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Thermo fluid effect of the urea thermal decomposition in a lab-scaled reactor



Department of Mechanical Engineering, Kyungpook National University, 80 Daehak-ro Buk-gu, Daegu, Republic of Korea

HIGHLIGHTS

• Conversion efficiency of reducing agent is investigated under 300 °C.

• Conversion efficiency is closely related with the thermo-fluid dynamics.

• NH₃ conversion was larger than that of HNCO unlike a theoretical urea thermolysis.

• Conversion difference of NH₃ and HNCO is affected by a convective heat transfer.

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ABSTRACT

Urea decomposition in the urea selective catalytic reduction (urea-SCR) system is achieved under a multiphase flow field coexisting with the injected urea solution droplets in the liquid phase, the inflow gas in the gas phase, and solid urea particles. For this reason, urea decomposition is closely connected with thermo-fluid dynamics as well as various chemical reactions. An experimental study was performed to investigate urea decomposition under various thermo-fluid conditions, with different temperatures and velocities of inflow gas, and urea solution quantities. The inflow gas conditions were similar to the exhaust conditions of a large marine engine. A urea solution injector of twin fluid type was designed, as well as a lab-scale exhaust system to control the temperature and velocity of the inflow gas. The spray performance of the designed urea solution injector was almost identical under all the experimental conditions used, so the spray's effect on the urea decomposition was minimized. The total conversion efficiency considered with both NH₃ and HNCO was closely related to the urea injection quantity and convective heat transfer of the inflow gas as well as the temperature of the inflow gas. The conversion efficiency of NH₃ was larger than that of HNCO under all experimental conditions, unlike for the theoretical thermolysis reaction. The difference between the conversion efficiency of NH₃ and HNCO was affected by convective heat transfer and urea injection quantity as well as the temperature of the inflow gas.

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1. Introduction

In selective catalytic reduction (SCR) technology, urea solution is widely used as an NH_3 storage compound because it is inexpensive and nontoxic. The urea solution injected into exhaust gas is evaporated and decomposed according to reactions (1) and (2) to yield the actual reducing agent NH_3 [1]:

Urea thermolysis : $CO(NH_2)_2 \rightarrow NH_3(g) + HNCO(g)$ (1)

 Great developments in urea-SCR technology have been made in the field of diesel cars, where its demand is relatively high. De Rudder [2,3] reported that the de-NOx efficiency with the urea-SCR system is associated with the interaction between the evaporation of urea solution, decomposition, and mixing with exhaust gas. Sivanandi [4] asserted that a wire-mesh mixer enhances the thermolysis of urea into NH₃ and isocyanic acid (HNCO). Birkhold et al. [5,6] reported the conversion rate of the reducing agent in diesel car exhaust pipes determined through numerical simulation. Dong et al. [7] described the formation of deposits during urea decomposition being affected by the spray quality of the injected urea solution. Studies on the control algorithm of the urea-SCR system have been conducted [8,9]. In addition, work on methods to increase the de-NOx efficiency in the SCR systems of diesel vehicles is in progress [10–16]. Moreover, many results on urea decomposition have





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^{*} Corresponding author. Tel.: +82 53 950 6570; fax: +82 53 950 6550. *E-mail address:* jghong70@knu.ac.kr (J.G. Hong).

been reported by chemical researchers, as in urea-SCR, the aqueous urea solution injected directly into the exhaust pipe yields the reducing agent through urea thermolysis and isocyanic acid hydrolysis. Koebel et al. [17] reported the relative amounts of urea, NH₃, and HNCO at the catalyst entrance according to the exhaust gas temperature. Yim et al. [18] found that the hydrolysis of HNCO is possible even in the absence of catalyst, although the rate was slow when the reaction temperature was sufficiently high (above 400 °C). The evaporation and decomposition of particles containing aqueous urea solution was investigated by Kontin et al. [19]. Urea decomposition and deposit formation have been investigated with micro reactors [20-23]. Fang et al. [24] asserted that deposit formation decreases catalytic performance not only by consuming part of the NH₃ produced during urea decomposition, but also by degrading the structural and thermal properties of the catalyst surface. Recently, there has been growing interest in the urea-SCR system for large marine ships because of the regulation of the International Maritime Organization (IMO) about NOx reduction [25]. In large marine engines, a turbocharger is used to enhance the engine power in general. The exhaust gas temperature of a large marine engine drops significantly through the turbocharger. Unfortunately, the urea-SCR system installation is restricted in front of the turbocharger because of its performance. For this reason, it is recommended that the urea-SCR system is installed after the turbocharger, even though the exhaust temperature is relatively low. Moreover, the quantity of NOx in the emissions is much larger than in those of a diesel car. Therefore, it is difficult to apply research on the urea-SCR system with a focus on the diesel car to that for large marine engines, because the exhaust gas temperature and NOx quantity differ significantly. Moreover, many studies on urea decomposition have been focused on the chemical reaction. Urea decomposition in the urea-SCR system is generated under a multiphase flow field coexisting with the injected urea solution droplets in the liquid phase, the inflow gas in the gas phase, and solid urea particles. Thus, it is thought that the urea decomposition is closely connected with thermo-fluid dynamics as well as with various chemical reactions. The purpose of this study is to investigate the urea decomposition related to thermo-fluid dynamics under experimental conditions similar to the exhaust gas conditions of a large marine engine. The lab-scale exhaust system and urea solution injector were designed for this study, and the conversion efficiencies of NH₃ and HNCO were measured under various thermo-fluid conditions by using this experimental apparatus. In this work, it was found that the urea decomposition is closely related to the urea injection quantity and convective heat transfer of the inflow gas as well as the temperature of the inflow gas.

2. Experimental setup and method

A schematic diagram of the experimental setup for investigating the spray performance of the urea solution injector is shown in Fig. 1. The experimental setup consists of a fluid supply system, a visualization system, and a measurement system. The spray experiment was conducted under atmospheric conditions. The fluid supply system consisted of a liquid pump (Longer Precision Pump, BT50-1J), which supplied the urea solution from the storage tank to the injector, and an air-assist supply device for atomization. The urea solution quantity was controlled by the number of revolutions per minute (RPM) of the liquid pump. The air compressor and regulator were used for making the pressurized assist air, and the volume flow rate of the assist air supplied to the injector was controlled by the needle valve that was positioned after the air regulator. The volume flow rate of the assist air was measured by an air flow meter (SMC, PF2A750). The air flow meter has a



Fig. 1. Schematic diagram of the experimental setup for measurement of spray characteristics.

margin of error of $\pm 5\%$ FS. The spray images were captured by using a CCD camera (Vieworks, VM-2M 35) with both a Nd:YAG laser (New wave, Solo II) and short-duration light (V-tek, LS-201) as the light source. A micro lens (Nikon, AF micro 105 mm) was used to take close-up pictures of the droplets. The macroscopic characteristics of the injected urea solution were analyzed from the spray images. The Sauter mean diameter (SMD) and the cumulative volume percent of droplets for the urea solution quantity were measured by using a technique based on that reported by Suzuki et al. [26,27].

Fig. 2 illustrates the experimental setup for measuring the urea decomposition. The modeled exhaust pipe (66 mm of diameter, insulated) was designed to investigate the urea decomposition. The urea solution injector was installed in parallel to the inflow gas in the center of the modeled exhaust pipe. A multicomponent FTIR gas analyzer (Gasmet, DX-4000) was used to analyze the urea decomposition. The FTIR spectra, calibrated to an ammonia concentration of 1000 ppm before the experiment, showed five significant peaks: one at approximately 3300 cm⁻¹, one at approximately 1600 cm⁻¹, and three in the region of 900–1150 cm⁻¹. The most intense peak (in the region of 910–1150 cm⁻¹) was chosen for ammonia quantification. The FTIR spectra for HNCO at 500 ppm showed three significant peaks: one at approximately 3500 cm^{-1} and two in the region of $2000-2300 \text{ cm}^{-1}$. The peak in the region of 2000-2240 cm⁻¹ was chosen for HNCO quantification. The sampling part of the FTIR gas analyzer was purged



Fig. 2. Schematic diagram of the experimental setup for measurement of urea decomposition.

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