SIMULTANEOUS OPTIMISATION OF DRYING PARAMETERS OF CERAMIC BODIES

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ABSTRACT

This study deals with the mineralogical, physico-chemical and geotechnical analyses of representative Aptian clays in the north-east of Tunisia. X-ray diffraction reveals a predominance of illite (50-60 wt.%) associated to kaolinite and interstratified illite/smectite. The accessory minerals detected in raw materials are quartz, calcite and Na-feldspar. The average amounts of silica, alumina and alkalis are 52, 20 and 3.5 wt.%, respectively. The contents of lime and iron vary between 4 and 8 wt.%. Physical analyses show that the cation exchange capacity is 34.1-45.7meq/100g of air-dried clay. The plasticity test shows medium values of plasticity index (16-28 wt.%). The linear drying shrinkage is weak (less than 0.99 wt.%) which makes these clays adapt to fast drying. The firing shrinkage and the expansion are limited. A lower firing and drying temperature can be translated into significant energy savings. Currently, these clays are used in the industrial process for the manufacturing of the earthenware tiles. For the better exploitation of the materials and improvement of production conditions, a mathematical formalism is established for the drying parameters. The regression models relate drying shrinkage (d), bending strength after drying (b) and residual moisture (r) with the moisture (m) and the pressing pressure (p).

Keywords: Drying Parameters, Regression Models, Physiochemical Properties, Tunisia

1. INTRODUCTION

There are huge studies that develop the clays characterization used for ceramic products (e.g., Ibrahim et al., 2004; Monteiro and Vieira, 2004; Lisboa et al., 2007; Dondi et al., 2008; Mahmoudi et al., 2008; Meseguer et al., 2010; Diko et al., 2011). In addition, many researches deal with the transformations of clay materials by firing. Indeed, the reactions in the phyllosilicates and accompanying minerals like quartz, feldspar and calcite are decisive to establish the final properties of the ceramic bodies (e.g., Jordán et al., 1999; Carretero et al., 2002; Matteucci et al., 2002; Zanelli et al., 2003; Mao et al., 2006). The disappearance and the neomineralisation of mineral bodies, such as the formation of wollastonite, gehlenite and anorthite is the subject of many works (e.g., Cultrone et al., 2001; Mansur and Mansur, 2003; Aras, 2004; Sousa and Hollanda, 2005; Cultrone et al., 2005; Ferrari and Gualtieri, 2006; Jordán et al., 2008; Jordán et al., 2009; Mahmoudi et al., 2010; Pardo et al., 2011). The influence of the heating rate on phase transformation and mullite formation is devoted by Castelein et al., 2001; Tulyaganov et al., 2002; Sedmale et al., 2006; Sahnoun et al., 2008). It’s clear that the firing stage is important and decisive in the manufacturing of ceramic bodies, but this step is preceded by a drying stage, which can influence and affect the later stages of production (Tari and Ferreira, 1998; Tari et al., 1999). Few studies only are developed in this way. Indeed, the main steps of the drying process consist in the evaporation of the free-water, obtained from the green shaped bodies. Consequently, the particles approach each other, accompanied by shrinkage. The drying behaviour depends on the humidity or moisture (m) and the pressing pressure (p) required for shaping.
K2 strength (b) was measured with a three-point flexural green and dried samples (ISO 10545-4). The bending values of bending strength were calculated by the equation:

\[ B = \frac{3FL}{2yz} \]

where: \( F \) = breaking load (kg), \( L \) = distance between supports (L = 29.67 mm), \( y \) = sample thickness (mm). At the distance between supports (L = 29.67 mm), \( y \) = sample thickness (mm). The bending strength (b) was measured by the three-point flexural method according to the norm ISO 10545. The average approach is to determine regression models of (d), (b) and (r) as function of (p) and (m) is considered.

2. MATERIALS AND METHODS

The measurements of drying shrinkage (d), bending strength after drying (b) and residual moisture (r) were determined in industry conditions. The raw materials were crushed (residue through a 425 \( \mu \)m sieve is less than 0.5 wt.%), moistened (~8 wt.%), mixed and sieved (Ø 2 mm) and then left to rest for 48 h to obtain homogeneous agglomerates. The paste was shaped by pressing and dried in an industrial fast horizontal roller drier (cycle = 12 min, T = 240°C). Ten green specimens (300×75×7 mm) were collected to measure drying parameters for each according to the European Standard (ISO 10545). The linear drying shrinkage (d) was evaluated using the formula: \( D = \frac{l_1 - l_2}{l_1} \times 100 \), where \( l_1 \) and \( l_2 \) are the measured length of green and dried samples (ISO 10545-4). The bending strength (b) was measured by the three-point flexural method according to the norm ISO 10545-4. The average values of bending strength were calculated by the equation:

\[ B = \frac{3FL}{2yz^2} \]

where: \( F \) = breaking load (kg), \( L \) = distance between supports (L = 29.67 mm), \( y \) = sample width (mm), \( z \) = sample thickness (mm). At the outgoing of the drier, the green ceramic bodies were weighed (m1) and dried at 105°C until constant weight (m2). The value of the residual moisture (r) was obtained as follows: \( R = \frac{(m_1-m_2)}{m_2} \times 100 \).

3. RESULTS

Illite is the main mineral (50-60 wt.%) but other minerals; quartz, kaolinite, interstratified illite/smectite, calcite and feldspar, are present in small quantities. Next, this study reveals that the average amounts of SiO2 and K2O are 51.9 and 3.4 wt.%, respectively. The amount of Al2O3 is in average of 19.6 wt.%. The contents of CaO vary between 4 and 8 wt.%. The average amounts of Fe2O3 is 6 wt.%. The grain size data indicate a silt-dominated assemblage. The value for plasticity index ranges from 16 to 28 wt.%. The firing shrinkage and the expansion are limited. The cation exchange capacity is 34.1-45.7 meq/100g of air-dried clay.

According to (Ferrari and Gualtieri, 2006), who used illitic clays for traditional ceramic and showed that the high amount of illite is necessary in ceramic mixtures but it provokes a larger percentage of glass phase, lower water absorption and makes the clays easy to dry. To judge the quality of ceramic paste, the mixture used to produce the ceramic bodies is composed of 80% of Apitian clays and 20% of chamotte.

4. DISCUSSION

With reference to the data obtained (Table 1), regression models can be thought for the drying parameters measured at different pressures (p) and moistures (m).

The mathematical Equation 1-3 are proposed in their canonical form as second-degree polynomials these Equations can further relate drying shrinkage (d), bending strength after drying (b) and residual moisture (r) with the moisture (m) and the pressure (p). The mathematical model is valid, when the error (difference between measured and calculated values) is uncorrelated and randomly distributed with a zero mean value and a common variance (Cornell, 2002; Myers and Montgomery, 2002; Correia et al., 2003).

\[
d(m,p) = \sum_{i=0}^{n} \alpha_i m^p \]

(1)

\[
b(m,p) = \sum_{i=0}^{n} \beta_i m^p \]

(2)

\[
r(m,p) = \sum_{i=0}^{n} \lambda_i m^p \]

(3)

where, d: Drying shrinkage; b: Bending strength after drying; r: Residual moisture; m is the wt.% of moisture (humidity); p is the pressing pressure (bar) applied upon the paste; \( \alpha_i \) ∈ \( \mathbb{R} \); \( \beta_i \) ∈ \( \mathbb{R} \); \( \lambda_i \) ∈ \( \mathbb{R} \) and i ∈ [0 2].

The experiment results are used to calculate the coefficients of the regression equations relating d, b and r with the values of m and p. The calculations are carried out with MATLAB 7.5.0:

\[
d(m,p) = -0.0006p^2 + 0.5220m^2 + 0.0006pm + 0.1858p + 5.5357m + 1.1601 \]

(4)

\[
b(m,p) = -0.0117p^2 + 7.2409m^2 - 0.0082pm + 4.9724p + 90.2445m - 234.5912 \]

(5)

\[
r(m,p) = -0.0023p^2 + 1.5080m^2 - 0.0005pm + 0.9707p - 18.2315m - 44.2979 \]

(6)
The Equation 4 to 6 are the final results and the values obtained from these mathematical models show that d, b and r are 0.35-0.92 wt.%, 0.51-2.23 N/mm$^2$ and 0.01-1.43 wt.%, respectively. The significance and the validity of the mathematical models can also be evaluated by comparing the experimental and the calculated values (Table 2). The difference between these values is low. It is on average 0.03% for drying shrinkage, 0.32 N/mm$^2$ for bending strength, or 0.01% for residual moisture. Diagrams (Fig. 1) show tolerable correlations between variables (R$^2 = 0.989$ for d, R$^2 = 0.986$ for b and R$^2 = 0.982$ for r). However, the prediction is good for all cases of drying shrinkage (d), bending strength (b) and residual moisture (r).

The margin of error between the calculated and the measured values is less than 0.07 wt.%, 0.77 N/mm$^2$ and 0.36 wt.% for d, b and r, respectively. This suggests that the developed model is valid, since the predicted values are consistent with the results obtained from experimental results and the regression equations are considered statistically significant.

The 3D plots are the graphical representation of the Equation 4 to 6 and allow for easy and rapid predictive estimate over the entire properties under investigation. Figure 2 and 3 show that b and r increase with the increase of (p) and (m) amounts.

On the contrary, the drying shrinkage (d) is conversely proportional to the pressure (p) and moisture (m) values (Fig. 4). Indeed, an important compaction of the paste reduces the movement of the particles. Moreover, when the evaporation is incomplete and part of water quantity stays in the pores it can prevent the drying shrinkage. Another way of visualizing the effect that changes in drying parameters (p and m) might have on the d, b and r and prove the interpretation of the statistical results through the use of response plots. In this way, the effect of each property can be visualized.

![Graphs showing correlations](image)

**Fig. 1.** Correlations between measured and calculated values, (d) drying shrinkage, (b) bending strength and (r) residual moisture.
Fig. 2. 3D plot of bending strength as function as the amounts of pressure and moisture

Fig. 3. 3D plot of residual moisture as function as the amounts of pressure and moisture
Fig. 4. 3D plot of drying shrinkage as function as the amounts of pressure and moisture

Table 1. The drying parameters: drying shrinkage (d), bending strength after drying (b), residual moisture (r) and pressure (p) applied upon the paste and moisture (m) of paste before drying

| Pieces | Pressure: p(Kg/cm$^2$) | Moisture: m (%) | Drying shrinkage: d (%) | Bending strength: b: (N/mm$^2$) | Residual moisture: r (%) |
|--------|-------------------------|-----------------|--------------------------|-------------------------------|------------------------|
| 1      | 185                     | 5.4             | 0.99                     | 1.28                          | 0.10                   |
| 2      | 190                     | 5.6             | 0.86                     | 1.43                          | 0.13                   |
| 3      | 195                     | 5.8             | 0.78                     | 1.58                          | 0.20                   |
| 4      | 200                     | 6.0             | 0.66                     | 1.73                          | 0.37                   |
| 5      | 205                     | 6.2             | 0.61                     | 1.76                          | 0.42                   |
| 6      | 207                     | 6.2             | 0.41                     | 1.98                          | 0.44                   |
| 7      | 207                     | 6.3             | 0.52                     | 1.88                          | 0.40                   |
| 8      | 210                     | 6.3             | 0.33                     | 1.94                          | 0.46                   |
| 9      | 210                     | 6.4             | 0.51                     | 1.99                          | 0.48                   |
| 10     | 215                     | 6.6             | 0.40                     | 2.05                          | 0.60                   |
| 11     | 220                     | 6.8             | 0.38                     | 2.10                          | 0.71                   |
| 12     | 225                     | 7.0             | 0.35                     | 2.15                          | 0.92                   |
| 13     | 230                     | 7.2             | 0.31                     | 2.18                          | 1.01                   |
| 14     | 235                     | 7.4             | 0.36                     | 2.22                          | 1.07                   |

Table 2. The errors between measured and calculated values: Drying shrinkage measured ($d_m$), drying shrinkage calculated ($d_c$), bending strength measured ($b_m$), bending strength calculated ($b_c$), residual moisture measured ($r_m$) and residual moisture calculated ($r_c$)

| Pieces | $d_m$ (%) | $d_c$ (%) | Error $d_c-d_m$ (%) | $b_m$ (N/mm$^2$) | $b_c$ (N/mm$^2$) | Error $b_c-b_m$ (N/mm$^2$) | $r_m$ (%) | $r_c$ (%) | Error $b_c-b_m$ (%) |
|--------|-----------|-----------|---------------------|-----------------|-----------------|---------------------------|-----------|-----------|---------------------|
| 1      | 0.99      | 0.92      | 0.07                | 1.28            | 0.51            | 0.77                      | 0.10      | 0.01      | 0.09                |
| 2      | 0.86      | 0.81      | 0.05                | 1.43            | 0.77            | 0.66                      | 0.13      | 0.13      | 0.00                |
| 3      | 0.78      | 0.71      | 0.07                | 1.58            | 1.03            | 0.55                      | 0.20      | 0.26      | 0.06                |
| 4      | 0.66      | 0.62      | 0.04                | 1.73            | 1.25            | 0.48                      | 0.37      | 0.39      | 0.02                |
| 5      | 0.61      | 0.54      | 0.07                | 1.76            | 1.46            | 0.30                      | 0.42      | 0.52      | 0.10                |
| 6      | 0.41      | 0.42      | 0.01                | 1.98            | 1.66            | 0.32                      | 0.44      | 0.56      | 0.12                |
| 7      | 0.52      | 0.53      | 0.01                | 1.88            | 1.52            | 0.36                      | 0.40      | 0.59      | 0.19                |
| 8      | 0.33      | 0.35      | 0.02                | 1.94            | 1.65            | 0.29                      | 0.46      | 0.61      | 0.15                |
| 9      | 0.51      | 0.48      | 0.03                | 1.99            | 1.65            | 0.34                      | 0.48      | 0.66      | 0.18                |
| 10     | 0.40      | 0.42      | 0.02                | 2.05            | 1.81            | 0.24                      | 0.60      | 0.81      | 0.21                |
| 11     | 0.38      | 0.38      | 0.00                | 2.10            | 1.95            | 0.15                      | 0.71      | 0.96      | 0.25                |
| 12     | 0.35      | 0.36      | 0.01                | 2.15            | 2.07            | 0.08                      | 0.92      | 1.11      | 0.19                |
| 13     | 0.31      | 0.35      | 0.04                | 2.18            | 2.16            | 0.02                      | 1.01      | 1.27      | 0.26                |
| 14     | 0.36      | 0.35      | 0.01                | 2.22            | 2.23            | 0.01                      | 1.07      | 1.43      | 0.36                |
5. CONCLUSION

The studied raw materials collected from Aptian clays of Jebel Ressas (north-east Tunisia) show as dominant mineral illite, with accessory phase of quartz, kaolinite, calcite, illite-smectite mixed layers and Na-feldspar. Chemical analysis indicates that these clays are rich in silica, alumina and alkalis. A significant amount of iron oxides is also detected. Physical analyses show that the cation exchange capacity is 34.10-45.7meq/100g of air-dried clay. The plasticity test shows medium values of plasticity index (16-28 wt.%). The linear drying shrinkage is weak (less than 0.82 wt.%) which makes these clays adapt to fast drying. The firing shrinkage and the expansion are limited. A lower firing and drying temperature can be translated into significant energy savings. The ceramic properties and industrial tests show appropriate industrial characteristics of these clays, which enable to find application in the production of bricks and earthenware tiles. On the other hand, regression models are established to quantify the amounts of drying parameters (drying shrinkage, bending strength after drying and residual moisture) as function as pressure applied upon the paste and moisture. The developed model is based overall on measured values of the drying parameters. These models could be instructive for specialists in geotechnical and industrial applications.

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