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EFFECT OF FLOW CONFIGURATION ON DARCIAN AND FORCHHEIMER PERMEABILITIES DETERMINATION IN A POROUS COMPOSITE TUBE

Hussain NAJMI¹

INSA-CVL, Univ. Orléans, PRISME EA 4229, F-18022 Bourges, France

Eddy EL TABACH²

Univ. Orléans, IUT-Bourges, PRISME EA 4229, F-18000 Bourges, France

Khaled CHETEHOUNA³

INSA-CVL, Univ. Orléans, PRISME EA 4229, F-18022 Bourges, France

Nicolas GASCOIN⁴

INSA-CVL, Univ. Orléans, PRISME EA 4229, F-18022 Bourges, France

François FALEMPIN⁵

MBDA, 1 avenue Réaumur 92358 Le Plessis-Robinson cedex, France

Using Fuel Cell on board of aircraft imposes to extract light species (such as Hydrogen and light hydrocarbons) from the liquid fuel which is stored and used, possibly at temperatures where a fuel pyrolysis occurs. Natural porosity of composite material could be used to filtrate the selected species. Hence the permeability of the porous media becomes one of the key parameter to be accurately measured. It is often determined experimentally in laboratory with disc samples (outlet of the flow is achieved through the porous material) and normal flow. However, this configuration is far from the realistic one consisting of tubes (a main flow is found additionally to the one through the material, tangential permeability). Therefore, the effect of a second outlet on the Darcy's and Forchheimer's permeabilities characterization should be studied (despite the permeability is an intrinsic property of the material itself and it should not be dependent on the test apparatus). This paper focuses on a new way of using an existing test bench for the determination of Darcy's and Forchheimer's permeabilities of C/SiC porous composite tube by taking two outlets into account. Operating parameters (temperature, pressure and mass flow rate) are measured for three different configurations: i) secondary outlet (S.O) is 0% open ii) S.O is 50% open and iii) S.O is 100% open. Then Darcy's and Forchheimer's permeabilities are computed by ISO and P² methods using a direct search algorithm. Obtained results from different methods are compared and discussed. They are in agreement with the literature data which guarantees the reliability of the test bench and of related measures.

Nomenclature

Acronym

<i>CMC</i>	= Ceramic Matrix Composite
<i>PO/SO ratio</i>	= ratio of the flowrate from the Primary Outlet over the one from the Secondary Outlet
<i>P.O</i>	= Primary outlet

¹ PhD student, CE Group, PRISME Laboratory, hussain.najmi@insa-cvl.fr.

² Associate Professor, DMS Group, PRISME Laboratory, eddy.el-tabach@univ-orleans.fr.

³ Tenure Professor, CE Group PRISME Laboratory, khaled.chetehouna@insa-cvl.fr.

⁴ Full Professor, CE Group, PRISME Laboratory, nicolas.gascoin@insa-cvl.fr, Senior AIAA Member.

⁵ Chief Engineer, Hypersonic Programs MBDA, francois.falempin@mbda-systems.com, Senior AIAA Member.

$S.O$ = Secondary outlet

Latin Letters

a_g = Grain area
 d_g = Grain diameter
 d_p = Pore diameter
 K_D = Darcian's permeability
 K_F = Forcheimer's permeability
 L = Length
 P_{inlet} = Pressure inlet
 P_{outlet} = Pressure outlet
 R_e = Reynolds pore number
 T = Temperature
 V = Velocity

Greek Letters

ε = Overall open porosity
 μ = Dynamic viscosity
 ρ = Density

I. Introduction

Using Fuel Cell on board of aircraft could be useful in the coming years for local energy production. The Solid Oxide Fuel Cell (SOFC) for example could be used, mostly if species like hydrogen and light hydrocarbons are available onboard. In the meantime, liquid fuel (stored for propulsion application when burned in the combustion chamber) may encounter temperature that pyrolyses it and that produces light species¹⁻⁵. Since the higher the H₂ or light hydrocarbons content, the longer the life time of the fuel cell, using heavy fuel would decrease the life time of the fuel cell. Thus, a separation of light/heavy species is required. To do so, the natural porosity of the composite materials which are used onboard may be used to separate light from heavy species and thus to increase the H₂ or light hydrocarbons concentration by filtration process.

Ceramic Matrix Composites (CMC) are usually used as a porous material with controlled permeability^{6,7,8,9} to sustain large heat load compared to metallic materials, while operating at much higher temperatures.

The species like H₂, C₂H₂ and C₂H₄ can even get separated from CH₄ by means of the selective permeation property of the CMC porous media¹⁰. Therefore, the fuel could get enriched with low ignition delay species in order to have a more efficient combustion. In the meantime, the filtrated film inside the combustion chamber would be generated with CH₄ mostly which is characterized by a high auto ignition delay compared to H₂, C₂H₂ and C₂H₄^{11,12}. Previous work shows that as the methane content increases there is increase in absorbance value which is clearly seen in IR spectrum around 3100 cm⁻¹¹³. Hence the generated film would thus play a positive role of an additional wall protection by absorbing a maximum of Infrared Radiation from the flame.

This selectivity property of CMC depends upon the permeability of the material or of the porous medium^{14,15}. Over the last decade, it has been recognized that the separation factor for gas pairs varies inversely with the permeability of the more permeable gas of the specific pair. Indeed, an analysis of the literature data for binary gas mixtures from the list of He, H₂, O₂, N₂, CH₄, and CO₂ reveals this relationship for these mixtures^{16,17}. Hence the determination of the permeability and studying the factor affecting it is of prime importance. Performance criteria for the filtration media are high flux rate, stability at variable temperature, pressure and high selectivity even in the presence of other gases and contaminants (e.g. CO, CO₂, CH₄, H₂O, H₂S, etc.).

The filtration process in aerospace sector can be found in other fields of application such as geology^{18,19}, petrochemical^{20,21} or fuel cell. For the fuel cell application, several literature works show that filtration process through membranes is a simple technique which is characterised by a low cost and by a low energy consumption as well as a high purity of hydrogen separation^{22,23,24,25,26}. But in current fuel cells, the temperature remains relatively low or moderate.

In previous works, a high pressure test bench with one inlet and one outlet configuration has been developed^{2,5}. It has been tested on different type of metal and composite disks to determine the Darcian's and Forchheimer's permeabilities with inert and reactive fluids at high temperature and pressure. Similar experimental setup has been used by several authors to determine the permeability of same type of sample geometry or of bar type specimens^{27,28,29}. Ilias et al. (1997) investigated the permeability and selectivity of hydrogen through a palladium

membrane at high temperature and low pressure. Tremblay et al. (2006) determined the permeability, diffusivity and solubility of polydimethylsiloxane, polyisoprene, polyoctenamer, and polyurethane membranes for pure nitrogen, helium, carbon dioxide, methane and formaldehyde. Papaioannou et al. (2015) evaluated carbon dioxide permeability for sintered supports with molten carbonate dual-phase membranes and its performances were found to be stable for more than 200 h of operation. Instead of so many researches on the porous media, still we face a lack in evaluating the correct flow through porous media. Permeability of the material is mostly determined through experiments using disk sample in laboratory, with normal flow. The outflow through the porous media is measured to determine the permeability of the material. Nevertheless, when using experimentally measured values to numerically simulate realistic engineering configurations, a major disagreement is found regarding the estimation of through flow. Hence, several hypothesis are possible, among which: thermal heterogeneity in the system and particularly in the porous material, material heterogeneity at macroscale, estimation of porous medium properties. Since the lab configuration (the porous disc is inserted into a tube which imposes the entire fluid to flow through the porous medium) is not exactly similar to a realistic one, a dedicated work appears necessary. In real engine, a perpendicular flow to the main one occurs in the cooling tube (cf. Figure 1) and the through flow (secondary outlet) is only a small percentage of the fluid that flows into the pipe from the inlet to the main outlet. Hence, it makes necessary to study the effect of a second flow outlet on the determination of Darcian's and Forchheimer's permeabilities (despite the fact that permeability is often considered as a characteristic of the material itself³⁰).

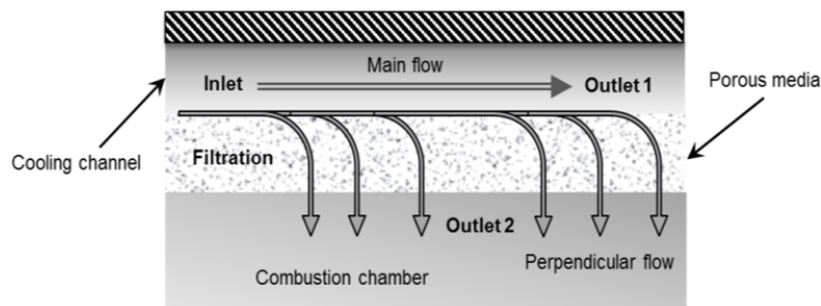


Figure 1: Overview of realistic flows through porous medium in transpiration cooling system (tangential permeation).

The aim of this paper is to determine Darcian's and Forchheimer's permeabilities of a permeable medium in a realistic configuration. Therefore, a porous tube made up of generic C/SiC composite is used as a porous medium and the effect of a second outlet on the Darcian's and Forchheimer's permeabilities is investigated. The next section is dedicated to the description of the experimental setup where different "Primary over Secondary flowrates" ratios (noticed PO/SO ratios) and three values of openings of the second outlet are studied. Two analytical methods for the determination of Darcian's and Forchheimer's permeabilities are given. The results of the porous composite tube permeabilities are presented and discussed in the third section of this paper.

II. Material and Methods

1. Permeation experiments and test methodology

The bench used here is composed of a porous tube connected to gas injection system. Porous tube is made up of C/SiC composite⁴⁸ having a square cross section area of 1 cm² and a length of 1 m. This tube has one inlet and two outlets (the main outlet "outlet-1" is noticed as Primary Outlet –P.O- and the one through the permeable medium "outlet-2" is called Secondary Outlet –S.O-, as seen on Figure 1). The pressure and the mass flow rate are measured on each end of the porous tube. The inlet is located at the left and the outlet-2 is all along the tube although outlet-1 is at extreme right. The injection system is capable of handling liquid as well as gaseous fuels with a high pressure pump (80 bar, 0.5 g.s⁻¹). In this study, nitrogen gas is used as fluid. Four sensors to monitor transient variations of mass flowrates, pressure and temperature are connected to a data acquisition system (Keithley 2700, 1 Hz, 16 bits, 48 channels). The mass flowmeter with a range of 0 to 3 g/s is placed at inlet in order to measure mass flowrate of fluid entering into the porous media. An additional Low flow-Brooks glass tube variable area flowmeter is used at P.O to estimate the through flowrate of S.O by subtracting the P.O flowrate from the inlet one. This Brooks flowmeter is calibrated for maximum flow of 25 mg/s for the nitrogen. Pressure transducer with a range of 0 to 60 bar is placed at inlet side after the mass flowmeter in order to measure the pressure of fluid (Pinlet) whereas both outlets (P.O and S.O) are kept at atmospheric pressure (1 bar). Flow from the primary outlet is controlled by

manually operated valve where three opening percentage values (0%, 50% and 100%) are selected. Hence, this imposes different PO/SO ratios depending on the pressure conditions. Figures 2 and 3 illustrate respectively a photograph of the developed permeation experimental setup and a schematic overview of the porous composite tube. ISO 4022 and P² methods for the determination of fluid permeabilities are followed for all the tests which are performed at room temperature and repeated at least three times.

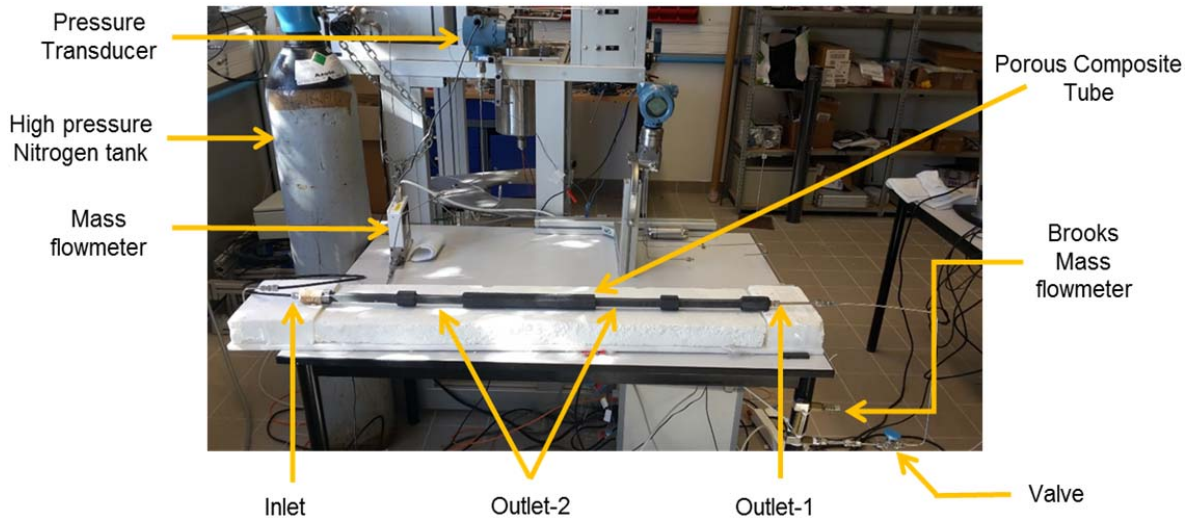


Figure 2: Photograph of the permeation test bench.

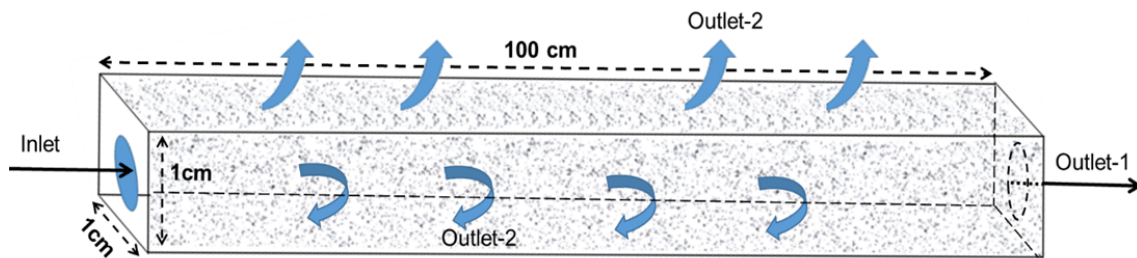


Figure 3: A schematic overview of the C/SiC porous composite tube.

2. Mathematical procedures of Darcian's and Forchheimer's permeabilities determination

Accurate description of fluid flow behavior in the porous media is essential and is described in several ways in various literature studies^{31,32,33}. Usually, Darcy's law depicts fluid flow behavior in porous media³⁴. According to Darcy's law, the pressure gradient is linearly proportional to the fluid velocity in the porous material. This linear relationship is valid only for very small flow velocities or low pressure gradients, typically for pore Reynolds number lower than one³⁵. As the Reynolds number (Re) approaches to a critical value (i.e. turbulent regime), the relationship becomes nonlinear but it gets quadratic due to inertial force. The one-dimensional Darcy equation can be written as:

$$\frac{dP}{dx} = -\frac{\mu V}{K_D} \quad (1)$$

where P is the pressure, x is the direction of fluid flow, μ is the fluid viscosity, V is the superficial velocity, and K_D is the darcian permeability. The transient behavior in porous media is described by Navier–Stokes equations whereas Brinkman's equation below is used to describe the macroscopic fluid flow in large range flow regimes:

$$\frac{\Delta P}{L} = \mu \frac{V}{K_D} + \rho \frac{V^2}{K_F} \quad (2)$$

where $\Delta P = P_{inlet} - P_{outlet}$ is the pressure drop through the porous medium, L is the external mean sample thickness, ρ is the inlet density (with respect to inlet pressure), K_D and K_F are the Darcy's and Forchheimer's terms. The right term of Eq. (2) is composed of two parts, one related to the Darcy's law for low velocity regime filtration (Darcian flows) and the quadratic one is related to the Stokes's law (non-Darcian flows) which takes into account the inertial effects related with flow resistance.

The Stokes equation is also called as Forchheimer's equation^{36,37}. Numerous other formulations of the Brinkman's equation can be found^{38,39,40,41}. Large range of flow regimes through porous media can also be described by using power laws⁴² and cubic laws^{43,44,45}.

In order to compute Darcian's and Forchheimer's permeabilities from the measured parameters on the experimental bench, the ISO 4022 norm urges to use the modified form of the above equation:

$$\frac{\Delta P}{L\mu V} = \frac{1}{K_D} + \frac{1}{K_F} \frac{\rho V}{\mu} \quad (3)$$

The dynamic viscosity, the density and the fluid velocity in the model given by Eq. (3) are calculated at the mean value of the inlet and the outlet pressures. This linear model (called ISO model) can be easily used for the experiments but not in case of Computational Fluid Dynamics codes or engineering which mostly rely on well-defined upstream conditions. It make necessary to use another method based only on inlet condition. To overcome this shortcoming, another method⁴⁶ which based on the use of a quadratic model (called P² model) is considered as a second method for the determination of Darcian's and Forchheimer's permeabilities. The P² model is written as:

$$\frac{\Delta P}{L} = \frac{P_{inlet}^2 - P_{outlet}^2}{2P_{inlet}L} = \mu \frac{V_{inlet}}{K_D} + \rho_{inlet} \frac{V_{inlet}^2}{K_F} \quad (4)$$

with $V_{inlet} = \frac{m}{\rho_{inlet}A}$ and $\rho_{inlet} = \frac{P_{inlet}}{rT}$ where r is the gas constant and T is room temperature.

Using both models, the determination procedure considered here can be stated as the following bound constrained minimization problem:

$$\begin{aligned} & \text{minimize } J(\xi) \\ & \text{subject to } \xi_l \leq \xi \leq \xi_u \\ & \text{with } J(\xi) = \frac{\|\eta_{th}(\xi) - \eta_{exp}\|}{\|\eta_{exp}\|} \end{aligned} \quad (5)$$

where $\xi = (K_D, K_F)$ is the decision variables vector that will be identified, ξ_l, ξ_u are respectively the lower and the upper bounds of these variables, $\|\cdot\|$ denotes the usual Euclidean norm, η_{th} is the theoretical pressure loss given by ISO model (Eq. 3) or P² model (Eq. 4) and η_{exp} is the experimental one.

A direct search algorithm provided by the "Global Optimization" Toolbox of Matlab® is used to solve the bound constrained optimization problem of relation (5). Unlike traditional optimization methods which need gradient or higher derivatives information to search for an optimal point, this optimization algorithm does not require any information about the gradient of the objective function. This direct search algorithm can be used to solve problems with no differentiable objective function or not even continuous, it searches a set of points around the current point, looking for a point where the objective function value is lower than the one at the current point^{47,48}.

III. Results and Discussion

As mentioned in previous sections, the Darcian's and Forchheimer's permeabilities of porous composite tube are determined by two different approaches (ISO method and P² method) and the influence of different secondary outlet openings is investigated. To achieve this determination, ISO method considers the evolution of the term $\frac{\Delta P}{L\mu V}$ as a

function of $\frac{\rho V}{\mu}$ whereas the P² method considers the evolution of $\frac{\Delta P}{L}$ versus V_{inlet} . Figure 5 presents these

experimental evolutions for three different P.O openings (0%, 50% and 100%). The curves are typical and very different because in case of turbulent flow the ISO method gives a linear trend while for P² method quadratic one is observed. However in case of a purely laminar flow (Darcian regime), the ISO method gives a horizontal line and a linear trend is observed for the P² method. In Figure 4 (left), it can be easily seen that for maintaining the same mass flow rate through the porous media the required inlet pressure increases slightly as the P.O opening increases. It is obviously because of the fact that as the P.O opens it creates a new pressure drop along the tube leading to a diminution in driving force. To overcome this, the pressure at inlet side should be increased. However, in order to have the same pressure drop across the tube for all three cases (0%, 50% and 100%) the inlet fluid velocity required is to be slightly lower in later case than the former which can be clearly observed in Figure 4 (right). It is also noted that as pressure drop over material thickness ratio reaches the value of $5 \times 10^7 \text{ Pa.m}^{-1}$ there is no much change in inlet fluid velocity which shows the existence of the turbulent regime expressed by the Forchheimer term.

This stagnation regime due to turbulent flow (slight velocity increase for major pressure drop increase) can be illustrated by the permeation images given as example in Figure 5 for fully closed P.O case. Initially at atmospheric pressure, there is no permeation as there is no driving force. Then having vaporised water and soap on the outer surface of the tube, when the inlet pressure increases, the soap bubble formation on the tube surfaces increases along the length of the porous tube. Bubbles appear near the inlet first and the inlet pressure increases, bubbles are found further along the tube. In addition, once inlet pressure reaches the value of 3 bar, the permeation occurs from all the four faces of the tube and become more uniform as it reaches a value of 4 bar.

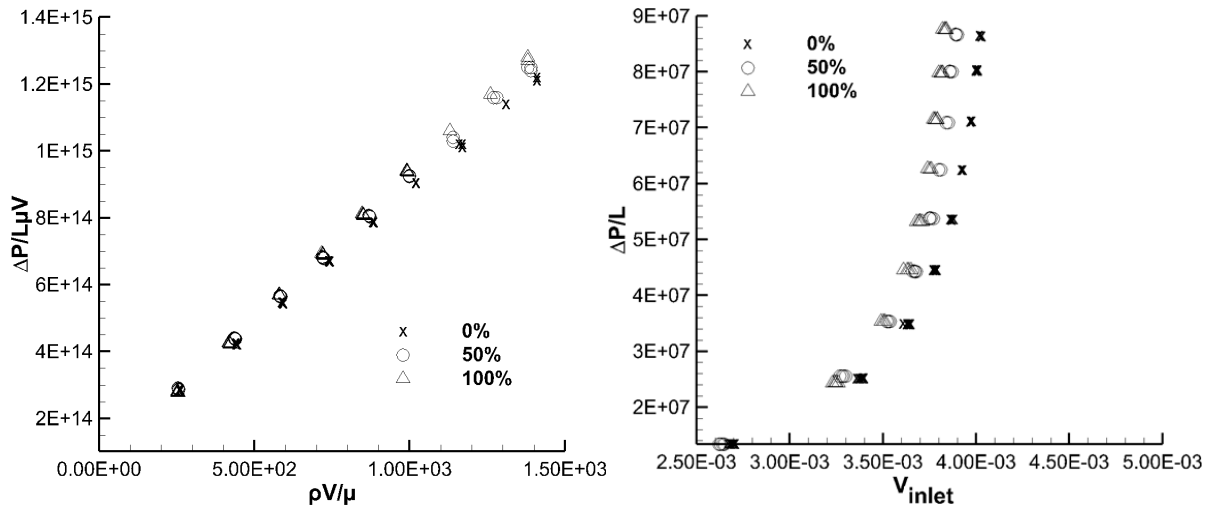


Figure 4. Evolutions of pressure loss in terms of $\rho V/\mu$ (left) and inlet fluid velocity (right).

A semi-quantitative post-treatment of the images of the bubbles formation is proposed in Figure 6 where the locally estimated S.O flowrate is divided by the entire S.O flowrate. Hence permeation distribution is given in percentage of locally permeating flow in order to express how the through flow is distributed over the entire tube length. It is seen that the central part presents a deficit in permeation which is due to the production process of the tube itself which required some structural parts (around 0.15 m, 0.30 m to 0.60 m and around 0.75 m). These structural additional parts are in the same material but the higher local thickness impacts the permeation locally. For pressure of about 4 bar, the permeation tends to be uniform while a major part of the through flow is found upstream for lower inlet pressure.

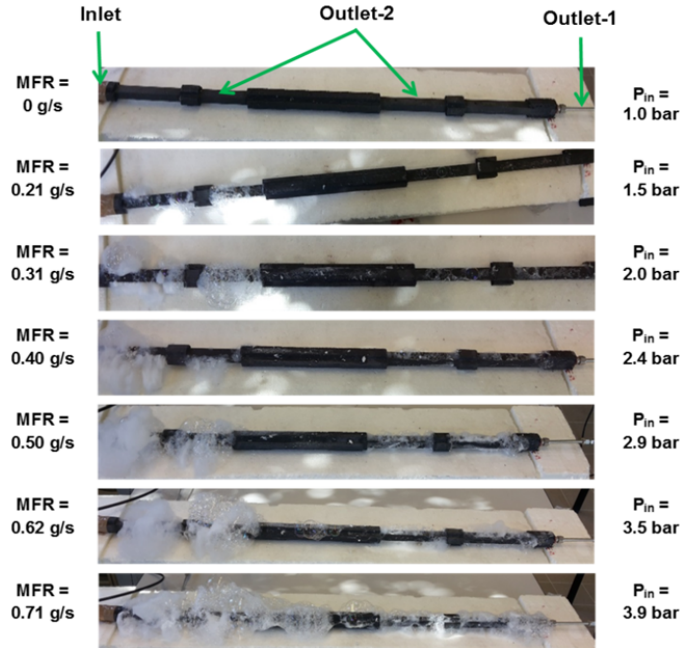


Figure 5. Step wise permeation images with respect to change in pressure and mass flowrate.

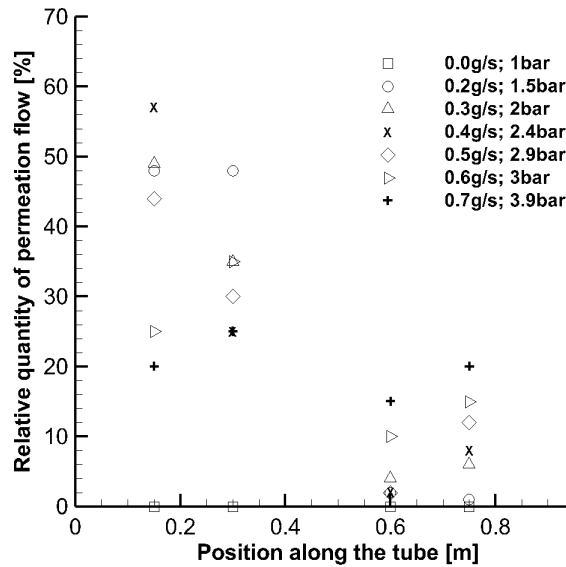


Figure 6. Permeation profile of the composite tube for 0% open case.

As described in earlier section, a direct search algorithm is used to minimize quadratic errors between experimental data and the functions of Eq. (3) and Eq. (4). ISO method (related to Eq. 3) and P^2 method (related to Eq. 4) are used to determine the Darcian's and Forchheimer's permeabilities for different P.Openings (0%, 50% and 100%) which are given in Table 1. A comparison between the two methods (ISO and P^2) shows that Darcian's permeabilities determined by P^2 method for three different P.O openings (0%, 50% and 100%) are slightly higher than Darcian's permeabilities determined by ISO method. The Forchheimer's permeabilities are very close to each other for both the methods in all three P.O openings. Determination of K_D by ISO method leads to a discrepancies of 2.2% and 17.2% for respectively 50% and 100% open cases. However for K_F coefficients, relative gaps of 10.8% and 6.9% is found respectively for 50% and 100% P.O opening. On the other hand, K_D determination by P^2 method leads to relative errors of 13% and 31% for respectively 50% and 100% open cases whereas relative changes of 7%

and 10.9% are observed for K_F terms. The permeability levels obtained in this paper are in quite agreement with those of literature for this kind of composite material (C/SiC). Indeed, Gascoin et al. (2013)⁴⁹ estimated Forchheimer's permeabilities between 10^{-10} and 10^{-14} m.

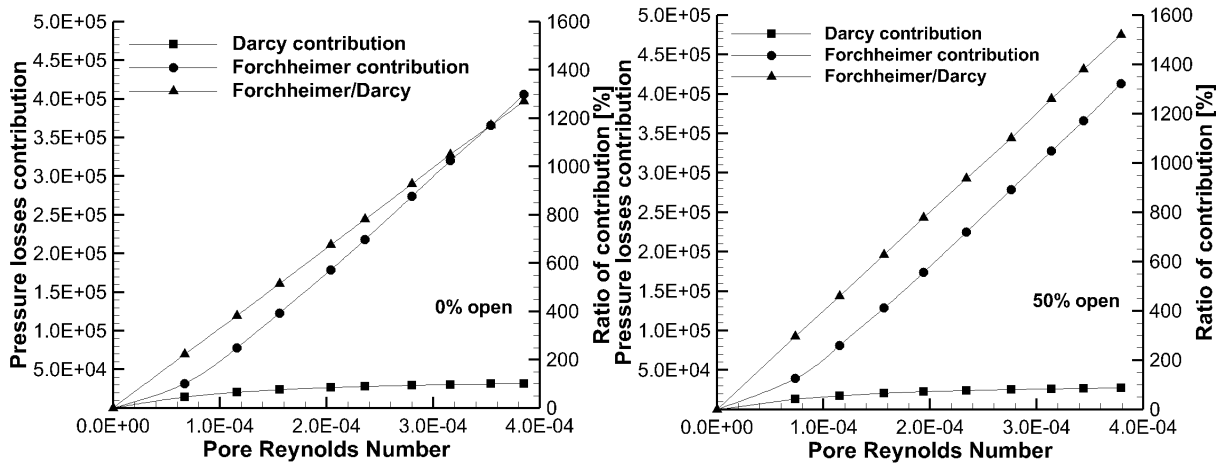
Primary outlet opening percentage	ISO method		P ² method	
	$K_D \pm \Delta K_D$ (m ²)	$K_F \pm \Delta K_F$ (m)	$K_D \pm \Delta K_D$ (m ²)	$K_F \pm \Delta K_F$ (m)
0%	$(1.35 \pm 0.3) \times 10^{-14}$	$(1.23 \pm 0.01) \times 10^{-12}$	$(1.40 \pm 0.3) \times 10^{-14}$	$(1.22 \pm 0.009) \times 10^{-12}$
50%	$(1.38 \pm 0.1) \times 10^{-14}$	$(1.11 \pm 0.006) \times 10^{-12}$	$(1.61 \pm 0.2) \times 10^{-14}$	$(1.14 \pm 0.006) \times 10^{-12}$
100%	$(1.63 \pm 0.4) \times 10^{-14}$	$(1.15 \pm 0.03) \times 10^{-12}$	$(2.04 \pm 0.6) \times 10^{-14}$	$(1.10 \pm 0.03) \times 10^{-12}$

Table 1. Darcian's and Forchheimer's permeability values by two methods at different secondary outlet openings for C/SiC composite material.

Even if measuring accurately these permeability levels is not so easy, we can see a small increase of the Darcy permeability while the tangential flow increase.

It may depend on the way of computing the density (here taken at entry conditions, the pressure drop can increase between the different PO levels, changing the local density to be theoretically taken into account).

In order to have better understanding of above statement and deeper insight of P.O influence on Darcian's and Forchheimer's term contributions on pressure losses, Figure 7 presents the evolutions of these contribution as well as their ratio as functions of pore Reynolds number. The pore Reynolds number is computed for metallic samples by using the relation $Re = \rho V d_p / \mu$ where $d_p = 4\varepsilon / (a_g (1 - \varepsilon))$ is the pore diameter, ε is the overall porosity of material, $a_g = 6/d_g$ is the grain area and d_g is the grain diameter. Let us notice that the grain diameter $d_g = 3.30 \mu\text{m}$ and porosity $\varepsilon = 11.05\%$ of the composite material is computed from a correlation between thickness, overall porosity and grain diameter deduced from the literature data (Gascoin 2011). It is observed from the set of plotted data that for porous composite tube, Darcian's term is always lower than the Forchheimer's term for the three different P.O openings (0%, 50% and 100%) showing that the flow is always in a turbulent regime. There is a decrease in Darcian's contribution as P.O opening increases and a relatively very low change is observed for Forchheimer's contribution. The Darcy's pressure term reaches its limit around unity for all cases (0%, 50% and 100% open) as seen in a previous work for different porous media (Gascoin 2011).



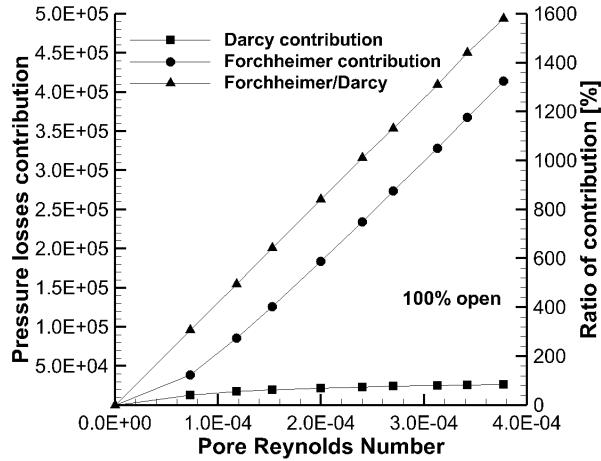


Figure 7. Permeation contributions plotted versus pore Reynolds number for different secondary outlet openings in porous composite tube.

IV. Conclusion

In many applications from aerospace to fuel cell, the permeability of materials is a key parameter that is generally determined experimentally. Lab test conditions (1 inlet/1outlet) are not always exactly those of realistic conditions of use of these materials (1 inlet/2outlets). Consequently, the present work intended to investigate how realistic configuration plays a role on Darcian and Forchheimer permeabilities determination. Additionally, two different procedures (ISO method and P^2 method) of permeability determination were compared. A C/SiC composite tube is used as a porous medium in this paper. It is found that determination procedures (ISO method and P^2 method) gives almost same results when the primary outlet is fully closed for both Darcian's and Forchheimer's permeabilities. After performing the tests for different P.O openings, it is observed that there is no much change in Forchheimer's permeability but there is a slight increase in the Darcian's one as the P.O opening increases. In order to understand more clearly the above statement, pore Reynolds number has been estimated and Darcian's and Forchheimer's contributions are plotted with respect to Reynolds number. It shows that the Darcian's contribution decreases whereas the Forchheimer's contribution remains more or less the same as P.O opening increases. These slight variations are mostly related to the experimental accuracies and further studies will be necessary to conclude on the best way to use the Brinkman equation in case of tangential permeation on a long tube, first under ambient temperature conditions and then in presence of temperature gradients. These experimental values obtained under realistic test conditions (2 outlets) are in agreement with existing data obtained under lab test conditions (1 outlet).

In future works, mathematical backgrounds will be developed to correlate Darcian's and Forchheimer's permeabilities to the selectivity of the porous materials since this would play a role in the fuel enrichment by separating H_2 from CH_4 for example. After that, the spatial selectivity distribution of several binary mixtures will be investigated for different variable parameters such as the type of porous media, the fluid temperature, the pressure and the time.

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