SINTEF Energy Research

Address:	NO-7465 Trondheim, NORWAY
Reception:	Sem Sælands vei 11
Telephone:	+47 73 59 72 00
Telefax:	+47 73 59 72 50

www.energy.sintef.no

Enterprise No.:
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Characterisation of the mixing behavior of hydrogen rich jets				
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ELECTRONIC FILE CODE		AUTHOR
071114101144		Sigurd Sannan, Mario Ditaranto, Alan R. Kerstein[1]
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DIVISION	LOCATION	LOCAL FAX
Energy Processes	Kolbjørn Hejes vei 1A	+47 73592889

An experimental study case to characterize the mixing behaviour of hydrogen rich jets is described. Both non-reactive mixing of hydrogen and hydrogen flame properties will be studied in the experiment. The proposed experiment is a three-stream mixing set-up that involves two planar jets with co-flows to the sides. Because of the large molecular diffusivity of hydrogen the effects of differential diffusion will particularly be studied. The results of the experimental study will in particular serve as a validation case for the novel LEM-EDC model under development at SINTEF/NTNU. In general, the results should aid in the development of all combustion models addressing differential diffusion and multi-scalar mixing. Experimental data on the mixing properties of hydrogen under various conditions is also expected to be valuable to developers of combustor designs adapted to the thermo-physical properties of hydrogen.

[1] Combustion Research Facility, Sandia National Laboratories, Livermore, CA 94551-0969, USA

1 INTRODUCTION

Combustion of hydrogen or hydrogen-enriched fuels is a central ingredient of the pre-combustion concept for capturing CO_2 in power generation from natural gas or coal. The idea is that the carbon of the fuel is removed prior to combustion, while the fuel heating value is transferred to hydrogen. The use of hydrogen in combustion processes is a challenging task, however, due to the specific thermo-physical properties of hydrogen. Although conventional gas turbines to a certain extent can handle hydrogen-enriched fuels, they are not designed and built for the use of such gas mixtures. The unique properties of hydrogen call for special care in the development of hydrogen combustion technology. Thus, hydrogen in its gaseous phase diffuses much faster than almost any other gas, and it is characterized by a high flame speed and an extremely wide flammability range. This means that issues such as flame holding, flashback, uncontrolled self-ignition, and safety must be resolved in the design of new burner configurations and combustion chambers for hydrogen. Furthermore, the high flame temperature of hydrogen may contribute to higher emissions of NO_x, as well as to enhanced thermal stresses on combustor materials. One way to resolve the temperature related problems is to lower the flame temperature by burning premixed lean mixtures of hydrogen and air. Up until now, the development of lean premixed combustion of hydrogen, with high efficiency and dry NO_x control, has been considered a prerequisite to the development of commercialized hydrogen gas turbines. However, problems related to auto-ignition and flashback have continued to remain a big challenge for gas turbine manufacturers.

A core element in the development of new combustor technology is the application of validated simulation tools that are able to give precise descriptions of the combustion processes. Commercial Computational Fluid Dynamic (CFD) tools of today are based on models that endeavor to capture the essence of the turbulent flow, the chemistry between reacting species, and the interactions between turbulence and chemical reactions. In general, however, the employed turbulence and mixing models rely on the large-scale characteristics of the turbulent flow and do not resolve the smallest scales of the flow at which the molecular mixing and chemical reactions take place. For this reason these simulation tools, based on state-of-the-art combustion models, have limited predictive capabilities in many practical engineering applications. The most accurate way of studying turbulent flows numerically is by a Direct Numerical Simulation (DNS), in which the flow field is resolved directly from the Navier-Stokes equations. In the DNS approach there is no need for turbulence modeling, but the computational cost associated with DNS is generally extremely high. Thus, the extreme range of length and time scales associated with high-Reynolds-number flows makes use of DNS prohibitively expensive, at least for the foreseeable future. It is therefore worthwhile to pursue methods that have the potential of resolving the turbulence-chemistry reactions of reactive flows at all scales, but at a more affordable computational cost than what is required for a DNS. The novel LEM-EDC model, currently under development at SINTEF/NTNU, is a model that does resolve the smallest scales of turbulent flows at affordable computational costs. The LEM-EDC is a hybrid model construction based on the Linear Eddy Model (LEM) formulated by Kerstein [1], [2] and the Eddy Dissipation Concept (EDC) developed by Magnussen *et al.* [4], [4]. In its original formulation, the LEM resolves the smallest scales of turbulent flows along one-dimensional domains. However, in the recently developed 3D-LEM code, which is a sub-structure in the LEM-EDC model, three orthogonally intersecting arrays of LEM domains are incorporated. Hence, the 3D-LEM provides DNS-like resolution in all three spatial directions. In this framework simulations can be performed for a fraction of the cost of a full DNS because the treatment is one-dimensional. The EDC combustion model is a central part of the in-house CFD code SPIDER at SINTEF/NTNU [5], [6]. The SPIDER code is based on the Reynolds Average Navier-Stokes (RANS) equations and can, based on an initial run of a reactive flow, provide input data profiles to the 3D-LEM. Various ways have been foreseen in which SPIDER and the 3D-LEM can be run iteratively and feed information back and forth between the two towards a converged solution. The ultimate goal, however, is a complete integration of the 3D-LEM sub-structure into the SPIDER code to extend the applicability of the tool.

The purpose of the proposed experiment described here and to be conducted at SINTEF/NTNU is to provide experimental data on hydrogen mixing and combustion for the validation of the novel LEM-EDC model. Thus, both non-reactive mixing of hydrogen, as well as hydrogen flame properties, will be studied in the experiment. In general, the data is expected to be useful also for other modeling developments that are endeavoring to provide accurate descriptions of turbulence-chemistry interactions in turbulent reactive flows. Experimental data on the mixing properties of hydrogen, and how these affect the combustion of this fuel, is additionally of great interest to developers of new combustor designs adapted to the particular thermo-physical properties of hydrogen. The effects of the large molecular diffusivity of hydrogen, known as differential diffusion, will particularly be studied. Many state-of-the-art combustion models make a simplifying equal-diffusivity assumption for all chemical species and hence neglect the effects of differential diffusion. However, when hydrogen is part of the reactive flow, such an assumption is prone to be more erroneous than usual. In the 3D-LEM substructure of the LEM-EDC model there is no need for an equal-diffusivity assumption since the model naturally accommodates any diffusivity given to the various species. Therefore, the LEM-EDC model is likely to capture the effects of differential diffusion. Another feature of the proposed experiment is that it is a three-stream mixing set-up that will provide data for comparison with and development of combustion models addressing multi-scalar mixing. Even though turbulent combustion problems of practical engineering interest usually do involve mixing of multiple streams, state-of-the-art combustion models cannot adequately describe turbulent mixing involving multiple streams. In the hybrid LEM-EDC model there is in principle no restriction on the number of chemically distinct streams, a property that suggests that the three-stream mixing should provide a particularly interesting validation case for this model. The preliminary assessment of the proposed experiment is that the results should aid in the development of all combustion models addressing differential diffusion and multi-scalar mixing. Furthermore, the experiment should provide fundamental insight into the mixing properties of hydrogen under various conditions where the effects of differential diffusion will be of particular interest.

2 DIFFERENTIAL DIFFUSION AND MULTI-SCALAR MIXING

As pointed out in the Introduction, hydrogen in its gaseous state has a number of properties that makes it behave very differently from natural gas or some other conventional fuel gas. One of these properties is the very large molecular diffusivity of hydrogen, which for instance causes hydrogen to diffuse about four times faster than air in hydrogen-air flames [7]. This is known as differential diffusion and is generally characterized by the different evolution of chemical species due to their different molecular diffusivities. Differential diffusion is clearly important when it comes to determining the mixing and chemical reaction rates of hydrogen with its oxidizer. The chemical reaction rate is determined by the instantaneous concentrations of the reactants at a given location of the flow, rather than just the means of the reactant concentrations. Therefore, in order to determine precise values for chemical reaction rates it is crucial to track all the reactants in the flow at all relevant length scales. In particular, it is important to resolve the flow at the smallest scales of the flow at which the molecular mixing and chemical reactions take place [8].

In most models of turbulent diffusion flames the effects of differential diffusion are neglected. It is often assumed, provided that the Reynolds number (Re) is sufficiently large, that the molecular diffusivities of all species are equal, in addition to that the effects of molecular diffusion are negligible compared to turbulent diffusion. In turbulent flames, however, the Reynolds number is often significantly reduced due to increased kinematic viscosities at the high temperatures resulting from substantial heat release. Thus, significant effects of differential diffusion in hydrogen jet flames into air have been reported in the literature [9], [10]. Furthermore, it has been shown for a non-reacting jet consisting of Freon and H_2 that differential diffusion can have significant effects on species concentrations even for Reynolds numbers as high as Re=20000 [11], [12]. Clearly, methods based on the assumption of equal diffusivity will not be capable of predicting species mass fractions accurately in the presence of significant differential diffusion. In this respect it should be noted that in the LEM-EDC model there is no need for the equal-diffusivity assumption since the model naturally accommodates any molecular diffusivity given to the different species.

In addition to addressing the effects of differential diffusion, the proposed experiment will provide mixing data for the development of mixing models based on multiple streams. In most combustion studies to date addressing the mixing behavior of chemical species, experimentalists have focused on two-stream mixing, where the mixing of fuel and oxidizer has been studied [13], [14]. In two-stream mixing, the limit of fast chemistry and simplifications such as the equal-diffusivity assumption lead to the definition of a single conserved scalar, the mixture fraction which describes the chemical state of the system. In this approach the mixture fraction represents the fraction of fuel mass at any given point. More generally, the mixture fraction can be defined on the basis of the species mass fractions and is used as a description of the composition of the multi-species mixture. Thus, with the given simplifying assumptions, all species mass fractions and their root-mean-square values then are functions of the mixture fraction [15]. When three chemically distinct streams are mixed, such as in the proposed experiment, it takes two independent mixture fractions to specify the mixture at a point [16]. The merit in performing a three-steam mixing process lies in the fact that it can contribute to the understanding of mixing of multiple streams of scalars. This is especially instructive in turbulent reactive flow problems, since the mixing of chemical species and their reactions usually in fact do involve multiple species. The hybrid LEM-EDC model can handle any number of chemically distinct streams, which in itself makes the model a promising approach to multi-scalar mixing. The power of the model, however, lies within its ability to capture and resolve the physical mechanisms of turbulent stirring, molecular diffusion and chemical reaction down to the Batchelor scale, the smallest scalar length scales of the flow. The 3D-LEM thus provides resolution at all relevant scales, including the smallest scales at which the chemical reactions take place, but a much smaller computational cost than a corresponding direct numerical simulation (DNS).

3 EXPERIMENTAL SET-UP

We here describe the experimental study to be carried out in detail. The proposed experiment involves two planar jets with co-flows to the sides, with the entire flow field confined. We first give a description of the flow configuration, followed by a subsequent discussion of the possible measurement techniques and their relevance to the experiment.

3.1 Flow configuration

The proposed validation case for the hybrid LEM-EDC model is a three-stream mixing set-up involving two planar jets. The experimental set-up is sketched schematically in Figure 1, which shows a 2-dimensional flow configuration for the exit section with two central slots for the planar jets. To the sides of the planar jets there are co-flows of air that provide three-stream mixing between the two central jets and the side flows. The entire flow of gases is assumed to be confined within a rectangular duct. The confinement is chosen to prevent highly flammable hydrogen gas from being emitted into the laboratory facilities, a situation that may be potentially hazardous. With the confinement the co-flows provide infinite reservoirs of fresh air that replenish the air that is mixed into the jet flows. In the case of a turbulent flame the co-flows will also aid in flame stabilization. The confinement of the flow means that special design care must be taken so that confinement effects will not impact the measurements. Thus, measurements must be taken at locations downstream where the flow does not feel the direct impact of the growth of the boundary layers from the confining walls normal to the slots and toward the center. These boundary layers break the planar symmetry of the system. Hence, the design will be such that the nozzle exits are sufficiently wide to allow substantial jet development before these boundary layers break the planar symmetry near the center of the flow. In the design of the confining duct it will also be taken into account that the boundary layers along the walls parallel to the slots should not be close to the jet at measurement locations.

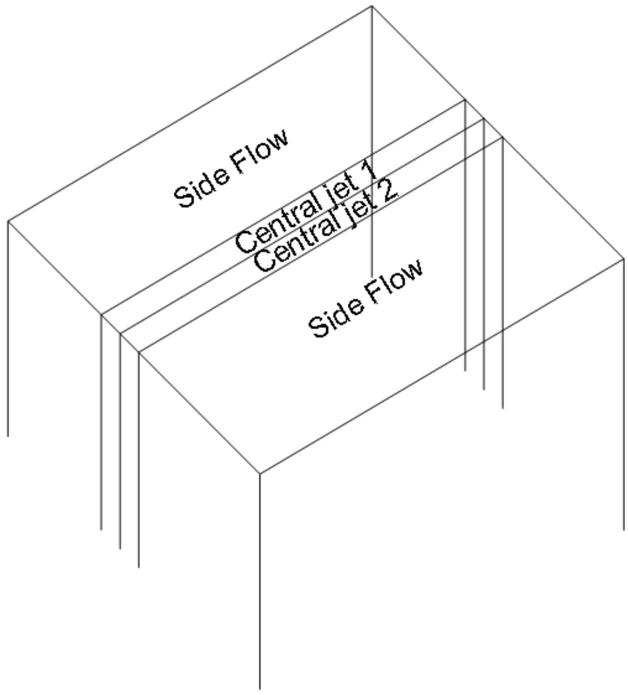

Figure 1 A geometric illustration of the flow field exit section with two central planar jets and co-flows to the side of the jets.

The proposed 2-dimensional flow configuration has several advantages compared to a turbulent round 3D-jet. Foremost, with planar jets it is easier to obtain a high degree of physical accuracy in the measurements since the centering of measurement points is less of an issue. Also, the proposed set-up avoids mixing layer curvature effects in the direction normal to the flow, as is the case in a turbulent 3D-jet. In addition, for a round 3D-jet there are limitations in the turbulence length scales for small jet diameters and it is necessary to go much further downstream to make measurements at locations where the flow field is fully developed. On the validation side, the planar symmetry of the flow means that both stand-alone 1D-LEM and the hybrid LEM-EDC may be applied to the flow configuration.

The described experimental set-up calls for a number of parameters to be studied during the investigation:

- The gas composition of the central planar jets
- The velocities of the planar jets
- The velocity profiles of the planar jets

The co-flows in the set-up will be taken to be air since this is the cheapest to use. For the planar jets the compositions may vary, but one of the jets will in some cases consist of pure H_2 while the second central jet may consist of N_2, O_2, H_2, CO_2, or some mixture thereof. In these cases the mixing of air species into the jet flows will be detected. The flow field will be left-right symmetric if the two jets both have the same velocity, density and viscosity. This is easiest to accomplish if the compositions of the jets are the same, but in principle this does not have to be the case. In some preliminary studies, however, both jets will consist of either H_2 or N_2, in which case the flow field is left-right symmetric if the velocities and the temperatures of the jets also are the same. The flow field will be left-right asymmetric if the velocities of the jets differ, or if the compositions are such that the jets have different densities and viscosities. In this case the boundary layers between the jets and the co-flows on either side will have different behaviour and properties and provide more non-trivial model testing than the symmetric case. In general, the velocities of the planar jets will be varied to study the entrainment of the co-flowing air into the jets, as well as the mixing characteristics. The variation of the velocity profiles of the planar jets makes for even more non-trivial validation cases for which the stand-alone 1D-LEM cannot be applied. However, moving away from the case where the inlet flow velocities are uniform along the slots represents a complication that enables a further testing of 3D-LEM and the LEM-EDC model. Flow conditioners will be used to introduce inlet velocity variations in the slot directions.

One consideration in the proposed set-up is fluid property differences in different streams. Since H_2 is lighter than other fluids and has higher diffusivity, pure hydrogen might introduce significant buoyancy and other dynamical effects in the flow field. The LEM-EDC model should be able to handle these effects because it is applied to combustion. These effects, together with differential diffusion of H_2 and the properties of three-stream mixing will contribute to a very interesting study case. However, it might also be worthwhile to consider some simpler cases that omit the complications of buoyancy effects. Therefore, side flows of air with the jets consisting of pure fluids or different mixtures of N_2, O_2, and CO_2 could serve as good candidates for this. In these cases, small but detectable amounts of H_2 can be added without changing the density significantly, which means that it should be possible to seeing the effects of differential diffusion without large density variations.

3.2 Measurement techniques

The possible measurement techniques to be used in the experiment are as follows:

- Laser Doppler Velocimetry for the 2D velocity components and the turbulence field
- Laser Rayleigh or Raman scattering for the main species local concentration profiles
- Planar Laser Induced Fluorescence (LIF) for instantaneous flow visualisations

2D Laser Doppler Velocimetry (LDV) is a non-intrusive method able to measure two components of the local and instantaneous flow field velocity. The typical data sampling frequency is high enough to obtain the point-wise characteristics of turbulence such as moment orders, the turbulent spectrum and Reynolds stresses. The principle is based on the Mie scattering properties of particles when illuminated by a laser beam. Therefore the gas streams must be seeded with micrometric size particles. The complete velocity and turbulence field is measured by moving the measurement control volume defined by four focusing and intersecting laser beams throughout the domain.

Rayleigh and Raman scattering are techniques based on the interaction of light and molecules. When a volume of gas is illuminated by a laser beam, it scatters light at intensity dependent on its molecular composition and temperature. The process is elastic for the Rayleigh part of the scattering and wavelength-shifted with regard to the excitation light for its Raman part. Both techniques can be used to measure the gas composition and the temperature in non-reacting flows. Although the techniques are possible in burning flows as well, the complexity is severely increased with combustion. The analysis of the Rayleigh and Raman signals have both pros and cons, but the main difference is in their scattering cross-sections which lead to a typical Rayleigh signal intensity three orders of magnitude stronger than that for Raman scattering. On the other hand, the signal of the Raman scattering is distributed over various wavelengths depending on each molecule composing the gas, and it is therefore directly proportional to the gas molecular concentration.

Planar imaging techniques can be achieved by using a pulsed laser. The beam can be shaped as a sheet and still contain enough energy to make the scattering well above the signal-to-noise ratio. Coupling the signal collection with an intensified CCD camera makes possible 2D imaging of scalars such as species concentrations, mixture fractions or temperature. Instantaneous mapping of scalar fields gives valuable information on the turbulence scales (their size and anisotropy), and their coexistence in a given area of the mixing flow. Another advantage of the planar technique is that once the collected signal has been reduced to a physical variable, for example species concentration, it is possible to calculate other important characteristics of the flow as the spatial concentration correlation functions. The scattering process investigated can be of the type Rayleigh or Raman, as described above, thus yielding instantaneous species concentrations or temperature fields, or it can be other laser-molecule interaction processes such as laser induced fluorescence (LIF). In combustion LIF is often used to probe intermediate species like OH, CH, and C_2 amongst others, or NO. These radicals are produced by the flame and serve also as a marker of the flame front, or of the region where they are generated. In non-reacting flows, seed gas must be added generally, as for example acetone which has strong fluorescent properties. In a similar manner, seeding one of the streams with micrometric size particles, the Mie scattering process becomes an indicator of the mixture fraction.

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REFERENCES

[1] Kerstein, A.R., “A linear eddy-model of turbulent scalar transport and mixing”, *Comp. Sci. and Tech.* 60, 391-421 (1988)

[2] Kerstein, A.R., “Linear-eddy modelling of turbulent transport. Part 6. Microstructure of diffusive scalar mixing fields”, *J. Fluid Mech.* 231, 361-394 (1991)

[3] Magnussen, B.F. and Hjertager B.H., “On Mathematical Modeling of Turbulent Combustion with Special Emphasis on Soot Formation and Combustion”, *16th Symposium (Internatl.) on Combustion*, The Combustion Institute, Pittsburg, Pennsylvania, pp. 719-726 (1976)

[4] Magnussen, B.F.,”On the structure of turbulence and a generalized eddy dissipation concept for chemical reaction in turbulent flow”, 19th AIAA Aerospace Science Meeting, St. Louis, Missouri, USA, Jan. 12-15 (1981)

[5] Melaaen, M.C.,”Analysis of curvilinear non-orthogonal coordinates for numerical calculation of fluid flow in complex geometries”, Ph.D. thesis, University of Trondheim, Division of Thermodynamics, 1990

[6] Gran, I.R., “Mathematical modeling and numerical simulation of chemical kinetics in turbulent combustion”, Ph.D. thesis, University of Trondheim, Division of Mechanics, Thermo- and Fluiddynamics, 1994

[7] Yueng, P.K. and Pope, S.B., “Differential diffusion of passive scalars in isotropic turbulence”, *Phys. Fluids A* 5, 2467 (1993)

[8] McMurtry, P.A., Menon, S., and Kerstein, A.R., “Linear Eddy Modeling of Turbulent Combustion”, *Energy & Fuels*, 7, 817-826 (1993)

[9] Drake, M.C., Pitz, R.W., and Lapp, M., “Laser measurements on non-premixed H_2-air flames for assessments of turbulent combustion models”, *AIAA J.* 24, 905 (1986)

[10] Chen, J-Y, Bilger, R.W., and Dibble, R.W., “PDF modeling of turbulent nonpremixed CO/ H_2/N_2 jet flames with reduced mechanisms”, *23rd Symposium (Internatl.) on Combustion*, The Combustion Institute, Pittsburgh, p. 775 (1990)

[11] Dibble, R.W., Kerstein, A.R., Long, M.B., Yip, B., and Lyons, K., in: T. Takeno (Ed.), Turbulence and Molecular Processes in Combustion, Elsevier Science, Amsterdam, 1993, pp. 303-310

[12] Dibble, R.W. and Long M.B, Investigation of differential diffusion in turbulent jet flows using planar laser Rayleigh scattering”, *Combust. Flame* 143, 644–649 (2005)

[13] Barlow, R.S. and Carter, C.D., “Relationships among Nitric Oxide, Temperature, and Mixture Fraction in Hydrogen Jet Flames”, *Combust. Flame* 104, 288–299 (1996)

[14] Barlow, R.S., Frank, J.H., Karpetis, A.N., and Chen J.-Y., "Piloted methane/air jet flames: Transport effects and aspects of scalar structure", *Combust. Flame*, 143, 433–449 (2005)

[15] Bilger, R.W., Stårner, S.H., Kee, R.J., "On reduced mechanisms for methane-air combustion in nonpremixed flames", *Combust. Flame* 80, 135-149 (1990)

[16] Klimenko, A.Y. and Pope, S.B., "The modeling of turbulent reactive flows based on multiple mapping condition", *Phys. Fluids* 15, 1907–1925 (2003)