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PROCEDURE FOR ASSESSMENT OF THE CLAYS SUITABILITY TOWARDS FAST DRYING

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Abstract: During the last 10 years it was shown that Deff – MR curve pattern is unique and very useful. This ability was firstly embedded in the procedure for setting up the fast drying regimes several years ago. This incredible pattern was used again in this invited paper. Actually, if the test samples formed from the same clay do not crack during drying in at least two isothermal experiments in which drying air temperature was different, while drying air humidity and velocity were constant, the unique line can be constructed. This line is linking corresponding Deff – MR curves through the same characteristic point. For simplicity reasons it was chosen to construct and use only the unique line pulled through characteristic E points. Its slope was proposed as the objective criteria for evaluation of the clays suitability for the rapid drying application. It was found that clays, for which the unique line slope is larger, had better dimensional and mechanical properties and were more suitable for the construction of the fast drying regimes.

Key words: fast drying, clay, effective moisture diffusivity, drying sensitivity, industrial monitoring.

1. INTRODUCTION

Drying of clay raw materials is correlated with various phenomena which happen simultaneously such as: mass transfer (from the product surface and throughout the formed product), heat transfer and dimensional variation which, is linked to the water loss and stress generation. Listed phenomena are closely related with the products and raw material intrinsic properties (its geometry, mineralogy, grain size distribution, density, etc.) and with the extrinsic properties of the drying air (humidity, temperature and velocity). That's why the global scale for the assessment of different clay raw materials suitability for standard drying application is still not reported. If the suitability of various raw materials for intensive drying utilization is analyzed the result is even worst.

Actually, many drying sensitivity techniques were reported and used as a tool for comparison of different clays. Bigot, Ratzenberg and Piltz method were mostly recognized [1-3]. However, one has to be aware that these tools beside the general “line up“ info usually do not provides other information’s including the suitability for rapid drying. That is in line with the fact that the same clay can be for example sensitive to drying according to Bigot and highly sensitive according to Piltz in the same time. Besides, it was already confirmed that clays can in some cases have different drying sensitivity for crack formation in traditional and fast drying regime [4].

Vasić has recently reported a novel, more objective and faster model for assessment of clay raw materials suitability for standard drying application in ceramic industry. This model has established a stable link with the interlayer water removal during heating, which is registered on DTA/TG curves at 2000C, and the intrinsic property of the clays expressed as clay mineral content [5].

The upgrades of the traditional and development of the innovative modern drying technologies for ceramic clay raw materials end especially roofing tiles or masonry elements is usually the subject of patents and innovation wrights. That’s why this scientific area is not covered with considerable number of scientific papers. During the last 25 years the three research groups have taken the most credits for transferring the procedures for increasing the energy efficiency of dryers and setting up the optimal drying regimes on the applicative industrial level. Nowadays it is possible to dry commercially some hollow clay products and clay blocks of large size and weight respectively for 5 and 9 hours. Since the ability to minimize the extrinsic influences is easier, it is evident that further research activities should be directed on intrinsic parameters optimization. It is expected that the future drying time reductions and energy savings is possible only if the influence of the intrinsic raw material properties on the internal moisture movement up to the product surface is much more discussed and understood. In other words, the ability to speed up the drying process is inevitably limited with the intrinsic products properties in which the raw material nature has a significant roll.

The German research group has reported a semi - conjugated model which was possible to simulate the full history of drying (evaporation rate and time dependent moisture, temperature and shrinkage profiles). This model was widely applied in ceramic industry predominantly as a tool for assessment of the drying upgrade investment profitability [6-7].

The Netherlands research group has also developed a semi-conjugated model based on the Kitcher receding front concept. Proposed methodology requires the estimation of the real industrial heat and mass coefficients as well as experimental determination of the limiting drying rate on full - scale industrial products in the specially designed modular, laboratory dryer [8]. This model was widely accepted and partly used in the designing process of Marcheluzo or Cosmec&Sacmi modular industrial semi - rapid dryers [9].

The Serbian research group has firstly reported the theory of moisture migration during isothermal drying. According to this theory the Deff – MR curve represent the complete record of internal moisture transport during drying [10]. The procedure for designing the theoretical non - isothermal drying regime in which the mass transport during drying was rigorously controlled was outlined too. The drying regime was segmented. The segments duration was calculated from the corresponding isothermal Deff – MR curves. Proposed rapid regimes were industrially validated in one domestic roofing tile factory. Dried tiles had better dimensional and mechanical properties [11]. The large number of experiments was identified as the objective problem, which had limited the application, of the Serbian model at the beginning. The Box-Wilkinson's orthogonal multi-factorial experimental design was used for the stated model disadvantage minimization. Drying air temperature, humidity and velocity were input parameters. The model outputs were equations for prediction of the first four drying segments duration. In other words, it was possible to calculate any desired drying segments duration which is in the range defined by the experiment matrix by entering the corresponding input values into the outputs model equations [12].

The pioneer attempt to assess the suitability of different clay mixtures for the fast drying process was reported by Zaccaron in 2021. Three clays (plastic, sandy and clay-stone) were selected. Their chemical (XRF), mineralogical (XRD) and particle size distribution were determined. The Box-Wilkinson's orthogonal multi-factorial experimental design was used to create 10 clay mixtures with different clay content. Extruded products were rapidly dried. The drying regime was predefined and has consisted of three zones (wet, natural and dry). Duration of the first two zones was respectively 40 and 30 % of the total drying time. The drying air parameters were respectively 300C, 90% and 1.5 m/s in the wet zone. The temperature, humidity and velocity in the natural zone were continuously changed until values of 900C, 2% and 4 m/s were reached. These values were kept constant in the final zone. The reference slow drying regime was used for comparison. The temperature within the dryer was the only drying air parameters which was controlled in the slow drying regime. Its value was set to 500C. Products were dried until constant mass was obtained. The content of coarser particles, moisture loss and drying shrinkage were outputs of the model. The raw material mixtures in which sandy and clay-stone were present were more suitable for the rapid drying application [13].

Even though various test methods, for raw material characterization and technological behavior estimation, are listed and described, in a systematic way [14-15] the objective criterion for assessment of raw material suitability towards rapid drying application is still not defined. The main objective of this invited paper was to define the scale for comparing the ability of clay raw materials to withstand the intensive drying conditions.

2. MATERIALS AND METHODS

Three clays which had different drying sensitivity according to Bigoth were chosen. The raw material was dried and afterwards simultaneously milled and moisturized. Dried material was sent on XRD and DTA/TG analysis. Powder diffractometer Philips PW-1050 was used for XRD analysis. Two instruments (TA-SDT-Q600 and Derivatograph C) were used for thermal analysis. Instrument one was used for Clays A and B while the instrument two was used for the clay C. Samples were heated in nitrogen atmosphere with the heating rate of 20°C/min up to 1000°C. Roofing tile samples (120x50x14 mm) were extruded on the laboratory press Hendle type 4. Drying plasticity and sensitivity were respectively determined in accordance with Feferkon and Bigoth methodology. Five isothermal experiments were proposed. The preset values of drying air velocity and relative humidity were 2 m/s and 75% respectively. The air temperature was set at 20°C and was then raised in each following experiment by 10°C. The accuracy of drying air regulation was defined in reference [10]. The roofing tile mass and linear shrinkage were recorded during drying. The accuracy of these measurements was 0.01 g and 0.2 mm. Procedure outlined in reference 10 and 16 was used for construction of the Deff – MR curves. The fact that all drying experiments were repeated 4 times has allowed us to determine the flexural strength on dried and fired samples.

Samples which did not cracked were further dried at 105°C ±5°C until no mass difference in two successive measurements was recorded. Remaining samples were then fired. The firing regime (heating rate up to 610°C and from 600 to 100°C) was reported in reference [11]. Roofing tiles were kept at 1000°C for 2h. EN 1024 and EN 538 standards, were used for determination of geometric (twist and chamber coefficients) and mechanical (flexural strength) properties of the dried and fired products.

3. RESULTS AND DISCUSSION

The XRD and DTA/TG results are shown at figure 1. Kaolinite, illite, and small amounts of montmorillonite were found in the raw material A. The raw material B was mostly consisted of kaolinite. Small amounts of Illite were also registered. The montmorillonite, illite, hydro-chlorite and kaolinite were detected in the raw material C. The plasticity and drying sensitivity results are indicating that the raw material A and C are problematic for drying. This is expected result which is caused by the presence of swelling minerals such as montmorillonite.

Table 1. Pfefferkorn Plasticity and Bigoth drying sensitivity

Clay	A	B	C
Moisture cont. after forming (wt. %)	23.10	23.17	24.59
Plasticity index	29.8	28.30	35.1
Pfefferkorn classification	Good	Good	highest
Moisture loss at critical point (wt.%)	9.81	6.41	12.00
Bigoth drying sensitivity	Sesitive	Poorly sensitive	Highly sensitive

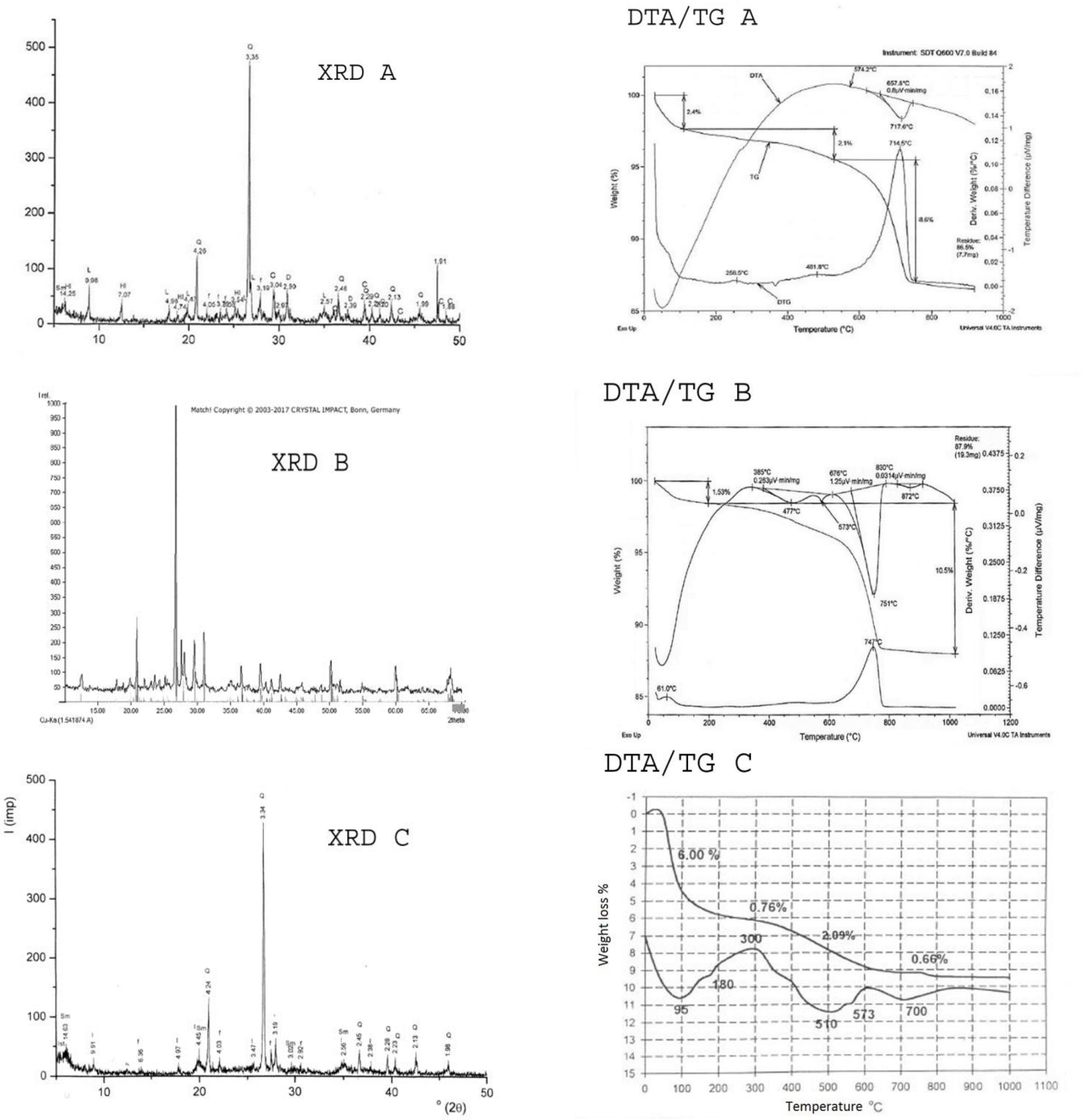
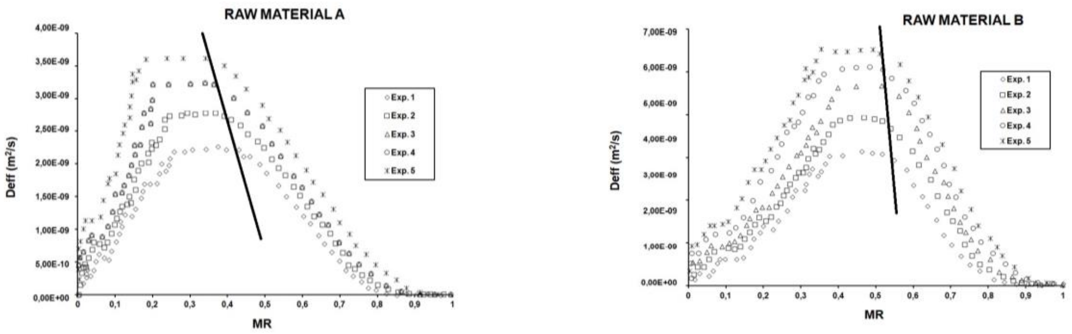


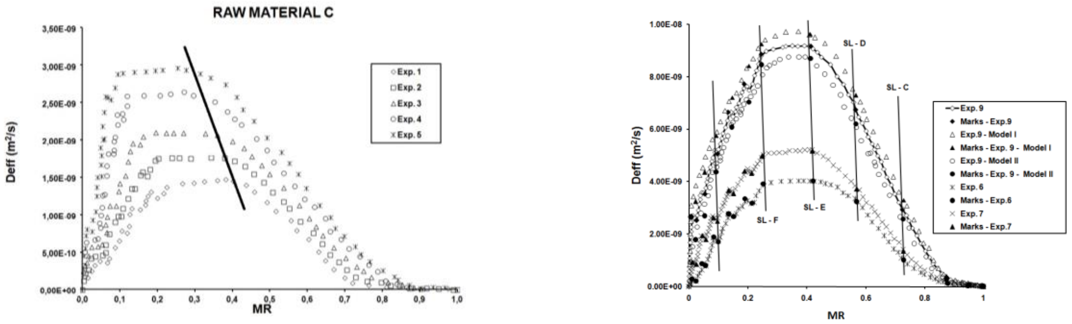
Fig. 1. XRD and DTA/TG

If products extruded from the same clay do not crack during drying in at least two isothermal experiments, in which drying air temperature is different while drying air humidity and velocity are constant, the unique line which passes through the same characteristic point belonging to different experiments can be constructed. If this procedure is repeated and other corresponding characteristic points are connected in the same way, a full set of parallel lines will be created (see figure 2d). It can be said that the registered pattern is the direct evidence that effective diffusivity is properly representing the overall mass transport of moisture. In other word, these pattern represents the complete and unique record of drying history in which all internal moisture transport mechanisms are distinquished. In accordance with the reference 11, the position of the characteristic points for cracked products dried on higher temperatures has to be somewhere on the unique line. The methodology for finding its exact position was reported in the same reference and was used in this study. Even though the drying body surface is not fully covered by a water film the constant drying rate period proceeds up to point E. This point is known as critical point. Final drying simulation outputs for all isothermal experiments and for each raw material are shown at figure 2. For simplicity reasons of not overloading the Deff – MR chart, with the full set of parallel lines only the unique line constructed through the characteristic point E was drawn.

Products extruded from the raw material B were dried without cracks. Cracks were registered on samples formed from raw material A and C in experiment 4 and 3 respectively. In all experiments the effective moisture diffusivity was rising as the drying air temperature was increased. It is obvious that this effect has not the same value for each of the analyzed raw materials. This is expected result, since the mineral composition as well as grain size distributions were different. The results given at figure 2 have pointed out that the unique line slope value is not the same for all three raw materials. In other words this line represents the perfect „finger print“ of any raw material. Up till now this incredible ability was just used in the process of finding the exact position of the characteristic drying segments for samples that were cracked during drying. Even though it was evident that the intrinsic nature of the raw material was built into this pattern so far this fact was not fully used for example as a tool for the evaluation of the clays suitability for the rapid drying application. That’s why the slope of the unique line E was proposed as criteria for comparison of various clay raw materials.



(a) SL line for raw material A (b) – SL line for raw material B



(c) – SL line for raw material C (d) SL pattern (Fig. 9 - reference 11)

Fig. 2. Determined unique SL lines

It is well known that the ideal drying sensitivity scale starts from kaolin and ends with montorillionite. In reality, raw material compositions B and C are representing a boundary situation closest to the ideally "pure" one. That was the reason why these two minerals were chosen in this study. The lowest value of the unique line slope was found in the case of raw material C while the highest value was registered for the raw material B. The unique line slope of any other raw materials will be between those two boundary values. Since the boundary unique lines (fig 2b and fig 2c) are defined the procedure for estimation of the raw material drying sensitivity is not experimentally demanded. Theoretically only two isothermal experiments are necessary to successfully finish the proposed procedure end to construct the unique line of the material which drying sensitivity is unknown (fig 2a).

Results of products shape regularity (twist and chamber coefficients) and mechanical properties of dried and fired products (flexural strength) are presented in Tables 2 and 3 respectively. The mean value of the twist and chamber coefficients calculated as described in EN 1024 shall comply with the requirements stated in Table 1 of the EN 1304. Results in which corresponding coefficients are less than 2% are acceptable. In accordance with the EN1304 the fired products shall be considered satisfactory if, when subjected to the test method described in EN 538 they support without breaking a load of at least 1200 N. The limiting flexural strength of dried product is defined as 600 N.

Table 2. Regularity of products shape

Raw material		A				B					C		
Experiment		1	2	3	4	1	2	3	4	5	1	2	3
Twist coefficient C (%)		0.33	0.60	1.25	1.88	0.25	0.33	0.45	1.10	1.60	0.90	1.75	1.98
Chamber coeff. R (%)	Longitudial	0.55	0.75	1.38	1.75	0.27	0.36	0.40	0.55	0.75	0.55	0.88	1.68
	Transferze	0.33	0.68	1.10	1.55	0.45	0.56	0.60	1.15	1.30	0.88	1.30	1.80

Table 3. Regularity of products shape

Raw material	A				B					C		
Experiment	1	2	3	4	1	2	3	4	5	1	2	3
DSFS* (N)	0.85	0.80	0.74	0.72	1.15	1.09	1.02	0.95	0.85	0.75	0.73	0.70
FSFS** (N)	1.44	1.35	1.33	1.26	1.65	1.58	1.55	1.49	1.44	1.39	1.30	1.29

* Dried samples flexural strength; **Fired samples flexural strength

It can be seen that twist and chamber coefficients is rising as the temperature is increased. The highest values are registered in the case of raw material C. The flexural strength is slowly decreasing with the increase of temperature for all raw materials. These results are expected and are in line with the registered raw material composition and detected drying sensitivity.

4. CONCLUSIONS

The novel method for evaluation of the clays suitability for the rapid drying application was reported in this paper. Since the intrinsic nature of the raw material is incorporated into the unique line it was seen as the perfect candidate for comparison of various clay raw materials. The lowest value of the unique line slope was found in the case of raw material C while the highest value was registered for the raw material B. The mineral composition of these two raw materials was closest to the ideal situation, in which only kaolinite or montmoriolinite were present alone. That's why the slope of these lines is on the opposite side of the proposed drying sensitivity scale. In other words, the limiting maximal and minimal slope values are defined in this paper. Since boundary values are known the estimation procedure is simple and comes down to determination of the unique line of the material which drying sensitivity is unknown. This means that only two isothermal experiments are theoretically necessary for determination of the drying sensitivity in accordance with the proposed model.

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