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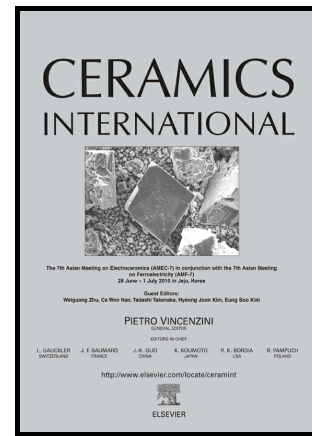
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Improvement of color quality and reduction of defects in the ink jet-printing technology for ceramic tiles production: a Design of Experiments study

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Abstract

The aim of the present work was to study the effect of different process parameters on the color and defects of tiles produced by ink-jet printing technology. The Design of Experiment approach was used to guarantee a rational planning of the experiments and to ensure objective conclusions through the statistical analysis of the data. Particularly, correlations between the process parameters and the quality of decorated tiles in terms of color and presence of surface defects were extensively investigated. Microstructural analysis was used to explain the results derived by the statistical analysis of the data obtained by the rational plan of the experiments allowing further insight in the structural features and mechanisms correlated to the macroscopic properties of the tiles. The study supplied an efficient way to control the final quality of the decorated tiles satisfying the quality standards required by the market demand.

Keywords

Ceramic tiles, ink jet printer, defects, color, design of experiments.

1. Introduction

Ceramic tiles industry has undergone significant changes in recent years to meet the latest market demands [1] [2] [3]. This trend has highlighted the need to control the entire production process to ensure an effective reproducibility of the final material in addition to maintaining high standards of quality. It is known that the production of ceramic tiles is a complex process influenced by many variables ranging from those directly related to the technology to those linked to the environmental conditions during production [4] [5]. These variables directly affect the final product in terms of color, dimension and presence of defects on the surface. [6].

One of the more recent innovations in the production of ceramic tiles regards the decorating technique, and in particular the introduction of inkjet printing technology that enables high quality printing on ceramic support and a wide range of aesthetics properties [7]. The limitations of the traditional decoration techniques such as screen printing done directly onto weak and fragile green bodies, resulting in significant quantities of breakages, inefficient screen printing to the edge of the article and inadequate screen resolution for trichromatic prints, leads to a rapid expansion of the Inkjet printing. Its advantages compared to the conventional methods [8] are mainly related to the fact that it is a digital process and a non-contact method therefore fragile substrates and non-flat substrates, which are difficult to be treated in the conventional printing methods, can be processed and a wide range of materials can be deposited on the substrate (pigments, dyes, glass frits and metallic particles).

Moreover the digital image definition and the flexibility of the process allow a more realistic representation of natural materials that is one of the main interesting effects in the tiles market.

Despite the advantages of this printing technique, its performance in ceramic production strongly depends on how well the printer is integrated in the production line. Different process variables can directly affect the final properties in terms of presence of defects, accuracy and reliability of color intensities over the whole decorated surface. The knowledge of the correlations existing between process variables and final aesthetic properties allows to control and therefore to improve both efficiency and quality of the manufacturing process. This skill represents an extremely powerful tool to design and optimize tiles with specific features encountering the market demands. To achieve this goal a strong collaboration between industrial partners (System S.p.A and Savoia S.p.A) and the research group expertizes in statistical analysis was established leading to an effective interconnection between all the parts.

The analysis of the quantitative relationships between process parameters and the aesthetic features of the ceramic product has been the objective of a wide research in the last two decades with the main aim to improve the color matching (prediction perspective), the automatic detection of both color and morphological defects. It is important to underline that in the inkjet printing technologies the color matching and its management is achieved by the Color Management System (CMS) software and therefore no operator intervention is required adding a more reproducible results if compared to the tintometric system used in the traditional analogic printing.”

Regarding the first task, many efforts were devoted in developing new mathematical and statistical framework and applying several instrumental techniques [9] [6] [10] [11]

[12] in order to increase the efficiency and the rate of quality control. As concern the second task the studies focused the attention on the influence of raw materials grade [13] [14], the firing temperature [15] [16] [17], the viscosity and the amount of deposited ink and of upper glazed [18] on the final colors of the ceramic tiles.

Moreover, M. Lassinanti Gualtieri et al [19], pointed out that the raw material mixture proportions in stoneware affect the amount of amorphous phase leading to a change in the chromatic aspects of the final product in terms of CIELab color parameters.

Regarding the surface grade and hence the aesthetic behaviour of a glazed tile, it was demonstrated the influence of the particle size distribution [20] and the composition of the original glassy matrix on microstructure and crystalline phase content on the surface of glass-ceramic glazes. [21] Moreover the formation of defects is hard to control and could derive by external factors, for example by pollution[22] or internal factors, in most cases the composition, that often leads to an unsuitable interactions among the glass components [23][24] .

The state of the art clearly reports that color rendering and the quality of surface are directly influenced by many factors but despite various attempts to investigate and analyse the quality of decorative tiles, a systematic and real in-line study was only preliminary performed by N. Erginel et al [18].

In this work, the Design of Experiments (DoE) [25], [26] strategy was used to provide an extensive evaluation, through mathematical models, of the correlations between the process parameters and the quality of decorated tiles in terms of color and presence of surface defects. In this way, an improvement of the in-line process control and then of the quality of the final products was ensured.

To achieve this goal several process variables such as temperature of green body (TGB) at the dryer exit, the amount of deposited engobe (AM1), glaze (AM2) and glass (AM3), the resolution of the printer (DPI), the type of glaze (GTY) and the maximum temperature of firing cycle (TMX) were considered in order to verify their potential effect (main or interaction) on the final aesthetic properties in terms of $L^*a^*b^*$ (CIELab). Moreover a *consensus panel* approach was used to obtain qualitative evaluation of the presence of defects in the final material. Further experimental analysis on the final products was conducted to explain the cause –effect mechanisms deriving by the DoE finding.

2. Experimental procedure

2.1 Methodologies and Process

A schematic layout of the tiles production line considered in this work is showed in Figure 1. During the experimental test we decide to keep constant all the variables (manufacturing line, printer, green body, density, viscosity and granulometry of deposited materials) except the ones directly considered in the DoE plan. All the tests were performed by using a dedicated industrial process line in which only the factors indicate in Table 1 were allowed to vary in specific ranges.

It is worth noting that to better control the TGB parameters a manual procedure using a pyrometer was implemented during the experimental test: tiles were blocked in the line after the dryer until the temperature reaches the value requested by the experimental plan.

The openings of the nozzles, positioned along the industrial lines were varied in order to control the amount of engobe (AM1), glaze (AM2) and glass (AM3) deposited on the

tile surface (60x30 cm²). It is important to know that 10 g of deposited materials corresponds to about 65 µm of thickness. The resolution (DPI) is controlled by setting the parameters of the printer with inkjet technology Creadigit (System Group SpA). Two commercial grade glazes (coded as GTY 1 and GTY 2) were used to test the effect of the composition on the final quality of printed tiles. According to conclusions already reported in literature [17] the firing temperature TMX ranges from 1200 to 1220 °C. The aesthetic properties were evaluated using a sample tile (coded as Chart) in which a standard decorative image was printed according to the test used for evaluating the performance of in-line digital printer. The Chart is a matrix divided into several cells in which different amount of ink and/or different types of colors are deposited (Figure 2). To obtain detailed picture of color rendering it was decided to investigate the four primary colors (cyan, brown, beige and yellow) used to print the first four columns (labelled respectively as C1, C2, C3 and C4). The values of color parameters used in the analysis were acquired in the rows labelled R28, R14 and R2 used as reference ones. The 12 cells used in this study are highlighted in Figure 2. For each column, the percentage of ink sprayed by digital printer gradually decreases from 100% of maximum capacity to 3%, moving from top to down of the Chart. This change of ink amount clearly generates different saturation of color between rows: the color became brighter going from top to down. The estimated amount of inks deposited in the three reference rows is approximately 80%, 45% and 5% (R28, 14 and 2 respectively).

2.2 Characterization

ISI hyperspectral scanner has been used to capture the charts of Chart with high resolution. Particularly, color in the specific area of the Chart was measured using a spectrophotometer (no portable) and the CIELab color coordinates (Color Eye XTH,

Greta Macbeth) obtained following the ASTM E 308. The ASTM E 308 provides a standard practice for computing the colors by using the CIE-System.

The CIE Color Systems use three coordinates (L , a^* and b^*) to place a color in a tridimensional space. Specifically, while L^* defines the lightness, a^* denotes the red/green value (negative values indicate green while positive values indicate red) and b^* the yellow/blue one (negative values indicate blue and positive values indicate yellow). Variations of these three parameters ΔL^* Δa^* Δb^* can be used to compare two colors: the total difference (distances in the CIELAB space diagram) can be stated as a single value, known as ΔE^* (eq. 1)

$$\Delta E_{ab} = \left[(DL^2) + (Da^2) + (Db^2) \right]^{1/2} \quad (eq.1)$$

The aesthetical evaluation of the tiles was performed through a consensual panel and the scores obtained were analysed by using statistical methods.

The parameters selected for this qualitative study are: the presence of white dots (Figure 3a), stains, i.e. limited and contiguous portions of region that have different shades, (Figure 3b), macro-roughness detachable by touch and by visual inspection (Figure 3c) and finally color vivacity (Figure 3d).

The results of the statistical analysis of the responses pointed out the cause-effect relationship between process parameters and the final aesthetical properties of the tiles (color and presence of defects on the surface) that were validated by ad hoc experimental characterizations on the inks used and the tiles microstructure. In particular, the viscosity of the inks at different temperature was studied by using Haake rotary viscometer, Rheostress1, with an external drive temperature monitoring.

Moreover scanning electron microscopy ESEM, Quanta 200, FEI, Oxford Instruments)

was used to characterize the microstructure of the glazed surface: cross-sections of the tiles, before and after firing.

The mineralogical composition of the two glazes named (GTY 1 and GTY 2) and the glass was determined by X-ray powder diffraction (XRD). For X-ray diffraction (XRD) the samples were crushed to a fine powder ($< 38 \mu\text{m}$) with pestle and mortar. XRD analysis was performed using a PANalytical X'Pert PROmdiffractometer (Cu- α radiation $\lambda = 1.5405 \text{ \AA}$). Diffraction patterns were acquired on finely ground samples for 2θ values ranging from 5 to 80 degrees.

2.3 Experimental design

In this work the Design of Experiment (DoE) approach was used to guarantee a rational planning of the experiments, which ensures the acquisition of the maximum significant information about the effect of the systematic and multivariate variation of the process variables on the final properties of materials. This method was largely used in the recent years in research [16], [19], development [27] and production [28].

In this study, seven parameters were considered as input factors and varied in a specific range (Table 1). For the categorical variables related to the type of the glaze, the two levels correspond to two different type of glaze. Furthermore, the other variables occurring in the industrial process and not specifically considered in this study are kept constant during all tests.

Fractional design with resolution V was selected for the study therefore 30 experiments (Table 2) were planned. The degree of fractional design resolution means that the main effects and the two-factors interactions are aliased with three-factor and higher interactions, as already reported in literature [29] [30]. Design-Expert v8 (Stat-Ease

Inc., U.S.) software was used and the Multiple Linear Regression (MLR) analysis was performed by using R code [31].

The $L^* a^* b^*$ parameters were collected for each color (cyan, brown, yellow and beige) of the 12 cells in the Chart as showed in Figure 2 (boxes); therefore 36 responses parameters for each of the 30 tiles were considered for the model evaluation.

For each specific defect, depending by the quality of the sample (from low to high), three different standards are selected. By comparison with the three standards all the samples were classified in four different groups and then each tile belonging to the subset was ordered from low to high score. In this way, an ordered classification (from 1 to 30) of the 30 samples was achieved by the panel test. Specifically, the classification number equal to 1 corresponds to the lowest score obtained for the investigated defect as well as the score equal to 30 corresponds to the tile with the higher value of the this defect.

The MLR analysis [32] performed for each tile points out that a linear model, as detailed in *eq. 2*, without quadratic terms (interactions negligible) can be used to correlate the independent variables (x_i) with the aesthetical properties of the tiles.

$$y = b_0 + \sum_{i=1}^n b_i x_i + \varepsilon \quad (\text{eq. 2})$$

The p-value is the statistical parameter used to evaluate the significance of the regression coefficient b_i and represents the probability that the coefficient is not significant.

The statistical analysis, presented hereafter considers the coefficients b_i obtained by using MLR having the p-value smaller than 0.05. The standard error on the coefficients is ranging between 0.1 and 0.3. The quality of the fit in terms of regression analysis and the prediction power of the models was evaluated by using the R^2 and R^2_{cv} (obtained by

leave-one-out method [33]) It is worth noting that most of the model present high R^2 and a good predictive power (R^2_{cv}). From an industrial point of view this represents an useful tool directly applicable in the process line to perform adjustment and improvements in order to guarantee required color effects in the final product.

3. Results and discussion

3.1 Characterization of the industrial materials

The X-Ray powder diffraction spectra of the glazes named GTY 1 and GTY 2 (Figures 4 and 5) show that the glaze GTY 2 is more vitreous with respect the GTY 1 and its spectrum presents a broad band around $2\theta = 25^\circ$ typical of a silicate glass with some crystalline peaks which can be attributed to quartz, kaolinite and corundum. The glaze GTY 1 is mainly crystalline and the observed peaks are related to the presence of albite (as main phase), kaolinite, calcite and corundum. The XRD analysis also performed on the glass applied at the end of the process shows a spectrum (not reported) similar to the GTY 2, mainly vitreous with crystalline peaks due to quartz, caolinite and corundum. The viscosity of the inks was evaluated in order to analyze their thermal behavior during the industrial process since it is one of the most important parameters in the tile decoration. Figure 6 shows that the viscosity of the inks decreases increasing the temperature.

3.2 Color analysis and model evaluation

As previously mentioned 36 models are evaluated by using the 30 tiles derived by the fractional design. Each coefficient of the models relates the color coordinate to process parameter as it has shown in *eq. 2*. The statistical parameters summarized in Table 3 demonstrate the validity of the models. The model coefficients for each color (cyan and brown in Figure 7 and beige and yellow in Figure 8) were reported in the same plot.

The most relevant coefficient of the models are discussed considering that two colors are perceivable different when their distance (ΔE) in the CIELab space is greater than two [34].

The first aspect that comes out from the analysis is that the darker color, cyan and brown, are mostly affected by DPI, GTY and AM3.

The negative value of the coefficients of DPI related to L^* and b^* and the positive one associated to a^* suggests that the lightness decreases while the color tone shifts to purplish blue using a resolution of 400 dpi instead of 200 dpi.

Due to the increasing of the thickness of upper glass (AM3) the cyan color turns to brightness (L^* coefficient positive) and more yellow tone (b^* coefficient positive) while the brown color turns to brightness and more greenish blue tone (a^* and b^* coefficients negative). Smaller effect on cyan and brown colors is also due to the type of glaze (GTY), in particular on a^* and b^* values (negative and positive effect respectively) for the cyan and L^* and b^* (positive and negative respectively) for the brown. Yellow and beige are mostly affect by DPI, AM3 and only marginally by AM1 (Figure 8). Either the type or the amount of glaze (GTY and AM2) seems to have no effect on these two tones as well as the TMX and TGB factors. Moreover, the models analysis reveals that only the row R28 and R14 are affected by this factors while R2 is mostly unaffected. An increase of the resolution print (DPI) produces a positive effect on a^* and b^* in the case of beige and only on b^* in the case of yellow while a negative effect was found on L^* in the case of beige and on b^* in the case of yellow. At high amount of engobe (AM1) the value of L^* slightly increases in both the cases (beige and yellow). It is worth noting that the maximum firing temperature (TMX) didn't play any significant effect on $L^*a^*b^*$ parameters. Moreover, the coefficients analysis clearly shows a weaker effect of

the thickness of engobe (AM1) and a negligible effect of the temperature of green body (TGB) on the $L^*a^*b^*$ parameters. Regarding the tones detected in rows 28 and 14, it was noted that they show quite similar trend in particular for the blue and brown colors.

3.3 Defects analysis

As concerning the presence of defects, the statistical analysis of the data deriving by the *consensus panel test* pointed out that only linear terms were found to be statistically significant and practically relevant. The R^2 and R^2_{cv} for the defects analysis (Table 4), indicate that all the models describe quite well the investigated responses notwithstanding the lower predictive power highlighted for the white dots defects. Figure 9 reports the regression coefficients of the linear models and the main conclusion that can be drawn is that increasing the TGB and AM3 the white dots defects result emphasized. Likewise, an increase of AM2 and DPI produces an opposite effect. The effect of TGB on white dots defect can be experimentally explained through the SEM micrograph illustrated in Figure 10 and the inks viscosity trend versus temperature reported in Figure 6.

The SEM image shows that for higher TGB the ink results mainly localized between the deposited engobe and glaze and not above the glaze as provided by the industrial process line. Thus the ink tends to percolate between the grains of the glaze emphasizing the white dots defect. This percolation phenomenon can be related to the fact that the ink viscosity decreases as a function of the temperature as reported in Figure 6. During the process, the temperature of the sample substrate ranges from 80 to 110°C at the dryer exit and decreases until 40 °C to 50 °C respectively in correspondence of the inkjet printing where the ink is deposited. From Figure 8 it is clear that at 50°C the viscosity of the ink is lower with respect to the one at 40°C thus it

is possible that an increase of the substrate temperature promotes the ink percolation resulting into an higher presence of white dot defects on the final material.

The stains and roughness data analysis suggest that using higher value of DPI and AM3 as well as the employment of the GTY 2 glaze type emphasizes these defects.

The presence of stains in the surface of the green tile due to high amount of glass is clearly evident in Figure 11, where the inhomogeneous distribution of the glass leads to the formation of droplets which lead to the presence of stains also after firing. The same effect is produced using the GTY 2 glaze instead of the GTY 1. It is also important to underline that this phenomenon become evident at visual inspection in the part of the Chart in which higher contents of ink are deposited (R28).

As concerns the roughness it is important to note that between the factors previously indicated the AM2 play a negative effect and in particular lower value of AM2 promote higher roughness of the printed surface. Concerning the role of GTY 2 in emphasize the roughness, SEM analysis performed on a selected tile using the GTY 2 glaze clearly pointed out the presence of structural defects as porosity, craters (Figure 12a). Details on porosity can be also obtained from Figure 12b that reports the cross section of the material: the presence of gases developed during the firing results entrapped as bubbles in the glaze (GTY 2) resulting in the increased macroscopic roughness of the tile. This finding confirmed that the bubbles formation is due both to the interaction among glaze layers and engobe and to the reactions at interface that lead to changes in the solubility of different components [23].

Figure 13 shows the cross section of two tiles with different roughness. The sample labelled as “a” contains 45 g of glaze GTY 1 and 15 g of glass and it has low score; the “b” contains 15 g of glaze GTY 2 e 45 g of glass and presents high roughness. It is

possible to observe that in the “b” sample the distribution of the glass seems not to be homogeneous. Here, higher amount of glass are deposited in the surface causing the higher roughness, as obtained by the DoE analysis.

As previously mentioned, using higher value of DPI and AM3 as well as the employment of the GTY 2 glaze type leads to obtain tiles with major defects in terms of stains and roughness. These results suggest that probably the low chemical affinity between the used glass and the ink as well as between GTY 2 with the inks and engobe are responsible of the observed defects.

Concerning the vivacity of tone highest value of DPI and lowest value of AM3 increases this parameter as well as the employment of the GTY 1 glaze. The three main variables affecting the vivacity are those that affect mostly the L^* a^* b^* parameters. This consideration confirms that the qualitative analysis provided by consensual panel can be successfully used to better explain and confirm the result obtained by quantitative analysis (CieLab parameters).

It is worth noting that while the color analysis performed by statistical evaluation of the $L^*a^*b^*$ parameters doesn't show significant correlation effects played by TGB and AM2, on the contrary, these two variables seems to be strongly connected to the presence of defects on the printed surface especially concerning the white dots and micro-roughness defects. In particular higher thickness of glaze, AM2, improves the quality of final product decreasing the aforementioned defects. Moreover high value of glass thickness, AM3, ensure high quality tile meanwhile high level of DPI decrease white points defects but aggravate the stain and the roughness problems.

4. Conclusions

In this work powerful statistical data analysis based on a DoE approach was applied to the inkjet printing of ceramic tile in a standard industrial process. The main interest was to find out regression models able to mathematically correlate the main industrial process variables with the final aesthetical properties of the tile such as color and presence of defects. The industrial process variables were taken fixed except the seven directly investigated in this work. The statistical analysis point out that different variables affect the final color of the tile with respect to the ones directly responsible of the presence of defects. Table 6 reports a summary of the effect of each process variable on the selected final properties.

It is important to note that for the $L^*a^*b^*$ parameters the main influence is played by the printer resolution (DPI) while no effect on the final color of the tile derive by the temperature of the green body (TGB). For the defects analyses it is possible to note that the engobe amount doesn't affect any of the investigated defects. Moreover the maximum temperature of firing (TMX) doesn't influence any final properties. From the application of DoE it was possible to derive mathematical models that can be directly used during the industrial production of tiles as useful beck march to perform “*in line*” adjustment to guarantee the satisfaction of the aesthetical requirements (standard of a specific tile production) for the market demand.

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REFERENCES

- [1] A. M. Berto, “Ceramic tiles: Above and beyond traditional applications” *J. Eur. Ceram. Soc.*, vol. 27, no. 2–3, pp. 1607–1613, Jan. 2007.
- [2] C. Ferrari, A. Libbra, A. Muscio, and C. Siligardi, “Design of ceramic tiles with high solar reflectance through the development of a functional engobe” *Ceram. Int.*, vol. 39, no. 8, pp. 9583–9590, 2013.
- [3] E. Sánchez, J. García-Ten, V. Sanz, and a. Moreno, “Porcelain tile: Almost 30 years of steady scientific-technological evolution” *Ceram. Int.*, vol. 36, no. 3, pp. 831–845, Apr. 2010.
- [4] R. Casasola, J. M. Rincón, and M. Romero, “Glass–ceramic glazes for ceramic tiles: a review” *J. Mater. Sci.*, vol. 47, no. 2, pp. 553–582, Oct. 2011.
- [5] “Digital decoration of ceramic tiles” *Acimac Notebooks*, 2009.
- [6] H. Elbehiery, A. Hefnawy, and M. Elewa, “Surface Defects Detection for Ceramic Tiles Using Image Processing and Morphological Techniques” 2005.
- [7] V. Sanz, Y. Reig, C. Feliu, Y. Bautista, C. Ribes, and M. Edwards, “Technical Evolution of Ceramic Tile Printing” *J. Imaging Sci. Technol.*, vol. 56, no. 5, pp. 1–7, Sep. 2012.
- [8] O. Watanabe, T. Hibino, and M. Sakakibara, “Development of an ink-Jet printing” *Qualicer*, pp. 1–10, 2012.

- [9] G. Peris-Fajarnés, P. Latorre, B. Defez, I. Tortajada, and F. Brusola, “Evaluation of color prediction models in the decoration of ceramic tiles” *J. Ceram. Soc. Japan*, vol. 116, no. 1349, pp. 146–152, 2008.
- [10] J. Prats-Montalbán and A. Ferrer, “Integration of color and textural information in multivariate image analysis: defect detection and classification issues” *J. Chemom.*, no. May, pp. 10–23, 2007.
- [11] S. Kukkonen, “Color features for quality control in ceramic tile industry” *Opt. Eng.*, vol. 40, no. 2, p. 170, Feb. 2001.
- [12] C. Boukouvalas, J. Kittler, R. Marik, and M. Petrou, “Automatic color grading of ceramic tiles using machine vision” *IEEE Trans. Ind. Electron.*, vol. 44, no. 1, pp. 132–135, 1997.
- [13] L. M. Schabbach, F. Bondioli, a. M. Ferrari, T. Manfredini, C. O. Petter, and M. C. Fredel, “Color in ceramic glazes: Analysis of pigment and opacifier grain size distribution effect by spectrophotometer” *J. Eur. Ceram. Soc.*, vol. 28, no. 9, pp. 1777–1781, Jan. 2008.
- [14] M. Llusar, A. Forés, J. A. Badenes, J. Calbo, M. A. Tena, and G. Monrós, “Color analysis of some cobalt-based blue pigments” *J. Eur. Ceram. Soc.*, vol. 21, no. 8, pp. 1121–1130, Aug. 2001.
- [15] F. Bondioli, T. Manfredini, and M. Romagnoli, “Color matching algorithms in ceramic tile production” *J. Eur. Ceram. Soc.*, vol. 26, no. 3, pp. 311–316, Jan. 2006.
- [16] Z. B. Ozturk and N. Ay, “An investigation of the effect of alkaline oxides on porcelain tiles using factorial design” *J. Ceram. Process. Res.*, vol. 13, no. 5, pp. 635–640, Oct. 2012.

- [17] L. M. Schabbach, F. Bondioli, A. M. Ferrari, T. Manfredini, C. O. Petter, and M. C. Fredel, "Influence of firing temperature on the color developed by a (Zr,V)SiO₄ pigmented opaque ceramic glaze" *J. Eur. Ceram. Soc.*, vol. 27, no. 1, pp. 179–184, 2007.
- [18] N. Erginel, B. Dogan, and N. Ay, "The Statistical Analysis of Coloring Problems Faced in Ceramic Floor Tile Industry" in *Key Engineering Materials*, 2004, vol. 264–268, pp. 1693–1696.
- [19] M. Lassinanti Gualtieri, M. Romagnoli, and A. F. Gualtieri, "Influence of body composition on the technological properties and mineralogy of stoneware: A DOE and mineralogical–microstructural study" *J. Eur. Ceram. Soc.*, vol. 31, no. 5, pp. 673–685, May 2011.
- [20] A. M. Bernardin, "The influence of particle size distribution on the surface appearance of glazed tiles" *Dye. Pigment.*, vol. 80, no. 1, pp. 121–124, Jan. 2009.
- [21] J. M. Rincón, M. Romero, J. Marco, and V. Caballer, "Some aspects of crystallization microstructure on new glass-ceramic glazes" *Mater. Res. Bull.*, vol. 33, no. 8, pp. 1159–1164, Aug. 1998.
- [22] M. Carlini, S. Castellucci, E. Allegrini, B. Giannone, S. Ferrelli, E. Quadraroli, D. Marcantoni, and M. T. Saurini, "Ceramic flaws: Laboratory tests and analysis using Scanning Electron Microscope to identify surface defects" *J. Eur. Ceram. Soc.*, vol. 34, no. 11, pp. 2655–2662, Sep. 2014.
- [23] M. Engels and S. Link, "Bubble control in ceramic glazes" *Qualicer*, pp. 219–235, 2004.
- [24] A. Tunali, "Industrial-Scale Application of Characterization Techniques to Improve Surface Quality of Opaque Floor Tile Glazes" *Acta Phys. Pol. A.*, 2014.

- [25] G. E. P. Box, W. J. Hunter, and J. S. Hunter, "Statistics for Experimenters: An Introduction to Design, Data Analysis, and Model Building" *Wiley Ser. Probab. Math. Stat.*, vol. 1st, 1978.
- [26] R. Leardi, "Experimental design in chemistry: A tutorial." *Anal. Chim. Acta*, vol. 652, no. 1–2, pp. 161–72, Oct. 2009.
- [27] K. Mac Namara, R. Leardi, and A. Sabuneti, "Fast GC analysis of major volatile compounds in distilled alcoholic beverages" *Anal. Chim. Acta*, vol. 542, no. 2, pp. 260–267, Jun. 2005.
- [28] H. Kang, J. Park, and K. Shin, "Statistical analysis for the manufacturing of multi-strip patterns by roll-to-roll single slot-die systems" *Robot. Comput. Integr. Manuf.*, vol. 30, no. 4, pp. 363–368, Aug. 2014.
- [29] D. Montgomery, *Design and analysis of experiments*. 2008.
- [30] C. C. Colbourn and J. H. Dinitz, *Handbook of Combinatorial Designs, Second Edition*, vol. 20065545. Chapman and Hall/CRC, 2006.
- [31] {R Core Team}, "R: A Language and Environment for Statistical Computing." R Foundation for Statistical Computing, Vienna, Austria, 2013.
- [32] D. C. Montgomery, E. A. Peck, and G. G. Vining, *Introduction to Linear Regression Analysis*. 2012.
- [33] R. G. Brereton, *Chemometrics: Data Analysis for the Laboratory and Chemical Plant*. Wiley & Sons, 2003.
- [34] Kuehni RG, Marcus RT. "An experiment in visual scaling of small color differences" *Color Research and Application*, pp. 83–91, 1979.

(eq.1)

$$y = y = b_0 + \sum_{i=1}^n b_i x_i + e \quad (\text{eq.2})$$

Figure captions

Figure 1. Scheme of components of the in-line manufacturing tile process

Figure 2. Chart target. C1:blue, C2:brown, C3:beige, C4:yellow. The box highlights the position of the cells used in the present study as reference

Figure 3. Defects on the printed tiles a) white dots b) stains c) roughness d) color vivacity

Figure 4. X-Ray powder diffraction of the glaze GTY 1

Figure 5. X-Ray powder diffraction of the glaze GTY 2

Figure 6. Viscosity of the inks as a function of the temperature

Figure 7. Significant regression coefficients (b_i) ($p < 0.05$) obtained in the color analysis of cyan and brown column

Figure 8. Significant regression coefficients (b_i) ($p < 0.05$) obtained in color analysis of beige and yellow column

Figure 9. Significant regression coefficients (b_i) ($p < 0.05$) obtained in the defects analysis

Figure 10. SEM micrograph of a tile obtained with higher value of TGB with ink positioned between the glaze and the engobe

Figure 11. Tiles before firing showing the glass inhomogeneous distribution

Figure 12. SEM micrograph of the a) tile surface with roughness defect showing open craters b) cross-section that shows the presence of bubbles inside the glaze.

Figure 13. Cross section of tiles a) with low score of roughness defect and b) with high score of roughness defect

Table 1. Factors and their levels

Factors	Notation	Low Level	High Level	Unit
Temp of Green Body	TGB	80	110	°C
Engobe Amount	AM1	60	90	g
Glaze Amount	AM2	15	45	g
Glaze Type	GTY	SM963	SM918	-
Printer resolution	DPI	200	400	dpi
Glass Amount	AM3	15	45	g
Max T of Firing Cycle	TMX	1200	1220	°C

Table 2. Experimental plan

Experiment	T. of green body	Engob Amount	Glaze Amount	Glaze type	Printer resolution	Glass Amount	Max. T of firing Cycle
	TGB (°C)	AM1 (g)	AM2 (g)	GTY	DPI (dpi)	AM3 (g)	TMX (°C)
1	80	60	15	SM693	400	45	1200
2	80	60	45	SM693	200	45	1200
3	110	60	45	SM693	400	15	1200
4	80	90	15	SM693	400	15	1200
5	110	90	15	SM693	200	45	1200
6	80	90	45	SM693	400	45	1200
7	80	90	45	SM693	200	15	1200
8	110	60	15	SM918	400	15	1200
9	80	60	15	SM918	200	45	1200
10	110	60	45	SM918	400	45	1200
11	80	60	45	SM918	400	15	1200
12	110	60	45	SM918	200	15	1200
13	110	90	15	SM918	200	15	1200
14	80	90	15	SM918	400	45	1200
15	110	90	45	SM918	400	15	1200
16	110	60	15	SM693	400	15	1220
17	110	60	15	SM693	200	45	1220
18	80	60	45	SM693	200	15	1220
19	110	90	15	SM693	200	15	1220
20	80	90	15	SM693	200	45	1220
21	110	90	15	SM693	400	45	1220
22	110	90	45	SM693	200	45	1220
23	80	90	45	SM693	400	15	1220
24	80	60	15	SM918	200	15	1220
25	80	60	15	SM918	400	45	1220
26	80	60	45	SM918	200	45	1220
27	110	60	45	SM918	400	15	1220
28	110	90	15	SM918	200	45	1220
29	110	90	45	SM918	400	45	1220

Table3. Statistical parameters for the 36 models obtained for L* a* b*.

	ROW	R^2			R^2_{cv}		
		L*	a*	b*	L*	a*	b*
CYANO	28	0.97	0.93	0.86	0.94	0.88	0.75
	14	0.98	0.95	0.95	0.96	0.91	0.90
	2	0.96	0.61	0.63	0.93	0.30	0.39
BROWN	28	0.96	0.89	0.95	0.94	0.78	0.90
	14	0.98	0.95	0.92	0.97	0.92	0.85
	2	0.94	0.94	0.93	0.89	0.89	0.88
BEIGE	28	0.97	0.98	0.93	0.95	0.97	0.88
	14	0.97	0.98	0.98	0.94	0.95	0.97
	2	0.84	0.92	0.97	0.73	0.86	0.93
YELLOW	28	0.75	0.97	0.98	0.52	0.95	0.96
	14	0.77	0.98	0.98	0.57	0.96	0.97
	2	0.78	0.79	0.89	0.63	0.64	0.83

Table 4. Statistical parameters for the 4 models obtained for the defects analysis

	R^2	R^2_{cv}
White dots	0.75	0.55
Stains	0.84	0.74
Roughness	0.83	0.70
Colour vivacity	0.89	0.80

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Table 5. Relationship between process parameters and the quality of final product highlighted by this study

Factors	Notation	CieLab	White Dots	Stains	Roughness	Vivacity
T of Green Body	TGB	No	Moderate	No	No	No
Engobe Amount	AM1	Low	No	No	No	No
Glaze Amount	AM2	Low	Moderate	No	Moderate	No
Glaze Type	GTY	Low	No	Moderate	High	Low
Printer resolution	DPI	High	Moderate	High	Moderate	High
Glass Amount	AM3	Moderate	Moderate	Moderate	Moderate	Moderate
Max T of Firing Cycle	TMX	No	No	No	No	No

Figure 1

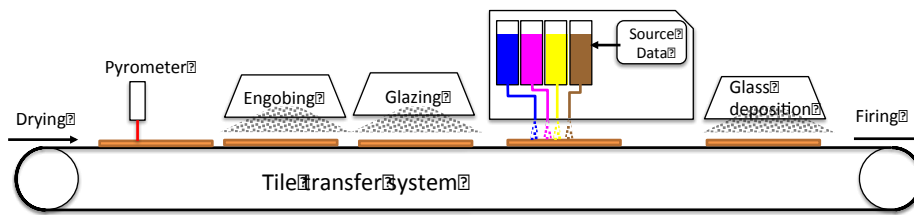


Figure 2

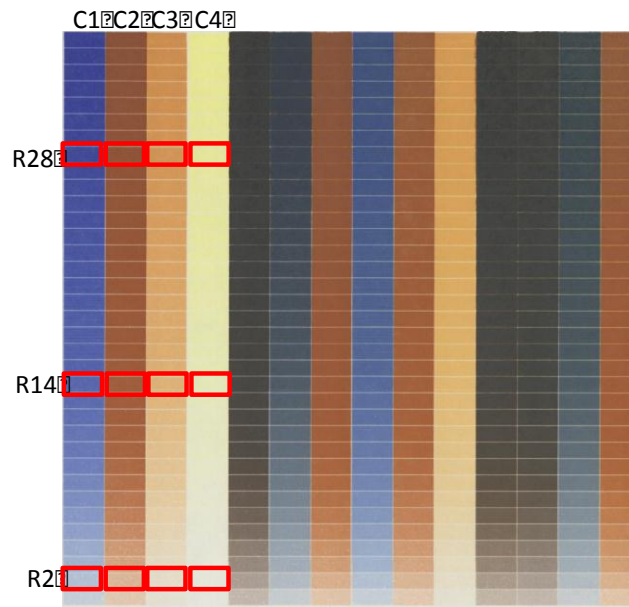


Figure. 3

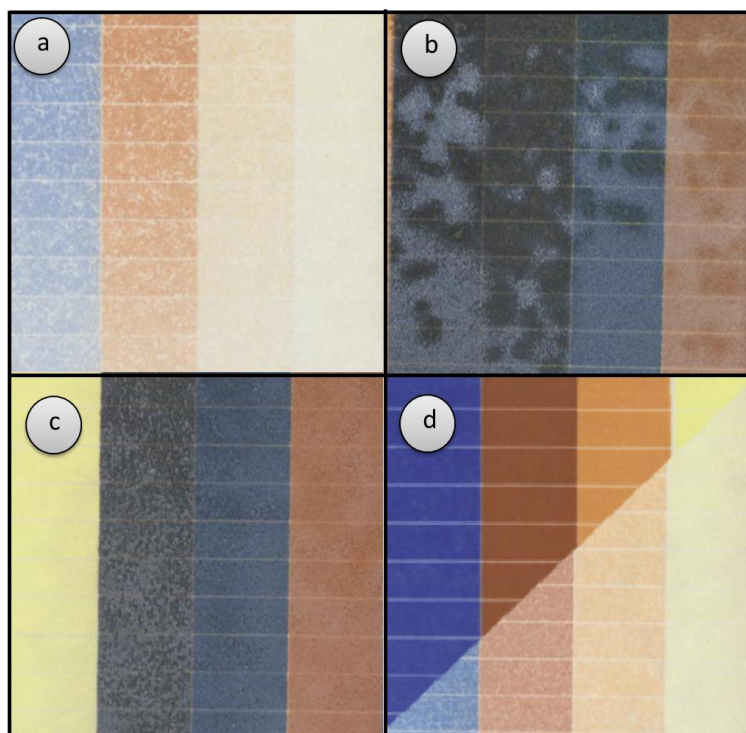


Figure 4

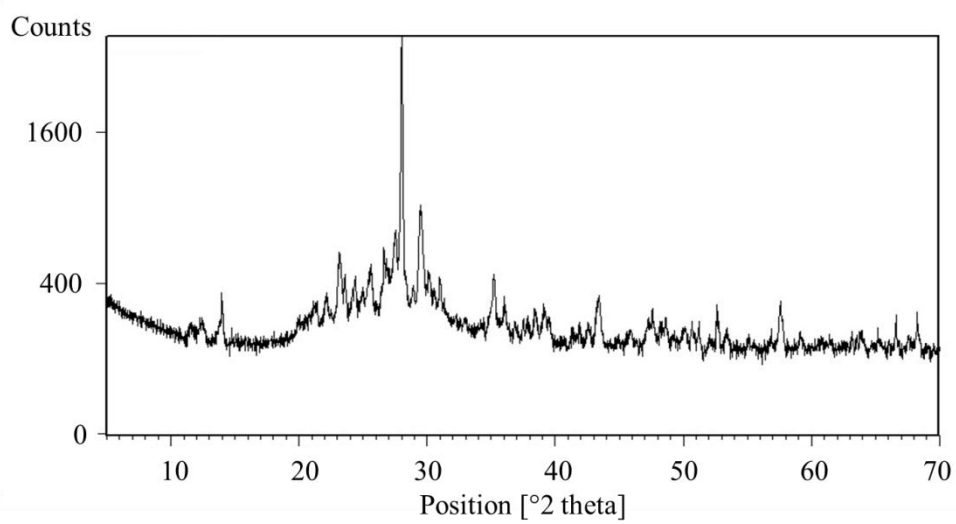


Figure 5

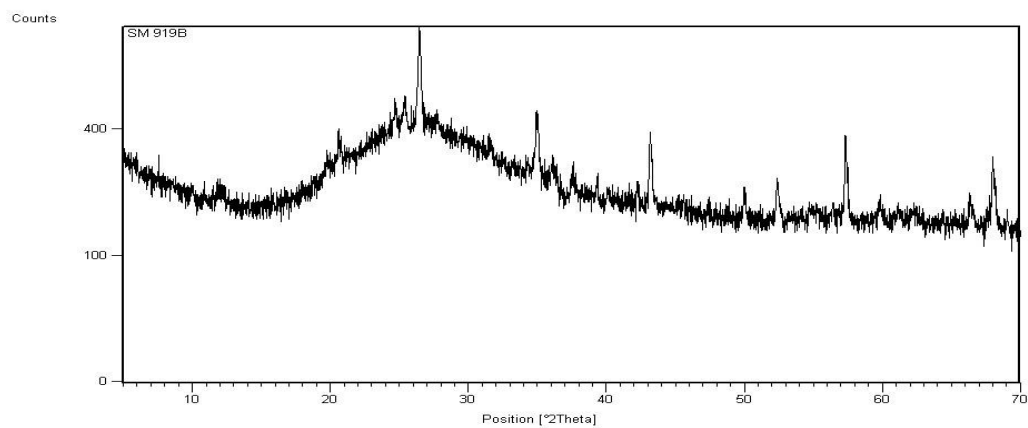


Figure 6

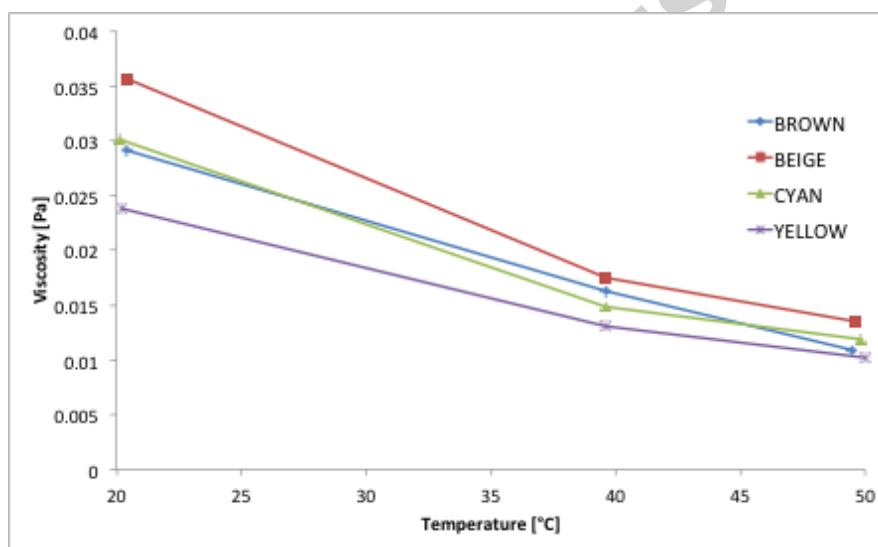


Figure 7

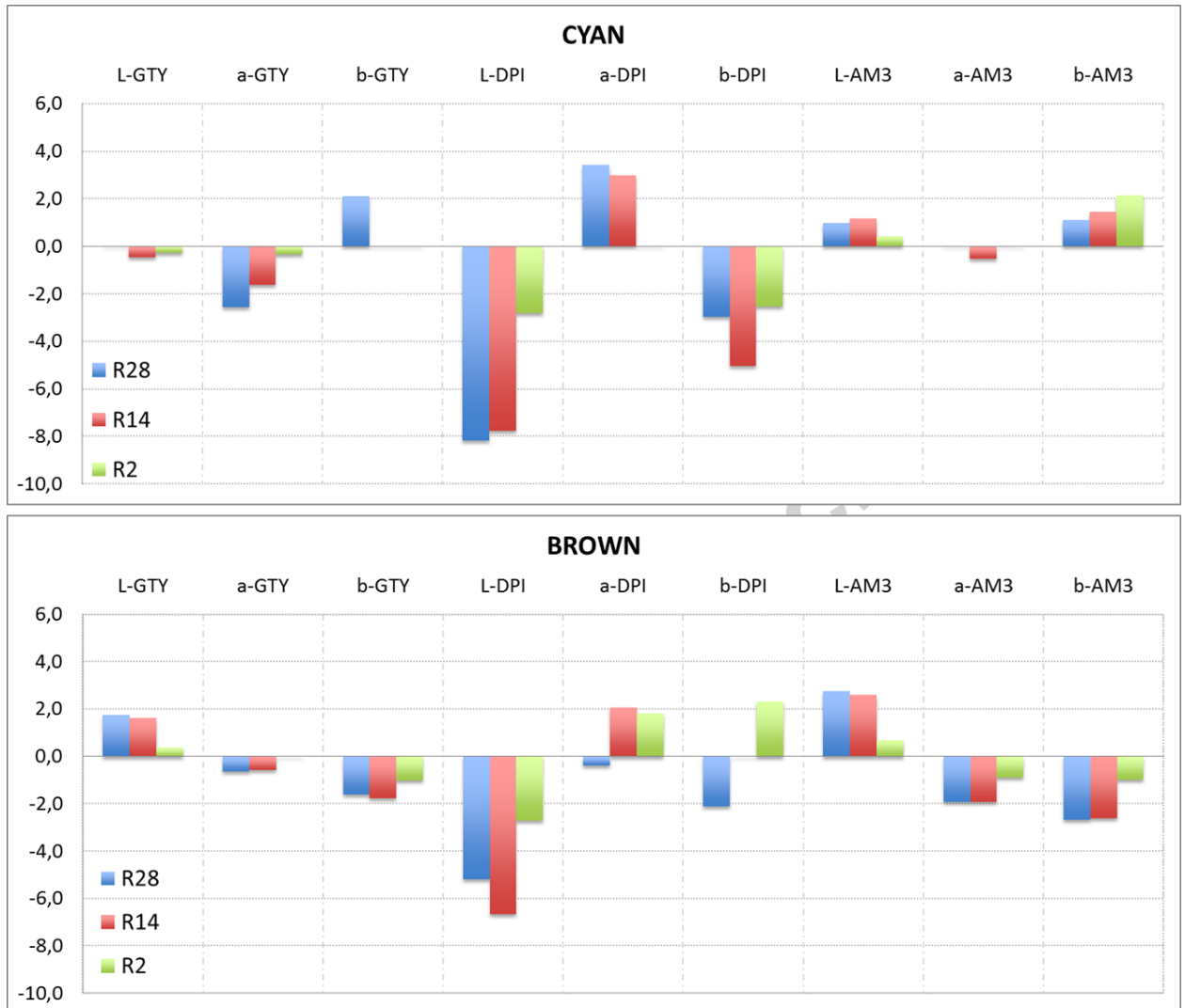


Figure. 8

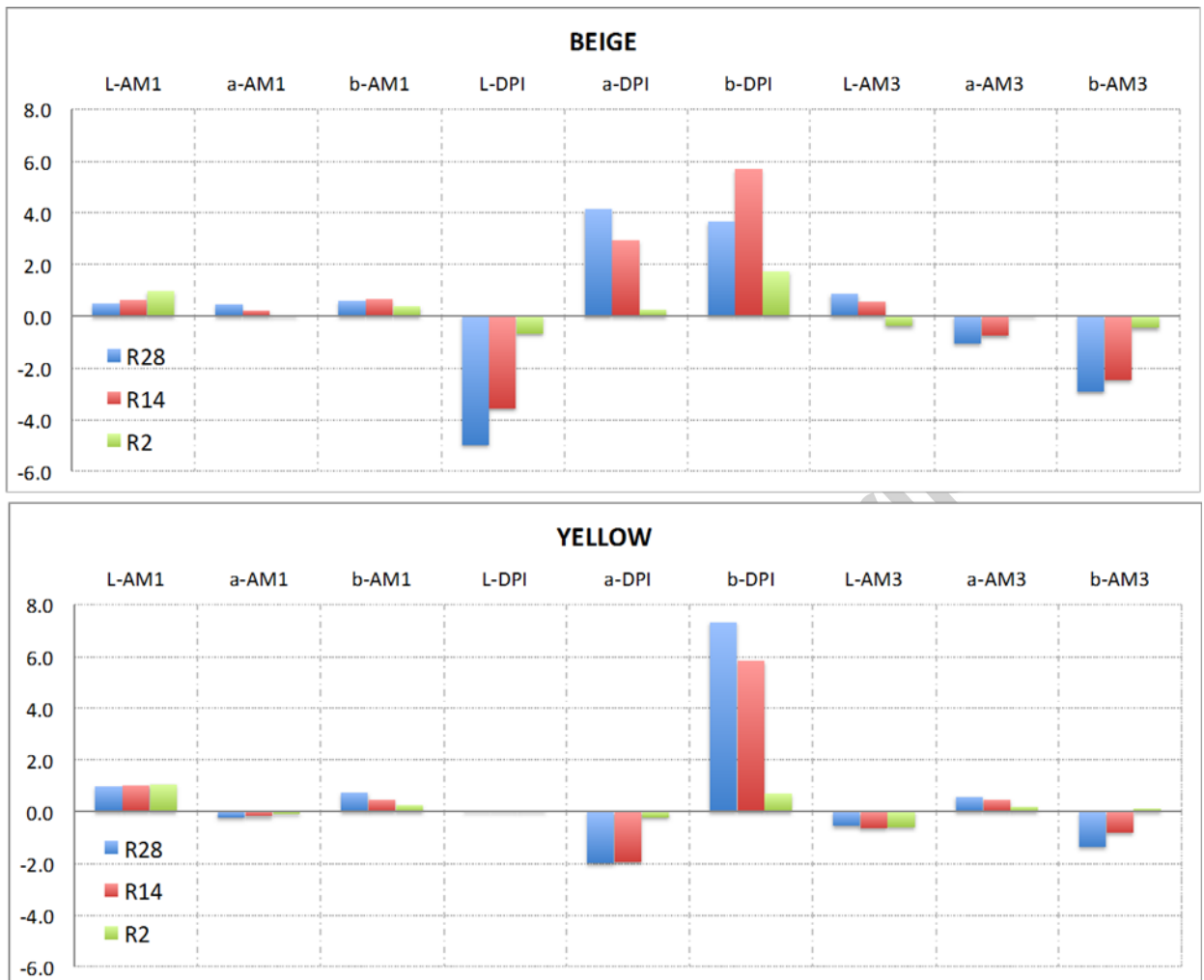


Figure 9

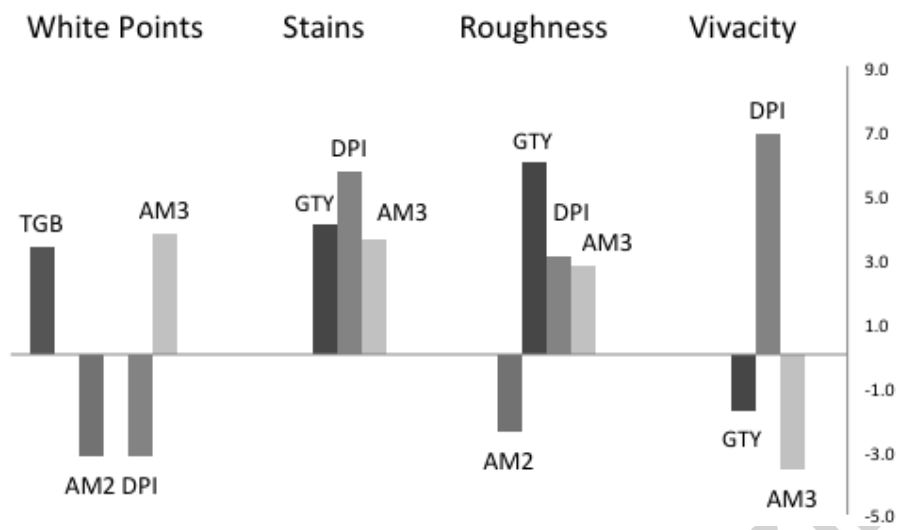


Figure 10

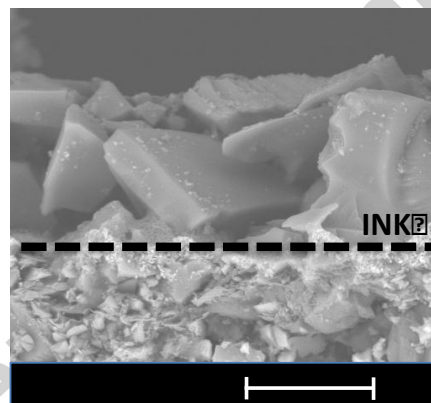


Figure 11



Figure 12

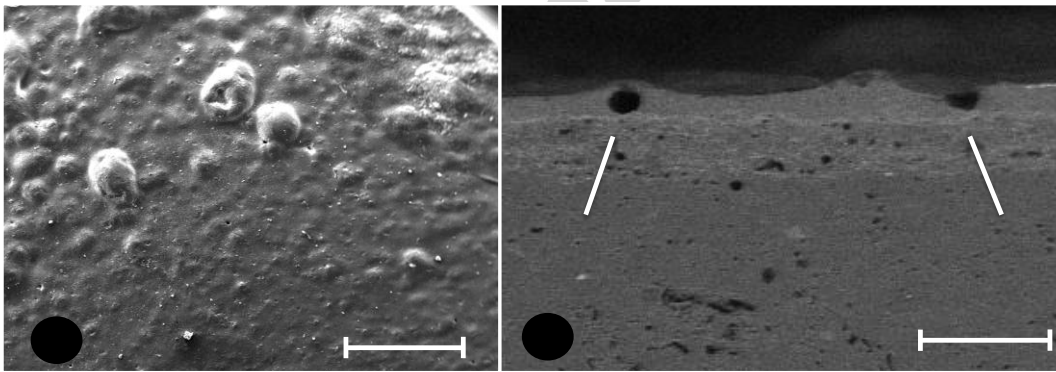


Figure 13

