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Effects of Inlet Flow Rates on Purge Durations in an Atomic Layer Deposition Process

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Abstract – The effects of inlet flow rates on the purge durations in an atomic layer deposition (ALD) process are investigated through the simulation of three-dimensional laminar multicomponent flow in viscous flow reactors. The operating pressure and temperature are 10 torr (1333 Pa) and 300 °C, respectively. Purge durations in reactors with inlet located on the top surface of the reactor are compared with those in a base reactor with one inlet at the bottom surface of the reactor. It is found that purge durations are reduced by an increase in the flow rates, but they are independent from the number of inlets if the flow rates are maintained equal among different reactors. One exception is the reactor with one inlet at the center of the top surface of the reactor, which experiences the longest purge durations, most likely due to the axisymmetric gas injection in this reactor. The acquired results will provide a better understanding about designing efficient viscous flow reactors to reduce both purge duration and gas consumption.

***Keywords*:** Atomic layer deposition; Viscous flow reactor; Purge duration; Multicomponent flow.

1. Introduction

Atomic layer deposition (ALD) is a key enabling nanotechnology to deposit ultrathin and highly conformal films on complex structures [1], [2]. ALD is a cyclic process, and each cycle consists of two precursor exposures with a purge between them. During the purging, unreacted precursors and reaction products from the previous precursor exposure are removed from the reactor, which is an essential step to avoid gas-phase reactions of precursors and defects in the deposited films [3], [4]. Viscous flow reactors with operating pressures of up to 10 torr (1333 Pa) are the most widely used ALD reactors [5]. However, despite deposition of high-quality films in these reactors, they are prone to limitations of long operational times, mainly due to long purge durations. As a supplement to the previous research by the author at [6], the present computational study is performed to investigate the effects of inlet flow rates on the purge durations in ALD viscous flow reactors.

2. Computational Model

All the reactors, shown in Fig. 1, include one outlet. The inlet and outlet diameters are 2.0 mm in all the reactors. The conventionally used ALD reactor is the base of comparison in this study and called "Base". It includes one inlet at the bottom surface of the reactor. The new reactors are top-inlet reactors (TIRs) with inlets at the top surface of the reactor. The TIR with one, two, and four inlets are shown as TIR-1, TIR-2, and TIR-4, respectively. The numerical procedure is described for deposition of Al_2O_3 from TMA, $Al(CH_3)_3$, and ozone, O_3, based on the global reaction $2Al(CH_3)_3 + O_3 \rightarrow Al_2O_3 + 3C_2H_6$. Pure argon (Ar) is used as the purge gas. The reaction mechanism is as follows [5]:

TMA exposure	$Al(CH_3)_3 + O^* \rightarrow Al(CH_3)_2^* + 0.5C_2H_6$	(1)
O_3 exposure	$O_3 + M \rightleftarrows O_2 + O + M$	(2)
	$O + O_3 \rightleftarrows 2O_2$	(3)
	$2Al(CH_3)_2^* + O \rightarrow Al(CH_3)OAl(CH_3)^* + C_2H_6$	(4)
	$0.5Al(CH_3)OAl(CH_3)^* + O \rightarrow O^* + 0.5C_2H_6 + 0.5(Al_2O_3)^B$	(5)

The asterisk and B superscripts represent surface and bulk species, respectively, and the remaining elements are gases. Since the focus of this work is to study purge durations, the reactions are neglected and only multicomponent flow transport inside the reactors is investigated. However, the precursor and gaseous products of reactions are pulsed into the reactor at each precursor exposure to keep the process similar to an ALD process. A timing-sequence of 0.5-0.5-1-0.5 is used for the cycle, corresponding to (i) pulsing TMA and C_2H_6 for 0.5 s, (ii) purging the reactor with pure argon for 0.5 s, (iii) pulsing O_3, O_2, O, and C_2H_6 for 1 s, and (iv) purging the reactor with pure argon for 0.5 s. This timing sequence is long enough to reach unchanged mass fractions in the reactor in the first and second precursor exposures and fill more than 99.9% of the reactor volume by pure argon at both purges. The inlet temperature is 27 °C. Inlet mole fractions of each gas in the first and second precursor exposures are 0.5 and 0.25, respectively. The inlet mole fraction of Ar in both purges is 1.0. The inlet precursor velocity for both precursor exposures is 0.6 m/s. However, the purge durations are investigated at different inlet purge velocities. The simulations are performed for a fixed operating pressure and temperature of 1333 Pa and 300 °C, respectively. The governing equations are transient continuity, momentum, energy, and species transport equations. The accuracy of applying the continuum flow with no-slip boundary conditions at the current operating conditions were demonstrated in [5]. After performing several grid and time-step independence tests, a grid structure with an element size of 0.35 mm and a time-step size of 0.01 s were selected for the simulation. The transport equations were solved using Ansys Fluent. The detailed explanations about the governing equations were provided in [6] and are omitted here for brevity.

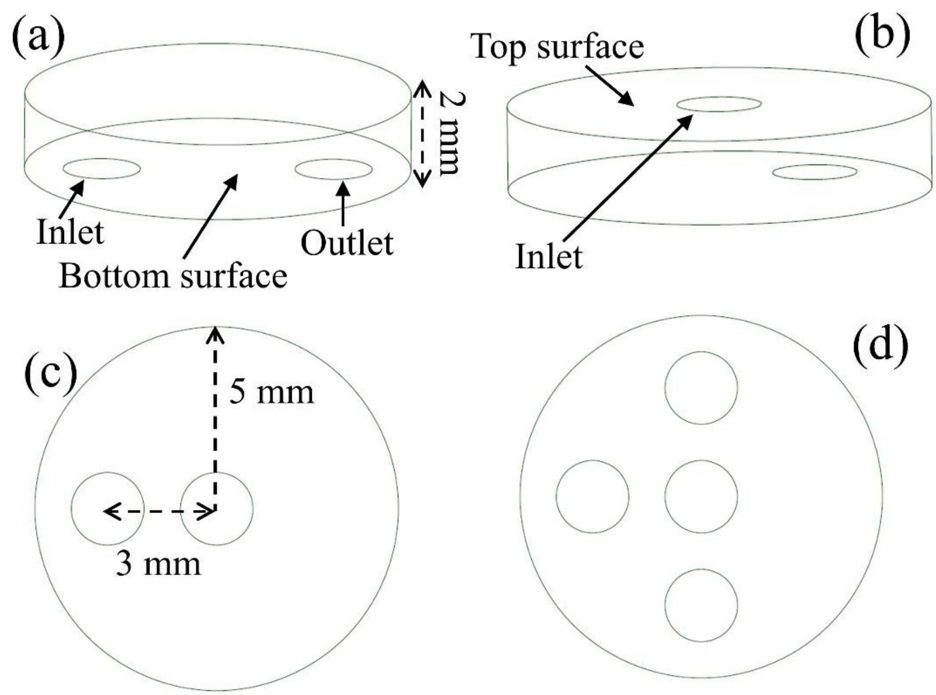

Fig. 1. (a) Three-dimensional view of the conventionally used reactor (Base); (b) Three-dimensional view of the top-inlet reactor with one inlet at the center of the reactor (TIR-1); (c) Top view of the top-inlet reactor with two inlets (TIR-2); (d) Top view of the top-inlet reactor with four inlets (TIR-4).

3. Results

The present study provides qualitative information about the dependency of purge durations on inlet flow rates in viscous flow reactors. Since the chemistry mechanism behind an ALD process is still not well understood, a quantitative prediction of the purge durations is not the focus of this research.

The mass fractions of all gaseous species inside a given reactor (Base) during one ALD cycle (i.e., 2.5 s) are illustrated in Fig. 2. When Ar is pulsed at each purge step, the remaining gases are removed from the reactor and mass fraction of Ar reaches to unity, which indicates the completion of purging. Fig. 3 illustrates the purge durations due to different inlet flow rates by assigning the inlet purge velocity of 4.2 m/s to an individual inlet manifold at each reactor. It is expected that an increase in the number of inlets leads to a shorter purge duration due to an increase in the flow rates. Basically, a sharper slope of the curve for the mass faction of Ar versus time indicates a shorter purging process. TIR-4 results in the shortest purge duration, and TIR-2 comes next. However, while the flow rates are the same in both Base and TIR-1, the latter reactor experiences longer purge durations, which most likely is due to different flow fields inside Base and TIR-1 resulting from the location of inlet manifold. The TIR-1 leads to an axisymmetric Ar injection and, in turn, a more uniform Ar distribution throughout the reactor volume, which may be the main reason for purging the reactor for a longer time.

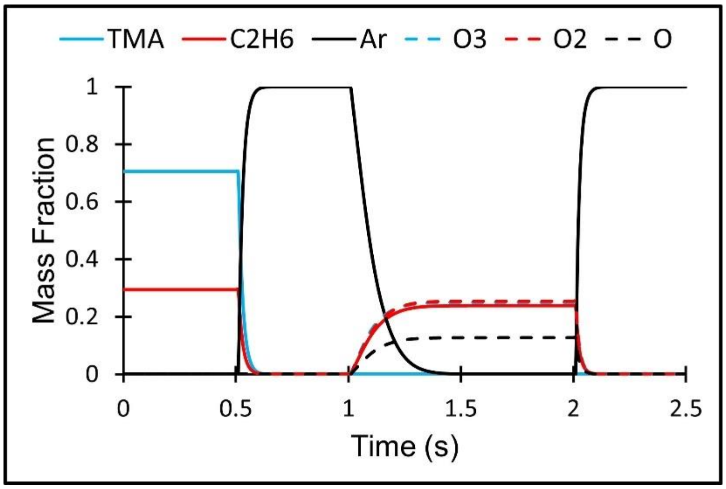

Fig. 2. Mass fractions of all gaseous species inside the Base reactor during one ALD cycle.

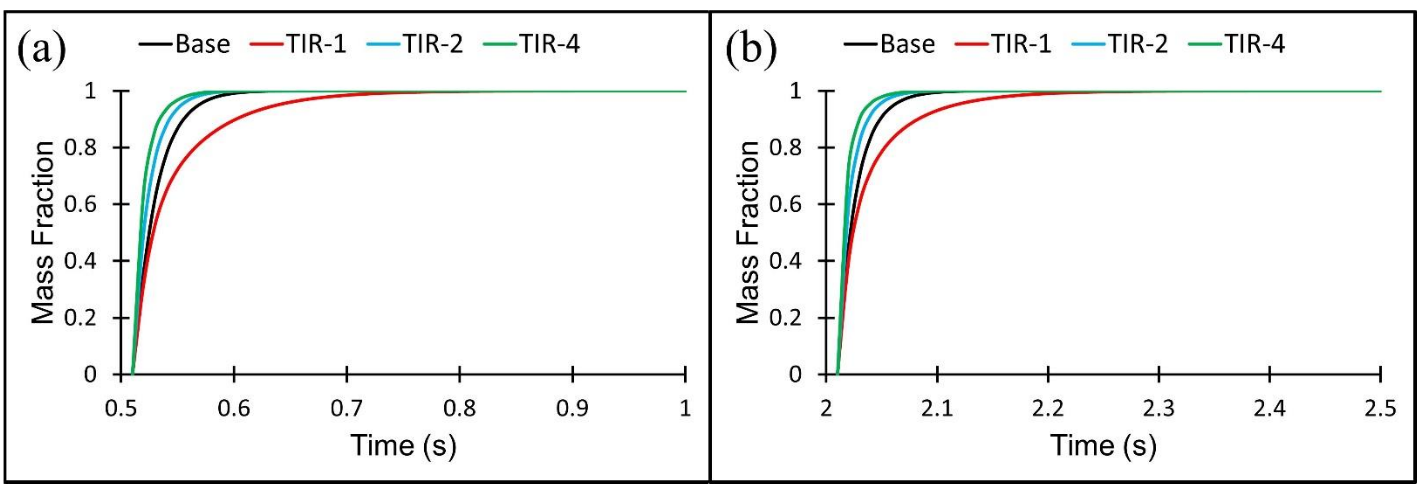

Fig. 3. Ar mass fractions at different inlet purge flow rates. Flow rates in TIR-4 and TIR-2 are four and two times larger than those in Base and TIR-1, respectively. (a) First purge; (b) Second purge.

Fig. 4 compares purge durations at a given flow rate. To achieve the same flow rates among all the reactors, the inlet purge velocity for TIR-4 and TIR-2 is reduced to 1.05 m/s and 2.1 m/s, respectively, and the inlet purge velocity at TIR-1 and Base is maintained at 4.2 m/s. The negligible differences in purge durations among TIR-4, TIR-2 and Base indicate an independency of purge durations from the number of inlets at a given flow rate. However, similar to Fig. 3, the purge durations in TIR-1 are longer than those in other reactors, which can be due to flow field resulting from the axisymmetric

Ar injection in TIR-1. More detailed flow field analysis in future research is required to address this finding in TIR-1. Combination of findings in Figs. 3 and 4 along with detailed research as a supplement to the present study will provide better understanding about designing efficient viscous flow reactors to reduce both purge durations and purge gas consumption.

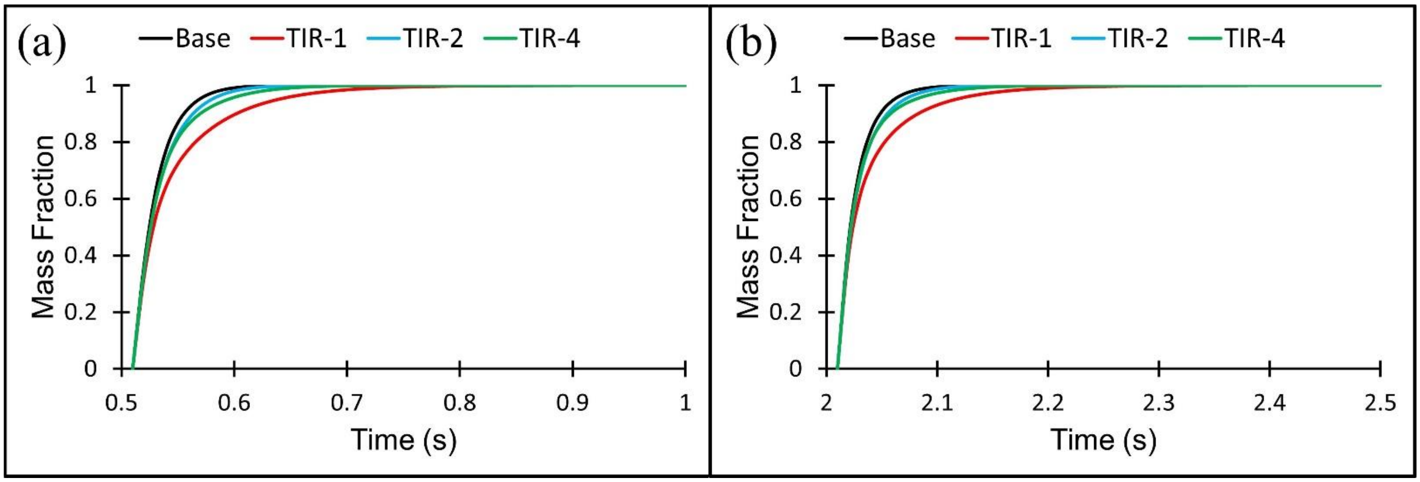

Fig. 4. Ar mass fractions at the same purge flow rate in different reactors. (a) First purge; (b) Second purge.

4. Conclusion

An increase in the inlet purge flow rate shortens the purge duration. However, at a given flow rate, changes in purge durations due to the number of inlets are negligible. Nevertheless, due to axisymmetric flow injection in TIR-1, the purge durations in this reactor are longer than other reactors. Although by increasing the flow rates, TIR-4 and TIR-2 reduce purge durations, the amount of consumed Ar at these reactors should be calculated to ensure the new reactors are economical because it is possible that despite a faster purging by TIR-4 and TIR-2, the amount of consumed Ar in these reactors is significantly higher than that in the Base (i.e., conventionally used reactor).

Acknowledgements

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