

A NEW LOOK AT OXYGEN PLASMAS - QUANTITATIVE SPECTROSCOPY FOR RIGOROUS TESTING OF MODELS

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Abstract. Despite many decades of study, models of discharges in molecular gases still lack accurate data on many key collisional processes, even for such “simple” and ubiquitous gases as O₂. Good data is lacking for near-threshold electron-impact dissociation, surface recombination, the role of metastables, of gas heating, of vibrational excitation, of energy transfer and surface thermal accommodation. Direct measurement of the rate constants of individual processes is a fastidious process, where it is even possible. As an alternative approach, we compare comprehensive measurements of internal plasma parameters to simulations for a plasma with relatively simple chemistry, namely a DC positive column discharge in pure O₂. This well-characterized, stable and uniform discharge is optimal for experiment-model comparison. Although it has been studied for a very long time, new experimental methods, including synchrotron Vacuum ultraviolet absorption spectroscopy and laser cavity ringdown absorption spectroscopy (CRDS), allow the densities of all the major species (atomic, molecular, in ground and excited states), as well as the gas translational temperature to be measured, with much-improved absolute accuracy, and with time resolution. Applied to (partially- and fully-) modulated discharges, these measurements provide unprecedented insight into the kinetic processes occurring (in the gas phase and at surfaces), and a profound test of the models. Whereas a model can be adjusted to fit a set of steady state measurements at one given set of operating conditions, trends with pressure and discharge current, and especially the temporal response to current modulation, are much harder to reconcile with an incorrect model. In practice, model failures often result from omission of key processes, or to neglect of their temperature-dependence. With the relatively simple chemistry occurring in pure O₂, with measurements of all principal species as well as the gas temperature, it is possible to identify the missing reactions, and even estimate their rates (and/or activation energies) by adjusting the model to fit the measurements.