Hydrophobic and optical properties of silica antireflective coating prepared via sol-gel method

Qianying Zhang, Hui Liu , Siyuan Zhao and Wanmeng Dong

School of Materials Science and Engineering, Shaanxi Key Laboratory of Green Preparation and Functionalization for Inorganic Materials, Shaanxi University of Science and Technology, Xi’an, 710021, People’s Republic of China

E-mail: liuhui@sust.edu.cn

Keywords: bifunctional coating, antireflection, self-cleaning, silica, dimethyl silicone

Abstract

A simple and low-cost process for the preparing of multifunctional coating is essential in practical applications. Here, alkali-catalyzed SiO2 antireflection coating was first prepared by using the sol-gel method, the tetraethyl orthosilicate (TEOS) and ammonia (NH₃·H₂O) were used as a raw material and catalyst, respectively. Then, a layer of dimethyl silicone was sprayed on the surface of the as-prepared primary coating to obtain a bifunctional coating of antireflection and hydrophobicity simultaneously. The experimental results shown that the peak transmittance of the as-prepared bifunctional coating was 98.11% in the wavelength range of 300–1500 nm, while the water contact angle of the prepared coating was 140°, which was caused by the introduction of hydrophobic methyl groups of dimethyl silicone. Besides, the tape adhesion test and pencil scratch test results shown that the coatings have excellent mechanical robustness. Therefore, this work provides a practical strategy for the preparation of bifunctional coatings with antireflective and hydrophobic properties.

1. Introduction

Reflection often exists at the interface of the transparent substrate, for example, the reflection of air-glass interface reduces the efficiency of solar modules [1–3]. In order to eliminate the unnecessary reflection, antireflection (AR) coatings have been widely researched due to their excellent antireflection properties [4]. In order to obtain an excellent AR property, the refractive index of the antireflection coating must be between the refractive index of the substrate and that of the air [5]. According to the interference principle and Fresnel formula, the refractive index of optical glass is 1.5 and the air is 1.0, which implies that the theoretical refractive index of the ideal single layer antireflection coatings is between 1.20 and 1.25 [6].

According to this physical principle, Magnesium fluoride (1.38) is the material with the lowest refractive index in nature [7], and still cannot achieve transmittance of 100% [8]. In addition, the practical application of fluorine materials is limited by the higher price and potential environmental hazards, which cannot meet the needs of large-scale production [9]. Fortunately, alternative scheme, such as through adjusting the pore structure of the coating materials to satisfy the above physical principle, [10, 11] it can be used to produce adjustable AR coatings on required optical elements to meet practical demand. Khan et al used glancing angle deposition (GLAD) to prepare a double-layer AR coating with periodically arranged SiO₂ nanostructures on a dense SiO₂ bottom layer [8]. By changing the deposition parameters to control the porosity of each layer, a double-layer coating with a refractive index between 1.08 and 1.46 can be easily obtained to achieve antireflection. Tao et al controlled the porosity of the coating by adjusting the molar ratio of tetraethyl orthosilicate (TEOS) and hexamethyldisilazane (HMDS) to obtain a double-layer antireflective coating with the refractive index of the bottom and top layers of 1.27 and 1.16, respectively [11]. And the transmittance of the as-prepared antireflective coating at 351 nm, 527 nm and 1053 nm can reach to 99.6%, 98.0% and 99.3%, respectively. These indicate that the required refractive index of the coating can be prepared by controlling the porosity to achieve antireflection performance and reduce the undesirable optical loss caused by the transparent substrate interface.
However, simple antireflection coating is often difficult to meet the needs of practical applications on susceptible polluted environment, especially for outdoor solar panels, architectural glass, automotive glass, etc [12, 13]. To maintain the transmittance of the transparent substrate requires regular manual cleaning, which requires considerable resources. Moreover, manual cleaning will cause great damage to the surface antireflection coating of the transparent substrate and accelerate the attenuation of transmittance [14, 15]. Therefore, in recent years, efforts have been made to impart self-cleaning functions to antireflective coatings. In nature, lotus leaf is typical hydrophobic surface with self-cleaning function, pollutants or dust on lotus leaf can be easily cleaned by rolling water drops. Related studies shown that the main factors affecting the hydrophobicity of lotus leaves are rough micro/nanostructure and waxy substances with low surface energy [12, 16–18]. According to this basic principle, researchers have successfully prepared various hydrophobic coatings. Yokoi et al deposited the 1H, 1H, 2H, 2H-perfluorodecyltrichlorosilane (PFDTS) on the alkaline treated polyester mesh by chemical vapor deposition (CVD) method, and sprayed the SiO2 nanoparticles modified by 1H, 1H, 2H, 2H-perfluoroctyltriethoxysilane (PFOTS) to obtain a transparent super-hydrophobic surface with a transmittance of 80% and a water contact angle of 150° [19]. Wu et al obtained a structure with hexagonal triangular protrusions and hexagonal rectangular micropillars by unidirectional rubbing and heating-assisted assembly technology, and then it was modified with PFOTS by CVD method to obtain a transparent superamphiphobic coatings [20]. Therefore, it proves that the hydrophobic coating can be obtained by using low surface energy material to modify rough surface. However, the large-scale industrial applications of hydrophobic coatings are often restricted due to the expensive raw materials, complex processes, or environmental pollution [4]. Therefore, a relatively simple and low-cost method was used in this work for preparing a bifunctional coating with antireflective and hydrophobic properties.

Herein, we used the sol-gel method to prepare a bifunctional coating with silica nanoparticles and dimethyl silicone as the bottom and top layer, respectively. The accumulation of silica nanoparticles on the bottom layer makes the coating have a certain porosity to obtain the antireflective property. At the same time, the introduction of hydrophobic methyl functional groups in the dimethyl silicone on the top layer makes the coating have good hydrophobicity and mechanical robustness. Therefore, this work provides a low-cost and simple method to obtain a bifunctional coating with antireflection and hydrophobic properties to meet the practical applications.

2. Experimental section

2.1. Reagents and chemicals
Tetraethyl orthosilicate (TEOS), anhydrous ethanol (EtOH, 99.7%), ammonia solution (NH3·H2O, 25%–28%) and dimethyl silicone were purchased from Tianjin Kermel Chemical Reagent Co., Ltd All of the reagents were used as received without further purification. The deionized water (H2O) was obtained by Millpore Water System (18.2 MΩ·cm).

2.2. Preparation of silica sol
Silica sol was prepared by a simple and low-cost sol-gel method. The typical fabrication process was carried out as follows: ethyl orthosilicate (7.5 ml) was dispersed in ethanol (36 ml) under magnetic stirring and recorded as solution A. The mixture of ammonia (1.1 ml), deionized water (1.5 ml) and ethanol (36 ml) was magnetically stirred at room temperature for 30 min as solution B. Then, solution B was dropped to solution A with magnetic stirring for 3h at room temperature. The resultant mixture was sustained and aged silica sol at room temperature and obtained silica sol.

2.3. Preparation of bifunctional coating
The glass was used as a substrate to prepared bifunctional coating, which was washed with deionized water and ethanol. Then, as-prepared silica sol was deposited on the glass substrate by a dip-coating technique to obtain the antireflection coating. Subsequently, amount of dimethyl silicone was sprayed on the as-prepared antireflection coating and heat treated at 450 °C for 1 h. The bifunctional coating was obtained by cooling to room temperature.

2.4. Characterization
Nano Particle size analyzer (NAMO-ZS, Malvern, UK) was analyzed the particle size and distribution of silica sol. X-ray diffraction (XRD) measurements were implemented on a D/max2200PC diffractometer using Cu Kα radiation (λ = 1.54178 Å). The plane and cross-sectional microstructure of the bifunctional coating were performed on a scanning electron microscope (SEM, S4800, Hitachi) under operating voltage of 5kV. Atomic force microscopy (AFM, SPI3800N/SPA400) was used to analyze the surface morphology of the bifunctional coating. Fourier transform infrared spectroscopy (FTIR, Vector–22, Bruker) was detected the chemical nature of the surface. Spectrum recording within the 400–4000 cm−1 frequency range. The transmission spectra of the
bifunctional coating was measured via a Varian Cary 5000 UV/vis-NIR spectrophotometer in the wavelength of 300–1500 nm. The variable angle spectroscopic ellipsometer (J. A. Woollam, USA) measured the thickness and refractive index of the coating. Water contact angle of the bifunctional coatings were measured at video optical contact angle (JC2000X, Germany) and 5 μl of the water droplets were dropped onto the coating surfaces.

3. Results and discussion

3.1. Structural and morphology analysis

For a certain coating material, the structure of the sol particles has a great influence on the refractive index of the coating. Particle size and its distribution are shown in figure 1(a). The particle size of the sol prepared in this experiment is about 30 nm, and the intensity distribution exhibited an obvious unimodal pattern. The PdI (polydispersity index) value is 0.3936, which indicates that the prepared monodisperse sol particles are uniformly distributed. The porous structure of the SiO₂ sol particles exercise a great impact on the antireflection performance of the coating. When the SiO₂ particles are too small, they are packed tightly and the pores formed are smaller. The large-diameter SiO₂ particles can construct more pore structures, which reduces refractive index of coating. However, the large SiO₂ particles are not conducive to preservation and prepared coating.

Figure 1. (a) Particle size intensity of silica sols. X-ray diffraction patterns of (b) antireflective coating and (c) bifunctional coating.

Figure 2. SEM and AFM images of (a)–(b) antireflective coating and (c)–(d) bifunctional coating.
Therefore, the sol prepared in this experiment has suitable particle size and uniform particle distribution, which is suitable for preparing antireflection coatings.

The XRD patterns reflect the composition and crystal structure of the as-prepared coating materials. Figures 1(b)–(c) shows the XRD patterns of antireflective coating and bifunctional coating. It can be seen that the two coatings show a characteristic peak around 20° to 30°, which confirmed the presence of amorphous silica [21]. And the amorphous silica was obtained by hydrolysis and polycondensation of TEOS under alkali-catalyzed. The XRD patterns of bifunctional coating is consist with antireflective coating, which indicating the composition did not changed with the addition of dimethyl silicone. In addition, the XRD patterns of the two coatings did not have other characteristic peaks, indicating the as-prepared coatings are purity. Therefore, above results indicating that antireflective coating and bifunctional coating have the property of amorphous structures. Amorphous silica is often used to prepare antireflective coatings because of its good mechanical properties, high firmness, excellent chemical stability and the accumulation of pore structure can reduce the refractive index of the material [22, 23]. In addition, the surface of amorphous silica contains more active hydroxyl, which can be used as site for functional molecules during surface modification [24–26].

The morphology and microstructure of the antireflective coating and bifunctional coating are show in figure 2. It can be seen from figures 2(a), (c), antireflection coating and bifunctional coating have a porous structure and accumulate with silica nanoparticles. The formation of silica nanoparticles is due to the TEOS occurred hydrolysis and condensation reaction under alkali-catalyzed and further formed micronuclei. The reactants continue to deposit and grow on the surface of the micronuclei and gradually formed nanoparticles [6, 27, 28]. It can be seen that figure 2(c) is more homogeneous than that of figure 2(a), this is because the dimethyl silicone has a graft reaction with the hydroxyl on the surface of the silica to fill the pores, so the surface of the bifunctional coating is smoother and the transmittance is slightly reduced, but the introduced dimethyl silicone is beneficial to the hydrophobic properties. Moreover, the surface roughness of as-prepared coatings is analyzed by AFM. In figures 2(b), (d), the root-mean-square (RMS) roughness of the antireflection coating and the bifunctional coating is 15.6 nm and 3.35 nm in an area of 5 × 5 μm, respectively. The decreased roughness of the bifunctional coating is related to the existence of dimethyl silicone on the surface. Dimethyl silicone is a kind of polyorganosiloxane with the chain structure of different polymerization degrees. It can deposit on the surface of the antireflection coating to fill the pores and reduce the surface roughness of the bifunctional coating, which is consist with the above argument. Therefore, combining the porous structure and the grafted dimethyl silicone can make the bifunctional coating have excellent antireflective and hydrophobic properties.

### 3.2. Optical properties of bifunctional coating

The transmittance can be used to evaluate the optical properties of the coating. The transmittance of different coatings were analyzed by UV/vis-NIR spectrophotometer. Figure 3(a) shows the transmittance spectra of the bare glass, antireflection coating and bifunctional coating in the range of 300–1500 nm. The transmittance of antireflection coating and bifunctional coating is greatly improved compared with that of bare glass, especially in the visible light range. This is because the porous structure of the stacked amorphous silica nanoparticles causes diffuse reflection of incident light, suppresses the reflection of incident light and improves transmittance [29]. Moreover, the transmittance of the bifunctional coating (98.11%) is slightly lower than that of the antireflection coating (98.68%). This is because the dimethyl silicone is deposited on the surface of the antireflection coating to make the surface of the bifunctional coating more homogeneous, which causes the dimethyl silicone to fill part of the pore structure of the coating surface, so that incident light cannot pass through the pores structure.

![Figure 3.](image)
Therefore, the incident light is reflected on the surface of the bifunction coating, which manifests as the transmittance of the dual-functional coating decreases. Additionally, the thickness of the antireflective coating (175 nm) and the bifunctional coating (214.3 nm) was measured with a variable angle spectroscopic ellipsometer. According to the formula of $2dn = \frac{1}{2} \lambda$, the thickness of the coating increased with the dimethyl silicone, so the peak transmittance of the bifunctional coating shifts to the long wave.

To obtain a coating with excellent transmittance, the refractive index of the coating must match the refractive index of the substrate as much as possible. The refractive indexes (at the wavelength of 632.8 nm) for the as-prepared coatings were analyzed by the Cauchy model of optical constants. Figure 3(b) shows the refractive index of the bare glass (1.501), antireflection coating (1.224) and bifunctional coating (1.229). It is worth noting that the dimethyl silicone layer slightly increases the refractive index of the antireflection coating, but it is still between 1.20 and 1.25. This indicates that compared to the SiO$_2$ antireflection coating, the refractive index of the dimethyl silicone layer by heating treatment is similar to that of the SiO$_2$ antireflection layer.

![Figure 4. SEM images of coating with different dipping times: (a) 1 time (b) 2 times (c) 3 times and (d) 4 times (The insets in the top right corner shown the corresponding cross-sectional view.).](image1)

![Figure 5. (a) Transmission spectra and (b) refractive index (at the wavelength of 632.8 nm) of bifunctional coating with different dipping times.](image2)
Therefore, the refractive index of the bifunctional coating does not change significantly and still has excellent optical properties.

The thickness of the coating has a great influence on the optical properties of the as-prepared bifunctional coating, thus the effect of coating times on optical properties of the coating was studied. Figure 4 exhibited the SEM images of the as-prepared coating with different dipping times. From the plane view, the bifunctional coatings have a uniform and compact surface morphology, and more and more cracks appear on the coating with the increase of dipping times. This is mainly because the evaporated moisture and solvent cause collapse of the porous structure during the heat-treated process. From the corresponding cross-sectional view inserted in the upper right corner, the thickness of the coating increases continuously (119.7 nm, 214.3 nm, 262.2 nm and 285.8 nm) and the degree of increases gradually decreases with the increase of dipping times. This is because silica nanoparticles accumulate on the surface of the glass substrate to form a hydrophilic silica coating under the action of adhesion, and the silica nanoparticles continue to deposit on the surface of the as-prepared silica coating when dip again in silica sol. When the attractive force between the silica nanoparticles is greater than the attraction force with the glass substrate, the silica nanoparticles are difficult to deposit continuously, so the thickness of the coatings does not change significantly as the increase of dipping times. Moreover, the dimethyl silicone molecules have graft reaction with the hydroxyl group and fill part of the porous structure, so there is no obvious delamination in the bifunctional coating. In this way, silicon nanoparticles can be stably adhered on the glass substrate to form a bifunctional coating with strong mechanical robustness [26, 30].

Figure 5(a) shows the transmittance spectra of bifunctional coating with different dipping times in the range of 300–1500 nm. Compared with that of the bare glass, the transmittance of the bifunctional coating increased obviously. This is because the loose porous structure of the bifunctional coating reduces the reflection of incident light, which leads to an increase of the transmittance [29]. Moreover, the transmittance of the coating shows fluctuations, and the number of wave crests and wave troughs increased with the increase of dipping times. This is because the thickness of the coating increases with the dipping times, and the optical path difference becomes larger. The appearance of constructive interference and destructive interference lead to the formation of wave crest and wave trough at the transmittance spectrum [31, 32]. Generally, the wave trough shows a lower transmittance, so it is necessary to choose dipping times with as few wave troughs as possible. At the same time, visible light and infrared light account for about 50% and 43% of the total solar radiation energy, respectively. So, the visible light and the near-infrared region also need more transmittance as much as possible. The results indicated that the bifunctional coating has the best transmittance when the dipping time is 2. This further confirms that preparing bifunctional coatings of appropriate thickness can achieve high transmission and thus can improve the utilization of solar radiation energy.

Figure 5(b) shows the refractive index of bifunctional coating with different dipping times. With the increase of dipping times, the refractive index of the bifunctional coating is 1.256, 1.229, 1.215 and 1.222, respectively. Compared with blank glass, the refractive index of the bifunctional coatings are between the theoretical
refractive index of the ideal antireflection coating, so the prepared bifunctional coatings have excellent antireflection properties. The difference in refractive index of the bifunctional coating is mainly caused by the random accumulation of silica nanoparticles with the dip-coating technique. From the analysis of the SEM images, it can be seen that as the increase of dipping times, the silica nanoparticles become more and more difficult to be deposited continuously, which may result in the scattering of incident light, and cause transmittance fluctuations and refractive index decrease. Combined with the coating technique and the transmission spectra, when the dipping time is 2, the obtained bifunctional coating has excellent optical properties and can be used on a transparent substrate with a refractive index about 1.5.

### 3.3. Self-cleaning properties of bifunctional coating

The self-cleaning properties of coatings can be obtained by controlling the wettability of the coating surface, and the wettability is usually characterized by the water contact angle. The water contact angle is less than 90° for hydrophilic coating and greater than 90° for hydrophobic coating. As shown in figure 6, the water contact angles of bare glass, antireflection coating, bifunctional coating and dimethyl silicone are 23°, 7°, 140°, and 53°, respectively. Antireflective coating has the smallest water contact angle and exhibits hydrophilicity, this is because the silica surface has amount of hydrophilic hydroxyl functional groups. According to the Wenzel model, when the coating’s surface has the hydrophilic group, the water contact angle decreases with increasing of the surface roughness. It is worth noting that the water contact angle of the bifunctional coating becomes larger and exhibits hydrophobic with the addition of dimethyl silicone, this because the methyl groups on the dimethyl silicone is hydrophobic. According to the ‘lotus effect’, the coating shows hydrophobicity when it has a rough structure and low surface energy. Therefore, the bifunctional coating with antireflective and hydrophobic properties is obtained by combining the rough structure of the silica nanoparticles and the low surface energy of dimethyl silicone.

The self-cleaning properties of bifunction coating is due to the effect of dimethyl silicone on its surface, and the thermal decomposition process of dimethyl silicone is greatly affected by temperature, so it can be said that
the water contact angle of the coating is also closely related to temperature [33, 34]. Therefore, the influence of different temperatures on the wettability of bifunctional coatings was explored. Figure 7(a) shows the water contact angle of the bifunctional coating at different temperatures. The result shows that the water contact angle of the bifunctional coating can reach to 140° at 450 °C, and the water contact angle exhibits a decreasing trend with the temperature increases. When the temperature rises to 550 °C, the water contact angle of the coating drops sharply to 5°, indicating that the coating is super-hydrophilic. This phenomenon can be explained by FTIR analysis.

The chemical nature of bifunctional coating at different temperatures were studied by FTIR spectra. Figure 7(b) shows the FTIR spectra of bifunctional coating at 400 °C to 550 °C. The absorption peaks at 870 cm⁻¹, 2903 cm⁻¹ and 2960 cm⁻¹ are the stretching vibration of Si–C and C–H bonds, respectively [35, 36], which is the reason for the hydrophobicity of the coating. When the temperature at 550 °C, the absorption peaks of Si–C and C–H bonds disappeared. The results show that the alkyl groups on the coatings are completely oxidized at 550 °C, which causes the complete disappearance of low surface energy and the coating becomes super-hydrophilic. The result suggesting that the hydrophobicity of the bifunctional coating is due to the methyl groups on the dimethyl silicone, so the functional group of the coating can be controlled by temperature to obtain different hydrophilic or hydrophobic coatings.

To intuitively illustrate the self-cleaning function for the bifunctional coating, figures 7(c)–(f) shows photographs of the bifunctional coating and the antireflection coating under simulated muddy water pollution. The muddy water droplets maintain a complete spherical shape on the bifunctional coating and spread out on the antireflection coating. This phenomenon proves that the bifunctional coating has good hydrophobicity and
can achieve self-cleaning effect through the rolling of the water droplets. The bifunctional coating can greatly reduce the natural pollution of the transparent substrate and decrease the waste of material and human resources in the cleaning process, so as to extend the life of the coating.

3.4. Mechanical robustness of coatings

Mechanical robustness is a very important factor in the practical application of coatings. In order to evaluate the mechanical properties of the coatings, pencil hardness and 3M tape adhesion tests were carried out. Generally, if a coating is attached to the substrate surface by van der Waals interactions, it usually could not stand for pencil hardness tests [5]. As shown in figures 8(a)–(b), bifunctional coating has the most of the silica particles on the glass substrate after 5H pencil hardness test, which indicates that the dimethyl silicone significantly enhances the adhesion between nanoparticles and the adhesion of nanoparticles with the substrate. Additionally, figures 8(c)–(f) shows the surface morphology of antireflective coating and bifunctional coating after the 3M tape adhesion test. Compared with the anti-reflective coating, no obvious peeling is observed in the bifunctional coating, which means that the adhesion of the bifunctional coating is 5A (ASTM D3359 standard) [37, 38]. This further confirms that dimethyl silicone makes the bifunctional coating has excellent mechanical robustness.

4. Conclusion

In summary, we used a simple sol–gel method to prepare a bifunctional coating on the surface of the glass substrate with antireflective and hydrophobic property. The results show that the bifunctional coating has a peak transmittance of 98.11% in the visible and near-infrared wavelength range. At the same time, the water contact angle of the bifunctional coating is 140° and has an outstanding hydrophobic property, which is conducive to the self-cleaning of the coating. Moreover, the dimethyl silicone not only improves the self-cleaning performance of the coating, but also promotes the mechanical robustness. Pencil hardness test and 3M tape adhesion test proved that the bifunctional coating has a good adhesion to the substrate, this is significance for the practical application of the coating. Therefore, this study provides a feasible strategy for the preparation of bifunctional coatings with broad application prospects.

Acknowledgments

We acknowledge financially support from the National Science Foundation of China (51272147), the Natural Science Foundation of Shaanxi Province (2015JM5208) and the Graduate Innovation Found of Shaanxi University of Science and Technology.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

ORCID iDs

Hui Liu https://orcid.org/0000-0002-5966-1191

References

[1] Sun X, Tu J, Li L, Zhang W and Hu K 2020 Preparation of wide-angle and abrasion-resistant multi-layer antireflective coatings by MgF2 and SiO2, mixed sol Colloid Surface A. 602 125106
[2] Zou X et al 2018 One-step sol-gel preparation of ultralow-refractive-index porous coatings with mulberry-like hollow silica nanostructures Surf. Coat. Tech. 341 57–63
[3] Keshavarzi R, Molabahrami N, Afzali N and Omrani M 2020 Improving efficiency and stability of carbon-based perovskite solar cells by a multifunctional triple-layer system: antireflective, uv-protective, superhydrophobic, and self-cleaning Sol. RRL 4 2000491
[4] Zhang C and Xu Y 2020 Transparent and hydrophobic hexylene-bridged polyvinyletheroxide/SiO2 composite coating with tunable refractive index and its application for broadband antireflection Thin Solid Films 701 137944
[5] Xu L, Geng Z, He J and Zhou G 2014 Mechanically robust, thermally stable, broadband antireflective, and superhydrophobic thin films on glass substrates ACS Appl Mater Inter. 6 9029–35
[6] Xia B, Yan L, Li Y, Zhang S, He M, Li H, Yan H and Jiang B 2018 Preparation of silica coatings with continuously adjustable refractive indices and wettability properties via sol-gel method RSC Adv. 8 6091–8
[7] Zhang X, Xia B, Ye H, Zhang Y, Xiao B, Yan L, Lv W, Li J, and Jiang B 2012 One-step sol-gel preparation of PDMS-silica ORMOSILs as environment-resistant and crack-free thick antireflective coatings J. Mater. Chem. 22 13132
[8] Khan S, Wu H, Li J, Chen L and Zhang Z 2018 Bilayer SiO2 nanorod arrays as omnidirectional and thermally stable antireflective coating Adv. Eng. Mater. 20 1700942
[9] Dumas L, Quesnel E, Pierre F and Bertin F 2002 Optical properties of magnesium fluoride thin films produced by argon ion-beam assisted deposition J. Vac. Sci. Technol. A 20 102–6
[10] Zou L, Li X, Zhang Q and Shen J 2014 An abrasion-resistant and broadband antireflective silica coating by block copolymer assisted sol-gel method Langmuir 30 10481–6
[11] Yao C, Zou X, Reddy K M, Zhang L and Jiang B 2019 A hydrophobic ultralow refractive-index silica coating towards double-layer broadband antireflective coating with exceptionally high vacuum stability and laser-induced damage threshold Colloid Surf. A 563 340–9
[12] Sethi S K and Manik G 2018 Recent Progress in super hydrophobic/hydrophilic self-cleaning surfaces for various industrial applications: a review Polym-Plast Tech. Mat. 57 1932–52
[13] Tavakoli M M, Tsui K H, Zhang Q, He J, Yao Y, Li D and Fan Z 2015 Highly efficient flexible perovskite solar cells with antireflection and self-cleaning nanostructures ACS Nano. 9 10287–95
[14] Zhang L, Xue C H, Cao M, Zhang M M, Li M and Ma J Z 2017 Highly transparent fluorne-free superhydrophobic silica nanotube coatings Chem. Eng. J. 320 244–52
[15] Kong J H, Kim T H, Kim J H, Park J K, Lee D W, Kim S H and Kim J M 2014 Highly flexible, transparent and self-cleaning superhydrophobic films prepared by a facile and scalable nanoparticle formation technique Nanoscale. 6 1453–61
[16] Liu Z, Xu J, Wang Z, Yu Z, Weng Z and Yu H 2018 Nanosecond laser-induced underwater superoleophobic and underoil superhydrophobic mesh for oil/water separation Langmuir 34 2981–8
[17] Ni X J, Jiang G C and Yang L L 2018 The synthesis of super-hydrophobic nano-silica and its effect to the surface wettability of sandstone Mater. Sci. Forum. 917 140–4
[18] Dong S et al 2020 Springtail-inspired superamphiphobic ordered nanohoodoo arrays with quasi-doubly reentrant structures Small. 16 2000779
[19] Yokoi N, Manabe K, Tenjimbayashi M and Shioratori S 2015 Optically transparent superhydrophobic surfaces with enhanced mechanical abrasion resistance enabled by mesh structure Acs Appl. Mater. Inter. 7 4809–16
[20] Wu Y, Zeng J, Shi Y, Chen M and Wu L 2018 Large-area preparation of robust and transparent superomniphobic polymer films ACS Nano 12 10338–46
[21] Sun X, Li L, Xu X, Song G, Tu J, Yan P, Zhang W and Hu K 2020 Preparation of hydrophobic SiO2/PTFE sol and antireflective coatings for solar glass cover Optik 212 164704
[22] Cai S, Zhang Y, Zhang H, Yan H, Lv H and Jiang B 2014 Sol-gel preparation of hydrophobic silica antireflective coatings with low refractive index by base/acid two-step catalysis ACS Appl. Mater. Inter. 6 11470–5
[23] Li X, Zou L, Wu G and Shen J 2014 Laser-induced damage on ordered and amorphous sol-gel silica coatings Opt. Mater. Express 4 2478
[24] Woold S, Encinas N, Vollmer D and Butt H J 2017 Stable hydrophobic metal-oxide photocatalysts via grafting polydimethylsiloxane brush Adv. Mater. 29 1604637
[25] Xue C H, Tian Q Q, Jia S T, Zhao Q L, Ding Y R, Li H G and An Q F 2020 The fabrication of mechanically durable and stretchable superhydrophobic PDMS/SiO2 composite film RSC Adv. 10 94966–19473
[26] Ge D, Yang L, Zhang Y, Rahmawan Y and Yang S 2014 Transparent and superamphiphobic surfaces from one-step spray coating of stringed silica nanoparticles/sol solutions Part. Part. Syst. Char. 31 763–70
[27] Yoldas B E 1982 Deposition and properties of optical oxide coatings from polymerized solutions Appl. Optics. 21 2960–4
[28] Guo Q Z, Liu Y, Yang M Y, Wang J H and Su X P 2017 Super-durable closed-surface antireflection thin film by silica nanocomposites Sol. Energ. Mat. Sol. C. 170 143–9
[29] Yamada Y, Iizuka H and Mizohata N 2020 Silicon nanocone arrays via pattern transfer of mushroomlike SiO2 nanopillars for broadband antireflective surfaces ACS Nano Mater. 3 4321–40
[30] Xi R, Wang W, Wang X, Lv J, Li X, Li T, Zhang X and Du X 2020 Ultrafine nano-TiO2 loaded on dendritic porous silica nanoparticles for robust transparent antifogging self-cleaning nanocoatings Ceram. Int. 46 23651–61
[31] Gale G O 1951 Constructive and destructive interference Am. J. Phys. 19 321–32
[32] Boosnorp P and Visser M 2012 Compound transfer matrices: constructive and destructive interference J. Math. Phys. 53 012104
[33] Grassie N and Macfarlane I G 1978 The thermal degradation of polysiloxanes—I. Poly(dimethylsiloxane) Eur. Polym. J. 14 411–475
[34] Shen Z, Hou C, Liu S and Guan Z S 2014 Micro-nanostructured silicon-carbon composite coatings with superhydrophobicity and photoluminescence prepared by oxidative chemical vapor deposition J. Appl. Polym. Sci. 131 1–9
[35] Siddiqua A R, Li W, Wang F, Oü And Amiri-farí A 2021 One-step fabrication of transparent superhydrophobic surface Appl. Surf. Sci. 542 148534
[36] Kopani M, Mikula M, Kosnac D, Gregus J and Pinkci E 2017 Morphology and FT-IR spectra of porous silicon JEE. 68 53–7
[37] Yao L, Qu Z, Pang Z, Li J, Tang S, He J and Feng L 2018 Three-layered hollow nanospheres based coatings with ultra-high performance of energy-saving, antireflection, and self-cleaning for smart windows Small. 14 1801661
[38] Qu Z, Yao L, Ma S, Li J, He J, Mi L, Tang S and Feng L 2019 Rational design of HSNs/VO2 bilayer coatings with optimized optical performances and mechanical robustness for smart windows Sol. Energ. Mat. Sol. C. 200 109920