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Statistical Study and Modeling of the Effect of Phosphoric Acid Impurities on the Physical Quality of Ammonium Phosphate Determined from the Production Data Using Artificial Neural Network

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ABSTRACT

The monitoring of the physical properties of fertilizers presents a lot of interest at different levels. All the problems related to the behavior of the granules (segregation, spreading, granulation, hardness...) need a better characterization of the fertilizers in order to understand and/or predict them.

The scientific community is led to develop a variety of modeling approaches for, on the one hand, the understanding of the dynamics of impurities originating from phosphoric acid during the manufacture of fertilizers, and on the other hand, the evaluation of the effectiveness of measurements and data monitoring from production and the fluctuations in the physical quality of the fertilizers that follow.

In this investigation of the influence of elements such as Fe, Al, Mg, F-, Si, Na, K and Cl on fertilizer quality, we gathered and followed-up data from production lines of Ammonium phosphate fertilizers for several months.

The aim of the current work is the statistical study and modeling of the phosphoric acid impurities effect on the physical quality of fertilizers such as granulation yield, grain size, and the average diameter of granules D50, based on the analysis of data from different production lines by applying the Artificial Neural Network (ANN) approach. In addition models, were constructed using Multiple Linear Regression (MLR) and Principal Components Analysis (PCA).

Key Words: Ammonium phosphates fertilizers, Impurities, Granulation yield, Principal Components Analysis (PCA), Artificial Neural Network (ANN).

1. INTRODUCTION

Ammonium phosphate fertilizer is produced by simple reaction (neutralization) between ammonia and phosphoric acid resulting in the formation of the basic salt.

Several works were carried out to define and control the effect of impurities from phosphoric acid on the physicochemical properties of ammonium phosphates fertilizers (Michael Lloyd & FIPR, 2004; Dillard et al. 1982; Lapshin et al. 2016; Lebbar et al. 2016; BARTOS. et al. 1992; Campbell et al. 2006).

Mechanistic simulation models, based on a large number of production parameters and complex deterministic mathematical functions predicting the behavior of impurities in the phosphate-acid-fertilizer system are not yet fully mastered,

Considerable research efforts have been done by Valiulis & Simutis (2009); Siminovich & Joao (2014) and Zhang et al. (2017) in the field of modeling, optimization and control of the industrial granulation of fertilizers. Different sources of knowledge have been exploited, such as physical phenomena and statistical analysis of process parameters.

Hence the need for flexible and rapid decision support systems to be put in place facing the diversity of situations encountered on the field.

This necessity, leads us to develop statistical models based on a limited number of production variables.

The objective of the study presented herein is to improve the fertilizer granulation yield on the basis of the prediction of the adequate phosphoric acid profile based on various variables that may influence the particle size of the finished product.

A series of research questions were then asked:

- What is the relationship between the granulometric profile of the fertilizer and the profile of the phosphoric acid used as raw material?
- What are the predominant influencing factors in the evolution of granulation efficiency during fertilizer production?
- Would it be possible to envisage a decision support tool based on a statistical analysis of the production data for the estimation of the phosphoric acid profile allowing the improvement of the granulation yield of the fertilizers?

2. METHODOLOGY

2.1. STUDY ZONE

In order to have relatively similar fertilizers in their granulometric profiles for a first study covering a period from 2010 to 2015 allowing the collection of a considerable amount of data, we chose to focus on a single type of fertilizer, in our case the mono ammonium phosphate (MAP).

2.2. CREATION OF A DATABASE

The first step is the creation of a database of pairs of profiles of acids and corresponding MAP fertilizers, together with variables that may influence the yield of granulation during production: Phosphorus content (expressed as P_2O_5), solid content (TS), Calcium content (expressed as CaO), Magnesium content (expressed as MgO), Iron content (expressed as Fe_2O_3), Aluminum content (expressed as Al_2O_3), Sodium content (expressed as Na_2O), Fluorine content (F), sulfate content (expressed as SO_3) and chlorine content (Cl^-).

A series of specific variables are also developed to improve our forecasting objectives such as granulometric analysis of fertilizers: granulation yield (RdG), particle sizes $> 2mm$ (d2), $> 2.5mm$ (d25), $> 3.15mm$ (d31), $> 4 mm$ (d4).

For this purpose, the following databases were used:

- A 54% P_2O_5 phosphoric acid database comprising a quantitative description of the different chemical elements present from the weekly characterization of 89 acid profiles carried out between 2010 and 2015.
- A Mono Ammonium Phosphate Database (MAP std): comprising information on the weekly granulometric analysis of 89 fertilizer profiles carried out between 2010 and 2015 matching the same sampling periods of the 89 Studied phosphoric acid profiles.

The evaluation of these databases requires the cross-checking of the operating conditions of sampling, the latter taking place continuously during the manufacture. Their state will therefore be influenced by the respective sampling dates of the acid and fertilizer. But they will also depend on the duration between the sampling and the effective time of processing.

To integrate these temporal factors into a point-in-time variable we proceed by the weekly evaluation of characteristic functions of the involved processes.

2.3. ANALYSIS OF THE DATABASE

Based on the dataset, a statistical approach is applied to the selection of the most significant variables having an impact on the ammonium phosphate particle size distribution; this approach is based on the application of modeling tools such as Main components (PCA), multiple linear regression (MLR), (Baccini, 2010), and artificial neural network (ANN) (Dreyfus et al., 2002, Cornuéjols et al., 2002).

Knowing that our system is nonlinear, our model was constructed and developed using the JMP software by applying the approach of artificial neural networks.

3. RESULTS AND DISCUSSION

3.1. STATISTICAL APPROACH BY APPLYING PRINCIPAL COMPONENT ANALYSIS ACP

Principal Components Analysis (PCA) applies in all areas for the processing and analysis of quantitative data. This is the reference method that can be used to support other statistical techniques such as linear regression and artificial neural networks.

Malik et al. (2018) and Chaouki et al. (2015) are examples of the diversity of this approach application.

3.1.1. Inertia or percentage of information

The analysis of the results shows that most of the information is explained by the first two factorial axes. In the factor plane F1xF2, the eigenvalues of the two components F1 and F2 and their contribution to the total inertia are shown in Table 1.

Table1. Distribution of inertia between axes

Own values and Main Axes				
Axis	Own value s	% Inertia	% Cumulative	Histogram
1	4,7648	43,3144	43,3144	=====
2	2,7391	24,8998	68,2142	=====
3	1,2161	11,0546	79,2688	===
4	0,733	6,6629	85,9317	==
5	0,4114	3,74	89,6717	=
6	0,2975	2,7041	92,3758	
7	0,281	2,5543	94,9301	
8	0,2043	1,8574	96,7875	
9	0,1503	1,366	98,1535	
10	0,1136	1,0331	99,1866	
11	0,0895	0,8134	100	

The three axes taken into account for decoupling the correlations between variables related to spatial structures; alone account for 79.27% of the total information with 43.31% for axis 1, 24.89% Axis 2 and 11.05% for axis 3.

3.1.2. Analysis of Main and Additional Variables

Table 2 and 3 assembled hereafters allow to define the factors having a significant effect (high correlation: $r > 0.2084$ and close to 1) or non-significant (low correlation: $r \leq 0.2084$) on the granulation efficiency. Here, most of the information is explained by the first three factorial axes.

Table 2. Analysis of Main Variables

Var. Main and Additional.
Key Individuals
Significant correlations à 95,00 %
r = 0,2084 (89 - 2)

Variables	Axe 1- co s ²	Axe 2- co s ²	Axe 3- co s ²	Axe 4- co s ²	Axe 5- co s ²	Axe 6- co s ²	Axe 7- co s ²	Axe 8- co s ²	Axe 9- co s ²	Axe 10- - co s ²	Axe 11- - co s ²	Cumul co s ²
Pac	0,63	0,18	0	0,09	0	0	0	0,05	0,02	0,01	0,02	1
TS	0,77	0,04	0,01	0,06	0,01	0,01	0	0,01	0,08	0	0,01	1
CaO	0,79	0,05	0	0,02	0	0,06	0,01	0	0,03	0,02	0,01	1
Si	0,34	0,41	0,01	0,05	0,05	0,01	0,13	0,01	0	0	0	1
Fe	0	0,79	0	0,12	0,01	0	0,02	0	0	0,02	0,02	1
F	0,5	0,06	0,02	0,28	0,04	0,02	0,06	0,02	0	0	0	1
SO	0,67	0	0,19	0,01	0	0,01	0,03	0,07	0	0,03	0	1
Na	0,35	0,19	0,17	0,05	0,22	0	0	0,01	0	0	0	1
Mg	0,15	0,14	0,6	0,02	0,04	0,01	0,01	0,03	0	0	0,01	1
Al	0	0,64	0,2	0,04	0,03	0,04	0	0	0	0,01	0,01	1
Cl	0,57	0,23	0	0	0	0,13	0,03	0,01	0	0,02	0	1

Table 3. Analysis of Additional Variables

Variables	Axe 1- cos 2	Axe 2- cos 2	Axe 3- cos 2	Axe 4- cos 2	Axe 5- cos 2	Axe 6- cos 2	Axe 7- cos 2	Axe 8- cos 2	Axe 9- cos 2	Axe 10- cos 2	Axe 11- cos 2	Cumul cos 2
RdG	0,53	0,31	0	0	0,02	0	0,04	0	0,01	0	0,09	1
d4	0,21	0,08	0,57	0	0,09	0	0,03	0	0,01	0	0	1
d31	0,36	0,53	0,01	0,01	0,05	0	0	0,01	0,02	0	0	1
d25	0,47	0,36	0,03	0	0,05	0,01	0,01	0,01	0,04	0	0,03	1
d2	0,54	0,25	0,06	0	0,05	0	0,03	0	0,01	0	0,07	1

3.1.3. Correlation matrix

Table 4. Correlation matrix

Var. Princ. et Supp.																
Individus Principaux																
Corrélationssignificatives à 95,00 %																
r = 0,2084 (89 - 2)																
	Pac	TS_	CaO	Si_	Fe_	F_	SO_	Na_	Mg_	Al_	Cl_	RdG	d4_	d31	d25	d2_
Pac	1															
TS_	-0,8268	1														
CaO	-0,6442	0,7422	1													
Si_	-0,1278	0,3641	0,6137	1												
Fe (as Fe ₂ O ₃)_	0,3276	-0,1463	0,1821	0,4355	1											
F_	-0,3257	0,4712	0,588	0,6272	0,0601	1										
SO_	-0,6029	0,6413	0,7228	0,4225	0,0253	0,5009	1									
Na_	-0,2261	0,3132	0,5731	0,5418	0,2056	0,4752	0,6173	1								
Mg_	0,3898	-0,4474	-0,2858	-0,0395	0,3466	-0,3052	0,0113	0,1344	1							
Al_	0,2375	-0,0641	0,2033	0,4762	0,7619	0,1752	-0,0911	0,2089	-0,0371	1						
Cl_	-0,7802	0,7286	0,499	0,1334	-0,4477	0,3815	0,5886	0,2256	-0,4323	-0,2821	1					
RdG	0,544	-0,4729	-0,3062	0,0154	0,302	-0,2551	-0,3597	-0,1564	0,3759	0,2562	-0,4934	1				
d4_	0,2331	-0,2708	-0,1861	-0,0165	0,1247	-0,1939	-0,0344	0,0132	0,4984	-0,0936	-0,2547	0,3942	1			
d31	0,4933	-0,4335	-0,2011	0,1015	0,4855	-0,1737	-0,2882	-0,1475	0,3144	0,3741	-0,5209	0,6129	0,572	1		
d25	0,5633	-0,5353	-0,2891	0,0401	0,3938	-0,2768	-0,3171	-0,1448	0,4916	0,2538	-0,578	0,868	0,6096	0,8407	1	
d2_	0,54	-0,5005	-0,3421	-0,0199	0,2781	-0,2724	-0,3141	-0,1477	0,4862	0,1679	-0,4984	0,9464	0,6119	0,7431	0,9361	1

Examination of the correlation matrix between variables on the table 4 reveals the presence of a first set of variables, consisting of well correlated variables. which is:

- TS with P2O5 (-)¹;
- CaO with P2O5 (-), and TS (+)²;
- SiO2 with CaO (+);
- Fe2O3 with SiO2 (+): slightly significant correlation;
- F with TS, CaO and SiO2 (+);
- SO3 with P2O5 (-) and TS, CaO (+), slightly significant with F and SiO2 (+);

¹(-) : Low correlation

²(+) : Strong correlation

- Na₂O with CaO, F, SiO₂ and SO₃ (+);
- MgO with TS (-): slightly significant correlation
- Al₂O₃ with Fe₂O₃ and slightly significant with SiO₂ (+)
- Cl with P₂O₅ (-), SO₃ and TS (+), and slightly significant with CaO (+), Fe₂O₃ (-), and MgO
- RdG with P₂O₅ (+), TS and Cl (-),
- d₄ with MgO (+)
- d₃₁ with P₂O₅ (+), TS (-), Fe₂O₃ (+) and Cl (-)
- d₂₅ with P₂O₅ (+), TS (-), MgO (+) and Cl (-)
- d₂ with P₂O₅ (+), TS (-), MgO (+) and Cl (-)

In conclusion, the major elements influencing the particle size of ammonium mono phosphate are: P₂O₅ (+), TS (-), Fe₂O₃ (+), MgO (+) and Cl (-).

3.1.4. Correlation circle

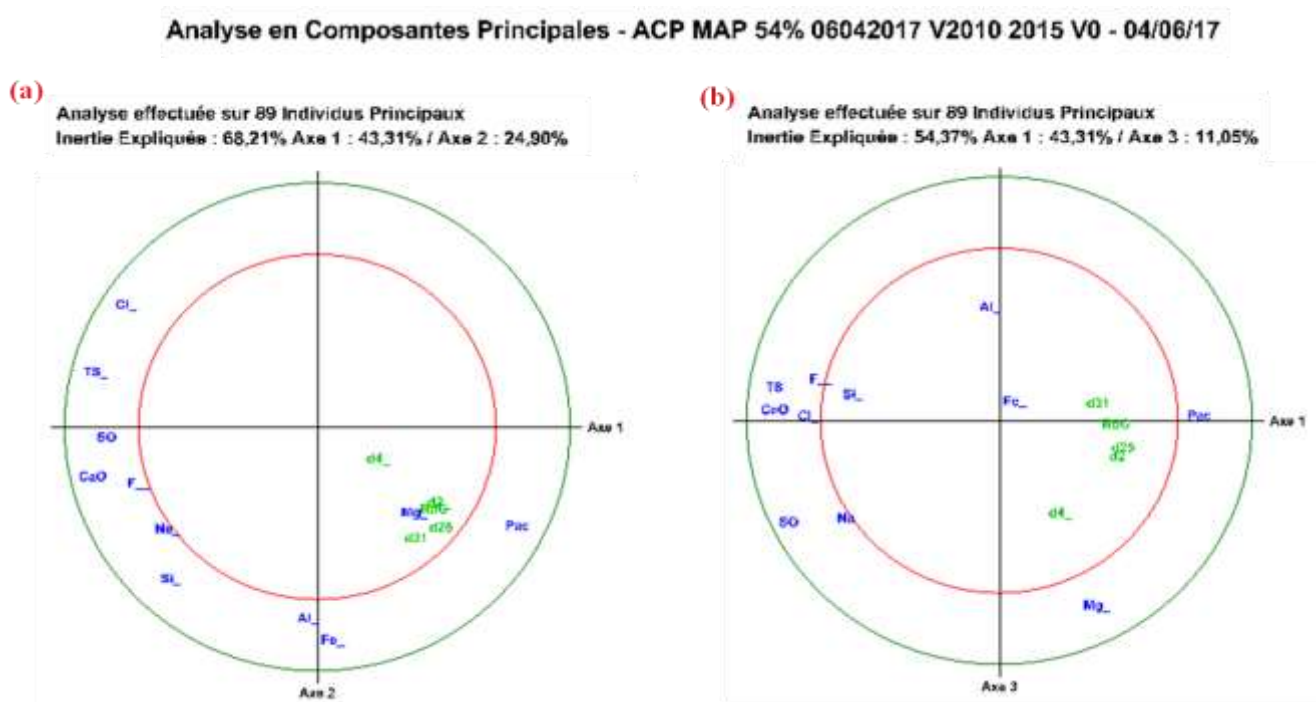


Fig.1. (a) Circle of correlations along axes 1,2 ; (b) Circle of correlations along axes 1,3

Examination of the correlation circles on the figure 1 shows that:

- The first component (F1), contributing by 43.31% inertia, is defined by the most significant factors: P₂O₅ (0.79) on the positive side, TS (0.88), CaO (0.89), F, SO₃ (0.82), and Cl⁻ (0.76) on the negative side
- With a 24.90% inertia, the second component (F2) is defined by the parameters: Si (0.64), Fe₂O₃ (0.89) and Al₂O₃ (0.80) on the positive side.
- With an inertia of 11.05% the 3rd component (F3) is defined by the parameter MgO (0.78) on the positive side.

3.2. STATISTICAL APPROACH BY APPLYING MULTIPLE LINEAR REGRESSION (MLR)

By applying multiple regressions to our system, we found that our system is nonlinear, and since the main composting analysis approach is reserved for linear systems,

We have opted for the application of suitable alternative statistical approach for non-linear systems called "Artificial Neural Networks"

3.3. *TATISTICAL APPROACH BY THE APPLICATION OF ARTIFICIAL NEURAL NETWORKS ANN*

Neural networks are tools for modeling nonlinear phenomena. Their development derives from the initial idea of imitating the structure and behavior of the human brain (Minsky & Papert 1969). In order to calibrate the ANN parameters, it is necessary to perform an Apprenticeship on a database of the phenomenon. This step is matched to the calculation of a non-linear regression. Apprenticeship is the process of adjusting the parameters of the different layers of the neural network so that the error on the output is as low as possible (Cornuéjols et al., 2002).

3.3.1. JMP processing software

The statistical analysis method used and included in the data processing by the JMP software is that of the least squares. This method makes it possible to compare experimental data, often error-free, with a mathematical model established to explain these data. In other words, it is to look for a relationship between the response “y” (granulation efficiency), dependent variable, and the factors “x” (P2O5, solid content (TS), CaO, MgO, Fe2O3, Al2O3, F, Na2O, SO3 and Cl-), explanatory variables, taking into account the errors observed in the mathematical model (given prediction). This statistical method thus provides several information about the established model (Dreyfus et al., 2002):

a) *Determination of the model structure*

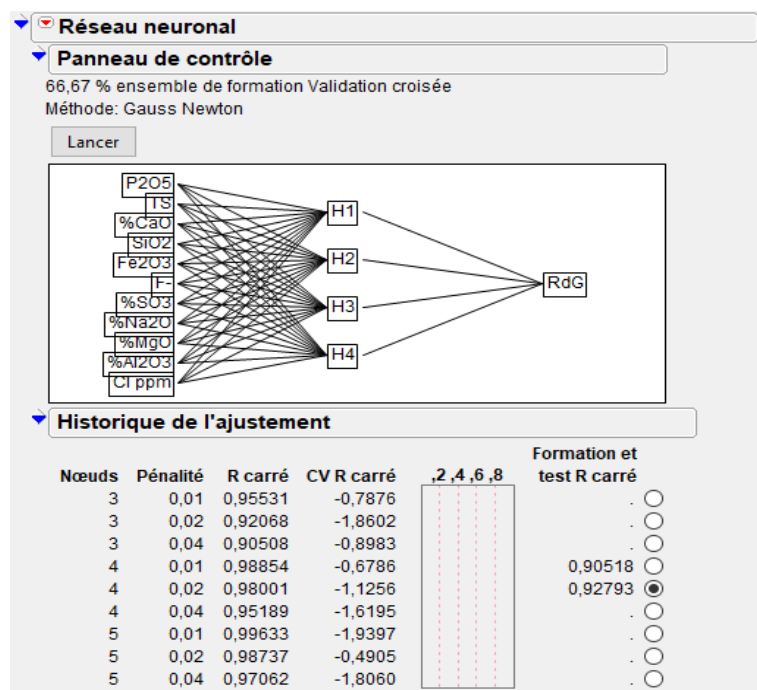


Fig.2. Structure of the artificial neural network (ANN)

We try to improve the quality of our predictions by adjusting artificial neural networks. The flexibility of this method of calculation will make it possible to demonstrate more complex nonlinear relations between our variables. We will pay particular attention to the development of architecture as simple as possible. We restrict ourselves to the training of networks with a single layer hidden, containing a minimum of nodes (Bishop 1995).

The structure of the model is determined as follows:

- **Entries /exits:** The input variables are chosen on the basis of the chemical analyzes results carried out on the 89 phosphoric acid profiles 54% such as P2O5, solid content (TS), CaO, MgO, Fe2O3, Al2O3, F, Na2O, SiO2, SO3 And Cl-.

The network will therefore include 11 inputs and 1 output node indicated by the RDG granulation yield (Fig.2).

Hidden layer: 5 networks whose architecture consists of 1 hidden layer whose number of nodes goes from 3 to 5 are adjusted independently. Networks whose R2-adjusted coefficients are maximum for a minimum number of nodes are

then selected (Fig.3). The network at 4 nodes is finally retained. An average coefficient of determination of 0.9279 is assigned following a total sequence of 9 adjustments.

b) Ajustement de Réseaux de Neurones Artificielles (RNA)

Apprenticeship is a phase of the development of a neural network during which the network behavior is modified until the desired behavior is obtained (granulation efficiency of 96%). Indeed, the connections between the neurons of 11 input variables cited in (a) have numerical weights; these weights reflect the strength, the importance of the related entries. The output of each neuron is a function of the weighted sum of its inputs. By considering that Apprenticeship is formulated as an optimal search in the weight space, we have adjusted the weights of our neuronal network iteratively (100 iterations) until the outputs are in agreement with the inputs, And in such a way as to minimize the sum of the squares of the deviations between the observed outputs and the desired outputs (fig. 3).

Furthermore, the graphs of figure 3 allow us to see if the prediction model conforms to the values obtained experimentally. If the model is perfect, a straight line is obtained with R^2 equal to 1.

3.3.2. Optimization study

3.3.3. For optimization, we used the optimization study tools that JMP processing software offers. However, in order to search for compromise zones between the factors that generate the adjustments leading to the desired response, we use the curves of isoresponses. The purpose of this tool is to obtain surface curves that reflect the variations of our response. After the determination of the model and the verification of its validity, the curves of isoresponses were plotted within the experimental domain. These curves represent planes for response surfaces, that is to say the graphical representation of the results (estimated model) in order to draw optimums.

Then, to find the exact optimal setting with a certain percentage of compromise, we use the "Desirability" function. This function makes it possible to give an exact optimal setting, varying between 0 and 1. Indeed, the value 0 is assigned when the factors lead to an unacceptable (undesirable) response and 1 when the response represents the desired maximum performance for the considered factors.

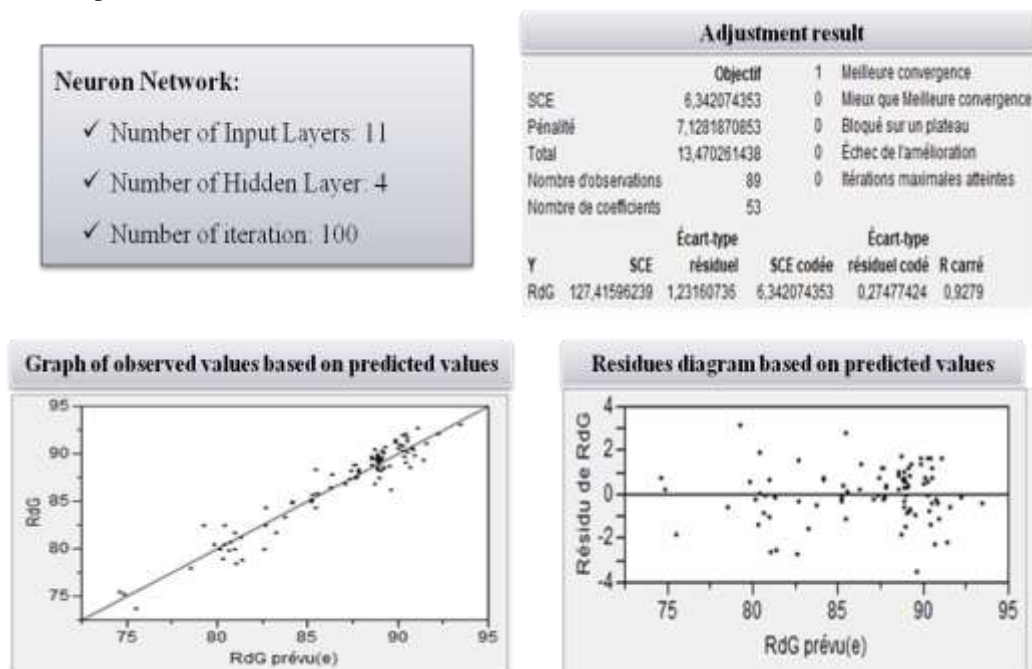


Fig.3. Adjustment results of neural network model

Several research works found in literature

applied this statistical approach in different studies related to chemistry, biology and environment which proves its performance and relevance in different fields. Among these studies, those carried out by Fadil et al. (2013), Petit et al. (2013), Gargouri1 et al. (2012), Ming et al. (2017), Sadoudi (2012) and Lauret (2016).

a) Profile of isoreponses

The step consists in fixing the two values for the two parameters P2O5 and SiO2 and proceeds by searching for the values of the other factors.

The white zone corresponds to the compromise zone for these two parameters in order to achieve the desired yield and fixing the contents of P2O5 and SiO2 at the values 50.02% and 5.15 respectively (fig.4). Fixing these two parameters allowed us to know the domains of variation of the other factors making it possible to achieve the desired yield.

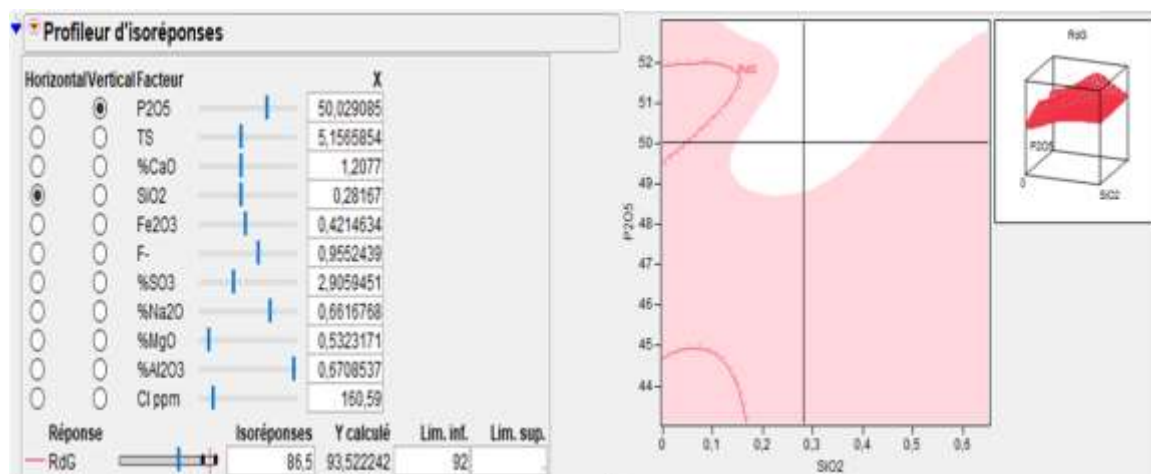


Fig.4. Example of isoreponses profiler of neural network model

b) Desirability study

The aim is to maximize the granulation yield so that it reaches a value of 96%. Reaching a 96% granulation yield is possible with a desirability of about 98.42% using the acid profile mentioned below as operating conditions (fig.5).

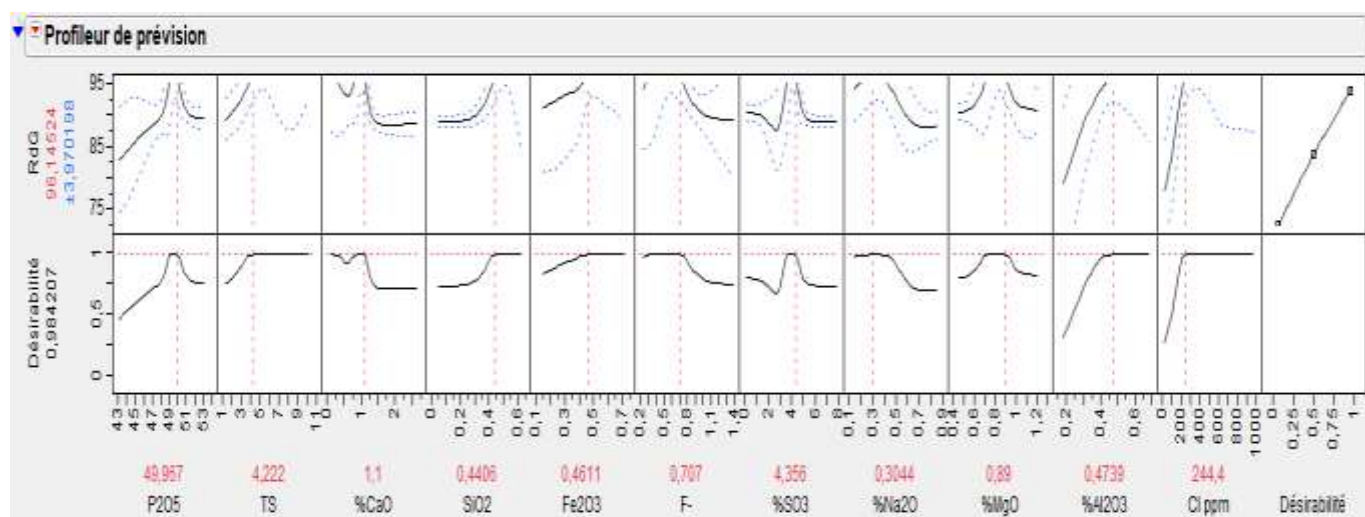


Fig.5. Prevision profiler of neural network model

4. CONCLUSION

The linear model could not be validated statistically; modeling neural networks for the nonlinear system gave us a model that will allow us to predict optimum conditions in the future and predict the best 54% phosphoric acid profile allowing a good granulation of mono ammonium phosphate fertilizers.

The coefficient of determination $R^2 = 92.67\%$ is significant. This value gives good compatibility between the experimental and predicted values of the adapted model.

This study allowed us to represent and develop a specification of the model to be deployed while respecting the following key points that are directly related to the manufacturing process of fertilizers on an industrial scale.

- Mastery of database sampling
- Predicting obstacles related to production performance parameters.
- Predicting the variation and diversity of the frequentative acid profile.
- Performing the most accurate forecasts possible.
- Adapting the model for each type of fertilizer.

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