Determination of Permeability for Refractories: From Standard Test Methods to Improved Interpretation Technique of the Experimental Data

L. Loison, E. de Bilbao, T. Tonnesen, J. Poirier, R. Telle

Transport properties of refractory materials are required to predict the infiltration behavior in contact with corrosive species. For this reason, the determination of permeability values needs to be accurate and inherent to the material, independently from the experimental conditions. The present paper introduces a methodology, used in the field of geology and transposed in this case to refractory samples, in order to obtain reliable intrinsic permeability values. In the first part, this approach is applied to a data set obtained with a permeameter in atmospheric mode as described in the standards. The corrected flow regime plot contributes to select the measurements performed in Darcy flow regime, to further yield the intrinsic permeability with a Klinkenberg plot. In the second part an improved equipment with backpressure mode enables to extend this approach to refractories with low permeability, when Darcy flow conditions cannot be reached in atmospheric mode. The values obtained from the corrected flow regime plot were compared with the ones yield by Klinkenberg plot to ensure the reliability of this method.

1 Introduction

Refractory microstructures determine the physical properties of the materials. For instance open porosity can influence the thermal shock behavior and thermal conductivity, as the grain size distribution can impact the strength of the material. Concerning the resistance to penetration of corrosive fluid pore size and permeability play a major role [1]. In the frame of FIRE project C2 [2], dedicated to the resistance of refractory in a corrosive environment, the research activities have focused on the accuracy of permeability measurement for a reliable prediction of the refractory performance. Permeability [3] is the property of a porous sample to let a fluid flow across a surface under a pressure gradient. This property is relevant as well for furnace reparation, where the unshaped refractory products have to dry in situ and the steam has to be released without explosive spalling. However, in the field of refractory materials, permeability measurements have often shown discrepancies or high standard deviations, since the criterions used to determine the flow regime occurring during the measurement are not always reliable [4–7].

The steady state technique based on Darcy's law is used in Standards ISO 8841:1991 [8], DIN EN 993-4 [9], and ASTM C577-7 [10], where the outlet is left at atmospheric pressure. Darcy's law is used to derive the permeability from pressure drop measurements at different fluid velocities:

$$\frac{P_i^2-P_o^2}{2P_{i,o}L}=\frac{\mu}{k}q_{i,o} \quad (1)$$

where P_i and P_o (Pa) are the inlet and outlet pressures, respectively. L [m] is the length of the sample in the flow direction. μ [Pa.s] is the dynamic viscosity of the fluid. $q_{i,o}$ [m/s] is the flow rate measured either at the inlet or at the outlet of the cell. Darcy's law allows for consistent value determination of the permeability k in the case of so-called Darcian flow regime conditions, i.e. rather at low flow rate. k may be expressed in [m^2], in [mD] (1 mD = 10^{-15} m^2), or in [nPm] (1 nPm = 10^{-13} m^2 = 100 mD). According to Klinkenberg [11], the gas-slippage at pores wall leads to a pressure dependent permeability larger than that would be measured with liquid. When the fluid velocity increases, inertial effects arise due the tortuosity of the pores network and the determination of the permeability value has to rely on the Forchheimer's equation [12]. Innocentini et al. [13] have observed that inertial effects arise for most refractories. The authors proposed to use Forchheimer number to derive the intrinsic permeability.

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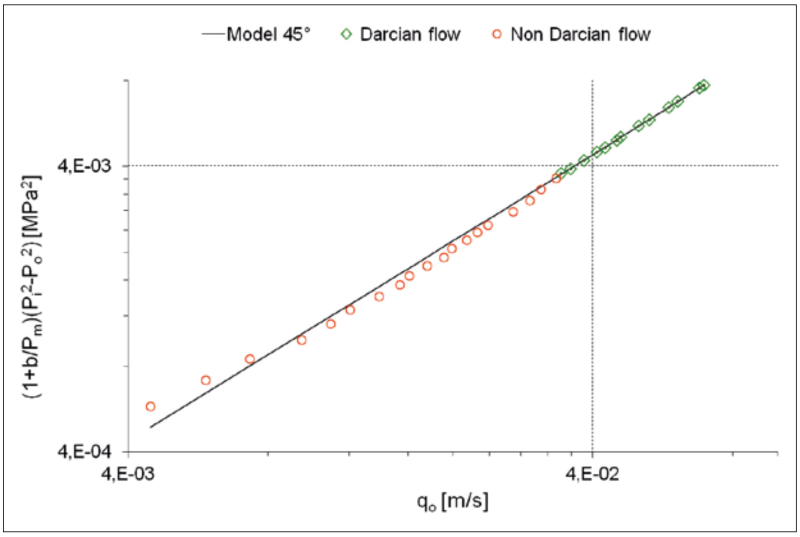

Fig. 1 Corrected Darcy plot of refractory A according to Dranchuk [14]

The standards are advising to determine three experimental data points for three pressure drops and to consider the average as the permeability value of the material, without taking into account the effect of gas velocity. Moreover, the three pressure drops have to be set low enough to neglect the gas compressibility. This presents the following drawbacks:

- Measurements may be performed while Darcian flow regime conditions have not been reached, and the calculation of the permeability based on Darcy's law is invalid
- Refractories with low permeability and high tortuosity may exhibit inertial effects even at this low flow rate and the standard methods do not explain how to manage the measurements in such conditions.

Applying standard method is therefore unsuitable since only an apparent permeability may be determined without controlling if Darcian conditions are fulfilled. Moreover intrinsic permeability cannot be accurately derived from the test.

The objective of this paper is to present two different ways of improvement for the determination of the intrinsic permeability. Two universities belonging to FIRE Federation collaborated toward the improvement of the permeability determination: The University of Orléans/FR and the RWTH Aachen University/DE.

A first solution involves the permeameter with the outlet at atmospheric pressure and performing measurements by increasing the inlet pressure, as suggested in the standard. The correction consists in a better interpretation of measurements data in order to distinguish clearly Darcian flow based on the work of Dranchuk et al. [14, 15]. The Dranchuk's method suggests to plot the log of the so-called corrected pressure drop versus the flow rate deriving Darcy and Klinkenberg's equations [11]:

$$Log\left\{(1+\frac{b}{P_m})(P_i^2-P_o^2)\right\} = Log\left\{\frac{2\mu P_o L}{k_\infty}\right\} + Log\{q_o\} \quad (2)$$

where k_∞ is the intrinsic permeability, $P_m = (P_i + P_O)/2$ is the mean pressure, and b [Pa] is the slippage factor defined by Klinkenberg. Since P_O is constant in atmospheric mode, the points must be aligned on a straight line with 45° slope. The slippage factor b is determined by an optimization procedure giving the best regression coefficient under the constraint of the slope equal to 1.

A second approach consists in modifying the permeameter to be able to apply an adjustable backpressure at the outlet [16]. This allows for increasing the mean pressure while keeping low flow rate to ensure Darcian conditions.

2 Materials and methods

Two refractories, supplied by Bony SA, were tested to illustrate these approaches. The first material (refractory A) was a low alumina content refractory composed of 43 mass-% of alumina, showing an open porosity of 22 %. The second material (refractory B) was an alumina-zirconia based refractory (Al_2O_3 = 50 mass-%, ZrO_2 = 29 mass-%) with an open porosity of 11 %. For refractory A, cylinders of 50 mm in height and 50 mm in diameter were cut. The permeability measurements were made based on the European Standard 993-1 recommendations and measurements were extended to obtain a data set composed of thirty pressure gradients at different fluid velocities. The methodology proposed by Dranchuk was applied on this data set prior to Klinkenberg plot to yield the intrinsic permeability. The value was compared to the one obtained following the standard processing of results. The second part focuses on an improved permeameter allowing tests at low constant mass flow rate with backpressure. This backpressure mode enables to ensure the Darcian flow regime. The intrinsic permeability value was obtained from both the corrected flow plot and the Klinkenberg plot to assess the consistency of this method. This procedure was applied to refractory B, previously cut into a cylinder of 40 mm in height and 35 mm in diameter.

3 Results and discussion

3.1 Selection of Darcian flow conditions

The first tests were performed on refractory A with a permeameter designed according to the standards. The outlet was left at atmospheric pressure and the inlet pressure increased up to 0,13 MPa. The interpretation technique proposed by Dranchuk was applied to select the measurements obtained in Darcian flow [14]. The Klinkenberg equation was used to yield the intrinsic permeability [11], which was compared with values obtained following the European Standard recommendation [9].

Fig. 1 enables to distinguish the flow regimes: the set of points can be split into two subsets. The first one includes the points for which the measurements are not fulfilling the Darcian conditions. The second subset includes the points aligning at 45° that are considered to fit the Darcian conditions with Klinkenberg effect. These measurements were therefore selected to apply the Klinkenberg equation for the identification of the intrinsic permeability (Fig. 2)). The linear regression yields an intrinsic permeability value of 1,69 D (1 D = 10^{-12} m²). The standard proposes to perform three measurements with low Pi-Po and to calculate the average. The average of three measurements belonging to the non Darcian flow set would yield the average permeability value of 1,88 D with a standard variation of 59 mD, i.e. 3,1 %. The standard asks for a deviation smaller than 5 % to ensure the legitimacy of the calculation based on Darcy's law. Even when the standard criteria are verified, the corrected flow plot shows Darcy flow conditions are unfulfilled for low pressure drop. The application of Darcy's law to yield permeability values from those measurement would be incorrect, however the standard do not provide any criteria to discharge values altered by inertial effects.

3.2 Backpressure mode

The first case presented above showed how to improve the analysis to get a reliable assessment of the intrinsic permeability. It has been shown that some points had to be excluded, as they were not measured in Darcian conditions. Moreover, for some refractories, test performed in atmospheric mode shows the transition from a not yet Darcian flow regime to a flow with inertial effects. In this case Dranchuk's method makes all the points to be eliminated and it is not possible to draw the Klinkenberg plot. In this work, an improved permeameter was developed allowing for the application of a backpressure at the outlet of the sample. This technique enables to increase the mean pressure inside the sample while keeping low flow rate. The gas density is then high enough to active Darcian flow regime and the flow rate is low enough to avoid inertial effects.

Fig. 3 shows the usual Darcy plot of the square pressures drop versus the outlet flow rate from measurements performed on material B in atmospheric mode. It can be seen that inertial effects arose even at low flow rate. Fitting a parabolic function gives the Forchheimer's parameters: k_1 = 18 mD, k_2 = 2,3 10^{-11} m. A second series of measurements was carried out by applying a backpressure at the outlet of the cell. It can be seen on Fig. 4 that all the measurements in atmospheric mode were not performed in Darcy flow rate with Klinkenberg effect. In contrast all points measured in backpressure mode show a very good alignment. It is then possible to derive the intrinsic permeability and the slippage factor either from Fig. 4 or by using the usual Klinkenberg

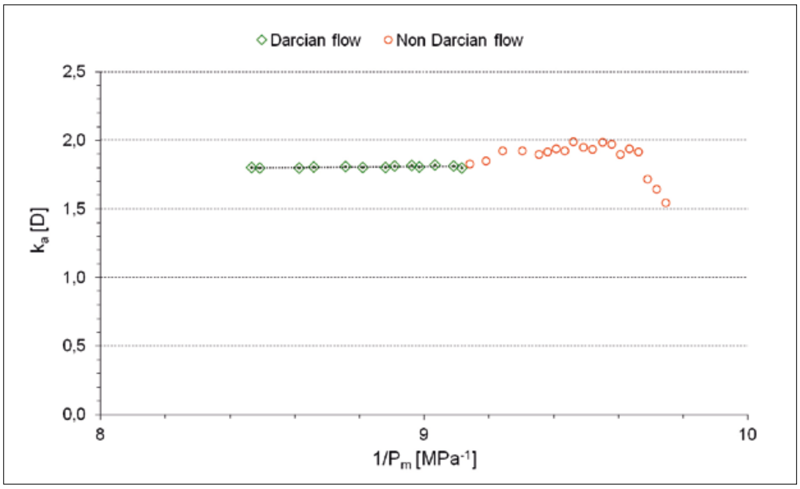

Fig. 2 Klinkenberg plot: determination of intrinsic permeability using measurements performed in Darcian flow regime conditions on refractory A

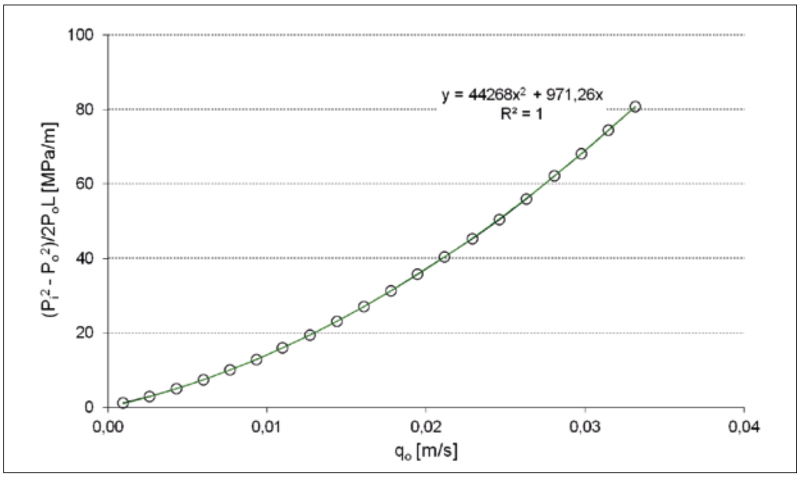

Fig. 3 Darcy plot: measurements performed on refractory B; the parabolic shape indicates that inertial effects arose

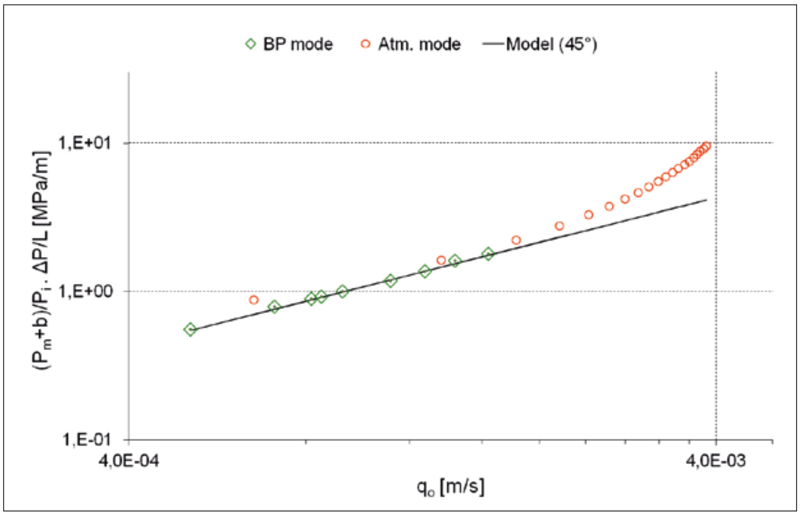

Fig. 4 Corrected Darcy plot of refractory B according to Dranchuk [14]: the straight line with its slope equal to 1 shows that any measurements in atmospheric mode was obtained in Darcian flow regime conditions (circles), all the points obtained in backpressure mode (diamonds) fulfilled the conditions

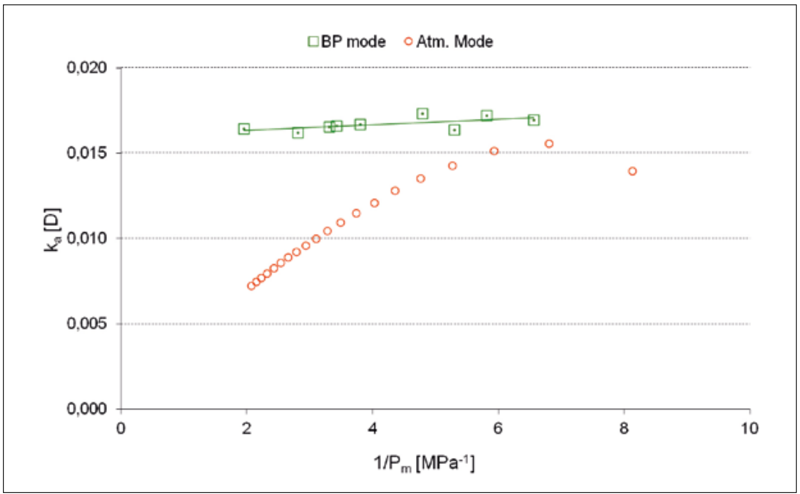

Fig. 5 Klinkenberg plot of refractory B: intrinsic permeability is derived from measurements performed in backpressure mode

Tab. 1 Comparison of the intrinsic permeability and slippage factor values between the corrected flow plot (Fig. 4) and the usual Klinkenberg plot (Fig. 5)

	Intrinsic Permeability [mD]	Slippage Factor [kPa]
From Fig. 4 [14]	16,4	7,96
From Fig. 5 [11]	16,0	10,1

plot (Fig. 5) as done previously with the refractory A. The values of permeability are corresponding while slippage factors show discrepancy (Tab. 1). It is worth noticing that the points are better aligned in Fig. 4 than in Fig. 5. In Klinkenberg plot (Fig. 5), the apparent permeability is derived from the pressures and from the flow rate, which is subjected to higher uncertainty of measurements. This additional calculation yields a higher scattering than the evaluation of the corrected pressure drop used in Fig. 4 (Y axis). This may explain the difference between the slippage factors. However, the intersect of the linear regression giving the intrinsic parameter is affected to a lower extent due to the wide range of inverse mean pressure obtained in backpressure mode.

4 Conclusion

Reliable permeability values are required to evaluate the resistance to physical infiltration of a refractory product. Although standards for permeability measurements do not consider the dependency on the fluid velocities and is based on a single point measurement, it was possible to have a reasonable estimation of the intrinsic permeability by improving the collection and interpretation of a data set. Considering a multi-point measurement of pressure gradient at different fluid velocities, a corrected regime flow plot enabled to distinguish Darcy flow regime. Thereby intrinsic permeability could be yield with satisfactory accuracy with the permeameter described in the standards. However, for refractories with low permeability, the Darcy flow conditions might not be obtained with the outlet at atmospheric pressure. In this situation, an improved permeameter offering flow rate control and backpressure allowed for accurate permeability measurement as it has been illustrated with a dense alumina-zirconia refractory brick. The corrected flow regime plot enabled to yield the intrinsic permeability and slippage factor as well, with lower dependence on the measurements uncertainty, thanks to the corrected pressure drop.

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