



Diffusion effects on the determination of surface catalysis in Inductively Coupled Plasma facility



Alan Viladegut*, Ümran Düzel, Olivier Chazot

von Karman Institute for Fluid Dynamics, Chaussée de Waterloo 72, B1640 Rhode-Saint-Genèse, Belgium

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ABSTRACT

Atomic recombination is an important process to consider when computing the heat flux transferred to the wall of a re-entry vehicle. Two chemical processes are influencing the species diffusion in the boundary layer surrounding a re-usable Thermal Protection System: gas phase reactions and catalytic recombination at the surface. The coupling between them is not normally taken into account when determining the catalytic recombination coefficient (γ) in plasma facilities. This work aims to provide evidence of such coupling based on both a theoretical analysis and an experimental campaign in the VKI-Plasmatron facility. Recombination coefficient measurements at off-stagnation point configuration on a linear copper calorimeter are provided. An evolution from a high-catalytic to a low-catalytic condition due to the boundary layer growth along the probe is observed. This result is consistent with a parametric analysis carried out using the in-house non-equilibrium boundary layer solver, which shows how the experimentally determined catalysis could be influenced by the amount of gas-phase recombination inside the boundary layer.

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1. Introduction

The re-entry of a space vehicle into the atmosphere normally takes a few minutes. Although this is a very short period of time if compared to the total time invested for the whole mission, the external design of the vehicle is a direct consequence of the type of re-entry and all the physics occurring on its surrounding. Typical re-entry velocities on Earth range between 7 and 11 km/s at the upper atmospheric layers. At these speeds, the kinetic energy of the gas is stored as internal energy when particles cross the shock wave that appears in front of the spacecraft, rising the gas temperature to thousands of Kelvins and dissociating the molecules into atomic and ionized species. This effect turns a high-speed situation into a high-temperature problem with chemical reactions. Therefore, re-entry space vehicles must be equipped with Thermal Protection Systems (TPS) capable to withstand such an extreme environment. Thus, the development and validation of accurate physical and chemical models describing the high-temperature phenomena is of major concern for both integrity of the payload and safety of the mission.

This work is focused on the diffusive effects confined inside the boundary layer that is growing along the body and, more particularly, on the mass diffusion of species towards or from the wall. Such diffusion is strongly interacting with both the chemical reactions occurring in the gas and the catalytically enhanced atomic recombination on the surface of the vehicle. The reason of being for this paper lies on the fact that there is an energy transfer from the gas to the wall caused by their chemical interaction. This is known as the gas-surface interaction phenomena and its modeling is required to quantify the heat flux during the design of the TPS.

The heat flux transferred by diffusion is defined in Eq. (1), where \vec{J}_{iw} and $h_{i,w}$ are both the diffusion flux and enthalpy of species i . It is only driven by the wall catalytic properties on reusable TPS through the boundary condition of the species conservation equation defined in Eq. (2).

$$\vec{q}_d = \sum_i^{N_s} \vec{J}_{iw} h_{i,w} \quad (1)$$

$$\vec{J}_{iw} \cdot \vec{n} = k_{i,cat} \rho_w c_{i,w} = \rho_w D_i \left. \frac{\partial c_i}{\partial \eta} \right|_w \quad (2)$$

This boundary condition is a simplification that only applies for binary mixtures and first order reactions, with ρ_w being the gas density at the wall, c_i and D_i are the mass fraction and mass

* Corresponding author.

E-mail addresses: alan.viladegut@vki.ac.be (A. Viladegut), umran.duzel@vki.ac.be (Ü. Düzel), olivier.chazot@vki.ac.be (O. Chazot).

diffusion coefficient of species i respectively, and η is the coordinate perpendicular to the wall. This expression is used here as an example to illustrate that the species diffusion velocity at the wall $(D_i/c_i(\partial c_i/\partial \eta))_w$ is imposed to match the speed of the catalytic reaction, characterized by the reaction rate constant $k_{i,cat}$. Therefore, the species production/depletion rates due to catalytic reactions is a balance between species diffusing to/from the surface and the chemical kinetic properties of TPS materials. For mixtures with more than two species, other diffusion models are available in literature [1,2].

The catalytic activity taking place at the wall is usually quantified by the so-called recombination coefficient γ (also known as recombination probability or catalyticity). Its experimental determination in high-temperature environments is critical for model validation and to improve TPS designs. The recombination coefficient was introduced for aerospace applications by Goulard [3] in 1958. He defined γ as the ratio between the atomic flux reacting at the surface and the atomic flux impinging at the surface, leading to Eq. (3); where k_B is the Boltzmann's constant, T_w the temperature at the surface and m_i the mass of particle i . Thus, the condition $\gamma \rightarrow 0$ is known as the non-catalytic wall and, conversely, $\gamma \rightarrow 1$ as the full-catalytic wall. Goulard's main contribution in the analysis of chemically reacting boundary layers was the possibility to introduce the partly catalytic boundary conditions at the wall in a decade in which either the under-predicting non-catalytic or the over-predicting full-catalytic boundary conditions were in use [4,5].

$$\gamma_i = k_{i,cat} \sqrt{\frac{2\pi m_i}{k_B T_w}} \quad (3)$$

The experimental determination of the recombination coefficient is suitable in binary mixtures with no gas-phase atomic recombination (frozen flow). Indeed, these ideal testing conditions are obtained by Balat-Pichelin [6–8], Driver [9,10] and Fletcher [11] due to the low pressure environments provided by their diffusion reactors (only a few pascals). However, the measurement of γ in larger plasma facilities is also of interest because relevant flight conditions need to be simulated. Since the peak heating conditions are reached under not so low pressure conditions, plasma wind tunnels such as the Plasmatron in the von Karman Institute (VKI) are designed to operate at pressures higher than the ones found in diffusion reactors, meaning that the presence of gas phase atomic recombination is likely to occur and frozen flow conditions might not be present during the tests. These reacting boundary layer conditions are inherent to larger facilities dedicated to TPS catalysis determination, and its comprehension is of major concern for both the ground-to-flight extrapolation activities being developed at VKI and the validation of gas-surface interaction models.

The problem of catalytic characterization under stagnant flow conditions is introduced by the Institute for Problems in Mechanics of Russian Academy of Sciences (IPM RAS) using the IPG-4 RF-Plasmatron [12]. The fact that surface kinetics can not be measured with one dedicated instrument makes the experimental determination of the recombination coefficient to rely on a combination of both heat flux measurements and CFD simulations that accurately reproduce the thermochemical environment in front of the TPS material sample. This means that the experimental determination of the catalytic recombination coefficient could appear influenced by both the diffusion and the chemical models implemented in the solver, leading to a γ parameter that might not behave consistently with the theoretical analysis proposed by Goulard [3]. Such “falsification” on the experimental measurements could be caused by the inaccuracy on both the chemical-diffusion models acting in the gas and the catalytic model at the wall of the sample. The flight extrapolation of the stagnant non-equilib-

rium boundary layer in the framework of the Local Heat Transfer Simulation concept [13,14] is required to properly predict on ground the recombination coefficient under flight relevant operating conditions. However, such relation between catalysis and flight is often disregarded when providing a quantification of γ in a plasma wind tunnel.

The diffusion effects influencing the determination of γ were already proposed in the past [15]. An experimental method for the recombination coefficient determination under dissociated nitrogen is presented by Vasilevskii [16], who determined catalytic values of beryllium under different chemical non-equilibrium regimes taking molybdenum and platinum as the non-catalytic and full-catalytic standards, respectively. Numerical heat flux predictions under pressures ranging from 0.1 to 0.9 atm and imposing different catalytic conditions assessed the sensitivity reduction of the heat flux with respect to catalysis from the almost frozen to the near equilibrium regimes. A constant value of $\gamma_{Be} = 0.01$ is reported inside this pressure range.

The purpose of this paper is to demonstrate that the different diffusion regimes, ruled by both volume and surface atomic recombination, can influence the experimentally determined recombination coefficient under the chemical non-equilibrium environment provided by the VKI-Plasmatron facility, making the reaction rate constant at the catalytic surface apparently linked with the gas-phase reactions. Results of catalytic recombination coefficient measurements on water-cooled copper calorimeters are shown at both stagnation and off-stagnation point configurations, showing inconsistencies with the γ -model implemented in CFD. The main advantage of testing with copper calorimeters is that surface temperature can be maintained constant at 350 K for any testing condition. The logic behind the experiment lies on the fact that, changing the probe geometry while keeping the same pressure and surface temperature conditions, one can isolate the diffusion effects and assess how they affect the determination of the recombination coefficient. The work is complemented with a parametric analysis using the in-house developed numerical tools dedicated to the reconstruction of the flow environment in the Plasmatron and from which catalysis is determined under stagnant flow configurations. Such analysis aims at understanding how both the gas-phase chemical regime and the surface catalytic activity in a non-equilibrium boundary layer influence the heat flux transferred to a cold wall.

The particular aspect of the paper lies on the fact that it shows experiments intended to isolate the diffusion phenomena during the quantification process of the recombination coefficient under both stagnation and off-stagnation point configurations in a plasma wind tunnel. This is done by keeping wall temperature and pressure constant for each test condition, and modifying the diffusion characteristics by either changing the probe geometry on a stagnant flow or looking at the evolution of the boundary layer along a flat plate. It also contributes on understanding the possible application limits of current γ -models widely implemented in CFD softwares through the combination of both heat flux measurements and numerical predictions.

2. Catalysis determination in non-equilibrium flows

To illustrate the problem regarding catalytic measurements under non-equilibrium boundary layers, it is worth to consider the work of Rosner [17], in which Broadwell's conductivity cell model [18] is generalized to include first order atom recombination at a cold surface of arbitrary catalytic activity. The flow within two parallel plates separated by a distance δ and enclosing a partially dissociated gas is analyzed. One of the plates is hot and under Local Thermodynamic Equilibrium (LTE) conditions, while the

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