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Accelerated photocatalytic CO_2 reduction using ruthenium-doped nickel–ZrO_2 photocatalyst

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1. Introduction

Photocatalytic conversion of CO_2 into fuels and raw materials is one of the options to create new carbon-neutral cycle. We previously reported Ni–ZrO_2 photocatalyst to convert $^{13}CO_2$ and H_2 into $^{13}CH_4$ at the rate of 0.98 mmol h^{-1} g_{cat}^{-1} [1]. Tuned photocatalyst Ag–ZrO_2 also selectively produced raw material CO [2]. This study reports accelerated CH_4 formation using ruthenium-doped Ni–ZrO_2 photocatalyst.

2. Experimental section

Ru-doped Ni–ZrO_2 photocatalyst was prepared via liquid phase reduction from Ru chloride, Ni chloride hexahydrated, and ZrO_2 by mixing with $NaBH_4$. The obtained sample is denoted as Ni–Ru–ZrO_2.

Ruthenium K-edge XAFS spectra were measured in the transmission mode at the Photon Factory Advanced Ring, High Energy Accelerator Research Organization (Tsukuba, Japan) on the NW10A beamline using a Si(3 1 1) monochromator, a Pt-coated mirror, and a piezo transducer.

The obtained Ru K-edge XAFS data were analyzed using the XDAP software package version 3.2.9 [3]. Multiple-shell curve-fit analyses were performed with the data obtained on the EXAFS using the theoretical amplitude and phase shift functions calculated using a multiple scattering code FEFF version 8.4 [4].

3. Results and discussion

Using CO_2 (2.3 kPa) and H_2 (22 kPa), the photocatalytic formation rate of CH_4 reached the maximum using Ni (10 wt %)–Ru (1.0 wt %)–ZrO_2 at the rate of 5.6 mmol h^{-1} g_{cat}^{-1}. The Fourier transform of the angular wave number k^3-weighted Ru K-edge EXAFS for the most active photocatalyst is depicted in Figure 1.

The curve fit to the data provided fit parameters of interatomic pair of Ru and Ni appeared at 0.175–0.244 nm (phase shift uncorrected, Figure 1): coordination number N = 3.6, interatomic distance R = 0.2446 nm, and Debye–Waller factor σ = 0.00521 nm, indicating the formation of dispersed atomic Ru atom at the surface of Ni nanoparticles.

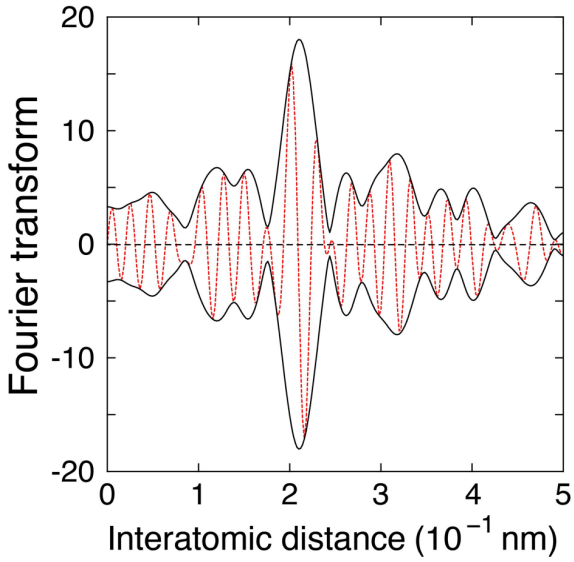

Figure 1. Fourier transform of the angular wave number k^3-weighted Ru K-edge EXAFS χ-function for the fresh Ni (10 wt %)–Ru (1.0 wt %)–ZrO_2 photocatalyst.

References

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