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Kinetic study on key reaction steps in methanol-to-olefin conversion: influence of catalyst topology

Jeroen Van der Mynsbrugge, Jeroen De Ridder, Karen Hemelsoet, Michel Waroquier and Veronique Van Speybroeck
Center for Molecular Modeling, Ghent University, Technologiepark 903, B-9052 Zwijnaarde, BELGIUM

Zeolite-catalyzed conversion of methanol to light olefins (MTO) is currently one of the most prominent alternatives to traditional cracking processes for the production of bulk chemicals such as ethene and propene. The search for a catalyst that offers maximal conversion and light-olefin selectivity is key to further optimizing the industrial process. This requires a thorough understanding of the underlying reaction mechanisms, which has proven a persistent challenge, despite considerable research efforts throughout the past decades.[1] Currently there is a consensus on an indirect mechanism in which olefin formation occurs through repeated methylation and/or cracking reactions of a pool of hydrocarbons present inside the zeolite pores. The predominant hydrocarbon pool species vary from small alkenes to bulky polymethylbenzenes, depending on the zeolite topology.[2]

While most studies originally focused on the archetypal MTO-catalysts H-ZSM-5 (MFI) and H-SAPO-34 (CHA), other zeolites such as H-ZSM-58 (DDR) and H-ZSM-22 (TON) are now also being investigated for their ability to convert methanol to olefins.[3,4] Earlier theoretical research established the paramount importance of methylation reactions, which were found to be the rate-determining steps in most olefin-producing routes.[5,6] Focusing on methylations, the current study uses quantum chemical calculations on extended cluster models that mimic the local environment of the active site to predict activation barriers and rate constants.[7] Comparing methylation kinetics in a selection of both established and newly-suggested catalysts offers new insights into the relation between their topological features and the activity of different species in the hydrocarbon pool, which is crucial for designing improved MTO-catalysts.

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E-mail: jeroen.vandermynsbrugge@ugent.be
www: http://molmod.ugent.be