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Изменения цветности неорганических пигментов традиционной китайской живописи под воздействием узких спектральных линий четырех хроматических компонент светодиодов белого цвета

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Музейное освещение предметов традиционной китайской живописи, весьма чувствительной к воздействию света, приводит к серьезным нарушениям их цветовой гаммы. Вследствие метамерии зрения и возможности управления интенсивностями излучения отдельных спектральных компонент светодиодов белого цвета возможна подстройка результирующего спектра их свечения под различные требования. Необходимо изучение воздействия различных монохроматических составляющих излучения белых светодиодов и определение правильных пропорций их интенсивностей, при которых вредное воздействие освещения на экспонаты минимизируется. Раздельно рассматривалось воздействие излучения четырех цветов (красный, желтый, зеленый и синий) на типичные неорганические красители. Измерялись координаты цвета в системе CIE XYZ и такие характеристики как доминирующая длина волны, фотометрическая яркость и условная чистота цвета различных пигментов при их освещении. Построены кривые изменения указанных параметров по 16 циклам испытаний и выявлены закономерности изменения цветности указанных комбинаций освещения и цветов пигментов. На основе этого путем расчета и сравнения изменений параметров цветности получены количественные данные о выцветании объектов живописи под воздействием указанных четырех хроматических компонент. Результаты предоставляют базовые данные для дальнейших исследований изменения и восприятия цветов объектов китайской традиционной живописи под действием музейного освещения.

Ключевые слова: *традиционная китайская живопись, изменение цвета, узкополосные спектры, белые светодиоды, музейное освещение.*

Chromaticity changes of inorganic pigments in traditional Chinese paintings due to narrowband spectra in four-primary white light-emitting-diodes

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Spectral power distributions of white light-emitting-diodes can be adjusted according to various demands. Thus, it is meaningful to explore the influence law of monochromatic lights mainly constructing white light-emitting-diodes. Four kinds of typical red, yellow, green, and blue monochromatic lights were used to illuminate typical inorganic pigments. CIE XYZ coordinates were examined periodically, and then the color

parameters — dominant wavelength, luminance, and excited purity, of the pigments were calculated through the data. The changing laws of colors of the five pigments under different monochromatic lights were obtained. The relative influence values of the four monochromatic lights were also acquired through calculating and comparing the changing degree of the color parameters. The achievements can provide data basis and reference for research on traditional Chinese paintings pertaining to color damage and rendering effect of the illumination.

Keywords: *Traditional Chinese paintings, color change, narrowband spectra, white light-emitting-diodes, museum lighting.*

OCIS codes: *140.3330, 150.2950, 230.3670, 010.1690*

INTRODUCTION

Traditional Chinese paintings (TCPs), which characterize huge storage and high value, suffer severe damage, like fading, color distortion, and color vanishing, due to radiation in museum illumination [1–5]. Controlling the spectral power distributions of light sources is the key to protect the paintings. Thus, it is significant to find a suitable method for evaluating the degree of color damage and choose the lowest damage light sources for different kinds of paintings.

For evaluating visible damage to paintings, color differences are generally used by researchers over the decades [3, 6–12]. Color differences can effectively express color changes of the pigments in paintings, and thus present damage extent of paintings under illumination. However, the change of color has three dimensions (hue, chroma, and value). An overall index of color difference fails to give details of the color change. Thus, specific information of color change, including hue (indicated by the main wavelength), chroma (the excited purity) and value (illuminance), is needed to show how the color exactly turns into, bluer or redder, more saturated or desaturated, brighter or darker?

A long-term illumination was conducted on traditional specimens of TCPs painted with inorganic pigments by typical light sources in museums-tungsten halogen, metal halide, and white light-emitting-diodes (WLED) [6]. The dominant wavelength, luminance, and excited purity were calculated by periodically tested color coordinates of pigments illuminated by various light sources. The influencing law of different pigments was obtained under various light sources and the influencing proportion of the tungsten halogen, metal halide, and WLED was respectively evaluated by different parameters: for the dominant wavelength, the ratio is 9.98:9.04:9.91, for the luminance, the ratio is 8.65:8.69:8.62, as for the excited purity, that is 8.54:9.05:8.68%. The WLED performed an overall low influence, however, the advantage was tiny.

Fortunately, we realized that WLEDs possess easy constructed spectrum which can be adjusted according to demands [13–16]. To date, the WLEDs satisfying low correlated color temperatures and high color rendering [5], are mainly composed of red, yellow, green, and blue wavebands [17–19], by adjusting the proportion of which can achieve the requirements; and the corresponding WLED is the namely red/yellow/green/blue (RYGB) four-primary WLED. Moreover, the four-primary mixture is frequently used to develop spectra of WLEDs achieving various functions [20, 21], *e.g.*, Ji Hye Oh et al. [22] developed a new type WLED according to the needs of circadian, color quality, and visual performance by the four-primary mixture. Accordingly, the quantitative damage law of the four wavebands plays an important role in manufacturing ideal light sources to illuminate TCPs.

Here, four typical monochromatic lights – blue (peak wavelength of 450 nm), green (510 nm), yellow (583 nm), and red (650 nm), were separately used to illuminate specimens of TCPs painted with classical inorganic pigments with color in cyan, yellow, red, white, and black. CIE XYZ coordinates of the pigments were periodically tested and transformed into parameters of the dominant wavelength, luminance, and excited purity to evaluate the specific color change. Changing curves of the three parameters varied with the exposure time and the quantified influential law was displayed to demonstrate the detailed influence. The technical roadmap is shown in Fig. 1.

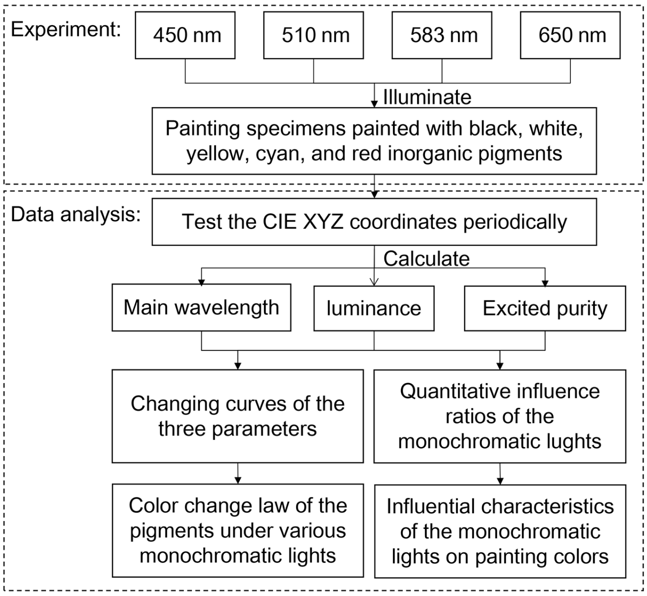

Fig. 1. The technical road map.

2. METHOD

2.1. Model of specimen

Four groups of specimens of the TCPs painted with inorganic pigments (iop-TCPs) were crafted by the traditional Chinese Painting Institute of the Tianjin University using traditional techniques and materials: firstly, the most typical inorganic pigments — ancient graphite (black in color), clam shell powder (white), azurite (cyan), cinnabar (red), and orpiment (yellow), were dissolved into water with the proportion of 1:1. Secondly, pigments were evenly sprayed on separate rice paper, which was then manually framed with wheat starch paste using traditional techniques to imitate the real iop-TCPs. Thirdly, five rice paper separately painted with five inorganic pigments were cut and then recombined to obtain four specimens, each containing the five pigments with the same size (4 cm by 4 cm). One of four specimens is presented in Fig. 2.

2.2. Experimental light sources

Four monochromatic lights with the peak wavelengths of 450, 510, 583, and 650 nm were produced by museum tungsten halogen lamp (PHILIPS 6423F0) cooperating with infrared cut-off filters and 20 nm narrow band-pass filters which peak at 450, 510, 583, and 650 nm (Specialized from Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences). To ensure the precision of the experiment, all of the spectra of monochromatic lights were detected via a spectrophotometer (Photo Research PR670) to determine their spectral irradiance distributions on the surfaces of the specimens (Fig. 3): the irradiance of each light source was kept the same and constant during the long-term experiment. We examined the irradiances of the light sources in every test cycles and changed the light source immediately once light fade occurred.

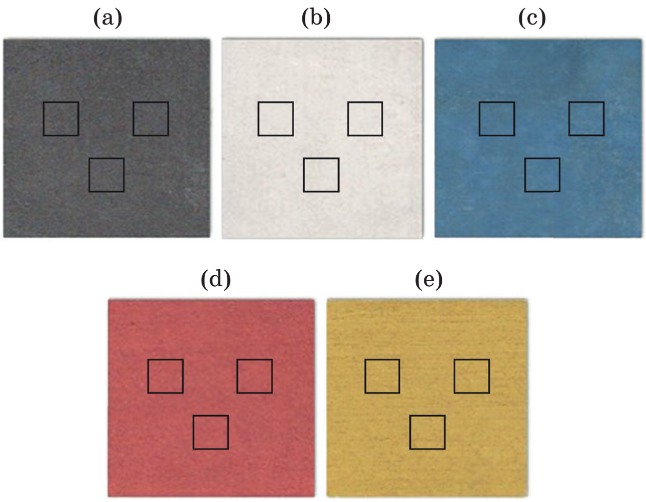

Fig. 2. One of four specimens with pigments (a) ancient graphite, (b) clam shell powder, (c) azurite, (d) cinnabar, (e) orpiment.

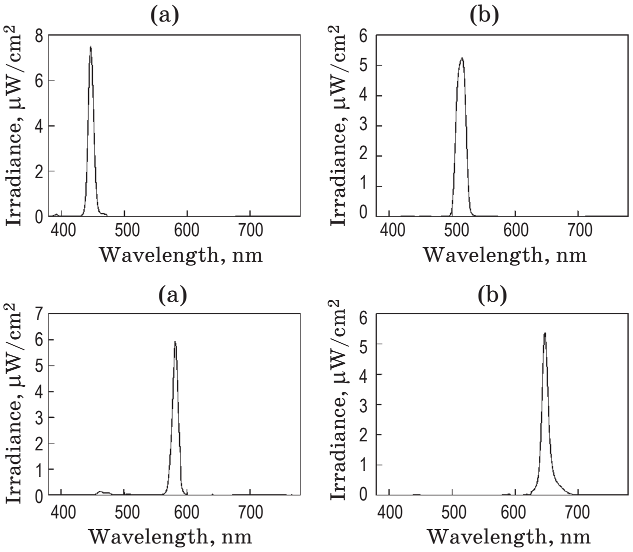

Fig. 3. Irradiance distributions of four monochromatic lights on the surface of the specimens (a) 450, (b) 510, (c) 583, (d) 650 nm.

In addition, color parameters in all cycles were tested under the D65 light source (OSRAM, L30W/965, 6500 ± 200 K) calibrated in National Institute of Metrology of China to conduct the color measurement.

2.3. Experimental scheme

The experiment was conducted in the Optical Laboratory of Tianjin University, the physical environment of which was kept at the required degree for preserving TCPs in the museum [23]: the whole laboratory maintained the temperature of 23 ± 0.5°, the humidity of 50% and the exchange rate of 0.5 per day.

Four monochromatic lights were installed on the upper part of the experimental bench, and four identical specimens were simultaneously put under the corresponding light sources. Irradiances on the surfaces of the four specimens were kept the same at 1 W/m^2 by adjusting the output power of the light sources. Four groups were kept independent by shade to avoid mutual interference.

Periodical illuminations on specimens were carried out, one illumination cycle contained six days with twelve hours in each, the experiment lasted sixteen cycles for 1152 illumination hours in total. Exposure of specimens accumulated with the increasing illumination hours. Experimental scheme is illustrated in Fig. 4.

After each cycle of illumination, the specimens were moved under the D65 standard light source; and color parameters of the specimens were measured by the standard test method of CIE (Commission International de l'Eclairage) [24] (Fig. 5).

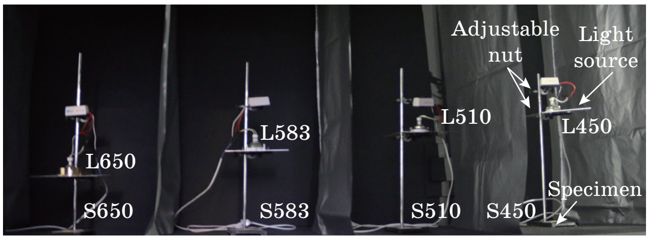

Fig. 4. Experimental device in the optical laboratory with constant physical environment. L represents the monochromatic light source, S represents specimen.

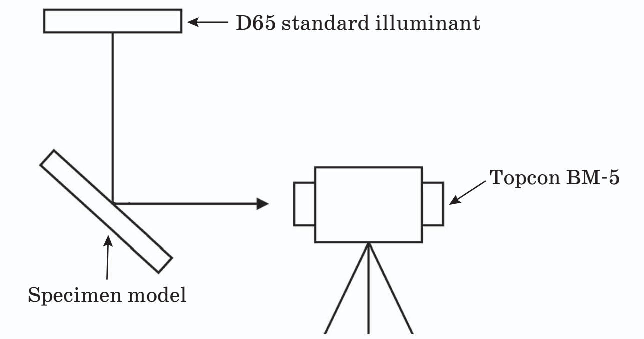

Fig. 5. Diagrammatic sketch of the test environment to measure the color parameters of the pigments by Luminance colorimeter (Topcon BM-5) under D65 standard illuminant.

The CIE chromaticity coordinates (*x*, *y*) and metric brightness values Y of four specimens were measured before and each cycle after the illumination, at test points marked in Fig. 2, achieved by Luminance Colorimeter (Topcon BM-5, with an accuracy of ±3%).

3. RESULTS

3.1. Changing curves of the color parameters

The dominant wavelengths and excited purity values of the pigments were calculated based on the (*x*, *y*), Y values. Changing curves of the dominant wavelengths, luminance, and excited purities of the 16 experimental cycles were plotted.

3.1.1. The dominant wavelength

Black (the ancient graphite) and white (clam shell powder) are achromatic colors without the dominant wavelengths, so the changes of dominant wavelengths of cyan (azurite), red (cinnabar), and yellow (orpiment) under various light sources were considered. Because of the visual characteristics of the human eyes and the nonuniformity of the CIE XYZ chromaticity diagram, the changing of the dominant wavelengths are nonuniform. For expressing the realistic changes of the dominant wavelengths in the changing curves, the vertical coordinates were made uneven as the CIE XYZ chromaticity diagram depicted. The changing curves of the dominant wavelengths of the azurite, cinnabar, and orpiment were illustrated in Fig. 6.

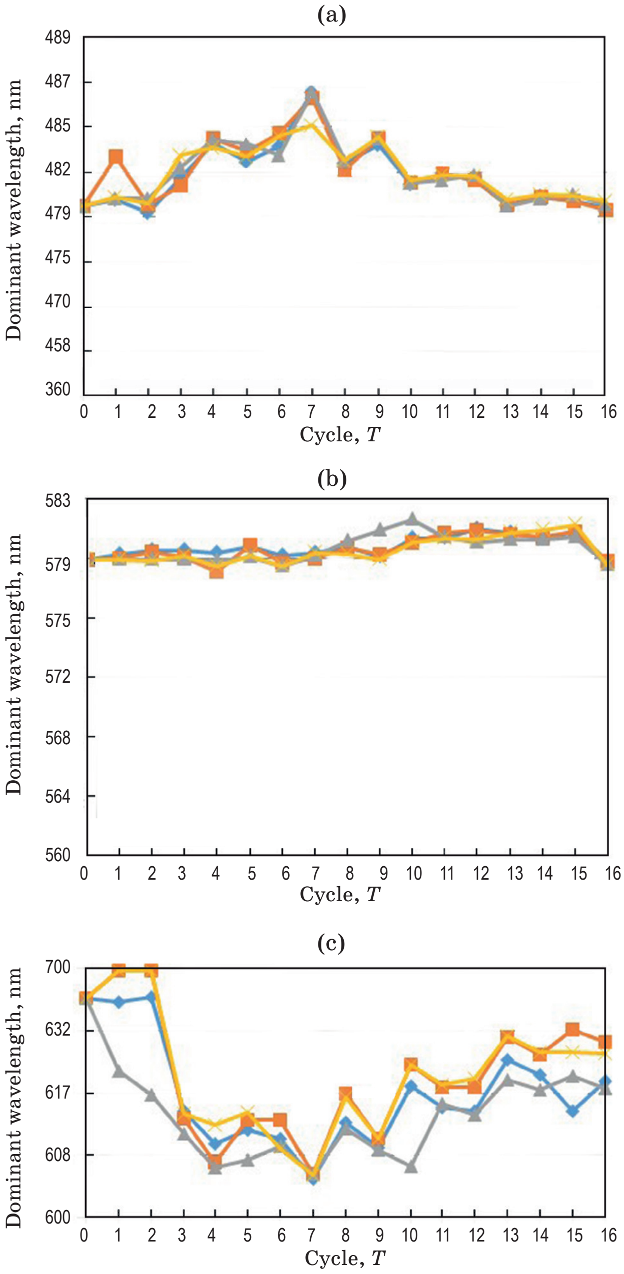

Fig. 6. Changing curves of the dominant wavelengths of (a) cyan (azurite), (b) yellow (orpiment), (c) red (cinnabar) under the illumination of the four monochromatic lights.

3.1.2. The excited purity

The changing curves of the excited purities of the azurite, cinnabar, and orpiment were demonstrated in Fig. 7.

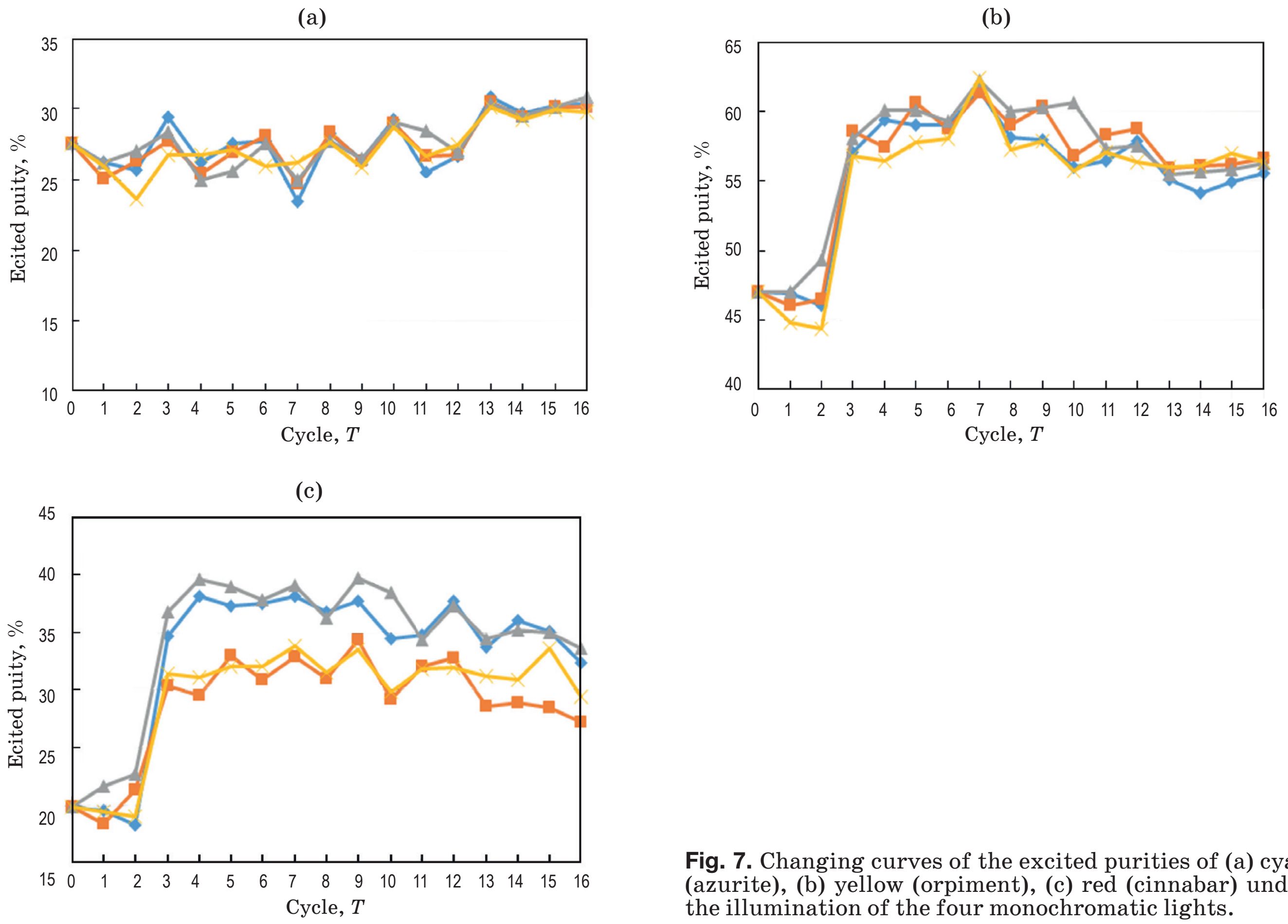

Fig. 7. Changing curves of the excited purities of (a) cyan (azurite), (b) yellow (orpiment), (c) red (cinnabar) under the illumination of the four monochromatic lights.

3.1.3. The luminance

The changing curves of the luminance of the ancient graphite, clam shell powder, azurite, cinnabar, and orpiment were plotted in Fig. 8.

3.2. Influence degrees of the monochromatic lights

The differences of the luminance, the dominant wavelengths, and the excited purities of the five inorganic pigments under the illumination of the four monochromatic lights were calculated after each cycle to compare with the initial values. And the average influencing values of the monochromatic lights in the three aspects were calculated to evaluate the comprehensive influencing degrees of the monochromatic lights on the colors of TCPs which can be basically represented by the colors of the five pigments. Calculating method is expressed by Eq. (1). The average results of the dominant wavelengths of the 16 cycles are shown in Table 1, the results of the excited purities are demonstrated in Table 2, for the luminance, in Table 3.

$$\begin{aligned}
\overline{\lambda} &= \frac{1}{16}\left(|\lambda_1-\lambda_0|+|\lambda_2-\lambda_0|+\dots+|\lambda_{16}-\lambda_0|\right),\\
\overline{Y} &= \frac{1}{16}\left(|Y_1-Y_0|+|Y_2-Y_0|+\dots+|Y_{16}-Y_0|\right), \qquad \textbf{(1)}\\
\overline{P_e} &= \frac{1}{16}\left(|P_{e1}-P_{e0}|+|P_{e2}-P_{e0}|+\dots+|P_{e16}-P_{e0}|\right).
\end{aligned}$$

Table 1. Average influences of the monochromatic lights on the dominant wavelengths of pigments

Wavelength, nm	Pigments			
	Cyan (azurite)	Yellow (orpiment)	Red (cinnabar)	Average
450	1.4324	0.4463	3.2627	1.7138
510	1.5850	0.4312	2.8117	1.6093
583	1.5418	0.4345	2.8366	1.6043
650	1.5218	0.3911	2.8136	1.5755

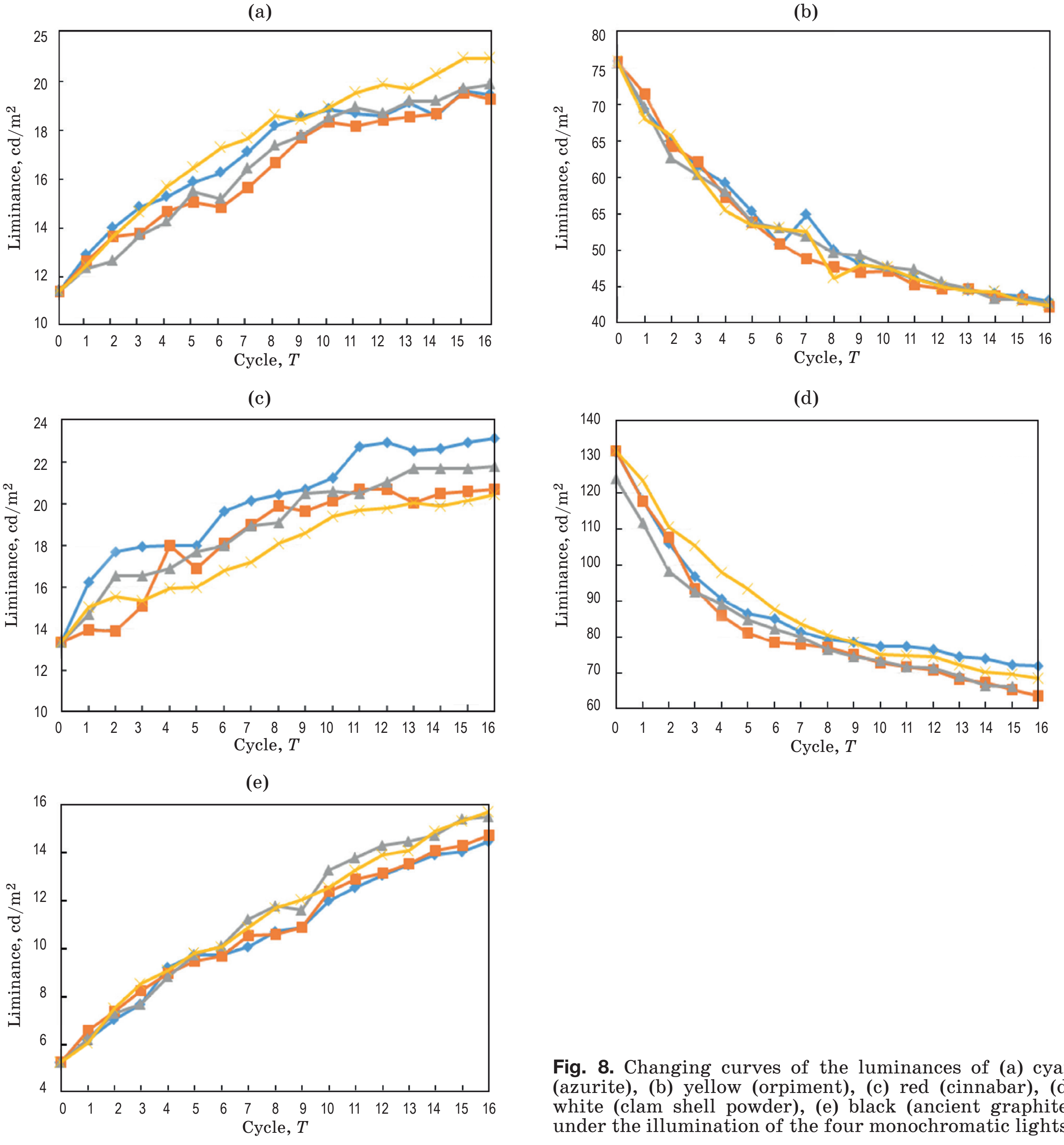

Fig. 8. Changing curves of the luminances of (a) cyan (azurite), (b) yellow (orpiment), (c) red (cinnabar), (d) white (clam shell powder), (e) black (ancient graphite) under the illumination of the four monochromatic lights.

Table 2. Average influences of the monochromatic lights on the excited purities

Wavelength, nm	Pigments			
	Cyan, %	Yellow, %	Red, %	Average, %
450	1.75	9.06	14.25	8.35
510	1.58	9.86	12.65	8.03
583	1.55	10.16	12.20	7.97
650	1.47	9.21	10.48	7.05

Table 3. Average influences of the monochromatic lights on the luminances

Wavelength, nm	Pigments					
	Cyan	Yellow	Red	White	Black	Average
450	5.8703	24.0637	7.0682	52.2831	5.6582	18.9887
510	5.2256	24.8703	5.2539	51.7373	5.8268	18.5828
583	5.4545	24.3708	5.8810	48.2200	6.3622	18.0577
650	6.4467	24.8312	4.6353	45.9954	6.3315	17.6480

4. DISCUSSION

4.1. Changing characteristics of the colors

(1) Cyan (azurite): we can see from Fig. 6a that the dominant wavelength of the azurite performs an overall stability with slight fluctuation, that indicates a minor change of the hue. As Fig. 7a shows, the excited purity changes a little, only increases slightly in the thirteenth period, that indicates a small increase of the saturation of cyan. In Fig. 8a, the luminance shows a linear increasing trend, that means, cyan has an apparent fading. The chemical composition of azurite is $2CuCO_3{\cdot}Cu(OH)_2$, which will decompose into CuO, H_2O, CO_2 in the air after suffering the illumination, and then occurs fading and discoloration due to the oxidation reaction.

(2) Yellow (orpiment): in Fig. 6b, the hue of the orpiment almost performs no change as the curves of the dominant wavelength of yellow are nearly straight during the whole experiment. The excited purity begins to raise from the first cycle and turn relatively smooth when the third cycle occurs, which tells us that the saturation of yellow increases after 72 hours of illumination, and the trend tends to be stable after being illuminated for 216 hours, as the Fig. 7b depicted. We can see from the Fig. 8b, that the luminance keeps decreasing dramatically after being illuminated, shows that the orpiment occurs an apparent deepening. The orpiment performs an overall higher discoloration than the azurite for the main component of the orpiment is As_2S_3, containing chemically unstable Fe_2S_3, which turns into yellow-green easily in normal temperature, and the illumination greatly enhances the reaction to change the saturation and luminance of the pigment.

(3) Red (cinnabar): the dominant wavelength begins to dramatically decrease from the second cycle to the fourth cycle. In the following cycles, the vertical values of the starting points and the ending points of the curves in Fig. 6c do not change a lot. But there are fluctuations in the curves, indicating that the hue changes from red to orange after the illumination of 144 hours. In Fig. 7c, the excited purity increases a lot from the second to the third cycle, turns into stabilization after that, which means the saturation of the red increases after 144 hours and tends to be stable after 216 hours. Figure 8c shows, that luminance keeps increasing during the whole experiment, representing the fading of red. The degree of discoloration of the cinnabar is higher than that of the azurite and lower than that of the orpiment, because the main component of the cinnabar is α-HgS, the chemical stability of which is between $2CuCO_3{\cdot}Cu(OH)_2$ and Fe_2S_3, and α-HgS easly changes into β-HgS under the influence of photochemical reaction to result in discoloration and fading.

(4) White (clam shell powder): as Fig. 8d shows, the luminance of the clam shell powder decreases dramatically during all the cycles, the extent of which is the largest in the five pigments, the clam shell powder has an extraordinary darken phenomenon. In theory, white can reflect lights with all the wavelengths, may absorb the least energy when the incident light is the same and suffers the least influence. However, the experiment gives an inverse result. The reason for the phenomenon is that the chemical property of the clam shell powder is rather unstable. $CaCO_3$ and $Ca_3(PO_4)_2$ mainly construct the clam shell powder. In them, $CaCO_3$ can easily decompose into CaO and CO_2 after suffering heat and reacting with the acid to generate saline content and CO_2, when deposited in the air for a long term. And the light and heat can aggregate the chemical reaction. In addition, $Ca_3(PO_4)_2$ reacts with the acid in the air producing phosphate to cause the deepening.

(5) Black (ancient graphite): in Fig. 8e, the luminance of the ancient graphite gradually increases, indicating the experience of fading and whitening, the degree of which is the most minimum in the five pigments. Theoretically, the black absorbs the photon energy of all the monochromatic lights. Compared to other colors, black should be the hugest influenced. But that is not so. The main reason for the minus change of the luminance is the organic hydrocarbon compound carbon, which is the basis of the ancient graphite, forming polyhedral carbon particles after suffering pyrolysis. The physical and chemical properties of polyhedral carbon particles are extremely stable, they can resist light, heat and acid-base. Thus, light damages the ancient graphite with the minimum extent.

4.2. Influencing degrees of the monochromatic lights

As Tables 1–3 demonstrate, the radiations of the monochromatic lights affect the most on the luminance, and the least on the dominant wavelength, which manifests that luminance change is the main reason for the color change of the iop-TCPs. Among the pigments, the luminances of black, cyan, and red, which are dark colors, increase indicating the fading, while white and yellow, which are light colors, decrease indicating the deepening due to the oxidation reaction.

We can see from the Tables 1–3, the average relative influencing values of the monochromatic lights (450, 510, 583, 650 nm) on the dominant wavelengths of the pigments are 1.7138, 1.6093, 1.6043, 1.5755 respectively; on the luminances the values are 18.9887, 18.5828, 18.0577, 17.6480; and for the excited purities, the values are 8.35, 8.03, 7.97, and 7.05%. Accordingly, the monochromatic lights with short wavelengths influence more on the dominant wavelengths, luminances, and excited purities of the pigments. According to the Plank formula $E = h\nu$, in which E represents the photon energy, h is Planck's constant, ν is the frequency of the photon, short wavelengths possess high frequency, and there is an inverse ratio between the wavelength and energy. Thus, the monochromatic lights with short wavelengths have larger energy and hence influence more on the pigments. The theory analysis corresponds to the experimental results.

5. CONCLUSION

The exposure, the photochemistry properties of the pigments, and the spectra of the light sources are the main components of illumination causing the color decay. In the experiment of illuminating inorganic pigments in traditional Chinese paintings by monochromatic lights, the exposure of the specimens was maintained the same, thus the factors that cause the color changes are the photochemistry properties of the pigments and the spectra of the light sources. According to the analysis of the experiment data, conclusions about the influential ways of the above factors are as follows:

(1) The photochemical properties contribute more to the color change. For example, white (clam shell powder) suffers a more apparent discoloration under the illumination of the four monochromatic lights because of the poor photochemical stability, although it absorbs the least photon energy. While black (ancient graphite) changes feebly because of the extreme chemical stability, though it absorbs the energy of all the wavebands. However, the light sensibility of specific pigments is their inherent attribute which is immutable.

(2) Different monochromatic lights influence differently on different pigments and generally, the monochromatic lights with shorter wavelengths perform a larger influence. Thus, when manufacturing the lowest damage WLEDs illuminating iop-TCPs, short waveband should be eliminated as much as possible. Because of the various construction of inorganic pigments in the three kinds of traditional Chinese heavy color paintings — meticulous heavy color painting, jinbi heavy color painting, turquoise heavy color painting, WLED spectra suitable for different kinds of inorganic pigment painted paintings can be developed according to the experimental results demonstrated in Tables 1–3. For example, the meticulous heavy color painting is colorfully painted with all typical inorganic pigments — ancient graphite (black), clam shell powder (white), azurite (cyan), cinnabar (red), and orpiment (yellow). When developing the WLED for this kind of painting, the comprehensive influence of the illumination on the five inorganic pigments should be considered. The result turns out to be that the average relative influence values of the 450, 510, 583, 650 nm monochromatic lights on the dominant wavelength of the meticulous heavy color painting are 1.7138, 1.6093, 1.6043, 1.5755 respectively; on the luminance, the values are 18.9887, 18.5828, 18.0577, 17.6480; and for the excited purity, the values are 8.35, 8.03, 7.97, and 7.05%.

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