# Preparation of broadband antireflective coating by sol-gel method

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A sol-gel method for the preparation of silica coating that varied in refractive index is developed. Silicon dioxide sol is obtained by hydrolysis and co-condensation reactions occurred in acid-catalyzed system. Surface morphology, refractive index, and transmission spectrum of the samples are studied. The results of transmission spectra of single-sided coatings show that the average transmittance of the samples increases about 4% compared with the uncoated one in the spectra range from 400 to 1 200 nm in the case of vertical incidence. For the double-sided coating the maximum transmittance is 99% at the wavelength of 840 nm.

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Antireflective coatings (ARCs) are widely used to increase transmission in industries such as photovoltaic, building, display, and ophthalmic lens[1−6]. For example, the efficiency of solar power plants for the production of electricity depends primarily on the intensity of the incident sunlight. A basic drawback of any transparent cover material used to minimize convection and radiation losses is the reflection of the incident sunlight at its surface[7], so ARCs are strongly needed in photovoltaic industry. Zero reflectance cannot be achieved with a single layer coating due to the absence of suitable low refractive index materials[8]. Many researches focus on a two-layer system in which a thin layer of material with a high refractive index is placed next to the substrate so as to make it appear to have a higher refractive index, hence the layer of low refractive index is more effective. Titanium dioxide and aluminum dioxide are common materials for these structures. However, a two-layer ARC has a narrower bandwidth though provides a lower reflectance at a given wavelength.

Porous silica are widely used as single layer ARCs because it can achieve lower refractive index than $MgF_2$ which has a refractive index of 1.38, the lowest for any solid material. Current single layer ARC technology relies primarily upon vacuum deposition techniques such as sputtering or physical and chemical vapor deposition[9,10]. High-quality vacuum deposited coatings are expensive and consequently limit application in cost sensitive industries. To solve this problem, the first and most common approach is the sol-gel route, in which chemical precursors (inorganic salts or metal alkoxides) are mixed with solvent and catalyst to cause hydrolysis and co-condensation[11]. Sol-gel coating equipment is simple, and the film can be operated at room temperature and atmospheric pressure. The films made by sol-gel method are highly uniform, microstructure controllable and adapted to different shape, size of substrates.

In this letter, we prepare porous silica film on K9 glass by sol-gel method, and applied polyethylene glycol (PEG) to increase the porosity of coatings which is directly relate to the refractive index. Surface morphology, refractive index, and transmission spectrum are studied. The lowest refractive index we achieved in our study is 1.26. The coating transmittance increases about 4% compared to the uncoated one in the spectra range from 400 to 1 200 nm. Double-sided coating further increases the transmittance to 99% at the wavelength of 840 nm.

Silica sol were prepared from tetraethyl orthosilicate (TEOS) according to the following procedure. Ethanol and TEOS (solution 1), ethanol , hydrochloric acid (HCl), and distilled water (solution 2) were stirred separately at room temperature for 30 min, then solution 1 was added to solution 2 dropwise for hydrolysis, and the resulting solution was used for coating. The molar ratio of ethanol, TEOS, distilled water, and HCl was 20:1:4:0.01. After aging for 3 days the sol should be filtrated before spin. To increase porous density of films, 0.3, 0.6, 0.9-g PEG 20000 were added to 10-ml as-prepared sol respectively. Sols were spun on K9 glass ($n$=1.516). The obtained coatings were heat-treated at 450 °C for 5 h. For double-sided coating, the first side coatings were sintered at 200 °C for 20 min before the second side was spin-coated to avoid destroying of the first side coating.

The surface morphologies of the coating films were observed with a field emission scanning electron microscope (FESEM, FEI Sirion-200, USA). Optical transmission spectra were measured with double-beam ultraviolet-visible spectrophotometer (Lamda-900, Perlin-Elmer, USA). Refractive indexes of films were investigated with ellipsometer (J. A. Woollam Co., USA).

Figure 1 shows micrographs of porous $SiO_2$ films. The coatings possess smooth and even surface. With increasing amount of PEG involved porous density increases

 

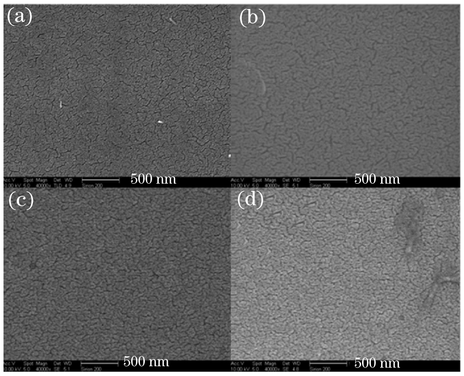


Fig. 1. SEM images of top view of ARCs prepared from (a) pure silica sol, (b) 10-ml silica sol with 0.3-g PEG, (c) 10-ml silica sol with 0.6-g PEG, and (d) 10-ml silica sol with 0.9-g PEG.

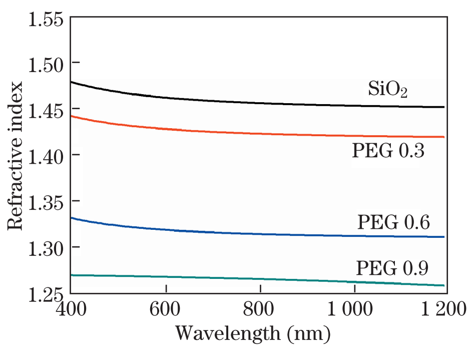


Fig. 2. (Color online) Refractive indexes of ARCs prepared from different sols.

obviously because more organic materials in coatings were removed when the films were heat-treated at 450 °C.

Refractive indexes of ARCs prepared from different sols are showed in Fig. 2. With increasing amount of PEG involved refractive index decreases. For a particular film the refractive index decreased slightly from 400 to 1 200 nm. When 0.9-g PEG was added to 10-ml silica sol we achieved the minimum refractive index of 1.26, which is very close to the theoretical requirement for zero reflection according to the formula $n = (n_0 n_{\mathrm{m}})^{1/2}$, where $n_0$ is the refractive index of atmosphere which is always defined as 1, and $n_m$ is the refractive index of the substrate, here is 1.516 for K9 glass, so when $n$=1.23 the reflection equals about zero.

At the wavelength of 550 nm the refractive indexes ranges from 1.47 to 1.27 (Fig. 3). By changing the amount of PEG we can prepare films with any refractive index within this range. According to the relationship of refractive indexes between ARCs and substrates the films can be applied to substrates with refractive index ranging from 2.16 to 1.69 with good performance.

Figure 4 shows the transmission spectra of uncoated and coated K9 glass, coatings with PEG compared with the pure silica coating. The results show that for the film prepared from pure silica sol, the ARC has a relatively broadband response throughout the spectrum from 400 to 1 200 nm, the average transmittance increased about 4% compared to the uncoated one. For the sample with 0.3-g PEG, the transmittance decreases compared to the pure silica one. Further addition of PEG results in ups and downs of transmittance due to the increase of film thickness causes interference of light. The sample with 0.9-g PEG, with the refractive index of 1.26, has the highest transmittance from 700 to 850 nm, and increases the maximum in transmittance of the uncoated K9 glass from 91.5% to 96% at wavelength of 820 nm. By changing the concentration and spin coating conditions the film can be controlled at an optimized thickness to remove the interference of light with no loss of performance, which will be investigated in the future.

Applications such as solar and optical windows may require an ARC with higher transmittance which can be achieved by double-sided coating. Figure 5 indicates that the transmittance of double-sided coating prepared by pure silica sol increases another 2%–3% in comparison to single-sided coating from 500 to 1 200 nm. At the

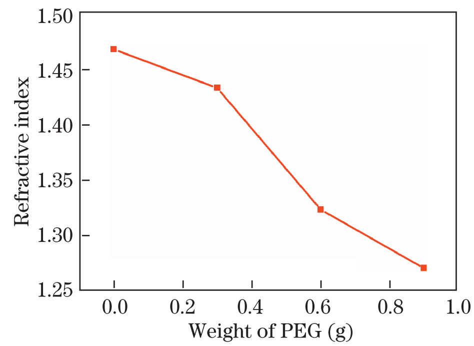


Fig. 3. Refractive indexes of ARCs at 550 nm.

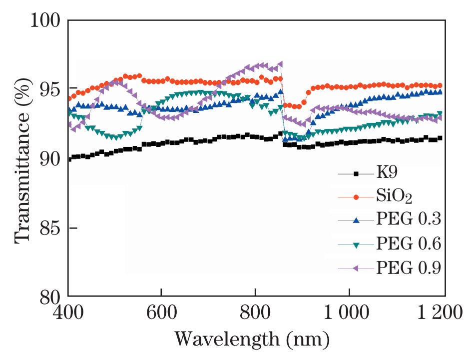


Fig. 4. (Color online) Transmission spectra of porous silica coatings on K9 glass, containing 0, 0.3, 0.6, and 0.9-g PEG in 10-mL sol in comparison to an uncoated K9 glass (All the curves have a mutation from 856 to 906 nm because the double-beam ultraviolet-visible spectrophotometer change detector at this range of wavelength).

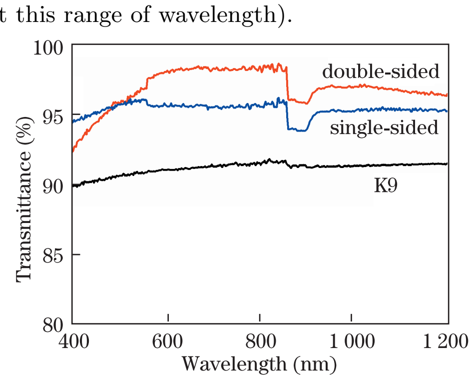


Fig. 5. (Color online) Transmission spectra of double-sided coating in comparison to single-sided coating and an uncoated K9 glass.

wavelength of 840 nm, the maximum transmittance is 99%.

In conclusion, porous silica films on K9 glass are prepared by sol-gel method, PEG is applied to increase the porosity of coatings which is directly relate to the refractive index. The lowest refractive index achieved is 1.26. The coatings transmittance increases about 4% compared with the uncoated one in the spectra range from 400 to 1 200 nm. Double-sided coating can increase the transmittance further to 99% at the wavelength of 840 nm.

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