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The pressure–decay technique for air permeability evaluation of dense refractory ceramics

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Abstract

The development of high-quality ceramic materials depends on the reliable evaluation of their structural, mechanical, thermal and fluid dynamic properties. However, particularly in the case of dense materials, the experimental evaluation of air permeability parameters involves much work and reliable measurements of high pressure and extremely low flow rate values. The objective of this study was to optimize the analytical routine of a new and promising permeametry technique, whose main advantages are its easy operation and fully electronic data acquisition. Tests carried out on a high-alumina ultra-low cement refractory castable sample showed that permeability constants k_1 and k_2 fitted reliably to Forchheimer's equation only when the original data set was screened at pressure intervals of 0.2 bar. It was also observed that the broader the pressure-drop range used for fitting purposes, the more reliable the resulting k_1 and k_2 values, even though permeability constants obtained by different techniques were best compared in similar pressure-drop ranges.

Keywords: Permeability; Characterization; Concrete

1. Introduction

Permeability parameters are particularly useful to evaluate the filtering performance of ceramic foams [1,2], the susceptibility of castables to mechanical failure during heat treatment [3,4] and the infiltration rate of corrosive liquids and gases in high-temperature processes [5].

The permeability of a given material is usually well represented by constants fitted according to a suitable fluid dynamic model. Innocentini et al. [5–9] have shown that Forchheimer's equation is particularly reliable for ceramic materials and can be expressed for airflow as [4]

$$\frac{P_i^2 - P_o^2}{2P_oL} = \left(\frac{\mu}{k_1}\right)v_s + \left(\frac{\rho}{k_2}\right)v_s^2 \quad (1)$$

where P_i and P_o are, respectively, the absolute pressures at the sample's inlet and outlet surfaces exposed to airflow; v_s is the superficial air velocity; L is the sample thickness; μ is the air viscosity and ρ is the air density calculated for P_o at the testing temperature (in this work, $T_o = 25$ °C and $P_o = 690$ mm Hg).

The terms k_1 and k_2, known as Darcian and non-Darcian permeability constants, are properties theoretically only of the porous medium and reflect, respectively, the influence of viscous and inertial interactions between fluid and the porous medium.

These constants are normally obtained by a traditional experimental procedure, where each measurement of air velocity (v_s) and air pressure (P_i and P_o) is carried out in a steady-state flow regime, after which a set of these values is fitted into Forchheimer's equation using the least-squares method. Although this procedure produces reliable permeability constants for ceramic materials, it is highly time consuming because data are manually acquired and sensitive equipment is often required to measure the very low air velocities and high pressures through dense samples.

For this reason, a new testing method, known as the pressure–decay technique, has been developed with the purpose of facilitating and speeding up the permeability evaluation of dense materials. This technique minimizes manual operations during data acquisition, reducing the total

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testing time and the number of parameter measurements that are required in steady-state permeametry.

Testing by the pressure–decay technique described by Innocentini et al. [9] is carried out using the same device applied in steady-state permeametry, with the sample fixed between two chambers. The inlet chamber is pressurized to a preset value, P_i, while the outlet chamber is exposed to the atmosphere (at constant pressure, P_o). At a given moment, the air supply is interrupted and the P_i pressure gradually decays as the air trapped in the inlet chamber flows through the sample to the atmosphere. The flow continues until P_i equals P_o, when $v_s = 0$. The only variable measured in this test is P_i, which is continuously recorded by computer as a function of time. The instant air velocity is not measured during the test, but can be estimated for each corresponding P_i value by [9]

$$v_s = -\left(\frac{V_c}{P_o A}\right)\frac{dP_i}{dt} \quad (2)$$

where V_c is the volume of the inlet chamber, A is the sample area perpendicularly exposed to the airflow and dP_i/dt is the derivative of the P_i-versus-time curve.

Analytical integration of Eqs. (1) and (2) with the initial conditions, when $t = t_o$ then $P_i = P_{io}$, directly gives the pressure dependence relative to time. Nevertheless, such a solution is very complex for the purpose of obtaining constants k_1 and k_2 through the fitting of experimental data [9]. On the other hand, several simplifications have already been proposed in the literature, as that described by Canon and Sander [4] who replaced Forchheimer's equation by Darcy's law as the permeability model adopted. Although in their case an analytical simple integrated solution was obtained, it did not represent physically the system, since Darcy's law does not suitably describe the permeability of dense refractory ceramics [5–7]. Similarly, the attempts of empirically fitting the pressure–decay curve prior to derivation/integration have also failed, mainly due to the lack of physical meaning of the fitting models.

For these reasons, the method usually adopted is the numerical derivation of Eq. (2) and its coupling with Eq. (1). This derivative can be obtained for each experimental data point j, according to the expression

$$\left(\frac{dP_i}{dt}\right)_j = \frac{P_{j+1} - P_{j-1}}{t_{j+1} - t_{j-1}} \quad (3)$$

where P_{j+1} and P_{j-1} are, respectively, the P_i pressures immediately after (t_{j+1}) and before (t_{j-1}) point j, at which the derivative is calculated.

A set of v_s, P_i and P_o values can thus be obtained by treating the pressure–decay curve according to Eqs. (2) and (3) and inserting them into Forchheimer's equation (Eq. (1)) to fit the k_1 and k_2 permeability constants.

Despite the abovementioned advantages, the pressure–decay technique requires data to be suitably treated in order to generate reliable permeability constants. The largest data set does not necessarily yield the most reliable constants [8], since the quality of the final fitting is highly dependent on the numerical calculation of the derivative dP_i/dt. For this reason, the lack of a suitable data analysis procedure has prevented a more effective use of the pressure–decay technique to evaluate the permeability constants of dense ceramic materials.

In this context, the purpose of this work was to propose an ideal analytical routine to treat the original data set generated by the pressure–decay technique, and thus produce reliable k_1 and k_2 values. A specific investigation was made of the minimum data set size and the optimum interval between consecutive data points required for a reliable analysis.

2. Experimental procedure

2.1. Sample preparation

High-alumina ultra-low refractory castable samples were prepared according to Andreasen's model for particle size distribution ($q = 0.26$), whose composition consisted of a mixture of a fine matrix (18.8 wt.%, $d_p < 100$ μm), coarse aggregates (79.2 wt.%, $d_p < 5.6$ mm) and high-alumina cement (2 wt.%). A 15-vol.% water content was added to the powders and aggregates during mixing. All the white fuse alumina used as aggregates, calcined alumina (A1000 SG, A3000 FL, APC3017 SG) and high-alumina cement (CA270) were supplied by Alcoa, Brazil and USA. The sample, a 2.2-cm-thick, 7.5-cm-diameter disk, was thermally treated at 1200 °C with a dwell time of 12 h.

2.2. Permeability tests

The sample under study was tightly fixed in a permeability device [7] between two chambers ($A = 1.257 \times 10^{-3}$ m^2 and $V_c = 5.62 \times 10^{-4}$ m^3). In the first test, 15 pairs of P_i and v_s values were manually acquired in a stationary flow regime and fed into Forchheimer's equation to fit k_1 and k_2 constants according to the least-squares method. The fluid properties used for the calculations were evaluated under ambient conditions ($\mu_{air} = 1.83 \times 10^{-5}$ Pa s and $\rho_{air} = 1.08$ kg/m^3).

The sample was then subjected to a pressure–decay test. The inlet chamber was pressurized and the air supply was interrupted at $P_i = 0.51$ MPa (gauge pressure of 5 bar). Air was allowed to flow through the sample into the outlet chamber, which was kept at a constant pressure of $P_o = 0.092$ MPa and at ambient temperature. The pressure P_i was measured with an electronic pressure transducer and recorded in a computer at 1-s intervals for 60 min. The data set collected in this test was then used for analysis as described below.

2.3. Analytical procedure

A computer routine (FORTRAN 90) was developed to screen the original pressure–decay data set based on two main parameters. The first concerned the interval between consecutive data points, which varied according to time (1–240 s) or pressure (0.05–0.5 bar) intervals. The second parameter involved the data range used for analysis, which varied according to the testing time (up to 60 min) or the pressure–decay range (up to 5 bar) employed during data acquisition. Each subset resulting from this procedure was individually treated to fit k_1 and k_2 constants, according to Eqs. (1)–(3).

In order to verify the influence of these screening parameters on the fitting quality of Forchheimer's equation, the deviation (σ) between k_1 and k_2 values resulting from each screened subset obtained by the pressure–decay technique and the values obtained by the steady-state technique was defined by

$$\sigma_{k_1}(\%) = 100\left|\frac{k_{1,\mathrm{pd}} - k_{1,\mathrm{ss}}}{k_{1,\mathrm{ss}}}\right| \quad (4)$$

$$\sigma_{k_2}(\%) = 100\left|\frac{k_{2,\mathrm{pd}} - k_{2,\mathrm{ss}}}{k_{2,\mathrm{ss}}}\right| \quad (5)$$

where $k_{1,\mathrm{pd}}$ and $k_{2,\mathrm{pd}}$ are the permeability constants obtained by the pressure–decay technique, and $k_{1,\mathrm{ss}}$ and $k_{2,\mathrm{ss}}$ are the constants obtained by the steady-state technique.

3. Results and discussion

Fig. 1a shows the original pressure–decay curve, with data collected every second during a 60-min test. As expected, the absolute pressure inside the chamber decreased gradually, following an exponential trend. Nevertheless, the resulting permeability curve shown in Fig. 1b displayed a high scattering level (correlation coefficient r^2=.3854), fitting very poorly to Forchheimer's equation compared to the curve obtained by the steady-state technique ($k_{1,\mathrm{ss}} = 1.56 \times 10^{-15}$ m^2 and $k_{2,\mathrm{ss}} = 1.95 \times 10^{-12}$ m, resulting in $\sigma_{k_1} = 711.8\%$ and $\sigma_{k_2} = 54.4\%$). The reason for this behavior was not physical, but instead was caused by the excessive density of data points in the original curve, which produced null or even positive derivative values for consecutive points that were too close to each other. Consequently, the air velocity (v_s), calculated by Eq. (1), displayed negative or null values along most of the data range, which, in addition to lacking any physical meaning, increased the scattering level and reduced the fitting quality of Forchheimer's equation.

In an attempt to minimize this effect, screening parameters as previously described were applied to reduce the density of points in the original curve. The first parameter applied was the time interval between consecutive points, which varied from 1 to 240 s. The total timespan in the screened data sets was kept unvaried (60 min).

As illustrated in Fig. 2a, the deviation for both permeability constants displayed a minimum value for a time interval of 30 s. Below this value, the point density, which was still high, precluded the calculation of reliable v_s values, increasing the deviation in the k_1 and k_2 values. On the other hand, for time intervals above 30 s, the excessive distance between successive points yielded low representative derivatives, particularly in the viscous-flow region (low P_{cham} and v_s), where the Darcian constant k_1 predominates. This explains the more significant deviation observed for k_1. The best fitting, found at 30-s intervals, produced, respectively, $k_{1,\mathrm{pd}}$ and $k_{2,\mathrm{pd}}$ values of 1.72×10^{-15} m^2 and 1.27×10^{-12} m ($\sigma_{k_1} = 10.0\%$ and $\sigma_{k_2} = 35.1\%$). Nevertheless, these deviation values were considered too high for fitting purposes; hence, the time spacing was not considered the best screening parameter to treat the pressure–decay data set.

The second screening parameter applied was the pressure interval between successive data points. For a pressure range of 5 bar, for instance, a pressure interval of 0.1 bar

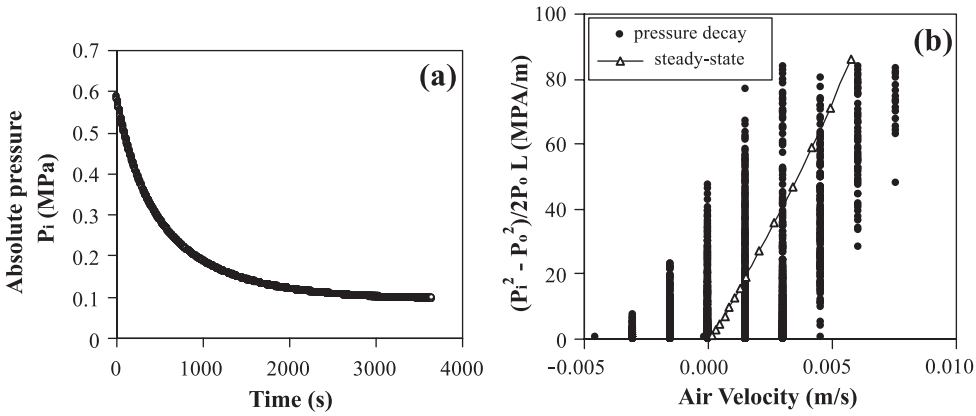

Fig. 1. Original data set acquired at 1-s intervals during 60 min. (a) Pressure–decay curve. (b) Permeability curves obtained by the pressure–decay and steady-state techniques.

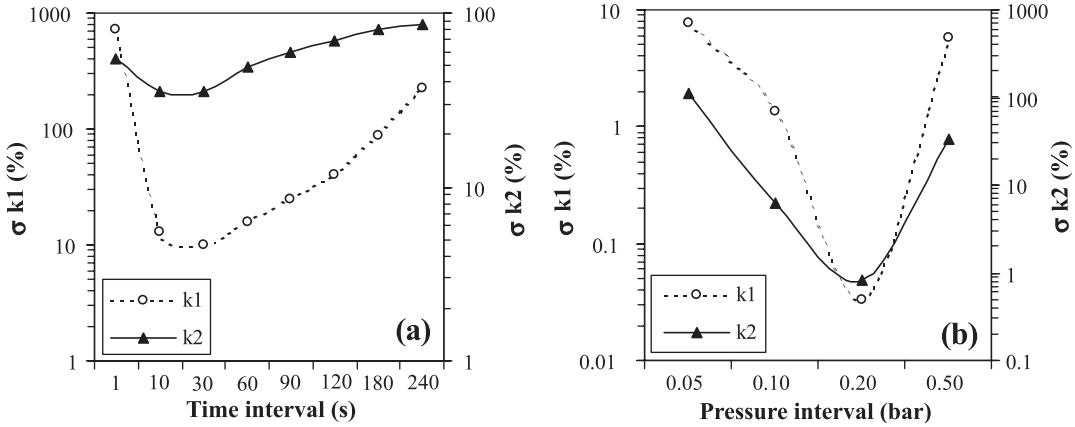

Fig. 2. Deviation between the pressure–decay and steady-state techniques found for k_1 and k_2 permeability constants when the original data set was treated according to (a) a time interval screen, (b) a pressure interval screen.

yielded a curve with approximately 50 points instead of the 3600 points in the original data set. The results of this analysis are illustrated in Fig. 2b.

Fig. 2b shows that the agreement between permeability constants obtained by the pressure–decay and the steady-state techniques improved significantly when the pressure interval screen was applied. The best fitting was achieved with a pressure spacing of 0.2 bar (a data set with 25 points), which yielded $k_{1,pd}$ and $k_{2,pd}$ values of 1.56×10^{-15} m^2 and 1.97×10^{-12} m ($\sigma_{k_1} = 0.03\%$ and $\sigma_{k_2} = 0.84\%$), respectively. For the lower pressure intervals (0.05 and 0.1 bar), the high data density resulted in a high scattering level. On the other hand, the highest pressure spacing applied (0.5 bar) yielded a permeability curve that was nonrepresentative, particularly in its inertial region dominated by the non-Darcian permeability constant k_2. This explains the higher deviation level observed for k_2 in Fig. 2b. Nonetheless, the original data set was apparently treated reliably according to the pressure interval screen. However, a previous analysis is highly recommended to define the best screen value based on the properties of the sample under study. This can easily be undertaken simply by checking the fitting quality of Forchheimer's equation through the correlation coefficient r^2, without the need for a comparison with k_1 and k_2 values obtained by the steady-state technique. For instance, the pressure interval chosen can be considered suitable for a given data set if the resulting fitting of Forchheimer's equation to the screened data points yields an r^2 coefficient higher than 0.99.

The next parameter analyzed was the data range used for fitting purposes. First, the original data set was divided into six subgroups, each 10 min longer than the previous one. The 10-min data set, for example, contained data acquired only in the 10 first minutes, while a 20-min-size set contained data from the first 20 min of collection. The data from the six subsets were screened at a time interval of 30 s, which was found to be the best value in the previous analysis. The results of this analysis are summarized in Fig. 3a.

Fig. 3a shows that the fitting quality was not improved by increasing the time range. In fact, the best fitting was found for data acquired only in the first 10 min of testing.

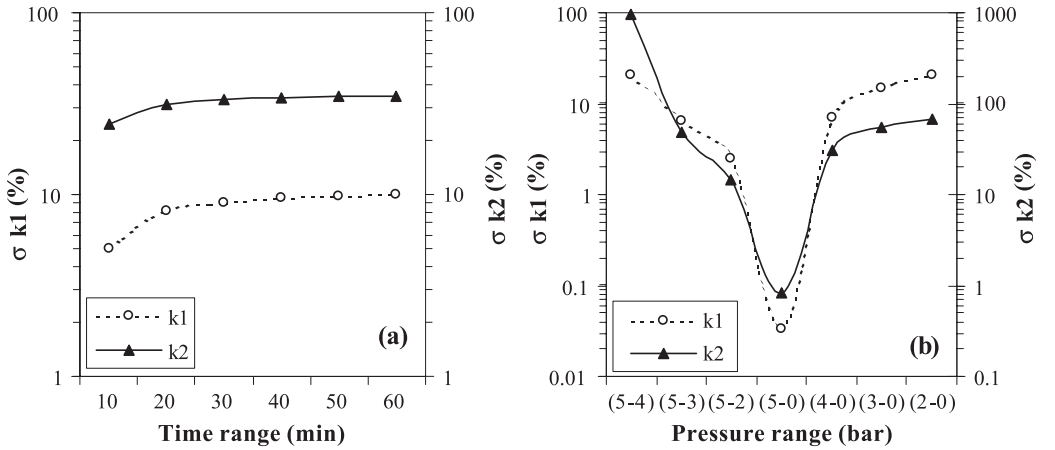

Fig. 3. Influence of the (a) time range and (b) pressure range used for data analysis on the deviation between the pressure–decay and steady-state techniques.

This effect is easily explained by the fact that the greatest pressure decay (from 5 to 1.71 bar) occurred in this 10-min period. The progressive inclusion of additional data points (a remaining pressure decay from 1.71 to 0.1 bar in the next 50 min) only resulted in a higher dispersion level in the permeability curve, particularly at low air velocities, shifting the deviation parameters σ_{k_1} and σ_{k_2} to a constant level of 10% and 35%, respectively. The conclusion here is that the choice of data range to fit permeability constants based on a time criterion is unreliable and should be avoided in practice. Furthermore, as shown by Innocentini and Pandolfelli [7] and Innocentini et al. [6,8], the comparison of permeability constants for different samples is actually reliable only if data are acquired in similar pressure-drop ranges. Therefore, data collection by the pressure–decay technique for different samples in a same time interval (for instance, during only 10 min) may not necessarily result in similar pressure-drop ranges, and hence in comparable k_1 and k_2 values, since the intensity of decay is directly proportional to the sample's permeability level.

As a final analysis, the original data set was split into seven subsets according to the pressure range of interest. For instance, the first subset included data points resulting from a pressure decay from 5 to 4 bar, the second subset contained data from 5 to 3 bar, etc. The data in all subsets were screened according to a pressure interval of 0.2 bar, which was defined as the best value, as previously demonstrated. Fig. 3b summarizes the results of this analysis.

As observed in Fig. 3b, except for the data set containing the entire pressure–decay range (5–0 bar), the deviation between constants obtained by both permeametry techniques was significantly high. This can be explained by the fact that the permeability constants obtained by the steady-state technique ($k_{1,ss}$ and $k_{2,ss}$) were also obtained in a pressure interval of 5 bar, which validates the conclusion that permeability constants obtained for different samples or by different techniques are comparable only if testing is performed in a similar pressure-drop range. Nevertheless, as discussed by Innocentini et al. [5,6], the broader the pressure-drop range used for fitting purposes, the more reliable the resulting permeability constants, regardless of the experimental technique applied [4].

Still in regard to the influence of the region of data analysis, Fig. 3b shows that the deviation in the subset, including points mainly in the inertial-flow zone (5–4 bar), was much higher for k_2 ($\sigma_{k_2}=941.8\%$) than for k_1 ($\sigma_{k_1}=20.7\%$). Similarly, for the data set with points mainly in the viscous-flow zone (2–0 bar), the deviation was still high for k_2 ($\sigma_{k_2}=66.7\%$) but remained practically constant for k_1 ($\sigma_{k_1}=20.4\%$). The main conclusion here is that the lack of data in the viscous-flow range (low v_s and P_{cham} values) is more deleterious to the overall fitting quality of Forchheimer's equation than is the lack of data in the inertial-flow range (high v_s and P_{cham} values).

Finally, Fig. 4 displays the optimized permeability curve resulting from the pressure–decay technique analyzed in this work. The best agreement was found with the steady-state curve screened at a pressure interval of 0.2 bar treated in the whole pressure range of 5 bar.

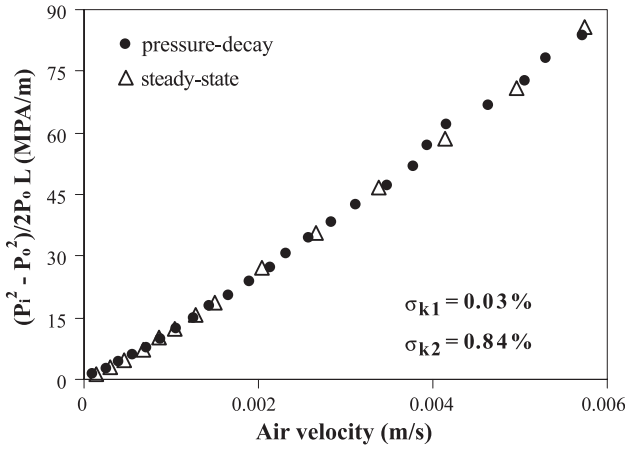

Fig. 4. Comparison between the steady-state permeability curve and the optimized curve resulting from the pressure–decay technique.

4. Conclusions

The objective of this study was to optimize the analysis procedure of a new and promising permeametry technique, whose main advantages are its easy operation and totally electronic data collection. It was found that permeability constants k_1 and k_2 are reliably fitted to Forchheimer's equation only when a pressure spacing screen is applied. The best screen value found in this work was 0.2 bar for data acquired in a pressure range of 5 bar. Nevertheless, a previous investigation is recommended to define the best screen value according to the properties of the sample under study. As for the amplitude of the data set, it was found that the larger the pressure-drop range used for fitting purposes, the more reliable the resulting k_1 and k_2 values, even though permeability constants obtained by different techniques are best compared in similar pressure-drop ranges. The lack of data in the viscous-flow range is apparently more deleterious to the overall fitting quality of Forchheimer's equation than the lack of data in the inertial-flow range.

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References

[1] M.D.M. Innocentini, V.R. Salvini, J.R. Coury, V.C. Pandolfelli, The permeability of ceramic foams, Am. Ceram. Soc. Bull. 78 (9) (1999) 78–84.

[2] A.P. Philipse, H.L. Schram, Non-Darcian airflow through ceramic foams, J. Am. Ceram. Soc. 74 (4) (1991) 728–732.
[3] W.E. Lee, R.E. Moore, Evolution of in situ refractories in the 20th century, J. Am. Ceram. Soc. 81 (6) (1998) 1385–1410.
[4] J. Canon, T. Sander, New test method for evaluating permeability of refractory castables, Refract. Appl. 2 (1) (1997 May) 6–7.
[5] M.D.M. Innocentini, A.R.F. Pardo, V.R. Salvini, V.C. Pandolfelli, How accurate is Darcy's law for refractories, Am. Ceram. Soc. Bull. 78 (11) (1999) 64–68.
[6] M.D.M. Innocentini, A.R.F. Pardo, V.C. Pandolfelli, Influence of air compressibility on the permeability evaluation of refractory castables, J. Am. Ceram. Soc. 83 (6) (2000) 1536–1538.
[7] M.D.M. Innocentini, V.C. Pandolfelli, Permeability of porous ceramics considering the Klinkenberg and inertial effects, J. Am. Ceram. Soc. 84 (5) (2001) 941–944.
[8] M.D.M. Innocentini, V.R. Salvini, J.R. Coury, V.C. Pandolfelli, Assessment of Forchheimer's equation to predict the permeability of ceramic foams, J. Am. Ceram. Soc. 82 (7) (1999) 1945–1948.
[9] M.D.M. Innocentini, A.R.F. Pardo, V.C. Pandolfelli, Modified pressure–decay technique for evaluating the permeability of highly-dense refractories, J. Am. Ceram. Soc. 82 (1) (1999) 220–222.