Simulation and Modeling of Uncertainties in the Calibration of an Fluorescence Chemical Spectrometer (FRX)

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Abstract— **Sensitivity analysis and uncertainty estimation are of major importance for the declaration of conformity of finished products. Models must be sought to analyze the test data. The main objective of this work is to establish reliable models to analyze our experimental data and validate them. So we have studied and used the Monte Carlo and Bootstrap simulation methods, we have been able to realize programs that calculate the uncertainty according to the ISO 8466 standard on X-ray fluorescence spectrometer samples from the URASM CRTI chemical analysis laboratory. Programs and interfaces are made with Matlab (GUI).**

Keywords—simulation; uncertainties; Monte carlo; Bootstrap; calibration

I. INTRODUCTION

In test laboratories and chemical analyzes of the metallic materials, the quality control of the applications is essential. The analysis of sensibility and the estimation of the uncertainties are of major importance for the declaration of conformity of finished products. It is necessary to analyze the data. The main objective of this work is to establish reliable models to analyze our experimental data and validate them. Equipment calibration involves applying a known standard value to the input of the measurement system to verify that the output corresponds to the expected value. There are several models that are used to adjust the calibration curve. The linear regression model is the best known. During calibration, it is necessary to validate the model. Laboratories are obliged to respect the accuracy of the analysis result. We must look for methods that would allow us to predict the standard deviations of the coefficients from a single measure of the measurement, without repeating the measurements. Uncertainty modeling is a very broad field of study. This requires a good foundation in statistical analysis. A very simple case study is the modeling of a calibration curve with the simple linear regression model, but there are other more complicated ones namely the Monte Carlo method and the Bootstrap method.

Our purpose is to realize the simulation by specific programs, to make a comparison between these methods then apply them to real data outcome of the chemical analyses of steels.

II. PRESENTATION OF THE SIMULATION

Numerical simulation is one of the tools for simulating real phenomena. It relies on the implementation of theoretical models often using mathematical techniques. As well as graphical interfaces allow the display (visualization) of the results (profits) of the calculations by computer generated images. The simulation is performed as follows:

- Generate data which are random variables according to specific models ·
- Execute the experience in a fast and economic way and the improvement of the forecasts.
- Solve the problems of probability and statistics.

To study the finite sample behavior of the test statistics, numerical simulation methods are used, taking advantage of the large computational capacity of the computers, which makes it possible to visualize the results graphically and numerically and thus to predict its behavior and its evolution [1]. Monte Carlo methods are certainly the most common simulation techniques and these are the ones we will use to study the numerical performances of the different tests.

Another alternative approach is developing today using bootstrap methods. The Bootrasp is a method of resampling. It is one of the revolutions of calculation method, it allows us to quantify uncertainties by calculating standard errors and confidence intervals as well as tests.

III. SIMULATION METHODS

A. Algebraic method of distribution of the errors

The purpose of curve fitting is to find the parameters of a mathematical model that describes a set of (usually noisy) data in a way that minimizes the difference between the model and the data. The most common approach is the "linear least squares" method, also called "polynomial least squares"[2].

B. Monte Carlo

The term Monte Carlo is used in many disciplines and refers to procedures where quantities of interest are approximated by generating many random realizations of any stochastic process and calculating any of their mean values [1] (Fig. 1).
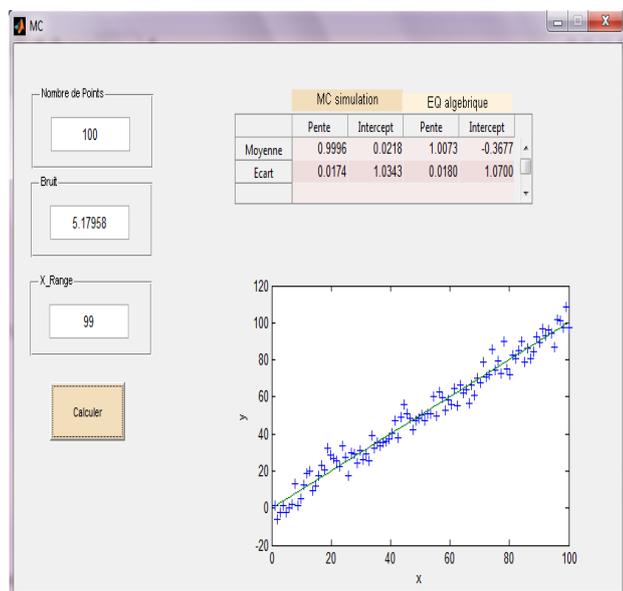

Fig. 1. MC method simulation

C. Bootstrap

Bootstrap: is a technique for performing statistical inference, based on the simulation of data from a limited number of observations, it is intended to facilitate the inference in complex situations where the analytical methods are not enough not [3]. The bootstrap can therefore be applied to any estimator other than the mean such as the median, the correlation coefficient between two random variables or the main eigenvalue of a covariance variance matrix. The bootstrap makes no assumptions about the underlying statistical distribution; hence its generality and power. [4]

D. COMPARISON OF ERROR PREDICTION METHODS

The compar.m program compares these three methods (Monte Carlo simulation, the algebraic method and the bootstrap method) for a linear adjustment of 100 first order points. Each method is repeated on different data sets with the same average slope, interception and random noise, then the SD (SDslope) and intercepts (SDint) were compiled and are shown in the following figure (Fig. 2).

On average, the average standard deviation ("mean SD") of the three methods is fine, but the algebraic and bootstrap methods fluctuate more than the Monte Carlo simulation each time this program is run because they are based on noise in a single 100-point data set, while Monte Carlo simulation reports the average of many data sets. Of course, the algebraic method is simpler and faster to compute than the other methods. However, an algebraic error propagation solution is not always possible, whereas the Monte Carlo and bootstrap methods do not depend on an algebraic solution and can easily be applied to more complicated curve situations, such as least squares non-linear iterative.
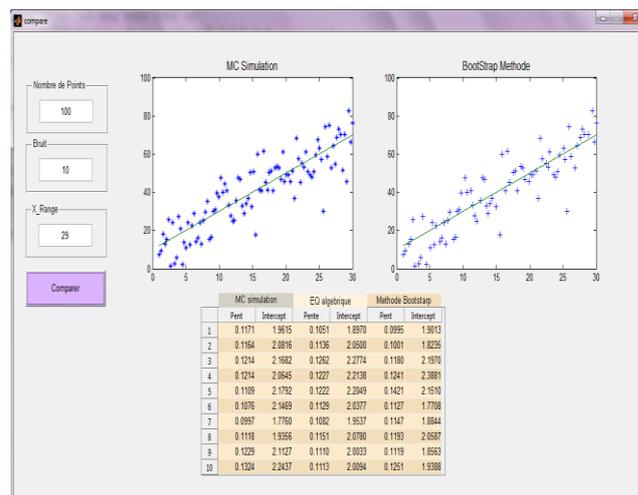

Fig. 2. Comparison of algebric, MC and Bootstrap methods

IV. TOOL OF SIMULATION "GUIDE MATLAB"

The language used to perform the simulation programs is Matlab with its IDE tool dedicated to the creation of graphical interfaces (GUI). This tool, called GUIDE (Graphical User Interface Development Environment), allows to design these graphical interfaces. HMIs (Human Machine Interfaces) are called Graphical User Interfaces (GUIs) in MATLAB. They allow the user, through graphical objects (buttons, menus, check boxes, ...) to interact with a computer program [5]. These interfaces make it easier for users to use programs.

V. APPLICATION ON REAL DATA

We applied the simulation on real data by considering standard samples of an X-ray fluorescence spectrometer from the URASM CRTI chemical analysis laboratory. The computer associated with the spectrometer is equipped with the SIEMENS SPECTRA3000 PC LEO 52X MAX chemical control and analysis software.

The quantitative analysis by X-ray fluorescence spectrometry is a basic analysis method, it allows to determine the quantitative composition of the various elements or combinations that enter the composition of the body to study. It consists of measuring the intensity of the element under study and positioning it on the calibration line drawn from the measurement of appropriate standards. The x-ray tube is powered by the x-ray generator. The x-ray tube's radiation excites the atoms of the sample and generates X-ray fluorescence radiation. The collimator selects a portion of this radiation and directs it onto a crystal. analyzer in the form of a parallel beam according to the BRAGG law:

$$n\lambda = 2d \, \mathrm{Sin}\theta$$

The BRAGG equation is verified for some values of θ. The analyzer crystal disperses the corresponding radiation that a proportional or scintillation counter collects located exactly at the 2θ angular positions. The intensity of wavelength radiation is expressed by the number of wavelength photons emitted in one second. The meters convert these photons into electrical pulses with a certain efficiency. A counting scale integrates for a predetermined time the number of electrical pulses. The result expresses in KCPS, numbers the intensity of a radiation.

A. Statistical evaluation of the linear calibration function

ISO 8466[6] is applicable to methods requiring calibration, it is specially designed for the evaluation of the analytical method and for the calculation of the performance characteristics of the calibration function. In order to obtain comparable analytical results and to serve as a basis for analytical quality control, the calibration and evaluation of analytical methods must be carried out in a uniform manner [7]. We represent some examples and windows of calculation of error with the Monte Carlo method of the program Tab_incert.m realized with Matlab (GUI) (Fig. 3, 4, 5).
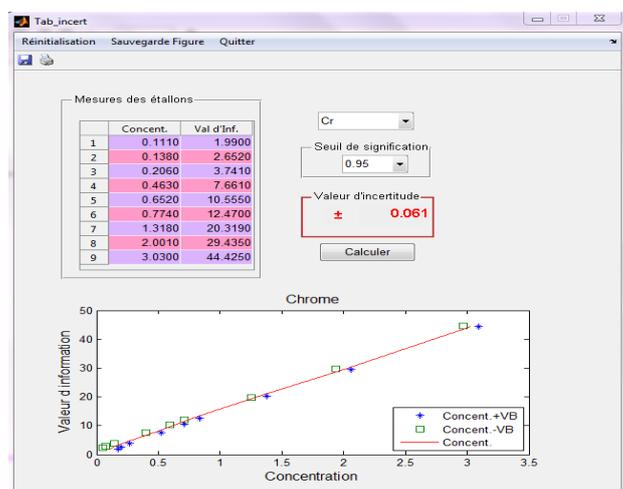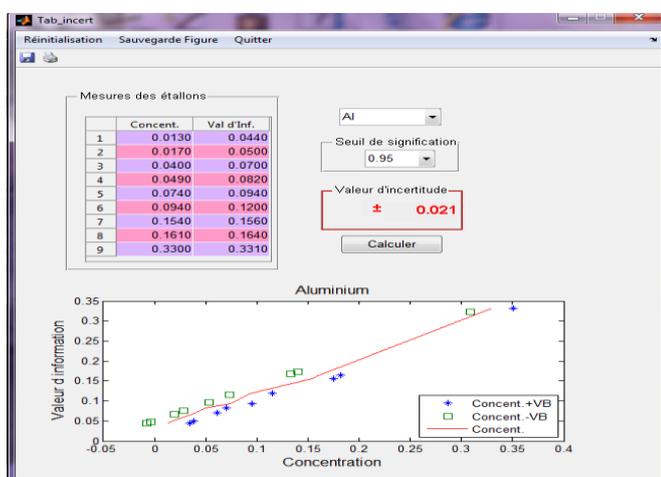

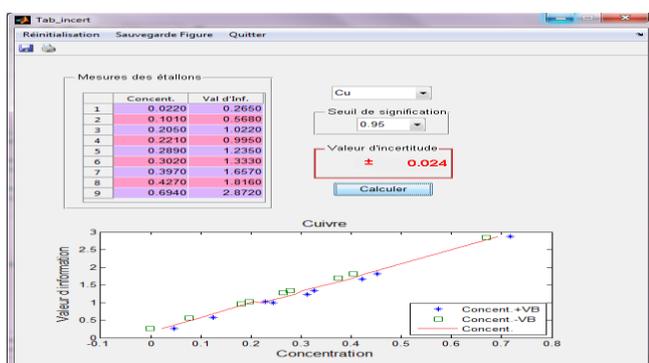Fig. 3. Applying the simulation to real data from an aluminum standard

Fig. 4. Applying the simulation to real data from a Copper standard

Fig. 5. Applying the simulation to real data from a Chrome standard

VI. Conclusion

The Monte Carlo method is a pillar of numerical simulation in statistical physics, nuclear physics and many other fields for the following reasons: it simplifies the calculations of uncertainties when the adjustment by the models is too complex, its efficiency increases when the number of points generated n increases. This MC simulation method does not depend on an algebraic solution and can be easily applied to more complicated curve situations, such as non-linear iterative least squares. The Bootstrap method still needs to be developed and applied to other measuring equipment as well.

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