1E12

The covalent bond formation between Al atoms in Al-clusters hydrides

○Pawel Szarek, Kouhei Watanabe and Akitomo Tachibana

Department of Micro Engineering, Kyoto University, Kyoto 606-8501, Japan

We investigated electronic and structural properties and the effect of hydrogenation of "cage" aluminum clusters by means of Rigged QED. The bonds types, energy based bond orders [1], redistribution of chemical potential [1] over the cluster structures and MO analysis were calculated for understanding of hydrogen absorbing properties. We found that hydrogen adsorption on surface area influences cluster structure that some Al atoms become covalently bonded (Fig.1). The hydrogen atoms at bridging positions seem to constitute three-center two electron covalent bonds with neighboring Al atoms. We also compared small Al_4 and corresponding B_4 clusters and their hydrides.

Recently developed Rigged QED theory allows to define local tensorial chemical force called electronic stress tensor [2]. This force traced in principal axis gives a measure of kinetic energy density, which on the frame of Rigged QED is divided into positive and negative regions separated by interface - turning point for electron. The negative – compressive stress gives positive contribution to kinetic energy density, while positive – tensile stress has negative contribution and when is found in interatomic region to form so called spindle structure it indicates covalent interaction (Fig.1) [2].

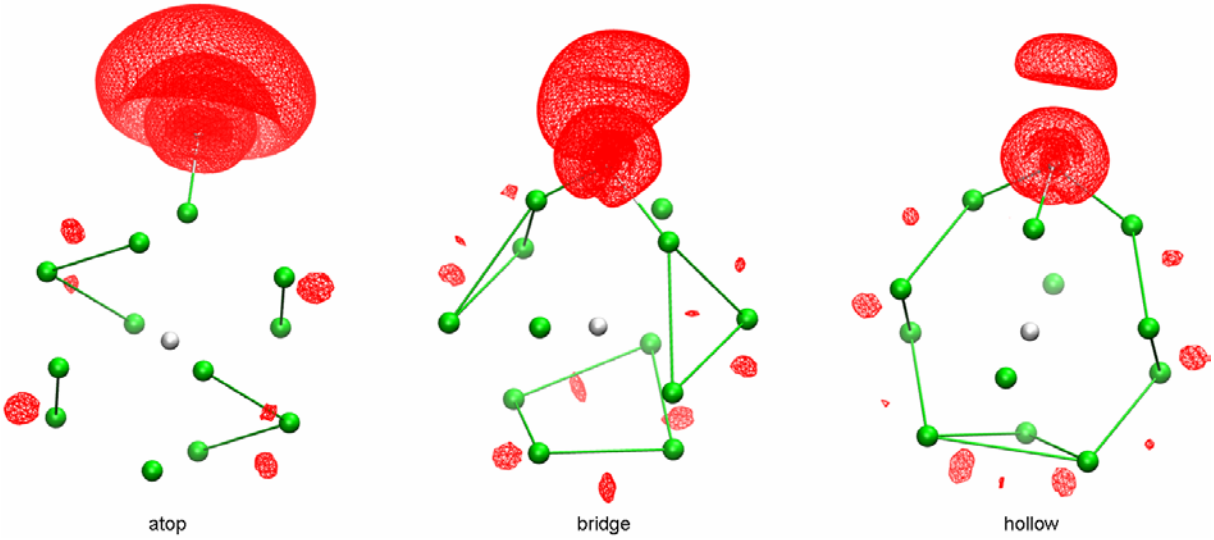

Fig. 1. The $Al_{12}MgH$ clusters, where Mg is central atom. The positive electronic stress (red regions) appears for different Al—Al bonds depending on hydrogen atom binding site indicating covalent interaction. The associated covalent bonds are distinguished with sticks. The "atop" structure possess ordinal Al—H covalent bond, the bridge structure is formed trough 3-centered-2-electron bond and hydrogen in hallow position is rather bonded to three Al atoms with electrons shered via covalent like manner.

Apart from stress tensor chemical force the divergence of latter leads to tension force. For stationary state, cancellation of tension in centain points of space defines stationary point a Lagrange point for electrons [2-5]. This point carries valuable interaction information [1]. The energy density and chemical potential analysis based on Lagrange point for non-relativistic limit of energy density [6] is source of strength and stability data such as energy based bond order indices [1]. There is a strong relation between these and structural parameters.

We have employed the Rigged QED theory to study hydrogen absorption properties of aluminium clusters. The hydrogen binding sites as well as the number of attached hydrogen atoms to the clusters have significant impact on electronic and chemical properties of Al-cages (Fig. 1).

Acknowledgements:

This work was supported by the Japanese Government – Monbukagakusho Scholarship through Kyoto University. The structural data were provided by Dr. David Henry at School of Applied Sciences, Materials Modelling and Simulation group, RMIT University.

References

[1] P. Szarek and A. Tachibana (2007) J Mol Model 13:651–663
[2] A. Tachibana (2004) Int J Quantum Chem 100:981–993
[3] Tachibana A (2001) J Chem Phys 115:3497–3518
[4] Tachibana A (2002) Energy density in materials and chemical reaction systems. In: Sen KD (ed) Reviews in modern quantum chemistry: a celebration of the contributions of Robert Parr, vol 2. World Scientific, Singapore, pp 1327–1366
[5] Tachibana A (1991) J Math Chem 7:95–110
[6] Tachibana A (2005) J Mol Model 11:301–311