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MoO_3-SnO_2催化剂上二甲醚低温氧化高选择性制备甲酸甲酯

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Low-temperature oxidation of dimethyl ether to methyl formate with high select catalysts

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摘要 定向设计并制备了多功能MoO_3-SnO_2催化剂,在常压连续流动固定床反应器上实现了二甲醚低温氧化高选择性制备甲酯的过程。考察了机械混合法、共沉淀法及沉淀浸渍法等不同制备方法对催化剂性能的影响。在沉淀浸渍法制备的MoO_3-S催化剂上,常压、160℃反应条件下,甲酸甲酯选择性达94.1%,DME转化率也达到了33.9%,并且产物中无CO_x生成。采用NTPD、CO_2-TPD及H_2-TPR对催化剂进行了表征,结果表明,表面酸性、碱性及氧化性的不同是造成催化剂反应性能差异的原另外,通过采用XRD、Raman及TEM对催化剂结构进行表征发现,晶粒粒径及金属氧化物MoO_3的存在状态等结构的差异是造化剂活性不同的主要原因。较小晶粒的催化剂和表面存在的低聚态MoO_3是致使催化剂活性提高的主要原因。

关键词: 二甲醚　低温　选择氧化　甲酸甲酯　氧化锡　氧化钼

Abstract: Low-temperature oxidation of dimethyl ether (DME) to methyl formate (MF) with high selecti was realized in a continuous flow fixed-bed reactor over the multifunctional MoO_3-SnO_2 catalysts design and prepared intentionally. The effect of the preparation methods including mechanical mixing, co-precipitation and co-precipitation-impregnation on the catalyst activity was investigated. The results sh that the selectivity to MF reaches 94.1% at 160℃ over the catalyst prepared by co-precipitation-impregnation, with DME conversion of 33.9% and absence of CO_x in the products. The results of NH_3-TPD CO_2-TPD and H_2-TPR characterizations indicated that the catalysts prepared by various methods are als obviously different in the surface acidic, alkaline and redox properties. The results of Raman, XRD and TE revealed that MoO_3 structure and particle sizes have a significant influence on the catalyst activity; sma particle size and oligomeric MoO_3 may be responsible for the high activity of the MoO_3-SnO_2 catalysts fr co-precipitation-impregnation in the selective oxidation of DME to MF at low temperature.

Key words: dimethyl ether　low-temperature　selective oxidation　methyl formate　MoO_3　SnO_2

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