

## Supporting information

### S1. The detailed boundary

*Inlet (Supercritical water): Velocity inlet boundary condition*

Velocity: 0.2563 m/s

Temperature: 660 K

Species mass fraction ( $\text{Ce}(\text{NO}_3)_3$ ): 0

Water: 1

*Inlet (Metal salt solution): Velocity inlet boundary condition*

Velocity: 0.013685 m/s

Temperature: 300 K

Species mass fraction ( $\text{Ce}(\text{NO}_3)_3$ ): 0.00326

Water: 0.99674

*Outlet (outlet vent boundary condition)*

Temperature: 300 K

Species mass fraction ( $\text{Ce}(\text{NO}_3)_3$ ): 0

Water: 0

*Inner wall boundary condition*

Coupled thermal boundary condition

Wall thickness (m)=0.00071

Heat generation rate ( $\text{w/m}^3$ ) =0

*Outer wall of the solid boundary*

Heat flux=0,

Wall thickness (m) =0

Heat generation rate ( $\text{w/m}^3$ ) =0

*Phase boundary conditions*

Reaction:  $(\text{CeO}_2)_{\text{phase 1}} = (\text{CeO}_2)_{\text{phase 2}}$

Reaction rate function: population balance

Drag coefficient:

Phase 2= phase 1 through schiller-naumann model

### S2. The finite rate-eddy dissipation model parameters

The mixture properties such as density, heat capacity, thermal conductivity and viscosity have been calculated using the mass averaged mixing law.

*Arrhenius rate*

Pre exponential factor= $7.0751 \times 10^{16}$

Activation energy (j/kgmol)= $1.6752 \times 10^8$   
Temperature exponent=0

*Mixing rate parameters*

A=4

B=0.5

### **S3. Population balance model parameters**

Number of bins=50

Ratio exponent=0.58

Minimum diameter=0.336 nm (ceria molecule size)

$K_v=0.5235988$

Initial conditions:

All bin fractions=0 ( there are no seed particles)

### **S4. UDFs for nucleation, diffusional growth rate, coagulation rate, and mass averaged density**

```
#include "udf.h"
```

```
#include "sg_pb.h"
```

```
#include "sg_mphase.h"
```

```
#include "mem.h"
```

```
/******UDF for nucleation rate******/
```

```
DEFINE_PB_NUCLEATION_RATE(nuc_rate, cell, thread)
```

```
{
```

```
  real J,c_ceria,c1,c2,c3,c4,c5,delta_g,f0,f1,f2,lamda_1,total_moles,total_vol,avg_mwt;
```

```
  real nav = 6.023e23; /* avagadro number */
```

```
  real pii=3.14;
```

```
  real int_tension=0.3; /*J/m2 surface energy ceria */
```

```
  real vmolecule = 1.943e-29; /* ceria molecule volume, m3 */
```

```
  real KB=1.38e-23; /*botlzman const, m2 kg s-2 K-1 */
```

```
  real pre_exp=1e34;
```

```
  real bulk_solub=1e-3; /* mol/m3, for ceria: */
```

```
  real T; /* line no 25 */
```

```
  Thread *tc = THREAD_SUPER_THREAD(thread); /*obtain mixture thread */
```

```
  Thread **pt = THREAD_SUB_THREADS(tc); /* pointer to sub_threads */
```

```
  Thread *tp = pt[P_PHASE]; /* primary phase thread */
```

```
  T = C_T(cell,tp); /* Temperature of primary phase in Kelvin */
```

```
  c1 = C_YI(cell,tp,0); /* hn03 massfraction, mwt=63. 01287, ro=1.504 */
```

```
  c2 = C_YI(cell,tp,1); /* ceria massfraction, Mwt ce02=172.115, ro=7.22 */
```

```
  c3 = C_YI(cell,tp,2); /* ceno3 massfraction, Mwt ceno33=326.116, ro=1.6 */
```

```

c4 = C_YI(cell,tp,3); /* h2 massfraction, mwt of h2=2, ro=0.00008189*/
c5 = C_YI(cell,tp,4); /* h2o massfraction, mwt=18, ro=1 */
total_moles = c1/63.01287+c2/172.115+c3/326.116+c4/2+c5/18; /* for 1gm of reaction
mixture */
total_vol= c1/1.504+c2/7.22+c3/1.6+c4/0.00008189+c5/1; /* units cc, for 1gm of reaction
mixture */
c_ceria= c2/total_vol*1/172.115*1e6; /* ceria concentration in mol/m3 */
lamda_1=c_ceria/bulk_solub; /* supersaturation */

f0=log(lamda_1);
f1= vmolecule/(KB*T*f0); /* post factor in critical gibbs free energy expression*/

delta_g=16/3*pii*pow(int_tension,3)*pow(f1,2); /* Critical Gibbs free energy, Reference:
Yoreo and Veklov */
f2=-(delta_g/(KB*T));

if (lamda_1 <= 1.)
{
J = 0.;
}
else
{
J = pre_exp*exp(f2);
}
return J;
}

/*****UDF for diffusional growth rate*****/

DEFINE_PB_GROWTH_RATE(growth_rate, cell, thread,d_1)
{
real
dif_coef,visc,cc_ceria,G,cc1,cc2,cc3,cc4,cc5,total_moles2,total_vol2,D_num,D_den,avg_mwt2,l
amda_2;
real pii=3.14;
real nav = 6.023e23; /* avagadro number */
real vmolecule = 1.943e-29; /* ceria molecule volume, m3: Molar volume= M/ro (Molecular
wt/density & Molecular volume
=Molar Volume/Navagrdo (172.115/ */
real KB=1.38e-23; /*botlzman const, m2 kg s-2 K-1 */
real d_molecule=2.69e-10;
real c_corect=2.5;
real bulk_solub=1e-3; /* mol/m3, for ceria: REFERENCE: Adschiri et al.,2001 */
Thread *tc = THREAD_SUPER_THREAD(thread); /*obtain mixture thread */
Thread **pt = THREAD_SUB_THREADS(tc); /* pointer to sub_threads */
Thread *tp = pt[P_PHASE]; /* primary phase thread */

```

```

real T;
  T = C_T(cell,tp); /* Temperature of primary phase in Kelvin */
  cc1 = C_YI(cell,tp,0); /* hn03 massfraction, mwt=63.01287, ro=1.504 */
  cc2 = C_YI(cell,tp,1); /* ceria massfraction, Mwt ce02=172.115, ro=7.22 */
  cc3 = C_YI(cell,tp,2); /* ceno3 massfraction, Mwt ceno33=326.116, ro=1.6 */
  cc4 = C_YI(cell,tp,3); /* h2 massfraction, mwt of h2=2, ro=0.00008189*/
  cc5 = C_YI(cell,tp,4); /* h2o massfraction, mwt=18, ro=1 */

total_moles2 = cc1/63.01287+cc2/172.115+cc3/326.116+cc4/2+cc5/18; /* for 1gm of reaction
mixture */
total_vol2=cc1/1.504+cc2/7.22+cc3/1.6+cc4/0.00008189+cc5/1; /* units cc, for 1gm of
reaction mixture */
cc_ceria= cc2/total_vol2*1/172.115*1e6; /* ceria concentration in mol/m3 */

lamda_2=cc_ceria/bulk_solub; /* supersaturation */

if (lamda_2 <= 1.)
  {
    G = 0.;
  }
else
  {
    visc=C_MU_L(cell,tp);
    D_num=KB*T*c_corect;
    D_den=3*pii*visc*d_molecule;
    dif_coef=D_num/D_den;
    G = 4*vmolecule*nav*dif_coef*cc_ceria/d_1;
  }
return G;
}

/*****UDF for coagulation rate*****/

DEFINE_PB_COALESCENCE_RATE(aggregation_kernel,cell,thread,d_1,thread_2,d_2)
{
real visc,brown_agg_kernel,shear_agg_kernel,agg_kernel,pre_factor;
real v1,v2,f1,f2;
real KB=1.38e-23; /*botlzman const, m2 kg s-2 K-1 */
real pii=3.14;
Thread *tc = THREAD_SUPER_THREAD(thread); /*obtain mixture thread */
Thread **pt = THREAD_SUB_THREADS(tc); /* pointer to sub_threads */
Thread *tp = pt[P_PHASE]; /* primary phase thread */
real T;
  T = C_T(cell,tp); /* Temperature of primary phase in Kelvin */

```

```

visc=C_MU_L(cell,tp);
pre_factor = 2*KB*T/(3*visc);
v1=pii/6*(d_1)*(d_1)*(d_1);
v2=pii/6*(d_2)*(d_2)*(d_2);
f1=1/pow(v1,0.3333)+1/pow(v2,0.3333);
f2=pow(v1,0.3333)+pow(v2,0.3333);

```

```

brown_agg_kernel=pre_factor*f1*f2;
real a1,b1,c1,grad_vel;
a1=C_U_G(cell,tp)[0];
b1=C_U_G(cell,tp)[1];
c1=C_U_G(cell,tp)[2];
grad_vel=pow((pow(a1,2)+pow(b1,2)+pow(c1,2)),0.5);
shear_agg_kernel=1/6*pow((d_1+d_2),3)*grad_vel;
agg_kernel=brown_agg_kernel+shear_agg_kernel;
return agg_kernel;
}

```

**/\*\*\*\*\*\*UDF for mass averaged density\*\*\*\*\*\*/**

```

DEFINE_PROPERTY(mass_wtd_rho,c,t)
{
  Material *sp;
  real rho;
  Property *prop;
  real sum = 0.;
  int i;

  mixture_species_loop(THREAD_MATERIAL(t),sp,i)
  {
    prop = (MATERIAL_PROPERTY(sp));
    rho = generic_property(c,t,prop,PROP_rho,C_T(c,t));
    sum += C_YI(c,t,i)*rho;
  }
  return sum;
}

```