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Fluid flow in complex porous media: Experimental data and IFU model predictions for water vapour permeability

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ABSTRACT

In this research work, an Intermingled Fractal Units' model (IFU's model) has been proposed with the aim of predicting water vapour permeability characteristics of porous rock, normally studied in gas reservoir field. IFU modelling approach commences with the reproduction of experimental pore size distribution with the help of diverse fractal base units mixed together. This procedure allows simulating the porous microstructure in terms of pore cumulative curve or pore size distribution. An analytical procedure is used for obtaining the permeability values. The studied rocks include limestone from South Sardinia quarries and have similar characteristics of those studied in gas reservoir field. They have been taken from four different parts (systems A, B, C and D) of the quarry for the assessment of the heterogeneity of this rock. From the experimental tests, the average water vapour permeability values are equal to 7.22 ± 1.22, 4.21 ± 1.51, 10.09 ± 1.11 and $10.12 \pm 2.42 10^{-12}$ $kg\ m^{-1}\ s^{-1}\ Pa^{-1}$ respectively for systems A, B, C and D, whereas predictions by IFU are respectively equal to 7.15 ± 1.55, 4.25 ± 1.45, 10.12 ± 1.22 and $9.98 \pm 2.18 10^{-12}$ $kg\ m^{-1}\ s^{-1}\ Pa^{-1}$. The comparison shows a good capacity of IFU procedure to predict water vapour permeability values.

1. Introduction

Gas permeability for porous media is a significant issue in various fields associated to catalyzing engineering (Wehinger et al., 2014), soil mechanics (Navarro et al., 2014), packaging (Samaniego-Esguerra and Robertson, 1991), filtration and ultrafiltration membranes (Huang et al., 2013), environmental protection (Gao and Li, 2016), restoration projects (Grover et al., 2016) as well as gas reservoir engineering (Baniasadi et al., 2015). During recent years, this last aspect is being increasingly investigated regarding the significant development in the assessment of reserves and reservoir performance (Baniasadi et al., 2015; Jang et al., 2016). In reality, the necessity of knowing the actual contents of fluid contained in the deposit is useful in planning the extraction activity. As a result, significant information is represented by the fluid flow kinetic through porous materials.

Barrer et al. (Barrer and Grove, 1951) studied the flow of gases and vapours in a porous medium by calculating flow rates, Knudsen and Poiseuille contributions to permeability. They proposed different equations, which present significant deviations from the behaviour highlighted by experimental tests.

Schofield et al. (1990) formulated an analytical procedure in order to predict water vapour flux through membrane distillation by taking into account Knudsen-Poiseuille transition region. A good agreement has been observed between experimental data and model predictions for various membranes and gases at sub-atmospheric pressures. The remarkable simplicity and accuracy of the proposed model makes it favourable over more rigorous models as an engineering equation for gas permeation (Schofield et al., 1990).

A representative study has been carried out by Albaalbaki et al. (Albaalbaki and Hill, 2012) for nanostructured materials. The proposed model developed an interfacial boundary condition for diffusion in composite materials that unify thermodynamic equilibrium, surface diffusion and interfacial exchange kinetics. The model demonstrates that water-vapour transport increases with RH, and that water-vapour transport is sensitive to the microstructural length scale. Calculations suggest that water vapour transport is maximised when the characteristic length scale is of the order of 100 nm (Albaalbaki and Hill, 2012).

Although a large number of models have been proposed during the last six decades, but water vapour transport phenomenon is still largely debated (Baniasadi et al., 2015). Often, these modelling

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approaches are characterised by several empirical correlations or they are limited to peculiar range of applications with the possibility of being generalised, as described by Baniasadi et al. (2015).

Some development has been made when it was clear that the application of new modelling approach must be based on a geometrical description (characteristic lengths) of the porous microstructure excluding the empirical terms. This task seems immediately difficult particularly because the systems are naturally fragmented, irregular and apparently chaotic for being reproduced by *Euclidean* geometry. Regarding this complication, the development of a new geometry having the capability of simulating morphological shapes observed in nature is a key to comprehend physical phenomena as regards to porous materials. In recent times, this role is covered by Fractal Geometry formalised by Mandelbrot in 1970s (Mandelbrot, 2004). The figures explaining the fractal geometry concepts are termed as *fractals* and this term is derived from Latin word, *fractus*, which means "broken", "indented", "irregular", etc. The fundamental characteristics of fractal figures are represented by the fact that the dimension may be non-integer. A sheet of paper is its practice example, which can be assimilated to a surface with Euclidean dimension equal to 2. While rolling this sheet of paper into a ball, the Euclidean dimension is not 2, and is not even 3, as it is not a volume. This dimension has been defined as a non-integer value between 2 and 3 by Fractal geometry. Similarly, Fractal geometry is capable of describing clouds, mountains and surfaces, which are not ascribable to spheres, geometric planes or cones respectively. Furthermore, fractals are constructed by a self-similarity iteration process. In reality, the extent of their irregularity is alike at all scales of observation (Falconer, 2003; Mandelbrot, 2004). This characteristic is very vital for generating figures, which may be assimilated to porous microstructures, particularly for those having dissimilar pore size classes.

Owing to these reasons, for example, fractal geometry has been utilised for explaining diverse characteristics behaviour of natural or advanced porous materials (Atzeni et al., 2008b; Pia et al., 2016a). Zhang (2008) investigated macro as well as micro-porous membranes used in diverse industrial areas. The fractal character is suggested by the disordered nature of microstructures. Box-counting method is used for calculating Fractal dimension. Contact gas permeation experiments with three porous hydrophobic membranes are performed for validating the model. Comparisons are made between the current model and those from references.

Carmeliet et al. (Carmeliet and Roels, 2001) proposed a multi-scale network approach for calculating the isothermal permeability of porous building materials covering hygroscopic and over-hygroscopic ranges. It is based on common moisture properties, such as water vapour permeability, which can be determined by standard experiments. The validation of this approach has been achieved by comparing experimental and simulation results of isothermal capillary absorption and drying processes in ceramic brick and calcium silicate.

Liu et al. (2015) suggested fractal length distribution fractal model to facilitate theoretically study fluid flow in fractured rock masses. Miao et al. (2015) investigated rocks having shear fractures and formalised a model for predicting permeability as a function of their fractal dimensions. The results indicate that the permeability values increase with the increase of the density of fractures as well as pore volume fraction. Zheng et al. (Zheng and Yu, 2012) studied gas permeability via matrix porous media entrenched with randomly distributed fractal-like tree networks. Gas permeability, calculated with the help of an analytical expression, is expressed as the function of fractal geometrical features. Indeed, the proposed model has no empirical constant. The model predictions are compared with simulating results as well as the available experimental data, and a fair harmony is found among them. Martunus Helwani et al. (2012) formalised a fractal permeability model for hydrogen diffusion across porous inorganic membranes. Porous microstructures are described with the help of surface fractal dimension as well as tortuous fractal dimension. Model predictions are in conformity with Kozeny–Carman equation for meso and micro-porosity region. Zheng et al. (2013) explained a fractal model for studying gas leakage rate through a bundle of tortuous capillaries. From the parametric effect study, it is established that geometrical-morphological characteristics and temperature influence the gas leakage significantly. The analytical expression for gas leak rate having specific physical significance reveals its physical principles. Thus, it is strongly recommended for resolving the gas leakage from a container.

A serial contribution to fractal modelling approach has been provided by Cai et al. by proposing a model of spontaneous imbibition on the basis of Hagen-Poiseuille flow. The proposed modelling approach, bearing in mind different shapes, sizes and tortuosity of the pores as well as the initial wetting-phase saturation, is capable of describing the imbibition process in several porous media, natural or artificial (Cai and Yu, 2011; Cai et al., 2015, 2014, 2012, 2010).

Recently, a new fractal approach, termed as Intermingled Fractal Units' model (IFU model), has been formalised to predict numerous characteristics of porous materials, for instance sorptivity, permeability, heat transfer and mechanical properties (Pia, 2016; Pia et al., 2016b, 2015, 2014). It is based on mixing together diverse fractal base units and non-porous parts with the intention of reproducing pore microstructures as pore size distribution and pore volume fraction, and of fractal or non-fractal porous materials. Experimental pore size distributions can be achieved with the help of image analysis, mercury intrusion porosimetry or BET technique. After the reproduction of these experimental pore cumulative curves or pore size distributions, an analytical procedure enable calculating fluid permeability. This approach is based on structural as well as geometric parameters without empirical constant and findings are particularly applicable to real case studied by variegated experimental porous microstructures.

In this work, IFU's model has been formalised for predicting the fluid flow into porous media. As a reference material to experimentally test this method, a calcareous rock has been used. This porous material possesses characteristics similar to those typically studied in gas reservoir field.

2. Materials and methods

The experimental study was conducted on limestones, named Cantone stone, from the quarries of Cagliari, in South Sardinia. This type of rock is similar to those studied in gas reservoir field.

For this research study, four blocks of limestone (A, B, C and D) having size of 40 × 40 × 10 cm were investigated. They have been taken from various parts of the quarry for the assessment of the heterogeneity of this rock.

For this purpose, the tests were conducted on samples apparently free from damage and macroscopic fossils' remains. Subsequently, the blocks were cut into cylinders of diameter equal to 13 cm and thickness equal to 2 cm, with the help of a rotating diamond blade saw. Each block was divided into eight samples (A1-8, B1-8, C1-8, D1-8).

Microstructural–morphological SEM investigation of porous limestones was conducted on conductive samples by a Zeiss Leo 50 XP apparatus function ning with 20 kV of accelerating voltage and an electron source of LaB6.

Mercury Intrusion Porosimetry Technique is used to measure porosimetric characteristics of stone microstructures (32 samples). The porosimeter is the Micrometrics AutoPore IV, which