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UPDATED REVIEW ON COUMARIN DERIVATIVES AND THEIR PHARMACOLOGICAL ACTIVITIES

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ABSTRACT

A significant class of naturally occurring secondary metabolites, coumarins are found in many plant components, such as roots, leaves, seeds, and fruits. Coumarins are categorised into various classes, including simple coumarins, furanocoumarins, pyranocoumarins, phenyl coumarins, and bicoumarins. They are structurally defined by a benzopyran-2-one nucleus and display a variety of oxygenation patterns. Due to their enormous biological potential, a great deal of study has been done on their synthesis using both traditional and contemporary methods, such as Pechmann, Perkin, Knoevenagel, and Wittig reactions, as well as green and catalytic techniques. Numerous pharmacological properties, such as antibacterial, anticoagulant, antioxidant, anti-inflammatory, and anti-hyperpigmentation effects, are exhibited by coumarin derivatives. These substances work by inhibiting enzymes, scavenging free radicals, and modifying cellular signalling pathways, among other processes. In addition to its uses in the food, cosmetics, and pharmaceutical industries, its wide therapeutic potential emphasises its significance in medicinal chemistry and drug development. This article offers a thorough examination of coumarins, emphasising their varied structural categories, including furanocoumarins and pyranocoumarins, as well as contemporary synthetic techniques such as Pechmann, Perkin, and Knoevenagel reactions. Moreover, it examines their considerable pharmacological potential, elucidating their functions as antibacterial, anticoagulant, antioxidant, and anti-inflammatory agents in medicinal chemistry and drug development.

KEYWORDS: Coumarin, Benzopyran derivatives, Synthetic methodologies, Pharmacological activities, Antibacterial and antioxidant properties.

INTRODUCTION

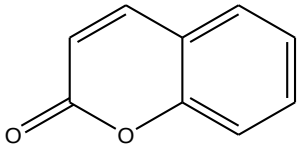
Coumarin constitutes a significant category of secondary metabolites that are extensively spread across the plant kingdom. They are characterised by a variety of oxygenation patterns on the benzopyran nucleus and exhibit a range of

biochemical and pharmacological activities.[1] Coumarins are prevalent in nature and are present in various plant roots, flowers, leaves, peels, seeds, and fruits as secondary metabolites.[2]

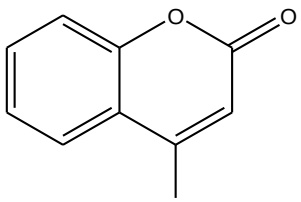
Chemistry of coumarin and its Derivatives

Structure

Coumarin and its derivatives serve as primary oral anticoagulants. Coumarin is insoluble in water; nevertheless, the 4-hydroxy substitution imparts weakly acidic characteristics to the molecule, rendering it water-soluble in mildly alkaline conditions.



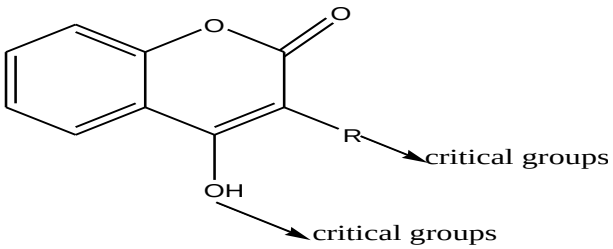
COUMARIN



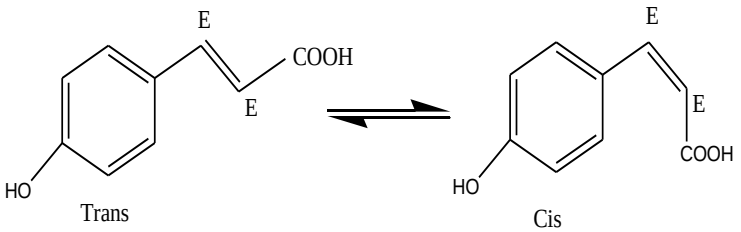
4-hydroxycoumarin

Structure-activity relation

Coumarin and 4-hydroxycoumarin lack anticoagulant properties. Link, who was the first to isolate and characterise bis-hydroxycoumarin (dicoumarol) from sweet clover, determined that the essential criteria for anticoagulant activity include a 4-hydroxy group, a 3-substituent, and a bis molecule.



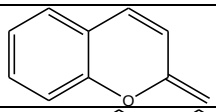
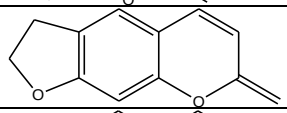
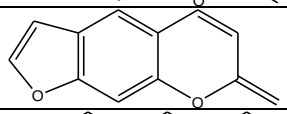
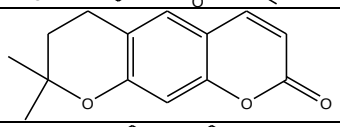
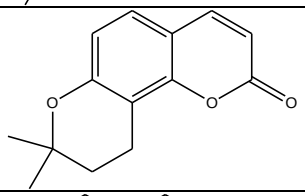
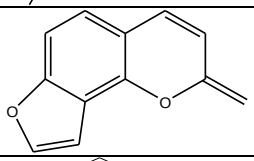
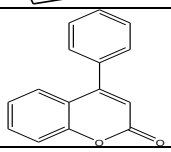
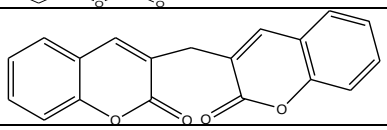
The coumarin structure originates from cinnamic acid through ortho-hydroxylation, trans-cis isomerisation of the side chain double bond, and lactonisation. The trans form is stable and cannot cyclize; thus, isomerisation must occur, implicating the enzyme isomerase.



Classification

Chemically, coumarins (2H-1-benzopyran-2-one) are classified within the lactone subgroup.[3] Coumarin is alternatively referred to as 1,2-benzopyrone or o-hydroxycinnamic acid-8-lactone.

Natural coumarins can be categorized into six fundamental groups: simple coumarins, furanocoumarins, pyranocoumarins (linear and angular types), dihydrofurano coumarins, phenyl coumarins, and bicoumarins (Table-1)[4]

Type of coumerin	Structure	Example
Simple coumarins		Esculetin, Coumarin
Dihydrofurano coumarins		Marmesin, rutaretin
furano coumarins		Marmalde, marmelosin
Pyrano coumerin Linear type		Xanthyletin
Pyrano coumerin Angular type		praeruptorin A,praeruptorin B, hyuganin G, hyuganin H,
Furano coumarins Angular type		Calanolide A, B, and F
Phenyl coumarins		disparpropylinol B
Bicoumarins		Dicoumarol

Since many natural items include this heterocyclic nucleus, chemical and medicinal chemists have long been interested in the synthesis of coumarin and its derivatives. They are frequently added to food, cosmetics, perfumes, and medications.

Synthetic Methodologies

2.1 Pechmann condensation

2.1.1 In the presence of acid catalysts, phenols and ketoesters condensed to create coumarins. The Pechmann-Duisberg reaction occurs when acetic acid derivatives are utilised. Additionally, as Scheme 1 illustrates, it requires the esterification of phenol and beta-keto ester in the presence of a protonic acid catalyst.[5]

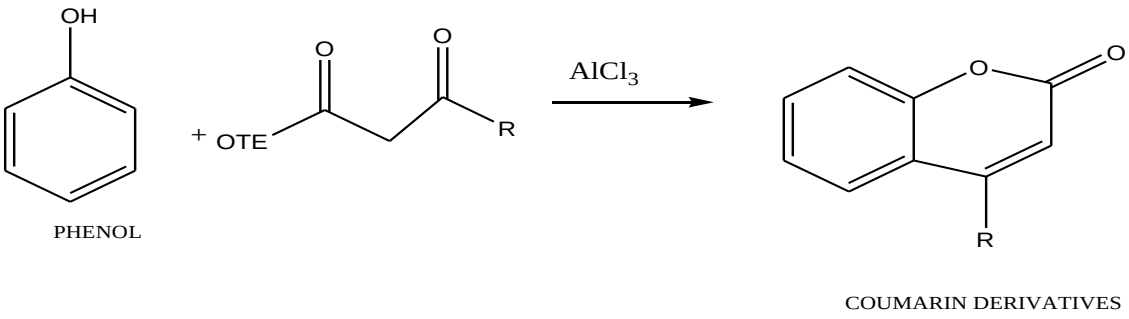


Figure 2: Pechmann-Duisberg condensation.

2.1.2 Neutral ionic liquids with a catalytic amount of acid have been employed for coumarin synthesis via Pechmann condensation of phenols and ethylacetoacetate under ambient conditions. The reaction was also successfully carried out at high temperature in i-butyl-3-methylimidazolium hexafluorophosphate ionic liquid, without the use of any acid catalyst.[6]

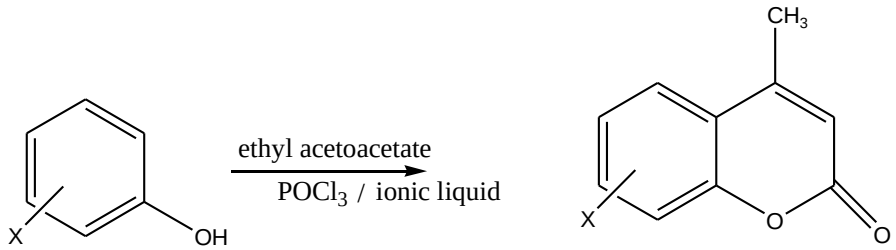


Figure 3: Pechmann condensation of phenols and ethyl acetoacetate in neutral ionic liquids catalysed by POCl3.

2.1.3 For the production of coumarin and its derivatives, a novel heterogeneous catalytic approach was devised using the Ti(IV)-doped ZnO matrix forming catalyst Zn0.925Ti0.075O, which has a high Lewis acidity and surface area. During the manufacture of coumarins, the catalyst exhibits high activity towards a wide range of substituted phenols with β-ketoesters, including ethyl acetoacetate, ethyl butyryl acetate, ethyl benzoyl acetate, and so on, in good yields over short reaction periods.[7]

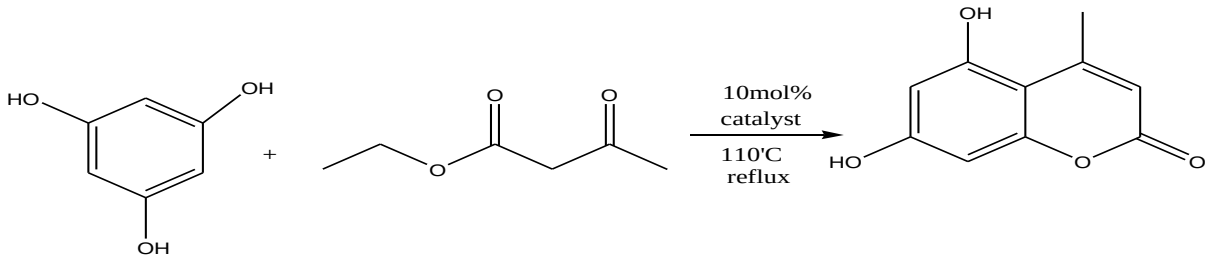


Figure 4: Heterogeneously Catalysed Pechmann Condensation Employing the Tailored.

Zn0.925Ti0.075O NPs: Synthesis of Coumarin

2.1.4 Mesoporous zirconium phosphate (m-ZrP) has strong catalytic activity for the condensation of phenols and ethyl acetoacetate in both traditional heating and microwave-assisted methods. It is employed as a solid acid catalyst for the manufacture of coumarins via the Pechmann condensation reaction.[8]

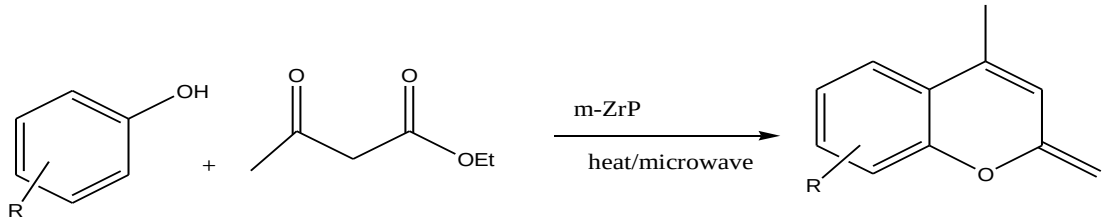


Figure 5: Mesoporous zirconium phosphate catalysed synthesis of coumarin derivatives through Pechmann condensation reaction.

2.1.5 A quick and effective solvent-free one-pot synthesis of coumarin derivatives employing FeF3 as a catalyst in Pechmann condensation reactions of phenols with ethyl acetoacetate under microwave irradiation. The compounds are produced in good yields using this one-pot synthesis on a solid inorganic substrate.[9]

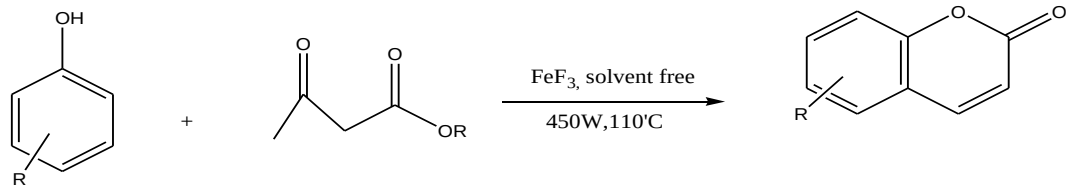


Figure 6: FeF3 catalysed the Pechmann reaction.

2.1.6 A green protocol has been developed for the synthesis of simple coumarins and linear pyrano[2,3-f] and [3,2-f]indoles by the reaction of phenol derivatives with β-ketoesters under ball milling at ambient temperature in the presence of methanesulfonic acid as a mild acid catalyst. The significant advantages of this procedure are high yields, scalability, no use of hazardous acids or solvents, shorter reaction time, ambient temperature, and low cost.[10]

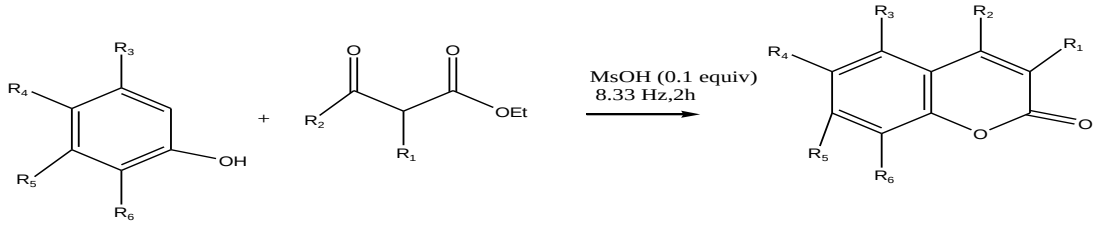


Figure 7: Mechanochemical synthesis of coumarins under solvent-free conditions.

2.1.7 Zirconyl chloride octahydrate supported by silica gel was discovered to be an effective and recyclable catalyst for the high turnover numbers and rates of synthesis of several biologically significant compounds. By directly reacting β-keto esters and phenol derivatives with a catalytic quantity of ZrOCl2.8H2O/SiO2 as Lewis acid at room temperature without the use of solvents, several substituted coumarins can be produced in high yield and purity.[11]

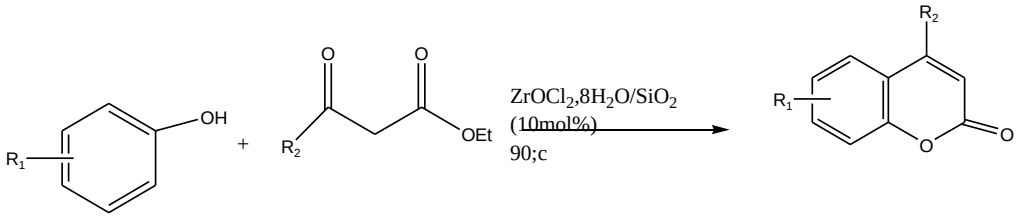


Figure 8: SynthesisofcoumarinderivativesbyPechmanncondensationusingβ-keto esters and phenol derivatives inthepresenceofZrOCl2.8H2O/SiO2.

2.1.8 Nitration of 7-hydroxy-4-methyl has produced a nitro coumarin derivative. Pechmann condensation of resorcinol with a β-ketoester was used to create coumarin.

By monitoring the reaction between resorcinol and ethyl, Amberlyst-15 serves as an effective and environmentally friendly catalyst. acetoacetate (1:1 mol ratio) at 110 °C in solvent-free circumstances with 0.2 g of Amberlyst-15. These nitro coumarin derivatives are significant and have biological activity in the industrial domains.

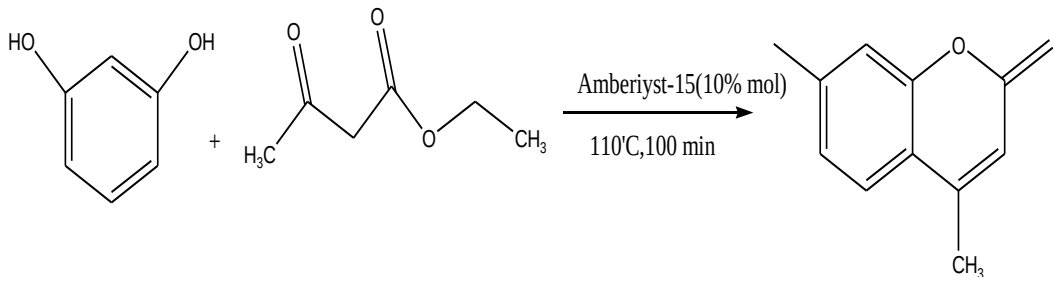


Figure 9: Pechmann condensation reaction of phenols with β-keto esters to substituted coumarins.

2.2. Witting Reaction

2.2.1 o-Hydroxybenzaldehydes on reaction with chloroacetyl chloride in the presence of pyridine, followed by the addition of triphenyl phosphine and base, gave coumarins

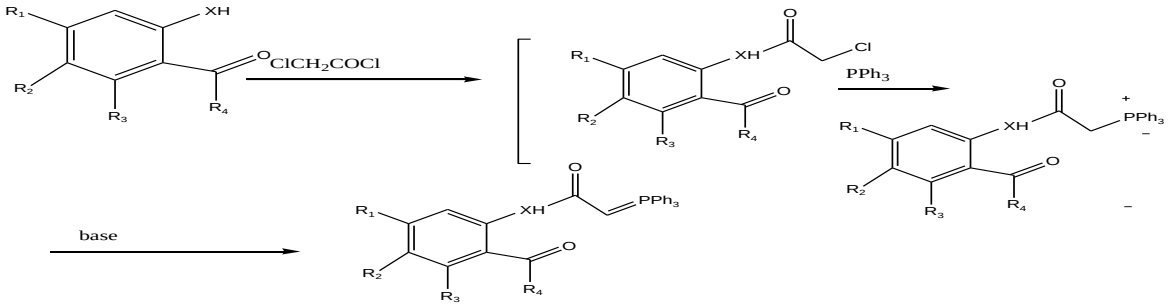


Figure 10: Intramolecular Wittig reactions.

2.2.2 Treatment of salicylaldehyde (2a) with ethyl diphenylphosphonoacetate (5a) at 0°C in the presence of NaI gave an unexpected intramolecular phosphonate derivative (a) in 33% yield along with the E-olefinated product (b) in 37% yield.[12]

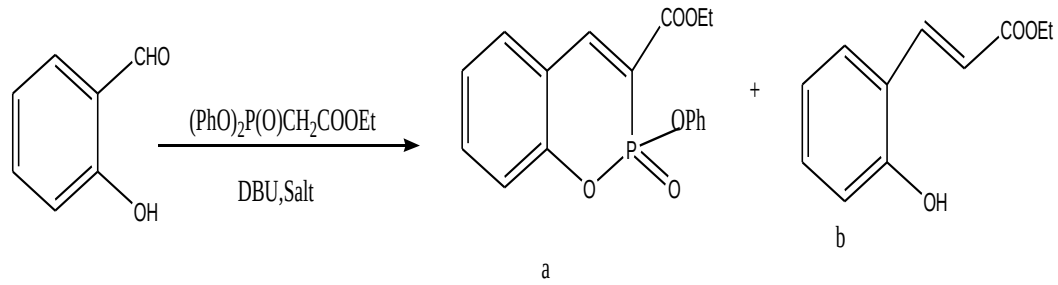


Figure 11: Reaction of Salicylaldehyde with (PhO)2P(O)CH2COOEt.

2.3 Perkin Reaction

2.3.1 Coumarin was synthesised by the Perkin reaction using salicylic acid, acetic acid and sodium acetate.[13]

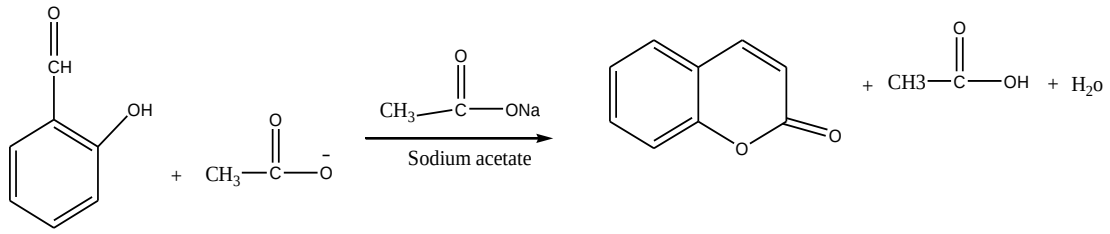


Figure 12: Reaction of salicylic acid with sodium acetate.

2.3.2 Using Perkin condensation, Augustine and colleagues created a one-pot synthesis of substituted coumarins. A model reaction of cyanoacetic acid and salicylaldehyde was selected.

To find the ideal reaction conditions, eight reactions with various parameters (time, temperature, and molarity of propylphosphonic anhydride T3P) were carried out. T3P, TEA (triethylamine), and n-BuOAc (butyl acetate) were present for every reaction.[2]

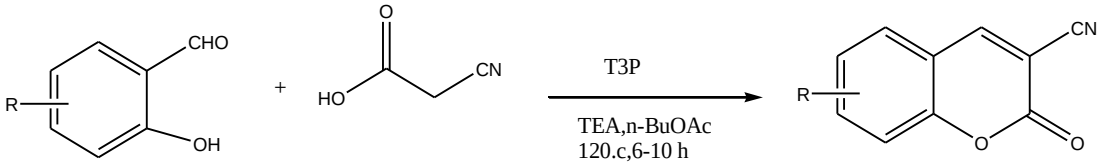


Figure 13: Reaction of salisylaldehyde with cyanoacetic acid.

2.4 Knoevenagel Reaction

2.4.1 When 4-substituted benzaldehydes and active methylene compounds undergo Knoevenagel condensation reactions, 1,3-dimethylimidazolium methyl sulfate, [MMIm][MSO4], an inexpensive, easily prepared, low-viscosity ionic liquid with low cytotoxicity, can function effectively as the solvent and catalyst without the need for a promoter other than a tiny quantity of water (≈2%). High yields of the required coumarins were obtained by heating the reagents and promoter at 90 °C in undried [MMIm][MSO4]

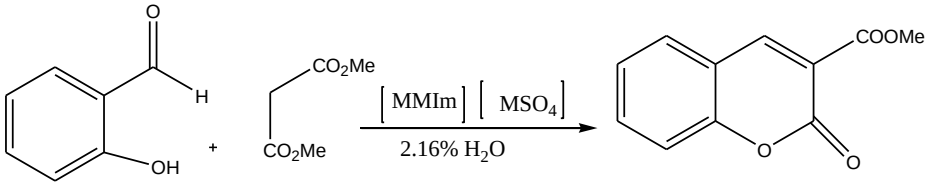


Figure 14: Synthesis of coumarins in undried [MMIm][MSO4] containing L-proline.

2.4.2 Through the Knoevenagel condensation reaction of an orthohydroxyaryl aldehyde and an activated β-dicarbonyl C-H acid in the presence of a recyclable ionic liquid 1,1,3,3-N, N, N', N'-tetramethylguanidinium trifluoroacetate, coumarin derivatives were produced in comparatively high yields using either classical heating conditions or microwave irradiation.[14]

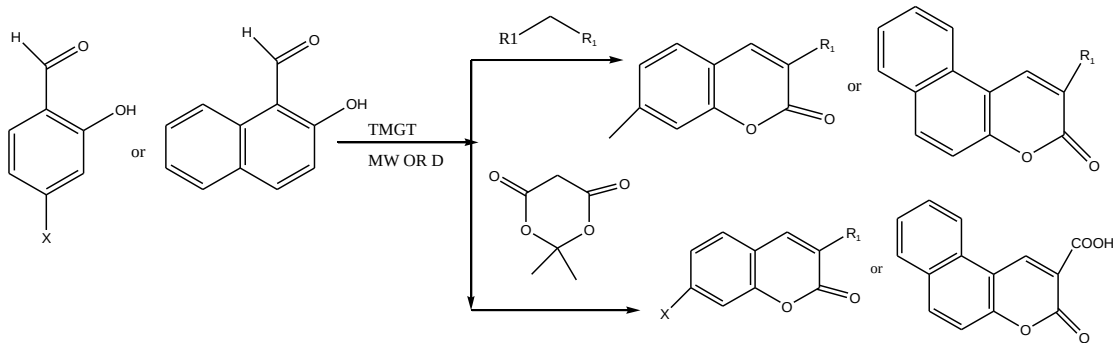


Figure 15: Coumarin-synthesis-via-Knoevenagel-condensation-reaction-in-1-1-3-3-N-N-N-N-tetramethylguanidinium-trifluoroacetate-ionic-liquid.

3. Pharmacological Applications

Derivatives of coumarin are reported to possess numerous therapeutic applications, including photochemotherapy, central nervous system stimulation, lipid-lowering effects, enzyme inhibition, antidiabetic properties, antioxidant activity, anti-inflammatory effects, anticoagulation, anticancer properties, and antifungal activity, among others.[15] The pharmacological applications of coumarins are illustrated in Fig 16.

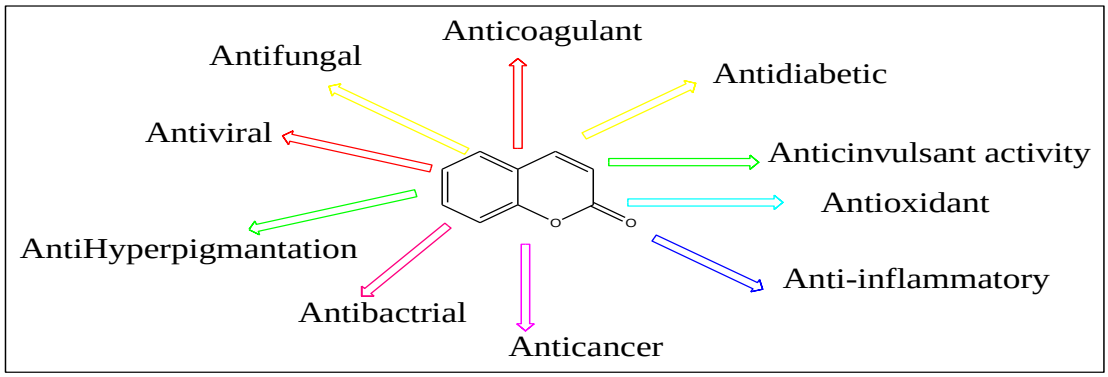


Fig 16: Pharmacological applications of coumarins.

4.1 Antibacterial activity

The compounds 6-hydroxy-7-methoxycoumarin, 7-hydroxy-5,6-dimethoxycoumarin, and 6,8-dihydroxy-5,7-dimethoxycoumarin were assessed against eight microorganisms, comprising three Gram-positive bacteria (Staphylococcus aureus, beta-hemolytic Streptococcus, and Streptococcus pneumoniae) and five Gram-negative bacteria (Escherichia coli, Klebsiella).

pneumoniae, Pseudomonas aeruginosa, Proteus mirabilis, and Haemophilus influenzae, employing the microdilution broth technique. The tested coumarins exhibited considerable variability concerning Growth inhibitory activity, with minimum inhibitory doses varying from 0.9 to 12.4 µM.[1] Coumarin inhibitors of DNA gyrase B containing an N-propargyloxycarbamate at C-30 of different 50,50. -Dialkylnoviose, including RU79115, was synthesised, and its antibacterial properties have been characterised. Introduction of dialkyl substituents at position 505. The positioning of noviose results in coumarin analogues with enhanced antibacterial activity both in vitro and in vivo.[16] Compounds comprising 1,2,3-triazole-substituted coumarins and 1,2,3-triazolyl or 1,2,3-triazolylalk-1-inyl-linked coumarin-2,3-furocoumarin hybrids were assessed for their in vitro antibacterial efficacy against Staphylococcus aureus, Bacillus subtilis, Actinomyces viscosus, and Escherichia coli bacterial strains. Demonstrated encouraging efficacy against S. aureus strains, in comparison to several contemporary antibiotic agents utilised in clinical settings, indicating favourable possibilities for further investigation.[17] Coumarin-3-carboxylic acid (3-CCA), identified from natural coumarins, had significant antibacterial efficacy against Acidovorax citrulli (Ac). To assess the efficacy of this molecule against plant bacterial pathogens and investigate its potential as a bactericidal lead drug, the activity of 3-CCA was evaluated against 14 plant pathogenic bacteria both in vitro and in vivo. The results indicated that 3-CCA showed significant in vitro efficacy against Ac, Ralstonia solanacearum, Xanthomonas axonopodis pv. Manihotis, X.

oryzae pv. oryzae, and Dickeya zeae, with EC50 values between 26.64 µg/mL and 40.73 µg/mL.[18] The synthesised compounds 4-hydroxycoumarin showed notable antibacterial activity against both Gram-positive and Gram-negative bacterial strains, with compound 4 exhibiting the most effective antibacterial activity against all tested bacterial species.

Structural changes, including the incorporation of nitrogen atoms and azomethane groups, likely augmented antibacterial effectiveness by enhancing membrane permeability and disrupting bacterial cellular activities.[19] Figure 18 demonstrates the competitive binding of coumarin derivatives to the ATP-binding pocket of the GyrB subunit, inhibiting the hydrolysis of energy required for DNA resealing. This immobilises the enzyme-DNA complex and inhibits supercoiling, resulting in irreversible DNA damage and bacterial cell mortality.

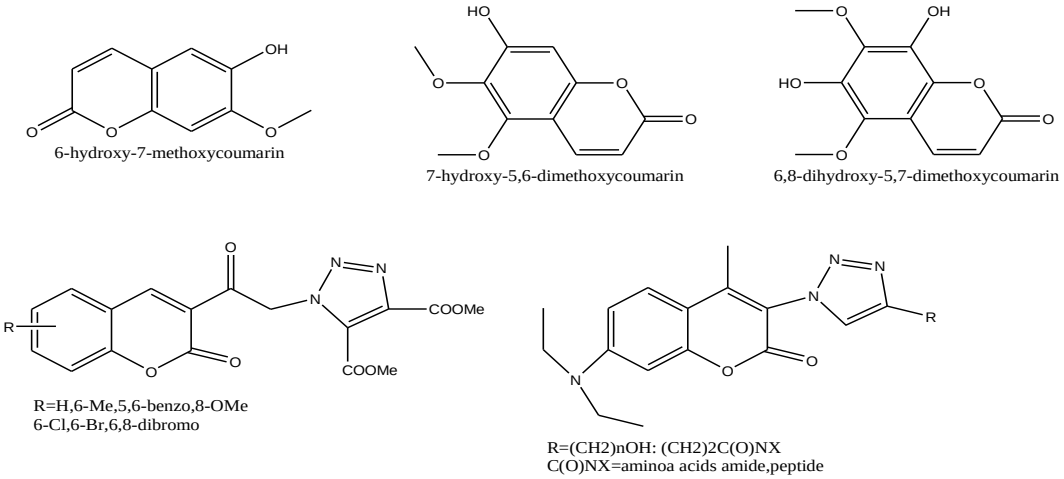


Fig 17: Antibacterial Coumarin derivatives.

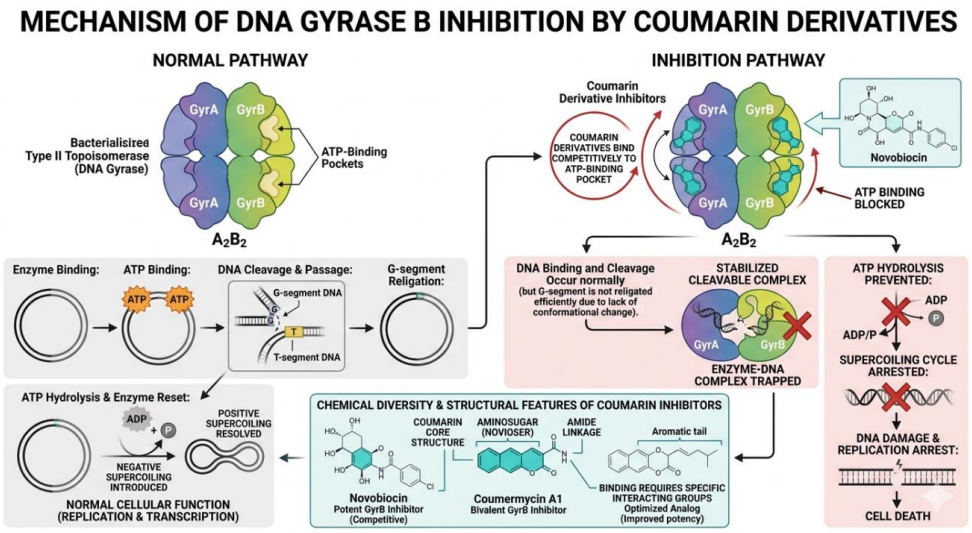


Figure 18: Antibacterial mechanism of Coumarin derivatives.

4.2 Anticoagulant activity

A prior endeavour was conducted by Chen et al. to identify platelet anti-aggregatory drugs. 4-hydroxycoumarin derivatives were produced through the alkylation of the hydroxyl group (Fig.18). The platelet anti-aggregatory efficacy of the synthesised compounds was examined against collagen (Co1)-induced, thrombin (Thr)-induced, arachidonic acid (AA)-induced, and platelet-activating factor (PAF)-induced platelet aggregation in washed rabbit platelets. Among the synthesised derivatives.[20] Lu et al. conducted a study examining the efficacy of three compounds, 6-8, containing a coumarin nucleus, against ADP-induced platelet aggregation. The three compounds demonstrated the ability to inhibit the active form of the GPIIb/IIIa complex. Rights reserved.The membrane of the platelets, thereby facilitating platelet

aggregation. Furthermore, the downstream signal transduction of the ADP receptor, encompassing calcium ion release and cAMP modulation, was likewise observed to be blocked by the three derivatives(Fig.19-21).[21] Katerina et al. conducted a study examining the antiplatelet activity, elicited by AA, collagen, and ADP, of previously produced simple 4-methylcoumarin derivatives.

4-methylcoumarin derivatives with hydroxyl groups at positions 5 and 7, especially those featuring a lipophilic side chain at C3, Fig.22 demonstrated platelet antiaggregatory activity comparable to acetylsalicylic acid in AA-induced aggregation (IC50 = 16.1 μM), with IC50 values of 17.5 μM and 23.3 μM, respectively. These compounds are promising candidates for the expansion of the existing repertoire of antiplatelet medications. Moreover, compounds 9a and 9b demonstrated COX-1 inhibitory action. They were determined to be more effective inhibitors than Acetylsalicylic Acid.[22]

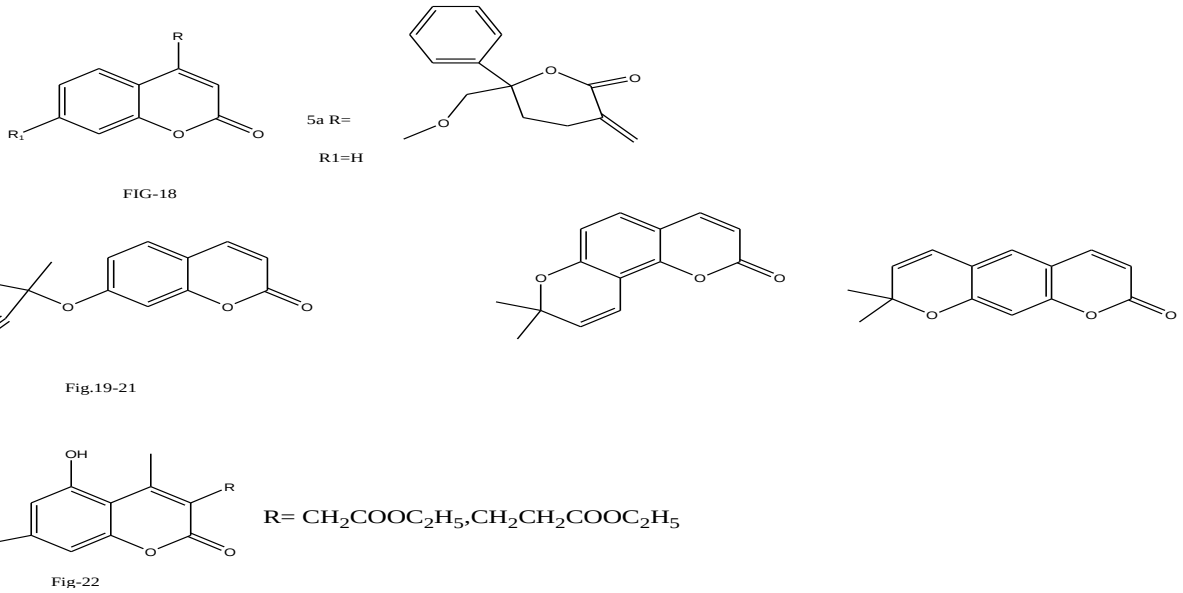


Fig 18: Anticoagulant Coumarin derivatives.

4.3 Antioxidant activity

Compounds 23 to 25 were evaluated for their antioxidant activity using the DPPH (2,2-diphenyl-1-picrylhydrazyl) radical scavenging assay at concentrations ranging from 200 μg/mL to 1000 μg/mL. The assay elucidates the capacity of antioxidants to neutralise free radicals and mitigate oxidative damage. Compound concentrations were varied to observe dose-dependent responses and assess the kinetics of the interaction between antioxidants and DPPH radicals.

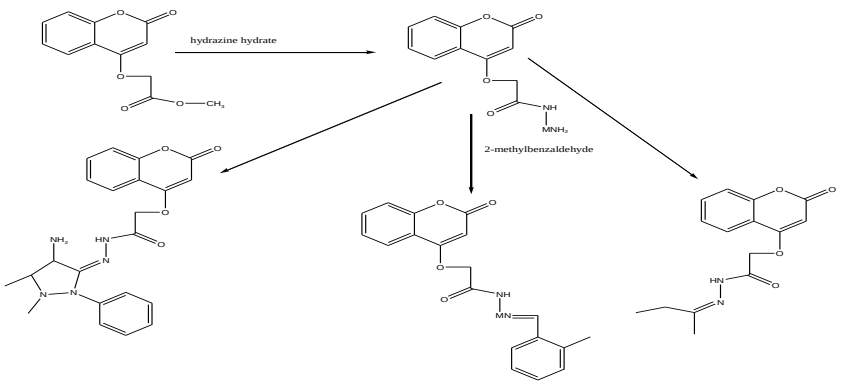


Fig 19: Antioxidant Coumarin derivatives.

4.4 Anti-inflammatory activity

In order to prevent an injury or illness from getting worse, the body uses inflammation as a defence mechanism. Overall, controlling inflammation is essential. Non-steroidal anti-inflammatory drugs are used to alleviate inflammation. Through various processes, a number of naturally occurring coumarins, including esculetin, umbelliferone, and scopoletin, have potent anti-inflammatory activity. A series of 3-methyl-1-phenyl-chromenol-[4,3-c]-pyrazole-4(1H)-ones were synthesized coumarin compounds. The mechanism of action of coumarin involves phagocytosis, the production of enzyme proteins, and the removal of oedema fluid. The anti-inflammatory properties of coumarin are used to treat oedema. Coumarins' role in anti-inflammatory activity can be divided into three categories: sensor level (MAP kinase), mediator level (prostaglandin inhibitor, inos inhibitor), and protective level (rheumatoid arthritis, atherosclerosisZA), as given in Fig.20.[15]

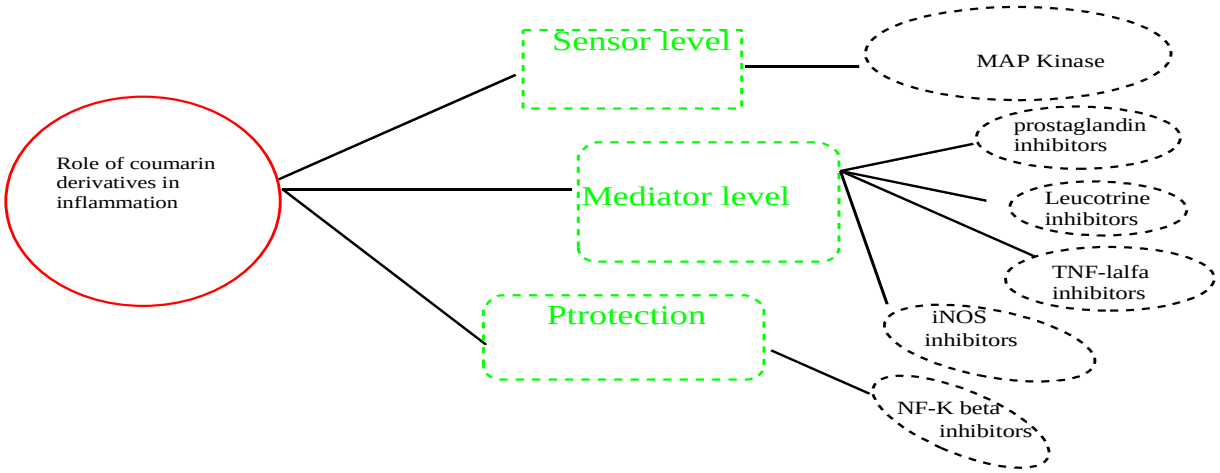


Fig 20: Role of coumarin derivatives in inflammation activity.

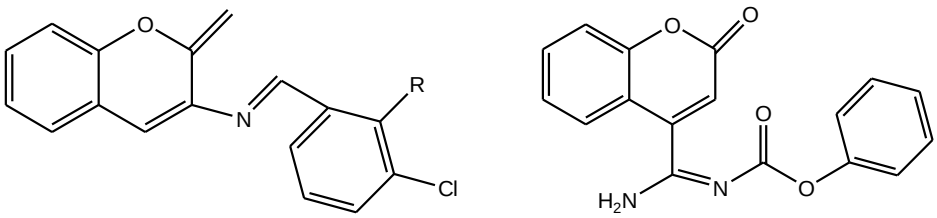


Fig 21: Anti inflammatory Coumarin derivatives.

4.5 Anti-Hyperpigmentation Coumarin

The coumarin derivatives coumophos and scandenin-robustic acid methyl ether are employed as components of antiperspirants and as additions in medications These formulations may be used to prevent the condition of undesired skin darkening, which may be brought on by melanocytes' production of melanin Fig 22.[15]

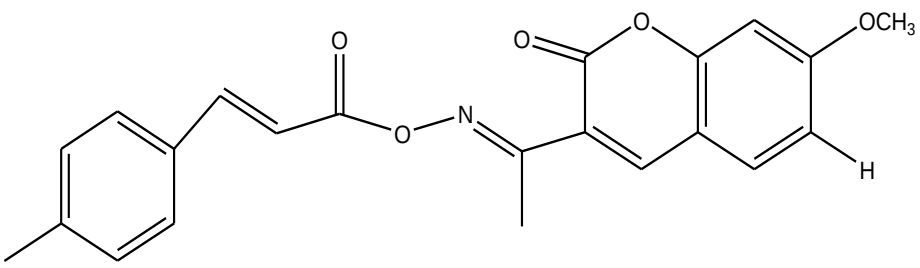


Fig 22: Anti-Hyperpigmentation Coumarin derivatives.

CONCLUSION

Coumarins constitute a "privileged scaffold" in medicinal chemistry, defined by a flexible benzopyran-2-one nucleus that facilitates wide oxygenation and substitution patterns. Although conventional synthetic methods, such as the Pechmann condensation, have progressed into effective, environmentally friendly techniques—employing ionic liquids, microwave irradiation, and recyclable catalysts—the clinical utilization of these derivatives remains constrained by challenges including inadequate aqueous solubility, swift metabolic degradation, and possible photosensitivity. The future of coumarin research is hopeful, especially with the advancement of nanotechnology-based delivery systems and the creation of multi-action hybrid molecules aimed at addressing complicated issues such as multi-drug resistant bacteria and neurodegenerative illnesses.

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