Mini-Review:

Enhancing Surface Properties Through the Applications of Silica Superhydrophobic Coating

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Abstract: *Superhydrophobic silica coatings have gained enormous attention due to their significant contribution to self-cleaning, anti-fouling, anti-icing, anti-corrosion, and moisture-resistance. This mini-review provides an overview of silica surface modification, including functionalization, roughness control, and deposition methods. Functionalization aims to reach hydrophobic properties by the application of low surface energy compounds. Surface roughness control at the micro- and nano-scale across different scales also results in various surface topographies. The type of deposition technique also influences the coating surface properties, including roughness texture, adhesion, and coating thickness. This paper also explains the challenges and gaps of research rarely reported by previous studies, such as the stability of coating in harsh environments, scalability, cost-effectiveness, and sustainability. Furthermore, this article also addresses promising future innovations in developing long-lasting coatings, multifunctional properties, and economic points of view.*

Keywords: *silica; superhydrophobic; modification; gaps; innovation*

■ INTRODUCTION

Superhydrophobic coatings have gained significant attention due to their potential in self-cleaning, anti-fouling, and moisture-resistant applications. Superhydrophobic has resulted from surface modification with particular chemical chains and hierarchical rough microstructures. A superhydrophobic surface allows liquid droplets to adhere and remove dirt particles instead of passing them through [1]. A superhydrophobic water surface exhibits a contact angle greater than 150°, with contact angle hysteresis less than 5°. These unique properties arise from their chemistry or surface texture [2].

Silica-based superhydrophobic coatings are popular for water repellency and several protective applications. Silica can play as a roughness component by providing low surface energy through silanization. Meanwhile, the presence of silica influences the scratch resistance of the surface. With a higher concentration of silica, the scratch resistance decreases [3]. Therefore, silica content in the coatings can be adjusted to have an optimum condition between achieving desired levels of hydrophobic properties and sufficient scratch resistance for effective practical application. Silica superhydrophobic coatings are versatile solutions to repel water and protect a surface. Various methods have been applied to drive silica with low surface energy, and stable hierarchical structures have been investigated. The use of silane and fluorinated chemicals in the surface modification of nano-silica has resulted in a contact angle of up to 159.2° [4]. Similarly, incorporating silica and polydimethylsiloxane (PDMS) resulted in coating with 156.4° and sliding angles less than 5° [5]. Both coating methods exhibited good self-cleaning ability, stability, and durability.

From 2016 through 2024, the development of silica-based superhydrophobic coatings has progressed from establishing mechanically robust silica–polymer systems toward scalable, environmentally friendly, and application coatings [6-8]. The coatings have been applied to various substrates, including metals, plastics, glass, fabric, paper, wood, and building materials [9]. The research evolution of silica superhydrophobic coatings is shown in Fig. 1. Despite significant progress, several critical challenges and gaps remain. These include the lack of long-term durability, poor mechanical robustness, limited scalability, and environmental concerns. Furthermore, there is limited research on how different silica morphologies and deposition techniques influence hydrophobic performance. It is essential to discuss these issues in order to demonstrate the commercial viability of silica-based superhydrophobic coatings.

■ SURFACE ENGINEERING OF SILICA

Functionalization Strategies

All highly-cited studies utilized fumed, colloidal, or sol-gel (particle-based) SiO_2; rarely are precipitated, mesoporous (MCM-41/SBA-15), or aerogel forms employed or compared directly [10-12]. Silica particles with uniform size, high purity, ease of control, and scalability are usually obtained via the sol-gel method. This method controls the reaction under acidic or basic conditions over metal alkoxides, such as tetraethylorthosilicate (TEOS), or inorganic salts (i.e., sodium silicate) as precursors. Some researchers also reported the synthesis of silica particles using mineral and waste-based materials such as bagasse ash, rice husk ash, silica sand, fly ash, kaolin, and waste palm kernel.

The hydrophilic properties of silica are generated from the hydroxyl groups attached to its surface. The nature of the silica particle surface can be changed from hydrophilic to hydrophobic through functionalization. Functionalization facilitates covalent bonding between silica particles and hydrophobic agents. Fig. 2 displays the reaction scheme of functionalizing silica with silane as the hydrophobic agent. The alkyl chains of silane replace silica's polar –OH groups, making the surface hydrophobic. Surface functionalization by fluorine-free

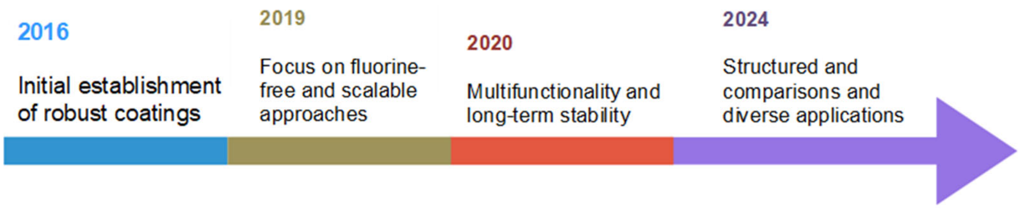

Fig 1. Evolution of durable silica superhydrophobic coatings

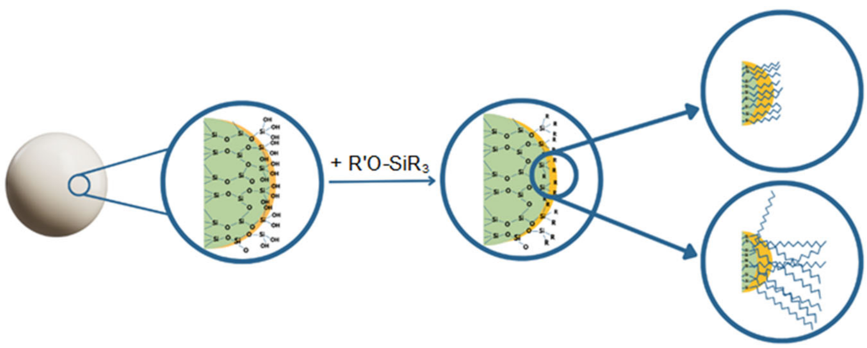

Fig 2. Functionalization of silica surface

approaches achieves contact angles up to 169° and sliding angles <1°, exceeding the fluorinated systems with environmental advantages [5,7,11]. The choice of precursor molecules, such as alkoxysilanes or others, is critical for determining the surface properties. Control reaction, including pH, temperature, and reaction time, is essential to achieve the desired level of hydrophobicity.

There is a synergistic mechanism between silica and low surface energy compounds. However, a further increase in modifier concentration reduces the contact angle due to an uneven coating surface. Some researchers use double silane, like Trimethylchlorosilane (TMCS) and Hexamethyldisilazane (HMDS) to reach maximum CA. The dosage of TMCS as a surface modifier is theoretically twice that of HMDS to achieve superhydrophobic properties [13]. Table 1 explains the various silica sources reported in the preparation of hydrophobic material studies. The silica source's influence on the contact angle of the hydrophobic layer is negligible. Hydrophobicity is more influenced by the type of surface functionalizing agent. Functionalization relies on the formation of self-assembled molecules. The longer the carbon chain of the surface functionalizing agent, the higher the hydrophobicity. However, a longer alkyl chain (C16) demonstrated a low surface height. It was predicted that a long alkyl chain would create self-assembly disorder. Most of the molecules formed a collapsed structure on the surfaces. Molecules tended to form a horizontal orientation on the surface, generating smooth surfaces and high surface energy [14]. The influence of silica concentration has been found to increase hydrophobicity. The silica amount, number, and size of silica aggregates lead to networks that fill the voids and grooves between the assembled silica [15]. If the silica concentration is higher than optimal, the surface morphology of the coating becomes smooth. High silica content fills surface pores and creates a smooth surface [14].

Roughness Control

Superhydrophobic comprises two substances: the roughness component and low surface energy providers. These special properties of superhydrophobic surfaces have made them suitable for applications in various industries, laboratories, and medical fields, such as self-cleaning coatings, anti-biofouling, and anti-icing coatings [3]. To invent new technologies, it is crucial to understand the features and potential applications of superhydrophobic surfaces.

Several methods, including chemical, physical, and combination methods, can fabricate rough superhydrophobic surfaces. Chemical methods involve chemical reactions for specific surface properties. A widely used example is chemical etching, where specific chemicals create rough surfaces. The etching reagents generate different topographies at different levels. Physical methods can achieve rough surfaces through

Table 1. The various silica sources used in the preparation of hydrophobic material

Silica source	Water contact angle using-							
	MTMS	PDMS	TMES	DDTMS	HMDS	TMCS	DMDEOS	SA
TEOS	132° [16]	156° [5], 162° [17]	152° [18]	152° [19]	161° [20]			
Sodium silicate	116° [16]					128.6° [21]		
Rice husk ash	116° [16]	160° [22]						145.3° [23]
Geothermal silica scaling waste						144° [13]		
Kaolin		105° [24]				152° [25]		149.4° [26]
Sugarcane bagasse waste ash							135° [27]	
Palm oil fuel ash		156° [28]						134° [29]
Paper sludge ash								153° [30]
Fly ash geopolymer		159° [31]		152° [32]				

MTMS: Methyltrimethoxysilane, PDMS: Polydimethylsiloxane, TMES: Trimethylethoxysilane, DDTMS: Dodecyldimethylchlorosilane, HMDS: Hexamethyldisilazane, TMCS: Trimethylchlorosilane, DMDEOS: Dimethyldiethoxysilane, SA: Stearic acid physical processes such as mechanical machining, sandblasting, and laser ablation. This method provides a surface with high roughness [33]. For example, sandblasting uses high-pressure air or water onto the surface [34], and laser ablation generates micro and nanostructures by material removal through laser beams. These methods can be applied to a wide range of materials, providing the required surface design.

Hybrid methods combine chemical and physical techniques to create a rough surface that is superhydrophobic. These methods combine surface roughness and superhydrophobicity. Nanoparticle deposition is an excellent instance, which consists of the induction of nanoparticles of different sizes to the surface for hierarchical structures with low surface energy [34]. Dual-size approach (micro/nano-silica blends) claimed to maximize roughness [7,35-36]. Combining micro- (0.5–2 µm) and nano-sized (20–200 nm) silica particles via sequential or simultaneous assembly and raspberry-like particle generating strategies to achieve a robust hierarchical roughness [37].

All methods generate the design of certain surfaces, as illustrated in Fig. 3. Single-scale roughness has surface features (like bumps or pores) that exist at only one scale, typically micro or nano. This surface provides limited roughness, which can somewhat enhance hydrophobicity when combined with a low surface energy coating. However, it often fails to achieve superhydrophobicity (water contact angle ≥ 150°) due to insufficient air trapping, which causes water droplets to penetrate the surface texture. Multiple-scale roughness, known as hierarchical surfaces, is a combination of micro- and nano-scale surface structures. These features significantly increase surface roughness and improve air trapping beneath water droplets. This promotes the Cassie-Baxter state, where water sits on top of surface asperities and air pockets, leading to superhydrophobicity. It mimics natural surfaces like lotus leaves. The last is multiple morphology, a surface composed of silica with varied shapes or structures, not just different scales. This could include spheres, rods, and sheets. The diversity of shapes adds complex surface topography, enhancing surface roughness and texture diversity. The rod-shaped and hollow silica nanoparticles contribute to higher surface roughness. This morphology can result in better water repellency and possibly additional functions like anti-fouling or self-cleaning. The tailoring morphology of these properties enables fine control over wetting properties. However, the surface design of multiple morphologies on the silica superhydrophobic coating is limited.

■ DEPOSITION TECHNIQUES

Deposition techniques control the growth and assembly of molecules, the thickness, and the homogeneity of coating solutions on surfaces. There are various methods available for synthesizing superhydrophobic coating materials, as shown in Fig. 4, for instance, spin coating, dip coating, spray coating, plasma technique, and chemical vapor. Most techniques have achieved beyond 150° contact angle, but spin coating achieves the highest, followed closely by spray

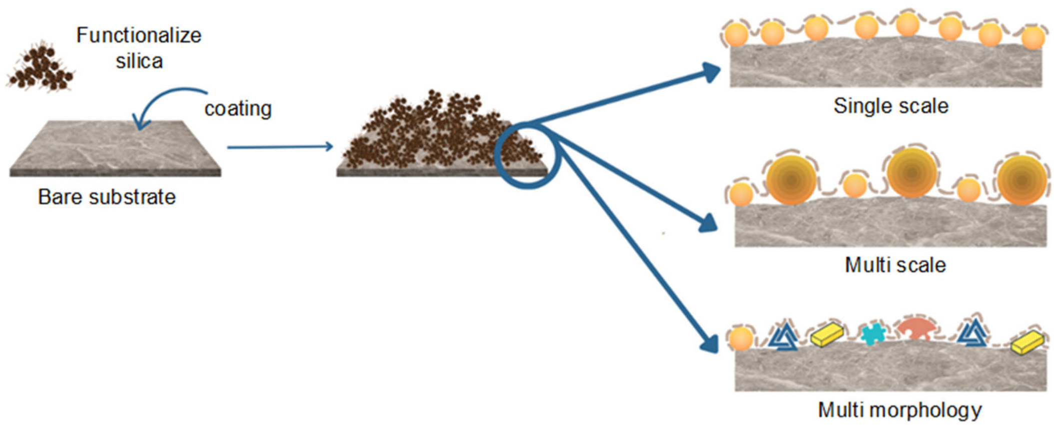

Fig 3. Roughness control of surface using functionalized silica

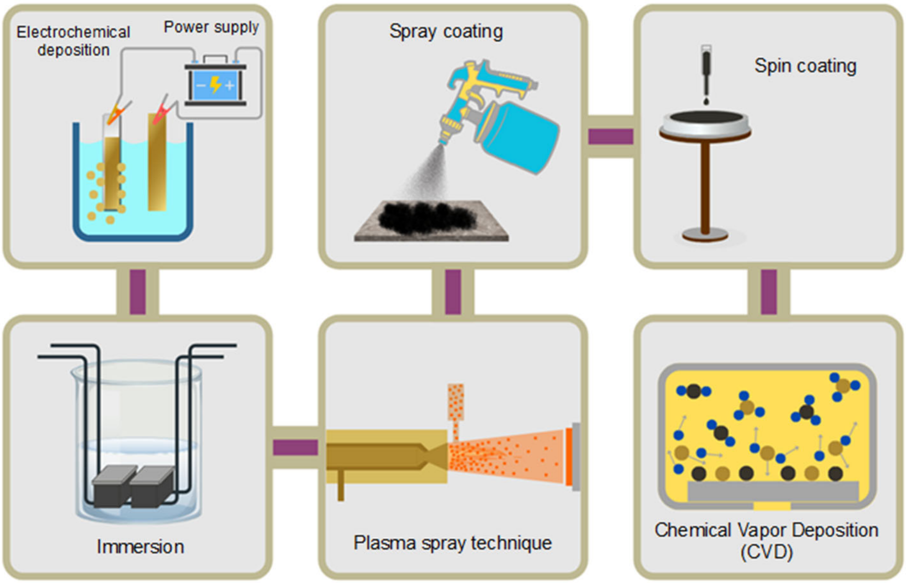

Fig 4. Schematic illustration of deposition techniques

and chemical vapor deposition. Chemical vapor deposition (CVD) provides more uniform films at lower particle concentrations, with superior adhesion and high transparency. The selection of substrate is crucial in the CVD process. This step is critical to ensure the adhesion and homogeneous coating. The plasma technique is suitable for developing superhydrophobic coatings for optical applications where the surface roughness can be adjusted during plasma treatment. However, due to process complexity, this method reaches a lower contact angle or less reproducible results [1].

Layer-by-layer deposition and self-assembly methods is commonly used to generate micro/nanostructures through electrostatic interactions between layers. The layer-by-layer process is a simple deposition technique that creates thin films on surfaces via different electrostatic interactions involving many layers [38]. This method leads to precise thickness and functionality. Various techniques, such as immersion, spin coating, spray coating, and electrochemical deposition, can be used for layer-by-layer surface modification [39]. Hence, other methods can be combined with layer-by-layer to attain the required surface properties and structures.

Sol-gel method reaches highly variable contact angles (115–165°), indicating strong dependence on process parameters [1]. The rate of hydrolysis and condensation reactions, pH, temperature, reaction time, reagent concentration, type and concentration of catalyst, molar ratios of metal precursors, aging time, and drying conditions were reported as factors influencing the properties of coated materials [40]. Several researchers also used wax solidification as a deposition technique. There are several critical factors to consider, such as particle size and wax emulsion concentration. Particle size distribution affects how much liquid wax forms droplets on the surface, which are crucial for achieving high water contact angles [41]. Other factors are the weight percent of wax emulsion, the catalyst amount, and the solvent type [42].

■ THE APPLICATION OF SILICA SUPERHYDROPHOBIC COATING

Researchers have widely fabricated superhydrophobic silica coatings for industrial anti-fog coatings, anti-icing surfaces, water repellency, oil and water separation, self-cleaning, and corrosion resistance. Improving corrosion resistance is one of the critical problems of today's society, and it causes significant damage [43-44]. The principal anti-corrosion performance of silica superhydrophobic coating is superhydrophobic surface layer prevents contact between ions and corrosive surfaces. In addition, this surface coating can reduce the real contact between the corrosive electrolyte and the surface [45]. A silica-based superhydrophobic coating incorporating AZ31B Mg metal alloy surface by spraying showed corrosion resistance [46]. The coating system provides a micro- and nano-scale dual hierarchical texture derived from silica particles. At the same time, the perfluorooctyltriethoxysilane (PFOTES) solution serves as a fluorine silane linker that will make the silica particles hydrophobic. This coating has a water contact angle value of 159.4° and a sliding angle of 1.2°, proving high corrosion resistance.

Superhydrophobicity as an anti-fog inhibits water condensation formation on surfaces such as goggles and glass. Superhydrophobic solution can be made using airless spray and crystal impact methods. The superhydrophobic surface structure allows water to form small droplets and easily roll over them without sticking. This enables these superhydrophobic surfaces to prevent water condensation on the surface and reduce the risk of condensation [47]. The anti-fog coating has been studied through superhydrophobic silica coating on glass substrates through layer-by-layer polystyrene@silica (PS@SiO_2) assembly with high-temperature calcination to remove the core PS. The developed silica coating has a self-cleaning ability and high transparency [48]. Under the condition of two assembly cycles, a superhydrophobic silica coating of WCA of 159 ± 2° with SA of 7 ± 1.5° and high transmittance of 85% was obtained. In addition, the anti-fogging of the superhydrophobic coating was also found. The small fog droplets formed on this coating quickly disappear in about 10 s due to the fast evaporation rate. A silica superhydrophobic coating with anti-icing properties and a water contact angle value of 163 ± 7.4° was successfully assembled from spin coating and chemical vapor deposition methods modified with PFOTES monolayer [49]. Commercial anti-icing products can delay the ice formation by 204 s, while the coated surface improved the icing delay by up to 289 s. This performance is superior to the bare substrate, which can only delay the icing process by 24 s.

The water-repellent properties were also developed in the textile industry. Cotton-based textiles can be modified using various techniques, such as sol-gel deposition of silica nanoparticles. Superhydrophobic fabrics are commonly used to separate oils from water. The manufacture of superhydrophobic composition coatings on the surface of fiberglass fabric was carried out through the formation of amino-silica particles and hydrophobization using octadecyltrichlorosilane [50]. The water-repellent and self-cleaning properties was also investigated in the conservation field. The development of hydrophobic coatings using silica combined with fatty acids was aimed at enhancing the surface properties of andesite stone. Due to hydrogen bonding, the silica-palmitic acid coatings on andesite demonstrated better stability and performance than silica-stearic acid coatings [51].

■ RESEARCH GAPS OF SUPERHYDROPHOBIC COATING

Comparative Gaps of Silica Types and Morphologies

Recent literature provides strong evidence that upscale silica-based superhydrophobic coatings primarily using fumed, colloidal, spheres, rods, and porous combined with low surface energy compounds can achieve robust water repellency. The silica shape and morphology influence the coating performance of the surface structure. Multiscale particles deliver superior mechanical and wetting durability across a range of environmental conditions. However, the effect of silica shape (rod, sphere, fiber, hollow, etc.) has not been studied. The optimized shape improves repellency, transparency, mechanical stability, and material compatibility.

Adhesion and Durability

The development of superhydrophobic coatings encounters considerable problems concerning adhesion and durability [52]. Poor adhesion is indicated by the ease of removing the coating, scratch, or damage by mechanical forces. While some coatings exhibit excellent mechanical stability, others degrade under a high pH environment or heavy abrasion [53]. These challenges require novel approaches to enhance adhesion properties while maintaining their superhydophobicity [54]. Moreover, existing superhydrophobic surfaces exhibit low durability. These films are generally very hydrophobic and have good self-cleaning properties in the short term. Contaminants on the surface tend to decrease the water-repellent properties of coatings. Water tends to spread over the hydrophilic surface. Some contaminants still remain on the surfaces, resulting in unclear surfaces. Most stability and aging tests stay 120 days outdoors or less than 1000 h under UV exposure. However, the long-term test period, which is longer than 6 months in the field, is incomplete. Commercial products or supporting industrial processes need long-term stability and durability with regard to superhydrophobicity [55-56].

Scalability and Cost-Effectiveness

Another gap is the scalability and cost-effectiveness of production methods. Scalability and production issues also pose significant barriers to the development of superhydrophobic coatings [52]. Hence, it is essential to note that these coatings can be reproduced on a large scale with consistent quality. This often requires specific control over surface structures and material composition for superhydrophobic coatings to retain their desired attributes. But, going from lab-scale to commercial production involves many aspects like economics, environmental, and engineering. Many current techniques, such as laser texturing, are complex and expensive, limiting their industrial applicability [57].

Environmental Issues

To develop high-performance superhydrophobic materials, several environmental concerns include sustainability aspects such as reducing contamination and improving cleanliness. The production processes and the choice of reactants may raise environmental issues, such as green chemistry. Traditional methods still use harmful chemicals with adverse environmental effects or hardly recycled properties [58]. Addressing these issues requires sustainable materials and manufacturing techniques like green chemistry principles using natural resources to make eco-friendly hydrophobic surfaces [59]. Maintaining a balance between performance, durability, and environmental health is desirable in developing superhydrophobic for a sustainable future.

Application Diversity

Many applications of superhydrophobic coating, such as self-cleaning, anti-icing, corrosion, optics, and drag reduction on varied substrates, have been reported. Furthermore, there will be applicative coatings with multifunctional properties such as simultaneous superhydrophobicity and antibacterial activity. While some studies have incorporated antibacterial agents like copper, further research is required to explore the modification of other antimicrobial materials into silica-based coatings [60]. Additionally, the lack of standardization in testing protocols for superhydrophobic coatings hinders the comparison of their performance in different studies.

■ THE FUTURE PROSPECT OF SILICA SUPERHYDROPOBIC COATING

Silica superhydrophobic coating, which has outstanding water-repellent properties, holds great promise in different industries. However, the promise shown by silica superhydrophobic coating still requires further validation of its real-world applicability. The excellent lab-scale results on silica waterproofness are far from the industrial scale because of many aspects, such as various surfaces, environmental exposure, and long-term behavior. It is crucial to address any limitations or challenges that might arise from silica-superhydrophobic.

The proposed strategies to fabricate the silica superhydrophobic coating are strict and aim to create robust superhydrophobicity through hierarchical structuring (dual/multi-scale and low surface energy surface treatments). The fluorine-free formulations and scalable fabrication using easy deposition techniques such as spray coating are promising for sustainable and large-scale applications. Incorporating anti-fouling and self-healing mechanisms into the coating could solve the durability issue. By combining self-healing properties, superhydrophobic coatings can repair damage and maintain water-repellent properties. The presence of anti-fouling properties implies the prevention of the accumulation of contaminants. This means the durability of coatings with anti-fouling properties that have extended periods over conventional superhydrophobic. Combining anti-fouling properties with self-healing mechanisms allows a promising application of superhydrophobic coatings in various industries.

The cost of implementing silica superhydrophobic coating is a challenge for specific industries. This technology requires an initial high-cost investment for small companies or startups. These technologies also require skilled technicians and specialized equipment, which affects their overall cost. An economic analysis must compare the price of silica superhydrophobic coatings with conventional paints. Although silica superhydrophobic coating shows excellent properties, the cost-effectiveness must be calculated. While the benefits are evident, the financial aspect also shows cost-effectiveness; these technologies are ready to be applied for widespread adoption across industries.

■ CONCLUSION

Silica superhydrophobic coatings have demonstrated potential applications in self-cleaning, anti-corrosion, anti-icing, and drag reduction. The development of surface functionalization, micro-nano roughness control, and deposition techniques has enhanced their hydrophobic performance. However, challenges remain in adhesion, scalability, and cost-effectiveness. Future research should focus on developing robust, environmentally friendly, cost-effective fabrication techniques that can be manufactured efficiently at the industry level.

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■ CONFLICT OF INTEREST

The authors declare that there is no conflict of interest.

■ AUTHOR CONTRIBUTIONS

Alfa Akustia Widati developed the concept, designed the manuscript, and analyzed data. Andina Fitriyah Salsabilah and Aisyah wrote and revised the manuscript. Zeni Rahmawati and Wan Nazwanie Wan Abdullah provided technical assistance in collecting data. All authors agreed to the final version of this manuscript.

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