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극초음속 유동에서의 표면 촉매 재결합: 수치해석적 기법 리뷰

Surface Catalytic Recombination in Hypersonic Flow: A Review of the Numerical Methods

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Abstract
This paper provides a general overview of surface catalytic recombination in hypersonic flow. The surface catalytic recombination phenomena is elaborated in terms of its general overview and numerical modeling associated with it. The general overview of the surface catalytic recombination phenomena describes the elementary surface reactions for the surface catalytic and the role of the surface catalytic recombination efficiency in the heat transfer determination. In the numerical modeling, the surface catalytic recombination is described based on the stagnation-point boundary layer analysis, and finite-rate surface reaction modeling. Throughout this overview manuscript, a general understanding of this phenomena is obtained and can be used as foundation for deeper application with the numerical computational fluid dynamics (CFD) flow solver to estimate the surface heat transfer in the hypersonic vehicles.

Keywords: Hypersonic Flow · Surface Catalytic · Heat Transfer · Numerical Methods

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1. Introduction

Measuring the total heat transferred that is experienced by a hypersonic vehicle is crucial during the design process of thermal protection material. The total heat transfer can be estimated from the previous flight data[1 - 3]. This approach is performed by measuring the total heat transfer in the hypersonic boundary layer during the real flight condition. However, this approach is difficult to be implemented, and not much data are available in the open literature.

As an alternative approach to measure the total heat transfer, an experiment conducted in hypersonic ground facilities is applied. This method, from many decades ago, has been conducted by many researchers to measure the heat transfer. One of the interested phenomena to study in that hypersonic ground facility experimental approach is the catalytic recombination phenomenon. The understanding of this phenomenon is essential since it has been indicated that the catalytic recombination provokes an additional heat flux around 30% of the total heat transfer for Earth re-entry case[4].

As the catalytic recombination measurement is concerned, one of the final aims is to measure the catalytic recombination efficient, which is defined as the ratio of the recombined atomic flux to the impinging atomic flux. The experimental measurements have been conducted in various ground facilities: side-arm[5], arc-jet[6-8], shock tube[9-11], solar radiation plasma generator[11].

One of the popular methods in the experimentally measurement of the catalytic recombination is by applying the local heat transfer approach. This approach requires a theory that predicts the total heat transfer due to the catalytic recombination. By comparing the measured heat transfer and theoretical heat transfer, the corresponding catalytic recombination efficiency can be estimated.

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Although a number of experimental studies that have been conducted, there is a scattering among the measured recombination efficiency values. To address the data scattering found in the catalytic efficiency values, a number of numerical methods have becomes another essential element in the study of the catalytic recombination phenomenon.

To name a few, surface kinetic modeling, molecular dynamics (MD) simulation, kinetic Monte Carlo (KMC) method have been rigorously studied by many researchers. These aforementioned modeling studies aim for not only resolving the scattering observed in the experimental measured data, but also getting a better physical understanding of the catalytic recombination phenomenon on the surface. However, despite the variation of the numerical methods, there is still lack of study that explains the relation among them and their comparison with the experimental approach.

In this review paper, the numerical approaches to compute the catalytic recombination efficiency are elaborated to provide an up-to-date comprehension about the catalytic recombination phenomena. The discussion will start from the fundamental aspects of the catalytic recombination phenomenon and continue with the appropriate numerical modeling. In the numerical modeling discussion, we will also include a review of the theoretical heat transfer theory that is mainly used in the local heat transfer approach in the experimental measurement.

2. Catalytic Recombination Phenomena

Like any other chemical phenomena, the catalytic recombination phenomenon is associated with some elementary steps. The elementary steps associated with the are listed as[12]: atomic diffusion to the surface, adsorption at the surface, chemical reaction at the surface, desorption from the surface, and diffusion away from the surface. The schematic of this elementary reaction is shown in depicted in Fig. 1. Besides the diffusion reaction, the remaining elementary steps are considered as the dominant reactions. Therefore, in many studies of the catalytic recombination phenomenon, much attention is given to model the adsorption, the surface reaction, and the desorption process.

The adsorption process is defined as the process in which the atoms becoming adhered into the surface. Based on type of bonding, this type of process can be classified into: physisorption and chemisorption. The physisorption process occurs due to the van der Waals force betweent the atoms and the surface, therefore it has lesser activation energy.

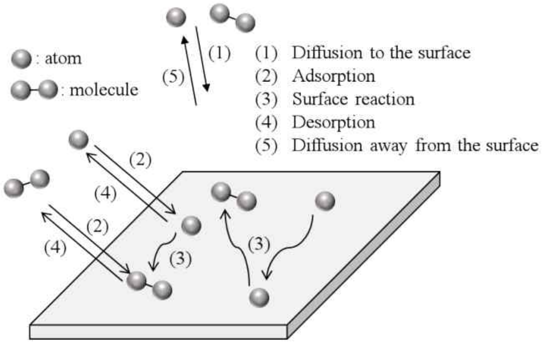

Fig. 1 Elementary steps during the catalytic recombination phenomena

The chemisorption process, on the other hand, occurs when the chemical bonding is formed and requires a higher activation energy. The adsorbed atoms, when are detached from the surface, is defined as the desorption process.

The surface reaction mainly involves the possible reaction paths that can contribute into the catalytic recombination phenomena. In the common approach, the surface reaction can be classified into two different mechanisms: Eley-Rideal (ER) and Langmuir-Hinshelwood (LH) mechanisms. In the ER recombination mechanism, the recombination takes place between the incoming atom and the adsorbed atom. LH recombination mechanism, on the other hand, takes place between two adsorbed atoms. Schematic of surface reactions during the recombination process is shown in Fig. 2.

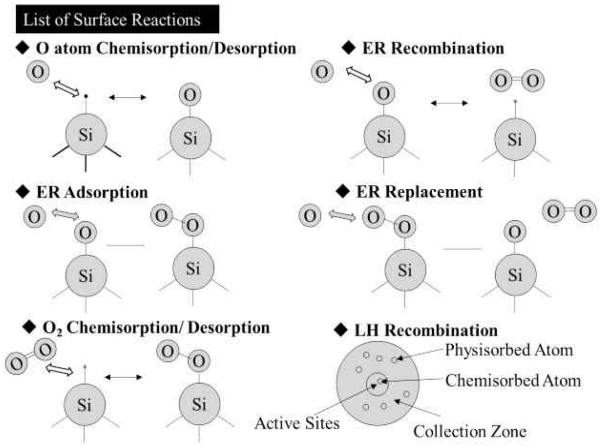

Fig. 2 Surface reactions during the catalytic recombination phenomena

The catalytic recombination process on the surface is mainly characterized by the catalytic recombination efficiency. This catalytic recombination efficiency γ is defined as the ratio of recombined atomic flux with the incoming atomic flux. The concept of finite catalytic recombination was firstly introduced by Goulard[13] in 1950s. In many computational fluid dynamics (CFD) application, the catalytic recombination efficiency is used as the input for the species wall boundary condition. Based on the recombination efficiency values, the catalytic boundary condition can be classified into: non-catalytic, finite-catalytic, and fully-catalytic. The schematic of these boundary conditions is shown in Fig. 3. The species concentration at the wall is calculated by imposing the species diffusion velocity equal to the catalytic velocity, shown in Eq. (1). In addition to the catalytic boundary condition, a local equilibrium wall[4] can also be implemented. In this specific wall boundary condition, the species concentration at the wall is assumed to be equal to that at local equilibrium condition.

$$k_i = \frac{\gamma}{4}\sqrt{\frac{8RT_w}{\pi M_i}} \qquad (1)$$

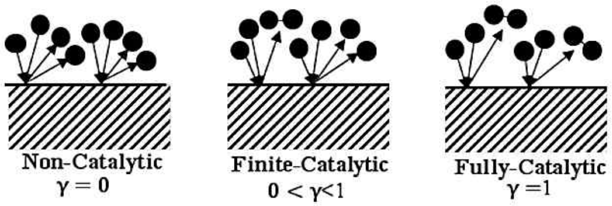

Fig. 3 Type of catalytic boundary condition

When dealing with the heat transfer calculation, an additional parameter called as energy accommodation factor (β) should be taken into account as well. This parameter is defined as the ratio of the energy transferred to the surface to the energy resulted from the recombination process[14]. Unlike the catalytic recombination measurement, the energy accommodation factor measurement only has some experimental measured values available in the open literature[15-17]. Often, the catalytic recombination efficiency is also defined by its apparent value, which is defined as $\gamma' = \gamma\beta$. In some cases, this value is also called as the global energy transfer recombination efficiency[18].

The catalytic recombination efficiency depends on several parameters such as temperature, pressure, and the surface morphology. The temperature dependence is often modeled based on the experimental measured data. Arrhenius-type reaction rate is chosen as the base equation on which pre-exponential factor and the activation energy is attained through the curve fitting procedure based on the experimental data. As a representative, the atomic oxygen catalytic recombination efficiency on a silica-like surface is shown in Table 1.

Table 1 Temperature dependence of oxygen catalytic recombination efficiency on silica-like surface

Author[Ref]	γ_O expression	Temperature [K]
Scott[19]	$16\exp(-10271/T)$	1400 – 1650
Kolodziej[20]	$40\exp(-11440/T)$	1435 – 1650
Rakich[21]	$0.8\sqrt{2\pi m_O/k_B T}$	1100
	$7\sqrt{2\pi m_O/k_B T}$	1600
Zoby[22]	$0.0094\exp(-658.9/T)$	800 – 1400
Gupta[23]	$8\exp(-8600/T)$	800 – 1400

The above Arrhenius-type catalytic modeling is also named as the specified reaction efficiency (SRE) model[24]. The conventional SRE model, at first glance, can be thought as a successful model since it predicts the catalytic recombination efficiency value. However, the SRE can only be applied at some temperature range, based on the curve-fitting data. Also, the SRE model lacks information about the reaction mechanism that supports the recombination process. Consequently, in the past decades, many further numerical studies have been attempted to fill the voids that SRE model has created. To achieve this objective, a more detailed catalytic recombination modeling is proposed. In the next section, an overview of this method is going to be explained.

3. Catalytic Numerical Modeling

In this section, we will consider two different subjects on the catalytic recombination modeling. In the first subsection, an overview of catalytic boundary layer theory is explained. This theory, reinforced with the experimental heat transfer, has been actively applied to compute the catalytic recombination efficiency.

In the second subsection, a more concentrated discussion on pure numerical modeling is elaborated. The numerical modeling aims to view the recombination mechanism from the elementary surface reactions described previously.

3.1 Stagnation Catalytic Boundary Layer

The hypersonic catalytic boundary layer has been studied since from 1950s[25, 26]. Fay and Riddell[26] formulated a stagnation heat transfer in the equilibrium flow based on the fully-catalytic assumption, show in Eq. (2). Goulard[13] later upgraded the stagnation heat transfer formulation by including the role of finite rate recombination. Goulard's approach has found its application in the experimental catalytic measurements that are based on heat transfer measurement[27-31].

$$q_{w} = 0.763\mathrm{Pr}^{-0.6}(\rho_w\mu_w)^{0.1}(\rho_s\mu_s)^{0.4} \left[1+(\leq^{0.52}-1)\frac{h_D}{h_s}\right](h_s-h_w)\sqrt{\left(\frac{du}{dy}\right)_s} \tag{2}$$

In his theory, Goulard[12] assumed that the thermal chemical equilibrium is reached at the boundary layer edge. Specifically, the considered geometry in the formulation is a hemisphere blunt body shown in Fig. 5. The boundary layer is assumed to be frozen so that no chemical reaction takes place inside. The solely allowed chemical reaction assumed to be occur at the wall, that is due to the heterogeneous catalytic recombination. The total stagnation heat transfer at the wall is comprised of the conductive heat transfer and the diffusive heat transfer.

$$q \tag{3}$$

To simplify the solution process, the boundary layer coordinate transformation is applied based on the Lees-Doronitzin's transformation [24], shown in Eq. (4) and Eq. (5). Imposing this coordinate transformation, the boundary layer governing equations are reduced into the differential equations. The subscript e denotes the flow properties at the boundary layer edge.

$$\eta = \frac{\mu_e}{(2s)^{1/2}}\int_0^y r_0\rho dy \tag{4}$$

$$s = \int_0^x \mu_e\rho_e u_e r_0^2 dx \tag{5}$$

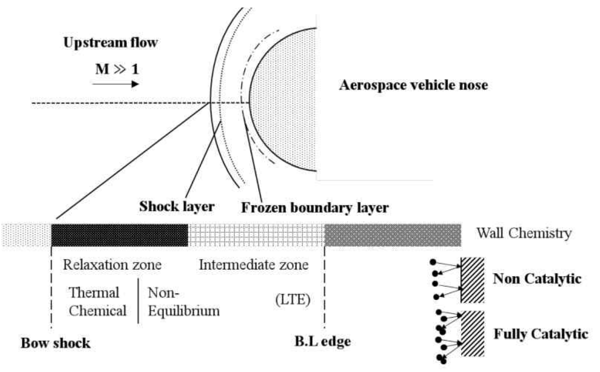

Fig. 4 Numerical modeling for catalytic recombination

The conductive heat transfer is computed based on the temperature distribution by coupling the momentum and energy equation in the boundary layer. This conductive heat transfer is expressed in Eq. (6), where Pr_w is the Prandtl number at the wall, β is the velocity gradient at the stagnation point.

$$q_{cond} = 0.48(2\beta\mu_e\rho_e)^{1/2}\mathrm{Pr}_w^{-2/3}\overline{h}_e \tag{6}$$

The diffusive heat transfer, on the other hand, is computed as the multiplication of the recombination heat released due to recombination with the diffusion flux. Expression for the diffusive heat transfer is shown in Eq. (7). The term $z(0)$ used in the expression is defined as the ratio of atomic mass fraction at the wall to that at the boundary layer edge.

$$\begin{aligned} q_{diff} &= h_R J_w = h_R k_w c_e z(0)\rho_w \\ &= h_R c_e \rho_w \frac{k_w}{\left(\frac{\mu_e\rho_e}{2\beta}\right)^{1/2}\frac{k_w Sc}{0.47Sc^{1/3}\mu_w}+1} \end{aligned} \tag{7}$$

At this point, it is important to look into some advantage and drawback of the Goulard's approach. The simple algebraic expressions shown in Eq. (6) and Eq. (7) provide an easy assessment of the total stagnation heat transfer. However, this approach is mostly appropriate in the binary gas mixture.

Using Goulard's theory in a multi-component gas mixture may lose its accuracy for predicting the total stagnation heat transfer. In the case of flow inside the boundary layer is not in the equilibrium state, a non-equilibrium approach is compulsory to compute the total heat transfer. As a consequence, to alleviate the problem, some improved method of Goulard's method have been proposed.

In the case of frozen boundary layer assumption, a non-equilibrium boundary layer approach has been extensively studied. For example, Inger[32] specifically considered the contribution that comes from the chemical reaction that may occur inside the boundary layer with binary gas consideration. Like Goulard, Inger also provided an algebraic formulation of stagnation heat transfer that depends on some important parameters like flow properties, body geometry, and gas recombination rate.

Not only the chemical non-equilibrium, but also the thermal non-equilbirium has been shown to have an influence in the catalytic phenomenon at the wall. The recombined molecules, when leaving the surface, are found to be in the excited state instead of in the ground state. Considering this fact, the vibrational kinetics and state-to-state modeling in the boundary has been lately investigated. As a representative, Armenise et al.[33, 34] performed a catalytic boundary layer study by considering each vibrational level of recombined oxygen as different species. In each species, a separated value for recombination coefficient is applied into the computation.

Regarding the limitation in the number of species in the Goulard's approach, Park[8] improved the Goulard's approach so that it can be applied for a tertiary gas mixture. Instead of using the Fick's binary diffusion law, the diffusion velocity for the tertiary gas mixture is computed according to the Chapman-Enskog approximation. The limitation of the Park's approach is that the third species is assumed to be inert so that no species variation is observed in the boundary layer

To determine the catalytic recombination efficiency using the Goulard-like approach, accurate values for the input parameters like flow properties at the boundary layer edge, the velocity gradient, and the measured heat transfer value is required[35-36]. The catalytic recombination efficiency is attained by finding the intersection between the experimental heat transfer value with the theoretical heat transfer. Figure 5 illustrates an example to compute the catalytic recombination efficiency from the Park's shock tube experimental data for copper material[9].

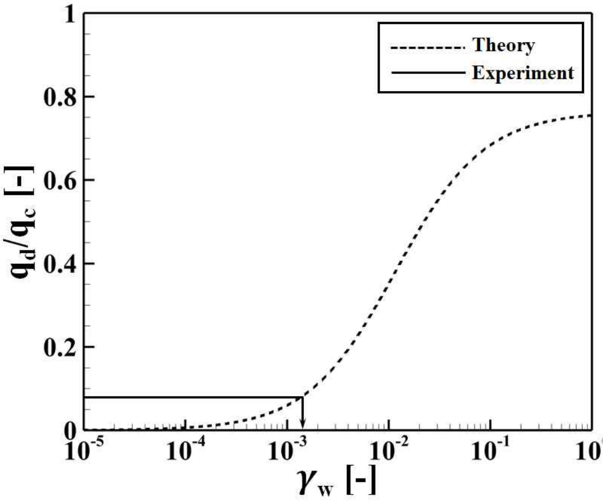

Fig. 5 Catalytic recombination theory with experimental heat transfer to obtain recombination efficiency

3.2 Gas Surface Interaction Modeling

The experimentally measured catalytic recombination efficiency is often used as the input parameter for catalytic boundary condition in the CFD computation. However, this type of boundary condition still lacks information about the reaction mechanism that build up the recombination process. Consequently, a different type of approach that is able to provide this kind of information is required. To be more exact, the interaction between gas and surface needs to be taken into account. One of the main method used in this modeling is by using the chemical kinetics modeling[37-41].

In the chemical kinetics approach, the aim is to model the interaction between the gas atoms and surface atoms. Basically, in its implementation, the chemical reactions governing the recombination mechanism are the important parameters. According to the chosen chemical reactions, the production rate for each species is computed.

A typical schematic representative of chemical kinetics modeling is shown in Fig. 6. In this example, for each considered chemical reaction, the chemical reaction rate can be defined. This chemical reaction depends on some quantity like: atomic gas concentration [A], atomic surface concentration $[A]_s$, and other parameters related to reaction rate constant. Generally, the adsorbed species concentration is solved for the steady state condition. The ratio of the adsorbed species concentration with the total available site concentration is defined as the surface coverage.

Depending on the number and type of chemical reactions at the surface shown in Fig. 2, a number of catalytic modeling studies have been done in the past recent decades.

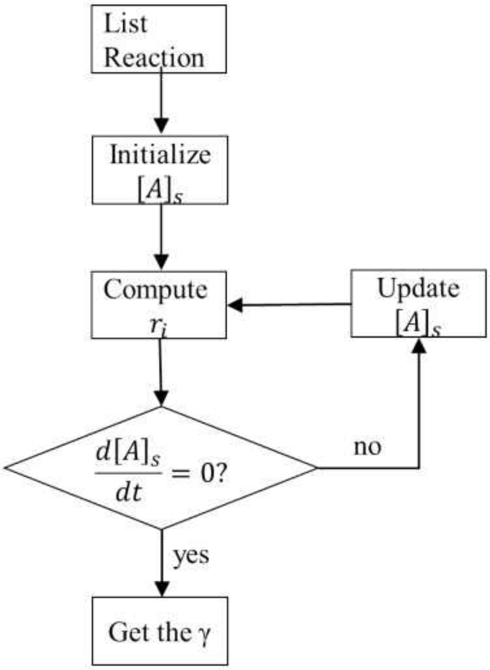

Fig. 6 Numerical method applied to obtain the recombination efficiency

A recent finite-surface catalytic modeling for silicon dioxide is proposed by Norman et al.[37] with the required reaction parameters obtained from the molecular dynamics simulation using the Reactive Force Field (ReaxFF) for the silica surface[42].

Table 2 Surface Reaction Rate Proposed by Norman et al.[37]

Reaction	Reaction Name
$O + E_s \leftrightarrow O_s$	Atomic Oxygen Adsorption
$O + O_s \leftrightarrow E_s + O_2$	Eley-Rideal Recombination
$O + O_s \leftrightarrow O_{2s}$	Oxygen Molecule Formation
$O + O_{2s} \leftrightarrow O_s + O_2$	Oxygen Molecule Replacement
$O_2 + E_s \leftrightarrow O_{2s}$	Oxygen Molecule Adsorption

In their updated work, Norman et al.[40] also proposed another set of surface reaction that is applicable for low surface temperature condition. Instead of dealing with the chemical adsorption, the low-temperature model implements the physical adsorption. An combined model based on these these two Norman's models are also proposed by Li et al.[38]. The validity of the model is highly dependent on how the parameters are defined. The rise of the molecular dynamics simulation for the atomic simulation in the hypersonic flow allows an estimation of these parameters.

4. Conclusions

An overview of the surface catalytic recombination phenomena in hypersonic flow is presented. Surface catalytic recombination is a process where two atoms hit the surface and recombine into the molecule due to the surface acts as a catalyst. A description of the numerical surface catalytic modeling is also given. A common method in modeling the surface catalytic recombination is by using the recombination catalytic efficiency. This method is mostly implemented in the numerical CFD calculation or stagnation heat transfer analysis with boundary layer. By combining with experimental measurement, the boundary layer approach can give an estimation for the material surface efficiency. Another numerical modeling described for the surface catalytic is based on the surface reaction modeling. This approach depends highly on the chosen surface reaction. With the present development of atomic modeling with molecular dynamics, a more accurate estimation of the reaction parameters for the surface reaction modeling can be obtained.

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<주요 연구 분야>

- 비평형 극초음속 공기열역학, 전산유체역학, 가스 표면 상호 적용