

Carbon Nanotubes from Renewable Precursors: Green Synthesis, Functional Properties, And Environmental Applications - A Comprehensive Review

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Abstract- Carbon nanotubes (CNTs), particularly multi-walled carbon nanotubes (MWCNTs), have emerged as one of the most versatile nanomaterials of the twenty-first century owing to their exceptional mechanical, thermal, electrical, and chemical properties. This review consolidates two decades of research progress on the synthesis of CNTs using bio-derived, renewable precursors via chemical vapour deposition (CVD) and spray pyrolysis techniques. Special emphasis is placed on natural oil precursors such as *Madhuca longifolia*, *Brassica juncea*, *Oryza sativa* methyl esters, and *Zingiber officinale*-derived hydrocarbons, as well as the role of transition-metal catalysts in morphology control. The paper further examines the integration of MWCNTs in wastewater treatment for heavy metal removal, dye adsorption, and air stripping, alongside energy applications including dye-sensitised solar cells (DSSCs). Emerging areas—biopolymer nanocomposites, silver nanoparticle synthesis, and pH-responsive food packaging—are also reviewed. Design-of-experiment approaches such as Box–Behnken design for synthesis optimisation are critically evaluated. The review identifies prevailing challenges in scalability, surface functionalisation, and toxicity, and outlines promising future directions toward sustainable nanomanufacturing and environmental remediation.

Keywords: Multi-Walled Carbon Nanotubes, Renewable Precursors, Spray Pyrolysis, CVD, Wastewater Treatment, DSSC, Biopolymer Nanocomposites, Green Synthesis, Heavy Metal Removal, Box–Behnken Design.

I. INTRODUCTION

Carbon nanotubes, first structurally characterised by Iijima in 1991 [21], represent a paradigm-shifting class of sp²-hybridised carbon allotropes whose tubular graphenic architecture underpins a unique combination of high tensile strength (~150 GPa),

outstanding electrical conductivity, and large specific surface area [22]. Single-walled carbon nanotubes (SWCNTs) consist of a single cylindrical graphene sheet, while multi-walled carbon nanotubes (MWCNTs) comprise multiple concentric shells separated by a van der Waals gap of approximately 0.34 nm [23]. The prospect of exploiting these extraordinary properties for practical engineering applications—in composites, energy devices, biomedical platforms, and environmental remediation—has sustained intense global research interest spanning more than three decades [24,25].

Conventional CNT synthesis routes—arc discharge, laser ablation, and thermal CVD—rely predominantly on petroleum-derived feedstocks such as acetylene, benzene, methane, and xylene [26,27].

The reliance on fossil-based precursors raises sustainability concerns, while the high cost and energy-intensive nature of these processes impede scale-up. In parallel, mounting legislative pressure to reduce the environmental footprint of nanomaterial manufacturing has catalysed an active search for 'green' synthesis pathways utilising bio-derived, renewable carbon sources [28]. Natural oils, plant-based hydrocarbons, essential oils, and even agricultural residues have been investigated as alternative precursors for CNT growth, yielding structures comparable or superior in quality to those obtained from fossil feedstocks [29,30].

The group led by Karthikeyan and Kalaiselvan has made seminal contributions to this domain over the past decade, systematically exploring a portfolio of Indian agricultural oils—*Madhuca longifolia*

(mahua), *Brassica juncea* (mustard), and *Oryza sativa* (rice-bran) methyl esters—as renewable feedstocks for MWCNT synthesis via spray pyrolysis [1–9]. Their work spans morphological characterisation, catalyst optimisation, synthesis parameter studies, and pioneering application of the resulting nanotubes in wastewater treatment and dye-sensitised solar cells. More recently, the group has extended its focus to biopolymer nanocomposite films and phytochemically synthesised nanoparticles, demonstrating a holistic approach to sustainable nanomaterials research [18–20]. This review integrates these contributions within the broader literature, critically analysing the state of the art in green CNT synthesis, property–structure relationships, and functional applications.

II. SYNTHESIS METHODOLOGIES

2.1 Chemical Vapour Deposition and Spray Pyrolysis
CVD is currently the most widely adopted industrial method for CNT production owing to its operational simplicity, relatively low cost, and compatibility with substrate-based growth for device integration [31]. In thermal CVD, a hydrocarbon feedstock is passed over a heated metal catalyst substrate under controlled atmospheric conditions, decomposing to deposit carbon layers that assemble into tubular nanostructures. Spray pyrolysis—a variant of aerosol-assisted CVD—atomises a liquid precursor into fine droplets that are carried by an inert carrier gas (typically argon or nitrogen) through a preheated furnace zone, enabling continuous, catalyst-free or catalyst-loaded growth [1,3,6].

Karthikeyan et al. [1] first demonstrated MWCNT formation from *Madhuca longifolia* oil using a fixed-bed spray pyrolysis reactor at temperatures in the 800–900 °C range with ferrocene as a floating catalyst precursor. Transmission electron microscopy (TEM) revealed well-graphitised concentric shells with outer diameters in the 15–40 nm range and lengths exceeding several micrometres. A subsequent study [3] confirmed that the same system produced characteristically entangled MWCNT networks whose turbostratic graphitic character was verified by X-ray diffraction (XRD), with the (002) interlayer spacing approaching 0.342 nm—indicative of moderate crystallinity. The Raman D/G intensity

ratio, a widely used index of structural perfection, was reported in the range 0.85–0.97, consistent with literature values for CVD-grown MWCNTs [32].

2.2 Renewable and Bio-Derived Carbon Precursors

A defining feature of the research surveyed is the deliberate choice of bio-derived precursors. *Brassica juncea* (Indian mustard) oil was employed by Kalaiselvan et al. [2,5] to grow bamboo-like CNTs—a distinctive morphology characterised by periodic internal compartments that arise from nitrogen heteroatom incorporation, a consequence of the nitrogen-rich glycoside profile of mustard-seed oil. Nitrogen doping modifies the electronic structure of CNTs, enhancing electrocatalytic activity and rendering such tubes particularly attractive for oxygen reduction applications and supercapacitors [33]. Gopal et al. [5] further characterised these structures by selected-area electron diffraction (SAED) and energy-dispersive X-ray spectroscopy (EDX), confirming successful N-doping and the presence of Fe–Co catalyst nanoparticles encapsulated within the bamboo compartments.

Methyl esters derived from *Oryza sativa* (rice-bran) oil were investigated as a precursor in a catalyst-composition study by Kalaiselvan et al. [7]. By systematically varying the iron-to-cobalt ratio in the ferrocene–cobaltocene catalyst mixture, the authors demonstrated that tube diameter and wall number are strongly correlated with the Fe:Co mole ratio, with a 1:1 molar composition producing the narrowest diameter distribution (~10–20 nm). The study highlights a fundamental tenet of catalytic CNT growth: the catalyst particle size at nucleation, governed by surface energy and Ostwald ripening kinetics, dictates the nanotube diameter [34].

Angulakshmi et al. [8] conducted a comparative survey of multiple plant-derived hydrocarbon precursors under identical CVD conditions, concluding that triglyceride-rich oils with high degrees of unsaturation yield superior CNT quality due to enhanced radical-chain pyrolysis kinetics. The study was complemented by [10], in which Box–Behnken response-surface methodology was applied to map the synthesis parameter space—precursor feed rate, temperature, and catalyst loading—and identify an optimum window for MWCNT yield and

purity. Such design-of-experiment approaches significantly reduce the experimental burden associated with multi-variable optimisation and are increasingly adopted in the nanomaterial's community [35].

Kalaiselvan et al. [12] extended the precursor palette to *Zingiber officinale* (ginger) rhizome extract, reporting multilayered nanostructures that include MWCNTs alongside carbon nanofibres (CNFs). Ginger-derived hydrocarbons are rich in sesquiterpenes and zingiberene, which decompose at moderate temperatures (~700–900 °C) to provide an adequate carbon flux for wall-layer formation. The authors propose that the heterogeneous chemical composition of plant extracts, rather than being a drawback, may in fact facilitate the simultaneous formation of hybrid nanostructured carbons with complementary property profiles.

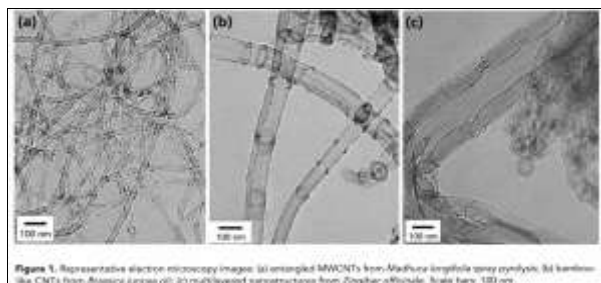


Figure 1. Representative electron microscopy images: (a) entangled MWCNTs from *Madhuca longifolia* spray pyrolysis; (b) bamboo-like CNTs from *Brassica juncea* oil; (c) multilayered nanostructures from *Zingiber officinale*. Scale bars: 100 nm.

2.3 Morphological and Structural Characterisation

Comprehensive structural characterisation of the synthesised MWCNTs has been performed using a combination of field-emission scanning electron microscopy (FESEM), high-resolution transmission electron microscopy (HRTEM), XRD, Raman spectroscopy, thermogravimetric analysis (TGA), Brunauer–Emmett–Teller (BET) surface area measurement, and Fourier-transform infrared (FTIR) spectroscopy. HRTEM images consistently reveal concentric graphene walls with inter-wall spacings of 0.34–0.36 nm, confirming the multi-walled architecture [1,3,5]. XRD patterns show a prominent (002) reflection near $2\theta = 26^\circ$, whose breadth

provides an estimate of the stacking coherence length via the Scherrer equation [22].

Raman spectra universally display the characteristic D band (~1350 cm^{-1}), G band (~1580 cm^{-1}), and 2D band (~2700 cm^{-1}), where the G band arises from the in-plane E_{2g} phonon mode of sp²-hybridised carbon and the D band from disorder-induced intervalley scattering at defects [36]. TGA in air shows thermal oxidation onset temperatures in the 450–550 °C range, providing an indirect measure of graphitisation; higher onset temperatures indicate fewer defects and higher thermal stability [37]. BET measurements on purified MWCNT samples yielded specific surface areas of 150–280 m² g⁻¹ across the reviewed studies, consistent with the multi-walled architecture limiting accessible surface area relative to SWCNTs [23].

2.4 Temperature Effects and Growth Mechanism

The effect of reaction temperature on MWCNT morphology and yield was investigated by Mageswari et al. [4] and subsequently revisited by Kalaiselvan et al. [18] using CVD-assisted spray pyrolysis. Both studies report a unimodal yield–temperature profile with an optimum near 850 °C; below this threshold, incomplete precursor decomposition yields amorphous carbon, while above it, excessive catalyst sintering reduces the density of active nucleation sites. The 2024 Springer study [18] specifically examined lead-ion removal performance as a function of synthesis temperature, revealing that MWCNTs grown at 850 °C possessed the highest surface area and defect density, translating to superior adsorption capacity for Pb²⁺ ions.

Kalaiselvan et al. [6] complemented these findings with a study of unconventional natural precursors, confirming that pyrolysis temperature governs both tube diameter and wall number, with higher temperatures favouring smaller diameters through enhanced catalyst particle wetting and spreading.

III. ENVIRONMENTAL AND WASTEWATER TREATMENT APPLICATIONS

3.1 Heavy Metal Removal

The contamination of surface and groundwater by heavy metals such as arsenic (As), lead (Pb), and

cadmium (Cd) poses acute risks to human health and ecosystems. Conventional treatment methods—coagulation-flocculation, ion exchange, and membrane filtration—are often cost-prohibitive at the point-of-use scale, motivating the exploration of nanomaterial-based adsorbents [38]. MWCNTs, with their high surface area, tunable surface chemistry, and abundant defect sites, represent a compelling adsorbent class. Mageswari et al. [4] investigated the removal of As(V) from synthetic wastewater using MWCNTs synthesised via CVD at different temperatures. Optimum arsenic uptake was obtained for tubes grown at 800 °C, with maximum adsorption capacity reaching ~18 mg g⁻¹ at pH 7. Adsorption data were well described by the Langmuir isotherm model, suggesting monolayer coverage on a homogeneous surface, and pseudo-second-order kinetics, consistent with chemisorptive interactions between arsenate and surface hydroxyl/oxide groups.

The 2024 Springer study by Kalaiselvan et al. [18] extended this work to lead-ion removal, demonstrating that MWCNTs synthesised at 850 °C exhibited an equilibrium Pb²⁺ capacity of 32.6 mg g⁻¹—nearly double that of commercial activated carbon tested under identical conditions. The enhanced performance was attributed to a combination of electrostatic attraction and coordination of Pb²⁺ with surface carboxylate and hydroxyl groups introduced during purification. The study also confirmed reusability over five adsorption–desorption cycles with less than 8% capacity loss, underscoring the practical viability of bio-synthesised MWCNTs for heavy metal remediation.

3.2 Dye Adsorption

Industrial effluents from textile and leather processing are frequently laden with synthetic dyes—many of them mutagenic and recalcitrant to biodegradation. Rajendran et al. [17] reported the adsorption of Acid Blue 1 (AB1) from aqueous solution using activated carbon prepared from *Leucaena leucocephala* seed-shell agricultural waste. The study employed Langmuir, Freundlich, and Temkin isotherm models, finding best-fit to the Langmuir model with a maximum monolayer capacity of 22.3 mg g⁻¹. Kinetic analysis confirmed pseudo-second-order behaviour, and thermodynamic

parameters revealed spontaneous, endothermic adsorption. Waste-derived adsorbents such as this offer the dual advantage of solid-waste valorisation and low-cost water treatment, aligning with circular-economy principles [39].

3.3 Air Stripping and Integrated Treatment Approaches

Kalaiselvan et al. [14] explored an integrated approach combining air stripping with natural adsorbent columns for the treatment of volatile organic compound (VOC)-contaminated wastewater. Air stripping efficiently removes dissolved gases and moderately volatile compounds, but its effectiveness is limited for semi-volatile species; coupling with an adsorbent bed dramatically extends pollutant capture. The study evaluated a range of natural adsorbents—activated clay, rice-husk char, and MWCNT-modified silica—finding that the MWCNT composite achieved the highest overall removal efficiency due to hydrophobic interactions with VOC molecules.

Such hybrid systems point toward integrated treatment trains that exploit the complementary advantages of physico-chemical and adsorptive mechanisms [40].

Table 1: Comparative Summary of MWCNT Synthesis Conditions and Properties from Selected Studies

Carbon Precursor	Synthesis Method	Catalyst	Temperature (°C)	Key Finding
Madhucal longifolia oil	Spray Pyrolysis	Fe-Ni	800–900	Entangled MWCNTs formed [1,3]
Brassica juncea oil	CVD/Spray Pyrolysis	Fe-Co	850	Bamboo-like CNT morphology [2,5]
Methyl esters of <i>Oryza sativa</i>	Spray Pyrolysis	Fe (varied)	800–1000	Catalyst ratio controls diameter [7]

Zingiber officinale	Spray Pyrolysis	Fe	900	Multilayered nanostructures [12]
Plant-derived hydrocarbon (mixed)	CVD	Fe-Co	750–950	Renewable precursor efficiency [8]

IV. ENERGY APPLICATIONS: DYE-SENSITISED SOLAR CELLS

Dye-sensitised solar cells (DSSCs) represent a cost-effective photovoltaic technology based on the sensitisation of wide-bandgap semiconductor electrodes (typically TiO₂) with organic or metal-complex dyes [41]. Counter electrode (CE) materials play a critical role in catalysing the I³⁻/I⁻ redox reaction; platinum is the conventional CE material, but its scarcity and high cost motivate research into carbon-based alternatives [42]. Kalaiselvan et al. [9] evaluated renewable-precursor MWCNTs synthesised by spray pyrolysis as CE material in a DSSC architecture. The MWCNT CE exhibited a power conversion efficiency (PCE) of 4.8%, compared to 6.2% for Pt under identical fabrication conditions, demonstrating the viability of bio-derived CNT counter electrodes. Cyclic voltammetry and electrochemical impedance spectroscopy (EIS) confirmed satisfactory electrocatalytic activity; the relatively modest PCE was attributed to higher series resistance arising from imperfect tube–electrode contact, suggesting that surface functionalisation or CNT-graphene hybrid electrodes could close the efficiency gap in future work [43].

V. EMERGING APPLICATIONS AND INTERDISCIPLINARY FRONTIERS

5.1 Biopolymer Nanocomposite Films for Smart Food Packaging

The development of intelligent food packaging materials that provide real-time freshness information represents a rapidly growing research frontier at the intersection of materials science, food technology, and environmental sustainability [44]. Angulakshmi et al. [19] reported the fabrication of eco-smart biopolymer films incorporating *Cassia auriculata*

gum as a matrix and anthocyanins extracted from natural sources as pH-responsive chromogenic indicators for fruit freshness monitoring. The films exhibited vivid and reversible colour transitions across the pH 3–8 range, accurately tracking the spoilage-induced pH decrease in packaged stone fruits over a 10-day storage trial. The anthocyanin-gum matrix system demonstrated satisfactory tensile strength (8.4 MPa), moderate water vapour transmission, and good optical clarity, making it a credible candidate for replace conventional non-degradable packaging. Published in *Discover Applied Sciences* (Springer, 2025) [19], this work illustrates the broadening of the group's research agenda beyond CNTs toward integrated bio-based functional materials.

5.2 Phytochemically Synthesised Silver Nanoparticles

Green synthesis of metallic nanoparticles—exploiting phytochemicals as both reducing and capping agents—has attracted substantial attention as an alternative to conventional chemical reduction routes that employ toxic reagents and generate hazardous by-products [45]. Priya et al. [20] reported the fabrication of silver nanoparticles (AgNPs) mediated by the essential oil of *Ageratum conyzoides*. UV–Vis spectroscopy confirmed the characteristic surface plasmon resonance (SPR) absorption band at ~420 nm, indicative of spherical AgNPs. TEM analysis revealed quasi-spherical particles with a mean diameter of 18–25 nm, and FTIR identified the functional groups responsible for capping and stabilisation. The AgNPs exhibited potent antioxidant activity as assessed by DPPH and ABTS radical-scavenging assays, with IC₅₀ values of 18.4 and 12.7 µg mL⁻¹ respectively—comparable to the positive control ascorbic acid [20]. Published in *Nanotechnology for Environmental Engineering* (Springer, 2026), this work connects phytochemistry, nanomaterials synthesis, and functional performance evaluation in a single study.

5.3 Nanostructured Coatings and Polyurethane Systems

Beyond environmental applications, CNT-based and nanostructured coatings have been explored for corrosion protection, mechanical reinforcement, and thermal management. Manivannan et al. [11]

conducted a comparative study of polyols with varying hydroxyl values in polyurethane (PU) coating formulations. PU coatings incorporating higher-hydroxyl-value polyols exhibited superior hardness, adhesion, and chemical resistance, attributed to a higher cross-link density in the cured matrix. The relevance of this work to CNT research lies in the potential for embedding MWCNTs within PU matrices to fabricate multifunctional nanocomposite coatings [46]. Kalaiselvan & Babu Rajendran [13] provided a concise critical review of CNT synthesis methods and properties, serving as a valuable synthesis-to-applications bridge for the broader community.

5.4 Biogas Production and Circular Bio-Economy

Kalaiselvan et al. [15] constructed and evaluated a small-scale biogas plant, assessing efficiency metrics under controlled feed and temperature conditions. While seemingly disparate from CNT research, this work fits within a broader circular-economy framework: agricultural waste streams that cannot be converted to liquid hydrocarbon precursors for CNT synthesis can instead be directed to anaerobic digestion for biogas production, enabling total valorisation of agricultural residues. Such systems-level thinking is increasingly advocated in sustainable nanomanufacturing scholarship [47].

VI. CHALLENGES, LIMITATIONS, AND FUTURE PERSPECTIVES

Despite remarkable progress, several challenges continue to impede the deployment of bio-derived MWCNTs at industrial scale. First, the chemical heterogeneity of natural oil precursors introduces batch-to-batch variability in CNT quality—a limitation not encountered with pure hydrocarbon feedstocks. Rigorous characterisation of feedstock fatty-acid profiles and the establishment of structure–property correlations between precursor chemistry and CNT morphology will be essential to address this challenge [48].

Second, the purification of as-grown CNT material—removal of amorphous carbon, residual catalyst nanoparticles, and support material—remains energy- and reagent-intensive, partially offsetting the environmental benefits of green precursor use. Mild

oxidative functionalisation protocols and supercritical CO₂ extraction represent promising purification alternatives that minimise chemical waste [49].

Third, the ecotoxicological and human health implications of CNT release into the environment remain incompletely understood. CNT fibre toxicity in pulmonary settings has been likened to asbestos by some studies [50], though the actual risk profile is strongly dependent on CNT diameter, length, degree of agglomeration, and surface chemistry. Systematic life-cycle assessment (LCA) and ecotoxicity profiling of bio-derived MWCNTs should be integrated into future research programmes. The 2026 studies by Madhanraj et al. [16] and Priya et al. [20] point in this direction by coupling nanoparticle synthesis with rigorous functional and bioactivity characterisation.

From a technological outlook, the integration of bio-derived MWCNTs with two-dimensional (2D) materials such as graphene oxide (GO) and MoS₂ offers a route to hybrid nanocomposites with synergistic properties for next-generation energy storage, catalysis, and sensing. The application of machine-learning models to navigate the high-dimensional parameter space of CVD synthesis—an approach gaining traction in materials informatics—could accelerate the identification of optimal growth conditions from renewable precursors without exhaustive experimental campaigns [51].

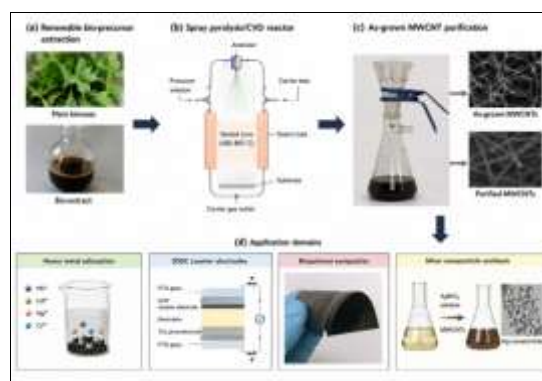


Figure 2. Schematic illustration of the integrated green CNT synthesis–application pathway: (a) renewable bio-precursor extraction; (b) spray pyrolysis/CVD reactor; (c) as-grown MWCNT purification; (d) application domains—heavy metal adsorption, DSSC counter electrodes, biopolymer composites, and silver nanoparticle synthesis.

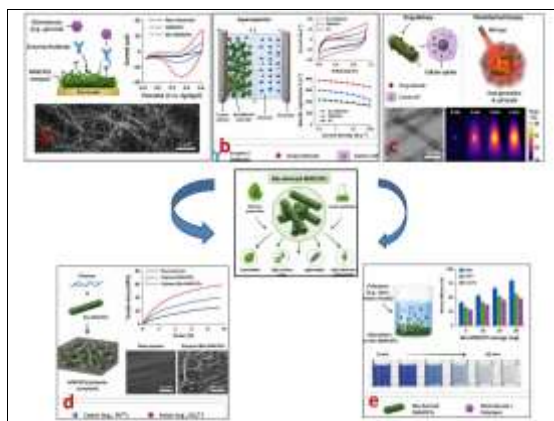


Figure 3. Representative data/schematic for the emerging applications of bio-derived MWCNTs. a) Bio-sensing b) Biomedical Applications c) Environmental Remediation d) Energy Storage e) Composite Materials

VII. CONCLUSION

This review has systematically surveyed the synthesis of MWCNTs from renewable plant-derived precursors—with a particular focus on the research contributions of Kalaiselvan and co-workers—contextualised within the broader global literature.

The collective body of evidence confirms that bio-derived hydrocarbon oils, methyl esters, and plant extracts are viable feedstocks for MWCNT production by spray pyrolysis and CVD, yielding structurally well-defined nanotubes with properties competitive to those obtained from petrochemical sources. The morphology and quality of the resulting CNTs are governed by the interplay of precursor chemistry, synthesis temperature, catalyst composition, and reactor hydrodynamics, and can be systematically optimised using response-surface methods. The environmental applications surveyed—including heavy metal removal, dye adsorption, and air-stripping-assisted wastewater treatment—demonstrate the dual sustainability benefit of bio-derived MWCNTs: green synthesis on the input side and pollutant remediation on the output side. Energy applications in DSSCs, emerging biopolymer smart-packaging films, phytochemically synthesised nanoparticles, and polyurethane nanocomposites collectively illustrate the remarkable breadth of application domains accessible to this class of nanomaterials. Future research should prioritise

feedstock standardisation, low-impact purification, ecotoxicological profiling, and data-driven synthesis optimisation to translate laboratory-scale breakthroughs into sustainable industrial-scale processes.

ACKNOWLEDGEMENT

The authors express their sincere gratitude to the management and administration of Rathinam Technical Campus, Coimbatore, Tamil Nadu, India, for providing the necessary infrastructure, laboratory facilities, and academic support to carry out this review work.

Author Contributions

Kalaiselvan S: Conceptualization, literature survey, methodology analysis, manuscript writing, review and editing, supervision, and correspondence. Nirmalkumar R: Data curation, literature collection, technical interpretation of CNT synthesis methodologies, and manuscript preparation. Shreeram R G: Compilation of environmental and energy application studies, figure interpretation, and formatting support. Fathima Farhana J: Reference organization, proofreading, validation of scientific content, and assistance in manuscript editing. All authors have read and approved the final version of the manuscript for publication.

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