Exhaust Gas Recirculation for Reducing the Oxides of Nitrogen in Spark Ignition Engines

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ABSTRACT. Spark-ignition engine exhaust gases contain oxides of nitrogen (NO_x), carbon monoxide (CO), and organic compounds that are unburned or partially burned hydrocarbons (HC). These exhaust emissions are presently subject to regulatory control. Of these harmful pollutants, NO_x emissions are the hardest to control under the current status of emission control technology.

The present paper deals with the use of the exhaust gas recirculation (EGR) for NO_x control. The NO_x concentration in the exhaust was studied experimentally, and theoretically using the method of element potential over a wide range of fuel-air ratios, percentage of exhaust gas recycled, and engine speeds. Comparison of the measured and calculated exhaust nitric oxide concentration shows good agreement over all operating conditions. To make the EGR method effective over a wide range of operating conditions, two EGR maps were constructed. They can be used as input data to a closed loop control system, to regulate the EGR rate required to reduce the NO_x emission to the reduction target.

Emissions generated by automotive engines are a major environmental problem, especially in urban areas. Because of environmental concerns, the Environmental Protection Agencies in the industrial countries have imposed very stringent emission standards to be met by the manufacturers of automotive engines. These standards represent one of the greatest challenges to face the industry. With continuing environmental pressures and renewed concerns over the potential health hazards of

Exhaust Gas Recirculation for Reducing the ...

automotive emission, these standards are unlikely to be relaxed. Therefore, further detailed study of the automotive emission problem is required.

Three exhaust emissions, hydrocarbon (HC), Carbon monoxide (CO), and Oxides of nitrogen (NO_x), from the automotive spark-ignition engines are presently subject to regulatory control. The major task of automotive emission control is basically to seek the best compromise between exhaust emission control and the two principle requirements of automotive engine, higher thermal efficiency (fuel economy) and power output.

 NO_x emission from spark ignition engines is one of the most difficult tasks. Nitric oxide NO is formed within the combustion chamber at peak combustion temperature and persists during expansion and exhaust in non-equilibrium amounts. Upon exposure to additional oxygen in the atmosphere, nitrogen dioxide NO_2 and other oxides of nitrogen may be formed. NO_2 is objectionable for several reasons: it is coloured gas which in turn contributes to decreased visibility. It also can absorb the solar energy and combine with unburned or partially burned hydrocarbon to form photo chemical smng which can reduce the ozone layer, causes damage to corps and releases a powerful eye irritant. Also studies Rai (1987), indicate that humans, when exposed to nitrogen dioxide concentration approximating 5 ppm for 10 minutes, are affected by an increased air way resistance. At extremely high doses, around 90 ppm for 30 minutes, nitrogen dioxide has been shown to produce pulmonary edema and other respiratory impairments. Accidental exposures to even higher concentrations for about 5 minutes have produced death within 2 days to 5 weeks.

Although NO is not irritant, it binds Hemoglobin in the blood. The binding strength is about 100 times stronger than that of CO and about 3 times stronger than that of No₂. By the oxidation of NO-Hb in the blood, methemoglobin forms, and its increase leads to lack of oxygen in the blood and to damage of the central nervous system.

Two approaches are currently used to reduce NO_x emissions. The first approach is to reduce the amount of NO_x produced by the combustion process itself. This may be done by the exhaust gas recirculation (EGR), retard spark timing, rich or lean air fuel-ratio, *etc.* One of these approaches, EGR is the most effective measure considering fuel economy and driveability, as revealed in past studies Kuroda and Nakajima 1978. In this system a fraction of the exhaust gases is recycled through a control valve from the exhaust to the engine intake system. The recycled exhaust gas is usually mixed with the fresh fuel-air mixture just below the carburetor throttle valve. EGR acts as an additional diluent in the unburned gas mixture, thereby

614

Abu-Qudais, M. et al.

reducing the peak burned gas temperature and hence NO formation rates.

The second approach to NO_x emissions control consists of the use of after-treatment devices Heywood 1983. These devices include flow through oxidation catalysts. If an engine is operated at all times with an air-fuel ratio at or close to stoichiometric, then both NO_x reduction, and CO and HC oxidation can be done in a single catalyst bed. The catalyst effectively brings the exhaust gas combustion to a near-equilibrium state at these exhaust condition. Such a catalyst is called a three-way catalyst.

Previous work

Alperstien and Bradow 1966, demonstrated experimentally many characteristics of the NO formation process. Using a rapid-acting sampling valve placed at different distances from the spark plug in the cylinder head of the a CFR engine, they showed that NO concentrations increase with crank angle at a given location after the flame front reaches the valve peak at crank angle approximately corresponding to peak pressure and then decay before freezing occurs. The kinetic nature of the NO formation process, the importance of the gradient in the burned gas properties, and the difference between a quiescent and swirl type of combustion chamber were all studied.

Newhall and Starkman 1967, examined the kinetics of NO formation, concluded that the Zeldovich mechanism, with the assumption that nitrogen and oxygen atoms were in equilibrium, was an adequate reaction scheme for engine conditions. They suggested that little NO decomposition would occur during expansion stroke. The measured NO concentration did not correlate well with equilibrium values corresponding to the average burned gas temperature at peak cylinder pressure.

Heywood *et al.* 1973, showed how exhaust NO concentrations could be calculated but were unable to obtained good agreement with measurements for rich mixtures. Muzio *et al.* 1971, showed how changes in engine speed, spark timing, and fuel-air ratio affected model predictions. The trends in NO concentration versus time in the burned gases matched qualitatively the sampling valve measurements of Newhall and Strakman 1967.

BMW company Walter *et al.* 1994, developed a new external EGR-system with sophisticated qualities in emission results under transient condition, in long term durability and engine response. A special kind of pressure controlled EGR-rates and an air diluted supply system guarantee a high stability of the combustion process and consequently engine emissions.

Exhaust Gas Recirculation for Reducing the ...

Gardiner *et al.* (1991) used the EGR system to improve in-cylinder mixture formation during cold starting. Tests using a special camshaft to obtain prompt EGR showed that successful starts using neat methanol could be achieved if ignition occurred during cranking. Thus, unlike the standard engine configuration, the ignition and starting limits with the prompt EGR system were essentially the same.

Blumberg and Kummar 1971, studied the effect of air-fuel ratio, spark timing, compression ratio, EGR percentage, residual gas temperature, inlet conditions, and engine geometry on NO emissions.

In this paper a theoretical model for predicting the NO_x concentration in the exhaust of a spark ignition engine is presented over a wide range of fuel-air ratios, engine speeds, and percentage of exhaust gas recycled. It is assumed that the unburned mixture are frozen in composition and the burned gas mixture are in equilibrium. The burned mixture composition calculations were done by adjusting the mole of each species until the minimum Gibbs function state is found. This method is called, method of element potential. Experimental work was done to look into NO_x emissions from the engine and to compare them with those predicted from the theoretical model.

Theoretical Model

It is good approximation for performance estimates in engine to treat the unburned mixture of fuel, air, and residual gases from the previous cycle as a frozen in composition, and the burned gases produced by combustion as in chemical equilibrium. The most complete models are based on polynomial curves fits to the thermodynamic data for each species in the mixture. The thermodynamic properties of each of these species have been fitted to data from the JANAF tables Gardiner *et al.* 1991. The specific heat, enthalpy, and entropy of species i are given by:

$$C_p = R(a_{i1} + a_{i2}T + a_{i3}T^2 + a_{i4}T^3 + a_{i5}T^4)$$
(1)

$$\bar{h}_{i} = RT \left(a_{il} + \frac{a_{i2}}{3} T + \frac{a_{i3}}{3} T^{2} + \frac{a_{i4}}{4} T^{3} + \frac{a_{i5}}{5} T^{4} \right)$$
(2)

$$\bar{s}_i = R \left(a_{il} \ln T + \frac{a_{i2}}{3} T + \frac{a_{i3}}{3} T^2 + \frac{a_{i4}}{4} T^3 + \frac{a_{i5}}{5} T^4 \right)$$
(3)

$$\bar{h} = \int \bar{C}_p dT$$
 and $\bar{s} = \int C_p \frac{dT}{T} - R \ln \frac{P}{P_0}$

Values of the a_{ij} for several species of interest, such as Iso-C₈H₁₈, H₂O, CO, CO₂, H₂, O₂, O, N₂, OH, NO, and H were taken from JANAF 1971. Two temperature ranges are given, the 300-1000 °K for unburned mixture property calculations, and 1000-5000 °K for burned mixture property calculations. The reference temperature is chosen to be 298 °K with the enthalpy of H_e, O₂, N₂, and C set to zero.

The properties of the unburned mixture are the sum of the properties of all n components

$$h_u = \sum_{i}^{n} Y_i \bar{h}_i$$
, $S_u = -R \ln \frac{P}{P_o} + \sum_{i}^{n} Y(\bar{S}_i - R \ln Y_i)$ (4)

The temperature of the unburned mixture at any point in the engine cylinder during the isentropic compression process was determined from fuel-air cycle calculations.

The burned mixture composition calculations is a chemical equilibrium problem. It can be formulated in several ways. If the concept of equilibrium constant is used, then it is necessary to identify the set of reactions that take place and to determine the associated equilibrium constants. A set of nonlinear algebraic equations must be solved to get the mole numbers of each species. If the system is large, it becomes a difficult task. Other methods adjust the mole of each species, consistent with atomic constraints, until the minimum Gibbs function state is found. Again, there are many variables involved, and great care must be taken to be sure that all mols are non-negative. The presence of rare species in the system complicates the problem.

In search of a better way to solve combustion equilibrium problems, Reynolds 1988, suggested the method of element potentials. This method uses theory to relate the mole fractions of each species to quantities called element potentials. There is one element potential for each independent atom in the system, and these element potential, plus the total number of mols in each phase, are the only variables that must be adjusted for the solution. In large problems this is much smaller than the number of species, and hence far fewer variables need to be adjusted. The present

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analysis, and the program, assumes that the gas phase is a mixture of ideal gases and that condensed phases are ideal solutions. These are good approximations for many practical problems of interest.

The Basic Theory of Element Potentials

The Gibbs function of a system is:

$$G = \sum_{j=1}^{s} \bar{g}_j N_j \tag{5}$$

where g_j is the partial molal Gibbs function and Nj is the number of mols of species j, and s is the total number of species in the system. Treating each phase as either a mixture of ideal gases or as an ideal solution, the partial molal Gibbs functions are given by:

$$\overline{g_i} = g_i (T, P) + RT \ln x_i \tag{6}$$

where g_j (T, P) is the Gibbs function of pure j evaluated at the system temeprature and pressure, x_j is the mole fraction of j in its phase, and R is the universal gas constant.

The atomic population constraints are:

$$\sum_{j=1}^{s} n_{ij} N_j = P_i \qquad i = 1, ..., a$$
(7)

Where n_{ij} is the number of i atoms in a j molecule, p_i is the population (mols) of i atoms in the system, and a is the number of different elements (atom types) present in the system. The equilibrium solution at the given T and P is the distribution of N_j that minimizes G, subject to the atomic constraints (7), for non-negative N_i .

For convenience, let $g_j = g_j$ (T, P)/RT, and seek the minimum of G/RT. Using equation (6) we find that, for arbitrary variations in the mole numbers

$$d \left[\frac{G}{RT}\right] = \sum_{j=1}^{s} \left(\bar{g}_{j} + \ln x_{j}\right) dN_{j} + \sum_{j=1}^{s} N_{j} \frac{1}{x_{j}} dx_{j}$$
(8)

Now denote the number of mols in phase m by N_m; then,

$$x_j = N_j / N_{(j)} \tag{9}$$

where $N_{(j)}$ is the number of mols in the phase containing species j. The second sum in equation (8) can therefore be replaced by

$$\sum_{m=1}^{p} \overline{N}_{m} \sum_{in \ m} \sum_{j=1}^{s} dx_{j}$$

where p is the total number of phases that might be present. This vanishes, because the mole fractions in each phase always sum to unity.

Now the dN_j are not all independent, because of the atomic constraints. Relationships among the dN_j are obtained by differentiating equation (7):

$$\sum_{j=1}^{s} N_{ij} \ dN_j = 0 \quad , \qquad i = 1, \dots, a$$
(10)

We must solve for the a restricted dN_j in terms of the s-a free dN_j , and then substitute these relationships into (8) in order to express the G variation in terms of freely variable N_j . This process is equivalent to subtracting multiples of equation (10) from equation (8):

$$d\left[\frac{G}{RT}\right] = \sum_{j=1}^{s} \left(\bar{g}_j + \ln x_j\right) dN_j - \sum_{j=1}^{\alpha} \lambda_i \sum_{j=1}^{s} n_{ij} dN_j$$
(11)

The λ_i are the multipliers required to drop out of the set of restricted dN_j from this equation. Thus, setting the coefficients of these dN_j to zero, then:

$$\bar{g}_j + \ln x_j - \sum_{i=1}^{\alpha} \lambda_i \ N_{ij} = 0$$
(12)

for the restricted j. With these dN_j absent from (11), the remaining dN_j may be freely varied, and at the minimum G point there must be no variations that change G to first order. This will be true only if the coefficient of each free dN_j is zero; hence, (12) also applies to the free j. So, for every species:

Exhaust Gas Recirculation for Reducing the ...

$$x_j = e^{-\tilde{g_j} + \sum_{j=1}^{\alpha} \lambda_j \ n_{ij}}$$
(13)

Equation (13) is the main result of the theory of element potentials for mixtures of ideal gases or for ideal solutions. It relates the phase mol fraction of each species to its value of g(T,P)/RT, to the atomic makeup of its molecule, and to a set of undetermined multipliers to be determined from the atomic constraints. The multiplier is called the element potential for i, atoms. Using equation (6), we see that

$$\frac{g_j}{RT} = \sum_{j=1}^{\alpha} \lambda_i \ n_{ij} \tag{14}$$

and hence λ_i represents the Gibbs function/RT per mole of i atoms. The λ_i are properties of the system, and can not be tabulated as functions of the atom or molecule, as can the g_i .

The values of the element potentials, are determined by the atomic constraints (7), rewritten as follows:

$$\sum_{j=1}^{s} n_{ij} \ \bar{N}_{j} \ x_{j} = p_{i} \ , \quad i = 1, \dots, a$$
(15)

using (13), this becomes a set of a equations for the a unknown - and the p unknown N_m . To this we add the p equations:

$$\sum_{j=1}^{s} x_{j} = 1 , \qquad m = 1, \dots, p$$
(16)

 $X = \{y \mid x\}$

Equations (15) and (16) must be solved simultaneously to determine the element potentials and phase mols. This might appear to be a difficult task, but it is possible to do it accurately and quickly.

In the computer program, the user selects the species to be included in each phase of the system, sets the atomic populations and state parameters. Then the equilibrium composition can be calculated using the method of element potentials, or a frozen composition can be specified. A series of calculation can be made over a

620

matrix of T and P values. For example when one mole of Iso-Octane C_8 H₁₈ is burned with air at equivalence ratio of 0.9 according to the following reaction:

$$C_8H_{18} + 13.9O_2 + 52.2N_2 \rightarrow 8CO_2 + 9H_2O + 1.39O_2 + 52.2N_2$$

The number of mols of the reluctant input into the program are the same as in the chemical reaction equation. The calculation of the properties (u,h,s,v) of this freezing mixture at the beginning of the compression is done by estimating the inlet temperature and pressure. To move to the end of the compression process in the airfuel cycle (reversible and adiabatic compression), the compression ratio and the estimated temperature must be specified. As a result the properties at this point are stored in a file. Then constant volume combustion under estimated maximum temperature will take place. By isentropic expansion of the burned gases that remain in chemical equilibrium, the properties at the end of the expansion stroke are stored in a file. Finally a reversible, adiabatic exhaust blow down process with the burned gases fixed in chemical composition, which represents the expansion of the exhaust gases to the atmospheric pressure.

The calculations were done for zero EGR percentage, and for different burned gas fractions (residual and recycled exhaust gases), in the unburned mixture.

Experimental Apparatus and Test Procedure

Figure (1) shows a schematic diagram of the test setup used in this study. The engine was four cylinder, 4-stroke, water cooled, FORD spark ignition engine. The basic engine specifications are given in Table 1.

Bore	8.1 cm
Stroke	5.33 cm
Speed range	1500-3000 rpm
Maximum power	22 Kw
Spark timing	Optimized for maximum torque

Table 1. Basic Engine Specifications



Fig. 1. Schematic diagram of the experiment set-up.

The exhaust gas recirculation (EGR) line connected the exhaust manifold with the intake through a valve and calibrated orifice. The pressure difference between the exhaust and the intake was sufficient to obtain an adequate flow. The flow rate was determined using the calibrated orifice and controlled manually through the valve.

An exhaust gas analysis system, P7450 Cusson, was used to measure the NO, NO_2 , CO, CO_2 , HC, and O_2 concentrations in the raw exhaust gases by taking a sample from down-stream of the exhaust manifold. The analyzer automatically and continuously determines the concentration of a particular component of interest in a complex gas mixture.

 NO_x concentration measurements were made in the following manner. The engine was allowed to warm up and stabilize for at 20 minutes. Measurements were first made without EGR over the equivalence ratio range 0.7-1.35 at different engine speeds. Experiments were then carried out with increasing EGR percentages at

several equivalence ratios. The spark timing was set for maximum brake torque. A sample was drawn through the analyzer sampling probe to the analyzer. The analyzer was calibrated using standard span gases.

Results and Discussion

Figure (2) shows the calculated and measured exhaust NO_x concentration as function of equivalence ratio at different EAR percentages. It can be shown from this figure that, the maximum point of NO_x concentration lies around the equivalence ratio of 0.9 where the flame temeprature is high and the O_2 molecules are available.



Fig. 2. Measured and calculated exhaust NOx concentration as function of equivalent ratio.

Also it shows that NO_x formation is reduced by increasing EGR rate due to lower combustion gas temperature and lower O_2 concentration, as a result of inert gases introduced into the combustion chamber. There is a good agreement between the

calculated and the measured values of NO_x concentration. The deviation in the calculated concentration from the measured one around the stoichiometric equivalence ratio is due to the heat loss from the combustion chamber walls which reduces the combustion gas temperature, hence the NO_x formation will be less, while for the theoretical one, it is an adiabatic combustion process which in turn results in higher combustion gas temperature. In the rich side, the deviation could be due to the freezing of NO_x chemistry during the expansion stroke after a substantial NO_x decomposition has occurred as found by Heywood (1973), which depends on engine operating conditions as well as whether the mixture element burned early or late during the combustion process. As a result, the final exhaust NO_x concentration for rich mixtures, depends upon the burned gas temperatures later in the expansion stroke after the charge is fully burned, while for the lean mixtures, it depends primarily upon the peak gas temperatures. The theoretical model assumed that the burned gases during the expansion stroke remain in chemical equilibrium until they are fixed in chemical composition at freezing point. This makes the prediction of exhaust NO_x concentrations for rich mixtures more difficult.



Fig. 3. Measured percent reduction of exhaust NOx emission and I.M.E.P as function of EGR percent.



Fig. 4. Calculated percent reduction of exhaust NOx emission and I.M.E.P as function of EGR percent.

Figures (3) and (4) show the measured and calculated percentage reduction in NO_x concentration and indicated mean effective pressure (I.M.E.P), which is proportional to the power output from the engine, as function of EGR percent. The reason of reduction in i.m.e.p. is that EGR increases the combustion duration by increasing the time for the flame to propagate across the charge inside the combustion chamber. Also as the EGR percentage increases, the combustion temperature decreases which inturns decreases the pressure inside the combustion chamber.

Figure (5) shows the effect of EGR on the measured hydrocarbon (HC) emission. It can be seen that the EGR increases HC emission due to increase in the amount of the charge burned during the expansion stroke, hence increases the possibility that part of this charge will be exhausted as unburned HC.



Fig. 5. Percent increase in the HC as function of EGR percentage at different equivalent ratios.

To determine the optimum EGR rate, two EGR maps were constructed, under the following conditions as an example,

NO_x reduction target HC increase I.M.E.P loss 1500 ppm. 20% at maximum. 10% or less.



These two maps, Figure (6) and (7), can be used as input data to a closed loop control system, to regulate the EGR rate required to reduce the NO_x emission to the

reduction target at any operating condition of the engine. The first map uses the engine speed and the equivalence ratio at full throttle condition to get the required EGR at which NO_x level can't exceed its constant value. The second map gets the EGR rate as function of engine speed, and at different throttle valve opening. To look at the NO_x emission during the acceleration and decleration conditions, the NO_x emission was measured during these conditions by recording the output from the analyzer in a strip chart recorder. It was found that these conditions fall within the second map. The NO_x emission also was measured during warming up period, where it was much less than 1500 ppm, which means that no EGR is required to control the NO_x emission during the warming up period.

Conclusion

– The use of EGR is effective in controlling of NO_x emission with less penalty on power output up to a certain limit of EGR.

- The effect of EGR on the HC emission and on the output power can be minimized with lean mixtures. Also by improving the combustion chamber design to get fast burning, which can be achieved by increasing the turbulence, as an example, to improve the mixing of the charge.

– The point in the cycle at which the NO_x chemistry freezes is different for lean and rich mixtures.

- The model described here for predicting exhaust NO_x emission from spark ignition engine adequately matches experimental measurements over a wide range of engine air-fuel ratios.

- According to the good agreement between the model and the experimental results, the model can be easily used to construct emission control maps.

Nomenclature

BSFC: Brake specific fuel consumption.

G: Gibbs free energy.

 $g_{\overline{i}}$: Partial molal Gibbs function of j molecule.

h-: Enthalpy of i speciy.

IMEP: Indicated mean of effective pressure.

S_i: Entropy of i speciy.

No_x: Mixture of NO and No₂.

y_i: Mole Fraction.

 λ_i : Multiplier called element potential for i atom.

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(Received 01/12/1994; in revised form 24/05/1997) تدوير الغاز العادم لتقليل انبعاث أكاسيد النيتروجين من محركات الاحتراق الداخلي ذي الاشتعال بالشرارة

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ان الطريقة الفعالة لتقليل انبعاث أكاسيد النيتروجين من محركات المركبات هي اعادة تدوير الغاز العادم لتخفيف شحنة الهواء والوقود النقية الداخلة إلى الحرك والتي غالبا ما تكون مصحوبة بحلقة تحكم مغلقة

لقد تم تطبيق نموذج نظري للتنبؤ بتركيز أكاسيد النيتروجين في الغاز العادم المنبعثة من الحرك ذي الاشتعال بالشرارة على نطاق واسع من نسب خليط الهواء والوقود ونسبة الغاز العادم المدور .

وقد تم عمل التجارب العملية على محرك ذي أربعة أسطوانات من نوع (Ford) على نطاق واسع من نسب خليط الهواء والوقود ونسبة تدوير الغاز العادم وكذلك سرعة الحرك . بالمقارنة بين القيم المقاسة والمحسوبة لتركيز أكسيد النيتروجين تبين ان هناك توافقا جيدا تحت جميع الظروف العملية .

ولجعل طريقة تدوير الغاز العادم فعالة ، وجد أنه من الضروري السيطرة على نسبة الغاز العادم المعاده حسب ظروف العمل للمحرك . ان نظام السيطرة الموجود هنا يحتاج إلى معرفة نسبة خليط الهواء والوقود وسرعة الحرك ثم الدخول إلى الخرائط لمعرفة نسبة الغاز المدور اللازمة ، وبمقارنة هذه القيمة Abu-Qudais, M. et al.

المقروءة من الخريطة مع القيمة المحسوبة من معرفة تركيز الأكسوجين في مدخل المحرك وبإستخدام تلك الحصلة ممكن بناء حلقة تحكم مغلقة عن طريق اشارة تغذية راجعه معتمدة على تركيز الأكسوجين في مدخل المحرك .