Utilization of Transport of Species and Heat Release to a DRG-Method-Based Reduction

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1 Introduction

Detailed reaction mechanisms are used for reproducing combustion phenomena precisely. Many reaction mechanisms modeling the oxidation of various hydrocarbons have been developed over the past few decades, and the sizes of the mechanisms have increased drastically as the target hydrocarbons have become larger. However, large reaction mechanisms disenable numerical simulations to be easy to converge because they include too many species and elementary reactions. Actually, it is unrealistic to carry out multidimensional simulations using recently-developed huge detailed mechanisms because of their high computational cost. Therefore, reduction of the size of a detailed reaction mechanism is one of the essential techniques to reduce computational cost for a detailed kinetics simulation.

The enormousness of a detailed mechanism is caused by its comprehensiveness in thermodynamic condition. Therefore, it is possible to make a skeletal mechanism from a detailed mechanism by removing unimportant species and reactions for particular states of interest. The directed relation graph (DRG) method [1, 2] is one of the techniques for mechanism reduction by evaluating the importance of each species in detailed mechanisms. The original DRG method is rather simple since it considers only chemical reaction rates in homogeneous systems. In realistic combustion phenomena, however, transport of species also has important roles, so Tosatto et al. [3] developed "the transport-flux-based directed relation graph method" that considers convection and diffusion velocity in addition to chemical reaction rates. In this study, we developed a new method including three factors; chemical reaction rates, transport of species, and heat release rate (HRR) of each elementary reaction step; for evaluating the importance of species based on the consideration that not only the reaction rate itself but also its heat release is important since the latter directly changes the local temperature that significantly affects reaction rates, which may cause self-acceleration. "The transport and heat release DRG method" proposed here is expected to rank the importance of species more correctly than the original DRG method and reduce the error caused by the mechanism reduction.

In this study, we validate the transport and heat release DRG method through numerical simulations of ethylene/air 1-D premixed flame and n-butane/air 1-D premixed flame. Laminar flame speed and flame structure obtained by a produced skeletal mechanism are compared with those by the original detailed mechanism. Various size of a skeletal mechanism is used for the validation.

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2 Reduction methodology

Firstly, we show the original DRG method proposed by Lu and Law [1, 2], which is the base of our present method. We use the original DRG method adding several modifications in order to apply it to sampling from a 1-D flame. In the method, the important species kept in the skeletal mechanism are determined by DRG which indicates dependence between two species. Fig. 1 describes a simple example of DRG. Each vertex on the graph means a species in a detailed mechanism. Edges between species are vectors defined as

$$r_{AB}{}^{J} = \frac{\sum_{i=1,II} |\nu_{A,i}\omega_i{}^{J}\delta_{Bi}|}{\sum_{i=1,II} |\nu_{A,i}\omega_i{}^{J}|}, \tag{1}$$

where $\nu_{A,i}$ is the net stoichiometric coefficient of species A in reaction i, ω_i is the net reaction rate of reaction i, J is the grid number, II is the total number of reactions in mechanism, and δ_{Bi} is defined as

$$\delta_{Bi} = \begin{cases} 1 & \text{if the } i\text{th reaction involves species B,} \\ 0 & \text{otherwise.} \end{cases} \tag{2}$$

$r_{AB}{}^{J}$ is the normalized contribution which means how much reaction rate of species A depends on the reaction including species A and B. Importance of each species $R_A^{J,T}$ at grid J is then quantified by the minimum value which exists in edges from target species T (i.e., starting species of graph search) to considering species. Each target species is selected from species considered to be important in the mechanisms, such as fuel, oxidizer, and major radical. In the end, resulting importance of each species is determined by $R_A{}^{J,T}$ of all grids and target species, and defined as

$$R_A = \max_{all\ J,T} \alpha_{J,T} R_A^{J,T}, \tag{3}$$

where $\alpha_{J,T}$ is a scaling factor proposed by Pepiot-Desjardins and Pitsch [4] and defined as

$$\alpha_{J,T} = \max_{all\ atoms\ a} \frac{\alpha_{a,J,T}}{\max\limits_{all\ J} \alpha_{a,J,T}}, \tag{4}$$

where $\alpha_{a,J,T}$ is a scaling coefficient and defined as

$$\alpha_{a,J,T} = \frac{N_{a,T}\left|P_{J,T} - C_{J,T}\right|}{P_{J,a}}, \tag{5}$$

where $N_{a,T}$ is the number of atom a in target species T, $P_{J,a}$ is the pseudo-production rate of atoms a at grid J, and $P_{J,T}$ and $C_{J,T}$ are the production rate and consumption rate of target species T at grid J, respectively.

Species are kept in skeletal mechanism if and only if

$$R_A > \varepsilon_{DRG}, \tag{6}$$

where ε_{DRG} is the user-specified threshold value.

The normalized contribution to consider transport of species is evaluated by the equation proposed by L. Tosatto et al. [3], which is

$$T_{AB}{}^{J} = \frac{\sum_{i=1,II} |\nu_{A,i}\omega_i{}^{J}\delta_{Bi}|}{\max\left\{\sum_{i=1,II}\left|\nu_{A,i}\omega_i{}^{J}\right|,\ \frac{1}{W_A\Delta x}\left(\left|\varphi_A{}^{J-1\cap J}\right| + \left|\varphi_A{}^{J\cap J+1}\right|\right)\right\}}, \tag{7}$$

where $\varphi_A{}^{J\cap J+1}$ is the transport flux (convection and diffusion) between grid J and $J+1$.

We propose the normalized contribution including the effect of HRR. This is because Eq. (7) cannot keep the reactions which have small reaction rates and, however, have large HRR value. In

order to increase the normalized contribution of species included in the reactions like that, ${T_{AB}}^J$ is compared with

$$ {Q_{AB}}^J = \frac{\left|\sum_{i=1,II} {\dot{q}_i}^J \delta_{A,B}\right|}{max\left\{{P_{q,A}}^J, {C_{q,A}}^J\right\}}, \tag{8} $$

where ${\dot{q}_i}^J$ is the HRR of reaction i at grid J, ${P_{q,A}}^J$ and ${C_{q,A}}^J$ are the heat production rate and the heat consumption rate of reactions involving species A, and $\delta_{A,B}$ is defined as

$$ \delta_{i,A,B} = \begin{cases} 1 & \text{if the } i\text{th reaction involves species A and } B, \\ 0 & \text{otherwise.} \end{cases} \tag{9} $$

${Q_{AB}}^J$ means how much the HRR of reactions involving A depends on B, and evaluates the error of HRR caused by removing B from the mechanism. Finally, ${T_{AB}}^J$ and ${Q_{AB}}^J$ are combined by

$$ {C_{AB}}^J = max\left\{{T_{AB}}^J, {Q_{AB}}^J {\alpha_Q}^J\right\}, \tag{10} $$

where ${\alpha_Q}^J$ is the scaling factor for heat defined as

$$ {\alpha_Q}^J = \frac{\dot{Q}^J}{\max\limits_{all\ J} \dot{Q}^J}, \tag{11} $$

where $\dot{Q}^J$ is the net HRR at grid J, and $\dot{Q}^J$ becomes unity at the grid in which maximum net HRR is observed.

In this paper, the original DRG method, the transport DRG method, and the transport and heat release DRG method use Eq. (1), (7), (10), respectively. Next section presents the validation of these three methods, and the effect of HRR on the performance of resulting skeletal mechanism is examined.

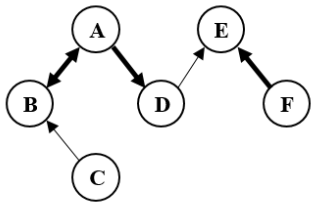

Fig. 1 Schematic drawing of DRG

3 Validation

We tested three types of DRG method mentioned in the preceding section through the simulation of 1-D premixed flame by CHEMKIN. Ethylene and n-butane were selected for fuel, and oxidizer was air. We used the detailed mechanism of ethylene developed by S.G. Davis, C. K. Law and H. Wang [5], which includes 469 reactions and 71 species. For n-butane/air flame, the detailed mechanism developed by Combustion Chemistry Centre in NUI Galway [6], which includes 1328 reactions and 230 species, was used. The sampling data for each DRG method were also produced by 1-D premixed flame with equivalence ratios of 0.7 to 1.5. In the case of n-butane, the grids which have the HRR of larger than 1 kJ/(cc-s) were selected for sampling use, because the grid used for sampling should have active reactivity. The initial temperature was 300 K for ethylene and 500 K for n-butane, and the initial pressure was 1 atm in all following results.

Firstly, we show the validation with ethylene/air premixed flame. Fig. 2 shows the laminar flame speed of ethylene/air flame. Fig. 2 (a) was obtained by the skeletal mechanism of 29 species reduced from 71 species. A large discrepancy exists between the original DRG method and the detailed

mechanism, while the other methods indicate similar results as the detailed one, and maximum relative error is about 5% for transport DRG method and 2% for transport and heat release DRG method. Fig. 2 (b) is the case of the skeletal mechanisms including 19 species. Obviously, the results of the original DRG method and the transport DRG method are completely differed from the detailed one, while the skeletal mechanism produced by transport and heat release DRG method well reproduce the result of detailed mechanism except for the case of high equivalence ratio. Such comparisons as shown in Fig. 2 were conducted with various sizes of skeletal mechanism and arranged in Fig. 3. Mean relative error is obtained by averaging relative errors on laminar flame speeds at equivalence ratios of 0.7 to 1.5. For transport and heat release DRG method, the mean relative error is smaller than the results of the other methods roughly regardless of the size of a skeletal mechanism.

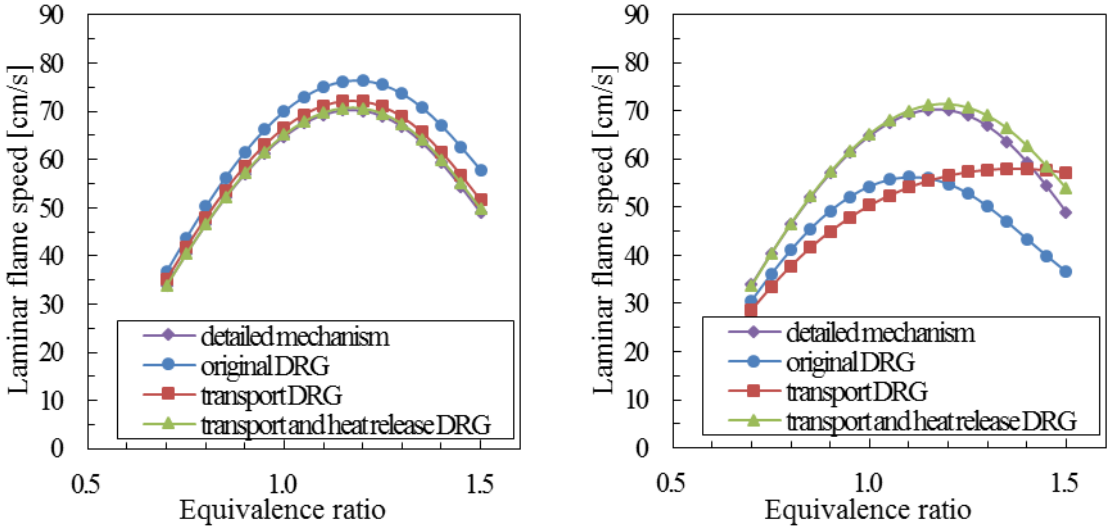

Fig. 2 Comparison of laminar flame speed of ethylene/air flame obtained by detailed mechanism and skeletal mechanisms. (a) and (b) use the skeletal mechanism of 29 species and 19 species, respectively.

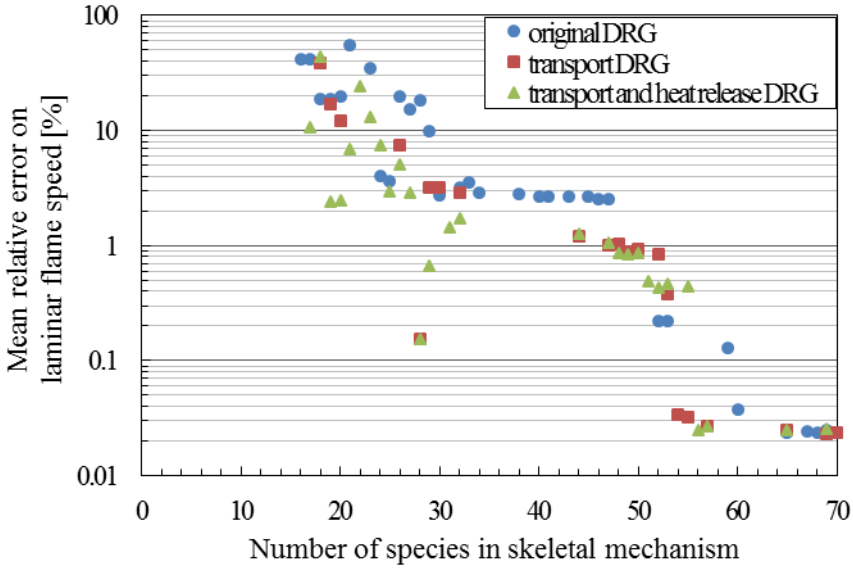

Fig. 3 Mean relative error on laminar flame speed of ethylene/air flame between the result of detailed mechanism and each size of skeletal mechanism.

The validation with n-butane/air premixed flame is shown for the example of reduction of larger detailed mechanism than ethylene. Fig. 4 shows the flame structure of n-butane/air premixed flame. The result of transport and heat release DRG method is considerably close to the result of detailed mechanism except for a little discrepancy found in HRR. The original DRG method and the transport DRG method also have good agreement with the result of the detailed mechanism in mole fraction and temperature, and, however, a large discrepancy is shown in HRR. Fig. 5 is the comparison of mean relative error on laminar flame speed for n-butane/air premixed flame. It cannot be mentioned easily which method is the most appropriate for yielding laminar flame speed because the ranking is changed by the size of the skeletal mechanism. However, in the range of relative error of 0.1 to 1.0%, which is useful for application, the transport and heat release DRG can make smaller skeletal mechanism than the others.

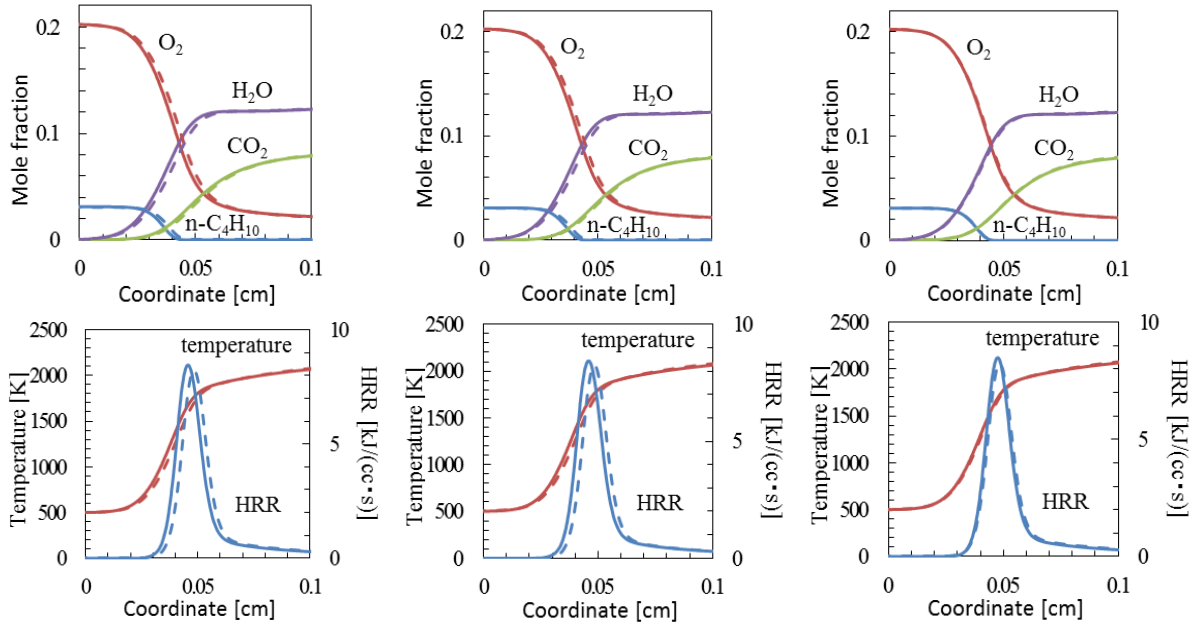

Fig. 4 Flame structure of n-butane/air 1-D premixed flame. (a), (b), (c) are the results of the original DRG, the transport DRG, the transport and heat release DRG, respectively. Solid lines are solved by skeletal mechanisms including 47 species, and dashed lines are yielded by detailed mechanism.

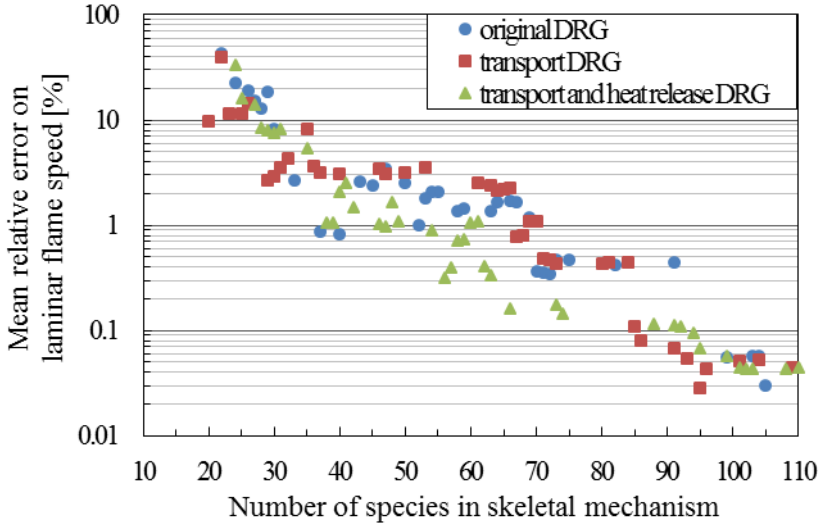

Fig. 5 Mean relative error on laminar flame speed of n-butane/air flame between the result of detailed mechanism and each size of skeletal mechanism.

4 Conclusion

In this study, we developed "the transport and heat release DRG method" for reaction mechanism reduction, which includes the consideration of transport of species and heat release rate in addition to reaction rate, and comparison of three methods, i.e., DRG, transport DRG, and transport and heat release DRG, was conducted using ethylene/air and n-butane/air 1-D premixed flames. As a result, the following knowledge was obtained.

- In the case of ethylene/air premixed flame, the laminar flame speed was well reproduced by the skeletal mechanism produced by transport and heat release DRG method.
- In the case of n-butane/air premixed flame, comparison of the skeletal mechanisms including 47 species shows that mole fraction and temperature were well reproduced by each method. However, the error of heat release rate by the present DRG was smaller than the others.
- In the case of n-butane/air premixed flame, the transport and heat release DRG produced smaller skeletal mechanism than the other methods in the range of the relative error on the laminar flame speed of 0.1 to 1.0%.

References

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