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Self-sustained catalytic combustion of carbon monoxide ignited by dielectric barrier discharge

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Abstract

This paper presents the results of a study of self-sustained catalytic combustion of CO ignited by dielectric barrier discharge (DBD) using $\text{Ce}_{0.5}\text{Zr}_{0.5}\text{O}_y/\text{TiO}_2$ (CeZr/Ti), $\text{CuZr}_{0.25}\text{O}_y/\text{TiO}_2$ (CuZr/Ti), and $\text{CuCe}_{0.75}\text{Zr}_{0.25}\text{O}_y/\text{TiO}_2$ (CuCeZr/Ti) catalysts. DBD excites and dissociates some of the reactant molecules in the gas phase. These are more easily adsorbed on the catalyst surface than are ground-state species, therefore induction begins at a lower background temperature than in thermal catalysis. CO is adsorbed on copper sites, therefore CeZr/Ti is inactive in CO ignition, but CuZr/Ti and CuCeZr/Ti achieve DBD ignition at 34 and 44 s, respectively, at a specific energy density (SED) of 1500 J/L. CO catalytic ignition by DBD involves two steps. The induction process is dominated by plasma catalysis. At the same SEDs, induction with CuCeZr/Ti begins earlier than those with CuZr/Ti, in good agreement with the reducibilities and oxygen-transfer properties of these catalysts. The ignition process is governed by thermal catalysis because the enhancement of external diffusion induced by increasing the temperature improves the reaction rate. CuZr/Ti provided more CO adsorption sites than did CuCeZr/Ti, contributing to shortening of the ignition delay. © 2016 by The Combustion Institute. Published by Elsevier Inc.

Keywords: Self-sustained catalytic combustion; Carbon monoxide; Ignition; Dielectric barrier discharge

1. Introduction

The off-gas produced during basic oxygen furnace steelmaking contains a considerable amount

of CO. Because a high concentration of oxygen in combination with CO is liable to cause explosions, the off-gas generated at steelmaking intermissions is not directly recovered as a fuel, and is often discharged into the atmosphere via conventional flare burners [1]. Our new method for using this energy in CO catalytic oxidation is based on methane self-sustained catalytic combustion as an alternative route for electric power generation. In this process, a mixture of methane and O_2 generates

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local hot spots on the catalyst surface, and then adjacent methane and O₂ molecules are dissociated, leading to a thermochemical runaway reaction, followed by self-sustained non-flame burning [2]. Such a combustion strategy is suitable for portable electricity generation, because of its high combustion efficiency in small-scale devices.

Dielectric barrier discharge (DBD)-assisted ignition and oxidation processes have recently drawn attention because they give rapid start-up under cold conditions [3]. This involves strong coupling of electron-induced chemistry and thermochemistry. Discharge produces electronic excitation, dissociation, and ionization of molecules, followed by plasma chemical reactions, and the background temperature increase not only induces thermochemical reactions, but also affects the plasma physics. Zhang et al. [4] identified the contributions of the effects of plasma chemistry and thermochemistry, and confirmed that the reactant conversions depend on both two processes by independently controlling the temperature and the reactor pressure, whereas the consecutive reactions depend mainly on the temperature and the corresponding thermochemistry. Mintusov et al. [5] reported that the temperature increase in air–C₂H₄ flows is caused by exothermic plasma chemical oxidation, which results in ignition when the flow temperature approaches the auto-ignition temperature. Jo et al. [6] found that the ratio of the plasma power to the heating input power is an important parameter for enhancing methane oxidation. The results can be characterized by limitedness, however, due to their methodology of a high-temperature plasma source and a narrow temperature range. Although many reports have linked plasma-assisted ignition of air–fuel flows, no statistical evidence of plasma-assisted catalytic ignition to achieve self-sustained combustion has yet been presented.

Our previous study showed that CuCe_{0.75}Zr_{0.25}O_y mixed oxides formed on ZSM-5 substrates show excellent catalytic activities because copper–ceria synergism improves the redox capability and the ceria–zirconium solid solution has a good oxygen storage capacity [7]. In this work, we achieved CO self-sustained combustion ignited by DBD for the first time, using CuCe_{0.75}Zr_{0.25}O_y/TiO₂ (CuCeZr/Ti), CuZr_{0.25}O_y/TiO₂ (CuZr/Ti), and Ce_{0.75}Zr_{0.25}O_y/TiO₂ (CeZr/Ti) catalysts with Cu:Ce:Zr molar ratios of 1:0.75:0.25. Inert TiO₂ was used as the substrate because of its excellent thermal stability and good dispersion of active species. The promotion of active phase dispersion, oxygen mobility, and redox properties caused by strong interactions among copper, cerium, and zirconium species will be discussed. We distinguished between the effects of electron-induced chemistry and thermochemistry by using DBD and temperature-programmed ignition (TPI), combined with various characterization techniques. The results obtained will be helpful

in introducing the use of plasma-assisted ignition for self-sustained catalytic combustion of CO in steelmaking off-gas.

2. Experimental specifications

2.1. Synthesis of catalysts

The TiO₂ support was pure anatase TiO₂ (Degussa). The CuCeZr/Ti, CuZr/Ti, and CeZr/Ti catalysts were prepared using an incipient wetness impregnation method. The TiO₂ gel support was impregnated with an aqueous solution containing appropriate amounts of copper, zirconium, and cerium nitrates. The catalysts were dried for 8 h at 120 °C and calcined in air for 6 h at 600 °C. The dried samples were crushed and sieved to 10–12 mesh. The copper content of the CuCeZr/Ti catalyst was fixed at 4 wt% and the molar ratio of Cu:Ce:Zr was 1:0.75:0.25. For the CuZr/Ti and CeZr/Ti catalysts, the molar ratios of Cu:Zr and Ce:Zr were 1:0.25 and 0.75:0.25, respectively.

2.2. Characterization

X-ray photoelectron spectroscopy (XPS) was performed using a Kratos Axis Ultra DLD spectrometer with a Mg K α X-ray source. Temperature-programmed H₂ reduction (H₂-TPR) experiments were performed using a Micromeritics 2920 II instrument with 5% H₂/Ar at a heating rate of 15 °C/min to 600 °C. Prior to reduction, the sample (ca. 200 mg) was pretreated in flowing Ar at 300 °C for 1 h. Temperature-programmed O₂ desorption (O₂-TPD) was also performed with the Micromeritics 2920 II instrument using 200 mg of sample. After O₂ (30 mL/min) saturation for 1 h, the gas was switched to Ar for 0.5 h, and TPD was performed by ramping the temperature at 10 °C/min to 600 °C in Ar (30 mL/min). Desorption of O₂ was detected using a thermal conductivity detector. In situ diffuse-reflectance infrared Fourier-transform (DRIFT) spectroscopy was performed using a Bruker Tensor 27 spectrophotometer. Self-supporting disks prepared from the sample powders were suspended in a quartz holder and then mounted in an IR cell (Tianjin XianQuan Industry and Trade Development CO., Ltd.) connected to a vacuum line. The disks were heated at 300 °C in N₂ for 1 h and evacuated to a pressure of 3 × 10⁻³ mbar for 1 h. The IR CO adsorption measurements were performed at CO pressures of about 20 Torr with the samples at room temperature. After exposure to CO, IR spectra were recorded at 30 s intervals until the spectrum was stable.

2.3. Experimental setup

Figure 1 shows a schematic diagram of the experimental setup. The system consisted of a continuous flow gas-supply system, electric furnace,

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