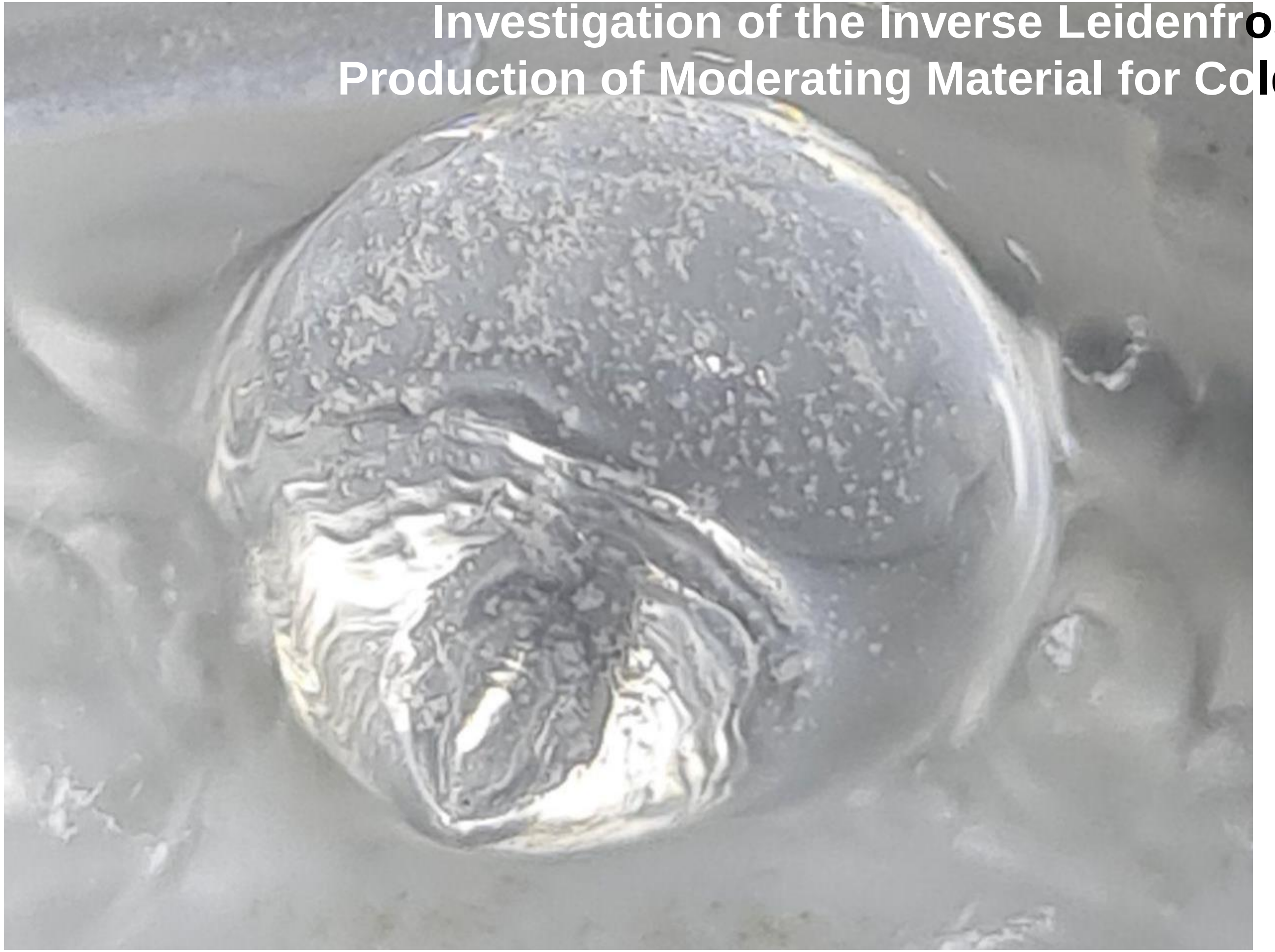
Investigation of the Inverse Leidenfrost Effect in the Production of Moderating Material for Cold Neutron Sources



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Current Challenges

Required		Current Capabilities	Issue
Production Needs	Baseline: 6 L in 4 weeks	6.7 L in 4 weeks	Current device meets only baseline needs; emergency production impossible
	Optimal: 10 L in 4 weeks		
	Future: 10 L/day for a source with 10x neutron flux		
Safety for Employees and Productivity		Routine work. Contact with liquid nitrogen and mesitylene mixture	Increased costs and risks with higher productivity
Scalability		Requires additional device	
Ease of Use		1 employee can manage 4 devices	
Defect and Size Control		Post-batch (330 ml beads)	One day lost if issues arise
Dry Storage		Not possible	Delay required for helium purge before loading

Relevance

1) **Solution to the problems described**
2) **Brings you closer to use Methane for Solid Frozen Beads**
   3–4x Increase in Cold Neutron Intensity
3) **Medicine and Biology**
   Freezing and transporting biological materials without contamination risks [1].
4) **Materials Science**
   Studying Rupert’s drops from various materials (sizes from mm to cm) [2].
5) **History and Archaeology**
   Exploring processes similar to lava interacting with cold surfaces during eruptions [3].
6) **Molecular Cuisine**
   Creating edible beads from liquids (e.g., compote).

[1] Inverse Leidenfrost Effect: Levitating Drops on Liquid Nitrogen / M. Adda-Bedia, S. Kumar, F. Lechenault, S. Moulinet, M. Schillaci, D. Vella // Langmuir 2016, 32, 17, 4179–4188
[2] Rupert's glass drops: Residual-stress measurements and calculations and hypotheses for explaining disintegrating fracture / W. Johnson, S. Chandrasekar // Journal of Materials Processing Technology, 31 (1992) 413-440
[3] Prince Rupert’s Drops: An analysis of fragmentation by thermal stresses and quench granulation of glass and bubbly glass / K. V. Cashman etc. // PNAS, 2022 y. Vol. 119 № 31

Objectives and Tasks

Objectives:

Develop a high-performance automated device using knowledge of physical processes involved in bead production

Tasks:

1) Investigate the physical processes occurring during beads production.
2) Conduct laboratory experiments to validate research findings.
3) Design a automated device to produce mesitylene beads with a target output of 1 liters in 1 hours, based on the obtained results.

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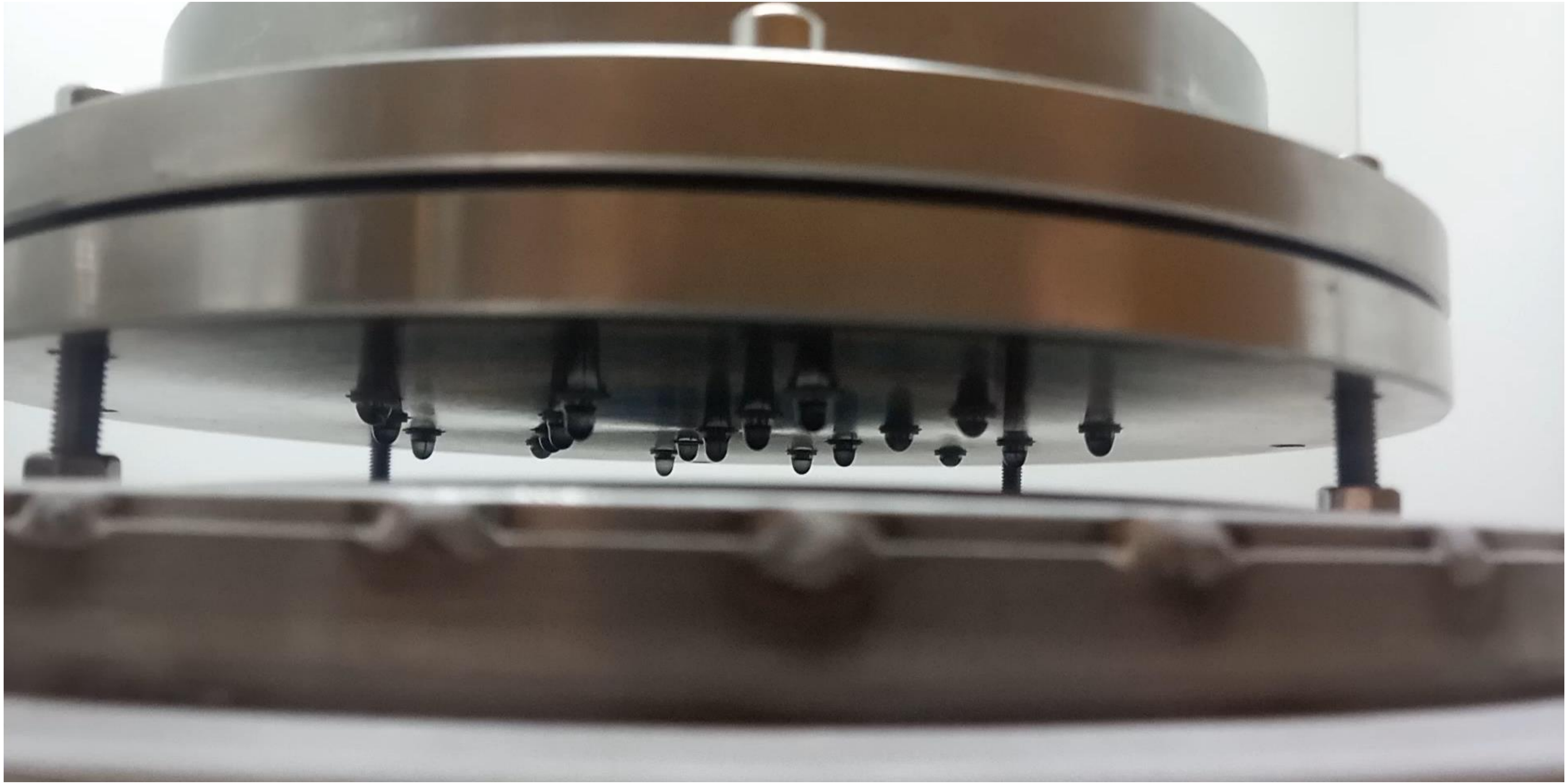
Technology description



Investigate the physical processes occurring during beads production

1) Formation of a drop
2) Heat exchange of liquid nitrogen with the surrounding space
3) Heat exchange of a drop with liquid nitrogen
4) Cooling and crystallization of a drop
5) Adhesive properties of balls

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Formation of a drop

1) Force balance:

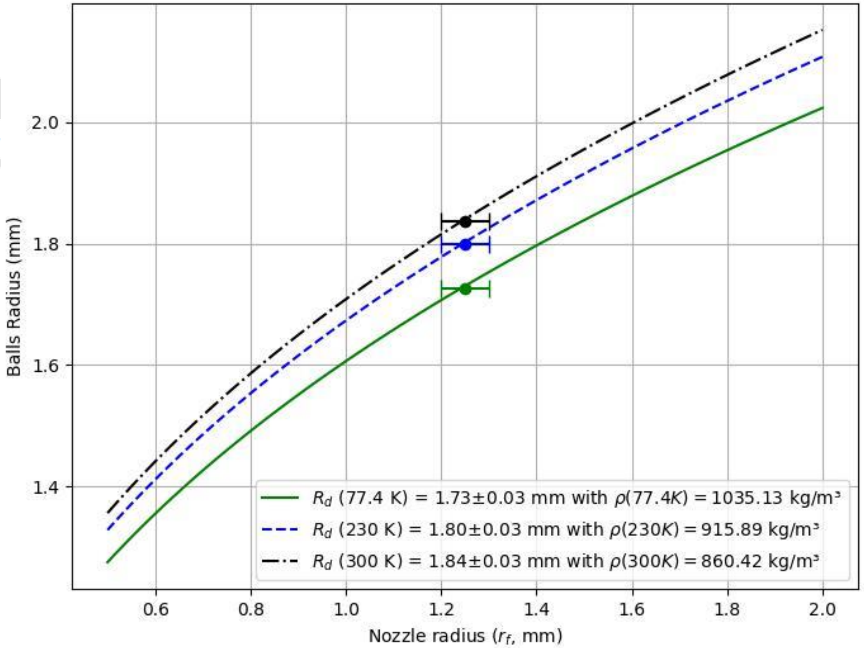
$$\sigma_d \int_{l_d} dl = g\rho_d \int_{V_d} dV \Rightarrow 2\pi\sigma_d r_f = g\rho_d V_d \Rightarrow V_d = \frac{2\pi\sigma_d r_f}{g\rho_d}$$

2) Droplet detachment radius:

$$R_d = \sqrt[3]{\frac{3\sigma_d r_f}{2g\rho_d}},$$

3) Correction to the bead radius based on density change:

$$m = \rho_d V_d = \rho_{df} V_{df} \Rightarrow V_{df} = \frac{\rho_d V_d}{\rho_{df}} \Rightarrow R_{df} = R_d \sqrt[3]{\frac{\rho_d}{\rho_{df}}},$$



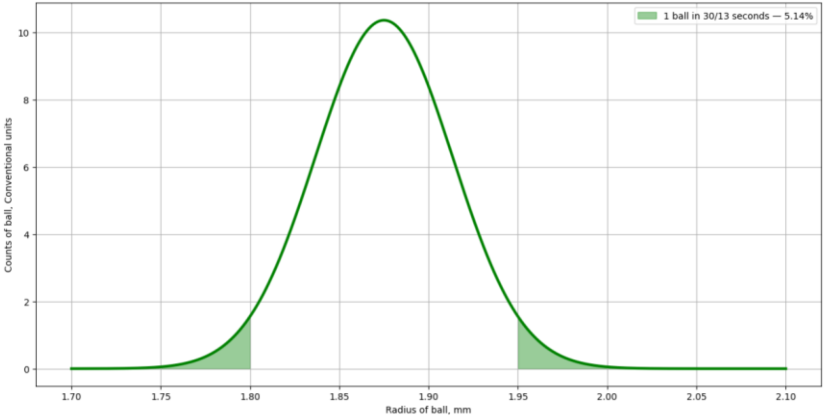
Formation of a drop

Nozzle Radius (mm)	Detachment Mechanism
$r_f < 2.04$	**Rayleigh-Plateau Instability**
$2.04 < r_f < 4.37$	Gravity > Surface Tension
$r_f > 4.37$	Taylor Instability

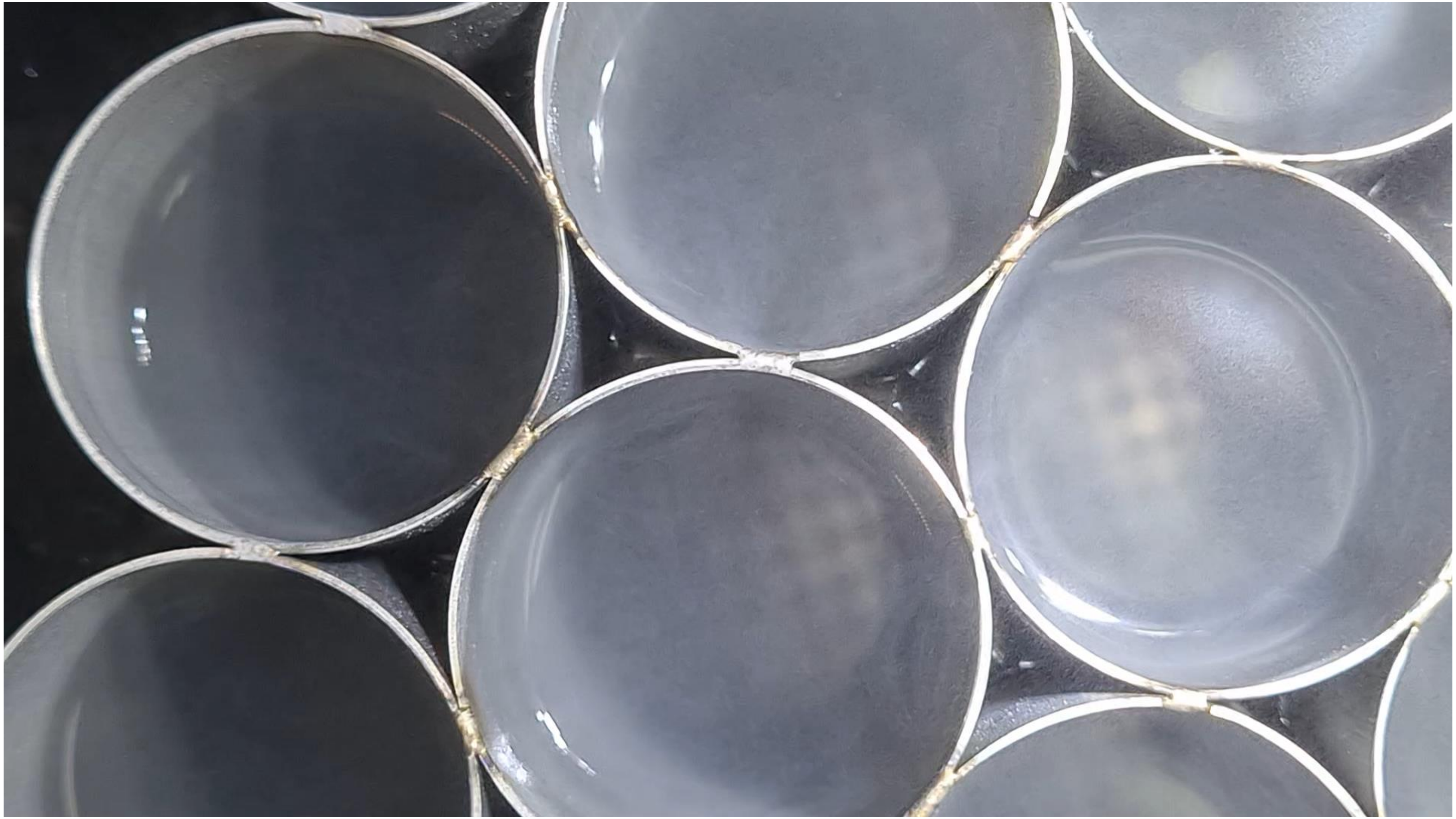
Dependence of droplet detachment mechanisms on nozzle radius for a mesitylene and meta-xylene mixture [1]

[1] A. I. Grigoriev, A. A. Zemskov, Detachment of a drop from a capillary under the action of gravity. Nauchnoe Priborostroenie 1, 50–58 (1991).

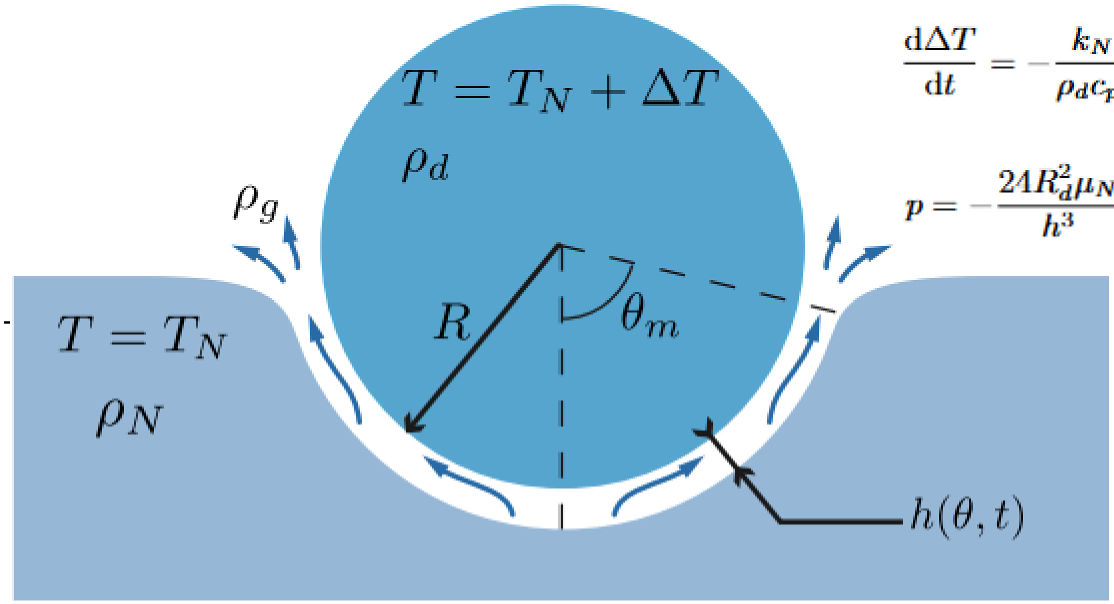
Therefore, the size distribution of balls can be described using a normal distribution with a mean ball radius of 1.875 mm and a standard deviation of 0.0385 mm



Heat exchange of liquid nitrogen with the surrounding space



Heat exchange of a drop with liquid nitrogen(Inverse leidenfrost effect)



$$\frac{\mathrm{d}\Delta T}{\mathrm{d}t} = -\frac{k_N A_d}{\rho_d c_{pd} V_d}\frac{\Delta T}{h},$$

Heat exchange of a steam pillow

Lubrication theory:

$$p = -\frac{24 R_d^2 \mu_N}{h^3}\left(\frac{\mathrm{d}h}{\mathrm{d}t} - \frac{k_N \Delta T}{\rho_g L_{vN} h} - \frac{\sigma \Delta {T_0}^4 \pi R_d^2}{\rho_g L_{vN} A_d}\right)\ln\left(\frac{\cos\left(\frac{\theta}{2}\right)}{\cos\left(\frac{\theta_m}{2}\right)}\right.$$

$\Delta T = 0$ and quasi-static limit:

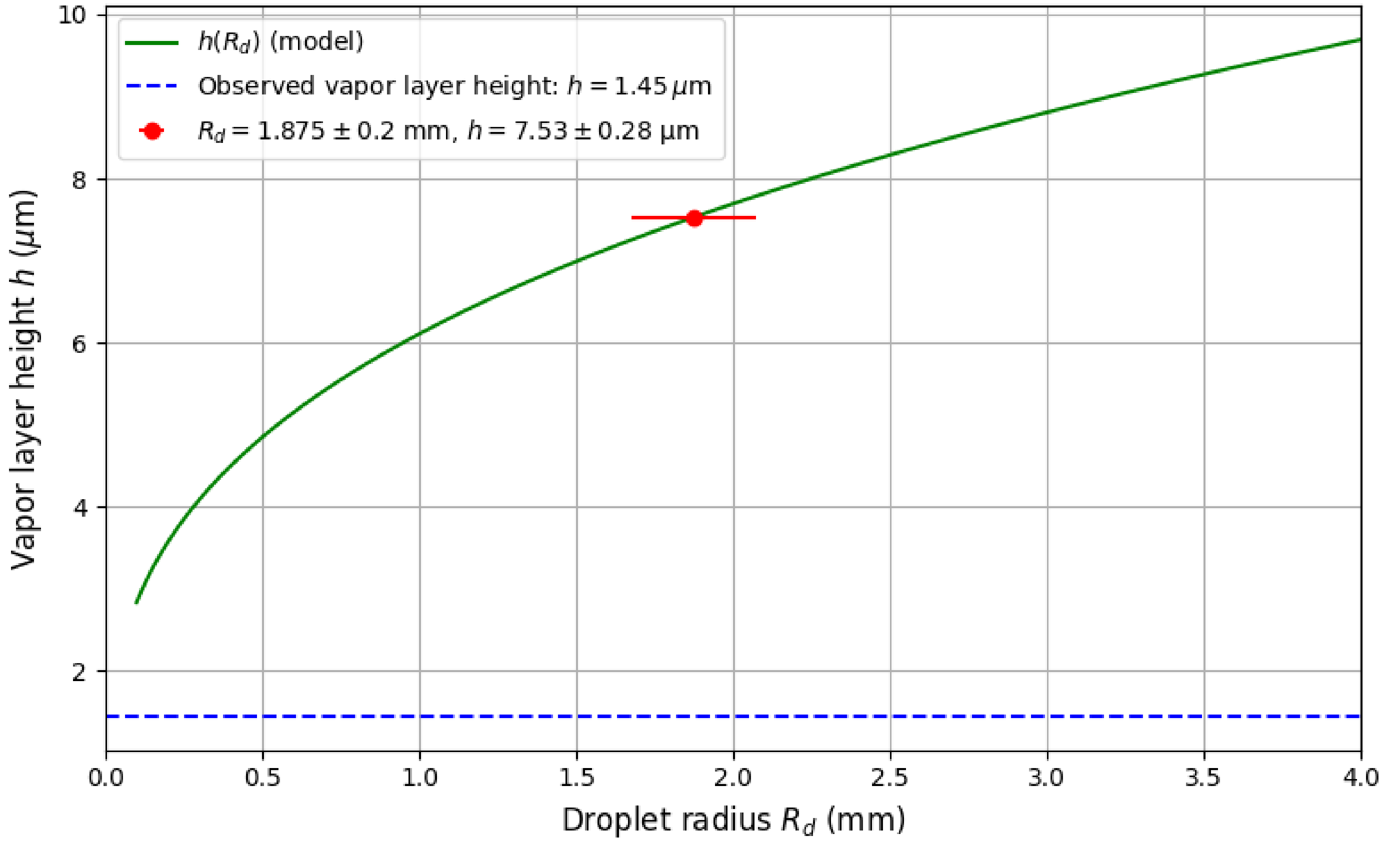
$$h = \sqrt[3]{\frac{3\pi^2 \mu_N \sigma \Delta T_0^4 R_d^6}{\rho_d g V_d \rho_g L_{vN} A_d}(3 + \cos 2\theta_m - 4\cos\theta_m)},$$

A Gauthier, CD R ´emi, D Lohse, D van der Meer, Self-propulsion of inverse leidenfrost drops on a cryogenic bath. Proc. Natl. Acad. Sci. 116, 1174–1179 (2019).

M Adda-Bedia, et al., Inverse leidenfrost effect: Levitating drops on liquid nitrogen. Langmuir (2016).

Inverse leidenfrost effect

Thickness of the constant vapor cushion caused by external radiation



Inverse leidenfrost effect

Duration of cooling of a liquid droplet to its melting temperature:

$$t_1 = C_{drop}\left\{(T_0 - T_N)^{\frac{1}{4}} - (T_{Fr} - T_N)^{\frac{1}{4}}\right\},$$

Duration of droplet crystallization:

$$t_2 = C_{drop}\frac{\lambda^f}{c_p}(T_{Fr} - T_N)^{-\frac{3}{4}},$$

Cooling of the beads to the Leidenfrost temperature of liquid nitrogen:

$$t_3 = C_{drop}\left\{\frac{c_p^s}{c_p}\left[(T_{Fr} - T_N)^{\frac{1}{4}} - (T_L - T_N)^{\frac{1}{4}}\right]\right\},$$

Duration of beads levitation:

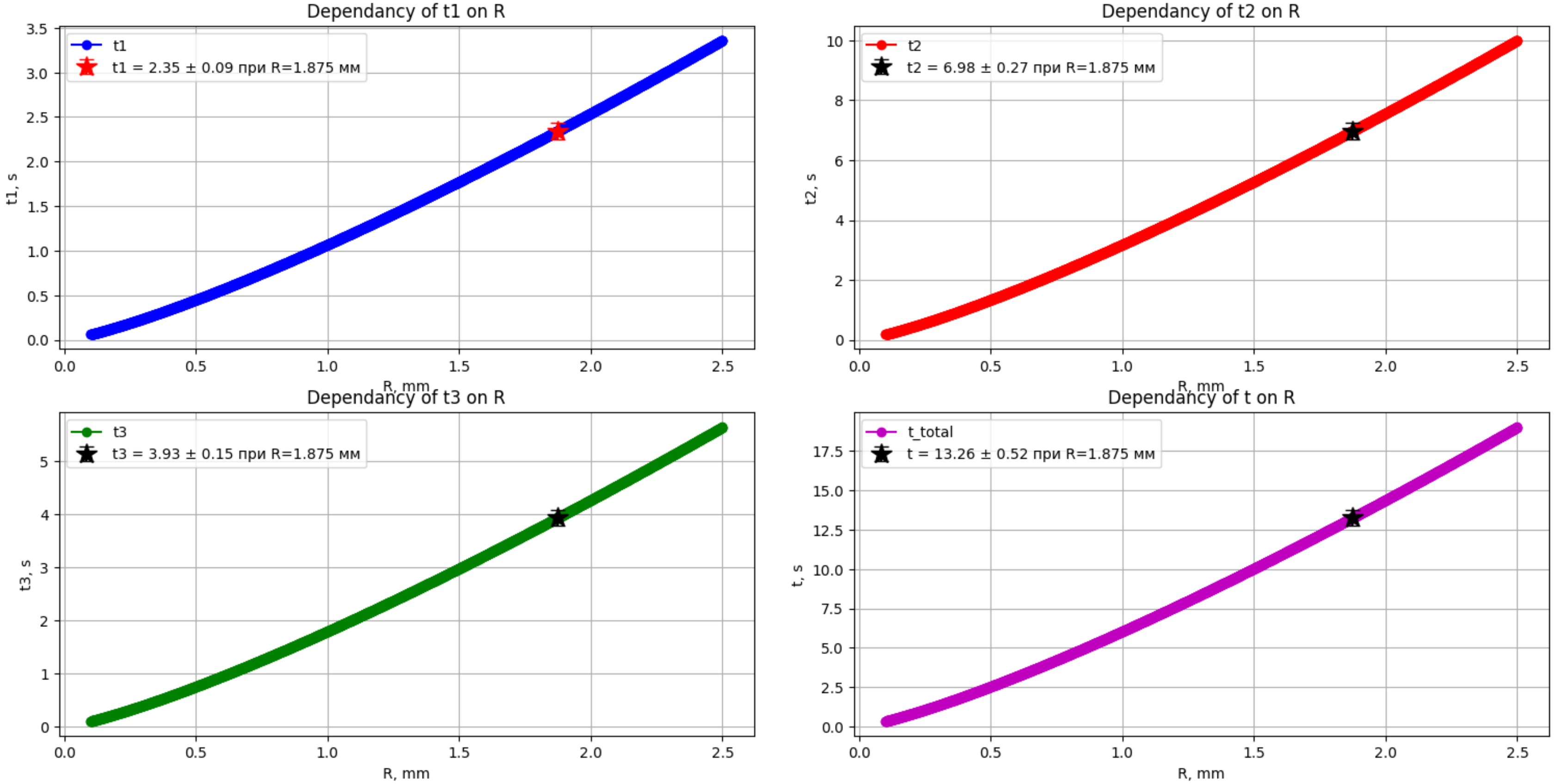
$$t_l = t_1 + t_2 + t_3 = \sum_{i=1}^{3} t_i$$

$$C_{drop} = \frac{4\rho_d c_p R}{3\eta}\left(\frac{9\mu R}{2\rho_d\rho_g g k^3 L^v}\right)^{\frac{1}{4}}$$

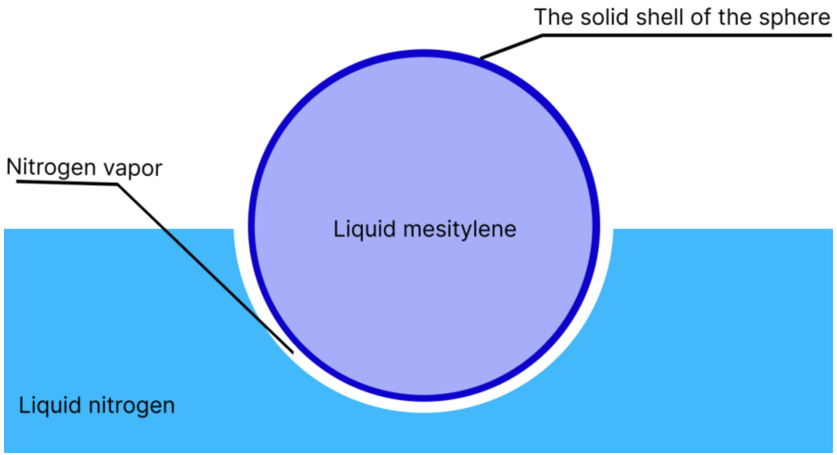
Theory	Experiment
2.35 s	3 +- 1 s
17 s	-
10 s	-
13.26 s	13 +- 1 s

Inverse Leidenfrost Effect: Levitating Drops on Liquid Nitrogen / M. Adda-Bedia, S. Kumar, F. Lechenault, S. Moulinet, M. Schillaci, D. Vella // Langmuir 2016, 32, 17, 4179–4188

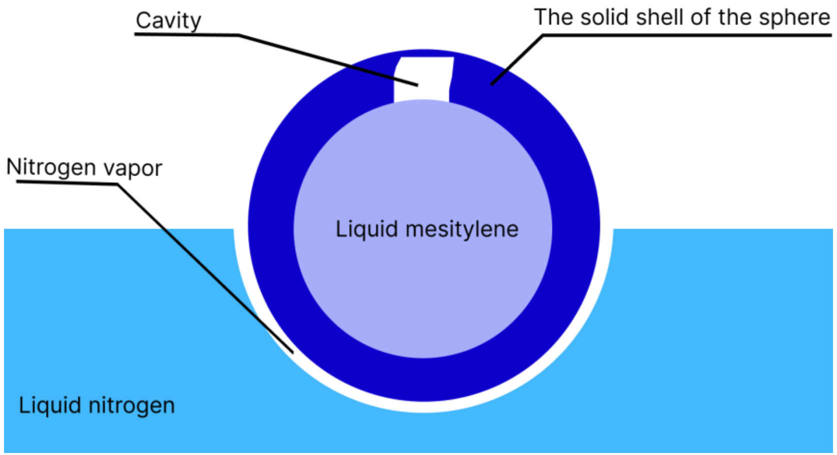
Inverse leidenfrost effect



Cooling and crystallization of a drop



Formation of a solid outer shell



Formation of a cavity inside the beads

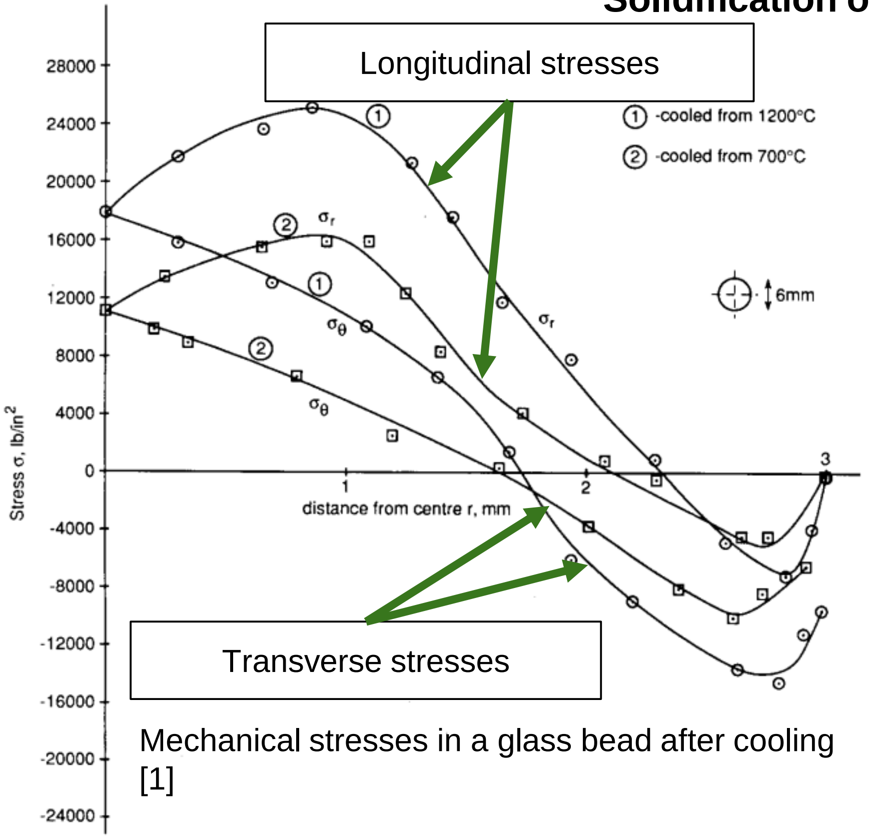
The shape of the bead, the shape of the cavity, and the size of the bead depend on: the intensity of heat exchange, the initial size of the droplet, and the initial temperature of the droplet

The bead will break during production if the heat exchange intensity exceeds the critical value

The size of the bead affects its shape, as in the case of the direct Leidenfrost effect [1]

[1] Geometry of the Vapor Layer Under a Leidenfrost Drop / J. C. Burton and etc. // Physical Review Letters, 2012, 109(7), 074301–617

Solidification of the Inner Layer



Mechanical stresses in a glass bead after cooling [1]

The outer layer solidifies without volume change

↓

Molecules tend to increase the material's density

↓

A mechanical stress gradient forms between the core and the shell

↓

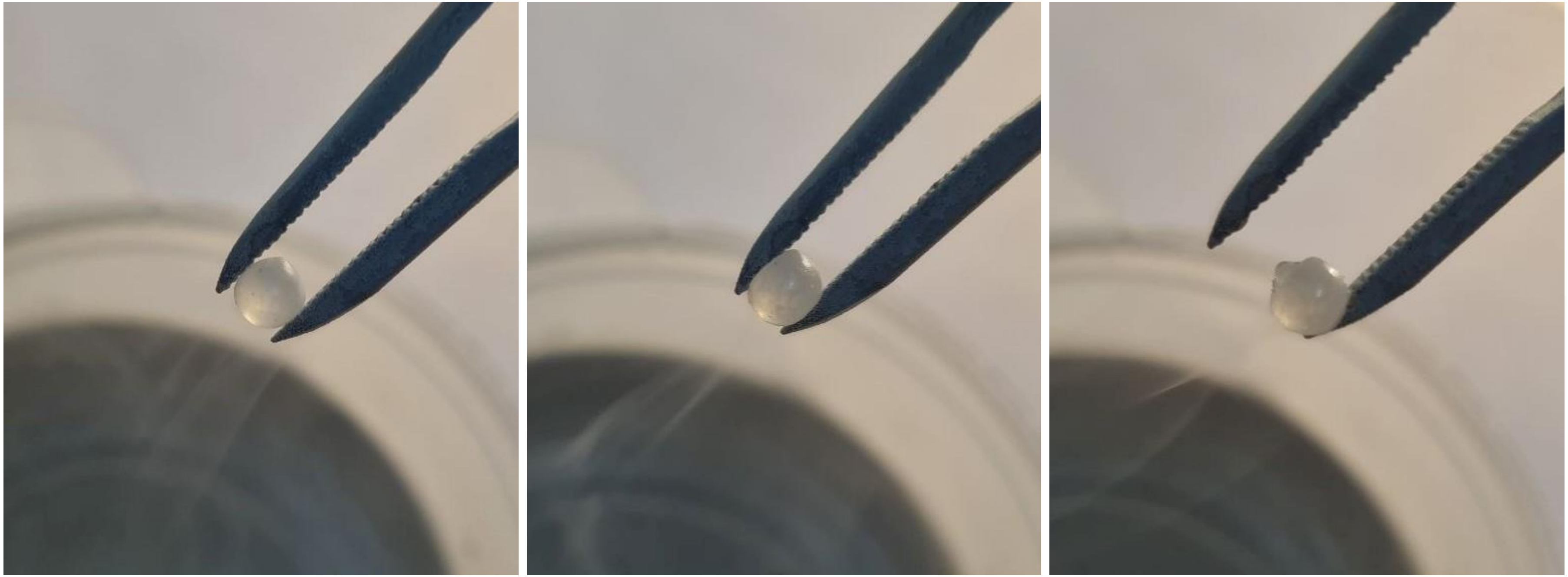
A crack forms. Mechanical stress decreases

↓

A cavity forms as the mechanical stress decreases proportionally to the growth [2]

[1] Residual-stress measurements and calculations and hypotheses for explaining disintegrating fracture // W. Johnson, S. Chandrasekar // Journal of Materials Processing Technology, 31 (1992) 413-440
[2] Площадь свободной поверхности как критерий хрупкого разрушения / Ерасов В.С., Орешко Е.И., Луценко А.Н. // Авиационные материалы и технологии. 2017. № 2 (47). С. 69–79

Adhesive properties of the beads



a) b) c)

Manifestation of plastic properties and high adhesion upon heating a frozen bead made from a mesitylene-m-xylene mixture: a) bead at 80 K in heated tweezers, b) plastic deformation of the bead under tweezers' pressure during heating, c) adhesion of the bead to the tweezers.

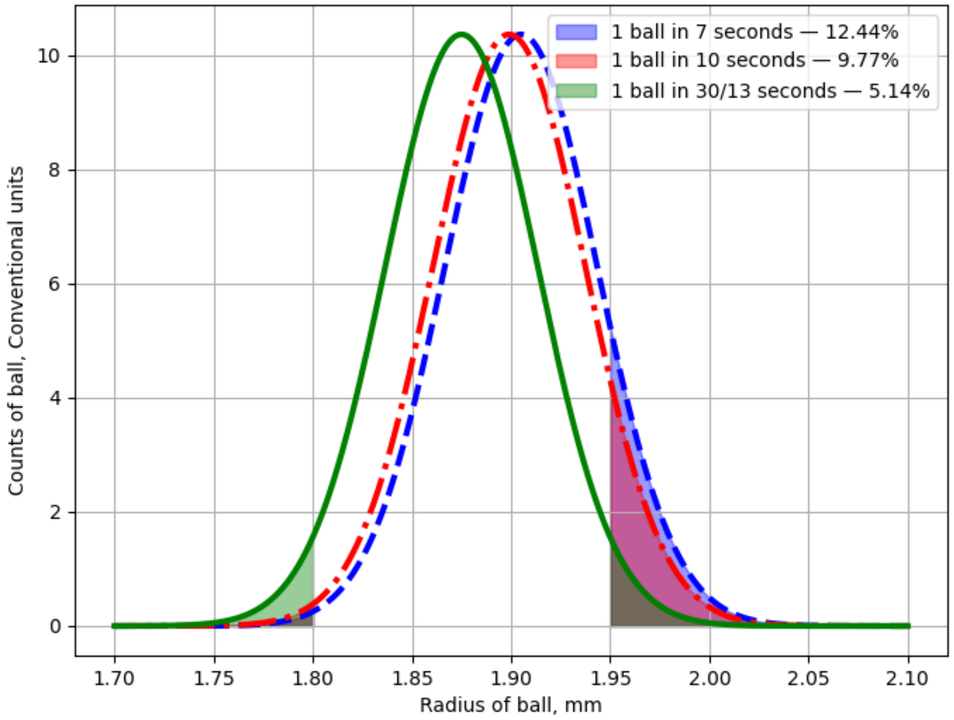
Enhancement of heat exchange between the bead and the vapor cushion

Parameters for the production of balls from a mixture of 280 mL.

Parameter	Value		
Duration of production of one ball (s)	30/13	10	7
Voltage and current supplied to the nichrome wire (V, A)	0, 0	15.69, 2	23.69, 3
Production time (h)	6	3	2
Frequency of adding liquid nitrogen (times/h)	1	3	3
Defective balls with a diameter greater than 3.9 mm (%)	2.5	8.5	12.5
Defective balls with a diameter less than 3.6 mm (%)	2.5	1.5	0.5

Enhancement of heat exchange between the bead and the vapor cushion

Schematic representation of the size distribution of balls at different levitation times. The shaded areas denote balls with diameters smaller than 3.6 mm and larger than 3.9 mm.



Comparison of Conventional and Automated Devices

	Conventional Device	Automated Device (Production is nearing completion)
Productivity	50 mL/h	1 L/h
Device preparation and shutdown	About 20 minutes	A few minute
Maintenance frequency	After producing a batch (1/3 liter of beads) + refilling liquid nitrogen every hour	Not required
Employee safety	Contact with liquid nitrogen and mesitylene mixture	Button press, remote control
Control of defective beads	After batch production	In real-time
Nitrogen-free storage capability	Not available	Possible

1 person per 4 devices (5 employees)
Variations in bead shape and size = potential issues with the dispenser
18 conventional devices < 1 Automated Device

Emergency bead production possible

Absence of employee contact with liquid nitrogen and the mesitylene mixture, as well as a significant reduction in routine work.

Thanks