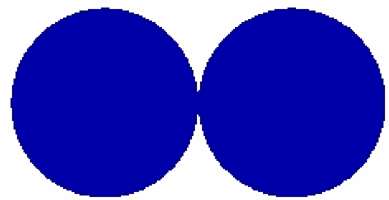
# The role of inertia in coalescence of drops in liquid-liquid emulsions



**Vishrut Garg**, Krishnaraj Sambath[*], Sumeet S. Thete[†], Hariprasad Subramani[*] and Osman A. Basaran

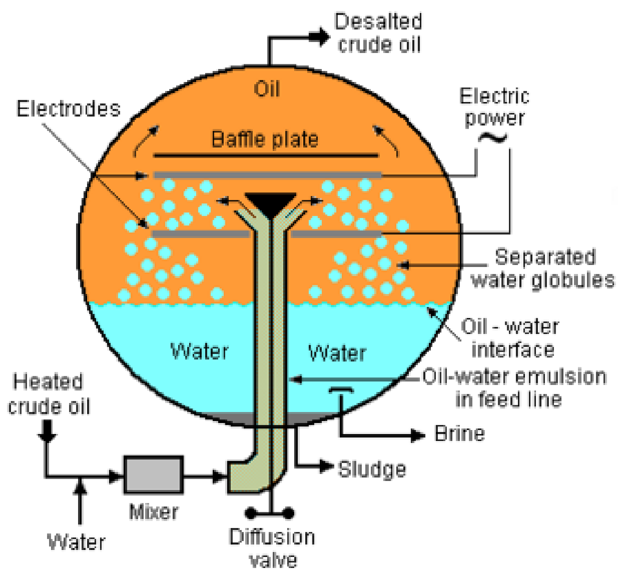
Davidson School of Chemical Engineering, Purdue University

Work supported/sponsored by:
Chevron, P2SAC and Purdue CoE

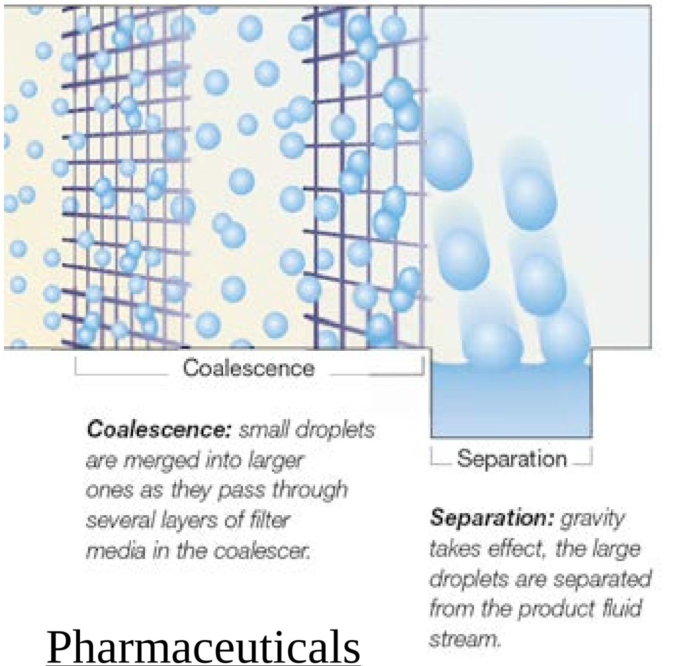
*Now at Chevron Corporation
† Now at Air Products and Chemicals, Inc.

# Liquid-liquid emulsions are ubiquitous

Crude oil desalters




Coalescers for L/L extraction



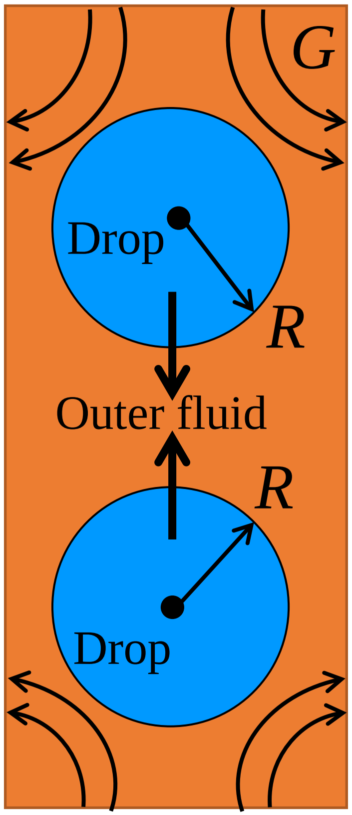

Food products


Pharmaceuticals


Intravenous lipid emulsions


Ointments

# The process of drop coalescence




- Two drops in another immiscible outer liquid separated by a certain distance are driven towards each other by an external force

- External force can be due to:
  1. Gravity
  2. Electric fields
  3. Flow imposed on outer fluid

- A key parameter in flow-induced coalescence of drops is the capillary number:

$$Ca = \frac{\mu_2 G R}{\sigma}$$

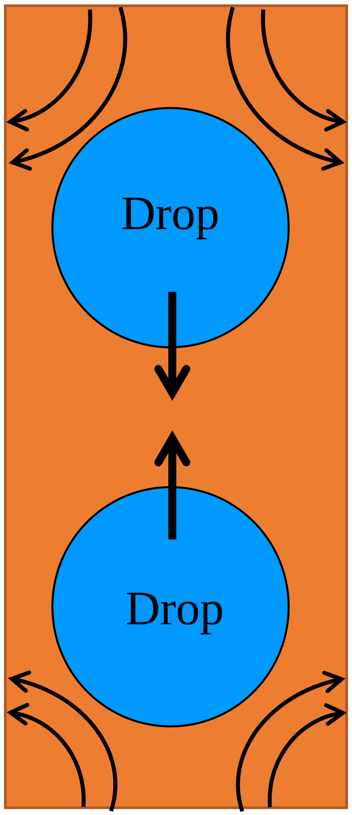
$\mu_2$ - viscosity of the outer liquid

$G$ - strain rate of the imposed flow

$\sigma$ - interfacial tension of the liquid-liquid interface

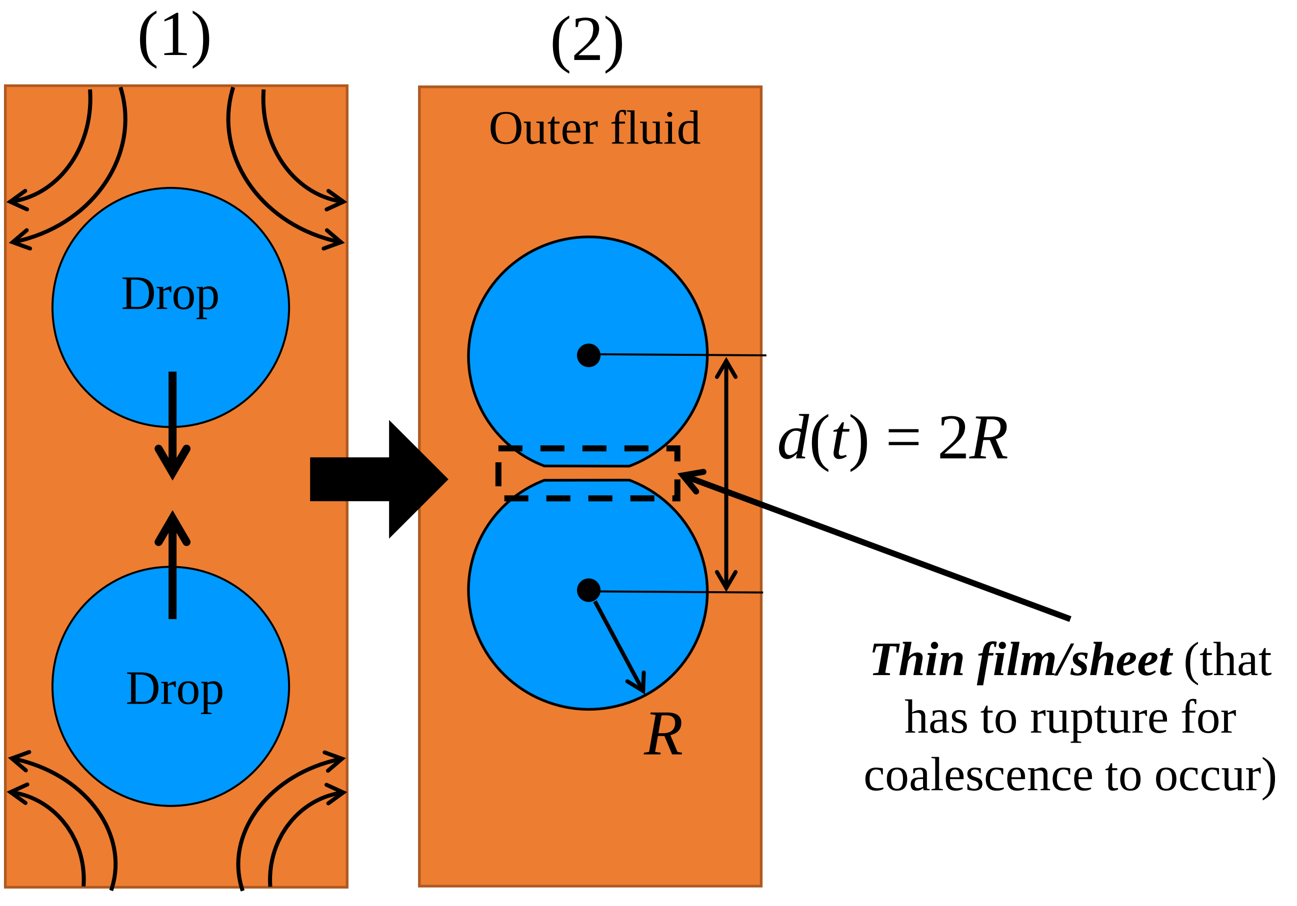
# The process of drop coalescence (1)

(1)

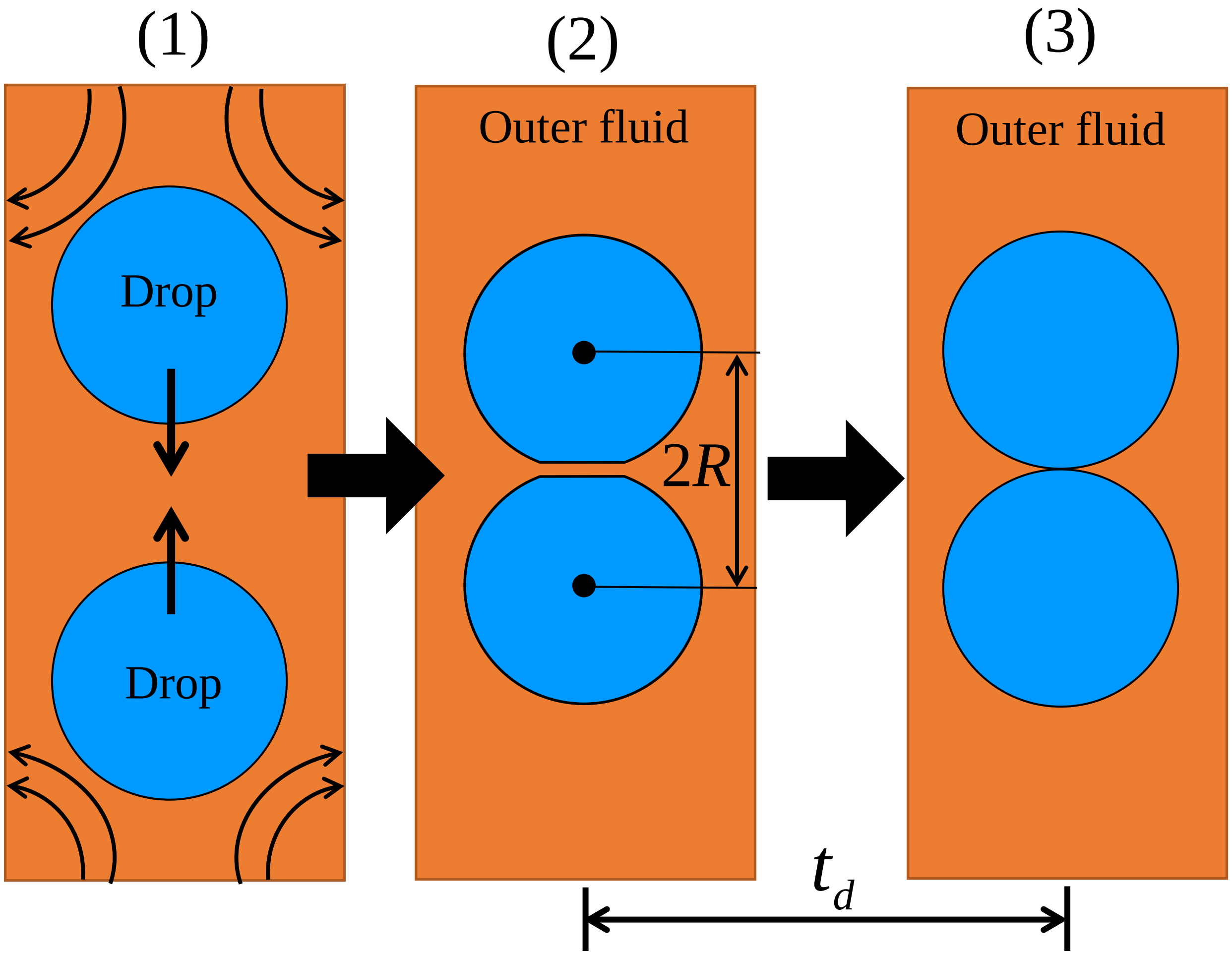



Two drops in an immiscible outer liquid are pushed or driven towards each other due to an imposed flow

# The process of drop coalescence (2)



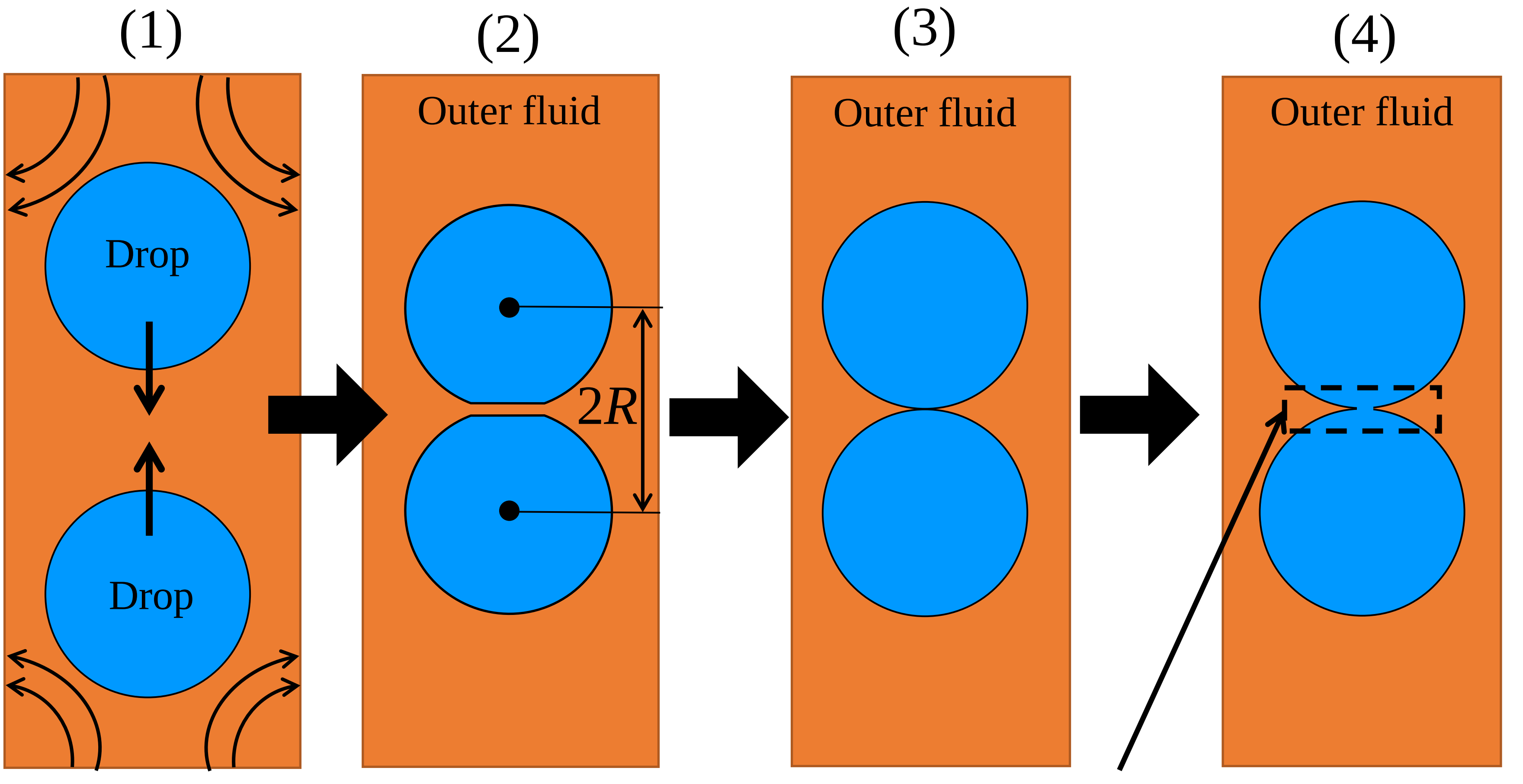
# The process of drop coalescence (3)




The interfaces become close enough for attractive van der Waals forces to cause coalescence of the two drops

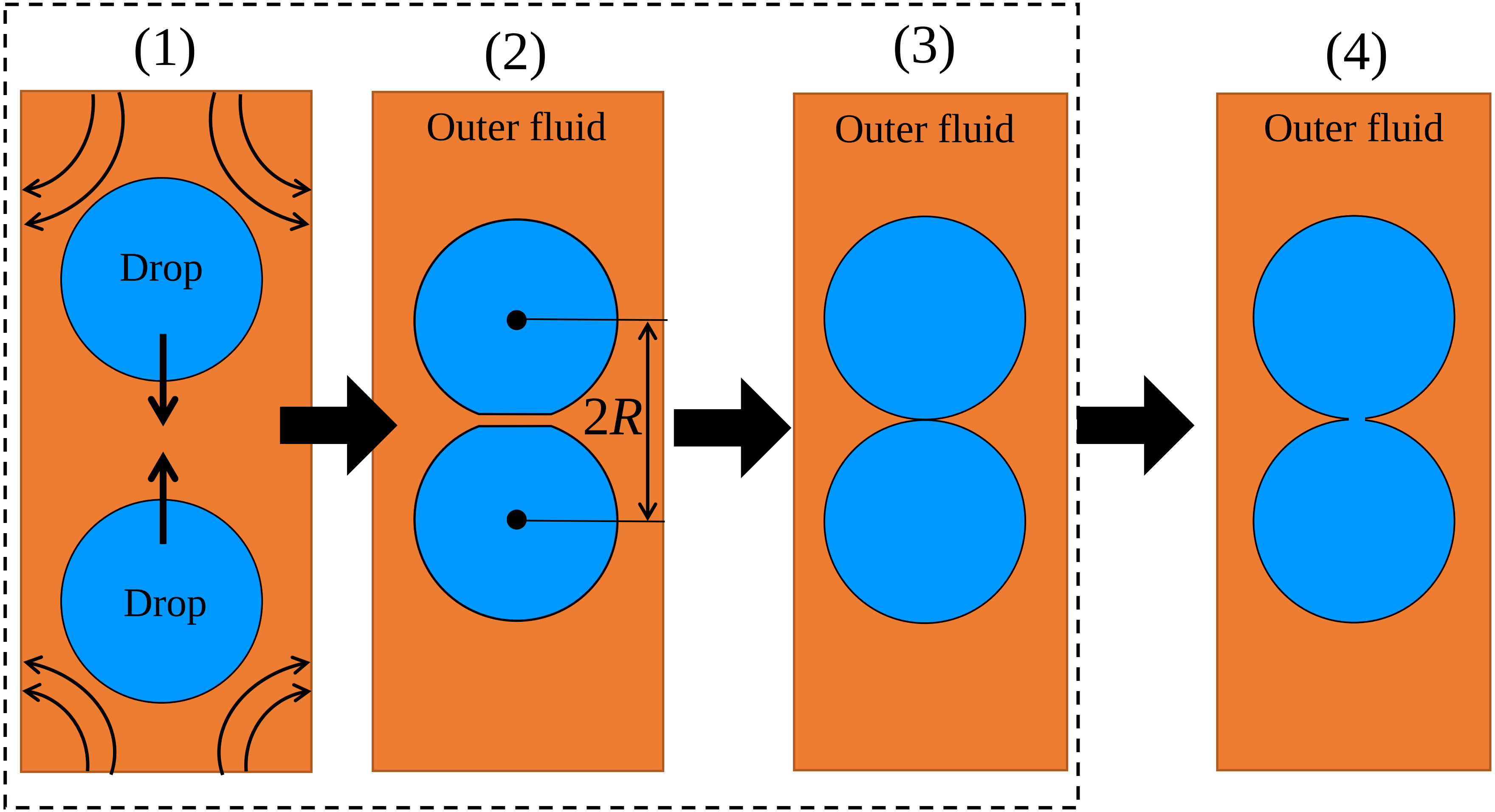
Time elapsed between time instants 2 and 3 is known as “drainage time”

# The process of drop coalescence (4)



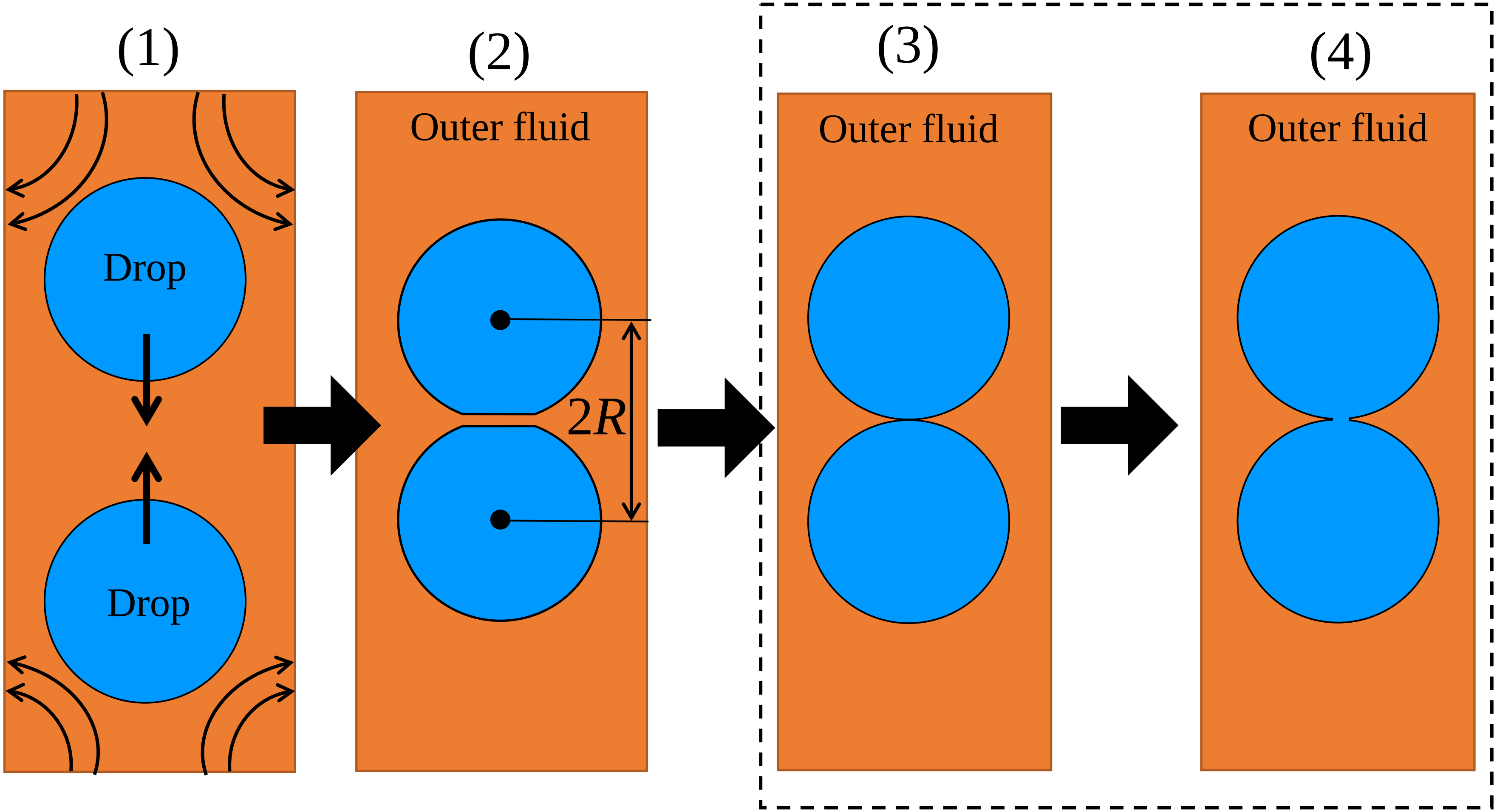

***Thin film/sheet*** with a growing hole (or a growing bridge that connects the two drops/bubbles)

# **Pre**-coalescence (subject of this talk)




***Singularity at the end of the process***

# **Post**-coalescence




***Singularity at beginning of process (see papers by Eggers, Stone, Lister, Nagel, Basaran, …)***

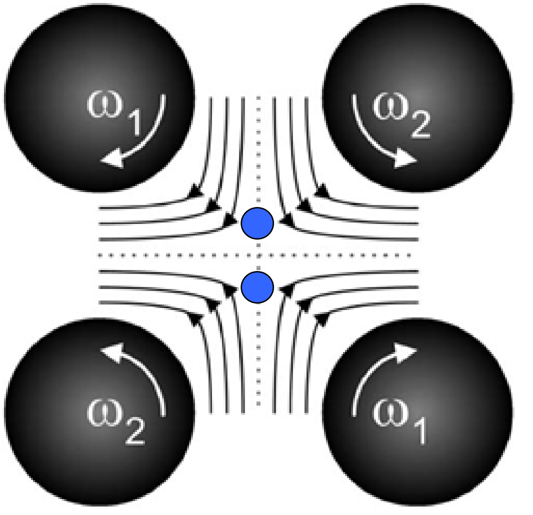
# Previous works on pre-coalescence

## L.G. Leal and coworkers (2001 – present)

- Extensive experiments using Taylor's four roll mill
- Boundary integral (BI) simulations
- Scaling theory for drainage time:

$$t_d \sim Ca^{4/3}$$

for drops with radius larger than 27 $\mu$m



Taylor's four roll mill

## M. Loewenberg and coworkers (2004 – 2013)

- BI simulations (role of internal flows in arresting drop coalescence)

## H. Meijer and coworkers (2006 – 2011)

- BI simulations
- Scaling theory

All previous studies considered **creeping (Stokes) flow** conditions

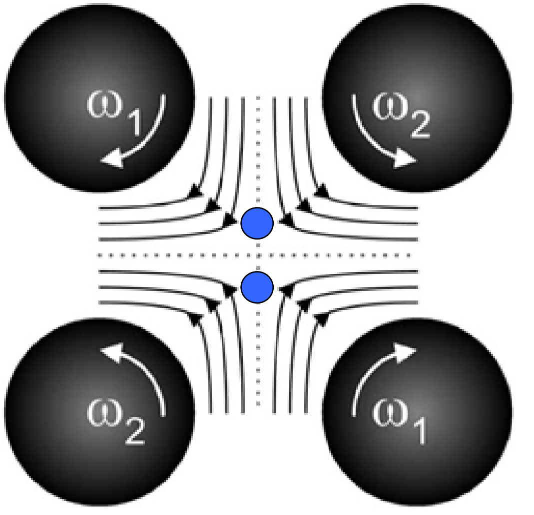
# Previous works on pre-coalescence

L.G. Leal and coworkers (2001 – present)

- Extensive experiments using Taylor's four roll mill
- Boundary integral (BI) simulations
- Scaling theory for drainage time:

$$t_d \sim Ca^{4/3}$$

for drops with radius larger than 27 $\mu$m




Taylor's four roll mill

M. Loewenberg and coworkers (2004 – 2013)

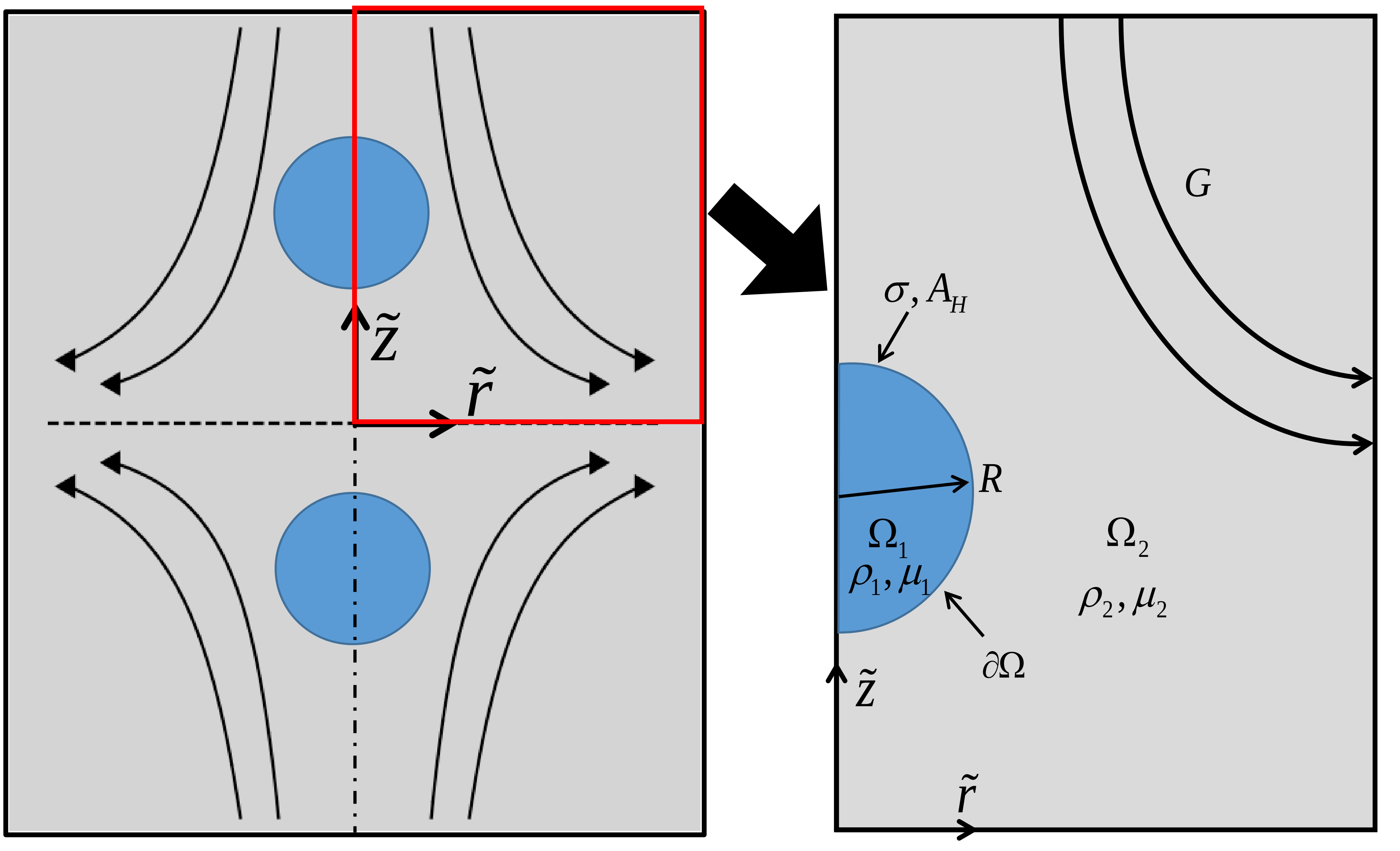
- BI simulations (role of internal flows in arresting drop coalescence)

H. Meijer and coworkers (2006 – 2011)

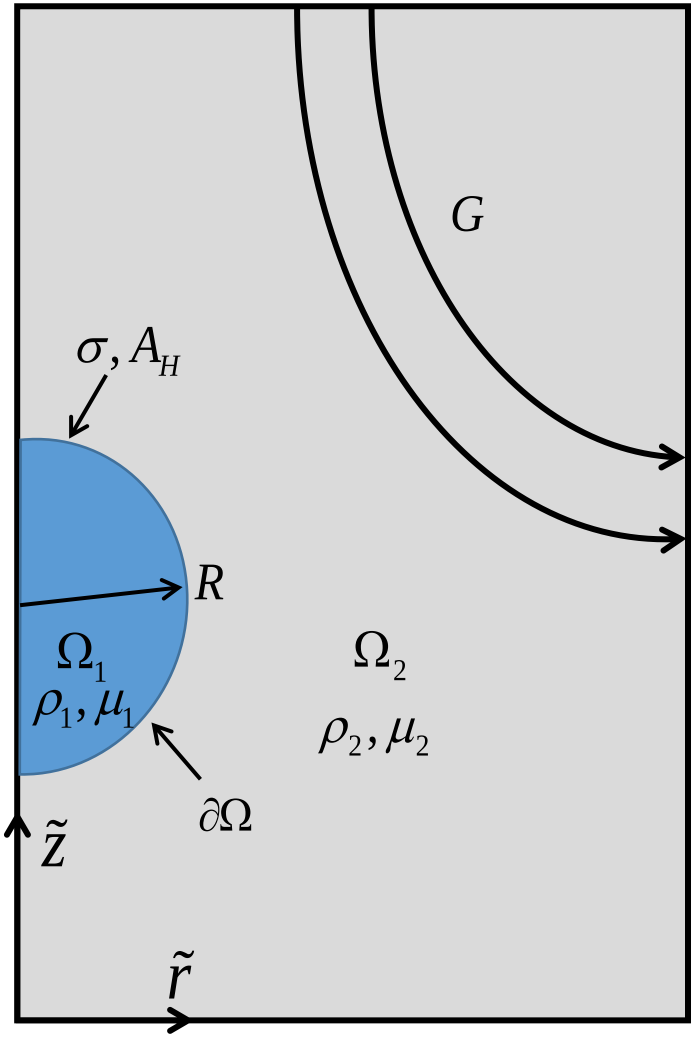
- BI simulations & scaling theory

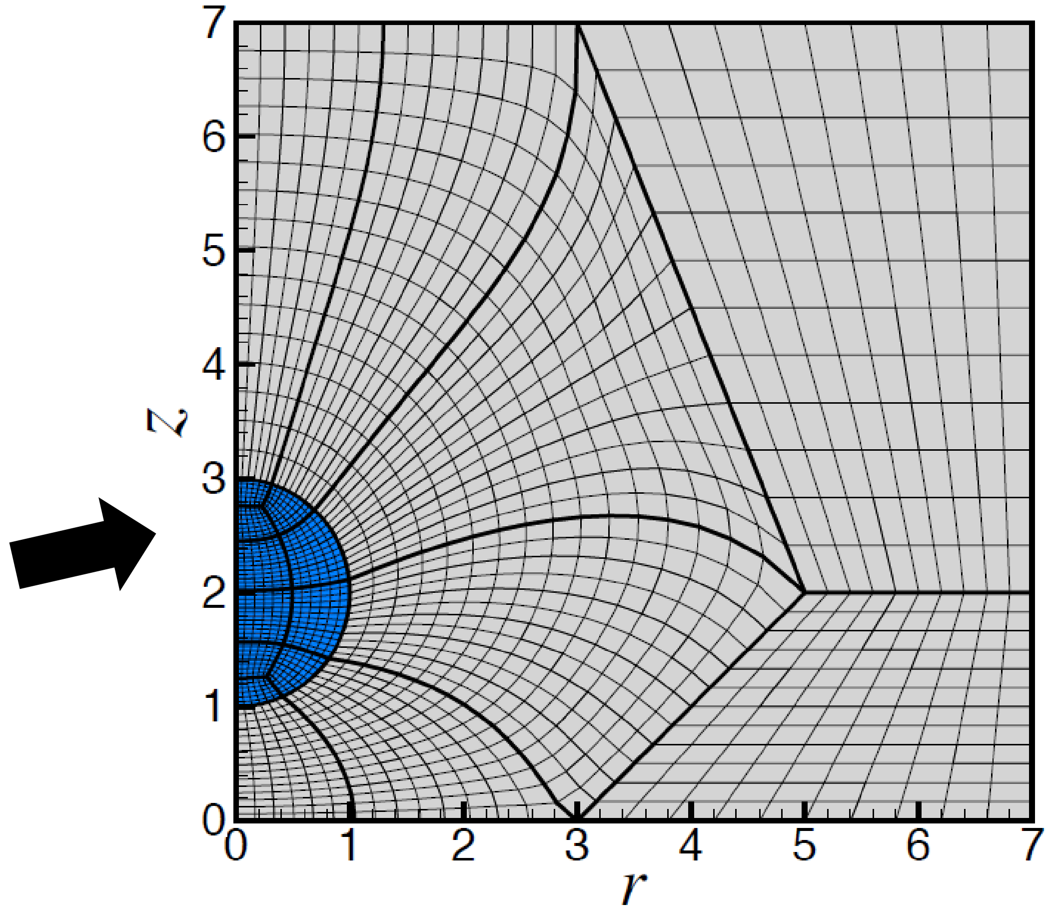
We study drop coalescence dynamics when **inertial effects are significant**

# Numerically solving the 3D axisymmetric problem (1)

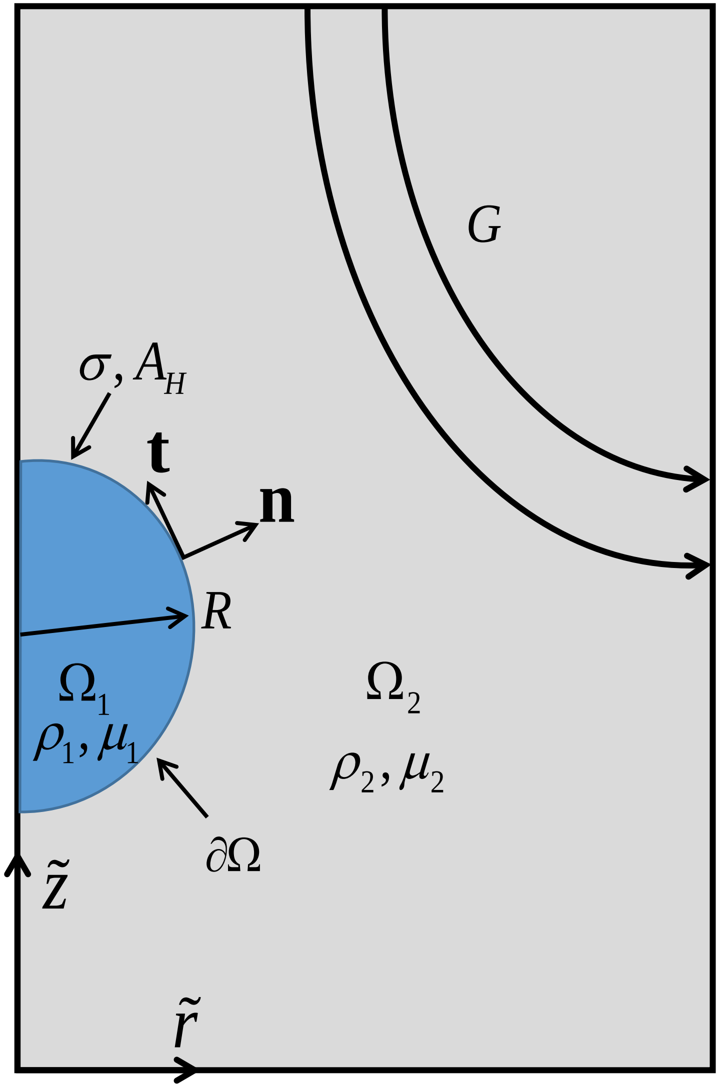


# Numerically solving the 3D axisymmetric problem (2)



# Mathematical formulation




**Navier-Stokes system**

$$\left.\begin{aligned} \nabla \cdot \tilde{\mathbf{v}}_i &= 0 \\ \rho_i \left( \frac{\partial \tilde{\mathbf{v}}_i}{\partial \tilde{t}} + \tilde{\mathbf{v}}_i \cdot \tilde{\nabla} \tilde{\mathbf{v}}_i \right) &= \tilde{\nabla} \cdot \tilde{\mathbf{T}}_i \end{aligned}\right\} \Omega_1 \cup \Omega_2$$

where $\tilde{\mathbf{T}}_i = -\tilde{p}_i \mathbf{I} + \mu_i \left[ \left( \tilde{\nabla} \tilde{\mathbf{v}}_i \right) + \left( \tilde{\nabla} \tilde{\mathbf{v}}_i \right)^T \right]$

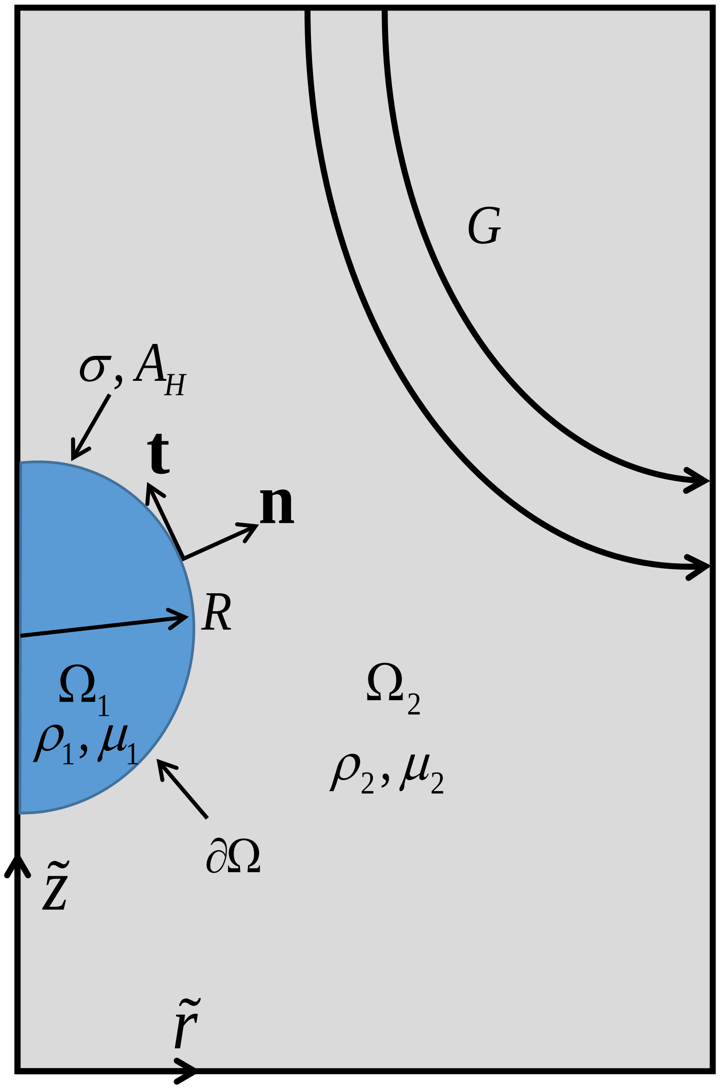
**Boundary conditions**

$$\left.\begin{aligned} \mathbf{n} \cdot \left( \tilde{\mathbf{v}}_i - \tilde{\mathbf{v}}_{s,i} \right) &= 0 \\ \mathbf{n} \cdot \left[ \tilde{\mathbf{T}}_i \right]_1^2 &= \left( 2\sigma \tilde{H} - \frac{A_H}{6\pi \tilde{h}(\tilde{\mathbf{x}})^3} \right) \mathbf{n} \end{aligned}\right\} \partial\Omega$$

where $h(\mathbf{x})$ – vertical separation between drops' interfaces

For typical liquids, $A_H = 10^{-21}$ to $10^{-18}\ J$

# Mathematical formulation




**Navier-Stokes system**

$$\left.\begin{aligned} \nabla \cdot \tilde{\mathbf{v}}_i &= 0 \\ \rho_i \left( \frac{\partial \tilde{\mathbf{v}}_i}{\partial \tilde{t}} + \tilde{\mathbf{v}}_i \cdot \tilde{\nabla} \tilde{\mathbf{v}}_i \right) &= \tilde{\nabla} \cdot \tilde{\mathbf{T}}_i \end{aligned}\right\} \quad \Omega_1 \cup \Omega_2$$

where $\tilde{\mathbf{T}}_i = -\tilde{p}_i \mathbf{I} + \mu_i \left[ \left( \tilde{\nabla} \tilde{\mathbf{v}}_i \right) + \left( \tilde{\nabla} \tilde{\mathbf{v}}_i \right)^T \right]$

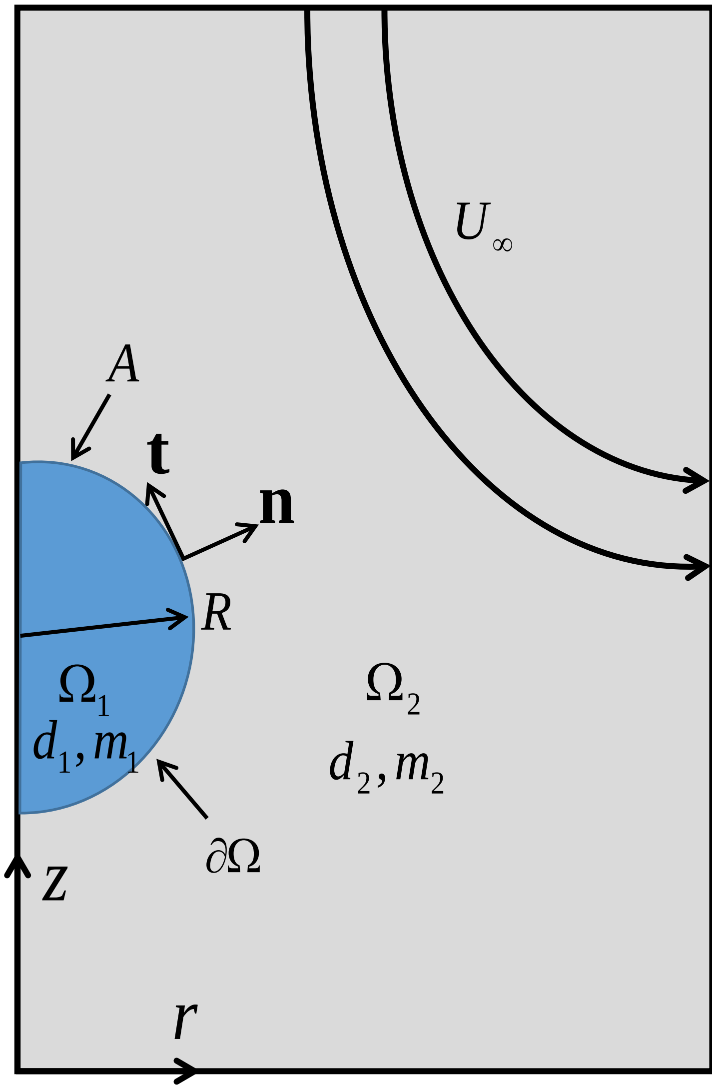
**Boundary conditions**

$$\left.\begin{aligned} \mathbf{n} \cdot \left( \tilde{\mathbf{v}}_i - \tilde{\mathbf{v}}_{s,i} \right) &= 0 \\ \mathbf{n} \cdot \left[ \tilde{\mathbf{T}}_i \right]_1^2 &= \left( 2\sigma \tilde{H} - \frac{A_H}{6\pi \tilde{h}(\tilde{\mathbf{x}})^3} \right) \mathbf{n} \end{aligned}\right\} \quad \partial\Omega$$

**Imposed bi-axial extensional flow**

$$\tilde{\mathbf{v}}_2(\tilde{\mathbf{x}}) = G \left( \frac{\tilde{r}}{2} \mathbf{e}_r - \tilde{z} \mathbf{e}_z \right) \quad \text{when } |\tilde{\mathbf{x}}| \to \infty$$

# Non-dimensionalization



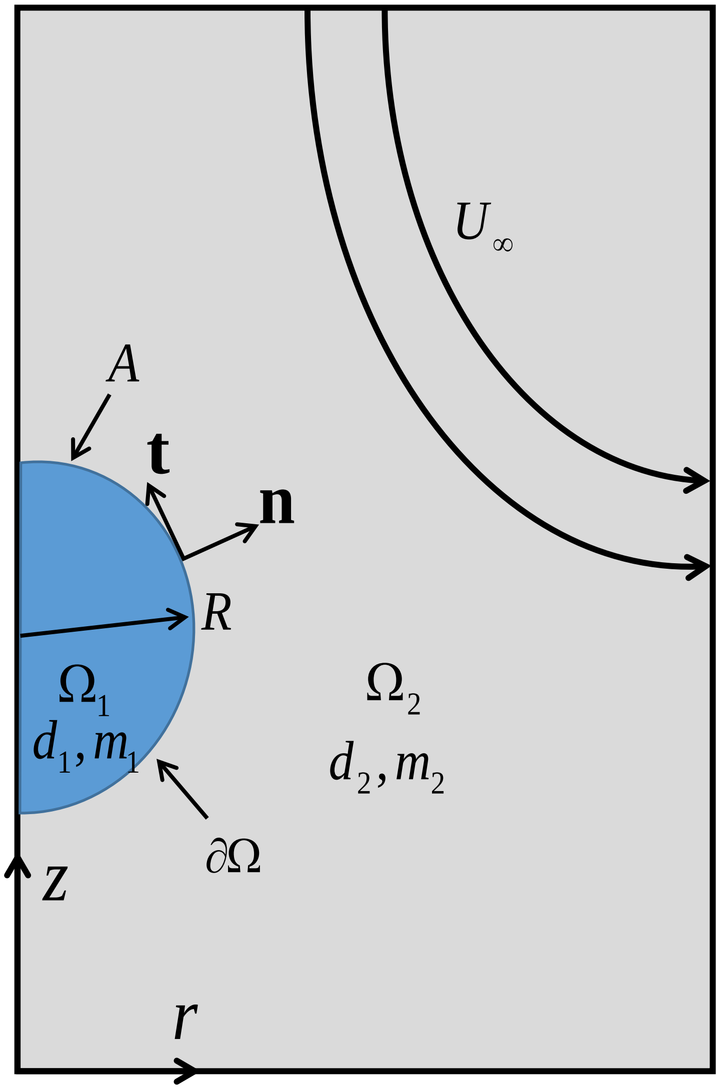

## Characteristic scales

$$l_c \equiv R\ ,\ t_c \equiv \left(\rho_1 R^3 / \sigma\right)^{1/2},\ v_c \equiv l_c / t_c\ ,\ \ p_c \equiv \sigma / R$$

| Key Dimensionless groups | | 1 mm water drops in oil |
|---|---|---|
| $U_\infty = G\left(\frac{\rho_1 R^3}{\sigma}\right)^{1/2}$ | Dimensionless velocity/strain rate | $10^{-4}$ to 0.1 |
| $A = \frac{A_H}{6\pi\sigma R^2}$ | Van der Waals force / Surface tension force | $10^{-13}$ |
| $m_i = \mu_i / \mu_1$ $\quad_{i=1,2}$ | Viscosity ratio | 10 |
| $d_i = \rho_i / \rho_1$ | Density ratio | 0.9 |
| $Oh = \frac{\mu_1}{\left(\rho_1 R\sigma\right)^{1/2}}$ | Ohnesorge number or dimensionless viscosity | 0.006 |

| | |
|---|---|
| $Ca = \frac{\mu_2 G R}{\sigma} \rightarrow$ $10^{-4}$ to 0.1 | $Re = \frac{G R^2 \rho_2}{\mu_2} \rightarrow$ 0.01 to 10 |

# Non-dimensionalization (2)



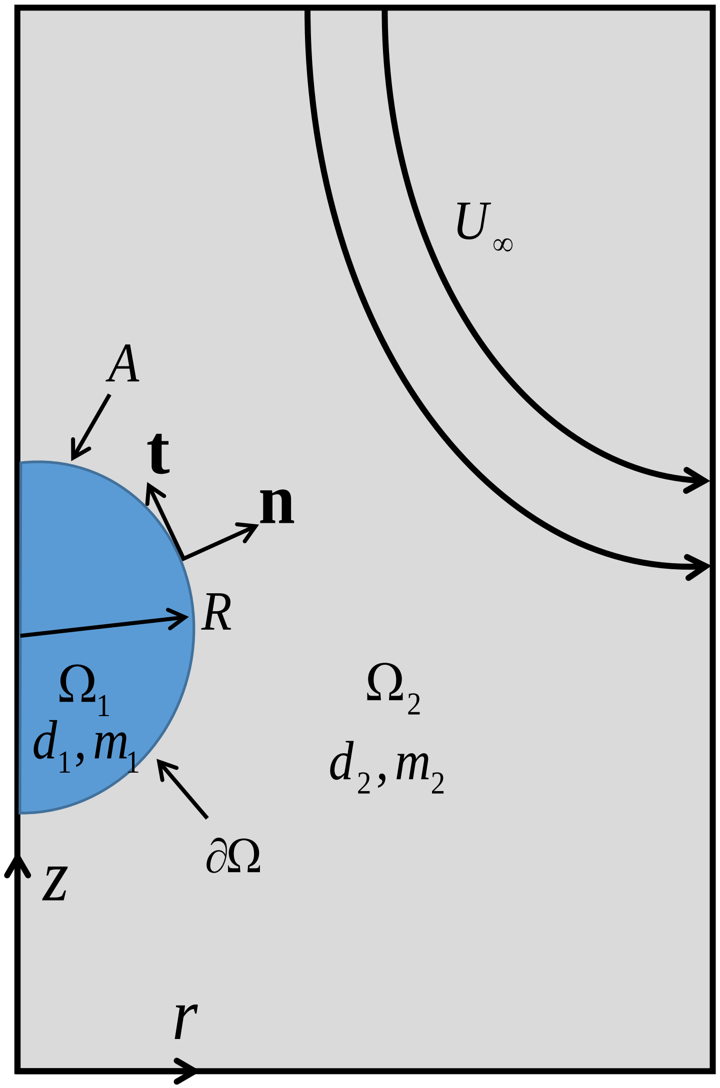

<u>Capillary number</u>

$$Ca = \frac{\mu_2 G R}{\sigma} = m_2 U_\infty Oh$$

<u>Reynolds number</u>

$$Re = \frac{\rho_2 (GR) R}{\mu_2} = \frac{U_\infty d_2}{m_2 Oh}$$

# Numerical simulations



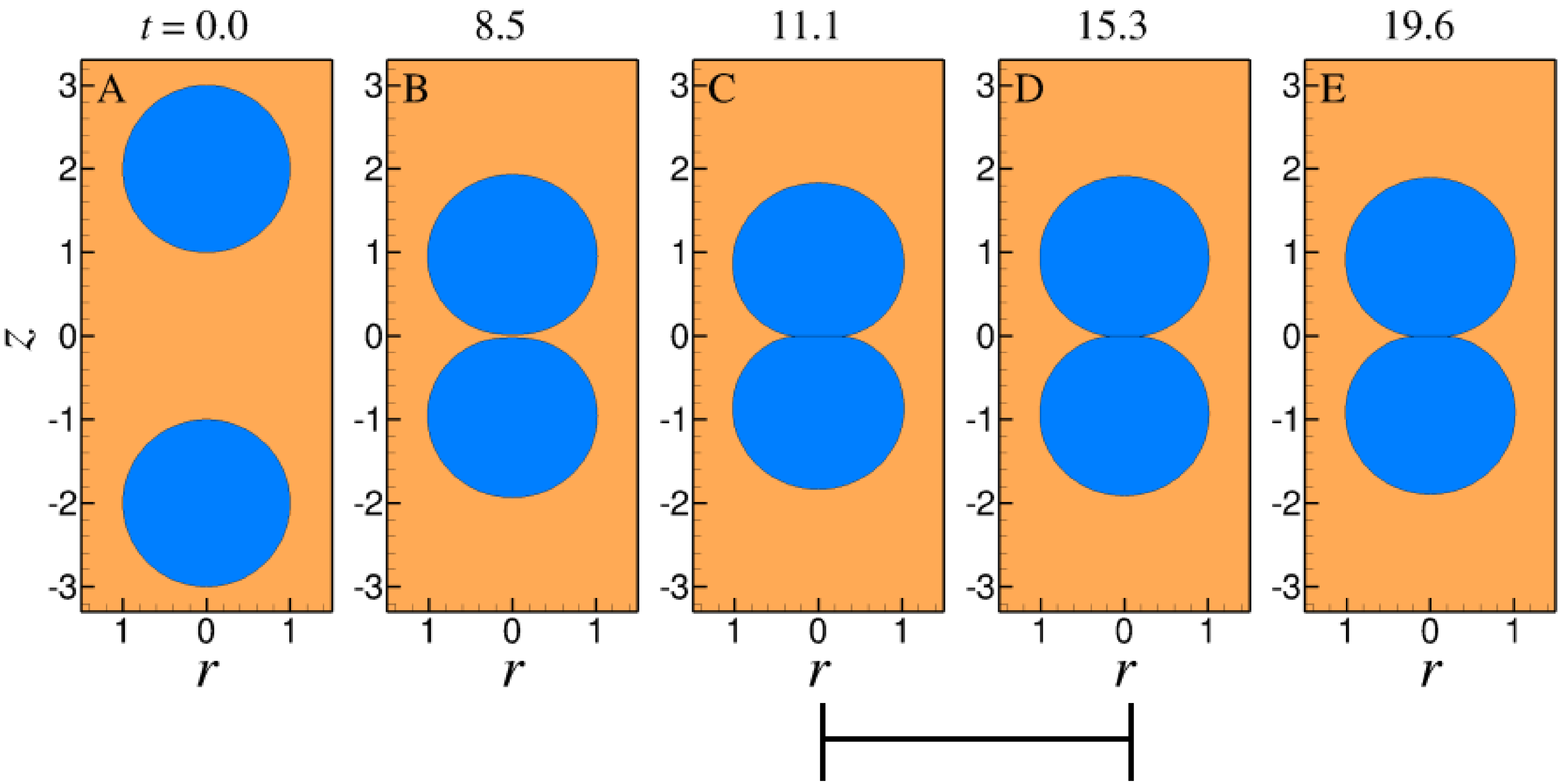

- Governing equations are solved numerically using a fully implicit **method of lines** (MOL), **arbitrary Lagrangian-Eulerian** (ALE) algorithm
- **Galerkin finite element method** (G/FEM) is used for spatial discretization
- **Adaptive time-difference** method is used for time integration
- **Elliptic mesh generation technique** is used to construct highly adaptive dynamic meshes which ensures accuracy over length scales that differ by **six (or more) orders of magnitude** **(this is the reason why commercial codes do not do a good job in solving this problem!)**

# Summary of what was done

- Drops bounce or rebound when inertia is not neglected
- *Unlike in experiments*, inertia can be "artificially" turned off in both fluids or just one of the two fluids
- Computations show that drop inertia is key to causing drop rebound
- Due to time limitations, we will skip to the end of the presentation
- The entire presentation is posted at the P2SAC web site for your perusal and study at your convenience
- A second presentation will also be posted on line that details what happens when surfactants or surface-active species are present

# Drop rebound occurs for *Re* = 1 flows

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$$

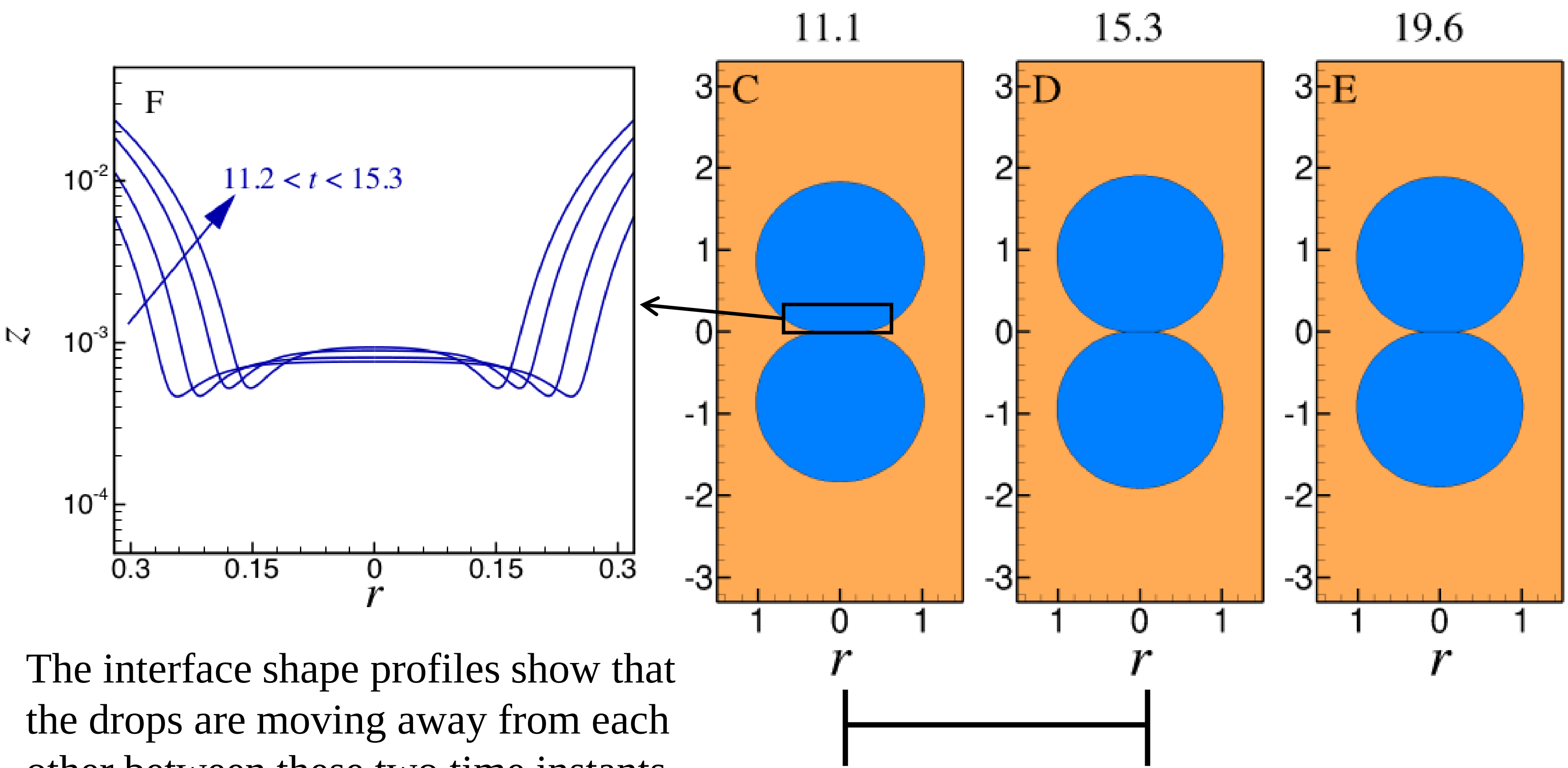



The Reynolds number for this case is *Re* = 1

The drops rebound during the mid stages before coalescing on second approach

# Drop rebound is evident from interface shapes

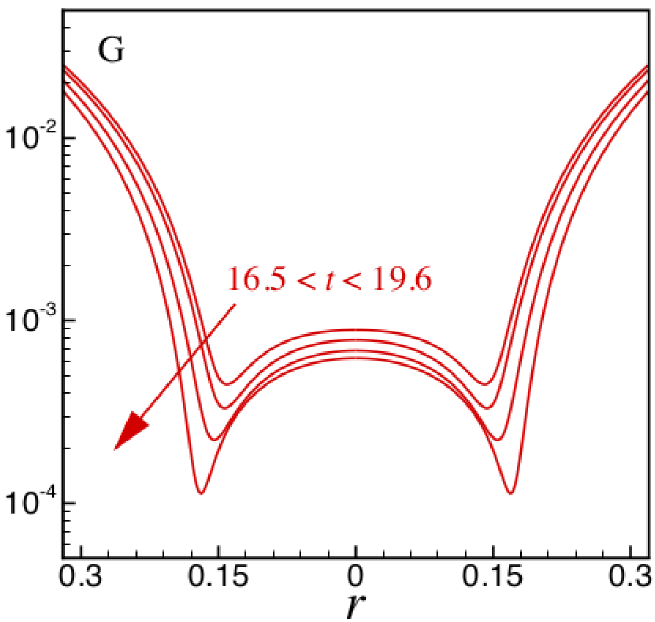
$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$$

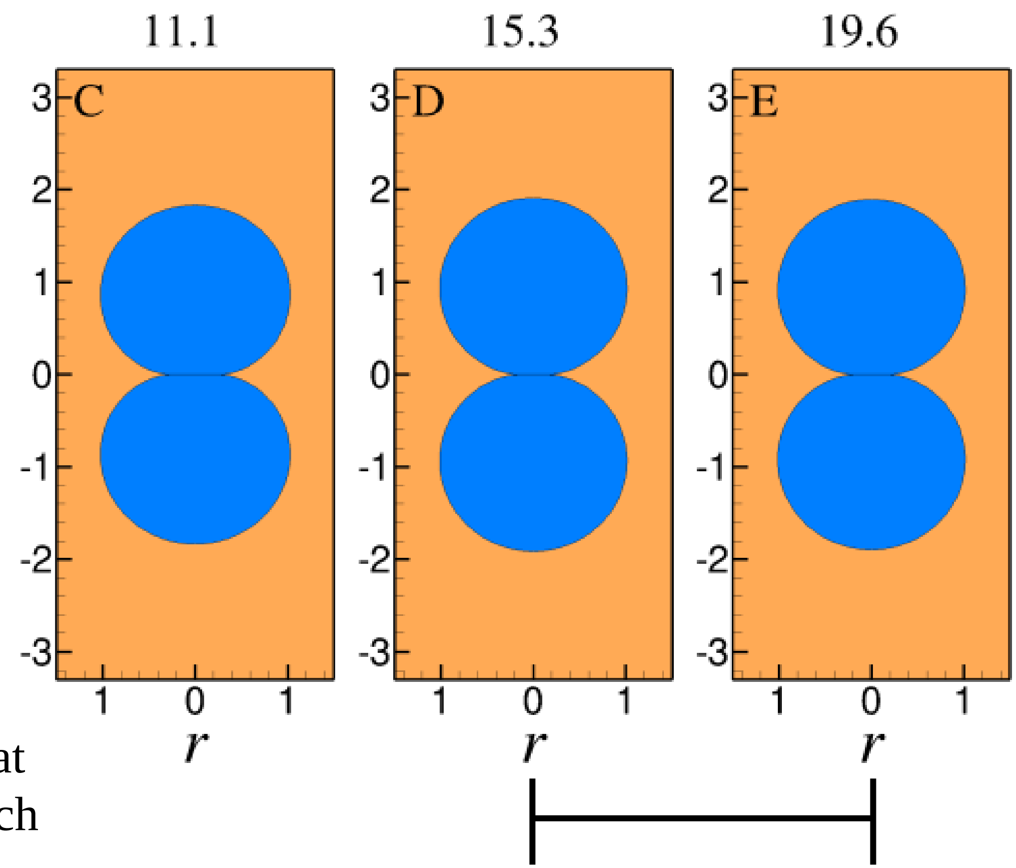



The interface shape profiles show that the drops are moving away from each other between these two time instants (so do their center-of-mass velocities)

# Coalescence occurs on second approach

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$$



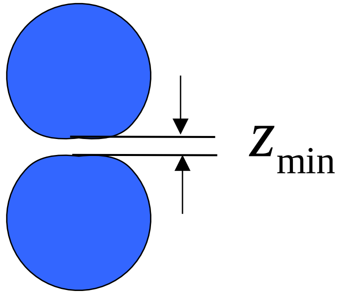



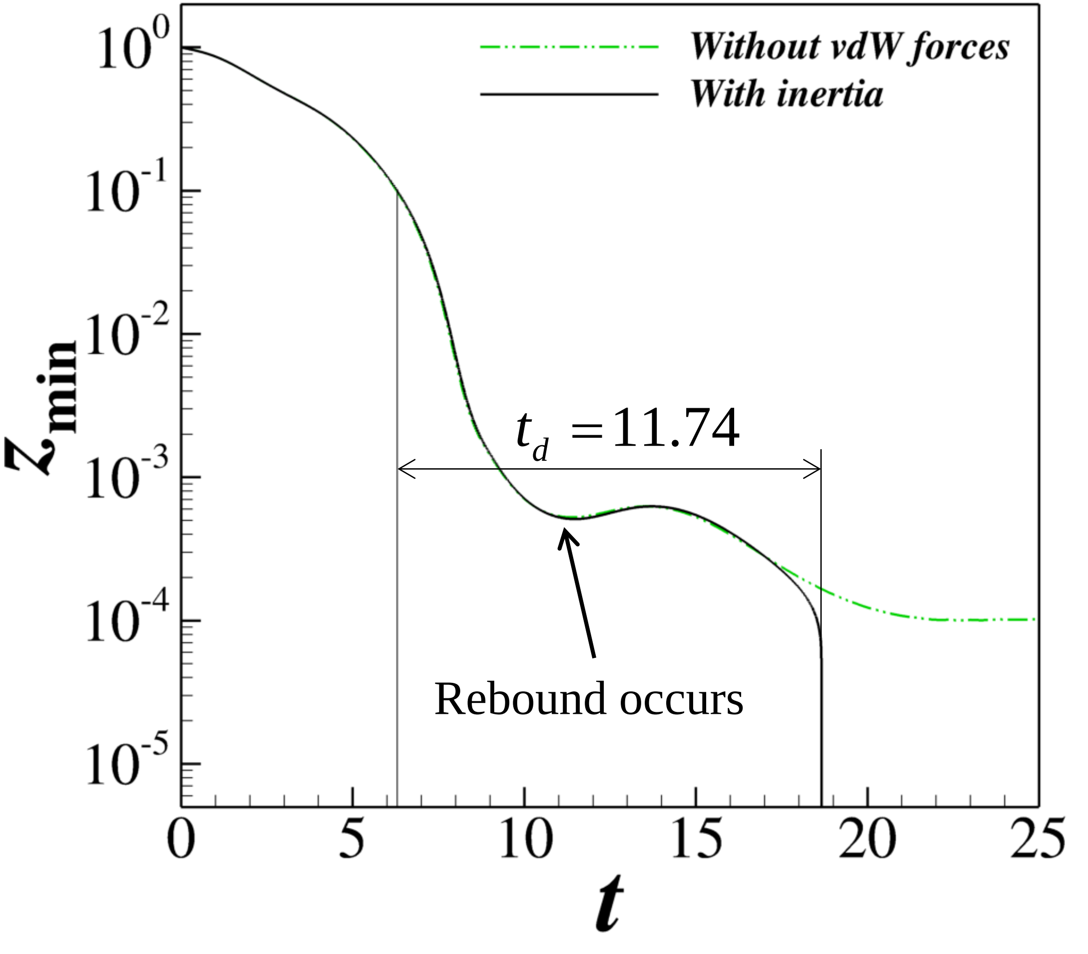

The interface shape profiles show that the drops coalesce on second approach as they get close enough for van der Waals forces to become significant

# Inclusion of inertia is essential for drop rebound

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$$

<u>*Minimum axial separation*</u>

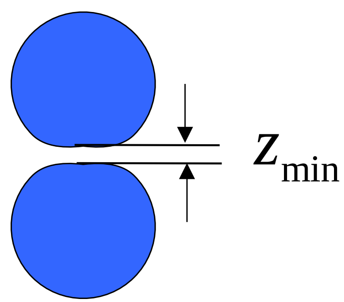


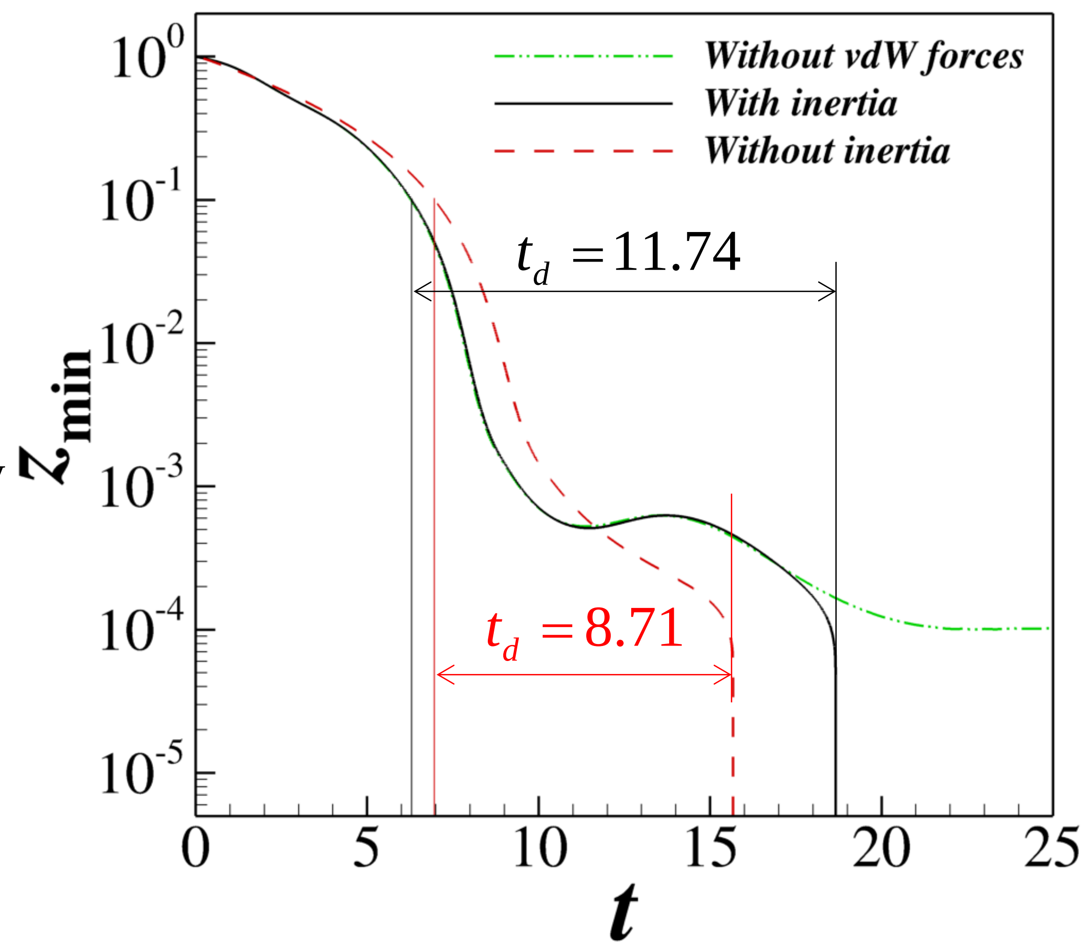



# Inclusion of inertia is essential for drop rebound

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99\times10^{-11}$$

*Minimum axial separation*

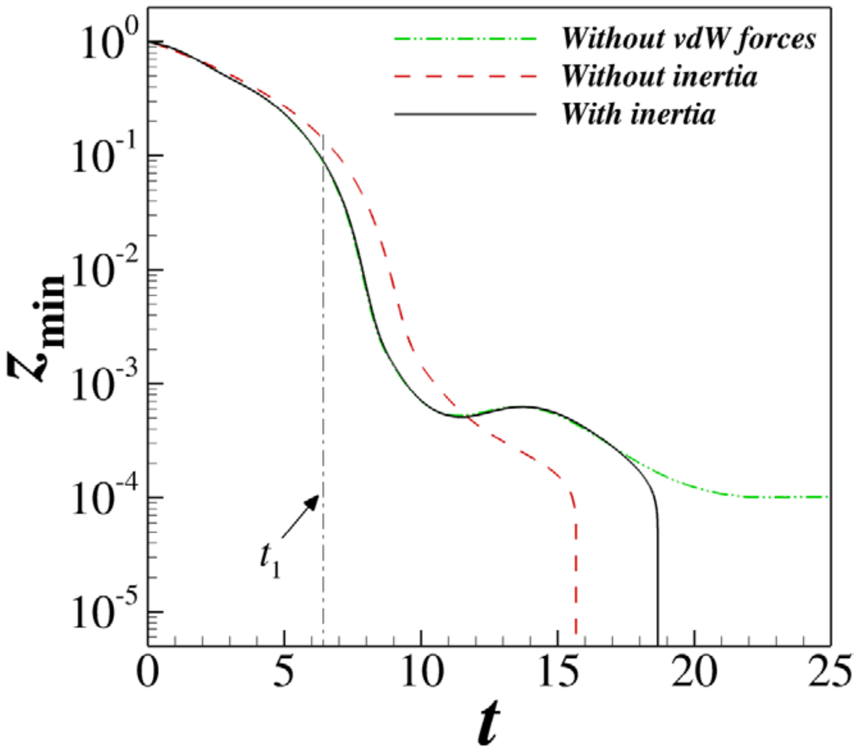


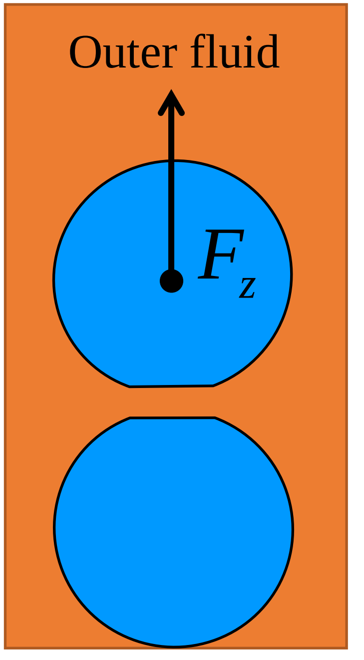




- If inertial terms are artificially "turned off" in the governing equations, the drops coalesce on first approach without rebound
- Drainage times are smaller if inertia is neglected

# Why does the presence of inertia cause rebound?

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99\times 10^{-11}$$



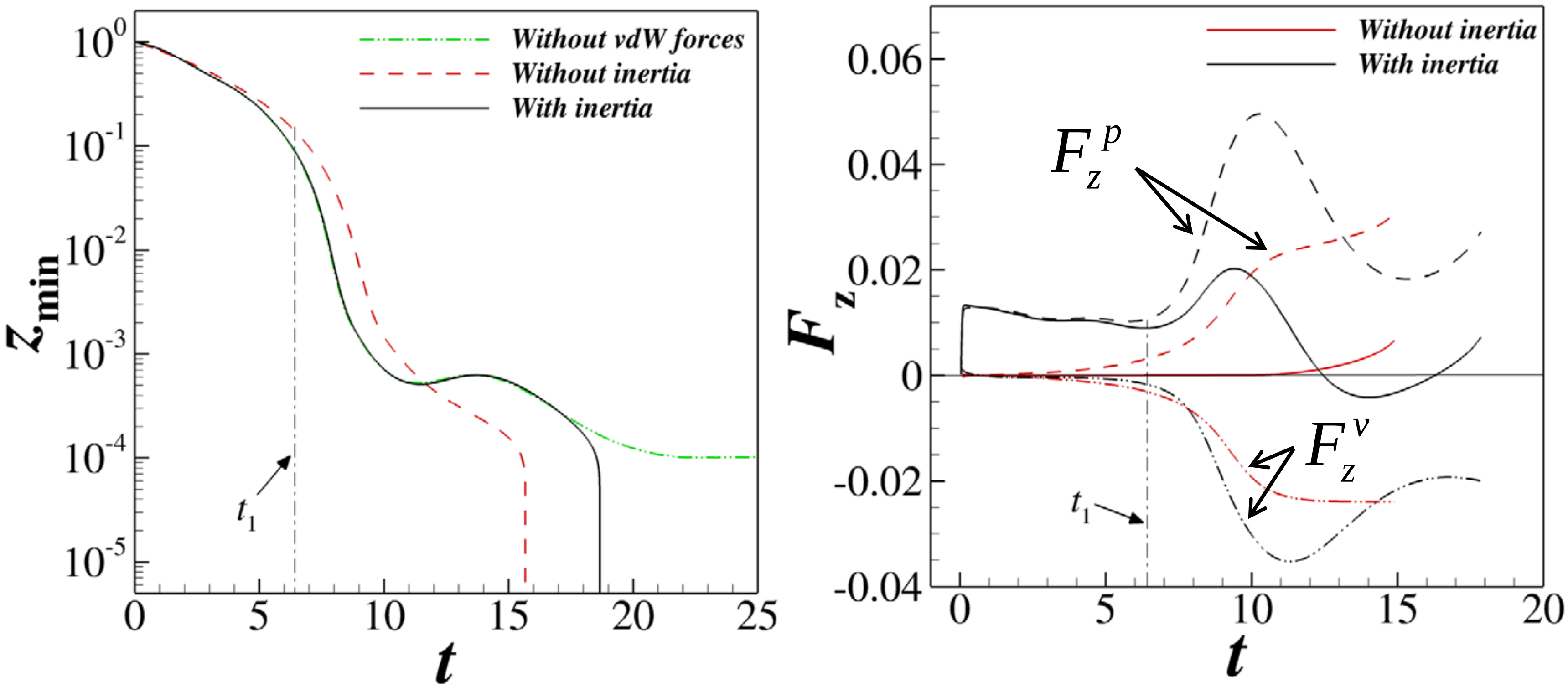




- We compute the net force exerted by the outer fluid on the top drop in the axial direction
- Contributions to this force due to pressure in the film and that due to viscous stress

$$F_z = \int_S \mathbf{n}\cdot\mathbf{T}_2\cdot\mathbf{e}_z dS = \underbrace{\int_S \mathbf{n}\cdot(-p_2\mathbf{I})\cdot\mathbf{e}_z dS}_{F_z^p} + \underbrace{\int_S \mathbf{n}\cdot m_2 Oh\left[\nabla\mathbf{v}_2 + (\nabla\mathbf{v}_2)^T\right]\cdot\mathbf{e}_z dS}_{F_z^v}$$

# Opposing forces acting on the drop

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$$

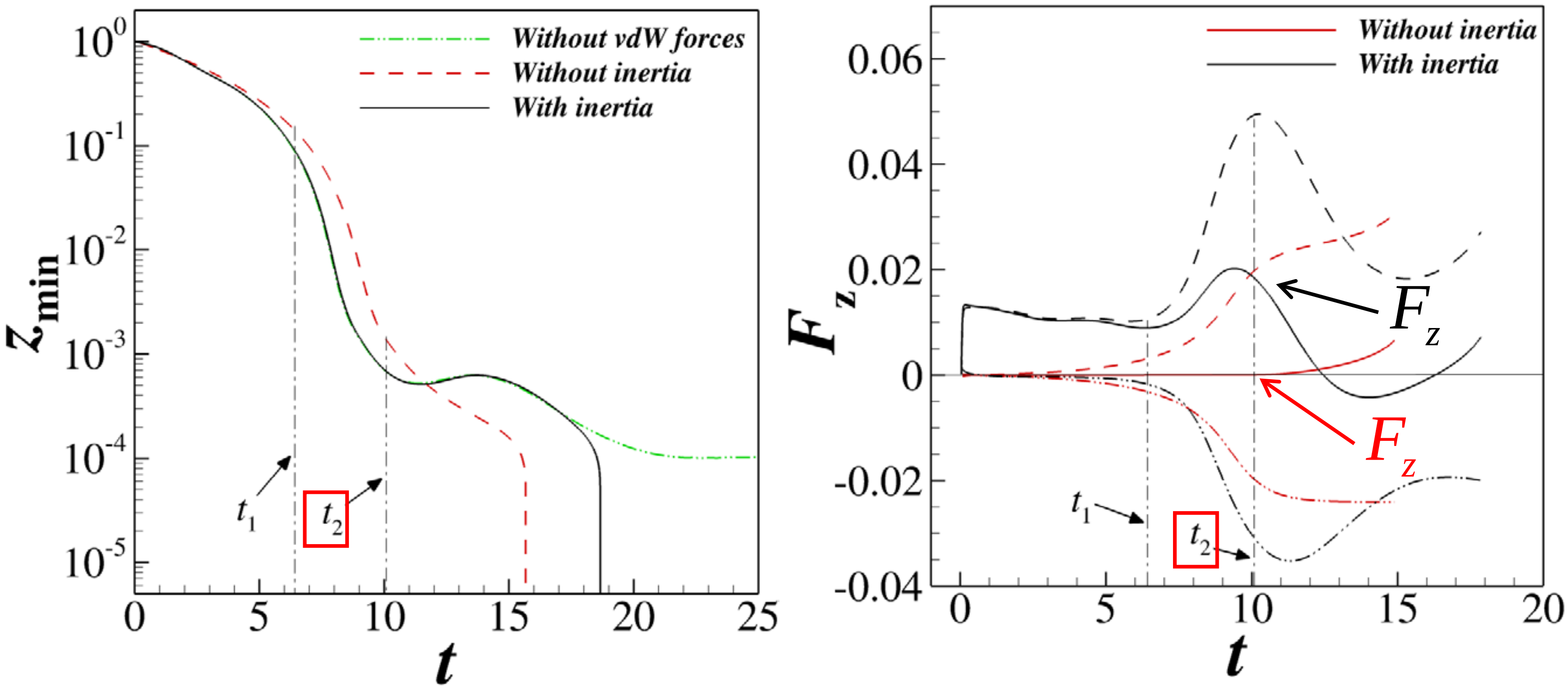



$$F_z = F_z^p + F_z^v$$

- Force due to hydrodynamic pressure in the film is always positive
- Viscous force is always negative

# Large positive $F_z$ when inertia is present

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$$

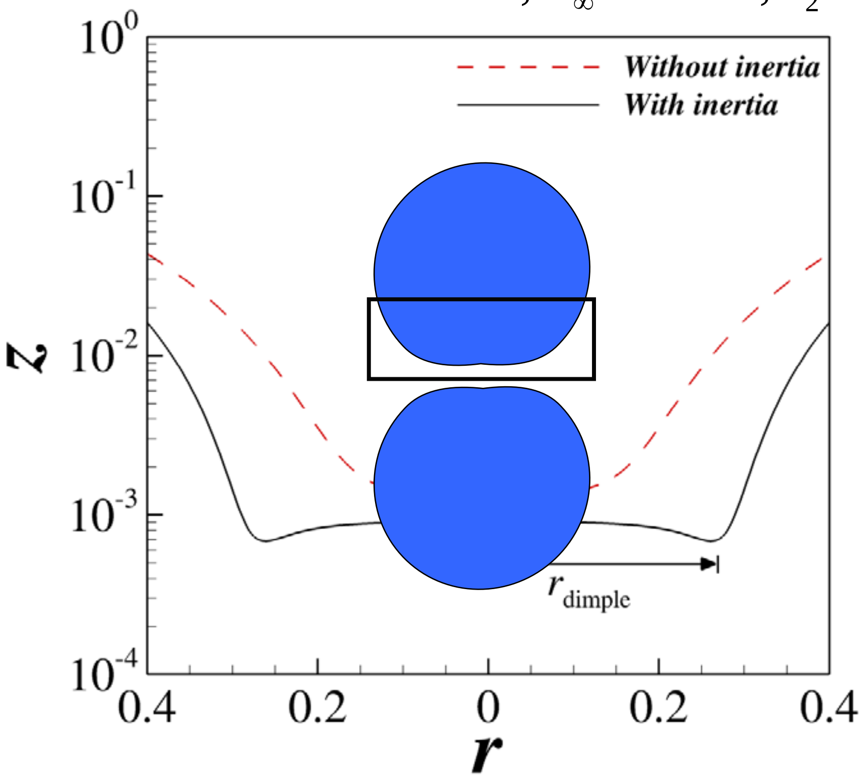


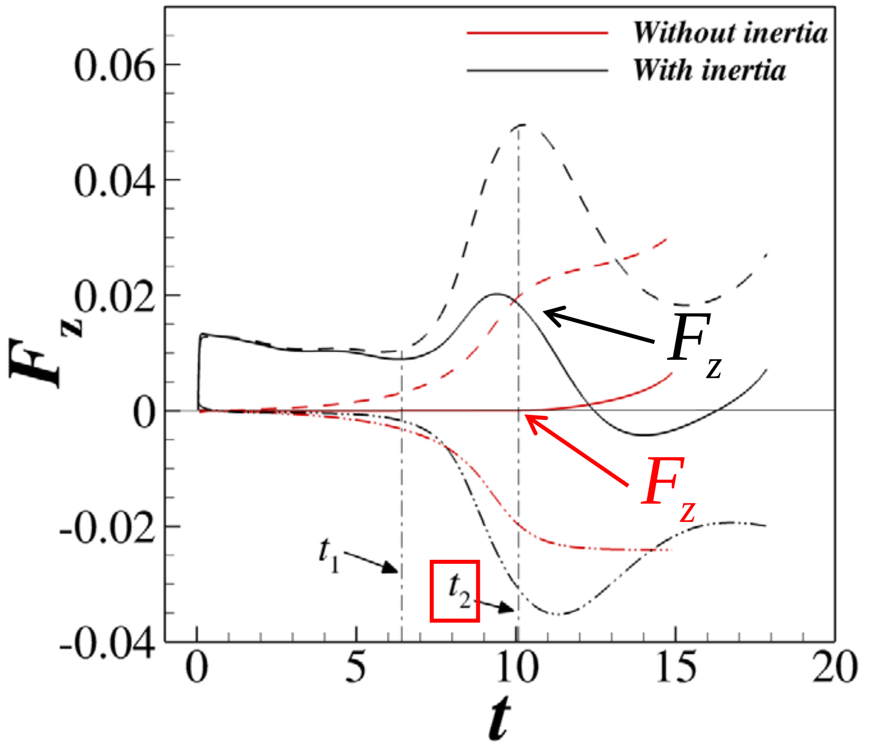

Large positive value of $F_z$ for case "**with inertia**" due to net positive difference between pressure and viscous forces pushes drops away from each other

Net zero value of $F_z$ for case "**without inertia**" as pressure and viscous forces in balance

# Large drop deformation when inertia is present

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99\times10^{-11}$$



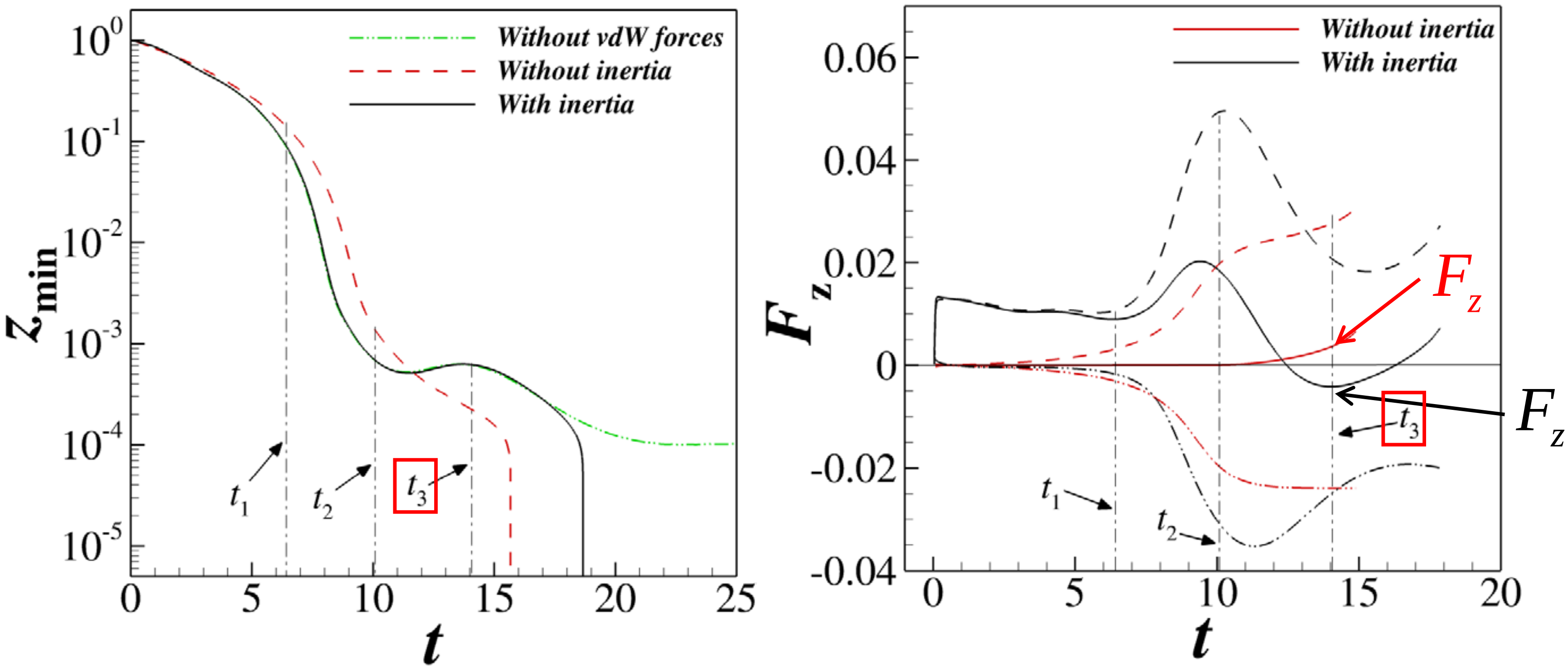




Large extent of interfacial deformation for case "**with inertia**" as compared to case "**without inertia**"

Large interfacial deformation a result of high pressure in film between the two drops

# Net negative $F_z$ after rebound forces 2nd approach

$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$$

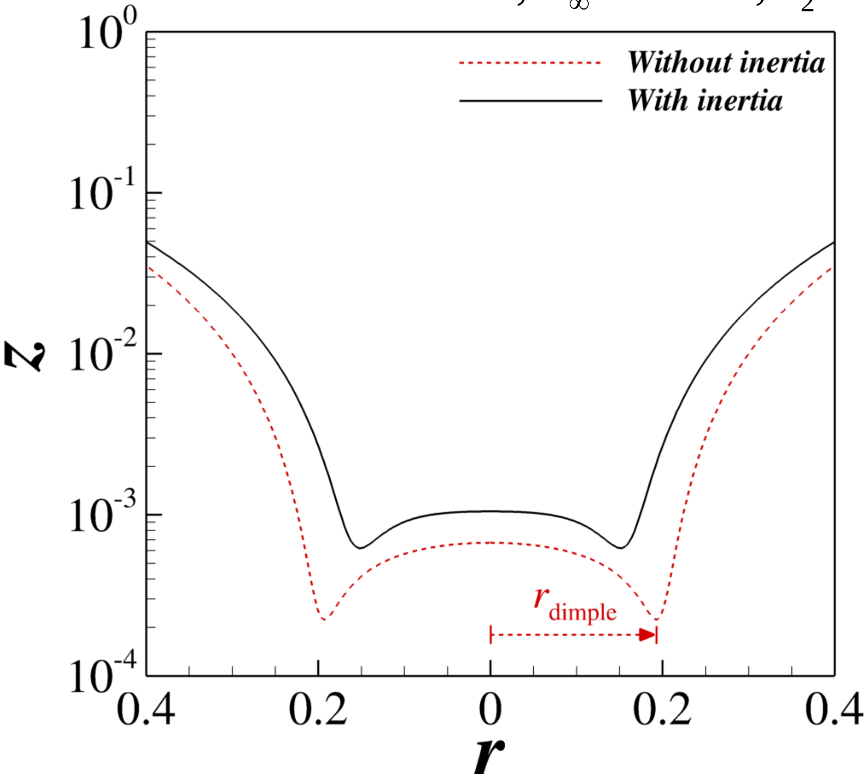


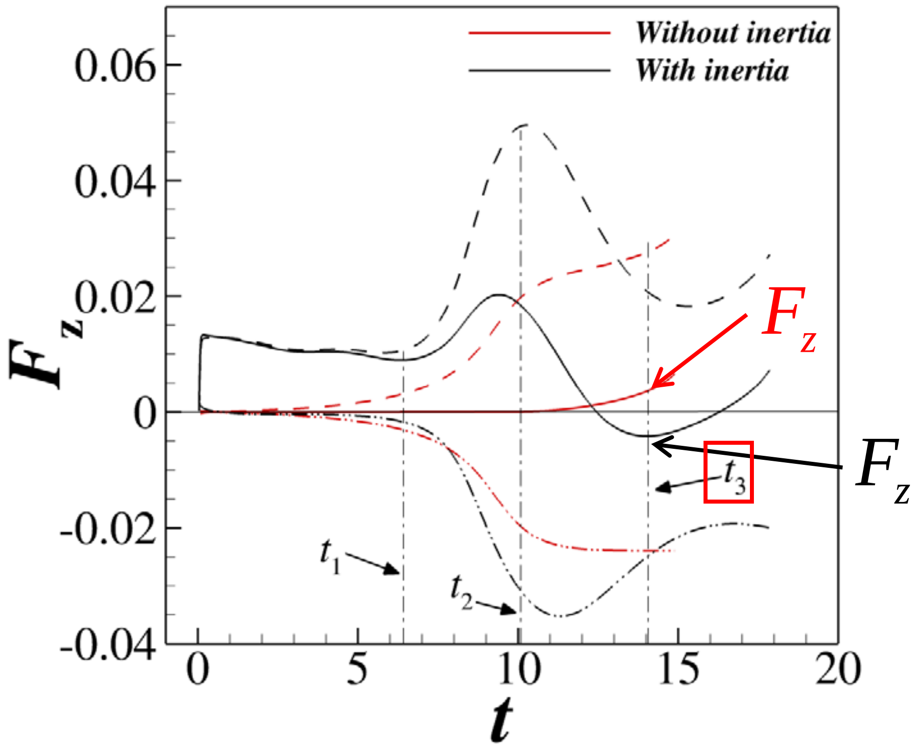

As drops move apart, viscous force dominates and results in negative value of $F_z$ for case “**with inertia**” that pushes them back together again

Small positive value of $F_z$ for case “**without inertia**” causes drops to slow down but van der Waals forces become significant at this separation and cause coalescence

# Net negative $F_z$ after rebound forces 2nd approach

$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99 \times 10^{-11}$



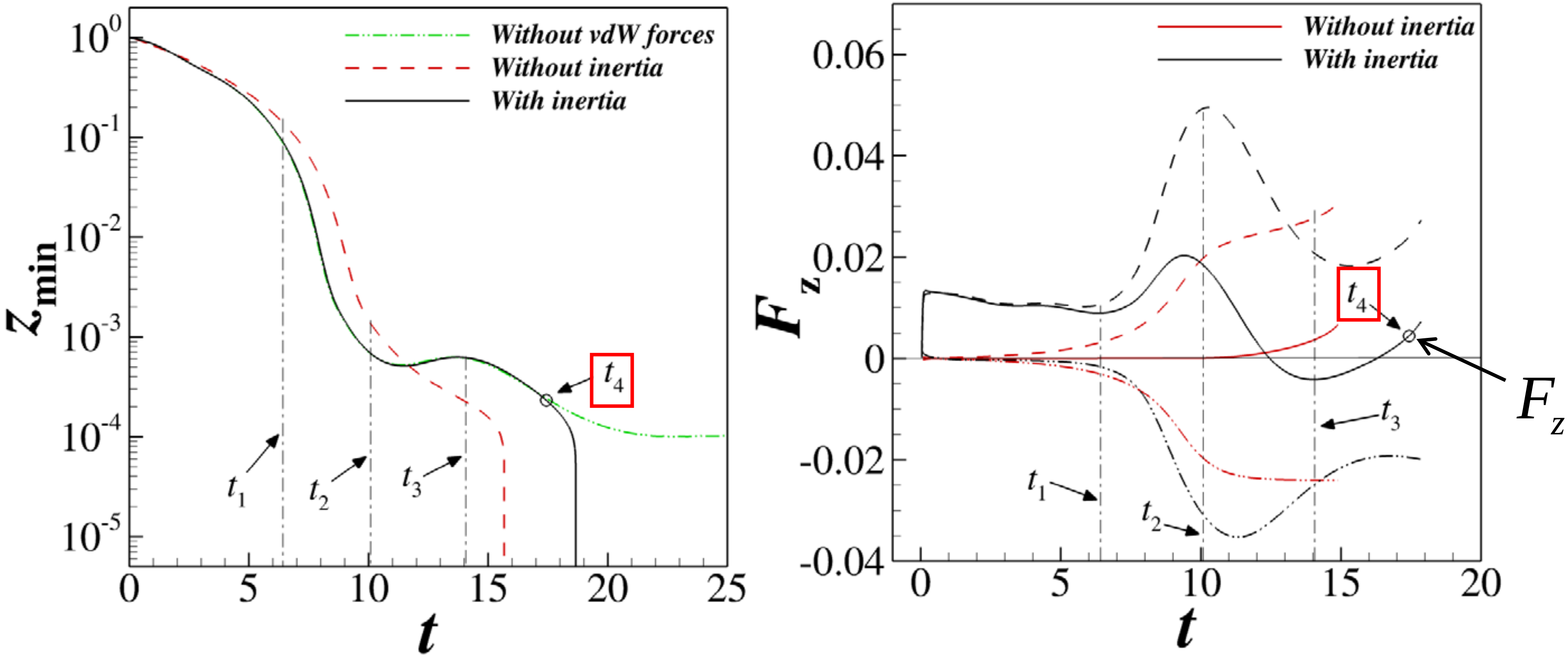




Extent of interfacial deformation $r_{dimple}$ larger now for case "**without inertia**" as compared to case "**with inertia**" as the drops have moved away from each other

Net force $F_z$ larger for case "**without inertia**"

# Coalescence on 2nd approach

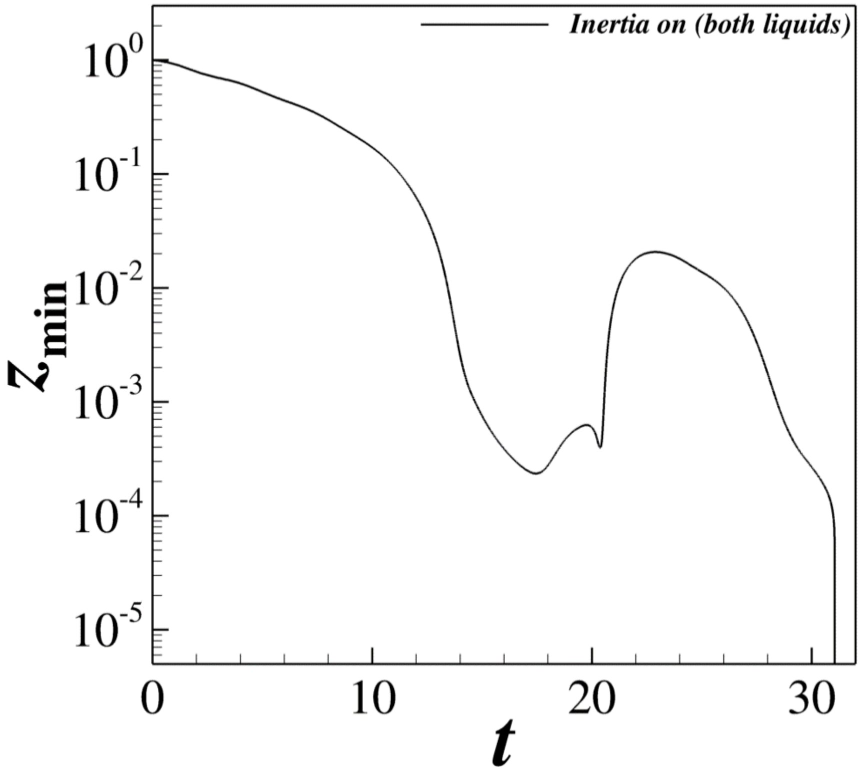
$$Oh = 0.02, U_\infty = 0.095, d_2 = 1.1, m_2 = 5.26, A = 4.99\times10^{-11}$$




As drops approach again, pressure builds up again and $F_z$ becomes positive again for case "**with inertia**" but this time van der Waals forces kick in and cause coalescence

# Which fluid's inertia is essential for rebound?

$$Oh = 0.023, U_\infty = 0.05, d_2 = 1.0, m_2 = 1.0, A = 10^{-10}$$

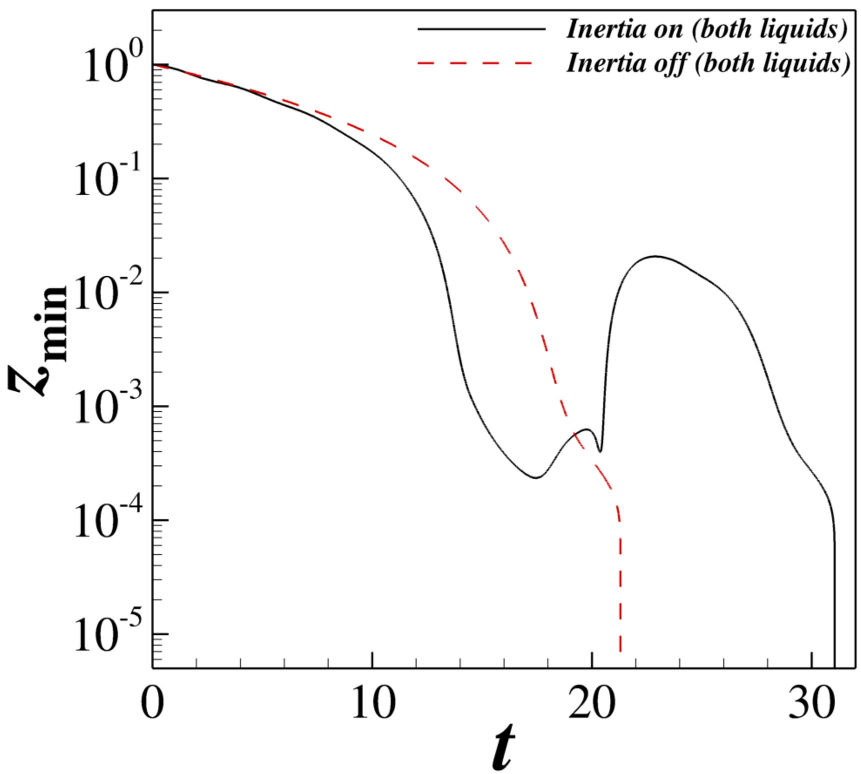



- For the physical case, the two drops rebound twice before coalescing on the third approach
- Extent of second rebound is much larger than the first

We consider a case where the two fluids are of equal density and viscosity, while the Reynolds number of the flow is $Re = 2.17$

# Which fluid's inertia is essential for rebound? ..(2)

$$Oh = 0.023, U_\infty = 0.05, d_2 = 1.0, m_2 = 1.0, A = 10^{-10}$$

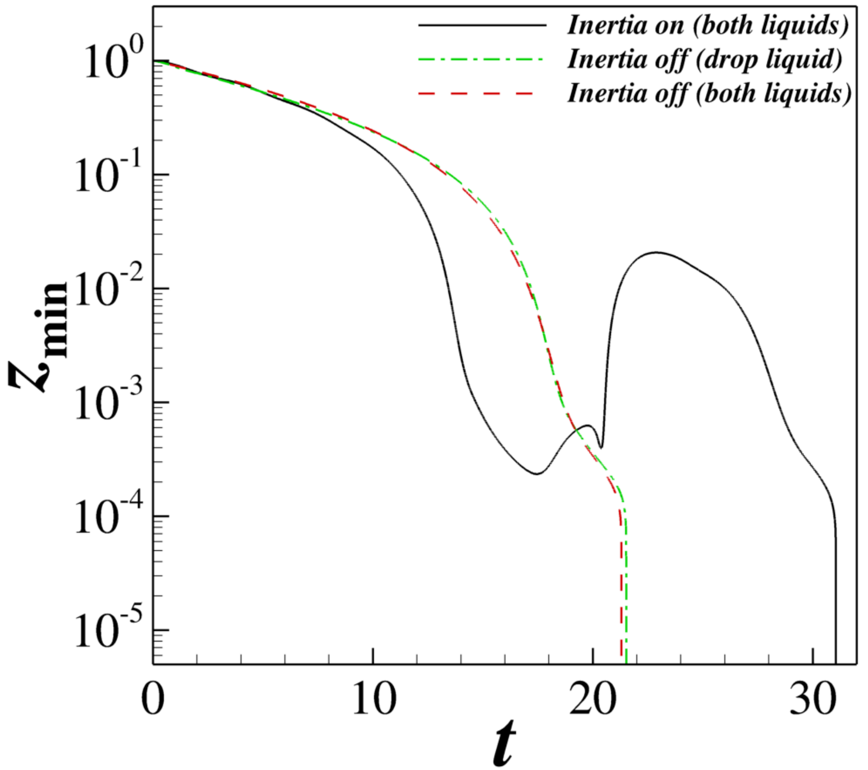



- For the physical case, the two drops rebound twice before coalescing on the third approach
- If inertia is "turned off" for both fluids, no rebound is observed

We consider a case where the two fluids are of equal density and viscosity, while the Reynolds number of the flow is $Re = 2.17$

# Which fluid's inertia is essential for rebound? ..(3)

$$Oh = 0.023, U_\infty = 0.05, d_2 = 1.0, m_2 = 1.0, A = 10^{-10}$$

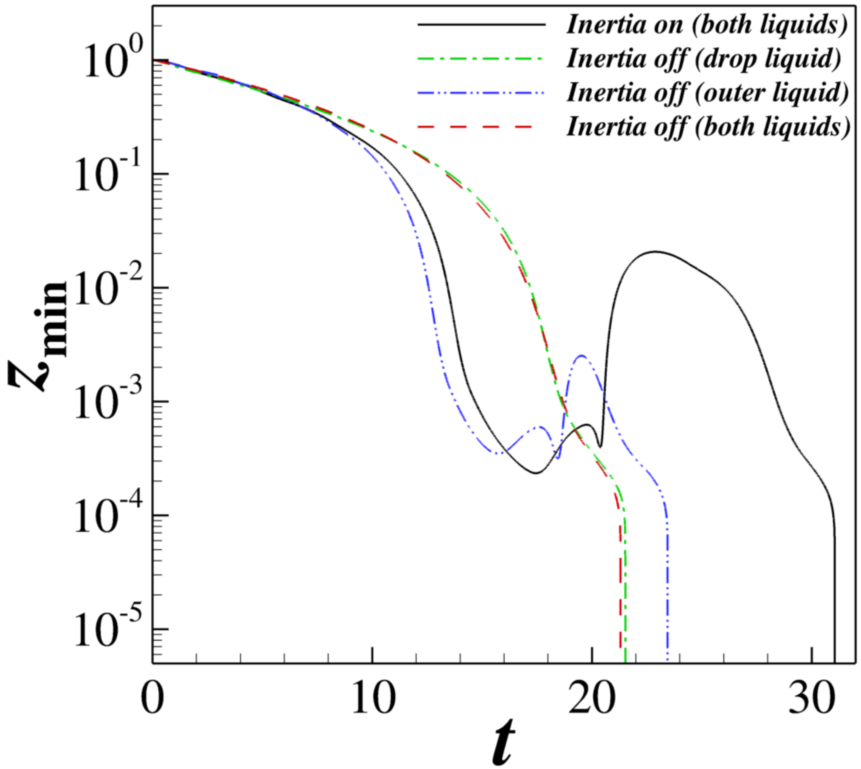



- For the physical case, the two drops rebound twice before coalescing on the third approach
- If inertia is "turned off" for both fluids, no rebound is observed
- If inertia is "turned off" for **only drop fluid,** it behaves **as if inertia is turned off for both fluids**

We consider a case where the two fluids are of equal density and viscosity, while the Reynolds number of the flow is $Re = 2.17$

# Which fluid's inertia is essential for rebound? ..(4)

$$Oh = 0.023, U_\infty = 0.05, d_2 = 1.0, m_2 = 1.0, A = 10^{-10}$$

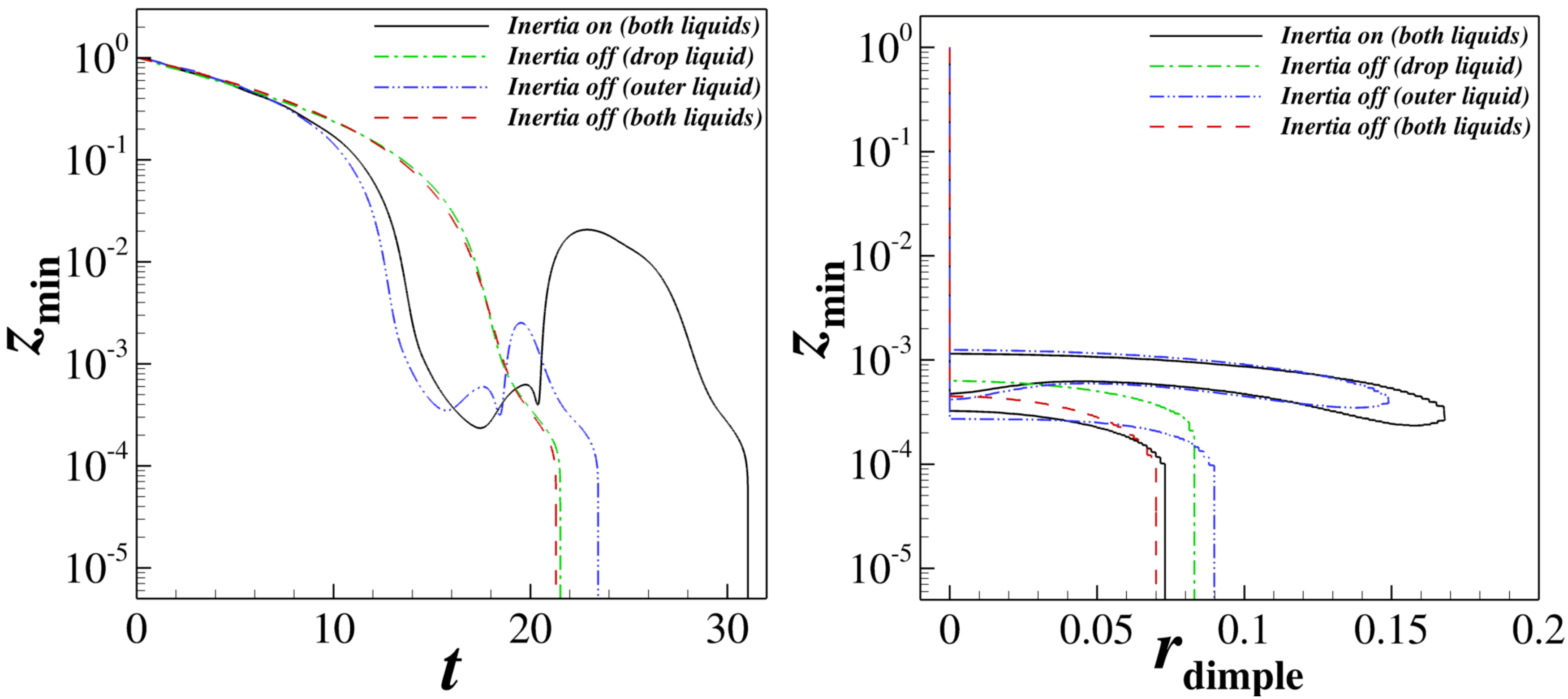



- For the physical case, the two drops rebound twice before coalescing on the third approach
- If inertia is "turned off" for both fluids, no rebound is observed
- If inertia is "turned off" for **only drop fluid,** it behaves **as if inertia is turned off for both fluids**
- If inertia is "turned off" for only outer fluid, **double rebound occurs** albeit to a modest extent

**Inertia of drop liquid** is crucial for drop rebound to occur, as its absence leads to Stokes-flow-like behavior

# Larger interfacial deformation when drop inertia present

$$Oh = 0.023, U_\infty = 0.05, d_2 = 1.0, m_2 = 1.0, A = 10^{-10}$$

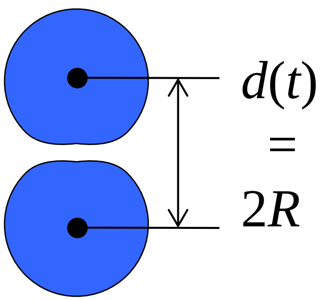



Larger extent of interfacial deformation at higher $z_{min}$ when drop fluid inertia is present leads to rebound of drops

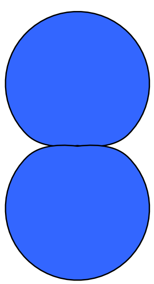
# Conclusions and impact on drainage times

Defined as:

*Beginning at*

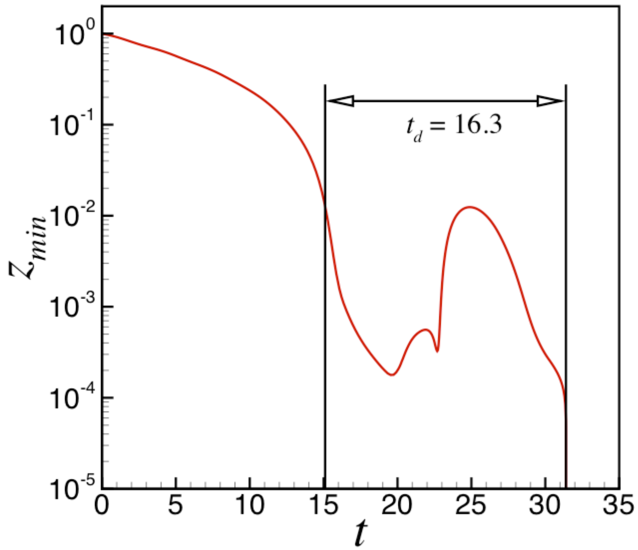


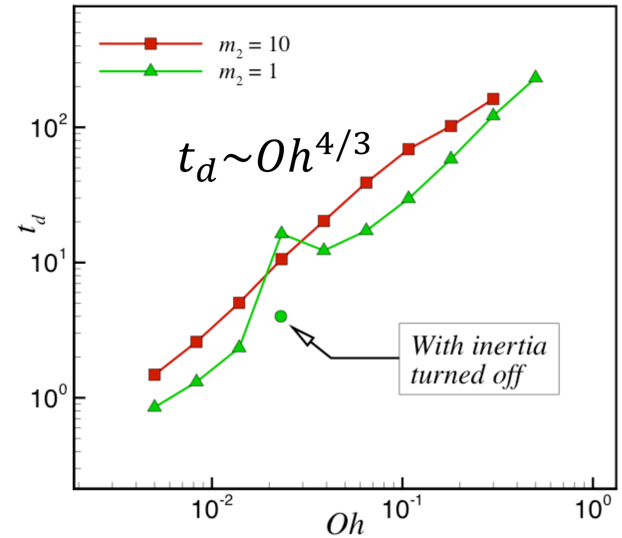

*Ending with*



*Coalescence*

$$Oh = 0.023, U_\infty = 0.05, d_2 = 1.0, m_2 = 1.0, A = 10^{-10}$$




- ***Inertia*** causes the droplets to rebound on first approach at intermediate values of *Oh* resulting in the non-monotonic variation of drainage time with *Oh*
- Accurate prediction/knowledge of drainage time is essential if the results of simulations are to be used in engineering calculations (e.g. population balances) and in engineering design

# Why the jump in $t_d$ at intermediate Oh?

- Two spheres in a nearly inviscid (i.e. very low viscosity) fluid would bounce upon colliding in the absence of van der Waals (vdW) forces
- Two spheres in a nearly inviscid fluid would stick to each other upon colliding because they can get close enough for vdW forces to become operative
- Two spheres that are driven toward each other in a very viscous fluid would slow down considerably as the pressure in the thin film separating them builds up but would ultimately coalesce without bouncing or rebounding due to vdW forces
- At intermediate Oh or viscosity, the spheres cannot get close enough on first approach for vdW forces to become large enough and therefore rebound due to the larger pressure that develops in the thin film on account of inertia