Supporting Information: Dynamic Catalyst
Restructuring During Carbon Nanotube Growth

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Figure SI1 shows evidence for lattice fringes in the Ni clusters at temperatures as high as ∼890K. Other examples can be found in Ref. 1.

Effect of dissolved C

The possible presence of dissolved C does not affect the conclusions of our continuum model. To verify this, we compute the diffusion coefficient of a fcc-Fe(110) surface with and without dissolved C. We use Fe instead of Ni due to the availability of tested Fe-C potentials in the literature, where Fe-Fe, Fe-C and C-C potentials are second moment tight-binding, Johnson and Lennard-
**Fig SI1.** ETEM snapshots of a Ni nanocrystal at ∼890K in ∼10⁻³ mbar C₂H₂. The strong reflections in the images fast Fourier transforms (insets) correspond to fcc Ni{111} planes.

**Fig SI2.** Arrhenius plot showing the negligible effect of C for self-surface diffusion in a fcc-Fe(110) slab, periodic in "x" and "z". A wide vacuum layer avoids self interaction in the "y" direction. The diffusion coefficient is calculated with the drift force method. The pristine slab consists of 12 layers, 7 free to move. A force $f=0.25\,\text{eV/nm}$ is applied to all Fe atoms in the upper free-layers in the "+z" direction. As a result, these acquire a velocity $v$. The surface diffusion coefficient is calculated as $D_s = v k_B T / f$ and plotted in black squares. When ∼5at% C is dissolved, the force on the Fe atoms is the same, while that on the C atoms is in the opposite direction to simulate the opposite flow of Fe and C during CNT growth and particle dewetting. $D_s$ of the Fe-C system is represented by red circles.

Jones. The behavior of C in Ni should follow the same trends as in fcc-Fe. Figure SI2 shows that dissolving up to ∼5at% C, the diffusion coefficient increases negligibly with respect to pure Fe.
Movies

Experimental

- **Movie S1.** Real time ETEM movie in 3:1 NH$_3$:C$_2$H$_2$ at 1.3mbar and 750K showing morphing of an elongated Ni particle dewetting from a CNF, with a final spherical shape. Fig. 1a in the main text plots the frames of this movie.

- **Movie S2.** Real time ETEM movie in 3:1 NH$_3$:C$_2$H$_2$ at 1.3mbar and 750K showing the escape of a Ni particle through a carbon nano-neck. Fig. 1b in the main text plots the frames of this movie.

- **Movie S3.** Real time ETEM movie in 3:1 NH$_3$:C$_2$H$_2$ at 1.3mbar and 750K showing the splitting of a Ni particle due to the formation of a carbon neck. Fig. 1c in the main text plots the frames of this movie.

Molecular Dynamics

- **Movie S4.** MD simulation of a Ni nanoparticle dewetting from a CNT via surface diffusion. Fig. 6a in the main text plots frames of this movie.

- **Movie S5.** MD simulation of a Ni nanoparticle dewetting from a CNT with a neck constriction. Fig. 6b in the main text plots frames of this movie.

- **Movie S6.** MD simulation showing the fracture of a back-anchored Ni nanoparticle caused by a Nichols-Mullins instability initiated by a neck constriction in the CNT. Fig. 6c in the main text plots frames of this movie.

References

