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One-step Formation of Acetone from Propene and Water on Ultrastable Y-type Zeolite Catalyst

Hiroshi MORI,+ Hisashi UENO, Noritaka MIZUNO,
Hidenori YAHIRO, and Masakazu IWAMOTO*
Catalysis Research Center, Hokkaido University, Sapporo 060
+Department of Industrial Chemistry, Miyakonojo National
College of Technology, Miyakonojo 885

Proton exchanged ultrastable Y-type zeolite showed a pronounced catalytic activity for the formation of acetone directly from propene and water. The formation rate of acetone was higher or comparable to those on MoO_3-based catalysts in the oxidation of propene by oxygen to produce acetone.

Zeolites catalyze a wide variety of reactions.1) Little is known of the application of zeolite to the formation of ketones directly from olefins and water through the combination of hydration and dehydrogenation.2) Here, we wish to report that the proton exchanged ultrastable Y-type zeolite efficiently catalyzed the formation of acetone directly from propene and water without oxygen, while other zeolites were much less active.

The parent zeolites, ZSM-5, ferrierite (denoted as F), and ultrastable Y-type (dealuminated by steaming, denoted as USY) zeolites were supplied by Tosoh Corporation and mordenite (denoted as M) and Y-type were obtained from the Catalysis Society of Japan. Proton exchanged zeolites were prepared as described previously.3) The reaction of propene and water or the dehydrogenation of 2-propanol was carried out in a conventional flow reactor below 433 K at an atmospheric pressure using 1 g of catalysts. The flow rates of propene and water vapor were 5.8 and 11.6 $cm^3 \cdot min^{-1}$, respectively, or those of 2-propanol and water vapor were 0.3 and 11.3 $cm^3 \cdot min^{-1}$, respectively. Total flow rate was 30 $cm^3 \cdot min^{-1}$ (N_2 balance). The reactants and products were analyzed by gas chromatography. Approximately steady formation of acetone and/or 2-propanol, which were main products, was attained after 8 h in the reaction of propene and water.

Catalytic activities are summarized in Fig. 1. H-USY showed the highest activity among the catalysts listed in Fig. 1. The activities of H-M and H-F zeolites were very low, though they were reported to be very active for the hydration of lower olefins.3,4) The activity for the dehydrogenation of 2-propanol was examined separately and found to increase with the increase of the activity for the formation of acetone from propene and water. The H_2 evolution was qualitatively confirmed by gas chromatography in the reaction of propene and water and in the dehydrogenation of 2-propanol. The formation of basic anion such as $Al(OH)_4^-$ or $AlO_2^- \cdot 2H_2O$ on zeolite by steaming has been reported.5) Therefore, acetone would be formed through the combination of the hydration of propene on acidic sites and the dehydrogenation of resulting 2-propanol on basic sites.

The addition of oxygen (10%) slightly increased the formation rate of acetone and little changed the selectivities to acetone. Moro-oka et al.6) and Ogasawara et al.7) have reported that MoO_3-based catalysts

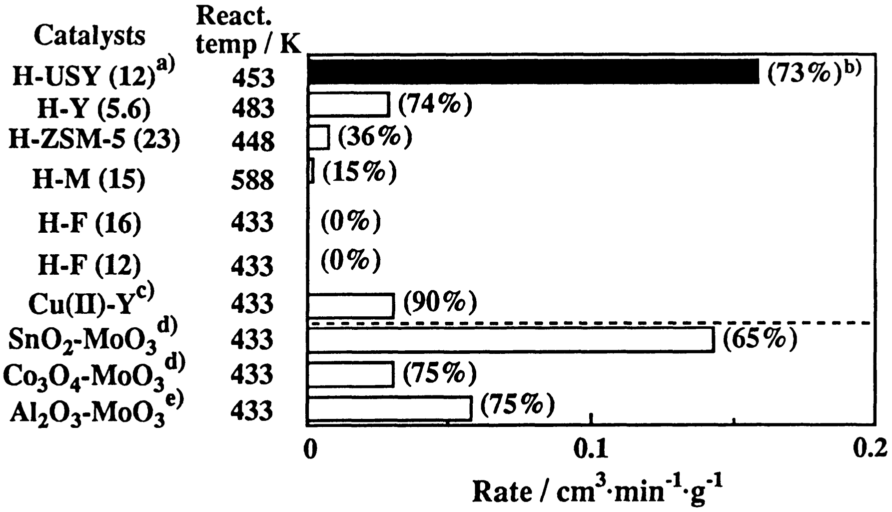

Fig. 1. Rates of acetone formation over various catalysts.
a) Figures in parentheses were SiO_2/Al_2O_3 ratios. b) Figures in parentheses were selectivities to acetone. c-e) Cited from refs. (2), (6), and (7), respectively. The partial pressures of propene and water on SnO_2-MoO_3 and Co_3O_4-MoO_3 were similar to those in the present work. Reactions on SnO_2-MoO_3, Co_3O_4-MoO_3, and Al_2O_3-MoO_3 were carried out with oxygen. The rate of Cu(II)-Y was estimated assuming that flow rate of propene is 10 $cm^3 \cdot min^{-1}$.

such as SnO_2-MoO_3 and Co_3O_4-MoO_3, and Al_2O_3-MoO_3 were active for the oxidation of propene by oxygen to produce acetone. Figure 1 reveals that H-USY has the activity for the formation of acetone comparable to or higher than the MoO_3-based catalysts even in the absence of oxygen and the selectivity to acetone (73%) was nearly equal to those of the MoO_3-based catalysts.

In conclusion, the novel route for the ketonization of olefins on H-USY, which was different from the oxidation of olefins by oxygen on MoO_3-based catalysts, was found.

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References

1) J. A. Rabo and G. J. Gajda, *Catal. Rev. Sci. Eng.*, **31**, 385(1989-90).
2) I. Mochida, S. Hayata, A. Kato, and T. Seiyama, *Bull. Chem. Soc. Jpn.*, **44**, 2282(1971).
3) M. Iwamoto, M. Tajima, and S. Kagawa, *J. Catal.*, **101**, 195(1986).
4) M. Iwamoto, H. Mori, M. Tajima, and S. Kagawa, *Nippon Kagaku Kaishi*, **1989**, 586; M. Iwamoto, H. Mori, M. Tajima, and S. Kagawa, " Proceedings of the International Symposium on Acid-Base Catalysis, Sapporo, 1988," ed by K. Tanabe, H. Hattori, T. Yamaguchi, T. Tanaka, Kodansha, Tokyo(1989), p. 453; E. Kikuchi, T. Matuda, K. Shimomura, K. Kawahara, and Y. Morita, " Proceedings of the 7th International Conference of Zeolites, Tokyo, 1986," ed by Y. Murakami, A. Iijima, J. W. Ward, Kodansha, Tokyo(1986), p. 771.
5) D. R. Corbin, R. D. Farlee, and G. D. Stucky, *Inorg. Chem.*, **23**, 2920(1984).
6) S. Tan, Y. Moro-oka, and A. Ozaki, *J. Catal.*, **17**, 132(1970); Y. Moro-oka, Y. Takita, and A. Ozaki, *ibid.*, **23**, 183(1971).
7) S. Ogasawara, S. Takahashi, A. Fukai, and Y. Nakada, *Kogyo Kagaku Zasshi*, **72**, 2244(1969).

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