

The effect of heavy naphtha on the blended gasolines and oxygenates

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ABSTRACT

The aim of this paper is to study the effects of the use of heavy naphtha on the blended gasolines and oxygenated gasolines, and enhancing the octane number using different types of alcohols as oxygenates. In addition, reducing the tailpipe emissions is the second aim of this paper. Preparing a different blends of gasoline's with four refinery streams which consists of reformate, isomerate, light naphtha and heavy naphtha in a different ratios gave us a low octane number, so *iso*-butanol and/or *n*-butanol were added as oxygenates by decreasing the isomerate, light naphtha and heavy naphtha ratios in the blends.

Under the environmental consideration, using *iso*-butanol and/or *n*-butanol blended with gasoline is better than methanol because of its renewability and less toxicity. Based on economic and environmental considerations in Egypt, we are interested in studying the effects of *iso*-butanol and/or *n*-butanol contents in the gasoline blends fuel on the engine performance and pollutant emission of a commercial spark ignition (SI) engine.

Key words: Gasolines and oxygenates, Heavy Naphtha.

INTRODUCTION

Recently, there are new trends in gasoline production in order to reduce pollution problems such as hydrocarbons and especially aromatic compounds emissions. This implies a decrease in the volatility of the gasoline and also its octane number. Research and Motor Octane numbers (RON, MON) constitute the main quality characteristics of the gasoline, as they provide a sensitive indication of the anti-knocking behaviour of the fuel. The higher the octane number the better the gasoline resists detonation and the smoother the engine runs. The effect of octane number on detonation has been investigated by several researchers since the octane number of a gasoline is a measure of its resistance to detonation¹⁻⁴. Other important technological properties of the commercial gasolines are the Reid Vapour Pressure

(RVP), American Society for Testing Materials (ASTM) distillation point's flash point, aromatic and sulfur content etc. These properties are monitored during production to ensure the required technological and environmental quality level of the final gasoline^[5]. Although the final gasoline has to meet all the product specifications RON and MON are considered to be the most important ones. This is especially true during the last decade, increasing the compression ratio of motor vehicle engines led to higher requirements in octane rating of the fuels.

Fuel additives are very important, since many of these additives can be added to fuel in order to improve its efficiency and its performance. One of the most important additives to improve fuel performance is oxygenates (oxygen containing organic compounds). Several oxygenates have been

used as fuel additives, such as methanol, ethanol, tertiary butyl alcohol and methyl tertiary butyl ether. Palmer reported that all oxygenated blends gave a better anti-knock performance during low speed acceleration than hydrocarbon fuels of the same octane range⁶. Alcohols have been considered as blending agents to raise the octane number of gasoline and have been used as anti-knock additives to gasoline⁷.

Exhaust emissions from engines are dependent on fuel composition^[8], air/fuel equivalence ratio⁹, driving conditions oxygen content and the chemical structure of additive^[10]. Since tetraethyl lead as gasoline's octane improver was banned in the United States on the first day of January in 1996, oxygenates, which have no differences in air toxicity of ozone forming potential¹¹, have been used to enhance gasoline's octane number, reduce summertime smog, wintertime carbon monoxide and volatile organic compounds with the provision of more complete fuel combustion in engines.

Although the decrease of exhaust emissions by applying oxygenates to engines is small relative to that by catalysts^[12], the fuels containing oxygenates and with aromatics replaced by isoparaffins can reduce hydrocarbon (HC), carbon monoxide (CO) and nitrogen oxides (NOx) emissions¹³. From the literature review, it is understood that alcohol-gasoline blended fuels can effectively lower the pollutant emission without major modifications to engine design^{14,15}.

RESULTS AND DISCUSSION

El-Kady, *et al.*¹⁶ studied the effects of light naphtha on some blended gasolines consists of reformat, isomerate and/or *n*-butanol.

In this manuscript, Heavy naphtha was added to these gasoline blends in different ratios and then some alcohols were added to these blends as oxygenates. The gasoline blends are shown in Table 1 and Table 2 shows the main specifications, octane number, ASTM distillation and the chemical composition of the four refinery streams.

Gasoline blends formulations

Reformat, isomerate, light naphtha and heavy naphtha were blended in two different ratios in volume, the first was 50%, 20%, 15% and 15% respectively yielding the gasoline blend number (1) and the second was 44%, 33%, 11.5% and 11.5% yielding the gasoline blend number (2). Table [3] will show the physical properties of these blends, whereas, ASTM distillation, volatility criteria and driveability index are listed also in this table, and the effects of the heavy naphtha on the composition and on the octane number are clearly visible in Table 3.

A marketed 90 RON gasoline (G) was supplied to be compared with the gasoline blends. Thus, specifications, octane number, chemical composition, ASTM distillation volatility criteria and driveability index were tested for this marketed gasoline sample and shown also at Table 3.

Table 1: The formulated gasoline blends

Formulated blends	Added oxygenate stream	Blended refinery gasoline blends	Designation of samples	Total
Gasoline blends	-	(Reformat Isomerate L. Naphtha H. Naphtha)	1,2	2
B-gasoline blends	<i>n</i> -Butanol	(Reformat Isomerate L. Naphtha H. Naphtha)	1B,2B,3B,4B	4
	<i>n</i> -Butanol <i>iso</i> -Butanol	(Reformat Isomerate L. Naphtha H. Naphtha)	5B,6B,7B,8B	4

Table 2: The main Specifications, octane number, ASTM Distillation and GC analyses of gasoline components

Gasoline components	Reformate	Isomerase	Light Naphtha	Heavy Naphtha
Specifications				
Density @ 15/4°C g/L	0.7950	0.6515	0.6883	0.7138
R.V.P Psi, Kg/cm ²	3.1 (0.22)	12.6 (0.89)	7.5 (0.53)	5.5 (0.39)
Sulphur, % wt (ppm)	0.009(9)	0.016(16)	0.008(8)	0.050(50)
Corrosion, Copper stripTest 3hrs @ 50 °C	1A	1A	1A	1A
Oxidation Stability, mint.	>480	>480	>480	>480
Antiknock Index:-				
RON	94	86	68	61
MON	82	78	60	55
(R + M) / 2	88	82	64	58
ASTM Distillation				
IBP	60	32	44	49
5%	82	37	56	58
10%	88	39	58	60
20%	96	40	60	65
30%	104	42	63	74
40%	111	44	66	84
50%	118	46	69	94
60%	125	49	71	104
70%	133	53	77	115
80%	144	58	82	124
90%	157	69	91	143
95%	166	73	102	155
FBP	180	79	106	163
Recovered, vol%	98.8	98.5	99.0	98.0
Loss, vol%	0.6	1.0	0.5	1.5
Residue, vol%	0.6	0.5	0.5	0.5
Chemical composition, % wt				
iso-Butane	0.000	0.173	0.035	0.000
n-Butane	0.649	1.654	1.061	0.153
iso-Pentane	1.721	36.542	19.776	0.606
n-Pentane	1.110	8.235	21.062	0.823
2,2-Dimethylbutane	0.314	14.142	0.686	0.036
Cyclopentane	0.139	1.523	2.266	0.145
2,3-Dimethylbutane	0.380	4.076	1.710	0.140
2-Methylpentane	2.444	12.192	10.383	0.809
3-Methylpentane	1.952	6.985	7.096	0.632
n-Hexane	2.854	4.051	13.240	1.568
Methylcyclopentane	0.987	2.748	6.683	1.214
Benzene	3.468	0.000	2.281	0.369
Cyclohexane	0.075	3.866	2.981	0.986
C ₇ ⁺	83.907	3.813	10.740	92.519
Total	100.00	100.00	100.00	100.00

Table 3: Specifications, Octane Number and ASTM Distillation volatility criteria of two refinery streams.

Gasoline components	Gasoline No		
	1	2	G
Reformate, vol%	50	44	-
Isomerase, vol%	20	33	-
Light Naphtha, vol%	15	11.5	-
Heavy Naphtha, vol%	15	11.5	-
Total Blend, vol%	100	100	-
Specifications:-			
Density @ 15/4 °C g/L	0.7468	0.7337	0.7608
R.V.P Psi, Kg/cm ²	6.04 (0.42)	6.90 (0.49)	5.60 (0.39)
Sulphur, % wt (ppm)	0.044(44)	0.045(45)	0.020(20)
Corrosion, Copper stripTest 3hrs @ 50 °C	1A	1A	1A
Oxidation Stability, min.	>480	>480	>480
Chemical composition, % wt.			
Total Aromatics	36.365	32.333	46.016
Total Iso-paraffins	35.030	39.869	33.293
Total Naphthenes	10.973	10.996	5.763
Total Olefins	0.443	0.312	0.528
Total Paraffins	17.189	16.492	14.400
Total Hydrocarbons	100	100	100
Benzene, % wt.	1.98	1.80	1.96
Antiknock Index:-			
RON	85	86	90
MON	79	80	85.5
(R + M) / 2	82	83	88
ASTM Distillation:-			
IBP	42	39	49
5%	56	50	69
10%	63	55	75
20%	71	62	83
30%	81	70	91
40%	91	79	101
50%	97	91	110
60%	107	95	120
70%	113	106	129
80%	130	122	142
90%	145	144	158
95%	156	155	176
FBP	174	171	185
Recovered, vol%	98.9	99.0	98.4
Loss, vol%	0.5	0.5	0.6
Residue, vol%	0.6	0.5	1.0
E70 vol. %	20	30	5
E100 vol. %	54	65	40
E150 vol. %	93	93	85
Driveability Index (DI), °C	531	500	600

It is clear from the table 3 that the percentage volume ratio for the reformats, light naphtha and heavy naphtha were decreased whereas the isomate percentage volume ratio increased to reduce the aromatic contents which are very high in the reformat. Additionally, increasing the isomate percentage was to compensate the loss of octane number.

Fig. 1 illustrates the effect of blend composition on volatility criteria via showing the full distillation profiles of these two hydrocarbon gasoline blends. E70, E100 or E150 (the volume percentage of gasoline distilled at 70, 100 or 150 °C respectively) for the gasoline blends are shown in table 3.

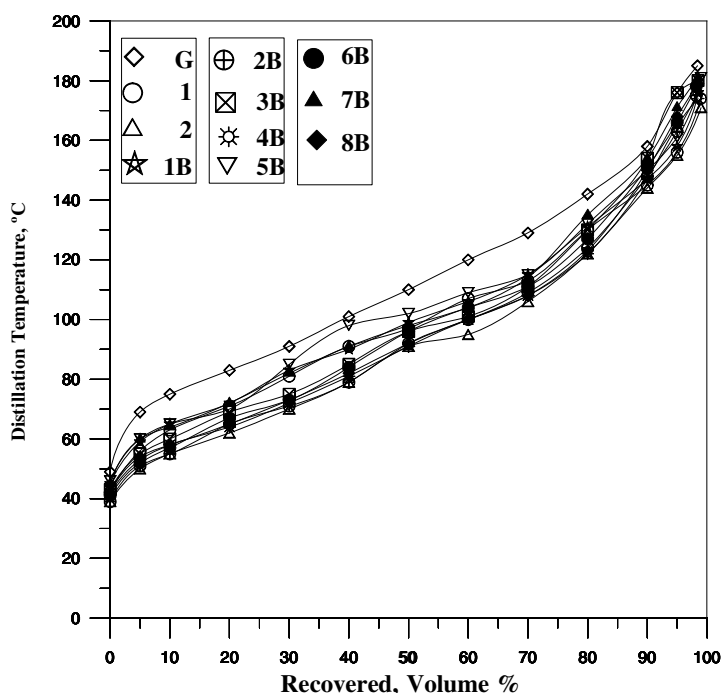


Fig. 1: Distillation profiles of [1B to 8B] and hydrocarbon gasoline blends [1,2] compared with (G).

Oxygenated gasoline blend in 2.0 wt. %

Gasoline blends No. (1B) and (2B) are oxygenated with *n*-butanol (94 RON) relative to O₂ in 2.0 weight percent which equal 9.25 volume percent. The employed hydrocarbon fuel components reformat, isomate, light naphtha and heavy naphtha were exchanged in sufficient quantities for the formulation of these two gasoline blends specified in table 4. Butanol was studied in a single cylinder engine as a fuel¹⁷.

Table 4 shows the specifications, chemical composition, distillation and octane numbers of formulation consisting of four refinery streams and *n*-butanol in 2.0 weight percent. Reid vapor pressure (RVP) values are between 6.0-6.4 psi. These RVP

values are relatively lower than that for Reformulated Gasoline (RFG) in U.S.A (RVP for California phase II RFG is 6.7-7.0 psi), then is required to be lower to reduce evaporate emissions and also reduces the amount of vaporization during cold start and warm-up¹⁸.

Aromatic contents 34.398-28.600 weight percent and trace amount of olefins, benzene weight percent range is 1.880-1.690 and 40 ppm sulfur content for the two blends. A copper corrosively standard ensures that the resultant oxygenated fuels will not create excessive corrosion in the vehicle fuel system. Oxidation stability also indicates that blending *n*-butanol will not alter the fuel's storage life.

Table 4: Specifications, Octane Number and ASTM Distillation volatility criteria of oxygenated four refinery streams with *n*-butanol and/or iso-butanol in 2.0 and 2.7 wt.%

Gasoline components	Gasoline No							
	1B	2B	3B	4B	5B	6B	7B	8B
Reformate, vol%	50	44	50	44	50	44	50	44
Isomerase, vol%	20	33	20	33	20	33	20	33
Light Naphtha, vol%	10.375	6.875	10.375	6.875	8.75	5.25	8.75	5.25
Heavy Naphtha, vol%	10.375	6.875	10.375	6.875	8.75	5.25	8.75	5.25
Hydrocarbon	90.75	90.75	90.75	90.75	87.5	87.5	87.5	87.5
Gasoline, vol%								
Added Oxygenate, vol%	9.25	9.25	9.25	9.25	12.5	12.5	12.5	12.5
Total Blend, vol%	100	100	100	100	100	100	100	100
Oxygenate type	n- BuOH	n- BuOH	n- BuOH	n- BuOH	n- BuOH	n- BuOH	n- BuOH	n- BuOH
Oxygen content, wt%	2.0	2.0	2.0	2.0	2.7	2.7	2.7	2.7
Specifications								
Density @ 15/4 °C g/L	0.7555	0.7422	0.7600	0.7427	0.7708	0.7589	0.7512	0.7381
R.V.P Psi, Kg/cm ² (0.42)	6.0 (0.45)	6.4 (0.41)	5.9 (0.44)	6.2 (0.40)	5.7 (0.41)	5.8 (0.44)	6.2 (0.46)	6.5
Sulphur, % wt (ppm)	0.012 (40)	0.040 (40)	0.040 (20)	0.020 (16)	0.016 (40)	0.040 (40)	0.040 (18)	0.018 (12)
Corrosion, Copper strip Test 3hrs @ 50 °C	1A	1A	1A	1A	1A	1A	1A	1A
Oxidation Stability, min.	480<	480<	480<	480<	480<	480<	480<	480<
Chemical composition, % wt.								
Total Aromatics	34.398	28.600	34.000	28.121	33.579	27.540	33.356	26.001
Total Iso-paraffins	33.904	38.946	34.411	39.714	35.052	38.160	35.574	40.559
Total Naphthenes	9.298	7.480	8.299	7.959	6.913	6.633	6.848	6.876
Total Olefins	0.210	0.116	0.401	0.527	0.066	0.088	0.375	0.512
Total Paraffins	12.950	15.804	13.619	14.306	10.210	14.928	10.852	13.497
Total Oxygenates	9.240	9.054	9.271	9.373	14.180	12.651	13.001	12.555
Total Hydrocarbons	100	100	100	100	100	100	100	100
Benzene, % wt.	1.880	1.690	1.712	1.573	1.870	1.571	1.512	1.021
Antiknock Index								
RON	88	89	88.5	90	89	90	90	92
MON	85	85	85.5	86.5	85	86	86.5	87.5
(R + M) / 2	86.5	87	87	88	87	88	88	90
ASTM Distillation								
IBP	45	39	43	42	46	41	45	42
5%	59	51	55	53	60	52	60	54
10%	65	55	60	58	65	57	64	58
20%	72	65	69	64	70	67	72	65
30%	83	71	75	72	85	73	82	73

40%	90	79	85	81	98	84	91	82
50%	99	92	96	91	102	96	98	92
60%	106	100	101	100	109	104	104	100
70%	115	109	111	108	115	111	114	108
80%	131	124	130	122	132	127	135	123
90%	147	148	154	147	150	151	154	150
95%	158	163	176	165	161	166	171	168
FBP	176	175	180	174	181	178	181	179
Recovered, vol%	98.5	98.2	98.5	98.5	99.0	98.2	98.5	98.5
Loss, vol%	1.0	1.0	1.0	1.0	0.5	1.0	1.0	1.0
Residue, vol%	0.5	0.8	0.5	0.5	0.5	0.8	0.5	0.5
E70 vol. %	17	27	22	27	19	25	18	31
E100 vol. %	53	60	58	60	45	55	59	60
E150 vol. %	91	91	89	91	90	90	88	90
Driveability Index (DI), °C 543		507	532	507	560	525	547	513

Table 5 : Tailpipe emissions

Blend	HC (PPM)	% Improvement	CO % (Vol.)	% Improvement	NOx (PPM)	% Improvement	CO ₂ (% Vol.)	% Improvement
G	830	-	6.80	-	4	-	11.50	-
1	950	(-) 14.46	5.25	(+) 22.79	3	(+) 25.00	10.23	(+) 11.04
2	915	(-) 10.24	5.08	(+) 25.29	2	(+) 50.00	10.02	(+) 12.87
1B	760	(+) 8.43	4.72	(+) 30.59	1	(+) 75.00	9.72	(+) 10.47
2B	713	(+) 10.10	4.56	(+) 32.94	1	(+) 75.00	9.45	(+) 15.48
3B	630	(+) 24.10	4.28	(+) 37.06	0	(+) 100.00	9.68	(+) 15.83
4B	520	(+) 37.35	4.10	(+) 39.71	0	(+) 100.00	9.21	(+) 19.91
5B	642	(+) 22.65	4.50	(+) 33.82	1	(+) 75.00	9.44	(+) 17.91
6B	617	(+) 25.66	4.39	(+) 35.44	1	(+) 75.00	9.19	(+) 20.09
7B	480	(+) 42.17	4.02	(+) 40.88	0	(+) 100.00	8.88	(+) 22.78
8B	411	(+) 50.48	3.85	(+) 43.38	0	(+) 100.00	8.53	(+) 25.83

Percent improvement is calculated as shown in the following equation:

$$\% \text{Improvement} = \frac{E_o - E_b}{E_b} \times 100$$

Where:

E_o = Emission of oxygenated blend.

E_b = Emission of the base fuel.

The range of RON is 88-89 respectively and MON is 85 for the two. Thus the antiknock index (AKI) was ranged from 86.5-87 respectively and related with the reformate percent due to its aromatic content which contributes more efficiently in antiknock property than isomerate¹⁹.

Using *n*-butanol and *iso*-butanol by 1:1 ratio in the gasoline blends and also in 2.0 weight percent will increase the (AKI) due to the highly RON of the *iso*-butanol (113 RON). The formulation of the two newly oxygenated blends (3B) and (4B) are shown in table 4.

The most important standards relating to driveability are octane number and volatility, but another specification must be taken into consideration as benzene percent which must not exceed than 2.0 weight percent, sulfur contents are below 50 ppm, oxidation stability larger than 480 minutes and corrosion problems must not be founded in these oxygenated gasolines. Considering the last specifications for these oxygenated gasoline blends (3B) and (4B), sulfur contents range are 20-16 ppm, oxidation stability larger than 480 minutes, corrosion, Copper strip results are 1A for all and the (R+M)/2 values are in the range 87-88. RVP values of these formulations are found to range between 5.9-6.2 psi; T50 and T90 are in the range 91-96°C and 154-147°C respectively. ASTM distillation, volatility criteria and driveability index are listed in table 4.

Oxygenated gasoline blend in 2.7 wt. %

Increasing the oxygen content to 2.7 weight percent which equal 12.5 volume percent with *n*-butanol will enhance the research octane number and improve the composition of the generated blends number (5B) and (6B).

Further addition of oxygenate results in more reduction in RVP values and an increase in antiknock value is observed. Table 4 which also show the distillation profiles, volatility characteristics and driveability indexes. The measured E100 values of these formulations are lower than all hydrocarbon blends as illustrated in fig. 1. In this figure, distillation curves at the bottom represent all hydrocarbon gasoline blends (1,2) which have the highest volatilities. Distillation curve at the top represent oxygenated gasoline blends No. 5B, and 6B, which have the lowest volatilities.

In continuous from table 4, oxygenated gasoline blends (5B) and (6B) have a range of aromatic contents 33.579-27.540 weight percent, trace amount of olefins and 1.870-1.751 weight percent benzene. Sulfur content 40 ppm for the two blends, copper corrosively standard ensures that the resultant oxygenated fuels will not create excessive corrosion in the vehicle fuel system. The antiknock performance of these blends observes that the range of RON and MON are 89-90 and 85-86 respectively for (5B) and (6B). Thus the (AKI) was ranged from

87-88 respectively. Oxidation stability also indicates that blending *n*-butanol will not alter the fuel's storage life.

Furthermore, adding *n*-butanol and *iso*-butanol by 1:1 ratio to the gasoline blends and also in 2.7 weight percent will reach the maximum RON that can be obtained in these blends which called (7B) and (8B).

The Reid vapour pressure (RVP) values are 6.2-6.5 psi and the aromatic contents are 33.356-26.001 weight percent for (7B) and (8B) respectively. Benzene content is 1.512 weight percent for oxygenated gasoline blend No. (7B) and 1.021 weight percent for (8B). Sulfur content 18-12 ppm, copper corrosively standard ensures that the resultant oxygenated fuels will not create excessive corrosion in the vehicle fuel system. The antiknock performance of these two blends observes that the range of RON and MON are 90-92 and 86.5-87.5 respectively for (7B) and (8B) yielding antiknock index (AKI) with values which are 88 and 90 respectively due to increasing the *iso*-butanol percent rather than 3B and 4B. Oxidation stability is over 480 minutes in its induction periods for all the three refinery gasoline blends. T50 and T90 which varied from 98-92°C and 154-150°C are slightly alerted in a narrow range for (7B) and (8B) respectively.

Other specifications as ASTM distillation, volatility criteria and driveability index are listed in table [4]. At 2.0 and 2.7 weight percent oxygen level, the measured E100 values of formulations (1B:8B) are relatively lower than hydrocarbon blend No. (2). E100 values are better illustrated in fig. 1 for all oxygenated gasolines (1B:8B) when compared with hydrocarbon blends No. (1,2) and marketed 90 RON gasoline (G). Alteration is less severe in front-end or tail-end volatilities (E70 and E150) than for E100.

Tailpipe emissions

The tailpipe emissions, the volatility properties of gasoline and unburned hydrocarbons in the exhaust were responsible for the majority of pollutants²⁰.

The four main emissions which are hydrocarbon (HC), carbon monoxide (CO), nitrogen

oxide (NO_x) and carbon dioxide (CO₂), were studied via comparison between marketed 90 RON gasoline (G) with blended (1,2) and the oxygenated gasolines (1B : 8B) as shown in table 5.

All of these blends results are in the specification of Egyptian Environmental Law 4/1994.

The net reduction in tailpipe hydrocarbon emissions (HC) for every oxygenated blend would be expected to reach the maximum values at 2.7 wt % oxygen in (8B). Variable improvements were shown in CO, NO_x and CO₂ emissions, but it is noticeable that the presence of *iso*-butanol enhances these emissions exclusively for NO_x emissions²¹.

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