Gas Phase Reaction with FT-ICR and Molecular Dynamics Simulation of Precursor Clusters for SWNTs

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In order to investigate the formation mechanism of SWNTs, gas phase reaction experiments of Ni/Co/C and Ni/Y/C mixed clusters by using FT-ICR (Fourier Transform Ion Cyclotron Resonance) mass spectrometer and molecular dynamics simulations of metal-containing carbon cluster formation were performed. Metal-carbon binary clusters were generated by the laser vaporization of Ni/Co or Ni/Y loaded carbon materials used for macroscopic production of SWNTs. In case of the Ni/Co loaded carbon material, there was not a trace of metal-carbon binary clusters in the positive mass spectrum and tiny signals of NiC_n^- and CoC_n^- were observed for negative spectrum as in Fig. 1(a). The chemical reaction of these clusters with NO was used as the probe of the structure of clusters. The reaction results with 10^{-7} Torr NO for 2 s and 10 s are shown in Fig. 1(b) and Fig. 1(c), respectively. The chemisorption of NO to NiC_n^- and CoC_n^- were observed in much faster rate than pure carbon clusters and La-carbon binary clusters. The high reactivity of NiC_n^- and CoC_n^- strongly suggests that Ni or Co atom is outside the carbon cage. The formation process of metal-carbon binary clusters was also studied using the molecular dynamics simulations. Based on DFT calculations of various forms of small clusters MC_n and M_n (M = La, Sc, Ni), multi-body classical potential function for M-C and M-M interactions were constructed with the Morse-type term and the Coulomb term as function of coordinate number of a metal atom. The growth process of carbon clusters from completely random vapor phase was simulated with and without 1% of metal atoms. Inclusion of La atoms enhanced the clustering process to the random caged carbon clusters, with a La atom inside the cage. However, a Ni atom on the face of the random cage delayed the annealing of the cage.

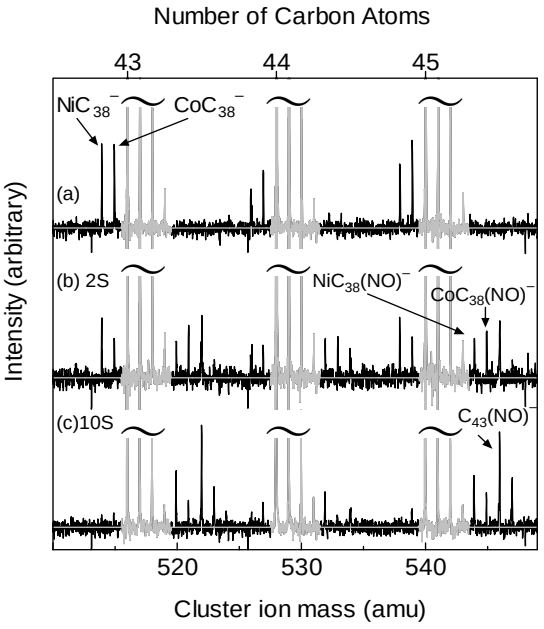

Fig. 1. Reaction of NiC_{38}^- and CoC_{38}^- with NO. Note that signal of pure carbon are drawn in gray lines.