

## Reformer Model Development for Hydrogen Production

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### Objectives

- Research and develop low cost, highly efficient hydrogen production from renewable sources
- Reduce the cost of distributed production of hydrogen from liquid fuels such as diesel and biomass oils

### Technical Barriers

This project addresses the following technical barriers from the Hydrogen Production section of the Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year R,D&D Plan:

- A. Fuel Processor Capital Costs
- B. Operation and Maintenance
- C. Feedstock and Water Issues
- D. Carbon Dioxide Emissions
- E. Control and Safety
- F. Feedstock Cost and Availability
- G. Efficiency of Gasification, Pyrolysis and Reforming Technology

### Approach

- Develop a two-part database that is the benchmark for developing and testing the elements of the reformer model, called submodels
- Derive submodels from one part of the database to be used for the reformer simulation
- Use the complement of the database to test the derived submodels
- Validate the entire model using experimental data
- Utilize the validated model to optimize and scale up reformers

### Accomplishments

- Created a two-part database portraying the evaporation and mixing of a large number of single-chemical-species-liquid drops in a turbulent flow
- Used one part of the database to develop computationally efficient models (called submodels) of the small scale interaction of the drops and flow
- Tested the submodels on the complement of the database to assess the interaction of the submodels with the large scales; found a model that is stable and accurate and reduces computational costs by a factor of 300

- Tested a model of multicomponent-liquid (i.e. mixture of many chemical species, as in diesel and biomass oils) drop existing in the literature and found that it has serious limitations
- Developed a computationally efficient improved model of the evaporation of a multicomponent-liquid drop when the fuel is a mixture of hundreds of chemical species

### **Future Directions**

- Validate the single-chemical-species-liquid model with experimental data for an evaporating spray
- Create a database of the evaporation and turbulent mixing of multicomponent-liquid drops to enable the extraction of submodels for complex-mixture liquids
- Extract submodels for complex-mixture liquids evaporation and turbulent mixing
- Test the interaction of the submodels for complex-mixture liquids evaporation and turbulent mixing with the large flow scales
- Validate the multicomponent-liquid model with experimental data for an evaporating spray
- Create a two-part database of the reforming reactions in turbulent flows in the presence of evaporating multicomponent-liquid drops to enable the extraction of submodels for the chemistry/turbulent-flow interaction
- Use one part of the database to extract submodels of the interaction between chemistry and turbulence
- Test these submodels on the complement of the database to assess the interaction between the submodels and the database
- Validate the reformer model (i.e. turbulent, reactive flow laden with evaporating drops) by comparing with experimental data

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### **Introduction**

Reforming is a chemical process wherein a hydrocarbon feedstock reacts with water to form H<sub>2</sub> and other gaseous chemical species. These reactions are induced in a container, called the reactor. In this reactor, the feedstock is introduced in the form of a spray of drops. Atomization of a liquid (i.e., the formation of drops) enlarges the surface area of the liquid, thereby promoting evaporation. The evaporated species reacts with steam in the presence of catalysts which enhance the reaction. The reactor operates at a temperature of 700°C to 850°C and pressures of 3 to 25 atmospheres, which favor the output of the reaction [1]. Concerns encountered with reforming for H<sub>2</sub> production are the efficiency of the process (e.g. optimization of the parameters for maximum yield), which must be high enough to render H<sub>2</sub> economically viable; the presence of coking reactions, which build carbon on the catalysts, thereby deactivating them; and reactor scale-up to industrial sizes.

The present study addresses all concerns of the process with the goal of understanding the details of the physical/chemical processes involved, so as to enable changes of the present operation that are based on physics/chemistry rather than circumstantial empirical correlations. This understanding will also be vital in proposing reactor scale-up.

### **Approach**

The approach is based on numerical simulations that are experimentally validated. The idea is that just as the aircraft industry uses simulations to design aircraft (and no longer performs costly wind-tunnel testing), the hydrogen industry can eventually design reformer reactors using simulations as well. However, in many ways the situation in a reformer is considerably more challenging to model than for the aircraft; in the reformer one has a turbulent two-phase (liquid drops and gas) reacting flow with phase change (drop evaporation). Moreover, the feedstock is invariably a complex mixture of hydrocarbons that cannot be effectively treated as a single chemical

species because different components of the mixture are responsible for different aspects of the process.

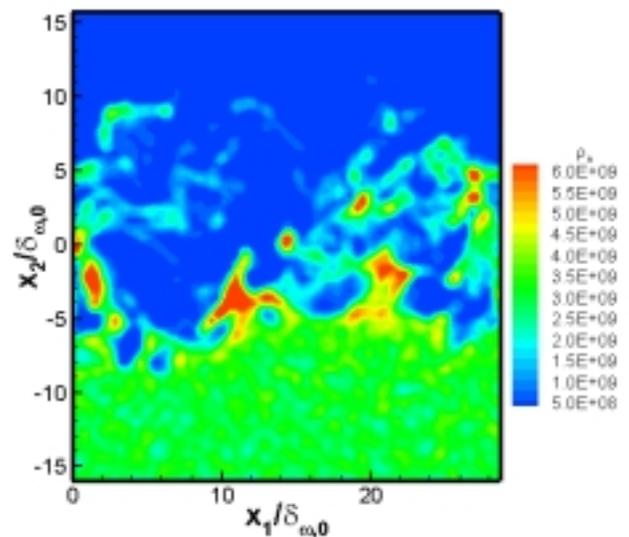
The methodology uses the contemporary direct numerical technique called DNS to achieve a transitional state, defined as a state displaying turbulent features. The DNS is performed in a domain much smaller than that of interest because currently and in the foreseeable future there is no large enough computer to simulate turbulent flows of practical interest; even with the small domain size, these DNS calculations are very computationally intensive, lasting thousands of computer hours. However, neither the small DNS domain size nor the costly computations are of concern because this simulation is supposed to be only a tool for model development. The models extracted from DNS are Subgrid Scale (SGS) models, and they represent the small-scale behavior of the flow. This small-scale behavior cannot be computationally resolved for flows of practical interest; therefore, SGS modeling is used. The simulations wherein the large scale of engineering interest are computed, and the small scales, all of which have a major impact on large scales, are modeled, is called Large Eddy Simulation (LES). These LESs are intended to be computationally much cheaper than the DNS, so as to be able to use them on a routine basis for engineering design. The goal is here to create DNS databases, develop SGS models, and ultimately perform LES. The predictive ability of LES crucially depends on the quality of the SGS models.

## Results

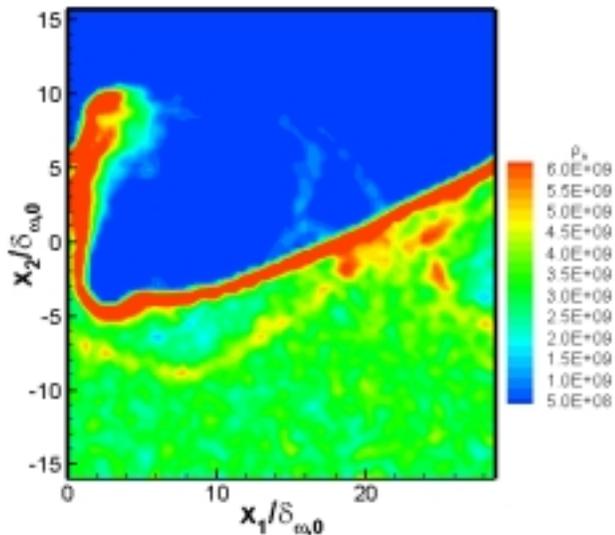
We have created DNS databases of transitional states both for gaseous flows and for flows containing millions of drops. For drop-laden flows, we have used in the computation either single-component (SC) or multicomponent (MC) drops. To model MC drop evaporation, we used a model already existing in the literature, which we showed contained limitations when exercised for drops evaporating in mixtures of air and hydrocarbon vapor. The limitations motivated us to develop a more robust model of MC drop evaporation, which was validated in its predictive capability [2].

SGS models were developed using one part of the DNS database and then tested on the complement

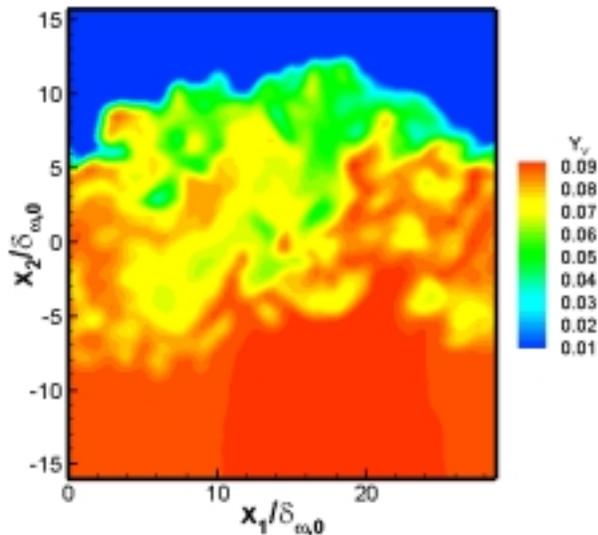
of the database. The results showed that currently used SGS models are inherently incorrect, in that they are unable to reproduce the DNS database [3]. Particular deficiencies are mostly seen in their inability to predict the correct drop distribution in the flow and the correct mixture fraction, which is essential for predicting reforming reactions. From the analysis of the database, we have developed accurate SGS models that are as computationally inexpensive as those incorrect ones that are currently used. This comparison is presented in Figures 1-6, which also include in the captions the computational times necessary to achieve these states starting from two segregated gaseous streams moving with respect to each other, where the bottom stream is laden with drops that may evaporate (the initial gas temperature is larger than the initial drop temperature). The initial mass loading with liquid drops is 50%. The results show that Figure 2 is only a caricature of Figure 1, rather than a replicate, and the same can be said of Figure 5 with respect to Figure 4. In contrast, Figure 3 replicates reasonably well Figure 1, and Figure 6 compares very favorably with Figure 4, and this at only a fraction of the computational time of the original simulation.



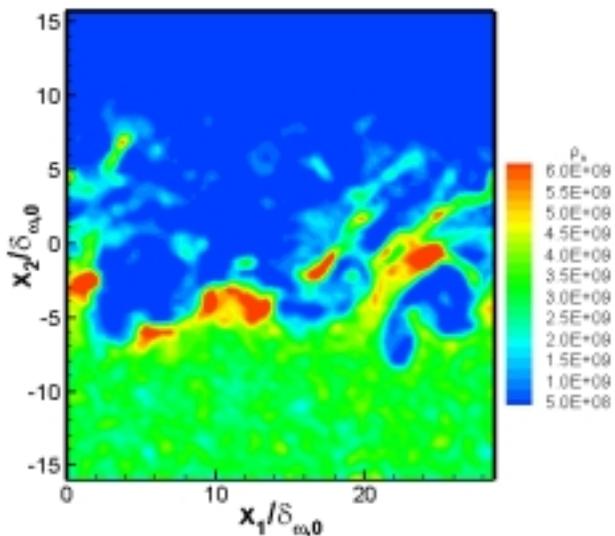
**Figure 1.** Drop number density (in  $\text{m}^{-3}$ ) obtained from a DNS. The figure illustrates the results from a computation that lasted 2981 hours of computer time. In the figure, the small scales are removed, and only one out of 8 drops is shown, in order to have an appropriate comparison with LES.



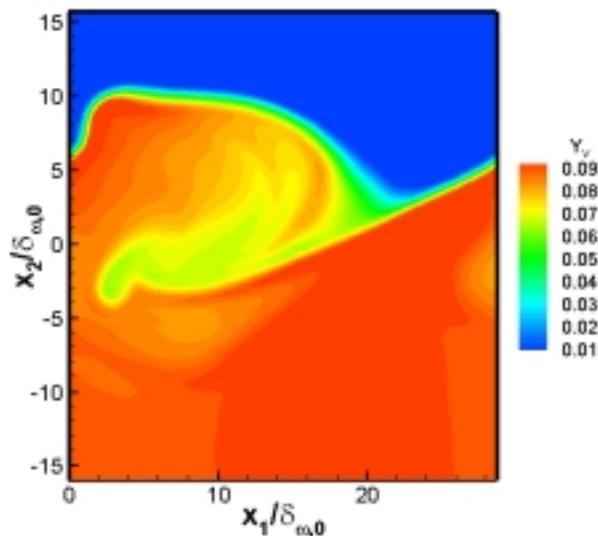
**Figure 2.** Drop number density (in  $m^{-3}$ ) obtained from a LES using currently used SGS models. The simulation lasted 37 hours of computer time.



**Figure 4.** Vapor mass fraction obtained from the same DNS as in Figure 1. Just as in Figure 1, the small scales are removed, and only one out of 8 drops is shown, in order to have an appropriate comparison with LES.



**Figure 3.** Drop number density (in  $m^{-3}$ ) obtained from a LES using the best SGS model developed in this program. The simulation lasted 37 hours of computer time (same number of hours as in Figure 2).

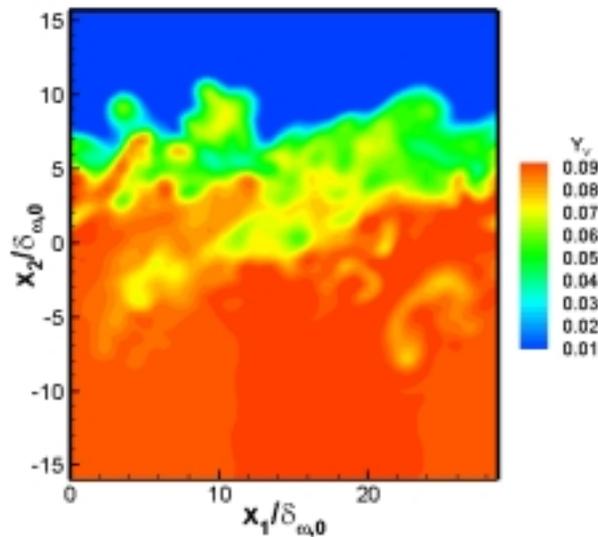


**Figure 5.** Vapor mass fraction obtained from a LES. The figure illustrates the results from the same computation as in Figure 2.

**Conclusions**

It is now apparent that current models are highly deficient in their portrayal of turbulent mixing and drop evaporation in a turbulent flow. The same can

be stated about the modeling of evaporating drops of complex hydrocarbon mixtures. Models developed in this project have the ability to remedy these deficiencies and will thus be used in further computationally efficient simulations.



**Figure 6.** Vapor mass fraction obtained from a LES. The figure illustrates the results from the same computation as in Figure 3.

## References

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- “A robust statistical model for the evaporation of multicomponent-fuel drops containing a multitude of chemical species”, (K. G. Harstad, P. LeClerc and J. Bellan), paper AIAA 2003-1321, presented at the 41<sup>st</sup> Aerospace Sciences Meeting, Reno, NV, 2003
- “The statistical modeling of multicomponent-fuel drop evaporation for fuels containing a multitude of species”, (K. G. Harstad, P. LeClerc and J. Bellan), presented at the Joint Meeting of the US Sections of the Combustion Institute, March 17-19, 2003
- “*A priori* study of models for large eddy simulations of drop-laden flows”, (N. Okong'o and J. Bellan), presented at the Joint Meeting of the US Sections of the Combustion Institute, March 17-19, 2003; also ILASS 2003, Monterey, CA, May 18-21, 2003
- “*A posteriori* study of models for large eddy simulations of drop-laden flows”, (A. Leboissetier, N. Okong'o and J. Bellan), ILASS 2003, Monterey, CA, May 18-21, 2003
- “Direct Numerical Simulations of Two-phase Laminar Jet Flows with Different Cross-Section Injection Geometries”, (H. Abdel-Hameed and J. Bellan), *Phys. Fluids*, 14(10), 3655-3674, 2002

## FY 2002 Publications/Presentations

- “Direct Numerical Simulations of Two-phase Laminar Jet Flows with Different Cross-Section Injection Geometries”, (H. Abdel-Hameed and J. Bellan), *Phys. Fluids*, 14(10), 3655-3674, 2002

**Special Recognitions & Awards/Patents**  
**Issued**

1. NASA Certificate of Recognition for each one of the papers above
2. Invited publication (appeared July 2002) by the Chemical Propulsion Information Agency Bulletin, sponsored by the DoD Information Analysis Center