High-Capacity Rotary Drum For Atomic Layer Deposition Onto Powders and Small Mechanical Parts In A Hot-Walled Viscous Flow Reactor[1]

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Abstract

We designed and benchmarked a new rotary drum that can be inserted into pre-existing ALD tubular reactors. The drum enables laboratory-scale coating of powder samples and small mechanical parts without the design complications of fluidized bed reactors. We found that rotation significantly decreased saturation time, and that (with rotation) saturation time scaled nearly proportionally to powder quantity. We were able to provide a homogeneous coating of Al_2O_3 on up to 75 grams of silica gel powder, surface area ~1500 m^2.

Rotary Drum Design

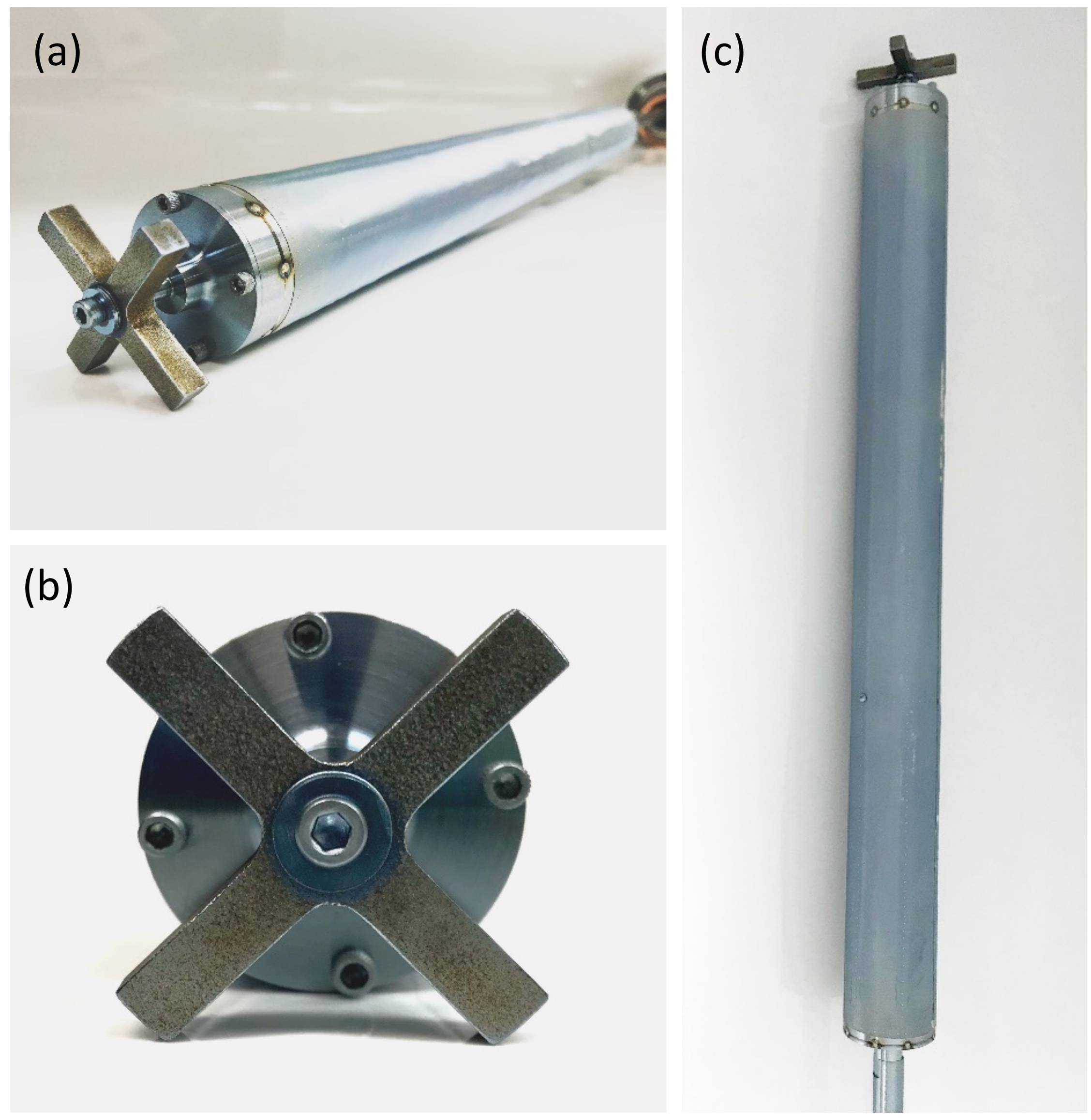

Figure 1: Rotary drum with a capacity of 100s of grams of powder (a) full view (b) end view (c) top view

The rotary drum is constructed of stainless-steel 325 mesh surrounding a ribbed cylinder 3.81 cm in diameter and 40.64 cm in length. The end (shown in figure 1(b)) is removable for loading and unloading of powder. The drum can be loaded into a tubular reactor, where an X-shaped support rests on the bottom of the reactor, allowing the drum to freely rotate. The drum is magnetically coupled to a variable speed servo motor powered by an Arduino-based speed controller.

Rotation Effect on Saturation Time

Saturation time decreased with rotation, demonstrating that mixing aided precursor transport into the powder bed.

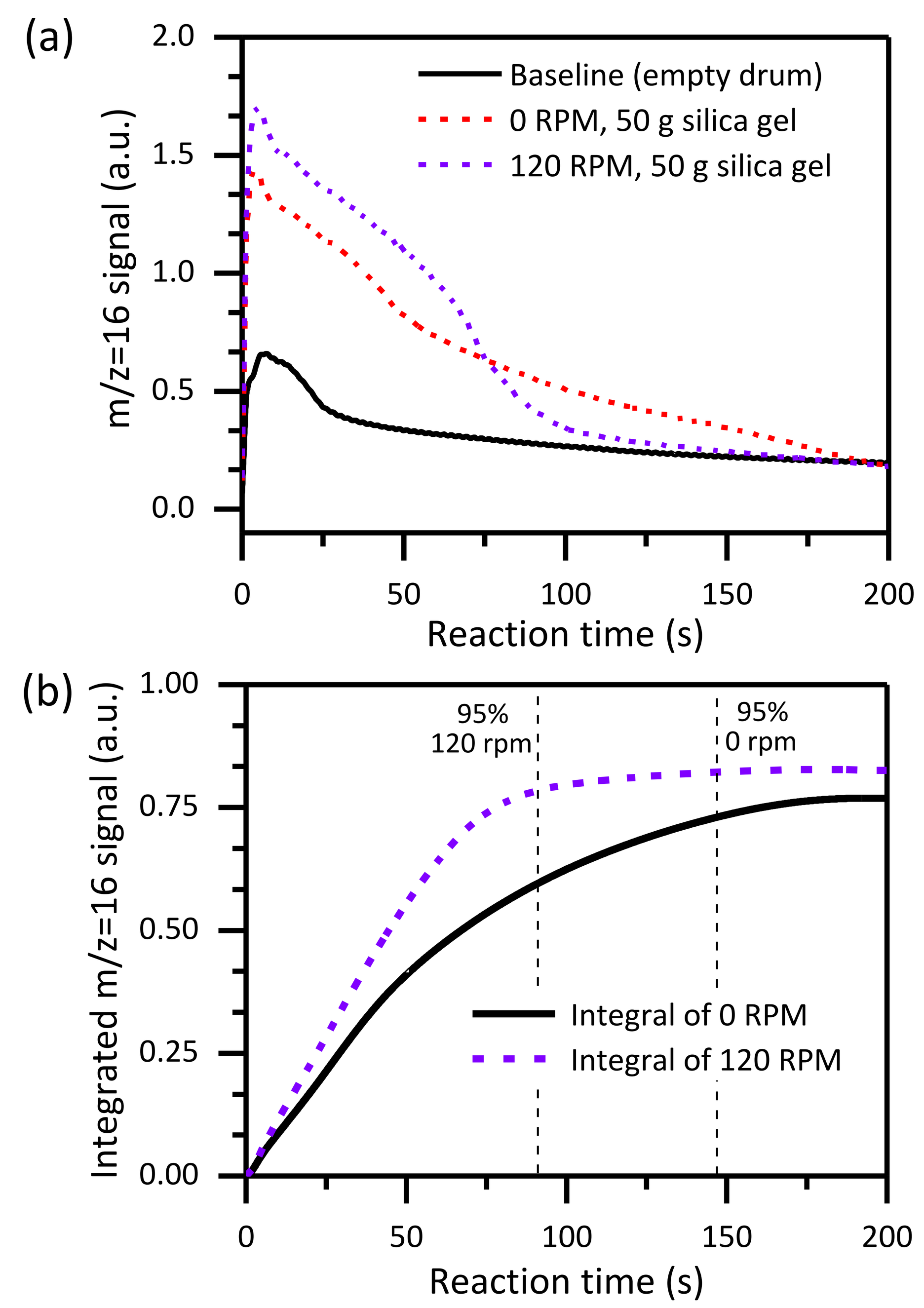

Figure 2: (a) Mass spectrometer m/z=16 signal during TMA dose (b) baseline-subtracted integrated values from (a), saturation time defined at 95% of maximum integral value

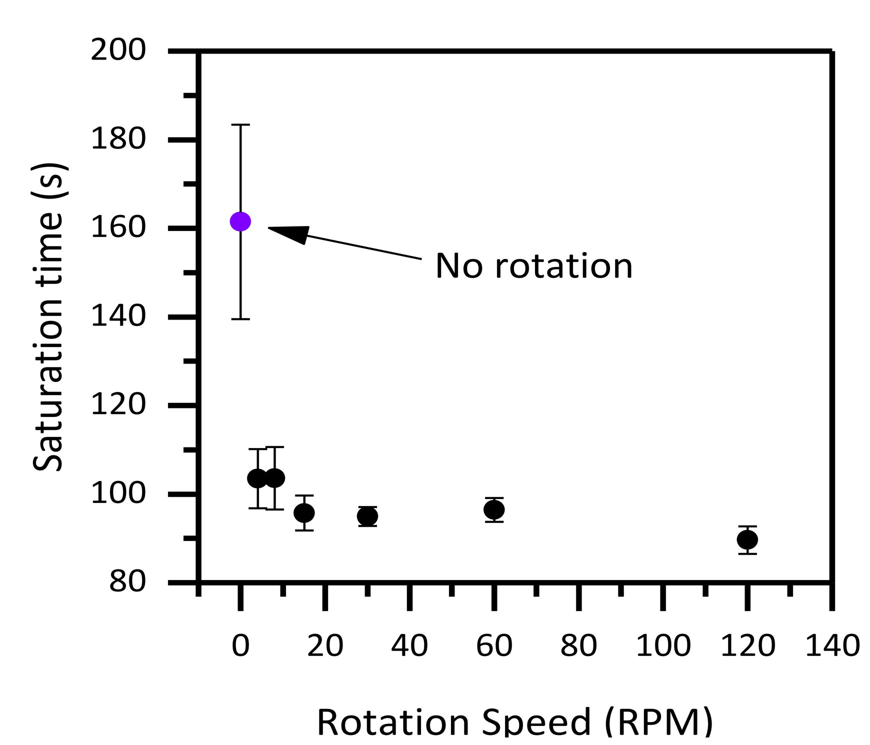

Figure 3: Saturation times (TMA dose) extracted from plots like Fig 2(b). Error bars represent 1 standard deviation calculated from duplicate measurements

Effect of Powder Quantity

Mass gains were approximately constant regardless of initial powder mass. We expected a linear saturation time response to increased initial mass. While the response is mostly linear, it appears that there may also be transport differences between H_2O and TMA within the silica gel.

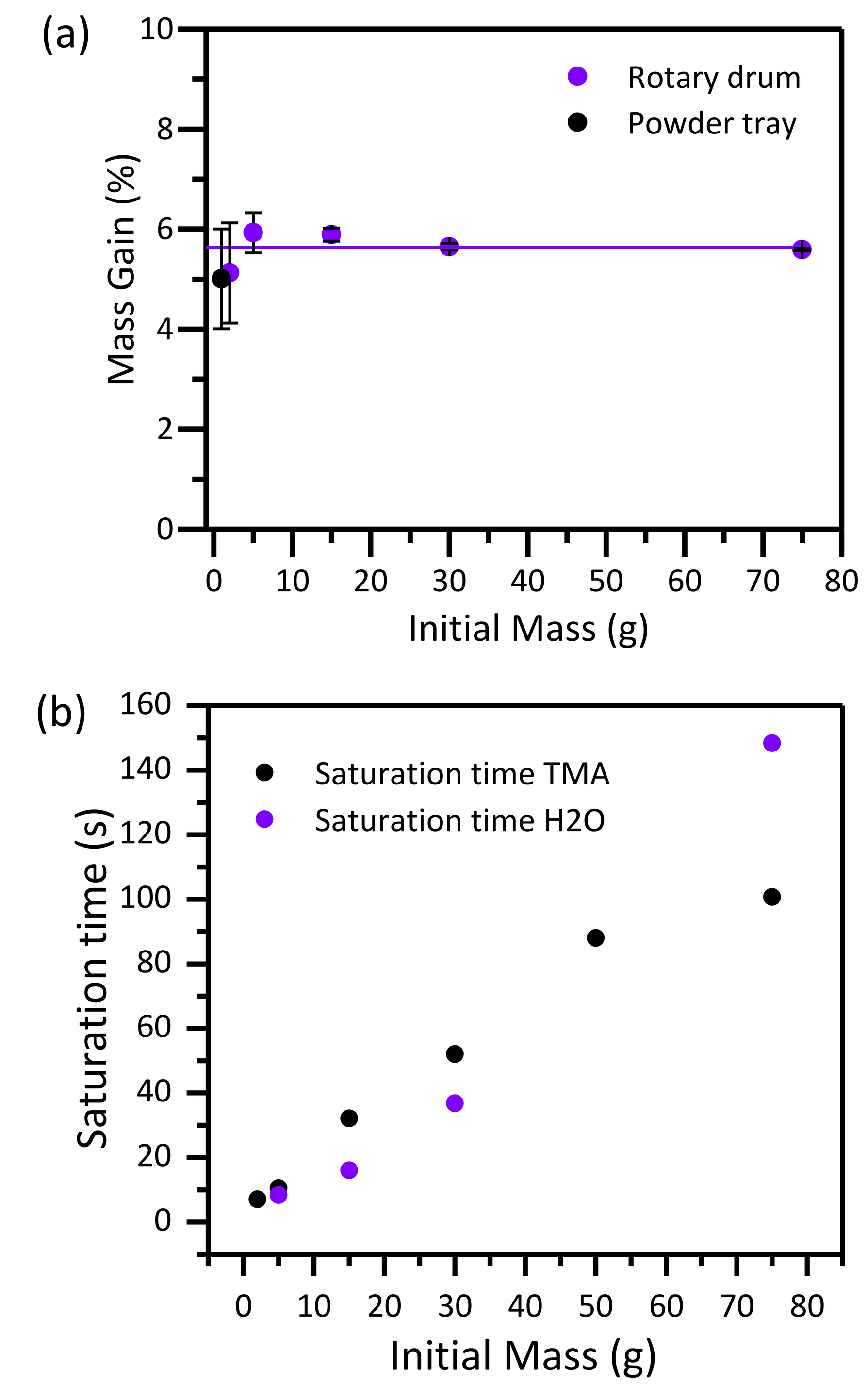

Figure 4: (a) Percentage mass gain after 5 cycles of TMA-H_2O ALD. Lower mass gains at 1 and 2 grams are attributed to the relative mass losses during powder transfer (b) saturation time versus powder mass

Table 1: X-ray photo electron spectroscopy data demonstrating comparable results for powder coated in new rotary drum compared to 1 gram of powder spread out in a tray

	Si 2p	Al 2p	O 1s	Al/Si
Powder Tray (1 g)	26.4	11.1	62.5	0.42
Rotary Drum (30 g)	23.3	14.2	62.6	0.61

Methods

We deposited Al_2O_3 on silica gel (Silicycle S10040T) with a surface area of 20 m^2/g and a primary particle size 75-200 microns via alternating doses of trimethylaluminum (TMA) and H_2O at 200 °C, reproducing previous work.[2] Reaction products were monitored using quadrupole mass spectrometry tracking a m/z ratio of 16, corresponding to methane released during the TMA-H_2O reaction. The background-subtracted QMS signal was integrated and saturation times were calculated at the time of 95% of maximum integrated value. A variety of rotation speeds and powder masses were tested to establish the relationship between these variables and saturation time. The pre- and post-deposition powder masses were measured to confirm constant mass gain. ALD on small nuts and bolts was performed to visually demonstrate homogeneous coating, and X-ray photoelectron spectroscopy (XPS) was performed to compare film quality to that of coating a small amount of powder spread out in a powder tray.

Visual Confirmation of Coating Homogeneity

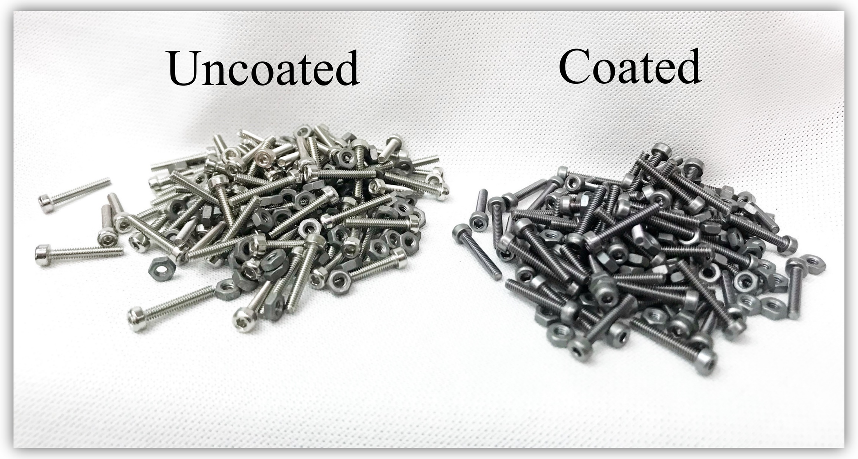

Figure 5: Nuts and bolts were coated with 500 cycles of TMA-H_2O in the rotary drum. The uncoated parts are shown at left; the coated parts are shown at right. The rotation of the drum enabled uniform coating (no spots left uncoated) of the parts

Acknowledgements

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2) Elam, J.W., Libera, J., Huynh, T., Feng, H., and Pellin, M. Atomic Layer Deposition of Aluminum Oxide in Mesoporous Silica Gel. J. Phys. Chem. 2010, 114. 17286-17292