



Full Length Article

Effect of oxygen and additives on thermal decomposition of aqueous urea solution

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ARTICLE INFO

Article history:

Received 12 June 2015

Received in revised form 1 March 2016

Accepted 5 April 2016

Keywords:

Aqueous urea solution

Thermal decomposition

Oxygen concentration

Na₂CO₃NaNO₃

ABSTRACT

The thermal decomposition of aqueous urea solution was studied in a lab-scale experimental system. The experimental research was focused on the effect of oxygen concentration and different additives on NH₃ yields, N₂O production, and CO concentration. Without oxygen or additives, the NH₃ yields increased gradually as the temperature increased from 473 K to 923 K and remained approximately 60% in the temperature range of 923–1073 K. Oxygen promoted the thermal decomposition process and enhanced the NH₃ yields in the temperature range of 473–773 K, but it brought about the oxidation of NH₃ and led to significant decline of the NH₃ yields when the reaction temperature was above 823 K. The addition of Na₂CO₃ promoted the NH₃ yields obviously, but it did not restrain the NH₃ oxidation at high temperatures. Furthermore, Na₂CO₃ reduced the production of N₂O and CO at 623–1073 K. Similar to Na₂CO₃, the addition of NaNO₃ also increased the NH₃ yields and decreased the N₂O and CO concentration.

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1. Introduction

Due to the combustion of fossil fuels in boilers and furnaces, nitrogen oxides (NO_x) emission has been a seriously environmental problem for decades [1]. NO_x results in photochemical smog, acid rain, and greenhouse effect, so NO_x control technologies including selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR) have been developed to reduce the pollutants within the stringent limit of emission regulations [2,3].

Various N-based reducing agents have been extensively studied and applied in SCR and SNCR processes with the most popular being ammonia (NH₃) [4,5]. In the earlier days, aqueous ammonia and anhydrous ammonia were commonly supplied as feedstock for thermal denitrification (DeNO_x) process, but they presented significant danger to human health as hazardous chemicals [6]. The production, transportation, storage and handling of aqueous ammonia and anhydrous ammonia trigger serious safety and environmental regulatory requirements for risk management plans, emergency response plans and release analysis [7]. Therefore, the application of aqueous urea solution (AUS) (NO_xOUT process) instead of ammonia has been investigated and applied since 1980 due to its

virtual nontoxicity as well as simple storage and handling properties [8–11].

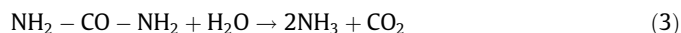
As reported in literature [12–14], during NO_xOUT process urea is first thermally decomposed into ammonia (NH₃) and isocyanic acid (HNCO) according to (1).



HNCO formed in reaction (1) then reacts with water and produce NH₃ and carbon dioxide (CO₂) according to (2).



By addition of reactions (1) and (2), the well-known overall reaction is obtained:



For reasons of simplicity, AUS is usually injected into the hot exhaust directly. However, the simple injection technique suffers from short residence time, leading to incomplete decomposition of AUS and causing a significant waste [15]. Moreover, N₂O and CO are easily generated when AUS is directly injected into a SNCR reactor [16–18]. Therefore, the technique of injecting AUS into a thermal decomposition reactor out of the main exhaust stream was developed. It allows much longer residence time for AUS decomposition and has attracted a lot of attention of industrialists and researchers [19–22].

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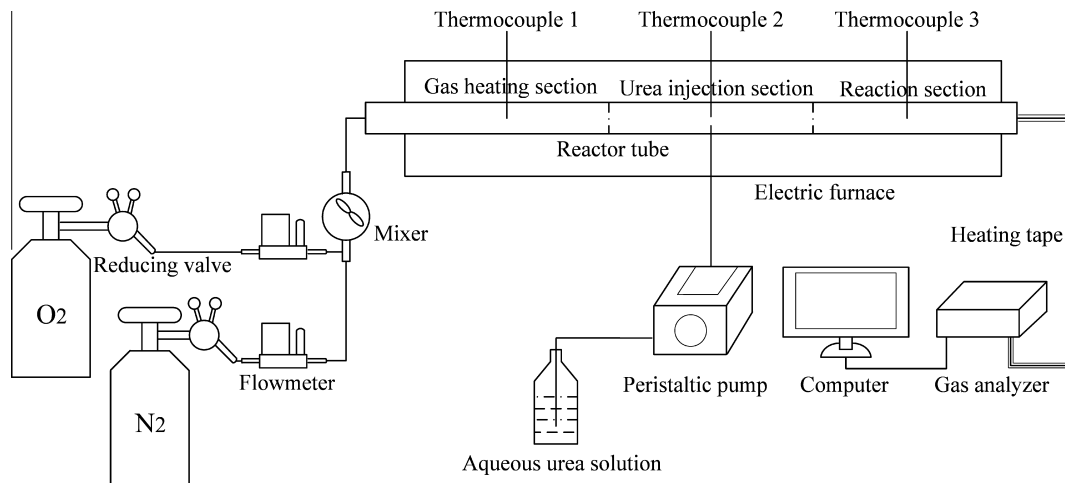


Fig. 1. Experimental setup.

Table 1
Feed rate and component concentration in the experimental cases.

Case	Solution (mL/min)	O ₂ (mL/min)	Urea (mol/L)	Na ₂ CO ₃ (mol/L)	NaNO ₃ (mol/L)
1	0.16	0	0.176	0	0
2	0.16	105	0.176	0	0
3	0.16	210	0.176	0	0
4	0.16	0	0.176	0.006	0
5	0.16	105	0.176	0.006	0
6	0.16	210	0.176	0.006	0
7	0.16	0	0.176	0	0.006
8	0.16	105	0.176	0	0.006
9	0.16	210	0.176	0	0.006

Note: The concentration of urea, Na₂CO₃, and NaNO₃ in the table stands for the molar concentration in the aqueous solution.

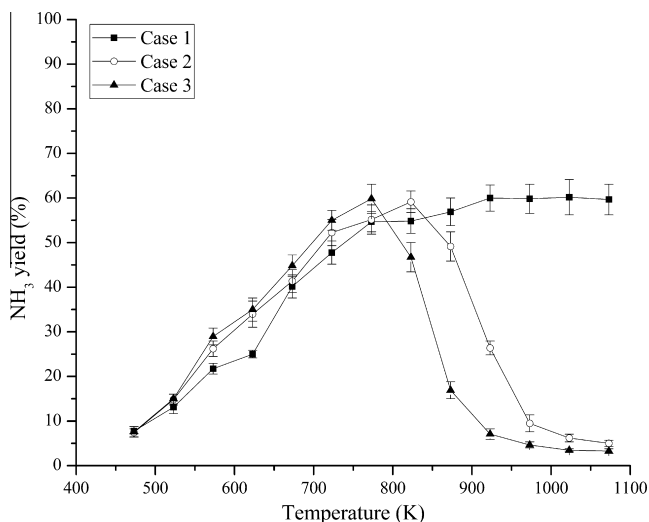


Fig. 2. NH₃ yields versus temperature of Cases 1, 2, and 3.

In the two steps of thermal decomposition of AUS, reaction (2) is rather slow because HNCO formed in reaction (1) is quite stable in the gaseous phase [13,23]. To promote reaction (2) and improve the NH₃ yields, a number of researchers have tested different types of materials to explore a high catalytic one for the thermal decomposition [24–26]. Lundström et al. [27] investigated the catalytic

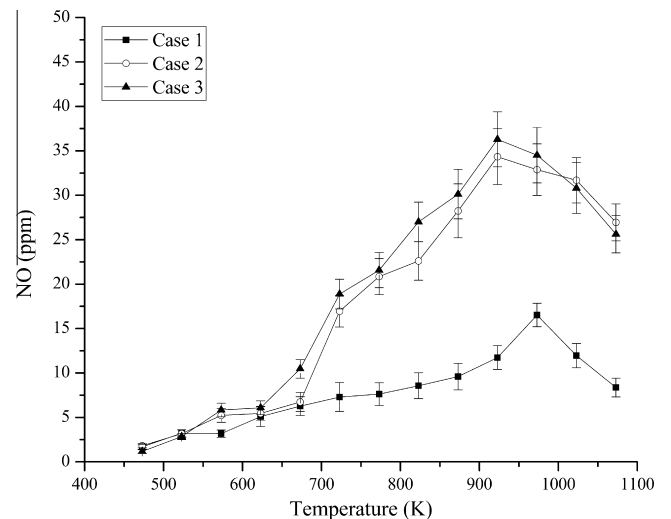


Fig. 3. NO concentration versus temperature of Cases 1, 2, and 3.

effect of titanium dioxide, Fe-Beta, and Alumina on AUS decomposition, and the experimental results showed that TiO₂ exhibited the best catalytic performance. Bernhard et al. [28] compared water-free catalytic urea thermolysis with catalytic thermal decomposition of AUS by experiments, and they concluded the order of hydrolysis activities: ZrO₂ > TiO₂ > Al₂O₃ > H-ZSM-5 > SiO₂ and the order of thermolysis activities: TiO₂ > H-ZSM-5 ≈ Al₂O₃ > ZrO₂ > SiO₂. Hauck et al. [29] explored the kinetics and surface chemistry of the HNCO hydrolysis with water on TiO₂ and the influence of other molecules present in the reactant stream such as NH₃, NO and NO₂ on this catalytic chemistry.

Although some catalysts performed effectively in increasing NH₃ yields [24–29], they seem not quite suitable for the SNCR process, where no catalysts are wanted. Actually, to enhance the NO removal in the SNCR process, various additives have been studied by experiments and by kinetic modeling [10,30,31]. Zamansky et al. [32] studied the effect of small amounts of sodium salts, and they found parts per million levels of sodium compounds could not only enhance NO removal but also broaden and deepen the effective SNCR temperature range. Guo et al. [33] developed and validated a reduced mechanism simplified from a detailed chemical kinetics mechanism, and the simulation results coincided qualitatively with the experimental data in an entrained flow reactor.

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