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Oxidation of Isobutane to Methacrolein over $Ga_2O_3/Bi_2Mo_3O_{12}$ Catalysts

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Catalytic performance and the surface character of the Ga_2O_3 supported Bi-Mo complex oxides were studied to achieve direct formation of methacrolein from isobutane. $Bi_2Mo_3O_{12}$ (α phase) and $Bi_2Mo_1O_6$ (γ phase) showed higher catalytic activity than $Bi_2Mo_2O_9$ (β phase) for isobutane partial oxidation. Supporting Ga_2O_3, which is an active catalyst for dehydrogenation of hydrocarbons, onto the oxides, enhanced the catalytic activity.

The optimum amount of supported Ga_2O_3 on $Bi_2Mo_3O_{12}$ was about 3 wt% for methacrolein formation. In the presence of oxygen, a remarkable amounts of hydrogen over Ga_2O_3 during the isobutane oxidation but no hydrogen was formed over Ga_2O_3/ $Bi_2Mo_3O_{12}$. It is confirmed from TPR that Ga_2O_3 and $Bi_2Mo_3O_{12}$ were not reduced until 550°C but the reduction of $Ga_2O_3/Bi_2Mo_3O_{12}$ started at 350-380°C. The on-set temperature in TPR of the Bi-Mo complex oxides decreased to 350-380°C from 500°C by the supporting Ga_2O_3 onto the oxides, and the catalysts after TPR measurement are composed of BiO, Bi, and MoO_2 in addition to $Bi_2Mo_3O_{12}$. These results suggest that the hydrogen spillover took place over supported catalyst.

$Ga_2O_3/Bi_2Mo_3O_{12}$ catalyst showed higher activity and high selectivity for methacrolein at 450°C. The improvement in the selectivity for methacrolein of the $Ga_2O_3/Bi_2Mo_3O_{12}$ may

be explained as following. Isobutane is adsorbed on the surface of Ga_2O_3 to form hydrogen atom and *t*-butyl fragment and both formed species migrates to $Bi_2Mo_3O_{12}$ surface. Migrated hydrogen may modify the $Bi_2Mo_3O_{12}$ surface property by the reaction with oxide ions, which is active for the deep oxidation resulting in high selectivity for methacrolein.
In the non-aerobic oxidation of isobutane over the $Ga_2O_3/Bi_2Mo_3O_{12}$ catalyst, the formation rate of CO_x significantly reduced, and methacrolein and isobutene were selectively obtained when the reduction degree of the catalyst was lower than 0.3% at 450° C.

Keywords: Isobutane oxidation, Methacrolein synthesis, Bismuth molybdate catalyst, Gallium oxide, Hydrogen spillover, Nonaerobic oxidation

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