

H-MCM-22 沸石分子筛中 Brønsted/Lewis 酸协同效应的 ^1H 和 ^{27}Al 双量子魔角旋转固体核磁共振研究

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摘要 采用各种固体核磁共振 (NMR) 技术详细研究了 H-MCM-22 分子筛中 Brønsted/Lewis 酸的协同效应. 二维 ^1H 双量子魔角旋转 (DQ-MAS) NMR 结果表明, 在脱铝 H-MCM-22 分子筛中 Brønsted 酸位 (骨架桥式羟基) 和 Lewis 酸位 (非骨架铝羟基) 之间是空间邻近的, 暗示着可能存在 B/L 酸协同效应. 二维 ^{27}Al DQ-MAS NMR 结果揭示了各种铝物种之间的空间邻近性, 表明 B/L 酸协同效应优先发生在 H-MCM-22 分子筛超笼中的骨架 T6 位铝和非骨架铝物种之间. 2-13C-丙酮探针分子实验发现, 因 B/L 酸协同效应而导致脱铝 H-MCM-22 分子筛酸性明显增强, 氘代吡啶探针分子实验也证实了在 H-MCM-22 分子筛的超笼中发生了 B/L 酸协同效应. 上述结果将有助于我们理解在脱铝 H-MCM-22 分子筛上发生的多相催化机理.

关键词: H-MCM-22 分子筛 脱铝 酸性 Brønsted/Lewis 酸协同效应 双量子魔角旋转核磁共振

Abstract: Brønsted/Lewis acid sites synergy in H-MCM-22 zeolite was studied by solid-state nuclear magnetic resonance (NMR). Two-dimensional ^1H double quantum-magic angle spinning (DQ-MAS) NMR revealed the details of the spatial relationship between the Lewis and the Brønsted acid sites in a dealuminated H-MCM-22 zeolite, which implied the existence of a Brønsted/Lewis acid sites synergy. Two-dimensional ^{27}Al DQ-MAS NMR was used to give the details of the spatial proximities of various aluminum species. The Brønsted/Lewis acid sites synergy occurred in the supercage of the H-MCM-22 zeolite between a T_6 site Al and extra-framework Al species. ^{13}C CP/MAS NMR of adsorbed acetone demonstrated that the spatial proximities of the Brønsted and Lewis acid sites led to a synergy that enhanced the Brønsted acid strength of the dealuminated zeolites. ^1H MAS NMR of adsorbed deuterated pyridine confirmed that the Brønsted/Lewis acid sites synergy occurred in the supercage of H-MCM-22. This finding is important for understanding the mechanism of acid-catalyzed reactions on H-MCM-22 zeolites.

Keywords: H-MCM-22 zeolite, dealumination, acidity, Brønsted/Lewis acid synergy, double quantum-magic angle spinning nuclear magnetic resonance

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