Atom recombination on surfaces in plasmas - an experimental study

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Surface-catalysed recombination of atoms and free radicals plays a key role in the plasma equilibrium for molecular plasmas in enclosed reactors. This process is routinely characterized by a surface reaction probability, γ, (between 0 and 1), which for simplicity is often assumed to be a constant. However, this parameter is nearly always poorly characterized, or indeed used as the adjustable variable to fit models to experimental data. Ab-initio theories are not currently able to predict surface reaction probabilities reliably, so they must be determined by in-situ measurements (either through the measurement of spatial mole-fraction gradients adjacent to the surface, or (more commonly) by time-resolved measurements in modulated plasmas).

We have made measurements of γ for oxygen atoms in pure O_2 plasmas in different plasma devices (a DC glow discharge in a borosilicate glass tube at pressures 0.5-8 Torr, in a radiofrequency capacitively-coupled plasma reactor with aluminium electrodes at pressures 1-6 Torr, and in an inductively coupled plasma at 5-100 mTorr. The time-resolved oxygen atom density was determined by time-resolved optical emission actinometry[1], two-Photon Laser-Induced Fluorescence and monomode laser Cavity Ring Down Spectroscopy (CRDS) of the forbidden O $^3P_2 \rightarrow {}^1D_2$ transition[2] at 630nm.

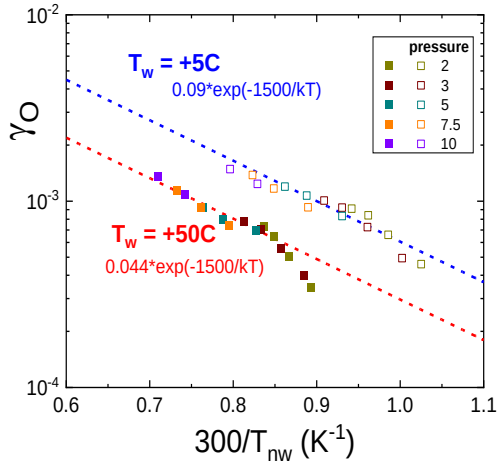

Figure. 1. *Oxygen atom surface recombination probability on borosilicate glass as a function of near-wall gas temperature.*

The values of γ obtained are not constant, but vary considerably with O_2 pressure, plasma density, plasma exposure time and air leak rate. At gas pressures above 150 Pa the recombination coefficient on both borosilicate glass and oxidised aluminium surfaces is small, of the order several 10^{-4}, and obeys an Arrhenius law with the gas temperature (activation energy of ~0.15eV) indicating an Eley Rideal mechanism. The oxygen atom decay in the afterglow is not exponential, but slows down with afterglow time. This indicates the presence of two types of surface reaction sites; strongly-bound chemisorbed O and weakly-bound physisorbed O. The latter become important for high incident atom fluxes, leading to non-exponential decays. At lower gas pressures the recombination coefficient becomes much bigger, of the order 0.1 in an inductively-coupled plasma at 10 Pa. These observations suggest that energetic ion bombardment causes activation of the surfaces, strongly increasing the surface recombination probability[3].

Références

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[3] Afonso J, Vialetto L, Guerra V and Viegas P *Journal of Physics D: Applied Physics* 2023 **57** 04LT01

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