ABSTRACT: Structural color as a revolutionary coloration strategy has been proposed to replace the traditional dyeing and printing process. However, the poor colorfastness and easy crack formation of structural colors on textile fabrics restrict their practical application at present. In this study, poly (tert-butyl acrylate-co-glycidyl methacrylate) (P(t-BA-co-GMA)) copolymers containing reactive epoxy groups with different mass ratios of tert-butyl acrylate (t-BA) and glycidyl methacrylate (GMA) were successfully synthesized, which were used to create structural colors on black polyester fabrics. The results showed that P(t-BA-co-GMA) nanospheres could form crack-free structural colors on polyester fabrics, and the colors vary with the mass ratio of t-BA and GMA to obtain five different colors. The different particle sizes of the different P(t-BA-co-GMA) nanospheres with different refractive indexes and the arrangement of short-range ordered and long-range disordered in microstructures may be the reason of different angle-independent structural colors on polyester fabrics. The P(t-BA-co-GMA) nanosphere structural colors on polyester fabrics possess good abrasion and washing colorfastness. This research provides the experimental basis for the development of crack-free amorphous photonic crystal structural color on fabrics with high colorfastness to promote the practical application of structural color in textile coloration.

1. INTRODUCTION

Dyes and pigments are widely used colorants for textile through dyeing and printing, which produce bright colors by selectively absorbing and reflecting certain wavelengths in the range of visible light.\textsuperscript{1−3} However, traditional dyeing and printing industry consumes a large amount of fresh water and energy and produces a large amount of waste water, which could not meet the clean production requirements of greenness, low-carbon emission, ecofriendliness.\textsuperscript{4,5} Therefore, there is an urgent need to develop a novel green and energy-saving textile colorization method.

In nature, there are vivid structural colors, such as peacock feather, wings of Morpho butterflies, and outer shells of jewel beetles.\textsuperscript{6,7} Structural color is formed by the interaction between light and the micro/nanostructure of materials, such as light dispersion, interference, and diffraction.\textsuperscript{8,9} As long as the micro/nanostructure of materials is not destroyed, its structural color will never fade. Structural color as a revolutionary coloration strategy has been proposed to replace the traditional dyeing and printing process.\textsuperscript{10,11} Artificial brilliant structural colors possess various advantages compared with traditional dyes and pigments,\textsuperscript{12−14} such as fadeless, high stability, and ecofriendly properties, which have been widely investigated in the recent decades.\textsuperscript{15−17}

Photonic crystals (PCs) are a kind of photonic nanomaterial with a photonic band gap, which was formed by closely packed periodic arrangement of materials with different reflective indexes and have received more and more attention for
creating structural colors and PCs have been reported to create the structural colors on smooth, seamless, and rigid solid substrates such as glass and silicon. However, textile fabrics consist of a network of natural or chemical fibers containing voids and gaps between fibers and yarns, which are quite different from the solid substrates. Hence, it is more difficult to create the structural colors on textile fabrics than on the solid substrates. Shao and co-workers have extensively studied the production of monodisperse PC structural colors on different kinds of textiles by using various methods. They also have studied the correlations between the structural colors of PCs and the base colors of textile.

Although various efforts have been made on the PC structural colors in the textile colorization field, it still faces a dilemma that artificial PC structural colors cannot meet the requirement of practical application. The restrictions of current dilemma that arti-structural colors in the textile colorization PCs and the base colors of textile. have studied the correlations between the structural colors of

2. RESULTS AND DISCUSSION

2.1. Characterization of the P(t-BA-co-GMA) Copolymer. The chemical structure of the P(t-BA-co-GMA) copolymer was characterized by FTIR, which is shown in Figure 1.

As can be seen from Figure 1, the curve of t-BA shows the peak at 2980 cm\(^{-1}\), which is attributed to the stretching vibration of \(-\text{CH}_2\). The peak at 1730 cm\(^{-1}\) is assigned to the stretching vibration of ester \(\text{C}=\text{O}\), and the absorption peak at 1630 cm\(^{-1}\) belongs to the stretching vibration of the \(\text{C}==\text{C}\) bond. The curve of GMA shows the saturated and unsaturated \(\text{C}==\text{H}\) stretching vibration peaks at 2980 and 2930 cm\(^{-1}\). There also exist the peaks at 1730 and 1630 cm\(^{-1}\) corresponding to the ester \(\text{C}=\text{O}\) bond and \(\text{C}==\text{C}\) bond, respectively. There exist the \(\text{C}==\text{H}\) deformation vibration peaks at 1450 and 1310 cm\(^{-1}\). The peak at 1161 cm\(^{-1}\) is assigned to the stretching vibration of the \(\text{C}==\text{O}\)–\(\text{C}\) bond. The peaks at 908 and 842 cm\(^{-1}\) are attributed to the skeletal vibration of the epoxy group. In the curve of \(\text{P}(\text{t-BA-co-GMA})\) copolymer, the peaks of the chemical shifts at 17.24 and 27.89 ppm belong to the carbon atoms of \(\text{GMA}\). The peak at 4.28 ppm is attributed to the protons in \(\text{GMA}\). As can be seen from Figure 2a, the peaks of the chemical shift at 1.26 and 1.43 ppm belong to the \(-\text{CH}_3\) protons of GMA and t-BA, respectively. The peaks at 2.08, 2.55, and 2.65 ppm are assigned to \(-\text{CH}\) and \(-\text{CH}_2\) protons in the main chain of \(\text{P}(\text{t-BA-co-GMA})\). The peak at 3.15 ppm is assigned to the \(-\text{CH}_2\) protons of the epoxy group. The peak at 3.91 ppm is attributed to \(-\text{CH}\) protons of the epoxy group, and the peak at 4.28 ppm is attributed to the \(-\text{CH}_2\text{OC}(==\text{O})\)–\(\text{C}\) protons in GMA.

As can be seen from Figure 2b, the peaks of the chemical shift at 17.24 and 27.89 ppm belong to the carbon atoms of \(-\text{CH}_3\) in GMA and t-BA, respectively. The peaks at 44.75 ppm are assigned to the carbon atoms of the main chain (\(-\text{C}==\text{C}==\text{CH}_2\)–\(\text{CH}_2\)–\(\text{CH}_3\)) in \(\text{P}(\text{t-BA-co-GMA})\). The peak at 48.72 ppm is assigned to the carbon atom of the epoxy group. The peak at 66.72 ppm is assigned to the carbon atom of \(-\text{O}==\text{CH}_2\) in GMA. The peak at 80.27 ppm is attributed to the carbon atom of \(-\text{O}==\text{C}==(\text{CH}_3)_2\) in GMA.
of FTIR, 1H-NMR, and 13C-NMR spectra, the P(t-BA-co-GMA) copolymer has been successfully synthesized.

The thermal stability of the P(t-BA-co-GMA) copolymer was determined by thermogravimetric−differential scanning calorimetry (TG-DSC), which is shown in Figure 3.

As can be seen from Figure 3, there exist two mass loss stages of P(t-BA-co-GMA). The first stage started from 227 °C and reached the maximum mass loss at 251 °C; the mass loss of this stage was 9.39%. The second stage with an 88.47% mass loss began at 346 °C and reached the maximum mass loss at 416 °C. There are two endothermic peaks in the DSC curve. The two peaks also correspond to the two mass loss stages of the TG curve. There is almost no mass loss below 200 °C. Therefore, the P(t-BA-co-GMA) copolymer possessed good thermal stability below 200 °C.

### 2.2. Optical Properties of P(t-BA-co-GMA) Structural Colors on Polyester Fabrics

The photographs of structural colors of P(t-BA-co-GMA) nanospheres on polyester fabrics with different mass ratios of t-BA and GMA are shown in Figure 4.

As can be seen from Figure 4, the structural colors of P(t-BA-co-GMA) nanospheres on polyester fabrics vary with the mass ratio of t-BA and GMA. The color of P(t-BA-co-GMA) nanospheres on polyester fabric was green when the mass ratio of t-BA and GMA was 1:1, and the color changed to purple when the mass ratio was 1.5:1. It changed to pink when the mass ratio increased to 2:1. The color turned to bluish violet when the mass ratio of t-BA and GMA was 2.5:1. The color of P(t-BA-co-GMA) nanospheres with a mass ratio of 4:1 was blue. Therefore, five different colors of P(t-BA-co-GMA) nanospheres on black polyester fabrics were obtained with different mass ratios of t-BA and GMA.

The reflectance spectra of P(t-BA-co-GMA) nanosphere structural colors on polyester fabrics with different mass ratios of t-BA and GMA are shown in Figure 5.

As can be seen from Figure 5, the reflectance curves of P(t-BA-co-GMA) nanospheres on polyester fabrics with different mass ratios of t-BA and GMA are significantly different. As for green color with a mass ratio 1:1, there exists a reflection peak at 510 nm with a reflectivity of 8.2%. When the mass ratio was 1.5:1 and for purple color, the reflectivity decreased from 400 to 550 nm, and then, it gradually increased after 550 nm. As for the pink color with a mass ratio of 2:1, the reflectivity decreased in the range of 400−520 nm, and then, it increased to 610 nm; after this, it decreased to the lowest value of 670 nm. For the bluish violet color with a mass ratio of 2.5:1, the reflection peak was at 410 nm. As for the blue color with a mass ratio of 4:1, the reflectance curves exhibit a peak at 450 nm. The different colors and reflectance curves of P(t-BA-co-GMA) nanospheres on polyester fabrics with different mass ratios of t-BA and GMA may arise from the different P(t-BA-co-GMA) nanospheres to form the different photonic bandgap.
gaps corresponding to different wavelengths of electromagnetic waves in the visible range.

2.3. Morphology and Particle Size Distributions of P(t-BA-co-GMA) Nanospheres. The scanning electron microscopy images of P(t-BA-co-GMA) nanospheres on polyester fabrics with different mass ratios of t-BA and GMA are shown in Figure 6.

As is shown in Figure 6, all the P(t-BA-co-GMA) nanospheres with different mass ratios of t-BA and GMA were successfully formed by closely packed coating on polyester fabrics. The P(t-BA-co-GMA) nanospheres can self-assemble into short-range ordered and long-range disordered amorphous colloidal crystals to present bright colors, which was also the origination of the vivid angle-independent coloration effect on polyester fabrics.

The particle size distributions of P(t-BA-co-GMA) nanospheres with different mass ratios of t-BA and GMA are shown in Figure 7.

As is shown in Figure 7, the peaks of the particle size distributions of P(t-BA-co-GMA) nanospheres with different mass ratios of t-BA and GMA are narrow and sharp, which indicates excellent monodispersity of P(t-BA-co-GMA) nanospheres. The diameters of P(t-BA-co-GMA) nanospheres vary with the different mass ratio of t-BA and GMA. The particle size of P(t-BA-co-GMA) nanospheres is the smallest (201.5 nm) when the mass ratio of t-BA and GMA is 4:1. As for the green color (with a mass ratio of 1:1), the particle size of nanospheres is 270.2 nm. The particle sizes of P(t-BA-co-GMA) nanospheres with other mass ratios are all over 300 nm.

The particle size reaches the largest value of 378.7 nm when the mass ratio of t-BA and GMA is 2.5:1. The different particle sizes of the P(t-BA-co-GMA) nanospheres with different mass ratios of t-BA and GMA may be the reason of different structural colors on polyester fabrics.

The mechanism of color formation of PCs could be explained by Bragg’s law. The maximum wavelengths ($\lambda_{\text{max}}$) of structural colors fit the Bragg’s equation:

$$m \lambda = 2nd \sin \theta$$

According to this equation, the $\lambda_{\text{max}}$ of the structural color is related to parameters of $n$ (average refractive index of the PCs), $d$ (the distance between the diffracting planes), and $\theta$ (the Bragg glancing angle between the incident light and the diffraction crystal planes). The one change of these parameters will result in different colors. There are two kinds of PCs: one is the conventional PCs (PCs) with both short-range and long-range order arrangements producing iridescent structural colors. The other one is amorphous PCs with short-range order and long-range disorder arrangements producing vivid angle-independent noniridescent structural colors. The P(t-BA-co-GMA) nanosphere structural colors belong to amorphous PCs, which are angle-independent. The structural colors of P(t-BA-co-GMA) nanospheres are related to the parameters of $n$ and $d$. The different P(t-BA-co-GMA) copolymers with different mass ratios of t-BA and GMA result in different $n$ values. Therefore, the different particle sizes of different P(t-BA-co-GMA) nanospheres and their different refractive indexes may be the causes of different colors.

2.4. Colorfastness of the Structural Colors on Polyester Fabrics. The colorfastness is one of the most important factors for the practical application of structural
colors in the textile field. The abrasion and washing colorfastness were investigated in this research. The P(t-BA-co-GMA) nanosphere structural colors on polyester fabrics with different mass ratios of t-BA and GMA before and after abrasion are shown in Figure 8.

![Figure 8](image)

**Figure 8.** Abrasion colorfastness of P(t-BA-co-GMA) nanosphere structural colors on polyester fabrics with different mass ratios of t-BA and GMA: (a) 1:1, (b) 1.5:1, (c) 2:1, (d) 2.5:1, and (e) 4:1.

As can be seen from Figure 8, the colors of P(t-BA-co-GMA) nanospheres on polyester fabrics with different mass ratios of t-BA and GMA before and after abrasion were almost the same. The reflectance spectra of P(t-BA-co-GMA) nanosphere structural colors after abrasion were also determined. The shape of the reflectance curves of all colors is similar to that before abrasion, and the reflection peaks of different reflectance curves are also similar to those before abrasion. The values of reflectivity at peaks of these structural colors after abrasion are also similar to those before abrasion, except for the green color, which was 0.32% lower than that before abrasion. These results show that P(t-BA-co-GMA) nanosphere structural colors on polyester fabrics possess good abrasion colorfastness.

The washing colorfastness of the structural colors on polyester fabrics is shown in Figure 9. As can be seen from Figure 9, the colors of P(t-BA-co-GMA) nanospheres on polyester fabrics with different mass ratios of t-BA and GMA slightly changed after washing. The shape and reflection peaks of the reflectance curves after washing are similar to those before washing. The values of reflectivity at peaks of these structural colors after washing slightly increase (less than 0.5%) compared with those before washing, while the pink color slightly reduced about 0.46% at the peak after washing. These results also demonstrate that the P(t-BA-co-GMA) nanosphere structural colors on black polyester fabrics have good washing colorfastness.

3. EXPERIMENTAL SECTION

3.1. Materials. Black polyester fabric with a density of 90 g/m² was purchased from Dongguan Humen Hengwei Cloth Shop (Guangdong, China). Glycidyl methacrylate (GMA, 97%), tert-butyl acrylate (t-BA), and sodium bicarbonate (AR, >99.8%) were purchased from Aladdin reagent (Shanghai) Co., Ltd. (Shanghai, China). Ammonium persulfate ((NH₄)₂S₂O₈, AR) was purchased from Wuxi Yasheng Chemical Co. Ltd. (Jiangsu, China).

3.2. Synthesis of P(t-BA-co-GMA). t-BA and GMA with different mass ratios and 0.05 g of sodium bicarbonate were evenly dispersed into a 150 mL round-bottom flask filled with 90 mL of deionized water. Then, N₂ gas was injected into the flask for 15 min. After this, the flask was put into a water bath of 80 °C, which was kept stirring for 30 min. Then, 0.15 mmol (NH₄)₂S₂O₈ was dissolved in 10 mL of deionized water, which was used as the initiator solution. After this, 10 mL of (NH₄)₂S₂O₈ solution was added into the above reaction flask using a syringe. The reaction was kept at 80 °C for 4 h. After this, the reaction solution was centrifuged at 11000 rpm for 40 min to obtain the solid product. The solid product was dried at 50 °C on a vacuum oven. The chemical structure of P(t-BA-co-GMA) is shown in Figure 10. The P(t-BA-co-GMA) copolymers with different masses and mass ratios of t-BA and GMA are shown in Table 1.

3.3. Preparation of Structural Color on Black Polyester Fabric. P(t-BA-co-GMA) copolymers (2 g) were added into a 50 mL conical flask with 20 mL of deionized water, which was magnetically stirred for 2 h to obtain the uniform nanosphere dispersion. The black polyester fabric with a diameter of 8 cm was placed on a sealed foamed plastic box with a 10 mL nanosphere dispersion was added into a WH-2000 ultrasonic nebulizer (Guangdong Yuehua Medical Device Factory Co. Ltd., China), which was sprayed out from the nozzle to form the atomization and deposit on the polyester fabrics. Then, the fabrics were dried with an oven at 40 °C for 10 min, and then, the structural colored fabrics were obtained.

3.4. Characterization. Fourier transform infrared spectroscopy (FTIR) and nuclear magnetic resonance (NMR) were used to characterize the chemical structure of the P(t-BA-co-GMA) copolymer. FTIR was conducted using an IRPrestige-21 (Shimadzu, JAPAN) FTIR spectrometer with KBr pellets under an air atmosphere. 1H-NMR was conducted with an Avance III HD spectrometer (Bruker, Germany) at room temperature using o-dichlorobenzene as the solvent. 13C NMR was conducted using solid-state nuclear magnetic resonance spectra, which were recorded on a Bruker 400 M spectrometer at resonance frequencies of 100.63 MHz using a 4 mm magic angle spinning (MAS) probe. The spinning rate and delay time were set to 10 kHz for the MAS and 2 s. The simultaneous TG analysis and DSC were performed with a NETZSCH STA 449F3 (NETZSCH, Germany) under a nitrogen atmosphere with a heating rate of 10 °C/min in the range of room temperature and 800 °C.

The particle size and distribution of P(t-BA-co-GMA) nanospheres were tested with the 0.01 g/L monodisperse
nanosphere dispersion using a Malvern nanometer particle size analyzer with a high sensitivity (Zetasizer Nano S90, Malvern Instrument Co. Ltd., UK). The morphology of structural colors on black polyester fabrics was viewed by using a Hitachi S-4800 scanning electron microscope. The reflectance spectra of structural colorized polyester fabrics were obtained using a Datacolor 650 spectrophotometer (Datacolor, USA) in the test range of 400–700 nm.

The abrasion colorfastness of the structural colors on polyester fabrics was determined by the sandpaper abrasion test method according to our previous research. The testing samples were placed face-up on sandpaper, and the black polyester fabrics were placed upon the colorized fabrics weighting 50 g by moving for 5 cm, and the changes of color and reflectance were used to evaluate the abrasion colorfastness. The structural colorized polyester fabrics were put into a 250 mL conical flask with 150 mL of deionized water, which was kept in a 40 °C constant temperature oscillating water bath for 2 h. After this, the samples were dried at 40 °C, and the washing fastness of the samples was determined by the changes of color and reflectance of structural colors.

4. CONCLUSIONS

In this study, the P(t-BA-co-GMA) copolymers containing reactive epoxy groups with different mass ratios of t-BA and GMA were successfully synthesized. P(t-BA-co-GMA) nanospheres can self-assemble into short-range ordered and long-range disordered PCs to present different bright crack-free structural colors on polyester fabrics by spray coating. The P(t-BA-co-GMA) nanospheres possess excellent monodispersity, and their particle sizes vary with the different mass ratio of t-BA and GMA to obtain five different colors on black polyester fabrics. The different particle sizes of the different P(t-BA-co-GMA) nanospheres with different refractive indexes and the arrangement of short-range ordered and long-range disordered in microstructures may be the reason of different angle-independent structural colors on polyester fabrics. The P(t-BA-co-GMA) nanosphere structural colors on polyester fabrics possess good abrasion and washing colorfastness. This research provides the experimental basis for the development of polymer PCs containing reactive groups and structural colorization on fabrics with high colorfastness to promote the practical application of structural color in textile coloration.

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Notes
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