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Effect of catalyst composition - (In, Fe, Co) oxides based on Al_2O_3 and ZSM-5 on their activity in propane dehydrogenation with CO_2, N_2O

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Propylene is the most important olefin with an annual production of roughly 8×10^7 t. A great deal of attention has been paid to the on-purpose propylene production technology, such as propane dehydrogenation in the presence of mild oxidants CO_2 and N_2O (PODH-CO_2, N_2O), due to their potential to make-up the shortfall of propylene supply left by conventional steam cracking of hydrocarbons where propylene is produced as a byproduct of ethylene [1].

The paper presents results on the effect of support and active component nature as well as the preparation method of oxide catalysts on their activity (C_3H_8 conversion, selectivity to C_3H_6) in the oxidative dehydrogenation of propane to propylene: In_2O_3-Al_2O_3 (YSZ) for PODH-CO_2 and Fe_2O_3 (Co_2O_3)/H-ZSM-5 for PODH-N_2O.

Data on porous structure of alumina based catalysts and their activity in the PODH-CO_2 are listed in the table below. Hydrothermal treatment of the catalyst In_2O_3-Al_2O_3(HT) synthesized by coprecipitation of indium and aluminum nitrates leads to increase of specific surface (S_{BET}) mesoporous structure, that results to higher propylene selectivity (S_{C3H6}) with the same propane conversion (X_{C3H8}) as compared to the sample coprecipitated without HT – In_2O_3-Al_2O_3 and the catalysts prepared by mechanical mixing of indium and aluminum nitrates In_2O_3-Al_2O_3 (MM) and by impregnation of alumina with indium nitrate In_2O_3/Al_2O_3. The mesopores improve the catalysts activity due to the better transport of the reactant and product molecules in PODH-CO_2 [2].

Porous structure (N_2 adsorption) and activity of In_2O_3-Al_2O_3 catalysts

Catalyst	S_{BET}, m^2/g	Total pore volume, cm^3/g	Micropore volume, cm^3/g	Mesopore diameter, nm	X_{C3H8}, %	S_{C3H6}, %
In_2O_3-Al_2O_3 (HT)	176	0.49	0.0012	12.385	8	51
In_2O_3-Al_2O_3	142	0.3034	0.0026	12.476	11	25
In_2O_3-Al_2O_3 (MM)	72	0.1055	0.00017	5.4416	13	26
In_2O_3/Al_2O_3	83	0.21	0.048	7.1	11	32

The catalyst supported on Y- stabilized zirconia In_2O_3/YSZ (S_{BET} = 50 м²/г) exhibits higher C_3H_8 conversion (23%) and the same selectivity to propylene (32%) in the PODH-CO_2 compared to In_2O_3/Al_2O_3. This could be attributed to the higher activity of zirconia support compared to alumina as was shown for ethane dehydrogenation to ethylene with CO_2 [3].

In PODH-N_2O over Fe_2O_3/H-ZSM5 catalyst the higher activity (X_{C3H8} = 53%, S_{C3H6} = 21%) is achieved at the temperature 400 °C. In the presence of Co_2O_3/H-ZSM5 catalyst the X_{C3H8} = 55% and S_{C3H6} = 31% are achieved at the higher temperature, 600 °C.

Thus, the activity of the catalysts in PODH-CO_2, N_2O depends on the nature of both the support (Al_2O_3, YSZ) and the active component (Fe_2O_3, Co_2O_3).

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