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Evaluation of New Type of Synthetic Filler for the Removal of NO_x

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In this study, a novel biological filler was prepared. The physicochemical properties and performances on treating NO_x were investigated compared with corncob and ceramsite in three biofilters. The new filler could provide nutrients for the microbe and could prevent the large fluctuate of pH. Compared with the two other materials, the new synthetic packing material (MCP) had a lowest pressure drop. The maximum removal capacities of NO_x could reach to 2318, 1115 and 624 mg m^{-3} h^{-1} for MCP, corncob and ceramsite filler when the EBRT was not less than 36 s. Besides, the experimental datum were modeled using the modified Michaelis-Menten model and K_s and V_m had been calculated from the model equation.

Keywords: Filler, Biofilter, Biodegradation, Pressure drop, Removal rate, NO_x.

INTRODUCTION

It is widely known that NO_x is one of the main toxic contaminants which could form photochemical smog, cause acid rain and be harmful to health. The NO_x could be removed by traditional methods such as SCR, SCNR, adsorption process and plasma activation methods. However, these technologies have high operation cost and complex operation. In case of the low concentration and high flow rate, the pollutants could be treated by green processes like biofiltration[1]. Biofiltration is good for environment as it does not generate undesirable byproducts and other forms NO_2^- and NO_3^- could be used as an energy source by microorganisms, but in the same time dissolved carbon source are needed[2]. Therefore, suitable fillers should be chose to satisfied the requirements of microorganisms, for most of the contaminations are biodegraded by them. Besides, the fillers also play key roles in air distribution as well as mass transfer[3]. Various investigations of fillers have been studied in biofilters, such as compost, peanut shells, sugarcane bagasse, metal oxides, lava rock and soil[4-7]. However, organic packing materials tend to compact and crack after operating 3-5 years and inert packing materials can not provide nutrients for microbe and could lead to problems if the biomass is excessive on the fillers[8-10]. Thus, an ideal filler should have specific physico-chemical properties like large surface area, suitable particle size, high porosity, good mechanical resistance, good water capacity, high pH buffer capacity and abundant nutrient sources[11,12]. Moreover, it had better contain functional microbial communities to short the start-up period[13,14]. In order to meet the characteristics of ideal fillers as much as possible, synthesized materials is being sought, such as UP20[5] and PVA[11]. For the purpose of overcoming the fillers without functional microorganisms or nutrient sources, a new synthetic packing material has been developed in our laboratory (called MCP)[15] (Fig. 1). It has demonstrated that this filler had a good pH buffering capacity and could provide nutrients, centrally, it contains functional microorganisms. The aim of this work was to test and compare the performance of MCP on NO_x treatment with an organic material (corncob) (Fig. 1)and an inert material (ceramsite) (Fig. 1).

EXPERIMENTAL

The filler (MCP) used in this work contained $CaCO_3$, compost, plant fibre, perlite and cementing materials. MCP filler was prepared as follows: (1) the compost was sieved between 20-40 mesh, then added into microbial communities suspension mixed with nutrients solution. (2) The colonized compost was mixed with the composed of $CaCO_3$, cement and perlite and plant fiber. After that, the mixture was stirred for 15 min. (3) The mixture of dried powders obtained above was added into water glass slowly, keeping on stirring for 20 min. (4) Extrusion had been performed with a special device (self-made). Corncob has been chosen for it is an organic material and has nutrients for microbe. Ceramsite is an inert material,

Fig. 1. Fillers used in this study: (a) MCP; (b) Corncob; (c) Ceramsite

having good mechanical resistance. Fig. 1 shows the packing materials before being packed into biofilters. It is interesting to compare this new filler with organic and inert materials. Table-1 presents some characteristics of the three fillers. The specific surface area and porosity were measured by a specific surface area analyzers (GeminiVI2390).

The experimental equipment used in this work is shown in Fig. 2. Three biofilters (BF_1, BF_2, BF_3) were constructed with PVC pipes (2 m in height and 0.16 m in inner diameter) in the same size. The three biofilters were filled with MCP, corncob and ceramsite, respectively. Each biofilter consisted of three sections and in the bottom was an aqua storage tank. The biofilter was packed with fillers to the height of 40, 30 and 30 cm from bottom to top. The height between two layers was 20 cm to redistribute the gas. There were four sampling ports along the biofilter for pressure and gas sampling. The polluted air was introduced from the bottom of the biofilters. The NO_x was generated by mixing clean air with the productions of $NaNO_2$ with 10 % H_2SO_4 in a static chamber. In the beginning, NO_x contains NO and NO_2, but most of the NO_2 would transform to NO with the passage of time. The NO_x concentration was controlled by the addition of $NaNO_2$. The inlet and outlet

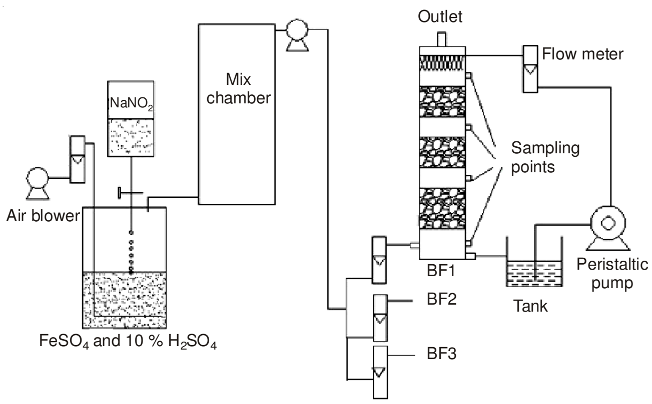

Fig. 2. Schematic diagram of the biofilter system for NO_x treatment

pollutant concentrations were measured using GT901-NO and GT901-NO_2 devices (Shenzhen, China). To maintain the humidity, spraying liquid (tap water without nutrients) was sprayed for 15 min (about 3 L) on the top of the biofilters every day. At the first-four days, the spraying period should be 15 min every 5 h. Besides, the pH of the aqueous phase had not been regulated during all the experiment. The pressure drop was measured using a Testo 510 device (Testo, German). To obtain the advantages of fillers carried the microbe, the three filters were not inoculated with microbe factitiously. And apart from polluted air and materials like MCP and corncob, no other nutritive sources and microorganisms were introduced into biofilters.

RESULTS AND DISCUSSION

Fig. 3 shows the relationship between the pressure drop and the superficial velocity. The results indicate that the pressure datum fit Michaelis-Menten equation pretty well. The correlation coefficients R^2 were 0.983, 0.979 and 0.968 for the biofilters packed with MCP, ceramsite and corncob, respectively. The three fillers showed different degrees of increase in the pressure drop. According to the pressure datum, the biofilter packed with corncob provided the highest pressure drop, with an average value of 9.4 Pa m^{-1} when the superficial velocity was 0.024 m s^{-1}. At the same velocity, the average pressure drops for biofilters packed with ceramsite and MCP reached to 5.8 and 4.5 Pa m^{-1}, respectively. Yang and Allen[16] showed that the pressure drop relied on the biofilter packing procedure. If the porosity of packing materials was little or the size was small, the pressure drop would be greater even for the same superficial velocity. The MCP size was the biggest and the bulk porosity was relatively large, so the pressure drop was small. The bulk density of the corncob (110 Kg m^{-3}) was lightest, but its water holding capacity was strong and it was easy to compact and crack, leading to diminish the porosity, so the pressure was the largest.

TABLE-1
SOME CHARACTERISTICS OF THE THREE FILLERS

	Size (mm)	Water holding capacity (% by weight)	Specific surface area (m^2 g^{-1})	Porosity (%)	Bulk Density (kg m^{-3})
MCP	12 × 20	47	3.28	30	471
Corncob [6]	8 × 10 × 12	70-80	4.11	> 50	110
Ceramsite	10 - 15	< 10	0.22	54	420-460

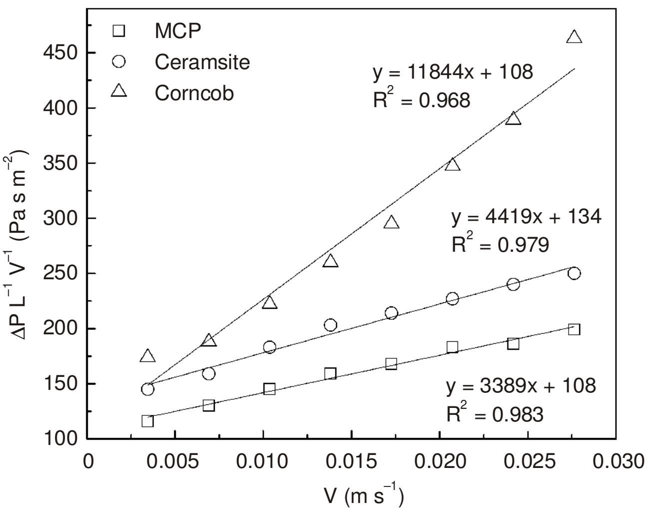

Fig. 3. Pressure drops for the three biofilters

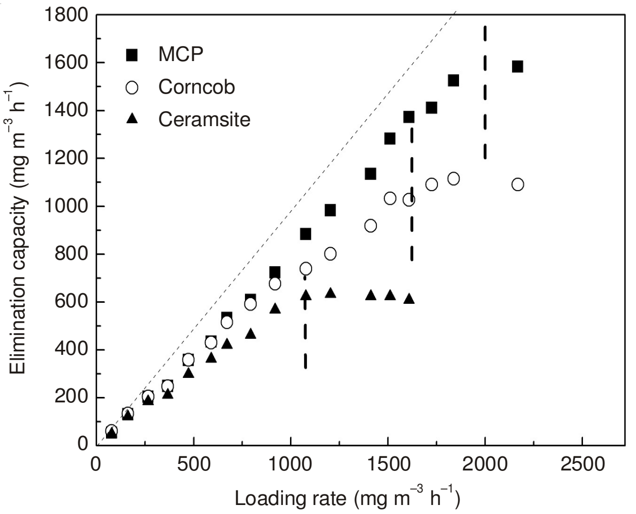

Fig. 5. Elimination capacity *versus* loading rate for three biofilters: EBRT = 36 s

Acidification was an obstacle to the treatment of acidic gas using conventional materials. Therefore, maintaining pH at a suitable level in the process was very important for biofilter's function[17]. Fig. 4 shows the evolution of the pH of fillers *versus* time for the three biofilters. The pH of MCP presented a slow decrease from about 9.5 to 8.7 throughout the 120 d, which provided a good buffer capacity for the biofilter, whereas the ceramsite and corncob biofilters dropped from 6.9 to 3.5 rapidly. The results presented that MCP showed better buffering capacities comprising with ceramsite. The MCP showed alkaline due to presence of sodium silicate and calcium carbonate. The acidity of NO_x introduced into the biofilter would be neutralized by the base of MCP avoiding large pH fluctuations. For the corncob and ceramsite, the acidoid was removed by the metabolism of microbe and the adsorption of water and the acidoid was accumulated.

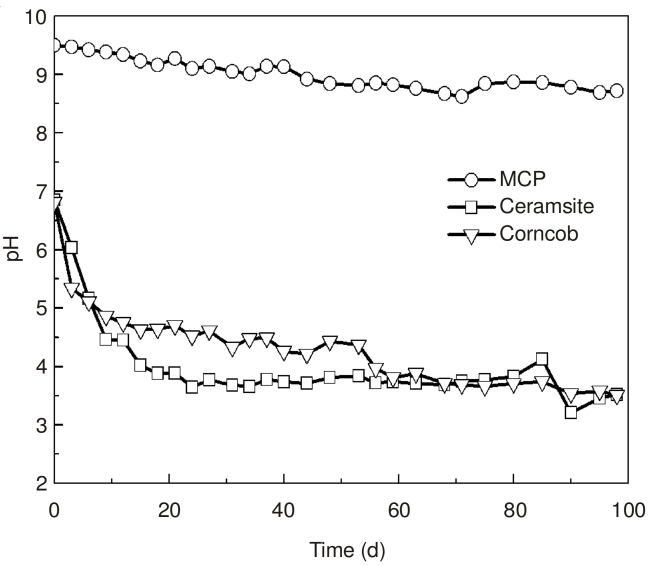

Fig. 4. pH Evolution of packing materials *versus* time for the biofilters

Previously reported parameters [loading rate (LR) in mg m^{-3} h^{-1}; removal efficiency (RE) in % and elimination capacity (EC) in g m^{-3} h^{-1}] were used to evaluate the performance of biofilter. Fig. 5 shows the evolution of the elimination capacity *versus* loading rate for the three biofilters. The results indicated that the highest removal capacity of MCP was about 2318 mg m^{-3} h^{-1} which was higher than corncob (1115 mg m^{-3} h^{-1}) and ceramsite (624 mg m^{-3} h^{-1}). The ratio of NO in the NO_x was much larger than NO_2 by detected. The reason might be that NO_x could react with H_2O and produce NO. However, NO was insoluble in water. Thus, the elimination rate of NO_x in biofilter mostly removed by microorganisms and the datum were modeled using a modified Michaelis-Menten model[5,18]. The treatment of NO_x, the biokinetic constants depended on the microorganisms immobilized on the materials. The relation of 1/R *versus* $1/C_{ln}$ is shown in Fig. 6 and the values of K_s and V_m could be calculated from the kinetic equation. The physical meaning of K_s corresponded to the concentration of H_2S, at which the reaction rate achieve $V_m/2$ and V_m was the maximal removal rate for NO_x. In other words, a filler having a small K_s value presented it had a strong affinity for NO_x. The reverses was also true. K_s could be obtained through the equations and the corncob value was the smallest which showed the adsorption capacity of corncob was the biggest. The V_m of the three biofilters were 0.039, 0.198 and 0.191 g m^{-3} h^{-1} for ceramsite, MCP and corncob, respectively. These mathematical model agreed with the experimental datum.

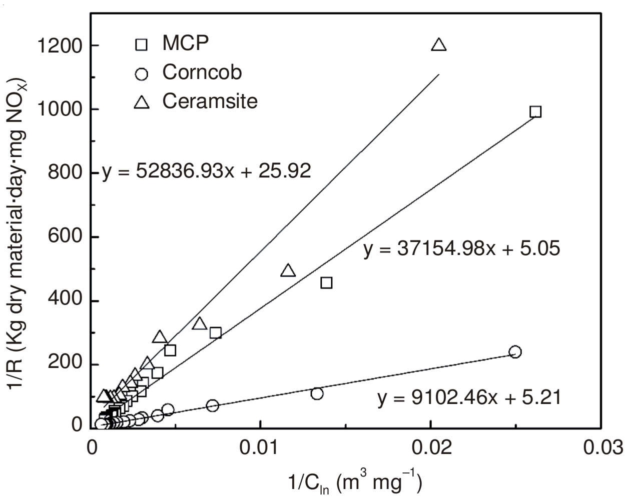

Fig. 6. 1/R *versus* $1/C_{ln}$ according to Michaelis-Menten type model

Conclusions

A new filler was synthesized and was used for the treatment of NO_x. According to the results, the following conclusions could be drawn:

(1) The pressure drop datum suited quite well to the modified Michaelis-Menten equation and the results showed that MCP provided a lowest pressures compared with ceramsite and corncob.

(2) Ceramsite and corncob had approximate neutral pH and MCP generated a basic pH. However, the pH of ceramsite and corncob dropped rapidly after introducing the NO_x and MCP had a better capacity for buffering the pH fluctuation.

(3) MCP was the best packing material to treat high NO_x concentrations. The maximum elimination capacities of MCP, corncob and ceramsite biofilters were about 2318, 1115 and 624 g m^{-3} h^{-1}, respectively.

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