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Research Article

Recent Developments in Pyranopyrazole Derivatives: Synthesis, Reactions, and Potential Pharmaceutical Applications

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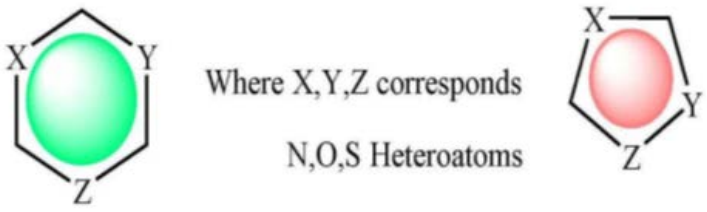
A B S T R A C T

In the recent years the science of assembling heterocyclic ring has made enormous strides. Heterocyclic compounds consist of very important key compounds in organic chemistry. Due to their various pharmaceutical properties resulting many researchers are syntheses of these compounds. A number of drugs containing simple heterocyclic or a combination of different heterocyclic moieties have been used these days as anticancer, antibacterial and antifungal agents. Because of the heteroatoms in the heterocycle compounds they are playing very role in the organic chemistry.



The presence of nitrogen atoms in the cyclic compounds increased their pharmaceutical efficiency which we can find in many natural compounds like vitamins, hormones and enzymes. The compounds which consist of five and six membered heterocyclic nitrogen containing systems such aspyrazole, imidazole, trizoles, thiozolidine, pyrazolidine etc. as far by the most important in the ongoing research for more efficacious drugs in the fields such as antibacterials, antifungal, anti-inflammatory, diuretics, antirheumatics and antihistaminic. Here we will shed light on Synthesis, Reactions and Biological Applications of Pyrazolopyrimidine Derivatives.

1. Introduction

The heterocyclic compounds are widely spread in nature and play an important role in life. Due to the characteristic properties, the heterocyclic compounds hold a large area in medicinal

chemistry. The chemistry of heterocyclic chemistry has been explored widely in the past two -three decades. The synthesis and the application of heterocyclic compounds of medium size rings became popular[1].

Heterocyclic compounds, bearing atoms of at least two different elements as a member of its ring have attracted considerable attention in the growth of pharmacologically active molecules and advanced organic materials Because of the heteroatoms in the heterocycle compounds they are playing very rule in the organic chemistry. The presence of nitrogen atoms in the cyclic compounds increased their pharmaceutical efficiency which we can find in many natural compounds like vitamins, hormones and enzymes[2].

2. Pyranopyrazole

Pyranopyrazoles are known since the 19th century and first synthesized in 1974 by Otto who synthesized it via cyclization of 4-aryliden-5-pyrazolone in the presence of base. Pyranopyrazole compounds, oxygen- and nitrogen-ring fused heterocycles, are important group of heterocyclic compounds with natural and synthetic molecules[3]. The synthesis of the heterocyclic compounds containing the pyranopyrazole moiety, is of great importance, besides its biological and medicinal properties. pyranopyrazoles have attracted the attentions of agrochemical research due to their fungicidal, bactericidal, and herbicidal properties.There are four isomeric structures for pyranopyrazole including: pyrano [2,3-c] pyrazole, pyran o[3,2-c] pyrazole , pyrano [3,4-c] pyrazole , and pyrano [4,3-c] pyrazole, but pyrano [2,3-c] pyrazole isomer is the most investigated one, on the other hand, reports on the preparation of other three pyranopyrazoles are rare[4].

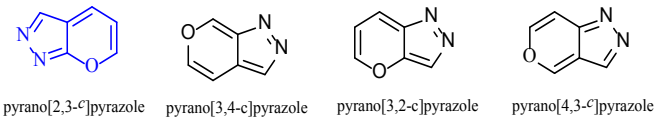


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Figure 1: Different chemical structures of pyrano pyrazoles.

3. Synthesis of Pyrano [2,3-c] pyrazoles

3.1. Two component synthesis of Pyrano [2,3-c] pyrazoles

Junek and Aigner treated tetracyanoethylene with pyrazol-5-one and 5-aminopyrazole to obtain pyrano[2,3- c]pyrazoles **(5)**, pyrazolo[3,4-b]pyridines **(6)** and dipyrazolylmalonodinitriles **(7)** respectively depending on reaction condition (**Figure 2**). 6-Amino-1,3-disubstituted-4,4-5-tricyanopyrano[2,3-c] pyrazole **(5)** was obtained by refluxing the appropriate pyrazolone and tetracyanoethylene in ethanol[5].

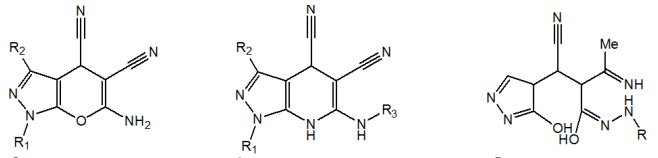
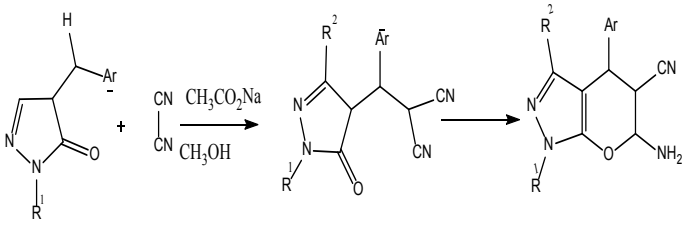


Figure 2: Two component synthesis of Pyrano [2,3-c] pyrazoles.

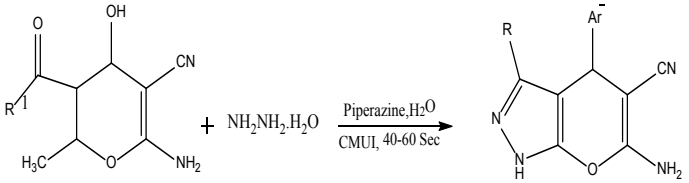
Otto refluxed 4-benzylide-pyrazol-5-one (**8**) with malononitrile (**9**) in methanol in the presence of sodium acetate catalyst to obtain pyrano [2,3-*c*] pyrazole (**11**)[6] (**Scheme 1**).



8 **9** **10** **11**

(**Scheme 1**)

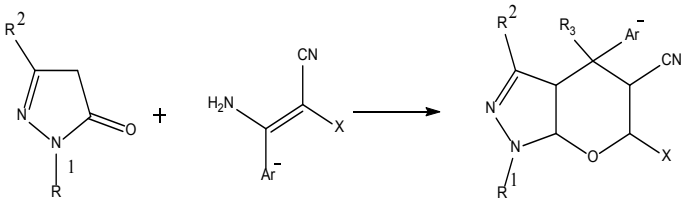
Water as a green solvent is the most environmentally friendly, safe and inexpensive choice to decrease pollution, toxicity and cost of a reaction. Peng and co-workers used pure aqueous media for reaction of 5- alkoxycarbonyl-2- amino-4-aryl-3-cyano-6-methyl-4*H*-pyrans (**12**) and hydrazine hydrate in the presence of a catalytic quantity of piperazine by three methods (i) heating (ii) exposing to microwave irradiation (iii) exposing to a combination of microwave and ultrasound irradiation where, the latter was found to be excellent in terms of yield within short time[7]. It was assumed that powerful ultrasound irradiation causes cavitations and high-velocity interparticle collisions, which cleaned the surface, thus mass transfer between two phases increased and the reaction completed fast without need of any organic co-solvent (**Scheme 28**).



12 13 14

Scheme 2

Abdou and co-workers, in a simple procedure, refluxed various alkene derivatives (16) and pyrazolones in piperidine containing ethanolic solution to produce a variety of pyranopyrazoles bearing carbonitrile, hydroxyl or a phenyl group at the 6-position (Scheme 3)[8].

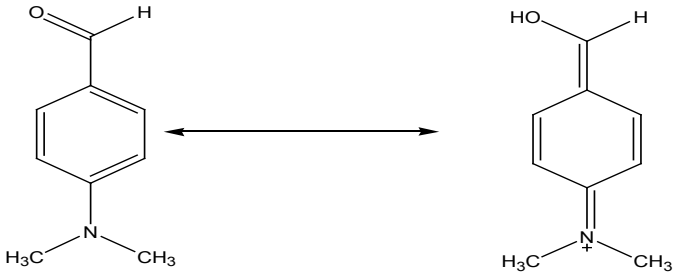


15 16 17

(**Scheme 3**)

3.2. Three components synthesis of Pyrano [2,3-c] pyrazoles

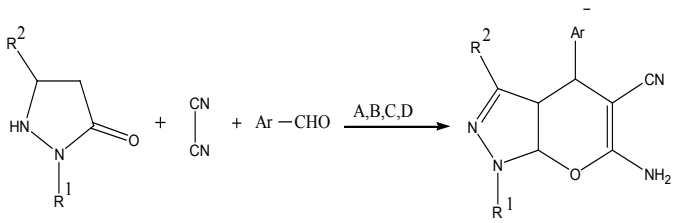
Most of these examples are used pyrazolone, aldehydes and malononitrile and allowed to react together under different reaction conditions to form a variety of pyranopyrazoles. Jin and co-workers added p-dodecylbenzenesulfonic acid (DBSA), as phase transfer catalyst, for uniform dispersion of reactants to get a better yield (84-94%). Initially, the reaction was tested in the absence of catalyst and yielded traces of product or no product as in case of 4-dimethylaminobenzaldehyde, which has strong electron donating dimethylamino group that has significant contributions of the quinoid resonance form, hence reactivity decreased **18-19** (**Figure 3**)[9].



18 19

Figure 3: Three components synthesis of Pyrano [2,3-c] pyrazoles.

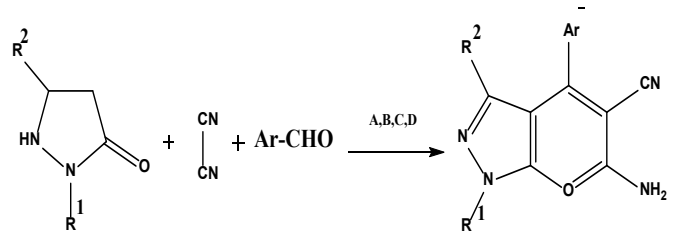
In another attempt, various PTC namely, TBAB, DBSA, sodium dodecyl sulphate (SDS) and HTMAB were tested for similar reactants where HTMAB was found best in term of yield[10]. The reaction conditions worked equally for aromatic aldehydes with electron-withdrawing and donating substituents, but did not proceed for aliphatic aldehydes probably, due to their low reactivity. Prajapati and co-workers refluxed substituted aldehydes, malononitrile and 1-(2,4-dinitrophenyl)-3-methylpyrazol-5-one in ethanol containing piperidine catalyst to give the respective pyranopyrazoles which were found to be good antibacterial agents[11].



20 21 22 23

(**Scheme 4**)

Pyranopyrazoles bearing a triflouromethyl group at the 3-position were obtained by reaction of aldehydes, malononitrile and trifluoromethylpyrazol-5-one, in water as solvent without catalyst at 90°c, in good yields in 3-5 h (Scheme 5)[12]. The yield of the product is not affected by the electronic nature of the aryl substituents.

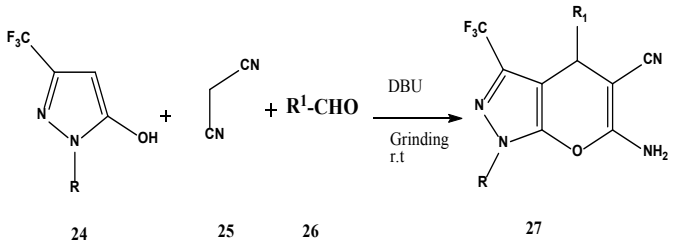


20 21 24 25

(**Scheme 5**)

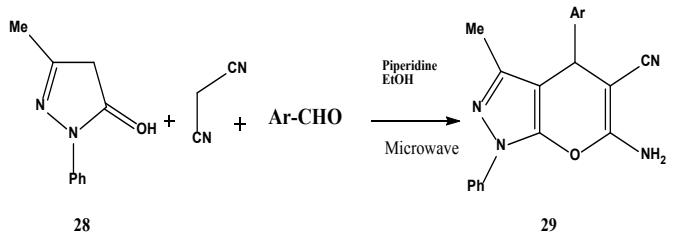
Bhavanarushiandco-workerspreparedflouropyranopyrazoles by grinding similar reactants in a pestle mortar using DBU as catalyst and established the molecular mechanism for DNA binding of resultant products (Scheme 6) [13].

Microwave irradiation to eliminate the need of heat, enhances the rate of reaction, is a widely applicable technique and has been used for the synthesis of pyranopyrazoles within 2-8 min in dry ethanol containing piperidine catalyst (**Scheme 7**)[14].



24 25 26 27

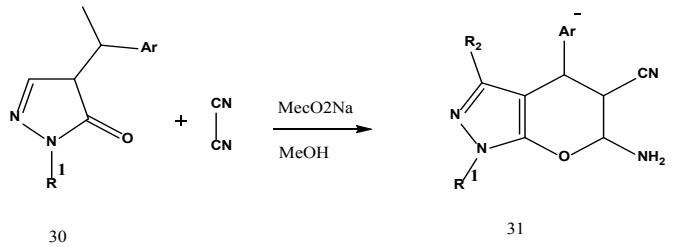
Scheme 6



28 29

Scheme 7

Diaminopyrano [2,3-c] pyrazoles were prepared at room temperature in ethanolic solvent containing secondary amine/ organic bases such as pyridine, piperidine and pyrrolidine[15]. The resultant compounds were found to be potential antibacterial agent while, some of them also exhibited antifungal activity (**Scheme 8**).



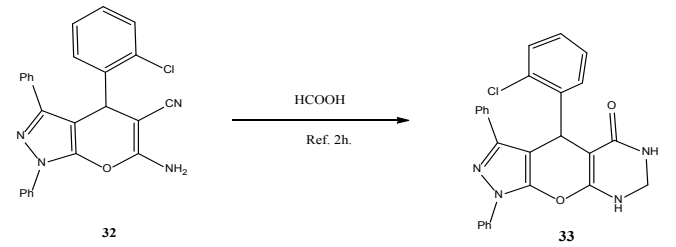
30 31

Scheme 8

4. Reactions of Pyranopyrazoles

4.1 Reaction with formic acid

Nassar etal[16] reported reaction of pyranopyazole derivative 32 with formic acid by reflux at high temperature to produce pyranopyrazolopyrimidinone derivative 33 [Scheme 9]

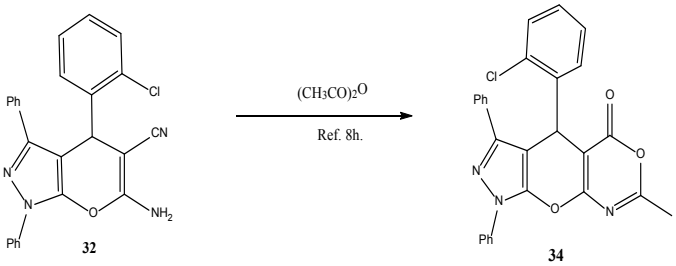


32 33

Scheme 9

4.2. Reaction with acetic anhydride

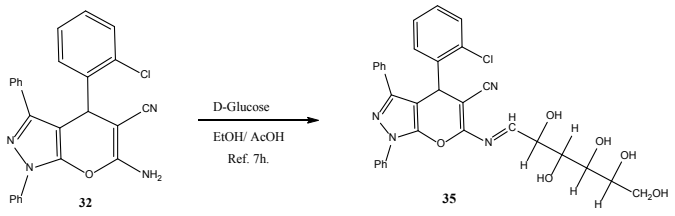
Nassar etal [16] reported reaction of pyranopyazole derivative 32 with acetic anhydride by reflux to produce pyranopyrazolooxazinone derivative 34 [Scheme 9].



32 34

5. Reaction with sugars

Nassar etal [16] reported reaction of pyranopyazole derivative 32 with d-glucose by reflux to produce sugar derivative 35 [Scheme 10]



5. Biological Activities

Pyranopyrazoles in general are biologically active and have remarkable antimicrobial, anticancer[17], anti-inflammatory, analgesic, anticonvulsant, anti-platelet[18], vasodilator, antifungal, potential Chk1 inhibitor[19] herbicidal16 and molluscicidal properties. Moreover, pyranopyrazoles were found to be effective inhibitors to steel corrosion 6 and as antioxidants for lubricant oil. Since these can lead to a variety of pyrano[2,3-c] pyrazoles by virtue of aryl and hetaryl aldehydes, hydrazines and malononitriles and other reactants, the researchers from time to time have subjected the novel synthesized compounds to diverse type of biological activities which may be summed up in the following: Tetrahydroquinolines derivatives being biological active anti-HIV, antibacterial, antifungal, antimalarial, antitrypanosomal, antitumor, psychotropic, anti-allergic, anti-inflammatory, and estrogenic agents, were incorporated with pyranopyrazoles to obtain potential biologically active compounds[20-63].

6. Conclusion

In conclusion, the research presented in this paper demonstrates the versatility and potential of pyranopyrazole derivatives in organic and pharmaceutical chemistry. The innovative synthesis methods, including environmentally friendly approaches and the use of phase transfer catalysts, have led to the efficient production of these compounds. The biological activities of pyranopyrazoles, especially their antibacterial properties, suggest their promise as candidates for drug development. Future research should focus on exploring the full therapeutic potential of these compounds, investigating their mechanisms of action, and developing more targeted applications in medicine. The continued exploration of pyranopyrazole derivatives is likely to yield significant contributions to the field of heterocyclic chemistry and pharmacology.

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