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The Effect of Diesel Exhaust Fluid Dosing on Tailpipe Particle Number Emissions

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Michael A. Robinson, Jacob Backhaus, Ryan Foley, and Z. Gerald Liu
Cummins Emission Solutions

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Abstract

Introduction of modern diesel aftertreatment, primarily selective catalytic reduction (SCR) designed to reduced NO_x, has increased the presence of urea decomposition byproducts, mainly ammonia, in the aftertreatment system. This increase in ammonia has been shown to lead to particle formation in the aftertreatment system. In this study, a state of the art diesel exhaust fluid (DEF)-SCR system was investigated in order to determine the influence of DEF dosing on solid particle count. Post diesel particulate filter (DPF) particle count (> 23 nm) is shown to increase by over 400% during the World Harmonized Transient Cycle (WHTC) due to DEF dosing. This increase in tailpipe particle count warranted a detailed parametric study of DEF dosing parameters effect on tailpipe particle count. Global ammonia to NO_x ratio, DEF droplet residence time, and SCR catalyst inlet temperature were found to be significant factors in post-DPF DEF based particle formation. Thermogravimetric analysis (TGA) of ammonia salt particles and urea decomposition byproducts indicate significant chance of measurement using the Particle Measurement Programme (PMP) Particle Number (PN) method. These DEF based particles were not intended to be addressed by the PMP PN methodology, but are found to be over 80% of PN post DPF.

Introduction

Potential environmental impacts and human health risks [1, 2, 3, 4, 5] associated with diesel PM and NO_x have resulted in stricter emission regulations, requiring the introduction of advanced aftertreatment systems. In Europe, heavy-duty emission standards introduced in 2005 have reduced diesel PM mass emissions by an order of magnitude, from 0.1 g/kW-hr to ~0.02 g/kW-hr. In addition to this tightening of mass emission standards, a new PM standard was introduced in Europe, regulating particle number (PN) in addition to PM mass. Extensive documentation is available on the Particle Measurement Programme (PMP), which defines the method in which the solid fraction of particles are counted for this new PN regulation [6, 7]. These tightening regulations have required the introduction of wall flow diesel particulate filters (DPFs), which effectively remove 99% of PM mass and some of volatile and semi-volatile species associated with PM [8, 9]. In addition to changes in PM regulations, NO_x emission standards have decreased rapidly over the last 20 years.

US and European heavy-duty emission regulations successfully reduced NO_x emission limits by an order of magnitude in 2010, requiring the introduction of selective catalytic reduction (SCR) systems utilizing a 32.5% urea water solution also known as diesel exhaust fluid (DEF). SCR technology was first used in stationary applications, typically with pure gas phase ammonia (NH_3), due to its high conversion efficiency of NO_x to N_2 and H_2O [10]. Diesel emission control application of SCR requires injection of DEF in the exhaust system, which thermally decomposes to isocyanic acid (HNCO) and NH_3 before passing over the SCR catalyst [11]. Control of the DEF injection system is not a trivial task, especially over challenging transient certification cycles requiring greater than 95% NO_x conversion (deNO$_x$) efficiency [12].

Stoichiometric injection of NH_3 (ammonia to NO_x ratio (ANR) = 1.0) is required to prevent excessive NO_x emissions (ANR < 1.0) or NH_3 slip (ANR > 1.0) downstream of the SCR catalyst. Due to difficulties in DEF dosing rate control, prediction of DEF droplet evaporation, and prediction of SCR catalyst NH_3 storage, balancing deNO$_x$ performance and NH_3 slip over transient cycles is still a technical challenge. Introduction of additional NO_x or NH_3 sensors have allowed for closed loop SCR control, but common architectures still rely on above stoichiometric ANR to maintain high deNO$_x$, requiring an ammonia oxidation catalyst (AMOX) to reduce tailpipe NH_3 slip.

The presence of excess NH_3 in these architectures has been shown to unintentionally contribute significantly to PM emissions. Retrofit of advanced aftertreatment systems on diesel vehicles using ultra low sulfur diesel fuel has shown to increase the concentration of nucleation mode particles [13]. Herner et al. report an order of magnitude increase of nucleation mode (~10 nm) particles at a

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