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Chemical and Biological Potentials of Chalcones: A Review

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Abstract

Plants from the natural world are linked with the treatment of different human ailments. This is due to the presence of different classes of chemical constituents. Flavonoids are one such class of natural constituents responsible for the activity of plants. Chalcones are a class of natural open chain flavonoids that are linked by a three carbon spacer between two aromatic rings. Chalcones and their heterocyclic analogues enjoy a range of biological activities such as antimicrobial, antioxidant, cytotoxic, anticancer, antiprotozoal, antihistaminic, antiulcer and anti-inflammatory activities which makes these compounds as a special attraction for investigation. Additionally the bielectrophilic, ketovinyl chain between the two rings is highly reactive and acts as an important chemical synthon for constructing different five, six and seven membered heterocyclic scaffolds containing different hetero atoms like nitrogen, oxygen and sulfur atoms by abridgment with a variety of binucleophilic reagents. This review highlights on the chemical and biological potentials of chalcones.

Keywords: Flavonoids; Chalcone; Biological Activities; Chemical Synthon; Bielectrophilic and Binucleophilic.

Introduction

Chalcone (Figure 1) is a generic term given to compounds bearing the 1, 3-diphenyl-2-propen-1-one framework and belong to the flavonoid family [1-3]. Chemically they are open-chain flavonoids in which the two aromatic rings are joined by a three carbonα,β-unsaturated carbonyl system. Chalcones are abundantly present in nature starting from ferns to higher plants[4] and a number of them are polyhydroxylated in the aryl rings. In plants, chalcones are converted to the corresponding (2S)-flavanones in a stereospecific reaction catalyzed by the enzyme chalcone isomerase. This close structural and biogenetic relationship between chalcones and flavanones explains why they often co-occur as natural products.

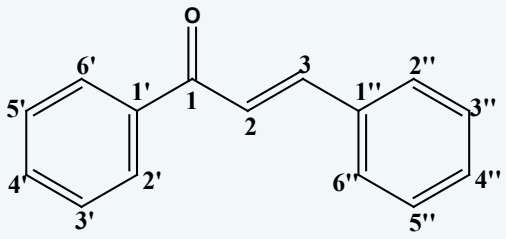


Figure 1. The general structure and numbering of chalcones.

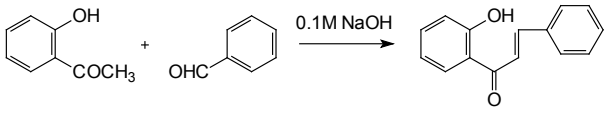
Figure 1. The general structure and numbering of chalcones.

All the chalcones give pink coloration with concentrated sulphuric acid (positive Wilson test)[5] and when a phenolic hydroxyl group is present, they give violet coloration with alcoholic ferric chloride solution. Chalcones on heating with traces of iodine in dimethylsulphoxide (DMSO) for two hours give the corresponding flavones. Chalcones were converted into the corresponding flavonols by their oxidation using hydrogen peroxide in methanolic sodium hydroxide solution and these flavonols showed a characteristic greenish yellow fluorescence in ethanolic solution as well as with concentrated sulphuric acid.

General Methods of Synthesis Of Chalcones

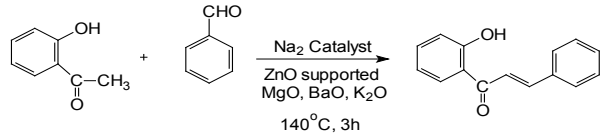
Chalcones can be obtained by the acid or base catalyzed aldol condensation of acetophenones with aromatic aldehydes[6-8].

2'-hydroxyacetophenone react with benzaldehyde in the presence of 0.1M NaOH to give the chalcone[9] (Scheme 1).

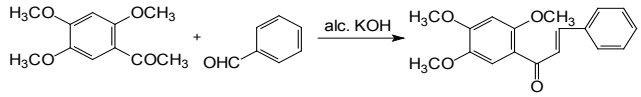


Liquid phase Claisen–Schmidt condensation between

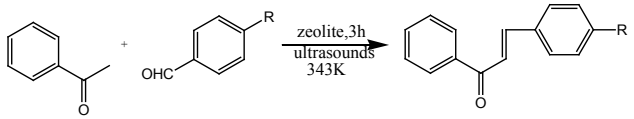
2′-hydroxyacetophenone and benzaldehyde was carried out over a zinc oxide supported metal oxide catalyst under solvent free conditions to form 2′-hydroxychalcone[10] (Scheme 2).



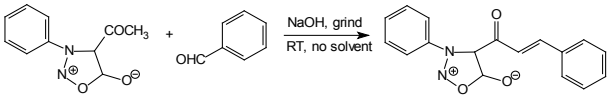
2′,4′,5′-trimethoxyacetophenone, when condensed with equimolar proportions of aromatic aldehydes in the presence of 30 % alcoholic alkali at room temperature yield chalcones[11] (Scheme 3).



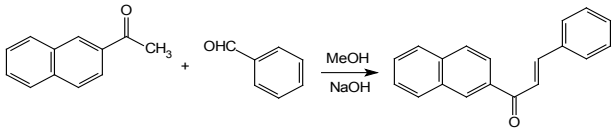
Claisen-Schmidt condensation between benzaldehyde and acetophenone by sonochemical and thermally activated reactions over zeolite as catalyst under solvent free conditions give chalcone[12] (Scheme 4).



4-acetyl-3-aryl-syndones when subjected to grinding with various aryl aldehydes in the presence of a base catalyst under solvent free conditions yield sydnone chalcones[13] (Scheme 5).



Condensation of 2-naphthylmethyl ketones with substituted aryl aldehydes in the presence of NaOH under methanol as solvent gave the corresponding chalcones[14] (Scheme 6).



Molecular Spectral Studies of Chalcones

Ultra Violet Spectroscopy

The major absorption band in chalcones (Band I) usually occurs in the range 340-390 nm, although chalcones lacking B-ring oxygenation may have their Band I absorption at considerably shorter wavelengths and Band II is usually a minor peak in the 220-270 nm region[15]. In the spectra of chalcones containing a free 4′′-hydroxyl group, the addition of NaOMe causes a 60-100 nm bathochromic shift of Band I with an increase in peak intensity. Chalcones without a 4′′-hydroxyl group but with either a free 2′′ or 4′- hydroxyl group also give, in the presence of NaOMe, a 60-100 nm bathochromic shift of Band I but without an increase in peak intensity[16, 17]. UV spectroscopy proved useful to distinguish between substituted chalcones and flavanones, which is not possible by EI mass spectrometry due to thermal isomerization of 2′-hydroxychalcones in the ion source[18, 19].

Infra Red Spectroscopy

The α, β-unsaturated carbonyl group, characteristic of a chalcone usually appear as a prominent band in between 1625-1650 cm^{-1} in its IR spectrum[20,21]. The region at which other absorption bands appear depends on the type of aromatic / heteroaromatic rings as well as the substituents present on these rings.

Nmr Spectroscopy

The H-α and H-β protons of chalcones occur as two doublets (J= 17 Hz) in the ranges 6.7 – 7.4 ppm (H- α) and 7.3 -7.7 ppm (H- β) in the ^{1}H NMR spectra[22]. The other aromatic protons usually appear in between δ 6.9-8.0, depending on the type of aromatic/ heteroaromatic ring and also based on the electronic effects of the substituents present on these rings. The large J value (17 Hz) clearly reveals the *trans* geometry for the chalcones. The carbonyl carbon of the chalcones usually appears between δ 188.6 and 194.6 in its ^{13}C NMR spectrum[23]. The α and β- carbon atoms with respect to the carbonyl group give rise to characteristic signals in between δ 116.1-128.1 and δ 136.9-145.4 respectively, which can also be readily identified by their characteristic appearance as a six line multiplet in the half resonance decoupled spectrum[24]. The presence of 2′-hydroxy group shifts the carbonyl carbon shift downfield by 3 ppm relative to corresponding acetoxy and methoxy compounds, presumably owing to hydrogen bonding. The β-hydroxy chalcones are a relatively small group of chalcones that occur naturally sometimes as the enol- tautomers of dibenzoylmethane derivatives. The extent of keto-enol tautomerism is largely solvent dependent, and nuclear magnetic resonance spectroscopy (NMR) provides one of the best methods to determine the ratio of the tautomers present. In the ^{1}H NMR spectra recorded in $CDCl_3$, the exchangeable proton of the β-OH of the enol tautomer appears as a 1H singlet at δ 16.0, whereas the α-CH_2 protons of the keto tautomer appear as a 2H singlet at δ 4.50. Another diagnostic resonance is the 1H methine singlet of the enol tautomer (α-CH), which is found at δ 6.5, with its corresponding C- α resonance at δ 90 to 92 in the ^{13}C NMR spectra[25, 26].

Mass Spectrometry

Cleavage of the heterocyclic C-ring via a retro Diels-Alder (RDA) mechanism represents an important fragmentation pathway in chalcones. RDA fission leads to two characteristic fragments, which provides useful information as to the number of hydroxyl, methoxyl and other substituents on each ring[27] (Scheme **7**). The same information was also obtained by a HPLC-tandam mass spectrometer system equipped with a heated

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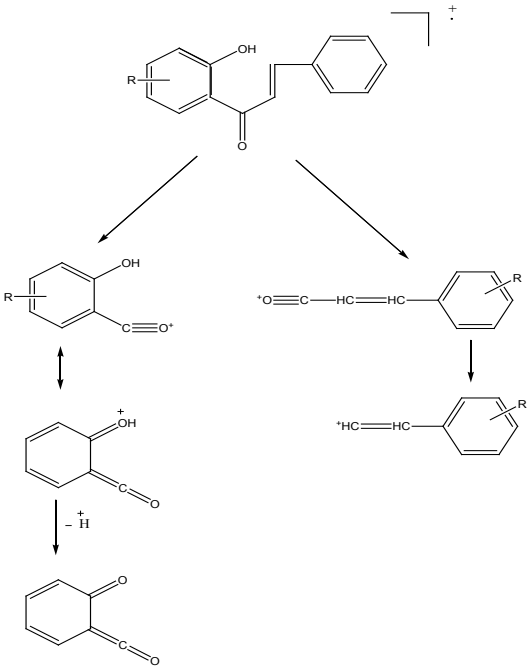
nebulizer- atmospheric pressure chemical ionization (APCI) interface[28].

Scheme 7. A typical fragmentation pattern of a 2'-Hydroxy chalcone.



Therapeutic Potential Of Chalcones

Chalcone is unique template that is associated with several biological activities (Figure 1.1) and is also well known for synthesizing various heterocyclic compounds[29]. They are secondary metabolites of terrestrial plants, precursors for the biosynthesis of flavonoids. The introduction of a halogen into the benzenoid part of these α, β-unsaturated ketones enhances their biological activity[30].

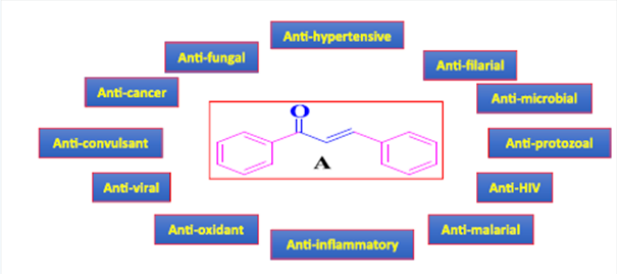


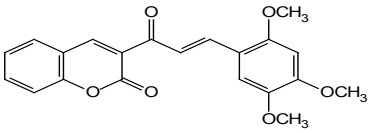
Figure 1.1. Different biological activities of chalcone analogues

The compounds with chalcone as backbone have been reported to possess varied biological and pharmacological activities[31], including antimicrobial, anti-inflammatory, analgesic, cytotoxic, antitumor, antimalarial, antitubercular, antiviral, anti-HIV, antiulcerative, antileishmanial, antioxidant, antiprotozoal, antihistaminic, antifedent, immunomodulatory, anticonvulsant, antihyperglycemic, antihyperlipidemic and antiplatelet activities. Thus chalcones continue to attract considerable scientific attention because of their association with a variety of biological activities. Given below is a brief account of various modifications reported on chalcones, which resulted in a variety of biological and pharmacological activities.

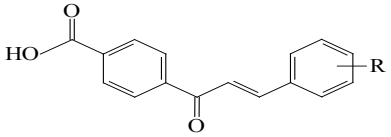
Antimicrobial activity

The antimicrobial activity of chalcones is being increasingly documented. Many research groups either isolated or synthesized chalcones that possess antimicrobial activity. The presence of a reactive α, β-unsaturated keto function in chalcones was found to undergo conjugate addition with a nucleophilic group like a thiol group in an essential protein, thus partly contributing for their antimicrobial activity, which may be altered depending on the type and position of the substituents on the aromatic rings.

Prasad *et al.*[32] synthesized 3-[1-oxo-3-(2, 4, 5-trimethoxyphenyl)-2-propenyl]-2H-1-benzopyran-2-ones **(2)** that showed significant antimicrobial activity against *B.subtilis, B.pumilis* and *E.coli* when tested at a concentration of 1000 µg/ml. The study revealed the importance of electron releasing groups such as hydroxyl and methoxyl groups in enhancing the activity. Chalcones with halogen substituents like bromine and chlorine contributed favorably to the antifungal activity.



Nielsen *et al.*[33] described the bioisosteric replacement of the essential 4'-hydroxy group in the hydroxychalcones with bioisosters of varied degrees of acidity which resulted in both more potent and more soluble compounds. Exchanging the hydroxyl group, particularly with a carboxy group resulted in a potent compound with a high aqueous solubility. Further optimization and SAR analysis resulted in soluble and potent carboxychalcones having dibromo or trifluoromethyl substitution on B-ring **(3).** The MIC values for these compounds were found to be 2 µM and 40 µM respectively when tested against the Gram-positive bacterium *Staphylococcus aureus.* A dibromo or trifluromethyl substitution on B-ring was found to enhance the lipophilic character, while the carboxy group on A-ring contributed to the required aqueous solubility.



Karthikeyan *et al.*[34] synthesized 3-aryl-1-(2,4-dichloro-5-fluorophenyl)-2-propen-1-ones **(4)** showing antimicrobial activity, again consistent with the observations that the halogens possess favorable lipophilic character required for antimicrobial activity.

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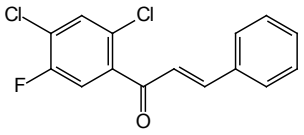
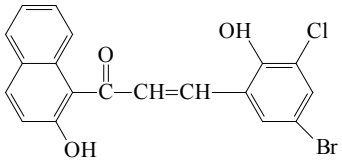
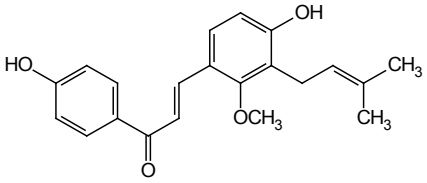


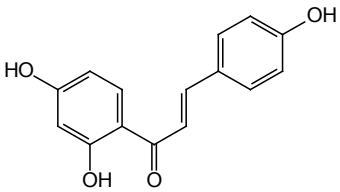
Prasad *et al.*[35] synthesized a chalcone **(5)** having a naphthalene moiety on one side and an aryl moiety having substituents on the other side, which showed significant antifungal activity against *A.niger* and *R.oryzae*. This compound can also be considered to provide optimal hydrophilic and hydrophobic properties as evidenced by hydroxyl groups and the halogens.



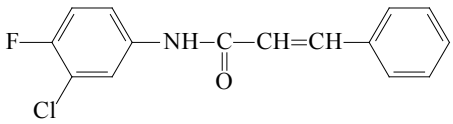
Tsukiyama *et al.*[36] isolated a retrochalcone, licochalcone-C **(6)** from *Glycyrrhiza infanta* which showed potent antibacterial activity.



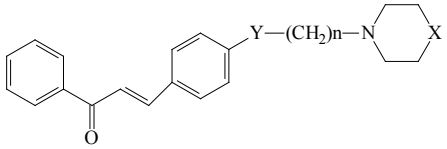
Machodo *et al.*[37] isolated isoliquiritigenine **(7)** which showed antibacterial activity.



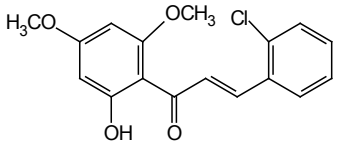
Rao *et al.*[38] synthesized chalcones having chlorine and fluorine substitution **(8)**, which showed antimicrobial activity.



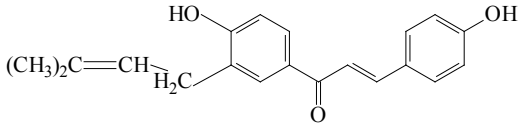
Nowakowska *et al.*[39] synthesized a series of substituted chalcones **(9)** and tested for their antibacterial and antifungal activities. The physico-chemical properties of these novel chalcones which contributed favourably to the observed activities were also determined.



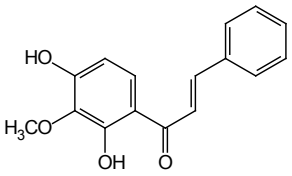
Boeck *et al.*[40] synthesized novel xanthoxylin-derived chalcones **(10)** showing antifungal activity.



Sohly *et al.*[41] isolated prenylated chalcones **(11)** from the leaves of *Malclura tinctoria* possessing antifungal activity.

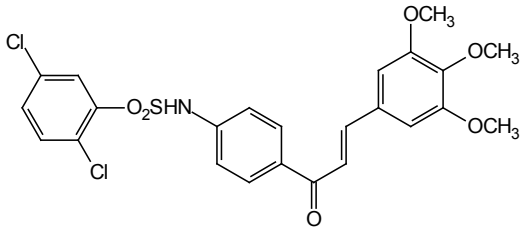


Stevaz *et al.*[42] isolated a 2', 4'-dihydroxy-3'-methoxychalcone **(12)** from the methanolic extract of *Zuccagnia punctata* which exhibited antifungal activity.

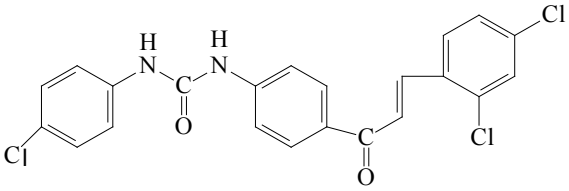


Antimalarial activity

Dominguez *et al.*[43] synthesized chalcones **(13)** with sulfonamide moiety possessing antimalarial activity.

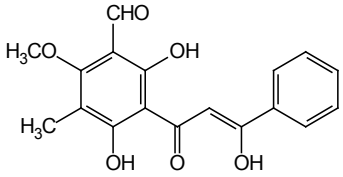


Dominguez *et al.*[44] synthesized phenylurenyl chalcones **(14)** that exhibited antimalarial activity.



Anti-HIV activity

Wu *et al.*[45] isolated a chalcone **(15)** that exhibited the anti-HIV activity with a good therapeutic index.



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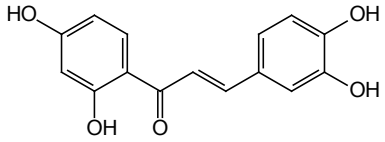
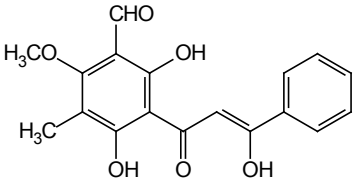
Xu *et al.*[46] isolated butein **(16)** possessing anti-HIV activity.

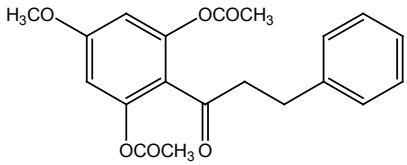


Nakagawa *et al.*[47] isolated a unique potent chalcone **(17)** from genus ***Desmos*** showing anti-HIV activity.

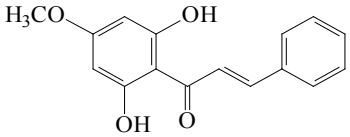


Antileishmanial activity

Hermoso *et al.*[48] synthesized a dihydrochalcone **(18)** having antileishmanial activity.

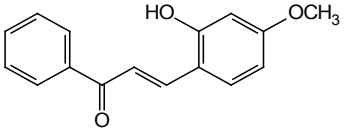


Santos *et al.*[49] synthesized 2',6'-dihydroxy-4'-methoxychalcone **(19)** that showed significant antileishmanial acitivity.

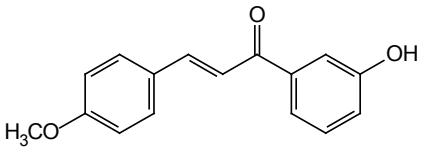


Antitubercular activity

Kumar *et al.*[50] synthesized a chalcone **(20)** that exhibited antitubercular activity.

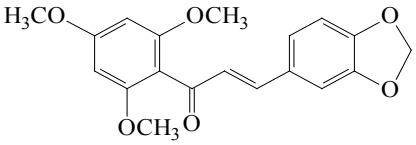


Kumar *et al.*[51] synthesized a chalcones (**21)** possessing antimycobacterial activity.

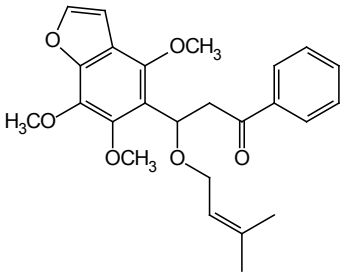


Antitumor activity

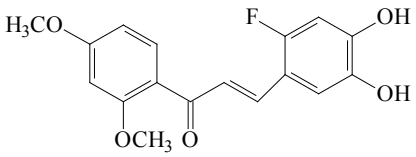
Moderate cytotoxicity was reported for a methylenedioxychalcone **(22)** isolated from the stem bark of *Millettia leucantha*[52].



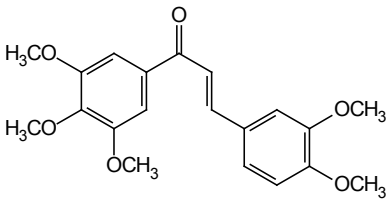
Francesco *et al.*[53] synthesized furanochalcones **(23)** possessing anticancer activity.



Miyataka *et al.*[54] have successfully designed and synthesized some new fluorinated 3,4-dihydroxychalcones and evaluated their biological activities with respect to anti peroxidation activity. All the fluorinated chalcones tested showed 5-lipoxygenase inhibition on rat basophilic leukemia-1. However, the 6-Fluoro-3,4-dihyroxy-2',4'-dimethoxychalcone **(24)** was the most effective compound in the *in vitro* assay using a human cancer cell line panel consisting of 39 systems.



Lawrence *et al.*[55] synthesized a methoxylated chalcone **(25)** possessing good cytotoxic activity.



Sato *et al.*[56] synthesized a fluorinated chalcone **(26)** exhibiting anticancer activity.

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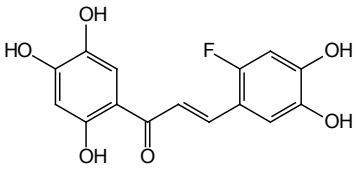
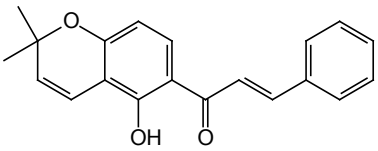
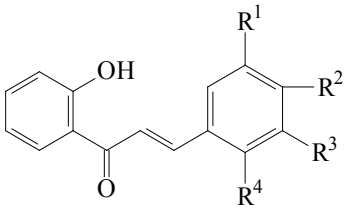


Cunha *et al.*[57] isolated the chalcone lonchocarpin **(27)** from the roots of *Lonchocarpus sericeus* which showed cytotoxic activity.



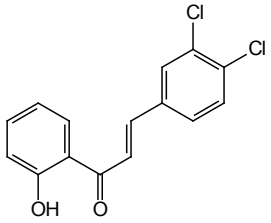
Rodrigo *et al.*[58] studied the relationship between the structural characteristics of synthetic chalcones and their antitumor activity. Treatment of Hep G2 cells for 24 h with synthetic 2′-hydroxychalcones **(28)** resulted apoptosis induction and dose-dependent inhibition of cell proliferation. The calculated reactivity indexes and the adiabatic electron affinities suggest a structure-activity relationship between the chalcone structure and the apoptosis in HepG2 cells.



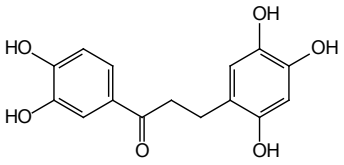
	R1	R2	R3	R4
1.	H	H	H	H
2.	OCH3	OCH3	H	H
3.	OCH3	H	H	OCH3
4.	OCH3	H	OCH3	H

Anti-inflammatory and Analgesic activities

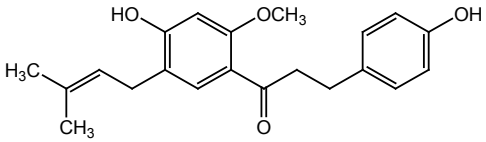
Shen *et al.*[59] synthesized a 2′-hydroxy-3,4-dichlorochalcone **(29)** possessing anti-inflammatory and cancer chemopreventive activity.



Ito *et al.*[60] isolated a reduced chalcone **(30)** having cyclooxygenase-2 inhibitory activity.

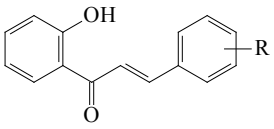


Zhao *et al.*[61] isolated dihydroxanthohumol **(31)** from fruits of *Mallotus philippinensis* which showed anti-inflammatory activity.

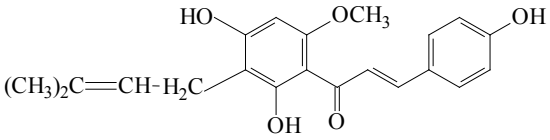


Antioxidant activity

Kostova *et al.*[62] synthesized 2'-hydroxychalcones **(32)** which showed antioxidant activity.

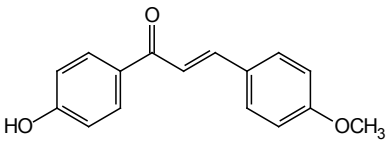


Miranda *et al.*[63] synthesized a prenylated chalcone **(33)** exhibiting antioxidant activity.

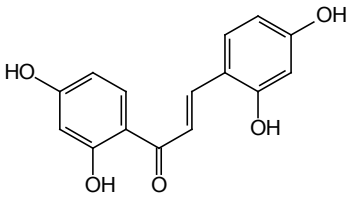


Miscellaneous activities

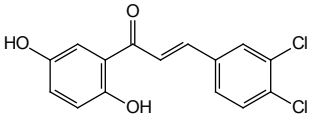
Satyanarayana *et al.*[64] synthesized a 4′-hydroxy-4-methoxychalcone **(34)** exhibiting antihyperglycemic activity.



Soliman *et al.*[65] synthesized a tetrahydroxychalcone **(35)** that showed potent tyrosine- kinase inhibitory activity.



Ko *et al.*[66] synthesized a chalcone **(36)** showing the inhibition of nitric oxide production in lipopolysaccharide-activated macrophages.



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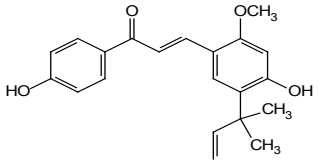
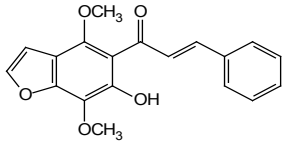
Barford *et al.*[67] isolated an oxygenated chalcone, lichochalcone A **(37)** from the roots of the Chinese liquorice, *Glycyrrhizae uralensis*, which showed immunosuppressive property.



Baell *et al.*[68] synthesized a chalcone **(38)** having potassium channel modulatory activity.



Chalcones As Synthon In Chemical Synthesis

Chalcones are resourceful precursor for the synthesis of heterocyclic compounds (Figure 2). Chalcones undergo cyclization reactions with different reagents to form diverse classes of heterocyclic compounds ranging from five membered to seven membered rings containing nitrogen, oxygen and sulfur heteroatoms. In the cyclization reactions the highly reactive bielectrophilic ketovinyl chain condenses with a variety of binucleophilic reagents to generate an assortment of heterocyclic systems such derivatives pyrazolines, phenylpyrazoline and isoxazole (5-membered heterocyclics),[69] derivatives aminopyrimidines and cyanopyridines (6-membered heterocyclics)[70] and derivatives of 1,5-benzodiazepines, 1,5-benzoxazepines, and 1,5-bezothiazepines (7-membered heterocyclics).[71]

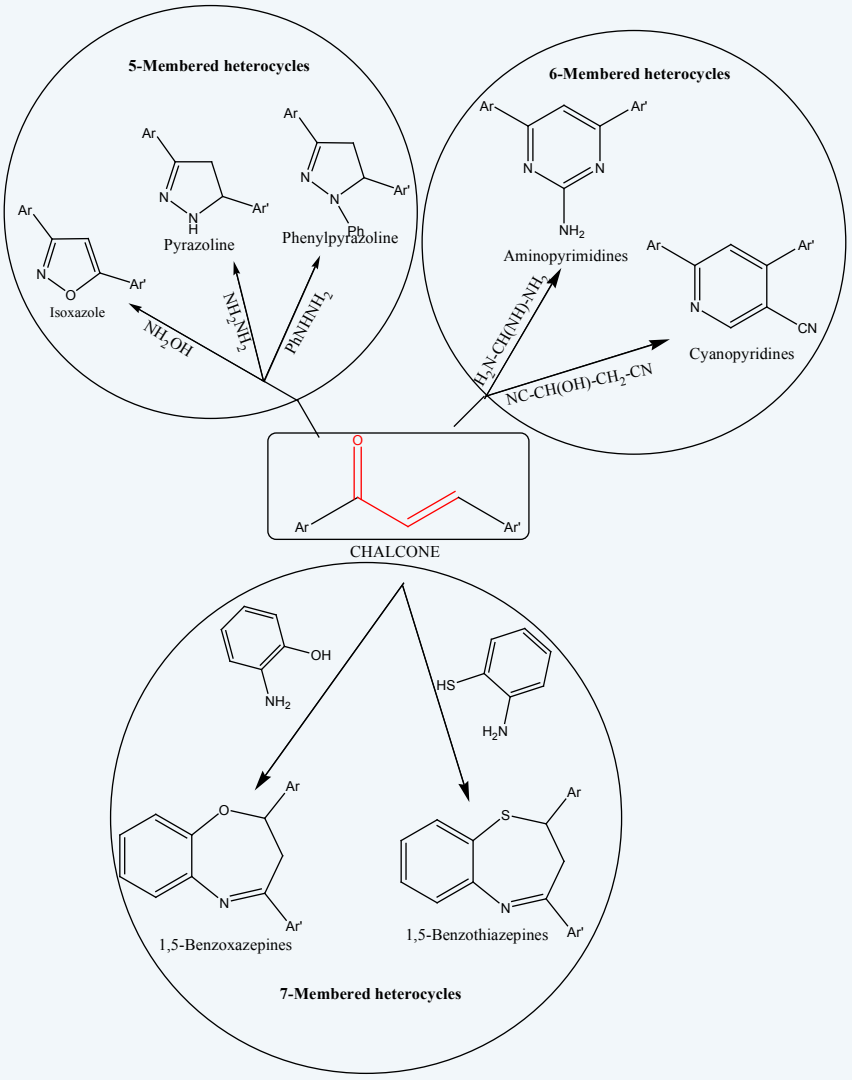


Figure 2. Synthetic potential of chalcone derivatives for the synthesis of heterocyclic analogues

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Conflicts of interest

The authors have no conflicts of interest.

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