

A Non-destructive Mineralogical Study of Nephritic Artifacts from Itbayat Island, Batanes, Northern Philippines

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ABSTRACT

A non-destructive mineralogical study of nephritic jade artifacts was carried out using a low-vacuum type scanning electron microscope (LVSEM) with an energy dispersive X-ray spectrometer (EDS). Six artifacts from a prehistoric workshop site dated between 2500 and 1300 yr BP at Anaro on Itbayat Island, Batanes Province, northern Philippines, were found to be composed of nephrite (tremolite-actinolite amphiboles) with zinc-bearing chromian-spinel as accessory minerals. The mineralogical characteristics of these nephrite artifacts are comparable with those of Fengtian (Taiwan) nephrite. The results indicate that the original materials for these artifacts were collected in the Fengtian area, near Hualien in eastern Taiwan.

Key Words: Non-destructive analysis, EDS, Nephrite, Artifact, Itbayat, Batanes, Taiwan

INTRODUCTION

Nephritic jade artifacts are commonly found in Neolithic archaeological sites in China (e.g., Huang 1975; Zhang 1995; Tang 1998), Taiwan (e.g., Chen 1998; Lien 1998, 2002; Liu 2003; Hung 2000, 2004) and some regions of Southeast Asia (e.g., Kano 1946; Beyer 1948;

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Fox 1970; Chin 1980; Sung 1989; Dizon 1998; Ha 1998; Nguyen 1998; Tang & Nguyen 2004; Hung *et al.* 2004; Hung & Iizuka 2004). Nephrite was used for tools such as axes, adzes, chisels and spearheads, and was also a highly symbolic stone, used for ornaments and probably ritual objects since Neolithic times. Because nephrite deposits occur rarely in nature, it is widely accepted that chemical analysis is a useful technique to trace specific geological sources or quarries. A sourcing study of nephrite is an important approach to comprehend the nature of an ancient culture.

Several methods of analysis have been applied to the study of nephritic artifacts, both destructive and non-destructive. Destructive mineralogical studies of nephrite artifacts can only be applied to performs, damaged or broken artifacts, because samples need to be cut and/or powdered before analysis. For precious nephrite artifacts, non-destructive Raman spectroscopy (*e.g.*, Xu *et al.* 1996; Lien *et al.* 1996) and non-destructive X-ray fluorescence (XRF) (Douglas 1996, 2003) have respectively been applied to determine mineralogical information and whole-rock chemical compositions. Recently, an advanced method of analysis has been established; combining techniques of PIXE (proton induced X-ray emission) and micro-Raman to measure the major elements of nephrite artifacts (Chen *et al.* 2004). However, many of these methods are not able to identify specific quarries clearly because of overlaps in the mineralogy and whole rock chemistry of many nephrites.

An electron probe micro-analysis (EPMA) has the advantage of being almost non-destructive, unlike most other techniques for chemical analyses, and is able to obtain the chemical compositions of both the nephrite matrix and the accessory minerals from a micrometer-sized sampling area. EPMA sourcing is an advanced technique that can identify specific nephrite sources, and it has been successfully applied in the case of Fengtian (Taiwan) nephrite (Iizuka & Hung 2005: *this issue*). However, it still requires some sample preparation, such as polishing and carbon coating, before analysis can take place.

The newly-developed low-vacuum type scanning electron microscope (LVSEM), equipped with the conventional tungsten (W) hairpin electron gun, combined with an energy dispersive X-ray spectrometer (EDS), can be applied as completely non-destructive technique. It can perform mineral identification and quantitative analysis of elements that occur at more than 1% by weight with easily operation. This is an excellent system for the study of precious ancient materials.

In a 2004 excavation in the Batanes Islands, between Taiwan and Luzon, Bellwood and Dizon found a nephrite workshop at the Anaro site on Itbayat Island (Fig. 1). A great deal more material (including nephrite adzes, discs and rings) has just been found by Bellwood, Dizon and Hung in further June-July 2005 investigations at Anaro, but this report only deals with material from the 2004 season. Small ornaments, possibly including *lingling-o* earrings of Iron Age type (Anaro has also produced some iron tools), were made at Anaro between 2500 and 1300 yr BP by drilling and cutting nephrite (Bellwood & Dizon 2005: *this issue*). This report presents the mineralogical characteristics of six worked nephrite samples from Anaro, using the

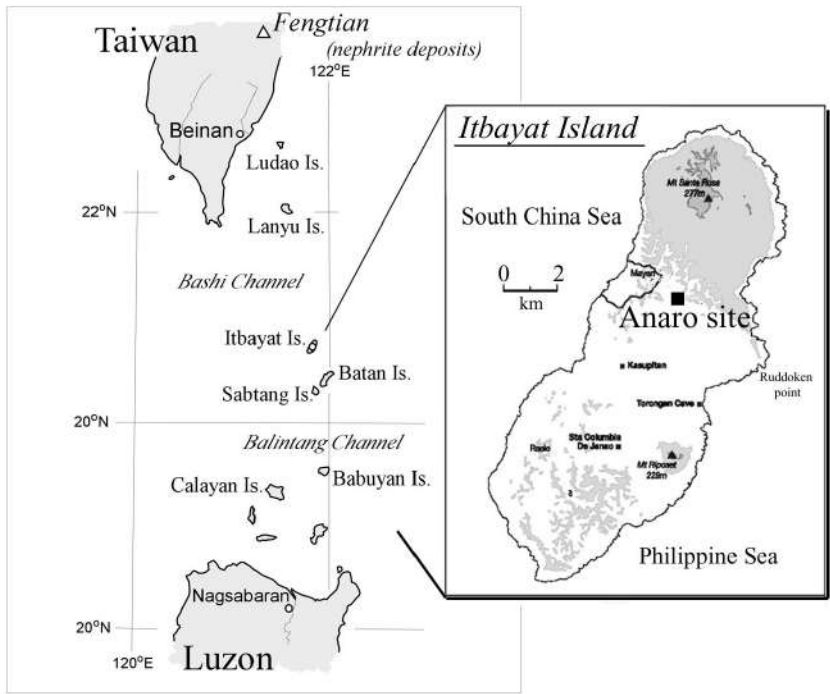


Fig. 1. Map of the Anaro site on Itbayat Island (right), and its location between southern Taiwan and northern Luzon (left).

non-destructive chemical analysis technique of LVSEM with EDS. The analytical results indicate that the nephrite (tremolite-actinolite) originated from the Fengtian source region in eastern Taiwan.

FENGtian (TAIWAN) NEPHRITE

The popular term "jade" is applied to two different metamorphic minerals, jadeite and nephrite, in mineralogy. Jadeite is Na-clinopyroxene and is very rare in the world, and no jadeite deposit has ever been reported in Taiwan. The nephrite is Ca-amphibole and the term "Taiwan jade" is applied to this.

Nephrite deposits are located west of the town of Fengtian in Hualien county, eastern Taiwan. Fengtian nephrite, characteristically green in color, occurs mainly in serpentinite

layers intercalated in black schist or muscovite-quartz schist in the greenschist facies (Tan *et al.* 1978). In Taiwan, since nephrite has been mined in the Fengtian area in the 20th century, it has become accepted that this area was the source for nephrite artifacts in prehistory (Wang *et al.* 1996; Lien *et al.* 1996; Tan *et al.* 1997; Huang & Chou 2001; Yui *et al.* 2001; Lien 2002).

Iizuka and Hung (2005: *this issue*) report a comparison of nephrites from Fengtian and some other East Asia and circum Pacific sources, in order to define their mineralogical characteristics, especially those that can reliably distinguish one source from others. Based on mineral identification and chemical analysis by EPMA, they conclude that Fengtian (Taiwan) nephrites can be discriminated on the following characteristics;

- (1) Fengtian nephrites are characteristically green in color and fibrous in texture, and are bearing black-colored Cr-spinel as an accessory mineral.
- (2) Fengtian nephrites are tremolite-actinolite amphiboles. Although the Mg/[Mg+Fe] ratio has a fairly wide range, it is under 0.93 in all cases (the median is 0.9).
- (3) Cr-spinel, which is a major accessory mineral, bears significant amounts of Zinc (Zn) and Manganese (Mn).

Based on these observations, Iizuka and Hung report that green nephrite artifacts of Fengtian source rock were widely distributed not only in Taiwan but also in the islands of Luzon and Palawan in the Philippines in prehistory (Hung *et al.* 2004; Hung & Iizuka 2004; Iizuka & Hung 2005: *this issue*). Unpublished research carried out in April 2005 also indicates that a lingling-o from Niah Cave in Sarawak is of Fengtian nephrite (report in prep.).

NON-DESTRUCTIVE ANALYSIS OF ANARO NEPHRITE ARTIFACTS BY LVSEM-EDS

The Samples

The six studied samples were excavated from the Anaro-3 site in central Itbayat Island during an excavation in March 2004 (Fig. 1). They are listed in Table 1. All of specimens are worked and broken pieces (Fig. 2). A description of the Anaro site and the implications of the findings there are presented by Bellwood and Dizon (2005: *this issue*).

Analytical procedure

Before SEM observation, the samples were well cleaned with distilled water for several hours in an ultra-sonic bath to remove dust and soil from their surfaces. Each sample was then rinsed in ethanol, and dried in oven overnight at 75°C. The samples were neither damaged nor coated.

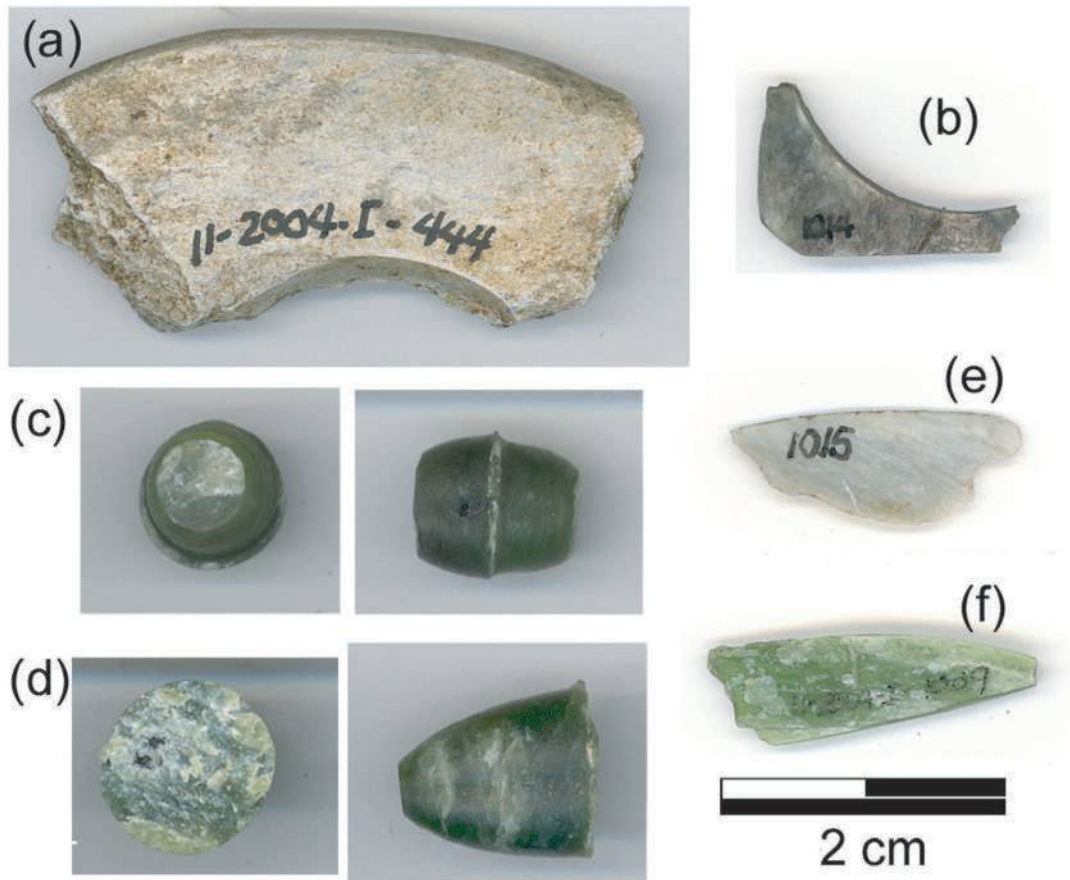


Fig. 2. The nephrite artifacts from Anaro.

- (a) Btn-1: II-2004-I 444: a fragment of a ring.
Cr-spinel inclusions (black in color) are observed on the surface of the brown-white nephrite. Estimated inner and outer diameters of the ring are 34 mm and 75 mm, respectively. Thickness is 5.8 mm, weight 10.4 g.
- (b) Btn-2: II-2004-I 1014. A worked fragment, 2.8 mm in maximum thickness, 0.6 g in weight.
- (c) Btn-3: II-2004-I 1013. Right: a drilled core from a *lingling-o* (barrel shaped), profile view. Cr-spinel grains (black in color) can be observed on the surface of the green nephrite, which is 10.5 mm in length. Left: a view of the right hand end of the above, 9.8 mm in diameter at the center, 6.0 mm at the base. The other end is 6.7 mm in diameter and the weight is 1.5 g.
- (d) Btn-4: II-2004-I 1019. A core of a *lingling-o* (bullet shaped—was it intended to become a projection?). Cr-spinel grains (black in color) can be observed on the surface of the green nephrite. Right: profile view, 14.0 mm long. Left: a view of the base (12.0 mm in diameter). The top is 3.7 mm in diameter, weight 2.8 g.
- (e) Btn-5: II-2004-I 1015. A fragment (knife?). Fibrous texture can be observed on the surface of this white colored-nephrite. 0.7-1.5 mm thick, weight 0.2 g.
- (f) Btn-6: II-2004-I-1009. A fragment (knife?). Fibrous texture can be observed on the surface of this green nephrite. 2.2 mm thick, weight 0.5 g.

A JEOL JSM-6360LV SEM equipped with an EDS (Oxford Instruments Ltd., INCA-300) was used. The analyzed points were selected on the relatively flat and well-polished surfaces under the electron image. Minerals were identified based on comparisons with the X-ray spectra of chemical-known minerals (see Appendix: Fig. A1). The chemical compositions of minerals greater than 1% by weight, both in the nephrite matrix and in the accessory mineral inclusions, were analyzed at the 1 μ m electron beam spot for 100 seconds. The quantitative data were corrected as oxide compositions, using the X-ray intensities of synthetic chemical-known minerals as standards; wollastonite for Si and Ca, corundum for Al, fayalite for Fe, and pyrope for Mg. The number of cations was calculated into the atoms per formula unit (a.p.f.u.) on the basis of 23 oxygen, and minerals were classified according to the nomenclature of amphiboles (Leake *et al.* 1997). The corrected data in the cation ratios are based on the ideal chemical formula of calcium amphibole, this being $\text{Ca}_2(\text{Mg,Fe})_5(\text{Si}_8\text{O}_{22})(\text{OH})_2$, where $\text{Mg}/(\text{Mg}+\text{Fe}) < 0.9$: actinolite; $0.9 >$ tremolite. All Fe is calculated as ferrous iron (Fe^{2+}) because ferric iron (Fe^{3+}) is rare in nephrite (Tan *et al.* 1978).

ANALYSIS RESULTS FOR NEPHRITE ARTIFACTS FROM ITBAYAT ISLAND

Surface observations:

Sample Btn-1 is brown to white (Fig. 2-a), Btn-2 is dark green to black (Fig. 2-b) and Btn-5 is white in color (Fig. 2-e). In samples Btn-3 (Fig. 2-c), 4 (Fig. 2-d) and 6 (Fig. 2-f), the color of the unweathered portion is mostly pale green to deep green, and weathered portions, mostly chlorite, are brown to gray in color. Most of the specimen surfaces were well polished by the ancient craftsmen and were no problem to analyze. Fibrous textures were observed on the surfaces of all specimens (Fig. 3). Cr-spinels (black in color) can be observed on the surfaces of specimens Btn-1, 3 and 4, as accessory minerals. The sizes of Cr-spinels range up to 50 μm .

Chemical compositions:

A representative X-ray spectrum by EDS from the matrix of nephrite (Btn-3) is shown in Figure 4. The results of the quantitative analysis of the six nephrite samples are also listed in Table 2 and plotted in Fig. 5. Regardless of color, the studied six specimens range from 0.92 to 0.83 in $\text{Mg}/[\text{Mg}+\text{Fe}]$ ratios, with the Si (a.p.f.u.) range being from 8.0 to 7.87. Thus, all samples are tremolite-actinolite and can be termed nephrite because of their fibrous textures. Their chemical ranges are comparable to those of Fengtian nephrite in Taiwan.

A representative EDS spectrum from Cr-spinel is shown in Fig 6. It shows strong intensities of Fe and Cr, and minor peak of a Zn. Based on the spectrum pattern, the accessory minerals can be identified Zn-chromite (Cr-spinel) (see Appendix A1(5)).

DISCUSSION

Nephrite artifacts are widely distributed in Neolithic sites in East and Southeast Asia. Characteristically Taiwan nephrite is green in color and was widely distributed in

Table 1. Nephrite artifacts from Itbayat Island, the Batanes, the Philippines

No.	collection No.	descriptions	color	texture*	mineral name	inclusions**
Btn-1	II-2004-I 444	fragment of ring	white- light brown/green	fibrous	tremolite-actinolite	Zn-chromite
Btn-2	II-2004-I 1014	worked piece	dark green	fibrous	tremolite-actinolite	no
Btn-3	II-2004-I 1013	barrel shaped core of <i>lingling-o</i>	green	fibrous	tremolite-actinolite	Zn-chromite
Btn-4	II-2004-I 1019	bullet shaped core of <i>lingling-o</i>	green	fibrous	tremolite-actinolite	Zn-chromite
Btn-5	II-2004-I 1015	fragment	light green- white	fibrous	tremolite-actinolite	no
Btn-6	II-2004-I-1009	fragment	green	fibrous	actinolite	no

*: surface texture was described by the SEM observation; **: inclusions were identified by EDS spectrum.

Table 2. Chemical compositions of nephritic artifacts.

sample No.	Btn-1				Btn-2								Btn-3				
analysis No.	1-1	1-2	2-2	2-3	1-1	1-2	1-4	5-1	5-2	5-3	5-4		2-3	2-5	1-5	1-9	1-10
wt%																	
SiO ₂	57.43	57.76	57.38	58.21	57.14	57.77	57.44	57.37	57.68	57.85	57.41		58.13	57.67	57.96	57.17	58.14
Al ₂ O ₃	0.26	0.06	0.19	0.19	0.01	0.04	0.23	0.34	0.08	0.41	0.25		0.13	0.25	0.04	0.48	0.15
FeO	4.64	4.04	3.56	3.78	5.18	4.08	4.03	3.66	3.71	3.58	3.45		3.75	3.63	3.65	4.36	3.75
MgO	22.35	22.64	23.76	21.20	20.56	20.81	22.05	23.83	23.25	22.74	23.55		23.33	23.54	23.42	23.46	23.47
CaO	13.38	13.42	12.38	14.81	16.48	16.04	14.32	11.89	12.87	12.71	12.60		12.00	12.12	12.36	11.84	11.74
Total	98.06	97.92	97.27	98.20	99.37	98.74	98.06	97.08	97.59	97.29	97.25		97.34	97.21	97.43	97.31	97.25
(O = 23)																	
Si	7.918	7.951	7.916	8.000	7.875	7.947	7.919	7.919	7.943	7.972	7.921		7.995	7.949	7.975	7.900	7.997
Al	0.043	0.009	0.030	0.030	0.001	0.007	0.037	0.055	0.013	0.066	0.040		0.021	0.041	0.007	0.078	0.025
Fe	0.535	0.465	0.411	0.434	0.597	0.469	0.464	0.423	0.427	0.413	0.398		0.431	0.418	0.419	0.503	0.431
Mg	4.589	4.642	4.882	4.341	4.220	4.265	4.529	4.899	4.768	4.669	4.839		4.780	4.833	4.799	4.829	4.809
Ca	1.976	1.978	1.830	2.180	2.433	2.362	2.114	1.757	1.898	1.876	1.861		1.767	1.790	1.821	1.752	1.729
cation total	15.061	15.045	15.069	14.985	15.125	15.050	15.063	15.053	15.050	14.995	15.059		14.994	15.031	15.022	15.061	14.990
Mg/(Mg+Fe)	0.896	0.909	0.922	0.909	0.876	0.901	0.907	0.921	0.918	0.919	0.924		0.917	0.920	0.920	0.906	0.918

sample No.	Btn-4						Btn-5						Btn-6					
analysis No.	2-2	2-4	2-5	1-4	b2-1	b2-2	a1-3	a2-1	s5-1	s5-2	s5-3	s5-4	4-2	4-3	4-4	4-5	3-2	3-3
wt%																		
SiO ₂	56.12	56.98	57.51	57.12	57.83	57.94	57.42	57.53	57.58	57.52	57.34	56.47	57.96	57.84	57.95	57.53	57.81	57.75
Al ₂ O ₃	0.60	0.74	0.53	0.62	0.65	0.57	0.79	1.81	0.78	0.65	1.37	4.61	0.68	0.67	0.74	0.51	0.69	0.26
FeO	8.73	6.39	7.24	5.96	4.79	3.48	4.44	5.03	3.86	4.54	4.36	4.84	5.91	4.87	4.84	4.98	5.03	5.00
MgO	18.29	21.26	19.28	21.88	22.10	22.89	22.50	19.91	21.83	22.67	21.86	17.98	21.22	22.15	21.66	22.44	21.53	21.65
CaO	17.08	13.11	15.11	12.65	12.24	12.07	12.11	12.96	13.33	11.99	11.90	10.94	12.38	12.02	12.45	12.28	12.80	13.67
Total	100.82	98.47	99.68	98.23	97.60	96.96	97.26	97.24	97.38	97.36	96.83	94.83	98.16	97.55	97.64	97.75	97.86	98.34
(O = 23)																		
Si	7.769	7.880	7.924	7.893	7.973	7.988	7.935	7.972	7.951	7.942	7.943	7.938	7.985	7.975	7.988	7.937	7.970	7.952
Al	0.098	0.121	0.086	0.101	0.106	0.093	0.129	0.295	0.127	0.106	0.223	0.763	0.111	0.109	0.121	0.082	0.112	0.043
Fe	1.010	0.738	0.834	0.688	0.552	0.401	0.513	0.583	0.446	0.524	0.504	0.568	0.680	0.561	0.557	0.575	0.580	0.576
Mg	3.771	4.379	3.958	4.503	4.538	4.701	4.632	4.108	4.490	4.661	4.511	3.764	4.355	4.549	4.448	4.612	4.422	4.441
Ca	2.533	1.941	2.230	1.871	1.807	1.782	1.792	1.923	1.972	1.772	1.765	1.647	1.827	1.776	1.837	1.815	1.889	2.015
cation total	15.182	15.059	15.033	15.057	14.974	14.965	15.001	14.881	14.985	15.005	14.946	14.680	14.959	14.970	14.951	15.021	14.974	15.027
Mg/(Mg+Fe)	0.789	0.856	0.826	0.867	0.892	0.921	0.900	0.876	0.910	0.899	0.899	0.869	0.865	0.890	0.889	0.889	0.884	0.885

Taiwan in prehistory (e.g., Kano 1946; Sung 1989; Chen 1996; Yui *et al.* 2001; Hung 2003, 2004). It is believed that the sources of Taiwan's green nephrite were in the Fengtian deposits in eastern Taiwan (Tan *et al.* 1978; Wang *et al.* 1996; Lien *et al.* 1996; Tan *et al.* 1997; Huang & Chou 2001; Lin *et al.* 2002; Hung 2003, 2004). Based on studies of mineral chemistry of nephrites from the Fengtian deposits and artifacts from archaeological excavations, it is apparent that ornaments of green nephrite found on Luzon and Palawan in the Philippines can be identified as Fengtian nephrite (Hung & Iizuka 2004; Hung *et al.* 2004; Iizuka & Hung 2005: *this issue*). Since Itbayat is an island formed around two basaltic volcanoes surrounded by uplifted coral reefs, it is clear that nephrite is an exotic mineral on this island.

Fengtian nephrite is composed of Fe-rich tremolite and actinolite (Mg/[Mg+Fe] ratios = 0.93-0.85) with Cr-spinel, which contains significant amounts of Mn (up to 9 wt.%)

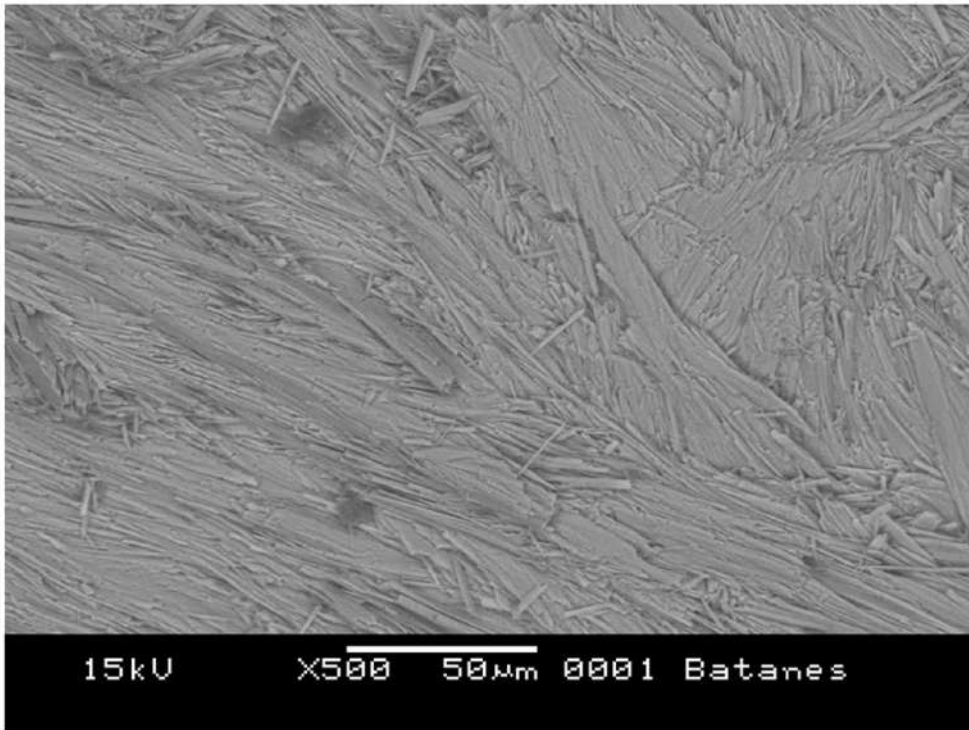


Fig. 3. Electron back-scattered photograph of the surface of nephrite artifact Btn-1: II-2004-I 444 (the large ring). Fibrous texture can be observed. Dark contrast portions are chlorite, which is identified by EDS. Scale bar is 50 μm .

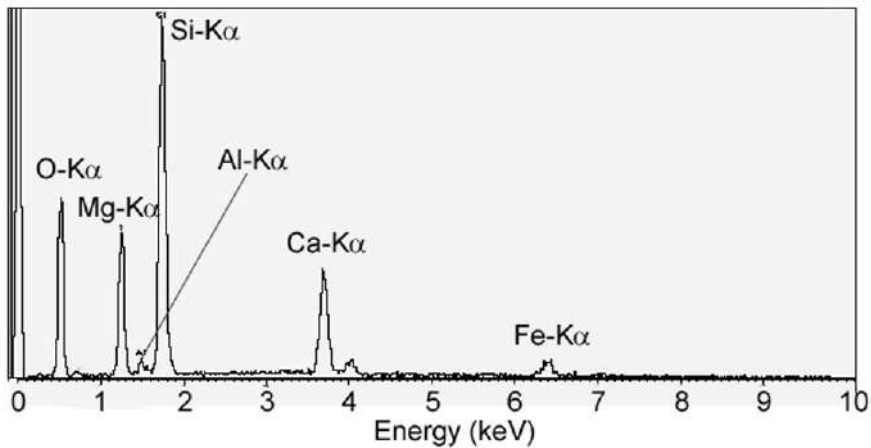


Fig. 4. A representative EDS spectrum from the surface of a nephrite artifact Btn-3: II-2004-I 1013 (the drilled core).

K α lines of oxygen (O), magnesium (Mg), aluminum (Al), silicon (Si), calcium (Ca), and iron (Fe) are identified from low-energy (left) to high-energy (right). The X- and Y-axes represent X-ray intensity and energy (keV), respectively.

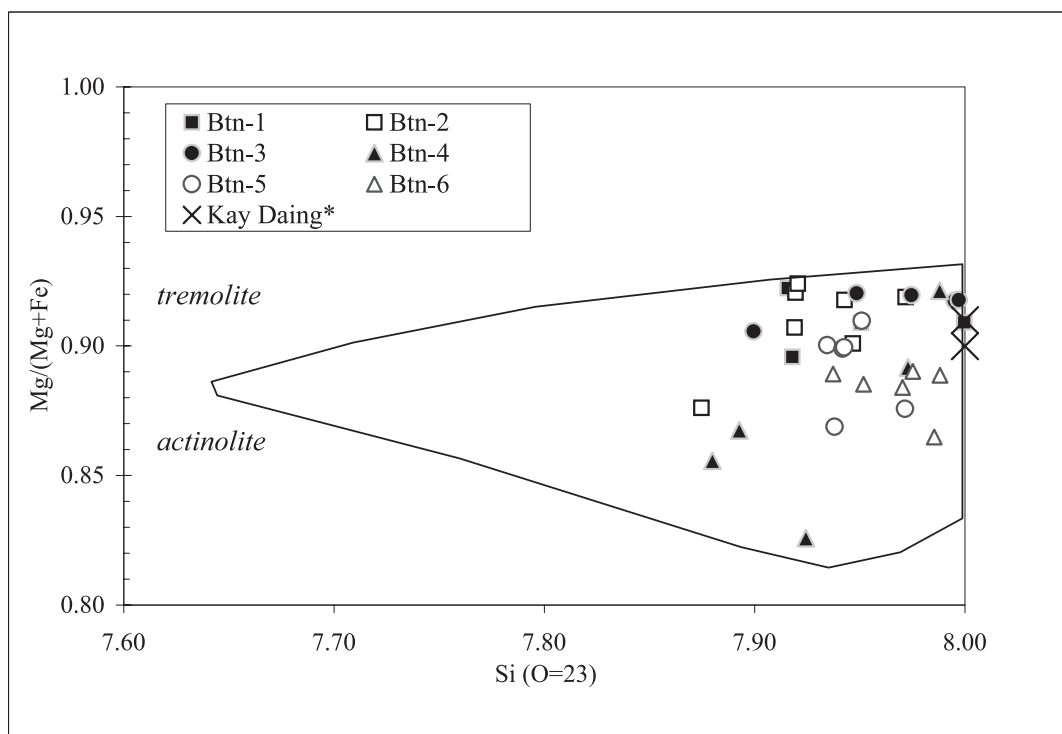


Fig. 5. Chemical compositions of nephrite artifacts from Itbayat Island.

Solid and open symbols represent nephrite with and without Zn-chromite, respectively. The chemical boundary between tremolite and actinolite is 0.90 in $\text{Mg}/[\text{Mg}+\text{Fe}]$ ratio (Leake *et al.* 1997). Analytical error is shown as the size of the symbol. The ranges for Fengtian nephrite are shown as an enclosed area (Iizuka & Hung 2005: *this issue*).*: Non-destructive analysis results of a bell-shaped bead from Kay Daing site in Batangas, southwestern Luzon (after Hung *et al.* 2004).

and Zn (up to 7 wt.%) as accessory mineral (Iizuka & Hung 2005: *this issue*). The analytical results show that the mineralogical characters of the six artifacts from Anaro are comparable to those of Fengtian nephrite (Fig. 5). It can be concluded, therefore, that the raw material of the analyzed nephrite artifacts was derived from Fengtian in eastern Taiwan.

As mentioned in Bellwood & Dizon (2005: *this issue*), a most remarkable discovery from the Anaro site was that of two drilled cores of green nephrite (samples Btn-3 and 4 in this report). A barrel-shaped Btn-3 appears to be a discard from drilling out the central hole in one of the thick *lingling-o* earrings that usually have three circumferential protuberances (see Iizuka & Hung 2005: Figs. 9-h and -i: *this issue*). A bullet-shaped Btn-4 is not so easily identified, but it looks like an early stage in the shaping of a possible projection, that broke from a core during manufacture. This type of thick earring with projections was called by Robert Fox (1970) the "Sa Huynh type *lingling-o*". Such earrings are noticeable amongst the green nephrite ornaments that belong to the Southeast Asian Metal Age and have been found around the South China Sea, particularly in Luda and Lanyu Islands (Taiwan), in the Tabon Caves on Palawan (Philippines), in Sa Huynh sites

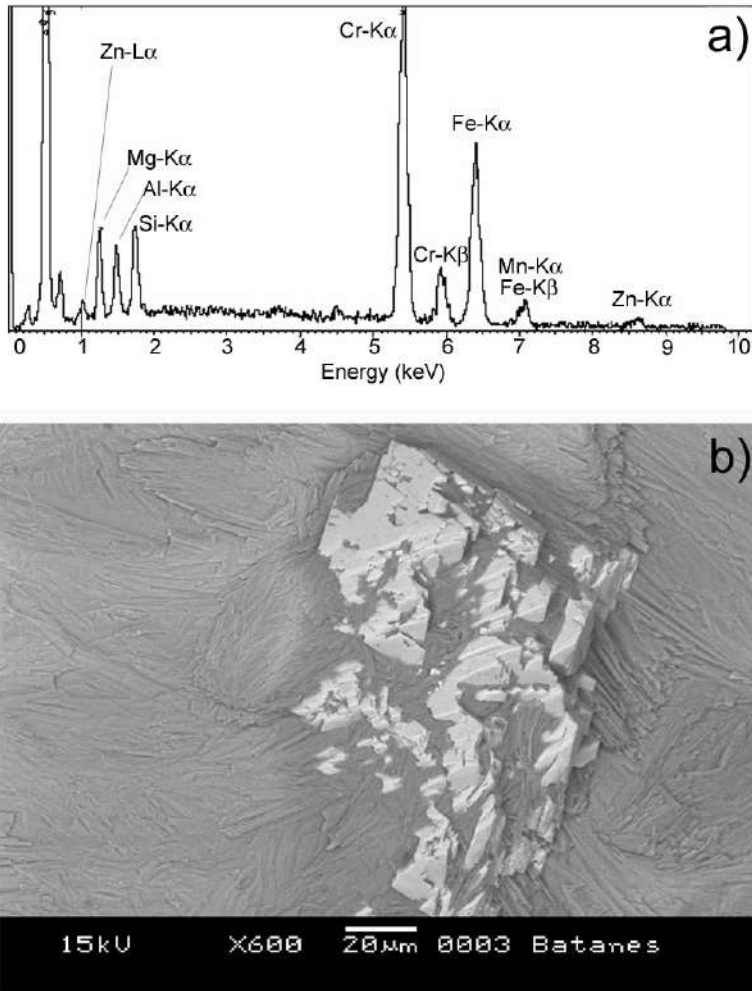


Fig. 6. Zn-chromite on nephrite artifact.

- (a) A representative EDS spectrum from Cr-spinel on the surface of nephrite artifact Btn-3: II-2004-I 1013 (the drilled core). $K\alpha$ lines of chromium (Cr), oxygen (O), iron (Fe), manganese (Mn) and zinc (Zn) are identified. The peaks of magnesium (Mg), aluminum (Al), silicon (Si), and calcium (Ca) are interferences from the nephrite matrix. The X-ray peaks of manganese are not clearly separated because the lines for the Mn $K\alpha$ and Mn $K\beta$ peaks are overlapped by the lines for Cr $K\beta$ and Fe $K\alpha$ respectively. However, both lines of Zn $K\alpha$ and $L\alpha$ are clearly marked in the Cr-spinel inclusions in Fengtian nephrite.
- (b) Electron back-scattered photograph of a Cr-spinel inclusion on the surface of a nephrite artifact (Btn-3: II-2004-I 1013). The white (high-contrast) part is a Zn-chromite inclusion. A fibrous texture is observed on the surface of this green nephrite. Scale bar is 20 μm .

in south-central Vietnam, and in Niah West Mouth in Sarawak (Malaysia) (Hung & Iizuka 2004). Although they have never found in Taiwan proper (Hung 2005: *this issue*), these sites are all related to the distribution of Austronesian-speaking people.

In addition to these two drilled cores, other pieces of worked Fengtian nephrite and pieces of slate (a meta-sedimentary rock), probably also from Taiwan, were found in Anaro. Given the quantity of new material recovered in the 2005 investigations (still to be analyzed), it is suggested that Anaro served from time to time as a nephrite workshop and that the raw materials were imported from eastern Taiwan, perhaps via Ludao and Lanyu. Some artifacts were perhaps made there for export onwards to Luzon and regions to the south. Although more detailed studies are needed on nephrite artifacts such as *lingling-o*, the mineralogical investigation reported here offers important information about cultural interaction between ancient Austronesian societies.

CONCLUSION

In a sourcing study of nephrite artifacts, it is very important to compare the chemical compositions of those artifacts with those of their expected original materials, without damage to precious artifacts. As demonstrated in this report, the non-destructive technique of LVSEM with EDS is useful not only for sample surface observation, but also for quantitative chemical analysis without conventional sample preparation, especially for precious artifacts.

The six analyzed artifacts were found to be composed of nephrite (tremolite-actinolite amphiboles) with Zinc bearing chromian-spinel as accessory minerals. The mineralogical characteristics of these nephrite artifacts are comparable to those of Fengtian (Taiwan) nephrite. The results suggest that the original materials for these artifacts were collected between 2500 and 1300 years ago in the Fengtian area in eastern Taiwan.

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APPENDIX: ELECTRON BEAM ANALYSIS

Principles of Mineral identification by SEM-EDS

The scanning electron microscope (SEM) performs the observation of material surfaces on sub-nanometer to micrometer scales. When the electron beam hits the surface of the specimen, imaging signals, such as secondary- and backscattered electrons, are emitted. Contrast of the secondary- and the backscattered electrons indicate difference of surface topography, and heterogeneity of the mean atomic weights (differences of chemical phases) of surface of specimens, respectively. At the same time, characteristic X-rays are also emitted as a result of the electron beam bombardment. The analysis of X-ray spectra and their intensities can yield both qualitative mineral identification and quantitative elemental analyses from a limited region of the specimen.

X-ray spectrometers which can be attached to a SEM are of two kinds; wavelength-dispersive spectrometers (WDS) equipped with diffracting crystals, and an energy-dispersive spectrometer (EDS). Both WDS and EDS techniques are well understood

(*cf.* Reed 1993, 1996; Goldstein *et al.* 2003). In general, accuracy of elemental analysis is better (RSD < 1%) with WDS equipment. This type is also known as EPMA (Electron Probe Micro Analyzer) or electron probe. On the other hand, EDS is quickly for qualitative analysis which is the identification of the elements that are present.

Common rock-forming minerals contain the elements of atomic numbers from 8 to 30 (Oxygen-Zinc) (*cf.* Railsback 2003). The K- and the L-spectra of these elements lie within the energy range 0.5-10 keV (kilo electron volt), and 1-2.5 keV, respectively. These energy ranges are commonly covered by standard EDS system. Mineral can be identified based on comparisons with the X-ray spectra of chemical-known minerals which indicate presence of elements and their X-ray intensities. EDS spectra of common rock-forming minerals, including *jade* and its related minerals, are shown in Fig. A1, as references of qualitative mineral identification.

Comparisons between LVSEM-EDS and WDS-EPMA

In order to verify the results of quantitative analysis using the LVSEM-EDS, a comparative study has been carried out. A sample of nephrite from the Fengtian source was analyzed by LVSEM-EDS without a carbon coat, and then re-analyzed by WDS-EPMA after carbon coating.

EDS (Oxford INCA-300) was operated with a 10 mm of the working distance and 35° X-ray take-off angle on SEM (JEOL JSM-6360LV) under the primary beam conditions of 15 kV acceleration voltage and 0.18 nA beam current at low-vacuum condition (25 Pascal). WDS-EPMA (JEOL JXA-8900R) was used with 15 kV acceleration voltage and 10 nA beam current under the vacuum condition of approx. 5×10^{-4} Pascal (Iizuka & Hung 2005: *this issue*). The number of cations was calculated into the atoms per formula unit (a.p.f.u.) on the basis of 23 oxygen, which is based on the ideal chemical formula of calcium amphibole, $\text{Ca}_2(\text{Mg,Fe})_5(\text{Si}_8\text{O}_{22})(\text{OH})_2$.

Results are shown in the Ca-amphibole (tremolite-actinolite) discrimination diagram (Fig. A2). For the same surface of each specimen, the results from both the LVSEM-EDS and the WDS-EPMA show similar variations in the plots of Si and the Mg/[Mg+Fe] ratio. Si (a.p.f.u.) ranges from 8.0 to 7.86 and from 8.0 to 7.87 in EDS and WDS, respectively. The Mg/[Mg+Fe] ratio ranges from 0.901 to 0.888 and from 0.918 to 0.871 in EDS and WDS, respectively. All overlap with the chemical ranges for the Fengtian nephrites, which are shown as an enclosed area. The result of quantitative analysis by LVSEM-EDS is thus consistent with the data from WDS-EPMA. Therefore, the non-destructive type of analysis can be used with confidence.

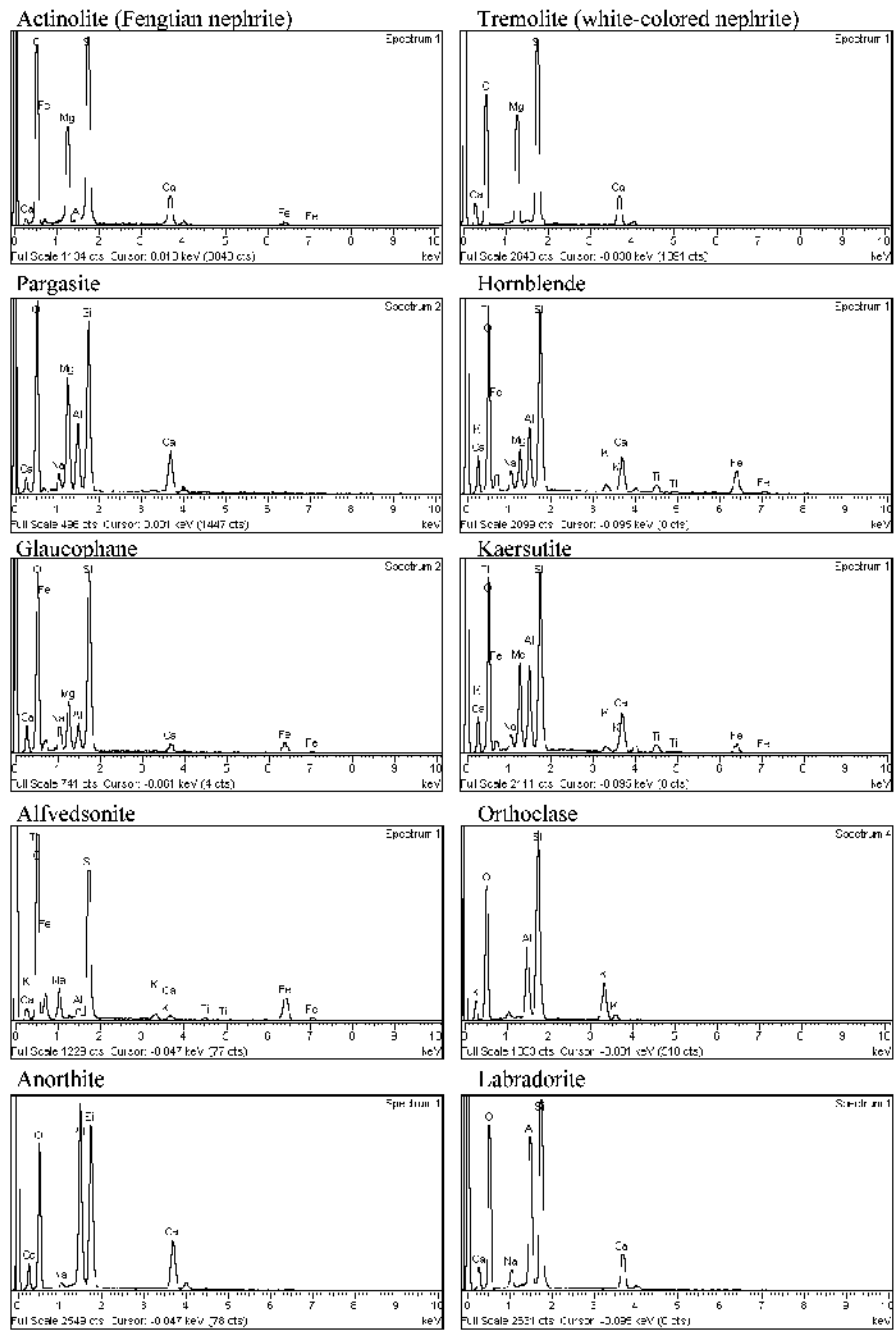


Fig. A1(1): EDS spectra of rock-forming minerals

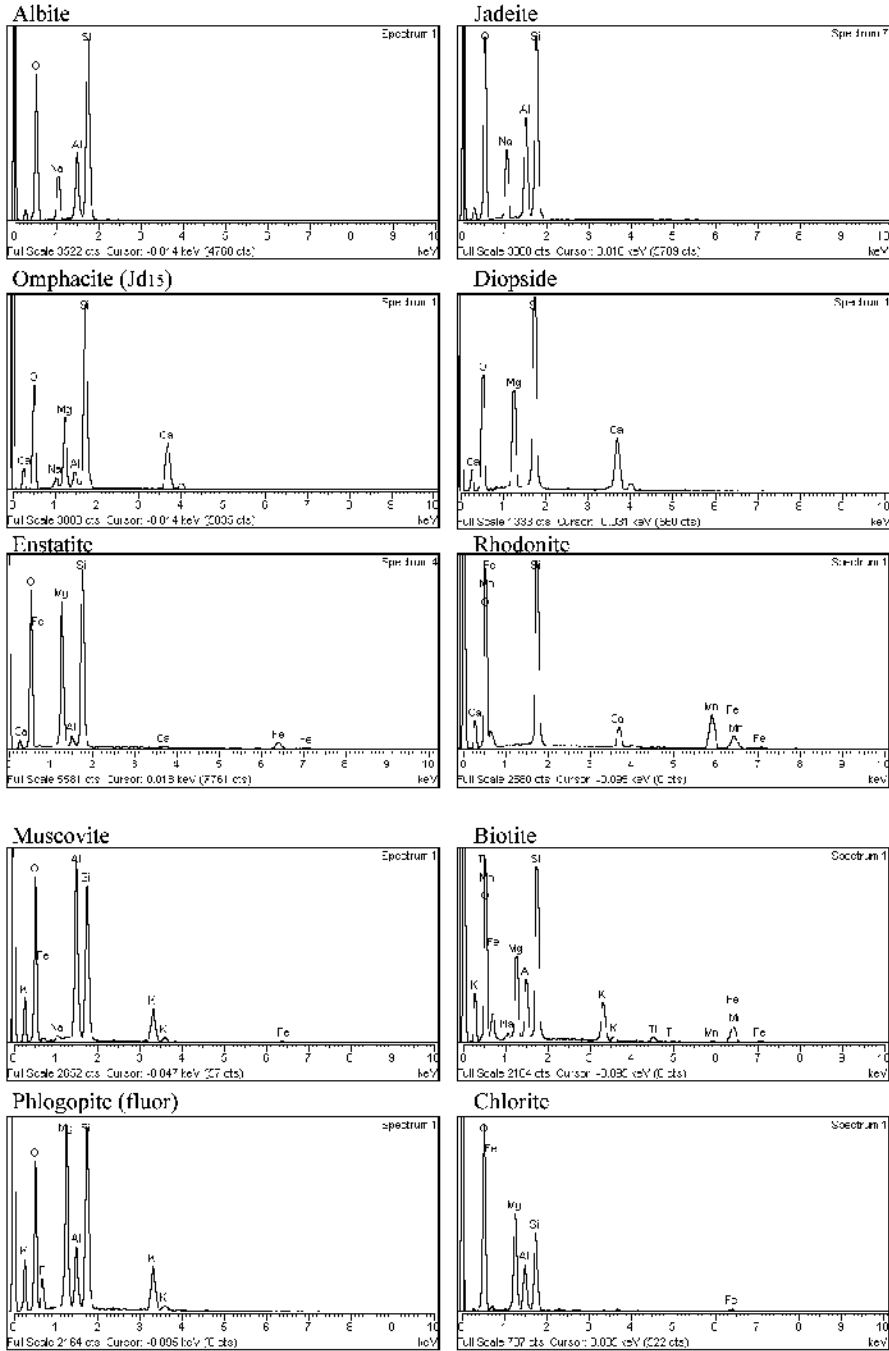


Fig. A1(2): EDS spectra of rock-forming minerals

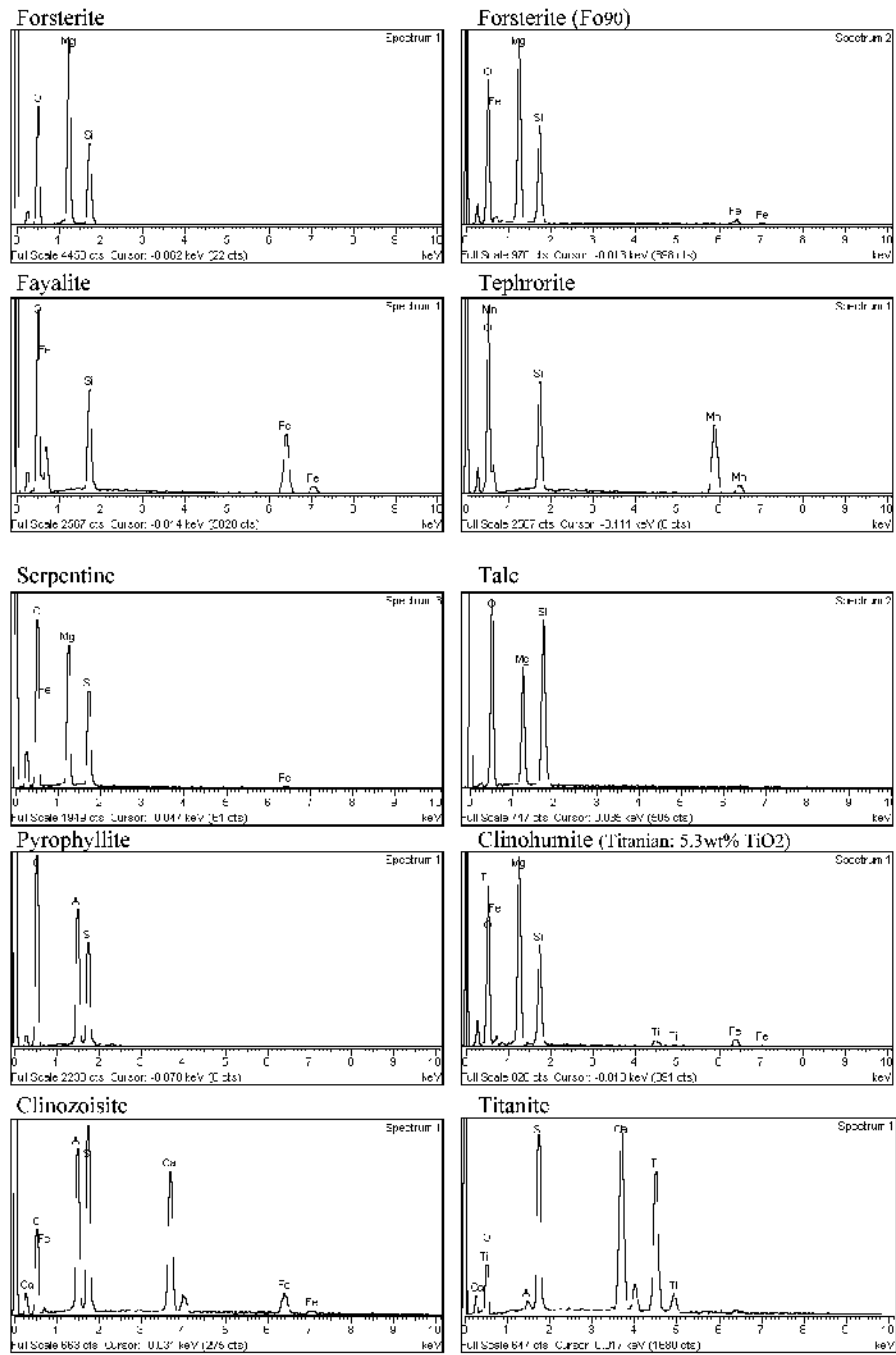


Fig. A1(3): EDS spectra of rock-forming minerals

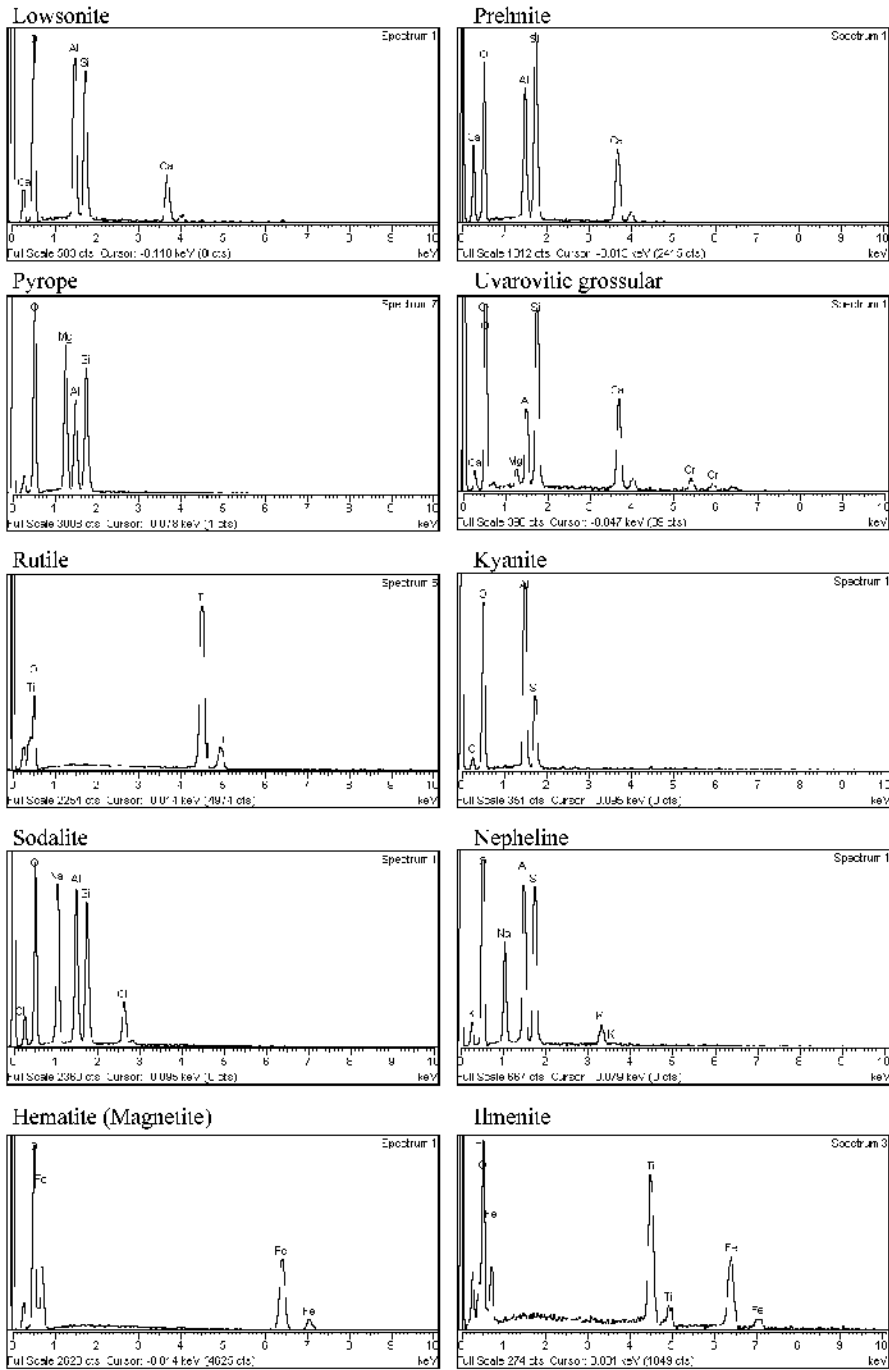


Fig. A1(4): EDS spectra of rock-forming minerals

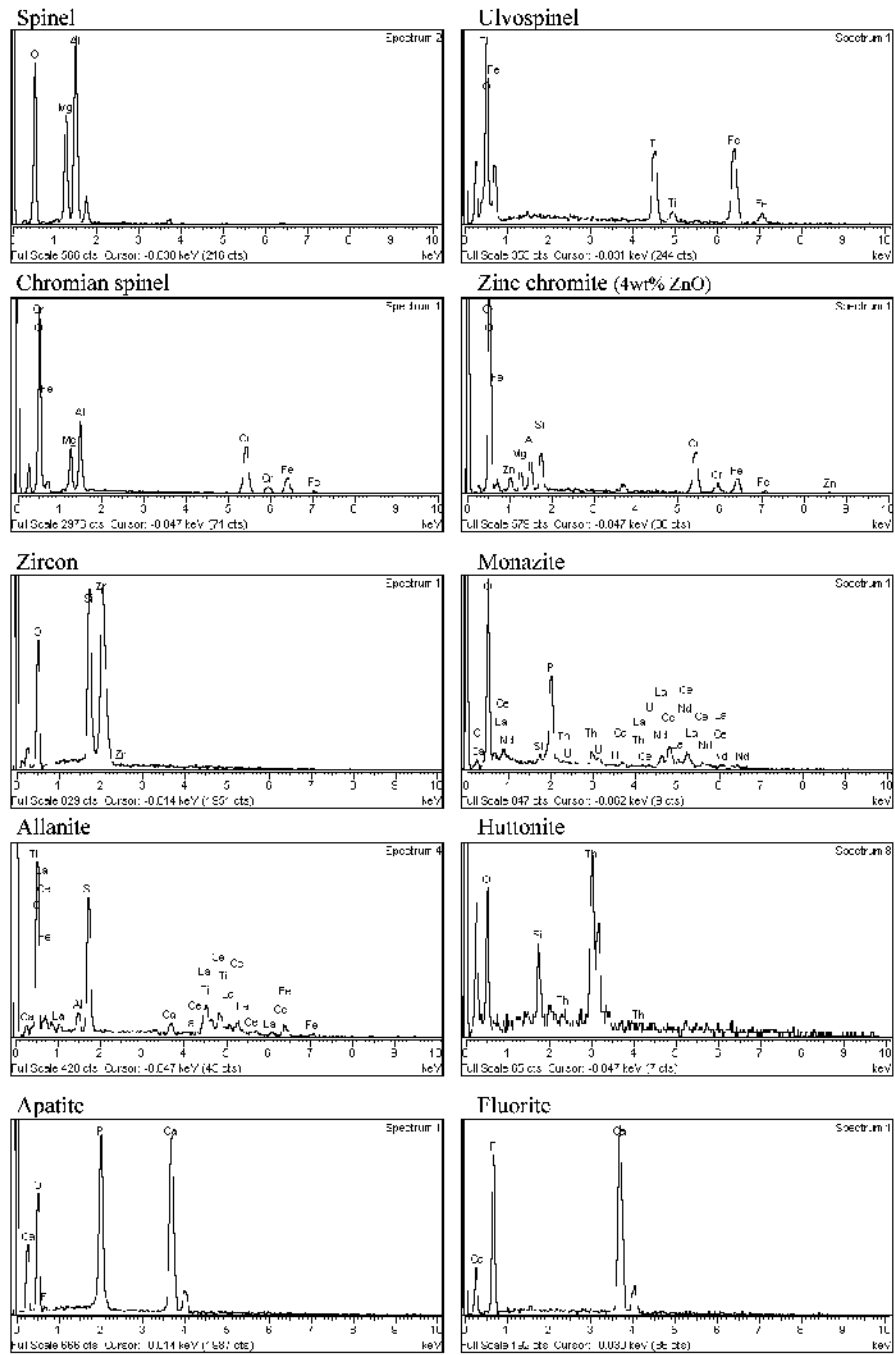


Fig. A1(5): EDS spectra of rock-forming minerals

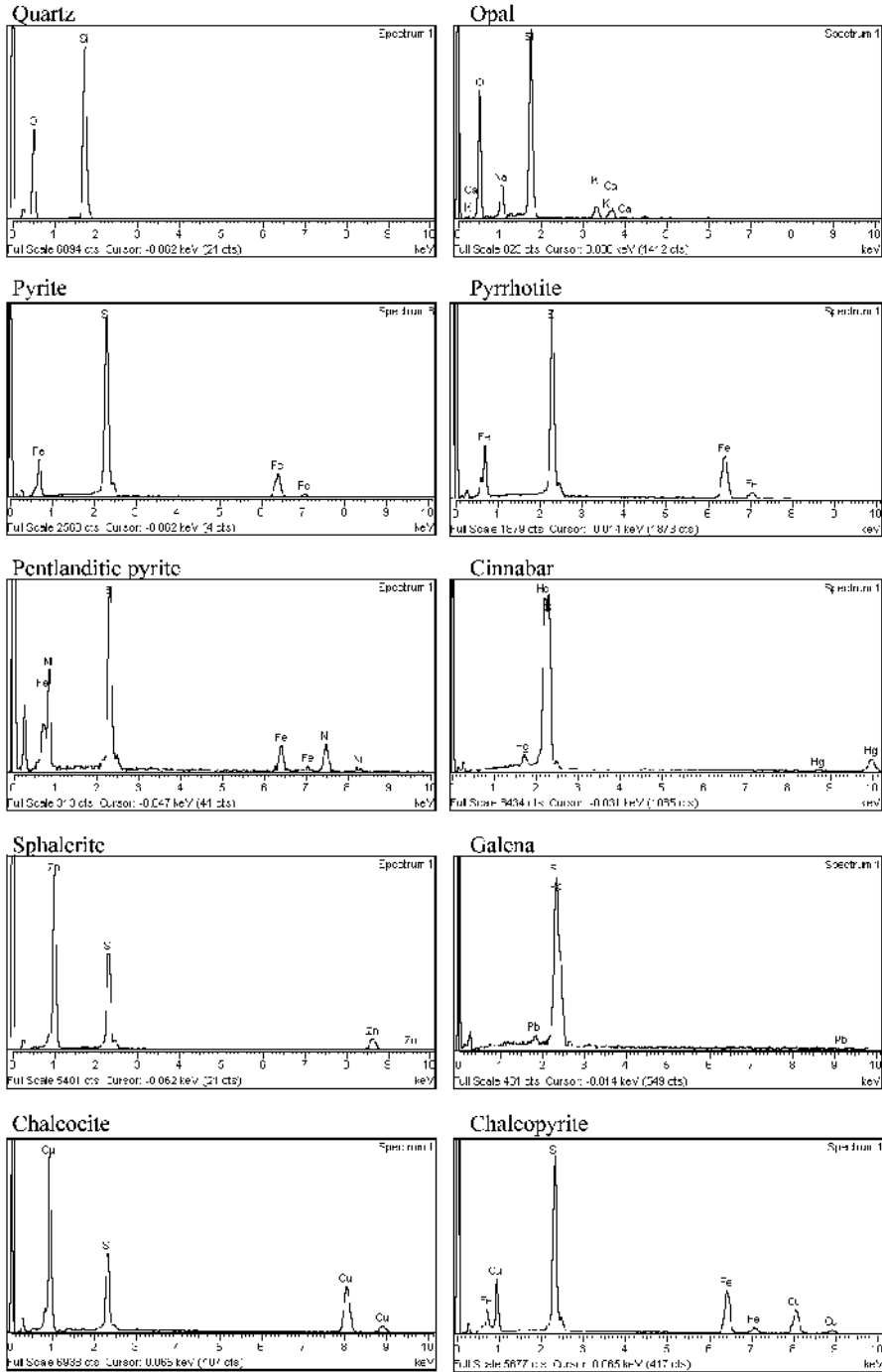


Fig. A1(6): EDS spectra of rock-forming minerals

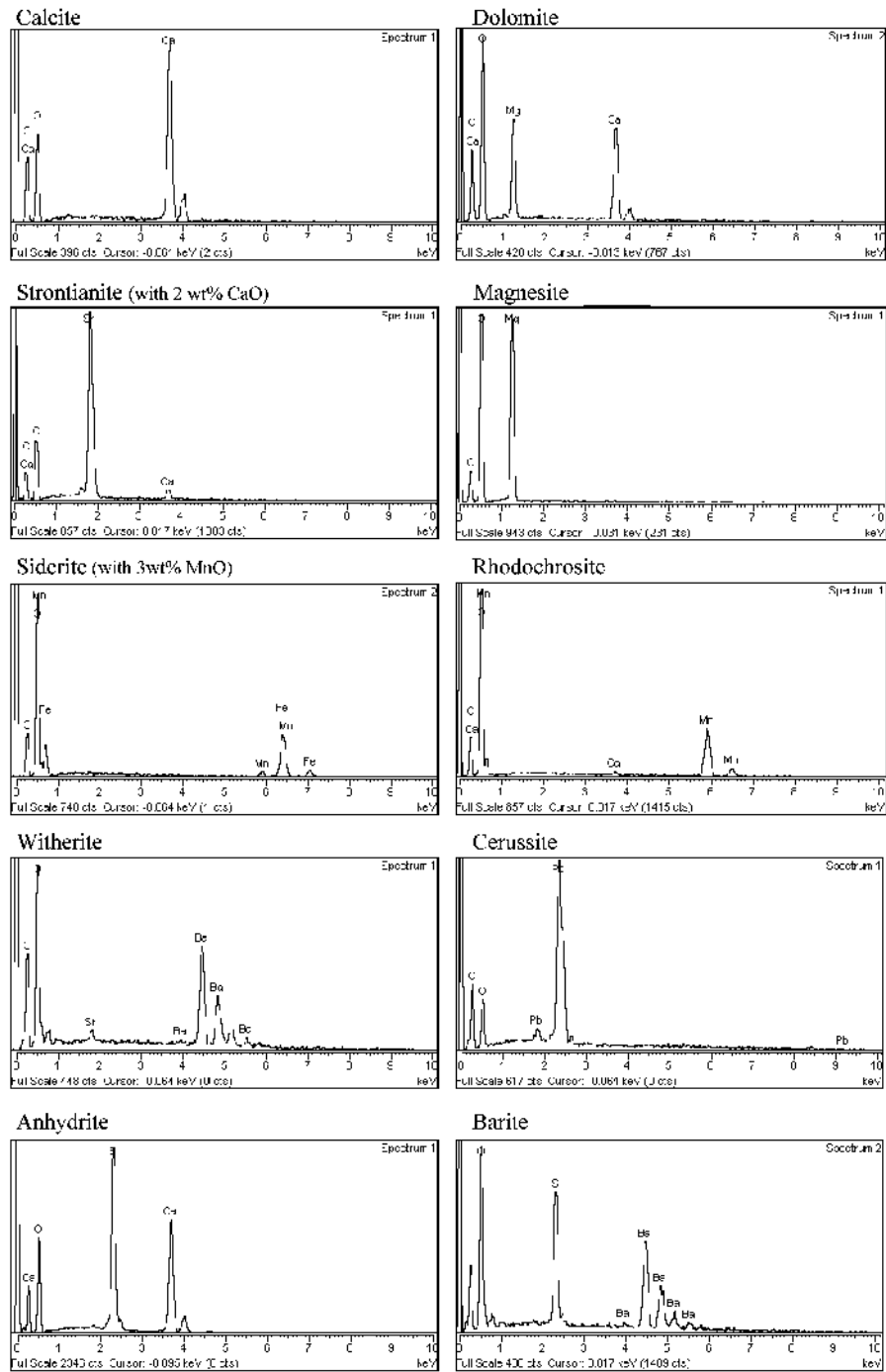
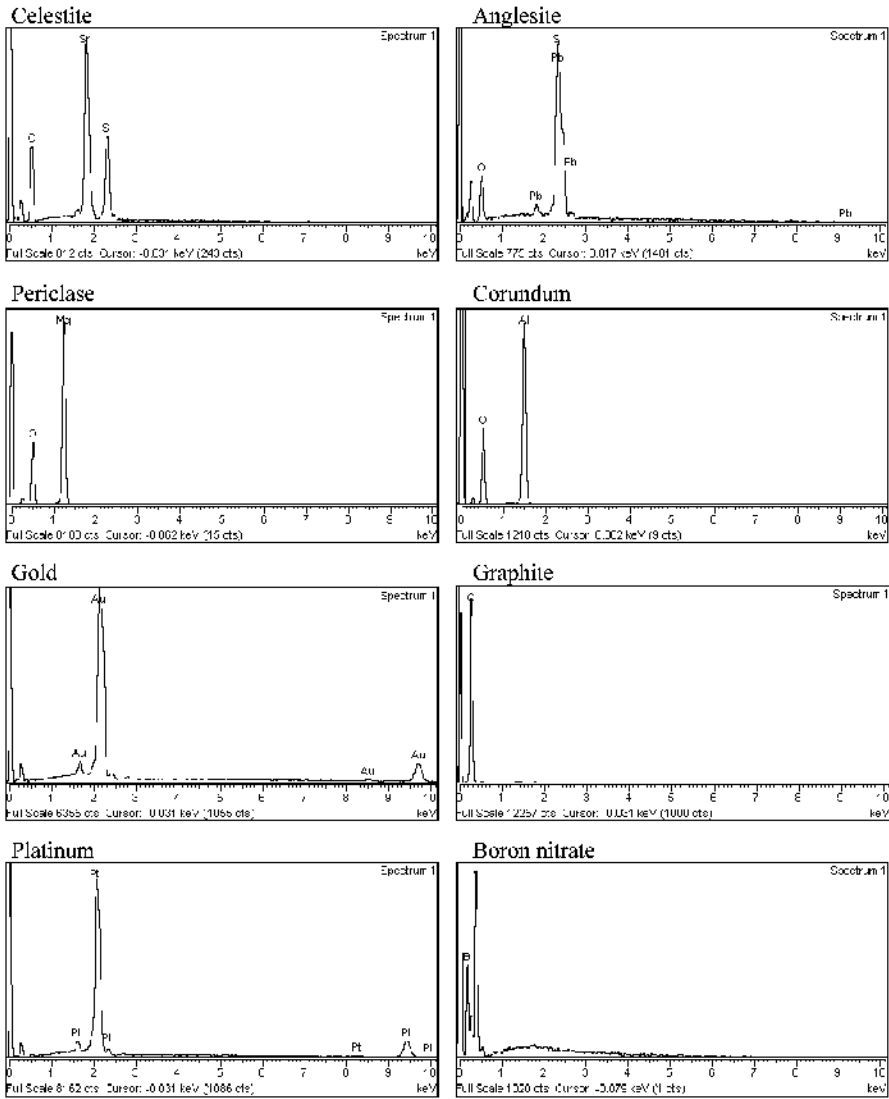


Fig. A1(7): EDS spectra of rock-forming minerals



FigA1(8): EDS spectra of rock-forming minerals

Fig. A1. EDS spectra of chemical-know minerals.

X- and Y-axes represent X-ray intensity and energy in kilo electron volt (keV), respectively. Analysis conditions (SEM: JEOL JSM-6360LV; EDS: Oxford INCA-300): 15 kV acceleration voltage, 0.18 nA beam current, approx. 1 μ m beam diameter, 50 sec. counting time, 25 Pa vacuum condition, using Si-Li detector with Be window and 0-10 keV energy band collection. Although all samples were not coated by carbon, a C-K α peak (at 0.277 keV) was observed from some samples but carbonates and graphite are excluded. It is caused by interference from epoxy resin, which was the sample mounting agent.

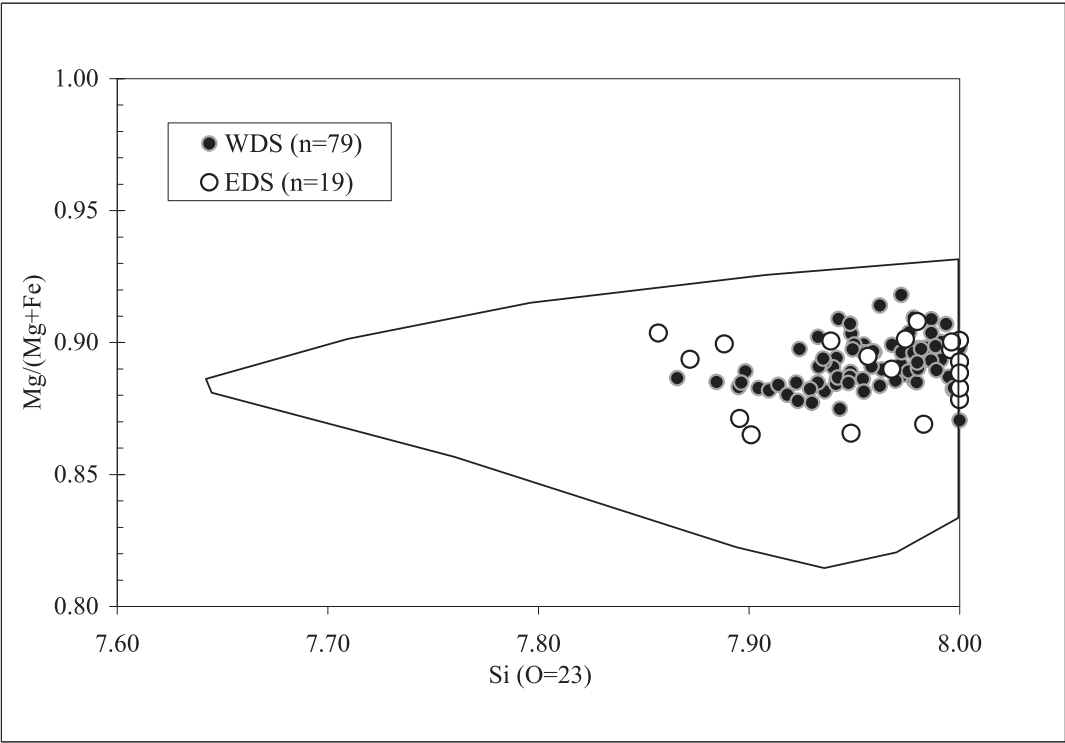


Fig. A2. The chemical composition of the internal standard of Fengtian nephrite. Solid and open symbols represent the results from WDS-EPMA and SEM-EDS, respectively. The chemical variations of Fengtian nephrites are shown as an enclosed area (Iizuka & Hung 2005: *this issue*).

菲律賓北部Batanes省Itbayat島出土玉器 之非破壞性礦物學研究

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本文利用低真空掃描電子顯微鏡（Low-Vacuum type Scanning Electron Microscope，簡稱LV-SEM）配備X射線光譜儀（Energy Dispersive X-ray Spectrometer，簡稱EDS）針對6件出土於菲律賓Anaro玉作坊遺址的玉器進行非破壞性的礦物分析，這處玉作坊的年代在距今2500年到1300年之間。分析結果發現這6件出土於Batanes省Itbayat島Anaro遺址的玉器都是軟玉，是由透閃石－陽起石所組成的角閃石礦物，並具有含鋅的鉻鐵礦包裹體。本文並將這6件玉器的礦物特徵與台灣東部豐田之玉料進行各方面的綜合性比較，分析結果證實這些出土於Itbayat島Anaro遺址的玉料都是來自於台灣東部的花蓮豐田地區。

關鍵字：非破壞性的礦物分析，X射線光譜儀，閃玉（軟玉），玉器，Itbayat島，Batanes群島，台灣。
