Spectrochimica Acta Part B 122 (2016) 127–131

Contents lists available at ScienceDirect

Spectrochimica Acta Part B

journal homepage: www.elsevier.com/locate/sab

The surface morphology analysis based on progressive approximation method using confocal three-dimensional micro X-ray fluorescence

Longtao Yi, Tianxi Sun, Kai Wang, Min Qin, Kui Yang, Jinbang Wang, Zhiguo Liu *

[a] The Key Laboratory of Beam Technology and Material Modification of the Ministry of Education, Beijing Normal University, Beijing 100875, China
[b] College of Nuclear Science and Technology, Beijing Normal University, Beijing 100875, China

ARTICLE INFO

Article history:
Received 29 October 2015
30 May 2016
Accepted 8 June 2016
Available online 9 June 2016

Keywords:
Confocal 3D MXRF
Surface topography
Progressive approximation method
Surface adaptive

ABSTRACT

Confocal three-dimensional micro X-ray fluorescence (3D MXRF) is an excellent surface analysis technology. For a confocal structure, only the X-rays from the confocal volume can be detected. Confocal 3D MXRF has been widely used for analysing elements, the distribution of elements and 3D image of some special samples. However, it has rarely been applied to analysing surface topography by surface scanning. In this paper, a confocal 3D MXRF technology based on polycapillary X-ray optics was proposed for determining surface topography. A corresponding surface adaptive algorithm based on a progressive approximation method was designed to obtain surface topography. The surface topography of the letter "R" on a coin of the People's Republic of China and a small pit on painted pottery were obtained. The surface topography of the "R" and the pit are clearly shown in the two figures. Compared with the method in our previous study, it exhibits a higher scanning efficiency. This approach could be used for two-dimensional (2D) elemental mapping or 3D elemental voxel mapping measurements as an auxiliary method. It also could be used for analysing elemental mapping while obtaining the surface topography of a sample in 2D elemental mapping measurement.

1. Introduction

Confocal three-dimensional micro X-ray fluorescence (3D MXRF) based on the polycapillary X-ray lens [1] is an excellent surface analysis technology. It has been widely used in the biological sciences, materials science, environmental science, archaeology [2–6], and other fields. The principle of 3D MXRF combined with two individual polycapillary X-ray lenses was proposed by Gibson and Kumakhov [7]. By using polycapillary X-ray lenses in both the excitation and detection channels, a confocal volume could be obtained. Because the polycapillary X-ray lens limits the viewing area of the detector, radiation outside of the volume and scattered radiation were shielded from the detector. Therefore, only radiation from the confocal volume could be detected.

In 2003, Kanngießer and Malzer [8] built the first confocal 3D MXRF setup at BESSY to analyse an ancient painting. In 2005, Kanngieger et al. [9] demonstrated the possibility of using a tabletop confocal 3D MXRF setup to study a painting nondestructively in the depth direction. Then, Tsuji and Nakano [10] used confocal 3D MXRF to analyse the 3D elemental mapping of K in an amaranth seed. Reiche et al. [11] applied confocal 3D MXRF analysis to the Portraits of Famous Men series. Patterson et al. [12] used confocal 3D MXRF to obtain a 3D image of a machined aerogel tube through a three-dimension voxel scanning. In the 3D image, the surface morphology of the machined aerogel tube was clearly shown. The above works applied confocal 3D MXRF to obtaining elemental depth profiles, elemental mapping or 3D image. However, none of them applied this technology to analysing surface morphology by surface scanning. Although there many other technologies can be applied to characterizing the surface topography of a sample, such as electron microscopy, scanning tunnelling microscopy, optical microscopy, and others, the above technology requires certain conditions and cannot be applied to in situ XRF analysis. In our previous studies [13,14], surface morphology analysis methods based on confocal 3D MXRF was established that could analyse the sample surface morphology without any preprocessing. However, the scanning efficiency was not ideal.

In this work, a surface morphology analysis method based on a progressive approximation method was proposed. With this method, the surface topographies of the letter "R" on a coin of the People's Republic of China and a small pit on a piece of painted pottery were obtained.

2. Experimental

2.1. Instrumentation

Fig. 1 (a) shows the confocal 3D MXRF setup. An X-ray source with a Mo target [XTG UltraBright Microfocus X-ray Source, Oxford, USA] was operated at 20 kV and 0.5 mA. The focal spot size of the X-ray tube

* Corresponding author.

http://dx.doi.org/10.1016/j.sab.2016.06.002
0584-8547/

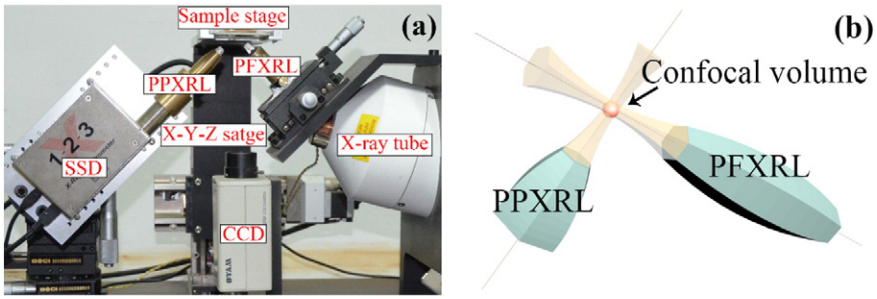

Fig. 1. (a) Experimental setup of the confocal 3D MXRF. (b) Schematic diagram of the confocal geometry.

was 16.45 μm at 20 kV. The polycapillary full X-ray lens (PFXRL) for X-ray irradiation was attached to the X-ray tube, and a polycapillary parallel X-ray lens (PPXRL) was attached to a silicon drift detector (*Si*-PIN) [X-123, Amptek, USA] (sensitive area: 6 mm^2, energy resolution: 145 eV at 5.9 keV). Each polycapillary lens was designed and manufactured by the Key Laboratory of Beam Technology and Material Modification of the Ministry of Education, Beijing Normal University. The input focal distance and output focal distance of the full lens were 52.9 mm and 11.5 mm, respectively. The focal distance of the half lens was 14.9 mm. The sizes of the focal spot of the half lens and full lens were experimentally determined to be 33 μm and 32.4 μm, respectively, at an X-ray energy of 17.4 keV (Mo Kα). Both polycapillary X-ray lenses were positioned in the confocal geometry, as shown in Fig. 1 (b). The micro volume overlaps the output focal spot of the PFXRL and the input focal spot of the PPXRL. The angle between the incident and detection beams was set to 90°. The sample stage was installed on an X–Y–Z stage [DS102 Series, SURUGA SEIKI, Japan], and the corresponding step resolution was 0.5 μm of each axis. A CCD camera was also used to confirm the relative position between the sample and the confocal volume.

2.2. Depth resolution

The depth resolution of the confocal 3D MXRF setup was evaluated by scanning a thin nickel film, prepared by electronic sputtering. The thickness of the film was 200 nm. The film was set on the sample stage and scanned perpendicularly to the surface of the film under a confocal set-up. The minimum step size was 2 μm, and the measurement time was 120 s per step. The depth resolution S can be estimated by the following equation [15]:

$$S = \sqrt{\left(S_F{}^2 - D_W{}^2\right)} \tag{1}$$

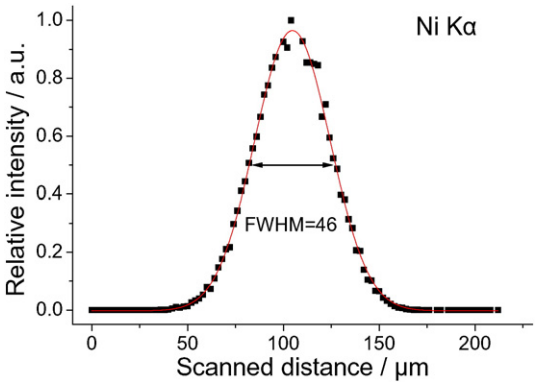

Fig. 2. Relative intensity curve measured for the thin film of Ni by the confocal 3D MXRF setup.

where S_F is the full width at half maximum (FWHM) value of the measured XRF intensity, and D_W is the thickness of the film.

Fig. 2 shows the measured XRF intensities of Ni Kα measured for the nickel film by the confocal 3D MXRF setup. The value of the FWHM is clearly shown in Fig. 2. Because the thickness is much smaller than the FWHM, the depth resolution can be denoted by it directly.

2.3. Theory and scanning process

The intensity distribution of the confocal volume fitted a Gaussian distribution. Its intensity along the z axis I(z) could be written in the following form:

$$\mathrm{I}(z) = ae^{\frac{-(z-z_0)^2}{2\sigma^2}} \tag{2}$$

where z_0 was the center of the confocal volume on the Z axis, and a and σ were the parameters of the confocal volume.

In the actual scanning process, only part of the confocal volume would be passed into the sample. Assuming that the sample is a homogeneous material, the absorption and excitation effects were not taken into account. The detected characteristic X-ray of the sample C(z) can be expressed as the integral of the intensity distribution on the Z axis and can be written in the following form:

$$\mathrm{C}(z) = \int_b^z \mathrm{I}(z)dz, \quad b<z<z_0 \tag{3}$$

where b was the lower boundary of the confocal volume.

By moving the confocal volume up and down, the detected characteristic X-ray of the sample will be changed. By observing the change in C(z), the sample surface can be determined. To maintain the confocal state, the sample instead of the confocal volume will be moved in the scan. By moving the sample, the confocal volume can be displaced perpendicular to the sample surface.

The flow chart of the scanning process is shown in Fig. 3. Initially, the scanning area, starting height Z_{start}, scanning range along Z-axis Z_{rang} and threshold value of XRF intensity $C_{\mathrm{threshold}}$ should be set manually. Then, the sample will be moved to two positions z_1 and z_2. The following relationships hold:

$$z_1 = Z_{start} - \frac{Z_{rang}}{2} \tag{4}$$

$$z_2 = Z_{start} + \frac{Z_{rang}}{2} \tag{5}$$

The XRF counts at the two positions can be denoted by C(z_1) and C(z_2). If the following conditions are satisfied:

$$\mathrm{C}(z_1)<C_{\mathrm{threshold}}<\mathrm{C}(z_2) \tag{6}$$