

Quantitative analysis results of CE-1 X-ray fluorescence spectrometer ground base experiment

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Abstract As the nearest celestial body to the earth, the moon has become a hot spot again in astronomy field recently. The element analysis is a much important subject in many lunar projects. Remote X-ray spectrometry plays an important role in the geochemical exploration of the solar bodies. Because of the quasi-vacuum atmosphere on the moon, which has no absorption of X-ray, the X-ray fluorescence analysis is an effective way to determine the elemental abundance of lunar surface. The CE-1 X-ray fluorescence spectrometer (CE-1/XFS) aims to map the major elemental compositions on the lunar surface. This paper describes a method for quantitative analysis of elemental compositions. A series of ground base experiments are done to examine the capability of XFS. The obtained results, which show a reasonable agreement with the certified values at a 30% uncertainty level for major elements, are presented.

Key words X-ray fluorescence, elemental abundance, lunar surface, quantitative analysis

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1 Introduction

Chemical compositions on planetary surfaces provide not only the geochemical information on the planetary surfaces but also the clues to the origins and evolutions of the celestial bodies. The remote X-ray fluorescence^[1–7] (XRF) detection has been proved to be a credible measure to derive the chemical compositions on the body's surface in many deep space missions^[5, 6].

To get a reliable result of the elemental abundances of the moon, the high quality orbital spectrums and careful ground-base calibrations are required. The Si-PIN detectors are adapted for XFS to get high energy resolution. And the fundamental parameters method (FP) will be used for quantitative analysis of the XFS data.

In this paper we briefly introduce the FP method. To examine the feasibility of the FP application, we carried out a series of Ground-base experiments, and applied the FP method to the rock samples, which were provided by the National Astronomical Observatories of China (NAOC). The results are in reason-

able agreement with their certified values.

2 Instrument and the FP method

The XFS is an instrument mounted on the CE-1 satellite, the first Chinese mission to the moon. It is composed of 21 Si-PIN sensors, where one is an X-ray solar monitor (XSM) with the energy band of 0.5–10 keV, four sensors are used for the low energy band of 1–10 keV (XLE) and the remaining sixteen are used for the high energy band of 10–60 keV (XHE). XSM is mounted towards the sun all the time for measuring the solar X-rays. Both XLE and XHE are mounted on the satellite with the entrance surface towards the lunar surface, so as to measure the fluorescence X-ray lines and the lunar diffuse background respectively. Both XSM and XLE have 125 μm thick beryllium windows, their energy resolutions are about 300 eV at 5.9 keV.

The intensity of the characteristic X-ray for a given element depends on the incident X-ray spectrum and the compositions of the surface matters. The elemental abundance can be deduced from the

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incident X-rays spectrum observed by XSM and the detected characteristic spectrum of the lunar surface by XLE with an approach called FP method^[8].

The samples were ground in the agate mortar and crushed into tablets. Providing the targets are several mm thick, the incident X-ray emitted by the X-ray tube is less than 8 keV, it is impossible for the incident X-ray to penetrate the samples. So we can adapt the following equations.

Based on the deduction of N. Broll, we can get the relationship between elemental concentration C_i and the relative intensity of the element's characteristic line R_i :

$$C_i = R_i(1 + \bar{\alpha}_{ij}C_j + \bar{\alpha}_{ik}C_k + \dots), \quad (1)$$

where α_{ij} refers to the influence of element j on the fluorescence intensity of element i . The effective coefficients in Eq. (1) are fixed for the standard sample and for samples which have composition values similar to the standard ones. Thus we can calculate the elemental abundance of an unknown sample by a standard sample whose chemical compositions are similar to the unknown ones.

In the FP method^[9], the X-ray fluorescence intensity of the primary X-rays induced process can be described as Fig. 1.

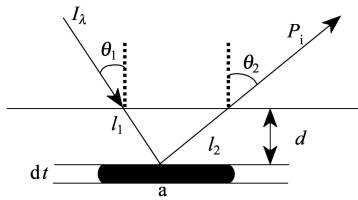


Fig. 1. The excited process of primary X-ray.

When the incident X-ray reaches the position a, its intensity is reduced to:

$$I_v = I_\lambda \exp(-\mu_{s,\lambda} \rho d / \cos \theta_1), \quad (2)$$

where $\mu_{s,\lambda}$ is the mass absorption of the sample for the X-ray whose wavelength is λ , and ρ is the density of the sample. Other variable terms are indicated in Fig. 1.

The primary X-ray intensity of element i at position a can be drawn from I_v :

$$I_c = I_v E_i C_i \mu_{i,\lambda} \rho dt / \cos \theta_1, \quad (3)$$

E_i is an excited factor composed of the absorption edge jump ratio, the transmission probability and the fluorescence yield.

As the primary X-ray of element i reaches the surface of the sample, its intensity becomes:

$$I_e = I_c \exp(-\mu_{s,\lambda_i} \rho d / \cos \theta_2) d\Omega / 4\pi. \quad (4)$$

The primary X-ray integral intensity of element i

is:

$$P_{i,\lambda} = E_i \frac{d\Omega}{4\pi \cos \theta_1} \frac{I_\lambda \mu_{i,\lambda}}{\mu_{s,\lambda} / \cos \theta_1 + \mu_{s,\lambda_i} / \cos \theta_2} C_i, \quad (5)$$

C_i is the weight fraction of element i in the sample.

For a pure sample:

$$P_{(i),\lambda} = E_i \frac{d\Omega}{4\pi \cos \theta_1} \frac{I_\lambda \mu_{i,\lambda}}{\mu_{i,\lambda} / \cos \theta_1 + \mu_{i,\lambda_i} / \cos \theta_2}. \quad (6)$$

The production process of secondary X-ray intensity of element i is indicated in Fig. 2.

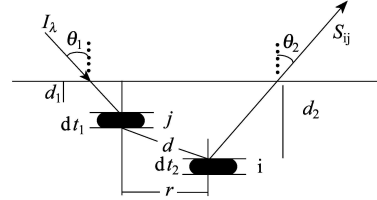


Fig. 2. The process of secondary fluorescence.

And the intensity of secondary fluorescence can be described as:

$$S_{ij,\lambda} = E_i \frac{d\Omega}{4\pi \cos \theta_1} C_i \frac{I_\lambda \mu_{i,\lambda}}{\mu_{s,\lambda} / \cos \theta_1 + \mu_{s,\lambda_i} / \cos \theta_2} e_{ij,\lambda} C_j = P_{i,\lambda} e_{ij,\lambda} C_j, \quad (7)$$

where

$$e_{ij,\lambda} = \left\{ \frac{1}{2} E_j \mu_{i,\lambda_j} (\mu_{j,\lambda} / \mu_{i,\lambda}) \times \left[\frac{1}{\mu_{s,\lambda} / \cos \theta_1} \ln \left(1 + \frac{\mu_{s,\lambda} / \cos \theta_1}{\mu_{s,\lambda_j}} \right) + \frac{1}{\mu_{s,\lambda_i} / \cos \theta_2} \ln \left(1 + \frac{\mu_{s,\lambda_i} / \cos \theta_2}{\mu_{s,\lambda_j}} \right) \right] \right\}_{\lambda_j}. \quad (8)$$

Considering the efficiency of the detector ε_i , the relative intensity of the peak can be written as:

$$R'_i = (P_{i,\lambda} + S_{ij,\lambda}) \varepsilon_i, \quad (9)$$

while for a pure elemental sample:

$$R'_{(i)} = P_{(i),\lambda} \varepsilon_i. \quad (10)$$

In the calculation, some fundamental parameters such as the fluorescence yield, the mass attenuation cross section, the transition probability and the jump ratio are adopted to calculate the fluorescence yield of the sample.

As we can see from the above equations, the term $E_i \frac{d\Omega}{4\pi \cos \theta_1}$ is an element and sample independent constant, called instrument constant, while $P_{i,\lambda}$ and $S_{ij,\lambda}$ are both relative to the abundance of the element i and the incident X-ray.

For polychromatic source, we can get the total intensity by the integral calculation in the incident X-ray interval.

The relative intensity of characteristic line of element i is:

$$R_i = \frac{I_i}{I_{(i)}} = \left(\frac{I_i}{I_{i,s}} \right)_{ms} \times \left(\frac{I_{i,s}}{I_{(i)}} \right)_{cal}, \quad (11)$$

$$I_i = P_i + \sum_j S_{ij}, \quad (12)$$

where I_i is the intensity of element i 's characteristic line in the unknown samples. $I_{(i)}$ is the calculated intensity of pure element i . $I_{i,s}$ is the intensity of element i 's line in the standard samples. Footprint ms indicates a value measured, while the footprint cal denotes the calculated value.

We adopted an iterative process to get the elemental concentrations of the unknown samples:

Assume that the initial value of i th elemental abundance C_i equals R_i , and set C_i as the input data for Eq. (12) to calculate the relative intensity R_i . Compare the calculated value R_i^{cal} with the measured value R_i^m , then correct the C_i with Eq. (13)

$$C'_i = (R_i^m / R_i^{cal}) \times C_i, \quad (13)$$

C_i is the value from the last iterative, and C'_i is the value after correction. The iterative process will continue until it comes to the convergence of C_i .

3 The ground base experiment

The XLE of the CE-1/XFS have been used for fluorescence analysis in the ground base experiments. The fluorescence generated by the X-ray beam bombarding the target, was collected by the XFS. Fig. 3 shows the schematic diagram of the experiment. The certified basalt (GB-07105) and amphibolite (GB-07122) were chosen as the standard samples. The unknown samples were provided by the NAOC, their elemental concentrations were certified by the Institute of Geochemistry Chinese Academy of Sciences.

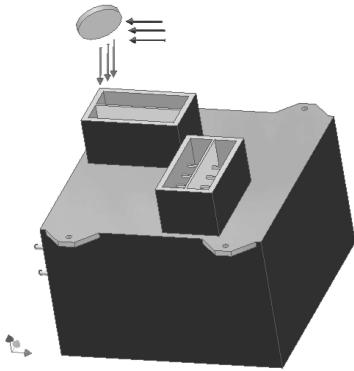


Fig. 3. The relative position of instruments.

It is very difficult to distinguish the peaks of the elements Mg, Al and Si, when a sample contains all these elements, because their characteristic lines' energies are very close and the energy resolutions of our

detectors are limited. Therefore we have to use multi-gaussian fit to draw the peak area of each element, to get the parameters of gaussian-fit, we have calibrated XLE with pure elemental samples excited by the X-ray tube. Table 1 lists the peak positions and FWHMs of pure elemental samples, derived from the calibration experiments described above. An accumulated spectrum analyzed by this way for a standard sample was shown in Fig. 4.

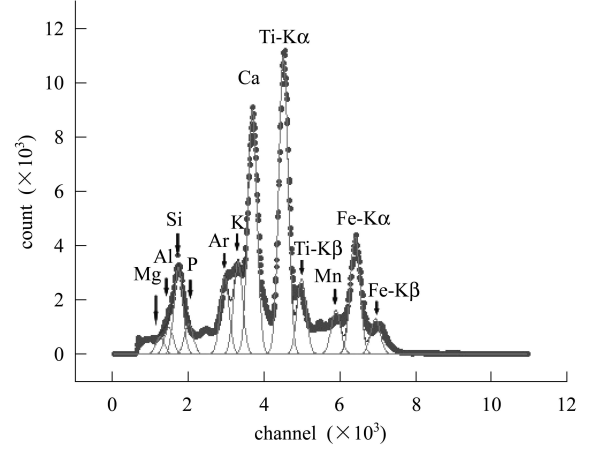


Fig. 4. The Gaussian fit spectrum of sample by the parameters in Table 1.

Table 1. The peak positions and FWHMs for elements in waveband 1–10 keV for one of the XLE.

element	characteristic K- α line/eV	peak position (ch)	FWHM (ch)
Mg	1253.60	111.61	25.61
Al	1486.70	132.83	25.72
Si	1739.98	155.89	25.83
P	2013.70	180.80	25.96
Cl	2622.39	236.21	26.23
Ar	2957.00	266.67	26.38
K	3313.80	299.15	26.54
Ca	3691.68	333.55	26.70
Ti	4510.00	408.05	27.06
Cr	5414.72	490.41	27.45
Mn	5898.00	534.40	27.65
Fe	6403.84	580.45	27.87
Ni	7478.15	678.25	28.31
Cu	8047.78	730.10	28.55
Zn	8638.86	783.91	28.79

By multi-gaussian fit to the spectrums of sample compositions, the intensities of the peak areas can be derived.

4 Results

We have developed a program to complete the iterative process mentioned above. Then the elemental concentrations of unknown samples can be derived by comparing the characteristic peak areas with the standard ones. The comparisons between the certified values and measured values are presented in Table 2.

Table 2. The comparison of the certified value and the calculated value of three unknown samples (where the C denotes the certified value, and M denotes the calculated value).

	sample1-C	sample1-M	sample2-C	sample2-M	sample3-C	sample3-M
Mg	4.27	4.91	4.221	3.88	3.18	4.11
Al	6.25	6.56	6.70	7.94	9.58	8.58
Si	18.44	20.78	21.85	22.24	22.32	23.01
P	0.28	0.38	0.09	0.41	0.19	0.38
K	1.11	1.29	0.34	0.73	1.20	1.50
Ca	4.59	5.34	6.84	8.72	5.34	6.51
Ti	5.39	5.23	3.07	3.44	1.19	1.29
Mn	0.16	0.13	0.15	0.14	0.10	0.12
Fe	12.22	11.27	10.58	7.62	7.83	9.48

As shown in the table, the calculated values of major elements are in agreement with the certified ones at a 30% uncertainty level except some minor components, which proves that the FP method is a reliable way to derive the quantity of the major chemical compositions of the samples on ground for our instrument.

5 Discussion

For the minor element phosphorus, its calculated values are much larger than the certified ones. This may be imputed to the small concentrations of the element phosphorus in the three samples, which means poor peaks to backgrounds ratios. To obtain more accurate concentrations of this element, we should nar-

row the exciting energy just above the absorbing edge of phosphorus to increase the intensity of its intensity. However as the concentrations of element phosphorus doesn't concern us, no further experiment is done.

As mentioned above, to get the quantitative results with the FP method, at least one standard sample is required. And the chemical compositions of the chosen standard samples should resemble the compositions of the unknown samples. It is impossible to realize in the CE-1 mission. To apply this method in this mission, we will do further work in simulating different lunar rock samples illuminated by different class of solar flare, using a software called Geant-4^[10]. Comparing the simulation spectrums to the similar spectrums accumulated by the XLE on the track, the elemental concentrations of the soil on the moon surface can be derived.

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