Effect of Surface Catalysis on Measured Heat Transfer in Expansion Tunnel Facility

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DOI: 10.2514/1.A3327

Nomenclature

\( h_0 \) = total enthalpy, MJ/kg
\( M \) = Mach number
\( Re \) = unit Reynolds number
\( T \) = temperature, K
\( U \) = velocity, m/s
\( \gamma \) = catalytic reaction efficiency, reactant loss efficiency
\( \rho \) = density, kg/m^3
\( \theta \) = angle from stagnation point, deg

I. Introduction

The effect of gas/surface interaction in the form of surface catalytic recombination occurring in hypersonic flows as the reacting gas behind the bow shock dissociates can have a significant impact on the resulting heat transferred to the surface. Recombination that occurs either in the boundary layer or locally at the surface increases the net level of heat transferred to the surface due to the chemical energy released from the recombination process. As vehicle speed and shock layer temperature increases, the component of heat transfer due to catalytic recombination can become a dominant portion of the total heat load on the surface. To understand the measured heat transfer rate in a ground test experiment, the catalytic phenomena contributing to the heat transferred to the test article must be characterized or found through other means to validate computational fluid dynamic (CFD) simulations or extrapolate the ground test measurements to flight.

A number of studies have been made in recent years regarding measurements made in reflected shock tunnels addressing the observed level of catalytic heating in air, carbon dioxide, and nitrogen environments [1–3]. In many cases, measured heat transfer rates have been observed to be consistent with very high efficiencies of recombination or nonphysical processes on the surface. One of the main conclusions from these works is that it is unclear if the observations about surface catalysis in reflected shock tunnel environments are in some way linked to the residual nonequilibrium, excitation, or chemical state of the freestream test gas (or at least inadequate description of it using current, state-of-the-art methods) [4]. The catalytic response observed in such tests is suspicious because it is not in line with general expectations from literature.

II. Background of Experimental Facility and Numerical Tools

A. Experimental Facility

The LENS-XX facility [5,6] is a new, large-scale expansion tunnel facility brought online during the past several years. Unlike reflected shock tunnels (which drive strong shocks through the test gas and stagnate it at very high pressures and temperatures), expansion tunnels do not stagnate the test gas at any time before it flows over the test article. Instead, the test gas is set into motion by a single shock of only weak-to-moderate strength, and the bulk of the enthalpy in the flow is added as kinetic energy directly by using an unsteady acceleration to increase the velocity. Using this technique, approximately 1 ms of test time is possible at very high-velocity conditions while maintaining a thermally cool gas state. To date, the LENS-XX facility has performed tests up to 8.4 km/s (36 MJ/kg). Determination of test conditions for this facility uses the CUBRC High Enthalpy Expansion Tunnel Analysis (CHEETAh) code described by MacLean et al. [7]. This code solves for the intermediate states of the primary and secondary wave systems incorporating equilibrium chemistry, ionization, and thermodynamic excitation mechanisms. This tool provides a simple and accurate way to determine the freestream condition using measured data from the facility for each run. Given the newness of the facility, we cannot yet hope to fully quantify the uncertainty of the freestream condition specification, but measurements of the freestream velocity using tunable diode laser absorption spectroscopy Doppler shift have so far always been found to agree with the prediction to within the accuracy of the measurement (<3%). This provides confidence in the quality of the freestream conditions.

B. Numerical Tool

The primary production CFD tool used for LENS facility design, design-of-experiment, and data validation is the Data-Parallel Line-Relaxation (DPLR) code [8] licensed by NASA Ames Research Center (ARC), Version 4.02.2. DPLR is a multiblock, structured, finite volume code that solves the reacting Navier–Stokes equations including generalized finite rate chemical and thermal nonequilibrium effects. As it is a general code, a large number of chemical, thermal, and transport models are available. By default in this analysis, transport properties are computed using a collision integral database [9] with the Yos mixing rule (Gupta et al. [10]) and self-consistent effective binary diffusion (SCEBD) method [11]. Chemical modeling uses the rates proposed by Park [12] with modified Zel’Dovich reaction rates proposed by Bose and Candler [13,14]. Thermal modeling assumes a single nonequilibrium vibrational temperature with the Park T – T\( _V \) coupling model [15].
The DPLR code has several options available to model surface catalysis at the solid surface boundary. Boundary conditions for each reacting species are individually computed from species mass balance at the surface, where species diffusion balances catalytic production for a nonablating wall and a self-consistent diffusion model enforces total mixture diffusion flux conservation. The noncatalytic wall boundary condition enforces zero catalytic production for each species, which implies that diffusion flux for each will be zero. The “super-catalytic” boundary condition sets a mixture composition explicitly at the surface that coincides with the lowest energy composition of the gas without regard for reaction kinetics or reactant availability. The super-catalytic boundary condition enforces a nonphysical set of catalytic production rates at the surface, but does provide an upper limit on energy release at the surface and thus a conservative estimate of heat transfer rate. The specified reaction efficiency (SRE) model enforces a user-specified efficiency \( \gamma \) for homogeneous catalytic reactions based on the fraction of reactant species that reach the surface for which a recombination event occurs. The SRE model relies on a single empirical parameter \( \gamma \) for each reactant species in the system that represents the effective loss of that reactant at the surface, which may, in general, be a function of any number of things including pressure, surface temperature, surface defects, surface reaction sites, surface roughness, gas composition, etc. In the case of the air systems studied here, this amounts to two independent reactions, \( \text{O} + \text{O} \rightarrow \text{O}_2 \) and \( \text{N} + \text{N} \rightarrow \text{N}_2 \), each with an associated efficiency \( \gamma_0 \) and \( \gamma_{N_2} \) (in this study, the two \( \gamma \) values were kept the same). A limitation of the SRE model for air systems is that the surface is considered inert to nitric oxide because its inclusion implies multiple outcomes for each reactant and could only be included by assuming a number of additional empirical parameters. Because the SRE model efficiency is not directly linked to a physical reaction pathway, a generalized, physics-based finite rate surface chemistry model [16,17] has been recently implemented in the DPLR code. However, such a model requires a significant number of parameters and has not yet gained widespread use in the literature.

C. Experimental Measurement Techniques

Accurate heat transfer measurements are made on cold-wall (300 K) stainless-steel models using several types of instruments. The primary gage is the platinum thin-film gage, which is a platinum resistance thermometer painted on a substrate of Pyrex. These gages can be installed isolated on a Pyrex island of between 1 and 3-mm in diameter on an otherwise metallic surface or painted along a continuous Pyrex strip to provide continuity of surface properties. A very thin layer of magnesium fluoride (MgF\(_2\)) is deposited on the surface of the gage before the initial installation. Each gage is individually calibrated across the expected temperature range. The second type of instrument is the chromel-constantan coaxial thermocouple. The thermocouple junction is formed by depositing a layer of chromium across the surface of the gage, which is the material exposed on the surface upon installation.

In both cases, the short runtime of the shock and expansion tunnel facilities allows the heat transfer to be reduced from measured surface temperature history using one-dimensional conduction on a semi-infinite solid. The short duration of flow in the facilities limits the temperature rise that will occur during the runtime. Ending temperature varies with conditions, but, for the types of conditions considered here, temperature rises for the thermocouples will be on the order of 10 K and for the thin-films on the order of 100 K over the approximately 1 ms time period.

Standard uncertainties in the heat transfer sensors are ±5% for thin-film gages and ±8% for coaxial thermocouple gages (using conduction properties specified by the manufacturer). In this work, we report error bars on each individual sensor using the standard deviation of that sensor during the averaging window, which incorporates sensor uncertainty as well as facility and freestream fluctuation. In most cases, the standard deviation is similar to or just slightly exceeds the quoted sensor uncertainty, providing some notional indications of the flow quality.

D. Expected Results from Literature

Unfortunately, literature on the specified reaction efficiencies of oxygen atom and nitrogen atom recombinations varies wildly, presumably from the bias inherent in each experimental setup and material sample source. We loosely may expect nitrogen atom recombination efficiencies to be anywhere from 0.0007 to 0.07 and oxygen recombination efficiencies to be from 0.005 to 0.2 for metallic surfaces [18–21]. A recent study by Marschall et al. [22] found nominal oxygen atom recombination efficiencies of 0.016 for stainless steel, 0.0053 for platinum, 0.046 for constantan, and 0.0068 for chromel, which seem reasonable median values to expect. It is also not fully clear yet whether the model material or the gage material most influences the effect of catalysis on the measured heat transfer, although some preliminary analysis suggests that the gage material may matter most [23]. Further, while removing the chromium layer on the thermocouples would be obvious by a breaking of the junction, it is not obvious how resilient the MgF\(_2\) coating on the thin-film gages is against the shearing force of the airflow.

III. Results

A number of measurements of surface heat transfer have been made on fundamental shapes in the LENS-XX facility targeting the assessment of catalytic response of our instrumentation to reacting flow conditions. The two main test articles, shown in Fig. 1, are a 7.6-cm (3 in.) diameter hemisphere nosetip probe and an 8.9-cm (3.5 in.) diameter planar cylinder model. The hemisphere model was instrumented with four thermocouple type gages located at positions of 0, 30, 50, and 70 deg from the stagnation point. The cylinder was instrumented with parallel rays of thin-film and thermocouple gages located at 0, 15, 30, 45, 60, and 75 deg from the stagnation line. A number of runs for different conditions and test gas compositions were made to test these two models in the LENS-XX facility. Run conditions selected for this study are listed in Table 1. By default, the model has been assumed to have a fully accommodated, isothermal wall temperature of 300 K for all cases. Further, the freestream test gases are quiescent (having no nonequilibrium thermodynamic excitation or chemical dissociation) for all cases. The Reynolds numbers for all conditions are sufficiently low that transition is not
expected, so laminar flow may be assumed to make CFD predictions to compare with the experimental data.

The measured and predicted heat transfer results on the hemisphere model for Runs 64 and 65 are shown in Figs. 2a and 2b. These two runs employed pure nitrogen as a test gas and the total enthalpy of the test flow was kept sufficiently low to minimize the effect of surface catalysis to the model. For these two runs, the thermocouple located at the 30 deg position on the hemisphere was not working, but the measured data is shown at the three other locations, including the stagnation point. As the results with the noncatalytic and supercatalytic boundary conditions show, the agreement between theory and data is very good for both runs. These results, where no significant catalytic heating component exists, demonstrate that the operation of the instrumentation and the determination of the freestream conditions are sufficiently accurate to make a good comparison of purely convective heating to a nosetip or blunt body in laminar flow in the LENS-XX facility.

The same hemisphere model with the same instrumentation was used in reacting air at 17 MJ/kg, where the comparison between the CFD prediction and the data is shown in Fig. 2c. The second gage located at 30 deg was included for this run (a wiring problem was repaired). Here, in addition to the noncatalytic and supercatalytic boundary condition prediction, the SRE model has been used by arbitrarily assuming a reaction efficiency, $\gamma = 0.01$, for both $N_2$ and $O_2$ homogeneous recombination that was empirically observed to match the data reasonably well. It may be readily noted that, in contrast to the observations from the reflected shock tunnel environment, the supercatalytic model significantly overpredicts the heat transfer rate.

For the cylinder model, the comparison between prediction and experiment for Run 77 (air at 10 MJ/kg) is shown in Fig. 3a and the comparisons for Runs 73 and 74 (air at 17 MJ/kg) are shown in Figs. 3b and 3c. For Run 77, the SRE model assuming a reaction efficiency, $\gamma = 0.01$, also best matches the data. For Runs 73 and 74, the best match is obtained assuming a reaction efficiency, $\gamma = 0.003$. Given that reaction efficiency is not associated with a particular physical process, there is no reason to presume that exactly the same value should hold across different conditions and different geometries. The response from both the thermocouples and the thin-film gage types was observed to be consistent for all of these cases. In fact, the thermocouple and thin-film at the stagnation point for Run 74 (Fig. 2c) are so consistent that it is difficult to differentiate them on the graph.

For the highest enthalpy Run 75 (air at 26.4 MJ/kg), the measured and predicted heat transfer distribution on the cylinder model is shown in Fig. 3d. As was observed in all the earlier cases, the measured heat transfer is significantly lower than the supercatalytic limit. For the four thin-films located at 15, 30, 60, and 75 deg, the measured heat transfer is again most consistent with the SRE model using $\gamma = 0.01$. At this condition, however, the three working thermocouples appear to show a bias toward lower heat transfer rates than the thin-films do. An SRE calculation with $\gamma = 0.002$ was found empirically to best-fit the thermocouple data. The reason for this discrepancy at this condition is not clear from the available data, but one possible explanation is that the two different exposed materials produce a different catalytic interaction at this condition that does not occur at the other conditions because of the different gas environments and surface temperature histories.

The thin-film at the stagnation point for Run 75 in Fig. 3d requires special consideration. This gage was constructed with a coating of magnesium fluoride that is four times thicker than the normal coating of 0.12-μm applied to thin-film gages. The measurement from this gage is definitively lower than the standard thin-film located at 15 deg and correlates better with the stagnation point thermocouple and the $\gamma = 0.002$ SRE calculation. Two possible explanations for this are that the thicker coating insulates the sensing element of the gage and lowers the observed heat transfer rate measured for short time-scales or that the coating applied to the gage results in a different catalytic production than the others because of differences in exposed surface topology. To further investigate this observation, a one-dimensional conduction analysis has been performed using the NASA Johnson Space Center CHAR [24] code by applying a constant heat flux of

<table>
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<th>Run #</th>
<th>$h_0$, MJ/kg</th>
<th>$Re_e$, $10^6$/m</th>
<th>$\rho$, kg/m$^3$</th>
<th>$U$, m/s</th>
<th>$T$, K</th>
<th>$M$</th>
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<th>model</th>
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<td>.37</td>
<td>.00105</td>
<td>4708</td>
<td>203</td>
<td>16.2</td>
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Fig. 2 Measured and predicted heat transfer results on hemisphere model.
500 W/cm² to the surface as a boundary condition. Gages were modeled with 0.12 and 0.48-μm of MgF₂ followed by a semi-infinite volume of Pyrex with the surface and the volume initially at 300 K. The heat flux at the interface station where the resistance element is located was monitored with time as shown in Fig. 4 over one millisecond, which is characteristic of the nominal steady-state test time for the LENS-XX facility for high-enthalpy conditions. As expected, the heat flux at the resistance element station for the case with the thinner (standard) coating increases more rapidly while the case with the thicker coating lags behind due to the longer heat soak time scale. When these calculated responses were averaged from 0.2 to 0.8-ms as might typically be done with an experimental signal (thus avoiding the very beginning and the very end of the run), a heat flux of 99.3% of the input heat flux is obtained for the 0.12-μm case and a heat flux of 97.2% of the input heat flux is obtained for the 0.48-μm case. The conduction analysis shows that there is a small bias toward a lower value that occurs with the use of thicker coatings over the resistance element, but this bias is less than 3% for the Run 75 stagnation point. The 15% lower value measured in the experiment far exceeds the 3% expected from conduction loss in this case. Although it is not possible to definitively say that the remaining 12% discrepancy in heat flux is due to a reduction in catalytic efficiency, this is the first time we have observed gages with different material characteristics behaving differently for the same reactive environment.

The results shown in this section use the same efficiency parameter γ for both N and O atom recombinations. In general, analysis of stagnation line profiles showed that much of the nitrogen recombination occurs within the gas phase of the boundary layer. Thus, there are fewer nitrogen atoms reaching the surface than oxygen atoms, but each nitrogen recombination event releases more energy than each oxygen atom event. For Run 75 shown in Fig. 3d, the contribution to the catalytic heating augmentation over the noncatalytic solution from oxygen and nitrogen atoms is roughly the same. By collecting additional experimental data with different gas mixtures, it would be possible to establish separate efficiencies for each element, but the goal of this effort was to demonstrate that a finite rate response is observed from the first few runs of data in the new LENS-XX facility and environment.

The SRE efficiencies found through the LENS-XX comparisons may be contrasted with a typical set of data on the cylinder model from the LENS-I reflected shock tunnel facility shown in Fig. 5. This condition has a total enthalpy of approximately 11 MJ/kg, and has the following freestream conditions: $U = 4464$ m/s, $\rho = 0.00379$ kg/m³, and $T = 656$ K. The freestream flow is partially dissociated with the following computed mass fractions: $c_{\text{N}_2} = 0.7397$, $c_{\text{O}_2} = 0.1905$, $c_{\text{NO}} = 0.0543$, and $c_{\text{O}} = 0.0155$. As the results show, the measured stagnation point heat transfer from the reflected shock tunnel environment using both thin-film and coaxial thermocouple sensors is above the predicted supercatalytic level of heating. The measured data trend somewhat toward the noncatalytic level at large angles from stagnation, but the reason for this trend remains unknown. Measurements in the LENS-I reflected shock tunnel facility on a spherical capsule shape at similar freestream
conditions showed macroscopic discrepancies between predicted and measured shock shape \([25]\), giving us further evidence to doubt our modeling of the state of the gas in the reflected shock tunnel environment.

IV. Conclusions

The first measurements in the new LENS-XX expansion tunnel facility have demonstrated the capability to make surface heat transfer measurements in the stagnation region of planar cylindrical and spherical nosetip flows with freestream total enthalpy from 4.5 to 7 km/s (10 to 26 MJ/kg). When measured heat transfer rates are compared with computational fluid dynamic simulations employing a specified reaction efficiency wall boundary condition and the noted set of thermal and chemical reaction rates and transport coefficient models, a reaction efficiency of between \(\gamma = 0.003\) and 0.01 was found to best match the available platinum/Pyrex thin-film gage measurements across the range of conditions. A reaction efficiency of \(\gamma = 0.002\) and 0.01 was found to best match the available chromium-plated chromel-constantan thermocouple measurements. Although reaction efficiency lacks physical interpretation, these values are within the median of the range published in the literature for general metallic surfaces. The levels of heat flux associated with complete catalytic recombination or higher that has been formerly measured in some reflected shock tunnel environments were not observed in this dataset.

Acknowledgments

This work was sponsored by the U.S. Air Force Office of Scientific Research under the Multidisciplinary University Research Initiative entitled Fundamental Processes in High-Temperature Hypersonic Flows, Prime Award FA9550-10-1-0563. The views and conclusions contained herein are those of the authors and should not be interpreted as necessarily representing the official policies or endorsements, either expressed or implied, of the U.S. Air Force Office of Scientific Research or the U.S. Government.

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G. Palmer
Associate Editor