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The Emerging of Nanocellulose as a New Generation of Water Purification Application

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Abstract: Water pollution has been identified to be the grand environmental challenge, that could be adversely affected by the rapid growth urbanization, industrialization and economic development. In parallel to the concept of green chemistry principles for sustainability, the development of eco-friendly and advanced treatment systems has become critically important to the threatening conditions of environmental contamination. Nanocellulose, a class of sustainable and carbon-neutral nanomaterials, has received specific highlight in this regard to their attractive properties, from the perspective of the including abundance of hydroxyl groups, light-weight, high aspect ratio, tailorable surface chemistry, exceptional mechanical flexibility, bio-degradability and environmental compatibility. The 3D interleaved nanonetworks and tailorable pore structure over the surfaces of nanocellulose enable a broad range of surface functionalization, resulting in numerous tunable characteristics and advanced functionalities. Within this context, this paper attempts to elucidate the driving causes and major impacts of water pollution, and the inherent advantages and limitations of available treatment approaches. The development of nanocellulose as a new emerging water treatment technique, together with the challenges and future perspectives are discussed and presented. These advanced functional nanocellulose materials are expected to foster cross-disciplinary development in the near future.

Keywords: Nanocellulose, Nanotechnology, Water pollution, Water and wastewater treatment

1. Introduction

Water, the foundation of human survival, ecological balance and sustainable socio-economic development, serves as the most essential linkage between community, environmental system and resource management. Protection and provision of safe, clean and affordable freshwater resource, for both potable or non-potable requirements, are the key messages from United Nations (UN) Sustainable Development Goal (SDG) 6 [1], with the global targets on water quality improvement, water pollution control, better water management and preservation of ecosystems. The rapid population growth, coupled with the growing industrialization have led to the alterations of composition, chemical structure, complexity, instability and toxicity, due to the diverse range of pollutants, not limited to conventional (nutrients, microbial pollutants, heavy metals, organic and inorganic compounds) and emerging water pollutants (pesticides, pharmaceuticals, drugs of abuse, antibiotics and hormones, endocrine disrupting compounds (EDCs), personal care products, artificial sweetener, surfactants, microplastic, persistent organic pollutants, and per- and polyfluoroalkyl substances), being released from point and diffuse sources into the environment [2]. The accumulation of water pollutants as a mixture pollutant in the environment appear to be highly hazardous than the individual entities, which may lead to undesirable synergistic consequences [3].

The existing natural or conventional treatment processes are not capable to remediate the new emerging contaminants, and reaching their limits for the simultaneously removal of mixed pollutants from the aquatic streams [4]. The adaptation of highly advanced nanotechnology to traditional process engineering offers new opportunities for the development of advanced water treatment process. Nanotechnology is an interdisciplinary field that strives to manage, produce and develop novel prospects of science, engineering and new approaches, at a nanoscale condition. Nanotechnology which applies nanoscale particles or nanomaterials holds significant potential to cost-effectively address the challenges of existing treatment technology systems, for the improvement of the overall efficiency of wastewater remediation processes [5]. Specific efforts have been devoted to the integration of green chemistry concept into the rapidly evolving field of nanotechnology, for the sustainable transformation of industrial or agricultural bio-residue into high quality nanocellulose. Nanocellulose, as the fundamental natural building block of cellulosic-based fibres, have fore-fronted to be a new generation of functionalized nanomaterials, which have attracted tremendous attention among the

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nanotechnology community. The highly rodlike nanocrystalline of nanocelluloses featuring their excellent mechanical property, low-density, renewability, sustainability and tailorable surface properties, present a great potential to revolutionize the new water and wastewater treatment applications [6]. Within this framework, this paper outlines an overview on the driving factors and impacts of water pollution. The advantages and constraints of the existing treatment technologies are presented. Moreover, the emergence of nanocellulose, together with their potential for water/ wastewater remediation applications are elucidated.

2.0 Structure Water Pollution and Its Detrimental Implications

Water pollution associated with water quality and water scarcity are exacerbated by the population explosion and its associated effects, notably in the forms of accelerated urbanization, industrialization and technological development, and the rapid economic development. Growing municipal, agricultural and industrial waste discharges, coupled with indiscriminate dumping, sewer lines leakage, accidental spillage/ seepage, and limited wastewater treatment system capacity (outmoded technologies and obsolete facilities), have emitted serious menace on increasing environmental quality deterioration, water pollution and sustainable development [7]. The combination pollution of multi-pollutants, specifically hazardous dyestuffs, organic and inorganic contaminants, alongside with waterborne microbial pathogens and fungi species, owing to their high tetra-toxicity, non-biodegradability, chemical stability, high lipophilicity, solubility, persistence and bio-accumulative behaviours, and long-range transport in various environmental matrices, represents a critical risk to the waterway-ecosystem and human health [8]. Researchers have reported the connections between water pollution and water-borne/ enteric disease incidence, notably presented as skin and eyes diseases, gastrointestinal illness (GI), hepatitis, cholera, dysentery, diarrhea, typhoid fever, respiratory tract infection, kidney damage, endocrine disorder and metabolic syndrome, a legacy which continues to plague most of the world's population [9]. Serious conflicts have arisen on the disorganized development and management of water resource, improper public engagement, consciousness and knowledge in pro-environmental behaviours, and a corresponding limited of legal and economic frameworks to promote water stewardship and conservation [10]. To achieve the sustainable degree of water resource usage and compliance with environmental and health regulations, recent effort has been focused on the development of advanced with robust, high-efficiency and economically viable water/ wastewater treatment alternatives, that are in line with its commitments to the Sustainable Development Goals' (SDG) 2030 targets [11].

3.0 Water Pollution Control Technologies

Effective water pollution control requires an in-depth understanding on the water contaminants, treatment processes, as well as the qualitative and quantitative water quality data before the corresponding strategy is elected [12]. These aforementioned techniques, notably natural-based solutions, filtration, disinfection, ion exchange, precipitation, coagulation, dialysis, foam flotation, advanced oxidation, membrane filtration, adsorption and photocatalytic degradation, have been developed for the removal of a variety of toxic pollutants from the water streams [13,14,15,16]. The widespread applicability and sustainability of these treatment technologies or their combination, however, may be restricted by the (a) economically affordability (investment, processing efficiency, operation and maintenance, and residuals management), (b) environmental sustainability (environmental protection, resources conservation, energy requirement, engineering expertise, water reuse, nutrient recycling, and seasonal and temporal variations), and (c) socially acceptability (public health protection, government policy and regulations, and human settlement), all of which have precluded their implementation under critical scenarios [17]. Table 1 outlines the advantages and limitations of the most commonly used water/ wastewater treatment techniques.

Table 1 - Advantages and limitations of different treatment techniques.

Category	Technique	Advantages	Limitations
Physical	Filtration	• Most common technique • Inexpensive	• May not effectively remove pathogens on its own
	Membrane filtration	• Easy to operate • Low space requirement • High efficiency	• Low flux • Low-selectivity • Membrane fouling • Periodic replacement of membrane • High cost • Incapable for large-scale application
	Dialysis	• No chemical required • Works under a wide pH range	• Membrane fouling • Require regular cleaning and maintenance • Limited to small-scale application
Chemical	Disinfection	• High water solubility • Inexpensive and readily available • Effective against most pathogens	• Corrosive • High generation of hazardous Cl_2 gas
	Coagulation/ Flocculation	• Low cost • Easy-to-operate procedure • High efficiency • Removal of fine, insoluble and colloidal contaminants • Significant reduction of COD and BOD_5 • Bacterial inactivation capability	• High chemical and energy consumption • Large quantity of hazardous sludge generation • High amount of chemicals requirement for pH adjustment • Increase water hardness

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Table 1 - Advantages and limitations of different treatment techniques (Continued).

Category	Technique	Advantages	Limitations
Chemical	Chemical precipitation	• Simple equipment • Integrated physico-chemical process • Adapted to high pollutant loads • Efficient for the removal of metal, fluoride and COD • Not metal selective	• High chemical consumption • Ineffective for the removal of the metal ions at trace concentration • Require an oxidation step if the metals are complexed • High sludge production, handling and disposal problems
	Advanced oxidation	• Eco-friendly • Non-hazardous • Good elimination of colour and odor • *In-situ* production of reactive radicals	• High cost • Oxidative by-products production • Laboratory scale
Biological	Phyto-remediation	• Low capital and energy requirement • Less carbon footprint and secondary waste generation • Environmental - friendly • Reclamation of wastewater and nutrient recovery	• Slow remediation duration • Low rate of contaminant removal • Require more land • Limited to shallow contaminants • High risk of ecological and human exposures
	Microbial remediation	• Low operational costs • Economically attractive • High removal of BOD_5 and bio-degradable organic matters • No secondary pollution	• Inconsistent and slow process • Requires management and maintenance of microorganisms and/or physico-chemical pre-treatment • Generation of biological sludge and uncontrolled degradation products • Complexity of the micro-biological mechanisms

Table 1 - Advantages and limitations of different treatment techniques (Continued).

Category	Technique	Advantages	Limitations
Biological	Activated sludge process	• High treatment efficiencies possible for BOD_5, COD, TSS, nitrogen and phosphorus • High flexibility • High effluent quality	• High cost of operation and maintenance requirements • Low pathogen removal • Poor decolorization • Sludge bulking and foaming • Requires killed personnel
Physico-chemical	Adsorption	• Simplicity • High effective process with fast kinetics • Large range of adsorbent material • Excellent quality of the treated effluent	• Weak selectivity • High cost • High generation of waste product • pH dependent • Chemical derivatization to improve adsorption capacity • Rapid saturation and clogging of the reactors • Regeneration is expensive and results in loss of materials
	Flotation	• Integrated physico-chemical process • Low level of sludge formation • High separation efficiency • Effective for inorganic and organic contaminants removal • Low energy requirement	• High cost of operation and maintenance • pH dependent
	Ion-exchange	• Low cost • High removal efficiency • High treatment capacity • Easy to use with other techniques • Applicable to different flow regimes • Remove fluoride up to 90-95% • Fast reaction kinetics • Operative with no loss of regeneration	• Influenced by the variety, yield and cost of exchangers • Rapid saturation and clogging of the reactors • Performance sensitive to pH • Not effective for disperse dyes

Table 1 - Advantages and limitations of different treatment techniques (Continued).

Category	Technique	Advantages	Limitations
Physico-chemical	Photo-catalysis	• Renewable energy source utilization • Highly efficient process • Excellent degradation effective • Elimination of wide spectrum of organic pollutants • Low sludge generation	• High initial cost of equipment and maintenance • Poor thermal stability • Limited lamp life • Difficult photocatalyst separation and recovery from solution

4.0 Emergent of Nanocellulose for Water Purification Application

Nanotechnology is a complex, emerging and inter-disciplinary field of material research targeted on the economic-society-ecology development. For the last few decades, the development of nanoscience technology as the top scientific agenda is impelling a rapid growth of different green approaches for environmental remediation, mainly ascribed to the simplicity of preparation technique, and design and manufacturing of chemical products, with minimum chemicals and energy consumption. This rising concern has attracted aesthetic attention to the practical use of agricultural residues, herbaceous crops, and non-wood fibers from forestry and agro-food industries as viable substitutions to the synthetic materials. Within the portfolio of commended biopolymer, cellulose, as a structural linear homopolysaccharide with the repeating units of β-1,4-D-glucopyranose, is the most abundant resources of polysaccharide in the higher plants, woods, specific algae, amoeba, bacteria, invertebrates and tunicates (Fig. 1). In the recent development of cellulosic-based materials, the recovery of crystalline nanofibrils from the native cellulose, or known as nanocrystalline cellulose/ nanocellulose (NC), has been highlighted to be a fascinating interest in both academia and industrial sectors, due to its unique multi-dimensional structure and exquisite characteristics as compared to the bulk precursors [18].

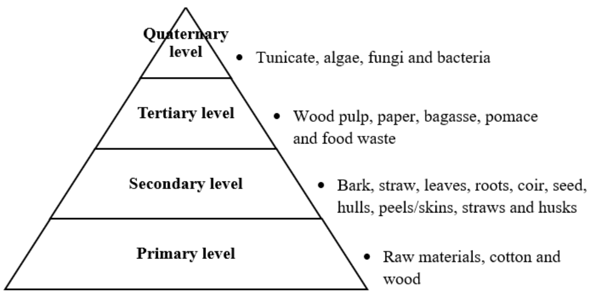

Fig. 1 - Sources of cellulosic material.

Nanocrystalline cellulose is a form of one-dimensional (1D) nanostructured cellulosic material, consisting of β-1,4-glucose and three (3) hydroxyls active at the C_2, C_3 and C_6 positions of glucopyranosyl ring, with at least one of its external dimensions (length or width/ diameter) ≤ 100 nm [18]. Table 2 lists a comparison of the three major types of NC, from the perspectives of sources, formation techniques and geometrical dimensions. According to the TAPPI standard terminology, the term nanocrystalline cellulose is generally classified into three hierarchical nano-structural forms, namely cellulose nanocrystals (CNCs), cellulose nanofibers (CNFs) and bacterial nanocellulose (BNC), presented by different alignment of alternating crystalline and amorphous regions, which in turn depended primarily on the cellulosic sources, synthesis procedures and processing conditions. The fundamental features of NCs reflect the key properties of cellulose (Table 3), making it a rising star in various applications, notably within the healthcare industries, biomedical engineering, cosmetics, electronic, construction, food packaging, functional additives and environmental applications.

Table 2 - Classification of NCs (adopted from TAPPI W1 3021) [19].

Type of NC	Synonyms	Typical source	Preparation technique	Geometrical dimension
Cellulose nano-crystals (CNCs)	Crystallites, cellulose whiskers, nano-whisker, nano-crystals, nano-wire, Nano-rod	Wood, non-woody residues, tunicates and bacteria	Acid hydrolysis, or hybrid of physical and chemical method	*D*: 5-70 nm *L*: 100-500 nm (from plant source); 100 nm to several micro-meters (from tunicates, algae and bacteria) *AR*: 5-50
Cellulose nano-fibers (CNFs)	Nano-fibrils, nano-fiber, nano-fibrillar cellulose	Wood, non-woody residues, tunicates and bacteria	Mechanical, chemical or enzymatic treatments	*D*: 5-30 nm *L*: 100-2,000 nm *AR*: ≥ 50
Bacterial nano-cellulose (BNC)	Bacterial cellulose, microbial cellulose, bio-cellulose	Micro-organisms	Bacterial synthesis	*D*: 20-100 nm *L*: > 1,000 nm (Nano-fiber network) *AR*: 100-150

Table 3 - Fundamental features of nanocrystalline cellulose.

Property	Description	Reference
Aspect ratio (*AR*)	• An important parameter for nanocrystalline cellulose is the aspect ratio, which could be defined as the ratio of length (*L*) to width (*W*)/ diameter (*D*) • It determines the anisotropic phase formation and reinforcing properties • The *AR* of CNCs is less than 50, but greater than 5 • The *AR* of CNFs is greater than 10	[20]

Table 3 - Fundamental features of nanocrystalline cellulose (Continued).

Property	Description	Reference
Size and morphology	• Core parameter to impact the functionality and property of engineered functional materials • CNCs consist of cylindrical, elongated, less flexible and rod-like nanoparticles, with 5-70 nm in width, and 100-500 nm in length • CNFs presents an entangled network structure with flexible, longer and wide nanofibers (5-30 nm in width and 100-2000 nm in length) • Expressed in term of shape, length (*L*), width (*W*), diameter (*D*), aspect ratio (*AR*) and average particle size/ size distribution	[21]
Crystallinity	• A parameter to image the crystallinity, crystal structure and polymorphic of nanocrystalline cellulose • Crystallinity index (*CI*) has been applied for the description of relative amount of crystalline material in cellulose, on the basis of the crystallographic two-phase model, for both crystalline (ordered) and amorphous (less ordered) regions • The crystallinity index of nanocrystalline cellulose ranges from 45 to 80 % • The crystalline structure of nanocrystalline cellulose may influence the thermal and mechanical/ physical properties of cellulose samples	[22]
Surface charge and chemistry	• Zeta-potential is a good indicator for surface charge measurement • Two different approaches to impart/ increase surface charges onto nanocrystalline cellulose: (a) extraction/preparation process; and (b) post-modifications. • Formation of surface functional groups is crucial to improve the dispersibility and re-dispersibility of nanocellulose	[23]
Hydrophilicity	• Nanocrystalline cellulose owns strong hydrophilicity, due to the presence of abundant hydroxyl groups or hydrophilic functional groups over the surface	[24]
Broad possibility of surface functionalization	• High possibilities of surface functionalization (amidation, oxidation, esterification, etherification, radical grafting or silylation), attributed to the presence of high densities of surface hydroxyl groups, for improvement of dispersion performance	[25]
Thermal degradation	• Thermal stability is directly related to the crystalline structure and crystallinity of nanocrystalline cellulose	[26]

The preparation of nanocellulose from cellulose source materials involves typically two main stages: (a) pre-treatment to remove non-cellulosic matrix materials, and (b) hydrolysis/ extraction process to isolate nanocellulose. Pre-treatment (Fig. 2) is the necessary step for the (a) reduction of biomass recalcitrance nature, (b) degradation of lignin sheath and hemicellulose, (c) structural modification of solid materials, (d) digestibility improvement of lignocellulosic components, (e) breaking down of inter-chain hydrogen bonds between cellulose and non-cellulosic compounds, (f) depolymerization of cellulose chain into NCs, and (g) minimization of the formation of inhibitory compounds [27]. In practise, a multitude of physical, chemical and biological, or hybrid of these pre-treatment approaches (Table 4) have been carried out.

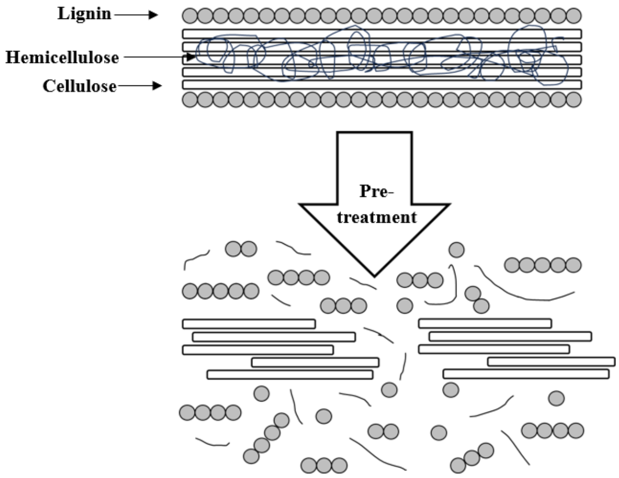

Fig. 2 - Pre-treatment process for NCs.

Table 4 - Pre-treatment techniques for NC preparation.

Pre-treatment	Description	Method
Physical	• Reduction of feedstock particle size by mechanical force • Retain most of its initial biomass compositions (cellulose, hemicellulose and lignin) after the treatment step	Grinding, milling, refining or high-pressure homogenization

Table 4 - Pre-treatment techniques for NC preparation (Continued).

Pre-treatment	Description	Method
Chemical	• Application of chemical agents, specifically acids, bases and oxidizers for the breaking of selected bondings within the biopolymer complex • Acidic pre-treatment removes and hydrolyzes up to 90 % of the hemicellulose fraction • The major effect of alkaline pre-treatment is the removal biomass lignin removal	Acid, alkali, ionic liquids, organosolv or mixed chemicals
Biological	• Application of microorganisms and enzymes for the depolymerization and degradation of lignin fraction	Bacteria, fungi and enzyme
Combined	• A combination of chemical attack and physical shear forces for the separation of microfibres from the targeted biomass • Removal of hemicellulose fraction	Extrusion, hydro-thermolysis and steam explosion

Extraction is a major processing step for NC production, in which the purified cellulosic contents are depolymerized into nanoscale products (CNCs and CNFs), with the desired characteristics. This extraction process enables the cleavage of glycosidic bonds or hydrogen bonds within the interfibrillar contacts, and disordered nanodomains of nanocellulose [28]. The three major extraction methods for the preparation of nanostructured cellulose are mechanical process, acid hydrolysis and enzymatic hydrolysis, of which acid hydrolysis has been accepted as the standard technique for industrial grade applications.

These natural-occurring cellulose-based nanomaterials own which fibrillar structures, with the sizes ranging from few nanometers to microns, have occupied a prominent time and immemorial position, due to its renewable, biodegradable, low-density, low-cost, excellent mechanical features, super-molecular structural properties, and abundant hydroxyl groups. The tailorable surface chemistry and functionality of NC materials have garnered spotlight for water treatment, organic and inorganic pollutants removal, and decontamination of microorganisms and virus from the aqueous environment. The removal efficacy of these NCs depends primarily on the nature and density of the available functional groups, that is proportional to the active surface sites in supporting the adsorption process, particularly the sulfate (SO_3^-), carboxyl (COO^-) and amine ($-NH_2$) groups. Table 5 summarizes previous studies on the application of NCs for the remediation of different water contaminants.

Cationic modification would enhance the uptake of direct, acid, sulfur (–S–, –S–S–, –Sn–), azoic (−N=N−) and vat dyes [29,30]. Anionic modification is the most prevalent method to enhance complexation and ion-exchange adsorption towards different hazardous metals ions [31,32]. Alternatively, amine modification would induce chelating action on the desired anionic pollutant via electrostatic interaction and ion exchange mechanisms under acidic conditions [24], or covalent bonding between the nitrogen atom (lone pair of electrons) with the metal ions.

Lim and Foo [6] reported a novel methodology to facilely prepared coffee residue (CR)-derived cellulose nanocrystals (CNCs) via hydrothermal technique. The hydroxyl, carbonyl and sulfate groups over the surface of CR-CNCs are the key factors of its excellent adsorption performance, demonstrating that the MB adsorption process governed by hydrogen bonding, electrostatic attraction, Van der Waals and ion-dipole interaction. They also reported the eventual interactions between CR-CNCs and chlorpyrifos, pertaining to the occurrence of π–π stacking interaction, hydrophobic force and hydrogen bonding at the interface, which can enhance the efficiency on the adsorption process.

In addition, nanocellulose has been proposed for the adsorptive removal of residual antibiotics, which are commonly found in industrial discharge, aquaculture and medicinal residues. The abundant surface -OH groups over NC surface as hydrophilic sites may accelerate nucleation and growth of inorganic particles, for the effective control of morphology, particle size and surface crystallinity. Such readily available surface –OH groups could be an ideal platform for the preparation of hybrid nanocomposites with different hierarchical structure. These OH groups can be tailored for selective and improved interactions through a variety of modification steps. The hydrophilicity of nanocellulose could be modified by intercalation of carboxylic acids, alcohols and amides into its cellulose chains. However, the hydrophilic nature of nanocelluloses might cause aggregation during the drying steps, due to the abundance of –OH groups over the surface, which might retard the adsorptive capacity [33].

Table 5 - Previous studies on the application of NCs for water purification.

Nano-cellulose	Surface functionality	Contaminant	Reference
CNCs	Sulfated group (SO_3^-)	Cationic dyes, anionic pesticides and antibiotic removal	[6,31,34, 35]
	Phosphorylated group (PO_3^{2-})	Metal cation removal	
	Carboxylated group (COO^-)	Cationic dyes and metal cation removal	
	Aminated group	Azo dye and metal cation removal	

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Table 5 - Previous studies on the application of NCs for water purification (Continued).

Nano-cellulose	Surface functionality	Contaminant	Reference
CNFs	Phosphorylated group (PO_3^{2-})	Cationic dye and metal cation removal	[36,37]
	Carboxylated group (COO^-) and aminated group	Metal cation removal	
BNC	Aminated, carboxylated (COO^-) and phosphorylated (PO_3^{2-}) groups	Metal cation removal	[38,39]

5.0 Challenges and Concluding Remarks

The selection of a treatment technique is usually site-specific, which could be altered according to the complex conditions, specifically the contaminated time, concentration and nature of the contaminant, the soil/ water and site characteristics, in which appropriate restoration and treatment plans may be formulated [40]. The relationship between risk evaluation and the suitable treatment technologies choice is critical direction for the future research, in which nanotechnology should be emphasized [41]. Within this context, the treatment performance under natural and real conditions needs to be examined [42]. Moreover, the long-term efficacy of the nanotechnology is largely beyond laboratory studies. Research addressing the long-term performance of water and wastewater treatment nanotechnologies is in a great need [43]. As a result, side-by-side comparison of nanotechnology enabled systems and existing technologies is challenging. Similarly, the adoption of innovative technologies from the perspective of preparation techniques, reusability and cost-effectiveness, would lead to broader public acceptance, that is crucial for market deployment [44].

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