# Viscosity Measurements of Molten Salts by Rotational Viscometry

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# Provide data needed to build a FOAK MSR by 2035

Argonne is supporting Developers by:

Generating high quality molten salt property data for key properties

Standardizing measurement methods

**The MSR industry needs reliable viscosity values to model the behavior of a molten salt in a reactor over the range of normal and accident conditions.**

**The viscosity of a molten salt directly affects its thermal hydraulic behavior in a reactor, including flow and heat transport.**

**Reliable measurements are necessary to support reactor design, licensing, and operation.**

# Key Salt Properties Being Measured at Argonne

- Phase transition temperatures
- Heat Capacity
- Thermal Diffusivity and Conductivity
- *Viscosity*
- Density
- Vapor Pressure
- Mass Diffusion Coefficients
- Activity Measurements
- Compositional analyses for major and minor elements, trace contaminants including dissolved oxygen

*Rotational Viscometer Installed in a Glovebox for Measuring Molten Salt Viscosity*

# Measuring Viscosity of Molten Salts

Both absolute and relative methods can be used for measuring molten salts:

Use materials compatible with high temperature and corrosive environments

Protect or isolate electronics from elevated temperatures

Make measurements in inert atmosphere environment

## Relative Methods – use comparisons to other liquids to determine viscosity values

- Capillary methods
- Falling and rolling body methods

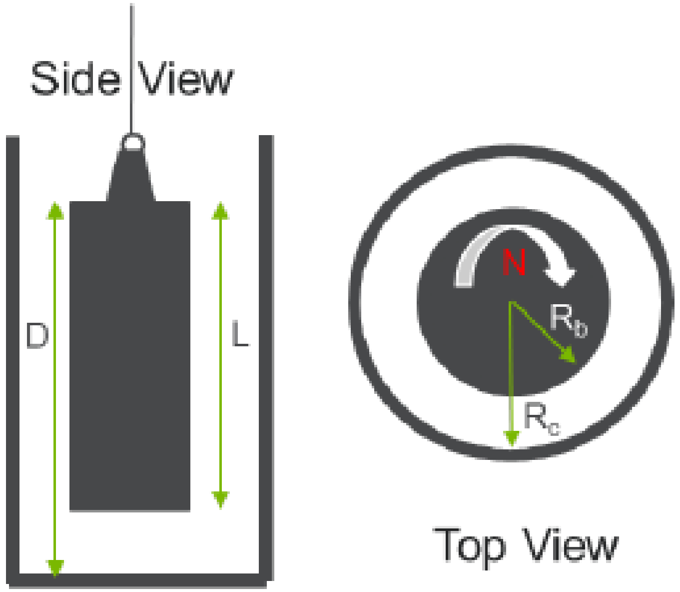
## Absolute Methods - measure dimensions and variables to determine viscosity and dependencies

- Oscillation methods
- Vibration methods
- **Rotational methods**

# Measuring Viscosity of a Molten Salt By Rotational Viscometry

Viscosity (μ) is the ratio of the shear rate ($\tau$) to the shear stress ($\gamma$) produced by an applied torque (M)

$$\mu = \frac{\tau}{\gamma} \qquad \tau = \frac{M}{2\pi R_b^2 L} \qquad \gamma = \frac{2\left(\frac{2\pi}{60}N\right)R_c^2}{\left(R_c^2 - R_b^2\right)}$$




The measured torque (**M**) required to rotate a spindle at a specific rotational velocity (**N**) is used to calculate viscosity as

$$\mu = \frac{M\left(R_c^2 - R_b^2\right)}{8\pi^2 R_c^2 R_b^2 L\left(\frac{N}{60}\right)}$$

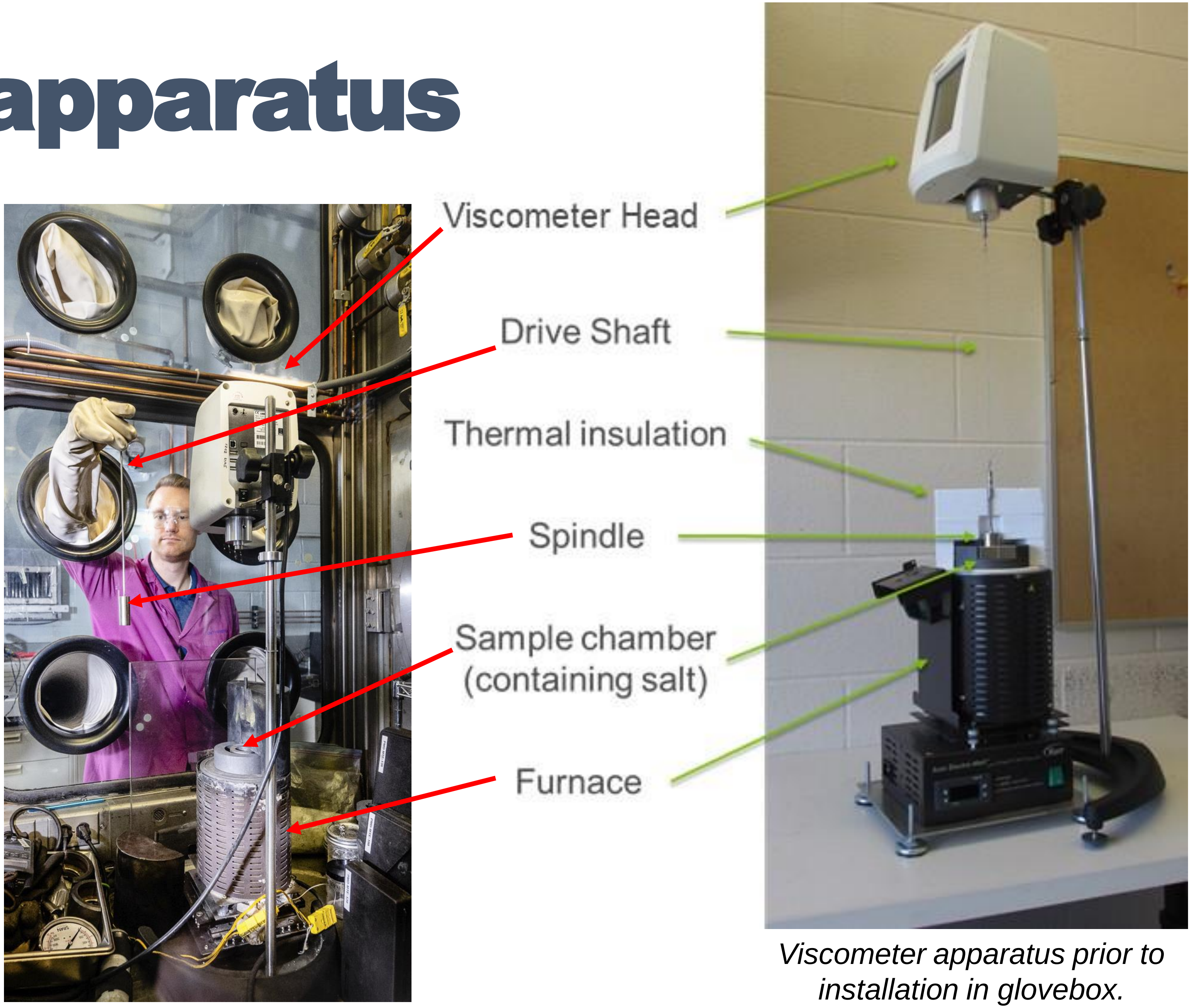
# Experimental apparatus

**Commercial viscometer head paired with custom metallic spindle and inert crucible**

**Thermal insulation to maintain uniform salt temperature**

**Furnace on x-y-z stage to facilitate alignment**

**Installed in a glovebox with argon atmosphere maintained at <1 ppm $H_2O$ and <5 ppm $O_2$**




*Viscometer apparatus installed in glovebox.*

*Viscometer apparatus prior to installation in glovebox.*

# Quality affecting aspects

System Requirements:

- Fluid must be incompressible
- Salt must be isothermal to avoid axial convective flow
- Spindle and crucible must be inert to the salt

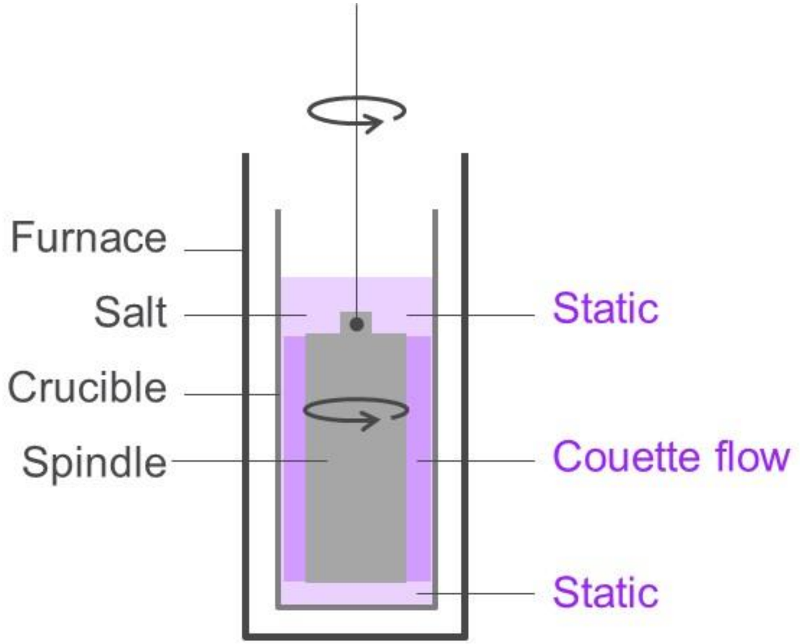
## Aspects affecting measured values and implemented appropriate controls:

### Fully-Developed Couette Flow

- Time necessary to achieve steady state laminar flow conditions
- Taylor Vortices effectively avoided by system design

### End Effects

- Depend on spindle depth in crucible and in salt




### Rotational Speed

- High speeds induce turbulence
- Responses at low speeds are affected by end effects

### Instrument wear

- Increases required torque
- Check bearing performance with reference oil regularly

# Fully Developed Couette Flow

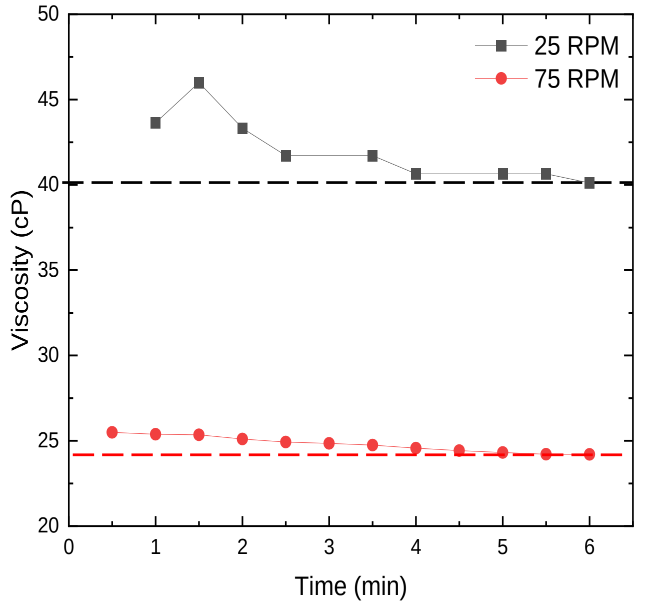
## Sufficient time should be allowed to achieve fully developed Couette flow conditions prior to measurement.

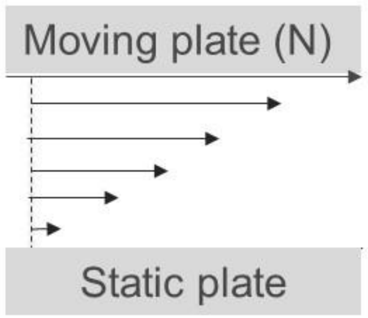
- Highly viscous salts and salts at lower temperatures require longer times to stabilize flow

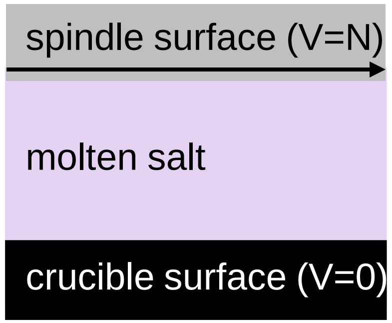
## Fully developed Couette flow is approximated by laminar flow between moving and static plates

- Re < 370 for Couette flow

*Measurements of a molten salt at various start up times, Stabilized flow values as indicated by dashed lines*




$$Re = \frac{\rho(T) N d_{spindle}(d_{crucible} - d_{spindle})}{\eta(T)}$$

$N$ is rotational velocity

## Crucible and spindle are designed to achieve laminar flow at a wide range of rotational speeds

# Avoiding Taylor Vortices

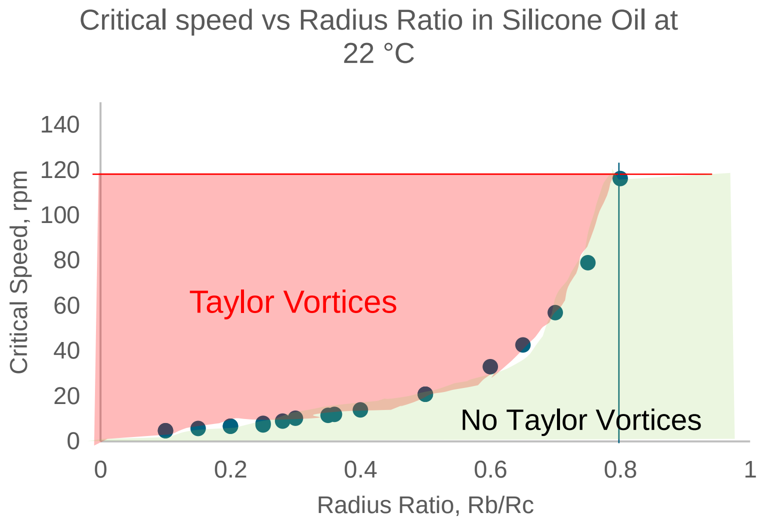
## Taylor vortices are turbulent flow in the annular region

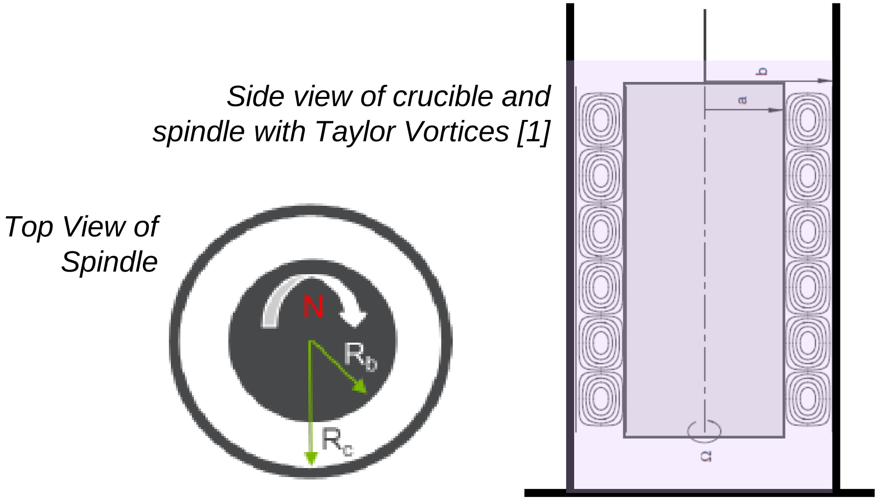
- Critical speed, above which Taylor vortices form, is a function of the ratio of the radius of the crucible and spindle ($R_b/R_c$), salt properties and temperature and should be determined for each salt and temperature

## Viscometer must be operated at rotational velocities below the critical speed to prevent turbulence. Laminar flow in the annulus is required to apply equation.

## Appropriately size spindle and crucible to avoid Taylor vortices over a wide range of rotational speeds.

- An $R_b/R_c$ of 0.8 avoids Taylor Vortices in Silicone oil at speeds up to 120 rpm.



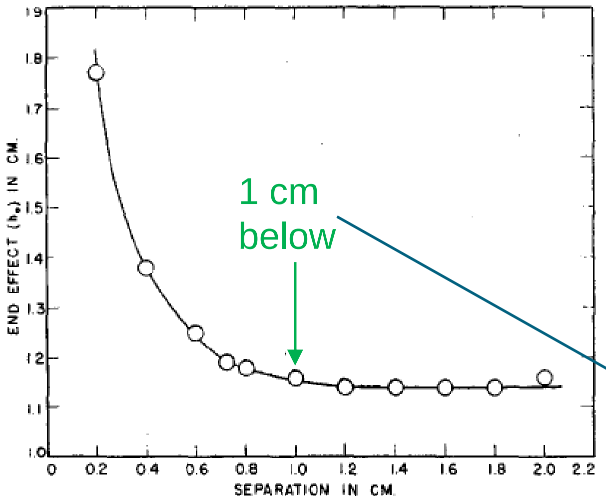




1. Childs, P.R.N. Chapter 6 - Rotating Cylinders, Annuli, and Spheres. In Rotating Flow, Oxford, UK,177–247 (2011)

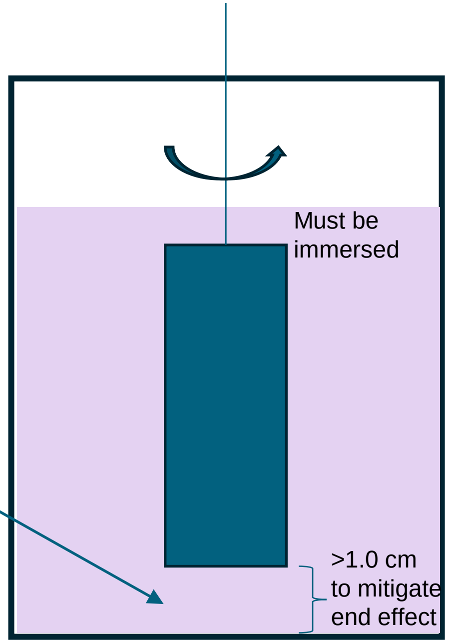
# Mitigating End Effects

## End Effect is non-laminar flow at the end of the spindle that alters flow in the annular region.

- Increasing the salt volume below the spindle mitigates the end effect.




[1] Variation in end-effect with separation between bottom of bob and cup.



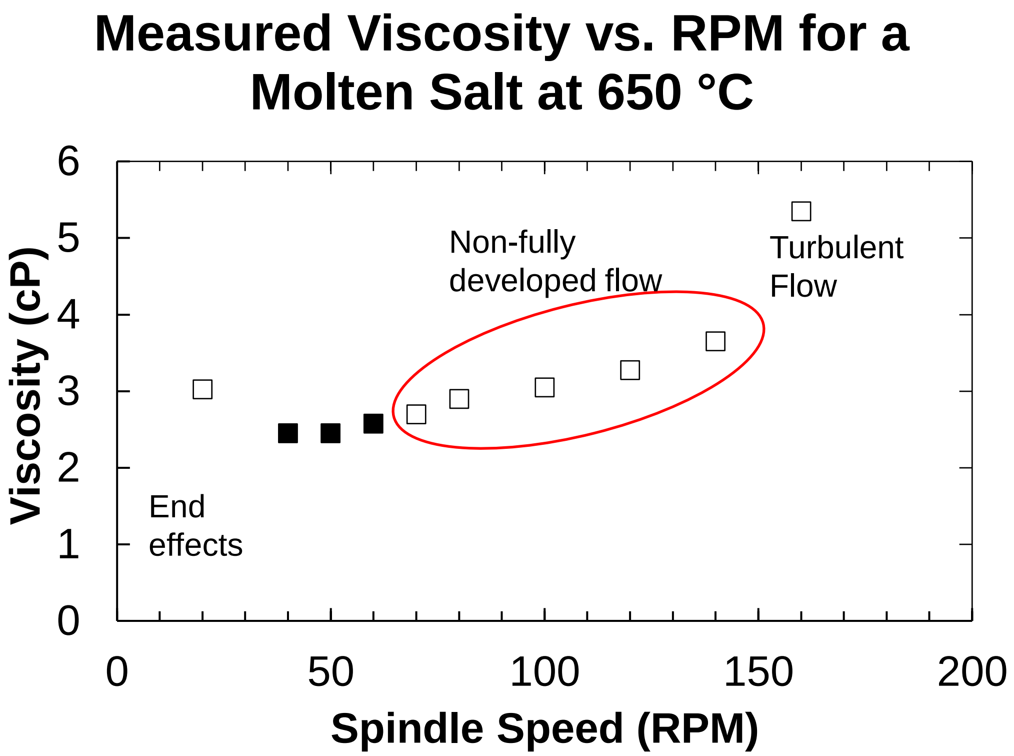

1. Lindsley and Fisher, *J. Appl. Phys.*, **18**, 11, (1947).

# Rotational Speed

## Viscosity depends on temperature and measured value is sensitive to rotational speed

- Speeds that are too low generate significant end effects
- Speeds that are too high generate turbulence
- Both end effects and turbulence increase measured torque and calculated viscosity, so minimum values are most reliable

## Take measurements at several spindle speeds to determine which values were measured under laminar flow for each salt mixture at every temperature



# Confirming Instrument Performance

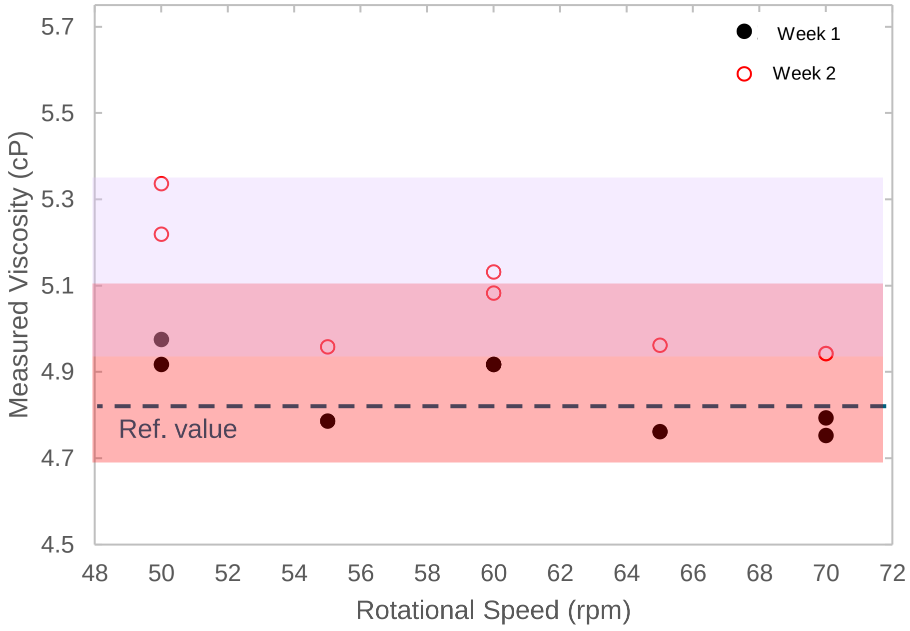
## Instrument performance must be regularly checked with a reference fluid to assess wear on the bearing.

- Spindle mass must be low enough to minimize wear on viscometer bearing but heavy enough to generate >10% maximum torque that can be measured by instrument.
- Silicone oil is used at room temperature in inert atmosphere to indicate excessive wear

## Measurements of a reference fluid are being used to assess uncertainty in measured values for salts.

- Using replicate measurements of reference fluid to determine precision of rotational viscometer system
- Differentiating between day-to-day repeatability and effects of bearing wear

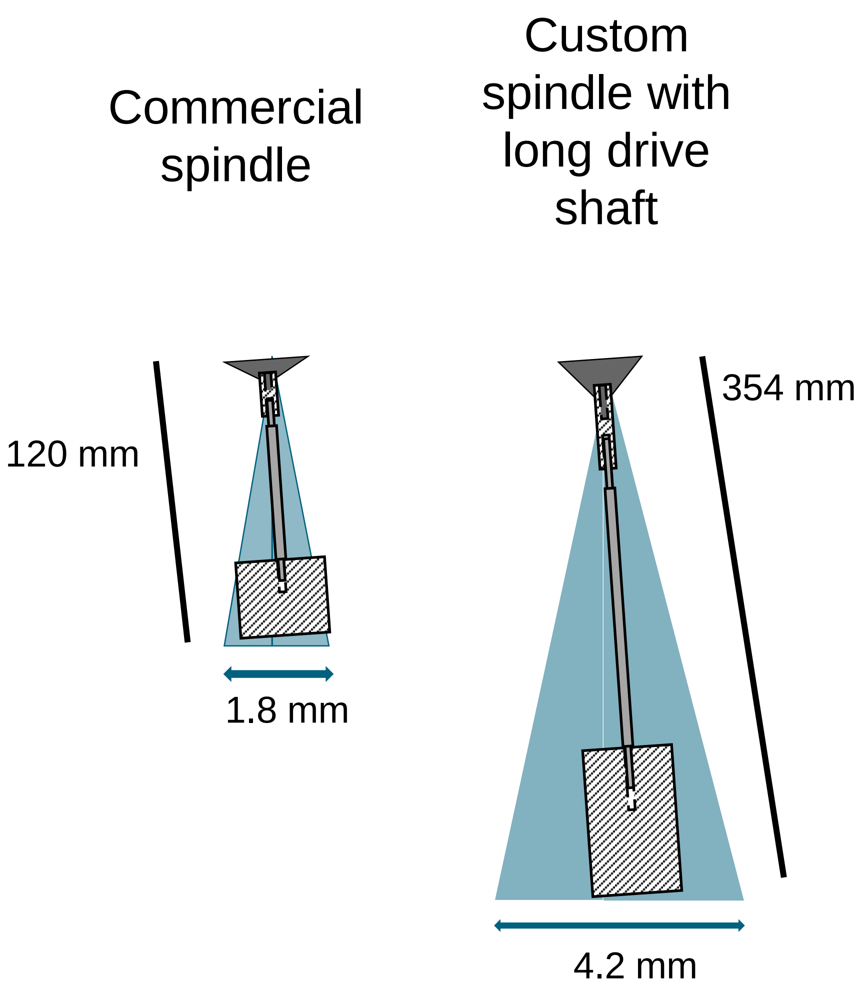
Reference Silicone Oil Measurements vs. Rotational Speed, Shaded Bands Represent One standard deviation




**Extensive use over one-week increased wear on the bearing and impacted performance.**

# Gyration

- **Long drive shaft is needed to avoid heating the viscometer head**
- **Gyration of the spindle is more significant with a  long drive shaft**
  - Decreases measurements precision
- **Precision of measurements of silicone oil with long drive shaft include effects of gyration**
  - Commercial spindle with short drive shaft is used to measure reference viscosity values for reference material (silicone oil) by a third party



# Measurement Controls and Calibrations

## Use calibrated measurement devices with known accuracy:

- **Measurements of spindle and crucible size**
  - Use calipers to measure dimensions
    - Check accuracy of calipers with certified gauge blocks
- **Depth Measurement**
  - Calculate immersion depth from salt mass, known density of the salt at measurement temperature and measured dimensions of crucible and spindle
  - Use calipers to measure the distance the viscometer head is lowered to achieve immersion depth

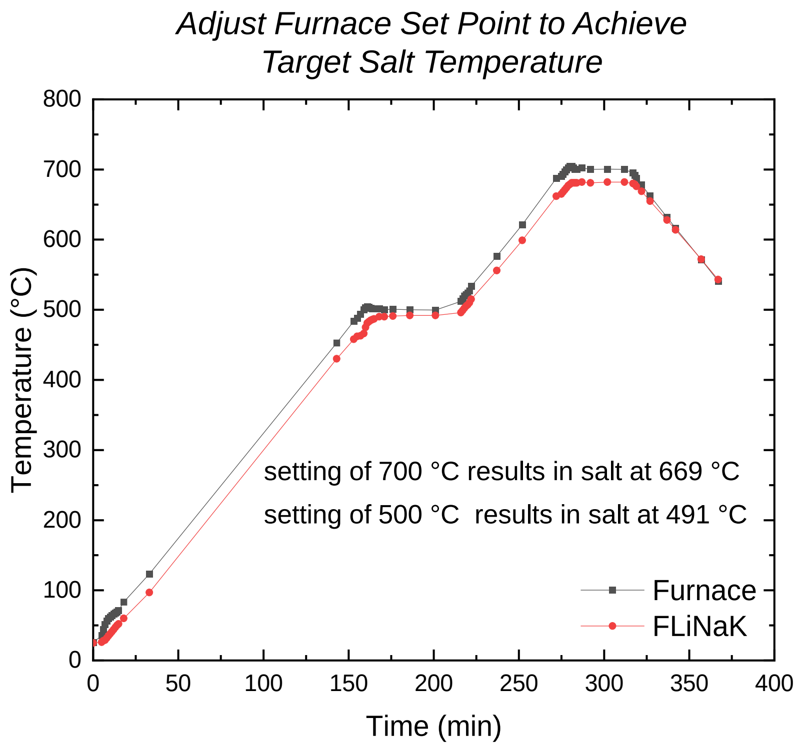
*Using calipers to measure a spindle*

# Temperature Measurement

## Determine furnace set point to achieve desired salt temperature

- Use calibrated thermocouples immersed in a test salt to determine furnace set point corresponding to the target salt temperature
- Confirm calibration of thermocouples with a dry block calibrator

## Using a thermocouple during measurement is not recommended as it will impact flow



# Uncertainty Quantification

Uncertainty in dimensions (Rc, Rb, and L), and measured torque and rotational speed (M and N) can be propagated to determine uncertainty in the measured value:

$$\mu = \frac{M\left(R_c^2 - R_b^2\right)}{8\pi^2 R_c^2 R_b^2 L\left(\frac{N}{60}\right)}$$

$$s_f = \sqrt{\sum \left(\frac{\partial f}{\partial x}\right)^2 s_x^2}$$

The combined uncertainty in measured torque and rotational velocity are being determined by measuring the viscosity of a reference fluid at room temperature.

Precision of measured viscosity is based on replicate measurements

# Measuring the Effect of Fission Products on Viscosity of Molten Salt

**Measuring viscosity of salt doped with fission products for comparison with measurements of the same salt without dopants.**

- **LiF-NaF-KF salt was doped with fission products at compositions representing low burn up**
  - **Inspired by depletion calculation results for MSRE**


***Doped FLiNaK Composition (mol %)***

| Component | Composition (mol %) |
|---|---|
| FLiNaK | 99.65 |
| $ZrF_4$ | 0.05 |
| Mo | 0.05 |
| $NdF_3$ | 0.05 |
| $CeF_3$ | 0.05 |
| CsF | 0.05 |
| CsI | 0.005 |
| $SrF_2$ | 0.05 |
| Ru | 0.05 |
| Te[a] | 0.005 |

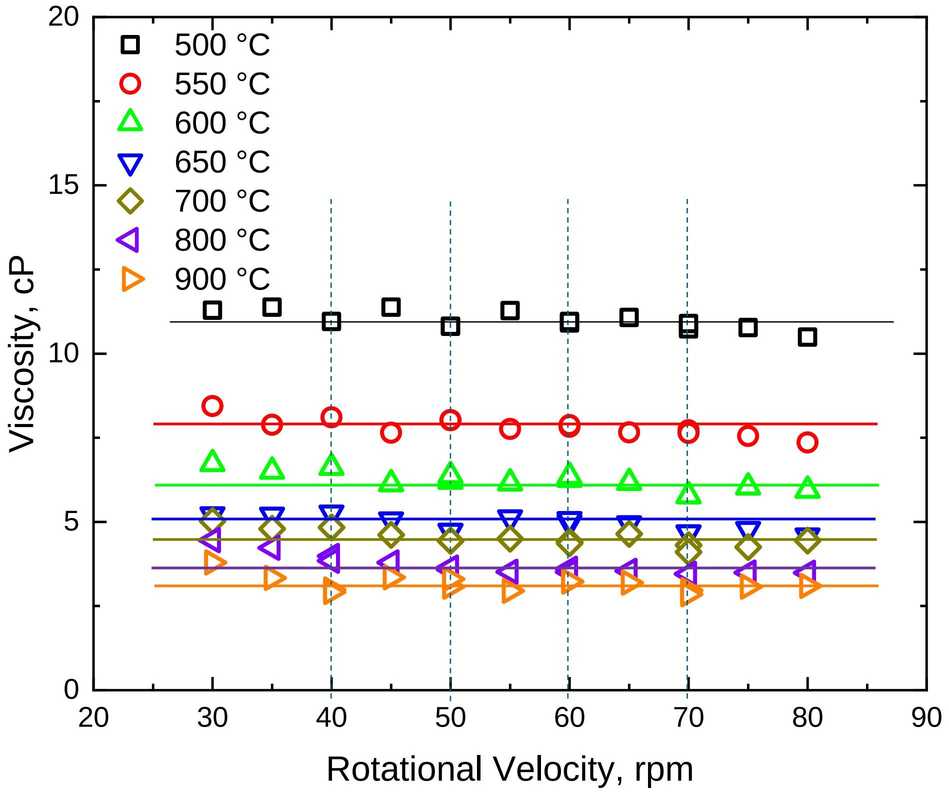
[a] Added as $Na_2Te$

# FY24 Viscosity Measurements by Rotational Method

## The viscosities of low-burnup FLiNaK salt were measured between 500 and 900 °C.

- Duplicate measurements made at 40, 50, 60, and 70 rpm indicate measurements are repeatable.
- Measured viscosities are independent of rotational velocity between 40 and 70 rpm, which is expected for Newtonian fluids.

*Low-burnup FLiNaK viscosity measured at different rotational velocities*



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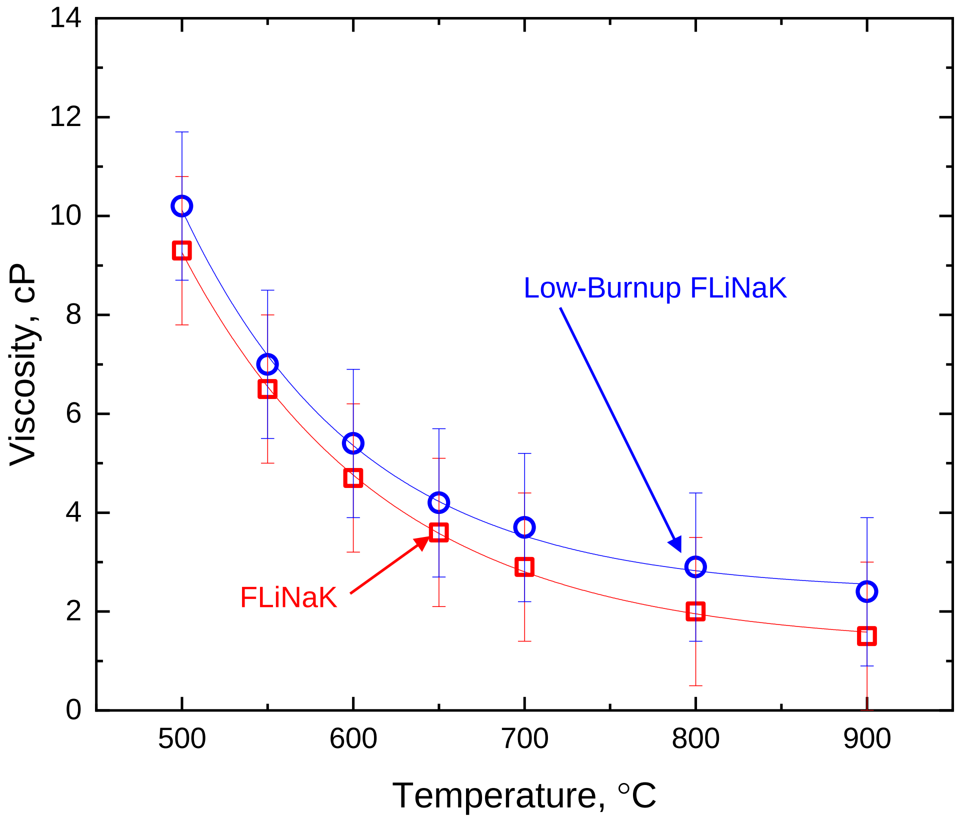
# FY24 Viscosity Measurements by Rotational Method

## The effects of fission products on viscosity

- Viscosities of low-burnup FLiNaK are higher than those of FLiNaK at each temperature, but differences are within the precision of the measurements.
- The temperature dependence of doped salt viscosity values is consistent with trends measured for FLiNaK and other salts.
- Error bars represent ranges of replicate measurements



# Developing Standard Test Methods

Standard test methods support:

Uniformity in calculating property values from measured values and in determining uncertainty.

Uniform application of the method across institutions will allow direct comparison of measured values.

Round-robin studies to validate property values for key salts.

**Argonne is leading an ASTM task group to develop standardized method for viscosity measurements of molten salts using a rotational viscometer (WK91221)**

**"A *set of standard methods* for measuring the thermal properties of molten salts will enable the generation of high-quality property data needed to qualify molten salt fuel and to design, license, and safely operate MSRs."**

**- from the MS properties uncertainty workshop report**

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M.A. Rose. *Molten Salt Thermal Property Uncertainty Workshop Report*. ANL/CFCT-23/32. September 2023.



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# WK91221 : Standard Test Method for Measurement of Viscosity of a Molten Salt using a Rotational Viscometer

1. Scope

   1.1 This method can be used to determine the viscosity of a molten salt (Pa·s), including salt mixtures, contaminated salts, and radioactive salts, at high temperatures (e.g., 900 °C) in a controlled atmosphere by using a rotational viscometer. The torque generated by the resistance of a spindle rotated at a constant rotational speed due to the resistance of the salt is measured.

   Controls necessary for accurate measurements are identified. These address the corrosive nature of molten salts, measurements of radioactive salts, elevated temperatures, and salt volatilization.

4. Summary of Method

   4.1 A cylindrical spindle is immersed in a molten salt contained in a cylindrical container at a known temperature to a known depth and rotated at a known rotational speed. The viscous drag on the immersed spindle is measured as torque at several rotational speeds. Torque values measured at different speeds are used to determine the range of speeds resulting in non-turbulent flow. The viscosity is determined using torques measured under stable Couette flow conditions and the shear rate is determined based on the dimensions of the spindle and crucible.

# Summary

**Quality-affecting aspects of measuring viscosity of a molten salt by using rotational viscometry have been identified and appropriate controls have been developed.**

**A standard test method for molten salt rotational viscometry is being developed through ASTM – Argonne is leading this effort.**

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- Government License Notice -the manuscript has been created by UChicagoArgonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357.

# Thank you

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