Features of oxygen metastable molecules kinetics in O2 plasma with increasing pressure

D. Lopaev[1], A. Volynets[1], T. Rakhimova[1], O. Proshina[1], A. Chukalovsky[1]

[1]*Skobeltsyn Institute of Nuclear Physics, Moscow State University, 119991 Moscow, Russia*

As known, all excited electronic states in O_2 molecule below the dissociation limit are metastable because the O_2 ground state in contrast to most of molecules is triplet and of negative parity. So the lowest $a^1\Delta_g$ and $b^1\Sigma_g^+$ states have rather low rates both of radiative decay (radiative lifetimes are ~4000s and ~12s respectively) and of nonresonant collisional quenching. In case of neutrals only specific reactions going through the quasi-resonant energy exchange between singlet O_2 states and states of the neutrals are able to provide high loss rate. It opens opportunities for accumulating high percent of O_2 molecules in the singlet states (especially in $O_2(a^1\Delta_g)$) in order to use it further for application: such as lasing in COIL or initiating combustion. The electrical discharges provides effective excitation of O_2 electronic states and especially the lowest singlet states since the excitation energy of the last is below characteristic electron temperature for most of the discharges. And seemingly one can apply the discharges for effective and fast production of $O_2(a^1\Delta_g)$, but most of plasmas is found to be rather ineffective in it especially with increasing pressure and power despite that estimated quenching by neutrals is still low under those conditions.

This report is just devoted to detail consideration of $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$ kinetics in O_2 plasma at increased pressures where experimentally observed rather fast quenching of these molecules unexplained from the viewpoint of known kinetic schemes of reactions. The research was carried out with O_2 rf (81 MHz) in the pressure range of 10-100 Torr. It was transversal discharge with external electrodes in quarts tube to exclude fast heterogeneous quenching on metallic surface. Extremely fast $O_2(a^1\Delta_g)$ quenching was experimentally observed in discharge while quenching in afterglow was rather slow. It is shown that processes with secondary electrons and oxygen atoms are responsible for this fast quenching. Direct and secondary electron processes provide fast establishing balance between the populations of O_2 states while fast reactive quenching of $O_2(b^1\Sigma_g^+)$ by O atoms shift the balance to the ground state. This effect increases with pressure because of increasing O density and significantly grows with discharge power due to increasing gas temperature stimulating $O_2(b^1\Sigma_g^+)$ reaction with atomic oxygen.

Thus it provides quite clear limitation on discharge systems for effective pumping of singlet oxygen. This limitation concerns as oxygen pressure (including mixtures) and discharge schemes that should provide low power density at increased pressure for avoiding fast loss of metastable O_2 molecules.