Nuclear Instruments and Methods in Physics Research B 267 (2009) 171–174

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Short Communication

Identification of origin of single aerosol particles using polycapillary X-ray lens

Tianxi Sun [a,b,c,*], Zhiguo Liu [a,b,c], Guanghua Zhu [a,b,c], Hui Liu [a,b,c], Qing Xu [d], Yude Li [a,b,c], Guangpu Wang [a,b,c], Hongbo Sun [a,b,c], Ping Luo [a,b,c], Qiuli Pan [a,b,c], Xunliang Ding [a,b,c]

[a] The Key Laboratory of Beam Technology and Materials Modification of Ministry of Education, Beijing Normal University, Beijing 100875, China
[b] Institute of Low Energy Nuclear Physics, Beijing Normal University, Beijing 100875, China
[c] Beijing Radiation Center, Beijing 100875, China
[d] Institute of High Energy Physics, Chinese Academy of Science, Beijing 100039, China

ARTICLE INFO

Article history:
Received 28 July 2008
Received in revised form 9 November 2008
Available online 3 December 2008

PACS:
78.70.En
41.50.th
91.67.gp

Keywords:
Micro-XRF
Capillary X-ray optics
Single aerosol particle analysis

ABSTRACT

A micro X-ray fluorescence (Micro-XRF) spectrometer based on a polycapillary focusing X-ray lens (PFXRL) and a laboratory X-ray source was designed to carry out the XRF analysis of single aerosol particles. The minimum detection limit (MDL) of this Micro-XRF spectrometer was 9 ppm for the Fe-Kα. The percentage of the particles of vehicle exhaust among aerosol particles was studied in Beijing, Chinese capital, during the test of odd–even driving restrictions for Beijing 2008 Olympics Games. This Micro-XRF spectrometer had potential applications in the analysis of single aerosol particles.

1. Introduction

Single particle analysis is a useful method of identifying origins of aerosol particles [1,2]. It can provide more detailed and reliable information of aerosol particles than bulk analysis. The main methods of the analysis of single aerosol particles are electron probe X-ray microanalysis (EPXMA), laser mass microanalysis (LAMMA), micro synchrotron X-ray fluorescence (Micro-SXRF) and micro proton-induced X-ray emission (Micro-PIXE) [1–5]. The MDLs of the EPXMA and LAMMA are higher than those of the Micro-SXRF and Micro-PIXE. Although the Micro-SXRF and Micro-PIXE can provide elemental analysis with μg/g sensitivity for almost all elements even in a micrometer size samples, they are large and expensive facilities. In fact, the XRF analysis of single aerosol particles can be carried out by using a laboratory Micro-XRF spectrometer based on a PFXRL [6–8]. It is well known that the distributions of the X-ray intensity in the focal spot of the PFXRL are Gaussian distribution. This is not helpful for quantitative analysis of single aerosol particles, because some elements are inhomogeneously distributed over the particles with different shapes and sizes [9]. Therefore, the studies on the application of the PFXRL in the analysis of single aerosol particles are now focused on how to obtain the accurate results of the quantitative analysis. In fact, if the Micro-XRF spectrometer based on the PFXRL is used to identify the origins of single aerosol particles, the quantitative analysis of single aerosol particles is not necessary. The reasons for this are as follows. In the origin identification of single aerosol particles, one is mostly interested in whether the elemental composition of an unknown aerosol particle is similar to that of a known origin aerosol particle, rather than the absolute value of the elemental compositions. Previous studies have revealed that particles from different pollution sources have different elemental composition characterized by its characteristic X-ray spectrum [2]. The pattern of the characteristic X-ray spectrum is considered as the chemical fingerprint of the single aerosol particle. The origins of single aerosol particles can be therefore identified by comparing the spectra of unknown single aerosol particles with the spectra of single aerosol particles from air pollution sources [2]. In our studies of the application of the Micro-XRF spectrometer based on the PFXRL in the origin identification of single aerosol particles, the method of directly identifying the origins of single aerosol particles by their characteristic X-ray spectra was used.

In this paper, the Micro-XRF spectrometer based on the PFXRL was proposed to be used in the origin identification of individual aerosol particles. The performances of the micro-XRF spectrometer

* Corresponding author. Address: Institute of Low Energy Nuclear Physics, Beijing Normal University, Beijing 100875, China. Tel.: +86 10 62207171; fax: +86 10 62208258.
E-mail address: stxbeijing@163.com (T. Sun).

0168-583X/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2008.11.032

Table 1
The parameters of the PFXRL.

Length (mm)	86.3
Input diameter (mm)	4.5
Output diameter (mm)	3.3
Focal distance at 17.4 keV (mm)	15.1
Focal spot size at 17.4 keV (μm)	33.7
Gain in flux density at 17.4 keV	3260
Number of the capillaries composing the PFXRL	270,000

based on a PFXRL were studied. As an example of the application of the micro-XRF spectrometer based on the PFXRL in the origin identification of single aerosol particles, the percentage of the particles of vehicle exhaust among aerosol particles was studied in Beijing, Chinese capital, during the test of odd–even driving restrictions for Beijing 2008 Olympics Games. The effects of air conditioning on the indoor air quality were investigated.

2. Experiments and results

2.1. Experimental setup

The Micro-XRF spectrometer based on the PFXRL is described in detail in [10]. The X-ray source is a Mo rotating anode X-ray generator (RIGAKU RU-200, 60 kV–200 mA) whose spot size is $300 \times 300\ \mu m^2$. The detector system is an XFlash® Detector 2001 RÖNTEC and RÖNTEC MAX Spectrometer. The energy resolution of this detector system is 140 eV at 5.9 keV. The parameters of the PFXRL are reported in Table 1.

2.2. MDLs of micro-XRF spectrometer based on PFXRL

The MDL can be calculated by the following formula [11]:

$$\mathrm{MDL} = 3 \cdot C_i N_B^{1/2} / N_A, \quad (1)$$

where C_i is the concentration of element i in the standard sample, N_B and N_A are the area of background and net peak of the analyzed element, respectively. The MDLs of the micro-XRF spectrometer based on PFXRL were 9 and 13 ppm for Fe-Kα and Cu-Kα, respectively, which were measured using standard samples prepared by pipetting standard solutions on a Mylar film with a thickness of 3 μm. The measurement time was 300 s when the operating voltage and current of the X-ray tube were 23.0 kV and 60.0 mA, respectively. The MDLs of the micro-XRF spectrometer based on PFXRL were higher than those of LAMMA, Micro-SXRF and Micro-PIXE, and lower than that of EPXMA.

3. Identification of origin of single aerosol particles

The origins of single particles were identified via the following steps. First, numerous single particles collected from the possible emission sources were analyzed by the micro-XRF spectrometer based on the PFXRL one by one. Their micro-XRF spectra were recorded in a database as the fingerprint library of the air pollution sources. Second, the particles collected from the environmental monitoring site were analyzed in the same way. Their micro-XRF spectra were recorded in another database as the fingerprint library of the environmental monitoring samples. Finally, the micro-XRF spectra in the fingerprint library of the environmental monitoring samples were compared with those of air pollution sources. Origins of the analyzed individual particles from the environmental monitoring site were directly recognized.

For sampling, a Battelle-type cascade impactor was used, which has eight size-fractionated stages with the size range of <0.25, 0.25–0.5, 0.5–1, 1–2, 2–4, 4–8, 8–16 and >16 μm, respectively. The samples were prepared within a sampling time limit so that the particles were well separated with a minimum separation distance which is greater than the diameter of the excitation source beam. The aerosol particles were collected on a Mylar film with a thickness of 3 μm. In order to avoid particles piling up on the Mylar film, the collected samples were firstly checked with a polarizing microscope with a total magnification of 1250.

In order to establish a fingerprints database of the air pollution sources, some typical emission sources were sampled. These sources were soil dust, vehicle exhaust and construction dust, respectively. Figs. 1–3 show some XRF spectra of a single aerosol particle collected on the stage with the size range of 2–4 μm mentioned above. As shown in these spectra, each emission source had its characteristic micro-XRF spectrum, and the relative peak intensities of the elements were different from each other. Therefore, micro-XRF spectra can be used to identify the sources of each measured particle collected from the environmental monitoring site.

For each particle collected on the stage with the size range of 2–4 μm, we recorded six X-ray spectra with the center of the focal spot of the PFXRL locating at six different positions on the analyzed particle, and regarded the average of these six spectra as the chem-

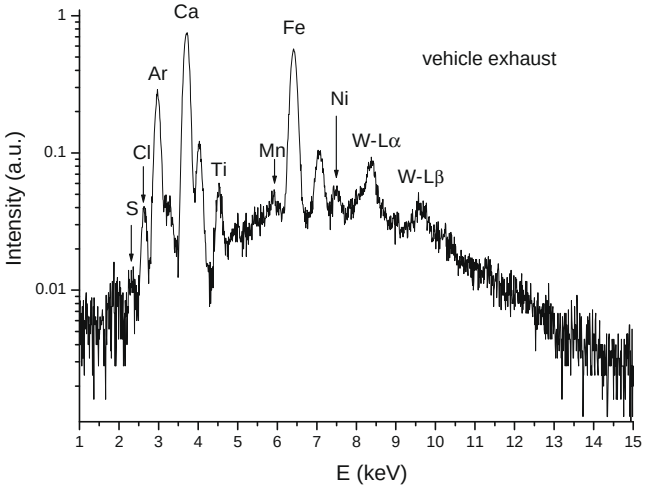

Fig. 1. XRF spectra of a single aerosol particle of vehicle exhaust. (W-Lα and W-Lβ are from the contaminations on X-ray source target).

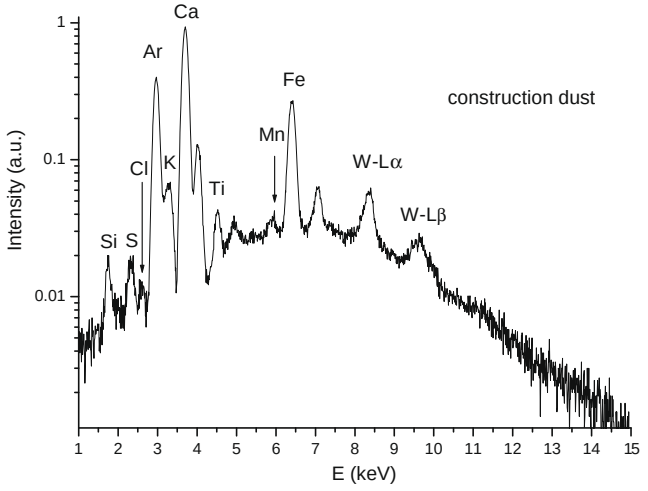

Fig. 2. XRF spectra of a single aerosol particle of construction dust. (W-Lα and W-Lβ are from the contaminations on X-ray source target).