

# SUPPLEMENTARY MATERIAL

accompanying the article

## Scale-up of Nanoparticle Synthesis by Flame Spray Pyrolysis: The High Temperature Particle Residence Time

by

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**Number of Pages: 10**

**Number of Tables: 3**

The supplementary material provides additional details of the employed CFD model, namely conservation equations, spray droplet heat and mass transfer, reaction rate expressions and thermodynamic properties, as well as fundamentals of the aerosol dynamics model.

### Conservation Equations

The heat, mass and momentum balances of the discrete and continuous phase are connected by two-way coupling in ANSYS Fluent.<sup>1</sup> This is accomplished by solving both phases alternatively until a converged solution is obtained. The momentum source term for the continuous phase is computed from the change of droplet momentum in a control volume:

$$\mathbf{F} = -\sum \left( \frac{18\mu C_D Re_d}{\rho_d d_d^2 24} (\mathbf{u}_d - \mathbf{u}_g) \right) \dot{m}_d \Delta t, \quad (\text{S1})$$

Where  $C_D$  is the drag coefficient and  $\Delta t$  the time step. The heat exchange is computed by examining the change in thermal energy of a particle in each control volume:

$$H = \frac{\dot{m}_{d,i}}{m_{d,i}} \left( -h_{ref} (m_{d,in} - m_{d,out}) - m_{d,out} \int_{T_{ref}}^{T_{d,out}} (c_{p,d} \partial T) + m_{d,in} \int_{T_{ref}}^{T_{d,in}} (c_{p,d} \partial T) \right), \quad (\text{S2})$$

Where indices *in* and *out* refer to cell entry and exit, respectively while *ref* indicates reference conditions. Droplet heat capacity is  $c_{p,d}$ . The mass exchange source term is the change of droplet mass as it passes through each control volume:

$$M = \frac{\Delta m_d}{m_{d,i}} \dot{m}_{d,i}, \quad (\text{S3})$$

### Spray Droplet Heat and Mass Transfer

The mass and momentum balance equations between the liquid and gas phase were solved similar to Gröhn et al.<sup>2</sup>:

$$\frac{\partial Q}{\partial t} = \pi d_d Nu_0 \tilde{k}_g (T_g - T_d) + \sum_j \left( \frac{\partial m_j}{\partial t} h_j \right), \quad (\text{S4})$$

$$\frac{\partial m_j}{\partial t} = -\varepsilon_j \pi \rho_g D_g d_d Sh^* \ln(1 + B_M), \quad (\text{S5})$$

for which the relative mass flux of each component is obtained from:

$$\varepsilon_j = \frac{\partial m_j}{\partial m} = \frac{Y_{j,s} (1 + B_M) - Y_{j,g}}{B_M}, \quad (\text{S6})$$

where  $B_M$  is the multi-component Spalding mass transfer number:

$$B_M = \left[ \left( 1 - \sum_j Y_{j,g} \right) - \left( 1 - \sum_j Y_{j,s} \right) \right] \left( 1 - \sum_j Y_{j,s} \right)^{-1}. \quad (\text{S7})$$

The modified Sherwood number based on Abramzon and Sirignano<sup>3</sup> is:

$$Sh^* = 2 + \frac{Sh_0 - 2}{F_M}, \quad (\text{S8})$$

with film thickness correction:

$$F_M = (1 + B_M)^{0.7} \frac{\ln(1 + B_M)}{B_M}. \quad (\text{S9})$$

The Sherwood and Nusselt numbers are computed as:

$$(Nu_0, Sh_0) = \frac{2 + 0.555\sqrt{Re}(Pr, Sc)^{1/3}}{\sqrt{1 + 1.232/(Re(Pr, Sc)^{4/3})}}. \quad (\text{S10})$$

The evaporation rate of each component was assumed to be proportional to its concentration at the droplet surface, computed according to Raoult's law. The heat transferred to droplets (eq. S4, first term) was calculated according to the Nusselt number,  $Nu_0$ , (eq. S10) after which the evaporation rate (eq. S5) and cooling by latent heat (eq. S3, second term) was computed from the modified Sherwood number (eqs. S8 – S10) and Spalding mass transfer number (eq. S7) for each component  $j$  (eq. S6). When the total vapor pressure at the droplet surface reaches the pressure of the surrounding gas, droplets start to boil and thus their temperature stays constant. Then, all heat transferred to the droplet is consumed by latent heat allowing to solve for mass transfer:

$$\frac{\partial m_j}{\partial t} = \frac{X_{j,s} \pi d_d Nu_0 \tilde{k}_g}{h_j} (T_g - T_d). \quad (\text{S11})$$

The multi-component evaporation model<sup>4</sup> assumes a well-mixed liquid phase<sup>5</sup> due to rapid mixing by internal boiling.<sup>6</sup>

### Reaction Rate Expressions and Thermodynamic Properties

One-step global Arrhenius rates,  $R_{k,r}$ , used for gas phase reactions were computed as:<sup>1</sup>

$$R_{k,r} = (v'_{k,r} - v''_{k,r}) \left( k_r \prod_j C_j^{(\eta'_{j,r} + \eta''_{j,r})} \right), \quad (\text{S12})$$

$$k_r = A_r e^{-E/R_g T}, \quad (\text{S13})$$

where  $C_j$  is the molar concentration ( $\text{kmol/m}^3$ ) of species  $j$  in reaction  $r$ ,  $\eta'_{j,r}$  is the rate exponent for reactant species  $j$  in reaction  $r$  while  $\eta''_{j,r}$  is that of product species. Similarly,  $\nu'_{k,r}$  is the stoichiometric coefficient for reactant species  $k$  in reaction  $r$  while  $\nu''_{k,r}$  is that of product species.  $A_r$  is the pre-exponential factor and  $E$  the activation energy ( $\text{J/mol}$ ). Apart from  $E$ ,<sup>7</sup> xylene oxidation is assumed similar to that of toluene.<sup>8,9</sup> Ideal-gas mixing laws<sup>1</sup> are applied for the gas-phase. Liquid-phase properties are calculated by mass weighted mixing laws<sup>1</sup> except for density and viscosity for which volume weighted mixing law<sup>1</sup> and Bingham correlation<sup>10</sup> are used, respectively. Values for gas phase reaction rates as well as gas and liquid phase thermodynamic properties are given in Tables S1-3, respectively.

**Table S1.** Coefficients used to compute gas phase reaction rates.

Reaction	Species	Rate Exponent, $\eta$	Stoichiometric Coefficient, $\nu$
Zr 2-ethylhexanoate combustion	$\text{C}_{24}\text{H}_{46}\text{O}_6\text{Zr}$	1	1
	$\text{O}_2$	1	33.5
	$\text{ZrO}_2$	0	1
Oxidation limited only by mass transport	$\text{CO}_2$	0	24
	$\text{H}_2\text{O}$	0	23
	$\text{C}_8\text{H}_{10}$	-0.1	1
Xylene combustion <sup>1,7</sup> $A_r = 8.997 \cdot 10^8$ $E = 8.368 \cdot 10^7$	$\text{O}_2$	1.85	10.5
	$\text{CO}_2$	0	8
	$\text{H}_2\text{O}$	0	5
	$\text{CH}_4$	0.2	1
Methane combustion <sup>1,11</sup> $A_r = 2.119 \cdot 10^{11}$ $E = 1.2552 \cdot 10^8$	$\text{O}_2$	1.3	2
	$\text{CO}_2$	0	1
	$\text{H}_2\text{O}$	0	2

**Table S2.** Thermodynamic properties of gaseous species.Polynomial values reported as  $A_1 + A_2T + A_3T^2 + A_4T^3 + A_5T^4$ .

Species	Heat Capacity, $J \cdot kg^{-1} \cdot K^{-1}$	Thermal Conductivity, $W \cdot m^{-1} \cdot K^{-1}$	Viscosity, $kg \cdot m^{-1} \cdot s^{-1}$	Standard Enthalpy, $J/kmol$	State
Carbon Dioxide <sup>1</sup>	$300 K > T < 1000 K$ : $A_1 = 4.299 \cdot 10^2$ $A_2 = 1.874 \cdot 10^0$ $A_3 = -1.966 \cdot 10^{-3}$ $A_4 = 1.297 \cdot 10^{-6}$ $A_5 = -4.000 \cdot 10^{-10}$	$1000 K \geq T < 5000 K$ : $A_1 = 8.414 \cdot 10^2$ $A_2 = 5.932 \cdot 10^{-1}$ $A_3 = -2.415 \cdot 10^{-4}$ $A_4 = 4.523 \cdot 10^{-8}$ $A_5 = -3.153 \cdot 10^{-12}$	$1.450 \cdot 10^{-2}$	$1.370 \cdot 10^{-5}$	$-3.935 \cdot 10^8$
	$300 K > T < 1000 K$ : $A_1 = 4.036 \cdot 10^2$ $A_2 = 9.057 \cdot 10^0$ $A_3 = -1.443 \cdot 10^{-2}$ $A_4 = 1.581 \cdot 10^{-5}$ $A_5 = -6.343 \cdot 10^{-9}$	$1000 K \geq T < 5000 K$ : $A_1 = 8.725 \cdot 10^2$ $A_2 = 5.305 \cdot 10^0$ $A_3 = -2.008 \cdot 10^{-3}$ $A_4 = 3.517 \cdot 10^{-7}$ $A_5 = -2.333 \cdot 10^{-11}$	$3.320 \cdot 10^{-2}$	$1.087 \cdot 10^{-5}$	$-7.489 \cdot 10^7$
Nitrogen <sup>1</sup>	$300 K > T < 1000 K$ : $A_1 = 9.790 \cdot 10^2$ $A_2 = 4.179 \cdot 10^{-1}$ $A_3 = -1.176 \cdot 10^{-3}$ $A_4 = 1.674 \cdot 10^{-6}$ $A_5 = -7.256 \cdot 10^{-10}$	$1000 K \geq T < 5000 K$ : $A_1 = 8.686 \cdot 10^2$ $A_2 = 4.416 \cdot 10^{-1}$ $A_3 = -1.687 \cdot 10^{-4}$ $A_4 = 2.997 \cdot 10^{-8}$ $A_5 = -2.004 \cdot 10^{-12}$	$2.420 \cdot 10^{-2}$	$1.663 \cdot 10^{-5}$	0
	$300 K > T < 1000 K$ : $A_1 = 8.348 \cdot 10^2$ $A_2 = 2.930 \cdot 10^{-1}$ $A_3 = -1.496 \cdot 10^{-4}$ $A_4 = 3.414 \cdot 10^{-7}$ $A_5 = -2.278 \cdot 10^{-10}$	$1000 K \geq T < 5000 K$ : $A_1 = 9.607 \cdot 10^2$ $A_2 = 1.594 \cdot 10^{-1}$ $A_3 = -3.271 \cdot 10^{-5}$ $A_4 = 4.613 \cdot 10^{-9}$ $A_5 = -2.953 \cdot 10^{-13}$	$2.460 \cdot 10^{-2}$	$1.919 \cdot 10^{-5}$	0
Water <sup>1</sup>	$300 K > T < 1000 K$ : $A_1 = 1.563 \cdot 10^3$ $A_2 = 1.604 \cdot 10^0$ $A_3 = -2.933 \cdot 10^{-3}$ $A_4 = 3.216 \cdot 10^{-6}$ $A_5 = -1.157 \cdot 10^{-9}$	$1000 K \geq T < 5000 K$ : $A_1 = 1.233 \cdot 10^3$ $A_2 = 1.411 \cdot 10^0$ $A_3 = -4.029 \cdot 10^{-4}$ $A_4 = 5.543 \cdot 10^{-8}$ $A_5 = -2.950 \cdot 10^{-12}$	$2.610 \cdot 10^{-2}$	$1.340 \cdot 10^{-5}$	$-2.418 \cdot 10^8$
	$150 K > T < 1500 K$ : $A_1 = 6.616 \cdot 10^1$ $A_2 = 4.413 \cdot 10^0$ $A_3 = -1.120 \cdot 10^{-3}$ $A_4 = -8.075 \cdot 10^{-7}$ $A_5 = -3.884 \cdot 10^{-10}$		$A_1 = -3.702 \cdot 10^{-3}$ $A_2 = 1.917 \cdot 10^{-5}$ $A_3 = 1.017 \cdot 10^{-7}$ $A_4 = -3.621 \cdot 10^{-11}$	$250 K > T < 1500 K$ : $A_1 = -4.246 \cdot 10^7$ $A_2 = 2.272 \cdot 10^{-8}$ $A_3 = -1.753 \cdot 10^{-12}$ $A_4 = -6.433 \cdot 10^{-16}$	$2.110 \cdot 10^7$
Zr 2-ethylhexanoate <sup>14,15</sup>	$1.200 \cdot 10^3$		$4.600 \cdot 10^{-3}$	$3.780 \cdot 10^{-6}$	$-1.530 \cdot 10^9$

**Table S3.** Thermodynamic properties of liquid species.Polynomial values reported as  $A_1 + A_2T + A_3T^2 + A_4T^3 + A_5T^4$ .

Species	Heat Capacity, $J \cdot kg^{-1} \cdot K^{-1}$	Thermal Conductivity, $W \cdot m^{-1} \cdot K^{-1}$	Density, $kg/m^3$	Latent Heat of Vaporization, $J/kg$	Saturation Vapor Pressure, $Pa$	Binary Diffusivity, $m^2/s$	Viscosity, $kg \cdot m^{-1} \cdot s^{-1}$	Surface Tension, $N/m$
Xylene <sup>12,13,16</sup>	$43 K > T < 516 K$ : $A_1 = 1.110 \cdot 10^3$ $A_2 = 4.164 \cdot 10^0$ $A_3 = -1.066 \cdot 10^{-2}$ $A_4 = 1.467 \cdot 10^{-5}$ $A_5 = 7.423 \cdot 10^{-23}$	$1.264 \cdot 10^{-1}$	$8.640 \cdot 10^2$	$3.440 \cdot 10^5$	$286 K > T < 616 K$ : $A_1 = 2.046 \cdot 10^6$ $A_2 = -2.698 \cdot 10^4$ $A_3 = 1.359 \cdot 10^2$ $A_4 = -3.096 \cdot 10^{-1}$ $A_5 = 2.692 \cdot 10^{-4}$	$7.416 \cdot 10^{-6}$	$286 K > T < 589 K$ : $A_1 = 1.463 \cdot 10^{-2}$ $A_2 = -1.149 \cdot 10^{-4}$ $A_3 = 3.496 \cdot 10^{-7}$ $A_4 = -4.788 \cdot 10^{-10}$ $A_5 = 2.466 \cdot 10^{-13}$	$2.820 \cdot 10^{-2}$
	$1.660 \cdot 10^3$	$1.360 \cdot 10^{-1}$	$1.120 \cdot 10^3$	$1.380 \cdot 10^5$	$^a 1.013 \cdot 10^5 \cdot (1.380 \cdot 10^5 / (R_g T))^2 \cdot \exp(-1.380 \cdot 10^5 / (R_g T))$	$2.850 \cdot 10^{-6}$	$^b 298 K > T < 343 K$ : $A_1 = 3.644 \cdot 10^1$ $A_2 = -4.366 \cdot 10^{-1}$ $A_3 = 1.971 \cdot 10^{-3}$ $A_4 = -3.973 \cdot 10^{-6}$ $A_5 = 3.014 \cdot 10^{-9}$	$1.420 \cdot 10^{-6}$

<sup>a</sup>Clausius–Clapeyron relation<sup>b</sup>Measured by VT550 Viscotester

## Aerosol Dynamics Model

The present CFD implementation of aerosol dynamics is based on the monodisperse population balance equations of Kruis et al.<sup>18</sup> which were used also by Mueller et al.<sup>19</sup> for the description of zirconia nanoparticle growth in spray flames. The evolution of the total agglomerate number ( $N$ ; eq. S14), area ( $A$ ; eq. S15) and volume ( $V$ ; eq. S16) concentration were modeled with transport equations where source terms are used to account for the actual aerosol dynamics.<sup>20</sup> This approach allows solving arbitrary flow fields and geometries. The system of equations describing the particle dynamics in steady state was implemented to ANSYS Fluent<sup>1</sup> through user defined functions as:

$$\frac{\partial(\mathbf{u}_g \rho_g N)}{\partial \mathbf{x}} - \frac{\partial}{\partial \mathbf{x}} \left( \frac{\mu_t \partial N}{Sc_t \partial \mathbf{x}} \right) = k_f - \frac{1}{2} \beta (\rho N)^2, \quad (\text{S14})$$

$$\frac{\partial(\mathbf{u}_g \rho_g A)}{\partial \mathbf{x}} - \frac{\partial}{\partial \mathbf{x}} \left( \frac{\mu_t \partial A}{Sc_t \partial \mathbf{x}} \right) = k_f a_0 - \frac{\rho(A - Na_s)}{\tau_s}, \quad (\text{S15})$$

$$\frac{\partial(\mathbf{u}_g \rho_g V)}{\partial \mathbf{x}} - \frac{\partial}{\partial \mathbf{x}} \left( \frac{\mu_t \partial V}{Sc_t \partial \mathbf{x}} \right) = k_f v_0. \quad (\text{S16})$$

The first two left-hand side terms in eq. S14-S16 describe the convection and diffusion of the particles in turbulent flow. The term  $k_f$  is the particle formation rate,  $v_0$  and  $a_0$  are the volume and the surface area of a  $ZrO_2$  monomer, respectively. The coagulation rate was computed with the Fuchs interpolation kernel,  $\beta$ , accounting for the fractal structure of the agglomerates with  $D_f = 1.8$ .<sup>18</sup> The agglomerate total surface area decreases by sintering towards more compact structures in the high temperature regimes of the flame. The  $\tau_s$  is the characteristic sintering time, defined as the time needed to reduce ~63 % of the excess surface area<sup>21</sup> over that of a completely fused aggregate,  $a_s$ . The primary particle diameter ( $d_p$ ), number of primary particles per agglomerate ( $n_p$ ) and agglomerate diameter of gyration ( $d_{gyr}$ ) are:<sup>18</sup>

$$d_p = \frac{6V}{A}, \quad (\text{S17})$$

$$n_p = \frac{6V}{\pi d_p^3}, \quad (\text{S18})$$

$$d_{gyr} = d_p n_p^{1/D_f}. \quad (\text{S19})$$

Sintering kinetics of zirconia<sup>22</sup> were described with the characteristic sintering time  $\tau_s$  and diffusion coefficient  $D_b$ :

$$\tau_s = 1.703 \cdot 10^{-3} \frac{R_g T d_p^4}{16 w D_b \gamma \Omega}, \quad (\text{S20})$$

$$D_b = D_0 \exp\left(\frac{-E}{R_g T}\right). \quad (\text{S21})$$

The parameters for equations (S20) and (S21) were obtained as: Grain boundary width  $w = 5 \cdot 10^{-10}$  m, grain boundary diffusion pre-exponential factor  $D_0 = 3.3 \cdot 10^{-5}$  m<sup>2</sup>/s and activation energy  $E = 1.88 \cdot 10^5$  J/mol<sup>23</sup> as well as  $\gamma = 1.545$  N/m for the average surface tension of tetragonal ZrO<sub>2</sub>.<sup>24</sup> The molar volume  $\Omega$  was computed from the molecular weight and bulk density of ZrO<sub>2</sub>. This sintering rate was selected among others based on comparison of model predictions to measured ZrO<sub>2</sub> primary particle diameters made by laboratory-scale FSP.<sup>2</sup>

## Nomenclature

$a_s$	Area of a completely fused aggregate (m <sup>2</sup> )
$a_0$	Surface area of ZrO <sub>2</sub> monomer (m <sup>2</sup> )
$A$	Total agglomerate area concentration (m <sup>2</sup> /kg <sub>gas</sub> )
$A_r$	Pre-exponential factor
$\beta$	Coagulation kernel (m <sup>3</sup> /s)
$B_l$	Break-up constant
$B_M$	Spalding mass transfer number
$c_p$	Heat capacity (J·kg <sup>-1</sup> ·K <sup>-1</sup> )
$C$	Molar concentration (kmol/m <sup>3</sup> )
$C_D$	Drag coefficient
$\Delta H^\circ_C$	Standard combustion enthalpy (kJ/L)
$d$	Diameter (m)
$D_b$	Grain boundary diffusion coefficient (m <sup>2</sup> /s)
$D_f$	Fractal-like dimension
$D_0$	Diffusion pre-exponential factor (m <sup>2</sup> /s)
$\varepsilon$	Mass flux fraction
$\eta$	Rate exponent

$E$	Activation energy (J/mol)
$F_M$	Film thickness correction factor
$\mathbf{F}$	Force vector (N)
$\gamma$	Surface tension (N/m)
$h$	Latent heat (J/kg)
$H$	Flow rate of enthalpy (J/s)
$k$	Forward rate constant
$\tilde{k}$	Thermal conductivity ( $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ )
$k_f$	Particle formation rate ( $\text{s}^{-1}\cdot\text{m}^{-3}$ )
$\lambda$	Wavelength (m)
$\mu$	Dynamic viscosity ( $\text{Pa}\cdot\text{s}$ )
$m$	Mass (kg)
$\dot{m}$	Mass flow (kg/s)
$M$	Mass exchange rate (kg/s)
$\nu$	Stoichiometric coefficient
$n$	Number
$N$	Total agglomerate number concentration ( $1/\text{kg}_{\text{gas}}$ )
$Nu_0$	Nusselt number
$\Omega$	ZrO <sub>2</sub> molar volume ( $\text{m}^3/\text{mol}$ )
$\Omega_w$	Wave growth rate ( $\text{s}^{-1}$ )
$\Phi$	Equivalence ratio
$Pr$	Prandtl number
$Q$	Heat (J)
$\rho$	Density ( $\text{kg}/\text{m}^3$ )
$R_g$	Universal gas constant ( $\text{J}\cdot\text{K}^{-1}\cdot\text{mol}^{-1}$ )
$R_k$	Global Arrhenius rate ( $\text{kmol}\cdot\text{m}^{-3}\cdot\text{s}^{-1}$ )
$Re$	Reynolds number
$Sc$	Schmidt number
$Sh_0$	Sherwood number
$Sh^*$	Modified Sherwood number
$SSA$	Particle specific surface area ( $\text{m}^2/\text{kg}$ )
$\tau_b$	Characteristic break-up time (s)
$\tau_s$	Characteristic sintering time (s)



$t$	Time (s)
$T$	Temperature (K)
$U$	Velocity vector (m/s)
$v_0$	Volume of ZrO <sub>2</sub> monomer (m <sup>3</sup> )
$V$	Total agglomerate volume concentration (m <sup>3</sup> /kg <sub>gas</sub> )
$w$	Grain boundary width (m)
$We$	Weber number
$\mathbf{x}$	Spatial position vector (m)
$X$	Mole fraction
$Y$	Mass fraction

#### *Sub-indexes*

$d$	Droplet
$g$	Gas
$gyr$	Gyration
$i$	Initial
$in$	Cell entry
$j,k$	Species index
$out$	Cell exit
$p$	Primary particle
$r$	Reaction index
$ref$	Reference
$s$	Droplet surface
$t$	Turbulent

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