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Nature-inspired nano-additives for Biofuel application – A Review

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ABSTRACT

The increasing demand and cost of conventional fuels have forced humanity to consider various alternative fuels. Low yield and massive cost are significant limitations in biofuel production. Nanotechnology has emerged as a prominent research area in the scientific community with a wide range of applications, including biofuels. This review discusses the specific approaches and methods to synthesize nanoparticles from natural materials. In addition, it summarizes the use of nanocatalyst, nano-additives and microbial enzymes to enhance biofuel properties and production processes. This review also highlights the effect of plant-based nanoparticles on diesel engines' combustion performance and emissions.

1. Introduction

The current crisis in the world's oil supply, production costs, and energy consumption in developing nations [1]. Climate change has become more pronounced due to environmental problems, including GHG (Greenhouse Gas) emissions, mainly CO_2 from the large-scale burning of fossil fuels [2]. The transportation sector will account for about 75 percent of the projected oil market by 2050, with an estimated 145 thousand gallons per day [3]. Many researchers have been looking into various techniques to shift from coal and crude oil-based fuel to renewable biofuel. There is also an urgent need for the scientific community to address energy and environmental problems through novel approaches. The various sustainable energy solutions include solar, wind, biomass, and geothermal plants. Additionally, efforts have been made to produce liquid or gaseous fuels or steam from waste biomass. However, technological and economic constraints continue to restrict the use of biomass for biofuels [4]. Innovative sustainable technologies are crucial for the rapid transition to a carbon-free economy [5].

Nanotechnology is an engineering science branch used to test emerging technical alternatives. The science of growth, synthesis and characterization of nanoparticles and their applications can be described simply as nanotechnology. Since adopting the term "nanotechnology," which focuses on the identification, preparation, and characterization of nanoparticles, many publications have been developed. It allows chemists, physicists, biotechnologists, and engineers to work at the cellular and molecular levels. It has tremendous potential for improving human life [6].

Nanotechnology has become a relevant topic of research in recent years. It has evolved substantially, offering various opportunities for industrial application. Nanotechnologies are increasingly used in telecommunications, chemical processing, cosmetics, fuel, paints, bioremediation, catalysts, pharmaceuticals, and life sciences. Nanoparticles have sophisticated scientific properties which can be applied for efficient bioenergy production [7]. Figure 1 describes the bibliographic mapping of Biofuel production with Nano-technology. Nanotechnology has many applications in biofuels, such as feedstock modification, catalytic production, and esterification reaction. Nanotechnology can provide an effective solution for biofuel by changing the characteristics of feedstocks. The application of nanotechnology to biofuel processing offers exciting solutions to some of the problems in this area.

2. Nanoparticle: Synthesis, preparation and characterization

This section studies nanoparticles and their shapes, properties, and synthesis methods. Figure 2 illustrates the various nanoparticle preparation methods [8]. Table 1 examines the most commonly used nanoparticles, their preparation process, and their characterizations. Nanoparticles, including carbon nanoparticles, magnetic materials,

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metal oxide nanoparticles, etc., are widely used in multiple applications.

These nano-sized materials are known for their unique properties, i.e., mechanical and physical properties, including reactivity, durability, elasticity, resilience, and good electrical and thermal conductivity [10]. The size, structure, and morphology of nanoparticles have an essential role in their properties. The preparation route depends on the resulting nanoparticles' shape, properties, and intended use. Over the past decade, research initiatives for developing different nanoparticles from green materials have been undertaken [9].

Enzymes are used to hydrolyze biofuels and to produce biofuel production from oil and fat. In this context, enzyme extraction or immobilization using nanostructures may be used to achieve successful catalytic activity and biocatalyst recovery. This method provides alternatives for adding magnetic properties to immobilized systems [7].

Novel research on using aluminum oxide nano-particles has been undertaken to improve bio-oil properties [11]. Various synthesizing techniques have been developed to monitor the results of Nanoparticles in shape, size, and morphology [12]. These include chemical vapor deposition (CVD), sol-gel processing, and mechano-chemical processes.

Inorganic nanoparticles, such as metal oxides, are usually produced by wet chemistry, chemical micro-emulsion, hydrothermal, sonochemical, and direct deposition. However, Carbon Nanoparticles are typically made by arc discharge (AD) and chemical vapor (CVD) techniques [13]. Nano-sheet graphene uses both chemical and thermal exfoliation techniques.

The adherence of particles to one another by weak forces causes the agglomeration of nanoparticles, resulting in (sub)micron-sized entities. An effective way to prevent agglomeration in nanoparticles is to add surfactant. A suitable plant extract can serve as a capping and reducing agent. Plant extract or organic fiber is used as a reduction or stabilization agent for the production of nanoparticles [14].

Nanoparticles may be used as attractive carriers to immobilize catalysts, allowing them to recover from the liquid process by filtering or centrifuging. For example, modified nano-sphere silica has been shown to remove pathogens from living microalgae and enhance lipid accumulation [15].

2.1. Nanoparticles – synthesis

Nanoparticles may be categorized by source, chemical composition, synthetic path, shape, size or composition, and application [16]. Production/preparing nanoparticles can generally be either a "bottom-up" approach or a "top-down" approach, as shown in Figure 1. The bottom-up method described by its name includes the formation of nanoparticles by single molecules or nanoparticle atoms, e.g., sedimentation and reduction techniques [17]. The substratum or source is either used by sedimentation or by reducing the production of nanoparticles. On the other hand, the top-down approach requires dividing bulk products into smaller units that are then transformed into appropriate nanoparticles [18].

2.2. Characterizations of Nanoparticles

Chemical properties, structure, size, and appearance play an important role in determining the characteristics of the nanoparticles that emerge from them. Nanoparticles are generally categorized by size, shape, and surface load using microscopic methods such as electron micro scanning, electron transmission, tunneling microscopy, and atomic energy microscopy. For surface features, molecular structure, chemical composition, nanoparticle size and shape, transmission electron microscopy and scanning electron microscope are essential [19]. Electron microscopy can resolve these disadvantages by scanning field emissions imaging samples that are not pre-treated; however, samples are still handled in most cases. Atomic force microscopy provides information on surface thickness, shape, hardness, and roughness [20]. However, atomic force microscopy overestimates the size of nanoparticles, mainly if the tip structure is more significant than that of nanoparticles. Nanoparticle atoms and molecules can be observed in a tunneling microscope scan by switching between the states to be seen, such as air, liquid state, or the gas environment [21].

Light scattering methods are used primarily to determine the size of nanoparticles. Dynamic light scattering (often referred to as photon compatibility spectroscope) is the most common technique for determining particulate size in colloidal suspensions [22]. The suspended particles are exposed to Brownian random motion; thus, the light

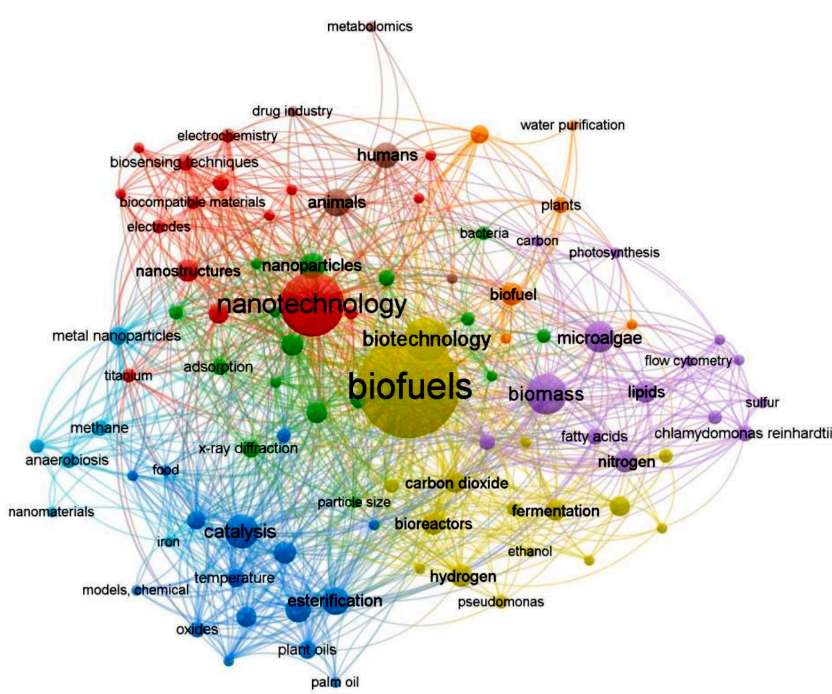

Fig 1. Bibliographic mapping on biofuel production with Nano-technology

scattering over the suspended particles makes it possible to determine the size of the particles. The calculation is based on observing light-dispersed fluctuation intensities with the particles during laser beam irradiation [23].

X-ray techniques enable absorption, fluorescence, spectroscopy, energy distribution, and diffraction. These techniques commonly provide data on surface and substrate structures, crystallographic structure, bonding status, or elementary composition [24]. Energy Dispersive X-Ray Spectroscopy and Photoelectron Spectroscopy are usually combined for simple measurement and quantitative research with either electron microscopy scanning or transmission microscopy [51]. The absorption of X-rays is used to identify untreated electronic states, chemical composition, and bonding conditions in nanoparticles. The relationship between the characteristics of a-synthesized nanoparticles and the standard provides data based on their structure and properties [25].

Reciprocal oscillations of their electrons use spectroscopic techniques in reaction to electromagnetic waves in Plasmon-resonance nanoparticles. UV-visible is used to define the characteristics of metal nanoparticles in terms of size, shape, and surface properties, particularly in the form of functional nanoparticles [26]. This needs to come from plasma nanoparticles, which absorb visible radiation from the near-infrared region depending on their shape and scale. This character can be correlated with the mutual oscillation of nanoparticles' surface electrons known as the surface plasmon resonance. Due to the characteristic surface plasmon resonance of nanoparticles, the dispersed nanoparticles in suspension have more points that could be used to obtain helpful information on the distribution of shapes and sizes of nanoparticles [27].

Nanoparticle tracking technology is one of the most common methods in particulate matter sizes below ~15–1250 nm, with a lower detection limit varying from the nanoparticle's refractive index. It uses a susceptible computer-loaded image camera to record light from each nanoparticle and monitors the Brownian movement of single nanoparticles to create separate proportional measurements [28]. Hyperspectral imagery is a further approach, which focuses on dispersing visible and infrared incident light through a vast dark field. Provides the strength of a single nanoparticle (size $<$ 08 nm) based on the NP form, with distribution patterns and spectral composition [29].

Quantifying nanoparticles is another significant step toward understanding nanoparticles' origin, behavior, and presence under different conditions. Various sensitive analytical methods were used to identify and classify nanoparticles, as described in the sections below [30].

Inductively coupled plasma mass spectrometry is usually used during nanoparticle synthesis as it helps to measure the concentration of the total colloidal solution product. Nanoparticle characterization of the shape has now been further extended. Previously, plasma mass spectrometry coupled inductively was used to determine the distribution of

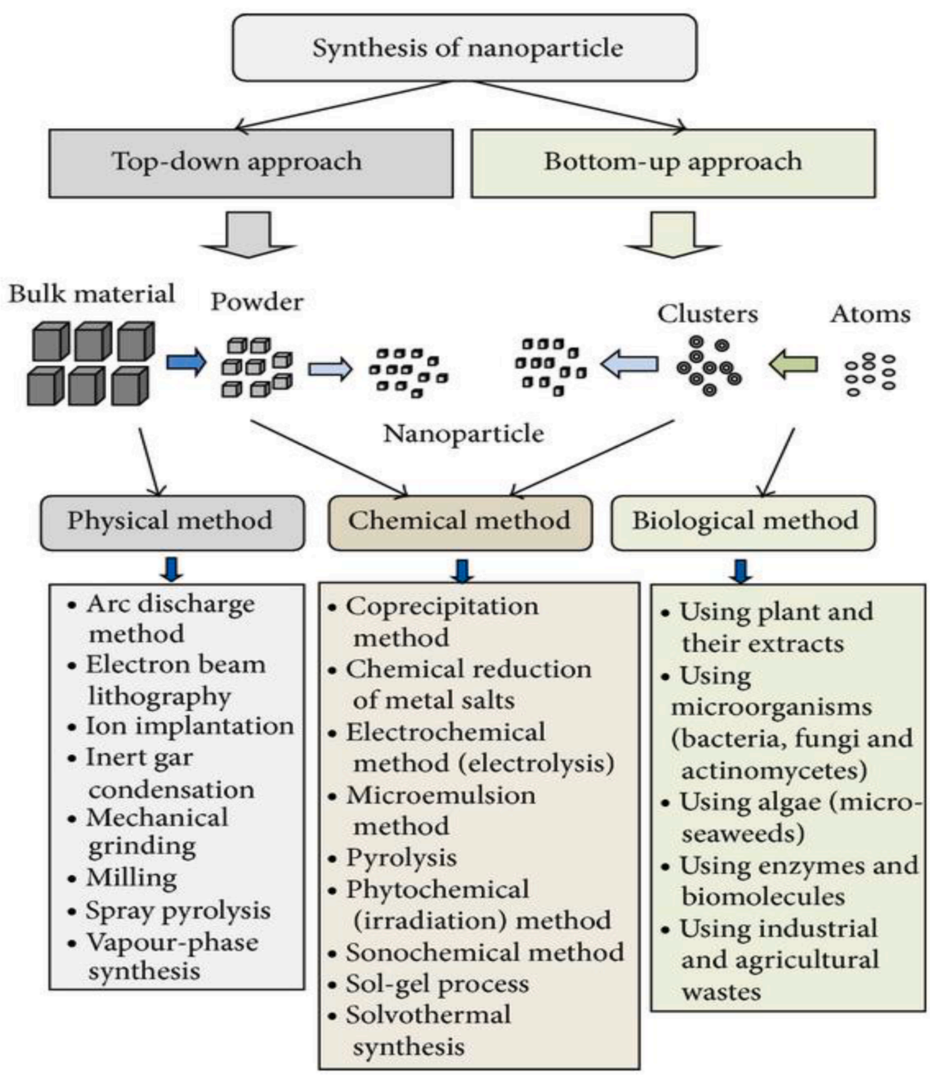

Figure 2. Nanoparticles preparation methods[14]

nanoparticles [31]. Thus, the plasma detection of various ion clouds caused by atomized nanoparticles induces a signal of resistance to the particle size detector. Inductively coupled plasma mass spectrometry is among the analytical methods used to measure nanoparticle concentration, volume, and size distribution [32]. It calculates the intensity signals generated by the ionization of the individual particle instead of the continuous ion flow to the plasma. Inductively coupled plasma optical emission spectroscopy was also used to study nanoparticles.

2.3. Nanotechnology in Biofuel Production

Biofuels are classified as first, second, third, and fourth generations on the basic principle of raw materials sources. The FAME converts sugar, starch, vegetable oil, and fats into bio-alcohol in first-generation biofuels [33]. First-generation biofuels are made from various feedstocks such as starch, sugar cane, animal fats, and vegetable oils. There are several questions about the use of such feedstocks. Second-generation biofuels have a harmful carbon content mainly dependent on lignocellulose biomass. Thus, the idea of second-generation biofuels is making headway in the global biofuel development scenario using non-food feedstocks. Although 2nd generations of biofuels have some advantages, such as waste material, there are also disadvantages, such as high production and maintenance costs and technical problems. In the 3rd and 4th generations, algae are used to convert lipids into biofuel. In the 3rd generation, algae are used explicitly for biofuel processing, whereas metabolic algae from oxygen photosynthetic micro-organisms produce carbon reservoirs in the 4th generation. Biofuel is made using a catalyst (chemical, magnetic, enzymatic) and the influence of several process parameters such as pH, temperature, and pressure plays a key role [34]. They are also classified as biofuel, which is chemically and biologically synthesized[35]. Hence, taking these facts into account, researchers developed appropriate bi-functional catalyst to increase the biofuels yield and solve the problems of mass production [36]. Because of this, the application of nanotechnology should overcome the difficulties mentioned above by providing an opportunity to change the characteristics of feedstock materials from biofuel. Nanoparticles have strategic applications in biofuel production due to their exceptional physiochemical properties. Several Nanoparticles have been used to produce biofuels, including titanium dioxide, ferrosoferric oxide, tin oxide, zinc oxide, and carbon with unique properties [37]. Furthermore, magnetic nanoparticles are widely used in biofuel processing due to their high surface volume fraction, quantum properties, and small-scale immobilizing properties. In addition, the most important properties of such nanoparticles must be easily retrieved by applying an appropriate magnetic field to the reaction mix [38].

3. Plant-based Nanoparticle

Metallic nanoparticles such as silver and their various components have become the main ingredient of daily use [39]. Since these Nanoparticles, which have a diameter of 100 nanometers, now play a significant role in our daily lives, there are also issues about the potentially harmful effects on humans and the climate. Nevertheless, these statistics generally neglect that nature is a qualified nanotechnologist, with numerous natural patterns existing in nature. Nanoparticles originate from natural sources, such as geological rocks, volcanoes, and human beings. Life ultimately changes the path to a microscopic cell and metabolizes tiny molecules [40]. Natural Nanoparticles need to be distinguished because they are either created or synthesized without human intervention in the environment, as well as "bio" and "nano" chemicals [41]. In another case, 'natural' refers to 'biological', which explicitly includes biological substances, as in 'natural products'.[42].

Table 2
Different Plant-based metallic nanoparticles

S. No	Plant	Plant part	Nano-particles	Size (nm)	Ref
1.	Euphorbia prostrata	Stem	Silver nano-particles	67-88	[91]
2.	Citrus sinensis	Stem	Silver nano-particles	122-124	[92]
3.	Cinnamon zeylanicum	Leaves	Silver nano-particles	18-22	[92]
4.	Euphorbia prostrata	Fruits	Gold nano-particles	12-16	[93]
5.	Dillenia indica	Leaves	Gold nano-particles	8-24	[93]
6.	Aloe vera	Leaves	Silver nano-particles	90-150	[94]
7.	Caria papaya	Fruits	Gold nano-particles	20-50	[95]
8.	Cinnamon zeylanicum	Seeds	Zinc oxide nano-particles	15-24	[95]
9.	Acalypha indica	Stem	Silver nano-particles	11-24	[95]
10.	Cinnamon zeylanicum	Seeds	Gold nano-particles	40-50	[96]

3.1. Inorganic Nanoparticles – Natural sources (Plant-Based)

So many inorganic Nanoparticles have been discovered on Earth, produced from natural sources. And these particles are regular, much more often than not ordinary. Volcanic powder mist produces a wide variety of miniaturized poly-dispersion sizes and Nanoparticles. Such particles range in size from 50 to 250 nm and are primarily made from iron and silicate composites [43]. These particles purposely circulate in the air, and exposure to pulmonary conditions cannot be prompt until they are solid. "Carbon Nano-tube" residue has been deliberately airborne and poses significant threats to animals and humans. Under a microscope, drinking water can be filled with nanoparticles of different sizes, almost like minute solid objects of some kind [44]. These samples are not typically of good quality and cannot be easily categorized and characterized. In the meantime, however, these inorganic substances have stimulated counterparts to generate a variety of related particles that generally rely on distilled nanoparticles such as Fe_3O_4 and MnO_2. Figure 3 illustrates the method of Metallic Nanoparticles plant-based

Table 1
Popular nanoparticles and their properties

Nanoparticles	Process	Procedure	Characteristics	Reference
$AgNO_3$ (Silver Nitrate)	Biosynthesis of chitosan as a stabilizing and reducing agent	Fourier Transform Infrared Spectroscopy (FTIR)	The maximum processing time for $AgNO_3$ at 100°C was 14h to produce 3–15 nm nanoparticles.	[17]
$ZnCl_2$ (Zinc Chloride)	Biosynthesis with Arbortritis Nyctanth extract	Transmission electron microscopy (TEM)	The size of the nanoparticles was 10–30 nm	[18]
$ZnC_4H_6O_4$ (Zinc Acetate)	Hydrothermal processes	X-ray crystallography (XRD)	Polycrystalline films with a hexagonal crystal structure 12 nm in size	[21]
ZnO powder (Zinc Oxide)	Chemical vapour deposition (CVD)	X-ray photoelectron spectroscopy (XPS)	Nanoparticles (nano-wires) of 3 nm in diameter and 45 nm in length	[25]
$Mg(NO_3)_2$ (Magnesium Nitrate)	spraying a solution on a heated surface (Spraying pyrolysis)	Ultraviolet–visible spectroscopy (UV–Vis or UV/Vis)	12 nm of crystalline size has been obtained	[26]

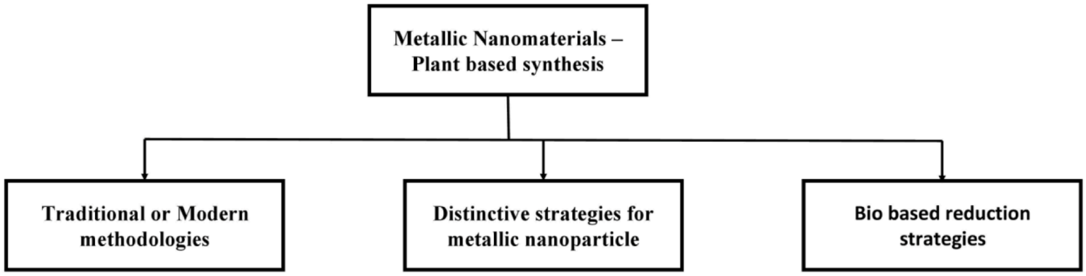

Figure 3. Metallic nanoparticles plant-based synthesis [43]

synthesis [45]. Likewise, the types of molecular stimuli are comparable in these "straightforward" physical cycles but are progressively regulated, often due to routine oxidation. One example was the oxidation of hydrogen sulfide gas found in volcanic eruptions or wells characteristic of many parts of the world. All the issues discussed need to be identified and considered the synthetic course of action [46].

3.1.1. Metallic Nanoparticles – Plant-based synthesis

3.1.1.1. Modern Methods. Metals have been seen as a symbol of strength, and some metals are still considered popular today as gold and silver. Gold was used for spiritual and psychological refinement in the eighteenth century [47]. Arabs use water made of gold-plated metal. In rural areas, the value of gold is still recognized as workers prepare their rice dishes with a gold pellet to compensate for the lack of minerals in the body. Gold is used to treat wounds and heal ulcers. In reality, silver nanoparticles, colloidal in nature, have been used as wound wrapping materials, as a tooth bond, and as a filtration system [48].

3.1.2. Silver nanoparticles Plant-based

Physical and chemical approaches were used in the long-term synthesis of silver nanoparticles. Furthermore, progress has now identified the fundamental role of biological processes [49]. Chemical and physical processes are energy-intensive and have high production costs. Synthesis of silver nanoparticles may involve the absorption of harmful chemicals on nanoparticle surfaces, which may cause toxicity problems by chemical processes such as hydrazine hydrate, ethylene glycol and di-methyl-formamide. Aqua chemicals thus cause a long-term accumulation of nano-chemical silver colloids, ensuring accommodation with the dimensional substance after shipment [50]. Plant derivatives for other biological systems, such as fungi and microorganisms, have been investigated for bio-based production of silver nanoparticles to be significantly better candidates because they do not require hazardous caps and reduced factors, heat, radiation, fungal/microbial strain, and expensive media to sustain microbial growth and nanoparticles [51]. Table 3 shows various types of plant-based silver nanoparticles. The chance of particulates or infection is often reduced during the creation and execution of areas. Several other scientists have investigated the role of many herbal metabolites in developing silver nanoparticles, such as amines, flavonoids, polyphenols, terpenoids, aldehydes, ketones, arabinoses, and galactose [52].

Several methods (such as human, chemical, enzyme, and physical) to combine nanoparticles are used. Mechanical techniques include ball sorting, heat dissipation, laser smashing, radioactive shaft, and amalgamation of nanoparticles. Synthetic systems employ high-frequency radiation operators that threaten human well-being. Figure 4 shows the plant-based Nano-particles synthesis [53].

The biochemical input of silver nitrate contributes to the organization of silver nanoparticles using vegetative tools. Plants also contain other essential biomolecular, such as proteins, amino acids, enzymes, and metals. Such bimolecular can then be used for the bio-reduction process [54]. The result of this metal-like gold was, thus, the transition and removal of Au+ redox catalysts for Au_0 nanoparticles. Figure 4 provides the various plants-base Nano-particles synthesis, and Table 3 shows the different types of plant-based nanoparticles.

Table 3
Different plant-based Silver nano-particles

S. No	Plant	Plant part	Silver Nano-particles specifications	Ref
1.	Euphorbia prostrata	Stem	6-14 nm	[97]
2.	Citrus sinensis	Root	64-80 nm	[97]
3.	Cinnamon zeylanicum	Leaves	15 nm	[97]
4.	Rosa rugose	Fruits	25-40 nm	[98]
5.	Piper betle	Leaves	35-41 nm	[98]
6.	Aloe vera	Stem	17-110 nm	[98]
7.	Parthenium hysterophorus	Fruits	24 nm	[98]
8.	Mangifera indica	Leaves	7-68 nm	[99]
9.	Acalypha indica	Leaves	14 nm	[99]
10.	Daucus carota	Root	9-25 nm	[101]

3.1.3. Gold nanoparticles – Plant-based

Nanoparticles derived from gold are used in several applications. The Au (3+) ion is decreased to the Au atom by connecting the electron to the molecule's surface, while the Au integrates and clumps to generate gold nanoparticles [55]. At the initial binding stage to the atoms, Au ions are highly likely to produce clumps of gold nanoparticles in medicine. Table 4 shows various types of plant-based gold nanoparticles. Bio-based metallic nanoparticle synthesis has some morphology and specified forms, such as hierarchies, triangles, hexagonal nano-parts, decahedrons, icosahedrons, nano-triangles, and node bands [56].

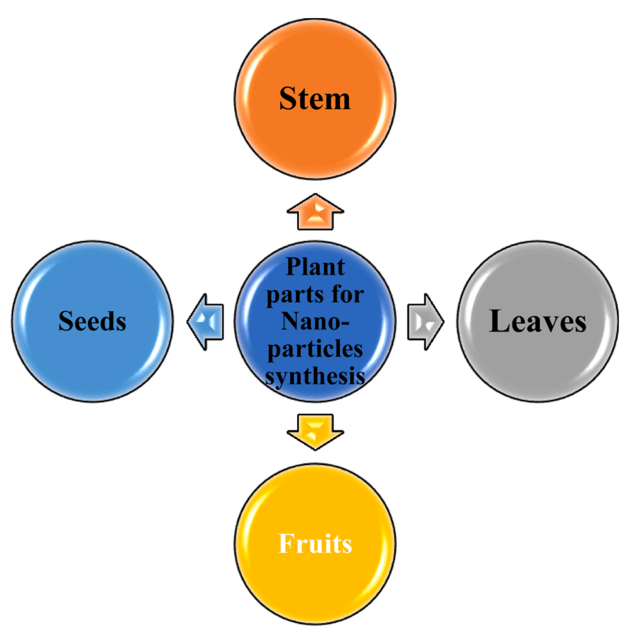

Fig 4. Plants based Nano-particles synthesis [94]

Table 4
Different Plant-based Gold nano-particles

S. No	Plant	Plant part	Gold Nano-particles specifications	Ref
1.	Punica granatum	Flower	12.75-36.84 nm	[102]
2.	Hygrophila spinosa	Root	12-18 nm	[102]
3.	Morinda citrifolia	Stem	64-78.5 nm	[102]
4.	Citrus maxima	Fruits	75 nm	[102]
5.	Coleus forskohlii	Stem	13-16 nm	[103]
6.	Argemone mexicana	Leaves	7-18 nm	[103]
7.	Abelmoschus esculentus	Fruits	6-20 nm	[103]
8.	Hibiscus sabdariffa	Leaves	6-35nm	[104]
9.	Ixora coccinea	Stem	98.5-115 nm	[104]
10.	Cassia fistula	Root	63.75-79 nm	[104]

Nanoparticles have various beneficial applications in different areas; thus, investigators are now concentrating on their forms.

3.1.4. Zinc oxide nanoparticles – Plant-based

Due to their other features, zinc oxide nanoparticles are even more fascinating, e.g., their processing is fast, easy to use, and easy to synthesize. Due to their wide use in optics, electronics, and biomedical systems, researchers are concerned with nanoparticle zinc oxides [57]. Nanoparticles made of zinc oxide have immense anti-inflammatory and injury healing properties. They have the characteristics of UV absorption and are also used in cosmetics such as sunscreen and lotions. These are now widely used in medicines such as anti-cancer, herbal products, antifungal, anti-bacterial, anti-diabetic, and livestock [58]. Table 5 shows various types of plant-based zinc oxide nanoparticles. This is often used in manufacturing rubber and cement, dentistry and arsenic, and in treating sulfur water. Nanoparticles containing zinc oxide have a lot of morphology, including nano-rods and nano-belts [59].

4. Nano-catalyst for Biofuel Production

Several articles, reviews and chapters on the use of Nanoparticles to promote the production of biofuels and the resulting fuel quality have been published. These studies can be examined from four perspectives: working conditions for feedstocks, sustainability prospects and production opportunities. Indeed, there are substantial feedstock-related costs associated with the production of biofuel. In recent years, there has been more concentration on exploring various aspects of nanocatalysts for different applications.

Nanocatalysts are a sustainable and environmentally sustainable biofuel production method that offers enhanced catalytic conversion, increased selectivity, economic processes, milder working conditions, and long-lasting catalysts [60]. Thus, it is essential to identify cost-effective feedstock and reliable technology for the marketing of biofuel [61].

Metal oxide nanocatalysts are widely used in many applications, including additives and materials. Nanoparticles such as calcium oxide, aluminum oxide, and magnesium oxide are now classified as more than 80% heterogeneous converting catalysts with lower oil content. In addition, nanoparticles can be directly used as heterogeneous catalysts to increase the biofuel conversion yield and fuel output.

Table 6 provides the list of nanocatalysts and their chemical description. Nanocatalysts lead in the latest research fields due to their excellent ability to deliver large surface areas. They have unique chemical and physical properties due to the size and higher density of the active catalytic sites [62].

These Nanoparticles have also been widely used in biofuel and gasification/pyrolysis processes. Catalyst classifications and the number of studies published on the nano-catalytic conversion of biofuels [63].

Table 5
Different Plant-based Zinc oxide nanoparticles

S. No	Plant	Plant part	ZnO Nano-particles specifications	Ref
1.	Cocos nucifera	Flower	15-35 nm	[105]
2.	Nephelium lappaceum	Root	7-60 nm	[105]
3.	Vitex negundo	Stem	115-135 nm	[105]
4.	Rosa canina	Fruits	55 nm	[106]
5.	Gossypium	Stem	23-86 nm	[106]
6.	Coptidis rhizoma	Leaves	77-101 nm	[108]
7.	Solanum nigrum	Fruits	96-120 nm	[108]
8.	Calotropis gigantea	Leaves	96-135nm	[108]

4.1. Applications of Nanocatalyst in Biofuel Production

The transformation of waste cooking oil into biofuel using heterogeneous nanocatalysts has received considerable attention. KF / CaO-NiO, CZO, FMSZ, and FMTZ acid catalysts were nanocatalysts tested as feedstocks using waste oil. These catalysts used FMSZ and FMTZ for feedstocks with a high FFA value and demonstrated better catalytic efficiency and higher recyclability than other catalysts [64]. In recent years, super-acid nanocatalysts have been involved in the development of biofuel as a subgroup of heterogeneous catalysts, as they are not only capable of solving several acid-homogeneous catalyst defects. The most efficient parameters for nanocatalyst processing have been developed in catalytic converters, preparation methods, and calcination procedures, based on the nanocatalyst studies documented for the production of biofuel [65]. Oil to alcohol molar ratio, temperature, stirring speed, catalytic processing, and reaction time were the main operating parameters that influenced biofuel development. Table 7 explains various feedstock and nanocatalysts used for the production of biofuel. The operating parameters used for oil/alcohol molar ratio, temperature, strain, catalyst feed, and reaction time was 2:6–1:42, 25–280 °C, 5–80 bar, 0.3–15 wt % , and 5–15 h, respectively. While the implementation of extreme operating conditions has resulted in higher catalytic efficiency and greater reusability of nanocatalysts, this is not a preferred solution for large-scale production processes [66].

The stability of catalysts is one of the most critical issues of heterogeneous catalysts for industrial applications. Some research studies have extensively examined the reusability of Nano catalysts and the number of pathways used for catalysts. In contrast, some studies have generally reported the high recyclability of catalysts without specific details. KF / CaO and KF / CaO – Fe3O4 Nano catalysts were the most active catalysts in the 18th and 88%, respectively, in the 26th cycle, with a Biofuel yield of 94% [67]. Based on numerous nano-crystalline CaO studies, the presence of nano-sized CaO as an extremely active fundamental catalyst for transesterification reactions is highly effective due to the high surface area associated with smaller crystalline nanoparticles. In short, the reason for low reusability catalysts is that the catalytic components do not have stability during the reaction and are leached into the reaction mix with the exact mechanism of the homogeneous catalysts [68]. Higher calcination temperatures can be used as a catalytic preparation method for leaching and can also be used to minimize components reaching higher catalytic surfaces and stable catalysts. Another definition relates to the catalytic deactivation of organic impurities and the formation of moisture and enolates caused by carboxyl de-protonation [68–69].

From a marketing perspective, the CaO, KF / CaO, and TiO2-ZnO

Table 6
Various Nanocatalysts

Catalyst	Chemical description
Alkali earth metal oxides	MgO, CaO
Transition metal oxides	ZnO, NiO
Mixed metal oxides	TiO_2–ZnO
Supported metal oxides	KF/Al2O_3, KF/CaO, CaO/Fe_3O_4

Table 7
Different Feedstocks and Nanocatalysts for the preparation of biofuel

S.No	Feedstock	Nano-Catalyst	Yield (%)	Reference
1.	Corn oil	Ti_2O - ZnO	97.5	[112]
2.	Sunflower oil	KF/CaO	94.3	[116]
3.	Neem oil	CaO	86.7	[116]
4.	Karanja oil	ZrO_2	91.5	[117]
5.	Palm oil	ZnO nano-particles	98.2	[118]
6.	Soybean oil	K_2O/Ti_2O	85.6	[119]
7.	Jatropha oil	Li-CaO	99.4	[121]
8.	Olive oil	Fe3O4/AI/Cs	93.8	[122]

nano-catalysts applied for Biofuel production have shown promising potential for implementing Biofuel scale-up processes [70]. It should be noted that requirements for higher catalytic efficiency and more excellent durability should be considered in evaluating effective Nano-catalyst marketing technology and other areas such as ease of service and probability of low-cost feedstocks and low-energy utilization [71]. While metal oxides improve the efficiency of nano-crystalline types rather than ordinary industrial catalysts, there are also issues, such as reusing catalysts, which require more research soon [72]. Figure 5 shows the mapping of Catalyst types and characterization.

4.2. Applications of Nano-catalyst in Biofuel Production

Nanoparticles may be used as a substrate for lipid aggregation, lipid separation, and transesterification processes or as the substrate for biofuel development [73]. Nanoparticles have been used as lipid immobilizers due to their wide surface area and can be quickly recycled from the liquid process by filtration or centrifugation. Due to the high lipid affinity of organic solvents, including hexane and methanol, the immobilization of organic solvent-like chemicals on nanoparticles allows for the recovery and removal of lipids [74]. In addition, the modified nano-silica allowed the extraction of living algae which could be returned for further accumulation of lipids. Some studies have also found that nanoparticles promote microalgae growth and enhance lipid accumulation. However, nanoparticles must be carefully treated to prevent adverse effects on microbes [75].

Transesterification is one of the most influential and straightforward technologies in biofuel development. In this context, different oils of different origins, including animal, plant or soil microorganisms, react with alcohol to generate methyl ester fatty acids [76]. These reactions occur under high pressure and temperature or moderate catalytic conditions. In this latter case, the acids and the base are often used as catalysts for biofuel processing on a laboratory and industrial scale. The acid and soap-forming corrosivity of the base catalyst paved the way for

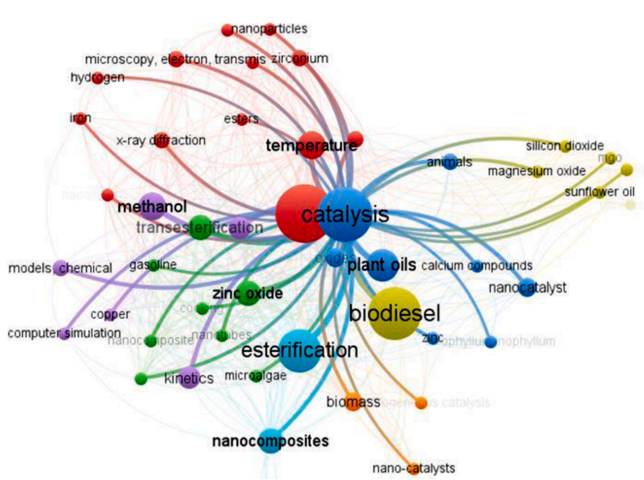

Figure 5. Mapping of Catalyst types and characterization

environmentally stable and effective biocatalyst lipase enzymes [77]. However, the cost of using this biocatalyst enzyme is considered the most significant constraint that hinders its efficiency. Various approaches have been developed to reduce total costs, such as lower output rates for lipase, quality improvements, and more than once re-use [78]. The above is seen as a realistic way to minimize running costs. In this context, lipase has been immobilized in different containers, such as fabric, acrylic resin, silica gel, hydrotalcite, and microorganisms. Due to their unique characteristics, such as the large surface-to-volume ratio, nanoparticles have been extremely interesting in recent years, providing a vast area for lipase immobilization [79].

The lipase function is 96% immobilized in nano-size silica after eight months of preservation, compared to 50% for free lipase activity. In addition, lipase immobilization on functional zirconia increased activity and remained eight times higher after re-use. Other nanoparticles, including carbon nitride, can also be used to avoid lipase [80]. Carbon nitride, one of the two-dimensional forms of Nanoparticles, can immobilize lipase to maximize its performance. Lipase immobilization nanosheets showed adequate processing, pH durability, thermo-stability, and recyclability [81]. The problem with immobilizing nanoparticles by a biocatalyst in recovering such products is inherently cumbersome. In some research, centrifugation has been used as a treatment modality; however, it takes much time and energy [82].

Magnetic nanoparticles have been a significant research priority in recent decades for the rapid recovery of all Nanoparticles and immobilized biocatalysts. Magnetic nanoparticles can be removed easily with an external magnet to improve the recovery and reusability of biocatalysts [83]. Magnet nanoparticles can be quickly retrieved using an external interest that increases the recovery and reusability of biocatalysts. The aggregation and lack of chemical interaction of magnetic nanoparticles are also significant drawbacks [84]. One way to overcome these problems is by coating magnetic nanoparticles with inorganic materials such as carbon and silica. Magnetic silica aerogels have been synthesized in a process subjected to chemical modifications and environmental strains of iron oxide nanoparticles, including sodium silicate byproducts. The total potential for adsorbing lipase by physical adsorption was approximately 92.64 mg / g [85].

In addition to the enzyme catalyst, heterogeneous catalysts that are solid acid or base have been studied in the production of biofuels. Nano-catalyst provides high catalytic efficiency and allows chemical insulation. The transesterification of Nano-catalysts produces biofuels. Nano-catalyst provides high catalytic efficiency and rapid product separation between the two [86]. It is known that the conventional catalyst is 50% of the Nano-catalyst required for similar reaction conditions. In addition, the process did not depend on moisture or the free fatty acid amount [87].

4.3. Role of Surfactants

Direct applications of nano-emulsions, have most notably in the pharmaceuticals (drugs), personal care (hygiene), healthcare (agrochemicals), and cosmetics industries. Surfactants are primarily employed in the production of nanoparticles for dispersion. Because of their non-equilibrium nature, nano-emulsions display various properties and behaviours influenced by their chemical composition and manufacturing process [88]. As a result of these new uses, more research into Nano-emulsion preparation optimization methods is now necessary [89]. Surfactants have a hydrophilic head and a long hydrophobic tail that distinguishes them. Many researchers used Nano emulsions and oxygenated additives with biodiesel blends to enhance the diesel characteristics [90]. As a result of their attraction to a broad spectrum of different substances, surfactant molecules are usually referred to as amphiphilic. Surfactants are required components in the preparation of Nano-emulsions because they aid in the reduction of interfacial tension. Because surfactants are present, newly formed droplets, do not combine to form larger droplets [91].

Very few studies were carried out on the lubricity of these ternary fuel blends used for improving engine characteristics.

Surfactants are categorized according to the hydrophilic proportion of their charge.

Amphoteric surfactants,

Nonionic surfactants,

Anionic surfactants,

Cationic surfactants.

It is possible to reduce effectiveness and maybe recoalescence by increasing the quantity of surfactants employed [92]. Because the surface tension of the whole is lower than that of the separate components, the combination has the potential to benefit. Smaller droplets can be formed by dissolving the surfactant in the dispersed phase instead of the uninterrupted liquid phase [93].

5. Effects of Nano-additives Biofuels in Internal Combustion Engine

The key feature of this research is to focus on using potential nano-additives with different fuels in CI engines [93]. This section discusses the following aspects of nanoparticles blended in biofuels.

Nano-additives as a catalyst for the enhanced biofuel characteristics

Improvement of diesel engine operating characteristics with nano-additives.

Improvement of diesel engine emission characteristics with nano-additives.

Effect of nano-particles on human health

5.1. Nano-additives as a catalyst for the enhanced biofuel production process

Many additives have been made available for several years to improve fuel properties, and attempts have been made to combine diesel and biofuel to test the functional properties [94]. Figure 6 explains the impacts of Nanoparticles as the catalyst. Mixing additives to both ordinary fuel and emulsion is necessary to prevent problems caused by cold weather in the engine and to increase the fuel performance [95]. Reactive solutions such as alkyl nitrates have been used for these issues but have been classified as highly corrosive and toxic. Several studies have used ignition promoters, oxygenated or catalytic additives to enhance fuel properties and maximize the performance of intermediate and medium cetane combustion [96].

Several studies have shown the use of additives for water-based biofuel emulsion fuels to increase production and mitigate harmful pollutants. First, the metal content of the emulsion interacts with water to create OH ions, reduce dust deposition and, secondly, minimize carbon oxidation [97]. Many studies have shown an interest in using aluminum nanoparticles as diesel and Biofuel fuel additives. In addition, aluminum nanoparticles are stable for catalytic reactions at high oxidizing temperatures [98]. Several researchers have recently conducted extensive studies on using carbon nanotubes and aluminum as fuel additives to improve fuel efficiency as much as possible.

In addition, when combined with a solvent, the nanoparticles tend to be agglomerated. Various extraction methods and spreading nanoparticles in the base fluid overcome binding forces after wetting the material [99]. To break agglomerates into aqueous and non-aquatic suspension systems, several researchers have widely embraced ultrasonic dispersion, leveraging the maximum potential of nano-sized materials for different applications [100]. Recently, scientists have systematically prepared mixed Biofuel fuel with ultrasonic machines. The same research group used ultrasonic and mechanical homogenizers to combine nanoparticles in water-Biofuel emulsion fuels. They also demonstrated that, unless the frequency of agitation of the mechanical homogenizer is increased, the stability of the prepared fuels will improve [101]. Several studies have suggested ambitious strategies to improve the strength of nano-additive emulsion fuels for the long-term use of engineered fuels.

5.2. Diesel engine operating characteristics using nano-additives

The scientific community has developed various innovative methods to eliminate toxic gas emissions from Internal combustion engines. One technique is using nano-additives in fuel as an ignition promoter or catalyst [102]. Researchers have recently confirmed that nano-additive blended fuels have increased thermal brake performance compared to renewable fuels. In addition, the ignition delay for nano-additive mixed biofuel fuels has been substantially reduced compared to renewable diesel and Biofuel fuels [103]. This resulted in decreased cylindrical pressure and gaseous emissions compared to renewable fuels for mixed biofuel nano-additives.

Biofuel emulsion-fueled nanoparticles have been tested in Compression Ignition (CI) engines. They found improved brake thermal efficiency, reduced peak cylinder pressure, reduced ignition speed, reduced release time, and a low-pressure improvement in nano-additive mixed biofuel emulsions compared to clean fuels [104]. Increased thermal brake efficiency, reduced peak cylinder pressure, shorter

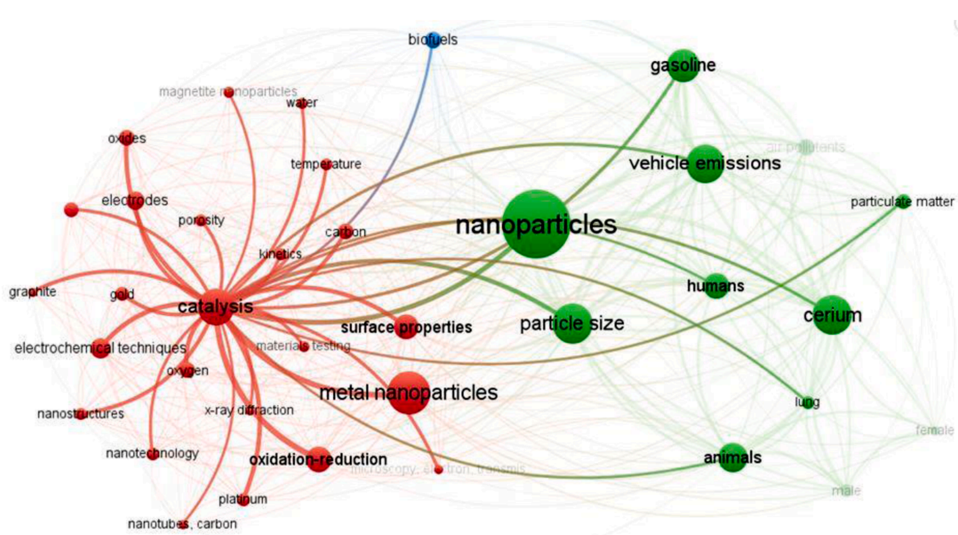

Fig. 6. Impacts of Nanoparticles as a catalyst

ignition delays, and reduced heat exhaust relative to clean fuels were observed. Due to the involvement of nano-additives in emulsions, the researchers have observed that a relatively homogeneous mixture of air-fuel and nano-additives could be achieved in the combustion chamber during combustion, thus increasing catalytic combustion and reaction quality [105].

Nano-additives mixed emulsions have significantly good emission properties. The combined effects of decreased un-burnt hydrocarbons and carbon monoxide emissions for mixed nano-additives for renewable biofuel have been observed. Producing potentially more robust catalytic combustion additives, air-fuel mixing was the reason for reducing emissions from nano-additive blended biofuel emulsion fuels [106]. It will also improve the CI engine's efficiency, emission, and combustion quality. Nano-science researchers are also currently researching the working properties of internal combustion engines when combining nano-additives with lubricants [107].

Nanoparticles' characteristics differ significantly from those of nanoparticles since they are considerably smaller [108]. Nanoparticles are attractive candidates for various energy engineering applications due to their quick oxidation, improved stability, lower melting point, strong heat output during combustion, lower heat of fusion, and enhanced heat transfer nature [109]. Nano additives have been demonstrated to influence various energy-related technical parameters [110] substantially. This fluid includes fluids used in propulsion systems and engine cooling fluids. As previously discussed in greater depth, the nano additives serve a range of uses [111].

Increase in oxygen content
Increase in fuel's oxidation stability
Increase in viscosity
Decrease in the ignition time and a reduction in the flashpoint
Increase in mechanical strength

According to several published literature, the Nanoparticles investigated were metallic nanopowders, polysaccharides, ceramic nanoparticles, non-organic chemicals, and carbon-based nanoparticles such as carbon nanostructures [112]. Oxide nanoparticles are extensively utilized in diesel and biofuel mixe as a dispersion agent. Metallic nanoparticles such as thallium and the rare earth metals ruthenium, palladium, and nickel, when used as catalysts, lower the amount of fuel required and pollutants discharged into the environment and complete the combustion process [113].

The addition of nano-AgO and nano-ZnO_2 to tamanu Biofuel was demonstrated to improve the kinematic viscosity of the biofuel. These additives showed potential benefits in enhancing kinematic viscosity by dispersing multi-walled carbon nanotubes [114]. They observed that employing multi-walled carbon nanotubes improved brake thermal efficiency while reducing the number of contaminants in exhaust streams. In a diesel engine that used graphite to diesel mix of Simaroubaceae methyl ester, there was a reduction in unburned hydrocarbons of 18.46 percent, carbon-no-oxides and nitrogen oxides of 45.63 percent and 14.96 percent, respectively. While using SAE50 fuel, this reduction will occur [115]. Nano fuel is a kind of fuel used in diesel engines. Compared to diesel fuel, the addition of nano-ZnO resulted in an 18% increase in efficiency and a 15% reduction in the fuel required for the brakes [116].

Studies on using nano-sized metals, nonmetals, natural, and mixed components in diesel engine base fuel have recently increased and reported in several recent works of literature. These studies have been published in scientific journals and research papers [118]. Fuel modification is one of the essential strategies for improving performance and lowering emissions. Fuel modification is one of the critical strategies when compared to significant alterations in the engine and exhaust gas emission [119]. Nanoparticles (NPs) increase the contact area to volumetric proportion, heat capacity, and mass diffusivity when distributed in a liquid media, all of which can be employed to improve the thermophysical characteristics of diesel. This allows diesel to be modified to have more acceptable thermo - physical characteristics [120].

Compared to pure diesel fuel, direct usage of biodiesel or biodiesel–alcohol mixes in diesel engines results in a loss of power. This is because Biodiesel and Biodiesel–alcohol blends have lower calorific values [121]. The calorific values of CH_3OH, C_2H_6O, C_3H_8O, C_4H_9OH, $C_5H_{11}OH$, and diesel fuels are as follows: The findings were 21.63 MJ/kg, 27.75 MJ/kg, 30.19 MJ/kg, 33.06 MJ/kg, 34.25 MJ/kg, and 44.6 MJ/kg. According to IS 1856 standards, blending diesel with biodiesel produced from jatropha, legumes, neem, and pine seed reduces the biodiesel's kinematic viscosity, pour point, and cloud point [122]. In that order, CH_3OH, C_2H_6O, C_3H_8O, C_4H_9OH, and $C_5H_{11}OH$ contribute 51.73 percent, 36.544 percent, 38.764 percent, 21.88 percent and 19.64 percent of the total oxygen content respectively. Increasing oxygen availability is the most critical factor impacting total production [123].

5.3. Impact of nano additives on engine emission characteristics

Academic researchers have recently focused on combustion quality improvements to reduce the amount of exhaust pollutants produced by conventional diesel engines using nanoparticle-loaded diesel–biodiesel fuel blends [117]. Because biodiesel–alcohol contains more oxygen, it burns at a higher temperature, releasing more nitrogen oxides into the atmosphere. These levels may be as much as 25% higher. Compared to diesel fuel, it decreases carbon monoxide emissions by up to 45 percent, hydrocarbon generation by 98 percent, and particulate matter emissions by about 85 percent [124]. According to a research study using a Ford V6 diesel engine running at full load and variable speed, the amount of hydrocarbon and carbon monoxide emissions, heat release rate, and in-cylinder pressure increased as the ethanol to gasoline ratio grew. Carbon dioxide and nitrates of oxides concentrations, on the other hand, are significantly reduced, oxidation time is decreased, and ignition delay is increased. As a result of these changes, the ignition delay has risen [125].

Adding methyl ester to palm-based biodiesel influenced the efficiency, ignition, and exhaust gas emissions of a direct injection-compression diesel engine that also ran on conventional biodiesel mixed with alcohol [126]. This experiment was carried out on a diesel engine with direct injection and compression ignition. Compared to regular biodiesel, the results revealed a slight rise in the quantity of gasoline used and improved brake thermal efficiency. The impact is increased when methanol is added to the mix [127]. Furthermore, the alcohol biodiesel blend dramatically increases the gasoline necessary to operate the engine. The temperature of the exhaust smoke produced by using either mixed biodiesel with ethanol and methanol or pure biodiesel is not significantly different. Compared to pure biodiesel combustion, the pressure rise generated by alcohol-blended biodiesel combustion is significantly smaller [128]. Nitrogen oxide emissions decrease by 35 and 23 percent, respectively, when biodiesel is blended with 18 percent ethanol and 22 percent methanol by volume, compared to pure biodiesel alone [129]. Even though methanol has no apparent influence on carbon dioxide emissions, there is a little reduction in Carbon-di-oxides. Following that, the particulate matter and Smoke discharge of the Alcohol blended biodiesel are significantly greater than that of pure Biodiesel. Table 8 shows the various nanoparticles' effects on engine emissions [130].

At a constant speed of 1900 rpm, an engine running on diesel and Biodiesel with 10%, 20%, 30%, and 50% ester blends is tested to see how different mixtures burn and how much particulate matter is released into the exhaust [131]. The ethanol injection enhances the diesel engine's high heat rate and cylinder pressure [132]. The effects of combining n-butanol and n-propanol in a diesel engine were investigated using an ER-8S CI diesel engine. However, the percentages of butanol and pentanol used in the experiment were 15% and 30%, respectively. Compared to a biodiesel-fueled engine, butanol-biodiesel blends improve brake thermal efficiency by 3.8 percent and brake fuel economy by 2.75–4.62 percent at medium and low engine speeds, respectively [133]. These effects on combustion parameters and the physiochemical aspects of particulate matter discharges were detected

Table 8
Impacts of various nanoparticles on engine emissions

S.No	Fuel Type	Nano-Particles	Engine emission CO (ppm)	NO_X (ppm)	PM	CO_2 (kg)	HC (ppm)	Ref
1.	Diesel	CNT	188	348	9.6	2.4	79	[58]
2.	Mauha Biodisel	Al_2O_3	84	287	8.4	2.1	43	[62]
3.	Jatropha Biodisel	TiO_2	93	282	8.5	1.6	32	[73]
4.	Thumba Biodeisel	$Al_2O_3 + CeO_2$	76	264	7.6	1.89	34	[78]
5.	Waste cooking oil	Ferrous Fluid	78	257	7.25	1.68	38	[92]
6.	HVO Biodiesel	Rh_2O_3	43	142	6.42	1.3	21	[97]
7.	Ricebran Biodiesel	CeO_2	81	298	6.83	2.1	18	[103]
8.	Rapeseed Biodiesel	MWCNTs	97	234	7.35	2.4	22	[105]
9.	Linseed Biodiesel	$Rh_2O_3 + TiO_2$	96	248	7.21	2.3	34	[126]
10.	Algae Biodiesel	Graphene oxide	51	230	6.5	1.8	23	[128]

when the engines were run at a constant speed and under three different load conditions. Increased brake thermal efficiency resulted in a 4% increase in specific fuel consumption and a reduction in particulate matter and elemental carbon emissions [134]. Table 8 highlights the effect of various nanoparticles on engine emissions.

5.4. Impacts of Nanoparticles on human health

In their most basic form, nanoparticles are powders or colloids with a substantially greater surface area than bigger particles. As a result of recent advances in nanoparticle production, Nanoparticles are increasingly employed in the energy industry. Because of their properties, nanoparticles are great candidates for various applications. In general, due to the smaller size of the nano-particles, the accidental inhalation by human may cause, lung inflammation, heart problems, allergic effects, DNA/RNA alternation damage etc. Studies in humans show that metal based nano-particles poses toxicity and affects the cardiovascular system. However, detailed analysis on the nano-particle emissions on human health is needed which is beyond the scope of the present study.

6. Conclusion and Future Perspectives

Plant-based nanoparticles' impressive physical, chemical and environmental characteristics have motivated researchers to use them in various applications, including fuels. This paper demonstrates the applications of different plant-based nanoparticles for fuel properties enhancement. The following is a summary of the main conclusions from this review.

It is important to develop simple, affordable, environmentally safe plant-based catalysts for the synthesis of renewable fuels from non-edible biomass feedstocks sources. The desirable qualities of plant-based nano-additives in biofuels, such as abundance, low cost, stability, good oxidation etc., are of significant importance.

The biocompatibility, renewable and eco-friendly aspects of the nature-based nano-additives pose a significant advantage over metallic-derived nano-particles. The unique thermal conductivity and rheological properties of nano-additives have attracted scientists to use them in automotive fuels.

Using nano-additives in automotive engines has improved ignition quality, increased output, cetane number, reduced toxic smoke and harmful pollutants. Its viability and potential in CI engines are due to the improved combustion behavior. In general, the efficiency, emission, and combustion characteristics of diesel engines have been improved with nano-additives.

However, several researchers have reported the problem of un-burnt nanoparticles in the exhaust, which may affect the smooth operation. Further, research on eliminating un-burnt nano-additives from the exhaust system must be carried out.

In the future, plant-based nano-additive will have large-scale applications in the fuel sector, overcoming the various commercial and technological challenges.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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