# The migration and re-distribution of Pt and their application in the activation of C-H bonds of ethane

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**Abstract:** The shale-gas boom in the United States provides an opportunity to develop advanced catalysts and processes to enable the direct conversion of light alkanes, major constituents of shale-gas, into value-added chemicals, in which C-H bond activation is the first step. We have developed advanced catalyst, which consist of atomically dispersed Pt species supported on zeolite, for the activation of C-H bonds of ethane at relatively mild conditions. Under selected reaction conditions, ethane can be selectively converted to ethylene or aromatics. The supported highly dispersed Pt can be prepared through the migration and re-distribution of Pt upon different post-synthesis treatments.

**Keywords:** shale-gas, ethane, aromatics.

## 1. Introduction

Owing to the low cost and increased supply of natural gas in the United States, the direct conversion of natural gas liquids (C2-C4) into aromatics is of great industrial importance, in which C-H bond activation is the key step. We have developed a new process to selectively convert ethane into ethylene or aromatics under relatively mild conditions. We can achieve >60% conversion of ethane with the BTX selectivity > 50%, under selected reaction conditions. Our process involves atomically dispersed Pt species supported on zeolite. Such atomically dispersed catalysts have attracted significant scientific attention recently, owing to the high atom efficiency that these catalysts can achieve, but also improve the selectivity of certain reactions. However, it is usually a challenge task to prepare this type of catalysts.[1]

Recently, Datye and co-authors have reported that the migration and trapping of Pt upon aging in air can be used to synthesize single-atom Pt catalysts.[2] However, this synthesis has to be performed at rather high temperature (800 °C) for long time. In NICE America, We have developed a novel approach to prepare atomically dispersed Pt species supported on zeolite, through the migration and re-distribution of Pt, promoted by post-synthesis treatments with various gases. The atomically dispersed Pt species consist of isolated Pt single atoms and sub-nanometer Pt nanoclusters, which are exceptional active for the C-H bond activation of ethane. This approach for the preparation of atomically dispersed catalysts, through the migration and re-distribution, may have wide applications in heterogeneous catalysis.

## 2. Experimental

The initial catalysts were prepared through wet impregnation with platinum tetraamine nitrate solution in a rotary evaporator. The migration and re-distribution of Pt was achieved through post-synthesis treatments with various gases. The formation of atomically dispersed Pt species through the migration and re-distribution were investigated in-situ by dynamic CO chemisorption, diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS), X-ray absorption near edge structure (XANES), Extended X-Ray absorption fine structure (EXAFS). The catalytic performance of these supported atomically dispersed Pt species in the C-H bond activation of ethane was studied using microreactor and GC.

## 3. Results and discussion

Our zeolite supported atomically dispersed Pt catalysts can selectively convert ethane to aromatics at relatively mild conditions. Under selected reaction conditions, the conversion of ethane is > 60%, with BTX selectivity >50%. Although carbon coke decreased the activity of the catalyst after several hour of reaction, the activity can be re-generated after coke burning. The activity and selectivity of this catalyst in the ethane aromatization reaction are show in the Figure 1.

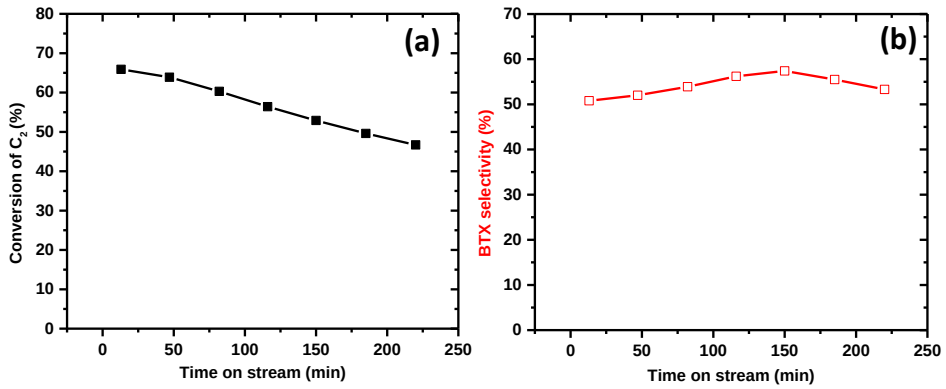


**Figure 1.** The catalytic performance of the zeolite supported atomically dispersed Pt species in the ethane aromatization reaction.

The formation of the atomically dispersed Pt species through the Pt migration and re-distribution, promoted by post-synthesis treatments, can be identified through various techniques. Figure 2 show the CO dynamic chemisorption spectra of the catalyst before Pt migration and after Pt migration. The figure clearly show that before Pt migration, Pt presents as nanoparticles, while after Pt migration, the atomically dispersed Pt species are formed.

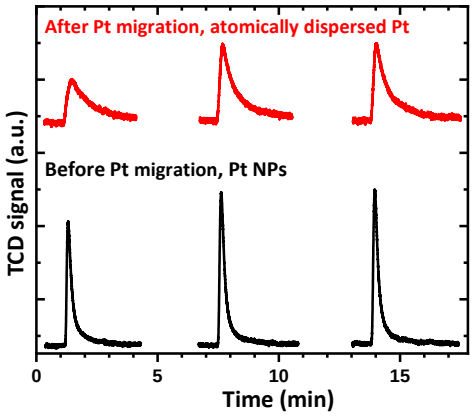


**Figure 2.** Dynamic CO chemisorption spectra of the sample before Pt migration (bottom), and after Pt migration (top).

## 4. Conclusions

A new novel approach has been developed to prepare supported atomically dispersed Pt catalysts, through Pt migration and re-distribution. The formed atomically dispersed Pt species supported on zeolite are highly active for the direct conversion of ethane to aromatics at relatively mild conditions. This approach may have wide applications in heterogeneous catalysis.

## References

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