

Development of an Ammonia Reduction Aftertreatment Systems for Stoichiometric Natural Gas Engines

Saroj Pradhan, Arvind Thiruvengadam, Pragalath Thiruvengadam, Berk Demircok, Marc Besch, Daniel Carder, and Bharadwaj Sathiamoorthy
West Virginia University

ABSTRACT

Three-way catalyst equipped stoichiometric natural gas vehicles have proven to be an effective alternative fuel strategy that has shown superior low NOx benefits in comparison to diesels equipped with SCR. However, recent studies have shown the TWC activity to contribute to high levels of tailpipe ammonia emissions. Although a non-regulated pollutant, ammonia is a potent pre-cursor to ambient secondary PM formation. Ammonia (NH₃) is an inevitable catalytic byproduct of TWC during that results also corresponds to lowest NOx emissions.

The main objective of the study is to develop a passive SCR based NH₃ reduction strategy that results in an overall reduction of NH₃ as well as NOx emissions from a stoichiometric spark ignited natural gas engine. The study investigated the characteristics of Fe-based and Cu-based zeolite SCR catalysts in storage, and desorption of ammonia at high exhaust temperature conditions, that are typical of stoichiometric natural gas engines. Results of the investigation showed that both, the Fe- and Cu-zeolite SCR adsorbed above 90% of TWC generated NH₃ emissions below 350-400°C SCR temperatures. Desorption or slipping of NH₃ was mostly observed at exhaust gas temperatures exceeding 400°C. In terms of NOx conversions, for a same cell density substrate, Fe-zeolite showed efficiency between 60-90% above temperatures of 300-350°C while Cuzeolite performed well at lower SCR temperature from 300°C below with conversion efficiency of greater than 50%.

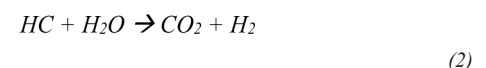
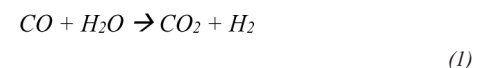
In order to efficiently reduce both NOx and NH₃ simultaneously over longer dynamic duty cycles it was found that an active regeneration strategy for the passive SCR system must be developed. To this extent, the study extended its objectives to develop an engine control strategy that results in stoichiometric ammonia production operation followed by brief lean operation to regenerate SCR brick using high NOx slip through TWC.

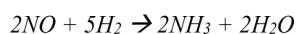
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INTRODUCTION

Considering the available technologies that meet the current stringent USEPA emissions standards for heavy-duty vehicles, natural gas fueled vehicles utilizing a TWC converter operating under stoichiometric conditions provide superior NOx benefits compared to SCR equipped diesels [1]. Monetary incentives and local regulations have increased the population of NGV in captive fleets such as transit buses and refuse trucks. The operating bandwidth of a TWC to maximize the simultaneous reduction of NOx, CO and THC is achieved by changing the operating air-fuel ratio of the engine between rich and lean by a process called as dithering [2]. The dithering strategy is sequenced between a narrow range of stoichiometric window. Under slightly lean operation, the catalyst formulation is able to oxidize HC and CO, and under slight rich condition reduces NOx into harmless products such as water (H₂O), carbon dioxide (CO₂) and nitrogen (N₂) [3].

However, studies have shown that under such rich operating conditions oxygen deficiency in the exhaust favors a water gas shift reaction and a steam reforming given by [Equations 1](#) and [Equation 2](#) reactions [4]. And, in such events, CO's which are not oxidized (due to lack of oxygen) reacts with water vapor to form CO₂ and hydrogen (H₂). Under suitable conditions the H₂ molecules further reacts with available NO and CO to form ammonia (NH₃) given by [equation 3](#) [5], a secondary unregulated emission known to be detected in both gasoline and natural gas vehicles equipped with a TWC aftertreatment systems [5] [6] [7] [8].





(3)

Gas phase NH_3 is considered to be a critical atmospheric pollutant contributing to the formation of airborne particulate matter [9]. These secondary formed particulate matter along with the atmospheric NH_3 have adverse health effects [5] and impact terrestrial vegetation [10].

Various studies carried out by WVU has indicated increasing trends in NH_3 emissions with higher vehicle miles. Figure 1 presents work-specific NO_x and NH_3 emissions along with the odometer readings consolidated from five different natural gas vehicles from a recent WVU chassis and on-road findings [7]. The first three vehicles (Veh 1, 2 and 3) represent goods movement vehicles and (Veh 4 and 5) represents transit bus vehicles. The results shown were conducted over Urban Dynamometer Drive Schedule (UDDS) chassis drive cycle including one from an in-use testing for the transit bus (Veh 5) as provided by the last two bars in the chart below. Although not conclusive due to limited data, the results do indicate an increase in NH_3 emission as a function of vehicle age and could be contributed to many different factors, one being the aging of the oxygen sensor causing sluggish response [7]. Additionally, degradation of a TWC over time have shown to negatively impact the catalyst performance. Researchers have shown that aging of catalyst are caused due to chemical and/or thermal aging mechanisms [11], and tend to impact oxidation rates [12], (CO, HC, & NO) conversion efficiencies, and light-off temperature [13].

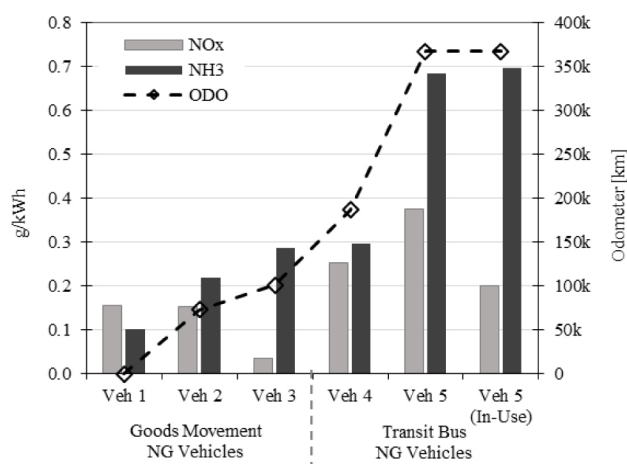


Figure 1. Work-specific NO_x and NH_3 emissions from natural gas vehicles

Elevated levels of NH_3 emission from HD NGV is a major concern due to its potency to form secondary PM. Also the substantial penetration of natural gas-fueled vocational vehicles within the urban cities of US, could potentially increase secondary PM concentration in urban, highly populated locations [14]. Although the market share for natural gas-fueled vehicles are significantly low when compared to commercial heavy-duty diesel-fueled, a 11.9% average annual growth rate is projected between 2011 and 2040 [1].

Multiple studies in the past have demonstrated coupled aftertreatment (TWC and SCR) system primarily for lean-burn gasoline applications to further achieve better fuel economy and satisfy emission standards [15] [16] [17]. Majority of these studies show similar pathway in which an SCR catalyst is employed as a passive ammonia storage

system that uses the NO_x slip from TWC during less frequent rich operation as a source to regenerate the stored ammonia. Likewise, WVU is investigating into pathways of a similar passive SCR system as a potential retrofit approach for abatement of both NH_3 and NO_x emissions from a stoichiometric NG engines.

Application of SCR technologies has been a suitable solution for modern-heavy-duty diesel application in controlling tailpipe NO_x emissions, however exhaust temperature becomes a key challenge in maintaining overall performance of the SCR depending on the type of catalyst used [18]. Catalyst formulation plays an important role in performance over different temperature range. For example, Cuzeolite based SCRs have operating range between (150-450°C) whereas Fe-zeolite based SCRs have between (350-600°C) [19]. Natural gas vehicles tend to operate at high temperatures, and from a recent on-road study conducted by WVU on a 2013 CNG vehicle show exhaust temperatures as high as 600°C measured right after the TWC [20]. This study looks in investigating the characteristics and effects of different catalyst formulation on NH_3 storage and NO_x conversion efficiency of a SCR system installed after an aged TWC and operated over dynamic loading of a typical natural gas engine.

As a further extension to this study, WVU is also working on developing SCR models NH_3 storage and adaptive decision making controllers which will then be utilized under the engine controls for precise air-fuel ratio patterns between rich mode (NH_3 production mode) and lean mode (NH_3 regeneration mode). Also, leading to a lean burn NG pathway using TWC as an on-board ammonia production system and eventually increasing the operating range of a stoichiometric NG engines for enhanced fuel economy. As catalyst aging viewed as huge performance factor for the TWC, a retrofitted passive SCR system with TWC with an on-board Ammonia Reduction Catalyst (ARC) controller can effectively lower the NO_x profile of NG through its useful.

EXPERIMENTAL SETUP

All measurements of the study presented herein were conducted at the Engine and Emission Research Laboratory (EERL) at West Virginia University. The EERL is part of West Virginia University's Center for Alternative Fuels, Engines and Emissions (CAFEE), and the transient engine dynamometer test cell is designed and operated according to recommendations outlined in the Code of Federal Regulations (CFR), Title 40, Part 1065.

Table 1. Test Engine Specification

Manufacturer	Cummins
Model/Year	ISL-G280/2008
Capacity	8.9L
Rated Power	209kW @ 2200RPM
Engine Hours	354k km (13,800 hrs)
Aftertreatment System	Three-way Catalyst (TWC)

An 8.9L natural gas engine equipped with its original TWC aftertreatment system was used for the transient dynamometer testing. The specification of the engine are presented in Table 1. The engine