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EFFECTS OF SURFACE CATALYSIS ON HEAT TRANSFER TO SHUTTLE ORBITERS

Howard A. Stine

Ames Research Center
Moffett Field, Calif. 94035

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EFFECTS OF SURFACE CATALYSIS ON HEAT TRANSFER TO SHUTTLE ORBITERS

by

Howard A. Stine

Abstract

The aerothermodynamic environment generated during much of the shuttle orbiter's entry flight consists of a flow of atoms and molecules. Oxygen is completely atomic and nitrogen is partly atomic and partly molecular.

Two kinds of interactions of atoms with the shuttle surface are reviewed. First, the catalytic efficiency of the surface can change the incident heat transfer rate by factors of from two to three, depending upon whether the surface is chemically inert or chemically active, over the enthalpy range of peak heating. A corresponding surface radiation equilibrium temperature change of about 240° is calculated to be possible. Second, although information is scarce, indications are that oxidation rates of metals under attack by oxygen atoms can be one to two orders of magnitude greater than those corresponding to attack by oxygen molecules.

Substantial improvement in shuttle operational capability, survivability, and longevity evidently can accrue if interaction of atomic oxygen with the shuttle surface can be suppressed.

INTRODUCTION

The Space Shuttle Orbiter is designed for many returns to earth along lifting entry trajectories that will limit the magnitudes of the incident aerodynamic heat transfer rate to those that will allow large areas to be cooled by radiation.[1] Along the trajectories to be flown, the zone of maximum aerodynamic heating will be encountered at altitudes between 175,000 and 250,000 ft.[2], where air density is such that the chemical reaction of dissociation of the air molecules into atoms on passing into a zone of high temperature will usually go to completion over large areas of the vehicle. On the other hand, dissociation will not always be followed by equilibrium recombination of the atoms into molecules in zones of lower temperature. Under these circumstances, the catalytic efficiency of the shuttle skin has important effects on the magnitude of convective heat transfer loads imposed on the vehicle.

Chemical reactions of dissociation and recombination are influenced not only by variations in the local "inviscid" flow in the neighborhood of the vehicle, but also by the dissipative and diffusion processes that take place in the boundary layers. In particular, as is well known, when altitude is increased from 200,000 ft. to 300,000 ft. and for speeds near satellite velocity, the flow in a shock layer and around the flanks of a blunt body will be largely dissociated, but the chemical recombination rate will vary from near equilibrium (recombination time fast compared to flow time) to completely "frozen" (flow time fast compared to recombination time).[3] At the higher altitudes, the controlling chemical reaction is that of dissociation behind the shock wave structure, but at the lower altitudes it is the recombination process in the boundary layer.[3,4] In the latter case, the energy of dissociation will not appear in the gas as sensible heat, for all practical purposes, unless three atoms participate in the so-called "homogeneous" recombination process, a rare event when the gas density is low. However, the third participant in the recombination process need not be a gas atom, and a "heterogeneous" recombination reaction can be triggered by the presence of a solid surface such as the vehicle wall. Heterogeneous reactions are thought to be promoted on the surfaces of ablating materials, for example, and the net result is that the convective heat load in a "frozen flow" ablation situation is not noticeably different from that to be expected in an equilibrium flow.[5]

On non-ablating surfaces, such as those contemplated for major areas of the shuttle orbiter, laboratory experiments have shown that, given the circumstances of frozen dissociated non-equilibrium, it is possible to realize significant reductions in the heat transfer rate to walls of low catalytic efficiency[6], that is, to surfaces that suppress heterogeneous recombinations of air atoms. Here, catalytic efficiency is defined as the ratio of the number of atoms that recombine on a surface per unit area and per unit time to the total number of atoms that strike the surface per unit area and per unit time. On an inert surface, for which the catalytic efficiency is zero, it has been estimated that heat transfer can be as little as 1/3 of the equilibrium value.[7] On the other hand, it is also possible in such flows to trigger excursions in heat transfer rate above the equilibrium value, as when a frozen flow over a surface of low catalytic efficiency encounters an area of high catalytic efficiency.[8]

The purposes of this note are: (1) To review some of the known effects of surface catalysis on convective heat transfer rate in shock layers, (2) To determine the extent of dissociated flow that might be expected over a shuttle-like configuration, and (3) To estimate the implications of non-equilibrium phenomena on the design of thermal protection system for the shuttle orbiter.

Little to be said is new, and this note represents principally a survey of research information on surface catalytic effects in non-equilibrium flows that might prove of value to shuttle operational capability, survivability, and longevity.

Catalytic Surface Heat Transfer Effects at Stagnation Points

A considerable body of literature, containing results both experimental and theoretical, has been generated during the past decade on the subject of surface catalytic effects upon stagnation-point heat transfer in frozen flows. Although differences of opinion exist as to the exact physical mechanism whereby recombination of dissociated gas atoms occurs at a solid wall,[9,10,11] it is abundantly clear from experiments that surface material and surface condition can have interestingly large effects on heat transfer at a stagnation point.[5,6,12]

One such set of experiments, unpublished data collected in arc-heated, chemically-frozen flows by Lewis A. Anderson of Ames Research Center, is shown on figure 1. This figure is a plot of stagnation-point heat transfer rate parameter against total enthalpy potential, and shows data taken on various stagnation-point calorimeters for the diatomic gases nitrogen and air. The calorimeter surfaces consist either of the material silicon dioxide (actually silicon monoxide is vacuum deposited on a metal surface, but it rapidly oxidizes to silicon dioxide) which has little tendency to promote recombination reactions, or the material nickel, which strongly promotes recombinations. In no case was the calorimeter surface temperature allowed to exceed 1000°K.

Figure 1 shows that the heat transfer to the nearly non-catalytic surface SiO_2 increases with enthalpy at a much lower rate than does that to the catalytically-active nickel surface once the gas dissociation threshold of about 3-4 x 10^3 BTU/lb is exceeded. Over the enthalpy range of shuttle peak heating (5,000 - 12,000 BTU/lb), the nickel surface absorbs heat at a rate two to three times that of the silicon dioxide surface.

Should such effects persist for large distances downstream of the stagnation point, these data suggest that large gains in performance of the shuttle thermal protection system could be realized were the panel system nearly non-catalytic. They also show that these benefits depend sensitively on enthalpy and that meaningful ground-based tests of thermal protection systems designed to have low catalytic efficiency must be carried out in facilities that can operate at enthalpy levels in the range from 8,000 to 14,000 BTU/lb.

Catalytic Surface Heat-Transfer Effects on Afterbodies

An interesting series of experiments was carried out by Sheldahl and Winkler[8] to explore the effects of discontinuities in surface catalytic efficiency on heat transfer that had previously been anticipated on theoretical grounds by Chung, Liu and Mirels.[13] The experimental results, which incidentally illustrate that catalytic effects are not phenomena confined locally to the vicinity of the stagnation point but can occur well downstream, are summarized on figure 2.

The test bodies, hemisphere-tipped cylinders, were fitted either with noses of high catalytic activity (copper) or with noses of low catalytic activity (silicon dioxide). Similarly, interchangeable cylindrical afterbodies of the same two surface materials were provided. Heat-transfer tests, limited to wall temperatures less than 1000°K, were carried out on the cylindrical afterbodies in an arc-heated stream of dissociated nitrogen at stagnation enthalpies that averaged about 12,000 BTU/lb with the results illustrated in figure 2.

Figure 2 shows, first, that with the all-copper body of high surface catalycity, the afterbody heat transfer rate distribution is not far from that which would be predicted for an equilibrium flow about a hemisphere-cylinder. When, however, the nose of silicon dioxide was substituted for the copper nose, the heat transfer rates on the copper afterbody increased by 50%. Note that the afterbody heat transfer rates in all cases are normalized with respect to a (constant) equilibrium value at the stagnation point so that the indicated differences in levels are real. This heat transfer rate increase, due to the recombination of the surplus of atoms carried over the afterbody in the chemically inert flow from the nose, was expected, on theoretical grounds,[13] to die out towards the equilibrium value a few body radii downstream of the discontinuity, but the experimental evidence[8] indicates that the region of augmented heat transfer rates persists farther downstream than theory would predict. These data suggest that, in frozen boundary layers, discontinuities in surface catalycity from low to high values can lead to higher local heating loads downstream than might be expected on the basis of equilibrium estimates.

When the afterbody of silicon dioxide was employed behind a nose material of either Cu or SiO_2, the afterbody heat transfer rates were consistently some 60% lower than the equilibrium rates, as illustrated in figure 2. Actually, the test with the catalytic (copper) nose shows a slightly lower heat transfer rate distribution than that with the non-catalytic (silicon dioxide) nose, but the decrease is small. This experiment shows that a silicon dioxide surface is highly effective in suppressing atom recombination under a frozen, laminar boundary layer and suggests that non-catalytic surfaces are of potential importance in reducing heating loads to the shuttle orbiter.

Shuttle Orbiter Environment

The shuttle orbiter as presently conceived is an airplane-like configuration such as is illustrated on figure 3. It is designed to return to earth through the entry corridor shown on figure 4. During the portion of its entry where the heat transfer rates, and thus the surface equilibrium temperatures, are the highest (i.e., the altitude range between 270,000 ft. and 175,000 ft., on figure 4), the free stream Mach number, M_∞ (figure 3), is estimated to range from 24 down to about 18. Angles of attack, α, can vary from about 60° to about 20°, depending upon the entry trajectory to be flown. During the course of the entry, the local Mach numbers adjacent to the lower surface, M_L, can vary from 3 to 15, and the local Mach numbers adjacent to the upper surface, M_u, can vary from perhaps 30 to 15. It is these local Mach number conditions; along with the corresponding local flow history; the local state conditions of pressure and temperature; transport properties of viscosity, thermal conductivity, and diffusion coefficient; and the rate processes of dissociation and recombination; that determine whether or not the boundary layers are out of chemical equilibrium and thus can respond to surface catalytic effects. Obviously, a very wide spectrum of local conditions can be expected.

The intent here is not to evaluate the specific states of local flow non-equilibrium for a specific shuttle orbiter configuration such as that shown in figure 3 throughout its entry profile, nor will an attempt be made to determine which of many alternative entry profiles will yield the most advantageous conditions of dissociation for permitting surface temperature reductions over catalytically inert walls. Rather, the point of view will be taken that specific examples for simple steady flow fields can serve to illustrate, at least qualitatively, the effects of surface catalycity on heat transfer rate with the anticipation that these phenomena will have application in alleviating thermal loads on the entering shuttle orbiter. In keeping with this intent the complicated flow field over the shuttle will be idealized to that over a slightly blunted, two dimensional flat plate at arbitrary angle of attack.

It is obvious that the flow over a blunt flat plate at sufficiently high angle of attack can be likened directly to the shock-layer - dominated flows alluded to in discussion of figures 1 and 2. At low-to-moderate angles of attack the situation is not as clear. Certainly, very near the nose, the shock layer concept will hold and the flow will be dissociated and frozen. Well downstream of the leading edge, however, the local flow field will be increasingly encroached upon by an outer flow that has passed through a progressively weaker oblique shock wave that might not produce dissociation. Thus, the embedded, dissociated flow generated by the shock wave at the nose can be expected to become diluted by the adjacent, cold undissociated gas. This is not to say, however, that dissociation and recombination reactions are not important for flight at low angles of attack. Rather they will now occur mainly in the high temperature regions within the boundary layer. This situation is illustrated in the two parts of figure 5.

Figure 5a is a map of the equilibrium, dissociated mass fraction to be expected behind the shock wave on a sharp flat plate at various angles of attack and flight velocities for altitudes between 200,000 and 250,000 ft. The figure shows that the shock wave will dissociate 30% or more of the air at velocities above 20,000 ft/sec. and angles of attack greater than 45°. However, interestingly large dissociation fractions ($\approx$ 20%, corresponding to an oxygen population of atoms) are not to be expected in the shock layer at the velocity of peak heat transfer rate at angles of attack less than about 34°.

The dissociation that might be expected to occur as a result of viscous heating in the boundary layer can be estimated in the following manner. For flow over a cold flat plate at low angles of attack, where the edge enthalpy, h_e, is small compared to the flow enthalpy, $V_e^2/2$, shear work will predominate over diffusion and conduction in determining the boundary layer temperature profile. Thus, assuming negligible longitudinal velocity gradients and enthalpy gradients, the boundary layer velocity and enthalpy profiles will be related by Crocco's integral, provided that Lewis number and Prandtl number are both set equal to unity.[14] With the Crocco relation, the peak boundary layer enthalpy can be found for a fixed wall enthalpy, angle of attack, and altitude and for a range of flight velocities with the aid of tabulated state and flow parameters for equilibrium air.[15,16] The equilibrium dissociated fractions corresponding to these calculated enthalpies are related to flight velocity and angle of attack as shown in figure 5b.

Figure 5b shows the dissociation fraction that can be generated by shear work in the high enthalpy region of the boundary layer after sufficient boundary layer run for the dissociation reaction to reach equilibrium. Here one sees that complete oxygen dissociation is expected down to angles of attack of 20° at the velocity of peak heating. The length of boundary layer run required to attain the plotted dissociation levels depends mainly upon the dissociation rate coefficient, which is in turn highly dependent on boundary layer density and temperature. To accurately determine the dissociation fraction as a function of length of run, rather complicated calculations are necessary for each flight condition.[17,18,19]

Calculations suggest, however, that dissociation relaxation will go to completion after from 10 to 60 feet of boundary layer run at shuttle flight conditions near peak heating, depending upon the altitude.[19] It also should be pointed out that the calculated magnitude of the dissociated fraction that can be generated in the boundary layer is very sensitive to the assumed values of Lewis number and Prandtl number.[18] A precise calculation of dissociation fraction is thought to require numerical techniques in which these parameters can be treated as variables within the boundary layer.[18]

Chemical State of Boundary Layer Along Entry Corridor

Figure 6 is an altitude-velocity plot that summarizes the three domains where the boundary layer in the local flow generated by a sufficiently strong shock wave can be characterized variously as being either chemically frozen, chemically reacting, or in chemical equilibrium.[19] The boundaries are admittedly fuzzy and depend upon flight attitude and the portion of the surface one chooses to examine. The boundaries shown correspond to flight at angles of attack above about 30° and to areas on the lower surface from the leading edges to between 10 and 50 feet downstream. The boundaries were estimated independently by Mr. Warren Winovich and Dr. Chul Park of Ames Research Center, and are adaptations and extensions of estimates reported previously.[7,20]

At altitudes above about 275,000 ft. dissociation of the free stream by the shock wave proceeds so slowly that it may not go to completion for many hundreds of feet downstream. Also, if the shock wave structure is so weak that dissociation temperatures are not reached in the shock layer as when flying at low angles of attack where dissociation takes place in the boundary layer, obviously the zone of reacting boundary layer can engulf the entry corridor.

Effects of Surface Catalytic Efficiency on Radiation

Equilibrium Temperature

To evaluate the possible benefits of surface catalytic effects to the shuttle orbiter, radiation equilibrium temperature distributions along a flat plate at angle of attack in a frozen hypersonic air flow were calculated for several (constant) values of surface catalytic efficiency, even though it is known that catalytic activity is a function of temperature.[21]

The computations, carried out by Dr. Chul Park of Ames Research Center, incorporate the assumptions of equilibrium dissociation through an oblique shock wave,[15] frozen inviscid local flow, laminar boundary layer with diffusion of species, and constant surface emissivity of 0.6. One of the results is shown on figure 7 for a free-stream velocity of 22,000 ft/sec., and altitude of 240,000 ft., an angle of attack of 55°, and various values of catalytic efficiency.

Four curves of wall temperature versus length Reynolds number are shown on figure 7. The upper one corresponds to a fully catalytic surface, and represents the case where all air atoms that strike the surface are recombined into molecules. For the next lower curve, the catalytic efficiency is assumed to be 10^{-3}, a value representative of a metal oxide such as Al_2O_3 that is impervious to atomic oxygen and nitrogen and one notes that now the surface temperature is consistently reduced some 50°C (90°F) below the equilibrium value. When the catalytic efficiency is further reduced to 10^{-4}, which means that only one recombination occurs per 10,000 atom impacts, the radiation equilibrium temperature is lowered an additional 80°C for a total reduction of about 130°C (234°F). A value of catalytic efficiency as low as 10^{-4} is probably within practical reach, since glassy materials such as pyrex, vycor, and quartz are reported to exhibit values near 10^{-4} at temperatures up to 700°K.[21] Materials with significantly lower catalytic efficiencies than 10^{-4} are not known, apparently. Happily, as figure 7 further shows, little additional reduction in surface temperature would appear possible at this flight condition even with a material having an inert surface.

Although this result applies to a laminar boundary layer, the same incremental surface temperature reduction is expected in a turbulent layer, although the absolute temperature would, of course, be higher.

ATOMIC OXYGEN ATTACK ON METALS

One of the hazards of flying within a cloud of oxygen atoms is a possible increase in oxidation rates of metals as compared to the rates in molecular oxygen. Unfortunately, little work has been done on this phenomenon at the background pressures and temperatures characteristic of shuttle entries, nor has the effect of growth of the oxide layer been taken into account, except for one instance wherein oxide formation on a nickel-chromium alloy under attack of oxygen molecules has been treated as an ablation process.[22] Results from a set of experiments to study attack of atomic oxygen on a refractory metal are shown on figure 8.

Figure 8 shows the oxidation probabilities measured by Rosner and Allendorf[23] for molybdenum when attacked either by atomic oxygen or by molecular oxygen. At surface temperatures expected for the major portion of the shuttle orbiter [$\leq$ 1300°K (1900°F)], one sees that attack by atomic oxygen is 1-1/2 orders of magnitude more likely than attack by molecular oxygen. Although data for nickel, chromium, columbium, etc. are not available, it is probable that a similar state of affairs will exist for these elements. Thus, it may prove mandatory to provide inert coatings to metallic portions of the shuttle skin to prevent its rapid deterioration by atomic oxygen. If such a coating were also non-catalytic, the heat load due to atom recombination could also be suppressed.

Concluding Remarks

An attempt has been made to review some of the known effects and consequences of non-equilibrium flow phenomena that can be expected to be encountered by the entering shuttle orbiter. It has been concluded that the oxygen in the atmosphere will be largely dissociated over most of the vehicle throughout the high-speed portion of its entry corridor. The possible advantages that will accrue to the thermal protection system if atomic recombination on the surface could be suppressed were outlined, and examples were given of effects of surface catalytic efficiency on heat transfer rate and surface temperature. Finally, an example of the grossly increased oxidation probability for metals attacked by atomic oxygen, as opposed to attack by molecular oxygen, was discussed.

Much has been left unsaid in this necessarily brief survey, and many questions remain to be answered on the subject of non-equilibrium flows. For example, the whole problem of non-equilibrium chemistry in a turbulent boundary layer remains to be solved, although qualitatively the same kinds of phenomena are expected as occur in laminar layers. Nor has anything been said about non-equilibrium effects in regions of separated flow such as will exist over the upper surfaces of the shuttle. Although much work has been done on the mechanism of surface catalysis, it has all been aimed toward increasing catalytic activity. Very little is known about the subject of "non-catalysis", and what property is required of the lattice structure of a material surface to render it inert to attack by foreign atoms and molecules.

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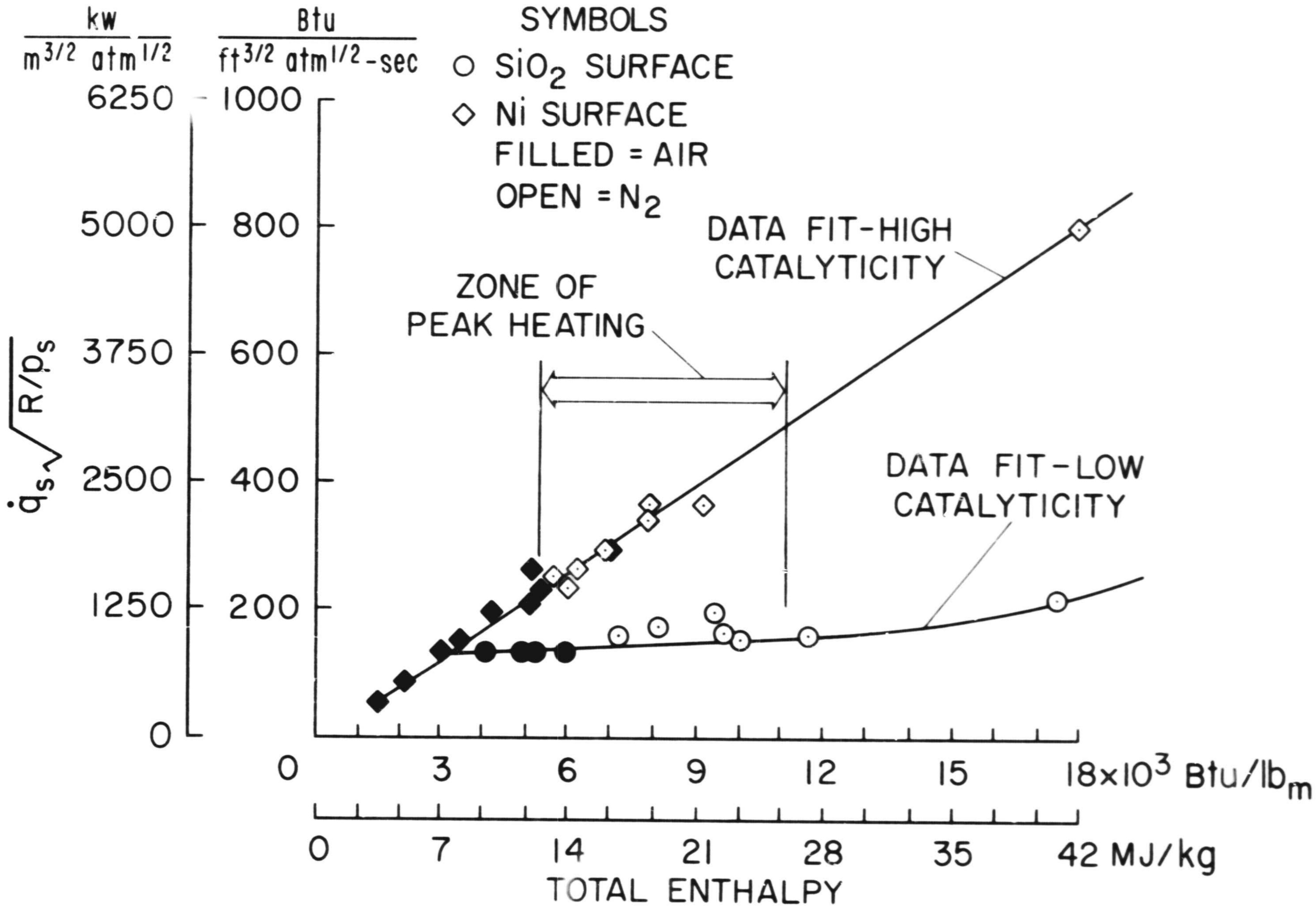


Figure 1.- Effect of surface catalytic activity on heat transfer rate, frozen boundary layer. $T_W \leq 1000°K$.

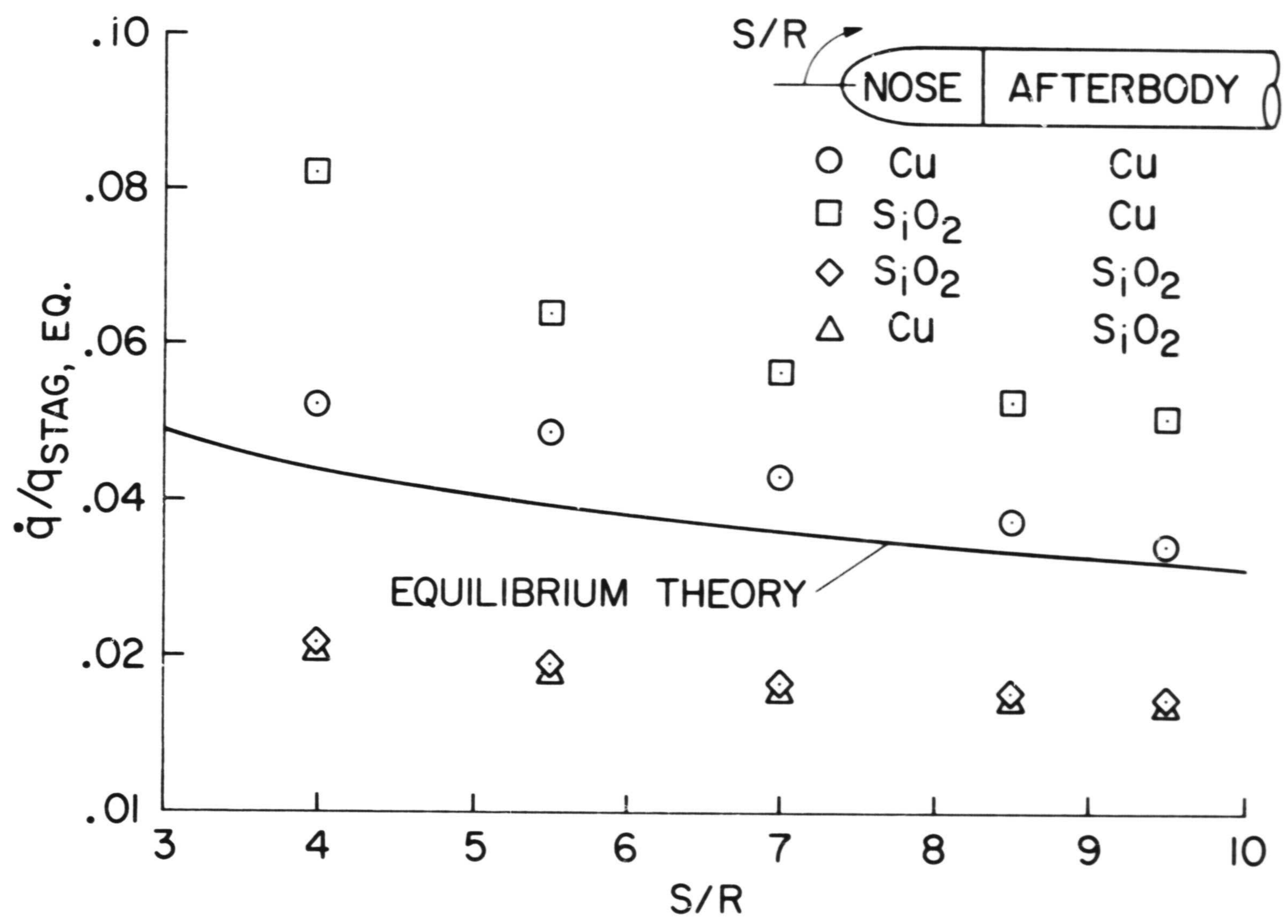


Figure 2.- Normalized afterbody heat transfer rates to bodies with various catalytic discontinuities. Total enthalpy = 12,000 BTU/lb.

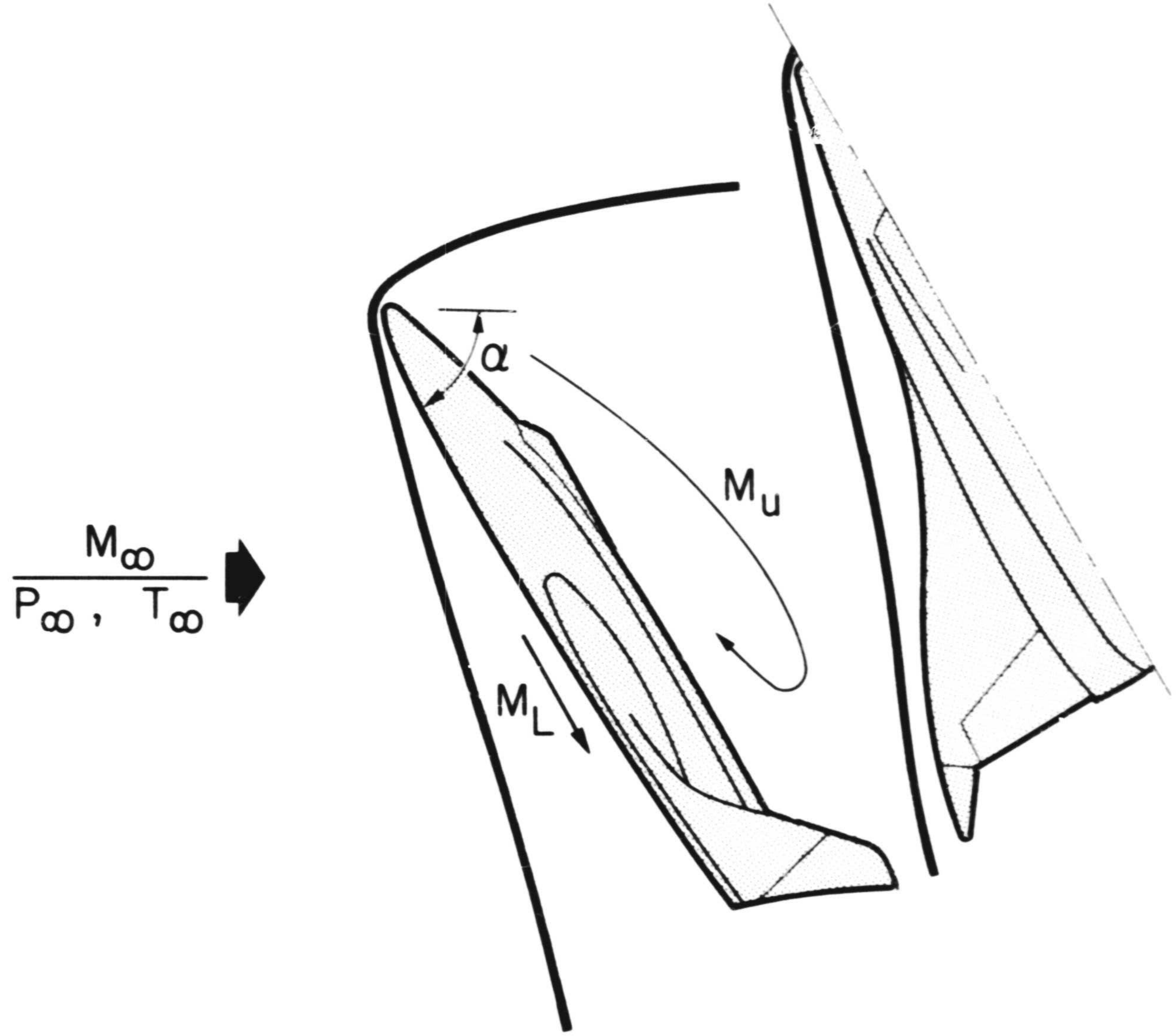


Figure 3.- A shuttle orbiter configuration.

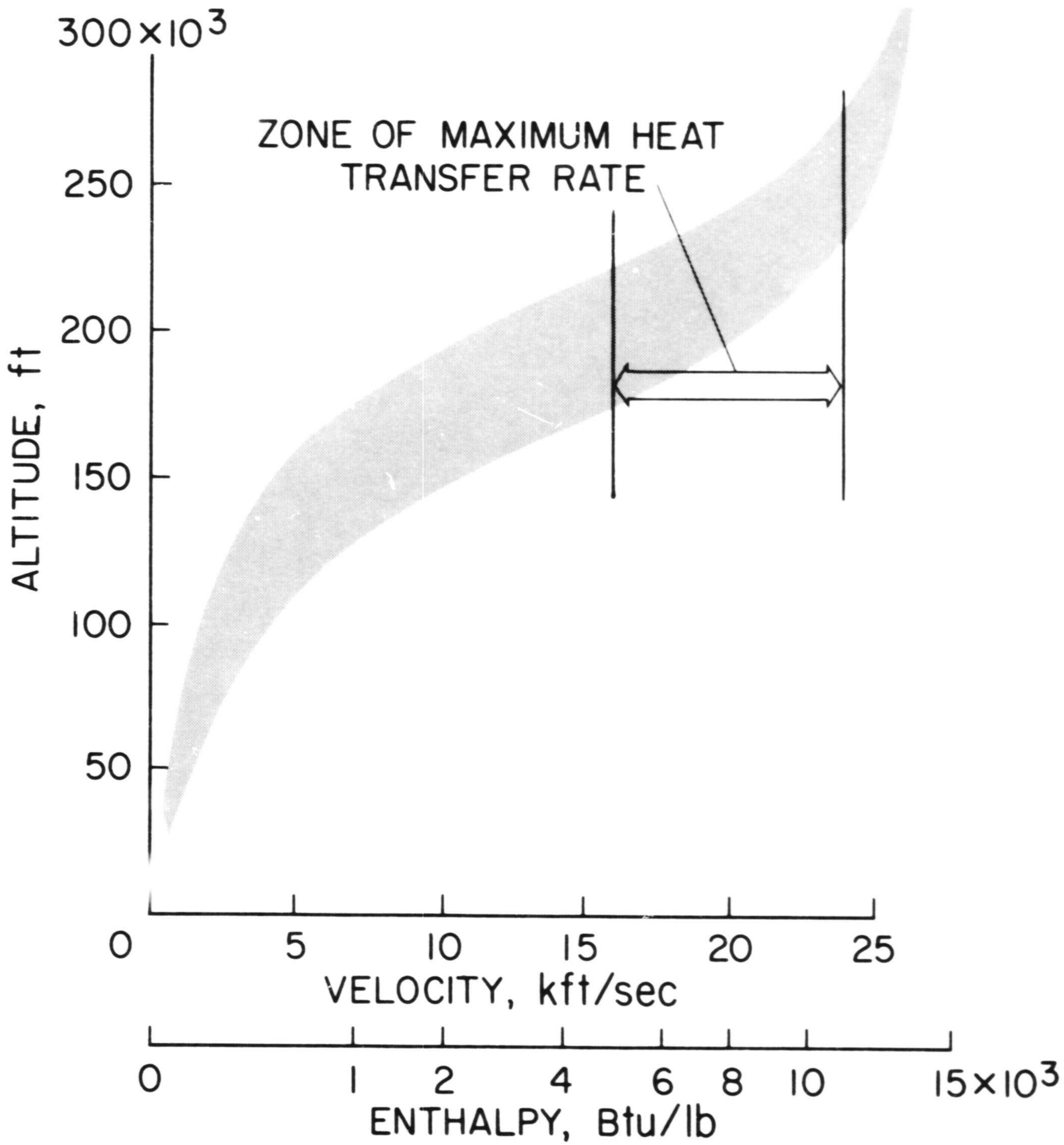
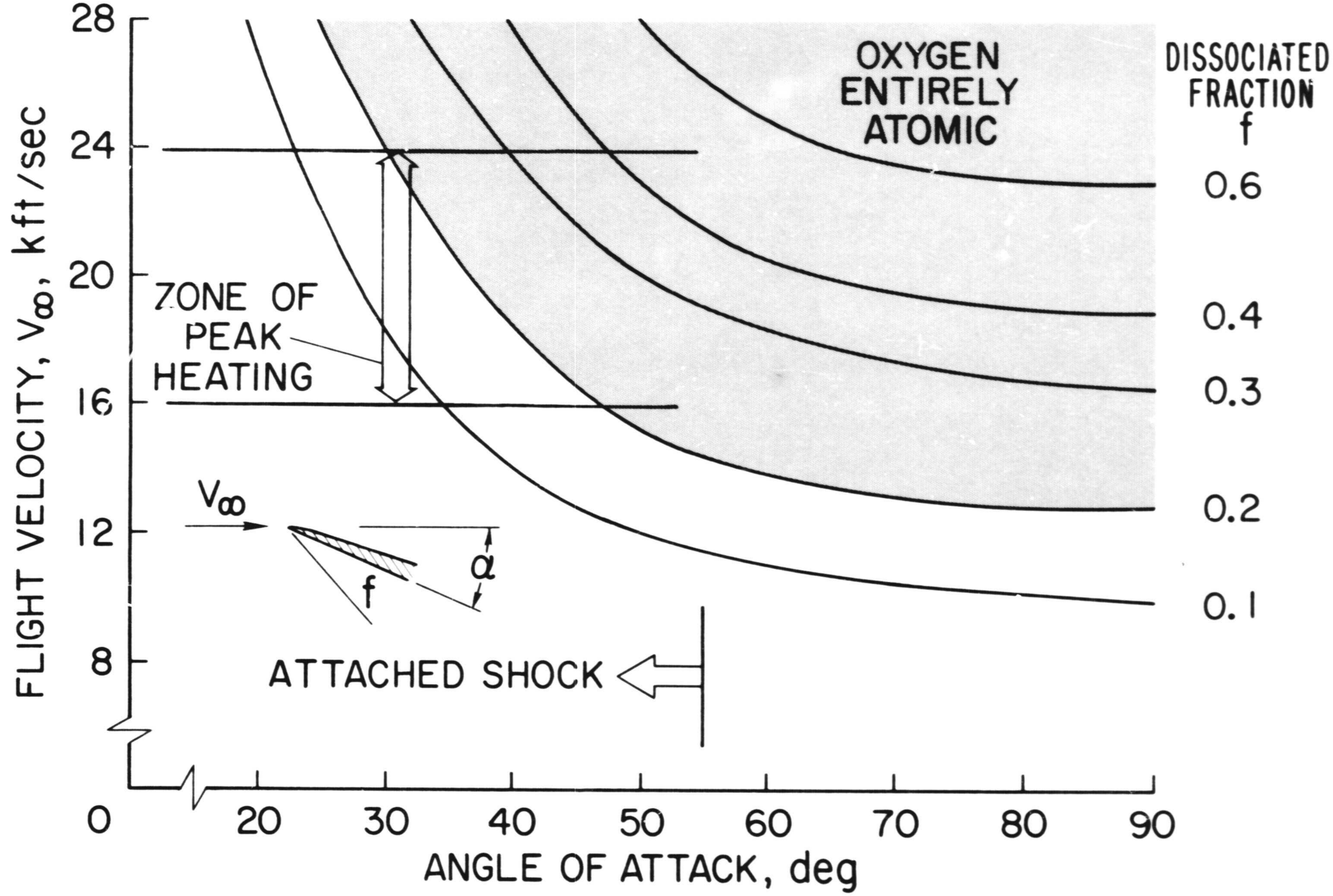
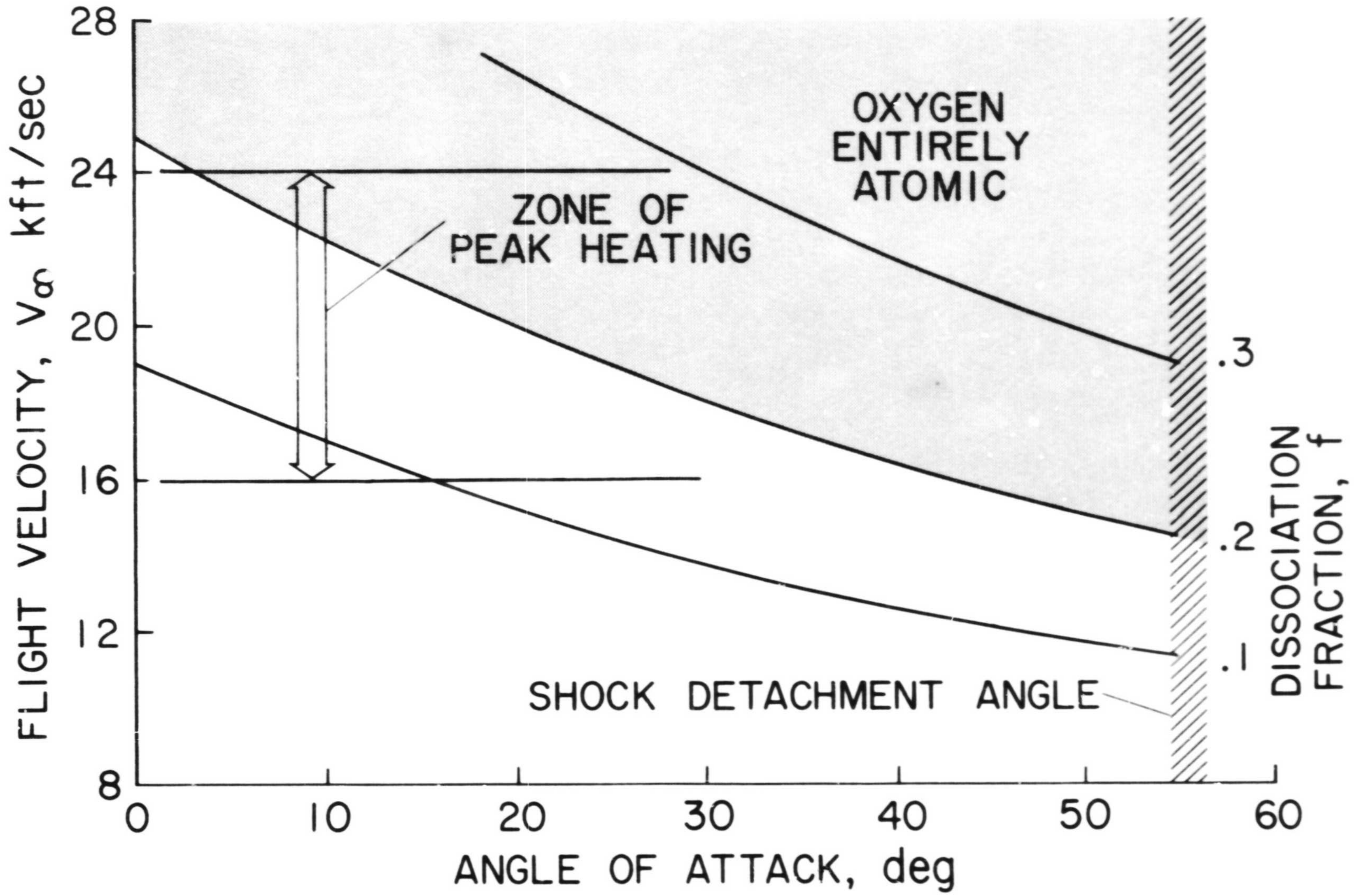


Figure 4.- Entry corridor for shuttle orbiter.



(a) Dissociation produced by bow shockwave.

Figure 5.- Effect of flight velocity and angle of attack on equilibrium dissociation at altitudes from 200,000 to 250,000 feet.



(b) Dissociation produced by boundary layer.

Figure 5.- Concluded.

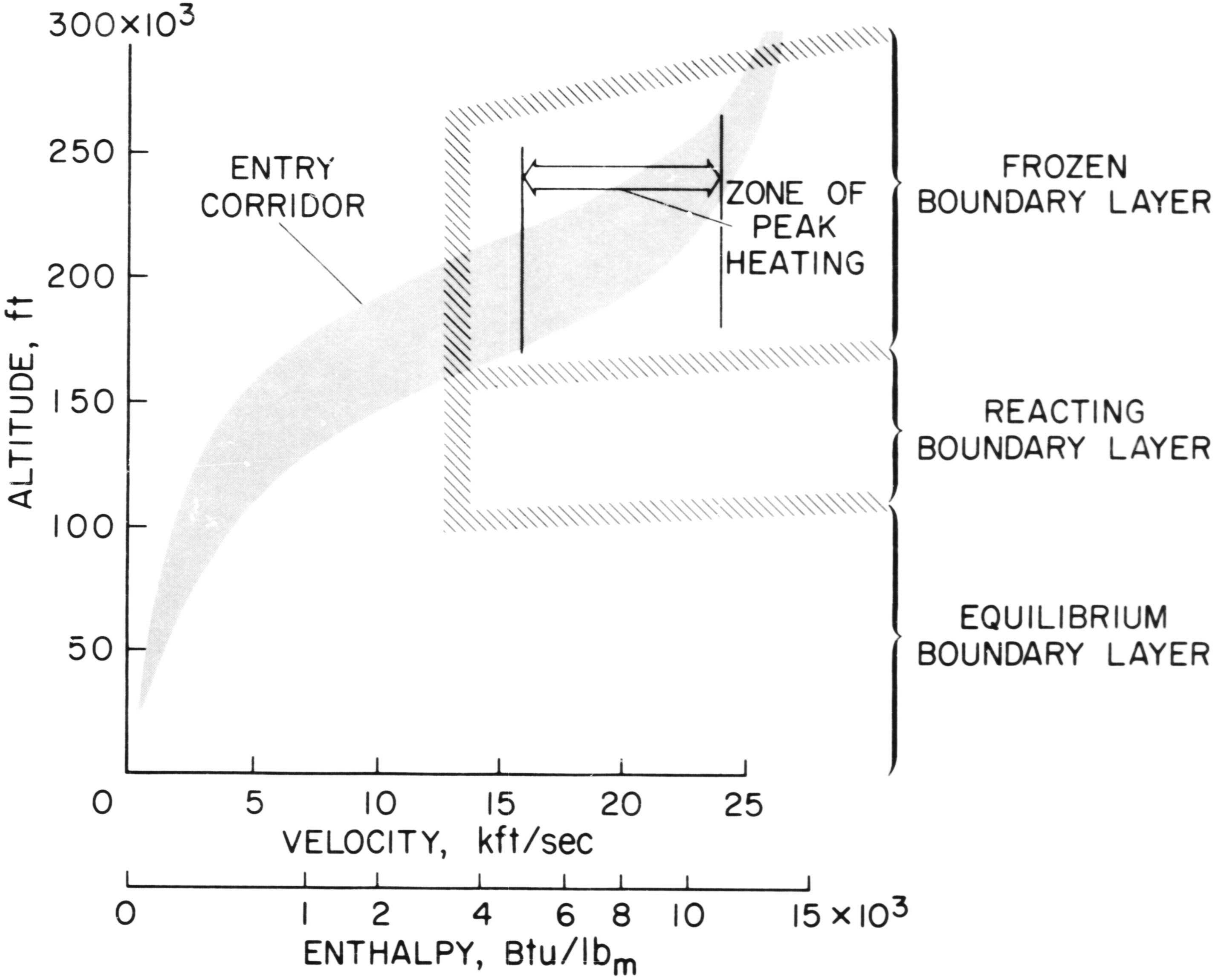


Figure 6.- Chemical state of boundary layer along shuttle orbiter entry corridor. $\alpha \geq 30°$.

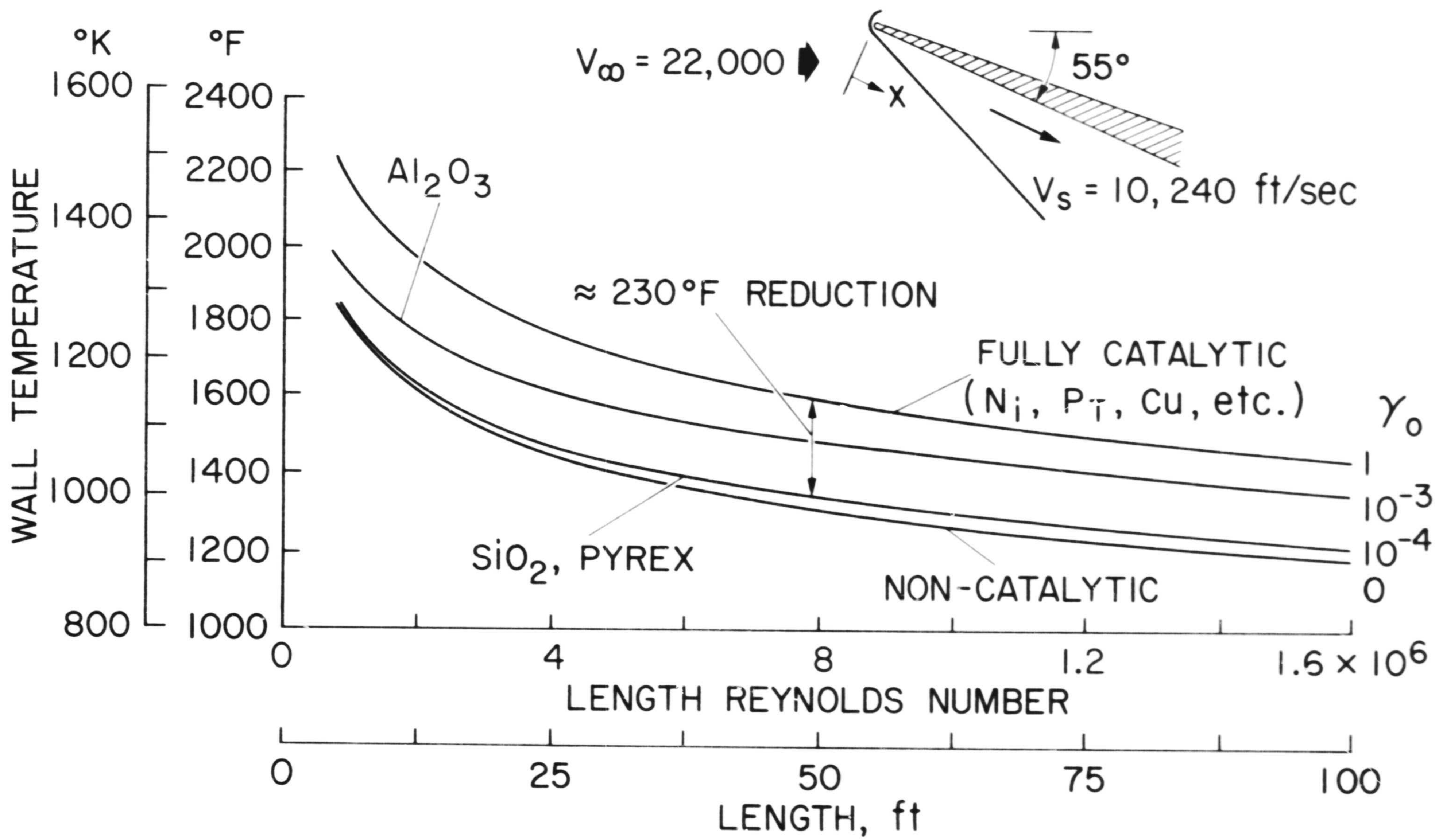


Figure 7.- Effect of surface catalytic efficiency on temperature of radiation-cooled flat surface, altitude = 240,000 feet, laminar flow.

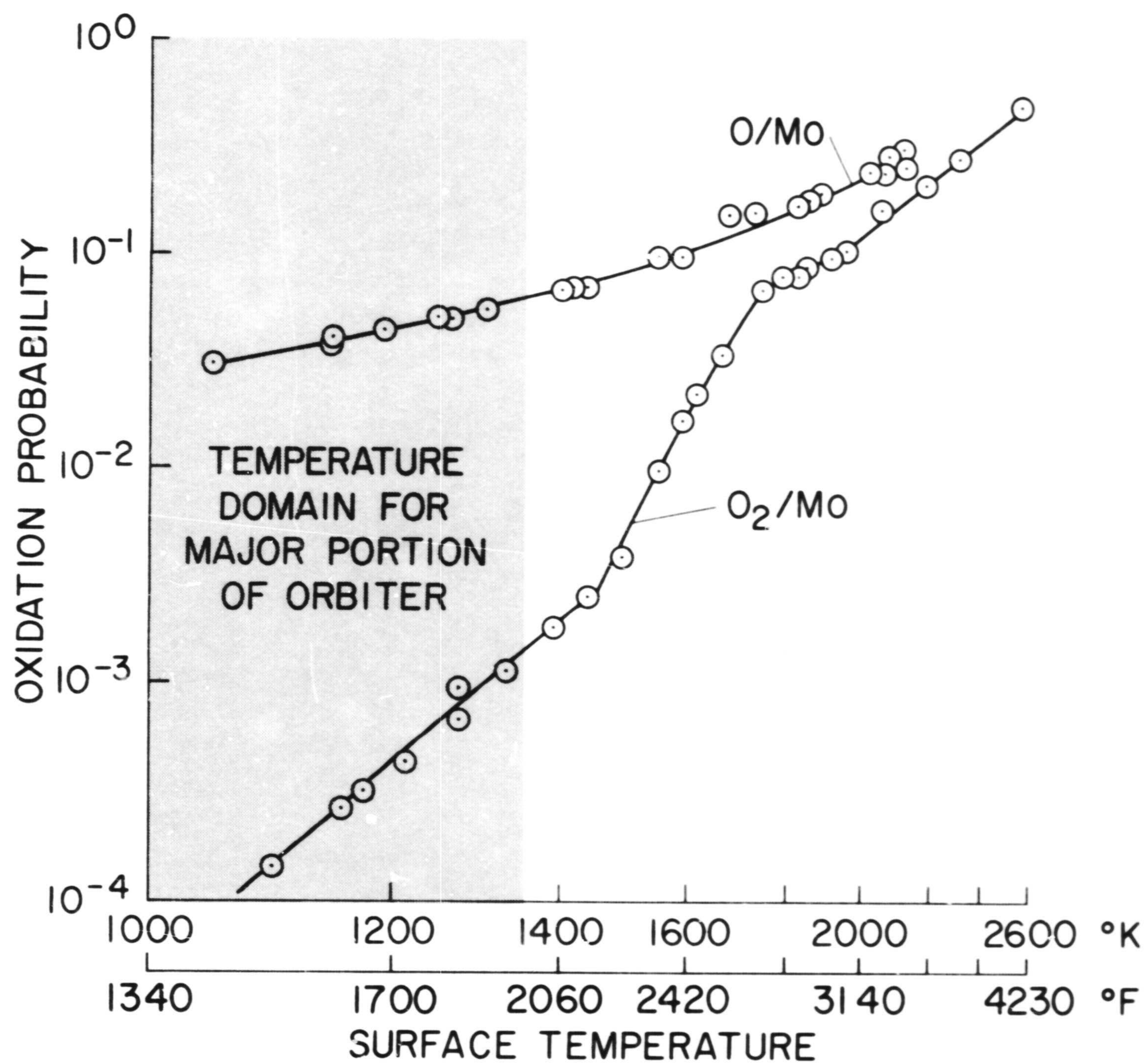


Figure 8.- Oxidation probabilities for attack of molybdenum by atomic and molecular oxygen.