

A DFT study on CO oxidative coupling to dimethyl oxalate over Pd-Me (Me=Cu, Al, Ag) catalysts¹

Bingying Han^a, Lixia Ling^{*b}, Maohong Fan^c, Ping Liu^d, Riguang Zhang^a, Baojun Wang^{*a}

^a Key Laboratory of Coal Science and Technology of Ministry of Education and Shanxi Province, Taiyuan University of Technology, Taiyuan 030024, P.R. China. E-mail address: wangbaojun@tyut.edu.cn

^b College of Chemistry and Chemical Engineering, Taiyuan University of Technology, Taiyuan 030024, P.R. China. E-mail address: linglixia@tyut.edu.cn

^c Department of Chemical and Petroleum Engineering, University of Wyoming, 1000 E University Ave, Laramie, WY 82071, USA. E-mail address: mfan@uwyo.edu

^d State Key Laboratory of Coal Conversion, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan 030001, P.R. China. E-mail address: pingliu@sxicc.ac.cn

CO oxidative coupling to dimethyl oxalate (DMO) is the crucial step in coal to ethylene glycol (CTEG) ^[1]. Palladium (Pd) catalysts have been proved to be effective catalysts for this process ^[2], and the (111) facet has been identified as the active plane in catalysis ^[3]. However, Pd catalyst is a noble metal catalyst resulting in a great increase in the cost of production, thus how to reduce the usage of Pd is an urgent problem. Bimetallic catalysts have been used in a great number of reactions to reduce usage of noble metal catalysts and keep or improve catalytic performance. Oxidative coupling of CO to DMO on Pd(111), Pd-Cu(111), Pd-Al(111) and Pd-Ag(111) surfaces have been systematically investigated by means of density functional theory (DFT) together with periodic slab models and micro-kinetic modeling. The favorable pathway for DMO synthesis on these catalysts starts from the formation of two COOCH₃ intermediates, followed by the coupling to each other, and the catalytic activity follows the trend of Pd-Al(111) > Pd-Ag(111) > Pd(111) > Pd-Cu(111). Additionally, the formation of DMO is far favorable than that of dimethyl carbonate (DMC) on the Pd-Al(111) surface, but it is exactly opposite on the Pd-Ag(111) surface. The results have been further demonstrated by the micro-kinetic modeling. Therefore,

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Pd-Al bimetallic catalyst can be applied in practice to effectively enhance the catalytic performance and greatly reduce cost.

Keywords: CO oxidative coupling to DMO; Pd(111); Pd-Cu(111); Pd-Al(111); DFT; Micro-kinetic modeling; Catalytic performance

References

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