Simulation of an Ozone Nozzle for Oxide Molecular-Beam Epitaxy at PARADIM

Joseph Dill
2020 PARADIM REU Intern @ Cornell
Intern Affiliation: Physics, University of Minnesota - Twin Cities

PARADIM REU Mentor: Dr. Darrell Schlom, Materials Science and Engineering, Cornell University

ABSTRACT

The Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials (PARADIM) operates a molecular-beam epitaxy system to grow oxide thin films. In this study, we develop a computer simulation of gas emission from a nozzle in a high vacuum environment, then simulate a new nozzle design and compare its performance to the existing nozzle. The new nozzle improves the average ozone flux on a 1x1 cm substrate by 117% when the pressure inside the growth chamber is 10^{-5} torr and 53% at 10^{-6} torr. Additionally, the new nozzle adequately oxidizes 36% of a 3-inch diameter substrate, compared to the 19% achieved with the existing nozzle.

INTRODUCTION

Molecular-beam epitaxy (MBE) is a thin film growth method in which separate molecular beams of reactants are deposited on a substrate in an ultra-high vacuum, allowing atomic-level control of the film contents. The growth of an oxide such as $YBa_2Cu_3O_7$ by MBE involves separate molecular beams of Y, Ba, Cu, and ozone. Molecular beams of the metals are created by heating crucibles containing each element. The ozone gas, however, is emitted towards the substrate surface from a nozzle. The activity of ozone is roughly a billion times that of molecular oxygen [1], allowing it to form $YBa_2Cu_3O_7$ without destroying the long mean-free-path needed for MBE. Thus, ozone is a common oxidant for oxide MBE.

At PARADIM, thin films can be grown by MBE on one of two substrate geometries: 1) 1x1 cm square or 2) 3-inch diameter circle. The existing nozzle emits a beam of ozone from a single long pipe aimed at the substrate's center. This design does not provide enough ozone at the outer edge of the 3-inch diameter substrate to fully oxidize the material being grown.

This study's objective is to simulate gas emission from a pipe in high-vacuum and design a new ozone nozzle that produces larger fluxes on the 1x1 cm substrate and a uniform flux on the 3-inch substrate. This study builds on previous simulations conducted by Kraisinger et al. [2], who also designed an ozone delivery nozzle for an oxide MBE system.

METHODS

In a high-vacuum environment, the angular distribution of gas emitted from a circular pipe orifice is a cosine distribution that becomes more focused as the pipe's length-to-diameter ratio increases [3]. The equations used to calculate this collimating effect are given in [4]. Mathematica was used to simulate the flux generated by given nozzle specifications. Each differential area element of the pipe orifice can be treated as a flow area emitting the same collimated distribution as the pipe at large [5]. Thus, the flux at any point on the substrate is obtained by integrating the flux contribution from each differential flow element across the entire pipe orifice. Additionally, the substrate platform rotates during growth, making roughly one revolution in the time required to deposit an atomic monolayer. The average flux at a given radius from the center of the substrate after a rotation is obtained by integrating the flux pattern around a circle of that radius concentric with the substrate.

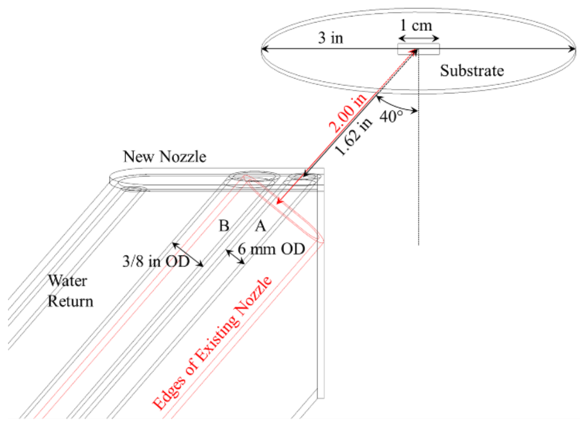

Figure 1: CAD drawing of the new nozzle. The size and position of the existing nozzle are shown in red.

The referenced emission equations apply to gas in molecular flow. Under typical growth conditions, ozone enters the nozzle in intermediate flow but exits in molecular flow. The length of the portion of the ozone delivery pipe in which the gas is in molecular flow is obtained by treating the pipe as two pipes in series. Gas is in intermediate flow in the first pipe and molecular flow in the second. Equations for the conductance of pipes in each regime are provided in [5]. By equating both pipes' throughput, one can solve for the length of the molecular regime pipe.

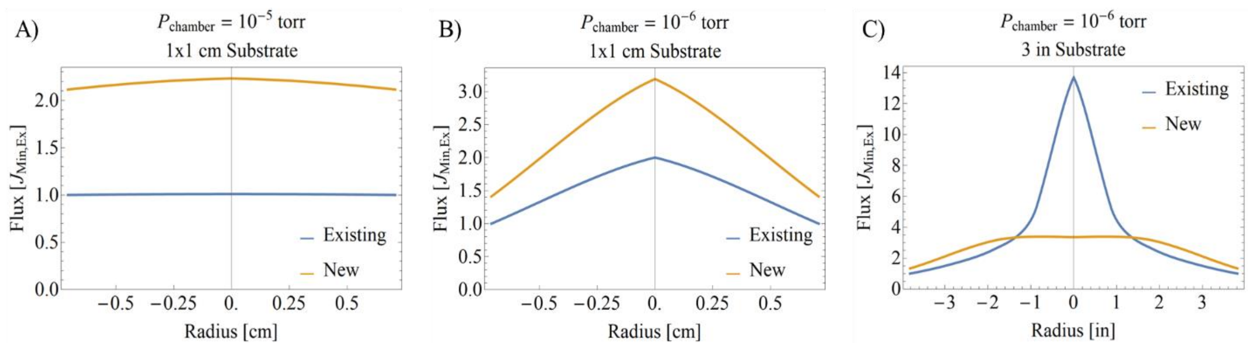

Figure 2: Simulated flux patterns, averaged over one rotation of the substrate, created by the existing (blue) and new (yellow) nozzle designs. The chamber pressure and substrate size for each plot are A) 10^{-5} torr, 1x1 cm; B) 10^{-6} torr, 1x1 cm; C) 10^{-6} torr, 3 in. In each graph, flux is plotted in units of the minimum flux from the existing nozzle for the respective substrate size and chamber pressure.

RESULTS AND CONCLUSIONS

All proposed new nozzle designs were constrained to the existing nozzle's position and orientation to avoid interference with other MBE system components. Additionally, the ozone must be water-cooled until it exits the pipe such that it does not dissociate into oxygen. So, all proposed nozzle designs were water-tight and included water supply and return pipes.

The final nozzle design, shown in Figure 1, is different from the existing one in two ways. Firstly, the nozzle casing has two edges, perpendicular and parallel to the substrate, rather than one edge perpendicular to the ozone delivery pipe and angled relative to the substrate. As shown in the figure, this modification decreases the total distance from the edge of the ozone delivery pipes to the substrate's center. The flux at a given point is inversely proportional to the squared distance from that point to the source of the flux substance. Hence, this modification increases the ozone flux at all points on the substrate surface.

Secondly, the width of the new nozzle is 2-7/8-inches while the existing pipe is only 1-inch wide. The wider nozzle can hold two ozone delivery pipes, one for each substrate geometry. Note that widening along this dimension does not interfere with any existing MBE system components.

Pipe A is intended for thin-film growth on 1x1 cm substrates. While the existing ozone delivery pipe has an outer diameter (OD) of 1/4-inch, pipe A only has a 6 mm OD, which creates a more concentrated beam. Figures 2A and 2B show the simulated flux pattern generated by the existing pipe and pipe A on the 1x1 cm substrate at two frequently used chamber pressures, 10^{-5} and 10^{-6} torr, respectively. The new nozzle increases the average flux across the 1x1 cm substrate by 117% when the chamber pressure is 10^{-5} torr and 53% when it is 10^{-6} torr.

Pipe B, designed for growths on 3-inch diameter substrates, has a 3/8-inch OD. It is aimed off-center such that, as seen in Figure 2C, the flux after rotation is uniform across a large portion of the substrate. With the existing nozzle, the flux at the center of the 3-inch substrate is larger than that at the edge of the substrate by a factor of 13.7. Pipe B improves the flux uniformity, decreasing this factor to 2.5.

FUTURE WORK

The nozzle described in this report will be built and installed in the oxide MBE system at PARADIM.

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