

Initial Reaction in CVD Nanotube Synthesis by FT-ICR

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SWNTs are expected for various applications from various physical and chemical characters based on the geometric unique structure. As to the macroscopic generation, the ACCVD technique appears to be one of best synthesis methods. However its synthetic mechanism has not yet been made clear, so fundamental research is necessary to generate high quality SWNTs. In this paper we have explored the basic reaction mechanisms of relatively large catalyst clusters of iron, cobalt and nickel with ethanol.

The details of the experimental facility have already been reported, but essentially cluster ions were generated by a laser ablation / supersonic expansion cluster beam source. Trapped ions were reacted with ethanol (RT, 1×10^{-8} Torr) for one or two seconds.

Fig.1 shows relative rate constant of each transition metal cluster with ethanol. In the case of alkali metal clusters, they show magic number behavior explained by super shell theory, however there is no report of magic number for transition metal clusters. However as shown in Fig.1, chemical reactivity shows remarkable cluster size dependence. It is also very interesting that most reactive cluster sizes increased by one or two electrons with increasing atomic number. In this experiment, the dehydrogenation reaction process of transition metal clusters (Fe, Co, Ni) also change in order of atomic number. In the case of iron, the reaction with the ethanol molecule was nothing but a simple chemisorption, and in case of nickel, four hydrogen atoms were dissociated from the clusters. However in case of cobalt, which is situated between iron and nickel on the periodic table, two kinds of reaction pattern were shown.

Fig.2 shows the reaction results of cobalt clusters with isotopically modified ethanol. In Fig. 2(a) it is clear that 4 hydrogen atoms are dissociated. In Fig. 2(b, c, d) another reaction is seen, but it must be a product of H/D exchange. It is important to decide which signal is original and which signal is a product of H/D exchange. In this paper we have determined the dominant signals represent the actual products of the reaction. Based on this idea, 3 H atoms and 1 D atom are dissociated in Fig. 2(b), and 2 H atoms and 2 D atoms are so in Fig.2(c). From these experiments we can clearly determine the dehydrogenation reaction path of cobalt cluster with ethanol molecule.

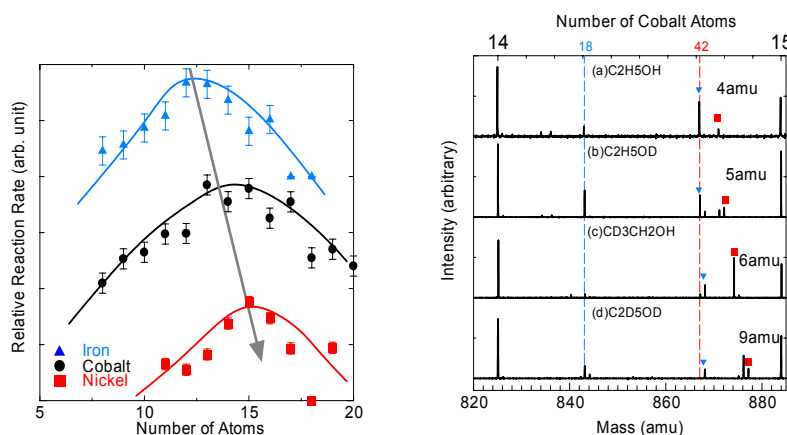


Fig. 1 Comparison of relative rate constant.

Fig. 2 Isotope experiment of cobalt clusters.

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