

The Bio-ethanol and ETBE Effects on Volatility Properties of Oxygenated Gasoline

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Abstract

The purpose of this paper is to analyze the Reid vapour pressure (RPV) and distillation curves of four synthetic gasoline partial substituted with 2-10 % v. ethanol or 4-15 % v. ETBE. Several parameters that describe the distillation curve were investigated. These parameters specify the percentage of the gasoline that evaporates at 70°C (E70), 100°C (E100), and 150°C (E150). The Vapor Lock Index (VLI) is also calculated from the RPV and E70 parameters. Finally, the advantages and disadvantages of ethanol and ETBE, as oxy-compounds for gasoline partial substitution are highlighted.

Key words: *bio-ethanol, ETBE, gasoline, volatility properties*

Introduction

The oxygenated bio components as substitutes for petroleum-based gasoline are increasingly used to increase the renewable energy content of road transport fuels. The Renewable Energy Directive (RED) (2009/28/EC) requires that 10% of sustainable produced renewable energy must be embedded into transport fuels by 2020 [10]. Only common products, such as ethanol and fatty acid methyl esters (FAME), seem to be available in sufficient quantities to meet the RED mandate. In conjunction of this Directive and as part of the same legislative package, the European Parliament introduced the revised Fuel Quality Directive (FQD) 2009/30/EC [11].

The directive provide a gradual reduction of greenhouse gases emissions per unit of energy supplied for the transportation sector with a 6 % reduction target over the 2010 - 2020 time frame. The FQD request an increase of the percentage of oxygenated bio components in fuel, up to a total content of oxygen of 3.7 % wt. According to the expected quality standard of commercial gasoline, will be allowed an increasing content of primary alcohols (methanol, ethanol, isopropyl alcohol, iso-butyl alcohol) as well as ether's content. The current European Norm EN 228 specification permitted a maximum of 2.7 % wt oxygen. Current and targeted oxygen content in gasoline are showed in Table 1.

The first oxygenated compound used as octanic additive for gasoline was methyl *tert*-butyl ether (MTBE). Nowadays, more than 85% of reformulated gasoline contained MTBE, because of the lowered price, low vapour pressure, total miscibility with gasoline, medium boiling point and reduction of fuel consumption [12]. Despite these huge advantages, MTBE is 30 times more soluble in water than hydrocarbons. This undesired property led to a ban on the use of MTBE in gasoline, starting with 2001, in the USA [24]. International experiences suggest using other ethers, such as ethyl *tert*-butyl ether (ETBE) for partial substitution of petroleum-based

gasoline. Recent research has indicated as favourable the usage of ETBE as a partially renewable component because in its synthesis bio ethanol can be used [9].

Table 1. EN 228 Standard specifications regarding the oxygen content and oxygenated compounds in gasoline [7, 14].

Characteristic	Current specifications	Target specifications
Oxygen content, % wt.	Max 2.7	3.7
Oxygen compounds content, % v:		
-methanol	3	3
-ethanol	5	10
-isopropyl alcohol	10	12
-iso-butyl alcohol	7	15
- tert-butyl alcohol	10	15
-ethers	15	22
-other oxygenated compounds	10	15

Bio alcohols were provided by researchers and accepted by refiners as components for gasoline substitution. Bio ethanol is preferred option in worldwide gasoline [26]. The use of ethanol as a bio component in gasoline has a set of drawbacks. Most important disadvantages are related to volatility and stability on contact with water [1, 3, 4].

Recent studies show favourable impact of the superior bio alcohols in gasoline blend [15]. Biobutanol have the advantage of lower consumption, a low content of oxygen in the molecule, a very low vapour pressure and much lower affinity towards water than ethanol [2, 8, 22, 27]. Butanol can be obtained by renewable materials through fermentation processes, but with lower capability and higher recovery costs than the bio ethanol production. For now, the bioethanol remain the most economical option.

The aim of this paper is the experimental determination of Reid vapour pressure (RPV) and distillation curves of two synthetic gasolines partial substituted with 2-10 % v. ethanol and other two synthetic gasolines partially substituted with 4-15 % v. ETBE. Several parameters that describe the distillation curve were investigated. These parameters specify the percentage of the gasoline that evaporates at 70°C (E70), 100°C (E100), and 150°C (E150). The Vapor Lock Index (VLI) is also calculated from the RPV and E70 parameters. By interpreting the experimental results and the calculated ones, the advantages and disadvantages of ethanol and ETBE are highlighted.

The European Gasoline Volatility Specifications

Quality standards mark a large number of properties which must be ensured by fuel suppliers. The EN 228 specifies the volatility requirements for gasoline in order to ensure good performance of vehicles. Different gasoline volatility classes are included in the EN 228 specification that include minimum and maximum volatility limits for summer and winter gasoline as well for seasonal transitions periods (Table 2).

From the point of view of the six volatility classes of commercial gasoline, classes *A* (RPV for summertime: 45.0-60.0 kPa) and *D/DI* (RPV for wintertime: 60.0-90.0 kPa) are relevant.

Table 2. The requirements of volatility classes according to EN 228 standard [17].

Volatility class	Vapour pressure, kPa	Evaporated at 70°C, E70, % v.	Evaporated at 100°C, E100, % v.	Evaporated at 150°C, E150, % v.	Final boiling point, °C, max	Distillation residue, % v., max	VLI (10VP+7E70) Index, max
A	45-60	20-48	46-71	75	210	2	-
B	45-70	20-48	46-71	75	210	2	-
C/C1	50-80	22-50	46-71	75	210	2	C (-) C1 (1050)
D/D1	60-90	22-50	46-71	75	210	2	D (-) D1 (1150)
E/E1	65-95	22-50	46-71	75	210	2	E (-) E1 (1200)
F/F1	70-100	22-50	46-71	75	210	2	F (-) F1 (1250)

Experimental Studies

The experimental research started with the preparation of four conventional gasoline blends, noted Base A, B, C and D. Three components were mixed in different proportions: the catalytic cracking gasoline, the catalytic reforming gasoline and an isomerization fraction. All conventional gasoline's were analysed from the point of view of volatility (distillation curves and Reid pressure vapour), total aromatics, olefinic and parafinic hydrocarbons. The results of the experimental determinations of the main gasoline characteristics, as well as the lab equipment used in these experiments, are presented in table 3.

Table 3. Conventional gasoline characterisation and experimental equipment.

Properties	Values and UM				Lab Equipment
	Base A	Base B	Base C	Base D	
Hydrocarbons content:					IROX 2000 Fuel Analyzer Portable Gasoline Analysis with MID-FTIR
-aromatic (Aro)	40.3% v.	36.3 % v.	38.1 %v .	33.9 % v.	
-olefinic (Ole)	8.6 % v.	3.2 % v.	12.9 % v.	11.5 % v.	
-saturated (Sat)	51.1 % v.	60.5 % v.	49.0 % v.	54.6 % v.	
Reid pressure vapour	56 kPa	59.8 kPa	56 kPa	63.7 kPa	MINIVAP VPS/VPSH Vapor Pressure Tester
Distillation curves:					MINIDIS Analyzer Portable Minidistillation
- Initial	44.8 °C	43.9 °C	41.1 °C	41.8 °C	
- E70	24.8 % v.	25.2 % v.	33.0 % v.	35.0 % v.	
- E100	43.0 % v.	45.1 % v.	51.5 % v.	53.0 % v.	
- E150	74.5 % v.	75.4 % v.	78.0 % v.	78.0 % v.	
- FBP	198.3 °C	196.4 °C	190.4 °C	195 °C	

In accordance with EN228 European standard, Base A conventional gasoline clearly belonging in A volatility class taking into consideration the RPV value and E70, E100 and E150 values, whilst gasoline B is placed at the superior limit of A class. Based on its RPV value, gasoline Base C seem to be placed in A volatility class, despite the E150 value which exceed the standard specification. Base gasoline D belongs in B volatility class.

Results and Discussions

The first volatility property analysis was the Reid pressure vapour of ethanol-gasoline and ETBE-gasoline blends. In order to demonstrate the way in which the oxygenated compound type affects the volatility properties of gasoline, we prepared oxygenated gasoline blends with ethanol and ETBE, respectively. Out of gasoline A and B blends with 2, 4, 6, 7, 10 % v. of ethanol and out of gasoline C and D blends with 4, 7, 10, 15 % v. of ETBE were prepared. In this study, the oxygenated blends (OB) of base gasoline (Base A and Base B) with ethanol (E) are symbolized as: OBA-E2, OBA-E4, OBA-E6, OBA-E7, OBA-E10 (numbers represent the ethanol concentration in OB). Similar symbolizations are for Base B, e.g. OBB-E2. The experimental results and the changes in RPV are presented according to the oxygen content of these blends in Table 4.

Table 4. Ranges of measured RPV and calculated change in RPV values at each ethanol concentration in OB.

Ethanol content, %v.	Oxygen content in OB, %wt.	RPV of OBA-E, kPa,	Change in RPV of OBA-E, kPa	RPV of OBB-E, kPa,	Change in RPV of OBB-E, kPa
0 (Base)	0.00	56.0	Base A	59.86	Base B
2	0.77	57.0	1.0	61.59	1.73
4	1.54	58.3	2.3	62.45	2.58
6	2.30	60.2	4.2	63.54	3.68
8	2.68	61.6	5.6	63.74	3.87
10	3.81	61.5	5.5	63.06	3.20

Reid pressure vapour for the OBA and OBB gasoline with 2-10 % v. ethanol are influenced by the vapour pressures of conventional gasoline, such as those of ethanol. The change in RPV increase by increasing the ethanol content in gasoline blends. By substituting base gasoline A with 6 % v. ethanol, we can obtain blends that, from the point of view of Reid pressure vapour, are situated at the superior limit of usage during summer. Blending gasoline A with approx. 8 % v. ethanol leads to obtaining 2.7 % wt oxygen content in gasoline and a vapour pressure that makes it suitable in the transition seasons [20]. In case of OB provided from Base B, all blends are suitable for transition seasons. It is can be pointed that the change in RPVs of OBB-E (in any proportion, but without exceeding the maximum oxygen content), are lesser than in case of OBA-E. The change in RPV values of ethanol-gasoline blends are implications in commercial gasoline formulation. In practice, conventional gasoline, with different C4 hydrocarbons proportion, are formulated just with the purpose of increasing vapour pressure. For summer blends of gasoline with ethanol, formulations with the C4 fraction must be avoided, or dealt with cautiously [21].

In order to discuss the experimental results, the oxygenated blends (OB) of base gasoline (Base C and Base D) with ETBE (Eb) are symbolized as: OBC-Eb4, OBC-Eb7, OBC-Eb10, OBC-Eb15 (numbers represent the ETBE concentration in OB). Similar symbolizations are for Base D, e.g. OBD-Eb4. The experimental results and the changes in RPV are presented according to the oxygen content of these blends in Table 5.

As a consequence of the low pressures of pure ETBE, vapour pressures for gasoline C or D oxygenated with ETBE decrease. According to EN 228 standard, judging only by the vapour pressure, OBs with ether are all placed in summertime classes. It is beneficial to note that through adding C4 hydrocarbons, their pressure can be increased up to suitable transition seasons or wintertime values [19].

Table 5. Ranges of measured RPV and calculated change in RPV values at each ETBE concentration in OB.

ETBE content, %v.	Oxygen content in OB, %wt.	RPV of OBC-Eb, kPa,	Change in RPV of OBC-Eb, kPa	RPV of OBD-Eb, kPa,	Change in RPV of OBD-Eb, kPa
0 (Base)	0.00	56.00	Base C	63.70	Base D
4	0.62	55.14	-0.86	60.10	-3.60
7	1.10	54.47	-1.53	58.03	-5.67
10	1.55	53.80	-2.20	56.70	-7.00
15	2.32	52.78	-3.22	54.62	-9.08

The distillation curves were determined experimentally for all oxygenated gasoline and base gasoline. Distillation curves for OBA-E, OBB-E compared with Base A and Base B gasoline are graphically represented in Fig. 1 and Fig. 2, respectively. In addition to increasing the vapour pressure of the ethanol-gasoline blends, ethanol also changes the shape of the blend’s distillation curves. This has the potential to impact the vehicle’s regulated emissions and driveability performance in cold and hot weather.

The distillation curves of gasoline A and B are strongly influenced by the presence, in different proportions, of ethanol. The distillation curves of gasoline blends with ethanol show a region that indicates the formation of azeotropes with minimum boiling temperature between ethanol and some light hydrocarbons in the gasoline [5, 13, 20]. The decrease in the boiling temperatures of gasoline-alcohols blends boiling temperatures in the azeotrope regions has a positive effect upon the vehicle easy start, but favours the appearance of vapour locks and increase of exhaust hydrocarbons [21].

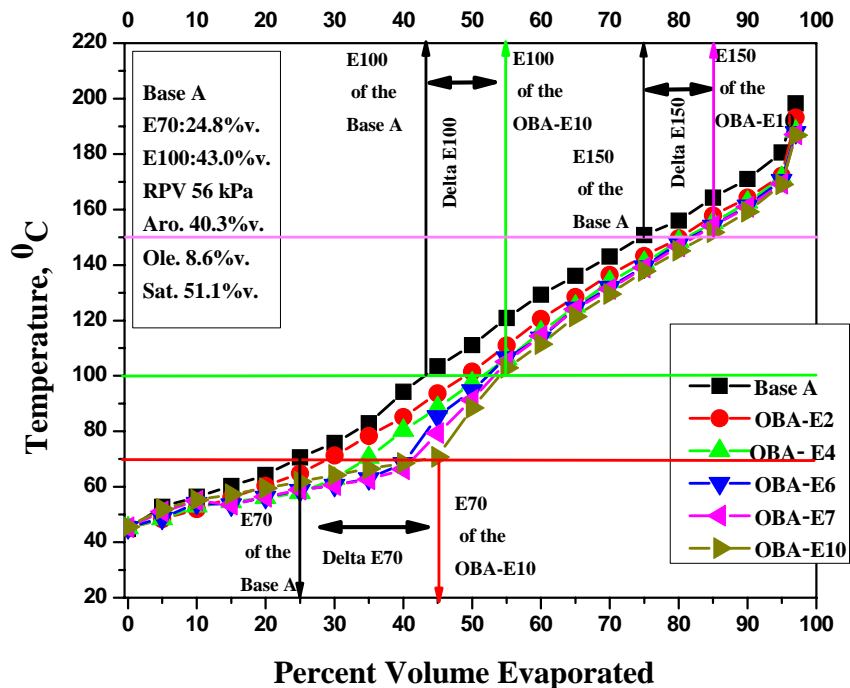


Fig. 1. Distillation curves of Base A and oxygenated blends of Base A with ethanol.

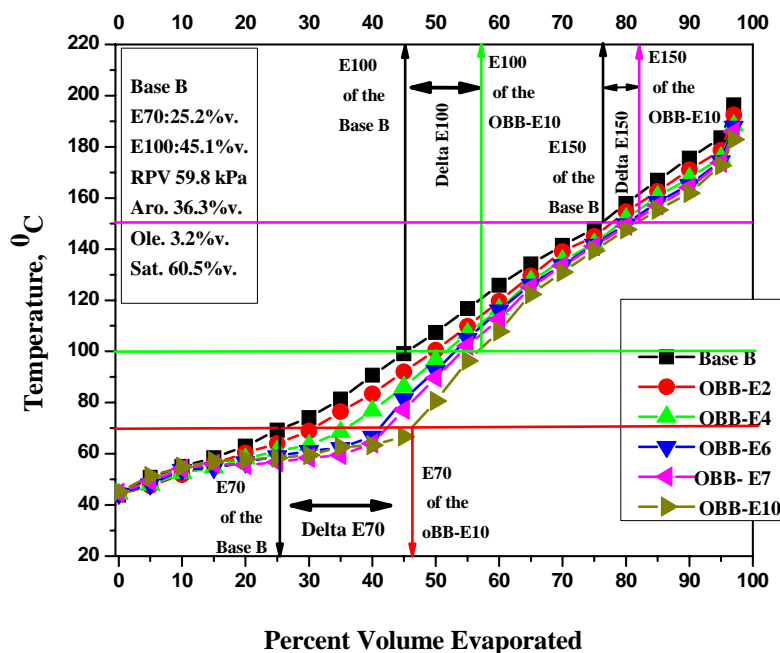


Fig. 2. Distillation curves of Base B and oxygenated blends of Base B with ethanol.

Between the distillation curves of oxygenated gasoline with ethanol and of oxygenated gasoline with ETBE there is a major difference (Fig. 3 and Fig. 4). The ETBE do not form azeotropes with the hydrocarbons in the gasoline; his only has an influence on the temperature at which the evaporation of hydrocarbons takes place. Hence, in the distillation curves of gasoline oxygenated with ETBE only the boiling temperatures of the pure ether and the chemical composition of conventional C and D gasoline are influenced.

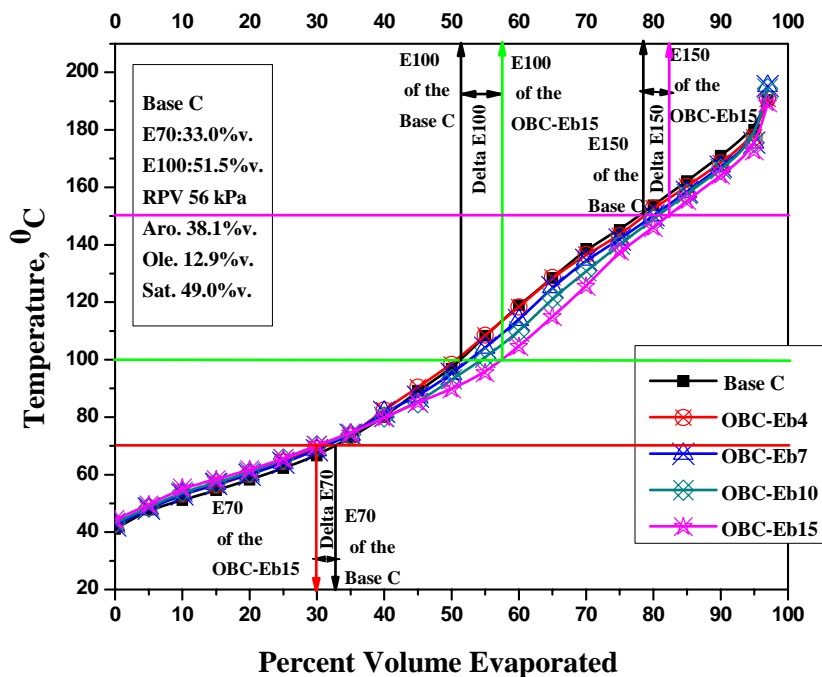


Fig. 3. Distillation curves of Base C and oxygenated blends of Base C with ETBE.

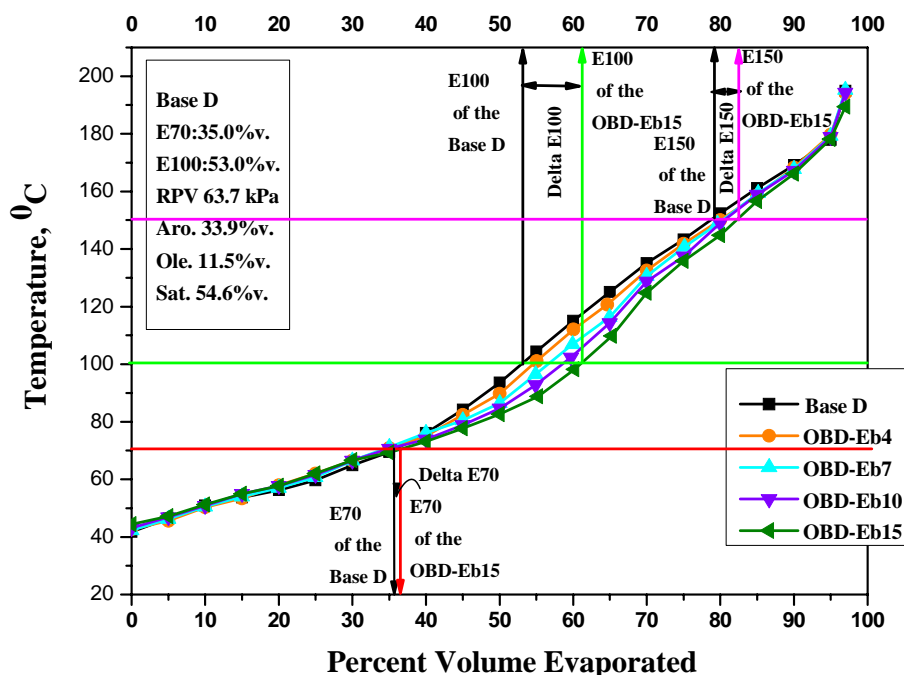


Fig. 4. Distillation curves of Base D and oxygenated blends of Base D with ETBE.

The difference between the behaviours of the ethanol and ETBE towards the distillation curves is better outlined by analyzing the values of evaporated percentage of the fuel at 70°C (E70), at 100°C (E100) and 150°C (E150). The comparison of these values that are representative for the distillation curves with the ones indicated in EN 228 standard allow us to classify the oxygenated gasoline according to the classes of volatility. Based on Figs. 1 and 2, the E70, E100 and E150 values for gasoline blends with ethanol are represented graphically in Fig. 5, Fig. 6 and Fig. 7, respectively.

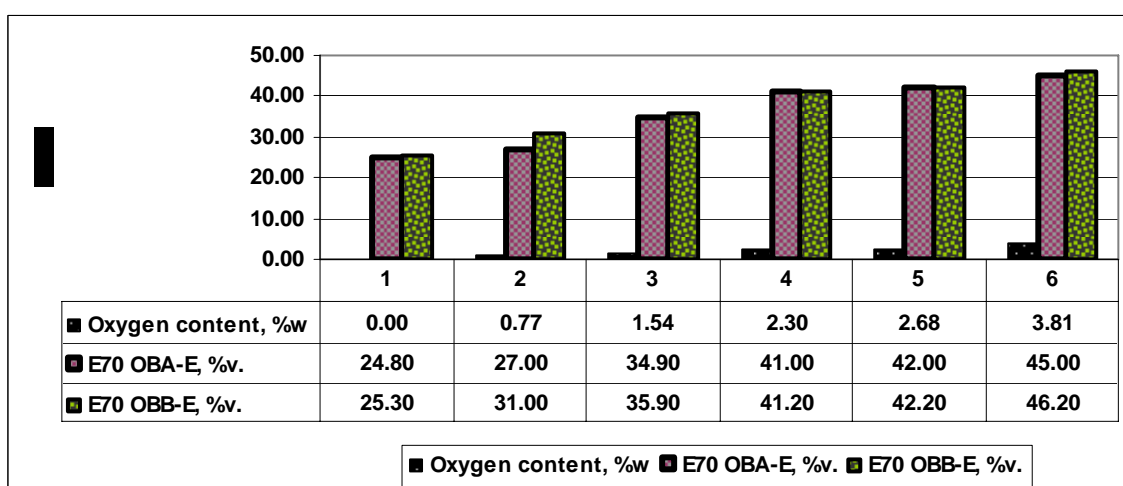


Fig. 5. Evaporated percentage of fuel at 70°C for conventional gasoline (Base A and Base B) and their oxygenated blends with ethanol

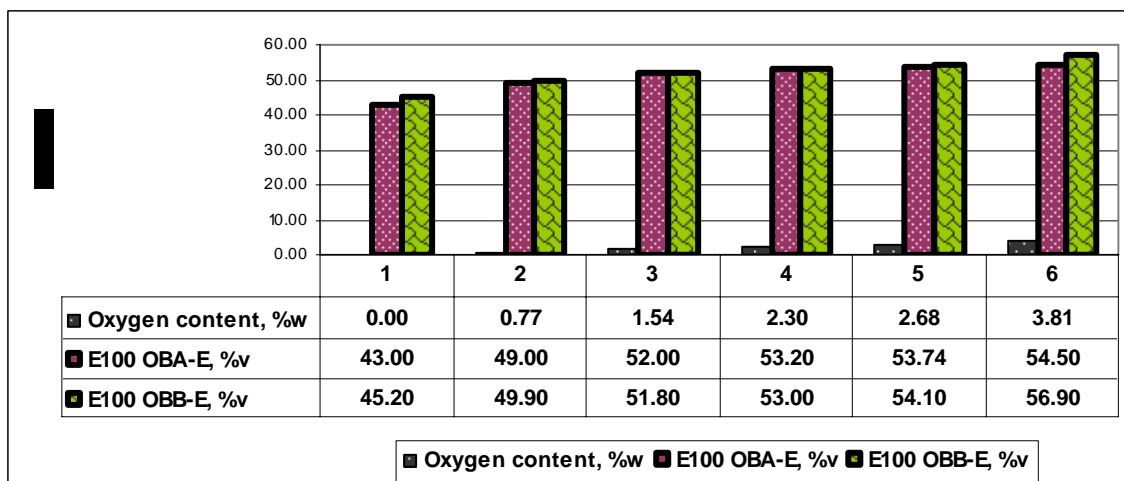


Fig. 6. Evaporated percentage of fuel at 100°C for conventional gasoline (Base A and Base B) and their oxygenated blends with ethanol

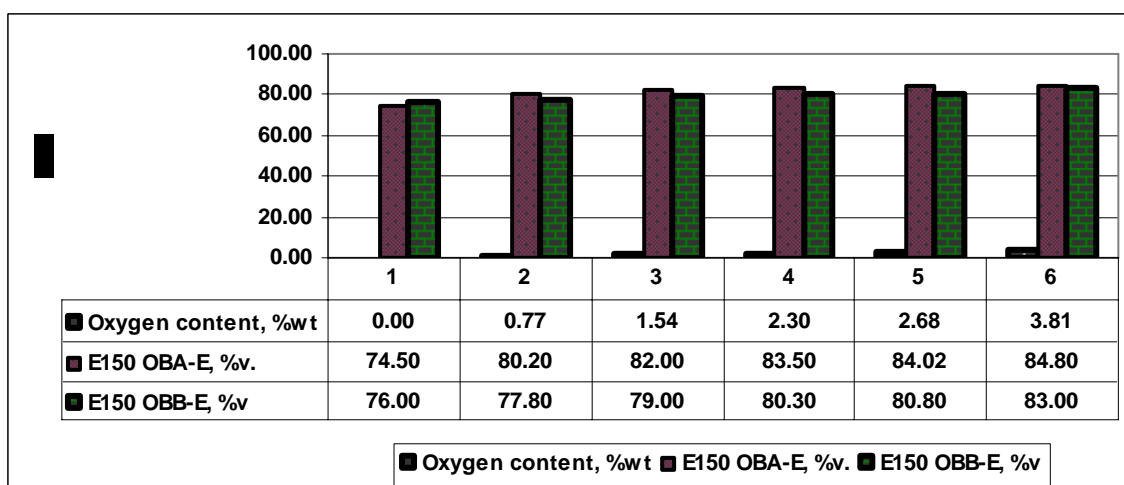


Fig. 7. Evaporated percentage of fuel at 150°C for conventional gasoline (Base A and Base B) and their oxygenated blends with ethanol

Fig. 5 shows an increase in E70 values of the oxygenated blends with increasing ethanol concentration. The E70 values are especially affected because the distillation curves of the ethanol-gasoline blends indicate the formation of azeotropes (constant boiling point mixtures) between ethanol and the light hydrocarbons C5-C8 comprising in the blends. In addition, the type of the base gasoline influences the E70 values of oxygenated gasoline, as follows: for the same content of ethanol, the E70 values for blends with B gasoline are slightly higher than the ones for A gasoline because B gasoline has a higher content of saturated hydrocarbons, which influences the region of minimum boiling points azeotropes. The changes in E70 values have positive and negative effect in the vehicle performance. Namely, the azeotrope regions have a positive effect upon the vehicle easy start, but favour the appearance of vapour locks and increase of exhaust hydrocarbons. From the point of view of the values of the evaporated percentage at 70°C, all gasoline-ethanol blends are enclosed in volatility classes A or B (i.e. in the domain of 20-48% v. values), according to the gasoline quality standard EN 228.

Similar effect but smaller increases in the E100 distillation values are observed with increasing ethanol content (Fig. 6). The presence of ethanol introduces some advantage by increasing evaporated percentages at 100°C, this signifies that the vehicle will offer an easier start-up and

lower fuel consumption. In terms of E100 values all the gasoline-ethanol mixtures were found to be in the domain of the 46-71% v., according to the gasoline quality standard EN 228.

Evaporation, at a temperature of 150°C, is favoured by the presence of ethanol. This is which translates into the following effects: a decrease in fuel consumption on long-trips, an enhancement in lubricating oil dilution and a decrease in engine deposits. Ethanol has a stronger contribution to the evaporation of the heavier hydrocarbons (Fig. 7). From the point of view of the quality standards of fuels EN 228, the evaporation percentage at a temperature of 150°C for all gasoline-ethanol blends is over 75.

The E70, E100 and E150 values for gasoline blends with ETBE are represented graphically in Fig. 8, Fig. 9 and Fig. 10 respectively.

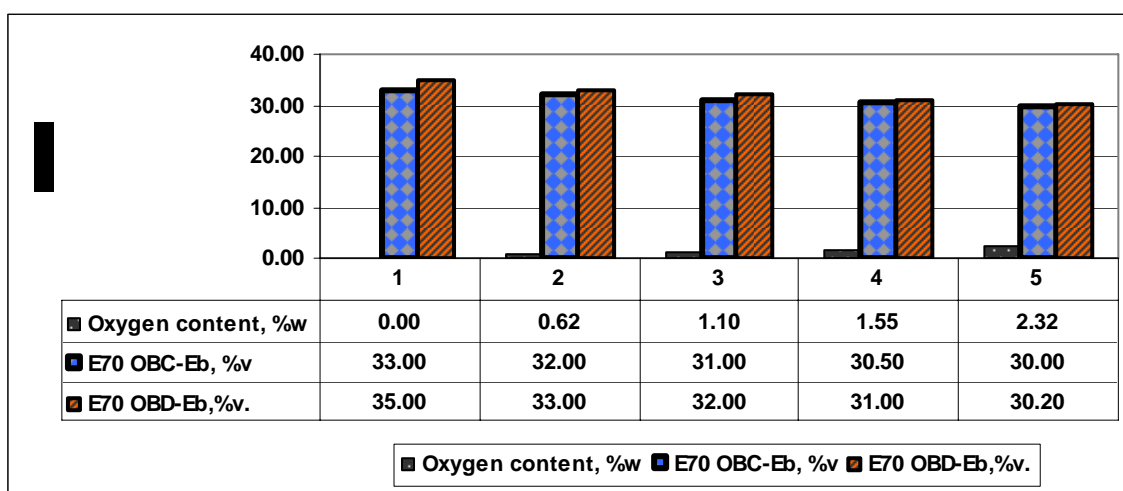


Fig. 8. Evaporated percentage of fuel at 70°C for conventional gasoline (Base C and Base D) and their oxygenated blends with ETBE

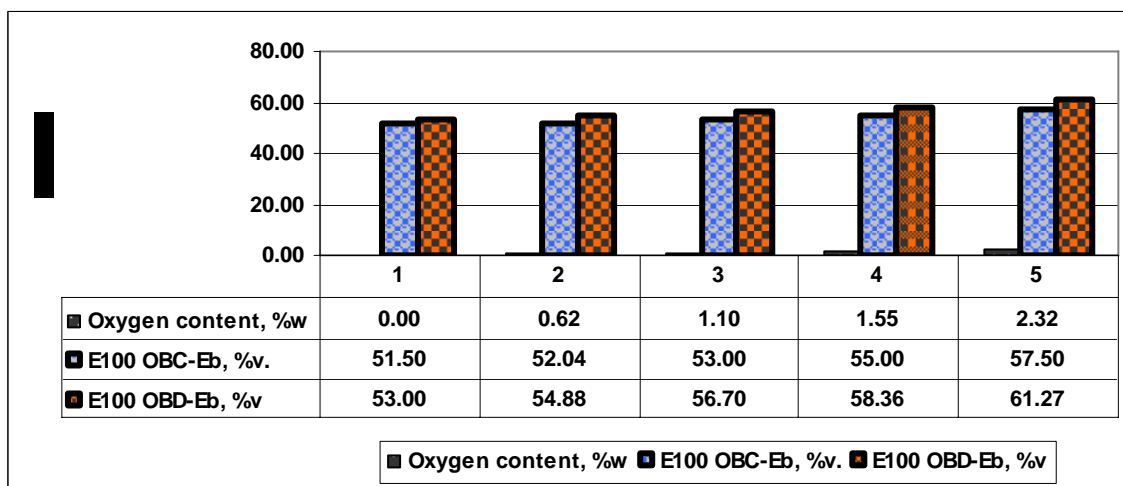


Fig. 9. Evaporated percentage of fuel at 100°C for conventional gasoline (Base C and Base D) and their oxygenated blends with ETBE

In the case of the gasoline oxygenated with ETBE, the evaporated percents at the temperature of 70°C slightly decrease with ETBE concentration increasing in the blends (Fig. 8). This behaviour is justified by the boiling temperature of ETBE (i.e., 72°C). The introduction of ETBE disadvantage in some way the evaporation of his mixtures with hydrocarbons from the curves of the gasoline distillation region around 70°C. Furthermore, it was observed that

gasoline oxygenated with ETBE, has a minimal influence on hydrocarbon evaporation in the start region of the distillation curve.

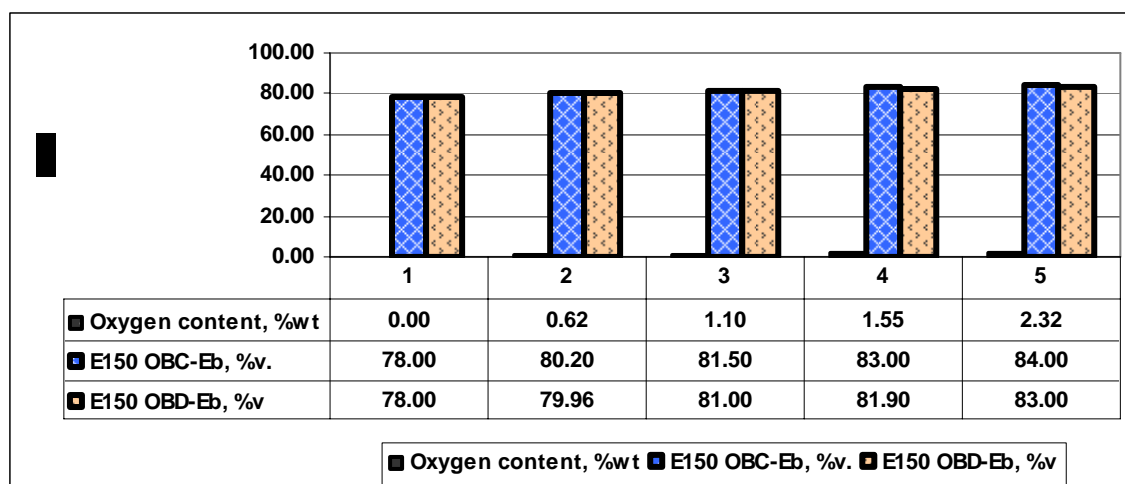


Fig. 10. Evaporated percentage of fuel at 150°C for conventional gasoline (Base C and Base D) and their oxygenated blends with ETBE

The E100 values of the oxygenated gasoline increase with ETBE concentration increasing in blends (Fig. 9). This influence is linked by the ETBE boiling point temperature. Thus, ETBE influences in a positive way the evaporation at the temperature of 100°C. The positive effect is more marked at gasoline D, more volatile than gasoline C. The gasoline oxygenated with ETBE, through the fact that it evaporates more readily at 100°C, as compared to conventional gasoline, means that it offers an easier start-up and lower fuel consumption.

The percentages evaporated at a temperature of 150°C from oxy-gasoline with ETBE increase proportionally with ETBE concentration. The effect is more visible with gasoline C (less volatile), rather than with gasoline D. In the case of gasoline oxygenated with ether, the percentage evaporated at a temperature of 150°C is over 75, according to standard EN228.

The finally volatility property is the vapour lock index *VLI*. This is the volatility properties that indicate the fuel tendency to form vapor plugs. *VLI* is calculated ($VLI = 10 \cdot VP + 7 \cdot E70$) based on the value of the vapour pressure experimentally determined and the evaporated percentages at a temperature of 70°C (Table 6).

Table 6. Vapour lock index *VLI*

Gasoline type	Base A	OBA-E2	OBA-E4	OBA-E6	OBA-E7	OBA-E10
<i>VLI</i>	733.60	759.00	827.30	889.00	910.00	925.00
Gasoline type	Base B	OBB-E2	OBB-E4	OBB-E6	OBB-E7	OBB-E10
<i>VLI</i>	775.72	832.89	875.75	923.82	932.77	947.71
Gasoline type	Base C	OBC-Eb4	OBC-Eb7	OBC-Eb10	OBC-Eb15	-
<i>VLI</i>	790.00	774.00	759.00	746.50	725.00	-
Gasoline type	Base D	OBD-Eb4	OBD-Eb7	OBD-Eb10	OBD-Eb15	-
<i>VLI</i>	882.00	832.00	804.00	784.00	757.40	-

Data from Table 6 shows that the VLIs of oxygenated gasoline with ethanol are higher than those of non-oxygenated gasoline (Base A and Base B). This is an expected behaviour because both RPVs and E70 values increase in ethanol-gasoline blends. An opposite effect is on VLI values of the ETBE-gasoline blends.

Conclusions

In this paper were evaluated the volatility properties of non-oxygenated and oxygenated gasoline. Experimental researches show that the volatilities of the blends are strongly depended by the base gasoline properties as well as the oxygenated compound type and its concentration. Regarding RPV values of the investigated oxygenated gasoline, our conclusions are in concordance with those of the recent studies and experimental research. Literature results showed increases in the RPV of ethanol-gasoline blends [6, 16, 18, 23, 25]. An opposite behavior was depicted in ETBE-gasoline blends; the RPV values decrease by adding ETBE in gasoline. Taking into account the experimental RPV results, all oxygenated gasoline are placed in the A volatility class, according to the EN 228 standard. But one major difference has from ethanol-oxygenated and ETBE oxygenated gasoline. In order to avoid the overlap of ethanol's effect with C4 fraction's effect on RPV of its blends, the formulation with C4 hydrocarbons fraction of the base gasoline dedicated to ethanol blends must be avoided (or dealt with cautiously). No same particularity was depicted in ETBE-gasoline blends.

The distillation curves of the gasoline oxygenated with bio-ethanol are disturbed over its entire ranges. All representative values of distillation curves (E70, E100 and E 150) increase. Despite that those values are placed in A volatility class, the ethanol-gasoline blends seem not to be suitable for summer's commercial fuels. Or rather, the future EN 228 standard must be adjusted in order to reflect more properly the ethanol effect over the volatility properties of oxygenated gasoline.

Oxy-gasoline with ETBE displays more beneficial volatility properties than bio-ethanol. ETBE, has a minimal influence on hydrocarbon evaporation in the start region of the distillation curve, but has positive influence (increase) on hydrocarbon evaporation in the middle and end region of the distillation curve. It is beneficial to note that through adding C4 hydrocarbons, their volatility can be increased. Such flexibility in ETBE-oxygenated gasoline favours them in commercial gasoline producers' options. ETBE has another advantage: in order to achieve a maximum content of oxygen in the gasoline, larger quantities of ETBE can be used.

In the future, partially renewable ETBE oxy-compound, have great potential to partial substitutes the conventional gasoline.

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Efectul bio-etanolului și ETBE asupra proprietăților de volatilitate ale benzinelor oxigenate

Rezumat

Scopul acestei lucrări este să analizeze presiunea de vapori și curbele de distilare a patru benzine sintetice substituite parțial cu 2-10 % vol. etanol sau 4-15 % vol. ETBE. Au fost investigați câțiva parametri care descriu curbele de distilare. Acești parametri specifică procentul evaporat din benzine la 70°C (E70), 100°C (E100) și 150°C (E150). De asemenea a fost calculat indicele de vapori (Vapor Lock Index -VLI) din mărimile RPV și E70. În final au fost subliniate avantajele și dezavantajele etanolului și ETBE ca și compuși oxigenați pentru substituirea parțială a benzinelor.