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DOI: 10.1016/j.fuel.2017.07.022

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Document Version Peer reviewed version

Citation for published version (Harvard):

Hergueta Santos-Olmo, C, Bogarra Mácias, M, Tsolakis, A, Essa, K & Herreros, J 2017, 'Butanol-Gasoline Blend and Exhaust Gas Recirculation, Impact on GDI Engine Emissions', *Fuel*, vol. 208, pp. 662-672. https://doi.org/10.1016/j.fuel.2017.07.022

Link to publication on Research at Birmingham portal

Publisher Rights Statement: Published version https://doi.org/10.1016/j.fuel.2017.07.022

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Butanol-Gasoline Blend and Exhaust Gas Recirculation, Impact on GDI Engine Emissions.

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6 Abstract

A potential approach for addressing simultaneous reductions in toxic pollutants, greenhouse gas emissions and fuel consumption in gasoline direct injection (GDI) engines is the use of renewable alternative fuels. Furthermore, the combination of cleaner fuels with well-established technologies such as Exhaust Gas Recirculation (EGR) can reduce pollutant emissions and improve engine's efficiency.

12 In this research, the effect of 33% v/v of butanol in EN228 commercial gasoline containing 5% 13 of ethanol (B33) and gasoline (B0) fuels under maximum admissible EGR rate at two steady state 14 engine load (low and medium) conditions has been investigated. B33 reduces engine out 15 carbonaceous emissions, while maintaining similar levels of nitrogen oxide emissions when 16 compared to standard gasoline combustion. However, the physical and chemical properties of butanol 17 (i.e. viscosity and heat of vaporization) showed a negative impact on carbon monoxide emissions at 18 low load due to combustion inefficiencies. The addition of EGR showed a general reduction of 19 gaseous emissions and particulate matter (except unburned hydrocarbons), a trend that was more 20 significant for B33 at medium load. In addition, transmission electron microscope (TEM) analysis 21 showed that B33 is formed by more similar primary particles than primary particles formed with 22 gasoline fuel. From the engine point of view, EGR improved both Brake Specific Fuel Consumption 23 (BSFC) and Brake Thermal Efficiency (BTE) for the studied fuels with respect to baseline conditions.

24 Keywords: GDI engine, Butanol, EGR, gaseous emissions, particulate matter, TEM

Nomenclature	
aTDC	After Top Dead Centre
BSFC	Brake Specific Fuel consumption
bTDC	Before Top Dead Centre
ВТЕ	Brake Thermal Efficiency
CAD	Crank Angle Degree
COV of IMEP	Coefficient of Variation of Indicate Mean Effective Pressure

dp ₀	Primary particle diameter
ECU	Engine Control Unit
EGR	Exhaust Gas Recirculation
EVO	Exhaust Valve Opening
GDI	Gasoline Direct Injection
GNMD	Geometric Number Mean Diameter
нс	Hydrocarbons
IVO	Intake Valve Opening
MFB	Mass Fuel Burned
РАН	Polycyclic Aromatics Hydrocarbons
PM	Particulate Matter
TEM	Transmission Electron Microscopy
ТНС	Total Hydrocarbons
TWC	Three Ways Catalyst

27 **1. Introduction**

28 Gasoline Direct Injection (GDI) engines are fuel efficient and contribute to the reduction of 29 carbon dioxide (CO₂) when compared to port fuel injection engines. The reasons are increased 30 compression ratio by the charge cooling effect of the direct fuel injection, lower pumping losses, 31 higher volumetric efficiency and more accurate injection control [1]. GDI engines also reduce 32 pre-ignition and knock tendency as the compression temperatures are lower, and thus an enhancement 33 in thermal efficiency by a reduction of heat losses can be achieved [2]. On the other hand, GDI 34 engines have reported to increase the concentration of the Particulate Matter (PM) emissions [3, 4]. 35 The main sources of PM formation in GDI engines are identified as fuel piston wetting, injector fuel 36 deposits and inadequate air-fuel mixing. Consequently, the diffusive combustion of rich-in-fuel areas 37 promotes PM formation [5, 6], and also wall wetting by fuel impingement also produces an increment 38 of unburned hydrocarbons (HCs) and carbon monoxide (CO) due to a significant grade of incomplete 39 combustion [7, 8]. For this reason, emission standards such as Euro 6c, which includes a strict limit of $6x10^{11}$ particles per kilometer and comes into force in September 2017 [9], are boosting the 40 development of new technologies to reduce emissions in GDI. 41

42 A feasible short-to-midterm solution for addressing additional emissions reduction with a 43 decreased in the demand of high quality fossil fuels is to use renewable bio-alcohols fuels such as

44 butanol, which is considered a second generation of renewable transportation fuel [10]. Butanol 45 provides complementary physicochemical properties to gasoline blends for decreasing regulated 46 emissions as well as improving combustion. Amongst these properties, higher octane number and 47 oxygen content extend the knock limit for advanced spark timings [11] and improve combustion 48 efficiency, respectively, leading to further CO and total hydrocarbons (THCs) reductions [3, 12]. 49 Furthermore, butanol's higher latent heat of vaporization results in further cooling charge effect in 50 GDI engines, which increases the volumetric and thermal efficiency [13]. The higher latent heat of 51 vaporization combined with its lower adiabatic flame temperature can also assist in NOx reduction 52 [14, 15]. By contrast, studies have reported an increase of NOx with 35% butanol and advanced ST 53 [16, 17], or even insignificant differences in NOx emission [18]. Conversely, butanol's high latent 54 heat of vaporization and viscosity can also have a negative effect on engine operability, associated 55 with engine's cold-start and ignition problems [19]. The lower energy density of bio-alcohols (22% 56 lower butanol than for gasoline) leads to a penalty in fuel consumption and, in most cases, Brake 57 Specific Fuel Consumption (BSFC) increases [20]. Butanol can also be an approach in satisfying the 58 regulated PM levels in modern engines due to its oxygen content, inhibiting particle formation and 59 promoting oxidation rates [21]. In the literature, but nol blends have been reported to reduce large 60 particles (40-60nm), while the formation of small particles (30nm) is promoted [22]. Transmission 61 Electron Microscopy (TEM) has extensively been used for the analysis of the PM size, morphology 62 and nanostructure of soot particles, as these parameters are directly related to the formation process of 63 the particles [23]. However, there are still limited studies on the morphology of butanol-gasoline fuels. 64 This information can provide a guide of the soot oxidation rate, as it depends on the aggregate surface 65 area to volume ratio. It has been estimated that the aggregate surface area to volume ratio, is inversely 66 proportional to primary particle diameter [24]. Therefore, small primary particles will lead to high 67 aggregate surface area favoring soot oxidation [25].

EGR is a widespread technique firstly used in diesel engines to limit thermal NOx formation rate by reducing combustion temperature [26]. However, this technology is primarily implemented in GDI engines to obtain high engine efficiency and to improve fuel economy, since throttling losses can be reduced at low/part load range [27]. EGR increases the overall charge mass so the volumetric efficiency and the total heat capacity is relatively raised, leading to reduced pumping losses and lower temperature of the in-cylinder walls [28]. EGR has also shown beneficial effects on decreasing the severity of knock in GDI engines [22], enabling to advance the spark timing that improves the combustion phasing and therefore, increasing the engine work output and the thermal efficiency [29]. However, EGR addition slows down the combustion speed, leading to prolonged combustions that occurs over a greater proportion of engine cycle, thereby, worsening combustion stability [30].

78 In terms of emissions, EGR is reported to improve regulated emissions except for THC, mainly 79 due to the high EGR heat capacity that reduces the HC oxidation rate and may cause engine to misfire 80 when high EGR rates are used [29, 31]. The effect of EGR on particle emissions in GDI engines has 81 been also reported. Depending on the engine condition and EGR ratio, some authors have reported 82 that EGR addition increases the accumulation mode of particles and reduce nucleation mode 83 (medium loads) [32]. This effect was attributed to the lower in-cylinder temperature that reduced the 84 soot oxidation rate. This effect appeared to be more significant than the decreased in primary carbon 85 particles formed by thermal pyrolysis and dehydrogenation reaction of fuel vapor droplets, and thus, 86 the accumulation mode is increased. However, other authors have found the opposite effect, with 87 reduced accumulation mode and increased nucleation mode when EGR ratio was higher than 12% [22, 88 32]. It was attributed to the low in-cylinder temperature that limited the primary particle formation by 89 thermal pyrolysis and dehydrogenation. Additionally, the abruptly increased in HC, which are highly 90 related to nucleation mode, promoted further the increment of small particles and consequently, the 91 reduction in accumulation mode relatively to nucleation mode. Others have found reduction of 46% 92 and 90% in particle number (PN) and solid particle number, respectively, at high load and slightly 93 rich condition that simulate the transient engine conditions with cooled EGR [33].

Although, the use of alcohols for replacing gasoline in the GDI engines has been studied, there is still a need to better understand the behavior and the potential benefits of butanol fuel in these engines in terms of emissions reduction and combustion performance. The aim of this investigation is to assess and to further the understanding on the effect of the utilization of high butanol fraction blends on combustion characteristics, gaseous and particulate matter emissions in GDI engines. The potential benefits of high-diluted combustion process with EGR are also investigated in conjunction 100 with the B33 fuel. Additionally, the analysis of the primary particle diameters of the PM agglomerates

101 emitted for both fuels has also been investigated through transmission electron microscopy (TEM).

102 2. Materials and Methods

103 An 2L air-guided, four cylinders turbocharged GDI engine manufacturer by Ford has been used 104 for this study. The engine was coupled to a 75kW AC dynamometer and inverter drive capable of 105 motoring and absorption/regeneration. The specifications are depicted in Table 1 and the schematic 106 of the engine and experimental set up is shown in Figure 1.

107

Table 1: GDI engine specifications

Engine Specifications				
Compression Ratio	10:1			
Bore×Stroke	87.5×83.1 mm			
Turbocharger	Borg Warner k03			
Rated Power	149kW at 6000rpm			
Rated Torque	300Nm at 1750-4500rpm			
Engine Management	Bosch ME 17			

108 The engine was operated at stoichiometric conditions where the oxygen concentration was 109 controlled by a Heated Exhaust Gas Oxygen (HEGO) sensor. An AVL miniature piezo-electric 110 pressure transducer referenced to the engine cycle utilizing a Baumer 720 pulse per revolution 111 magnetic encoder was used for in-cylinder pressure measurements, considering an average of 200 112 cycles. The in-cylinder pressures were acquired using an in-house LabView application. Fuel 113 consumption was monitored with a Rheonik RM015 Coriolis fuel meter. The fuel supply temperature 114 control was provided by a fuel conditioning unit sourced from CP Engineering, and set to 28°C for the 115 test. The OEM's calibration strategy to reduce pumping losses was the utilization of valve 116 overlapping to increase the residuals. This technique is known as internal EGR, where the intake 117 valve opening (IVO) was set at 11 CAD bTDC (Crank Angle Degree before Top Dead Centre), and 118 the exhaust valve closing (EVO) is at 57 CAD aTDC (Crank Angle Degree after Top Dead Centre). 119 The engine is also equipped with high pressure external EGR system (designed and implemented by 120 the University of Birmingham). The external EGR valves were controlled using a standalone control

unit that requires a pulse-width modulated input signal to specify the desired valve position, providedby a custom LabView application.

A Fourier Transform Infrared Spectroscopy (FTIR) 2100 MKS was used to measure gaseous emissions (CO, CO₂, NOx and THC). The sample was previously filtered to avoid potential damage of the optical lenses by PM, and pumped via a heated line maintained at 190°C to prevent any water and HC condensation. The EGR ratio was determined using eq.1 by utilising an AVL Digas 440 non-dispersive infrared analyzer, to measure the CO₂ concentration at the intake and exhaust.

$$EGR \ Ratio = \frac{CO_{2(intake)}}{CO_{2(exhaust)}} \cdot 100 \ (\%) \tag{1}$$

128 Particle size distribution measurements were carried out using a TSI scanning mobility particle 129 size composed by a series 3080 electrostatic classifiers, a 3081 Differential Mobility Analyzer and a 130 3775 Condensation Particle Counter. The sample flow and the sheath flow were set to 1 and 10 lpm 131 respectively. The distribution ranged from 7.5 to 294 nm. The sampling point was located pre-three 132 way catalyst (TWC). To prevent HCs and water condensation, the line temperature was also 133 maintained at 190 °C during the test. The samples were then diluted with air at a dilution ratio of 7-8, 134 using an ejector diluter system, fitted with a high efficiency particulate arrestance filter to 135 precondition the sample.



Figure 1: Schematic of the engine and instrumentation set up

A custom post-processing script was developed in Matlab to import and time-align the channels of data obtained by the multiple data acquisition sources. The Matlab function enabled a detailed combustion process analysis by means of in-cylinder pressure measurements, including heat release rate, mass fraction burned (MFB), in-cylinder pressure, total cycle indicated mean effective pressure (IMEP) and coefficient of variation (COV) of IMEP (%).

For the PM primary particles analysis 3.05mm TAAB Formvar coated cooper grids were used to obtain the sample. The grids were directly exposed in the engine exhaust pipe before the three-way catalyst. A JEOL 1200EX TEM LaB6 80keV operating voltage was used to obtain the micrographs for primary particle analysis.

146 **3. Experimental Procedure**

147 The engine was operated at two steady-state conditions from the New European Driving Cycle 148 for a mid-size/large family vehicle with 2L engine under urban driving operation: a) 35Nm/2100rpm 149 (low load) and b) 60Nm/2100rpm (medium load). Butanol-gasoline fuel was blended at the 150 University of Birmingham using standard EN228 gasoline with 5% (v/v) ethanol content (B0) and 151 pure n-butanol was used in the process. The fuel characteristics are listed in Table 2.

152

Table 2: Fuels	properties	[34-36]
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Property	Gasoline	n-Butanol
Chemical formula	$C_{5.88}H_{11.06}O_{0.1}{}^a$	$C_4H_{10}O$
Density (kg/m ³)	743.9 ^a	811
Research octane number	96.8 ^a	96
Motor octane number	85.2 ^a	85
Latent heat of vaporization (kJ/kg)	350	722
Lower Heating Value LHV (kJ/kg)	42.2 ^a	33.1
Auto-ignition temperature (°C)	~300	385
Laminar flame speed (m/s)	51	58.5
Viscosity (mm ² /s) at 40°C	0.4-0.8	2.63
Adiabatic flame temperature (K)	2370	2340

153

^a Provided by Shell

154 The blend chosen was 33% v/v (B33) of butanol in EN228 commercial gasoline containing 5%

155 of ethanol. An IKA C200 calorimeter was used to measure the lower heating value of B33. The result

obtained was 40.175 MJ/kg. The test was repeated three times to ensure reproducibility and repeatability, with a maximum relative standard deviation of 0.1% between the measurements.

158 The spark timing was varied to phase the MFB50% to the one obtained for baseline gasoline for 159 Maximum Brake Torque (8± 1.5 CAD aTDC timing) for comparison purposes (Table 3). Camshaft 160 timings were fixed at short overlap setting to avoid the presence of residuals in the combustion 161 chamber (IVO 11 CAD bTDC and EVC 8 CAD aTDC). The effect of butanol on gasoline was studied 162 under the influence of external EGR. The conditions must satisfy the constrain of remaining COV of 163 IMEP below 3% at maximum EGR, which is a reasonable measurement of a modern engine stable 164 operation [35]. Prior to the test, the engine was warmed up, starting the measurement at 95 ± 0.5 °C for 165 coolant and 95 ± 2 °C for oil. The intake air temperature was maintained at 45 ± 1 °C throughout the 166 experiment to reduce the test-to-test variability. Otherwise, the standard Engine Control Unit (ECU) 167 calibration settings were used such as injection timing. Confidence intervals using a 95% confidence 168 level, which reflects the reliability and repeatability, have been calculated for gaseous emissions. All 169 experiments were conducted on the same day for each condition to reduce the effect of day-to-day 170 variability of the engine and emissions equipment on the results obtained.

171

Table 3: E	ngine conditions	and ECU settings
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			Ignition Timing		Spark Timing	MFB50%
			CAD bTDC	EGK (%)	CAD bTDC	CAD aTDC
	BÜ	Baseline	304.5	0	30.75	9.4
35Nm/2100rpm	D 0	Max %EGR	304.5	17	+49.5a	9.12
	B33	Baseline	304.5	0	33	9.14
		Max %EGR	304.5	17	+50.25a	8.38
	В0 В33	Baseline	303.7	0	30	7.7
60Nm/2100rnm		Max %EGR	303.7	19	+49.5a	7.04
001(11)/21001 pill		Baseline	303.7	0	29.7	6.9
		Max %EGR	303 7	19	+50 75a	79

172

+ advance with respect to ECU settings

The study of the primary particle diameters (dp_0) for B33 and gasoline fuels where carried out at 60Nm/2100rpm. For the analysis of dp₀, manual measurements were performed, although it was only measured the apparently identifiable primary particles were measured. The recognition of the primary particles boundaries is somewhat subjective, and can lead to some fluctuations in the analysis of the results [37]. Additionally, the sample has to be large enough to ensure its log-normality distribution, as Gaddam et al. [38] have discussed. At least 240 primary particles were considered per fuel, located
in 17 different agglomerates randomly chosen in the TEM grids. One-sample non-parametric
Kolmogorov Smirnov test in the IBM statistical package for social sciences software (SPSS) was
performance to check the log-normality.

182 **4. Results**

183 *4.1 Combustion studies*

184 The in-cylinder pressure and heat release rate for B33 are overlapped with respect to those 185 obtained under gasoline fueling at 35Nm (Figure 2 (a)). In addition, the MFB50% timing was kept 186 identical (Figure 3 (a)) by slightly advancing the spark timing (Table 3), so comparisons of the two 187 fuels engine-out emissions can be made based on their chemistry. It has been previously reported that 188 the higher flame speed of butanol promotes stable combustion and improves the degree of constant 189 volume heat release [34]. However, at this engine load, it seems that the fuel physical properties (i.e. 190 viscosity and enthalpy of vaporization) dominate over the faster laminar burning velocity of butanol 191 (Table 2) resulting in advanced spark timing (2.25° CAD) to achieve the same MFB50% (Figure 3 (a)) 192 and slightly worse combustion stability as COV of IMEP indicates (Figure 4). It is thought that the 193 high viscosity of B33 impacts on the fuel spray atomization process, promoting more 194 in-homogeneous mixture and hence, it reduces combustion stability.

195 For the medium engine load of 60 Nm, the in-cylinder peak pressure and heat release rate of B33 196 were slightly higher compared to gasoline, which is in agreement with the results reported earlier in 197 literature [12, 22, 36, 39], at a comparable combustion phasing (Figure 3 (a)). The higher fuel spray 198 velocities, resulting from the increased engine load at 60 Nm, created a turbulent motion inside the 199 cylinder that promoted the homogeneity of the air-fuel mixture [39]. At this engine load, the higher 200 laminar burning velocity of butanol combined with an enhancement in fuel spray atomization provide 201 greater combustion quality when compared to gasoline, as observed by the lower COV of IMEP 202 (Figure 4).





Figure 2: (a) In-cylinder pressure and (b) heat release rate versus CAD for gasoline (B0) and B33 at 2100rpm





Figure 3: MFB for Gasoline and B33: (a) Baseline and (b) Max EGR at 2100rpm



The introduction of maximum applicable EGR rate (%) in B33 fueling at low and medium engine load slightly reduced the in-cylinder pressure and heat release with respect to maximum EGR with gasoline fuel (Figure 5). The B33 higher cooling charge capability together with the EGR negatively affected the fuel vaporisation of B33, and consequently the mixture homogeneity as reflected in the COV of IMEP (Figure 4), which leads to marginal peak reduction in both, the in-cylinder pressure and heat release rates.



Figure 5: (a) In-cylinder pressure and b) heat release rate versus CAD for Gasoline and B33 at maximum EGR
 and 2100rpm

215 Figure 6 (a) shows that the EGR enhances the BSFC for both fuels, in agreement with the 216 literature [8, 32]. When EGR is used, the throttle valve must be further opened in order to maintain the 217 oxygen level at stoichiometric ratio and therefore, the pumping losses are reduced. In all cases, the 218 BSFC of B33 combustion was higher in comparison to gasoline combustion. The brake thermal 219 efficiency, which is inversely proportional to the BSFC, and considers the lower heating value of the 220 fuel blends is shown in Figure 6 (b). There are no significant differences in the indicated thermal 221 efficiency which concludes that the higher BSFC in the case of the B33 blend is due to the lower 222 energy density of butanol compare to gasoline [36].

As it can be observed from the reduction of the exhaust gas temperatures (Figure 7), EGR brings the in-cylinder temperature down, enabling a reduction of the heat losses to the coolant and surroundings, and contributing in further improvements of the brake thermal efficiency (BTE) [22,
32]. Thus, EGR enhances BTE (Figure 6 (b)) for the gasoline and B33 combustion at both engine
loads. Although B33 worsens the BSFC compared to gasoline, the higher heat of vaporization of B33
can improve the volumetric efficiency and reduce the heat losses to the coolant and surroundings. The
physicochemical properties of butanol in gasoline blend is reflected in a BTE similar than for gasoline
as the differences are around 1% at both engine conditions (Figure 6 (b)).





Figure 6: a) Brake Specific Fuel Consumption BSFC and b) BTE with and without max %EGR ratio at 2100rpm.



233

234

Figure 7: Exhaust gas temperature at baseline and max % EGR at 2100 rpm

236 *4.2 Gaseous Emissions*

237 THCs and CO emissions provide a direct insight into the combustion process by contributing to 238 the evaluation of the combustion efficiency. The combustion of B33 at 35Nm resulted in slightly 239 lower THC emissions compared to gasoline (Figure 8). Although both the oxygen content of B33 and 240 its shorter carbon chain length are promoting higher oxidation rates to CO_2 , the poor butanol spray 241 and mixing properties at low load inhibit this oxygen effect, increasing CO emissions [40] through 242 incomplete combustion. A significant decrease in THC and CO is noticeable at 60Nm (Figure 9) with 243 the combustion of B33 compared to gasoline. The greater combustion stability, as it can be observed 244 from the COV of IMEP and, higher combustion temperature and heat release rate may be the reasons. 245 The exhaust gas temperature of B33 at 60Nm raised by nearly 10 °C (Figure 7) with respect to 246 gasoline combustion, which is an indication of higher in-cylinder temperature for B33. This resulted 247 in a THC and CO reduction of 12.6% and 4%, respectively, as greater oxidation rates of the THC can 248 be achieved during the combustion and exhaust stages.

249 The in-cylinder pressure increases with EGR, and as a result, the fuel is forced into the piston 250 ring crevices that then is released as unburnt HC emissions during the exhaust stroke [7]. 251 Furthermore, the reduction of combustion temperatures induced by the addition of EGR led to lower 252 heat release and combustion stability. This promotes lower oxidation of THC during the combustion 253 process and exhaust stroke. In addition, the lower mixing time available for fuel-air mixture due to 254 spark timing advance (a total average of 50 CAD advanced, see Table 3) can also result in poorer 255 mixture preparation, which worsens the oxidation of THC [32] for both fuels. The combustion of 256 gasoline at maximum EGR produced higher THC concentration for both engine conditions, even 257 when the stability and exhaust temperature was less favourable for B33. This effect has been reported 258 in literature for EGR rates up to 20%, THCs were found to decrease with the increasing bio-alcohol 259 content [22]. CO emissions were generally reduced with EGR addition compared to baseline 260 combustion for both fuels and engine conditions. A reason could be the reduction of liquid fuel [41] 261 due to a drop in pumping losses. Furthermore, THC oxidation is highly worsened respect to baseline 262 (Figure 8), which leads to lower CO and CO₂ emissions. CO emissions for B33 experienced a

significant reduction in both conditions at maximum EGR rate, accounting for 6.73% and 14.76% at
35Nm and 60Nm respectively, compared to gasoline.



265

266

Figure 8: Specific gaseous emission for baseline and maximum EGR ratio at 35Nm/2100rpm for B33 and

B0

267

268 NOx emissions keep a clear trend at 35 Nm where a decrease of 7.65% in NOx was observed for 269 B33; however, no significant reduction was seen at 60 Nm. At low load, the exhaust gas temperature 270 of both fuels (Figure 7) were the same, which can indicate a narrow difference of in-cylinder 271 temperature (source of thermal NOx formation). However, the exhaust gas temperature is not a direct 272 measurement of the in-cylinder temperature, and no accurate prediction can be made based on an 273 insignificant difference in exhaust gas temperature. Thus, it is thought that butanol's lower adiabatic 274 flame temperature of B33 could reduce the local combustion temperatures compared to gasoline, 275 being the reason for slightly NOx reduction [36, 42] at 35Nm. In addition, the marginally lower heat 276 release rate at the end of the combustion phase could potentially reduce the NOx emissions with 277 respect to gasoline. On the other hand, the reason for the lack of effect of B33 at 60 Nm could be the 278 better homogenisation and stability, enabling to gain higher combustion temperature and heat release 279 rate (Figure 2) as it can be predicted through the exhaust temperatures (Figure 7) in this case [14]. 280 EGR reduces NOx emissions for both fuels. The general reason is the higher overall heat

capacity in the combustion chamber limits the combustion temperature, and consequently the thermal
NOx formation rate. Comparing both engine conditions and fuels at maximum EGR, there was a clear

283 reduction in NOx of 37% at 60Nm by B33 (Figure 9), while there was not benefit in reduction of NOx 284 at low load (Figure 8). Assuming that the in-cylinder temperature with EGR is lower in both engine 285 conditions for B33, which is reflected in exhaust gas temperature (Figure 7), it could be thought that 286 NOx emission for B33 must be lower than in the gasoline case. However, this is not observed in both 287 engine conditions. The high viscosity of butanol together with the low injection pressure at 35Nm 288 worsens the spray pattern and hence, the air-fuel mixture quality. Consequently, the combustion of 289 B33 is more unstable compared to gasoline at low load. This has been reported to promote diffusive 290 flames (precursor of NOx emissions), and as a result, inhibiting the favourable influence of the 291 cooling charge effect of B33 for NOx inhibition [28].





293 The combustion of B33 provided a reduction in CO_2 emissions at both loads when compared to 294 gasoline. The decrease in CO₂ emissions with the butanol blend is due to the higher H/C and O/C ratio 295 of B33 with respect to gasoline fuel. The CO₂ reduction is more noticeable at low load which could be 296 attributed to a lower conversion of THC to CO₂ due to the lower combustion efficiency and lower 297 carbon fuel content. With EGR, CO₂ experienced a reduction for both fuels. The EGR improved 298 BSFC (Figure 6 (a)) and, consequently; a lower mass of fuel is needed to maintain the engine 299 operation condition. In addition, the incomplete combustion of THCs, which increases its emissions, 300 may be another cause of CO₂ reduction. At both conditions with EGR, B33 produced lower CO₂ emissions, which can be again ascribed to its higher H/C and O/C ratios and improved volumetric
efficiency due to the greater cooling charge effect with respect to baseline operation.

303 4.3 PM Emissions

304 Particle number size distribution from the combustion of B33 and gasoline with and without the 305 addition of EGR at maximum rates are plotted in Figure 10. At low engine load (Figure 10 (a)) during 306 baseline combustion, a unimodal particle distribution. Firstly, B33 provided a significant reduction in 307 particle concentration compared to gasoline, accounting for 60% when the peak was considered at 308 35Nm. At this engine condition, gasoline particle size distribution is displaced to larger diameters 309 with respect to B33 results. The combustion of B33 presents a peak governed by nucleation mode 310 particles with the geometric number mean diameter (GNMD) being at 30nm, exhibiting a reduction in 311 size, mass and number of particles relative to gasoline. The accumulation mode of particles has a 312 close relation to the polycyclic aromatic hydrocarbons (PAHs) as it has been reported in literature [1, 313 3, 43, 44]. Thus, the addition of butanol to the gasoline, reduces the aromatics content in the fuel 314 blend, and consequently, the number of particles with high GNMD is decreased, favoring the 315 increased number of nuclei particles [39]. In addition, the presence of oxygen as part of the alcohol 316 fuel reduces the soot formation rate and enlarges the oxidation rate during the combustion process. 317 Hence, the reductions of number and GNMD for B33 combustion decrease the number of interaction 318 between the particles, leading to lower surface growth, coagulation and aggregation processes, and 319 therefore the presence of large hydrocarbons for the formation of the accumulation mode particles 320 decreases.

Similar results were found at 60 Nm (Figure 10 (b)), but in this case the reduction in the peak of the particle distribution was even more notable, accounting for 81% with respect to gasoline. This engine condition promoted a more homogeneous air-fuel mixture for B33, which resulted in higher combustion efficiency and stability (Figure 4). This enhancement in the stability was reflected in higher in-cylinder pressure and heat release that indicate higher combustion temperatures shown by exhaust gas temperatures in (Figure 7). Therefore, the soot and THC oxidation rate is increased, enabling a greater difference between gasoline and B33 at 60Nmwith respect to 35Nm.



Figure 10: Effect of B33 and maximum EGR ratio on particle size distribution: a) 35Nm/2100rpm and b)
 60Nm/2100rpm

330 A general reduction of particle number and size was achieved under EGR conditions. A 331 reasonable explanation for lower PM formation can be attributed to the fact that EGR generally 332 improves engine efficiency and fuel consumption (Figure 6). Therefore, for a given engine load, less 333 fuel is required into the cylinder compared to baseline condition, leading to proportionally less PM 334 being formed [31]. For both fuels and conditions tested, the particle distribution peak shifts towards 335 smaller particles sizes with the introduction of EGR, indicating a growth in proportion of finer 336 particles in benefit of the reduction in particulate mass. The application of EGR led to a 15-20% 337 reduction in the particle concentration peak of the distribution for both fuels and engine operating 338 conditions. Furthermore, the temperature was reduced by EGR, and consequently primary carbon 339 particles formed by thermal pyrolysis and dehydrogenation reaction of fuel vapor may have been 340 decreased [45]. The consequence is that overall concentration decreased with EGR for both fuels.

341 4.4 Primary Particle Diameter (d_{p0}) Analysis

As reported in the previous sections, the butanol blends reduce considerably the concentration of PM independently of the engine condition and the size of the final agglomerate. Gasoline PM presents aciniform-shape formed by several nearly spherical primary particles. In this section, an analysis of the effect of butanol in the primary particle diameter (d_{p0}) has been performed at 60Nm/2100rpm, in which the PM concentration was more significantly reduced. An example of the TEM micrograph of particles collected from the GDI engine exhaust for both fuels is shown in Figure 11. Previous studies
reported the presence of different types of particles in GDI engines: i) nearly-spherical HCs droplets,
ii) solid spherules as small as 6 nm, iii) 'wet' diesel-like particles and iv) 'dry' diesel like particles
[46, 47]. These types of particles were also found in B33 in this research work.





352 The log-normal primary particle size distribution for B33 and gasoline obtained by IBM 353 statistical package for social sciences (SPSS) software are shown in Figure 12. B33 particle 354 agglomerates are formed by similar mean diameter primary particles as the normal log shows. At this 355 engine condition, B33 combustion led to higher in-cylinder pressure (Figure 2, and thus, likely higher in-cylinder temperature (as the exhaust temperature trends also indicates) that could increase the rate 356 357 of particle formation (although also the rate of oxidation), favoring the formation of primary particles 358 with higher diameters [23]. However, the presence of oxygen in B33 seems to counteract this effect, 359 maintaining the size of the formed primary particles the same as the ones recorded from the gasoline 360 combustion. The oxygen content in bio-alcohols fuels can reduce the soot formation rate and enhance 361 the formed particles oxidation rates during the combustion process, [21]. These results are similar 362 than the found for diesel bio-alcohol fuels combustion as reported in literature [48, 49], where in these 363 cases, bio-alcohols even showed slightly lower primary particles diameters due to the oxygen content. 364 Therefore, the oxygen content in B33 is likely to be helpful to limit the rate of primary particles 365 formation.



Figure 12: Primary particles size distributions at 60Nm/2100rpm for: a) Gasoline and b) B33 Additionally, the mean primary diameter and standard deviation achieved from the primary particles size distribution for B33 and gasoline are plotted in Figure 13. The average diameter for B33 was in 28.96 nm while gasoline showed a diameter 29.1 nm, which was also found in our previous work for gasoline [23]. B33 formed a slightly smaller primary particle but the standard deviations overlap. Therefore, it can be concluded that the differences in primary particle diameter are negligible in this study.



Figure 13: Average primary particle diameter

375 4.4 Summary of Results

The effect of B33 with and without EGR are shown in Table 4, to present qualitatively the benefits of B33 in comparison to gasoline baseline combustion.

379

380

Table 4: Performance combustion of B33 combined with EGR with respect to gasoline combustion: blue + positive,

		BSFC	BTE	COV of IMEP	СО	THC	NOx	CO ₂	РМ		
B33		Baseline			-	-	+		++	++	
	35Nm/2100rpm	17% EGR	-			+		++	+	+	dp_0
	B33	Baseline	-		+	+++	+		+	+++	
	601Nm/2100rpm	19% EGR	-			+		++	++	++	

red - negative and - - - insignificant effect.

381 **5. Conclusions**

382 The effect of butanol 33% v/v in EN 228 commercial gasoline containing ethanol 5% v/v (B33) 383 and EGR rate on combustion characteristics and regulated emissions in a multi-cylinder GDI 384 production engine has been investigated. The influence of the butanol addition on the engine 385 combustion characteristics and emissions was dependent on engine operating (i.e. load) conditions. 386 At low engine loads, butanol's physical properties (e.g. high viscosity) are more influential on the 387 combustion performance than its chemical properties (e.g. higher flame speed). Consequently, the 388 combustion of B33 was observed to be more unstable due to the deteriorated fuel spray atomization 389 and in-homogeneous air-fuel mixture, also contributing to the marginally increased carbon monoxide 390 emissions. Conversely, butanol's shorter carbon chain and its oxygen content help to reduce the rest 391 of the emissions. As the engine load was increased, and hence the fuel injection pressure, the 392 combustion performance of B33 was improved through greater air-fuel mixture quality and 393 potentially improved spray atomization; that was reflected in an overall reduction of gaseous and 394 particulates matter emissions.

Transmission Electron Microscope (TEM) analysis showed that B33 did not increase the primary particle diameters, even when the in-cylinder temperature was higher. This confirmed that the oxygen content of butanol could limit the rate of soot formation and at the same time promote soot oxidation. Therefore, in terms of the obtained soot agglomerate surface area to volume ratio, gasoline and its blends with butanol are expected to have the same impact on the soot oxidation process during
the regeneration stages of the Gasoline Particulate Filters. It was shown that B33 was an effective fuel
to reduce most of the legislated emissions in both engine conditions, while maintaining engine brake
thermal efficiency (BTE).

403 The addition of EGR provided a general improvement of BTE and brake specific fuel 404 consumption (BSFC) for both fuels and was beneficial to B33 since greater engine-out emissions 405 reduction was achieved.

The research work presented here has shown that high percentages of butanol in gasoline blends combined with EGR technology can be a potential solution in GDI engines for reducing legislated emissions while maintaining the BTE compare to gasoline. However, it is anticipated that the calibration and injection systems of the engine would have to be adapted to minimize its limited performance at low loads, since the physical properties can predominate over its advantageous chemical properties.

412 <u>Acknowledgments:</u> C.H.S.O. would like to thank University of Birmingham for his scholarship. M.B.M. 413 would like to thank EPSRC (Grant No: 1377213) for providing her scholarship. Innovate UK (Technology 414 Strategy Board) is acknowledged for supporting this work with the project "CO₂ Reduction through Emissions 415 Optimisation" (CREO: ref. 400176/149). The Advantage West Midlands and the European Regional 416 Development Fund as part of the Science City Research Alliance Energy Efficiency Project are also 417 acknowledged for supporting the research work.

418 **References**

F. Zhao, M.C. Lai, and D.L. Harrington, Automotive spark-ignited direct-injection gasoline engines. *Prog. Energ. Combust.*, 1999. 25(5): p. 437-562.

- 421 2. A.C. Alkidas, Combustion advancements in gasoline engines. *Energ. Convers. Manage.*, 2007.
 422 48(11): p. 2751-2761.
- G. Karavalakis, et al., The impact of ethanol and iso-butanol blends on gaseous and particulate
 emissions from two passenger cars equipped with spray-guided and wall-guided direct injection SI
 (spark ignition) engines. *Energy*, 2015. 82: p. 168-179.
- 4264.D.C. Quiros, et al., Particle effective density and mass during steady-state operation of GDI, PFI, and427diesel passenger cars. J. Aerosol Sci., 2015. 83: p. 39-54.

- 428 5. C. Wang, et al., Impact of fuel and injection system on particle emissions from a GDI engine. *Appl.*429 *Energ*, 2014. 132: p. 178-191.
- 430 6. J.E. Ketterer and W.K. Cheng, On the Nature of Particulate Emissions from DISI Engines at
 431 Cold-Fast-Idle. *SAE J-Automot.Eng.*, 2014. 7(2): p. 986-994.
- 432 7. A.O. Hasan, et al., Control of harmful hydrocarbon species in the exhaust of modern advanced GDI
 433 engines. *Atmos.Environ*, 2016. **129**: p. 210-217.
- 434 8. D.A. Fennell, Exhaus Gas Fuel Reforming for Improved Gasoline Direct Injection Engine Efficiency
 435 and Emissions Mechanical Engineering at University of Birmingham, Birmingham, 2014.
- 436 9. F. Steimle, et al. "Systematic Analysis and Particle Emission Reduction of Homogeneous Direct
 437 Injection SI Engines", <u>2013-01-0248</u>, 2013.
- 438 10. S. Heyne and S. Harvey, Assessment of the energy and economic performance of second generation
 439 biofuel production processes using energy market scenarios. *Appl.Energ*, 2013. 101: p. 203-212.
- B. Deng, et al., The heat release analysis of bio-butanol/gasoline blends on a high speed SI (spark
 ignition) engine. *Energy*, 2013. 60: p. 230-241.
- 442 12. Z. Chen, et al., Impact of higher n-butanol addition on combustion and performance of GDI engine in
 443 stoichiometric combustion. *Energ. Convers. Manage*, 2015. 106: p. 385-392.
- J.M. Bergthorson and M.J. Thomson, A review of the combustion and emissions properties of
 advanced transportation biofuels and their impact on existing and future engines. *Renew.Sust.Energ.Rev.*, 2015. 42: p. 1393-1417.
- 447 14. R. Feng, et al., Experimental study on SI engine fuelled with butanol–gasoline blend and H2O
 448 addition. *Energ. Convers. Manage*, 2013. **74**: p. 192-200.
- B.R. Wigg, A Study on the Emissions of Butanol Using a Spark Ignition Engine and their Reduction
 Using Electrostatically Assisted Injection. Mechanical Engineering at University of Illinois,
 Urbana-Campaign, 2011.
- 45216.B. Deng, et al., The challenges and strategies of butanol application in conventional engines: The453sensitivity study of ignition and valve timing. *Appl.Energ*, 2013. **108**: p. 248-260.
- M. Pechout, M. Mazac, and M. Vojtisek-Lom. "Effect of Higher Content N-Butanol Blends on
 Combustion, Exhaust Emissions and Catalyst Performance of an Unmodified SI Vehicle Engine",
 SAE, <u>2012-01-1594</u>, 2012.
- T. Wallner, S.A. Miers, and S. McConnell, A Comparison of Ethanol and Butanol as Oxygenates
 Using a Direct-Injection, Spark-Ignition Engine. *J.Eng.Gas.Turb.Power*, 2009. **131**(3): p.
 032802-032802.
- 460 19. G. Broustail, et al., Comparison of regulated and non-regulated pollutants with iso-octane/butanol and
 461 iso-octane/ethanol blends in a port-fuel injection Spark-Ignition engine. *Fuel*, 2012. 94: p. 251-261.
- 462 20. F. Catapano, et al. "Effects of Ethanol and Gasoline Blending and Dual Fueling on Engine
 463 Performance and Emissions", SAE, <u>2015-24-2490</u>, 2015.
- 464 21. E.J. Barrientos, et al., Particulate matter indices using fuel smoke point for vehicle emissions with
 465 gasoline, ethanol blends, and butanol blends. *Combust.Flame*, 2016. 167: p. 308-319.
- Z. Zhang, et al., Combustion and particle number emissions of a direct injection spark ignition engine
 operating on ethanol/gasoline and n-butanol/gasoline blends with exhaust gas recirculation. *Fuel*,
 2014. 130: p. 177-188.

469	23.	M. Bogarra, et al., Impact of exhaust gas fuel reforming and exhaust gas recirculation on particulate
470		matter morphology in Gasoline Direct Injection Engine. J. Aerosol Sci., 2017. 103: p. 1-14.
471	24.	T.L. Barone, et al., Inertial deposition of nanoparticle chain aggregates: Theory and comparison with
472		impactor data for ultrafine atmospheric aerosols. J. Nanopart. Res., 2006. 8(5): p. 669-680.
473	25.	P. Karin, et al., Morphology and oxidation kinetics of CI engine's biodiesel particulate matters on
474		cordierite Diesel Particulate Filters using TGA. Int. J. Automot. Techn., 2017. 18(1): p. 31-40.
475	26.	K.H. Yoo, et al. "Experimental Studies of EGR Cooler Fouling on a GDI Engine", SAE Technical
476		Paper, <u>2016-01-1090</u> , 2016.
477	27.	G. Fontana and E. Galloni, Experimental analysis of a spark-ignition engine using exhaust gas recycle
478		at WOT operation. Appl.Energ, 2010. 87(7): p. 2187-2193.
479	28.	H. Wei, et al., Gasoline engine exhaust gas recirculation – A review. Appl.Energ, 2012. 99: p. 534-544.
480	29.	J.M. Luján, et al., Influence of a low pressure EGR loop on a gasoline turbocharged direct injection
481		engine. Appl. Therm. Eng, 2015. 89: p. 432-443.
482	30.	W. Zeng, M. Sjöberg, and D.L. Reuss, Combined effects of flow/spray interactions and EGR on
483		combustion variability for a stratified DISI engine. Proc. Combust. Inst, 2015. 35(3): p. 2907-2914.
484	31.	D. Fennell, J. Herreros, and A. Tsolakis, Improving gasoline direct injection (GDI) engine efficiency
485		and emissions with hydrogen from exhaust gas fuel reforming. Int. J. Hydrogen Energ, 2014. 39(10):
486		p. 5153-5162.
487	32.	T. Lattimore, et al., Investigation of EGR Effect on Combustion and PM Emissions in a DISI Engine.
488		Appl.Energ, 2016. 161: p. 256-267.
489	33.	M. Hedge, et al., Effect of EGR on Particle Emissions from a GDI Engine. SAE Int. J. Engines 2011. 4:
490		р. 650-666.
491	34.	C. Tornatore, et al., Optical diagnostics of the combustion process in a PFI SI boosted engine fueled
492		with butanol-gasoline blend. <i>Energy</i> , 2012. 45 (1): p. 277-287.
493	35.	R. Scarcelli, et al. "Cycle-to-Cycle Variations in Multi-Cycle Engine RANS Simulations", SAE
494		Technical Paper, <u>2016-01-0593</u> , 2016.
495	36.	A. Irimescu, et al., Combustion process investigations in an optically accessible DISI engine fuelled
496		with n-butanol during part load operation. Renew. Energ, 2015. 77: p. 363-376.
497	37.	K. Kondo, et al. "Uncertainty in Sampling and TEM Analysis of Soot Particles in Diesel Spray Flame",
498		SAE Technical Paper, <u>2013-01-0908</u> , 2013.
499	38.	C.K. Gaddam and R.L. Vander Wal, Physical and chemical characterization of SIDI engine
500		particulates. Comb. Flame, 2013. 160(11): p. 2517-2528.
501	39.	R. Daniel, et al., Ignition timing sensitivities of oxygenated biofuels compared to gasoline in a
502		direct-injection SI engine. Fuel, 2012. 99: p. 72-82.
503	40.	E. Galloni, et al., Performance analyses of a spark-ignition engine firing with gasoline-butanol blends
504		at partial load operation. Energ. Convers. Manage, 2016. 110: p. 319-326.
505	41.	M. Bogarra, et al., Study of particulate matter and gaseous emissions in gasoline direct injection engine
506		using on-board exhaust gas fuel reforming. Appl. Energ, 2016. 180: p. 245-255.
507	42.	S.S. Merola, et al., Optical diagnostics of early flame development in a DISI (direct injection spark
508		ignition) engine fueled with n-butanol and gasoline. Energy, 2016. 108: p. 50-62.
509	43.	X. Gu, et al., Emission characteristics of a spark-ignition engine fuelled with gasoline-n-butanol
510		blends in combination with EGR. Fuel, 2012. 93: p. 611-617.

511	44.	J.M. Storey, et al., Novel Characterization of GDI Engine Exhaust for Gasoline and Mid-Level
512		Gasoline-Alcohol Blends. SAE Int. J. Fuels Lubr, 2014. 7: p. 571-579.
513	45.	T. Alger, et al., The Role of EGR in PM Emissions from Gasoline Engines. SAE Int. J. Fuels Lubr,
514		2010. 3 : p. 85-98.
515	46.	P. Karjalainen, et al., Exhaust particles of modern gasoline vehicles: A laboratory and an on-road
516		study. Atmos. Environ., 2014. 97: p. 262-270.
517	47.	T.L. Barone, et al., An analysis of direct-injection spark-ignition (DISI) soot morphology. Atmos.
518		<i>Environ.</i> , 2012. 49 : p. 268-274.
519	48.	M.A. Fayad, et al., Manipulating modern diesel engine particulate emission characteristics through
520		butanol fuel blending and fuel injection strategies for efficient diesel oxidation catalysts. Appl. Energ.,
521		2017. 190 : p. 490-500.
522	49.	H. Yang, et al., Experimental investigation into the oxidation reactivity and nanostructure of
523		particulate matter from diesel engine fuelled with diesel/polyoxymethylene dimethyl ethers blends.
524		<i>Sci. Rep.</i> , 2016. 6 : p. 37611.