

The Potential Environmental Benefits of Utilising Oxy-Compounds as Additives in Gasoline, a Laboratory Based Study

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1. Introduction

Resolving the conflict between growth oriented powers, which tend to extend polluting emissions has become a focus of 21st century politics.

The growth of transportation requirements in modern society implies the consumption of large quantities of fuel. It is a fact that fossil fuel reserves have dramatically dropped and can no longer sustain the ever growing demand. The need to protect existing crude oil resources has also fuelled the search for alternative renewable energy resources that are compatible with auto fuel. Furthermore these fuels must face another challenge: through their burning in the vehicles' engines they have to reduce polluting exhaust emissions.

An innovating solution, through which the relation between transportation and the environment is reconciled, is the substitution of auto fuel with different proportions of oxygenated biocomponent. The introduction of a certain oxygen percentage in auto gasoline started in 1970, with the purpose of increasing the number of octanes of gasoline, as a replacement of tetraethyl lead (TEL). The first oxygenated compound used as an octanic additive for gasoline was methyl *tert*-butyl ether (MTBE). Shortly after introduction, it was discovered that using MTBE leads to a reduction of polluting exhaust emissions. For a gasoline with 15% MTBE, carbon monoxide emissions are reduced by 10-15%, nitric oxide by 1.0-1.7%, and total hydrocarbon emissions are reduced by 10-20% (Song et al., 2006). 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 (He et al., 2003). However, MTBE is 30 times more soluble in water than hydrocarbons. This undesired property has proved in time that, when MTBE, reformulated gasoline leaks from underground storage tanks or auto tanks, it moves through soil to groundwater thus contaminating it. The subsequent environmental and human health problems led to a ban on the use of MTBE in gasoline, starting with 2001, in the USA (Poulopoulos & Philippopoulos, 2001).

In Europe, the USA's point of view regarding the use of MTBE is not fully agreed upon. The benefits of using MTBE as a gasoline additive, from the point of view of improving the

quality of fuel and the reduction of exhaust emissions in the atmosphere are more obvious than the drawback created by possible leaks from the storage tanks (Osman et al., 1993; Tavlarides et al., 2000; Zervas et al., 2004). Nevertheless, social pressures have determined the promotion of other oxygenated compounds. The first solution provided by researchers and accepted by refiners and the users was to revert back to use of ethanol. Tests of exhaust emissions of vehicles fuelled by gasoline oxygenated with 10% ethanol have shown a reduction of 4.7-5.8% in carbon monoxide and 5-15.3% total hydrocarbons emissions, as well as a reduction in the emitted levels of nitric oxide. The reduction of exhaust emissions also depends on the fuel burning efficiency, operating conditions of the engine (speeds and charges), ethanol content in the gasoline and the air/fuel ratio (He et al., 2003). Still, using ethanol as a biocomponent in gasoline has its set of drawbacks. The heat value of ethanol is less than that of gasoline. Consequently, the heat value of ethanol blended gasoline fuels will decrease when the proportion of ethanol increases (Hsieh et al., 2002). The most controversial aspects are related to volatility and stability on contact with water (Aakko & Nylund, 2004; Bayraktar, 2005; Cataluña et al., 2008). Recent studies and experimental research motivated by the presence of ethanol in fuel showed increases in the vapour pressure of reformulated gasoline, which is dependent upon the content of saturated hydrocarbons, the vapour pressure of the base fuel and ethanol content in the mixture (da Silva et al., 2005; Martini et al., 2007; Muzíková et al., 2009; Pospíšil et al., 2007; Pumphrey et al., 2000; Rosca et al., 2009). 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 (D'Ornellas, 2001; Hsieh et al., 2002; Neagu et al., 2010). This in turn significantly contributes to reformulated gasoline volatility, prevalence of vapour lock and the loss of emissions by evaporation and increased acetaldehyde emissions (Poulopoulos & Philippopoulos, 2001; Zervas et al., 2001, 2002). Recent studies concerning the use of superior alcohols in reformulated gasoline have shown surprising results. Isopropyl alcohol as well as the butanols can be extracted from the same renewable materials as the ethanol, through fermentation processes, but with lower capability and higher recovery costs. Although obtaining these bioalcohols seems to be uneconomical, using them in auto gasoline produces more favourable results than the addition of ethanol (Brekke, 2007). Biobutanols have the advantage of lower consumption (it has almost the same energy content as the petroleum-based gasoline: 26-27 MJ/litre of butanol and 32-33 MJ/litre gasoline), research octane number 94, has a low content of oxygen in the molecule and thus can be mixed in a bigger proportion with gasoline (up until 16%), a very low vapour pressure (VOC emissions are reduced) and much lower affinity towards water than ethanol (Arnold, 2008; Szulczyk, 2010).

Ethers' role as a substitute for auto gasoline has increased since the 1990s. Since ether has a lower vapour pressure than ethanol it also causes a reduction of essential organic compounds in the exhaust gases. Directly connected to the essential exhaust compounds is the level of the ozone and its role in global warming.

The position of oxygenated biocomponents as substitutes for petroleum-based gasoline consolidated with the fight for reducing the emissions of greenhouse gases (Aakko & Nylund, 2004; He et al., 2003; Szklo et al., 2007). Carbon dioxide emitted by the burning of oxygenated biocomponents can be remedied to a large extent by growing plants that can absorb the carbon dioxide. The introduction of an oxygen percentage in gasoline is legally sustained and mandatory in all countries of the European Union. At the end of 2008, the

European Parliament introduced a legislative package in the area of energy and climatic changes which sets targets for reducing greenhouse gases in the period of time following the Kyoto Protocol (2008-2012), with medium (2020) and long (2050) term targets. A part of this legislative package is the revised Fuel Quality Directive 2009/30/EC (EC, 2009). The directive appeals the gradual reduction of GHG emissions per unit of energy from fuel and energy supplied for the transportation sector with a 6% reduction target over the 2010 - 2020 time frame. Another request of the directive is to increase the percentage of oxygenated compounds in fuel, up to a total content of oxygen of 3.5 %weight. The changing of the total content of oxygenated compounds, as well as other quality specifications of fuels was stipulated by revising international standards (Table 1). The quality of commercial gasoline in Europe is set by EN 228 Standard (Dixson-Declève & Szalkowska, 2009).

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

Table 1. EN 228 Standard specifications regarding the content of oxygenated compounds in gasoline

The quality standard of commercial gasoline predicts the growth of oxygenated compounds content, primary alcohols (methanol, ethanol, isopropyl alcohol, iso-butyl alcohol), as well as ether compounds. The role of the ether oxygenated compounds (with five or more carbon atoms in the molecule) in gasoline formulation will be decisive because their content could increase from 15 up to 22 vol.%. International experiences related to using methyl *tert*-butyl ether (MTBE) and *tert*-amyl-methyl ether (TAME) as additives for commercial gasoline suggest using other ethers as well, such as ethyl *tert*-butyl ether (ETBE) and *tert*-amyl ethyl ether (TAEE) 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 production process bioethanol can be used, just like in the case of TAEE synthesis.

Extensive research on the properties of various blends of biocomponents in petroleum-based fuel has been conducted, in particular research that emphasises the advantages and disadvantages of using these products in existing vehicles' engines as well as in the new vehicles' engines. At the request of the auto manufacturers, at least until 2013, the oxygen content in gasoline will remain at a maximum 2.7 weight%.

In the first part of this research the physical-chemical properties of oxygenated compounds, primary alcohol and ethers, are presented, as well as their main properties on which petroleum-based and oxygenated gasoline are accepted as commercial products.

The second part is dedicated to a laborious experimental study on the volatility and octane properties of gasoline oxygenated with different proportions of primary bioalcohols and ether. Our research aimed, firstly, on the experimental determination of vapour pressure and distillation curves of synthetic gasoline partially substituted with 2-10% vol methanol, ethanol, isopropanol, tert-butanol and other synthetic gasoline partially substituted with 4-15 vol.% MTBE, TAME, ETBE, TAEE (Neagu et. al., 2010, 2011). Based on these properties, vapour lock indexes were calculated. By interpreting the experimental results and the calculated ones, the advantages and disadvantages of each oxygenated compound are highlighted. In the final part of this study, a comparison between the investigated properties of the oxygenated gasoline with bioalcohols and those with ethers is presented.

2. The oxy-compounds used as bio-substitutes for petroleum-based gasoline

A relatively recent classification divides biocomponents into alcohols and ethers. The main difference between the two categories is that in the alcohols, each hydroxyl functional group

Alcohol	MeOH	EtOH	IPA	TBA
CAS number	67-56-1	64-17-5	67-63-0	75-65-0
Chemical formula	CH ₄ O	C ₂ H ₆ O	C ₃ H ₈ O	C ₄ H ₁₀ O
Molecular weight, g/mol	32.04	46.07	60.10	74.12
Density (at 20 C), g/cm ³	0.792	0.789	0.786	0.781
Boiling temperature, °C	64.7	78.3	82.3	82.3
RVP, kPa	31.7	16.0	12.6	5.5
Oxygen, wt. %	49.9	34.7	26.6	21.6
Solubility in water	miscible	miscible	miscible	miscible
Blending RON/MON	122-133/92	121-130/96	117-118/95-98	105-109/94-95

Table 2. The physical and chemical properties of fuel oxygenates type alcohols as partial substitutes compounds of commercial gasoline (Lesnik, 2002; Nylund et al., 2008)

Ether	MTBE	TAME	ETBE	TAEE
CAS number	1634-04-4	994-05-8	637-92-3	919-94-8
Chemical formula	C ₅ H ₁₂ O	C ₆ H ₁₄ O	C ₆ H ₁₄ O	C ₇ H ₁₆ O
Molecular weight, g/mol	88.15	102.18	102.18	116.20
Density, g/cm ³	0.741	0.764	0.752	0.750
Boiling temperature, C	55.2	86.3	72.2	101.42
RVP, kPa	55	10	28	NA (not available)
Oxygen, wt. %	18.15	15.7	15.66	13.77
Solubility in water	4.8 g/100 g	1.2 g/100 g	1.2 g/100 g	NA
Blending RON/MON	115-118/101	109-112/98-99	117-119/102-103	NA

Table 3. The physical and chemical properties of fuel oxygenates type ethers as partial substitute compounds of commercial gasoline (Lesnik, 2002; Nylund et al., 2008).

(-OH) is bound to a carbon atom, usually connected to other carbon or hydrogen atoms, and in ethers, each oxygen atom is linked to two alkyl groups. Among the alcohols used into the formulation of commercial gasoline are the following: MeOH (methanol), EtOH (ethanol), IPA (iso-propyl alcohol) and TBA (tert-butyl alcohol). In Table 2 are included some physical and chemical properties of alcohols.

The ethers most commonly used as substitutes for gasoline are: MTBE (methyl *tert*-butyl ether), TAME (*tert*-amyl methyl ether), ETBE (ethyl *tert*-butyl ether), TAEE (*tert*-amyl ethyl ether). Among the ethers, MTBE is the most widely used oxygenate compound, followed by ETBE and TAME. In Table 3 are included some physical and chemical properties of ethers.

3. Commercial gasoline properties

Quality standards indicate a large number of properties that must be met by suppliers of fuel. Drivers are generally unaware of the complexity of fuel products, and the manufacturers' efforts to produce high quality fuel that can satisfy consumer needs whilst also generating low emissions. From the point of view of the drivers it is important only if the engine has an easy start in the cold season, warms up rapidly, the engine has adequate power without knocking, provides good fuel economy, there are no deposits or corrosion in the fuel system and last but not least, if the fuel is cheap.

Below we will present some of the most important characteristics of gasoline and how they affect driving performance.

Fuel volatility is the gasoline's tendency to vaporize. In cold weather, gasoline is blended to vaporize easily. In the warm season, the gasoline is blended to vaporise less easily in the combustion chamber of an engine in order to prevent vapour lock or other hot-fuel handling problems and to minimize evaporation, which contributes to an increase in hydrocarbon emissions. According to EN228 European standard the properties that are being used to measure gasoline volatility are: Reid vapour pressure, distillation profiles and Vapour Lock Index (VLI).

The vapour pressure of a fuel must be high enough to be able to supply a smooth start of the engine, but not too high so that it does not contribute to the appearance of a vapour lock or excessive evaporate emissions. *The distillation curve* of gasoline is a graphic representation of the variation in the boiling temperature according to the volume percentage of distilled (evaporated). The gasoline is made of a variety of chemical compounds which evaporate at different temperatures. The more volatile components evaporate at lower temperatures, while the less volatile ones evaporate at higher temperatures.

The various regions of the distillation curve can be correlated with the gasoline and driving performance:

- the starting area should provide: easier cold starting and when warm, avoid vapour plugs and have low emissions;
- the middle area should provide: rapid warm-up and smooth running, fuel economy for short-trip, good engine power;
- the final area should provide: fuel economy for long-trips, freedom from engine deposits, minimal dilution of the lubricating oil and low exhaust emissions of volatile organic compounds.

On the basis of the evaporated percentage at a temperature of 70°C (E70) and of Reid pressure vapour, a parameter used to control vapour lock and other hot-fuel handling problems, can be calculated; namely the VLI (*Vapour Lock Index*).

$$VLI = 10 \cdot VP + 7 \cdot E70 \quad (1)$$

The European Union's EN228 gasoline specification controls the volatility of gasoline by setting limits for the vapour pressure, distillation profile (evaporated percentage at three temperatures and final boiling point), the percentage of maximum distillation residue and VLI properties (Table 4). The European Standard employs six volatility classes according to the summer, winter and transition periods.

Volatility class	Vapour pressure kPa	Evaporated at 70°C, E70, % vol	Evaporated at 100°C, E100, % vol	Evaporated at 150°C, E150, % vol	Final boiling point, °C, max	Distillation residue, % vol, 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)

Table 4. The requirements of volatility classes according to EN 228 standard (Motor Gasolines Technical Review, 2009)

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/D1 (RPV for wintertime: 60.0-90.0 kPa) are relevant.

The octane number characterises the antiknock performances of auto gasoline. There are two laboratory tests through which the number of octanes is determined: a test that is made on an engine with a single cylinder in mild-knocking conditions and low speed and which has as a result the *research octane number* (RON) and a test that is made on the same type of engine, but in high speed and high-temperature knocking conditions and which has as a result the *motor octane number* (MON) of tested gasoline. A fast way to estimate the number of octanes is using the IROX 2000 Fuel Analyzer Portable Gasoline Analysis with MID-FTIR, from Grabner Instruments.

4. Volatility and octane properties of oxygenated gasoline. Experimental and discussions

Of the primary alcohols, as biocomponents to partially substitute commercial gasoline, the most studied is bioethanol. Many experimental studies underlined that, through the combustion of bioethanol-hydrocarbons blends, emissions through evaporation and the prevalence of vapour lock increases. Both effects are due to modifications to the volatility properties of bioethanol oxygenated gasoline. Based on these drawbacks of bioethanol, we aim to develop an experimental study that addresses all of the gasoline volatility properties oxygenated with primary alcohols (1 to 4 carbon atoms in the molecule). In particular the volatility properties of iso-propyl alcohol and tert-butyl alcohol gasoline blends are investigated, because they have been less studied. In order to have a complete picture of the effect of all bio-alcohols, we have included in the experimental study other important properties of mixtures with gasoline, such as antiknock. Finally, by comparing the properties of volatility (Reid vapour pressure, distillation curves and parameters characteristic curves of distillation and vapour lock index) and the octane numbers (RON and MON) of oxygenated gasoline with each of the primary alcohols (methanol, ethanol, iso-propyl alcohol and tert-butyl alcohol), we can argue the advantages and disadvantages of using each of them as biocomponents. The interpretation of the experimental results also includes the effect of the bioalcohols over oxygenated gasoline behaviour in current vehicles, as well as some considerations relating to exhaust emissions.

The ethers synthesized from methanol (derived from natural gas) and C4 hydrocarbons, specifically MTBE or TAME, have practically demonstrated their benefits as octane additives for auto gasoline. Less studied has been their effect upon the volatility properties of gasoline blends. Over the last decade the interest in the two ethers, ETBE and TAEE has increased. Most importantly, they are considered as partially renewable, being synthesized from bioethanol and the C5 hydrocarbons fraction. Details regarding their impact over the octane and volatility properties of gasoline blends are less known. To compare, the effect of every type of ether on volatility and octane properties, an experimental was designed using MTBE, TAME, ETBE and TAEE. The experimental results highlight the advantages and disadvantages of every type of ether. Also, in the case of these oxygenated gasolines, the behaviour in vehicle engines is interpreted and assessments on the loss emissions by evaporation are made.

The first step of the experimental research was to prepare four conventional gasoline blends, noted 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), octane numbers, 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 5.

4.1 Bioalcohols or ethers gasoline mixtures volatility

The volatility properties of gasoline are very important characteristics which affects the vehicle's driveability. For any type of gasoline, these properties depend on the hydrocarbons classes, their concentration and distribution in gasoline. In the case of

oxygenated gasoline, Reid pressure vapour as well as the distillation curves are greatly influenced by the type and content of the oxygenated compound.

In order to demonstrate the way in which the oxygenated compound type affects the volatility properties of gasoline, we prepared conventional gasoline blends with primary alcohols and ethers, respectively. Out of gasoline A and B blends with 2, 4, 6, 7, 10 vol% of every alcohol (methanol, ethanol, iso-propyl alcohol and tert-butyl alcohol) were prepared. Out of gasoline C and D blends with 4, 7, 10, 15 vol% of every ether (MTBE, TAME, ETBE, TAAE) were prepared.

Properties	Values and UM				Lab Equipment
	Gasoline A	Gasoline B	Gasoline C	Gasoline D	
Hydrocarbons content:					IROX 2000 Fuel Analyzer Portable Gasoline Analysis with MID-FTIR
-aromatic	40.3% vol.	36.3 % vol.	38.1 % vol.	33.9 % vol.	
-olefinic	8.6 % vol	3.2 % vol	12.9 % vol	11.5 % vol	
-saturated	51.1 % vol	60.5 % vol	49.0 % vol	54.6 % vol	
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 % vol	25.2 % vol	33.0 % vol	35.0 % vol	
- E100	43.0 % vol	45.1 % vol	51.5 % vol	53.0 % vol	
- E150	74.5 % vol	75.4 % vol	78.0 % vol	78.0 % vol	
-FBP	198.3 °C	196.4 °C	190.4 °C	195 °C	
Octane numbers:					IROX 2000 Fuel Analyzer Portable Gasoline Analysis with MID-FTIR
-MON	85.0	85.7	85.0	85.0	
-RON	95.6	96.0	95.0	95.5	

Table 5. Conventional gasoline characterisation and experimental equipment

Each gasoline-alcohol or gasoline-ether blend was analysed from the point of view of the hydrocarbons content, the volatility properties, the octane numbers and the total amount of oxygen. In oxygenated gasoline, the oxygen content is determined with IROX 2000 Fuel Analyzer Portable Gasoline Analysis with MID-FTIR, as represented in Fig. 1 and Fig. 2. It is obvious the fact that, in the case of oxygenated gasoline, once the amount of alcohol or ether increases, the oxygen amount increases as well. EN 228 standard for commercial gasoline quality limits for now the total amount of oxygen to 2.7 weight%. According to this limit, we analysed the volatility properties of oxygenated gasoline with bioalcohols or ethers. When interpreting the experimental volatility properties, we took into consideration the criteria after which oxygenated gasoline are being framed in the specified EN228 standard volatility classes.

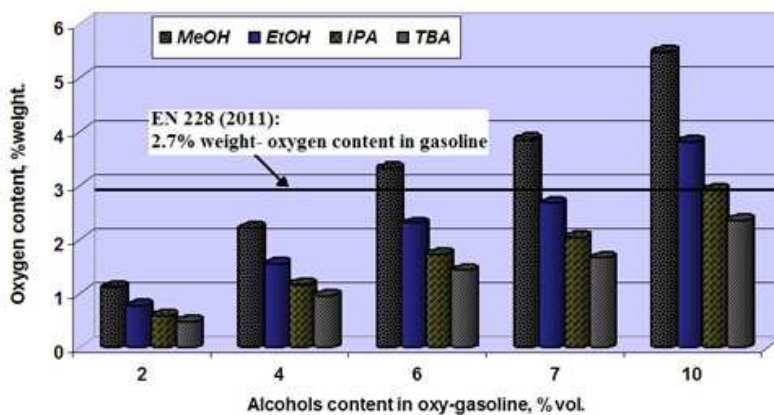


Fig. 1. The total oxygen content in oxygenated gasoline with primary alcohols

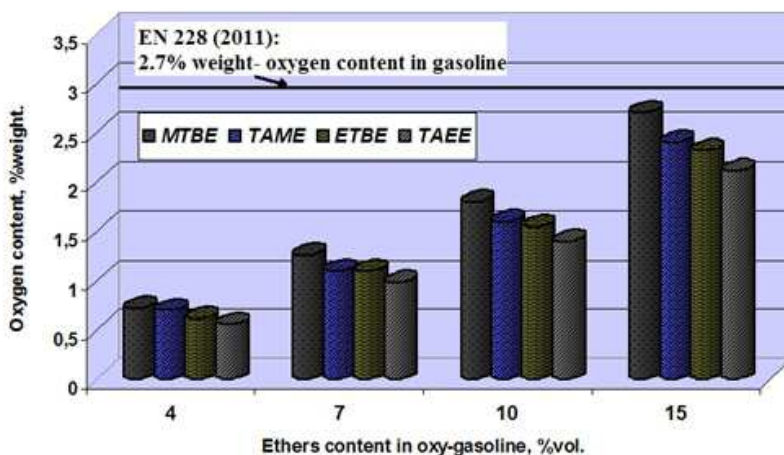


Fig. 2. The total oxygen content in oxygenated gasoline with ethers

The first volatility property analyses was the Reid pressure vapour of alcohol-gasoline and ether-gasoline blends. The experimental results are graphically represented according to the oxygen content of these blends (Figs. 3- 4). Reid pressure vapour for the two gasoline blends (A and B) with 2-10 vol.% primary alcohols are influenced by the vapour pressures of conventional gasoline, such as those of primary alcohols. Both conventional gasolines are a part of A volatility class taking into consideration the values of the vapour pressures and in accordance with EN228 European standard, whilst gasoline B is placed at the superior limit of this class (Table 5). By substituting conventional gasoline A with 4 vol% methanol or with 6 vol% 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 aprox. 8 vol% ethanol leads to obtaining a 2.7 weight% oxygen content in gasoline and a vapour pressure that makes it suitable in the cold season (Fig 3).

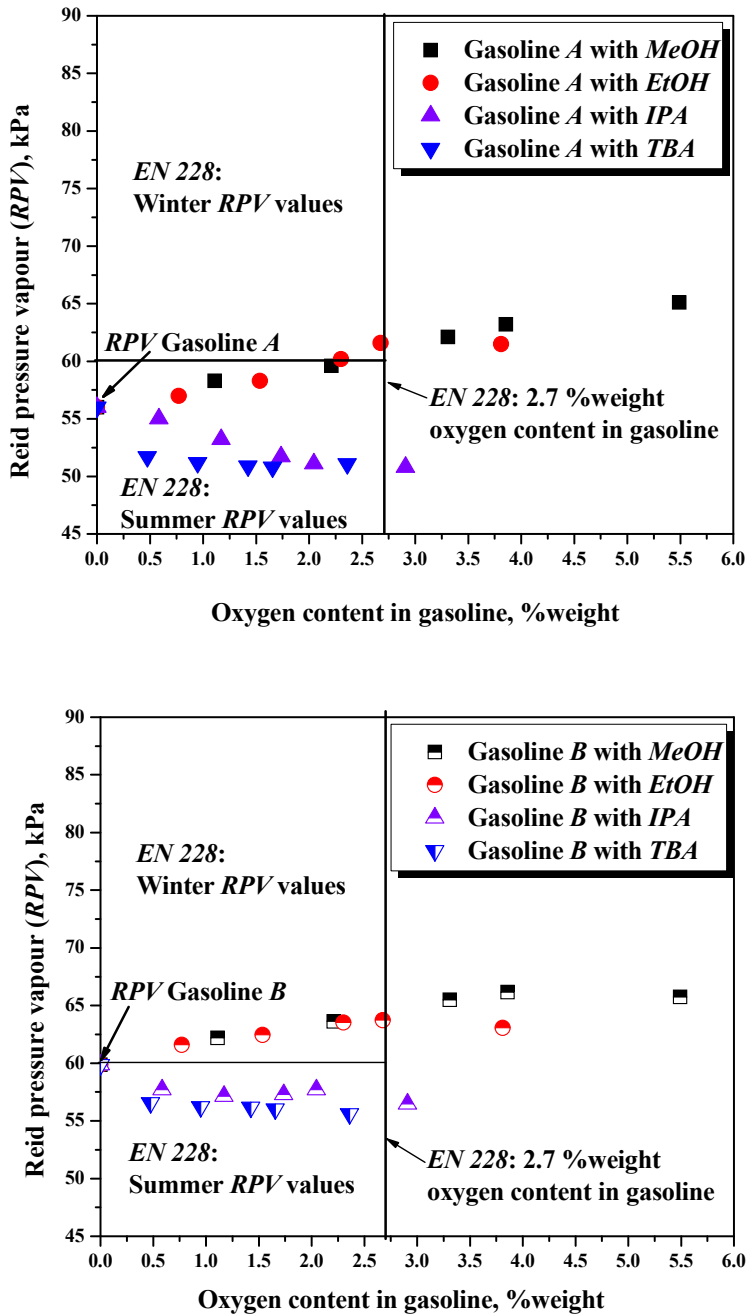


Fig. 3. Variations of Reid pressure vapour with the oxygen content of alcohol-gasoline blends

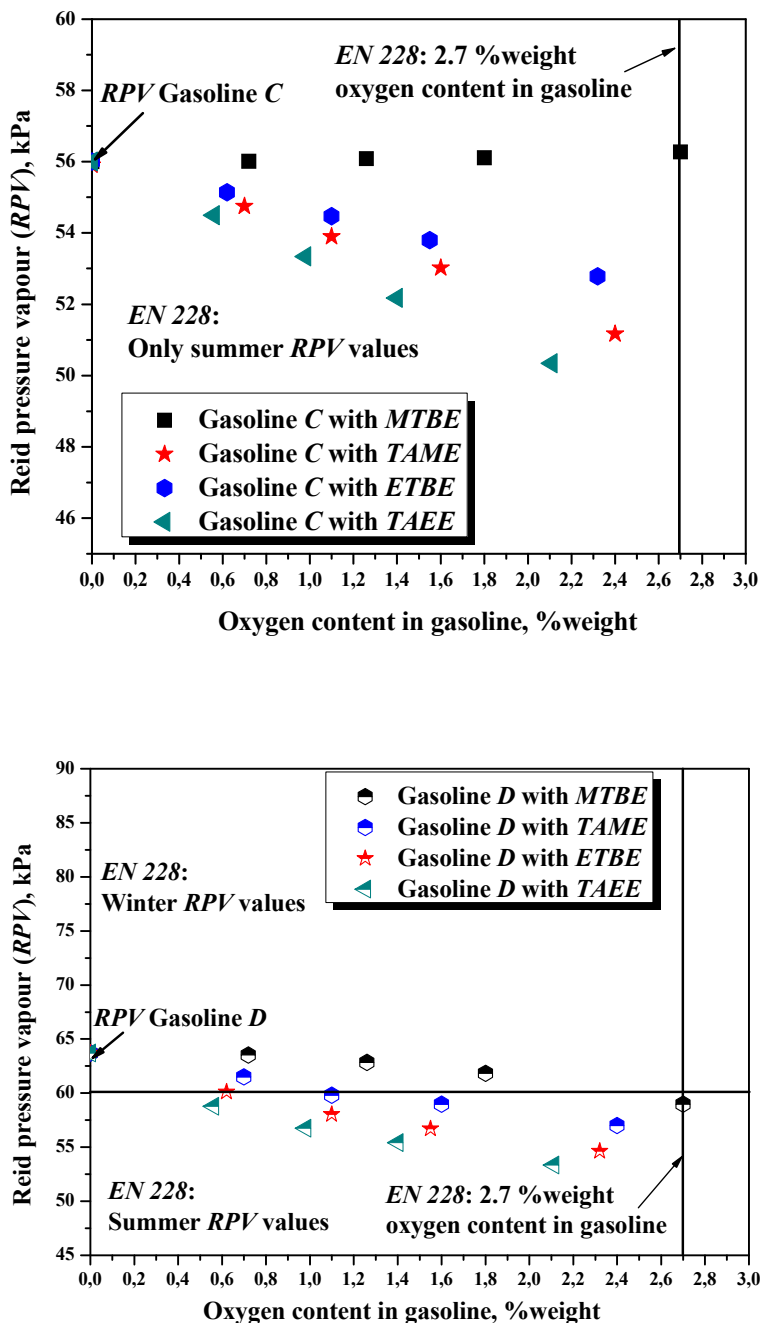


Fig. 4. Variations of Reid pressure vapour with the oxygen content of gasoline-ether blends

In contrast, by oxygenating gasoline B with methanol or ethanol (in any proportion, but without exceeding the maximum oxygen content), we only obtained gasoline with a vapour pressure over 60kPa, suitable for consumption in the cold season (Fig. 3). Therefore, in both cases, blending gasoline with methanol or ethanol causes an increase in the vapour pressure. This fact has 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 methanol or ethanol, formulations with the C4 fraction must be avoided, or dealt with cautiously. In the case of substituting conventional gasoline with IPA or TBA, regardless of the oxygenated compound content, the vapour pressure decreases. Nevertheless, gasoline-IPA or gasoline-TBA blends are in the same class volatility as with conventional gasoline. This fact is favourable because substituting conventional gasoline with different IPA or TBA proportions does not replace gasoline additives with a C4 fraction, as is desired by the refiners. Another positive aspect of IPA or TBA in oxygenated gasoline is that they can substitute large proportions of conventional gasoline without exceeding the maximum oxygen content allowed in the blends. Blends of 9-10 vol.% IPA or 10 vol.% TBA can be used in the case of gasoline such as A type, and 8-9 vol.% IPA or 10 vol.% TBA in the case of gasoline similar to B type (Neagu et al., 2010).

In experimental research of the volatility of blends of hydrocarbons with ethers, gasoline C and D were used. Gasoline type C is in A volatility class (for summertime) taking into consideration the vapour pressures, whereas gasoline type D is in B volatility class. For gasoline C with 4-15 vol.% MTBE, Reid pressure vapour increases with the ether content (Fig 4). This is a consequence of the fact that pure MTBE has a similar vapour pressure as gasoline C (Tables 3 and 5). Regardless of the MTBE content, gasoline C blends are situated in the A volatility class, just like conventional gasoline. In the case of gasoline D, oxygenated with MTBE, vapour pressure decreases, and at 15 vol.% MTBE, vapour pressures falls into A volatility class (Fig 4). As a consequence of the low pressures of pure ethers, vapour pressures for gasoline C or D oxygenated with TAME, ETBE and TAEE decrease. According to EN 228 standard, judging by the vapour pressure, gasoline oxygenated with ethers are placed in summertime classes. It is beneficial to note that through adding C4 hydrocarbons, their pressure can be increased up to suitable wintertime values. Such flexibility in using ether oxygenated gasoline, especially those with ETBE or TAEE favours them in commercial gasoline producers' options. These ethers have yet another advantage: in order to achieve a maximum content of oxygen in the gasoline, larger quantities than MTBE and even larger than for primary alcohols can be used (Neagu et al., 2011).

The distillation curves were determined experimentally for all oxygenated gasoline. Out of the large number of experimental results, distillation curves for gasoline A and B, non-oxygenated and oxygenated with 10 vol.% alcohol (Fig. 5) and for gasoline C and D, non-oxygenated and oxygenated with 15 vol.% ether (Fig. 6) are graphically represented.

The distillation curves of gasoline A and B are strongly influenced by the presence, in different proportions, of primary alcohols. This observation is exemplified, in the case of gasoline oxygenated with 10% vol. primary alcohols, in Fig. 5. In these curves three distinct regions can be identified, as in the case of conventional gasoline, but with different influences and names, like: azeotropes with different minimum boiling temperature regions, transition region and dilution region.

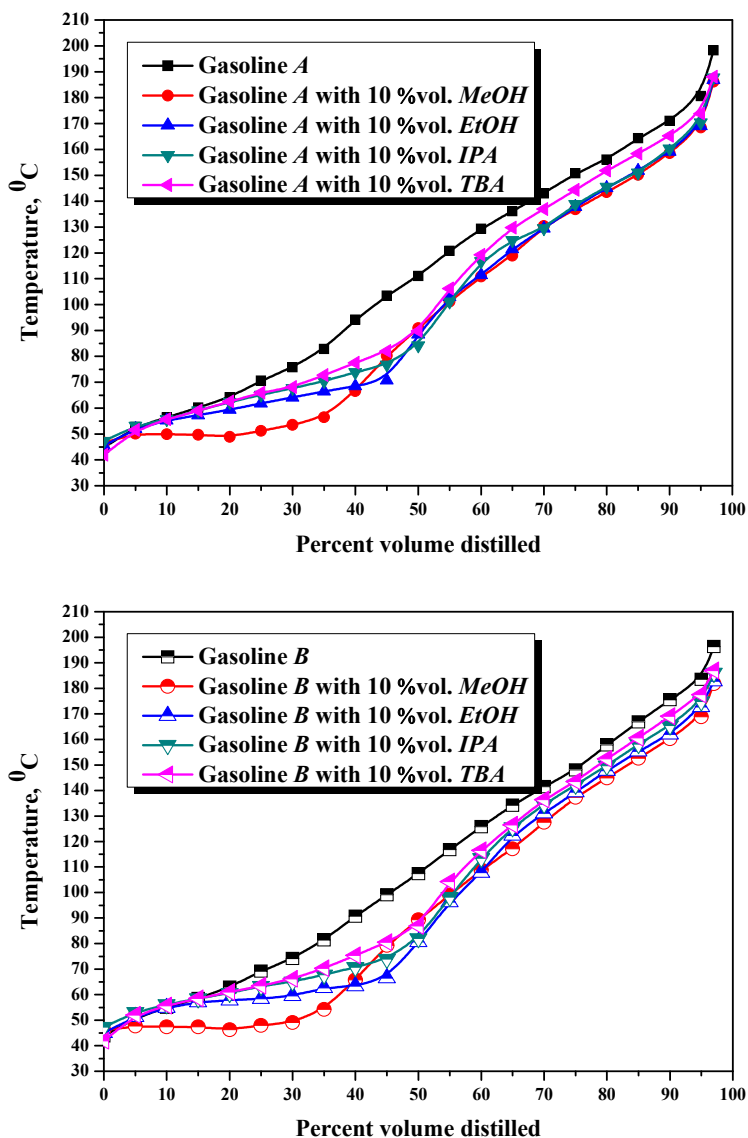


Fig. 5. The influence of alcohols over the distillation curves of oxy-gasoline A and B

The presence of azeotropes with minimum boiling temperature inside the distillation curves of gasoline-alcohol blends is explained by the fact that light hydrocarbons C5-C8 (alkanes, alkenes, aromatics) in the gasoline with polar compounds such as primary alcohols form azeotropes with a minimum boiling temperature. The decrease in the boiling temperatures of gasoline-alcohol blends 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.

Between the distillation curves of oxygenated gasoline with alcohols and of oxygenated gasoline with ethers there is a major difference. The ethers do not form azeotropes with the hydrocarbons in the gasoline; they only have an influence on the temperature at which the evaporation of hydrocarbons takes place. Hence, in the distillation curves of gasoline oxygenated with ethers only the boiling temperatures of the pure ethers and the chemical composition of conventional C and D gasoline are influenced.

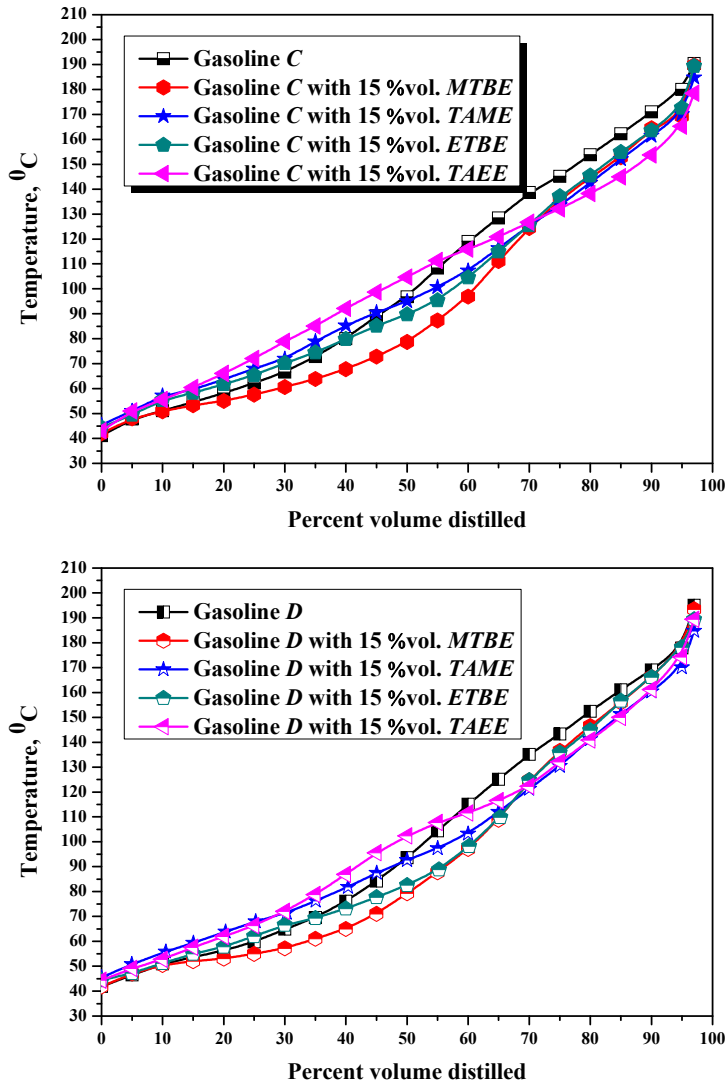


Fig. 6. The influence of ethers over the distillation curves of oxy-gasoline C and D

The difference between the behaviours of the alcohols and ethers towards the distillation curves is better highlighted by analyzing the respective values of the evaporated percentage of the fuel at certain temperatures: 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 will allow us to incorporate the oxygenated gasoline within the classes of volatility.

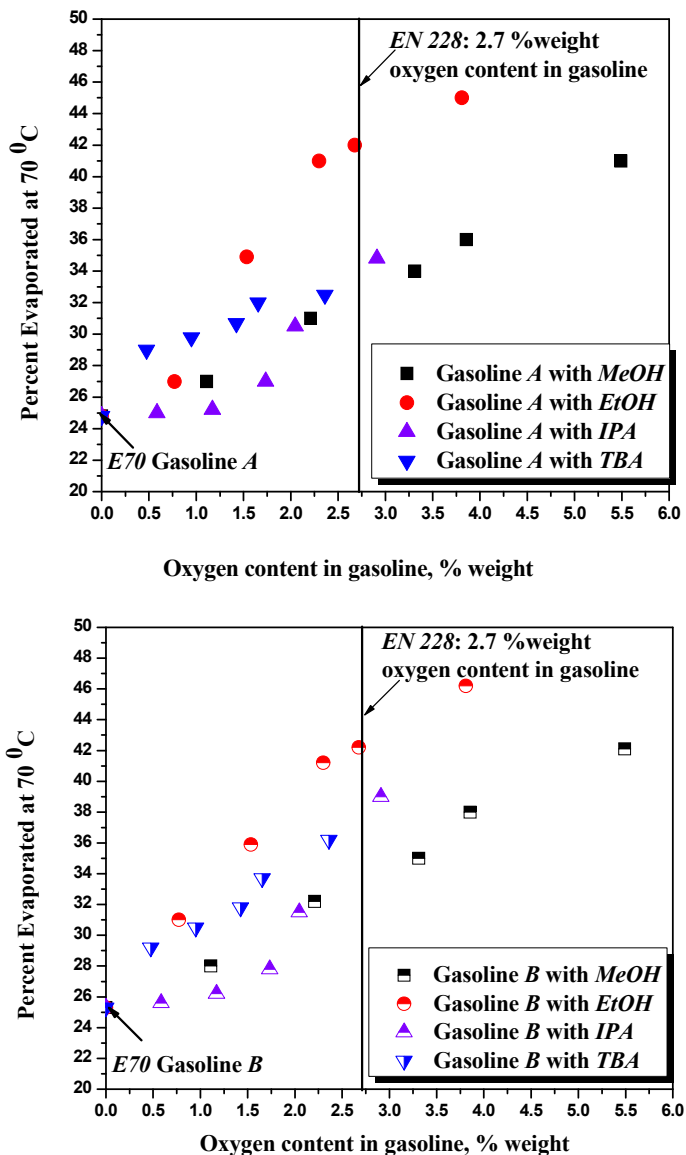


Fig. 7. The influence of alcohols over the percentage evaporated at 70°C of oxy-gasoline A and B

The type of alcohol strongly influences the E70 values of A or B gasoline mixtures (Fig. 7). The effect of the replacement of gasoline with ethanol on the E70 value is more obvious than in the cases in which gasoline is replaced with methanol. The explanation is that at 70°C the azeotropes with minimum boiling temperature of the light hydrocarbons from the ethanol gasoline evaporate. In the case of methanol, the problem of azeotropes evaporation at 70°C does not exist. But methanol, which has its boiling point at 64.7°C, favours a slight evaporation of the hydrocarbons that has boiling temperatures around 70°C. For IPA or TBA gasoline, the boiling point of the alcohols being the same (Table 2), the desired effect on the E70 value is due to the different oxygen content and chemical structures of these alcohols.

Furthermore, the type of the base gasoline influences the E70 values of the primary alcohols, as follows: for the same type and same content of alcohol, the E70 values for blends with B gasoline are higher than the ones for A gasoline because B gasoline has a higher content of saturated hydrocarbons, which influences the region of azeotropes with minimum boiling temperatures which it forms with each alcohol.

From the point of view of the values of the evaporated percentage at 70°C, all gasoline-alcohol blends are enclosed in volatility class A, i.e. in the domain of 20-48% vol. values, according to the gasoline quality standard EN 228.

In the case of the gasoline oxygenated with ethers, the evaporated percents at the temperature of 70°C increase in the presence of the MTBE and decrease in the presence of the TAME, ETBE or TAEE (Fig. 8). This behaviour is justified by the boiling temperature of ethers (Table 3). The introduction of MTBE, which has the lowest boiling point among the studied ethers (55°C), allows a light evaporation of his mixtures with hydrocarbons from the curves of the gasoline distillation region around 70°C. Instead, TAEE, which has the highest boiling point (101.42°C), like TAME, which has a boiling point at 86.3°C, will have as an effect a decrease in the hydrocarbons percentages that could evaporate at this temperature. Due to the fact that ETBE has a boiling point close to 70° C (72° C), the introduction of a high proportion of this ether (in present 15%vol., but will increase up to 22%vol. according to the Revised Fuel Quality Directive 2009/30/EC) has a minor effect on E70. From the point of view of the engine operation, the oxygenated gasoline with MTBE improved the cold ignition of the vehicle through the fact that it helps with the evaporation of the hydrocarbons from the start region of the distillation curve, but without increasing vapour lock. Instead, the gasoline oxygenated with TAEE or TAME, will evaporate harder, but the addition of a supplementary additive (C4+ light hydrocarbons) will correct this deficiency. Furthermore it was observed that gasoline oxygenated with ETBE, has a minimal influence on hydrocarbon evaporation in the start region of the distillation.

The presence of alcohols introduces a significant advantage by increasing evaporated percentages at 100°C, this signifies that the vehicle will offer an easier start-up and lower fuel consumption. Methanol, because it has the lowest boiling point among the primary alcohols, will favour less the evaporation of the gasoline hydrocarbons, at 100°C. Instead, ethanol (boiling point at 78.3°C), isopropyl alcohol and tert-butyl alcohol (boiling point 82.3°C) provide benefits in terms of hydrocarbons evaporation from the area of 100°C (Fig. 9). In the case of B oxygenated gasoline, the volatility of the base gasoline is a factor that favours hydrocarbon evaporation at a temperature of 100 °C. From the point of view of the

values of the evaporated percentage at the temperature of 100°C, all the gasoline-alcohols mixtures were found to be in the domain of the 46-71% vol., according to the gasoline quality standard EN 228.

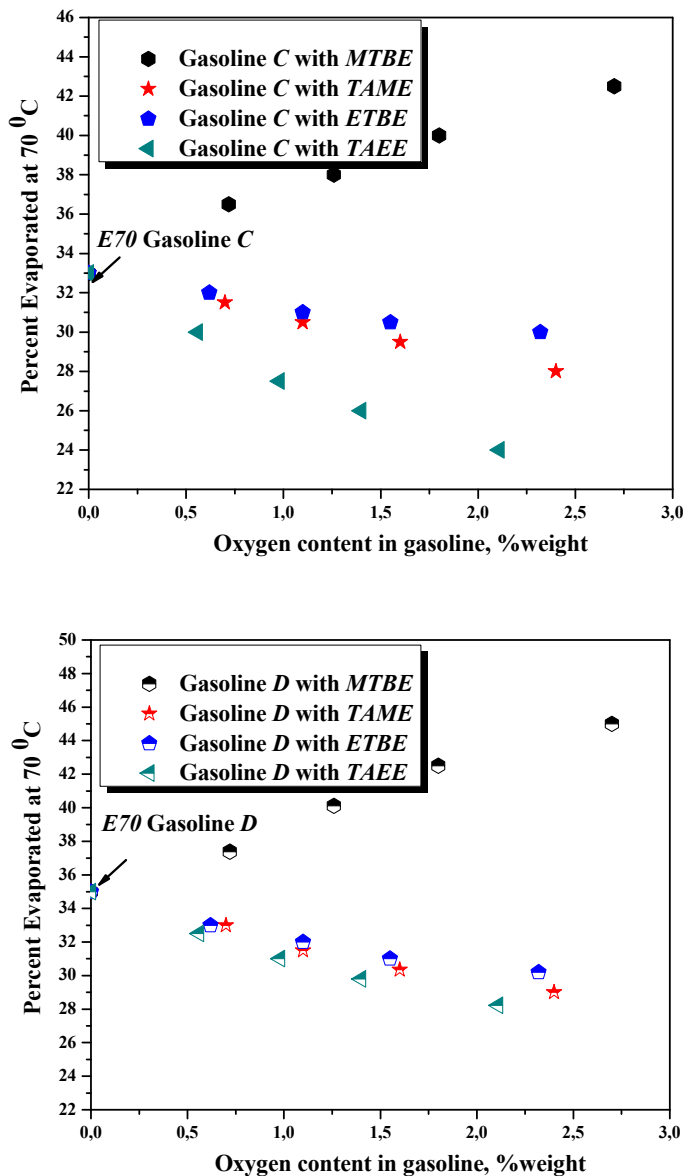


Fig. 8. The influence of ethers over the percentage evaporated at 70°C of oxy-gasoline C and D

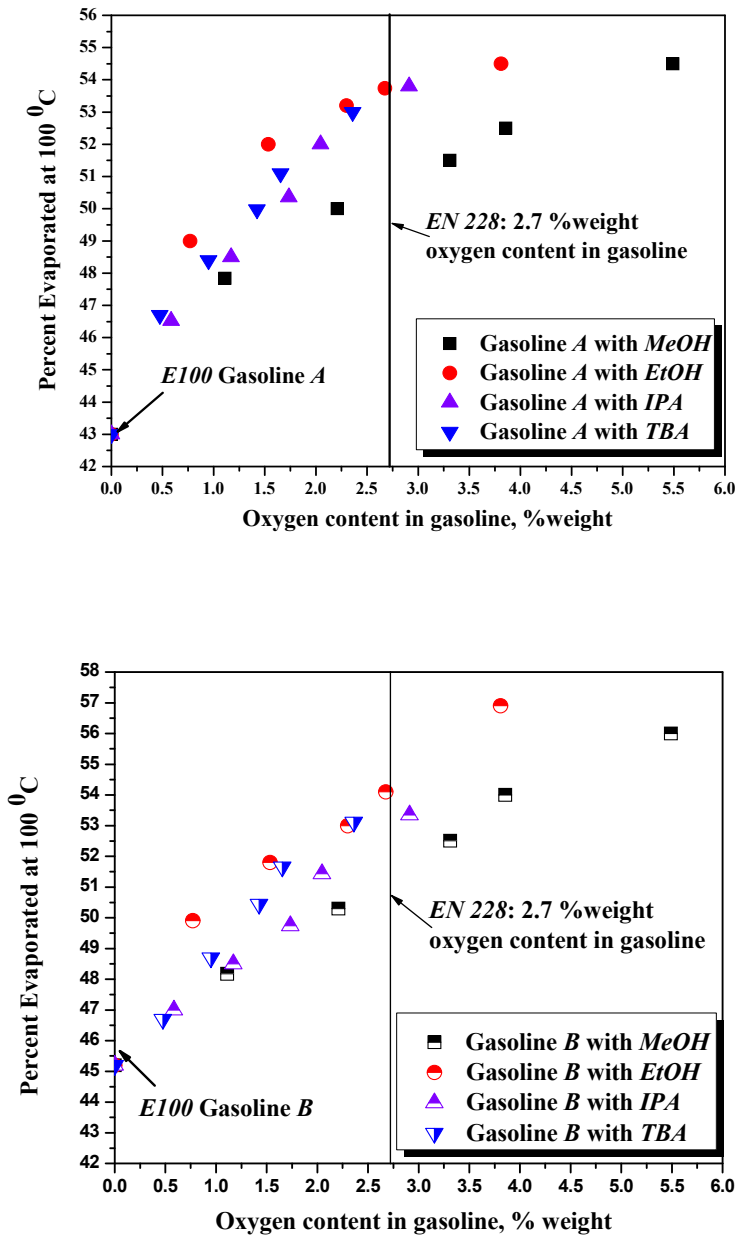


Fig. 9. The influence of alcohols over the percent evaporated at 100°C of oxy-gasoline A and B

The evaporated percents at the temperature of 100°C increase in the presence of ethers with the boiling point lower than 100°C, namely MTBE, ETBE or TAME (Fig. 10). The influence of these ethers on E100 is inversely proportional to their boiling temperatures. Instead, TAAE (the boiling point of the pure component is 101.42°C) influences in a negative way the evaporation at the temperature of 100°C. The negative effect is more marked at gasoline C, less volatile than gasoline D. The gasoline oxygenated with MTBE, ETBE or TAME, 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.

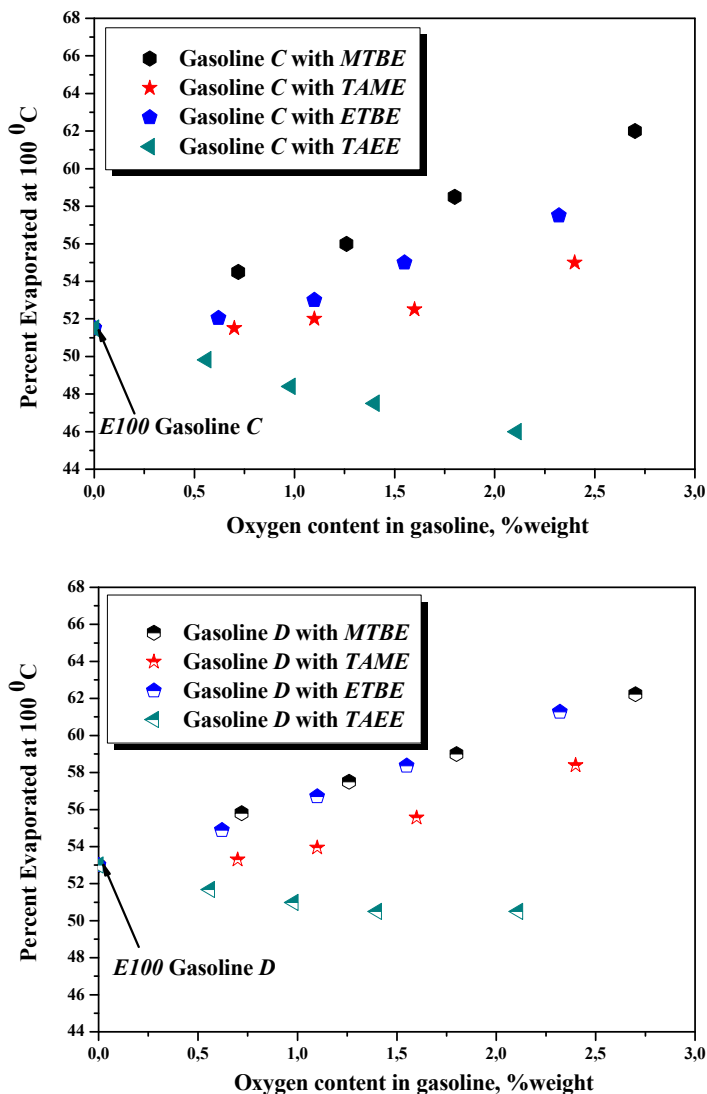


Fig. 10. The influence of ethers over the percent evaporated at 100°C of oxy-gasoline C and D.

Evaporation, at a temperature of 150°C, is favoured by the presence of primary alcohols, 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. The boiling temperatures of alcohols, and also their chemical structures, are the main factors that trigger the evaporation of hydrocarbons at 150°C. Adding methanol in fuels, less than 4% vol., has a rather smaller effect over the evaporation of the heavier hydrocarbons in the gasoline, which occurs at around 150°C. On the other hand, ethanol and IPA make a stronger contribution to

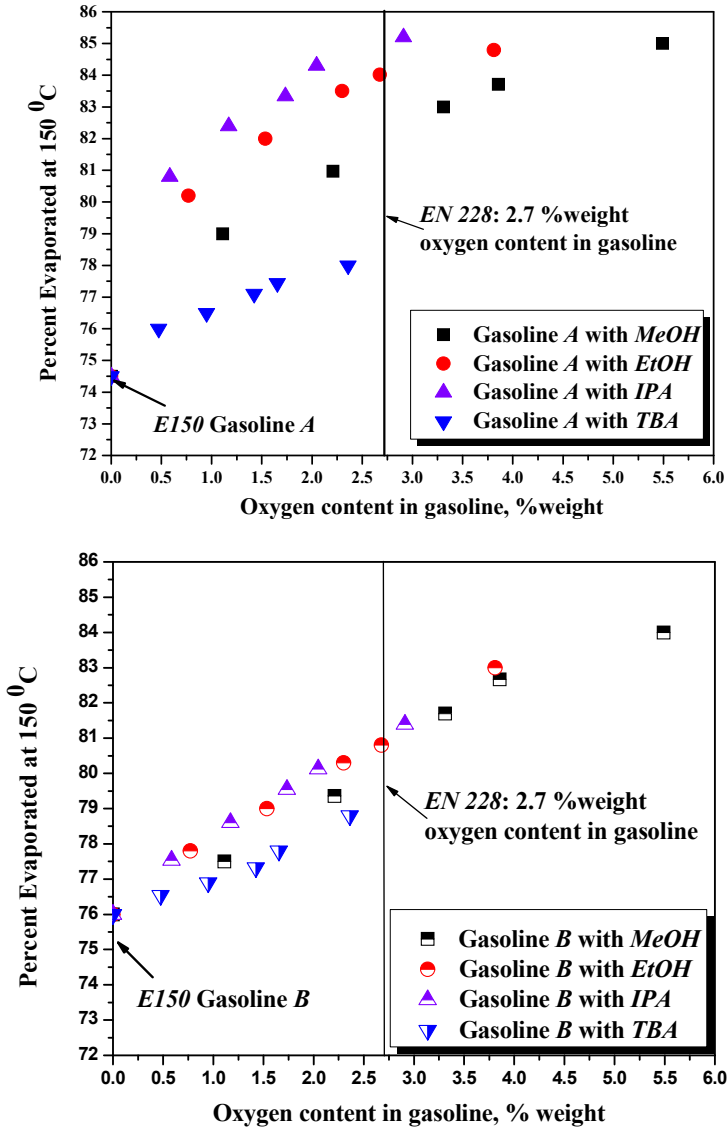


Fig. 11. The influence of alcohols over the percent evaporated at 150°C of oxy-gasoline A and B

the evaporation of the heavier hydrocarbons due to the fact that their boiling point is somewhat higher (Fig.11). One would have expected a similar effect from butanol also, but we believe that its effect is very much reduced because of its branched chemical structure. 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-alcohol blends is over 75.

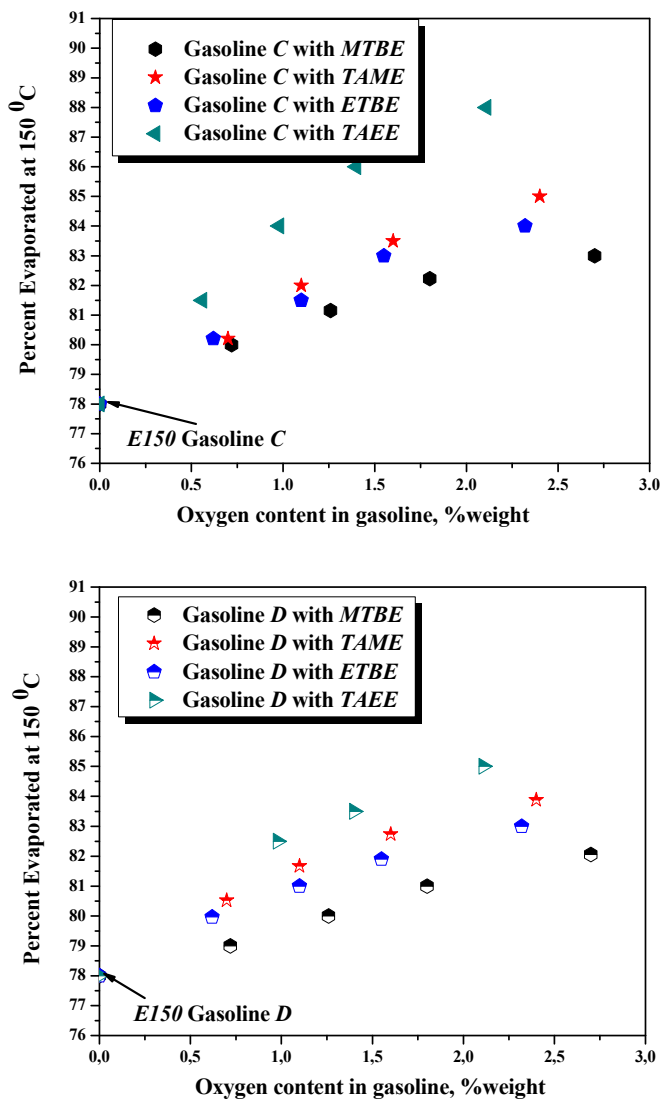


Fig. 12. The influence of ethers over the percent evaporated at 150°C of oxy-gasoline A and B

The percentages evaporated at a temperature of 150°C from oxy-gasoline with the ethers under study increase proportionally with each volume increase of each ether. The increase

in E150 is proportional with the boiling points of ethers. TAAE (with boiling point at 101.42°C) provides the best contribution to the evaporation of gasoline at a temperature of 150°C – the effect is more visible with gasoline C (less volatile), rather than with gasoline D. In the case of gasoline oxygenated with ethers, the percentage evaporated at a temperature of 150°C is over 75, according to standard EN228.

Another volatility property is the vapour lock index VLI. This property is calculated with Eq. (1), based on the value of the vapour pressure experimentally determined and the evaporated percentages at a temperature of 70°C (Figs. 13 and 14).

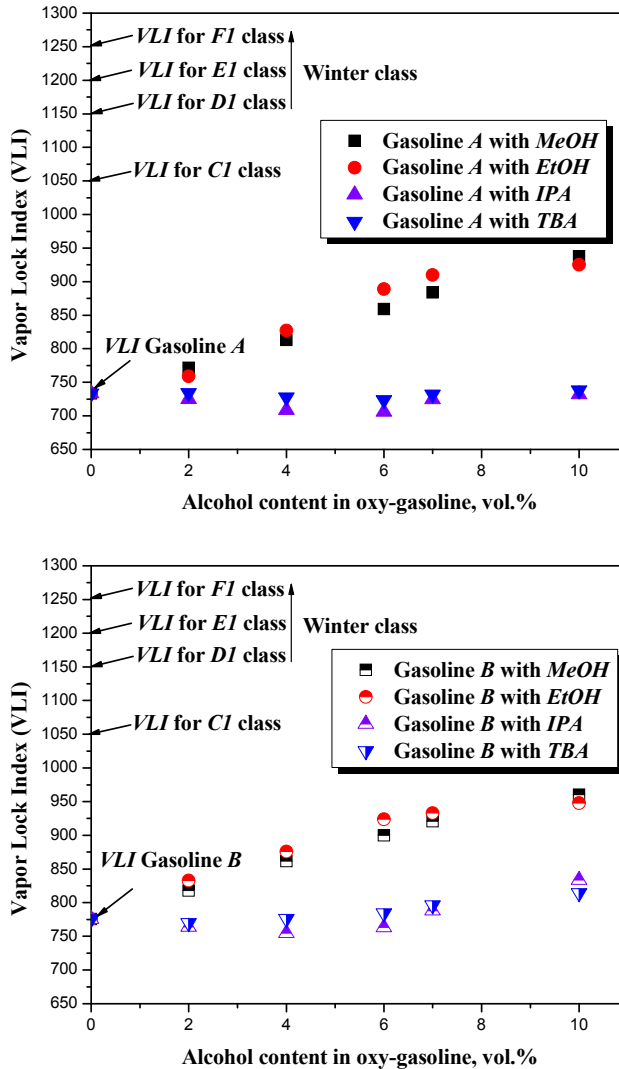


Fig. 13. Vapour lock index (VLI) of oxy- gasoline with alcohols

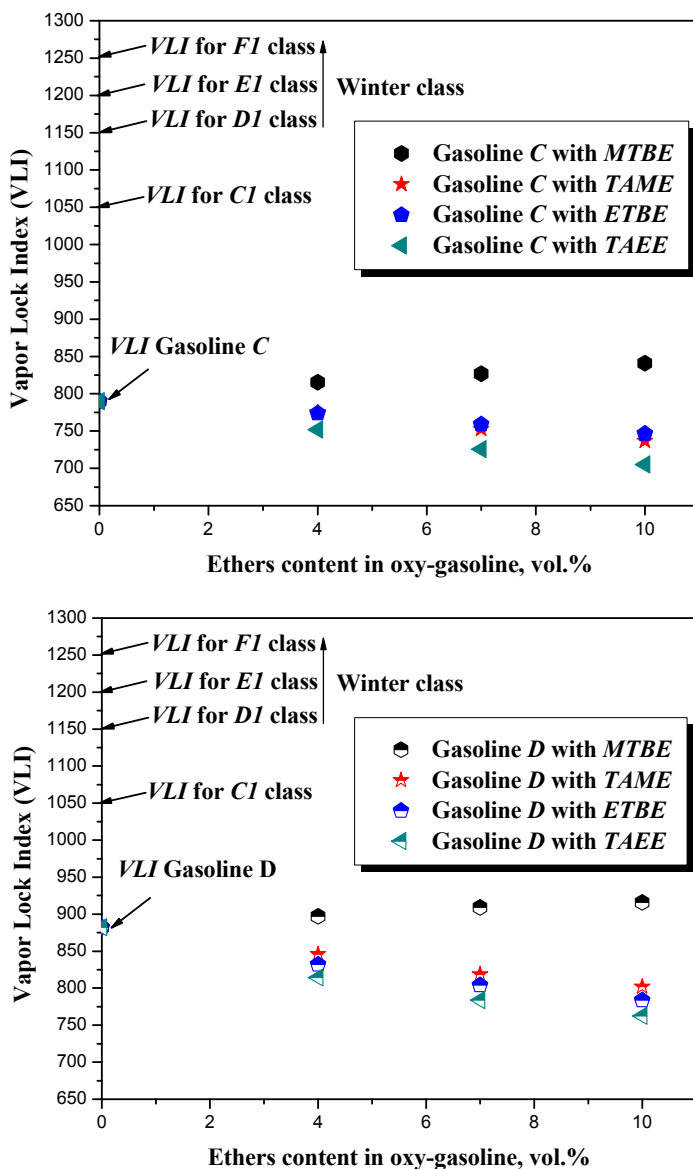


Fig. 14. Vapour lock index (VLI) of oxy-gasoline with ethers

From the point of view of the vapour lock index VLI, oxy-gasoline with alcohols, but also with MTBE, presents an increase in the tendency of forming vapour locks, unlike conventional gasoline. On the other hand, oxy-gasoline with TAME, TAEE or ETBE indicates a decrease in the vapour lock index, just as Reid pressure vapours and evaporated percentage at a temperature of 70°C decreased.

4.2 Octane properties of blends between gasoline and bio-alcohols or ethers

From the very beginning the introduction of a particular oxy-compound in commercial gasoline (1970 MTBE), was mainly to achieve an octane value increase. Both primary alcohols and all other ethers have an increased octane value, which may suggest that the effect of all oxy-compounds is that of having a positive in-take on octane value of blends. In order to sustain this idea, the engine octane value (MON) and Research octane value (RON) were experimentally determined using IROX 2000 Fuel Analyzer Portable Gasoline Analysis with MID-FTIR. The experimentally-determined results are correlated with the oxygen content in gasoline, as shown in the Tables 6 and 7.

Oxygen content in gasoline, % weight	Gasoline A with MeOH		Gasoline B with MeOH	
	MON	RON	MON	RON
1.11	0.32	0.83	0.39	0.74
2.21	0.85	1.50	0.77	1.48
3.31	1.12	2.34	1.16	2.22
3.86	1.45	2.62	1.36	2.62
5.49	1.88	3.59	1.79	3.43
Oxygen content in gasoline, % weight	Gasoline A with EtOH		Gasoline B with EtOH	
	MON	RON	MON	RON
0.77	0.34	0.71	0.29	0.66
1.40	0.62	1.27	0.73	1.32
2.30	0.98	1.92	0.98	1.98
2.68	1.19	2.38	1.15	2.33
3.81	1.70	3.23	1.51	3.06
Oxygen content in gasoline, % weight	Gasoline A with IPA		Gasoline B with IPA	
	MON	RON	MON	RON
0.59	0.18	0.49	0.14	0.38
1.17	0.46	0.81	0.39	0.74
1.74	0.78	1.34	0.63	1.26
2.05	0.86	1.44	0.73	1.50
2.91	1.30	2.28	1.12	2.07
Oxygen content in gasoline, % weight	Gasoline A with TBA		Gasoline B with TBA	
	MON	RON	MON	RON
0.48	0.16	0.25	0.10	0.20
0.95	0.40	0.42	0.37	0.40
1.43	0.53	0.64	0.56	0.60
1.66	0.70	0.73	0.74	0.79
2.36	0.95	0.96	0.93	1.00

Table 6. The effect of the type of alcohol and oxygen content on net increase of octane numbers in gasoline.

Table 6 shows that both motor octane numbers and research octane numbers increase with increasing alcohol content. The biggest increase is upon replacement of gasoline with methanol, followed by ethanol, isopropanol, tert-butyl alcohol. Net increase in octane number for gasoline-alcohol blends is smaller than with conventional gasoline of higher

octane number. For example: net the increase of the MON with gasoline A substituted by 10 % vol. IPA is 1.30 octane, while for gasoline B is 1.12 octane. Net increase of the RON with gasoline A substituted by 10% vol. IPA is 2.28 octane, while for gasoline B is 2.07 octane.

Oxygen content in gasoline, % weight	Gasoline C with MTBE		Gasoline D with MTBE	
	MON	RON	MON	RON
0.72	0.64	0.88	0.46	0.83
1.26	1.12	1.54	1.01	1.50
1.80	1.60	2.20	1.36	2.15
2.70	2.40	3.30	2.10	3.23
Oxygen content in gasoline, % weight	Gasoline C with TAME		Gasoline D with TAME	
	MON	RON	MON	RON
0.70	0.60	0.50	0.27	0.31
1.10	0.85	0.80	0.70	0.56
1.60	1.00	1.50	1.06	1.00
2.40	1.80	2.30	1.50	1.80
Oxygen content in gasoline, % weight	Gasoline C with ETBE		Gasoline D with ETBE	
	MON	RON	MON	RON
0.62	0.68	0.92	0.59	0.90
1.10	1.14	1.61	1.12	1.58
1.55	1.62	2.30	1.48	2.25
2.32	2.42	3.45	2.26	3.38
Oxygen content in gasoline, % weight	Gasoline C with TAEE		Gasoline D with TAEE	
	MON	RON	MON	RON
0.56	0.40	0.40	0.20	0.22
0.98	0.70	0.70	0.49	0.49
1.40	1.00	1.00	0.80	0.89
2.11	1.50	1.50	1.20	1.43

Table 7. The effect of the type of ether and oxygen content on net increase of octane values in gasoline

Both motor octane numbers and research octane numbers increase as the content of each ether increases (Table 7). Octane numbers vary as follows: ETBE>MTBE>TAME>TAEE. Net increase of octane numbers with gasoline-alcohol blends are smaller than with conventional gasoline of higher octane values and with bigger aromatics content (gasoline C). For example: net increase of MON with gasoline C substituted by 15 % vol. ETBE is 2.42 octane, while for gasoline D is 2.26 octane. Net increase of RON with gasoline C substituted by 15 % vol. ETBE is 3.45 octane, while for gasoline D is 3.38 octane.

Comparing the effects of alcohols and ethanol on octane values of oxy-gasoline, the conclusion is as follows: for motor octane numbers, it is more effective to use ether oxy-compounds, while for research octane numbers it is more effective to use ethanol, and also ETBE and MTBE.

5. Conclusion

In today's era, where various and major changes in climate emerge ever more often, and where humanity tends to offer more and more project-like solutions to using regenerable raw material, primary alcohols (with 1 or 4 carbons/molecule) and ethers (with 5 to 7 carbons/molecule) play a leading role. There are still a number of issues that need to be resolved before they will be commonly used as bio-components in gasoline for vehicles, which implies additional research and tests run on engines. Last but not least, the future of these bio-components depends essentially on legislative aid and the evolution of the automobile industry. Thus, the formulation of commercial gasoline quality to meet all quality prescriptions, and at the same time to contribute essentially to reducing air pollution, is a continuous challenge for all researchers and refiners.

A major effect on vehicle engines, and also on exhaust gas is played by the volatility and antiknock performances of oxy-gasoline. The present study is based on a very laborious experimental research design. We determined and analyzed in detail the volatility properties (Reid pressure vapour, distillation curves, and vapour lock index) and octane properties (motor octane number and research octane number) of two synthetic conventional gasoline oxygenated with 2-10% vol. primary alcohols (methanol, ethanol, isopropyl alcohol, tert-butyl alcohol). Also, we have determined and analyzed these properties for 2 other synthetic conventional gasoline oxygenated with 4-15% vol. ethers (MTBE, TAME, ETBE, TAEE). The present study contributes to highlighting the advantages and disadvantages of using oxy-compounds (primary alcohols or ethers, as partial substitutes for petroleum-based gasoline).

The main volatility property of oxy-gasoline is the Reid pressure vapour. From the point of view of the refiners, this property makes all the difference between summer gasoline and winter gasoline. From the point of view of the vapour pressure, oxy-gasoline with methanol or ethanol is recommended during winter, as the vapour pressure is higher. Conventional gasoline, with an initial vapour pressure of 60 kPa, oxygenated with MTBE, will have an even higher vapour pressure. In this case, this gasoline can be used efficiently in winter with no need to use additives with C4+ hydrocarbon fractions. On the other hand, oxy-gasoline with IPA, TBA, TAME, ETBE, TAEE, that have low vapour pressure, can be used in summer, but if a C4 hydrocarbons fraction is used, the gasoline can reach the vapour pressure required by the quality standard of winter gasoline. Such flexibility in using oxy-gasoline with ethers, and especially with ETBE or TAEE will offer a top position in any producer's choices for gasoline. Apparently, it seems that IPA and TBA alcohols are substitutes which can ensure, for the gasoline they are oxygenating, the proper flexibility from the point of view of volatility correction. Although these alcohols are obtained from renewable material, are not, YET, beneficial. For this reason, we consider that the possibility of their being used beneficially in the future is strictly dependant on developing future technologies that can produce them in sufficient quantities and at reasonable costs. Depending on the development of the production industry, isopropanol and tert-butyl alcohol (or other butanols) may be, for example, used in a blend with bio-ethanol in order to correct the deficiencies associated with high volatility of oxy-gasoline.

Gasoline distillation curves are modified when combined with oxy-compounds. With bioalcohols, distillation curves indicate an azeotrope region. In this region, evaporation of

hydrocarbons occurs much faster than with conventional gasoline. This is a beneficial aspect, as it aids the vehicle start up on ignition, even at cold temperatures. The disadvantage of this is that it increases the frequency of vapour plugs and evaporative emissions.

Ethers do not form azeotropes with gasoline hydrocarbons. Based on their boiling point, ethers influence hydrocarbon evaporation differently along distillation curves and vehicles operating in all gears. MTBE ensures evaporation along the whole distillation curve of oxy-gasoline, and especially volatile gasoline. ETBE, although it slightly affects evaporation at 70°C, has the advantage that it ensures evaporation for the rest of the distillation curve regardless of the gasoline type. TAME is not at all advantageous for evaporation at 70°C, but it is advantageous at 100°C. TAAE influences evaporation at 70°C and 100°C, but is far more advantageous for evaporation at 150°C.

Other volatility properties such as vapour lock index VLI offers an idea on the gasoline's tendency to form vapour plugs. In fact, VLI combines vapour pressure and a property for the beginning part of the distillation curve, E70. Unlike classic gasoline, oxy-gasoline with methanol or ethanol are more likely to form vapour plugs. This tendency is rather smaller with oxy-gasoline with IPA or TBA. Oxy-gasoline with ethers does not tend to form vapour plugs, and VLI values are rather smaller than VLI values for non oxy-gasoline (except those with MTBE). This behaviour is completely justifiable, as oxy-gasoline with TAME, ETBE or TAAE have E70 values and smaller vapour pressure than conventional gasoline.

In conclusion, oxy-gasoline with ethers displays more beneficial volatility properties than primary alcohols, especially than bio-ethanol.

From the point of view of antiknock properties, oxy-gasoline with ethers have higher octane numbers than alcohols. On the other hand, only MTBE and ETBE would lead to similar research octane numbers to those of oxy-gasoline with ethanol.

Ethers, specifically ETBE which is an oxy-compound partially renewable, have great potential to be used - in the future - as partial substitutes for gasoline. Alone, or combined with bio-ethanol, they enhance both volatility properties and octane numbers in gasoline for commercial use.

In the light of the information presented herein, we believe that we brought a significant contribution to the domain, clarifying a number of aspects associated with the current and potential usage of oxy-compounds in auto gasoline formulation.

6. Acknowledgement

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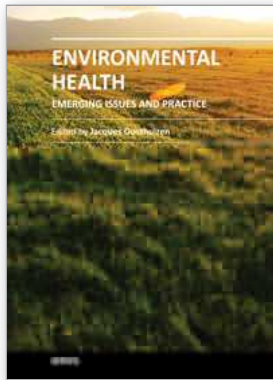
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Environmental health practitioners worldwide are frequently presented with issues that require further investigating and acting upon so that exposed populations can be protected from ill-health consequences. These environmental factors can be broadly classified according to their relation to air, water or food contamination. However, there are also work-related, occupational health exposures that need to be considered as a subset of this dynamic academic field. This book presents a review of the current practice and emerging research in the three broadly defined domains, but also provides reference for new emerging technologies, health effects associated with particular exposures and environmental justice issues. The contributing authors themselves display a range of backgrounds and they present a developing as well as a developed world perspective. This book will assist environmental health professionals to develop best practice protocols for monitoring a range of environmental exposure scenarios.

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