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ADVANCES IN SPECTRAL ANALYSIS USING ARTIFICIAL NEURAL NETWORKS

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Abstract

*Artificiel Neural networks (ANNs) have a powerful representational capacity and ability to handle with any multi-*input multi-output mapping problem, e.g. in clustering, pattern recognition and identification areas, particulary when combined with some *a priori* knowledge and statistical point of view. They can be useful in spectrometry for the uranium enrichment measurement methods by example, where numerous approaches like models fitting or experts analysis are limited. These depend on the radiation measured : the methods most widely used developped over the past 20 years were based on the counting of the 185,7-keV peak with a sodium iodide scintillation detector or the 163,4-keV peak of ^{235}U. But these methods depend critically of the source-detector geometry. A means of improving the above conventional methods is to reduce the region of interest : it is possible by focusing at the region called $K\alpha X$ where the three elementary components are present. The measurement of these components in mixtures leads to the isotope ratio $\frac{^{235}U}{^{235}U+^{236}U+^{238}U}$.
In this paper we explore statistical orientations and their consequences on "neural" parameters. We show these decisions are induced by a log-linear model, a special case of a *GLIM* (*G*eneralized *LI*near *M*odel) and correspond to a *Maximum Likelihood Estimation* problem.

NOUVELLES METHODES D'ANALYSE SPECTRALE PAR RESEAUX DE NEURONES

Résumé

Les réseaux de neurones sont une classe de modèles inspirés des systèmes biologiques. Ils sont composés d'un grand nombre de processeurs interconnectés entre eux. Ce fonctionnement collectif (parallèle) est apprécié pour des domaines d'application couvrant la reconnaissance de formes, la classification ou la modélisation. Leur emploi présente plusieurs avantages : ils sont simples à programmer, peu sensibles aux données erronées, contiennent des non-linéarités et sont mis en oeuvre sans fournir un modèle mathématique proprement dit, par opposition aux modèles paramétriques. L'objectif de cette communication est de présenter les résultats obtenus par les techniques neuronales pour le traitement des spectres γ dans le calcul de l'enrichissement en ^{235}U. Les techniques conventionnelles s'appuyent sur un nombre fini de pics γ ou X et exigent un étalonnage préalable de l'instrumentation pour chaque échantillon (les conditions géométriques source-détecteur étant très différentes). On peut s'en affranchir en se focalisant sur la région dite $K\alpha X$ ou trois composantes seulement sont présentes: l'^{235}U à 89,9 et 93,3-keV , l'^{238}U à 92,4 et 92,8-keV, enfin une contribution due à une auto-fluorescence X à 94,7 et 98,4-keV. La quantification de ces trois éléments doit conduire au taux désiré $\frac{^{235}U}{^{235}U+^{236}U+^{238}U}$.

Nous établissons ici que les réseaux de neurones permettent le calcul du taux d'enrichissement pour des échantillons en géometrie infinie. Nous montrons que le couplage des techniques neuronales avec les méthodes issues de l'analyse statistique des données permet d'améliorer la qualité de toute modélisation neuronale .

1 Introduction

A new approach to the multivariate calibration problem is described here. The method use ANN which, like most statistical methods, are capable of processing vast amounts of data and making predictions that are sometimes surprisingly accurate ; this does not make them intelligent in the usual sense of the word. ANNs learn in much the same way that many statistical algorithms do estimation. But in contrast to usual methods of automatic spectra analysis, ANNs use full parallel computing, are simple to implement, not very sensitive to outliers, contain nonlinearities.

Generally, training by Neural Networks needs a priori knowledge to optimize performances : in the choice of architecture which is very similar to the choice of a statistical model of data ([1],[2]), in the choice of the cost-function to minimize ; with such choices, it is possible to orientate learning towards particular properties.

This paper is concerned with ANNs for data analysis and application to parameters estimation in uranium enrichment measurement. The spectrometric methods used depend most of the time on the radiation measured. These can then be classified in three categories, according to the principle on which they are based.

The most widely used procedures are based on the countings of the ^{235}U energy peak, with collimated geometry ([3], [4], [5], [6]). They have been developed over the past 20 years. The counting of the 185.7 -keV peak, recorded with a sodium iodide scintillation detector, requires a previous calibration with known enrichment standards. Provided that the sample measured is similar to the standards and the measurements conditions are constant, the counting rate for the 185.7 keV energy peak is proportional to the enrichment. Very often the samples measured have infinite thickness, i.e. their thickness is such that a further increase in its value has no effect on the emission of the 185.7-keV radiation. Under these conditions, with appropriate collimation, it is possible to be independent of matrix effects for all so-called infinitely-thick samples. Although they are easy to use, these methods have some inconveniences. They depend very critically on the source-detector ensemble geometry and a bias, due to modifications of the background curve under the peak, can also appear at low enrichments. In addition, the 186.2 keV peak attributable to the presence of radium can perturb the data recorded for the 185.7-keV peak. Thus, for measuring uranium concentrations in minerals, some [7] have preferred to use another peak characteristic of ^{235}U, the 163.4-keV peak.

Similary to plutonium isotopic composition measurement, some workers have suggested the use of several γ-ray peaks for determining uranium enrichment, provided that the ^{238}U is in equilibrium with its daughters [8], [9]. In fact, these methods imply a self calibration based on a limited number of peaks. These are mainly the peaks with γ-ray energies of 84.2, 143.8, 163.4, 185,7, 205.3 keV for ^{235}U, and 63.3, 92.4, 92,8, 766.4 and 1001.3 keV for ^{238}U. This autocalibration and the need of fixed conditions represent the major difficulties in these methods.

A means of mitigating the above problem is to reduce the region of interest so that the variation in the efficiency response of the detector is limited. This is possible by considering the region called $K_\alpha X$ where γ-ray (gaussian distribution) and X-ray peaks (Voigt function) are superposed. The measurement of these components in the complex spectrum leads to the enrichment value. This measurement is now carried out with this complex spectrum using the *L*east *S*quares *P*rinciple *LSP*, or by a Fourier transform of the complex spectrum [10]. It is precisely in this context that the ANNs appear to be a useful tool for the characterisation of so-called infinitely thick samples. Indeed, for statisticins, ANNs belong to evaluation techniques for non-parametric models (still called *tabula rasa*). In contrast to usual classical spectral analysis methods, ANNs are full parallel computing, are simple to implement, not very sensitive to outliers and contains non-linearities to deal with any multi-input multi-output mappings.

In the following, we present a description of the experimental procedure in Sec.II. Section III covers the supervised learning. In Sec. IV, we discuss the salient features of the training method and the criteria for stopping the training.

2 Experimental procedure

2.1 preliminaries

Similary to plutonium isotopic measure processes, it should be possible to use all the spectrum to compute enrichment ; but, it means establishing the efficiency of the detector in a relative manner for one sample, the response being the product of the 4 elementary components:

- impulsionnal response of the detector,

- the tranfer of this response to the sample,
- the attenuation by screens between them,
- auto-absorption in the sample.

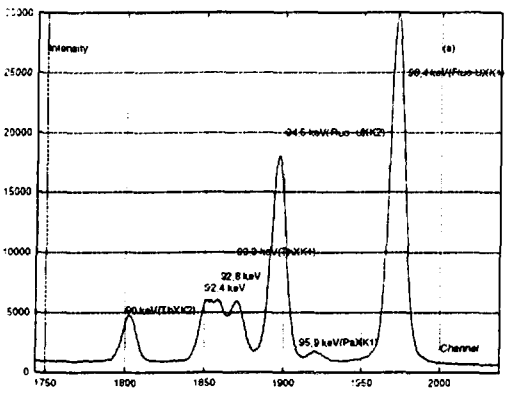

Figure 1: Principal useful X- and γ-rays in the spectral analysis of the $K_\alpha X$ region.

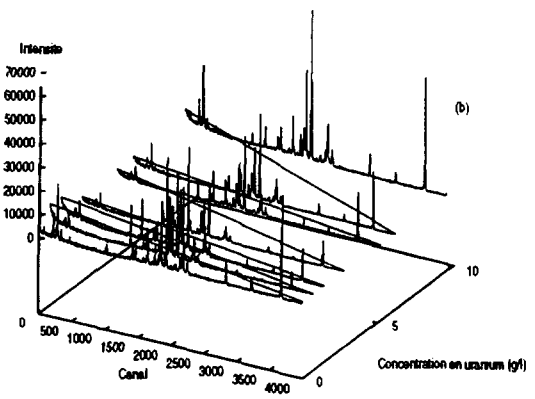

Figure 2: 3D-Representation of the UO_2 spectra set.

The sensibility response of the sensor is varying critically with energy. It depends of a great number of parameters, particulary of the dimensions of the sample, the composition of materials. In the case of uranium, this is still more complex because the response is definite by a number a peaks too short. We go beyond these limitations by focusing on the $K_\alpha X$ region from 83 to 100 keV (figure 1). This region contains exhaustive information to measure ^{235}U and ^{238}U, and at a first sight, the global response can be considered as constant. The signal is however very complex due to several interfering X and γ-raies. These can be retained in the following manner:

- ^{235}U and daughters : 84,21 keV (γ^{231}Th), 89,95 keV (γ^{231}Th), ($ThK_{\alpha_2}X$), 92,28 keV ($PaK_{\alpha_2}X$), 93,35 keV ($ThK_{\alpha_1}X$), 95,86 keV ($PaK_{\alpha_1}X$)
- ^{238}U and daughters : 83,30 keV (γ^{234}Th), 92,28 keV ($PaK_{\alpha_2}X$), 92,38 keV (γ^{234}Th), 92,79 keV (γ^{234}Th), 94,65 keV ($UK_{\alpha_2}X$), 95,86 keV ($PaK_{\alpha_1}X$), 98,43 keV ($UK_{\alpha_1}X$), 99,85 keV (γ^{234}Pa)
- Uranium X-ray fluorescence : 94,65 keV ($K_{\alpha_2}X$), 98,43 keV($K_{\alpha_1}X$).

In the classical way, the processing of this region is done by dealing 3 elementary pattern : one due to the presence of ^{235}U and its daughter products, the second corresponding to ^{235}U and its daughter products, the third corresponding to X self-fluorescence. The measuring of these components leads to the ratio $\frac{^{235}U}{^{235}U+^{238}U}$. The final value of uranium enrichment is established by taking into account ^{234}U contribution. The procedures suggested by Kull & Gunaven [4], and Neuilly [11] are based on ordinary linear regression with enrichment chosen as the independent variable. The error analysis presented does not assess the estimates of the regressors. Liggett [12] investigate *M*aximum *L*ikelihood *E*stimators (*MLE*) but does not apply them. In each case, the data are analyzed on the assumptions of constant variance and the normally distribution of the random errors (due to the randomness of radioactive decay). It will be shown the ANNs build a mapping with the learning examples during the training period. They are able of interpolation providing that the new example is close to the learning database. Extrapolation is not possible: the response for a distant pattern will be wrong a priori.

2.2 Samples measurement

Six UO_2 standards with different uranium enrichment in infinite thickness have been measured several times by γ-spectrometry to test neural procedure. These are bare cylindrical pellets, with certified enrichment value, detailled in table 2:

The Ge(HP) planar detector used in the γ-ray spectroscopy system had the following characteristics: surface=2.00 cm^2, depth= 1,00 cm, for a efficiency of 190 eV at 6 keV and 480 eV at 122 keV.

All the measurement were made in similar conditions, i.e. with 0.05 keV/channel. Ten 20000 seconds counts were made for each enrichment. The presence of ^{234}U is minor : the $\frac{^{234}U}{^{235}U}$ relative ratio vary from 0,5 to 1,1% accordings to the measurement procedure, i.e. using or not the 53,2 and 120,9 keV peaks for ^{234}U.

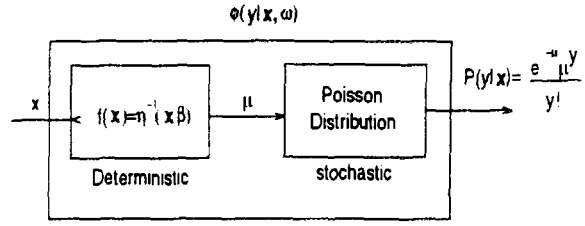

Figure 3: Poissonian Model of a γ spectrum channel.

3 Neural Network and Training method

3.1 Using ANN : Methods

In this section, we briefly summerize basic concepts and necessary notations. Our aim is not the presentation of the neural network theory, but to present the place of the connectionnist approach in γ-spectrometry problems. Table ?? displays much of our notation. More details can be found in a paper to be published [13]. During the last two decades, the method of *M*aximum *L*ikelihood (*ML*) has become the most widely followed approach to the problem of parameters estimation.

The basic idea is fairly simple. The $K_\alpha X$ region to be considered is divided into 210 channels. Let $\chi = (\mathbf{x}, \mathbf{y}^*)_{i=1}^N$ be the set of observations, where the inputs $\mathbf{x}$ are simply the enrichment values and $\mathbf{y}^* = (y_1, y_2, \ldots y_M)$ the outputs photon counts vector. A reasonnable model of countings in multi-channel spectra is to take the desired output $\mathbf{y}^*$ as the mean of a poissonian process. The assumed probability model is :

$y_i \rightsquigarrow$ independent Poisson $(\mu_i), i = 1, \ldots, n$
$\mu = (\mu_1, \ldots, \mu_n)$
link function: $\mu_i = e^{\eta_i}$
linear predictor : $\eta_i = \sum_{j=1}^p \beta_j x_{ij}$

The linear predictor given by is linked with the random component $\mu_i = e^{\eta_i}$ of moments $E(y_i)$ and constant variance σ^2 in the ordinary linear models. The objective of ANNs is to construct a suitable statistical model (see figure ??) which, when subjected to a ^{235}U enrichment spectrum, produces an output $\mathbf{y}$ which approximates the exact uranium enrichment ratio. The principal idea of the connectionist approach is to substitute a neural model and the learning procedure of the network for classical fitting algorithms.

The parameters of this model are the connection weights of the neural mapping. They are found from a goodness-of-fit criterion between expected data and predicted values, according to Nelder et Wedderburn's works [2] on *G*eneralized *LI*near *M*odel (*GLIM*). Let $\mathbf{y}^*$ have a density function $\phi(\mathbf{x}, \omega)$, where ω is some vector of parameters to be estimated. When $\phi(\mathbf{x}, \omega)$ belongs to the exponential family, the maximisation of ϕ with repect to ω can be recast in an optimisation problem using the sufficient statistic for $\mathbf{y}^*$.

The expected μ_i are derived from the enrichment values x by a exponential tranformation. For this problem, the likelihood is $f(\mathbf{y} \mid \mu) \propto \prod_i \frac{e^{-\mu_i} \mu_i^{y_i}}{y_i!}$, and we define the log-likelihood by :

$$\ell(\mu \mid \mathbf{y}^*) = lnf(\mathbf{y}^* \mid \mu) + c = \sum_i (y_i ln \frac{y_i}{\mu_i} - y_i + \mu_i) \tag{1}$$

We have set the constant so that $\ell(\mathbf{y}^* \mid \mu) = 0$ when $\mu = y$. $\ell(\mu \mid \mathbf{y}^*)$ is the *cross-entropy* cost function. The usual maximisation of $\ell(\mu \mid \mathbf{y})$ with respect to β_j :

$$\frac{\partial \ell(\mathbf{y}^*, \mu)}{\partial \beta_j} = \frac{d\ell}{d\mu} \frac{d\mu}{d\beta_j} \tag{2}$$

leads to the optimal parameters vector β (this is the *Maximum-likelihood* principle) :

$$\Delta\beta = \alpha' \mathbf{x}(\mathbf{y} - \mu) \tag{3}$$

if α is a user parameter. We recognize the stochastic error gradient descent rule used in Back-Propagation (BP). The learning heuristic can be derived from the previous equations (1) and (3) and allow an iterative online training of the network through iterative update of the connection weights.

3.2 Neural Architecture

The choice of the right architecture is mainly intuitive and imply arbitrary decisions. In general, ANNs are a collection of simple computational units interlinked by connections. The number of units can be very large and the connections intricate. Figure 4 gives a diagrammatic representation.

The notational convention is that the square represents a computational unit into which the x_j's are fed and multiplied by the respective ω_j's. Nodes are analogous to neurons in biological systems. Each node has a series of weighted inputs, ω_i which are output from other nodes. The inputs of the nodes are analogous to synapses, and the weights corresponds to the strengh of the synaptic connection. The resulting products are added in the unit and hardlimited by a sigmoïdal activation function. The most popular one is the logistic function $f(x) = \frac{1}{1+e^{-\frac{x}{\theta}}}$[1].

(1)

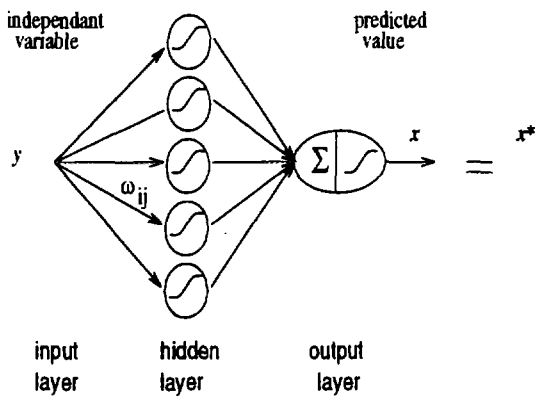

Figure 4: MLP 3-5-1 with nonlinear threshold.

We create a *M*ulti-*L*ayered *P*erceptron (*MLP*), representation of a very familiar statistical construct : the Multivariate Multiple Nonlinear Regression. The essence of the construction set in the role of the hidden layer. A network without hidden units will be unable to perform the necessary multi-input multi-output mappings, in particular in the case of non-linear problems. Input pattern can always be encoded, if there are enough hidden units, in a form so that the appropriate output pattern can be generated from the corresponding input pattern.

From the χ set of examples, the learning of the network consists in the modification of the synaptic weights in order to minimize an objective function (eq. 3) in relation to the set of examples that will be presented to the network. Cost function represents the discrepancy between desired output, say $\mathbf{y}^{\mathrm{d}}$, and predicted output $\mathbf{y}$ of the model.

To determine the optimal set of conditions on

[1] This function has an output value in the range from 0 to 1, where x is the weighted sum of the inputs plus a bias term, and θ is an adjustable parameter which serves to modify the shape of the sigmoid curve and expresses the nonlinearity of the node's operation [14]

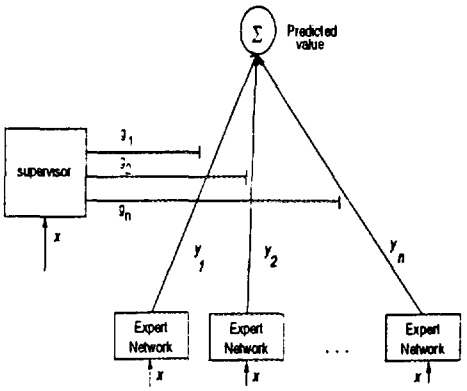

Figure 5: Mixtures of Experts model.

the architecture and the network parameters, we investigated the effect of data-processing, learning rate, and the number of hidden nodes on the network training performance. In this paper we have compared 2 regressions models using ANN with back-propagation of errors : the *MLP model*, where the input are spectral data and the *GMN* (*G*eneral *M*ulti *N*et)(figure ??) where the input are the enrichment values.

The MIXTURE OF EXPERTS(MEX) depicted in figure 5) consists of 210 independant fully-connected networks 1-5-1 : an input unit leads up through one layer of 5 hidden units to an output layer of a single unit that corresponds to the desired counts. Each expert is an observator, trying to find a "signal" in a large amount of noise due to radioactive decay, the variance of each count being proportional to the level and thus depends on the enrichment of a particular sample and on the background level of the particular observation. The input is fed with concentration values. One expert is put for one channel of the $K_\alpha X$ region. The competition between neural model's outputs leads to the choice of the most appropriate concentration, according a SOFT-MAX procedure. The basic idea is that competition leads to specialization.

When the network configuration w is given, we can define the likelihood function $L(p/w) = \prod_i^{210} p(y^i/x^i$ where $p(y^i/x^i, w)$ is the likelihood of getting the expected value y^i for given input data x^i and w. Such conditional distribution could be defined as finding $p_i = p(y^i/x^i, w)$ so that the likelihood of the training set is maximized. By rewriting $L(p/w)$, we arrive at the *Gibbs canonical distribution* on the ensemble of networks[2] :

[2] This is also equivalent to maximizing the Lagrangian of the entropy $-\sum_i p_i \ln p_i$, with the mean cost $< E >$ as constraint.

$$p(y^i/x^i, w) = \frac{e^{-\beta\epsilon_i}}{\sum_j e^{-\beta\epsilon_j}} \qquad (4)$$

where the distance ϵ_i is given by $\epsilon_i = (\mu_i - y_i + y_i \ln \frac{y_i}{\mu_i})$ and $\bar{C} = \sum_i^{210} p_i c_i$; the c_i are predicted estimation of enrichment given by an observator and $\bar{C}$ is the predicted "average" value. It has been demonstrated this procedure, called *inverse model* is particulary efficient in the case of few data and a A-onto-B function. The folllowing section presents the results obtained with MEX, compared to those obtained with a MLP : 3-6-1.

3.3 Discussion of the Results using ANN

Redundances in the data-set enrichments present one main advantage : as we measure more than one response for each case, information from all the measured responses can be combined to provide more precise parameter estimation and to determine a more realistic model.

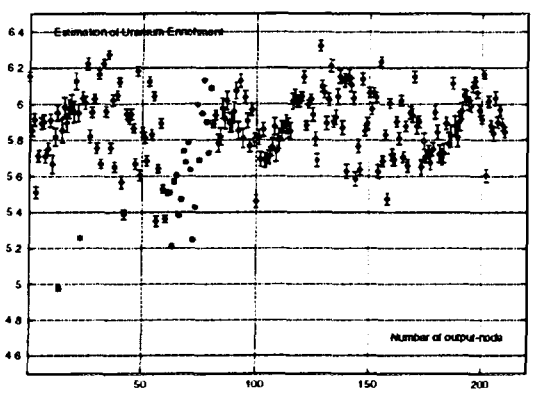

Figure 6: Cloud of enrichment values predicted by each expert compared to expected 5.785 %

In all simulations, the measure of the system's performance is the Mean Square Error. The bias rates are compared in table 1 and on figure 7.

Figure 7 compares the results of the two models. The bias between the predicted and the desired enrichments is plotted for each of the 65 samples. The darkest line is put for the MEX. The results suggest that the strong dispersion of the bias with MLP is significantly attenuated when MEX is applied. This judgement must be moderated for the 6.122-enrichment-ratio samples.

The figure 6 is concerned with the Multi-Expert model. The plotted points are predicted enrichment value (one for each of the 210 experts) when a $5.785\% - {}^{235}U$ spectrum is presented to the MEX

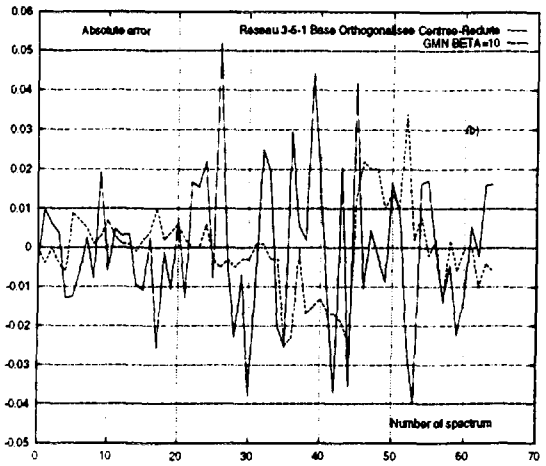

Figure 7: Absolute bias in the enrichment estimation.

model. The credit assignement procedure on these 210 contributions is supervised to produce a final estimation. In the most right column of table (1) can be seen the final predicted values of the simulations with MEX. Compared with the MLPs, this shows that MEX method is really reliable : for example the bias between the predicted and the calculated 2.785% enrichments range from 2,784 to 2,790%. As noted above; after 32 000 successful training passes, the larger bias happens for 5,111 and 6.122% enrichments. This relative lack of precision can be ascribed to the small size of the training dataset.

A comparison of the absolute bias curves suggest that of the two systems studied, the Mixtures of Experts is capable of showing the most robust performance.

The virtues of the MEX algorithms include :

- accurate incorporation of a good physical model.
- representation is easier to interpret.
- automatic inclusion of constraints on all parameters through Gibbs measure.
- a measure of the quality of reconstruction.

Although our algorithm can capture stochastic variation in photon counts and yields more accurate results, it is iterative and therefore slower compared to Fourier-based methods, which are generally preferred in practice. But, with the advent of more powerful computers, the argument favoring MEX become more compelling.

This method is at the same time very general and very specific. It is very general in the sense that no hypothesis is made on the aspect of the spectra : it does not depend if the spectra are well resolved or

not, if they are very likely or not, if you select most significative areas of spectrum only (MLP models) or a global part of the spectrum (MEX model). No physical model is required whereas "classical" procedures may use three Lorentzians for example. But, at the same time, the method is very specific because the ANN must learn representative spectra of the family spectra to identify. Furthermore, other tests proved to us that ANNs are resistant to noise Presently, we must put the blame on the excessively short size of the training dataset.

4 Conclusions

A new approach to the multi-variate calibration problem has been described here, in applying ANNs to uranium enrichment measurement. A neural network is shown to be able to predict ^{235}U concentrations, taking into account, non-linear relationship between spectral response with analyte concentration. The ANN calibration results are especially interesting because they remain reliable in the general case despite the presence of non-linear instrumental artefacts.

Our results appear to be at the state of art in automated quantifying methods for isotopes in a mixture of components in the case of fixed experimental conditions. The basic principle is to use input and output data to provide information on how to set the parameters where no definite mathematical model can be assigned *a priori*. Thus we have adaptative prediction. This requires that the network parameters be set correctly for the work to be carried out as desired. This method has been demonstrated as a reliable tool for dealing with data from low resolution detectors even in under adverse conditions and has been already successfully used by [15] in a X-ray fluorescence application. Final network with connections and weighting functions could be easily implemented using commercial digital processing hardware.

But, there is no single learning procedure which is appropriate for all tasks. It is of fundamental importance that special requirements of each task are analyzed and that appropriate training algorithms are developed for families of tasks. However an efficient use of the networks requires as careful as possible analysis of the problem, an analysis that is often ignored by impatient users.

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Declared enrichment	MLP 3-5-1	MEXs $\beta = 10$
0.711%	0.691-0.723	0.702-0.710
1.416%	1.394-1.426	1.406-1.416
2.785%	2.732-2.822	2.784-2.790
5.111%	5.066-5.148	5.112-5.136
6.122%	6.105-6.162	6.088-6.112
9.548%	9.531-9.570	9.542-9.552

Table 1: Min-Max of calculated Enrichments of MLP and MEX

Diameter(cm)× Height(cm)	$\frac{U}{O}$ ratio (g/g%)	Stated enrichment $(10^{-2}g.g^{-1})$	$\frac{^{235}U}{^{235}U+^{236}U}$ ratio $(10^{-2}g.g^{-1})$
1,30× 2,00	88,00	0,7112 ±0,004	0,7112
1,30× 1,90	88,00	1,416 ±0,001	1,416
0,80× 1,10	88,00	2,785 ±0,004	2,786
0,80× 1,02	87,96	5,111 ±0,015	5,112
0,80× 1,00	87,98	6,222 ±0,018	6,225
0.92× 1,35	87,90	9.548 ±0.04	9,558

Table 2: Percentage Enrichments of UO_2 standards

Symbole	
$\mathbf{y}^*$	desired output vector independantly distributed with means $E(\mathbf{y}) = \mu$
$\mathbf{x}$	input data
η	linear predictor $\eta = \beta\mathbf{x}$
μ	predicted output of the activation node $(\mu_1, \mu_2, \ldots \mu_N)$
β	unknown parameter vector $(\beta_1, \beta_2, \ldots \beta_p)$ to be estimated
ω	neural connection parameter matrix
$f(\mathbf{y} \mid \mu)$	likelihood
$\ell(\mu \mid \mathbf{y}^*)$	log-likelihood
$\ell(\mathbf{y}^* \mid \mathbf{y}^*)$	log-likelihood for an exact fitting
α	learning rate