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Abstract. *Interval Analysis is a new tool, well known in Automatic Control, very powerfull for solving estimation problem. This appraoch is especially suited for models whose outputs is nonlinear in their parameters, a situation which is the rule for models based on prior physical knowledge but where most available methods fail to provide any guarantee as the global validity of the results obtained. It may be useful for NaI detector measurement system commonly used by the EURATOM safeguards system. One of the problems encountered during the in-field measurements of this type is the strong influence of the thickness and type of the material of container on the measurement results. We explore some possible solutions and validate these solutions on the real JRC Ispra PERLA dataset.*

Key words: interval analysis; parameter estimation; nonlinear model; enrichment measurement.

1 Introduction

This paper deals with parameter estimation in an error bounded context. This new approach based on interval analysis, is proposed to compute guaranteed estimates of the parameter vector such that the error between the experimental data and the model outputs belong to some predefined feasible sets.

*Example 1 (*Parameter estimation in $\gamma-$spectrometry). *Let $X \in \mathbb{R}^p$ be the vector of measurements performed by measuring e.g. $\gamma-$rays activity from an uranium sample for which the enrichment E has to be determined. This vector may correspond to the countings (scalar) X_1 and X_2 of the full energy peak at 185,7 keV and 1001 keV, $X = (X_1, X_2)$. We assume that these data are to be described by the output of a linear model $E(X) = p_0 + p_1 X_1 + p_2 X_2$ with unknown parameter vector $p = (p_0, p_1, p_2)$. To determine the unknown coefficients of dependence p_0, p_1 and p_2, many observations are made. For each observation $i = 1, \ldots, n$ numerical data*

$$E_i; X_{1i}, X_{2i} \tag{1}$$

are recorded. We then try to solve the equations

$$E_i = p_0 + p_1 X_{1i} + p_2 X_{2i}, \quad i = 1, \ldots, n. \tag{2}$$

When the number of observations n is greater than the number of unknowns, the linear system usually has no solution. Nevertheless, one would like to find the best possible *values of p_0, p_1 and p_2 for the unsolvable system (2). Of the many senses in which the expression* the best possible *can be defined, the commonest is the least square sense: we look for the value of p_0, p_1 and p_2 that minimizes the sum of the squares of differences:*

$$\phi \equiv \sum_{i=1}^{n} (E_i - p_0 - p_1 X_{1i} - p_2 X_{2i})^2. \tag{3}$$

With the notations

$$E = \begin{bmatrix} E_1 \\ E_2 \\ \vdots \\ E_n \end{bmatrix}, A = \begin{bmatrix} 1 & X_{11} & X_{12} & X_{13} \\ 1 & X_{21} & X_{22} & X_{23} \\ . & . & \vdots & . \\ 1 & X_{n1} & X_{n2} & X_{n3} \end{bmatrix}, p = \begin{bmatrix} p_1 \\ p_2 \\ p_3 \end{bmatrix}$$

with A^T = the transpose of A, ϕ is minimized by a unique *vector p estimated by $\hat{p} = (A^T A)^{-1} A^T E$ (if $\det(A^T A) \neq 0$).* □

Many other criteria ϕ have also been considered such as any criteria given by the maximum likelihood or bayesian approaches under various hypotheses on the noise (see [8]). In this example where the error is affine in the parameters, an explicit formula is available to compute an optimal parameter vector. Most often, however (e.g. when the model output is nonlinear with respect to the parameters), no explicit formula can be provided for the best value of the parameter in the sense of the chosen criterion. A local optimization is then usually performed iteratively, starting from some initial value of the criterion.

*Example 2 (*Nonlinearity). *For instance, the enrichment model could be described by the equation $E(X) = 7f(p_1 X_1) + 2,9 f(p_1 X_2)$. Here, $f(\cdot)$ is a $S-$shape function defined by $f(x) = \frac{1}{(1+\exp(-x))}$, which is almost linear around zero.* □

However, such optimization of a scalar criterion has several drawbacks: (i) the choice of the initial value relies on guess work, (i) no guarantee of convergence of the global optimum of the criterion can be provided, (iii) if there are several values of the estimated parameters that correspond to the same value of the criterion, the algorithm picks one of them, (iv) one is not interested in the *optimal* value but would rather like to characterize the set of all values that are acceptable in a sense to be specified, (v) no reliable evaluation is provided of the precision with which the estimated value of the parameters is obtained.
This is why we shall follow a different route, and look for the set of models that are *acceptable* instead of looking for the model that is optimal in the sense of a given criterion. *Acceptability* can be tackled by listing all the properties that the model should satisfy, *e.g.* a serie of (possibly nonlinear) equalities to be satisfied by the parameters.

*Example 3 (*Some acceptability conditions in spectrometry*). A very realistic assumption is provided by the fact the concentrations are necessarily non-negative numbers !* □

Once these conditions of acceptability have been defined, one is interested in characterizing the set S of all parameter vectors such that the model is acceptable. Starting from some prior feasible set for the parameter vector under the form of axis aligned box in the parameter space, we wish to characterize –in a guaranteed way– the posterior feasible set for the parameters, *i.e.* the set of all values that are consistent with the conditions of acceptability [5]. This will be performed with the help of an algorithm described in section 2. It is, for instance, possible to characterize the set of all parameters that are consistent with the data in the sense that the error between the data and the corresponding model outputs fall within some known prior bound, considered in section 2.2. Section 3 addresses the problem of ^{235}U enrichment estimation out of a spectrum obtained with a NaI detector with different absorbing materials between sample and detector.

2 Interval computation

2.1 Basic principles

Interval arithmetic treats intervals as a new kind of numbers, on which classical arithmetical operations such as $+, -, *, /, \sin, \exp, \ldots$ can be performed.

Definition 1 (Interval). *An interval $[x]$ is a closed and connected subset of $\mathbb{R}$. Let define $[x] = \{x \in \mathbb{R} \mid \underline{x} \leq x \leq \overline{x}, \overline{x} \in \mathbb{R}, \underline{x} \in \mathbb{R}\}$, where $\overline{x}$ and $\underline{x}$ are the upper and under bounds of $[x]$*[4].

Intervals are basic constituents to describe uncertainty, as probability laws are for a statistical description. A box $[\boldsymbol{x}]$ of $\mathbb{R}^p$ (or vector of intervals) is a cartesian product of p intervals, *i.e.* $[\boldsymbol{x}] = [\underline{x_1}, \overline{x_1}] \times \ldots \times [\underline{x_p}, \overline{x_p}] = ([x_1], \ldots, [x_p])^T$. The width $w([\boldsymbol{x}])$ of a box $[\boldsymbol{x}]$ is the length of its largest side.

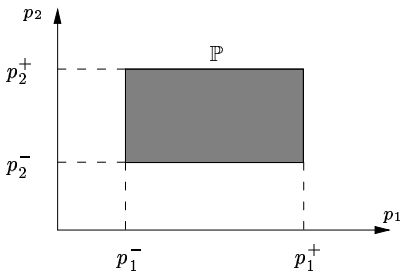

Fig. 1. A 2-dimensional box $[p]$.

Example 4. Let x, y and z 3 real variables such that $z = x + y$. Suppose that x and y are uniformly distributed $P(x) = \frac{1}{5}$ on $x \in [0, 5]$ and $P(y) = \frac{1}{2}$ on $y \in [1, 3]$. Then, to compute the $P(z)$ we need the joint distribution $P(z)$. If x and y are independent, then the calculus is trivial (the joint distribution is the product of the marginal ones), but in the usual case, such calculus is complex. On the contrary, suppose that the uncertainty associated to x and y is given by an interval, then $z \in [z] = [0 + 1, 5 + 3] = [1, 8]$. □

Note that the product $[x] \times [y]$ is defined as follows $\{x \times y \mid x \in [x], y \in [y]\} = [\min(\underline{x} \cdot \underline{y}, \underline{x} \cdot \overline{y}, \overline{x} \cdot \underline{y}, \overline{x} \cdot \overline{y},), \max(\underline{x} \cdot \underline{y}, \underline{x} \cdot \overline{y}, \overline{x} \cdot \underline{y}, \overline{x} \cdot \overline{y},)]$.

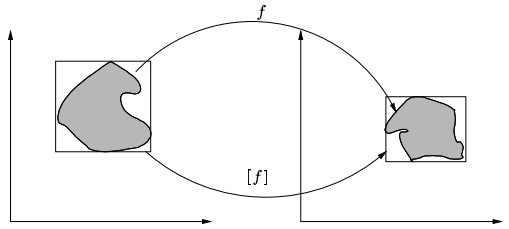

Fig. 2. Minimal inclusion function $[f]$ of a function f.

Definition 2 (Inclusion function). $[f]$ is an *inclusion function* of the vector function f if, for any box $[\boldsymbol{x}]$, $[f]([\boldsymbol{x}])$ is also a box such that

$$\forall [\boldsymbol{x}], f([\boldsymbol{x}]) \triangleq \{f(\boldsymbol{x}) \mid \boldsymbol{x} \in [\boldsymbol{x}]\} \subset [f]([\boldsymbol{x}]). \quad (4)$$

and $w([\boldsymbol{x}]) \to 0 \Rightarrow w([f]([\boldsymbol{x}])) \to 0$[5]. △

The interval function$[f]([\boldsymbol{x}])$ is thus a box (see Fig. 2) that contains $f(\boldsymbol{x})$, *i.e.* the envelopping box of $f([\boldsymbol{x}])$. The inclusion function $[f]$ for f is minimal if for any $[\boldsymbol{x}]$, $[f]([x])$ is the *smallest* box that contains $f(\boldsymbol{x})$. The notion of inclusion function is the key idea of interval analysis: (i) it extends the notion of application to intervals, (ii) and makes possible uncertainty propagation for a large class of nonlinear functions.

Example 5. For instance, let $f : \mathbb{R}^2 \to \mathbb{R}$, defined by $f(x_1, x_2) = \frac{x_1 - x_2}{x_1 + x_2}$, with $x_1 \in [-1, 2]$ and $x_2 \in [3, 5]$. Then, the inclusion function associated to f is $[f]([x_1], [x_2]) = \frac{[-1,2]-[3,5]}{[-1,2]+[3,5]} = [-3, -\frac{1}{7}]$. □

It is easy to compute for usual elementary functions.

2.2 Constraint satisfaction problem

A *constraint satisfaction problem* (CSP) is defined by

- a set of real vector variables $\boldsymbol{y}_1, \ldots, \boldsymbol{y}_n, \boldsymbol{p}$,
- a set of domains (generaly boxes) $[\boldsymbol{y}_1], \ldots, [\boldsymbol{y}_n], [\boldsymbol{p}]$ assumed to contain the variables $\boldsymbol{y}_1, \ldots, \boldsymbol{y}_n, \boldsymbol{p}$,
- and a set of constraints linking all these variables.

The solutions of a CSP are defined as the set of n-tuple $(\boldsymbol{y}_1, \ldots, \boldsymbol{y}_n, \boldsymbol{p})$ such that $\boldsymbol{y}_1 \in [\check{\boldsymbol{y}}_1], \ldots, \boldsymbol{y}_n \in [\check{\boldsymbol{y}}_n], \boldsymbol{p} \in [\check{\boldsymbol{p}}]$ and such that the constraints be satisfied [2]. In an bounded error estimation problem, the

[4] A real number is an interval such that $\overline{x} = \underline{x} = x$.

[5] The last condition is only needed to ensure convergence.

$\boldsymbol{y}_i, 1 \leq i \leq n$, ($n$ is the number of observations) can be any uncertain variables : a noisy data, a time measurement, an input, a perturbation,.... The domains $[\boldsymbol{y}_1], \ldots, [\boldsymbol{y}_n]$ can correspond to interval measurement (or error bars) or any *prior* knowledge on the variable bounds. The constraints can be the equations of the models, and/or any identifiability constraints [9][6]. For instance, in the case of an affine model described by $z = \frac{1}{p_1 x} + \frac{1}{p_2 x}$, one can add the condition $p_1 \geq p_2$ so that the model be identifiable. In the following, $\boldsymbol{p}$ plays the role of the unknown parameter vector. The problem is denoted in a short form by

$$\boldsymbol{y} = \mathcal{F}(\boldsymbol{p}), \boldsymbol{y} \in [\boldsymbol{y}], \boldsymbol{p} \in [\boldsymbol{p}]. \tag{5}$$

A value of a variable is *consistent* with the Eq. (5) if one can instantiate the other variables in their domains such that the relation $\boldsymbol{y} = \mathcal{F}(\boldsymbol{p})$ is satisfied. In the present problem estimation, f is given, $\boldsymbol{p}$ play the role of the unknown parameters and $\boldsymbol{y}$ of the set of *all* measurements:

$$\begin{array}{lcl} \boldsymbol{p} & \leftrightarrow & p_1, p_2 \\ \boldsymbol{y} & \leftrightarrow & x_1, z_1, \ldots, x_i, z_i, \ldots, x_n, z_n \end{array}$$

The initial domain of the parameter vector $\boldsymbol{p}$ is $[-\infty, \infty]$, *i.e.* no prior information is available on the parameters. The constraints on the variables are given by a set of (non)linear equations, *e.g.*

$$\begin{cases} y_1 = f(x_1, \boldsymbol{p}) \\ \quad \vdots \\ y_n = f(x_n, \boldsymbol{p}) \end{cases} \tag{6}$$

which computes all outputs from the chosen model f. *Consistency methods* (also called *constraint propagation*) based on intervals have been proposed (see [1]) to reduce efficiently the variable domains of a CSP without loosing any solution. Among all the proposed methods, the simplest one consists to decomposed the set of constraints in primitive constraints and to contract whenever it is possible by removing values in the domains that are inconsistent with the other domains.
Let us illustrate the method on the problem given in the introduction.

*Example 6 (*Consistency estimation in $\gamma-$spect.*). Each of the constraint $z_i = p_0 + p_1 x_1 + p_2 x_2$ is decomposed in 3 elementary operations (such that only one operator is involved at each statement) by adding intermediate variables (*forward propagation*):*

$$\begin{cases} a_i = p_1 x_{1i} \\ b_i = p_2 x_{2i} \\ z_i = p_0 + a_i + b_i \end{cases} \tag{7}$$

*where a_i, b_i are intermediate variables. We get now $n \times 3$ primitive constraints. Reading Eq. (7) from the end to the beginning (*backward propagation*), we isolate each variables of the right hand side and write the corresponding statement in Eq. (8)*

$$\begin{cases} a_i = p_1 x_{1i} \\ p_1 = \frac{a_i}{x_{1i}} \\ x_{1i} = \frac{a_i}{p_1} \\ b_i = p_2 x_{2i} \\ p_2 = \frac{b_i}{x_{2i}} \\ x_{2i} = \frac{b_i}{p_2} \\ e_i = p_0 + a_i + b_i \\ a_i = e_i - p_0 - b_i \\ b_i = e_i - p_0 - a_i \\ p_0 = e_i - b_i - a_i \end{cases} \tag{8}$$

Equations (7) and (8) should be rewritten by replacing each variable and each operator or function by their interval counterpart. At each iteration, the interval domain computed has to be intersected with its previous value using interval arythmetic. For our example, we get in merging the intervalized forward and backward equations:

$$[a_i] := [a_i] \cap [p_1][x_{1i}] \tag{9}$$
$$[p_1] := [p_1] \cap \frac{[a_i]}{[x_{1i}]} \tag{10}$$
$$[x_{1i}] := [x_{1i}] \cap \frac{[a_i]}{[p_1]} \tag{11}$$
$$[b_i] := [b_i] \cap [p_2][x_{2i}] \tag{12}$$
$$[p_2] := [p_2] \cap \frac{[b_i]}{[x_{2i}]} \tag{13}$$
$$[x_{2i}] := [x_{2i}] \cap \frac{[b_i]}{[p_2]} \tag{14}$$
$$[e_i] := [e_i] \cap [p_0] + [a_i] + [b_i] \tag{15}$$
$$[a_i] := [a_i] \cap [e_i] - [b_i] - [p_0] \tag{16}$$
$$[b_i] := [b_i] \cap [e_i] - [a_i] - [p_0] \tag{17}$$
$$[p_0] := [p_0] \cap [e_i] - [a_i] - [b_i] \tag{18}$$
$$\tag{19}$$

These equatons are called solution equations. We have $10 \times n$ solution equations in our problem. The final algorithm is obtained by repeating several times the merged algorithm for all the data set while the contraction is sgnificant: The first equation (9) guarantee that if the $[a_i]$ domain is replaced by $[a_i] \cap [p_1][x_{1i}]$, then no solution is lost. By applying this type of contraction for each of the $10 \times n$ solution equations, one obtain at the end an equilibrium. □

In summary the contraction algorithm alternates a *forward propagation* step and a *backward propagation* step.

*Example 7 (*Constraint propagation on intervals*).* Consider the following interval problem :

$$\begin{cases} xy = z, z \in [-\infty, \infty], \\ x^2 = z \quad x \in [-10, 10] \text{ et } y \in [1, 3]. \end{cases} \tag{20}$$

Backward propagation of the intervals $[x]$ and $[y]$ on the Eq. (20) gives

$$\left.\begin{array}{rl} [-10, 10] \times [1, 3] = [-30, 30] & = [z] \\ [-10, 10]^2 \qquad\quad = [0, 100] & = [z] \end{array}\right\} \Rightarrow z \in [0, 30].$$

[6] For instance, stability requirement were cast in the framework of set inversion for blind source separation in Vigneron *et al.* [9].

Algorithm Contract	
Inputs:	$[p_0], [p_1], [p_2], [x_{1i}], [x_{2i}], [z_i], i = 1, \ldots, n$
Init:	$[p_0] = [p_1] = [p_2] = [-10000, 10000]$
Repeat	
	For $i = 1$ to n
1	$[a_i] := [a_i] \cap [p_1][x_{1i}]$
2	$[p_1] := [p_1] \cap \frac{[a_i]}{[x_{1i}]}$
3	$[x_{1i}] := [x_{1i}] \cap \frac{[a_i]}{[p_1]}$
4	$[b_i] := [b_i] \cap [p_2][x_{2i}]$
5	$[p_2] := [p_2] \cap \frac{[b_i]}{[x_{2i}]}$
6	$[x_{2i}] := [x_{2i}] \cap \frac{[b_i]}{[p_2]}$
7	$[e_i] := [e_i] \cap [p_0] + [a_i] + [b_i]$
8	$[a_i] := [a_i] \cap [e_i] - [b_i] - [p_0]$
9	$[b_i] := [b_i] \cap [e_i] - [a_i] - [p_0]$
10	$[p_0] := [p_0] \cap [e_i] - [a_i] - [b_i]$
	EndFor
While the contraction is significant	
Output:	$[\boldsymbol{p}]$

Forward propagation of the interval $[z]$ on the Eq. (20) gives

$$[x] = \pm\sqrt{[0,30]} = [-5.5, 5.5]$$
$$[x] = \frac{[0,30]}{[1,3]} = [0,10]$$

and so on... while the contraction is significant. □

This contraction is very fast but, most of the time, all inconsistent values have not been removed, *i.e.* the contraction is not optimal.

2.3 Subpaving in the solution space and bissection

In this section, we show how it is possible to control the precision of the contraction. When the fixed point is reached, domains still contains large parts of inconsistent values. Bissections (procedure Bisect) should then be performed to eliminate them. The strategy to follow, proposed by Jaulin and Walter [4] consists to split the box of interest $[p]^{(k)}$ (at iteration k) into smaller boxes in order to eliminate inconsistent values whenever needed until the width of the considered box becomes smaller than some required parameter ρ. In other words, the analysis of the space of interest is performed by building sets of non-overlapping boxes as represented on figure 3.

Let $\mathbb{P}$ be the set of solution to characterize. We shall say that the box $[\boldsymbol{p}]^{(k)}$ is *feasible* if $\mathcal{F}([\boldsymbol{p}^{(k)}]) \subset [\boldsymbol{y}] \Rightarrow [\boldsymbol{p}^{(k)}] \in \mathbb{P}$ and *unfeasible* if $\mathcal{F}([\boldsymbol{p}^{(k)}]) \cap [\boldsymbol{y}] = \varnothing \Rightarrow [\boldsymbol{p}^{(k)}] \cap \mathbb{P} = \varnothing$. In all other cases, $[\boldsymbol{p}^{(k)}]$ is said *undeterminate*. Note that Figure 3 plots 2 type of subpavings (computed iteratively by our algorithm): p^- which contains all (dark grey) boxes that are proved feasible, p^+ containing all indeterminate (grey) boxes. From the subpavings it is easy to bracket $p^+ \cup p^-$ guaranteed to contain the portion of $\mathbb{S}$ of interest. Interval computational methods are used to bracket the set of interest. Let $\boldsymbol{p}^{(k)}$ be the box considered at the iteration k, ρ the *required* accuracy. A stack is used to store the boxes under consideration during the processing. A stack is dynammical structure on which the only 3 operations are possible.

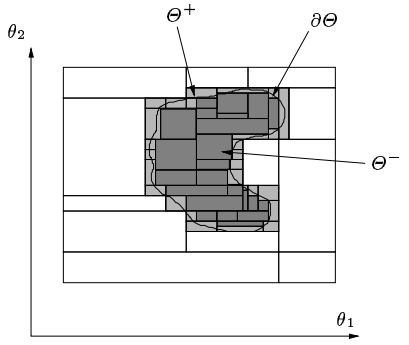

Fig. 3. Feasibility of boxes.

*Example 8 (*Solution characterization by bisection). *Consider again the example 6. The resulting algorithm* Sis *(for Set Inversion Spectra) is the following:*

Algorithm Sis	
Inputs[a]:	$[\check{\boldsymbol{p}}_0], [\check{\boldsymbol{p}}_1], [\check{\boldsymbol{p}}_2], [\check{x}_{1i}], [\check{x}_{2i}], [\check{z}_i], i = 1, \ldots, n$
Init:	$[\check{\boldsymbol{p}}_0] = [\check{\boldsymbol{p}}_1] = [\check{\boldsymbol{p}}_2] = [-10000, 10000]$
Repeat	
Constract	$([\check{\boldsymbol{p}}^{(k)}], [\check{x}], [\check{z}_i], i = 1, \ldots, n)$
	For $i = 1, \ldots, n$
1	If $[p_0^{(k)}] + [p_1^{(k)}][\check{x}_{1i}] + [p_2^{(k)}][[\check{x}_{2i}] \subset [\check{z}_i]$, append $[\boldsymbol{p}]^{(k)}$ to subpaving p^-. Go to step 4
2	If $[p_0^{(k)}] + [p_1^{(k)}][[\check{x}_1] + [p_2^{(k)}][\check{x}_2] \cap [\check{z}_i] = \varnothing$, discard $[\boldsymbol{p}]^{(k)}$. Go to step 4
3	If $w([\boldsymbol{p}]^{(k)}) \leq \rho$, then append $[\boldsymbol{p}]^{(k)}$ to subpaving p^+ else Bisect $[\boldsymbol{p}]^{(k)}$ into $[\boldsymbol{p}']$ and $[\boldsymbol{p}'']$ and stack the two resulting boxes
4	Sis$([\boldsymbol{p}'], [\check{x}_{1i}], [\check{x}_{2i}], [\check{\boldsymbol{z}}])$; Sis$([\boldsymbol{p}''], [\check{x}_{1i}], [\check{x}_{2i}], [\check{z}_i])$.
	EndFor
While the contraction is significant	
output:	$[\hat{\boldsymbol{p}}], [\hat{x}_{1i}], [\hat{x}_{2i}], [\hat{z}_i], 1 \leq i \leq n$;

[a] The symbol $\check{}$ defines prior intervals and $\hat{}$ intrval after contraction.

After completion, the union of all $[\boldsymbol{p}^{(k)}]$'s provide an estimation for the set $\mathbb{P} = \mathcal{F}^{-1}([\boldsymbol{y}]) \cap [\boldsymbol{p}]$. From the list of consistent boxes, it is possible to get an accurate approximation of he smallest domains for the $\boldsymbol{p}$'s and the x_i, z_i's that are consistent with the prior domains $[\boldsymbol{p}]$ and the constraints.

3 Test case: uranium enrichment

3.1 Physics recall

In Nuclear Materials Management and Control, in Safeguards and Waste Management, Non Destructive Assay (NDA) of fissile and fertile material is widely used to evaluate ^{235}U enrichment. The basic metrological parameters of the measurement process involves the use of several $X-$ and $\gamma-$ray peaks identified in the 60 to 200 keV region. The remaining

part of the gamma spectrum is not introduced in the data analysis. This part of the spectrum can contain, however, important information concerning the parameter of interest. Recently active neutron interrogation techniques have been upgraded by measuring in parallel the interrogating neutrons, after passing through the sample to be assayed, (transmission) and the fission neutrons resulting from neutron induced fission (emission) [3]. For measurement methods resulting in one measured signal for the determination of the parameter of interest, the calibration procedure is quiet obvious. The measurement results, however, are sometimes no satisfactory. This is mainly due to the fact that the basic metrological parameters of the process are difficult to identify and hence should be interpreted by multiparametric calibration curves. Examples of such measurement techniques are the determination of the Uranium enrichment based on the detection of the 185,6 keV line emitted during the decay of ^{235}U and active neutron interrogation for the determination of the ^{235}U mass in Uranium bearing sample.
The basic measurement apparatus is a collimated γ-ray detector. Measurements on a set of U_3O_8 and UO_2 samples were performed with a standard measurement system consisting of a Teledyne $3''x3/4$ NaI detector, and a conventional spectroscopic measurement chain. The spectra were accumulated in a 1K memory covering an energy region from 10 keV to 1100 keV. A 20 mm thick Pb collimator with a 20 mm collimating hole in it was used. The resolution was 11,7 % at 185,7 keV and 6,4 % at 1001,0 keV. This region includes the uranium $KX-$rays and $\gamma-$lines from U and its daughters up to the 1001 keV line.
The uranum enrichment E is proportional to the *net* 186 keV count rate R which is given by $R = X_1 - fX_2$, where X_1 and X_2 are two energy regions (this equation represents the substraction of a background from the gross rate in the chosen 186 keV peak energy region), hence we have

$$E = aRF\exp(\mu_C\rho_C t_C) = a\exp(\mu_C\rho_C t_C)(X_1 - fX_2),$$

μ_C, ρ_C are the linear photon absorption coefficient of the sample container at the essay energy, t_C the single wall thickness of the sample container. The factor F reflects the matrix effects. For a fixed geometry ficed spacer the count rate should be linear with enrichemnt only up to 20%. The enrichment can then be written $E = \alpha X_1 + \beta X_2$, where the calibration constant α and β now include the container attenuation and the matrix factor contains all the geometric factor.by measurements of n It has been shown in [10] that the gain problem, the *dead-time* problem and additional spacers have a strong impact on statistical uncertainties of α and β.

3.2 Experimental data

As an illustration of SIS combined with forward-backward propagation, consider the 2-parameters problem of example 6. The PERLA dataset contains 1500 spectra, taken over a period of about one year, measured with a planar detector EG& G type, using the standard NIM electronics [7]. The spectra were taken in various experimental conditions. The spectra cover a wide range of isotopic composition, chemical form, actinide concentration and plutonium/uranium mass.
The quality of the recorded spectra, with respect to stability and resolution are demonstrated in Figure 4.

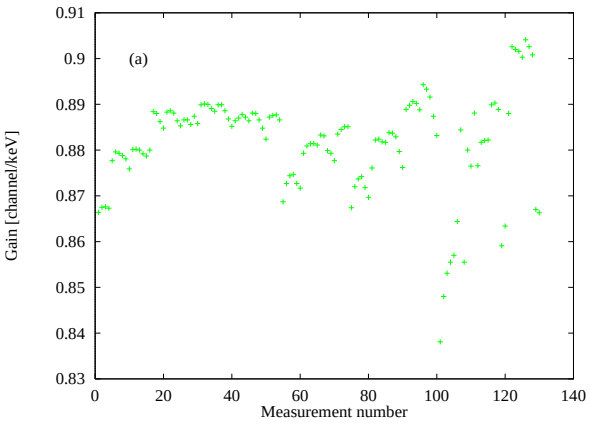

Fig. 4. Dependance of the gain [channel/kev] of the system with the measurement number.

The table 1 is a selection of 56 $U-$samples of the dataset. The value of interest is the ^{238}U concentration (z_i in %), the measured values are 2 the peak areas $x_{1i}, x_{2i}, \quad 1 \leq i \leq 56$ at 185,6 keV and 1001 keV. The uncertainties are almost only based on counting statistics, supposed to be poisonian. This is a highly

z_i	$[x_{1i}^-; x_{1i}^+]$	$[x_{2i}^-; x_{2i}^+]$
94,89%	[113829;144075]	[316901;317272]
94,89%	[129991;158562]	[429135;429425]
94,89%	[131751;161789]	[418386;418684]
⋮		
64,60%	[191945;275871]	[507324;508268]
64,60%	[224105;327653]	[577138; 578305]

Table 1. Enrichment with respect to the 2 peak areas at 185,6 keV and 1001 keV.

favourable problem for using interval analysis.
The isotopic compsition of the samples is summerized in Table 2. The prior boxes for the parameters

Sample	^{234}U	^{235}U	^{236}U	^{238}U
PERLA HEU 35	0,13	35,00	0,27	64,60
PERLA HEU 45	0,34	45,55	0,18	53,93
PERLA LEU 5.0	0,05	5,03	0,04	94,89

Table 2. Uranium isotopic coposition

p_0, p_1 and p_2 may be arbitrarily large, by example $[p_0] = [p_1] = [p_2] = [-1000; 1000]^3$ *i.e. no prior* information is available on the parameter. The algorithm SIS given in section 2.3 can be used to characterize the solution domain $[\boldsymbol{p}]$ such that the whole set of constraints $z_i = p_0 + p_1 x_{1i} - p_2 x_{2i}, \quad 1 \leq i \leq 56$ be satisfied.

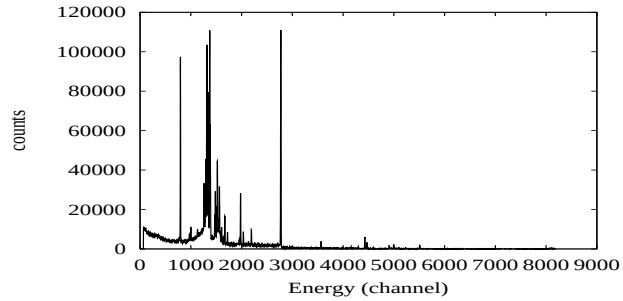

Fig. 5. PERLA Spectra with 61% Pu (File `PU61S28.ASC`).

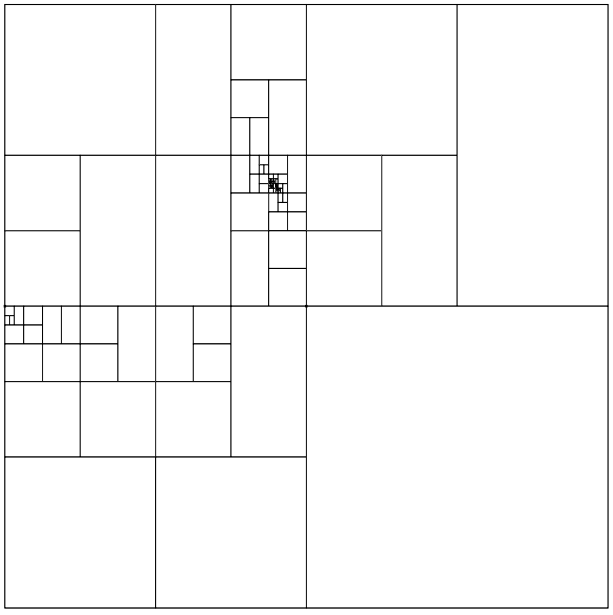

Fig. 6. Contracted domains of the parameters (p_0, p_1) in performing SIS algorithm.

Results After completion, the contracted intervals in the figure 6 include *ALL* the values of the parameters. A large number of bisections have to be performed, but the computing time is about 0,5 seconds in this case. There is 4 distinct solution domains for which the parameters are consistent with respect to the constraints (see equations 9-18).

4 Conclusion

For the first time, this paper studies the application of interval analysis to parameter estimation in spectrometry problems. The so-called SIS algorithm is capable to enclose efficiently all consistent values for the unknown parameter vector inside a box in a error bounded context. An illustrative example has shown the efficiency of the approach. The size of the dataset was forced to be small to compare with classical learning algorithms.

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