

Simulation of thermal ignition of an ozone-oxygen mixture in a cylindrical vessel

Ralf Deiterding

Institute of Mathematics, Technical University Cottbus, Germany

e-mail: deiterding@math.tu-cottbus.de

Technical Report NMWR-00-4

Nov 15, 2000

1 Introduction

Developing multi-dimensional numerical methods for instationary chemically reacting gaseous flows is a challenging task. Validation is usually very time consuming and up to now only few comparative results for non-equilibrium chemistry are available.

In this report, we simulate the laser induced ignition process of an ozone-oxygen mixture in a cylindrical vessel. The laser pulse is modeled by an additional source term in the energy conservation equation. The flow in the vessel is cylindrical symmetric and can be described by two-dimensional conservation laws. This particular example has been proposed in [8] as a viscous computation, but comparable results can also be obtained by utilizing generalized Euler equations. The reaction mechanism is remarkable small and uses 3 different chemical species and only 6 elementary reactions. Qualitative correct numerical results can be obtained at moderate computational costs and can serve as a first test case for a two-dimensional method.

By treating the additional term that arises due to cylindrical symmetry within each conservation law as a nonstiff source term, each transport scheme that solves the two-dimensional cartesian equations can be employed. In this case, stiff source terms due to chemical reaction and nonstiff source terms have to be integrated simultaneously.

2 Governing equations

We consider the three-dimensional Euler equations with chemical reactive source terms. We assume a cylindrical symmetric flow that allows the dimensional reduction of the system of conservation laws. The equations then take the following form:

$$\begin{aligned} \partial_t \rho_i &+ \partial_r(\rho_i u) &+ \partial_z(\rho_i w) &= W_i \dot{\omega}_i - \frac{1}{r}(\rho_i u) & i = 1, \dots, K \\ \partial_t(\rho u) &+ \partial_r(\rho u^2 + p) &+ \partial_z(\rho u w) &= & - \frac{1}{r}(\rho u^2) \\ \partial_t(\rho w) &+ \partial_r(\rho u w) &+ \partial_z(\rho w^2 + p) &= & - \frac{1}{r}(\rho u w) \\ \partial_t(\rho E) &+ \partial_r[u(\rho E + p)] &+ \partial_z[w(\rho E + p)] &= \dot{q} & - \frac{1}{r}[u(\rho E + p)] \end{aligned}$$

We utilize a continuity equation for the partial density ρ_i of each gaseous species. The mass production rate due to chemical reaction is written as $W_i \dot{\omega}_i$ (see appendix A.1 for the actual

reaction mechanism). We denote the velocity in radial direction by u , the velocity in axial direction by w and the total energy per unit mass by E . The hydrodynamic pressure p is evaluated by Dalton's law for mixtures of ideal gases [2]. The additional source term \dot{q} in the energy conservation equation is used to model the ignition source.

3 Geometry

We use the two-dimensional domain $\Omega = \{(r, z) \mid 0 < r < R_0 \wedge 0 \leq z \leq Z_0\}$. The radius of the vessel is $R_0 = 4$ mm and its length is $Z_0 = 8$ mm.

4 Initial conditions

The computational domain is filled with a motionless perfectly stirred $O_2:O_3$ -mixture of molar ratio 0.28:0.72 at a pressure of 0.34 bar. The vector of state for the initial data is given in tab. 1.

ρ [kg m ⁻²]	0.50058
u [m s ⁻¹]	0.0
w [m s ⁻¹]	0.0
p [Pa]	34000.0
T [°K]	298.0
$X_{O_2} : X_{O_3} : X_O$	0.720000 : 0.280000 : 0.0
$Y_{O_2} : Y_{O_3} : Y_O$	0.631579 : 0.368421 : 0.0
W [kg mol ⁻¹]	$3.64786 \cdot 10^{-2}$

Table 1: Initial data within whole computational domain.

A temporary source term \dot{q} in the energy equation is used to model the thermal ignition process by a laser pulse in axial direction [8]. It is defined by

$$\dot{q} = \begin{cases} \frac{D_s}{\tau_s} \left(1 - \beta \frac{z}{Z_0}\right) \exp\left\{-\left(\frac{r}{r_s}\right)^8\right\} & \text{for } 0 \leq t \leq \tau_s \\ 0 & \text{for } t > \tau_s \end{cases} .$$

The thermal source \dot{q} is constructed as a smooth function linear in z that converges monotonically to 0 for $r > r_s$. During the initial heating period $t \leq \tau_s$ the source terms \dot{q} essentially rises the total energy density in a cylindrical ignition volume around the z axis with radius r_s . The parameters for the actual computation are

$$D_s = 3.2 \cdot 10^{-5} \text{ J m}^{-3}, \quad \tau_s = 1.0 \text{ } \mu\text{s}, \quad r_s = 1.37 \text{ mm}, \quad \beta = 0.4 .$$

At the end of the heating period at $t = 1.0 \text{ } \mu\text{s}$ the temperature has its maximal value at $r = 0, z = 0$ with approx. 1090°K. In order to simulate absorption [8], the energy deposition decreases in axial direction to approx. 802°K at $r = 0, z = Z_0$.

5 Boundary conditions

Symmetry boundary conditions at $r = 0$. Solid walls elsewhere.

6 Simulation

The simulation ends at $t_{end} = 20 \mu s$.

7 Comparative data

We employ a fractional step method and alternate between solving the homogeneous two-dimensional hydrodynamic transport equations and two different systems of ordinary differential equations that are integrated successively for stiff and nonstiff source terms. Within each grid cell the systems are solved separately with standard ODE methods. For the pure hydrodynamic transport we solve

$$\begin{aligned} \partial_t \rho_i &+ \partial_r(\rho_i u) &+ \partial_z(\rho_i w) &= 0 & i = 1, \dots, K \\ \partial_t(\rho u) &+ \partial_r(\rho u^2 + p) &+ \partial_z(\rho u w) &= 0 \\ \partial_t(\rho w) &+ \partial_r(\rho u w) &+ \partial_z(\rho w^2 + p) &= 0 \\ \partial_t(\rho E) &+ \partial_r[u(\rho E + p)] &+ \partial_z[w(\rho E + p)] &= 0 \quad . \end{aligned}$$

The wave propagation method of R. J. LeVeque [7] in combination with an approximate Riemann solver of Roe type for mixtures of real gases is utilized [4]. In the next step we integrate the nonstiff source terms due to symmetry and the ignition source using a two-step Runge-Kutta method. The system of ODE's that has to be solved is:

$$\begin{aligned} \partial_t \rho_i &= -\frac{1}{r}(\rho_i u) & i = 1, \dots, K \\ \partial_t(\rho u) &= -\frac{1}{r}(\rho u^2) \\ \partial_t(\rho w) &= -\frac{1}{r}(\rho u w) \\ \partial_t(\rho E) &= -\frac{1}{r}[u(\rho E + p)] + \dot{q} \end{aligned}$$

Finally, we integrate

$$\partial_t \rho_i = W_i \dot{\omega}_i(\rho_1, \dots, \rho_K, T) \quad i = 1, \dots, K \quad (1)$$

to incorporate the reactive source terms. ODE systems that arise in chemical kinetics are usually stiff, hence we employ a semi-implicit Rosenbrock-Wanner method of fourth order with automatic stepsize adjustment [5]. Note that ρ, E, u and w remain constant during integration of (1).

Whenever the temperature T has to be calculated from the conserved variables the implicit equation

$$\sum_{i=1}^K \rho_i h_i(T) - \mathcal{R}T \sum_{i=1}^K \frac{\rho_i}{W_i} - \rho E + \rho \frac{u^2}{2} + \rho \frac{w^2}{2} = 0$$

is solved utilizing Newton's method [2].

Numerical Fluxes	18.3%	
Source integration	49.2%	
Boundary update	21.0%	(19.9%)
Recomposition	4.3%	(0.3%)
Interpolation	1.2%	
Conservative Fixup	3.1%	(2.3%)
Clustering	0.4%	
Output	0.3%	
Not explicitly measured	2.2%	

Table 2: Decomposition of computational time for the entire calculation. The portions of parallel communication in relation to the whole computational time are displayed in brackets.

The reference computation employs a parallelized blockstructured adaptive mesh refinement algorithm (AMR) to reduce the computational costs [3, 1]. The adaptive calculation uses 2 levels of refinement with a uniform refinement factor of 2. The highest resolution corresponds to a uniform 640×320 grid. As refinement criterions scaled gradients of ρ , p , Y_{O_2} and Y_{O_3} are used. The computation is carried out in a parallel environment and requires 3.8 h real time on 6 nodes of a Pentium-III-PC-Cluster with 450 MHz. Tab. 2 shows the decomposition of the computational time for this calculation. Fig. 1-4 show the results of this reference computation. Exemplary snapshots of the blockstructured adaption are displayed in fig. 5.

References

- [1] M. Berger and P. Colella. Local adaptive mesh refinement for shock hydrodynamics. *J. Comput. Phys.*, 82:64–84, 1988.
- [2] R. Deiterding. Generalized euler equations with non-equilibrium chemistry. Technical Report NMWR-00-2, Brandenburgische Technische Universität Cottbus, Oct 2000.
- [3] R. Deiterding. Object-oriented design of an AMR-algorithm for distributed memory computers. In *Proc. of 8th Int. Conf. on Hyperbolic Problems, submitted*, Magdeburg, Feb. 2000.
- [4] B. Grossmann and P. Cinella. Flux-split algorithms for flows with non-equilibrium chemistry and vibrational relaxation. *J. Comput. Phys.*, 88:131–168, 1990.
- [5] P. Kaps and P. Rentrop. *Numerische Mathematik*, 33:55–68, 1979.
- [6] R. J. Kee, F. M. Rupley, and J. A. Miller. *Chemkin-II: A Fortran chemical kinetics package for the analysis of gas-phase chemical kinetics*. SAND89-8009, Sandia National Laboratories, Livermore, California, Sep 1989.
- [7] R. J. LeVeque. Wave propagation algorithms for multidimensional hyperbolic systems. *J. Comput. Phys.*, 131(2):327–353, 1997.
- [8] U. Maas and J. Warnatz. Simulation of thermal ignition processes in two-dimensional geometries. *Zeitschrift für Physikalische Chemie Neue Folge*, 161:61–81, 1989.

A Chemical kinetics

A.1 Reaction mechanism

The ozone-oxygen mechanism from [8] is displayed in tab. 3.

						A	β	E_{act}		
						[cm, mol, s]		[cal mol ⁻¹]		
1.	O	+ O	+ M	→	O ₂	+ M	0.290E + 18	-1.00	0.00	
2.	O ₂	+ M		→	O	+ O	+ M	0.681E + 19	-1.00	118645.00
3.	O ₃	+ M		→	O ₂	+ O	+ M	0.950E + 15	0.00	22706.00
4.	O ₂	+ O	+ M	→	O ₃	+ M		0.332E + 14	0.00	-1171.15
5.	O	+ O ₃		→	O ₂	+ O ₂		0.520E + 13	0.00	4158.77
6.	O ₂	+ O ₂		→	O ₃	+ O		0.427E + 13	0.00	98926.20

Third body efficiencies: $f(\text{O}) = 1.14$, $f(\text{O}_2) = 0.40$, $f(\text{O}_3) = 0.92$

Table 3: Ozone-oxygen mechanism from [8] in units for Chemkin-II.

A.2 Thermodynamic data

Necessary thermodynamic data are extracted from the Chemkin-II data base [6]. The employed thermodynamic fits are valid for a temperature range from 300°K to 5000°K.

	W [g mol ⁻¹]
O	15.999400
O ₂	31.998800
O ₃	47.998200

Table 4: Molecular weights of involved species [6].

A.3 Units

In our computational code we utilize the Chemkin-II library to evaluate chemical production rates. To avoid permanent conversion the code internally employs the fixed unit system determined by Chemkin-II [6]. In this report, we use SI-units for initial conditions and reference data. Tab. 5 gives the necessary factors to convert initial data and output into the Chemkin-II unit system.

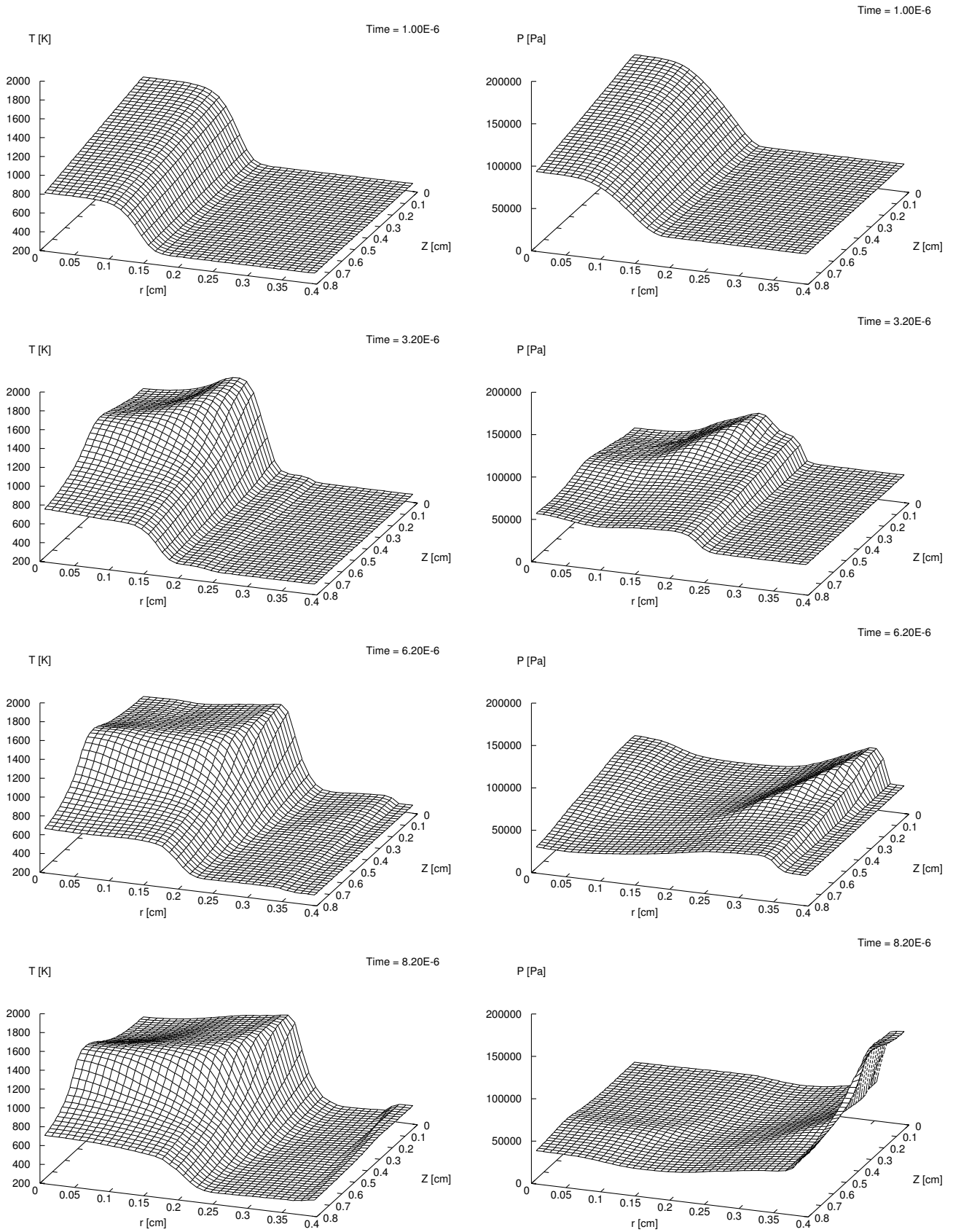


Figure 1: Distribution of T and p in 1st half of simulation time.

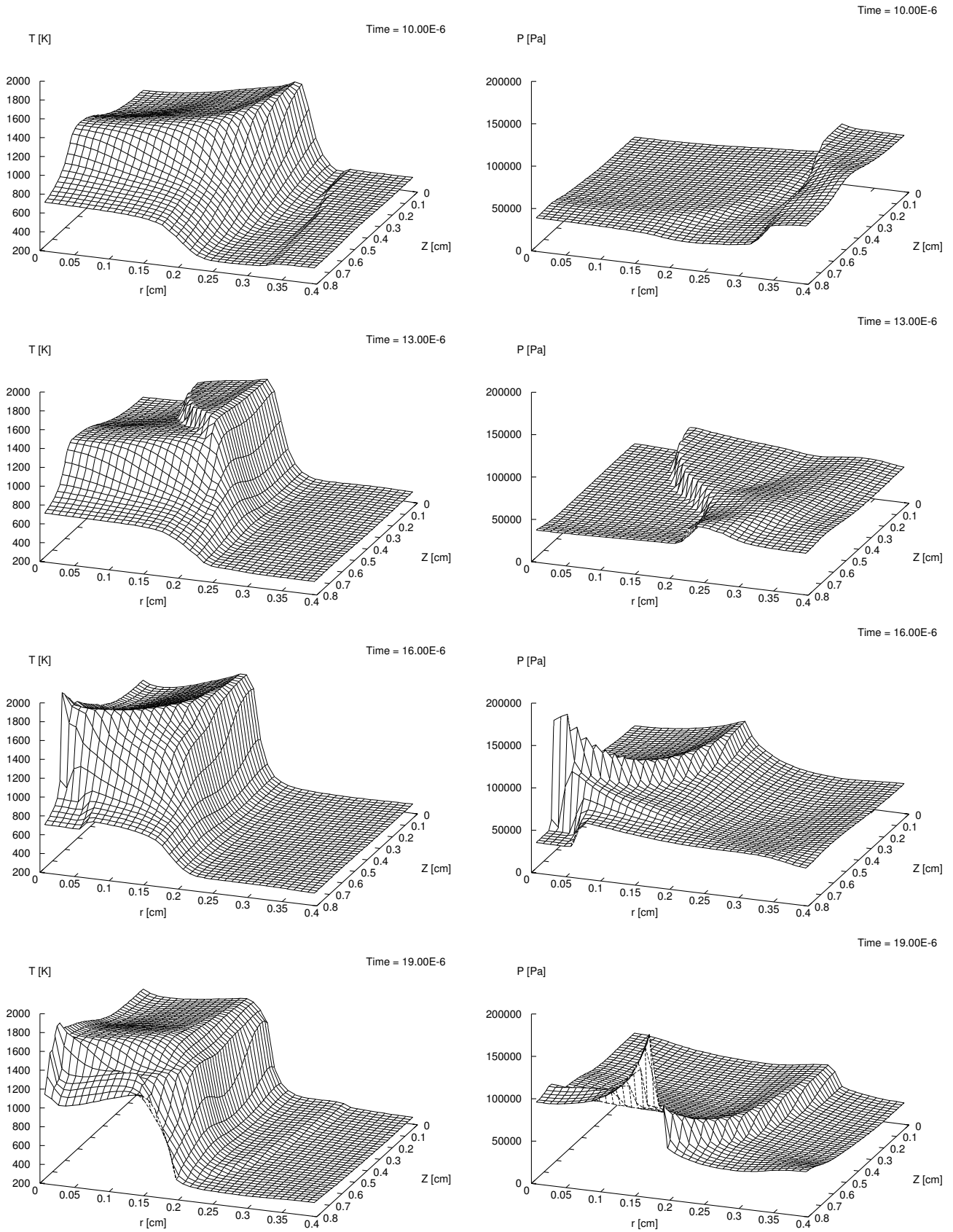


Figure 2: Distribution of T and p in 2nd half of simulation time.

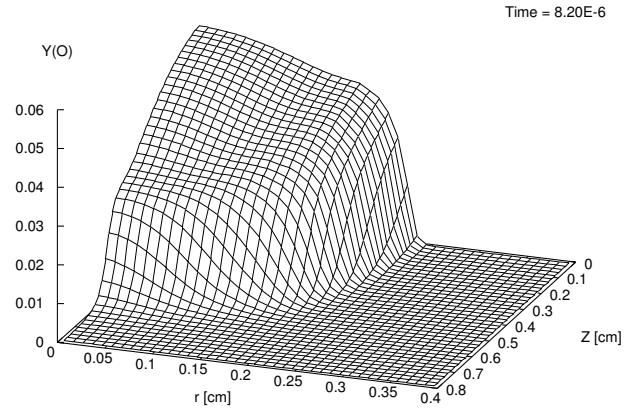
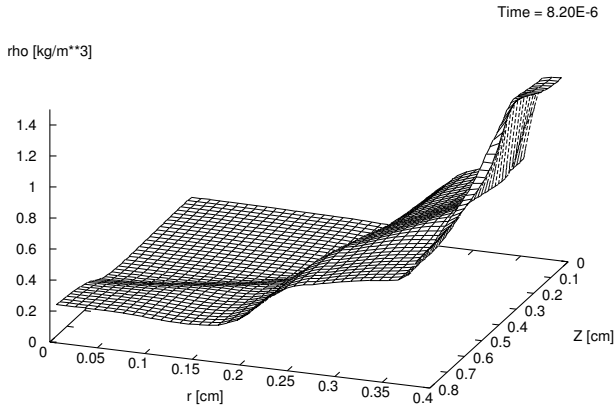
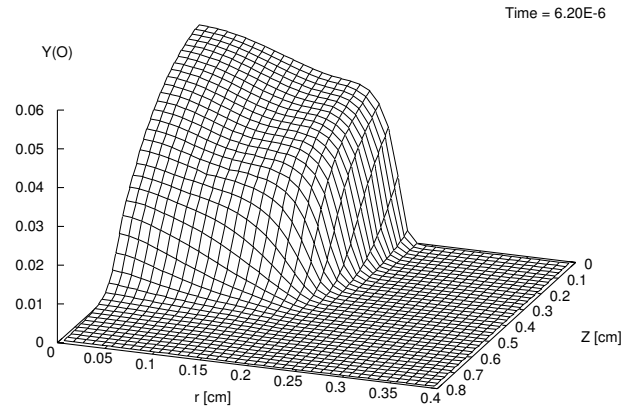
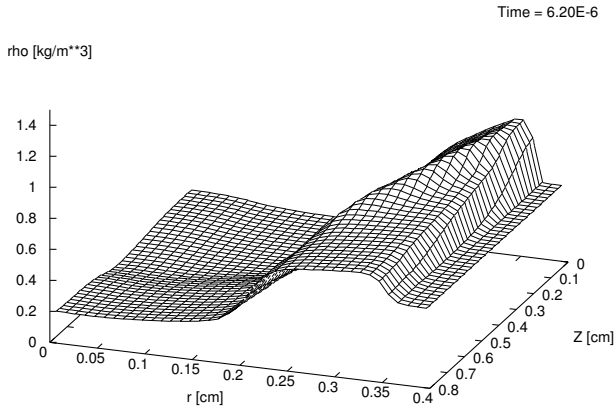
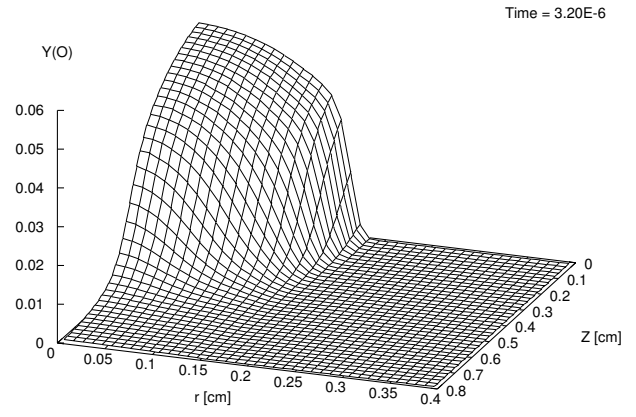
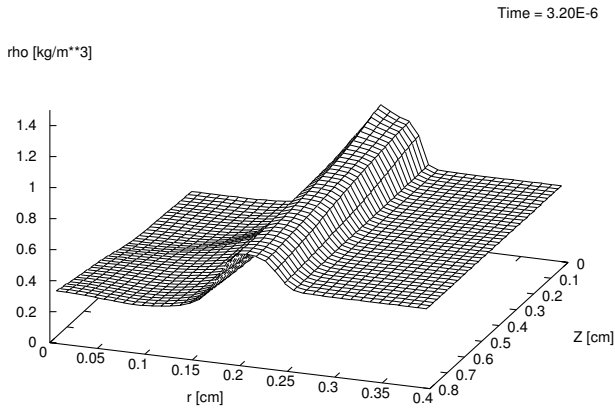
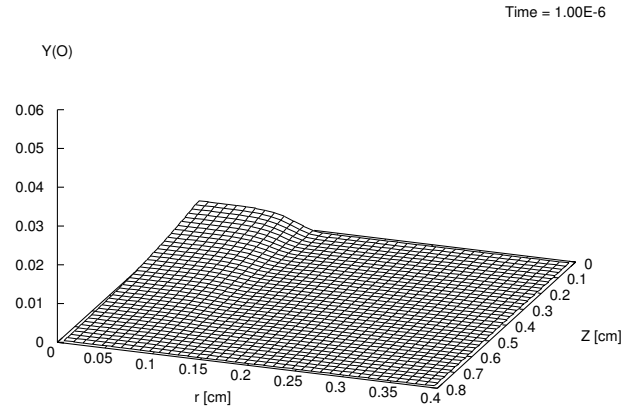
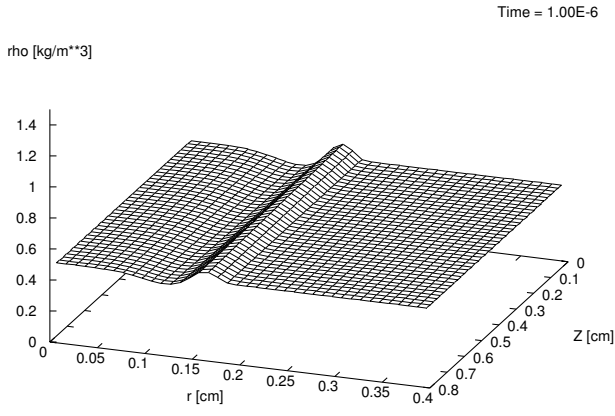


Figure 3: Distribution of ρ and Y_O in 1st half of simulation time.

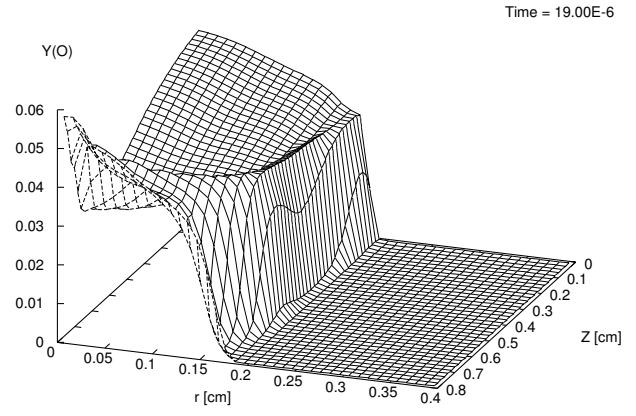
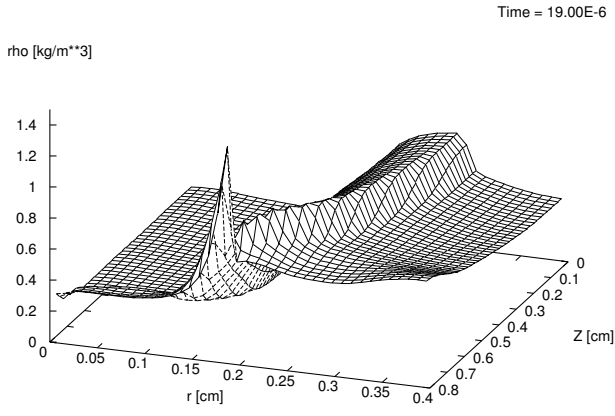
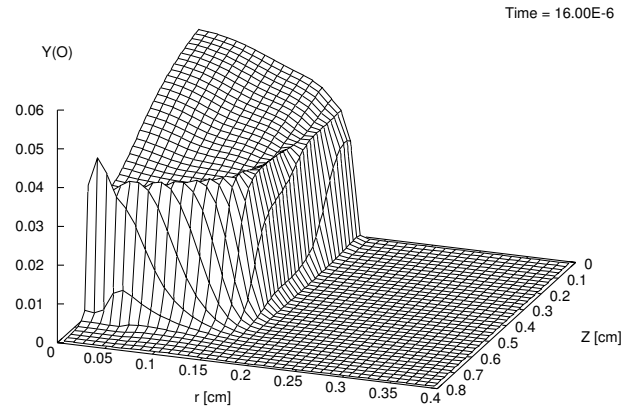
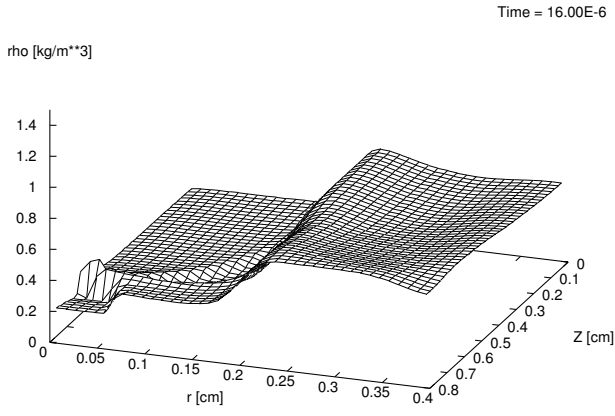
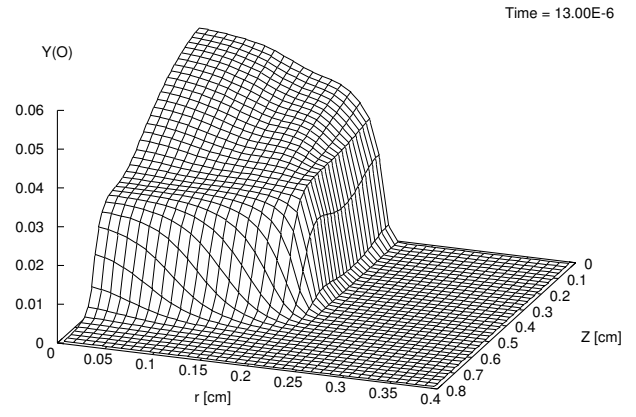
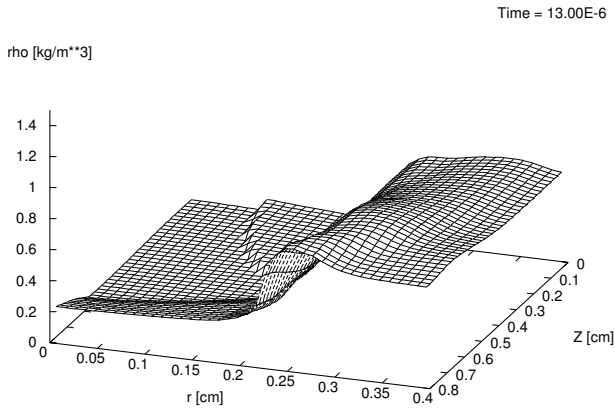
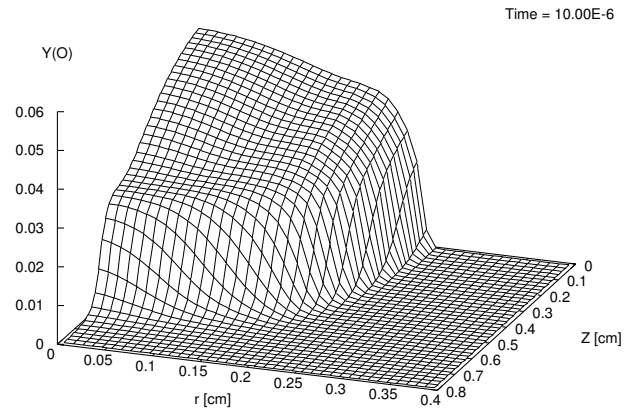
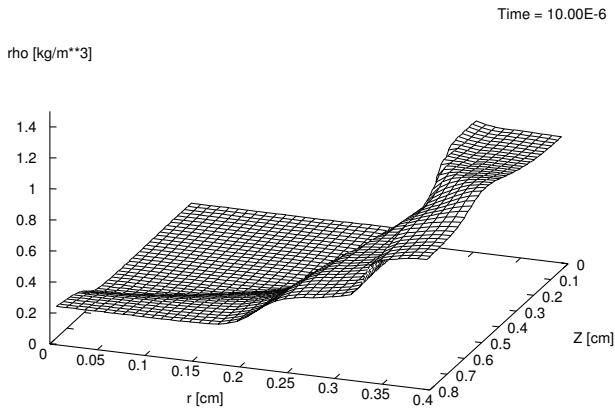


Figure 4: Distribution of ρ and Y_O in 2nd half of simulation time.

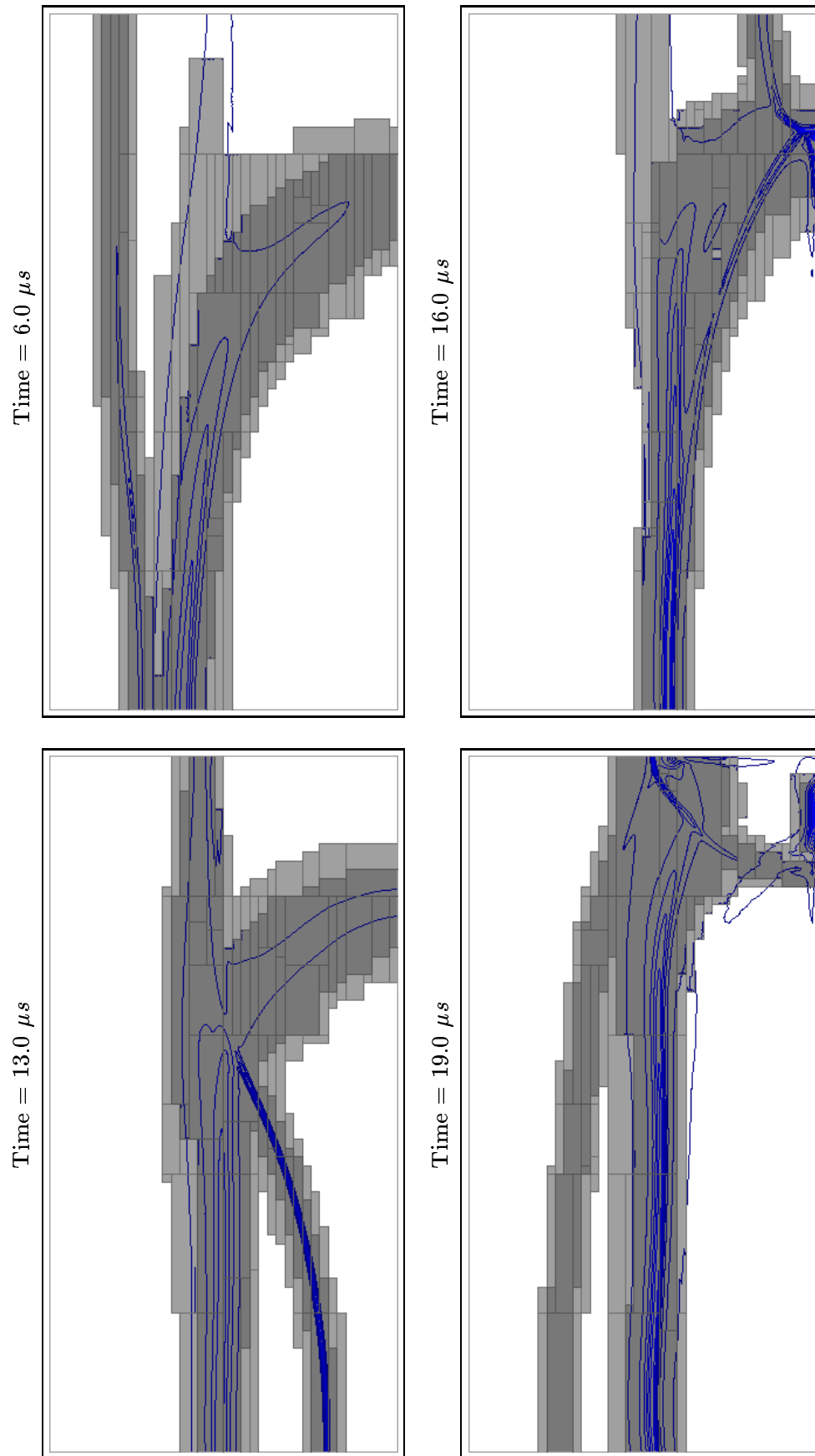


Figure 5: Isolines of density on 1st (light grey) and 2nd (dark grey) refinement level.

	Chemkin-II (g, cm, mol, s)	SI (kg, m, mol, s)	Factor [†]
ρ	g cm^{-3}	kg m^{-3}	10^3
u	cm s^{-1}	m s^{-1}	10^{-2}
e, E	erg g^{-1}	J kg^{-1}	10^{-4}
h, h^f	erg g^{-1}	J kg^{-1}	10^{-4}
c_v, c_p	$\text{erg g}^{-1} \text{K}^{-1}$	$\text{J kg}^{-1} \text{K}^{-1}$	10^{-4}
ρE	erg cm^{-3}	J m^{-3}	10^{-1}
p	dyne cm^{-2}	N m^{-2}	10^{-1}
T	K	K	-
W	g mol^{-1}	kg mol^{-1}	10^{-3}
$\dot{\omega}$	$\text{mol cm}^{-3} \text{s}^{-1}$	$\text{mol m}^{-3} \text{s}^{-1}$	10^6
\mathcal{R}	$8.31441 \cdot 10^7 \text{ erg mol}^{-1} \text{K}^{-1}$	$8.31441 \text{ J mol}^{-1} \text{K}^{-1}$	10^{-7}
A^*	$(\text{cm}^3 \text{mol}^{-1})^{r-1} \text{s}^{-1}$	$(\text{m}^3 \text{mol}^{-1})^{r-1} \text{s}^{-1}$	$(10^{-6})^{r-1}$
β	-	-	-
E_{act}	cal mol^{-1}	J mol^{-1}	4.18392
\mathcal{R}_{act}^{**}	$1.98723 \text{ cal mol}^{-1} \text{K}^{-1}$	$8.31441 \text{ J mol}^{-1} \text{K}^{-1}$	4.18392

[†] Conversion factor from units used in Chemkin-II into SI-units. * r denotes the reaction order.

** used for activation energy within Chemkin-II.

$1 \text{ erg} = 1 \text{ g cm}^2 \text{ s}^{-2}$, $1 \text{ J} = 1 \text{ kg m}^2 \text{ s}^{-2}$, $1 \text{ dyne} = \text{g cm s}^{-2}$, $1 \text{ Nm}^{-2} = 1 \text{ Pa} = \text{kg m}^{-1} \text{ s}^{-2}$

$1 \text{ erg} = 10^{-7} \text{ J} = 2.3901 \cdot 10^{-8} \text{ cal}$, $1.01325 \cdot 10^5 \text{ Pa} = 1.01325 \cdot 10^6 \text{ dyne cm}^{-2} = 1 \text{ atm}$

Table 5: Conversion of Chemkin-II- into SI-units.