The Influence of Chemical and Mechanical Effects on the Stability and Strength of Formvar Tents

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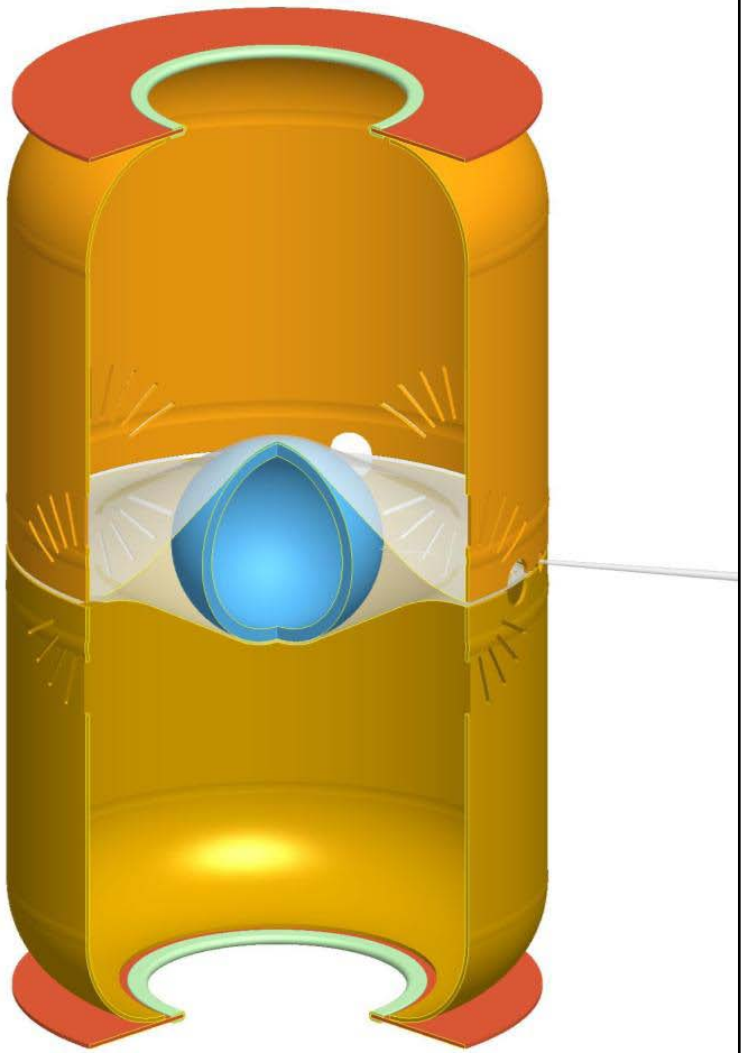
Polymer tents are used to mechanically support the fuel capsule within the hohlraum

Schematic of Hohlraum, capsule & tent

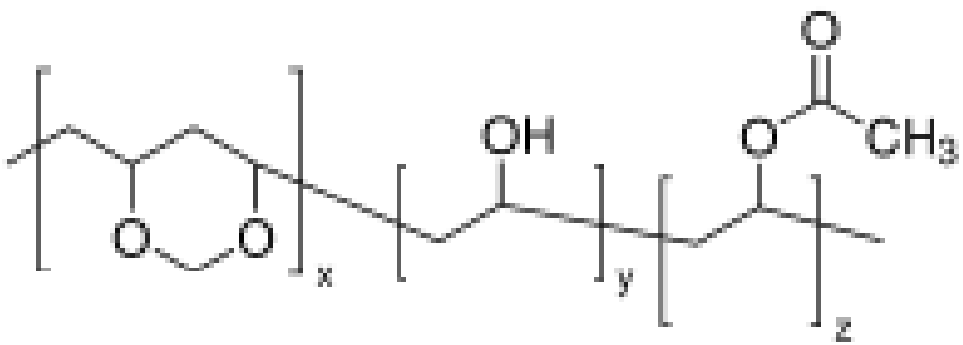


Tents have stringent mechanical and performance specifications

- **Tent deflection without tearing: >1 mm**
- **Typical tent thickness ≈110 nm**
- **Capsule centering: –10 µm axial, 12 µm radial**
- **Low mass, and low Z material**
- **Holes (for gas equilibration) – size/location vary with target type**

Formvar is used as the baseline tent material, at LLNL, due to its high elongation and ease of fabrication

Formvar is polymer containing three functional groups



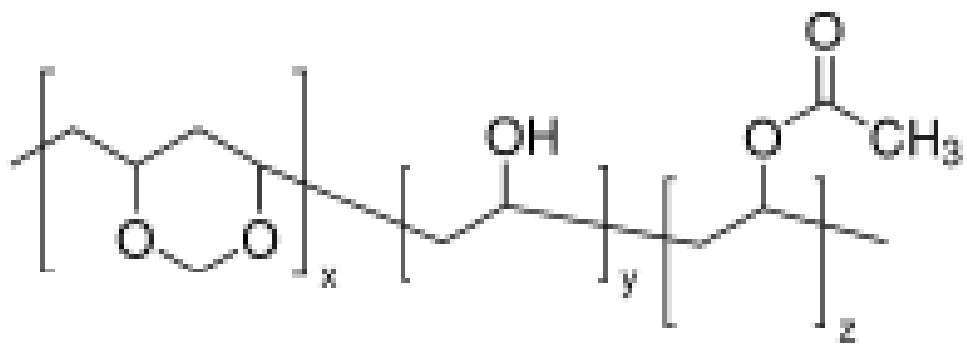
Polyvinyl Acetal **Polyvinyl Alcohol** **Polyvinyl Acetate**

Material	Elongation (Yield)	Elongation (Break)	Tensile Strength (MPa)	Flexural Strength (MPa)	Water Abs in 24 hrs	Molecular Weight $x10^3$
Formvar 15/95	7%	50%	42-59	117-124	1.2%	24-40

* All values from Monsanto data sheet

The successful use of Formvar, as a tent material, depends on both mechanical and materials issues

Formvar



- **Mechanical effects**
- **Material performance**
  - **The length of the polymer chains (e.g. x + y + z)**
  - **The relative distribution of monomers along the polymer chains** $\left[\frac{x}{(x+y+z)}, \frac{y}{(x+y+z)}, \frac{z}{(x+y+z)}\right]$
  - **Intermolecular forces (factors which would enhance or retard the slippage of polymer chains past each other)**

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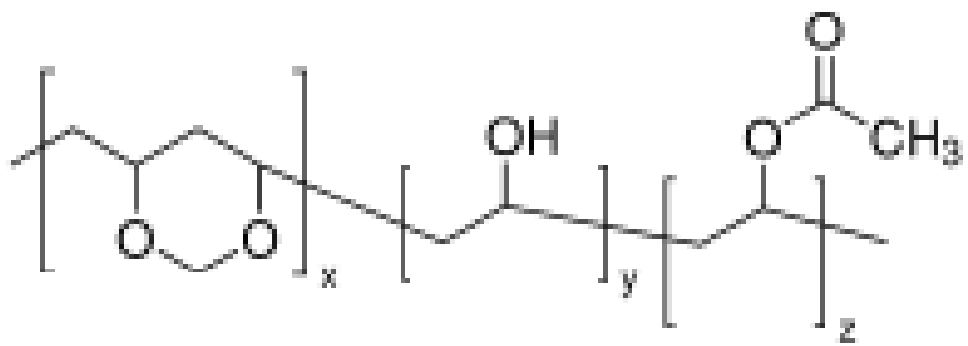

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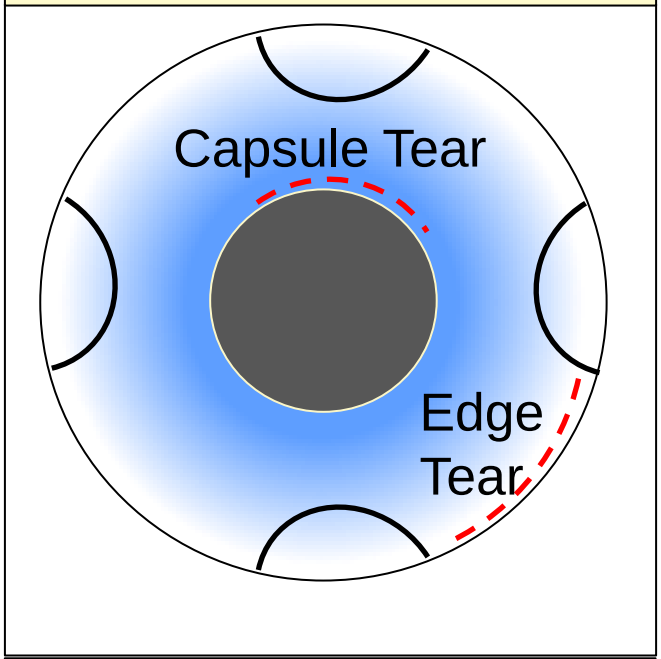
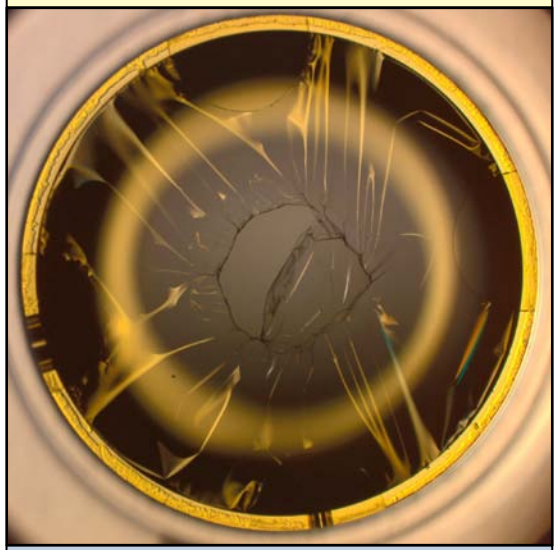
The mode of tent failure indicates whether performance is limited by material issues or mechanical issues

Typical types of tent failures



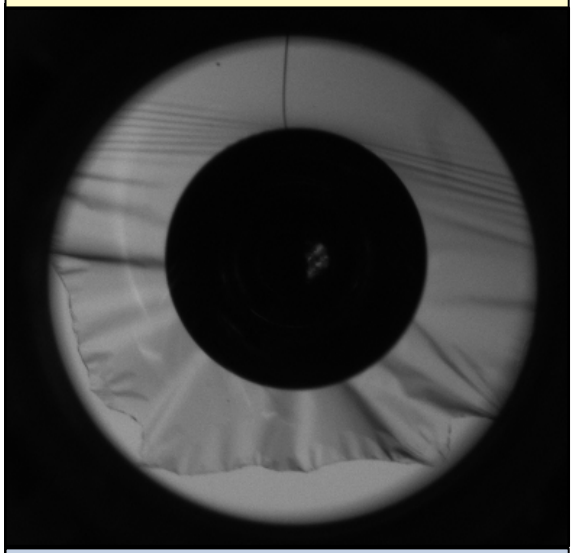
During closing, forces should be highest in the center

Tent failure at capsule



Indicative of material failure

Tent failure at edge

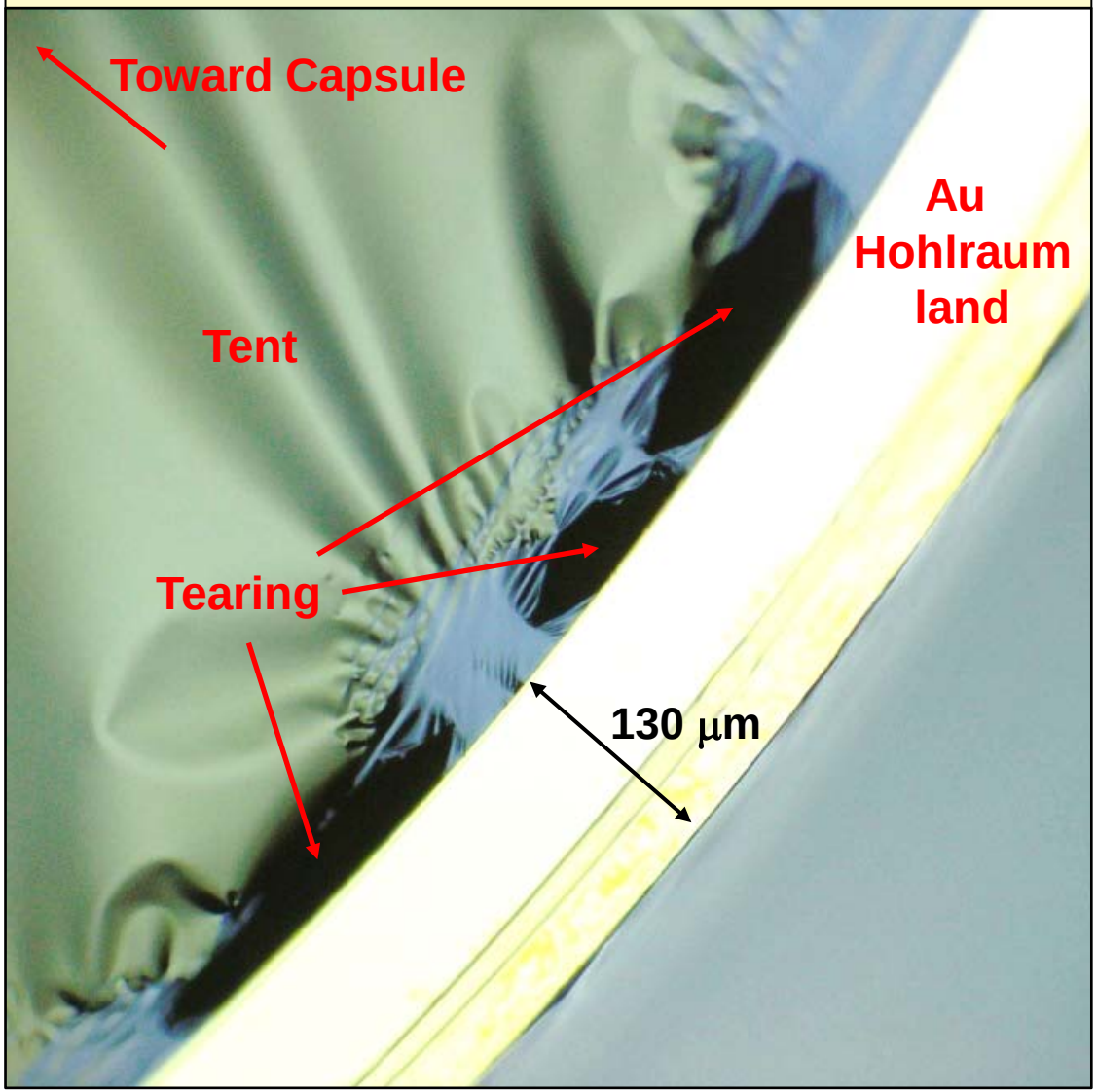


Indicative of mechanical issues

In a well designed and executed system, tent performance will be limited by material properties, rather than mechanical properties

Edge failures suggest that mechanical, rather than chemical effects are limiting tent performance

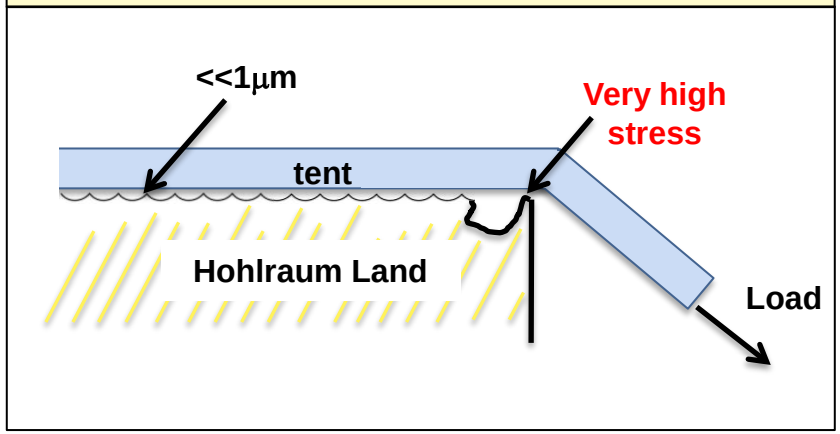
Top view of tent tear near Au hohlraum land edge



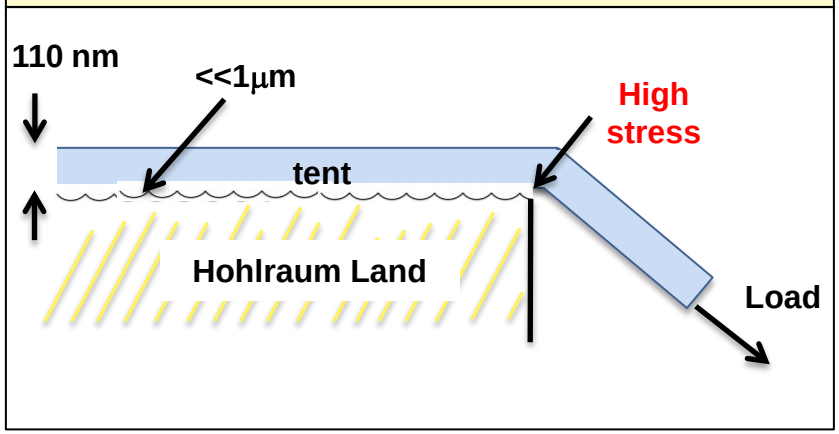
Asperities on Hohlraum lands can lead to a stress rise and thus lead to tent ripping

High edge stress sources

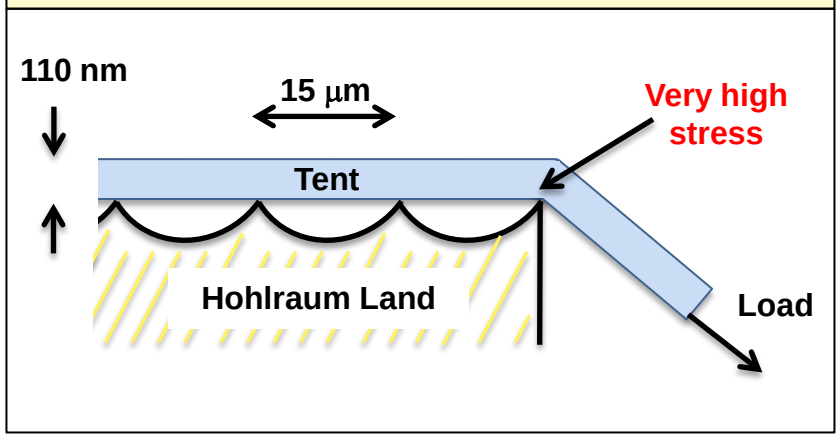
1) Land damage during insertion



2) Sharp edge by design

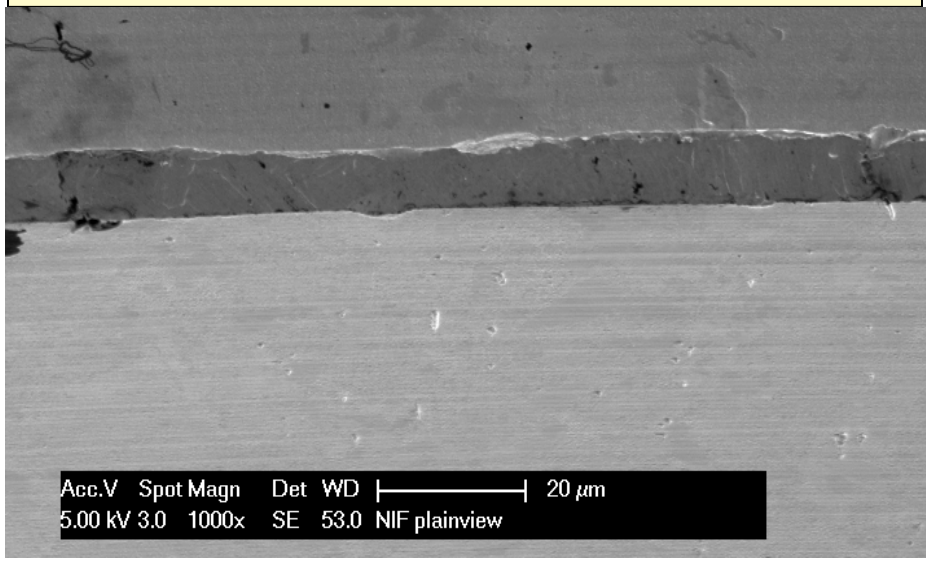


3) Manuf. tooling marks

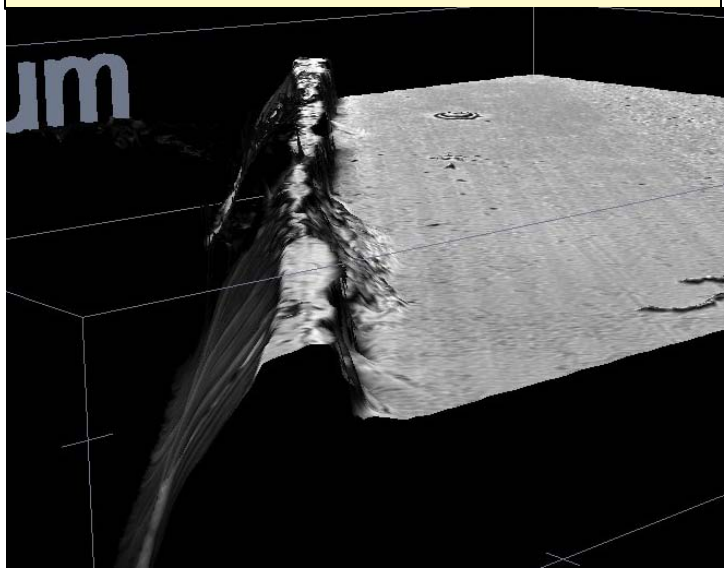


Sharp edges can also be caused by created by the interaction of the Hohlraum with assembly tooling

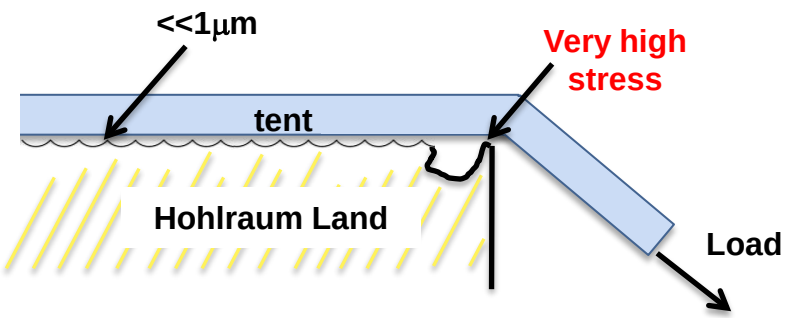
Ridges created from damaged tooling can be 0.5 mm long



Leica image shows ridges are 2.5 µm tall

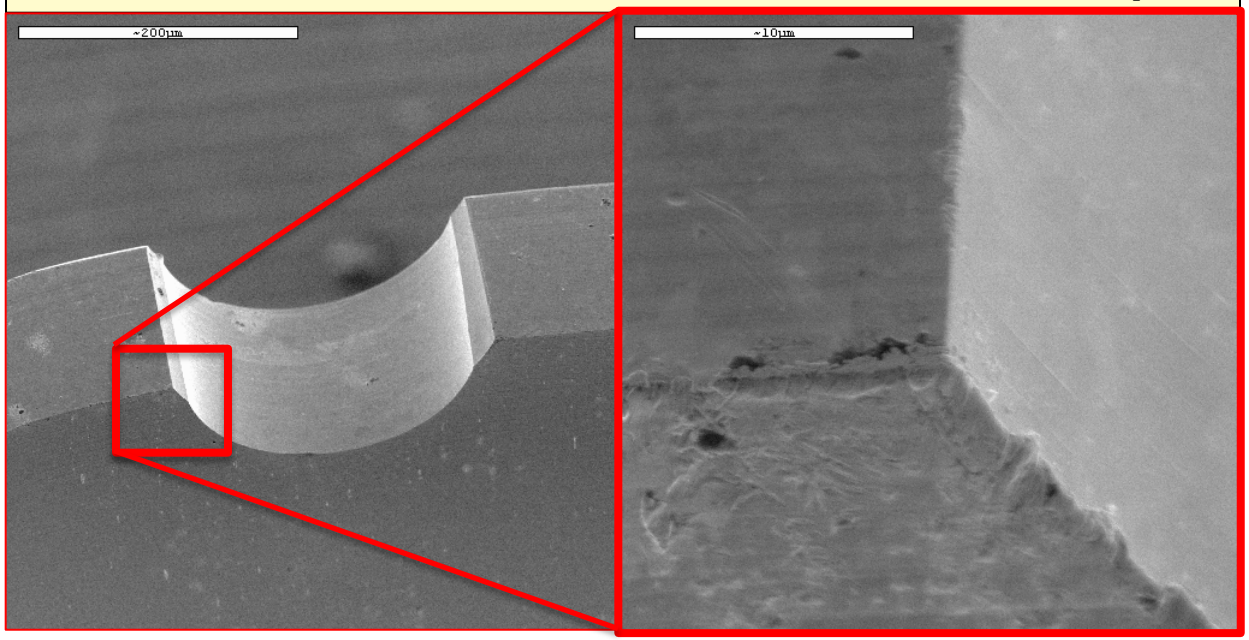


Land damage during insertion

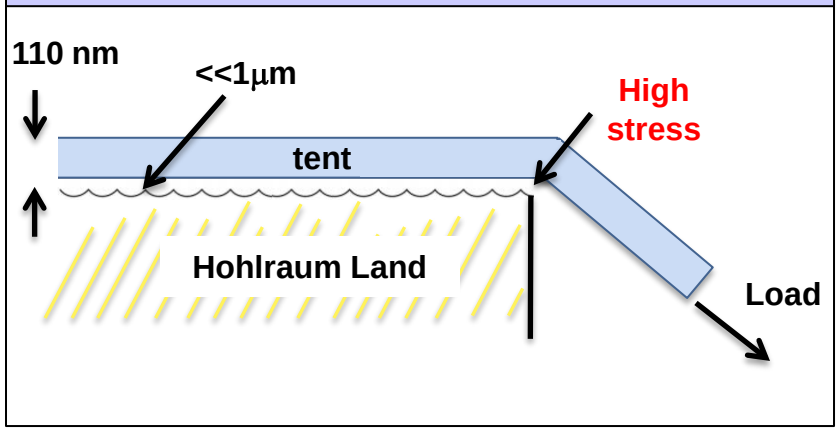


The details of the mechanical contact between the hohlraum and the tent are important

Radius of curvature on backmachined HRs is 1 μm



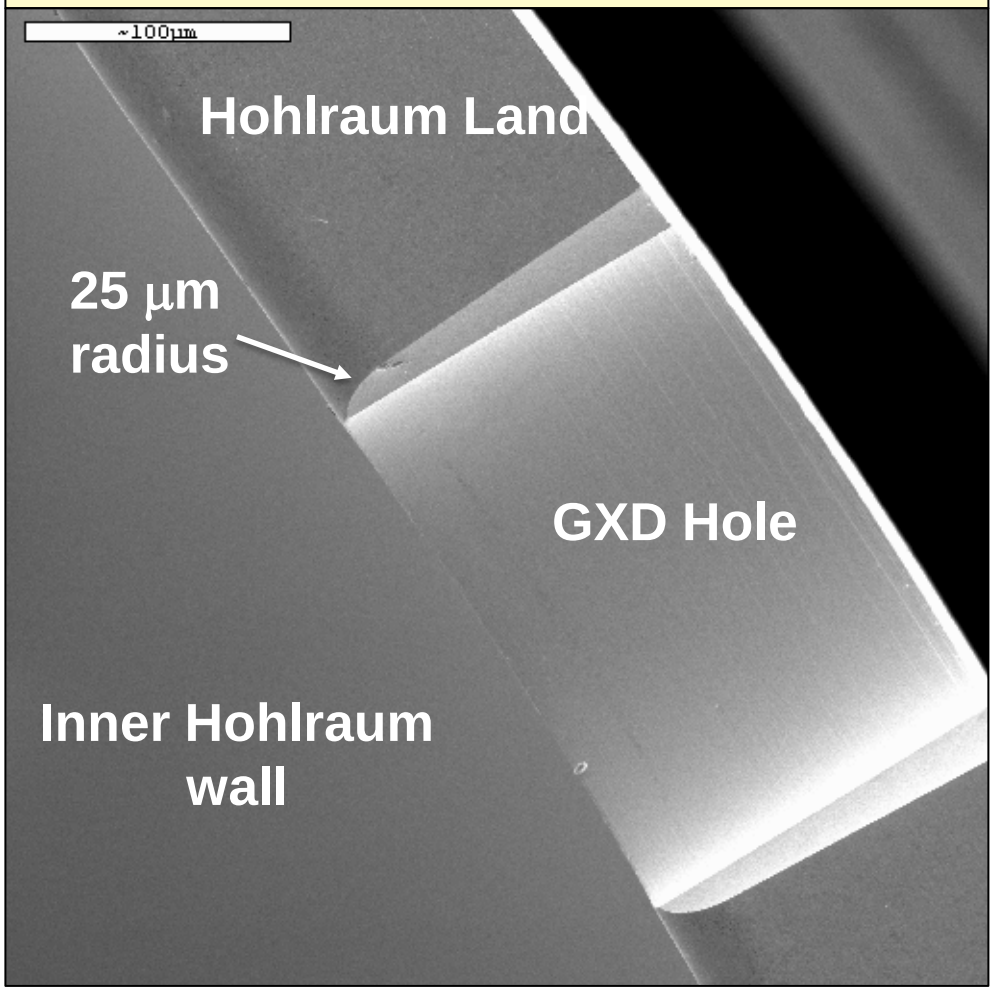
Early hohlraum designs had a very sharp edge



A sharp land, and localized asperities present an opportunity for failures of the tent material at the edge of the hohlraum

Radiused hohlraums & improved Hohlraum insertion tooling are being used to mitigate asperity-induced tent failures

SEM image of radiused hohlraum land



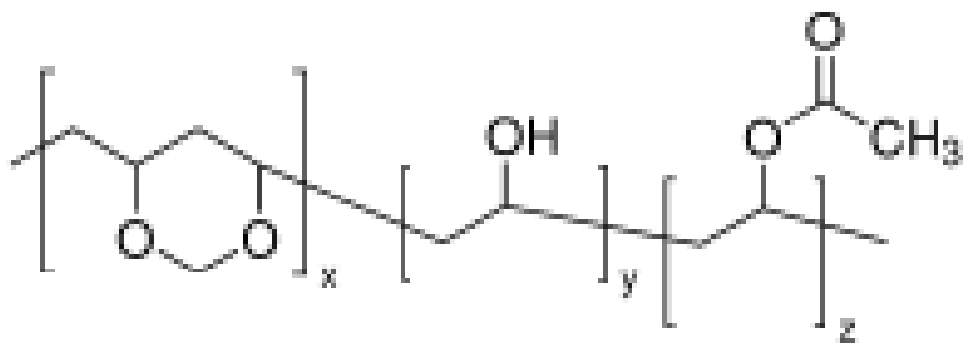
- Offline testing was used to qualify the used of radiused hohlraums
- Change in failure mode suggests stress rise at edge has been reduced

	Failure Mode
Radiused Hohlraums	100% Center
Sharp Hohlraums	44% Center 56% Edge

- Radiused Hohlraums have been in full production since Jan 2011

The successful use of Formvar, as a tent material, depends on both mechanical and chemical issues

Formvar



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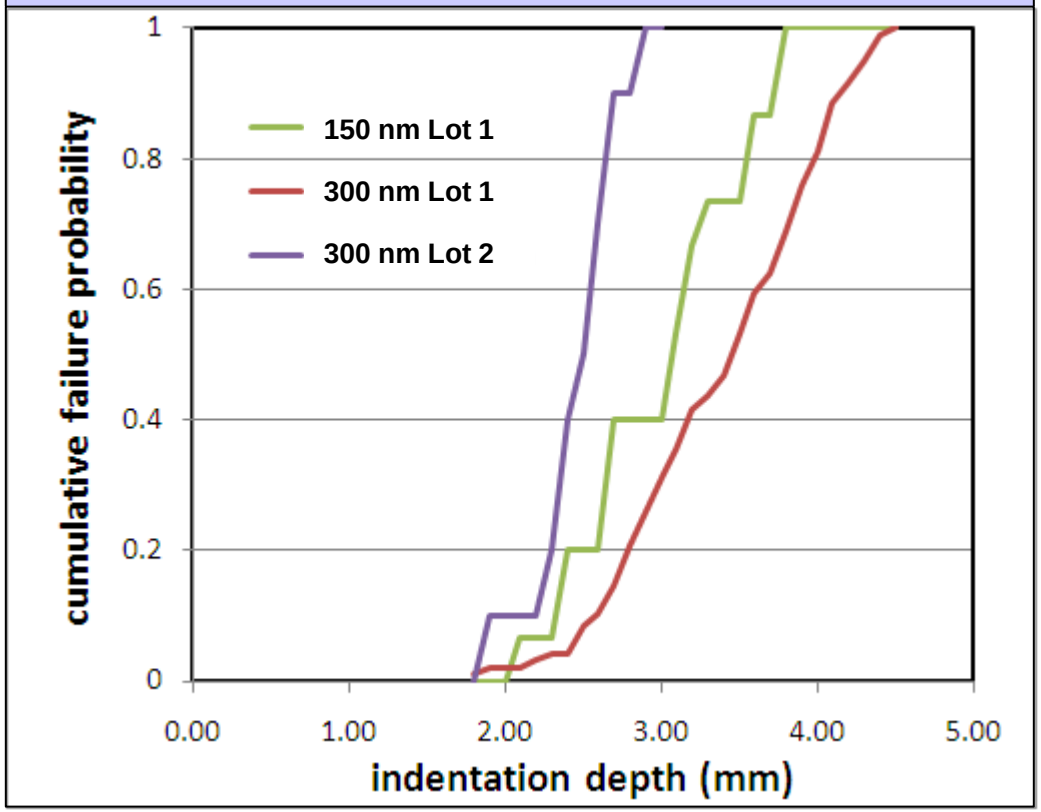
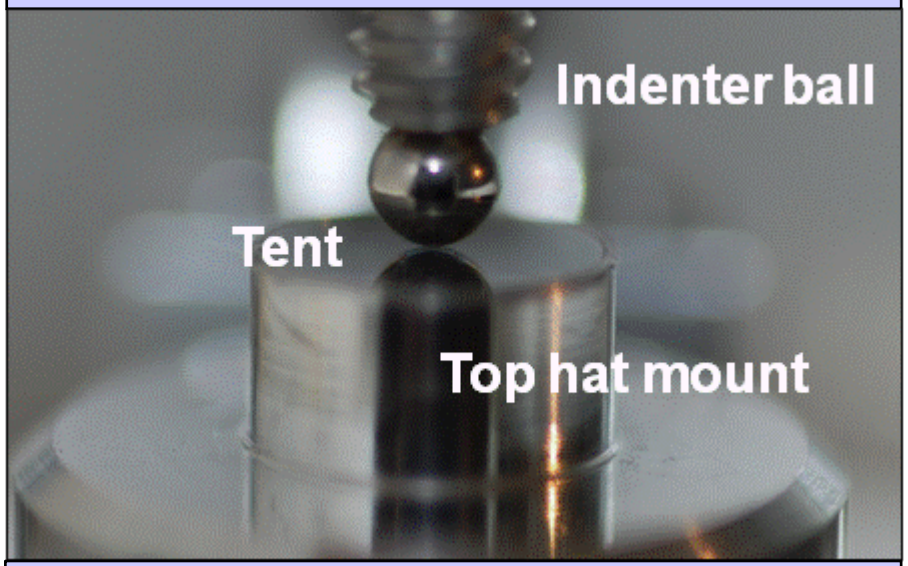
Mechanical testing reveals consistent lot to lot variations in the performance of the polymer used for tent fabrication

Cumulative failure probabilities for two lots of material



Testing is used to characterize material strength prior to use

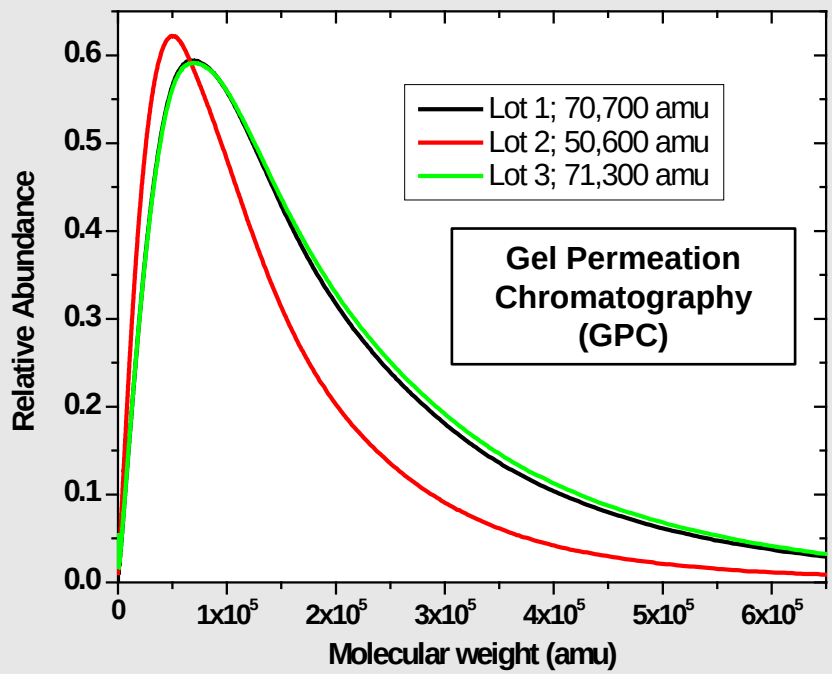


Deflection at failure as well as modulus, visco-elasticity, are characterized

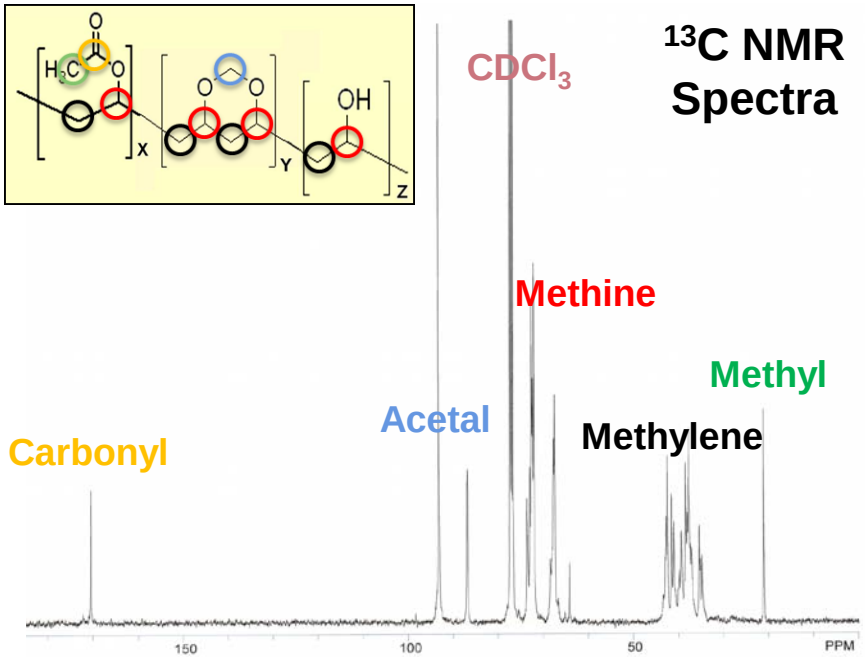
Lot to lot variations in material used for the fabrication of NIF targets is controlled by characterization prior to use

Mechanical properties of polymers can vary as a function of the molecular weight and functional groups

Molecular Weight Distribution



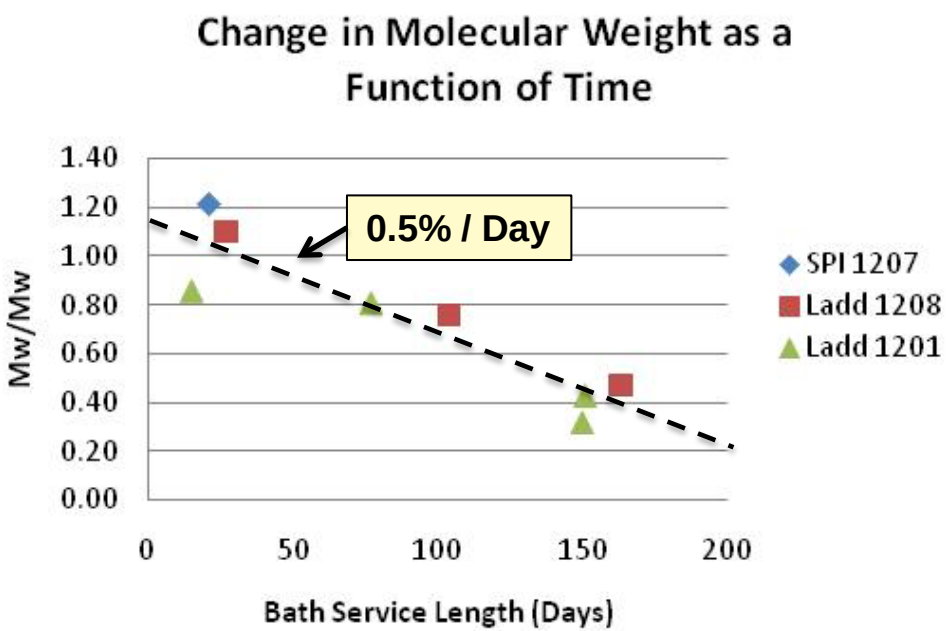
Distribution of Copolymers

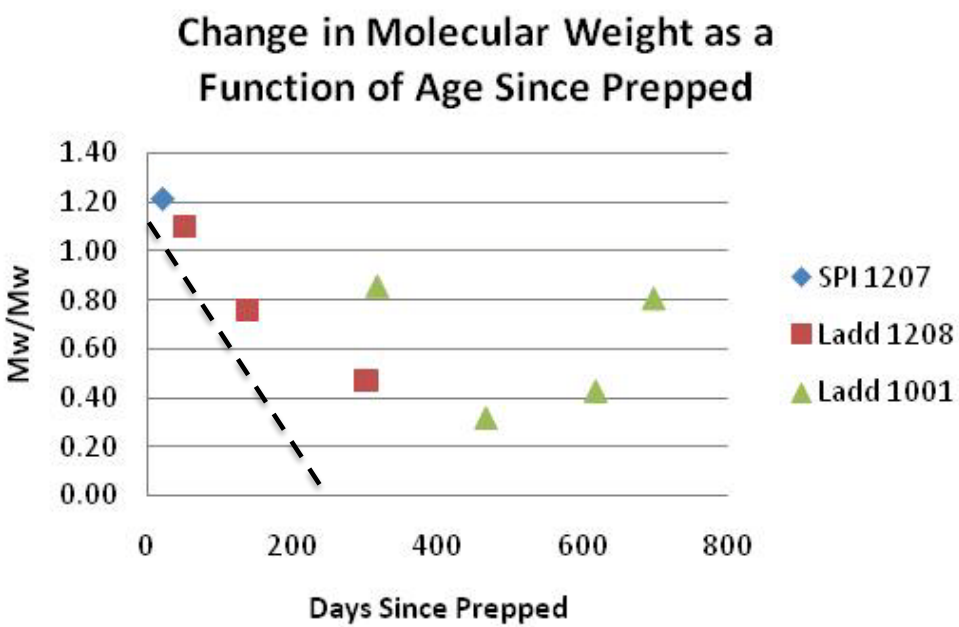


Batch	Designation	Acetate (x)	Acetal (y)	Alcohol (z)
Lot 1	Ladd 1001	14.6%	78.6%	6.8%
Lot 2	Ladd 1208	13.8%	78.7%	7.5%
Lot 3	SPI 1207	15.2%	78.1%	6.7%

The principle difference see between the Formvar and the remaining lots of resin is the lower molecular weight

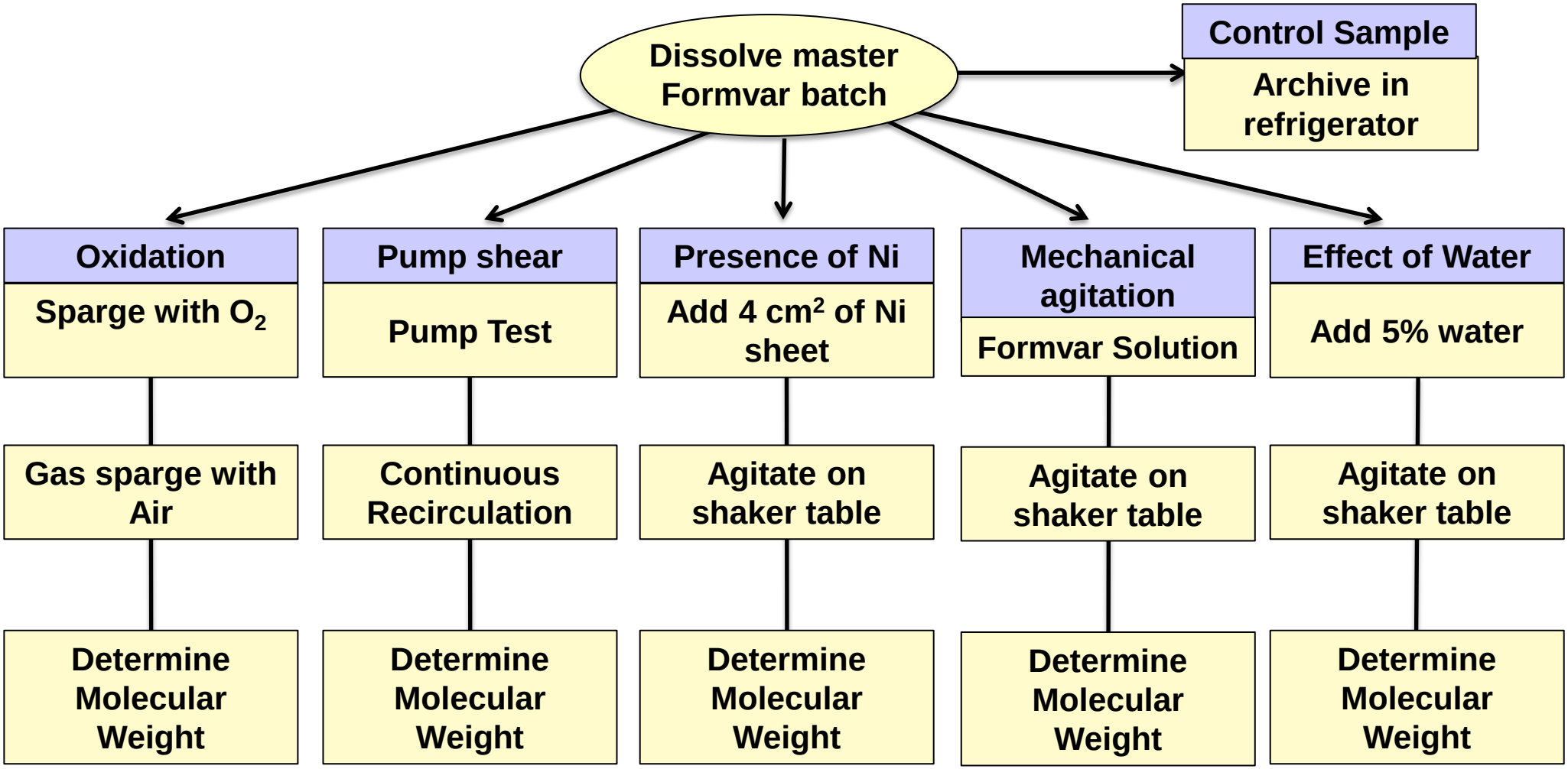
We have also, found that the molecular weight of the Formvar in the coating baths decreases as a function of time



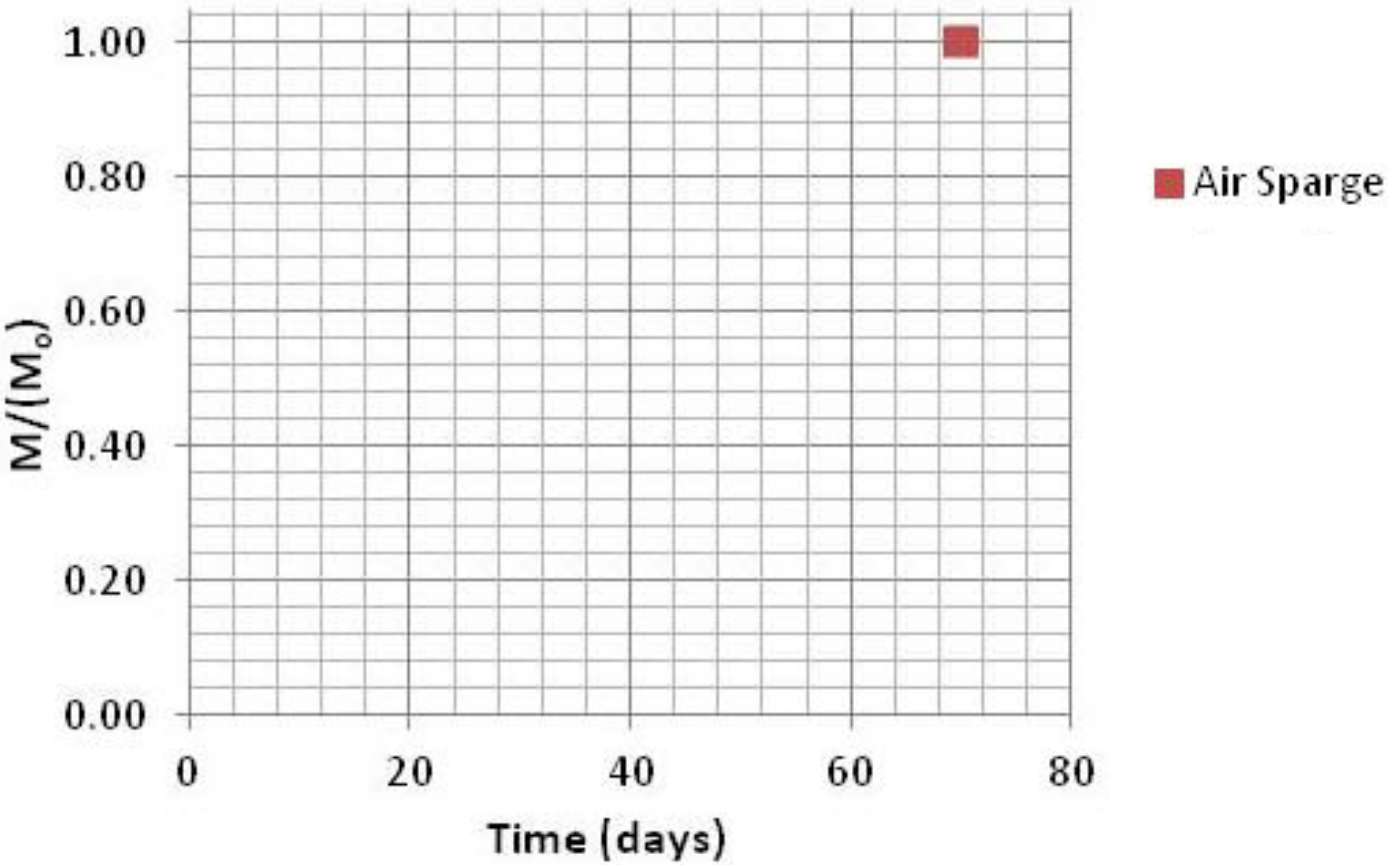


The reduction in the molecular weight of the polymer appears to be correlated to time in the bath, rather than simply chronological age

Several environmental factors *might* cause polymer chain scission; which is responsible?

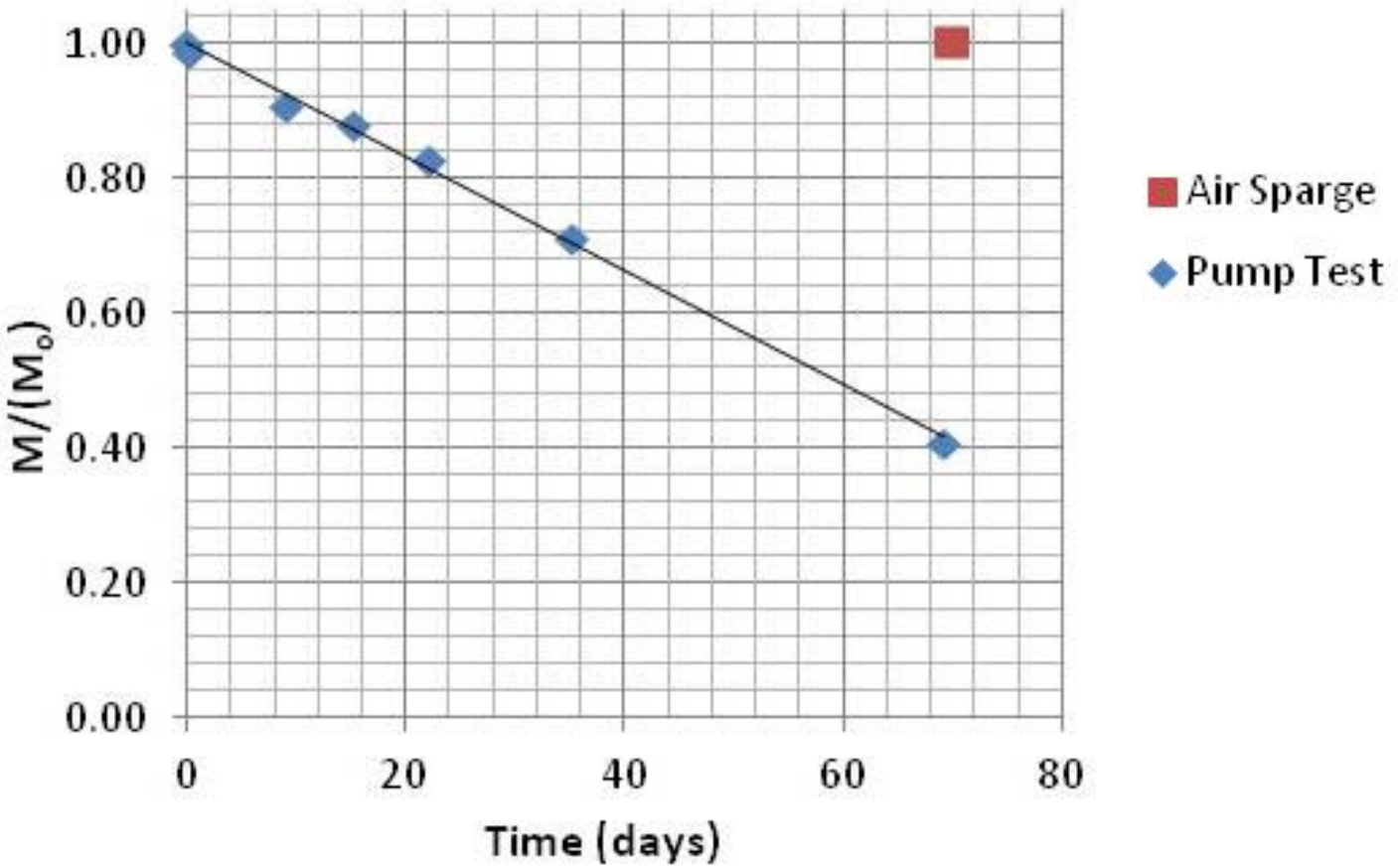


Air sparging was used to test the polymer stability relative to oxidation



No degradation the molecular weight, due to air oxidation, of the polymer was seen over a two month period

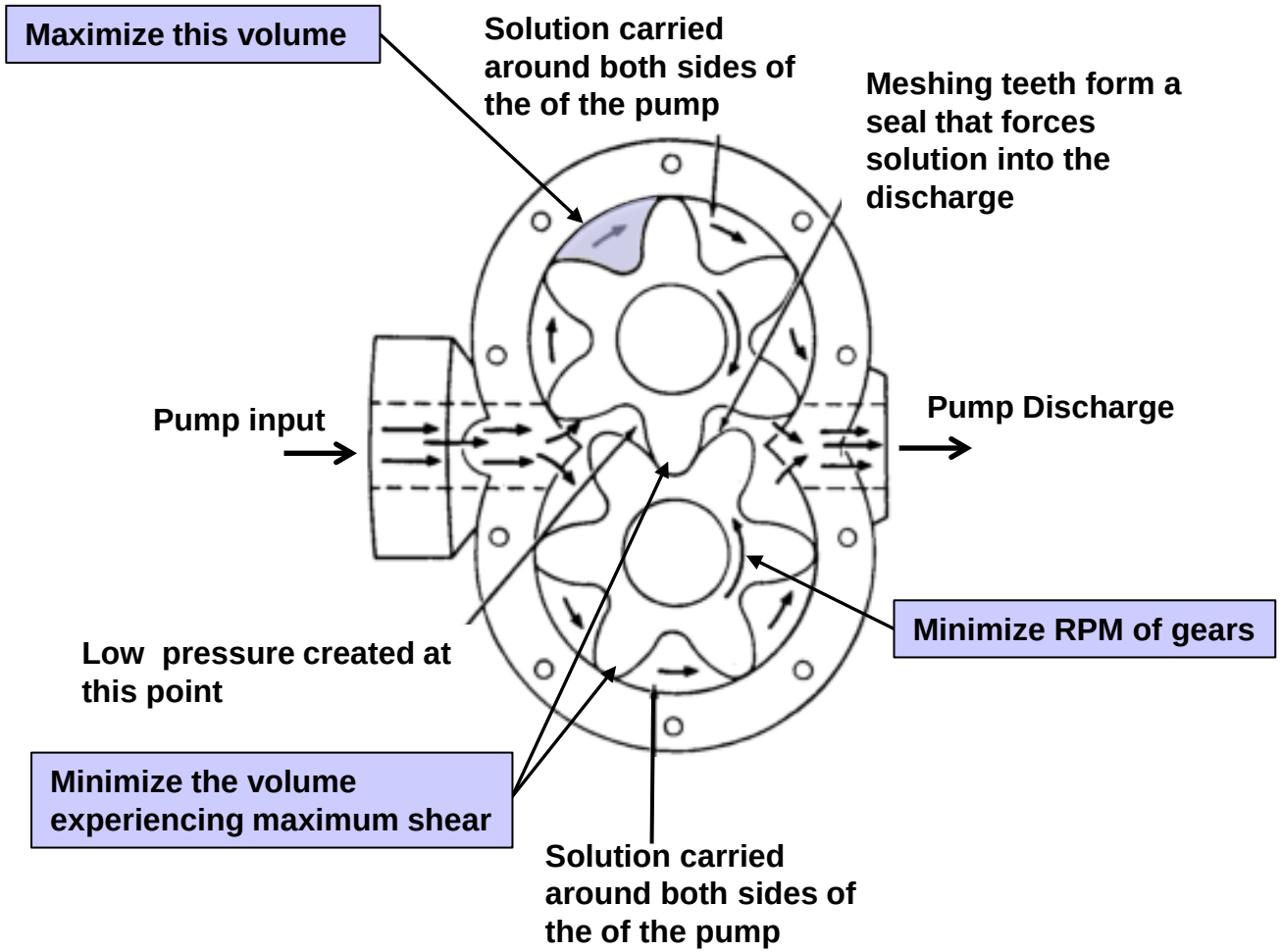
Pump shear leads to degradation of the molecular weight of the Formvar polymer



The rate of shear establishes a limit on the lifetime of solutions that are used for tent fabrication

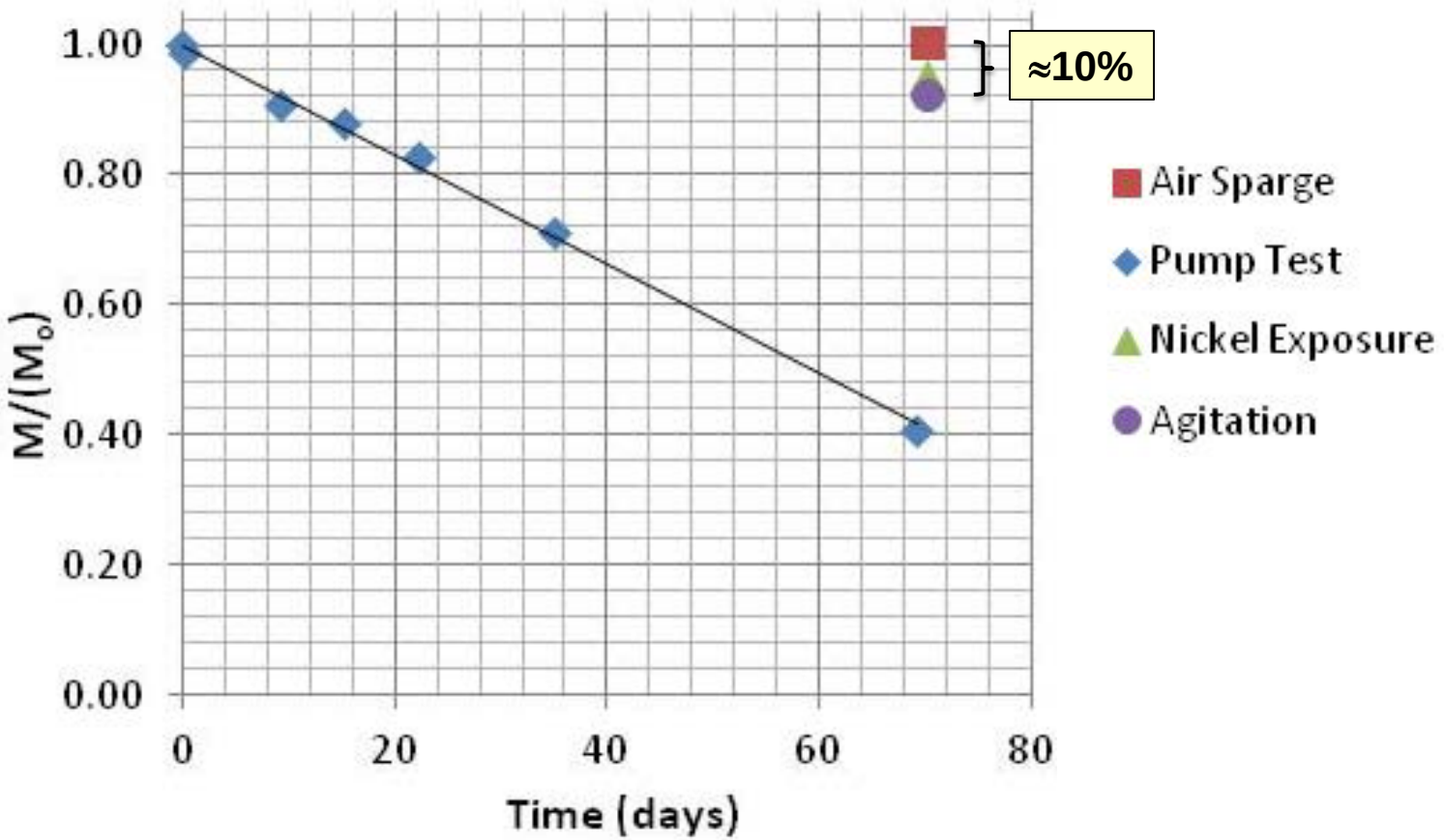
Pump shear leads to degradation of the molecular weight of the Formvar polymer

Gear Pump used to circulate polymer solution

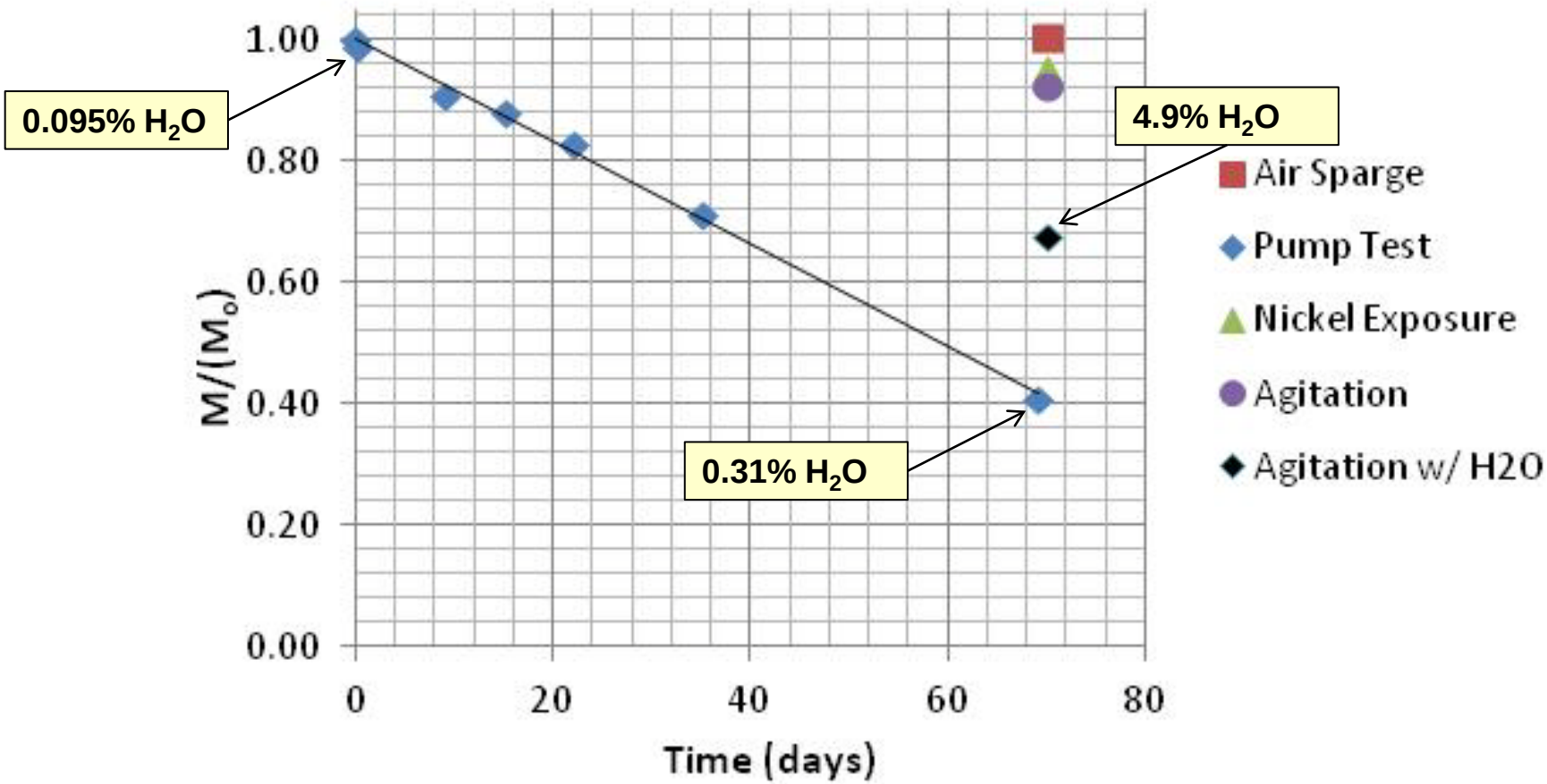


The rate of shear establishes a limit on the lifetime of solutions that are used for tent fabrication

The shear associated with even simple agitation is enough to slightly degrade the polymer's molecular weight



In addition to shear hydrolysis, upon exposure to water, also leads to a loss in the molecular weight of the polymer



In addition to the degradation in molecular weight, hydrolysis will increase the fraction of polyvinyl alcohol and decrease the fractions of polyvinyl acetate and polyvinyl formal in the polymer

These results suggest that spin coating, may be an attractive alternative to meniscus coating

- No pumping system is required
  - Concerns about to molecular weight degradation due to shear are avoided
  - Concerns regarding solution carry over, changes in solution concentration and system dead volume are avoided
- Solutions are stored under dry, inert conditions alleviating concerns about molecular weight degradation of the coating solutions due to hydrolysis
- Small foot print / standard commercially available equipment
- Fast straight forward processing

Conclusion and Summary

- Careful attention to the interaction between the mechanical aspects of the hohlraum and the performance of the tent material are required for successful fielding of tent materials for NIF targets

- Mechanical effects of importance include
  - Mechanical design of the hohlraum edge
  - Fabrication details
  - Assembly and fixturing details

- Intrinsic properties of the tent material that have also been found to be of importance include variation in the molecular weight of the polymer arising from
  - Lot to lot variations
  - Degradation due to pump shear
  - Degradation due to hydrolysis

NIC

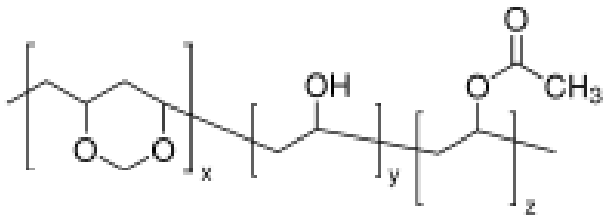
Other candidate materials are being considered as a back up

Material	Modulus (GPa)	Failure strain	Material commercially available	Processing capabilities available	Time to readiness
Formvar 15/95	2.8-3.1	50%	Yes	Yes	Ready
Butvar	1.9-2.3	70-110%	Yes	Yes	4-6 months
Enhanced Formvar*	>2.9	>50%	No	Yes	6-9 months
Polyimide (Luxel)	3.2-7.5	8-40%	Yes	No	6-12 months
Single sheet Graphene	1000	19%	No	No	Very long

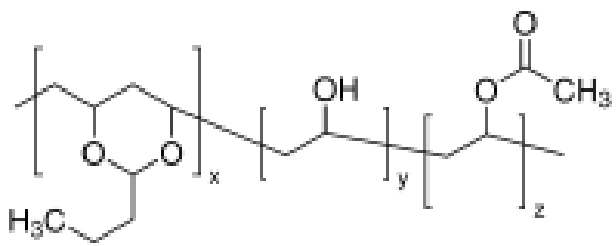
*Enhanced Formvar would have larger molecular weight than commercial Formvar

Selected properties of Formvar and Butvar Resins

Formvar



Butvar



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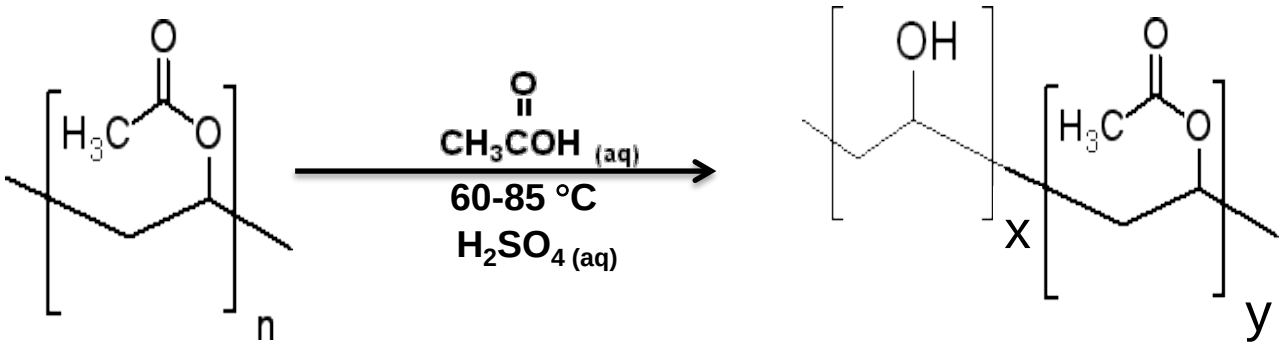
Formvar 15/95	7%	50%	42-59	117-124	1.2%	24-40
Butvar B72	8%	70%	48-55	83-90	0.5%	170-250
Butvar B76	8%	110%	32-39	72-90	0.3%	90-120
Butvar B90	8%	100%	39-46	76-83	0.5%	70-100
Butvar B98	8%	110%	29-46	76-83	0.5%	40-70

Butvar resins potentially offer considerable advantage with respect to elongation and water absorption compared to Formvar

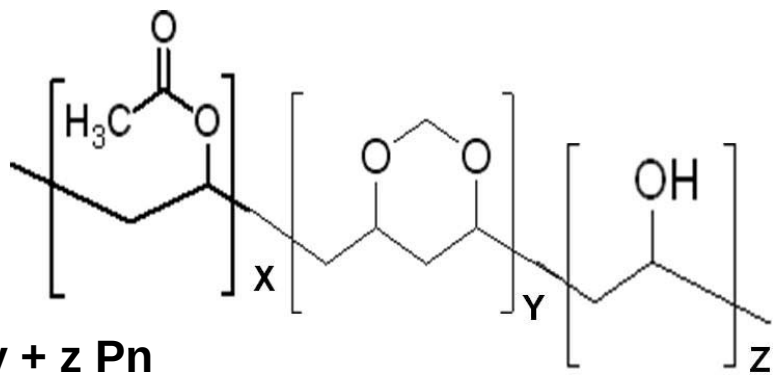
Formvar is synthesized from polyvinyl acetate using an acid catalyzed reaction involving formaldyde in aqueous media

First acid catalyzed hydrolysis is used to hydrolyze a fraction of the acetate groups on the polymer background

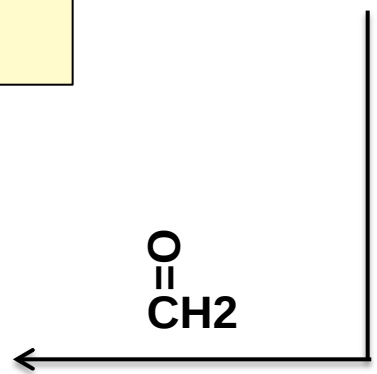


x+y=n

Second a fraction of the resultant alcohol groups are acetalized with formaldehyde



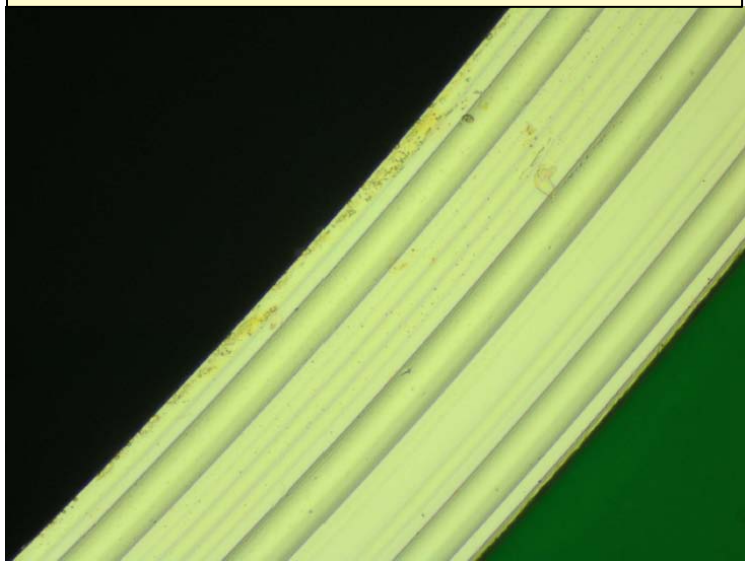
x + y + z Pn



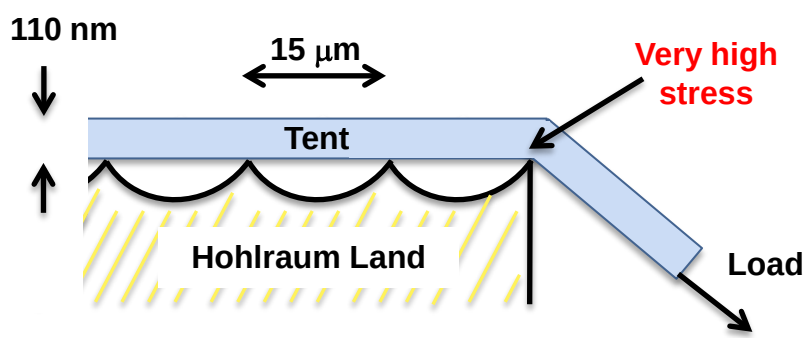
Altering the ratio of acetic acid, water and formaldehyde used during synthesis would allow the composition of the monomers within the polymer to be modified to maximize material performance

The hohlraum fabrication can introduce asperities that can lead to premature tent failure

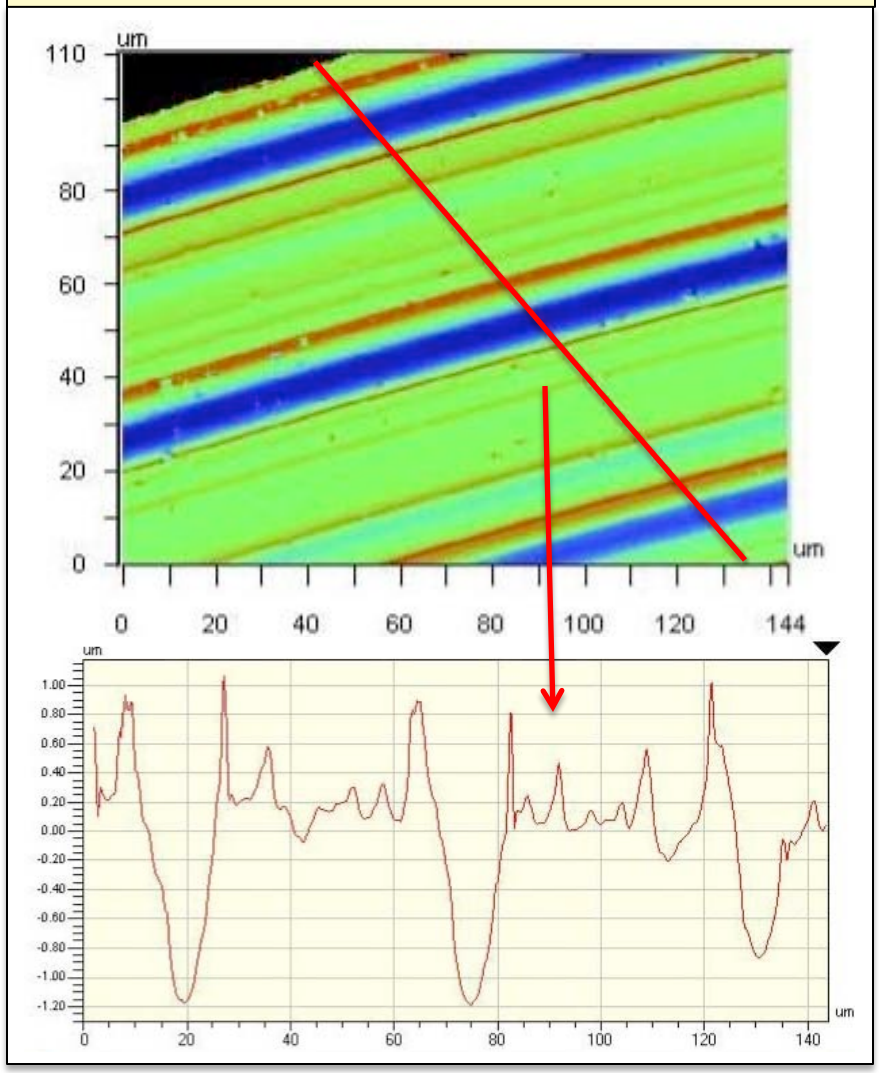
Grooves created by change of machining program



Grooves enhance edge sharpness

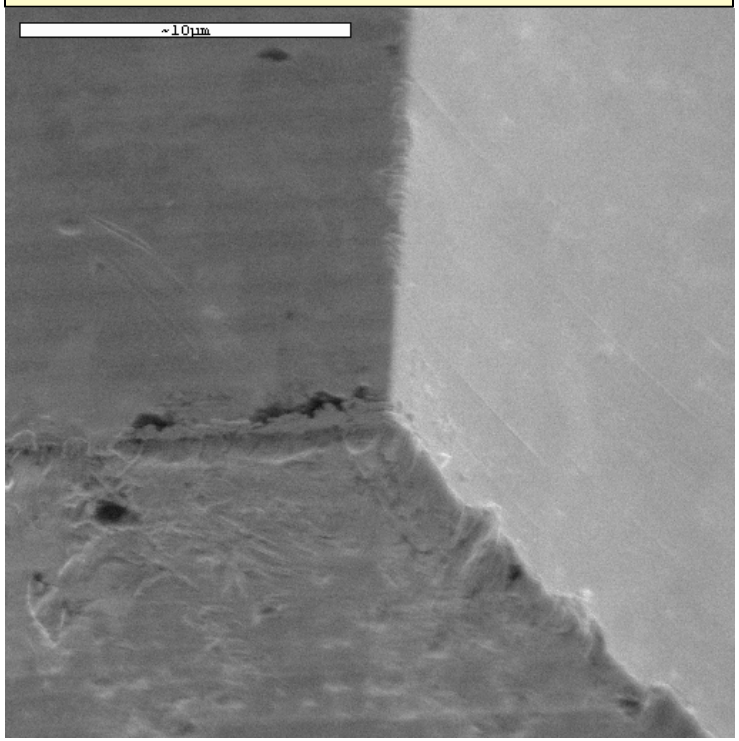


White light interferometry shows groove height of 1.5 µm

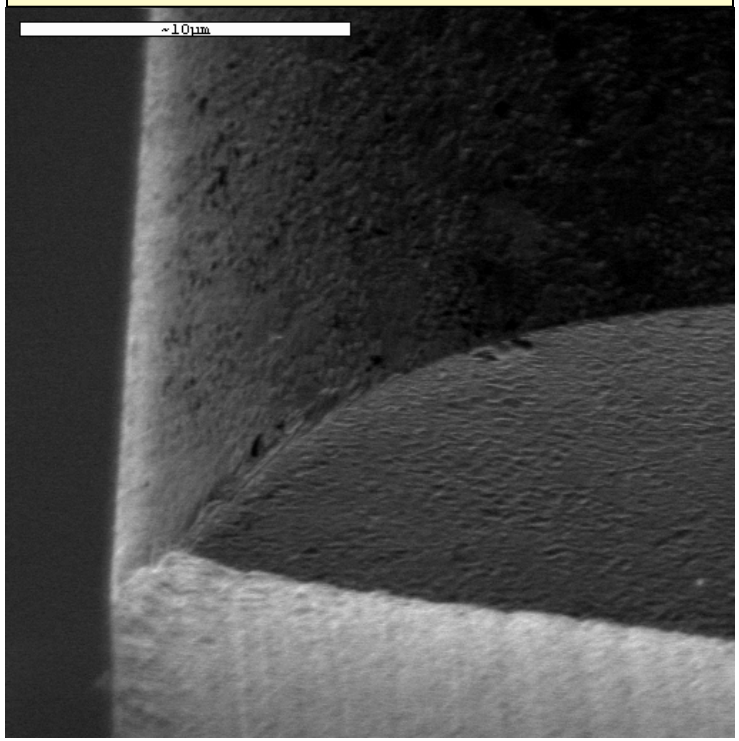


Radiused Hohlraums & improved Hohlraum insertion tooling mitigate asperity-induced tent failures

Original HR edge



HR with 25 µm radius



- **radius increases edge stability, decreases edge stress rise**
- **improved tooling does not touch inner edge**
- **tooling is now monitored and cleaned regularly**

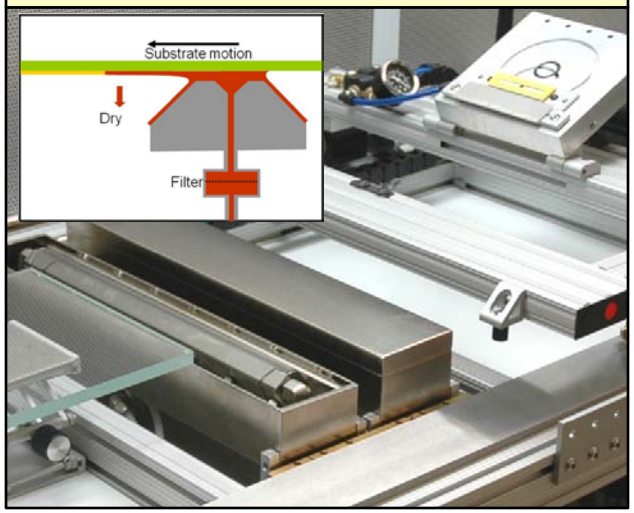
Offline tests	**Failure Mode**
Radiused Hohlraums	**100% Center**
Sharp Hohlraums	**44% Center 56% Edge**

Formvar's sensitivity to environment, material chemistry and mechanical asperities have lead to target yield loss

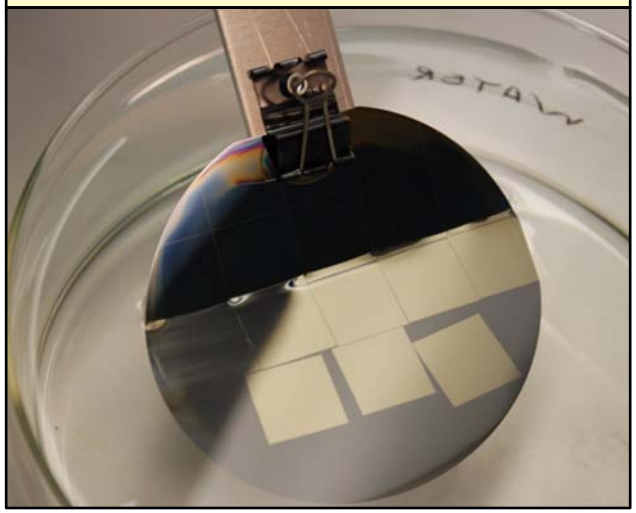
Effect	Category	Physical Effect /Mechanism	Impact	Mitigation	Status
1. H_2O partial pressure	Environment	H_2O absorption relaxes and weakens tent	Capsule centering; Tent wrinkling	Humidity control (40%±5%) & 2 week tent lifetime	✓
2. Glue vapor		Volatiles cause viscoplastic relaxation	Capsule centering	Gas Purge	✓
3. Asperities during tenting	Mechanical	Asperities cause stress rise that exceed failure stress	Tent tear	Radiused hohlraums/ Improved tooling for hohlraum Insertion	✓
4. Lot-to-lot variation	Chemical	Tent strength changes likely due to change in molecular weight dist.	Tent tear	QA of molecular weight (GPC) & side group conc (NMR)	✓
5. Formvar solution aging		Tent strength decreases with age; Mechanism TBD	Tent tear	Weekly tent mechanical testing & 2 month solution lifetime	✓

Tent Fabrication Process

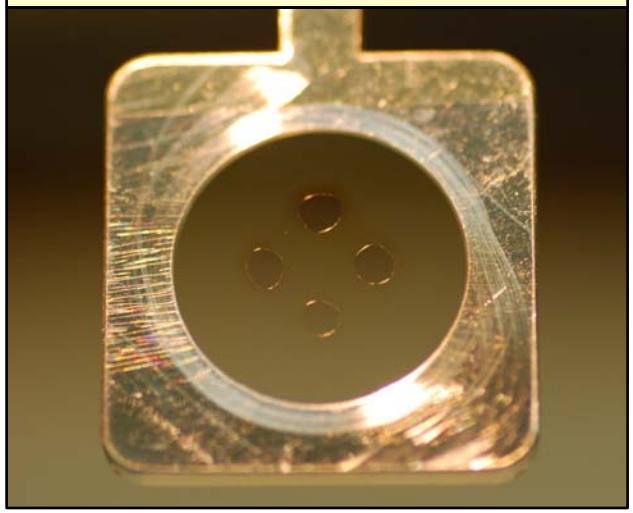
1. Meniscus Coating



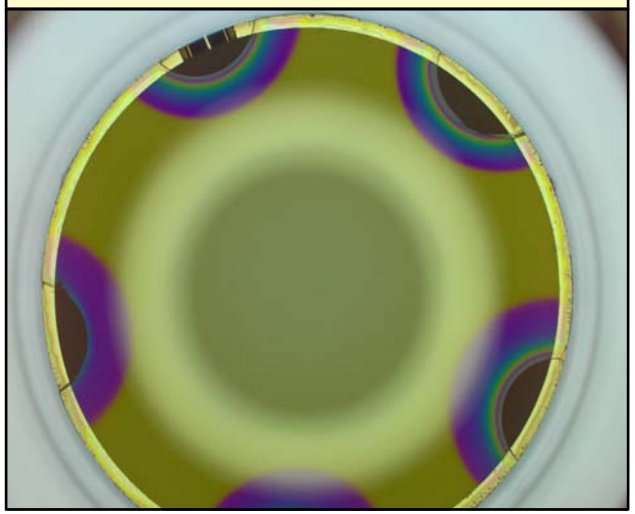
2. Liftoff



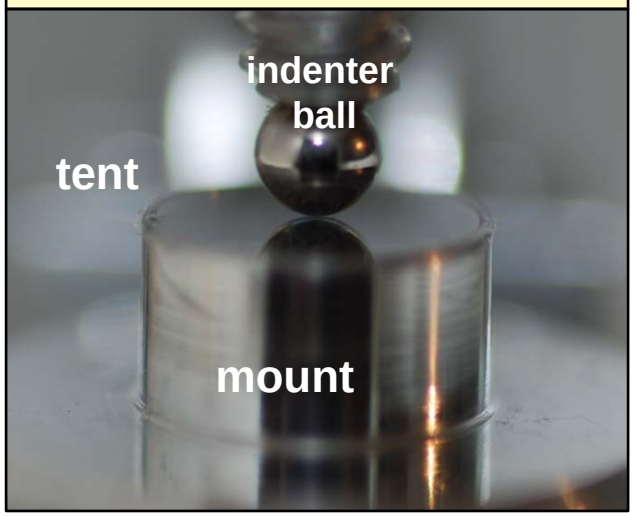
3. Mount & Hole burn



4. Glue on Hohlraum

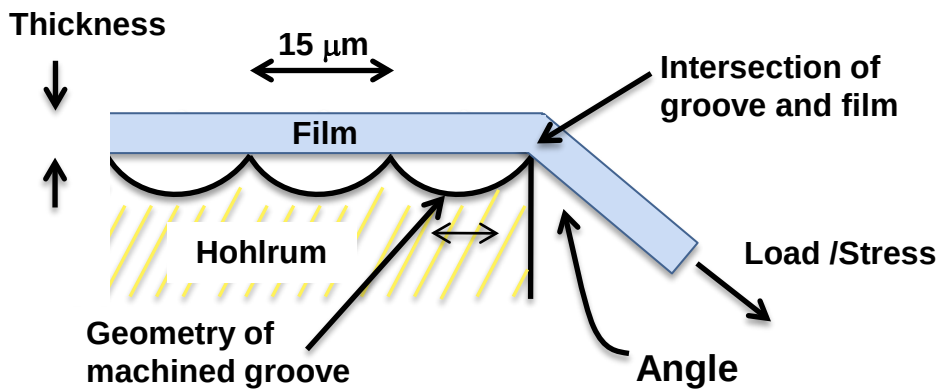


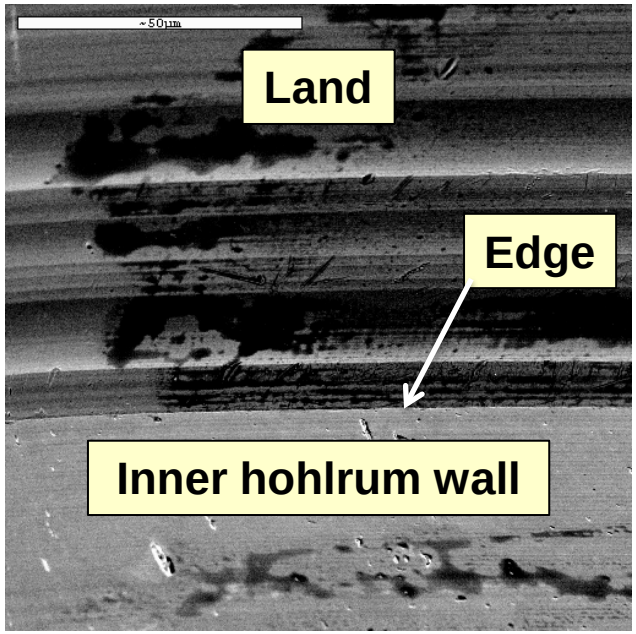
Offline QC tent strength



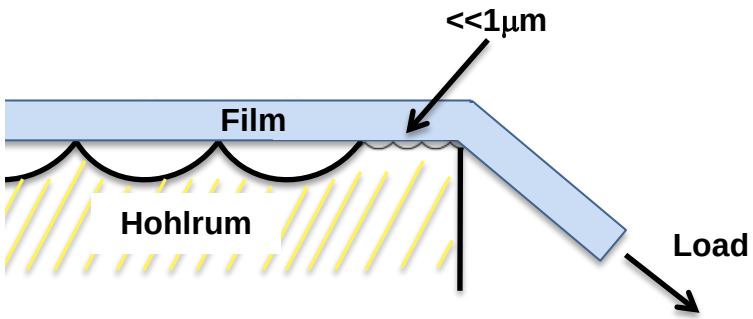
Premature tent failure have occurred at the edge of the hohlraum, several factors might influence the tear point of a film stretched over a hohlrum edge

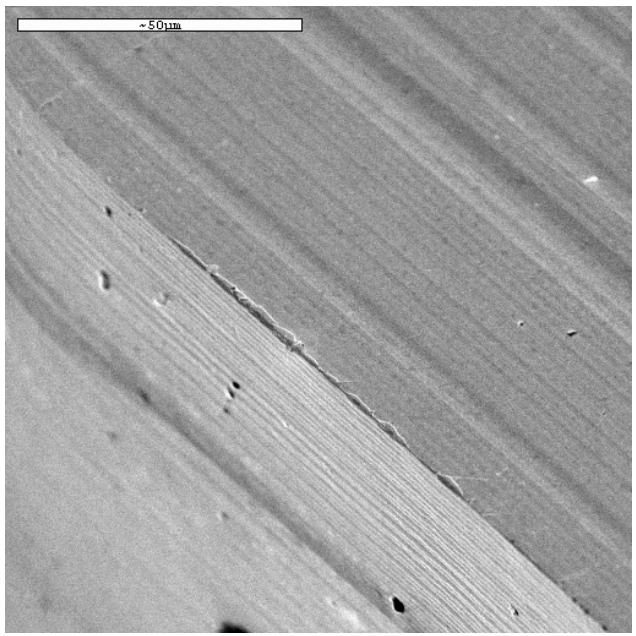
26990005 Failed Early in Offline Test

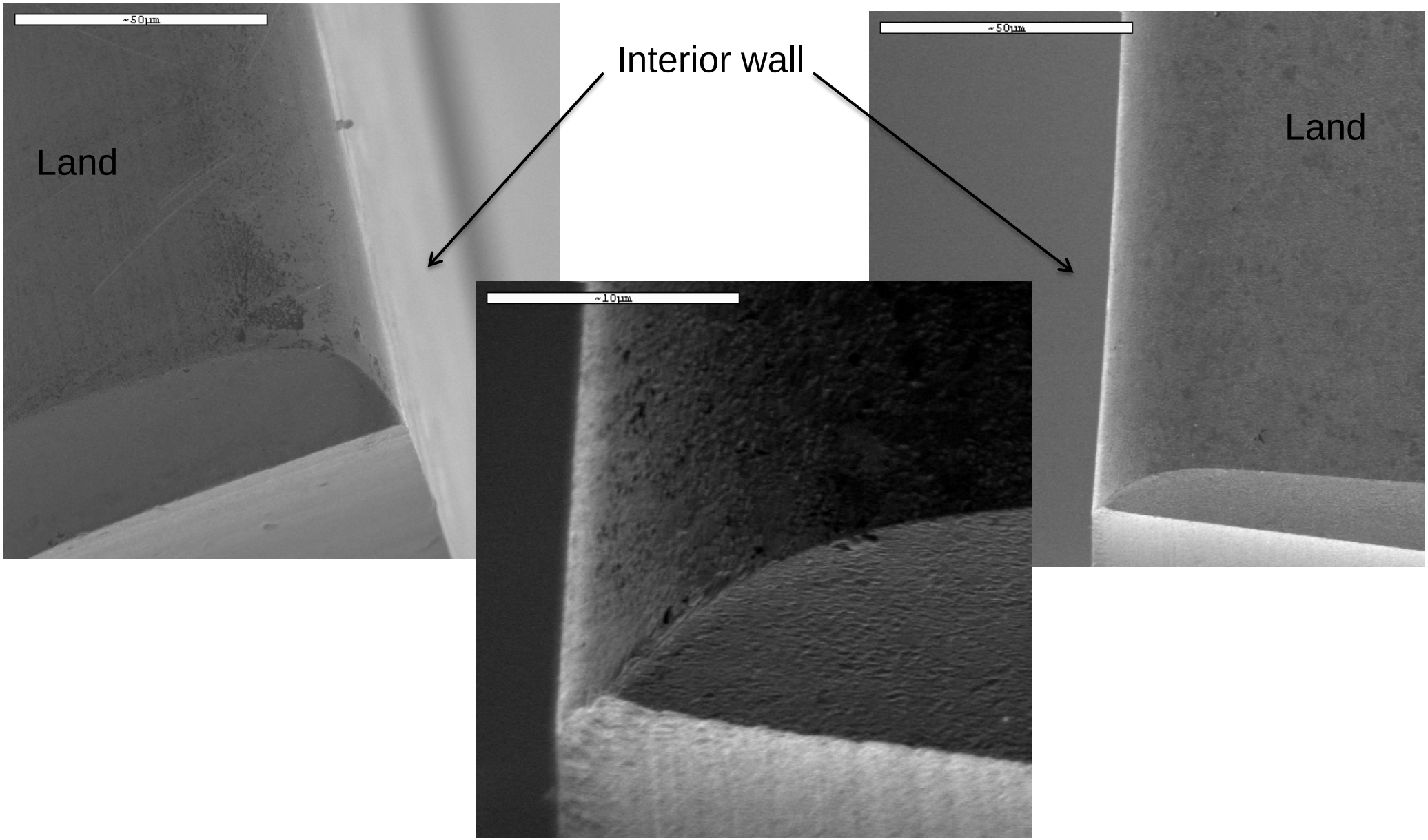




26990008 Failed Late in Offline Test





At no point do machining groove >1 μm intersect the edge of the hohlrum

Radiused hohlraum S/N 15180 exhibited similar high quality surfaces to that seen in S/N 15179