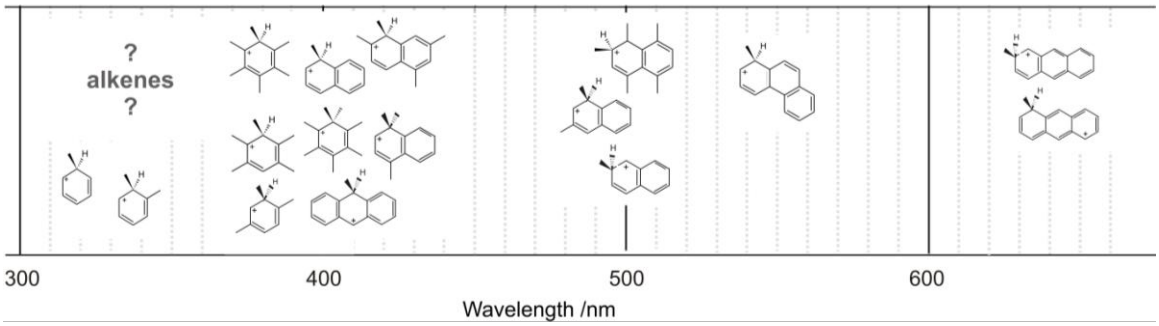
A theoretical assignment scale of aromatic and aliphatic MTO intermediates

K. Hemelsoet[1]; Q. Qian[2]; K. De Wispelaere[1]; T. De Meyer[1]; A. Van Yperen-De Deyne[1]; M. Waroquier[1] ; B. M. Weckhuysen[2]; V. Van Speybroeck[1]

[1] Center for Molecular Modeling (CMM); Ghent University; Belgium
[2] Inorganic Chemistry and Catalysis; Debye Institute for Nanomaterials Science; Utrecht University; The Netherlands

The optical absorption properties of an extended set of hydrocarbons are calculated using up to date time-dependent density functional theory. Alkenes and (poly)aromatics of increasing size and methylation degree are examined, since these species are an essential part – i.e. the hydrocarbon pool (HP) - of a working catalyst for the Methanol-to-Olefins (MTO) process.[1] To determine which species contribute to the experimental in-situ UV/Vis bands, a systematic series of both neutral and cationic HP species are analyzed. A molecular dynamics simulation study of the organic molecule is essential. During such simulation the flexibility is fully taken into account and the effect of temperature and environment on the UV/Vis spectra is determined by performing TD-DFT calculations at various snapshots of the molecular dynamics run. The theoretical energy absorption scale can help to assign absorption bands determined in in-situ UV/Vis spectra to structurally different species.[2] In addition, a new analysis procedure is presented to link changes in optical spectra to the vibrational modes and specific geometrical features of the organic compounds. The proposed methodology is of general use for characterization of intermediates in catalytic processes.



[1] Olsbye, U., Svelle, S., Bjørgen, M., Beato, P., Janssens, T. V. W., Joensen, F., Bordiga, S. and Lillerud, K. P., Angew. Chem. Int. Ed. 51 (2012) 5810.

[2] Hemelsoet, K., Qian, Q., De Meyer, T., De Wispelaere, K., De Sterck, B., Weckhuysen, B.M., Waroquier, M. and Van Speybroeck, V., Chem. Eur. J. (2013) doi: 10.1002/chem..201301965.

E-mail: Karen.Hemelsoet@UGent.be

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