

Spraylets - a Way to Simulate Spray Autoignition

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Abstract

A new approach for spray ignition simulation is presented. Since the numerical effort of simulating an evolving spray within a hot and pressurized ambient from shortly after injection until ignition is interminable, the problem is tackled by splitting the simulation into two tasks. A CFD package is used to calculate a two phase flow through a simulation domain based on a configuration model. From this two phase flow droplet track data and gas phase values are extracted to serve as variable boundary conditions for the second task, the one dimensional single droplet ignition simulations, which are fully transient and based on detailed chemistry, named *spraylet*. The obtained ignition delay times can be transformed into spatial distributions of ignition probability.

While the CFD part is handling spray formation, turbulence, temperature, pressure and global vaporization within the confinements, the *spraylet* calculations take care of droplet related physics and chemistry. The reworked model is able to reproduce single and multistage ignition and simulation can be performed beyond the limit of complete evaporation.

The software framework for this approach has been implemented using a dummy function as substitute for the *spraylet* calculations still under development. The dummy function, which is intended for testing purpose only, comprises a very simple ignition criterion based on a threshold temperature.

1. Introduction

Simulating spray combustion, prevalent in many technical applications, eyes are usually laid on jet breakup and spray formation, vaporization and mixture formation, flame propagation and formation of pollutants. Detailed modeling of the ignition process as the transition from the physical mixing process into the chemical reaction process is apparently underrepresented. Starting with a perfectly separated system of a liquid fuel and a gaseous oxidizer and aiming for a most homogeneous gaseous pre-mixture prior to combustion, the ignition process is of great importance in all high-pressure/high temperature applications as e.g. in LPP combustion in gas turbines or in HCCI combustion in internal combustion engines. In these cases ignition shall not happen until a minimal degree of pre-mixture is achieved to allow for lean but stable flames with lowest rates of formation of thermal nitric oxides. In most cases ignition is dealt with as a single step transition into combustion initiated through sufficient residence time of a mixture in hot ambiance. Recent research on single droplet self-ignition accompanied by microgravity drop tower experiments revealed that ignition of hot spray systems encounters complicated temperature and pressure dependent endothermic and exothermic precursor reactions finally leading to a hot flame [7, 10, 12, 13]. The induction of exothermic precursor reactions, often called cool-flames, is strongly temperature dependent and less dependent on pressure or mixture ratio. The burning lifetime of

these reactions leading to sufficient radical pooling and chain-branching reactions for hot ignition depends on ambient pressure and on the local mixture ratios more than on temperature. Ignition is the final result of a proper balance between the appropriate mixture ratio at appropriate temperature and after sufficient residence time. All this dependent on the given elevated pressure conditions [10]. Numerical studies on single droplet ignition of n-heptane in a virtually finite volume to serve a finite overall equivalence ratio showed that induction time is the shortest at a certain initial droplet size (30 μm at $\Phi=0.5$). This induction time coincides with droplet lifetime. Initial sizes larger than this size encounter longer induction times but the mixture will mostly burn in the diffusive regime while droplets smaller than this diameter also show longer induction times but the mixture will mostly burn in the lean premixed regime [6, 7].

In a technical spray it is obvious that droplet size distribution and droplet-droplet- as well as droplet-premixture interactions additionally affects the induction process. A couple of neighboring droplets e.g. form a rich but relatively cool mixture at the inbound volume and a leaner but warmer mixture on the outbound side. Smaller already vaporized droplets form an inhomogeneous lean gaseous mixture on which the larger droplets vaporization and ignition proceeds. Up to date the numerical simulation of self-ignition of single droplets of simple monocomponent or binary fuels is possible with satisfying detailedness regarding the physical

processes and the chemical kinetics. However, this already reaches the limits of acceptable numerical effort. Currently there is no numerical model that can reliably predict autoignition in an evolving technical spray. Some spray models with detailed kinetics were introduced by Gutheil but they assume the spray as uniform [4], mono- or bi-disperse and in a closed volume [5]. Brute force strategies to extend the single droplet 1-D simulation to sprays must fail because the numerical effort exceeds the limits of available and foreseeable computing power. Extrapolating the actual 1-D single droplet simulation efforts into spray simulations, taking the necessary grid resolution, the time steps, the number of droplets and the calculation volume into account would lead to a calculation time in the order of 10^9 years for a single injection cycle. Therefore this paper presents a numerical approach that keeps the droplet related physical and chemical detailedness on the 1-D level joining it to the 3-D information gained from commercial CFD-simulations on spray formation, vaporization and mixture formation. Proper coupling with exchange of transient boundary conditions allows for spray ignition simulations to predict the most probable instants of ignition.

2. Description of the Spray Autoignition Model

In general, modeling of spray ignition involves modeling of two phase flows. In most cases this will be a gas-liquid flow. The objective of a simulation of spray ignition is the prediction, if, when and where a (lean) spray will ignite within a hot and pressurized ambience. Thereby, flow velocities, turbulence, temperature, pressure, vaporization and low- and high temperature reaction kinetics have to be taken into account.

The model proposed is tackling the problem by splitting the task into two parts. The first part consists of a large scale CFD simulation. Within this simulation a so-called steady spray will be computed. More exactly it is the calculation of a steady state solution for a two phase flow within a simulation domain based on a configuration model. This can be a combustion chamber of a gas turbine, a cycle engine or an experiment. The simulation considers flow, turbulence, temperature and (global) vaporization on a coarse grid with cell sizes several orders of magnitude larger than a droplet. Chemistry is not taken into account.

The calculation of a two phase flow requires the numerical solution of conservation equations for the gas and the liquid phase simultaneously. The gas phase is treated in Eulerian frame of reference, just like a single phase flow. Regarding the liquid phase the simulation is based on the Discrete Droplet Method (DDM) by Dukowicz [2]. This is a statistical model, which operates by solving the ordinary differential equations for the track, momentum, heat and mass transfer. The model handles single droplets each being a member of a group of identical,

non-interacting droplets. Therefore each calculated droplet describes the behavior of the group it represents.

The droplets are introduced into the simulation domain with initial position, diameter, velocity and temperature. At this point nozzle/atomization sub models can be utilized, or the spray can be defined as fully developed using a spray pattern. The droplet-gas momentum exchange, turbulent dispersion and evaporation of droplets are considered. Secondary break-up of droplets and droplet collision/coagulation are neglected because the spray is assumed to be dilute. The vapor of the evaporating droplets is used as a source term of an additional transport equation for the vapor void fraction in Eulerian frame of reference.

The droplets are tracked in Lagrangian frame of reference through the numerical grid of the gas phase. Therefore the resolution of the droplet tracks can be orders of magnitudes finer than the grid size. Full two-way coupling between the gas and the liquid phase is taken into account. In configurations where the influence of the liquid phase on the gas phase is negligible the dispersed phase can be calculated independently.

To this point the simulation can be performed in the described manner using commercial CFD-codes. The packages enable the user to calculate a gas phase flow field, that is a vector field of velocities, and several scalar fields such as temperature, pressure and mixture fraction. Secondly an arbitrary number of droplet trajectories can be generated. By combining this data one can obtain the scalar field values along the droplet tracks in form of data tables. Due to the large number of droplets in a spray computer power is easily exceeded. To reduce the number of spraylets required to be calculated average trajectories can be aggregated for each size range from a bundle of trajectories. This is a promising approach for predicting ignition delay and location of sprays. However the precision depends on the size range classification of the droplets.

The generated data tables serve as input for 1-D simulations of droplet ignition on a microscopic scale. Backing the similar flamelet approach (0-D) we call this spraylet. This is the second part of the proposed approach to simulation of technical spray ignition.

The spraylet calculation simulates a single droplet in a fully transient way. The one dimensional grid covers a liquid and a gas phase separated by a phase boundary. The simulation considers the physical processes of heat exchange, evaporation and diffusion. The chemistry is treated using detailed or reduced kinetics as described elsewhere [12]. The species mass fraction and the temperature from the gas phase are synchronized against the global scalar field values from the large scale simulation. By using this variable boundary conditions within the spraylet code the droplet and its vicinity virtually moves along a trajectory within the large scale simulation. Finally,

after a certain ignition delay, the spraylet indicates ignition. These ignition delay times are the primary output of the spraylet module.

With the help of the trajectory data the ignition delay times can be mapped into points of ignition in the large scale simulation domain. Each spraylet calculation delivers one ignition location (in case ignition actually takes place in the spraylet domain). Thus with increasing number of spraylet calculations one gains a more and more accurate spatial distribution of probability density for the ignition event to take place.

2.1 Large scale CFD simulation, modifications and extensions

Simulating the two phase flow inside a domain based on a configuration model is a task that can be accomplished using commercially available software packages. For the demand on the model to be applicable to real combustion machinery this is very beneficial. Reflecting on different CFD environments there are some common problems that require extensions to the CFD packages.

2.1.1 Data export and phase coupling

The data required as input for the variable boundary conditions of the spraylet calculation is based on the gas- and dispersed liquid phase data of the large scale CFD simulation. In order to generate the droplet track data in combination with the scalar field values, data from both phases need to be combined. As mentioned before the grid resolution for the gas phase calculation is low against the droplet size in order to keep the numerical effort low. On the other hand the spatial resolution of the droplet tracks is high (same order of magnitude as the droplet diameter).

This may cause the scalar field functions of temperature, pressure and mass fraction to be uncontinuous along droplet tracks. When a highly resolved droplet trace crosses a boundary between two adjacent cells the corresponding scalars jump from the first cell value to the second. In order to smooth scalar field values along droplet tracks interpolation has to be done.

The simplest interpolation method, that is operating on n-dimensional and irregularly spaced data, is Inverse Distance Weighting (IDW).

Further details regarding this subject given in [3].

Using the IDW interpolation the gas phase data was coupled to the liquid phase track data, so that the resulting tables can be used as input to the spraylet calculations.

2.1.2 Droplet lifetime and ignition delay

As the simulation shall be applicable to lean prevaporized premixed combustion systems the spraylet code is able to calculate the autoignition process beyond droplet lifetime. This feature has to be considered in the CFD calculation too. When

computing the dispersed (liquid) phase the trajectories of the droplets are limited in length due to droplet lifetime. When, on the other hand, the spraylet code is able to compute autoignition events that occur at a later time, additional data from CFD is needed as spraylet boundary condition. In practice this means the trajectories have to be extended beyond complete evaporation of the droplet.

Extension of trajectories can only be established by elongation of droplet lifetime, since the dispersed phase is calculated in a Lagrangian frame of reference. A solution to this problem is the modification of the evaporation model within the CFD calculation. The standard evaporation model within CFD environments follow the D^2 -rule. After heating up to boiling temperature the temporal derivative of the droplet surface is assumed to be constant. Because the droplet scales with D^2 one gets $d(D^2)/dt = -k$. Here k is an evaporation coefficient. Integration yields to $D(t) = (C - kt)^{0.5}$. This model was modified in such a manner that an evaporating droplet shrinks until it reaches a size which won't be allowed to evaporate any further in the CFD calculation to prevent the droplet center point from vanishing, this in turn prevents the CFD code from losing track of the center point of the cloud of evaporated fuel. With this inert droplet core trajectories can be extended at will. The rest size is very small against initial droplet diameter, so that the mass defect can be neglected.

Most CFD Packages enable the user to implement self made sub-models by user defined functions. With the described modification a virtual vessel, in which a spraylet calculation takes place, can be tracked beyond evaporation. This enables handling of cases in which the ignition delay is longer than droplet lifetime.

2.2 Small scale spraylet simulation

The spraylet calculation is performed by the separate spraylet module which deals with just a single droplet-trajectory context which represents a size class of droplets. Each spraylet will be calculated independently from the others, so an arbitrary number of spraylets can be calculated in parallel.

The model used by the spraylet module is based upon a combination of the open vessel droplet ignition model [12, 13] with constant pressure and of the closed vessel droplet ignition model [7] with pressure variation and appropriate boundary conditions.

Due to the fact that the single droplets are part of the spray environment the open vessel model is enhanced by ambient boundary conditions and pressure variation introduced by the results of the CFD calculation depending on the application e.g. gas turbines or cycle-based combustion engines.

The model is fully transient. A fuel droplet is located at the center of a spherical domain (virtual

vessel) which on its part is surrounded by the ambient valid for the current moment and position of the droplet on its trajectory. The model domain is split into three sub-domains: the liquid phase, the spraylet gas phase and the ambient, the CFDs part of the gas phase, the so called CFD gas phase (Fig. 1 and Fig. 2).

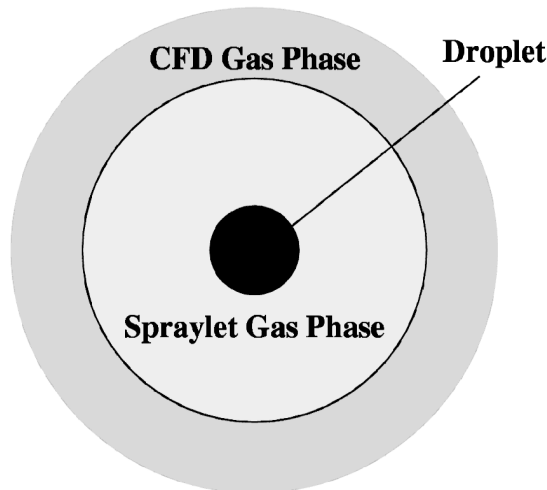


Fig.1 Arrangement of the three phases in the spraylet model.

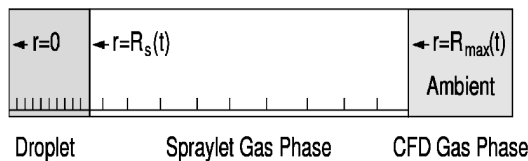


Fig.2 1D spraylet calculation domain.

The ambience has further influence on the process, as shown later, and may not only taken as an ordinary boundary condition as in the open vessel model. After complete vaporization only the two gas phases remain.

For the liquid and the spraylet gas phase mass, energy and species conservation are applied. Momentum conservation is neglected in both phases. A radial velocity inside the liquid and spraylet gas phase appears due to the consideration of convective transport mechanism induced by the evaporation process (Stefan flow). Radiant heat flux, viscosity and body forces are not taken into account.

2.2.1 Liquid phase

Inside the liquid phase no chemical reactions happen: $\dot{w}_i = 0$. The liquid phase consists only of fuel components. Diffusion of gas into the liquid phase is neglected. Concentration changes due to diffusion applying Fick's law based on the diffusivity that is calculated with a combination of the methods of Vignes and Tyn, Calus [9]. The mixture heat conductivity is calculated by the Filippov equation in case of a multi component fuel and the method of

Latini et al. otherwise. Heat capacity and absolute enthalpy calculation applies the Rowlinson method. Finally the Peng-Robinson equation is used for compression factor Z of the liquid. An equidistant grid is applied. Symmetric boundary condition at $r=0$ in the center of the droplet and phase boundary condition at the droplet surface are assumed. A description of these methods is given in [9].

2.2.2 Spraylet gas phase

In contrast to the CFD- and the liquid phase calculations the spraylet gas phase must consider the chemical reactions. Here, various reaction mechanisms can be implemented, detailed or reduced, for various fuels, single, two or multicomponent. Diffusion is calculated using the method of Oran and Boris [8] and diffusion coefficients are calculated by the working equation of Chapman and Enskog given in [9]. The heat conductivity of the mixture is calculated by the Coffee and Heimel method [1] and the conductive heat flux by $q_T = -\lambda_m \partial T / \partial r$. The cubic equation of state of Peng-Robinson is used. An unstructured grid is applied to the spraylet gas phase with an initially higher resolution near the droplet surface. Phase boundary condition at the inner and ambient boundary condition at the outer boundary are assumed.

2.2.3 Phase boundary

The surface temperature and species concentrations at the phase boundary are calculated by assuming fugacity equilibrium including the calculation of surface tension for the liquid-gas interface [9]. In addition the mass and species flux are applied at the liquid/gas boundary layer.

2.2.4 CFD gas phase

The CFD gas phase is a kind of abstractum. The scalar values representing this phase are larger scale information of the gas phase in the neighborhood extracted from the CFD calculations along the trajectory of the current spraylet. It is taken as the ambience of the spraylet. The data extracted from the CFD calculation along the trajectory are temperature, pressure, density of mixture and fuel concentration. The data are temporally interpolated from the table entries through linear approximation.

2.2.5 Coupling between the two gas phases

The variables of the CFD gas phase are handled in different ways. The value for pressure is taken from the CFD code for the spraylet gas phase due to the fact that pressure is a global attribute (for low velocities) and therefore the CFD code is the expert code for this item. Because chemical reactions are neglected in the CFD code temperature and therefore density can't be reproduced locally in the neighborhood of the spraylet. Hence the expert code (for local data) for temperature and density is the spraylet code. Because the temperature rise through

exothermal reactions happens locally and the spray is dilute no effect to the global gas phase is assumed until ignition. Finally the fuel concentration has to be dealt with. While the CFD gas phase contains a global fuel concentration, the spraylet gas phase is described by several species concentrations. This must be transformed into fuel concentrations by splitting each species into its fuel and air part followed by a weighted summation at each grid point.

Assuming that the CFD code and the spraylet calculation must describe the same situation at the droplet position on different length scales they have to be coupled for conformity. For realization both data sets become adopted and scaled to the same length scale. As the first step the CFD code must interpolate the data from its coarse grid to the position of the spraylet. In order to meet halfway the average of the fuel concentration in the spraylet gas phase (applying the trapezoidal rule) is compared to the value of the CFD gas phase. Before calculating a time step the average fuel concentration must be calculated and the size of the spraylet gas phase is adjusted as follows:

When the average fuel concentration of the spraylet gas phase is lower than the CFD-value which is the situation in **figure 3** it is assumed that the outer regions of the spraylet gas phase are outdated and will be dropped (**Fig. 4**). This to an extent until the average fuel concentration of the spraylet gas phase matches the value of the CFD gas phase.

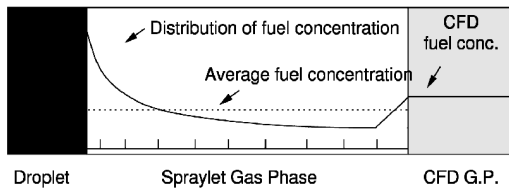


Fig. 3 Average fuel concentration of the spraylet gas phase is lower than the CFD-value (as a droplet moves into richer spray zones)

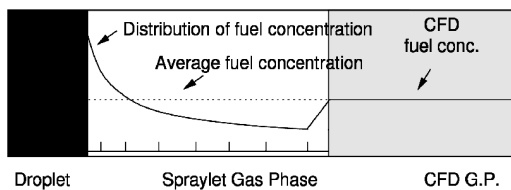


Fig. 4 Average fuel concentration matched spraylet gas phase size (richer case)

In the other case, when the average fuel concentration of the spraylet gas phase is higher than in the CFD gas phase (**Fig. 5**) the spraylet gas phase has to be extended until the average values match (**Fig. 6**). The species fractions are kept constant in the added region.

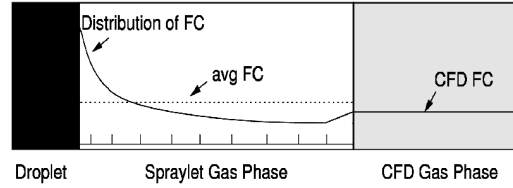


Fig. 5 Average fuel concentration of the spraylet gas phase is higher than the CFD-value (as a droplet moves into leaner spray zones)

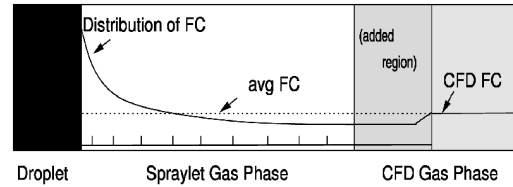


Fig. 6 Average fuel concentration matched spraylet gas phase size (leaner case)

As grid points are removed or added with proceeding time the spraylet gas phase grid has to be unstructured to avoid information losses due to interpolation. This would be necessary when keeping the grid structure or the applied metric. A special problem arises when the CFD fuel concentration is lower than the minimum of the distribution of the fuel concentration inside the spraylet gas phase (**Fig. 7**). In this case a 0 fuel concentration is added until the average values match again (**Fig. 8**).

The size of the added region will be limited by estimated species propagation speed and the maximal time step size that will be used for the next time step.

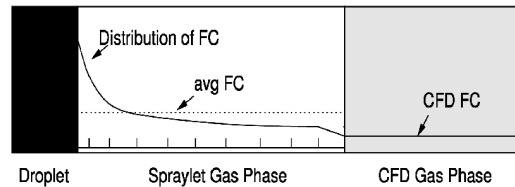


Fig. 7 CFD gas phase fuel concentration is lower than the minimal fuel concentration in the spraylet gas phase

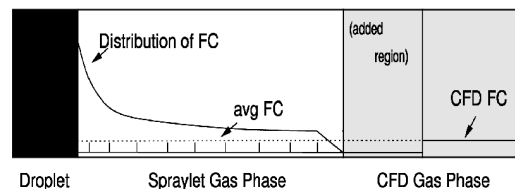


Fig. 8 Average fuel concentration matched spraylet gas phase size (0 fuel added zone)

3. Test Results

The described framework for spraylet based simulation of spray ignition has been implemented. However the spraylet model itself is work in progress by the time of this writing. Therefore a dummy module has been created as substitute for the spraylet module. The dummy module features the same interface regarding the external boundary conditions

and delivers ignition delay times, so that it can easily be replaced by the spraylet code.

Within this dummy module droplets traveling through a temperature field are monitored and whenever trespass a certain threshold temperature T_{ig} a timer is started. If a droplet moves into cooler regions below the threshold value before the timer reaches a certain value

t_{crit} nothing happens. Otherwise it ignites. After processing a certain number N of droplets the droplet with the lowest ignition time is marked. This is one virtual experiment. After processing M virtual experiments one gains a distribution of ignition times. With the droplet track data it is easy to map ignition times into ignition points. This gives a spatial distribution of ignition points.

Such a distribution is shown in Fig. 9. It shows the result of a simulation based on the experiment section of ZARM Hot Wind Tunnel (HWT) [11] (HWT setup: Jet-In-Crossflow with gas phase flow in direction of cylindrical axis (z-coordinate) and liquid phase injection upwards (y-coordinate), diameter=350 mm, $v_{inlet} = 75$ m/s, $T_{inlet} = 723.15$ K, $p = 2$ MPa, $v_{injection} = 180$ m/s, $T_{injection} = 300$ K).

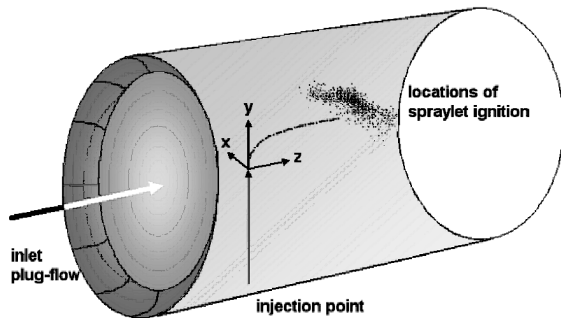


Fig.9 Simulation domain of HWT experiment section

Please note that this simulation does not claim to be realistic, because the dummy module is a not justifiable simplification of the spraylet calculation. This very simple dummy for the spraylet code depends only on the temperature information from the CFD. The distribution in Fig. 9 is based on 1000 virtual experiments, each consisting of 1000 tracked droplets, which are representing a Rosin-Rammler distributed spray (min. diameter $0.1 \mu\text{m}$, max. diameter $120 \mu\text{m}$, mean diameter $30 \mu\text{m}$, spread parameter 1, mass flow rate $= 35$ g/s). Ignition threshold temperature $T_{ig} = 720$ K and critical time

$$t_{crit} = 1 \text{ ms.}$$

From the distribution probability density functions in x, y and z direction with the simulation domain were calculated. The results are depicted in Fig. 10, 11 and 12.

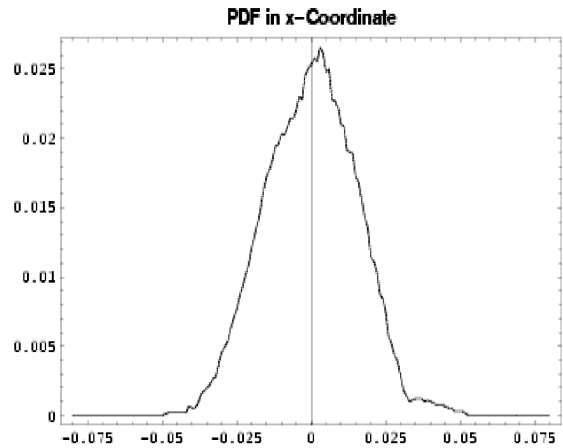


Fig.10 Probability density function for ignition probability. Spray simulation in HWT domain in x direction [m]

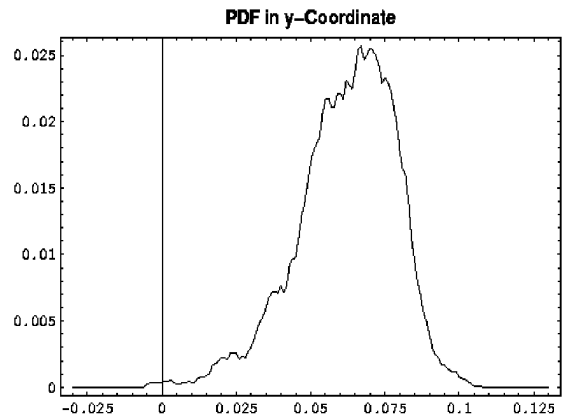


Fig.11 Probability density function for ignition probability. Spray simulation in HWT domain in y direction [m]

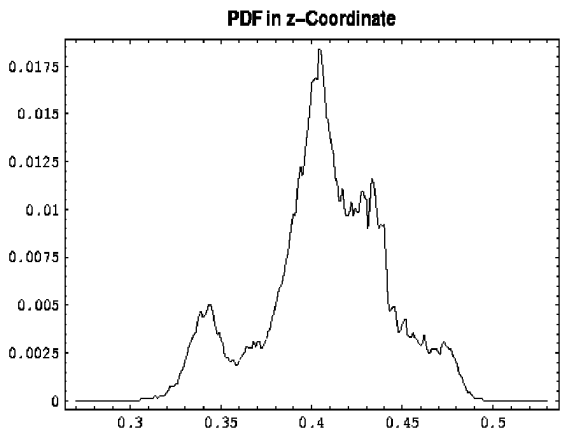


Fig.12 Probability density function for ignition probability. Spray simulation in HWT domain in z direction [m]

4. Conclusion

The split-tasks spraylet simulation enables the application of chemically detailed 1-D droplet ignition simulations to 3-D sprays in technical applications. With this the advantages of detailed but extensive micro-scale droplet calculation are made available for macro-scale spray simulations. It is

expected that reliable predictions of conditions for autoignition, knock or flashback are now generally possible. The validation by comparison with experiments is the next task. The split-tasks simulation is open for user defined reaction mechanisms and for the implementation of different droplet vaporization models including those for supercritical conditions.

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References

- 1) T.P. Coffee and J.M. Heimerl, Transport algorithms for premixed, laminar steady-state flames., *Combustion and Flame*, **43**, 273–289, 1981.
- 2) J. K. Dukowicz, A particle-fluid numerical model for liquid sprays., *Journal of Computational Physics*, **35**, 229–253, 1980.
- 3) N. I. Fisher, T. Lewis, and B. J. J. Embleton, Statistical Analysis of Spherical Data. , Cambridge University Press, 1987.
- 4) E. Gutheil, Numerical investigation of the ignition of dilute fuel sprays including detailed chemistry., *Combustion and Flame*, **93**, 239–254, 1993.
- 5) E. Gutheil, Numerical analysis of the autoignition of methanol, ethanol, n-heptane and n-octane sprays with detailed chemistry., *Combust. Sci. and Tech.*, **105**, 265–278, 1995.
- 6) O. Moriue, M. Mikami, N. Kojima, and C. Eigenbrod, Numerical simulations of the ignition of n-heptane droplets in the transition diameter range from heterogeneous to homogeneous ignition., *Proceeding of the Combustion Institute*, **30**, 1973–1980, 2004.
- 7) O. Moriue, S. Schnaubelt, Ch. Eigenbrod, and H.J. Rath, Numerical simulation of the ignition of a single fuel droplet in air with finite volume., *In Proc. 9th Int. Conf. on Liquid Atomization and Spray Systems*, ICLASS 2003, Sorrento, Italy, 2003.
- 8) E. S. Oran and J. P. Boris, Detailed modelling of combustion systems., *Progr. Energy Combust. Sci.*, **7**, 1–72, 1981.
- 9) R. C. Reid, J. M. Prausnitz, and B. E. Poling, The Properties of Gases & Liquids. McGraw-Hill, New York, 4th edition, 1988.
- 10) S. Schnaubelt, Numerische Analyse des Selbstzündungsverhaltens einzelner Brennstofftropfen. PhD thesis, Fachbereich Produktionstechnik, University of Bremen, 2005.
- 11) S. Schnaubelt, C. Eigenbrod, and H.J. Rath, Measurement of autoignition characteristics of technical fuel sprays under machine-identical conditions in a hot windtunnel., *Twenty-Ninth International Symposium On Combustion*, Abstracts of Work-In-Progress Posters, 405, 2002.
- 12) S. Schnaubelt, O. Moriue, T. Coordes, C. Eigenbrod, and H. J. Rath, Detailed numerical simulations of the multistage selfignition process of n-heptane isolated droplets and their verification by comparison with microgravity experiments., *Proceedings of the Combustion Institute*, **28**, 953–960, 2000.
- 13) M. Tanabe, M. Kono, J. Sato, J. Koenig, C. Eigenbrod, F. Dinkelacker, and H. J. Rath, Two stage ignition of n-heptane isolated droplets., *Combust. Sci. and Techn.*, **108**, 103–119, 1995.

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