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THE USE OF METHANOL IN TRANSPORTATION

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THE USE OF METHANOL IN TRANSPORTATION

ABSTRACT

Information is presented to supplement the January, 1974 LLL report (UCID-16442) to the AEC on methanol fuel utilization and its environmental impact.

INTRODUCTION

This report is intended to supplement UCID-16442, which contained the Laboratory's contribution to the AEC methanol report. The chief thrust of this earlier document was methanol fuel utilization and its environmental impact. This report's primary emphasis is to explain the interwoven facts of methanol with other fuels, engines, and emissions. A simplified version of those relationships is presented along with an outline of our beginning efforts for a "Coal to Cars" program.

FUEL TYPES AND CHARACTERISTICS

History

Coal-based fuels including methanol* have emerged as an alternative to petroleum fuels where petroleum supplies were limited and the economics permitted. Great Britain, with a relatively high standard of living, no petroleum, and an available coal supply, has used coal-based fuels, particularly benzole, for some time. This coal tar distillate consists of about 70% benzene, 25% toluene, and 5% xylene.¹ At first, benzole was mixed with gasoline. More recently, a blend of mostly benzole with some high grade gasoline has served as a high-octane premium fuel for high compression engines. The first fuel required some minor carburetor modifications, the second fuel, none.^{†1}

*Methanol, methyl alcohol, and methanol alcohol are used here as synonymous terms.

[†]Emissions were not considered.

The use of alcohols as transportation fuels also has historical precedent. Extensive usage in Europe, Central America, and South America was evident prior to World War II.² An article written in the U.S. in 1936³ questioned the reasoning behind 11 European countries requiring some use of alcohol as a motor fuel. (In 1934, the 11 countries consumed 600,000 U.S. tons of alcohol as motor fuel.) Apparently, the need for a capability for munitions production and self-sufficiency during future hostile actions was not clear from this side of the Atlantic. The U.S. also had considerable distribution of alcohol-blends in the Midwest during the 1930's.⁴ The differences in fuel/air requirements were seemingly ignored, which probably caused some drivers irritation, but then again, do-it-yourself adjustments were common with auto owners. Ethanol was often proposed as a fuel to stimulate greater agricultural production during periods when food consumption and production were balanced unfavorably to industry. Published reports were for the use of alcohol if the financial support was from agricultural industries or areas and against its use if backed by petroleum interests — a logical difference of opinion in a free enterprise society with unlimited resources.

Methanol blends have long been used in racing. Alcohol, 80%; acetone, 10%; nitrobenzene, 5%; and gasoline, 5% is a typical motorcycle racing fuel. Pure methanol used in a turbocharged piston engine produces a startling 850 hp in an Indianapolis racing machine of quite limited displacement.

Alcohols have been mixed with benzoles. The London General Omnibus Company did extensive tests on alcohol-benzole blends and found a 50-50 mixture very satisfactory for London buses.*

Coal gas was used extensively during World War II by England, New Zealand, Australia, and Germany. Several reactor designs exist in which the retort is carried on the vehicle (or trailer) and the coal and water are converted to gas as needed. The gas was stored in inflatable bags on the automobile roof.

* The article implied that the fuel was not being used because of the high price of alcohol.

Methanol and Other Fuels

We emphasize methanol fuel* in this report because with today's technology the methanol process has the highest efficiency in converting coal to a liquid fuel. Methanol has an energy recovery factor between 50-55%, while other fuels, such as synthetic gasoline, result in about 20% additional loss and have an energy recovery factor between 40-45%.⁵ Undoubtedly, improvements will be made in the processes to increase the yields, but as long as methanol remains ahead of the field by several percent, it does not seem likely it will be replaced. A word of caution should be added here as the clear preference for methanol is qualified by today's technology, meaning proven, ready-to-go chemical processing techniques. Other methods of converting coal to clean fuels, such as liquifaction[†] appear promising but are at best in the pilot plant stage. The step from small experiments to large industrial plants is a giant one and involves risks, unknowns, and perhaps most importantly — time.

For comparison purposes, Table 1 lists some of the fuels that can be made from coal along with some of their properties.^{6,7} Synthesis gas from coal is CO_2 , CO , and H_2 , and, on a laboratory scale, all the products listed in the table could be constructed from these basic building blocks. Once we leave the laboratory, however, and embark on a production program of 10^{16} Btu's of energy yearly (the amount of gasoline currently consumed by vehicles in America), the economics, availability of raw materials, convenience, social acceptance, safety restraints, pollution effects, and production byproducts will narrow the field to a few practical choices.

To understand Table 1, some interesting points about hydrocarbon fuels should be noted: higher hydrogen-to-carbon ratios have higher heat values; longer chain molecules (i.e., higher x and y in the formula C_xH_y) have higher boiling points and lower octane ratings; branched chain or

* A fuel containing methanol, plus other compounds formed during methanol synthesis, and necessary additives.

[†] Liquifaction is the process in which hydrogen content of the coal is increased, changing it physically from a solid to a crude oil type liquid. This liquid is amenable to production of synthetic gasoline.

Table 1. Possible hydrocarbon fuels for transportation.^{6,7}

Fuel		Boiling point (°F)	Octane rating ^a	High heat value (Btu/lb)	Ideal Air/Fuel ratio (wt)
PARAFFINS					
CH ₄	Methane	-259	~110	23,850	17.2:1
C ₂ H ₆	Ethane	-127	~104	22,400	16.0:1
C ₃ H ₈	Propane	-44	100	21,650	15.6:1
C ₄ H ₁₀	Butane	31	92	21,300	15.4:1
C ₅ H ₁₂	Pentane	97	61	21,100	15.3:1
C ₈ H ₁₈	Octane	258	-17	20,750	15.1:1
C ₈ H ₁₈	Iso-octane	211	100	20,580	15.1:1
2,2,4 trimethyl pentane (standard for gasoline comparisons)					
NAPHTHENES					
C ₅ H ₁₀	Cyclopentane	121	83	18,780	14.7:1
C ₆ H ₁₂	Cyclohexane	177	77	18,710	14.1:1
AROMATICS					
C ₆ H ₆	Benzene	177	~110	18,160	13.2:1
C ₇ H ₈	Toluene	231	~104	18,300	13.4:1
C ₈ H ₁₀	Xylene-m	285	~105	18,500	13.6:1
OLEFINS					
C ₃ H ₆	Propene	-54	85	21,000	14.7:1
C ₄ H ₈	Butene-1	20	80	20,900	14.7:1
ALCOHOLS					
CH ₃ OH	Methanol	148	98	9,770	6.4:1
C ₂ H ₅ OH	Ethanol	173	99	12,780	9.0:1
C ₃ H ₇ OH	Propanol	208		14,500	10.5:1
C ₄ H ₉ OH	Butanol	244	~100	15,500	11.1:1
OTHER					
H ₂	Hydrogen			61,030	34.2:1
~C ₇ H ₁₇	Automotive gasoline		86-100	21,050	15.3:1
	Diesel fuel			19,000	
~C ₇ H ₅	Coal			10-15,000	

^aThe octane number of a fuel is a measure of that fuel's resistance to combustion knock. Normal combustion processes have flame speeds or chemical reactions proceeding at 50 to 150 ft/sec velocities. The unburned gases ahead of this flame front are subject to an increase in temperature and pressure and, if spontaneous inflammation takes place, the rapid pressure rise is the characteristic knock. A comparison is made with isooctane rated at 100 and heptane rate at 0.

more complex compounds have higher octane ratings (i.e., isooctane vs octane); alcohols have much lower heat values than similar C-H compounds because of the OH radical; and longer chain alcohols more nearly approach the heating value of other CH compounds as the effect of the OH radical becomes less important. Note that for the alcohols the Air/Fuel (A/F) ratio steadily increases with molecular weights. These points are graphically present in Fig. 1, which compares paraffin hydrocarbons to the critical compression ratio.⁸ The relationship between the critical compression ratio and octane is indicated in Fig. 2. Engine combustion chambers can vary the curve somewhat, with current U.S. engines at 8.5 compression ratio and 91 octane. Figure 3 compares the critical compression ratio of a variety of hydrocarbons to the number of carbon atoms in the ring or chain.

The lower boiling point fuels listed in Table 1 (hydrogen, methane, propane, etc.) require pressurized containers and/or lowered temperatures for storage, and their use in an engine is in the form of gas mixed with air. These gases have storage problems because more space is required; a weight problem because of heavier vessels; and a cost problem because all vessels are more complicated than the simple and hard-to-beat gasoline tank.

Higher boiling point compounds are used in a vapor (gas) and liquid mixture with air; gasoline is an example. The presence of a liquid phase with much higher density allows more charge and thus more power (~10-15%) to be utilized in a given engine size, but it also increases the difficulty of minimizing exhaust pollutants over normally encountered temperature ranges. A/F ratios are usually near stoichiometric;* a slightly richer mixture aids ignition and flame propagation but results in increased unburned hydrocarbons and carbon monoxide in the exhaust.

Still higher molecular weight compounds with less volatility are usually metered directly into the combustion chamber and ignited or

* Stoichiometric is the term used to designate the exact chemical proportions needed to obtain a complete chemical reaction.

Fig. 1. The critical compression ratios of pure paraffin hydrocarbons (600 rpm and 350°F manifold).⁸

Fig. 2. Relationship of critical compression ratio and octane for typical engines.¹

Fig. 3. The critical compression ratios of pure hydrocarbons (600 rpm, 150° manifold, 350° F coolant).⁸

compressed to self-ignition temperatures. Combustion chamber fuel injection in the diesel engine is one method, continuous burning turbine engines another. These systems can tolerate much leaner fuel mixtures (high A/F ratio).

Energy density and convenience of handling are two other important considerations in fuel selection. Methanol, like gasoline, is a liquid and has a fairly high energy density compared to the alternatives. Table 2 lists some of today's often referred to replacements for gasoline. The table indicates some natural groupings of the fuels. Gasoline and the alcohols (Group 1) are stable liquids at normal temperatures and pressures, and thus, are suitable to the existing national fuel distribution system. A second group includes gases such as propane and butane, which can be liquified by moderate pressures or small decreases in temperature. They are probably an acceptable fuel substitute, but their use would require huge capital expenditures for the construction of new distribution and storage facilities. A third group consists of gases, such as hydrogen and methane, which require high pressures or cryogenic temperatures for storage in any significant quantities. For effective use, the many problems created by this group need to be solved, and a large capital investment would be required. They are definitely fuels of the future.

Blended Fuels

Alcohol blends with gasoline have been proposed since the early 1900's. They were initially suggested as a way to increase the consumption of agricultural and wood products. Combinations of alcohol and gasoline have been tried to increase power and octane rating. For instance, methanol and water were injected into supercharged aircraft engines in World War II when emergency power was needed. Methanol up to 25%¹⁰ and ethanol up to 20%¹¹ have been tried experimentally in an attempt to understand and alter the combustion and emission products. Figure 4 indicates the change in A/F ratios needed to accommodate various methanol-gasoline blends. Note that additions of alcohol cause the A/F ratio to decrease. The often suggested and tried addition of alcohol to a gasoline-fueled engine with a

Table 2. Alternate fuels compared to gasoline.^{1,6,7,9}

Fuel	Density (lb/ft ³)	Energy density		Amount equivalent to 20 gal of gasoline		Weight of tank with fuel equivalent to 20 gal of gasoline (lb)
		(Btu/lb) (low heat value)	(Btu/ft ³)	(gal)	(lb)	
GROUP 1						
Gasoline Liquid	43.8	19,080	835,700	20	117	150
Methanol Liquid	49.7	8,640	429,400	38.9	258	310
Ethanol Liquid	49.6	11,550	572,900	29.2	194	235
Ammonia Liquid	48.1	8,000	384,800	43.3	279	335
GROUP 2						
Propane Liquid, -44°F/100 psi	31.8	19,900	632,800	26.4	112	180
Butane Liquid, 31°F	34.6	19,680	682,000	24.5	113.5	-
GROUP 3						
Hydrogen						
Gas, 2000 psi	0.667	51,980	34,650	482	43.0	4600
Gas, 3000 psi	0.89	51,980	46,500	359	43.0	-
Liquid, -423°F	4.43	51,980	230,300	72.6	43.0	300
Metal Hydrides						
Mg ₂ Ni·Hx	110	4,350	479,900	34.8	513	625
MgH ₂ (40% void vol)	54.7	3,950	216,200	77.3	565	-
Methane						
Gas, 2000 psi	7.09	21,500	152,400	110	104	1100
Gas, 3000 psi	11.1	21,500	218,000	76.6	104	1100 ^a
Liquid, -259°F	26.6	21,500	573,000	29.2	104	-

^aSpace needed for four gas cylinders, 1 × 4 × 8 ft.

Fig. 4. A/F relationships for various methanol-gasoline blends.

fixed A/F ratio makes the mixture leaner as the proportion of alcohol is increased. Viscosity and density changes have unpredictable effects on some carburetors; but many pre-emission era systems, adjusted for maximum power and performance (lower A/F ratio than stoichiometric), run very well on blends up to 20% ethanol.* A recent report¹² showed several types of automobiles produced between 1967 and 1972 have improved fuel economy, decreased CO emissions, and in eight out of nine cars, a reduction in exhaust gas temperatures with blends up to 15% methanol.

Blending of alcohol and gasoline can make a very desirable fuel. Small alcohol percentages (less than 20%) can increase the antiknock qualities of low octane fuels (below 98) and substitute for antiknock compounds such as tetraethyl lead (TEL). Blending can promote a smoother combustion process as the high heat of vaporization on compression of the fuel's liquid portions somewhat reduces the initial temperature and pressure. Partial decomposition of the alcohol before arrival of the flame front also probably takes place. This can absorb up to 20% of the heat of combustion energy and return it to the cycle during the burning process. There is also a greater tolerance for lean conditions before the flame is quenched, probably because of hydrogen production during dissociation. This allows the reaction to proceed further into the more remote combustion chamber volumes, which will reduce carbon deposits, and hopefully, hydrocarbon and carbon monoxide emissions.

An interesting situation has been developing in this country since 1971 regarding the antiknock compounds that have been blended with gasoline to increase the octane. An article in American Scientist¹³ discusses the effects of the Clean Air Act, the national fuel policy, and a number of other interrelated problems which are causing the fuel and auto industries to initiate several far-reaching changes. The article states that the compression ratio (CR) of auto engines has been reduced from 9.5 to 8.5,

* An ethanol curve drawn like Fig. 4, would have a 100% point at 9.0/1 rather than 6.4/1, indicating the total system is somewhat less sensitive to blend variations.

causing a 12%* decrease in fuel economy. The 91 Research Octane Number (RON) fuel needed for the 8.5 CR engines has been obtained from 88.5 RON raw pool gasoline by the addition of lead additives. The decision to remove lead, as it contaminates the catalyst used to control emissions, will force the refining process to produce higher quality (91 RON) fuel at a cost of a \$2-4 billion capital investment with an increase in cost to the motorist of \$6.5-8 billion (\$65-85 per vehicle) per year. An additional penalty is the increase of 6% in total crude oil that is necessary to produce the higher quality fuels.

Continuing beyond the article's intent to indicate the cost of our present policy decisions, consider the second shock wave of turning to a new and additional source of fuel in the near future. It seems an appropriate time to start methanol production from coal, use it as the octane booster rather than the higher quality gasoline, and invest the savings in development of the new fuel source.

Alcohol-gasoline blends will benefit from alcohol's solvent action on the gasoline-derived gums and varnishes that tend to collect in the carburetor, intake manifold, valves and fuel lines. On the negative side, there is some evidence that alcohol corrodes some pot metals and occasionally, iron and copper materials.¹

Although alcohol and gasoline of high aromatic content are completely miscible, a problem arises with the addition of water. Depending on the alcohol, temperature, and blend, the water causes a two-phase separation; the alcohol, water, and some higher aromatic portions of the gasoline settle to the bottom of the tank. With ethanol and premium gasoline, separation occurs with 0.1-2.5% water in a temperature range from 0°-100°F.³ Methanol seems to be an order of magnitude less tolerant of water than ethanol.¹⁴ Additives to increase water tolerance have been

* The change in otto cycle thermal efficiency is not this high; however, to provide performance, the effects of increasing the engine displacement, added weight, etc. are totaled. Auto manufacturers have also stated this type of loss.

used, such as benzole, benzene, acetone, and butyl alcohol. J. Bolt noted in his paper¹⁵ that Europe has been using blends for years, so evidence exists that the problems are not insuperable.

Methanol and water as a blend could have an important role in methanol marketing. In the last few months, there has been a renewal of interest in adding water to gasoline with some questionable news stories reporting very large decreases in fuel consumption. One individual has succeeded in actually homogenizing water and gasoline with a high pressure pump. Oldsmobile* in the sixties had water injection available on some models; I believe those models also included the exhaust-turbo charger. The overall effects of water additions to the gasoline combustion process have been discussed in a paper by Nicholls, El-Messiri, and Newhall.¹⁶ On a single-cylinder experimental engine, additions of water (into the intake manifold) of 50-75% of the weight of fuel, reduced the fuel consumption by a few percent and were acceptable in amounts up to 150% with little or no gain in economy. At that point the water had little effect depending on A/F ratio (rich conditions - a few percent greater consumption; lean conditions - a few percent less). The importance of their experimental work, however, was not the small, but significant, reduction in fuel consumption but a drastic reduction in nitric oxide emissions. To quote their abstract, "For a water injection rate 1-1/4 times the fuel consumption rate, nitric oxide reductions of over 90% were achieved". They attributed this reduction to the addition of water because "the molar energy density of the combustible charge is effectively reduced."¹⁶ These data lead to the conclusion that water-methanol blends should be investigated. An important plus over gasoline-water systems is their miscibility, which could allow them to be mixed in the fuel tank. The importance of the reduction in nitric oxide is discussed in the emissions and control section of this report.

* Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U.S. Atomic Energy Commission to the exclusion of others that may be suitable.

Decomposed Fuels

Decomposed or dissociated fuels are not presently used in any system today outside of research projects,¹⁷⁻¹⁹ but they may prove to be a solution to fuel and pollution problems. Complex compounds of hydrocarbon fuels or their derivatives can be broken down (in a process similar to coal gasification) by heat and pressure with water to form simple products, CO and H₂. These products can then be burned under controlled conditions to produce minimal objectionable emissions. If exhaust heat can be utilized in the dissociation, an additional advantage of increased efficiency may be realized. The drawback is that expensive and perhaps bulky additional equipment is required. The possibility, however, that low grade fuels may be made acceptable by this method should support considerable research effort in this area.

FUELS AND THEIR EFFECTS ON ENGINE DESIGN

Methanol and Gasoline

Narrowing the discussion to a specific comparison of methanol and gasoline to get a view of the differences in the two fuels, we have:

<u>Factors</u>	<u>Methanol</u>	<u>Gasoline</u>
High heat of combustion/lb	9700 Btu	21,000 Btu
A/F ratio	6.4:1	~15:1
Heat of vaporization/lb of fuel	502 Btu	117 Btu*
Heat of vaporization of fuel/lb of air consumed	78.4 Btu	7.8 Btu*
Boiling point	148°F	Variable (100-400°F)
Vapor pressure vs temperature	Fixed curve	Variable

Other items are as follows. The power available from a methanol-fueled engine is within ±10% of a gasoline-fueled engine depending on amount of the fuel that is in a vapor state with the air charge (liquid phase-more power; vapor phase-less power). Flame velocity is higher with methanol at high (lean) A/F ratios.²⁰ Lean conditions are less likely to cause misfire (no ignition of mixture) with methanol.²⁰

* Isooctane

The differences can be corrected by a few engine modifications:

- The engine fuel supply system for methanol must have about twice the capacity of a comparable gasoline supply system.
- If the engine design requires partial or fully vaporized fuel, much more heat must be supplied for methanol fuel vaporization.
- Since methanol fuel liquid phase will probably be the greater portion of the total fuel supplied because of its singular vapor pressure and higher heat of vaporization, greater care will be required in the liquid distribution to individual cylinders.

Two advantages of methanol over gasoline are: one, methanol fuel has a high octane rating (98) and neglecting emission results, the compression ratios and thus efficiency could be increased; and two, the higher flame velocity and extended misfire limits (under lean conditions) could produce fewer emissions.

The major disadvantages are cold engine starts will be more difficult to obtain, possible corrosiveness, and increased fuel bulk.

Spark Ignition Engines

Carburetion is probably the least expensive and least precise method of metering and mixing fuel. With perhaps a single model exception, this system is used on all U.S. production. As previously stated, carburetors now in use could tolerate a small amount of alcohol blended with gasoline, probably 10%.²¹

Above 10%, a carburetor retrofit is possible to adjust the fuel flow to a chosen blend. The upper limit of this simple modification is determined by the need for additional heat to the intake manifold to account for the change in the heat of vaporization. To obtain the additional heat, some manifold designs will be more suitable than others, and the problem of adjustment becomes more difficult. Still higher alcohol contents require increased heat exchange between the exhaust manifold and the intake manifold, and either a heat supply system for starting or an

additional volatile fuel fraction capable of an Air/Vapor (A/V) flammable ratio* at the temperatures encountered.

Fuel injection systems, unlike carburetors, do not attempt homogeneity of the air-fuel mixture outside of the combustion chamber. The fuel is forced, under pressure, through nozzles placed either in the intake manifold or in the combustion chamber. The first method, with nozzles placed close to the intake valve, is rapidly being accepted in the European automotive industry as a method of meeting the U.S. emission laws while still providing economy and performance. These designs can be adapted to provide the proper A/F ratios for other fuels by changing the amount of fuel injected.

The California Air Resources Board Report of January 9, 1974, which is a summary of 1974 model year vehicles approved for purchase in California, listed the following fuel-injected engines without other notations (Table 3). All other autos listed had engine modifications, air injection, exhaust gas recycle, or thermal reactors, as one or more exhaust control systems incorporated into the design. Using low emission producing methanol

Table 3. Fuel-injected engines approved by the 1974 California Air Resources Board Report.^a

Manufacturer	Engine family	Engine size CID	Models
Bayerische Motoren Werke Ag.	BMW 121 FI	121.3	BMW 2002 tii
Porsche	I	163.9	911T, 911S, 911 Carrera
Renault	807	95.5	Renault 17 Gordini Cpe
Saab-Scania	BE20	121	Saab LE, EMS, CCZ, and 4-door sedan
Volkswagen	3	120	914/4 roadster

^aThe VW 412 which is essentially the same engine as the 914 appears second only to the Honda Civic on the EPA economy data, a startling achievement when weights are compared (2300 vs 1500 lb).

* Air/Vapor (A/V) ratios are more significant than A/F ratios when lower temperatures or starting is discussed. A/V ratios refer only to vaporized fuel as the liquid portions do not contribute to the flammable mixture or start.

fuel with the low emission producing fuel-injected car could have very positive effects on air pollution.

The second method of fuel injection, combustion chamber injection, is capable of precise fuel metering and is similar to diesel systems in refinement and cost. An engine employing injection of this type can run at much higher A/F ratios than spark ignition engines using carburetion. No present production is known, but the Honda CVCC (compound vortex controlled combustion) could be declared an indirect descendant as it utilizes the stratified charge* principal discovered in the combustion chamber injection engines years ago. Ford PROCO (Programmed Combustion) and Texaco have similar development programs.

Compression Ignition Engines

Space and time do not permit a complete coverage of the multitude of engine designs conceived over the years. Presently ranking second in popularity to the SI Otto cycle engine is the diesel or compression ignition (CI) engine, the prime mover of heavy equipment. It is inherently more efficient, particularly at part throttle, than the spark ignition engine as it operates at higher compression ratios and unrestricted air intake. It is also capable of combustion at very lean mixtures because of the stratified charge combustion; however, weight per horsepower output and initial costs have kept it from other than a token usage in automobiles. A new trend toward diesel engines in cars may be in the offing with Peugeot's new diesel which started production this year. At 1357 cc and 45 hp, it is a smaller engine than their 504 diesel and operates at 5000 rpm, which is very high for a compression ignition engine. The station wagon weight with this engine installed is only one hundred

*Stratified charge is the term used for a localized rich mixture (low A/F) in the combustion chamber which burns at low initial temperatures with the flame front proceeding outward to lean mixtures resulting in low total emissions.

pounds more than the gasoline version and will compete in cost with European economy autos. Recorded mileage, obtained from the severe European Driving Code (DIN 70030) test, was 40 mpg.

Contrary to one or two articles recently published in the literature, methanol is not a fuel for all types of engines. It can be a substitute for gasoline in spark ignition engines, but it is not a substitute for diesel oil. The CI engine relies on a fuel that is self-igniting and requires low temperature, spontaneous combustion characteristics. This is opposite to the high octane requirements for which methanol is noted. One possibility, however, for utilizing methanol in the CI engine, is to blend methanol with a second fuel that can contribute the low temperature ignition traits.

Other old and new concepts, such as turbines, steam cars, and Stirling engines, can be categorically grouped as continuous fuel burners. These engines have a wider tolerance to variation in fuel characteristics. They can use methanol or heavy hydrocarbon fuels, such as diesel fuels. They also have flexibility in the A/F ratio required; therefore, designing for excess air consumption will lower emissions.

EMISSIONS AND CONTROLS

Federal and State Standards

The federal standards developed in response to the 1970 Clean Air Act are shown in Table 4. This list is the result of the amendment of July 2, 1971

By FTP in this table is meant federal test procedure. The driving cycle is the California 7-mode cycle repeated nine times. Pollutant concentrations in the exhaust are analyzed continuously throughout the 16-min test. Concentrations in each mode are multiplied by weighting factors to give grams per mile. This is not a true mass emissions measurement.

Table 4. Federal emission-control requirements for light-duty vehicles.²²

Model year	Pre-1968 ^a		1968	1970	1971	1972	1973	1975 ^b	1976 ^b
Test procedure	FTP	CVS-C	FTP	FTP	FTP	CVS-C	CVS-C	CVS-CH	CVS-CH
Emissions (g/mi)									
Hydrocarbons	10	17	3.4	2.2	2.2	3.4	3.4	0.41	0.41
Carbon monoxide	77	125	35.0	23.0	23.0	39.0	39.0	3.4	3.4
Nitrogen oxides	4-6	6	NR ^c	NR	NR	NR	3.0	3.0	0.4
Evaporative losses (g/test)	40		NR	NR	6.0	2.0	2.0	2.0	2.0

^aUncontrolled vehicle except for crankcase blowby control.

^bSince modified.

^cNR - No requirement.

By CVS-C in this table is meant constant volume sampling procedure - cold. This is a cold start mass emissions test. The vehicle stands for 12 h at 70°F before engine startup. The driving cycle is a 23-min, 7.5 mi non-repetitive pattern. A constant fraction of the exhaust flow is collected in a bag, and concentration measurements at the end of the test give true mass emissions in grams per mile.

By CVS-CH in this table is meant constant volume sampling procedure - cold-hot. This includes both cold and hot starts with weighted mass emissions. The vehicle stands at constant temperature for 12 h at 70°F before engine startup. The driving cycle is the 23-min pattern used in CVS-C. After a 10-min shutdown, the engine is restarted and the first 505 sec of the driving cycle repeated. A constant fraction of the exhaust flow is collected: the first 505 sec in a "cold transient bag"; the next 864 sec in a "stabilized bag"; and the repeat 505 sec in a "hot transient bag." Emissions in cold and hot transient bags are weighted 0.43 to 0.57 respectively, and added to emissions in stabilized bag to give true mass emissions.

The standards shown in Table 4 provide the required 90% reduction in 1975 vehicle hydrocarbon and carbon dioxide emissions over emissions produced in 1970 vehicles, and the required 90% reduction in 1976 vehicles in nitrous oxides emissions over those produced in the 1971 vehicles. The latter restriction is intended to preserve the overall national limit

for nitrogen dioxide, which is $100 \mu\text{g}/\text{m}^3$ of air. In June, 1972, the EPA announced that only Los Angeles and Chicago exceeded the 100- μg limit rather than about 200 areas previously listed. This questions the validity of the original drastic reductions.* Also, the law provides a "hardship" clause for complying with the 1975 standards and as a result enforcements have been delayed. The standards early this year were:

	<u>HC</u>	<u>CO</u>	<u>NO_x</u>
1975 Federal	1.5	15.0	3.1
1975 California	0.9	9.0	2.0
1976 Federal	0.41	3.4	2.0
1977 Federal	0.41	3.4	0.4

Late April changes have the 1975 Federal standards continuing through 1978 with the 1976 limits slated for 1979, and the 1977 list becoming effective in 1980 or later.† The limits include the car's 5-yr or 50,000-mi performance record.

The industry's response to these ever tightening restrictions has been one of painful acceptance, using a variety of fixes while researching better solutions. To date, there hasn't been a breakthrough the public and buyer would like. Most research seems to indicate hard work and refinement will be the answer, not a pot of gold at the end of the rainbow.

A change in fuel composition will have a profound effect on the nature of the emitted pollutants and to a lesser extent, the emitted quantities. This in turn will have a direct bearing on the difficulty in meeting and interpreting the Clean Air Act standards. The hydrocarbon emissions are related to the specific fuel compounds²³ and vary greatly in their ability to produce smog²⁴ and to cause adverse reactions in plants and animals. A change to methanol should greatly reduce the variety of

*The 1968-1971 overall reductions were 85%. The 90% additional decrease is a total improvement of 98% over the precontrol era.

†Talk given by John Brogan, director, Alternate Automotive Power System Division, EPA, April 24, 1974 in Livermore, California.

emission compounds. Some reports indicate a 70-90% reduction in HC. Nitric oxides also tend to be reduced.²⁵ Reported reduction in aldehydes have varied with the researcher^{19,25} but have been equal to or greater than the gasoline byproduct.

California initiated its emission control program in 1966. The Federal and California state laws are slowly merging into an identical set of requirements. Most automobile and other light vehicles in the U.S. now have crankcase-ventilation controls (initiated in California in 1963), evaporative controls, HC and CO controls, and NO_x controls to comply with federal and state standards.

The crankcase-ventilation control vents the crankcase fumes into the carburetor. The evaporative control has a fuel tank which is vented through activated charcoal and in addition there is a closure on the carburetor when the engine is not running. For the hydrocarbon and carbon monoxide controls, timing is retarded, particularly at low rpm and cold engine conditions. Carburetors are set on the lean side with close tolerance range. Exhaust gas is kept at high temperature with additional air for further oxidation added to exhaust gases by means of an engine driven pump. These fixes are inadequate for the new 1975 standards and industry is resorting to catalytic mufflers for further reductions. For the NO_x control, the exhaust gas is recirculated. This amounts to dilution of the combustible mix with the inert exhaust gas lowering the peak temperatures. This reduces the thermal and volumetric (breathing) efficiency of the engine. Further reductions to meet the 0.4 g/mi NO_x emission level required in 1980 depend on new design breakthroughs, such as Honda's stratified charge engine.

Combustion and Emission Relationships

The relationships with which we are primarily concerned are those between combustion and carbon monoxide, hydrocarbons, and the oxides of nitrogen. Some researchers^{26,27} support the following explanation of

combustion and emission product relationships while others disagree.²⁸ However, until the existing literature can be extensively reviewed and further work continued under our control, the following approach will be assumed.

CO and Combustion

Since CO is a product of incomplete combustion, this product is expected to increase in an engine which lacks sufficient air, that is, has a low A/F ratio. There also can be localized areas in the combustion chamber which cool below that temperature needed to continue the oxidation reaction. Engines can be designed by some simple rules for low CO and HC emissions: (1) High volume-to-surface ratio in combustion chamber. (The Wankel is a low ratio example.) (2) No cold surface areas to quench the combustion process (conversely no hot spots to develop premature self-ignition.) (3) No small, crack-like volumes that cannot support combustion. (4) Sufficient turbulence to create good fuel mixing and promote flame propagation. (Conversely, not too high a turbulence to cause excessive scrubbing action on the walls and high heat loss.) (5) If the fuel enters the chamber all or partly in liquid phase, complete vaporization, mixing, and then burning must be promoted. (A difficult cold start problem unless compression can supply the heat.) (6) Proper intake, exhaust, and ignition timing are required. Note that, of the three, only the ignition timing can be changed to accommodate various load and speed variations in present engine designs.

Different fuels have been found to influence CO production in some research. Pefley et al.¹⁹ found a factor of 10 reduction over gasoline on a research engine operating on methanol near stoichiometric conditions. This indicates methanol's ability to continue burning under more severe combustion boundary conditions such as near the cool walls of the chamber.

HC and Combustion

It is strongly apparent in some research^{29,30} that the species and quantity of hydrocarbon products vary with the fuel burned. Another

important finding noted in these papers^{24,29,30} was the wide differences in the hydrocarbon product species in their ability to produce photo-chemical smog and other environmental contaminations. Jackson's paper³⁰ and the accompanying discussions gave some indication of the large amount of work that must be done to be aware of all the facets of this problem. The species differences and their effects are not yet recognized in the control laws but must be taken in account eventually as a greater variation of fuels and engine designs are proposed. The engine modifications suggested to lower CO emissions will also result in lower HC emissions.

NO_x and Combustion

The time-temperature history of the combustion along with the available oxygen determine the NO_x produced. Higher temperatures encourage nitrogen in the air to react with oxygen in a chemical process parallel with fuel combustion. The chemical kinetics of this process were studied theroretically and experimentally³¹ and show that large differences in temperature exist during combustion which markedly influence the NO production. Time of an engine cycle is faster than necessary to approach NO chemical equilibrium, so formation becomes time dependent as well. Decomposition of NO formed early with the flame front at high temperatures does not have time to take place as the gases cool. Fuel rich ratios also limit the availability of oxygen. Air rich ratios increase NO production until such quantities are added to have a cooling effect on the peak temperatures.

Methanol fuel used in the liquid state should show a decrease in NO products because the high heat of vaporization can contribute to lower combustion temperatures. As stated previously, the energy of dissociation might also be used to smooth out the peak temperature, if the kinetics could be controlled.

LAWRENCE LIVERMORE LABORATORY RESEARCH

We came to the following conclusions by examining past work on methanol as a transportation fuel:

- Methanol is second only to synthetic gasoline as the most desirable and likely transportation fuel to replace natural petroleum products.
- The problem of blending gasoline and alcohol and performing sufficient testing to prove it is a viable solution has not been addressed by industry. The country-wide distribution of such blends is a part of this problem.
- The use of pure methanol has been studied by several groups.

The major problem in its use is cold weather starting.

Both blending and cold starting problems are being investigated in our laboratory as a small continuing effort. We are also investigating fogged fuels and electronic control. These investigations are a part of the LLL "Coal to Cars" program.

Blend Program

We have been working with new 1973 and 1974 test vehicles that have been modified with instrumentation and auxiliary fuel tanks. The test vehicles include a 1973 International pickup truck and a 1974 Plymouth Valiant. A Porsche 914 air-cooled, fuel-injected engine is also being used. Blends of up to 20% have been tried. The higher blends cause rough operation (missing). The ten percent blend doesn't seem to have any negative effects, including any recordable change in exhaust temperatures. After further testing, the 10% blend will be added to selected cars in the LLL fleet. The fleet consists of about 500 vehicles, which travel 2-1/2 million mi and consume about 300,000 gal of gasoline each year, under a variety of circumstances. Hopefully, half of the fleet will operate with the blend for a 2-yr period. Careful records are kept on the fleet and results of this program will be most valuable in proving blend practicality.

Electronic Control

In addition to testing methanol and methanol-gasoline blends, we have a program to develop a closed-loop electronic system for both fuel and emission control. The Porsche engine and the 1974 Valiant are being used in these tests. With closed-loop control, exhaust gases can be analyzed for combustion performance and thus fuel can be closely controlled for lowest emissions. We are trying to adapt the Bosch open-loop system, shown in Fig. 5, as well as develop our own designs. The dotted line in Fig. 5 is the addition to the Bosch circuit that will make it closed-loop.

After this project got underway, we learned that Bendix is working on a similar closed-loop system for gasoline. They have developed a sensor that measures oxygen content in the exhaust gases and produces a sharp change in voltage at stoichiometric. (See Fig. 6.)³² Their operation is probably identical to the solid-electrolyte, zirconium dioxide device that was proposed by R.W. Taylor of LLL in a technical note (IMTN No. 12) of February 10, 1970. The device, which is oxygen sensitive, allows passage of ionized oxygen from a surface in contact with a gas containing oxygen to a surface in contact with a gas containing less oxygen (a partial pressure difference), provided the material is at 900-1500°F (~500-800°C). The resulting change in the generated voltage provides the sensing of free oxygen in the exhaust gases. Since this gives an indication of the original A/F ratio, the system has great promise in emissions control.^{33,34} Progress in this area will be discussed in a subsequent report.

Fig. 5. The open-loop control system. The dotted line shows the closed-loop system.

Fig. 6. Characteristics of the oxygen sensor.³⁰

Fogged Fuel

The underlying problem with methanol operation in autos is the high heat needed for vaporization in a carbureted engine. This heat is not available on starting and initial warmup (without extensive modification such as electric heaters, etc.). Under normal circumstances and with the same percent vapor-to-liquid ratio, a methanol fuel requires 10 times the heat of gasoline. An alternative is to keep the fuel in liquid form but break it into small-size particles. This causes it to transport as a gas but does not require the high heat of vaporization. The phase change to vapor then occurs in the combustion chamber.

Many carburetor prototypes have attempted this sort of thing in the past. Turbulence has been used with higher velocity air streams. The 1937 Graham Supercharger improved the mix greatly and provided a higher engine input pressure. Recently, two or three systems employing electrically-driven sonic plates have been designed and are now in the experimental stages.

We call the fuel system being developed now in bench tests here at the laboratory a "fogged fuel" system. Details of the device and experimental results will be included in a later report. The design, however, is independent of the differences in vaporization heats of fuels, so could work equally well with gasolines or alcohols. First runs of a prototype on a small engine generator have increased the fuel efficiency about 50% at idle, 16% at half load, and 21% at full load. Admittedly, these engines have very simple carburetors but, nevertheless, the condition of the fuel in the fogged fuel prototype must be a more acceptable quality than the standard carburetor. Work is now proceeding to adapt the unit to a vehicle engine for further tests.

CONCLUDING REMARKS

An interim fuel is needed to supply our transportation needs until continuing sources of energy can be found and utilized. Coal is the largest remaining fossil fuel resource. With present technology, conversion

of this coal to clean fuel appears to be best accomplished through synthesized methane and methanol.

Availability of a transportation fuel in quantity from coal will not be instantaneous. Conversely, all petroleum will not disappear overnight. The sum of rising coal production and declining petroleum availability along with their attendant costs will determine the course of the transportation industry.

Society is in bondage to its huge investment in refineries, distribution, servicing, and highway networks which cannot be easily abandoned. The vehicles themselves, because of their short lifetime, are probably the most flexible element of the whole. Even so, these will not undergo sudden mutation but will, of necessity, be subject to a form of selective breeding. Where this selection will lead is unknown at this time. The Environmental Protection Agency predicts that the present conventional engine types will survive through the mid-eighties, with some modifications, followed by the gas turbine or Stirling engine along with a constantly increasing usage of electrics. Regardless of the path reality takes us from this prediction, transportation fuels will be needed well beyond the year 2000.

For the immediate future, the use of methanol would have the least shock on the automotive and related industries. It is a liquid about as hazardous as gasoline and similar in other respects; it can be blended with gasoline and has the potential of less emissions than gasoline. With slight engine modifications, it will perform well in gasoline-powered vehicles.

Transportation in this country has relied on petroleum products, mainly gasoline, and to substitute a new fuel will be difficult. Methanol fuel cannot be a step-function substitute. As methanol fuel synthesizing plants are completed, the production can gradually reduce our petroleum imports and reduce the need to draw on our dwindling petroleum reserve.

near the plants and coal fields, so the fuel type could vary with the locale as well as with time. Thus, it may be more economically feasible to prepare for a country-wide variation in fuel variety than to supply a specific blend of fuel nationally which must change as the supplies change.* Additionally, other sources, such as the forest and agricultural industries, that can produce methanol or ethanol alcohol may be important if the price of fuel rises to make manufacture profitable.

The task of the transportation industry is to prepare to use a variety of these fuels and to use them efficiently while simultaneously reducing pollution. Drastic changes in the present highway system and vehicle design are not possible on the time frame we have to prepare for non-petroleum fuels. Fortunately, the present internal combustion engine, modified in some details and equipped with sophisticated but hopefully reasonably priced electronic controls, can be adapted to the fuels of the near future.

* For some possible phasing programs, see discussion in UCID 16442.³⁵

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