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Supporting Information

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Incremental Growth of Short SWNT Arrays by Pulsed Chemical Vapor Deposition

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1. Time resolved reflectivity (TRR) curve corresponding to uncoordinated growth of carbon nanotubes.



Fig. 1S. TRR curve, corresponding to uncoordinated growth of nanotubes (see Fig. 4b). The reflectivity does not show interference fringes as in the case of coordinated growth. The insert shows the magnified view of the reflectivity change during the first gas pulse.



2. SEM image corresponding to uncoordinated growth of carbon nanotubes



Fig. 2S. Cross sectional SEM image of single wall carbon nanotubes in the case of uncoordinated growth.



3. Modeling of Gas Dynamics

In this work, we have employed general purpose CFD (Computational Fluid Dynamics) software called MFIX (Multiphase Flow with Interphase eXchanges).^[1-3] We have utilized only the single-phase capabilities of this software and the following continuity, momentum, species, and energy equations are solved in cylindrical coordinates.

Continuity Equation

$$\frac{\partial}{\partial t}(\rho_g) + \frac{\partial}{\partial x_j}(\rho_g U_{gj}) = 0 \tag{1}$$

Momentum Equation

$$\frac{\partial}{\partial t} \left(\rho_g U_{gi} \right) + \frac{\partial}{\partial x_j} \left(\rho_g U_{gj} U_{gi} \right) = - \left(\frac{\partial p_g}{\partial x_i} \right) + \frac{\partial \tau_{gij}}{\partial x_j} + \rho_g G_i$$
(2)

where,

$$\tau_{gij} = \mu_g \left[\left(\frac{\partial U_{gi}}{\partial x_j} + \frac{\partial U_{gj}}{\partial x_i} \right) - \frac{2}{3} \frac{\partial U_{gi}}{\partial x_i} \delta_{ij} \right]$$
(3)

Species Equation

$$\frac{\partial}{\partial t} \left(\rho_g X_{gn} \right) + \frac{\partial}{\partial x_j} \left(\rho_g U_{gj} X_{gn} \right) = \frac{\partial}{\partial x_i} \left(D_{gn} \frac{\partial X_{gn}}{\partial x_i} \right) \tag{4}$$

Energy Equation

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$$\rho_g C_{pg} \left[\frac{\partial T_g}{\partial t} + U_{gi} \frac{\partial T_g}{\partial x_i} \right] = -\frac{\partial}{\partial x_i} \left(k_g \frac{\partial T_g}{\partial x_i} \right) - \left[\frac{\partial p_g}{\partial t} + U_{gi} \frac{\partial p_g}{\partial x_i} \right]$$
(5)

The above equations are spatially discretized using a finite volume technique (first-order upwinding in this case) and an implicit backward

Euler method is used for time discretization. At each time step, MFIX uses Picard fixed point iteration to solve the set of coupled, highly nonlinear equations that arise from the discretization of transport and conservation laws.



Fig. 3S. Boundary conditions used in calculations.

The set of nonlinear equations is linearized using the SIMPLE^[4] formulation of fluid velocity correction and pressure correction. For each variable, a system of sparse, non-symmetric linear equations corresponding to a regular seven-point stencil on a logically rectangular grid are solved.

The computational setup of the pulse reactor is shown in Fig. 9 a (not to scale). The mass-flow rate along with species composition is prescribed at the inlet. The temperature profile on the wall is prescribed and is of the following functional form: $1000/cosh(4x-2L)^{0.7}$. Symmetric boundary conditions are prescribed at the centerline and this problem is solved as a 2D axisymmetric case (Fig. 3S).

This case is run with very small time-steps (100 μ s) to resolve the injected pulse width of 1ms, and the pulse is injected after 0.5 seconds. By 0.5 seconds the solution has reached stationary state starting with initial conditions (reference temperature, constant axial



velocity corresponding to the mass flow rate, and radial velocity set to zero). The properties of the gases are calculated using the BURCAT thermochemical database.

Nomenclature:

 $\rho_g = gas \ density; U_{gj} = j \ component \ of \ gas \ velocity; \ p_g = gas \ pressure; \ \tau_{gij} = shear \ stress \ tensor; \ \mu_g = gas \ viscosity; \ G_i = gravity \ vector \ in \ i \ direction - in \ this \ case \ taken \ to \ be \ zero; \ \delta_{ij} = dirac - delta \ function; \ X_{gn} = nth \ gas \ species \ mass \ fraction;$ $D_{gn} = diffusion \ coefficient \ of \ the \ nth \ gas \ species;$ $C_{pg} = heat \ capacity \ of \ the \ gas \ mixture; \ k_g = thermal \ conductivity \ of \ gas \ mixture$

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