A PRELIMINARY ASSESEMENT OF MICROWAVE BASED DRYING FEASABILITY USING THERMOGRAVIMETRY ANALYSIS

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ABSTRACT

Due to the climate changes, the processing of refractory materials is under fine scrutiny to find out opportunities to increase its efficiency and reduce direct and indirect emissions. The drying of monolithics is a complex subject where long drying schedules are used as the main strategy to mitigate the risks of explosive spalling. In various applications such as wood and food drying, microwaves have been used to increase efficiency and reduce induced damage to the dried materials. The current work aims to propose a preliminary but innovative, experimental comparison of the refractory castables drying under distinct heating sources, namely conventional (electrical resistance) and microwave heating, using a novel adapted thermogravimetry setup followed by numerical methods. Estimations of energy consumption and emissions are also presented to assess the feasibility of microwave as the main source for drying refractory castables.

1 INTRODUCTION

Monolithic refractories are mostly cast by the consumer at the working place[1]. These materials present several advantages when compared to the shaped ones, such as ease of installation, lack of expansion joints and the possibility of automate their application[1, 2]. Despite these features, the complex rheological control required to their conformation, generally involves the addition of a liquid phase, which for hydraulic bonded refractories is water. For drying of these products care must be taken to avoid, cracks, spalling, or even explosions, in more extreme cases. This phenomenon known as "explosive spalling" is still not fully understood[3], however, it is currently accepted that it occurs due to a combination of two mechanisms, the thermomechanical stresses and the pressurization of water vapor, which results in a sudden release of the stored elastic energy[3].

The most common strategy to avoid the damage effects is using long heating protocols[4]. This approach is based in semi-empirical relationships and poses a great challenge to the efforts to decrease the energy consumption and, consequently, the carbon footprint of refractory castables, as the longer time of the slow heating schedules lead to greater emissions. Additionally, the productivity halt decreases the profitability of the transformation process where the refractory is applied. Thus, recently studies focused on how to properly model this phenomenon to design safe and efficient heat up curves[5, 6], as well as increase the understanding of additives such as polymeric fibers[7], and even the drying itself by using direct imaging techniques[8].

The current work proposes a new development front, based on the promising results suggested by Nippon Steel[9] and Gong et al.[10], on considering using microwaves for the drying of refractory castables. This study differs from the ones aforementioned by focusing on the thoroughly understanding of the differences between conventional and microwave heating, via and adapted novel thermogravimetry equipment and numerical simulations. Finally, energy consumption estimative are also considered to understand whether the increased complexity and investment costs could overcome the likely advantages of shifting from conventional to microwave drying.

2 MATERIALS AND METHODS

2.1 Microwave Thermogravimetry Equipment

The principle of working of thermogravimetry analysis (TGA) is based on the weight variation of a given sample with the increase of temperature[4]. Fundamentally, this concept can be applied with distinct heating methods and due to cost effectiveness and reproducibility, the use of electric resistance furnaces is more usual. In the present work a TGA setup is based on a microwave oven, which leads to a notoriously different heating process.

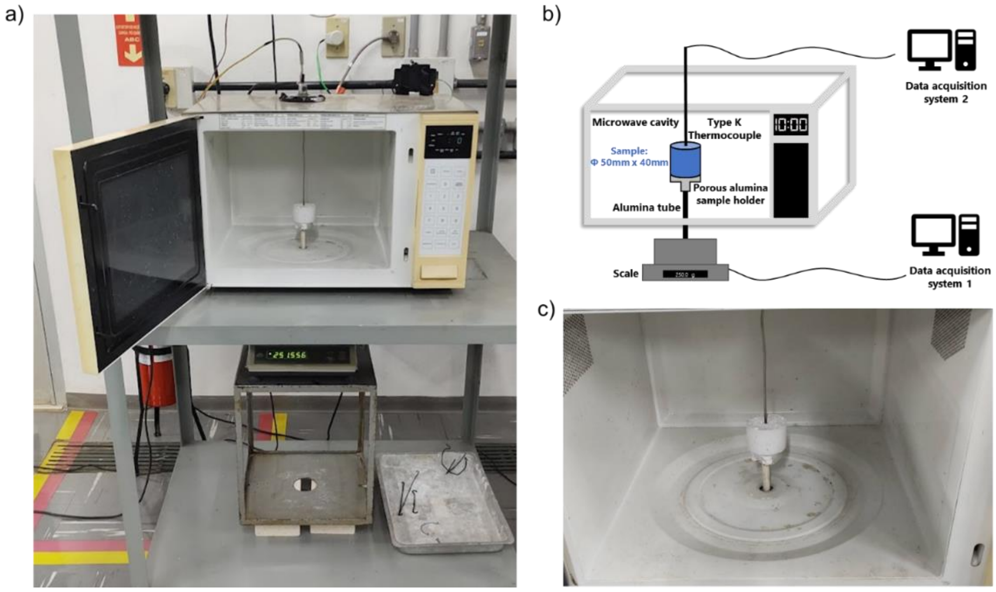
Given the larger wavelengths of microwave radiation (around 12.2 cm for the 2.45 GHz frequency used for kitchen ovens and Wi-Fi signals), its photons are less energetic than thermal radiation, and thus, can penetrate deeper in semi-opaque materials[11]. Thus, a volumetric heating can occur and due to thermal losses to the cold environment surrounding the sample, the innermost positions are hotter than the surface of the material[11]. Additionally, different molecules have distinct absorption behavior, ranging from reflecting microwaves (such as metals) to absorbing it and increasing its temperature (such as silicon carbide)[12]. Thus, in the case of partially saturated porous medium, microwaves can be of great interest as the regions in the sample that will heat the most are those containing water[11].

Based on that, the current setup comprises a system capable of carrying out TGA analysis on samples with the same sizes of the conventional TGA (CTGA) results. This system is based on a scale placed under the microwave, an alumina tube used to sustain the sample and a thermocouple, which plays a critical role in the equipment.

Due to the nature of microwaves, the air surrounding the sample is transparent and don't have its temperature increased. Thus, opposed to the conventional TGA where one can define a furnace and sample temperature (which are considerably different), for the microwave TGA (MWTGA) the heating schedule is followed directly by the sample. Besides the complexities of measuring the temperature without affecting the mass values obtained by the scale, the greatest challenge is on the thermocouple itself. Being a metallic material that reflects the microwaves, the latter can be leaked and also generate sparks or even sustained plasma[13]. To solve this problem the thermocouple used was grounded and a rubber piece was placed to support it on the top of the microwave equipment. Additionally, two independent computers are used as a strategy to decrease signal loss due to electromagnetic interference, as described in Figure 1.

To sustain the long drying curves used in this work, auxiliary fans were adapted to the microwave equipment to avoid excessive heating which triggers the thermal protection system and shuts down the wave generator (i.e. the magnetron). Finally, for thermal insulation, the support between the alumina tube and the sample is made of porous alumina insulating material, limiting the temperatures of the alumina tube to only 40°C – 60°C at the final temperature of 625°C.

Figure 1: Experimental layout of the microwave TGA equipment, a) photograph of the system, b) schematics of the components and c) detail of the cavity and sample inside of it.



For the tests in the current work, a high alumina castable with 5 wt.% of calcium aluminate cement and a packing distribution coefficient q = 0.21, following Andreasen's particle model, was prepared following the methods described by Cunha et al[6]. The samples are molded as ⌀ 50mm x 40mm cylinders with a hole with ⌀ 6mm x 20mm at its center for positioning the thermocouple. The composition is described in Table 1.

Table 1: 5CAC High-alumina refractory castable composition.

Raw Materials		Composition (wt.%)
Tabular Alumina (D<6mm)	Almatis	74
Calcined and reactive alumina	CL370, Almatis	11
	CT3000SG, Almatis	10
Calcium Aluminate Cement	Secar 71, Imerys	5
Distilled Water		4.5
Dispersant	Castament FS60, Basf	0.2

After 24h of curing at room temperature inside closed plastic bags with a water containing beaker, the samples were stored and analyzed by both the microwave and conventional TGA systems using a 5°C/min heating rate from room temperature up to 625°C with a dwell time of 20 minutes. The mass loss was normalized by the initial weight of the sample and its derivate with respect to the sample's temperature was obtained via finite differences approximation.

2.2 Numerical Simulations

The simulations were based on the single-phase model proposed by Bažant et al.[3] and the microwave heating followed the description proposed by Gong et al.[10]. The mass conservation and thermal energy balance are described by Equations 1 and 2, respectively

$$\frac{\partial \rho_{ev}}{\partial t} = \nabla \cdot \left(\frac{\kappa}{g} \nabla P_v\right) + \frac{\partial \rho_{deh}}{\partial t} \tag{1}$$

where ρ_{ev} is the mass of free water per kilogram of concrete, t the time, κ the intrinsic permeability, g the acceleration of gravity (9.81 m/s²), ρ_{deh} the mass of chemically adsorbed water released and P_v is the pressure inside the pores, the primary variable of the model.

$$\frac{\partial T}{\partial t} \rho C_p = \nabla \cdot (\lambda \nabla T) - C_l \frac{\kappa}{g} \nabla P_v \cdot \nabla T + \Delta H_{ev} \frac{\partial \rho_{ev}}{\partial t} + \Delta H_{deh} \frac{\partial \rho_{deh}}{\partial t} + Q_{MW} \tag{2}$$

where ρ is the density of the refractory castable, C_p is its specific heat, λ is its thermal conductivity, C_l the specific heat of liquid water, ΔH_{ev} and ΔH_{deh} the enthalpy of evaporation and dehydration, respectively, and T the temperature, the primary variable. Q_{MW} is ignored when considering the conventional TGA case and for the microwave one, it is given by Equation 3.

$$Q_{MW} = P(A_w \rho_{ev} + A_c \rho) \tag{3}$$

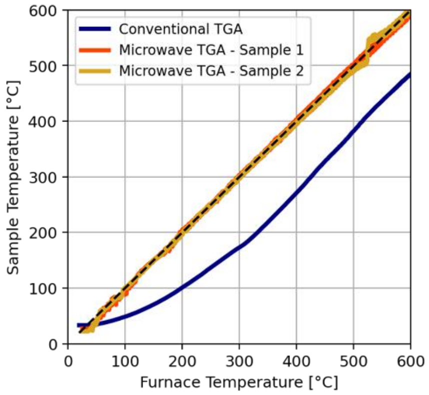
where A_w and A_c are the coefficient of microwave absorption of water and castable respectively, and based on Gong et al.[14], they were assumed to be equal to 1040 J/(Kg s) and 13 J/(Kg s). P is the power of the microwave defined by the PID controller to follow the defined heating protocol. For more details on the implementation of the model, the properties used to simulate the 5CAC composition and its numerical solution, the reader is referenced to Cunha et al.[6].

3 RESULTS AND DISCUSSION

3.1 Microwave Thermogravimetry

The difference between the furnace temperature and the value at the center of the sample is an estimative of the thermal gradient observed in the sample. Figure 2 shows such difference for the conventional TGA. The values for the microwave TGA are also presented, however as the surrounding air inside the microwave cavity is not heated, the furnace temperature is actually the programmed values (hence the quotation marks). It should be noted that this not mean that the thermal gradient in the sample during MWTGA is null, especially when considering that the surfaces of the sample will be losing heat to the cold air. It was even observed after the end of the MWTGA test that the thermocouple hole glowed while the rest of the sample was still opaque.

Figure 2: Furnace temperature (set temperature scheduled for the microwave TGA) and sample temperature profile when heated at 5°C/min.



When considering the mass loss, Figure 3 (a) shows that the overall behavior was similar for the conventional and microwave TGA, leading to a total mass loss of around 4.2 wt.%. It is possible to observe that the CTGA sample wasn't able to reach the final temperature of 625°C.

Figure 3: (a) Mass loss of the sample for the conventional TGA and Microwave TGA, (b) derivative of mass loss with respect to the sample's temperature.

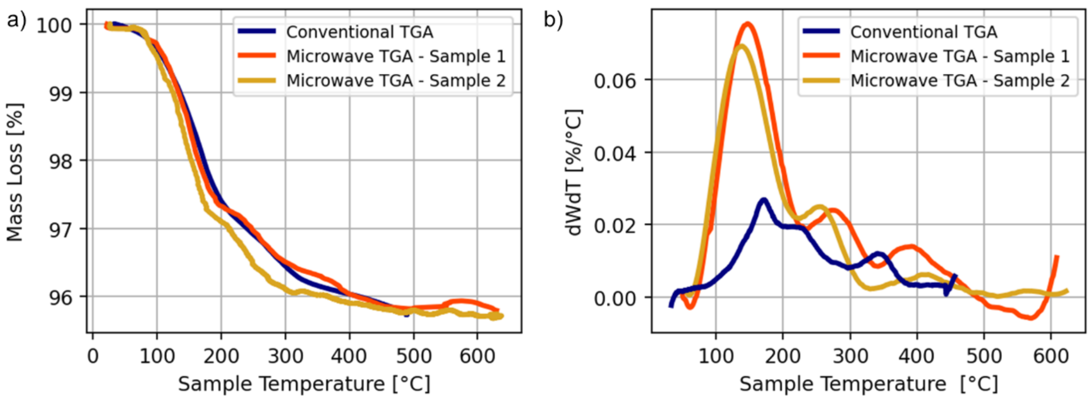
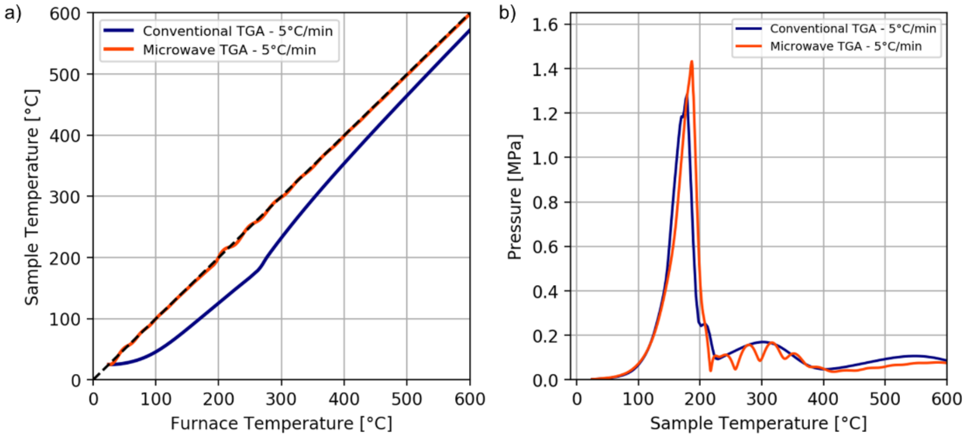


Figure 3 (b), which presents the derivative of the mass loss with respect to the sample temperature highlights that the first mass release peak occurs almost 30°C earlier for the MWTGA when compared to the CTGA. Additionally, the first dehydration peak is convoluted with the free water for the conventional drying, whereas it is clearly separated for the microwave TGA. This shows that modifying the temperature gradients in the sample by changing the heating method can impact the water removal dynamics. To further investigate these aspects, numerical simulations are considered next.

3.2 Numerical Simulations

The numerical results depicted in Figure 4 (a) suggest some temperature difference between the furnace and the sample similar to the one observed experimentally in Figure 2, due to the different boundary conditions. The PID controlled temperature of the MWTGA presents a larger oscillation around 200°C, due to the free water removal.

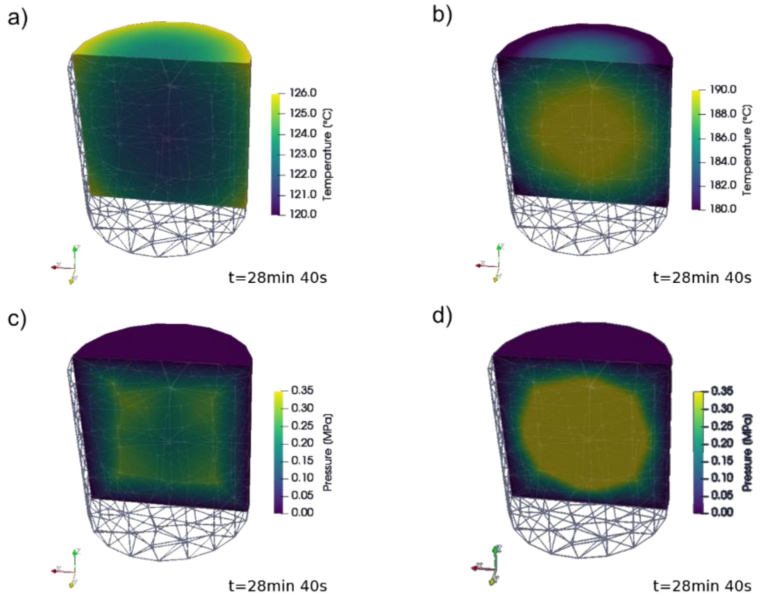
Figure 4: (a) Furnace temperature (set temperature scheduled for the microwave TGA) and the sample temperature profile predicted when heated at 5°C/min and (b) pressure evolution at the center of the sample.



Regarding the pressure evolution on the microwave TGA, a peak 11% higher than the conventional one can be seen. That can be related to the real heating rate of 5°C/min that is directly applied to the center of the sample by the microwave heating. As in the case of the mass loss (Figure 3), the separation of the free water withdrawal and the dehydration one can be seen in the case of MWTGA, whereas for the CTGA, a shoulder appears around 215°C. Another distinct feature of the microwave case is the effect of the PID oscillations on the pressure peaks related to dehydration. This highlights an interesting potential of MW heating, as the heating protocols are not limited to constant positives heating rates and plateaus, but it can be precisely controlled including regimes of momentary cooling.

Finally, Figure 5 shows the temperature and pressure distribution for the conventional and microwave TGA. It is clear that the conventional heating has a negative thermal gradient from the surface towards the innermost positions as seen in Figure 5 (a), whereas the exactly opposite is seen for the microwave case, Figure 5 (b). Also, at that time, the MWTGA sample is roughly 60°C hotter than the CTGA. The pressure distribution on both samples is similar with higher values at the center of the sample. Due to the higher temperature at this specific time, the microwave case shows higher pressure.

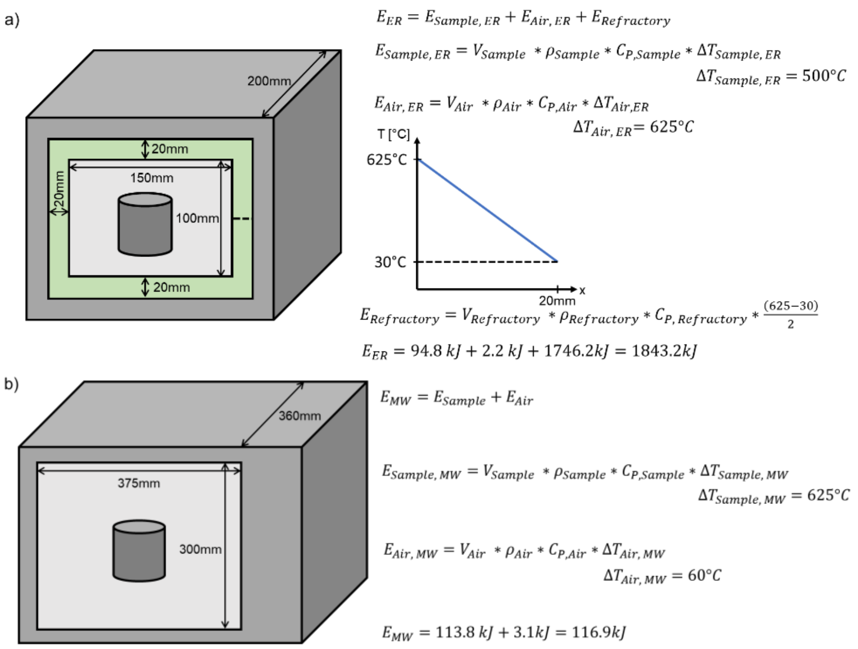
Figure 5: Temperature and pressure distribution at 28 minutes and 40 seconds of the test for conventional TGA, (a) and (c) and for microwave TGA (b) and (d), respectively.



3.3 Energy consumption

This last section presents the estimation of energy consumption for the two thermogravimetry methods. The TGA equipment geometry as well as the main hypothesis and the calculations can be seen in Figure 6.

Figure 6: Estimation of energy consumption of a) electric heating and b) microwave TGA equipment. The refractory assumed in the electric furnace was a vermiculite plate with density of 2300kg/m³ and a specific heat of 1100 J/(kg K).



The energy consumed to heat the refractory walls in the conventional case is the main responsible for the almost 15 times higher total consumption. This shows the potential for higher efficiency of microwaves for drying of pre-shaped products. When considering the CO_2 emissions of such methods a simplified estimative considering EPA Greenhouse equivalence calculator[14] shows that the CTGA would result in 0.221 kg of CO_2, versus 0.014 kg of CO_2 for the MWTGA.

When considering the consumption of energy for the drying of real refractory castable linings, other important aspects should be considered. In conventional drying, natural gas is burned to heat up the refractory. Using microwaves for that propose could decrease the total consumption at this stage (even if at higher temperatures the natural gas was still used), especially when considering that clean energy electricity could be used to power the magnetrons.

4 CONCLUSIONS

The drying of refractory castables is a burdensome stage in the processing of this class of materials. The likelihood of explosive spalling leads to long and less efficient heat-up protocols that decrease productivity and waste a lot of energy. Microwaves interact in a different way than conventional heating, inducing a volumetric increase of temperature. This can promote changes on the mass release as seen from both thermogravimetry tests and numerical simulations. Such results points towards promising alternatives specially when considering that for the specific case of conventional and microwave TGA, an energy consumption decrease of 93% was observed. Further understanding on how to properly conduct microwave drying is still required, as well as more information on how such a different heating method can impact the real-world consumption of energy and carbon dioxide emissions. An important tool was set (MWTGA) which will certainly help to find out various novelties and innovations.

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