Low angle-dependent structural colours from redispersion of silica photonic crystal film

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Abstract. Structural colour has attracted more and more attentions owing to its advantages of non-fading, environment friendly and non-toxic. According to its behaviour with respect to the viewing angle, there are two groups: iridescent (angle-dependent) and nor iridescent (angle-independent). However, many applications in textile and fashion require stable colour which means the colour shows low angle dependent. In this work, 314nm silica nanoparticle was synthesized by using a solvent varying technic which based on Stöber method. Iridescent structurally coloured film was directly self-assembled by gravity sedimentation of silica nanoparticle. Then, film was triturated and redispersed by ultrasonic dispersion process. Low angle-dependent structurally coloured film was then obtained by the redispersed silica nanoparticles. The effect of redispersion time on the structural colours was studied. The result shows that a twenty-minute redispersion gives a less bright but low angle-dependent structural colour film.

1. Introduction
Colour is a significant information carrier between environment and living organisms. A well-known example is chameleons have unique ability to change colours when in danger or during social interactions [1]. Colour also plays a vital role in human society in decoration, textiles, information transform and more. In general, colour of two mechanisms constitutes the colourful daily life. The first one is the selectively absorption, reflection, and transmission light of a specific wavelength. In essence, it comes from the transition of electrons between molecular orbitals. Flowers, paints, pigments and dyes generate colour in this way [2]. The second one is resulting from scattering, interference or diffraction of light from micro- or nanostructures, which also called structural colour [3]. This colour is quite common in nature. For example, the precious opal gemstone, the peacock feathers and hair of sea mouse [4]. The first method suffered from poor coloration quality, fading and toxicity, while the latter one provide a no-fading, environmental friendly colour in a wide range wavelength [5].

Photonic crystal is one kind of structural colour, which can be divided in two groups: iridescent and non-iridescent [6]. The first one changes colour when the viewing angle changed. And the latter offers colour independent of viewing angle. Non-iridescent photonic crystal possesses a quasi-ordered arrangement of colloid particles [7,8], and has increasingly attracted the attention of researchers owing to their wide range of potential applications [9]. For instance, Huifang Shen et al. [10] reported the formation of mechanically stable non-iridescent structural colour coating by using a thermal assembly method. This facile fabricating provides the application including textile coloration, painting, display and other colour-related fields.

In this paper, uniform SiO$_2$ nanoparticles were prepared by a solvent varying method [11]. Then, a shorted range ordered arrangement colloid films were obtained by redispersion. The relationship
between the colour of photonic crystals and the diameters of microspheres or viewing angles was investigated, which was in great agreement with the Bragg’s law of diffraction. Especially, we designed and did experiments to study the relationship between time of the redispersion and colour. A conclusion was found that the longer the redispersion time was, the lighter but less angle-dependent colour was. The most suitable condition was 20 minutes for redispersion.

2. Experiment

2.1. Material
Tetraethyl orthosilicate (TEOS, >99%) was purchased from Macklin. Ammonia solution (NH₃, 25% in H₂O) purchased from Merck KGaA and ethanol (EtOH, ≥99.9%) which was obtained from Yonghua Chemical Co., the hydrolyzing agent water (H₂O) was purchased from Watsons Group (Hong Kong) Ltd. All the materials were used as received without any further purification.

2.2. Synthesis of silica nanoparticles
Uniform silica nanoparticles were synthesized by a solvent varying method as previously described. A mother solution containing ammonia solution, hydrolyzing agent water and certain amount of ethanol was prepared in a 250 ml round-bottomed flask under a vigorous stirring in a water bath. When the temperature of the bath reached to a set temperature, TEOS was then added into the solution immediately. The solution was stirred for at least 2 hours until the reaction completed. The diameter of the particles was controlled by just varying the volume of ethanol.

2.3. Fabrication of photonic films
Photonic crystal silica films were obtained by using gravity sedimentation. Firstly, 5 ml of silica suspension was added to Perti dish, after drying at room temperature, photonic crystal film was self-assembled. Then, 5 ml of ethanol was added to the Petri dish containing the film. This suspension was redispersed by ultrasonic machine for a period of time (10mins – 30mins). A new photonic crystal film was prepared after drying at room temperature.

2.4. Characterisation
The diameter and size distribution of the silica colloid particles were determined using dynamic light scattering (DLS, Zetasizer Nano ZS, Malvern Instrument, Worcestershire, UK). The image of the films was captured using camera (SONY, A7 III, Minato City, Japan) and the digital camera of microscope (BH200M, Huiguang Technology Co., Ltd., Suzhou, China).

3. Results and discussion

3.1. Synthesis of silica nanoparticles
It is well known that the diameter of the particle is a significant element of colour of the film, so controlling the particle size is the first priority. In this article, silica nanoparticle of 325 nm was synthesized.
3.2. Angle-dependent colour of silica films

In order to obtain the angle-dependent photonic crystal film of silica nanoparticles, a sedimentation method was applied. The as-synthesized silica nanoparticles dispersion was casted on a Petri dish. Solvent evaporated at room temperature and a photonic crystal film composed of silica nanoparticles was formed. Photos of films were taken and showed as follows. Figure 3 showed the pictures of the same film in a changing viewing angle.
Incident angle $\theta$ ($0^\circ$~ $60^\circ$)

Figure 3. Photographs of the iridescent silica film

Obviously, figure 3 shows that the same film in different viewing angle. It is clear that when viewing angle changed, colour of films also changed from red to blue. According to the Bragg-Snell equation:

$$\lambda = \frac{2d_{(111)} \sqrt{n_{eff}^2 - \sin^2 \theta}}{111}$$  \hspace{1cm} (1)

where $\lambda$ is the wavelength of the colour, $d_{(111)}$ is the lattice spacing of the photonic crystal structure, $\theta$ is the incident angle [11].

According to the equation, when the incident angle increased, the wavelength will decrease, so that colour of films observed changed from red to blue (this is known as blue-shift).

3.3. Low angle-dependent colour of silica nanoparticles

In order to obtain low angle-dependent photonic crystal films, redispersion of silica nanoparticles was subjected to the angle-dependent films. Angle-dependent film was triturated and the certain fresh solvent (EtOH) was added to the fragmented photonic crystals, and ultrasonic dispersion was applied to the solution. The redispersion time was controlled as 10 minutes, 20 minutes, and 30 minutes. And then, redispersed suspension evaporated and dried out at room temperature again so that a processed photonic crystal film was obtained. Figure 4 shows photograph of crystal films after ultrasonic dispersion at normal incident. Figure 5 shows photograph of the photonic crystal film prepared by 20 minutes redispersion in a different viewing angle.

As it shows in Figure 4, photonic crystal films with redispersion process performed a dimmer colour than the original film, but the hue of the colour remains red as the original film. Comparing the processed films, it is obvious that a 20-minutes process gives a much brighter than the others, and all processed films was duller than the original film. The decrease of the saturation of the film might cause by the aggregation of parts of silica nanoparticles. As the particles isolated and packed during the first sedimentation, it is hard to isolated the particles once they self-assembles into a film [12]. Although it is hard to isolate all the particles once they self-assembles into crystal film, the film can be smashed into tiny blogs with ordered silica nanoparticles, which is called short-range-ordered-long-range-disordered structure. So the duller film was obtained.
Figure 5 shows pictures of the 20 minutes redispersion film in a changing viewing angle. It can be shown in Figure 1, when incident angle changed from 0° to 60°, colour changed from red to yellow to blue. But in Figure 3, colour was changed from red to orange to yellow. The range of colour change was compressed, which means low angle dependence colour was formed.

4. Conclusion
In this study, silica nanoparticles with 325 nm were synthesized by a solvent varying technique based on Stober method. Low angle-dependent structural colours were prepared by redispersion of silica PC films using ultrasonic machine. The original PC films without redispersion showed iridescent angle-dependent structural colour from red to blue, while the PC films after redispersion showed low angle-dependent structural colours from red to yellow. The optimum ultrasonic time to achieve the highest quality low-angle dependent coloured films was 20 minutes, as the structural colour was the most vivid and saturated.

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