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Synthesis of Pyrazole Derivatives A Review

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Abstract
This review deals with various method of synthesis of pyrazole derivatives the pyrazole derivatives have numerous pharmacological activities from few decades pyrazole molecule synthesis has became an important strategy for development of drug like substances. So these type five membered heterocyclic compounds have targeted for formulation of new pharmaceutical active ingredient. Here an attempt made to overview of different methods of synthesis of pyrazole derivatives and their synthetic methods.

Keywords: Pyrazole, pheny pyrazole, pyrazole carbothiamide, pyrazole carboxamide

Introduction

The pyrazole derivatives active as cytotoxic [1] and having antitumor activity [2-3] as theses have broad spectrum against cancer cell hence act as anticancer agent [4-7]. The pyrazole derivatives are function as genotxic agent [8] that aims to induce DNA Damage of cancer cell. It is an important drug for treatment of lung cancer [9] and it also possess antileukemic activity [10]. Particularly in field of medical science and drug innovation the derivatives of pyrazole play a significant role as it found to possess Inhibitor of Angiogenesis [11-12] Anti-angiogenic activity [13] it is vital cancer fighting agent they block growth of blood vessel that supports tumor growth which play noteworthy role in the treatment of cancer by considering its applications in the field of cancer research and cancer treatment protocol the synthesis pyrazole derivative became more important strategy for development of life saving drugs . Recently several drugs have been developed from pyrazole derivatives the drug like Celecoxib acts as anti-inflammatory agent and remonabunt used in the treatment of obesity [14]. Some important synthetic methods of pyrazole derivatives are reviewed here

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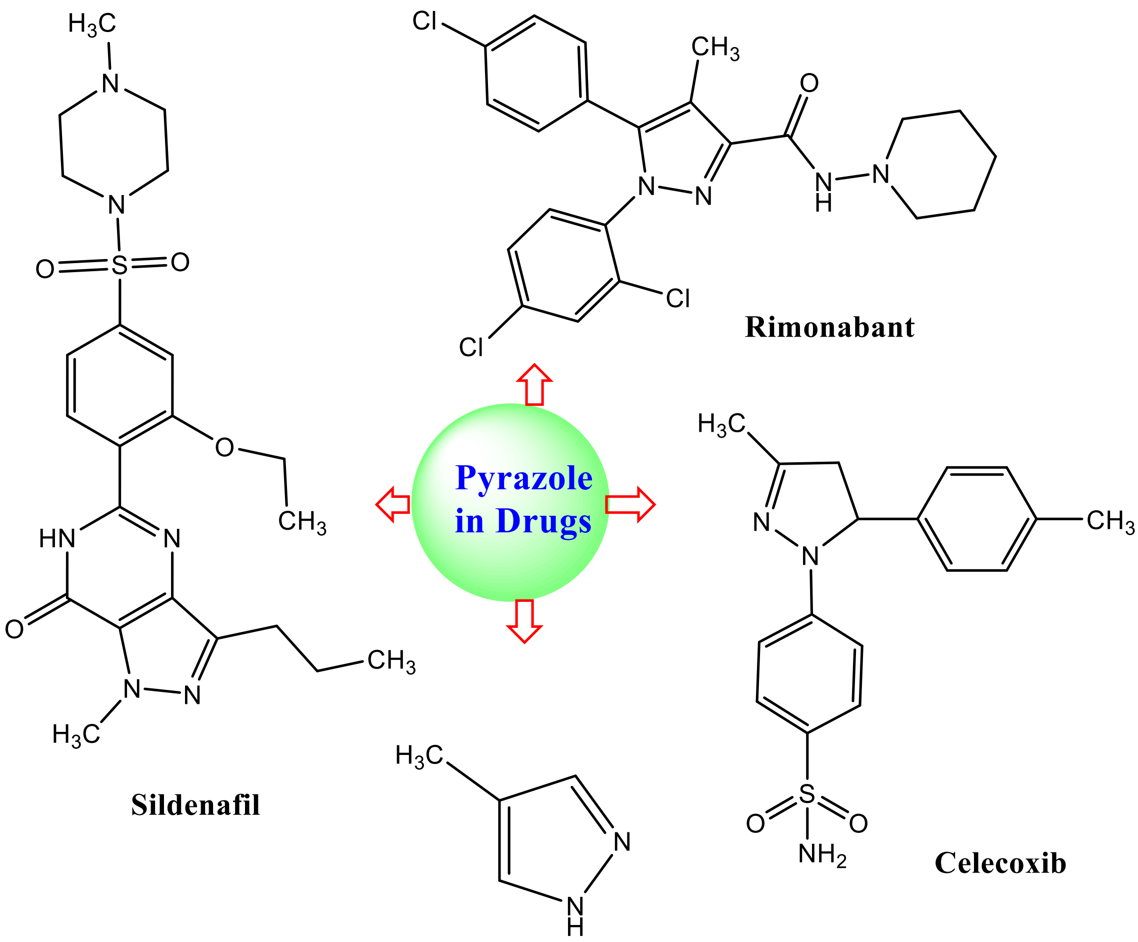
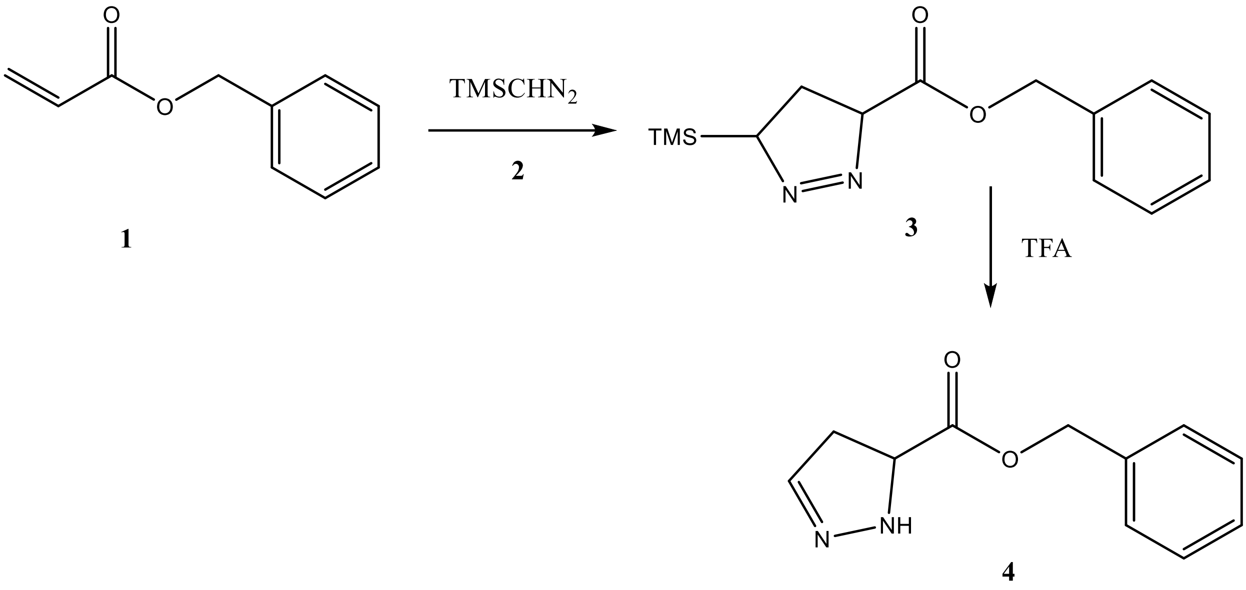
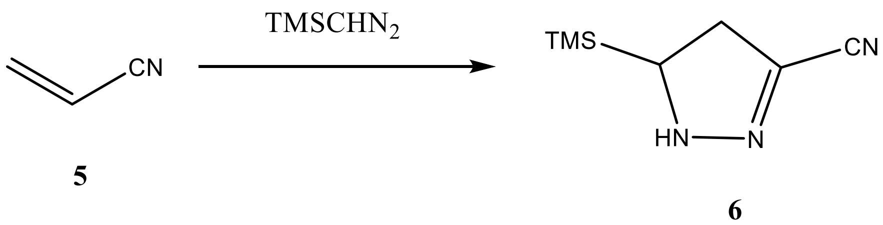


Fig: Drug molecule containing pyrazole scaffold

Jin Hee Ahn et al **[15]** have been extensively studied and developed Method of synthesis of pyrazole **4** by reaction of benzyl acrylate **1** with trimethyl silane diazomethane 2 converted in to an intermediate 3 which on further treatment with tetra flouro acetic acid afforded to substituted pyrazole **4** shown in (**Scheme-1)**. Similarly cyanoethene 5 on treatment with reagents trimethyl silane diazomethane **2** leading to cyclisation with formation of pyrazole derivative **6** (**Scheme-2)**

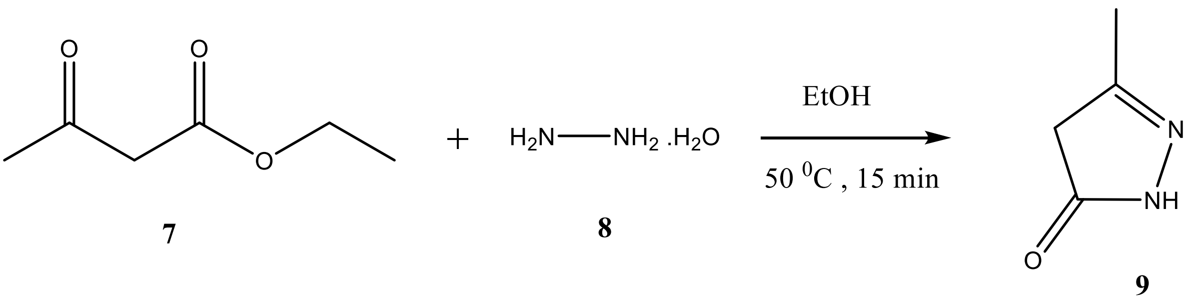


Scheme 1



Scheme 2

R Mallikarjun Rao et al [16] have formulated ketopyrazole derivative **9** in this synthesis ethyl-3oxobutanoate **7** condensed with hydrazine hydrate **8** in presence of ethanol as solvent refluxed reaction mixture for 15 min at 50 ^{0}C temperature converted in to compound **9** (**Scheme-3)**



Scheme 3

Kumar S et al [17] have synthesized amino phenyl substituted pyrazole derivative this have achieved by condensation reaction of chalcone **10** with hydrazine hydrate **11** in presence of tertiary butyl hydroxide

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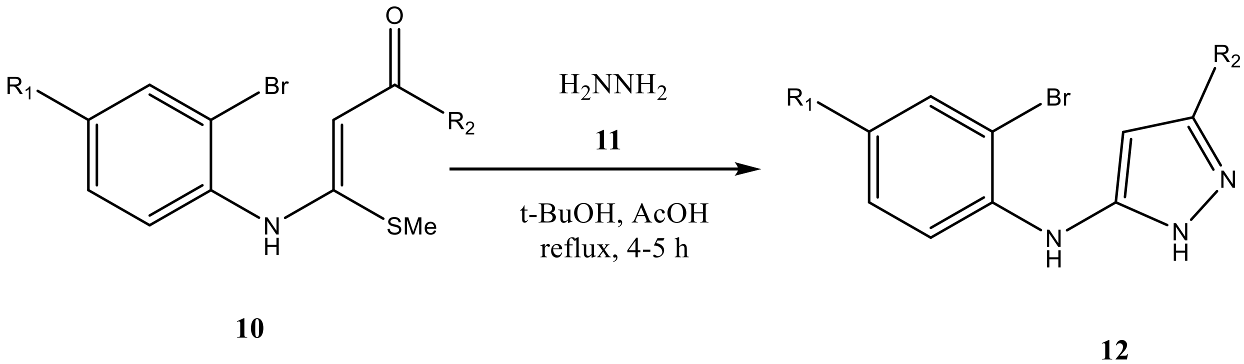
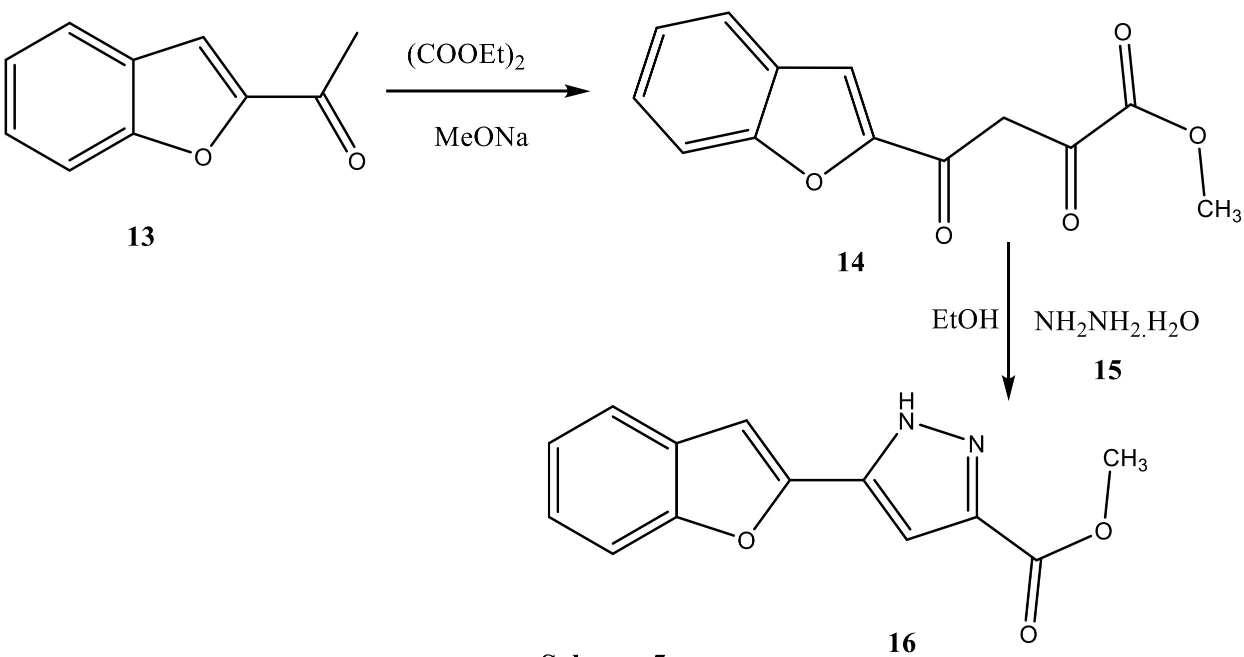
and in acidic medium and reaction mixture was refluxed for 4-5 hours results in formation of pyrazole derivative **12** **(Scheme-4)**



Scheme 4

Naqui-Jahan Siddiqui and co workers [18] have develop synthsis of benzofuran substituted pyrazole from 1-(benzofuran-2-yl)ethan-1-one **13** which on treatment with diethyl oxalate in presence of sodium methoxide as a base , which initiated formation of benzofuran carbanion which on attacked on diethyl oxalate and converted in to compound **14**. Which is further condensed with hydrazine hydrate **15** in presence ethanol resulted in to formation pyrol derivative **16**. **(Scheme-5)**



Scheme 5

Zhibing Wu et al [19] have devised synthesis of pyrol derivative **20** by reaction of dicorbonyl compound **17** on treatment with triethoxy methane in presenc of acetic unhydride converted in to compound **18** which

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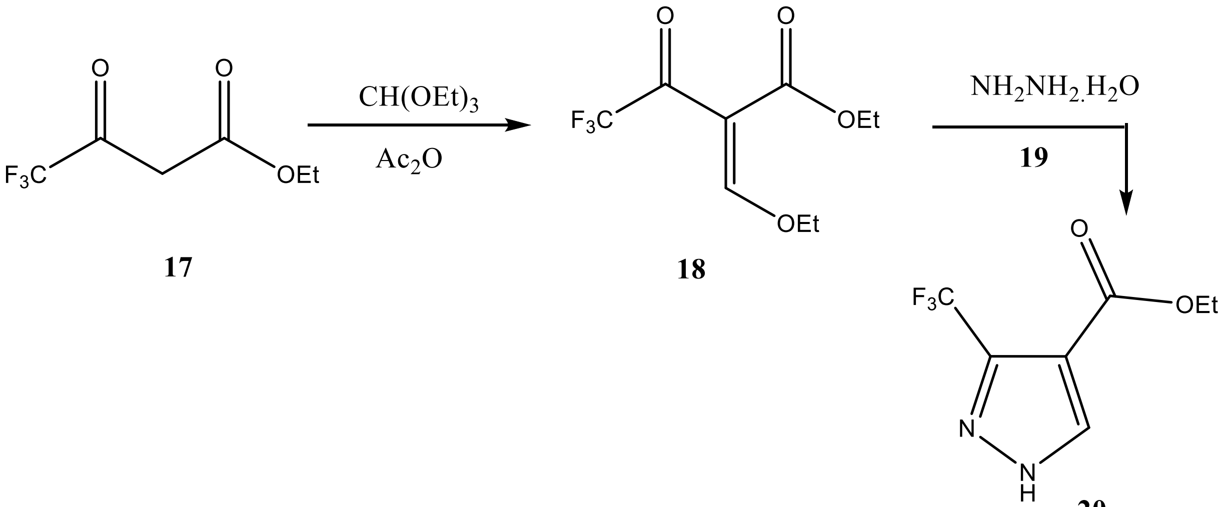
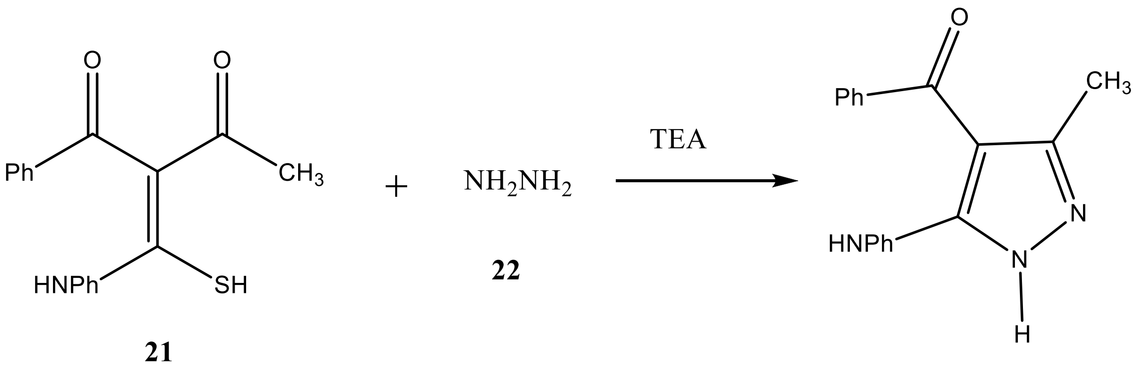
is further reacted with hydrazine unhydrate **19** due to ringclosure reaction resulted in to formation of pyrol derivative **20** **(Scheme-6)**



17 **18** **20**

Scheme 6

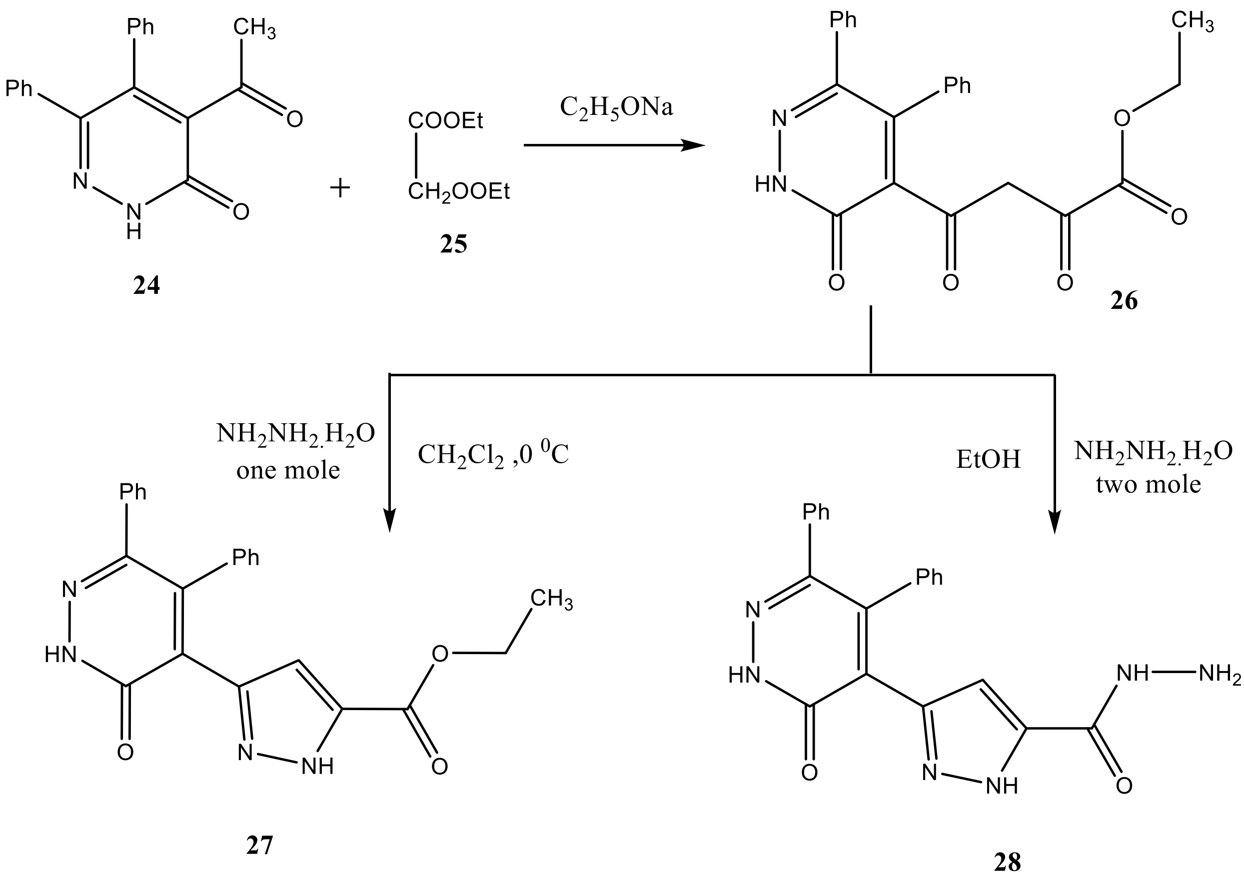
Fadda A A, et al [20] have develop method of formulation of compound **23** in this synthetic protocol substituted chlacone **21** and hydrazine condensed in presence of basic condition the base used in this protocol was tri ethyl amine furnished in to substituted pyrazole derivative **23** **(Scheme-7)**



21 **22** **23**

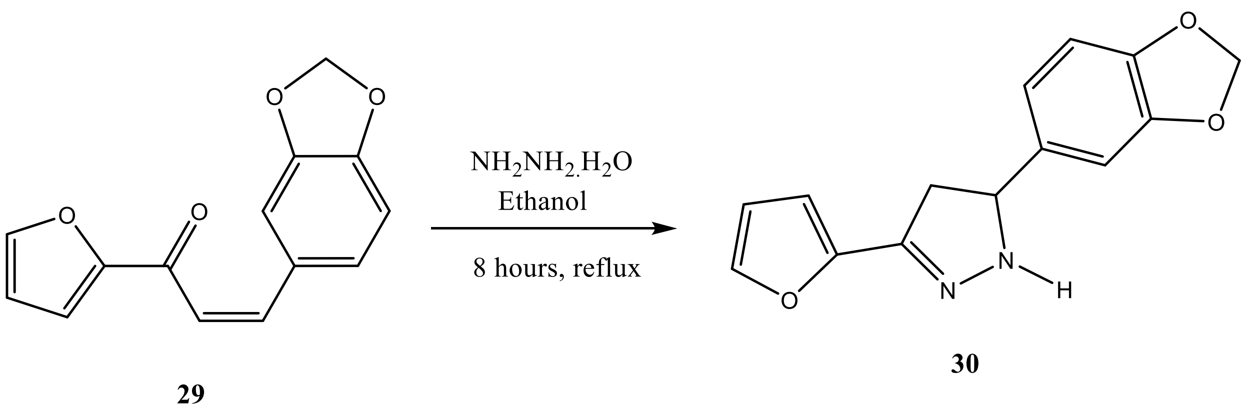
Scheme 7

F A Yassin et al [21] have developed synthesis of pyrimidine substituted pyrazole in this synthetic protocol started from 4-acetyl-5,6-diphenylpyridazin-3-one 24 reacted with ethyl 2-(ethylperoxy)acetate **25** in presence of sodium ethoxide afforded to compound **26** which is treated with hydrazine hydrate at two different conditions in first reaction condition methylene chloride is use as solvent and 1 mole of hydrazine hydrate was used at 0 ^{0}C temperature converted in to pyrazole **27**. In second strategy solvent used as ethanol with 2 mole of hydrazine hydrate and converted in to pyrazole derivative **28** **(Scheme-8)**



Scheme 8

Vidhya K R et al [22] have devised synthesis of furanyl substituted pyrazole derivative **30** by reaction chlacone **29** with hydrazine hydrate in presence of ethanol furnished in to compound **30**. **(Scheme-9)**



Scheme 9

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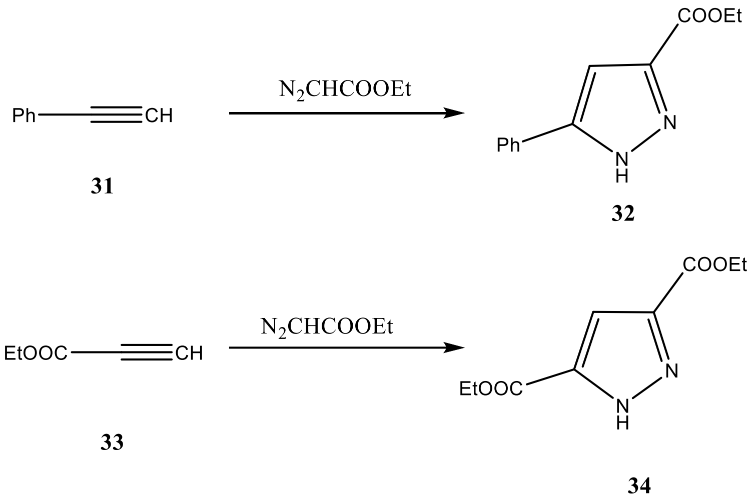
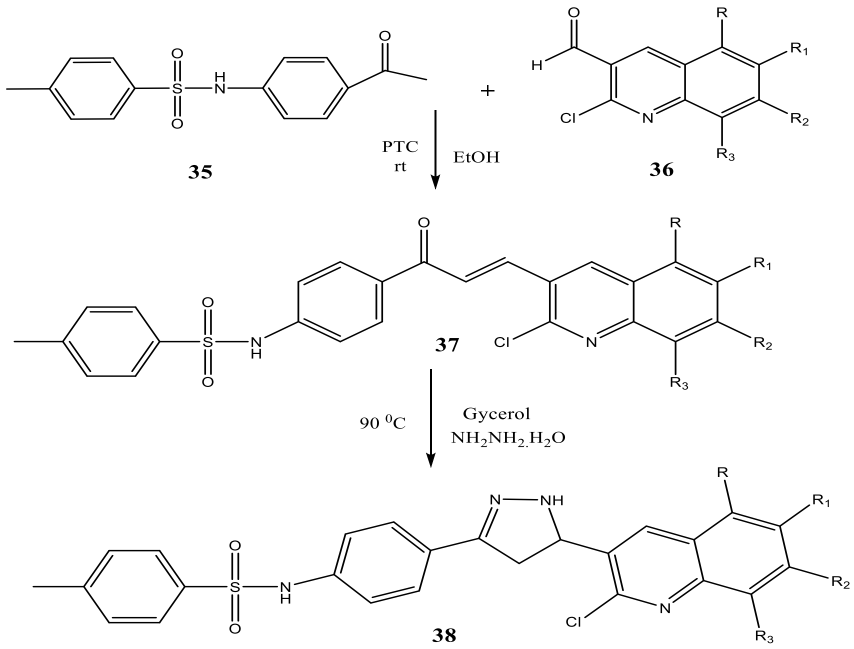
Krishna P R et al [23] have develop synthesis by reaction of ethynylbenzene **31** with ethyl 2-diazoacetate converted in to pyrazole **32** similarly cycloaddition type reaction between compound 32 with ethyl 2-diazoacetate resulted in to compound **34**. **(Scheme-10)**



Scheme 10

Vasant B Jagrut et al [24] have formulated preparation substituted pyrazole **38** by reaction of ketone **35** with an aldehyde **36** in presence of Phase transfer catalyst in ethanol solvent and reaction conducted at room temperature afforded to chalcone **37** . This on treatment with hydrazine hydrate and in presence of glycerin and at 90 ^{0}C temperature converted in to compound **38**. **(Scheme-11)**



Scheme 11

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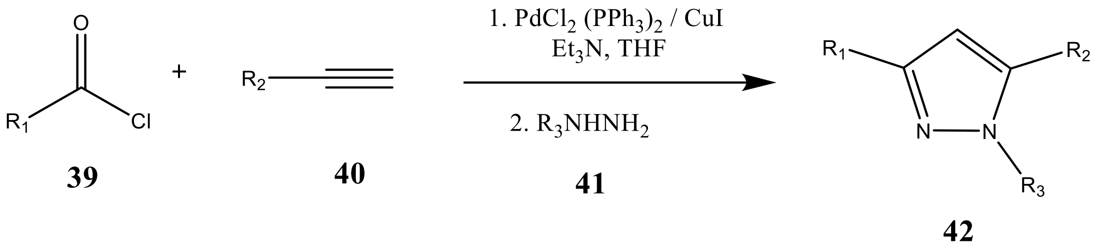
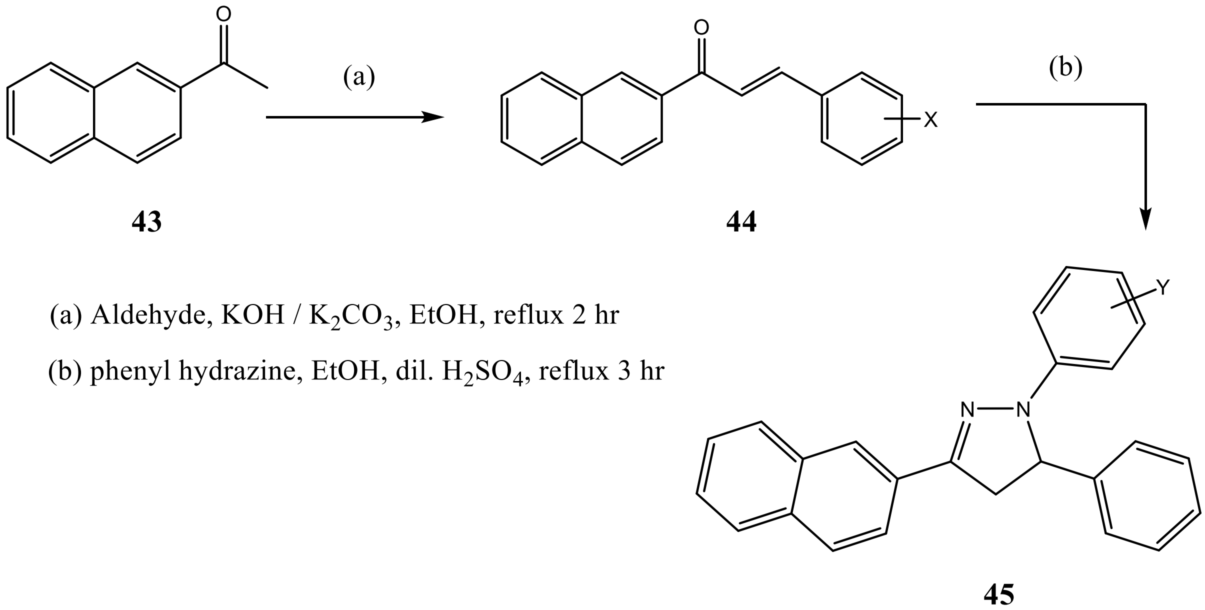
Phenyl pyrazole

Hai-Liang, Huan-Feng Jiang et al [25] have developed synthetic protocol in this process acyl chloride **39** reacted with terminal alkyene **40** and substituted hydrazine **41** in presence of palladium chloride with triphenyl phospine and cupric iodide triethyl amine and tetra hydro furan as solvent converted in to N-substituted pyrazole derivative **42** (**Scheme-12)**



Scheme 12

Beena Varghese, Saleh N Al-Busafi et al [26] have formulation of compound **45** by reaction of ketone **43** with benzaldehyde furnished in to chalcone **44** which on further condensed with phenyl hydrazine converted in to N-phenyl substituted pyrazole derivative **45** (**Scheme-13)**



Scheme 13

Alinezhad H, Tajbakhsh M et al [27] develop synthesis of N-substituted pyrazole by condensation reaction of dicorbonyl **46** with phenyl hydrazine **47** in presence of silica in acidic condition and N-bromo succinamide resulted in to the formation of compound **48** and compound **49** (**Scheme-14)**

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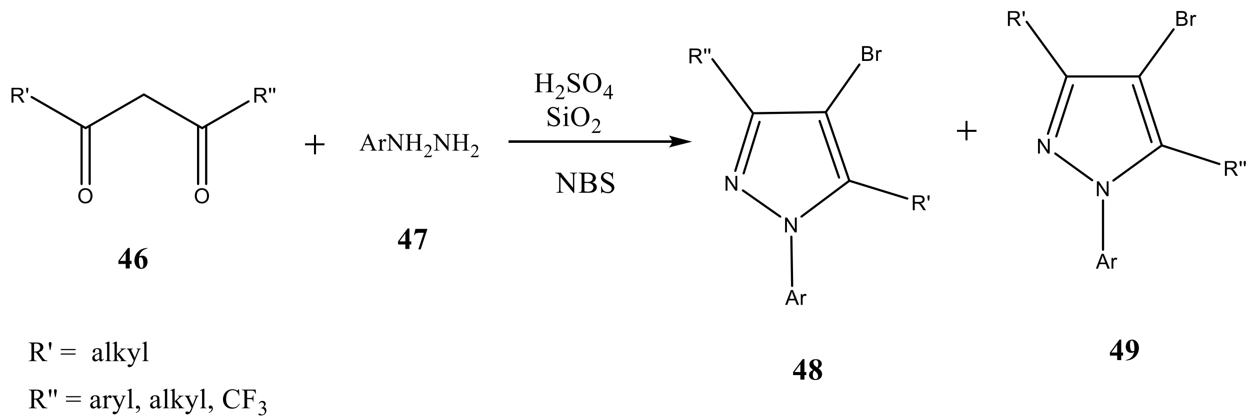
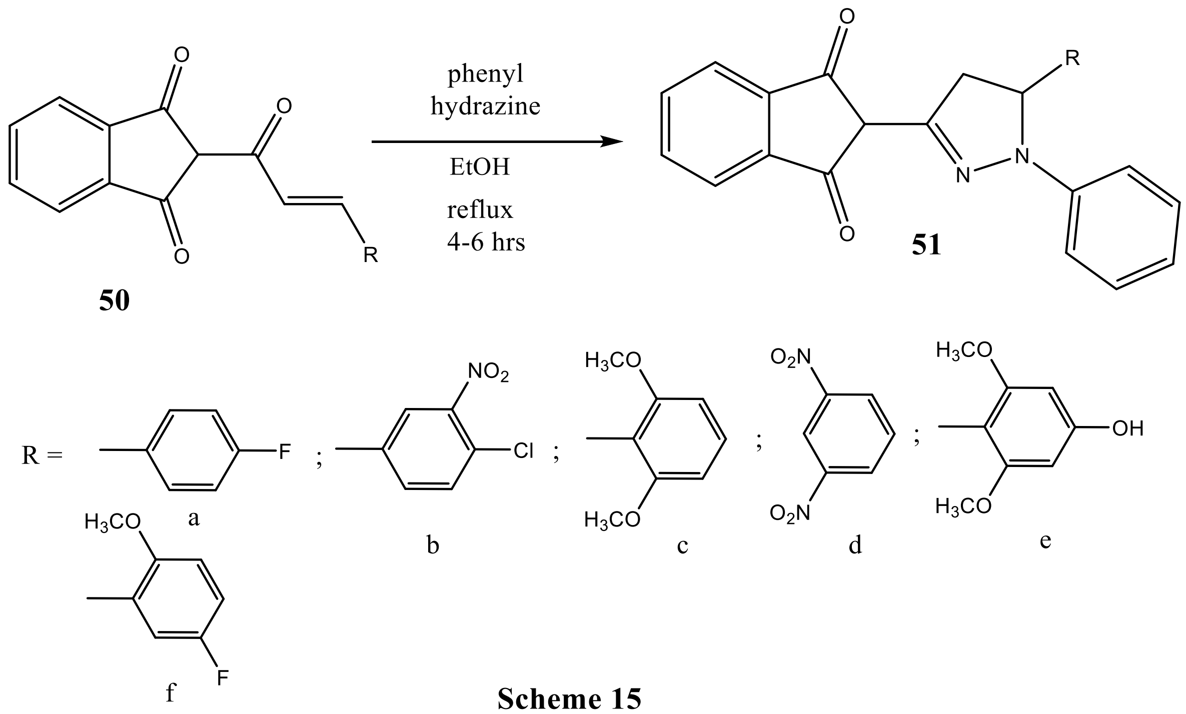


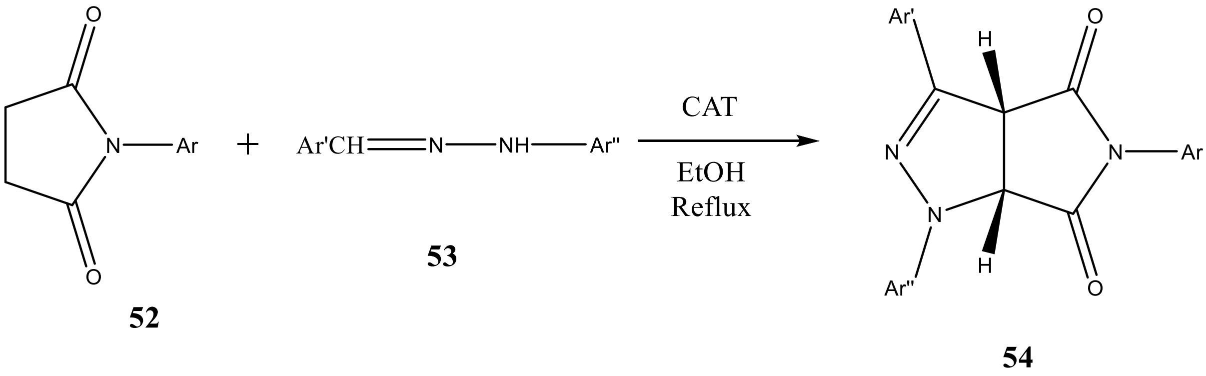
Scheme 14

Siddharth S Desai et al [28]have developed synthesis of N-phenyl substituted pyrazole have carried out by reaction of chalcone **50** with Phenyl hydrazine which is then refluxed in presence of ethyl alcohol for 4-6 hours converted in to compound **51(Scheme-15)**



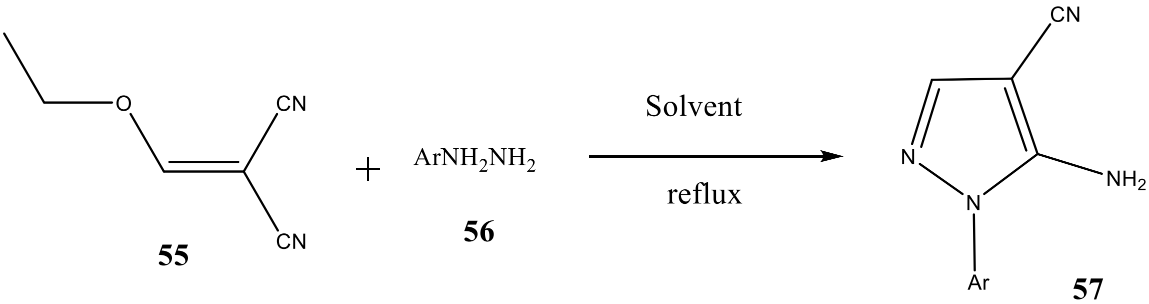
Scheme 15

Umesha K B et al [29] have developed synthesis of N- phenyl substituted pyrazole from 1,3 dipolar cycloaddition reaction of N-phenyl succinamide **52** and 1-benzylidene-2-phenylhydrazine **53** in presence of catalyst Chloroamine-T (CAT) and refluxed in ethanol afforded to N-substituted pyrazole derivative **54 (Scheme-16)**



Scheme 16

Silvana C Palem et al [30] have reported synthesis of phenyl pyrazole by reaction of 2-(ethoxymethylene) malononitrile **55** with phenyl hydrazine after refluxed it has been converted in to phenyl pyrazole **57** **(Scheme-17)**



Ar = a: C_6H_5, b: 4-F-C_6H_4, c:C_6F_5, d: 4-CF_3-C_6H_5, e: 2,6-Cl_{2-4}-C_6H_2, f:4CH_3O-C_6H_4

Scheme 17

Sreedhar B et al [31] have reported synthesis of N- phenyl pyrazole developed N-arylation strategy in this synthetic method pyrazole **61** on treatment with arylboronic acid **62** in presence of cupric oxide as a catalyst in presence of methanol as solvent and reaction was carried out at room temperature resulted in to N- phenyl pyrazole **63**(**Scheme-18)**

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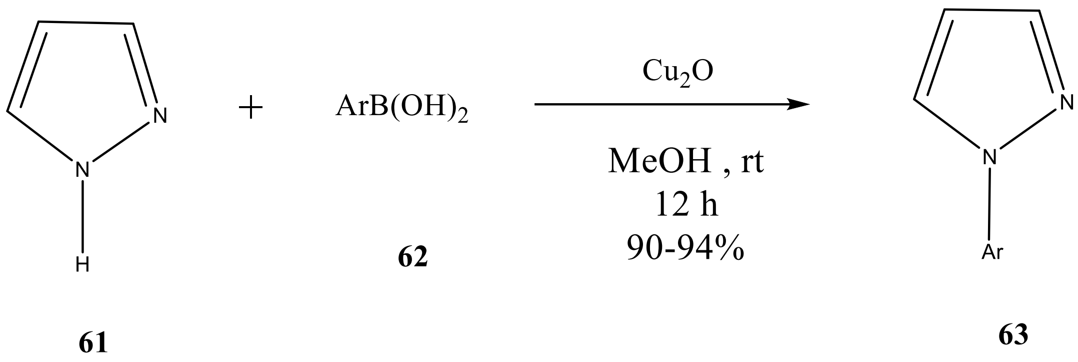
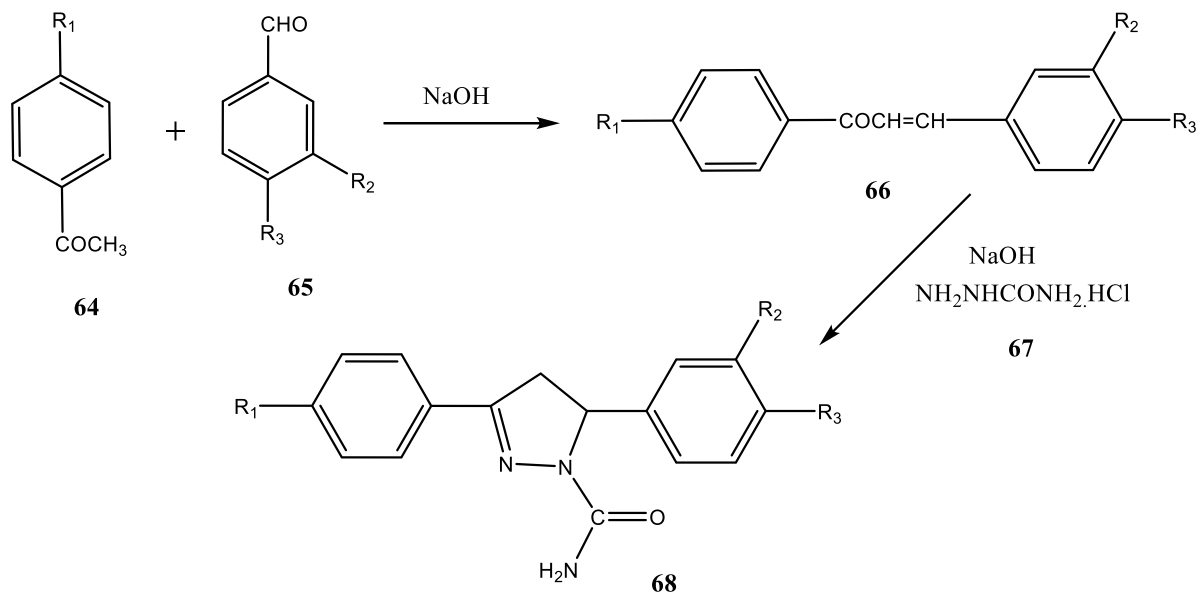


Scheme 18

Pyrazole carboxamide

Anees S Siddiqui et al [32] have developed synthesis of pyrazole carboamide by formation of chalcone **66** as an intermediate which have been synthesized from ketone **64** and aldehyde **65** in presence of base. Then the chalcone on condensation with semicarbazide **67** in presence of base converted in to pyrazole derivative **68** **(Scheme-19)**



Scheme 19

Bhagwat Jadhav et al [33] have developed synthesis of condensed pyridine pyrazole carboxamide derivative by reaction of amino pyridine **69** with ethyl 2-chloro-3-oxobutanoate **70** in presence of ethanol and refluxed it for 80 ^{0}C temperature converted in to 6-bromo-2,7,8-trimethylimidazo[1,2-a] pyridine-3-carboxylate **71** which on lithium hydroxide a soft base in presence of tetra hydro furan and water converted

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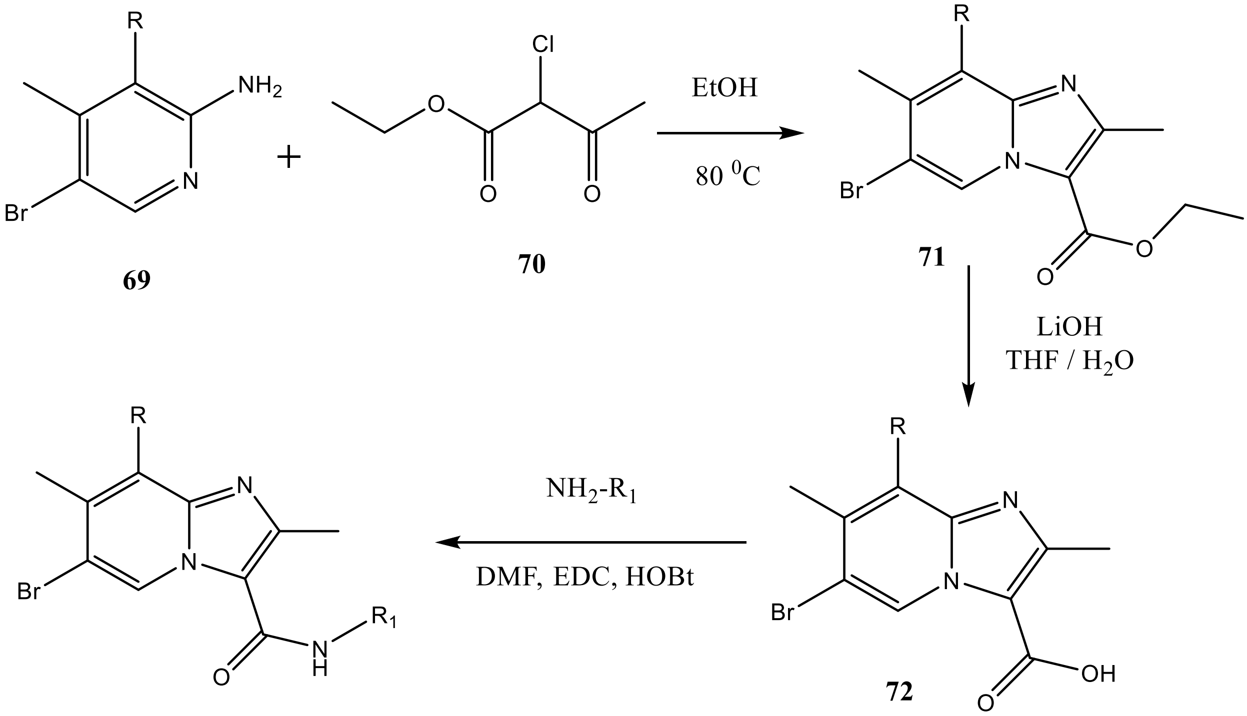
in to carboxylic acid **72** which on treatment with primary amine converted in to pyridine pyrazole carboxamide derivative **73.** **(Scheme-20)**

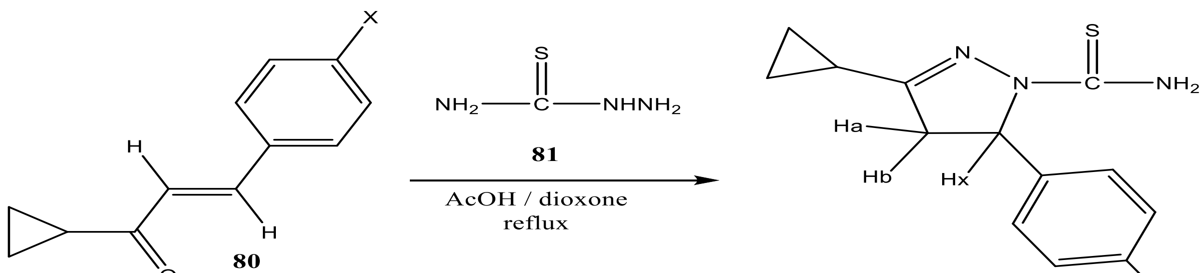


69 70 71 72 73

Scheme 20

Pyrazole carbothioamide

Essam Mohamed Sharshira et al [34] have develop new method of synthesis pyrazole carbothioamide **82** by reaction of chalcone **80** with thiosemicarbazide **81** in presence of acetic acid converted in to compound **82** **(Scheme-21)**



80 82

X = H, -OCH3, -CH3, -NO2

Scheme 21

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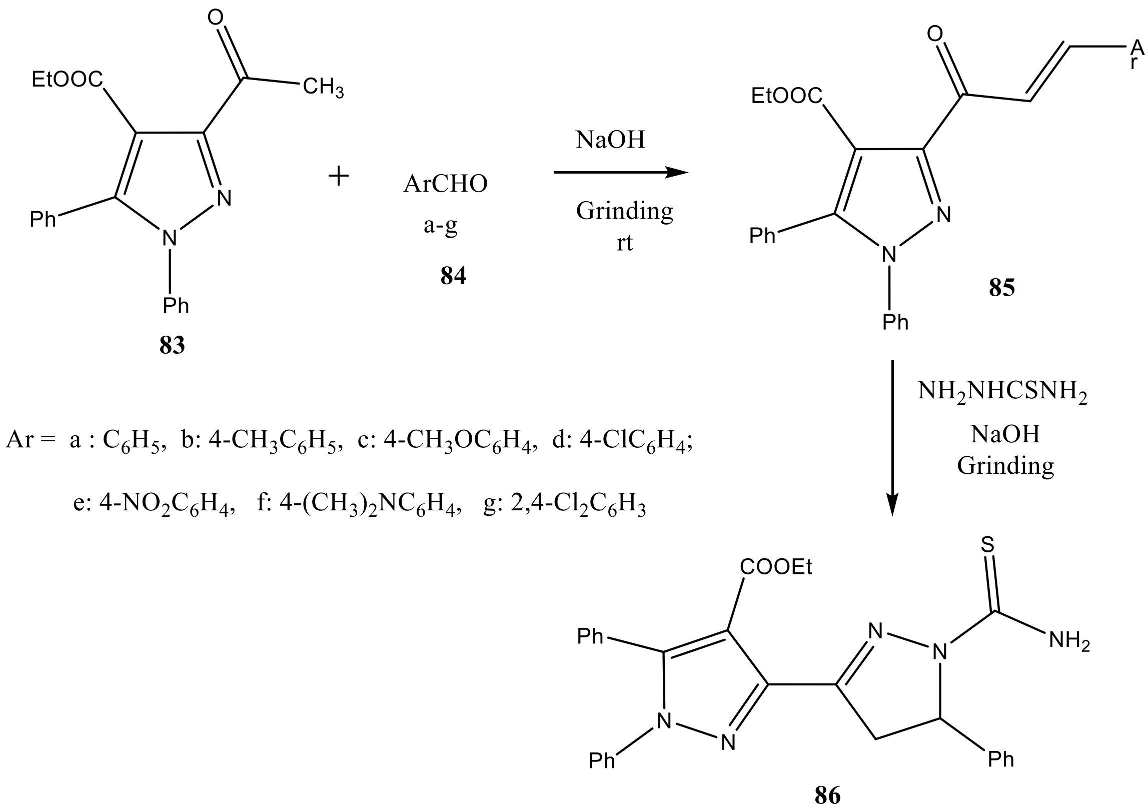
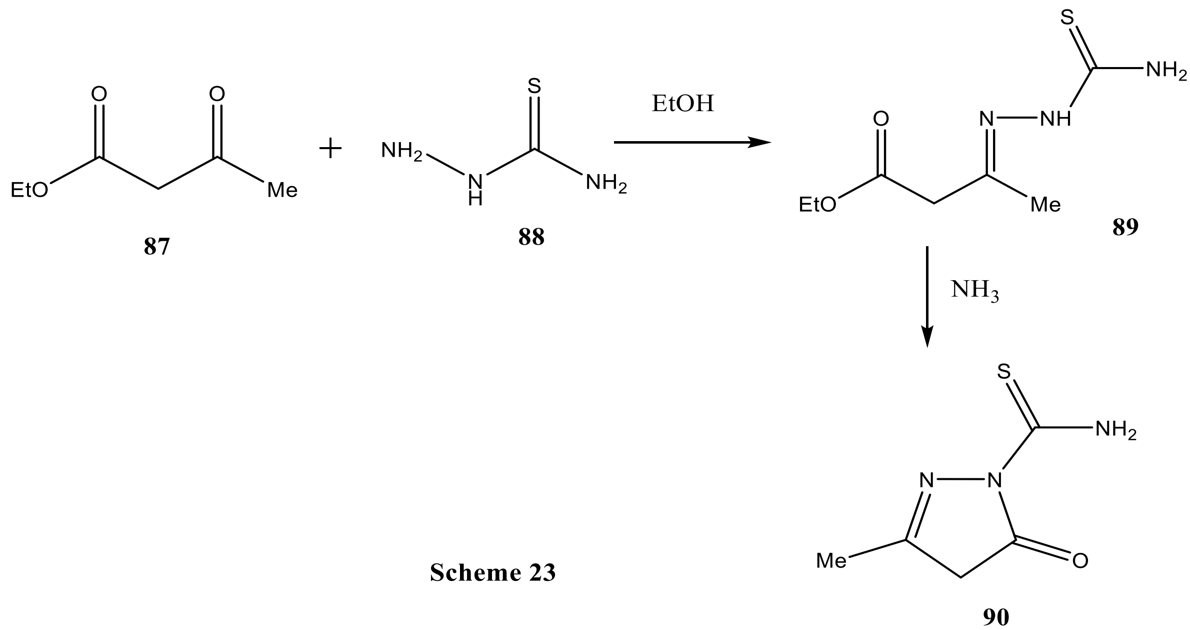
Sobhi Gomha et al [35] have synthesized pyrazole substituted pyrazole carbothioamide from condensation reaction of pyrazole ketone **83** with aldehyde by use of environment friendly method like grinding technique in presence of catalytic amount sodium hydroxide converted in to compound **86**(**Scheme-22)**



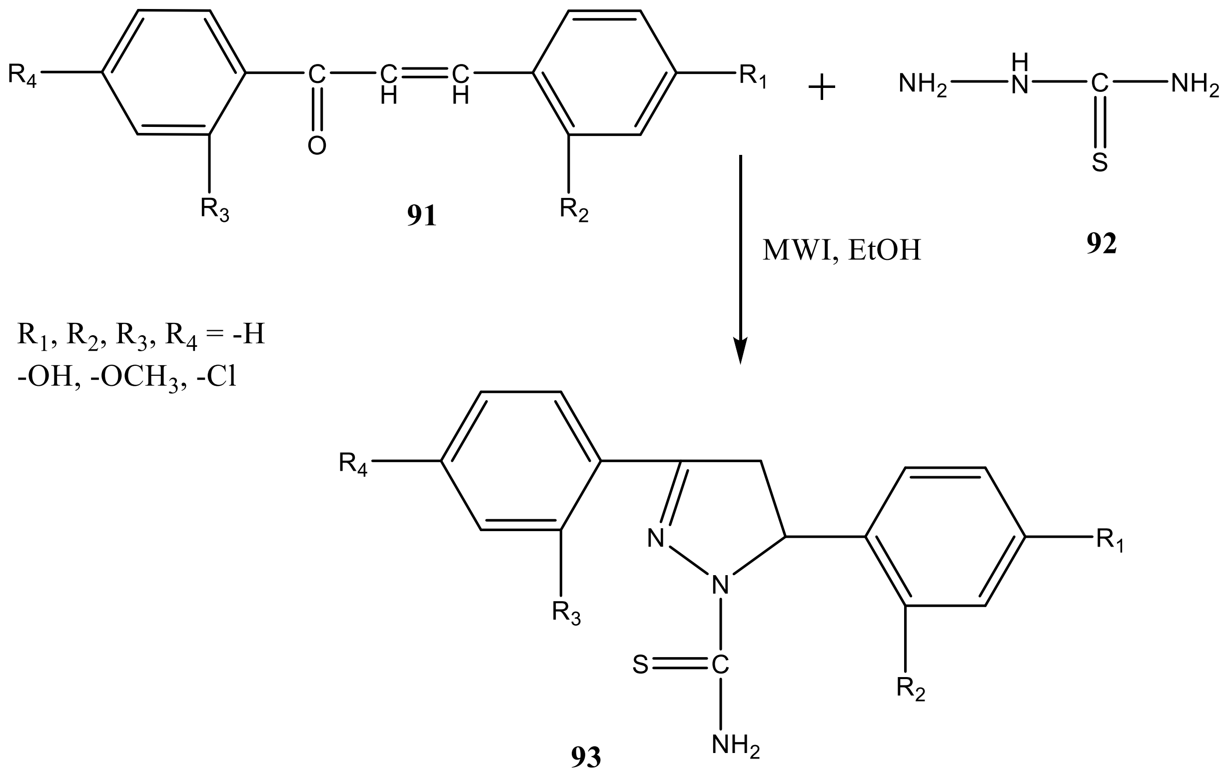
Scheme 22

Ebrahim Abdel-Galil et al [36] have formulated synthesis of pyrazole carbothiomide by reaction of ethyl 3-oxobutanoate **87** with thio semicarbazide **88** afforded to **89** which on further treatment with ammonia afforded to pyrazole carbothiomide **90.** (**Scheme-23)**



Scheme 23

Rashmi Sharma et al [37] have devised method of synthesis of pyrazole carbothiamide from reaction of chalcone **91** with thiosemicarbazide in microwave assisted reaction furnished in to compound **93** **(Scheme-24)**



Scheme 24

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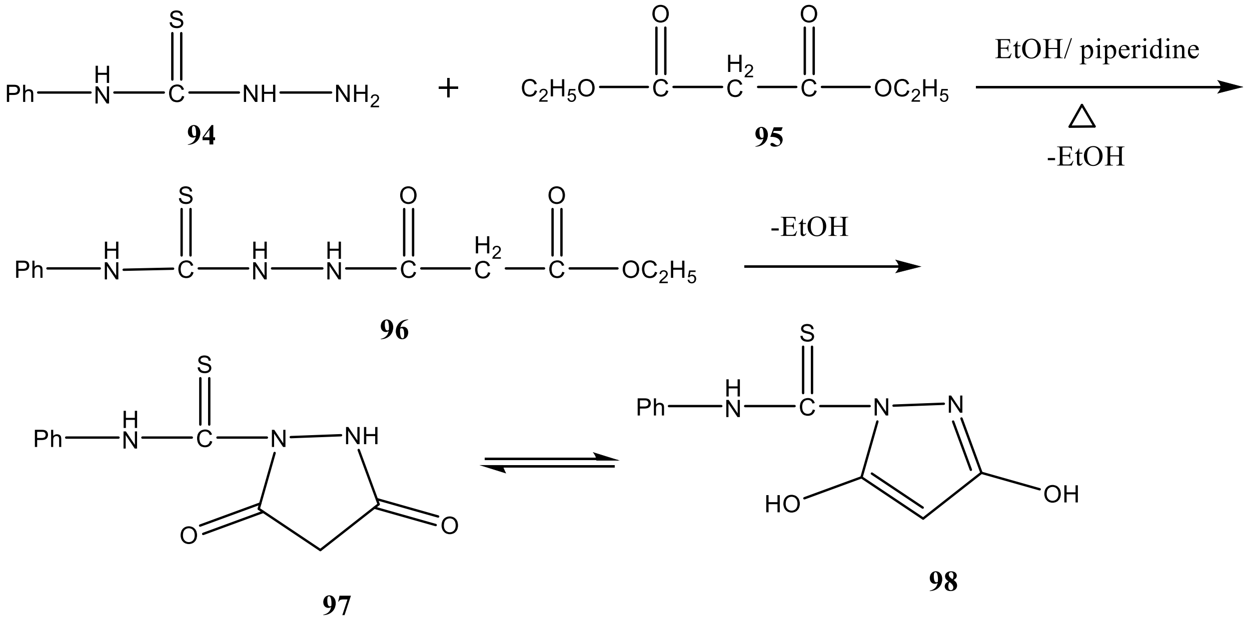
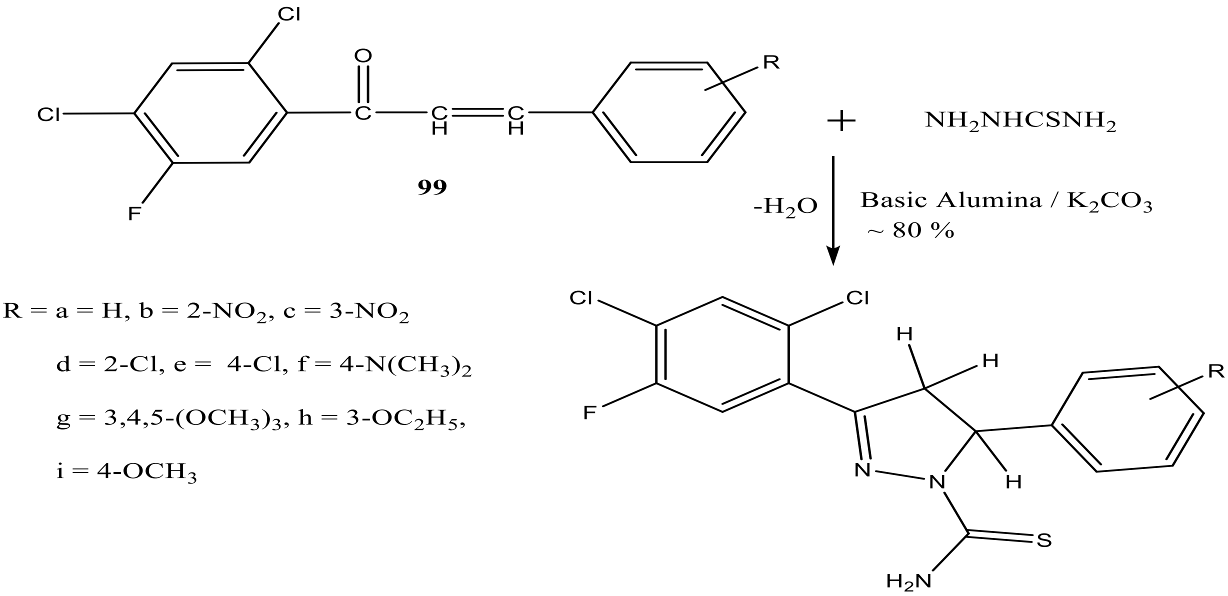
Abdel Sayed N I et al [38] have developed synthesis of pyrazole carbothioamide by condensation reaction of phenyl substituted thiosemicarbazide with diethyl malonate **95** in presence of piperidine afforded to intermediate **96** which is further converted inn to compound **97** which on rearranged to pyrazole carbothioamide compound **98** (**Scheme-25)**



Scheme 25

Patel V M et al [39] have prepared pyrazole carbothioamide by reaction of chalcone **99** with thiosemicarbazide in presence of basic alumina converted in to compound **100** (**Scheme-26)**



Scheme 26

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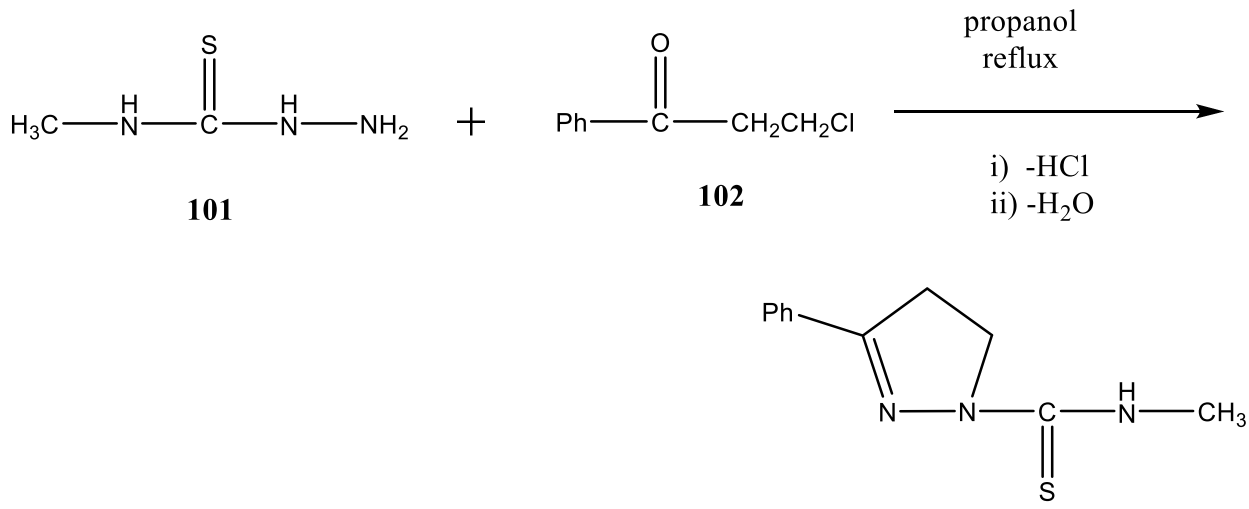
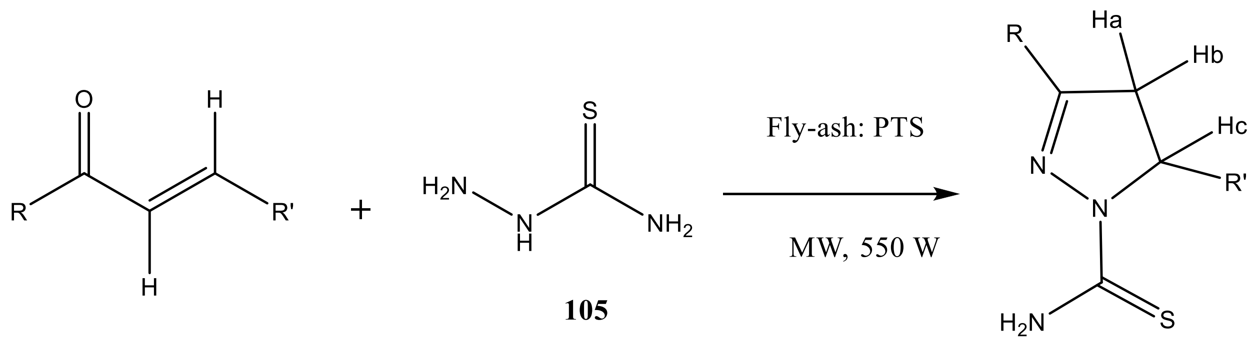
Tomita Y et al [40] have developed formulation of pyrazole thiosemicarbazide from reaction of methyl thiosemicarbazide with ketone **102** converted in to compound **103** (**Scheme-27)**



Scheme 27

Thirunarayanan G et al [41] have developed synthesis of pyrazole carbothiamide from reaction of chalcone **104** and thiosemicarbazide **105** in presence of fly –ash as catalyst in microwave irradiation converted in to compound **106** (**Scheme-28)**



104

105

106

Scheme 28

Nagihan Beyhan et al [42] have formulated synthesis of compound **110** by reaction of ketone and aldehyde in presence of sodium hydroxide afforded to chalcone **109** which on further treatment with thiosemicarbazide **109** in basic medium in presence of ethanol as solvent furnished in to pyrazole carbothiomide **110** (**Scheme-29)**

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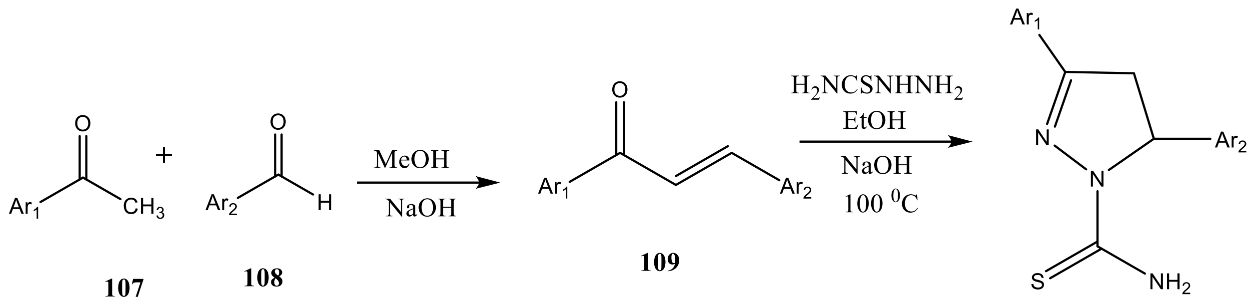

107 108 109 110

Scheme 29

Conclusion

This review reveals that the Pyrazole a versatile molecule many research groups have paid their attention in synthesis of their derivative for discovery of various biological activities. Dicorbonyl compounds, chalcone derivatives, nitriles are important starting material for synthesis of pyrazole derivatives. The ammonia derivatives like hydrazine, phenyl hydrazine, Semicarbazide and thiosemicarbazide have employed for cyclisation and formation of pyrazole moiety with diverse structural features. That enhances applicability of pyrazole nucleus in various field.

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