



PLANT DERIVED CATALYSTS MEDIATED GREEN SYNTHESIS OF BIOACTIVE HETEROCYCLIC COMPOUNDS

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ABSTRACT

Heterocyclic compounds are indispensable in medicinal chemistry because nitrogen-, oxygen-, and sulfur-containing rings not only play a crucial role in determining molecular geometry, polarity, hydrogen bonding capacity, and basicity, but also modulating interaction with biological targets. Conventional synthesis of heterocycle compounds, however, require hazardous corrosive acids or bases, toxic metal catalysts, volatile organic solvents, prolonged heating, and complex purification. Plant-derived catalysts provide a renewable and sustainable alternative, utilizing acidic fruit juices, aqueous botanical extracts, agricultural-waste ashes, natural biopolymers, and plant-mediated metal or metal-oxide nanoparticles. This review explains the utilizing of this sustainable catalytic system in the construction of different bioactive heterocycles such as pyrans, chromenes, pyrazoles, isoxazolones, quinazolinones, pyrimidines, imidazoles, indoles, and benzimidazoles. Organic acids such as Citric acid ($C_6H_8O_7$), Malic acid ($C_4H_6O_5$), and Ascorbic acid ($C_6H_8O_6$) promote carbonyl activation and cyclocondensation. Plant-ash extracts contain alkaline species including potassium carbonate (K_2CO_3), calcium oxide (CaO), calcium carbonate ($CaCO_3$), and magnesium oxide (MgO), which facilitate Knoevenagel Condensation, Michael addition, and ring cyclization. Plant phytochemicals can also reduce or stabilize CuO , ZnO , Fe_3O_4 , and Ag^0 nano-catalysts used in different coupling and multicomponent reactions. These systems simplify isolation and lower the need for hazardous reagents, as shown in by representative balanced equations and structural schemes. However, there are still significant challenges, including catalyst variability, unclear active-site chemistry, metal leaching, inadequate green-metric reporting, scale-up restrictions, and limited biological validation. Prior to broad industrial use, standardized catalyst preparation, mechanistic controls, pharmaceutical impurity analysis, and quantitative sustainability assessment are necessary.

KEYWORDS: Green Chemistry; Heterocyclic compounds; Plant-based Catalysts; Agricultural-waste ash; biogenic nano-catalysts; Pharmaceutical Synthesis.

INTRODUCTION

Heterocycles are cyclic organic compounds in which at least one ring atom is present as an element other than carbon. Nitrogen heterocycles such as pyridine (C_5H_5N), pyrimidine ($C_4H_4N_2$), imidazole ($C_3H_4N_2$), pyrazole ($C_3H_4N_2$), indole (C_8H_7N), and benzimidazole ($C_7H_6N_2$) are especially important in pharmaceutical chemistry. Oxygen-containing systems such as pyran (C_5H_6O), chromene (C_9H_8O), and coumarin ($C_9H_6O_2$) are also widely investigated. Their heteroatoms participate in hydrogen bonding, ionic interactions, metal coordination, and dipole-driven recognition;

consequently, small structural changes can substantially alter biological activity, selectivity, solubility, and metabolic stability.^[1-3]

The environmental profile of heterocycle synthesis is frequently less favorable than its scientific importance. Traditional methods may employ concentrated H_2SO_4 or HCl , strong bases such as $NaOH$ or KOH , transition-metal salts, chlorinated solvents, N,N -dimethylformamide, or high-temperature reflux. Linear synthesis further increases waste because intermediates must be isolated, washed, dried, and purified before the

next reaction. Green chemistry seeks to prevent this burden through catalysis, safer solvents, renewable feedstocks, high atom economy, energy-efficient activation, and avoidance of unnecessary derivatization.^[1,2]

Plant-based catalysis is attractive because botanical materials contain chemically active organic acids, polyphenols, flavonoids, reducing sugars, amino acids, enzymes, and mineral salts. A fruit juice may act as a Brønsted-acid catalyst; an agricultural-waste ash may supply basic carbonate or oxide species; and a plant extract may reduce and cap metal nanoparticles. These systems can be combined with one-pot multicomponent reactions, water or ethanol, microwave irradiation, ultrasound, or solar heating. The purpose of this review is to examine the underlying chemistry, representative heterocyclic transformations, pharmaceutical relevance, sustainability performance, and unresolved research requirements of plant-based catalytic synthesis.^[3,4]

Review methodology

A narrative literature review was conducted using peer-reviewed journal articles and authoritative green-chemistry literature. Search concepts included “plant extract catalyst,” “fruit juice catalyzed heterocycle,” “plant ash 4H-pyran,” “biogenic CuO N-arylation,” “green multicomponent heterocycle,” and “pharmaceutical heterocyclic synthesis.” Priority was

given to studies that clearly identified the botanical source, reactants, heterocyclic product class, catalyst role, reaction medium, yield or reuse performance, and product characterization method. General claims were interpreted cautiously because catalyst composition and biological outcomes vary substantially among studies. The present paper is therefore a critical review rather than an experimental report or a systematic meta-analysis.

1. Chemical basis of plant-based catalysis

a. Acidic fruit juices and aqueous extracts

Lemon, lime, tomato, tamarind, and related plant materials contain weak organic acids that can activate electrophiles without the handling hazards associated with concentrated mineral acids. Lemon juice commonly contains citric acid ($C_6H_8O_7$), ascorbic acid ($C_6H_8O_6$), and smaller amounts of malic acid ($C_4H_6O_5$). These acids protonate aldehyde or ketone carbonyl oxygen, increasing the partial positive charge at carbon and accelerating attack by amines, amides, hydrazines, hydroxylamine, or activated methylene nucleophiles. Proton transfer also supports dehydration and imine or iminium-ion formation during cyclocondensation.^[3,5]

Stepwise citrate ionisation: $H_3Cit \rightleftharpoons H^+ + H_2Cit^- \rightleftharpoons 2H^+ + HCit^{2-} \rightleftharpoons 3H^+ + Cit^{3-}$ *citrate framework*

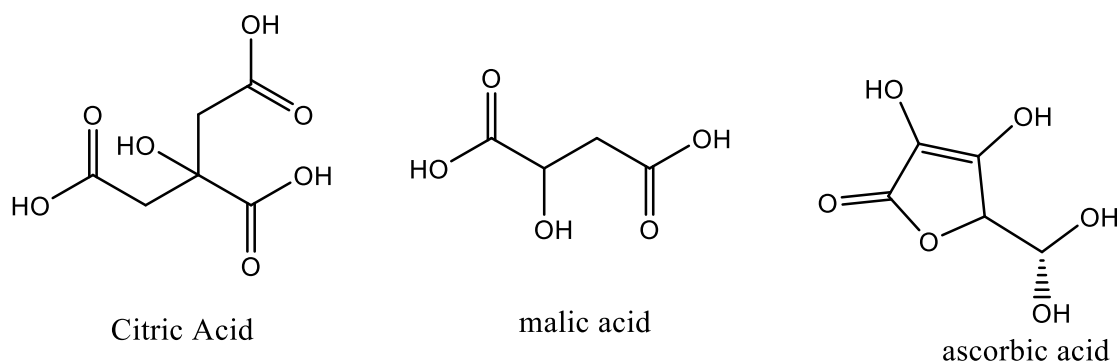


Figure 1: Representative organic acids contributing to the Brønsted-acid character of plant juices.

Crude juice is not a single-component catalyst. Its performance depends on pH, titratable acidity, water content, maturity, cultivar, storage, and filtration. A scientifically reproducible protocol should therefore report botanical identity, extraction procedure, pH, catalyst volume, and—where possible—the concentrations of major organic acids. Comparing the juice with an aqueous solution of pure citric acid at the same pH can reveal whether catalysis results mainly from acidity or from cooperative action among multiple phytochemicals.

b. Agricultural-waste ash as a basic catalyst

When peels, seeds, shells, stems, or leaves are dried and combusted under controlled conditions, their inorganic constituents remain as ash. Water extracts of these ashes

may contain K^+ , Ca^{2+} , Mg^{2+} , carbonate (CO_3^{2-}), bicarbonate (HCO_3^-), hydroxide (OH^-), phosphate, and silicate species. The exact composition varies with the plant, soil, washing procedure, and calcination temperature. Carbonate hydrolysis produces hydroxide ions, allowing the extract to deprotonate malononitrile, β -dicarbonyl compounds, and related C–H acids.^[6,7]

Base generation in ash extract: $CO_3^{2-} + H_2O \rightleftharpoons HCO_3^- + OH^-$ OH^- initiates enolate or carbanion formation.

Activated methylene deprotonation: $CH_2(CN)_2 + OH^- \rightleftharpoons ^-CH(CN)_2 + H_2O$ malononitrile forms a resonance-stabilized nucleophile.

Water extract of *Tamarindus indica* seed ash has been used as both reaction medium and basic catalyst for one-pot synthesis of 4H-pyrans, with reported reuse for four cycles.^[6] Water extract of mango peel ash has been applied under microwave irradiation to pyrano[2,3-*c*]pyrazoles and pyrazolyl-4H-chromenes.^[7] Such methods valorise agricultural residue, but the overall environmental benefit depends on controlled combustion, energy use, washing volume, ash disposal, and the absence of toxic trace elements.

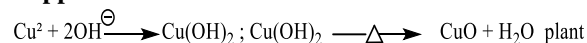
c. Plant-mediated metal and metal-oxide nano catalysts

Plant polyphenols, reducing sugars, proteins, and organic acids can complex metal ions and regulate nucleation during nanoparticle preparation. For silver nanoparticles, phytochemical oxidation supplies electrons for reduction of Ag^+ to metallic Ag^0 . For metal oxides such as CuO, the extract can promote precursor formation, restrict aggregation, and remain as a surface capping layer. The resulting high-surface-area catalysts may support cross-

coupling, oxidation, reduction, and multicomponent cyclisation.^[8,9]

Representative silver reduction: $\text{Ag}^+ + \text{e}^- \rightarrow \text{Ag}^0$
electrons are supplied by oxidised phytochemicals

Copper oxide formation



plant molecules assist nucleation and stabilization
Ocimum sanctum-mediated CuO nanoparticles have been reported for N-arylation of amides, amines, pyrrole, indole, imidazole, benzimidazole, and carbazole.^[8] *Tamarix gallica* leaf extract has similarly been used to prepare CuO for ligand-free N-arylation of nitrogen heterocycles.^[9] Because the intended products may enter pharmaceutical screening, inductively coupled plasma analysis or an equivalent method should quantify residual Cu, Ag, or other metals in the isolated compounds.

Table 1: Representative plant-derived catalytic systems for heterocyclic synthesis.

Plant-derived catalyst	Principal chemical species	Representative transformation	Green features / evidence
Citrus limon juice	$\text{C}_6\text{H}_8\text{O}_7$, $\text{C}_6\text{H}_8\text{O}_6$, $\text{C}_4\text{H}_6\text{O}_5$; H^+ donor	Isoxazol-5(4H)-ones and pyrano[2,3- <i>c</i>]pyrazoles	Renewable aqueous catalyst; hydroalcoholic medium ^[5]
Tamarind seed ash extract	K_2CO_3 , CaCO_3 , MgO and soluble alkaline ions	Three-component 4H-pyran synthesis	Water as medium; catalyst reused for four cycles ^[6]
Mango peel ash extract	Carbonate/oxide-rich alkaline mineral fraction	Pyrano-pyrazoles and pyrazolyl chromenes	Agro-waste valorisation; microwave activation; simple filtration ^[7]
<i>Ocimum sanctum</i> -mediated CuO	CuO nanoparticles with phytochemical surface layer	C–N coupling and N-arylation of N-heterocycles	Ligand-free catalysis and broad substrate scope ^[8]
<i>Tamarix gallica</i> -mediated CuO	CuO nanoparticles	N-arylation of nitrogen-containing heterocycles	Recoverable heterogeneous catalyst ^[9]
Lemon juice + solar heating	Organic acids; concentrated solar radiation	2-Aryl/heteroaryl-2,3-dihydroquinazolin-4(1H)-ones	Up to 97% yield in 10 min for the reported model system ^[10]
Coconut, tomato or sweet-lime juice	Organic acids, salts and biomolecules	Three-component substituted isoxazoles	One-pot synthesis with in-vitro bioevaluation ^[11]

2. Representative heterocyclic synthesis pathways

a. 4H-Pyrans and chromenes

A common 4H-pyran route combines an aromatic aldehyde (Ar-CHO), malononitrile [$\text{CH}_2(\text{CN})_2$], and a cyclic 1,3-dicarbonyl compound such as dimedone ($\text{C}_8\text{H}_{12}\text{O}_2$). The basic plant-ash medium first generates a malononitrile carbanion. Knoevenagel condensation forms an arylidene malononitrile intermediate, which accepts Michael addition by the enolate of the 1,3-dicarbonyl compound. Intramolecular O-cyclisation and tautomerization then furnish the substituted 2-amino-4H-pyran or chromene framework.

Knoevenagel step: $\text{Ar-CHO} + \text{CH}_2(\text{CN})_2 \rightarrow \text{Ar-CH=C(CN)}_2 + \text{H}_2\text{O}$ base-catalysed condensation.

Cyclisation sequence: $\text{Ar-CH=C(CN)}_2 + \text{cyclic 1,3-dicarbonyl} \rightarrow 2\text{-amino-4-aryl-4H-pyran-3-carbonitrile}$
Michael addition followed by O-ring closure.

The value of this sequence is operational rather than merely chemical: three substrates are combined in one vessel, most atoms appear in the product, and precipitation can avoid column chromatography. Substituent effects remain important. Electron-withdrawing groups on Ar-CHO generally increase carbonyl electrophilicity and can accelerate the initial condensation, whereas strongly steric substituents may retard both condensation and ring closure.

b. Pyrano[2,3-*c*] pyrazoles

Pyranopyrazoles are commonly assembled from an aldehyde, malononitrile, ethyl acetoacetate ($\text{C}_6\text{H}_{10}\text{O}_3$), and hydrazine hydrate ($\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$). Hydrazine and ethyl acetoacetate first generate a pyrazolone intermediate

with elimination of ethanol ($\text{C}_2\text{H}_5\text{OH}$). In parallel, the aldehyde and malononitrile undergo Knoevenagel condensation. C–C bond formation between these intermediates, followed by O-cyclisation, gives a 6-amino-pyrano[2,3-c] pyrazole-5-carbonitrile derivative. Lemon juice can facilitate carbonyl activation, whereas mango-peel-ash extract can provide the complementary basic pathway.^[5,7]

Pyrazolone formation: $\text{C}_6\text{H}_{10}\text{O}_3 + \text{N}_2\text{H}_4 \cdot \text{H}_2\text{O} \rightarrow$ pyrazolone intermediate + $\text{C}_2\text{H}_5\text{OH} + \text{H}_2\text{O}$ condensation of ethyl acetoacetate with hydrazine.

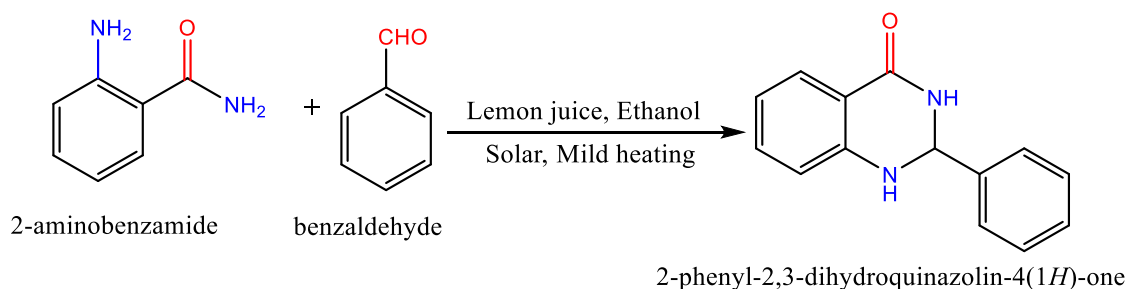


Figure 2: Representative acid-catalyzed formation of 2-phenyl- 2,3-dihydroquinazolin-4(1H)-one.

d. N-Arylation of nitrogen heterocycles

Plant-mediated CuO catalysis extends sustainable synthesis from ring construction to ring functionalisation. In an Ullmann-type N-arylation, a nitrogen heterocycle reacts with an aryl halide (Ar-X , where $\text{X} = \text{I}$ or Br) in the presence of base. Surface Cu sites facilitate C–X activation and C–N bond formation. The base converts

the acidic N–H group into a more nucleophilic nitrogen species and neutralises the hydrogen halide by-product.^[8,9]

General coupling: $\text{Ar-X} + \text{Het-NH} + \text{base} + \text{CuO} \rightarrow \text{Ar-N(Het)} + \text{base} \cdot \text{HX}$ Het = imidazolyl, indolyl, benzimidazolyl, etc.

Balanced example: $\text{C}_3\text{H}_4\text{N}_2 + \text{C}_6\text{H}_5\text{I} \rightarrow \text{C}_9\text{H}_8\text{N}_2 + \text{HI}$ imidazole + iodobenzene $\rightarrow$ 1-phenylimidazole

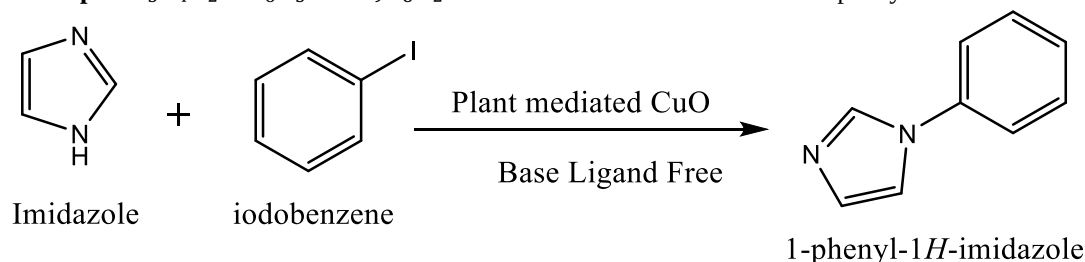


Figure 3: Representative plant-mediated CuO-catalysed N-arylation of imidazole.

3. Pharmaceutical relevance

Heterocyclic rings are often described as privileged scaffolds because they provide compact, three-dimensional arrangements of hydrogen-bond donors, acceptors, aromatic surfaces, and ionisable atoms. Pyrimidine derivatives are explored as kinase inhibitors, antivirals, antibacterials, and anticancer agents. Pyrazoles and pyranopyrazoles are associated with anti-inflammatory, antimicrobial, antioxidant, and enzyme-inhibitory research. Quinazolinone and dihydroquinazolinone derivatives have been studied for antimicrobial, anticonvulsant, anti-inflammatory, and antiproliferative activity, while imidazole and

benzimidazole systems occur in numerous clinically important pharmacophores.^[4]

Plant-catalyzed methods can accelerate medicinal-chemistry library generation because multicomponent reactions provide several substitution points in a single operation. Changing the aromatic aldehyde, activated methylene compound, hydrazine derivative, or aryl halide produces structurally related analogues for structure–activity relationship analysis. The lower cost and simpler equipment may also improve access to compound synthesis in resource-limited research environments.

However, a compound should not be described as a pharmaceutical merely because it contains a recognized heterocyclic ring or shows one preliminary biological response. DPPH radical scavenging, agar diffusion, molecular docking, or single-cell-line cytotoxicity are screening methods, not proof of therapeutic value. Credible pharmaceutical development requires complete spectroscopic characterization, purity assessment, normal-cell selectivity, target validation, dose–response analysis, ADME profiling, genotoxicity evaluation, in-vivo testing, formulation studies, and reproducible manufacture. Residual plant metabolites or metals must be excluded as the cause of apparent bioactivity.

4. Quantitative evaluation of sustainability

Terms such as natural, biodegradable, and eco-friendly are insufficient without quantitative comparison. Yield measures product formation, but it does not account for solvent, excess reagents, purification materials, energy, or catalyst manufacture. A robust assessment should report atom economy, reaction mass efficiency, E-factor, process mass intensity, solvent intensity, energy per gram of product, catalyst turnover, and verified reuse performance.^[1,2]

Table 2: Green chemistry metrics applicable to plant-catalyzed heterocyclic synthesis.

Metric	Chemical expression	Interpretation
Atom economy (AE, %)	$AE = [M_r(\text{desired product}) / \Sigma M_r(\text{stoichiometric reactants})] \times 100$	Measures the theoretical fraction of reactant atoms retained in the product.
Reaction mass efficiency (RME, %)	$RME = [\text{mass of isolated product} / \text{mass of reactants charged}] \times 100$	Includes yield and stoichiometric excess but normally excludes solvents.
E-factor	$E = \text{mass of waste} / \text{mass of product}$	Lower values indicate less waste; the definition of water should be stated.
Process mass intensity (PMI)	$PMI = \text{total mass input} / \text{mass of product}$	Includes reactants, catalysts, solvents, water, and processing materials.
Turnover number (TON)	$TON = \text{moles of product} / \text{moles of active catalyst}$	Measures total catalytic productivity.
Turnover frequency (TOF)	$TOF = TON / \text{reaction time}$	Measures catalytic productivity per unit time.

The catalyst boundary must be defined carefully. For a plant-ash process, drying, combustion, aqueous extraction, filtration, and disposal should be included. For nanocatalysts, metal salt manufacture, extract preparation, washing, calcination, catalyst loss, and metal-contaminated effluent must be counted. A high-yielding reaction can still have poor PMI if it uses large solvent volumes or chromatography. Conversely, a moderately yielding process may be attractive if the product crystallizes directly, the solvent is recycled, and the catalyst retains activity over many cycles.

5. Limitations and future research priorities

The principal scientific limitation is catalyst variability. Organic-acid content changes with cultivar and maturity, while ash composition changes with soil, plant part, washing, and combustion temperature. Plant-mediated nanoparticle size and surface chemistry depend on extract concentration, metal-ion ratio, temperature, pH, and ageing time. Studies should therefore report botanical authentication, collection conditions, extraction ratio, pH, conductivity, total acidity or alkalinity, major ion composition, phenolic content where relevant, and catalyst storage stability.

Mechanistic evidence is often incomplete. Appropriate controls include reactions without catalyst, reactions at matched pH using a pure acid or base, hot-filtration tests for heterogeneous catalysts, poisoning experiments,

kinetic analysis, and catalyst characterization before and after reuse. For ash extracts, ion chromatography or elemental analysis can identify the dominant alkaline species. For CuO, Ag, ZnO, or Fe₃O₄ catalysts, XRD, XPS, electron microscopy, surface-area measurements, and leaching analysis should be correlated with activity.

Scale-up requires more than increasing flask volume. Microwave penetration, solar intensity, ultrasound distribution, mixing, heat transfer, and product precipitation can change with reactor size. Continuous-flow reactors, immobilised biopolymer catalysts, magnetic recovery, and fixed-bed nanocatalysts may offer better control. Pharmaceutical translation also requires good manufacturing practice-compatible catalyst specifications, residual-metal limits, validated impurity profiles, batch reproducibility, solvent recovery, and safe biomass sourcing.

Future work should integrate green synthesis with medicinal evaluation from the beginning. Compound libraries should be screened with positive and negative controls, normal-cell comparators, orthogonal target assays, and early ADME-toxicity models. Life-cycle assessment should compare the plant-based route with the best conventional route rather than with an intentionally inefficient benchmark. This combination of mechanistic chemistry, quantitative sustainability, and pharmaceutical quality will determine whether plant

catalysts become practical manufacturing tools rather than attractive laboratory demonstrations.

6. CONCLUSION

Plant-based catalysts provide chemically diverse tools for sustainable heterocyclic synthesis. Fruit-derived organic acids such as $C_6H_8O_7$, $C_4H_6O_5$, and $C_6H_8O_6$ promote carbonyl activation, dehydration, and ring closure. Agricultural-waste ash supplies CO_3^{2-} , OH^- , K^+ , Ca^{2+} , and related mineral species that catalyze Knoevenagel condensation and Michael addition. Plant phytochemicals also assist the formation and stabilisation of CuO, Ag^0 , ZnO, and Fe_3O_4 nanocatalysts for coupling and multicomponent reactions. These systems have been applied to pharmaceutically relevant heterocycles and their derivatives such as pyrans, chromenes, pyrazoles, isoxazoles, quinazolinones, imidazoles, indoles, and benzimidazoles. The advantage of this protocol includes one-pot operation, renewable catalyst sources, agricultural-waste valorization, reduced hazardous reagent consumption, and simplified isolation. Nevertheless, reproducibility, mechanistic certainty, metal-leaching control, scale-up, and rigorous biological validation remain essential. Plant-based catalytic system will make a credible contribution to pharmaceutical sustainability only when its chemical performance and environmental benefit are measured with the same precision expected of conventional synthetic methods.

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