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دراسة انبعاثات المحرك العامل على وقود الإيثانول والغازات المسرطنة

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الخلاصـة

يستخدم الإيثانول بشكل واسع كوقود حيوي وهو ناتج من الكتلة الحيوية كالذرة وقصب السكر والشوندر السكري، وقد أثبتت الأبحاث أن الإيثانول يحسن أداء محرك الاحتراق الداخلي، ويعتبر وقوداً صديقاً للبيئة. ويؤدي استخدام الإيثانول إلى تخفيض (أول وثاني أكسيد الكربون وأكاسيد الآزوت والمركبات الهيدروكربونية والجسيمات الصلبة) بالمقارنة مع الديزل والبنزين، وبالطبع فإن الأمر يتعلق باستراتيجية التشغيل. لكن استخدام الإيثانول يؤدي لزيادة كبيرة في انبعاث الألدهيدات، خاصة الأستالدهيد والفورمالدهيد، وهي غازات مسرطنة. الهدف من هذا البحث هو دراسة المؤشرات التي تؤثر على الألدهيدات المنبعثة من محرك عامل على الإيثانول باستخدام برنامج المحاكاة CHEMKIN-PRO المتقدم.

Missions of ethanol fueled engine and carcinogenic gases

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ABSTRACT

Ethanol is used widely as a bio-fuel. It is produced from bio-mass (such as corn, sugar cane and sugar beet. Research has proved that ethanol improves performance of the internal combustion engine and it considered as an environmental friendly fuel. Use of Ethanol reduces the emission (CO_2, CO, NO_x, HC, PM) compared to diesel and gasoline fuel, but this depends on the strategy of operation. However, using Ethanol increases the emissions of aldehydes dramatically, especially acetaldehyde and formaldehyde, which are carcinogenic.

The purpose of this paper is to study the parameters that affect aldehydes emitted from Ethanol fueled engines by using CHEMKIN-PRO advanced simulation program.

Keywords: Aldehyde; bio-fuel; emissions; engine; ethanol

INTRODUCTION

Ethanol is used widely as bio-fuel. It is produced from bio-mass such as corn, sugar cane and sugar beet. Research has proved that ethanol improves the performance of internal combustion engine and is considered as environmental friendly fuel.

The first internal combustion engine was invented in 1826 by Samuel Morey, which was fueled by Ethanol and Turpentine. When Otto cycle was invented, the engine was designed to work on Ethanol and Gasoline as fuel. Henry Ford produced the first Ford car in 1896, which was fueled by pure Ethanol. In the 1920s, Ethanol was used as an additive to Gasoline for increasing Octane number (www.fuel-testers.com/ethanol_fuel_history.html). During World War II, Ethanol was used as fuel in many countries such as USA and Germany (www.liquidsunenergy.com/learning/ppt/russ.pdfl). In the 1970's, oil crisis has pushed the governments to re-invest in Ethanol production and use it as fuel in the engine. Brazil is one of the most important countries to develop production of Ethanol from sugar cane and flexible fuel vehicle (FFV) car production that works on the variable mix of Ethanol and gasoline (De Souza Nascimento, *et al.*, 2010). Another country is USA, who produces Ethanol from corn, to use it as E85 (85% Ethanol and 15% gasoline) as fuel in the engine, because E85 is competitive and does not need to modify the gasoline engine in addition to improving the combustion and reduce emissions, but the improvement is limited because of the low compression ratio of gasoline engine (modelengineeringwebsite.com/First_I_C_engine.html).

Ethanol utilization specifications

US-EPA researches about engines fueled with Ethanol conclude the following:

1. High octane number (RON=108) that allows working on high compression ratio (CR=19.5:1). (Brusstar, 2003)
2. Improvement of the thermal efficiency because of the compression ratio (CR). The thermal efficiency is about 40% when working on CR=19.5:1, which is close to similar diesel engine(Brusstar, 2003). As shown in Figure 1 and Figure 2.

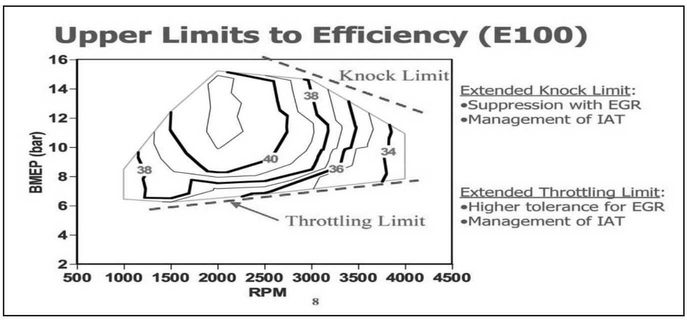

Fig. 1. Brake Thermal Efficiency of EPA engine (Brusstar, 2003)

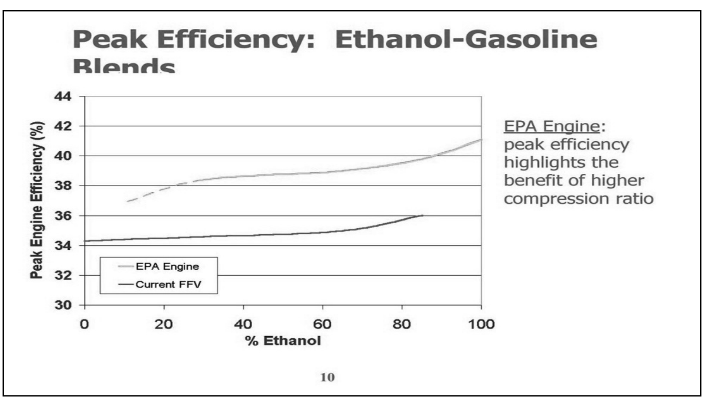

Fig. 2. Peak thermal efficiency of EPA engine and FFV engine depending on the percentage of Ethanol blended with gasoline (Brusstar, 2003)

3. Higher laminar flam velocity that help to mix Ethanol with air and reduce the need of throttling on light load. (Brusstar, 2003)
4. Higher heat of vaporization compared to gasoline makes the charge density higher and reduces the power loss. (Brusstar, 2003)
5. The emissions of CO, NO_x, HC & PM are lower than similar diesel engines

and are not worse than similar gasoline engines since the combustion is better (Ethanol contain oxygen) and the heat of combustion is lower, so the NO_x emission is lower. (Brusstar, 2003)

6. Ethanol is economic fuel comparing with diesel especially when using low-pressure port fuel injection system PFI. (Brusstar, 2003)
7. The annual cost of using a car fueled with Ethanol is similar to a gasoline fueled car. (Brusstar, 2003)

Experimental study

Description of the Experimental Setup and Testing Procedure

In this study, the experiments were performed on a Prodit computerized set engine (Figure 3), single cylinder 4 strokes, variable compression ratio, which can work on spark ignition (SI) and compression ignition (CI) gasoline engine. The engine specification is given in Table 1.

Table 1. Main characteristics of the test engine

Engine Type	Prodit 1 Cylinder
Bore ×Stroke, (mm)	90 × 85
Displacement Volume, (cc)	451
Compression Ratio	4:1÷17.5:1 variable
Max. Torque, (Nm@rpm)	38@2800
Max. Power, (kW@rpm)	12@3400
Max. Speed, (rpm)	3600
Cooling System Liquid	Water

Fig. 3. Prodit experimental set

Electrical Load was used in the experiments, which is electrical motor set on the axle of the engine.

Instruction computer by rigorous utility software (ICARUS) was used as control, analysis and calculation system, which measure: revolution number n, torque C, Air mass flow q_{va}, fuel consumption q_{vf}, compression ratio p, pressure in cylinder P_c.

This software collects all data from the sensors; analyze it and calculate the performance parameters and give it to the user through its built in software in Prodit Set (Figure 3).

The performance and emission parameters were measured by using pure Ethanol and Ethanol blended with Gasoline (E85 & E50) and compared the results with regular Gasoline. The properties of ethanol fuel are given in Table 2.

Table 2. Chemical and physical properties of used Ethanol

Density	0.79g/cm3
Soluble in water (20°C)	Soluble
Melting point	-114.5°C
PH (10g/l H2O, 20°C)	7
Flash point (1013 hPa)	78.3°C
Ignition temperature	425°C
Vapor pressure (20°C)	59 hPa
Viscosity (20°C)	1.2 mPa
Purity (G.C)	min 99.9%
Water	max. 0.1%
Filtered by fiulter	0.2µm

The gas emissions were measured by using three different emission analyzers: KANE (Gas analyzer), KANE (KM9106EM Quintox emissions), and Tecnotest 488.

The three analyzers are designed to measure CO_2, CO, NO_x, HC and PM in the exhaust, simultaneously for regular fuel (liquid and gas).

Engine performance parameters and emissions - results and comparisons

The experiment was done with speeds ranging between 1500rpm -3000rpm. Compression Ratio was 11.5:1, because the experiment set did not work on the spark ignition system above CR=12:1 and CR=11.5:1 is suitable for ethanol and it is under the knock limit for gasoline. We have obtained the results as shown in Table 3.

Table 3. Engine performance parameters with various types of fuel

	Gasoline	E100	E85	E50
Peak Power [kW]	9.68	6.17	14.40	7
Peak Torque [N.m]	36.74	27.38	45.74	28.38
Peak Specific fuel consumption [kgfuel/kW]	0.23	0.25	0.57	0.26

We used gasoline as base of comparison, as the compression ratio was used for gasoline was under 10.5:1, but we used it to search the limit of performance and for fixing the base of comparison.

We have got the best power and torque, when using E85, but the fuel consumption increased dramatically. This problem was solved by using another control strategy to decrease the fuel consumption, knowing that the fuel consumption increased when the engine worked at low speed under 2000 rpm between 2500-3000 rpm. The specific fuel consumption (the mass of consumption per 1 kW of engine power) was 0.18 kg_{fuel}/kW which is less than gasoline (0.22 kg_{fuel}/kW), E100 (0.24 kg_{fuel}/kW), E50 (0.22 kg/kW), in the same range of speed. Also the peak Torque for E85 was achieved at high speed (2800rpm) when it was achieved for gasoline, E100 and E50 at low speed (1850, 1450 and 1800 rpm in order).

Performance with E50 was bad because the power, torque and fuel consumption were less than gasoline and E85, thus making it not applicable in the world fuel markets. The performance with E100 was also bad. However, to get more power from it, we need higher compression ratio as EPA engine.

At the same time, we measured the emissions with three gas analyzers which was mentioned above. CO_2, CO, NO_x and HC were measured two times, first in summer at temperature 35°C and in winter (about 15°C) 2009-2010. The average emissions are shown in table 4.

Table 4. Average emissions

	Gasoline	E100	E85	E50
CO2 %	5.3	5.3	5.8	4.5
CO %	2.62	1.6	0	0.6
NOx [ppm]	24	29	18	30
HC [ppm]	632	396	565	199

The experiment set was not provided with catalyst, so the results are only for comparison.

- CO emission decreased when Ethanol was used because Ethanol contains (oxygen that makes combustion better (as shown in the Figure 4). The E85 was the best one in this field too. CO_2 increased when E85 is used, because CO decreased to zero. Since the higher percentage of Ethanol in E85 means higher percentage of oxygen, the combustion of E85 was better than the combustion of E50 and CO will be less.

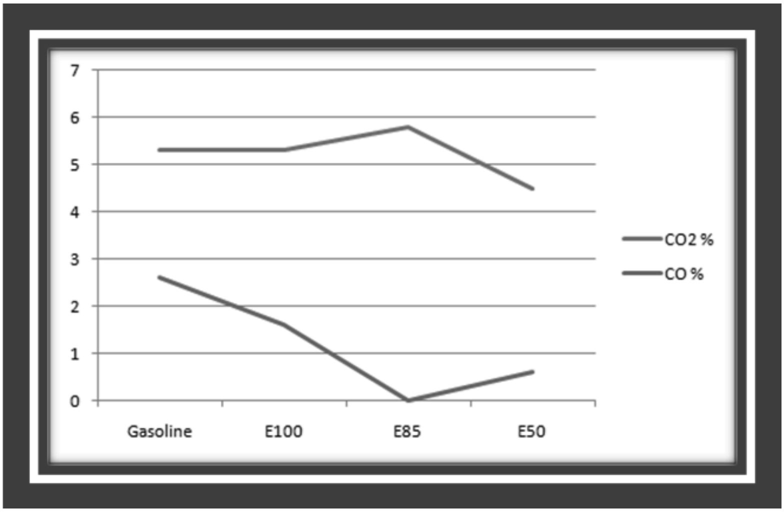

Fig. 4. Comparison of CO and CO_2 emissions from various type of fuel (gasoline, E100, E85, E50) in Damascus experiment

- NO_x emissions were low (as shown in the Figure 5) as the heat of burning of Ethanol was low and NO_x was not emitted, the E85 is the best in this field too. NO_x emissions at E50 was larger than gasoline because E50 contained oxygen that will be added to the air oxygen; the additional oxygen reacts with the N_2 to form NO_x.

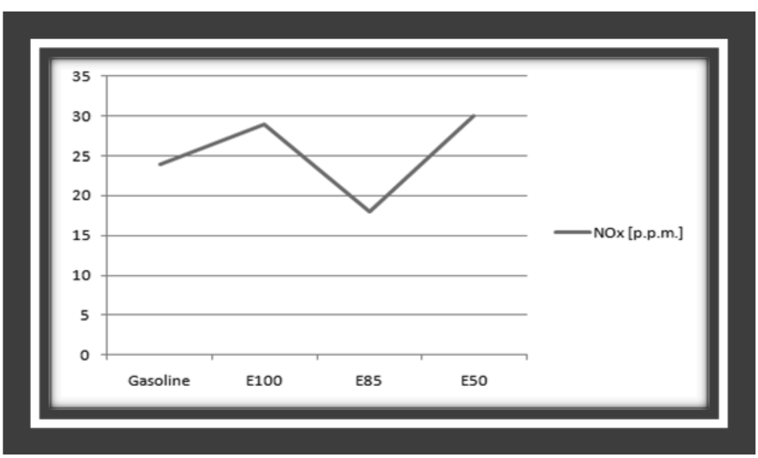

Fig. 5. Comparison of NO_x emissions from various type of fuel (gasoline, E100, E85, E50) in Damascus experiment

- HC emissions were high (as shown in the Figure .6). When a high compression ratio was used, the total hydro carbons increased with gasoline fueled engine. Using Ethanol decreased the HC: the best fuel was E50, but as shown above, using E50 had bad effect on the engine performance. Hence E100 is better.

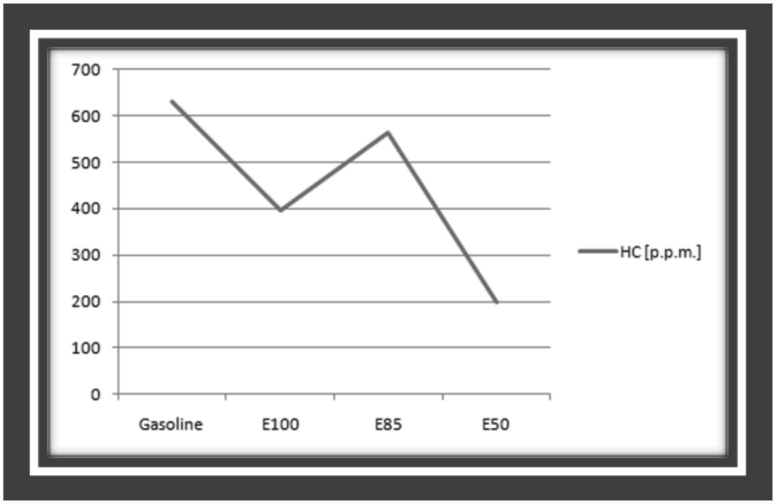

Fig. 6. Comparison of HC emissions from various type of fuel (gasoline, E100, E85, E50) in Damascus experiment

The result proved Ethanol is environmental friendly and using catalyst will make the emissions lower.

Emitted Aldehydes and its effect on human health

During the last three decades, many research have proved that using Ethanol as fuel increased emission of aldehydes, especially acetaldehyde and formaldehyde in small quantities.

The research of auto/oil air quality improvement program (AQIRP) in Dearborn – USA in 1989 confirmed that using E10 increased the emissions of acetaldehyde and formaldehyde comparing to other types of fuel. The Research concluded that the reason for increase in emissions of acetaldehyde and formaldehyde is partial oxidization of Ethanol in internal combustion engine.(Reuter *et al.*, 1992)

The research at Hennepin County, Minnesota State – USA (1993-1996) used E95 and concluded with the same results. (Frailey, 1998) At the same period EPA-USA research using E85 and E10 has also proved that using Ethanol blended with gasoline increased the emission of acetaldehyde and formaldehyde (Peter, 1995).

National renewable energy laboratory, Colorado State, USA has used 400 cars to compare the emission of cars fueled with gasoline, ethanol, methanol and CNG. The results of emitted acetaldehyde and formaldehyde of Ethanol fueled cars proved the same results, as shown in Figures 7 & 8. (Coburn, 2008)

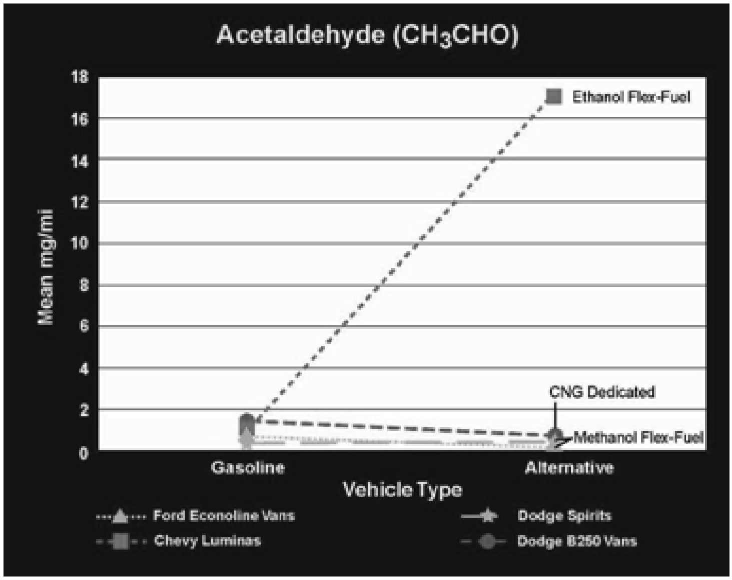

Fig.7. Acetaldehyde emissions in NREL experiment (Coburn, 2008)

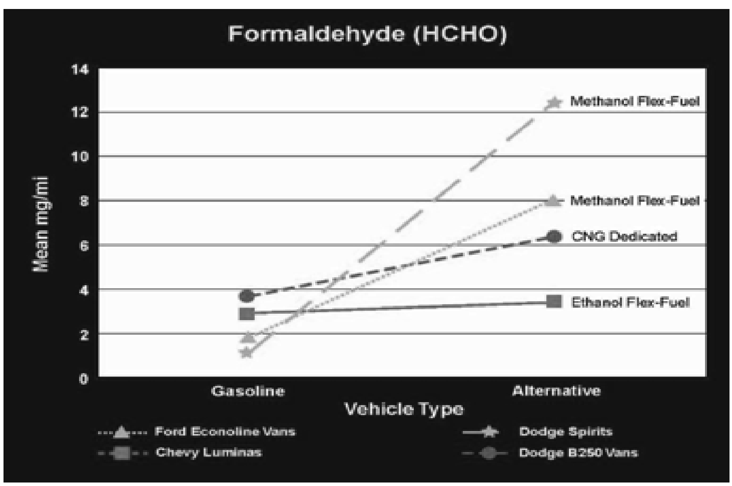

Fig. 8. Formaldehyde emissions in NREL experiment (Coburn, 2008)

Also several researches in this field have realized and concluded the same problem (NAFTC 2008; Poulopoulos, 2001; Jia, 2005; Song, 2005; Yang, 2012; Magnusson, 2010; Magnusson, 2002) that Acetaldehyde and Formaldehyde are toxic and are carcinogenic gases. (Committee on Aldehydes, 1981; World Health Organization, International Agency for Research on Cancer, 1999; United States Environmental Protection Agency, 1992; World Health Organization, International Agency for Research on Cancer, 2006). The chemical reactions of ethanol are shown in Table 5.

Table 5. Ethanol reactions (Jun, 2010)

Reaction	Equation
Sufficient steam supply	$C_2H_5OH+3H_2O \rightarrow 2CO_2+6H_2$
Insufficient steam supply	$C_2H_5OH+H_2O \rightarrow 2CO+4H_2$ $C_2H_5OH+2H_2 \rightarrow 2CH_4+H_2O$
Dehydrogenation	$C_2H_5OH \rightarrow C_2H_4O+H_2$
Acetaldehyde decomposition	$C_2H_4O \rightarrow CH_4+CO$
Acetaldehyde steam reforming	$C_2H_4O+H_2O \rightarrow 3H_2+2CO$
Dehydration	$C_2H_5OH \rightarrow C2H_4+H_2O$
Coke information	$C_2H_4 \rightarrow$Polymeric deposit (Coke)
Decomposition	$C_2H_5OH \rightarrow CO+CH_4+H_2$ $2C_2H_5OH \rightarrow C_3H_6O+CO+3H_2$ $C_2H_5OH \rightarrow 0.5CO_2+1.5CH_4$
Reaction of decomposition products	
Methanation	$CO+3H_2 \rightarrow CH_4+H_2O$ $CO_2+4H_2 \rightarrow CH_4+2H_2O$
Methane decomposition	$CH_4 \rightarrow 2H_2+C$
Boudouard reaction	$2CO \rightarrow CO_2+C$
Water gas shift reaction (WGSR)	$CO+H_2O \rightarrow H_2+CO_2$

As shown in Table 5, the emitted acetaldehyde from burning Ethanol is raised from Alcohol Dehydrogenises (ADH) (World Health Organization, International Agency for Research on Cancer, 2006)

Decreasing emitted aldehydes of I.C. engine by using CHEMKIN-PRO

Simulation experiment setup

To study the effective parameters that decrease the emitted aldehydes of internal combustion engines fueled with Ethanol, we used CHEMKIN-PRO advanced program.

Reaction design empowered transportation manufacturers and energy companies to achieve their clean technology goals with comprehensive, easy-to-use software simulation tools, chemical models and expert consulting services. Reaction design is the exclusive developer and distributor of CHEMKIN and CHEMKIN-PRO software; the de facto standards for modeling and simulating gas-phase and surface chemistry.

Today, world's leading companies are using detailed chemistry in their development processes as a means to improve product efficiency, reduce emissions, shorten design cycles and lower experimental costs. While the CHEMKIN simulation tool offers a wide variety of detailed chemical models that fit most chemistry processes, there are specific processes that may require enhanced or customized features to accurately emulate physical conditions. Reaction Design provides custom CHEMKIN model development for companies in the energy, transportation, materials and chemical processing fields. Customization projects range from enhancing the features of existing CHEMKIN models to creating entirely new models that suit unique applications or operating conditions.

This program gives the opportunity to study homogenous charge compression ignition (HCCI) engine that can be fueled with Ethanol and has a good reputation as an environmental friendly engine. Figure 9 compares between HCCI engine with the others. (Sun, 2005; Fengjun, *et al.,* 2010; U.S. Department of Energy, 2001; Motyl, 2003)

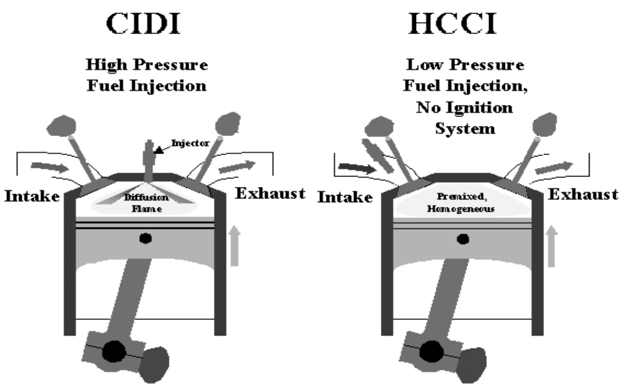

Fig. 9. Combustion systems (Fengjun, 2010)

The parameters shown in Table 6 are chosen to discover which one affects the emissions of acetaldehyde and formaldehyde in CHEMKIN-PRO simulation study.

Table 6. HCCI engine parameters in CHEMKIN-PRO simulation study

Parameters, unit	
Engine compression ratio	10:1 - 17:1
Engine cylinder displacement volume, cm^3	550
Engine connecting rod to crank radius ratio	2
Engine speed, rpm	1500
Equivalence ratio	0.8-0.9-1-1.1-1.2
Initial gas temperature, °C	20-85
Initial gas pressure, atm	1

Marinov study (Motyl, 2003; Marinov, 1998; www.pls.llnl.gov/data/docs/science_and_technology/chemistry/combustion/ethanol_mech.txt)is used as basic data in the program. We used it to study three types of fuel, Ethanol 100% (E100), and Ethanol 95% (5% water) and Blended Ethanol with Hydrogen (99% Ethanol, 1% Hydrogen).

Results and Comparison

Table 7 shows the results of using Ethanol E100 in HCCI engine. The results proved that the amount of acetaldehyde is more than formaldehyde. The changing CR does not affect the amount of acetaldehyde and formaldehyde, but when the equivalent Ratio α is increased, the amount of acetaldehyde and formaldehyde is increased. The initial gas temperature is the most effective parameter when the temperature of the injected ethanol is low, the acetaldehyde and formaldehyde become very low, but we have to study the effects of these parameters on the performance of the engine, which was the goal of my PHD study in the Faculty of Mechanical and Electrical Engineering, Damascus University.

Table 7. Results of using E100

α	CR	Formaldehyde, Mole/cm^3/sec	Acetaldehyde, Mole/cm^3/sec	Formaldehyde, Mole/cm^3/sec	Acetaldehyde, Mole/cm^3/sec
		Initial Gas Temperature, °C			
		20°C		85°C	
0.8	10	1.46E-56	4.01E-55	9.70E-44	3.53E-42
0.8	17	1.46E-56	4.01E-55	9.70E-44	3.53E-42
0.9	10	1.63E-56	4.56E-55	1.08E-43	4.32E-42
0.9	17	1.63E-56	4.56E-55	1.08E-43	4.32E-42
1	10	1.80E-56	5.10E-55	1.20E-43	5.18E-42
1	17	1.80E-56	5.10E-55	1.20E-43	5.18E-42
1.1	10	1.96E-56	5.65E-55	1.31E-43	6.10E-42
1.1	17	1.96E-56	5.65E-55	1.31E-43	6.10E-42
1.2	10	2.13E-56	6.20E-55	1.42E-43	7.08E-42
1.2	17	2.13E-56	6.20E-55	1.42E-43	7.08E-42

Table 8 shows the results of using commercial Ethanol (95%) in HCCI engine. The results are similar to E100, but the amount of water in Ethanol increases the amount of acetaldehyde and formaldehyde in engine’s emission.

Table 8. Results of using Ethanol 95%

α	Formaldehyde, Mole/cm³/sec	Acetaldehyde, Mole/cm³/sec	Formaldehyde, Mole/cm³/sec	Acetaldehyde, Mole/cm³/sec
	Initial Gas Temperature, °C			
	20°C		85°C	
0.8	1.80E-56	9.57E-51	1.11E-43	1.18E-40
0.9	2.00E-56	1.07E-50	1.24E-43	1.44E-40
1	2.19E-56	1.19E-50	1.36E-43	2.01E-40
1.1	2.38E-56	1.31E-50	1.47E-43	1.72E-40
1.2	2.56E-56	1.42E-50	1.59E-43	2.32E-40

Some studies (Davis, 2001; Davis, 2002) proved that adding hydrogen to ethanol improves the combustion, decreases the amount of emissions, improves efficiency, gets lean operation and avoids the cold start problem.

Table 9 shows the results of using blended Ethanol with even 1% hydrogen. The results are similar to E100, but using hydrogen increases dramatically the amount of acetaldehyde and formaldehyde in engine's emission.

Table 9. Results of using Ethanol+1%H2

α	Formaldehyde, Mole/cm³/sec	Acetaldehyde, Mole/cm³/sec	Formaldehyde, Mole/cm³/sec	Acetaldehyde, Mole/cm³/sec
	Initial Gas Temperature, °C			
	20°C		85°C	
0.8	9.13E-52	2.31E-43	5.89E-39	3.65E-34
0.9	1.15E-51	2.86E-43	7.31E-39	4.53E-34
1	1.42E-51	3.47E-43	8.86E-39	5.49E-34
1.1	1.71E-51	4.12E-43	1.05E-38	6.51E-34
1.2	2.03E-51	4.80E-43	1.23E-38	7.60E-34

CONCLUSION

The studies about Ethanol fueled engine proved that ethanol has a great potential to act as an environmental friendly fuel, but using ethanol increases the emitted acetaldehyde and formaldehyde, which are carcinogenic gases.

This problem happened in all kinds of engines (Typical one, EPA, FFV, HCCI).

Using CHEMKIN-PRO as simulation program for HCCI engine conclude the following:

1. Using Ethanol in HCCI engine. The results proved that the amount of acetaldehyde is more than formaldehyde.

2. The changing CR does not affect the amount of acetaldehyde and formaldehyde.
3. When the equivalent Ratio (α) is increasing the amount of acetaldehyde and formaldehyde is increasing. As shown in fig. 10 and fig. 11.

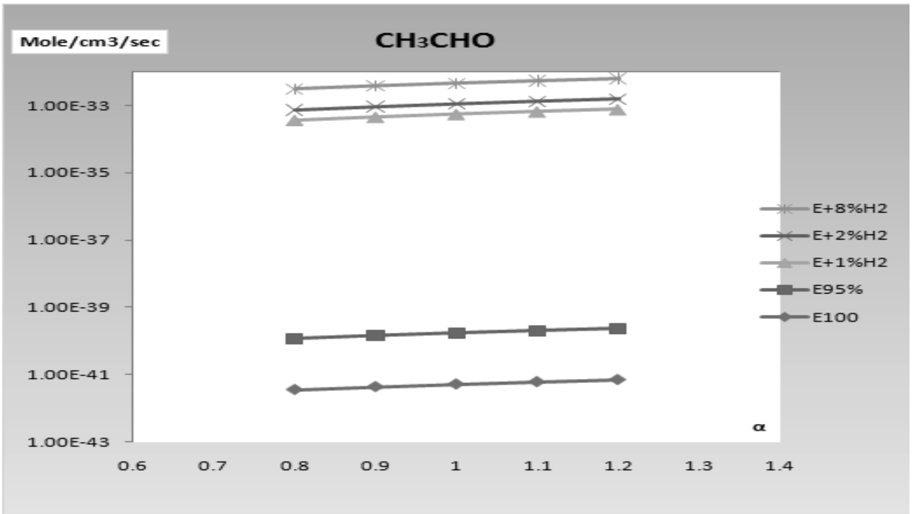

Fig. 10. Acetaldehyde emissions of HCCI engine as function of equivalent Ratio

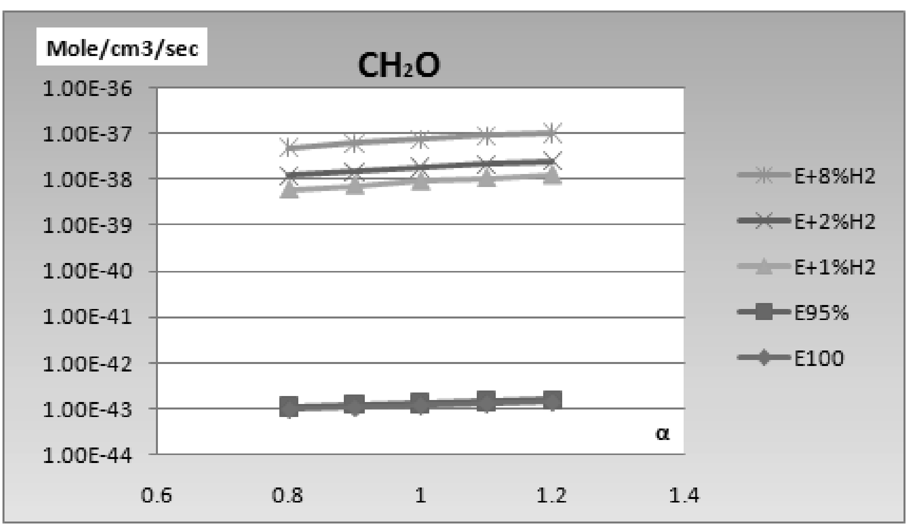

Fig. 11. Formaldehyde emissions of HCCI engine as function of equivalent ratio

4. The initial gas temperature is the most effective parameter (as shown in Figure 12 and Figure 13). When the temperature of the injected ethanol is

low, the acetaldehyde and formaldehyde become very low. But, we have to study the effects of these parameters on the performance of the engine, which is the goal of this PHD study in the Faculty of Mechanical and Electrical Engineering, Damascus University.

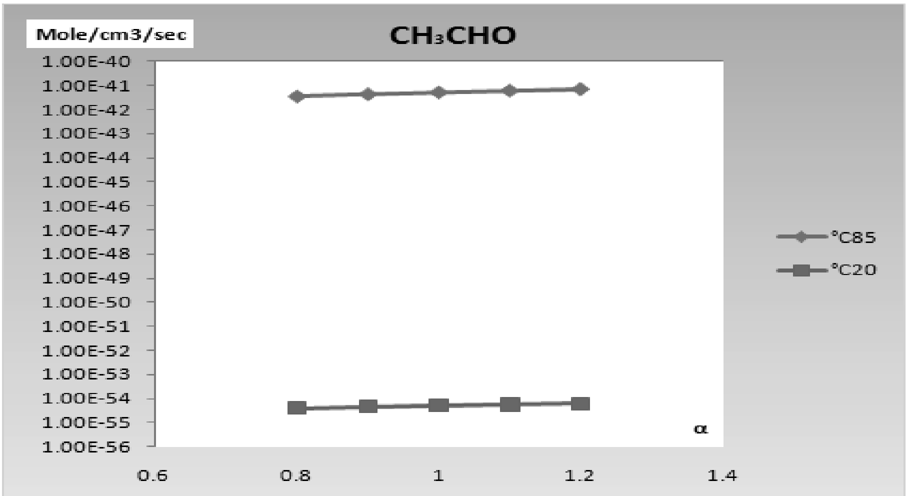

Fig. 12. Acetaldehyde emissions of HCCI engine fueled with Ethanol as function of equivalent ratio and initial gas temperature (20°C and 85°C)

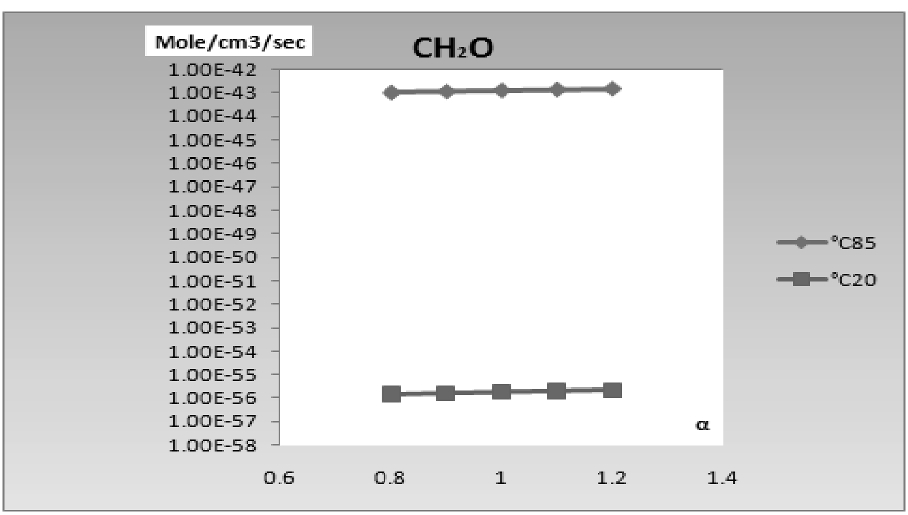

Fig. 13. Formaldehyde emissions of HCCI engine fueled with Ethanol as function of equivalent ratio and initial gas temperature (20°C and 85°C)

5. Using Ethanol makes the amount of acetaldehyde and formaldehyde less than E95 and Ethanol blended with hydrogen.

Future Study

In the frame of PHD study "Analytical and applied study for the methods of treating and reducing acetaldehyde and Formaldehyde emission of the Ethanol fueled engine", the author prepared an applied study to analyze the exhaust gases emitted by spark ignition internal combustion engine fueled by ethanol, to search the quantity of formaldehyde and acetaldehyde by chemical sampling (NIOSH Manual of Analytical Methods, FORMALDEHYDE: METHOD 3500) and the factors that affect the emitted formaldehyde and acetaldehyde.

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