

Treatment of differently weighted particles in reactive re-entry flows with DSMC

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Abstract. A molecular relaxation model, a dissociation model, and a recombination model for gas phase particles have been developed making use of the Conservative Weighting Scheme. The models are discussed in detail and are verified on basis of single cell simulations. Verification results show a strong increase of computational efficiency for the resolution of trace species' macroscopic properties. Additionally, several catalysis models are discussed with respect to expected pros and cons. Analysis results allow for identification of the most promising candidate. Implementation of catalysis is currently ongoing.

Keywords: dsmc, particle weight, high enthalpy flow, re-entry simulation

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1. INTRODUCTION

The Direct Simulation Monte Carlo (DSMC) method is a stochastic particle approach for the computational treatment of highly rarefied flows [1]. The basic idea of DSMC is to simulate gas flows as a particle stream whereas each simulated particle may represent a number of real particles. These particles move through the flow field and interact with walls and they perform collisions with each other. One feature of DSMC is the separation of movement and collision. Both movement and collision processes are treated subsequently. For the movement the classic equations of motion are solved deterministically. The critical modelling concerns the collision.

Two main DSMC mechanisms allow the simulation of particle systems on large spatial and temporal scales: One simulated particle represents a huge number of real particles, and particle interactions a reduced from a deterministic interaction principle to a probabilistic one. The former leads to a significant reduction of the total particle number N_{sim} , the latter reduces the computational load with respect to the particle number from a N_{sim}^2 to a linear dependency ($\sim N_{sim}$). In standard DSMC, each simulated particle represents W real particles, i.e. the physical particle density in a cell of volume V is represented by $n_{real} = N_{sim}W/V$ where the weighting factor W is mostly constant and the same for all particles of all species. However, in case there is a large difference between two mole fractions such that at least one species is considered as trace species, computation becomes either too inaccurate or too expensive. The former is caused by the small number of simulated trace particles which induces strong statistical scattering in the solution. Compensation by increase of simulated trace particle number increases significantly the overall computational load as the overall particle number N_{sim} is increased given that W is equal and constant for all particle of all species.

In 1994 Bird [1] introduced the varying particle weight principle. However, his Non-Conservative Weighting Scheme does not conserve momentum and energy exactly but only on average such that representative results can be obtained only under equilibrium conditions where many collisions take place.

Boyd introduced 1996 [2] the CWS method (Conservative Weighting Scheme). It contains a particle splitting, a collision, and a merging step. At the beginning, the *larger* collision partner is split into a particle with the same weight as the *smaller* collision partner, and a residual particle. During the collision step the equally weighted particles collide using standard DSMC algorithms. After assigning the post-collisional velocities to the collision partners the previously separated particles are merged. One can show that, in the merging step, the energy is not conserved exactly. The energy difference obeys the relation $\Delta E \sim \Phi(1 - \Phi)$ where $\Phi = W_j/W_i \leq 1$ is a measure of the particle weight difference. However, energy can be conserved *on average* by cumulating ΔE during each time step in each cell and by distributing the energy difference to equally weighted particles in the corresponding cell during a subsequent collision with $W = W_{max}$.

2. GAS PHASE INTERACTION PROBABILITIES

The general probability evaluation procedure differs from the well known *Constraint Probability (CP)* method in which the interaction numbers are computed first, followed by a loop over the currently considered interaction types (collision, excitation, ionization, molecular relaxation,...) and the corresponding interaction numbers. Instead, the implemented *Natural Sample Size (NSS)* method evaluates within a loop over *all* collision pairs in the given cell the individual collision probabilities P_c for each pair. If positive (i.e. if $P_c > R$ where R is an equally distributed random number), the correct interaction type is identified [3] and carried out. On average, both methods CP and NSS lead to the same collision rates.

The collision probability is computed via the standard scheme for differently weighted particles as introduced by Bird [1] and reads for a simple gas

$$P_c = \frac{N^2}{2} \frac{W}{S_{NSS}} \frac{\Delta t}{V} \sigma g \quad (1)$$

with S_{NSS} representing the number of pairs which equals in the simplest case $N/2$. However, having differently weighted particles, W needs to be replaced by the larger particle weight.

The relaxation probabilities do not depend on the individual particle weights. In fact, at higher energies the individual probabilities for rotational and vibration relaxation depend only weakly on the collision energy such that, for the sake of simplicity, they can be assumed constant ($P_{rot} = 0.2, P_{vib} = 0.02$), see e.g. [3].

The reaction probabilities also do not depend on particle weights. This allows to decide on reactive interactions right before any split or merge algorithms are executed. General details on the implemented reaction probability models which are used for the reaction of equally weighted particles can be found e.g. in [3, 4].

3. GAS PHASE INTERACTION MODELS

3.1. Relaxation

In [2] Boyd points out that inertial energy exchange for weighted particles can also be simulated using the splitting-merging technique. Care needs to be taken for the merging process of a molecule. Rotational and vibrational energies should be distributed without violating energy conservation. This can easily be realized for the rotational degree of freedom. However, for vibration the situation is different due to its discrete nature. In fact, the exactly conserved vibrational energy leads mostly to non-existing energy states. For instance, let $i = 1.7$ describe the numerical energy state according to a given vibrational energy value as a consequence of the merging process. By comparison of 0.7 with an equally distributed random number $R \in [0, 1]$ it is decided which state is occupied. This fast algorithm does not conserve energy exactly ($\Delta E_v \neq 0$), but on average over many collisions under equilibrium conditions.

3.2. Reaction

Wu et al. [5] presented a model for dissociation of gas phase molecules and the adsorption of particles. Adsorption is a sub-element of catalysis which is discussed in a subsequent section. In this section, the models for gas phase dissociation and recombination are discussed.

3.2.1. Dissociation

Dissociation is a two body problem in which either one or both particle(s) can dissociate (given that both particles are molecules and for both molecules a dissociation reaction has been defined). One can identify four relevant configurations when modelling differently weighted particles. However, the configuration can be reduced to two if the collision partner is defined non-reactive whereas only the first particle is reacting (dissociating). In short, the instantaneous dissociation of both molecules is neglected here.

The remaining two configurations regard the distribution of the particle weights. Note, the dissociating particle is called *first* here although index-wise it can be the second stored in the memory. Also and for the sake of simplicity, the collision partner is being considered an atom although the same algorithm applies if the collision partner is a molecule

1. $W_1 > W_2$: Let's assume $W_1 = 100$ and $W_2 = 1$. After the molecule is split into $W_1' = 99$ and $W_1'' = 1$, the dissociation is treated with standard DSMC algorithms leading to three particles (all atoms) with $W_2 = W_3 = W_4 = 1$. Since $W_1'' = 1$ does not exist any more, $W_1' = 99$ cannot be re-merged. Therefore, this configuration of particle weights generates at least one additional particle. Even more particles might be generated due to after-splitting which is an additional splitting process of the reaction products: If e.g. $W_3 > W_3^0$ where W_3^0 represents the initially defined particle weight of the species to which this particle with index 3 belongs, then W_3/W_3^0 new particles are created.

After-splitting is based on a division operation instead of a subtraction (as in the pre-splitting). This process was introduced in order to resolve trace species in case one (or more) of the products belong(s) to a previously defined trace species. However, in the given implementation mass is not conserved exactly. If e.g. $W_3/W_3^0 = 10.3$ then it is randomly decided whether 10 or 11 particles are created. Therefore, mass is conserved only on average over many dissociation reactions.

2. $W_1 < W_2$: Let the collision partner be split into $W_2' = 99$ and $W_2'' = 1$. Now, the collision partner is re-merged after the dissociation of the first particle. Without after-splitting this chemical reaction obeys the same particle number growth as in standard DSMC.

Two core rules have been followed in order to make the potentially occurring variations of weight pairings treatable:

1. The collision partner never reacts and is always re-merged in case it has been split.
2. The dissociation products are always evaluated regarding after-splitting but are never merged with other particles.

3.2.2. Recombination

The non-radiating atom recombination is considered here where the recombination energy is assigned to a randomly chosen (third) particle in the vicinity of the short-living pseudo particle composed of two atoms [3].

The same two core rules as in case of dissociation apply here which reduces the eight potentially occurring particle weight configurations to four. In the following we consider the third particle an atom although it can be a molecule as well. A third rule applies for the recombination process: The third particle is also split (if necessary) and, if split, later re-merged. In fact, splitting is based on the minimal particle weight such that even two out of three particles might be split at the beginning. Hence, this model of recombining particles with different particle weights mostly leads to an increased number of particles in the simulation domain instead of reducing it.

1. $W_{1,2} = W_{min} \neq W_3$: Only the third particle is split and re-merged after the recombination.
If no after-splitting takes place, the particle number is reduced by one.
2. $W_2 = W_{min} \neq W_3$ or $W_1 = W_{min} \neq W_3$: The two larger particles are split on basis of W_{min} yielding two new particles of which one will be re-merged (W_3) and the other will remain in the system (W_1' or W_2'). Splitting is done using the already described algorithms. Recombination as such is treated with standard DSMC.
The overall particle number might stay constant if the recombined molecule does not undergo after-splitting.
3. $W_{min} = W_3 < W_{1,2}$: Both educt particles are split in order to process the recombination with standard DSMC algorithms. However, since the residuals of the educts are probability-wise supposed to undergo some kind of interaction, an additional elastic scattering process is applied to them. Without after-splitting, this case leads to an increase of simulation particle number by one.
4. $W_{min} = W_{1,3} \neq W_2$ or $W_{min} = W_{2,3} \neq W_1$: The larger educt is split in the usual way.
The overall particle number remains constant without after-splitting.

4. CATALYSIS MODELS

Catalysis describes the adsorption of atoms by the surface and the subsequent recombination, followed by an emission of the created molecule. Both adsorption and recombination increase the heat transfer to the surface and are crucial

for the heat load estimation during atmospheric high enthalpy manoeuvres. The most important catalysis mechanism is the Eley-Rideal (ER) mechanism which is based on the recombination of an adsorbed atom with an gas phase atom, followed by molecule emission.

In fact, standard DSMC models for surface chemistry need to be extended in case of $W_{in} \neq W_{ad}$ where the indices describe the *incoming* and the *adsorbed* particle. Several ER catalysis models are proposed and discussed here of which one has been chosen for implementation and evaluation. All three models are based on the assumption that sheath growth is independent of the target material. This implies that *all* atomic gas phase particles (e.g. N and O in air) can be adsorbed regardless of their species assignment as long as their species dependent adsorption coefficients are non-zero, and the already adsorbed particles of species i do not affect the adsorption coefficient of species j and vice versa. The latter assumes that the adsorption of gas phase particles does not affect surface properties.

Independently of the weight configurations, one needs to distinguish between the adsorption and the recombination. In standard DSMC, the adsorption coefficient $\alpha \in [0, 1]$ is compared to a random number $R \in [0, 1]$ in order to decide whether adsorption occurs. Once an atom sticks to the surface the adsorption is prohibited until the surface element is freed by recombination. For detailed recombination probability modelling in standard DSMC please refer to [3].

The following strategies for the treatment of differently weighted particles in DSMC catalysis are discussed:

1. Assuming air as environment, both N and O are abundant species which can be initiated with maximum particle weight. Therefore, adsorption and recombination can be treated with standard DSMC algorithms, adding just an after-splitting step right after molecule emission in order to resolve the NO species.

Pros:

- (a) This approach demands minimum modification in the code.

Cons:

- (a) Neither adsorption, nor recombination are resolved better over time since N and O have initially large weights. In fact, due to splitting without re-merging in chemical reactions, the particle weights of N and O might be different. Therefore, either W_{in} , or W_{ad} , or or both would need to undergo pre-splitting, otherwise all pairings where $W_{in} \neq W_{ad}$ would have to be ignored in terms of catalysis. Therefore, weight configurations in which $W_{in} \neq W_{ad}$ demand a more smooth like adsorption and recombination process which would increase the resolution over time of both processes. In [5], gas phase adsorption was modelled on basis of non-vanishing adsorption coefficients which might be much smaller than unity. In order to speed up computation Wu et al. proposed to split the incoming particle on basis of α and to adsorb *every* particle according to αW_{in} whereas the residual $(1 - \alpha)W_{in}$ remains in the gas phase.

2. In order to apply standard DSMC algorithms to the ER mechanism $W_{in} = W_{ad}$ needs to be fulfilled naturally (first group), or enforced (second and third group). Therefore, gas phase atoms are allowed to be adsorbed constantly according to their adsorption coefficients [5]. Recombination is prohibited unless a certain weight is reached by the adsorbed species, cumulated such that each adsorbable species is represented by one adsorbed particle with growing W_{ad} . Hence, the active site is *free* until a pre-defined particle weight $W_{ad,max}$ has been reached for the pre-defined ER reaction. Once $W_{ad} \geq W_{ad,max}$, the active site is defined *not free* for adsorption such that only recombination steps can take place. Given that, potential recombination pairs are evaluated according to P_{rec} . If $P_{rec} > R$, the eventually bigger gas phase particle is split and recombination is carried out. The residual remains in the solid phase. Once $W_{ad} < W_{ad,max}$, adsorption is allowed and recombination is prohibited again.

Pros:

- (a) This model generally allows for $W_{in} \neq W_{ad}$ configurations.
- (b) Sheath growth (and energy and momentum transfer) is much better resolved.
- (c) The user has control over the degree of resolution.
- (d) Introducing a weight limit simplifies the decision path regarding the adsorption *and* recombination probability of the incoming atom. Only one process can take place by definition.
- (e) Limiting the sheath growth is reasonable as the number of active sites available for adsorption are limited [6].

Cons:

- (a) The adsorption is treated differently than the recombination step which represents an inconsistency. As a consequence, recombination as such is resolved not necessarily better than in standard DSMC.
- (b) Pre-defining a particle weight for one (or all) surface reaction(s) makes the approach rigid since the ideal $W_{ad,max}$ value is not known a priori.
- (c) Allowing for $W_{ad,max}$ related weight adaptation would not necessarily neutralise the aforementioned issues.

- (d) During adsorption phase, recombination is prohibited. During recombination phase, adsorption is prohibited. This might affect the resolution of both processes in a negative way.

This method should lead on average to a slight net increase of simulation particle number compared with standard DSMC catalysis (see [3]) where particles are adsorbed completely and remain adsorbed until recombination occurs. Contrarily to that, particle matter is adsorbed here more or less constantly without reducing the simulation particle number in the simulation domain. However, certain downsides of the second model might be addressed in the following way:

3. The same as model 2 but both processes adsorption and recombination are allowed to take place concurrently. The decision is trivial in case $W_{ad} \geq W_{ad,max}$ or $W_{ad} = 0$. In all other cases it is reasonable to primarily check for recombination since adsorption can occur mostly at any time, i.e. with a very high probability.

Pros:

- (a) Adsorption and recombination should be resolved much better.
- (b) Sheath growth is still limited.
- (c) Decision path concerning adsorption vs. recombination is clear.

Cons:

- (a) $W_{ad,max}$ is unknown a priori but still needs to be defined, see above.
- (b) P_{rec} is computed before any pre-splitting (same as in gas phase chemistry). However, if the gas phase particle is split to undergo recombination, the residual remains in the gas phase without any further interaction. This is currently not covered by the positive evaluation of P_{rec} which is independent of the particle weight.
- (c) The solution to the aforementioned drawback is trivial for $W_{in} < W_{ad}$. However, it is non-trivial for $W_{in} > W_{ad}$: According to which rules should the incoming particle be split? In a first approximation, the non-trivial configuration can be ignored.

Due to these considerations the third model has been chosen for implementation. This activity is ongoing.

5. VERIFICATION RESULTS

In this section the models and their implementations are verified. The collision model is well established such that we focus on the verification of the relaxation and reaction models only. All simulations are based on thermal non-equilibrium states which relax towards an equilibrium point inside an adiabatic box filled with a $N - N_2$ mixture.

5.1. Relaxation

Molecular nitrogen N_2 represents the abundant species (mole fraction $\Psi_{N_2} = 0.9999$), N is the trace species ($\Psi_N = 0.0001$). Initial temperatures are $T_N = 1000\text{ K}$, $T_{N_2, tra} = 10000\text{ K}$, $T_{N_2, rot} = T_{N_2, vib} = 100\text{ K}$.

Figure 1 (a) shows the successful equilibration towards an equilibrium temperature. As expected, the translational degree of freedom (DOF) relaxes with maximum rate, followed by the rotational DOF. The vibrational DOF equilibrates with minimum rates.

Figure 1 (b) shows a relevant signal obtained with the new approach with respect to standard DSMC. The models are abbreviated with vPW and ePW according to *varying Particle Weight* and *equal Particle Weight*. Essential simulation differences are summarized in table 1.

The additional algorithms increase the simulation time per particle by approx. 30% whereas the overall simulation time is nearly the same given the same iteration numbers. The difference in the translational temperature signal is depicted on the right in figure 1. The noise level in standard DSMC is very high due to the small number of atoms in the box. Scattering amplitude is about 30 – 40% with respect to the improved signal. Given that, a simulation

TABLE 1. Comparison of simulation time for standard DSMC (ePW) and the new model (vPW).

	$W_{N_2} / 10^6$	$W_N / 10^6$	$N_{N_2} / -$	$N_N / -$	Total time / s	Time per particle / $10^{-3}s$
<i>ePW</i>	3.5	3.5	~ 550000	~ 55	949	~ 1.8
<i>vPW</i>	10	10^{-3}	~ 200000	~ 200000	994	~ 2.4

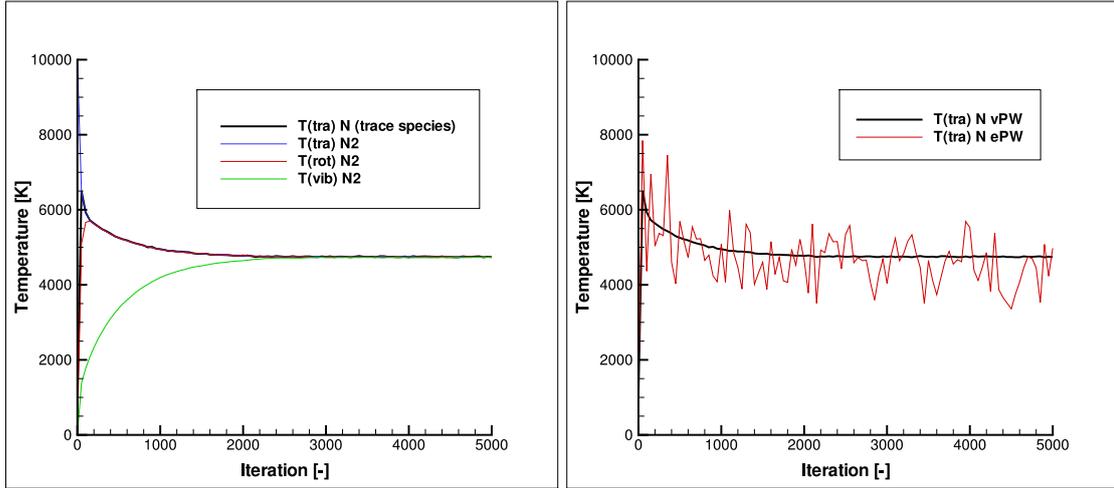


FIGURE 1. (a) Temperature convergence for the non-reactive test case. (b) Translational temperature signal: new model (*vPW*) vs. standard DSMC (*ePW*).

with standard DSMC leading to the same signal quality as *vPW* would last about 1000 *h* due to an estimated amount of 2×10^9 N-particles in the system. Despite the increased mean computational time per particle, the *vPW* based computational speed-up for the current test case is about 99.97%.

5.2. Dissociation

Similar but not identical parameters have been chosen for verification simulations, only the differing values are mentioned. Also here, atomic nitrogen *N* is the trace species. The initial temperature T_{tra,N_2} has been changed to 8000 K. In the new model, *N* has a particle weight of $W_N = 10^4$ leading to a particle number of $2 \cdot 10^5$. In comparison, $W_{N_2} = 10^7$ leads to the same number of particles.

The left in figure 2 shows the evolution of the temperatures of trace species *N* and background species *N*₂ over time. Again, all temperatures converge towards one single temperature value with qualitatively correct relaxation rates. A more detailed analysis will be addressed in subsequent research activities.

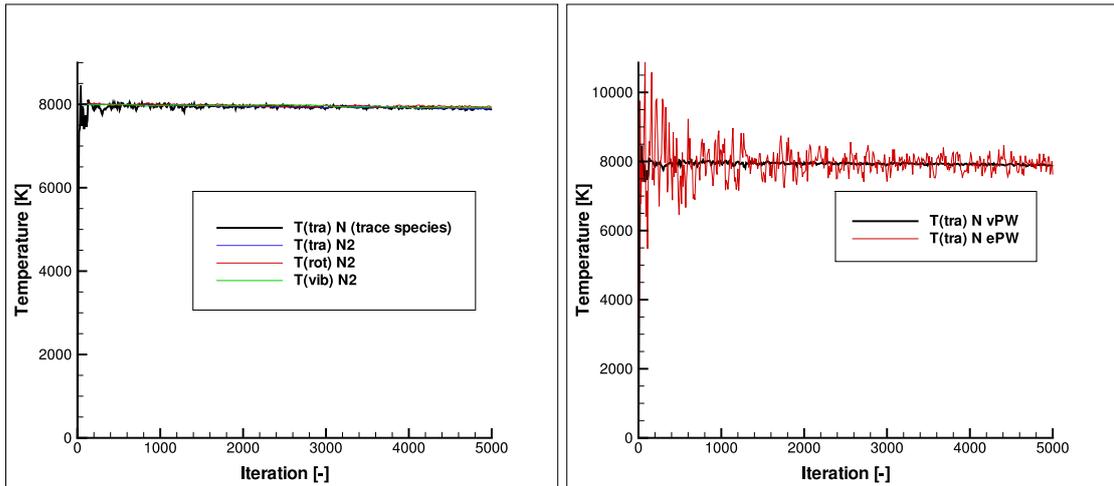


FIGURE 2. (a) Temperature convergence. (b) Translational temperature signal: new model (*vPW*) vs. standard DSMC (*ePW*).

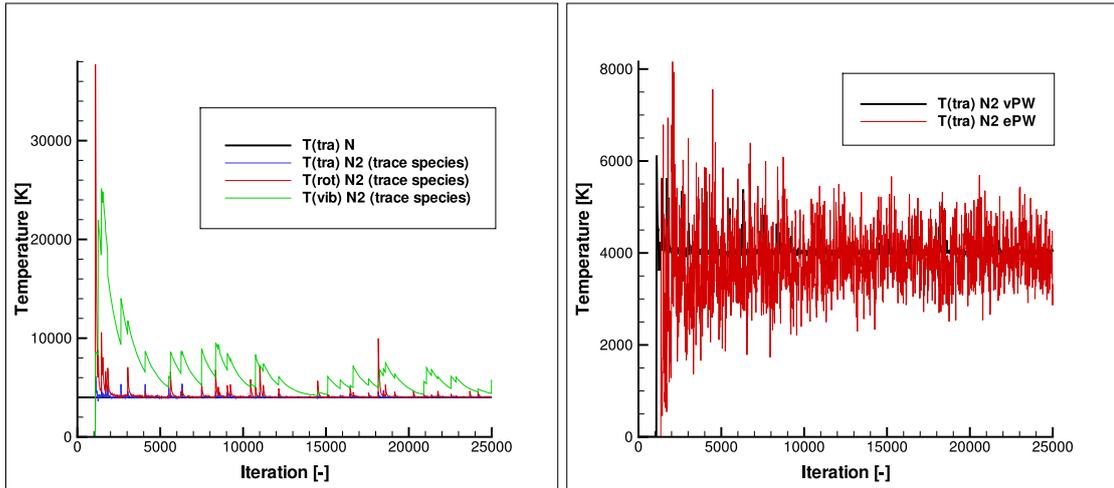


FIGURE 3. (a) Temperature convergence. (b) Translational temperature signal: new model (vPW) vs. standard DSMC (ePW).

Figure 2 at the right compares the translational temperature signals of the new approach (vPW) vs. standard DSMC (ePW). Again, the signal quality has improved significantly.

5.3. Recombination

In the used code the recombination process algorithmically cannot be realized without allowing for dissociation processes. Therefore, both recombination and dissociation (including the relaxation processes) take place according to slightly differing parameters: Now, N_2 is the trace species which, in the vPW model, is resolved according to $W_{N_2} = 10^4$ yielding $2 \cdot 10^5$ trace species particles. The initial temperature of the background species equals $T_N = 4000$ K.

The temperatures converge to a single value (figure 3 (a)) with qualitatively correct convergence rates. All red peaks (rotational DOF) coincide with the green peaks (vibrational DOF). Therefore, this is attributed to recombination processes in which trace species particles were used as third particles. However, there might be an interaction with the vibrational energy conservation modelling in which the final vibrational level is chosen on basis of random numbers.

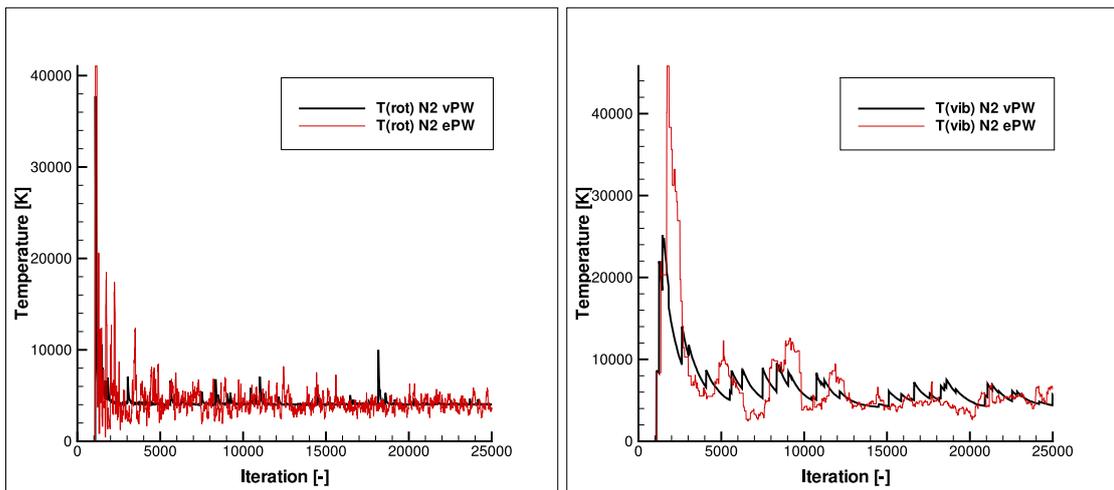


FIGURE 4. (a) Rotational temperature signal: new model (vPW) vs. standard DSMC (ePW). (b) Vibrational temperature signal: new model (vPW) vs. standard DSMC (ePW).

The translational temperature of the trace species element (black curve) is resolved much better than with standard DSMC (red curve), see figure 3 (b). The graph shows that signal peaks are also in the translational DOF. Another significant difference is related to the after splitting in case of vPW which leads to an increase of trace species particles. Contrarily to ePW after the first recombination process, the calculation of a temperature value for N_2 does give a non-zero number such that the black curve rises before the red. In fact, the appearance of the red curve marks the second recombination process due to the non-zero signal.

Concerning the signal quality, for the rotational DOF as shown in figure 4 (a) the observation is similar although the signal quality improvement is less significant compared with the translational DOF. Again, the black line contains some peaks which presumably represent recombination processes in which a trace species particle is used as third particle, yielding a jump in the distribution function.

For the vibrational DOF a signal quality improvement is observed only *between* the peaks of the black curve, see figure 4 (b). Alternatively, the peaks can originate from the fact that in the current implementation the vibrational energy is not conserved exactly but only on average. This might lead to that kind of jump in the vibrational temperature and will be addressed in upcoming reports.

6. DISCUSSION AND OUTLOOK

The developed gas phase reaction models and the corresponding rules were implemented and verified on basis of adiabatic single cell (box) simulations. Not surprisingly, the simulation time per particle was slightly increased. However, as in case of the non-reactive test case, the total simulation time was decreased by approx. 99% if one estimates the particle numbers necessary to get the same signal quality with standard DSMC.

Three catalysis models were developed and discussed in detail, starting with a very simple but not very physical type. However, the third model is the most realistic one but not necessarily computationally most expensive (compared with the second model). Both surface related processes are allowed to occur concurrently - adsorption and recombination, allowing for dynamic sheath growth.

Some model aspects need improvement or at least intense elaboration:

- The conservation of vibrational energy in the gas phase reaction models. The ΔE_v can generally be added to ΔE which stores already energies which cannot be conserved explicitly during each collision (CWS model). In the present implementation the *vibrational* energy gap is either added to or removed from the system such that energy can be conserved only on average over many collisions. For instance, the residual energy caused by the discrete character of the vibrational states can be stored in the general ΔE variable whose value is then coupled into a subsequent collision of equally weighted non-trace particles with $W = W_{max}$.
- The after-splitting algorithm based on weight division operation in combination with the probabilistic approach for particle weight derivation causes a violation of mass conservation. On average and under equilibrium conditions this should not be critical. However, problems might arise under real flow conditions such that this model needs modification (or replacement).
- The third particle set aside for energy conservation in non-radiative recombination processes is arbitrarily chosen from the particle repository. However, particles with very low particle weights should not be used for this purpose otherwise very high and non-physical particle energies in different degrees of freedom might be obtained. Correspondingly, the peaks in the temperature signals cannot be fully interpreted on basis of the current implementations.

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