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**BIO-OXIDATION OF A MODEL VOC IN AIR**

by  
**Jeong Seop Shim**

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**A Thesis  
Submitted to the Faculty of  
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Master of Science in Environmental Science**

**Department of Chemical Engineering, Chemistry and  
Environmental Science**

**January 1994**

APPROVAL PAGE

BIO-OXIDATION OF A MODEL VOC IN AIR

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## ABSTRACT

### Bio-Oxidation of a Model VOC in Air

by  
Jeong Seop Shim

This study was performed to find a satisfactory regime of operation for the vapor phase bio-oxidation of ethyl alcohol, a model volatile organic compound (VOC), in a specially designed bioreactor. Ethanol was selected as a model compound representing bakery oven gas pollution. A spirally wound bioreactor module was used within which was immobilized a mixed bacterial culture from aerobic sludge. The activated sludge from municipal wastewater treatment plants readily attached with no pretreatment on the surface of the spiral biosupport which is a polymeric sheet.

The parameters studied were air flow rate and inlet concentrations of ethanol. Ethanol was injected, along with air, into a water reservoir prior to being fed into the bioreactor. The vapor and liquid concentrations in the reservoir were measured continuously and reached an equilibrium state. The reaction rates for all runs were determined. An optimal vapor temperature was observed for the environment of the microorganisms.

As expected, the reaction rate varied with air flow rate and vapor temperature. An optimal air flow rate which was used for the action of microorganisms with gaseous ethyl alcohol and oxygen was 20 L/min (retention time: 1.45 min). At this flow rate, the desirable vapor temperature in the reservoir was between 27 °C and 30 °C. At 20 L/min of air flow, a local maximum reaction rate was maintained at about 44 to 50 mg of ethanol per minute for this 28 hour run at this feed injection level. The vapor concentration at the inlet in typical runs from this series at this flow rate reached equilibrium levels ranging

from about 1,000 ppmv to 1,700 ppmv within the first four hours of the 28 hour runs. The air flow rates for this series ranged from 7.52 to 40 L/min, while the total amount of ethanol fed to the system per minute was kept constant.

At higher inlet feed concentrations, the reaction rates increased. For this series, at 2.34 L/min of air flow (retention time: 12.35 min), the maximum inlet vapor concentration reached about 7,000 ppmv within 6 hours. The removal efficiency was 99 percent, equivalent to about 30 mg of ethanol/min due to the low air flow rate. Also, after a 6 hour run at 7.52 L/min of air flow (retention time: 3.84 min), about 6,000 ppmv at the inlet was converted to 24 ppmv at the outlet. The removal efficiency was 99 percent, equivalent to about 87 mg of ethanol/min. This is about 3 times the corresponding rate at 2.34 L/min of air flow.

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This thesis is dedicated to my parents and my wife.

## ACKNOWLEDGMENT

I would like to express my sincere gratitude to my adviser Dr. Sam Sofer for his valuable guidance and encouragement during the entire course of this thesis and special thanks are due to Dr. Richard Trattner and Dr. Barbara Kebbekus for serving as members of the committee of my thesis.

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## CHAPTER 1

### INTRODUCTION

#### 1.1 Bio-Oxidation for Removing Volatile Organic Compounds in Off-Gas

Until recently, air pollution control engineers and scientists have been mainly interested in automobiles and major stationary sources such as refineries and large coating operations for the control of volatile organic compounds (VOCs) as precursors of photochemical air pollution. In ozone build-up areas, the studies on air pollution control are increasingly focusing on smaller stationary sources that emit approximately a few hundred pounds of VOCs per day. As an example, environmental protection regulations for San Francisco Bay and Southern California area now require large commercial bakeries to control gaseous ethanol which is produced from the dough fermentation generated during the baking process [1].

Most off-gases from industrial facilities, waste disposal, and food processing activities have been treated by biofiltration techniques which are generally defined as processes that use compost, peat, bark, soil, or mixtures of these substances as the filter medium. These media serve as support systems for microbial populations. VOCs present in off-gas are trapped in the support material and eliminated biologically as carbon sources for the microorganisms. Biofiltration of contaminated off-gas has been considered a new technology in North America. This is partly because incineration, water and chemical scrubbing, and activated carbon adsorption have been mainly used as air pollution control methods.

Bio-oxidation by using a spirally wound biosupport which is a polymeric sheet, can be considered a new air pollution control technique that utilizes microorganisms, immobilized on the surface of the sheet, to degrade VOCs. As a gas stream passes tangentially through the space which is formed between sheets by winding a sheet, the

pollutants are absorbed or trapped into a liquid biolayer attached on the surface, and oxidized. In this technique, the term "bio-oxidation" is preferred rather than biofiltration because this is not a filtering technique. Thus, this reactor can be named the air bioreactor for bio-oxidation of air pollutants like VOCs. Also, the term "biosupport" is more valid than "biofilter medium".

Typically, compounds which can be easily degraded by bio-oxidation are lightweight, water soluble, and contain oxygen atoms: alcohols, aldehydes, ketones, etc. For aromatic and halogenated compounds, which may be more difficult to degrade biologically, inoculation with specific microbial species, additional nutrients and possibly an additional carbon source may be required [2]. In addition, gaseous VOCs are inherently more biodegradable than solids and liquids because they are molecularly dispersed, and because air has a higher oxygen content than water.

### **1.2 Characteristics of Immobilized Activated Sludge**

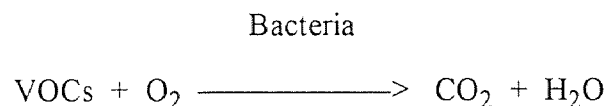
In the last two decades, the development of immobilization techniques has been one of the major changes for bioreactor designs. Using immobilized microorganisms is more advantageous than using free microorganisms because immobilized cells can be much more resistant to high concentrations of toxic chemicals. Immobilization leads to easier recovery and subsequent reuse of biomass [3-5]. Additional advantages such as superior mass transfer at higher cell densities, no washout problems, and capability to operate in a continuous fashion have been demonstrated for the reactors operating on immobilized activated sludge.

Activated sludge from a municipal waste water treatment plant usually contains a variety of microorganisms, which are able to eliminate a broad spectrum of organic compounds. Aerobic degradation of the target pollutants occurs in the biofilm. The biofilm thickness depends upon three factors: biofilm growth due to substrate utilization, sloughing, and decay of biofilm due to bacterial metabolic activity. When the biomass is



attached on the external surface, the biofilm growth is self regulated. The biofilm reaches a critical point beyond which it cannot sustain additional growth. This is due to insufficient diffusive transport of the substrate for maintaining energy. The excess biomass is also removed continuously through a sloughing process. Consequently the removal efficiency of a bioreactor will never be reduced due to excessive growth, and it can be operated over a much longer time [6].

Bio-oxidation relies predominantly on heterotrophic organisms that use organic waste gas constituents as carbon and energy sources. Growth and metabolic activity of microorganisms on the biosupport depend primarily on the presence of dissolved oxygen in the biofilm, the absence of compounds that are toxic to microorganisms, the availability of nutrients, sufficient moisture, and desirable ranges for temperature and pH. The transformation process from the complete biodegradation of VOCs in air can be expressed as follows:



### 1.3 Operating Parameters for Bio-Oxidation in the Air Bioreactor

The performance of immobilized cell bioreactors depends not only on the relevant microbial or enzymatic kinetics, but also on physical process parameters. The reaction kinetics depend on parameters like biomass concentration, dissolved oxygen, and substrate concentration. In order for the bioreactor to operate efficiently, it must be provided with optimal environmental conditions such as moisture, temperature, air flow rates and surface area for biosupport, which make the resident microbial population achieve and maintain high degradation rates.

Maintaining an optimum moisture content on the biosupport is the major operational requirement for the air bioreactor. In case of biofiltration, to prevent the decrease of microbial activity from drying out of a filter material, a moisture content of

between 40 percent and 60 percent is desirable [7]. Pre-humidification used in this study in order to supply water vapor and decrease the drying out rate. Additionally, because the biosupport is hydrophilic, it absorbs a small amount of water.

Bio-oxidation relies predominantly on the activity of mesophilic microorganisms. For optimum results, it is recommended that the vapor temperature in a gas stream be maintained between 20°C and 40°C [1]. In case of higher or lower temperature than in this range, preheating or cooling of the raw gas is required to maintain the optimal temperature before entering a reactor.

In this study, the air flow rates are listed which supply moisture and bring about the best reaction rates which are maintained by choosing the proper retention times. In the spirally wound bioreactor, it is necessary to consider fluid-flow phenomena like laminar or turbulent flow, which depend on air flow rates through the spiral space of the support. "At low velocities fluids tend to flow without lateral mixing, and adjacent layers slide past one another like playing cards. There are neither crosscurrents nor eddies. At higher velocities turbulence appears and eddies form, which lead to lateral mixing [8]." Thus the lateral mixing effect plays a very important role to allow the reaction of oxygen and substrate with microorganisms on the surface of the spiral biosupport.

#### **1.4 Advantages of an Enclosed Reactor with a Spiral Biosupport**

The air bioreactor developed in this study can be used in a flow-through configuration which offers many advantages. Unlike the bed or column type reactor, this configuration allows less room for channeling. Diffusion limitation is restricted largely to the biofilm attached to the biosupport, as opposed to gel-immobilized systems where internal diffusional resistance also plays a role. Oxidation rates of VOCs from industrial waste air vary from fractions of a second in incinerators to seconds in chemical scrubbers to minutes or days in biofilters. The rapid reactions such as incinerators and chemical scrubbers require fuel, chemicals and maintenance. In slower, inexpensive reactions, biofilters require large

reactor volumes and bed areas (e.g. soil or compost beds) [9]. The spirally wound biosupport, however, has a high surface area and high porosity which allows higher biomass loading, and hence smaller reactor volumes. Bio-oxidation in the air bioreactor requires residence times in the order of minutes.

A closed system bio-oxidation reactor is appropriate where minimum maintenance is required and temperature and humidity can be controlled. Also, monitoring of the effluent is easier than in an open system such as a large filter.

## CHAPTER 2

### LITERATURE SURVEY

H. Brauer [10] states that biological purification of waste gases is a more economical process than conventional chemical reactions which generally require elevated temperatures and pressures. This reason is that most of the microbial oxidation occurs at normal temperatures and pressures. Additionally, the oxygen necessary for microbial transformation of the waste components is normally present in the waste gas and is easily introduced into the water phase without any great expenditure of energy.

Ottengraf [7] mentions that the appropriate composition of the solid phase such as peats, composts, etc. and the viable organisms present in the waste gas prevents aging of microorganisms and causes them to maintain a relatively high activity during a long period of time (years).

The effective biofilm thickness for biodegradation depends on transport phenomena such as the mass flow of degradable components into the filter material. The components are absorbed into the water phase of the biofilm and diffuse into the water layer of the biolayer in which a concentration gradient is created. During the diffusion process, at a certain depth in the biolayer no components are left and the degradation process stops. The effective biofilm thickness is the aqueous portion in which biological activity is present. Thus it is always less than or equal to the thickness of the real water film [11].

Rakesh Govind *et al.* [12] developed a novel biofilter for aerobic biodegradation of VOCs and observed the major differences between the novel aerobic biofilter and other immobilized cell bioreactors such as typical packed beds containing support materials. One of the differences is that the straight passes by the flow of gas/liquid phase of their reactor enable the excessive biomass growth to leave the novel biofilter, in comparison to packed

beds, due to the shear forces exerted by the flow of liquid through the straight passages, minimizing the pressure drop.

Rittmann *et al.* [13] described a variable-order model of bacterial-film kinetics which provides an explicit analytical solution for a fixed biofilm flux. It is a mathematical approximation based on a verified, fundamental model that includes liquid-layer mass transport, Monod kinetics, and biofilm molecular diffusion.

Jennings *et al.* [14] developed a mathematical model for percent removal of a pure, nonabsorbable, biodegradable substrate in a submerged biological filter using the nonlinear Monod expression for the substrate utilization rate. One of their conclusions is that the approximation by first order kinetics of the nonlinear Monod expression yields more accurate results over a much wider range of bulk concentrations than that of zero order kinetics. Ottengraf [15] remarked for Jennings' mathematical model that in a zero order analysis they neglected a reaction-free zone in the biolayer which can occur at sufficiently high substrate utilization rates.

Ottengraf and Van Den Oever [16] have developed a model for biofilters, in which the biodegradation in the biolayer is expressed by diffusion transport with a zero-order reaction (i.e. in which degradation velocity is independent of the concentration of the components to be degraded.) They also mentioned that the mass transfer rates in the gas-phase filter beds are much higher than in liquid-phase filter beds. The reason is that the interface resistance in the gas phase can be neglected by assuming the equilibrium state in the concentration between the biolayer interface and the bulk gas.

Ottengraf *et al.* [17] carried out experiments for the biological elimination of volatile xenobiotic compounds such as aromatic compounds and chlorinated hydrocarbons (e.g. 1,2-dichloroethane, xylenes, styrene, methyl acrylate, dimethylformamide, acrolein) in biofilters. From their analysis they concluded that the biodegradation of all the xenobiotic compounds investigated corresponded to zero-order reaction kinetics. In spite

of this fact, they stated that the elimination capacity of a continuously operating biofilter bed depends on the both organic load of the filter bed and the gas flow rate.

For the removal of dichloromethane from waste gases using a biological trickling filter, Diks and Ottengraf [18,19] developed a simplified model, the "Uniform-Concentration-Model", which was shown to predict the elimination performance close to the numerical solutions of the model equations at various conditions. They also found from experimental and theoretical results that the relative flow direction (e.g. co-current and counter-current flow) of the mobile phases did not significantly affect the performance in the trickling filter system.

Arora and Umphres [20] reported that biofiltration of waste water consistently removes BOD and often more than at design predictions. This was from the evaluation results of activated biofiltration and activated biofiltration/activated sludge technologies for 19 treatment plants.

As two principal types of biological deodorization processes of odorous air, Pomeroy [21] described bodenfilters with composts or soils and packed towers similar to trickling filters. As a third deodorizing device, he also mentioned an activated sludge tank, through which a waste air stream is blown.

Kampbell *et al.* [22] studied the removal of a mixture of light hydrocarbons, primarily propane, n-butane and isobutane, which result from the venting of propellant gas injected into product containers during the filling process, using a prototype soil bioreactor. They reported that the total hydrocarbon removal starting at concentrations below the half-saturation constant, resulted in a Lineweaver-Burk plot, and could be described by first-order kinetics. At higher substrate concentrations, the biodegradation rate is limited by the microbial activity to metabolize the organic compounds. Thus the rate becomes independent of the organic concentration (zero-order kinetics).

Prokop and Bohn [23] used a soil bed system to control high intensity odors from a rendering plant. They indicated that it was at least equivalent and probably superior to

other known odor control methods such as incineration and wet scrubbing. They also concluded that it was accomplished at significantly less cost to install and operate.

Bohn [24] proposed that the soil or compost filter for the removal of malodorous gases provides sorptive surface, structural support, nutrients and water for microorganisms. According to types of filter materials and the properties of influent gases, he mentioned related problems and suggested the ways to resolve them.

In 1966, Carlson and Leiser [25] studied the mechanisms of action on and design of soil beds to control sewage odor problems which are prevalent during warm summer temperatures, by long detention times in transit, by lack of aeration and by the close proximity of residences to sewer system vents. They suggested a greenhouse shelter and a buffered irrigation system to provide better control of environmental conditions in the soil filter.

V. Utgikar *et al.* [26] developed a mathematical model describing the steady state biodegradation of VOCs on activated carbon in a biofilter. They demonstrated the use of the model in design of the biofilter for a given load of leachates.

## CHAPTER 3

### OBJECTIVES

The primary objective of this study has been to find a satisfactory regime of operation for the bio-oxidation of gaseous ethanol, a model VOC, using a specially designed, spirally wound bioreactor in which the activated sludge has been immobilized on the surface of a polymeric sheet. The specific objectives are :

1. To evaluate the capability of the bioreactor to eliminate high concentrations of gaseous ethanol under constant flow rates,
2. To demonstrate the importance of flow rate for maximizing reaction rate,
3. To determine an optimal air flow rate for bio-oxidation while ethanol is fed at constant rate,
4. To find a region in which bio-oxidation depends on zero-order reaction kinetics when the substrate concentration is increased to a level above the critical load,
5. To determine the maximum reaction rate at an optimal air flow rate by varying the injection concentration of ethanol,
6. To observe the effect of vapor temperature on the performance of microorganisms in the air bioreactor, and
7. To record the equilibrium ethanol concentrations of the gas and liquid phases in the water reservoir.



## CHAPTER 4

### MATERIALS AND EXPERIMENTAL METHODS

#### 4.1 Microorganisms and Immobilization

16 Liters of activated sludge were obtained from the Parsippany Troy Hills Water Pollution Control Plant (NJ). The sludge was sieved through a 297  $\mu\text{m}$  opening screen, and washed once with 0.5% saline. The sludge was acclimated with 6 ml of pure ethanol with constant air bubbling for one day.

Dry biomass weight of the activated sludge was determined by drying eight samples taken from the washed activated sludge in the oven at 120 °C for 24 hours. In this experiment, total weight attached on the spirally wound biosupport which was a polymeric microporous sheet was 105 g of dry biomass.

Total volume of the washed activated sludge with tap water was 71 liters. It was recirculated overnight to attach the biomass on the bioreactor, using a water pump and 1.5 psi of water pressure.

#### 4.2 Target Compounds and Analytical Methods

To make an artificial waste gas, 95% denatured ethanol, one of the volatile organic compounds found in bakery oven gas, was primarily used in this experiment. Isopropanol and ammonium hydroxide, from which offensive odors emanate, were also used temporarily only to confirm whether their odors can be removed by the spiral biosupport. It was determined only by the sense of smell that deodorization took place.

Ethanol in vapor was sampled with a gas tight syringe from each sampling glass holder at the inlet and outlet of the bioreactor, shown in Figure 1. Two ml in vapor samples were taken. The gaseous ethanol was analyzed on a Perkin Elmer Model 8500 Gas Chromatograph. A stainless steel column (6' x 1/8", Supelco) packed with 1% SP-

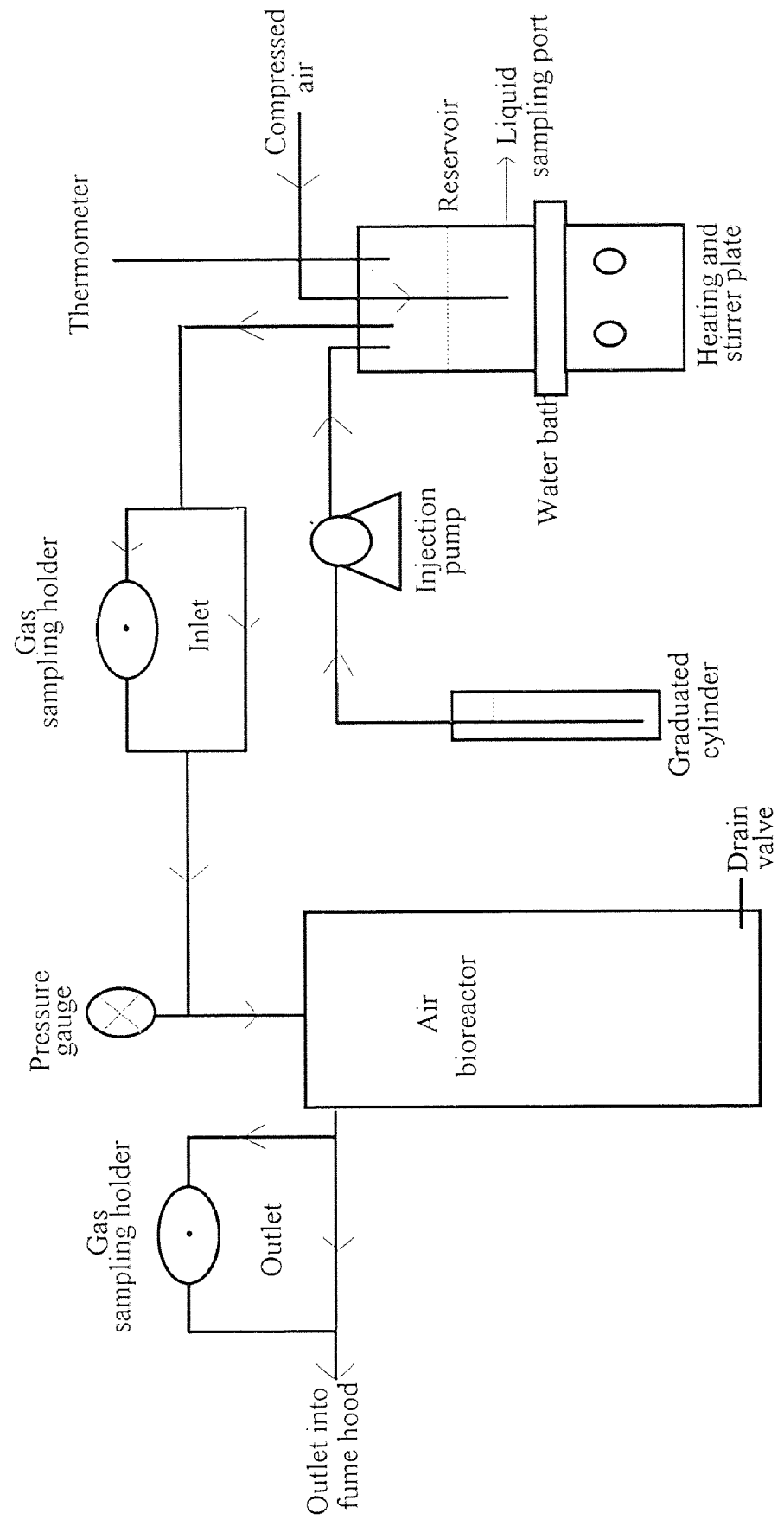


Figure 1 Experimental Setup of the Air Bioreactor for VOC Control in Air

1000 TM on 60/80 mesh Carbopack® B was used at an oven temperature of 140 °C. The detector used was a flame ionization detector (FID), and helium was used as a carrier gas.

#### 4.3 Polymeric Microporous Biosupport for Bio-Oxidation

The physical and chemical properties of the polymeric microporous sheet used as a biosupport in this study are as follows: 60-65 % porosity, 0.4-0.6  $\mu\text{m}$  pore size, hydrophilic in character, and 55 % silica, 45 % PVC and carbon (trace) in composition [6].

The polymeric sheet used to attach the biomass was wound in a spiral configuration with a spacing of 0.25 inches. The ribbed sheet was 20 feet long and 29 inches wide. The spirally wound sheet was sealed at the edges with two round plastic epoxy plates which gave it mechanical strength and rigidity. Each plate was 10.5 inches in diameter. Finally, the reactor was enclosed in a plastic cylindrical body.

Total inside volume of the bioreactor was 41 liters. The capacity to fill the reactor with water was 30 liters because the volume of 11 liters was occupied by the wet polymeric sheet. The volume of the spirally wound space, the space to oxidize VOC by the biomass attached on the sheet surface, was 28.9 liters. This was considered to be the true volume for bio-oxidation. Total surface area of the sheet was 13,920 square inches (about 9  $\text{m}^2$ ), considering both sides of the sheet. The available surface area for immobilizing the biomass was 13,055 square inches (8.42  $\text{m}^2$ ), since the outermost area of the cylindrical wrapped sheet remained essentially unavailable for use.

Supposing that the total biomass was distributed uniformly on the sheet surface, the biomass density was 8 mg of dry biomass per square inch of sheet surface (105 g of dry biomass per 13,055 sq. in. of the available polymeric sheet surface).

#### 4.4 Experimental Setup of the Air Bioreactor for Input VOC Control

Experimental setup of the air bioreactor is shown in Figure 1. The sampling holders made with glass are installed to take gas samples at the inlet and outlet, before and after the reactor, using a by-pass.

To make artificial waste gas and pre-humidified feed, a reservoir of 2.34 liters in size was used. Also, a heating and stirrer plate was used to control the vapor temperature, by heating the reservoir in an aluminum container for the water bath. Initial water volume in the reservoir was kept at about 1 liter, and the initial concentration of ethanol in the liquid was 2,000 mg/L in most experiments. For the continuous injection of ethanol, a peristaltic pump was used at 1 ml/min throughout this study.

Before starting every experiment, the initial solution in the reservoir was heated and purged with air for about 30 minutes until a constant vapor temperature could be maintained. Thus, the initial concentration of ethanol in the reservoir was reduced from 2,000 mg/L to approximately 1,600-1,800 mg/L. The reduced concentration showed slight differences according to the air flow rate and the heating temperature. To vaporize ethanol and water in the reservoir and supply oxygen for microorganisms, compressed air was purged through sintered glass tubes in the reservoir liquid.

#### 4.5 Operating Parameters of the Air Bioreactor

In this study, operating parameters are the air flow rate, the injected ethanol concentration and the vapor temperature at inlet. The air flow rates and the retention (or contact) time for bio-oxidation are shown in Table 1.

The vapor temperatures used were 23°C, 27°C and 30°C. Also, small changes of the vapor temperature were experienced when using different air flow rates.

The feed mixture of ethanol and water was injected into the reservoir, ranging from 25,000 mg/L to 120,000 mg/L. The ethanol solution from a covered graduated

cylinder was continuously injected at 1 ml/min until the vapor concentration in the reservoir reached a steady state level in the gas phase.

**Table 1** The Retention Time of the Vapor Stream in the Bioreactor

Air flow rate (L/min.)	Retention time (min.)
2.34	12.35
4.76	6.07
7.52	3.84
10	2.89
20	1.45
30	0.96
40	0.72

( The operating volume of the bio-oxidation in the reactor is 28.9 liters. )

## CHAPTER 5

### RESULTS AND DISCUSSION

#### 5.1 Effect of the Air Flow Rate for Bio-Oxidation

When the air flow rate was increased, equilibrium gaseous ethanol concentrations at the inlet of the reactor were decreased accordingly, as shown in Figure 2. All data are the values of the equilibrium state of each experiment, resulting from separate runs.

For 50,000 mg/L of ethanol feed concentration, the increase of air flow rate from 7.52 L/min to 40 L/min showed that the inlet vapor concentration was decreased from about 2,100 ppmv to 540 ppmv (Figure 2).

For 100,000 mg/L of injected ethanol concentration, the inlet vapor concentration was reduced from about 7,000 ppmv to 4,200 ppmv when the air flow was increased from 2.34 L/min to 7.52 L/min. For this series of experiments, Figures 3 and 4 show that the vapor concentration at the inlet was reduced to much lower concentrations upon bio-oxidation. The removal efficiencies are over 99 percent.

Figure 5 shows the reaction rate at different air flow rates. The maximum reaction rate for this series was about 60 mg of ethanol per minute at 7.52 L/min of air flow. In this case, the increase in the air flow rate shows the increase of the bio-oxidation rate at the same conditions, and this can be due to two reasons. First, the amount of gaseous ethanol carried by higher air flow rates is increased. Secondly, at higher velocities in the spirally wound sheet, the turbulent flow leads to a mixing effect that can cause the microorganisms on the surface of the sheet to be exposed to ethanol and oxygen more rapidly. In other words, the bio-oxidation rate can be increased by the more rapid and efficient mass transfer between the biofilm and the gas stream until the air flow rate reaches an optimal point. At this point, the absorption of feed and oxygen into, and the production of CO<sub>2</sub> out of, the biofilm are accelerated due to the turbulent air flow.

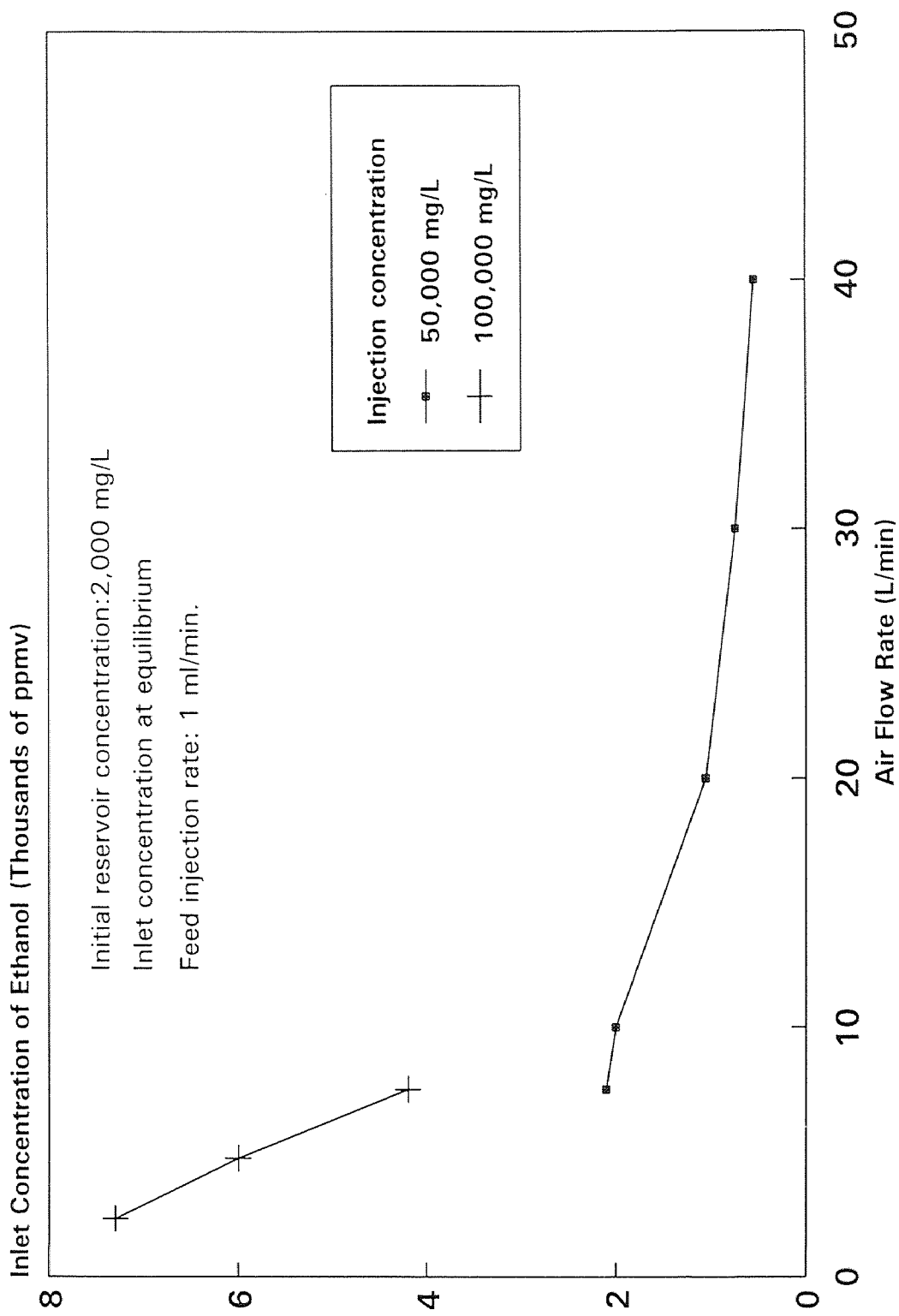


Figure 2 Variation of Inlet Vapor Concentration at Different Air Flow Rates

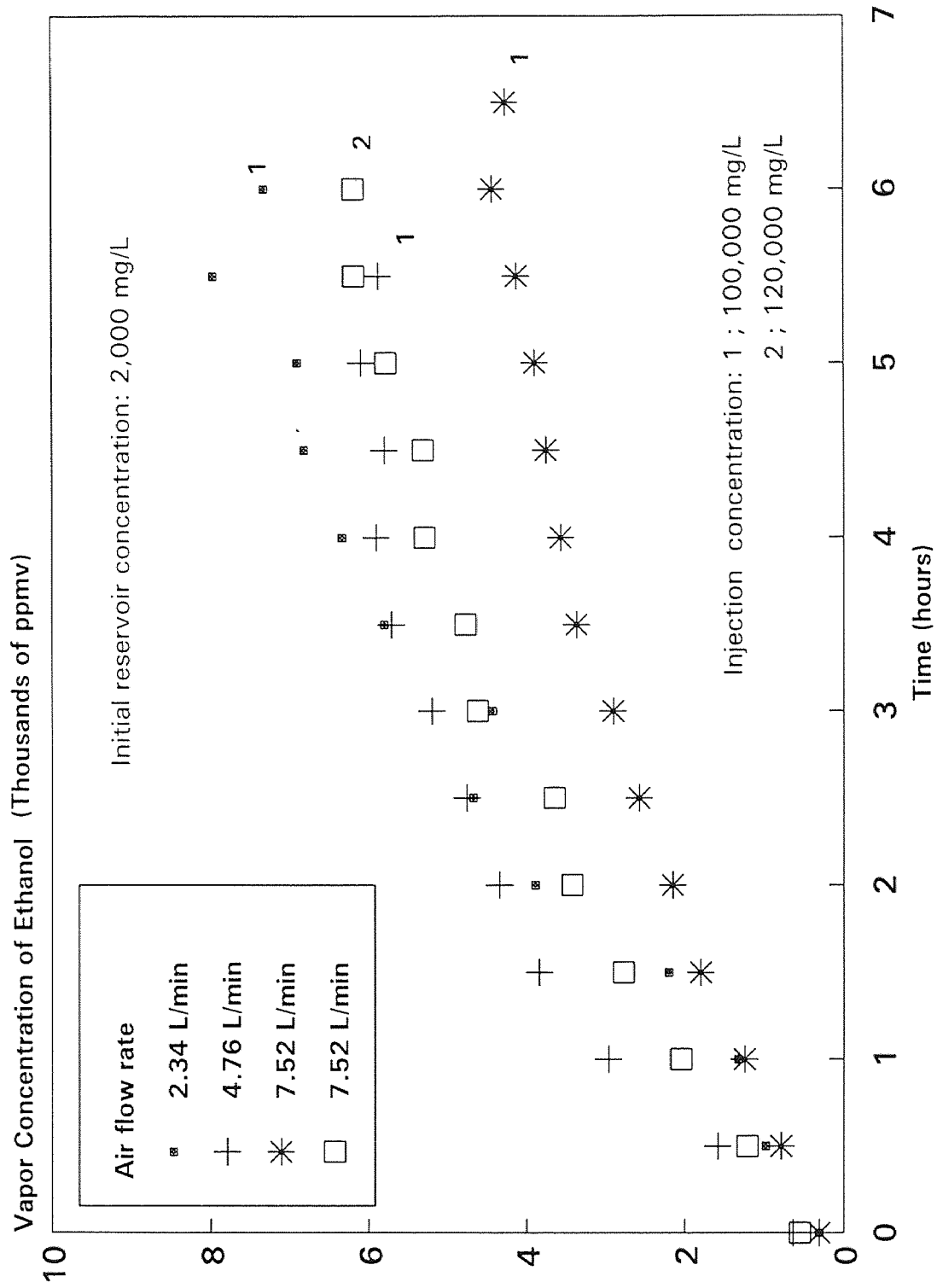


Figure 3 Bioreactor Inlet Vapor Concentrations at Different Air Flow Rates and High Ethanol Liquid Feed Injection Concentration



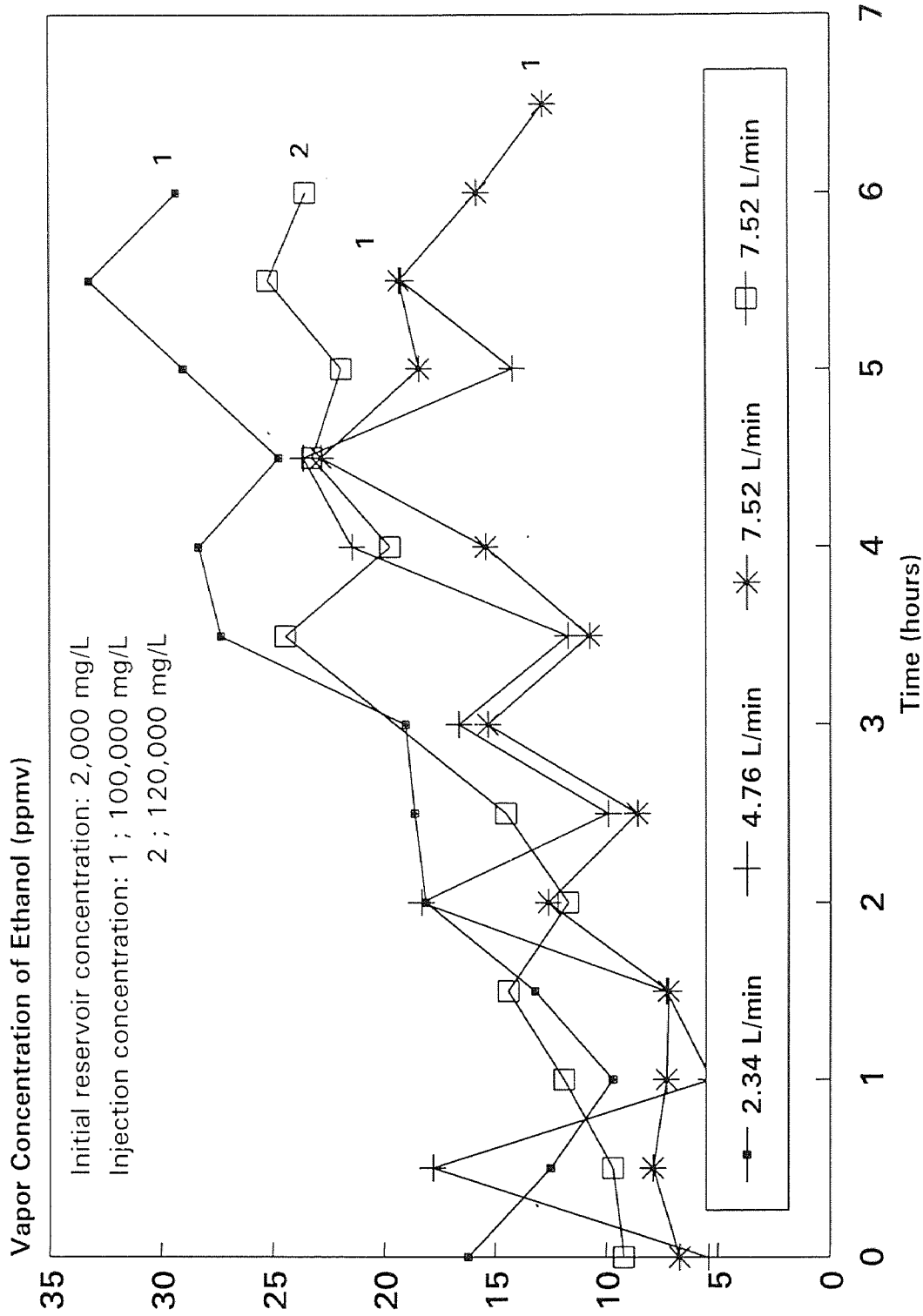


Figure 4 Outlet Vapor Concentrations at Different Air Flow Rates with Vapor Feed Concentrations Ranging from 4,200 ppmv to 7,000 ppmv

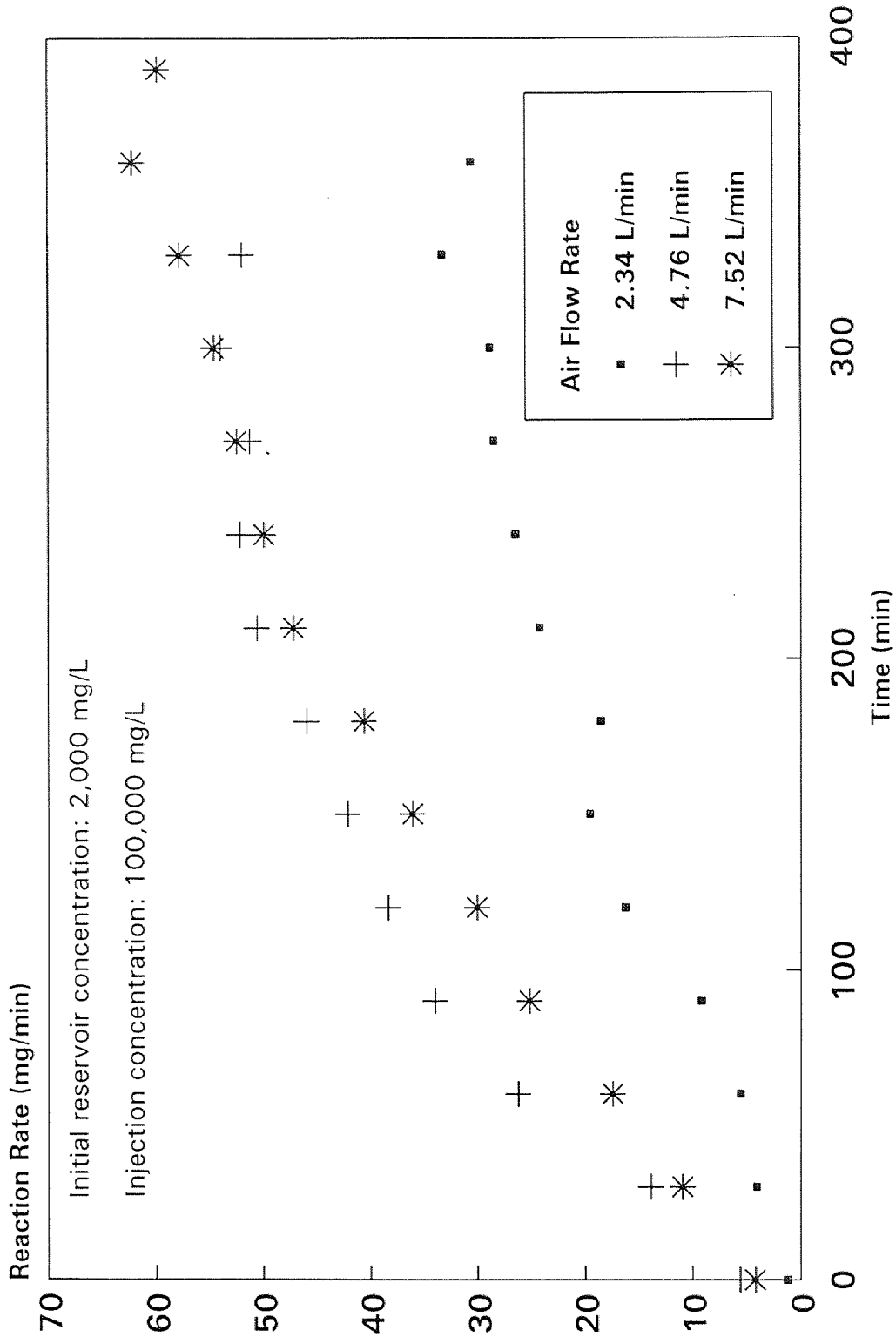


Figure 5 Reaction Rate at Different Air Flow Rates

Figures 6, 7 and 8 combine to summarize results of using air flow rates ranging from 7.52 L/min to 40 L/min under a constant liquid feed injection of ethanol (50,000 mg/L) for the 6 - 8.5 hour run. Varying with air flow rates, the inlet vapor concentrations ranged from about 2,200 ppmv to 570 ppmv at the steady state after 6 hours, as shown in Figure 8. From Figure 7, the apparent optimal air flow rate was 20 L/min. This gave a maximum reaction rate (point 3, Figure 7) as compared to points 1 and 2. However, points 4 and 5 represent lower rates, due to lower ethanol vapor concentrations, as will be shown later.

Additionally, the outlet vapor concentrations of ethanol at the higher air flow rates (30, 40 L/min) were higher than those at the lower air flow rates (7.52, 10, 20 L/min), as shown in Figure 9. Figure 10 shows that the removal efficiency decreases at 30 L/min and 40 L/min. But, the lower air flow rates show about 99 percent removal efficiency. The removal efficiency is defined as the reaction rate expressed as a percent of the inlet organic loading rate. Also, Figure 11 shows the reaction rate vs the organic loading rate at different air flow rates. The maximum reaction rate (about 40 mg of ethanol per minute) was at 20 L/min of air flow after 5.5 hours. At 30 L/min and 40 L/min air flows, the reaction rates reduce to about 37 mg/min and 30 mg/min, respectively. For the best treatment of VOCs in air, the outlet vapor concentration should be considered.

## 5.2 Vapor-Liquid Equilibrium Data in the Reservoir

Binary vapor-liquid equilibrium data of ethanol are available in the literature. However, when air is bubbled through the liquid, the system is no longer binary (Figure 12). Therefore, these data for the air flow rate of 20 L/min are presented for use by engineers and applied scientists interested in these values. As can be seen from the figure, the data are consistent for three separate runs.

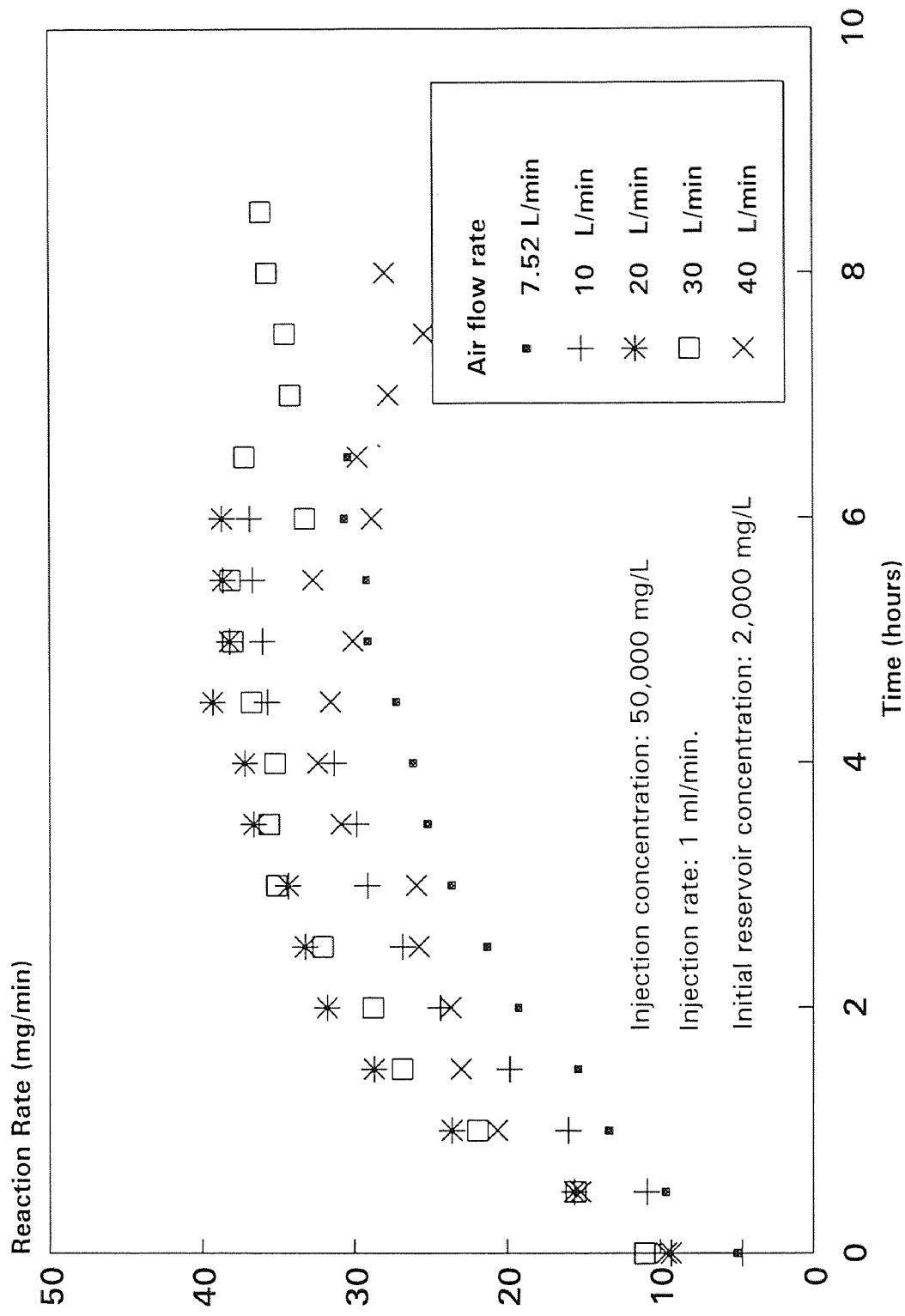


Figure 6 Determination of the Optimal air flow rate for Bio-Oxidation at 50,000 ppm Feed Injection

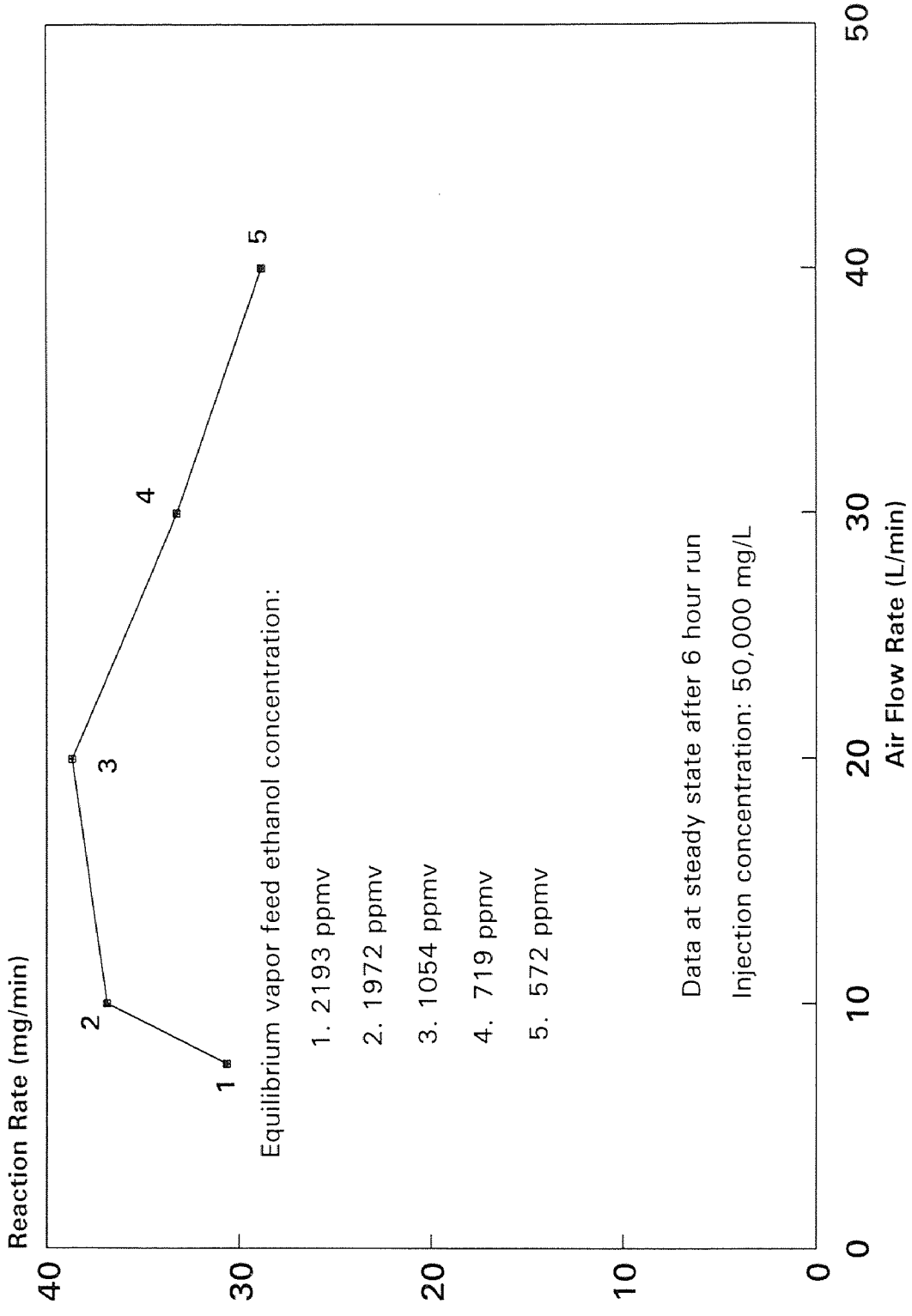


Figure 7 Reaction Rates for the Air Flow Rates in Figure 6

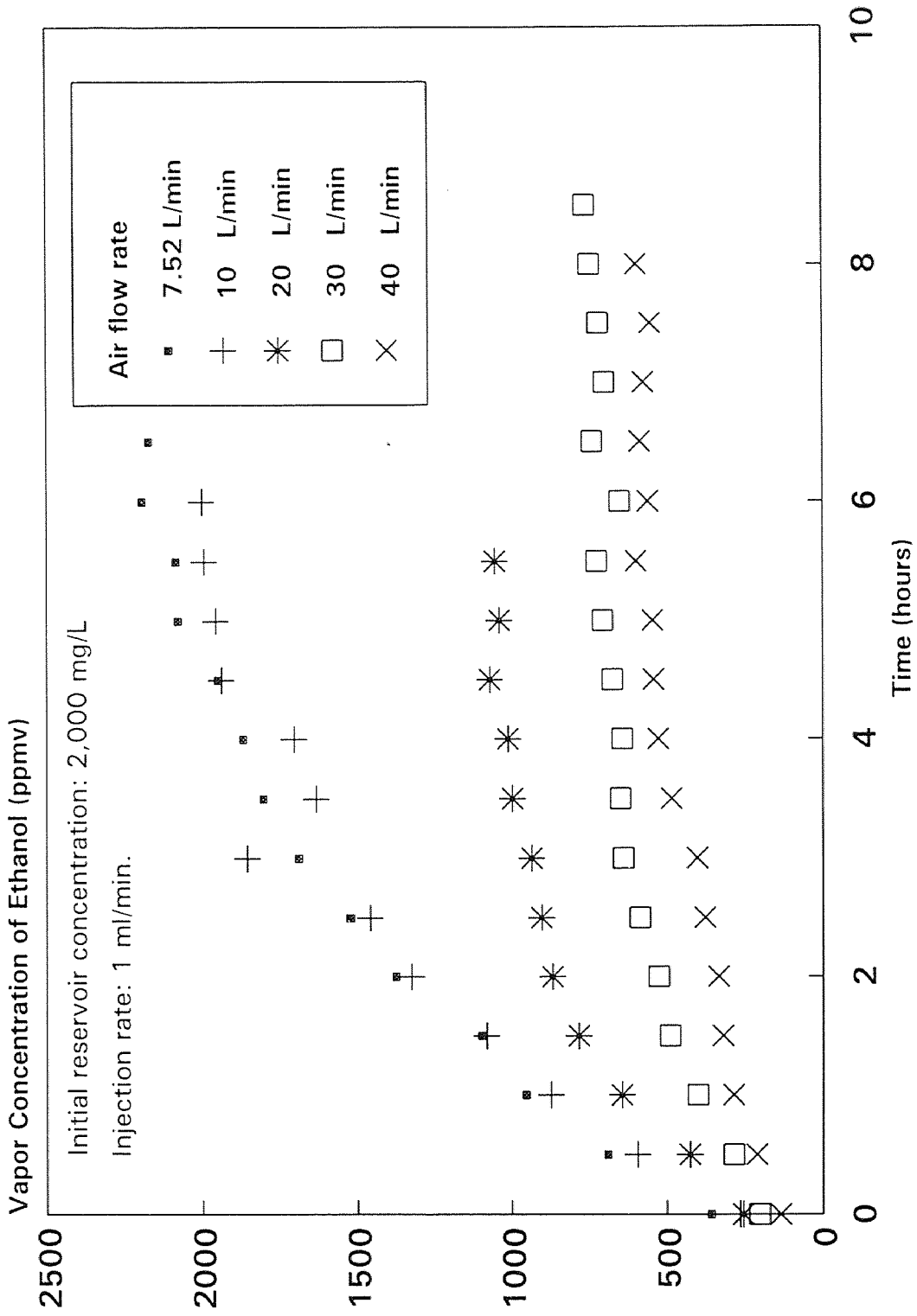


Figure 8 Inlet Vapor Concentrations at Different Air Flow Rates and 50,000 mg/L of the Injection Concentration

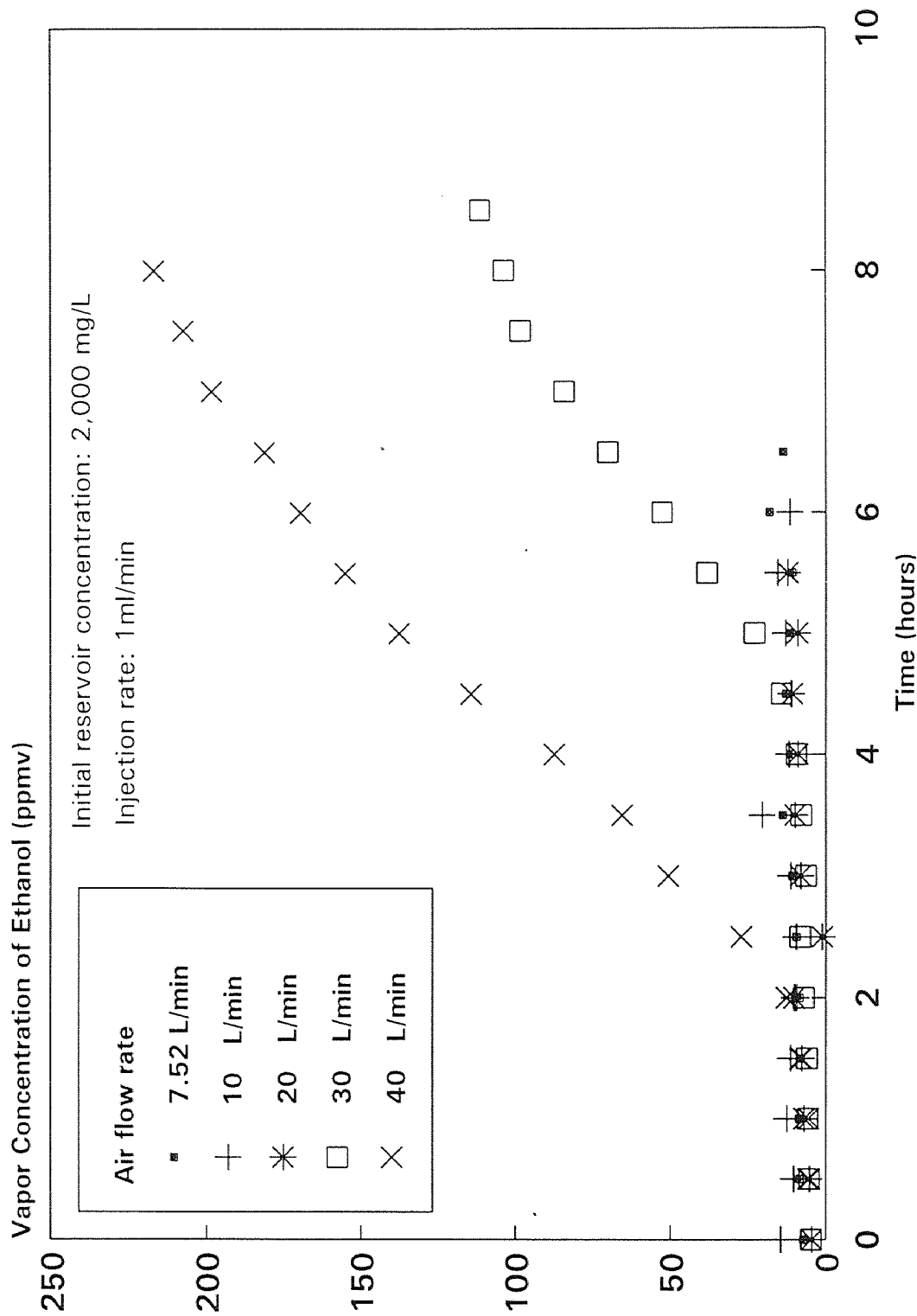


Figure 9 Outlet Vapor Concentrations at Different Air Flow Rates and 50,000 mg/L of the Injection Concentration

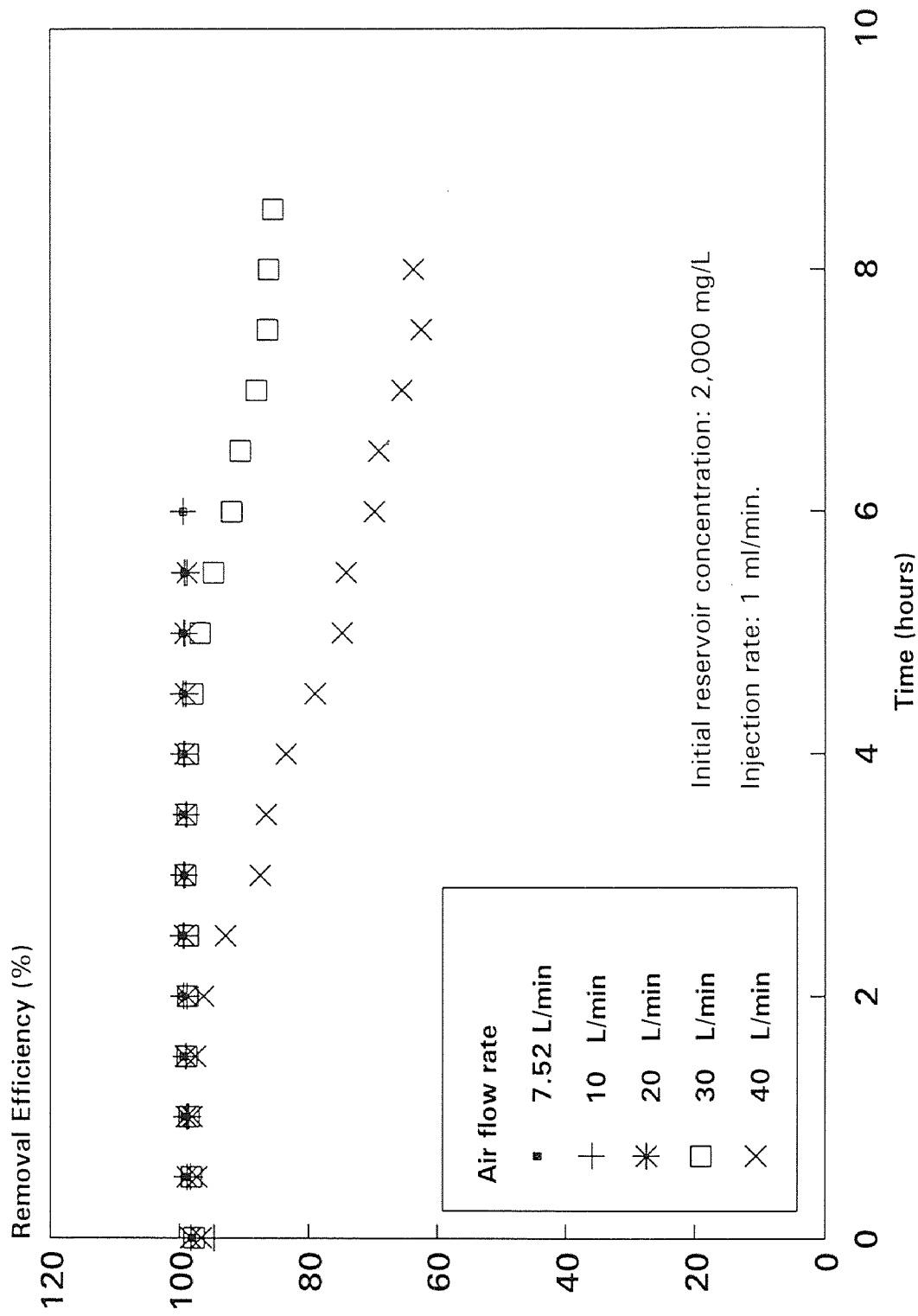


Figure 10 Removal Efficiency of the Gaseous Ethanol in the Air Bioreactor at Different Air Flow Rates and 50,000 mg/L of Injection Concentration



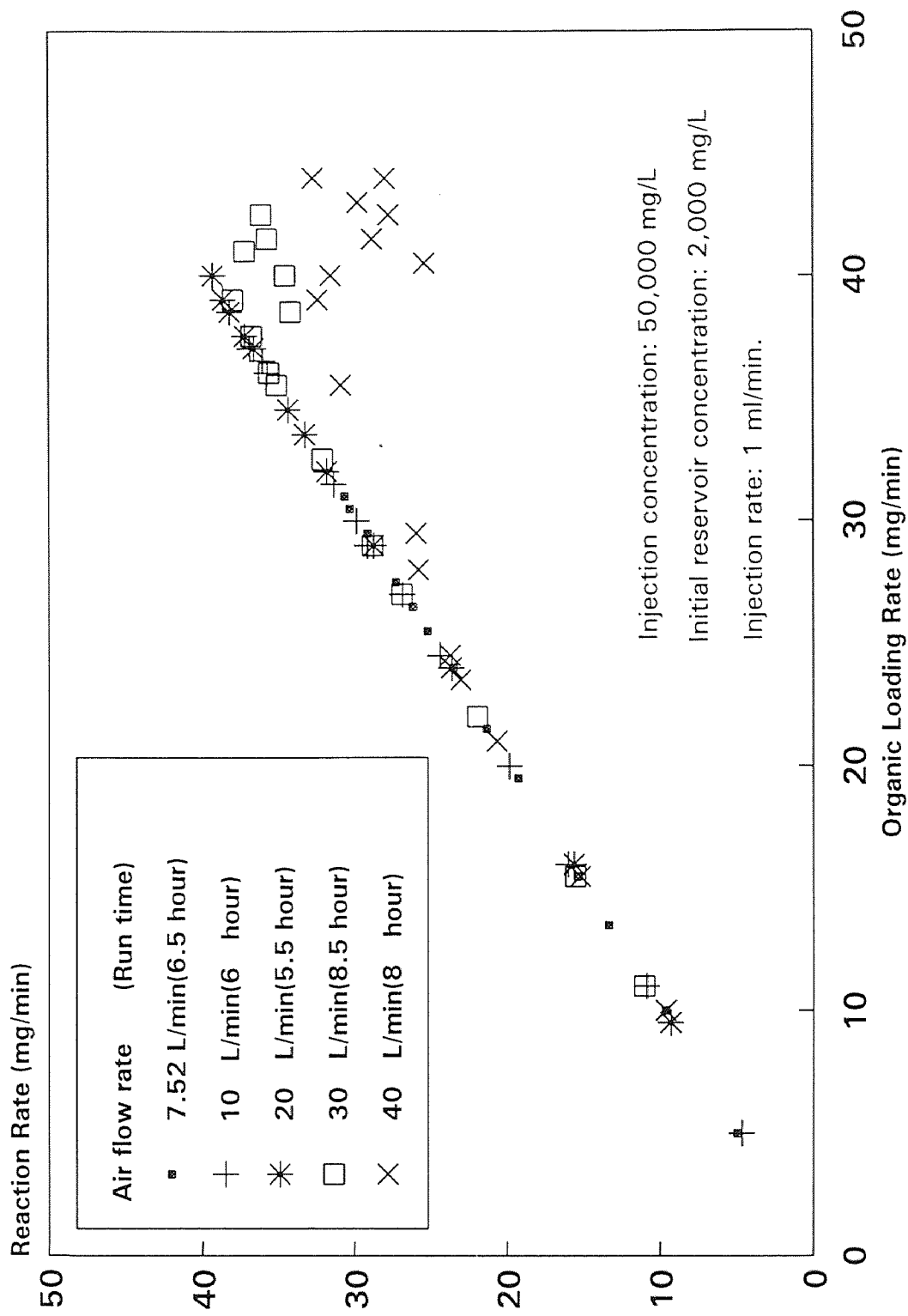
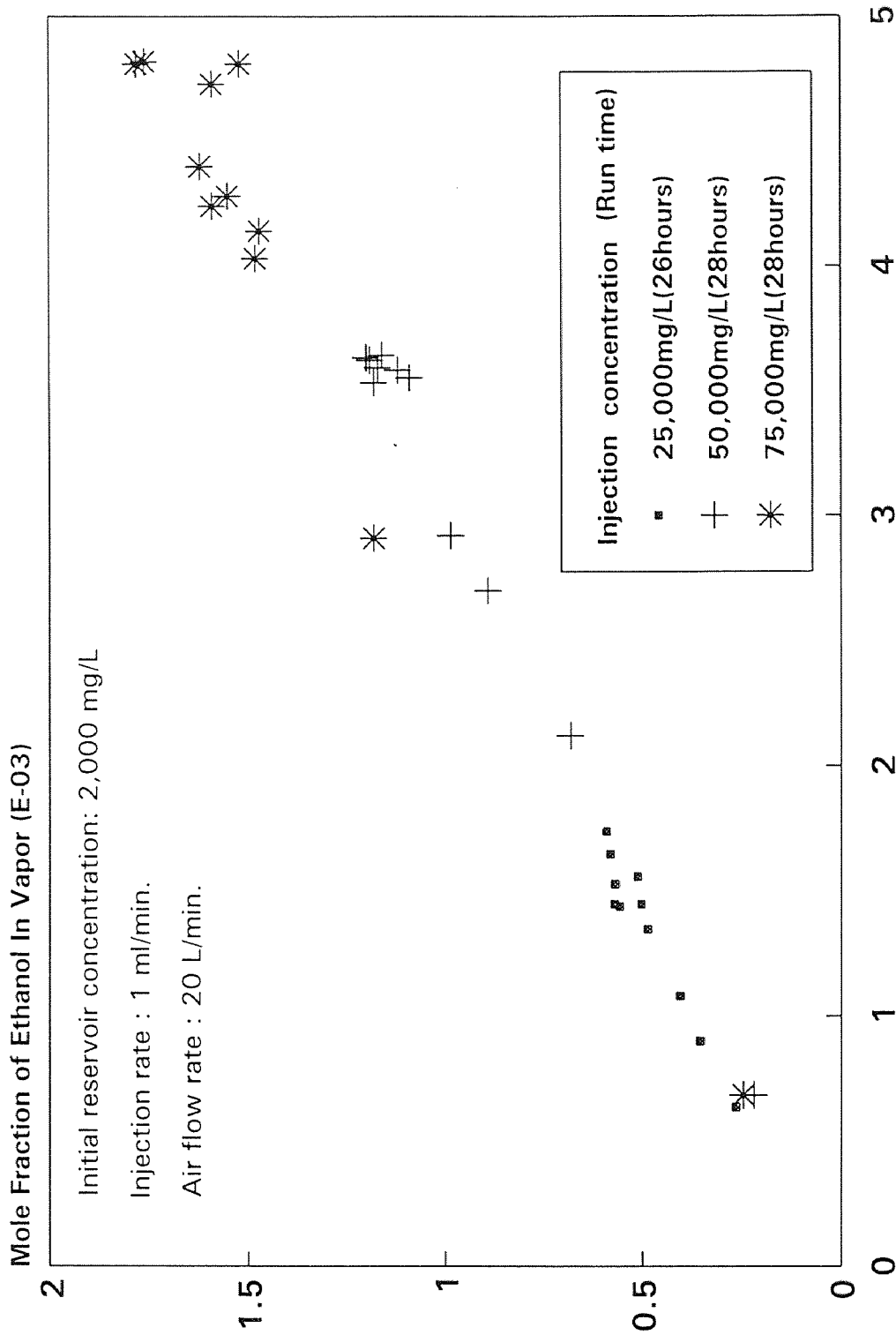


Figure 11 Reaction Rate vs Organic Loading Rate at Different Air Flow Rates



**Figure 12** Equilibrium Ethanol Concentrations in Vapor and Liquid in the Reservoir at Different Injection Concentrations

### 5.3 Effect of the Substrate Concentration for Bio-Oxidation

Figure 13 shows the reaction rate of the bioreactor as a function of the organic loading rate to the bioreactor. The reaction rate and the organic loading rate are given per unit time. The results are obtained from several combined experiments of 25,000, 50,000 and 75,000 mg/L in the feed concentration of ethanol at the same air flow rate (20 L/min), along with concentrations of 20,000, 50,000, 100,000 and 120,000 mg/L at 7.52 L/min. The reaction rate remained nearly constant at 20 L/min when the organic loading rate (the substrate concentration) was increased to a level above the critical load (about 55 mg of ethanol / min), which corresponded with the maximum value (about 50 mg of ethanol / min) of the reaction rate. Thus, the gaseous ethanol in this region is eliminated according to zero-order reaction kinetics. However, at 7.52 L/min, this plateau was not yet reached, even at reaction rates of over 80 mg/min.

Also, for the duration of 28 hour runs for each of these feed injection levels, the maximum reaction rate at 20 L/min of air flow was maintained at about 44 to 50 mg of ethanol per minute (2.64 - 3 g/hour), as shown in Figure 14. The vapor concentrations of ethanol at the inlet at this air flow rate reached equilibrium levels ranging from about 1,000 ppmv to 1,700 ppmv within the first four hours of the 28 hour runs (Figure 15). The outlet vapor concentrations in these experiments showed big differences between 75,000 mg/L and the other two injected concentrations after approximately 6 hours. When 75,000 mg/L of ethanol was injected into the reservoir, the inlet vapor concentration was converted from about 1,700 ppmv to 420 ppmv. In case of 25,000 mg/L and 50,000 mg/L, the outlet concentrations were about 5 to 8 ppmv (Figure 16). This is possibly due to substrate inhibition at the higher concentrations.

From the above experiments, Figure 17 shows each removal efficiency of gaseous ethanol in the air bioreactor: about 73 % for 75,000 mg/L feed, about 99 % for 25,000 mg/L and 50,000 mg/L.

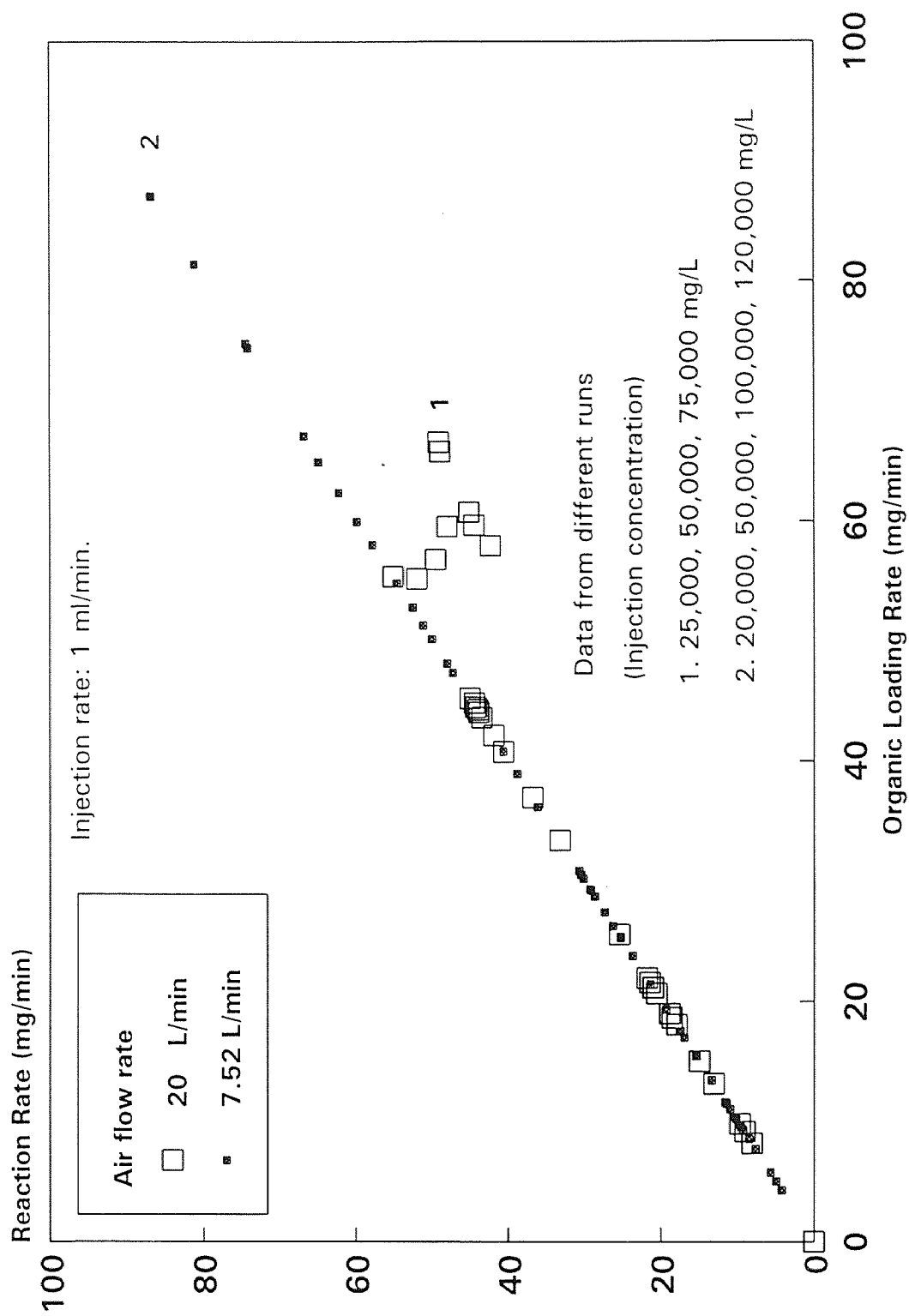


Figure 13 Reaction Rate vs Organic Loading Rate in the Air Bioreactor

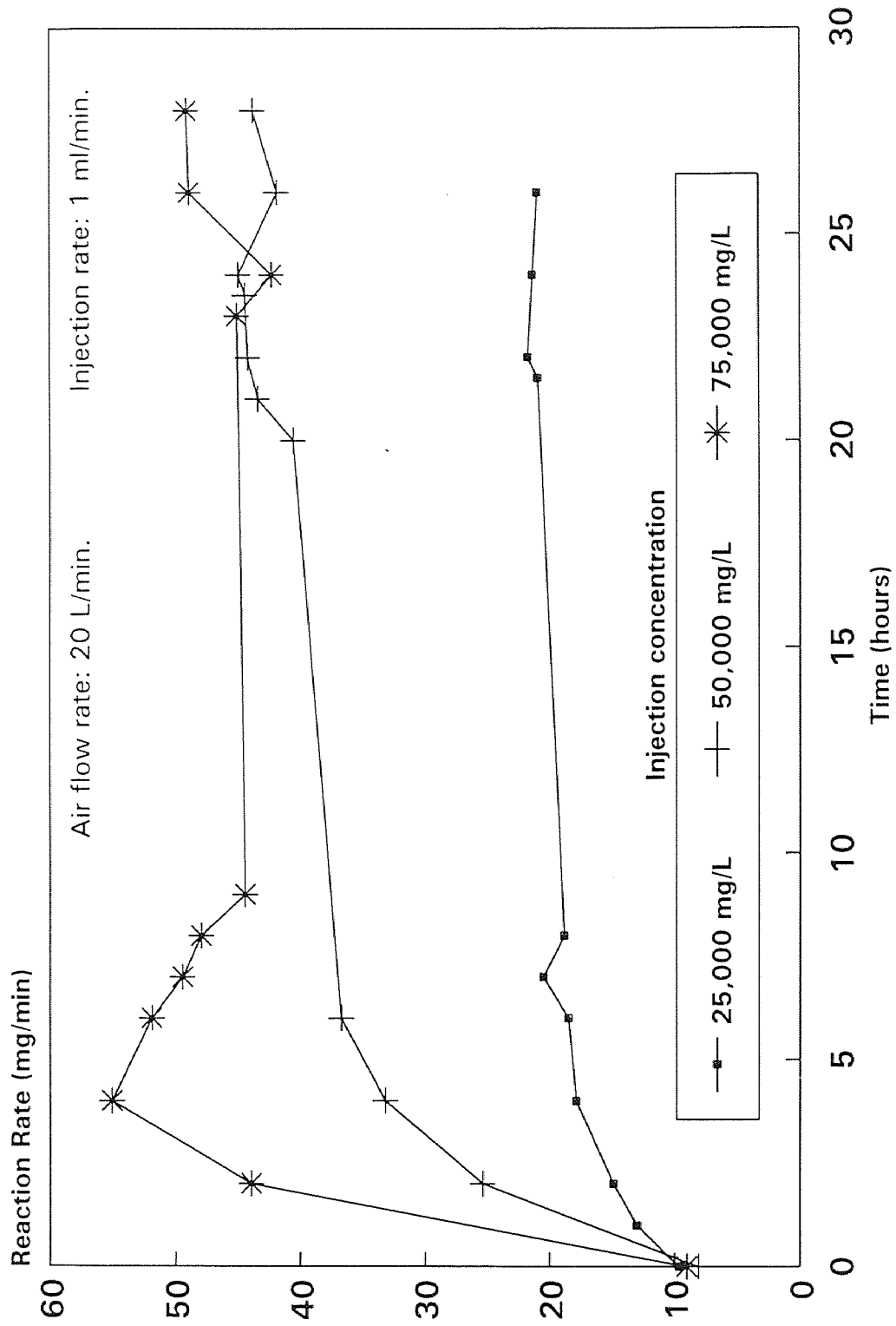


Figure 14 Reaction Rate at 20 L/min of Air Flow and Different Injection Concentrations

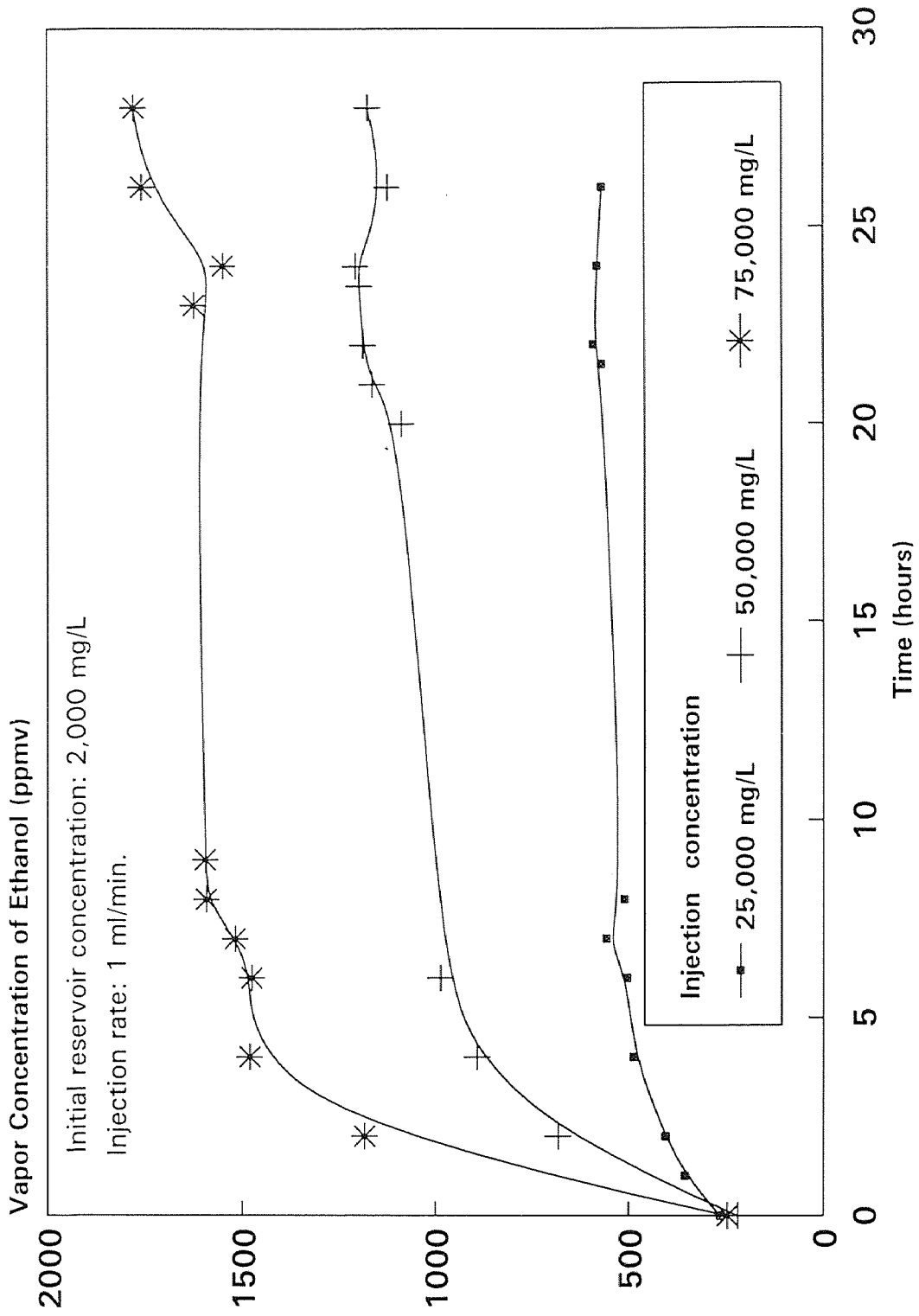


Figure 15 Inlet Vapor Concentration of Ethanol at 20 L/min of Air Flow

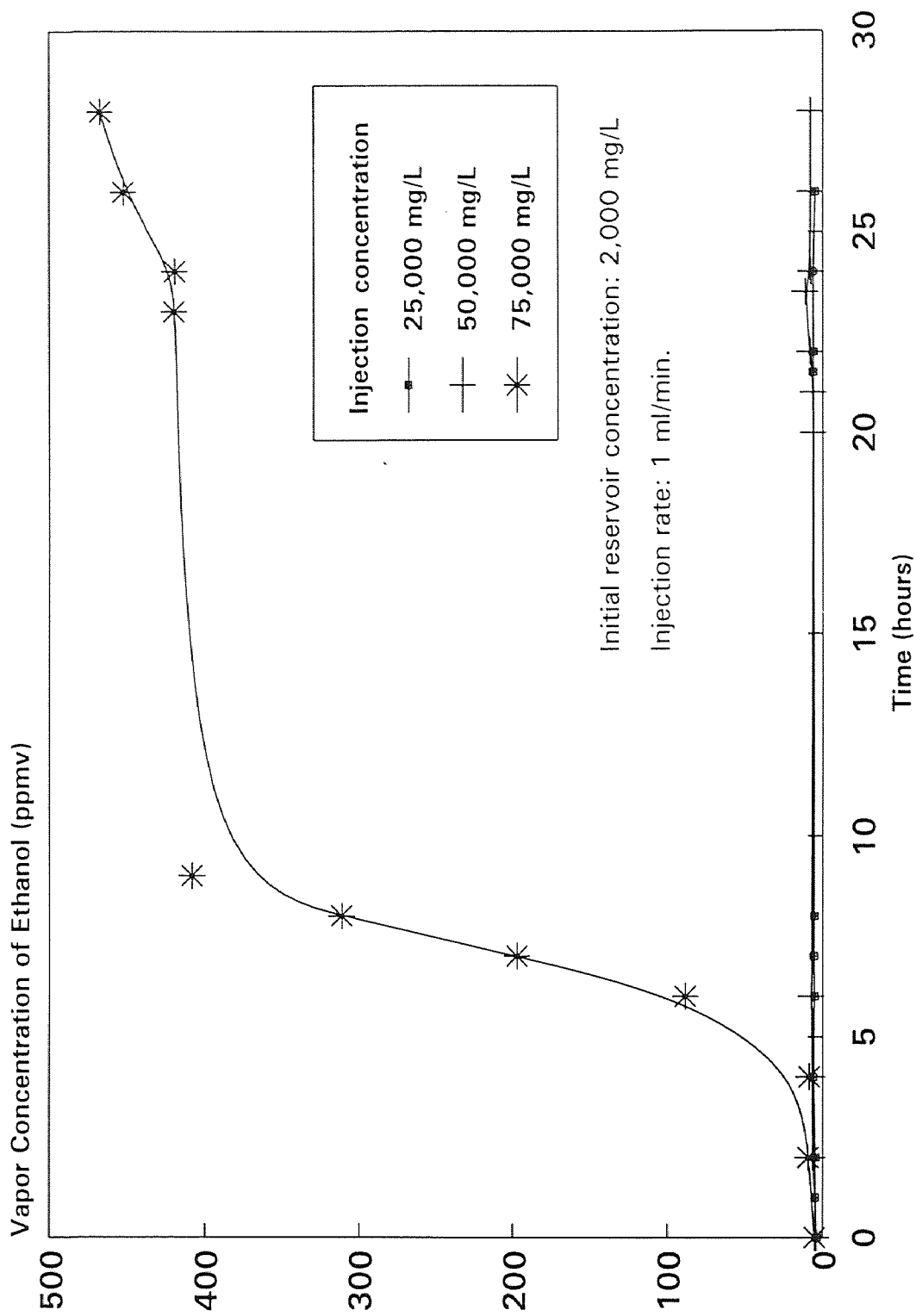


Figure 16 Outlet Vapor Concentration of Ethanol at 20 L/min of Air Flow

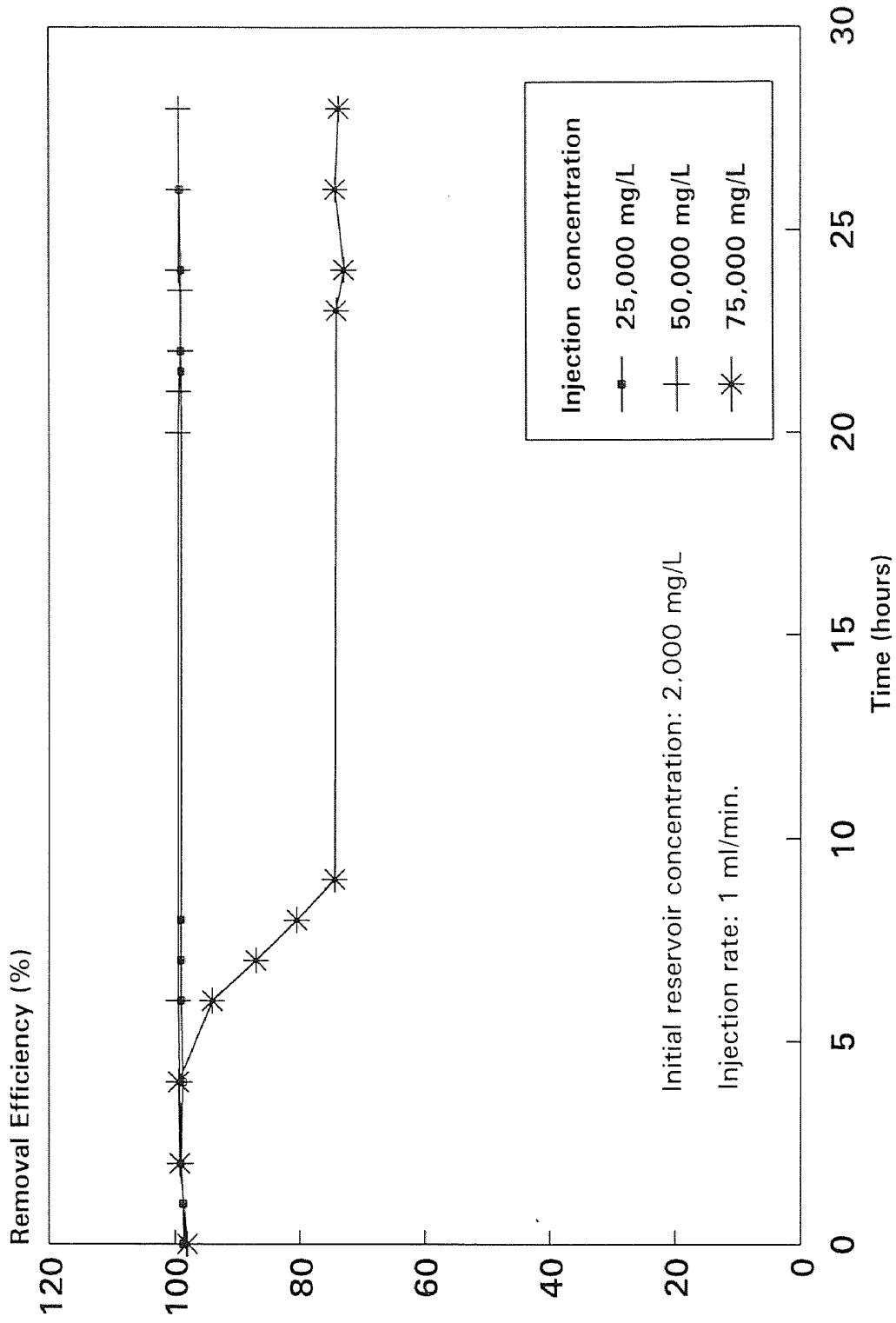


Figure 17 Removal Efficiency of Gaseous Ethanol in the Air Bioreactor at 20 L/min of Air Flow and Different Injection Concentrations



Figure 18 shows the reaction rates at 7.52 L/min of air flow with various concentrations of ethanol injected into the reservoir. The injected concentrations ranged from 20,000 mg/L to 120,000 mg/L. The reaction rates were increased according to the increase of the injected concentration of ethanol. Thus, at 7.52 L/min of air flow for the 4 - 6.5 hour run, the reaction rate can be more than 87 mg of ethanol per minute (5.22 g/hour) by increasing the feed concentration of ethanol.

#### **5.4 Variation of the Optimal Vapor Temperature for the Air Bioreactor**

Most of microorganisms for bio-oxidation are mesophilic, showing the good activity between 20°C and 40°C. In this study, the determination of a desirable vapor temperature is investigated for the best biological activity in the air bioreactor, changing the inlet vapor temperatures of 24°C, 27°C and 30°C for 28 hours.

The range between 27°C and 30°C at the inlet was preferable to 24°C as shown in Figures 19 and 20. In this range, the maximum reaction rate was about 42 to 44 mg of ethanol per minute when 50,000 mg/L of ethanol solution was injected. The removal efficiency ranged from about 94 % to 99 %. At 24°C, the reaction rate decreased gradually after 6 hours. After 28 hours, the removal efficiency was about 92 %.

#### **5.5 General Observations for the Operation of the Air Bioreactor**

The air bioreactor was operated under the different conditions for about four months until all results presented in this study were obtained. Total amounts of organic compounds loaded to the bioreactor during this period were about 1,430 g of the carbon sources which were mainly ethanol, including small amounts of isopropanol. In other words, the liquid volume fed was 1,908 ml of ethanol which is 95 % in purity and 0.789 in density.

The equilibrium ethanol concentrations of the gas and liquid phases in the reservoir were compared in this study. Figure 21 shows the equilibrium concentrations of ethanol as a function of the air flow rate at 50,000 mg/L of injection concentration. The numerical

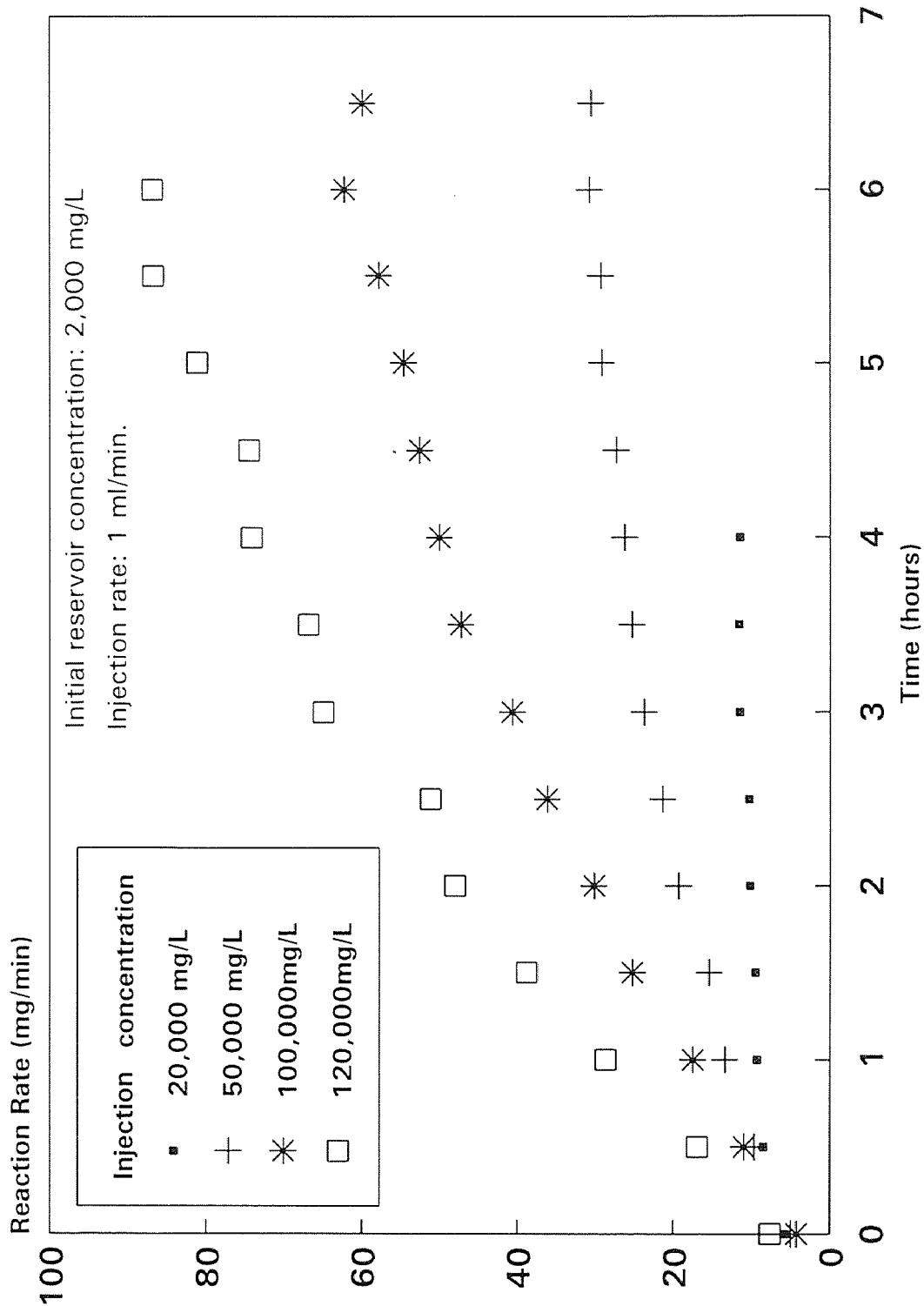


Figure 18 Reaction Rate at 7.52 L/min of Air Flow and Different Injection Concentrations

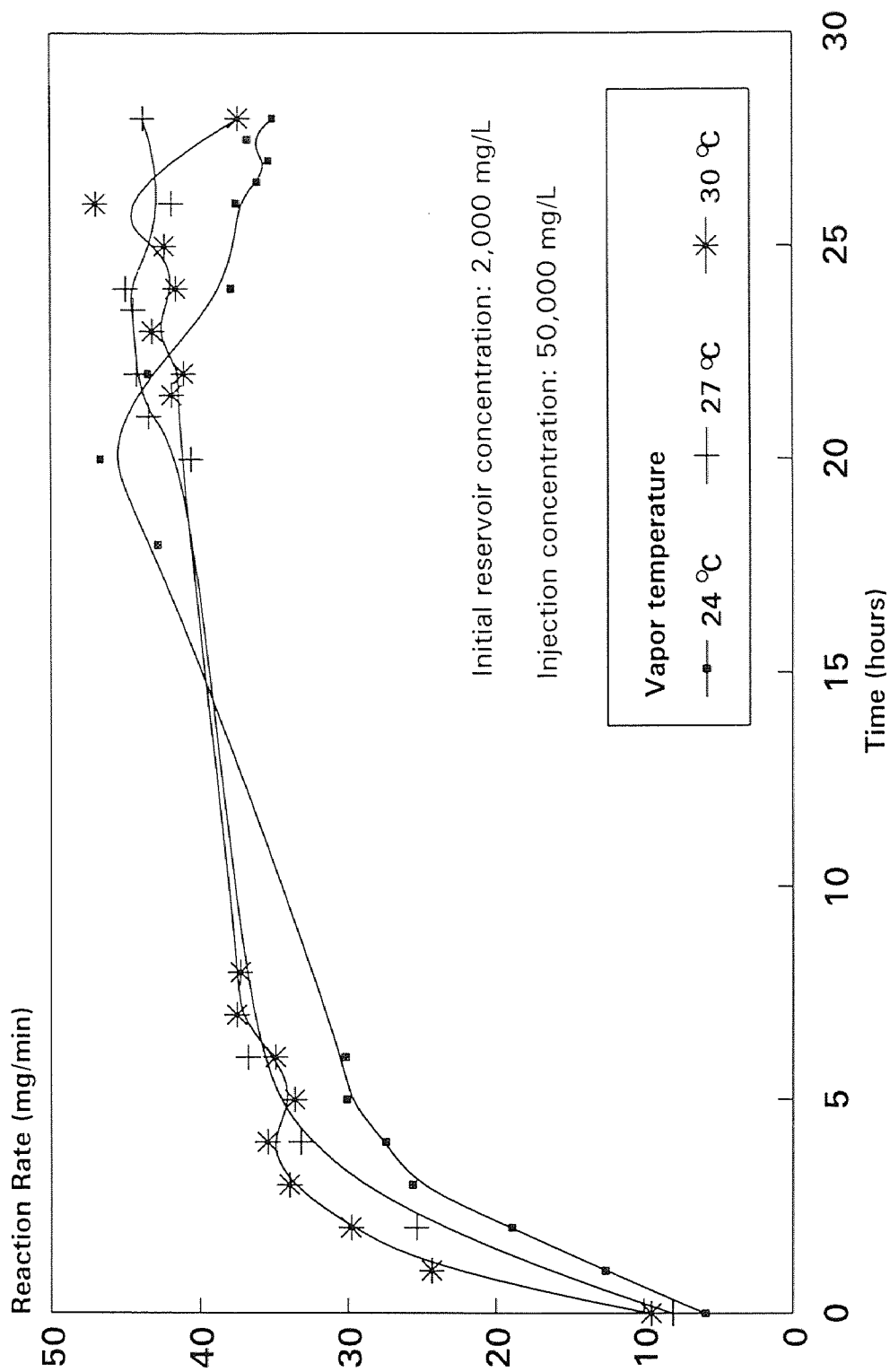


Figure 19 Reaction Rates at Various Vapor Temperatures for Bio-Oxidation

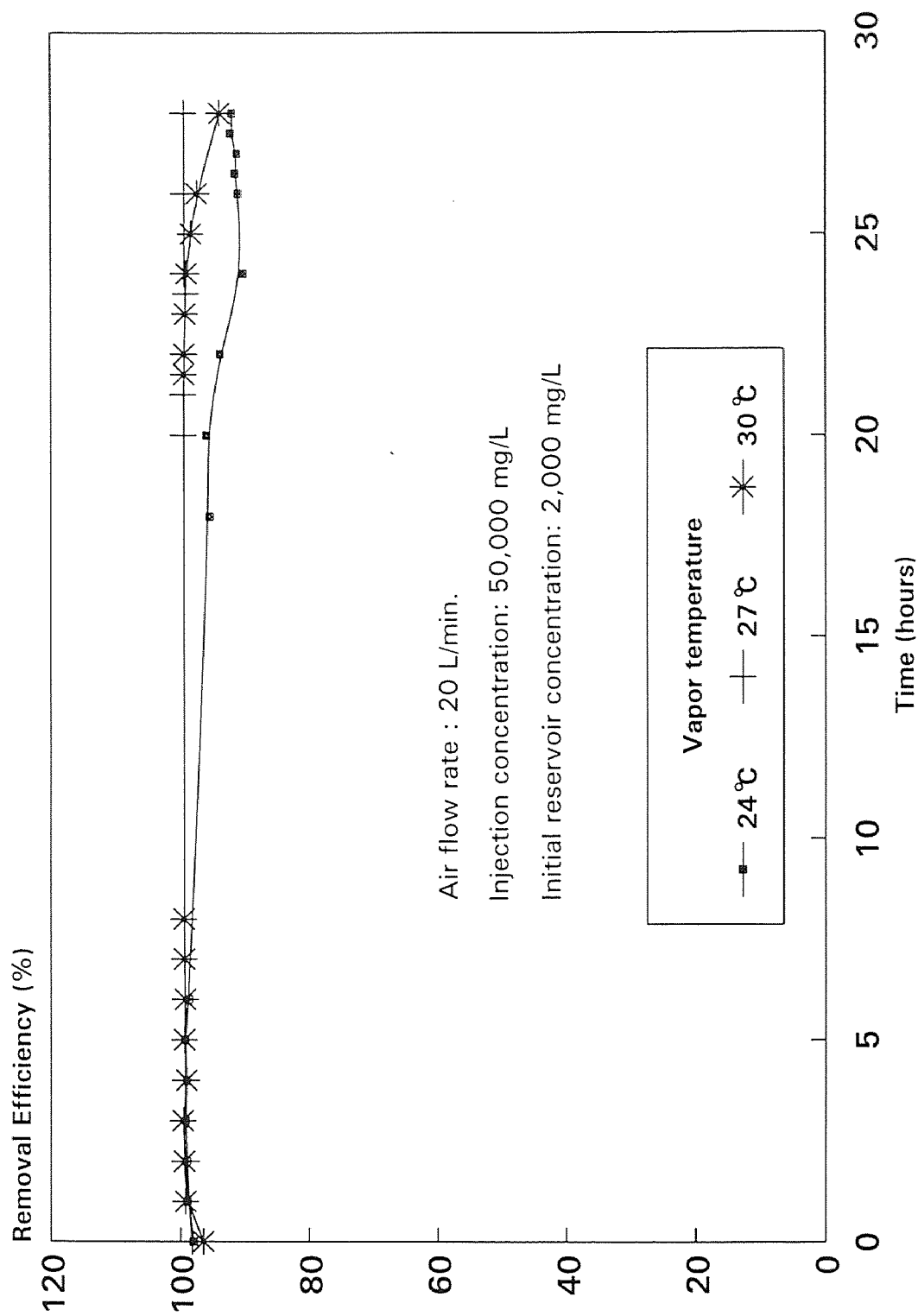
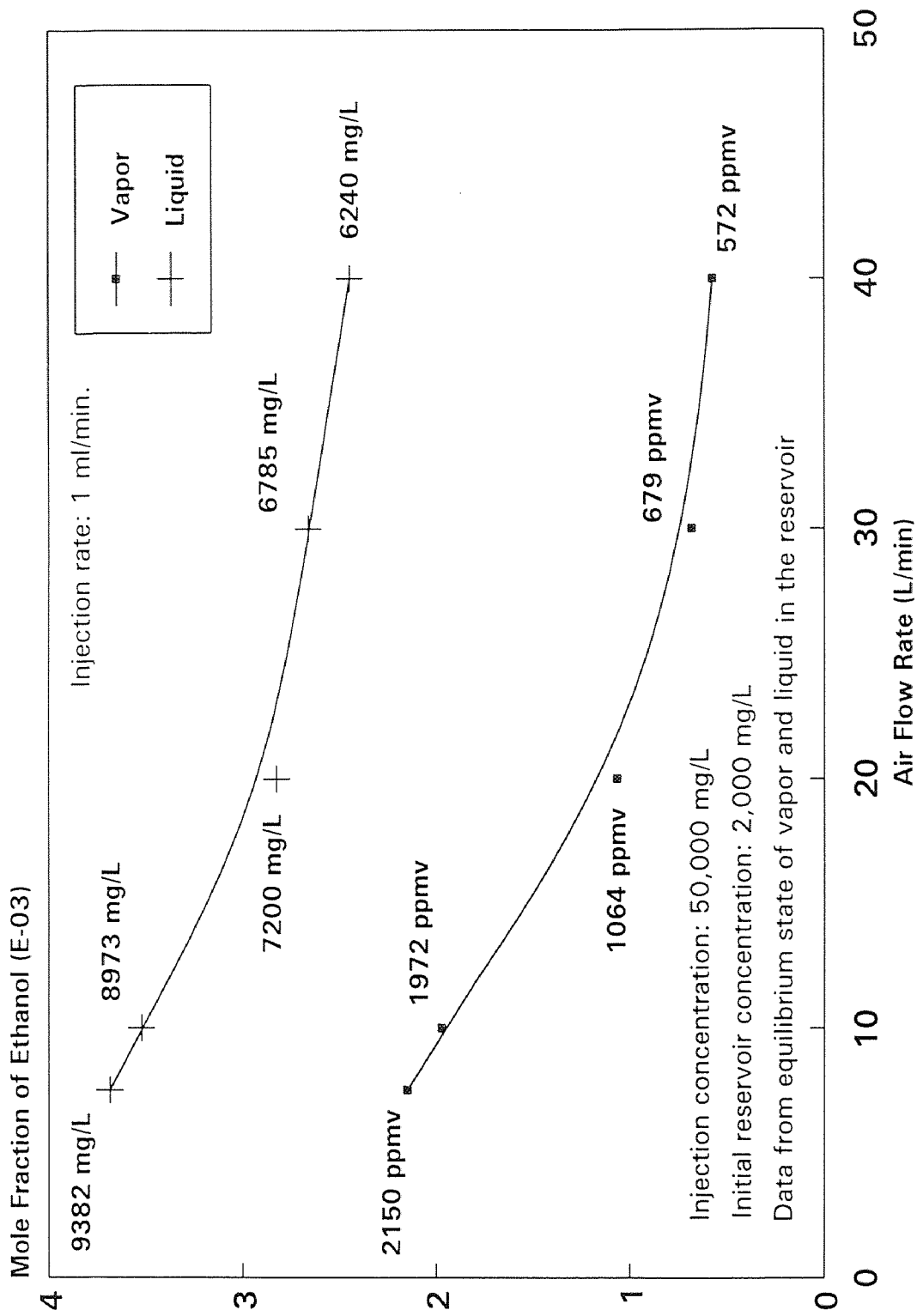


Figure 20 Removal Efficiency of Gaseous Ethanol at Different Vapor Temperatures



**Figure 21** Comparison of Ethanol Concentrations in Vapor and Liquid in the Reservoir at Different Air Flow Rates

values are summarized in Table 2. At the range from 7.52 L/min to 40 L/min of air flow, the ratios of mole fractions of ethanol in liquid and vapor ranged from 1.71 to 4.28.

It was found only by the sense of smell that the offensive odor emanated from isopropanol and ammonium hydroxide could be eliminated by the bioreactor.

A minor operating problem of the bioreactor was found throughout these experiments. Condensed water accumulated on the bottom of the reactor. This water tended to be stagnant and anaerobic without aeration. As a result, a musty, slightly unpleasant odor was generated from the collected bottom water, especially when the high concentration of ethanol was loaded to the bioreactor. Thus, an idea for the most complete treatment of the gaseous VOCs, needs to be recommended in this study.

**Table 2** Comparison of the Vapor and Liquid Concentrations of Ethanol at the Equilibrium State in the Reservoir

Air flow rate (L/min.)	Mole fraction ( $\times 10^{-3}$ )		Concentration		Ratio of liquid to vapor as the mole fraction
	Liquid	Vapor	Liquid (mg/L)	Vapor (ppmv)	
7.52	3.687	2.150	9,382	2,150	1.71
10	3.525	1.972	8,973	1,972	1.79
20	2.826	1.064	7,200	1,064	2.66
30	2.662	0.679	6,785	679	3.92
40	2.447	0.572	6,240	572	4.28

Injected ethanol concentration : 50,000 mg/L

Initial reservoir concentration : 2,000 mg/L

## CHAPTER 6

### CONCLUSIONS AND RECOMMENDATIONS

This study has demonstrated the capability of the spirally wound air bioreactor to degrade a model VOC in air. In the bioreactor, the high concentrations (4,200 - 7,000 ppmv) of gaseous ethanol were converted to the very low concentrations (13 - 30 ppmv) with air flow rates of from 2.34 to 7.52 L/min for the 6 hour runs (Figures 3 and 4).

The reaction rate varied with the air flow rates. 20 L/min of air flow was determined as an optimal one for low ethanol loading for air flow rates of from 7.52 L/min to 40 L/min.

The vapor temperature observed as best was between 27°C and 30°C.

At 20 L/min, the maximum reaction rate was maintained at about 44 to 50 mg of ethanol per minute for 28 hour runs.

The bio-oxidation in the air bioreactor depended on the zero order reaction kinetics above the critical load of the substrate, at 20 L/min.

At 7.52 L/min, reaction rates as high as 87 mg/min were observed, with 99 percent removal efficiency. It is recommended that regimes of higher loading be investigated in future studies.

At the equilibrium state between gas and liquid phase in the reservoir, the ethanol concentration in each phase was observed. The molar ratio of liquid to vapor in the ethanol concentration ranged from 1.71 to 4.28 at 7.52 - 40 L/min of air flow.

To prevent the unpleasant odor from the accumulated water in the reactor bottom, the recirculation of the bottom water to the reservoir is recommended.

In further studies, it is necessary to observe reaction rates at 30 and 40 L/min of air flow, varying injection concentrations of ethanol, and to show the potentiality of the air bioreactor to eliminate other air pollutants.



## APPENDIX

### Sample Calculations

1. Sample calculations for ethanol vapor concentration (ppmv) and organic loading rate (mg of ethanol / min) into the air bioreactor:

[Example]

- Air flow rate : 20 L/min
- A sample volume for the analysis of a gas chromatograph : 2 ml (using a gas syringe)
- Room temperature : 28 °C
- A peak area of the gas chromatograph : 106.0058

[Calculations]

By the equation of  $P_1V_1 / T_1 = P_2V_2 / T_2$ ,

$P_1 = 760$  mmHg,  $T_1 = 273$  °K,  $V_1 = 22.4$  L (volume of 1 mole gas at 0 °C, 1 atm)

$P_2 = 760$  mmHg,  $T_2 = (273 + 28)$  °K,  $V_2 = 24.7$  L (volume of 1 mole gas at 28 °C, 1 atm)

\* Equation of ethanol vapor concentration (ppmv):

$$\text{ppmv} = (\text{moles of ethanol} / \text{moles of air}) \times 10^6 \text{ -----(1)}$$

- Moles of air ( volume of the sample: 2 ml ):

$$= 0.002 \text{ L} / 24.7 \text{ L} = 8.1 \times 10^{-5} \text{ -----(2)}$$

- Moles of ethanol in 2ml of the gas syringe (Y):

by a linear regression with calibration data of a standard ethanol

$$Y = ( 8.524 \times 10^{-10} )X + ( 9.08 \times 10^{-11} ) \text{ -----(3)}$$

where X = a peak area of a gas chromatograph

From the calculations 1, 2 and 3, the vapor concentration of ethanol (ppmv):

$$\begin{aligned}
 &= (\text{moles of ethanol in 2 ml of gas} / 8.1 \times 10^{-5}) \times 10^6 \\
 &= (1.235 \times 10^{10}) Y \\
 &= (1.235 \times 10^{10}) \{ (8.524 \times 10^{-10})X + (9.08 \times 10^{-11}) \} \\
 &= 10.5271 X + 1.1214 = 10.5271 (106.0058) + 1.1214 \\
 &= \mathbf{1,117 \text{ ppmv}}
 \end{aligned}$$

- Mass of ethanol for organic loading rate ( 1 mole of ethanol = 46.07 g ):

$$M_1 = (\text{moles of ethanol}) \times 46,070 \text{ mg} \text{ -----(4)}$$

- At 20 L/min of air flow, moles of ethanol in 20 L of gas per minute ( $M_2$ ):

$$M_2 (\text{moles /min}) = (20 \text{ L} / 0.002 \text{ L}) \times Y = 9.04501 \times 10^{-4} \text{ -----(5)}$$

From the calculations 4 and 5, the organic loading rate (mg of ethanol / min) into the bioreactor at 20 L/min of air flow:

$$\begin{aligned}
 &= M_2 \times 46,070 \text{ mg} = (9.04501 \times 10^{-4} \text{ moles / min}) \times 46,070 \text{ mg} \\
 &= \mathbf{41.67 \text{ mg / min}}
 \end{aligned}$$

2. A sample calculation for the removal efficiency of gaseous ethanol in the air bioreactor

[Example]

- Organic loading rate at the inlet of the bioreactor: 41.67 mg of ethanol /min
- Discharged organic rate at the outlet of the bioreactor: 6.77 mg of ethanol / min

[Calculation]

$$\begin{aligned}
 \text{The removal efficiency (\%)} &= \{ (41.67 - 6.77) / 41.67 \} \times 100 \% \\
 &= \mathbf{83.75 \%}
 \end{aligned}$$

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