A SEMIEMPIRICAL STUDY OF PROPYLENE ADSORPTION ON
EtInd$_2$ZrCH$_3^+$/$\text{SiO}_2$ (111)

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Abstract

An Extended Hückel Method (ICONC) which includes core-core repulsion to the energy terms was used to characterize propylene adsorption on an active site model of SiO$_2$-supported metallocene catalysts for olefin polymerization.

Keywords: SiO$_2$-supported catalyst, propylene adsorption modeling, catalysts modeling

INTRODUCTION

Molecular orbital studies of the Extended Hückel type (EHMO) while being less precise than ab initio calculations differentiate more readily between contributions from different origin, such as specific bending or repulsive interactions, to overall energy changes.

Olefin insertion in homogeneous catalyst systems [1] and α-agostic interaction in electron deficient metal alkyl species [2] have been the subject of MO theoretical studies in recent years. As usual for the mechanism of olefin polymerization, the first step is the olefin coordination on a deficient site followed by the subsequent insertion into the metal polymer bond through a cis opening. It has been shown that upon coordination a prochiral olefin such as propylene may give rise to non superposable re and si coordination for an
isotactic specific homogeneous catalyst like EtInd$_2$ZrCH$_3^+$ [3]. However, in the case of SiO$_2$ supported zirconocene, there are several restrictions to adsorption, because one side of the active site is occupied by the support. For these systems, the MO studies are really scarce and they should be able to help in understanding the reason for the low productivity obtained with such catalysts.

The purpose of this paper is to theoretically study propylene adsorption on a SiO$_2$-supported metallocene active site model: EtInd$_2$ZrCH$_3^+$/SiO$_2$ (111).

THEORETICAL METHOD

The program used (ICONC) [4] includes repulsive terms to the energy. The total energy of adsorbed species was calculated as the electronic energy difference the adsorbed molecule (propylene) at a finite distance from the surface of a EtInd$_2$ZrCH$_3^+$/SiO$_2$ (111) cluster and the same energy when the same molecule is far away from the surface.