Applied Energy 205 (2017) 964–975

Contents lists available at ScienceDirect

Applied Energy

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

Characterization of the urea-water spray impingement in diesel selective catalytic reduction systems

Yujun Liao[a], Panayotis Dimopoulos Eggenschwiler[a,*], Daniel Rentsch[b], Francesco Curto[a], Konstantinos Boulouchos[c]

[a] Automotive Powertrain Technologies Laboratory, Empa, Swiss Federal Laboratories for Materials Science and Technology, CH-8600 Dübendorf, Switzerland
[b] Functional Polymers Laboratory, Empa, Swiss Federal Laboratories for Materials Science and Technology, CH-8600 Dübendorf, Switzerland
[c] Aerothermochemistry and Combustion Systems Laboratory, ETH Zurich, CH-8092 Zürich, Switzerland

HIGHLIGHTS

- The spray wall impingement is significant for low pressure sprays.
- The gas flow entrains or evaporates mainly fine droplets below 90 µm.
- Impinging liquid undergoes local boiling and forms a film, where urea partly crystalizes.
- Liquid accumulation leads to solid deposit formation under all conditions investigated.
- NMR analysis reveals cyanuric acid as the major solid deposit component.

ARTICLE INFO

Keywords:
Selective catalytic reduction NOx
Urea water solution spray impingement
Solid deposits formation
Optical diagnostics
Nuclear magnetic resonance spectroscopy

ABSTRACT

Exhaust after-treatment selective catalytic reduction (SCR) systems based on urea-water solution are state-of-the-art technologies mitigating NOx emissions for diesel and lean combustion systems. Major challenges for implementing the systems are high NOx reduction performance, uniform mixture formation and solid deposits formation. This study presents a detailed analysis of the urea-water spray wall impingement and influences on reducing agent distribution and deposit formation, thus system performance. High speed images provide detailed information of the impingement process. Moreover, impinging spray mass flux distribution and droplet size distribution have been quantified under typical diesel exhaust flow conditions. The work has been performed with a commercial 3-Hole pressure-driven injector dosing into a flow channel.

Under all tested conditions, the impingement is significant. At gas flow conditions of 300 °C, 200 kg/h, 35.6% of the injected fluid impinges on the opposed wall due to entrainment and evaporation. The entrainment level has been found to scale logarithmically with the gas flow momentum, related correlations are provided. Having an integrated analysis of a mechanical patternator and non-intrusive Phase Doppler Anemometry (PDA) results, it is concluded that droplets below 20 µm are completely entrained or evaporated. The impingement rate is gradually increasing with increasing droplet diameter up to 90 µm, while almost all larger droplets reach the opposed wall. High speed imaging shows in detail liquid film formation, film transport, liquid accumulation, nucleate boiling, urea crystallization and melting, as well as thin film evaporation prior to solid deposit formation. The spray impingement leads to liquid film formation. Relevant dimensions have been evaluated with digital image processing. Direct relationship of liquid accumulation to the solid deposit formation has been identified. The permanent solid deposit is consisting of cyanuric acid, biuret, ammelide and ammeline as identified by nuclear magnetic resonance spectroscopy.

1. Introduction

Diesel engines relying on lean combustion are widely used in the transportation sector due to their high energy conversion efficiency. Concerning heavy duty applications, freight transport and mobile machines, there are no potential alternatives to diesel engines in the next decades. However, they produce harmful pollutants, such as NOx and particles. NOx is one of the major and most harmful environmental

* Corresponding author at: Panayotis Dimopoulos Eggenschwiler, Empa, Ueberlandstrasse 129, 8600 Duebendorf, Switzerland.
E-mail address: Panayotis.Dimopoulos@empa.ch (P. Dimopoulos Eggenschwiler).

http://dx.doi.org/10.1016/j.apenergy.2017.08.088
Received 4 March 2017; Received in revised form 11 May 2017; Accepted 11 August 2017
Available online 20 August 2017

pollutants induced by the transportation sector. They lead to the acid rain and formation of ground level ozone, which leads to health and environmental problems. Methods to reduce NOx emissions include in-cylinder techniques and exhaust after-treatment systems. In-cylinder techniques which aim to lower peak temperatures tend to decrease engine efficiency and increase soot formation. Therefore, exhaust selective catalytic reduction (SCR) systems are currently the promising technique to reduce NOx emissions without sacrificing engine efficiency [1]. SCR systems rely on injection of ammonia precursor in the exhaust gas. In real applications, 32.5% urea-water solution (UWS, trademark AdBlue) is used as an ammonia precursor because of its non-toxicity and lowest freezing point. The UWS injection is followed by the evaporation of water from spray droplets. Thereafter, thermal decomposition of urea and hydrolysis of isocyanic acid take place [1–3], resulting in the production of ammonia. Ammonia reacts with NOx to water and nitrogen.

The main challenges for the implementation of mobile urea-SCR systems include homogeneous distribution of urea [4–6] as well as mitigation of solid deposit formation [7–9]. Because of compact design requirements of the exhaust pipe and relatively long time scales of urea thermal decomposition [10], the spray impingement on the exhaust pipe or on a mixer is unavoidable [2]. As the wall temperature drops below a certain threshold, liquid film starts forming [2]. Evaporation from the wall film leads to further cooling and increasing risk of deposit formation such as cyanuric acid, biuret, and ammelide [3,11–13].

Numerical simulation efforts concerning SCR sprays [2,14–22] are more established in the literature. However, they deliberately point out that there is a lack of experimental data in this field. Only recently a limited number of experimental data on SCR sprays has been available [10,23–28]. SCR pressure-driven injectors operated at low pressures don't allow secondary breakup when injected into the elevated cross flow [23] and produce relatively large droplets [10,22,28]. Therefore, spray wall impingement occurs and results in a spray cooling flux of several MW/m^2 during the injection duration [29]. Due to the spray cooling effect, liquid film formation occurs below a critical wall temperature as visualized by [14,24]. However, the fundamental aspects of the spray wall impingement and the phenomenology of the deposit formation process are not thoroughly reported. Meanwhile, several application oriented papers have clearly pointed out detrimental effects of solid deposits in real applications [7,9,30,31].

In the field of UWS spray impingement, two kinds of studies exist in the literature: single droplet evaporation/impact [24,32,33] or numerical simulation of the spray/wall impact [2,14,34]. Multi-stage evaporation behavior of UWS droplets have been observed [32,33]. The first stage is featured by water evaporation following the D^2 law, thereafter micro-explosion results in more complex behavior. Numerical simulation efforts [2,14] have implemented a regime map into the modelling of the urea spray impingement based on the kinetic properties of droplets and the wall temperature. They have described different modes of single droplet wall impact. To our knowledge so far, no results have been reported concerning the macro properties of droplet impact as a whole spray.

On the other hand, the spray impingement has been studied in diverse applications, like fuel spray impingement [35–37], steel quenching [38] and electronics cooling [39]. Previous studies have indicated that, the impingement flux has the highest influence on the spray impingement heat transfer [38,40–42]. Typical optical techniques, Mie scattering [23] or backlight imaging [24], having described the spray patterns while impacting, are not able to provide quantification of the impinging spray mass load on the wall. This is decisive for the spray wall heat transfer and the following deposit formation.

The present paper is a systematical investigation of the UWS spray impingement process under diesel-typical conditions. The impingement process is described both qualitatively and quantitatively. The impinging spray mass flux distributions were quantified by using a mechanical patternator. Phase Doppler Anemometry (PDA) was applied to characterize the impinging droplet size distributions under various conditions. Optical visualization experiments were conducted to observe the phenomena associated with the impingement process, such as liquid film formation, urea crystallization and deposit formation. Solid deposits were analyzed by nuclear magnetic resonance spectroscopy. Results of this study provide new insights into the UWS spray impingement process which can be used for the lay-out of efficient after-treatment systems, and offer ample possibilities for validating numerical simulations.

2. Experimental setup and methods

Measurements were conducted in a flow lab, which was designed for the experimental investigation of SCR systems as described in detail [10,28]. In the present work, a commercial SCR injector was mounted on the top of the measurement chamber, 50° inclined to the gas flow direction. It was a pressure-driven injector with three 190 μm nozzle holes arranged evenly on a 1.9 mm diameter ring. The injection pressure was regulated to 9 bar.

Optical film visualization experiments were performed to study the phenomena associated with spray impingement on a plate under various exhaust-typical conditions. The stainless steel plate, having an averaged surface roughness of 0.5 μm, was placed 14 mm (measured from the front surface) above the channel bottom as shown in Fig. 1. The impinged plate had a dimension of 200 mm × 80 mm and a thickness of 4 mm. The impingement processes were captured by a high speed CMOS camera (Casio EX-FH100) working at 30 frames per second at a resolution of 640 × 480 pixels. The camera was mounted 352 mm vertically upwards of the impinged plate. The illumination was provided by a LED lamp covered by a milk glass to diffuse light. For all visualization experiments, the injection frequency was set to 1 Hz, while two different injection durations were studied: 60 ms and 120 ms. The total number of injections was limited to 150. Adblue (32.5% urea-water solution) was used as the injection fluid.

Image processing was conducted in Matlab for determining the liquid film area. RGB images were firstly converted to grayscale images. The spatial intensity distribution of the grayscale images was corrected by subtracting background image (one image before injection) as shown in Fig. 2b. Median filter (5 × 5) was applied to the corrected images to remove the small features on the plate. The processed images were binarized by a threshold value, thus separating the wetted area from the plate (Fig. 2d). Thereafter, every connected region was attributed to a label and only the pixels at the front cone region were kept (Fig. 2e and f).

Patternator is an in-house developed instrument for measuring spray mass flux distributions. A perforated plate with 24 probes, each of which is connected to a PVC bottle through a silicon hose, was pressed against the channel tightly. A vacuum pump is linked to all collecting bottles through a distributor to create a slight suction for avoiding capillary effects in the hoses. The 24 probes were arranged in three rows of 8 probes each as shown in Fig. 3a. The probe matrix has a spacing distance of 10 mm in the flow direction and 15 mm in the transversal direction. Each probe has an inner diameter of 5 mm. To refine the measurement grid, the flow channel was traversed on a two-axis sliding system, while the patternator was fixed on the working table. In this manner, a resolution of 2 mm × 2 mm was obtained. Each data point was averaged over 1700 injections. The injection strategy was set to 3 Hz of 150 ms to speed up the measurement. The injected fluid is demineralized water to avoid blockages in the hoses or probes. The mass flux displayed in results section is calculated as follows:

$$\dot{m} = \frac{m}{n t_{inj} A} \tag{0}$$

where $\dot{m}$ is the mass flux, m the total mass collected per probe, n the number of injections collected, t_{inj} the duration of each injection and A the probe tip opening area.