A Novel Integrated Scheme for Stabilizing Biological Oxidation Process Performance in Treating Waste Gas Streams

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Abstract: Most off-gas or treatment streams for VOCs that originate in industrial processes have variable flowrates and transient loadings, which limit the handing efficiency of the biological oxidation processes. For this purpose, an adsorption system was designed and installed in series before the biological oxidation process. Basically, the adsorption system consisted of two fixed beds which are alternately pressurized and depressurized, in which a two-step cycle, i.e., adsorption and desorption was simply achieved. The results obtained revealed that the fixed two-bed adsorption system served successfully as a buffering unit to flatten and reduce the VOC concentration in the treated air which allowed long-term stable performance of the biological oxidation process.

Introduction

Non-methane hydrocarbons (NMHC), usually called volatile organic compounds (VOCs), have been a major health concern due to their adverse effects on both human and the environment. Biological oxidation processes, e.g., biofiltration, have recently emerged as an attractive option for controlling VOCs emissions from industrial processes due to their cost effectiveness (1-3). However, a number of challenges face biological oxidation processes in real application. Typically, most off-gas or treatment streams for VOCs that originate in industrial processes have variable flowrates and transient loading, which limit the handling efficiency of biological oxidation processes (4-9). Nevertheless, the regulatory community generally expects emission controls to be capable of maintaining adequate treatment.

For this purpose, a fixed two-bed adsorption system was proposed to flatten and reduce the contaminant concentration in the treated air (10). In this study, the effectiveness of a novel integrated technology of a fixed two-bed adsorption unit followed by a biofilter, i.e., Trickle Bed Air Biofilter (TBAB) was evaluated. The TBAB system is a known technology that provides consistent nutrient and pH control to optimize the biological oxidation of the waste gas (1, 9, 11). The TBAB system was designed for harnessing the natural degrading abilities of microorganisms to biochemically oxidize organic contaminants into environmentally benign end products such as carbon dioxide and water (12). The two-bed adsorption unit was designed for operating in a two-step cycle, i.e., feeding (adsorption) and purging (desorption), in a fixed bed of adsorbents using gas pressure variation as the principal operating parameter. Regeneration of adsorbents was involved in the same process. Figure 1 shows a typical two-bed with two-step cycle for two-bed adsorption unit in this study.

The experiments were conducted to evaluate the performance of the integrated scheme of an adsorption unit followed by a TBAB under transient loading of toluene. Its overall performance was compared to that of a stand alone TBAB (control).

Experimental Methods

The experimental work was performed on two lab-scale reactors for controlling toluene as a single contaminant. Toluene is a common solvent employed in the industry and a major component in paints and varnishes. One system consisted of a two-bed adsorption unit followed by a biofilter. The other system was a control unit, in which a biofilter was solely operated. The experimental setup is shown in Figure 2.

Adsorption Unit

The system was designed for operation in a two-step cycle, i.e., feeding (adsorption) and purging (desorption) within two adsorption beds. To determine the size of adsorption unit, the design methodology proposed by Moe and Li (13) was accepted for this study. The EBRT in the two-bed adsorption was designed to be 5.6 sec (at air flow rate of 2.22 L min⁻¹) with corresponding total volume of two cylindrical beds of 2.06×10⁻⁴ m³. Each bed was constructed of stainless steel with an external diameter of 2.54 cm (1 inch) and a length of 20.3 cm (8 inch). The beds were packed with bituminous base BPL activated carbon (Calgon Carbon Co, apparent density = 0.85...
g/mL). Freundlich parameters for selected carbon was experimentally determined, $K = 339.2 \text{ (mg/g)/(L/mg)}^{1/n}$ and $1/n = 0.314$ based on isotherm equilibrium adsorption by using the constant volume approach method (14).

Cyclic operation was generated through an electrically operated 4-way solenoid valve (ASCO 8342G 701, Florham Park, NJ), which was controlled by an electronic timer (Digi 42A-120; GRASSLIN Controls Corp., Mahwah, NJ). The cycle duration was set at 8 hours providing that each bed had 4-hour feeding and 4-hour purging. The air supplied to the system was purified with complete removal of water, oil, carbon dioxide, VOCs, and particles by Balston FTIR purge gas generator (Paker Hannifin Corporation, Tewksbury, MA). Liquid VOC was injected via syringe pumps (Harvard Apparatus, model NP-70-2208, Holliston, MA) into the air stream where it vaporized, and entered the equalizing vessel before the adsorption bed for isolating the adsorption bed from unexpected concentration fluctuations in the upstream air supply. Sampling ports were installed for both the feed and exhaust gases. The air valve was installed to introduce the supplemental fresh air within the two adsorption beds. If necessary, it allows gas pressure to reduce in the other fixed bed where desorption occurs.

**Biofilter Unit**

Two independent parallel trickle bed air biofilters (TBAB) were employed. The biofilters were constructed of seven cylindrical glass sections with an internal diameter of 76 mm and a total length of 130 cm (corresponding EBRT of 1.23 min at an air flow rate of 2.22 L/min). Each section was equipped with a sampling port that extends to the center of the column. Each biofilter was packed with pelletized diatomaceous earth biological support media (Celite® 6 mm R-635 Bio-Catalyst Carrier; Celite Corp., Lompoc, CA) to a depth of about 60 cm. The pellets were made from sintered diatomaceous earth and are therefore principally silica (SiO$_2$). Their physical properties were demonstrated in a previous study (15) performed at University of Cincinnati. The biofilter was seeded with an aerobic microbial culture pre-acclimated to toluene, which had been obtained from a previous operation of the biofilter (10). The buffered nutrient solution was circulated by a stainless steel gear pump from a 20 L feed tank through a solenoid operated 3-way valve and back to the feed tank. The feed was sprayed as a fine mist onto the top of the medium bed through a spray nozzle. Details about the formulation of the buffered nutrient solution can be found in our previous publications (9, 12). The biofilters were maintained at a constant operating temperature of 20 °C in a constant temperature chamber and were operated in a co-current gas and liquid downward flow mode.

Since the biofilter performance decreased substantially, coincident with the buildup of back pressure caused by the accumulation of excess biomass within the biofilter bed (15, 16), periodic *in-situ* upflow backwashing was coordinated as biomass control strategy for removing excess biomass in the biofilter. Backwashing was conducted while the biofilter was off line by using 18 L of the buffered...
nutrient solution to induce full medium fluidization at about 50% bed expansion for a defined time period. The duration and frequency for the backwashing were set at one hour once a week. Details about backwashing were well described elsewhere (9, 15).

To simulate the transient contaminant loading in the industry, a square wave change of toluene concentration considered in this study is depicted in Figure 3. Gas phase samples for VOC analysis were taken with gas-tight syringes. These VOC concentrations were measured by using a gas chromatograph (GC) equipped with a flame ionization detector (FID) detector.

Results and Discussion
Experimentally measured desorption profile for transient loading of toluene as single solute is depicted in Figure 4. As shown, the net effect of the
Figure 4. Desorption profiles of two-bed adsorption: The dotted lines indicate the time of cyclic operation of two-bed adsorption unit. Transient loading condition studied consists of 10-hour square wave change of toluene concentration followed by 14-hours starvation without toluene loading a day, which means pure air through the system.

Figure 5. Overall removal performances of integrated unit and control unit with time after backwashing: The data presented are the averages each day (10 hours/day) for three cycles of backwashing, which was conducted once a week.

adsorption system reduced toluene concentration in the treated air, and also the effluent was attenuated below the 250 ppmv, at which the biofilter effectively provided consistent 99% efficiency toluene removal (17). Interestingly, toluene continued to be detected in the effluent even during the period without toluene loading. This effluent response in the adsorption unit would allow stabilizing biofilter performance.

Figure 5 summarizes the overall removal performance of the integrated unit and the control unit with time after backwashing as biomass control. The data presented denotes the average values for a period of three cycles of backwashing. As shown in the integrated unit, no acclimation periods were apparently observed and subsequently over 99% removal efficiencies were consistently attained during the
Overall toluene removal performance without backwashing: The data are the 8-hour average for each day, the two-hour of reacclimation period was excluded in the calculation.

Figure 6. Overall toluene removal performance without backwashing: The data are the 8-hour average for each day, the two-hour of reacclimation period was excluded in the calculation.

experimental period. However, for the control unit, more than 2 hours of acclimation was observed on day 1 when backwashing was conducted, and also no less than 2 hours of acclimation was perceived after 14-hour starvation each day. After acclimation, subsequent removal performance in the control unit fluctuated with removal efficiencies ranging from 85% to 99%. The difference of performance between the two units clearly indicates that the adsorption unit provided an important role in stabilizing the biofilter performance. The biofilter performance was found to be strongly affected by inlet loading conditions (see Figure 5b).

After a period of three cycles of backwashing, the two units continued to be operated without backwashing for the purpose of investigating the effect of backwashing as biomass control on the removal performances of both biofilters. As shown in Figure 6, the overall performances of both units were found to deteriorate with time. Prolonged operation primarily resulted in formation of biomass not actively involved in contaminant removal, and subsequently the biofilter subject to excessive biomass formation faces clogging with decreasing contaminant removal efficiency (15, 16). This indicates that the control of biomass was necessary for attaining stable, long term high removal efficiencies for the biofilter. For this reason, periodic backwashing of the biofilter with medium fluidization was coordinated in this study. It is worthwhile to note that the integrated unit deterioration impact was much less as compared to the control unit due to less contaminant loading on the biofilter in the integrated unit.

Conclusion

A two-step cycle of adsorption and desorption involved in a fixed two-bed adsorption system successfully performed its function as a buffering unit for fluctuating inlet loadings, and subsequently achieved its goal to stabilize the performance of the biological oxidation process. When the integrated unit of a fixed two-bed adsorption followed by a biofilter was coordinated with periodic backwashing as biomass control, removal efficiencies above 99% were consistently attained.

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