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# MATHEMATICAL MODELLING OF THE CONSTITUENTS' CONCENTRATIONS OF AISI 304 STAINLESS STEEL SAMPLES THAT DIFFUSE INTO SIMULATED ACIDIC ENVIRONMENTS

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**Abstract:** *The purpose of this research was to study the mathematical modelling of dependence between testing parameters and some metallic elements diffused into simulated acidic environments. Diffusion processes occurring at the contact between AISI 304 stainless steel samples and simulated acidic environments were analyzed to fulfill this goal. In order to process the experimental data by statistical and mathematical methods, the following steps were taken: diffusion testing of Cr, Mn, $^{56}$Fe and Ni elements from AISI304 stainless steel samples into solutions with concentration of 3%, 6% and 9% acetic acid; chemical analysis of corrosive solutions using mass spectrometry and inductively coupled plasma method (ICP-MS); the results were processed using ANOVA method and thus resulting the mathematical models that describe the dependence between variables. A polynomial model with independent variables was used to obtain the mathematical models: temperature of acidic simulated solutions ($X_1$), testing time ($X_2$), stirring grade of environment ($X_3$) and the dependent variables (response function): $Y_1$ - concentration of chromium (Cr), $Y_2$ - concentration of manganese (Mn), $Y_3$ - concentration of iron ($^{56}$Fe) and $Y_4$ - concentration of nickel (Ni) found in corrosive environments. After having made the analysis of variance ANOVA, a model having the lowest P value (critical probability) for all variables was chosen. The regression coefficient was determined to verify the validity of each mathematical model.*

**Keywords:** *stainless steel, simulated environments, diffusion, ANOVA method, mathematical model.*

## 1. Introduction

It is known that food raw materials have their own natural metal content. In case when there are additions to this natural content, the diffusion from metal surfaces coming into direct contact can cause serious health problems or a change unwanted organoleptic characteristics of the finished product. These metals are found in absolutely all foodstuffs in lower or higher concentrations, depending on various circumstances [1]. A mathematical model that describes the physical processes which occur at the contact of some material from the food processing chain is considered a very important tool that can replace, at least partially, the experimental investigations which are expensive and require longer time. The models thus obtained aimed at complying with the conformity assessment of specific regulations concerning migration limits and at describing the time variation of the values of parameters. Having in view the advantages of this tool, in recent years, numerous studies have been undertaken for this purpose [2-7]. The research described in the literature highlights a relatively limited approach of the diffusion systems consisting of materials intended to come into contact with foodstufs.

Firstly, it is useful to present a schematic illustration of the mathematical formulation stages of this problem and the finding of adequate solutions is shown in Figure 1 [8].

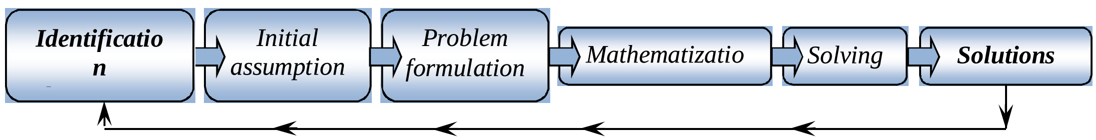


**Fig. 1. The problem formulating stages and finding solutions [8]**

The general mathematical model solutions thus obtained provide a quantitative and qualitative perspective on how the parameters affect the phenomenon; further research could be focused on the application of these basic equations [8].

The aim of this work was to study the mathematical modelling of dependence between the constituents' concentrations of AISI 304 stainless steel samples that diffuse into simulated acidic solutions and the testing parameters. Similar researches have been conducted for advanced characterization behavior of AISI321 stainless steel samples in acidic environments [4].

## 2. Matherials and methods

### *2.1. Metallic samples*

In this research metallic samples made of AISI304 stainless steel grade were used. The samples sizes were of 40±0.5 x 40±0.5 x 1 mm and they were established by the Ministerial Decree of 21.03.1973, which stipulates that the ratio of exposed surface of the stainless steel samples and solution volume should be between 0.5 ... 2 [9].

The chemical composition of metallic samples (according to the *SR EN 10088-2:2005* Romanian standard) is shown in Table 1.

**Table 1**

**Chemical composition of AISI304 stainless steel samples (wt %)**

| Fe | C | Mn | P | S | Si | Cr | Ni |
|---|---|---|---|---|---|---|---|
| 67 | 0.07 | 1-2 | 0.045 | 0.03 | 0-1 | 17-19 | 8-10.5 |

A systeM *µScan* (manufactured by *NanoFocus* - Germany) was used to determine roughness of the metallic samples. Measurement and calculation of usual surface parameters were made according to *DIN EN ISO 4287* and *DIN EN ISO 4288* standards. The surface mean roughness of AISI304 stainless steel samples was $\overline{R_a}$ = 0.598875±0.0125 $\mu m$.

### *2.2. Corrosive environments*

All solutions were freshly prepared with quality analytical chemical reagents. Given the fact that acidic environments are one of the most aggressive environments in the food processing industry, within the experiment we used 3%, 6% and 9% $CH_3COOH$ in double distilled water (according to Italian D. M. of 21-03-1973) [9]. Glacial acetic acid (*Sigma-Alorich*, Germany) was used to prepare the experimental solutions.

### *2.3. Testing method*

To cover a domain as broad as that of using AISI304 stainless steel, three testing parameters were chosen. To determine the variation levels of testing parameters, the usual values encountered in the processing industry were considered.

The levels of the three parameters used in the experiment are shown in Table 2.

**Table 2**

**Variation levels of testing parameters**

| Levels / Parameters | Minimum level | Central level | Maximum level |
|---|---|---|---|
| **Temperature solution,** [°C] | 22 | 28 | 32 |
| **Testing time,** [min] | 30 | 60 | 90 |
| **Stirring environment,** [r/min] | 0 | 125 | 250 |

The samples were kept in the oven to make the experiments in stationary environment and at temperatures different from the room temperature one (*22°C*). A magnetic heat stirrer *Heidolph MR Hei-Tec* (Heidolph, Germany) was used to make the experiments at the temperature of *28°C*, *34°C* respectively and in stirred environment.

### *2.4. Chemical analysis of corrosive environments*

Inductively coupled plasma mass spectrometry (ICP-MS) was used to analyze the chemical composition of corrosive environments, both before having used them in the corrosion tests, and after having made them, taking into consideration the adavantages of this method as compared to other analytical ones.

### *2.5. Mathematical modelling of the constituents' concentrations*

ANOVA method, known as *the variance analysis* was appealed to in order to find a mathematical model as accurate as possible which describe the diffusion phenomena.

This technique was chosen due to the fact that it is highly recommended when studying a larger number of levels of independent variables, providing higher accuracy of the effect that the independent variables have upon the dependent ones as well as of the their joint effect.

Having in view that some practical values (minimum values) of responses are to be determined, it is necessary to establish some interdependences capable of describing both the nature and the extent of the influences taken into consideration (see also [4]).

To develop the predictive-mathematical models the *Design Expert* software (trial version) was used.

In order to obtain the mathematical models a polynomial model with independent variables was used: acidic simulated solutions temperature ($X_1$), testing time ($X_2$), stirring grade of environment ($X_3$) and the dependent variables (response function): $Y_1$ - concentration of chromium (*Cr*), $Y_2$ - concentration of manganese (*Mn*), $Y_3$ - concentration of iron ($^{56}Fe$) and $Y_4$ - concentration of nickel (*Ni*) found in corrosive environments. After having made the analysis of variance ANOVA, a model having the lowest P value (*critical probability*) for all variables was chosen. The regression coefficient was determined to verify the validity of each mathematical model.

## 3. Results and discussion

The chemical analysis of corrosive environments was intended to determine the concentration of Cr, Mn, Fe and Ni elements migrated from AISI304 stainless steel samples.

### *3.1. Mathematical modelling of metallic constituent concentrations migrated into 3% $CH_3COOH$ solutions*

The statistical summary of mathematical models is shown in Table 3.

**Table 3**

**Summary statistic of mathematical models proposed for the dependent variables**

| *Dependent variable* | *P* | *Model* | *Standard deviation [σ]* | *R-Squared [$R^2$]* | *Adjusted R-Squared [$R^2$ adjusted]* |
|---|---|---|---|---|---|
| $Y_1$ | 0.0070 | Quintic | 8.59 | 0.9967 | 0.9783 |
| $Y_2$ | 0.0190 | Quintic | 1.71 | 0.9964 | 0.9765 |
| $Y_3$ | 0.0490 | Quintic | 323.04 | 0.9910 | 0.9300 |
| $Y_4$ | 0.0099 | Quintic | 7.82 | 0.9964 | 0.9765 |

The mathematical model that defines the variation of chromium concentration (dependent variable) depending on the testing parameters (independent variables) is shown in Equation 1.

$$Y_1 = 7.96 + 3.83 \cdot X_1 - 3.72 \cdot X_2 - 1.28 \cdot X_3 + 3.06 \cdot X_1^2 + 6.06 \cdot X_2^2 + 3.06 \cdot X_3^2 + 17.75 \cdot X_1 \cdot X_2 - 45.25 \cdot X_1 \cdot X_3 - 0.25 \cdot X_2 \cdot X_3 + 25.08 \cdot X_1^2 \cdot X_2 + 48.92 \cdot X_1^2 \cdot X_3 + 15.25 \cdot X_2^2 \cdot X_3 + 9.58 \cdot X_2 \cdot X_3^2 - 9.50 \cdot X_1 \cdot X_2 \cdot X_3 + 5.42 \cdot X_1^2 \cdot X_2^2 + 30.25 \cdot X_1^2 \cdot X_2 \cdot X_3 + 43.42 \cdot X_1^2 \cdot X_3^2 + 21.00 \cdot X_1 \cdot X_2^2 \cdot X_3 - 26.25 \cdot X_1 \cdot X_2 \cdot X_3^2 - 9.58 \cdot X_2^2 \cdot X_3^2 \quad (1)$$

In order to check the validity of the mathematical model proposed, the *experimental values* vs *model values* were plotted (Figure 2).

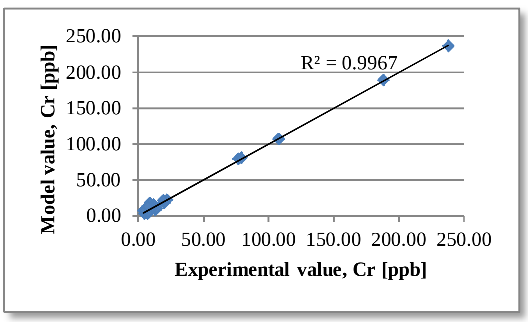


**Fig. 2. Validity check of the $Y_1$ (*chromium concentration*) mathematical model**

The mathematical model of the $Y_2$ response function is shown in Equation 2.

$$Y_2 = 4.04 - 0.59 \cdot X_1 - 0.35 \cdot X_2 + 1.39 \cdot X_3 + 0.38 \cdot X_1^2 + 0.68 \cdot X_2^2 + 0.18 \cdot X_3^2 + 3.18 \cdot X_1 \cdot X_2 - 8.20 \cdot X_1 \cdot X_3 + 0.30 \cdot X_2 \cdot X_3 + 4.18 \cdot X_1^2 \cdot X_2 + 8.98 \cdot X_1^2 \cdot X_3 + 4.24 \cdot X_1 \cdot X_2^2 - 7.88 \cdot X_1 \cdot X_3^2 - 0.47 \cdot X_2^2 \cdot X_3 + 1.95 \cdot X_2 \cdot X_3^2 - 0.63 \cdot X_1 \cdot X_2 \cdot X_3 + 0.83 \cdot X_1^2 \cdot X_2^2 + 4.02 \cdot X_1^2 \cdot X_2 \cdot X_3 + 8.35 \cdot X_1^2 \cdot X_3^2 + 3.43 \cdot X_1 \cdot X_2^2 \cdot X_3 - 2.73 \cdot X_1 \cdot X_2 \cdot X_3^2 - 0.40 \cdot X_2^2 \cdot X_3^2 \quad (2)$$

The validity of the $Y_2$ mathematical model proposed was plotted by *experimental values* vs *model values* (Figure 3).

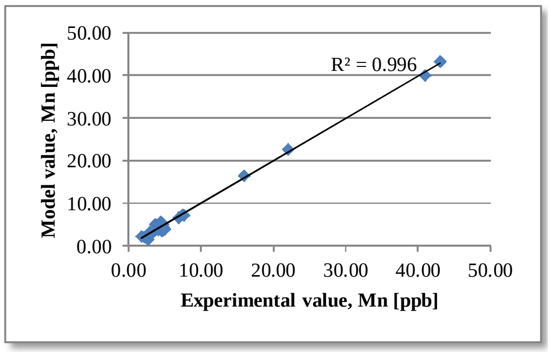


**Fig.3. Validity check of the $Y_2$ (*manganese concentration*) mathematical model**

The mathematical model of the $Y_3$ response function is shown in Equation 3.

$$Y_3 = 394.07 - 88.33 \cdot X_1 - 1.67 \cdot X_2 + 116.67 \cdot X_3 - 56.11 \cdot X_1^2 + 123.89 \cdot X_2^2 - 1.11 \cdot X_3^2 + 437.50 \cdot X_1 \cdot X_2 - 887.50 \cdot X_1 \cdot X_3 + 0.002 \cdot X_2 \cdot X_3 + 372.50 \cdot X_1^2 \cdot X_2 + 1012.50 \cdot X_1^2 \cdot X_3 + 637.50 \cdot X_1 \cdot X_2^2 - 762.50 \cdot X_1 \cdot X_3^2 - 115 \cdot X_2^2 \cdot X_3 + 55 \cdot X_2 \cdot X_3^2 + 430 \cdot X_1 \cdot X_2 \cdot X_3 + 194.17 \cdot X_1^2 \cdot X_2^2 + 305 \cdot X_1^2 \cdot X_2 \cdot X_3 + 1019.17 \cdot X_1^2 \cdot X_3^2 + 772.5 \cdot X_1 \cdot X_2^2 \cdot X_3 + 52.50 \cdot X_1 \cdot X_2 \cdot X_3^2 - 253.3 \cdot X_2^2 \cdot X_3^2 \quad (3)$$

The validity of the $Y_3$ mathematical model proposed was plotted by *experimental values* vs *model values* (Figure 4).

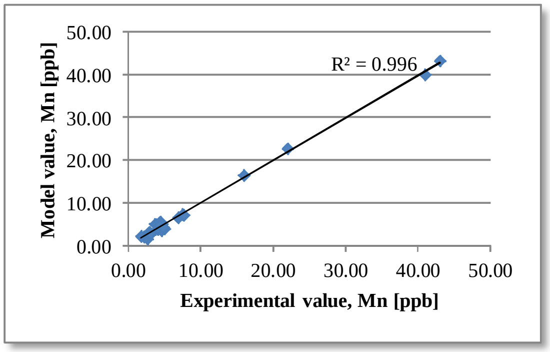


**Fig. 4. Validity check of the $Y_3$ (*iron concentration*) mathematical model**

The mathematical model of the $Y_4$ response function is shown in Equation 4.

$$Y_4 = 20.44 \cdot X_1 - 8.39 \cdot X_2 + 2.00 \cdot X_3 - 7.56 \cdot X_1^2 - 0.86 \cdot X_2^2 + 0.64 \cdot X_3^2 + 15.25 \cdot X_1 \cdot X_2 - 24.75 \cdot X_1 \cdot X_3 - 2.25 \cdot X_2 \cdot X_3 + 26.83 \cdot X_1^2 \cdot X_2 + 37.50 \cdot X_1^2 \cdot X_3 + 8.60 \cdot X_1 \cdot X_2^2 - 37.40 \cdot X_1 \cdot X_3^2 + 9.00 \cdot X_2^2 \cdot X_3 + 10.83 \cdot X_2 \cdot X_3^2 - 13.63 \cdot X_1 \cdot X_2 \cdot X_3 + 14.23 \cdot X_1^2 \cdot X_2^2 + 28.88 \cdot X_1^2 \cdot X_2 \cdot X_3 + 34.23 \cdot X_1^2 \cdot X_3^2 + 2.63 \cdot X_1 \cdot X_2^2 \cdot X_3 - 23.88 \cdot X_1 \cdot X_2 \cdot X_3^2 + 4.03 \cdot X_2^2 \cdot X_3^2 \quad (4)$$

The validity of the $Y_4$ mathematical model proposed was plotted by *experimental values* vs *model values* (Figure 5).

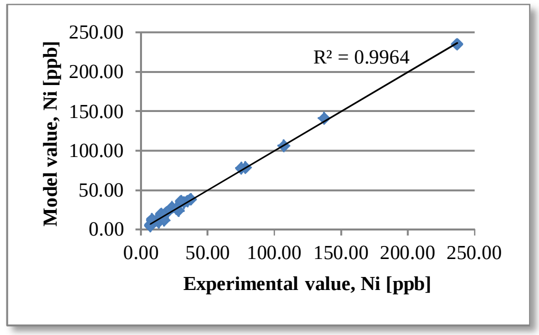


**Fig. 5. Validity check of the $Y_4$ (*nickel concentration*) mathematical model**

### *3.2. Mathematical modelling of metallic constituent concentrations migrated into 6% $CH_3COOH$ solutions*

Statistical summary of mathematical models to describe the found dependent variables Cr, Mn, $^{56}Fe$ and Ni is shown in Table 4.

**Table 4**

**Summary statistic of mathematical models proposed for the dependent variables**

| *Dependent variable* | *P* | *Model* | *Standard deviation [σ]* | *R-Squared [$R^2$]* | *Adjusted R-Squared [$R^2$ adjusted]* |
|---|---|---|---|---|---|
| $Y_1$ | 0.0400 | Cubic | 2.67 | 0.9618 | 0.9006 |
| $Y_2$ | 0.0480 | Quintic | 0.19 | 0.9999 | 0.9978 |
| $Y_3$ | 0.0015 | Quintic | 17.25 | 0.9979 | 0.9866 |
| $Y_4$ | 0.0018 | Quintic | 27.76 | 0.9890 | 0.9553 |

The mathematical models that define the studied response functions (dependent variables) depending on testing parameters (independent variables) are presented in the following equations.

$$Y_1 = 19.67 + 2.50 \cdot X_1 + 7{,}72 \cdot X_2 + 4{,}28 \cdot X_3 - 4.33 \cdot X_1^2 + 2.00 \cdot X_2^2 + 0.17 \cdot X_3^2 + 0.083 \cdot X_1 \cdot X_2 1.50 \cdot X_1 \cdot X_3 + 3.00 \cdot X_2 \cdot X_3 - 3.58 \cdot X_1^2 \cdot X_2 - 0.67 \cdot X_1^2 \cdot X_3 + 3.25 \cdot X_1 \cdot X_2^2 - 1.00 \cdot X_1 \cdot X_3^2 + 2.83 \cdot X_2^2 \cdot X_3 - 0.33 \cdot X_2 \cdot X_3^2 + 2.13 \cdot X_1 \cdot X_2 \cdot X_3 \quad (5)$$

The validity of the $Y_1$ mathematical model proposed was plotted by *experimental values vs model values (Figure 6).*

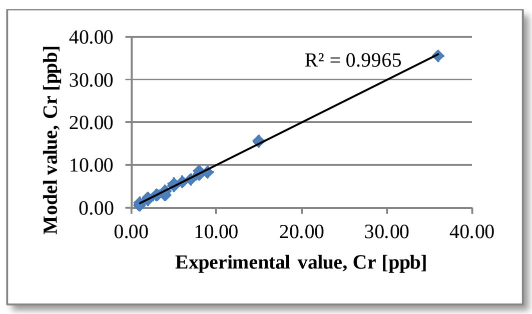


**Fig. 6. Validity check of the $Y_1$ (*chromium concentration*) mathematical model**

The mathematical model of the $Y_2$ (Mn concentration) response function is shown in Equation 6.

$$Y_2 = 3.87 - 1.60 \cdot X_1 + 4.70 \cdot X_2 + 0.75 \cdot X_3 + 0.54 \cdot X_1^2 + 3.04 \cdot X_2^2 - 1.11 \cdot X_3^2 + 0.30 \cdot X_1 \cdot X_2 + 3.05 \cdot X_1 \cdot X_3 + 1.92 \cdot X_2 \cdot X_3 - 4.25 \cdot X_1^2 \cdot X_2 - 3.10 \cdot X_1^2 \cdot X_3 + 1.75 \cdot X_1 \cdot X_2^2 - 1.55 \cdot X_1 \cdot X_3^2 + 1.72 \cdot X_2^2 \cdot X_3 - 1.73 \cdot X_2 \cdot X_3^2 + 1.83 \cdot X_1 \cdot X_2 \cdot X_3 - 4.77 \cdot X_1^2 \cdot X_2^2 - 0.32 \cdot X_1^2 \cdot X_2 \cdot X_3 + 2.83 \cdot X_1^2 \cdot X_3^2 - 1.42 \cdot X_1 \cdot X_2^2 \cdot X_3 + 1.15 \cdot X_1 \cdot X_2 \cdot X_3^2 - 0.49 \cdot X_2^2 \cdot X_3^2 + 2.68 \cdot X_1^2 \cdot X_2^2 \cdot X_3 + 3.00 \cdot X_1^2 \cdot X_2 \cdot X_3^2 + 2.85 \cdot X_1 \cdot X_2^2 \cdot X_3 \quad (6)$$

The validity of the $Y_2$ mathematical model proposed was plotted by *experimental values vs model values (Figure 7)*.

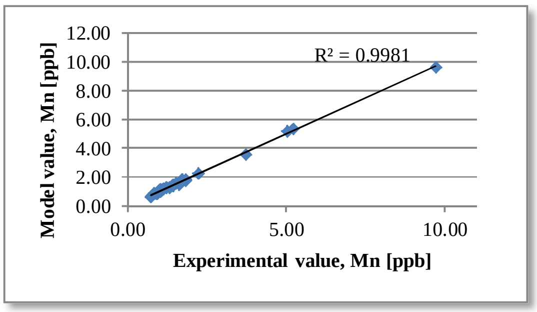


**Fig. 7. Validity check of the $Y_2$ (*manganese concentration*) mathematical model**

The mathematical model of the $Y_3$ ($^{56}Fe$ concentration) response function is shown in Equation 7.

$$Y_3 = 167.41 - 16.11 \cdot X_1 + 143.33 \cdot X_2 + 80.00 \cdot X_3 + 123.89 \cdot X_1^2 + 128.89 \cdot X_2^2 + 18.89 \cdot X_3^2 - 70 \cdot X_1 \cdot X_2 - 72.50 \cdot X_1 \cdot X_3 - 12.50 \cdot X_2 \cdot X_3 - 90 \cdot X_1^2 \cdot X_2 + 57.50 \cdot X_1^2 \cdot X_3 + 31.67 \cdot X_1 \cdot X_2^2 - 0.83 \cdot X_1 \cdot X_3^2 - 2.50 \cdot X_2^2 \cdot X_3 - 22.50 \cdot X_2 \cdot X_3^2 + 0.002 \cdot X_1 \cdot X_2 \cdot X_3 - 88.33 \cdot X_1^2 \cdot X_2^2 + 10 \cdot X_1^2 \cdot X_2 \cdot X_3 - 5.83 \cdot X_1^2 \cdot X_3^2 + 152.50 \cdot X_1 \cdot X_2^2 \cdot X_3 - 5.00 \cdot X_1 \cdot X_2 \cdot X_3^2 - 5.83 \cdot X_2^2 \cdot X_3^2 \quad (7)$$

The validity of the $Y_3$ mathematical model proposed was plotted by *experimental values vs model values (Figure 8)*.

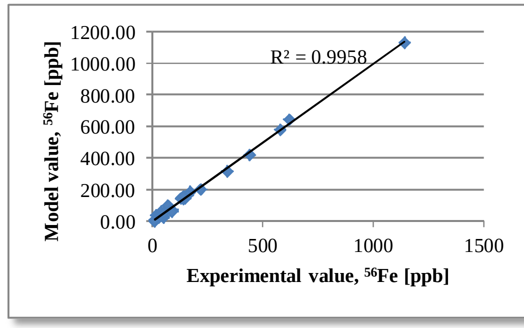


**Fig. 8. Validity check of the $Y_3$ (*iron concentration*) mathematical model**

The mathematical model of the $Y_4$ (Ni concentration) response function is shown in Equation 8.

$$Y_4 = 311.44 - 24.67 \cdot X_1 - 181.50 \cdot X_2 + 41.17 \cdot X_3 + 7.50 \cdot X_1^2 + 24.33 \cdot X_2^2 - 8.83 \cdot X_3^2 - 69.25 \cdot X_1 \cdot X_2 + 6.33 \cdot X_1 \cdot X_3 - 15.67 \cdot X_2 \cdot X_3 + 97.25 \cdot X_1^2 \cdot X_2 - 1.50 \cdot X_1^2 \cdot X_3 + 35.75 \cdot X_1 \cdot X_2^2 - 8.00 \cdot X_1 \cdot X_3^2 + 7.50 \cdot X_2^2 \cdot X_3 + 12.50 \cdot X_2 \cdot X_3^2 - 7.38 \cdot X_1 \cdot X_2 \cdot X_3 \quad (8)$$

The validity of the $Y_4$ mathematical model proposed was plotted by *experimental values vs model values (Figure 9)*.

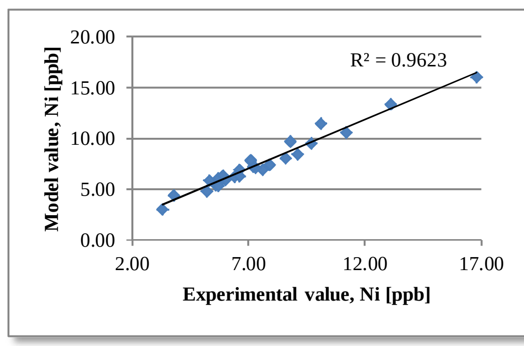


**Fig. 9. Validity check of the $Y_4$ (*nickel concentration*) mathematical model**

### *3.3. Mathematical modelling of metallic constituent concentrations migrated into 9% $CH_3COOH$ solutions*

Statistical summary of mathematical models to describe the found dependent variable Cr, Mn, $^{56}Fe$ and Ni is shown in Table 5.

**Table 5**

**Summary statistic of mathematical models proposed for the dependent variables**

| *Dependent variable* | *P* | *Model* | *Standard deviation [σ]* | *R-Squared [$R^2$]* | *Adjusted R-Squared [$R^2$ adjusted]* |
|---|---|---|---|---|---|
| $Y_1$ | 0.010 | Quintic | 1.06 | 0.9965 | 0.9771 |
| $Y_2$ | 0.020 | Quintic | 0.21 | 0.9981 | 0.9876 |
| $Y_3$ | 0.035 | Quintic | 41.82 | 0.9960 | 0.9730 |
| $Y_4$ | 0.040 | Cubic | 0.89 | 0.9823 | 0.9220 |

The mathematical models that define the studied response functions are presented in the following equations.

$$Y_1 = 8.67 + 0.33 \cdot X_1 + 2.17 \cdot X_2 + 0.17 \cdot X_3 - 6.00 \cdot X_1^2 - 2.50 \cdot X_2^2 - 0.50 \cdot X_3^2 + 2.00 \cdot X_1 \cdot X_2 + 0.25 \cdot X_1 \cdot X_3 + 1.50 \cdot X_2 \cdot X_3 + 1.00 \cdot X_1^2 \cdot X_2 + 1.25 \cdot X_1^2 \cdot X_3 + 4.00 \cdot X_1 \cdot X_2^2 + 0.75 \cdot X_1 \cdot X_3^2 + 3.00 \cdot X_2^2 \cdot X_3 + 6.00 \cdot X_2 \cdot X_3^2 + 3.00 \cdot X_1 \cdot X_2 \cdot X_3 + 6.00 \cdot X_1^2 \cdot X_2^2 + 1.25 \cdot X_1^2 \cdot X_2 \cdot X_3 + 0.75 \cdot X_1^2 \cdot X_3^2 + 3.50 \cdot X_1 \cdot X_2^2 \cdot X_3 + 2.00 \cdot X_1 \cdot X_2 \cdot X_3^2 + 1.50 \cdot X_2^2 \cdot X_3^2 \quad (9)$$

The validity of the $Y_1$ mathematical model proposed was plotted by *experimental values vs model values (Figure 10)*.

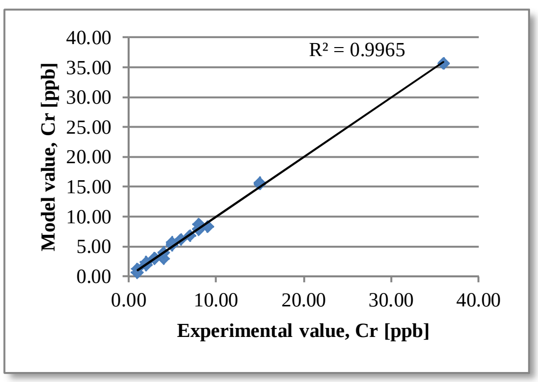


**Fig. 10. Validity check of the $Y_1$ (*chromium concentration*) mathematical model**

The mathematical model of the $Y_2$ (Mn concentration) response function is shown in Equation 10.

$$Y_2 = 1.46 - 0.00005 \cdot X_1 + 1.21 \cdot X_2 + 0.13 \cdot X_3 - 0.22 \cdot X_1^2 + 0.88 \cdot X_2^2 - 0.12 \cdot X_3^2 + 1.15 \cdot X_1 \cdot X_2 + 0.025 \cdot X_1 \cdot X_3 + 0.63 \cdot X_2 \cdot X_3 - 0.33 \cdot X_1^2 \cdot X_2 + 0.26 \cdot X_1^2 \cdot X_3 + 0.93 \cdot X_1 \cdot X_2^2 + 0.11 \cdot X_1 \cdot X_3^2 + 0.91 \cdot X_2^2 \cdot X_3 + 0.042 \cdot X_2 \cdot X_3^2 + 0.93 \cdot X_1 \cdot X_2 \cdot X_3 + 0.25 \cdot X_1^2 \cdot X_2^2 + 0.20 \cdot X_1^2 \cdot X_2 \cdot X_3 + 0.17 \cdot X_1^2 \cdot X_3^2 + 0.90 \cdot X_1 \cdot X_2^2 \cdot X_3 + 0.10 \cdot X_1 \cdot X_2 \cdot X_3^2 + 0.025 \cdot X_2^2 \cdot X_3^2 \quad (10)$$

The validity of the $Y_2$ mathematical model proposed was plotted by *experimental values vs model values (Figure 11)*.

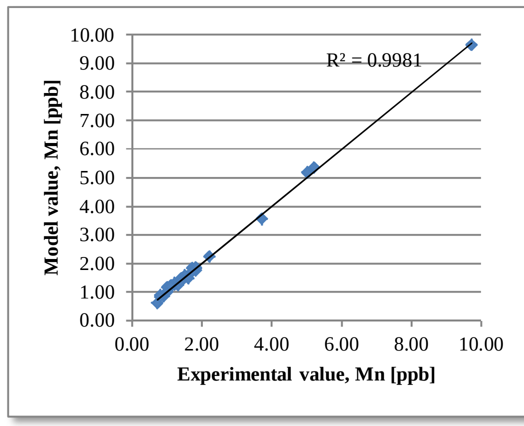


**Fig. 11. Validity check of the $Y_2$ (*manganese concentration*) mathematical model**

The mathematical model of the $Y_3$ ($^{56}Fe$ concentration) response function is shown in Equation 11.

$$Y_3 = 147.78 - 18.89 \cdot X_1 + 190.00 \cdot X_2 + 57.78 \cdot X_3 - 66.67 \cdot X_1^2 + 83.33 \cdot X_2^2 - 16.67 \cdot X_3^2 + 157.50 \cdot X_1 \cdot X_2 - 47.50 \cdot X_1 \cdot X_3 + 100.00 \cdot X_2 \cdot X_3 - 42.50 \cdot X_1^2 \cdot X_2 + 40.83 \cdot X_1^2 \cdot X_3 + 180.83 \cdot X_1 \cdot X_2^2 - 9.17 \cdot X_1 \cdot X_3^2 + 53.33 \cdot X_2^2 \cdot X_3 - 15.00 \cdot X_2 \cdot X_3^2 + 105.00 \cdot X_1 \cdot X_2 \cdot X_3 + 12.50 \cdot X_1^2 \cdot X_2^2 - 7.50 \cdot X_1^2 \cdot X_2 \cdot X_3 + 77.50 \cdot X_1^2 \cdot X_3^2 + 162.50 \cdot X_1 \cdot X_2^2 \cdot X_3 + 7.50 \cdot X_1 \cdot X_2 \cdot X_3^2 - 20.00 \cdot X_2^2 \cdot X_3^2 \quad (11)$$

The validity of the $Y_3$ mathematical model proposed was plotted by *experimental values vs model values (Figure 12)*.

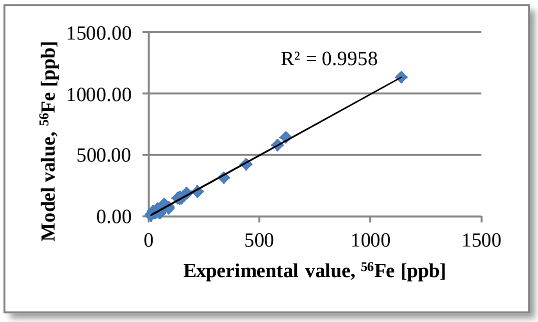


**Fig. 12. Validity check of the $Y_3$ (*iron concentration*) mathematical model**

The mathematical model of the $Y_4$ (Ni concentration) response function is shown in Equation 12.

$$Y_4 = 8.44 + 1.09 \cdot X_1 + 1.28 \cdot X_2 + 1.74 \cdot X_3 - 1.49 \cdot X_1^2 - 0.23 \cdot X_2^2 + 0.37 \cdot X_3^2 + 1.07 \cdot X_1 \cdot X_2 + 0.59 \cdot X_1 \cdot X_3 + 0.76 \cdot X_2 \cdot X_3 - 0.47 \cdot X_1^2 \cdot X_2 - 0.21 \cdot X_1^2 \cdot X_3 + 0.0083 \cdot X_1 \cdot X_2^2 + 0.91 \cdot X_1 \cdot X_3^2 + 0.24 \cdot X_2^2 \cdot X_3 + 0.72 \cdot X_2 \cdot X_3^2 + 1.18 \cdot X_1 \cdot X_2 \cdot X_3 \quad (12)$$

The validity of the $Y_4$ mathematical model proposed was been plotted by *experimental values vs model values (Figure 13)*.

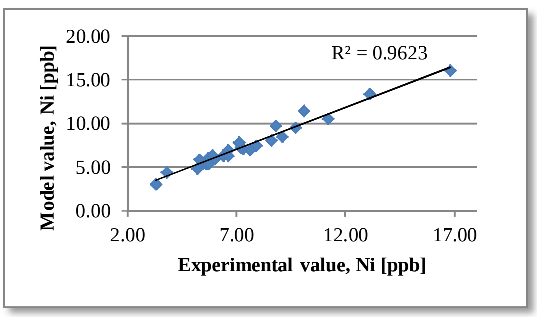


**Fig. 13. Validity check of the $Y_4$ (*nickel concentration*) mathematical model**

## 4. Conclusions

ICP-MS method and ANOVA method were used to establish the relationships between migration test parameter values and Cr, Mn, $^{56}$Fe and Ni concentrations found in solutions.

Besides the fact that these instrumental analysis techniques provide the checking of mathematical models' validity for each dependent variable, we can also draw the conclusion that they are highly performing and accurate instruments that can be used in estimating the values of objective function (see also [4]).

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