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Review Article

RECENT TREND ON KNOWN PYRAZOLONE ENDOWED DERIVATIVES: ANOVERVIEW

G. NEELIMA[1*], T. ASHA[1], S. RUKMINI[1], G. K. BRUNDHA[1], SREERAMA RAJASEKHAR[2]

[1]Department of Pharmaceutical Analysis, Sri Venkateswara College of Pharmacy (Autonomous), RVS Nagar, Chittoor 517127, Andhra Pradesh, India, [2]Department of Pharmaceutical Chemistry, Sri Venkateswara College of Pharmacy (Autonomous), RVS Nagar, Chittoor 517127, Andhra Pradesh, India

Email: grandhe.neelima37@gmail.com

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ABSTRACT

Pyrazolone is a lactam ring with five-membered and 2-Nitrogen and 1-ketonic groups in its structure. Antimalarial, antifungal, antimycobacterial, antibacterial, anti-inflammatory, anticancer, gastric secretion stimulatory, antidepressant and antifilarial properties are also found in pyrazolones. They are also used in the extraction and separation of various metal ions, as well as precursors for dyes, pigments, insecticides and chelating agents. The excellent therapeutic qualities of pyrazolone-related medicines have prompted medicinal chemistry to develop a slew of new chemotherapeutic treatments. The synthesis of pyrazolone and numerous structural reactions may be done in a variety of ways, which opens up a lot of possibilities in the field of medicinal chemistry. This article mainly reports the chemistry, novel synthesis and biological activity of Pyrazolones and their derivatives.

Keywords: Pyrazolone, Synthesis, Chemotherapy, Heterocyclic and Biological activity

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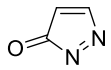
INTRODUCTION

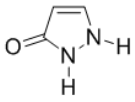
Pyrazolone is a 5-membered heterocyclic possessing two nitrogen atoms that are very close together. It is a carbonyl [C=O] pyrazole derivative with an extended carbonyl group. Compounds containing this functional group are utilized in analgesics and dyes in the new economy.



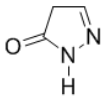
Pyrazolone

Heterocyclic compounds have gained popularity in recent years due to their pharmacological properties. Pyrazolones are valuable because of their broad range of biological activity and their versatility as synthetic tools for the development of a variety of bioactive compounds.

Pyrazolones are a group of chemicals with the nucleus of [1]H-pyrazol-3-ol and pyrazolin-3-one that have been studied for their diverse properties and applications. The analgesics and the antipyretic effect of the pyrazolone analogue have attracted a lot of attention since Knorr's synthesis of antipyrine in 1883. The discovery of these properties prompted researchers to create new pyrazolone compound with comparable properties but better therapeutic effect.

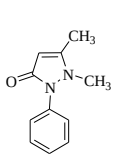


[1]H-Pyrazol-3-ol

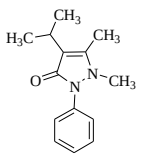


Pyrazolin-5-one

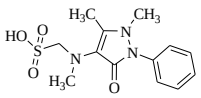
Drug like pyrazolone candidates with antibacterial [1], antioxidant [2], antitubercular [3], anticancer [4], antifungal [5], CNS impact, anti-inflammatory activity [6], and other therapeutic properties have been produced in medicinal chemistry research. Various synthetic pyrazolone compounds have potential core active moiety like, *analgesic and antipyretic*–antipyrine, propyphenazone, *anti-inflammatory*–metamizole, *uricosuric*–sulfinpyrazone. Meanwhile, the medicinal chemist has been drawn to attention by SAR research, and a state of analogues for a number of targets has been produced.



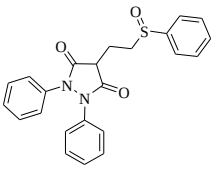
Antipyrine



Propyphenazone



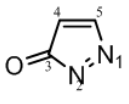
Metamizole



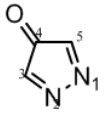
Sulfinpyrazole

The condensation of hydrazine hydrate and ethyl acetoacetate is a recognized way for synthesizing of pyrazolone derivatives. Dimethylformamide [7], methanol [8], dioxane [9], and ethanol [10] can be used as a solvent for this transformation.

3-Pyrazolone and 4-pyrazolone are two isomers of pyrazolone.

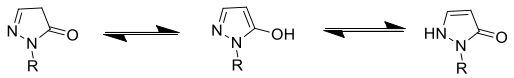


3-Pyrazolone



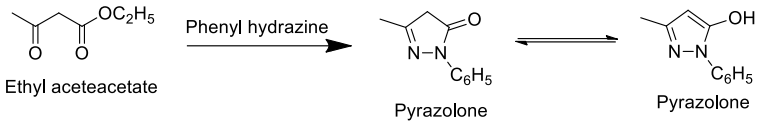
4-Pyrazolone

Lactam-lactim and imine-enamine tautomerism can be used to interconvert these isomers; photochromism is common in these conversions. The 3-Pyrazolone isomers can be stabilized by N-alkyl (or) N-aryl substituents in pyrazolone derivatives.



Synthesis of pyrazolone

Ludwig knoor [11] reported the first pyrazolone synthesis in 1883, using a condensation reaction between ethyl aceto acetate and phenylhydrazine.



Scheme 1: Synthesis of pyrazolone

Synthesis of 3-methyl-pyrazole-5-one

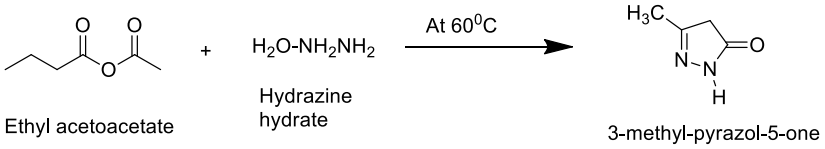
All methyl and phenyl Pyrazolone compounds were prepared with minor modification. In a 250 ml conical flask, 32.5g of ethylacetoacetate was placed and stirred magnetically while a solution of hydrazine hydrate was slowly added drop by drop (12.5g hydrazine hydrate in 20 ml ethanol). As the temperature of the reaction mixture rises during the reaction, it was kept at 60 °C. After 1 h of constant stirring, a crystalline deposit was formed. As the product crystallized, the mixture was chilled in an ice bath and the solid was filtered through a Buchner funnel before being washed in cold alcohol. The result was a white colourless crystal that was dried, recrystallized from ethanol.

Synthesis of 3-methyl-1-phenyl-pyrazolone derivative

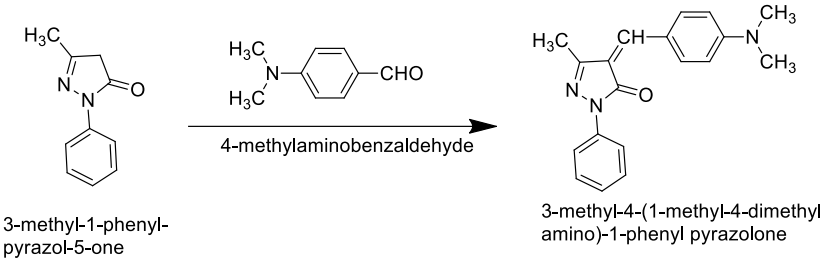
A 100 ml round bottom flask was filled with about 3.5g of synthesized pyrazolone and 60 ml of freshly prepared 20% Sodium hydroxide ethanolic solution, which was agitated for 30 min in a magnetic stirrer. 2.98g of 4-dimethyl aminobenzaldehyde was added to the reaction mixture and stirred for another 8-10 h with thin-layer-chromatography monitoring the reaction's completion. After that the reaction mixture was placed in crushed ice and neutralised with dilute HCl to precipitate the result, which was then frozen overnight. It was then filtered, dried, and recrystallized to purify it.

Synthesis of pyrazolone from chalcones

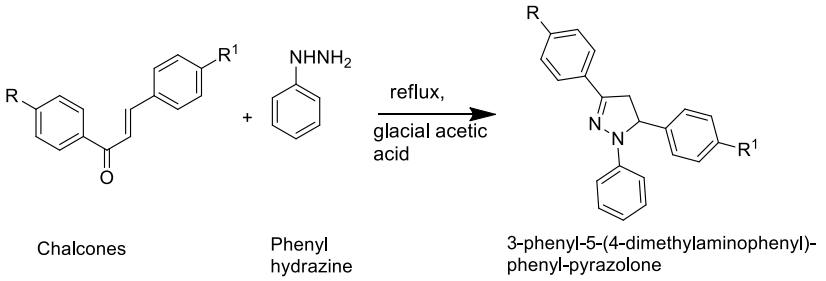
In an 80 ml glacial acetic acid, a mixture of synthesized 5.02g chalcones and 4.32g phenyl hydrazine was refluxed for 8 hours. As the solid precipitated, the mixture was cooled and poured over crushed ice, and the solid mass was filtered in the Buchner funnel, washed with cold water to remove the acid, dried and recrystallized from ethanol.



Scheme 2: Synthesis of 3-methyl-pyrazolone-5-one



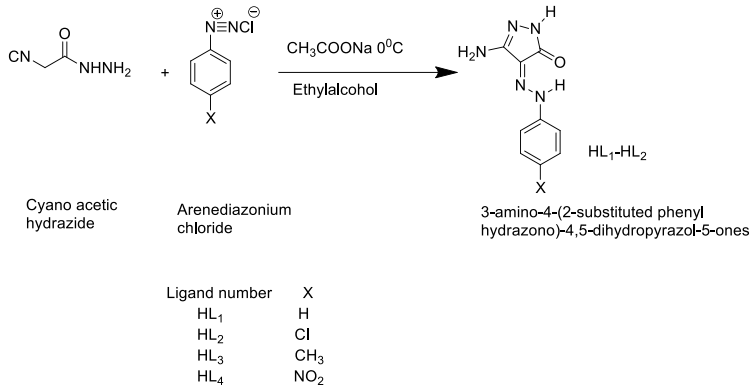
Scheme 3: Synthesis of 3-methyl-1-phenyl-pyrazolone derivatives



Scheme 4: Synthesis of pyrazolone from chalcones

Synthesis of 3-amino-4-(2-Substituted phenylhyrazono)-1-H-pyrazol-5(4H)-one

In the presence of sodium acetate trihydrate (1.36g, 0.01M) a freshly made solution of cyano acetic hydrazide (0.99g, 0.01M) in ethanol (50 ml) was linked. In an ice bath (at 0-5 °C), 0.01M arenediazonium chloride (made by diazotizing corresponding aniline derivative) was coupled with cold sodium nitrite solution (10 ml,1M)in 6M HCl (6 ml). The reaction mixture was refrigerated for three hours. The precipitating chemical was filtered and dried after the precipitation. The desired ligands 3-amino-4-(2-substituted phenylhydrazono)-4,5-dihydropyrazol-5-ones (HL[1-4]) are obtained by refluxing ethanolic arylhydrazone solution for 2 h.



Ligand number	X
HL_1	H
HL_2	Cl
HL_3	CH_3
HL_4	NO_2

Scheme 5: Synthesis of 3-Amino-4-(2-Substituted Phenylhydrazono)-1-H-Pyrazol-5(4H)-one

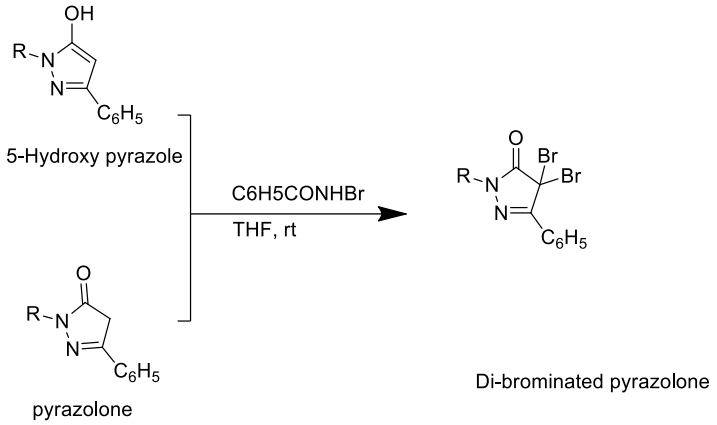
Synthesis of dibromopyrazolones

Halogenated pyrazolones have previously been found to be valuable synthetic intermediates for the manufacture of dyes, fused and Spiro heterocyclic compounds. Bromo acetic acid, bromine water, N-bromo succinimide (NBS) can all be used to make brominated pyrazolones. Using pyrazolone or hydroxyl pyrazolones and N-bromobenzamide, Huang *et al.* [12], produced di-bromopyrazolones with a product yield of 90%.

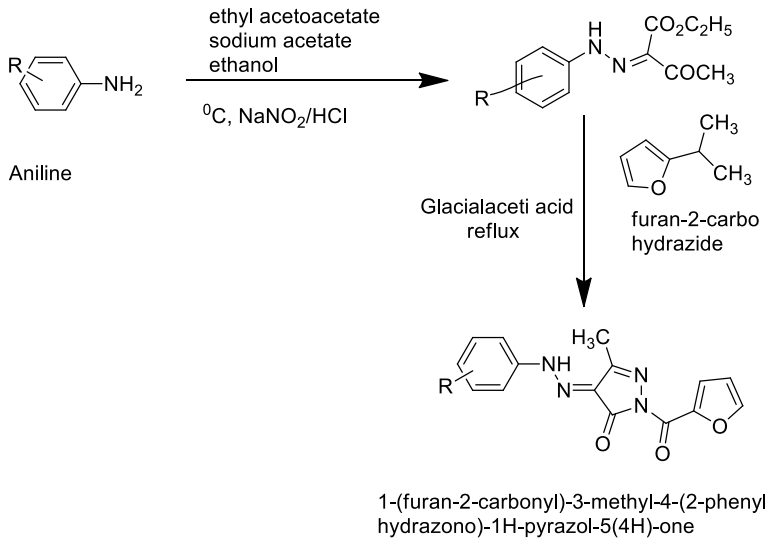
Synthesis of pyrazolone derivatives

Synthesis of 1-(Furan-2-Carbonyl)-3-Methyl-4-(2-Pheynl Hydrazono)-1H-Pyrazol-5(4H)-one by mannich reaction

The Mannich reaction of different ethyl has been reported by Shah *et al.* [13]. Furan-2-carbohydrazide was used to make 2-substituted phenyl hydrazono-3-oxobutyrates,1-(furan-2-carbonyl)-3-methyl-2-(phenyl hydrazono)-4-(phenyl hydrazono)-4-1H pyrazol-5(4H)-one.



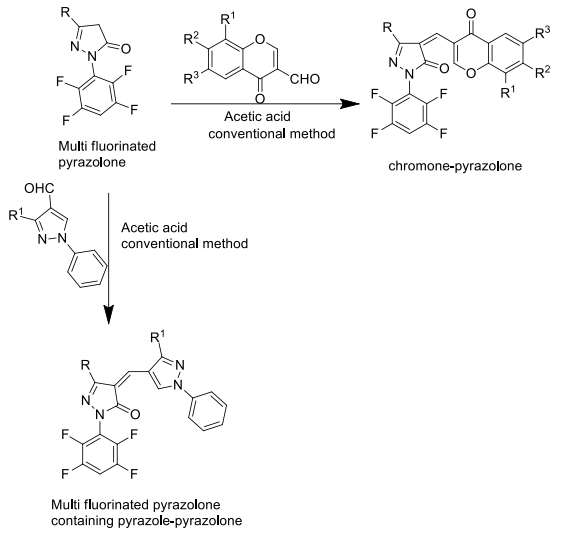
Scheme 6: Synthesis of dibromopyrazolones



Scheme 7: Synthesis of 1-(Furan-2-Carbonyl)-3-Methyl-4-(2-Phenyl Hydrazono)-1H1Pyrazol-5(4H)-one by mannich reaction

Synthesis of 2-Aryl-5-Methyl-2,3Dihyro-1H-3-pyrazolones using cul nanoparticles

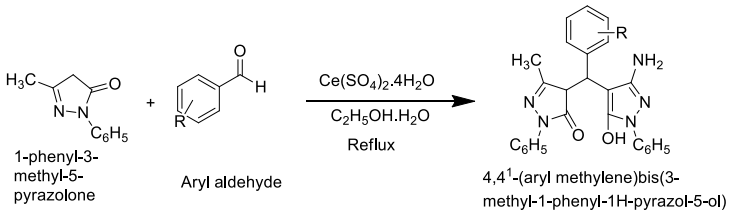
The knoevenagel condensation procedure was used by Gadhave *et al.* [14], to synthesize a series of novel fluorine-containing pyrazole-pyrazolone and chromone-pyrazole from multifluorinated pyrazolone. All of the compounds were created using a combination of conventional heating and ultrasonic irradiation. The ultrasonication approach was shown to be more efficient than the traditional heating method.



Scheme 8: Synthesis of 2-Aryl-5-Methyl-2,3 Dihydro-1H-3-Pyrazolones using cul nanoparticles

Synthesis of 4,4′-(Aryl methylene)-Bis(3-Methyl-1-phenyl-1H-pyrazol-5-ol) by using $Ce(SO_4)_2.4H_2O$

Mosaddegh [15] and his colleagues, the reaction of aryl aldehyde and 1-phenyl-3-methyl-5-pyrazolone in the presence of a catalytic amount of $Ce(SO_4)_2.4H_2O$ as reusable and environmentally friendly catalyst in water/ethanol solution was reported to be effective in producing 4,4-(arylmethylene)bis(3-methyl-1-phenyl-1H pyrazol-5-ol). High yields, a fast reaction time, easy set-up and reusability of the catalyst are all advantages of this approach.



Scheme 9: Synthesis of 4,4[1]-(Arylmethylene)Bis-(3-Methyl-1-Phenyl-1H-Pyrazol-5-ol) by using Ce(So4)

Synthesis of 4,4′-Arylmethylene)Bis(1H-Pyrazol-5-ols) using ceric ammonium nitrate as catalyst

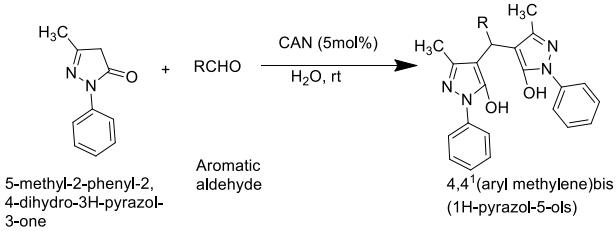
Sujatha *et al.* [16], described the preparation of 4,4-(arylmethylene)bis(1H-pyrazol-5-ols) by a tandem knoevenagel-micheal reaction of two equivalent of 5-methyl-2-phenyl-2,4-dihydro-3H-pyrazol-3-one with different aromatic aldehydes in water catalysed by ceric ammonium nitrate.

Synthesis of photochromic pyrazolones based on photochromish by employing posphor $Sr_2P_2O_7$

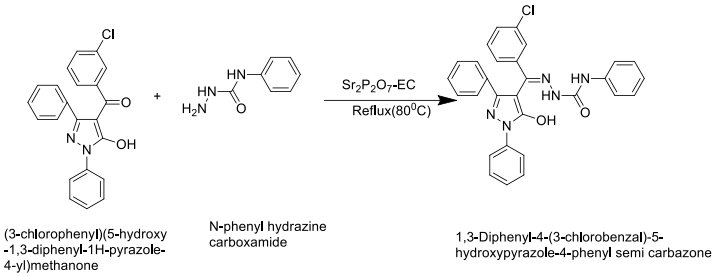
Using the phosphor $Sr_2P_2O_7$ co-doped with europium ion and chlorine ion ($Sr_2P_2O_7$-EC), Lui *et al.* [17], created a novel solid-state reversible fluorescence photoswitching system(FPS) based on photochromism of photochromic pyrazolones. As the fluorescent dye and the photochromic molecule, (3-chlorophenyl) (5-hydroxy-1,3-diphenyl-1H-pyrazol-4-yl) methanone reacts with N-phenyl hydrazine carboxamide to create 1,3 diphenyl-4-(3-chlorobenzal)-5-hydroxy pyrazole-4-phenyl semicarbazone.

Synthesis of new pyrazolones containing a phenothiazine unit

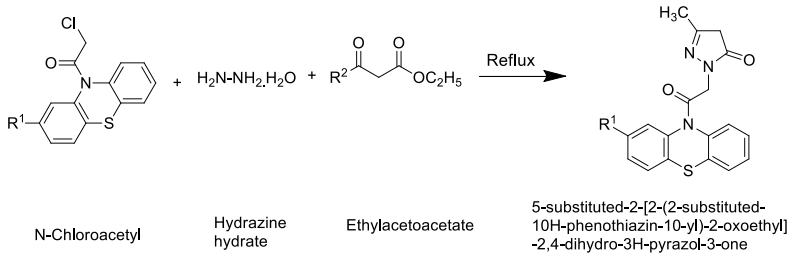
A straightforward one-pot technique for the synthesis of novel 5-substituted-2-[2-(2-substituted-10H-phenothiazin 10-yl)-2-oxoethyl] has been reported by Baciu-Atudosie *et al.* [18]. N-chloroacetyl compound, ethyl acetoacetate and hydrazine hydrate react to form 2,4-dihydro-3H-pyrazol-3-one with a phenothiazine unit.



Scheme 10: Synthesis of 4,4[1]-(Arylmetylene)Bis(1H-Pyrazol-5-ols) using ceric ammonium nitrate as catalyst



Scheme 11: Synthesis of photochromic pyrazolones based on photochromism by employing phosphor $Sr_2P_2O_7$



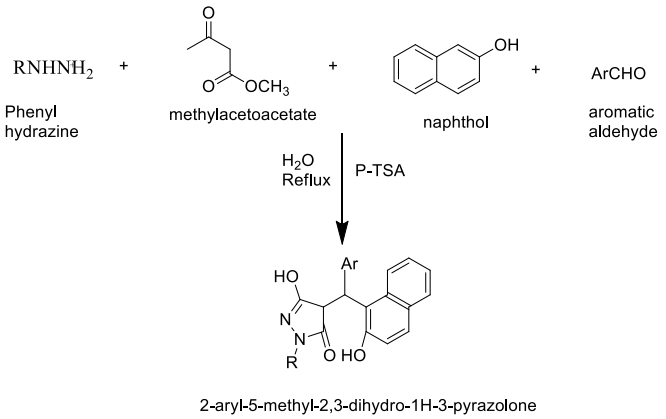
Scheme 12: Synthesis of new pyrazolones containing a phenothiazine unit

Synthesis of 2-Aryl-5-methyl-2,3-Dihydro-1H-3-pyrazolones in the presence of P-Tsa

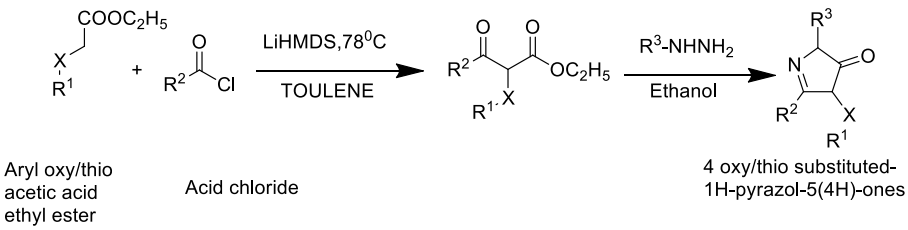
In a one-pot, the four-component sequential reaction of phenyl hydrazine, methyl acetoacetate, naphthol and aromatic aldehydes in the presence of p-toluenesulphonic acid in water, Gunasekaran *et al.* [19] reported the synthesis of a sequence of 2-aryl-5-methyl-2,3dihydro-1H-3-pyrazolones.

Synthesis of 4-Oxy/Thio substituted pyrazolones *via* cross-claisen condensation

According to Ragavan *et al.* [20] aryl oxy/thioacetic acid ethyl ester were synthesized via an efficient cross-claisen with acid chlorides forming an intermediate, which was then converted into 4-oxy/thio substituted-1H pyrazol 5(4H)-ones by the addition of hydrazine or hydrazine derivatives.



Scheme 13: Synthesis of 2-Aryl-5-Methyl-2,3-Dihyro-1H-3-pyrazolones in the presence of P-Tsa

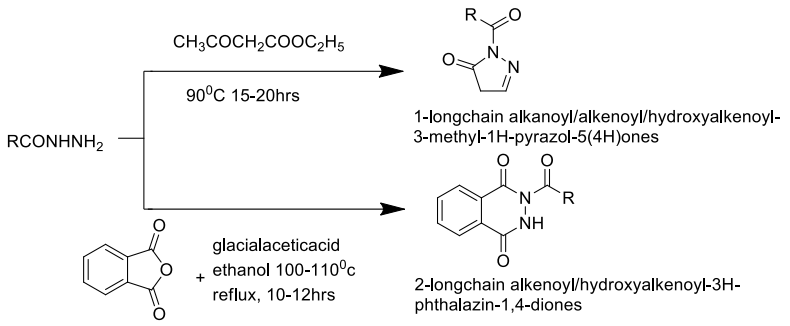


Scheme 14: Synthesis of 4-Oxy/Thio substituted pyrazolones via cross-claisen condensation

Synthesis of Hydroxyalkenoyl-3-Methyl-1H-Pyrazol-5(4H)-ones

Two unique series of 1-long chain alkanoyl/alkenoyl/hydroxyalkenoyl-3-methyl-1H-pyrazol-5(4H)-ones and 2-long chain alkenoyl/hydroxyalkenoyl-3H-phthalazin-1,4-diones have been described by Ahmad *et al.* [21]. The reaction of ethylacetoacetate with pthalic anhydride and hydrazides produces

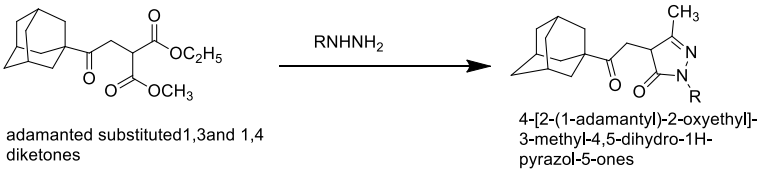
it. The cyclization reaction between ethylacetoacetate and hydrazides produces compound 1-long chain alkanoyl/alkenoyl/hydroxyalkenoyl-3-methyl-1H-pyrazol-5(4H)-ones. The reaction of phthalic anhydride and hydrazides in 100% ethanol or glacial acetic acid yielded compound 2-long chain alkenoyl/hydroxyalkenoyl-3H phthalazin-1,4-diones.



Scheme 15: Synthesis of hydroxyalkenoyl-3-methyl-1H-pyrazol-5(4H)-ones

Synthesis of dihydropyrazolones containingan adamantane fragment

Various pyrazole and dihydropyrazolones with an adamantane fragment were synthesized by Konkov *et al.* [22] from adamantyl-substituted 1,3 and 1,4-diketones, ethyl 4-(1-adamantyl)-2-R-4-oxobutanates (R=CN,Ac) and ethyl 2-(1-adamantyl)-2(1-adamantyl carbonyl)-4-oxo-4-phenyl butanoate. 4-[2-(1-adamantyl)-2-oxoethyl)-4-[2-(1-adamantyl)-2-oxoethyl]-4-[2-(1-adamantyl)-2-oxoethyl]-4-[2-(1-adamantyl)-2-oxoethyl]-4-[2-3-methyl-4,5-dihydro-1H-pyrazol-5-ones.



Scheme 16: Synthesis of dihydropyrazolones containing an adamantane fragment

Synthesis of bisindolyl pyrazolone derivatives

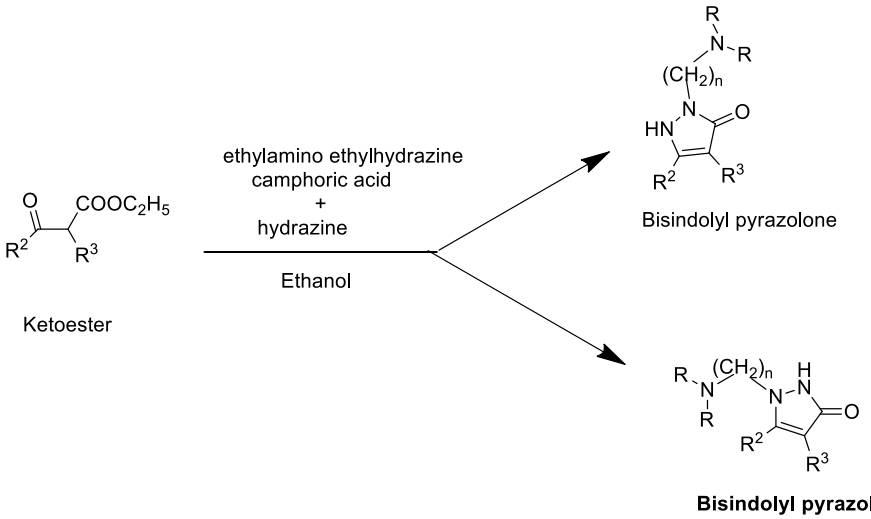
By reacting substituted ketoester, camphoric acid and ethyl amino ethyl hydrazine in ethanol medium. Bran *et al.* [23] described a series of bisndolyl pyrazolone derivatives.

Synthesis of nanosteroidal pyrazolones

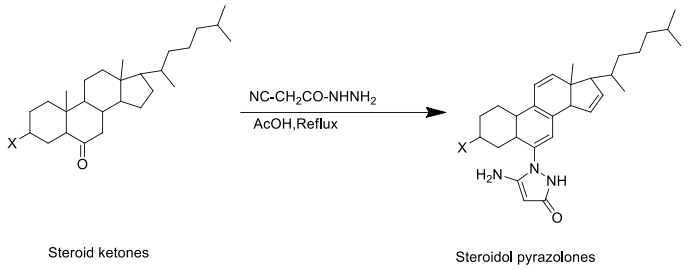
A convenient synthesis of a new series of nano steroidal pyrazolones was reported by Shamsuzzaman *et al.* [24]. Cyanoacetohydrazide was added in an equimolar ratio to a solution of steroidal ketones in acetic acid, for 7 h the reaction mixtures was stirred under reflux.

Synthesis of benzylpyrazolyl coumarins

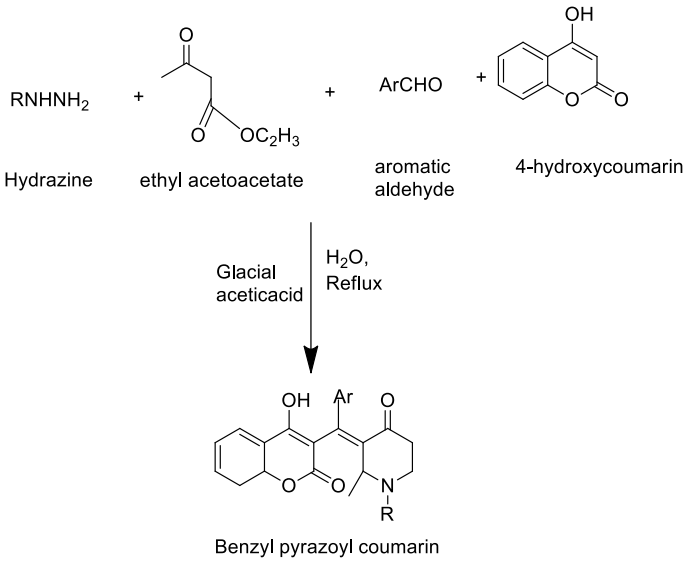
Ghosh *et al.* [25] reported a glacial acetic acid catalysed reaction for the combinatorial synthesis of highly functionalized benzylpyrazolyl coumarin prepared by a green pot four-component reaction in water medium under refluxing conditions involving aryl hydrazine/hydrazine hydrate, ethylacetoacetate, aromatic aldehydes and 4-hydroxy coumarin.



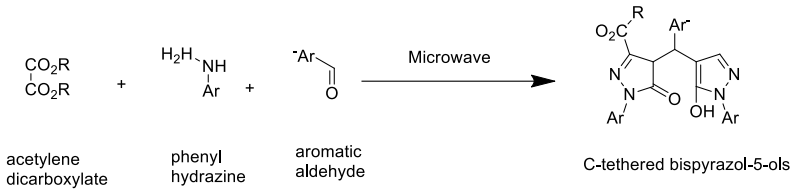
Scheme 17: Synthesis of bisindolylpyrazolone derivatives



Scheme 18: Synthesis of nanosteroidal pyrazolones



Scheme 19: Synthesis of benzylpyrazolyl coumarins



Scheme 20: Synthesis of C-tethered bispyrazol-5-ols

Synthesis of C-tethered bispyrazol-5-ols

Tu *et al.* [26] have been integrated C-fastened bispyrazol-5-ols through multicomponent domino responses of acetylenedicarboxylate, phenylhydrazine and sweet-smelling aldehydes under microwave illumination.

CONCLUSION

According to research, pyrazolone derivatives have antibacterial, anti-inflammatory, analgesic, anticancer, and antitubercular properties. The numerous synthetic approaches used to generate a physiologically rich pyrazolone moiety are discussed in this paper. Slight changes to the substituents on the basic pyrazolone nucleus can further boost the activity. This page serves as a valuable resource for future study on the bioactive pyrazolone ring as well as a tool for the development of improved therapeutic drugs.

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AUTHORS CONTRIBUTIONS

All the authors have contributed equally.

CONFLICT OF INTERESTS

The authors have no conflicts of interest regarding this investigation.

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