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STRUCTURAL FEATURES OF GEM MINERALS AND IDENTIFICATION AND EVALUATION

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Abstract: Gem has long been chased because of its cultural and historical significance and beauty, and the identification and evaluation of its structural features have also been paid attention to. Plum blossom jade and emerald were identified and analyzed by means of the conventional detection method, infrared spectroscopy and X-ray powder diffraction analysis. The results showed that plum blossom jade with black and brown background and red, white and brown amygdaloid body spots had a diameter of 1–10 mm, a relative density of 2.69–2.75 g/cm^3 and a refractive index of 1.52–1.55, and its minerals contained water molecules, quartz, low albite, anorthoclase and andesite. It was observed that emerald was green, greasy, glassy, transparent, with a relative density of 2.66–2.78 g/cm^3 and 1.58 refractive index, and its minerals contained water molecules; there were two kinds of arrangement in crystal channels. The main mineral component of emerald was beryl, and its purity was high.

***Keywords*:** *plum blossom jade, emerald, infrared spectroscopy, X-ray diffraction*

1. INTRODUCTION

In China, gem culture has existed long, and its origin can be roughly divided into three reasons. The first one is production demand (Majzlan et al. 2016). At the time when human civilization has just started, a large number of production tools are needed to support production activities. Ordinary rocks are not suitable and their hardness is also

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not up to the standard, but some gems have relatively high hardness, suitable for expanding production needs. The second reason is inheritance of culture and status (Başıbüyük 2018). Because of its hardness, gem is used as tools of production; hence they have the color of cultural heritage. At the same time, some gems are considered to have "destiny" because of their inherent color patterns. After being carved, the gems are favored by those in power, thus possessing the symbolic significance of status. The last reason is aesthetics (Serdari 2017). With the development of social productivity, people's attention gradually turned to living quality after the problem of clothing and food was solved, and gems are carefully carved as their beautiful patterns conform to aesthetics. In modern times, gem still has considerable value. For example, the production of natural gems with beautiful patterns is relatively rare, which ensures the value of hard currency, and because of the non-renewability of natural gems, it has the nature of preservation or even appreciation (Zaitsev et al. 2016). Gems also have the value of ornamentation and decoration after polishing, and some gems are added with value of etiquette status because of historical factors; more importantly, gems have research value as the formation of natural gems usually has a great connection with geological changes (Liu et al. 2015). Analyzing the structure of natural gems can understand the geological changes of gem origin, and even summarize the law of exploring gem origin. In addition, the carving process of gems can also reflect the social style and experience of that time and provide theoretical basis for historical research. Caucia et al. (2015) have studied the physical and chemical properties of some opals from the Akali Mine in Peru by means of optical analysis, hydrostatics, refractive index and X-ray diffraction and found that opals had green, blue and pink colors, had greasy luster, and had no color change. X-ray Powder Diffraction (XRPD) analysis showed that opals in the Akali Mine were CT types with variable contents of cristobalite and tridymite. Chen et al. (2016) studied the mineral composition of imitation Turquoise which is a new product in the market by traditional gemological method, infrared absorption spectrometry and X-ray diffraction and found that the imitation had no natural color characteristics and mainly consisted of enstatite and quartz. The physical characteristics and mineral composition of plum blossom jade and emerald were identified and analyzed by means of conventional detection, infrared spectroscopy and X-ray powder diffraction analysis. This study can provide an effective reference for the study of structure of gem mineral and improve the accuracy of gem identification to ensure the stable of the gem market.

2. PRINCIPLE OF LARGE-SCALE MINERAL DETECTING INSTRUMENT

2.1. PRINCIPLE OF X-RAY POWDER DIFFRACTION ANALYSIS

X-ray powder diffraction (Ishikawa et al. 2014) can be used for analyzing the composition of gem mineral. X-ray powder diffraction analysis includes single crystal diffraction and polycrystal diffraction. Single crystal diffraction is mainly used for

detecting the crystal structure of single crystal materials. Polycrystal diffraction mainly detects bulk materials such as powders and polycrystals. X-ray diffraction is one of the simplest and most effective methods for qualitative and quantitative analysis of the phase of testing materials. Its principle is that different crystals have different structural parameters, such as cell size, lattice arrangement and atomic arrangement. When X-ray irradiates these crystals at a certain frequency, X-ray diffraction occurs and forms diffraction lines, which are singular and correspond to the structural characteristic parameters of crystals. When minerals contain more than two kinds of crystals, the characteristics of diffraction lines of crystals will not interfere with each other, but simply superimpose. Based on the above principle, a database of crystal-diffraction lines can be established. The crystal composition in samples can be rapidly analyzed by comparing the diffraction lines of samples with the database. The expression of qualitative analysis of phrase of X-ray diffraction (Galović and Peh 2014) is:

$$l = \frac{n\lambda}{2\sin\alpha}, \tag{1}$$

where l stands for interplanar spacing, which is correlated to crystal structure, n stands for diffraction series, λ stands for incident wavelength, and α stands for diffraction angle. The phrase of sample was analyzed based on interplanar spacing and relative strength.

2.2. PRINCIPLE OF INFRARED SPECTROSCOPIC ANALYSIS

As a kind of crystal, gems have lattices and keep vibration, deflection and interaction at all times (Liu et al. 2015). Microscopically, the lattice always vibrates in its own region. The vibration form of a single lattice is in a random state, which can not be accurately described. However, the overall lattice vibration in a crystal can be described by statistical method. Electrons at different lattice sites will transit in the vibration process of crystal lattice, and energy will be absorbed or released during the transition process. Infrared light with a specific frequency has a corresponding fixed energy. When the energy reaches the energy required for the transition of lattice electrons, lattice electrons will absorb the energy of infrared photons for the transition and produce the corresponding infrared absorption spectra (Ling et al. 2015). Different crystal lattices need to absorb different energy frequencies; hence the infrared absorption spectra produced are also different. In this paper, two main points should be paid attention to when gem minerals were analyzed by infrared spectroscopy. Firstly, the infrared light used for analysis needs to reach the energy frequency required for the lattice electron transition of gem minerals. The sec-

ond point is whether the molecular structure of gem minerals is symmetrical or not; if symmetrical, there will be no coupling effect under infrared light, that is, gems have no infrared activity; otherwise coupling effect will be produced under infrared light, i.e., gems have infrared activity. Infrared spectroscopy has the advantages of high sensitivity, accuracy and repeatability in the analysis of structure of gems mineral (Krasnoshchekova et al. 2015). In this paper, the infrared spectrum of gems can be analyzed to get five important information for gems identification: hydroxyl groups in minerals, impurity atoms in minerals, identification of artificial gemstones, identification of gems with similar appearance and identification of antique gems.

3. STRUCTURAL CHARACTERISTICS OF PLUM BLOSSOM JADE AND EMERALD

Plum blossom jade is named for its pattern like plum blossom. It is also named Ruyang jade because of its origin in Ruyang, Henan province, China. Among many gem minerals, plum blossom jade is a unique gem mineral in China. Its formation process is as follows. During volcanic eruption, magma wrapped a large number of air bubbles. When magma cooled, bubbles formed different shapes of pores. Over time, other minerals gradually accumulated in the pore, forming "plum blossom" spots on plum blossom jade. The Mohs hardness of plum blossom jade ranges from 6.5 to 7. Its petrological name is black amygdaloidal andesite. The plum blossom spots are plagioclase. At the micro level, they are interwoven by microcrystalline plagioclase and vitrified glass flakes. Its interwoven form is interwoven into felts. Because of their micro-interwoven structure, the spots are very tough. The main mineral composition includes quartz, epidote, calcite, etc. Branches in plum blossom jade connect "spots" together, which are cracks produced by magma cooling and are gradually filled with silicate and carbonate during the formation process.

Emerald, also known as cat's eye, is mainly produced in Colombia, Russia, India and China. The petrological classification of emerald is beryl. It is green because of the large amount of chromium ions. The rock strata where emeralds are formed are often squeezed by stress in the process of geological change. Therefore, emeralds are usually in a broken state or have cracks. Complete emerald is rare. The cracks are filled with different content during the formation process. The origin of emeralds can be judged according to the content. The main chemical composition of emerald is Be_3Al_2 [Si_6O_{18}]. At the micro level, the crystal is hexagonal columnar and hexagonal bipyramid, and the single crystal can aggregate into granules. At the macro level, emerald has glass luster, is green transparent or translucent, has 7.5 Mohs hardness, and

has visible conchoid crack section.

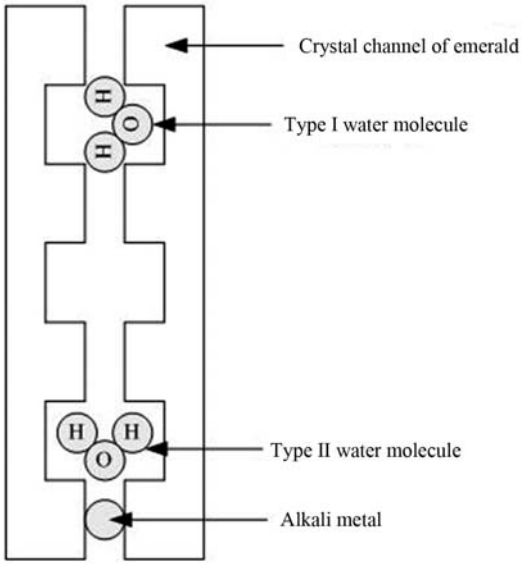

Fig. 1. The arrangement of water molecules in crystal channels of emerald

As shown in Fig. 1, there are channels between crystals and water molecules in the channels as emeralds are usually cracked. There are two arrangement types of water molecules in the channels. One is that the symmetrical axis of water molecules is perpendicular to the channel, which is called type I water molecule. The other is that water molecules rotate due to the influence of alkali metal electric field in minerals, whose symmetrical axis is parallel to the channel. It is called type II water molecule. Its absorption intensity of infrared light is proportional to the content of alkali metals in minerals.

4. TEST MATERIALS AND METHODS

4.1. TEST SAMPLES

Three raw plum blossom jades and three raw emeralds were selected; the raw plum blossom jades were from Ruyang, Henan, and the raw emeralds were from Malipo, Yunnan, China. The samples are shown in Figs. 2–7.

Fig. 2. Plum blossom jade sample A1

Fig. 3. Plum blossom jade sample A2

Fig. 4. Plum blossom jade sample A3

Fig. 5. Emerald sample B1

Fig. 6. Emerald sample B

Fig. 7. Emerald sample B3

4.2. DETECTING INSTRUMENTS AND METHODS

Experimental instruments included X-ray diffractometer, 1° divergence slit and anti-scatter slit, 0.2 mm acceptance slit, Cu target, solid-state counter, Fourier transform infrared spectrometer (Pottermcintyre et al. 2014) and sample crusher.

Experimental methods

(1) X-ray powder diffraction analysis: Firstly X-ray diffractometer was set according to operation manual, and then the parameters were set: the tube voltage was 40 kV and the current was 40mA; the representative part of the sample was crushed by the sample crusher and then grinded repeatedly in a mortar until there was no granular sensation. The sample powder was put into the sample sink of the diffractometer and scanned at a scanning angle of 6–80°, 0.1 s every step and 0.02° for one step.

(2) Infrared spectroscopic analysis: Firstly, the representative parts of the samples were crushed by a crusher and then ground in a mortar until no granular sensation was observed. Then, potassium bromide was added. The blended sample powder was crushed into thin sheets by a tablet press and put into the sample sink of the spectrometer. The infrared spectrometer used magnetic resonance imaging (MIR) light source (Galović 2016). After sampling, the samples and background were scanned for 16 seconds at a scanning speed of 10 kHz.

(3) Conventional detection: The surface characteristics of the gem minerals were observed by a microscope and briefly described; the relative density of the samples was detected using hydrostatic method, and its expression (Galović 2016) was:

$$\text{Relative density of sample} = \frac{\textit{Weight}\ \text{of sample in the air}}{\textit{Weight}\ \text{of sample in the air} - \textit{Weight}\ \text{of sample in 4 °C water}}. \quad (2)$$

The refractive index of the samples was detected using a refractometer.

5. RESULTS

5.1. CONVENTIONAL DETECTION RESULTS OF SAMPLES

As shown in Table 1, plum blossom jade minerals was black and brown, with many spots on the surface generally. The spots were amygdaloid body in the shape of round, oval, bean-pod and connected cloud. The visual diameter of the amygdaloid body was between 1 mm and 10 mm, and they were red, white and brown. The majority of emerald mineral samples were brown minerals containing iron. The green part was emerald, which seemed like glass grease, and was slightly transparent and contained gas-liquid inclusions.

Table 1. The color and structural characteristics of plum blossom jade and emerald

Sample No.	Color	Structural characteristics
A1	Red spots, brown background	Spots were amygdaloid body in the shape of round, oval and bean-pod, with a diameter between 1 mm and 10 mm; most of the spots were red, and some was white.
A2	White spots, black background	Spots were amygdaloid body in the shape of oval and bean-pod, with a diameter between 1 mm and 10 mm; they were white.
A3	Brown spots, black background	Spots were amygdaloid body in the shape of connected clouds, with a diameter between 1 mm and 10 mm; they were brown.
B1	Green, brown base	The overall structure was dominated by brownish minerals containing iron; the green part was emerald with glassy luster.
B2	Green, brown base	The overall structure was dominated by brownish minerals containing iron; the green part was raw emerald with glassy luster, and the cross section seemed greasy.
B3	Green, brown base	The overall structure was dominated by brownish minerals containing iron; the green part was slightly transparent raw emerald with glassy luster, containing gas-liquid inclusions.

As shown in Table 2, the relative density of plum blossom jade minerals was about 2.69–2.75 g/cm^3, and the refractive index was between 1.52 and 1.55. The relative density of different plum blossom jade mineral samples was similar, but there were some differences. The main reason might be that the main components and structures of minerals were the same, but the structures of the other components were different. The relative density of emerald minerals was between 2.66 and 2.78 g/cm^3, and the refractive index was about 1.58. Different emerald minerals had different relative densities, which basically follows the rule that the darker the green, the greater the relative density. The reason was that the color of emerald was provided by metal ions. The darker the color, the higher the content of metal ions, i.e., the higher the relative density.

Table 2. The relative density and refractive index of plum blossom jade and emerald

Sample No.	Weight in the air/g	Weight in 4 □ water/g	Relative density (g/cm^3)	Refractive index
A1	4.1023	2.5828	2.6998	1.525
A2	4.1258	2.6098	2.7215	1.551
A3	4.1125	2.6146	2.7456	1.531
B1	3.1212	1.9492	2.6631	1.581
B2	3.1524	2.0043	2.7458	1.584
B3	3.4512	2.2090	2.7784	1.583

5.2. INFRARED SPECTROSCOPIC ANALYSIS OF SAMPLES

The infrared absorption spectra of plum blossom jade minerals were basically the same. As shown in Fig. 8, the plum blossom jade minerals had seven infrared absorption bands. The infrared absorption wavenumber was 3409 cm^{-1}, 1036 cm^{-1}, 788 cm^{-1}, 649 cm^{-1}, 590 cm^{-1}, 532 cm^{-1} and 462 cm^{-1}, from large to small. 3409 cm^{-1} was induced by the common stretching vibration. 1036 cm^{-1} was induced by the asymmetric stretching vibration of silicon-oxygen key. 788cm^{-1} was induced by the stretching vibration of silicon-silicon key. 649 cm^{-1} was induced by the bending vibration of oxygen-silicon (aluminum)-oxygen key. 590 cm^{-1} was induced by the stretching vibration of silicon-aluminum key. 532 cm^{-1} was induced the common coupling vibration of oxygen-silicon-oxygen key and sodium-oxygen key; the former was bending vibration and the latter was stretching vibration. 462 cm^{-1} was induced by the common coupling vibration of oxygen-silicon-oxygen key and sodium-oxygen key. Vibration key position inducing wavenumber 1036 cm^{-1}, 788 cm^{-1}, 649 cm^{-1}, 590 cm^{-1} and 532 cm^{-1} belonged to albite. Though the vibration key position inducing 462 cm^{-1} was the same with 532 cm^{-1}, the vibration key position inducing 462 cm^{-1} belonged to daphnite.

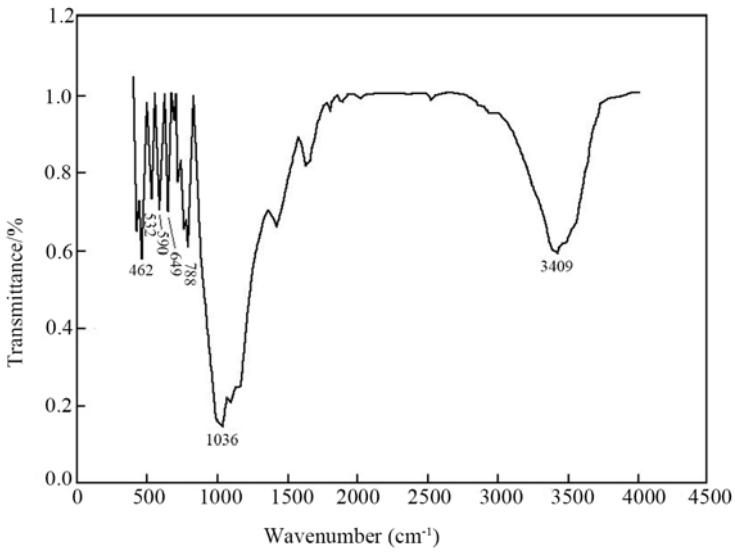

Fig. 8. The infrared transmission spectrum of plum blossom jade minerals

The infrared absorption spectra of emerald mineral were nearly the same. As shown in Fig. 8, the absorption peak of 5274 cm^{-1} was caused by the co-vibration of water molecules and mineral components in emerald minerals, and the absorption peak band around the absorption peak was called hydration frequency vibration absorption band. The absorption peaks of absorption wavenumber 1632 cm^{-1}, 3595 cm^{-1} and 3697 cm^{-1}

were caused by the bending vibration and stretching vibration of water molecules in minerals. The arrangement of water molecules in 3697 cm^{-1} absorption peak was classified as type I, and it was asymmetrical stretching vibration. The arrangement of water molecules in 3595 cm^{-1} absorption peak was classified as type II, and it was symmetrical stretching vibration. The arrangement of water molecules in 1632 cm^{-1} absorption peak was classified as type II, and it was bending vibration. Besides the absorption spectra induced by vibration of water molecule, there was also absorption peak between 2500 cm^{-1} and 3000 cm^{-1}; the comparison of standard spectra of emeralds suggested that the absorption peak was a deliberately added oil absorption peak, which was used for simulating the means of improving the quality of emerald in the market.

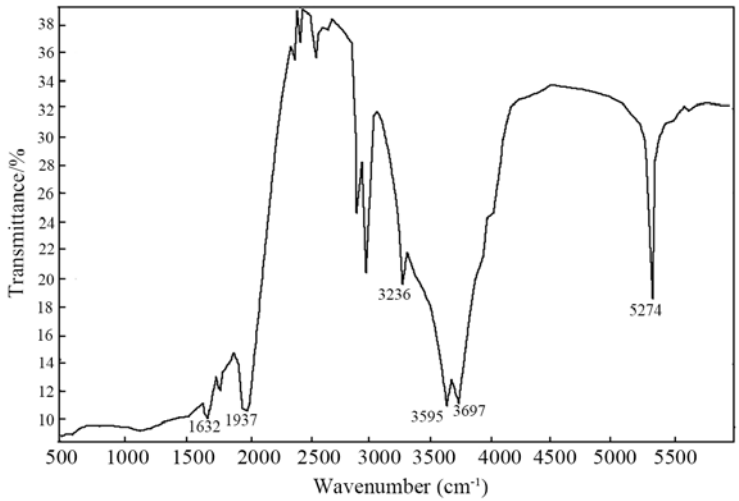

Fig. 9. The infrared transmission spectrum of emerald minerals

5.3. X-RAY POWDER DIFFRACTION ANALYSIS

As shown in Fig. 10, the d value and relative strength of the main peak in the diffraction spectrum of plum blossom jade were compared with those of different minerals in the standard database, which could help qualitatively analyze the mineral components in plum blossom jade minerals; d value refers to interplanar crystal spacing, which is a characteristic parameter of crystal.

The spectra of plum blossom jade minerals showed that there were eight main peaks. Diffraction peaks with 4.2545 d value and 25.0% relative strength, 3.3431 d value and 99.9% relative strength, 1.8175 d value and 14.9% relative strength and 1.5411 d value and 10.6% relative strength had slight differences with the diffraction peaks of quartz; therefore the minerals contained quartz. Diffraction peaks with 4.0165 d value

and 14.9% relative strength and with 3.1643 d value and 13.1% relative strength were slightly different with the diffraction peaks of low albite. Diffraction peaks with 3.6654 d value and 11.5% relative strength were slightly different with the diffraction peaks of anorthoclase; therefore the minerals contained anorthoclase. Diffraction peaks with 3.1906 d value and 28.8% relative strength had little differences with diffraction peaks of andesite; therefore the minerals contained andesite.

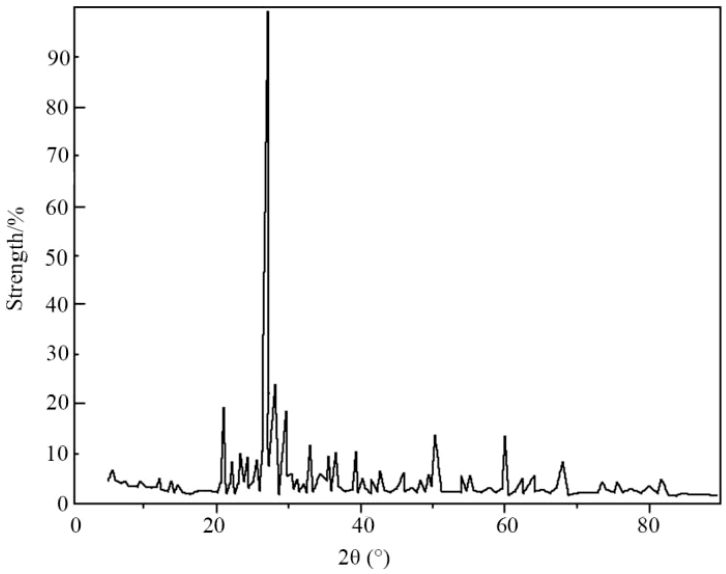

Fig. 10. The X-ray diffraction spectrum of plum blossom jade minerals

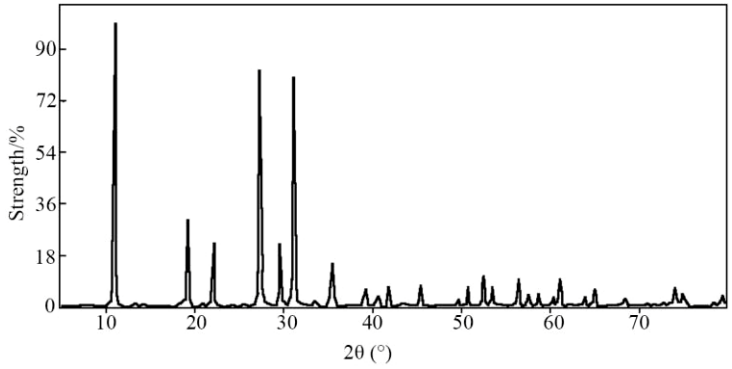

Fig. 11. The X-ray diffraction spectrum of emerald minerals

As shown in Fig. 11, there were three obvious diffraction peaks in the X-ray diffraction diagram of emeralds, diffraction peak with 8.0045 d value and 100% relative

strength at 11.044° diffraction angle, diffraction peak with 3.2601 d value and 92.8% relative strength at 27.335° diffraction angle and diffraction peak with 2.8731 d value and 80.5% relative strength at 31.117°. The comparison between Fig.e 11 and diffraction spectra of minerals in standard database (Tungpalan et al. 2015) suggested that the diffraction peak position of emerald minerals was basically consistent with that of No. 07-0430, i.e., beryl. It demonstrated that beryl was the main mineral component of emerald minerals and emerald minerals had high purity.

Comparing the results obtained by conventional detection method, infrared spectroscopy and X-ray powder diffraction, it can be seen that the conventional detection method could only identify the general properties of gemstone minerals, including density, hardness and other general characteristics, which are not unique for gemstone minerals. If the conventional detection method is used, the identification mainly relies on experience, and the accuracy is difficult to guarantee. However, infrared spectroscopy and X-ray powder diffraction identified minerals according to the characteristics of elements and crystal structure in minerals. For gemstones, the elements and crystal structure are unique. Therefore, the two methods mentioned above can accurately identify different gemstone minerals and even identify the authenticity of gemstones.

6. CONCLUSION

In this study, plum blossom jade minerals and emerald minerals were selected as test samples, and the structural characteristics and composition of the two gem minerals were analyzed by means of conventional detection, infrared spectroscopic analysis and X-ray powder diffraction. The results obtained are as follows. Plum-blossom jade minerals had black and brown background, with amygdaloid body spots, and they were mostly red, white and brown, with variable shapes; the diameter of plum blossom jade was between 1 mm and 10 mm, the relative density was between 2.69 and 2.75 g/cm^3, and the refractive index was between 1.52 and 1.55. Brown iron was the main component of emeralds, and the emerald part was green, greasy and subtranslucent; the relative density of emeralds was between 2.66 and 2.78 g/cm^3, and the refractive index was about 1.58. It was inferred from the infrared spectrum and X-ray diffraction spectrum of plum blossom jade that plum blossom jade minerals contained water molecules, quartz, low albite, anorthoclase and andesite. It was inferred from the infrared spectrum and X-ray diffraction spectrum of emeralds that there were water molecules in emerald minerals, and water molecules were arranged in two ways in crystal channels. The comparison between the X-ray diffraction diagram and standard spectrum demonstrated that the main mineral component of emerald samples was beryl and emeralds had high purity. Only two kinds of gem minerals were identified in this study, and more kinds of gem minerals need to be identified in the future to verify the effectiveness of the proposed methods.

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