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Demonstration of Oxygen-Enriched Combustion System on a Light-Duty Vehicle to Reduce Cold-Start Emissions

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ABSTRACT

The oxygen content in the ambient air drawn by combustion engines can be increased by polymer membranes. The authors have previously demonstrated that 23 to 25% (concentration by volume) oxygen-enriched intake air can reduce hydrocarbons (HC), carbon monoxide (CO), air toxics, and ozone-forming potential (OFP) from flexible-fueled vehicles (FFVs) that use gasoline or M85. When oxygen-enriched air was used only during the initial start-up and warm-up periods, the emission levels of all three regulated pollutants [CO, nonmethane hydrocarbons (NMHC), and NO_x] were lower than the U.S. EPA Tier II (year 2004) standards (without adjusting for catalyst deterioration factors). In the present work, an air separation membrane module was installed on the intake of a 2.5-L FFV and tested at idle and free acceleration to demonstrate the oxygen-enrichment concept for initial start-up and warm-up periods. A bench-scale, test set-up was developed to evaluate the air separation membrane characteristics for engine applications. On the basis of prototype bench tests and from vehicle tests, the additional power requirements and module size for operation of the membrane during the initial period of the cold-phase, FTP-75 cycle were evaluated. A prototype membrane module (27 in. long, 3 in. in diameter) supplying about 23% oxygen-enriched air in the engine intake only during the initial start-up and warm-up periods of a 2.5-L FFV requires additional power (blower) of less than one horsepower. With advances in air separation membranes to develop compact modules, oxygen enrichment of combustion air has the potential of becoming a more practical technique for controlling exhaust emissions from light-duty vehicles.

INTRODUCTION

A significant portion of the HC and CO emissions emitted by light-duty passenger vehicles occurs immediately following the start-up of the engine, because the engine block and exhaust manifold are cold and the conversion efficiency of the catalytic converter is low (when catalyst is below its light-off temperature). To compound the problem, most vehicles run fuel-rich after a cold start for better start-up and operating driveability. Also considered as potential sources of HCs from the SI engine are piston-ring crevices, poor mixture preparation, absorption of oil films, flame quenching and lower post-flame reaction rates, thermal inertia and heat loss, and valve leakage [1]. In the case of FFVs operating on alcohol fuels, catalytic control of unburned alcohol and aldehydes poses additional problems, particularly with increasing vehicle mileage [2].

With growing demand for air quality improvements in urban areas, stringent regulations are now being put on light-duty passenger vehicle exhaust emissions. To obtain compliance with California's LEV/ULEV and the U.S. Environmental Protection Agency's (EPA's) Tier II standards, substantial reductions in HC and CO emissions during the initial start-up and warm-up periods are required. Many studies have been directed toward meeting these standards with improvements in combustion and/or catalytic converter performance [3,4]. The use of oxygen-enriched combustion air has been considered as one technique to reduce HC and CO emissions from SI engines, in particular during initial start-up and warm-up periods [5,6]. Oxygen-enrichment of the combustion air can lead to faster and nearly complete combustion by providing higher oxygen-to-fuel ratios. Oxygen-enriched combustion air can reduce the engine-out HC and CO emissions rapidly, even when the engine is cold, and it helps to minimize converter limitations during the initial period. However, NO_x emissions would be higher if oxygen enrichment were used continuously, because of accompanying higher combustion temperatures. If oxygen-enriched intake air were used only during the initial start-up and warm-up periods, the advantages of lower HC and CO emission levels could be obtained while keeping NO_x emission levels sufficiently low [6].

OVERVIEW OF PREVIOUS WORK AT ARGONNE

The authors have previously reported that oxygen-enriched intake air (nominal 23 or 25%) can reduce both engine-out and converter-out FTP emission levels, particularly during cold-phase operation [6,7]. Figure 1 shows the significant reductions in engine-out total hydrocarbons (THC) and CO emissions during the initial 127 s of the cold-phase FTP cycle ("first cycle or hill"), using 23 or 25% oxygen-enriched intake air from an SI-engine-powered FFV running on Indolene fuel. When 23% oxygen-enriched air was used only during the cold phase (505 s) or 25% oxygen-enriched air was used only during the initial 127 s of the cold phase (and ambient air was used in the intake for the rest of the FTP cycle), the emission levels of all three regulated pollutants (CO, NMHC, and NO_x) were lower than the Tier II standards (without adjusting for catalyst deterioration factors). Concentrations of regulated air toxics (benzene, formaldehyde, acetaldehyde, and 1,3-butadiene) and the OFP (on the basis of MIR factors) were reduced by about 23 and 33%, respectively, using 23 and 25% oxygen-enriched air. Converter-out, THC, and CO emissions from the EPA's off-cycle (REP05, having rapid accelerations and decelerations during the driving cycle) were reduced by about 60 to 70% with moderate oxygen enrichment of intake air (up to 23% oxygen). In the case of an FFV operating on M85, the FTP converter-out, weighted averages of formaldehyde emissions with 23% or 25% oxygen-enriched intake air were lower than 10 mg/mile. During all the previous tests, bottled oxygen was employed to increase the oxygen content of ambient air to 23 or 25%. On the basis of test results, it appears that oxygen enrichment of intake air has the potential of becoming a viable technology for controlling exhaust emissions from light-duty vehicles, but the success of practical vehicular applications depends on the development of a less complex membrane system to supply the desired oxygen-enriched air in the engine intake.

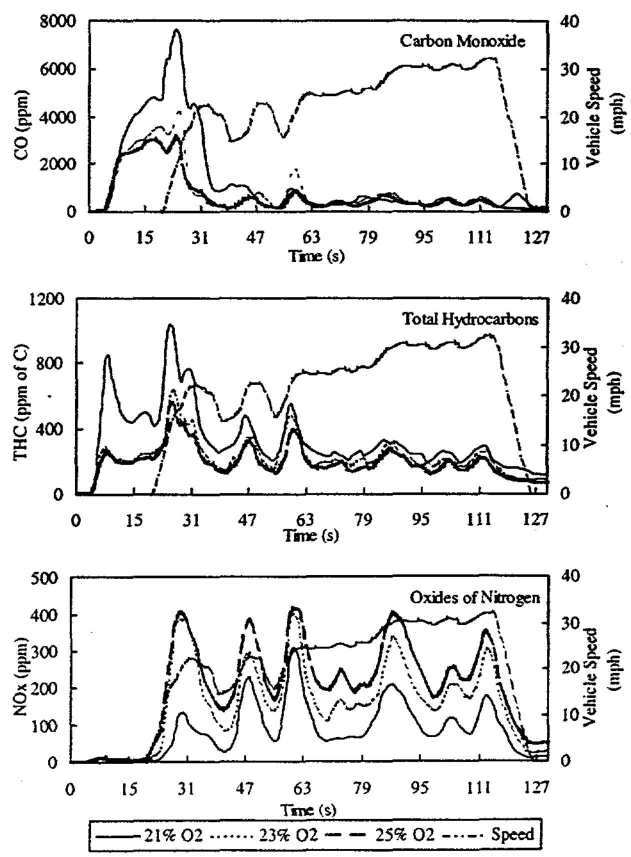


Fig. 1 Engine-out CO, THC, and NO_x emissions during the first 127 s of the cold-phase FTP cycle with Indolene fuel

RESEARCH OBJECTIVES

The objective of the present work is to demonstrate the operation of an air separation membrane supplying oxygen-enriched air in the engine intake of a light-duty passenger car. Another objective is to evaluate the membrane operating power requirements on the basis of prototype bench tests and from vehicle tests. The principles and operating characteristics of the air separation membrane are briefly reviewed. Vehicle tests are designed to show whether engine intake manifold vacuum could be utilized to maintain the required pressure differential across the membrane. The module size and additional power needed to operate the auxiliary equipment of the membrane during the initial start-up and warm-up periods of a light-duty vehicle are also evaluated.

MEMBRANE CHARACTERISTICS

The air delivered to engines can be enriched in oxygen by selective permeation through nonporous, polymeric membranes via a "solution-diffusion" mechanism. In this mechanism, air molecules dissolve into the nonporous membrane and then diffuse across it into a porous support layer. The ambient air travels across the membrane by means of a pressure gradient at the opposite ends of the module. The pressure differential across the membrane provides the driving force for the absorption and diffusion of oxygen and nitrogen molecules through the membrane material. Oxygen can diffuse more rapidly and becomes enriched in the low-pressure permeate stream, while nitrogen is concentrated in the retentate stream.

The performance of an air separation membrane module depends on the coated membrane material's intrinsic properties (permeability and selectivity) and on such other parameters as membrane polymer structure, skin thickness, coating morphology, geometry of the fibers (porous support), fiber dimensions, flow pattern, feed direction, feed conditions, percent recovery (or stage cut, permeate flow over feed flow), cartridge type, packaging density, and arrangement of separators. The influence of these key parameters on the performance of membranes can be found in References 8 and 9. The selection of a membrane to achieve the desired oxygen-enriched air flow is evaluated in terms of the power required to maintain the differential pressure across the membrane and the amount of space it occupies.

The geometry of the porous support (flat film or hollow fiber) influences the manner in which the module is packaged. Hollow fiber geometry (see Figure 2), which is self-supporting and offers excellent packaging compared to flat films, appears to be more suitable for automotive applications. Hollow fiber sizes ranging from 50 to 1000 μm are currently available. For compact modules, the skin thickness of a membrane coating on a porous hollow fiber support is critical; typically, this would be on the order of 1000 to 3000 Å, with 400 to 1000 Å attainable in more finely tuned membranes. The usable cartridges consist of coated hollow fibers arranged in small bundles and are sealed with an adhesive into header plates at opposite ends of the module, such that a large number of fibers could be included. Typically, a hollow fiber membrane module resembles a shell-and-tube geometry as shown in Figure 2.

Air separation membrane units can be operated in one of three modes: vacuum, pressure, or mixed modes. In the vacuum mode, the feed air is pressurized to only slightly above atmospheric pressure (about 1 to 5 psig), and a vacuum is maintained on the permeate side of the membrane. The retentate is vented at atmospheric pressure. The vacuum mode is typically more energy-efficient than the pressure mode, primarily because a vacuum is applied only on the permeate (product stream). However, because of the limited differential pressure, the vacuum mode requires a larger membrane area for a given flow rate than does the pressure mode. In the pressure mode, the feed air is typically pressurized (by an air compressor) to several atmospheres, while the permeate is maintained at about atmospheric pressure. Higher driving forces are obtained in this mode because the differential pressures are higher than those of the vacuum mode, resulting in reduced membrane area requirements. However, the pressure mode is more energy-intensive, because both permeate and retentate have to be compressed to higher pressures. Finally, in the mixed mode, the feed air is pressurized, and a partial vacuum is maintained on the permeate side to increase both the compression ratio and differential pressure, and thus, the oxygen concentration. The oxygen-enriched air stream from the permeate is then mixed with an ambient air stream to obtain the desired air flow and oxygen concentration. The selection of a preferred mode of operation depends on the economic tradeoffs among membrane area costs, auxiliary equipment costs, input power requirements (to maintain required pressure/vacuum), and system compactness.

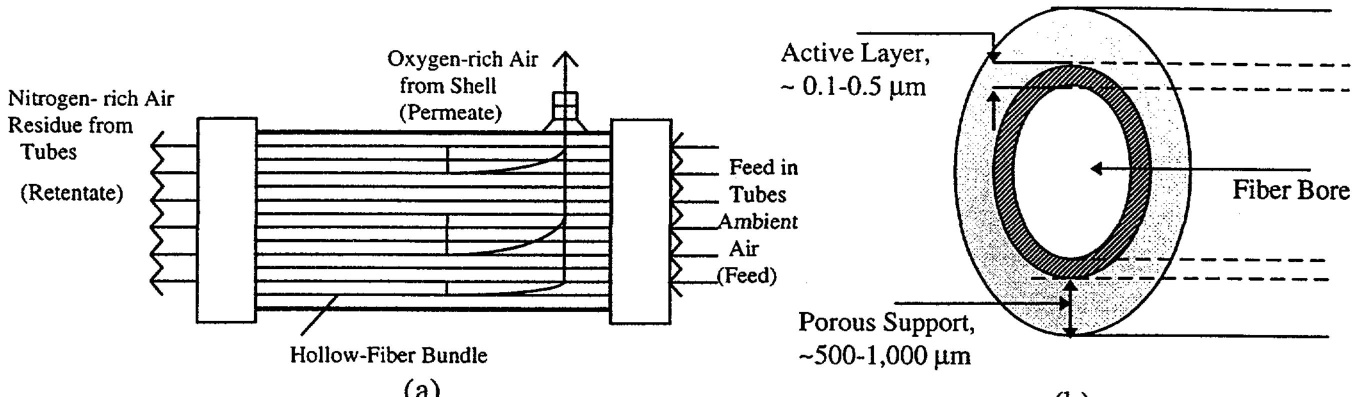


Fig. 2 Schematic of a counter-flow hollow-fiber membrane with shell-and-tube arrangement: (a) module geometry; (b) hollow-fiber geometry

EXPERIMENTAL TEST PROCEDURE

The experimental investigations consist of membrane prototype bench tests and demonstration of the membrane on a vehicle, as described below.

Membrane Prototype Bench Test Set-Up

It is important to examine various operating factors of the oxygen membrane module closely before installing it in an engine. Also, the design and operating conditions should be optimized for a specific application, such as a passenger car engine. In order to evaluate the performance of a membrane device under simulated engine operating conditions, a membrane bench test set-up was developed. Figure 3 shows the schematic test set-up to evaluate the prototype air separation membranes.

A hollow-fiber membrane module developed at Compact Membrane Systems, Inc. (CMS), was employed in the present tests. The module consists of a number of hollow fibers coated with a membrane material of Perfluoro-2-2-dimethyl-1-3 dioxole copolymerized with tetrafluoroethylene [10]. The physical properties of the coated membrane material and module are given in Table 1. Since the intended application is for use during the initial start-up and warm-up periods of light-duty vehicle operation, power requirements of the auxiliary equipment and size of the membrane module are critical for competing with other emissions reduction technologies. Hence, the vacuum mode was selected as the mode of operation because it offers the lowest power requirements (vacuum is applied only on the product stream) with respect to other methods. In order to examine the membrane performance characteristics, tests were conducted at different feed flow rates (ranging from 20 to 50 scfm) and at different permeate vacuum conditions (0 to 25 in. Hg). At each operating condition, the pressure (or vacuum at the permeate), temperature, flow rate, and oxygen concentration were recorded at the feed, permeate, and retentate sides of the membrane. Mass and oxygen balances were done to verify the test data. At any given operating condition, the absolute pressure ratio across the membrane is defined as the ratio between the average tube (feed and retentate) pressure and the average shell pressure. The power requirements of the blower and vacuum pump were calculated on the assumption of a constant 70% working efficiency.

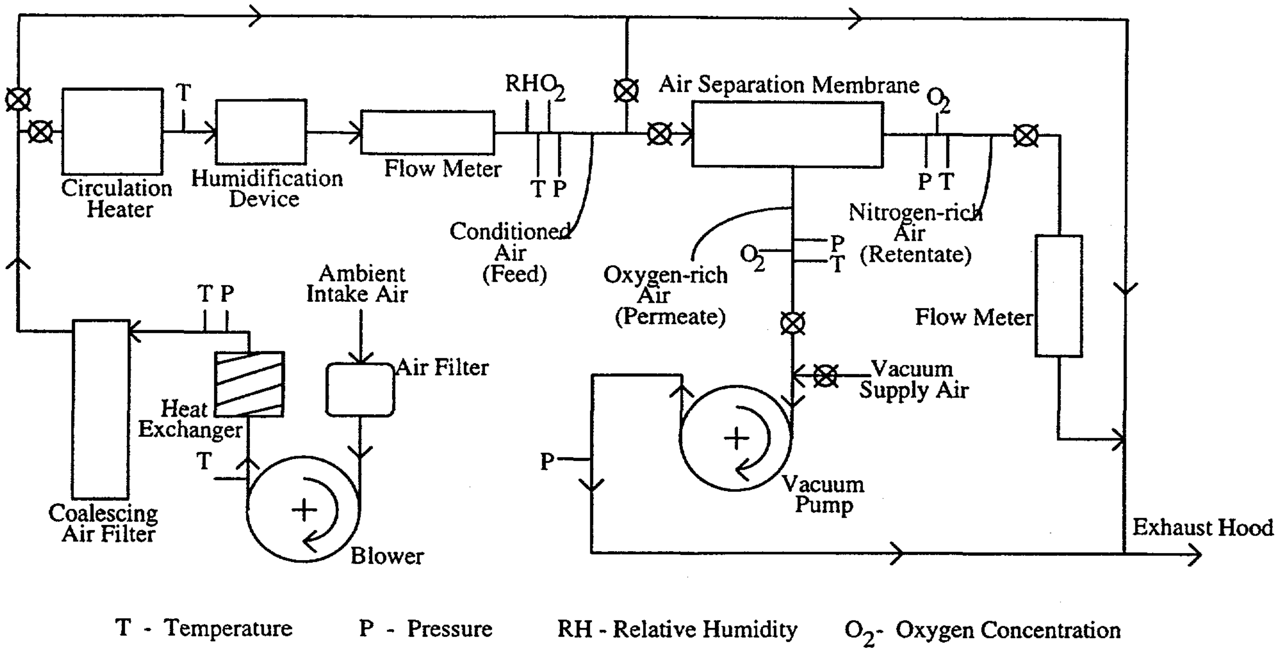


Fig. 3 Schematic bench set-up for testing prototype air separation membranes

TABLE 1 Properties of CMS membrane module

Property	Value
Permeability	
(cm^3(STP)-cm/cm^2-s-cm Hg $\times 10^{10}$)	
Oxygen	990
Nitrogen	480
Water vapor	4500
Selectivity (O_2/N_2)	2.05
Membrane skin thickness (μm)	0.3
Fiber geometry	Hollow
Fiber dimensions (OD/ID) (μm)	660/500
Membrane coated surface area (m^2)	5.6
Module dimensions (length $\times$ diameter), cm	68.5 $\times$ 7.6

Vehicle Demonstration Test Set-Up

One of the research objectives of the present work is to examine whether intake manifold vacuum could be utilized (instead of using an additional vacuum pump on the permeate side of the membrane) to maintain the required pressure differential across the membrane. Another objective is to examine the transient response of the membrane under engine operating conditions. In order to fulfill these objectives, the prototype membrane tested earlier on the bench set-up was installed on the engine intake. The existing air filter and air box were removed, and the intake manifold was connected directly to the permeate side of the membrane. An external air filter and electrically driven blower were connected on the feed side of the membrane to supply ambient air at different flow rates, under different feed pressure conditions. The PCV system was modified to draw intake air directly from the intake manifold without having the air pass through the crankcase. The purge line from the carbon canister to the engine was disconnected at the canister and relocated to a port at the intake manifold. The fuel tank vapor line was connected to a remote canister. This configuration caused the engine to draw only oxygen-enriched air from the permeate side of the membrane. The pressure (and vacuum), flow rate, and oxygen concentration were measured at the feed, permeate (intake manifold), and retentate sides of the membrane. The schematic vehicle test set-up is shown in Figure 4, and a photograph of the system is shown in Figure 5.

During the initial tests, the blower was switched off, and the engine was started with the membrane on the intake manifold to examine the engine breathing capacity and to verify the engine idle speed stability. In the second set of experiments, feed flow rate and pressure were maintained nearly constant, and the

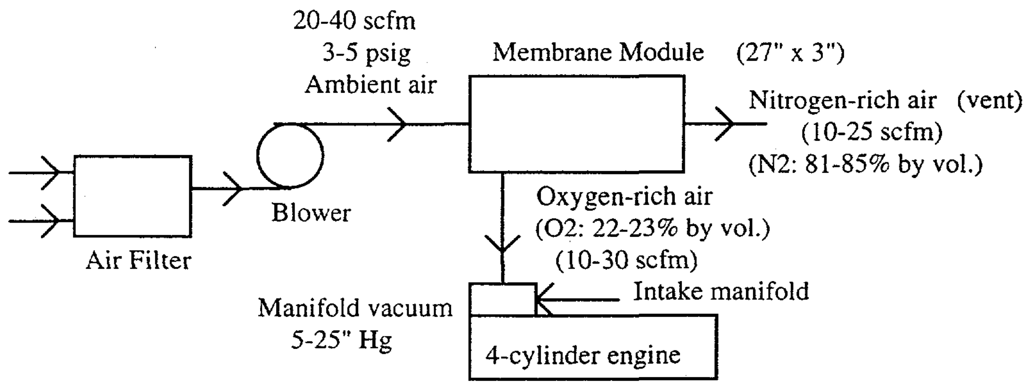


Fig. 4 Schematic test set-up of engine intake system with the membrane supplying oxygen-enriched air

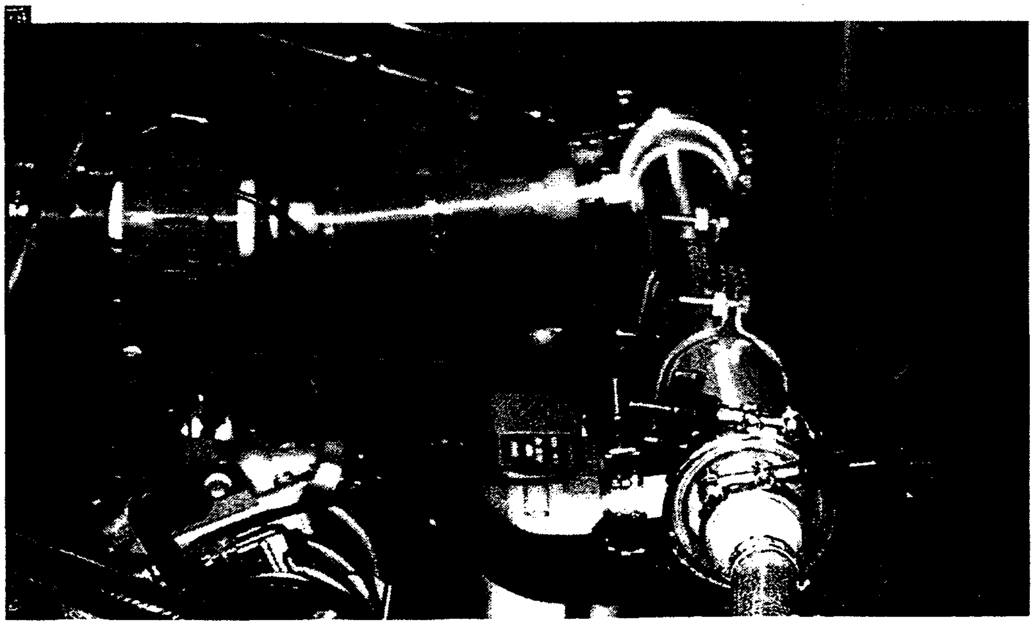


Fig. 5 Photograph of the membrane connected at the intake of a passenger car

position of the gas pedal (under free acceleration of the vehicle) was varied to obtain different intake manifold vacuum conditions. The position of the gas pedal was held nearly constant at any set intake manifold vacuum condition. By varying the vacuum at the permeate side, different pressure ratios across the membrane were established, supplying different oxygen-enriched air flows and oxygen content to the engine. No exhaust emissions data were recorded, because the investigators had prior knowledge of experimental data (replicated) with bottled oxygen for a similar (23%) oxygen-enrichment level.

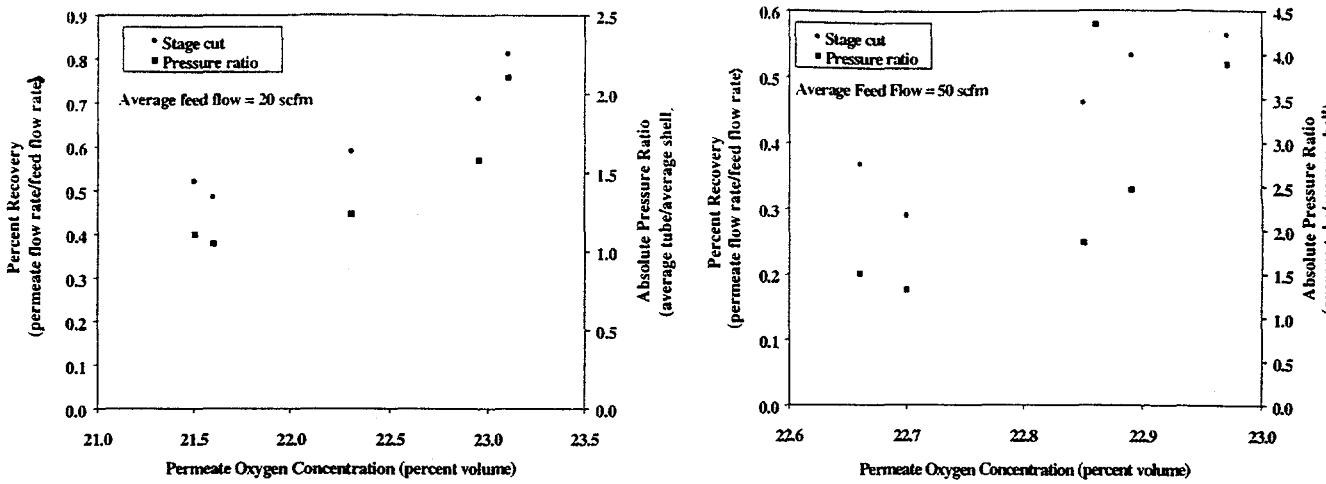
RESULTS AND DISCUSSION

Prototype Membrane Bench Tests

The permeability of a given gas is an intrinsic property of the membrane, while the degree of separation between the gases depends on the relative permeabilities of the gases to be separated. For a given membrane surface area and membrane material (permeability), the gas flow rate across the membrane is a function of trans-membrane partial pressure difference. Also, the rate of gas transport across the membrane is inversely proportional to the skin thickness of the membrane (active) layer, as described by Fick's law: $N_i = \frac{P_{ri} A \Delta P}{\delta}$, where N_i is the flow rate of gas i (cm^3 of STP/s); P_{ri} is the intrinsic permeability of gas i in the membrane (cm^3 of STP-cm/cm^2-s-cm of Hg); A is membrane area (cm^2); δ is the membrane separating barrier thickness (cm); and ΔP is the trans-membrane partial pressure difference of gas i (cm of Hg).

In a given membrane module (constant surface area) and at a constant feed flow rate, different permeate flow and concentration levels can be obtained by changing the absolute pressure ratio across the module. Figure 5 shows the permeate oxygen concentration and percent recovery obtained by varying the absolute pressure ratio across the membrane (between tube and shell side of the module) at two different feed flow rates (average flow rates of about 20 and 50 scfm). At both feed flow rates, as the absolute pressure ratio increases, the percent recovered from the feed and the permeate oxygen concentration level increase. But at higher absolute pressure ratios, the increase in permeate oxygen concentration is marginal compared with the increase in permeate flow. Since the maximum degree of separation is limited by the skin thickness of the membrane active layer (Eq. 1), any further increase in differential pressure causes more permeation (flow) than separation. The maximum oxygen concentration obtained under both cases is only about 23% by volume. It was observed that at a higher average feed flow rate of 50 scfm, the absolute pressure ratio required to cause a similar degree of separation (about 23% oxygen concentration) compared to a lower average feed flow rate of 20 scfm increased from 2.1 to 3.9. The permeate flow (or percent recovered from the feed flow) increased with the increase in absolute pressure ratio at both the average feed flow rates of 20 and 50 scfm. The maximum permeate flows recovered at 23% oxygen concentration were about 19 and 30 scfm, corresponding to the average feed flow rates of 20 and 50 scfm, respectively. Similar permeate flow and concentration trends were obtained at other feed flow rates tested. Test results suggest that there exists an optimum condition where higher permeate flow and concentration levels could be obtained at a modest absolute pressure ratio. In order to attain higher permeate flow and concentration levels with a given membrane material, higher membrane surface areas are required. This can be achieved by simply increasing the number of fibers in the module (larger module) and/or decreasing the thickness of the active layer.

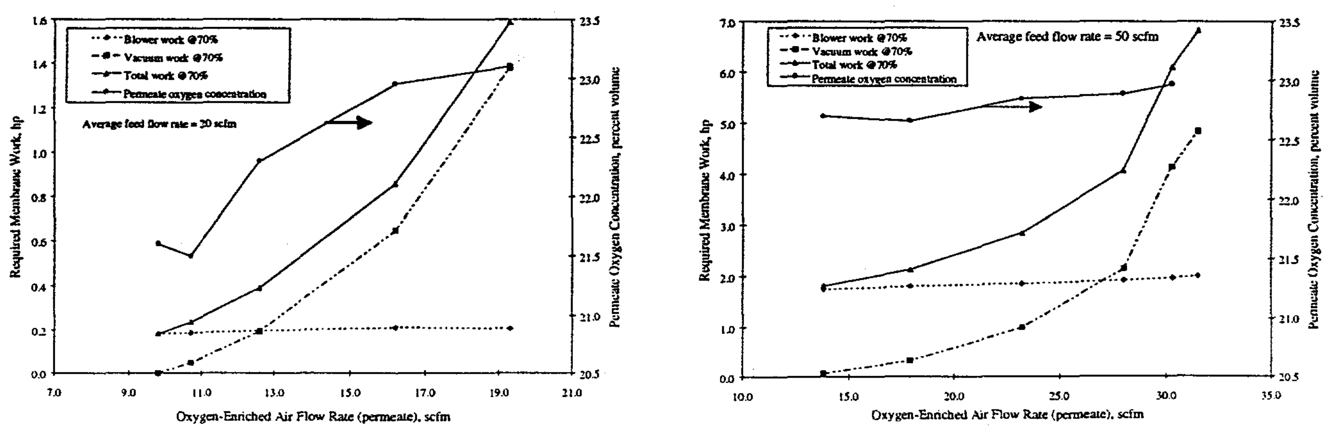
In order to maintain a pressure differential across the membrane to cause air separation, auxiliary equipment (such as a blower and a vacuum pump) are required (membrane operating in vacuum mode)



(a) (b)

Fig. 6 Variation of permeate oxygen concentration with percent recovery and absolute pressure ratio (a) average feed flow = 20 scfm; (b) average feed flow = 50 scfm

at the feed and permeate sides, respectively. The purpose of maintaining a positive air flow at the feed is to overcome the pressure loss in the tubes so that separation sweep occurs along the entire length of the tubes. In the absence of a positive feed, a lower pressure (vacuum) at the permeate causes incomplete sweep (poor separation) such that air will be pulled from both sides of the tubes (feed and retentate). Therefore, it is necessary to maintain a positive air flow at the feed and a lower-pressure (vacuum) level at the permeate for good separation to occur. The total membrane driving force is the sum of the power required by the blower and vacuum pump to maintain their respective pressure (or vacuum) conditions. The power requirements to attain different permeate output levels (oxygen concentration and flow) at average feed flow rates of 20 and 50 scfm are shown in Figure 7.



(a) (b)

Fig. 7 Power requirements of the membrane module operating in vacuum mode (a) average feed flow = 20 scfm; (b) average feed flow = 50 scfm

Power requirements of the blower remained constant (about 0.2 hp with 20 scfm, 1.8 hp with 50 scfm) regardless of the permeate output, because feed flow rate and pressure were held constant. Conversely, power requirements for the vacuum pump increased with permeate output, because the corresponding absolute pressure on the permeate was decreased. To obtain nominal 23% oxygen-enriched air and maximum flows of about 19 and 30 scfm (corresponding to 20 and 50 scfm feed flow) at the permeate, the total power requirements of the membrane are about 1.6 and 6.0 hp, respectively. A tradeoff appears to exist between the permeate output and total power requirements. Membrane operating conditions and auxiliary equipment size should be custom-optimized for any given application.

Vehicle Demonstration Tests

In preliminary vehicle tests with the membrane on the intake manifold, the engine started with no difficulty, without the help of an external blower. The intake manifold vacuum was able to produce the pressure differential across the membrane and sustain a steady idle speed. However, there was little oxygen enrichment of air passing though the engine intake, due to the poor separation sweep across the membrane (discussed earlier). When a positive, constant feed flow (about 35 scfm) was maintained with the help of an external blower, the vacuum at the intake manifold generated an adequate pressure differential across the membrane for separation. Under free acceleration of the vehicle, the gas pedal was adjusted to maintain a maximum of about 21 in. Hg at the intake manifold. During both engine idle and free acceleration, the membrane could supply enough oxygen-enriched air to the intake for the engine to breath freely for rapid acceleration. There were no engine hunting (breathing) problems; the transient response characteristics of the membrane were found to be very satisfactory. In the absence of an additional vacuum pump (dispensed with manifold vacuum), the power requirements of the membrane are low, because only blower power for maintaining a positive feed is needed.

Figure 8 shows the external blower power required to maintain a constant feed flow of about 35 scfm to obtain different permeate outputs corresponding to different vacuum levels. Less than one horsepower is sufficient to obtain permeate flow of about 20 to 25 scfm at about 23% oxygen-enriched air. Since the intended application is to use oxygen-enriched air only during the initial start-up and warm-up periods for light-duty vehicles (about 20 to 60 s after key-on), the present module and a blower rated to produce 30 to 40 scfm air flow at 4 psig pressure would be sufficient.

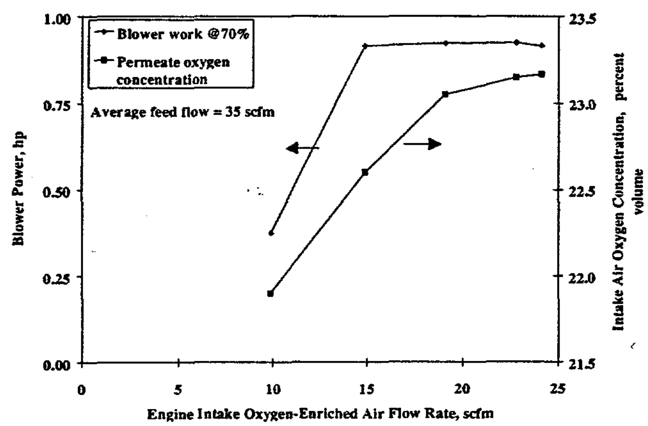


Fig. 8 External blower power requirements during the initial start-up of the vehicle

The humidity of ambient air does not appreciably affect membrane performance, because water vapor readily permeates across the membrane (see Table 1 for comparison of permeabilities). However, membranes are highly susceptible to oil; an oil film significantly affects the sorption characteristics. An electronically controlled bypass can be designed so that the membrane can be activated only during the initial 20 to 50 s after key-on; thereafter, ambient air directly from the air filter could be used. At the time of writing, a membrane coating thickness of 0.1 μm has been attained with a new coating technique at CMS, resulting in more compact modules and higher separation factors (producing >26% oxygen concentration) with considerably lower operating power requirements [11]. A small blower, electrically driven by an alternator, and a compact membrane module (about the size of an air filter, with advanced membranes) can be suitably packaged underneath the hood of a car.

SUMMARY

Prototype air separation membranes are available to supply oxygen-enriched combustion air for SI-engine-powered, light-duty vehicles to potentially reduce regulated exhaust emissions, particularly during the initial start-up and warm-up periods of vehicle operation. Bench-scale tests on a prototype membrane indicate that several membrane physical and operating parameters need to be optimized to obtain the desired oxygen-enrichment and air flow levels. A membrane module 27 in. long and 3 in. in diameter is sufficient to deliver about 23% oxygen-enriched intake air (with an air flow up to about 25 scfm) during the initial operating period of a 2.5-L FFV; less than one horsepower of additional power is needed to operate the blower. With rapid advances in the development of newer membrane materials and better coating techniques (≤0.1 μm thickness), oxygen enrichment of combustion air has the potential to become a more practical technique for controlling exhaust emissions from light-duty vehicles, in particular during the initial start-up and warm-up periods (up to 60 s from key-on).

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