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Gallic Acid and its Derivatives: A Review of their Antioxidant Properties and Applications for Fossil Fuel Preservation

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Abstract

For many industrial and home applications, fossil fuels serve as the primary energy source. They are, however, prone to oxidative breakdown when exposed to air, light, heat, and metal catalysts, which results in the development of dangerous compounds such gums, sediments, acids, and peroxides. These products have the potential to negatively impact fuel quality, performance, and storage stability. Effective antioxidants that can neutralize free radicals and prevent oxidation are therefore required. Because of its capacity to donate hydrogen atoms and create stable radicals, gallic acid (3,4,5-trihydroxybenzoic acid) is a naturally occurring phenolic molecule with considerable antioxidant activity. Gallic acid and its derivatives have received a lot of attention for their potential use as antioxidants in a variety of industries, including food, cosmetics, medicine, and biotechnology. Gallic acid and its derivatives' chemistry, biology, and production, as well as their antioxidant mechanisms and uses for preserving fossil fuels, are all covered in this review. We also go over the benefits and drawbacks of using these substances as fossil fuel antioxidants, and we highlight the difficulties that now face their development and optimization as well as the potential that lie ahead.

Key words: Gallic acid; Antioxidants; Fossil fuels; Oxidative degradation; Phenolic compounds

1. Introduction

When fossil fuels are burned, oxygen reacts with carbon and hydrogen to produce water, carbon dioxide, heat, and light. However, sulfur and nitrogen in the fuel and air also create pollutants like sulfur dioxide (SO_2) and nitrogen oxides (NO_x). These pollutants can lead to acid rain, which damages structures, harms vegetation, and disrupts aquatic ecosystems. [1-8].

This review paper on gallic acid and its derivatives explores their antioxidant properties and applications, particularly in fossil fuel preservation. While focusing on these areas, the paper also highlights the broader relevance of the United Nations Sustainable Development Goals (SDGs). By investigating the role of gallic acid in various industries, the review supports SDG 12 on responsible consumption and production, SDG 9 on sustainable innovation and industry, and SDG 13 on environmental protection and climate action. Additionally, it emphasizes the importance of scientific research and technological advancements for sustainable development, aligning with the SDGs' call to foster innovation for a more sustainable future.

1.1. Effect of oxidation and degradation of fossil fuels:

Oxidation and degradation have an impact on the traits and issues of fossil fuels. The elderly ones emit greenhouse gases. Performance and stability provide evidence of how well fossil fuels can produce and sustain energy. Degradation and oxidation reduce their efficacy, dependability, longevity, compatibility, and safety **[9, 10]**.

Factors influencing the oxidation of fossil fuels:

Free radicals and reactive oxygen species (ROS) are molecules or atoms that can oxidize fossil fuels by taking their hydrogen atoms or electrons. They can be formed by oxygen, heat, light, microorganisms, or catalysts. Catalysts are substances that speed up the oxidation process by activating oxygen or other molecules, or by generating more radicals from water or hydrogen peroxide **[11, 12]**.

1.2. Gallic acid and its derivatives

Benzene rings with one or more hydroxyl groups attached are chemical compounds that make up the family of "phenolic acids," which includes the naturally occurring substance gallic acid. Gallic acid's chemical formula, "$C_6H_2(OH)_3COOH$", reflects the presence of three hydroxyl groups and one carboxylic acid group. Gallic acid and its derivatives exhibit a variety of "biological activities", including anti-inflammatory, anti-cancer, anti-microbial, and antioxidant properties. Various industrial applications, including those in food additives, preservatives, cosmetics, colors, and pharmaceuticals, are also possible for them **[13-20]**. **Table 1** showed the various industrial applications of gallic acid and its derivatives. **Figure 1** displays the percentage of applications for various gallic acid derivatives. Esters of gallic acid (green color) are used most frequently and have 7 applications (23.3%) in a variety of industries, including the food preservative, pharmaceutical, cosmetic, and chemical industries. The second most frequent use is for gallic acid itself (red color), which has six applications (20%) in a variety of industries including antioxidant, anticancer, antibacterial, antiulcer, anti-cholesterol, and drug metabolizing enzyme inhibitor. Ionic gallate (blue color) is used in two (6.7%) applications, including one as a carbonic anhydrase inhibitor and one as a

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chondro-protective action. There is just one application (3.3%) for each of the remaining gallic acid derivatives across all industries (Grey for Gallic acid and its catechin derivatives have one application as drug metabolizing enzyme inhibitor; Brown for Gallic acid and its ester derivatives have one application as anti-carcinogenic.; Yellow for Gallic acid and its ester derivatives have one application as anti-mutagenic.; Magenta for Gallic acid and its ester derivatives have one application as antiangiogenic. Silver color for gallic acid and its ester derivatives have one application as fuel additive. Cyan color for gallic acid and its ester derivatives have one application as anti-inflammatory.

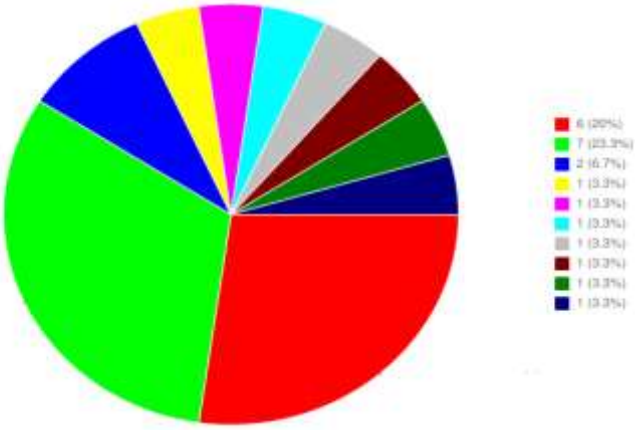


Figure 1. Applications of gallic acid and its derivatives.

Table 1. Applications of gallic acid and its derivatives.

#	Application	Gallic acid derivative	Source
1	Antioxidant	Gallic acid &Esters of gallic acid	[21-24]
2	Anticancer	Gallic acid	[21]
3	Antifungal	Gallic acid	[21]
4	Antibacterial	Gallic acid	[21]
5	Antiviral	Gallic acid	[21]
6	Antiulcer	Gallic acid	[21]
7	Anti-cholesterol	Gallic acid	[21]
8	Anticarcinogenic	Gallic acid Esters of gallic acid	[25, 26]
9	Antimutagenic	Gallic acid & Esters of gallic acid	[25, 26]
10	Antiangiogenic	Gallic acid & Esters of gallic acid	[25, 26]
11	Antiinflammatory	Gallic acid & Esters of gallic acid	[25, 26]
12	Drug metabolizing enzyme inhibitor	Gallic acid and its catechin derivatives	[22]
13	Food preservative	Esters of gallic acid	[22]
14	Cosmetic ingredient	Esters of gallic acid	[22]
15	Pharmaceutical agent	Esters of gallic acid	[22]
16	Chondro-protective effect	Ionic gallate	[23]
17	Carbonic anhydrase inhibitor	Ionic gallate	[23]
18	Antidiabetic activity	Ionic gallate	[23]
19	Cathepsin D inhibitor	Ionic gallate	[23]
20	Antimicrobial	Gallic acid and its ester derivatives	[25, 26]
21	Antifungal	Gallic acid and its ester derivatives	[25, 26]
22	Biofuel production	Gallic acid	[27]
23	Fuel additive	Esters of gallic acid	[21,23]
24	Lubricity enhancer	Gallic acid and its esters	[24]

1.2.1. Extraction Techniques for gallic acid:

There are numerous ways to extract gallic acid from natural sources, including maceration, percolation, soxhlet extraction, ultrasound-assisted extraction (UAE), microwave-assisted extraction (MAE), supercritical fluid extraction (SFE), enzyme-assisted extraction (EAE), etc. Additionally, pyrogallol and phloroglucinol can be used as starting materials for the chemical or enzymatic synthesis of gallic acid from other molecules. Utilizing a variety of processes including esterification, polymerization, condensation, complexation, etc., gallic acid derivatives can be made from gallic acid or other precursors. Recrystallization, column chromatography, fractional distillation, and other processes can be used to purify gallic acid and its derivatives. Methods like spectroscopy (UV-Vis, IR, NMR) can be used to characterize gallic acid and its derivatives [28-31].

1.3. Gallic acid occurrence, chemical, physical and biological activity

Occurrence:

Gallic acid is a naturally occurring substance that can be found in a variety of fruits, plants, and nuts. Gallnuts, sumac, witch hazel, tea, oak bark, grapes, berries, pomegranate, and mango are a few typical sources of gallic acid. Microbes like fungus and bacteria are also capable of producing gallic acid. Gallic acid derivatives can be made synthetically or from natural sources, such as gallic acid or other precursors [14, 32, and 33].

Gallic acid's chemical formula is $C_6H_2(OH)_3COOH$, and its molecular weight is 170.12 g/mol. The benzene ring has a carboxylic acid group at position 1, three hydroxyl groups at positions 3, 4, and 5, and three hydroxyl groups at positions 3, 4, and 5. Its four tautomeric forms are keto-enol (KE), enol (E), quinone-hydroquinone (QH), and semiquinone (SQ). The kind and location of the substituents on the carboxylic acid group or the benzene ring determine the structure of the derivative of gallic acid. Typical types of gallic acid derivatives include gallates (esters), gallotannins (polymers), ellagitannins (dimers), and metal complexes [34].

Chemical properties: Gallic acid is an acidic, insoluble, white or yellowish substance. It has a strong potential as an antioxidant and can create many compounds with various properties. Examples of derivatives of gallic acid include gallates, ellagitannins, metal complexes, and galllotannins [35]. **Scheme 1** indicates the gallic acid and its derivatives.

The biological activity of gallic acid and its derivatives include anti-inflammatory, anti-microbial, antiviral, anti-cancer, and neuroprotective properties. They are capable of modifying several signaling pathways, removing free radicals, chelating metals, and controlling gene expression. Additionally, they may find use in the culinary, cosmetics, and pharmaceutical industries [23, 31, 36- 38].

1.2. Reactions of gallic acid

By esterification, methylation, or other changes, gallic acid can produce a number of derivatives. Examples include:

- **Esterification:** Gallic acid can be esterified to create methyl gallate, ethyl gallate, propyl gallate, octyl gallate, lauryl gallate, and other esters by reacting with alcohols or other organic acids. These esters can be employed as antioxidants in food, cosmetics, medicines, and fossil fuels since they are more stable and soluble than gallic acid [39- 42].

-**Methylation:** Gallic acid can be methylated to create derivatives like 3-methoxygallic acid, 4-methoxygallic acid, 5-methoxygallic acid, 3,4-dimethoxygallic acid, 3,5-dimethoxygallic acid, 4,5-dimethoxygallic acid, etc. by reacting with methylating chemicals like dimethyl sulfate or methyl iodide. As intermediates in the synthesis of other chemicals, these derivatives can have diverse pharmacological and antioxidant properties from gallic acid [13, 43, and 44].

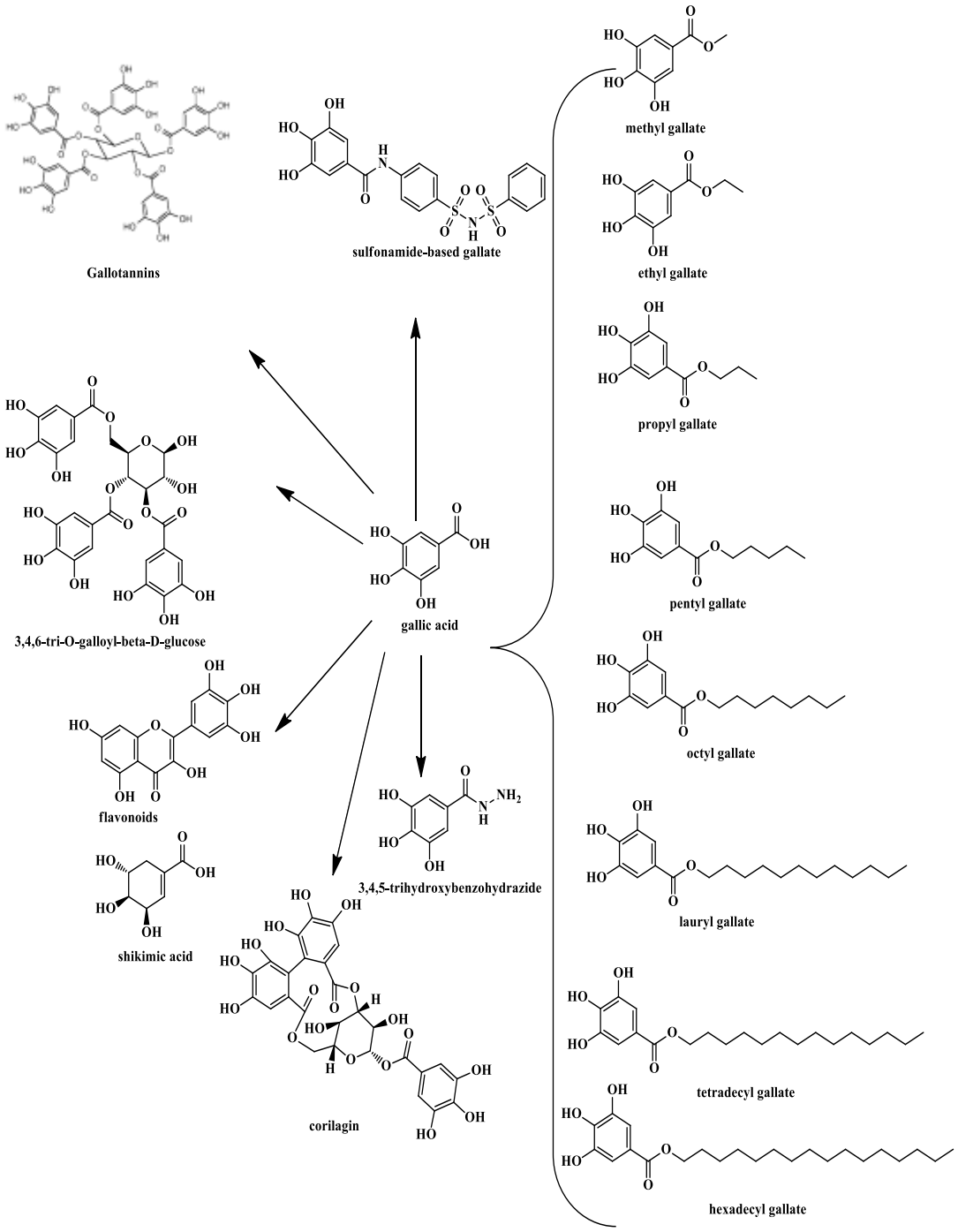
- **Oxidation:** Gallic acid can be oxidized by a variety of oxidizing substances, including oxygen, hydrogen peroxide, potassium permanganate, etc. to generate oxidized derivatives such gallic acid hydrazide, gallic aldehyde, pyrogallol, etc. These derivatives can function as intermediates in the synthesis of other substances and can have chemical and biological properties that are distinct from those of gallic acid [45, 46].

- **Reduction:** Gallic acid can be reduced by a variety of reducing agents, including sodium borohydride, sodium dithionite, etc. to generate reduced derivatives such 3-deoxygallic acid, 4-deoxygallic acid, 5-deoxygallic acid, etc. These derivatives can function as intermediates in the synthesis of other substances and can have chemical and biological properties that are distinct from those of gallic acid [45-48].

- **Hydrolysis:** Acids or water can hydrolyze gallic acid to create hydrolyzed derivatives such 3-hydroxybenzoic acid, 4-hydroxybenzoic acid, 5-hydroxybenzoic acid, etc. When manufacturing other chemicals, these derivatives can be utilized as intermediates because they can have different chemical and biological properties from gallic acid [49, 50].

- **Condensation:** Gallic acid can condense with other substances like formaldehyde, acetaldehyde, phenols, etc. to create condensed derivatives such ellagic acid, gallotannins, ellagitannins, etc. These derivatives can be employed as natural products or antioxidants and have molecular weights that are higher than gallic acid and antioxidant activity [50, 52].

- **Complexation:** Gallic acid can combination with metal ions like iron, copper, zinc, etc. to create gallic acid metal complexes. These complexes can be utilized as dyes or catalysts and can differ from gallic acid in terms of color and catalytic activity [44, 53-55].



Scheme 1. Gallic acid and its derivatives.

Gallic acid can form different types of derivatives by modifying its hydroxyl or carboxylic acid groups. Some common types of gallic acid derivatives are:

Gallates: are esters of gallic acid with phenols or alcohols. For instance, methyl gallate, propyl gallate, octyl gallate, and dodecyl gallate are aliphatic alcohol-based esters of gallic acid. Catechins are flavonoids that can be found in tea and other plants, and catechin gallates are esters of gallic acid with catechins [56-60].

Gallotannins: They are polymer component of gallic acid, which also contain glucose or other sugars. They fall under the category of hydrolyzable tannins, which can be hydrolyzed enzymatically or in an acidic environment to produce gallic acid and sugars. They are abundant in plants and possess astringent and antioxidant qualities [61-65].

Ellagitannins: A class of hydrolyzable tannins that can be converted into ellagic acid and sugars through acidic or enzymatic hydrolysis. Ellagic acid is a dimer of gallic acid, linked by a carbon-carbon bond and two lactone rings. Found widely in plants, ellagitannins have demonstrated antioxidant and anticancer activities. [65-70].
Metal complexes: They are substances created when gallic acid or its derivatives combine with metal ions like iron, copper, zinc, or silver. They are used as catalysts, antibacterial agents, dyes, and in photography [71-75].

1. Fossil fuels antioxidants
Antioxidants can stop or delay the oxidation of other molecules. During oxidation, free radicals are produced as a result of electron loss. Free radicals are highly reactive and unstable atoms or molecules. They can damage the chemical structure and function of lipids, proteins, DNA, and other biomolecules [76 – 79].
Fossil fuels are hydrocarbons formed from prehistoric organic materials and are commonly used as energy sources for heating, lighting, transportation, and industrial activities. However, when exposed to air, heat, light, or metal catalysts, fossil fuels undergo oxidation. This process can produce peroxides, aldehydes, ketones, acids, and other compounds that can reduce the fuel's quality and efficiency. Additionally, these byproducts may increase the emissions of harmful substances such as carbon monoxide, hydrocarbons, nitrogen oxides, particulate matter, and smoke [80-84].
Antioxidants are therefore crucial and pertinent for use in fossil fuels since they can:
1. Prevent the creation of hazardous chemicals that could damage the stability, storage, transport, and combustion of fossil fuels by inhibiting or delaying the oxidation of those fuels [85-89].
2. Enhance engine performance and fuel efficiency by lowering fuel use, friction, wear, corrosion, and deposits [90-94].
3. Lower the emissions of greenhouse gases and other harmful substances that might cause cancer, acid rain, pollution, respiratory illnesses, and global warming in order to lessen their negative effects on the environment and health concerns [95-97].

2.1. Applications of gallic acid and its derivatives antioxidants:
Gallic acid and its derivatives can act as antioxidants for industrial fluids, heating systems, and fossil fuels. They can prevent or slow down oxidation and the formation of harmful chemicals. They can also improve the quality, performance, efficiency, and safety of these items. They can also reduce their consumption and emissions, and protect the environment and human health [98].

2.1.1. Mechanism of using gallic acid as antioxidant.
Gallic acid and its derivatives can act as antioxidants for fossil fuels by scavenging free radicals produced during the burning of hydrocarbons, such as hydroxyl radicals (•OH), peroxyl radicals (ROO•), and alkoxyl radicals (RO•). They can also stop lipid peroxidation, a process that starts a chain reaction that produces additional free radicals and dangerous substances. Additionally, they have the ability to bind metal ions that catalyze oxidation reactions, such iron and copper. In order to prevent oxidative damage to the fuel, they can also regulate endogenous antioxidant enzymes like superoxide dismutase (SOD), catalase (CAT), and glutathione peroxidase (GPx) [13, 98].
Gallic acid and its derivatives can reduce the oxidation of fossil fuels and the emissions of pollutants by: Preventing or delaying the development of peroxides, aldehydes, ketones, acids, and other chemicals that can reduce the quality and efficiency of the fuel. By lowering fuel consumption, friction, wear, corrosion, and deposits, one can increase engine performance and fuel economy. Decreasing the emissions of harmful compounds such carbon monoxide (CO), hydrocarbons (HC), nitrogen oxides (NOx), sulfur dioxide (SO2), particulate matter (PM), and smoke, as well as greenhouse gases like carbon dioxide (CO_2) and methane (CH_4) [13].

2.1.2. Forms of antioxidants in fossil fuel:
Phenolic anti-oxidants: These are substances that include a benzene ring together with one or more hydroxyl groups. By giving hydrogen atoms or electrons, they are able to scavenge free radicals. The metal ions that can catalyze oxidation reactions can also be chelated by them. Tert-butylhydroquinone (TBHQ), propyl gallate (PG), butylated hydroxytoluene (BHT), butylated hydroxyanisole (BHA), gallic acid (GA), and others are phenolic antioxidants [99, and 100].
Amine antioxidants: Antioxidants with one or more amino groups connected to an aromatic or aliphatic chain are known as amines. By contributing hydrogen atoms or electrons, they can neutralize free radicals. They have the ability to bind metal ions and prevent them from catalyzing. Diphenylamine (DPA), phenylenediamine (PPD), di-tert-butyl-p-cresol (DTBC), and N,N'-di-sec-butyl-p-phenylenediamine (DBPC) are a few examples of amine antioxidants [101, and 102].
Vitamin antioxidants: These are substances with structures and actions resembling those of vitamins. By giving hydrogen atoms or electrons, they are able to scavenge free radicals. In addition, by lowering other antioxidants back to their active states, they can renew them. Vitamins that act as antioxidants include beta-carotene (provitamin A), ascorbyl palmitate (vitamin C ester), and alpha-tocopherol (vitamin E) [103; 104]
Gallic acid and its derivatives have been shown in several experimental experiments to have antioxidant properties in a variety of fossil fuels, including gasoline, diesel, biodiesel, jet fuel, etc. Some examples include:
Sutanto and Nasikin [105] investigated the solubility and antioxidant properties of a pyrogallol derivative as a potential additive for biodiesel. The researchers prepared the derivative by reacting pyrogallol with methyl linoleate in the presence of a radical, resulting in a molecule with increased solubility. The solubility test and various tests for antioxidant potential were conducted, including acid value determination and the Rancimat test. The results showed that the pyrogallol derivative exhibited higher solubility and acid value stability in palm oil biodiesel compared to pyrogallol, tert-butylhydroquinone (TBHQ), and gallic acid. Additionally, the derivative demonstrated a higher Rancimat induction time, indicating superior performance under accelerated

oxidation conditions. These findings suggest the potential of the pyrogallol derivative as an effective antioxidant additive for biodiesel.

Varatharajan & Pushparani [106] addresses the issue of maintaining the stability of biodiesel fuels over an extended period. Biodiesel is susceptible to oxidation, resulting in the formation of insoluble gums that can clog fuel filters. The article highlights the importance of incorporating appropriate antioxidants to improve the storage stability of biodiesel. The selection of antioxidants depends on factors such as the chemistry of antioxidants, the composition of biodiesel, the presence of transition metals, and storage temperature. The study emphasizes the need for further research to identify effective antioxidants for different types of biodiesel fuels.

Longanesi et al [107] discussed the factors promoting oxidation, including molecular composition, metal contamination, temperature, and light exposure. It also explores the benefits of hydrogenated vegetable oil (HVO) and the analytical techniques used to study oxidation. Additionally, they addressed the influence of higher pressure injection systems on deposit formation, emphasizing that unsaturated biodiesel components are not solely responsible.

2.2. Factors that influence the antioxidant efficiency of gallic acid and its derivatives in fossil fuels

Gallic acid and its derivatives are natural compounds that can prevent or delay the oxidation of fossil fuels by free radicals and ROS. Their antioxidant performance depends on many factors that influence their characteristics and behavior in the fuel systems. They have benefits and drawbacks as antioxidants for fossil fuels, and they need to be optimized and researched to enhance their effectiveness and sustainability. [21, 23, 98, 108-115].

3. Determination of the antioxidant activity of gallic acid and its derivatives in fossil fuels:

There are some methods for measuring the antioxidant activity of gallic acid and its derivatives in fossil fuels, such as:

- **Induction period:** The induction period is the amount of time needed for free radicals and ROS to start oxidizing fossil fuels. Different methods, including differential scanning calorimetry (DSC), pressure differential scanning calorimetry (PDSC), and the Rancimat method [116, 117]; can be used to determine the induction duration. By delaying or inhibiting the oxidation of fossil fuels, the induction duration may reflect the antioxidant activity of gallic acid and its derivatives.
- **Gum content:** The quantity of sludge or insoluble deposits created during the oxidation of fossil fuels by free radicals and ROS is known as the gum content. Several methods [118, 119] like ASTM D381 [120], ASTM D873 [121], or IP 131 [122] can be used to determine the gum content. By lowering or limiting the formation of deposits or sludge, the gum content can serve as a proxy for the antioxidant activity of gallic acid and its derivatives.
- **Oxidation stability:** The ability of fossil fuels to withstand oxidation by free radicals and reactive oxygen species (ROS). Different methods, such as ASTM D2274 [123], ASTM D525 [124], ASTM D2272 [125], or EN 14112 [126], can be used to determine the oxidation stability. By improving or maintaining the quality and performance of fossil fuels, the oxidation stability can represent the antioxidant activity of gallic acid and its derivatives.
- **Acid value:** The amount of acid produced as a result of free radicals and reactive oxygen species oxidizing fossil fuels is known as the acid value. There are numerous methods that can be used to determine the acid value, including ASTM D664 [127], ASTM D974 [128], and IP 139 [129]. Gallic acid and its derivatives have antioxidant properties that can be reflected in the acid value by reducing or preventing the production of acid.
- **Peroxide value:** The peroxide value is the quantity of peroxides produced when free radicals and ROS oxidize fossil fuels. Different methods, such as ASTM D3703 [130], ASTM D2344 [131] or ISO 3960 [132], can be used to determine the peroxide value. By reducing or preventing the generation of peroxides, the peroxide value can indicate the antioxidant activity of gallic acid and its derivatives**.**

Accelerated oxidation tests: are techniques that can quickly and practically measure the antioxidant effectiveness and the quality and performance of fossil fuels under real oxidation and degradation conditions. The disadvantage of these techniques is that they may not be consistent, reproducible, or comparable across different laboratories or systems. They may also be influenced or interfered by various factors such as temperature, pressure, oxygen concentration, catalysts, etc.

3.1. Techniques for describing composition, characterization, and behavior of gallic acid and its derivatives in fossil fuels:

3.1.1. Differential scanning calorimetry (DSC):

DSC can be used to quantify the oxidation and decomposition of fossil fuels by looking for exothermic peaks that signify the beginning and growth of oxidation reactions [133]. Thermodynamics, antioxidants, and the kinetics of fossil fuels can all be studied using DSC. Although DSC is sensitive, accurate, and illuminating, it is also pricey, difficult, and demands several samples as well as pricy equipment. Various elements that affect the outcomes may also have an impact.

3.1.2. Gas chromatography (GC):

GC is a technique that separates and analyzes a sample by its volatility and polarity. It can measure the oxidation and degradation of fossil fuels by identifying and quantifying their components and changes. GC is selective, accurate, and informative, but it is also time-consuming, requires sample preparation and suitable column and detector. It could also be affected by various factors that could cause peak problems [134-136].

3.1.3. Mass spectrometry (MS):

- MS is a method that ionizes a sample and analyzes each of its constituent parts according to their mass-to-charge ratio. By identifying and quantifying their chemical fragments and isotopes, it can gauge the oxidation and breakdown of fossil fuels.

Although MS is sensitive, accurate, and illuminating, it is also costly, difficult, and labor-intensive and necessitates sample preparation and appropriate ionization and detection techniques [136-138].

3.1.4. The Molecular Docking of Gallic Acid and Its Derivatives

Gallic acid and its derivatives exhibit various biological functions, including anti-inflammatory, anti-cancer, antioxidant, and antibacterial effects. Molecular docking, a computational technique, was used to predict the binding affinity of ligands to target proteins. In this study, molecular docking was performed using AutoDock Vina, with crystal structures of target proteins obtained from the Protein Data Bank (PDB). Energy minimization was carried out using Gaussian 09, and ligands were prepared with ChemDraw. Binding affinity was calculated using the inhibition constant (Ki) and binding energy (ΔG).

The outcomes demonstrated that gallic acid and its derivatives had strong binding affinity for the intended proteins. With a ΔG value of -8.1 kcal/mol and a Ki value of 0.13 μM, gallic acid had the highest binding affinity towards cyclooxygenase-2 (COX-2). The derivatives likewise demonstrated strong binding affinity for COX-2, with G values between -7.5 and -8.0 kcal/mol.

Gallic acid is a type of phenolic acid that is present in a wide variety of plants, including gallnuts, sumac, witch hazel, tea leaves, oak bark, etc. It contains antioxidant qualities and might also be analgesic, anti-inflammatory, antibacterial, anti-cancer, and have other health advantages.

Gallic acid and its derivatives can be evaluated using a variety of computational techniques, including as molecular docking, molecular dynamics simulations, quantum mechanics calculations, and density functional theory (DFT) studies.

3.1.4.1. Molecular docking:

Arsianti et al [139] presents the design and screening of gallic acid derivatives as potential inhibitors of malarial dihydrofolate reductase (DHFR) using in silico docking. The researchers designed fourteen gallic acid derivatives and modeled their three-dimensional structures. Through docking simulations and amino acid analysis, they identified three top-ranked compounds, with compound 12 (octyl gallate) showing the strongest interaction and greatest inhibitory activity against malarial DHFR. The study suggests that compound 12 holds promise as a potential candidate for the development of new antimalarial agents, Figures 2, 3.

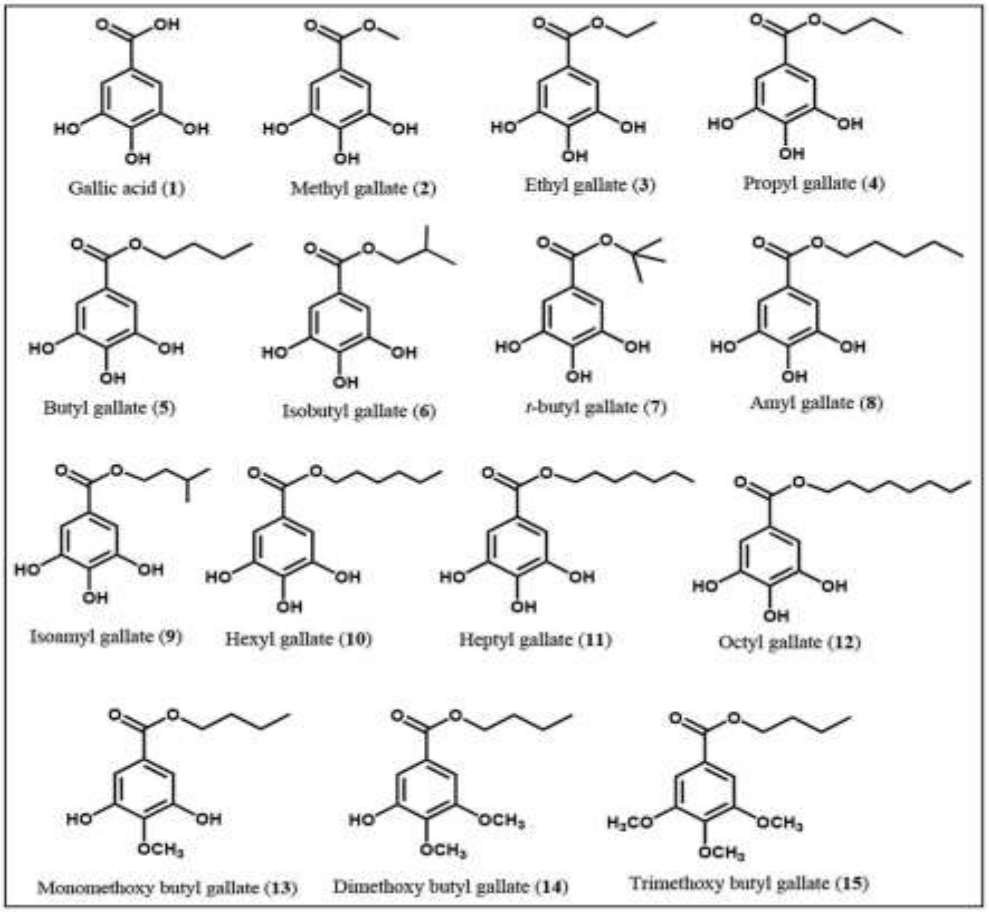


Figure 2 Chemical structure of gallic acid (1) and its derivatives (2-15) [139]

Humaedi et al [140] investigates the potential of gallic acid and its derivatives as inhibitors of BRAF colon cancer through in silico molecular docking. The study examines the stability, affinity, and interaction of gallic acid and five derivatives as ligands with the BRAF protein. The results indicate that the derivatives have lower Gibbs energy values compared to gallic acid, suggesting higher stability. Additionally, the derivatives exhibit greater affinity and stronger interaction with the catalytic site of BRAF colon cancer compared to gallic acid. Among the derivatives, (2-hydroxy)-benzylgallate shows the highest stability and strongest interaction with BRAF, making it a promising candidate for colon cancer drug development, Figure 4.

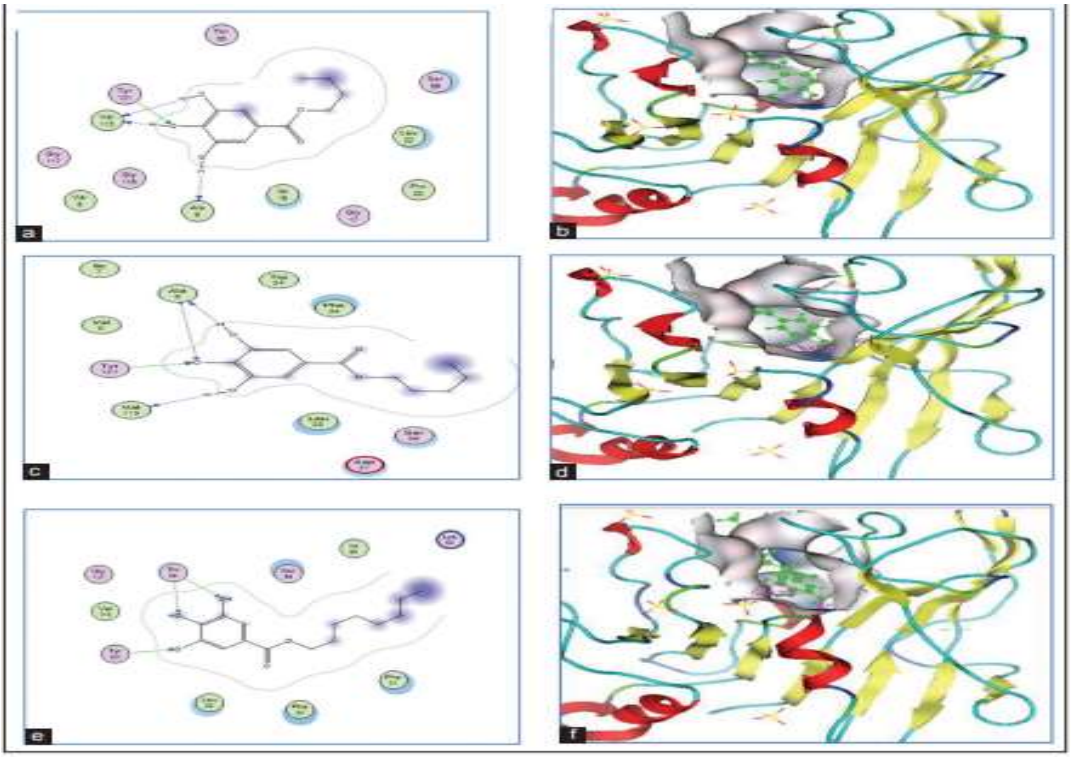


Figure 3 Two-dimensional and three-dimensional complex interaction of derivative 5 (a, b), 8 (c, d), and 12 (e, f) with dihydrofolate reductase [139]

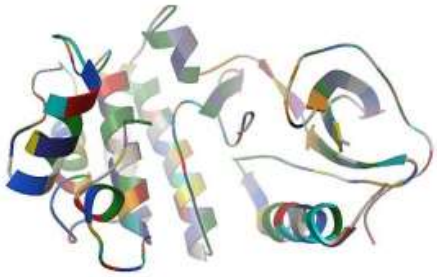


Figure 4 Crystal structure BRAF [140].

Kokila et al [141], explores the antihyperglycemic properties of gallic acid derived from the methanol extract of Thunbergia mysorensis flowers. The study demonstrates that the extract effectively inhibits α-glucosidase, α-amylase, and aldose reductase enzymes, which play a crucial role in postprandial hyperglycemia management. Gallic acid is identified as the primary active component of the extract, and it exhibits significant inhibition of the target enzymes. Molecular simulations reveal stable interactions between gallic acid and the active sites of the enzymes. These findings suggest that gallic acid derived from Thunbergia mysorensis could serve as a potential natural antidiabetic drug, warranting further in vivo and clinical studies.

Raghi et al. [142] explores the inhibitory activity of Gallic acid derivatives fused with 1,3, 4-Oxadiazole moieties against the ABL receptor, which is implicated in Chronic Myeloid Leukemia (CML). The study employs molecular electrostatic potential maps (MESP), molecular docking, and molecular dynamics (MD) simulations to evaluate the structural features and drug likeness of these derivatives. The compounds exhibit promising ABL kinase inhibitory activity and acceptable pharmacokinetic properties, making them potential candidates for the development of new CML drugs.

Umar et al. [143] presents molecular docking studies aimed at identifying potential therapeutic compounds against the non-structural proteins (nsp3, nsp5, nsp12, nsp13, and nsp14) of the novel coronavirus (SARS-CoV-2). The researchers docked 16 derivatives of gallic acid against these proteins and evaluated their binding energies. One derivative, 4-O-(6-galloylglucoside), displayed lower binding energy values than control drugs against multiple target proteins. Pharmacokinetics

screening revealed that this derivative could be metabolized by the liver and has high plasma protein binding. The study suggests that 4-O-(6-galloylglucoside) could be a promising inhibitor against the studied SARS-CoV-2 proteins, but further studies such as molecular dynamic simulation, Figure 5, in vivo, and in vitro experiments are needed to validate these findings.

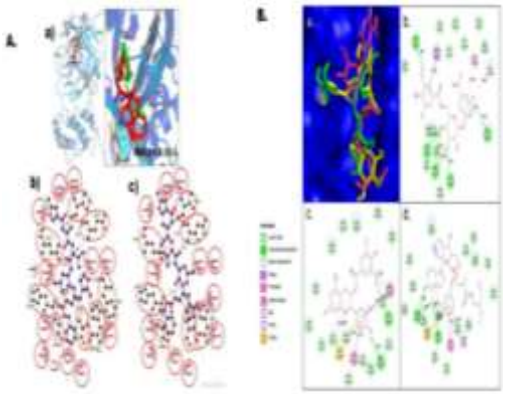


Figure 5 Molecular docking studies of gallic acid derivatives against main protease (Mpro) of SARS-Cov-2. a Protocol validation of molecular docking experiment using AutoDock Vina, PyMOL, and LigPlot+ . (a) Comparison of binding modes for re-docked ligand (red) vs. co-crystallized ligand (green) shown as stick representation. Amino acid residues interaction with (b) co-crystalized and (c) re-docked ligand accomplished in LigPlot+ . b Binding mode and molecular interaction of hit ligands with Mpro. (a) Surface representation of Mpro (PDB: 6 LU7) show the binding mode of docked 3-O-(6-galloylglucoside) (yellow), epicatechin gallate (green), and remdesivir (pink). 2D interaction of (b) 3-O-(6-galloylglucoside), (c) epicatechin gallate, and (d) remdesivir [143].

3.1.4.2. Molecular dynamics simulations:

Zhan et al [144] investigates the binding interaction between gallic acid (GA) and lysozyme (LYS) using a combination of molecular dynamics (MD) simulation and spectroscopic techniques. The results indicate that GA forms a stable complex with LYS, with hydrogen bonding and hydrophobic interactions being the main driving forces. The calculated binding energies suggest that van der Waals forces and electrostatic interactions play a significant role in the interaction, Figure 6. The study provides valuable insights into the pharmacological mechanism of GA and offers a reference for future research in this field.

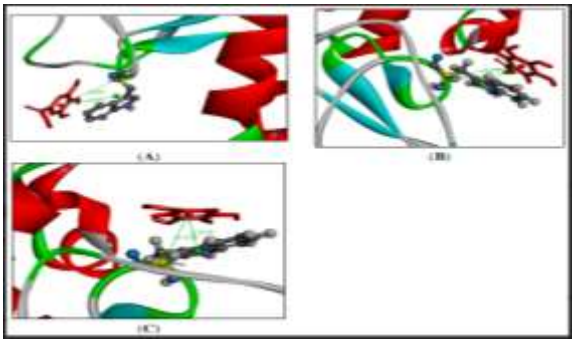


Figure 6 Distance map of GA to Trp62 of LYS, only picked Trp62 as a example. (A) The interaction mode between GA and LYS before MD simulation; (B) The interaction mode between GA and LYS after 298 K MD simulation; (C) The interaction mode between GA and LYS after 310 K MD simulations. (The ligand structure is represented using red stick model, and the red ball is the centroid of GA. The distances are remarked using green lines. The residue Trp62 is represented using a ball and stick model, the green ball represents the centroid of Trp and the yellow ball represents the Cα of Trp) [144].

Nie et al [145] investigates the inhibitory effects of gallate moiety on the formation of Aβ amyloid aggregates, which are associated with Alzheimer's disease. Through molecular dynamics simulations, the study reveals that gallic acid (GA), containing the gallate moiety, effectively prevents the conformational changes of Aβ1–40 monomers and inhibits the formation of β-sheet structures. The binding between GA molecules and Aβ1–40 monomers involves both hydrophilic and hydrophobic amino acid residues. Furthermore, the study demonstrates that polar interactions play a stronger role in the binding than nonpolar interactions. These findings provide molecular-level insights into the mechanisms by which gallate moiety contributes to the anti-amyloidogenic effects of polyphenols, offering potential implications for the development of therapeutic strategies for Alzheimer's disease.

Zhang et al [146] identified natural thrombin inhibitors from traditional Chinese medicine (TCM) and evaluate their biological activity and binding characteristics. Through a combination of molecular docking, thrombin inhibition assay, surface plasmon resonance (SPR), and molecular dynamics simulation, gallic acid was identified as a direct thrombin inhibitor with significant inhibitory effects on thrombin-induced platelet aggregation. The binding studies revealed that gallic acid interacted

with thrombin with a KD value of 8.29 lmol/L. Molecular dynamics and binding free energy analysis provided insights into the mechanism of thrombin inhibition by gallic acid, Figure 7. The study concluded that gallic acid could serve as a potential natural thrombin inhibitor, providing a basis for further research and development in this area.

Cappelli et al. [147], explores the solvation of gallic acid in water and acetonitrile through spectroscopic analysis. The study utilizes classical and quantum mechanical approaches to predict IR, UV, and NMR spectra and compares them with experimental data to validate the solvation models. The effects of hydrogen bonding and bulk solvent on the spectroscopic properties are evaluated. The authors employed a combination of continuum-only, discrete, and mixed continuum/discrete solvation models based on quantum-mechanical and classical molecular dynamics solute-solvent clusters. The study provides insights into the structural characteristics and spectroscopic properties of gallic acid, which can contribute to a better understanding of its biochemical structure-activity relationships and its potential applications in various fields.

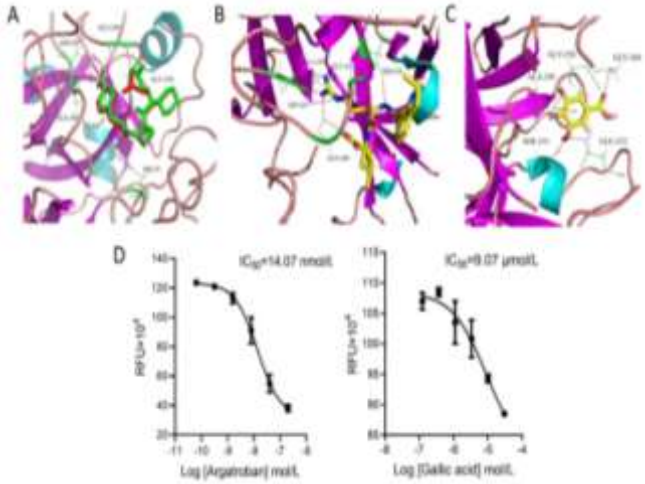


Figure 7 . Binding mode of ligand MEL (A), argatroban (B) and gallic acid (C) in the inhibitor-binding site of thrombin. The green and red sticks represent the re-docked and cocrystallized conformations of ligand MEL, respectively. The red dotted lines represent the hydrogen bond interactions between the ligand and thrombin. Key residues are shown and the red dotted lines represent the hydrogen bond interactions between the inhibitors and thrombin. Yellow, white, red and blue atoms represent carbon, hydrogen, oxygen and nitrogen atoms, respectively. (D) Dose-response curves of thrombin inhibition for gallic acid and argatroban, respectively [146].

4. Quantum mechanics calculations:

Badhani and Kakkar [148] explored the structural and chemical properties of gallic acid using density functional theory (DFT) calculations. The study investigates the intramolecular interactions, determines the pKa values of gallic acid anions, and examines the influence of external factors such as pH and dielectric of the medium on the molecular orbitals and electronic spectra. Additionally, the paper simulates and validates the IR and NMR spectra of gallic acid. The researchers also analyze the global and local reactivities of gallic acid, assessing its susceptibility to nucleophilic, electrophilic, and radical attacks. The study provides insights into the structural and electronic characteristics of gallic acid, which are important for developing pharmacologically potent molecules.

Vahedi et al, [149] investigated the interaction mechanism between gallic acid (GA) and α-Chymotrypsin (α-CT) using spectroscopic methods, computational docking, and molecular dynamics (MD) simulation. The results show the formation of a stable complex between GA and α-CT, with fluorescence spectra analysis indicating a static quenching mechanism. The binding constants suggest moderate affinity between GA and α-CT. CD findings reveal alterations in the protein's secondary structure upon interaction with GA. Enzyme activity assays demonstrate a significant decrease in α-CT's activity in the presence of GA, indicating its role as an effective inhibitor. Molecular docking and MD simulations provide insights into the optimal binding site and stability of the α-CT-GA complex.

Pardeshi et al, [150] presents a comprehensive investigation on the molecular interactions and spectroscopic properties of Gallic acid (GA) and acrylic acid (AA) in Gallic acid imprinted polymers (MIPs). The study utilizes density functional theory (DFT) calculations to optimize the structures of GA, AA, and their complex, and examines the effect of the porogen acetonitrile (ACN) using the polarizable continuum model (PCM). The results reveal the formation of a stable GA-AA complex through intermolecular hydrogen bonding. The vibrational spectra simulations are compared with experimental FT-IR spectra, demonstrating the accuracy of the computational approach. Moreover, the study determines the optimal GA-AA mole ratio for the synthesis of MIPs for GA.

Guendouzi, et al, [151] investigated the formation of an inclusion complex between gallic acid (GA) and β-cyclodextrin (β-CD) at a 1:1 stoichiometry ratio. β-CD is known for its ability to encapsulate bioactive compounds, providing protection and improved solubility. The study aims to understand the complexation mechanism and the capability of β-CD to encapsulate GA in both gas and solution phases. Different quantum mechanical methods, including HF/6-31G* and density functional theory (DFT) with dispersion correction, were compared to evaluate the importance of dispersion forces and hydrogen bonding in the interaction. The stability of the optimized complex geometries was assessed using the super molecule method. Various techniques such as frontier molecular orbital (FMO) theory, global indices of reactivity, condensed natural bond orbital

(NBO) analysis, and molecular docking were employed to confirm the inclusion complex and examine the nature of hydrophobic interactions during the complexation process.

Ghouari, et al [152] investigated a new polymorph of gallic acid monohydrate (GAM-VII) for its potential application in the pharmaceutical industry. The synthesis, crystal structure, and computational quantum investigations of GAM-VII were presented. The structural analysis reveals that the presence of water molecules and moderate intermolecular interactions involving carboxyl groups contribute to the formation and stability of this polymorph. A comprehensive comparison is made with the other six known GAM polymorphs, focusing on molecular conformations, hydrogen bonding, and intermolecular interactions. The Hirshfeld surface method is employed to analyze the supramolecular assemblies and quantify the contributions of different intermolecular interactions. The study also provides insights into the reactivity process of GAM-VII through theoretical calculations, including dipole moment, ionization, chemical potential, electronegativity, and electrophilicity index.

Zhan et al. [153] conducted molecular dynamics simulations on the GA-LYS complex to study its binding mechanism. They found that temperature affects GA
binding to LYS, with less protein flexibility at lower temperatures. Molecular docking confirmed the interaction was driven by hydrogen bonding, van der Waals forces, and hydrophobic interactions, consistent with spectroscopy results. Using the MM-PBSA method, they showed the complex remained stable during the simulations.
Additionally, they observed that the binding strength of GA to LYS weakened at higher temperatures(310K) compared to 298K.

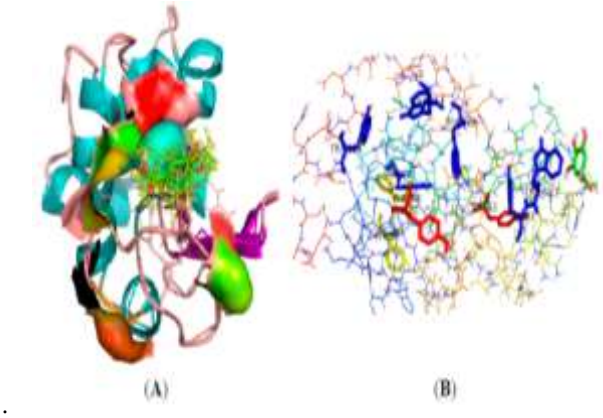


Figure 8 Interaction of GA with LYS. (A) Electron density and hydrophobic surface map between GA- LYS, only residues around 20.0 Å of the ligand are displayed; (B) Interaction mode between GA and LYS. The ligand structures are represented using a stick model, the blue sticks represented Trp residues, the red sticks represented Tyr residues, the yellow sticks represented Phe residues [153].

5. Advantages and disadvantages of using gallic acid and its derivatives as antioxidants in fossil fuels

It is possible to examine and contrast the following benefits and drawbacks of employing gallic acid and its derivatives as antioxidants in fossil fuels:
The benefits are Free radicals and reactive oxygen species (ROS) can oxidize and degrade fossil fuels, whereas gallic acid and its derivatives are organic, biodegradable, and renewable sources of antioxidants. In addition to chelating metal ions and modulating cellular signaling pathways, they can scavenge free radicals, prevent lipid peroxidation, and limit combustion of lipids. By extending the induction period, oxidation stability, viscosity, density, etc. of fossil fuels, they can also enhance their quality and efficiency. They may also possess a variety of biological and pharmacological properties, including those that are anticancer, antifungal, antibacterial, antiviral, antiulcer, and anticholesterol, among others.
Gallic acid's and its derivatives' low solubility, stability, and availability in fossil fuels are disadvantages. In the interfacial region of the emulsions, they may be quickly oxidized or destroyed by oxygen, light, heat, metal ions, or microorganisms. Depending on their dosage, structure, and surrounding circumstances, they may also exhibit a dual function as an antioxidant and prooxidant. Additionally, they might have negative consequences like cytotoxicity, genotoxicity, allergenicity, or disruption of other molecules or enzymes.

6. Potential uses and effects of gallic acid and its derivatives' as antioxidants in numerous fields:

Advantages of using gallic acid

Gallic acid and its derivatives are antioxidants that can improve the quality, performance, and longevity of fossil fuels and the equipment that uses them. They can prevent or reduce the harmful effects of oxidation and other chemical reactions in fossil fuels, as indicated in the following paragraphs:
- **Transportation:** By enhancing the fluidity, flow-ability, lubricity, etc. of fossil fuels, gallic acid and its derivatives might enhance their transportation. Additionally, they can guard against the corrosion, fouling, clogging, etc. that may happen as a result of the oxidation and degradation of fossil fuels in transportation pipelines and tanks. They can also prevent the production of hazardous substances like aldehydes, ketones, esters, ethers, and other gases from occurring when fossil fuels are burned.
- **Environment:** By lowering the environmental effect and reliance on synthetic antioxidants that may be hazardous, non-biodegradable, or non-renewable, gallic acid and its derivatives can enhance the environment. Additionally, they can lessen the

harm that fossil fuel oxidation and degradation may do to the environment by releasing noxious substances into the air, water, or soil, such as acids, peroxides, hydroxyl radicals, and other hazardous substances. Additionally, they may encourage the use of antioxidants derived from various plant sources or manufactured using sustainable and renewable methods.

- **Health:** By lowering the risks for illnesses and conditions brought on by contact with or consumption of oxidized and degraded fossil fuels, which may have cytotoxic, genotoxic, allergic, or carcinogenic effects, gallic acid and its derivatives can improve health. Additionally, they can have a number of biological and pharmacological advantages, including, but not limited to, anticancer, antifungal, antibacterial, antiviral, antiulcer, and anti-cholesterol properties.

Limitations and potential drawbacks of using gallic acid

Limitations and potential drawbacks of using gallic acid and its derivatives as antioxidants may vary depending on the context and application, here are some general considerations:

Stability and Shelf Life: Gallic acid and its derivatives may exhibit limitations in terms of stability, especially under certain environmental conditions such as exposure to light, heat, or pH variations. This can affect their efficacy as antioxidants and their shelf life in practical applications.

Compatibility and Interactions: Compatibility issues may arise when incorporating gallic acid and its derivatives into different products or systems. They may interact with other components or ingredients, leading to changes in taste, color, or stability. Understanding the compatibility and potential interactions is crucial for successful application.

Extraction and Production Challenges: Obtaining gallic acid and its derivatives from natural sources or synthesizing them can present challenges. Extraction methods may require specific solvents or conditions, and large-scale production might be costly or complex. These factors can impact the availability and cost-effectiveness of these antioxidants.

Sensitivity to Processing Conditions: Gallic acid and its derivatives may be sensitive to processing conditions, such as high temperatures or certain chemical reactions. This sensitivity can result in the degradation or alteration of their antioxidant properties, limiting their effectiveness in specific applications or processing techniques.

Regulatory Considerations: Depending on the industry and application, there may be regulatory considerations or restrictions on the use of gallic acid and its derivatives as antioxidants. Compliance with regulations related to safety, labeling, dosage, and maximum allowable concentrations may be necessary.

Potential Interference with Other Processes: While gallic acid and its derivatives exhibit antioxidant properties, they may also interfere with other processes or reactions. For example, in certain industrial processes or biological systems, they might impact enzymatic reactions or interfere with desired chemical transformations.

7. Gallic acid and its derivatives' potential as antioxidants in fossil fuels: Opportunities and challenges

Fossil fuels, such as gasoline and diesel, are prone to oxidative degradation, leading to the formation of harmful byproducts and reduced fuel quality. Gallic acid, a naturally occurring phenolic compound, possesses strong antioxidant properties due to its ability to scavenge free radicals and inhibit oxidation reactions. Its derivatives, including esters and polymers, can be tailored to enhance stability and solubility in fuel matrices. These antioxidants have demonstrated promising results in mitigating fuel degradation and improving fuel performance, such as reducing deposit formation, minimizing corrosion, and extending fuel shelf life. However, challenges remain, including the optimization of antioxidant dosage, compatibility with different fuel compositions, and potential side effects on combustion efficiency and emissions. Additionally, the interactions between gallic acid derivatives and other fuel additives need to be thoroughly investigated. Despite these challenges, gallic acid and its derivatives offer a promising avenue for developing effective antioxidant strategies in fossil fuels, contributing to the overall sustainability and longevity of fuel systems.

8. New Directions for further research of gallic acid and its derivatives as antioxidants in fossil fuels

More research is needed on gallic acid and its derivatives' antioxidant properties in fossil fuels in:

Improving their characteristics: improving their solubility, stability, and availability by using different formulations and optimizing the pH and the ratios of the components.

Characterizing them: determining their chemical composition and behavior and studying their role in preventing oxidation and degradation by using advanced methods.

Evaluating them: measuring their antioxidant activity and effectiveness based on various factors and comparing the results of different systems and methods.

Optimizing them: developing models for their distribution, concentration, and structure by using computer-based methods and predicting and controlling their antioxidant and pro-oxidant effects when they interact with other molecules or enzymes.

9. List of Abbreviations:

Abbreviation	Full Form
•OH	Hydroxyl Radicals
ASTM	American Society for Testing and Materials
BHA	Butylated Hydroxyanisole
BHT	Butylated Hydroxytoluene
CAT	Catalase
CD	Cyclodextrin
CH4	Methane
CO	Carbon Monoxide
CO_2	Carbon Dioxide
DBPC	N,N'-Di-sec-butyl-p-phenylenediamine
DFT	Density Functional Theory
DG	Dodecyl Gallate
DPA	Diphenylamine
DSC	Differential Scanning Calorimetry
DTBC	Di-tert-butyl-p-cresol
E	Enol
EAE	Enzyme-assisted Extraction
EG	Ethyl Gallate
EN	European Patent Specification
GA	Gallic Acid
GADs	Gallic Acid Derivatives
GC	Gas Chromatography
GPx	Glutathione Peroxidase
HC	Hydrocarbons
hCA IX	Human Carbonic Anhydrase IX
HAS	Human Serum Albumin
IP	Institute of Petroleum Test Methods
IR	Infrared
ISO	International Organization for Standardization
KE	Keto-enol
Ki	Inhibition Constant
MAE	Microwave-assisted Extraction
MG	Methyl Gallate
MS	Mass Spectrometry
NMR	Nuclear Magnetic Resonance
NOx	Nitrogen Oxides
OG	Octyl Gallate
PDB	Protein Data Bank
PDSC	Pressure Differential Scanning Calorimetry
PG	Propyl Gallate
PM	Particulate Matter
PPD	Phenylenediamine
QH	Quinone-hydroquinone
RMSD	Root Mean Square Deviation
RMSF	Root Mean Square Fluctuation
RO•	Alkoxyl Radicals

10. Conclusions:

Gallic acid and its derivatives, with their potent antioxidant properties and ability to prevent the oxidative degradation of fossil fuels, present promising candidates for use as antioxidants in fossil fuel preservation. While their synthesis, formulation, characterization, assessment, and optimization can still be improved, there are significant opportunities for advancement in this field. This study aims to stimulate further interest and innovation by providing a comprehensive review of the current knowledge and identifying research gaps regarding the use of gallic acid and its derivatives as antioxidants in fossil fuels.

11. Conflicts of interest

The authors have no conflict of interest.

12. Formatting of funding sources

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