

Fabrication of sub-wavelength structures on silicon dioxide

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In this reported work, nanosphere lithography (NSL) and inductively coupled plasma reactive ion etching (ICP-RIE) are combined to successfully fabricate a sub-wavelength structure (SWS) on a glass substrate, achieving broadband antireflection and increasing the transmittance of incident light through the glass. The experimental results show that the SWS surfaces with 180 nm width and 50 nm height could be fabricated onto glass. The mean reflectance of a blank glass is 5.81% in the wavelength range of 400–950 nm, 3 min of ICP-RIE combined with NSL reduce the mean reflectance to 3.5% and increases the mean transmittance from 92.3 to 94.3%. An additional coat of a 200 Å-thick gold layer on the 30 s etched surface sample reduces the transmittance in the visible light range (400–700 nm) to 36.6%, which is 2.25 times larger than that at the infrared range (700–950 nm). The proposed novel fabrication technology has the advantage of being low cost, and the fabricated nanodot array structure, which is gold coated, can be used on an insulated window.

1. Introduction: Recently, studies have been performed on the sub-wavelength structure (SWS) of silicon material [1–4], aimed at reducing the reflectance of the silicon wafer by fabricating a microscaled or nanoscaled structure onto the wafer surface. The SWS has a periodicity smaller than the wavelength of light, so that a strong antireflection effect can be produced [5]. In addition, the SWS is generated on a transparent material to minimise light reflection and maximise transmission [6–8]. Traditionally, antireflection coating (ARC) technology is intended to suppress reflectance on transparent material. However, preparing the ARC requires complex theoretical calculations and the thickness must be monitored to high precision. ARC is a different material than the substrate with less mechanical stability than the SWS.

The SWS can be fabricated on substrates by lithography process followed by using an etching technique. For defining arrayed nanoscaled patterns, the approaches include deep ultraviolet lithography [9], electron-beam lithography [10], interference lithography [11] and nanoimprint lithography [12]. Although such methods can define the nanoscaled pattern precisely, those techniques are expensive with respect to equipment and fabricating process. Excepting these expensive approaches, the nanoscaled array pattern can also be defined by nanosphere lithography (NSL) [13, 14], and the pattern can be determined effectively by appropriately selecting the sphere size and controlling the number of nanospheres per layer.

In this Letter, we present an approach which integrates NSL and inductively coupled plasma reactive ion etching (ICP-RIE) to yield a SWS on glass, followed by a coated metal layer to be applied on an insulated window. Here, we employed the microscope slide as substrate, which is an inexpensive common glass. The process parameters developed in this study have significant potential for applications in the fields of windshields, building glass and cooler doors.

2. Design and fabrication: The schematic fabrication process of a sub-wavelength structured glass array is shown in Fig. 1, which mainly consists of NSL and ICP-RIE. A common glass (microscope slides) (1.2 mm thick) was used as substrate and then reduced to a surface area of 1 cm². First, we generated a monolayer of nanospheres using a nanosphere lithography solution which was prepared by a 2.5% (w/v) suspension of polystyrene nanospheres (200 nm diameter) (Polysciences, Inc.), methanol and Triton X-100 in a volume ratio of 800:400:1. The masked glass samples were prepared using ICP-RIE with

parameters of a mixture of two gases at a ratio of C₄F₈/He:12/84 sccm and constant pressure of 10 mTorr, ICP etching power of 1500 W and RF etching power of 120 W to yield a submicron structure array with a periodic arrangement. Then, PS nanospheres were removed by applying a piranha solution (H₂SO₄:H₂O₂ = 3:1) for 10 min. Using this method, the AR surface constructed by nanodot arrays eventually formed. Furthermore, a gold layer was coated on a sample surface to enhance reflectance in the infrared wavelength range.

A scanning electron microscope (SEM, HITACHI S-4300) and scanning probe microscopy (SPM) (Veeco Dimension-3100) were employed to observe the micrographs generated by the etch process and the experimental results were discussed. Finally, a spectrometer (B&W BRC-112E) was used to evaluate reflectance in the wavelength range of 400–950 nm for blank and processed samples.

3. Experimental results: Fig. 2 shows the nanosphere array, which was fabricated by spin-coating processes. The arrayed nanospheres are in a near-perfect arrangement, but some defects are observed in the monolayer of the nanosphere film, such as a polygonal crystalline lattice as well as voids and disordered grain boundaries. The formation of these defects is a complex process since many experimental parameters, such as surface roughness, ionic strength, surface tension and charge on the nanospheres surface influence the formation process.

The etched samples with various process times from 10 to 180 s are shown in Fig. 3. Fig. 3a indicates that the arrayed nanostructure had an etching depth of 10 nm when the etching time was 10 s. Fig. 3a also presents six nanopores periodically arranged around the nanodots after ICP-RIE etching; these results indicate that the nanospheres were not damaged during the 10 s ICP-RIE process. When the etching time was increased to 30 s, the diameter of the etched nanodots was about 180 nm, which is smaller than the minimum wavelength of the test light source (400 nm), and the etching depth is approximately 35 nm (Figs. 3b and 4a). When the etching time is greater than 10 s, the six pores expanded to connect with each other and a nanodot structure was generated at the centre of the arrangement of the six pores. Figs. 3c and 4b show that the height of the arrayed structures increases to about 54 nm and the etched surface on the nanodot had a slight roughness for etching times of 60 s. Figs. 3b–e indicate that the height of the arrayed nanodots is almost the same when the etching time exceeds 60 s. On the other hand, the etched surface on the nanodot top gradually became rougher, but the grass-like structures could be

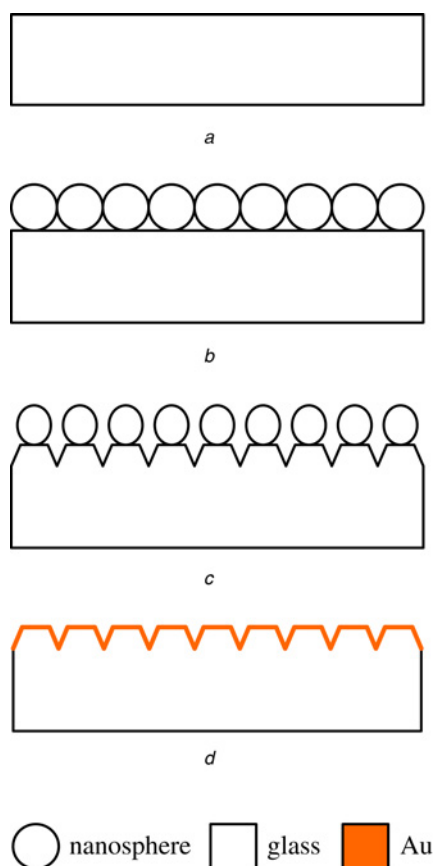


Figure 1 Schematic of SWS fabrication processes
a Using glass as substrate
b Nanospheres deposited on substrate
c RIE process applied to form arrayed nanostructure
d Removal of nanospheres and coated on Au layer

helpful in suppressing reflection. The rough surfaces would be because of PS nanospheres which disappeared after the etching process of 60 s. Fig. 3*f* shows a sample with an etching time of 3 min. Random coral-like structures appear on the glass surface, and no sphere-like definitions can be observed. The etching morphology using microscope slides is very different from our previous work [15], which employs quartz as a substrate and yields arrayed nanocone structures 190 nm in height and 108 nm in diameter. The difference could be attributed to microscope slides containing impurities such as Na, Mg and Ca (Fig. 5). Those impurities serve as a native etching mask, which disturbs the etching reaction on silicon oxide.

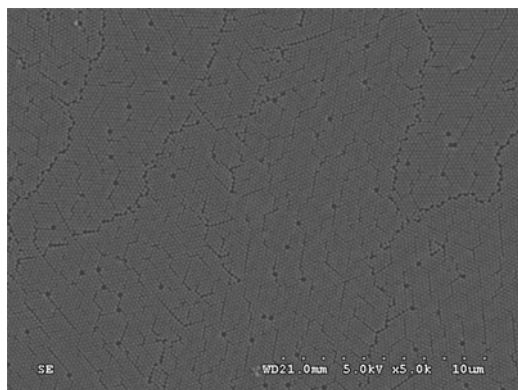


Figure 2 SEM micrographs of nanosphere array deposited by spin coater

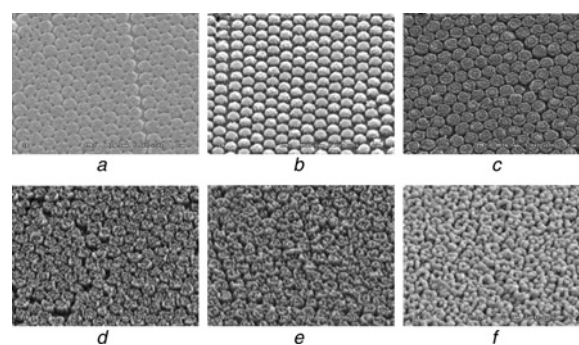


Figure 3 SEM micrographs of arrayed structures formed after C_4F_8 ICP-RIE for different etching times

a 10 s
b 30 s
c 60 s
d 90 s
e 180 s
f 300 s

The spectrometer obtains the reflectance and transmittance spectra in the range of 400–950 nm to investigate etched samples with different morphology (Fig. 6*a*). Fig. 6*a* indicates that the mean reflectance of a blank glass is 5.81% in the wavelength range of 400–950 nm. If nanospheres of 200 nm were coated on the substrate surface, the reflectance increased to 6.42%. After 30, 90 and 120 s of ICP-RIE, the mean reflectance is reduced to 5.08, 4.81 and 4.48%, respectively. Etching for 180 s yields the best anti-reflection effect, and the mean reflectance is 3.5%. The transmittance improved as the etch time increased (Fig. 6*b*). The mean transmittance (400–950 nm) of a blank glass and etched sample with 180 s is 92.3 and 94.3%, respectively. Therefore the improvements in reflectance and transmittance are only 2.31 and 2%. The

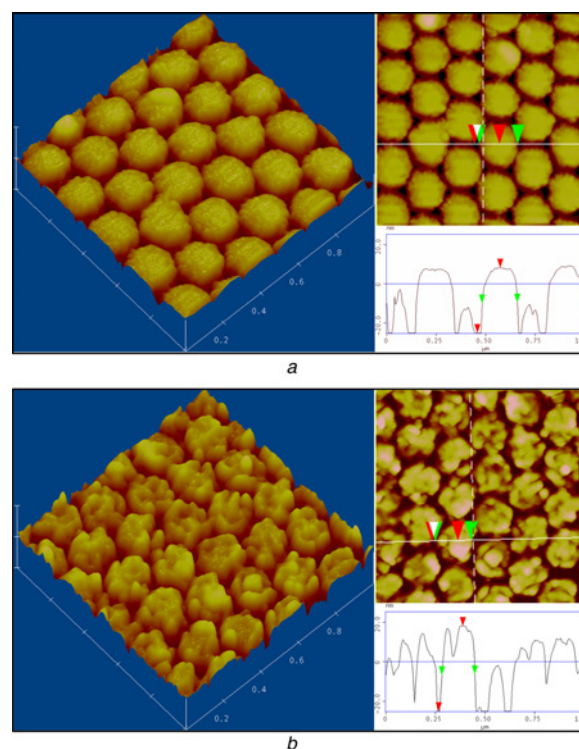


Figure 4 AFM images of nanodot arrays processed by ICP-RIE for 30 and 60 s

a 30 s
b 60 s

result could be because of quick exhaustion of the nanospheres during the ICP-RIE process; hence, the height of the arrayed structure cannot effectively increase. Hadobás *et al.* [5] report that the

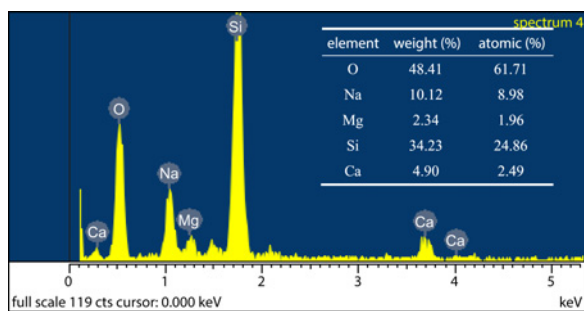


Figure 5 EDS analysis of substrate material

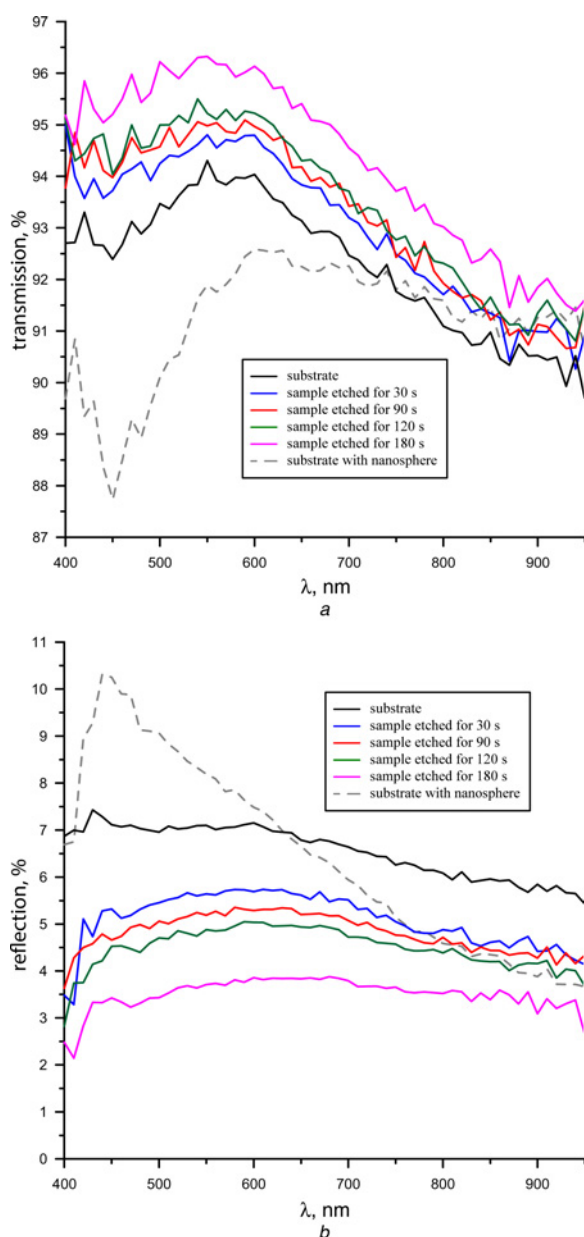


Figure 6 Transmission and reflection of slide glass coated by nanospheres and etched for various times
a Transmission
b Reflection

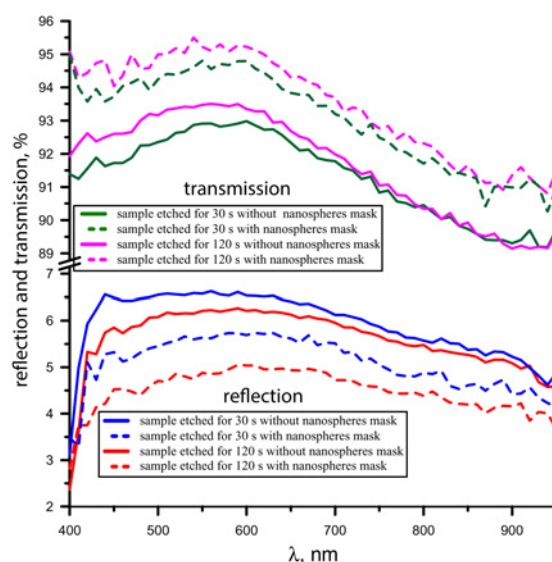


Figure 7 Reflection and transmission of samples etched with and without nanospheres

reflectance of incident light varies with the depth of the periodic structure, indicating that the effect on suppressing reflectance is limited. We prepared a blank glass to process ICP-RIE to examine whether the arrayed nanostructure suppresses reflectance. Fig. 7 indicates that the mean reflectance of the sample etched for 30 and 120 s without NSL are 5.93 and 5.60%, respectively, which are higher than that etched with nanosphere (5.08 and 4.48%, respectively). The NSL defined samples also have higher transmittance than that etched without the nanosphere.

Hass [16] indicates that gold has a higher reflectance at the infrared range (>700 nm) compared with the visible wavelength range (400–700 nm). For this reason, we deposited a gold thin film of 50 nm on samples according to the best parameters [8]. Fig. 8a shows that a 50 nm gold layer on SWS samples increases reflectance significantly in the broadband. In addition, the increment in the infrared range is higher than that in the visible range. The result suggests that gold coating of 50 nm insulates heat well. However, the transmission of 50 nm gold samples is simultaneously more than 10%. Hence, the 50 nm coating cannot be used to insulate windows. Consequently, we attempted to reduce the thickness of gold film to 20 nm. For the samples coated with 20 nm of gold, the transmittance at broadband is about half of the uncoated gold. The mean transmittances for all samples are similar in the visible light range (36.6, 37.1, 38.6 and 40.0% for samples etched using 30, 90, 120 and 180 s, respectively). For the infrared range, the sample etched for 30 s had the lowest mean transmittance of 16.6% compared with the other three samples (34.0, 35.5 and 40.1% for the 90, 120 and 180 s etched samples, respectively). Hence, the sample etched for 30 s displayed the largest difference of mean transmission between the infrared range and visible light range. At the same time, for the mean reflectance, the 30 s etched sample also yielded the largest difference between the infrared range (54.6%) and visible light range (33.1%). The results indicated that the sample etched for 30 s, using 20 nm gold, could be used to insulate glass.

4. Conclusion: We have developed an effective approach for depositing a SWS directly onto a low-cost glass substrate. As anticipated, the proposed approach resulted in an orderly arranged nanosphere monolayer on a 1×1 cm sample. When an ICP-RIE etching time of 60 s is applied, the arrayed nanodots are about 50 nm in height and 180 nm in diameter. The impurities inside the glass result in a rougher nanodot top surface as the etching

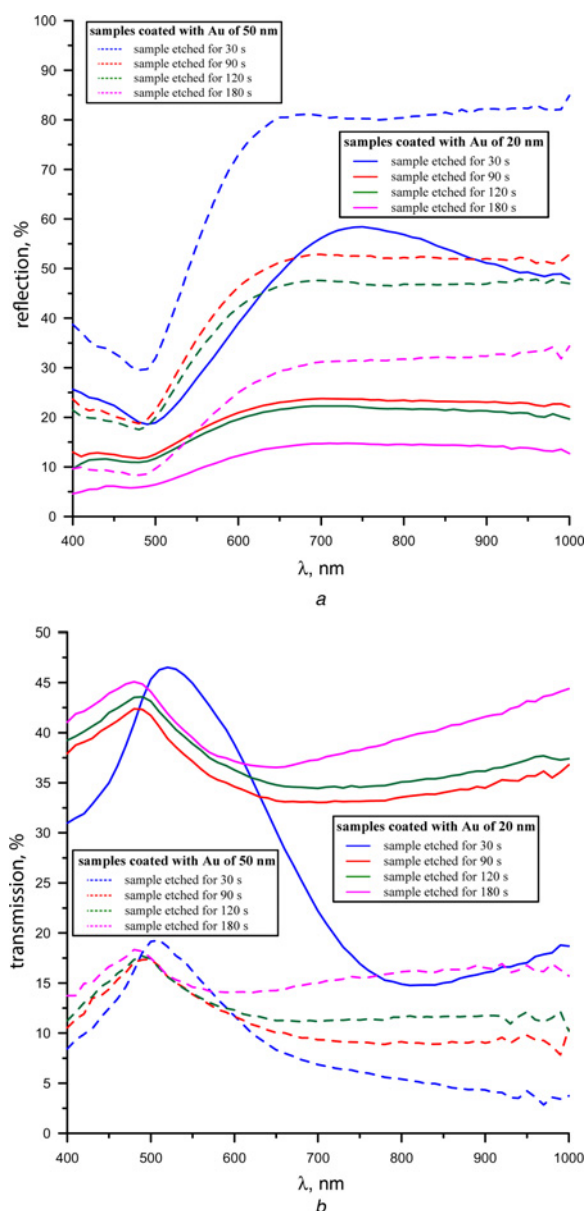


Figure 8 Reflection and transmission of samples coated with Au of 20 and 50 nm
a Reflection
b Transmission

time increases. For the optical measurement, the reflectance decreases and transmittance rises with increasing etching time. The mean reflectance (400–950 nm) of a 180 s etched glass can be reduced from 5.81 to 3.5%. When a 20 nm Au thin film has been deposited onto the 30 s etched periodic nanostructure, the

mean transmittance is only 16.3% in the infrared range (700–950 nm) and 36.6% in the visible range (400–700 nm). The mean transmittance in the visible light range is 2.25 times higher than that in the infrared range, indicating that a SWS coated with a gold layer is suitable for heat-insulated windows.

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6 References

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