Mass and Thermal Accommodation Coefficients: Interfacial Transfer Efficiency

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Introduction

The Mass and Thermal Accommodation Coefficients (MAC and TAC, Eq. 1) are parameters which quantify the efficiency of mass and thermal energy transfer across an interface.

$$MAC_i = \frac{J_{actual}}{J_{theoretical}} \qquad TAC_i = \frac{T_{reflected} - T_{bulk}}{T_{surface} - T_{bulk}} \qquad \text{Eq. 1}$$

Literature values reported for the MAC and TAC over the past half century consistently vary up to three orders of magnitude, creating skepticism toward incorporating data into predictive models.

The authors have developed a preliminary model which predicts both the MAC and TAC over a large temperature range (300 to 3000 K). Interesting deviations from equilibrium emerge at combustion conditions, suggesting increased importance of the elucidation of the MAC and TAC.

Model Development

MAC

The theoretical maximum mass flux at an interface is described using tenets from the kinetic theory of gases. The rate of molecular collisions with a solid or liquid surface is defined as:

$$J_{theoretical} = \frac{n_g \bar{c}}{4}, \qquad \text{Eq. 2}$$

which redefines the MAC as:

$$MAC_i = \frac{J_{actual}}{\frac{n_g \bar{c}}{4}}. \qquad \text{Eq. 3}$$

Under rarefied conditions, where collisions with the interface are significantly more abundant than collisions with other molecules, both condensation and evaporation (directional corrections incorporated) are defined by Eq. 3.

A deviation from equilibrium (where the mole fraction of species *i* deviates from one) can be predicted by subtracting the actual from the equilibrium flux. After rearrangement, the actual interfacial mole fraction of species *i* is predicted by:

$$x_i^{actual} = \frac{MAC_i x_i^{condensed} n^{condensed} \bar{c}^{condensed} - 4J_{theoretical}}{MAC_i n_i^{gas} \bar{c}^{gas}} \qquad \text{Eq. 4}$$

TAC

The TAC deals in terms of energy transfer from a molecule to an interface upon collision, but is expressed, as seen in Eq. 1, in terms of temperatures. This is done valid via use of the Boltzmann approximation. Similar to the MAC, the TAC may be expressed in terms of the kinetic theory of gases. Analogously, departure of the surface temperature from the equilibrium condition may be as:

$$\frac{T^{gas} - T^{condensed}}{\left(T^{gas}\right)^{1/2}} = \frac{2q^{gas}\pi^{1/2}}{P(8R)^{1/2}\sum_i x_i m_i}\frac{1}{TAC_i} \qquad \text{Eq. 5}$$

MAC and TAC Data Acquisition

Droplet Train Flow Reactor (DTFR)

Currently one of the most accurate methods of measuring the MAC, is the DTFR (Figure 1) pioneered by Li and Davidovits (2001). The DTFR drops monodisperse water droplets through a chamber filled with trace isotopic gas species. The isotopic gas uptake is measured, and related via a three-step kinetic expression to the MAC. Representative MAC data obtained on specific compounds are included in Table 1.

Table 1: Representative values obtained for the MAC

Molecule	MAC (273 K)
Acetone	0.026
Methanol	0.056
Ethanol	0.049
HCl	0.233
H_2O_2	0.151
HNO_3	0.234

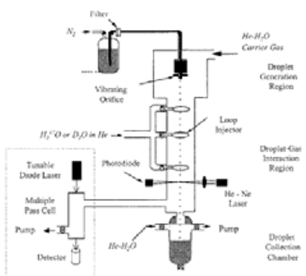

Figure 1: Droplet Train Flow Reactor Apparatus, Li and Davidovits (2001)

Droplet Growth and Evaporation Rates

Shaw and Lamb (1999) measured the evaporation rate and the liquid-solid (water-ice) homogeneous nucleation rate of water droplets by Mie light-scattering techniques. Combination of the two types of measurements greatly reduces data ambiguity.

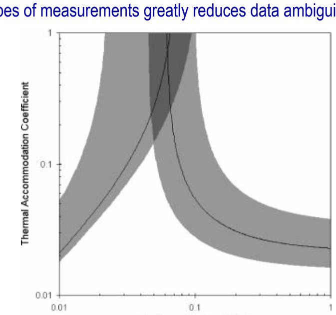

Figure 2: Graph schematic of most probably combinations of MAC and TAC based of measured nucleation and condensation rates (condensation coefficient is equivalent with the MAC). Shaw and Lamb (1999)

Theory Predictions

When Eqs.4 and 5 are simultaneously solved with the equations for steady, one-dimensional heat and mass transfer, the model can predict droplet surface temperature, heat flux, and mass flux as functions of the bulk or ambient temperatures and the MAC and TAC. Results are shown below in Figures 3, 4, and 5. Here α and β represent the MAC and inverse of the TAC (1/TAC).

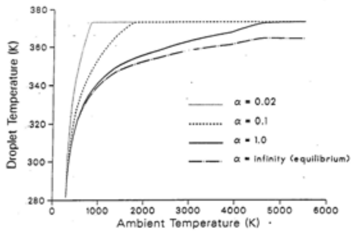

Figure 3: Droplet Temperature as a function of ambient temperature with the TAC held constant at unity.

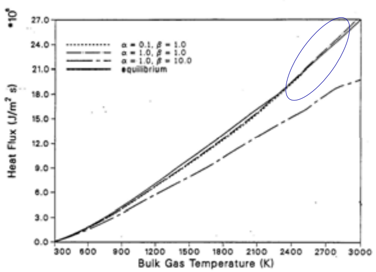

Figure 4: Heat flux as a function of ambient temperature with the varying MAC and TAC. Notice the deviation from equilibrium at high bulk gas temperatures.

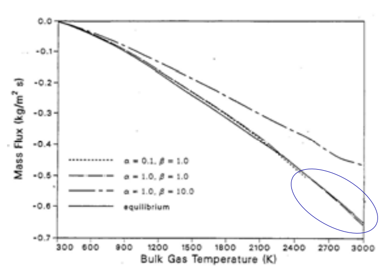

Figure 5: Mass flux as a function of ambient temperature with the varying MAC and TAC. Again, notice the deviation from equilibrium at high bulk gas temperatures.

Proposed Experimental Apparatus

To validate the results predicted by the proposed theory, a simple apparatus has been devised.

A thermistor with a diameter approximately 100 nm across will be placed beneath a liquid surface in a closed container under a vacuum at 10^{-6} mmHg. Liquid evaporation will be controlled at a constant rate via a micro pump to allow the interface to slowly recede over the perimeter of the thermistor.

The surface temperature profile surrounding a sphere as a function of time will be obtained.

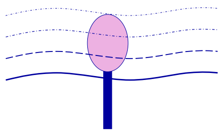

Figure 6: Schematic of proposed experimental apparatus to measure the temperature profile at the surface of a sphere.

Theory Comparison with Existing Data

The authors are currently finding different avenues to modify existing MAC and TAC data to validate theory predictions. Both data obtained on gas-solid and vapor-liquid interfaces are being considered.

Helpful Materials

The following list of articles and book chapters provide a thorough review of work-to-date on the MAC and TAC.

•Shaw R., Lamb D., (1999) *J. Chem. Phys.*, 111 (23), 10659

•Nathanson G., Davidovits P., (1996) *J. Phys. Chem.*, 100, 13007

•Saxena S., Joshi R., (1981), *Thermal Accommodation and Adsorption Coefficients of Gases*, McGraw-Hill, Library of Congress Cataloging in Publication Data (chapters 1-4)

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