Available online at www.sciencedirect.com

ScienceDirect

Journal of the European Ceramic Society 30 (2010) 2451–2455

www.elsevier.com/locate/jeurceramsoc

Development of coloured glazes for tile applications using Taguchi's method

A.O. Castela [a,*], A.T. Fonseca [b], P.Q. Mantas [c]

[a] *Endeka Ceramics, 3780-290 Anadia, Portugal*

[b] *Dep. Ceram. Glass Eng., University of Aveiro, 3810-193 Aveiro, Portugal*

[c] *Dep. Ceram. Glass Eng./CICECO, University of Aveiro, 3810-193 Aveiro, Portugal*

Received 26 November 2009; received in revised form 4 May 2010; accepted 17 May 2010
Available online 12 June 2010

Abstract

The development of coloured glazes produced for ceramic tiles is normally based on intuitive methods, where a lot of trials are usually necessary to obtain the desired colour.

The aim of this paper was to propose a new approach to develop coloured glazes produced for tile applications using Taguchi's method. With this method, the best producing conditions can be achieved by an appropriate selection of the controlled factors – the pigments – and their levels – the quantities of the pigments – to obtain the desired colour and production costs were thus reduced.

Two standard colours were selected under the conditions of the customer. The experience was planned and the results were treated according to the requirements described by Taguchi's method. The trials were selected by an orthogonal array of smaller resolution. Using Taguchi's method, the number of trials was substantially reduced; consequently the production costs and the loss to society also.
© 2010 Elsevier Ltd. All rights reserved.

Keywords: C. Colour; D. Glass ceramic; Taguchi's method

1. Introduction

The conventional method to develop coloured glazes for ceramic tiles, used in many companies, is based on intuitive approaches involving the knowledge about the behaviour of pigments in glazes available in the company. Pigments and the type of glaze (opaque or transparent glazes) are chosen to match the required colour (called the standard colour) according to the experience of the technician. The difference between the standard colour and the developed colour is obtained by a colorimeter.

Another approach for colour matching is based on the Kubelka–Munk theory.[1–4] This theory is used to predict the colour of a glaze containing more than one pigment. Kubelka–Munk theory suggests an approach that offers possibilities for relating the visible reflectance data of a glaze to reflectance data on the individual pigments contained in that glaze. To do this, it is necessary a spectrophotometer, which is not normally available in companies. Colorimeters have the ability to detect and measure small differences between samples that are nearly alike in colour and are less expensive than spectrophotometers. This is why they are used in most cases.

The use of colorimeters requires a considerable number of experiments to obtain the standard colour, which represents a high cost to the company. Glazes approved by the laboratory are then produced in high scale and retested in the client's conditions. At this point, they often need small adjustments (corrections) before being delivered to the client. These corrections significantly increase the costs of the glazes production and decrease de quality of the product because its quality depends not only on its characteristics, but also on its costs of production, development and possible corrections and reparations. To compete successfully in the global marketplace, organizations must have the ability to produce variety of high quality, low cost products that satisfy customer's needs.[5,6]

Geinichi Taguchi introduced several statistical concepts which allow to improve the quality of manufactured goods.

* Corresponding author. Present address: Dep. Ceram. Glass Eng./CICECO, University of Aveiro, 3810-193 Aveiro, Portugal.
E-mail address: anacastela@ua.pt (A.O. Castela).

0955-2219/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2010.05.016

Taguchi's method is a robust engineering one, which has a large applicability. Many Japanese manufacturers (as automobile) have used his approach for improving the product and the process quality with success.

Taguchi's method focuses on improving the fundamental function of the product or process. It may be the most powerful method available to obtain products or process at the lowest cost possible and minimally sensitive to factors causing variability. The cost of a product or process is related with its quality. A product is as better for society as lower is its cost; a lower cost represent a lower loss of quality.[5–7]

The quality of a product depends on factors, some of them being controllable and others not. Uncontrollable factors are called noise factors. The controllable factors and their levels are those selected by the technician who makes the experiences or controls the process. The levels represent the quantities or qualities of the controllable factors. Any uncontrolled factor disturbing the characteristics of the product or the process is a noise factor. Products should be designed in a way to be immune to uncontrollable factors. After the selection of the factors and their levels, these are introduced in an orthogonal array. The choice of the orthogonal array depends on the number of controllable factors and levels. This array helps to determine the best combination of factors and levels with a small number of experiments. It also allows evaluating the interaction between factors, particularly important in case of formulation of colours.[5,7–10]

To analyse the results, Taguchi uses signal-to-noise ratio (S/N ratio). The S/N ratio quantifies the variation between the target value and the obtained result. The S/N ratio depends on the characteristics of the product and can be found for "lower is better", "nominal is best" and "larger is better", depending on what is the aim of the experience. If, for example, the desired product is related to a target size, the S/N ratio is found by the expression "nominal is best"; if the desired product is related to a characteristic of efficiency, the S/N ratio is found by the expression "higher is better". In this work, it must be used the "lower is better" case, because the desired colour must be as close as possible to the standard one, i. e, the difference between colours should tend to zero. In this case, the S/N ratio is expressed by: $S/N_{LB} = -10\,\mathrm{Log}_{10}\left(\sum_{i=1}^{n} y_i^2/n\right)$ is expression (1); where y is the observed values and n is the number of observations. The values of S/N ratio are expressed by decibel. The experimental results are analysed by using the S/N ratio values as the objective characteristics. It is calculated the mean of the S/N values to each level of each factor. The optimal level of each factor is the level with the highest S/N ratio mean. Then, it is performed an analysis of variance (ANOVA) to verify which factors are statistically significant. With the analysis of the S/N values and ANOVA it is possible to predict the optimal combination of factors.[5,7–11]

The aim of its work was to develop a method which can be used in most companies without the knowledge of the pigments spectral reflectance. In this way, it was evaluated the ability of the Taguchi method in the development of two coloured glazes for tile applications using a colorimeter in order to measure the difference of colours. The developed colours was a green one (reference 14C) and a grey one (reference 14B). It is expected a reduction of the number of experiments necessary to develop new coloured glazes for tile applications and to prevent after production corrections.

Table 1
Chemical elements (besides oxygen) and references for the pigments used in this work.

Reference	Colour	Chemical elements
A	Yellow	Zr, Si, Pr
B	Blue	Zr, Si, V
C	Black	Fe, Cr, Co
D	Pink	Zr, Si, Fe

2. Experimental procedure

To produce the coloured glazes, four pigments were selected, black, pink, yellow and blue, and a transparent base glaze was used for glaze 14C and on opaque base glaze for glaze 14B. These base glazes are normally used by the client. The chemical elements and the references of the pigments are given in Table 1.

Pigments were weighted in an AND HF 3000 g (±0.01 g) balance and then mixed with the base glaze in a balls fast mill, with a capacity of 250 g, for 5 min. The density of the base glazes was 1820 g l^{-1} and the drain velocity, measured by Ford viscosity (ASTM D1200), was 500 s l^{-1}. These are good client conditions for the application of the base glazes. After milling, the coloured glazes were applied to ceramic bodies and then fired in the client's kiln (Barbieri & Tarozzi, 1996) in a firing cycle of 1095 °C, for 34 min. After firing, the colour control was performed using CIElab system by using of a colorimeter (Datacolor Color Tools). ΔE values were obtained through the measures of L* (black and white), a* (green–red) and b* (yellow–blue) parameters. ΔE measures the difference between the standard colour and the one under analysis in terms of the tone and intensity using a light source.[12] It was calculated the values of S/N ratio using expression (1) and analysed the results of the means and of the variance (ANOVA) (Appendix A).

3. Results and discussion

A series of steps, the planning, the conduction and the analyses and interpretation of the results was performed. It is important to know the purpose of the experience, i. e., what the desired product is and what factors actuate on the characteristics of the product (controllable and uncontrollable factors).[5,7–10] The controlled factors and levels were, respectively, the pigments and their amounts. It was selected two levels for each factor and it was chosen an orthogonal array with the lowest resolution: a L_4 array, when it was selected three factors with two levels, and a L_8 array, when it was selected four (or more) factors with two levels. Therefore, the experimental trials were arranged in a L_4 orthogonal array to obtain the 14B coloured glaze and a L_8, to obtain the other glaze (14C). If it was used a full factorial experimental design, it would be necessary to do eight trials (2^3) for glaze 14B and sixteen trials (2^4) for the 14C one.

Table 2
Controlled factors (pigments) and levels (quantities of pigments) for 100 g of opaque glaze to obtain the 14B coloured glaze.

Factors	Level 1 (g)	Level 2 (g)
A (yellow)	0.15	0.18
B (blue)	1.40	1.70
D (pink)	0.80	1.10

Table 3
Trials to obtain the coloured glaze 14B, the ΔE determinations and the S/N values.

Trials	D (pink)	B (blue)	A (yellow)	ΔE	S/N
1	1	1	1	1.3	−2.3
2	1	2	2	2.2	−6.8
3	2	1	2	2.1	−6.4
4	2	2	1	1.1	−0.8

The results of these trials were used to evaluate the influence of the factors and to determine their optimum combinations. It was calculated the means of the S/N values for each level, designated by level 1 mean and level 2 mean, for each entry (factor or interaction of factors). The higher the difference between level 1 and level 2 means, the higher the influence of that factor (or interaction of factors) on the composition. The best combination is that one formed by the highest S/N mean values. Then, it was made an analysis of variance (ANOVA) over these S/N values.[7] The ANOVA helps to choose the pigments with significant influence on the desired colour. If the effect of one pigment is significant, its level should be the same as in the best combination of factors, but if it is not significant, its level should be the lowest one in order to reduce costs.[5,7–10] In the following, the two coloured glazes are analysed separately.

3.1. 14B coloured glaze

In order to obtain the 14B coloured glaze it was selected three pigments (Table 1), yellow (A), blue (B) and pink (D). The desired colour of this glaze is grey: with blue and pink it is possible to obtain this colour and yellow is used to regulate the colour and the tonality. Table 2 shows the factors and the selected levels for this colour. They were mixed with 100 g of an opaque base glaze.

Table 3 shows the trials that were performed according to a L_4 orthogonal array. It was used an L_4 array because it was selected three factors and two levels. The experimental ΔE values and the calculated S/N ratios are also presented in this table.

It can be seen in Table 3 that the trial with the best result (that with the higher value of S/N) is trial number 4, with combination $D_2B_2A_1$.

The best combination has to be close to the standard colour, but costs must also be minimized. Therefore, it is necessary to analyse the result of the means of the S/N values and then the variances. Table 4 presents the S/N means resulting from the trials. It is possible to see in this table and in Fig. 1 the difference between level 1 and level 2 of each factor. The highest variation between levels is found for the yellow pigment (A) and the lowest is found for the blue one (B). Therefore, the variation of A is more significant to obtain the desired colour than the variation of B. The best combination is obtained by selecting, for each factor, the level with the highest mean value. In this case it is the $D_2B_2A_1$ combination, already found in trial number 4 (Table 4).

Table 4
S/N mean values for level 1 and level 2 for each factor of 14B coloured glaze and ranking of the factors. The ranking is based on the "difference between means", from the most influencing factor (#1 in the ranking) to the less one (#3).

	D (pink)	B (blue)	A (yellow)
Level 1 mean	−4.56	−4.36	−1.55
Level 2 mean	−3.64	−3.84	−6.65
Difference between means	0.93	0.52	5.09
Ranking	2	3	1

Table 5
ANOVA results of S/N of the obtained values, presented in Table 3 for the 14B coloured glaze. The columns have the following meanings: DF, degrees of freedom, SS, sum of square, MS, mean square, *F*, fisher value, *P*, probability.

Factor	DF	SS	MS	*F*	*P* (%)
D (pink)	1	0.86	0.86	3.14	2.17
(B) (blue)	(1)	(0.27)	(0.27)		
A (Yellow)	1	25.94	25.94	94.66	94.80
Error+ B	1	0.27	0.27		3.04
Total	3	27.07			100

Table 5 shows the ANOVA results. The variance of the B factor was much lower than the variance of A and of D and, consequently, it was pooled in the error.[7] The contribution of pigment A was about 95% and only this factor has a calculated value of F higher than the tabulated one for a 95% confidence level (161.44). Because of this, the quantities of the other pigments on the final combination can be the lower ones, reducing in this way the costs associated with their amounts. Therefore, the best combination for this glaze is $D_1B_1A_1$. This was already tried in trial number 1 (Table 4) and the result is still good: this combination shows $\Delta E = 1.3$ (S/N = −2.3), which is close to 1.1, the ΔE value for $D_2B_2A_1$ combination. Therefore, the optimum combination of factors to obtain this glaze is $D_1B_1A_1$, the combination with lower costs and probably less prone to after production corrections since we are using the lowest amount of pigments.

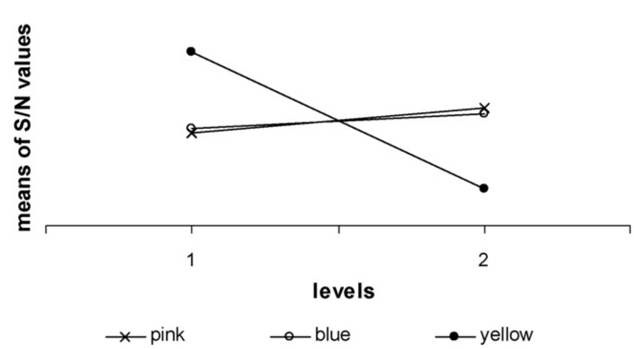

Fig. 1. Graph which represents the influence of each factor in the development of 14B coloured glaze.

Table 6
Controlled factors (pigments) and levels (amounts of pigments) for 100 g of transparent glaze to obtain the 14C coloured glaze.

Factors	Level 1 (g)	Level 2 (g)
A (yellow)	5.00	6.00
B (blue)	3.00	3.50
C (black)	0.03	0.05
D (pink)	0.00	0.10

Table 7
Trials to obtain the coloured glaze 14C, ΔE results and S/N ratio. The empty columns were used to study the interactions between factors (A × B, A × C and B × C).

Trials	A	B	A × B	C	A × C	B × C	D	ΔE	S/N
1	1	1	1	1	1	1	1	3.6	−11.1
2	1	1	1	2	2	2	2	2.2	−6.8
3	1	2	2	1	1	2	2	4.4	−12.9
4	1	2	2	2	2	1	1	3.9	−11.8
5	2	1	2	1	2	1	2	4.3	−12.7
6	2	1	2	2	1	2	1	4.9	−13.8
7	2	2	1	1	2	2	1	5.9	−15.4
8	2	2	1	2	1	1	2	1.8	−5.1

3.2. 14C coloured glaze

Table 6 shows the pigments (factors) and their amounts (levels) selected to obtain the 14C coloured glaze. It was chosen the yellow and the blue pigments to obtain the green colour and this colour and its tonality were regulated by the pink and black pigments (Table 1). The pigments were mixed with 100 g of a transparent glaze.

The trials were arranged according to a L_8 orthogonal array, the one shown in Table 7, due to the fact that it was selected four pigments with two levels. The empty columns were used to study the interactions between factors (A × B, A × C and B × C). Table 7 shows the ΔE values and S/N results calculated to "lower is better". The S/N mean results and their differences are shown in Table 8 and in Fig. 2.

In Table 7, it is possible to verify that the best result is achieved by combination number 8, $A_2B_2C_2D_2$. By the "difference between the means" of levels (Table 9), it is observed that the C and D pigments and the A × B interaction are the factors that more significantly influence the desired 14C coloured glaze. The green colour is obtained by the mixing of colours A (yellow) and B (blue) and it is the interaction of them (A × B) that mostly affects the desired colour, besides the corrections induced by C (black) and D (pink). As done before for colour 14B, the best combination is the one for which the factors are in the levels with the highest S/N values. Looking in Table 8, the best combination for this case is $(A \times B)_1C_2D_2$. It must be now analysed the meaning of level $(A \times B)_1$.

In Table 7, it is possible to see that the level $(A \times B)_1$ corresponds to the trials where both A and B factors are in the same level, i. e, where we have the combinations A_1B_1 or A_2B_2. Therefore, to obtain the 14C coloured glaze the best combinations are $A_1B_1C_2D_2$ and $A_2B_2C_2D_2$.

The last result is confirmed by the ANOVA (Table 9). The variance of B is the lowest and it was pooled in the error.[7] The ANOVA results indicate that C, D and the A × B interaction are the highest contributions to the composition of the desired colour, with contributions between 19.97% and 26.84%. The values of the calculated F reveal that these factors, with F higher than the tabulated F for 95% confidence level ($F_{0.95;1;1} = 161.44$), have a significant influence in the composition. If a factor is indicated to have a significant influence on the

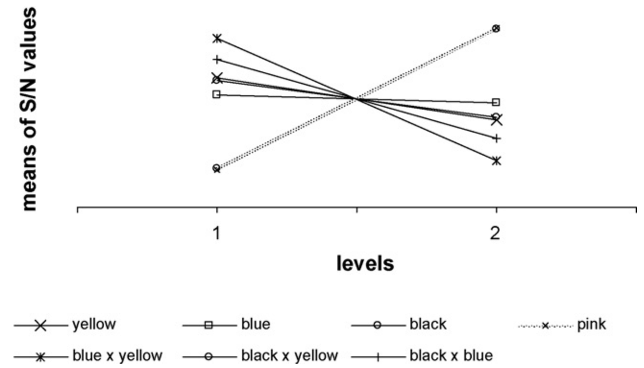

Fig. 2. Graph which represents the influence of each factor in the development of 14C coloured glaze.

Table 9
ANOVA results of S/N of the obtained values, presented in Table 7 for 14C coloured glaze. The columns have the following meanings: DF, degrees of freedom, SS, sum of square, MS, mean square, *F*, fisher value, *P*, probability.

Factor	G.L.	SS	MS	*F*	*P* (%)
A (yellow)	1	2.34	2.34	32.05	2.26
(B) (blue)	(1)	(0.07)	(0.07)		
A × B	1	20.06	20.06	274.13	19.97
C (black)	1	26.29	26.29	359.34	26.20
A × C	1	1.85	1.85	25.35	1.77
B × C	1	8.44	8.44	115.34	8.35
D (pink)	1	26.92	2.92	368.00	26.84
Error+ B	1	0.07	0.07		14.60
Total	7	85.98			100

Table 8
S/N mean values for level 1 and level 2 for each factor of 14C coloured glaze and ranking of the factors. The ranking is based on the "difference between means", from the most influencing factor (#1 in the ranking) to the less one (#7).

	A (yellow)	B (blue)	A × B	C (black)	A × C	B × C	D (pink)
Level 1 mean	−10.67	−11.12	−9.62	−13.02	−10.72	−10.18	−13.04
Level 2 mean	−11.75	−11.30	−12.79	−9.39	−11.70	−12.23	−9.37
Difference between means	1.08	0.19	3.17	3.63	0.96	2.05	3.67
Ranking	5	7	3	2	6	4	1

final results, it should be used its level that better agrees with the desired target. Now, it must be choose the optimum composition, either $A_1B_1C_2D_2$ or $A_2B_2C_2D_2$. Following Taguchi, the optimum composition should be the one corresponding to lower costs.[5,8] Looking to Table 7, it can be seen that composition $A_1B_1C_2D_2$ (trial 2), gives a ΔE value ($\Delta E = 2.2$) higher than $A_2B_2C_2D_2$ composition, with $\Delta E = 1.8$ (trial 8). It is a rule inside the company that $\Delta E = 2.0$ is a threshold value for a colour under development, above which other trials should be done. Therefore, although composition $A_1B_1C_2D_2$ should present a lower cost only due to the lower amounts of pigments A and B, it must be ruled out due to its high ΔE value. Then, the optimum composition to obtain the desired green (colour 14C) is $A_2B_2C_2D_2$.

The two coloured glazes, $A_1B_1D_1$ for colour 14B and $A_2B_2C_2D_2$ for colour 14C, were approved for production in high scale. As already mentioned in the introduction, prior to deliver to the client, they were both retested in the client's conditions for a final assessment and no corrections were necessary.

It must be emphasized that, during this work, it was not possible to include other controllable factors on the design of experiments, since they were related to the client's conditions. During the time scale of months the colour of the ceramic body and slight variations of the firing conditions (the maximum temperature of firing or the firing atmosphere) can influence the colour of the glaze. The number of experiences to develop the new colours was low. Comparing with previous similar cases, it is possible to estimate 30–40% reduction in the number of trials necessary to achieve the goals. Due to the limited cases studied, it is not possible to conclude about the efficiency of the method to reduce the need for corrections after the high scale production.

4. Conclusion

Two new coloured glazes were developed in the company (Endeka ceramics) and they were fired in its client company.

Using Taguchi's Method, the number of trials was reduced when compared with the usual try-and-error method, where it is usual to do several aleatory trials to obtain the desired colour. The orthogonal array is a fractional factorial involving several factors which depend on the characteristic of the product. The orthogonal array, the means analysis and the ANOVA where used to study the effect of the factors and its interactions and allows to choose the optimum combination. It is thus possible to choose the combination which presents the desired characteristics and involves the lower costs. In this way, the production costs were reduced, the lost to society is lower and therefore the quality of the product is higher.

Appendix A. Expressions to ANOVA[10–12]

Variation	Between samples (BS)	Within samples (WS – error)	Total
Grades of freedom	$K-1$	$N-K$	$N-1$
SS (sum of squares)	$\sum N_j(\bar{y}_i-\bar{y})^2$	$\sum(\bar{y}_{ij}-\bar{y}_j)^2$	$\sum(y_i-\bar{y})^2$
MS (mean squares)	$\frac{\sum N_j(\bar{y}_i-\bar{y})^2}{K-1}$	$\frac{\sum(\bar{y}_{ij}-\bar{y}_i)^2}{N-K}$	
F	$\frac{MS(BS)}{MS(WS)}$		
P (%)	$\frac{SS(BS)-MS(WS)}{SS(total)}\times 100\%$		

References

1. Murdock SH, Wise TD, Eppler RA. Predicting the color of a ceramic glaze. *American Ceramic Society Bulletin* 1990;**69**:228–30.
2. Eppler RA, Eppler DR. *Glazes and glass coatings*. USA: The American Ceramic Society; 2000.
3. Schabbach LM, Bondioli F, Ferrari AM, Petter CO, Fredel MC. Colour in ceramic glazes: efficiency of the Kubelka–Munk model in glazes with a black pigment and opacifier. *Journal of the European Ceramic Society* 2009;**29**:2685–90.
4. Bondioli F, Manfredini T, Romagnoli M. Color matching algorithms in ceramic tile production. *Journal of the European Ceramic Society* 2006;**26**:311–6.
5. Ross PJ. *Taguchi techniques for quality engineering*. 2nd ed. New York: McGraw-Hill; 1996.
6. Taguchi G, Chowdhury S, Taguchi S. *Robust engineering*. USA: McGraw-Hill; 1999.
7. Roy R. *A primer on the Taguchi method*. SME – Society of Manufacturing Engineers; 1990.
8. Garcia-Diaz A, Philips D. *Principles of experimental design and analysis*. 1st ed. London: Chapman & Hall; 1995.
9. Taguchi G. *Introduction to quality engineering*. Tokyo: Asian Productivity Organization; 1989.
10. Taguchi G. *System of experimental design Taguchi*, vol. I. EUA: Don Clauising, Technical Editor for the English Edition; 1988.
11. Breyfogle III FW. *Implementing Six Sigma*. 2nd ed. Hoboken: John Willey & Sons; 2003.
12. Voelz HG. *Industrial color testing: fundamentals and techniques*. Weinheim: VCH; 1995.