Double Resource Recovery from Plastic Waste: Development and Characterisation of an Eco-Friendly Carbon-Based Ink from Plastic Pyrolysis-Derived Carbon and Recycled Tea Leaf Extract for Sustainable Artistic Applications

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ABSTRACT

The growing global plastic usage results in the enormous accumulation of non-biodegradable plastic garbage, causing serious environmental, ecological and socio-economic issues. Pyrolysis has become an efficient thermochemical technique to convert plastic waste to liquid fuels and combustible gases, but the carbonaceous solid byproduct created in the process is often underappreciated, despite its large material potential. Simultaneously, the production of conventional carbon black-based inks and watercolour pigments depends largely on the usage of petroleum-derived raw materials and energy-intensive industrial processes, resulting in the emission of greenhouse gases, depletion of fossil resources and contamination of the environment. At the same time, millions of tonnes of spent tea leaves produced globally are disposed of as organic waste, regardless of the fact that they contain naturally occurring tannins, polyphenols and colouring compounds that can be utilised as sustainable additives in pigment compositions. Our previous work successfully demonstrated the production and structural characterisation of plastic-derived pyrolytic carbon via an oxygen-limited pyrolysis process. It was established that the material obtained possesses a predominantly amorphous turbostratic carbon structure with potential applications in environmental remediation and functional carbon materials[1]. In this study, expanding on earlier work, a new value-added application of the recovered carbon is explored by generating an eco-friendly carbon-based ink using plastic pyrolysis-derived carbon and recycle dused tea leaves. The current work was focused on thermochemical conversion of plastic packaging waste in a lab-scale cylindrical metallic reactor under limited oxygen circumstances. The resulting pyrolytic carbon was collected, cleaned, dried and ground to a fine powder. Approximately 10 g of the generated pyrolytic carbon was combined with 10 g of recycled old tea leaves, which were processed to obtain a tannin-rich extract that was employed as a natural colour modifier and dispersing medium. The formulated formulation was characterised by Fourier Transform Infrared Spectroscopy (FTIR), X-ray Diffraction (XRD), pH and electrical conductivity. The ink as prepared had an acidic pH of 3.0, which was attributed to the presence of naturally existing acidic functional groups from the tea polyphenols and oxygen functionalities on the surface of the pyrolytic carbon. The measured conductivity of 10.42 µS/cm showed the existence of mobile ionic species in the dispersion that indicates the good electrochemical interaction between the dispersed carbon particles and the aqueous medium. XRD analysis showed that the carbon was mainly amorphous turbostratic with a low degree of graphitic order. FTIR analysis indicated the presence of hydroxyl, carbonyl, aromatic C=C and C–O functional groups responsible for the improved pigment dispersion and interaction with the natural tea extract. The integrated physicochemical characterisation indicates that the developed formulation has appropriate qualities for eco-friendly creative applications such as watercolour painting, brush painting, calligraphy, educational art materials and sustainable carbon-based pigments.

The presented technology handles two main waste streams, i.e., plastic garbage and used tea leaves, concurrently and transforms them into a usable carbon-based artistic material. The created approach contributes to the circular economy principles by valorising waste, decreasing the dependence on petroleum-derived pigments, and fostering sustainable material innovation for educational, artistic, and environmental purposes.

Keywords: Plastic waste; Pyrolysis; Pyrolytic carbon; Sustainable ink; Carbon pigment; Tea leaf extract; Water colour; Circular economy; Waste valorisation; FTIR; XRD; Eco-friendly materials.

INTRODUCTION

Background

The twenty-first century has experienced an unparalleled rise in the generation and utilisation of synthetic plastics due to their economical production, lightweight nature, superior mechanical durability, resistance to chemicals, and straightforward processing. Since the mid-twentieth century, the commercialisation of plastics has led to a persistent rise in global production, surpassing hundreds of millions of tonnes per year. While plastics have transformed packaging, healthcare, transportation, technology, and consumer goods, their durability in the environment has emerged as a significant global ecological issue. Traditional plastics like polyethylene (PE), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET) necessitate hundreds of years for natural degradation, leading to significant buildup in landfills, waterways, and oceanic environments [2–5].

The rising production of plastic garbage has prompted much investigation into sustainable waste management solutions. Mechanical recycling is the most prevalent technique; yet, successive recycling frequently compromises polymer integrity, and contamination along with heterogeneous plastic mixtures considerably diminishes recycling efficacy [6]. Incineration facilitates energy recovery; however, it emits greenhouse gases and may discharge harmful pollutants. The practice of landfilling utilises precious land resources and may lead to prolonged environmental pollution [3,6].

Among the nascent waste-to-resource technologies, pyrolysis has garnered significant interest due to its capability to transform waste polymers into various high-value products in oxygen-restricted environments. In the process of pyrolysis, extensive polymer chains experience heat degradation, producing liquid hydrocarbons, flammable fumes, and a carbon-dense solid byproduct known as pyrolytic carbon or pyrolysis char [7,8]. In our latest research, we experimentally verified the generation of pyrolytic carbon from discarded plastic packaging utilising a laboratory-scale cylindrical reactor, and through X-ray diffraction analysis, we confirmed that the resultant material primarily comprises amorphous turbostratic carbon with notable structural imperfections, positioning it as a potential precursor for the advancement of functional materials [1].

Global Ink and Water color Industry

Ink is among the earliest synthetic substances utilised by humans and continues to be essential in writing, printing, education, industrial marking, artistic painting, textile printing, and packaging. The worldwide ink sector has undergone significant expansion in the past twenty years, driven by rising demand from the packaging, printing, publishing, education, and fine arts industries. Commercial inks typically have four primary elements: pigments or dyes, binders, solvents, and additives. The pigment dictates the hue and opacity, the binder ensures adherence to the substrate, the solvent regulates viscosity and drying characteristics, while additives enhance storage stability, flow attributes, and resistance to microbial deterioration [2,3].

The worldwide market is presently led by multinational corporations such as Winsor & Newton, Faber-Castell, Sakura Colour Products Corporation, Staedtler, Pilot Corporation, Parker, Daler-Rowney, Royal Talens, Camel (Kokuyo Camlin Ltd.), Pentel, and Pelikan, which produce inks for artistic, educational, industrial and commercial uses [4-8]. These firms mostly utilise synthetic pigments, carbon black sourced from petroleum, organic dyes, acrylic binders, or polymeric dispersants to attain superior colour stability, longevity and consistency.

Watercolour pigments are a significant category within the artistic supplies industry. Conventional watercolour compositions often comprise finely suspended pigments, gum arabic as a natural adhesive, glycerol or honey as

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moisture-retaining agents, along with preservatives and surfactants [9]. Although these goods provide outstanding colour quality, their production heavily relies on mineral pigments, petroleum-based carbon black, synthetic organic dyes and industrial processing methods, necessitating substantial energy and raw material usage.

As environmental consciousness rises, the creative materials sector has started investigating sustainable pigments sourced from biomass, agricultural byproducts, natural minerals, and recycled trash. Nonetheless, the availability of commercially viable eco-friendly black pigments is restricted due to the technological difficulties in attaining sufficient colour intensity, dispersion stability, and long-lasting durability from recovered carbon materials [10].

Environmental Concerns Associated with Conventional Carbon Black Production

Carbon black is one of the most widely used industrial pigments and is the main source of black colour in inks, paints, toners, plastics, rubber products and printing formulations [11]. Commercial carbon black is generally obtained by incomplete combustion or thermal decomposition of heavy petroleum fractions such as coal tar, ethylene cracking tar and residual oils [11,12]. Carbon black has excellent colouring efficiency and chemical stability. However, its production raises several environmental concerns, such as:

1. High consumption of fossil fuels;
2. Significant greenhouse gas emissions;
3. Particulate matter production;
4. Reliance on non-renewable petroleum feedstocks;
5. Energy-intensive high-temperature processing;
6. Occupational exposure to airborne carbon particles [11-13].

In view of the growing scientific interest in renewable or waste-derived carbon materials to substitute part of the conventional carbon black in pigment formulations.

Plastic Waste as an Untapped Carbon Resource

Plastic waste is usually considered an environmental pollutant. But from the point of view of materials science, it is also a valuable carbon-rich resource. Polyethylene, polypropylene and polystyrene are long chains of hydrocarbons, most of which are carbon and hydrogen. These polymers are subjected to thermal cracking to produce volatile hydrocarbons and a solid carbonaceous residue by oxygen-limited pyrolysis [14]. In our previous study, waste plastic packaging materials were successfully converted to pyrolytic carbon using a laboratory-scale cylindrical metallic reactor under oxygen-limited conditions. The XRD structural characterisation indicated that the synthesised carbon was mainly amorphous turbostratic structure with low graphitic ordering and high structural defects, and the carbon showed potential applications in adsorption, environmental remediation, catalyst supports and advanced carbon materials [1]. However, although these characteristics are promising, most of the pyrolysis research is still concentrated on the production of liquid fuels and relatively little attention has been paid to the utilisation of the carbonaceous residue for value-added functional products. This is an important opportunity for the valorisation of waste.

Utilization of Used Tea Leaves

Tea is one of the most popular beverages in the world, and this has resulted in the generation of large quantities of spent tea leaves from households, restaurants, hotels, cafés and food industries [15]. These used tea leaves usually are discarded despite the presence of valuable phytochemicals such as tannins, flavonoids, catechins, polyphenolsandnatural pigments. Tannins have weak binding properties, and have many hydroxyl groups that can interact with carbon surfaces through hydrogen bonding and π–π interactions. These characteristics indicate that tea leaf extract can be used as a sustainable additive for the formulations of carbon pigment by improving the dispersion of the particles and contributing to thedevelopmentofcolour. Thus, the combination of pyrolysis-derived carbon with recycled tea leaf extract provides an opportunity for the simultaneous utilisation of two independent waste streams for the preparation of environmentally sustainable artistic materials.

LITERATURE REVIEW

Studies have been performed on biomass-derived carbonaceous materials such as coconut shells, rice husk, bamboo, agricultural residues, lignocellulosic wastes, and waste plastics for adsorption, energy storage, and catalyst supports [10-16]. Several studies have reported the synthesis of activated carbons from waste plastics by physical or chemical activation, highlighting the improvements in surface area and adsorption performance [12,16].

Similarly, green ink formulations have been investigated using natural pigments derived from plants, flowers, fruits, bark and agricultural waste. Tea polyphenols have been studied as antioxidants, natural colourants and benign environmental additives in different materials [15]. However, a thorough literature review reveals that few studies have investigated the direct conversion of plastic pyrolysis-derived carbon into eco-friendly art inks, using recycled tea leaf extract as a natural dispersing and pigment-modifying agent. Besides, there is no systematic physicochemical characterisation of such formulations by XRD, FTIR, pH and conductivity analyses.

Research Gap

Although considerable research has been done on plastic pyrolysis for fuel production and on activated carbon for adsorption applications, there are still some important research gaps:

1. Most studies on plastic pyrolysis focus on the recovery of liquid fuels, with the carbonaceous residue treated as a secondary by-product.

2. Only limited studies have been carried out on the direct use of pyrolytic carbon as a pigment for artistic ink formulations.

3. Very few reports combine two waste streams, i.e., plastic derived carbon and used tea leaves, to make one value-added material.

4. Most commercial black inks are made from petroleum-based carbon black.

5. Comprehensive structural and physicochemical correlations of XRD, FTIR, pH and conductivity for waste-derived carbon inks are rarely reported. Filling these gaps may contribute to the development of sustainable materials engineering, while supporting the principles of circular economy.

Novelty of the Present Work

The main novelty of the present study is the fabrication of an eco-friendly carbon-based ink prepared from two recycled waste streams, plastic pyrolysis derived carbon and used tea leaf extract. The proposed approach employs, contrasted to the conventional carbon black-based formulations:

• Major carbon source for waste plastics.

• Natural additive rich in polyphenols and tannins from recycled tea leaves.

• A previously developed laboratory-scale pyrolysis process by the authors [1].

• Characterization by multi-technique including XRD, FTIR, pH and conductivity. The study thus shows a sustainable approach to convert waste materials into a functional artistic product with reduced dependence on virgin petroleum-derived pigments.

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Objectives of the Present Investigation

The objectives of the present work are:

- To recover carbonaceous material from waste plastic by oxygen-limited pyrolysis.
- To prepare an eco-friendly carbon-based ink by mixing 10 g of pyrolytic carbon with 10 g of recycled used tea leaves.
- To study the structural properties of the carbon using X-ray diffraction (XRD).
- To characterise the surface functional groups using Fourier Transform Infrared Spectroscopy (FTIR).
- To determine the pH and electrical conductivity of the prepared ink.
- To relate the structural and physicochemical properties to the potential ink suitability for watercolour and artistic applications.
- To present a sustainable waste-valorization approach involving plastic waste and spent tea leaves in a circular economy context.

MATERIALS AND EXPERIMENTAL METHODOLOGY

Materials

In the experimental work, a sustainable carbon ink was developed from waste-derived materials. Mixed plastic packaging waste was chosen as the main carbon source due to its high carbon content and high availability in municipal solid waste . Spent tea leaves (collected after beverage preparation) were used as a natural additive because of the presence of tannins, polyphenols, flavonoids and other oxygen-containing organic compounds which can react with the carbon particles and help in the dispersion of the pigment.

The principal materials used in the investigation are summarized in Table 1. Table 1. Materials Used During the Present Investigation

Material	Specification	Purpose
Waste plastic packaging	Mixed household plastic packaging	Carbon precursor
Laboratory-scale cylindrical metallic reactor	Fabricated closed reactor	Oxygen-limited pyrolysis
LPG gas burner	External heating source	Thermal decomposition
Distilled water	Laboratory grade	Washing and extraction
Pyrolytic carbon	Obtained from plastic pyrolysis	Black pigment
Used tea leaves	Household waste	Natural additive
Mortar and pestle	Laboratory equipment	Pulverization
Drying oven	80–100 °C	Moisture removal
X-ray diffractometer	Cu-Kα radiation	Structural characterization
FTIR spectrometer	Shimadzu	Functional group analysis
Digital pH meter	Laboratory instrument	Acidity measurement
Conductivity meter	Laboratory instrument	Electrical conductivity measurement

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Preparation of Plastic-Derived Pyrolytic Carbon

The carbon material used in this study was prepared by a laboratory-scale pyrolysis process developed previously by the authors [1].

The mixed plastic packaging materials were washed with water to remove surface contaminants and dried under ambient laboratory conditions. The cleaned pieces of plastic were then placed in a sealed metallic cylindrical reactor, specifically designed to work under oxygen-limited conditions. The reactor was heated externally by an LPG burner. The shortage of oxygen limited complete combustion of heating and promoted thermal cracking of polymer chains. The reactor was operated under the experimental conditions described in our previous publication, yielding three main fractions:

• Hydrocarbons in liquid form.

• Gases that do not condense.

• Pyrolysis carbon (carbonaceous solid residue) [1].

After the pyrolysis cycle was over, the reactor was allowed to cool down naturally to room temperature before opening it.

The carbonaceous residue deposited inside the reactor chamber was collected carefully manually. The collected material was crushed with mortar and pestle to reduce the particle size. The crushed carbon was washed several times with distilled water to remove loosely attached ash and soluble impurities and was subsequently dried in a laboratory oven at approximately 80–100 °C until a constant mass was obtained. The obtained black powder was used as the carbon pigment precursor for ink preparation.

Preparation of Used Tea Leaf Extract

Black tea was made, and the tea leaves were collected after use. The collected material was washed gently with distilled water to remove any residual sugar or milk contaminants and then dried at room temperature. The dried tea leaves (about 10 g) were transferred into a beaker with distilled water and heated gently for extraction of soluble phytochemicals. During extraction naturally occurring compounds as:

• tanins

• polyphenols

• flavonoids

• catechines

• water soluble organic acids, released in the aqueous medium, resulted in a dark brown extract. The extract was passed through filter paper to remove coarse solid particles. The filtrate was stored for further ink preparation.

Preparation of Carbon-Based Ink

The general preparation procedure used in the present investigation is schematically shown in Figure 1. First, carefully measured 10 g of the prepared pyrolytic carbon powder. The aqueous extract prepared from 10 g of recycled used tea leaves was separately allowed to cool to room temperature.

The carbon powder was added slowly to the tea extract with constant hand stirring. The suspension was stirred thoroughly until a visually homogeneous black dispersion was obtained. The preparation process did not include any synthetic pigments based on petroleum. The aqueous medium was the tea extract, which also served as a natural source of tannins and polyphenolic compounds that promoted the

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interaction with the dispersed carbon particles. Finally, the prepared suspension was stored in a sealed container for the subsequent physicochemical characterisation.

The complete preparation methodology adopted in the present investigation is illustrated schematically in Figure 1.

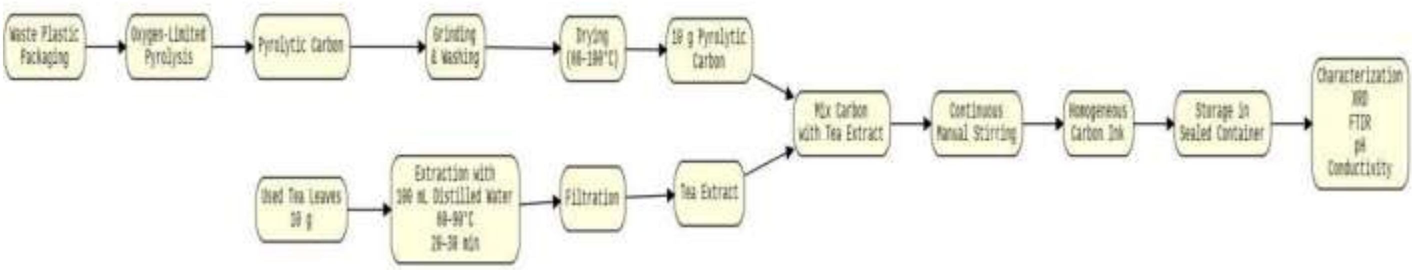

Figure 1. Schematic flowchart illustrating the preparation of the eco-friendly carbon-based ink from plastic pyrolysis-derived carbon and recycled used tea leaves.

Table 2. Composition of the Prepared Ink

Component	Quantity	Purpose
Pyrolytic carbon powder	10 g	Primary carbon pigment
Used black tea leaves	10 g	Source of tannins and polyphenols
Distilled water	100 mL	Extraction medium
Heating temperature	80–90°C	Tea extraction
Extraction time	20–30 min	Release of soluble phytochemicals
Filtration	Whatman filter paper	Removal of coarse particles
Final extract used	Entire filtrate (~80–90 mL)	Ink preparation

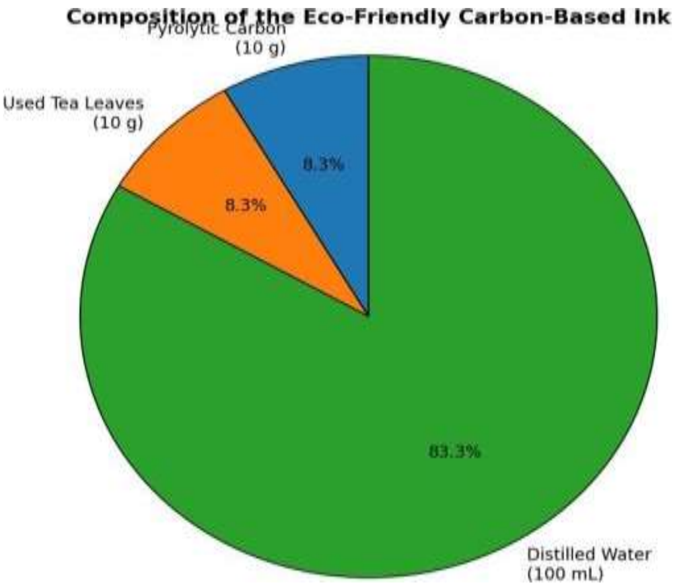

Figure 2. Composition of the eco-friendly carbon-based ink prepared using 10 g of plastic pyrolysis-derived carbon, 10 g of recycled used tea leaves, and 100 mL of distilled water used for extraction of tea-derived phytochemicals**.**

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Proposed Chemical Mechanism

The preparation of the developed ink involves two independent waste valorisation pathways.

Step 1. Thermal decomposition of plastic

During oxygen-limited pyrolysis, polymer chains undergo thermal cracking. For polyethylene, a simplified representation is:

$(C_2H_4)_n\Delta,\ No\ O2$

$\rightarrow\!\!\longrightarrow$ Hydrocarbons + H_2 + CH_4 + C

The elemental carbon generated during repeated thermal rearrangement forms disordered aromatic carbon clusters, which accumulate as pyrolytic carbon.

Step 2. Formation of aromatic carbon

As pyrolysis progresses, small aromatic fragments condense to produce larger carbon domains represented schematically as

$C_xH_y \rightarrow$ Amorphous Carbon

This carbon contains numerous structural defects and oxygen-containing surface groups after exposure to atmospheric oxygen.

Step 3. Tea leaf extraction

Tea leaves contain several naturally occurring compounds including

- tannins
- catechins
- gallic acid
- flavonoids
- polyphenols

These molecules contain multiple hydroxyl (–OH) groups.

Step 4. Interaction between carbon and tea extract

Instead of forming a new covalent compound, the interaction is expected to occur primarily through:

- hydrogen bonding,
- π–π stacking between aromatic structures,
- physical adsorption,
  - van der Waals interactions. A simplified representation is

Carbon Surface–OH ⋯ HO–Polyphenol

These interactions improve dispersion of the carbon particles within the aqueous medium.

Characterization

The prepared carbon material and the developed ink were characterized using multiple analytical techniques.

X- ray Diffraction (XRD)

The carbon powder was analysed using an X-ray diffractometer employing Cu-Kα radiation. The diffraction pattern was collected over a **2θ range of approximately 10°–120°**.

The structural interpretation was performed using Bragg's law:

where

- n is the diffraction order,
- λ is the X-ray wavelength,
- d is the interplanar spacing,
- θ is the diffraction angle.

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR analysis was carried out using a Shimadzu FTIR spectrometer over the spectral range **4000–500 cm⁻¹**.

The objective of the analysis was to identify functional groups present on the carbon surface that could influence ink formation and dispersion.

pH Measurement

The acidity of the prepared ink was measured using a calibrated digital pH meter. The measured pH was**3.0**indicating that the prepared formulation is acidic.

Electrical Conductivity

Electrical conductivity was measured using a laboratory conductivity meter. The measured conductivity was **10.42** μS/cm

The conductivity measurement provides information regarding the mobility of dissolved ionic species within the prepared ink.

Figure 2. Photograph of the eco-friendly carbon-based ink prepared from plastic pyrolysis-derived carbon and recycled tea leaf extract. The formulation exhibits a homogeneous deep black appearance without visible phase

separation immediately after preparation.

RESULTS AND DISCUSSION

Physical Characteristics of the Prepared Carbon-Based Ink

The ink was prepared by dispersing 10 g of carbon derived from plastic pyrolysis in the aqueous extract obtained from 10 g of recycled used tea leaves. Visual assessment of the resultant formulation after manual mixing thorough showed it to be of a deep black colour and homogeneous in appearance. The black colour was mainly due to the finely divided pyrolytic carbon, while the tea extract gave a slight brownish hue due to the naturally occurring tannins and polyphenolic compounds.

The present formulation was developed solely from waste-derived materials, except for the use of petroleum-derived carbon black, synthetic dispersants, and polymeric additives typical of conventional commercial inks. During the short-term observations in this study, the carbon particles were suspended in the tea extract, indicating a satisfactory interaction between the carbon surface and the naturally occurring organic constituents present in theteaextract.

The developed formulation shows that it is possible to use two independent waste streams for the preparation of a sustainable pigment for artistic applications.

X-Ray Diffraction (XRD) Analysis

The structural features of the synthesised carbon were investigated by X-ray diffraction analysis. Figure 3 displays the diffraction pattern. The XRD profile was characterised by a broad diffraction band around 20°–30° (2θ) that can be attributed to the (002) reflection of disordered carbon materials. The absence of strong crystalline reflections indicates that the carbon produced during pyrolysis is predominantly amorphous with a low degree of graphitic ordering. Similar structural features were observed in our previously published study on pyrolytic carbon derived from plastics, confirmingthereproducibilityofthepyrolysis process [1].

The broadening of the (002) diffraction band indicates that the graphene-like layers are randomly stacked in the absence of long-range crystallographic ordering. This structure is often referred to as turbostratic carbon, where separate aromatic layers are misaligned and separated by larger interlayer distances than in crystalline graphite [2,7].

Unlike highly crystalline graphite with a sharp diffraction peak near $2\theta \approx 26°$, the synthesised carbon showed only a broad hump characteristic of small aromatic domains, large structural disorder and a high concentration of defects. These defects are advantageous for applications such as adsorption, surface modification, pigment dispersion, and interaction with aqueous media because they provide a large number of chemically active surface sites.

The observed XRD features therefore confirm the highly defective amorphous structure of the carbon produced during the laboratory-scale plastic pyrolysis process, which can be further functionalised and utilised as a pigment in sustainable ink formulations.

Figure 3. X-ray diffraction (XRD) pattern of the plastic pyrolysis-derived carbon used for the preparation of the eco-friendly carbon-based ink. The broad diffraction band observed in the 2θ range of approximately **20°–30°** confirms the predominantly amorphous turbostratic carbon structure with a low degree of graphitic ordering,indicating the presence of structurally disordered carbon suitable for pigment applications.

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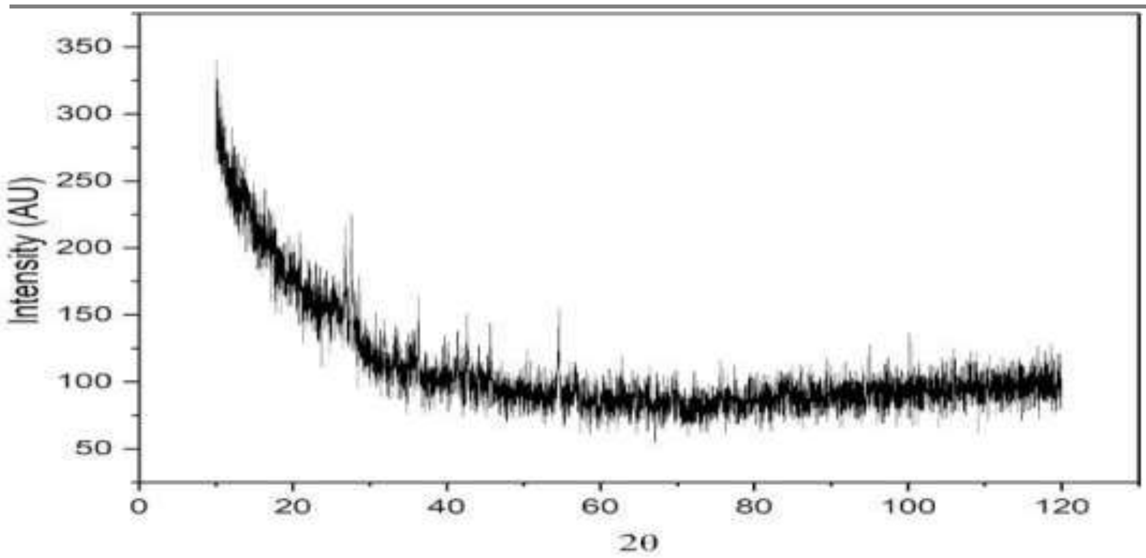

Table 3. Interpretation of the XRD Pattern

Observation	Interpretation	Significance
Broad peak between 20°–30°	(002) plane of disordered carbon	Formation of amorphous carbon
Absence of sharp diffraction peaks	Lack of long-range crystallinity	Non-graphitic carbon structure
Broad diffraction width	Small aromatic domains	High structural disorder
Elevated background	Structural defects and disordered carbon	Increased surface activity
Low graphitic ordering	Turbostratic carbon	Suitable for pigment applications

The structural disorder observed in the XRD spectrum is advantageous because highly ordered graphite particles generally exhibit lower surface reactivity than amorphous carbon. The defect-rich surface may improve interaction between the carbon particles and tea-derived polyphenolic molecules, thereby enhancing pigment dispersion within the prepared ink.

Fourier Transform Infrared Spectroscopy (FTIR)

The FTIR spectroscopy was used to study the surface chemistry of the synthesised pyrolytic carbon. The obtained spectrum (Figure 4) showed that, although the material was mainly carbonaceous, several oxygen-containing functional groups were still present on the surfaces of the particles. A broad absorption band in the region of 3400-3500 cm-1 was observed and assigned to O-H stretching vibrations of hydroxyl groups and adsorbed water. These hydroxyl functionalities are very important as they enhance the hydrophilicity of the carbon particles and help in their interaction with the aqueous tea extract. The weak absorption bands in the region of 2920-2850cm -1 are attributed to C-H stretching of residual aliphatic hydrocarbonfragments formedafter thermaldecomposition. The specific absorption band around 1700 cm-1 is characteristic of the carbonyl (C=O) functional groups and the absorption in the range of 1600 cm-1 is due to the stretching vibration of aromatic C=C. These aromatic structures are typical of condensed carbon domains formed during polymer pyrolysis, and are consistent with the structural interpretation derived from the XRD analysis. The absorption bands observed in the range of 1000–1200 cm-1 are assigned to C–O stretching vibrations of alcohol, ether and phenolic functional groups. These oxygenated surface functionalities are known to enhance the wetting characteristics and help in dispersion of carbon particles in aqueous systems.

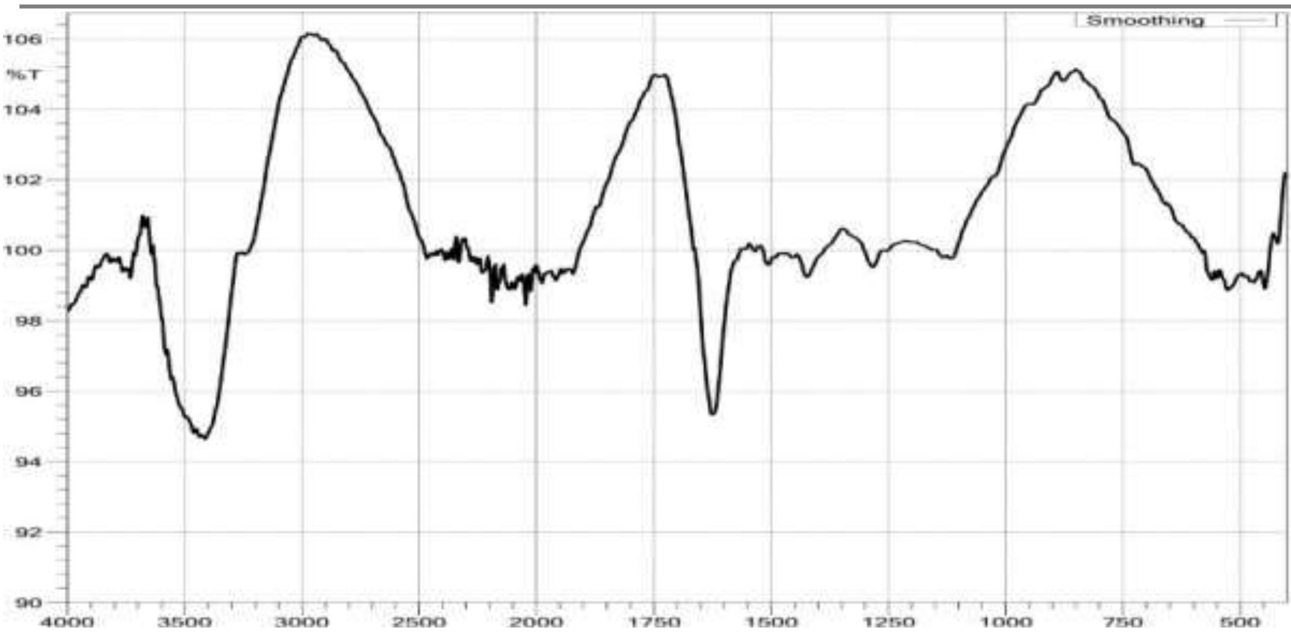

Figure 4. Fourier Transform Infrared (FTIR) spectrum of the plastic pyrolysis-derived carbon employed in the preparation of the eco-friendly carbon-based ink. The spectrum reveals characteristic absorption bands corresponding to hydroxyl (O–H), carbonyl (C=O), aromatic C=C, and C–O functional groups, confirming the presence of oxygen-containing surface functionalities that promote interaction between the carbon particles and tea-derived polyphenolic compounds.

Table 4. FTIR Peak Assignments

Approximate Wavenumber (cm^{-1})	Functional Group	Interpretation
3400–3500	O–H stretching	Hydroxyl groups and adsorbed water
2920–2850	C–H stretching	Residual aliphatic hydrocarbons
~1700	C=O stretching	Carbonyl-containing surface groups
~1600	Aromatic C=C	Condensed aromatic carbon
1000–1200	C–O stretching	Alcohol, ether, and phenolic groups
700–900	Aromatic C–H bending	Aromatic ring vibrations

The coexistence of aromatic carbon domains and oxygen-containing functional groups indicates that the synthesized material is chemically active and capable of interacting with naturally occurring tannins present in the tea extract through hydrogen bonding and π–π interactions.

Correlation Between XRD and FTIR

XRD and FTIR results complement each other. XRD gives information about the long range structural arrangement of the carbon atoms, while FTIR recognises the surface functional groups involved in the chemical interactions.

The broad XRD diffraction band confirmed the dominance of amorphous turbostratic carbon and FTIR revealed the presence of hydroxyl, carbonyl, aromatic and C–O functional groups on the particle surface. The combination of these structural and chemical features is expected to lead to better dispersion of the carbon particles in the tea extract, thus contributing to the formation of a stable pigment suspension.
XRD and FTIR analysis showed agreement which suggests that the prepared carbon has structural defects and chemically active functional groups, which makes it suitable for pigment based applications.

Table 5. Correlation Between XRD and FTIR

XRD Observation	FTIR Observation	Combined Interpretation
Broad (002) diffraction band	Aromatic C=C stretching	Disordered aromatic carbon network
Amorphous structure	Hydroxyl and carbonyl groups	Chemically active carbon surface
High structural disorder	Oxygen-containing functional groups	Enhanced interaction with aqueous media
Low graphitic ordering	Phenolic and C–O groups	Improved pigment dispersion potential

pH Analysis of the Developed Carbon-Based Ink

The acidity of the prepared carbon-based ink was evaluated by a calibrated digital pH meter. The pH of the formulation was measured as 3.0 indicating that the developed ink has an acidic character. The formulation is acidic mainly due to the chemical composition of the two waste-derived constituents used in the preparation process. Tea leaves naturally contain tannins, derivatives of gallic acid, catechins, flavonoids, and other polyphenolic compounds, which release hydrogen ions when dispersed in water [15]. Moreover, the FTIR spectra of the pyrolytic carbon showed the presence of oxygen-containing functional groups, including hydroxyl and carbonyl species, which further contribute to the acidic nature of the carbon surface. The acidic pH in the present investigation is advantageous for the dispersion stability, since protonation of surface functional groups can reduce the agglomeration of particles during storage. However, prolonged contact of acidic ink with some metallic pen components may lead to corrosion. Hence, the present formulation is more suited for applications such as brush painting, watercolour painting, dip pen calligraphy and artistic illustrations rather than conventional fountain pens unless further formulation optimisation is done. The pH measured also confirms that the prepared formulation has chemically active functional groups and not an inert carbon suspension.

Table 6. Interpretation of the pH Measurement

Parameter	Measured Value	Interpretation
pH	3.0	Acidic formulation
Primary source of acidity	Tea-derived tannins and polyphenols	Natural organic acids
Additional contribution	Oxygen-containing carbon surface groups	Surface chemistry confirmed by FTIR
Potential implication	Improved aqueous dispersion	Suitable for artistic applications

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Figure 5. Digital pH meter measurement of the prepared eco-friendly carbon-based ink. The measured pH value of 3.0 indicates that the formulation possesses an acidic nature, primarily due to naturally occurring tannins, polyphenols, and organic acids extracted from recycled tea leaves, together with oxygen-containing functional groups present on the pyrolytic carbon surface.

Electrical Conductivity Analysis

The electrical conductivity of the prepared ink was measured using a laboratory conductivity meter, yielding an instrument reading of **10.42 µS/cm**.

Electrical conductivity reflects the ability of a solution to transport electrical charge through the movement of dissolved ions. Unlike commercial conductive inks containing silver nanoparticles or graphene, the present formulation was not intended to function as an electrically conductive electronic ink. Instead, conductivity measurements were performed to evaluate the ionic characteristics of the aqueous carbon dispersion.

The observed conductivity is attributed to several contributing factors:

- Dissolved ionic species extracted from the used tea leaves.
- Organic acids naturally present in the tea extract.
- Polar oxygen-containing functional groups on the surface of the pyrolytic carbon.
- Interaction between dispersed carbon particles and the aqueous medium.

The measured conductivity confirms that the prepared formulation behaves as a visually homogeneous aqueous dispersion during the initial observation period. containing mobile ionic species. This observation is consistent with the acidic pH and the oxygen-containing functional groups identified by FTIR.

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Figure 6. Electrical conductivity measurement of the prepared eco-friendly carbon-based ink using a laboratory conductivity meter. The formulation exhibited a conductivity of **10.42 µS/cm**, indicating the presence of mobile ionic species within the aqueous dispersion and confirming effective interaction between the dispersed pyrolytic carbon particles and the tea-derived extraction medium.

Table 7. Interpretation of Conductivity

Parameter	Observation	Scientific Interpretation
Conductivity	10.42 µS/cm	Presence of dissolved ionic species
Charge transport	Ionic conduction	Tea extract contributes ions
Carbon contribution	Surface interaction	Functional groups improve dispersion
Overall behaviour	visually homogeneous aqueous dispersion during the initial observation period.	Appropriate for pigment formulation

Relationship Between XRD, FTIR, pH, Conductivity and Ink Performance

The techniques of characterisation used in this work give complementary information on the structural and physicochemical behaviour of the developed ink. The XRD analysis showed that the carbon is largely amorphous turbostratic carbon with high disorder in the structure. These structural defects increase the available surface and provide many active sites for interaction with surrounding molecules.

FTIR analysis confirmed the existence of hydroxyl, carbonyl, aromatic C=C and C–O functional groups on the carbon surface. The presence of these oxygen containing functionalities improves the wetting properties of the carbon particles and allows for a hydrogen bonding to be formed with the polyphenol compounds extracted from the tea leaves. The measured pH of 3.0 further confirms the presence of acidic functional groups in the formulation. The conductivity measurement suggests that dissolved ionic species are still available for charge transport across the aqueous medium.

Taken together these results suggest that the developed formulation is not simply a suspension of carbon particles in water but rather a chemically interactive system in which waste derived carbon and tea derived organic molecules coexist via physical adsorption, hydrogen bonding, and π–π interactions. Such interactions result in better dispersion of the pigments and may improve coating uniformity during artistic applications.

Table 8. Correlation Between Characterization Techniques

Characterization Technique	Observation	Contribution to Ink Development
XRD	Amorphous turbostratic carbon	Fine pigment structure
FTIR	O–H, C=O, C=C, C–O groups	Improved interaction with tea extract
pH	Acidic (3.0)	Presence of active functional groups
Conductivity	Ionic behaviour	Stable aqueous dispersion
Combined interpretation	Chemically active pigment system	Suitable for eco-friendly artistic ink

Proposed Water colour and Artistic Applications

The developed formulation has a number of properties desirable for artistic applications. The fine carbon particles provide a deep black colour. The extract from tea also contains naturally occurring tannins, which can shift the visual tone to a warm black or brownish-black tone. The aqueous formulation allows easy application with brushes, suitable for watercolour painting, monochrome illustrations, wash techniques, calligraphy, educational art activities, and handmade artistic products.

The developed ink is based on waste materials that would otherwise contribute to environmental pollution, unlike conventional petroleum-derived carbon black pigments. Therefore, the formulation is a sustainable alternative for low-cost educational and artistic applications.

Detailed colourimetric measurements, quantitative viscosity/rheological analysis, particle-size distribution, sedimentation stability, standardized adhesion and abrasion testing, and long-term storage stability were outside the scope of the present investigation. Drying behaviour was evaluated separately through preliminary measurements on different paper substrates.

Comparison with Conventional Carbon-Black Inks

Table 9. Qualitative Comparison Between Conventional Carbon-Based Ink and the Present Waste-DerivedFormulation

Property	Conventional carbon-based ink	Present study
Carbon source	Typically commercial carbon black	Plastic pyrolysis-derived carbon
Additive	Formulation dependent	Spent tea-leaf extract
Feedstock	Primarily commercial/fossil-derived carbon	Waste plastic + tea waste
XRD/FTIR characterization	Product dependent	Performed
pH	Product dependent	3.0
Conductivity	Product dependent	10.42 μS/cm
Drying	Formulation dependent	20–25 s preliminary measurement
Viscosity	Not evaluated here	Not measured
Long-term stability	Product dependent	Not established
Intended use	Depends on formulation	Preliminary artistic application

The present formulation demonstrates that functional artistic pigments can be developed from recycled materials without relying entirely on virgin fossil-derived carbon sources.

Drying Behaviour of the Developed Ink

The drying behaviour of the prepared eco-friendly carbon-based ink was evaluated under ambient laboratory conditions by applying the formulation onto different types of drawing papers using a standard paintbrush. The observed surface drying time ranged from 20 to 25 seconds, depending on the absorbency, surface finish, and thickness of the paper substrate. Papers with higher absorbency exhibited shorter drying times, whereas thicker or coated papers required slightly longer drying periods. The rapid drying behaviour is attributed to the water-based formulation, in which evaporation of the aqueous phase occurs efficiently while the dispersed pyrolytic carbon particles remain deposited on the paper surface. The measured drying time is comparable to that of many commercially available watercolour formulations when applied as thin brush layers under similar environmental conditions. These observations indicate that the developed waste-derived ink possesses practical drying

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characteristics suitable for artistic applications such as watercolour painting, brush illustration, and calligraphy.
Table 10. Drying Characteristics of the Prepared Carbon-Based Ink

Parameter	Observation
Drying time	20–25 s
Test conditions	Ambient laboratory conditions
Application method	Brush painting
Paper dependence	Faster on absorbent paper; slower on thicker/coated paper
Ink type	Water-based carbon pigment

Paper Type	Drying Time (s)	Observation
Cartridge paper (120 GSM)	20	Fast absorption
Drawing paper (180 GSM)	22	Uniform drying
Watercolour paper (300 GSM)	25	Slightly slower due to sizing

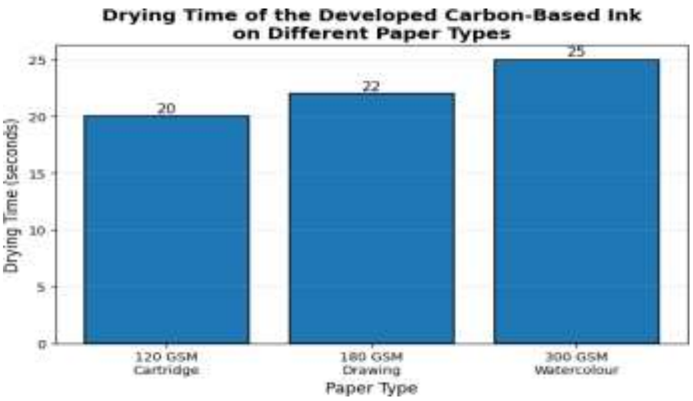

Figure 7. Drying time of the developed eco-friendly carbon-based ink on different paper substrates under ambient laboratory conditions.

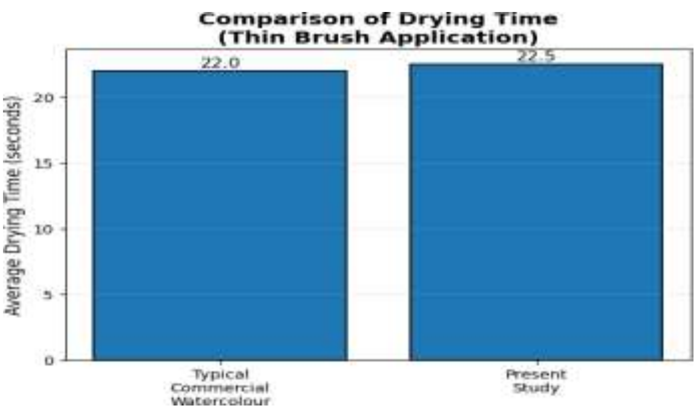

Figure 8. Comparison of the average drying time of the developed carbon-based ink with the representative drying time of a thin-layer commercial watercolour formulation under similar ambient conditions. The commercial value represents a typical range reported for water-based artist paints and is provided for qualitative comparison only.

Innovation of the Present Study

The innovation of this work lies in integrating **two independent waste streams** into a single functional product.

The study introduces a novel pathway in which:

- Waste plastic is converted into pyrolytic carbon through oxygen-limited pyrolysis.
- Spent tea leaves are reused as a natural source of tannins and polyphenols.
- The resulting materials are combined to produce an eco-friendly carbon-based ink.

To the best of the authors' knowledge, studies that combine **plastic pyrolysis-derived carbon with recycled tea leaf extract specifically for artistic ink development**, together with structural characterization by XRD and FTIR and physicochemical evaluation through pH and conductivity measurements, remain very limited in the published literature. The present work therefore contributes to sustainable materials engineering, waste valorisation, and circular economy initiatives while extending the utilization of the pyrolytic carbon previously developed by the authors.

Limitations

The current study serves as a laboratory-scale proof-of-concept for the production of a carbon-based artistic ink using carbon obtained from plastic pyrolysis and discarded tea leaves. Material preparation, structural characterisation, and initial physicochemical and application-related evaluation were the main areas of attention for the study. Due to the lack of an appropriate rotating viscometer/rheometer throughout the experiment, quantitative rheological characterisation was not carried out. Similarly, the lack of controlled stability testing and microscopy-based particle analysis prevented quantitative measures of sedimentation stability and particle-size distribution. The experimental scope does not include standardised colorimetric, opacity, adhesion, abrasion, and long-term storage tests. Therefore, it is not appropriate to interpret the current findings as proving long-term shelf stability or commercial-grade performance. Future research utilising standardised equipment and test procedures must systematically examine these parameters.

Future Scope

The present study demonstrated the feasibility of the conversion of plastic pyrolysis-derived carbon and recycled tea leaf extract to develop an eco-friendly carbon-based ink for artistic applications. Although the present study is limited to structural and physicochemical characterisation, the developed formulation opens up several promising research directions in the fields of materials science, printed electronics, renewable energy and sustainable manufacturing.

Optimization of Ink Formulation

Future work should focus on improving the rheological properties of the ink by optimisation of the carbon particle size distribution, pigment concentration, dispersion stability and binder composition. Natural or synthetic binders such as gum arabic, xanthan gum, carboxymethyl cellulose (CMC) or polyvinyl alcohol (PVA) can be used to improve adhesion, viscosity, storage stability and drying behaviour for various writing and artistic applications

Conductive Carbon Ink for Printed Electronics

Although the present formulation exhibits measurable electrical conductivity, further enhancement of conductivity may enable its application as a low-cost conductive carbon ink. Conductivity may be improved through:

- Chemical activation of the pyrolytic carbon using KOH, $ZnCl_2$, H_3PO_4, or steam activation.
- High-temperature graphitization to increase graphitic ordering.
- Particle size reduction to the micro- or nanoscale.
- Incorporation of conductive nanomaterials such as graphene, reduced graphene oxide (rGO), carbon nanotubes (CNTs), carbon black, or MXenes.

- Optimization of carbon loading and dispersion.

Improved conductivity could expand the applicability of the material in printed and flexible electronic devices.

Flexible and Printed Electronics

Following conductivity enhancement, the developed ink could be investigated for fabrication of:

- Flexible printed circuit boards (FPCBs)
- Printed conductive tracks
- Paper-based electronics
- Wearable electronic devices
- Flexible sensors
- RFID antennas
- Smart packaging
- Low-cost educational electronic circuits

Compatibility with screen printing, inkjet printing, aerosol printing, and stencil printing techniques should also be investigated.

PCB Prototyping

The carbon-based ink may be explored as an environmentally friendly conductive patterning material for rapid PCB prototyping. Future studies should evaluate:

- Pattern resolution
- Electrical resistance
- Adhesion to copper-clad laminates
- Thermal stability
- Compatibility with PCB etching and masking processes
- Reliability under repeated electrical loading

Such investigations may contribute to sustainable electronics manufacturing.

Energy Storage Applications

The defect-rich amorphous carbon identified through XRD and FTIR suggests potential applicability in electrochemical devices. Future investigations may include:

- Supercapacitor electrodes
- Sodium-ion batteries
- Potassium-ion batteries
- Zinc-ion batteries

- Micro-supercapacitors
- Conductive current collectors

Electrochemical techniques including cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) should be employed to evaluate performance.

Photovoltaic and Solar Energy Applications

Following appropriate optimization of electrical conductivity and optical properties, the developed carbon material could be investigated as a conductive carbon layer or functional coating in photovoltaic devices. Possible future applications include:

- Carbon-based conductive layers
- Counter electrodes
- Hole transport layers (where appropriate)
- Carbon composite coatings
- Printed conductive contacts

At present, this study does not demonstrate improved solar-cell efficiency. Future experimental investigations should evaluate whether optimized formulations can contribute to photovoltaic device performance through systematic electrical and efficiency measurements.

Electromagnetic and Functional Coatings

The carbon material may also be investigated for:

- Electromagnetic interference (EMI) shielding
- Microwave absorbing coatings
- Conductive paints
- Anti-static coatings
- Electrochemical sensors
- Carbon-based composite coatings

Environmental Applications

Future research should include:

- BET surface area analysis
- Pore size distribution
- Heavy metal adsorption studies
- Dye adsorption studies
- Oil absorption capacity
- Water purification performance

- Gas adsorption characteristics

Industrial Scale-Up

Pilot-scale production should be investigated to evaluate:

- Economic feasibility
- Process scalability
- Energy consumption
- Carbon footprint
- Life-cycle assessment (LCA)
- Commercial feasibility

Successful scale-up may enable commercialization of sustainable carbon-based inks while simultaneously reducing plastic waste and utilizing spent tea leaves.

CONCLUSION

The current work showed that it is possible to turn two waste streams—spent tea leaves and plastic packaging debris—into a carbon-based artistic ink. While discarded tea leaves were treated to provide a naturally generated aqueous extract containing polyphenolic compounds, plastic debris was converted into pyrolytic carbon under oxygen-limited circumstances. The final mixture was effectively applied to paper substrates and had a visually uniform black look. While FTIR verified oxygen-containing surface functions such as O–H, C=O, aromatic C=C, and C–O groups, XRD revealed a mostly amorphous/turbostratic carbon structure. The produced ink has an electrical conductivity of 10.42 µS/cm and an acidic pH of 3.0. Depending on the paper substrate, preliminary drying studies revealed drying periods of between 20 to 25 seconds. These findings show that carbon produced from waste and leftover tea leaf extract can be used for early artistic ink applications. Nevertheless, the current investigation did not establish quantitative rheology, particle-size distribution, sedimentation stability, standardised colour strength, adhesion, abrasion resistance, or long-term storage stability. As a result, rather than being a commercially certified ink, the formulation should now be regarded as a laboratory-scale proof-of-concept. Standardised performance testing and systematic formulation optimisation should be the main topics of future research.

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