Modelization and Optimization by Experimental Design Method of the Carbonation of Raw Sugar Juice

R. Lotfi[1], R. Diany[2], D. Zakarya[3]

[1]Faculty of Science and Technology of Mohammedia-Morocco

[1]Correspondence: Laboratory of Materials, Catalysis and Valorization of Natural Resources Faculty of Science and Technology - BP 146 Mohammedia; 20650- Tel.: 212523314705- Fax: 212523315353 - E-mail: *lotfirabiaa[at]yahoo.fr*

Abstract: ***Carbonation by lime milk and carbon dioxide, used in the refining of raw sugar, was studied using a plan of experiments. This study allowed us to establish a mathematical model that describes the influence of the variables: carbon dioxide richness, alkalinity level, acidity level, Brix rate, sugar juice heating temperature and liming level and their interactions on the response: loss of loads. The use of this model in the space of variables allowed to define the optimal economic conditions (X1 = 3g/l, X2 = 0.07g/l, X3 = 44.73 p.p.m, X 4 = 64 Brix, X5 = 4.7g/l, X6 = 12 kg/h, and X7 = 87°C for obtaining clear refined sugar and with a pressure difference (P) maintained at 0.5 bars.***

Keywords: Sugar juice, modelization, optimization, carbonation, experimental design

1. Introduction

The refining of raw sugar is a process that produces refined sugar from raw sugar (FAUCONNIER and BASSEREAU D, 1970). The latter consists of sucrose crystals coated with a syrup film containing organic (CLARKE, M. A. 1996) and mineral impurities. This process requires several operations such as refining and scrubbing to remove external and internal impurities to the crystals through the carbonation procedure, which consists of introducing lime milk with sweet juice and carbon dioxide to form a calcium carbonate precipitate capable of securing the majority of both organic and mineral impurities. Then discoloration by ion exchange resins (THEOLEYRE et al. 1999).

In order to control the impact of carbonation of sugar juice, it is necessary to study the variation of the parameters involved and influencing this treatment, such as the carbon dioxide content, the level of liming, the level of alkalinity and acidity (HCl used for boiler washing), the starch content (from the sugar plant), the high starch levels pose filtration problems (ANYANGWA et al., 1993), Brix (dry matter content in 100g of sweet water), liming level (amount of lime milk introduced) and heating temperature of sugar syrup. In this work, on the one hand, we present the procedure that describes the carbonation of sugar juice, on the other hand, we will present the statistical analysis of the results obtained from an experimental data table converted into a plan of experiments using the Statgraphics. The results of this analysis enabled us to establish a mathematical model and to determine the optimal experimental conditions.

2. Materials and Methods

2.1 Material

The refining process used consists of the following equipment: UV-visible single-beam spectrophotometer from 320 to 1000 nm Jenaway N1972, DMA48 densimeter, Mettler PM 2000 balance of 1% accuracy for weighing sugar, thermometer, pH-metre RS232C refractometer of the following characteristics, to measure the color at 0.04%: halogen light source, synthetic sapphire prism, stainless steel sample chamber.

3. Experimental Method

Carbonation is the process of precipitation of calcium carbonate, this precipitate allows the removal of impurities. Lime milk is fed into the sugar juice solution and carbon dioxide is pumped into the water channel. top of the boiler, which combines with water to form carbonic acid. It reacts immediately and neutralizes the dissolved lime in the syrup at a temperature between 65°C and 75°C and a pH between 8.5 and 10.

$CO2 + H2O \rightarrow H2CO3$
$CaO + H2O \rightarrow Ca(OH)2$
$Ca\ (OH2)2 + H2CO3 \rightarrow 2H2O + CaCO3$

After carbonation, filtration is carried out to further remove ash and coloring matter. The study of the experiment plan and the statistical calculations were carried out using the Statgraphics software.

4. Design Experimental and Statistical Analysis

The experimental design is frequently used in the field of agriculture, agri-food, biology and chemistry (BOX et al. 1978). One of its objectives is to establish a mathematical model between the measured response and a number of variables that influence. In this work, we will present the raw sugar refining process and we will present an experimental design that allows us to establish an analytical expression linking the response: loss of loads by measuring the difference in pressure P (between the input pressure at the first boiler set at 3.8 bar and the average of the pressures measured after the five boilers pass through) to the influential factors: the level acidity level (X_1), alkalinity level (X_2), starch content (X_3), Brix (X_4), liming level (X_5), carbon dioxide content (X_6), and sugar syrup heating temperature (X_7), taking into account their interactions (Box and DRAPPER, 1987), and statistical analysis (GOUPPY, 1992), of these results obtained from this plan based on unplanned industrial data, which we collected in a table and converted by the software Statgraphics into a design of experiments. Table 1 presents data from the 38 industrial experiments.

Table 1: Experimental data collected and analytical results

Expérience	X_1	X_2	X_3	X_4	X_5	X_6	X_7	Y_{exp}	Y_{cal}
1	2,4	0,03	5,26	63	4,5	13	92	1,28	1,30
2	2,4	0,02	2,63	64	4,1	13	84	1,3	1,26
3	2,4	0,05	71,05	63	3,8	13	86	1,4	1,42
4	2,4	0,06	78,94	62	4,8	13	86	1,08	1,02
5	2,4	0,04	26,31	64	4,1	12,4	85	1,45	1,38
6	2,4	0,02	15,78	64	4,3	12,4	85	1,44	1,60
7	2,4	0,06	5,26	65	4,8	12,4	85	1,44	1,43
8	2,4	0,05	57,89	64	4,5	13	87	1,94	2,00
9	2,4	0,06	21,05	64	4,5	13	87	1,62	1,62
10	2,4	0,07	60,52	63	5,1	13	87	1,42	1,46
11	2,4	0,04	13,15	61	4,2	13	85	1,68	1,68
12	2,4	0,05	15,78	62	4,9	13	86	1,4	1,43
13	2,4	0,04	107,9	64	4,3	11,6	85	1,2	1,21
14	2,4	0,05	65,78	63	4,5	12,9	87	2,66	2,65
15	2,4	0,08	113,15	62	4,4	12,9	87	2,5	2,50
16	2,4	0,07	255,26	63	4,6	12,6	88	2,16	2,02
17	3	0,04	26,31	65	4	12,2	87	0,52	0,83
18	3	0,03	18,42	63	3,4	12,2	87	0,84	0,85
19	3	0,03	21,05	63	3,7	12,2	85	0,86	0,62
20	3	0,03	10,52	61	4,2	12	88	0,68	0,68
21	3	0,04	44,73	64	4,1	11,2	87	0,72	0,71
22	3	0,05	57,89	64	4,3	11,2	88	0,67	0,69
23	3	0,04	39,47	64	4,8	11,2	85	0,68	0,61
24	3	0,03	110,52	63	4,3	12,4	87	0,52	0,88
25	3	0,05	13,15	64	4,1	12,4	87	0,6	0,77
26	3	0,04	21,05	64	4,1	12,4	87	0,88	1,10
27	3	0,04	10,52	63	3,8	11,6	87	0,82	1,06
28	3	0,06	26,31	64	3,9	11,6	86	0,97	0,87
29	3	0,05	10,52	65	4,7	11,6	84	1,1	1,58
30	3	0,06	26,31	63	4,5	10,8	86	0,84	0,84
31	3	0,05	36,84	64	4,1	10,8	85	1,64	0,86
32	3	0,05	26,31	64	4,1	12	87	0,8	0,97
33	2	0,03	39,47	64	4	12,2	86	0,9	0,88
34	2	0,02	73,68	66	3,8	12,2	87	0,86	0,81
35	2	0,03	15,78	65	3,5	12,2	91	0,82	0,85
36	2	0,03	18,42	65	4,1	12,2	87	0,84	0,82
37	2	0,05	65,78	65	3,9	12,2	83	0,82	1,20
38	2	0,05	31,58	65	4,2	12,2	87	1,2	1,57

This table displays information about the measured pressure difference values and the estimated values using the adjusted model.

5. Results

All operating conditions, and the results obtained (pressure difference) of refined sugars, are grouped in Table 1. This model consists of 36 terms: 1 constant, 7 linear, 7 square and 21 rectangular terms known as interaction terms. The terms of this model are easily calculated by the least squares method (Table 2)

The model equation (equation 1) is written as follows:

$\Delta P_{\mathbf{cal}} = 1{,}74 - 15X_1 - 2{,}15\ X_2 - 0{,}77X_3 - 4{,}5\ X_4 + 14{,}6\ X_5 - 50X_6 + 2{,}4X_7$ (équation 1)
$- 2{,}4\ X_1^2 - 63\ X_2^2 - 52.10^{-5}\ X_3^2 - 0{,}03\ X_4^2 + 0{,}3\ X_5^2 - 0{,}1\ X_6^2 - 4.10^{-4}\ X_7^2$
$+ 26{,}7\ X_1X_2 + 0{,}015\ X_1X_3 + 0{,}56\ X_1X_4 - 0{,}01\ X_1X_5 - 0{,}28X_1X_6 - 0{,}08\ X_1X_7$
$- 0{,}4\ X_2X_3 + 9{,}1\ X_2X_4 - 3{,}7\ X_2X_5 + 7{,}7\ X_2X_6 +{}_5 + 0{,}77\ X_2X_7 - 10^{-4}\ X_3X_4$
$+ 0{,}01\ X_3X_5 + 0{,}01\ X_3X_6 + 0{,}01\ X_3X_7 - 0{,}02\ X_4X_5 + 0{,}2X_4X_6 + 0{,}004\ X_4X_7$
$- 0{,}05\ X_5X_6 - 0{,}14\ X_5X_7 + 0{,}3\ X_6X_7$

With:
X_1: acid, X_2: alkalinity, X_3: starch, X_4: Brix: X5: liming, X_6: CO_2
and X_7: temperature

Based on this equation, the estimated values of ΔP ($\Delta Y_{cal} = \Delta P_{\mathbf{cal}}$) and corresponding residues $e_i = P_{exp} - P_{cal}$ (Table 1)

The estimate of the variance of the experimental error (s_r^2) is obtained by dividing the sum of the squares $\Sigma\ e_i^2$ of the residue by the number of degrees of freedom ν (number of degrees of freedom = number of experiments - number of model coefficients) (BENOIST et al., 1994

$$sr^2 = 0.0164351 \text{ (table 2)}$$

The significance of the effects is estimated Table (3) by comparing the value of Snedecor experimentally estimated (F_{exp}) to the value of Snedecor critical ($F_{0\cdot1}(1, 36) = 2.8503$) (BOX AND DRAPPER, 1987) to ν 1 = 1 and ν= 36 degrees of freedom, for a probability of 90%.

The experimental Snedecor factor is obtained by dividing the mean square (CMu) by the variance of the experimental error (sr2) Table 4:

$$Fexp = CMu/sr^2$$

The estimate of the individual mean square (CMu) is obtained by dividing the sum of the squares of each coefficient (SS) by its degree of freedom ($\nu_u = 1$):

$$CM_u = SSu / \nu_u$$

The estimate of the sum of the squares of the coefficients (SSu) is obtained by multiplying the square of the coefficient (bu) by the sum of the squares of the values of X_u:

$$SSu = b_u^2 \sum X_{iu}^2$$

With: b_u = polynomial model coefficient.
e_i = residue of the previous experiment: ei = P(exp) - P (cal).
sr^2 = variance of residue: $sr^2 = \sum e_i^2/\nu$
CM_u = average square of the b_u coefficient.
SS_u = sum of the squares of the b_u coefficient.

ν = degree of freedom = number of experiments - number of model coefficients.

Table 2: Estimation of model factors associated with loss of loads

Parameter	*Estimation*	*Erreur-type*
b_o	1,74791	0,937235
b_1	-15,2536	6,10929
b_2	-19,8272	5,18108
b_3	-2,14739	1,68624
b_4	5,86838	4,57342
b_5	1,70685	1,71557
b_6	-12,03	5,1095
b_7	4,03919	1,62014
b_1b_1	-1,21637	0,624635
b_1b_2	0,800955	0,324672
b_1b_3	1,93674	1,18782
b_1b_4	17,8257	6,33997
b_1b_5	-0,010766	0,340726
b_1b_6	-0,302465	0,982746
b_1b_7	-0,375425	0,905835
b_2b_2	-0,4072	0,498059
b_2b_3	-3,44837	1,18465
b_2b_4	17,4157	5,75299
b_2b_5	-0,189536	0,635429
b_2b_6	0,508873	0,400765
b_2b_7	0,209089	0,873581
b_3b_3	-1,68129	0,504665
b_3b_4	-1,05371	1,16361
b_3b_5	1,85786	0,980237
b_3b_6	2,61617	1,12111
b_3b_7	8,02126	2,14797
b_4b_4	-9,04954	4,41381
b_4b_5	-0,852893	1,61748
b_4b_6	16,1754	5,60673
b_4b_7	1,33887	0,789123
b_5b_5	-0,161721	0,38485
b_5b_6	-0,092128	0,520513
b_5b_7	-1,08992	0,897874
b_6b_6	0,883242	0,59315
b_6b_7	3,10976	1,05639
b_7b_7	-1,39024	0,542006

Table 3: Signification of variables in relation to experimental dispersion

Source of variation	*Sum of squares*	*ν*	*Mean square*	*F_{exp}*	*P-Value*	*Signification*
b_1	0,102456	1	0,102456	6,23	0,0412	**
b_2	0,240688	1	0,240688	14,64	0,0065	***
b_3	0,0266536	1	0,0266536	1,62	0,2435	NS
b_4	0,02706	1	0,02706	1,65	0,2403	NS
b_5	0,0162685	1	0,0162685	0,99	0,3529	NS
b_6	0,0911062	1	0,0911062	5,54	0,0508	NS
b_7	0,102154	1	0,102154	6,22	0,0414	**
b_1b_1	0,0623238	1	0,0623238	3,79	0,0925	*
b_1b_2	0,100023	1	0,100023	6,09	0,0430	**
b_1b_3	0,0436934	1	0,0436934	2,66	0,1470	NS
b_1b_4	0,129925	1	0,129925	7,91	0,0261	***
b_1b_5	0,0000164	1	0,00001640	0,00	0,9757	NS
b_1b_6	0,0015568	1	0,00155682	0,09	0,7672	NS
b_1b_7	0,0028230	1	0,00282308	0,17	0,6909	NS
b_2b_2	0,0008621	1	0,00086212	0,05	0,8254	NS
b_2b_3	0,139259	1	0,139259	8,47	0,0226	***
b_2b_4	0,150614	1	0,150614	9,16	0,0192	***
b_2b_5	0,0014622	1	0,00146226	0,09	0,7741	NS
b_2b_6	0,026498	1	0,026498	1,61	0,2448	NS
b_2b_7	0,0009415	1	0,00094151	0,06	0,8177	NS
b_3b_3	0,182412	1	0,182412	11,10	0,0126	***
b_3b_4	0,0134771	1	0,0134771	0,82	0,3953	NS
b_3b_5	0,059039	1	0,059039	3,59	0,0999	*
b_3b_6	0,0894971	1	0,0894971	5,45	0,0523	**
b_3b_7	0,229194	1	0,229194	13,95	0,0073	***
b_4b_4	0,0690875	1	0,0690875	4,20	0,0795	**
b_4b_5	0,0045696	1	0,00456969	0,28	0,6143	NS
b_4b_6	0,136793	1	0,136793	8,32	0,0235	***
b_4b_7	0,0473106	1	0,0473106	2,88	0,1336	*
b_5b_5	0,0029021	1	0,00290218	0,18	0,6869	NS
b_5b_6	0,0005148	1	0,00051486	0,03	0,8645	NS
b_5b_7	0,0242177	1	0,0242177	1,47	0,2642	NS
b_6b_6	0,0364421	1	0,0364421	2,22	0,1801	NS
b_6b_7	0,142422	1	0,142422	8,67	0,0216	***
b_7b_7	0,108129	1	0,108129	6,58	0,0373	**
Residue (total)	0,115046	7	0,0164351			
Total (corr.)	11,4282	42				

***: significant at 1% ($F_{0.01}$(1.36) = 7.39); **: significant at 5% (F0.05(1.36) = 4.11); *: significant at 10% (F0.01(1.36) = 2.85), NS: not significant.

Table 4 of the analysis of variance breaks down the variability of the pressure difference into separate rows for each effect. He then tests the statistical significance of each effect by comparing the quadratic mean with an estimate of the experimental error. In this case, 12 effects are significant at the 90.0% confidence level.

It appears that only X_1 acidity, X_2 alkalinity, X_7 temperature, interactions, X_1X_2, X_1X_4, X_2X_3, X_2X_4, X_3X_7, X_6X_7, X_3X_7 and X_3X_3,, X_7X_7 square terms are significant.

Therefore, for a 90% threshold of significance, the equation of the mathematical model is written:

P_{cal} = 1.74 - 15 X1 - 19.82 X2 + 4.04 X7 - 1.68 X32 - 1.39 X72
(±6.11) (±5.18) (±5.11) (±0.50) (±0.50)
+ 0.81 X1X2 +17.82 X1X4 - 3.45 X2X3+ 17.42 X2X4 (équation 2)
(±0.32) (±6.34) (±1.18) (±5.75)
+ 8.02 X3X7 + 16.18 X4X6 + 3.11 X6X7
(±2.15) (±5.61) (±1.06)

The values in parentheses and below each coefficient of the model represent the errors calculated by Statgraphics software.

6. Discussion

The study of equation (2) shows that under the optimal conditions presented in table (5), the calculated pressure difference is 0.5132, whereas the pressure difference obtained experimentally at this point is in the order of 0.52, thus enabling the mathematical model to be validated.

The geometric representation (GOUPY, 1999) of the response P in the space of variables X_4 (Brix) and X_3 (starch content) is shown on [Fig.1]. By decreasing the starch content and decreasing the Brix, the pressure difference decreases to the target value (0.5 bars).

Table 4: Optimal conditions for maintaining ΔP at 0,5 bars

Facteur	*Bas*	*Haut*	*Optimum*
Acidité (g/l)	3,0	3,0	3,0
Alcalinité (g/l)	0,07	0,08	0,07
Amidon (ppm)	18,42	255	44,73
Brix	64,0	64,0	64,0
Chaulage (g/l)	4,7	4,7	4,7
Gaz carbonique (kg/h)	12,0	12,0	12,0
Température (°C)	87,0	87,0	87,0

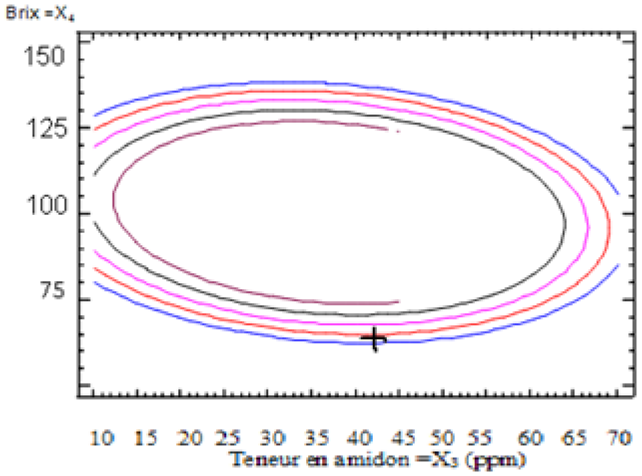

Figure 1: Pressure difference iso-response curves [equation 2]

$X_1 = 3$, $X_2 = 0.07$, $X_5 = 4.7$, $X_6 = 12$, and $X_7 = 87$.

- P = 0.0; 0.8; 1.6; 2.4; 3.2 (from outside to inside)

7. Conclusion

The application of the experimental design, based on the actual experimental data, allowed us to establish a mathematical model representative of the impact of the carbonation stage on the refining of raw sugar. The representation of this model allowed us to predict the optimal conditions for obtaining refined sugar with a pressure difference of less than 0.5 bar, thus presenting an economic interest to the raw sugar refining industry from different sources.

References

[1] ANYANGWA, E. M., KAPSEU, C., ELEMVA, M.et MUSONGE, P. 1993. The effect and removal of starch in the sugar refining industry. International Sugar Journal, 95 (1134), 210-213.
[2] BENOIST D., TOURBIER S., TOURBIER S.G., 1994. Plans
[3] d'expériences: Construction et analyses. Techniques et documentation, Lavoisier, Paris.
[4] BOX G.E.P., Hunter W.G., Hunter J.S., 1978. Statistics for experimenters , An introduction to Design, Data Analysis and Model Building, Wileys, New-York.
[5] R.FAUCONNIER R., BASSEREAU D., 1970. La canne à sucre. MAISONNEUVE G.P, et LAROSE. Paris
[6] BOX G. E. P. DRAPPER, N.R., 1987. Empirical Model-Building and response surfaces, Wileys, New York.
[7] CLARKE, M. A., 1996. Sugarbeet and sugarcane polysaccharides. Sugar processing research, 368-388. New Orleans, Louisiana, Etats-Unis
[8] FAUCONNIER R., BASSEREAU D., 1970. La canne à sucre. MAISONNEUVE G.P, et LAROSE. Paris
[9] GOUPY J., 1999. Plans d'expériences : Pour surfaces de réponses. Dunod, Paris.
[10] THEOLEYRE M.A., CARTIER S., DECLOUX K., 1999. Couplage de la décoloration et de la nanofiltration des éluats de regénération en raffinerie de canne. AVH Association -6th Symposium-Reims