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In-situ ETEM Studies of Fe Catalyst NPs Formation under Molecular or Radicals/Activated Hydrogen Environments for the Growth of SWCNTs

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Catalyst design plays vital roles in the structurally relevant chemical reactions. Revealing the catalyst structure and associated chemical activity in the reactive environment and at the atomic scale is imperative for the rational design of catalysts as well as for the investigation of reaction mechanism [1,2]. However *in situ* characterization at the atomic scale at high temperature is still a great challenge. [3]-[5]. The present study is focused on the realization of *in situ* TEM observations of Fe catalyst film dewetting and catalytic nanoparticles (NPs) formation under molecular and/or radical/ activated hydrogen environments. The formed NPs exhibit different sizes depending on the temperature and type of reactive gas. The experiments are performed within a modified environmental transmission electron microscope-ETEM equipped with a Cs image aberration-corrector; a special sample holder and customized CVD gas sources that allow sending collimated beams of molecules/radicals with different partial pressure.

From the experimental point of view the analysis of the recorded images at different temperatures and exposure times allowed us observing that the Fe film dewetting is strongly related to both the nature of hydrogen environment and the temperature conditions. Thus, through *in-situ* TEM heating process under 3sccm ($1.4\ 10^{-4}$ mbar) of molecular H2 environment we notice that the Fe film dewetting starts at 400°C leading to the formation of big crystalline Fe nanoparticle (see Figure1). We have observed that the Fe NPs size increases, 20nm size, as the temperature increases while keeping the same H2 flow. On the contrary, under 6sccm ($2.5\ 10^{-3}$mbar) activated hydrogen environment the Fe film dewetting initiate only at higher temperature, around 500°C with the formation of smaller crystalline Fe Nps with sizes ranging from 1nm up to 5nm(see Figure2).The formation of such small nanoparticle is related to the presence of hydrogen radicals which tend to block the metal atoms surface diffusion by creating small defects within the support which pin the forming nanoparticle and prevent their agglomeration.

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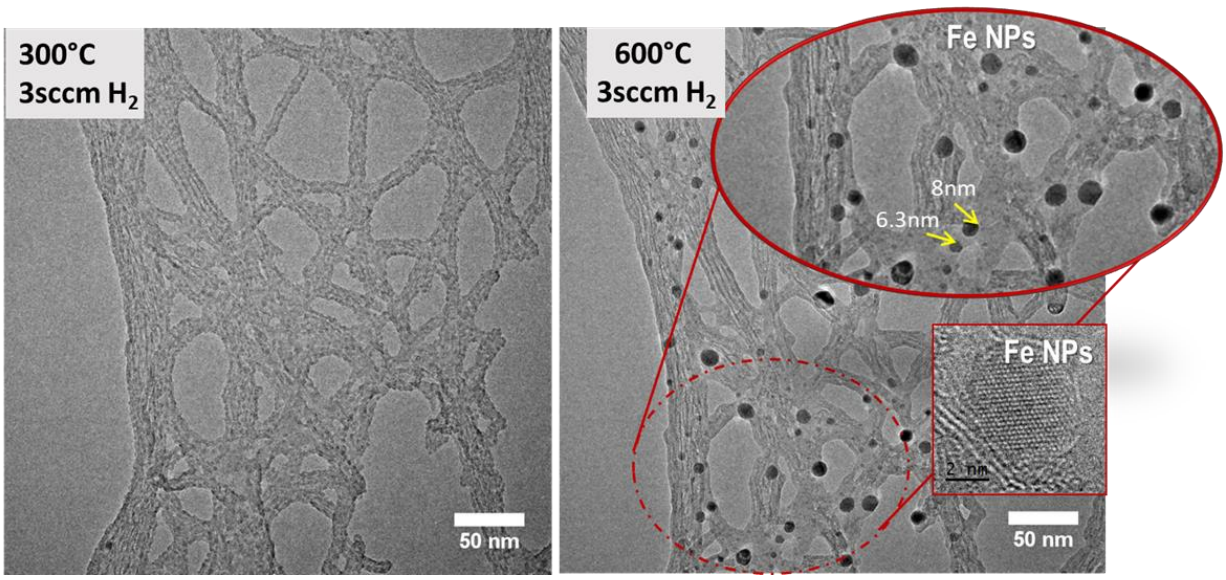

Figure 1. *In-situ* TEM analysis of Fe film dewetting through heating process under 3sccm molecular H2 environment illustrating the formation of Fe NPs.

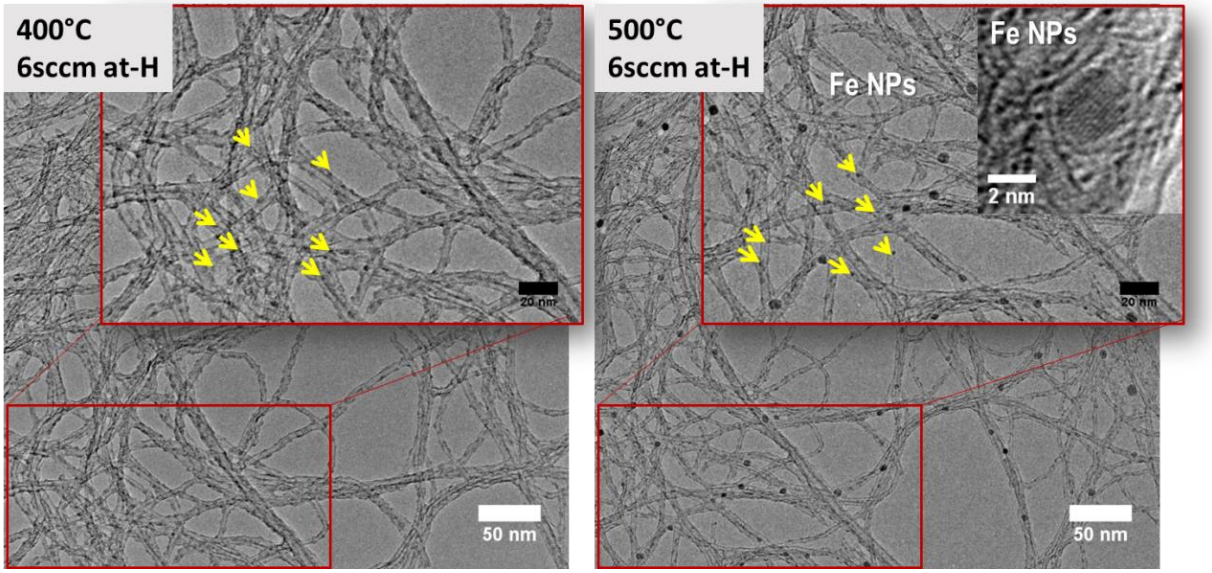

Figure 2. *In-situ* TEM analysis of Fe film dewetting through heating process under 6 sccm activated H2 environment illustrating the formation of small Fe metallic NPs.

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