A Global Pathway Selection Method for the Reduction of Detailed Chemical Kinetic Mechanisms

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Introduction

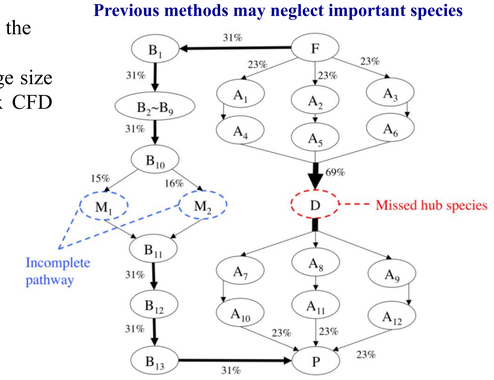
Challenge of combustion simulation

- Numerical simulations play an increasingly important role in the study of combustion
- However, the extremely expensive CPU time due to the large size of the detailed mechanisms prohibits their use in complex CFD simulations

The reduced mechanism

- One strategy to alleviate the computational complexity is the reduction of the detailed mechanism by trimming the less important species;
- The selection of this abundant set needs the knowledge of the complex chemistry relationship;
- However, the coupling relationship between species through one or multiple reaction steps makes this task difficult.
- For the multi-component fuel the challenge is even greater due to the coupling among the components

Previous methods may neglect important species



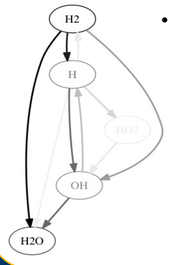
Comparison with previous methods

	Directed Relation Graph (DRG)	Pair element Flux	Path Flux Analysis (PFA)	**Global Pathway Selection (GPS)**
Reference	Lu et al, 2005	He et al, 2008	Sun et al, 2010	**The present work**
Strategy	Expand the set of important species iteratively	Identify the set of important species simultaneously	Expand the set of important species iteratively	Identify a set of hub species, then pick up the ones on their pathway
Flux considered	Net reaction rate	Element flux	Net reaction rates of cons. and prod.	Element flux of cons. and prod.
Generation considered	One generation	One generation	Multi-generation	Up-to-infinity generation

Methodology

Step1. Construct element flux graph

- The weight of the edge represents the flux rate (mole/s) of atoms of the considered element passing through this edge from all possible reactions
- The graph can be constructed either using integral of reaction rate, or instantaneous values. Therefore applicable to one-the-fly reduction



- The graph is a easy representation of the physical facts, for example, hydrogen atoms come out from H_2 is approximately equal to that entering to H_2O

Step2. Identify hub species

- The nodes have significant total outgoing/incoming flux act as hubs of the flux network, thus are kept in the reduced mechanism
- Essentially, this step identifies the species that have significant relationship with initial reactant, through up-to-infinity generation, as all atoms must be converted from initial reactants, not affecting by the number of reaction steps it takes
- This provides a very easy way to consider the infinite-generation relation and avoid the "missed hub species" issue, which was time-consuming in previous method such as PFA

Step3. Select global pathways

- For each identified hub species, the pathways important to its production and consumption will be searched and kept to resolve the incomplete pathway issue
- For example, the global pathway of C2H5CO in the combustion of n-heptane identified from carbon flux graph is

$nC_7H_{16} \rightarrow C_7H_{15} \rightarrow C_7H_{15}O_2 \rightarrow C_7H_{14}OOH \rightarrow C_7H_{14}OOHO_2 \rightarrow nC_7(Ket) \rightarrow C_2H_5CHO \rightarrow C_2H_5CO \rightarrow CO \rightarrow CO_2$

- This step is achieved by algorithm from graph theory (Yen's k shortest pathways algorithm)

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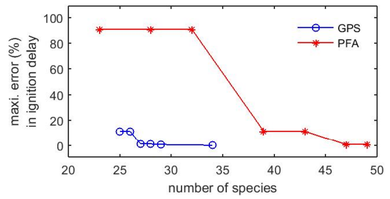
Results & Discussion

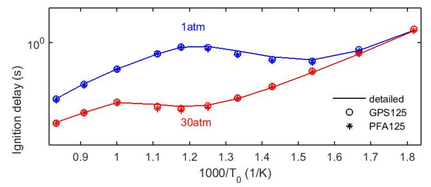
- The present GPS method is demonstrated by comparing the accuracy of the reduced mechanism with previous methods.
- The accuracy is measured by comparing the simulation with detailed mechanism on autoignition delay, perfect-stirred reactor, laminar flame speed, or flame structure. Various fuels and broad range of operating conditions are considered

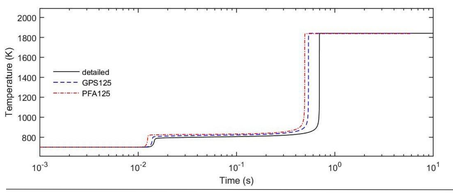
Methane



- GPS can reduce the number of species from 53 to around 20 but keep high accuracy, either using integral or instantaneous form
- Compared to PFA, GPS shows high accuracy over wide range of reduced mechanism size. The reason is that C_2H_6, an important species, is neglected by PFA

n-Heptane

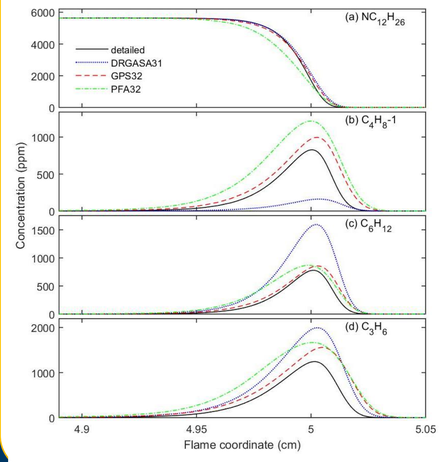




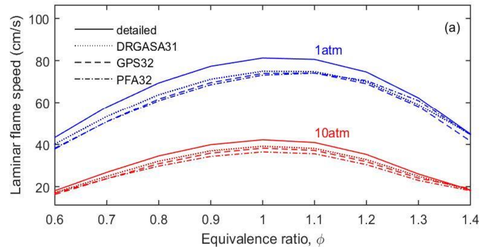
- GPS can reduce the number of species from 1034 to 125 species, but predicts the negative temperature coefficient (NTC) behavior and two-stage ignition process accurately
- Compared to PFA, the skeletal mechanism generated by GPS could be more accurate at the low temperature region

n-Dodecane

- GPS is further compared with DRG aided sensitivity analysis, which is very time consuming but is believed to very effective, as the error is measured directly

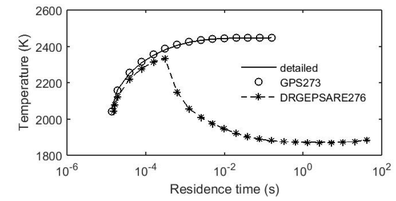


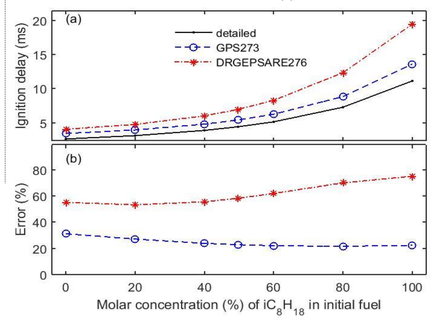
- GPS can achieve similar accuracy of similar mechanism size without this time-consuming process.
- Furthermore, GPS can be more accurate for predict of flame structure, as this is usually not validated by sensitivity analysis, but important in complex reacting flows.



Multi-component

- The practical fuel is usually mixture of several components. New pathways not favored for neat fuel may appear
- Previous reduction methods may over emphasize one component whose reaction pathways are more straightforward, or whose fraction is higher.
- GPS, however, can minimize this risk. Tests are conducted on the mixture of toluene, n-heptane and iso-octane
- Compared to previous DRG method with error propagation, sensitivity analysis, and CSP reaction elimination (DRGEPSARE), GPS shows much higher accuracy
- DRGEPSARE may neglect the consumption pathway of important species and results in abnormal PSR behavior, but GPS won't have such issue





Conclusions

1. GPS considers the up-to-infinite generation coupling relation between species to identify the important species, thus can resolve the issues encountered by previous methods;
2. Compared to previous methods, GPS could show higher accuracy for the reduction of mechanism, and save the reduction time for various fuel (gas or liquid, neat or multi-component) and operating conditions;
3. GPS can be applied to either global or local (on-the-fly) mechanism reduction.