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Antireflection/antifogging coatings based on nanoporous films derived from layered double hydroxide

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ABSTRACT

Multifunctional mixed metal oxide (MMO) thin films on quartz substrates have been fabricated by layer-by-layer assembly of layered double hydroxide (LDH) nanoparticles and poly(sodium styrene 4-sulfonate) (PSS) followed by calcination, which exhibited properties of both antireflection (AR) and antifogging (AF). The AR and AF performances are related to the low refractive index and superhydrophilicity originating from the nanoporous structure. The influences of deposition cycle, LDH particle size and calcination temperature on AR and AF behavior were thoroughly studied. A maximum transmittance of 98.7% at 650 nm and a minimum time of 0.2 s for a droplet to spread flat (water contact angle \sim 0°) were achieved with LDH particle size of 110 nm, 10 cycles of deposition and calcination temperature of 450°C. Therefore, this work provides a facile approach for the fabrication of multifunctional coatings, which can be potentially used in photovoltaic devices, optical lens and underwater imaging systems.

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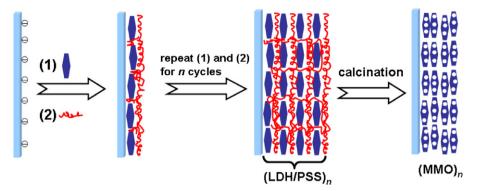
1. Introduction

Antireflective (AR) coatings have recently attracted much interest for their applications in photovoltaic, displaying devices and all kinds of optical lenses [1-3], owing to their ability to enhance the transmittance of light and remove ghost images. The principle of AR coatings is based on the destructive interference of reflected light from air-film and film-substrate interfaces. An ideal homogeneous single-layer AR coating satisfies the following conditions [4]: (1) the thickness of the coating is $\lambda/4$, where λ is the wavelength of the incident light; (2) $n_c = (n_a n_s)^{1/2}$, where n_c , n_a and n_s are the refractive indices of the coating, air and substrate, respectively. For a glass substrate ($n_s = \sim 1.5$), the refractive index of AR material should be \sim 1.22. However, nature materials with such low refractive index are either rare or expensive to obtain in thin film form. As a substitute, nanoporous materials are usually chosen as AR coatings, since the introduction of the nanopores can reduce the refractive index of the coatings and satisfy the AR requirement [5–7]. The refractive index of porous coatings can be well tailored by controlling the percentage of pores introduced, in which larger percentage of pores leads to lower refractive index. Up to now, many methods have been developed to obtain nanoporous film materials for use as AR coatings, including sol-gel process [8], phase-separation [9], sacrificial porogen approach [10–12], layer-by-layer (LBL) deposition of nanoparticle multilayers [13,14], plasma-enhanced chemical vapor deposition [15] and deposition of nanorods or nanowires [16,17]. Compared with other methods, the LBL assembly technique holds a great potential in the fabrication of AR coatings because of its simplicity in preparation of films with large area and its facility in depositing films on non-flat surfaces [18,19].

Although the materials and methods for the preparation of porous AR coatings are diverse, there are still several serious issues should be concerned for the application of the resultant AR coatings, especially the influence of water vapor existing in the external environment. Water vapor molecules condense and form water droplets on the surface, resulting in scattering and reflection of light, which in turn decreases the transmittance of the AR coating. A superhydrophilic coating, on which the water contact angle is less than 5° within 0.5 s as soon as a water droplet contacts such surface, can solve this problem [20-22]. Superhydrophilic coatings can significantly suppress the fogging behavior by the rapid spread and flow of water droplets on their surface and therefore eliminate the light scattering caused by water droplets. In fact, AR coatings combined with an antifogging (AF) capability are highly desired in daily used eyeglasses, swimming goggles, periscopes, lenses in endoscopic surgery, and so forth.

Layered double hydroxides (LDH) are layered anionic clays generally expressed by the formula $[M_{1-x}^{2+}M_{\chi}^{3+}(OH)_2](A^{n-})\cdot mH_2O$, where M^{2+} and M^{3+} are di- and trivalent metal cations and A^{n-} is a counter anion [23–26]. The host structure consists of brucite-like layers of edge-sharing $M(OH)_6$ octahedra, and the partial substitution of M^{3+} for M^{2+} results in positively-charged host layers, balanced by the interlayer anions [27–30]. It is well-known that calcination of LDH at intermediate temperatures in the range

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Scheme 1. Schematic illustration for the fabrication of MMO nanoporous coatings.

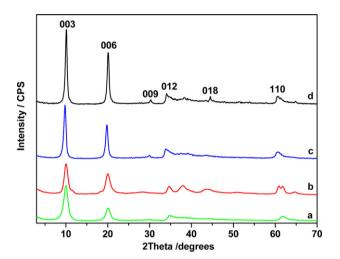


Fig. 1. XRD patterns for the MgAl-LDH materials prepared at $110\,^{\circ}$ C with different aging time: (a) 4 h, (b) 8 h, (c) 18 h and (d) 48 h.

300–600 °C leads to the formation of mixed metal oxides (MMO) which possess porous structures and large specific surface areas as well as good thermal stability [31–33]. In our previous work [34], a nanoporous MMO coating was fabricated by calcination of

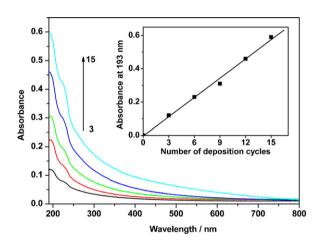


Fig. 3. UV–vis absorption spectra of the $(LDH-25/PSS)_n$ (n=3-15) films assembled on quartz glass substrates. The absorbance at 193 nm is plotted against the bilayer number in the inset.

 $(LDH/PSS)_n$ ('n' denotes the number of deposition cycles) multilayer films on quartz glass substrate, which exhibited erasable AR properties with excellent mechanical stability and good adhesion to substrates. However, detailed AR properties of the MMO coating

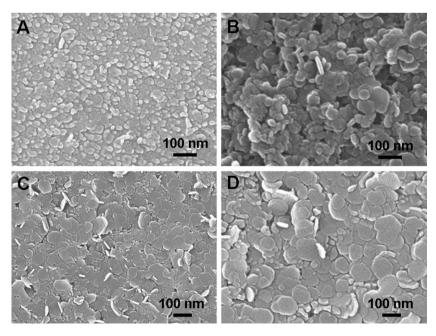


Fig. 2. SEM images for the MgAl-LDH materials prepared at 110 °C with different aging time: (a) 4 h, (b) 8 h, (c) 18 h and (d) 48 h.

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