TOWARDS INTELLIGENT SPECTRUM ANALYZING SYSTEM FOR INDUSTRY-ORIENTED PIXE

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TOWARDS INTELLIGENT SPECTRUM ANALYZING SYSTEM FOR INDUSTRY-ORIENTED PIXE

S. Iwasaki, K. Murozono, K. Ishii, and M. Kitamura

Department of Quantum Science and Energy Engineering,Tohoku University, Sendai 980-77, Japan

Outstanding features of a new method called 'pattern analysis method' are its analysis speed and easy handling, therefore enabling us to automatize the PIXE spectrum analysis. We address the following tasks within the framework of the pattern analysis method towards realization of more intelligent system for the industrial PIXE: adoption of more detailed models for all the reference X-ray spectra of element, of continuum from the Bremsstrahlung process and of the summing effect; real time inference of the content elements based on the iterative Bayes' theorem, which could be effective in the selection of candidate elements and monitoring the status of the spectrum accumulation; use of the suitable X-ray absorber with accurate data for penetration rate, being indispensable to make PAM analysis more reliable; and development of iterative method for thick sample PIXE where various non-linear effects become significant.

INTRODUCTION

Industrial applications of PIXE have not been exploited as expected partly due to the difficulty in the analysis of PIXE spectra. The conventional peak analysis has necessitated analysts to be highly experienced, because of the complexity of PIXE spectra and existence of frequent overlaps of peaks to be separated, and of the ambiguity in the estimation of the continuous background.

The present authors have developed a new method called 'pattern analysis method',[1,2] hereafter PAM. Outstanding features of this method are its analysis speed and easy handling; therefore it could enable us to automatize the process of the spectrum analysis, although there are some problems to be solved in order to apply the method in an actual system.

In this paper, we address the remained tasks of this approach, and also the prospects of the intelligent pattern analysis system applied to future industrial purposes.

PATTERN ANALYSIS METHOD AND ITS IMPROVEMENT

Pattern Analysis Method

The function of PAM is the linear transformation of a vector x , a measured spectrum of a sample, in the spectra space spanned by x_is each of which is the reference spectrum of the single element i (a continuum component due to the Bremsstrahung processes included), into another space of the elemental concentrations, y_i as $y=X+x$. Here, X+ is the Moore-Penrose pseudo-inverse matrix of X which is composed of x_is. X+ is easily obtained by several mathematical algorithms such as the neural network[1], and the singular value decomposition[2], etc. Thus, our task in the analysis is only to multiply the known matrix X+ by the measured spectrum x. In this approach, each component x_i is considered as a fundamental pattern of the measured spectra.

Fig.1 shows an example of the performance of PAM for various liver samples, which have beam measured under the same experimental condition. All measured spectra for a bovine liver, two mouse livers and two rat livers are successfully reproduced with a set of common reference spectra as the results of PAM,[2] though the simplest single Gaussian peak function model has been adopted. Prior the PAM analysis, the candidate content elements among the all possible elements of the atomic numbers from 11 to 82 in the samples and the shape of the continuum reference spectrum were determined from the preliminary analysis of the spectrum for the first bovine liver. The candidate content elements were chosen by the peak search with a priori knowledge on the liver samples. As the continuous background shape, we took a combination of a polynomial and two exponential functions (one is for the higher energy tail, and the other for gamma rays), as done in the SAPIX code.[3] The fitting was performed in an interactive way; we adjusted the model parameters of the background until an acceptable agreement was achieved by observing the fitted result.

Improvement of Reference Spectra Model

Peak Function Model

First, we should adopt more realistic peak function model, i.e., a single Gaussian accompanies by an exponential tail and a plateau with an escape peak as suggested in the literature.[4] A revised model calculation including above components was generally in good agreement with the experimental data, especially in the lower energy region. However, due to the ambiguity of the parameter values given in the literature for the above model, the fitting was not improved as expected for some cases. Therefore, further searches for some model parameters may be necessary using

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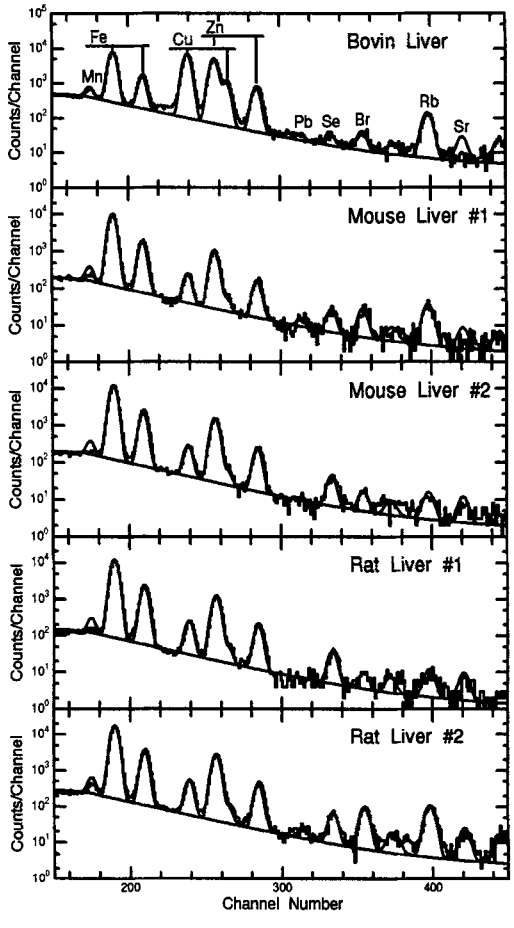
the generalized least-squares method[5] for the multi-elemental standard samples.

Bremsstrahung Components Model

The above mentioned model for the continuum background also introduces uncertainty in the results because there are large ambiguities in the selection of fitting region



in the spectrum, and of the fitting points which are usually

FIGURE 1. PIXE spectra from samples of a bovine liver, two mouse livers and two rat livers, compared with the results of the pattern analysis method.

chosen at some extremal points between peaks. There is no physical base for the fitting functions and region, and the degree of fitting is easily changed by the level of the experiences of the analysts and status of the object spectra.

According to Ishii and Morita,[6] the continuous background is understood mainly as the sum of various Bremsstralung processes, such as atomic Bremsstrahung, secondary electron Bremsstralung, and others. They gave a comprehensive formulation for them. A theoretical model calculation based on this formulation reasonably reproduced the continuous spectrum from a thin Mylar film in shape and amplitude. This theoretical approach could reduce the ambiguity in the estimation of the continuous components.

Sum Coincidence Continuum

The possibility exists of two photons entering the detector within a sufficiently short time interval that they are recorded as a single event. The outcome is a continuum above each peak in the spectrum, culminating in a pile up peak at energy of the sum of the peak energies. In the framework of PAM, we should include the inevitable summing components of the prominent peaks into the reference spectra. The spectral shape of the summing continuum can be estimated using a statistical model.

Evaluation of Penetrability of X ray Absorber

In PAM, the correct evaluation of the penetration rate of the absorber used is indispensable, because the shape of the reference spectra varies due to the penetrability. This effect should be taken into account in the preparation phase of the reference spectra in the system.[2] Sera, et al.[7] pointed out that, e.g., in the case of Mylar absorber, the model calculation based on the data base values for the photoelectric cross sections gave fairly reliable estimation, but, the experimental penetrability data derived from their proposed procedure was more desirable because the procedure was easy, adaptable for any type of absorber, and free from the geometrical uncertainty in the calculation.

TOWARDS INTELLIGENT SYSTEM

Automatic Selection of Candidate Elements

The pre-selection of the candidate elements as mentioned above is necessary in PAM to avoid the ill-conditioned results: spurious elements or elements with negative concentration values.[2] Although the selection using the knowledge of the samples is not so large drawback of the PAM method, it is desirable to find an easier method without or with minimum process. This could be solved by a new approach to the element inference based on the Bayes' theorem. This approach was first developed for the neutron spectrum unfolding.[8]

In this approach, we repeatedly estimate the plausibility or appropriateness of the content elements (i.e., the relative concentration of the elements) from the outcome of the pulse height data. Actually, we accumulate the a posteriori probability for each possible element given by the Bayes formula every time of the event (one count) of the X-ray pulse height data taking. The accumulated posterior probabilities were renormalized (the sum of them equals to

unity) and used as the new a priori probabilities for the estimation the next event. We start the estimation at the first event with the prior of the equal probability for every element in the sample, i.e., with the uniform prior distribution. The posterior probabilities gradually change as increase the number of events, and those of the probable elements in the sample approach to the approximate true relative concentration values, whilst those for the almost all improbable elements decrease to infinitely small values. Sometime, a small number of exceptional elements whose references spectra are quite similar to those of the true content elements remain. These elements can be removed from the candidate lists using the knowledge of the samples.

Thus, the list of candidate elements with the approximate concentration values would be given by this method. The intensities of the Bremsstrahlung's and others continua, relative to those of the reference ones are also inferred by the same way. This method was applied to a PIXE spectrum of a stainless steel type 316 (SUS316) sample, giving a consistent result with other analysis methods.[9]

As mentioned above, the estimation is made on the detection event basis during the measurement. In addition to the candidate element selection, such (quasi-) real-time mode analysis presenting approximate results would be quite efficient and useful for the monitoring the status of the data accumulation especially in the industrial application.

Optimization of X-Ray Absorber

Sera, et al[10] recently proposed the method where they use various types of absorbers aggressively in order to suppress a few prominent peaks. This method improved the signal to background (pile up events) ratio of the spectra, considerably, and enabled them to detect many heavier trace elements especially in the mineral samples. This idea can be applied to other types of samples. We have, actually, used a special absorber for the example case shown in Fig.1, which was a pierced Mylar absorber of 1000 μm in thickness called funny filter. The strong K-X ray peaks from a major element potassium in the liver samples were effectively suppressed, while the lower energy X-rays from the lighter elements survived through the small aperture of the filter. This type of absorber was first used to suppress an ill-condition appeared in the case of a serum sample with no absorber; the result of the indium contained as the internal standard was overestimated due to the strong interference of the potassium K-X ray peaks with the indium L-X ray peaks and partly due to the inadequacy of such simple peak shape model. The suppression of the indium L-X rays relative to its K-X ray peaks solved the ill-condition by increasing the orthogonality between the reference spectra of the potassium and indium.[2]

Future PIXE system should equip a mechanism to choose and set a specific absorber among various type of absorbers to optimize the data taking condition. It would not be difficult to build an intelligent system for the selection of the optimum absorber using the combination of the previously mentioned quasi-real-time analysis method and a small knowledge-base system.

PAM for Thick Sample Cases

If we aim to apply PIXE in the industrial field, we should consider the thick target cases where the various non-linear effects will occur, such as, the energy loss of the incident protons, absorption of the emitted X-rays in the sample, and secondary fluorescence effect in the sample. The matrix correction for the yield of the trace elements become significant in this case. In the case of the full stop thick sample, a thick sample of SUS316, we tested an iterative method to reach a set of correct reference spectra in thick sample, starting from a set of reference spectra in each pure elemental thick target. The latter spectra are sometime quite different with those of the thin target cases, and can be estimated from the model calculation considering above effects. The final obtained reference spectra could interpret the measured thick SUS316 spectrum well, and the result of the elemental concentrations was consistent with the case of the thin sample case.[11]

The above approach was in an ideal case (uniform and smooth surface) of the thick samples. Usually we have no information on the surface of the sample and inside of the matrix. It is known that the useful information can be obtained from the observation of proton spectrum back scattered from the sample surface like Rutherford back scattering (RBS). This kind of information is of particular importance if the sample surface contains lights elements whose X-rays could not observed by usual X-ray detector. Detailed analysis of the observed proton spectra we can deduce more useful information on the matrices.

CONCLUSIONS

In this paper, we discussed the status of the PAM system for the industrial application of PIXE, and prospects towards intelligent PAM system.

The present paper is summarized as follows:

- More detailed models for the reference spectrum of element, of the continuum from the Bremsstrahlung process and of the sum effect are necessary.
- Quasi-real time analysis based on the iterative Bayes' inference could be effective in the selection of candidate elements and monitoring the status of the spectrum accumulation. This method could also be a basic and indispensable technique for the intelligent system, meaning an adaptable system to the change of the status of the data

collection and to various types of samples.

- Use of suitable X-ray absorber is efficient to make the PAM analysis more reliable. The X-ray absorber selection guide and measurement method of the X-ray penetration rate in absorbers have almost been developed.
- Concerning the application of PAM to thick samples, the iterative method is promising although the status of the development is preliminary. Suitable combination of the proton spectrum measurement to PIXE could be useful.

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