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Effects of natural gas composition on performance and regulated, greenhouse gas and particulate emissions in spark-ignition engines

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ABSTRACT

In vehicles fueled with compressed natural gas, a variation in the fuel composition can have non-negligible effects on their performance, as well as on their emissions. The present work aimed to provide more insight on this crucial aspect by performing experiments on a single-cylinder port-fuel injected spark-ignition engine. In particular, methane/propane mixtures were realized to isolate the effects of a variation of the main constituents in natural gas on engine performance and associated pollutant emissions. The propane volume fraction was varied from 10 to 40%. Using an experimental procedure designed and validated to obtain precise real-time mixture fractions to inject directly into the intake manifold. Indicative Mean Effective Pressure, Heat Release Rate and Mass Burned Fraction were used to evaluate the effects on engine performance. Gaseous emissions were measured as well. Particulate Mass, Number and Size Distributions were analyzed with the aim to identify possible correlations existing between fuel composition and soot emissions. Emissions samples were taken from the exhaust flow, just downstream of the valves. Opacity was measured downstream the Three-Way Catalyst. Three different engine speeds were investigated, namely 2000, 3000 and 4000 rpm. Stoichiometric and full load conditions were considered in all tests. The results were compared with pure methane and propane, as well as with natural gas. The results indicated that both performance and emissions were strongly influenced by the variation of the propane content. Increasing the propane fraction favored more complete combustion and increased NO_x emissions, due to the higher temperatures. In all tests, natural gas showed the highest PN values. At high speeds, adding propane increased the number of particles between 5 and 30 nm, highlighting the relevance of the ultrafine particles. Smaller differences were recorded at low speeds.

1. Introduction

Over the past several years, road transportation has seen significant advances and new alternative technologies are rapidly emerging. Energy production and storage, electric drive systems, and fuel cell technologies all seem able to find a significant place in the automotive marketplace [1–5]. However, it would be a mistake to believe that such technologies will completely replace conventional internal combustion engines in short time [6]. The need for practical mid-term solutions that can meet new fuel economy and emissions standards has pushed the development of new technologies for internal combustion engines, comprising innovative combustion techniques [7–10] as well as their control strategies [11–17].

In addition, alternative fuels are being promoted and developed to replace traditional fuels [18–22]. Natural gas represents one of the most concrete alternatives to conventional petroleum fuels (especially in the heavy-duty vehicle segment) since it produces significantly lower emissions, such as particulate matter (PM) and oxides of nitrogen (NO_x), than conventional diesel engines [8,23–25]. For these reasons, over the past eight years, 50% of the transport bus fleet in Brisbane, Australia, has been gradually converted from diesel to CNG. In New Delhi, India, one of the most polluted cities in the world, the entire transport fleet was converted to CNG in 2003 resulting in some improvement in air quality in terms of suspended particulate matter, CO, SO_2, and NO_x [26,27].

For CNG vehicles, one issue is that a variation in the fuel composition can have non-negligible effects on the combustion process

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Nomenclature

$\dot{m}_i$	mass flow rate
$\dot{m}_{exp}$	measured mass flow rate
m_{CH_4}	methane injected mass
$m_{C_3H_8}$	propane injected mass
χ_{CH_4}	methane volume fraction
$\chi_{C_3H_8}$	propane volume fraction
MW_{CH_4}	methane molecular weight
$MW_{C_3H_8}$	propane molecular weight
ζ_{CH_4}	constant in Eq. (6) relative to methane
$\zeta_{C_3H_8}$	constant in Eq. (6) relative to propane
Γ	corrected mass flow rate with respect to the injection frequency
$\Gamma_{95\%}$	corrected mass flow rate measured when the DOI is equal to 95%
Γ_{ref}	value of the corrected mass flow rate measured at the reference injection pressure
D_L	deviation from the Linearity
A_{inj}	nozzle cross section area of the injector
p_i^0	total injection pressure
$p_{i,ref}^0$	reference total injection pressure, equal to 3.5 bar
T_i^0	total injection temperature
R_i	individual gas constant
γ_i	specific heat ratio
f_{inj}	injection frequency
f_{inj}^{ref}	reference injection frequency
Δt_i	duration of injection
Γ	corrected mass flow rate

a, **b**	correction constants in Eqs. (4) and (6)
i	refers to either methane or propane

Definitions/Abbreviations

ABDC	After Bottom Dead Center
ATDC	After Top Dead Center
BBDC	Before Bottom Dead Center
BTDC	Before Top Dead Center
CNG	Compressed Natural Gas
COV	Coefficient Of Variation
DOC	Duration of Combustion
DOI	Duration Of Injection
HHR	Heat Release Rate
IMEP	Indicating Mean Effective Pressure
MBF	Mass Burned Fraction
MN	Methane Number
NO_x	Oxides of Nitrogen
PM	Particulate matter
PN	Particle number
PSDs	Particle Size Distributions
TUHCs	Total Unburned Hydrocarbons
TWC	Three-Way Catalyst
UHCs	Unburned Hydrocarbons
VPR	Volatile Particle Remover

[28–32]. In fact, natural gas is a mixture of various hydrocarbon molecules: the principal component is methane and its volume fraction can vary from 55.8% to 98.1%; the main heavy hydrocarbons present in natural gas are ethane, which can vary between 0.5% and 13.3% (by volume), and propane, in amounts varying between 0% and 23.7% (by volume) [33]. Diluents such as N_2 and CO_2 are also present in significant fractions. There are also trace levels of sulfur compounds, often added as odorants, and hydrocarbons larger than C_3 [30]. The components concentration change with geographical source, time of year, and treatments applied during production or transportation [34].

Previous studies have shown that changes in natural gas composition can impact emissions, as well as engine performance [28–32]. Karavalakis et al. [35] reported that natural gases with higher heating value exhibited higher fuel economy on an energy equivalent basis. Higher flame speeds and higher adiabatic flame temperatures can be obtained with larger amounts of ethane and propane in natural gas, producing more efficient combustion [30,35,36]. A reduction in Total Unburned Hydrocarbon (TUHC) emissions was seen for fuels with higher hydrocarbon contents [29,30]. Some researchers report increases in TUHC emissions with increased ethane and propane concentration [37], although these results are not consistent with other previous studies. NO_x emission levels were clearly influenced by the fuel composition, with low Methane Number (MN) natural gases resulting in higher NO_x emissions [29–32,35]. McTaggart-Cowan et al. [30] suggested that it was due to the increased adiabatic flame temperature with a higher fraction of ethane and propane, since NO_x are generated predominantly through the strongly temperature-dependent thermal NO mechanism [38]. The authors of such work found that a 1% change in adiabatic flame temperature resulted in a 5% change in NO_x emissions. CO is another combustion by-product that is sensitive to fuel composition, but discordant results have been reported in literature [30,35].

Furthermore, current emission regulations emphasize the need to control greenhouse gas emissions from on-road sources, and consequently there is a need to control methane, as well as CO_2 emissions, from natural gas vehicles [39]. Methane is not toxic and not relevant to ozone-forming potential, but it shows a global warming potential 25 times higher than CO_2 [29]. In general, higher methane emissions were recorded for higher MN fuels [29] and this might be due to the fact that methane is less reactive than higher chain hydrocarbons, so it is more likely that higher amounts survive the combustion process [40]. Higher CO_2 emissions were recorded for natural gases having higher fraction of higher hydrocarbons [35].

McTaggart-Cowan et al. [30] found that relatively high levels of ethane and propane in natural gas can significantly increase Particulate Matter (PM) emissions. In such a study, both black carbon and volatile PM emissions were claimed to be increased by an increase in ethane and propane contents, and other studies have confirmed this trend [41]. The presence of hydrocarbons with longer chains or more complex structures can enhance PM precursor formation in the reaction zone [42,43], including C_2 species, such as the ethyl radical (C_2H_5) and acetylene (C_2H_2) [41] which are the most abundant gaseous hydrocarbon species in regions where soot is formed in laminar premixed flames [44,45].

There is a lack of information about the effect that a variation of natural gas composition can produce on Particle Number density (PN) and Size Distribution (PSD) functions. Karavalakis et al. [29] recently reported some measurements, but a clear and exhaustive understanding of the phenomenon is still needed. Therefore, substantially more work is required to understand the effects of the heavier hydrocarbons on particle formation in natural gas engines.

The present study aims to isolate the influence that hydrocarbons heavier than methane have on natural gas combustion. Ethane and propane are the other two hydrocarbons that are present in a relevant amount in natural gas. However, propane, more