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Co K-edge EXAFS analysis of cobalt-based catalyst

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Introduction

When hydrocarbons were produced from a feed containing CO and H_2 using a cobalt-based catalyst (Co catalyst) in a pilot scale plant, the CO conversion rate decreased over the course of the reaction.

In this study, we investigated the chemical states and structures of the active species cobalt on the spent Co catalysts from the pilot scale plant by way of Co K-edge EXAFS measurements in order to determine the cause of the decrease in CO conversion over the course of the reaction.

Experimental

For the Co K-edge EXAFS measurements, we prepared spent Co catalysts which had been used for different times on stream in the pilot scale plant. Before the EXAFS measurements, the spent catalysts were packed in vacuum-sealed polyethylene bags to prevent exposure to the ambient air. The EXAFS measurements were conducted in transmission mode on station NW10A at the Photon Factory Advanced Ring (PF-AR).

Results and Discussion

The CO conversion rate of the Co catalysts used in the pilot scale plant decreased as the time on stream increased (Fig. 1). To determine why, we analyzed the Co species on the catalysts using Co K-edge EXAFS.

The Co K-edge EXAFS Fourier transforms (FTs) of the

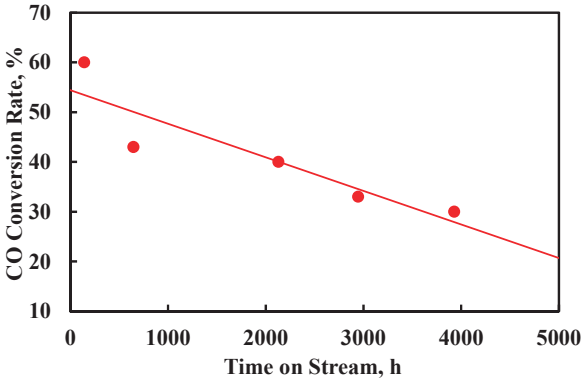

Fig. 1: Relationship between time on stream and CO conversion rate

spent Co catalysts obtained at different times on stream are shown in Fig. 2. Co-Co peaks (originating from the active species, Co metal) were observed in the FTs of the Co K-edge EXAFS for all catalysts, regardless of time on stream. Furthermore, the Co-Co peak intensities varied depending on the time on stream. We thus focused on the Co-Co peak intensities of the EXAFS FTs of the spent Co catalysts.

As a result, it was found that the Co-Co peak intensity in the FT of the EXAFS increased as the time on stream increased (Fig. 3). This indicated that the coordination number of the Co metal on the spent catalyst increased or possible aggregation of the Co metal particles as the time on stream increased.

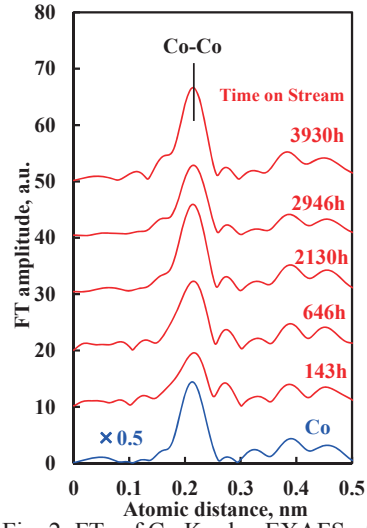

Fig. 2: FTs of Co K-edge EXAFS of Co spent catalysts.

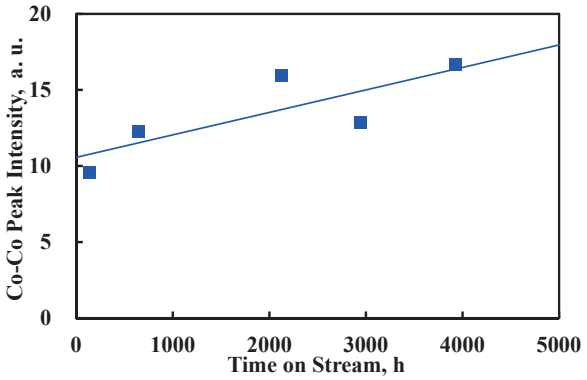

Fig. 3: Relationship between time on stream and Co-Co peak intensity

Therefore, the Co K-edge EXAFS measurements suggested that aggregation of the Co metal particles on the spent Co catalysts might be one cause of the decrease in the CO conversion rate over the course of the reaction in the pilot scale plant.

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