Open Access Library Journal
2022, Volume 9, e9045
ISSN Online: 2333-9721
ISSN Print: 2333-9705

Research on the Influence of Collimator on the Inversion Algorithm of NDP Measurement

Miao Feng, Peng Wu, Hongjie Chen, Baolin Song

College of Nuclear Technology and Automation Engineering, Chengdu University of Technology, Chengdu, China
Email: 774254767@qq.com

How to cite this paper: Feng, M., Wu, P. Chen, H.J. and Song, B.L. (2022) Research on the Influence of Collimator on the Inversion Algorithm of NDP Measurement. *Open Access Library Journal*, **9**: e9045.
https://doi.org/10.4236/oalib.1109045

Received: June 25, 2022
Accepted: July 23, 2022
Published: July 26, 2022

Abstract

The neutron depth profile analysis (NDP) technique can obtain the depth distribution information of ^{10}B, ^{6}Li, ^{7}Be, ^{22}Na and other light core elements near the surface without loss. It is widely used in the influence of Li^{+} on the performance of lithium batteries. When using NDP technology to measure lithium concentration in lithium batteries, the distribution of target nuclide concentration with sample surface depth can be obtained by measuring energy spectrum inversion. In this paper, the principle of measuring lithium ion concentration in lithium batteries with the NDP method is analyzed, and a collimator is added to the measurement system. Through the research and evaluation of four typical lithium ion concentration distributions, it is shown that the collimator can effectively improve the reliability of the inversion algorithm when applied to the inversion calculation of NDP measurement.

Subject Areas

Radiation Measurement

Keywords

Collimator, NDP Measurement, Inversion Algorithm

1. Introduction

The lithium battery is a secondary battery, its positive and negative poles can make Li^{+} detached and embedded, and the process is reversible [1]. The neutron depth profile analysis (NDP) can be used in situ characterization of lithium battery anode and nondestructive measurement of Li^{+} concentration [2]. At present, NDP technology is mainly applied to the research of lithium battery electrolyte and anode materials in foreign countries, while the application of NDP technology in lithium batteries is still insufficient in China. In order to improve the spatial resolution of detected lithium ion concentration in most NDP measurements, the detection solid angle is reduced by increasing the position distance between the detector and the sample. Meanwhile, the ions received by the detector are very limited, which affects the detection efficiency. The collimator can limit the particle beam and increase the duty ratio. The performance of the collimator depends on the geometric transparency of the porous narrow channel of the collimator, the size and uniformity of each small hole, the thickness and strength of the hole wall, the length of the channel, the surface smoothness and straightness, etc. [3]. The earlier collimator was bulky and large with a large aperture and a wall thickness of 5 cm. In the 1980s, the development of the handicraft industry made the thin collimator appear, and the hole wall thickness reduced to half of the original. In the late 1980s, the collimator produced by micro-hole casting method had a uniform aperture and uniform shape, which greatly improved the resolution. In the 1990s, a variety of shape collimators appeared. Studies by many scholars show that the optimized collimator design can effectively improve the resolution and detection efficiency of the front-end system [4] [5] [6]. The most direct way to maximize detection efficiency is to align collimator holes with detector pixels [7] [8]. So far, many scholars have done a lot of research on collimators, detectors and reconstruction algorithms, but the research on collimators used in NDP measurement is little. Therefore, this paper proposes to increase the collimator in the NDP measurement system to improve detection efficiency.

In the NDP lithium ion concentration measurement system, in addition to the application of collimator, it is also necessary to use a certain algorithm to inverse the measured energy spectrum to obtain the depth distribution of lithium ion concentration. At present, the common inversion algorithms of NDP are probabilistic iterative method, SVD singular value decomposition for least squares method, and linear regularization method. Cao and Zhao *et al.* used the improved algorithm combining regularization theory and Chahine iterative algorithm to reconstruct the particle size distribution in 2015, which improved the stability and smoothness of the inversion results [9]. When using NDP technology to measure lithium concentration in lithium batteries, the distribution of target nuclide concentration with sample surface depth can be obtained by measuring energy spectrum inversion. Therefore, this paper discusses the influence of the inversion algorithm on the lithium ion concentration in the NDP measurement by the collimator through the simulation experiment. When using NDP technology to measure lithium concentration in lithium batteries, the distribution of target nuclide concentration with sample surface depth can be obtained by measuring energy spectrum inversion. Therefore, this paper discusses the influence of the inversion algorithm on the lithium ion concentration in the NDP measurement by the collimator through the simulation.

2. Theory

When the lithium battery is measured by NDP, the neutron capture reaction occurs after the Li element in the lithium battery sample captures the neutron. The reaction equation is as follows:

$$^{6}\text{Li} + \text{n} \rightarrow {}^{3}\text{H}^{+} + {}^{4}\text{He}^{2+} \tag{1}$$

The energy of $^3H^+$ generated by Equation (1) is 2727.9 keV, and the energy of α particle is 2055.5 keV. $^3H^+$ is emitted in all directions, and ions travel through the lithium battery medium and lose energy. Some ions reach the surface of the lithium battery. So $^3H^+$ has spatial and concentration information of Li^+. Some ions travel to the surface of the lithium battery sample after losing a certain amount of energy. According to these $^3H^+$ residual energies, the initial position of the nuclear reaction between Li element and neutron can be calculated in order to obtain the concentration of Li^+ at the corresponding depth.

The energy loss law of heavy charged particles in matter can be expressed by Bethe-Block formula.

$$S_{ion} = \left(-\mathrm{d}E/\mathrm{d}x\right)_{iom} = \frac{4\pi \mathrm{z}^2 \mathrm{e}^4}{m_0 v^2} \cdot NB \tag{2}$$

$$B = Z\left[\ln\left(2m_0 v^2/I\right) - \ln\left(1-\beta^2\right) - \beta^2\right] \tag{3}$$

S_{ion} is the loss rate of ionization energy; e is electron stationary charge; z is the charge number of incident charged particles; m_0 is the electron static mass; v is the velocity of incident charged particles; n is the number of atoms per unit volume of lithium battery medium; i is the average Equivalent ionization potential of lithium battery medium; $\beta = v/c$, for the non-relativistic particle $v > c$, β can be ignored, where c is the speed of light. According to the Formula (2) and Formula (3), it can be seen that the ionization energy loss rate of $^3H^+$ is smaller than that of α particle, so the penetration distance of $^3H^+$ is longer than that of α particle, which is more conducive to the spatial and concentration analysis of Li^+ in lithium batteries. Therefore, $^3H^+$ was selected as the analytical ion in this paper.

Inversion Algorithm is a Method of Reason Inversion from Result [10]. At present, most NDP calculation methods adopt inversion method [11], however, different algorithms are selected in the calculation and processing, and the distribution of the depth change of the sample surface obtained by different algorithms will be different. Therefore, the selection of the inversion algorithm is a key step in the NDP technology. Selecting the appropriate algorithm in the measurement of lithium battery samples can make the Li^+ concentration obtained by inversion calculation more reliable.

When NDP measures lithium batteries, the count of the i_{th} channel is y_i, which detected by the detector has an integral relationship with the longitudinal distribution $C(x)$ of lithium ion concentration:

$$y_i = \int_0^{xmax} C(x) R_i(x)\,\mathrm{d}x,\ i = 1, \cdots, N \tag{3}$$

The response function $R_i(x)$ depends on the model to predict the ion energy distribution reaching the detector, and Equation (3) is the first kind of Freholm

Equation. The method used to obtain the longitudinal distribution of source strength is the inversion problem that will be encountered in many fields. There are different core problems, which depend on the actual physical process. In NDP, it is very difficult to calculate $C(x)$ according to Formula (1) by using the count y_i obtained by the known multi-channel spectral analyzer MCA because only finite $C(x)$ is known and countless $S(x)$ values are required in the range of $x = 0$ to $x = x_{\max}$. Therefore, the lithium battery samples are divided into j layers, and the matrix relationship between the counting and the longitudinal distribution of Li^+ concentration can be written as follows:

$$\begin{bmatrix} R_{1,1}R_{1,2}\cdots R_{1,j} \\ R_{2,1}R_{2,2}\cdots R_{2,j} \\ \vdots \\ R_{i,1}R_{i,2}\cdots R_{i,j} \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \\ \vdots \\ C_j \end{bmatrix} = \begin{bmatrix} Y_1 \\ Y_2 \\ \vdots \\ Y_i \end{bmatrix} \tag{4}$$

$$RC = Y \tag{5}$$

In the above formulas, R_{ij} represents the probability that a $^3H^+$ in the j_{th} layer generates a count on the i_{th} channel of the multi-channel detector; R is the energy spectrum response matrix of $i \times j$; C is the concentration distribution of Li^+ ions; and Y is the measured energy spectrum data. The inversion task is to solve the concentration distribution C of Li^+ ions according to the spectral response matrix R and the measured spectral data Y.

In the commonly used inversion algorithm of NDP, SVD singular value decomposition solves the least square method and linear regularization method for inversion, which has the disadvantages of poor stability and strong oscillation [11]. The inversion result of probability iterative method is not ideal when the concentration changes, and it is easy to fall into the trap of local optimum. Simple Principle of Particle Swarm Optimization, easy implementation, fast convergence and global optimization, so this paper chooses particle swarm optimization algorithm as inversion algorithm.

3. Instrument

Nowadays, lithium-ion batteries have been applied to all aspects of human production and life. In the 3C field, most mobile communication devices such as mobile phones and tablets use lithium-ion batteries. In order to ensure that the positive and negative electrodes are not short-circuited inside the battery, separators are also included in the lithium ion battery system using liquid electrolyte. During the charge and discharge process, the positive and negative electrodes generate electronic gain and loss, and the positive and negative materials of lithium ion batteries are reversible to embed and remove Li^+ to balance the charge. This kind of lithium ion battery can be charged and discharged repeatedly is like a rocking chair with positive and negative electrodes at both ends. Li^+ is like a small person running back and forth on the rocking chair, which is vividly called rocking chair battery [12].

Organic liquid electrolytes used in commercial lithium ion batteries are toxic, flammable and leaky, bringing security challenges [13]. Thin-film lithium battery is a kind of all-solid-state battery with high safety and good compatibility. It replaces the liquid electrolyte and separator with inorganic solid electrolyte, which can inhibit the growth of lithium dendrite during charging and discharging to avoid damaging the cathode structure of the battery, and avoid the influence of liquid composition on the measurement of lithium ion concentration [14]. In this paper, the lithium battery model reference X. Liu *et al.*'s NDP measurement lithium battery experiment used all-solid-state thin film lithium battery [15]. NDP measurement lithium battery experiment is shown in **Figure 1**.

When the battery was charged and discharged, most of the $^3H^+$ produced by Li^+ capture reaction with neutron still remained in the electrolyte, only a few Li^+ migrated to the electrode, and the $^3H^+$ concentration in the electrolyte was higher than that in the electrode. With the increase of charging and discharging time, Li^+ continuously migrates to the electrode. At this time, the concentration of Li^+ on the electrode increases continuously until the Li^+ in the electrode reaches saturation. Therefore, it is necessary to ensure that the measurement time is long enough to ensure that the inversion algorithm correctly inverts and calculates the depth distribution of lithium ion concentration in lithium battery [16].

Weng Fenghua clearly pointed out that the image quality depends largely on the performance of collimator in 2016. The collimator parameter size is closely related to the nuclide energy, the collimator designed by ideal collimator method can reduce flux error very well [13]. Wen Yuqin *et al.* studied the influence of collimators with different speed limiting apertures on the measurement results in 2021. According to the different incident ray energy, the aperture size and shielding thickness of collimator hole were selected, and the optimum size of collimator was established [17], Avoid producing small angle scattered rays from collimator, which provides a reference for the design of collimator in this paper.

In this paper, collimator material is selected in photosensitive resin [17]. According to the current processing technology level, the collimator with radius r = 2.0 mm has better performance, so r = 2.0 mm is designed. The shape of the collimating hole is honeycomb, and the spacing of the collimating holes is 1.0 mm. If the collimating device is manufactured by metal processing or laser processing, it is easy to cause the deformation and fracture of the material. Therefore, the collimating device used in this experiment is processed by 3D printing. The design and physical drawings of the collimator are shown in **Figure 2**.

4. Results and Discussions

In this experiment, the inversion of the depth distribution of Li^+ concentration with different shapes can make the inversion of the depth distribution of lithium concentration in lithium battery more accurate, so as to improve the reliability of the experiment. In order to study the influence of the designed collimator on

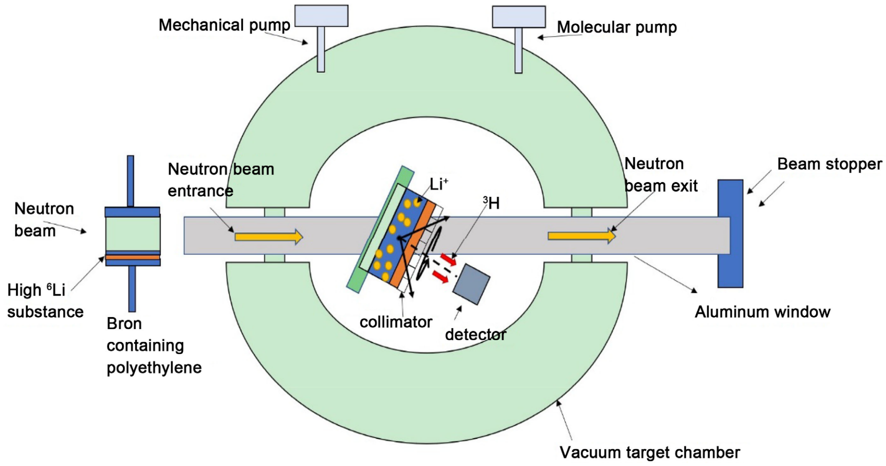

Figure 1. NDP measurement lithium battery experimental system diagram.

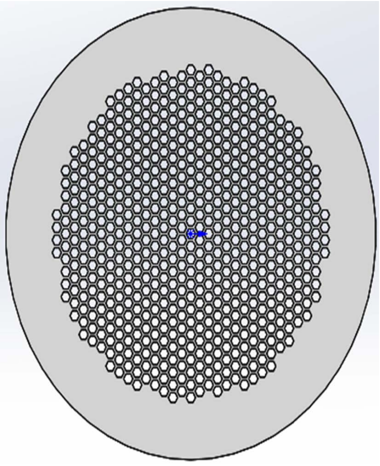

Figure 2. Design diagram and physical diagram of collimator.

the results of the inversion algorithm, this experiment assumed four typical depth distributions of Li^+ concentration: uniform distribution, linear distribution, power function distribution and sawtooth distribution.

The count data is obtained by Monte Carlo simulation software, and the inversion results before and after alignment are compared by particle swarm algorithm. The results are normalized. The comparison results are shown in **Figure 3**. The particle swarm algorithm can better inverse the linear and power function Li^+ concentration depth distribution without collimator, but the inversion results of uniform and serrated are not ideal. After the collimator is added, not only the linear and power function Li^+ concentration depth distribution can be well inverted, but also the inversion results that are not well consistent with the uniform and sawtooth distribution are improved. The inversion depth distribution is in good agreement with the reference depth distribution. It can be found that the inversion accuracy of the particle swarm algorithm is greatly improved after the collimator is used.

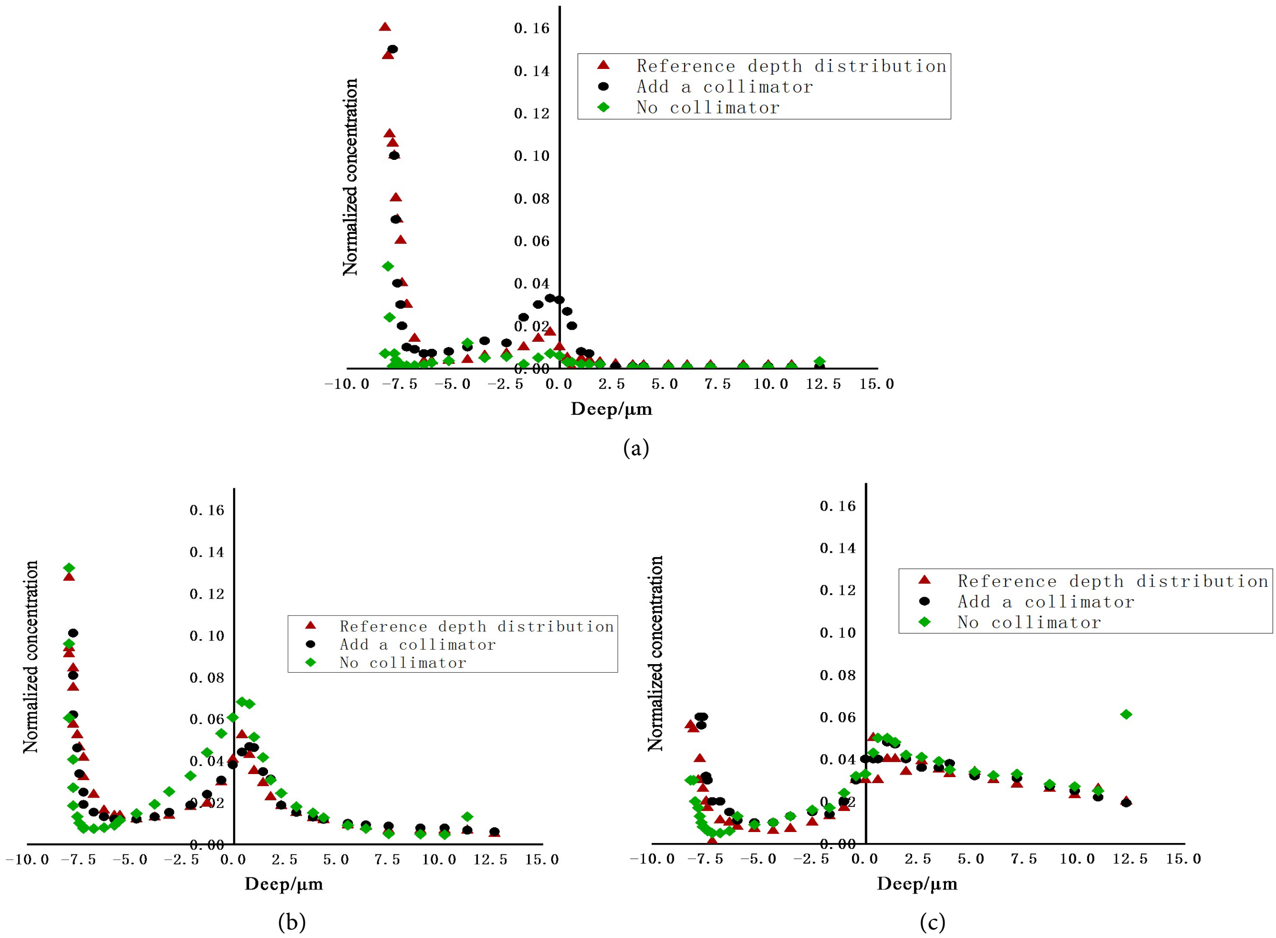

Figure 3. The inversion results of particle swarm algorithm. (a) The results of 0h energy spectrum; (b) The results of 5 h energy spectru; (c) The inversion results of particle swarm algorithm.

It can be seen from Figure 3(a) that the collimator is not ideal for 0 h inversion results. Although the concentration coincidence at the left boundary is improved, the overall calculation results are too large. The reason for this result is that at 0 h, the lithium battery has just begun to charge and discharge, and the concentration of Li^+ in the battery is low, which leads to the low concentration of $^3H^+$ produced by neutron capture reaction. The particle swarm algorithm can't invert the low concentration of Li^+ very well. In practical applications, sufficient measurement time should be ensured in order to improve the accuracy of the results. Therefore, the collimator can improve the reliability of the inversion algorithm under sufficient measurement time.

It can also be seen from Figure 3(b) and Figure 3(c) that the inversion results before collimation are not in good agreement with the reference depth distribution in the electrolyte region, and there are also some differences between the inversion results and the reference depth distribution in the electrode region. After the collimator is used, the concentration distribution trend in the electrolyte region is greatly corrected, and the distribution in the potential region is more consistent with the reference depth distribution.

5. Conclusion

In this paper, the principle of NDP measuring lithium ion concentration of lithium battery and the principle of NDP measuring lithium ion concentration depth distribution inversion algorithm are proposed. The experimental platform and simulation platform are designed to study the influence of collimator on the inversion algorithm of NDP measurement. Comparing the simulated spectrum of four different Li^+ concentration depth distributions with and without collimator, the obtained spectrum is inverted. The results show that adding a collimator to the NDP measurement system can improve the accuracy of the inversion calculation of lithium particle concentration.

Conflicts of Interest

The authors declare no conflicts of interest.

References

[1] Kim, J., Park, K. and Cho, G. (2019) Multi-Radioisotope Identification Algorithm Using an Artificial Neural Network for Plastic Gamma Spectra. *Applied Radiation and Isotopes*, **147**, 83-90. https://doi.org/10.1016/j.apradiso.2019.01.005

[2] Xiao, C.J., *et al*. (2013) Preliminary Study on Neutron Depth Profile Technology of CARR Reactor. 13*th National Symposium on Activity Analysis Technology*, Emei, 22 September 2013, 3.

[3] Feng, Q.X. (1982) Manufacturing Method of Honeycomb Collimator for Cesium Beam Tube.

[4] Charlotte, R., *et al*. (2010) Simulation-Based Evaluation and Optimization of a New CdZnTe Gamma-Camera Architecture (HiSens). *Physics in Medicine and Biology*, **55**, 2709-2726. https://doi.org/10.1088/0031-9155/55/9/019

[5] Trinci, G., *et al*. (2010) A New Variable Parallel Holes Collimator for Scintigraphic Device with Validation Method Based on Monte Carlo Simulations. *Nuclear Instruments and Methods in Physics Research Section A*: *Accelerators*, *Spectrometers*, *Detectors and Associated Equipment*, **621**, 406-412. https://doi.org/10.1016/j.nima.2010.05.023

[6] Wangerin, K., Du, Y.F. and Jansen, F. (2011) CZT Performance for Different Anode Pixel Geometries and Data Corrections. *Nuclear Instruments and Methods in Physics Research Section A*: *Accelerators*, *Spectrometers*, *Detectors and Associated Equipment*, **648**, S37-S41. https://doi.org/10.1016/j.nima.2010.12.240

[7] Suzuki, A., *et al*. (2013) A Four-Pixel Matched Collimator for High-Sensitivity SPECT Imaging. *Physics in Medicine and Biology*, **58**, 2199-2217. https://doi.org/10.1088/0031-9155/58/7/2199

[8] Suzuki, A., *et al*. (2013) High-Sensitivity Brain SPECT System Using Cadmium Telluride (CdTe) Semiconductor Detector and 4-Pixel Matched Collimator. *Physics in Medicine and Biology*, **58**, 7715-7731. https://doi.org/10.1088/0031-9155/58/21/7715

[9] Cao, L.X., *et al*. (2015) Particle Size Distribution Inversion Based on Improved Chahine Iterative Algorithm. *Infrared and Laser Engineering*, **44**, 2837-2843.

[10] Wang, Y.F. (2007) Calculation Method of Inversion Problem and Its Application. In: 2006 *Academic Annual Meeting of Institute of Geology and Geophysics*, Chi-

nese Academy of Sciences, Beijing, 2.

[11] Hnatowicz, V., Vacík, J. and Fink, D. (2010) Deconvolution of Charged Particle Spectra from Neutron Depth Profiling Using Simplex Method. *Review of Scientific Instruments*, **81**, Article ID: 073906. https://doi.org/10.1063/1.3463692

[12] Lu, Y.T. (2017) Preparation and Energy Storage Characteristics of All-Solid-State Thin Film Lithium Battery. University of Electronic Science and Technology of China, Chengdu.

[13] Feng, C., *et al.* (2020) Theoretical Study on the Design of Angle Collimator in High Energy Proton Photography. *Acta Physical Sinica*, **69**, 72-78.

[14] Yu, Z., *et al.* (2020) Research on Prediction of Lithium Battery Cycle Performance Based on BP Neural Network in Laboratory. *Jiangxi Chemical Industry*, No. 3, 93-95.

[15] Liu, M., Cheng, Z., Qian, K., *et al.* (2019) Efficient Li-Metal Plating/Stripping in Carbonate Electrolytes Using a $LiNO_3$-Gel Polymer Electrolyte, Monitored by Operando Neutron Depth Profiling. *Chemistry of Materials*, **31**, 4564-4574. https://doi.org/10.1021/acs.chemmater.9b01325

[16] Cong, S. (2019) Quantitative Method of Neutron Depth Profile Technology in CARR Reactor. Chengdu University of Technology, Chengdu, 56.

[17] Wen, Y.Q., *et al.* (2021) Effect of Collimator of MC Simulated High Purity Germanium Detector on X-Ray Spectrum Measurement. *Acta Metrologica Sinica*, **42**, 245-249.