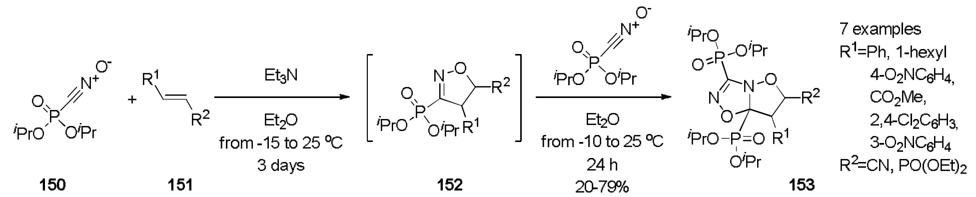
6.1. Formal [3+2]-cycloaddition

Conti *et al.*[118] prepared isoxazolyl-3-phosphonates and isoxazolinyl-3-phosphonates **149** by the formal [3+2]-cycloaddition of (diethoxyphosphoryl)formonitrile oxide **148** to terminal alkenes and alkynes (Scheme 36). This process is characterized by high regioselectivity (on the average, 99% of the 5-substituted product), but the yields are not higher than 48%. The reaction scope appears to be quite broad since various alkyl and aryl groups in the ene component were tolerated. Notably, nitrile oxide **148** was synthesized in two steps from diethyl (hydroxymethyl)phosphonate **146** through diethyl (hydroxyimino)methylphosphonate intermediate **147**.

Using this approach, Shen *et al.*[119] studied the formal [3+2]-cycloaddition of phosphorylnitrile oxide **150** to internal alkenes **151** (Scheme 37). The authors isolated annulated 7,7a-dihydro-6*H*-isoxazolo[2,3-*d*][1,2,4]oxadiazoles **153** as the major products, which were generated through the stepwise cycloaddition of nitrile oxide **150** to double bonds of alkene **151** and isoxazolyl **152**. Electron-deficient alkenes bearing aryl and alkyl substituents were efficient in reactions giving the target products in yields of up to high.

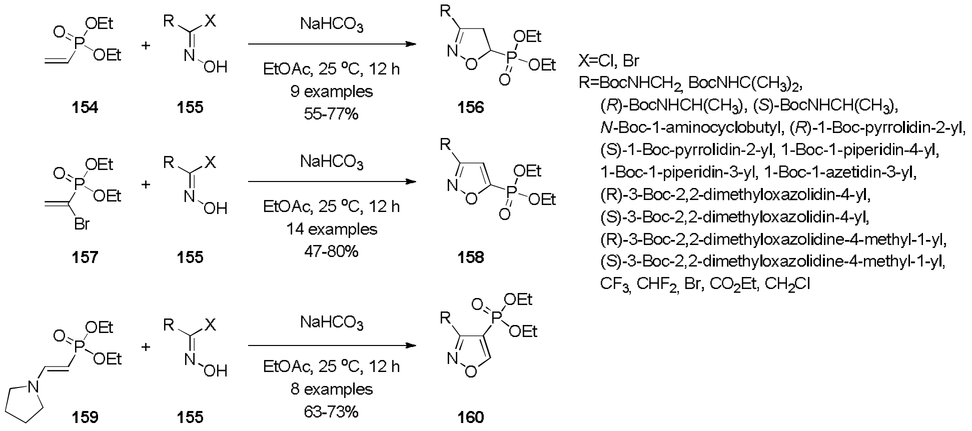


Scheme 36. Formal [3+2]-cycloaddition of (diethoxyphosphoryl)formonitrile oxide to alkenes and alkynes.



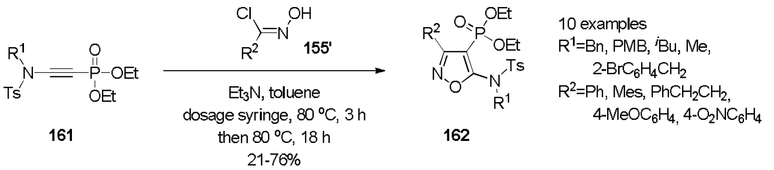
Scheme 37. Double [3+2]-cycloaddition of phosphorylformonitrile oxide to alkenes.

Grygorenko and co-workers[120] performed the reaction of vinylphosphonates **154**, **157**, and **159** with substituted α-halonitrile oxides **155** (Scheme 38). A series of isoxazolyl-4-phosphonates **156**, isoxazolyl-5-phosphonates **158**, and isoxazolinyl-5-phosphonates **160** was synthesized. This reaction is quite general in scope and allows for a very facile preparation of a variety of substituted isoxazoles in good yields. The substituent pattern of the reaction product is determined by the vinylphosphonate used. Thus, the reactions employing reagents without additional substituents or those containing a substituent at the 1 position (**154** and **157**) afford products bearing a phosphonate group at the 5 position; the reaction of vinylphosphonate **159** containing a substituent at the 2 position gives 4-phosphorylated isoxazoles. Vereshchagina *et al.*[121] used this approach to prepare tetramethyl 3-(4-fluorophenyl)-4,5-bis(isoxazolyl)phosphonate from tetramethyl acetylenediphosphonate.



Scheme 38. Formal [3+2]-cycloaddition of nitrile oxides to vinylphosphonates.

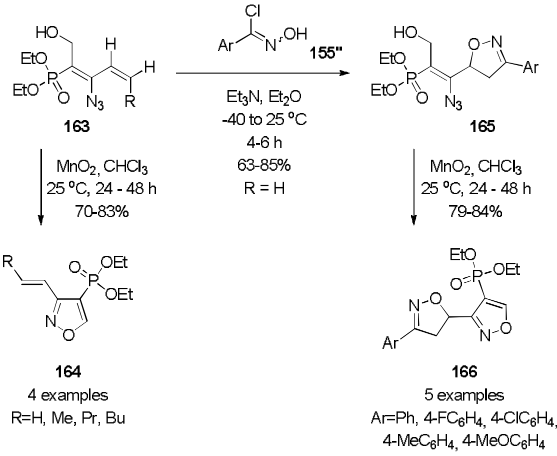
Perez *et al.*[122] used the formal [3+2]-cycloaddition of nitrile oxides **155'** to [(*N*-tosylalkyl)ethynyl]phosphonates **161** as an approach to synthesize 3-substituted 5-aminoisoxazolyl-4-phosphonates **162** (Scheme 39). This reaction can be performed with reagents containing different substituents, in particular benzyl, alkyl, and aryl groups, except those with additional double bonds.



Scheme 39. Formal [3+2]-cycloaddition of nitrile oxides to [(*N*-tosylalkyl)ethynyl]phosphonates.

6.2. Cyclization of γ-keto and hydroxy vinyl azides

Brel[123] proposed an interesting approach to the synthesis of isoxazolyl-4-phosphonates **164** *via* the intramolecular cyclization of γ-hydroxy vinyl azides **163** (Scheme 40). The manganese dioxide-mediated oxidative cyclization of 3-hydroxy vinyl azides was found to be efficient in the synthesis of alkene-bearing phosphoryl-substituted isoxazoles in good yields. Due to the presence of an additional double bond in 3-hydroxy vinyl azide intermediate **163**, one more isoxazole moiety can be introduced *via* [3+2]-cycloaddition involving nitrile oxides **155''** to prepare polyheterocyclic system **166** from intermediate **165**. It is worth noting that only one double bond (which is not substituted by the phosphonate moiety) of the diene is involved in this reaction.

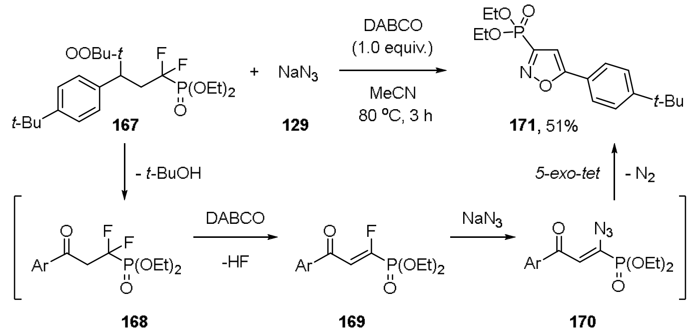


Scheme 40. Oxidative cyclization of 3-hydroxy vinyl azides.

Recently, Li *et al.*[124] showed that the DABCO-promoted heterocyclization of β,β-dihalo peroxides **167** with sodium azide **129** is an efficient route to the synthesis of 3-phosphoryl isoxazoles (Scheme 41). Compound **171** was obtained from peroxide **167** in moderate yield under mild conditions. The reaction is believed to occur as the stepwise Kornblum-DeLaMare rearrangement and the elimination of fluorine from carbonyl intermediate **168** to form alkene **169**. Finally, the fluorine replacement with sodium azide followed by the *5-exo-tet* cyclization of intermediate **170** affords product **171**.

7. Oxazoles

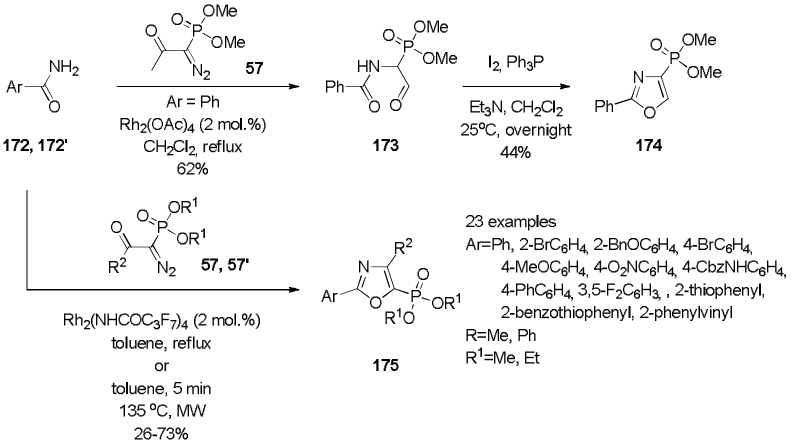
Phosphorylated oxazoles were first described in 1973-1975 by Drach *et al.*[125-128] The following synthetic approaches to phosphoryl-substituted oxazoles are worth mentioning: the metal-catalyzed reaction of α-diazo-β-ketophosphonates with nitriles,[129] reactions between phosphorylazirines and carboxylic acids derivatives,[130,135,136] the reaction of isocyanomethylphosphonates with carboxylic acid chlorides,[131-134] reaction of 1-dichloroacetylamino-2,2,2-trichloroethylphosphonates with secondary amines,[137,139] and thionyl chloride mediated intermolecular cyclization of (1-acylamino-2-oxo-2-phenylethyl)-phosphonates.[138] In recent years, these methods were further improved and new methods were developed based on the direct phosphorylation of the oxazole ring. These approaches are discussed below.



Scheme 41. DABCO-promoted cyclization of β,β-difluoro peroxides with sodium azide.

7.1. Formal [3+2]-cycloaddition

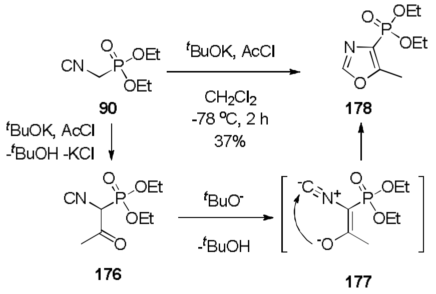
Moody and co-workers[140-141] studied the rhodium-catalyzed synthesis of phosphorylated oxazoles from arylamides and the Ohira-Bestmann reagent (or its analogues) (Scheme 42). The reaction pathway depends on the nature of the catalyst used. Thus, the $Rh_2(OAc)_4$-catalyzed reaction of benzamide **172'** with keto azide **57** under mild conditions affords phosphorylated ketoamide **173**, which can be transformed into oxazolyl-4-phosphonate **174** in reasonable yields by the treatment with iodine in the presence of triphenylphosphine. Meahwhile, the $Rh_2(NHCOC_3F_7)_4$-catalyzed reaction of aryl amides **172** with azides **57** and **57'** was shown to be efficient for the regioselective synthesis of oxazolyl-5-phosphonates **175**. Under microwave-assisted conditions, these compounds were obtained in low to good yields.



Scheme 42. Rhodium-catalyzed reaction of arylamides with the Ohira-Bestmann reagent.

7.2. Cyclization of α-isonitrile-β-ketophosphonates

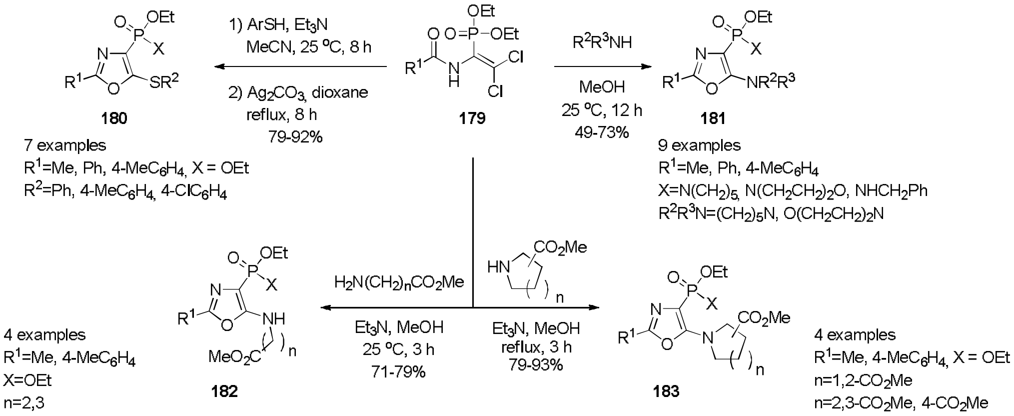
Dang *et al.*[19] described the assembly of 5-methyloxazolyl-4-phosphonate **178** from isonitrile **90** and acetyl chloride (Scheme 43). This reaction involves the acylation of isocyanophosphonate **90** at the methylene group, the enolization of the resulting ketone **176**, and the intramolecular attack of the oxygen atom on the isocyano group of intermediate **177** to give the final product **178** in reasonable yield.



Scheme 43. Assembly of 5-methyloxazolyl-4-phosphonate from isonitrile and acetyl chloride.

7.3. Cyclization of *N*-acyl-1-phosphoryl-2,2-dichloroethyleneamines

A series of studies by Brovarets and co-workers[142-149] were devoted to the synthesis of phosphorylated oxazoles based on the reaction of *N*-acyl-1-phosphoryl-2,2-dichloroethyleneamines **179** with *N*- and *S*-nucleophiles (Scheme 44). Amines, amino acid esters (both linear and cyclic), and thiols (in this case, additional activation by silver carbonate is required for heterocyclization) can be employed as nucleophiles in the cyclization reaction providing 3-substituted 4-phosphorylated oxazoles **180-183**. A variety of alkyl- and aryl-substituted enamines were tolerated under the reaction conditions giving oxazoles in moderate to excellent yields.

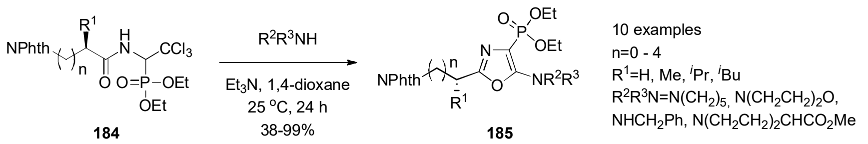


Scheme 44. Reaction of *N*-acyl-1-phosphoryl-2,2-dichloroethyleneamines with nucleophiles.

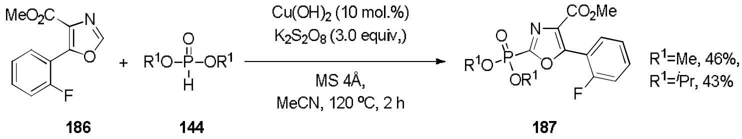
The same group described the reaction of *N*-acyl-1-phosphoryl-2,2,2-trichloroethylamines (as an example compound **184**) with *N*-nucleophiles in the presence of triethylamine as an efficient approach to 4-phosphorylated oxazoles.[142-143, 148-150] The intramolecular cyclization believed to occur through the *in situ* generation of phosphoryl-2,2-dichloroethyleneamine intermediated mentioned above (Scheme 45). The approach potential is quite good with respect to scope of amines which can be involved in the reaction. As an example, the authors applied this approach to prepare 4-phosphoryloxazoles **185** containing an alkylamino group at the 2 position, including those bearing a chiral center.

7.4. Phosphorylation of oxazoles

The phosphorylation of oxazoles are relatively poorly studied. Recently, Singh and co-workers[92] described two examples of the Cu(II)-catalyzed direct radical phosphorylation of 4,5-substituted oxazoles at the 2 position (Scheme 46). In the presence of $K_2S_2O_8$ and $Cu(OH)_2$, electron-deficient oxazole **186** reacted with dialkyl phosphonates **144** to form products **187**. However, it should be noted that this transformation occurs in low yields at high temperature. Moreover, the phosphorylation of the oxazole ring at the 4 and 5 positions is still unknown.



Scheme 45. Reaction of *N*-acyl-1-phosphoryl-2,2,2-trichloroethylamines with nucleophiles.



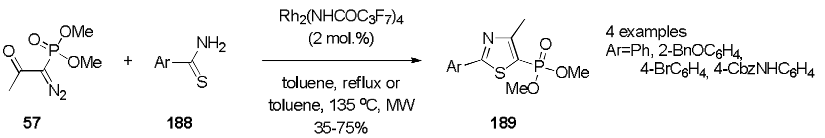
Scheme 46. Direct phosphorylation of the oxazole derivative.

8. Thiazoles

The synthesis of phosphoryl-substituted thiazoles was first described by Dawson and Burger.[151] The following synthetic approaches are worth mentioning: the reaction of α-bromo-β-ketophosphonates with thioamides,[151-152] the cyclization of linear phosphorylated precursors with different reagents,[153-154] the reaction between isocyanomethylphosphonates and carbon disulfide,[132] the reaction between allenyl isothiocyanate and phosphites,[155] the reaction of metalated thiazoles with P–Cl-containing compounds,[156-157] and the electrophilic phosphorylation of thiazoles.[158-159] In the past decade, the studies were focused on the development of methods for the direct phosphorylation of the thiazole core, while the synthesis of thiazolylphosphonates using the Ohira-Bestmann reagent was described in a single publication.

8.1. Formal [3+2]-cycloaddition

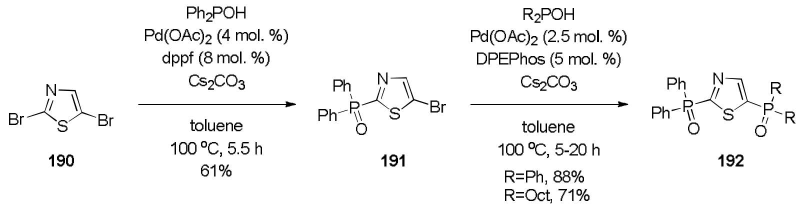
Moody and co-workers[141] described the reaction between Ohira-Bestmann reagent **57** and thioamides **188** in the presence of $Rh_2(NHCOC_3F_7)_4$ giving thiazolyl-5-phosphonates **189** (Scheme 47). Electron-withdrawing and electron-donating aryl-substituted thioamides participated in the heterocyclization. However, this reaction affords the target products in low yields.



Scheme 47. Reaction between the Ohira-Bestmann reagent and thioamides in the presence of $Rh_2(NHCOC_3F_7)_4$.

8.2. Phosphorylation of thiazoles

The phosphorylation of thiazoles was performed only at the 2 position. The works in this field up to 2019 are considered in a review by Chen.[32] Recently, Zakirova *et al.*[160] developed the Pd(II)-catalyzed synthesis of mono- and bis(phosphorylated) thiazoles (Scheme 48). The catalyst $Pd(OAc)_2$ proved to be particularly efficient in the stepwise phosphorylation of 2,5-dibromothiazole **190** with phosphine oxides. The yields of target nonsymmetric bis(tertiary phosphine oxides) **192** were higher when using DPEPhos instead of dppf, which was used for the synthesis of (5-bromothiazol-2-yl)diphenylphosphine oxide **191**. The reaction was general for diphenyl- and dioctylphosphine oxides. The products were obtained in good to high yields.



Scheme 48. Pd(II)-catalyzed synthesis of mono- and bis(phosphorylated) thiazoles.

9. Conclusions

In conclusion, the phosphorylation is definitely the most general route to the synthesis of phosphoryl-substituted five-membered aromatic *N,O,S*-heterocycles. Such methods as the Arbuzov reaction (despite more than a century history), the palladium-catalyzed cross-coupling of diethylphosphonate with halogen-substituted heterocycles, and the C(sp^2)H-phosphorylation of heterocycles with diethylphosphonates promoted by one-electron oxidants were extensively developed in the past decades. Due to a wide range of available phosphorylating agents and the possibility of performing the reaction in a nucleophilic, electrophilic, or radical manner, the phosphorylation has attracted great attention in fundamental organic chemistry. However, the necessity of finding conditions for each particular type of heterocycles, severe reaction conditions (high temperature, the presence of strong bases and acids), and high cost of the catalysts significantly limit the practical application of this approach.

The strategy of the synthesis of phosphoryl-substituted five-membered heterocycles, which is based on the construction of the heterocyclic core using phosphoryl-containing reagents, employs mainly the formal cycloaddition and intramolecular cyclizations. Most of the methods developed in this area in the past decade do not have a general character. However, the application of versatile reagents, such as the Wittig-Horner, Horner-Wadsworth-Emmons, Ohira-Bestmann, and Seyferth-Gilbert reagents, in the synthesis of phosphoryl-substituted heterocycles has attracted increasing interest. It was shown that these compounds are efficient precursors in the synthesis of different phosphorylated five-membered heterocycles, including such important pharmacophores as pyrazoles, 1,2,3-triazoles, oxazoles, thiazoles, *etc.* Their evident advantages are the simplicity of handling, commercial availability, and predictable reactivity.

Since there are a few examples of the synthesis of certain phosphorylated five-membered heterocycles, while synthetic approaches to phosphorylated derivatives of a wide range of basic *N,S,O*-heterocycles are absent, further research in this field is a long-term challenge. Clearly, the development of synthetic methods based on the phosphorylation of heterocycles and the heterocyclization involving phosphoryl-substituted reagents is of fundamental research interest. The design of new versatile multifunctional reagents for various chemical transformations has a great promise in the synthesis of phosphoryl-substituted heterocycles. The detailed investigation of such new reagents would provide the basis for a new area in the synthesis of nitrogen-, sulfur-, and oxygen-containing heterocyclic compounds with different ring sizes and different combinations of heteroatoms and also for the preparation of new complex heterocyclic systems and assemblies.

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