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Effects of the sulfur content of liquefied petroleum gas on regulated and unregulated emissions from liquefied petroleum gas vehicle

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HIGHLIGHTS

- Sulfur content effects in LPG on the performance of LPG vehicle were investigated.
- Regulated and unregulated time-resolved emissions increased during cold start phase.
- Most of the mass emissions increased as the sulfur content of the LPG increased.
- However, Fuel economy was not affected by the sulfur content of the LPG fuel.

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ABSTRACT

A more stringent standard for sulfur in liquefied petroleum gas (LPG) is currently being considered by the Ministry of Environment of Korea. Therefore, we investigated the characteristics of exhaust emissions from liquid phase LPG injection passenger car depending on the sulfur content of the LPG. Using a chassis dynamometer test with exhaust gas analyzers and Fourier-transform infrared spectroscopy, regulated emissions of carbon monoxide, non-methane hydrocarbons, and nitrates were investigated, as well as unregulated emissions of CH_4, N_2O, NH_3, and SO_2. Vehicle tests were carried out using Federal Test Procedure 75 (FTP-75) and Highway Fuel Economy Test (HWFET) cycles and time-resolved exhaust emissions and mass emissions were analyzed. The concentrations of regulated and unregulated time-resolved emissions increased during the cold start phase (Phase 1) of the FTP-75 cycle, in which the catalyst does not reach the light-off temperature. In the FTP-75 and HWFET cycles, most of the regulated and unregulated mass emissions increased as the sulfur content of the LPG increased. The SO_2 concentration in the emissions increased in proportion to the sulfur content in the LPG, particularly in the high-speed portion of the FTP-75 cycle.

1. Introduction

Liquefied petroleum gas (LPG or autogas) is a byproduct of the crude oil refinery process and mainly consists of propane (C_3H_8) and butane (C_4H_{10}). The composition of LPG for vehicle use varies by country; the standards for LPG in Korea are shown in Table 1. In Korea, the main component of LPG as a vehicular fuel is butane, and propane is added at 15–35 mol% to improve the ability to start the vehicle in the winter. Except during this season, the propane content is restricted to < 10 mol%. The sulfur content of LPG is limited to < 40 ppm.

LPG is known as a clean fuel, because it contains less carbon than gasoline. LPG vehicles typically produce lower CO_2 emissions than gasoline vehicles, as well as reductions of up to 40% and 60% for hydrocarbon (HC) and carbon monoxide (CO) emissions, respectively [1]. In addition, the concentration of nanoparticles emitted from LPG engines is much lower than that from gasoline engines [2].

Recently, many countries have promoted the use of LPG as an environmentally beneficial and economical alternative fuel. Currently, about 2.5 million LPG vehicles, representing 13% of the registered vehicles, are being driven in Korea due to the relatively low price of LPG fuel. About 80% of these vehicles are passenger cars.

Liquid phase LPG injection (LPLi) and gas phase LPG injection (LPGi) type LPG vehicles have been produced in the Korean market. Mixer type LPG vehicles having low performance are no longer produced. Because the LPLi vehicles inject the liquid phase LPG into the engine intake port, it is possible to control the fuel flow rate precisely and the volumetric efficiency of the engine is increased by reducing the intake air temperature due to vaporization of the

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Table 1
Automotive LPG fuel standards in Korea.

Property	Until December 31 2008	After January 1 2009
Sulfur content (wt ppm)	100 max	40 max
Vapor pressure (40 °C, MPa)	1.27 max	1.27 max
Density (15 °C, kg/m^3)	500–620	500–620
Copper strip corrosion (1 h at 40 °C)	1 max	1 max
Residue on evaporation of 100 mL (mL)	0.05 max	0.05 max
Propane content (mole %) November 1–March 31	15–40	15–40 (35)[a]
April 1–Oct 31	10 max	10 max

[a] Effective on December 1 2010.

LPG. Therefore, fuel economy and power are greater and the exhaust emissions of the LPLi vehicles are lower than those of conventional mixer type LPG and LPGi vehicles [3]. In LPGi vehicles, the liquid phase LPG in the evaporator is vaporized and the gas is injected into the engine intake port. The performance of LPGi vehicles is lower than that of LPLi vehicles, because their volume efficiency is lower due to superheating of the LPG gas in the evaporator and because the fuel flow rate is less controlled than in an LPLi engine [4].

LPG contains a small amount of sulfur after refining from crude oil, and this sulfur concentration is increased by odorants that contain sulfur. Sulfur poisons the three-way catalyst of LPG and gasoline vehicles, which reduces the conversion efficiency of the catalyst [5,6] and results in increased hazardous emissions [7,8]. SO_2 also contaminates the exhaust oxygen sensor, which interferes with precise feedback controlling the air to fuel ratio. Alliance and Association of International Automobile Manufacturers (AIAM) have reported that emissions of HC, CO, and NO_x were reduced by more than 20% in low and ultralow emissions vehicles when the sulfur content in gasoline decreased from 100 to 30 ppm [9].

The Ministry of Environment in Korea has gradually reduced the allowable sulfur content in the quality standards for vehicular fuel. In 2009, the allowable sulfur content in LPG was lowered to < 40 ppm from < 100 ppm. This limit, however, is still much higher than 10 ppm, which is the sulfur content limit for gasoline and diesel [10]. Therefore, a more stringent standard is currently being considered by the Ministry of Environment. In the future, lean burn and/or LPG direct injection (LPDI) engines are expected to be introduced to improve vehicle fuel economy. Ultra-low sulfur fuel may be required to improve the durability of the $DeNO_x$ catalyst used in these new vehicles, because this catalyst is much more sensitive to sulfur than the three-way catalyst [11].

In this study, regulated and unregulated exhaust emissions from an LPLi vehicle were investigated for LPG fuels with sulfur contents of 4 ppm (sulfur free level) and 40 ppm. Emissions analyzed included regulated gases such as CO, non-methane hydrocarbons (NMHC), and NO_x and unregulated gases such as CH_4, N_2O, NH_3, and SO_2. Among these gases, CH_4 and N_2O are greenhouse gases whose global warming potentials are 21 and 310 times greater than that of CO_2, respectively [12]. Ammonia is a toxic gas that directly affects human health and is also a secondary particulate matter precursor, along with SO_2 and NO_x.

2. Experimental setup

2.1. Test vehicle and test cycles

An LPLi type LPG passenger car was tested with the Federal Test Procedure 75 (FTP-75) cycle and the Highway Fuel Economy Test (HWFET) cycle. The specifications of the test vehicle are shown in Table 2.

Table 2
Specifications of the test vehicle.

Fuel injection type	LPLi
Displacement	1998 cc
Model year	2010
Odometer	7300 km
Catalyst type	Three-way catalyst
Manufacturer	Kia
Model	K5

The FTP-75 cycle is based on the urban dynamometer driving schedule (UDDS) with an average speed of 34.12 km/h. It consists of three phases: in Phase 1, a cold start cycle that simulates driving after a cold start runs for 5.78 km in 505 s; in Phase 2, a stabilized cycle simulates driving in a stable state and runs for 6.29 km in 865 s; and in Phase 3, a hot start cycle, in which the engine is stopped and started again after Phase 2, consists of the same speed profile as Phase 1. The HWFET cycle is a highway driving cycle whose running distance and average speed are 16.45 km and 77.7 km/h, respectively.

To ensure that the test vehicle was in a normal state, a preliminary test was carried out using the FTP-75 cycle, which is the emissions certification test cycle in Korea. The test vehicle's emissions met the Ultra Low Emissions Vehicles (ULEV) standard for LPG vehicles in Korea, as shown in Table 3.

2.2. Test fuels

As noted in Section 1, the composition of propane and butane in LPG depends on the season, in accordance with the quality standards for LPG in Korea. In this study, LPG fuels with 4 and 40 ppm sulfur were used as test fuels; the propane and butane contents were held constant to exclude the effects of compositional change on exhaust emissions. The properties of the LPG fuels tested are shown in Table 4. The test fuels were stored in their original 85-L containers. When a test fuel was replaced with another, the remaining fuel in the fuel line of the engine was flushed out by driving Phases 1 and 2 of the FTP-75 cycle.

2.3. Test equipments and emissions analysis

A 48-inch single-roll chassis dynamometer (ECDM-48L; MAHA-AIP), a gaseous emissions analyzer (MEXA-9200; Horiba), and a full-flow constant volume sampling exhaust gas analyzer system (CVS-9100; Horiba) were used to measure emissions of CO_2, NMHC, CO, NO_x, and CH_4. Simultaneously, unregulated emissions (N_2O, SO_2, NH_3) were measured by Fourier-transform infrared (FTIR) spectroscopy, as shown in Table 5. The experimental set-up is shown schematically in Fig. 1.

3. Results and discussions

3.1. Time-resolved emissions

Fig. 2 shows the catalyst-out emissions concentrations for regulated and unregulated emissions of the test vehicle measured on a real-time basis during the FTP-75 cycle using LPG fuel with a sulfur content of 40 ppm. In general, engine-out CO from a spark-ignited engine is produced mainly from incomplete combustion under rich mixture conditions; engine-out NMHC is produced both during non-combustion and incomplete combustion. Generally, these gases are reduced by the three-way catalyst when the engine is operated under a stoichiometric air-to-fuel ratio. However, the concentrations of CO and NMHC significantly increased