Removal of dissolved benzaldehyde from contaminated water stream using granular iron slag by-product in the permeable reactive barrier technology

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ABSTRACT

This work explores the suitability of iron slag solid waste as a sorbent in the permeable reactive barrier technology in order to achieve sustainability principles for remediating groundwater contaminated with benzaldehyde. Numerical modeling was able to simulate the measured breakthrough curves under various values of flowrates and initial concentrations with a coefficient of determination greater than 0.98. This model is based on solving of advection–dispersion–sorption equation using computer solution (COMSOL) Multiphysics 3.5a. Results certified that there was a direct relation between longevity of the barrier measured by breakthrough time and bed depth; however, this longevity was decreased dramatically with an increase of inlet concentration of contaminant and flowrate of flow. Hydraulic conductivity was recognized to remain approximately constant which certified that there was no precipitation occurred through the removal process. Finally, the dissolved calcium oxide (formed 14% of iron slag) enhanced the sorption process because calcium can compose the cation bridge between benzaldehyde and iron.

Keywords: Benzaldehyde; Groundwater; Sustainable; Contamination; Permeable reactive barrier; Iron slag

1. Introduction

Water contamination with organic compounds is a popular environmental problem that resulted from the activities of different industries like mining, processing of metal, generation of power, electroplating, and pigment [1–8]. These compounds can penetrate the subsurface environment and transport through unsaturated-saturated zones of the soil. Frequently, the migration of organic contaminants (called non-aqueous phase liquids (NAPLs)) is accompanied by the formation of three types of plumes; namely, NAPL core, dissolved plume formed by dissolution and vapor plume resulted from volatilization. The dissolved plume can cause significant contamination for groundwater which is considered more reliable from the surface water. Benzaldehyde (C_7H_6O) is the easiest and the most industrial useful members of the aromatic aldehyde families. It exists in nature as combined forms like glycosides in almonds, apricots, cherries, and peaches seed. This compound is

utilized in the foods, beverages, pharmaceuticals, perfumes, soaps, and dyestuff industry [9]. Benzaldehyde releases to the environments in emission from the combustion processes like diesel and gasoline engine, incinerator, and wood burnings. It was made in the atmospheres through photo-chemicals oxidations of toluene and other aromatics hydrocarbon.

Many technologies and theories were devoted to the efficient elimination of organic compounds and other pollutants from the surrounding ecosystem [10,11]. One of them, permeable reactive barrier (PRBs) selected to be the best achievable approach for treating wide broad of the contaminant released to the surfaces and, consequently, reached to the subsurface water because of its lower cost, high efficacy and environmentally friendly behavior [12,13]. It requires the emplacement of an *in-situ* reactive bed with hydraulic conductivity comparable to the ambient aquifer especially the sandy soil. The bed will purify the groundwater through the capturing of the contaminants under the natural hydraulic gradient. Accordingly, the *in-situ* remediation of groundwater by this technique is more attractive than handling large quantities of contaminated water to the above-ground that will be associated with high expenses for operation and maintenance [14,15]. To ensure the high removal of the contaminants within the reactive materials, substantial exploration and investigation for characteristics of these materials must be achieved [16–18]. The selection of the best suitable reactive mediums primarily depends on the sorption capacity, hydraulic conductivity, availability, low-cost, long-term stability, and environmental compatibility [19]. Based on the target contaminants that required to remove from the subsurface environment, sorbents applied in the PRB must be easily available, not generated secondary hazardous by-products, and cost-effective with reactivity and hydraulic conductivity remain efficient as a function of the time [20]. Activated carbon, zeolite, and other conventional sorbents have been utilized for treating polluted streams [21,22]. Although these sorbents are generally suitable for the removal of organic pollutants and the high cost limited their widespread uses [23,24]. The adsorption of the sodium salt of *p*-phenol sulfonic acid, sulphanilic acid, and benzaldehyde from water, base or acid solutions onto activated carbon cloth by *in-situ* UV-spectrophotometric method has been identified. Kinetics of adsorption was followed over 90 min and the first-order rate law introduced a good fit for kinetic data [25]. The ability of granular activated carbon for adsorption of benzaldehyde from aqueous solutions was studied under the effects of adsorbent dose, contact time, and temperature on adsorption capacity. The work proved that the adsorption capacity of sorbent for benzaldehyde was decreased with the increase of temperature. Also, pseudo-second-order and Temkin models were found suitable to represent this adsorption process [9]. The equilibrium adsorption resulted from the interaction of activated carbon with benzaldehyde, aniline, and benzoic acid was investigated. The results showed that the Hill and Koble–Corrigan isotherm models could be effectively described the adsorption process depended on the values of the mean relative percent error, least-square parameter, and Durbin–Watson Test. Furthermore, the results proved that benzoic acid was adsorbed more than the other compounds

[26]. Finding the ability of iron slag for removing of benzaldehyde from wastewater with identification, the predominant mechanisms of the uptake process was elucidated. By applying the Langmuir model, the batch tests proved that the maximum uptake capacity of iron slag was 118.25 mg/g. The main mechanism for the removal of benzaldehyde onto iron slag was the physical sorption based on the analysis of isotherm and kinetic sorption data and thermodynamically, the process was spontaneous and endothermic [27].

Hence, finding the non-conventional materials with low costs that have a good ability in the reduction of pollution became a good topic for many studies. By-products released from several industries such as bottom ash, fly ash, and iron slag, as well as agricultural wastes like rice husk and tree bark, maybe tested as a reactive bed for PRB. The previous records signified that the annual world production of iron slag reached 50 million tons which consider by-products from the industry of iron and steel [28,29]. The generated quantities were found an environmental problem because they consider persistent wastes that remain for long period; so, the application of this slag as a sorbent in PRB is more attractive in the terms of sustainable development. The existence of benzaldehyde in groundwater might be because of the accidental release of the petroleum products and their subsequent migration to the subsurface environment or spillage of these compounds from the storage tanks constructed underground level. The petroleum products; especially, the benzaldehyde compound can cause extreme damage to humans and other life forms. Therefore, their existences in the groundwater resource often require urgent action to remediate the contaminated locations or at least to control the source of contamination. Accordingly, the major aims of this work are; (1) to monitor the reactivity and permeability of the iron slag by-product in the PRB technology for restriction the migration of benzaldehyde in the subsurface environment based on the mathematical and experimental models, and (2) identification of predominant mechanisms responsible of benzaldehyde removal due to its interaction with adopted sorbent depended on the spatially and temporally dissolution of calcium oxide from iron slag in the aqueous solution.

2. Materials and methods

2.1. Materials

Iron slag produced as a by-product from the steel industry was selected to be the sorbent material applied in the PRB to remove the dissolved plume of benzaldehyde from artificial polluted water based on the column tests. These tests aim to find the contaminant transport in a packed column for simulation of PRB role in capturing of benzaldehyde front. Iron slag was collected from the factories of steel and iron available in Babylon Governorate/Iraq. It is washed with distilled water for removing fine powder and, thereafter, dried at 105°C for 24 h. This material with grain size of (0.6–1 mm) has bulk density, actual density, surface area (Brunauer–Emmet–Teller, BET), porosity, mean diameter, hydraulic conductivity, ash content and pH of 2,026 kg/m^3, 3,414 kg/m^3, 0.2571 m^2/g, 0.41, 0.775 mm, 2.69 × 10^{-3} m/s, 10% and 8%, respectively. The surface area was measured

using surface area analyzer, BET method, Quantachrome, USA while the actual density analyzer, Gas Pycnometer, S/N:0429010-1849, USA utilized to measure the actual density. The porosity of the iron slag – packed bed was evaluated by the water evaporation method. The calcium oxide forms approximately 14% from the composition of the iron slag and this compound will be specified the predominant mechanism responsible for the sorption process.

At room temperature, the benzaldehyde was dissolved in the ethanol and the resulted solution must be mixed with distilled water to obtain the water contaminated with a specific concentration of this contaminant. This concentration was measured by high-performance liquid chromatography (HPLC, Shimadzu 2010, Japan). The pH of the water samples was changed by the addition of 0.1 M HNO$_3$ or NaOH as needed.

2.2. Experimental procedure

The adsorber continuous set up utilized in this work was manufactured of four similar acrylic columns in row configuration. The wall thickness of each column equal to 5 mm, while the inner diameter and length dimensions have the values of 5 and 50 cm, respectively (Fig. 1). Column experiments are assumed that the migration of contaminants in the one dimensional (1D) for uniform flow of water. This assumption seems fair for the present situation because the choice of small diameter for the used column was to ensure that the transport of contaminant would be in the direction of flow and no radial migration will occur. Also, using iron slag with diameter ranged from 0.6 to 1 mm means that the velocity of flow will not change along with the bed depth; so, the uniform flow will be predominant. For analyzing both reactivity and hydraulic performance, several parameters such as bed longevity, hydraulic conductivity, and calcium concentration are monitored through this migration which reflects the real operation of PRB. The columns were packed with iron slag without introducing any aquifer because the aim of the continuous tests is to evaluate the efficacy of this material as PRB in the elimination of contaminant concentration. The packed bed was fed with distilled water from the bottom of the column to avoid the entrapped air. The up-flow column test was performed at room temperature.

After the saturation of iron slag bed with distilled water, the polluted water was introduced to the column with a certain value of flow rate under constant hydraulic gradient. The hydraulic conductivity coefficient for packed bed was determined with time by applying Darcy's law using hydraulic gradient and accumulative effluent of water. Ports designated as P1, P2, P3, P4, and P5 are equipped with each column situated from column bottom at 0, 10, 20, 40, and 50 cm respectively. These ports are utilized to withdrawn the periodic water samples from the central axis of the bed with aid of syringes in order to monitor the contaminant concentrations.

3. Governing equation for contaminant migration

Three familiar processes governed the migration of dissolved contaminants in the subsurface environment specifically; advection, dispersion, and sorption. The migration of solute as a result of flowing water represents the "advection" while "dispersion" means the transport of solute due to varied pathways; however, this transportation will accompany a reduction in solute concentration due to "sorption" process. The last process can be represented by the "reaction term" which is identified by the isotherm model. One dimensional advection–dispersion–sorption equation takes the following form [30]:

$$D_z \frac{\partial^2 C}{\partial z^2} - V_z \frac{\partial C}{\partial z} - \frac{\rho_b}{n} \frac{\partial q}{\partial t} = \frac{\partial C}{\partial t} \qquad (1)$$

where ρ_b is the bed-bulk density (kg/cm³), n is the bed porosity, D_z is the dispersion coefficient (m²/s), V_z is the flow velocity and C is the contaminant concentration (mg/L) at distance z and time t.

The q represents the quantity of contaminant sorbed on the iron slag (mg/g). A previous study conducted by Faisal et al. [27] was included the development of best sorption isotherm for a description of the interaction of iron slag with aqueous solution contaminated with benzaldehyde through batch tests. The results proved that the sorption data can be fitted with the "Langmuir model" as clear in Eq. (2). Combination of Eq. (1) with the Langmuir model [Eq. (2)] will give the general equation [Eq. (3)] for benzaldehyde migration:

$$q = \frac{118.25 \times 0.052 \times C}{1 + 0.052 \times C} \qquad (2)$$

$$D_z \frac{\partial^2 C}{\partial z^2} - V_z \frac{\partial C}{\partial z} = R \frac{\partial C}{\partial t} \qquad (3)$$

where R is the retardation factor equal to $\left(1 + \frac{\rho_b}{n}\left(\frac{118.25 \times 0.052}{\left(1 + 0.052 \times C\right)^2}\right)\right)$. The last equation was solved by the finite element method using computer solution COMSOL Multiphysics 3.5a (2008).

4. Results and discussion

4.1. Longitudinal dispersion coefficient

The effective coefficient of longitudinal dispersion for the iron slag bed was determined by the tracer experiment. This experiment is included the filling of the column with 50 cm of this slag in the dry condition. Tracer test required to apply of deionized water with 1 g/L NaCl under various values of seepage velocity (0.01, 0.02, 0.03 and 0.04 cm/min) with corresponding flow rate (5, 10, 15 and 20 mL/min) respectively. Because the movement of the groundwater in the porous medium is mostly identical to the laminar flow (Reynolds number < 1–10) [31], the values of the velocities were chosen to satisfy this type of the flow. As a function of the elapsed time, the concentration of this tracer was measured by electrical conductivity based on "the electrical conductivity of sodium chloride solution increased
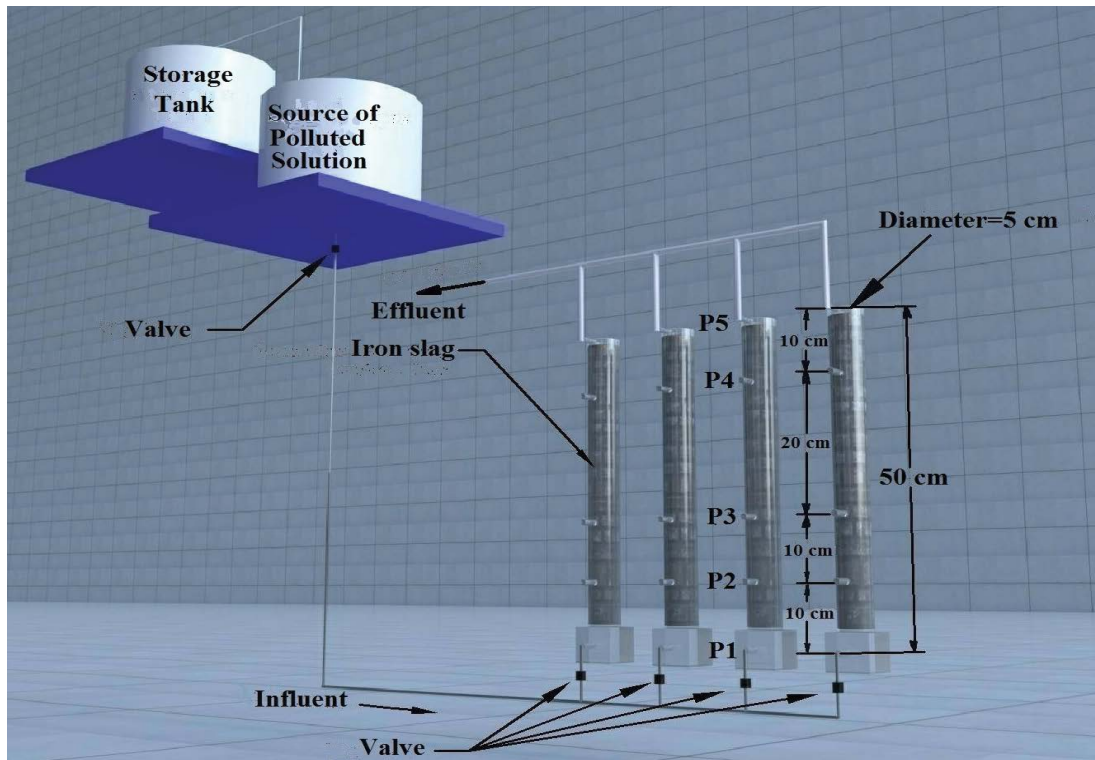
Fig. 1. Schematic representation of the experimental set-up used in the continuous tests.

linearly with increasing its concentration". The value of D_L is specified as below [32]:

$$D_L = \frac{1}{8}\left[\frac{\left(z_0 - Vt_{0.16}\right)}{\left(t_{0.16}\right)^{0.5}} - \frac{\left(z_0 - Vt_{0.84}\right)}{\left(t_{0.84}\right)^{0.5}}\right]^2 \qquad (4)$$

where D_L is the longitudinal dispersion coefficient; z_0 is the distance along the z-axis (L); V is the pore velocity; $t_{0.84}$ and $t_{0.16}$ represent the times corresponding for C/C_0 of 0.84 and 0.16, respectively.

Measured values of D_L at V for iron slag bed are shown in Fig. 2 with a linear relationship as follows:

$$D_L = 18.08V + 0.0275 \; R^2 = 0.9903 \qquad (5)$$

So, the dispersivity is 18.08 cm which considers the main input parameter required in the COMSOL program to calculate the value of the dispersion coefficient in the transport equation (Eq. (3)).

4.2. Benzaldehyde migration in iron slag bed

The results obtained from column tests were discussed by monitoring of the following parameters; (1) variation of C/C_0 as a function of the time (i.e., breakthrough curves) at ports P2 (10 cm), P3 (20 cm), P4 (40 cm) and P5 (50 cm) for a period not exceeded 1,200 h, and (2) variation of both hydraulic conductivity (K) and calcium concentration (Ca) with the operation time. The present study is applicable for one-dimensional contaminant transport with a uniform flow of water for the homogenous porous medium by taking the advection, hydrodynamic dispersion, and sorption processes into account.

4.2.1. Breakthrough curves and modeling

• *Effect of inlet concentration*: Figs. 3 and 4 draw the influence of variation the inlet concentration in the range of 300–500 mg/L at certain values of flowrate and bed depth on the shape of the breakthrough curves. The saturation rate of the slag bed is depended primarily on the magnitude of the concentration gradient. The breakthrough curve shape is frequently less pronounced at lower inlet concentration of benzaldehyde indicating that the occurrence of slow adsorption; however, this shape will be more steepness at high concentration and this will enhance the bed saturation with a contaminant in a short period of time. Also, reductions in the coefficients of mass transfer and/or diffusion may occur at a lower concentration gradient and this will support the low transport of contaminants within the pores of the bed [33], which definitely leads to increase of the saturation time. The values of breakthrough time (t_b) and saturation time (t_s) (opposite to the C/C_0 = 5 and 95% respectively) are increased with the decrease of the inlet concentration and this will effect on the value of adsorbed quantities of contaminant within the bed column q_b and q_s. For example, the decrease of C_0 from 500 to 300 mg/L will increase the t_b from 470 to 750 h for flowrate and bed depth equal to 5 mL/min and 50 cm
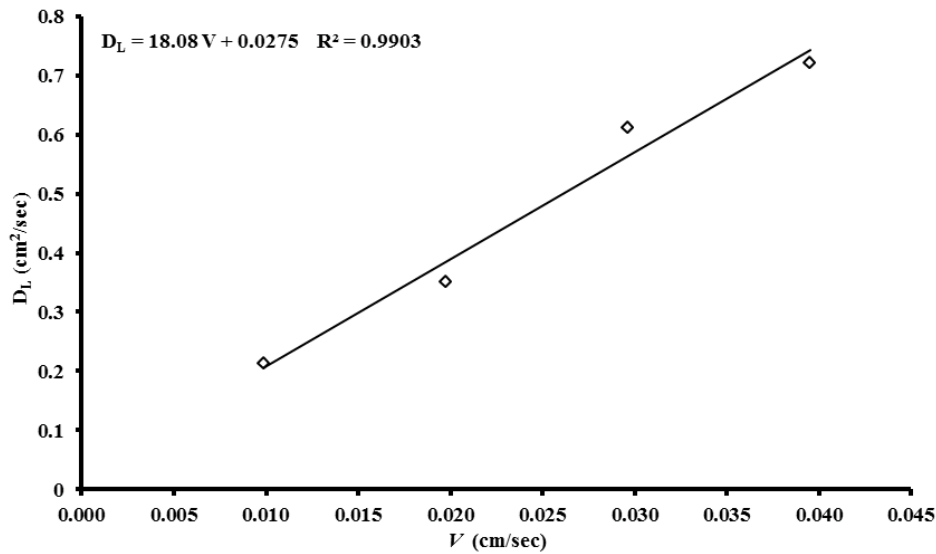
Fig. 2. Longitudinal dispersion coefficient vs. mean pore velocity relation for iron slag bed.

respectively. Conversely, the mass transfer zone can be reduced due to an increase of C_0 and, consequently, this will increase the driving force [34]; however, a similar trend was mentioned in the previous studies [35].

• *Effect of flow rate*: The sorption of benzaldehyde onto granular iron slag was investigated with a flow rate equal to 5 and 10 mL/min for initial concentrations described previously. The measured breakthrough curves of the contaminant uptake for adopted flow rates at different times and locations (i.e., ports) along each packed column are shown in Figs. 3 and 4. It is clear that there is a significant reduction in the breakthrough time due to an increase in flow rate and steeper curves can be observed. This means that the retention time is not sufficient and this causes the escape of polluted water before equilibrium occurrence. Limited diffusion of the contaminant into the sorbent pores at a higher flow rate may be the cause of this behavior [36,37]. The transport of the contaminant from the liquid phase to the zone surrounding the iron slag particles will cause the evolution of the concentration gradient at the solid–liquid interface which pushes the solute to be sorbed through this zone. The efficacy of sorption may be reduced as a result to increase in the water velocity because this will reduce the adhesion between the solute and iron slag [38]. For example, the variation of flow rate from 5 to 10 mL/min will decrease the t_b from 750 to 370 h for C_0 and bed depth of 300 mg/L and 50 cm, respectively.

• *Effect of the bed depth*: Figs. 3 and 4 show the variation of the bed depth from 10 to 50 cm, and the results certified that this increase will improve the bed sorption capacity for solute under the same operational conditions. The increase of bed depth will delay the appearance of the benzaldehyde front; accordingly, this can be increased the breakthrough time. For example, the decrease of bed depth from 50 to 10 cm with cause a significant decrease in t_b from 750 to 195 h for flow-rate and C_0 equal to 5 mL/min and 300 mg/L, respectively.

• *Numerical modeling*: Earth science module, solute transport, saturated porous media, transient analysis in the COMSOL Multiphysics 3.5a (2008) was applied to solve the transport equation. Parameters and constants of the granular iron slag used in the one-dimension simulation (i.e., system dimensions and properties with boundary and initial conditions) are inserted in Table 1. Comparisons between the predicted values by COMSOL and measured results for solute concentrations during the migration of the contaminant plume at different time intervals for a specified flow rate are also depicted in Figs. 3 and 4. A good agreement can be observed between the model predictions and experimental results with a coefficient of determination (R^2) not less than 0.98. So, the present model can be used to predict the propagation of the contaminant front in the field-scale situation when the granular iron slag is used as PRB.

4.2.2. Monitoring of hydraulic conductivity and calcium concentration

Hydraulic conductivities of the iron slag bed packed in the column at specified periods of time were calculated and are plotted in Fig. 5. It is obvious that the hydraulic conductivity was approximately remained constant during the duration not exceeding 200 h for adopted contaminants with an average value equal to 0.00253 cm/s. Accordingly, this period was sufficient to give a clear vision about the variation of hydraulic conductivity coefficient and there is no justification to monitor this parameter along with all duration of each experiment. The stabilization of hydraulic conductivity means that the voids (i.e., areas) prepared for flowing water remained constant because no precipitates are formed within the slag bed.

Figs. 6 and 7 illustrated the variation of calcium content with time as a result of the migration of groundwater contaminated with benzaldehyde at various values of initial
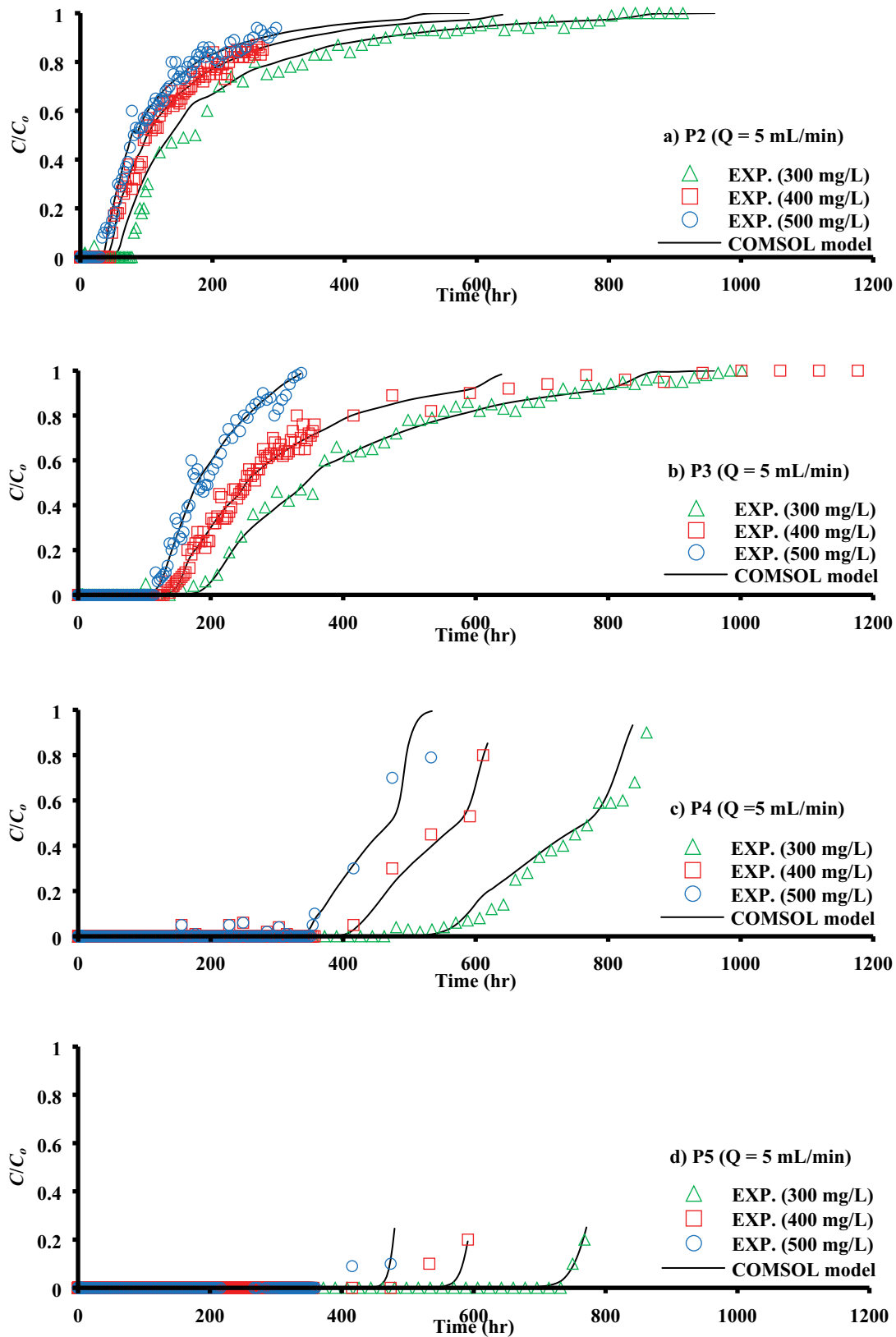
Fig. 3. Experimental and COMSOL predictions of breakthrough curves for the transport of benzaldehyde at different values of initial concentration with flow rate = 5 mL/min.
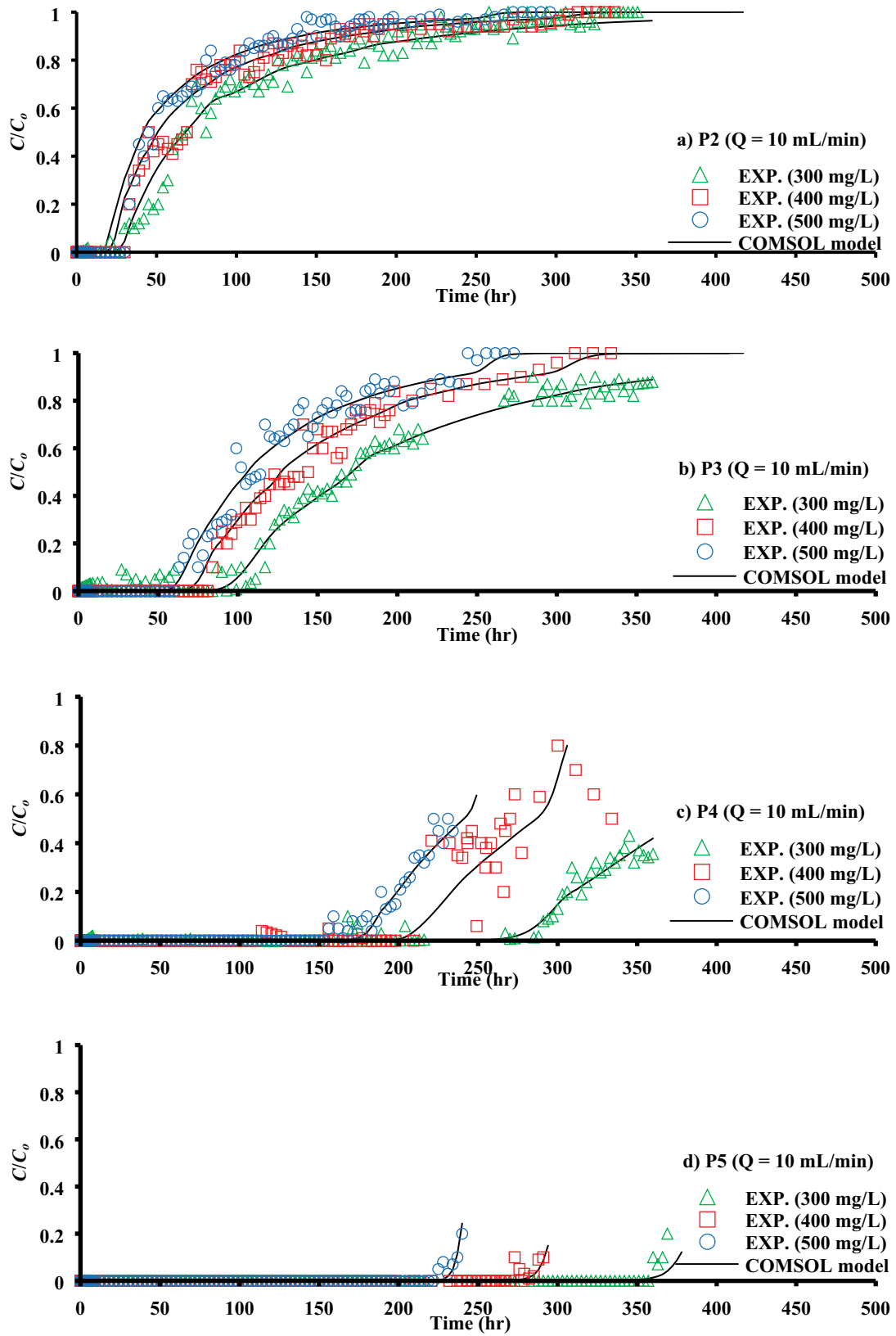
Fig. 4. Experimental and COMSOL predictions of breakthrough curves for the transport of benzaldehyde at different values of initial concentration with flow rate = 10 mL/min.

Table 1
Measured parameters and related constants used in the modeling of C_7H_6O transport in 1D columns

Parameter	Value
Barrier bed depth (cm)	50
Porosity of barrier (n_B)	0.41
Longitudinal dispersivity (α_L, cm)	18.08
Bulk density (kg/m³)	2,026
Initial concentration (mg/L)	Zero
Concentration at $z = 0$ (mg/L)	300, 400, 500
Advective flux $\left(\dfrac{\partial C}{\partial z} \right)$ at $z = 50$ cm	Zero

concentration and bed depth for flow rate values of 5 and 10 mL/min respectively. The dissolution of calcium oxide in the solution and the profile change can describe the adsorption mechanism. The source of calcium concentration in the aqueous solution definitely results from the dissolution of calcium oxide presented in the composition of iron slag with percentage reached to 14% as mentioned previously. These figures signify that the calcium profile is divided into two distinct parts. The first one is remarked by the stabilization of calcium concentration while a clear decrease in the second part as a function of the time can be observed. This behavior can be attributed to the equilibrium phenomena between dissolved calcium oxide and calcium uptake on the surface of the adsorbent for the first part. However, the increase of benzaldehyde concentration as seen in Figs. 3 and 4 correspondings to the second part appeared in Figs. 6 and 7 can be the main reason for the decrease of calcium concentration because the calcium will form a cation bridge between

benzaldehyde and iron slag as mentioned by previous study Faisal et al. [27].

5. Conclusions

The results proved that the iron slag by-product material had a good ability in the elimination of the benzaldehyde from contaminated water and it can be applied effectively in the permeable reactive barrier technology. COMSOL Multiphysics 3.5a software was able to simulate the breakthrough curves of the benzaldehyde front measured at certain locations along the iron slag bed with a coefficient of determination (R^2) greater than 0.98. The lower initial concentration of benzaldehyde (300 mg/L), lower flow rate (5 mL/min) and higher packed depth (50 cm) achieved the highest longevity (i.e., breakthrough time) of the barrier with value reached to 750 h; however, the hydraulic conductivity remained approximately constant around the average value of 0.00253 cm/s. The calcium oxide forms approximately 14% from the composition of the iron slag and breakthrough curves for dissolved calcium proved that there was equilibrium between dissolved calcium oxide and calcium uptake on the iron slag for a duration approximately within the breakthrough time. Thereafter, the calcium formed a cation bridge between benzaldehyde and iron slag; so, this causes a clear decrease in the calcium concentration.
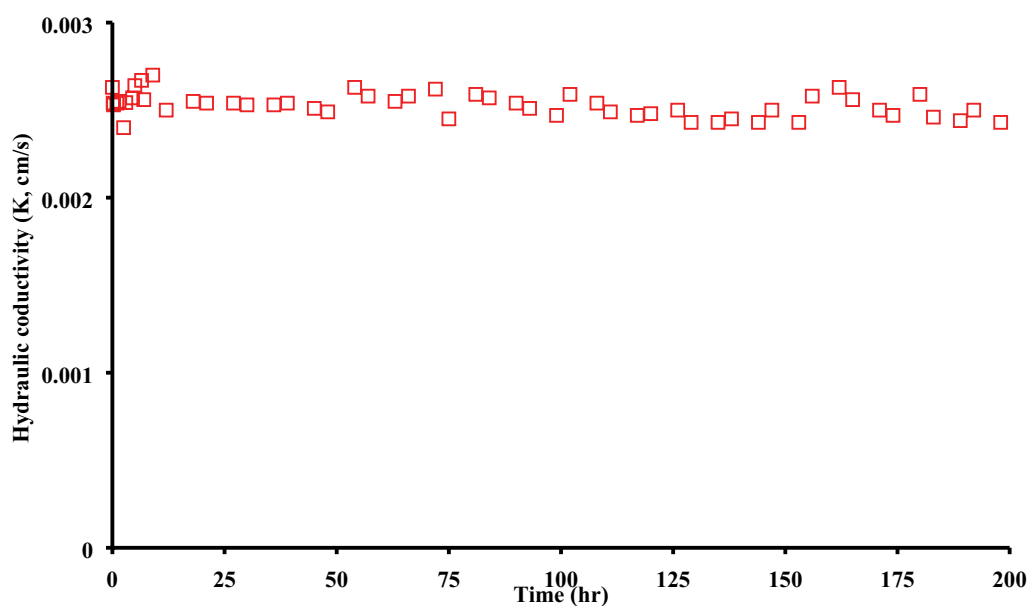
Fig. 5. Variation of hydraulic conductivity as a function of the travel time associated with the transport of groundwater contaminated with benzaldehyde in the columns packed with iron slag bed.
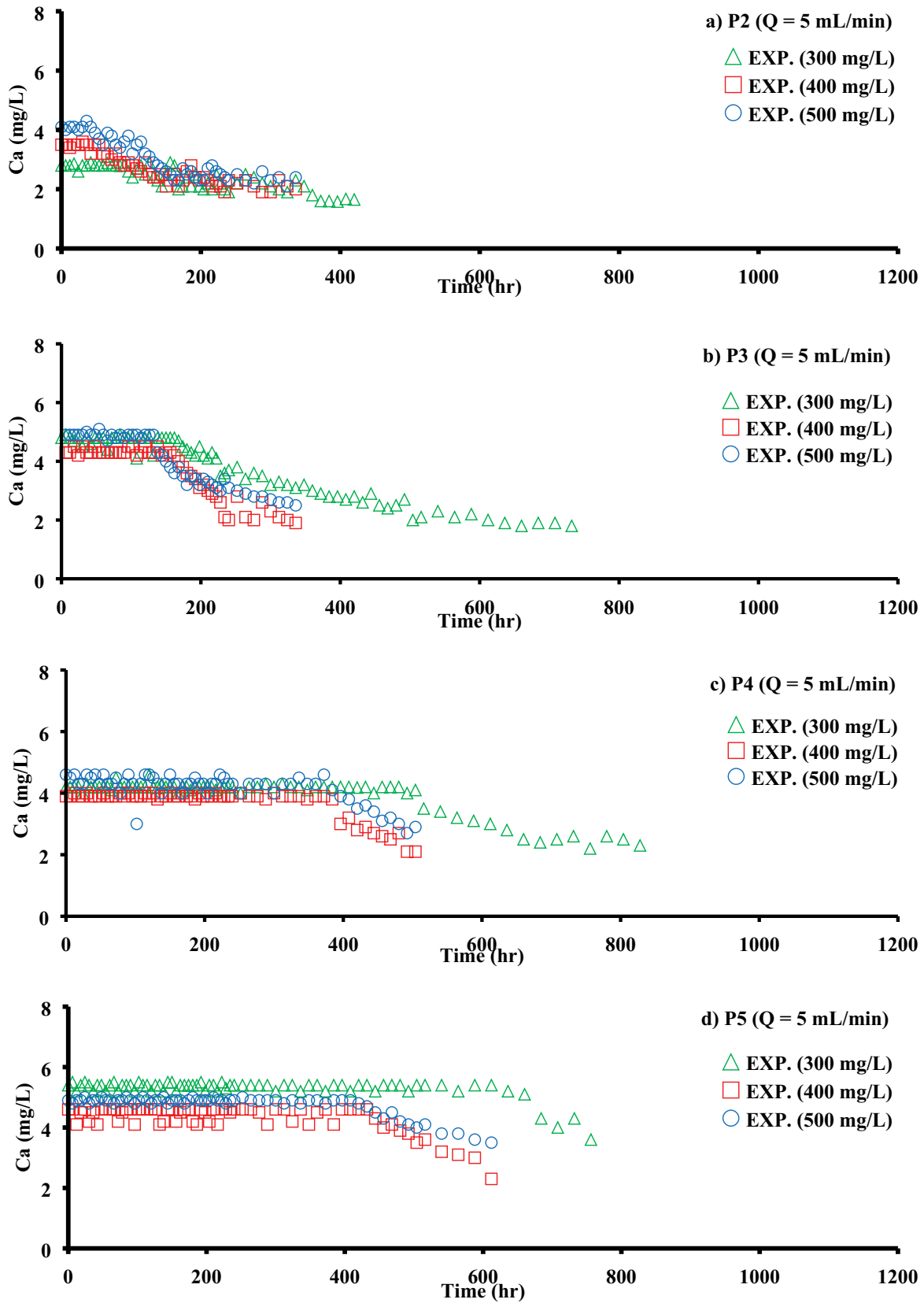
Fig. 6. Variation of calcium concentration dissolved in effluent water as a function of time due to the transport of aqueous solution contaminated with benzaldehyde at different values of initial concentration with flow rate = 5 mL/min.
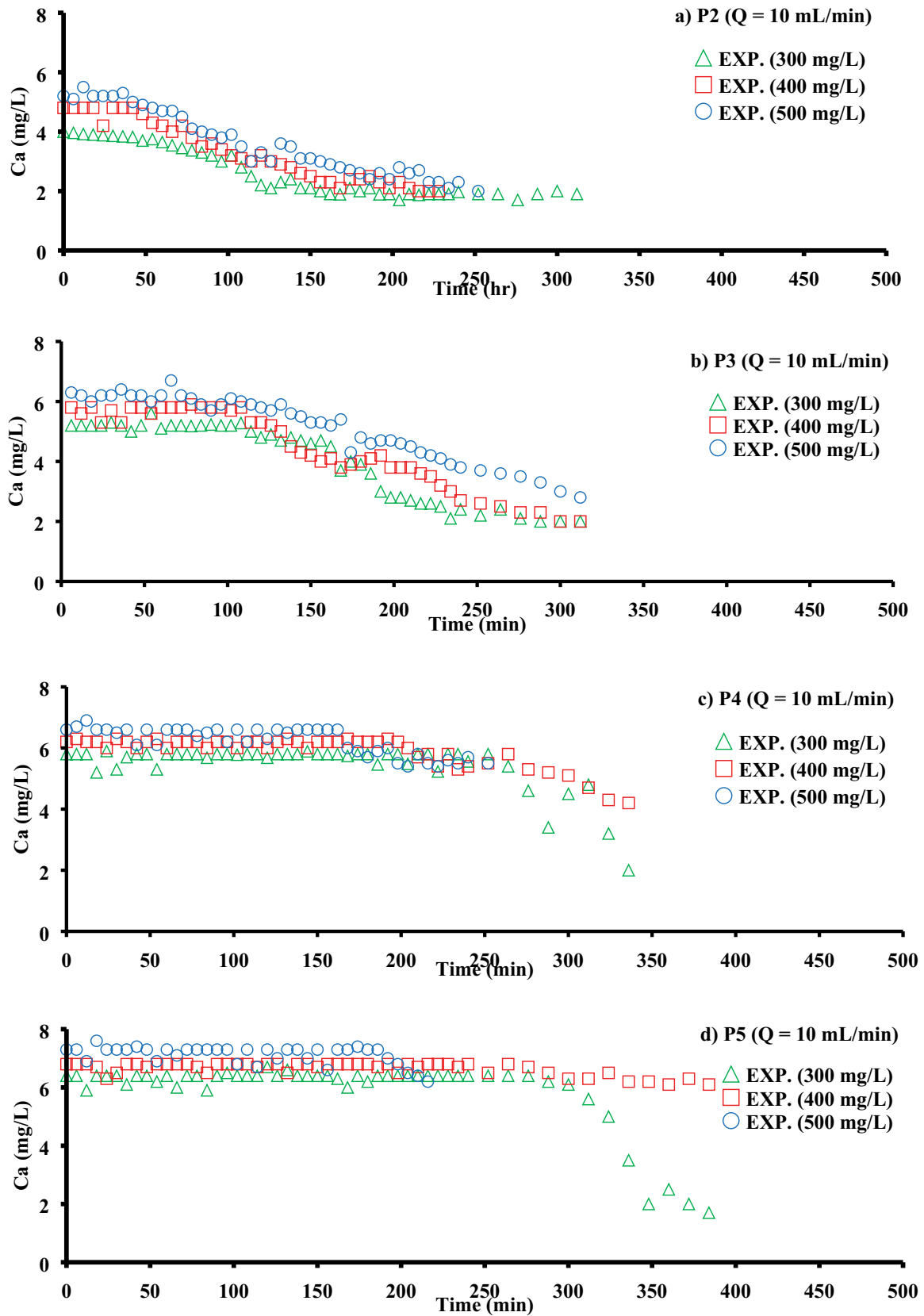
Fig. 7. Variation of calcium concentration dissolved in effluent water as a function of time due to the transport of aqueous solution contaminated with benzaldehyde at different values of initial concentration with flow rate = 10 mL/min.

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