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CRYOGENIC LASER FUSION TARGET MATERIALS*

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Laser fusion target designs exhibit improved performance when the fusion fuel, a deuterium-tritium (DT) mixture, is frozen into a uniform, solid shell The formation of such a shell requires rapid isothermal cooling of the target to cryogenic temperatures. The cooling rate must be sufficiently fast to prevent significant, gravitationally driven downward flow of the DT as it passes from the gaseous through the liquid state. Because it is not possible to measure the uniformity of a solid DT layer in opaque, multi-shell targets, we have modeled such targets to calculate the cooling rate and, hence, the expected thickness uniformity of the DT shell. The presented results provide target designers with practical guidelines for the selection of materials and configurations, which will assist in the fabrication of high-quality cryogenic targets.

INTRODUCTION

Materials used in laser fusion targets span the periodic table from hydrogen to uranium. The targets are usually constructed by assembling concentric spherical shells of the various materials around a fuel core filled with a deuterium-tritium (DT) mixture, shown in Fig. 1. A general discussion of this target is given elsewhere [1]. The target fuel core is typically a 100-μm-diameter glass microballoon having a wall thickness of 1 μm and containing up to 10 ng of DT. Targets having the DT fuel frozen as a uniform layer onto the inner surface of the fuel core are of particular interest [2]. These targets are expected to generate a

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higher fusion yield for the same incident laser energy because a solid DT layer sourrounding a void can be compressed more easily than can a gas-filled sphere.

A number of materials problems arise for cryogenic fusion targets. The mechanical integrity of a multimaterial target fabricated at 300 K, then cooled to 4 K, depends on proper assembly techniques because the thermal expansion coefficient of each material is different. This fabrication problem, and the issue of workable adhesives, has been discussed previously [3]. A second materials problem, and the topic of this paper, concerns the ability to produce the required uniform layer of DT ice in the fuel core. Because the uniformity of the solid DT shell is strongly dependent on the rate of target cooling, each shell's geometry and thermal transport characteristics are important. However, it is not possible to optically measure the uniformity of a DT layer in an opaque, multishell fusion target, and we have, therefore, modeled such targets to calculate the cooling rate of the fuel core and hence the expected thickness uniformity of the solid DT layer. We will briefly describe, first, the processing technique for the cryogenic target, then the heat-flow model, and, last, we will present calculated cooling-rate results for some general multishell, multimaterial cryogenic laser fusion targets.

CRYOGENIC TARGET PROCESSING

A highly successful technique termed fast isothermal freezing (FIF) has been developed at the Los Alamos Scientific Laboratory to produce quality cryogenic laser fusion targets [4]. The FIF method requires that the target be rapidly, but uniformly, cooled from a temperature at which the fuel is a gas ($\sim$ 40 K) to a temperature at which the fuel is a solid ($<$ 19.7 K). As a result, the DT gas uniformly condenses into a liquid

shell on the inner surface of the fuel core and then freezes into place in this configuration before significant gravitationally driven liquid flow occurs. The temperature-entropy phase diagram for DT, shown in Fig. 2, contains the general path A-B-C-D-E followed as the target fuel core cools. Starting at A, where the DT is all gas, an isochore is followed to B where liquefaction begins. Between B and C, liquid and gas coexist. At C, the liquid to solid phase transition begins. The DT liquid is completely solidified at D. From D to E, gas continues to freeze. Gravitationally driven liquid flow can occur whenever liquid exists--while the path B-C-D is transversed.

The nonuniformity (NU) produced in the frozen DT shell, defined as the difference between the maximum and minimum shell thickness divided by the minimum shell thickness, depends on the time liquid is present, on the diameter of the fuel core, and on the mass of DT contained within the fuel core. Table I shows the predicted DT shell NU for three fuel-core diameters for different lengths of time the DT is liquid [5]. The room-temperature DT fill pressure is 10 MPa (100 atm) in each case. A 100-μm-diameter fuel core filled with DT to 10 MPa and frozen in 25 ms or a 500-μm-diameter fuel core filled to 10 MPa and frozen in 100 ms will each produce a NU of 0.10.

The FIF method relies on locally heating the target in an isothermal, helium-filled cryogenic cell with a focused, low-power laser beam. The laser vaporizes the DT fuel; then, when shuttered, the target cools rapidly with the subsequent uniform condensation and solidification of the DT onto the inner surface of the fuel core. The density of helium gas surrounding the target controls the cooling rate.

The FIF technique has produced solid DT shells whose uniformity has been measured quantitatively in transparent, bare fuel cores and in

transparent, two-shell targets. Interferograms of these cryogenic targets have been obtained with a simple device [6] and then compared with results generated by a ray-tracing computer code [7]. In these transparent targets, the FIF method reproducibly forms solid DT layers with a NU $\leq$ 0.12, the sensitivity limit of this comparison. Because it is not possible to measure the solid DT layer's uniformity in opaque, multishell targets interferometrically, and because no other diagnostic techniques are available to measure the uniformity of a thin, low-Z shell (DT) inside one or more opaque high-Z shells (gold, for example), we have developed a computer model for such targets to calculate the cooling rate of the fuel core and hence the expected thickness uniformity of the solid DT layer.

HEAT-FLOW MODEL -- CONDENSATION

Although laser fusion targets are radially symmetric and therefore can be mathematically modeled in one dimension, the cooling rate of a multishelled target cannot be solved analytically. This inability from the strong temperature-dependence of both the thermal conductivity and the specific heat of most materials over the temperature range of 4 to 40 K and from the thermal interaction of each layer in a multimaterial target.

Referring to Fig. 2, the problem of calculating the time liquid exists in a fuel core can be separated into two parts. The first part consists of determining the time required to transverse from B to C, and the second part, the time required to transverse from C to D. The sum of these two times is the quantity of interest used to predict the NU of the frozen DT shell.

The time needed for the DT gas to condense into a liquid shell (B-C in Fig. 2) is discussed first. In the formulation of this general, nonlinear, boundary-value problem of heat transfer in a multishell sphere with

temperature-dependent thermal properties, the following assumptions were made:

1. Thermal conduction is the only mode of heat transfer between the sphere and its cryogenic environment;
2. Heat conduction is radially symmetric, thus one-dimensional;
3. Each shell is isotropic and constitutionally homogeneous;
4. All shells are isometric;
5. No interfacial thermal resistance exists between adjoining layers;
6. Temperatures change slowly relative to the computational time step, allowing the calculation of thermal transport properties of each layer to follow continuously;
7. Transport properties of saturated DT are acceptable for conservative computation of condensation time; and
8. The environmental temperature is constant at distances far from the target.

The one-dimensional heat equation for a multishell sphere with temperature-dependent thermal properties is

$$\frac{\partial T_\ell}{\partial t} = \alpha_\ell(T) \left[\frac{\partial^2 T_\ell}{\partial r^2} + \frac{2}{r} \frac{\partial T_\ell}{\partial r} \right] \qquad (1)$$

for $r_\ell \leq r \leq r_{\ell+1}$ ← where $\ell = 1, 2, \ldots m$, m is the number of spherical layers and the thermal diffusivity of layer ℓ is

$$\alpha_\ell(T) = \left[\frac{k(T)}{\rho(T) C_p(T)} \right]_\ell \quad ; \qquad (2)$$

here $k(T)$ is the thermal conductivity, $\rho(T)$ is the density, and $C_p(T)$ is the specific heat of shell ℓ.

Equation (1) is subject to the following initial conditions:

$$T_\ell\ (r,\ o) = T_i = 40\ K \qquad \text{for} \qquad 0 \leq r \leq b \tag{2a}$$

$$T_{m+1}\ (r,\ o) = T_\infty = 4\ K \qquad \text{for} \qquad b < r \leq R \text{ and } R >> b, \tag{2b}$$

where $b \equiv$ the target radius and $\ell = 1, 2, \ldots m$.

Equation (1) is subject to three boundary conditions. At the target center, symmetry requires that

$$\left.\frac{\partial T_1}{\partial r}\right|_{r=o} = 0 \qquad . \tag{3a}$$

Continuous heat flux at the shell interfaces is represented by

$$k_\ell(T_\ell)\ \frac{\partial T_\ell}{\partial r} = k_{\ell+1}\ (T_{\ell+1})\ \frac{\partial T_{\ell+1}}{\partial r} \qquad . \tag{3b}$$

The boundary condition imposed very far from the target is

$$\left.\frac{\partial T_{m+1}}{\partial r}\right|_{r=R} = 0 \qquad \text{where } R >> b \qquad . \tag{3c}$$

An exact numerical solution to the above set of equations was obtained from a standard finite-difference formulation [8,9]. Stable, physically realistic results for all times were obtained with a Crank-Nicholson Scheme employing a time dependent weighting factor [10]. With a constantly spaced nodal mesh, the transient target temperature distribution had minimal dependence on the time step used. Therefore incorporation of a variable time step into the finite-difference scheme had the added benefit of minimizing computational effort. Less than one minute of CDC 3600 cpu time was needed for each multishell target evaluated.

The numerical solution of Eq. 1 for the DT condensation time in a two-shell target is shown in Fig. 3. The family of curves, reduced

temperature vs reduced radius, plots the condensation time of DT gas in a 100-μm-diameter, 1-μm-thick glass fuel core positioned inside a second 1-μm-thick glass shell. The target, initially at T_i = 40 K, cools to T_F = 19.7 K in 63 ms. Condensation times for other target configurations and/or target materials are shown in Fig. 4. In general, thicker shells or larger target diameters increase the DT condensation time. Also shown in the figure are the temperature-dependent thermal properties of the target materials used in the calculations.

Because the DT condensation times are strongly affected by the helium diffusivity, and because thermal-property data for cryogenic, gaseous helium at low pressures are scarce [11], the diffusivity relationship $\alpha(T) = (2.9 + 141\ T)10^{-7}$ cm^2/s was obtained by matching calculated and experimental results of the freezing time for a bare, 100-μm-diameter, glass fuel core (the upper left target in Fig. 4). As a result, all condensation times given in Fig. 4 are relative and correspond to a fixed helium exchange-gas density. The condensation times can be changed by changing the density of the gaseous helium.

Although metal shells are not considered in Fig. 4, condensation-time results for such shells have been obtained. The effect on condensation time of adding a metal shell is minimal: no significant time increase is seen, due to the relatively high thermal conductivity of metals compared to that of other target materials.

HEAT FLOW MODEL - SOLIDIFICATION

The time required to freeze a liquid DT shell is the second part of the liquid-existence-time problem. This time is indicated by the path C to D in Fig. 2. As stated, the freezing time, when added to the condensation

time, allows us to predict the layer nonuniformity of solid DT in a target fuel core.

The freezing time of a liquid DT shell of outer radius b, initially at the fusion temperature T_F = 19.7 K, is found by calculating the solid-liquid interfacial boundary location as a function of time. At t = 0, the surface temperature of the liquid shell is instantaneously changed to, and held constant at, T_∞ = 4 K. The liquid begins to freeze, moving the liquid-solid interface inward. The position of this interface x = F(t) is used in calculating the freezing time of the liquid shell.

The dimensionless differential equation describing the temperature distribution is

$$\frac{\partial^2 u}{\partial x^2} = \frac{\partial u}{\partial \tau} \quad \text{for } 0 < x < F(\tau) \text{ and } \tau > 0 \quad . \tag{4}$$

The dimensionless variables are defined as:

$$\tau = \frac{\alpha_F t}{b^2} \qquad \text{for time,} \tag{5}$$

$$x = \frac{b-r}{b} \qquad \text{for position,} \tag{6}$$

$$u(x,\tau) = \frac{r}{b}\left[\frac{T-T_F}{T_\infty - T_F}\right] \qquad \text{for temperature,} \tag{7}$$

$$F(\tau) = \frac{b-F(t)}{h} \qquad \text{for interface,} \tag{8}$$

$$\text{and} \quad L = \frac{L_F}{C_p(T_F - T_\infty)} \qquad \text{for enthalpy,} \tag{9}$$

where α_F is the heat of fusion.

The initial conditions for Eq. 4 are:

$$u(x,0) = 1 \text{ and } F(0) = 0 \quad . \tag{10}$$

At the outer surface of the shell, the boundary condition $u(0,\tau) = 1$ is imposed for all time. At the moving interface $x = F(\tau)$, the boundary conditions are:

$$u(F(\tau),\tau) = 0 \tag{11a}$$

and $$\frac{\partial u(F(\tau),\tau)}{\partial x} = -1.\left[1 - F(\tau)\right]\frac{\partial F(\tau)}{\partial \tau} \tag{11b}$$

An approximate solution to the heat-flow problem involving a liquid-solid phase change in spherical bodies, as described by the above set of equations, has been formulated [12]. This approximate solution was used to compute freezing times of liquid DT shells. Thermodynamic values used here for DT were [13]:
$T_F = 19.7$ K, $\rho = 0.224$ g/cm^3, $C_p = 9.275$ J/gK and $L_F = 47.7$ J/g. The results of this calculation, shown in Fig. 5, indicate that the DT fusion time is short (< 1 ms) compared to typical DT condensation times (≥ 10 ms). Consequently, layer nonuniformities will arise during DT condensation rather than during DT solidification. Also shown in Fig. 5 is the predicted aspect ratio (shell thickness vs shell diameter) for a DT fuel core filled to a pressure of 10 MPa at 300 K.

SUMMARY

Model calculations have been performed to determine the length of time liquid DT is present during the formation of cryogenic laser fusion targets. Our results make it possible to predict the thickness uniformity of solid DT shells. As shown, gas condensation occurs at a much slower rate than does the subsequent solidification of the liquid shell.

Some typical condensation-time results, shown in Fig. 4, indicate that larger targets, or targets having thick low-thermal-conductivity shells,

contain liquid DT substantially longer than do bare fuel cores. To form a solid DT shell of desired uniformity, the density of the helium exchange gas surrounding a target can be adjusted. As stated, metal shells do not adversely affect the ability to produce a uniform layer of DT ice in the fuel core.

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FIGURE CAPTIONS

Fig. 1. Multishell laser fusion target.

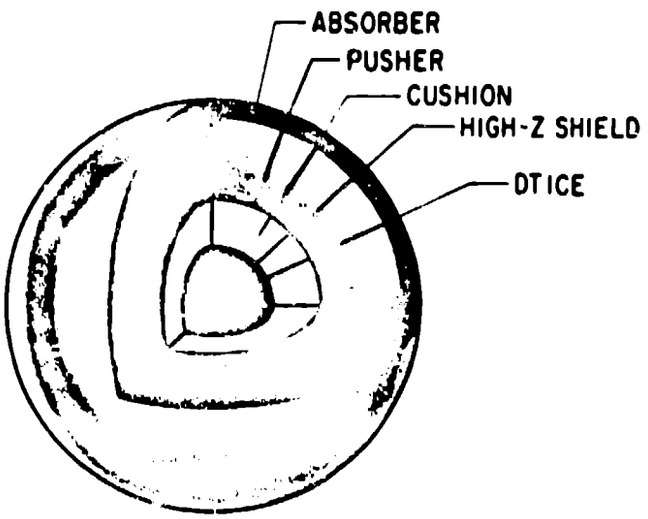
Fig. 2. Temperature-entropy phase diagram for DT.

Table I. Calculated DT shell nonuniformity.

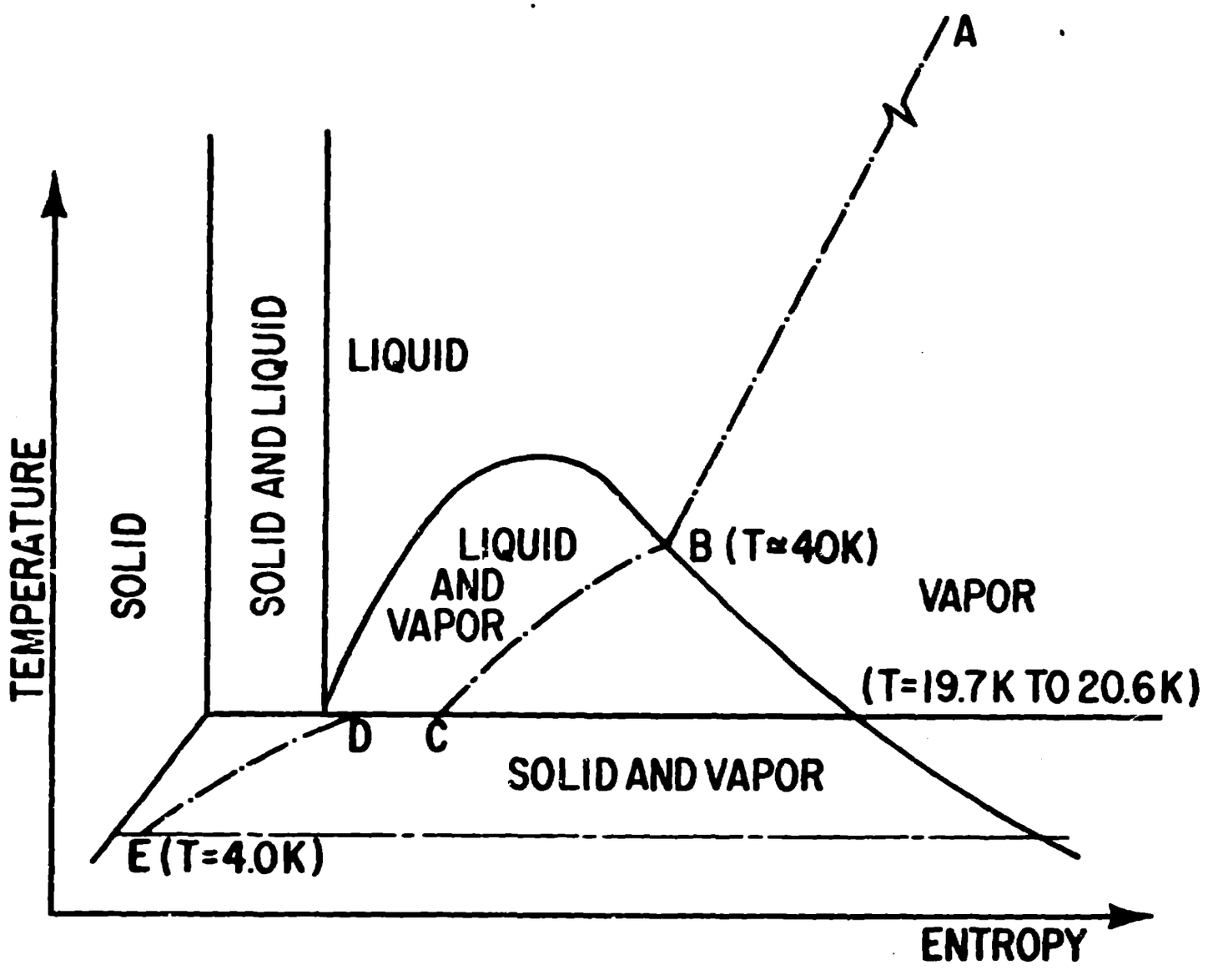
Fig. 3. DT condensation time calculation for a two-shell target.

Fig. 4. DT layer condensation times of various targets.

Fig. 5. Freezing times of liquid DT shells.

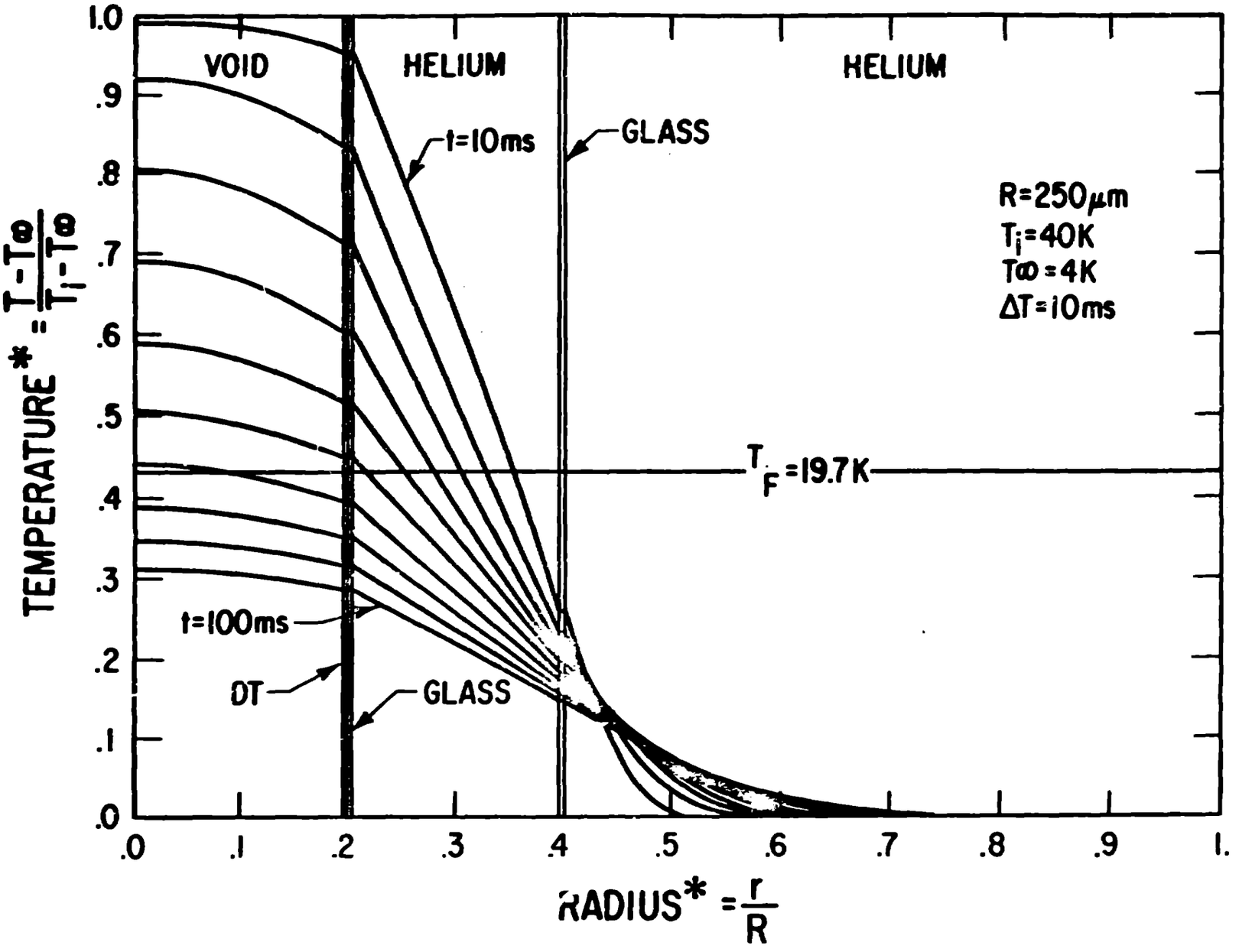


OVERALL DIAMETER ≃ 2mm


A
LIQUID
SOLID AND LIQUID
SOLID
TEMPERATURE
LIQUID
AND
VAPOR
B (T≈40K)
VAPOR
(T=19.7K TO 20.6K)
D
C
SOLID AND VAPOR
E (T=4.0K)
ENTROPY

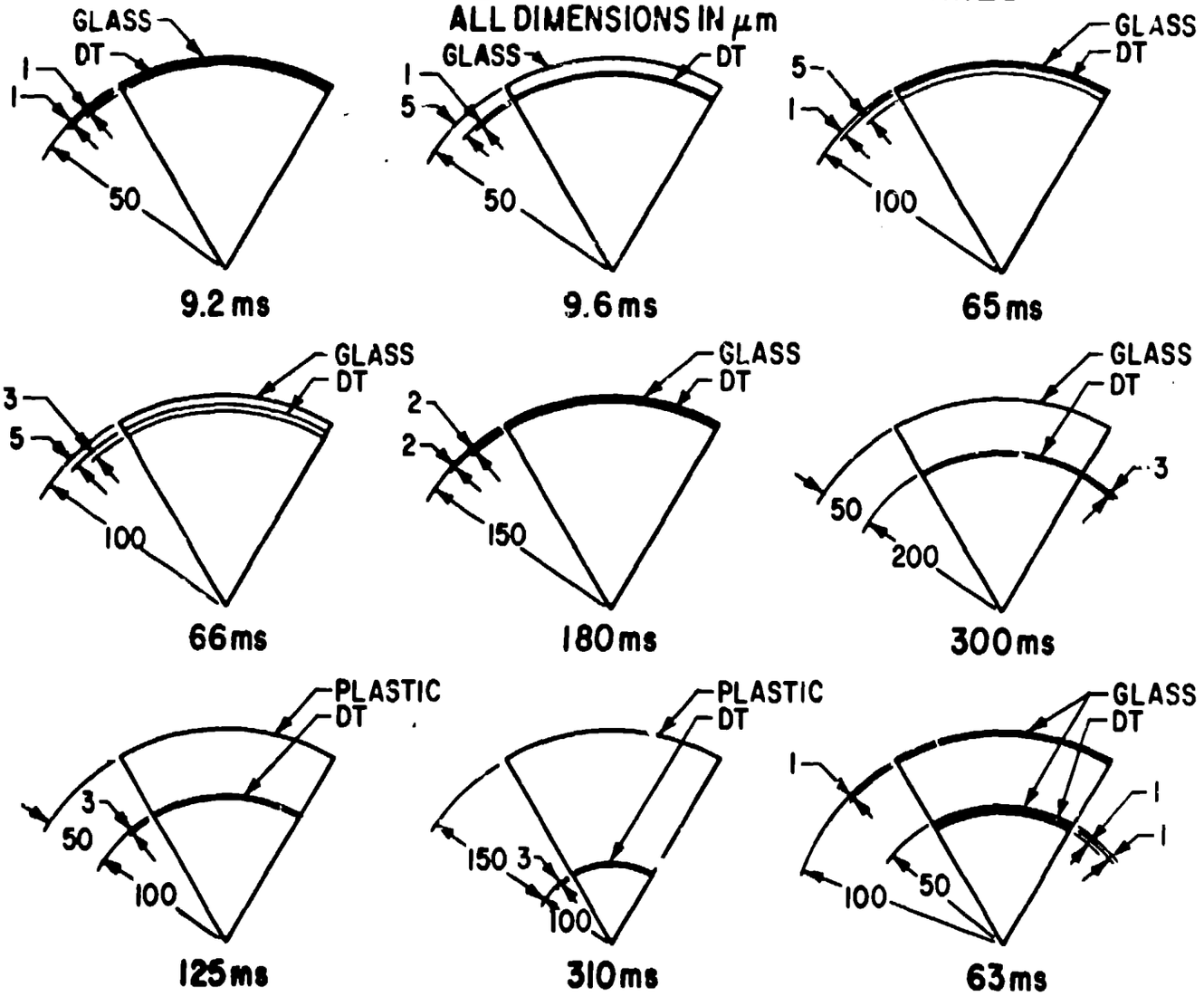
Fuel Layer Nonuniformity (NU)

LIQUID EXISTENCE TIME (ms)	FUEL CORE DIAMETER (FILL PRESSURE = 10 MPa)		
	100 μm	300 μm	500 μm
10	0.04	0.01	0.01
20	0.08	0.03	0.02
30	0.12	0.04	0.02
40	0.18	0.06	0.04
50	0.22	0.07	0.04
60	0.28	0.09	0.06
70	0.32	0.11	0.06
80	0.38	0.13	0.08
90	0.42	0.14	0.08
100	0.48	0.16	0.10

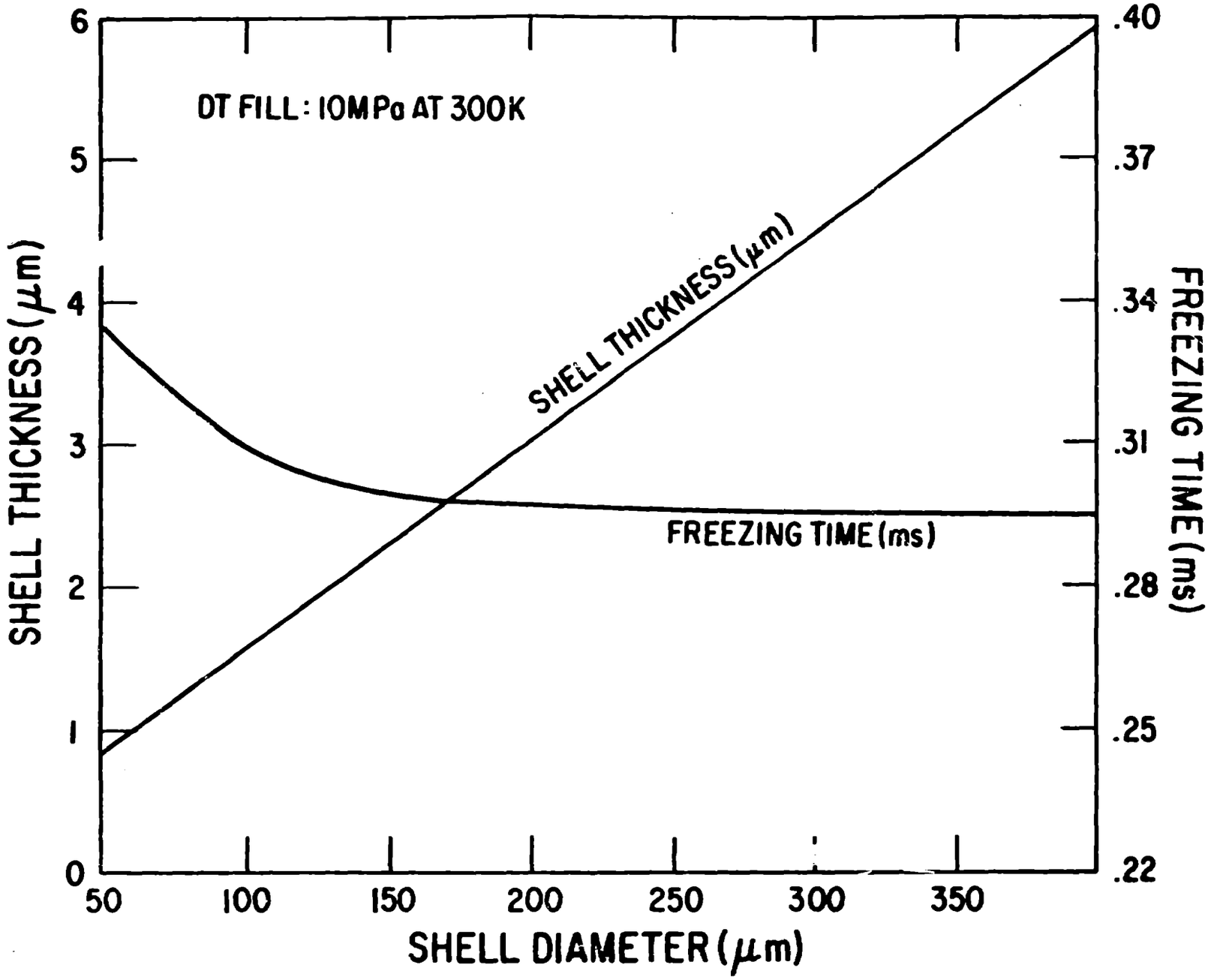

VOID
HELIUM
HELIUM
t=10ms
GLASS
R=250μm
T_i=40K
T∞=4K
ΔT=10ms
T_F=19.7K
t=100ms
DT
GLASS
TEMPERATURE* = (T-T∞)/(T_i-T∞)
RADIUS* = r/R
1.0
.9
.8
.7
.6
.5
.4
.3
.2
.1
.0
.0
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.

SAMPLE TARGETS - CONDENSATION TIMES

ALL DIMENSIONS IN μm



TRANSPORT PROPERTIES		
MATERIAL	α (cm^2/s)	k ($W/cm \cdot K$)
HELIUM	$(2.900+141T)10^{-7}$	$(62.33+8.520T)10^{-7}$
DT(LIQUID)	$(8.6328-0.15247T)10^{-4}$	$(7.1246+0.2333T)10^{-4}$
DT(GAS)	5.0×10^{-4}	2.0×10^{-6}
GLASS	$(4.5676-0.09768T)10^{-2}$	$(6.775+0.4276T)10^{-4}$
PLASTIC	$(2.65-0.01T)10^{-3}$	$(5.65+0.043T)10^{-2}$


DT FILL: 10MPa AT 300K
SHELL THICKNESS (μm)
FREEZING TIME (ms)
SHELL DIAMETER (μm)
0
1
2
3
4
5
6
50
100
150
200
250
300
350
.22
.25
.28
.31
.34
.37
.40