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ORIGINAL ARTICLE

Combined effect of unsaturated soil condition and soil heterogeneity on methylene blue adsorption/desorption and transport in fixed bed column: Experimental and modeling analysis

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Abstract Several series of batch and fixed bed column experiments have been carried out to study the methylene blue sorption and transport in sandy and clay soil under different experimental conditions. The non linear forms of the pseudo-second order kinetic model and the Freundlich and Langmuir isotherm models were used to quantify the maximum adsorption capacity of the used materials. The effects of several experimental factors (flow rate, initial concentration, saturation condition of soil and soil heterogeneity) on the breakthrough curves have been undertaken. Experimental data revealed that the breakthrough curves depend on flow rate, methylene blue inlet concentration, saturation condition of soil and soil heterogeneity. A two dimensional model based on Richards equation and advection–dispersion equation coupled with adsorption model was developed. A comparative analysis between this model and Thomas model showed the effectiveness of advection–dispersion model to describe the experimental breakthrough curves and particularly in an unsaturated heterogeneous medium. The advection dispersion model reproduces perfectly the transfer mechanisms in porous media and seems to be a useful tool to better understanding the physical processes and the effect of capillary barrier of methylene blue transport in unsaturated heterogeneous soil. The result shows also that the soil heterogeneity has a significant effect on methylene blue adsorption through unsaturated layered media. Furthermore, in the interface between two layers with different hydrodynamic proprieties, the adsorption capacity increases with a decrease in kinetic adsorption rate caused by the decrease in pore water velocity.

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1. Introduction

With the fast development in industrial scale, the problem of water and soil pollution has become more serious. The use of organic dyes in many industrial products may threaten the water systems. For instance, the methylene blue (MB) is a cationic dye which is found in many industrial effluents (textile, cosmetic industries, paper and plastic). It is an important contaminant in soil and water bodies and it may induce health problems (Xing et al., 2010). Hence, the removal of MB is fundamental to ensure the non contaminated water supplies. Several adsorbents were used to remove the MB such as the activated carbons (Yang and Qiu, 2010; Foo and Hameed, 2011), Kapok fiber (Liu et al., 2012), chitosan clay composite (Auta and Hameed, 2014), modified bamboo powders (Guo et al., 2014), Natural zeolite (Wang et al., 2006), sludge (Mitrogiannis et al., 2015), swelling clay (Li et al., 2011), char (Makrigianni et al., 2015) and organo-illite/smectite clay (Wang et al., 2013). The activated carbon is the most used adsorbent for dye removal, but it is costly. As an alternative solution, many low cost adsorbents are examined to replace activated carbons. These adsorbents should be easily available and could be regenerated. Sand is used as an adsorbent in removing dyes (Rauf et al., 2008) and the removal of methylene blue (Bukallah et al., 2007; Dotto et al., 2015). Also, the clay minerals are a good adsorbent with low cost which are still receiving attention because of the various applications in industrial scale due to their attractive adsorption proprieties. The adsorption capacities of these clay minerals for dyes depend on the clay proprieties, the adsorbate and the experimental conditions (Şahin et al., 2015). The sorption to subsurface materials is one of the major processes that dominate the dye transport in soil. This process can be performed using batch and column experiments (Auta and Hameed, 2014; Zhang et al., 2011; Hamdaoui, 2006), but the dynamic adsorption systems (column experiments) are preferred because they describe well the dye adsorption capacity (Reza and Ahmaruzzaman, 2015). Sorption is also one of the most important processes which reduce the chemical infiltration in soils, but it isn't the only dominating process that controls the MB migration in continuous fixed bed column. The MB particles move in the pore space of the medium due to advection by the fluid flow and it is dispersed according to the transport process. Therefore, the mass transfer which occurs along the continuous fixed bed column during MB migration includes three processes: (i) fluid flow, (ii) mass transport and (iii) physical and chemical adsorption. Several experimental factors influence the MB infiltration into the soil. Therefore, the breakthrough curves (BTCs) depend on the fixed bed configuration, flow rate, feed concentration, pH, temperature, adsorbent density, and other variables (Reza and Ahmaruzzaman, 2015). Besides these influencing factors, the soil heterogeneity plays an important role in reducing chemical product infiltration. The presence of capillary barrier influences the transfer of both water and pollutant (Winiarski et al., 2013). The adsorption in the vicinity of the capillary barrier increases and the interface between both porous media became an ideal area for particle detention (Prédélus et al., 2015). In unsaturated medium the transport process is more sophistical than saturated medium due to the presence of air in poral space. This can cause more retention in unsaturated soil (Tian et al., 2011). The high retention capacity in unsaturated soil causes retardation in breakthrough curve profile. The retardation factor increases in soil under dry conditions compared to the porous media under moist conditions (Sadasivam and Reddy, 2015). Indeed, the presence of moisture in soil results in an important decrease in the equilibrium adsorption capacity of soil. Moreover, the increase in moisture content in porous media may also decelerate the transport processes of air phase existing in the pore volume because molecular diffusion in water is slower than in air. The modeling of these breakthrough curves is essential to predict the adsorption behavior using parameters obtained from experimental studies. In fact, several models have been proposed for the correlation of breakthrough curve data obtained in dynamic adsorption process. For example, Thomas model (Thomas, 1944) is one among these several models used to describe the solute adsorption in fixed bed column. It was successfully used for the prediction of breakthrough curves and provides good agreement between experimental and calculated data (Uddin et al., 2009). It has been proved that Thomas model is the most suitable one to describe MB adsorption in column experiment on to modified straw adsorbent (Zhang et al., 2011), Leaf powder (Han et al., 2009), jackfruit leaf powder (Uddin et al., 2009) and natural zeolite (Han et al., 2007). However to model the transport in column adsorption, the advection dispersion model was chosen (Sadasivam and Reddy, 2015) to fit the experimental breakthrough curves obtained under different moisture conditions. In this work, a comparative numerical analysis of two models for breakthrough curves modeling has been studied using experimental data. To model the MB transfer in porous media, it is prerequisite to know the hydrodynamics and solute transport parameters. These parameters have been determined by characterizing the porous media used in this study. The advantage of the advection–dispersion compared to the Thomas model is that the different physical processes can be coupled and solved simultaneously. Furthermore, this model coupled with Richard equation and adsorption kinetic equation can be applied to a variety of materials and takes into account the porous media characteristics, the hydrodynamics and the solute proprieties. Another advantage of this model is the capabilities to study the desorption experiments and to evaluate the adsorbed concentration in solid phase.

The main goal of this study is (i) to study the adsorption–desorption and transport behavior of MB in two soils with different properties (sand and clay) using batch and column experiments; and (ii) to test the capabilities of both models to simulate non equilibrium sorption and transport of MB in soils under different experimental conditions.

2. Materials and methods

2.1. Soil and solutions

Both samples (sand and clay) used in this study were collected from an industrial zone in the region of Sousse in Tunisia. The particle size distribution of sand and clay and the exchange capacity (CEC) of clay were measured by laser diffraction particle size analyzer (Microtrac S3500) and by the Metson method (AFNOR NF X31-130), respectively. The mineralogy of the sand and clay samples was established by X-ray