Fabrication of Bragg Mirrors by Multilayer Inkjet Printing

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Bragg mirrors are widely applied in optical and photonic devices due to their capability of light management. However, the fabrication of Bragg mirrors is mainly accomplished by physical and chemical vapor deposition processes, which are costly and do not allow for lateral patterning. Here, the fabrication of Bragg mirrors by fully inkjet printing is reported. The photonic bandgap of Bragg mirrors is tailored by adjusting the number of bilayers in the stack and the layer thickness via simply varying printing parameters. An ultrahigh reflectance of 99% is achieved with the devices consisting of ten bilayers only, and the central wavelength of Bragg mirrors is tuned from visible into near-infrared wavelength range. Inkjet printing allows for fabricating Bragg mirrors on various substrates (e.g., glass and foils), in different sizes and variable lateral patterns. The printed Bragg mirrors not only exhibit a high reflection at designed wavelengths but also show an outstanding homogeneity in color over a large area. The approach thus enables additive manufacturing for various applications ranging from microscale photonic elements to enhanced functionality and aesthetics in large-area displays and solar technologies.

1. Introduction

Bragg mirrors (also known as dielectric mirrors) have been widely investigated due to their fascinating capability of optical manipulation and structural coloration. A Bragg mirror is a device based on a 1D photonic crystal (1DPC), consisting of a structure where the refractive index (RI) is periodically distributed along one dimension in space.\(^\text{[1]}\) Bragg mirrors can be built by alternating two materials with different RI or tuning the porosity of one material.\(^\text{[2]}\) Due to the multilayer interference effect, Bragg mirrors possess a photonic bandgap (PBG) analogous to the electronic bandgap in semiconductors.\(^\text{[3]}\) Electromagnetic waves at specific frequencies cannot propagate inside these media. By tailoring the stacking sequence, layer thickness, and material composition, it is possible to tune the photonic stopband to meet almost any desired characteristic, e.g., wavelength-selective filters and ultrahigh reflectivity mirrors. Dielectric laser mirrors and dichroic beamsplitters are essential discrete elements of different optical systems. Distributed Bragg reflectors (DBR) are used in various integrated optical components and systems.\(^\text{[4]}\) In addition, Bragg mirrors are essential components in highly sensitive optical sensors\(^\text{[5]}\) and are applied as functional and colorful coatings for solar cells.\(^\text{[6]}\) Furthermore, high-reflectance photonic crystals are widely applied in the field of radiative cooling.\(^\text{[7]}\)

Various materials have been used to fabricate Bragg mirrors, including inorganic,\(^\text{[8]}\) organic,\(^\text{[3,9]}\) and hybrid materials.\(^\text{[10]}\) The contrast in the RI of the constituent materials is preferably higher in favor of fabricating more efficient Bragg mirrors. Different fabrication methods have been applied to manufacture Bragg mirrors: Physical vapor deposition (PVD)\(^\text{[11]}\) and chemical vapor deposition (CVD)\(^\text{[12]}\) are widely used in industry. CVD-based dielectric mirrors are used for realizing vertical-cavity surface-emitting lasers (VCSELs).\(^\text{[13]}\) This technology, however, is very specialized and time-consuming. In addition, both methods need to be combined with lithographic patterning to obtain lateral structures. More recently, solution-based processes such as spin coating,\(^\text{[14,15]}\) dip coating,\(^\text{[15]}\) blade coating,\(^\text{[16]}\) self-assembly methods,\(^\text{[17]}\) and holographic photopolymerization\(^\text{[18]}\) have emerged as attractive methods.

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striations and comets which lead to poor stack quality.\textsuperscript{[19]} Blade coating, on the other hand, can be used for large-scale production; however, it is very challenging to have good control of the thin film quality.\textsuperscript{[20]} Using self-assembly is time-consuming, and also it is hard to land with high reproducibility.\textsuperscript{[21]} Moreover, the lateral definition of Bragg mirrors is not possible with any of the abovementioned approaches. A more detailed comparison of different solution processing methods is shown in Table S1 (Supporting Information).

Inkjet printing, a straightforward, versatile, low-cost, and high-throughput technique,\textsuperscript{[22]} is a highly promising method for fabricating Bragg mirrors. First, it provides a cost-effective solution and has the potential to be implemented in large-scale roll-to-roll manufacturing. Second, lateral patterning of Bragg mirrors down to the <100 μm range is feasible, thus enabling the maskless additive manufacturing of optical filter/mirror arrays. Besides, the Bragg mirror printing process can be easily transferred to different substrates. Last but not least, the fast prototyping of Bragg mirrors can be realized by inkjet printing. However, it is very challenging to develop suitable inks and establish a reliable multilayer fabrication process. In terms of the formulation of the inks, they should contain constituents that give a relatively high RI contrast while meeting the orthogonal solubility condition. In addition, every layer needs to be as uniform as possible to ensure a uniform multilayer stack. Moreover, it is crucial to have precise control and a high reproducibility on the film thickness so as to achieve superior optical performance, e.g., high reflectance of the Bragg mirrors.

To the best of our knowledge, we report herein the first fully digitally fabricated Bragg mirrors prepared by a commercial desktop inkjet printer. We use a high RI contrast material pair of printable nanoparticulate titanium dioxide (TiO\textsubscript{2}) and poly-(methyl methacrylate) (PMMA) in inks. An ultrahigh reflectance of ≈99% was achieved with ten bilayers only, while 99% was achieved with ten bilayers only, while 99% was achieved with ten bilayers only, while 99% was achieved with ten bilayers only, while however, it is very challenging to develop suitable inks and establish a reliable multilayer fabrication process. In terms of the formulation of the inks, they should contain constituents that give a relatively high RI contrast while meeting the orthogonal solubility condition. In addition, every layer needs to be as uniform as possible to ensure a uniform multilayer stack. Moreover, it is crucial to have precise control and a high reproducibility on the film thickness so as to achieve superior optical performance, e.g., high reflectance of the Bragg mirrors.

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2. Results and Discussion

A typical Bragg mirror stack architecture can be represented as A[(HL)\textsuperscript{N}]S, where A, H, L, and S denotes air, high RI layer, low RI layer, and substrate, respectively, and N is the number of bilayers. Under normal incidence, the position of the fundamental PBG (\(\lambda_0\)) in such a design is given by the following relation:\textsuperscript{[23]}

\[
\lambda_0 = 2(n_1 d_1 + n_L d_L)
\] (1)

In the case of a quarterwave stack, i.e., \(n_1 d_1 = n_L d_L = \lambda_0/4\), the reflectance (\(R_0\)) at \(\lambda_0\) can be calculated as:\textsuperscript{[24]}

\[
R_0 = \frac{\left(\frac{n_{11}/n_{12}}{n_{11}/n_{12}}\right)^{2N} - n_L}{\left(\frac{n_{11}/n_{12}}{n_{11}/n_{12}}\right)^{2N} + n_L}
\] (2)

where \(n_{11}\) and \(n_{12}\) are the RI of H and L layers, \(d_{11}\) and \(d_L\) are the thickness of H and L layers, respectively. \(n_L\) is the RI of the substrate. The stopband width (\(W\)) for the quarterwave case can be calculated using the following equation:\textsuperscript{[25]}

\[
W = \frac{4}{\pi} \lambda_0 \arcsin\left(\frac{n_{11} - n_L}{n_{11} + n_L}\right)
\] (3)

The expressions above show that the central wavelength depends only on the optical thickness of both H and L layers. Hence, it can be tuned by changing the material composition and even simpler by the layer thickness. The maximum reflectance is only influenced by the number of layers with the same constituent materials, and a higher reflectance can be obtained by stacking more layers. In principle, the layer thickness can be controlled in an inkjet printing process by modifying the printing parameters. However, key challenges are the nanometric control of the thickness, the uniformity of the printed layers, and the choice of solvents such that intermixing of L and H layers is avoided.

Here, TiO\textsubscript{2} and PMMA were chosen as the constituent materials due to their widespread usage in optical applications and their high contrast in RI. The fabrication process of our Bragg mirrors was completed by alternately printing the PMMA and TiO\textsubscript{2} layers on top of each other, as illustrated in Figure 1a. In order to be applied as suitable inks for the inkjet printing process, TiO\textsubscript{2} nanoparticle dispersion with a UV curable organic matrix as host, as well as a PMMA ink were developed. The rheological properties of the PMMA and TiO\textsubscript{2} composite inks are shown in Table 1. The shear viscosity of the PMMA and TiO\textsubscript{2} composite ink was measured at 28 and 27 °C, respectively, as shown in Figure S1 (Supporting Information). To determine the jettability of the inks, which is related to their rheological parameters such as viscosity, surface tension, and inertial force, a set of dimensionless physical constants, the Reynolds (\(Re\)), Weber (\(We\)), and inverse Ohnesorge (\(Z\)) numbers, are used:\textsuperscript{[26]}

\[
Re = \frac{\rho v l}{\eta}
\] (4)

\[
We = \frac{\rho v^2 l}{\gamma}
\] (5)

\[
Z = \frac{1}{\text{Oh}} = \frac{Re}{We} = \frac{\sqrt{\eta l}}{\gamma}
\] (6)

where \(\gamma\), \(\eta\), and \(\rho\) are the surface tension in (N m\textsuperscript{-1}), dynamic viscosity (in Pa s), and density (in kg m\textsuperscript{-3}) of the ink, respectively, \(v\) is the velocity (in m s\textsuperscript{-1}), \(l\) is the characteristic length (in m, which is the nozzle diameter here), and \(Oh\) is the Ohnesorge number. The droplet velocity of the PMMA ink was 4.0 m s\textsuperscript{-1}, and of the TiO\textsubscript{2} composite ink was 3.8 m s\textsuperscript{-1}. The nozzle orifice...
size is 21.5 µm. It is proposed that the $Z$ number has to be in the range of $1 < Z < 10$ for stable drop generation.\cite{26a} When $Z$ is too low, the ink is too viscous to be ejected by the nozzles, whereas a high $Z$ value indicates a large number of satellite droplets with the primary drop. In addition, a drop needs sufficient energy to overcome the liquid/air surface tension at the nozzle, and this gives a minimum $We$ value of 4.\cite{26a} Moreover, the threshold for the onset of droplet splashing on the substrate needs to be considered, and a well-established experimental threshold was proposed to be $We^{1/3}Re^{1/4} > 50$ for a flat and smooth surface.\cite{27} Accordingly, a map can be constructed in the $Re$–$We$ parameter space with the variables and limiting factors listed above, as shown in Figure 1b. The locations of both PMMA and TiO$_2$ composite inks are within the white area of the printable region, indicating that both inks are printable by inkjet printing.

At room temperature, PMMA showed a very low solubility in the solvent used for TiO$_2$, as shown in Figure S2 (Supporting Information). Therefore, it ensures that the previously printed PMMA layer is dissolved to a minor extent during the next TiO$_2$ printing step and vice versa. The RIs of both materials were measured by spectroscopic ellipsometry and are shown in Figure 1c. The RI of a printed TiO$_2$ layer is 2.08 at 380 nm and 1.87 at 780 nm, while at these wavelengths, the RI of a PMMA layer is 1.49 and 1.48, respectively. This gives a high RI contrast ranging from 0.59 to 0.39 in the visible light range and renders these materials suitable to be used as high index and low index layers in the Bragg mirror. The measured extinction coefficients of the two constituent materials are shown in Figure 1d. The measured extinction coefficients are zero in the wavelength range of 370 nm to 1200 nm, indicating very low absorption and negligible scattering in the corresponding wavelength range.\cite{28}

The details of the ellipsometry measurement can be found in Figures S3 and S4 (Supporting Information).

In order to obtain different spectra with $\lambda_0$ over the whole visible light range, the thickness of each layer was adjusted by changing the printing resolution, i.e., the deposition volume per unit area. During printing, at least ten nozzles were used for jetting, and the quality factor was chosen as two, which means two nozzles are used to print one vertical line. An example schematic of the nozzle arrangement can be found in Figure S5 (Supporting Information). By these means, the variation in individual droplet volume was minimized in each printing round, and a high-precision control in film thickness was achieved. The relation between the layer thickness of both materials and the number of printed droplets is shown in Figure S6 (Supporting Information). With the number of printed droplets increasing, the thickness of the thin film increases, and the reflectance spectrum shifts toward the long-wavelength range. The normalized reflectance spectra of TiO$_2$ composite thin films with different thicknesses can be seen in Figure 2a, where on a $2 \times 2$ cm$^2$ area, the number of printed droplets increases from $N_0 = 309$ 135 to $N_4 = 322$ 503, with a constant interval $\Delta N = 3342$. The measured reflectance spectra

Table 1. Rheological properties of the PMMA and TiO$_2$ composite inks.

| Ink                  | Density [g cm$^{-3}$] | Surface tension [mN m$^{-1}$] | Viscosity [mPa s] |
|---------------------|-----------------------|-------------------------------|-------------------|
| PMMA ink            | 1.04                  | 38.45                         | 5.05              |
| TiO$_2$ composite ink | 0.94                  | 29.89                         | 2.62              |
were fitted to the simulation results to extract the thickness information of the single layer, and the fitting results are shown in Figure S7 (Supporting Information). With the number of printed droplets increasing from \( N_0 \) to \( N_4 \), the peak wavelength of the reflectance spectrum increases from \( 472.8 \) to \( 496.8 \) nm, as can be seen in Figure 2b. This indicates an increase in the film thickness from \( 69.2 \) to \( 72.5 \) nm and a thickness resolution of \( 0.825 \pm 0.150 \) nm. The thickness resolution can be further reduced to lower magnitudes by decreasing \( \Delta N \) and diluting the ink. After printing, the PMMA layer went through a drying process in reduced pressure condition, which can also be replaced by an integrated photo-crosslinking in future work when more advanced materials are used. And the TiO\(_2\) layer was hardened by UV curing. The UV-curable polymer matrix (PM) also helped to enhance the adhesion of the TiO\(_2\) layer without the need for a high sintering temperature. This UV curing step is an essential process, as when the UV radiation dose was not sufficient during exposure, an intermixing of the subsequent PMMA layer with the TiO\(_2\) layer was observed.

In order to realize a highly reflective mirror, a larger number of bilayers had to be deposited on top of each other. For achieving a reflectance of more than 98\%, ten bilayers are needed by calculations according to Equation (2). Figure 2c shows the cross-sectional scanning electron microscopy (SEM) image of our printed Bragg mirror with ten bilayers. The bottom PMMA layer was intentionally designed to have a relatively larger thickness to ensure a closed PMMA film on the substrate. Meanwhile, this has little impact on the final optical property of the Bragg mirrors due to its similar RI as the glass substrate. Especially for a high sintering temperature. The bandwidth of the PBG becomes larger with increasing \( \rho_0 \), as predicted by Equation (3). In addition, the Bragg mirrors show a significant drop in transmittance in both constituent materials and the glass substrate. The Bragg mirror whose \( \rho_0 \) is at \( 416 \) nm, a fall in the left edge of the stopband was obvious. The CIE color coordinates

Figure 2. a) Normalized reflectance spectra of single TiO\(_2\) composite thin films with different thicknesses on glass substrates. The reflectance spectrum shifts toward the longer wavelength range with the number of printed droplets (\( N_0, N_1, ..., N_4 \)). b) The peak wavelength of the reflectance spectra (gray line) and the calculated layer thickness (orange line) with respect to the number of droplets printed in the same area. The number of printed droplets has an equal step of \( \Delta N \approx 334.2 \). c) Cross-sectional SEM image of a printed ten-bilayer Bragg mirror. The \( H \) and \( L \) layers were partially colored for clarity. Scale bar: 500 \( \mu \)m. d) SEM image of the top layer of the ten-bilayer Bragg mirror. Scale bar: 1 \( \mu \)m.
of the four fabricated Bragg mirrors in the visible light range can be found in Figure S12 (Supporting Information). Figure 3c exhibits reflectance spectra with $\lambda_0$ at 561 nm, showing the effect of the number of bilayers in the stack. As can be seen, when the Bragg mirror consists of one bilayer only, the reflectance peak is broad and is $\approx 20\%$. Then it increases to $\approx 60\%$ with three bilayers, and the value exceeds $80\%$ with five bilayers. We further pushed this limit to $96\%$ with eight bilayers and $99\%$ with ten printed bilayers. In addition, the central wavelengths of these different stacks show a minor difference within a few nanometers only, indicating that the layer thickness can be well controlled by carefully choosing working nozzles and the corresponding inkjet printing parameters. Using the measured thickness and the RI values in Figure 1c, simulations on the reflectance spectra were also carried out. The layer thicknesses of respective PMMA and TiO$_2$ layers were assumed constant in the simulation except for the bottom PMMA layer. The comparison between the simulated and measured spectra can be found in Figure 3d, proving an excellent agreement on the central wavelength, reflectance peak, and bandwidth. The slight increase in the side interference peaks might result from the detuning of the Bragg mirror and the random thickness variation.[20a,29] Furthermore, the angle dependence of the fabricated Bragg mirror at 561 nm can be found in Figure S13 (Supporting Information), where a larger angle of incidence leads to a blue shift.

Our inkjet-printed Bragg mirrors can be used not only as high reflectivity mirrors but also as dichroic beamsplitters. Figure 4a shows the dichroic wavelength filtering property of the printed Bragg mirror. When holding the sample upstanding, a white light beam was incident on the stack from the right side. The fraction of the blue light was mainly reflected back to the same side of the incident light, while the rest components in the spectrum passed through and formed an orange shadow of the Bragg mirror on the left side. An idea of the vast possibilities of our printed Bragg mirrors for large-area applications is given in Figure 4b–d. Figure 4b is a photograph of the fabricated Bragg mirrors printed in 2 $\times$ 2 cm$^2$ square forms on 2.5 $\times$ 2.5 cm$^2$ glass substrates. The 5 $\times$ 5 array was arranged in a way that the number of bilayers increased from one to five in the vertical direction. In the lateral direction, the thickness of the layers increases, and thus $\lambda_0$ shifts from the purple to red region from left to right. From this image, we can see that the printed Bragg mirrors cannot only be used as a high reflection or dichroic mirror, but also in terms of the visual and aesthetic aspects they can present a homogeneous color impression, and the brightness of the color is clearly increasing with an increasing number of layers.

After the inks have been developed, the inkjet printing process can be transferred to a large variety of substrates. Only the printing parameters for the first PMMA layer need to be adjusted according to the wettability of the substrate. Therefore, Bragg mirrors can be printed not only on small and rigid substrates but also in specific patterns on large and flexible substrates. Bragg mirrors were printed on 12 $\times$ 12 cm$^2$ polyester (PET) foils, in the letter pattern of “Printed 1DPC” and two large background rectangles with different $\lambda_0$. The printed Bragg mirror is shown in its relaxed and bent status in Figure 4c,d, respectively. The deformability of our printed Bragg mirrors, therefore, allows for their applications on curved surfaces with a high degree of freedom. The excellent color homogeneity over a large area and the high reflectance give the inkjet-printed Bragg mirrors a massive potential for a large variety of applications. Furthermore, the inkjet printing process of the TiO$_2$ nanoparticle composite ink is compatible with other nanofabrication processes, such as nanoimprinting. It could therefore
be used for the deposition of dielectrics in various applications, e.g., nanocomposite metasurfaces.[30]

3. Conclusion

We have demonstrated the first fully digitally manufactured Bragg mirrors by multilayer inkjet printing. The central wavelength was tuned from the purple to infrared spectral range, i.e., from 416 to 808 nm. The central wavelength was tuned by tuning the layer thickness via changing printing resolutions, and the maximum reflectance was adjusted by controlling the number of bilayers. The reflectance peak reached up to ≈99% when ten bilayers were printed. Without the need for a high annealing temperature, Bragg mirrors were successfully printed on glass substrates as well as on flexible PET foils in designed patterns. The printed Bragg mirrors showed a good color homogeneity and an overall high quality in optical property, making inkjet printing a highly competitive candidate for the large-scale fabrication of high-quality Bragg mirrors. Inkjet printing offers a fast, simple, material-saving, and low-cost fabrication route. Inkjet-printed Bragg mirrors, large or small, patterned or unpatterned, can be used in numerous fields as functional and decorative optical components. We foresee vast applications, ranging from additive manufacturing of integrated photonic systems (e.g., in sensing systems) to large-area applications such as aesthetically appealing photovoltaics.

4. Experimental Section

Substrate Preparation: The 2.5 × 2.5 cm² glass substrates (soda lime glass) and 12 × 12 cm² PET foils (Puetz Folien) were cleaned in an ultrasonic bath in deionized water, acetone, and isopropanol for 10 min each. Then the substrates were treated by oxygen plasma in a plasma chamber (PlasmaFlecto 30, Plasma technology) with a power of 100 W for 10 min.

Ink Preparation and Inkjet Printing: PMMA with a molecular weight of 65 000 Da (PSS-polymer) was dissolved in 1,3-dimethoxybenzene (≥98%, Sigma–Aldrich) to achieve a concentration of 40 mg mL⁻¹, and 10% hexylbenzene (97%, Sigma–Aldrich) was added to mitigate the coffee ring effect. TiO₂ ink was prepared by diluting the TiO₂ nanoparticle dispersion (RF-10-UV, Avantama) with ethylene glycol monopropyl ether (99.4%, Sigma–Aldrich) to reach a final concentration of 3.8 wt.%. The concentrations of the inks were determined to reach the target film thickness range with suitable printing parameters. Before printing, both inks were placed in an ultrasonic bath for 5 min and then filtered using PTFE filters with a pore size of 0.2 μm. The inkjet printer (PixDro LP50) was equipped with 10 pL cartridges (Fujifilm Dimatix). During printing, at least ten nozzles were used for jetting. The substrate temperature was set at 24 °C. The printhead temperature was set at 27 and 28 °C for TiO₂ and PMMA ink, respectively. The waveforms were custom-made for up to 2.5 kHz jetting frequency. Both waveforms were single peak waveforms with a maximum voltage of 22 V, because the inks have been
developed to be suitable for inkjet printing. First, a PMMA layer was printed directly on the substrate, and then the layer was dried at 10 mbar for 2 min and then placed on a hotplate at 50 °C for 5 min. TiO2 layer was subsequently printed on top of the PMMA layer, and this layer was first dried at ambient temperature for 2 min, and then prebaked on a hotplate at 100 °C for 5 min, UV-cured for 10 min with a UV-LED light source (GC 77, Hamamatsu), and subsequently post baked at 100 °C for 10 min. The following PMMA and TiO2 layers were alternately printed and processed in the same way. The thickness of each layer was controlled by changing the printing resolution, which determines how much volume is finally deposited on a unit area. The printing resolutions were in the range of 550–900 dots per inch (dpi) for TiO2 ink and 500–700 dpi for PMMA ink. The whole fabrication process was completed in a cleanroom with the environment temperature at 21–22 °C and humidity at 40–50%.

Characterization and Simulation: The refractive indices and extinction coefficients of the materials have been measured by ellipsometry (VASE ellipsometer, J.A. Woollam). The cross-sectional SEM images and the SEM images of the top layer of the Bragg mirrors were obtained by SUPRA 55 (Carl Zeiss) at 3 kV. The sample was prepared by focused ion beam (FIB) milling (Zeiss Crossbeam 1540 EsB) for the cross-sectional SEM measurements. The cross-sectional SEM image was partially colored using the outline trace function of CorelDraw software in a layer-by-layer manner. The original SEM image can be seen in Figure S14 (Supporting Information). The reflectance (under an angle of 8°) and transmittance spectra were measured with a spectrophotometer (Lambda 1050+ UV–vis–NIR, PerkinElmer) with an embedded integrating sphere. The reflectance spectra in the angle dependence measurement were measured with the same spectrophotometer without an integrating sphere. The refractive indices used for simulation were obtained by the ellipsometer measurement, and the thickness values were measured using a stylus profilometer (DektakXT, Bruker). The optical simulation of the Bragg mirrors was accomplished using the simulation program “Optical” based on the transfer matrix method.[11] The surface morphology was measured by AFM (NanoWizard, Bruker Nano). The surface RMS roughness was analyzed with Gwyddion. The surface tension of the ink was measured by a contact angle measuring system (OCA 50, DataPhysics Instruments) under ambient conditions. The viscosity of the ink was measured using a viscometer (m-VROC, RheoSense).

Statistical Analysis: The effective refractive indices of the films of the constituent materials were determined by fitting the ellipsometric parameters to a Tauc-Lorentz oscillator and a roughness layer. The mean square error (MSE) is 1.5 (three angles fitted simultaneously). The ellipsometric statistical analysis was carried out using the program WVASE (J.A. Woollam). All normalized reflectance spectra were normalized with respect to the maximum value. Data were presented as mean ± standard deviation (SD).

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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