ABSTRACT

This thesis focusses on reactive processes catalysed by zeolites to obtain and convert BTX aromatics. These compounds, together with light olefins, are the pillars of the petrochemical and organic chemical industry, as they are the raw materials to produce basic intermediates, such as polyester monomers, polyamides, engineering plastics and intermediate products for the manufacture of detergents, pharmaceuticals, fertilizers, and explosives.

Currently, the aromatics and light olefins are obtained mainly from petroleum refining, natural gas or coal. Along the last decades, natural gas conversion into higher value chemicals has gained importance, such as olefins and aromatics. The main reasons are its high hydrogen to carbon (H/C), its composition, which is independent of the source, and its availability, as world reserves are larger than those of oil, especially if shale gas is considered. Therefore, the methane aromatization reaction (MDA) is studied as a route for obtaining aromatics. The most widely used catalyst for this reaction is Mo/H-ZSM-5. Thus, the effect of various parameters on the catalyst activity has been studied, such as the physico-chemical properties of the parent sample and the metal component incorporation procedure, in order to find the optimum catalyst Mo/ZSM-5. Moreover, this study has allowed correlating successfully amount and nature of the two active centers involved in this reaction with methane conversion and product distribution. On the other hand, one of the mayor drawbacks of this reaction is the strong deactivation of the catalyst; therefore, a study of the regeneration possibilities has been carried out and has allowed the design of an effective protocol to prolong the catalyst life and with the aim of expanding its industrial applications. Furthermore, other medium pore and multipore zeolitic structures have been studied as alternative to ZSM-5 zeolite, and their catalytic behavior has been successfully correlated with their topologies.

Regarding the transformation and redistribution of the BTX aromatics, alkylation, disproportionation and isomerization reactions have been studied, which employ acid zeolites as catalyst to obtain alkylaromatics with high interest for the petrochemical industry, such as xylenes, ethylbenzene, cumene, ethyltoluene and cymene. The main zeolites used in these processes at commercial scale are ZSM-5, MCM-22, Beta and MOR. Therefore, new zeolitic materials have been studied considering the molecular dimension of reactants, products and reaction intermediates, as well as the dimensions of the zeolite's pore topologies. Multipore zeolites have also been included in this study, due to the potential "molecular traffic control" type of selectivity that can be improved by these structures presenting interconnected channel apertures of different size. The best performing zeolitic structures for these processes are ITQ-13, ITQ-22, ITQ-39 and ITQ-47, which due to their good catalytic behavior, could be considered as an attractive alternative to be used as catalysts on commercial scale.