

INFLUENCE OF HUMIDITY ON PHOTOCHEMICAL OZONE GENERATION WITH 172NM XENON EXCIMER LAMPS

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Abstract. The reaction kinetics of photochemical ozone (O₃) generation in humid air and Oxygen (O₂) using efficient, narrow band Vacuum Ultra Violet (VUV) 172nm Xenon excimer lamps is discussed. Trace amounts of water (H₂O) vapor in the process gas leads to Hydroxyl (OH) and Hydroperoxy (HO₂) radical formation. These radicals drive a catalytic O₃ destruction cycle limiting O₃ saturation concentration. This catalytic O₃ destruction cycle was included into a quantitative kinetic model describing photochemical O₃ production. Experimental O₃ saturation concentrations obtained with coaxial VUV driven photochemical O₃ generators compare satisfactorily well with the models predictions.

1. INTRODUCTION

Short wavelength Vacuum Ultra Violet (VUV) radiation ($\lambda < 180\text{nm}$) emitted from VUV lamps can photo split Oxygen molecules (O₂) with high cross sections and thus initiate ozone (O₃) formation [1]. The photochemistry of VUV induced O₃ production is well known due to studies of stratospheric O₃ [2]. Since lamps operate independently of the photo reactor region, a wide feed-gas parameter range is possible. Since nitrogen (N₂) is transparent to this VUV light, very clean O₃ without any nitrogen oxide (NO_x) contamination is generated. New Xenon excimer 172nm VUV lamps can overcome all the shortcomings of 185nm low pressure mercury (LPM) lamps presently used to drive photochemical O₃ generators [3], emitting only narrow band VUV radiation around 172nm, with up to 40% wall plug efficiency [4, 5]. This allows system size independent O₃ yields of up to 200g/kWh. Since VUV radiation photo splits O₃ with only a slightly higher cross section than O₂, high O₃ saturation concentrations ($\leq 10\text{wt\%}$ in air, $\geq 30\text{wt\%}$ in O₂) [1], that favorably compare to values achievable with modern silent discharge O₃ generators ($\leq 5\text{wt\%}$ in air, $\leq 20\text{wt\%}$ in O₂), are theoretically possible. Unfortunately, first experiments generating O₃ with 172nm UV light using O₂ as a feed gas, showed O₃ saturation concentration of only 5wt% [1] or less [6]. Now, the discrepancy between theory and experiment can be explained and overcome by including the influence of water vapor into the rate equation system describing O₃ generation.

2. VUV DRIVEN COAXIAL O₃ GENERATOR IN DRY AIR AND O₂

A schematic of a VUV photochemical O₃ generator is shown in fig. 1. A graphic plan of all the reactions can be seen in fig. 2. A Xenon excimer lamp with $\varnothing = 2r_0$ and a VUV irradiance of 50mW/cm² is placed coaxially in a tube with $\varnothing = 2R$ containing O₂ and N₂ at densities $n(\text{O}_2, t=0)$ and $n(\text{N}_2)$, resulting in a VUV Photon flux of:

$$\Phi(r) = \Phi_0 \cdot (r_0/r) \cdot \exp(-(r-r_0)/\delta) \quad (1)$$

$$\delta = (\sigma_1 \cdot n(\text{O}_2, t=0))^{-1} \quad (2)$$

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