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Non-invasive in situ concentration determination of fluorescent or color tracers and pollutants in a glass pore network model

Constantinos V. Chrysikopoulos*, Christina C. Plega, Vasileios E. Katzourakis

Department of Civil Engineering, Environmental Engineering Laboratory, University of Patras, Patras 26500, Greece

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ABSTRACT

This study presents a non-invasive imaging method for in situ concentration determination of conservative tracers and pollutants in a two-dimensional glass pore network model. The method presented is an extension to the work by Huang et al. [1], and Thomas and Chysikopoulos [2]. The method consists of fabricating the glass pore network model using a photolithography technique, conducting flowthrough contaminant transport experiments, taking digital photographs at various times of the two-dimensional pore network under ultraviolet or visible light source, and determining the spatially-distributed pollutant concentrations by measuring the color intensity in the photographs with comparative image analysis. Therefore, the method is limited to fluorescent or colored pollutants and tracers. The method was successfully employed to in situ concentration determination of uranine and red color tracers.

1. Introduction

Several non-invasive imaging methods have been developed in the recent years for a variety of solute transport and porous media characterization. The majority of these methods employ optical, magnetic resonance, and gamma radiation techniques. An excellent comparison of the various non-invasive imaging methods available for subsurface contaminant migration applications was presented in the review paper by Werth et al. [3]. For direct and clear viewing of solute and particle movement, as well as two-phase immiscible flow within a complex pore space, micromodels are often employed instead of packed columns. Micromodels are transparent networks of pores and constrictions that simulate some of the complexities of natural porous media. Numerous visualization flow and transport in porous media studies presented in the literature employ etched micromodels [4–12] or monolayers of glass beads [13–15].

An imaging procedure using ultraviolet (UV) illumination has been reported by Huang et al. [1] in order to evaluate the transport of fluorescein in a transparent plastic (Perspex) box with internal dimensions 18 × 28 × 1 cm, packed with glass beads. The procedure consisted of illuminating the fluorescein with UV light, capturing the emitted fluorescence by a camera, and converting image intensities to fluorescein concentrations. The procedure was shown to be simple and moderately accurate, but limited to fluorescent tracers. Recently, Thomas and Chrysikopoulos [2] developed a method for accurate in situ measurement of conservative tracer and colloid concentrations in columns packed with glass beads. The method consists of fabricating clear sintered glass-bead-packed columns, by taking digital photographs of the column under a UV light source, and determining concentrations by measurement of the fluorescence intensity of the tracer or colloids in the photographs. The method was shown to be as accurate as standard effluent sampling and is capable of capturing concentration data at multiple time steps, but it is limited to one-dimensional packed columns. The objective of this research is to extend the work by Huang et al. [1], and Thomas and Chrysikopoulos [2] to the case of in situ measurement of conservative red color (rc) tracer and uranine (u) concentrations in two-dimensional etched micromodels under ultraviolet or visible light sources and to eliminate errors that could occur in previous studies due to image blurring caused by optical dispersion through glass-bead-packed porous medium, and solute distribution within the thickness of the porous medium.

2. Procedures and materials

2.1. Micromodel fabrication

The selected micromodel pore network was carefully designed with AutoCAD® and was printed on a high quality transparent sheet of acrylic film. The transparent pore network pattern, shown in

* Corresponding author. Tel.: +30 2610 996531; fax: +30 2610 996573.
E-mail address: gios@upatras.gr (C.V. Chrysikopoulos).

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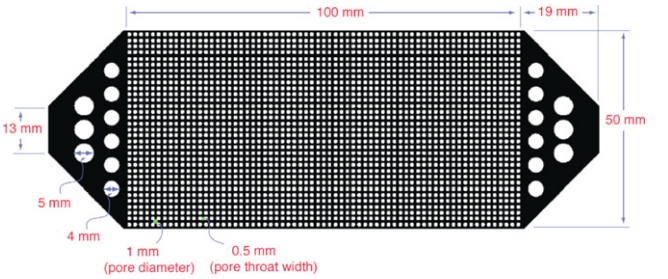

Fig. 1. Computer image ("mask") of the pore network micromodel. The dark areas correspond to the voids of the micromodel. There are 67 pore bodies along the x-direction is 67 and 34 along the y-direction.

Fig. 1, was used as a photographic "mask". Therefore, the dark areas in the mask represent the void spaces of the micromodel. The dimensions of the pore network are L_x = 100 mm in length and L_y = 50 mm in width. The diameter of each pore (pore body) is 1 mm, and the width of the narrow channels (or pore throats) connecting the pores in a square lattice arrangement is 0.5 mm. The larger voids at the two ends of the micromodel were included in order to aid in the uniform distribution of the incoming fluids.

The micromodel was constructed with the photolithographic technique developed by McKellar and Wardlaw [16] with certain modifications initially proposed by Payatakes and co-workers [17–19]. The method consists of a photo-imaging procedure followed by chemical etching of the glass. Two pieces of mirror glass with approximate dimensions 21 cm in length and 12 cm in width were placed in 600 mL single-distilled water (sdH_2O) solution containing 200 g NaOH for a 24 h time period, in order to remove the protective layer of the mirror and to expose the copper. Then, the mirrors were gently washed with sdH_2O, and were placed in a dark and dust-free room where the copper surfaces were sprayed with Positive Resist (Cramolin, Germany) until a visible film had built up. Subsequently, the mirrors were placed for 30 min in a furnace, which was preheated at 70 °C, in order to allow the film to dry. The Positive Resist was used because it offers a convenient way to accurately copy any illustration onto a great variety of materials and ensures relatively fast drying and high sharpness. Each glass essay was allowed to cool off, and the network "mask" was adapted on the surface with the Positive Resist film. The glass essays were exposed for 45 min to ultraviolet light in a custom-made large wooden container were the visible Positive Resist film was polymerized. Subsequently, the glass essays were retained in dark room, and they were placed in a sdH_2O bath containing 7 g/L NaOH for a few minutes (<10 min) to dissolve the non-polymerized Positive Resist film. The precise bathing period was determined by visual inspection under the red light (Philips darkroom lamp 230 V). This was the last visible-light sensitive step. Next, the glass essays were washed with sdH_2O and placed in a HNO_3 solution bath (200 mL HNO_3 65% and 230 mL sdH_2O) for approximately 10 s to dissolve the copper surface area, which is no longer covered by the Positive Resist film. Then, pre-heated wax was carefully spread to form a layer over the glass essay area surrounding the pore network that should not be etched. Additional wax was placed at the four edges of the glass essay in order to form a thick wall capable of retaining approximately 30 mL of fluids at the top surface of the glass essay. Subsequently, 20 mL of HF solution (prepared with 75 mL HF 90% in 25 mL sdH_2O) were poured on the waxed surface of the glass essay for 5 min. The glass essays were thoroughly washed with sdH_2O and the wax was carefully removed. Subsequently, the glass essays were placed in a HNO_3 solution until all of the remaining copper was removed. Then, the transparent glass essays were washed with sdH_2O. Prior to sintering, two holes were carefully drilled in one of the two etched glasses in order to create the necessary inlet and outlet openings of the micromodel. Finally, the desired two-dimensional micromodel was constructed by sintering the two etched glasses in a programmable furnace. Based on numerous preliminary sintering tests, the furnace was programmed to the following temporal temperature variation: 2 min at 40 °C, 6 h at 400 °C, 2 h at 500 °C, 2 h at 600 °C, 30 min at 690 °C, and 30 min at 600 °C. Scanning electron microscope (SEM) images of the etched micromodel are presented in Fig. 2. However, it should be noted that one significant limitation of the method presented is that pores with radius <20 μm are quite difficult to be etched with great precision on the glass essay.

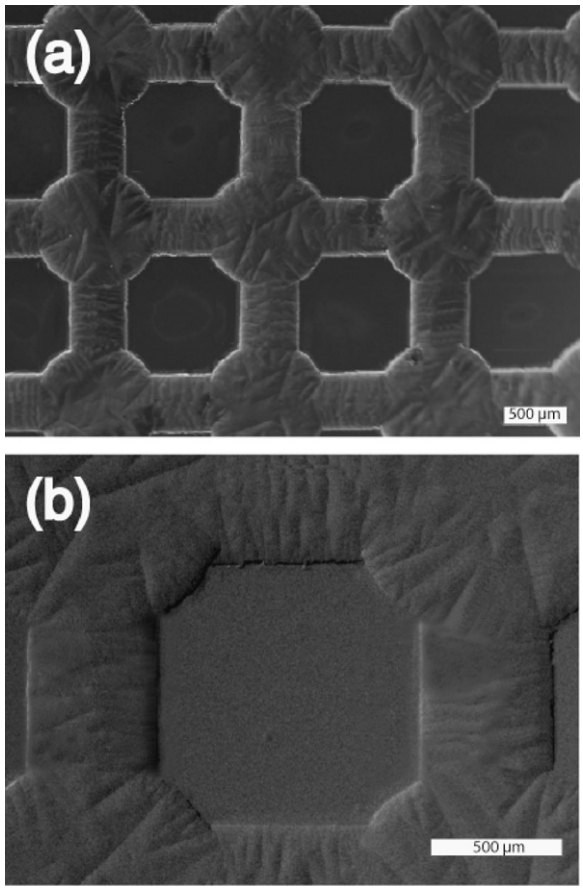

Fig. 2. Scanning electron micrographs of (a) a selected micromodel section, and (b) four connected pores in a square lattice.

The porosity, pore volume, pore depth, and cross sectional area of the micromodel, listed in Table 1, were determined by both simple geometric considerations and by direct use of the line scan option of SEM. However, it should be noted that the pore depth value listed in Table 1 is a spatially averaged value, because the micromodel fabrication technique employed does not guarantee a spatially uniform pore depth. The intrinsic permeability, k [L^2], of the micromodel was determined by conducting flow through

Table 1
Parameters of the pore network micromodel.

Parameter	Symbol	Value
Length	L_x	10 cm
Width	L_y	5 cm
Porosity	θ	59.2%
Pore depth	L_p	390.2 μm
Pore volume	V_p	1.184 cm^3
Cross sectional area	A_m	1.95×10^{-5} m^2
Intrinsic permeability	k	2.23×10^{-10} m^2