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Analysis of Crystallinity and Physical Properties of the Bio-solar Gemstone

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Abstract – Bio-solar is one of the natural gemstones found in the province of Aceh in Indonesia. Nevertheless, detail information on this gemstone is still unknown. The purpose of this work is to analyze the chemical composition, crystallinity, specific gravity, and hardness of the bio-solar gemstone from Aceh. X-ray diffraction has been used to determine the chemical composition and crystallinity of the sample. The hardness of the sample was measured by using Diamond Selector II. The specific gravity of the sample was determined by the water displacement method. Our results show that the bio-solar gemstone from Aceh is composed of CaO, SiO_2, MgO, Al_2O_3, Fe_2O_3, TiO_2, and Ni_2O_3 phases. It is found that the bio-solar gemstone is a crystalline material, not amorphous. The average crystallite size of this gemstone is 35.3 nm (353 Å). The specific gravity of bio-solar gemstone is found to be 3.09 – 3.34. Its hardness is 3 to 4 Mohs. Our analysis suggests that the bio-solar can be classified as a soft vesuvianite gemstone.

Keywords: gemstone, jade, vesuvianite, bio-solar

Introduction

The gemstone is a mineral crystal which has been mostly used as jewelry. One of the favorite gems is jade (Walker, 1991). Chinese people believe that jade can make good health and long life. Jade price can exceed the price of gold (New York Times, 2010). Meanwhile, natural jade is rare; therefore, its price has become expensive. Consequently, only high-class people use natural jade, and it has become a symbol of nobility and perfection (Desjardins, 2015). For those reasons, people try to search for natural jade everywhere in the world. Jade has been found in several countries, including in Indonesia. Jade has been defined to consist mostly of jadeite or nephrite (Harlow *et al.*, 2005). Jadeite is a pyroxene group that includes Na(Al, Fe)-Si_2O_6 (Morimoto, 1988; Prewitt *et al.*, 1966; Htein *et al.*, 1994). In the meantime, nephrite is an amphibole group which is composed of Ca_2 (Mg, Fe)$5Si_8O_{22}(OH)_2$ (Liu *et al.*, 2011). Nephrite can be classified into two types that are nephrite-tremolite and nephrite-antigorite. Serpentine is a type of jade. Sometimes, it is called a serpentine jade. Its color can be green, yellowish-green, or brownish-green. Two kinds of serpentine are antigorite and chrysotile. Vesuvianite is also known as jade. It is often called as California jade or American jade. Vesuvianite is also known as idocrase. Sometimes, idocrase is used for gemstone-quality vesuvianite. According to web mineral data, the chemical composition of vesuvianite gemstone is SiO_2, Al_2O_3, MgO, CaO, and H_2O (webmineral.com). However, Groat et al. found that the chemical composition of vesuvianite is SiO_2, Al_2O_3, MgO, CaO, FeO, Fe_2O_3, and MnO (Groat *et al.*, 1992).

The research on jade has been conducted continuously. In Turkey, Murat *et al.* characterized the purple stone, which mainly contained SiO_2 (63.54%) and Al_2O_3 (20.17%), and Na_2O (6.71%). The specific gravity of this gemstone was found to be 3.24 to 3.42. This Turkish gemstone was classified as jadeite-jade (Murat *et al.*, 2012). In Indonesia, the community continues to explore precious stones, including in the province of Aceh. Some gemstones were discovered in Nagan Raya, Aceh (Tempo, 2015). A previous study classified gemstones

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from the region of Aceh in Indonesia as jadeite, nephrite, serpentine, and vesuvianite based on their colors and hardness (Nurul *et al.*, 2014). They suggested that the colors and hardness of the gemstones were influenced by impurity mineral compounds (Nurul *et al.*, 2014). One of a well-known gem from the province of Aceh is called bio-solar. It is so beautiful, as shown in Figure 1. Its color is dark yellow. A recent study showed that this bio-solar gemstone contains 59.8% of CaO, 19.7% of SiO_2, 11.1% of Fe_2O_3, 7.5% of Al_2O_3, and 1.3% of NiO. Based on its chemical composition, the bio-solar gemstone from Aceh can be classified as a type of vesuvianite jade (Akmal *et al.*, 2016). However, detail information of this gemstone, such as crystallinity, specific gravity, and hardness, is still unknown. According to the previous study, the vesuvianite gemstone is crystallite, having the tetragonal atomic structure (Anthony *et al.*, 2010). Its values of specific gravity and hardness are in the range 3.32 – 3.43 and 6 – 7 Mohs, respectively (Anthony *et al.*, 2010). The objectives of this study are to understand the chemical composition, crystallinity, specific gravity, and hardness of the bio-solar gemstone. The results are presented in this paper.

Figure 1. Bio-solar gemstone from Nagan Raya, Aceh

Materials and Methods

The bio-solar gemstone samples were purchased from the Aceh Gemstone Market in Tingkeum, Aceh Besar District, where they are initially from the district of Nagan Raya in the Aceh province, Indonesia. Minerals identification and the crystallite size of bio-solar gemstone were determined by using x-ray diffraction (Shimadzu, D7000). The sample of bio-solar gemstone for x-ray diffraction measurement was crushed into powder. The measurement condition was at room temperature, x-ray wavelength λ = 1.5406 Å, continuous scan, scan range (2θ) from 10 to 80 degrees with each step of 0.02 degrees. The sample size for specific gravity and hardness measurements was 17 mm x 15 mm x 7 mm. Four samples were used in the analyses. The hardness of bio-solar gemstone was measured by using Diamond Selector II. The specific gravity of the bio-solar gemstone was determined by using the water displacement method. Electronic balance with high sensitivity was used in determining the specific gravity.

Results

Our result of x-ray diffraction (XRD) measurement of the bio-solar gemstone is shown in Figure 2. There were eleven sharp peaks observed in our XRD measurement. The highest peak was seen at the Bragg angle (2θ) of 32.48 degrees. The kind of minerals (a type of oxide) contained in the bio-solar gemstone was determined by comparing the XRD data to JCPDS (Joint Committee on Powder Diffraction Standards).

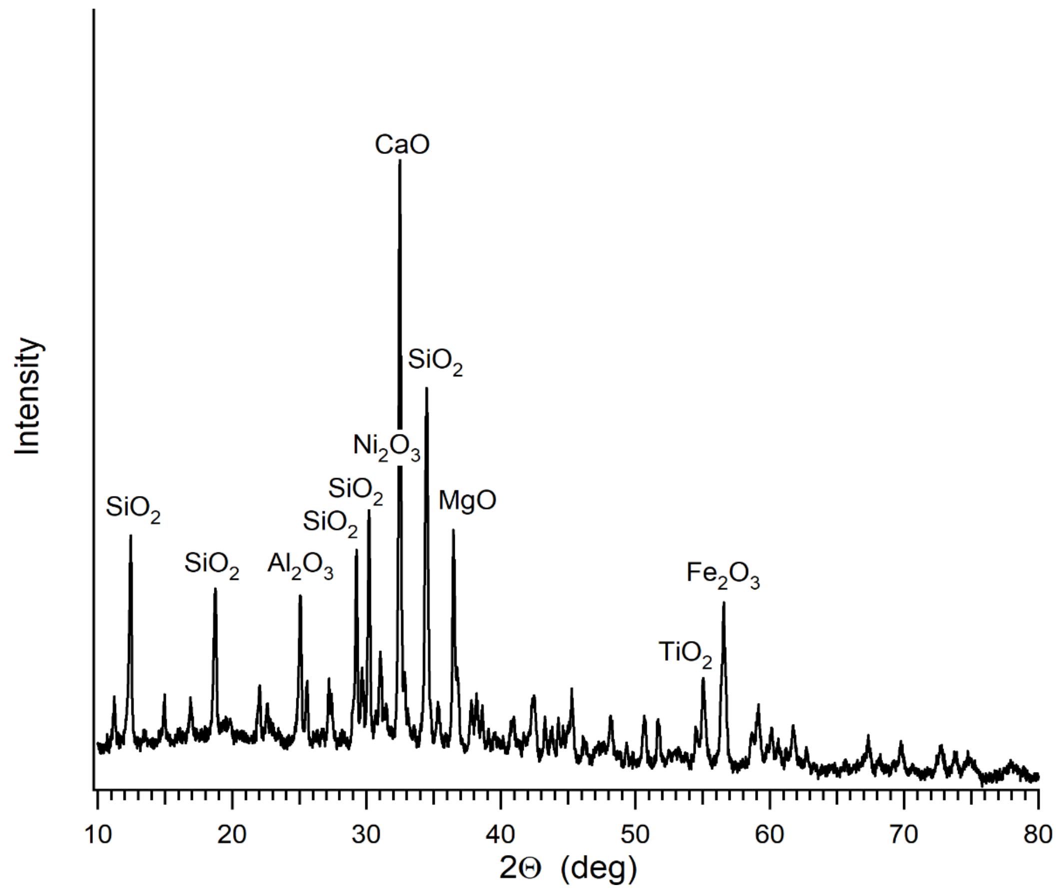

Figure 2. X-ray diffraction data from the bio-solar gemstone

The comparisons between our XRD data and JCPDS data are shown in Table 1. By comparing the experimental data to JCPDS data, we found that the highest peak at 32.48 degrees is the CaO phase. The second highest peak was observed at 34.46 degrees, which is the SiO_2 phase. Some other peaks having SiO_2 periods were found at the Bragg angles of 30.18 degrees, 29.24 degrees, 12.46 degrees, and 18.74 degrees. The MgO, Fe_2O_3, Al_2O_3, and TiO_2 phases were observed at the Bragg angles of 36.48 degrees, 56.56 degrees, 25.56 degrees, and 55.06 degrees, respectively. There was also a peak seen at 31.02 degrees, which is the Ni_2O_3 phase. As shown in Table 1, our experimental data are in good agreement with the JCPDS data.

Table 1. The comparison of XRD data from bio-solar gemstone and JCPDS data

No	2θ (degrees)		Intensity	Phase	hkl
	Experiment	JCPDS			
1	32.48	32.300	100	CaO	111
2	34.46	34.645	66	SiO_2	642
3	30.18	30.248	49	SiO_2	533
4	36.48	36.932	45	MgO	111

5	12.46	12.923	45	SiO_2	220
6	29.24	29.150	42	SiO_2	620
7	18.74	18.315	36	SiO_2	400
8	25.56	25.839	35	Al_2O_3	012
9	56.56	56.575	33	Fe_2O_3	211
10	55.06	55.134	28	TiO_2	202
11	31.02	31.575	24	Ni_2O_3	002

Our XRD results (see Figure 2) revealed that the bio-solar gemstone is crystallite, not amorphous. It is a well-ordered crystal. The crystallite size of the bio-solar gemstone can be calculated by using equation (1) (Suryanarayana *et al.*, 1998).

$$D = \frac{k.\lambda}{\beta.\cos(\theta)} \quad (1)$$

Where, k = 0.95 (constant), λ = 0.15406 nm (the wavelength of x-ray used in the experiment), θ = the Bragg angle in degrees, β = the full width at half maximum (FWHM) in radian, D = the crystallite size of sample in nm.

The full width at half maximum was calculated by fitting the Lorentzian function to the experimental XRD data, as shown in Figure 3. By using the equation (1) above, the crystallite size of the CaO phase is found to be 45.97 nm (459 Å). The crystallite size of SiO_2 is 33.77 nm (337 Å), which is smaller than the crystallite size of CaO. The crystallite size of MgO is 38.39 nm (384 Å). The crystallite size of Al_2O_3 is 42.99 nm (429 Å), which is almost the same as the crystallite size of CaO. However, the crystallite sizes of Fe_2O_3 and TiO_2 are rather small, 23.80 nm (238 Å), and 27.02 nm (270 Å), respectively. The crystallite size of phases contained in the bio-solar gemstone is listed in Table 2.

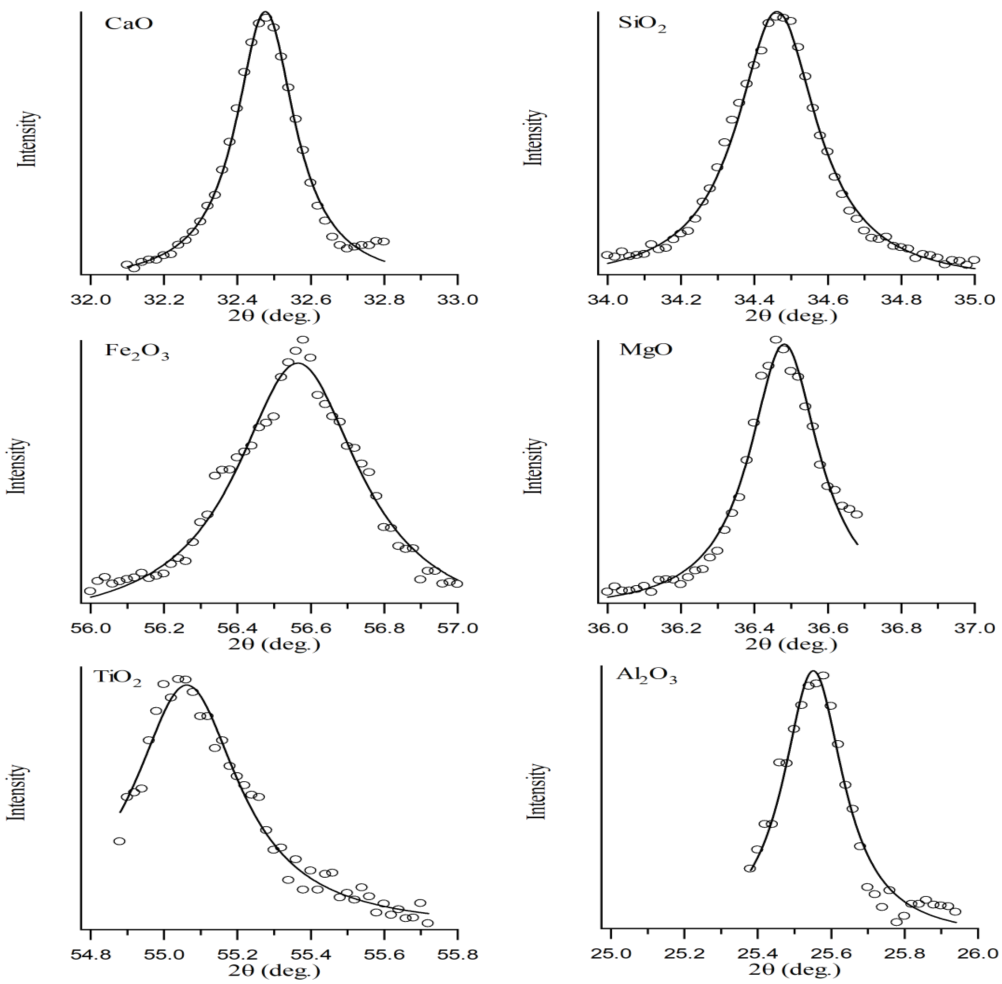

Figure 3. The comparison of the XRD data (circles) and calculated the best fit of Lorentzian function (solid lines) for the bio-solar gemstone

Table 2. The crystallite size of the bio-solar gemstone

No	Phase	2θ (deg)	FWHM (deg)	D (nm)
1	CaO	32.48	0.19	45.97
2	SiO_2	34.46	0.26	33.77
3	MgO	36.48	0.23	38.39
4	Fe_2O_3	56.56	0.40	23.80
5	Al_2O_3	25.56	0.20	42.99
6	TiO_2	55.06	0.35	27.02
	The average crystallite size (nm)			35.33

The specific gravity (SG) and hardness of the bio-solar gemstone have been measured. Our measurement results showed that the specific gravity of the bio-solar gemstone is in the range of 3.09 – 3.34. The hardness of the bio-solar gemstone is found to be in the range of 3 to 4 Mohs.

Discussion

The comparison between minerals contained in the bio-solar gemstone with some other gemstones is listed in Table 3. Mark √ indicates the type of oxides contained in the gemstones.

Table 3. The comparison of bio-solar with selected gemstones

Type of oxide	Bio-solar (this study)	Jadeite (Prewitt *et al.*)	Nephrite Actinolite (Evans *et al.*)	Nephrite Tremolite (Ballinaro *et al.*)	Serpentine Clinochrysotile (Pollastri *et al.*)	Serpentine Antigorite (Brindley *et al.*)	Vesuvianite (Groat *et al.*)
SiO_2	√	√	√	√	√	√	√
Al_2O_3	√	√	-	-	-	-	√
Na_2O	-	√	-	-	-	-	-
MgO	√	-	√	√	√	√	√
CaO	√	-	√	√	-	-	√
FeO	-	-	√	-	-	-	√
Fe_2O_3	√	-	-	-	-	√	√
TiO_2	√	-	-	-	-	-	√
MnO	-	-	-	-	-	-	√
Ni_2O_3	√	-	-	-	-	-	-

The present study of bio-solar gemstone contains CaO, SiO_2, MgO, Al_2O_3, Fe_2O_3, TiO_2, and Ni_2O_3. Meanwhile, jadeite contains SiO_2, Al_2O_3, and Na_2O. The mineral composition of bio-solar is quite different than that of jadeite. Thus, the bio-solar is not a type of jadeite. This situation is the same for nephrite actinolite, nephrite tremolite, serpentine clinochrysotile, and serpentine antigorite. Groat *et al.* found that vesuvianite contains SiO_2, Al_2O_3, MgO, CaO, FeO, TiO_2, and Fe_2O_3 (Groat *et al.*, 1992). All minerals included vesuvianite are found in the bio-solar gemstone, except FeO and MnO. However, the amount of FeO and MnO are small; a particular sample (vesuvianite from Mexico) has 0.06% of MnO and 0.68% of FeO (Groat *et al.*, 1992). There is a peak of Ni_2O_3 observed in our XRD data. It was noted NiO (1.24%) in bio-solar gemstone by using X-Ray Fluorescent (XRF) (Akmal *et al.*, 2016). There is a small discrepancy between our result and Akmal's result, which could be due to the sensitivity of XRD and XRF instruments.

The size of crystallite reflects the crystallinity that defines the degree of long-range order in a material which affects the properties of a material. The ideal crystallite size is usually between 500 – 1000 nm. However, the crystallite size is often much smaller than that because of the imperfection of material such as impurity, defect, dislocation (Suryanarayana *et al.*, 1998). The study of the crystallite size of the gemstone is very limited in the literature. For example, the crystallite size of opal gemstone from California USA was found to be 15 nm (150 Å) (Sanders, 1975). The crystallite size of amethyst gemstone from Central Kalimantan was 14 nm (Suastika *et al.*, 2017). A recent study found that the crystallite size of gold nanoparticles was 18 – 24 nm (Ogundare *et al.*, 2019). As shown in Table 3, CaO is the most significant crystallite size among all other phases in bio-solar that is 45.97 nm (459.7 Å). The average crystallite size of bio-solar is 35.33 nm (353 Å), which is far below the ideal size, which could be due to impurity and other imperfection in bio-solar. However, it is larger than the crystallite sizes of amethyst, opal, and gold nanoparticles.

The specific gravity (SG) of the bio-solar gemstone is compared to other gemstones, as shown in Table 4. The SG of bio-solar is 3.09 – 3.34, which is smaller than the SG of jadeite gemstones (Leaming, 1978; Anthony *et al.*, 2010; Franz *et al.*, 2014). The SG of nephrite actinolite is 3.03 – 3.24 that is the same as the SG of the bio-solar gemstone. Meanwhile, the chemical composition of bio-solar is different than that of nephrite actinolite (see Table 3). The SG of bio-solar is higher than the SGs of serpentine clinochrysotile and serpentine antigorite. The bio-solar gemstone is slightly smaller than the SG of vesuvianite from the previous study by Anthony *et al.* (2010). Nonetheless, the chemical composition of bio-solar is about the same as that of vesuvianite gemstone of the prior research by Groat *et al.* (1992).

Table 4. The comparison of the SG and hardness of bio-solar gemstone and other gemstones.

Kind of Gemstone	Specific Gravity (SG)	Hardness (Mohs)
Jadeite (Anthony *et al.*, 2010)	3.24 – 3.43	6 – 7
Jadeite (Franz *et al.*, 2014)	3.25 – 3.40	-
Jadeite (Leaming, 1978)	3.24 – 3.43	-
Nephrite actinolite (Anthony *et al.*, 2010)	3.03 – 3.24	5 – 6
Nephrite actinolite (Leaming *et al.*, 1978)	2.95 – 3.01	-
Nephrite tremolite (Anthony *et al.*, 2010)	2.99 – 3.03	5 – 6
Serpentine clinochrysotile (Anthony *et al.*, 2010)	2.53	2.5
Serpentine antigorite (Anthony *et al.*, 2010)	2.65	2.5 – 3.5
Vesuvianite (Anthony *et al.*, 2010)	3.32 – 3.43	6 – 7
Bio-solar (present study)	3.09 – 3.34	3 – 4

The comparison between the hardness of bio-solar with some selected gemstone from previous studies is shown in Table 4. The hardness of bio-solar is 3 – 4 Mohs, which is much smaller than the hardness of jadeite. The hardness of bio-solar is also lower than the hardness of nephrite. However, the hardness of bio-solar is larger than the hardness of serpentine. The previous study reported that the hardness of vesuvianite is 6 – 7 Mohs (Anthony *et al.*, 2010). The hardness of bio-solar gemstone is significantly lower than that of vesuvianite from the previous study.

Meanwhile, we have shown above that the chemical composition of the bio-solar is about the same as that of vesuvianite. Previous work showed that the impurity could affect the hardness of the crystal (Verma *et al.*, 2014). Sayan et al. found that impurities influenced the hardness of NaCl crystal. The K ion impurity increased the hardness of crystal. But, the Ni ion impurity decreased the hardness of NaCl crystal (Sayan *et al.*, 2001). Based on these studies by Verma ((2014) and Sayan (2001), we speculate that the hardness of bio-solar gemstone is low compared to the regular vesuvianite because of impurities in it such as Ni_2O_3 which was observed by our XRD data.

Conclusion

Our x-ray diffraction analysis showed that the bio-solar gemstone from Aceh Indonesia contains CaO, SiO_2, MgO, Al_2O_3, Fe_2O_3, TiO_2, and Ni_2O_3 phases. The chemical composition of the bio-solar gemstone is about the same as that of vesuvianite. The bio-solar gemstone is crystallite. Its average crystallite size is 35.3 nm (353 Å). The specific gravity of bio-solar gemstone is 3.09 – 3.34, which is about the same as those of vesuvianite.

These results suggest that the bio-solar gemstone from Aceh province in Indonesia is a vesuvianite gemstone. However, its hardness is 3 – 4 Mohs, which is slightly soft compared to the regular vesuvianite.

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