

# 负载型 $ZrO_2$ 催化苯甲醛 Meerwein-Ponndorf-Verley 反应中的载体效应

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**摘要** 采用浸渍法制备了 Si-MCM-41 和 Al-MCM-41 (Si/Al = 50) 介孔分子筛,  $SiO_2$ ,  $\gamma-Al_2O_3$  及 MgO 等负载的  $ZrO_2$  催化剂, 考察了其在以异丙醇为氢源苯甲醛 Meerwein-Ponndorf-Verley (MPV) 还原反应中的催化活性, 并与纯  $ZrO_2$  的催化活性进行对比。同时, 采用 X 射线衍射、N<sub>2</sub> 吸脱附法、X 射线光电子能谱、紫外-可见漫反射光谱和吡啶原位吸附红外光谱等手段表征了催化剂。结果表明,  $ZrO_2$  负载于 Si-MCM-41, Al-MCM-41 和  $SiO_2$  后, 催化活性明显提高, 这归因于  $ZrO_2$  与载体间存在强相互作用形成 Zr-O-Si 键, 使催化剂表面 Zr-OH 数量显著增多, Lewis 酸中心强度增强, 并出现 Brønsted 酸中心, 三种催化剂的活性高低次序是 5% $ZrO_2$ /Si-MCM-41 > 5% $ZrO_2$ /Al-MCM-41 > 5% $ZrO_2$ / $SiO_2$ 。而 5% $ZrO_2$ / $Al_2O_3$  和 5% $ZrO_2$ /MgO 基本无催化活性, 可归因为  $ZrO_2$  与  $\gamma-Al_2O_3$  的弱相互作用使 5% $ZrO_2$ / $Al_2O_3$  的酸性与  $\gamma-Al_2O_3$  类似,  $ZrO_2$  与 MgO 的强相互作用使 5% $ZrO_2$ /MgO 基本无酸性。

**关键词:** 载体效应 Meerwein-Ponndorf-Verley 反应 氧化锆 MCM-41 介孔分子筛 苯甲醛

**Abstract:** A series of zirconia catalysts supported on Si-MCM-41 and Al-doped MCM-41 (Si/Al = 50) mesoporous molecular sieves, silica,  $\gamma-Al_2O_3$ , and MgO were prepared by the wet impregnation method. The catalytic activities of these materials in the Meerwein-Ponndorf-Verley reduction (MPV) of benzaldehyde with 2-propanol as reducing agent were investigated, and compared to that of hydrous zirconia. The materials were characterized by X-ray diffraction, nitrogen adsorption-desorption, X-ray photoelectron spectroscopy, UV-Vis diffuse reflectance spectroscopy, and Fourier transform infrared and thermal desorption of pyridine. Loading zirconia on Si-MCM-41, Al-MCM-41, and  $SiO_2$  gave improved catalytic activity. This is attributed to a strong interaction of zirconia with the support to form Si-O-Zr bonds, which gave a significant increase in the amount of exposed Zr-OH groups and stronger Lewis acidity as well as an appearance of Brønsted acid sites. The activity of 5% $ZrO_2$ /Si-MCM-41 was the highest, followed by those of 5% $ZrO_2$ /Al-MCM-41 and 5% $ZrO_2$ / $SiO_2$ . However, 5% $ZrO_2$ / $Al_2O_3$  and 5% $ZrO_2$ /MgO gave very low catalytic activities. This is ascribed to that the acidities of 5% $ZrO_2$ / $Al_2O_3$  and the  $\gamma-Al_2O_3$  support were similar due to the weak interaction of zirconia with  $\gamma-Al_2O_3$ , and 5% $ZrO_2$ /MgO had no acidity because of the strong interaction between zirconia and MgO.

**Keywords:** support effect, Meerwein-Ponndorf-Verley reduction, zirconia, MCM-41 mesoporous molecular sieve, benzaldehyde

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