Study on the Correlation between Fe^{2+} and Peridot's Yellow Green Color and Quality Evaluation of Color Based on CIE1976 $L^*a^*b^*$ Uniform Color Space

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Abstract. Based on CIE1976 $L^*a^*b^*$ uniform color space, 54 peridot samples' color were measured under D65 light source. The component analysis results showed that all samples are forsterite and chrysolite. It showed that with the increase of Fe^{2+}, peridot's color turns to yellow. But Fe^{2+} has no significantly correlation with chroma. By applying clustering analysis and discriminant analysis, the results showed that peridot's yellow green color were divided well into eleven categories by K-means clustering analysis, and ANOVA showed L^*, a^* and b^* values among various categories were significantly different (sig＜0.001). A linear discriminant function model of the eleven categories of peridot's yellow green color was established by Fisher discriminant analysis which had excellent performance of discriminant with the model checking accuracy of 98.1%. Based on lightness and chroma, peridot's yellow green color was graded into seven levels, consisting of Fancy Vivid(FV), Fancy Intense(FI), Fancy Deep(FDe), Fancy Light(FL), Fancy Dark(FDa), Light(L), Very Light(VL).

1. Introduction

Peridot, known as "the gem of the sun", is one of the earliest gems for decorating. Peridot was firstly found in Egypt about 3000 years ago. Some important peridot deposits were found in Arizona, Colorado, Hawaii and Mogok. In China, we had found high quality peridot deposits in Damaping and Jiaohe areas[1-3].We can get the information such as the causes and the character of the mantle rock from olivine because it is the main composition of the mantle rock [4]. It also because peridot have a excellent crystallinity and pure chemical composition, researchers can get more information from it. That's why the high quality peridot is becoming more and more precious.

It's generally believed that Fe^{2+} is the main color-causing ion in peridot[5]. However, the description to peridot's color is too abstract to understand and the contribution of Fe^{2+} to the color of peridot remains unclear. We use Electro Probe Microanalyzer(EPMA) to analyze the chemical composition of peridot. Based on the uniform color space CIE1976 $L^*a^*b^*$, the color of peridot is characterized by X-Rite SP62. We discuss the correlation between Fe^{2+} and lightness, chroma, hue of peridot's color.

For colored gemstones, color is the most important factor to evaluate its quality. To a large extent the price of colored gemstones was determined by the color quality. For a long time, people evaluate gems only by naked aye and experience. But with the development of image processing technique and the application of chromatic in gemology, the way of color evaluation changed from experience to quantitative characterization, which makes the gems evaluation more scientific. The reason why we use the uniform color space CIE1976 $L^*a^*b^*$ is the difference of yellow-green color we calculated based on this color space was close to the difference we observe by naked eye. So we quantitative characterized peridot's color based on the uniform color space CIE1976 $L^*a^*b^*$[6-13], combining with K-means clustering analysis and Fisher discriminant analysis to classify peridot's color and evaluate its quality, which provides reference to the evaluation of peridot's color.

2. Samples and Experiments

54 pieces peridot with well purity were selected as samples, all these gemstones are from 4mm×6mm×3mm to 6mm×8mm×4mm, and they are all yellow-green color under the naked eye.

Micro-area chemical analyze at State Key Laboratory of Continental Tectonics and Dynamics in Institute of Geology, Chinese Academy of Geological Sciences. We use JXA-8100 Electro Probe Microanalyzer(EPMA) to analyze. The test conditions: accelerating voltage of 15 KV, beams of 2×10-8A, beam diameter of 5um, spectrographic time of 10s, ZAF correction, SPI composite sample correction.

We use X-Rite SP62 to analyze peridot's color, based on the uniform color space CIE1976 $L^*a^*b^*$, collecting the reflected signal on the surface of the sample by integrating sphere. The test conditions: reflection, not include the specular reflection, D65 standard light source, the observer view of 2°, measuring range from 400nm to 700nm, measuring time less than 2.5s, wavelength interval of 10nm, the voltage is 220V, the current is 50Hz.

3. Results

3.1. Electron microprobe analysis

The test results indicate that the chemical composition of 54 samples are close to each other, which mainly are FeO, MgO, SiO_2 and small amout of NiO_2. The mass fraction of $FeO_{(T)}$ is 7.620%-10.421% with an average of 9.034%, and MgO is 47.344%-50.593% with an average of 48.575%. According to the research of peridot's composition by the predecessors, the mass fraction of MgO is 28.11%-55.31% and FeO is 3.02%-33.58%[5], the peridot samples we test are within the range of the predecessors' study. We use Geokit to analyze the composition data, finding the mineral composition mainly are forsterite(Fo) and fayalite(Fa). Forsterite is ranging from 88.89% to 92.00%, with an average of 90.43%. Fayalite is ranging from 7.91% to 10.93%, with an average of 9.44%(Table 1). According to the composition plots of peridot(Fig.1) we know that these peridot samples are chrysolite and forsterite. And according to the research of Zhen jianping et al., the samples we test are fertile mantle type peridots[5]. We use anionic method and make oxygen atom number is 4 to calculate the crystal chemical formula of peridot. The cations the number of which is less than 0.01 are not included in the crystal chemical formula. The crystal chemical formula we got is $(Fe_{0.16\text{-}0.22}Mg_{1.74\text{-}1.85}Ni_{0.00\text{-}0.01})_{1.94\text{-}2.02}Si_{0.99\text{-}1.03}O_4$.

Tab.1 Result of microprobe analysis of peridot

	FeO	MgO	Fo	Fa
Content range （%）	7.620-10.421	47.344-50.593	88.89-92.00	7.91-10.93
Average（%）	9.034	48.575	90.43	9.44

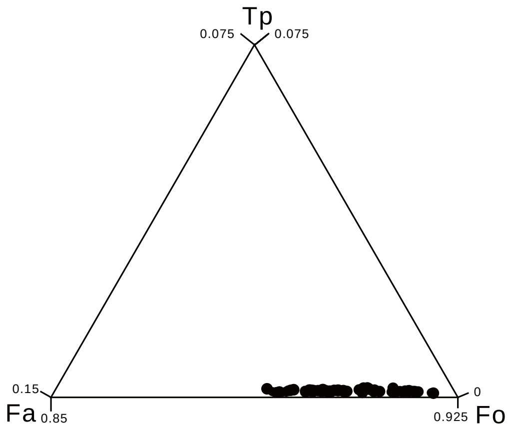


Fig.1 Composition plots of peridot

3.2. Color index measurement

3.2.1. Test result

Based on the uniform color space CIE1976 $L^*a^*b^*$, 54 peridot samples were tested under D65 standard light source. As is shown in the Fig.2, lightness $L^* \in (56.52, 69.39)$, i.e. it is middle level of lightness. And $a^* \in (-18.46, -12.26)$, $b^* \in (26.67, 41.96)$, in which is yellow-green color area. Chromite $C^* \in (29.38, 45.22)$, i.e. it is middle level of chromite. Hue angle $h° \in (109.30, 117.70)$.

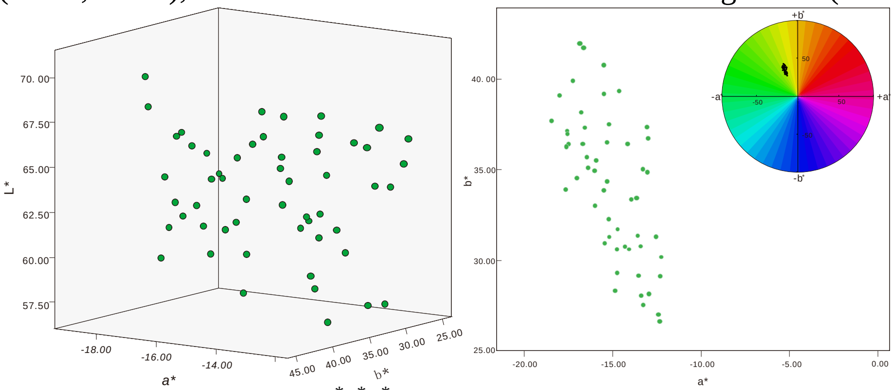


Fig.2 Peridots' color plots in CIE1976 $L^*a^*b^*$ uniform color space and chromaticity diagram(L^*=65)

3.2.2. The correlation between peridot's color constituents

There exists extremely notable linear ration between C^* and b^*(Fig.3a), R^2 = 0.984. But there are no significant correlation between C^* and a^*. It indicates that the chromite of peridot mainly controlled by yellow tones. h° and L^* are positively correlated (Fig.3b), i.e. hue angle increases with lightness, and peridot turns green. That is the same as what we observe by naked eyes.

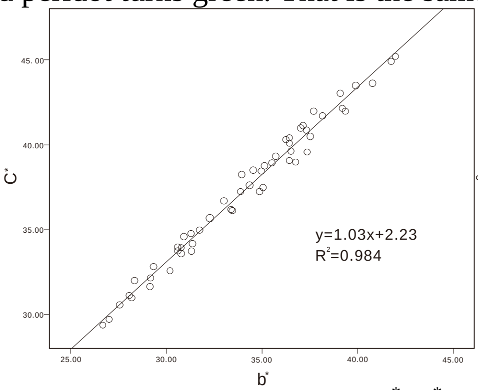


Fig.3a Scatter diagram of C^*- b^*

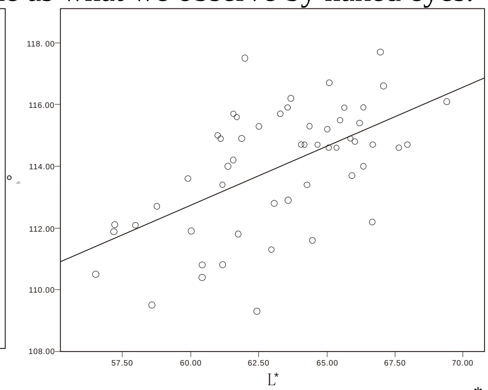


Fig.3b Scatter diagram of h°- L^*

3.3. Contribution of Fe^{2+} to peridot's color

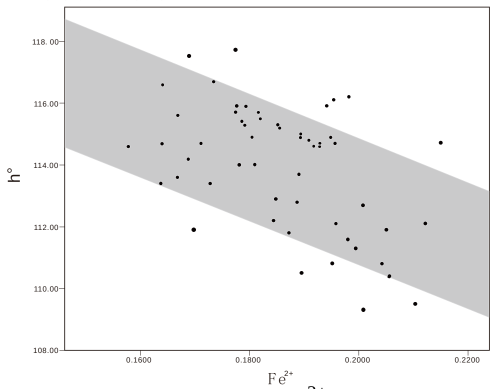


Fig.4 Correlation between Fe^{2+} and hue angle

We analyze the correlation between Fe^{2+} and the color data, finding Fe^{2+} has no significant correlation with L^* and C^*, but it is negatively correlated with h°. As is shown in the Fig.4, though the data is scattered, it shows a certain linear correlation, which indicating that h° decrease with the increase of Fe^{2+}, i.e. peridot's hue may turn to yellow tone gradually with the increase of Fe^{2+}. Besides, yellow tone has a higher lightness than green tone, therefore Fe^{2+} has indirect effects on the lightness of peridot, which is the reason why the correlation coefficient in Fig.3b isn't high enough.

3.4. Color classification and quality evaluation

According to the research of Du Hongmei et al., clustering analysis and discriminant analysis method is very effective for the classification of the gem colors, and takes an example of jade to demonstrate its feasibility[14]. Clustering analysis is a statistical analysis technology which divides research object into relative homogeneous group. Discriminant analysis, one of the important methods of multivariate statistical analysis, which is according to the information provided by the known classes of samples to summarizes the regularity of the various types and establish a discriminant formula and criterion to identifying the type of new sample points. We use these two methods to classify peridot's color. We based on the uniform color space CIE1976 $L^*a^*b^*$ to quantitate peridot's color and use K-means clustering analysis method and Fisher discriminant analysis method of SPSS22.0 statistical analysis software to find a objective and effective solution to classify peridot's color.

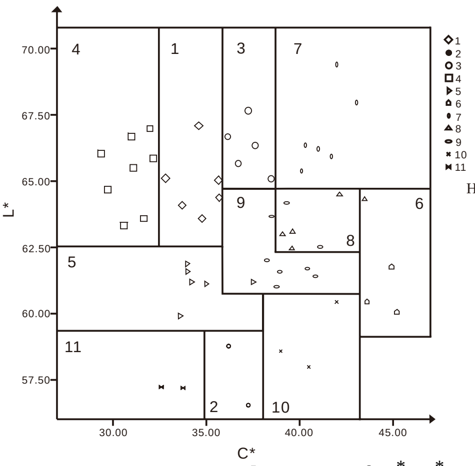


Fig.5a Scatter diagram of L^*-C^*

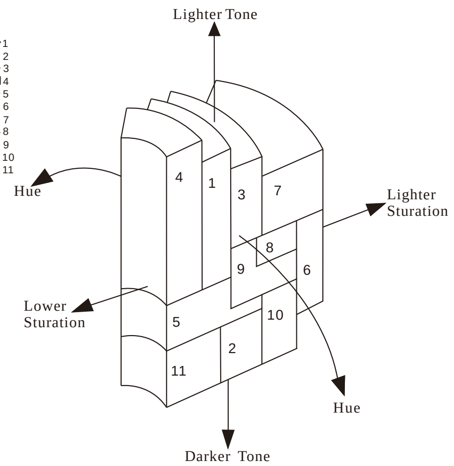


Fig.5b Color stereo of peridot

Firstly, we make L^*,a^*,b^* as variable factors and divided 54 peridot samples' yellow-green color into eleven types by K-means clustering analysis method. Eleven Types of peridot's L^*,a^* and b^* exist significant difference with all sig$<$0.001, indicating a excellent classification result. Then we use Fisher discriminant analysis method to analyze these eleven types of color, finding all sig$<$ 0.001 as well, indicating that discriminant variables reflect the classification characteristics well. We plug the all value of L^*,a^*,b^* into Fisher discriminant formula, finding the accuracy as high as 98.1% which indicating that this discriminant model is effective for the classification of the peridot's yellow-green color. The result of classification shown as scatter diagram(Fig.5a) and color stereo(Fig.5b).

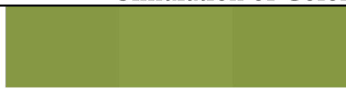
Through clustering analysis and discriminant analysis for the classification of the peidot's color, in combination with its commercial value and reference the classification of color grade of international color diamond to classify peridot's yellow-green color. For the case that the samples' tonal difference is small, we consider classifying peridot's color with L^* and C^*.

The high quality peridot require pure color(green), vivid and homogeneous distribution. Therefore we take C^* as first and L^* as second to evaluate peridot's color. L^* too high may lead to peridot pale in color while too low will cause peridot dark. Hence peridots which have middle lightness are higher quality than which of same chroma. Considering that eleven types of color are too much to evaluate color quality easily and the evaluation system of color gems often chooses five or seven levels, so we decide to merge eleven types color into seven types.

We use clustering analysis eleven types of color again. When we make the classification result is five or seven types, it turns out that both type-6 and type-10 are in the same level. However, the quality of type-6 is better than type-10 because type-6 is a type of middle lightness and high chroma, that's not consistent with the facts. So it's not suitable to divide peridot's color into five levels. When we divide the eleven types of color into seven levels, it turns out that type-4 and type-1, type-2 and type-11, type-8 and type-9, type-3 and type-7 are in the same level respectively. We have found two types of color in the same level are similar to each other by comparing lightness and chroma of each type. Therefore we decide dividing eleven types of peridot's color into seven levels.

Highest quality color should have a middle lightness and a very high chroma. As is shown in the Fig.6, we know that type-6 is the best color among all, type-8 and type-9 which have a middle lightness and a high chroma are the second. In the same way from good to bad in the order are type-10, type-7 and type-3, type-2 and type-11, type-5. Type-1 and type-4 are the worst color types for they have a very high lightness and a low chroma even presenting a gray tone. We name these seven levels color from good to bad in the order as Fancy Vivid(FV), Fancy Intense(FI), Fancy Deep(FDe), Fancy Light(FL), Fancy Dark(FDa), Light(L), Very Light(VL). It should be considered the feeling of the naked eyes for the process of color evaluation is also the process of color objectification and color ordering[15-16], thus we use Photoshop CC to simulate each peridot sample's yellow-green color and show them in the Table.2.

Tab.2 Various types of olivine color quality division

Level	Color	Basis of	Simulation of Color
Ⅰ	Fancy Vivid(FV)	Very high C^*, middle L^*	
Ⅱ	Fancy Intense(FI)	High C^*, middle L^*	
Ⅲ	Fancy Deep(FDe)	High C^*, low L^*	
Ⅳ	Fancy Light(FL)	Middle C^*, high L^*	
Ⅴ	Fancy Dark(FDa)	Low C^*, low L^*	
Ⅵ	Light(L)	Low C^*, high L^*	
Ⅶ	Very Light(VL)	Low C^*, very high L^*	

4. Conclusion

(1) The content of fosterite(Fo) in the peridot samples is 88.89%-92.00%, which is indicating the peridot samples are chrysolite and forsterite.

(2) The yellow-green hue difference is small in the peridot samples. There exists extremely notable linear ration between C^* and b^*. But there are no significant correlation between C^* and a^*. Therefore peridot's color is controlled by the yellow hue. Hue angle(h°) and lightness(L^*) are positively correlated, i.e. hue angle increases with the increase of lightness, peridot turns to green.

(3) Hue angle(h°) decrease with the increase of Fe^{2+}, i.e. peridot's hue may turn to yellow gradually with the increase of Fe^{2+}.

(4) The results of variance analysis shows well when we divide peridot's color into eleven categories by clustering analysis and discriminant analysis. We grade these eleven categories of color into seven levels based on the color difference, in order, Fancy Vivid, Fancy Intense, Fancy Deep, Fancy Light, Fancy Dark, Light, Very Light.

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