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DETECTION OF OUTLIERS IN GAS CENTRIFUGE EXPERIMENTAL DATA

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ABSTRACT

Isotope separation in a gas centrifuge is a very complex process. Development and optimization of a gas centrifuge requires experimentation. These data contain experimental errors, and like other experimental data, there may be some gross errors, also known as outliers. The detection of outliers in gas centrifuge experimental data may be quite complicated because there is not enough repetition for precise statistical determination and the physical equations may be applied only on the control of the mass flows. Moreover, the concentrations are poorly predicted by phenomenological models. This paper presents the application of a three-layer feed-forward neural network to the detection of outliers in a very extensive experiment for the analysis of the separation performance of a gas centrifuge.

1. INTRODUCTION

Any experimental data set has an associated error, which may be systematic, random or, in most cases, both. Correct use of these experimental data requires a careful analysis of the associated errors, which indicates the reproducibility, the representativeness and the reliability of the data obtained. However, this analysis can be an arduous task in the case of a very complex process, where uncontrolled or unmonitored parameters influence the results or where large stochastic deviations are observed, resulting in a very dispersed data set [1].

Several procedures have been proposed for the treatment and analysis of experimental data sets, based on statistics or on the physics of the process [2], [3]. Nonetheless, those techniques can be difficult to apply. For instance, when the physics of the process is not very well known, i.e., it cannot be totally represented by means of equations, or when there is little data under similar operational condition. Many processes have these characteristics, such as the experimental investigation and development of ultracentrifugation uranium enrichment, [4].

The detection of gross errors using neural network technique will be performed on an experimental data set from isotope separation tests of ultracentrifuges. These data were originated in the process of evaluation and optimization of the ultracentrifuges developed at the Centro Tecnológico da Marinha em São Paulo (CTMSP) with the collaboration of the Instituto de Pesquisas Energéticas e Nucleares (IPEN). In an isotope separation test of a centrifuge, UF_6 gas with a known isotope composition is fed into the centrifuge, that is operated in different flows and pressures conditions. Similar results, with differences due to technological changes, were published by Zippe [5] and Jordan [6].

2. THEORETICAL BASIS

The numerical estimate of the separation performance of a centrifuge requires the calculation of the internal flow, that is only feasible through mathematical simplifications [7], [8], [9], [10]. On the other hand, it is very difficult to relate the parameters used to describe numerically the countercurrent with the physical devices used in the centrifuge [4], [7], [11].

2.1. Uranium Enrichment with a Gas Ultracentrifuge

A gas ultracentrifuge, is basically a long, thin vertical cylinder (rotor), rotating around its axis at a high velocity, inside a case under vacuum. The process gas, assumed to be a binary isotopic mixture with $^{235}UF_6$ and $^{238}UF_6$, inside the rotor is subjected to centrifugal force, thousands times stronger than gravity. A pressure gradient is established in the radial direction, increasing from the center to the rotor wall. That pressure distribution is slightly dissimilar for the different isotopes because it is proportional to mass. This results in a partial separation in the radial direction. A countercurrent axial flow multiplies the radial separation.

The separation performance of a gas ultracentrifuge depends on [12], [13], (a) the characteristics of the ultracentrifuge, given by the angular velocity, length and diameter of the rotor; (b) the operational conditions, given by the feed and extraction mass flows and by the mass hold-up (indirectly measured by the pressure at the product or at the waste extraction); (c) the axial feed position inside the rotor and (d) the intensity and profile of the countercurrent axial flow. The countercurrent flow is induced by mechanical drives as the rotating gas hits stationary obstacles inside the rotor (scoop or baffles) and by thermal drives, e.g., different temperatures at the end caps and along the rotor wall.

3. EXPERIMENTAL DATA

The bench plant is composed of an ultracentrifuge and a UF_6 container, interconnected by pipes and valves, where instruments and control valves are properly located to control and to monitor the whole process of injection and extraction of the process gas UF_6 in the ultracentrifuge. The operational condition is defined by the pressures in the feed p_F, product p_P and waste p_W lines; by the feed flow rate F and extractions of the product P and waste W, or the cut $\theta = P/F$. For each operational condition, product and waste samples are collected and sent to the mass spectrometer to determine their abundance ratios, R_P and R_W, respectively. An isotope separation test consists in the operation of an ultracentrifuge under different operational conditions, defined by three of the process variables (feed flow rate F, product pressure p_P and the cut θ). Thus, several groups of data are generated and each of them is denominated a separation experiment. They are composed of the controlled variables that define the operational condition; the other process variables measured, which are the observed answers response; and the respective separation parameters calculated from the experimental values. These experiments can be divided into groups of experiments obtained under similar conditions [11], [14].

Seven hundred and ninety-one experiments were performed; however some of them were eliminated due to insufficient material for sample analysis, resulting in 764 experiments. The test was programmed to cover the domain of interest of the controlled variables, consisting of eight values for feed flow F, seven values for cut θ and five values for pressure p_P. These data were organized and analyzed according to usual statistical procedures, taking into consideration the macroscopic mass balance, cluster analysis and the statistical analysis of the errors in the experimental results, as described below. Then, a neural network technique was employed.

3.1. Macroscopic Mass Balance

During the isotope separation test, the macroscopic mass balance was constantly verified by comparing the cut obtained through measurement of product and waste flows, and the cut obtained from the abundance ratio resulting from Eq. (1):

$$\theta = \frac{(R_F - R_W)}{(R_P - R_W)} \cdot \frac{(R_P + 1)}{(R_F + 1)} \tag{1}$$

Experiments where the difference between these two values of cut was greater than 0.03 were rejected and repeated latter. This verification checks the possibility of an error in the sampling procedure, which uses an external element (the sample vessel) and bypasses the material flow coming from the ultracentrifuge to the sampling system, that may disturb the steady-state ultracentrifuge condition. It also checks the calibration of some instruments.

This procedure aimed to the experiments with gross errors. Those 176 experiments from the original data set were excluded. Thus, the experimental data set remained with 588 experiments.

3.2. Statistical Analysis of the Errors

3.2.1. Statistical analysis by hypothesis test

The 588 remaining experiments were clustered by process primary variables F and θ, then $\mathbf{R_P}$ and $\mathbf{R_W}$ averages were obtained. The error of the experimental data was calculated in relation to these averages. For example, if a given group has three samples (three experiments), one has a $\mathbf{R_P}$ average of $\mathbf{R_{Pavgj}}$ and $\mathbf{e_{Rpi}}$ is the difference between $\mathbf{R_{Pi}}$ and $\mathbf{R_{Pavgj}}$. The same approach for $\mathbf{R_W}$ was followed. The analysis was then conducted for the errors in $\mathbf{R_P}$ ($\mathbf{e_{Rp}}$) and in $\mathbf{R_W}$ ($\mathbf{e_{Rw}}$). The resulting errors were assumed to be a random sample of normal random variables $\mathbf{e_{Rp}}$ and $\mathbf{e_{Rw}}$, where $\mathbf{e_{Rpavg}}$ and $\mathbf{e_{Rwavg}}$ are the averages. The statistical test, referred to as the extreme deviate statistic, involves the difference between the extreme value and the sample mean value, where $\mathbf{Y_{Rp}}$ and $\mathbf{Y_{Rw}}$ are defined as the residual.

Statistical analysis was performed based on a hypothesis test for means, which considers a confidence interval estimate with a confidence level of 95%, and Anscombe suggests the rules, giving $\boldsymbol{c}$ explicitly in terms of the F distribution in Himmelblau [2].

Using this procedure, 15 experiments were suspected of contain gross errors in $\mathbf{e_{Rp}}$**,** and nine in $\mathbf{e_{Rw}}$**.**

4. NEURAL NETWORKS

The application of neural networks for the simulation of chemical and nuclear processes, specifically for isotope separation with the gas ultracentrifuge, is of great interest due to the nonlinearity of these processes [4], [11], [14]. The success of this kind of modeling depends heavily on knowledge of the main variables affecting the process and the availability of a good data base with the necessary information on the desired domain. This work uses this technique to identify the outliers based on the neural network scheme of modeling defined by Migliavacca [4]. It is based on the idea that experimental points that are more difficult to be predicted by NN after few thousands presentations, may be outliers. The software for training the neural network was developed in FORTRAN by the Laboratório de Simulação e Controle de Processos do Departamento de Engenharia Química da Universidade de São Paulo [15]. All data were included in the training data set and during the training process several thousand iteration were performed. Figures 3 and 4 show the error between the experimental and calculated values of Rp and Rw, respectively, in sequential order.

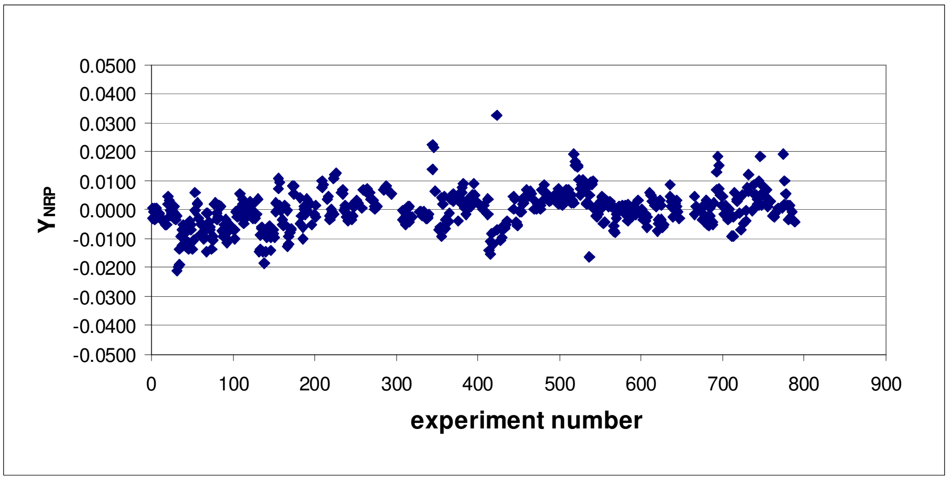

Figure 3. Residual $\mathbf{Y_{Rp}}$ of the experimental data in sequential order

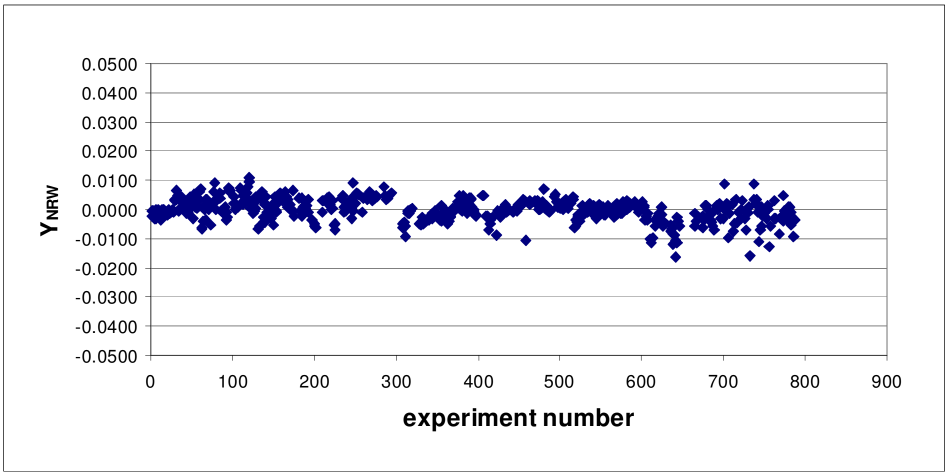

Figure 4. Residual Y_{Rw} of the experimental data in sequential order

The execution of the experiments took several months, and a worsening of the calibration of some instruments (the flow meters) occurred. Around the experiments coded 294 to 308, a severe error was detected through verification of the mass balance, and thus, the instruments were recalibrated. The same problem occurred after experiment number 604. These problems were also identified when using the neural network. These graphics show three different regions of experiments. Thus, the data set was divided into three groups of experiments, for the three different bias observed in the sequential experimental residues:

- First group: from experiment number 1 to 294
- Second group: from experiment number 308 to 604
- Third group: from experiment number 605 to 788

Table 1. Hypothesis test for means

Groups	1		2		3	
N° of points	239		236		113	
C	3.100919		3.100468		3.061339	
Variable	e_{NRP}	e_{NRW}	e_{NRP}	e_{NRW}	e_{NRP}	e_{NRW}
Minimum	-0.0143	-0.01022	-0.02127	-0.01051	-0.01102	-0.01463
Maximum	0.01175	0.00871	0.02701	0.0089	0.01534	0.01132
Mean	-0.001195	-0.00656	0.000581	0.000354	0.000643	0.000192
Std dev S	0.004680	0.003247	0.005261	0.002743	0.004835	0.004235
Mean + c*S	0.013325	0.009412	0.015731	0.008857	0.015444	0.013155
Mean - c*S	-0.015715	-0.010724	-0.016893	-0.008149	-0.014158	-0.012771

The points where errors appear to be scattered far from the majority are probably outliers [16]. The residues were analyzed applying statistical test described above, according to Himmelblau [2]. The results are presented in Table 1.

Applying the neural network to the first group of the data set, corrected previously to include a bias, no suspect experiment (with gross error) was detected. Using the same methodology for the second group, seven experiments with gross error in the first run were detected, as shown in Figures 5 and 6.

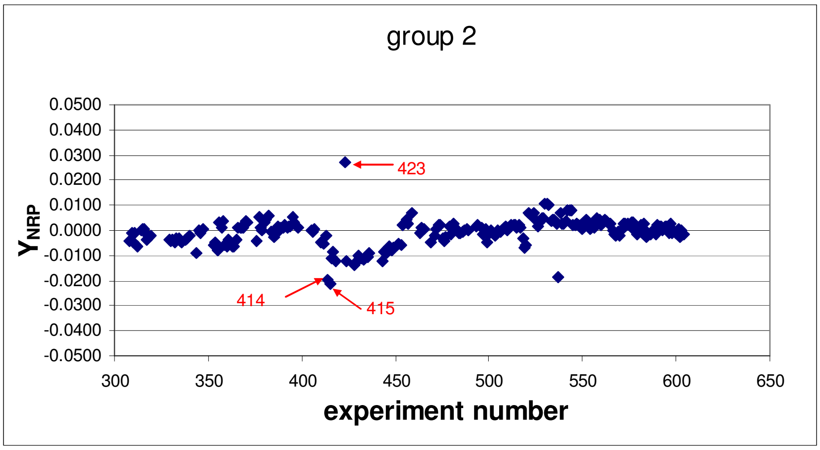

Figure 5. Residual Y_{Rp} of the experimental data in sequential order for the second group in the first run

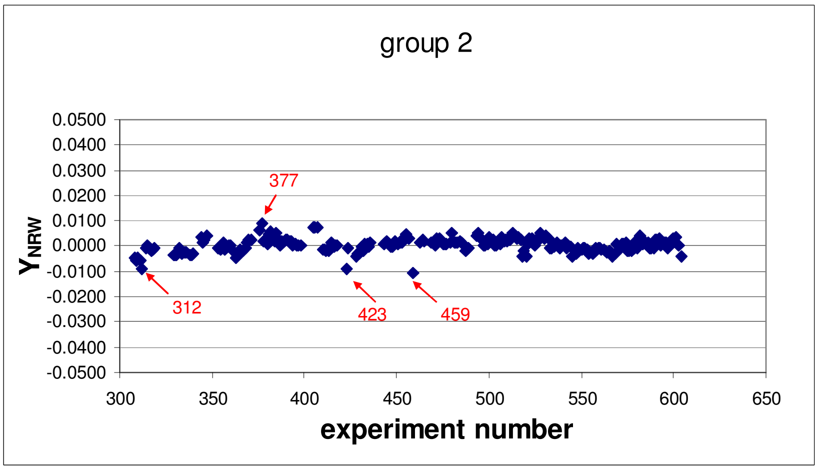

Figure 6. Residual Y_{Rw} of the experimental data in sequential order for the second group in the first run

Seven points were excluded from the data set and then the neural network was applied again. At the second run, two additional experiments containing gross error were identified and eliminated from the data set. The same procedure was followed until no experiment with gross error was detected. A total of 13 experiments were eliminated from the second group. At the third group the experimental error was higher than it was at the two other groups. In this case, only one experiment was eliminated.

Thus, after the neural network analysis of the three groups, a new, unique data set was formed with 574 experiments. This means that 14 suspect experiments were eliminated. Four of them are justified by statistical analysis. Four others experiments, that were identified in the first run, can be justified by small deviation at the steady state operational conditions. The neural network approach showed the capability to identify two different classes of errors: instrument bias and outliers.

5. CONCLUSIONS

The neural network model has been shown to be a very attractive tool for identifying systematic and gross error. The statistical methods applied could identify some experiments suspected of being outliers. A direct comparison between the statistical analysis and neural network analysis was not possible, since only the NN was able to identify groups with different bias. Statistical analysis of the points with bias is not reliable.

The operation of an ultracentrifuge is a very complex process. Under many conditions high variability may occur. Some abnormal results may appear and can be misleading with gross error. So, all the results from any technique employed must be verified by the research staff.

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