DRYING OF REFRACTORY CASTABLES – HOW COMPLEX THE MODELLING NEEDS TO BE? A NUMERICAL AND EXPERIMENTAL STUDY

M. H. Moreira[(1)]*, S. Dal Pont[(2)], R. F. Ausas[(3)], T. M. Cunha[(1)], A. P. Luz*, [(1)], V. C. Pandolfelli[(1)]

[(1)] Federal University of Sao Carlos, Graduate Program in Materials Science and Engineering, Sao Carlos, SP, Brazil

[(2)] CNRS, Grenoble INP, 3SR, Université Grenoble Alpes, 38000 Grenoble, France

[(3)] Institute of Mathematical and Computer Sciences, University of São Paulo, 13566-590, São Carlos, SP, Brazil

ABSTRACT

Calcium aluminate cement (CAC)-bonded castables have numerous benefits when compared to monolithic refractories. Their main drawback is their long initial heating, when the physical and chemically-bonded water are released. If the mass flux of water to the ambient is lower than the vapor generation, the gas pressure can increase well above the material's strength resulting in cracks and spalling of the ceramic lining. In order to avoid this, optimized heat up curves are needed, and one promising methodology by using numerical models. Most of these calculations are based on tools used for simulating Portland cement concrete structures on fire and they vary both in complexity and their basic assumptions. Thus, the main question that remains is how complex such models need to be in order to capture the fundamental aspects of this phenomenon. The present work aimed to solve this issue by implementing multiple numerical tools reported in the literature with distinct complexity levels and fundamental assumptions and by also conducting neutron tomography tests on a CAC-bonded castable. It was possible to see that the pressure values predicted by such methodologies are equivalent and the water content predicted is qualitatively similar within the models and with the experimental values, indicating that the simplest approach might be used for predicting the castables' drying.

INTRODUCTION

Refractory monolithics were developed as an option to shaped products, as they offer several distinct features such as a greater degree of freedom to adjust and install the composition and, as a consequence, a more detailed control over the properties of the final product. These characteristics lead to several benefits and, finally, over time it has been one of the most used classes of refractories[1].

Aiming to increase the service time and performance of these linings, the design of monolithic compositions is usually based on the particles' packing optimization allied to the used of additives, that control the rheological behavior of the matrix components due to modifications in the surface chemistry of the finer particle size fractions[2]. As a result, important improvements in the castables' properties could be obtained, such as higher

mechanical strength, lower porosity and also higher resistance to corrosion.

As a side effect, the well packed microstructures led to materials with reduced permeability, which during the initial stages of the lining preparation, i.e., the drying step, can pose great technical challenges for hydraulic bonded castables.

Such fact can be better understood when analyzing how the refractories' drying process takes place. Due to great capillary pressures, drying at room temperature is practically impossible, even with forced convection[3, 4]. Thus, the common practice is to split the first heating of the castable in two main steps: drying and firing.

The first one is the most critical, especially for compositions with low permeability. The benefits of using thermal gradients to enhance the mass transport can also be responsible for inducing the steam pressurization, and if this gas is not released to the external environment, the forces inside the material can surpass its mechanical strength, leading to cracks and ultimately to explosive spalling[4, 5].

This phenomenon is associated with two main processes: the moisture clog and thermomechanical stresses, as described in Fig. 1. The moisture clog is a local permeability decrease, usually identified ahead of the drying front, due to the condensation of liquid water on the cooler and deeper regions of the refractory structure. This can lead to a sudden pressurization that generates a tensile stress.

If this was the only force acting on the refractory, cracking around those regions would occur. However, in practice it is well known that such phenomenon is more violent and the pieces that are spalled are thrusted to several meters. Thus, the vapor pressure can only be seen as a trigger, which can rapidly release the potential energy from the thermomechanical stresses due to uneven heating of the refractory wall.

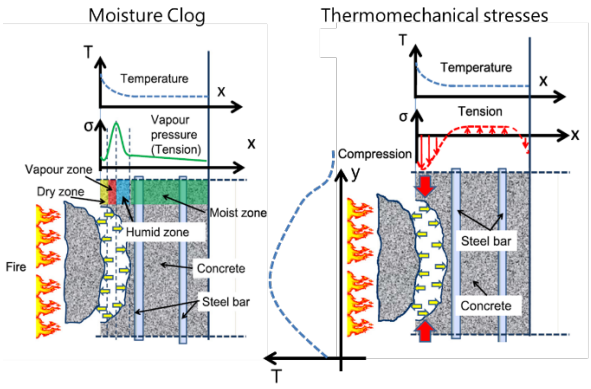

Fig. 1. The current understanding of the explosive spalling is based on the moisture clog event and thermomechanical stresses. Adapted from [6].

This problem is not exclusive to the drying of refractory materials. Several partially saturated porous media can suffer intense damage when subjected to high temperatures. In fact, most of the fundamental studies and proposed models for the evaluation of this process were originated from the civil engineering community as they are concerned by the fire resistance of common Portland cement concrete structures[3, 4].

The study of the refractories' drying is a challenging endeavor. Firstly, it is size dependent and most of the times, laboratory samples' scales are very different from the sizes found for castables' pieces designed for industrial applications. Also, the properties that affect the behavior of the materials during drying are difficult to measure. Finally, the drying is a multidisciplinary subject and demands a collaborative work from different specialists.

As a result, several numerical models were developed over time. They are based on distinct assumptions and can vary in complexity and ease of using. Fig. 2 summarizes the main categories that can be used to classify these numerical tools.

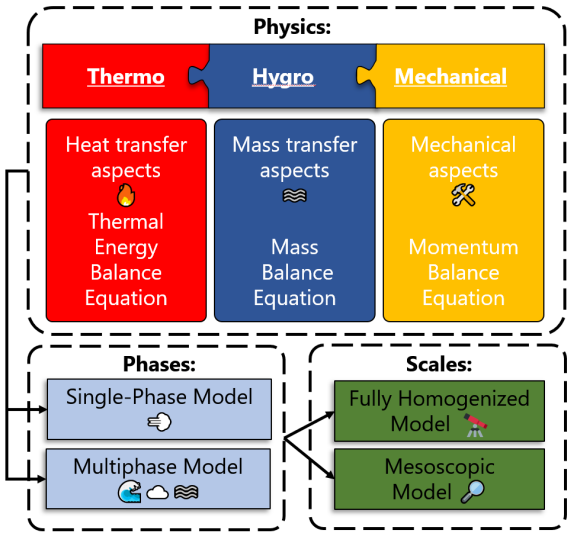

Fig. 2. Summary of classifications and categories of numerical models commonly used for studying the drying.

Depending on the physics that are considered, a numerical tool to predict the drying behavior can be classified as a thermal, thermohygral or thermohygromechanical model. As the complexity increases, so does the kind of responses that the simulation can offer. A straight thermal model can only describe the temperature distribution on the ceramic wall. However, this cannot be completely reliable to predict the generated steam pressure inside a dense structure, as even if one can, in theory, use the Antoine's law to compute the vapor pressure at a given moment and time. This latter would only indicate a maximum value. Furthermore, as the water vapor is not hermetically sealed, it can still escape from the material, leading to a pressure decrease[4].

On the other hand, the thermohygro models consider both the thermal energy balance and the mass transfer. This enables the prediction of the water distribution inside the material at a given time. Also, it is possible to obtain the pressure distribution.

Finally, the mechanical aspects can be considered in the thermohygromechanical models, which indicates wether the material was damaged or not for a given pressure values.

It should be noted that the coupling between the distinct physics can also be further classified into fully coupled models or semi-coupled (also known as one-way coupled) ones. This indicates if a given primary variable will only affect a single physics model or not (for instance, consider the coupling between the hygro and mechanical aspects – a one-way coupling would be to only consider the effect of the pressure on the stress whereas a fully coupled model would also consider the effect of the stress in the material's permeability)[4].

A second way to categorize a model is by the number of considered phases. The two main concurrent paradigms are the single-phase and multiphase models. It is argued that a single-phase model can lead to unrealistic predictions of the relative humidity[6], whereas a multiphase one would be too complex and rely on a high number of properties (that are so challenging to measure) that it would render it unfeasible.

Finally, the last way to organize the families of models is to consider the scales of each numerical tool. The majority of the published works are based on the homogenization techniques, which leads to fully homogenized models. This category does not consider explicitly the heterogeneities contained in the castables' microstructure. The second one is the mesoscopic models, which can consider such aspects[3, 4].

Thus, given the great number of distinct approaches to model the drying behavior of refractory castable, each one with distinct advantages and drawbacks, it is necessary to compare and consider which model satisfies the needs of a given application. To achieve this, the current work presents a comparison of three thermohygral fully homogenized models considering single and multiphase approaches.

MATERIALS AND METHODS

The current section will briefly introduce the models and the analysis that will be carried out. For a more in-depth presentation of the models the reader is referenced to the works of Moreira et al. [4,7].

The basic principle from which the models are derived from are the conservation equations from the Classical Irreversible Thermodynamics (CIT) [4], which states that the time derivative of a quantity plus the gradient of its flux is equal to any sources or sinks of this particular quantity present in the region of interest. Mathematically this is given by Equation 1.

$$\frac{\partial}{\partial t}(density) + \nabla \cdot (flux) = source \qquad (1)$$

Regarding the mass conservation, there are three options to consider: (i) the mass balance of moisture, a quantity that represents both the liquid water and vapor – the approach of the single-phase model (SP); (ii) the distinct phases individually inside the porous medium and neglect the capillary pressure that develops at the meniscus interface between the liquid and gas phases – the multiphase model that neglects the capillary pressure (NCP); (iii) the distinct phases and also the effect of the capillary pressure (CCP).

Each approach will yield an equation (or a set of equations, for the multiphase models) that describes the mass balance. Such considerations will also have an influence on the resulting thermal energy conservation equation (also broadly known as the heat transfer equation), as regardless of the assumption, the convection will introduce the mass flux effect on the heat transfer equation.

An important aspect is how one can model the flux of the moving phase on such approaches. For the SP model, the mass flux is given by Darcy's law, whereas in NCP and CCP, besides the Darcy's law, the diffusion of the molecules in the gas phase is also accounted. Finally, the NCP model also considers the movement of adsorbed water inside the material.

The resulting set of partial derivative equations are then numerically solved using the finite element method (FEM). The computer code elaborated in the present work is based on the FEniCS platform[8], a framework based in C++ and Python that is free and *open source.* The results are plotted also using Python.

Three distinct analyses will be presented: (i) 1D Portland cement concrete benchmark based on the pressure, temperature and mass (PTM) results by Kalifa et al. [9]; (ii) 2D Portland cement concrete benchmark based on the neutron tomography results by Dauti et al. [10]; and (iii) 2D Simulation of a high-alumina CAC-bonded castable (5CAC)[11] using the SP model and considering the data obtained via neutron tomography.

RESULTS AND DISCUSSION

Analysis (i) - 1D Portland Cement Concrete Benchmark

The first set of results is based on the simulation of a slab (30 x 30 x 12 cm^3) of Portland cement concrete, when it was heated from one of its sides and had its temperature and pressure profiles measured.

Fig. 3 shows the temperature evolution at different depths of the sample. It is observed that the three evaluated models are all equivalent, with minor deviations on their thermal prediction. It may also be pointed out that the experimental values are in tune with the overall trend of presenting lower temperatures at the innermost positions of the sample.

This is an important result as it shows that the influence of the different assumptions of the models have a low impact on the thermal prediction.

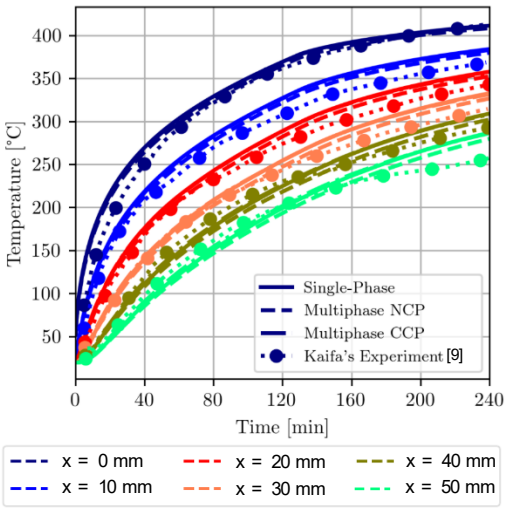

Fig. 3. Temperature increase calculated by the models and also measured by Kalifa et al.[9].

Next, Fig 4. presents the gas pressure evolution at different positions inside the samples, which was calculated by different numerical approaches. Additionally, a comparison of the simulations with the experimental values are also provided.

Overall, all three models were able to predict the moment of the pressurization event. However, the data derived from SP and CCP models led to pressure peak results that were closer to the experimental ones. However, NCP model underpredicted the pressure values and also displayed a distinct change in the inclination of the pressurization curve.

Another important aspect is that all the models were not able to capture the pressure decay that follows the peak value. That is related to the fact that the models only consider thermal and hygral physics (i.e., all the models are thermohygro), without taking into account the mechanical damage effects, which could increase permeability and reduce the obtained pressure values [7, 10].

It should be noted that the great correspondence between the CCP model and the experimental values are also related to the fact that the input parameters, used in all three models, were tuned by Dauti et al[10].

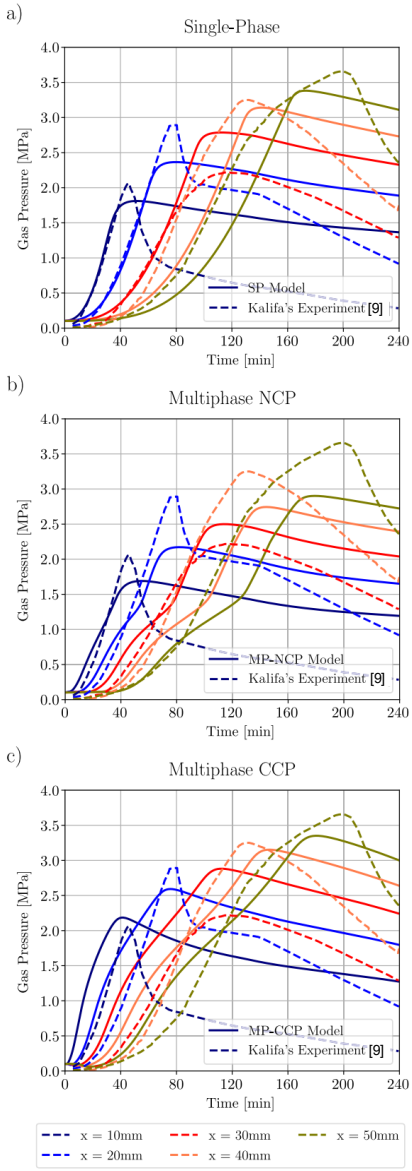

Fig. 4. Pressurization prediction by (a) the single-phase model (SP), (b) the multiphase model that neglects the capillary pressure (NCP) and (c) the multiphase one that considers the capillary effects.

Analysis (ii) - 2D Portland Cement Concrete Benchmark

The PTM (pressure, temperature and mass) experiments are widely applied as a validation test for the drying numerical models. However, they are prone to some effects of the

pressure and temperature sensors themselves that are embedded in the samples' microstructure [10].

As an alternative, direct imaging techniques, such as neutron tomography, are suited to monitor the moisture content distribution during the concrete's samples heating process and they are also representative of the behavior of the material without any sensors embedded into it. The only drawback is that the pressure is not measured. Thus, next, a comparison of experimental data with the models' results will be based on the variations of the water content of Portland cement concrete samples.

The evaluated sample is a cylinder with 30 mm in diameter and 60 mm in height, and it was heated by an infrared radiator source that was placed close to the top surface of the sample during the neutron tomography measurements.

Fig. 5 shows the results of the water content variation of the three evaluated models and the ones obtained experimentally. It was observed that the simulations predicted similar drying front velocities (i.e., the position where there is a sudden variation of the water content) as the one detected in the neutron tomography.

Some distinct features of the model's predictions could also be observed, such as a higher lateral drying for the CCP – probably related to the capillary effects, which is not observed in the NCP and SP models. Finally, the drying of the SP model was more intense on the top bottom of the sample

It is, however, clear that all three models qualitatively agree on the moisture distribution content, and depending on the level of detail that one aims to achieve, a simpler model can be safely used to predict the pressure values, at least to a first degree.

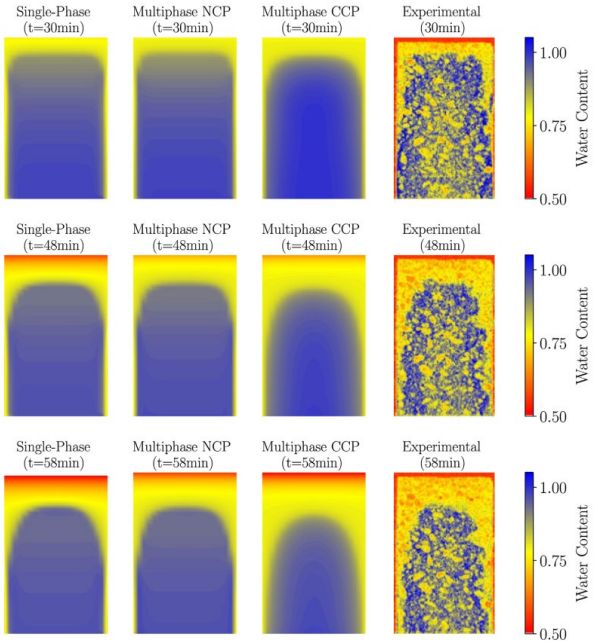

Fig. 5. Water distribution evolution calculated with the models and obtained in the neutron tomography test, reported by Dauti et al[10].

Analysis (iii) - Simulation of the 5CAC castable and comparison with Neutron Tomography test results

The last set of analysis is based on the simulation of a 5 wt.% CAC-containing castable composition (for details on the particle size distribution and the properties used, the reader is referenced to the work by Cunha et al[11]) using the SP model and the comparison of the results with neutron tomography data.

Two distinct tests using the same conditions from Dauti et al[10] were performed: (i) with a ceramic casing around the sample, to prevent any mass flux in the radial direction and provide a unidirectional drying, and (ii) without any casing.

Fig. 6 presents the results of water content for the sample with the ceramic casing. It is observed that the drying front velocity is closely predicted by the model, considering both the principal front moving from the top to the bottom surface, as well as the secondary front that develops on the cold surface.

It should be noticed, however, that the water accumulation (where the water content is higher than one) that occurs at the innermost positions are not predicted by the single-phase model. This is an important feature verified during the drying, which is also known as the *moisture clog*.

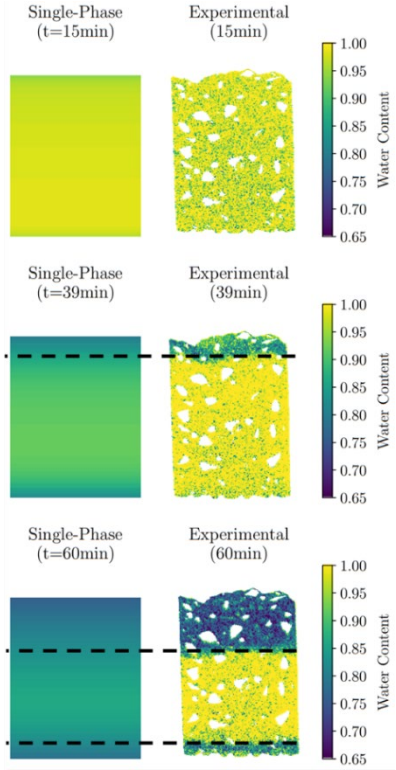

Fig. 6. Water distribution evolution calculated with the SP model and the one obtained by the neutron tomography test with ceramic casing.

However, it is important to notice that the sorption isotherm (i.e., the state function that defines the physically adsorbed water inside the material) for the 5CAC composition was not measured. Also, the boundary conditions could be further enhanced to better fit the experiments.

Following, Fig. 7 shows the analysis of the sample with no casing. It is possible to see that once again the drying front velocity was well captured and even its blunt shape was reproduced by the model. Also, as in the previous case, the water accumulation was underestimated by the numerical simulation.

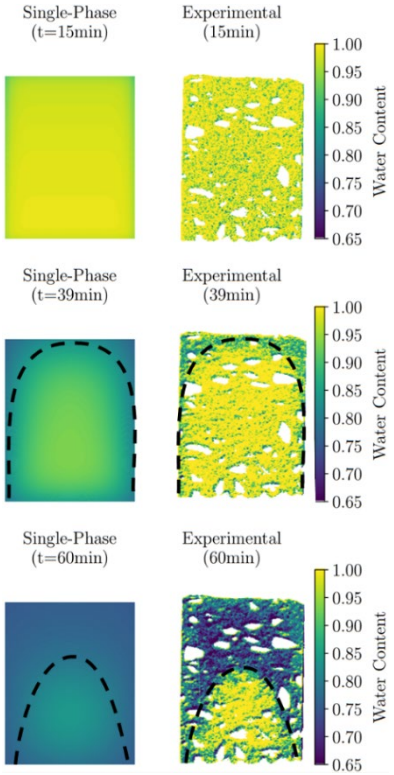

Fig. 7. Water distribution evolution calculated with the SP model and the ones obtained in the neutron tomography test for a sample without the ceramic casing.

However, in this specific configuration, the radial mass transport can result in the *beam hardening effect*, which is an artificial increase on the water content at the innermost positions due to the screening of more energetic neutron rays. Consequently, the difference between the water content in the center of the sample, for the caseless condition could be even smaller when comparing the numerical results and the real values.

The main outcome of these last two qualitative comparisons is that even for very different conditions (with and without the ceramic casing) the simplest SP model is still capable of representing the main features of the drying phenomena.

CONCLUSIONS

There are several numerical methods that aim to accurately predict the behavior of

castables during drying. Most debates over the adoption of one methodology were mainly theoretical, and thus, the current work aimed to directly compare the results derived from those common approaches with experimental data.

By using different experimental results, it was verified that the most complex multiphase model that considered the capillary pressure (CCP) could estimate the pressure peaks values, their time and also the moisture accumulation ahead of the drying zone.

Meanwhile, the model that neglected the capillary pressure underestimated the pressure levels and were not able to highlight the details of the moisture distribution.

Finally, the single-phase model was the simplest approach, and still it was able to capture the main aspects of the drying, such as the pressure levels and the drying front velocity. One observed drawback is that this model could not reproduce the moisture clog effect, neither for the Portland cement nor the 5CAC composition. On the other hand, it was able to qualitative show the effect of using of a ceramic casing, showing that for technological applications, this simplest approach can aid to optimize the drying of refractories.

Further studies focused on more precise measurements of the 5CAC castable properties and the temperature distribution inside the sample during the drying process could also result in better agreements between the theoretical predictions and experimental observations. Finally, the main outcome is that the best model for the drying is based on the necessities of the end-user and each model has advantages and specific drawbacks

ACKNOWLEDGMENTS

This study was financed in part by the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) - Finance Code 001. The authors would like to thank the Fundação de Amparo à Pesquisa do Estado de São Paulo - FAPESP (grant number: 2021/00251-0 and 2019/07996-0), Finally, the authors are greatly thankful for FIRE support to carry out this work.

REFERENCES

1. C. Schacht, Refractories Handbook, Mechanical Engineering, CRC Press (2004)
2. A.P. da Luz, M.A.L. Braulio, V.C. Pandolfelli, Refractory Castable Engineering, vol. 756, Goller Verlag GmbH, (2015)
3. Z.P. Bažant, M. Jirásek, Creep and Hygrothermal Effects in Concrete Structures, [225], Springer, The Netherlands (2018)
4. Moreira, M. et al. Main trends on the simulation of the drying of refractory castables - Review. Ceramics International, Elsevier, vol. 47 [20] pp. 28086-28105 (2021)
5. G. Palmer, et al. The accelerated drying of refractory concrete – Part 1: a review of current understanding, Refractories Worldforum 6 [2] pp 75–83 (2014)
6. D. Gawin, et al., What physical phenomena can be neglected when modelling concrete at high temperature? A comparative study. Part 2: comparison between models, Int. J. Solid Struct. 48 [13] pp 1945–1961 (2011)
7. Moreira, M., et al., Direct comparison of multi and single-phase models depicting the drying process of refractory castables. Open Ceramics, Elsevier, [6], pp. 100111, (2021)
8. M. S. Alnaes, et al., The FEniCS Project Version 1.5, Arc. of Num. Software [3] (2015)
9. P. Kalifa, et al., Spalling and pore pressure in HPC at high temperatures, Cement Concr. Res. 30 [12] pp 1915–1927 (2000)
10. D. Dauti, et al., Modeling of 3D moisture distribution in heated concrete: from continuum towards mesoscopic approach, Int. J. Heat Mass Tran. [134] pp 1137–1152 (2019)
11. Cunha, T. et al., Drying behavior of steel-ladle lining refractory castables under continuous heating rate. Ceramics International, Elsevier, vol. 48 [1] pp. 1142-1151, (2022)