Fuel xxx (2015) xxx–xxx

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

The formation and physical properties of the particle emissions from a natural gas engine

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HIGHLIGHTS

- Natural gas engine particle emissions were studied in engine dynamometer.
- Exhaust particle size distribution, volatility and electric charge were measured.
- Particle features suggest that they form originally in vicinity of engine cylinders.
- Size range 1–5 nm is relevant for natural gas engine emitted particle emissions.

ARTICLE INFO

Article history:
Received 30 June 2015
Received in revised form 29 August 2015
Accepted 1 September 2015
Available online xxxx

Keywords:
Fine particle emission
Particle formation
Natural gas
Internal combustion engine

ABSTRACT

Natural gas engine particle emissions were studied using an old gasoline engine modified to run with natural gas. The tests were steady-state tests performed on two different low loads in an engine dynamometer. Exhaust particle number concentration, size distribution, volatility and electric charge were measured. Exhaust particles were observed to have peak diameters below 10 nm. To get the full picture of particle emissions from natural gas engines, size range 1–5 nm is relevant and important to take into consideration. A particle size magnifier (PSM) was used in this engine application for measuring particles smaller than 3 nm and it proved to be a useful instrument when measuring natural gas engine exhaust particles. It is concluded that the detected particles probably originated from the engine cylinders or their vicinity and grew to detectable sizes in the sampling process because a small fraction of the particles were observed to carry electric charge and the particles did not evaporate totally at 265 °C.

1. Introduction

The usage of natural gas and the interest towards it have risen because of its smaller environmental effects compared to diesel and gasoline and its improved availability due to shale gas. As a fast reserve power for renewable energy sources such as wind and solar power, engine power plants have and will become more important. Natural gas is also an attracting alternative for diesel in piston engines in energy production and transport due to its potentially smaller carbon dioxide and particulate mass emissions [1].

Fine particles can possess a threat to human health. There is wide epidemiological evidence that especially particles smaller than 2.5 μm increase mortality already at moderate mass concentrations [2–4]. Mortality is connected with cardiovascular diseases and combustion related particles. Ultrafine particles smaller than 100 nm have larger surface area per unit mass and they can penetrate deeper into lungs when inhaled. Typically they do not contribute significantly to particle mass but can be significant in terms of number concentration.

Particle emissions of natural gas engines have not been studied in detail until recently, and current research in the field is concentrated on natural gas buses. Size distribution measurements have shown the size of natural gas emission particles to be in the nanoparticle size range (below 50 nm) [5–7] both in transient and in steady driving conditions. Particle size distribution peaks smaller than 12 nm have been measured by Hajbabaei et al. [8], Jayaratne et al. [1] and Hallquist et al. [9]. Limitations in instrument detection limits could be the reason for them not reporting even smaller peak diameters; especially the size distributions reported by Hajbabaei et al. [8] indicate the particles to exist also below the detection limit of the instrument. No research has been

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http://dx.doi.org/10.1016/j.fuel.2015.09.003
0016-2361/

reported about natural gas engine exhaust particles in the size range below 4 nm. However, if a substantial fraction of exhaust particles is smaller than 4 nm, the whole picture is not obtained if those particles are not taken into account. The smallest nanoparticles are potentially hazardous because of their ability to enter human central nervous system [10].

The number concentrations of particles emitted by natural gas engines are not necessarily small when compared to diesel engines in spite of small soot particle formation in the engines. Jayaratne et al. [11] and Hallquist et al. [9] raise a concern that particle number emissions from CNG (compressed natural gas) buses are an order of magnitude larger than from diesel buses with no after-treatment although the mass concentration of particles is substantially smaller. The maximum concentration in exhaust gas is measured to be $3 \cdot 10^5$ 1/cm^3 [5] or $1 \cdot 10^7$ 1/cm^3 [6] when the subject of experiment has been a large scale natural gas engine with maximum power more than 298 kW and $1.3 \cdot 10^6$ 1/cm^3 for a natural gas bus equipped with an oxidation catalyst at maximum load [1]. The particle number emission factors for CNG buses are of order of magnitude 10^{14} 1/km [9,1].

Physical and chemical characteristics of natural gas engine exhaust particles have previously been measured by e.g. Bullock and Olfert [12], Jayaratne et al. [7] and Yoon et al. [13]. Less than 5% of the particulate matter volume has been observed to remain non-volatile at 100 °C for a homogeneous charge compression ignition engine [12] and for CNG buses, approximately 85% of the exhaust particles are volatile at 100 °C and 98% at 250 °C [7]. The density of natural gas particulates has been found out to be 850 kg/m^3 [12]. A filter collection study by Yoon et al. [13] reveals that natural gas buses emit particles that contain elemental and organic carbon, aromatic hydrocarbons, and lubricating oil originating calcium, phosphorus, zinc, magnesium and sulfur.

Studies on natural gas combustion in flame or commercial burners can possibly provide information about natural gas engine combustion process and related particulate formation without the effect of lubricating oil. Methane fueled cook stove and burner particulates are carbonaceous nanoparticles with modal diameter at 2–4 nm [14]. According to Wagner et al. [15], the diameter of the particles emitted by natural gas fired domestic gas cookers are about 6–10 nm. On the other hand, Murr and Soto [16] defined by TEM study that natural gas burners emit particulates that are larger carbon aggregates with diameter at micron scale. Particles generated in ethylene flame have sizes of about 2–10 nm [17].

In the future the need to reduce the emissions also from natural gas engines might become more important. Therefore they need techniques to reduce their particle emissions. If the origin, formation process and the properties of the emissions are known, reducing them will be easier. In this study, detailed measurements about the primary particle emission characteristics of an engine running on natural gas were performed, including measurements of particles with diameter below 4 nm. The emissions of a natural gas engine were studied using a small-scale retrofitted natural gas engine. The original goal of the project behind the study was to imitate the emissions of a large-scale natural gas engine power plant. A small-scale engine was used in order to save space and fuel and to be able to run long detailed measurements and to test different exhaust after-treatment systems cost-efficiently in later phases of the project. This article focuses on exhaust particle formation phenomena in a natural gas engine, but not in any specific engine type.

2. Experimental

2.1. Engine, fuel and lubricating oil, and driving conditions

The engine used in the experiments was a passenger car gasoline engine that was modified to run with natural gas by installing natural gas injection nozzles in its intake manifold [18]. The original engine was an in-line, 4-cylinder, model-year 1999 engine with a total displacement of 1998 cc and maximum power 100 kW/5500 rpm. The engine was run in an engine dynamometer without exhaust after-treatment.

The engine was operated at two different engine modes which were chosen so that the gaseous emissions of the test engine were similar to the gaseous emissions of a natural gas engine power plant at two different constant loads. Carbon monoxide and NO_x levels were searched with engine adjustments but hydrocarbons were needed to add into the exhaust gas at one of the engine modes in order to produce the exhaust composition of the power plant engine that was imitated. The specification of the test engine modes are presented in Table 1. All aerosol measurements reported in this article were performed at both engine modes.

The fuel used was Russian pipeline natural gas with high methane content. It contained 97.2 vol% methane, 1.37% ethane, 0.17% propane, 0.07% other hydrocarbons, 0.9% nitrogen and 0.2% carbon dioxide. The sulfur content of the fuel was below 1.5 ppm. The lubricating oil was produced by Neste Oil Plc. The content of sulfur, calcium, phosphor and zinc were 1280 mg/kg, 2580 mg/kg, 721 mg/kg and 700 mg/kg, respectively. The mass percentage of sulfated ash was 0.97% for the lubricating oil. Viscosity at 40 °C was 68.4 mm^2/s and density was 852.7 kg/m^3.

2.2. Sampling system and measurement setup

Particle emission sampling system consisted of a porous tube diluter (PTD, [19,20]), a residence time tunnel and an ejector diluter (Dekati Ltd.). Sample was taken from the exhaust pipe after an exhaust gas heating unit to a dilution system. Residence time in the primary dilution system, i.e. in the PTD and the residence time tunnel, was 2.6 s. The dilution ratio (DR) over the porous tube diluter during the measurements was set to be as small as 6 because the particle number concentrations in the exhaust line were low and close to the detection limit of used aerosol instruments (especially EEPS and Nano-SMPS). The secondary dilution ratio over the ejector diluter was 4, resulting in a total dilution ratio of 24.

Particulate mass measurements were completed as filter collections and the sampling was done using an AVL SPC 427 Smart Sampler partial flow dilution system. The filter collections followed standard ISO 8178-1:2006(E). Particulate mass was collected on a pair of 70 mm diameter Pallflex TX40-HI20WW filters using a 1.5 g/s sample flow and a dilution ratio of 10. The sample gas temperature at the sample filter was 43–49 °C. Sample filters were weighed in a special weighing room in which the temperature and humidity were controlled according to ISO 8178-1:2006(E). Sartorius SE2-F ultramicro scale was used for filter weighing.

Particle size distribution and number concentration were measured using three instruments that together covered a wide particle mobility size range. A particle size magnifier (PSM, Airmodus Inc.) together with a condensation particle counter (CPC 3775, TSI Inc.) was used to measure size range 1.7–7.2 nm. A scanning mobility particle sizer (Nano-SMPS [21]) equipped with DMA 3085 and UCPC 3025 (TSI Inc.) was used in size range 3–60 nm.

Table 1
The specifications of the engine modes.

	Higher torque	Lower torque
Test engine torque	60 N m	35 N m
Test engine speed	2700 rpm	3100 rpm
Residual O_2	6%	6%
Combustion air consumption	100 kg/h	95 kg/h
Hydrocarbon additions	No	Yes

Please cite this article in press as: Alanen J et al. The formation and physical properties of the particle emissions from a natural gas engine. Fuel (2015), http://dx.doi.org/10.1016/j.fuel.2015.09.003