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# One-step synthesis of SiO<sub>2</sub> nanomesh for antireflection and self-cleaning of solar cell



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## A B S T R A C T

SiO<sub>2</sub> nanomaterials are widely used for antireflection and self-cleaning, but the preparation process is usually complex and time-consuming. Hence, we present a facile one-step synthesis of a hydrophobic two-dimensional SiO<sub>2</sub> nanomesh by tuning the reaction temperature using dodecylamine as a catalyst. SiO<sub>2</sub> nanomesh has the advantages of an adjustable refractive index, simple preparation process, and low cost, which affords both antireflection and self-cleaning functions for solar cells. Two types of per-ovskite solar cells were used to verify the stability and universality of the SiO<sub>2</sub> nanomesh coatings. The antireflection effect of the SiO<sub>2</sub> nanomesh is found to increase the current density of both perovskite solar cells fabricated at 500 °C and 150 °C, with the efficiency increased by 4.48% and 4.79%, respectively.

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1. Introduction

Organic-inorganic halide perovskite materials have made great progress in photovoltaic devices due to the advantages of a high absorption coefficient, long charge carrier recombination lifetime,

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high charge carrier mobility, and tunable band gap [1–2]. Perovskite solar cells (PSCs) have achieved an efficiency record of 25.7% after only ten years of development, which is comparable to that of silicon solar cells [3]. Many valid strategies have been proposed to further improve the performance of PSCs, including energy level alignment, additive engineering, defect passivation, etc [4–6]. However, the problem of light reflection loss in PSCs has not been satisfactorily solved so far [7].

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Since PSCs are generally fabricated on glass or polymer films (such as PET, PEN, PDMS) [8–11], which all have a low refractive index, a material with a lower refractive index is required to reduce light reflection [12]. SiO<sub>2</sub> has been widely used in self-cleaning and antireflection for the above substrates due to the advantages of a low and tunable refractive index, controllable size, wide raw material reserves, and easy modification [13–16]. However, large-size SiO<sub>2</sub> is not suitable for antireflection on a PSC substrate due to the strong light scattering brought by the considerable roughness [17,18].

Chemical preparations of small-size (<50 nm) SiO<sub>2</sub> nanoparticles demand harsh conditions and long periods [19–22]. For example, Cecilia Agustín-Sáenz et al. [20] prepared silica sols for glass antireflection by the sol-gel method, which typically required two steps and several days of aging. However, preparation based on vapor deposition and other methods is relatively dependent on equipment; and is likely to desire the use of dangerous gases [23–25]. For example, Myungkwan Song et al. [23] used PECVD (Plasma Enhanced Chemical Vapor Deposition) to fabricate a SiO<sub>2</sub> light trapping structure on PET for organic solar cells, which was carried out in a mixed gas of HMDSO (hexamethyldisiloxane)-O<sub>2</sub>-Ar. In addition, since the stability of perovskite material is greatly affected by water and high temperature [2], many methods for preparing SiO<sub>2</sub> antireflection films on PSC are limited by aqueous phase or high-temperature treatment [18–20,22,26].

To solve these problems, we first proposed a method to hydrolyze tetraethyl orthosilicate (TEOS) with dodecylamine (DDA) as a catalyst [27–29], synthesizing SiO<sub>2</sub> nanomesh by one-step temperature regulation for antireflection and self-cleaning of PSC. Our results show that the SiO<sub>2</sub> nanomesh has excellent hydrophobic and antireflection performance. Benefiting from the increased short-circuit current resulting from the antireflection property, the efficiencies of two kinds of PSCs prepared at 500 °C and 150 °C are found to be increased by 4.48% and 4.79%, respectively, proving the stability and universality of the SiO<sub>2</sub> nanomesh.

#### 2. Experiments and methods

#### 2.1. Materials

Tetraethyl orthosilicate (TEOS, 99.9%), and dodecylamine (DDA, 98%) were purchased from Aladdin, China. Anhydrous ethanol (EtOH, 99.7%) was supplied by Guangfu Chemical Reagent Factory, China. Methylammonium iodide (CH<sub>3</sub>NH<sub>3</sub>I, 99.5%) and lead iodide (Pbl<sub>2</sub>, 99.9%) were acquired from Xi'an Polymer Light Technology Co., Ltd., China. Dimethyl formamide (DMF, 99.8%), dimethyl sulfoxide (DMSO, 99.8%), and chlorobenzene (CB, 99.5%) were acquired from Sigma-Aldrich. The ITO-coated glasses, high-temperature (HT) and low-temperature (LT) TiO<sub>2</sub> precursor agents, and spiroMeOTAD were purchased from MaterWin New Materials Co., Ltd., China.

#### 2.2. Synthesis of the SiO<sub>2</sub> nanomesh

The method of controlling the temperature to inhibit the hydrolysis of the silicon source was used to synthesize a  $SiO_2$  nanomesh. Briefly, 3 ml of TEOS and 35 ml of EtOH mixed solution were uniformly dispersed by ultrasonication, and then the mixed solution was quickly added to a flask containing 20 ml of EtOH, 35 ml of distilled water, and 0.4 g of DDA under high-speed stirring. The reaction was continued under stirring for 4 h with different water bath temperatures. The products were centrifuged 6 times with EtOH and distilled water alternately, for 10 min each time at 12,000 rpm.

#### 2.3. Preparation of perovskite solar cells

The glass substrates were sequentially cleaned in an ultrasonic bath consisting of diluted detergent, deionized water, acetone, and isopropanol for 15 min in each step, followed by ultraviolet ozone treatment for 20 min. The TiO<sub>2</sub> layer was prepared by spin-coating using HT TiO<sub>2</sub> agent on ITO glass at 4000 rpm for 30 s with annealing at 500 °C for 60 min or LT TiO<sub>2</sub> agent on glass at 3000 rpm for 30 s with annealing at 150 °C for 30 min. ITO glasses was used as the substrates for the two types of TiO<sub>2</sub> for the convenience of comparison. Samples were treated with ultraviolet ozone for 15 min and then transferred into a glove box. The MAPbI<sub>3</sub> layers were rapidly deposited onto TiO<sub>2</sub> substrates by a spin coating process at 500 rpm for 5 s and 4000 rpm for 30 s. During the MAPbI<sub>3</sub> layer spin-coating process, chlorobenzene was injected onto the substrate starting at the 20th second. After several minutes, all the samples were then annealed at 100 °C for 20 min with a cover plate. Waiting for the sample to cool to room temperature, the SpiroOMeTAD precursor was then deposited on MAPbI<sub>3</sub> layers by spin coating at 5000 rpm for 30 s. Finally, Au electrodes were deposited by vacuum thermal evaporation.

#### 2.4. Preparation of SiO<sub>2</sub> antireflection coatings

Pre-fabricated SiO<sub>2</sub> powder dispersed in isopropanol solution (10 mg/ml) was sonicated for 20 min. A total of 60  $\mu$ L of SiO<sub>2</sub> suspension was drop cast onto the front surface of the PSCs, and SiO<sub>2</sub> nanomesh coatings were prepared by spin-coating at 2000 rpm for 30 s. The samples were then annealed at 70 °C for 10 min on a heating table.

#### 3. Results and discussion

 $SiO_2$  nanomesh was synthesized with TEOS as the silicon source and DDA as the catalyst. The total reaction is mainly divided into (1) hydrolysis reaction, (2) condensation reaction, and (3) polymerization reaction. First, TEOS (Si(OEt)<sub>4</sub>) undergoes a hydrolysis reaction in an alcohol/water mixed solution to generate silanol groups (Si(OEt)<sub>4-x</sub>(OH)<sub>x</sub>). After that, a dehydration condensation reaction occurs between hydroxyl groups to produce siloxane oligomers. At the same time, Si(OEt)<sub>4</sub> can also undergo a dealcohol condensation reaction with Si(OEt)<sub>4-x</sub>(OH)<sub>x</sub>. Finally, the condensation and polymerization reactions further induce the nucleation and growth of SiO<sub>2</sub> particles within a certain reaction period.

$$Si(OEt)_4 + xH_2O \Rightarrow Si(OEt)_{4-x}(OH)_x + xEtOH$$
 (1)

$$2Si(OEt)_{4-x}(OH)_x \rightleftharpoons (EtO)_{8-2x}(Si-O-Si)_1(OH)_{2x-2} + H_2O$$
(2)

$$Si(OEt)_4 + Si(OEt)_{4-x}(OH)_x \rightleftharpoons (EtO)_{7-x}(Si-O-Si)_1(OH)_{x-1} + EtOH$$
(3)

Owing to the sharp increase in the reaction rate due to an increased temperature, the hydrolysis reaction is rapidly accelerated. Rising the temperature to close to the boiling point of ethanol results in evaporation of the solvent. Consequently, the effective collision probability between DDA and the reactants is greatly reduced, resulting in the restrained condensation and polymerization reactions [30,31]. In addition, evaporation of the solvent gives the nucleated particles upward momentum to overcome aggregation difficulties. As a result, two-dimensional (2D) nanomesh instead of three-dimensional (3D) structures are produced in a limited reaction time. SiO<sub>2</sub> nanomesh has a remarkable hydrophobic performance, because DDA remains in the interior and surface of the SiO<sub>2</sub> product during the reaction.

Typical TEM images of SiO<sub>2</sub> micro/nano products under different reaction temperatures are shown in Fig. 1. For temperatures below 35 °C, regular SiO<sub>2</sub> microspheres can be obtained. At 20 °C, flocculent residues can be observed on the surface of SiO<sub>2</sub>, indicating that the reaction is not complete at this temperature, and the SiO<sub>2</sub> spheres can grow larger. After the temperature reaches 35 °C, it is difficult to obtain spherical 3D products. As the temperature is further increased to 45 °C, both the monodispersity and size of the product are further reduced, forming a 2D nanomesh structure with interconnected small nanoparticles, since the increased temperature leads to an increased number of nucleations, resulting in a decrease in the size of the product [30]. When the temperature exceeds 45 °C, the size of the product increases slightly, and the yield decreased sharply. Noted that almost no product appears at 55 °C, which is not involved in the characterizations.



**Fig. 1.** TEM images of the SiO<sub>2</sub> nanomesh prepared temperature at (a) 20 °C, (b) 25 °C, (c) 30 °C, (d) 35 °C, (e) 40 °C, (f) 45 °C, (g) 50 °C, (h) 55 °C.

To explore the effect of different reaction temperatures on the SiO<sub>2</sub> products, we analyzed the products prepared at 25 °C, 35 °C, 45 °C, and 50 °C by infrared (IR) spectroscopy and ultraviolet-visible (UV-Vis) absorption spectroscopy. Fig. 2a draws the IR spectrum for the SiO<sub>2</sub> product synthesized using DDA as the catalyst. The peaks at 2860 cm<sup>-1</sup> and 2931 cm<sup>-1</sup> are attributed to typical C-H bond stretching, which is mainly derived from DDA considering the efficient removal of residual TEOS after multiple high-speed centrifugation washes. The UV-Vis absorption spectra in Fig. 2b exhibit that the SiO<sub>2</sub> products attract a gradual decay in absorption intensity with increasing preparation temperature, achieving a minimum value at 45 °C; however, a slight increase in absorption at 50 °C is observed over the wavelength range of 360–800 nm. Since the antireflection structure for solar cells requires weak parasitic absorption, SiO<sub>2</sub> nanomesh prepared at 45 °C is the best appropriate candidate.

To clarify the elemental and chemical states of the  $SiO_2$  nanomesh, Fig. 3a demonstrates the survey (X-ray photoelectron spectroscopy) XPS data measured from 0 to 1250 eV. The peaks observed on the curves of  $SiO_2$  nanomesh are assigned to Si, O, N, and C elements in Fig. 3(b-e). The uniformly symmetric single peaks located at 103.3 eV and 532.7 eV can be indexed to Si 2p and O 1s, respectively, which are attributed to Si-O bonds [32], confirming the existence of amorphous  $SiO_2$  in the product. The weak peak with a binding energy of 401.7 eV is attributed to N 1s [33], which can only be derived from DDA. The C 1s spectrum contains two sets of peaks at 284.6 eV and 285.8 eV corresponding to C—C and C—N bonds, respectively, which are consistent with the characteristic peak of the N 1s spectrum.

The SiO<sub>2</sub> volume fraction can be adjusted by calcining to remove DDA, thereby changing the optical properties [34–36]. SiO<sub>2</sub> was calcined in a tube furnace for 2 h at 300 °C, 450 °C, 600 °C, and 750 °C. The mass loss after calcination is drawn in Fig. 4a. When calcined at 300 °C, the mass losses for the microspheres and nanomesh is 14.83% and 21.98%, respectively. With increasing calcination temperature, the two mass losses gradually approach each other, reaching values of 26,15% and 28,01% at 750 °C, since the DDA as well as a little water inside the microspheres requires higher energy to escape compared to that for the SiO<sub>2</sub> nanomesh. In order to further verify the formation mechanism of silica products at different temperatures, the products were further calcined at 750 °C. As shown in Fig. 4b, the mass losses for the SiO<sub>2</sub> products prepared at 20–50 °C proceed through multiple stages with increasing preparation temperature. As the diameter of the SiO<sub>2</sub> microspheres decreases, the mass loss reaches a minimum value of 24.04%. The mass loss of the product at 35 °C rises sharply to 47.92% due to poor spherical structure and monodispersity, and a minimum value of 28.01% is obtained again when the synthesis temperature reaches 45 °C, with the product forming a nanomesh structure at this temperature. SiO<sub>2</sub> nanomesh does not produce structural changes at 50 °C; however, the sizes of the linked particles are increased, which leads to an improvement in the mass loss during calcination.

Due to the excellent monodispersity of the pre-prepared SiO<sub>2</sub> microspheres, uniformly arranged single-layer SiO<sub>2</sub> microspheres coatings can be obtained by the spin-coating method [37], as shown in Fig. 5a. The SiO<sub>2</sub> nanomesh was fabricated based on the same method, and the pores of the SiO<sub>2</sub> nanomesh are shaded to a certain extent in Fig. 5b. The coating films of both structures show good hydrophobicity with water contact angles of 98° for SiO<sub>2</sub> microspheres (Fig. 5c) and 89° for SiO<sub>2</sub> nanomesh (Fig. 5d), indicating the good performance for self-cleaning and prevention of water ingress. Owing to the excellent hydrophobicity and chemical inertness, the SiO<sub>2</sub> nanomesh film can be used to improve the durability of PSCs. However, the water contact angles are reduced to 21° and 0° after calcination at 750 °C, demonstrating the



Fig. 2. (a) IR spectra and (b) UV–Vis absorption spectra of SiO<sub>2</sub> products prepared at different temperatures.







Fig. 4. (a) Mass loss of SiO<sub>2</sub> nanostructures at different annealing temperatures, (b) Mass loss of SiO<sub>2</sub> materials prepared at different temperatures annealed at 750 °C.



Fig. 5. SEM images of (a) SiO<sub>2</sub> microspheres (25 °C) and (b) SiO<sub>2</sub> nanomesh (45 °C) coated ITO glass substrate. The water contact angles of (c) SiO<sub>2</sub> microspheres and (d) nanomesh.

hydrophobic properties contributed by DDA. Although theoretically the calcined mesoporous  $SiO_2$  nanomesh has better optical properties with a tunable refractive index, the calcined  $SiO_2$  nanomesh also loses the hydrophobic self-cleaning function. Furthermore, the  $SiO_2$  nanomesh structure is destroyed after calcination, as shown in Fig. S1, but the dispersion is greatly improved. Therefore, calcined  $SiO_2$  nanomesh is not suitable for application in perovskite solar cells, but can be applied in other crucial fields, such as chemical carriers, drug delivery, and radiative cooling [38–40]. The adsorption-desorption isotherms and corresponding pore size distributions of  $SiO_2$  nanomesh are exhibited in Fig. S2.

The antireflection performance of the  $SiO_2$  product for ITO glass is plotted in the transmission spectrum, as shown in Fig. 7. The transmittance of the ITO glass coated with  $SiO_2$  microspheres is severely reduced, because  $SiO_2$  microspheres induce a significant increase in the roughness of the glass surface, enhancing light scattering. In contrast, the roughness of the  $SiO_2$  nanomesh is hardly



Fig. 6. Transmittance spectra of  $SiO_2$  nanospheres and nanomesh coated ITO glass substrate.

increased, but the reflection of the air-glass interface is reduced due to the appropriate refractive index. Compared with the blank one, the transmission of ITO glass with  $SiO_2$  nanomesh coating is improved over the wavelength of 300–800 nm, and the largest improvement in the transmission from 76.45% to 81.73% occurs at a wavelength of 440 nm.

The transmission of the low-roughness SiO<sub>2</sub> nanomesh coating can be explained by the equivalent refractive index corresponding to the SiO<sub>2</sub> volume fraction in the film [12]. The effective refractive index ( $n_{\text{eff}}$ ) of the SiO<sub>2</sub> nanomesh coating can be obtained according to the volume fraction  $\alpha$  using Eq. (4).

$$n_{eff} = \left[ (1 - \alpha)n_{air}^2 + \alpha n_{SiO_2}^2 \right]^{1/2}$$
(4)

$$R = \left(\frac{n_{\rm air} n_{\rm glass} - n_{\rm eff}^2}{n_{\rm air} n_{\rm glass} + n_{\rm eff}^2}\right)^2 \tag{5}$$



**Fig. 7.** The effective refractive index of the SiO<sub>2</sub> nanomesh coating (left), calculated reflectance for quarter-wavelength SiO<sub>2</sub> nanomesh coating on glass (right).

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The interference cancellation of the incident and reflected light at the two interfaces achieves the ideal antireflection effect when the thickness (d) of the antireflection film satisfies the quarterwavelength relationship  $(d = \frac{\lambda}{4n_{\text{eff}}})$ . The reflection of the air-glass interface adjusted by the SiO<sub>2</sub> nanomesh under a quarterwavelength thickness is obtained according to Eq. (5). Here the refractive index of air (n<sub>air</sub>) is 1, and the refractive index of glass (n<sub>glass</sub>) is 1.5. When the volume fraction of the SiO<sub>2</sub> nanomesh varied from 0.35 to 0.55 is an ideal interval, as drawn in Fig. 6, the reflection is almost reduced from 4.3% to 0%. Therefore, a suitable antireflection coating can reduce the absolute value of the reflectance for PSCs by more than 4% considering the air-glass interface and other interfaces.

Two kinds of PSCs were fabricated using HT TiO<sub>2</sub> (500 °C) and LT TiO<sub>2</sub> (150 °C) agents to verify the universality of SiO<sub>2</sub> nanomesh on rigid and potential flexible devices [41]. As shown in Fig. 8a-b, the PSC has a typical 5-layer structure, including a glass substrate with an ITO conductive layer, followed by TiO<sub>2</sub> as an electron transport layer, MaPbI<sub>3</sub> as the active layer, SpiroOMeTAD as the hole transport layer, and an Au layer as the top electrode. To compare the effects of different TiO<sub>2</sub> layers on PSCs, Fig. 8c-d plots the plane



Fig. 8. SEM cross-sectional image of the PSC. SEM images and corresponding grain size distributions of perovskite films with (c) HT and (d) LT TiO<sub>2</sub> substrates.



Fig. 9. (a) XRD (X-Ray Diffraction) pattern and (b) PL (photoluminescence) spectrum of perovskite thin film on TiO<sub>2</sub> layer.

scans and grain size distributions for perovskite thin films prepared on TiO<sub>2</sub> layers. The average grain size of the perovskite film on the HT and LT TiO<sub>2</sub> layer is 617 nm and 242 nm, respectively.

The crystallization of perovskite films can also be observed from the XRD spectra. Fig. 9a shows that the MAPbI<sub>3</sub> film prepared on HT TiO<sub>2</sub> substrate possesses the stronger peaks than that on LT TiO<sub>2</sub> substrate located at 14.2° and 28.5° corresponding to the (110) and (220) crystal planes, which verifies the better perovskite crystallization for the MAPbI<sub>3</sub> film on HT TiO<sub>2</sub> than that on LT TiO<sub>2</sub> [42]. Therefore, the emission peak at 742 nm for the MaPbI<sub>3</sub> perovskite film on HT TiO<sub>2</sub> is weaker than that on LT TiO<sub>2</sub> [41] based on better crystalline properties and less radiation loss in Fig. 9b.

The photovoltaic performance of the PSCs coated with  $SiO_2$  nanomesh was tested for under a standard light intensity of 100 mW/cm<sup>2</sup>. Representative *J*-*V* curves of MaPbI<sub>3</sub>-type PSCs based on different  $TiO_2$  layers are plotted in Fig. 10a-b, and the corresponding photovoltaic performance parameters are listed in

Table 1. The perovskite device fabricated using a mature HT TiO<sub>2</sub> agent has a  $I_{SC}$  (short-circuit current density) of 21.18 mA/cm<sup>2</sup>,  $V_{\rm OC}$  (open-circuit voltage) of 1.098 V, FF (fill factor) of 74.48%, and  $\eta$  (efficiency) of 17.18%. Due to the antireflection effect of the SiO<sub>2</sub> nanomesh, the  $\eta$  of PSC is increased by 4.48%, reaching a value of 17.95%. The improvement in  $\eta$  is contributed by the contribution of J<sub>SC</sub>. As displayed in Fig. S3, the current density contribution is calculated to be 1.03 mA/cm<sup>2</sup> according to the transmittance difference in Fig. 6, which is slightly higher than 0.95 mA/cm<sup>2</sup> determined from the *I-V* curve. The small difference is mainly due to the weak absorption of perovskite near a wavelength of 800 nm and the structural difference between PSC and ITO glass [43,44]. Similarly,  $\eta$  is increased from 15.88% to 16.64% due to the SiO<sub>2</sub> nanomesh coating for the PSC structured by LT TiO<sub>2</sub>, corresponding to a percentage improvement of 4.79%. To verify the stability of the PSCs. 10 sets of the same short-circuit current density and efficiency parameters are plotted in Fig. 10c-d. Generally, the stability of PSCs prepared by LT TiO<sub>2</sub> is better than



**Fig. 10.** *J-V* curves of (a) High-temperature and (b) Low-temperature fabricated perovskite solar cells without and with SiO<sub>2</sub> nanomesh coatings. Distribution of (c) current density and (d) efficiency parameters of perovskite solar cells with and without SiO<sub>2</sub> nanomesh coatings.

Table 1
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Performance parameters of perovskite solar cells decorated with SiO<sub>2</sub> nanomesh.

Fabricating temperature	SiO <sub>2</sub> nanomesh coating	V <sub>oc</sub> (V)	J <sub>sc</sub> (mA/cm <sup>2</sup> )	FF (%)	η (%)
500 °C	W/O	1.089	21.18	74.48	17.18
	With	1.089	22.13	/4.4/	17.95
150 °C	W/O	1.096	20.90	69.36	15.88
	With	1.096	21.88	69.35	16.64

that of HT TiO<sub>2</sub>. The improved performance for both of these two PSCs demonstrates the excellent applicability and stability of the SiO<sub>2</sub> nanomesh.

#### 4. Conclusions

In conclusion, we have developed a temperature-controlled method based on DDA as a catalyst to prepare the hydrophobic SiO<sub>2</sub> nanomesh in one step. The method has the advantages of simple operation, short preparation time, and no dependence on complex equipment. The water contact angle of the SiO<sub>2</sub> nanomesh reaches 89° without further modification, providing excellent self-cleaning performance for PSCs. In addition, the SiO<sub>2</sub> nanomesh coating leads to an efficiency increase of 4.48% and 4.79% for PSCs prepared at 500 °C and 150 °C, respectively, which is based on the remarkable optical antireflection performance of the SiO<sub>2</sub> nanomesh. This improvement is weakly dependent on the reference solar cells, which provides vital guidance for efficient solar cells.

#### **CRediT** authorship contribution statement

Daxue Du: Conceptualization, Investigation, Methodology, Data curation, Writing - original draft. Fengyan Wang: Investigation, Conceptualization. Dezhao Zhang: Data curation, Formal analysis. Jiahao Bao: Data curation. Yunhao Fan: Formal analysis. Yikai Guo: Validation. Wenzhong Shen: Resources, Funding acquisition. Haiyan Wang: Writing - review & editing, Project administration.

#### Data availability

Data will be made available on request.

#### **Declaration of Competing Interest**

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

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jcis.2022.10.157.

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