Preparation of Silica-Coated ZnO with Oxygen Vacancy to Enhance the Performance of Photocatalytic Methane Oxidation

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Methane has a significantly higher global warming potential compared to carbon dioxide. Consequently, converting methane to carbon dioxide through oxidation reactions represents an effective strategy for mitigating global warming. However, due to the low reactivity of methane, this reaction typically requires high-temperature conditions over 200 °C. The application of photocatalysis to methane oxidation has been investigated, and among various photocatalytic materials, ZnO has been studied as a photocatalyst for methane oxidation due to its low cost and superior performance compared to other photocatalysts including TiO_2.[1] However, its poor thermal stability has significantly hindered the mechanistic studies and applications of ZnO. In this study, ZnO@SiO_2 core-shell photocatalysts with high thermal stability were synthesized, and the photocatalytic performance was investigated.

The ZnO@SiO_2 core-shell photocatalysts (ZnO@SiO_2) were synthesized by two-step processes. First, ZnO nanoparticles were prepared by thermal decomposition of zinc oxalate dihydrate at 350 °C. Then, the prepared ZnO nanoparticles were coated with SiO_2 via hydrolysis of tetraethyl orthosilicate.

The particle growth during the heat treatment at 600 °C was significantly suppressed with the SiO_2 coating while severe aggregation proceeded in pristine ZnO from 11.1 to 49.8 nm, which were evaluated by the XRD line width. Moreover, the high-temperature treatment increased the absorption in the visible range in ZnO@SiO_2 and ZnO, which can be attributed to the defects in ZnO. Figure 1 shows the results of the photocatalytic methane oxidation using a fixed bed flow system. The bare ZnO before and after the heat treatment exhibited very low photocatalytic activity for methane oxidation under visible light irradiation. In contrast, the ZnO@SiO_2 photocatalyst showed a higher methane conversion under visible light. This high activity is likely to be achieved by simultaneously suppressing particle growth and introducing defects.

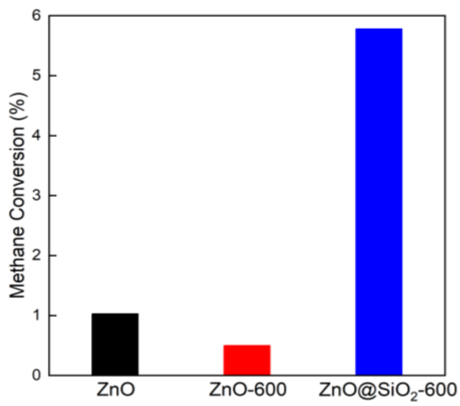

Fig. 1 Methane conversion in photocatalytic methane oxidation on ZnO, ZnO treated at 600 °C (ZnO-600), and ZnO@SiO_2 treated at 600 °C (ZnO@SiO_2-600) under visible light irradiation.

1) X. Chen et al., *Nat. Commun.*, **2016**, *7*, 12273.