

Article

Strategies to Introduce n-Butanol in Gasoline Blends

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Abstract: The use of oxygenated fuels in spark ignition engines (SIEs) has gained increasing attention in the last few years, especially when coming from renewable sources, due to the shortage of fossil fuels and global warming concern. Currently, the main substitute of gasoline is ethanol, which helps to reduce CO and HC emissions but presents a series of drawbacks such as a low heating value and a high hygroscopic tendency, which cause higher fuel consumption and corrosion problems, respectively. This paper shows the most relevant properties when replacing ethanol by renewable n-butanol, which presents a higher heating value and a lower hygroscopic tendency compared to the former. The test matrix carried out for this experimental study includes, on the one hand, ethanol substitution by n-butanol in commercial blends and, on the other hand, either ethanol or gasoline substitution by n-butanol in E85 blends (85% ethanol-15% gasoline by volume). The results show that the substitution of n-butanol by ethanol presents a series of benefits such as a higher heating value and a greater interchangeability with gasoline compared to ethanol, which makes n-butanol a promising fuel for SIEs in commercial blends. However, the use of n-butanol in E85 blends substituting either gasoline or ethanol may cause cold-start problems due to the lower vapor pressure of n-butanol. For this reason, a combined substitution of n-butanol by both gasoline and ethanol is proposed so that n-butanol can be used without start problems.

Keywords: n-butanol; gasoline; E85; interchangeability; density; heating value; vapor pressure

1. Introduction

The gradual depletion of fossil fuels along with concern about global warming have led to the use of biofuels in internal combustion engines (ICEs) [1]. Among various biofuels, bio-alcohols have been investigated as alternative engine fuels because of their potential for improving engine performance and reducing pollutant emissions [2]. Bio-alcohols such as ethanol and butanol can reduce the life-cycle greenhouse emissions, due to their biological production processes. In fact, ethanol is normally used in spark ignition engines (SIEs) replacing gasoline [3–6]. Both of the aforementioned alcohols have also been used in compression ignition engine (CIEs) as a diesel partial substitute [7–10].

This paper focuses on the use of alcohols in SIEs, in which the most used bio-alcohol is ethanol, which is very common in many countries. The main advantage of this fuel is the reduction of CO and HC emissions when used in SIEs as a gasoline substitute [5,11]. Particularly, ethanol has been proved to reduce the extra emissions of CO and HC during cold-start conditions with respect to hot engine conditions [4]. In direct-injection SIEs, which have been gaining prominence in the last few years, ethanol has been shown to reduce NOx emissions slightly [3] and PM emissions considerably [6]. The lower heating value of ethanol is much lower than that of gasoline fuel, which causes higher fuel consumption. However, the thermal brake efficiency has been shown to slightly increase [12]. Ethanol could also present corrosion problems in the injection system due to its hygroscopic tendency. Ethanol also presents low lubricity [13], which can cause problems in gasoline direct-injection engines. Finally,

according to Rodríguez-Antón et al. [14], the addition of ethanol increases the vapor pressure of the blend only up to 35% by volume. A higher ethanol content would reduce the vapor pressure that leads to start problems [4].

n-butanol is considered the most promising bio-alcohol because of its numerous advantages over short-chain alcohols (methanol and ethanol mainly), including a higher energy density (higher lower heating values and higher density), lower viscosity, and higher lubricity. Additionally, n-butanol is much less hygroscopic and corrosive than ethanol. Besides, n-butanol seems to have a very interesting potential because its properties are very similar to those of gasoline; therefore, it would be compatible with the current fuel distribution infrastructure. n-butanol is a second-generation biofuel since it can be produced from lignocellulosic or waste biomass, with lower lifecycle greenhouse emissions than ethanol [15]. Currently, it can be produced through either an acetone–n-butanol–ethanol (ABE) fermentation process or an isopropanol–n-butanol–ethanol (IBE) fermentation process [16].

However, n-butanol has lower vapor pressure compared to ethanol and obviously compared to gasoline, which is a very evaporative fuel. This drawback could cause cold-start problems in SIEs when n-butanol is used in high proportions.

Some authors [3,17–19] have reported studies using gasoline-butanol blends in SIEs. Costagliola et al. [17] tested 10% n-butanol (as well as different ethanol contents) in a port-fuel injection (PFI) engine showing similar benefits in CO, HC, and PM emissions as with ethanol, but also similar increases in carbonyl emissions. Galloni et al. [18] tested gasoline and n-butanol (20% and 40% butanol mass percentage) also in PFI engine. When using a B40 blend, power delivered at same engine speed decreased about 13%, mainly due to the difference between heating values. HC and NOx emissions decreased slightly, whereas CO emissions did not suffer any significant change when using B40. Again in a PFI engine, Li et al. [19] also observed lower CO (4.2%), HC (18.9%), and NOx (5.5%) emissions when using B30 compared to gasoline as a baseline fuel keeping the engine load constant at 300 kPa of the brake mean effective pressure. Fournier et al. [3] tested gasoline blended with butanol, butanol–ethanol, and butanol–ethanol-acetone, with butanol contents up to 40% in volume, in both direct and port-fuel injection (PFI), showing that benefits in HC emissions in PFI engines turned into HC increases in direct-injection engines.

In this study, ethanol is replaced by n-butanol in commercial ethanol–gasoline blends, as well as in E85 blends (85% ethanol–15% gasoline by volume). Some properties of the blends were measured in order to compare the use of n-butanol instead of ethanol in commercial blends for their use in SIEs. The values of the different properties measured must be within the limits considered in EN 228 and EN 15376 standards. In summary, this paper discusses the advantages and drawbacks of using n-butanol instead of ethanol in different percentages and blends considering current European regulations. Vapor pressure, limited in both the E85 standard (CEN/TS 15293) and the gasoline standard (EN 228) was revealed as the most restrictive property. Within this European legal framework, the use of oxygenated compounds such as methanol, ethanol, isopropyl, ter-butyl, isobutyl alcohols, and ethers (with five or more carbon atoms) is explicitly considered. However, n-butanol should be included within the group of “other oxygenated compounds” despite having a similar octane number than gasoline, and closer polarity to that of gasoline, and thus greater miscibility than ethanol.

2. Experimental Procedure and Fuels

The whole test matrix (shown in Figure 1) carried out in this experimental study is divided into three submatrices (red, blue, and green dots) in order to study the most appropriate strategy for the use of n-butanol in commercial blends. First, commercial gasoline–ethanol blends in which ethanol (up to 10% by volume) was replaced by n-butanol (up to 16% by volume) were studied, keeping the oxygen content (limited by 3.7% by mass) constant, as limited by the EN 228 standard. Moreover, similar tests were carried out limiting the oxygen content to 6 and 7.4% by mass, considering possible future regulations. This submatrix is marked with red dots in Figure 1. Second, considering E85 as a baseline fuel (marked in yellow), ethanol was replaced by n-butanol up to 50% by volume of ethanol

(minimum value regulated by EN 15376), leading to a blend of 50% ethanol, 35% n-butanol, and 15% gasoline by volume. This submatrix is marked with blue dots in Figure 1. Third, considering again E85 as a baseline fuel, gasoline was progressively replaced by n-butanol up to 15% by volume, thus removing gasoline from the blend. This submatrix is marked with green dots in Figure 1. Finally, some extra dots were carried out for a more detailed study of vapor pressure, which will be explained in the corresponding section. These extra dots are marked in purple.

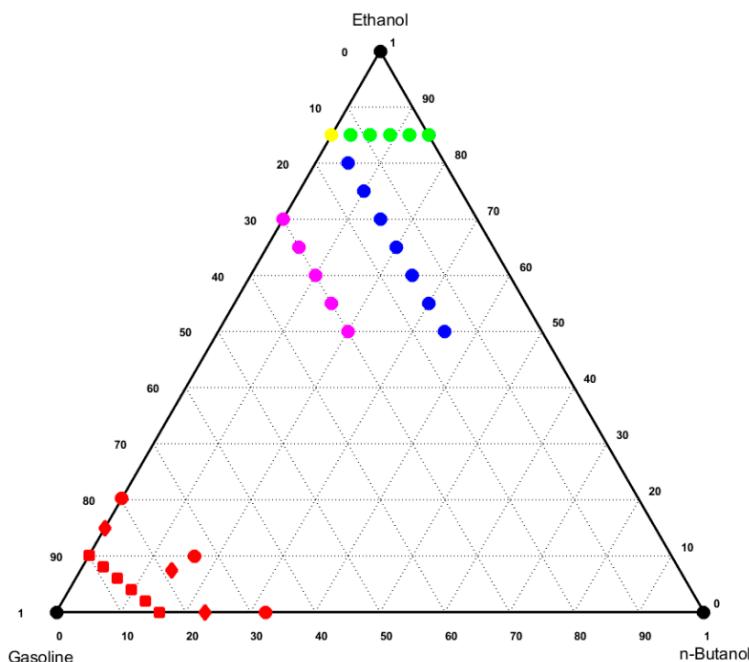


Figure 1. Test matrix carried out in the experimental study.

The main characteristics of pure alcohols (ethanol and n-butanol) and gasoline are shown in Table 1. These selected results are in agreement with those obtained in the literature. For instance, the lower heating value, the density, and the vapor pressure are very similar to those obtained by Elfasakhany [20] and Aleiferis et al. [21] for ethanol and n-butanol fuels. However, the vapor pressure values for gasolines are variable, since gasoline is a seasonal fuel, so its properties depend on the season of the year. The experimental procedure and the devices used in this study to measure the properties described in the results section are explained below. Density was measured following EN ISO 3675 standard at 15 °C using a densimeter. A higher heating value was measured with a bomb calorimeter (Parr 1351) according to UNE 51123 standard. Finally, vapor pressure was measured at a temperature of 38 °C with a commercial device (Eralytics, model Evarap EV01) according to EN 13016-1. Research Octane Number (RON) measurements for gasoline, E10 (10% ethanol by volume in gasoline blend), and Bu16 (16% n-butanol by volume in gasoline blend) were carried out in a CFR engine following the ASTM D 2699-15a standard. However, a CFR engine is out of range when operating either with pure alcohols such as ethanol or n-butanol or with blends with high alcohol content. For this reason, ethanol and n-butanol values were taken from the literature [18].

With regard to fuels, the gasoline used in this study was supplied by the Repsol company, and its main characteristic is the absence of oxygenated additives such as ETBE and MTBE in order to properly observe the effect of alcohol addition. Butanol was supplied by Green Biologics Ltd. (Milton Park, UK), as a member of the Consortium of Butanext Project (see Acknowledgments), and ethanol was supplied by PanReac ApplChem.

Table 1. Specifications of tested fuels.

Properties	Method	Gasoline ^a	Ethanol ^a	n-Butanol ^a
Purity (% v/v)		-	99.5	99.5
Density at 15 °C (kg/m ³)	EN ISO 3675	726.0	793.0	808.7
Viscosity at 40 °C (cSt)	EN ISO 3104	0.40–0.80	1.08	2.63
Higher heating value (MJ/kg)	UNE 51123	44.01	26.71	34.17
Higher heating value (MJ/L)		31.95	21.18	27.63
C (% wt)		86.00	52.14	64.86
H (% wt)		14.00	13.13	13.51
O (% wt)		0.00	34.73	21.62
Water solubility (ppm wt)	EN ISO 12937	<0.1 ^b	Fully miscible	<7.7 ^b
Standard enthalpy of vaporization (kJ/kg)		380–500 ^b	904 ^b	716 ^b
Stoichiometric fuel/air ratio		1/14.7	1/9.0	1/11.2
CFPP (°C)	EN116	-	<−51	<−51
Octane number	ASTM D-2699	95.8 ^c	107.0 ^d	96.0 ^d
Vapor pressure (kPa)	EN 13016-1	60	15	2
Flash point (°C)	ISO 2719	−45.00	13.00	36.17

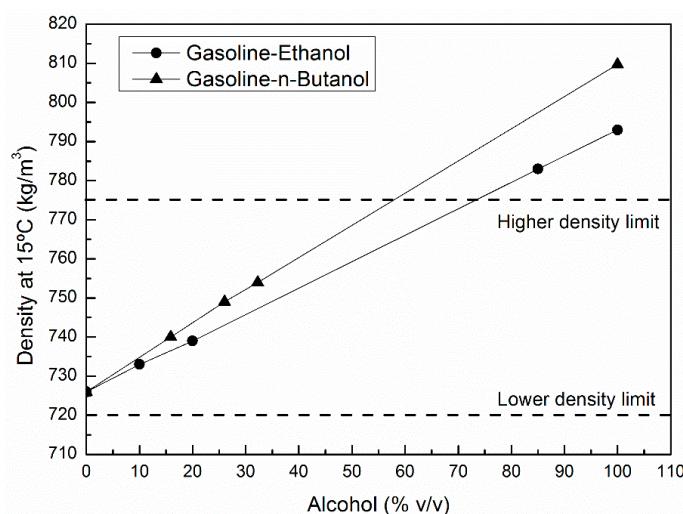
^a data measured at University of Castilla-La Mancha; ^b taken from reference [22]; ^c data provided by Repsol company, Spain; ^d taken from Reference [18].

3. Results and Discussion

3.1. Density and Lower Heating Value

The density of blends is usually given by a volume-weighted average of the densities of the components (all measured at the same temperature). However, some exceptions to this behavior, excess volume, can be found due to a large mismatch in the molecular/compositional structure of the components.

In this work, besides the test scheduled in the test matrices, some extra-blends were tested to evaluate possible non-lineal tendencies when small quantities of oxygenated fuels were added with respect to the original test matrix. As observed in Figure 2, n-butanol has a higher density than ethanol, and both alcohols have a higher density than gasoline. In European countries, gasolines are usually made to have densities close to the lower limit (upper and lower limits established in EN 228 standard are shown in Figure 2 with dashed lines) for two reasons: first, the strong dieselization of the vehicle market, leading to an extension of the diesel distillation range (and thus to an extension of the diesel density range); second, such a density allows for the further blending with oxygenates, which usually have higher densities. In any case, only lower butanol concentrations with respect to ethanol could be used in the blends to fulfill the above-mentioned standard.

**Figure 2.** Density versus alcohol content.

With regard to heating value, an almost linear trend with the alcohol content (Figure 3) can be observed when both ethanol and n-butanol are used. However, n-butanol has a higher heating value than ethanol in both mass and volume basis, as observed in Figure 3. For this reason, n-butanol blends are expected to reduce the fuel consumption in an SIE vehicle with respect to ethanol blends [3], and would therefore increase the vehicle's mileage. The effect of alcohol as a fuel component on the engine efficiency has been reported to be minor [3,17,20] and is not expected to modify this trend.

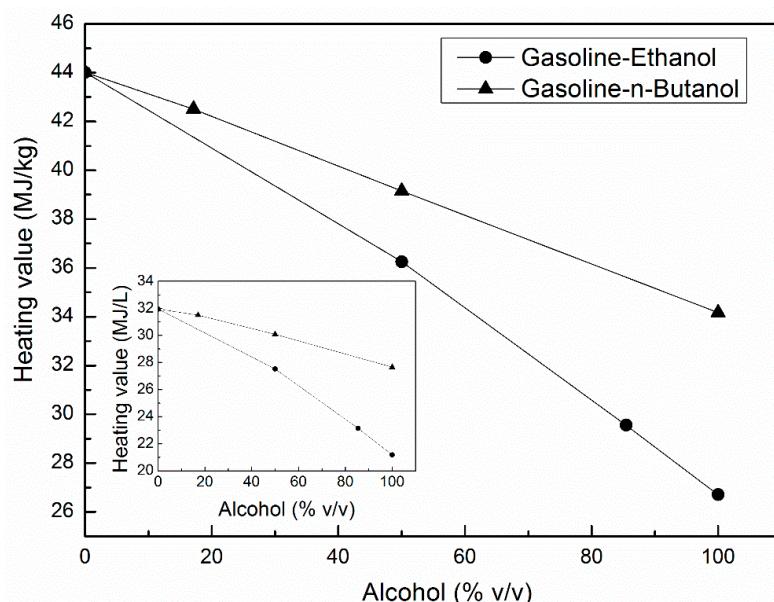


Figure 3. Heating value versus alcohol content in mass basis and volume basis.

As shown in Figure 4, when the ethanol content increases, the octane number of the blend also increases. This could derive into an advantage, either because the compression ratio of the engine could be increased in a modified engine design, leading to improved engine efficiency, or because additives improving the octane number could be avoided. Differently to ethanol, butanol presents a similar value of octane number compared to gasoline and, thus, modifying the butanol content in gasoline blends would not require re-design nor a reformulation of additives. Consequently, butanol, compared to ethanol, can be considered as more interchangeable with gasoline.

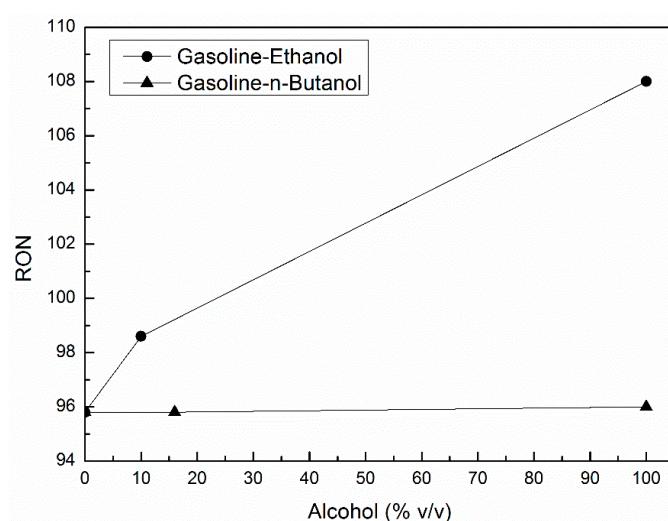


Figure 4. Octane number of ethanol and butanol blends with gasoline.

3.2. Vapor Pressure

With regard to vapor pressure, four standards were used for the vapor pressure tests. Specifically, two American standards (ASTM D5191 and ASTM D6378) and two European standards (EN 13016-1 and EN 13016-2) were followed. Standard EN 13016-1 was selected because gasoline standard EN 228 indicates that vapor pressure tests must be made at 37.8 °C. This standard allows for a vapor pressure increase when gasoline is replaced by ethanol up to 10% by volume as shown in Table 2. This increase is motivated by its azeotropic behavior.

With regard to Submatrix 1, a sharp decrease in vapor pressure is observed in Figure 5 when ethanol is replaced by n-butanol keeping the oxygen concentration constant. This decrease is observed for all oxygen limitations. This drop is due mainly to the lower vapor pressure of n-butanol compared to that of ethanol. It can be observed that such a decrease is more effective for lower oxygen content than for high oxygen content. Nevertheless, substantial decreases in vapor pressure would require high n-butanol contents (and thereby O₂ content).

Table 2. Vapor pressure overrun authorized by the EN 228 standard for different ethanol additions.

Ethanol Content (% v/v)	Authorized Vapor Pressure Overrun (kPa)
0	0
1	3.7
2	6
3	7.2
4	7.8
5	8
6	8
7	7.9
8	7.9
9	7.8
10	7.8

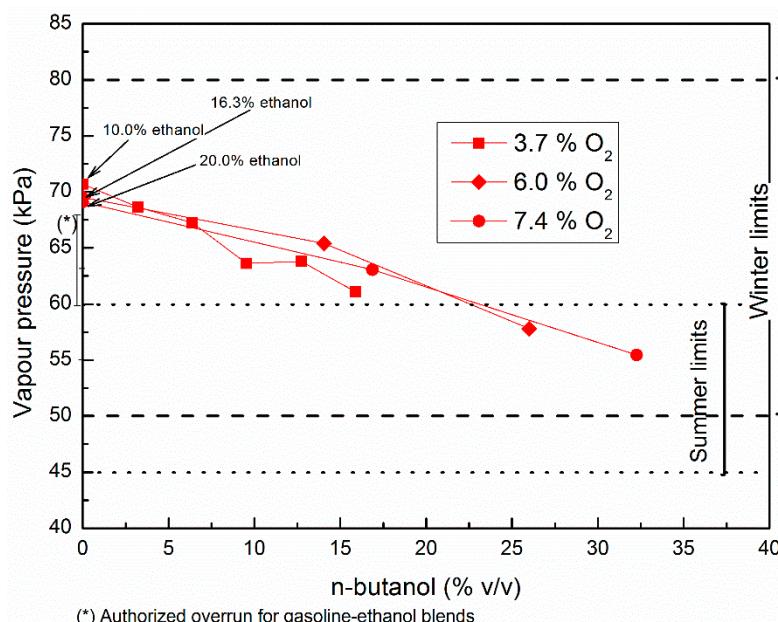


Figure 5. Vapor pressure versus percentage of n-butanol in Submatrix 1.

Following EN 228 standard, summer vapor limits were defined between 45 and 60 kPa, whereas winter limits were established between 50 and 80 kPa (shown in Figure 6 with a shaded area).

All the gasoline–ethanol–butanol blends tested in Submatrix 1 fulfill the vapor pressure limits established in the EN 228 standard for the winter season (as shown in Figures 5 and 6), so cold-start problems should not arise.

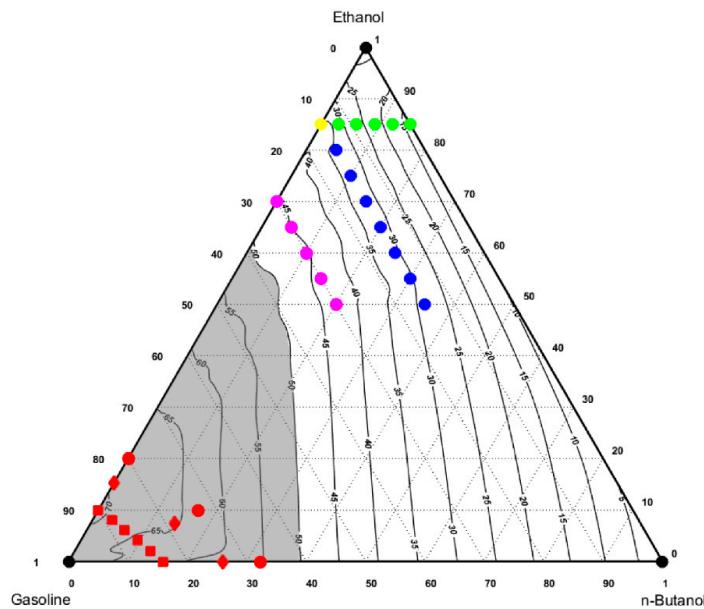


Figure 6. Vapor pressure range (shaded area) accepted by EN 228 in winter.

On the other hand, vapor pressure limitation is more restrictive in summer, as shown in Figures 5 and 7. None of the gasoline–ethanol blends would fulfill EN 228 requirements unless the vapor pressure overrun (shown in Table 2) is considered. On the contrary, gasoline–n-butanol blends fulfill the EN 228 standard for much lower alcohol concentrations compared to ethanol, so no overrun authorization would be needed. It is important to remark that the gasoline donated by the Repsol company is a fuel commercialized in winter and therefore not prepared for use in the summer.

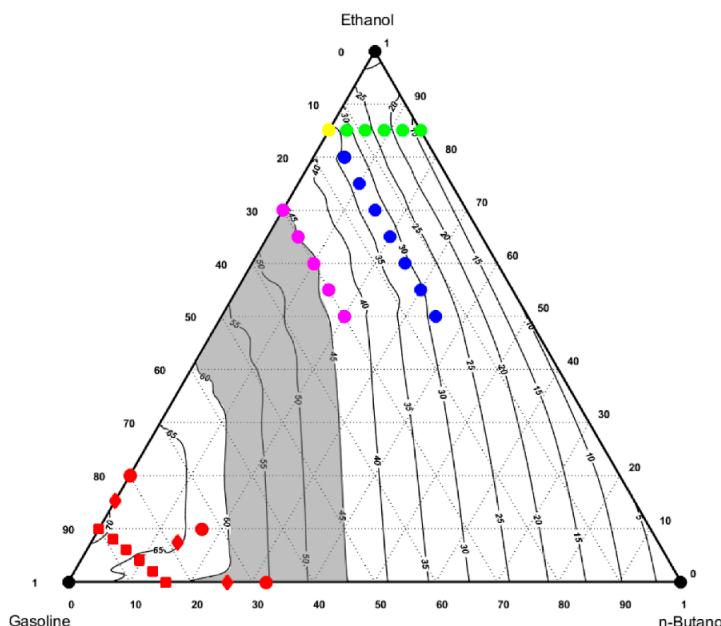


Figure 7. Vapor pressure range (shaded area) accepted by EN 228 in the summer.

With regard to E85 blends (Submatrices 2 and 3), when 15% (volume basis) of gasoline is kept constant (Figure 8, squared symbols and Figure 9, blue dots), and butanol content is increased to replace ethanol by up to 50% (volume basis) according to the EN 15376 limits, lower vapor pressure values are obtained. This means that replacing ethanol by n-butanol in E85 blends could cause cold-start problems despite the lower enthalpy of vaporization of butanol, and due to the lower vapor pressure of n-butanol [4,23–25]. When n-butanol substitutes gasoline (Figure 8, triangle symbols and Figure 9, green dots), cold-start problems are expected to be even more relevant, thus requiring a second fuel tank with a more volatile fuel for the start. This drawback is expected to be less relevant in direct-injection engines due to the increased cylinder pressure and temperature conditions. Indeed, as can be observed in Figure 9 (shaded area), the replacement of either gasoline or ethanol by n-butanol would move the blend out of the limits established by the EN 15376 standard. For this reason, some extra blends were tested using E70 (70% ethanol and 30% gasoline) as a baseline fuel. In this case, the replacement of ethanol (up to 50% by volume according to the EN 15376 standard limits) by butanol keeping the gasoline content constant allows for the fulfillment of the EN 15376 standard (Figure 9) despite the decrease in vapor pressure (circle symbols, Figure 8).

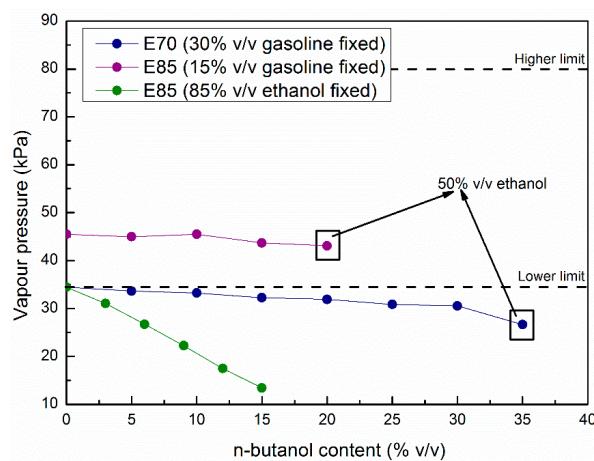


Figure 8. Vapor pressure versus percentage of n-butanol in Submatrices 2 and 3.

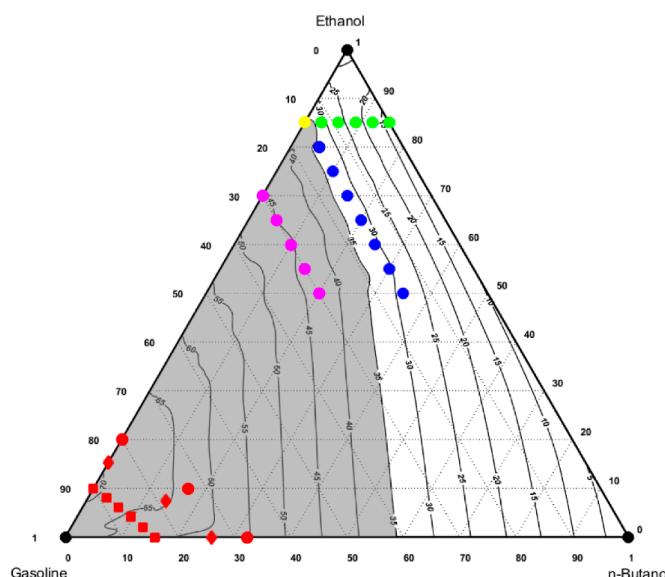


Figure 9. Vapor pressure range (shaded area) accepted by CEN/TS 15293/2011.

4. Conclusions

The main properties established in standards EN228 (for gasoline fuels) and CEN/TS 15293/2011 (for E85 blends) were measured for n-butanol–ethanol–gasoline blends and butanol–E85 blends. Fuels used in SIEs can contain up to 3.7% O₂ by mass to be commercialized. In this frame, the substitution of ethanol (up to 10% by volume) by n-butanol (up to 16% by volume) presents a series of benefits such as a higher heating value or a lower hygroscopic tendency compared to ethanol blends, which makes n-butanol a promising fuel for SIEs. The higher heating value for butanol–gasoline blends would reduce the engine fuel consumption. When butanol is blended with gasoline, the vapor pressure decreases, and consequently, the blends fulfill EN 228.

However, the use of n-butanol in E85 blends substituting either gasoline or ethanol can cause cold-start problems despite the lower enthalpy of vaporization of n-butanol. To avoid these problems, ternary blends where both gasoline and ethanol are simultaneously replaced with n-butanol should be used. As an example, if the baseline fuel is E70 (70% ethanol and 30% gasoline by volume), any replacement of ethanol by n-butanol would fulfill EN 15376, and cold-start problems should therefore not arise.

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Author Contributions: Magín Lapuerta conceptualized the study and organized the manuscript, Rosario Ballesteros conducted and processed the experiments and revised the laboratory conditions, Javier Barba processed the experiments and wrote the manuscript. All authors edited and approved the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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