DFT study of the reaction mechanism of N2O decomposition on Au3+/0/- clusters

Lin-Yu Wu[1], Cheng Chen[2], Lan Luo[3], yongcheng wang[4], and Bing Yin[5]

[1]College of Chemistry and Chemical Engineering, Northwest Normal University,
[2]College of Chemistry and Materials Science, Northwest University,
[3]College of Chemistry and Materials Science, Northwest University,
[4]College of Chemistry and Chemical Engineering
[5]Northwest University Xi'an

May 5, 2020

Abstract

In this paper, the reaction mechanism of Au3+/0/- clusters with N2O was studied by density functional theory (DFT) calculations. The analysis of the potential energy surfaces showed that the Au3 neutral cluster exhibited highest catalyze activity on the decomposition of N2O, energy barrier is only 11.60 kcal/mol. The corresponding energy barriers for Au3- and Au3+ are 28.51 and 58.79 kcal/mol, respectively. The effects of Au3+/0/- clusters assistance analyzed using the activation strain model indicated that the dissociation of the N-O bond depends on the interaction energy of Au3 clusters and N2O. The electron transfer from the Au3+/0/- cluster to the N2O facilitates the dissociation of N-O bond. The analysis of frontier molecular orbitals (FMO) indicated that only the interaction of HOMO-LUMO interactions is strongly enough and there is sufficient orbital overlap between the Au3+/0/- clusters and N2O, electron transfer can occur and activation of N2O can be achieved. The study of thermodynamic processes showed that there is evident correlation between the binding energy of the Au3O+/0/- cluster oxides and the barrier energy of the reaction. Therefore, the orbital interactions between Au3+/0/- and N2O and thermodynamic driving force have a great influence on the reaction. These results enrich our understanding of the catalytic dissociation of N2O by Au-cluster-based catalysts.

Hosted file

`12-07--IJQC.docx` available at https://authorea.com/users/288112/articles/413192-dft-study-of-the-reaction-mechanism-of-n2o-decomposition-on-au3-0-clusters