

ASSESSMENT OF AEROTHERMAL HEATING AUGMENTATION ATTRIBUTED TO SURFACE CATALYSIS IN HIGH ENTHALPY SHOCK TUNNEL FLOWS

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ABSTRACT

The effect of gas/surface interaction in making CFD predictions of convective heating has been considered with application to ground tests performed in high enthalpy shock tunnels where additional heating augmentation attributable to surface recombination has been observed for nitrogen, air and carbon dioxide flows. For test articles constructed of stainless steel and aluminum, measurements have been made with several types of heat transfer instrumentation including thin-film, calorimeter, and coaxial thermocouple sensors. These experiments have been modeled by computations made with the high quality, chemically reacting, Navier-Stokes solver, DPLR and the heating results compared. Some typical cases considered include results on an axisymmetric sphere-cone, axisymmetric spherical capsule, spherical capsule at angle of attack, and two-dimensional cylinder. In nitrogen flows, cases considered show a recombination probability on the order of 10^{-3} , which agrees with published data. In many cases in air and CO_2 , measurements exceeding the predicted level of convective heating have been observed which are consistent with approximately complete recombination (to O_2/N_2 or CO_2) on the surface of the model (sometimes called a super-catalytic wall). It has been recognized that the conclusion that this behavior is tied to an excessively high degree of catalytic efficiency is dependent on the current understanding of the freestream and shock-layer state of the gas.

INTRODUCTION

In high enthalpy flows where chemical dissociation occurs in the shock layer of the vehicle, a significant fraction of the total heating to the surface can result from recombination on a catalytic material. A primary goal of ground tests in shock tunnels and other high enthalpy facilities is often to make measurements of convective heating in environments where shock/shock and shock/boundary-layer interactions, roughness augmentation, transitional heating, turbulence, and/or re-laminarization are important. In the ground test environment, metal models with specialized heat transfer instrumentation are employed for this purpose to isolate the convective heating component. The catalytic response of the particular TPS material chosen for the flight heat shield is considered separately to properly design the flight vehicle. However, the catalytic response of the metallic ground test model and

instrumentation, which may be different than the corresponding flight vehicle TPS system, must be characterized to isolate and accurately describe only the convective component, minimize vehicle conservatism and maximize vehicle performance or payload.

Currently, CUBRC operates the 48", LENS-I and LENS-II reflected shock tunnels [1], and the LENS-XX expansion tunnel [2]. In reflected shock tunnel facilities the stagnant test gas is shock-heated to very high enthalpy and then accelerated, while, in the expansion tunnel, kinetic energy is added directly to minimize chemical dissociation that can occur in the freestream of reflected shock tunnels at enthalpies above 5 MJ/kg. The capabilities of the current set of CUBRC facilities are shown in Fig 1. The LENS reflected shock-tunnel facilities were developed primarily to study the full-scale, hypervelocity flow physics of interceptors and air-breathing engine configurations. The scale and flow duplication capabilities of LENS are such that these vehicles can be studied at their full scale, inclusive of effects such as transition to turbulence, turbulent mixing from cross-flow jets and thrusters, duplicated flow chemistry, and other effects that are difficult or impossible to simulate at cold-flow or sub-scale conditions. The reflected shock tunnel facilities have been used to perform studies of fundamental flow phenomena like the double cone with success and to solve practical flight problems on vehicles like the Shuttle STS vehicle.

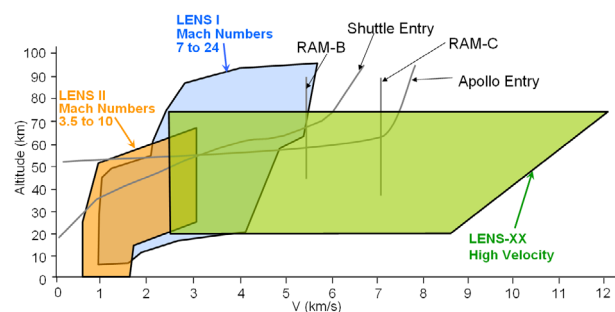


Figure 1. Velocity-Altitude Duplication Capabilities of CUBRC LENS Facilities

SURFACE BOUNDARY CONDITIONS AND NUMERICAL METHODS

All ground test studies in the LENS facilities are extensively calibrated and validated with numerical tools. The primary CFD tool used is the DPLR code provided by NASA Ames Research Center. DPLR is a

multi-block, structured, finite-volume code that solves the reacting Navier-Stokes equations including finite rate chemistry and finite rate vibrational non-equilibrium effects. This code is based on the data-parallel line relaxation method [3] and implements a modified (low dissipation) Steger-Warming flux splitting approach [4] for the convection terms and central differencing for the diffusion terms. Finite rate vibrational, rotation, and chemical relaxation is modeled in the code via the techniques described by Candler [5].

Surface catalysis is also a central issue in the study of real-gas effects in ground test environments. We have already shown the effects of catalysis on the measured heat transfer in a shock tunnel environment for a CO₂ test gas, where catalytic heating can be as large in magnitude as the convective heating, or more than 50% of the total [6]. In air, the effect of catalytic heating can be typically 20–40% of the total, and in nitrogen flows for LENS conditions, catalytic heating is usually no more than 15% of the total.

To model the effects of catalytic heating numerically, two extremes exist. The non-catalytic surface boundary condition enforces zero gradients of species densities at the surface. Since large levels of chemical energy remains trapped by dissociated atoms, this boundary condition will always predict the lowest level of surface heating. The super-catalytic boundary condition provides the highest possible level of heating because the mixture is returned to its lowest possible chemical energy state at the wall, meaning that the maximum amount of chemical energy has been returned to the other energy modes. The super-catalytic boundary is non-physical in that it does not identify a specific mechanism by which the recombination occurs, but assumes that it has occurred by some unidentified process without consideration of rate mechanics. In the case of short-duration facilities like LENS, the surface temperature of the model never rises appreciably above room temperature (at least as a fraction of total temperature), so the super-catalytic condition corresponds to a fully-accommodated chemical state in both CO₂ and air.

The DPLR code provides several models of finite-rate surface reactions for a variety of thermal protection and ground test materials, but the levels of catalysis predicted using these models generally under-predict the level of catalysis observed in the experimental measurements. Several careful experimental studies by Myerson [7] and Hartunian and Thompson [8,9] among others have demonstrated this effect in shock tunnel environments on metal surfaces. Currently, a satisfactory model of the detailed rate processes causing the observed catalysis effects on surface heat transfer under these conditions is unknown.

EXPERIMENTAL MEASUREMENT TECHNIQUES AND MODEL PREPARATION

With few exceptions, models developed for ground testing in the LENS facilities are made either of stainless steel or aluminum with inserted heat transfer or surface pressure instrumentation. CUBRC uses several types of heat transfer instrumentation to make measurements in both high and low enthalpy flows. An example of a model with all types of instrumentation installed in the surface is shown in Fig 2.

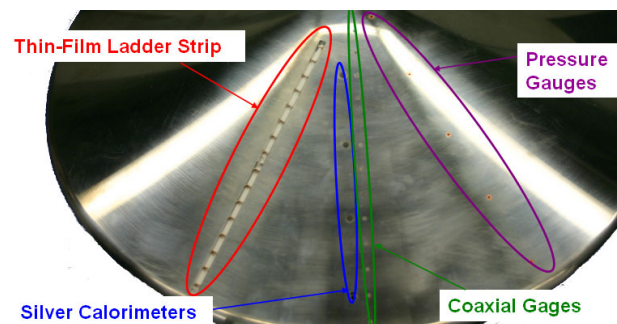


Figure 2. Sphere-cone Model Showing Gage Installation Techniques

Thin-film gages are a platinum resistance thermometer painted on a base of pyrex. The gage is coated with a layer of magnesium fluoride that is deposited on the surface of the gage. Although it is probable that the magnesium fluoride wears away after repeated runs in the facility and exposes the platinum underneath, there is no evidence that the catalytic behavior of the instrument changes at all with use. Thin-film gages can be installed either in small circular islands of pyrex within an otherwise metallic surface or in a continuous strip (a “ladder”) of pyrex providing continuity of the surface properties. Thin-film gages are the most accurate method of heat transfer measurement, but are most fragile and subject to damage. This type of gage is shown in Fig 3(a).

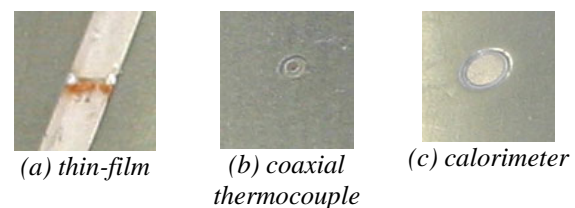
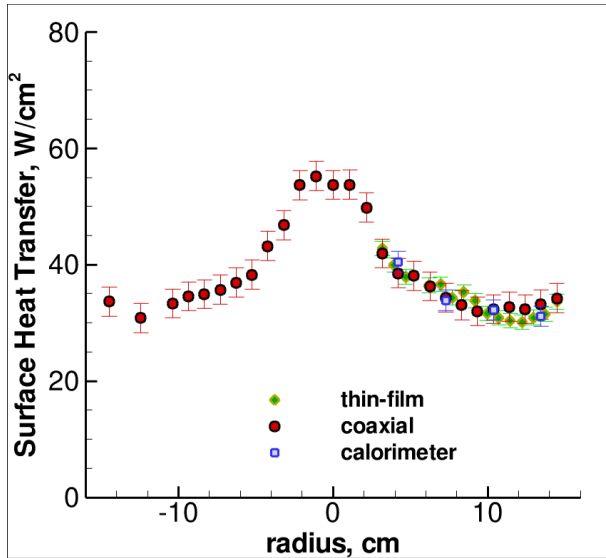


Figure 3. Types of Heat Transfer Instrumentation Employed in Making Aerothermal Measurements in High Enthalpy Flows

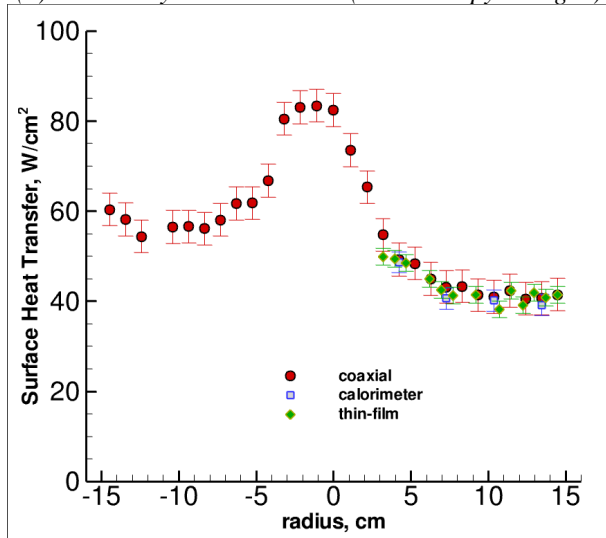
Coaxial thermocouples are also employed to measure the surface temperature. The thermocouples are composed of chromel and constantan with an insulating substrate between them. One-dimensional conduction is insured because the short duration runtime insures that heat does not have sufficient time to extend into the surrounding material. A coating of chromium is plated

over the thermocouple, helping form the junction. The chromium is the surface material exposed to the gas. This type of gage is shown in Fig 3(b).

Calorimeters made of pure silver are also employed to measure heat transfer. Temperature history is essentially monitored with a thin-film type sensor on the back face of the calorimeter. The calorimeter mass is insulated on the sides with a thin layer of pyrex to isolate it from the surrounding metal. A very thin coating of chromium is applied to the top of the silver. This type of gage is shown in Fig 3(c).



(a) Non-catalytic Environment (low enthalpy nitrogen)



(b) Catalytic Environment (dissociated CO_2)

Figure 4. Response of All Types of Heat Transfer Measurements on a Sphere-Cone Shape for Catalytic and Non-catalytic Flows

In each case where instrumentation is installed on an island in the metallic surface, a discontinuity in the catalytic response of the surface as seen by the flow can

exist. This catalytic jump effect can exhibit a change to the measured heat transfer as the species profiles of the boundary layer adjusts to the new surface. However, for each different gage with a different catalytic response, the magnitude of this effect will presumably be different. No such differences have been observed so far in any environment where dissociated species exist in the shock layer. Heating Measurement profiles are shown in Fig 4 for two cases. In Fig 4(a), a very low enthalpy nitrogen flow is shown where the gas remains in its molecular form and no dissociation occurs either in the facility nozzle or the shock layer. No possible catalytic heating exists in this environment and the response of the gages is due to convection only. In Fig 4(b), a case with carbon dioxide is shown where significant dissociation is occurring in the flow. CFD prediction suggests that as much as half of the total possible heat transfer to the body can be due to catalytic reactions. In this case as well, all three types of heat transfer instrumentation display the same level of heating to within the accuracy of the gages. No changes due either to different catalytic efficiencies or catalytic jump can be detected.

Before each run in the facility, the surface of the model is cleaned with acetone. Prior to each firing, the test chamber is evacuated and sits under vacuum for approximately one hour at levels typically around 10- μm Hg. This presumably encourages at least some desorption of contaminants on the surface that remain after the cleaning including water vapor.

Some consideration has been given to the transient response of the surface when the pulse of high energy flow first impinges on the model. This is expected to be rapid even in the timescale of the facility operation, but experimental evidence corroborates this assumption. A typical temperature history and corresponding heat transfer measurement from a high enthalpy air run is shown in Fig 5 for a thin-film gage in the stagnation region of a blunt body. As the heat transfer history shows, the level is steady for the entire window of time, indicating that any transient effects occurring on the surface are either very much faster or very much slower than the steady-state time period. It is also interesting to note the temperature rise during the run, which initially begins at room temperature (300 K) and rises about 200 K during the runtime. Work by Marschall, et al. [10] tested several substances including platinum in a side-arm reactor to estimate the catalytic response of these types of instruments. Their findings showed very little activity on platinum for either pure oxygen recombination or carbon monoxide oxidation at 300K but a significant activity was observed at 250°C for O_2+CO recombination. This finding suggests that thin-film gages might see an increase in heat transfer as the surface temperature rises during the run if there is a

significant concentration of molecular oxygen in the carbon monoxide flow, but heat transfer remains constant for both air and CO₂ flows as shown.

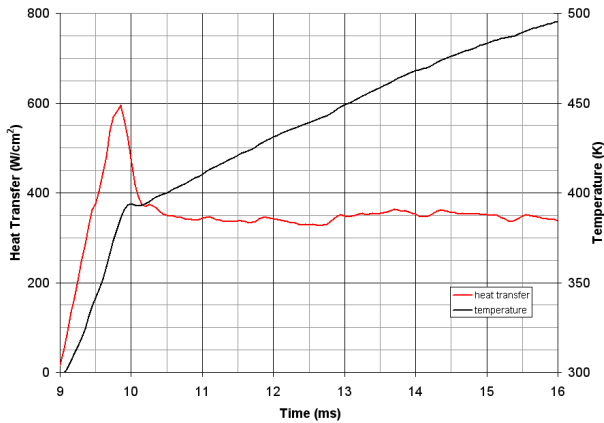


Figure 5. Typical Temperature and Heat Transfer Time History for Thin-film Gages in Catalytic Environments

Several test geometries are shown in Fig 6 that are considered in the following sections. Each of these shapes is blunt enough to dissociate the test gas in the shock layer for test conditions in the LENS facilities and is representative of real vehicles of current interest to the hypersonics community.

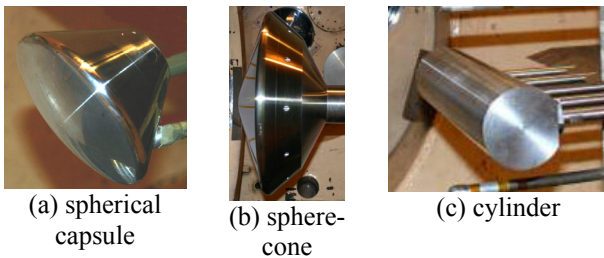


Figure 6. Test Geometries Considered in Results Sections of This Work

RESULTS IN NITROGEN GAS

Three runs were analyzed for which measurements on the capsule model shown in Fig 6(a) were made in pure nitrogen as a test gas and nominal total enthalpy near 10 MJ/kg. Discussion and tabulation of the freestream conditions are given by MacLean et al. [11]. In pure nitrogen there is a significant level of vibrational energy in the freestream because of the slow relaxation of nitrogen in the expanding gas, but this is adequately described with the use of a simple harmonic oscillator (SHO) model and a single vibrational temperature.

The solutions to these cases were computed with a two species (N₂, N), single reaction nitrogen model using several surface species boundary conditions. In the specific case of a nitrogen system, the catalytic wall considers only the single homogeneous reaction of atomic nitrogen recombining to form molecular

nitrogen. There is no competition for available surface sites, heterogeneous reactions, etc. as in more complex scenarios. The super-catalytic wall boundary enforces complete recombination to molecular nitrogen (N₂) while the finite rate wall allows for the recombination of a specified percentage of the atomic nitrogen reaching the surface with a recombination probability for the reaction.

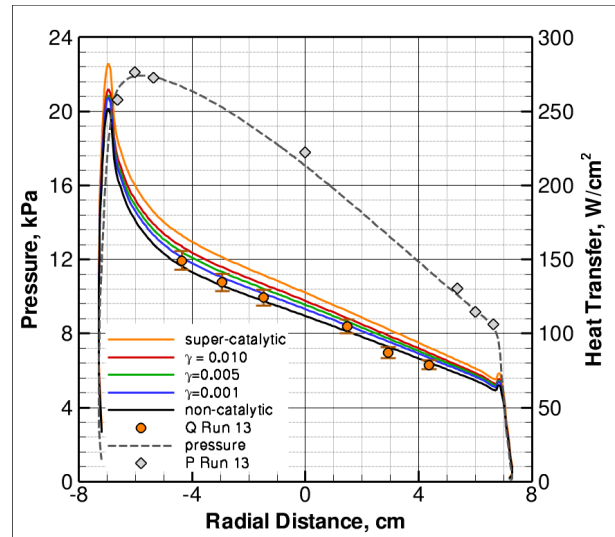


Figure 7. Predicted and Measured Surface Heating and Pressure for Spherical Capsule for Run 13

The comparison of measurement and prediction for Run 13 at a total enthalpy of 10.4 MJ/kg and angle of attack of 28° is shown in Fig 7. The percentage of catalytic heating on the main body where the measurements are made is about 15% as determined by the difference between the super-catalytic and non-catalytic wall solutions. In this case, the heat transfer measurements correlate well with the non-catalytic prediction. However, when considering a range of finite rate catalytic surface results, values of as much as $\gamma=0.001$ is not significantly different than the non-catalytic level and can be seen to match the heating data just as well when considering the measurement uncertainty. It is also important to note that the super-catalytic solution does NOT describe the measured heating adequately. Values of $\gamma=0.01$ and above also do not seem to match the data.

The comparison for Run 14 at a total enthalpy of 10.3 MJ/kg also contains a catalytic heating component of about 15% of the total as shown in Fig 8. Here, the mean data clearly does not match either the super-catalytic or the non-catalytic solutions. Tests with a finite-rate catalytic boundary revealed that a recombination efficiency of $\gamma=0.01$ best matches the data. This is somewhat higher than the level found for the previous case, but still on the same order of magnitude.

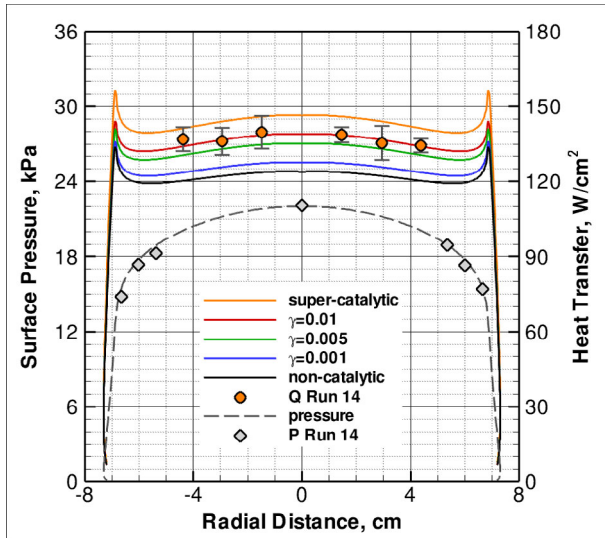


Figure 8. Predicted and Measured Surface Heating and Pressure for Spherical Capsule for Run 14

Run 18, despite being at 9.0 MJ/kg total enthalpy, shows considerably less dissociation both in the freestream and in the hot shock layer. The compared surface data is shown in Fig 9 for this case. Here, the percentage of catalytic heating is about half of the previous cases (<10%), which is consistent with the reduced dissociation observed in the flowfield. At less than 10%, the catalytic heating component borders on insignificant as illustrated by the uncertainty in the measurements. The mean measurements seem to correlate best with the super-catalytic solution, but a finite rate recombination efficiency of $\gamma=0.01$ is also well within the uncertainty bounds of the data. Even values as low as $\gamma=0.001$ are not significantly out of bounds.

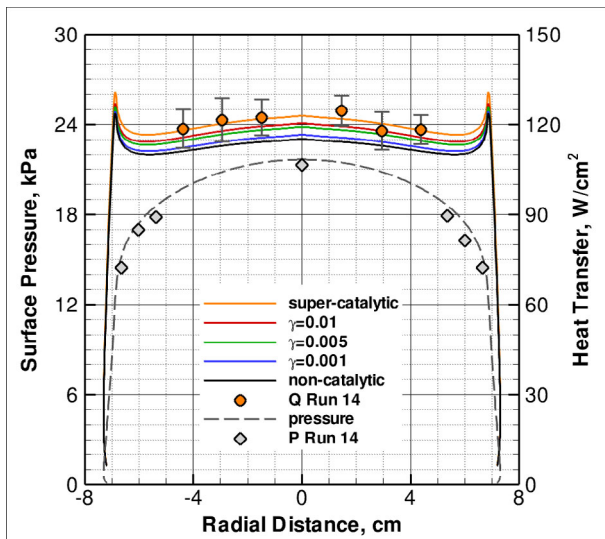


Figure 9. Predicted and Measured Surface Heating and Pressure for Spherical Capsule for Run 18

In summary, for a simple catalytic system of pure nitrogen where a single recombination mechanism occurs on the surface of the model, values between 0.001 and 0.01, or on the order of 10^{-3} were found to best match the available data. A search of literature for a 300K stainless steel surface revealed a surprisingly large range of acceptable recombination efficiency spanning 0.0007 to 0.07 [12]. The results shown here are right in the middle of the accepted range for nitrogen flows.

RESULTS IN AIR TEST GAS

For several cases considered in air on the same spherical capsule shown in Fig 6(a), the conclusions about catalytic activity in high enthalpy airflows are very different than was observed for nitrogen. Here, the surface kinetics are more complex since both atomic oxygen and atomic nitrogen can potentially adsorb onto the surface. In reality, for flows up to 10 MJ/kg, very little atomic nitrogen is created in the flow and the chemical energy is mainly in the form of atomic oxygen and nitric oxide.

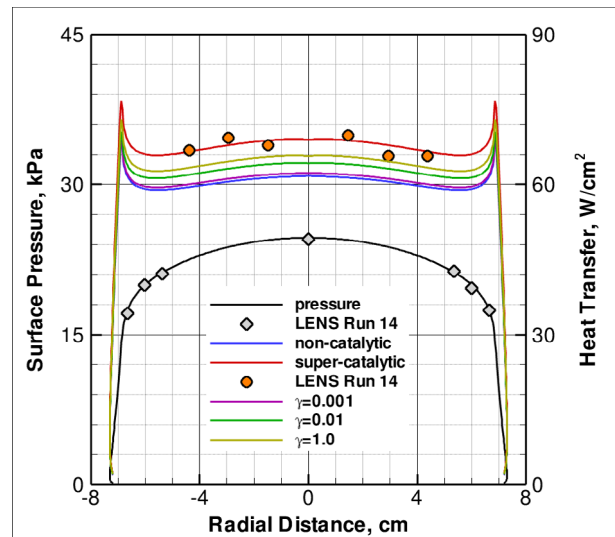


Figure 10. Predicted and Measured Surface Heating and Pressure for Spherical Capsule for 5 MJ/kg Air

At an enthalpy of 5 MJ/kg the comparison of measured and predicted heat transfer is shown in Fig 10. Here, the case was simulated with non-catalytic and super-catalytic wall boundaries as before. The data shows very good agreement with the super-catalytic solution, which implies that both atomic oxygen and nitric oxide are removed by the surface. DPLR models homogeneous surface reactions only, so the surface is considered non-catalytic to nitric oxide for the finite-rate solutions. The maximum value of $\gamma=1.0$ implies that every single oxygen atom which reaches the surface is converted to molecular form. As can be seen, the predicted heating for that case is still insufficient and

breakdown of nitric oxide must also be considered to match the available data. Although oxygen atoms are known to be very reactive and recombination probabilities as high as 0.1 have been reported [13,14], complete recombination seems very dramatic. Additionally, several authors have reported on the effect of nitric oxide in surface chemistry in competition with nitrogen and oxygen atom recombination [15-17], but here it appears that a mechanism to describe nitric oxide reduction as in automotive catalytic converters is required.

When a much higher enthalpy is considered, as shown in Fig 11 at about 15 MJ/kg, a very different result occurs. The total enthalpy for this condition is greater than the level at which flow can be tailored in the shocktube, but measurements of freestream velocity [18] showed that, even at this condition, the total enthalpy of the flow is within a few percent of the target level and remains steady for several milliseconds. So, despite the very high enthalpy of the facility, a low heat transfer is measured that seems to fall significantly below even the non-catalytic level.

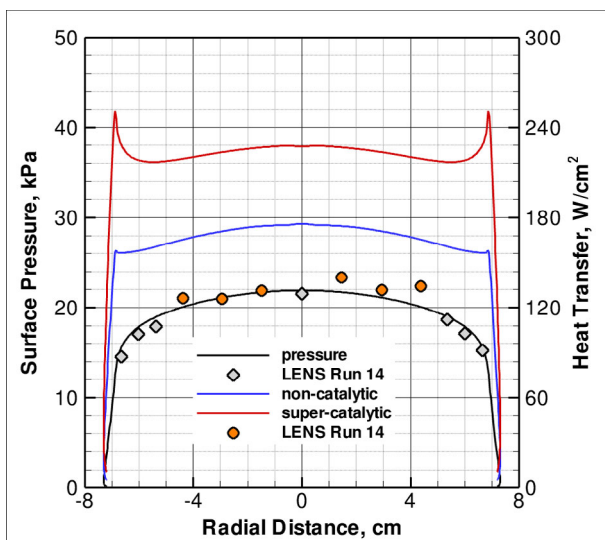


Figure 11. Predicted and Measured Surface Heating and Pressure for Spherical Capsule for 15 MJ/kg Air

However, comparison of the predicted shock shape of the body to the shape measured with a Schlieren image reveals that the CFD does not accurately predict the correct standoff distance in the stagnation region. This is shown in Fig 12. The CFD solution predicts stand-off considerably less than measured. The analysis of Fay and Riddell [19], for example, shows that stagnation point heat transfer decreases with increasing shock stand-off distance. This implies that the CFD solution might be predicting higher heat transfer loads than if it were able to match the larger stand-off distance. In any case, the inability to accurately predict shock stand-off distance, combined with measurements of nitric oxide

temperature and concentration in the freestream that are vastly different than predicted [20], suggests that a strong non-equilibrium state exists in the freestream that is not adequately described by current physical models. This effect was observed in work looking at the non-equilibrium effects in carbon dioxide flows in the facility [21]. In any case, it is not possible to make any definitive conclusions about the surface catalytic response for this flow until the shock shape can be accurately modeled.

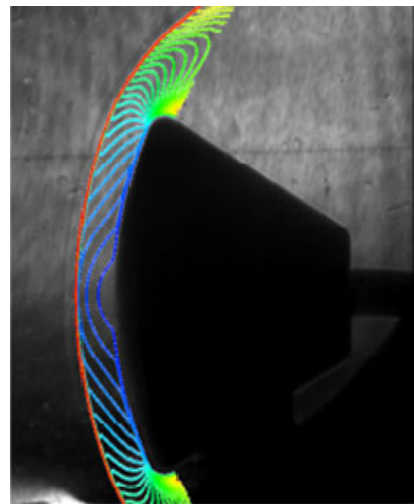


Figure 12. Predicted and Measured Shock Shape for Spherical Capsule in 15 MJ/kg Air

Results on the circular cylinder shape shown in Fig 6(c) at a total enthalpy of 11 MJ/kg showed good agreement with the shape of the shock [20]. The corresponding heat transfer comparison is shown in Fig 13. Here, as at 5 MJ/kg on the capsule model, the super-catalytic wall best matches the data in the stagnation region, although there is limited evidence that as the flow accelerates, the last few gages seem to show some decrease in catalytic heating. At the stagnation point, the finite-rate catalytic wall boundary with maximum recombination efficiency ($\gamma=1.0$) gives a value only slightly less than the super-catalytic solution, since, at this higher enthalpy, nitric oxide exists in lower quantity in the shock layer.

The reason for the differences between the spherical capsule result and the cylinder result may be due to several things such as shape streamline patterns, boundary layer growth, etc. However, most importantly, the freestream density for the cylinder case is approximately three times as for the spherical capsule runs at similar enthalpies. This and other evidence collected during this work suggests that the effect of non-equilibrium in the freestream is largely mitigated by increased facility pressure. Apparently, the additional molecular collisions in the nozzle expansion produce a state that is closer to the predicted freestream condition.

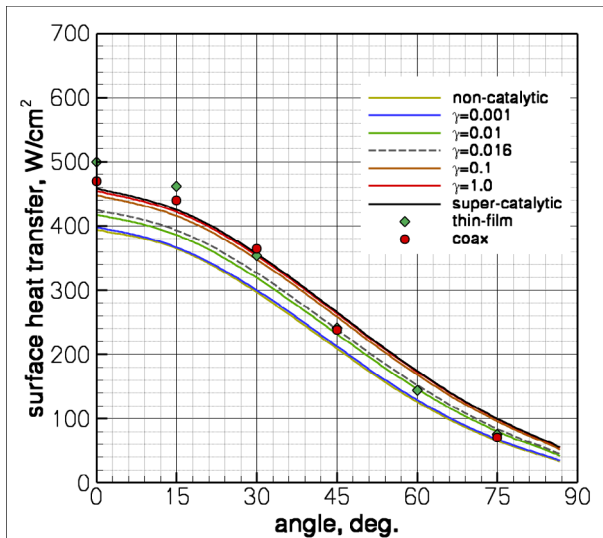


Figure 13. Predicted and Measured Heat Transfer for 8.83-cm Diameter Cylinder at 11 MJ/kg Air

RESULTS IN CARBON DIOXIDE TEST GAS

In carbon dioxide flows, the greatest potential for catalytic heating exists of any of the test gas mixtures considered here. Analysis of non-equilibrium effects in carbon dioxide flows have already shown the dramatic effect of non-equilibrium energy on the expanding gas and freestream thermochemical state [21]. The effect of non-equilibrium in the freestream for a low pressure, moderate total enthalpy (5 MJ/kg) condition in CO₂ over a sphere-cone shape shown in Fig 6(b) is shown in Fig 14 which plots the predicted shock stand-off distance as a function of the percentage of total enthalpy of the freestream which is trapped in either vibrational or chemical forms. The nominal conditions for this case predicts approximately 15% chemical energy and equilibrium vibration, which under-predicts the shock stand-off by more than a factor of two times. To match shock stand-off distance and shape with this simplistic treatment of the freestream, a minimum of 43% of the total enthalpy of the flow must reside in non-equilibrium energy.

Shown in Fig 15 are the corresponding super-catalytic heat transfer predictions for a series of these possible freestream conditions with percentages of the total enthalpy contained in the vibrational modes. On the conic frustum of the vehicle, the agreement between the super-catalytic wall models and the data is very good, again noting that all types of sensors produce a consistent response. Near the stagnation point on the spherical portion of the body, the different freestream conditions have a dramatic influence on the heating prediction, with freestream conditions best matching the shock shape on the body also showing the best agreement between the data and the super-catalytic solutions.

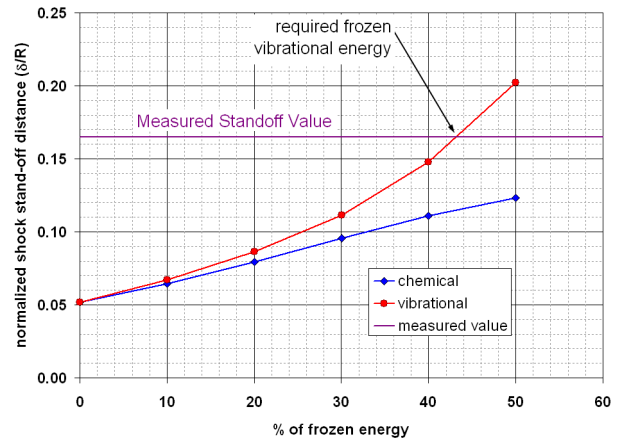


Figure 14. Comparison of Effect of Non-equilibrium Freestream Energy Content on Shock Stand-off Distance in 5MJ/kg CO₂

Although the evidence suggests that the super-catalytic wall boundary more closely represents the data when the predicted shock is properly aligned with the observed shape in the experiment, it is too difficult to make a judgment about the catalyticity of the surface for this flow since the freestream conditions in the facility for this particular condition in CO₂ are so obviously unknown.

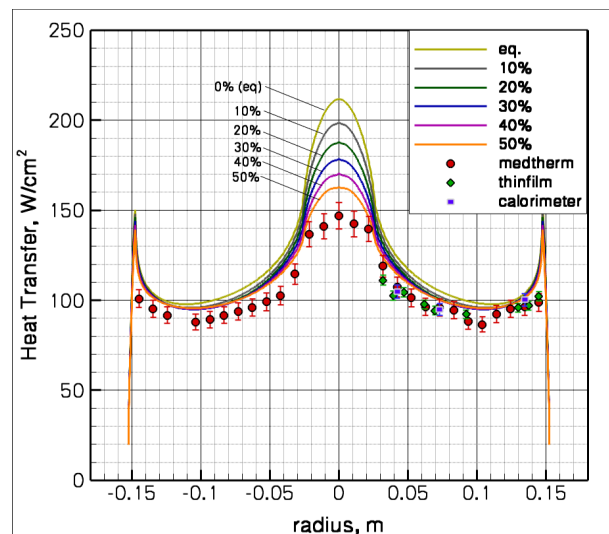


Figure 15. Predicted Super-catalytic and Measured Heat Transfer for Sphere-cone in 5 MJ/kg CO₂ for Several Non-equilibrium Energies

CONCLUSIONS

In summary, we have shown that the catalytic response on test articles in ground tests for aerothermal characterization is a central issue for blunt body vehicle design. In several cases in a reflected shock tunnel facility where chemical dissociation and thermodynamic excitation occurs in low pressure conditions, the catalytic response of the surface is difficult to interpret

without additional understanding of the freestream state of the gas.

In cases in nitrogen, catalytic recombination probability was found to be most likely between 0.001 and 0.01, a finding that is centered within the range of published values in the literature. In air at conditions where the shock shape of the test article was properly replicated by CFD, the response was always found to correspond to super-catalytic heating, implying both complete recombination of atomic oxygen but also nitric oxide on the surface. In cases in air and CO₂ where the shock shape was not accurately predicted by CFD, the significantly higher heating predicted by the super-catalytic wall compared to the data was found to be tied directly to the under-prediction of shock standoff distance. An accurate assessment of the surface response of the instrumentation in these flows is impossible to make.

Future work in this area will consist of making a similar database of measurements in the LENS-XX expansion tunnel facility, where we can generate similar conditions with little or no freestream non-equilibrium in order to assess the impact that phenomenon has on the surface heating profiles.

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