

Article

Study on the Color-Influencing Factors of Blue Iolite

Xin Liu and Ying Guo * 

School of Gemmology, China University of Geosciences, Beijing 100083, China

* Correspondence: guoying@cugb.edu.cn

Abstract: This article explores the factors that influence the color of blue iolite, which is the gem-quality variety of the cordierite mineral. The X-Rite SP62 portable spectrophotometer was used to measure color in the CIELAB color space. ED-XRF and UV-vis were used for analysis. The results show that blue iolite contains the chromophore elements Fe and Mn, but the effect on the color of iolite is not significant due to the low Mn content. The lightness L^* and the hue angle h° are mostly determined by its Fe content. UV-vis spectra show that iolite has a broad absorption band near 570 nm caused by the charge transfer between Fe^{2+} on the octahedron and Fe^{3+} on the T_1 1 tetrahedron and results in the color of blue iolite. Each different standard light source has different degrees of influence on the color parameters of iolite, and the hue angle h° is the most influenced.

Keywords: blue iolite; CIE LAB color space; ED-XRF; UV-vis spectroscopy; standard light source



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1. Introduction

Cordierite is a mineral with a long history. More than a thousand years ago, the Norse used the strong polarization effect of cordierite to determine the position of the sun during cloudy days. Cordierite is a typical metamorphic mineral, formed after moderate to high thermal metamorphism, mainly in gneiss, schist with a high aluminum content, and altered igneous rocks. It can also be found in some granites. In regional metamorphic rocks, granulite and gneisses, cordierite coexists with other Mg-Fe minerals, such as garnet, biotite, orthopyroxene, spinel or staurolite. It can also coexist with aluminum-rich minerals, such as sillimanite, sapphire and corundum [1].

Cordierite is used as a gemstone, which is known as “iolite” in the gem and jewelry industry. It is also known as “water sapphire” because of its sapphire-like color and is widely used in industry for its excellent properties, but gem-quality cordierite is also considered to be a rare gemstone variety. Worldwide, gem-quality cordierite is mostly found in India, Sri Lanka, Madagascar, the United States (California, Idaho, and Wyoming), Canada, and Greenland [2]. Among them, India is an important market for iolite gem trading. Iolite occurrences have been reported from high-grade metamorphic rocks of the Eastern Ghat Mobile Belt of the states of Odisha and Andhra Pradesh and in metamorphic rocks of Karnataka and Tamil Nadu [3].

Iolite is highly transparent, usually a blue to blue-violet color, with strong pleochroism visible to the naked eye: blue-violet iolite shows light purple, dark purple and yellow-brown pleochroism; blue iolite shows colorless to yellow, blue-gray and dark purple pleochroism. According to previous studies on the spectrum and color properties, the cause of the color can be attributed to the charge transfer between Fe^{2+} on the octahedron and Fe^{3+} in the edge-shared T_1 1 tetrahedra in the iolite [4–6].

In addition, some external factors such as light source and background will also affect the appearance of the gemstone color. For example, color-changing garnets can change from red to green under different color temperature light source conditions. According to the national standard (GB/T 20146-2006) and the requirements of jewelry quality evaluation as well as the uniformity of lighting sources, D65 (6500 k), A (2856 k) and F2 (4150 k) were

selected to explore the influence of different standard light sources on the color appearance of blue iolite in this paper.

The CIE 1976 $L^*a^*b^*$ uniform color system is the most popular system for color measurement and analysis, currently recommended by the International Commission on Illumination (CIE) [7–10]. The color space has good color uniformity, so that the visual distance between colors is proportional to the Euclidean distance between color coordinates, and is fully consistent with the subjective law that the visual color difference in the red and green directions is less than the visual color difference in the yellow and blue directions [11]. The system consists of color coordinates a^* and b^* and the lightness L^* ; the chroma C^* and the hue angle h° can be calculated from a^* and b^* :

$$C^* = [(a^*)^2 + (b^*)^2]^{1/2} \quad (1)$$

$$h^\circ = \arctan(b^*/a^*) \quad (2)$$

In recent years, colorimetry has played an important role in gemology, mainly involving materials such as jadeite [12–14], diamond [15], sapphire [16], peridot [17,18], amethyst [19], tourmaline [20,21], chrysoberyl [22], color-changing garnet [23–26], amber [27] and cubic zirconia. However, evaluation and analysis of factors influencing iolite color have been scarce so far. In this paper, the color of iolite is quantitatively characterized using the CIE 1976 $L^*a^*b^*$ uniform color space, and the effects of transition elements, by means of UV–vis spectroscopy, and light source changes on the color are discussed.

2. Materials and Methods

2.1. Materials

Twenty gem-quality natural iolite oval faceted, with a uniform color transition from light blue to blue–violet were selected for this work. No inclusion in the inner part of the sample as seen by the naked eye. The gemological properties of iolite are summarized in Table 1 and some of the samples are shown in Figure 1.

Table 1. Gemological properties of iolite samples.

Properties	Results
Color	Light blue to blue–violet
Weight	0.553–0.811 ct
Refractive indices	1.532–1.551
Specific gravity	2.409–2.596
Pleochroism	Dichroism visible to the naked eye
	Strong to medium blue or blue–violet; colorless to yellow
Microscopic inclusions	Needle-like or columnar gas–liquid inclusions
	Fingerprint-like inclusions

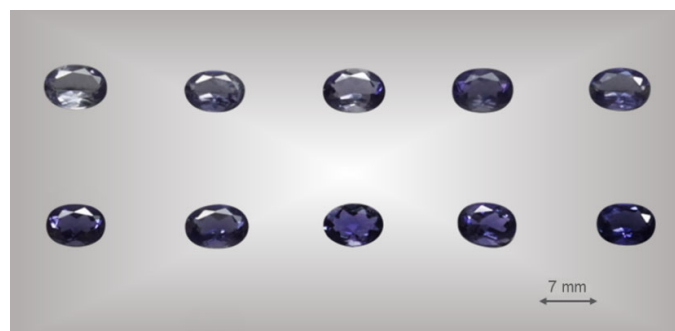


Figure 1. Some of the samples used in this study under standard D65 light. All samples are taken from the tabletop.

2.2. Methods

2.2.1. Colorimetric Analysis

The color parameters of iolite were determined by an X-Rite SP62 portable spectrophotometer (X-Rite, Grand Rapids, MI, USA). Based on the CIE 1976 $L^*a^*b^*$ uniform color space, an integrating sphere is used to collect the reflected signal from the sample surface. The test conditions are as follows: reflection, excluding specular reflection; D65 (6504 K) standard light source illumination; 10° observer field of view; measurement range, 400–700 nm. The final color data were averaged over three tests; the test area for a single sample is a circle with a diameter of 6 mm.

2.2.2. ED-XRF

Semi-quantitative elemental testing was carried out using an EDX-7000 energy dispersive X-ray fluorescence spectrometer (SHIMADZU, Kyoto, Japan). The test conditions are as follows: atmosphere, vacuum; electron beam energy 2.2 keV; X-ray energy range 3.5–35 keV; detector, Si (Li); collimator, 1 mm.

2.2.3. UV-Vis Spectroscopy

Absorption spectra in the UV–vis range were recorded using a PE-Lambda 950 UV–vis spectrophotometer (PERKINELMER, Waltham, Massachusetts, USA). The test conditions are as follows: transmission method; measurement range 300–800 nm; data interval 1 nm; scan speed 266.75 nm/min; cycles 1 s; detector settings 0.2 s.

3. Results

3.1. Color Quantification

The color of 20 iolites was quantitatively characterized using a X-Rite SP62 portable spectrophotometer based on the CIE 1976 $L^*a^*b^*$ uniform color space with a Munsell N9 color card as the background under a standard D65 light source. As shown in Figure 2, the color of all samples was measured with the tabletop up vertically in this paper, and the overall color parameters of iolite are obtained after taking the average value three times. Results show the color range of iolite: the lightness $L^* \in (29.47, 67.84)$, color coordinate $a^* \in (3.00, 7.97)$, color coordinate $b^* \in (-20.25, -6.87)$, the chroma $C^* \in (7.60, 21.72)$ and the hue angle $h^\circ \in (287.17, 295.25)$.

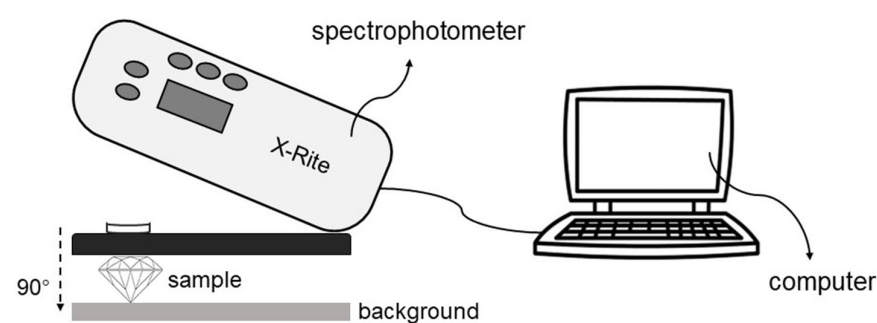


Figure 2. The color measurement device for this experiment.

Through the correlation analysis of the color parameters of 20 iolites, it is found that there is no obvious good correlation between the color coordinates a^* and b^* and the hue angle h° , indicating that the color coordinates a^* and b^* are not the main factors affecting the change of the hue h° of iolite. This paper focuses on the correlation between color coordinates a^* and b^* and the chroma C^* . After linear fitting, the relationship is as follows (Figure 3):

$$C^* = 2.415a^* + 1.906 \quad (R^2 = 0.957)$$

$$C^* = -1.090b^* - 0.175 \quad (R^2 = 0.999)$$

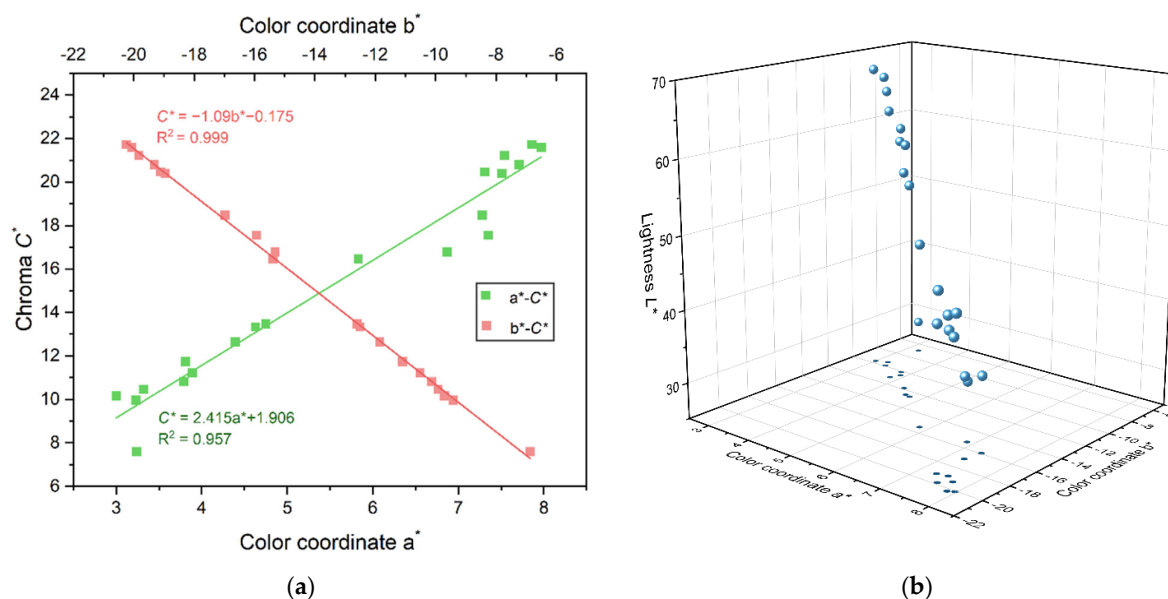


Figure 3. (a) Relationship between color coordinates a^* and b^* and the chroma C^* . (b) A total of 20 iolite blue plots in the CIE 1976 $L^*a^*b^*$ uniform color space.

The goodness of fit R^2 is 0.957 and 0.999, respectively, which means that there is a highly significant linear positive and linear negative correlation between the color coordinates a^* and b^* and the chroma C^* , that is, the chroma C^* of most iolites increased with the increase in the color coordinate a^* and decreased with the increase in the color coordinate b^* , and the color of iolite became darker, transitioning from light blue to blue–violet.

3.2. Chemical Composition

Cordierite, which gemmologically takes the name of iolite, is a silicate mineral with a tetrahedral framework, as shown in Figure 4. The crystal structure of iolite consists of different types of tetrahedra and octahedra. The octahedral M-site is usually occupied by medium-sized cations such as Mg^{2+} , Fe^{2+} , Mn^{2+} or Li^+ , T_1 and T_2 tetrahedra are usually occupied by small-radius cations Al^{3+} as well as Si^{4+} . Al^{3+} located at T_{11} and T_{26} sites, while Si^{4+} at T_{16} , T_{21} and T_{23} sites. Furthermore, sometimes Be and a small amount of Mg^{2+} , Fe^{2+} , and Fe^{3+} can enter the distorted T_{11} site. The octahedron is edge connected to the T_1 tetrahedron, and both are connected to the T_2 tetrahedron through the shared anion O^{2-} . The T_2 tetrahedra are co-angled and form a six-membered ring structure, infinitely along the c-axis to form broad channels in which large-radius cations Na^+ , K^+ , as well as H_2O and CO_2 can be located. The upper and lower hexahedral rings are staggered by about 30° and connected by T_2 tetrahedra [28–32]. Therefore, the chemical equation for iolite can be written as: $(M)_2(T_1)_2(T_2)_2(T_2)_2(T_2)_2(T_1)_2(T_1)_6 O_{18}, (H_2O, CO_2)$.

ED-XRF can be used for a rapid and non-destructive detection of the major elements in iolite, and the data show that the main components of the samples are: Si (47.98–50.372 wt%), Al (34.065–35.667 wt%) and Mg (9.194–14.031 wt%), and also contain the minor elements Fe (1.848–5.18 wt%) and Mn (0.019–0.28 wt%), where $w(Fe_2O_3)$ represents the total iron content (Table 2). The analyses show that the content of Mg gradually decreased with the increase in Fe, while the content of Mn and Al changed relatively flatly. Based on a linear fit to the Fe–Mg content in iolite (Figure 5):

$$w(FeO_{tot}) = 15.936 - 1.35w(MgO) \quad (r = -0.919 \quad R^2 = 0.844)$$

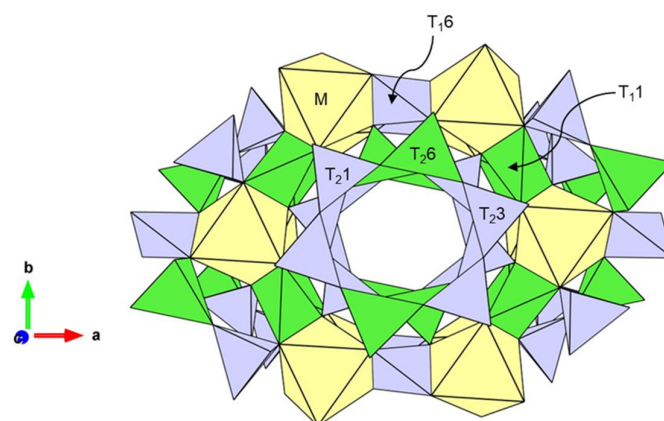


Figure 4. The crystal structure of iolite (from Christian Bertoldi [33]).

Table 2. The ED-XRF data of iolites.

Sample	Iol-1	Iol-4	Iol-7	Iol-5	Iol-11	Iol-12	Iol-14	Iol-18	Iol-20
Simulated color									
L^*	67.07	64.85	60.44	58.92	55.72	49.06	46.85	35.89	29.47
C^*	10.46	9.96	10.82	11.22	12.64	21.22	21.72	18.48	7.60
h°	288.50	288.93	290.51	290.29	290.33	290.81	291.21	293.21	295.25
Oxides (wt%)									
SiO ₂	48.758	48.372	49.193	48.907	50.045	49.201	48.856	49.204	49.089
Al ₂ O ₃	34.727	34.585	34.065	34.835	34.462	34.510	35.117	34.787	35.012
MgO	13.186	14.031	13.342	12.982	11.623	12.544	12.615	11.433	9.249
Fe ₂ O ₃	2.026	1.848	2.432	2.393	2.643	2.259	2.493	3.556	5.180
SO ₃	1.150	0.936	0.834	0.689	0.947	1.285	0.844	0.885	1.026
MnO	0.071	0.020	0.033	0.097	0.074	0.060	0.034	0.042	0.280
Others	0.082	0.208	0.101	0.097	0.206	0.141	0.041	0.093	0.164
Total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00

Among them, r is the Pearson correlation coefficient, which is used to describe the degree of linear correlation between two variables; R^2 represents the goodness of fit and is used to describe the degree of fit of the regression line to the observations, i.e., the percentage of variability in the dependent variable explained by the regression equation. A high negative correlation exists between $w(\text{FeO}_{\text{tot}})$ and $w(\text{MgO})$, the decrease in the Mg content can explain 84% of the increase in the Fe content. It indicates that Fe^{2+} is the main form of Fe element present in iolite, which is consistent with the results of Charles A. Geiger [34]. The remaining iron is present in T_{11} tetrahedra as Fe^{3+} to account for its blue color and strong pleochroism and a small amount of Fe^{3+} can also displace Al^{3+} in the composition [35]. The Mg^{2+} in the secondary composition can also be replaced by a small amount of Mn^{2+} , which also explains the discrete gray spots in Figure 5.

3.3. UV–Vis Spectroscopy Characteristics of Iolite

The UV–vis absorption spectrum of iolite is shown in Figure 6. All samples show a broad absorption band centered at 570 nm in the visible region; as the color deepens, this absorption becomes progressively stronger. It is assumed that this absorption band is attributed to the charge transfer between Fe^{2+} on octahedra and Fe^{3+} in the co-sided

T₁1 tetrahedra [4], and leads to the blue appearance of iolite. Three weak absorption peaks are found in the violet-blue region, the absorption at 390 nm corresponds to the d-d orbital spin-forbidden transition of Fe³⁺ (⁶A_{1g} → ⁴T_{2g}) [36,37], and the absorption at 441 nm is generated by the d-electron transition of Fe³⁺ from the ground state ⁶A₁ energy level to the excited state ⁴A_{1g} and ⁴E_g energy levels. Yan Q [38] et al. attributed the peak of pyrope-spessartine garnets near 424 nm to Mn²⁺ and six-fold spin-forbidden transitions of Fe³⁺ (⁶A_{1g} → ⁴A_{1g} + ⁴E_g). Therefore, it is assumed that the peak at 423 nm in iolite is also caused by a combination of Mn²⁺ and Fe³⁺. In addition, the absorption of the sample near 500 nm in the blue–green region is associated with the spin-forbidden transition of Fe²⁺.

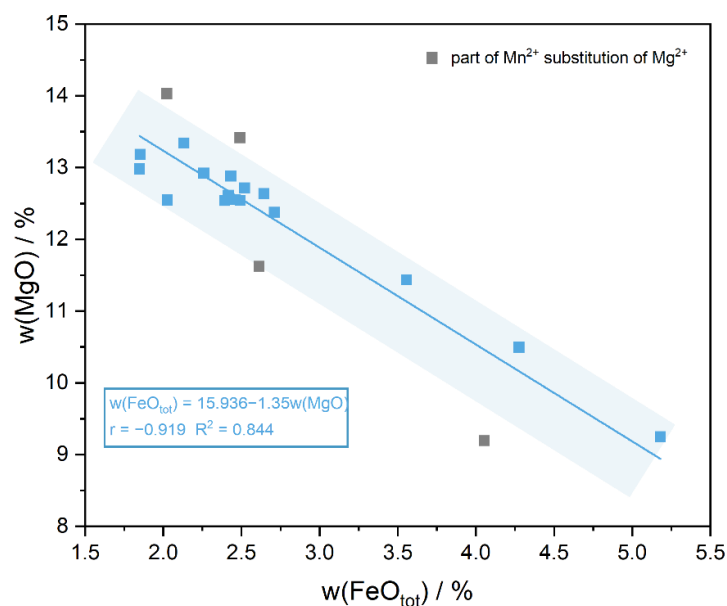


Figure 5. $w(\text{FeO}_{\text{tot}})$ is significantly negatively correlated with $w(\text{MgO})$, and the discrete gray points are due to a small amount of Mg^{2+} substitution by Mn^{2+} .

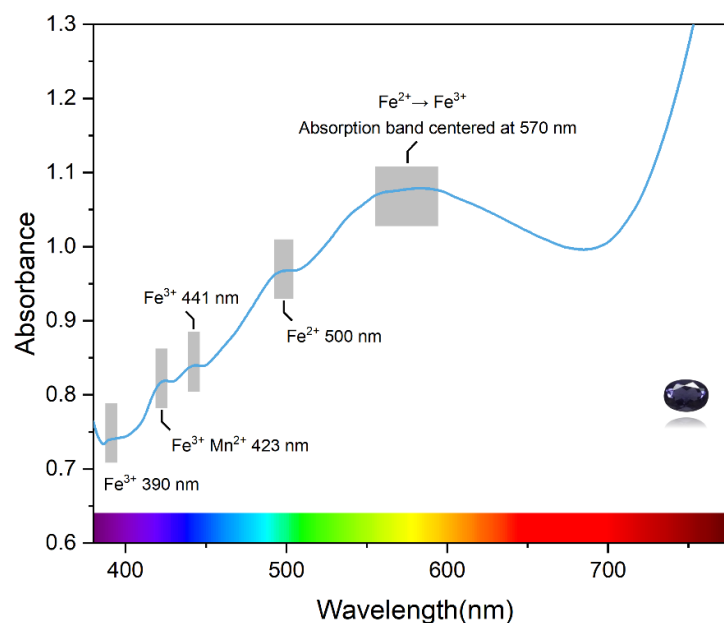


Figure 6. UV-vis spectroscopy of the iolite.

4. Discussion

4.1. Relationship between Fe, Mn and Color

Both Fe and Mn are transition metal elements that are often associated with gemstone color. We explored the correlation between the content of transition metal elements and each color parameter of 20 iolite samples by bivariate correlation analysis. The test is two tailed and the linearity between the two datasets can be determined by the Pearson correlation coefficient r : the correlation is weak or absent when $|r| < 0.3$; the correlation is low when $0.3 \leq |r| < 0.5$; the correlation is medium when $0.5 \leq |r| < 0.8$; the correlation is high when $|r| \geq 0.8$ (Table 3).

Table 3. Results of bivariate correlation analysis.

Oxides (wt.%)		L^*	C^*	h°
FeO _{tot}	r	−0.831 **	0.005	0.877 **
	sig	0.000	0.984	0.000
MnO	r	−0.387	−0.219	0.439
	sig	0.092	0.354	0.053
FeO _{tot} + MnO	r	−0.822 **	−0.010	0.869 **
	sig	0.000	0.967	0.000

The results showed a high correlation between $w(\text{FeO}_{\text{tot}})$ and the lightness L^* ($r = -0.831^{**}$) and the hue angle h° ($r = 0.877^{**}$) in blue iolite. It shows that the concentration of iron has a significant effect on the lightness L^* and the hue angle h° . With the increase in the Fe content, the lightness L^* of the sample decreased almost linearly, the hue angle h° showed an overall increasing trend (Figure 7), the color of iolite appeared from light to dark blue. Thus, iolite with a high iron content exhibits a highly saturated blue color, accompanied by a lower lightness. The correlations between $w(\text{MnO})$ and color parameters were all relatively weak. In order to explore whether Mn has a contribution to the color of iolite, a bivariate correlation analysis was performed between the Fe + Mn contents and the color parameters, r instead resulting in a decrease in the values of r . It indicates that Mn has less effect on the color of iolite, which may be related to the low content of Mn in the samples, so Fe is the main color element in iolite.

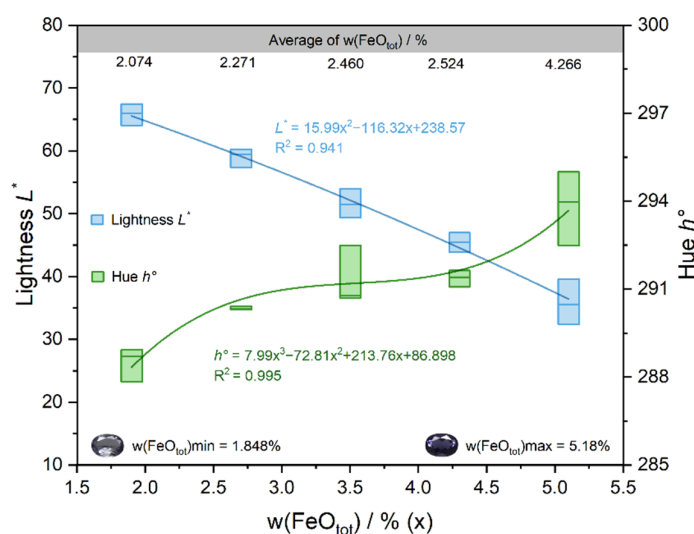


Figure 7. Relationship between $w(\text{FeO}_{\text{tot}})$ and color parameters.

4.2. Color Causes of Blue Iolite

Mn^{2+} and Fe^{3+} have the same electronic structure, both are chromophores, and will only produce obvious color at higher concentrations [39]. Mn^{2+} is less abundant in iolite,

compared to Fe^{2+} , only a small amount of Mn^{2+} can replace Mg^{2+} on the octahedral M-site. So Mn^{2+} does not contribute significantly to the color of iolite. Additionally, whether it is Mg-rich iolite or Fe-rich iolite, the Fe^{3+} content is low: the amount of Fe^{3+} in tetrahedra is usually less than 0.004 a.p.f.u. [35]. Fe^{3+} absorbs to a weaker extent, and the charge transfer in the visible light region is more intense than the color caused by dispersed metal ions [40]. Thus, Fe^{3+} also has a weak effect on the color of iolite. In iolite, the role of Fe^{3+} contributes more to the charge transfer, which shifts the color of the iolite from blue to violet, so that the color of iolite is more due to the intervalence charge transfer between Fe^{2+} and Fe^{3+} .

4.3. Influence of Standard Light Source on Color Appearance

Using the X-Rite SP62 portable spectrophotometer, based on the CIE 1976 $L^*a^*b^*$ uniform color space, with the Munsell N9 color card as the background, the color parameters of 20 iolites were tested under D65, F2 and A light sources. One-way ANOVA was used to explore the impact of different standard light sources on the color appearance of iolite, the results are shown in Table 4. When the significance coefficient p is <0.05 , it is considered to have a significant impact.

Table 4. ANOVA results for the color parameters.

Color Parameters	df	Mean Square	F	p
L^*	2	1.557	0.020	0.980
a^*	2	60.629	28.995	0.000
b^*	2	19.542	0.924	0.406
C^*	2	22.884	0.990	0.381
h°	2	771.901	379.442	0.000

It is suggested that different standard light sources have no significant effect on the lightness L^* ($p = 0.980$), a little effect on the color coordinate b^* ($p = 0.406$) and the chroma C^* ($p = 0.381$), but the greatest effect on the color coordinates a^* ($p = 0.000$) and the hue angle h° ($p = 0.000$). F represents the between-group variance, and the F of the hue angle h° is the largest (379.442), indicating that the light source change has the greatest impact on the hue angle h° .

Different light sources have different spectral energy distributions, which determine the color-rendering properties by influencing its rendering indices, and ultimately produce different colors when irradiated to the gemstone. As shown in Figure 8a,b, when the light source is switched from D65 to F2 and A, the hue angle h° of iolite shows a downward trend. The energy of the daylight source D65 is concentrated in the blue zone, which helps iolite to produce a blue hue, so that iolite has the largest hue angle h° under D65. The spectral energy of the fluorescent light sources F2 and A are concentrated in the yellow and red regions, respectively, the blue hue of the iolite is disturbed, hence the hue angle h° becomes smaller.

Under the three standard light sources, the color coordinates of iolite are positive for a^* and negative for b^* . When the light source is switched from D65 to F2 or A, the a^* decreases and the red density decreases. The b^* is the smallest under F2 with the highest blue concentration, and the largest under A with the lowest blue concentration, both make iolite present the highest chroma C^* under F2. At the same time, F2 has higher energy in the purple wavelength region, and its color light has a purple light part. When the purple light is superimposed on the blue–violet iolite samples, the samples show a fuller blue–violet color, so under the F2 light source, the chroma C^* is the largest. A light source is a color light with higher energy in a longer wavelength range, its color light tends to be orange–red. When the orange–red light is superimposed on the blue iolite samples, the chroma C^* is reduced. Therefore, the D65 light source is suitable for the evaluation of iolite, while the F2 light source is suitable for the sale and display of iolite because it can show the blue color of iolite more effectively.

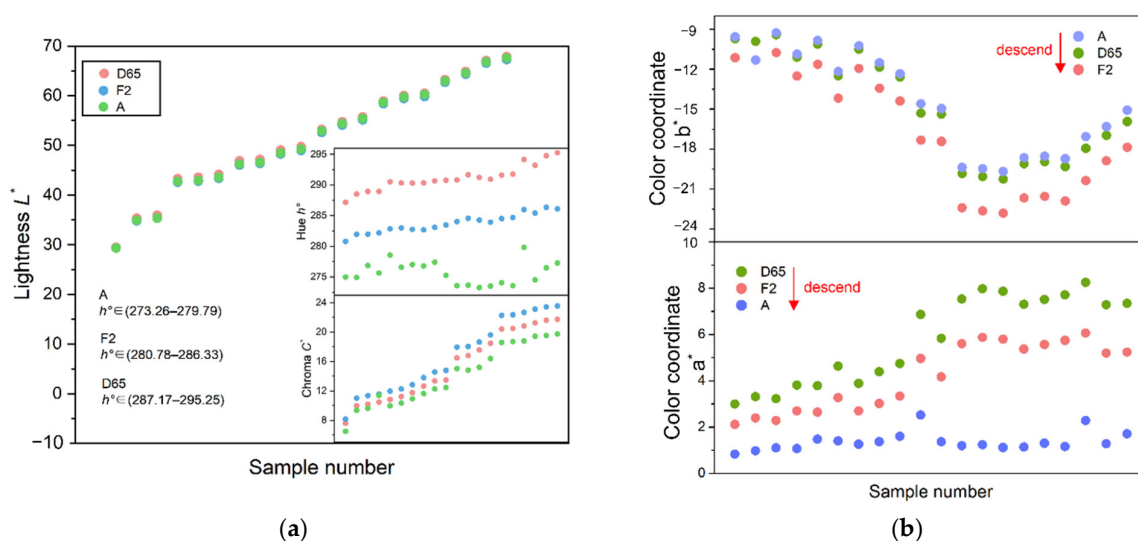


Figure 8. Comparison of color parameters under three kinds of standard light sources. (a) Comparison of the lightness L^* , the chroma C^* , and the hue angle h° ; (b) comparison of color coordinates a^* and b^* .

5. Conclusions

- (1) The color coordinates a^* and b^* were the main factors affecting the variation of the chroma C^* .
- (2) Fe is mainly present as Fe^{2+} and controls the lightness L^* and the hue angle h° , while Mn does not have a significant effect on the color of iolite.
- (3) The intervalence charge transfer between Fe^{2+} and Fe^{3+} is responsible for the blue color of iolite.
- (4) The color parameters of iolite are affected by the change of light sources, and the hue angle h° is the most affected.

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Conflicts of Interest: The authors declare no conflict of interest.

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