Journal of Air Pollution and Health (Spring 2016); 1(2):69-76

Original Article

Available online at http://japh.tums.ac.ir

EVALUATION OF FORMALDEHYDE REMOVAL FROM CONTAMINATED AIR BY USING A BIOTRICKLING FILTER REACTOR IN A CONTINUOUS CONDITION

Amirreza Talaiekhozani[1], Mohammad Reza Talaie[2], Mohamad Ali Fulazzaky[3]*

[1] Department of Civil and Environmental Engineering, Jami Institute of Technology, Isfahan

[2] Department of Chemical Engineering, Faculty of Engineering, University of Isfahan, Isfahan, Iran

[3] Institute of Environmental and Water Resources Management, Water Research Alliance, Universiti Teknologi Malaysia, 81310 UTM Skudai, Johor Bharu, Malaysia

ARTICLE INFORMATION

Article Chronology:

Received 24 November 2015

Revised 27 January 2016

Accepted 12 March 2016

Published 31 May 2016

Keywords:

Biotrickling filter reactor, formaldehyde removal, mathematical model, air pollution control

CORRESPONDING AUTHOR:

mrtalaie@eng.ui.ac.ir

Tel: (+98 31) 52636319

Fax: (+98 31) 52636320

ABSTRACT:

Introduction: Formaldehyde is a toxic, mutagen and teratogen chemical, and it is suspected to be carcinogenic to humans. As a result, it can be classified as a noxious air pollutant which must be removed from any formaldehyde-contaminated air stream before releasing to the atmosphere. Biological methods, particularly biotrickling filtration, have recently attracted a great deal of attention due to low operating cost and high removal efficiency.

Materials and Methods: In this study, the biotrickling filtration of air stream contaminated by formaldehyde was investigated in a continuous system. The main objective of this study was to investigate formaldehyde biotrickling processes in continuous mode, culminating in the development of a mathematical relation to correlate volumetric mass transfer coefficient versus the Reynolds number of gas flow. During these experiments, formaldehyde removal efficiencies of 97, 97.5 and 96.5% were achieved for the gas flow rates of 90, 291 and 1512 l/h, respectively. Biotrickling filter reactor (BTFR) was employed for this continuous experiment.

Results: The results obtained in this study show that the mass transfer from gas phase to the gas-wet biomass interphase is the controlling step, and the reaction rate is not an important factor in this case for a wide range of gas flow rates and gas velocities.

Conclusions: The analyses demonstrate that external mass transfer is the controlling step compared to diffusion through liquid phase and bio-reactions. The results also show that using smaller but longer beds resulting in higher velocities enhances the mass transfer rate and hence the removal efficiency.

INTRODUCTION

The continuous discharge of a variety of chemicals through wastewater, stack gases, or solid waste has such a serious impact on the environment that it can prevent industries from sustainable development. The aim of this study is to develop a new method to manage and treat contaminated air by using a biotrickiling filter reactor. Formaldehyde (FA) has a wide range of applications in various industries and is frequently

Please cite this article as: Talaiekhozani AR, Talaie MR, Fulazzaky MA. Evaluation of Formaldehyde removal from contaminated air by using a biotrickling filter reactor in a continuous condition. Journal of Air Pollution and Health. 2016; 1(2): 69-76.

released into atmosphere in the form of contaminated air. Formaldehyde is known as a toxic and mutagen compound that can cause malformation of the embryo. Formaldehyde has high solubility in water, which is almost equal to 40 g/L and is prone to appear in drinking waters or even edible foods [1]. Although formaldehyde has a mortal effect on microorganisms, many researchers have made attempts to use biological methods to remove it from waste air [2,12]. It has been proven that biological methods are able to remove formaldehyde under both aerobic and anaerobic conditions. Different mechanisms have been proposed to explain the anaerobic biodegradation of formaldehyde based on the observation of its intermediate products. According to Gonzalez et al. (1999), formaldehyde is first biologically turned into formic acid and methanol. Second, formic acid and methanol are biologically converted to methane and carbon dioxide by microorganisms [1, 13].

Another mechanism was proposed by Oliveira et al. (2004). Based on this mechanism, formaldehyde is transformed into volatile fatty acids, especially formic acid. Then all produced volatile fatty acids are converted to acetic acid. Finally, acetic acidis biologically turned into methane and carbon dioxide under anaerobic condition [1, 14]. Aerobic formaldehyde removal can occur through two possible pathways. The first consists of converting formaldehyde to methanol and formic acid by utilizing the formaldehyde dismutase enzyme ($2HCHO \rightarrow CH_3OH+HCOOH$) produced by particular microorganisms. The second is based on converting the formaldehyde to formic acid by the formaldehyde dehydrogenase enzyme ($HCHO \rightarrow HCOOH$) when the microorganisms are capable of producing the enzymes of formaldehyde dehydrogenase [1]. Both methanol and formic acid can be biologically converted to water and carbon monoxide.

Five different biological methods including biofilters (BFs), biotrickling filter reactors (BTFRs), bioscrubbers (BSs), biomembrane reactors (BMRs) and suspended growth reactors (SGRs) have been developed for treatment of waste air. Among these five methods, BTFRs have been gained more attention due to its advantages such as the ability to treat acidic gases, the capability of controlling pH, long lifetime of its supporting materials and low-pressure drop [3]. BTFR is an economical and environmentally friendly method to remove contaminants that are highly soluble in water. BTFRs have been widely used for removal of volatile organic compounds (VOCs). Traditionally, biofilters are used to treat exhausted gases with VOC concentrations below 10 mg/L [15]. A BTFR is an attached growth biological reactor that is commonly packed with artificial supporting materials. Microorganisms make a biological active biofilm layer on the surface of the supporting materials. Microorganisms for survival need energy, carbon and nutrient sources as well as water. In a BTFR, pollutants are energy and carbon sources. A nutrient solution, which is composed of water and required chemicals, is circulated around biofilms to prepare enough nutrients and water for microorganism activities [3]. Studies demonstrated that highly soluble pollutants in water are removed by two mechanisms in BTFRs [4]. Pollutants are divided into three parts when they are injected into BTFRs. The first part of pollutants is solved into the nutrient solution, which are finally removed by microorganism activities in biofilms. The second part can be directly absorbed by some microorganisms like fungi (Section 2). Only a small part of pollutants can escape from BTFR without a reaction (Section 3) [16].

Although several studies have been conducted on the removal of VOCs from contaminated gases using BTFR systems, the effect of synthetic contaminated air stream (SCAS) velocity on formaldehyde removal has not yet been investigated. Also, a reliable correlation for biotrickling design has not yet been introduced in the literature. This study was conducted (1) to evaluate velocity effect on biological formaldehyde removal in a continuous bioreactor, and (2) to develop a correlation to predict formaldehyde removal efficiency as a function of gas retention time. A BTFR was used for continuous experiments.

MATERIALS AND METHODS

Evaluation of continuous treatment of air polluted with formaldehyde

The present study was conducted with a laboratory-scale BTFR (Fig.1). Small pieces of a polyurethane pipe of 1 cm length and 0.5 cm diameter were used as supporting material. This type of supporting material provides a 90% void space in a BTFR bed. In this study SCAS was produced by bubbling fresh air through a vessel containing 37% formaldehyde solution known as "formalin". As formaldehyde is a very volatile component, a significant amount of formaldehyde was evaporated and transferred into the bubbles of passing air. Therefore, the leaving air stream was saturated by formaldehyde. SCAS flow rate was measured using a flow meter, and then it was sent to the BTFR for biological degradation of formaldehyde. SCAS preparation details were depicted in Fig.1.

As can be seen in Fig.1, the BTFR has four sampling ports (1, 2, 3 and 4) starting from the bottom of the biofilter bed, located at the heights of 5, 15, 25 and 40 cm, respectively. These ports were prepared to collect samples of solution to determine the formaldehyde concentration along the bioreactor bed. Fig.2 shows a photograph of the BTFR. Aerobic granular sludge (AGS) collected from a municipal wastewater treatment plant in Isfahan, Iran, was used to inoculate the BTFR system in start-up stage. The experiments, which were conducted at room temperature (between 20 and 25℃), were designed to examine the effect of gas velocity on removal efficiency of a BTFR. In the continuous system the nutrient solution had constant concentration and was continuously circulated through the biofilter bed [15]. For our experiments, the nutrient solution flow rate was adjusted at 50 l/h by using a peristaltic pump equipped with a mechanical clock switch. This clock allowed nutrient solution to be injected into the bed with a period of 15 min/h.

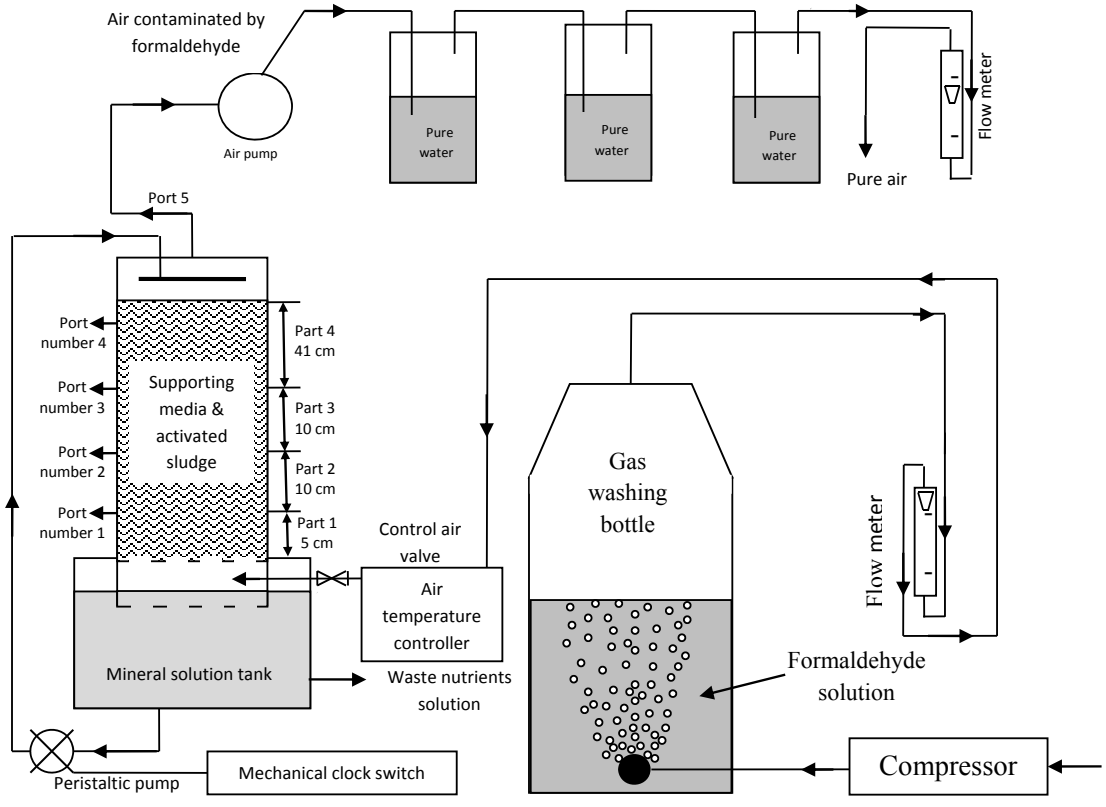

Fig.1. Schematic of the continuous BTFR and synthetic contaminated air stream (SCAS) production systems

In the first stage, the experiment began and continued for 90 days growing microorganisms, which were adapted to consume formaldehyde as the sole source of carbon, on the supporting materials. After passing the microorganisms growth and adaptation stage, the experiment continued for another 80 days, during which the desired experimental data were collected. The aqueous formaldehyde concentrations were determined using chemical oxygen demand (COD) analysis on the nutrient solution. In order to evaluate the effect of gas velocity on BTFR performance, the steady-state formaldehyde removal efficiencies were determined for the volumetric air flow rates (VAFR) of 90, 291 and 1512 l/h. The formaldehyde concentration in the input SCAS remained constant at 490 mg/L during all experiments. In order to determine the formaldehyde concentrations in the gas stream along the biofilter bed, air samples were collected from ports 1 through 4, and were sent to a backwash system using a Champion Air Pump-AAP model vacuum pump (Gardner Denver Pumps, Inc.,) (see Fig. 1). The temperatures of the gas stream at inlet, middle and outlet of the biofilter bed were measured steadily using a digital thermometer. All of the above-mentioned experiments were replicated twice for precision assurance.

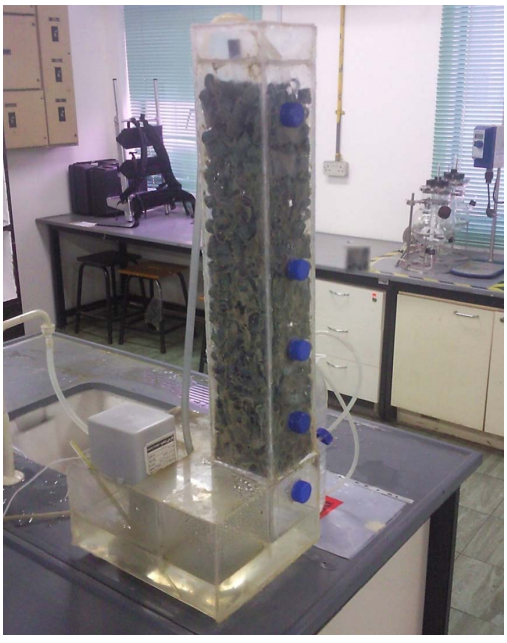

Fig.2. Photograph of BTFR

Analytical methods

The formaldehyde concentration in the nutrient solution was determined by means of COD concentration measurement, which was conducted using a microwave digestion and spectrophotometric method [17]. For this purpose, the Environmental Protection Agency (EPA) method was employed [18]. The treated SCAS from the outlet of the BTFR system was passed through three consecutive bottles containing a specific level of distilled water (see Figs. 3 and 4) for a particular period of time. The pressure head required to pass this stream through these three columns of water was supplied by a vacuum pump (Champion Air Pump-AAP Model) installed at the outlet of this measurement system. In the end, the contents of these three bottles were mixed together, and then the COD of this combined mixture was measured. COD can be converted to the formaldehyde concentration according by the reaction of $CH_2O + O_2$ à $CO_2 + H_2O$ [19]. The above-mentioned COD-to-formaldehyde conversion was confirmed by measuring several formaldehyde concentrations using gas chromatography (GC), via Varian gas chromatograph CP-3800 (Varian, Inc., Walnut Creek, CA, USA).

Biomass concentrations were measured in mg/L of dried solids from each part of the biofilter bed based on the selection of 10 pieces of the supporting material, dried at 104°C for 24 h. The presence of microorganisms in the different parts of the biofilter bed was identified according to a conventional biochemical method [20].

Table 1. Operational conditions of GC to measure FA concentration in the SCAS

Column	DB-WAX 123-7033, 30 m × 0.32 mm, 0.50 μm
Carrier	Helium at 36 cm/s, measured at 35 ℃
Oven	35 ℃ isothermal
Injection	Split, 200 ℃ Split ratio, 1:100
Detector	FID, 300 ℃ Nitrogen makeup gas at 30 mL/min

RESULTS AND DISCUSSION

So far the parameters such as pollutants concentration, gaseous retention time, gas moisture, nutrient concentrations, temperature, biofilter bed type, pH and gas-liquid flow rates ratio have been taken into account as the influencing parameters on the removal efficiency of a BTF [4, 21]. However, the effect of air velocity on BTFR performance has not been fully investigated yet. Fig.3 shows the formaldehyde removal percentage variation versus gas residence time for three different flow rates of 90, 210 and 1512 l/h. Because the BTFR diameter is constant, the immediate effect of gas flow rate variation is the change in gas velocity. The gas velocities corresponding to these three flow rates are 0.0039, 0.0126 and 0.0656 m/s, respectively. This figure reveals that rising gas velocity in BTFR while maintaining residence time increases the removal efficiency significantly. It can be attributed to the turbulence mixing effect which is intensified by velocity rise. This indicates that the gas-phase mass transfer plays an essential part in overall mass transfer rate. Talaiekhozani (2015) reviewed different models introduced in literature and demonstrated the key role of the gas-phase mass transfer on overall mechanism [22]. This conclusion supports the fact that the determination of the external mass transfer rate is an important step in the design of a BTFR for formaldehyde removal from an air stream. In order to determine the contribution of external mass transfer to the removal process, it was assumed that the biological degradation reactions and liquid diffusion are so fast that the formaldehyde concentration at the gas interphase is nearly zero. This assumption leads to Eq. (1) expressing mass balance over an axial differential control volume in the BTFR:

$$C_g.u.A\big|_x - C_g.u.A\big|_{x+dx} = k_g.a\,(C_g - 0)A.dx \quad (1)$$

Where C_g is formaldehyde concentration in gas phase, u is gas velocity, A is BTFR cross-sectional area and $k_g.a$ is volumetric mass transfer coefficient in gas phase. Rearranging Eq. (1) yields the following differential Eq. (2):

$$\frac{dC_g}{C_g} = -k_g.a.\frac{1}{u}.dx \quad (2)$$

Integrating the above equation results in Eq. (3) expressing the relationship between gas-phase formaldehyde concentration through BTFR (C_g) and its respective residence time (t_r):

$$\ln C_g = -(k_g.a).t_r + C \quad (3)$$

Where t_r is retention time and C is a constant. Fig.4 shows the plots of $ln(C_g)$ versus retention time for three different gas flow rates. As can be seen, these plots followed a linear trend. It demonstrates that the assumption of rapid reactions is in line with the fact observed in this experiment.

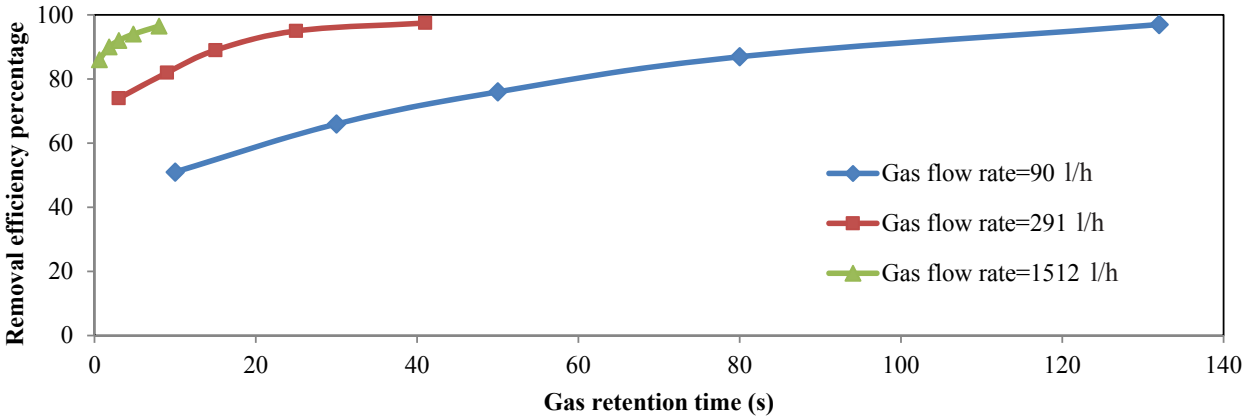

Fig.3. Formaldehyde removal efficiency versus gas retention time in the continuous BTFR for gas flow rates of 90, 291 and 1512 l/h

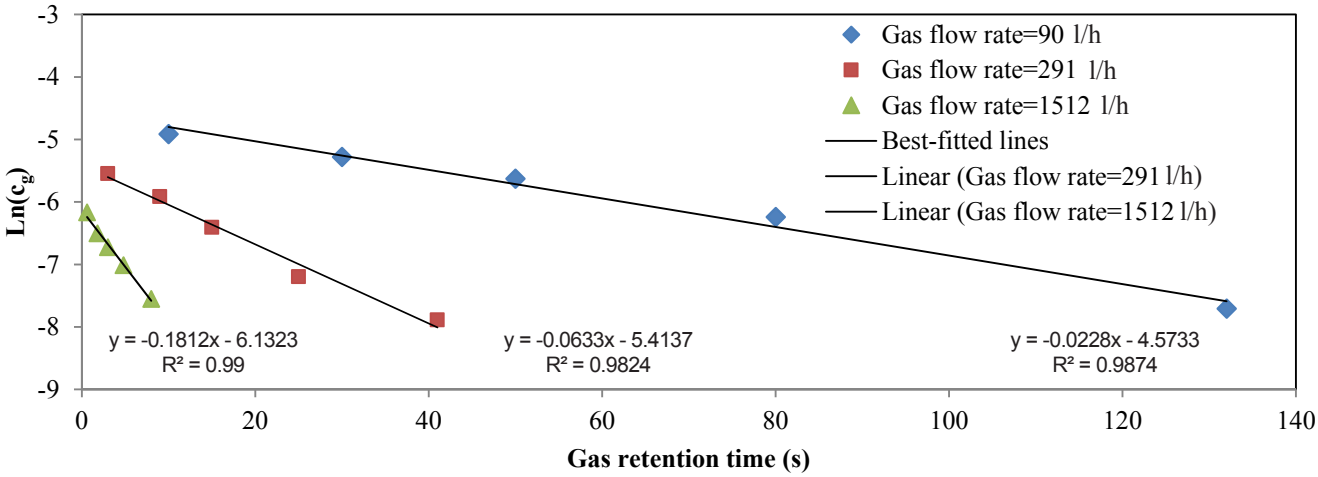

Fig.4. The plot of $ln(C_g)$versus gas retention time

The slopes of these lines are $-(k_g.a)$ at the gas velocities of 0.0039, 0.0126 and 0.0656 m/s, corresponding to the gas flow rates of 90, 291 and 1512 l/h, respectively. Fig.5 shows the plot of $k_g.a$ versus gas velocity. Because mass transfer coefficient is expressed in terms of Reynolds number (Re) rather than velocity, it was correlated to *Re*, and the results are given by Eq. (4):

$$k_g.a = 0.0028Re^{0.728} \quad (4)$$

In the above equation $k_g.a$ is calculated in s^{-1} and Reynolds number was calculated based on superficial gas velocity. The exponent of Reynolds number in the correlations found in literature for mass transfer coefficient ranges from 0.59 to 0.83 for packed beds and empty tubes, respectively [23]. As can be seen, the calculated value of 0.728 for Reynolds number exponent in this study falls within the range of those reported in the relevant literature.

The above equation is a handy tool for sizing BTFR employed for formaldehyde removal.

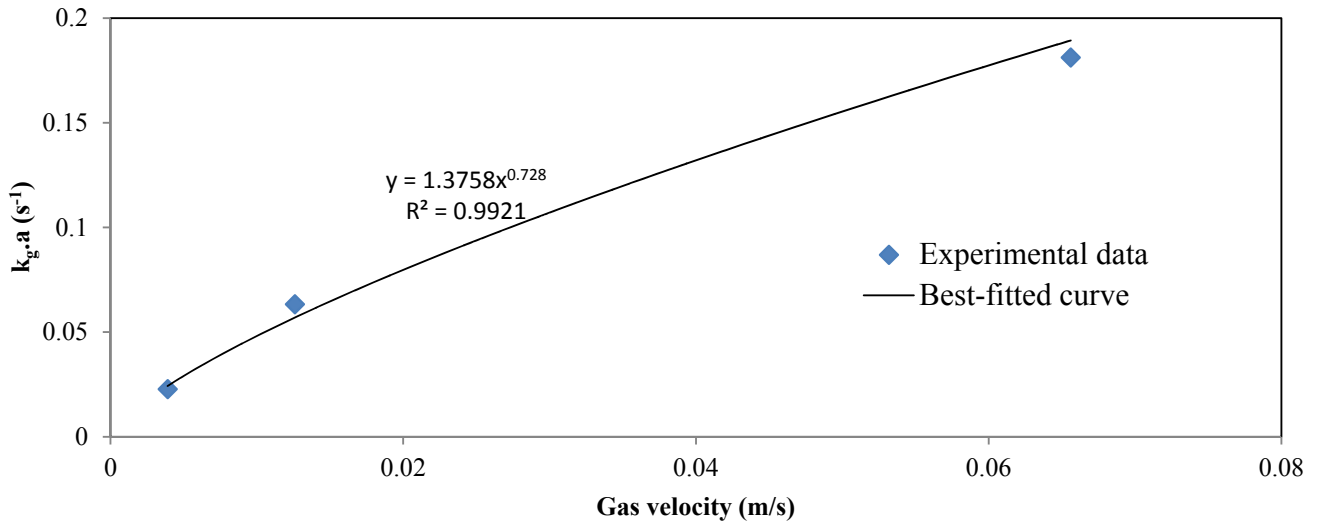

Fig.5. The variation of $k_g.a$ with gas velocity

CONCLUSIONS

In this study, formaldehyde is removed from formaldehyde-contaminated air in a continuous BTFR system. This system shows a high ability to remove more than 95% of the inlet formaldehyde. The analyses made demonstrate that external mass transfer is the controlling step compared to diffusion through liquid phase and bio-reactions. As a result, a correlation was developed to calculate volumetric mass transfer coefficient in a BTFR containing adapted microorganisms. This correlation can be used to design BTFR systems employed for formaldehyde removal accurately, provided the same size and type of packing are used as those utilized in this experiment. The results also shows that using smaller but longer beds resulting in higher velocities enhances the mass transfer rate and hence the removal efficiency. However, the costs are higher due to pressure drop and hence higher energy consumption for driving air blower/compressor. Thus, the economical optimization is required to obtain the optimum size of the formaldehyde-removal BTFRs for a particular application.

FINANCIAL SUPPORTS

UniversitiTeknologi Malaysia (UTM) financially supported this study (Vot. 00H89).

COMPETING INTERESTS

The authors declare that there is no conflict of interest that would prejudice the impartiality of this scientific work.

ACKNOWLEDGEMENTS

The authors gratefully acknowledge financial support from UTM-GUP Grant (Vot. 00H89), Universiti Teknologi Malaysia. We also sincerely thank the Jami Institute of Technology, Isfahan, Iran for providing laboratory facilities.

ETHICAL CONSIDERATIONS

Authors are aware of, and comply with, best practice in publication ethics specifically with regard to authorship (avoidance of guest authorship), dual submission, manipulation of figures, competing interests and compliance with policies on research ethics. Authors adhere to publication requirements that submitted work is original and has not been published elsewhere in any language.

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